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ON THE HYPER-GEOMETRIZATION OF RELATIVISTIC PHASE-SPACE FORMALISM III

By

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(Presented by A. Kónya — Received 30. V. 1967)

In the first two parts of this paper the definition and the geometrical structure of relativistic phase-space were investigated; later the idea of hypergeometrization was discussed reformulating several previous results. Now, the suggested method will be used to develop a relativistic kinetic theory of gases. First different definitions of the distribution function, then the derivation of Boltzmann's relativistic transport equation will be discussed and the suggested general results will be checked in special cases previously investigated by several authors. Finally, having ISRAEL's theory in mind, the outline of the foundation of a general relativistic thermodynamics will be proposed.

In order to find a way to characterize the collective behaviour of a macrophysical system of gaseous particles one has to introduce one of the most important devices of kinetic and statistical theories, the *distribution function*, whereby the mathematical framework to render possible the calculations of its macroscopic quantities of states can be developed.

Two different philosophies are concealing behind the usual interpretations of the distribution function both in relativistic and non-relativistic cases. According to the first one the distribution function $\mathcal{F}(x, p)$ — strictly speaking $\mathcal{F}(x, p)d\Omega$ — denotes the number of particles in the volume-element dV of the configuration-space, the momentum components of which lie in the specified volume-element dP of the momentum-space. The didactical power of this familiar definition should not be doubted and anybody knows what it means, but it is horrifying to a mathematician to think of the paradoxical argumentation usually following this definition in several excellent text-books of kinetic and statistical theories, according to which the measure of $d\Omega$ has to be small enough to master the limiting process needed, but big enough to contain the necessary number of particles for a statistical treatment. All the conceptual difficulties of this definition can easily be overcome in terms of the theory of probability where the distribution function means the probability density to find the phase-point of one of the gaseous particles in the volume-element $d\Omega$ of the phase-space. As a further advantage of the second philosophy it may be regarded that it provides a possibility for developing kinetic and statistical theory in the language of field theories.

Keeping the dimensionality of the relativistic phase-space [42]* and the definition of the relativistic phase-space volume-element in mind [43], one can immediately observe that the distribution function $\mathcal{F}(x, p)$ — defined only provisionally above — has another dimension as the distribution function of the non-relativistic theory. As a matter of fact, we have to discuss the relation of the relativistic distribution function to the non-relativistic one.

After the definition of the distribution functions in the cases of different particular gaseous systems the explicit expression of this fundamental state quantity of the gaseous systems has to be determined. This may be done either in terms of combinatory methods or by finding the general differential equation representing the external and internal dynamic interactions as well as the influence of the stochastic forces induced by the binary elastic collisions of the particles, the solution of which gives the distribution function. The first method was already used by **Jüttner** [1] developing his relativistic gas theory, the second when the more up-to-date kinetic theories of relativistic gases have been treated. Also in the following, the latter method will be discussed based on the framework of the hyper-geometrized version of the relativistic phase-space. Finally, the outline of the foundation of a general kinetic theory of gaseous systems will be proposed.

§ 5. New derivation of the relativistic Boltzmann's transport equation

In order to derive Boltzmann's transport equation first of all we have to formulate the underlying equations of motion for single gaseous particles to find the equations of phase-space trajectories. Then we have the possibility to suggest a definition for the distribution function, or rather to formulate the problem in field theoretical terms based on the definition of an adequate field of probability.

5.1. *Equations of motion as the equations of phase-space trajectories.* The problems connected with the equations of motions of the gaseous particles were investigated in Section 3.1 and the definition of the curves of line-element spaces was treated in Section 2.3, respectively. Keeping these considerations in mind in the framework of the suggested theory where the geometrization of the phase-space is carried out in terms of line-element geometry, obviously, the relativistic trajectories of the gaseous particles have to be represented by the set of equations

$$\frac{dx^\mu}{d\tau} = p^\mu, \tag{5.1.1}$$

---

* Numeration of paragraphs, formulae and references will be continued in this part, too and our notations are, of course, co-ordinated with those of the previous parts [42, 43] as well.

$$\frac{Dp^\mu}{d\tau} = K^\mu, \tag{5.1.2}$$

where here and in the following it is again supposed that $m_0 = 1$ and τ means the parameter of the length of arc being the proper time of the gaseous particles along different world-lines of configuration-space.

The solution of this set of differential equations is

$$x^\mu = x^\mu(\tau) \tag{5.1.3}$$

which, according to the definition (2.3.2) of the curves in general line-element spaces, can be regarded as the trajectories of the gaseous particles.

In this way the influence of external fields K_μ can be regarded either in the case of perfect gas systems or — having **Israel**'s proposal in mind [9] — in the case of systems of gaseous particles with gravitational interactions. However, to render it possible to take into account also the stochastic interactions among gaseous particles being characteristic from the point of view of the collective behaviour of the macroscopic system, in addition, one has to find the relativistic generalization of the collision integral, to complete the derivation of the transport equation.

5.2. *The field of probability describing the collective properties of the gas systems; the definition of the distribution functions.* The volume and the volume-element of the $(4 + 3)$-dimensional relativistic phase-space, respectively, differs not only in its geometrical but also in its physical dimension from that of the $(3 + 3)$-dimensional non-relativistic phase-space possessing the physical dimension of "ergsec". Should the phase-points $\{x, p\}$, or $\{x, \xi\}$ be regarded as variables of probability and a probability field $\mathcal{F}(x, p)$ or $\mathcal{F}(x, \xi)$ be defined over the relativistic phase-space, then the interpretation of these density functions, of course, will be quite different from that of non-relativistic phase-probability.

The difference of the meaning between the distribution functions mentioned above can be found in the fact that while in the non-relativistic theory the time-coordinate plays the part of a parameter whereon the phase-points $\{\mathbf{x}(t); \mathbf{p}(t)\}$ depend, in the formalism of the relativistic configuration-space the time- and space-relations are already considered due to the geometrical structure of the space-time continuum. As a matter of fact, while in the non-relativistic theory the natural motion of the phase-space means the continuous sequence (one-parameter group) of transformations of the non-relativistic $(3 + 3)$-dimensional phase-space onto itself, in the relativistic theory the gaseous particles are moving along their world-lines and the distributions of the phase-points from instant to instant are represented by distributions of the phase-points on the sequence of successive $(3 + 3)$-dimensional space-like hypersurfaces. Consequently, the problem of the non-relativistic theory: "what is the probability of finding the phase-point at an instant of time in a phase-space volume-element?" has to be replaced in the relativistic theory by the following one: "what is the probability of crossing the world-line of the gaseous particle and the (3 + 3)-dimensional surface-element of the space-like hypersurface corresponding to the instant of time?". While in the non-relativistic theory phase-probability was explicitly dependent on time only in the case of non-stationary processes in the relativistic case the distribution function $\mathscr{F}(x, p)$ has to state, what is the probability of crossing the trajectory through a certain hypersurface in a given time-interval.

Let us suppose that *the probability of finding the phase-point* $\{x, p\}$ *or rather* $\{x, \xi\}$ *in a volume-element* $d\Omega$ is determined by

$$\begin{aligned} dw(x, \xi) &\overset{\text{def}}{=} \mathscr{F}(x, \xi)\, d\Omega = \mathscr{F}(x, \xi)\, m_0^3\, dV\, d^3\xi = \\ &= \mathscr{F}(x, p)\, dV dP = \mathscr{F}(x, p)\, d\Omega \overset{\text{def}}{=} dw(x, p) \end{aligned} \tag{5.2.1}$$

being invariant of the complete group $\mathcal{G} = \mathcal{G}_x \times \mathcal{G}_\xi$ *of the transformations*

$$x^{\mu'} = x^{\mu'}(x_\mu),\ \xi_{i'} = \xi_{i'}(\xi_i)\ \left(\Delta \equiv \frac{\partial(x^{0'}, x^{1'}, x^{2'}, x^3)}{\partial(x^0, x^1, x^2, x^3)} \neq 0,\ D \equiv \frac{\partial(\xi_{1'}, \xi_{2'}, \xi_{3'})}{\partial(\xi_1, \xi_2, \xi_3)} = \pm 1\right). \tag{5.2.2}$$

In order to obtain a deeper insight into the meaning of our definition one has to remember the explicit expression (4.5.21) of the relativistic phase-space volume-element. Consider an infinitesimal phase-tube of finite length generated by the natural motion of neighbouring phase-points bounded by the (3 + 3)-dimensional surface-element $d\Omega_0^{(\sigma')}$ on the space-like hypersurface σ' and by a similar surface-element $d\Omega_0^{(\sigma'')}$ on the space-like hypersurface σ''. Let us suppose that the volume-element in our above definition is a sheet of this phase-tube of a thickness of $d\tau$ denoted in the following by

$$\begin{aligned} d\Omega^{(\sigma)} &= d\Omega_0^{(\sigma)}\, d\tau = m_0^{-1}(p_\lambda n^\lambda)\, \varepsilon p\, d\varepsilon\, d\omega\, d\mathscr{V}^{(\sigma)}\, d\tau = \\ &= (p_\lambda n^\lambda)\, \varepsilon\xi\, d\varepsilon\, d\omega\, d\mathscr{V}^{(\sigma)}\, d\tau\,, \end{aligned} \tag{5.2.3}$$

where p_λ is the tangent of the world-line of the gaseous particle and n^λ means the unit normal vector of the hypersurface-element on the space-like σ hypersurface at their crossing-point at the instant of time τ. In fact,

$$dw^{(\sigma)}(x, p) = \mathscr{F}_{(\sigma)}(x, p)\, d\Omega^{(\sigma)} = \mathscr{F}_{(\sigma)}(x, p)\, d\Omega_0^{(\sigma)}\, d\tau = \mathscr{F}_{(\sigma)}(x, \xi)\, d\Omega_0^{(\sigma)}\, d\tau \tag{5.2.4}$$

means the probability that the trajectory of the gaseous particle considered crosses the surface-element $d\Omega_0^{(\sigma)}$ *of the space-like hyper-surface* τ *at the interval* $(\tau, \tau + d\tau)$ *of its proper-time.*

Having the definition (4.5.21) of $d\Omega^{(\sigma)}$ and that of $d\Omega$ [eq. (4.3.2)] in mind, we know that in the rest system of reference $\mathscr{K}^\circ$ if the σ hypersurface coincides with the hyper-plane $x^\circ =$ const. and the inhomogeneous direction co-ordinates are referred to the λ^+-triad, then the latter is a special case of the first one: $d\Omega = d\Omega^{(\sigma)}$. Owing to the convention that in the case of an internal inversion in respect to the origin of the λ^+-triad the normal direction of the σ hypersurface has not to be changed, $d\Omega$ is a pseudoscalar of the transformations of our general group (5.2.2) unlike $d\Omega$ which is a scalar. Here, in the following, we shall use the scalar volume-element $d\Omega^{(\sigma)}$, but we beware of the redefinition of $d\Omega$, namely, such a change of the normal direction of the σ hypersurface would entail time-reversal which could be problematic in some applications of the method. As the probabilities (5.2.1) and (5.2.3) are per definitionem invariant, keeping the transformation properties of $d\Omega$ and $d\Omega^{(\sigma)}$ in mind, it is easily obtained that the probability field $\mathscr{F}(x, p)$ is pseudo-scalar and the probability field $\mathscr{F}_{(\sigma)}(x, p)$ is a scalar.

The trajectories of the gaseous particles are time-like curves. Therefore, they cross certainly the space-like σ hypersurfaces. Let the probability $dw^{(\sigma)}(x, p)$ be integrated over the whole σ hypersurface then

$$w^{(\sigma)}(\tau) \overset{\text{def}}{=} \int_\sigma d\Omega_0^{(\sigma)} \mathscr{F}_{(\sigma)}(x, p) \tag{5.2.5}$$

is obtained, where $w^{(\sigma)}(\tau)d\tau$ means the probability that the trajectory crosses the space-like σ hypersurface in the proper time-interval $(\tau, \tau + d\tau)$. As a matter of fact,

$$\int_{-\infty}^{+\infty} d\tau\, w^{(\sigma)}(\tau) = \begin{cases} 1, \; \textit{if } w^{(\sigma)}(\tau) \textit{ is integrable} \\ \quad \textit{along the world-line;} \\ \infty, \; \textit{if } w^{(\sigma)}(\tau) = \text{const.} \end{cases} \tag{5.2.6}$$

This means, if the statistical weight of the phase-points along the trajectories — e.g., due to the finite life-time of the gaseous particles — is variable, so that $w^{(\sigma)}(\tau)$ differs only on a finite section of the world-line from zero, or for $\tau \to -\infty$ and $\tau \to +\infty$ decreases exponentially, then $\mathscr{F}_{(\sigma)}(x, p)$ can be normalized in the usual way:

$$\int d\Omega^{(\sigma)} \mathscr{F}_{(\sigma)}(x, p) = 1\,, \tag{5.2.7}$$

where the integration has to be extended over the whole phase-space.

However, if the life-time of the gaseous particles is infinite — as is usual in the case of gas-systems with unchanging number of particles $N =$ const. — the trajectories are running from $\tau = -\infty$ to $\tau = +\infty$, it can be supposed that the statistical weight of the phase-points along the whole world-line is constant and as a result the normalization of type (5.2.7) looses its meaning.

But, in this case each space-like σ hypersurface is crossed by the trajectories and it is a less stimulating problem in which time-interval $(\tau, \tau + d\tau)$ the crossing takes place. Since, in this case it is rather interesting to formulate our problem in the following way: *"what is the probability that the trajectory crosses the hypersurface σ through it surface-element $d\Omega_0^{(\sigma)}$?"*. Certainly, this may be the reason that only the problem formulated in this way has been investigated by previous authors.

Let the life-time of the gaseous particles be denoted by τ^* and, e.g., we suppose that τ is a variable of probability with Poisson-distribution. Furthermore, let us suppose that the crossing of the trajectory and the hypersurface σ takes place at the instant of the proper time τ_0, then the probability density function $\mathscr{F}_{(\sigma)}(x, p)$ may be given in the form

$$\mathscr{F}_{(\sigma)}(x, p) \sim \exp\left\{- |\tau - \tau_0|/\tau^*\right\} f_{(\sigma)}(x, p)\,. \tag{5.2.8}$$

In the case of gaseous particles with infinite life-time, the crossing at an instant of time τ_0 can certainly be realized. Therefore, it seems to be reasonable to suppose that

$$\mathscr{F}_{(\sigma)}(x, p) = \delta(\tau - \tau_0)\, f_{(\sigma)}(x, p)\,, \tag{5.2.9}$$

namely, in this case

$$\begin{aligned}\int_{-\infty}^{+\infty} d\tau \int_\sigma d\Omega_0^{(\sigma)}\, \mathscr{F}_{(\sigma)}(x, p) &= \int_{-\infty}^{+\infty} d\tau \int_\sigma d\Omega_0^{(\sigma)}\, \delta(\tau - \tau_0) f_{(\sigma)}(x, p) = \\ &= \int_{\sigma^*} d\Omega_0^{(\sigma^*)}\, f_{(\sigma^*)}(x, p)\,,\end{aligned} \tag{5.2.10}$$

where σ^* means the space-like hypersurface containing the phase-point corresponding to the parameter τ_0 and, of course, the normalization of this $(3 + 3)$-dimensional integral can be carried out.

$f_{(\sigma)}(x, p)$ denotes — in both cases — the probability density function of the event that the trajectory crosses the space-like hypersurface σ at the instant τ of the proper time in its point $\{x, p\}$ and $\{x, \xi\}$, respectively. By introducing an adequate factor of normalization also in the case of $\mathscr{F}_{(\sigma)}(x, p)$ defined by (5.2.8), it can be supposed that for any given space-like hypersurface the normalization

$$\int_\sigma d\Omega_0^{(\sigma)}\, f_{(\sigma)}(x, p) = 1\,, \tag{5.2.11}$$

or in terms of the inhomogeneous direction co-ordinates

$$\mathscr{W}[f_{(\sigma)}] \overset{\text{def}}{=} \int_\sigma d\Omega_0^{(\sigma)}\, f_{(\sigma)}(x, \xi) = 1 \tag{5.2.12}$$

is fulfilled.

In spite of the fact that — having non-stationary processes in mind — the investigation of systems of gaseous particles with finite life-time suggests a lot of very fascinating questions, this general problem will not be treated in the following.

Consider the special expression (5.2.9) of the "reduced" distribution function $f_{(\sigma)}(x, \xi)$ and let us determine the probability $f_{(\sigma)}(x, \xi) d\Omega_0^{(\sigma)}$ that the phase-point of the gaseous particle investigated can be found in the $(3+3)$-dimensional invariant phase-space volume-element; i.e., the probability that the trajectory of the particle crosses the mentioned surface-element of the space-like hypersurface σ.

Due to the fact that the conditions of normalization (5.2.11) and (5.2.13) are invariants of the transformations of the general group $\mathcal{G}$ of the relativistic phase-space, the probability field $f_{(\sigma)}(x, p)$ or $f_{(\sigma)}(x, \xi)$ has to fulfil the stationary condition

$$\delta \mathscr{W} [f_{(\sigma)}] = 0 , \tag{5.2.13}$$

based on which the continuity equation of the probability field can be obtained.

Having the infinitesimal transformation

$$x^{\mu\prime} = x^\mu + dx^\mu , \quad \xi_{i'} = \xi_i + \xi_i \tag{5.2.14}$$

in mind, by the usual definition of the variation of the functional $\mathscr{W}[f_{(\sigma)}]$

$$\delta \mathscr{W}[f_{(\sigma)}] \stackrel{\text{def}}{=\!=} \int_{\Omega_0^{(\sigma)\prime}} d\Omega_0^{(\sigma)\prime} f'_{(\sigma)}(x' \xi') - \int_{\Omega_0^{(\sigma)}} d\Omega_0^{(\sigma)} f_{(\sigma)}(x, \xi) , \tag{5.2.15}$$

due to the expansion up to the first order in dx^μ and $d\xi_i$

$$\int_{\Omega_0^{(\sigma')}} d\Omega_0^{(\sigma)\prime} f'_{(\sigma)}(x', \xi') \approx \int_{\Omega_0^{(\sigma)}} d\Omega_0^{(\sigma)} f_{(\delta)}(x, \xi) + \int_{\Omega_0^{(\delta)}} d\Omega_0^{(\sigma)} \left\{ \frac{\partial f_{(\sigma)}}{\partial x^\mu} dx^\mu + \frac{\partial f_{(\sigma)}}{\partial \xi_i} d\xi_i \right\} \tag{5.2.16}$$

and to the familiar supposition that dx^μ and $d\xi_i$ on the boundary of $\Omega_0^{(\sigma)}$ are vanishing, the equation of continuity can be obtained either as

$$D f_{(\sigma)} \stackrel{\text{def}}{=\!=} \frac{\partial f_{(\sigma)}}{\partial x^\mu} dx^\mu + \frac{\partial f_{(\sigma)}}{\partial \xi_i} d\xi_i , \tag{5.2.17}$$

or in its final form

$$\frac{D f_{(\sigma)}}{d\tau} = \frac{\partial f_{(\sigma)}}{\partial x^\mu} p^\mu + \frac{\partial f_{(\sigma)}}{\partial \xi_i} \frac{d}{d\tau} \xi_i = 0 . \tag{5.2.18}$$

It will be shown that this equation of continuity is exactly the relativistic generalization of Boltzmann's transport equation without the collision term. The general transport equation will be derived in the next Section 5.3.

Finally, let us discuss the connection between the reduced distribution function $f_{(\sigma)}(x, p)$ of the relativistic theory and that of non-relativistic one denoted by

$$f_{nr}(x; p) \equiv f_{nr}\,[t, \mathbf{x}; \mathbf{p}] \equiv f_{nr}\,[\mathbf{x}(t); \mathbf{p}(t)] = f_{nr}[\mathbf{x}; \varepsilon, \boldsymbol{\nu}]\,, \qquad (5.2.19)$$

where in the last version of f_{nr} ε means again the energy of the gaseous particle considered and $\boldsymbol{\nu}$ is a unit vector having the same direction as its three-momentum directed into the infinitesimal solid angle $d\omega$.

It is well known that the distribution function of the non-relativistic theory determines the probability that the phase-point $\{\mathbf{x}(t); \mathbf{p}(t)\}$ can be found in the volume-element $d\varepsilon d\omega d\mathscr{V}$ of the phase-space. Moreover, the invariant (3 + 3)-dimensional phase-space volume-element $d\Omega_0^{(\sigma)}$ on the hypersurface σ defined pro unit length of arch along the world-line of the gaseous particles fulfils Liouville's Theorem just as the non-relativistic phase-space volume-element does. Consequently, if in the rest frame of reference $\mathscr{K}^\circ$ one introduces local pseudo-Euclidean metric, then keeping the expression (4.5.24) of $d\Omega^0_{(\sigma)}$ in mind the connection

$$f_{(\sigma^\circ)}(x, p)\, d\Omega_0^{(lpE)} \equiv f_{(\sigma^\circ)}(x, p)\, \varepsilon p\, d\varepsilon\, d\omega\, d\mathscr{V}^{(\sigma^\circ)} = f_{nr}[\mathbf{x}, \varepsilon, \boldsymbol{\nu}]\, d\varepsilon\, d\omega\, d\mathscr{V} \quad (5.2.20)$$

may be obtained. Owing to the fact that in this case

$$d\mathscr{V}^{(\sigma^0)} \equiv d^3 x \equiv d\mathscr{V}$$

— keeping the normalization condition (2.2.1) of the four-momentum in mind — one gets

$$f_{(\sigma_0)}(x, p) = (\varepsilon p)^{-1} f_{nr}\,[\boldsymbol{\alpha}; \varepsilon, \boldsymbol{\nu}] = \varepsilon^{-1} \{\varepsilon^2 - m_0^2\}^{-1/2} f_{nr}\,[\mathbf{x}; \varepsilon, \boldsymbol{\nu}]\,. \quad (5.2.21)$$

Since $f_{(\sigma)}(x, p)$ is a scalar, this gives its value in any other frame as well; alternatively, it can be used to infer the transformation properties of

$$f_{nr}\,[\mathbf{x}; \varepsilon\, \boldsymbol{\nu}]\,.$$

In order to simplify our writing the label σ of the distribution functions $f_{(\sigma)}(x, p)$ and $f_{(\sigma)}(x, \xi)$ will be omitted in the following.

5.3. *The collision integral and the relativistic transport equation of Boltzmann.* In order to characterize the dynamics of the binary collisions of the gaseous particles, we consider an elastic collision between two particles with rest mass $m_0 = 1$. The initial momenta p^μ and $'p^\mu$ and the final ones $\tilde{p}^\mu$ and $'\tilde{p}^\mu$ all satisfy the normalization condition

$$g_{\mu\nu}\, p^\mu\, p^\nu = 1 \qquad (5.3.1)$$

and the conservation of four-momenta is expressed by

$$p^{\mu} + {}'p^{\mu} = \tilde{p}^{\mu} + {}'\tilde{p}^{\mu}. \tag{5.3.2}$$

In terms of the corresponding inhomogeneous direction co-ordinates — denoted by ξ_i, $'\xi_i$, $\tilde{\xi}_i$ and $'\tilde{\xi}_i$, respectively — the conservation law may be written as

$$\xi_i + {}'\xi_i = \xi_i + {}'\tilde{\xi}. \tag{5.3.3}$$

Denote $\mathscr{W}(\xi, {}'\xi; \tilde{\xi}, {}'\tilde{\xi})$ the probability density that a particle with the momentum p^{μ} (resp. ξ_i) via the considered elastic collision with a second particle with momentum $'p^{\mu}$ (resp. $'\xi_i$) changes its momentum into $\tilde{p}^{\mu}$ (resp. $\tilde{\xi}_i$) and simultaneously the second one into $'\tilde{p}^{\mu}$ (resp. $'\tilde{\xi}_i$). Let us suppose that such a collision means contact interaction among the particles which takes place at a point $\{x^{\mu}\}$ of the configuration space.

The probability that an arbitrary particle considered is in the neighbourhood dV of the point $\{x^{\mu}\}$ of the configuration-space with inhomogeneous direction co-ordinates in the range $(\xi_i, \xi_i + d\xi_i)$, i.e., with phase-point in the volume-element $d\Omega = \sqrt{-g}\, d^4x d^3\xi$ is given by $f(x, \xi)\sqrt{-g} d^4x d^3\xi$. Furthermore, the probability that this particle collides just at the point $\{x^{\mu}\}$ with an other gaseous particle with inhomogeneous direction co-ordinates in the range $('\xi_i, {}'\xi_i + d'\xi_i)$ is determined by $f(x, \xi) f(x, {}'\xi) \sqrt{-g} d^4x d^3\xi d^{3}{}'\xi$, since the probability that the second particle is at the point $\{x^{\mu}\}$ of the configuration-space where the collision takes place with inhomogeneous direction co-ordinates in the volume-element $d^{3}{}'\xi$ of the momentum-space is yielded by $f(x, {}'\xi) d^{3}{}'\xi$. Owing to the definition of $\mathscr{W}(\xi'\xi; \tilde{\xi}'\tilde{\xi})$ this means, however, that the probability that via the considered collision the two particles change their inhomogeneous direction co-ordinates into $\{\tilde{\xi}_i\}$ and $\{'\tilde{\xi}_i\}$ being in the volume-elements $d^3\tilde{\xi}$ and $d^{3}{}'\tilde{\xi}$ of the momentum-space, respectively, is obviously obtained by

$$\mathscr{W}(\xi, {}'\xi; \xi, {}'\tilde{\xi}) f(x, \xi) f(x, {}'\xi) \sqrt{-g}\, d^4 x d^3 \xi d^3\, {}'\xi d^3\, \tilde{\xi} d^{3}{}'\tilde{\xi} \tag{5.3.4}$$

being just the *transition probability* corresponding to the momentum changes of the particular binary collision considered. But, the momentum of the second particle may be arbitrary, therefore, in order to determine the *total probability of all possible binary collisions* at the point $\{x^{\mu}\}$ of the space-time continuum of the considered particle with phase-point in the volume-element $d\Omega$ of the phase-space with the other particles of the gaseous system, one has to integrate over the inhomogeneous direction co-ordinates $\{'\xi_i\}$, $\{\tilde{\xi}_i\}$ and $\{'\tilde{\xi}_i\}$, respectively. This means that the total probability to be looked for is given by

$$f(x, \xi) \sqrt{-g}\, d^4 x d^3 \xi \int d^3\, {}'\xi \int d^3\, \tilde{\xi} \int d^3\, {}'\tilde{\xi}\, \mathscr{W}(\xi, {}'\xi; \tilde{\xi}, {}'\tilde{\xi}) f(x, {}'\xi)\, \delta(\xi + {}'\xi - \tilde{\xi} - {}'\tilde{\xi}), \tag{5.3.5}$$

where in terms of the Dirac-function

$$\delta(\xi + {}'\xi - \tilde{\xi} - {}'\tilde{\xi}) \overset{\text{def}}{=} \prod_{i=1}^{3} \delta(\xi_i + {}'\xi_i - \tilde{\xi}_i - {}'\tilde{\xi}_i)\,, \tag{5.3.6}$$

due to eq. (5.3.3), the conservation of momenta is taken into account.

As an inverse collision to the binary collision with momentum change $\{\xi, {}'\xi\} \to \{\tilde{\xi}, {}'\tilde{\xi}\}$ the elastic collision with momentum change $\{\tilde{\xi}, {}'\tilde{\xi}\} \to \{\xi, {}'\xi\}$ can be regarded, the transition probability of which — having the analogous eq. (5.3.4) in mind — can be obtained as

$$\mathscr{W}(\tilde{\xi}, {}'\tilde{\xi}; \xi, {}'\xi) f(x, \tilde{\xi}) f(x, {}'\tilde{\xi}) \sqrt{-g}\, d^4 x d^3 \tilde{\xi} d^3 {}'\tilde{\xi} d^3 \xi d^3 {}'\xi; \tag{5.3.7}$$

i.e., the total probability of the inverse processes are given by

$$\sqrt{-g}\, d^4 x d^3 \xi \int d^3 \tilde{\xi} \int d^3 {}'\tilde{\xi} \int d^3 {}'\xi\, \mathscr{W}(\tilde{\xi}, {}'\tilde{\xi}; \xi, {}'\xi) f(x, \tilde{\xi}) f(x, {}'\tilde{\xi})\, \delta(\tilde{\xi} + {}'\tilde{\xi} - \xi - {}'\xi)\,. \tag{5.3.8}$$

By the binary collisions the stationarity of the probability field $f(x, \xi)$ will in general be destroyed. This means that the probability of finding the particle with phase-point in the volume-element $d\Omega$ due to the collisions will be decreased and due to the inverse collisions will be increased, respectively; i.e., the change of $f(x, \xi)$ pro unit of the proper time τ can be given in terms of

$$\begin{aligned} \frac{Df}{d\tau} = &- \int d^3{}'\xi \int d^3\tilde{\xi} \int d^3{}'\tilde{\xi}\, \mathscr{W}(\xi, {}'\xi; \tilde{\xi}, {}'\tilde{\xi}) f(x, \xi) f(x, {}'\xi)\, \delta(\xi + {}'\xi - \tilde{\xi} - {}'\tilde{\xi}) + \\ &+ \int d^3{}'\xi \int d^3\tilde{\xi} \int d^3{}'\tilde{\xi}\, \mathscr{W}(\tilde{\xi}, {}'\tilde{\xi}; \xi, {}'\xi) f(x, \tilde{\xi}) f(x, {}'\tilde{\xi})\, \delta(\tilde{\xi} + {}'\tilde{\xi} - \xi - {}'\xi)\,. \end{aligned} \tag{5.3.9}$$

We have not any reason to suppose that the transition probability of inverse binary collisions would not be the same as the original ones. Therefore, the obvious assumptions of *microscopic reversibility*

$$\mathscr{W}({}'\xi, \xi; {}'\tilde{\xi}, \xi) = \mathscr{W}(\xi, {}'\xi; \xi, {}'\tilde{\xi}) \quad \text{and} \quad \mathscr{W}(\tilde{\xi}, {}'\tilde{\xi}; \xi, {}'\xi) = \mathscr{W}(\xi, {}'\xi; \xi, {}'\tilde{\xi})\,, \tag{5.3.10}$$

respectively, may be suggested.

Bearing in mind this microscopic reversibility as well as the positivity of the Dirac-function (5.3.6), the right-hand side of eq. (5.3.9) can be written in the form:

$$\begin{aligned} \left[\frac{Df}{d\tau}\right]_{\text{coll}} \overset{\text{def}}{=} &\int d^3 {}'\xi \int d^3 \tilde{\xi} \int d^3 {}'\tilde{\xi}\, \mathscr{W}(\xi, {}'\xi; \tilde{\xi}, {}'\tilde{\xi}) \times \\ &\times \{f(x, \xi) f(x, {}'\xi) - f(x, \tilde{\xi}) f(x, {}'\tilde{\xi})\}\, \delta(\xi + {}'\xi - \tilde{\xi} - {}'\tilde{\xi})\,. \end{aligned} \tag{5.3.11}$$

This means, however, that owing to eq. (5.2.10) the above result (5.3.9) has the final form:

$$\frac{\partial f}{\partial x^\mu} p^\mu + \frac{\partial f}{\partial \xi_i} \frac{d}{d\tau} \xi_i = - \left[\frac{Df}{d\tau} \right]_{\text{coll}} \tag{5.3.12}$$

being exactly *the general Boltzmann's transport equation of relativistic homogeneous gas systems in arbitrary metrical space-time continuum with any intrinsic dynamical symmetry properties.*

5.4. *Relativistic transport equation in particular cases recently investigated.* Considering an otherwise general homogeneous gaseous system without internal dynamical anisotropy, the underlying geometrical background of the hyper-geometrized phase-space formalism is a Riemannian space-time continuum. If the special frame of reference (4.1.10) and the orientation (2.4.6) of the λ^+-triad is introduced — due to the definition (3.2.4) of the inhomogeneous direction co-ordinates — we have

$$\xi_i = p_i = g_{i\mu} p^\mu \tag{5.4.1}$$

and the transport equation (5.3.12) can be written either in the form:

$$\frac{\partial f}{\partial x^\mu} p^\mu + \left\{ \frac{\partial f}{\partial p_0} \frac{\partial p_0}{\partial p^i} + \frac{\partial f}{\partial p^i} \right\} \frac{dp^i}{d\tau} = - \left[\frac{Df}{d\tau} \right]_{\text{coll}}, \tag{5.4.2}$$

or as

$$\frac{\partial f}{\partial x^\mu} p^\mu + \frac{\partial f}{\partial p^\mu} \frac{dp^\mu}{d\tau} = - \left[\frac{Df}{d\tau} \right]_{\text{coll}}. \tag{5.4.3}$$

However, based on eq. (5.1.2) in this case the relativistic Boltzmann's equation can be put in the final form:

$$p^\mu \nabla_\mu f + K^\mu \frac{\partial f}{\partial p^\mu} = - \left[\frac{Df}{d\tau} \right]_{\text{coll}} \left(\nabla_\mu \overset{\text{def}}{=} \frac{\partial}{\partial x^\mu} - \widetilde{\Gamma}^{\varkappa}_{*\mu} \frac{\partial}{\partial p^\varkappa} \right) \tag{5.4.4}$$

which, with other argumentation in mind, was proposed by KEN-ITI GOTO [4].

As a further specialization the pseudo-Euclidean space-time background has to be dealt with when eq. (5.3.12) is reduced to

$$\frac{\partial f}{\partial x^0} p^0 + \frac{\partial f}{\partial x^i} p^i + \frac{\partial f}{\partial p^i} \frac{dp^i}{d\tau} = - \left[\frac{df}{d\tau} \right]_{\text{coll}}. \tag{5.4.5}$$

But, in this particular case the components of the four-momentum are

$$p^\mu \equiv \{m, mv^i\}, \qquad \left(m = m_0 \{1 - v^2\}^{-1/2}\right) \tag{5.4.6}$$

so that in terms of the familiar three-vector formalism with the abbreviations

$$\boldsymbol{\nabla} \overset{\text{def}}{=} \{\partial/\partial x^i\}, \boldsymbol{\nabla}_v \overset{\text{def}}{=} \{\partial/\partial v^i\}, \mathbf{v} \overset{\text{def}}{=} \{v^i\}, \mathbf{K} \overset{\text{def}}{=} \{K^i\} \quad (i = 1, 2, 3) \tag{5.4.7}$$

it can finally be obtained that

$$\frac{\partial f}{\partial t} + v^0 \mathbf{v} f + \frac{1}{m} \mathbf{K}^{\circ} \boldsymbol{\nabla}_r f = -\frac{1}{m} \left[\frac{df}{d\tau} \right]_{\text{coll}} \tag{5.4.8}$$

which is the familiar form of Boltzmann's transport equation.

A detailed discussion of the connections between different particular forms of the relativistic Boltzmann's equation and their solutions in the case of special gaseous systems lies outside the scope of this paper. Such investigations were most recently published by Abonyi [5].

5.5. *Outline of a general relativistic thermodynamics.* In order to develop a generalized relativistic thermodynamics of gaseous systems with intrinsic dynamical anisotropy it seems to be most straightforward to bear in mind the celebrated theories suggested by Kluitenberg, De Groot and Mazur [44], Israel [9] and others [45, 46].

The *mass-current, stress energy-momentum tensor* of the considered homogeneous gaseous system with N particles in the hyper-geometrized space-time, i.e., the configuration space, is usually defined by

$$M^{\mu}(x) \overset{\text{def}}{=} N \int dP f(x, p)\, p^{\mu} \tag{5.5.1}$$

and

$$T^{\mu\nu}(x) \overset{\text{def}}{=} N \int dP f(x, p)\, p^{\mu} p^{\nu}, \tag{5.5.2}$$

respectively. It can be proved that these fundamental quantities of the statistical thermodynamics statisfy the conservation laws:

$$M^{\mu}_{\cdot\,|\mu} = 0\,, \tag{5.5.3}$$

as well as

$$T^{\mu\nu}_{\cdot\,\cdot\,|\nu} = 0\,, \tag{5.5.4}$$

where the vertical bar denotes covariant differentiation. The latter eq. (5.5.4) means the relativistic formulation of the first law of thermodynamics having in mind isolated systems, i.e., systems in absence of external fields of forces [39]; however, the gravitational interactions of the particles are taken into account in terms of a background gravitational field determined in self-consistent way [9].

Indeed, let $\Psi(x, p)$ be an arbitrary tensor function of the line-element (x, p), then multiplication of the Boltzmann's equation (5.4.4) without external field ($K^{\mu} \equiv 0$) by $\Psi(x, p)$ and its integration yields:

$$\int dP\,\Psi(x,p)\,p^{\mu}\frac{\partial f}{\partial x^{\mu}} = \mathscr{J}\,[\Psi] \tag{5.5.5}$$

with

$$\begin{aligned}\mathscr{J}[\Psi] &\overset{\text{def}}{=} -\int dP\,\Psi(x,p)\left[\frac{Df}{d\tau}\right]_{\text{coll}} = \\ &= -\int dP\int d'P\int d\tilde{P}\int d'\tilde{P}\,\Psi\,\{f\,{}'f - \tilde{f}\,{}'\tilde{f}\}\times \\ &\quad \delta(\xi + {}'\xi - \tilde{\xi} - {}'\tilde{\xi})\,\mathscr{W}(\xi,{}'\xi;\tilde{\xi},{}'\tilde{\xi})\,.\end{aligned} \tag{5.5.6}$$

$$\begin{aligned}\mathscr{J}[\Psi] &= -\frac{1}{4}\int d'P\int d'P\int d\tilde{P}\int d'\tilde{P}\,\mathscr{W}\{\Psi + {}'\Psi - \\ &\quad - \tilde{\Psi} - {}'\tilde{\Psi}\}\,\delta(\xi + {}'\xi - \tilde{\xi} - {}'\tilde{\xi})\,\{f\,{}'f - \tilde{f}\,{}'\tilde{f}\}\,,\end{aligned} \tag{5.5.7}$$

where the symmetry relations (5.3.10) have been used and the obvious abbreviations

$$\Psi \overset{\text{def}}{=} \Psi(x,p),\quad f \overset{\text{def}}{=} f(x,p),\quad {}'\Psi \overset{\text{def}}{=} \Psi(x,{}'p),\quad {}'f \overset{\text{def}}{=} f(x,{}'p)\ \text{etc.} \tag{5.5.8}$$

were introduced. Owing to the conservation of momenta (5.3.2) one can immediately see that

$$\mathscr{J}\,[\Psi] = 0 \quad \text{if } \Psi = 1 \quad \text{or } p^{\mu}, \tag{5.5.9}$$

based on which the statements of eqs. (5.5.3) and (5.5.4) are proved.

The *entropy flux vector* is usually defined by

$$S^{\mu}(x) \overset{\text{def}}{=} -Nk\int dPf(x,p)\,ln\,\{f(x,p)\}\,p^{\mu}\,, \tag{5.5.10}$$

where k is Boltzmann's constant and it may also be obtained that

$$S^{\mu} = \Theta_{\nu}\,T^{\mu\nu}\,, \tag{5.5.11}$$

where owing to eq. (3.4.5)

$$\Theta^{\mu} \overset{\text{def}}{=} l^{\mu}/T \tag{5.5.12}$$

is a time-like vector, the so-called *temperature vector* being T the absolute temperature.

Using eqs. (5.5.5) and (5.5.7) with

$$\Psi = \ln\{f(x,p)\} \tag{5.5.13}$$

one finds

$$S^{\mu}_{\cdot|\mu} \geqq 0 \tag{5.5.14}$$

which expresses *Boltzmann's H-theorem*, i.e., the *second law of thermodynamics.*

In the case of gaseous systems with intrinsically isotropic dynamical interactions the underlying line-element space reduces into a Riemannian space-time and the considerations above do not mean any generalization. However, in the general case if in the line-element space the absolute parallelism of line-elements exists a field of directions

$$v^{\mu} = v^{\mu}(x) \tag{5.5.15}$$

may be introduced satisfying eq. (3.4.6) and the usual Riemannian formalism can be translated into the language of the line-element spaces in terms of the *osculate Riemannian spaces* which is defined by means of the postulates:

(a) The metrical fundamental tensor $\bar{g}_{\mu\nu}(x)$ of the osculate Riemannian space is identical in respect of the field of directions $v^{\mu}(x)$ with the metrical fundamental tensor of the line-element space, i.e.,

$$\bar{g}_{\mu\nu}(x) \equiv g_{\mu\nu}(x, v(x)) \tag{5.5.16}$$

and owing to the homogeneity of zero order in the variable v^{μ}

$$\bar{g}_{\mu\nu}(x) = g_{\mu\nu}(x, l(x)) = g_{\mu\nu}(x, p(x)); \tag{5.5.17}$$

(b) The geodetic lines of both spaces osculate each other;

(c) The invariant differential and the covariant derivatives of vectors are identical in both spaces;

(d) The tensors of principal curvature of both spaces are the same [15, 17].

Of course, the above considerations may also be obtained in terms of the λ-triad formalism. The definition of the most important thermodynamic quantities, such as mass-current, stress energy-momentum tensor and entropy-flux vector can be given, e.g., as follows

$$\mathscr{M}_i(x) = N m_0^3 \int d^3 \xi f(x, \xi)\, \xi_i\,, \tag{5.5.18}$$

$$\mathscr{T}_{ik}(x) = N m_0^3 \int d^3 \xi f(x, \xi)\, \xi_i\, \xi_k \tag{5.5.19}$$

and

$$\mathscr{S}_i(x) = N m_0^3 \int d^3 \xi f(x, \xi) \ln \{f(x, \xi)\}\, \xi_i\,, \tag{5.5.20}$$

respectively.

More detailed discussion of these problems connected with the relativistic generalization of statistical thermodynamics keeping the results and different suggestions of recent authors [6, 44—46] in mind will be published elsewhere.

I am very grateful to Professor H. Fröhlich for many helpful discussions and for his kind interest in these investigations. I am also indebted to Professor G. Marx for his stimulating criticism concerning the problems connected with

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REFERENCES

42. J. I. Horváth, Acta Phys. Hung. **24**, 205, 1968.
43. J. I. Horváth, Acta Phys. Hung. **24**, 347, 1968.
44. G. A. Kluitenberg, S. R. De Groot and P. Mazur., Physica **19**, 689, 1079, 1953; **20**, 199, 1953; **21**, 148, 169, 1954.
45. S. R. De Groot, Nucl. Energy., (C); *Plasma Physics*, Vol. **2**, 188, 1961.
46. G. Neugebauer, Wiss. Zs. d. Univ. Jena, **13**, 209, 1964; **15**, 161, 1966.

О СВЕРХ-ГЕОМЕТРИЗАЦИИ РЕЛЯТИВИСТСКОГО ФАЗОВО-ПРОСТРАНСТВЕННОГО ФОРМАЛИЗМА III

Я. И. ГОРВАТ

Резюме

В первых двух частях данной работы исследовались определение и геометрическая этруктура релятивистического фазового пространства; позднее дискутировалась идея сверхгеометризации, причем некоторые из прежних результатов вновь сформулировались. В настоящее время предложенный ранее метод применялся для развития релятивистической кинетической теории газов. Дискуссия начинается с разными определениями функции распределения, далее выводится релятивистическое уравнение переноса Больцманна и полученные общие результаты проверяются в специальных случаях, ранее исследованных несколькими авторами. Наконец, базируясь на теории Израиля, предлагается очертание основ общей релятивистической термодинамики.

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УЧЁТ АНГАРМОНИЧЕСКИХ ЭФФЕКТОВ ВЫСШИХ ПОРЯДКОВ В КРИСТАЛЛАХ

Н. М. ПЛАКИДА и Т. ШИКЛОШ
ОБЪЕДИНЕННЫЙ ИНСТИТУТ ЯДЕРНЫХ ИССЛЕДОВАНИЙ, ЛАБОРАТОРИЯ ТЕОРЕТИЧЕСКОЙ ФИЗИКИ, ДУБНА, СССР

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В работе развит метод учёта ангармонических членов высших порядков в потенциальной энергии кристалла. На основе уравнений для функций Грина найден спектр частот колебаний решётки в псевдогармоническом приближении, а также определено их затухание.

Введение

Обычно при рассмотрении ангармонических эффектов в кристаллах ограничиваются учётом ангармонических членов низших порядков — третьего и четвертого (см. например, [1]), которые учитывают по теории возмущений. При этом наблюдается относительно хорошее согласие теории и эксперимента вплоть до температур, равных примерно половине температуры плавления [1], что свидетельствует о достаточно слабом взаимодействии нормальных колебаний решетки, которые определяются из гармонического приближения. Однако, в некоторых случаях, как например, вблизи температуры фазового перехода, в квантовых кристаллах при низкой температуре, при изучении колебаний легкой слабосвязанной примеси и др. необходимо учесть высшие ангармонические члены в потенциальной энергии кристалла. В связи с этим в работах [2] была предложена «псевдогармоническая» теория кристаллов, основанная на построении эффективного самосогласованного гармонического гамильтониана, приближенно описывающего ангармонический кристалл. По существу, эта теория учитывает изменение частоты нормальных колебаний за счет ангармонических членов в потенциальной энергии, но в ней не рассматривается взаимодействие между нормальными колебаниями, найденными в «псевдогармоническом» приближении. Однако это взаимодействие может приводить к сильному затуханию нормальных колебаний и модель слабосвязанных «псевдогармонических» фононов будет плохо описывать реальный ангармонический кристалл.

В настоящей работе мы сформулируем приближенный метод учёта ангармонизмов высших порядков в кристаллах, позволяющий относительно просто определить спектр частот колебаний кристалла, а также учесть их затухание, связанное с простейшими процессами взаимодействия. При этом здесь мы ограничимся формальным вычислением сдвига частоты фононов и их затухания, а численные оценки для различных моделей рассмотрим в отдельной работе.

1. Гамильтониан ангармонического кристалла

Будем исходить из весьма общей модели ангармонического кристалла. Рассмотрим кристаллическую решетку из N атомов, гамильтониан которой может быть записан в виде:

$$\mathcal{H} = \sum_{l} \frac{\mathbf{P}_l^2}{2M_l} + U(\mathbf{R}_1, \ldots, \mathbf{R}_N)\,, \tag{1.1}$$

где $\mathbf{P}_l$ — оператор импульса, $\mathbf{R}_l = \langle \mathbf{R}_l \rangle + \mathbf{u}_l$ положение атома с массой M_l в узле $\boldsymbol{l}$, равновесное положение которого $\langle \mathbf{R}_l \rangle = \boldsymbol{l}$ определяется из условий равенства нулю средней силы, действующей на данный атом:

$$\frac{\partial}{\partial \boldsymbol{l}} \langle U(\mathbf{R}_1 \ldots \mathbf{R}_N) \rangle = 0\,, \tag{1.2}$$

где среднее $\langle \ldots \rangle$ берется по равновесному состоянию кристалла с гамильтонианом (1.1). Если имеются внешние силы, создающие напряжения в кристалле, то (1.2) следует приравнять этим силам $\mathbf{f}_l$. Таким образом, $\mathbf{u}_l = \mathbf{R}_l - \langle \mathbf{R}_l \rangle$ определяет малое смещение атома из положения равновесия при заданной температуре T. Поэтому потенциальная энергия U в (1.1) может быть разложена в ряд по этим смещениям:

$$U(R_1, \ldots, R_N) = U_0 + \sum_{n=1}^{\infty} \frac{1}{n!} \sum_{l_1 \ldots l_n} \Phi_{l_1 \ldots l_n}^{\alpha_1 \ldots \alpha_n} u_{l_1}^{\alpha_1} \ldots u_{l_n}^{\alpha_n}, \tag{1.3}$$

где

$$\Phi_{l_1 \ldots l_n}^{\alpha_1 \ldots \alpha_n} = \left[\frac{\partial^n}{\partial l_1^{\alpha_1} \ldots \partial l_n^{\alpha_n}} U(\mathbf{R}_1, \ldots, \mathbf{R}_N) \right]_{\mathbf{u}_i = 0}.$$

Обычно, квадратичная форма по операторам P_n, u_n:

$$H_0 = \sum_{l} \frac{\mathbf{P}_l^2}{2\,M_l} + \frac{1}{2} \sum_{l_m} \Phi_{lm}^{\alpha\beta} u_l^{\alpha} u_m^{\beta} \tag{1.4}$$

— гармоническая часть гамильтониана (1.1), диагонализуется с помощью ортогонального преобразования:

$$u_l^{\alpha} = \sum_{k} \frac{e_k^{\alpha}(l)}{(2\,M_l\,\omega_k)^{1/2}} A_k\,, \tag{1.5а}$$

$$P_l^{\alpha} = \frac{1}{i} \sum_{k} \left(\frac{M_l\,\omega_k}{2} \right)^{1/2} \overset{+}{e}{}_k^{\alpha}(l)\; B_k\,, \tag{1.5б}$$

где вектора нормальных колебаний $e_k^a(l)$ образуют полный и ортонормированный базис:

$$\begin{aligned} \sum_k \overset{+}{e}{}_k^\alpha(l)\, e_k^\beta(l') &= \delta_{l,l'}^{\alpha\beta}, \\ \sum_l \overset{+}{e}{}_k^\alpha(l)\, e_{k'}^\alpha(l) &= \delta_{k,k'} \end{aligned} \tag{1.6}$$

а частоты нормальных колебаний ω_k определяются из уравнения на собственные значения:

$$\sum_{l'} \Phi_{ll'}^{\alpha\beta} \frac{1}{\sqrt{M_l M_{l'}}} e_k^\beta(l') = \omega_k^2\, e_k^\alpha(l)\,. \tag{1.7}$$

В случае решетки без примесей нормальные колебания имеют вид плоских волн, так что вектора

$$e_k^\alpha(l) = \frac{1}{\sqrt{N}} e^{i\mathbf{kl}}\, w^\alpha(\mathbf{k}, j)\,, \tag{1.8}$$

где $w^a(\mathbf{k}, j)$ определяет нормальное колебание с волновым вектором $\mathbf{k}$ сорта j. Операторы A_k и B_k в (1.5) могут быть представлены через операторы рождения и уничтожения фононов $\overset{+}{a}_{kj}$ и a_{kj}:

$$\begin{aligned} A_{\mathbf{k}j} &= a_{\mathbf{k}j} + a_{-\mathbf{k}j}^+ = A_{-\mathbf{k}j}^+\,, \\ B_{\mathbf{k}j} &= a_{\mathbf{k}j} - a_{-\mathbf{k}j}^+ = -\,B_{-\mathbf{k}j}^+\,. \end{aligned} \tag{1.9}$$

Совершая преобразование (1.5), гамильтониан (1.1) запишем в виде:

$$\mathscr{H} = H_o + H_a\,, \tag{1.1a}$$

$$H_o = \frac{1}{4} \sum_k \omega_k (A_k^+ A_k + B_k^+ B_k)\,, \tag{1.10}$$

$$H_a = \sum_{n=1,3,\ldots}^{\infty} \frac{1}{n!} \sum_{k_1\ldots k_n} V_n(k_1 \ldots k_n)\, A_{k_1} \ldots A_{k_n}\,, \tag{1.11}$$

где

$$V_n(k_1 \ldots k_n) = \sum_{l_1 \ldots l_n} \Phi_{l_1 \ldots l_n}^{\alpha_1 \ldots \alpha_n} \prod_{i=1}^{n} \frac{e_{ki}^{\alpha_i}(l_i)}{(2M_{l_i}\, \omega_{ki})^{1/2}}\,. \tag{1.12}$$

В ряде случаев хорошим приближением является модель кристалла с парными силами:

$$U(\mathbf{R}_1, \ldots, \mathbf{R}_N) = \frac{1}{2} \sum_{l \neq m}^{N} \Phi_{lm}(\mathbf{R}_l - \mathbf{R}_m)\,. \tag{1.13}$$

Вводя фурье-образ потенциала взаимодействия между l-ым и m-ым атомами в виде:

$$\Phi_{lm}(q) = \frac{1}{\Omega} \int d^3 R \, \Phi_{lm}(\mathbf{R}) \, e^{-i\mathbf{q}\mathbf{R}}, \tag{1.14}$$

где интегрирование ведется по объему кристалла Ω, потенциал (1.13) представим в виде:

$$\begin{aligned} U(\mathbf{R}_1, \ldots, \mathbf{R}_N) &= \frac{1}{2} \sum_{l \neq m}^{N} \sum_{q} \Phi_{lm}(q) \, e^{i\mathbf{q}(\mathbf{l}-\mathbf{m})} \, e^{i\mathbf{q}(\mathbf{u}_c - \mathbf{u}_m)} = \\ &= \frac{1}{2} \sum_{l \neq m}^{N} \sum_{q} \Phi_{lm}(q) \, e^{i\mathbf{q}(\mathbf{l}-\mathbf{m})} \sum_{n=0}^{\infty} \frac{1}{n!} \left\{ iq(u_l - u_m) \right\}^n . \end{aligned} \tag{1.15}$$

Если совершить преобразование (1.5), то гамильтониан примет прежний вид (1.10), (1.11), где функция

$$V_n(k_1 \ldots k_n) = \frac{1}{2} \sum_{l \neq m} \sum_{q} \Phi_{lm}(q) \, e^{i\mathbf{q}(\mathbf{l}-\mathbf{m})} \prod_{i=1}^{n} iq^{\alpha} \, b^{\alpha}_{lm}(k_i) , \tag{1.16}$$

$$b^{\alpha}_{lm}(k) = \left(\frac{e^{\alpha}_{k}(l)}{(2 \, M_l \, \omega_k)^{1/2}} - \frac{e^{\alpha}_{k}(m)}{(2 \, M_m \, \omega_k)^{1/2}} \right) . \tag{1.17}$$

Таким образом, гамильтониан (1.1) может быть записан в общем виде (1.10), (1.11), причем в некоторых случаях функции взаимодействия могут быть выбраны в более простом виде (1.15) или (1.16). При этом равновесные положения атомов $\boldsymbol{l} = \langle \mathbf{R}_l \rangle$ при данной температуре T следует определять из условий (1.2).

2. Уравнения для функций Грина. Псевдогармоническое приближение

При определении спектра частот колебаний решетки, а также при исследовании ряда равновесных свойств кристалла достаточно найти однофононную функцию Грина (см. например, [3]). Чтобы сделать изложение более простым, мы рассмотрим сначала уравнение для функции Грина в координатном представлении, выбрав модель кристалла с парными силами (1.15). Мы будем пользоваться функциями Грина типа [4, 5]

$$G^{\alpha\beta}_{ll'}(t - t') = \langle\langle u^{\alpha}_{l}(t); \, u^{\beta}_{l'}(t') \rangle\rangle = - \, i\Theta(t - t') \, \langle [u^{\alpha}_{l}(t), u^{\beta}_{l'}(t')]_{-} \rangle . \tag{2.1}$$

Из уравнений движения для операторов $u_l(t)$ и $P_l(t)$ в гейзенберговском представлении с гамильтонианом (1.1), (1.15):

$$\begin{aligned} i \frac{d}{dt} u^{\alpha}_{l}(t) &= \frac{i}{M_l} P^{\alpha}_{l}(t) , \\ i \frac{d}{dt} i P^{\alpha}_{l}(t) &= \sum_{m \neq l} \sum_{q} \Phi_{lm}(q) \, e^{i\mathbf{q}(\mathbf{l}-\mathbf{m})} \, iq^{\alpha} \, e^{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)} \end{aligned} \tag{2.2}$$

получаем следующее уравнение для функции Грина (2.1):

$$\begin{aligned} M_l \left(i\frac{d}{dt}\right)^2 G_{ll'}^{\alpha\beta}(t-t') &= \delta(t-t')\,\delta_{ll'}^{\alpha\beta} + \\ &+ \sum_{m\neq l}\sum_{q} \Phi_{lm}(q)\, e^{i\mathbf{q}(\mathbf{l}-\mathbf{m})}\, iq^{\alpha} \langle\langle e^{i\mathbf{q}(\mathbf{u}_l-\mathbf{u}_m)};\ u_{l'}^{\beta}(t')\rangle\rangle . \end{aligned} \tag{2.3}$$

Очевидно, что записывая уравнение для функции Грина в правой части (2.3) мы придем к более сложной функции Грина и т. д., в результате будем иметь бесконечную цепочку уравнений, описывающую движение N атомов в решетке. Решение такой системы не представляется возможными и поэтому необходимо воспользоваться некоторым приближением. Предполагая, что корреляция в движении атомов уменьшается по мере увеличения их числа, функцию Грина в (2.3) представим в виде разложения по однофононным, двухфононным и т. д. функциям Грина, описывающим корреляцию двух, трех и т. д. фононов. Ограничиваясь низшими функциями Грина, получим:

$$\begin{aligned} \langle\langle e^{i\mathbf{q}(\mathbf{u}_l-\mathbf{u}_m)};\ u_{l'}^{\beta}(t')\rangle\rangle &= \sum_{n=1}^{\infty}\frac{1}{n!}\langle\langle\{i\mathbf{q}(\mathbf{u}_l-\mathbf{u}_m)\}^n;\ u_{l'}^{\beta}(t')\rangle\rangle \approx \\ \approx \sum_{n=1}^{\infty}\frac{1}{n!}&\langle\{i\mathbf{q}(\mathbf{u}_l-\mathbf{u}_m)\}^{n-1}\rangle\, n\, \langle\langle i\mathbf{q}(\mathbf{u}_l-\mathbf{u}_m);\ u_{l'}^{\beta}(t')\rangle\rangle + \\ + \sum_{n=1}^{\infty}\frac{1}{n!}&\langle\{i\mathbf{q}(\mathbf{u}_l-\mathbf{u}_m)\}^{n-2}\rangle\,\frac{n(n-1)}{2}\,\langle\langle\{i\mathbf{q}(\mathbf{u}_l-\mathbf{u}_m)\}^2;\ u_{l'}^{\beta}(t')\rangle\rangle , \end{aligned} \tag{2.4}$$

где множители n и $n(n-1)$ в правой части (2.4) появляются при подсчете числа возможных однофононных и двухфононных функций Грина. Подставляя (2.4) в (2.3) и выполняя суммирование по n получаем следующее уравнение для функции Грина:

$$\begin{aligned} M_l \left(i\frac{d}{dt}\right)^2 G_{ll'}^{\alpha\beta}(t-t') &= \delta(t-t')\,\delta_{l,l'}^{\alpha\beta} + \sum_{m}\tilde{\Phi}_{lm}^{\alpha\gamma}\, G_{ml'}^{\gamma\beta}(t-t') + \\ + \frac{1}{2}\sum_{m\neq l}\sum_{q}\Phi_{lm}(q)\, e^{i\mathbf{q}(\mathbf{l}-\mathbf{m})}&\langle e^{i\mathbf{q}(\mathbf{u}_l-\mathbf{u}_m)}\rangle\, iq^{\alpha}\, iq^{\gamma}\, iq^{\delta}\times\langle\langle(u_l^{\gamma}-u_m^{\gamma})\,(u_l^{\delta}-u_m^{\delta});\ u_{l'}^{\beta}(t')\rangle\rangle , \end{aligned} \tag{2.5}$$

где мы ввели «псевдогармоническую» силовую постоянную:

$$\tilde{\Phi}_{lm}^{\alpha\gamma} = \begin{cases} \displaystyle\sum_{q}\Phi_{lm}(q)\, e^{i\mathbf{q}(\mathbf{l}-\mathbf{m})}\, q^{\alpha} q^{\gamma}\,\langle e^{i\mathbf{q}(\mathbf{u}_l-\mathbf{u}_m)}\rangle & \text{при } l\neq m \\ \displaystyle-\sum_{q}\sum_{n\neq l}\Phi_{ln}(q)\, e^{i\mathbf{q}(\mathbf{l}-\mathbf{n})}\, q^{\alpha} q^{\gamma}\,\langle e^{i\mathbf{q}(\mathbf{u}_l-\mathbf{u}_n)}\rangle & \text{при } l = m \end{cases} \tag{2.6}$$

Очевидно, что подобное же уравнение мы получим и в случае более общего вида потенциальной энергии (1.3), причем в этом случае

$$\tilde{\Phi}^{\alpha\gamma}_{lm} = \frac{\partial^2}{\partial l_\alpha \, \partial m_\gamma} \langle U(\mathbf{R}_1, \ldots, \mathbf{R}_N) \rangle . \tag{2.6a}$$

Таким образом, мы приходим к результатам псевдогармонической теории [2] при учёте только первого члена в разложении многофононной функции Грина (2.4). Спектр частот колебаний решётки определяется полюсами фурье-образа функции Грина $G^{\alpha\beta}_{ll'}(\omega)$:

$$G^{\alpha\beta}_{ll'}(t-t') = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\omega(t-t')} \, G^{\alpha\beta}_{ll'}(\omega) \, d\omega \, , \tag{2.7}$$

и в псевдогармоническом приближении определяется из уравнения:

$$\sum_m \{ M_m \, \omega^2 \, \delta^{\alpha\beta}_{l,m} - \tilde{\Phi}^{\alpha\gamma}_{lm} \} \, G^{\gamma\beta}_{ml'}(\omega) = \delta^{\alpha\beta}_{l,l'} \tag{2.8}$$

При этом, в псевдогармоническом приближении

$$\langle e^{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)} \rangle = e^{-\frac{1}{2} \langle [\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)]^2 \rangle} , \tag{2.9}$$

так что необходимо определить лишь однофононную корреляционную функцию, которая согласно спектральной теореме [4] определяется через функцию Грина:

$$\langle u^\gamma_m \, u^\alpha_l \rangle = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{[-2 \, Im \, G^{\alpha\gamma}_{lm}(\omega + i\varepsilon)]}{e^{\omega/\Theta} - 1} \, d\omega \, . \tag{2.10}$$

Таким образом, система уравнений (2.6), (2.8)—(2.10) определяет свойства ангармонического кристалла в псевдогармоническом приближении [2].

Равновесные положения атомов $\boldsymbol{l} = \langle \mathbf{R}_l \rangle$ могут быть определены при усреднении уравнений движения (2.2):

$$i \frac{d}{dt} \langle i\mathbf{P}^\alpha_l \rangle = \sum_{m \neq l} \sum_q \Phi_{lm}(q) \, e^{i\mathbf{q}(\mathbf{l}-\mathbf{m})} \, iq^\alpha \langle e^{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)} \rangle = 0 \, . \tag{2.11}$$

Видно, что это уравнение совпадает с условием (1.2) равенства нулю средней силы, действующей на атом $\boldsymbol{l}$ в направлении α. Заметим при этом, что в псевдогармоническом приближении, когда выполняется соотношение (2.9), все четные степени в разложении потенциальной энергии определяют частоты колебаний решетки согласно (2.6), (2.8), а нечетные степени — равновесное положение атомов согласно (2.11). Результаты гармонического приближения немедленно получаются, если взять первый член разложения в (2.9) т. е. положить

$$exp\{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)\} \approx 1\cdot$$

При учете следующего члена в разложении (2.4) для многофононной функции Грина — двухфононной функции Грина, спектр колебаний решетки уже не определяется простым уравнением (2.8) — необходимо рассмотреть уравнение (2.5). При этом помимо дополнительной переновмировки частоты, возникает затухание колебаний, соответствующее возможным процессам распада возбуждения. Записывая уравнение для двухфононной функции Грина

$$G^{(1)\alpha\gamma\beta}_{lmn'}(t - t') = \langle\langle u^{\alpha}_{l}(t)\, u^{\gamma}_{m}(t);\, u^{\beta}_{n'}(t')\rangle\rangle \tag{2.12}$$

мы получим систему трех уравнений для функции (2.12) и ещё двух функций, полученных заменой операторов $\mathbf{u}_l$ на $\mathbf{P}_l$, решение которой не может быть выполнено в общем виде. Поэтому, в функции Грина (2.12) необходимо перейти к нормальным координатам согласно (1.5), Удобнее однако, совершить это преобразование в исходной функции (2.1), и рассмотреть уравнения движения для этой фужкции в импульсном представлении.

3. Затухание колебаний решетки

Совершая преобразование (1.5), гамильтониан системы запишем в виде (1.10), (1.11), Рассмотрим уравнение для однофононной функции Грина (см. [6]):

$$G_{kk'}(t - t') = \langle\langle A_k(t);\, A^{+}_{k'}(t')\rangle\rangle, \tag{3.1}$$

которая непосредственно связана с функцией Грина (2.1) преобразованием (1.5):

$$G^{\alpha\beta}_{ll'}(t - t') = \sum_{kk'} \frac{e^{\alpha}_{k}(l)\, e^{+\beta}_{k'}(l')}{2(M_l\, M_{l'}\, \omega_k\, \omega_{k'})^{1/2}}\, G_{kk'}(t - t')\,. \tag{3.2}$$

Из уравнений движения для операторов $A_k(t)$, $B_k(t)$ в гейзенберговском представлении с гамильтонианом (1.10), (1.11):

$$\begin{gathered} i\frac{d}{dt} A_k(t) = \omega_k\, B_k(t)\,, \\ i\frac{d}{dt} B_k(t) = \omega_k\, A_k + 2 \sum_{n=1,3\,\ldots}^{\infty} \frac{1}{(n-1)!} \sum_{k_2 \ldots k_n} V_n(-k, k_2 \ldots k_n)\, A_{k_2} \ldots A_{k_n} \end{gathered} \tag{3.3}$$

получаем уравнение для функции Грина (3.1):

$$\left\{\left(i\frac{d}{dt}\right)^2 - \omega_k^2\right\} G_{kk'}(t-t') = \delta(t-t')\, 2\,\omega_k\, \delta_{k,k'} + \\ + 2\omega_k \sum_{n=2}^{\infty} \frac{1}{n!} \sum_{k_1\ldots k_n} V_{n+1}(-k, k_1\ldots k_n) \langle\langle A_{k_1}\ldots A_{k_n}; A^+_{k'}(t')\rangle\rangle \tag{3.4}$$

(Здесь и далее в случае решетки без примесей, когда имеет место закон сохранения импульса, $-k = \{-\mathbf{k}, j\}$, $\delta_{k,k'} = \Delta(\mathbf{k}-\mathbf{k}')\,\delta_{j,j'}$.

Многофононную функцию Грина в (3.4) представим в виде разложения по функциям Грина низшего порядка, как и в (2.4)

$$\sum_{k_1\ldots k_n} V_{n+1}(-k, k_1\ldots k_n)\langle\langle A_{k_1}\ldots A_{k_n}; A^+_{k'}(t')\rangle\rangle \approx \\ \approx \sum_{k_1\ldots k_n} V_{n+1}(-k, k_1, k_2\ldots k_n)\langle A_{k_2}\ldots A_{k_n}\rangle\, n\, \langle\langle A_{k_1}; A^+_{k'}(t')\rangle\rangle + \\ + \sum_{k_1\ldots k_n} V_{n+1}(-k, k_1,\ldots k_n)\langle A_{k_3}\ldots A_{kn}\rangle \frac{n(n-1)}{2} \langle\langle A_{k_1} A_{k_2}; A^+_{k'}(t')\rangle\rangle, \tag{3.5}$$

где мы воспользовались симметрией функции $V_n(k_1\ldots k_n)$ по перестановке индексов k_i. Подставляя это разложение и переходя к фурье-представлению функций Грина согласно (2.7), получим

$$(\omega^2 - \omega_k^2)\, G_{kk'}(\omega) = 2\,\omega_k\,\delta_{k,k'} + \\ + 2\omega_k \sum_p P(-k, p)\, G_{pk'}(\omega) + \omega_k \sum_{pp'} Q(-k, p, p')\, G^{(1)}_{pp',k'}(\omega) \tag{3.6}$$

где мы ввели функции

$$P(-k, p) = \sum_{n=1}^{\infty} \frac{1}{n!} \sum_{k_1\ldots k_n} V_{n+2}(-k, p, k_1\ldots k_n)\langle A_{k_1}\ldots A_{k_n}\rangle, \tag{3.7}$$

$$Q(-k, p, p') = \sum_{n=0}^{\infty} \frac{1}{n!} \sum_{k_1\ldots k_n} V_{n+3}(-k, p, p', k_1\ldots k_n)\langle A_{k_1}\ldots A_{k_n}\rangle. \tag{3.8}$$

Составим уравнение для двухфононной функции Грина

$$G^{(1)}_{pp'k'}(t-t') = \langle\langle A_p(t)\, A_{p'}(t); A^+_{k'}(t')\rangle\rangle \tag{3.9а}$$

При этом нам потребуются также следующие функции Грина:

$$G^{(2)}_{pp',k'}(t-t') = \langle\langle B_p(t)\, A_{p'}(t); A^+_{k'}(t')\rangle\rangle, \tag{3.9б}$$

$$G^{(3)}_{pp',k'}(t-t') = \langle\langle B_p(t)\, B_{p'}(t); A^+_{k'}(t')\rangle\rangle. \tag{3.9в}$$

В правых частях уравнений для этих функций Грина, подобно уравнению (3.4), появляются снова многофононные функции Грина. Для них мы примем простейшее приближение:

$$\sum_{k_1\dots k_n} V_{n+1}(-p, k_1\dots k_n)\,\langle\langle C_{p'} A_{k_1}\dots A_{k_n}; A^+_{k'}(t')\rangle\rangle \approx$$
$$\approx \sum_{k_1\dots k_n} V_{n+1}(-p, k_1\dots k_n)\,\langle C_{p'} A_{k_2}\dots A_{k_n}\rangle\, n\,\langle\langle A_{k_1}; A^+_{k'}(t')\rangle\rangle, \tag{3.10}$$

где $C_{p'}$ — оператор $A_{p'}$, или $B_{p'}$,. Пользуясь также приближением:

$$\langle C_{p'} A_{k_2}\dots A_{k_n}\rangle = \sum_{i=2}^{n} \langle C_{p'} A_{ki}\rangle \left\langle \prod_{j\neq i}^{n} A_{kj}\right\rangle \delta_{-p',ki}\,, \tag{3.11}$$

после несложных преобразований получаем следующую систему уравнений для фурье-обрьазов функций Грина (3.9):

$$\omega G^{(1)}_{pp',k'}(\omega) = \omega_p\, G^{(2)}_{pp',k'}(\omega) + \omega_{p'}\, G^{(2)}_{p'p,k'}(\omega)\,, \tag{3.12а}$$

$$\begin{aligned}\omega G^{(2)}_{pp',k'}(\omega) &= \omega_p\, G^{(1)}_{pp',k'}(\omega) + \omega_{p'}\, G^{(3)}_{pp',k'}(\omega) + \\ &+ 2\,\langle A^+_{p'} A_{p'}\rangle \sum_{p_1} Q(-p, -p', p_1)\, G_{p_1 k'}(\omega)\,,\end{aligned} \tag{3.12б}$$

$$\begin{aligned}\omega G^{(3)}_{pp',k'}(\omega) &= \omega_p\, G^{(2)}_{p'p,k'}(\omega) + \omega_{p'}\, G^{(2)}_{pp',k'}(\omega) + \\ &+ 2\,(\langle A^+_{p'} B_{p'}\rangle + \langle B_p A^+_p\rangle) \sum_{p_1} Q(-p, -p', p_1)\, G_{p_1 k'}(\omega)\,.\end{aligned} \tag{3.12в}$$

где $Q(-p, -p', p_1)$- функция (3.8). Решая эту систему уравнений, находим функцию Грина (3.9а):

$$G^{(1)}_{vp',k'}(\omega) = 2F(p, p', \omega) \sum_{p_1} Q(-p, -p', p_1)\, G_{p_1 k'}(\omega) \tag{3.13}$$

где (ср. [6]):

$$\begin{aligned}F(p, p', \omega) &= \frac{N_p + N_{p'}}{2}\, \frac{\omega_p + \omega_{p'}}{\omega^2 - (\omega_p + \omega_{p'})^2} - \frac{N_p - N_{p'}}{2}\, \frac{\omega_p - \omega_{p'}}{\omega^2 - (\omega_p - \omega_{p'})^2} + \\ &+ \frac{\widetilde{N}_p + \widetilde{N}_{p'} + 2}{2} \left\{ \frac{\omega}{\omega^2 - (\omega_p + \omega_{p'})^2} - \frac{\omega}{\omega^2 - (\omega_p - \omega_{p'})^2} \right\},\end{aligned} \tag{3.14}$$

где

$$N_p = \langle A^+_p A_p\rangle\,, \quad \widetilde{N}_p = \langle A^+_p B_p\rangle\,.$$

Заметим, что в низшем порядке функции N_p и $\widetilde{N}_p$ равны:

$$N_p \approx \langle A^+_p A_p\rangle_0 = 2\,\langle a^+_p a_p\rangle_0 + 1\,,\ \widetilde{N}_p \approx \langle A^+_p B_p\rangle_0 = -1\,,$$

так, что второй член в (3.14) обращается в нуль.

Окончательно, подставляя (3.13) в уравнение (3.6) для функции Грина (3.1) получаем следующее выражение:

$$G_{kk}(\omega) = \frac{2\,\omega_k}{\omega^2 - \omega_k^2 - 2\,\omega_k\, M_k(\omega)}\,, \tag{3.15}$$

где массовый оператор $M_k(\omega)$ равен

$$M_k(\omega) = \mathbf{P}(-\,\boldsymbol{k}, \boldsymbol{k}) + \sum_{pp'} |Q(-\,\boldsymbol{k}, \boldsymbol{p}, \boldsymbol{p}')|^2\, F(\boldsymbol{p}, \boldsymbol{p}', \omega)\,. \tag{3.16}$$

Полюса функции Грина (3.15) определяют спектр частот колебаний решётки ϵ_k:

$$\epsilon_k^2 = \omega_k^2 + 2\,\omega_k\, \mathrm{Re}\, M_k(\epsilon_k) \tag{3.17}$$

с затуханием — обратным временем жизни:

$$\begin{aligned} \Gamma_k(\epsilon_k) &= -\,\frac{\omega_k}{\epsilon_k}\, Im\, M_k(\epsilon_k + i\varepsilon) = \\ &= -\,\frac{\omega_k}{\epsilon_k} \sum_{pp'} |Q(-\,\boldsymbol{k}, \boldsymbol{p}, \boldsymbol{p}')|^2\, Im\, F(\boldsymbol{p}, \boldsymbol{p}', \epsilon_k + i\varepsilon)\,. \end{aligned} \tag{3.18}$$

Сразу же замечаем, что первый член массового оператора — функция $\boldsymbol{P}(-\boldsymbol{k},\boldsymbol{k})$ — определяет спектр частот в псевдогармоническом приближении, как и уравнение (2.8):

$$\begin{aligned} (\epsilon_k^{\circ})^2 &= \omega_k^2 + 2\omega_k\, \boldsymbol{P}(-\,\boldsymbol{k}, \boldsymbol{k}) = \\ &= \sum_{lm} \Phi_{lm}^{\alpha\beta} \frac{1}{(M_l\, M_m)^{1/2}}\, e_{-k}^{\alpha}(\boldsymbol{l})\, e_k^{\beta}(\boldsymbol{m}) + \\ &+ \sum_{lm} \frac{1}{(M_l\, M_m)^{1/2}}\, e_{-k}^{\alpha}(\boldsymbol{l})\, e_k^{\beta}(\boldsymbol{m}) \sum_{n=1}^{\infty} \frac{1}{n!} \sum_{l_1 \ldots l_n} \Phi_{lml_1\ldots l_n}^{\alpha\beta\alpha_1\ldots\alpha_n} \left\langle \prod_{i=1}^{n} u_{l_i}^{\alpha_i} \right\rangle, \end{aligned} \tag{3.19}$$

где мы воспользовались определениями (1.5), (1.7), (1.12). Помимо этого возникает также перенормировка частоты за счет возможных процессов распада возбуждений, которые описываются второй частью массового оператора (3.16) и которая приводит также к конечному времени жизни — затуханию (3.18). Очевидно, если бы мы учли следующие члены в разложении (3.5), то возникло бы дополнительное смещение частоты фононов и затухание. Однако, учёт процессов более высокого порядка — четырех, пяти фононных и т. д. — представляет существенные трудности и мы не будем их рассматривать.

Особенно простой вид выражение для массового оператора принимает в случае парных сил (1.15), когда функции (3.7), (3.8) имеют вид:

$$P(-k,p) = -\frac{1}{2}\sum_{lm}\sum_{q}\Phi_{lm}(q)\, e^{i\mathbf{q}(\mathbf{l}-\mathbf{m})}\, q^{\alpha} q^{\beta}\, b^{\alpha}_{lm}(-k)\, b^{\beta}_{lm}(p) \times \times \left\{\langle e^{i\mathbf{q}(\mathbf{u}_l-\mathbf{u}_m)}\rangle - 1\right\}, \tag{3.7a}$$

$$Q(-k,p,p') = -\frac{i}{2}\sum_{lm}\sum_{q}\Phi_{lm}(q)\, e^{i\mathbf{q}(\mathbf{l}-\mathbf{m})}\, q^{\alpha} q^{\beta} q^{\gamma}\, b^{\alpha}_{lm}(-k)\, b^{\beta}_{lm}(p) \times \times\, b^{\gamma}_{lm}(p')\, \langle e^{i\mathbf{q}(\mathbf{u}_l-\mathbf{u}_m)}\rangle . \tag{3.8a}$$

Если для вычисления средних в (3.7а) (3.8а) воспользоваться псевдогармоническим приближением, то согласно (2.9) функция (3.7а) будет определяться только четными ангармонизмами, а функция (3.8а) — нечетными. Таким образом, в псевдогармоническом приближении массовый оператор (3.16) определяется всеми четными ангармонизмами, учтенными в первом порядке теории возмущений, и всеми нечетными ангармонизмами, учтенными во втором порядке теории возмущений в методе двухвременных функций Грина (см. [5], § 29). Результаты работы [6] получаются при учёте первых членов в разложении (3.7), (3.8), т. е. при учёте только четвертого и кубического членов в ангармонической части гамильтониана (1.11).

Для определения равновесного положения атомов следует воспользоваться усредненным уравнением движения для операторов (3.3):

$$i\frac{d}{dt}\langle B_k(t)\rangle = 2\sum_{n=1,2\ldots}^{\infty}\frac{1}{(n-1)!}\sum_{k_2\ldots k_n} V_n(-k,k_2\ldots k_n)\langle A_{k_2}\ldots A_{k_n}\rangle = 0\,. \tag{3.20}$$

При этом для решетки без примесей выполняется закон сохранения квазиимпульса: $\mathbf{k}=0$, $\mathbf{k}_2+\ldots+\mathbf{k}_n=0$.

Вычисляя корреляционные функции в псевдогармоническом приближении, т. е. полагая

$$\langle A_{k_1}\ldots A_{k_n}\rangle = \delta_{n,2s}\prod_{i=1}^{s}(2\,\mathrm{s}-1)!!\,\langle A^{+}_{k_i} A_{k_i}\rangle, \tag{3.21}$$

где $(2s-1)!!$ — произведение нечетных чисел от 1 до $(2s-1)$, и пользуясь спектральной теоремой:

$$\langle A^{+}_{k} A_{k}\rangle = \frac{1}{2\pi}\int_{-\infty}^{\infty}\frac{[-2\, Im\, G_{kk}(\omega+i\varepsilon)]}{e^{\omega/\Theta}-1}\, d\omega\ , \tag{3.22}$$

приходим к самосогласованной системе уравнений (3.15), (3.16), (3.7), (3.8), (3.20), определяющей свойства ангармонического кристалла при учёте высших порядков ангармонизма. При этом, в отличие от псевдогармонического приближения (2.8), перенормированные возбуждения (317) обладают конечным затуханием (3.13), что позволяет установить пределы применимости псевдогармонического приближения.

4. Обсуждение

Полученные нами уравнения позволяют рассмотреть свойства ангармонического кристалла при учёте высших порядков ангармонизма. Псевдогармоническое приближение (2.8) определяет спектр частот колебаний решетки при учёте всех чётных порядков ангармонизма. Отметим, что это уравнение, записанное в координатном представлении, особенно удобно при исследовании спектра колебаний решеток с примесями (см. [7]). Как видно из определений (2.6) и (2.6а), учет ангармонизмов высоких порядков может приводить к существенному изменению «псевдогармонических» силовых постоянных для легкой слабосвязанной примеси, поскольку её среднеквадратичное смещение $\langle u_1^2 \rangle$ может значительно превышать среднеквадратичные смещения $\langle u_0^2 \rangle$ атомов основной решетки.

Учет простейшего процесса распада одного фонона на два в самосогласованном поле остальных фононов (расцепление (3.5)) позволяет найти дополнительную перенормировку частоты за счет ангармонизмов нечетных порядков, а также определить затухание этих возбуждений (3.18). В области фазовых переходов это затухание может оказаться весьма большим и концепция слабосвязанных псевдогармонических фононов может оказаться неверной. Сама область фазовых переходов может быть найдена из самосогласованного решения уравнений, определяющих спектр колебаний и корреляционные средние, когда колебания решетки становятся неустойчивыми.

Заметим, наконец, что полученные нами уравнения в разделах 2 и 3 позволяют предложить некоторый эффективный ангармонический гамильтониан, содержащий лишь ангармонизм третьего порядка, где силовые постоянные определяются через корреляционные функции самосогласованным образом. Действительно, применим приближение типа (2.4) к гамильтониану (1.1), (1.15):

$$\begin{aligned}\sum_{n=1}^{\infty} \frac{1}{n!} \{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)\}^n \simeq \sum_{n=2}^{\infty} \frac{1}{n!} \{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)\}^2 \frac{n!}{(n-2)!2!} \langle \{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)\}^{n-2} \rangle + \\ + \sum_{n=3}^{\infty} \frac{1}{n!} \{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)\}^3 \frac{n!}{(n-3)!3!} \langle \{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)\}^{n-3} \rangle = \qquad (4.1) \\ = \left(\frac{1}{2} \{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)\}^2 + \frac{1}{3!} \{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)\}^3 \right) \langle e^{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)} \rangle .\end{aligned}$$

В результате получаем следующий эффективный ангармонический гамильтониан:

$$\begin{aligned}\mathscr{H}_{\text{эф.}} = \sum_l \frac{P_l^2}{2M_l} + \frac{1}{2} \sum_{lm} \tilde{\Phi}_{lm}^{\alpha\beta} u_l^\alpha u_m^\beta + \\ + \frac{1}{2} \sum_{l \neq m} \sum_q \Phi_{lm}(q) e^{i\mathbf{q}(\mathbf{l}-\mathbf{m})} \frac{1}{3!} \{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)\}^3 \langle e^{i\mathbf{q}(\mathbf{u}_l - \mathbf{u}_m)} \rangle,\end{aligned} \qquad (4.2)$$

где $\tilde{\Phi}^{\alpha\beta}_{lm}$ определяется соотношением (2.6). Обобщение на случай непарных сил (1.3) производится тривиально, однако гамильтониан будет иметь громоздкий вид. С помощью этого гамильтониана легко получаются уравнения для функций Грина (2.1) и (3.1) найденные в разделах 2 и 3. Добавление эффективного кубичного ангармонического члена в (4.2) позволяет учесть ангармонизмы нечетных порядков в псевдогармонической теории, определяемой первыми двумя членами в (4.2). Отметим, что в гамильтониане (4.2), а также в расцеплениях (2.4), (3.5) по существу просуммированы диаграммы типа:

и поэтому при построении диаграмной техники на основе гамильтонина (4.2) ти диаграммы не следует учитывать.

В заключение нам хотелось бы поблагодарить С. В. Тябликова за предложение темы и обсуждения, а также Г. Конвента, который обратил наше внимание на работы [2].

Литература

1. R. A. Cowley, Adv. in Phys., **12,** 421, 1963.
2. D. J. Hooton, Phil. Mag., **46,** 422, 1955; Z. Phys., **142,** 42, 1955.
   T. R. Koehler, Phys. Rev. Lett., **17,** 89, 1966.
   M. H. Cohen, R. M. Martin, preprint: "The Pseudoharmonic Theory of Lattice Vibrations", 1966.
3. L. J. Sham, Phys. Rev., **139,** A1189, 1965.
4. Д. Н. Зубарев, УФН, **71,** 71, 1960.
5. С. В. Тябликов, Методы квантовой теории магнетизма, Наука, М. 1965.
6. K. N. Pathak, Phys. Rev., **139,** A1569, 1965.
7. A. A. Maradudin, Rep. Prog. Phys. **28,** 331, 1965.

ACCOUNT OF THE HIGHEST ORDER ANHARMONIC EFFECTS IN CRYSTALS

By

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Abstract

A method of accounting the highest order anharmonic terms in the crystal potential energy is developed. On the basis of the equation for Green functions a frequency spectrum of the lattice vibrations in pseudoharmonic approximation is found. The damping of the lattice vibrations is also determined.

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DELOCALIZATION AND CORRELATION OF ELECTRONS IN THE MOLECULES OF SATURATED COMPOUNDS. I.

CONSIDERATION OF THE DELOCALIZATION BY THE MO AND GEMINAL METHOD

By

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In the geminal method the delocalization energy is not usually considered whereas the major portion of the correlation energy may be taken into account.

In the MO theory only delocalization energy is considered. The estimates show that, at least in hydrocarbons, the correlation energy is more important than the delocalization one. Therefore the localized geminals can be energetically more advantageous than the delocalized molecular orbitals. To take into account delocalization in the geminal product scheme the AO of the neighbouring bonds are added to the localized geminals.

1. Introduction

It is well-known that the HF (Hartree—Fock) scheme is based on the molecular orbital description even though we are concerned with large saturated molecules with strongly localized bonds. On the other hand we have the localized geminal method [1—3] in which the wave function is taken in the following form:

$$\Phi = \mathscr{A}\{\varphi_A(12)\,\varphi_B(34)\ldots\}, \tag{1}$$

where the functions $\varphi_k(12)$ describe the valence bond electrons or lone pairs. The question, which we would like to pose is the following: taking into account the approximate character of both methods, which of these methods can be applied more successfully to the large saturated molecules with a strongly specified individuality of the bonds (for example, to hydrocarbons). For an answer to this it is necessary to define the criterion for comparing both methods. A similar idea was expressed some years ago by Levinson [4] who defined the energy criterion for comparing the MO method with the localized valence bond method.

In most saturated compounds the deviations from the additivity principle are small which indicates a relatively small role for the delocalization and a

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strongly specified individuality for the valence bonds. The latter fact creates the difficulties for a justification of the applicability of the delocalized MO to the molecules of saturated compounds [5], at least in those cases when one can talk about the relatively small effects leading to the deviation from the additivity principle.

Although the semiempirical treatment of the delocalization energy in the MO—LCAO theory gives a good agreement with experimental data [6—9], it is not yet possible to consider it as confirmation of the applicability of delocalized MO. It is enough to indicate that the semiempirical estimate of the inductive effect only without delocalization energy also gives a good result [10].

Let us note that the LENNARD-JONES "equivalent orbital" theory [11, 12] is really only a formal method of associating certain orbitals with certain bonds in saturated molecules. It does not in itself give any direct method for finding just how localized these orbitals are or how they differ from one molecule to another [13].

On the other hand, the localized geminal may be associated with individual valence bonds and the correlation between valence electrons may be considered, which is important for the heats of formation [14, 15]. Moreover, as was shown by one of the authors [16], in the geminal product scheme it is possible to take into account the inductive effect and the transferability of geminals from one molecule to another.

To take into account delocalization in the geminal method the AO of the neighbouring bonds are added to the localized geminals.

To answer the above-mentioned question it is necessary to analyse those approximations, which are common to both methods, from the point of view of their application to molecules of saturated compounds and to make some estimate of the delocalization and correlation energies.

2. Estimation of the delocalization energy in the MO theory

Let us introduce the following definitions.

a) MO is localized if one includes only AO (among them hybrid orbitals), concerned with a certain bond.

b) MO is delocalized if one includes AO of two or more bonds.

In the simpler case, when the valence bond is formed by two AO, the localized and delocalized MO is of the following type [8]:

$$\varphi_k = C_k \psi_k + C_{k+1} \psi_{k+1} \qquad (k = 1, 2, \ldots 2n-1), \tag{2}$$

$$\tilde{\varphi}_i = \sum_{k=1}^{2n}{}' \tilde{C}_{ik} \psi_k \qquad (k = 1, 2, \ldots 2n). \tag{3}$$

In the case of the localized MO the task is reduced to the solution of the "n"-secular equations of 2^{nd} order, and in the case of the delocalized, to the solution of the secular equation of the $2n^{\text{th}}$ order.

The delocalization energy may be considered as a small quantity if two conditions are fulfilled: *a)* resonance integrals of the interactions between AO of different bonds are small in comparison with such integrals between AO of the same bond; *b)* the differences between the energies of the ground states of the localized MO are smaller then their excitation states.

Let us estimate the delocalization energy semi-empirically assuming that the deviation from additivity principle is due to changes in delocalization energy. The secular equation for ethane (for example) has the following form, with some simplifying conditions, the meaning of which is clear from the determinant structure:

$$\left|\begin{array}{cccccc:cc:cccccc}
x & 1 & 0 & 0 & 0 & 0 & 0 & 0 & \multicolumn{6}{c}{} \\
1 & x & 0 & m & 0 & m & m & 0 & \multicolumn{6}{c}{} \\
0 & 0 & x & 1 & 0 & 0 & 0 & 0 & \multicolumn{6}{c}{0} \\
0 & m & 1 & x & 0 & m & m & 0 & \multicolumn{6}{c}{} \\
0 & 0 & 0 & 0 & x & 1 & 0 & 0 & \multicolumn{6}{c}{} \\
0 & m & 0 & m & 1 & x & m & m & \multicolumn{6}{c}{} \\
\hdashline
0 & m & 0 & m & 0 & m & x & \gamma & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & \gamma & x & m & 0 & m & 0 & m & 0 \\
\hdashline
\multicolumn{6}{c:}{} & 0 & m & x & 1 & m & 0 & m & 0 \\
\multicolumn{6}{c:}{} & 0 & 0 & 1 & x & 0 & 0 & 0 & 0 \\
\multicolumn{6}{c:}{} & 0 & m & 0 & m & x & 1 & m & 0 \\
\multicolumn{6}{c:}{0} & 0 & 0 & 0 & 0 & 1 & x & 0 & 0 \\
\multicolumn{6}{c:}{} & 0 & m & 0 & m & 0 & m & x & 1 \\
\multicolumn{6}{c:}{} & 0 & 0 & 0 & 0 & 0 & 0 & 1 & x
\end{array}\right| = 0\,, \tag{4}$$

where

$$x = \frac{\alpha = E}{\beta}\,, \tag{4'}$$

β is the resonance integral between AO of the same bond CH ($\beta_{\mathrm{C-H}}$),

$$\gamma = \frac{\beta_{\mathrm{C-C}}}{\beta_{\mathrm{C-H}}}$$

is a parameter characteristic of the difference between the resonance integrals of the C—C and C—H bonds.

$$m = \frac{\beta_{\mathrm{C}}}{\beta_{\mathrm{C-H}}}$$

a small parameter characteristic of the resonance integral of the interaction of two hybrid orbitals of the same carbon atom.

It is assumed that the Coulomb integrals of C and H atoms are equal to each other.

Parameters β, γ, m can be calculated with the help of empirical data for the heats of formation of the molecules [7]. If, for the energy of the system, we take the total energy of the occupied MO, then the expressions correct to m^2 for the heats of formation of the molecules take the following form:

$$\mathrm{CH}_4: \quad \mathscr{E} = 2\beta\left(4 + \frac{3}{2}m^2\right) = 392{,}86 \text{ ccal/mol}, \tag{5}$$

$$\mathrm{C}_2\mathrm{H}_6: \quad \mathscr{E} = 2\beta\left[(6+\gamma) + 3\left(\frac{1}{2} + \frac{1}{1+\gamma}\right)m^2\right] = 667{,}02 \text{ ccal/mol}, \tag{5$'$}$$

$$\mathrm{C}_3\mathrm{H}_8: \quad \mathscr{E} = 2\beta\left[(8+2\gamma) + \left(\frac{7}{4} + \frac{1}{4\gamma} + \frac{5}{1+\gamma}\right)m^2\right] = 943{,}61 \text{ ccal/mol}. \tag{5$''$}$$

The delocalization energy $E_{\mathrm{del.}}$ corresponds to terms proportional to m^2*. Calculated from expressions (5—5″), the values of the parameters and delocalization energies are equal to:

$$\beta = 44{,}77 \text{ ccal/mol}, \quad \gamma = 0{,}45, \quad m^2 = 0{,}284,$$

$$\mathscr{E}_{\mathrm{del.}}(\mathrm{CH}_4) = 0{,}06038 \text{ at. u.} = 37{,}889 \text{ ccal/mol}.$$

It is interesting to remark that the value of the delocalization energy for a methane molecule obtained in [13] is only ~5ccal/mol.

3. Computation of delocalization in the geminal product scheme

The geminal $\psi(12)$ can be considered localized if in its bilinear expansion in atomic (hybrid) orbitals only atomic orbitals connected to the same bond are computed. Delocalized geminal $\tilde{\psi}(12)$ must be built from AO of different bonds. However, strong delocalization is energetically disadvantageous because in this case the geminals can take into account only an insignificant part of the correlation energy. It is reasonable to use the limited delocalized geminals which are little different from the localized geminals

$$\begin{aligned}\tilde{\psi}_k(12) = &\sum_{i_1 i_2} (A^k_{i_1 i_2} + B^k_{i_1 i_2})\, i^k_1(1)\, i^k_2(2) + \sum_{m(\neq k)} \Big[\sum_{i_1 j_1} B^{km}_{i_1 j_1}(1 + I\hat{I}_{i_1 j_1})\, i^k_1(1)\, j^m_1(2) + \\ &+ \sum_{j_1 j_2} B^{mm}_{j_1 j_2}\, j^m_1(1)\, j^m_2(2) + \sum_{m'(k,m)} \sum_{j_1 j_2} B^{mm'}_{j_1 j_2}(1 + I\hat{I}_{j_1 j_2})\, j^m_1(1)\, j^{m'}_2(2)\,,\end{aligned} \tag{6}$$

---

* With these assumptions the conclusion about the proportionality of $\mathscr{E}_{\mathrm{del.}}$ to m^2 has a general character. The absence in the expression for $\mathscr{E}_{\mathrm{del.}}$ of the terms of "m"th order can be proved easily by reducing the secular equation to nth order for the occupied MO and using Vieta's theorem.

where i^k, j^m are AO of "k" and "m" bonds. "$A^k_{i_1 i_2}$" are coefficients of the bilinear expansion of the localized geminals [17]. "B" are small coefficients responsible for delocalization. $\Pi_{j_1 j_2}$ is permutation operator equal to zero if $j_1 = j_2$. The bilinear combination (6) describes the general case of the computation of nonfundamental structures by the valence bond method.

Analogously to [17], the energy of the delocalized geminals can be represented in the form:

$$\widetilde{E}_k = \frac{\widetilde{\mathscr{E}}_k}{\widetilde{N}_k}, \tag{7}$$

$$\widetilde{\mathscr{E}}_k = \int \widetilde{\psi}_k(12)\, \hat{\mathscr{E}}_k\, \widetilde{\psi}_k(12)\, d(1)\, d(2), \tag{8}$$

where $\hat{\mathscr{E}}_k$ is the model operator for a geminal.

Taking into account (6) integral (8) is decomposed into a bilinear combination which includes the matrix elements of type:

$$\begin{aligned} \mathscr{E}[{}^{k}_{i_1}{}_{i_2}{}^{k}_{i_3}{}_{i_4}] &= (1 + \hat{\Pi}_{i_1 i_2})(1 + \hat{\Pi}_{i_3 i_4}) \int i_1^k(1)\, i_2^k(2)\, \hat{\mathscr{E}}_k\, i_3^k(1)\, i_4^k(2) d(1)\, d(2) = \\ &= \varepsilon[{}^{k}_{i_1}{}_{i_2}{}^{k}_{i_3}{}_{i_4}] + \sum_{m(\neq k)} \widetilde{N}_m^{-1} \widetilde{T}_{km}[{}^{k}_{i_1}{}_{i_2}{}^{k}_{i_3}{}_{i_4}]\,. \end{aligned} \tag{9}$$

$\widetilde{T}_{km}[i_1^k i_2 i_3^k i_4]$ denotes the matrix element of the interaction operator of the "k" geminal with the "m" one.

$\mathscr{E}[i_1^k i_2 i_3^k i_4]$ is the matrix element of the operator $\hat{\mathscr{E}}_k$ considering the inner interaction in the geminal and its interaction with the ionic cores of the system.

Matrix elements $\mathscr{E}^{(\eta)}[i_1^k i_2 i_3^k i_4]$ can be characterized by some parameter "η" denoting how many pairs of AO with the same electronic index components in the integral (8) are connected to different bonds. For a matrix element $\mathscr{E}^{(0)}[i_1^k i_2 i_3^k i_4]$, "$\eta$" is equal to zero. In general, the greater the value of parameter "η" the smaller is the matrix element.

Application of the usual variation procedure gives the following system of equations for a definition of the small coefficients "B":

$$\sum_{i_1 i_2} \left(B^k_{i_1 i_2} \mathscr{D}^{(0)}[{}^{k}_{i_1}{}_{i_2}{}^{k}_{i_3}{}_{i_4}] + \mathscr{A}^k_{i_1 i_2}\, \delta \widetilde{\mathscr{D}}\, [{}^{k}_{i_1}{}_{i_2}{}^{k}_{i_3}{}_{i_4}]\right) + \sum_{m(\neq k)} \sum_{i_3 j_3} B^{km}_{i_3 j_3} \mathscr{D}^{(1)}[{}^{k}_{i_1}{}_{i_2}{}^{k}_{i_3}{}^{m}_{j_3}] = 0, \tag{10}$$

$$\begin{aligned} \sum_{i_1 i_2} \mathscr{A}^k_{i_1 i_2} \mathscr{D}^{(2)}[{}^{k}_{i_1}{}_{i_2}{}^{m}_{j_1}{}^{m'}_{j_2}] + \sum_{i_1 j_3} B^{km}_{i_1 j_3} \mathscr{D}^{(1)}[{}^{k}_{i_1}{}^{m}_{j_3}{}^{m'}_{j_1}{}^{m}_{j_2}] + \sum_{i_1 j_3} B^{km}_{i_1 j_3} \mathscr{D}^{(1)}]{}^{k}_{i_1}{}^{m'}_{j_3}{}^{m}_{j_1}{}^{m'}_{j_2}] + \\ + \sum_{j_3 j_4} B^{mm'}_{j_3 j_4} \mathscr{D}^{(1)}[{}^{m}_{j_1}{}^{m'}_{j_2}{}^{m}_{j_3}{}^{m'}_{j_4}] = 0, \end{aligned} \tag{10'}$$

$$\sum_{i_1 i_2} \mathscr{A}^k_{i_1 i_2} \mathscr{D}^{(1)}[{}^{k}_{i_1}{}_{i_2}{}^{k}_{i_3}{}^{m}_{j_3}] + \sum_{i_1 j_1} B^{km}_{i_1 j_1} \mathscr{D}^{(0)}[{}^{k}_{i_1}{}^{m}_{j_1}{}^{k}_{j_3}{}^{m}_{i_3}] = 0\,, \tag{10''}$$

where

$$\mathscr{D}^{(0)}\left[{}^{k\;\;k}_{i_1i_2i_3i_4}\right] = \mathscr{E}^{(0)}\left[{}^{k\;\;k}_{i_1i_2i_3i_4}\right] - E_k\, N\left[{}^{k\;\;k}_{i_1i_2i_3i_4}\right], \tag{11}$$

$$\delta\mathscr{D}\left[{}^{k\;\;k}_{i_1i_2i_3i_4}\right] = \sum_{m(\neq k)} \Delta T_{kl} + (E_k - \widetilde{E}_k)\, N^{(0)}\left[{}^{k\;\;k}_{i_1i_2i_3i_4}\right]. \tag{12}$$

$\sum\limits_{l(\neq k)} \Delta T_{kl}$ is the change in the interaction energy of "k" geminal with others because of their delocalization. The delocalization energy is equal to

$$\begin{aligned} E^k_{\text{del.}} = E_k - \widetilde{E}_k = -\frac{1}{N_k}\Bigg\{\sum_m \Bigg(2 \sum_{i_1i_2}\sum_{i_3i_4} \mathscr{A}^k_{i_1i_2}\, B^{km}_{i_3j_3}\mathscr{D}^{(1)}\left[{}^{k\;\;km}_{i_1i_2i_3j_3}\right] + \\ + \sum_{i_1j_1}\sum_{i_2j_2} B^{km}_{i_1j_1}\, B^{km}_{i_2j_2}\, \mathscr{D}^{(0)}\left[{}^{km\;km}_{i_1j_1i_2j_2}\right]. \end{aligned} \tag{13}$$

In the expressions (10) and (13) the terms of the same order are retained. It is assumed that the delocalization weakly affects the electronic structure and that therefore ΔT_{km} is small and matrix elements (12) can be considered of the same order as the delocalization energy (13). The coefficients $B^{km}_{i_1j_1}$ are small of the first order and the coefficients B^k_{i,i_2}, $B^{mm'}_{j_1j_2}$ and the delocalization energy $E_{\text{del.}}$ are small of the second order, respectively, relative to the parameter "η". Using some simplified suppositions [18] the energy of the molecule can be represented in the form

$$E = \sum_k \left(\frac{\varepsilon_k}{N_k} + \sum_{m(>k)} \frac{1}{N_k N_m}\left[4\,\mathscr{K}_{km} - 2\,\mathscr{L}(-k-k-m-m-)\right]\right), \tag{14}$$

$$\varepsilon_k = \int \psi_k(12)\ \hat{\varepsilon}_k\, \psi_k(12)\, d(1)\, d(2), \tag{15}$$

$$\hat{\varepsilon} = \hat{h}_1 + \hat{h}_2 + \frac{1}{z_{12}}, \tag{15'}$$

$$\mathscr{K}_{km} = \int [\psi(12)]^2 \frac{1}{z_{13}} [\psi_m(34)]^2\, d(1) \ldots d(4), \tag{16}$$

$$\mathscr{L}(-k-k-m-m-) = \int \hat{\mathscr{L}}_{km} \psi_k(12)\, \psi_k(23)\, \psi_m(34)\, \psi_m(41)\, d(1) \ldots d(4), \tag{17}$$

$$\begin{aligned} \hat{\mathscr{L}}_{km} = \sum_{p=1}^{4} \hat{h}_p + \sum_{p=1}^{3} \sum_{q(>p)}^{4} \frac{1}{z_{pq}} - \left(\frac{\varepsilon_k}{N_k} + \frac{\varepsilon_m}{N_m} - 4\,\frac{\mathscr{K}_{km}}{N_k N_m}\right) + \\ + \sum_{s(\neq k,m)} \left(\hat{T}_{ks}(12) + \hat{T}_{sm}(34)\right), \end{aligned} \tag{17'}$$

where $\hat{h}_p$ is the one-electron operator of kinetic energy and potential interaction, N_k is a normalization constant of the geminal. Replacing in expressions (15—17) localized geminals ψ_k (12) by delocalized (6) ones and keeping only the significant coefficients $A^k_{i_1i_2}$ and $B^{km}_{i_1j_1}$ we obtain the expression for the delocalization energy

$$
\begin{aligned}
E_{\mathrm{del.}} = E - \widetilde{E} = -2 \sum_{k} \sum_{m(\neq k)} \Bigg(& \sum_{i_1 i_2} \sum_{i_3 j_3} \mathscr{A}^{k}_{i_1 i_2} B^{km}_{i_3 j_3} W_{km}[i_1 i_2 i_3 j_3] + \\
& + \frac{1}{2} \sum_{i_1 j_1} \sum_{i_3 j_3} B^{km}_{i_1 j_1} B^{km}_{i_3 j_3} Q_{km}[i_1 j_1 i_3 j_3] \Bigg) ,
\end{aligned}
\tag{18}
$$

$$
\begin{aligned}
W_{km}[i_1 i_2 i_3 j_3] = \sqrt{2} \int \Bigg[(\hat{\varepsilon}_k - \varepsilon_k) + \sum_{m(\neq k)} \hat{T}_{km} \Bigg] & i^k_1(1)\, i^k_2(2)\, i^k_3(1)\, j^k_3(2)\, d(1)\, d(2) - \\
& - \int \hat{\mathscr{L}}_{km}\, i^k_1(1)\, i^k_2(2)\, i^k_3(2)\, j^k_3(3)\, \psi_m(34)\, \psi_m(41)\, d(1) \ldots d(4) ,
\end{aligned}
\tag{19}
$$

$$
\begin{aligned}
Q_{km}[i_1 j_1 i_3 j_3] = \int \Bigg[(\hat{\varepsilon}_k - \varepsilon_k) + \sum_{m(\neq k)} \hat{T}_{km} \Bigg] & i^k_1(1)\, j^k_1(2)\, i^k_3(1)\, j^k_3(2)\, d(1)\, d(2) - \\
& - \int \hat{\mathscr{L}}_{km}\, j^k_1(1)\, j^k_3(3)\, i^k_1(2)\, i^k_3(2)\, \psi_m(34)\, \psi_m(41)\, d(1) \ldots d(4) ,
\end{aligned}
\tag{20}
$$

$$
\hat{T}_{km} = \int [\psi_m(34)]^2\, d(3)\, d(4) \left(\frac{2}{z_{12}} + \frac{2}{z_{23}} - 4\, \mathscr{K}_{km} \right) .
\tag{21}
$$

In the expressions (19) and (20) terms of the same order relative to the parameter "η" are kept (for $W_{km}\,\eta = 1$, for $Q_{km}\,\eta = 0$). The use of the variation principle gives the following system for the determination of the coefficients $B^{km}_{i_1 j_1}$:

$$
\sum_{i_2 j_2} B^{km}_{i_2 j_2} Q_{km}[i_1 j_1 i_2 j_2] + \sum_{i_3 i_4} \mathscr{A}^{k}_{i_3 i_4} W_{km}[i_3 i_4 i_1 j_1] = 0 .
\tag{22}
$$

Further simplification can be achieved by converting to one-electron approximation

$$
\widetilde{\psi}_k(12) = k(1)\, k(2) + \sum_{m(\neq k)} \frac{B_{km^*}}{\sqrt{2}} [k(1) m^*(2) + m^*(1)\, k(2)],
\tag{23}
$$

where k (1) is the MO of the "k" bond and m^* (1) is the MO of the first excited state of the "m" bond. The MO excited state is necessarily included through the introduction of the Pauli exclusion principle. It should be noted that if in (23) we used the MO m (1) of the ground state, then W_{km} would be equal to zero. The expressions (18)—(20) and (22) in this case are simplified to

$$
B_{km^*} = - \frac{W_{km^*}}{Q_{km^*}} ,
\tag{24}
$$

$$
E_{\mathrm{del.}} = \sum_{k} \sum_{m(\neq k)} \frac{W^2_{km^*}}{Q_{km^*}} ,
\tag{25}
$$

$$
\begin{aligned}
W_{km^*} = \sqrt{2} \big[& (\langle h/km^* \rangle - \langle h/kk \rangle \langle km^* \rangle) + (\langle kk/km^* \rangle - \\
& - \langle kk/kk \rangle \langle km^* \rangle) + 2 \sum_{l(\neq k)} (\langle km^*/ll \rangle - \langle kk/ll \rangle \langle km^* \rangle) -
\end{aligned}
\tag{26}
$$

$$-(\langle h/mm^*\rangle + \langle kk/mm^*\rangle + \langle mm/mm^*\rangle + \\ + 2\sum_{l(\neq k,m)} \langle mm^*/ll\rangle)\langle km\rangle - \langle km/mm^*\rangle],$$

$$Q_{km^*} = (\langle h/m^*m^*\rangle - \langle h/kk\rangle) + (\langle kk/m^*m^*\rangle - \langle kk/kk\rangle) + \\ + 2\sum_{l(\neq k)} (\langle m^*m^*/ll\rangle - \langle kk/ll\rangle) - \langle mm^*/mm^*\rangle . \tag{27}$$

The estimate of delocalization energy is made for the methane molecule according to the results of paper [19]. The calculations give the following values:

$$Q_{km^*} = 0{,}6948\,, \quad W_{km^*} = 0{,}0560\,, \quad B_{km^*} = -0{,}0806\,,$$
$$E_{\mathrm{del.}} = 0{,}0541 \text{ at. u. } 33{,}9 \text{ ccal/mol}.$$

4. Conclusion

Taking into account the estimates made we can try to answer the above mentioned question. In this paper we restrict ourselves to the discussion of the results on the example of the methane molecule.

The total correlation energy of the methane molecule according to [20—21] is 0.34—0.36 atomic units. Undoubtedly, the correlation energy of electron pairs of the C—H bonds provides the greater part of this value. Thus, if the competitive approximations are considered as the localized geminals and delocalized MO's, then the first of them should be supposed energetically more favourable because the total correlation energy of the valence pair is proved to be more than the delocalization energy. Probably, it is possible to extend this conclusion to the other saturated molecules and, in the first place, to hydrocarbons. If, in fact the localized geminals describe the system better than delocalized MO's, then this raises a doubt about the physical substantiation of the application of MO's, for studying the effects leading to deviation from the additivity principle.* Let us note that the calculation of other physical properties, and in particular, of the nuclear spin—spin coupling constants, which can be a more sensitive measure of the delocalization of electron binding, is of interest. The other paper of this series will be devoted to this and also to the calculation of pair and intergeminal correlation.

In conclusion one of us (R.G.N.) wishes to thank Dr. E. KAPUY for helpful discussions during his stay at the Research Group for Theoretical Physics of the Hungarian Academy of Sciences, in Budapest.

* This conclusion does not extend to calculations of the ionisation potentials because MO well describes the motion of the ionic vacancy [18]. Therefore, the fact of the dependence of the ionization potential on the length of the molecule cannot be used, as is done in [13], as an argument in favour of the necessity of the delocalization computation in neutral molecules.

REFERENCES

1. A. C. Hurley, J. E. Lennard-Jones and I. A. Pople, Proc. Roy. Soc., **A220,** 444, 1953.
2. E. Kapuy, Acta Phys. Hung., **9,** 237, 1958.
   J. Chem. Phys., **44,** 956, 1966.
3. I. Ts. Ljast, Zh. Strukt. Khim. USSR, **5,** 92, 1964.
4. I. B. Levinson, Litov. Fiz. Sbornik USSR, r. 1 No 3—4, 309, 1961.
5. C. A. Coulson, "Valency", Clarendon Press, Oxford, 1952.
6. G. Klopman, Helvet. Chim. Acta, **45,** 711, 1962.
7. G. Klopman, Tetrahedron, **19,** 111, 1963.
8. K. Fukui et al., Bull. Chem. Soc. Japan, **34,** 442, 1961.
9. И. Ц. Ляст, Р. Г. Нусратуллин, Сборник «Химия нероорганических соединений.» т. 9 (в печати).
10. M. J. Dewar and H. N. Sohmeising, Tetrahedron, **11,** 96, 1960.
11. J. E. Lennard-Jones, Proc. Roy. Soc., **A198,** 1, 1949.
12. J. E. Lennard-Jones and G. G. Hall, Proc. Roy. Soc., **A205,** 357, 1951.
13. J. A. Pople, D. P. Santry, Mol. Phys., **7,** 269, 1963—64.
14. K. Pitzer, Adv. Chem. Phys., **2,** 59, 1959.
15. O. Sinanoglu, Adv. Chem. Phys., **6,** 315, 1964.
16. R. G. Nousratoullin, Acta Phys. Hung., **24,** 163, 1968.
17. I. Ts. Ljast, Zh. Strukt. Khim. USSR, **7,** 80, 1966.
18. I. Ts. Ljast, Zh. Strukt. Khim. USSR, **6,** 271, 1965.
19. I. Ts. Ljast, R. G. Nousratoullin and T. S. Jampolskaja, Zh. Strukt. Khim. USSR, (to be published).
20. E. Clementi, J. Chem. Phys. **39,** 487, 1963.
21. K. D. Karlson and P. N. Skarcke, J. Chem. Phys., **40,** 612, 1964.
22. H. G. Heiht, Theoret. Acta Chim., **1,** 133, 1963, ibid. 223, 1963.

ДЕЛОКАЛИЗАЦИЯ И КОРРЕЛЯЦИЯ ЭЛЕКТРОНОВ В МОЛЕКУЛАХ НАСЫЩЕННЫХ СОЕДИНЕНИЙ. I.

Учет делокализации методом МО и джеминалей

И. Т. ЛЯСТ и Р. Г. НУСРАТУЛЛИН

Резюме

В методе джеминалей энергия делокализации обычно не учитывается в то время как большая часть корреляционной энергии может быть учтена.

В методе МО рассматривается только энергия делокализации. Оценки показывают, что, по крайней мере, в углеводородах, энергия корреляции играет большую роль чем энергия делокализации. Поэтому локализованные джеминали могут быть энергетически более выгодными чем делокализованные МО.

Для учета делокализации в методе джеминалей атомные орбиты соседних связей обавлятся к локализованным джеминалям.

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BARRIER PENETRABILITIES AND REDUCED WIDTHS FOR α-DECAY IN THE MEDIUM HEAVY REGION

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Alpha-decay barrier penetrabilities and reduced widths were calculated using all experimental disintegration energies and half-lives available in the medium heavy region. Some problems arising in numerical work is discussed and an extrapolation formula for penetrability vs. energy is given. The agreement between the relative values of the "experimental" as well as theoretical reduced widths has been found to be reasonable.

The nuclear α-decay barrier penetrability factor as defined by Rasmussen [1] is

$$P = \exp\left[-2\int_{\Omega} \frac{\sqrt{2M}}{\hbar}\sqrt{V(r)+\frac{2Ze^2}{r}+\frac{\hbar^2}{2Mr^2}l(l+1)-E}\,dr\right], \tag{1}$$

where

$$V(r) = -1100\exp\left[-\frac{r_f-1{,}17\,A^{1/3}}{0{,}574}\right]\mathrm{MeV} \tag{2}$$

is the real part of the Igo potential [2] for α-nuclear interaction for $|V| \leq 10$ MeV. In Eqs. (1) and (2) the symbols have the following meanings:

E : total decay energy,
Z : atomic number of the daughter nucleus,
A : mass number of the daugther nucleus,
M : reduced mass of α-particle,
l : orbital angular momentum carried off by the α-particle,
r_f : distance in fermis,
$\hbar, e$: Planck constant over 2π and electronic charge, respectively.

The integration in Eq. (1) is to be extended over the real values of the integrand. The limits, that is the values of r where the integrand vanishes are sometimes called inner and outer turning points.

The total decay energy E can be obtained from the α-particle energy by adding to it nuclear recoil and screening correction (see e.g. [3]).

Some remarks on numerical work

In evaluating the WKBJ integral the following procedure was used: The domain of integration was divided into three parts. The first part (I) is between the inner turning point and the maximum of the integrand; the third (III) is an interval of length 2 fermis measured backwards from the outer turning point. The remaining part is the second one (II). The values of the integral over the different domains were calculated by using Simpson's rule with constant divisions 0.01 fermis in (I) and (III) as well as 0.2 fermis in (II). This procedure was suggested by the fact that the area under the portion (II) of the integrand is much larger than that under other portions and the integrand varies much slower far from the turning points than near them.

Another possibility would be to calculate the integral by a modified Simpson's rule [1] to take into account the $\sqrt{x}$ behaviour of the integrand at the ends. However, this modification may occasionally give rise to some irregularities in the convergency of the approximate sums if the number of divisions is small. In fact, by dividing the barrier region into 128 equal parts only, one cannot reach, in general, the accuracy of 1% indicated in [1]. An illustrative example can be found in Table I. From a physical point of view this fact has no significance — of course — because the experimental results and the WKBJ approximation itself are rather inaccurate, but explains — in part — the slight differences between the results for P obtained by Rassmussen [1], Sandulescu [47], Kohlmann [4] and us.

The calculations were carried out by an M-20 computer in Dubna, USSR. To check the results an independent programme was made and used on an ICT computer in Budapest.

It is to be noted that the value of P is very sensitive to variations in the input parameters and constants. For example, if measuring the % error, a given error in M would yield an error hundred times larger in P. For this reason we list the values of the constants used.

$$e = 4.80273 \times 10^{-10} \text{ esu},$$

$$\hbar = 1.05439 \times 10^{-27} \text{ erg sec}.$$

Quantum energy as well as mass conversion factors:

$$1 \text{ MeV} = 1.60209 \times 10^{-6} \text{ erg}.$$

$$1 \text{ atomic mass unit} = 1.65975 \times 10^{-24} \text{ g}.$$

The values of M were calculated by the formula

$$M = 4{,}0027 \frac{A}{A+4}. \tag{3}$$

Table I

The values of the WKBJ integral (I) as well as the penetration factor (P) at different numbers of divisions of the barrier region (n) calculated by the method described in [1] when comparing them to the values obtained from the present procedure (I_0 and P_0) for Gd^{148}, $E = 3.27$, $l = 0$

n	$\frac{\lvert I - I_0 \rvert}{I_0} \cdot 100\,\%$	$P \cdot 10^{30}$	$\frac{\lvert P - P_0 \rvert}{P_0} \cdot 100\%$
128	$5.76 \cdot 10^{-2}$	7.8144	3.94
256	$1.09 \cdot 10^{-2}$	7.5736	$7.36 \cdot 10^{-1}$
512	$1.53 \cdot 10^{-4}$	7.5176	$9.71 \cdot 10^{-2}$
1024	$1.65 \cdot 10^{-3}$	7.5266	$1.11 \cdot 10^{-1}$
2048	$4.98 \cdot 10^{-4}$	7.5158	$3.47 \cdot 10^{-2}$
		$P_0 = 7.5183 \cdot 10^{-30}$	

The number 4.0027 is the mass of the α-particle in atomic mass units. (The omission of the term 0.0027 in the denominator shifts the value of M a little closer to the "true value", where this expression in the quotation marks denotes the reduced mass calculated by using exact mass data instead of mass numbers).

The results of the calculations

Tables II, III, IV and V contain the results obtained for the penetrability P as well as reduced width δ^2. Owing to the fact that the majority of nuclides in the Tables have no definite spin assignment so far, all the penetrabilities were calculated for $l = 0$ case. In calculating δ^2 the following definition was used [1]

$$\delta^2 = \frac{h \ln 2}{P \cdot T_\alpha}\,, \tag{4}$$

where T_a is the alpha partial half-life.

Alpha particle energies, total and alpha partial half-lives or branching ratios are presented as given in the references. The values of the inner turning points as well as penetrabilities are given for four, while reduced widths for three figures irrespective of the accuracies of their input parameters. In the last column only original references are listed, although in some cases the data were taken from the compilation tables of ESKOLA [39]. If more than one reference is presented, it is always the first that contains information on the energy. When alpha particle energies were corrected for nuclear recoil and electron screening a rounding off was applied in every case to four significant figures. So, the data in the third column are exactly the input energies for the WKBJ integral. No errors were calculated for P and δ^2. If such an error is

Table II

Barrier penetration factors and reduced widths for even—even nuclei, $l = 0$. The asterisk(*) means estimated value

Nuclide	α-particle energy (MeV)	Total α-decay energy (MeV)	Total half-life	α-partial half-life or α/total decay ratio	Inner turning point (fermi)	Barrier penetration factor	Reduced width (MeV)	References
${}_{58}\mathrm{Ce}^{142}_{84}$	~ 1.5	1.562	$5.1 \cdot 10^{15}$y	$5.1 \cdot 10^{15}$y	8.420	$7.453 \cdot 10^{-49}$	$2.390 \cdot 10^{4}$	[42]
${}_{60}\mathrm{Nd}^{144}_{84}$	1.83±0.02	1.902	$(2.1 \pm 0.4) \cdot 10^{15}$y	$(2.1 \pm 0.4) \cdot 10^{15}$y	8.439	$9.464 \cdot 10^{-44}$	0.457	[23]
${}_{62}\mathrm{Sm}^{146}_{84}$	2.46±0.02	2.550	$(7.4 \pm 1.5) \cdot 10^{7}$y	$(7.4 \pm 1.5) \cdot 10^{7}$y	8.470	$8.854 \cdot 10^{-36}$	0.139	[40]
${}_{62}\mathrm{Sm}^{148}_{86}$	2.14±0.03	2.220	$(1.2 \pm 0.3) \cdot 10^{13}$y	$(1.2 \pm 0.3) \cdot 10^{13}$y	8.489	$3.050 \cdot 10^{-40}$	0.0248	[5]
${}_{64}\mathrm{Gd}^{148}_{84}$	3.18±0.01	3.290	>35y	(93±5.8)y	8.503	$1.119 \cdot 10^{-29}$	0.0873	[6], [7] [17]
${}_{64}\mathrm{Gd}^{150}_{86}$	2.73±0.01	2.826	$(2.1 \pm 0.3) \cdot 10^{6}$y	$(2.1 \pm 0.3) \cdot 10^{6}$y	8.517	$4.048 \cdot 10^{-34}$	0.107	[6]
${}_{64}\mathrm{Gd}^{152}_{88}$	2.14±0.03	2.220	$(1.08 \pm 0.08) \cdot 10^{14}$y	$(1.08 \pm 0.08) \cdot 10^{14}$y	8.527	$5.593 \cdot 10^{-42}$	0.150	[43]
${}_{66}\mathrm{Dy}^{150}_{88}$	4.23±0.02	4.369	(7.20±0.10) min	0.18±0.02	8.548	$2.342 \cdot 10^{-23}$	0.0510	[9]
${}_{66}\mathrm{Dy}^{152}_{86}$	3.627±0.008	3.748	(2.37±0.02) h	(0.50±0.2)y	8.556	$2.519 \cdot 10^{-27}$	0.0721	[11]
${}_{66}\mathrm{Dy}^{154}_{88}$	2.870±0.020	2.969	>10y	$(2.9 \pm 1.5) 10^{6}$y	8.560	$4.700 \cdot 10^{-34}$	0.0666	[11], [14]
${}_{63}\mathrm{Er}^{152}_{84}$	4.80±0.02	4.953	(10.7±0.5) sec	$0.9^{\pm 0.05}_{-0.20}$	8.575	$2.396 \cdot 10^{-21}$	0.101	[12]
${}_{70}\mathrm{Yb}^{154}_{84}$	5.33±0.02	5.497	(0.39±0.04) sec	0.98*	8.601	$7.369 \cdot 10^{-20}$	0.0978	[15]
${}_{72}\mathrm{Hf}^{158}_{86}$	5.27±0.02	5.433	(3±0.5) sec	0.80*	8.635	$4.619 \cdot 10^{-21}$	0.165	[34]
${}_{72}\mathrm{Hf}^{174}_{102}$	2.50±0.03	2.584	$(2.0 \pm 0.4) \cdot 10^{15}$y	$(2.0 \pm 0.4) \cdot 10^{15}$y	8.770	$4.119 \cdot 10^{-43}$	0.110	[43]

$_{78}Pt_{96}^{174}$	6.03 ± 0.01	6.201	(0.7 ± 0.2) sec	0.8*	8.823	$2.098 \cdot 10^{-20}$	0.156	[35]
$_{78}Pt_{98}^{176}$	5.74 ± 0.01	5.902	(6.0 ± 0.5) sec	0.014	8.841	$1.336 \cdot 10^{-21}$	0.00500	[35]
$_{78}Pt_{100}^{178}$	5.44 ± 0.01	5.594	(21.3 ± 1.5) sec	0.013	8.858	$6.094 \cdot 10^{-23}$	0.0287	[35]
$_{78}Pt_{102}^{180}$	5.14 ± 0.01	5.286	(50 ± 5) sec	0.003	8.875	$2.085 \cdot 10^{-24}$	0.0825	[35]
$_{78}Pt_{104}^{182}$	4.82 ± 0.03	4.957	(2.5 ± 0.5) min	$2.3 \cdot 10^{-4} \cdot 2^{\pm 1}$	8.892	$3.901 \cdot 10^{-26}$	0.113	[13]
$_{78}Pt_{106}^{184}$	4.48 ± 0.02	4.608	(20 ± 2) min	$1.5 \cdot 10^{-5} \cdot 2^{\pm 1}$	8.907	$3.502 \cdot 10^{-28}$	0.102	[13]
$_{78}Pt_{108}^{186}$	4.23 ± 0.02	4.352	(2.0 ± 0.2) h	$1.4 \cdot 10^{-6} \cdot 2^{\pm 1}$	8.926	$7.776 \cdot 10^{-30}$	0.0717	[13]
$_{78}Pt_{110}^{188}$	3.93 ± 0.01	4.044	(10.2 ± 0.3) d	$(3.0 \pm 0.6) \cdot 10^{-7}$	8.943	$4.751 \cdot 10^{-32}$	0.0205	[13]
$_{78}Pt_{112}^{190}$	3.18 ± 0.02	3.277	$(5.4 \pm 0.6) \cdot 10^{11}$y	$(5.4 \pm 0.6) \cdot 10^{11}$y	8.946	$5.541 \cdot 10^{-39}$	0.0304	[13]
$_{78}Pt_{114}^{192}$	~ 2.6	2.684	$\sim 10^{15}$y	$\sim 10^{15}$y	8.954	$2.545 \cdot 10^{-46}$	357	[37]
$_{82}Pb_{122}^{204}$	~ 2.6	2.683	$1.4 \cdot 10^{17}$y	$1.4 \cdot 10^{17}$y	9.069	$2.032 \cdot 10^{-49}$	$3.19 \cdot 10^{3}$	[16]

Table III

Barrier penetration factors and reduced widths for even—odd nuclei $l = 0$. The asterisk(*) means estimated value

Nuclide	α-particle energy (MeV)	Total α-decay energy (MeV)	Total half-life	α-partial half-life or α/total decay ratio	Inner turning point (fermi)	Barrier penetration factor	Reduced width (MeV)	References
${}_{62}\mathrm{Sm}^{147}_{85}$	2.231 ± 0.010	2.314	$(1.08 \pm 0.02) \cdot 10^{11}$y	$(1.08 \pm 0.02) \cdot 10^{11}$y	8.477	$7.017 \cdot 10^{-39}$	0.120	[6], [19]
${}_{62}\mathrm{Sm}^{149}_{87}$	1.84 ± 0.05	1.911	$(4 \pm 2) \cdot 10^{14}$y	$(4 \pm 2) \cdot 10^{14}$y	8.494	$1.792 \cdot 10^{-45}$	126	[5]
${}_{64}\mathrm{Gd}^{149}_{85}$	3.016 ± 0.010	3.121	(8.3 ± 1.7) d	$(5.5 \pm 1.7) \cdot 10^{3}$y	8.512	$3.596 \cdot 10^{-31}$	0.0459	[20], [21]
${}_{64}\mathrm{Gd}^{151}_{87}$	2.60 ± 0.03	2.692	120 d	$4.2 \cdot 10^{7} \cdot 2^{\pm 1}$y	8.528	$1.310 \cdot 10^{-35}$	0.165	[21], [22]
${}_{66}\mathrm{Dy}^{151}_{85}$	4.074 ± 0.008	4.208	(17.7 ± 0.5) min	(5.4 ± 0.2) h	8.557	$2.751 \cdot 10^{-24}$	0.0536	[11], [24]
${}_{66}\mathrm{Dy}^{153}_{87}$	3.464 ± 0.008	3.580	(6.75 ± 0.15) h	$(3.0 \pm 0.3) \cdot 10^{-5}$	8.565	$1.431 \cdot 10^{-28}$	0.0247	[11], [9]
${}_{68}\mathrm{Er}^{153}_{87}$	4.67 ± 0.02	4.819	(36 ± 2) sec	$0.95^{+0.05}_{-0.20}$	8.586	$5.414 \cdot 10^{-22}$	0.140	[12]
${}_{70}\mathrm{Yb}^{155}_{85}$	5.21 ± 0.02	5.373	(1.65 ± 0.15) sec	0.90*	8.612	$2.235 \cdot 10^{-20}$	0.0700	[15]
${}_{72}\mathrm{Hf}^{157}_{85}$	5.68 ± 0.02	5.854	(0.12 ± 0.03) sec	1*	8.635	$2.661 \cdot 10^{-19}$	0.0898	[34]
${}_{78}\mathrm{Pt}^{175}_{97}$	5.95 ± 0.01	6.118	(2.1 ± 0.2) sec	0.8*	8.834	$1.020 \cdot 10^{-20}$	0.107	[35]
${}_{78}\mathrm{Pt}^{177}_{99}$	5.51 ± 0.01	5.666	(6.6 ± 1) sec	0.003	8.847	$1.253 \cdot 10^{-22}$	0.0104	[35]
${}_{78}\mathrm{Pt}^{179}_{101}$	5.15 ± 0.01	5.296	(33 ± 4) sec	0.001	8.862	$2.238 \cdot 10^{-24}$	0.0388	[35]
${}_{78}\mathrm{Pt}^{181}_{103}$	5.02 ± 0.02	5.162	(51 ± 5) sec	0.0006	8.885	$4.938 \cdot 10^{-25}$	0.0683	[35]
${}_{78}\mathrm{Pt}^{18}_{10}$	4.74 ± 0.03	4.875	(6.5 ± 1) min	$0.95 \cdot 2^{\pm 1}$y	8.902	$1.386 \cdot 10^{-26}$	0.00690	[13]

Table IV

Barrier penetration factors and reduced widths for odd—even nuclei $l = 0$. The asterisk(*) means estimated value

Nuclide	α-particle energy (MeV)	Total α-decay energy (MeV)	Total half-life	α-partial half-life or α/total decay ratio	Inner turning point (fermi)	Barrier penetration factor	Reduced width (MeV)	References
$_{61}\mathrm{Pm}^{145}_{84}$	2.24 ± 0.04	2.324	(17.7 ± 0.4) y	$(2.8 \pm 0.6) \cdot 10^{-9}$	8.458	$6.722 \cdot 10^{-38}$	0.214	[25]
$_{63}\mathrm{Eu}^{147}_{84}$	2.91 ± 0.01	3.013	(24 ± 2) d	$(2.2 \pm 0.6) \cdot 10^{-5}$	8.489	$1.683 \cdot 10^{-31}$	0.181	[6], [26]
$_{65}\mathrm{Tb}^{149}_{84}$	3.974 ± 0.006	4.106	(4.1 ± 0.05) h	(36 ± 7) h	8.535	$2.363 \cdot 10^{-24}$	0.00936	[27],[28], [29]
$_{65}\mathrm{Tb}^{149m}_{84}$	3.99 ± 0.03	4.122	(4.3 ± 0.2) min	$(2.5 \pm 0.5) \cdot 10^{-4}$	8.536	$2.969 \cdot 10^{-21}$	$9.36 \cdot 10^{-4}$	[9]
$_{65}\mathrm{Tb}^{151}_{86}$	3.409 ± 0.006	3.524	$\left(18.5 \begin{matrix} \pm 1.1 \\ 0.5 \end{matrix}\right)$ h	$(4.4 \pm 0.2) \cdot 10^{2}$ y	8.545	$2.224 \cdot 10^{-28}$	$9.28 \cdot 10^{-4}$	[27], [24]
$_{67}\mathrm{Ho}^{151}_{84}$	4.51 ± 0.02	4.656	(35.6 ± 0.4) sec	0.2 ± 0.05	8.561	$2.508 \cdot 10^{-22}$	0.0642	[36]
$_{67}\mathrm{Ho}^{151m}_{84}$	4.60 ± 0.02	4.748	(42 ± 4) sec	$0.28 \cdot 2^{\pm 1}$	8.565	$7.548 \cdot 10^{-22}$	0.0253	[36]
$_{67}\mathrm{Ho}^{153}_{86}$	3.92 ± 0.03	4.048	(9 ± 2) min	$(3 \pm 2) \cdot 10^{-3}$	8.570	$7.288 \cdot 10^{-26}$	0.219	[36]
$_{67}\mathrm{Ho}^{155}_{88}$	3.96 ± 0.03	4.088	(16.5 ± 0.5) min	0.0078	8.602	$1.463 \cdot 10^{-25}$	0.154	[38]
$_{69}\mathrm{Tm}^{152}_{84}$	5.11 ± 0.02	5.271	(1.58 ± 0.15) sec	0.90*	8.590	$2.308 \cdot 10^{-20}$	0.0707	[15]
$_{71}\mathrm{Lu}^{155}_{84}$	5.63 ± 0.02	5.804	(0.07 ± 0.02) sec	1*	8.165	$4.614 \cdot 10^{-19}$	0.0887	[34]
$_{83}\mathrm{Bi}^{199}_{116}$	5.47 ± 0.06	5.614	25 min	140 d	9.083	$5.913 \cdot 10^{-25}$	$4.01 \cdot 10^{-4}$	[18]

Table V

Barrier penetration factors and reduced widths for odd—odd nuclei $l = 0$. The asterisk(*) means estimated values

Nuclide	α-particle energy (MeV)	Total α-decay energy (MeV)	Total half-life	α-partial half-life or α/total decay ratio	Inner turning point (fermi)	Barrier penetration factor	Reduced width (MeV)	References
${}_{63}\mathrm{Eu}_{85}^{148}$	2.63 ± 0.03	2.724	(50 ± 11) d	$(1.6 \pm 0.5) \cdot 10^{7}$y	8.495	$1.677 \cdot 10^{-34}$	0.0339	[10]
${}_{65}\mathrm{Tb}_{85}^{150}$	3.492 ± 0.005	3.610	(3.15 ± 0.20) h	$\left(180 \begin{matrix} +300 \\ -120 \end{matrix}\right)$y	8.533	$9.788 \cdot 10^{-28}$	$5.16 \cdot 10^{-4}$	[31]
${}_{67}\mathrm{Ho}_{85}^{152}$	4.38 ± 0.02	4.522	(2.36 ± 0.16) min	$0.30 \cdot 2^{\pm 1}$	8.572	$4.970 \cdot 10^{-23}$	0.122	[36]
${}_{67}\mathrm{Ho}_{87}^{154}$	4.12 ± 0.03	4.253	(5.6 ± 0.2) min	0.017	8.592	$1.510 \cdot 10^{-24}$	0.0960	[38]
${}_{71}\mathrm{Lu}_{85}^{156}$	5.43 ± 0.03	5.598	~ 0.5 sec	0.7*	8.623	$6.936 \cdot 10^{-20}$	0.0579	[34]
${}_{83}\mathrm{Bi}_{115}^{198}$	5.83 ± 0.06	5.981	7 min	10 d	9.082	$3.092 \cdot 10^{-23}$	$1.07 \cdot 10^{-4}$	[18]

needed, one can easily estimate it by using the energy and half-life errors indicated, as well as formula (5).

Up till now only two nuclides have been found in the medium heavy region where the alpha spectra have fine structures. First GROMOV, MAHUNKA and FÉNYES [27] found an alpha line in the Tb fraction which was identified later as an alpha group from Tb^{149} to an excited state of Eu^{145} [31]. At the same time CHUMIN et al. [31] found another alpha group from Tb^{151} to an excited state of Eu^{147}. For explaining the disintegration of Tb^{149} and Tb^{145m} two decay schemes were proposed (not perfectly consistent with each other) by CHUMIN et al. [31] as well as MACFARLANE [41]. We have accepted and united the two schemes as can be seen in Fig. 1. For the ground state of Tb^{149} the $3/2^+$ assignment was retained. Beside considerations based on the shell model the following facts support the acceptance of this scheme. 1) For all the Eu isotopes of known ground state spin and parity a $5/2^+$ value was assigned [22]. These Eu isotopes, with the exception of Eu^{155}, have first excited states $7/2^+$. The energies of the first excited states of Eu^{151}, Eu^{149} and Eu^{147} are 22, 150 and 229 keV, respectively, while the excited state of Eu^{145} is 331 keV. For the same nuclei the second excited states $11/2^-$ have energies in the same sequence 197, 497 and 625 keV, so it is not probable that the excited state of the Eu^{145}, in question, has a spin value 11/2. Otherwise, if one had assigned a spin 11/2 (and negative parity) for this state the corresponding reduced width would prove to be too small. 2) For all the Tb isotopes of known ground state spin and parity a $3/2^+$ value was assigned. It is clear that the metastable level of the Tb^{149} must have a high spin, so the $11/2^-$ assignment seems to be obvious.

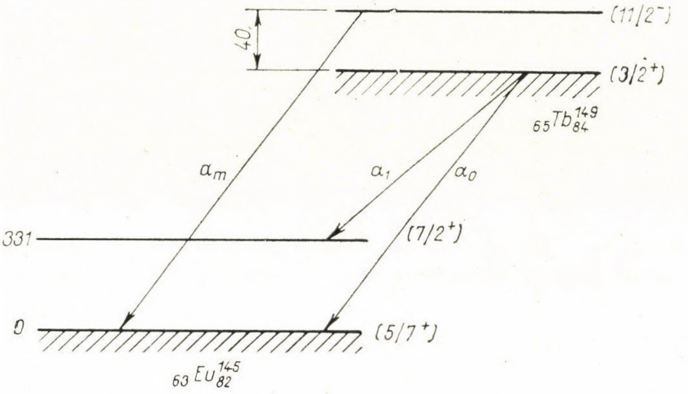

Fig. 1. Decay scheme for $Tb^{149,149m} \xrightarrow{\alpha} Eu^{145}$

It is to be noted that by inspecting Fig. 1 and the different transition probabilities one would expect that alpha-decay takes place from Tb^{149m} to the excited state of Eu^{145}, too, with an intensity about 10^{-3} relative to the ground state transition.

As for the disintegration of Tb^{151} we accepted the decay scheme to be found in [31] supposing a $3/2^+$ assignment for the ground state of Tb^{151} following the above considerations (see Fig. 2).

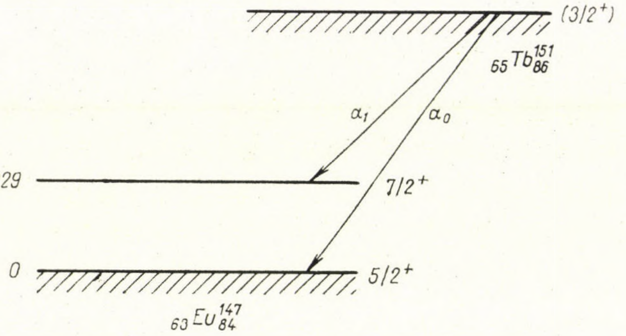

Fig. 2. Decay scheme for $Tb^{151} \xrightarrow{\alpha} Eu^{147}$

In Table VI the penetrability and reduced width values are presented for these Tb isotopes as well as for Pt^{178} with $l = 2$ [35]. In the seventh column the orbital angular momentum carried off by the alpha particle and the change in parity can be found.

Extrapolation for penetrability vs energy

It happens fairly frequently that a new value measured for E is believed to be more accurate than the previous ones and one desires to calculate quickly the new value of P for this energy. In such cases one can use the following approximate formula:

$$P(E) = P(E_0) \cdot \exp\left[\frac{0{,}988\sqrt{MZ}}{\varepsilon^{3/2}}(E - E_0)\right], \tag{5}$$

where $\varepsilon = \max\{E, E_0\}$ and the energies E_0, E are measured in MeV. In Table VII we presented some values of P calculated by Eq. (5) in order to have some indication concerning the accuracy of this extrapolation. One can infer that the error of this extrapolation practically does not exceed the value 1%, provided that $|E - E_0| \leq 30$ keV.

Table VI

Barrier penetration factors and reduced widths for some nuclei with $l \neq 0$. For the sake of the definiteness we used the values $3 \cdot 10^{-4}$(*) as well as $1 \cdot 10^{-3}$(**) when calculating the respective reduced widths

Nuclide	α-particle energy (MeV)	Total α-decay energy (MeV)	Total half-life	α-partial half-life α/total decay ratio	Relative intensities $I\alpha_1/I\alpha_0$	$\Delta\pi$, ΔI	Inner turning point (fermi)	Barrier penetration factor	Reduced width (MeV)
${}_{65}\mathrm{Tb}^{149}_{84}$ (α_0)	3.974 ± 0.006	4.106	(4.1 ± 0.05) h	(36 ± 7) h	$(2.5—4.0) \cdot 10^{-4}$*	2 no	8.519	$1.208 \cdot 10^{-24}$	0.0183
${}_{65}\mathrm{Tb}^{149}_{84}$ (α_1)	3.644 ± 0.005	3.767	(4.1 ± 0.05) h	—		2 no	8.507	$6.724 \cdot 10^{-27}$	$9.87 \cdot 10^{-4}$
${}_{65}\mathrm{Tb}^{149m}_{84}$ (α_m)	$3{,}99 \pm 0{,}03$	4,122	$(4{,}3 \pm 0{,}2)$ min	$(2{,}5 \pm 0{,}5) \cdot 10^{-4}$		3 yes	8,504	$7{,}792 \cdot 10^{-25}$	0,00357
${}_{65}\mathrm{Tb}^{151}_{86}$ (α_0)	3.409 ± 0.006	3.524	$\left(18.5 \begin{matrix} +1.1 \\ -0.5 \end{matrix}\right)$ h	$(4.4 \pm 0.2) \cdot 10^{2} y$	$(0.9—1.2) \cdot 10^{-3}$**	2 no	8.530	$1.126 \cdot 10^{-28}$	$1.83 \cdot 10^{-3}$
${}_{65}Tb^{151}_{86}$ (α_1)	3.183 ± 0.005	3.292	$\left(18.5 \begin{matrix} +1.1 \\ -0.5 \end{matrix}\right)$ h	—		2 no	8.522	$1.335 \cdot 10^{-30}$	$1.55 \cdot 10^{-4}$
${}_{78}\mathrm{Pt}^{178}_{100}$	5.28 ± 0.01	5.430	(21.3 ± 1.5) sec	0.0007		2 no	8.840	$5.542 \cdot 10^{-24}$	0.0170

Table VII

Some representative values of P extrapolated from E_0 to E_i by formula (5) · P(exact) denotes the value of P calculated by numerical methods described in the text

Nuclide	Energy E (MeV)	$\|E_0 - E\|$ keV	P(exact)	$P(E_i)$extrapolated from $P(E_0)$exact	Accuracy of extrapolation %
Nd^{144}	E_0 1.932		$3.354 \cdot 10^{-43}$		
	E_1 1,902	30	$9{,}464 \cdot 10^{-44}$	$9.487 \cdot 10^{-44}$	0,24
	E_2 1.872	60	$2.590 \cdot 10^{-44}$	$2.683 \cdot 10^{-44}$	3.59
Sm^{147}	E_0 2.303		$4.885 \cdot 10^{-39}$		
	E_1 2.293	10	$3.506 \cdot 10^{-39}$	$3.495 \cdot 10^{-39}$	0.31
	E_2 2.283	20	$2.511 \cdot 10^{-39}$	$2.501 \cdot 10^{-39}$	0.40
	E_3 2.273	30	$1.795 \cdot 10^{-39}$	$1.790 \cdot 10^{-39}$	0.28
	E_4 2.314	11	$7.017 \cdot 10^{-39}$	$7.041 \cdot 10^{-39}$	0.34
Pt^{184}	E_0 4.628		$4.683 \cdot 10^{-28}$		
	E_1 4.608	20	$3.502 \cdot 10^{-28}$	$3.474 \cdot 10^{-28}$	0.80
	E_2 4.588	40	$2.614 \cdot 10^{-28}$	$2.578 \cdot 10^{-28}$	1.38

Comparison between theory and experiment

The "experimental" reduced widths calculated in this paper were roughly compared with the theoretical widths of MANG [44] for even—even nuclides ($l = 0$). According to MANG the reduced widths can be expressed in the following form:

$$\delta^2 = \frac{1}{16} N_p (2 j_p + 3 - N_p) N_n (2 j_n + 3 - N_n) R, \tag{6}$$

where $N_p(N_n)$ is the number of protons (neutrons) in the last unfilled subshell of total angular momentum $j_p(j_n)$, while R is a rather complicated function of the quantum numbers n_i, l_i and the energy. If one would like to calculate the (relative) δ^2 values only for those nuclides which have the same unfilled proton and neutron subshell one can consider R to be constant. The reason for doing so is the fact that n_i, l_i are constants within a subshell while the energy dependence of R is very weak. Introducing the notation $2j_p + 1 = N_{pm}(2j_n + 1 = N_{nm})$ for the maximum number of protons (neutrons) that can be contained by the subshell we get

$$\delta^2 \approx C N_p (N_{pm} + 2 - N_p) N_n (N_{nm} + 2 - N_n), \tag{7}$$

where C is a constant.

In Fig. 3 the N—Z plane was divided into different "subshell-regions" according to the level sequence of the NILSSON scheme [45] at zero deformation.

Following formula (7) and the "experimental" reduced widths the values of C have been calculated. In each region we took the average of the C values and examined the deviations of the individual C's from this average.

In region I we used the data of Sm^{146}, Gd^{148}, Gd^{150} and Gd^{152} (the average of C's can be found in Fig. 3). The constancy of C is fulfilled within 30%. The value of C for Nd^{144} is considerably larger than the average but it is not impossibly high if taking into account the error of δ^2 for this nuclide. The value of C for Sm^{148} also deviates from this average but we feel that the accuracy of the decay energy in this case is rather questionable considering the fact that the decay energy dos not fit the different energy systematics [46].

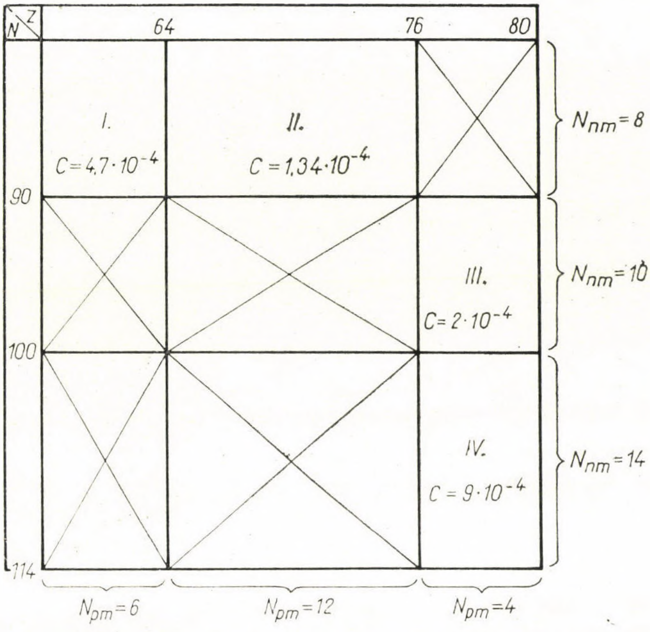

Fig 3. The average of the C values in the different "subshell-regions" (see the text)

In region II the constancy of C is surprisingly good. The average deviation is less than 10% and even the maximum of the deviations does not exceed 18%. (We used the data of Dy^{150}, Dy^{152}, Dy^{154}, Er ^{152}, Yb^{154} and Hf^{158}).

In regions III (Pt^{174}, Pt^{176}, Pt^{178}) and IV (Pt^{180}, Pt^{182}, Pt^{184}, Pt^{186}, Pt^{188}, Pt^{190}) the constancy of C is not fulfilled. The deviations are one half (III) and one (IV) order of magnitude, respectively. However, this is not astonishing because far from the magic numbers the shell model loses its validity. (Besides, there is some uncertainty in the branching ratios, but how much is not known exactly [35].)

Otherwise, one can see that for even—even nuclei the values of δ^2 vary between limits differing from each other by less than one order of magnitude. With the exception of a few questionable cases it is true that $0.02 < \delta^2 < 0.17$ if measuring δ^2 in MeV.

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REFERENCES

1. J. O. RASMUSSEN, Phys. Rev., **113,** 1593, 1959.
2. G. IGO, Phys. Rev. Letters, **1,** 72, 1958.
3. J. O. RASMUSSEN, Alpha-Decay in Alpha-, Beta- und Gamma-Ray Spectroscopy edited by K. Siegbahn, Vol. 1, p. 701, North Holland Publishing Company, Amsterdam, 1965.
4. L. KOHLMANN and T. VÖRÖS, Canad. J. Phys., **44,** 1479, 1966.
5. M. KARRAS, Ann. Acad. Sci. Fenn., **AVI.**, 65, 1960.
6. A. SIIVOLA, Ann. Acad. Sci. Fenn., **AVI.,** 109, 1962.
7. A. M. FRIEDMAN, J. MILSTED and A. L. HARKNESS, Bull. Am. Phys. Soc., **8,** 525, 1963.
8. I. OGAWA, T. DOKE, M. MIYAJIMA and A. NAKAMOTO, Nucl. Phys., **66,** 119, 1965.
9. R. D. MACFARLANE and D. W. SEEGMILLER, Nucl. Phys., **53,** 449, 1964.
10. K. S. TOTH, T. H. HANDLEY, E. NEWMAN and I. R. WILLIAMS, Phys. Rev., **136,** B 1233, 1964.
11. I. MAHUNKA and T. FÉNYES, Izv. Akad. Nauk, SSSR Ser. Fiz., **29,** 1121, 1965.
12. R. D. MACFARLANE and R. D. GRIFFIOEN, Phys. Rev., **131,** 2176, 1963.
13. G. GRAEFFE, Ann. Acad. Sci. Fenn., **AVI.,** 128, 1963.
14. R. D. MACFARLANE, J. Inorg. Nucl. Chem., **19,** 9, 1961.
15. R. D. MACFARLANE, Phys. Rev., **136,** B 941, 1964.
16. W. RIEZLER and G. KAUW, Z. Naturforschg., **13a,** 904, 1958.
17. J. O. RASMUSSEN, S. G. THOMPSON and A. GHIORSO, Phys. Rev., **89,** 33, 1953.
18. H. M. NEUMANN and I. PERLMAN, Phys. Rev., **78,** 191, 1950.
19. K. VALLI, J. AALTONEN, G. GRAEFFE, M. NURMIA and R. PÖYHÖNEN, Ann. Acad. Sci. Fenn., **AVI.,** 177, 1965.
20. I. MAHUNKA, M. MAHUNKA and T. FÉNYES, Jadernaja Fiz., **2,** 201, 1965.
21. A. SIIVOLA and G. GRAEFFE, Nuclear Phys., **64,** 161, 1965.
22. LANDOLT-BÖRNSTEIN, New-Series, Group I., Vol. 1., Springer, Berlin, 1961.
23. A. ISOLA and M. NURMIA, Z. Naturforschg., **20a,** 541, 1965.
24. R. D. MACFARLANE, UCRL-9566, 46, 1960.
25. M. NURMIA, P. KAURAUEN and A. SIIVOLA, Phys. Rev., **127,** 943, 1962.
26. R. W. HOFF, J. O. RASMUSSEN and S. G. THOMPSON, Phys. Rev., **83,** 1068, 1951.
27. K. YA. GROMOV, I. MAHUNKA, M. MAHUNKA and T. FÉNYES, Izv. Akad. Nauk SSSR, Ser. Fiz., **29,** 194, 1965.
28. K. S. TOTH, S. BJØRNHOLM, M. H. JØRGENSEN and O. B. NIELSEN, J. Inorg. Nucl. Chem., **14,** 1, 1960.
29. K. S. TOTH and J. O. RASMUSSEN, Nucl. Phys., **16,** 474, 1960.
30. K. S. TOTH, Ph. D. THESIS, UCRL-8192, 1958.
31. V. G. CHUMIN, K. YA. GROMOV, B. MAKHMUDOV and ZH. T. ZHELEV, Preprint, Dubna, JINR-E-2721, 1966.
32. A. M. FRIEDMAN, J. MILSTED and A. L. HARKNESS, Bull. Am. Phys. Soc., **8,** 525, 1963.
33. T. DOKE, Canad. J. Phys., **40,** 607, 1962.
34. R. D. MACFARLANE, Phys. Rev., **137,** B1448, 1965.
35. A. SIIVOLA, Nucl. Phys., **84,** 385, 1966; UCRL-16673, 1966 and UCRL-11828, 21, 1964.
36. R. D. MACFARLANE, R. D. GRIFFIOEN, Phys. Rev., **130,** 1491, 1963.
37. W. PORSCHEN and W. RIEZLER, Z. Naturforschg., **11a,** 143, 1956.

38. R. D. MACFARLANE and R. D. GRIFFIOEN, Bull. Am. Phys. Soc., **6,** 287, 1961.
39. P. ESKOLA, Compilation of Data on Alpha Activities with $Z > 50$, $N < 126$ Preprint of the Univ. of Helsinki, Finland, 1966.
40. M. NURMIA, G. GRAEFFE, K. VALLI and J. AALTONEN, Ann. Acad. Sci. Fenn., **AVI.,** 148, 1964.
41. R. D. MACFARLANE, Phys. Rev., **126,** 274, 1962.
42. W. RIEZLER and G. KAUW, Z. Naturforschg., **12a,** 665, 1957.
43. R. D. MACFARLANE and T. P. KOHMAN, Phys. Rev., **121,** 1758, 1961.
44. H. J. MANG., Phys. Rev., **119,** 1069, 1960.
45. M. A. PRESTON, Physics of the Nucleus, Addison-Wesley Pbl. Comp., Inc. Reading, Massachusetts, Palo Alto—London.
46. T. FÉNYES and Z. BŐDY, Acta Phys. Hung., **16,** 299, 1964.
47. A. SANDULESCU and T. VÖRÖS, Physica **31,** 83, 1965.

БАРЬЕРНЫЕ ПРОНИЦАЕМОСТИ И РЕДУЦИРОВАННЫЕ ШИРИНЫ ДЛЯ α-РАСПАДА В СРЕДНЕ-ТЯЖЕЛОМ РАЙОНЕ

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Резюме

Применением всех экспериментальных значений энергии распада и периода полураспада, имеющихся в средне-тяжелом районе, вычисляются барьерные проницаемости и редуцированные ширины α-распада. Дискутируются некоторые проблемы, вытекающие из цифровых вычислений, дается экстраполяционная формула для проницаемости в зависимости от энергии. Соответствие между относительными значениями «экспериментальной» и теоретической редуцированных ширин считается удовлетворительным.

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SPLITTINGS OF d^n-CONFIGURATIONS IN LIGAND FIELDS OF PENTAGONAL AND HEXAGONAL SYMMETRIES

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The number, the symmetry quantum numbers and spin multiplicities of the many-electron states arising from d^n-electron configurations ($n = 1, 2, 3, \ldots, 10$) in weak and strong ligand fields of pentagonal and hexagonal symmetries have been determined.

1. Introduction

In previous papers [1], on group-theoretical basis, investigations were carried out to find out about the number, symmetry species and spin multiplicities of the states occurring by the splitting of strong field configurations, arising from d^n-configurations, under the influence of tetragonal (D_4), trigonal (D_3) and rhombic (D_2) symmetries. In addition to the octahedral case [2] and the symmetries mentioned, the pentagonal and hexagonal symmetries are those which still appear among the coordination compounds of transition metals, e.g. in the sandwich molecules (complexes of dicyclopentadiene and dibenzene). Though, in some cases, by using trigonal or octahedral approximations [3], and in others by assuming D_{5h} or D_{5d} and D_{6h} [4] and even $D_{\infty h}$ [5] symmetries, calculations have already been carried out for such compounds, however, systematic studies on the splittings in fields of pentagonal and hexagonal symmetries are still lacking.

In the present paper, the calculations based on the method given in previous communications [1] have been extended to ligand fields of D_5 and D_6 symmetries.

2. Classification of the splitting products

As a starting point, the linear combinations of the linearly independent $s, p, d, f, \ldots$ etc. wave functions each belonging to one irreducible representation of the group $G (G = D_5, D_6)$ are determined. In this way, the m-dimensional ($m = 1, 3, 5, 7, \ldots$ etc.) space of these functions breaks up into subspaces which are invariant under the operations of the group G. Let us denote the representation valid in the m-dimensional reducible space with $\Gamma(G)$, then $\Gamma(G)$ will break up into irreducible representations, Γ_i, in the following form:

$$\Gamma(D_5, D_6) = \sum_i a_i \Gamma_i .$$

This means that the single electron states are split. The spaces of the irreducible representations are spanned by the linear combinations of the original functions. For the designation of these bases the same but lower case letters are used as for the corresponding representations, and for that of the many-electron (molecule) states, the corresponding capital letters, e.g. in D_5 symmetry a p- or d-electron can stay in one of the states a_2 and e_1 or a_1, e_1 and e_2, and accordingly, the splitting of the states P or D results in the molecule states of the symmetries A_2 and E_1 or A_1, E_1 and E_2, resp. Although, in the subsequent discussion, only the d^n-configurations and the interactions of such electrons will be dealt with, for the sake of completeness, all the splittings, occurring in weak fields, of orbitals or many-electron states appearing in the range of the complexes in question, will be given as well (Appendix 1). The results of such calculations are collected in Tables 1 and 2.

Table 1

Type of orbital (term)	Value of $l(L)$	Splittings in weak field of D_5 symmetry	Number of splitting products
S	0	A_1	1
P	1	$A_2 + E_1$	2
D	2	$A_1 + E_1 + E_2$	3
F	3	$A_2 + E_1 + 2E_2$	4
G	4	$A_1 + 2E_1 + 2E_2$	5
H	5	$A_1 + 2A_2 + 2E_1 + 2E_2$	7
I	6	$2A_1 + A_2 + 3E_1 + 2E_2$	8

Table 2

Type of orbital (term)	Value of $l(L)$	Splittings in weak field of D_6 symmetry	Number of splitting products
S	0	A_1	1
P	1	$A_2 + E_1$	2
D	2	$A_1 + E_1 + E_2$	3
F	3	$A_2 + B_1 + B_2 + E_1 + E_2$	5
G	4	$A_1 + B_1 + B_2 + E_1 + 2E_2$	6
H	5	$A_2 + B_1 + B_2 + 2E_1 + 2E_2$	7
I	6	$2A_1 + A_2 + B_1 + B_2 + 2E_1 + 2E_2$	9

3. Electronic interactions

In the case of d-electrons,* in compliance with the Pauli principle, two electrons can stay in state a_1 and four in each of states e_1 and e_2. The many-electron functions can be prepared from the product functions corresponding to this principle. The possible different products are the so-called strong field configurations. Generally, the many-electron functions belonging to a given strong field configuration span a space still reducible, the degeneracy of which is removed to an extent determined by the symmetry of the actual ligand field; the strong field configuration splits. To determine the splittings in D_5 and D_6 fields, first the dimension of the space (total degeneracy) formed by the functions of the strong field configuration, i.e. the number r of the linearly independent functions belonging to this space, has to be determined:

$$r = \frac{2m!}{(2\,m - n)\,!n!}\,,$$

where $2m$ denotes the number of electrons necessary to fill up the state belonging to the representation of m-dimension and n that actually staying on it. In the next step, the spurs of the reducible representation obtained in the space of the strong field configuration are to be determined, for which purpose, the transformation properties of the corresponding functions (Appendix 2) have to be established. On the basis of considerations referring to the possible strong field configurations, it is evident that the knowledge of the transformation properties is required only for the configurations $(e_1)^2$, $(e_2)^2$, $(e_1)(e_2)$, $(e_1)^2(e_2)$, $(e_1)(e_2)^2$ and $(e_1)^2(e_2)^2$, since the splitting of any other configuration can be deduced from these. In the possession of the spurs, the splitting products of the strong field configurations can easily be classified by the irreducible representations. The spin multiplicities of the splitting products are also determined in the way given in previous papers [1].

The splittings of d^1—d^{10} electron configurations in strong fields of D_5 and D_6 symmetries are given in Tables 3—12. In the first columns of the Tables there are the strong field configurations of a given d-electron configuration, in the second ones the corresponding total sums of degeneracy numbers, in the thirds the splitting products (many-electron states) and in the last columns the strong field configurations in d^{10-n}, i.e. the conjugate pairs of those in d^n, are given.

With the aim of making comparisons between the results, the numbers of the many-electron states — grouped by electron configurations and symmetries — have been presented in the Table 13.

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* Entirely the same considerations are, of course, valid for other electrons.

Table 3

D_5 symmetry

d^1	Total sums of degeneracy numbers	Many-electron states	d^9
(a_1)	2	${}^2A_1$	$(a_1)\ (e_1)^4\ (e_2)^4$
(e_1)	4	${}^2E_1$	$(a_1)^2\ (e_1)^3\ (e_2)^4$
(e_2)	4	${}^2E_2$	$(a_1)^2\ (e_1)^4\ (e_2)^3$

Table 4

D_5 symmetry

d^2	Total sums of degeneracy numbers	Many-electron states	d^8
$(a_1)^2$	1	${}^1A_1$	$(e_1)^4\ (e_2)^4$
$(e_1)^2$	6	${}^1A_1 + {}^3A_2 + {}^1E_2$	$(a_1)^2(e_1)^2(e_2)^4$
$(e_2)^2$	6	${}^1A_1 + {}^3A_2 + {}^1E_1$	$(a_1)^2(e_1)^4(e_2)^2$
$(a_1)\ (e_1)$	8	${}^{1,3}E_1$	$(a_1)\ (e_1)^3(e_2)^4$
$(a_1)\ (e_2)$	8	${}^{1,3}E_2$	$(a_1)\ (e_1)^4(e_2)^3$
$(e_1)\ (e_2)$	16	${}^{1,3}E_1 + {}^{1,3}E_2$	$(a_1)^2(e_1)^3(e_2)^3$

Table 5

D_5 symmetry

d^3	Total sums of degeneracy numbers	Many-electron states	d^7
$(e_1)^3$	4	${}^2E_1$	$(a_1)^2(e_1)\ (e_2)^4$
$(e_2)^3$	4	${}^2E_2$	$(a_1)^2(e_1)^4(e_2)$
$(a_1)^2(e_1)$	4	${}^2E_1$	$(e_1)^3(e_2)^4$
$(a_1)^2(e_2)$	4	${}^2E_2$	$(e_1)^4(e_2)^3$
$(a_1)\ (e_1)^2$	12	${}^2A_1 + {}^{2,4}A_2 + {}^2E_2$	$(a_1)\ (e_1)^2(e_2)^4$
$(a_1)\ (e_2)^2$	12	${}^2A_1 + {}^{2,4}A_2 + {}^2E_1$	$(a_1)\ (e_1)^4(e_2)^2$
$(e_1)^2(e_2)$	24	${}^2A_1 + {}^2A_2 + {}^2E_1 + 2{}^2E_2 + {}^4E_2$	$(a_1)^2(e_1)^2(e_2)^3$
$(e_1)\ (e_2)^2$	24	${}^2A_1 + {}^2A_2 + 2{}^2E_1 + {}^4E_1 + {}^2E_2$	$(a_1)^2(e_1)^3(e_2)^2$
$(a_1)\ (e_1)\ (e_2)$	32	$2{}^2E_1 + {}^4E_1 + 2{}^2E_2 + {}^4E_2$	$(a_1)\ (e_1)^3(e_2)^3$

Table 6

D_5 symmetry

d^4	Total sums of degeneracy numbers	Many-electron states	d^6
$(e_1)^4$	1	${}^1A_1$	$(a_1)^2(e_2)^4$
$(e_2)^4$	1	${}^1A_1$	$(a_1)^2(e_1)^4$
$(a_1)^2(e_1)^2$	6	${}^1A_1 + {}^3A_2 + {}^1E_2$	$(e_1)^2(e_2)^4$
$(a_1)^2(e_2)^2$	6	${}^1A_1 + {}^3A_2 + {}^1E_1$	$(e_1)^4(e_2)^2$
$(a_1)\ (e_1)^3$	8	${}^{1,3}E_1$	$(a_1)\ (e_1)\ (e_2)^4$
$(a_1)\ (e_2)^3$	8	${}^{1,3}E_2$	$(a_1)\ (e_1)^4(e_2)$
$(e_1)^3(e_2)$	16	${}^{1,3}E_1 + {}^{1,3}E_2$	$(a_1)^2(e_1)\ (e_2)^3$
$(e_1)\ (e_2)^3$	16	${}^{1,3}E_1 + {}^{1,3}E_2$	$(a_1)^2(e_1)^3(e_2)$
$(a_1)^2(e_1)\ (e_2)$	16	${}^{1,3}E_1 + {}^{1,3}E_2$	$(e_1)^3(e_2)^3$
$(e_1)^2(e_2)^2$	36	$2{}^1A_1 + {}^{3,5}A_1 + 2{}^3A_2 + 2{}^1E_1 + {}^3E_1 +$ $+ 2{}^1E_2 + {}^3E_2$	$(a_1)^2(e_1)^2(e_2)^2$
$(a_1)\ (e_1)^2(e_2)$	48	${}^{1,3}A_1 + {}^{1,3}A_2 + {}^{1,3}E_1 + 2{}^1E_2 +$ $+ 3{}^3E_2 + {}^5E_2$	$(a_1)\ (e_1)^2(e_2)^3$
$(a_1)\ (e_1)\ (e_2)^2$	48	${}^{1,3}A_1 + {}^{1,3}A_2 + 2{}^1E_1 + 3{}^3E_1 + {}^5E_1 +$ $+ {}^{1,3}E_2$	$(a_1)\ (e_1)^3(e_2)^2$

Table 7

D_5 symmetry

d^5	Total sums of degeneracy numbers	Many-electron states
$(a_1)\ (e_1)^4$	2	${}^2A_1$
$(a_1)\ (e_2)^4$	2	${}^2A_1$
$(e_1)^4(e_2)$	4	${}^2E_2$
$(e_1)\ (e_2)^4$	4	${}^2E_1$
$(a_1)^2(e_1)^3$	4	${}^2E_1$
$(a_1)^2(e_2)^3$	4	${}^2E_2$
$(e_1)^3(e_2)^2$	24	${}^2A_1 + {}^2A_2 + 2{}^2E_1 + {}^4E_1 + {}^2E_2$
$(e_1)^2(e_2)^3$	24	${}^2A_1 + {}^2A_2 + {}^2E_1 + 2{}^2E_2 + {}^4E_2$
$(a_1)^2(e_1)\ (e_2)^2$	24	${}^2A_1 + {}^2A_2 + 2{}^2E_1 + {}^4E_1 + {}^2E_2$
$(a_1)^2(e_1)^2(e_2)$	24	${}^2A_1 + {}^2A_2 + {}^2E_1 + 2{}^2E_2 + {}^4E_2$
$(a_1)\ (e_1)^3(e_2)$	32	$2{}^2E_1 + {}^4E_1 + 2{}^2E_2 + {}^4E_2$
$(a_1)\ (e_1)\ (e_2)^3$	32	$2{}^2E_1 + {}^4E_1 + 2{}^2E_2 + {}^4E_2$
$(a_1)\ (e_1)^2(e_2)^2$	72	${}^6A_1 + 2{}^4A_1 + 3{}^2A_1 + 2{}^{2,4}A_2 + {}^4E_1 + {}^4E_2 + 3{}^2E_1 + 3{}^2E_2$

Table 8

D_6 symmetry

d^1	Total sums of degeneracy numbers	Many-electron states	d^9
(a_1)	2	${}^2A_1$	$(a_1)\ (e_1)^4(e_2)^4$
(e_1)	4	${}^2E_1$	$(a_1)^2(e_1)^3(e_2)^4$
(e_2)	4	${}^2E_2$	$(a_1)^2(e_1)^4(e_2)^3$

Table 9

D_6 symmetry

d^2	Total sums of degeneracy numbers	Many-electron states	d^8
$(a_1)^2$	1	${}^1A_1$	$(e_1)^4(e_2)^4$
$(e_1)^2$	6	${}^1A_1 + {}^3A_2 + {}^1E_2$	$(a_1)^2(e_1)^2(e_2)^4$
$(e_2)^2$	6	${}^1A_1 + {}^3A_2 + {}^1E_2$	$(a_1)^2(e_1)^4(e_2)^2$
$(a_1)\ (e_1)$	8	${}^1E_1 + {}^3E_1$	$(a_1)\ (e_1)^3(e_2)^4$
$(a_1)\ (e_2)$	8	${}^1E_2 + {}^3E_2$	$(a_1)\ (e_1)^4(e_2)^3$
$(e_1)\ (e_2)$	16	${}^1B_1 + {}^3B_1 + {}^1B_2 + {}^3B_2 + {}^{1,3}E_1$	$(a_1)^2(e_1)^3(e_2)^3$

Table 10

D_6 symmetry

d^3	Total sums of degeneracy numbers	Many-electron states	d^7
$(e_1)^3$	4	${}^2E_1$	$(a_1)^2(e_1)\ (e_2)^4$
$(e_2)^3$	4	${}^2E_2$	$(a_1)^2(e_1)^4(e_2)$
$(a_1)^2(e_1)$	4	${}^2E_1$	$(e_1)^3(e_2)^4$
$(a_1)^2(e_2)$	4	${}^2E_2$	$(e_1)^4(e_2)^3$
$(a_1)\ (e_1)^2$	12	${}^2A_1 + {}^{2,4}A_2 + {}^2E_2$	$(a_1)\ (e_1)^2(e_2)^4$
$(a_1)\ (e_2)^2$	12	${}^2A_1 + {}^{2,4}A_2 + {}^2E_2$	$(a_1)\ (e_1)^4(e_2)^2$
$(e_1)^2(e_2)$	24	${}^2A_1 + {}^2A_2 + 3\,{}^2E_2 + {}^4E_2$	$(a_1)^2(e_1)^2(e_2)^3$
$(e_1)\ (e_2)^2$	24	${}^2B_1 + {}^2B_2 + 3\,{}^2E_1 + {}^4E_1$	$(a_1)^2(e_1)^3(e_2)^2$
$(a_1)\ (e_1)\ (e_2)$	32	$2\,{}^2B_1 + {}^4B_1 + 2\,{}^2B_2 + {}^4B_2 + 2\,{}^2E_1 + {}^4E_1$	$(a_1)\ (e_1)^3(e_2)^3$

Table 11

D_6 symmetry

d^4	Total sums of degeneracy numbers	Many-electron states	d^6
$(e_1)^4$	1	$^1A_1$	$(a_1)^2(e_2)^4$
$(e_2)^4$	1	$^1A_1$	$(a_1)^2(e_1)^4$
$(a_1)^2(e_1)^2$	6	$^1A_1 + {}^3A_2 + {}^1E_2$	$(e_1)^2(e_2)^4$
$(a_1)^2(e_2)^2$	6	$^1A_1 + {}^3A_2 + {}^1E_2$	$(e_1)^4(e_2)^2$
$(a_1)\ (e_1)^3$	8	$^{1,3}E_1$	$(a_1)\ (e_1)\ (e_2)^4$
$(a_1)\ (e_2)^3$	8	$^{1,3}E_2$	$(a_1)\ (e_1)\ {}^4(e_2)$
$(e_1)^3(e_2)$	16	$^{1,3}B_1 + {}^{1,3}B_2 + {}^{1,3}E_1$	$(a_1)^2\ (e_1)\ (e_2)^3$
$(e_1)\ (e_2)^3$	16	$^{1,3}B_1 + {}^{1,3}B_2 + {}^{1,3}E_1$	$(a_1)^2\ (e_1)^3(e_2)$
$(a_1)^2(e_1)\ (e_2)$	16	$^{1,3}B_1 + {}^{1,3}B_2 + {}^{1,3}E_1$	$(e_1)^3(e_2)^3$
$(e_1)^2(e_2)^2$	36	$3{}^1A_1 + {}^{3,5}A_1 + {}^1A_2 + 2{}^3A_2 + 3{}^1E_2 +$	$(a_1)^2(e_1)^2(e_2)^2$
$(a_1)\ (e_1)^2(e_2)$	48	$+\ 2{}^3E_2$	$(a_1)\ (e_1)^2(e_2)^3$
$(a_1)\ (e_1)\ (e_2)^2$	48	$^{1,3}A_1 + {}^{1,3}A_2 + 3{}^1E_2 + 4{}^3E_2 + {}^5E_2$	$(a_1)\ (e_1)^3(e_2)^2$
		$^{1,3}B_1 + {}^{1,3}B_2 + 3{}^1E_1 + 4{}^3E_1 + {}^5E_1$	

Table 12

D_6 symmetry

d^5	Total sums of degeneracy numbers	Many-electron states
$(a_1)\ (e_1)^4$	2	$^2A_1$
$(a_1)\ (e_2)^4$	2	$^2A_1$
$(a_1)^2(e_1)^3$	4	$^2E_1$
$(a_1)^2(e_2)^3$	4	$^2E_2$
$(e_1)^4(e_2)$	4	$^2E_2$
$(e_1)\ (e_2)^4$	4	$^2E_1$
$(e_1)^3(e_2)^2$	24	$^2B_1 + {}^2B_2 + 3{}^2E_1 + {}^4E_1$
$(e_1)^2(e_2)^3$	24	$^2A_1 + {}^2A_2 + 3{}^2E_2 + {}^4E_2$
$(a_1)^2(e_1)\ (e_2)^2$	24	$^2B_1 + {}^2B_2 + 3{}^2E_1 + {}^4E_1$
$(a_1)^2(e_1)^2(e_2)$	24	$^2A_1 + {}^2A_2 + 3{}^2E_2 + {}^4E_2$
$(a_1)\ (e_1)^3(e_2)$	32	$2{}^2B_1 + {}^4B_1 + 2{}^2B_2 + {}^4B_2 + 2{}^2E_1 + {}^4E_1$
$(a_1)\ (e_1)\ (e_2)^3$	32	$2{}^2B_1 + {}^4B_1 + 2{}^2B_2 + {}^4B_2 + 2{}^2E_1 + {}^4E_1$
$(a_1)\ (e_1)^2(e_2)^2$	72	$4{}^2A_1 + 2{}^4A_1 + {}^6A_1 + 3{}^2A_2 + 2{}^4A_2 + 5{}^2E_2 + 2{}^4E_2$

Table 13

Configuration	Free atom	Without multiplets					Free atom	With multiplets					Total splitting
		O_h^*	D_5	D_3,D_6	D_4	D_2		O_h^*	D_5	D_3,D_6	D_4	D_2	
d^1, d^9	1	2	3	3	4	5	2	4	6	6	8	10	10
d^2, d^8	5	11	15	17	19	25	9	19	27	31	33	45	45
d^3, d^7	8	20	30	33	37	50	20	48	72	80	88	120	120
d^4, d^6	16	43	60	67	76	100	34	87	126	139	158	210	210
d^5	16	43	60	67	76	100	44	110	152	170	192	252	252

* The results of Jorgensen [2].

Appendix 1. Splittings in weak fields

The character tables of the groups D_5 and D_6 are given in Tables 14 and 15. The corresponding reducible spur-systems determined by the Bethe method [6] are shown in the Tables 16 and 17. The results presented in Tables 1 and 2 have been obtained by the reduction of these reducible representations.

Table 14

D_5	E	$2C_5$	$2C_5^2$	$5C_2$
A_1	1	1	1	1
A_2	1	1	1	-1
E_1	2	$2b$	$2d$	0
E_2	2	$2d$	$2b$	0

where $\alpha = \frac{2\pi}{5} = 72°$, $b = \cos\alpha = \frac{\sqrt{5}-1}{4}$ and $d = \cos 2\alpha = -\frac{\sqrt{5}+1}{4}$.

Table 15

D_6	E	$2C_6$	$2C_3$	C_2	$3C_2'$	$3C_2''$
A_1	1	1	1	1	1	1
A_2	1	1	1	1	-1	-1
B_1	1	-1	1	-1	1	-1
B_2	1	-1	1	-1	-1	1
E_1	2	1	-1	-2	0	0
E_2	2	-1	-1	2	0	0

Table 16

D_5 symmetry

Value of $l(L)$	Type of orbital (term)	E	$2C_5$	$2C_5^2$	$5C_2$
0	S	1	1	1	1
1	P	3	$[2(1+b)]^{1/2}$	$-[2(1+b)]^{-1/2}$	-1
2	D	5	0	0	1
3	F	7	$-[2(1+b)]^{1/2}$	$[2(1+b)]^{-1/2}$	-1
4	G	9	-1	-1	1
5	H	11	1	1	-1
6	I	13	$[2(1+b)]^{1/2}$	$-[2(1+b)]^{-1/2}$	1

Table 17

D_6 symmetry

Value of $l(L)$	Type of orbital (term)	E	$2C_6$	$2C_3$	C_2	$3C_2'$	$3C_2''$
0	S	1	1	1	1	1	1
1	P	3	2	0	-1	-1	-1
2	D	5	1	-1	1	1	1
3	F	7	-1	1	-1	-1	-1
4	G	9	-2	0	1	1	1
5	H	11	-1	-1	-1	-1	-1
6	I	13	1	1	1	1	1

Appendix 2. Splittings in strong fields

The one-dimensional ones of the matrices of the irreducible representations in D_5 and D_6 are equal with the corresponding characters (see Tables 14 and 15). One of each two-dimensional representations are the following: in D_5

E_1:

$$E \sim \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}; \qquad C_5^1 \sim \begin{pmatrix} b & -a \\ a & b \end{pmatrix}$$

$$C_5^2 \sim \begin{pmatrix} b & a \\ -a & b \end{pmatrix}; \qquad (C_5^1)^2 \sim \begin{pmatrix} d & -c \\ c & d \end{pmatrix}$$

$$(C_5^2)^2 \sim \begin{pmatrix} d & c \\ -c & d \end{pmatrix}; \qquad C_2^1 \sim \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}$$

$$C_2^2 \sim \begin{pmatrix} -d & -c \\ -c & d \end{pmatrix}; \qquad C_2^3 \sim \begin{pmatrix} -b & a \\ a & b \end{pmatrix}$$

$$C_2^4 \sim \begin{pmatrix} -b & -a \\ -a & b \end{pmatrix}; \qquad C_2^5 \sim \begin{pmatrix} -d & c \\ c & d \end{pmatrix}$$

E_2:

$$E \sim \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}; \qquad C_5^1 \sim \begin{pmatrix} d & -c \\ c & d \end{pmatrix}$$

$$C_5^2 \sim \begin{pmatrix} d & c \\ -c & d \end{pmatrix}; \qquad (C_5^1)^2 \sim \begin{pmatrix} b & a \\ -a & b \end{pmatrix}$$

$$(C_5^2)^2 \sim \begin{pmatrix} b & -a \\ a & b \end{pmatrix}; \qquad C_2^1 \sim \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}$$

$$C_2^2 \sim \begin{pmatrix} -b & a \\ a & b \end{pmatrix}; \qquad C_2^3 \sim \begin{pmatrix} -d & c \\ c & d \end{pmatrix}$$

$$C_2^4 \sim \begin{pmatrix} -d & -c \\ -c & d \end{pmatrix}; \qquad C_2^5 \sim \begin{pmatrix} -b & -a \\ -a & b \end{pmatrix}$$

where

$$a = \sin\alpha = \frac{\sqrt{10 + 2\sqrt{5}}}{4};$$

$$c = \sin 2\alpha = \frac{\sqrt{10 - 2\sqrt{5}}}{4}.$$

in D_6

E_1:

$$E \sim \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}; \qquad C_6^1 \sim \begin{pmatrix} f & g \\ -g & f \end{pmatrix}$$

$$C_6^2 \sim \begin{pmatrix} f & -g \\ g & f \end{pmatrix}; \qquad C_3^1 \sim \begin{pmatrix} -f & g \\ -g & -f \end{pmatrix}$$

$$C_3^2 \sim \begin{pmatrix} -f & -g \\ g & -f \end{pmatrix}; \qquad C_2 \sim \begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix}$$

$$C'^1_2 \sim \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}; \qquad C'^2_2 \sim \begin{pmatrix} f & g \\ g & -f \end{pmatrix}$$

$$C'^3_2 \sim \begin{pmatrix} f & -g \\ -g & -f \end{pmatrix}; \qquad C''^1_2 \sim \begin{pmatrix} -f & g \\ g & f \end{pmatrix}$$

$$C''^2_2 \sim \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}; \qquad C''^3_2 \sim \begin{pmatrix} -f & -g \\ -g & f \end{pmatrix}$$

E_2:

$$E \sim \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}; \qquad C_6^1 \sim \begin{pmatrix} -f & -g \\ g & -f \end{pmatrix}$$

$$C_6^2 \sim \begin{pmatrix} -f & g \\ -g & -f \end{pmatrix}; \qquad C_3^1 \sim \begin{pmatrix} -f & g \\ -g & -f \end{pmatrix}$$

$$C_3^2 \sim \begin{pmatrix} -f & -g \\ g & -f \end{pmatrix}; \qquad C_2 \sim \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$$

$$C'^1_2 \sim \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}; \qquad C'^2_2 \sim \begin{pmatrix} f & g \\ g & -f \end{pmatrix}$$

$$C'^3_2 \sim \begin{pmatrix} f & -g \\ -g & -f \end{pmatrix}; \qquad C''^1_2 \sim \begin{pmatrix} f & -g \\ -g & -f \end{pmatrix}$$

$$C''^2_2 \sim \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}; \qquad C''^3_2 \sim \begin{pmatrix} f & g \\ g & -f \end{pmatrix}$$

where

$$f = \frac{1}{2} \quad \text{and} \quad g = \frac{\sqrt{3}}{2} .$$

In the following, by way of illustration, the procedure is given for the strong field configuration $(e_1)^2$ in D_5 symmetry. The spurs of the corresponding reducible representation can be determined by the transformation properties of the antisymmetrized functions spanning the space of the $(e_1)^2$ configuration (Table 18). These antisymmetrized functions are as follows:

$$\varphi_1 = 2^{-1/2}\,[e_1^+(1)_1\, e_1^+(2)_2 - e_1^+(2)_1\, e_1^+(1)_2]\,,$$
$$\varphi_2 = 2^{-1/2}\,[e_1^+(1)_1\, e_1^-(2)_1 - e_1^+(2)_1\, e_1^-(1)_1]\,,$$
$$\varphi_3 = 2^{-1/2}\,[e_1^+(1)_2\, e_1^-(2)_2 - e_1^+(2)_2\, e_1^-(1)_2]\,,$$
$$\varphi_4 = 2^{-1/2}\,[e_1^-(1)_1\, e_1^-(2)_2 - e_1^-(2)_1\, e_1^-(1)_2]\,,$$
$$\varphi_5 = 2^{-1/2}\,[e_1^-(1)_1\, e_1^+(2)_2 - e_1^-(2)_1\, e_1^+(1)_2]\,,$$
$$\varphi_6 = 2^{-1/2}\,[e_1^+(1)_1\, e_1^-(2)_2 - e_1^+(2)_1\, e_1^-(1)_2]\,,$$

where $e_1^+(j)_k$ denotes the wave function of the j-th electron of $+1/2$ spin, belonging to the k-th row of the representation E_1. By reducing the corresponding spur-system

$$\Gamma(e_1)^2 = 1A_1 + 3A_2 + 1E_2$$

is obtained which gives the spin multiplicities as well:

$$\Gamma(e_1)^2 = {}^1A_1 + {}^3A_2 + {}^1E_2 .$$

The splittings of the configurations $(e_2)^2$, $(e_1)(e_2)$, $(e_1)^2(e_2)$, $(e_1)(e_2)^2$ and $(e_1)^2(e_2)^2$ were determined in the same way as given above, by studying the transformation properties of the corresponding antisymmetrized wave functions. The splittings of other configurations can easily be traced back to those described above.

Table 18

D_5	E	C_5^1	C_5^2	$(C_5^1)^2$	$(C_5^2)^2$
φ_1	φ_1	φ_1	φ_1	φ_1	φ_1
φ_2	φ_2	$b^2\varphi_2 + a^2\varphi_3 - ab(\varphi_5 - \varphi_6)$	$b^2\varphi_2 + a^2\varphi_3 + ab(\varphi_5 - \varphi_6)$	$d^2\varphi_2 + c^2\varphi_3 - cd(\varphi_5 - \varphi_6)$	$d^2\varphi_2 + c^2\varphi_3 + cd(\varphi_5 - \varphi_6)$
φ_3	φ_3	$a^2\varphi_2 + b^2\varphi_3 + ab(\varphi_5 - \varphi_6)$	$a^2\varphi_2 + b^2\varphi_3 - ab(\varphi_5 - \varphi_6)$	$c^2\varphi_2 + d^2\varphi_3 + cd(\varphi_5 - \varphi_6)$	$c^2\varphi_2 + d^2\varphi_3 - cd(\varphi_5 - \varphi_6)$
φ_4	φ_4	φ_4	φ_4	φ_4	φ_4
φ_5	φ_5	$ab(\varphi_2 - \varphi_3) + b^2\varphi_5 + a^2\varphi_6$	$-ab(\varphi_2 - \varphi_3) + b^2\varphi_5 + a^2\varphi_6$	$cd(\varphi_2 - \varphi_3) + d^2\varphi_5 + c^2\varphi_6$	$-cd(\varphi_2 - \varphi_3) + d^2\varphi_5 + c^2\varphi_6$
φ_6	φ_6	$-ab(\varphi_2 - \varphi_3) + a^2\varphi_5 + b^2\varphi_6$	$ab(\varphi_2 - \varphi_3) + a^2\varphi_5 + b^2\varphi_6$	$-cd(\varphi_2 - \varphi_3) + c^2\varphi_5 + d^2\varphi_6$	$cd(\varphi_2 - \varphi_3) + c^2\varphi_5 + d^2\varphi_6$
$\chi(e_1)^2$	6	$1/2[7 - \sqrt{5}]$	$1/2[7 - \sqrt{5}]$	$1/2[7 + \sqrt{5}]$	$1/2[7 + \sqrt{5}]$

REFERENCES

1. F. J. GILDE and M. I. BÁN, Acta Phys. et Chem. Szeged, **5,** 3, 1959; Magy. Fiz. Folyóirat **8,** 95, 1960; Acta Phys. Hung., **12,** 13, 1960.
2. C. K. JORGENSEN, Acta Chem. Scand., **9,** 116, 1955.
3. J. W. LINNETT, Trans. Far. Soc., **52,** 904, 1956; A. D. LIEHR and C. J. BALLHAUSEN, Acta Chem. Scand., **15,** 775, 1961.
4. A. D. LIEHR and C. J. BALLHAUSEN, Acta Chem. Scand., **11,** 207, 1957; J. D. DUNITZ and L. E. ORGEL, J. Chem. Phys., **23,** 954, 1955; E. RUCH, Rec. Trav. Chim., **75,** 638, 1956; R. S. BERRY, J. Chem. Phys., **35,** 29, 2025, 1961.
5. F. A. MATSEN, J. Am. Chem. Soc., **81,** 2023, 1959.
6. H. BETHE, Ann. Phys., (5) **3,** 133, 1929.

C_2^1	C_2^2	C_2^3	C_2^4	C_2^5
$-\varphi_1$	$-\varphi_1$	$-\varphi_1$	$-\varphi_1$	$-\varphi_1$
$-\varphi_2$	$-d^2\varphi_2-c^2\varphi_3+$ $+cd(\varphi_5-\varphi_6)$	$-b^2\varphi_2-a^2\varphi_3-$ $-ab(\varphi_5-\varphi_6)$	$-b^2\varphi_2-a^2\varphi_3+$ $+ab(\varphi_5-\varphi_6)$	$-d^2\varphi_2-c^2\varphi_3-$ $-cd(\varphi_5-\varphi_6)$
$-\varphi_3$	$-c^2\varphi_2-d^2\varphi_3-$ $-cd(\varphi_5-\varphi_6)$	$-a^2\varphi_2-b^2\varphi_3+$ $+ab(\varphi_5-\varphi_6)$	$-a^2\varphi_2-b^2\varphi_3-$ $-ab(\varphi_5-\varphi_6)$	$-c^2\varphi_2-d^2\varphi_3+$ $+cd(\varphi_5-\varphi_6)$
$-\varphi_4$	$-\varphi_4$	$-\varphi_4$	$-\varphi_4$	$-\varphi_4$
φ_5	$cd(\varphi_2-\varphi_3)+$ $+d^2\varphi_5+c^2\varphi_6$	$-ab(\varphi_2-\varphi_3)+$ $+b^2\varphi_5+c^2\varphi_6$	$ab(\varphi_2-\varphi_3)+$ $+b^2\varphi_5+a^2\varphi_6$	$-cd(\varphi_2-\varphi_3)+$ $+d^2\varphi_5+c^2\varphi_6$
φ_6	$cd(\varphi_2-\varphi_3)+$ $+c^2\varphi_5+d^2\varphi_6$	$ab(\varphi_2-\varphi_3)+$ $+a^2\varphi_5+b^2\varphi_6$	$-ab(\varphi_2-\varphi_3)+$ $+a^2\varphi_5+b^2\varphi_6$	$cd(\varphi_2-\varphi_3)+$ $+c^2\varphi_5+d^2\varphi_6$
-2	-2	-2	-2	-2

РАСЩЕПЛЕНИЯ d^n-КОНФИГУРАЦИЙ В ПОЛЯХ ЛИГАНДОВ С ПЕНТАГОНАЛЬНОЙ И ГЕКСАГОНАЛЬНОЙ СИММЕТРИЕЙ

М. БАН, ДЬ. ДЭМЭТЭР и Ф. ГИЛЬДЕ

Резюме

Методами теории групп определяются многоэлектронные (молекулярные) состояния, возникающие под действием сильных и слабых полей лигандов с пентагональной (D_5) и гексагональной (D_6) симметрией в случае d^n-электронных конфигураций ($n = 1, 2, \ldots, 10$).

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MODIFICATION OF MULTIPLE SCATTERING THEORY IN VIEW OF RECENT HIGH CELL-SIZE MEASUREMENTS

By

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The paper contains a reformulation of the Voivodic—Pickup formulation of the scattering theory. Modifications are introduced for the correct introduction of the finite size limit and also for the treatment of inhomogeneous materials.

Agreement with the experimental data for nuclear emulsions at high cell sizes is clearly better with the reformulated version than with the old version.

1. Introduction

A. Hossain et al. [1, 2] (1961) found a value for the scattering constant K in nuclear emulsions at a large cell-size ($t \geq 2$ cm) lower than the value predicted by Voivodic—Pickup [3]. These authors based their calculations on both Williams [4] and Molière [5] theories. The discrepancy between the experimental and theoretical values was later confirmed by Pal and Roy [6] (1963), Chasnikov et al. [7] and Bozóki et al. [8] (1966). Bozóki et al. in their recent paper [8] compare the experimental evidence with the same Voivodic—Pickup calculations. They agree with Hossain et al. [1, 2] that a modification of the theory should be tried.

Voivodic and Pickup obtain the same results for the scattering constant dependence on cell-size whether they employed the theories of multiple scattering of Molière [5], Snyder and Scott [9], or Williams [4]. They use the Williams formulation for the calculations because of its mathematical simplicity, and introduce the Molière factor for the transitions between Born approximation and Rutherford regions. We introduce our modifications in the Voivodic—Pickup formulation for the same reason (simplicity) and also because the Voivodic—Pickup version is that most frequently employed today by experimenters. It is possible in principle to apply similar modifications to the Molière and Snyder and Scott theories. This will be examined in a following work.

In this paper we modify the Voivodic—Pickup formulation and recalculate the value of the scattering constant for Ilford Emulsions as a function of cell-size. A general formula for the calculation of the average angle α of multiple scattering as a function of the thickness for any material is also given.

The results of our calculations are now in reasonable agreement with available experimental evidence.

2. The Voivodic—Pickup formulation of the theory

According to VOIVODIC—PICKUP formulation of the WILLIAMS theory, the average angle of scattering $\bar{\alpha}$ is the sum of two contributions:

$$\bar{\alpha} = \bar{\alpha}_1 + \bar{\alpha}_2 , \tag{1}$$

where:

1) $\bar{\alpha}_1$ is the arithmetic mean value of the deflections α_1 (whose distribution is very nearly Gaussian) resulting from collisions having angles smaller than $\Phi_1 \cdot \Phi_1$ is defined by:

$$\int_{\Phi_1}^{\pi} P(\Phi)\, d\Phi = 1 , \tag{2}$$

where

$$P(\Phi) = k/\Phi^3 .$$

All angles, except where it is explicitly stated to the contrary, are given in δ units:

$$\delta = \frac{2\, Ze^2\, N^{1/2}\, t^{1/2}}{p\beta c} \tag{3}$$

for single charged incident particles.

So, the expression for Φ_1 is

$$\Phi_1 = (\pi/2)^{1/2} \tag{4}$$

and that for $\bar{\alpha}_1$

$$\bar{\alpha}_1 = \left(\log_e \frac{\Phi_1^2}{\Phi_{\min}^2}\right)^{1/2} = (\log_e M)^{1/2} = \left(\log_e \frac{M_B}{1 + \gamma^2/0.31}\right)^{1/2} , \tag{5}$$

where

$$\Phi_{\min} = 1.75\, \frac{\lambda}{a}\, (1 + \gamma^2/0.31)^{1/2} \text{ (radians)} , \tag{6}$$

$\Phi_{\min}$ is the lower limit for Φ imposed by electron screening; other symbols are defined in [3].

2) $\bar{\alpha}_2$ gives the contribution of the single scattering tail. The upper and the lower limits for α_2 are:

$$\bar{\alpha}_2\,(\max) = \bar{\gamma}_2 , \qquad \bar{\alpha}_2\,(\min) = (1 - e^{-1})\, \bar{\gamma}_2 , \tag{7}$$

where

$$\bar{\gamma}_2 = \int_0^\infty P(\alpha_2)\,\gamma(\alpha_2)\,d\alpha_2\,, \tag{8}$$

$$\gamma_2(\alpha_2) = \int_0^{\alpha_2} P_1(\alpha_1)\,(\alpha_2 - \alpha_1)\,d\alpha_1\,. \tag{9}$$

The expressions for $P_1(\alpha_1)$ and $P_2(\alpha_2)$ are given in [4].

WILLIAMS calculated $\bar{\gamma}_2$ with a numerical approximation method (see Appendix to his paper [4]) only for a thickness of one centimeter of lead. He also calculated the derivative $(d\bar{\gamma}_2/d\bar{\alpha}_1)$ for one centimeter of lead, and he supposed this derivative to be constant.

The final result obtained in this way by WILLIAMS [3, 4] is

$$\bar{\alpha} = 0.80\,\bar{\alpha}_1 + 1.45\,. \tag{10}$$

WILLIAMS assumes the existence of a limiting value for $\bar{\alpha}$ due to finite size:

$$\bar{\alpha} = \bar{\alpha}_b = \left(\log_e \frac{\Phi^2_{\text{max}}}{\Phi^2_{\text{min}}}\right)^{1/2} = (19.5 - 3.1\log_{10} Z)^{1/2}\,. \tag{11}$$

3. Proposed modification of the Voivodic—Pickup formulation

It seemed to us worth while to take into account the cut off due to finite size effects in the determination of $\bar{\alpha}$, that is, to perform the integration in (2) and (5) only up to the value Φ_{max} instead of up to the value ∞.

According to the indications given by WILLIAMS in a later paper [13], we must introduce in Φ_{max} the modification due to the use of the Born approximation. This modification is the same used by VOIVODIC and PICKUP [3] for M. The expression for Φ_{max} we shall use is:

$$\Phi_{\text{max}} = \lambda\, b^{-1}(1 + \gamma^2/0.31)^{1/2}\,(\text{radians}) = \tag{12}$$

$$= 98\,\beta r_e^{-1} N^{-1/2} Z^{-4/3} t^{-1/2}(1 + \gamma^2/0.31)^{1/2}\,(\delta - \text{units})\,, \tag{13}$$

where t is in cm.

The assumption of Φ_{max} as an actual limit for Φ is justified by the fact that for $\Phi > \Phi_{\text{max}}$ the distribution law for the projected deflection is no longer the Rutherford-like expression:

$$P(\Phi) = \pi\Phi^{-3}\,, \tag{14}$$

because the charge acting on the particle when it penetrates the nucleus is only part of the nuclear charge.

As a consequence, the actual distribution law falls to zero very quickly when $\Phi > \Phi_{\text{max}}$ and we may reasonably assume:

$$P(\Phi) = 0 \tag{15}$$

for $\Phi > \Phi_{\max}$.

The introduction of the upper limit $\Phi_{\max}$ will introduce the following modifications in the theory: ($\overline{\alpha}'$, $\overline{\alpha}_1$, $\overline{\alpha}_2$, $\overline{\gamma}_2'$, Φ_1' indicate the modified quantities)

a) Redefinition of Φ_1.

The definition of Φ_1 will be modified as follows:

$$\int_{\Phi_1'}^{\Phi_{\max}} P(\Phi)\, d\Phi = 1\,, \tag{16}$$

from which:

$$\Phi_1' = \left(\frac{\pi\Phi_{\max}^2}{2\,\Phi_{\max}^2 + \pi}\right)^{1/2}. \tag{17}$$

The formula (17) reduces to (4) when $2\Phi_{\max}^2 \gg \pi$.

Consequently the average angle $\overline{\alpha}_1$ becomes:

$$\overline{\alpha}_1' = \left(\log_e \frac{\Phi_1'^2}{\Phi_{\min}^2}\right)^{1/2} = \left(\log_e \frac{M_B}{1+\gamma^2/0.31}\;\frac{1}{1+\pi/2\,\Phi_{\max}^2}\right)^{1/2} \tag{18}$$

or

$$\overline{\alpha}_1' = \overline{\alpha}_1(1+\pi/2\,\Phi_{\max}^2)^{-1/2}. \tag{19}$$

b) Calculation of $\overline{\gamma}_2$.

Using $\Phi_{\max}$ as new upper limit, the expression of $\overline{\gamma}_2$ becomes:

$$\overline{\gamma}_2' = \int_{\Phi_1'}^{\Phi_{\max}} P_2(\alpha_2)\, d\alpha_2 \int_0^{\alpha_2} P_1(\alpha_1)\,(\alpha_2 - \alpha_1)\, d\alpha_1\,. \tag{20}$$

The analytical integration of $\overline{\gamma}_2$ (see Appendix) gives:

$$\begin{aligned}\overline{\gamma}_2' = \overline{\alpha}_1' [\exp(-\Phi_1^2/\pi\overline{\alpha}_1^2) - 1] + \frac{\pi\overline{\alpha}_1'}{2\,\Phi_{\max}^2}\{\exp(-\Phi_1^2/\pi\overline{\alpha}_1^2) - \\ - \exp(-\Phi_{\max}^2/\pi\overline{\alpha}_1^2)\} + 1/2\overline{\alpha}_1'\{E_i(-\Phi_{\max}^2/\pi\overline{\alpha}_1^2) - E_i(-\Phi_1^2/\pi\overline{\alpha}_1^2)\} + \\ + 2\,\pi/\Phi_1\,\Psi_0\left(\sqrt{\frac{2}{\pi}}\,\frac{\Phi_1}{\overline{\alpha}_1'}\right) - 2\,\pi/\Phi_{\max}\,\Psi_0\left(\sqrt{\frac{2}{\pi}}\,\frac{\Phi_{\max}}{\overline{\alpha}_1'}\right)\,.\end{aligned} \tag{21}$$

Also here as in [4] we have:

$$\overline{\alpha}_2'(\max) = \overline{\gamma}_2'\,, \tag{22}$$

$$\overline{\alpha}_1'(\min) = \overline{\alpha}_2'(1 - e^{-1})\,. \tag{22'}$$

As a final result we have

$$\overline{\alpha}' = \overline{\alpha}_1' + \overline{\alpha}_2'. \tag{23}$$

Let us examine the limiting case $t_2 \to \infty$.

From (17), owing to the fact that $\Phi^2_{\max}$ is proportional to t^{-1}, we have

$$\lim_{t\to\infty} \Phi_1' = \Phi_{\max}. \tag{24}$$

As a consequence:

a) from (18)

$$\lim_{t\to\infty} \overline{\alpha}_1' = \overline{\alpha}_b\,, \tag{25}$$

where $\overline{\alpha}_b$ is the Williams old limit value defined by (11);

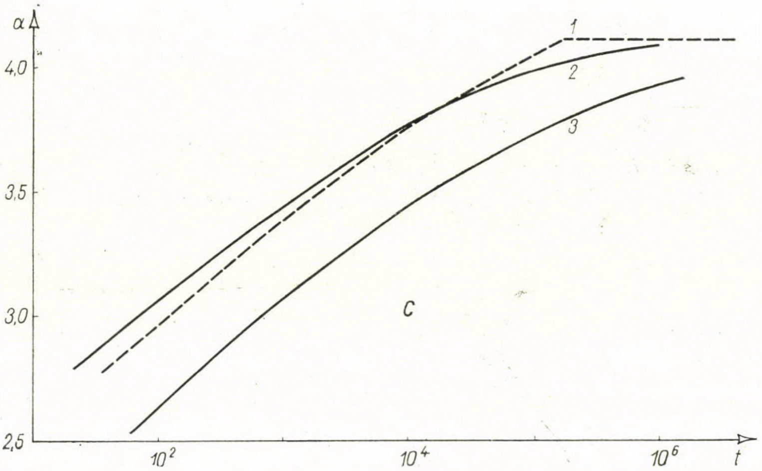

Fig. 1. $\overline{\alpha}_{\text{theor.}}$ in δ-units as a function of target thickness t in μm, for carbon. Curve 1 : Williams original calculation, 2 : Our calculation (upper limit), 3 : Our calculation (lower limit)

b) from (20)

$$\lim_{t\to\infty} \overline{\gamma}_2' = 0 \tag{26}$$

being the integral made between two identical limits.

So, from (23), (25), (26) we obtain:

$$\lim_{t\to\infty} \overline{\alpha}' = \overline{\alpha}_b. \tag{27}$$

The finite size limit is so approached asymptotically in our reformulation of the theory.

We have calculated $\overline{\alpha}_b$ with a more exact approximation of the coefficients, so obtaining instead of (11) the expression:

$$\overline{\alpha}_b = (19.27 - 3.07 \log_{10} Z)^{1/2}. \tag{27'}$$

In Figs. 1, 2, 3 $\overline{\alpha}'$ is plotted for carbon, aluminum and lead (calculated from (19), (21), (23) for $\beta \approx 1$). In these Figures $\overline{\alpha}_{\text{Will}}$ is also plotted.

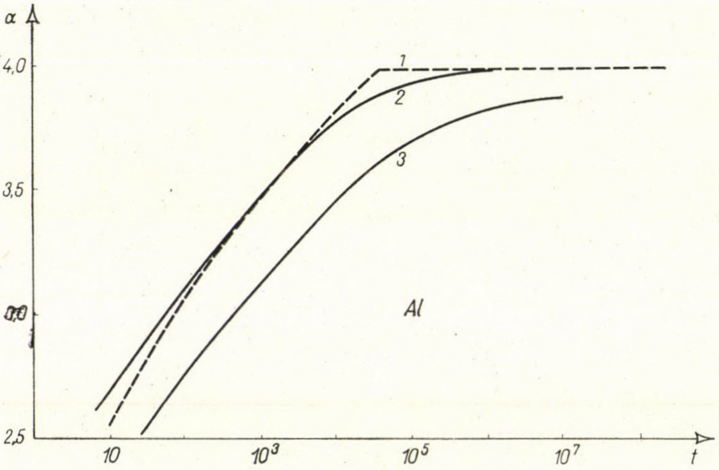

Fig. 2. $\bar{\alpha}_{\text{theor.}}$ in δ-units as a function of target thickness in μm, for aluminum. Curve 1 : WILLIAMS original calculation, 2 : Our calculation (upper limit), 3 : Our calculation (lower limit)

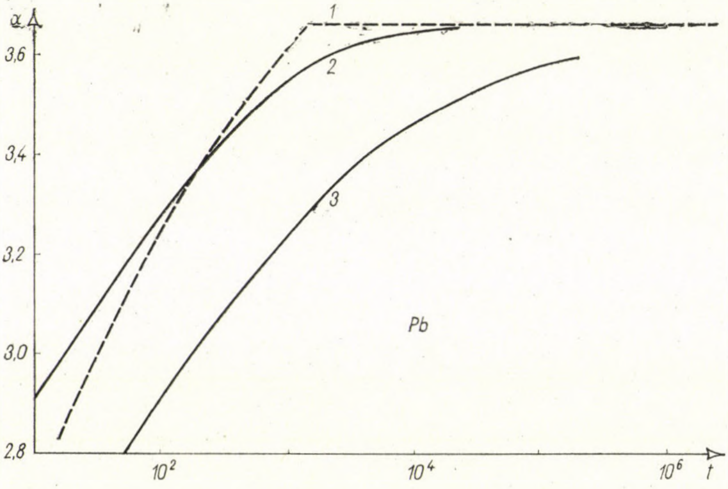

Fig. 3. $\bar{\alpha}_{\text{theor.}}$ in δ-units as a function of target thickness in μm, for lead. Curve 1 : WILLIAMS original calculation, 2 : Our calculation (upper limit), 3 : Our calculation (lower limit)

4. Application to the case of nuclear emulsions

In the case of nuclear emulsions it is usual to introduce a scattering constant K in the place of $\bar{\alpha}$ (in δ units) by means of the relation:

$$K = (2/3)^{1/2} \cdot (180/\pi) \cdot (t/100)^{-1/2} \cdot p\beta \cdot \bar{\alpha}' \cdot \delta \, \frac{\text{degrees} \times \text{Mev}/c}{(100\mu)^{1/2}}\,, \tag{28}$$

t is measured in microns. The factor $(2/3)^{1/2}$ comes from the use of the chord method of measurements.

Using (18), (21), (23) and (28) we can obtain K for a given element, characterized by a given Z and a given concentration of n atoms per cc.

Nuclear emulsions, however, are a mixture of several elements each characterized by its own Z_i and n_i. In order to obtain the value of K for inhomogeneous materials we have preferred to use standard straight-forward approximation instead of MOLIÈRE's mixtures method:

$$K^2 = \Sigma_i K_i^2 , \tag{29}$$

where

$$K_i = (2/3)^{1/2} (t/100)^{-1/2} (180/\pi) \cdot p \cdot \beta \cdot \bar{\alpha}'_i \cdot \delta_i = 0.1347 \cdot 10^{-11} \cdot (n_i Z_i^2 \bar{\alpha}_i^2)^{1/2} . \tag{30}$$

We recall that $\bar{\alpha}'$ is the sum of two terms $\bar{\alpha}'_1$ and $\bar{\alpha}'_2$ and that $\bar{\alpha}'_2$ may take any value between $\bar{\gamma}'_2$ and $(1-e^{-1})\bar{\gamma}'_2$. As a consequence we give two limiting values for K:

$$K(\max) = 0.1347 \cdot 10^{-11} \cdot [\Sigma_i n_i Z_i (\bar{\alpha}'_{1i} + \bar{\gamma}'_{2i})^2]^{1/2} , \tag{31}$$

$$K(\min) = 0.1347 \cdot 10^{-11} \cdot \{\Sigma_i n_i Z_i [\bar{\alpha}'_{1i} + \bar{\gamma}'_{2i} (1 - e^{-1})]^2\}^{1/2} . \tag{32}$$

WILLIAMS decided to use the value

$$\bar{\alpha}_2(\text{Will}) = 0.94 \, \bar{\gamma}_2 . \tag{32'}$$

We prefer to admit the possibility that $\bar{\alpha}'_2$ may assume any value between the limiting values and leave the comparison with experience to decide the exact value.

Notice that:

$$\lim_{t \to \infty} K(\min) = \lim_{t \to \infty} K(\max) = 30.74 = K_b . \tag{33}$$

This value of K_b can also be obtained using (27').

Formulae (31) and (32) are plotted in Fig. 4, for $\beta \simeq 1$ (curves 2 and 3, respectively).

Our value for K_b (30.74) is slightly lower than the value used by VOIVODIC and PICKUP ($K = 31.2$) because we have preferred to treat the inhomogeneity by means of (29) rather than with the MOLIÈRE method of mixtures.

In Fig. 4 the K calculated by VOIVODIC—PICKUP [3] are plotted (using (32')) (curve 1).

In the same Fig. 4 the experimental values of K obtained by several authors [6, 7, 8, 10, 11] are also plotted. We have taken only measurements which have $\beta \geqq 0.98$ and which have not used cut-off procedure. These two conditions are not restrictive in the high cell-size region but have greatly reduced the number of measurements we could accept in the low cell-size region. Measurements of different authors which were made at equal or near cell-sizes and which were statistically compatible were put together by us, as indicated in Table I. Measurements which correspond to the intermediate cell-size region (1 mm. $< t <$ 1 cm) were not included because spurious scattering is too important in that region [14].

We have also calculated K (V.P.) using for $\bar{\alpha}_2$ the lower value $(1-e^{-1})\bar{\gamma}_2$ (curve 4 in Fig. 4).

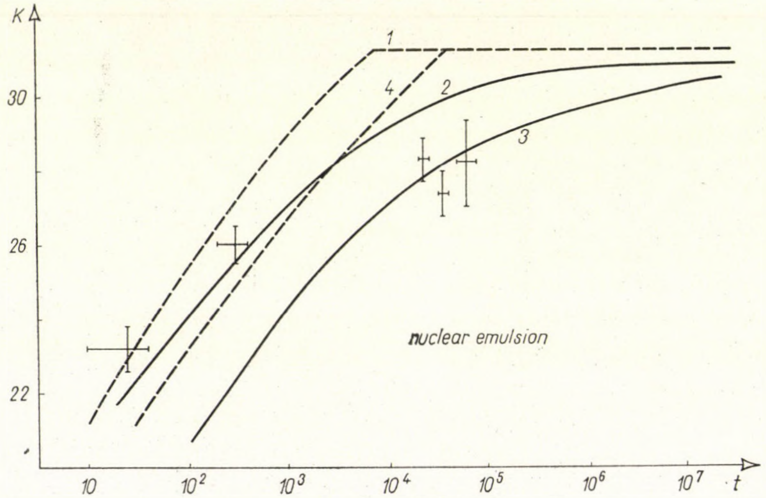

Fig. 4. Multiple scattering constant K in degrees. MeV/c. $(100\ \mu m)^{-1/2}$, as a function of cell-size t (in μm). Curve 1 : Voivodic—Pickup formulation (for $\bar{\alpha}_3 = 0.94\ \bar{\gamma}_3$), 2 : Our calculation (upper limit), 3 : Our calculation (lower limit), 4 : Voivodic—Pickup formulation (for $\bar{\alpha}_2 = (1-e^{-1})\bar{\gamma}_2$)

Table 1

t(mm)	K(average)	Ref.
0.025 ± 0.015	23.2 ± 0.6	[10]
0.30 ± 0.1	26.0 ± 0.5	[11]
22.5 ± 2.5	28.3 ± 0.6	[2, 8]
35.0 ± 5.0	27.3 ± 0.6	[8, 2, 7]
65.0 ± 15.0	28.1 ± 1.1	[8, 6]

5. Conclusions

For the high cell size region, the experimental values fall between curves 2 and 3, that is inside the band calculated by us and definitely not in the band which corresponds to the VOIVODIC—PICKUP theory (region between curves 1 and 4).

In the low cell-size region our upper curve 2 approaches VOIVODIC—PICKUP upper curve 1 and both curves fit the experimental values reasonably.

We conclude that the available experiments suggest that at high cell-sizes the lower curve is to be preferred in any case, but definitely our lower curve 3 fits the data much better than VOIVODIC—PICKUP lower curve 4.

It would be interesting for measurements to be made in the high cell-size region in other elements such as Al, C, Pb, to check the conclusions we arrived at in the case of nuclear emulsions. The already existing measurements [12, 13] give sometimes conflicting results. They seem to indicate, however, that at least in Pb the experimental values are lower than curve 1.

6. Appendix

According to WILLIAMS [4] (Appendix B), the contribution of scattering through angles $\Phi > \Phi_1'$ is related to the quantity $\overline{\gamma}_2'$ expressing the average deflection when the particle suffers only one collision with $\Phi > \Phi_1'$.

$\overline{\gamma}_2'$ is given by

$$\overline{\gamma}_2' = \int_{\Phi_1'}^{\Phi_{\max}} P_2(\alpha_2)\, d\alpha_2 \int_0^{\alpha_2} P_1(\alpha_1)\,(\alpha_2 - \alpha_1)\, d\alpha_1\,, \tag{34}$$

where α_2 is the projected deflection Φ with the condition $\Phi > \Phi_1'$ (which imposes the lower limit to the integration on α_2). Of course α_2, being the symbol for the values of $\Phi > \Phi_1'$, is subjected to the condition $\Phi < \Phi_{\max}$, so that the upper limit of integration is $\Phi_{\max}$.

Relation (34) can be rewritten in the form:

$$\begin{aligned} \overline{\gamma}_2' = & \int_{\Phi_1'}^{\Phi_{\max}} P_2(\alpha_2)\, d\alpha_2 \int_0^{\Phi_1'} P_1(\alpha_1)\,(\alpha_2 - \alpha_1)\, d\alpha_1 + \\ & + \int_{\Phi_1'}^{\Phi_{\max}} P_2(\alpha_2)\, d\alpha_2 \int_{\Phi_1'}^{\alpha_2} P_1(\alpha_1)\,(\alpha_2 - \alpha_1)\, d\alpha_1 = \overline{\gamma}_{21}' + \overline{\gamma}_{22}'\,. \end{aligned} \tag{35}$$

$\overline{\gamma}_{21}'$ can be calculated directly as follows:

$$\begin{aligned} \overline{\gamma}_{21}' & = 2/\overline{\alpha}_1' \int_{\Phi_1'}^{\Phi_{\max}} \alpha_2^{-3}\, d\alpha_2 \int_0^{\Phi_1'} \exp\left(-\alpha_1^2/\pi\overline{\alpha}_1'^2\right)(\alpha_2 - \alpha_1)\, d\alpha_1 = \\ & = \overline{\alpha}_1' \left[\exp\left(-\Phi_1'^2/\pi\overline{\alpha}_1^2\right) - 1\right] + 2\pi\left(\Phi_1'^{-1} - \Phi_{\max}^{-1}\right) \Psi_0 \left(\sqrt{\frac{2}{\pi}} \cdot \frac{\Phi_1'}{\overline{\alpha}_1'}\right), \end{aligned} \tag{36}$$

where ψ_0 is the error function:

$$\psi_0(x) = (2\pi)^{-1/2} \int_0^x \exp(-t^2/2)\, dt. \tag{37}$$

In order to calculate $\bar{\gamma}'_{22}$ we may observe that the integration field is triangular. So we may use the Dirichlet inversion formula:

$$\begin{aligned}
\bar{\gamma}'_{22} &= \int_{\Phi'_1}^{\Phi_{\max}} P_2(\alpha_2)\, d\alpha_2 \int_{\Phi'_1}^{\alpha_2} P_1(\alpha_1)(\alpha_2 - \alpha_1)\, d\alpha_1 = \\
&= \int_{\Phi'_1}^{\Phi_{\max}} P_1(\alpha_1)\, d\alpha_1 \int_{\alpha_1}^{\Phi_{\max}} P_2(\alpha_2)(\alpha_2 - \alpha_1)\, d\alpha_2 = \\
&= 2/\bar{\alpha}'_1 \int_{\Phi'_1}^{\Phi_{\max}} \exp(-\alpha_1^2/\pi\bar{\alpha}_1^2)\, d\alpha_1 \int_{\alpha_1}^{\Phi_{\max}} (\alpha_2 - \alpha_1)\, \alpha_2^3\, d\alpha_2 = \\
&= (2\bar{\alpha}'_1)^{-1} \{E_i(-\Phi_{\max}^2/\pi\bar{\alpha}_1'^2) - E_i(-\Phi_1'^2/\pi\bar{\alpha}_1'^2)\} + \frac{\pi\bar{\alpha}_1}{2\Phi_{\max}^2} \{\exp(-\Phi_1'^2/\pi\bar{\alpha}_1^2) - \\
&\quad - \exp(-\Phi_{\max}^2/\pi\bar{\alpha}_1^2)\} + 2\pi/\Phi_{\max} \left\{\Psi_0\left(\frac{\Phi'_1}{\bar{\alpha}'_1}\sqrt{\frac{2}{\pi}}\right) - \Psi_0\left(\frac{\Phi_{\max}}{\bar{\alpha}'_1}\sqrt{\frac{2}{\pi}}\right)\right\},
\end{aligned} \tag{38}$$

where E_i is the exponential integral:

$$E_i(-x) = -\int_x^{\infty} \exp(-t) \cdot t^{-1}\, dt. \tag{39}$$

The final expression of $\bar{\gamma}'_2$, i.e. the sum of (36) and (38), is given in Section 3. The expression of $\bar{\gamma}_2$ given in Section 2 may be obtained from our expression as a limiting case when:

$$\begin{aligned}
&\Phi_{\max} \to \infty, \\
&\Phi'_1 \to \Phi_1 = (\pi/2)^{1/2}.
\end{aligned} \tag{40}$$

REFERENCES

1. A. Hossain, M. F. Votruba and A. Wataghin, Il Nuovo Cimento, **22,** 308, 1961.
2. A. Hossain, M. F. Votruba, A. Wataghin and D. Evans, Il Nuovo Cimento, **22,** 861, 1961.
3. L. Voivodic and E. Pickup, Phys. Rev., **85,** 91, 1952.
4. E. J. Williams, Proc. Royal Soc., A **169,** 531, 1938.
5. G. Moliere, Z. Naturforsch., **2a,** 133, 1947.
6. Y. Pal and A. K. Roy, Il Nuovo Cimento, **27,** 960, 1963.
7. J. J. Chasnikov, Z. V. Anzon, Z. S. Takibaev and J. S. Strelecov, J. E. T. P. **45,** 29, 1963.
8. G. Bozóki, E. Gombosi, L. Jenik, E. Nagy and M. Sahini, Acta Phys. Hung., **20,** 133, 1966.
9. H. S. Snyder and W. T. Scott, Phys. Rev., **76,** 220, 1949.
10. C. Violet, Phys. Rev., **104,** 1454 1956.
11. K. Gottstein, M. G. K. Menon, J. H. Mulvey, C. O'Cellaigh and O. Rochat, Phil. Mag., **42,** 708, 1951.
12. N. I. Oleson, K. T. Kao and H. R. Crane, Phys. Rev., **60,** 378, 1941.
13. C. W. Sheppard and W. A. Fowler, Phys. Rev., **57,** 283, 1940.
14. A. Marzari Chiesa and A. Wataghin, Suppl. Nuovo Cimento, **26,** 279, 1962.

МОДИФИКАЦИЯ ТЕОРИИ МУЛЬТИПЛЕТНОГО РАССЕЯНИЯ С ТОЧКИ ЗРЕНИЯ НОВЫХ ЯЧЕЕК БОЛЬШОГО РАЗМЕРА

А. АГНЕСЕ, Г. ПАОЛИ, М. СКОТТО и А. ВАТАГИН

Резюме

В работе содержится переработка формулировки Войводич—Пикап теории рассеяния. Модификации вводятся для корректного введения границы конечного размера и для рассмотрения неоднородных материалов.

Совпадение с экспериментальными данными для ядерных эмульсий при ячейках большого размера явно лучше в случае применения преобразованного вида теории по сравнению со случаем применения старой версии.

Acta Physica Academiae Scientiarum Hungaricae, Tomus 25 (1), pp. 83—90 (1968)

EXPERIMENTELLE ANORDNUNGEN MIT A. P. M.-PRINZIP FÜR POLARISATIONSMESSUNGEN

Von

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INSTITUT FÜR KERNFORSCHUNG DER UNGARISCHEN AKADEMIE DER WISSENSCHAFTEN (ATOMKI), DEBRECEN

(Vorgelegt von A. Szalay. — Eingegangen 14. XI. 1967)

Der Artikel beschreibt zwei Messanordnungen, die auf dem a. p. m. (associated particle method)-Prinzip beruhen und für Polarisationsmessungen entwickelt wurden. Durch den in Koinzidenz erfolgenden Nachweis der Neutronen und ^{3}He-Teilchen der D(d, n) ^{3}He-Reaktion ist es möglich, einen Neutronenstrahl genau definierter Energie und Richtung zu erhalten; ausserdem ist mit der Registrierung der ^{3}He-Teilchen eine einwandfreie Monitorisierung gewährleistet.

1. Einleitung

Bei der Untersuchung von Kernprozessen, die die DD-oder DT-Reaktion als Neutronenquelle verwenden bzw. sich mit den beiden Reaktionen selbst befassen, hat sich aus mehreren Gründen die sog. »associated particle method« (im Folgenden mit a. p. m. bezeichnet) als sehr geeignet erwiesen. Hierbei werden die Neutronen der DD- bzw. DT-Reaktion in Koinzidenz mit den gleichzeitig entstehenden ^{3}He- bzw. ^{4}He-Rückstosskernen nachgewiesen.

Durch Anwendung dieser Methode kann ein Neutronenstrahl genau bekannten Flusses, sowie definierter Energie und Richtung erhalten werden. Das ergibt sich einmal aus der Tatsache, dass die geladenen ^{3}He- oder ^{4}He-Teilchen mit einem 100%-igen Wirkungsgrad nachgewiesen werden können, wodurch der Neutronenfluss genau bestimmt ist, und zum anderen daraus, dass die geometrische Anordnung von Helium- und Neutronendetektoren, sowie die Kinematik der Reaktion die Energie und Richtung der Neutronen eindeutig definieren. Ausserdem erreicht man durch die Koinzidenzmethode eine bedeutende Verringerung des störenden Untergrundes, der von gestreuten oder in Selbsttargeten entlang der Beschleunigungsröhre erzeugten Neutronen, sowie deren Gammastrahlung verursacht wird. Für Flugzeitmessungen bietet sich diese Methode ebenfalls an, weil der Nachweis der Rückstosskerne genau den Zeitpunkt der Neutronenerzeugung angibt.

Die genannten Eigenschaften der a. p. m. stimmen im wesentlichen mit den Forderungen an die Messapparatur überein, die man im Falle von Polarisationsmessungen stellen muss; besonders wichtig sind hier die Kollimation des Neutronenstrahls, die genaue Monitorisierung sowie ein möglichst geringer Untergrund, da der zu messende Effekt sich im allgemeinen nur wenig aus dem

Untergrund heraushebt. Das legt die Anwendung der a. p. m. auch für Polarisationsmessungen nahe.

1.1. *Probleme bei der Anwendung der a. p. m.*

Die Schwierigkeit der Entwicklung bzw. der Anwendung der a. p. m. bei der D(*d*, *n*)^{3}He-Reaktion besteht darin, dass man die mit den Neutronen koinzidierenden ^{3}He-Rückstosskerne von den elastisch am Target gestreuten Deuteronen, sowie den Protonen und Tritonen der D(*d*, *p*)T-Konkurrenzreaktion getrennt nachweisen muss. Die Trennung dieser Teilchengruppen erfolgt einmal über die Auswahl der ^{3}He-Teilchen aus dem Energiespektrum des Detektors und zum anderen, das trifft auf die Deuteronen zu, durch ein Trennungsverfahren noch vor dem Detektor.

Um den Detektor vor der grossen Zahl der gestreuten Deuteronen zu schützen und weiterhin die Übersteuerung der Elektronik, sowie eine Verzerrung des Spektrums durch »pile-up«-Effekte zu verhindern, verwendet man zumeist Schutzfolien (Al, Ni) vor dem Detektor. Diese Folien dienen gleichzeitig als Lichtschutz gegen Licht vom Target. Im Zusammenhang mit der Anwendung von Folien zum Abbremsen der Deuteronen muss noch gesagt werden, dass die Wahl der Folienstärke sehr kritisch ist. Die ^{3}He-Teilchen sollen die Folie mit einem möglichst geringen Energieverlust durchlaufen, während die Deuteronen ausgesiebt werden sollen, andererseits ist der Energieunterschied zwischen diesen Teilchen verhältnismässig gering. Ausserdem gibt es für die Folienstärke eine prinzipielle Grenze, da die Deuteronen- und ^{3}He-Energie nicht proportional wachsen; bei Winkeln über 85° für das ^{3}He-Teilchen fällt die ^{3}He-Energie sogar mit steigender Deuteronenenergie. Das ermöglicht die Verwendung von Folien nur bei niedrigen ($E_d < 150$ keV) Deuteronenenergien.

In der Literatur haben sich eine ganze Reihe von Autoren mit der a. p. m. befasst, z. B. [1—8]. In den früheren Arbeiten [3—8] werden als Helium-Detektor ausschliesslich Szintillationszähler mit organischen oder anorganischen Kristallen verwendet. Die Nachteile der Szintillationszähler sind aber eine starke Gammaempfindlichkeit und ein schlechtes Energieauflösungsvermögen, was sich zusammen mit der Anwendung von Folien, die ja zu einer wesentlichen Energieverbreiterung der durchgehenden Teilchenstrahlung führen, besonders nachteilig auswirkt. Der Vorteil der Szintillationszähler (besonders im Falle von organischen Szintillatoren) besteht aber darin, dass mit ihnen ein sehr schneller Koinzidenzkreis (10^{-8} sec.) ausgebaut werden kann.

In neueren Arbeiten [1, 2] kommen des besseren Energieauflösungsvermögens wegen vorwiegend Halbleiterdetektoren zum ^{3}He-Nachweis zur Anwendung. Damit können die Protonen und Tritonen ohne Schwierigkeiten von den ^{3}He-Teilchen abgetrennt werden. Gegen die gestreuten Deuteronen werden auch hier wieder teils Folien [1] (bei niedrigen E_d-Energien) und teils ein magnetisches oder elektrostatisches Ablenksystem [2] verwendet.

Im Folgenden beschreiben wir zwei auf den oben genannten Varianten beruhende a. p. m.-Systeme mit Halbleiterdetektoren, die für Messungen der Polarisation der DD-Neutronen sowie des totalen Wirkungsquerschnittes im entsprechenden Energiebereich entwickelt wurden.

2. Experimentelle Anordnung

Die Geometrie der Versuchsanordnung wurde durch die Polarisationsmessungen festgelegt und ist auf Abb. 1 bzw. Abb. 2 angegeben. Der Winkel zwischen dem beschleunigten Deuteronenstrahl und dem der nachgewiesenen ^{3}He-Rückstosskerne beträgt 112°, so dass der koinzidierende Neutronenstrahl etwa unter einem Winkel von 50° zum einfallenden Deuteronenstrahl austritt [9], wo der maximale Polarisationsgrad der DD-Neutronen zu erwarten ist [11]. Diese geometrischen Beziehungen gelten für beide gebauten Messanordnungen. Abb. 1 legt die erste Anordnung in Skizze dar.

Die ^{3}He-Teilchen wurden mit einem Siliziumhalbleiterdetektor registriert, der einen spezifischen Widerstand von 2000 Ω cm und eine Fläche von 12,56

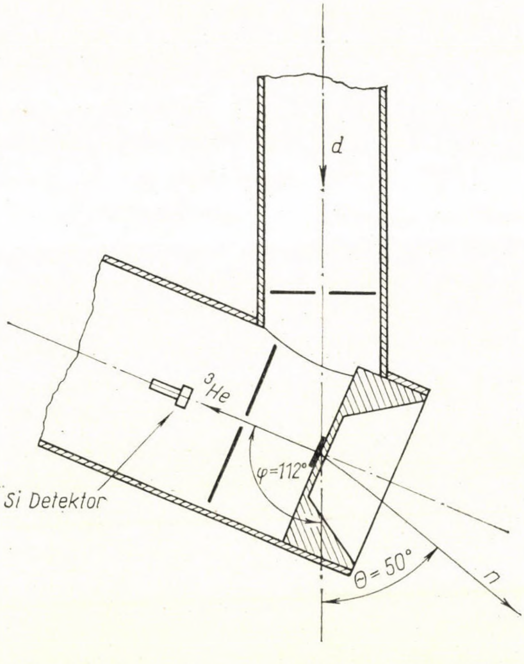

Abb. 1. Die geometrische Anordnung des a. p. m.-Systems. Der Halbleiterdetektor wird durch eine Al-Folie gegen die in grosser Zahl am Target gestreuten Deuteronen geschützt. Die Anordnung ist nur bei niedrigen Beschleunigungsspannungen anwendbar

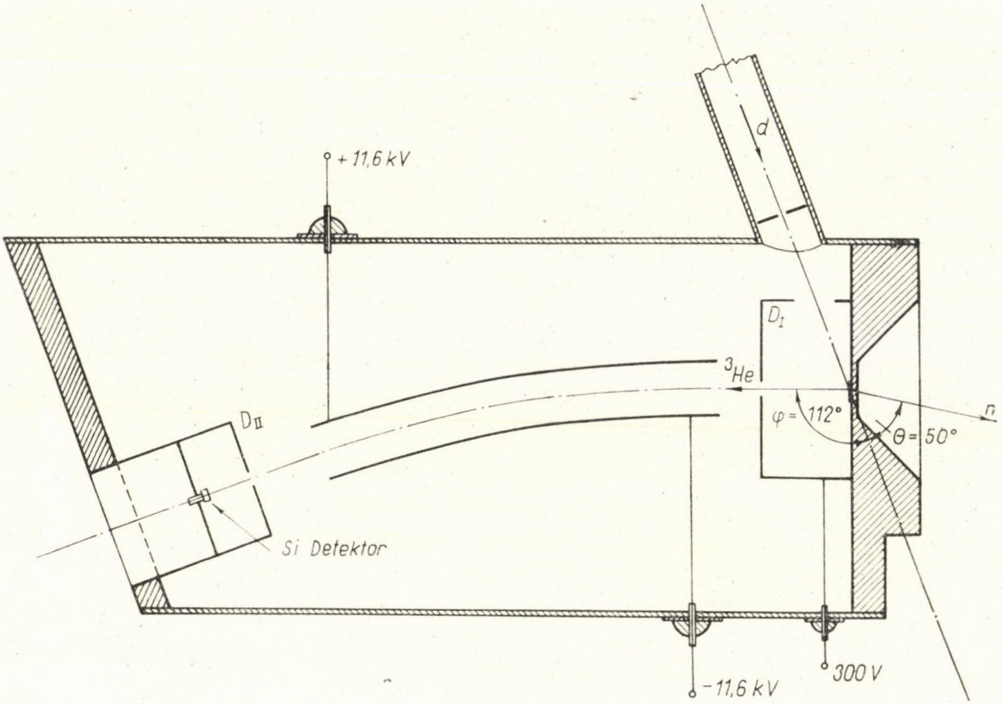

Abb. 2. a. p. m.-System mit elektrostatischer Ablenkung. Hier werden die Teilchen im Feld eines Zylinderkondensators elektrostatisch abgelenkt und der Si-Detektor so vor den gestreuten Deuteronen geschützt

mm^2 besass. Die Entfernung Detektor—Target betrug 36 mm. Die Al-Folie hatte eine Stärke von 0,18 mg/cm. Bei der Wahl der Folienstärke sind wir einen Kompromiss eingegangen, bei dem die Zahl der Deuteronen ausreichend gesenkt wurde, der Energieverlust und die Energieverbreiterung des ^{3}He-Strahles aber klein blieben.

Für die Messungen bei höheren Deuteronenenergien wendeten wir ein elektrostatisches Ablenksystem an. Sein prinzipieller Aufbau ist in Abb. 2 dargestellt.

Mit Hilfe eines ersten Diaphragmas wurden die unter $112^0 \pm 2{,}5^0$ austretenden ^{3}He-Teilchen ausgewählt. Dieses Diaphragma war isoliert am Targethalter befestigt und erhielt eine negative Spannung von — 300 V, um die bei der Abbremsung des beschleunigten Deuteronenstrahls im Target entstehenden Elektronen zurückzustossen, da diese sonst eine Ionisation des Restgases im Ablenkraum und einen Zusammenbruch der Ablenkspannung hervorrufen würden. Der Teilchenstrahl durchlief darauf das Feld eines Zylinderkondensators (mittlerer Radius 50 cm, Sektorausschnitt 20°, Plattenabstand 2 cm) und gelangte durch ein zweites Diaphragma hindurch in den Detektor. An das Plattenpaar des Ablenksystems wurde je nach Höhe der Beschleunigungsspannung eine symmetrische Spannung von ± 9 kV bis ± 13 keV geführt. Die Ablenkspannung wurde dabei so gewählt, dass die He-Teilchen, die bei einer bestimmten maximalen Deuteronenenergie zu der mittleren Energie der Neutronen korrespondieren, bei senkrechtem Eintritt das Plattenpaar auf einen Kreisbogen von $r = 50$ cm durchlaufen; z.B. betrug sie bei $E_d = 190$ keV, d.h. $E_d = 137$ keV, 11,6 kV.

Die Impulse des Halbleiterdetektors wurden über einen ladungsempfindlichen Vorverstärker (V.V.) in einen differenzierenden und integrierenden Hauptverstärker (H.V.I) geleitet. Es war schwierig, den Vorverstärker vor dem hochfrequenten Feld der Ionenquelle des Neutronengenerators und vor Kriechströmen zu schützen. Durch sorgfältige Abschirmung, sowie dicke Kupfererdleitungen gelang es, diese anfänglichen Störeffekte völlig zu beseitigen.

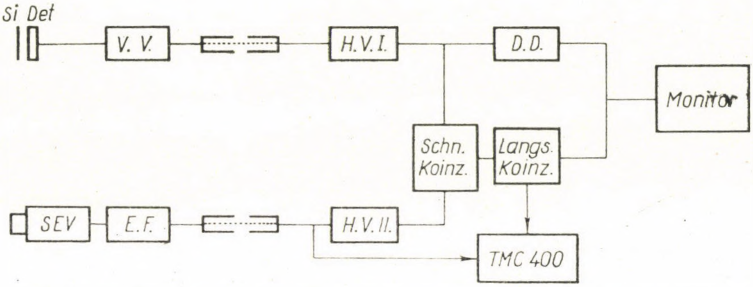

Abb. 3. Das Blockschema der verwendeten Elektronik

Die Neutronen wurden mit einem Stilbenkristall (∅ 3,7 cm, Länge 3,5 cm) detektiert. Dem Stilbenkristall schloss sich ein M12FS60 — Zeiss-SEV an. Die Impulse des SEV gelangten über einen Emitterfolger (E.F) einmal in den TMC 400 — Kanalanalysator und zum anderen nach entsprechender Verstärkung (H.V.II) in einen Koinzidenzkreis vom Typ eines Zeitamplitudenkonverters (Schn. Koinz.)

Der zweite Eingang des Koinzidenzkreises bekam seine Steuerung vom Hauptverstärker des Halbleiterdetektors (H.V.1). Die Koinzidenzanordnung verfügte auch über einen langsamen Zweig (langs. Koinz.) wodurch es möglich war, die koinzidierenden Impulse mit einem im Stromkreis des Halbleiterdetektors befindlichen Einkanalanalysators (D. D) auch energetisch zu selektrieren. Der Ausgang des langsamen Koinzidenzkreises lieferte den Torimpuls für den Vielkanalanalysator. So traten im Neutronenspektrum nur Impulse von den Neutronen auf, die mit vom Analysator des Halbleiterdetektors ausgewählten ^{3}He-Teilchen in Koinzidenz waren. Das Zeitauflösungsvermögen des schnellen Koinzidenzkreises betrug 10^{-7} sec. Diese Grenze in der Auflösung wurde durch das »Jitter« der Impulse des Halbleiterdetektors verursacht. Die Monitorisierung geschah mit einem an den Analysator des Halbleiterdetektors angeschlossenen Zähler (Monitor).

Die Messungen erfolgten am 300 kV-Generator des Institutes [10]. Der beschleunigte und durch ein Diaphragma ausgeblendete Deuteronenstrahl von 40 μ A traf unter einem Winkel von 22° auf ein dickes Ti—D-Target, wodurch sich ein elliptischer Targetfleck mit einer grossen Halbachse von 3 mm und einer kleinen von 1 mm bildete. Dieser Winkel wurde gewählt, damit die ^{3}He-Kerne senkrecht aus dem Target austreten und ihr Energieverlust im Target selbst so niedrig als möglich gehalten wird.

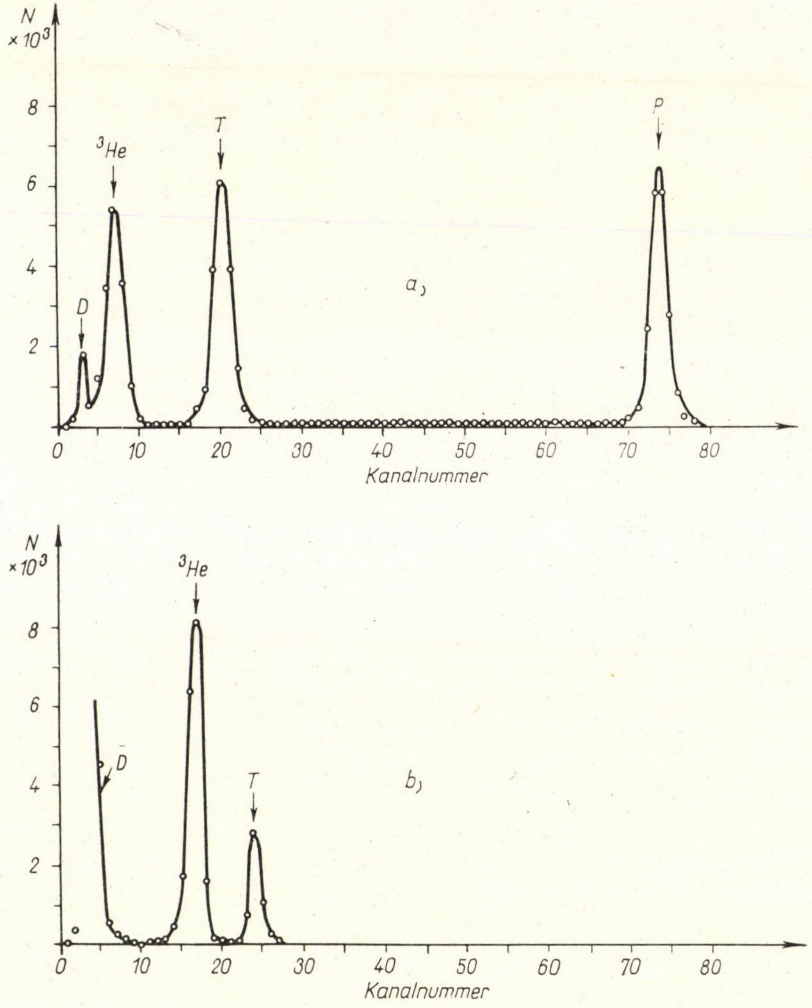

Abb. 4. Impulshöhenspektrum der geladenen Teilchen der DD-Reaktionen *a)* mit der Anordnung von Abb. 1 bei $E_d = 140$ keV aufgenommen; *b)* mit der Anordnung von Abb. 2 bei $E_d = 190$ keV aufgenommen

3. Experimentelle Ergebnisse

Abb. 4a zeigt ein typisches Impulshöhenspektrum der geladenen Teilchen der DD-Reaktion, das bei 140 keV Deuteronenenergie hinter einer Al-Schutzfolie vom Halbleiterdetektor erhalten wurde.

Abb. 4b gibt das gleiche Spektrum bei $E_d = 190$ keV wieder, das mit dem Halbleiterdetektor (∅ 5 mm) nach dem Ablenksystem aufgenommen wurde. Bei den niedrigsten Energien erscheinen die gestreuten Deuteronen, dann folgen mit steigender Energie die ^{3}He-Teilchen, sowie die Protonen und Tritonen aus der D(d, p)T-Reaktion. Die einzelnen Gruppen treten in beiden Spektren ausgezeichnet, voneinander getrennt auf, wobei die Trennung im Falle der Anwendung des Ablenksystems selbstverständlich die vollkommenere ist. In Abb. 4b fehlen die Protonen der D(d, p)T-Reaktion, da diese auf Grund ihrer hohen Energie kaum abgelenkt wurden und so den Detektor nicht »sahen«. Die Tritonen wurden ausserdem nur noch in geringem Masse in den Detektor gelenkt oder gestreut. In beiden Fällen gelangten jedoch noch elastisch gestreute Deuteronen in den Detektor, doch war ihre Zahl nicht bedeutend. Beim Vergleich der beiden Spektren muss beachtet werden, dass die Energiewerte der einzelnen Teilchengruppen nicht übereinstimmen, da diese einmal bei verschiedenen Beschleunigungsspannungen aufgenommen wurden und zum anderen die Anwendung der Al-Folie (Abb. 4a) zur Verschiebung des Energiespektrums führte.

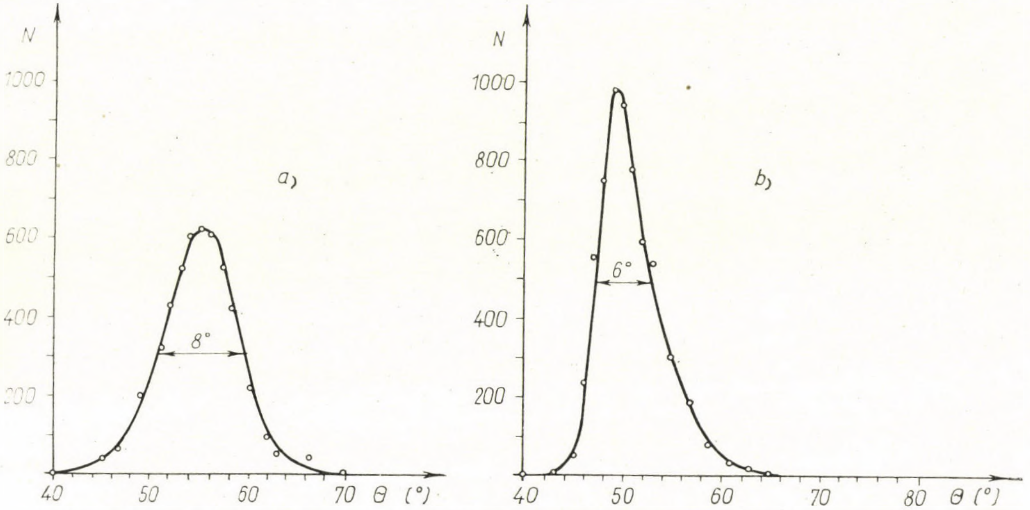

Abb. 5. Die Winkelverteilung der DD-Neutronen bei Anwendung der a. p. m.-Systeme. *a)* gemessen mit der Anordnung von Abb. 1 bei $E_d = 140$ keV; *b)* gemessen mit der Anordnung von Abb. 2 bei $E_d = 190$ keV.

Abb. 5 zeigt die Neutronenstrahlprofile für die beiden verwendeten Messanordnungen. Hierbei wurde die Zahl der Neutron—^{3}He-Koinzidenzen für eine bestimmte Monitorzahl in Abhängigkeit vom Winkel des Neutronendetektors zum beschleunigten Deuteronenstrahl um das Target als Mittelpunkt gemessen. Die Entfernung Target — Neutronendetektor betrug 40 cm. Die Beschleunigungsspannung betrug in dem einen Fall 140 keV (Abb. 5a), im anderen 190 keV (Abb. 5b).

Die Form des Neutronenprofils hängt einmal von dem durch den ^{3}He-Detektor erfassten Raumwinkel und zum anderen von der Breite der Reaktionsenergie der Deuteronen ab. Bei einem dicken Target ergibt sich durch die Abbremsung der Deuteronen im Target eine kontinuierliche Reaktionsenergie von Null bis $E_{d\max}$ was, wie von Bell et al. [3] und Monier et al. [1] gezeigt wird, zu einem asymmetrischen Neutronenprofil führt. Die auf Abb. 5b infolge der besseren Auflösung deutlicher auftretende Asymmetrie, ist ebenfalls der Verwendung eines dicken Targets zuzuschreiben.

Die beschriebenen Messanordnungen haben wir für die Messung der Polarisation der DD-Neutronen im Energiebereich 100 keV $< E_d <$ 250 keV angewendet.

Wir danken dem Direktor des Institutes, Herrn Prof. A. Szalay, für die ausgezeichneten Arbeitsbedingungen und das Interesse, das er unserer Arbeit entgegenbrachte, Herrn Dr. Gy. Csikai für die Anregung zu dieser Arbeit und für seine nützlichen Ratschläge, sowie den wissenschaftlichen Mitarbeitern I. Hunyadi und G. Pető für die während der Bearbeitung des Themas dargebrachte Hilfe, sowie für ihre nützlichen Diskussionen.

LITERATUR

1. L. F. C. Monier, G. E. Tripard u. B. L. White, Nucl. Instr. and Meth., **45**, 282 1966.
2. L. W. Put, C. Bot, W. J. Coenders, J. W. Koene u. J. Blok, Physica, **32**, 1397, 1966.
3. R. A. I. Bell, N. G. Chapman u. P. B. Johnson, Nucl. Instr. and Meth., **33**, 13, 1965.
4. W. R. Dixon u. J. H. Aitken, Nucl. Phys., **24**, 456, 1961.
5. P. L. Okhuysen, E. W. Bennett, J. B. Ashe, u. W. E. Millett, Rev. Sci. Instr., **29**, 982, 1958.
6. W. Franzen, P. Huber u. L. Schellenberg, Helv. Phys. Acta, **28**, 328, 1955.
7. А. П. Бордуля, С. П. Ситько, В. И. Стрижак, ПТЭ, **№ 3**, 48, 1965).
8. T. H. Hsu u. J. M. Robson, Nucl. Instr. and Meth., **39**, 8, 1966.
9. M. Oria u. A. Sorriaux, Rapport CEA-R2779 (1965).
10. P. Bornemisza-Pauspertl, ATOMKI Közlemények, **VII**, 47, 1965.
11. R. J. Blin-Stoyle, Proc. Phys. Soc., **A64**, 700, 1951.

ЭКСПЕРИМЕНТАЛЬНЫЙ ПРИЕМ ДЛЯ ИЗМЕРЕНИЯ ПОЛЯРИЗАЦИИ, ОСНОВАННЫЙ НА ПРИНЦИПЕ a. p. m.

ХАРАЛЬД ПРАДЭ и ДЬЁРДЬ МАТЭ

Резюме

В работе описываются два измерительных приема, которые базируются на принципе a. p. m. (метод связанных частиц) и разрабатываются для измерения поляризации. Через проявление в совпадении нейтронов и ^{3}He-частиц реакции D (d, n)^{3}He имеется возможность получить нейтронный поток вполне определенной энергии и направления. Кроме этого, регистрацией ^{3}He-частиц обеспечивается безупречный контроль выхода нейтронов.

Acta Physica Academiae Scientiarum Hungaricae, Tomus 25 (1), pp. 91—97 (1968)

APPLICABILITY OF THE STATISTICAL MODEL FOR EXPLAINING THE RATIO OF THE (n,n') TO $(n,2n)$ CROSS SECTIONS

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(Presented by A. Szalay. — Received 19. XII. 1967)

The cross sections for the reactions $Tl^{203}(n, 2n)Tl^{202}$, $Hg^{204}(n, 2n)Hg^{203}$, $Pb^{204}(n, 2n)Pb^{203m}$ $Y^{89}(n, n')Y^{89m}$, $Zr^{91}(n, 2n) + Zr^{90}(n, n')Zr^{90m}$, $Au^{197}(n, n')Au^{197m}$, and $Pb^{208}(n, 2n) + Pb^{207}(n, n')Pb^{207m}$ have been measured at neutron energy $15 \pm 0{,}3$ MeV, the results are 1680 ± 210, 2230 ± 300, 860 ± 180, 594 ± 130, 820 ± 200, 280 ± 64, and 1340 ± 174 mb, respectively. The cross section ratios of (n, n') to $(n, 2n)$ reactions are considerably higher than the values predicted by the statistical model. Features of $(n, 2n)$ reactions are in agreement with evaporation mechanism, so the most probable explanation of these disagreements is the deviation of the actual spectrum from the evaporation spectrum supposed in the calculations. According to the present results, the competition of inelastic scattering can be important in the case of the heaviest elements too, so the WEISSKOPF assumptions are not applicable for nuclei with relatively low neutron excess.

For the interpretation of the N—Z dependence of $(n, 2n)$ reactions it is necessary to investigate the mechanism of the competing (n, n') processes.

1. Inelastic scattering

In the bombarding neutron energy region 5—15 MeV the inelastic scattering leading to highly excited states follows evaporation mechanism [1—5, 41]. Only few data are available for the spectrum of the scattered neutrons belonging to the low lying excited levels. According to the measurements of COON [6] in the case of 14 MeV bombarding energy the scattered neutrons show an increasingly forward peaked angular distribution with increasing mass number. The values of cross sections given for scattered neutrons of 9—14 MeV in the angular region 40—180° are small but because of the poor energy resolution and the lack of data in the angular region 0—40° it is not possible to draw a definite conclusion for the total cross section of the direct effect. (By "direct effect" we mean processes which are beyond the framework of the statistical model.) In the case of zirconium the direct effect was estimated to be 20% by AHN [7] which was attributed to the effect of the magic number of neutrons in Zr^{90}. According to the measurements on Bi [8], Ta, W, Pb and Th [9] the excitation cross sections of the low lying levels are considerable even in the case of the heaviest elements.

The measurements of **Perkin** [10] as well as **Scherrer** et al. [11] show that the gamma-spectrum produced by fast-neutron scattering differs considerably from the spectrum shape calculated on the basis of the statistical model. **Clarke** and **Cross** [12] and **Stelson** et al. [13] have proved that the low lying collective levels are strongly excited by the scattering of 14 MeV neutrons in the whole mass number region. **Towle** and **Owens** [14] came to a similar conclusion for the neutron energy 7 MeV. The deformation parameters obtained are in good agreement with those obtained from charged particle scattering.

Bonazzola [15] has demonstrated the core-excitation of $S^{32}(P^{31})$ and $Al^{27}(Si^{28})$ nuclei in (n, n') processes which was obtained previously in proton scattering [16].

These data support the assumption that direct processes play an important role in fast-neutron scattering but they are insufficient for estimating the total cross section of direct effects.

2. (n, 2n) reactions

Barr et al. [17] observed a tendency of $N-Z/A$ dependence at the ratio σ_{nM}/σ_{ne}, where σ_{nM} is the cross section of the neutron emission calculated from $(n, 2n)$ cross sections on the basis of the statistical model and σ_{ne} is the cross section of nonelastic scattering.The results of **Pearlstein** [18] and **Breunlich** et al. [19] support the observation of these systematic deviations between the theoretical and experimental cross sections as a function of $N-Z/A$ as well as $N-Z$. **Csikai** and one of the authors [21] demonstrated a tendency of $N-Z$ dependence of $(n, 2n)$ cross sections obtained experimentally and they established its extent. To give an explanation of the $N-Z$ dependence they [20, 22, 23] supposed a concurrence of processes which cannot be accounted for by evaporation mechanism.

For the cross section of inelastic scattering a reliable lower limit can be obtained by measuring the activity of isomeric decay of the stable nuclei. The aim of this work is to investigate the applicability of the statistical model, for interpreting the data obtained by the above method in order to make the mechanism of the competing (n, n') processes clearer.

3. Experimental procedures and results

The cross sections were measured by activation method. The bombarding neutrons of 15.0 ± 0.3 MeV were produced by the 300 kV cascade generator of the ATOMKI [34] in $d + t$ reaction. The samples were transported by a pneumatic rabbit tube to the measuring system in the case of short half-lives. Fig. 1

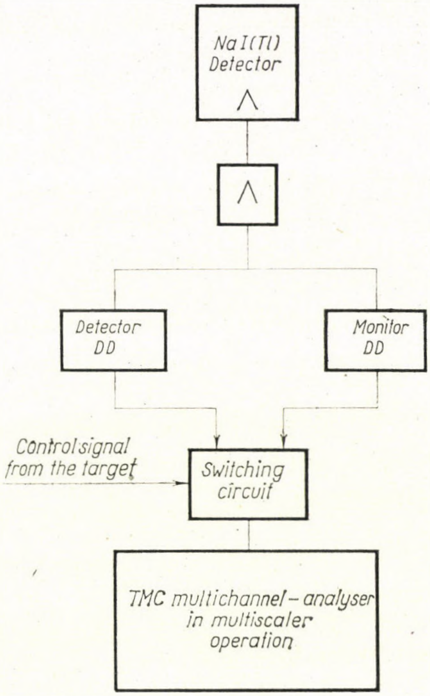

Fig. 1. Block diagram of the measurements

Table 1

Cross sections measured at energy 15.0 ± 0.3 MeV

Target	Reaction	σ(mb)	Monitor reaction	Method
Y_2O_3	$Y^{89}(n,n')Y^{89m}$	594±130	$Pr^{141}(n, 2n)$ 2050±180 mb	γ : 915 keV
Zr	$Zr^{91}(n,2n)Zr^{90m}$ $+Zr^{90}(n,n')Zr^{90m}$	820±200	$Pr^{141}(n, 2n)$ 2050 ± 180 mb	γ: 2.3 + 2.2 MeV
Au	$Au^{197}(n,n')Au^{197m}$	280± 64	$Pr^{141}(n, 2n)$ 2050±180 mb	γ : 279 keV
Pb	$Pb^{207}(n,n')Pb^{207m}$ $+Pb^{208}(n, 2n)Pb^{207m}$	1340±174	$Pr^{141}(n, 2n)$ 2050±180 mb	γ : 570 and 1060 keV
Pb	$Pb^{204}(n, 2n)Pb^{203m}$	860±180	$Pr^{141}(n, 2n)$ 2050±180 mb	γ : 825 keV
$TlNO_3$	$Tl^{203}(n, 2n)Tl^{202}$	1680±210	$Y^{89}(n, 2n)$ 1010±80 mb	γ : 440 keV
$HgCl_2$	$Hg^{204}(n, 2n)Hg^{203}$	2230±300	$Y^{89}(n, 2n)$ 1010±80 mb	γ : 279 keV

shows the block diagram of the measuring device. The neutron flux was monitored by the pulses corresponding to gamma-energies higher than 2 MeV. Details of the "long" irradiation technique and measurement have been described previously [22].

The relative photo peak efficiency of the gamma-spectrometer was determined by using isotopes of known decay scheme (Na^{22}, Na^{24}, Cl^{34}, Sc^{44}, Y^{88}). The data for the decay schemes have been taken from the Nuclear Data Sheets [29]. The obtained data are in good agreement with previously published data excepting for the Y^{89m}. For the half-life of Y^{89m} we have measured 17 ± 0.6 sec. The cross sections obtained are presented in Table 1. The indicated errors contain the error of the cross section of the monitor reactions, the error arising from the gamma-detection and the statistical error.

4. Discussion

According to Fig. 2 if the $N-Z/A$ value is higher than 0.1 it is only the inelastic scattering which could cause a competition for $(n, 2n)$ reactions. The survey in the Section 1 shows that the part of the spectrum belonging to low energy neutrons can be fitted well to the evaporation spectrum shape. The shape of the excitation functions of the $(n, 2n)$ reactions is in agreement practically [17, 20, 23, 30, 35] with the form given by the WEISSKOPF estimation using a nuclear temperature of $T = \sqrt{E_n/0.115\ A}$. Fig. 3 shows the ratios of the theoretically and experimentally obtained $(n\ 2n)$ cross sections at saturation.

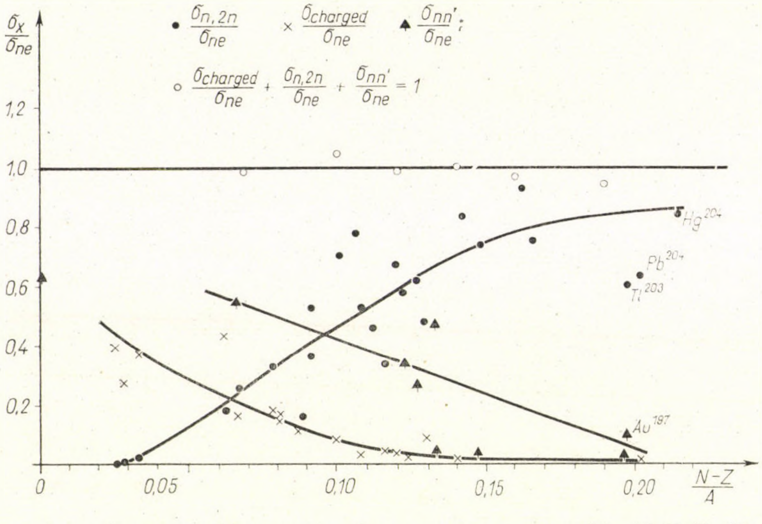

Fig. 2. Comparison of cross sections measured at a neutron energy 14—15 MeV. (Data: from [28, 31, 22, 36—39] and from Tables 1 and 2; σ_{ne}: [40].)

The theoretical values were extrapolated by the **Weisskopf** estimation [32] from the experimental values measured at 14.1 MeV. The agreement is satisfactory in the whole mass number region taking into account the 15—20 % error of the experimental results.

At the same time according to the data of Table 2 the experimental $\sigma_{n\,n'}/\sigma_{n\,2n}$ ratios are considerably higher than the values calculated from the above formula. The deviation is extremely high in the cases of Au^{197} and Rh^{103}. The ratios obtained experimentally could be accounted for only by supposing such a high nuclear temperature as would contradict the experimentally obtained values for it. The most probable explanation of these deviations is that the spectrum of scattered neutrons in the region under the $(n, 2n)$ threshold deviates from the evaporation shape. In Table 2 the ratios are indicated, showing the extent of the supposed direct effect.

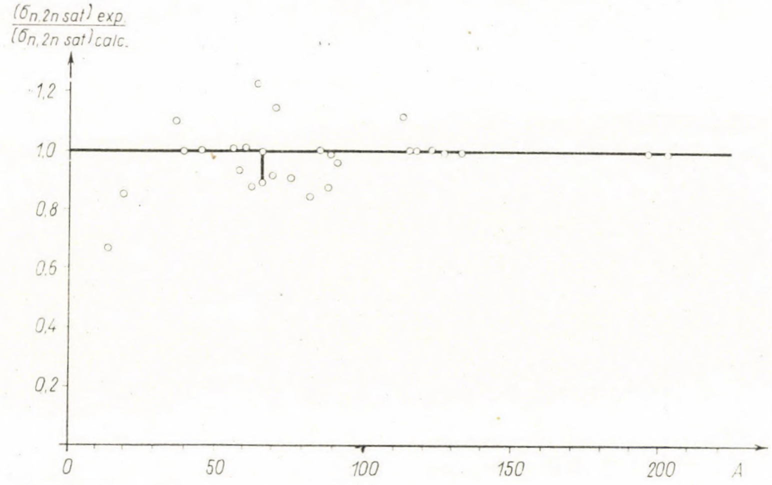

Fig. 3. Comparison of experimental saturation values of $(n, 2n)$ cross-sections with those calculated by the **Weisskopf** estimation, using the experimental data measured at 14,1 MeV, assuming $T = \sqrt{E_n/0{,}115A}$ for nuclear temperature

The competition of inelastic scattering observed previously [21] in the medium heavy mass number region can be significant also in the case of heavy elements according to the present cross section measurements on Au^{197}, Tl^{203}, Pb^{204}.

If neutron spectrum measurements were available over the whole energy region, it would contribute appreciably to our understanding of the above problem.

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Table 2

The experimental lower limits of the $\sigma_{n,n'}/\sigma_{n,2n}$ cross section ratios compared with the values calculated on the basis of the statistical model

Target	$\sigma_{n,n'}$ (mb)	Ref.	$\sigma_{n,2n}$ (mb)	Ref.	$\sigma_{n\,n'}/\sigma_{n,2n'}$ (exp)	$\sigma_{n,n'}/\sigma_{n,2n}$ (calc)	Lower limit of the direct effect%
Fe^{56}	660	(24)	500	(28)	1.32	0.504	23
Br^{79}	266	(25)	1140	(33)	0.233	0.112	9
Y^{89}	594	present measurement	1010	(22)	0.588	0.411	8
Rh^{103}	508	(26)	780	(28)	0.651	0.056	34
In^{115}	81	(27)	1540	(28)	0.053	0.034	1.6
Yb^{176}	16.7	(25)	1810	(28)	0.009	0.001	1
Au^{197}	280	present measurement	2150	(28)	0.130	0.002	10
Pb^{204}	76.5	(22)	1575	(22)	0.048	0.001	3.5

REFERENCES

1. O. A. Salnikov et al., Jadernaja Fizika, **4,** 1154, 1966.
2. A. V. Malyshev et al., Nucl. Phys., **76,** 232, 1966.
3. J. Koralewski et al., Report No 591 (XA) PL (1964).
4. K. R. Graves and L. Rosen, Phys. Rev., **89,** 343, 1953.
5. S. G. Buccino et al., Nucl. Phys., **60,** 17, 1964.
6. J. H. Coon et al., Phys. Rev., **111,** 250, 1958.
7. S. H. Ahn and J. H. Roberts, Phys. Rev., **108,** 110, 1957.
8. L. Rosen and L. Stewart, Phys. Rev., **99,** 1052, 1955.
9. S. Pearlstein and E. J. Winhold, J. Nucl. Energy A/B 19, 497, 1965.
10. J. L. Perkin, Nucl. Phys., **60,** 561, 1964.
11. V. E. Scherrer et al., Phys. Rev., **91,** 497, 1953.
12. R. L. Clarke and W. G. Cross, Nucl. Phys., **53,** 177 1964; Nucl. Phys., **A 95,** 320, 1967.
13. P. H. Stelson, R. L. Robinson, H. J. Kim, J. Rapeport and G. R. Satchler, Nucl. Phys., **68,** 97, 1965.
14. J. H. Towle and R. O. Owens, Nucl. Phys., **A100,** 257, 1967.
15. G. C. Bonazzola et al., Nucl. Phys., **68,** 169, 1965; Nuovo Cimento, (10) **38,** 144, 1965.
16. F. Perey et al., Phys. Letters, **4,** 25, 1963.
17. D. W. Barr, Cl. Browne and J. S. Gilmore, Phys. Rev., **123,** 859, 1961.
18. S. Pearlstein, Nucl. Sci. and Engineering, **23,** 238, 1965.
19. W. Breunlich, P. Hille, B. Karlik and S. Tagessen, Int. Conf. on the Study of Nucl. Structure with Neutrons, Antwerpen, c. 117, 1965.
20. G. Pető, P. Bornemisza-Pauspertl, J. Károlyi, Acta Phys. Hung., **24,** 93, 1968.
21. J. Csikai and G. Pető, Physics Letters, **20,** 52, 1966.
22. J. Csikai and G. Pető, Acta Phys. Hung., **23,** 87, 1967.
23. G. Pető, Thesis (1967) Debrecen.
24. B. A. Benetsky and I. M. Frank, Jadernaja Fizika, **3,** 861, 1966.
25. P. Bornemisza-Pauspertl and P. Hille, Sitz. Ber. Österr. Akad. Wiss. 1967 (in print).
26. W. Nagel and A. H. W. Aten, J. Nucl. Energy, **20,** 475 1966.
27. W. Nagel and A. H. W. Aten, Physica, **31,** 1091, 1965.
28. Neutron Cross Sections, BNL-325, Second Edition Vol. 1—3. 1964—1966.
29. Nuclear Data Sheets, Nat. Acad. Sci., Nat. Res. Council, Washington, 1961.
30. J. Csikai, Int. Conf. on the Study of Nucl. Structure with Neutrons, Antwerpen, c. 102, 1965.

31. P. Jessen et al., Fast Neutron Cross Sections, Univ. Hamburg, 1965.
32. J. M. Blatt and V. I. Weisskopf, Theoretical Nucl. Phys., Wiley, New York, 1952.
33. B. Grimeland et al., Phys. Rev., **137**, 8878, 1965.
34. P. Bornemisza-Pauspertl, ATOMKI Közl., **7**, 47, 1965.
35. J. Csikai, private communication.
36. N. W. Glochert et al. Nucl. Phys., **73**, 349, 1965.
37. S. Tagessen und P. Hille, Acta Phys. Austriaca, 23, 31, 1966.
38. B. A. Benetsky and I. M. Frank, Jadernaja Fizika, 3, 861, 1966.
39. K. G. Broadhead et al., Phys. Rev., **139B**, 1525, 1965.
40. N. N. Flerov and V. M. Talysin, J. Nucl. Energy, **4**, 529, 1957.
41. M. Maruyama et al., Int. Conf. on the Study of Nucl. Structure, Tokyo, 1967.

ПРИМЕНИМОСТЬ СТАТИСТИЧЕСКОЙ МОДЕЛИ ДЛЯ ОБЪЯСНЕНИЯ ОТНОШЕНИЯ ПОПЕРЕЧНОГО СЕЧЕНИЯ (n,n') К СЕЧЕНИЮ $(n,2n)$

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Резюме

Измерялись поперечные сечения для реакций Tl^{203} $(n, 2n)$ Tl^{202}, Hg^{204} $(n, 2n)$ Hg^{203}, Pb^{204} $(n, 2n)$ Pb^{203m} У^{89} (n, n') У^{89m}, Zr^{91} $(n, 2n) +$ Zr^{90} (n, n') Zr^{90m}, Au^{117} (n, n') Au^{197m} и Pb^{208} $(n, 2n) +$ Pb^{207} (n, n') P^{207m} при нейтронной энергии 15 ± 0,3 Мэв, для чего получены значения 1680 ± 210, 2230 ± 300, 860 ± 180, 594 ± 130, 820 ± 200, 280 ± 64 и 1340 ± 174 мб соответственно. Отношение поперечных сечений реакций (n, n') и $(n, 2n)$ значительно больше полученного на основе статистической модели. Характер реакций $(n, 2n)$ согласуется с механизмом испарения, таким образом более надежное объяснение данного несоответствия заключается в расхождении действительного спектра от спектра испарения, предположенного при вычислениях.

По отношению данных результатов можно установить, что конкуренция неупругого рассеяния может быть значительной и в случае более тяжелых элементов, и так предположения Вейскопфа неприменимы для ядер с относительно небольшим избытком нейтронов

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THE HYDRODYNAMICAL MODEL OF WAVE MECHANICS IV

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Continuing the works [1], [2], [3] the hydrodynamical model of wave mechanics will be extended to the hydrodynamical model of wave equation including spin-orbit coupling. Instead of the wave function hydrodynamical variables will be introduced and it will be shown that there is a one-to-one correspondence between the wave function and the hydrodynamical variables satisfying certain reasonable physical requirements. The equation of motion for these new variables will be derived in a subsequent paper.

1. Introduction

In the previous papers ([1], [2], [3]) it has been shown that both the Schrödinger and the Pauli equation containing the external electromagnetic field can be transformed into equations of hydrodynamical variables and there is a one-to-one correspondence between the hydrodynamical variables and the wave function. In the present paper we begin to establish the hydrodynamical model of the Pauli equation completed with the spin-orbit coupling term.

Let us first summarize the principles followed in developing the hydrodynamical model. Assuming that the wave equation for the wave function ψ is given, the hydrodynamical variables and their equation of motion will be required to satisfy the following reasonable requirements:

1) The wave function and the hydrodynamical variables must be able to be calculated from each other unambiguously at any time. In this mapping the wave function and the hydrodynamical variables will be called corresponding quantities.

2) If the values of the wave function and that of the complete set of corresponding hydrodynamical variables at a time $t = 0$ are given as $\psi(0)$ and $D(0)$, then the values of these quantities at time t [the values of $\psi(t)$ and $D(t)$] can be calculated from the wave equation and from the hydrodynamical equations, respectively. These $\psi(t)$ and $D(t)$ are required to be corresponding quantities in the sense of the above requirement 1.

3) Applying symmetry transformations for the hydrodynamical variables they have to transform like the corresponding classical quantities. (Thus, for instance, the hydrodynamical variable describing the velocity of the medium should transform as a vector and has to be gauge invariant, a.s.o.)

4) *a)* By integrating the equations of motion of the hydrodynamical variables over the whole space terms that can be interpreted in accordance with classical physics should be obtained.

b) The hydrodynamical variables must be equal to the experimentally obtained values of the quantities represented by them.

Let us complete the above requirements with the following: It is evident that hydrodynamical variables may be constructed from ψ and ψ^+ in various ways, provided the variables introduced satisfy requirements 1, 2 and 3. The restriction basically important from the point of view of physics is implied in 4*a)* and *b)*. The hydrodynamical model of the Pauli equation shows that even provided the conditions of 1, 2, 3 and 4*a)* are fulfilled there remains in the hydrodynamical quantities a certain arbitrariness which is sufficient — and also necessary — for satisfying 4*b)* and thus for describing correctly the experimental results.

It should be emphasized, however, that the existence of hydrodynamical variables satifying the above four requirements is by no means evident. In the present paper we shall show how hydrodynamical variables satisfying condition 1 can be associated with the wave function ψ obeying the wave equation in which the spin-orbit coupling term is included. The equations of motion and the fulfilment of conditions 2, 3 and 4 will be treated in a subsequent paper.

2. Wave equation

Consider a particle with charge e, mass m and magnetic moment $\boldsymbol{m}$, moving in an external electromagnetic field described by the field strengths $\boldsymbol{E}$ and $\boldsymbol{B}$. The interaction energy of a particle at rest with the electromagnetic field is:

$$\mathscr{E}_1 = -\boldsymbol{m}\cdot\boldsymbol{B}\,. \tag{1}$$

In the case of a moving particle, however, an additional electric dipole moment of the form

$$\boldsymbol{p}_0 = \frac{1}{c}\,\boldsymbol{v}\times\boldsymbol{m} \tag{2}$$

arises which also interacts with the electric field. Thus, in addition to $\mathscr{E}_1$ the following interaction energy appears

$$\mathscr{E}_2 = -\boldsymbol{p}_0\cdot\boldsymbol{E} = -\frac{1}{c}(\boldsymbol{v}\times\boldsymbol{m})\,\boldsymbol{E}\,. \tag{3}$$

Besides electromagnetic interaction, as a result of the Thomas precession a further interaction occurs. Let the particle move in a system of reference S_1

at time t with velocity $\boldsymbol{v}$ and at time $t + \Delta t$ with velocity $\boldsymbol{v} + \Delta \boldsymbol{v}$. Let S_2 and S_3 be the rest system of the particle at times t and $t + \Delta t$. Even if the Lorentz transformations from system S_1 to S_2 and similarly from S_1 to S_3 do not contain any rotation, the transformation from S_2 to S_3 generally does. Thus, the angular momentum of the particle must rotate by a certain angle while it moves from the system S_2 to S_3. The angular velocity of this precession has been given by THOMAS as (see e.g. [4])

$$\omega_0 \approx \frac{1}{2\,c^2}\,\boldsymbol{v} \times \frac{d\boldsymbol{v}}{dt} \approx \frac{e}{2\,mc^2}\,\boldsymbol{v} \times \boldsymbol{E}\,. \tag{4}$$

If the internal angular momentum of the particle is $\boldsymbol{s} = (mc/e)\boldsymbol{m}$ the energy due to the Thomas precession will be

$$\mathscr{E}_3 = -\,\boldsymbol{s} \cdot \omega_0 = -\,\frac{1}{2\,c}\,\boldsymbol{m}(\boldsymbol{v} \times \boldsymbol{E})\,. \tag{5}$$

Thus, taking into account (1), (3) and (5) we get for the interaction energy:

$$\mathscr{E}_1 + \mathscr{E}_2 + \mathscr{E}_3 = -\,\boldsymbol{m} \cdot \boldsymbol{B} + \frac{1}{4\,c}\,\boldsymbol{m}(\boldsymbol{v} \times \boldsymbol{E} - \boldsymbol{E} \times \boldsymbol{v})\,. \tag{6}$$

Here the last term we have written in a symmetrical form in order to get a Hermitian Hamiltonian in the corresponding quantum mechanical expression (see [9]). Adding to (6) the term

$$\mathscr{E}_0 = \frac{1}{2}\,mv^2 + e\Phi$$

containing the kinetic and the electrostatic energy, we arrive at the following expression of the energy:

$$\mathscr{E} = \frac{1}{2}\,mv^2 + e\Phi - \boldsymbol{m}\boldsymbol{B} + \frac{1}{4\,c}\,\boldsymbol{m}(\boldsymbol{v} \times \boldsymbol{E} - \boldsymbol{E} \times \boldsymbol{v})\,.$$

The transition to the quantum mechanical Hamiltonian can be performed by substituting

$$\boldsymbol{m} = \frac{e\hbar}{2\,mc}\,\boldsymbol{\sigma}\,, \quad \boldsymbol{v} = \frac{1}{m}\left(-i\hbar\,\mathrm{grad} - \frac{e}{c}\,\boldsymbol{A}\right), \tag{7}$$

where $\boldsymbol{\sigma}$ is the vector built up from the Pauli matrices [3] and $\boldsymbol{A}$ is the vector potential of the electromagnetic field. Thus, the wave equation will have the form

$$\mathscr{H}\psi = i\hbar\frac{\partial\psi}{\partial t}\,, \tag{8}$$

with

$$\psi = \begin{pmatrix}\psi_1\\ \psi_2\end{pmatrix}$$

and

$$\begin{aligned}\mathscr{H} = \frac{1}{2m}\left(-i\hbar\,\mathrm{grad} - \frac{e}{c}\boldsymbol{A}\right)^2 + e\Phi - \mu\,(\boldsymbol{\sigma}\cdot\boldsymbol{B}) + \\ + \lambda\boldsymbol{\sigma}\left[\left(-\,i\hbar\,\mathrm{grad} - \frac{e}{c}\boldsymbol{A}\right)\times\boldsymbol{E} - \boldsymbol{E}\times\left(-\,i\hbar\,\mathrm{grad} - \frac{e}{c}\boldsymbol{A}\right)\right].\end{aligned} \tag{9}$$

Here we introduced the constants:

$$\mu = \frac{e\hbar}{2\,mc}\,, \qquad \lambda = \frac{e\hbar}{8\,m^2c^2}\;.$$

The last term of the Hamiltonian arising from $\mathscr{E}_3$ describes the spin-orbit coupling, while the first three terms are the Hamiltonian of the Pauli equation. It is to be noted that the expression obtained from the Dirac equation for spin-orbit coupling is identical with the form derived above [5].

3. Mass density and density of momentum

In order to account for the mass density and for the density of momentum we have to formulate the continuity equation. Multiplying (8) from the left by $\psi^+ = (\psi_1^*\ \psi_2^*)$ and subtracting from it the conjugated equation multiplied from the right by ψ, we arrive at the equation

$$\frac{\partial\varrho_m'}{\partial t} + \mathrm{div}\,\varrho_m'\boldsymbol{v}' = 0\,, \tag{10}$$

where

$$\varrho_m' = m\,\psi^+\psi\,, \tag{11}$$

and

$$\varrho_m'\boldsymbol{v}' = \frac{\hbar}{2\,i}\left(\psi^+\nabla\psi - (\nabla\psi)^+\psi\right) - \frac{e}{c}\psi^+\boldsymbol{A}\psi - 2\,m\lambda\,(\psi^+\boldsymbol{\sigma}\psi)\times\boldsymbol{E}. \tag{12}$$

From the above continuity equation, however, ϱ_m' and $\varrho_m'\boldsymbol{v}'$ cannot be determined uniquely. Various, non-equivalent distributions of mass density and of momentum density can be given satisfying the continuity equation.

In order to examine this ambiguity let us rewrite equation (10) in the form

$$\sum_{\nu=0}^{3} \frac{\partial j'_\nu}{\partial x_\nu} = 0\,, \tag{13}$$

where

$$j'_0 = \varrho'_m = \varrho'_m\,, \quad \boldsymbol{j}' = \varrho'_m \boldsymbol{v}'\,, \quad \text{and} \quad x_0 = t\,.$$

Let $k_{\nu\mu}(\boldsymbol{r}, t)$ be an arbitrary screw-symmetrical four by four matrix depending on time and coordinates and let us form the vector

$$j''_\nu = \sum_{\mu=0}^{3} \frac{\partial k_{\nu\mu}}{\partial x_{\nu\mu}}\,. \tag{14}$$

As $k_{\nu\mu}$ is a screw-symmetrical matrix it is easy to see that if j'_ν satisfies the continuity equation (13), then

$$j_\nu = j'_\nu + j''_\nu \tag{15}$$

satisfies it too.

It can be shown that the form (14) of j''_ν is the most general expression satisfying (13) (See [6]).

Let us represent the six independent components of the matrix $k_{\nu\mu}$ by two arbitrarily chosen vectors $\boldsymbol{a}$ and $\boldsymbol{b}$, i.e. $k_{\nu\mu}$ be

$$k_{\nu\mu} = \begin{pmatrix} 0 & -a_1 & -a_2 & -a_3 \\ a_1 & 0 & b_3 & -b_2 \\ a_2 & -b_3 & 0 & b_1 \\ a_3 & b_2 & -b_1 & 0 \end{pmatrix}, \tag{16}$$

where the components of $\boldsymbol{a}$ and $\boldsymbol{b}$ are arbitrary functions of time and coordinates. Substituting (16) into (14) and singling out the components $\nu = 0$ and $\nu = 1, 2, 3$, we have

$$j'_0 \equiv \varrho''_m = -\operatorname{div} \boldsymbol{a} \tag{17}$$

and

$$\boldsymbol{j}'' \equiv \varrho''_m \boldsymbol{v}'' = \operatorname{rot} \boldsymbol{b} + \frac{\partial \boldsymbol{a}}{\partial t}\,. \tag{18}$$

Consequently, if instead of ϱ' and $\varrho'\boldsymbol{v}'$ we introduce the mass density ϱ_m and the density of momentum $\varrho_m \boldsymbol{v}$

$$\varrho_m = \varrho'_m - \operatorname{div} \boldsymbol{a}\,, \tag{19}$$

$$\varrho_m \boldsymbol{v} = \varrho'_m \boldsymbol{v}' + \operatorname{rot} \boldsymbol{b} + \frac{\partial \boldsymbol{a}}{\partial t} \tag{20}$$

these new quantities also satisfy the continuity equation.

The actual values of $\boldsymbol{a}$ and $\boldsymbol{b}$ will be determined by the condition that the value of the mass density and that of the density of momentum have to fulfil requirements 4*a*) and 4*b*) given in Chapter 1. Accordingly in the case of the Pauli equation we had to use (see [3])

$$\boldsymbol{a} = 0\,, \quad \boldsymbol{b} = \frac{\hbar}{4} \psi^+ \boldsymbol{\sigma} \psi . \tag{21}$$

These values can be used — as will be justified later — also in the problem of spin-orbit coupling, i.e. the hydrodynamical variables obtained using (21), as well as the related equations of motion satisfy restrictions 1—4.

Thus, using (11), (12) and (21) we get for (19) and (20)

$$\varrho_m \equiv m\varrho = m\psi^+ \psi\,, \tag{22}$$

$$\boldsymbol{v} = \frac{1}{\varrho} \frac{\hbar}{2mi} (\psi^+ \nabla\psi - (\nabla\psi)^+ \psi) - \frac{e}{mc} \boldsymbol{A} + \frac{1}{\varrho} \frac{\hbar}{4m} \operatorname{rot} \boldsymbol{s} - \frac{2\lambda}{\varrho} \boldsymbol{s} \times \boldsymbol{E}\,, \tag{23}$$

where the notations $\varrho = \psi^+\psi$ and $\boldsymbol{s} = \psi^+\boldsymbol{\sigma}\psi$ have been used.

4. Effective charge density and density of effective current

Since in the expression of mass density and that of momentum density (22) and (23) $\boldsymbol{a} = 0$ has been chosen, the mathematical possibility given by (19) and (20) has been only partially exploited. However, in the expression of the effective charge density and in that of the density of the effective current there will appear — as will be seen — the divergence and the time derivative of a vector.

Let us consider now a medium of electric charge density $\varrho_e(\boldsymbol{r}, t)$, velocity $\boldsymbol{v}(\boldsymbol{r},t)$, magnetic moment density $\boldsymbol{M}(\boldsymbol{r}, t)$ and electric moment density $\boldsymbol{P}(\boldsymbol{r},t)$. The charge and the current producing the same electromagnetic field as the above magnetically and electrically polarized charged medium will be called effective charge and effective current, respectively (see [7])

$$\varrho_{\text{eff}} = \varrho_e - \operatorname{div} \boldsymbol{P}\,, \tag{24}$$

$$\boldsymbol{j}_{\text{eff}} = \varrho_e \boldsymbol{v} + c \operatorname{rot} \boldsymbol{M} + \frac{\partial \boldsymbol{P}}{\partial t}\,. \tag{25}$$

The quantities ϱ_e and $\varrho_e\boldsymbol{v}$, i.e. the charge density and the current density can be obtained from (22) and (23) as

$$\varrho_e = e\psi^+ \psi = e\varrho\,, \tag{26}$$

$$\varrho_e \boldsymbol{v} = \frac{e\hbar}{2\,mi}\left(\psi^+ \nabla\psi - (\nabla\psi)^+ \psi\right) - \frac{e}{mc}\,\varrho_e \boldsymbol{A} + c\,\frac{\mu}{2}\,\mathrm{rot}\,\boldsymbol{s} - 2\,e\lambda(\boldsymbol{s}\times\boldsymbol{E})\,. \tag{27}$$

Taking into account the result of the Einstein—deHaas experiment and the fact that in the ground state of the hydrogen atom the value of the magnetic moment of the electron is μ, in the case of the Pauli equation a permanent magnetic moment density

$$\boldsymbol{M} = \frac{\mu}{2}\,\boldsymbol{s} \tag{28}$$

has to be assumed. This value of $\boldsymbol{M}$ is also valid for the Pauli equation when completed with the spin-orbit coupling. In this case, however, the electric moment density $\boldsymbol{P}$ has to be also taken into account, as a moving magnetic dipole density corresponds to an electric dipole distribution. The expression of $\boldsymbol{P}$ can be obtained with the aid of equation (6), writing it in the form

$$-\,\boldsymbol{mB} + \frac{1}{4\,c}\,\boldsymbol{m}\,[\boldsymbol{v}\times\boldsymbol{E} - \boldsymbol{E}\times\boldsymbol{v}] = -\,\boldsymbol{mB} - \boldsymbol{pE}\,, \tag{29}$$

where

$$\boldsymbol{p} = \frac{1}{4\,c}\,(\boldsymbol{v}\times\boldsymbol{m} - \boldsymbol{m}\times\boldsymbol{v})\,. \tag{30}$$

The right side of (29) describes the interaction of the magnetic moment with the magnetic field and similarly the interaction of the electric moment with the electric field. Using (7) we get from (30) for the electric moment density

$$\boldsymbol{p} = \lambda\psi^+\left[\left(-\,i\hbar\nabla - \frac{e}{c}\,\boldsymbol{A}\right)\times\boldsymbol{\sigma} - \boldsymbol{\sigma}\times\left(i\hbar\overleftarrow{\nabla} - \frac{e}{c}\,\boldsymbol{A}\right)\right]\psi, \tag{31}$$

where the symbol ∇ acting backwards on ψ^+ is denoted by $\vec{\nabla}$.

Inserting (28) and (31) into (24) and (25) we have for the density of effective charge and of effective current, respectively

$$\varrho_{\mathrm{eff}} = e\,\psi^+\psi - \lambda\,\mathrm{div}\left\{\psi^+\left[\left(-\,i\hbar\nabla - \frac{e}{c}\,\boldsymbol{A}\right)\times\boldsymbol{\sigma} - \boldsymbol{\sigma}\times\left(i\hbar\overleftarrow{\nabla} - \frac{e}{c}\,\boldsymbol{A}\right)\right]\psi\right\}, \tag{32}$$

$$\begin{aligned} \boldsymbol{j}_{\mathrm{eff}} = {} & \frac{e\hbar}{2\,mi}\left(\psi^+ \nabla\psi - (\nabla\psi)^+\psi\right) - \frac{e^2}{mc}\,\psi^+\psi\boldsymbol{A} + c\mu\,\mathrm{rot}\,\boldsymbol{s} - 2\,e\lambda\boldsymbol{s}\times\boldsymbol{E} + \\ & + \lambda\,\frac{\partial}{\partial t}\left\{\psi^+\Big[\left(-\,i\hbar\nabla - \frac{e}{c}\,\boldsymbol{A}\right)\times\boldsymbol{\sigma} - \boldsymbol{\sigma}\times\left(i\hbar\overleftarrow{\nabla} - \frac{e}{c}\,\boldsymbol{A}\right)\psi\right\}. \end{aligned} \tag{33}$$

It is to be noted that as we have shown in [6], the expression of ϱ_{eff} and $\boldsymbol{j}_{\mathrm{eff}}$ given in (32) and (33) can be obtained from the Hamiltonian (9) directly.

5. Hydrodynamical variables

The investigation of the density of momentum and of the effective current density has shown that the electron can be represented by a flowing charged medium having a magnetic polarization and an electric polarization arising as a result of motion. Thus, for the hydrodynamical variables the following quantities will be used:

a) The mass density of the medium

$$\varrho_m(\boldsymbol{v}, t) = m\,\psi^+\psi\,. \tag{34}$$

It is to be noted that if the physical problem requires the quantities

$$\varrho \equiv \psi^+\,\psi \quad \text{and} \quad \varrho_e \equiv e\psi^+\,\psi$$

can be used instead of (34).

b) The velocity distribution of the medium

$$\begin{aligned}\boldsymbol{v}(\boldsymbol{r}, t) = \frac{1}{\varrho}\,\frac{\hbar}{2\,mi}\,(\psi^+\,\nabla\psi - (\nabla\psi)^+\,\psi) - \frac{e}{mc}\,\boldsymbol{A} + \\ + \frac{1}{\varrho}\,\frac{\hbar}{4\,m}\,\mathrm{rot}\,(\psi^+\,\boldsymbol{\sigma}\psi) - \frac{2\,\lambda}{\varrho}\,\psi^+\,\boldsymbol{\sigma}\,\psi\times\boldsymbol{E}\,.\end{aligned} \tag{35}$$

c) The vector

$$\boldsymbol{s}(\boldsymbol{r}, t) = \psi^+\,\boldsymbol{\sigma}\psi \tag{36}$$

characterizing the magnetic moment distribution of the medium. It seems to be convenient to introduce besides $\boldsymbol{s}$ a unit vector

$$\boldsymbol{T}(\boldsymbol{r}, t) = \frac{1}{\varrho}\,\boldsymbol{s} \tag{37}$$

pointing in the direction of the magnetic polarization [3].

d) The vector

$$\boldsymbol{P}(\boldsymbol{r}, t) = \lambda\psi^+\left[\left(-\,i\hbar\,\nabla - \frac{e}{c}\,\boldsymbol{A}\right)\times\boldsymbol{\sigma} - \boldsymbol{\sigma}\times\left(i\hbar\,\overleftarrow{\nabla} - \frac{e}{c}\,\boldsymbol{A}\right)\right]\psi \tag{38}$$

representing the density of the electric polarization.

The vector

$$R(r,t)=\frac{1}{\varrho}P(r,t) \tag{39}$$

also may be used instead of P.

It is to be noted that P is not independent of the other hydrodynamical variables, i.e. it can be expressed by means of ϱ, v and T. However, in order to fulfil requirement 4*a)* P has to be treated as an independent variable.

6. Connection between hydrodynamical variables and the wave function

It will be shown that at any time t there exist essentially unambiguous relations between the components of the wave function and the variables ϱ, v, T, P introduced above, i.e. the wave function and the corresponding hydrodynamical quantities fulfil requirement 1.

It is obvious that with the help of the equations (34), (35), (36) and (38) the hydrodynamical variables can be obtained uniquely from a given wave function satisfying the wave equation (8) at some fixed value of the potentials A and Φ. However, the reverse problem requires a more detailed discussion: we have to consider (following the procedure developed in [3]) whether it is possible to associate the values of ψ_1 and ψ_2 unambiguously with the given values of hydrodynamical variables at fixed values of the potentials A and Φ. (The influence of possible gauge transformations on the relation between the hydrodynamical variables and the wave functions will be dealt with in a subsequent paper.)

In order to simplify the investigation of the relation between the hydrodynamical variables and the wave function let us represent the two complex components of ψ by four real functions of time t and coordinates r as

$$\psi_1=R\cos\frac{\vartheta}{2}\,e^{i\left(S-\frac{1}{2}\varphi\right)},$$

$$\psi_2=R\sin\frac{\vartheta}{2}\,e^{i\left(S+\frac{1}{2}\varphi\right)}.$$

We shall investigate now what kind of relation can be found between R, S, ϑ, φ and the hydrodynamical variables.

Inserting the above form of ψ into (37) we get for the components of T:

$$T_1=\sin\vartheta\cos\varphi\,,\quad T_2=\sin\vartheta\sin\varphi\,,\quad T_3=\cos\vartheta\,.$$

From these we have for ϑ and φ:

$$\vartheta = -\frac{i}{2} \ln \frac{\dfrac{i T_3}{\sqrt{T_1^2 + T_2^2}} - 1}{\dfrac{i T_3}{\sqrt{T_1^2 + T_2^2}} + 1}$$

and

$$\varphi = -i \ln \frac{T_1 + T_2}{\sqrt{T_1^2 + T_2^2}} ,$$

where the positive sign of the square roots has to be taken.

In order to express the velocity of flow in terms of the scalar functions R, S, ϑ, φ we substitute the above form of ψ_1 and ψ_2 into (35) and find

$$\boldsymbol{v} = \frac{\hbar}{m} \operatorname{grad} S - \frac{\hbar}{2m} \cos \operatorname{grad} \varphi - \frac{e}{mc} \boldsymbol{A} + \frac{\hbar}{4m} \frac{1}{\varrho} \operatorname{rot} \boldsymbol{s} - 2 \lambda \boldsymbol{T} \times \boldsymbol{E} .$$

Integrating both sides along a line connecting the coordinate vector $\boldsymbol{r}_0$ of an arbitrary point with $\boldsymbol{r}$ in a way not crossing singular points, we get

$$S = \frac{m}{\hbar} \int_{\boldsymbol{r}_0}^{\boldsymbol{r}} \left[\boldsymbol{v} + \frac{e}{mc} \boldsymbol{A} + \frac{\hbar}{2m} \cos \vartheta \operatorname{grad} \varphi - \right.$$
$$\left. - \frac{\hbar}{4m} \frac{1}{\varrho} \operatorname{rot} \boldsymbol{s} + 2 \lambda \boldsymbol{T} \times \boldsymbol{E} \right] d\boldsymbol{r} + S_0(\boldsymbol{r}_0, t) ,$$

where S_0 is a quantity which depends on the choice of $\boldsymbol{r}_0$ but is independent of the vector $\boldsymbol{r}$.

If the lines of integration from fixed $\boldsymbol{r}_0$ to $\boldsymbol{r}$ avoid singular points, the value of the integral becomes independent of the path of integration, i.e. S is a single-valued function. Whereas, choosing as path of integration a closed curve, surrounding singularities, the integral does not give zero and, thus, a multivalued function of S is obtained. The wave function, however, can even in this case unambiguously be determined from the hydrodynamical variables, i.e. remains a single-valued function even if S is multivalued, provided the values of S in the same point differ by integer multiples of 2π only. From this we get the condition

$$\oint \boldsymbol{v}\, d\boldsymbol{r} = 2 \pi \frac{\hbar}{m} k + \oint \left[- \frac{c}{mc} \boldsymbol{A} - \right.$$
$$\left. - \frac{\hbar}{2m} \cos \vartheta \operatorname{grad} \varphi + \frac{\hbar}{4m} \frac{1}{\varrho} \operatorname{rot} \boldsymbol{s} - 2 \lambda \boldsymbol{T} \times \boldsymbol{E} \right] d\boldsymbol{r} , \tag{40}$$

where

$$k = 0, \pm 1, \pm 2, \ldots .$$

Equation (40) can be taken as an auxiliary condition for hydrodynamical variables, i.e. they cannot be prescribed arbitrarily, since they have to satisfy the auxiliary condition (40). It must be emphasized, however, that it is sufficient to choose the hydrodynamical variables so as to fulfil equation (40) at the moment $t = 0$, since — as can be proved — the auxiliary condition if satisfied at $t = 0$, will remain satisfied at any time later.

Thus, for fixed electromagnetic potentials the hydrodynamical quantities satisfying the auxiliary condition determine a single-valued wave function (apart from the time dependent S_0 function contained in the expression of S). However, when we want to satisfy requirement 2 and substitute the wave function expressed in terms of the hydrodynamical variables in the wave equation it can be shown after a tedious calculation that it reduces to an identity provided the hydrodynamical variables obey the equation of motion and the constant of integration S_0 is taken to be a definite function apart from an additive constant S_0.

In the present paper we have succeeded in introducing hydrodynamical variables satisfying requirement 1. In a subsequent paper we shall proceed to derive equations of motions of hydrodynamical variables and prove that the variables introduced and their equations of motion satisfy requirements 2, 3 and 4.

REFERENCES

1. L. Jánossy and M. Ziegler, Acta Phys. Hung., **16**, 37, 1963.
2. L. Jánossy and M. Ziegler-Náray, Acta Phys. Hung., **16**, 345, 1964.
3. L. Jánossy and M. Ziegler-Náray, Acta Phys. Hung., **20**, 233, 1966.
4. J. D. Jackson, Classical Electrodynamics, John Wiley and Sons Inc. New-York, London, 1962.
5. M. Huszár, Acta Phys. Hung. **23**, 225, 1967.
6. Huszár Miklós and Náraynė Ziegler Mária, KFKI Közlemények, to be published (in Hungarian).
7. L. Jánossy, Acta Phys. Hung., **20**, 67, 1966.

ГИДРОДИНАМИЧЕСКАЯ МОДЕЛЬ ВОЛНОВОЙ МЕХАНИКИ, IV

М. ХУСАР и МАРИЯ ЦИГЛЕР-НАРАИ

Резюме

Продолжением работ (1, 2, 3) гидродинамическая модель волновой механики распространяется на гидродинамическую модель волнового уравнения, включающего спин-орбитальное взаимодействие. Вместо волновой функции вводятся гидродинамические переменные и показывается, что между волновой функцией и гидродинамическими переменными, удовлетворяющими некоторым рациональным физическим требованиям, имеется однозначное соответсвие. Уравнение движения для этих новых переменных выводится в следующей работе.

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COMMUNICATIONES BREVES

A THEORETICAL STUDY OF THE ULTRAVIOLET ABSORPTION SPECTRA OF BIPHENYL

By

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Introduction

The UV spectrum of biphenyl has been the subject of many experimental and theoretical studies [2], [7], [11], [12], [18]. The reduced interaction of the two benzene rings in the non-planar compound is responsible for the deviations in the absorption spectrum of the hindered compound from that expected for the planar model. Then attempts have been made to correlate the electronic absorption spectrum with the angle of twist. This is also the aim of this work.

Method and parameters

The semiempirical treatment due to **Pariser**, **Parr** and **Pople** [15], [16] has been extensively used for the calculation of electronic properties of conjugated molecules. We used in this work a variable β modification of this method, introduced by **Nishimoto** and **Forster** [13].

The configuration interaction included the four highest occupied orbitals and the four lowest empty ones.

The benzene rings were assumed to be perfect hexagons of side 1.4 Å joined by a $1-1'$ bond of constant length 1.5 Å for all values of the angle of twist Θ, the long molecular axis lying along the y direction and the $x-y$ plane containing one of the rings.

The two-centre core integrals, the distances and the two-centre repulsion integrals between neighbouring atoms were adjusted at every interation by means of the relations:

$$\beta_{\mu\nu} = -0{,}53\, p_{\mu\nu} - 2{,}030$$

for all bonds except the $1-1'$ bond.

$$r_{\mu\nu} = -\,0{,}18\, p_{\mu\nu} + 1{,}517\,; \quad \gamma_{\mu\nu} = \frac{14{,}397}{a_{\mu\nu} + r_{\mu\nu}}\,. \qquad [14]$$

We used the following empirical parameter values:

$$I_c = 11{,}16\text{ ev}; \quad \gamma_c = I_c - A_c = 11{,}13\text{ ev}. \qquad [6]$$

Following the usual practice, $\beta_{\mu\nu}$ was set equal to zero for all atoms μ non-bonded to ν. For bonded atoms, the initial value of $\beta_{\mu\nu}$ was -2.39 ev. For the determination of β_{112} with the planar configuration, use was made of the approximate proportionality of the $\pi - \pi$ overlap integral S and β [10]. Thus, the value of β for the benzene ring bonds was reduced by the factor $S_{1.4}/S_{1.2} = 0.859$ [9]

$$\beta_{11'}(\Theta = 0°) = -2{,}39 \times 0{,}859 = -2{,}05\text{ ev},$$

$$\beta_{11'}(\Theta > 0°) = \beta_{11'}(\Theta = 0°) \times \cos\Theta.$$

Results and discussion

The experimental data were taken from an atlas of UV absorption spectra [19]. The experimental values of the oscillator strength f were estimated from the absorption curves by means of the relation:

$$f = 4{,}32\, 10^{-9}\, \varepsilon_{\max}\, \Delta\bar{\nu}, \qquad [17]$$

where $\Delta\bar{\nu}$ is the band width (in cm^{-1}) at half-maximum extinction.

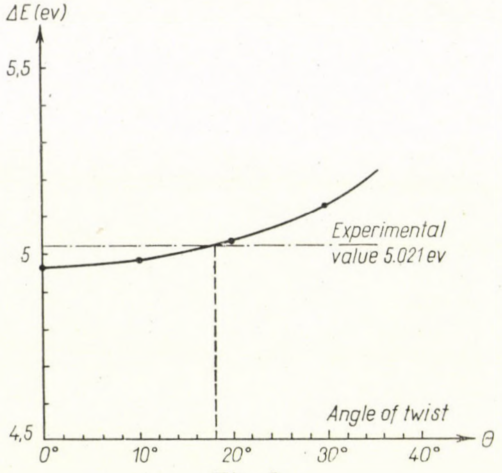

Fig. 1

The calculations were made for the values 0°, 10°, 20° and 30° of the angle of twist Θ. The results for the transition energies and intensities are summarized in Table 1. In Fig. 1 the calculated transition energy of the first band versus the angle Θ is plotted.

Table 1

Orbital jump	$\Theta = 0°$			$\Theta = 10°$			$\Theta = 20°$			$\Theta = 30°$			Experimental data		
	ΔE (ev)	f	pol.	ΔE (ev)	f	pol.	ΔE (ev)	f	pol.	ΔE (ev)	f	pol.	ΔE (ev)	f	pol.
$\varphi_5 \to \varphi_7$	4.637	0.	x	4.643	0.	x, z	4.663	0.	x, z	4.695	0.	x, z			
$\varphi_4 \to \varphi_7$	4.685	0.	—	4.690	0.	z, x	4.705	0.	z, x	4.729	0.	z, x			
$\varphi_6 \to \varphi_7$	4.960	0.85	y	4.979	0.84	y	5.035	0.82	y	5.130	0.77	y	5.021	0.4	y
$\varphi_6 \to \varphi_9$	6.162	0.	—	6.174	0.01	z, x	6.210	0.04	z, x	6.268	0.08	z, x			
$\varphi_3 \to \varphi_7$	6.223	0.	—	6.223	0.	—	6.223	0.	—	6.222	0.	—			
$\varphi_6 \to \varphi_8$	6.430	1.01	x	6.437	1.0	x, z	6.456	0.97	x, z	6.490	0.92	x, y	6.174	1.31	
$\varphi_5 \to \varphi_8$	6.503	1.31	y	6.504	1.31	y	6.504	1.32	y	6.504	1.32	y			
$\varphi_6 \to \varphi_{10}$	6.590	0.	—	6.597	0.	—	6.606	0.	—	6.621	0.	—			
$\varphi_4 \to \varphi_9$	7.025	0.	y	7.025	0.	y	7.025	0.	y	7.025	0.	y			
$\varphi_5 \to \varphi_9$	7.023	0.	y	7.023	0.	y	7.023	0.	y	7.023	0.	y			

X-ray diffraction studies show that in the crystalline state biphenyl is planar [5]. **Dale** [4] has measured the spectrum of solid biphenyl by the pressed-KCl disc technique and finds a band with a maximum at 4.9 ev. **Suzuki** [18] has corrected this value for the "normal red shift" observed when samples are measured in KCl discs and suggests that planar biphenyl should show the characteristic band at 4.97 ev. This value agrees well with our results (4.96 ev).

Electron diffraction results [1] indicate that in the vapour phase Θ has a value of about 42°. Probably for biphenyl in solution the value of Θ should be intermediate. Our theoretical curve (Fig. 1) gives a value of 18° for biphenyl dissolved in light petroleum.

The first band is found to be long-axis polarized, a fact about which there is general experimental agreement [3], [8], [18]. As usual, the calculated value of the oscillator strength is some two times too large.

Wenzel [20] has proposed that this intense band is a superposition of a strong and a weak transition corresponding to the 5.9 ev and 4.76 ev bands of benzene, respectively. The existence of this weak band has been corroborated by **Dale** [4]; it may be assigned to be a transversely polarized transition.

The two lowest transitions $\varphi_5 \rightarrow \varphi_7$ and $\varphi_4 \rightarrow \varphi_7$ that have their transition moments along the x and z axes are probably responsible for this "hidden" transition.

The theoretical energies for the second band are less accurate than for the first band, but the oscillator strengths agree well with experiment.

REFERENCES

1. A. **Almenningen** and O. **Bastiansen**, Klg. Norske Videns. Selk. Skrifter 4, 1958.
2. G. H. **Beaven**, Steric effects in conjugated systems; editor G. W. Gray p. 22. Butterworths, London, 1958.
3. R. **Coffman** and D. S. **Mc. Clure**, Canad. J. Chem., **36,** 48, 1958.
4. J. **Dale**, Acta Chem. Scand., **11,** 650, 1957.
5. J. **Dhar**, Indian J. Phys. **7,** 43, 1932, Proc. Nat. Inst. Sci. India, **15,** 11, 1949.
6. J. **Hinze** and H. H. **Jaffe**, J. Am. Chem. Soc., **84,** 540, 1962.
7. A. **London**, J. Chem. Physics, **13,** 396, 1945.
8. D. S. **Mc Clure**, Canad. J. Chem., **36,** 59, 1958.
9. R. S. **Mulliken**, C. A. **Rieke**, D. **Orloff** and H. **Orloff**: J. Am. Chem. Soc., **17,** 1248, 1949.
10. R. S. **Mulliken**, C. A. **Rieke** and W. G. **Brown**, J. Am. Chem. Soc., **63,** 41, 1941.
11. J. N. **Murrell**, J. Chem. Soc. p. 3779, 1956.
12. J. N. **Murrell** and H. C. **Longuet-Higgins**, Proc. Phys. Soc. London A, **68,** 601.
13. K. **Nishimoto** and L. S. **Forster**, Theoret. Chim. Acta, **3,** 407, 1965; **4,** 155, 1966.
14. K. **Nishimoto** and N. **Mataga**, Z. Physik. Chem. (Frankfurt), **12,** 335, 1957.
15. R. **Pariser**, and R. G. **Parr**, J. Chem. Physics, **21,** 466, 767, 1953; **24,** 250, 1956.
16. J. A. **Pople**, Trans. Faraday Soc. **49,** 1375, 1953.
17. C. **Sandorfy**, Electronic spectra and quantum chemistry, p. 103, Prentice-Hall, Englewood Cliffs, N. J. 1964.
18. H. **Suzuki**, Bull. Chem. Soc. Japan, **32,** 1340, 1350, 1959.
19. UV atlas of organic compounds (Weinheim. Verlag Chemie, 1966).
20. A. **Wenzel**, J. Chem. Physics, **21,** 403, 1953.

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MAGNETORESISTIVITY OF *n*-TYPE GaAs SINGLE CRYSTALS IN THE IMPURITY SCATTERING RANGE

By

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Much useful information on the band structure and scattering mechanism of charge carriers in semiconductors can be obtained by measurements of magnetoresistivity. Most magnetoresistivity measurements of the group $A^{III} B^{V}$ crystals have been performed on InSb single crystals, and relatively few measurements have been made on other crystals, e.g. on GaAs.

The first measurements of magnetoresistivity of GaAs single crystals were made by J. T. EDMOND et al., [1], O. V. EMELYANENKO et al., [2], and M. GLICKSMAN [3]. These measurements were performed at room temperature and at 77° K on *n*-type single crystals, with electron concentrations of $10^{16}-10^{17}$ cm^{-3} and room temperature mobilities of 3000—4000 cm^2/Vs. It has been found that the longitudinal magnetoresistivity was negligibly small, [2], [3], and the transverse effect was isotropic, [3], in accordance with the well known model of the conduction band of GaAs having a spherical minimum in the middle of the Brillouin zone [4]. The magnetoresistivity was proportional to the square of the magnetic field up to 10.000 Gauss [1]. The above conclusions were verified recently by D. FRITSCH and H. WEISS [5], on measuring the magnetoresistivity of higher purity *n*-type crystals, (room temperature mobility $\mu = 6000$ cm^2/Vs) at 77° K. The results of the measurements of A. F. KRAVCHENKO and H. Y. FAN [6], who found anisotropic transverse magnetoresistivity in *n*-type GaAs in the temperature range of 77—300° K, are not consistent with the foregoing results, as was pointed out in [5]. This anisotropy was subsequently interpreted by A. F. KRAVCHENKO et al., [7], on the basis of a special anisotropic scattering mechanism due to the segregation of impurities. R. K. WILLARDSON et al., [8], have measured the magnetoresistivity of pure *n*-type GaAs crystals at 300, 196, and 77° K temperatures. The room temperature electron concentration was 4×10^{15} cm^{-3}, the mobility was 6900 cm^2/Vs. The experimental results were interpreted on the basis of mixed acoustic phonon and ionized impurity scattering, with dominant phonon contribution. However, at room temperature and below, the acoustic phonon scattering is negligible compared with the polar and impurity scatterings [4].

In recent work the magnetoresistivity of impure n-type GaAs single crystals has been investigated in the impurity scattering range. Transverse magnetoresistivity was measured at 77° K temperature, in low magnetic fields (4000 Gauss). In addition, the Hall effect and conductivity were measured at 77° K and at room temperature.

The characteristics of the samples and the experimental results are shown in Table 1.

Table 1

Sample No	R_H(300°K) cm³/As	μ_H(300°K) cm²/Vs	R_H(77° K) cm³/As	μ_H(77° K) cm²/Vs	ξ(77° K) measured	ζ(77° K)	ξ(77° K) calculated
102/2	−37.4	2360	−81.1	2050	0.42	+1.2	0.43
102/3	−36.0	3500	−41.0	3200	0.27	+2.25	0.34
102/4	−33.7	2370	−29.7	1980	0.42	+3.00	0.30
105/11	−326	2650	−489	4220	0.51	−0.9	0.54
105/12	−108	1870	−290.5	4400	0.34	−0.3	0.52
F/1	−151.6	3500	−249.3	4140	0.30	−0.1	0.51
F/11	−144.0	3020	−196.5	3280	0.35	+0.2	0.50

The room temperature electron concentration can be determined only approximately because of the simultaneous effect of two scattering mechanisms, i.e. polar and impurity scatterings. However, assuming that the scattering factor is $r = 1$, then the electron concentration of crystals of group 102 is of the order of 2×10^{17} cm^{-3}, and of crystals of group F is of the order of 4×10^{16} cm^{-3}.

At 77° K the magnetoresistivity was proportional to the square of the magnetic field up to 4000 Gauss, $\Delta\varrho/\varrho \sim B^2$. Table 1 contains the values of $\xi = \Delta\varrho/(\varrho\, \mu^2 B^2)$.

The quadratic dependence on the magnetic field is a natural consequence of the fact that the measurements were performed in small magnetic fields, i.e. $(\omega\tau)^2 \approx (\mu_H B)^2 \ll 1$, where ω is the cyclotron frequency, τ is the relaxation time of the electrons.

Since at 77° K temperature only one scattering mechanism, the ionized impurity scattering is dominant, the experimental results can be interpreted quantitatively using the proper formulae, and also taking into account the degeneracy of conduction electrons. Considering the ionized impurity scattering, the relaxation time

$$\tau = \frac{16\,\pi\,(2\,m^*)^{1/2}\,\varepsilon^2\,E^{3/2}}{e^4 N_I[\ln(1+\beta^2) - \beta^2/(1+\beta^2)]}\,, \tag{1}$$

$$\beta^2 = \frac{8\,m^*\,\varepsilon kTE}{\hbar^2\,e^2\,n}\,,$$

where the meaning of symbols is the following

m^*	effective mass of electrons,
ε	dielectric constant,
n	electron concentration,
N_I	concentration of ionized impurities, in compensated crystals it is equal to the sum of donor and acceptor concentrations,
E	electron energy,

and others are used as usual.

Taking into account the Fermi—Dirac statistics, the electron mobility [9]

$$\mu = \frac{32 \sqrt{2}\, \pi \varepsilon^2 (kT)^{3/2} F_2(\zeta)}{e^3\, m^{*1/2} N_I \left[\ln (1+\beta_0^2) - \beta_0^2/(1+\beta_0^2)\right] F_{1/2}(\zeta)}\,, \tag{2}$$

where $F_n(\zeta)$ is the Fermi integral, ζ is the reduced Fermi level, and

$$\beta_0^2 = \frac{8m^*\, \varepsilon (kT)^2}{\hbar^2\, e^2\, n}\, x_0\,, \tag{3}$$

where x_0 is the root of equation

$$\frac{x_0+3}{x_0-3} = \exp(x_0 - \zeta)\,.$$

The Hall constant

$$R_H = -\frac{r}{ne}\,, \tag{4}$$

$$r = \frac{\langle \tau^2 \rangle}{\langle \tau \rangle^2} = \frac{3\, F_{7/2}(\zeta)\, F_{1/2}(\zeta)}{4\, [F_2(\zeta)]^2}\,, \tag{5}$$

where

$$n = N_c\, F_{1/2}(\zeta) = 4\,\pi \left(\frac{2\, m^*\, kT}{h^2}\right)^{3/2} F_{1/2}(\zeta)\,. \tag{6}$$

The coefficient of magnetoresistivity

$$\xi = \frac{\Delta \varrho}{\varrho \mu_H^2\, B^2} = \frac{8\, F_5(\zeta) F_2(\zeta)}{9\, [F_{7/2}(\zeta)]^2} - 1\,. \tag{7}$$

The reduced Fermi level, ζ, can be determined from the measured Hall coefficient, using Eqs. (4), (5) and (6). Table 1 contains the values of ζ determined from these equations. The coefficient of magnetoresistivity, ξ, can be evaluated from (7). Table 1 contains these values, too.

The measurements of samples belonging to the group 102 gave the following results: for sample 102/2 the measured and calculated values agreed well, whereas in the case of samples 102/3 and 102/4 the measured values were lower and higher, respectively, than the calculated ones. The same observations can be made in connection with the crystals belonging to group 105.

Taking into account that the absolute values of the measured magnetoresistivities were extremely small (1—2‰), so that the experimental errors were high (according to our estimations 20—30%), the agreement is satisfactory.

On the other hand for crystals of group *F*, the differences between the calculated and measured values are greater. The compensation degree of the three groups of crystals as calculated from the mobilities, was roughly equal, and thus, could not be the cause of the observed differences.

Other scattering mechanisms can appreciably lower the magnetoresistivity, as observed in the samples of group *F*. Neutral impurity scattering, or scattering by structural defects may be such a mechanism. Investigations of crystalline defects in our crystals are in progress.

We wish to express our acknowledgements to Dr. G. GERGELY for suggesting the experimental work, and to Dr. E. PAPP of the Research Institute for Metallurgical Industry, Budapest, for supplying the crystals.

REFERENCES

1. J. T. EDMOND, R. F. BROOM and F. A. CUNNEL in Rept. Meeting on Semiconductors, p. 109, Rugby, 1956.
2. О. В. Емельяненко, Д. Н. Наследов, ЖТФ, **28,** 1117, 1958.
3. M. GLICKSMAN, J. Phys. Chem. Sol., **8,** 511, 1959.
4. H. EHRENREICH, Phys. Rev., **120,** 1951, 1960.
5. D. FRITSCH and H. WEISS in Semiconductors and Semimetals Vol. 1. p. 373. Ed. R. K. Willardson, 1966.
6. A. F. KRAVCHENKO and H. Y. FAN, Proc. Int. Conf. Semiconductors, p. 737. Exeter, 1962.
7. A. F. KRAVCHENKO and V. S. SARDARYAN, phys. stat. sol., **17,** 479, 1966.
8. R. K. WILLARDSON and J. J. DUGA, Proc. Phys. Soc., **75,** 280, 1960.
9. E. H. PUTLEY, The Hall Effect and Related Phenomena, London, 1960.

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MEAN AMPLITUDES OF VIBRATION FOR AN XY_4 MOLECULAR MODEL WITH D_{2d} SYMMETRY: APPLICATION TO PHOSPHATE ION

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A method has been developed on the basis of symmetry considerations for the determination of mean amplitudes of vibration for an XY_4 molecular model of slightly distorted tetrahedron possessing a D_{2d} symmetry and applied to the phosphate ion. The available four fundamental frequencies of B_2 and E species have been considered and the mean-square amplitude quantities have been computed at room temperature. The results are discussed briefly.

Introduction

For the four classes of XY_4 molecules and ions having highly symmetric configurations, namely, a regular tetrahedron with the symmetry point group T_d, a tetragonal square plane belonging to the symmetry point group D_{4h}, a tetragonal pyramid coming under the C_{4v} symmetry, and a slightly distorted tetrahedron with the symmetry point group D_{2d}, a theory for the determination of mean amplitudes of vibration was first developed by CYVIN [1] utilizing the symmetry coordinates for molecules of the first type, on the basis of which many molecules and ions have been studied by several investigators [2—16]. Later on, a method was proposed by NAGARAJAN [17] on the basis of the principle postulated by CYVIN [18] for the determination of mean amplitudes of vibration for molecules of the second type and this was applied to many ions [17, 19] and xenon tetrafluoride [20—22]. On the basis of the principle postulated by CYVIN [18], a method utilizing symmetry coordinates was proposed by NAGARAJAN [23] for the determination of mean amplitudes of vibration for the molecules of the third type and applied to the skeletal vibrations of stable pentaborane. No method has so far been developed for the fourth type and applied to any system. Hence, it is aimed here to develop a method for the determination of mean amplitudes of vibration for the XY_4 molecular model of slightly distorted tetrahedron with the D_{2d} symmetry by the CYVIN method [18] and then apply it to the phosphate ion so that the results of the present

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study should be very useful for the interpretation of the results of electron diffraction studies and for the evaluation of normal frequencies in other related ions.

Mean amplitudes of vibration

A molecule or ion of an XY_4 type possessing a slightly distorted tetrahedron with the symmetry point group D_{2d} gives rise, according to the relevant symmetry considerations and selection rules [24, 25], to nine vibrational degrees of freedom constituting only seven fundamental frequencies. They are distributed under the various irreducible representations as follows:

$$2A_1(\nu_1, \nu_2) + B_1(\nu_3) + 2B_2(\nu_4, \nu_5) + 2E(\nu_{6a}, \nu_{7a} \text{ or } \nu_{6b}, \nu_{7b}),$$

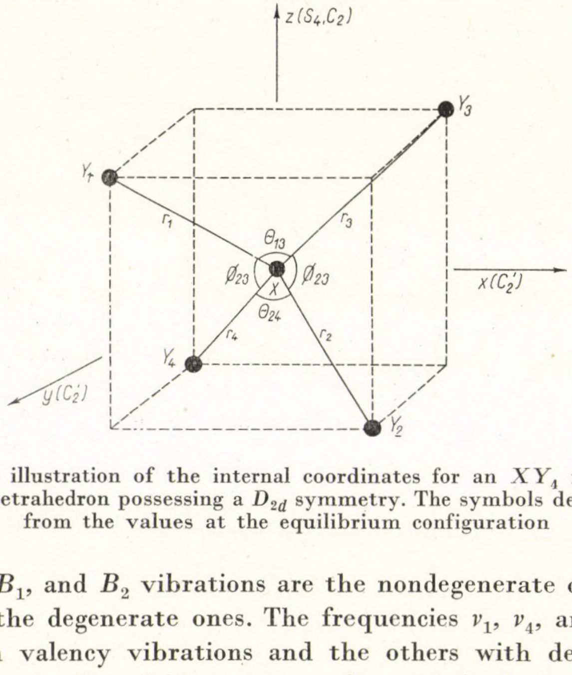

Fig. 1. Geometric illustration of the internal coordinates for an XY_4 molecular model of slightly distorted tetrahedron possessing a D_{2d} symmetry. The symbols denote the deviations from the values at the equilibrium configuration

where the A_1, B_1, and B_2 vibrations are the nondegenerate ones while the E vibrations are the degenerate ones. The frequencies ν_1, ν_4, and ν_{6a} or ν_{6b} are associated with valency vibrations and the others with deformations. The vibrations under the B_2 and E species are allowed in both the infrared absorption and Raman spectra while those of A_1 and B_1 species are allowed only in the Raman spectrum but forbidden in the infrared absorption spectrum. In order to construct the symmetry coordinates, the following set of internal coordinates has been introduced: Θ_{13} and Θ_{24} are the $Y-\hat{X}-Y$ bending coordinates along the $z(S_4, C_2)$ axis; Φ_{12}, Φ_{14}, Φ_{23}, and Φ_{34} are the $Y-\hat{X}-Y$ bending coordinates along the $x(C_2')$ and $y(C_2')$ axes; and r_1, r_2, r_3, and r_4 are the $X-Y$ stretching coordinates (see Fig. 1). With the help of these internal coordinates the following set of symmetry coordinates satisfying the conditions of normalization, orthogonality and transformations of the vibration species concerned has been constructed, including one redundancy:

$$
\begin{aligned}
S_1(A_1) &= (r_1 + r_2 + r_3 + r_4)/2\,,\\
S_2(A_1) &= R(2\Theta_{13} + 2\Theta_{24} - \Phi_{12} - \Phi_{14} - \Phi_{23} - \Phi_{34})/\sqrt{12},\\
S_0(A_1) &= R(\Theta_{13}+\Theta_{23}+\Phi_{12}+\Phi_{14}+\Phi_{23}+\Phi_{34})/\sqrt{6} = 0 \text{ (redundant)},\\
S_3(B_1) &= R(\Phi_{12} - \Phi_{14} - \Phi_{23} + \Phi_{34})/2\\
S_4(B_2) &= (-r_1 + r_2 - r_3 + r_4)/2\,,\\
S_5(B_2) &= R(\Theta_{13} - \Phi_{24})/\sqrt{2}\,,\\
S_{6a}(E) &= (r_1 + r_2 - r_3 - r_4)/2,\\
S_{6b}(E) &= (r_1 - r_2 - r_3 + r_4)/2\,,\\
S_{7a}(E) &= R(-\Phi_{12} + \Phi_{35})/\sqrt{2}\,,\\
S_{7b}(E) &= R(-\Phi_{14} + \Phi_{23})/\sqrt{2}\,,
\end{aligned}
$$

where r_i is the change in the length of the ith $X-Y$ bond; $\Theta_{i,i+2}$ the change of the angle between the r_i and r_{i+2} bonds; and Θ_{ij} the change of the angle between r_i and r_j bonds along the $x(C_2')$ and $y(C_2')$ axes. The reason for the existence of two kinds of bendings is due to the presence of an S_4 symmetry element on one of the three Cartesian axes. It is seen here that the angle displacements are multiplied by the equilibrium bond length $R(X-Y)$ in order to keep the dimensions of the mean-square amplitude quantities referring to the angle bending the same as those of the quantities due to the bonded and non-bonded atom pairs. These symmetry coordinates are the same as those of MOONEY and TOMA [24] except for the changes in the notations of the bending coordinates.

On the basis of the symmetry coordinates constructed above for this molecular model, the symmetrized mean-square amplitude in terms of the mean-square amplitude quantities have been obtained, and they are given as follows:

$$
\begin{aligned}
\Sigma_{11}(A_1) &= \langle s_1^2\rangle = \sigma_r + 3\,\sigma_{rr}\,,\\
\Sigma_{22}(A_1) &= \langle s_2^2\rangle = (2\,\sigma_\Theta + \sigma_\Phi + 2\,\sigma_{\Theta\Theta} - 8\,\sigma_{\Theta\Phi} + 2\,\sigma_{\Phi\Phi} + \sigma_{\Phi\Phi'})/3\,,\\
\Sigma_{12}(A_1) &= \Sigma_{21}(\mathrm{A}_1) = \langle s_1\,s_2\rangle = 2\,(\sigma_{r\Theta} + \sigma_{r\Theta'} - \sigma_{r\Phi} - \sigma_{r\Phi'})/\sqrt{3}\,,\\
\Sigma_{33}(B_1) &= \langle s_3^2\rangle = \sigma_\Phi - 2\,\sigma_{\Phi\Phi} + \sigma_{\Phi\Phi'}\,,\\
\Sigma_{44}(B) &= \langle s_4^2\rangle = \sigma_r - \sigma_{rr}\,,\\
\Sigma_{55}(B_2) &= \langle s_5^2\rangle = \sigma_\Theta - \sigma_{\Theta\Theta}\,,\\
\Sigma_{45}(B_2) &= \Sigma_{54}(B_2) = \langle s_4\,s_5\rangle = \sqrt{2}\,(-\sigma_{r\Theta} + \sigma_{r\Theta'})\,,\\
\Sigma_{66}(E) &= \langle s_{6a}^2\rangle = \langle s_{6b}^2\rangle = \sigma_r - \sigma_{rr}\,,
\end{aligned}
$$

$$\Sigma_{77}(E) = \langle s_{7\mathrm{a}}^2 \rangle = \langle s_{7\mathrm{b}}^2 \rangle = \sigma_{\Phi} = \sigma_{\Phi\Phi'} ,$$

$$\Sigma_{67}(E) = \Sigma_{76}(E) \langle s_{6\mathrm{a}}\, s_{7\mathrm{a}} \rangle = \langle s_{6\mathrm{b}}\, s_{7\mathrm{b}} \rangle = \sqrt{2}\,(-\sigma_{r\Phi} + \sigma_{r\Phi'}) ,$$

where σ_r is the mean-square amplitude quantity due to the bonded atom pair $X-Y$, σ_{rr} the quantity due to the interaction of bonded atom pairs, σ_{Θ} the quantity due to the bending $Y-\hat{X}-Y$ in the $z(S_4, C_2)$ axis, σ_{Φ} the quantity due to the bending $Y-\hat{X}-Y$ in the $x(C_2')$ axis as well as the $y(C_2')$ axes; $\sigma_{\Theta\Theta}$ the quantity due to the interaction of $Y-\hat{X}-Y$ bendings in the $z(S_4, C_2)$ axis having no common side; $\sigma_{\Phi\Phi}$ the quantity due to the interaction of $Y-X-Y$ bendings having common side in the $x(C_2')$ and $y(C_2')$ axes; $\sigma_{\Phi\Phi'}$ the quantity due to the interaction of $Y-\hat{X}-Y$ bendings having no common side in the $x(C_2')$ and $y(C_2')$ axes; $\sigma_{\Theta\theta}$ the quantity due to the interaction of $Y-\hat{X}-Y$ bending in the $z(S_4, C_2)$ axis and $Y-\hat{X}-Y$ bending in the $x(C_2')$ or $y(C_2')$ axis; $\sigma_{r\Theta'}$ the quantity due to the interaction of bonded atom pair $X-Y$ with the adjacent $Y-\hat{X}-Y$ bending in the $z(S_4, C_2)$ axis; $\sigma_{r\Theta}$ the quantity due to the interaction of the bonded atom pair $X-Y$ with the non-adjacent $Y-\hat{X}-Y$ bending in the $z(S_4, C_2)$ axis; $\sigma_{r\Theta}$ the quantity due to the interaction of the bonded atom pair $X-Y$ with the adjacent $Y-\hat{X}-Y$ bending in the $x(C_2')$ or $y(C_2')$ axis; and $\sigma_{r\Phi'}$ the quantity due to the interaction of the bonded atom pair $X-Y$ with the nonadjacent $Y-\hat{X}-Y$ bending in the $x(C_2')$ or $y(C_2')$ axis. In addition to these quantities due to the bonded atom pair, bendings and the respective interaction terms, the analytical expressions for the following mean-square amplitude quantities for the nonbonded atom pairs have been obtained in terms of the mean-square amplitude quantities due to the bonded atom pair, bendings, and the respective interaction terms and they are given as follows:

$$\begin{aligned}
\sigma_p &= 2\,(\sigma_r + \sigma_{rr}) \sin^2(\Theta'/2) + \sigma_{\Theta} \cos^2(\Theta'/2) + 4\,\sigma_{r\Theta} \sin(\Theta'/2) \cos(\Theta'/2) ,\\
\sigma_{pp} &= 4\,\sigma_{rr} \sin^2(\Theta'/2) + 4\,\sigma_{r\Theta'} \sin(\Theta'/2) \cos(\Theta'/2) + \sigma_{\Theta\Theta} \cos^2(\Theta'/2) ,\\
\sigma_q &= 2\,(\sigma_r + \sigma_{rr}) \sin^2(\Phi'/2) + \sigma_{\Phi} \cos^2(\Phi'/2) + 4\,\sigma_{\cdot\Phi} \sin(\Phi'/2) \cos(\Phi'/2) ,\\
\sigma_{qq} &= 4\,\sigma_{rr} \sin^2(\Phi'/2) + 2\,(\sigma_{r\Phi} + \sigma_{r\Phi'}) \sin(\Phi'/2) \cos(\Phi'/2) + \sigma_{\Phi\Phi} \cos^2(\Phi'/2) ,\\
\sigma_{qq'} &= 4\,\sigma_{rr} \sin^2(\Phi'/2) + 4\,\sigma_{r\Phi'} \sin(\Phi'/2) \cos(\Phi'/2) + \sigma_{\Phi\Phi'} \cos^2(\Phi'/2) ,\\
\sigma_{pq} &= (\sigma_r + 3\,\sigma_{rr}) \sin(\Phi'/2) \sin(\Phi'/2) + (\sigma_{r\Theta} + \sigma_{r\Theta'}) \sin(\Phi'/2) \cos(\Theta'/2) ,\\
&\quad + (\sigma_{r\Phi} + \sigma_{r\Phi'}) \sin(\Theta'/2) \cos(\Phi'/2) + \sigma_{\Theta\Phi} \cos(\Phi'/2) \cos(\Phi'/2) ,
\end{aligned}$$

where σ_p is the mean-square amplitude quantity due to the non-bonded atom pair Y_1-Y_3 or Y_2-Y_4; σ_{pp} the quantity due to the interaction of non-bonded atom pairs Y_1-Y_3 and Y_2-Y_4; σ_q the quantity due to the non-bonded atom pair Y_1-Y_2 or Y_1-Y_4 or Y_2-Y_3 or Y_2-Y_4; σ_{qq} the quantity due to the interaction of non-bonded atom pairs; Y_1-Y_2 and Y_1-Y_4 or Y_2-Y_3 and

$Y_3 - Y_4$ having a common side; $\sigma_{qq'}$ the quantity due to the interaction of non-bonded atom pairs $Y_1 - Y_4$ and $Y_2 - Y_3$ having no common side; and σ_{pq} the quantity due to the interaction of non-bonded atom pair $Y_1 - Y_3$ or $Y_2 - Y_4$ with all the other non-bonded atom pairs. In the above expressions the angles Θ' and Φ' are the values for the equilibrium configuration. It has to be mentioned that a difference in the quantity due to the bending along the S_4 axis from that of the bending which lies skew to that S_4 axis has been assumed while a difference in the quantities due to the interaction of bonded atom pairs required for the D_{2d} symmetry has been assumed to be negligible and hence all the quantities due to the interaction of bonded atom pairs are the same.

Normal frequencies

In order to construct the secular equations giving the normal frequencies in terms of the mean-square amplitude quantities, the basic equation $| \Sigma G^{-1} - E \Delta | = 0$ postulated by CYVIN [18] has been adopted for the present study. On the basis of this, the secular equations obtained for this molecular model are given as follows in terms of the symmetrized mean-square amplitude matrices, kinetic energy matrices and fundamental frequencies:

For A_1 type vibrations:

$$\Delta_1 + \Delta_2 = [\Sigma_{11} + (1/3)\, \Sigma_{22}]\, \mu_y^{-1},$$

$$\Delta_1 \Delta_2 = (\Sigma_{11} \Sigma_{22} - \Sigma_{12}^2)\, (1/3)\, \mu \quad .$$

For B_1 type vibration:

$$\Delta_3 = (1/3)\, \Sigma_{33}\, \mu_y^{-1} .$$

For B_2 type vibrations:

$$\begin{aligned} \Delta_4 + \Delta_3 = (1/3)\, [(8\, \mu_x + 3\, \mu_y)\, \Sigma_{44} + (1/2)\, (4\, \mu_x + 3\, \mu_y)\, \Sigma_{55} - \\ - 8\, \mu_x \Sigma_{45}]\, (4\, \mu_x + \mu_y)^{-1}\, \mu_y^{-1}, \end{aligned}$$

$$\Delta_4 \Delta_5 = (\Sigma_{44} \Sigma_{55} - \Sigma_{45}^2)\, (1/2)\, (4\, \mu_x + \mu_y)^{-1}\, \mu_y^{-1} .$$

For E type vibrations:

$$\begin{aligned} \Delta_6 + \Delta_7 = (1/3)\, [(8\, \mu_x + 3\, \mu_y)\, \Sigma_{66} + (1/2)\, (4\, \mu_x + 3\, \mu_y)\, \Sigma_{77} - \\ - 8\, \mu_x \Sigma_{67}]\, (4\, \mu_x + \mu_y)^{-1}\, \mu_y^{-1}, \end{aligned}$$

$$\Delta_6 \Delta_7 = (\Sigma_{66} \Sigma_{77} - \Sigma_{67}^2)\, (1/2)\, (4\, \mu_x + \mu_y)^{-1}\, \mu_y^{-1} .$$

In the above equations Δ has been related to the normal frequency ν in the form

$$\Delta_i = (h/8\pi^2 \nu_i) \coth(h\nu_i/2\,kT)\,,$$

where h is Planck's constant, k the Boltzmann constant and T the temperature in degrees Kelvin.

Application to phosphate ion

The infrared spectrum of the solid yttrium phosphate crystal YPO_4 was studied by MOONEY and TOMA [24] and it was found that the cations in the crystal are located along the axes which coincide with the S_4 axis of the phosphate ion PO_4^{3-} in the D_{2d} symmetry and this perturbation is caused by an interaction of the cations with the phosphate ion PO_4^{3-} with the greatest effect occurring with those vibrational modes associated with deformations along the S_4 axis of the anion. According to the group theoretical arguments for a tetrahedral XY_4 ion in a crystal, the B_2 vibration is a function of the bendings lying along the S_4 axis and independent of the bendings lying skew to this S_4 axis. On the basis of these facts the fundamental frequencies were assigned from the observed infrared spectrum for the phosphate ion of slightly distorted tetrahedron possessing a D_{2d} symmetry and the force constants were then calculated on the basis of group theoretical method by MOONEY and TOMA [24]. Their observed fundamental frequencies are given as follows: $\nu_4(B_2) = 1070$ cm^{-1}, $\nu_5(B_2) = 643.1$ cm^{-1}, $\nu_6(E) = 1031$ cm^{-1}, and $\nu_7(E) = 526.3$ cm^{-1}. The aim here is to employ these four fundamental frequencies in the theory developed above and evaluate the mean-square amplitude quantities by assuming both angles involved in this model to be tetrahedral and considering a value 1.685 Å for the phosphorus—oxygen bond.

The secular equations giving the normal frequencies in terms of the mean-square amplitude quantities were constructed at room temperature for the B_2 and E species with the help of the symmetrized mean-square amplitude matrices, kinetic energy matrices, and the fundamental frequencies in cm^{-1} given by MOONEY and TOMA [24]. Since the angle in the S_4 axis deviates very slightly from the angles lying skew to that S_4 axis, both angles were assumed to be tetrahedral, and a value 1.685 Å for the phosphorus—oxygen bond was considered in the calculations of kinetic energy matrices. There are two equations with three unknowns under the B_2 as well as E species. It is not possible to evaluate all the three elements unless some restricting approximations are adopted. When the off-diagonal element was neglected under each species, the equations resulted in imaginary values for the diagonal elements. Hence, the off-diagonal element was introduced and the equations were solved in the manner described by TORKINGTON [26] and the three elements under each species were evaluated. The obtained values of the symmetrized mean-square amplitude matrices in Å^2 are given as follows: $\Sigma_{44}(B_2) =$

0.0020879, $\Sigma_{55}(B_2) = 0.0055489$, $\Sigma_{45}(B_2) = 0.0007965$, $\Sigma_{66}(E) = 0.0022749$, $\Sigma_{77}(E) = 0.0066478$ and $\Sigma_{67}(E) = 0.0004750$. Since the same expression $(\sigma_r - \sigma_{rr})$ is obtained under the B_2 and E species for Σ_{44} and Σ_{66}, an average value 0.0021814 is considered here to represent the mean-square amplitude quantity σ_r by ignoring σ_{rr}. From this, the calculated value of the mean amplitude of vibration for the $P—O$ bond is given as 0.0467 Å at room temperature. If the interaction quantities due to bendings are neglected, Σ_{55} represents σ_Θ and Σ_{77} represents σ_Φ. Accordingly, the mean-square amplitude quantity due to bending in the S_4 axis is smaller than that of the bending along the skew to that S_4 axis. Similarly, the nondiagonal element Σ_{45} represents the interaction of the bonded atom pair with the bending in the S_4 axis while Σ_{66} represents the interaction of the bonded atom pair with the bending in the $x(C_2')$ or $y(C_2')$ axis. From the values obtained here it is seen that the quantity due to the interaction of the bonded atom pair with the bending in the S_4 axis is greater than that of the quantity due to the interaction of the bonded atom pair with the bending along the skew to the S_4 axis. The values presented here would be very useful in the interpretation of the results of electron diffraction studies and for the evaluation of normal frequencies in other related systems.

REFERENCES

1. S. J. Cyvin, J. Mol. Spectroscopy, **5,** 38, 1960; **6,** 333, 338, 1961.
2. N. Brearley and S. Sundaraam, Z. Physik Chem. Neue Folge, **32,** 219, 1962.
3. G. Nagarajan Bull. Soc. Chim. Belges, **71,** 347, 361, 1962, **72,** 647, 657, 1963.
4. K. Sathianandan, K. Ramaswamy and F. F. Cleveland., J. Mol. Spectroscopy, **8,** 470, 1962.
5. G. Nagarajan, Indian J. pure appl. Phys., **2,** 17, 145, 205, 1964.
6. G. Nagarajan, Acta Phys. Austr., **18,** 23, 1964.
7. G. Nagarajan, Bull. Soc. Chim. Belges, **73,** 768, 1964.
8. Y. Anantharama Sarma, S. Sundaram and F. F. Cleveland, J. Mol. Spectroscopy, **13,** 67, 1964.
9. S. J. Cyvin, J. Brunvoll, B. N. Cyvin and E. Meisingseth, Bull. Soc. Chim. Belges, **73,** 5, 1964.
10. G. Nagarajan, Acta Phys. Polon., **28,** 875, 1965.
11. G. Nagarajan, Czech. J. Phys., **B16,** 157, 1966.
12. G. Nagarajan, Z. Naturforschg., **21a,** 244, 1966.
13. A. Muller and G. Nagarajan, Z. Naturforschg., **21b,** 508, 1966.
14. A. Muller, G. Nagarajan and A. Fadini, Z. anorg. allg. Chem., **347,** 269, 1966.
15. G. Nagarajan and A. Muller, Z. anorg. allg. Chem., **349,** 82, 1967.
16. J. R. Durig and G. Nagarajan, Monats. fur Chemie, **98,** 1967 in press.
17. G. Nagarajan, J. Mol. Spectroscopy, **12,** 198, 1964.
18. S. J. Cyvin, Spectrochim. Acta, **15,** 828, 1959.
19. G. Nagarajan, Acta Phys. Austr., **24,** 20, 1966.
20. G. Nagarajan, Acta Phys. Austr., **18,** 11, 1964.
21. G. Hagen, Acta Chem. Scand., **21,** 465, 1967.
22. W. A. Yeranos, Mol. Phys., **12,** 529, 1967.
23. G. Nagarajan, Indian J. pure appl. Phys., **4,** 347, 1966.
24. R. W. Mooney and S. Z. Toma, J. Chem. Phys., **46,** 3364, 1967.
25. G. Herzberg, "Infrared and Raman Spectra of Polyatomic Molecules", D. Van Nostrand Company, New York, 1960.
26. P. Torkington, J. Chem. Phys., **17,** 357, 1949.

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THE HARTREE ENERGY OF THE VALENCE ELECTRON OF K

By

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The Hartree and the Hartree—Fock methods are those most frequently used in the theoretical examination of atomic structure. Till now they have been used to determine the total energy and the charge distribution.

In the present paper we determined the exchange energy of the valence electron of K in the ground state (4*s*). We carried out the computation in the following way. We calculated the Hartree energy of the valence electron and subtracted this value from the Hartree—Fock energy to obtain the exchange energy. The Hamiltonian of the ground state of the valence electron is*

$$H = -\frac{1}{2}\Delta + U\,, \tag{1}$$

where Δ is the Laplace operator and U the potential energy of the valence electron. From the value of T-total $2Z_p$ given in the HARTREE Table [1]

$$U = \frac{T-2}{r} + \frac{2}{r}\,. \tag{2}$$

The energy of the valence electron in the (4*s*) state

$$E = \int_0^\infty \bar{\varphi}_{4s}^* H \bar{\varphi}_{4s}\, dr\,, \tag{3}$$

where $\bar{\varphi}_{4s}$ is the orthonormal eigenfunction of the valence electron and it can be written as the following sum

$$\bar{\varphi}_{4s} = N \sum_{i=1}^{4} A_i\, \varphi_{is}\,, \tag{4}$$

where N is the normalization constant, A_i were derived by the Schmidt method of orthogonalization and φ_{is} are the normalized radial wave functions of the electrons in the 1*s*, 2*s*, 3*s*, 4*s* states.

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* The expressions are written in this paper using atomic units.

The $1s$, $2s$, $3s$ wave functions given in the Hartree Table were approximated analytically to obtain the integrals that we used for the evaluation of the energy. The approximating functions are

$$\varphi_{is} = a_{is}\, r^{i}\, e^{-\lambda_i r}\,. \tag{5}$$

The normalized radial eigenfunction of the valence electron was derived by a minimization procedure of the energy of the $4s$ state, based on a method that has been used recently by GOMBÁS [2].

The orthogonalization and normalization conditions by which N and A_i were determined are the following.

The normalization condition

$$\int_0^\infty (\bar{\varphi}_{4s})^2\, dr = 1\,. \tag{6}$$

The orthogonalization conditions

$$\int_0^\infty \bar{\varphi}_{4s}\bar{\varphi}_{1s}\, dr = 0\,; \int_0^\infty \bar{\varphi}_{4s}\bar{\varphi}_{2s}/dr = 0\,; \int_0^\infty \bar{\varphi}_{4s}\bar{\varphi}_{3s}\, dr = 0\,, \tag{7}$$

where $\bar{\varphi}_{1s}$, $\bar{\varphi}_{2s}$, $\bar{\varphi}_{3s}$ are the orthonormal eigenfunctions of the $1s$, $2s$, $3s$ states, respectively. They have been composed from the normalized radial wave functions of the particular states.

In calculating the energy, the first term of the potential energy has been obtained numerically by the Hartree Table and the second term has been integrated analytically. The value of energy obtained in this way is

$$E = -0.1369 \frac{e^2}{a_0} = -\, 3.728 \text{ eV}.$$

The Hartree—Fock energy of the valence electron in the $4s$ state is

$$E = -4.341 \text{ eV}.$$

The difference between these two values is the exchange energy

$$E_A = -0.613 \text{ eV}.$$

REFERENCES

1. D. R. HARTREE and W. HARTREE, Proc. Roy. Soc. London A, **166**, 1938.
2. P. GOMBÁS, Theoretica Chimica Acta, **5**, 112, 1966.

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C^{13} NUCLEAR MAGNETIC RESONANCE IN CALCITE*

By

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Infrared absorption spectra [1] of inorganic carbonates and nitrates obtained on single crystals at liquid helium temperature have shown large numbers of bands not attributable to fundamentals or combinations of fundamental frequencies of the anions. Almost all these bands are readily interpreted as summation bands of fundamental frequencies with successive levels of a librating oscillator. The libration is considered to represent a planar torsional oscillation of the anion about the trigonal axis. To obtain additional information on this libration, the C^{13} nuclear magnetic resonance is studied in calcite and the results of the study are reported in this note.

A varian dual purpose NMR spectrometer operating at 10.7 megacycles at a steady magnetic field of 10.000 Gauss. The first derivative of the resonance absorption spectrum was recorded at liquid helium temperature using a specially designed cryostat, described elsewhere [2]. The steady magnetic field was modulated with a small magnetic field oscillating at a frequency of 40 c/s. The amplitude of the modulating field was kept low to avoid artificial broadening effects. The radio frequency power level was kept sufficiently low to avoid saturation of the spin system. The sample of calcite was taken in the form of powder.

The line width (defined as the interval between maximum and minimum slopes of the resonance absorption curve) was found to be 4.50 ± 0.02 Gauss at liquid helium temperature.

In the present studies, the line width of the NMR absorption spectrum is essentially due to nuclear magnetic dipole—dipole interaction since the sample is diamagnetic and the nuclear quadrupole moment of the C^{13} nucleus is zero. The line shape can, in principle, be calculated using the nuclear dipole—dipole interaction formula [3]. The calculation is rather tedious and the analysis of the NMR spectrum becomes much simplified by calculating the second moment (or the root mean square width) of the resonance absorption spectrum. This can be done by graphical integration or numerical integration

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* The experimental part of this research was carried out at the University of Maryland, College Park, Maryland, U.S.A.

and the recording of the first derivative of the resonance absorption spectrum turns out to be advantageous [4]. The spectral width as measured by the second moment is a sensitive function of the inter-nuclear distances and the relative orientations of the interacting nuclear moments. At sufficiently low temperatures, where all molecular and sub-molecular translational and rotational motions have either ceased or slowed down sufficiently not to affect the spectral width, the second moment (ΔH^2) of the rigid lattice configuration can be readily calculated from the Van Vleck's second moment formula [5]

$$(\Delta H^2) = (3/2)\, I(I+1)\, g^2 \beta^2 N^{-1} \sum_{j>k} (3\cos^2 \Theta_{jk} - 1)^2\, r_{jk}^{-6}, \tag{1}$$

where I is the nuclear spin, g the nuclear g factor, β the nuclear magneton, N the number of nuclei in the system whose interactions are considered and over which the sum is taken, r_{jk} the distance between nuclei j and k, and Θ_{jk} is the angle between the internuclear vector r_{jk} and the direction of the applied magnetic field. If the sample is taken in the form of powder, as in the present experiment, the angular factor $(3\cos^2\Theta_{jk}\text{-}1)^2$ is to be replaced by its isotropic numerical average of 4/5 in order to account for the random orientation of the crystallites in the powdered sample. If the powdered sample contains other species of nuclei, besides those at resonance, the Van Vleck's second moment formula becomes

$$\begin{aligned}(\Delta H^2) = {} & (6/5)\, I(I+1)\, g^2 \beta^2 N^{-1} \sum_{j>k} r_{jk}^{-6} + \\ & + (4/15)\, \beta^2 N^{-1} \sum_{j f} I_f (I_f + 1)\, g_f^2\, r_{jf}^{-6},\end{aligned} \tag{2}$$

where I_f is the spin number of nucleus of another species and $g_f \beta I_f$ is its magnetic moment.

If in the sample of the solid, the molecules or atomic groups undergo rapid rotational motion (or libration which is a restricted form of rotation) about one or more axes, the nuclear dipole—dipole interaction will now be a function of time and its time average will in general be small compared with the rigid lattice value. This results in narrowing the resonance absorption spectrum. The second moment formula for powdered samples where there is rotational or vibrational motion is given by [6]

$$\begin{aligned}(\Delta H^2) = {} & (3/10)\, I(I+1)\, g^2 \beta^2 N^{-1} \sum_{j>k} (3\cos^2 \gamma_{jk} - 1)^2\, r_{jk}^{-6} + \\ & + (1/15)\, \beta^2 N^{-1} \sum_{j,f} I_f (I_f + 1)\, g_f^2 (3\cos^2 \gamma_{jf} - 1)^2\, r_{jf}^{-6},\end{aligned} \tag{3}$$

where γ_{jk} is the angle between the axis of rotation and the vector r_{jk} and γ_{jf} has similiar meaning with respect to other species of nuclei.

To apply the method of moments to the present problem, it is necessary to calculate the experimental and theoretical values of the second moment of the resonance absorption spectrum. The experimental value is obtained by graphical or numerical integration of the recorded spectrum and, hence, it is necessary to have a good signal to noise ratio since the second moment can be greatly influenced by the wings of the absorption spectrum where the signal is weak. Since the natural abundance of C^{13} nuclei is only 1%, the signal to noise ratio is rather poor at the wings and hence the calculation of second moment may not be accurate, whereas the line width can be measured accurately. The line width is found to be 4.50 ± 0.02 Gauss at liquid helium temperature. It is, therefore, necessary to calculate the line width from other considerations both for rigid lattice and librating anion configuration of calcite. The line shape in the present case is found to be approximately Gaussian and the ratio of line width to the square root of second moment is about three for Gaussian lines [7]. The theoretical value of the second moment of calcite for rigid lattice can be calculated from formula (2). From the crystal structure of calcite [8], it can be easily seen that important contributions to the second moment arise from oxygen atoms surrounding the carbon atom in the carbonate ion. The other carbon atoms in the lattice are too far apart to make any appreciable contribution since the second moment falls off as r^{-6}, r being the distance between the nuclei; and the Ca^{40} nuclei do not have any magnetic moment. Using the crystal structure data of calcite [8], the second moment for rigid lattice is 5.46 Gauss^2. The line width $\delta H = 3(\Delta H^2)^{1/2}$ and for the present problem it comes out to be 7.02 Gauss which is far in excess of the experimental value of 4.50 Gauss. On the other hand, if libration is assumed to be present about the trigonal axis, it can be seen from geometrical considerations that the angle γ_{jf} of formula (3) is approximately 90° so that the second moment for libration should be reduced by a factor $^1/_4$. The calculated value of the second moment for libration of the anion about the trigonal axis comes out to be approximately 1.36 Gauss^2 and the estimated line width is about 3.06 Gauss. This is much closer to the experimental value of 4.50 Gauss. These approximate considerations support the view that libration could take place about the trigonal axis in calcite at liquid helium temperature. The experiment should be performed with calcite samples enriched in C^{13} and O^{17} to get an accurate value of the experimental second moment so that the libration about the trigonal axis can be established unequivocally.

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REFERENCES

1. R. A. Schroeder, C. E. Weir and E. R. Lippincott, J. Chem. Phys., **36**, 2803, 1962.
2. A. S. Rao, Ph. D. Thesis, University of Maryland, College Park, Md, U. S. A., 1964.
3. G. E. Pake, J. Chem. Phys., **16**, 327, 1948.
4. G. E. Pake and E. M. Purcell, Phys. Rev., **74**, 1184, 1948.
5. J. H. Van Vleck, Phys. Rev., 74, 1168, 1948.
6. H. S. Gutowsky and G. E. Pake, J. Chem. Phys., **18**, 162, 1950.
7. A. Abragam, The Principles of Nuclear Magnetism, Oxford University Press, p. 108, 1961.
8. R. L. Sass, R. Vidal and J. Donohue, Acta. Cryst., **10**, 567, 1958.

CORRIGENDA

Correction to the article
“ON A NEW EXPANSION OF WAVE FUNCTIONS OF 2N-ELECTRON SYSTEMS”

(Acta Physica Academiae Scientiarum Hungaricae, *24*, 301—306, 1968)

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In this paper formula (13) on p. 306 should read

$$\psi_{J1}(1,2)\,\psi_{K1}(3,4)\,\psi_{L1}(5,6)\,\psi_{M1}(7,8) \longrightarrow \varphi_{J\varkappa}(1)\psi_{Kk}(2,3,4)\,\varphi_{L\lambda}(5)\psi_{Mm}(6,7,8), \tag{13}$$

for all $J < L$, $K < M(J,L \neq K,M), \varkappa, k, \lambda$ and m.

and should be placed immediately below formula (12) on p. 305.

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UREY–BRADLEY FORCE CONSTANTS, MEAN AMPLITUDES OF VIBRATION, SHRINKAGE EFFECT AND CORIOLIS CONSTANTS IN IF_5 AND IOF_5

By

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Force constants of the molecules IF_5 and IOF_5 have been calculated using the Urey—Bradley force field. The theory of mean square amplitude matrices is applied to these cases and the parallel and perpendicular mean square amplitudes at 300°K are determined. The perpendicular amplitudes are employed for calculating the BASTIANSEN—MORINO shrinkage effects. The Coriolis constants of these molecules are also evaluated.

Introduction

The molecules IF_5 and IOF_5 possess C_{4v} symmetry. The numbering of atoms and orientation of the principal axes are given in Figs. 1 and 2. The symmetry coordinates and frequencies used in the present study are the same as those given by BEGUN, FLETCHER and SMITH for IF_5 [1] and SMITH and BEGUN for IOF_5 [2].

Urey—Bradley force field

The application of the Urey—Bradley force field gives the following *F*-matrices for the two molecules. The values of the off-diagonal elements will reduce to zero.

For IF_5:

$$F_{11} = K_R + 4(t_0^2 F'_{16} + s_0^2 F_{16}),$$

$$F_{22} = K_r + 4s^2 F_{12} + t_1^2 F'_{16} + s_1^2 F_{16},$$

$$F_{33} = H_\beta - s_0 s_1 F'_{16} = t_0 t_1 F_{16},$$

$$F_{44} = K_r + 4t^2 F'_{12} + t_1^2 F'_{16} + s_1^2 F_{16},$$

$$F_{55} = F_{33},$$

$$F_{66} = H_\alpha - s^2 F'_{22} + t^2 F_{12},$$

$$F_{77} = K_r + 2(t^2 F'_{12} + s^2 F_{12}) + t^2 F'_{16} + s_1^2 F_{16},$$

$$F_{88} = F_{33},$$

$$F_{99} = F_{66},$$

where K and H represent the stretching and bending constants. F and F' are the repulsion constants between the atoms indicated by subscripts attached to F. $F' = -0.1\ F$ as assumed by SHIMANOUCHI. t_0, s_0, etc. are the usual constants employed in the Urey—Bradley field, which depend upon the structural parameters of the molecule.

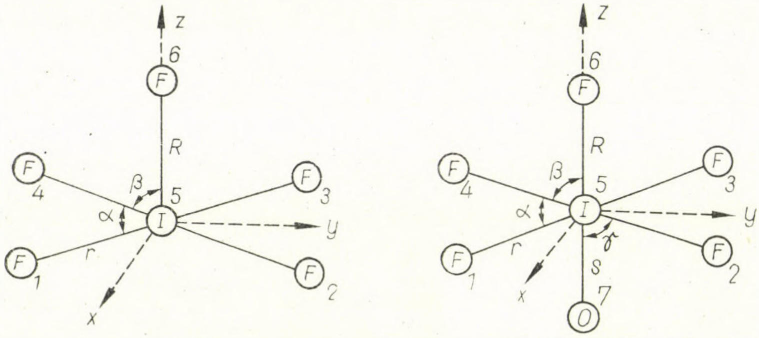

Fig. 1. Internal coordinates, numbering of atoms and orientation of principal axes in IF_5 and IOF_5

For IOF_5:

$$F_{11} = K_S + 4(t_{21}^2\, F_{17}^2 + s_{21}'\, F_{17})\,,$$

$$F_{22} = K_R + 4(t_{01}^2\, F_{16}' + s_{01}^2\, F_{16})\,,$$

$$F_{33} = K_r + 4s_{11}^2\, F_{12} + t_{10}^2\, F_{16}' + s_{10}^2\, F_{16} + t_{12}^2\, F_{17}' + s_{12}^2\, F_{17}\,,$$

$$F_{44} = \frac{1}{2}\,[H_\beta - s_{01}\, s_{10}\, F_{16}' + t_{01}\, t_{10}\, F_{16} + H_\gamma - s_{21}\, s_{12}\, F_{17}' + t_{21}\, t_{12}\, F_{17}]\,,$$

$$F_{55} = K_r + 4t_{11}^2\, F_{12}' + t_{10}^2\, F_{16}' + s_{10}^2\, F_{16} + t_{12}^2\, F_{17}' + s_{12}^2\, F_{17}$$

$$F_{66} = F_{44}\,,$$

$$F_{77} = H_\alpha - s_{11}^2\, F_{12}' + t_{11}^2\, F_{12}\,,$$

$$F_{88} = K_r + 2(t_{11}^2\, F_{12}' + s_{11}^2\, F_{12}) + t_{10}^2\, F_{16}' + s_{10}^2\, F_{16} + t_{12}^2\, F'_{7} + s_{12}^2\, F_{17}\,,$$

$$F_{99} = H_\gamma - s_{21}\, s_{12}\, F_{17}' + t_{21}\, t_{12}\, F_{17}\,,$$

$$F_{10,10} = H_\beta - S_{01}\, S_{10}\, F_{16}' + t_{01}\, t_{10}\, F_{16}\,,$$

$$F_{11,11} = F_{77}\,.$$

Parallel and perpendicular amplitudes

The mean square amplitudes σ_s' for any pair of atoms is obtained by the solution of the secular equation $|\, \Sigma G^{-1} - E\, \Delta\, | = 0^3$ and using the relations between Σ_s' and σ_s' which, in the present case, are as follows:

For IF_5:

$$\sigma_R = \Sigma_{11}\,; \quad \sigma_r = \frac{1}{4}(\Sigma_{22} + \Sigma_{44} + 2\Sigma_{77}),$$

$$\sigma_\alpha = \frac{1}{4}(\Sigma_{66} + 2\Sigma_{99}),$$

$$\sigma_\beta = \frac{1}{4}(\Sigma_{33} + \Sigma_{55} + 2\Sigma_{88}),$$

$$\sigma_{rr} = \frac{1}{4}(\Sigma_{22} - \Sigma_{44})\,; \quad \sigma_{\alpha\alpha} = -\,\Sigma_{bb}/4\,,$$

$$\sigma_{\beta\beta} = \frac{1}{4}(\Sigma_{33} - \Sigma_{55})\,,$$

$$\sigma_{1\cdots 2} = \frac{1}{q^2_{1\cdots 2}}\left[\frac{r^2}{2}(\Sigma_{22} + \Sigma_{44} + 2\Sigma_{77}) + \right.$$
$$\left. + \frac{r^2}{4}(\Sigma_{66} + 2\Sigma_{99}) + \sqrt{2}\, re\, \Sigma_{79})\right],$$

$$\sigma_{1\cdots 6} = \frac{1}{q^2_{1\cdots 6}}\left[\frac{r^2}{4}(\Sigma_{22} + \Sigma_{44} + 2\Sigma_{77}) + R^2\Sigma_{11} + \right.$$
$$+ \frac{Rr}{4}(\Sigma_{33} + \Sigma_{55} + 2\Sigma_{88}) + rR\Sigma_{12} +$$
$$\left. + \frac{1}{2} r(rR)^{1/2}(\Sigma_{45} - \Sigma_{23} + 2\Sigma_{78})\, R(rR)^{1/2}\Sigma_{13}\right],$$

where $q_{1..j}$ represents the nonbonded distance between atoms i and j.

For IOF_5:

$$\sigma_S = \Sigma_{11}\,; \quad \sigma_R = \Sigma_{22}\,,$$

$$\sigma_r = \frac{1}{4}(\Sigma_{33} + \Sigma_{55} + 2\Sigma_{88}),$$

$$\sigma_\alpha = \frac{1}{4}(\Sigma_{77} + 2\Sigma_{11,11}),$$

$$\sigma_\beta = \frac{1}{8}(\Sigma_{44} + \Sigma_{66} + 4\Sigma_{10,10}),$$

$$\sigma_\gamma = \frac{1}{8}(\Sigma^4_4 + \Sigma_{66} + 4\Sigma_{99}),$$

$$\sigma_{rr} = \frac{1}{4}(\Sigma_{33} - \Sigma_{55})\,; \quad \sigma_{\alpha\alpha} = -\frac{1}{4}\Sigma_{77}\,,$$

$$\sigma_{\gamma\gamma} = \sigma_{\beta\beta} = \frac{1}{8}({\Sigma^4}_4 - \Sigma_{65}),$$

$$\sigma_{1\cdots 2} = \frac{1}{4}(\Sigma_{33} + \Sigma_{55} + 2\Sigma_{88}) + \frac{1}{8}(\Sigma_{77} + 2\Sigma_{11,11}) + \frac{1}{\sqrt{2}}\Sigma_{8,11},$$

$$\sigma_{1\cdot\cdot 6} = \frac{1}{q^2_{1\cdot\cdot 6}}\Big[r^2\sigma_r + R^2\sigma_R + Rr\,\sigma_\beta + rR\,\Sigma_{23} + {} + \frac{r(rR)^{1/2}}{2\sqrt{2}}(\Sigma_{34} + \Pi_{56} + 2\sqrt{2}\,\Sigma_{8,10}) + \frac{R(rR)^{1/2}}{\sqrt{2}}\Sigma_{24}\Big],$$

$$\sigma_{1\cdots 7} = \frac{1}{q^2_{1\cdot\cdot 7}}\Big[r^2\sigma_r + S^2\sigma_S + Sr\,\sigma_\gamma + S + \Sigma_{13} - {} - \frac{r(rS)^{1/2}}{2\sqrt{2}}(\Sigma_{34} + \Sigma_{56} - 2\sqrt{2}\,\Sigma_{89}) - \frac{S(rS)^{1/2}}{\sqrt{2}}\Sigma_{14}\Big].$$

The generalised mean square amplitudes which include parallel and perpendicular mean square amplitudes are evaluated by the method suggested by Morino and Hirota [4]. The symmetry coordinates S are related to the Cartesian displacement coordinates X by the transformation $S = BX$. An inverse transformation gives $X = AS$ with $A = \mu\,\tilde{B}G^{-1}$, where μ is the reciprocal of the masses of the atoms and G is the kinetic energy matrix. Evaluation of X yields the parallel mean square displacements $\langle(\Delta z)^2\rangle$ and the perpendicular mean square displacements $\langle(\Delta x)^2\rangle$ and $\langle(\Delta y)^2\rangle$ in terms of Σ elements.

Shrinkage effects

The shrinkage effect in molecules observed from electron diffraction studies by Bastiansen et al. [5] may be calculated spectroscopically as has been pointed out by Morino [6]. In the case of IF_5 the shrinkage occurs in the linear bond (1—3) or (2—4) and in the nonbonded distance between any two of the atoms 1, 2, 3 and 4. In the case of IOF_5 there is additionally the shrinkage along the axial bond 6—7. If δ represents the shrinkage then

$$-\delta_{13} = K_{13} - K_{15} - K_{35},$$

$$-\delta_{1\cdots 2} = K_{12} - \sqrt{2}\,K_{15},$$

$$-\delta_{67} = K_{67} - K_{65} - K_{57},$$

where

$$K = \frac{\langle(\Delta x)^2\rangle + \langle(\Delta y)^2\rangle}{2r_e}\cdot\langle(\Delta x)^2\rangle \text{ and } \langle(\Delta y)^2\rangle$$

are known from previous calculations for each bonded or nonbonded distance and r_e is the corresponding equilibrium distance. The results are given in Table IV.

Table I

Force constants of IF_5 and IOF_5 (md/Å)

Molecule	GVFF [1] and [2]		UBF	
IF_5	f_r	3.64	K_r	3.706
	f_R	4.68	K_R	4.679
	f_α	0.17	H_α	0.188
	f_β	0.63	H_β	0.476
IOF_5	f_r	4.42	K_r	4.483
	f_R	4.60	K_R	4.601
	f_S	6.99	K_S	6.990
	f_α	0.20	H_α	0.196
	f_β	0.55	H_β	0.449
	f_γ	0.44	H_γ	0.367

Table II

Mean square amplitudes of vibration (in $Å^2$) of IF_5 and IOF_5

	IF_5	IOF_5
σ_r	0.003821	0.001677
σ_R	0.001368	0.001368
σ_S	. .	0.001560
σ_α	0.017573	0.015460
σ_β	0.005084	0.007230
σ_γ	. .	0.011338
σ_{rr}	0.000075	0.000017
$\sigma_{\alpha\alpha}$	—0.005643	—0.004655
$\sigma_{\beta\beta}$	0.000000	—0.000180
$\sigma_{\gamma\gamma}$	—	—0.000180
$\sigma_{1\ldots2}$	0.012607	0.009028
$\sigma_{1\ldots6}$	0.005208	0.005036
$\sigma_{1\ldots7}$	. .	0.006910

Coriolis constants

The Coriolis coupling constants if IF_5 and IOF_5 for the $e \times e$ coupling are calculated by the method introduced by **Meal** and **Polo** [7]. The C^z elements derived in the present cases are as follows:

IF$_5$:

$$C^z_{77} = -2\mu_1\,;\quad C^z_{88} = -(\mu_I + \mu_F)/2E^2\,,$$
$$C^z_{99} = -4\mu_I\,;\quad C^z_{78} = \mu_I/E\,,$$
$$C^z_{79} = \sqrt{1}\,(\mu_F + 2\mu_I);\quad C_{89} = -\sqrt{2}\,\mu_I/E\,.$$
$$C^z_{88} = -2\mu_I\,;\quad C^z_{99} = -(\mu_I + \mu_0)/2H^2\,,$$
$$C^z_{10,10} = -(\mu_I + \mu_F)/2E^2;\quad C^z_{11,11} = -4\mu_I\,,$$
$$C^z_{89} = \mu_I/H\,;\quad C^z_{8,10} = \mu_I/E\,,$$
$$C^z_{8,11} = \sqrt{2}\,(\mu_F + 2\mu_I);\quad C^z_{9,10} = -\mu_I/2EH\,,$$
$$C^z_{9,11} = -\sqrt{2}\,\mu_I/H;\quad C^z_{10,11} = -\sqrt{2}\,\mu_I/E\,,$$

where

$$E = \frac{1}{2}(R/r)^{1/2}\,,\quad H = \frac{1}{2}(S/r)^2\,.$$

From the values of the L-matrix elements obtained on the basis of the force constants of a general force field, the Coriolis constants are calculated from the formula $\zeta = L^{-1}\tilde{L}C^{-1}$.

From the general formula obtained by LORD and MERRIFIELD [8] for the zeta sum of molecules possessing C_{4v} symmetry one gets the zeta sum as $2\,I_A/2I_B$ for IF$_5$ and $2\,I_A/2I_B + 1$ for IOF$_5$.

Table III

Generalised mean-square amplitudes of IF$_5$ and IOF$_5$ (Å^2)

Atom pair/Bond	$\langle(\Delta z)^2\rangle$		$\langle(\Delta x)^2\rangle$		$\langle(\Delta y)^2\rangle$	
	IF$_5$	IOF$_5$	IF$_5$	IOF$_5$	IF$_5$	IOF$_5$
5—1 (I—F_1)	0.003821	0.001677	0.006840	0.006464	0.003379	0.006194
1...2 ($F_1 \ldots F_2$)	0.012668	0.009028	0.009718	0.006686	0.006755	0.006847
5—6 (I—F_6)	0.001368	0.001368	0.001015	0.002894	0.001015	0.002894
1...6 ($F_1 \ldots F_6$)	0.005208	0.005036	0.004117	0.003920	0.007762	0.009096
5—7 (I—0)	—	0.001570	—	0.005131	—	0.005131
1...7 ($F_1 \ldots 0$)	—	0.006910	—	0.004866	—	0.011711

Atom pair/Bond	$\langle(\Delta x)\ (\Delta y)\rangle$		$\langle(\Delta x)\ (\Delta z)\rangle$		$\langle(\Delta y)\ (\Delta z)\rangle$	
	IF$_5$	IF$_5$	IF$_5$	IOF$_5$	IF$_5$	IOF$_5$
5—1 (I—F_1)	0.000000	0.000000	0.000000	0.000000	—0.000149	0.000044
1...2 ($F_1 \ldots F_2$)	—0.000717	0.000045	0.000000	0.000000	0.000000	0.000000
5—6 (I—F_6)	0.000000	0.000000	0.000000	0.000000	0.000000	0.000000
1...6 ($F_1 \ldots F_6$)	0.000000	0.000000	—0.000105	—0.000209	0.000000	0.000000
5—7 (I—0)	—	0.000000	—	0.000000	—	0.000000
1...7 ($F_1 \ldots 0$)	—	—0.000353	—	0.003139	—	0.000684

I_A and I_B are the moments of inertia about the symmetry axis and any other principal axis of the molecule, respectively.

The values of the Coriolis constants obtained in this investigation are given in Table V. Due consideration of the signs of the C elements has been taken in fixing the signs of the zeta values. It can be seen from the Table that the values very nearly satisfy the sum rule for these molecules.

Table IV

Shrinkage effects (Å)

δ_{ij}	IF_5	IOF_5
$\delta_{1\cdots 2}$	0.00094	0.00105
$\delta_{1\cdots 3}$	0.00462	0.00415
$\delta_{6\cdots 7}$	—	0.00357

Table V

Coriolis coupling constants of IF_5 and IOF_5

	IF_5	IOF_5	
ζ_{77}	0.2941	ζ_{88}	0.3085
ζ_{88}	0.6630	ζ_{99}	0.5780
ζ_{99}	-0.2455	$\zeta_{10,10}$	0.8981
ζ_{78}	0.2221	$\zeta_{11,11}$	-0.1808
ζ_{79}	0.9370	ζ_{89}	-0.0279
ζ_{89}	-0.1783	$\zeta_{8,10}$	-0.9034
		$\zeta_{8,11}$	-0.3175
		$\zeta_{9,10}$	0.0979
		$\zeta_{9,11}$	-0.0771
		$\zeta_{10,11}$	-0.3377

Results and discussion

The structural parameters used in the present study are: $r = 1.86$Å, $R = 1.75$Å, and $\alpha = \beta = 90°$ in both molecules. Also $S = 1.70$Å, $\gamma = 90°$ in IOF_5. Normal coordinate analysis on the basis of general valence force field has been carried out by **Begun** et al. [1] for IF_5 and by **Smith** and **Begun** [2] for IOF_5. The frequencies reported by them are employed in this investigation. The present values of the Urey—Bradley force constants are given in Table I along with the general valence force constants for comparison. As the above authors have noted in their analysis some of the F-elements, although identical, have slightly different values in different species. This arises due to some unknown interaction term which should be included in the force field. Thus, in the present study, in IF_5, $F_{33} = F_{55} = F_{88}$ and $F_{66} = F_{99}$; and in IOF_5, $F_{33} = F_{55} = F_{88}$, $F_{44} = F_{66}$ and $F_{77} = F_{11,11}$. But the secular equations yield slightly different values. In these cases, either the average of the values or, the most reasonable one out of them, is chosen. It may be noted that according to the Urey—Bradley model, forces of interaction are not explicitly obtained in both molecules. It is possible that they are very weak due to the bonds being mutually perpendicular. However, some interaction force constants have been reported in the general valence treatment.

The notable feature in the mean amplitude of vibration (Tables II and III) is that the stretching amplitude of the equatorial bond is considerably less in IOF_5 as compared to IF_5. The axial I—F bond amplitude is the same in both molecules.

REFERENCES

1. G. M. Begun, W. H. Fletcher and D. F. Smith, J. Chem. Phys., **42**, 2236, 1965.
2. D. F. Smith and G. M. Begun, J. Chem. Phys., **43**, 2001, 1965.
3. S. J. Cyvin, Acta Polytechnica Scandinavica, Ph. 6.279/1960.
4. Y. Morino and E. Hirota, J. Chem. Phys., **23**, 737, 1955.
5. A. Almenningen, O. Bastiansen and T. Munthe-Kaas, Acta Chem. Scand., **10**, 261, 1956.
6. Y. Morino, Acta Cryst., **13**, 1107, 1960.
7. J. H. Meal and S. R. Polo, J. Chem. Phys., **24**, 1119, 1956.
8. R. C. Lord and R. E. Merrifield, J. Chem. Phys., **20**, 1348, 1952.

СИЛОВЫЕ ПОСТОЯННЫЕ УРИ—БРЭДЛИ, СРЕДНИЕ АМПЛИТУДЫ ВИБРАЦИИ, СТЯГИВАЮЩИЙ ЭФФЕКТ И ПОСТОЯННЫЕ КОРИОЛИСА В IF_5 и IOF_5

К. ФЕНКАТЕСВАРЛУ и Ц. ПУРУШОТАМАН

Резюме

Используя силовое поле Ури—Брэдли, в работе вычисляются силовые постоянные молекул IF_5 и IOF_5. К данному случаю применяется матричная теория средних квадратичных амплитуд и при 300°К определяются параллельные и перпендикулярные средние квадратичные амплитуды. Перпендикулярные амплитуды используются для определения стягивающего эффекта Бастиансена—Морино. Далее оцениваются постоянные Кориолиса этих молекул.

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ОБ ЭФФЕКТАХ ТОЖДЕСТВЕННОСТИ ЧАСТИЦ В ЯДЕРНЫХ РЕАКЦИЯХ

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ИНСТИТУТ ЯДЕРНОЙ ФИЗИКИ МОСКОВСКОГО ГОСУДАРСТВЕННОГО УНИВЕРСИТЕТА,
МОСКВА, СССР

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Выведены простые формулы позволяющие учитывать обменные эффекты в амплитуде ядерной реакции $T(a, b)F$ в общем случае.

1. Хотя рассмотрению обменных эффектов в ядерных реакциях посвящено большое число работ (см. [1, 2]) и содержащиеся в них ссылки), конкретные результаты, полученные в этих работах, применимы лишь в случае наиболее простых реакций (p, p'), (n, p) и (d, p).** Ни в одной из работ нет простых формул, позволяющих учитывать обменные эффекты в амплитуде ядерной реакции $T(a, b)\,F$ в общем случае. В настоящей работе, исходя из результатов Экштейна [3], мы вывели такие формулы и в качестве примера применили их к реакции (d, α).

2. Согласно Экштейну [3], амплитуда реакции $i \to f$ M_{fi}, связанная с матричным элементом S-матрицы соотношением $(\hbar = c = 1)$

$$S_{fi} = \delta_{fi} - i\,(2\pi)^4\,\delta\left(\sum_k p_k^{(i)} - \sum_l p_l^{(f)}\right) M_{fi}\,, \tag{1}$$

дается формулами

$$M_{fi} = \langle (H - E)\,\hat{\Phi}_f \,|\, \hat{\Psi}_i^{(+)} \rangle \tag{2a}$$

$$= \langle \hat{\Psi}_f^{(-)} \quad H - E)\,\hat{\Phi}_i \rangle\,, \tag{2b}$$

где H — полный гамильтониан системы, E — полная энергия, $\hat{\Phi}_i$ и $\hat{\Phi}_f$ — полностью антисимметричные по переменным тождественных частиц норми-

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[*] Постоянное место работы: Центральный Институт Физических Исследований АН ВНР.

[**] Заметим, что выражение (A.32) для амплитуды реакции, приведенное в книге Тобокмана [2], справедливо лишь в том случае, когда внутренние волновые функции ядер представляются в виде произведения одночастичных ортонормированных волновых функций. В общем случае оно отличается от приведенных ниже выражений (12) на постоянный множитель, связанный с нормировкой волновых функций.

рованные «базисные» волновые функции «групп каналов» i и f (определение «групп каналов» см. в [3]),

$$\begin{aligned} \hat{\Psi}_i^{(+)} &= [\,1 + (E - H + i\eta)^{-1}(H - E)\,]\hat{\Phi}_i\,, \\ \hat{\Psi}_f^{(-)} &= [\,1 + (E - H - i\eta)^{-1}(H - E)]\,\hat{\Phi}_f\,, \qquad (3 \\ \eta &\to +0. \end{aligned}$$

Дифференциальное сечение реакции $i \to f$ с двумя частицами в начальном и конечном состояниях выражается через M_{fi} с помощью формулы

$$\frac{d\sigma}{d\Omega} = \frac{m_i\, m_f}{4\pi^2}\; \frac{k_f}{k_i}\, |M_{fi}|^2\,,$$

где $m_i(m_f)$ и $k_i(k_f)$ — приведенная масса и импульс относительного движения в начальном (конечном) состоянии.

Разобьем гамильтониан H обычным образом на две части $H = H_i + V_i = H_f + V_f$ и введем функции Φ_i и Φ_f, удовлетворяющие уравнениям

$$(H_i - E)\,\Phi_i = 0, \quad (H_f - E)\,\Phi_f = 0, \tag{4}$$

и представляющие собой произведение нормированных антисимметризованных внутренних волновых функций ядер в начальном и конечном состояниях, соответственно, и плоских волн с единичной амплитудой, описывающих их относительное движение.

Наша цель состоит в построении из перестановочных операторов P_n операторов P_i и P_f таких, что

$$\hat{\Phi}_i = C_i\, P_i\, \Phi_i\,, \;\; \hat{\Phi}_j = C_f P_f \Phi_f \tag{5}$$

(C_i и C_f — нормировочные коэффициенты). Используя эти соотношения мы преобразуем формулы (2) к виду, удобному для практических расчетов.

3. Рассмотрим реакции

$$a + T \to b + F.$$

Для определенности будем работать в формализме изоспина, то-есть будем считать, что имеется только один сорт частиц. Предположим, что ядра a и T состоят из a и $(A - a)$ нуклонов и обозначим через i совокупность пространственных, спиновой и изоспиновой координат i-го нуклона. Предположим, далее, что внутренние волновые функции ядер a и T зависят от совокупностей переменных $(1, 2, \ldots, a)$ и $(a + 1,\; a + 2, \ldots, A)$, соответственно.

Оператор P_{aT}, производящий антисимметризацию по переменным всех нуклонов $(1, 2, \ldots, A)$ имеет вид (см., например [4], гл. 6)

$$P_{a+T} = \sum_n \sigma_n P_n \,, \tag{6}$$

где P_n — оператор перестановки координат нуклонов, $\sigma_n = 1(-1)$ для четной (нечетной) перестановки и суммирование идет по всем $A!$ перестановкам координат $(1, 2, \ldots, A)$. Определим аналогично операторы P_a и P_T, производящие антисимметризацию по переменным $(1, 2, \ldots, a)$ и $(a+1, a+2), \ldots, A)$ соответственно. Тогда оператор P_{a+T} может быть представлен в виде

$$P_{a+T} = P_{aT} \cdot P_a \cdot P_T \,, \tag{7}$$

где

$$P_{aT} = 1 - \sum_{i,\alpha} P_{i\alpha} + \sum_{\substack{i< j\\ \alpha<\beta}} P_{i\alpha} P_{j\beta} - \sum_{\substack{i<j<k\\ \alpha<\beta<\gamma}} P_{i\alpha} P_{j\beta} P_{k\gamma} + \ldots \,. \tag{8}$$

$P_{i\alpha}$ — оператор транспозиции, переставляющий переменные пары нуклонов i и α, причем

$$i, j, k = 1, 2, \ldots, a \,; \quad \alpha, \ \beta, \ \gamma = a+1, \quad a+2, \ldots, \ A,$$

то-есть оператор P_{aT} осуществляет антисимметризацию по переменным нуклонов, входящих в состав разных ядер a и T. В справедливости соотношений (7), (8) легко убедиться путем проверки, что в произведении $P_{aT} \cdot P_a \cdot P_T$ не встречаются дважды одна и та же перестановка и общее число членов равно $A!$ Заметим, что в (7) порядок операторов несущественен, то-есть

$$P_{a+T} = P_{aT} \cdot P_a \cdot P_T = P_a \cdot P_T \cdot P_{aT} \,. \tag{7'}$$

В силу эрмитовости операторов P_n оператор P_{aT} также эрмитов: $P_{aT}^{+} = P_{aT}$.

Вводя операторы P_b и P_F, производящие антисимметризацию по переменным $(1, 2, \ldots, b)$ и $(b+1, b+2, \ldots, A)$ нуклонов ядер b и F соответственно, можем представить оператор P_{a+T} в виде

$$P_{a+T} = P_{bF} \cdot P_b \cdot P_F \,, \tag{7''}$$

где оператор P_{bF} определяется соотношением, аналогичным (8):

$$P_{bF} = 1 - \sum_{i,\alpha} P_{i\alpha} + \sum_{\substack{i<j\\ \alpha<\beta}} P_{i\alpha} P_{j\beta} - \ldots ; \quad \begin{array}{l} i, \ j, \ldots = 1, 2, \ldots, b \,; \\ \alpha, \ \beta, \ldots = b+1, b+2, \ldots, A. \end{array} \tag{8'}$$

Формулы (8) и (8′) определяют одну из возможных форм искомых операторов P_i и P_f в (5) для реакций с двумя частицами в начальном и конечном состояниях. Используя вместо плоских волн относительного движения в волновых функциях Φ_i и Φ_f нормированные на единицу квазимонохроматические пакеты, нетрудно показать, что правильно нормированные полностью антисимметричные волновые функции $\hat{\Phi}_i$ и $\hat{\Phi}_f$ имеют вид

$$\hat{\Phi}_i = N_{aT}^{-1} \quad P_{aT}\Phi_i\,, \qquad \hat{\Phi}_f = N_{bF}^{-1/2} P_{bF} \cdot \Phi_f\,, \tag{9}$$

где

$$N_{aT} = \binom{A}{a}, \qquad N_{bF} = \binom{A}{b}\,. \tag{10}$$

Заметим, что иногда при выполнении антисимметризации удобно пользоваться формулой

$$P_{aT} \cdot P_a = \left(1 - \sum_{\alpha=2}^{A} P_{1\alpha}\right)\left(1 - \sum_{\beta=3}^{A} P_{2\beta}\right) \cdot \ldots \cdot \left(1 - \sum_{\omega=a+1}^{A} P_{a\omega}\right), \tag{11}$$

которая получается из сравнения соотношения (7) с соотношением, полученным в результате a-кратного тождества

$$P_{a+T} = \left(1 - \sum_{\alpha=2}^{A} P_{1\alpha}\right) P'_{a+T}\,,$$

где P_{a+T} и P'_{a+T} — антисимметризаторы по переменным $(1, 2, \ldots, A)$ и $(2, 3, \ldots, A)$, соответственно.

4. Используя соотношения (3), (9), (10) и учитывая, что гамильтониан H симметричен относительно перестановок координат частиц, из (2) получаем четыре эквивалентных выражения для матричного элемента реакции $T(a, b)\,F$ с учетом обменных эффектов:

$$M_{fi} = (N_f/N_i)^{1/2} \langle \Phi_f | V_f P_i | \Psi_i^{(+)} \rangle \tag{12a}$$

$$= (N_i/N_f)^{1/2} \langle \Phi_f | V_f P_f | \Psi_i^{(+)} \rangle \tag{12b}$$

$$= (N_f/N_i)^{1/2} \langle \Psi_i^{(-)} | P_i V_i | \Phi_i \rangle \tag{12c}$$

$$= (N_i/N_f)^{1/2} \langle \Psi_f^{(-)} | P_f V_i | \Phi_i \rangle\,, \tag{12d}$$

где

$$N_i = N_{aT}\,, \quad N_f = N_{bF}\,, \quad P_i = P_{aT}\,, \quad P_f = P_{bF}$$

и волновые функции $\Psi_i^{(+)}$ и $\Psi_f^{(-)}$ даются выражениями

$$\Psi_i^{(+)} = [1 + (E - H + i\eta)^{-1} V_i]\,\Phi_i\,,$$
$$\Psi_f^{(-)} = [1 + (E - H - i\eta)^{-1} V_f]\,\Phi_f\,. \tag{13}$$

Подставляя в (12) конкретные выражения (8) и (8′) для операторов P_i и P_f и собирая одинаковые члены, возникающие благодаря антисимметричности функций Φ_i и $\Psi_i^{(+)}$ (Φ_f и $\Psi_f^{(-)}$) повнутренним координатам ядер a и T (b и F), можно получить удобные формулы для практических расчетов. Предположим, что $a \leqslant b$. Тогда оператор $P_i = P_{aT}$ в матричном элементе (12а) можно заменить на оператор

$$P_i \to \binom{A-b}{0}\binom{b}{a-0} - \binom{A-b}{1}\binom{b}{a-1} P_{1A} + \binom{A-b}{2}\binom{b}{a-2} P_{1A}\, P_{2(A-1)} - \\ - \ldots + (-1)^n \binom{A-b}{n}\binom{b}{a-n} P_{1A}\, P_{2(A-1)} \cdot \ldots \cdot P_{n(A-n+1)}\,, \qquad (14)$$

$$n = \min(a,\ A-b), \quad a \leq b\,,$$

то-есть

$$M_{fi} = \left[\binom{A}{b}\Big/\binom{A}{a}\right]^{1/2} \Big\langle \Phi_f(1,2,\ldots,\ a,\ldots,\ b\,|b+1,\ldots,A)| \cdot \\ \cdot V_f(1,2,\ldots,b\,|b+1,\ldots,A)\left[\binom{A-b}{0}\binom{b}{a-0} - \binom{A-b}{1}\binom{b}{a-1} P_{1A} + \right. \\ \left. + \ldots + (-1)^n \binom{A-b}{n}\binom{b}{a-n} P_{1A}\, P_{2(A-1)} \cdot \ldots \cdot P_{n(A-n+1)}\right] \cdot \qquad (15) \\ \cdot |\Psi_i^{(+)}(1,2,\ldots,a|\,a+1,\ldots,b,\ldots,A)\Big\rangle\,.$$

Здесь волновые функции антисимметричны, а потенциал V_f симметричен по отдельным совокупностям переменных, разделенным вертикальной чертой. Аналогичные выражения можно написать для матричных элементов (12*b*), (12*c*) и (12*d*).

Если мы работаем не в формализме изоспина, то-есть имеется два сорта частиц, то полученные выше формулы надо применять отдельно для протонов и отдельно для нейтронов.

5. В качестве примера рассмотрим реакцию $T(d, \alpha) F$.

а) Формализм изоспина. В этом случае $a = 2$, $b = 4$ и с помощью формулы (15) получаем

$$M_{fi} = \left[\frac{(A-2)(A-3)}{3\cdot 4}\right]^{1/2} \Big\langle \Phi_f(1,2,3,4\,|\,5,\ldots,A)\Big|\, V_f(1,2,3,4\,|\,5,\ldots,A) \cdot \\ \cdot \left[6 - 4(A-4)\, P_{1A} + \frac{1}{2}(A-4)(A-5)\, P_{1A}\, P_{2(A-1)}\right]\Big| \cdot \qquad (16) \\ \cdot \Psi_i^{(+)}(1,2\,|\,3,4,5,\ldots,A)\Big\rangle.$$

в) В случае антисимметризации отдельно по протонам и отдельно по нейтронам имеем $a_p = 1$, $b_p = 2$ и $a_n = 1$, $b_n = 2$, соответственно. В этом случае с помощью формулы (15) получаем

$$\begin{aligned} M_{fi} = \frac{1}{2}\left[(N-1)(Z-1)\right]^{1/2} \langle \Phi_f (n_1, n_2 \,|\, p_1, p_2 \,|\, n_3, \ldots, n_N \,|\, p_3, \ldots, p_z)| \cdot \\ \cdot V_f (n_1, n_2 \,|\, p_1, p_2 \,|\, n_3, \ldots, n_N \,|\, p_3, \ldots, p_Z) \left[2 - (N-2) P^{(n)}_{1N}\right] \cdot \\ \cdot \left[2 - (Z-2) P^{(p)}_{1Z}\right] | \Psi^{(+)}_i (n_1 \,|\, p_1 \,|\, n_2, n_3, \ldots, n_N | \, p_2, p_3, \ldots, p_Z) \rangle , \end{aligned} \tag{17}$$

где $n_i(p_i)$ — совокупность пространственных и спиновой координат i-го нейтрона (протона), $N(Z)$ — полное число нейтронов (протонов) в системе $d + T$, операторы траспозиций $P^{(n)}_{1N}$ и $P^{(p)}_{1Z}$ переставляют координаты нейтронов и протонов, соответственно.

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ЛИТЕРАТУРА

1. F. S. Levin, Nucl. Phys., **46,** 275, 1963; S. Edwards, Nucl. Phys., **47,** 652, 1963; M. Bolsterli, Phys. Rev., **131,** 883, 1963.
2. W. Tobocman, Theory of direct nuclear reactions, Oxford Univ. Press, 1961.
3. H. Ekstein, Phys. Rev., **101,** 880, 1956.
4. С. Швебер, Введение в релятивистскую квантовую теорию поля, ИИЛ, Москва, 1963.

ON THE EFFECT OF PARTICLE EXCHANGE IN NUCLEAR REACTIONS

By

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Starting from the general formulas of Ekstein which take into account the effect of antisymmetrization, practically useful expressions are given in this paper for the amplitudes of nuclear reactions containing two nuclei both in the initial and final states.

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ЗАМЕЧАНИЕ ОТНОСИТЕЛЬНО ПРИНЦИПА ОСЛАБЛЕНИЯ КОРРЕЛЯЦИИ

С. А. СМОЛЯНСКИЙ

КАФЕДРА ТЕОРЕТИЧЕСКОЙ ФИЗИКИ САРАТОВСКОГО УНИВЕРСИТЕТА, САРАТОВ, СССР

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Показано, что принцип ослабления корреляции является следствием предположения об эргодичности системы. Это позволяет указать границы применимости принципа ослабления корреляции, которые оказываются несколько более широкими, чем обычно предполагаемые.

Известный в статистической механике необратимых процессов принцип ослабления корреляции Боголюбова [1] вводится из интуитивных соображений. Ниже показано, что принцип ослабления корреляции можно рассматривать как следствие из предположения об эргодичности системы. Это позволяет фиксировать границы применимости принципа. Ниже для определенности будет рассмотрен лишь случай пространственно-однородной системы бесконечного числа одинаковых частиц, поведение которых описывается уравнениями классической механики с функцией Гамильтона, не зависящей от времени. Необходимое обобщение может быть легко выполнено.

Систему с бесконечным числом степеней свободы можно описывать с помощью предельного значения N — частичной функции распределения $F_N(t, \{q\})$, где $q \equiv (x, p)$, в N/V — пределе. Запишем уравнение движения для $F_N(t, \{q\})$ через оператор Лиувилля

$$\frac{\partial F_N(t, \{q\})}{\partial t} = -i L_N F_N(t, \{q\}) . \tag{1}$$

Пусть $L_N = L_N^0 + L_N^1$, где L_N^0 — оператор Лиувилля N невзаимодействующих частиц:

$$L_N^0 = -i \sum_{i=1}^{N} \frac{p_i}{m} \frac{\partial}{\partial x_i} . \tag{2}$$

Определим далее s — частичную функцию распределения ($s \leqslant N$):

$$F_s(t, \{q\}) = \int F_N(t, \{q\}) \prod_{i=s+1}^{N} dq_i \tag{3}$$

и перейдем к представлению взаимодействия (см., напр. [2]):

$$F_N(t,\{q\}) = e^{-iL_N^0}\, F_N^{b3}(t,\{q\}) \equiv S_t^{(N)}\, F_N^{b3}(t,\{q\}),$$
$$F_N^{b3}(t,\{q\}) = S_{-t}^{(N)}\, F_N(t,\{q\}) = S_{-t}^{(N)}\, e^{-iL_N t}\, F_N(0,\{q\}). \tag{4}$$

В формуле (3) выполним N/V — предельный переход:

$$F'_з(t,\{q\}) = \lim_{V,N\leftarrow\infty} \int F_N(t,\{q\}) \prod_{i=s+1}^{N} dq_i\,, \tag{5}$$

где F'_s есть N/V — предел функции F_s. Оператор сдвига $S_t^{(N)}$, определяемый соотнощением (4), запищем в следующем виде: $S_t^{(N)} = S_t^{(S)} \cdot S_t^{(N-s)}$ и учтем, что из (4) и (5) следует

$$F'_s(t,\{q\}) = \lim_{V,N\to\infty} S_i^{(s)}\, F_s^{b3}(t,\{q\}) = S_t^{(s)}\, F_s'^{b3}(t,\{q\})\,. \tag{6}$$

Здесь было использовано то обстоятельство, что оператор $S_t^{(N-s)}$, действующий на переменные, по которым производится интегрирование, оставляет инвариантным несобственный интеграл в N/V — пределе:

$$\int_{-\infty}^{\infty} S_t^{(n)} f(t;q_1,\ldots,q_n)\, dQ_n = \int_{-\infty}^{\infty} f(t;q_1,\ldots,q_n)\, dQ_n\,.$$

Заметим, что согласно предположению об эргодичности системы один из пределов $\lim\limits_{t\to\pm\infty} F'_s(t,\{q\})$ существует. Но тогда существует и соответствующий предел $\lim\limits_{t\to\pm\infty} F_s'^{b3}(t,\{q\})$, поскольку согласно определению оператор $S_t^{(s)}$ есть оператор сдвига, действующий в $\{x\}$ — подпространстве фазового пространства [1]. Последний предел, однако, нетрудно вычислить, выполнив преобразование Лапласа над $F_s'^{b3}(t,\{q\})$ по формуле

$$\varphi_s(\xi) = \xi \int_0^{\infty} e^{-\xi t}\, F_s'^{b3}(t,\{q\})\, dt \tag{7}$$

и воспользовавщись теоремой Абеля [3]:

$$\lim_{t\to\infty} F_s'^{b3}(t,\{q\}) - \lim_{\xi\to 0} \Phi_s(\xi)\,. \tag{8}$$

Аналогично можно вычислить и предельное значение $F_s'^{b3}(t,\{q\})$ при $t \to -\infty$.

Чтобы иметь возможность провести формальное вычисление интеграла (7), воспользуемся некоторыми спектральными свойствами оператора Лиувилля [4]. Пусть

$$L_N\, \psi_{\lambda_N} = \lambda_N\, \psi_{\lambda_N}\,, \tag{9}$$

где ψ_{λ_N} и λ_N — собственные функции и соответствующие собственные значения оператора L_N. За редким исключением спектр оператора Лиувилля является непрерывным и неограниченным [5]. Пользуясь обычным предположением о полноте системы $\{\psi_{\lambda_N}\}$ [4], разложим $F_N(t, \{q\})$ в ряд по собственным функциям оператора L_N:

$$F_N\big(t, \{q\}\big) = \int C_{\lambda_N}(t)\, \psi_{\lambda_N}\big(\{q\}\big)\, d\lambda_N\,, \tag{10}$$

где интегрирование проводится в смысле Стилтьеса. В случае, если оператор L_N имеет лишь непрерывный спектр, λ_N не зависит от N и интегрирование проводится в обычном смысле. С помощью (1) находим:

$$F_N\big(t, \{q\}\big) = \int C_{\lambda_N}\big(\{q\}\big)\, e^{-i\lambda_N t}\, d\lambda_N\,, \tag{11}$$

где $C_{\lambda_N}(\{q\}) = C_{\lambda_N}(0)\, \psi_{\lambda_N}(\{q\})$. Теперь в (7) можно выполнить формальное интегрирование по t. Прежде всего с помощью (5), (6) и (11) перепишем формулу (7) в следующем виде:

$$\Phi_s(\xi) = \xi \int_0^\infty dt \,\Big\{ e^{-\xi t}\, S_{-t}^{(s)} \lim_{V,N\to\infty} \int \Big[\int C_{\lambda_N}\big(\{q\}\big)\, e^{-i\lambda_N t}\, d\lambda_N \Big] \prod_{i=s+1}^{N} dq_i \tag{12}$$

(заметим, что пределы $\lim\limits_{t\to\infty}$ и $\lim\limits_{V,N\to\infty}$ переставлять нельзя). Пусть

$$\int C_{\lambda_N}(\{q\}) \prod_{i=s+1}^{N} dq_i = K_{\lambda N}^{(S)}(\{q\})\,, \tag{13}$$

где индекс s указывает, что функция $K_{\lambda_N}^{(s)}(\{q\})$ определена в соответствующем $6s$-мерном фазовом подпространстве. Пусть далее

$$\lim_{V,N\to\infty} \int K_{\lambda N}^{(s)}(\{q\})\, e^{-i\lambda_N t}\, d\lambda_N = \int K_\lambda^{(s)}(\{q\})\, e^{-i\lambda t}\, d\lambda\,, \tag{14}$$

где $\lambda = \lim\limits_{V,N\to\infty} \lambda_N$, $K_\lambda^{(s)} = \lim\limits_{V,N\to\infty} K_{\lambda_N}^{(s)}$. Тогда с помощью (13) и (14) получим формально:

$$\Phi_s(\xi) = -\xi \int \frac{K_\lambda^{(s)}(\{q\})}{i(L_s^0 - \lambda) - \xi}\, d\lambda\,. \tag{15}$$

В соответствии с теоремой Абеля [8] здесь нужно перейти к пределу $\xi \to 0$. При этом видно, что задача вообще говоря является нестационарной (обычно этот вывод делается в результате привлечения адиабатической гипотезы [6] и что при $\xi \to 0$ $(t \to \infty)$ уравнение (9) должно переходить в $L_N^0\, \psi_{\lambda_N}^0 = \lambda_N \psi_{\lambda_N}^0$.

Таким образом, предположение об эргодичности системы приводит с необходимостью к выводу, что взаимодействие в системе должно выключаться при $t \to \infty$. При этом F'_N должна распадаться на произведение одночастичных функций распределения $F'_1(t; q_i)$. Тогда, учитывая (8), получим

$$\lim_{t \to \pm\infty} S^{(s)}_{-t} F'_s(t, \{q\}) = \lim_{t \to \pm\infty} S^{(s)}_{-t} \prod_{i=1}^{s} F'_1(t; q_i) \,. \tag{16}$$

При $t \to -\infty$ полученное выражение совпадает с обычной формулировкой принципа ослабления корреляции для пространственно однородных систем одинаковых частиц. Выбор такого граничного условия приводит к правильному выбору частного решения цепочки уравнений ББГКИ, удовлетворяющего требованиям H-теоремы. Напротив, граничное условие при $t \to \infty$ приводит к уравнению, отличающемуся от уравнения кинетического типа знаком перед столкновительным членом [7].

Приведенные соображения оправдывают привлечение в статистической механике необратимых процессов адиабатической гипотезы [2, 6, 8] и позволяют определенным образом интерпретировать принцип ослабления корреляции. В работе [9] было показано, что Лаплас — образ $\Phi_s(\xi)$, определяемый формулой (7), можно интерпретировать как усредненную по времени функцию $F_s'^{\,вз}(t, \{q\})$ по промежутку $[0, T = 1/\xi]$. Таким образом, граничные значения (16) можно рассматривать как результат такого «усреднения по Лапласу» по промежутку времени $[0, \infty)$ или $(-\infty, 0]$ s — частичной функции распределения $F'_s(t, \{q\})$, записанной в представлении взаимодействия.

Принцип ослабления корреляции обычно применяется для систем, между частицами которых действуют силы отталкивания, монотонно убывающие с расстоянием [1, 10]. Приведенное рассмотрение основывается исключительно на предположении об эргодичности системы, и поэтому требует несколько менее жестких условий для применимости принципа ослабления корреляции (об условиях, при которых справедливо предположение об эргодичности системы, см., напр., в работах [11, 12]).

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ЛИТЕРАТУРА

1. Н. Н. Боголюбов, Проблемы динамической теории в статистической физике. М.—Л., 1946.
2. R. Aronson, Phys. Rev., **145**, 46, 1966.
3. Ван дер Поль Б., Бремер Х., Операционное исчисление на основе двустороннего преобразования Лапласа. М., 1952.
4. И. Пригожин, Неравновесная статистическая механика, М. 1964.
5. R. Zwanzig, Phys. Rev., **144**, 170, 1966.
6. Д. А. Киржниц, Полевые методы теории многих частиц, М., 1963.

7. E. Cohen, T. Berlin, Physica, **26**, 145, 1962.
8. R. Aronson, J. Math. Phys., **7.** 221, 1966.
9. W. Kohn, J. Luttinger, Phys. Rev., **108**, 590, 1957.
10. К. П. Гуров, Основания кинетической теории, М., 1966.
11. G. Sandri, Ann. of Phys., **24**, 332, 1963.
12. S. Golden, H. C. Longuet-Higgins, J. Chem. Phys., **33**, 1479, 1960.

COMMENTS ON THE PRINCIPLE OF ATTENUATION OF CORRELATION

By

S. A. SMOLYANSKY

Abstract

It is shown that the principle of attenuation of correlation is assumed to be due to the ergodicity of the system. This makes it possible to show the range of application of the principle of attenuation of correlation, which is slightly wider than generally assumed.

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NACHWEIS DER WELLENEIGENSCHAFT DES Li_7^+-IONS DURCH STREUUNG AN MOLEKÜLEN

Von

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FACHSCHULE FÜR OPTIK UND FOTOTECHNIK BERLIN*

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Streuversuche mit Li_7^+-Ionen (30 und 40 keV) an freien Molekülen (CCl_4, $SnBr_4$) ergaben in der Intensitäts-Winkel-Verteilung neben den durch die Polarisation hervorgerufenen Maxima die Überlagerung einer weiteren Folge von Maxima. Diese nur mit subtilen Messmethoden nachweisbaren Maxima unterscheiden sich von den anderen durch ihre grössere Breite und geringere Abnahme der Höhe mit steigender Ordnung. Sie liegen annähernd bei den Streuwinkeln, die sich aus der Theorie der Gasinterferenzen nach Debye—Ehrenfest unter Verwendung der Wellenlänge der gestreuten Li_7^+-Ionen errechnen.

Einleitung

Nach de Broglie ist einem Teilchen der Masse m und der Geschwindigkeit v die Wellenlänge

$$\lambda = \frac{h}{m \cdot v} \tag{1}$$

zugeordnet. Diese Beziehung wurde bisher für Elektronen [1], Neutronen[2], He-Atome [3] bzw. H_2-Moleküle [4] experimentell verifiziert. Mit 17,7 keV Protonen an CCl_4-Molekülen durchgeführte Streuversuche führten jedoch nur zu Andeutungen einiger Beugungsmaxima höherer Ordnung [5]. Eine vollständige Analyse war wegen des sehr intensiven Streuuntergrundes nicht möglich. Andere Versuche zur Beugung von langsamen Protonen (mehrere 100 eV) an polykristallinen Metallschichten oder schnellen Protonen (40 keV) an Einkristallen (Kalkspat u. a.) schlugen fehl, da entweder die Winkelauflösung der Beugungsapparatur unzureichend oder die streuenden Objekte zu ungenau bekannt waren [6].

Da einwandfreie Ergebnisse bei Beugungsversuchen mit schweren geladenen Partikeln nur in unzureichendem Masse vorliegen, wurde versucht, die Welleneingenschaft des Li_7^+-Ions durch Beugung an Molekülen quantitativ nachzuweisen.

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* Berlin, 1 Berlin 10, Einsteinufer 43/53.

Theorie der Ionenstreuung unter Berücksichtigung der Atomanordnung im Molekül

Die theoretische Winkelverteilung schneller Elektronen (1. BORNsche Näherung) nach elastischer Streuung an regellos orientierten Gasmolekülen ist nach DEBYE und EHRENFEST* gegeben durch [8]

$$I(\vartheta) = I_0 \cdot \left(\frac{2m_0 \cdot e_0^2}{\hbar^2}\right) \cdot \frac{1}{q^4} \cdot \sum_i \sum_j \left(Z_i - F_i(q)\right)\left(Z_j - F_j(q)\right) \cdot \frac{\sin(q \cdot l_{ij})}{(q \cdot l_{ij})}, \tag{2}$$

$$q = \frac{4\pi}{\lambda} \cdot \sin\frac{\vartheta}{2}. \tag{3}$$

Hierin bedeuten $I(\vartheta)$ die Intensität der gestreuten Strahlung, I_0 die Intensität der Primärstrahlung, ϑ den Winkel zwischen Primärstrahl und gestreutem Strahl im Schwerpunktsystem, m_0 die Masse, e_0 die Ladung und λ die Wellenlänge des Elektrons, $h = 2\pi\hbar$ das PLANKsche Wirkungsquantum, l_{ij} den Abstand des Atoms i vom Atom j im streuenden Molekül, Z_i die Ordnungszahl des Atoms i und F_i sein Formfaktor für Röntgenstrahlen.

Die Gleichungen (2) und (3) geben die bei Streuung schneller Elektronen (einige 10 keV) an Molekülen experimentell beobachteten Intensitäts-Winkelverteilungen sehr genau wieder [8]. Diese Beziehungen können unter der Voraussetzung, dass die BORNsche Näherung gültig bleibt, auf die Streuung von einfach geladenen *Ionen* an Molekülen angewandt werden.** Für die Streuwinkel der Maxima in der Intensitäts-Winkelverteilung folgt dann näherungsweise für kleine Streuwinkel ($\sin\vartheta \simeq \vartheta$)

$$\vartheta_{\mathrm{max,n}} \simeq \frac{\lambda}{2\pi} \cdot q_{\mathrm{max,n}} \qquad n = 1, 2 \ldots \tag{4}$$

Da die $q_{\mathrm{max,n}}$-Werte aus den $\vartheta_{\mathrm{max,n}}$-Werten, die bei der Streuung von Elektronen an Molekülen beobachtet werden, bekannt sind, lassen sich nach (4) die erwarteten Streuwinkel für Ionen bekannter Wellenlänge berechnen.

Für die Streuung von 30 keV-Li_7^+-Ionen ($\lambda = 6{,}32 \cdot 10^{-12}$ cm) an CCl_4- und SnBr_4-Molekülen [11, 12] errechnen sich *Streuwinkel* $\vartheta^{\mathrm{ber}}_{\mathrm{max\,n}}$ von 10^{-4} bis 10^{-3} rad (vgl. Tab. 1). Diese Streuwinkel bzw. -Maxima werden durch die

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* Die Streuformel wurde von DEBYE und EHRENFEST ursprünglich für die Streuung einer ebenen, elektromagnetischen Welle (Röntgenstrahlung) an Molekülen unabhängig voneinander berechnet [7] und von WIERL [8] durch Hinzufügung von Formfaktoren auf Elektronen erweitert.

** Der Einfluss der Formfaktoren $F(q)$ auf die Lage der Intensitätsmaxima ist sehr gering. Durch Weglassen der Formfaktoren in Gl. (2) ergeben sich q_{max}-Werte, die von denen nach Gl. (2) abweichen (vgl. [9]). Eine Änderung der Formfaktoren durch Übergang von Elektronen zu Ionen wird deshalb von untergeordneter Bedeutung sein.

Atomanordnung im Molekül hervorgerufen und im folgenden abgekürzt als »Strukturwinkel« bzw. »Strukturmaxima« bezeichnet. Im Gegensatz dazu werden die durch die Polarisation der Moleküle hervorgerufenen Streuwinkel bzw. -Maxima (vgl. [9, 10]) als »Polarisationswinkel« bzw. »Polarisationsmaxima« bezeichnet.

Experimentelle Ermittlung der »Strukturmaxima« und Diskussion

Die bisher durchgeführten Streuversuche [9] weisen nur in einem Punkt auf die mögliche Existenz von »Strukturmaxima« hin: Bei der Streuung von Li_7^+-Ionen an CCl_4-Molekülen ist das zwischen 3. und 4. »Polarisationsmaximum« liegende Minimum stets so schwach ausgeprägt, d. h. die Photometerkurve berührt in dieser Stelle nicht die tangentielle »Untergrundkurve« (vgl. Abb. 4 in [9]), dass hier die Überlagerung eines weiteren, breiten Maximums denkbar ist. Offenbar sind die bei der Ionenstreuung auftretenden »Strukturmaxima« wesentlich schwächer ausgeprägt als bei der Elektronenstreuung. Aus diesem Grunde wurden besondere Massnahmen zum Hervorheben der »Strukturmaxima« ergriffen und die Versuchtstechnik verfeinert.

Zunächst wurden in einer einfachen Streuapparatur (Abb. 1 in [9]) 30 keV-Li_7^+-Ionen an CCl_4-Molekülen gestreut und Diagramme mit wesentlich längerer (mehr als 10-facher) Belichtungszeit hergestellt, als zur optimalen Sichtbarmachung der »Polarisationsmaxima« notwendig ist. Dann wurden die auf Ilford Q1-Platten aufgenommenen Streudiagramme mit einem photographischen Abschwächer* behandelt und im Lichtmikroskop vergrössert photographiert (vgl. Masstab in Abb. 1a). Auf den so erhaltenen Aufnahmen sind bereits 2—3 »Strukturmaxima« zu erkennen (Abb. 1a). Dagegen sind auf den schwächer belichteten Streudiagrammen, die in gleicher Weise behandelt wurden, nur die »Polarisationsmaxima« angedeutet (Abb. 2a). Schliesslich wurden die in Abb. 1a und 2a dargestellten Streudiagramme (bei schneller Rotation um ihren Mittelpunkt, um die Störung durch die Körnigkeit des Photomaterials zu reduzieren) photometriert (Abb. 1b und 2b).

Die Photometerkurve des stark belichteten Streudiagramms (Abb. 1b) lässt deutlich die breiten »Strukturmaxima« II, III, IV, V neben den schmalen »Polarisationsmaxima« 1, 2, 3 erkennen. Lediglich das I. »Strukturmaximum« (gestrichelt hervorgehoben) wird vom 3. »Polarisationsmaximum« störend überlagert, so dass dessen Lage nicht mit der gleichen Genauigkeit wie die der übrigen bestimmt werden kann. Die »Strukturmaxima« höherer Ordnung werden nicht mehr gestört, da die Höhen der »Polarisationsmaxima« mit

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* Es wurde ein unterproportionaler Abschwächer benutzt, d. h. kleine Schwärzungen wurden stärker abgeschwächt als grosse Schwärzungen [13].

grösser werdendem Streuwinkel stärker als die der »Strukturmaxima« abnehmen.

Die Photometerkurve des schwach belichteten Streudiagramms (Abb. 2b) lässt nur noch die »Polarisationsmaxima« deutlich erkennen. Hier entspricht die Intensitäts-Winkelverteilung annähernd den unter den früheren Versuchsbedingungen erhaltenen.

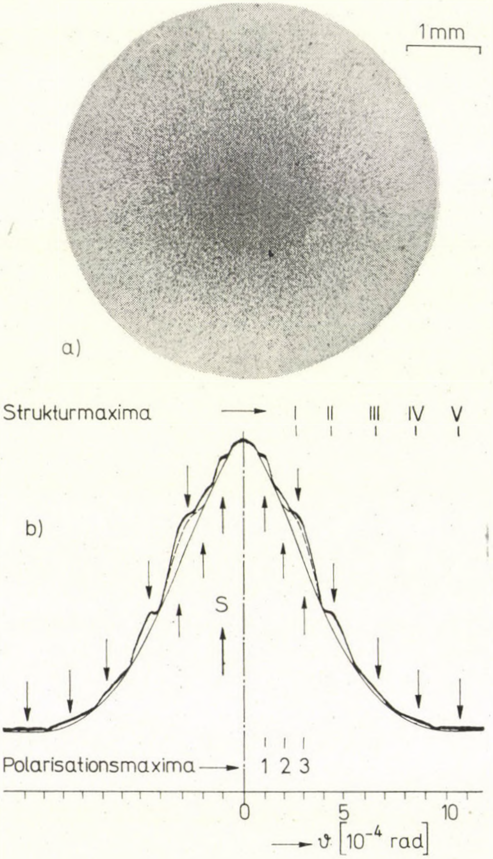

Abb. 1

Zur Überprüfung der mit der einfachen Streuapparatur erhaltenen Ergebnisse wurden die »Strukturmaxima« auch mit der Streuapparatur mit Bildwandler (vgl. [9]) und einem rotierenden Sektor, der die Streuintensitäten bei kleinen Streuwinkel reduziert [14], reproduziert. Auch hier waren die relativ breiten »Strukturmaxima« neben den schmalen »Polarisationsmaxima« deutlich zu erkennen.

Die Auswertung zahlreicher Messungen ergibt die in Tab. 1 dargestellten »Strukturwinkel« $\vartheta_{\max,n}^{\text{exp.}}$. Die experimentell gefundenen und die berechneten

»Strukturwinkel« stimmen innerhalb der Fehlergrenzen von etwa 5–8% überein.

Zur Vervollständigung des experimentellen Materials wurde noch die Streuung von 30 keV-Li_7^+-Ionen an Sn Br_4-Molekülen untersucht (vgl. Tab. 1).

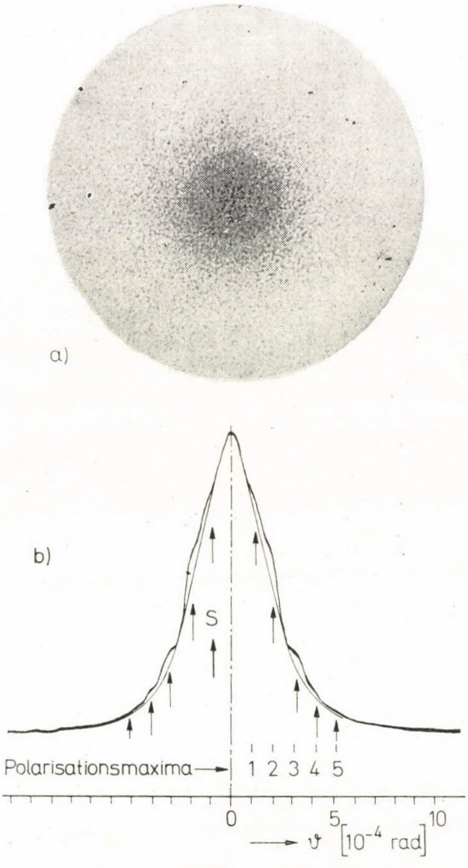

Abb. 2

Hier sind die gemessenen »Strukturwinkel« durchschnittlich um etwa 11% kleiner als die berechneten Werte. Die Abweichungen sind grösser als die geschätzten Fehlergrenzen und kommen evtl. dadurch zustande, dass die aus Elektronen-Streuexperimenten ermittelten Werte $q_{\max,n}$ nicht genau ermittelt worden sind.*

Neben den durch die Polarisation des Moleküls hervorgerufenen Intensitätsmaxima treten also auch Intensitätsmaxima auf, welche in annähernder

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* Die Diskrepanz könnte auch dadurch erklärt werden, dass wegen der Änderung der Bindungskräfte durch die Polarisation des Streuers die Atomabstände im Molekül vergrössert werden.

Tabelle I

Vergleich zwischen berechneten und gemessenen »Struktur-Winkeln« bei der Streuung von 30 keV Li^+_7-Ionen an CCl_4- und $SnBr_4$-Molekülen

Streuer	Ordnung der Strukturmaxima	$q_{max,n}$ [Å^{-1}]	$\vartheta^{ber}_{max,n}$	$\vartheta^{exp}_{max,n}$	$\Delta\vartheta$ [%]
			[10^{-4} rad]		
CCl_4	$n =$ I	2,76	2,78	(2,70)	(—3)
	II	4,94	4,98	4,65	—5
	III	7,34	7,40	6,80	—8
	IV	9,34	9,41	8,70	—7
	V	11,79	11,82	(10,70)	(—9)
			mittlere Abweichung:		—6,6%
$SnrB_4$	I	2,02	2,04	—	—
	II	3,49	3,52	3,17	—10
	III	5,34	5,38	4,63	—14
	IV	6,58	6,63	5,74	—13
	V	8,31	8,37	7,42	—11
	VI	9,90*	10,00	9,28	— 9
	VII	11,26	11,35	—	—
			mittlere Abweichung:		—11,4%

* Der $q_{max,n}$-Wert für $n =$ VI wurde aus dem theoretischen Kurvenverlauf der Intensitäts-Winkelverteilung ermittelt, da hierfür kein experimenteller Wert vorliegt [11].

Übereinstimmung mit der Theorie von DEBYE und EHRENFEST durch die Anordnung der Atome im Molekül verursacht werden. Damit ist die DE BROGLIE-Beziehung auch für Li^+_7-Ionen verifiziert.

LITERATURVERZEICHNIS

1. G. P. THOMSON, Nature (London), **120**, 802, 1927.
2. H. HALBAN and P. PREISWERK, C. R. Acad. Sci., **203**, 73, 1936.
3. O. STERN, Naturwiss., **17**, 391, 1929.
4. I. ESTERMANN und O. STERN, Z. Phys., **61**, 95, 1930.
5. H. J. YEARIAN, J. Chem. Phys., **8**, 24, 1940.
6. Y. SUGIURA, Sci. Pap. Ist. Phys. Chem. Res., **16**, 29, 1931; A. J. DEMPSTER, J. Chem. Phys., **8**, 24, 1940.
7. P. DEBYE, Ann. Phys., **46**, 809, 1915; P. EHRENFEST, Amst. Akad., **23**, 1122, 1915.
8. R. WIERL, Ann. Phys., **8**, 521, 1931; L. O. BROCKWAY, Rev. Mod. Phys., **8**, 231, 1936.
9. H. BOERSCH und G. FORST, Z. Phys., **169**, 239, 1962.
10. H. BOERSCH und G. FORST, Z. Phys., **176**, 221, 1963.
11. I. L. KARLE and J. KARLE, J. Chem. Phys., **17**, 1052, 1949.
12. M. W. LISTER and L. E. SUTTON, Trans. Faraday Soc., **37**, 393, 1941.
13. E. LAU und E. G. KIND, Optik, **18**, 646, 1961.
14. G. TRENDELENBURG, Naturwiss., **21**, 173, 1933; CHR. FINBAK, Avh. Norsk Vitensk-Akd. Oslo, Mat. Naturw. Klasse, No. 13, 1937.

ДОКАЗАТЕЛЬСТВО ВОЛНОВЫХ СВОЙСТВ ИОНА Li_7^+ РАССЕЯНИЕМ НА МОКЕКУЛАХ

Г. ФОРСТ

Резюме

Эксперимент по рассеянию ионов Li_7^+ (30 и 40 Кэв) на свободных молекулах (CCl_4, $SnBr_4$) показывает, что в распределении интенсивности по углам наряду с максимумами, обусловленными поляризацией, имеется и наложение других серий максимумов. Эти максимумы, обнаруженные только уточненными методами исследования, отличаются от других своей большей шириной и медленным снижением высоты с увеличением порядка. Они располагаются приблизительно при углах рассеяния, вычисленных на основе теории интерференции газов Дэбая-Эренфеста путем использования длины волн рассеянных ионов Li_7^+.

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NEGATIVE PARITY BARYON RESONANCES IN THE SU(6) SYMMETRY

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The decay ratios of negative parity baryon resonances are compared with prediction of $SU_W(6)$ symmetry by assigning these resonances to the 540 supermultiplet. Some "semi-empirical" mass formulae are also investigated. It turns out that the present experimental data are not inconsistent with such an assignment. Nevertheless, we believe that not all the negative parity baryon resonances of spins $1/2^-$, $3/2^-$, $5/2^-$ fit into the 540-plet without contradicting experimental values.

I. Introduction

At present there is experimental evidence for many resonances with various spin and parity, but the assignment of these resonances to SU(6) supermultiplets has not been successful in every case. In particular the situation regarding negative parity baryon resonances is still rather unclear.

The following negative parity baryon resonances are known [1]:

J^P	YI	Mass (MeV)	Width (MeV)
$5/2^-$	$N^*_{1/2}$	1670	140
$5/2^-$	Y^*_1	1768	89 ± 12
$3/2^-$	$N^*_{1/2}$	1525	105
$3/2^-$	Y^*_1	1660	50
$3/2^-$	Y^*_0	1518.8	16 ± 2
$3/2^-$	Y^*_0	1700	40 ± 10
?? $3/2^-$	$\Xi^*_{1/2}$	1815	16 ± 8
$1/2^-$	$N^*_{1/2}$	1570	130
$1/2^-$	$N^*_{1/2}$	1700	240
$1/2^-$	$N^*_{3/2}$	1670	180
$1/2^-$	Y^*_0	1405	35
? $1/2^-$	Y^*_0	1670	18

??: The spin parity poorly known, ?: dubious state.

The evidence for assigning a certain set of baryon resonances to an SU(6) supermultiplet comes from the decays, mass relations and spin-parity properties. There have been many attempts to assign the resonances $1/2^-$, $3/2^-$, $5/2^-$ to SU(6) supermultiplets. We investigate the possibility of assigning the known baryon resonances with spins $1/2^-$, $3/2^-$, $5/2^-$ to one given irreducible representation of static SU(6) or static SU(6)⊗SU(6) group. **Bég** and **Singh** [2, 3] have studied the $3/2^-$ resonances in a 70^--plet. This representation has the following SU(3)⊗SU(2) content

$$70 = (10,2) \oplus (8,4) \oplus (8,2) \oplus (1,2).$$

Therefore, $3/2^-$ resonances fit into this representation, if they form an SU(3) octet. Such an octet was proposed a long time ago as the γ octet.

Bég and **Singh** studied only the mass relations. The consistent assignment did not succeed.

Gatto, **Maiani** and **Preparata** [4] have investigated these resonances in the supermultiplet mixing model considering also the mass relations. In this model the physical particles are treated as bound states of quarks. Therefore, they belong to reducible representations. On the ground of the predictions obtained it is impossible to assign the physical particles into kinetic supermultiplets unambiguously. **Meshkov** and **Yodh** [5] were the first to investigate these particles from the point of view of decay ratios.

In a relativistic SU(6) symmetry model for collinear processes the $SU(6)_W$ group is the maximal possible symmetry group (see for example [6]); at the same time in the quark model the negative parity baryon resonances can be treated as *s* wave bound states of four quarks and one antiquark ($qqqq\bar{q}$ systems). This assumption gives the possible representations for the classification.

We must study the irreducible representations occurring in the tensor product

$$\underline{6} \otimes \underline{6} \otimes \underline{6} \otimes \underline{6} \otimes \underline{6}^* .$$

Therefore, we must choose between the representations of $\underline{1134}$, $\underline{700}$, $\underline{560}$, $\underline{540}$, $\underline{70}$, $\underline{56}$, $\underline{20}$.

Meshkov and **Yodh** have considered the predictions for the decay ratios, coming from the assignment of the resonances $Y_1^*\ (1765)^{5/2^-}$ and $Y_0^*\ (1520)^{3/2^-}$ to an $\underline{1134}$-plet.

The conclusion, which can be drawn from their results is that these resonances can be fitted consistently into an $\underline{1134}$-plet, though lately the experimental data have been modified into "wrong direction".

With the present experimental data the investigation of the $\underline{700}$-plet is inconclusive.

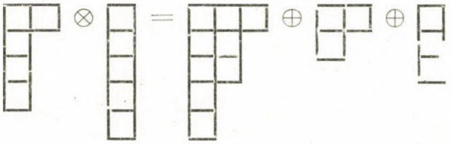
We have studied the consequences of choosing the 540-plet. For the sake of simplicity, simple "semi-empirical" mass relations were used, which had already been proved to be correct in other cases [7]. The $SU(6)_q \times SU(6)^{q}_{\bar{q}}$ has an irreducible representation

$$(105', 6^*).$$

(In the parentheses the first number gives the multiplicity with respect to $SU(6)_q$ and the second number with respect to $SU(6)_{\bar{q}}$.)

This representation can be split into SU(6) invariant subspaces as follows:

$$\underline{105'} \otimes \underline{6'} = \underline{540} \oplus \underline{70} \oplus \underline{20}$$



If we restrict ourselves to such two-body decays of negative parity baryon resonances in which the particles of the final state belong to $\underline{56}^+$ baryon and $\underline{35}^-$ meson supermultiplets of static SU(6), respectively, requiring $SU(6)_W$ symmetry it turns out that there is only one coupling constant. The simple mass formula and the single coupling constant makes it possible to analyse the assignment to a $\underline{540}$-plet.

II. Mass relations

The $SU(3) \times SU(2)$ content of 540-plet is

$$\underline{540} = (\underline{27},\underline{4}) \oplus (\underline{27},\underline{2})_{\mathrm{I}} \oplus (\underline{27},\underline{2})_{\mathrm{II}} \oplus (\underline{10}^*,\underline{4}) \oplus (\underline{10},\underline{4}) \oplus (\underline{10},\underline{2}) \oplus (\underline{10},\underline{2}) \oplus (\underline{8},\underline{6}) \oplus$$

$$\oplus (\underline{8},\underline{4})_{\mathrm{I}} \oplus (\underline{8},\underline{4})_{\mathrm{II}} \oplus (\underline{8},\underline{4})_{\mathrm{III}} \oplus (\underline{8},\underline{2})_{\mathrm{I}} \oplus (\underline{8},\underline{2})_{\mathrm{II}} \oplus (\underline{8},\underline{2})_{\mathrm{III}} \oplus (\underline{1},\underline{6}) \oplus (\underline{1},\underline{4}) \oplus (\underline{1},\underline{2})$$

Because of the presence of three (8.2) and (8.4) there are many possibilities for mixing. The physical states are eigenstates of the mass operator, therefore the consideration of mass formulae is important also from this point of view: they determine the mixing in part.

A systematic approach dealing with the masses in static SU(6) has been developed in [8]. A summary of different mass formulae can be found in [7]. Since

$$\underline{540} \otimes \underline{540}^* = \underline{34496} \oplus \underline{29700} \oplus \underline{19845} \oplus \underline{19845}^* \oplus \underline{14715} \oplus \underline{14715}^* \oplus 2\cdot\underline{12250} \oplus$$

$$\oplus 2\cdot\underline{12250}^* \oplus \underline{11340} \oplus \underline{6720} \oplus 2\cdot\underline{5670} \oplus \underline{5670}^* \oplus 4\cdot\underline{3969} \oplus 6\cdot\underline{3675} \oplus 2\cdot\underline{3200} \oplus$$

$$\oplus 2\cdot\underline{3200}^* \oplus \underline{2695} \oplus \underline{1134} \oplus \underline{1134}^* \oplus 4\cdot\underline{189} \oplus 2\cdot\underline{175} \oplus 3\cdot\underline{35} \oplus 1$$

the case of the 540-plet is rather complicated; the corresponding mass formula contains 11 parameters. These problems can be simplified only with assumptions which do not come from the group structure. In [7] there are proposed mass relations in U chain which we can obtain from the general relations neglecting some terms.

These formulae are:

$$M = a + c\,J(J+1) + dY + e\left[2\,S(S-1) - C_2^{(4)} + \frac{1}{4}\,Y^2\right] + g\left[I(I+1) - \frac{1}{4}\,Y^2\right], \tag{A}$$

$$M = a + c\,J(J+1) + dY + e\left[2\,S(S+1) - C_2^{(4)} + \frac{1}{4}\,Y^2\right] + g\left[I(I+1) - \frac{2}{5}\,C_2^{(4)}\right]. \tag{B}$$

In the representations 35, 56, 189, 405 these mass formulae agree very well with the experimental data, although they can be considered only as semi-empirical mass relations.

Another simple possible mass formula is proposed by Lipkin as follows:

$$M = M_0 + b \cdot S,$$

where S is the strange quark number. In the case of particles with spins $5/2^-$ in $(qqqq\bar{q})$ systems the strange quark number is proportional to the strange quark spin. Knowing the states of the U chain the mass difference of two $5/2^-$ resonances can be calculated. In the $SU(4) \otimes SU(2)_S \otimes U(Y)$ decomposition the content of $\underline{540}$ is:

$$\underline{540} = (\underline{64},\underline{2})^{\circ} \oplus (\underline{36},\underline{1})^{-1} \oplus (\underline{36}^*,\underline{1})^{+1} \oplus (\underline{20}',\underline{3})^{+1} \oplus (\underline{20}'^*,\underline{3})^{-1} \oplus (\underline{20}',\underline{1})^{+1} \oplus$$
$$(\underline{20}'^*,\underline{1})^{-1} \oplus (\underline{15},\underline{2})^{2} \oplus (\underline{15},\underline{2})^{-2} \oplus (\underline{10},\underline{2})^{\circ} \oplus (\underline{10}^*,\underline{2})^{\circ} \oplus (\underline{6},\underline{4})^{\circ} \oplus (\underline{6},\underline{2})^{\circ}_{a} \oplus (\underline{6},\underline{2})^{\circ}_{b} \oplus$$
$$\oplus (\underline{4}^*,\underline{3})^{+1} \oplus (\underline{4},\underline{3})^{-1} \oplus (\underline{4},\underline{1})^{-} \oplus (\underline{4}^*,\underline{1})^{1},$$

where the first number in the parentheses gives the multiplicity with respect to SU(4), the second number that with respect to $SU(2)_S$ and the upper index is the eigenvalue of Y. The states in the eigenvalue of Y. The states in the $SU(2)_I \otimes SU(2)_N \otimes SU(2)_S \otimes U(Y)$ decomposition can be found in Table II.

For the resonance $5/2^-\ N^*_{1/2}\,(1670)$ the strange quark spin is 1, and for $5/2^-\ Y^*_1\,(1768)$ it is 1/2. Therefore taking b as positive, Lipkin's formula gives $N^*_{1/2}$ and Y^*_1 in the wrong order.

Table I

$R^{e\varepsilon}_{[a\alpha,b\beta,c\gamma]d\delta} = \varepsilon_{abc}\,\varepsilon^{\varepsilon\varrho}\,c^{e}_{d,\alpha\beta\gamma\delta\varrho} +$	(8,6)
$+\frac{1}{\sqrt{3}}\,\varepsilon^{\varepsilon\varrho}\,\varepsilon_{abc}\,\delta^{e}_{d}\,S_{\alpha\beta\gamma\delta\varrho} +$	(1,6)
$+ [\delta^{\varepsilon}_{\alpha}\,\varepsilon_{bcr}\,K^{er}_{ad,\beta\gamma\delta} + \varepsilon_{car}\,\delta^{\varepsilon}_{\beta}\,K^{er}_{bd,\gamma\alpha\delta} + \varepsilon_{abr}\,\delta^{\varepsilon}_{\gamma}\,K^{er}_{cd,\alpha\beta\gamma}] +$	(27,4)
$+\frac{1}{\sqrt{2}}\,[\varepsilon^{\varepsilon\varrho}(\delta^{e}_{a}\,P_{bcd,\varkappa\delta\varrho}\,\varepsilon_{\beta\gamma} + \delta^{e}_{b}\,P_{cad,\beta\delta\varrho}\,\varepsilon_{\gamma\varkappa} + \delta^{e}_{c}\,P_{abd,\gamma\delta\varrho}\,\varepsilon_{\alpha\beta})] +$	(10,4)
$+\frac{1}{\sqrt{2}}\,[\varepsilon_{abr}\,\varepsilon_{cds}\,\varepsilon_{\alpha\omega}\,\varepsilon_{\beta\varrho}\,\varepsilon_{\gamma\delta}\,P^{*ers,\varepsilon\omega\varrho} + \varepsilon_{bcr}\,\varepsilon_{ads}\,\varepsilon_{\beta\omega}\,\varepsilon_{\gamma\varrho}\,\varepsilon_{\alpha\delta}\,P^{*ers,\varepsilon\omega\varrho} +$ $+ \varepsilon_{car}\,\varepsilon_{bds}\,\varepsilon_{\gamma\omega}\,\varepsilon_{\alpha\varrho}\,\varepsilon_{\beta\delta}\,P^{*er,\varepsilon\omega\delta}] +$	(10*,4)
$+\sum_{i=1}^{3}\gamma_i\,[(2\,x_i + y_i)\,(\delta^{e}_{c}\,\varepsilon_{abr} + \delta^{e}_{a}\,\varepsilon_{bcr} + \delta^{e}_{b}\,\varepsilon_{car})\,\delta^{\varepsilon}_{r}\,d^{r}_{id,\alpha\beta\gamma} +$ $+ (2\,x_i - y_i)\,\delta^{\varepsilon}_{\alpha}(\delta^{e}_{d}\,\varepsilon_{bcr}\,d^{r}_{ia,\delta\beta\gamma} + \delta^{\varepsilon}_{\beta}\,\varepsilon_{car}\,d^{r}_{ib,\delta\gamma\varkappa} + \delta^{\varepsilon}_{\beta}\,\varepsilon_{bar}\,d^{r}_{ic,\delta\varkappa\beta}) +$ $+ x_i\,\delta^{\varepsilon}_{\alpha}\,\delta^{e}_{a}(\varepsilon_{dbr}\,d^{r}_{ic,\beta\gamma\delta} - \varepsilon_{dcr}\,d^{r}_{ib,\beta\gamma\delta}) +$ $+ x_i\,\delta^{\varepsilon}_{\beta}\,\delta^{e}_{b}(\varepsilon_{dcr}\,d^{r}_{ia,\gamma\alpha\delta} - \varepsilon_{dar}\,d^{\gamma}_{ic,\gamma\varkappa\delta}) + x_i\,\delta^{\varepsilon}_{\gamma}\,\delta^{e}_{c}\,(\varepsilon_{dar}\,d^{r}_{ib,\alpha\beta\delta} -$ $- \varepsilon_{dbr}\,d^{r}_{ia,\alpha\beta\delta}) + y_i\,\delta^{\varepsilon}_{\alpha}(\delta^{e}_{c}\,\varepsilon_{dbr} - \delta^{e}_{b}\,\varepsilon_{dcr})\,d^{r}_{a,\gamma\beta\delta} +$ $+ y_i\,\delta^{\varepsilon}_{\beta}(\delta^{e}_{a}\,\varepsilon_{dcr} - \delta^{e}_{c}\,\varepsilon_{dar})\,d^{r}_{ib,\varkappa\gamma\delta} + y_i\,\delta^{\varepsilon}_{\gamma}\,(\delta^{e}_{b}\,\varepsilon_{dar} - \delta^{e}_{a}\,\varepsilon_{dbr})\,d^{r}_{ic,\beta\varkappa\delta} +$ $+ 3\,z_i\,\varepsilon_{abc}\,\{4\,\delta^{\varepsilon}_{\delta}\,d^{e}_{id,\alpha\beta\gamma} -$ $- (\delta^{\varepsilon}_{\alpha}\,d^{e}_{id,\beta\gamma\delta} + \delta^{\varepsilon}_{\beta}\,d^{e}_{id,\gamma\alpha\delta} + \delta^{\varepsilon}_{\gamma}\,d^{e}_{id,\alpha\beta\delta})\} + z_i\,\varepsilon_{dbc}\,\{4\,\delta^{e}_{\alpha}\,d^{e}_{ia,\delta\gamma\beta} -$ $- (\delta^{\varepsilon}_{\beta}\,d^{e}_{ia,\delta\varkappa\gamma} + \delta^{\varepsilon}_{\gamma}\,d^{e}_{ia,\delta\varkappa\beta} + \delta^{\varepsilon}_{\delta}\,d^{e}_{ia,\alpha\beta\gamma})\} + z_i\,\varepsilon_{dac}\,\{4\,\delta^{\varepsilon}_{\beta}\,d^{e}_{ib,\delta\varkappa\gamma} -$ $- (\delta^{\varepsilon}_{\gamma}\,d^{e}_{ib,\delta\beta\varkappa} + \delta^{\varepsilon}_{\delta}\,d^{e}_{ib,\delta\beta\gamma} + \delta^{\varepsilon}_{\delta}\,d^{e}_{i\alpha,\varkappa\beta\gamma})\} + z_i\,\varepsilon_{dbc}\,\{4\,\delta^{\varepsilon}_{\gamma}\,d^{e}_{ic,\delta\beta\varkappa} -$ $- (\delta^{\varepsilon}_{\alpha}\,d^{e}_{ic,\delta\gamma\beta} + \delta^{\varepsilon}_{\beta}\,d^{e}_{ic,\delta\gamma\alpha} + \delta^{\varepsilon}_{\delta}\,d^{e}_{ic,\alpha\beta\gamma})\} +$	(8,4)
$+\frac{1}{\sqrt{210}}\Big\{\varepsilon_{abc}\,\delta^{e}_{d}\Big[\delta^{\varepsilon}_{\delta}\,s'_{\alpha\beta\gamma} + \frac{7}{8}\,\varepsilon^{\varepsilon\varrho}\,(\varepsilon_{\alpha\delta}\,s'_{\beta\gamma\varrho} + \varepsilon_{\beta\delta}\,s_{\gamma\alpha\varrho} + \varepsilon_{\gamma\delta}\,s'_{\alpha\beta\gamma})\Big] +$ $+ \varepsilon_{dbc}\,\delta^{e}_{a}\Big[\delta^{\varepsilon}_{\alpha}\,s'_{\beta\gamma\delta} + \frac{7}{8}\,\varepsilon^{\varepsilon\varrho}(\varepsilon_{\beta\varkappa}\,s'_{\gamma\delta\varrho} + \varepsilon_{\gamma\alpha}\,s'_{\beta\delta\varrho} + \varepsilon_{\delta\alpha}\,s'_{\beta\gamma\varrho})\Big] +$ + **cyclical permutation in** $(a\varkappa.\,b\beta.\,c\gamma)\Big\} +$	(1,4)

$$\begin{aligned}&+\sum_{i=1}^{2}\beta_i\,\{u_i\,\varepsilon_{adr}\,\varepsilon_{\alpha\delta}\,(L^{er}_{ibc,\gamma}\,\delta^{\varepsilon}_{\beta}-L^{er}_{ibc,\beta}\,\delta^{\varepsilon}_{\gamma})+v_i\,\delta^{\varepsilon}_{\delta}\,L^{er}_{iab,\gamma}\,\varepsilon_{cdr}\,\varepsilon_{\alpha\beta}+\\&+w_i\,\varepsilon_{abr}\,\varepsilon_{\delta\gamma}(\delta^{\varepsilon}_{\beta}\,L^{er}_{idc,\alpha}+\delta^{\varepsilon}_{\alpha}\,L^{er}_{icd,\beta})\}+\\&+\text{cyclical permutation in }(a\alpha,b\beta,c\gamma)+\end{aligned}\tag{27,2}$$

$$\begin{aligned}&+\frac{1}{\sqrt{3}}\left(\delta^{\varepsilon}_{\delta}\,\delta^{e}_{a}\,Q_{bcd,\alpha}\,\varepsilon_{\beta\gamma}-\frac{1}{2}\,\delta^{\varepsilon}_{\alpha}\,\delta^{e}_{a}\,Q_{bcd,\delta}\,\varepsilon_{\beta\gamma}\right)+\\&+\text{cyclical permutation in }(a\alpha,b\beta,c\gamma)\end{aligned}\tag{10,2}$$

$$\begin{aligned}&+\frac{1}{2\sqrt{3}}\,\varepsilon_{abr}\,\varepsilon_{cds}(\delta^{\varepsilon}_{\beta}\,\varepsilon_{\alpha\varrho}+\delta^{\varepsilon}_{\alpha}\,\varepsilon_{\beta\varrho})\,Q^{*ers,\varrho}\,\varepsilon_{\gamma\delta}+\\&+\text{cyclical permutation in }(a\alpha,b\beta,c\gamma)+\end{aligned}\tag{10,2*}$$

$$\begin{aligned}&+\Sigma_i\,\alpha_i\,\{(y'_i+z'_i)\,b^{r}_{id,\alpha}\,\varepsilon_{\beta\gamma}\,\delta^{\varepsilon}_{\delta}(\delta^{e}_{c}\,\varepsilon_{abr}+\delta^{e}_{b}\,\varepsilon_{acr})+y'_i\,b^{r}_{ia,\delta}\,\varepsilon_{\beta\gamma}(\delta^{e}_{c}\,\varepsilon_{dbr}+\delta^{e}_{b}\,\varepsilon_{dcr})\,\delta^{\varepsilon}_{\alpha}+\\&+b^{r}_{ia,\gamma}\,\varepsilon_{\beta\delta}\,\delta^{\varepsilon}_{\alpha}\,[(-2\,y'_i-5\,z'_i)\,\varepsilon_{bdr}\,\delta^{e}_{c}+\\&+z_i\,\varepsilon_{\beta cr}\,\delta^{e}_{d}]+b^{r}_{ia,\beta}\,\varepsilon_{\gamma\delta}\,\delta^{\varepsilon}_{\alpha}\,[(2\,y'_i+5\,z'_i)\,\varepsilon_{cdr}\,\delta^{e}_{b}+z'_i\,\varepsilon_{bcr}\,\delta^{e}_{d}]+\\&+x_i\,b^{e}_{ia,\alpha}\,\varepsilon_{dbc}(\varepsilon_{\beta\delta}\,\delta^{\varepsilon}_{\gamma}+\varepsilon_{\gamma\delta}\,\delta^{\varepsilon}_{\beta})-\frac{1}{4}\,\delta^{e}_{a}\delta^{\varepsilon}_{\alpha}\Big[\varepsilon_{\gamma\delta}[(3\,x_i+2\,y_i+4\,z_i)\,\varepsilon_{dcr}\,b^{r}_{ib,\beta}\\&+(4\,y'_i+6\,z'_i)\,\varepsilon_{bcr}\,b^{r}_{i\alpha,\beta}+(2\,y'_i+6\,z'_i)\,\varepsilon_{dbr}\,b^{r}_{ic,\beta}]-\\&-\varepsilon_{\beta\delta}\,[(3\,x'_i+2y\;+4\,z'_i)\,\varepsilon_{dbr}\,b^{r}_{ic,\gamma}-(4\,y'_i+6\,z'_i)\,\varepsilon_{bcr}\,b^{r}_{id,\gamma}+\\&+(2\,y'_i+6\,z'_i)\,\varepsilon_{dcr}\,b^{r}_{ib,\gamma}]+2\,y'_i\,\varepsilon_{\gamma\beta}]+2\,y'_i\,\varepsilon_{\gamma\beta}\,[\varepsilon_{dcr}\,b^{r}_{ib,\delta}+\varepsilon_{dbr}\,b^{r}_{ic,\delta}]\Big]+\\&+\text{cyclical permutation in }(a\alpha,b\beta,c\gamma)\}+\end{aligned}$$

$$\begin{aligned}&+\frac{1}{12\sqrt{2}}\,[\varepsilon_{abc}\,\delta^{e}_{\alpha}(\delta^{\varepsilon}_{\beta}\,\varepsilon_{\delta\alpha}+\delta^{\varepsilon}_{\alpha}\,\varepsilon_{\delta\beta})\,s''_{\gamma}+\varepsilon_{dbc}\,\delta^{e}_{a}\,\delta^{\varepsilon}_{\alpha}(\varepsilon_{\beta\delta}+\\&+\varepsilon_{\gamma\delta})\,s''_{\beta}+\varepsilon_{dbc}\,\delta^{e}_{a}(\varepsilon_{\alpha\beta}\,\delta^{\varepsilon}_{\gamma}+\varepsilon_{\alpha\gamma}\,\delta^{\varepsilon}_{\beta})s''_{\delta}+\\&+\text{cyclical permutation in }(a\alpha,b\beta,{}_{s}\gamma).\end{aligned}\tag{1,2}$$

$\alpha_1=\dfrac{1}{2\sqrt{546}}$ $x_1{}'=10$ $y'_1=4$ $z'_1=-1$

$\alpha_2=\dfrac{1}{2\sqrt{2210}}$ $x'_2=34$ $y'_2=-28$ $z'_2=7$

$\alpha_3=\dfrac{1}{3\sqrt{170}}$ $x'_3=0$ $y'_3=7$ $z'_3=-6$

$\beta_1=\dfrac{1}{4\sqrt{2}}$ $u_1=3$ $v_1=2$ $w_1=1$

$$\beta_2 = \frac{1}{4\sqrt{6}} \qquad u_2 = 5 \qquad v_2 = -2 \qquad w_2 = -1$$

$$\gamma_1 = \frac{1}{2\sqrt{114}} \qquad x_1 = -1 \qquad y_1 = 1 \qquad z_1 = 0$$

$$\gamma_2 = \frac{1}{2\sqrt{10}} \qquad x_2 = -9 \qquad y_2 = -7 \qquad z_2 = +2$$

$$\gamma_3 = \frac{1}{2\sqrt{285}} \qquad x_3 = 4 \qquad y_3 = 1 \qquad z_3 = -3$$

The definition of (m, p) SU(3) ⊗ SU(2) tensors is unambiguous, except for the tensors of (8,2); (8,2); (8,2); (8,4); (8,4) (8,4); (27,2); (27,2).

The definitions of these tensors can be found in Table Ia. With that definition the states of different irreducible representations are orthogonal.

Table Ia

$$d_1 = \frac{1}{3\sqrt{10}} d'_1 \qquad (8,4)_1$$

$$d_2 = \frac{1}{3\sqrt{114}} (d'_1 - 2\, d'_2) \qquad (8,4)_2$$

$$d_3 = \frac{1}{6\sqrt{285}} (16\, d'_1 - 13\, d'_2 + 19\, d'_3) \qquad (8,4)_3$$

where

$$d'^{\;b}_{1a\;,\alpha\beta\gamma} = \varepsilon^{b\;u}(R^{r\omega}_{a\alpha,t\beta,u\omega,r\gamma} + R^{r\omega}_{a\alpha,t\gamma,u\omega,r\beta}) + \text{cyclical permutation in } (\alpha, \beta, \gamma) - \frac{1}{3}\,\delta^b_a\,\text{Trace}$$

$$d'^{\;b}_{2a\;,\alpha\beta\gamma} = \varepsilon^{btu}(R^{r\omega}_{t\beta\;,u\omega,r\gamma,a\alpha} + R^{r\omega}_{t\beta,u\omega,r\alpha,a\gamma}) + \text{cyclical permutation in } (\alpha, \eta, \gamma) - \frac{1}{3}\,\delta^b_a\;\text{Trace}$$

$$d'^{\;b}_{3a\;,\alpha\beta\gamma} = \varepsilon^{btu}(R^{r\omega}_{u\omega,r\gamma,a\alpha,t\beta} + R^{r\omega}_{u\omega,r\alpha,a\gamma,t\beta}) + {} + \text{cyclical permutation in } (\alpha, \beta, \gamma) - \frac{1}{3}\,\delta^b_a\,\text{Trace}$$

$$b_1 = \frac{\sqrt{7}}{2\sqrt{39}} b'_1 \qquad (8,2)_1$$

$$b_2 = \frac{1}{6\sqrt{1105}} (29\, b'_1 - 26\, b'_2) \qquad (8,2)_2$$

$$b_3 = \frac{1}{12\sqrt{85}} (22\, b'_1 + 9 b'_2 - 34\, b'_3) \qquad (8,2)_3$$

where

$$b'^{b}_{1a} = \varepsilon^{\varrho\pi}\,\varepsilon^{btu}\,R^{\gamma\omega}_{a\alpha t\varrho,u\omega,r\pi} - \frac{1}{3}\,\delta^{b}_{a}\,\varepsilon^{\varrho\pi\,ptu}\,R^{r\omega}_{p\alpha,tv,u\omega,r\pi}$$

$$b'^{b}_{2a} = \varepsilon^{\varrho\pi}\,\varepsilon^{btu}\,R^{\gamma\omega}_{t\varrho,u\omega,r\pi.a\alpha} - \frac{1}{3}\,\delta^{b}_{a}\,\mathrm{Trace}$$

$$b'^{\,b}_{3a} = \varepsilon^{\varrho\pi}\,\varepsilon^{b\,tu}\,R^{\gamma\omega}_{t\varrho,u\omega,r\alpha,a\pi} - \frac{1}{3}\,b^{b}_{a}\,\mathrm{Trace}$$

$$L_1 = \frac{1}{8}\,L'_1 \tag{27,2)$_1$}$$

$$L_2 = \frac{1}{8\sqrt{3}}\,L'_3 \tag{27,2)$_2$}$$

where

$$L'^{\,cd}_{1\,ab,\alpha} = \varepsilon^{\varrho\varkappa}(\varepsilon^{crs}\,R^{d\omega}_{r\omega,,a\varrho,b\varkappa.s\alpha} + \varepsilon^{drs}\,R^{c\omega}_{r\omega,a\varrho,b\varkappa,s\alpha}) - \mathrm{Trace\,SU(3)}$$

$$L'^{\,cd}_{2\,ab,\alpha} = \varepsilon^{\varrho k}(\varepsilon^{crs}\,R^{d\omega}_{r\alpha,a\varrho,bk,r\omega} + \varepsilon^{drs}\,R^{c\omega}_{s\alpha,a\varrho,bk,s\alpha}) - \mathrm{Trace\,SU(3)}$$

For the sake of simplicity, we assume that formulae (A) and (B) are also probably good approximations in the 540-plet as "semi-empirical" relations. The mixing of different states of subspaces of SU(3)⊗SU(2) is determined in part by that requirement.

The decomposition of the 540-plet with respect to SU(3) × SU(2) can be found in Table I.

III. The baryon-meson vertex, decay amplitudes

We propose to assign the resonances $3/2^-$ and $5/2^-$ to a 540-plet. For two-body decays we can derive relations from the investigation of the three point function. For collinear processes it is known that the maximal possible symmetry, consistent with Lorentz invariance is the $\mathrm{SU}(6)_W$ symmetry. Therefore, we require the $\mathrm{SU}(6)_W$ symmetry for vertex functions.

In order to define the particles in terms of quarks, we use $\mathrm{SU}(6)_\sigma$ (static SU(6)). We need, therefore, an $\mathrm{SU}(6)_q \otimes \mathrm{SU}(6)_{\bar{q}}$ symmetry from which we can pick up both an SU(6)s and an $\mathrm{SU}(6)_W$.

We have first to identify the $\mathrm{SU}(6)q \times \mathrm{SU}(6)_{\bar{q}}$ representation. We have chosen $(\underline{105}, \underline{6}^*)$, which decomposes under each SU(6) into

$$\underline{105'} \otimes \underline{6}^* = \underline{540} \oplus \underline{70} \oplus \underline{20}\,.$$

The W—S transition will mix all these representations. The $\mathrm{SU}(6)_\sigma$ $\underline{56}^+$ representation belongs to $(\underline{56}.\underline{0})$ as $\mathrm{SU}(6)_q \otimes \mathrm{SU}(6)_{\bar{q}}$ representation and $\underline{35}^-$ belongs to $(\underline{6}.\underline{6}^*)$

$$\underline{6} \otimes \underline{6}^* = \underline{35} \oplus \underline{1}\,.$$

Therefore the $\mathrm{SU}(6)_W$ invariant vertex turns out to be

$$g\, P_W^{[AB]C}\, B_{W,\{BCD\}}\, M_{WA}{}^{D}$$

where $\mathrm{P}_W^{[ABC]}$, $\mathrm{B}_{W,\{BCD\}}$, $\mathrm{M}_{WA}{}^{D}$ are the tensors of the $\underline{70}, \underline{65}, \underline{35}$ $\mathrm{SU}(6)_W$ representations, respectively. The W—S transition will bring in the tensors of the $\underline{540}$, $\underline{20}$ and $\underline{1}$ representations, so that we can give the vertex function in terms of the states of the $\mathrm{SU}(6)_\sigma$ in U-chain. Fitting the coupling constant we can determine the widths of the two-body decays and the branching ratios.

If, in the final state, there are only particles with spins $1/2^+$ and 0^-, the baryons $3/2^-$ and $5/2^-$ can decay only in a *d* wave, while for the baryons $1/2^-$ only the *s* wave channel is open. Thus, in these two cases the dimensions of coupling constant are different. The difficulty has been circumvented by introducing two different coupling constants. In the decay widths k^5/m^2 (*d*-wave) and k/m^2 (*s*-wave) phase corrections have been taken into account, where k is the momentum of the final products in the centre of the mass system and m is the mass of the decaying particle.

IV. Classification; results

The assignment of the resonances $5/2^-$ $\mathrm{N}^*_{1/2}$ (1670) and $5/2^-$ Y^*_1 (1768) to the 540-plet is unique. The coupling constant has been calculated by fitting the experimental value of the width of the $5/2^-$ Y^*_1 (1768) resonance. For the $5/2^-$ $\mathrm{N}^*_{1/2}$ (1670) the predicted width is reasonable. The branching ratios do not contradict the experimental values, although the $\Sigma\,\pi$ decay of the resonance $5/2^-$ Y^*_1 seems to be more forbidden than our model indicates.

For the resonances $3/2^-$ $\mathrm{N}^*_{1/2}$ (1525) and $3/2^-$ Y^*_0 (1518), $3/2^-$ Y^*_0 (1700) we have various possibilities since there are five states in the U-chain for each of them with identical quantum numbers J, Y, I (see Table II). These resonances have been assigned by "optimal fitting". Since the mass formula is independent of the N spin, the linear combinations of states differing only in N spin can be physical states. On classifying $3/2^-$ Y^*_0 (1518) this fact has been taken into account (see Tables II—IV).

The assignment of the resonances

$$3/2^- Y_1^*(1660), 3/2^- \Xi_{1/2}^{-*}(1815), 1/2^- N_{1/2}^*(1570),\ 1/2^- N_{1/2}^*(1700),$$
$$1/2^- N_{3/2}^*(1670),\ 1/2^- Y_0^*(1405),\ 1/2^- Y_0^*(1670)$$

Table II

SU(4) states	J	Y	I	N	S	$C_2^{(4)}$	Mass
$\vert 8,6 >$	5/2	±1	1/2	3/2	1	39/4	1670* 1870
$\vert 8,6 >$	5/2	0	1	2	1/2	15	1770*
$\vert 8,6 > + \sqrt{2}\,\vert 1,6 >$	5/2	0	0	2	1/2	15	1610
$\sqrt{2}\,\vert 8,6 > - \vert 1,6 >$	5/2	0	0	0	3/2	5	1793
$\vert 27 >$	3/2	±2	1	1	1/2	8	1660 2060
$\vert 10 >$	3/2	±2	0	1	1/2	8	1500 1900
$\vert 27,4 > + \vert 10,4 >$	3/2	±1	3/2	3/2	0	55/4	1760 1960
$\vert 27,4 > - \vert 10,4 >$	3/2	±1	3/2	1/2	1	39/4	1600 1800
$6\sqrt{19}\,\vert 27 > + 2\sqrt{95}\,\vert 10^* > - 3\sqrt{57}\,\vert 8 >_1 - \sqrt{5}\,\vert 8 >_2 - 8\sqrt{2}\,\vert 8 >_3$	3/2	+1	1/2	1/2	1	15/4	1703
$3\sqrt{19}\,\vert 27 > \pm 3\sqrt{95}\,\vert 10^* > + 6\sqrt{57}\,\vert 8 >_1 + 6\sqrt{5}\,\vert 8 >_2 - 9\sqrt{2}\,\vert 3 >_3$	3/2	±1	1/2	3/2	0	55/4	1525* 1725
$3\sqrt{19}\,\vert 27 > - \sqrt{95}\,\vert 10^* > + 8\sqrt{5}\,\vert 8 >_2 + 7\sqrt{2}\,\vert 8 >_3$	3/2	+1	1/2	3/2	1	39/4	1565
$6\sqrt{19}\,\vert 27 > - 2\sqrt{95}\,\vert 10 > - 3\sqrt{57}\,\vert 8 >_1 + 5\sqrt{5}\,\vert 8 >_2 + 2\sqrt{2}\,\vert 8 >_3$	3/2	−1	1/2	1/2	1	15/4	1865
$3\sqrt{19}\,\vert 27 > + \sqrt{95}\,\vert 10 > - 4\sqrt{5}\,\vert 8 >_2 - 13\sqrt{2}\,\vert 8 >_3$	3/2	−1	1/2	3/2	1	39/4	1765
$- 2\sqrt{95}\,\vert 10^* > + 4\sqrt{5}\,\vert 8 >_2 - 25\sqrt{2}\,\vert 8 >_3$	3/2	+1	1/2	1/2	1	39/4	1565
$2\sqrt{95}\,\vert 10 > + 16\sqrt{5}\,\vert 8 >_2 - 5\sqrt{2}\,\vert 8 >_3$	3/2	−1	1/2	1/2	1	39/4	1765
$3\sqrt{19}\,\vert 27 > - \sqrt{95}\,\vert 10^* > + 57\vert 8 >_1 - 7\sqrt{5}\,\vert 8 >_2 + \sqrt{2}\,\vert 8 >_3$	3/2	+1	1/2	3/2	0	39/4	1615
$3\sqrt{19}\,\vert 27 > + \sqrt{95}\,\vert 10^* > + 57\vert 8 >_1 - \sqrt{5}\, 8 >_2 + 11\sqrt{2}\,\vert 8 >_3$	3/2	−1	1/2	3/2	0	39/4	1815
$\vert 27 >$	3/2	0	2	1	1/2	15	1995

$\sqrt{114}\,\vert\,27> + \sqrt{95}\,\vert\,10> - \sqrt{95}\,\vert\,10^*> - 4\sqrt{5}\,\vert\,8>_2 + 6\sqrt{2}\,\vert\,8>_3$	$3/2_S$	0	1	2	1/2	15	1670
$42\sqrt{19}\,\vert\,27> - 7\sqrt{38}\,\vert\,8>_1 - 14\sqrt{30}\,\vert\,8>_2 + 42\sqrt{3}\,\vert\,8>_3 + 10\sqrt{133}\,\vert\,1>$	$3/2_A$	0	0	1	1/2	5	1752
$3\,\vert\,8>_2 + \sqrt{10}\,\vert\,8>_3$	3/2	0	0	1	1/2	5	1752
$2\sqrt{114}\,\vert\,27> - 2\sqrt{95}\,\vert\,10> - 2\sqrt{95}\,10^*> + \sqrt{57}\,\vert\,8>_1 + 9\sqrt{5}\,\vert\,8>_2 - 4\sqrt{2}\,\vert\,8>_3$	3/2	0	1	1	1/2	9	1835
$2\sqrt{114}\,\vert\,27> + 2\sqrt{95}\,\vert\,10> + 2\sqrt{95}\,\vert\,10^*> + \sqrt{57}\,\vert\,8>_1 + 3\sqrt{5}\,\vert\,8>_2 - 14\sqrt{2}\,\vert\,8>_3$	3/2	0	1	1	1/2	9*	1835
$\sqrt{114}\,\vert\,27> - \sqrt{95}\,\vert\,10> + \sqrt{95}\,\vert\,10^*> - 2\sqrt{57}\,\vert\,8>_1 - 2\sqrt{5}\,\vert\,8>_2 + 3\sqrt{2}\,\vert\,8>_3$	3/2	0	1	0	3/2	5	1860
$\sqrt{114}\,\vert\,27> - 3\sqrt{95}\,\vert\,10> + 3\sqrt{95}\,\vert\,10^*> + 8\sqrt{57}\,\vert\,8>_1 - 12\sqrt{5}\,\vert\,8>_2 + 18\sqrt{2}\,\vert\,8>_3$	$3/2_S$	0	1	1	1/2	15	1680
$\sqrt{95}\,\vert\,10> + \sqrt{95}\,\vert\,10^*> - 12\sqrt{5}\,\vert\,8>_2 - 20\sqrt{2}\,\vert\,8>_3$	$3/2_A$	0	1	1	1/2	15	1680
$\sqrt{19}\,\vert\,27> - 4\sqrt{38}\,\vert\,8>_1 - 8\sqrt{30}\,\vert\,8>_2 + 24\,\vert\,8>_3 + 5\sqrt{133}\,\vert\,1>$	3/2	0	1	1	1/2	15	1520*
$3\sqrt{27} > - 4\sqrt{2}\,\vert\,8>_1 - 7\,\vert\,1>$	3/2	0	0	2	1/2	15	1520
$3\sqrt{19}\,\vert\,27> - 2\sqrt{38}\,\vert\,8>_1 + 2\sqrt{30}\,\vert\,8>_2 - 6\sqrt{3}\,\vert\,8>_3 - \sqrt{133}\,\vert\,1>$	3/2	0	0	1	3/2	5	1700*
	1/2	±2	1	1	1/2	8	1603 2000
	1/2	±2	1	0	1/2	8	1600 2000
	1/2	±2	0	1	1/2	8	1445 1845
$\vert\,10> + \vert\,27>_2$	1/2	±1	3/2	1/2	0	55/4	1705 1905
	1/2	±1	3/2	1/2	1	39/4	1750 1950
	1/2	±1	3/2	1/2	0	39/4	1800 2000
	1/2	±1	1/2	3/2	1	39/4	1510 1710
	1/2	±1	1/2	1/2	1	39/4	1610 1810

Table II (continued)

SU(4) states	J	Y	I	N	S	$C_8^{(4)}$	Mass
$\frac{21}{\sqrt{546}} \vert 8 >_1 + \frac{29}{\sqrt{2210}} \vert 8 >_2 - \frac{6}{\sqrt{170}} \vert 8 >_3 - \vert 10^* > - \frac{\sqrt{3}}{2\sqrt{5}} \vert 27 >_1 - \frac{\sqrt{3}}{2\sqrt{5}} \vert 27 >_2$	1/2	±1	1/2	1/2	0	39/4	1560 1760
	1/2	±1	1/2	1/2	1	15*/4	1650 1850
$\frac{3}{2\sqrt{547}} \vert 8 >_1 - \frac{7}{2\sqrt{2210}} \vert 8 >_2 + \frac{6}{\sqrt{170}} \vert 8 >_3 - \frac{1}{2} \vert 10^* > - \frac{\sqrt{3}}{2\sqrt{5}} \vert 27 >_1 - \frac{\sqrt{3}}{2\sqrt{3}} \vert 27 >_2$	1/2	±1	1/2	1/2	0	15*/4	1700 1900
	1/2	±1	1/2	1/2	0	55*/4	1420 1620
	1/2	0	2	1	1/2	15	1940
	1/2	0	2	0	1/2	15	1940
	1/2	0	1	1	1/2	9	1781
	1/2	0	1	1	1/2	9*	1781
	1/2	0	1	0	1/2	6	1856
	1/2	0	1	0	1/2	5	1856
	1/2	0	1	1	1/2	15	1620
	1/2	0	1	1	1/2	15	1620
	1/2	0	1	0	1/2	15	1620
$\frac{2\sqrt{7}}{\sqrt{13}} \vert 8_1 > + \frac{2\sqrt{3}}{\sqrt{1105}} \vert 8 >_2 - \frac{3}{\sqrt{155}} \vert 8 >_3 + \sqrt{\frac{3}{5}} \vert 27 >_1 + \frac{1}{\sqrt{5}} \vert 27 >_2 - \sqrt{2} \vert 1 >$	1/2	0	0	1	1/2	15	1465
	1/2	0	0	0	1/2	9	1625
	1/2	0	0	0	1/2	9*	1625
$-\frac{2\sqrt{7}}{\sqrt{13}} \vert 8 >_1 - \frac{2}{\sqrt{3315}} \vert 8 >_2 + \frac{1}{\sqrt{255}} \vert 8 >_3 + \frac{\sqrt{3}}{2\sqrt{5}} \vert 27 >_1 + \frac{1}{2\sqrt{5}} \vert 27 >_2 - \frac{1}{3\sqrt{2}} \vert 1 >$	1/2	0	0	1	3/2	5	1650
	1/2	0	0	1	1/2	5	1660
	1/2	0	0	1	1/2	5	1660

Table III

States of 70_W $\vert SU(3) \otimes SU(2) ; J_3 >$	States of $540_\rho - 70_\sigma - 20_\sigma$ $\vert SU(6) ; SU(3) \otimes SU(2) ; J_3 >$
$\vert 10,2 ; 1/2 >$	$\sqrt{\frac{2}{3}} \vert 540 ; 10,4 ; 1/2 > + \sqrt{\frac{1}{3}} \vert 540 ; 10.2 ; 1/2 >$
$\vert 8,4 ; 3/2 >$	$- \sqrt{\frac{1}{5}} \vert 540 ; 8,6 ; 3/2 > + \frac{1}{2\sqrt{10}} \vert 540 ; (8,4)_1 ; 3/2 \rangle -$
	$- \frac{115}{2\sqrt{114}} \vert 540 ; (8,4)_2 ; 3/2 > - \frac{\sqrt{3}}{\sqrt{95}} \vert 540 ; (8,4)_3 ; 3/2 > -$
	$- \frac{1}{2} \vert 70 ; 8,4 ; 3/2 >$
$\vert 8,4 ; 1/2 >$	$\sqrt{\frac{3}{10}} \vert 540 ; 8,6 ; 1/2 > + \frac{1}{6\sqrt{10}} \vert 540 ; (8,4)_1 ; 1/2 > -$
	$- \frac{5}{2\sqrt{114}} \vert 540 ; (8,4)_2 ; 1/2 > - \frac{1}{\sqrt{280}} \vert 540 ; (8,4)_3 ; 1/2 > -$
	$- \frac{1}{6} \vert 70 ; 8,4 ; 1/2 > + \frac{1}{6} \vert 70 ; 8,2 ; 1/2 > +$
	$+ \frac{2}{3\sqrt{7}} \vert 20 ; 8,2 ; 1/2 > - \frac{19}{6\sqrt{182}} \vert 540 ; (8,2)_1 ; 1/2 > +$
	$+ \frac{7\sqrt{15}}{2\sqrt{442}} \vert 540 ; (8,2)_2 ; 1/2 > - \frac{\sqrt{5}}{\sqrt{102}} \vert 540 ; (8,2)_3 ; 1/2 >$
$\vert 8,2 ; 1/2 >$	$\frac{1}{6\sqrt{2}} \vert 540 ; (8,4)_1 ; 1/2 > + \frac{7}{2\sqrt{114}} \vert 540 ; (8,4)_2 ; 1/2 > -$
	$- \frac{2\sqrt{5}}{\sqrt{57}} \vert 540 ; (8,4)_3 ; 1/2 > - \frac{23}{6\sqrt{182}} \vert 540 ; (8,2)_1 ; 1/2 > +$
	$+ \frac{3\sqrt{15}}{2\sqrt{442}} \vert 540 ; (8,2)_2 ; 1/2 > + \frac{\sqrt{10}}{\sqrt{51}} \vert 540 ; (8,2)_3 ; 1/2 > +$
	$+ \frac{1}{6} \vert 70 ; 8,4 ; 1/2 > - \frac{1}{6} \vert 70 ; 8,2 ; 1/2 > -$
	$- \frac{2}{3\sqrt{7}} \vert 20 ; 8,2 ; 1/2 >$
$\vert 1/2 ; 1/2 >$	$\frac{2\sqrt{5}}{3\sqrt{7}} \vert 540 ; 1,4 ; 1/2 > - \frac{2}{3} \vert 540 ; 1,2 ; 1/2 > -$
	$- \frac{1}{3} \vert 70 ; 1,2 : 1/2 > - \frac{2\sqrt{2}}{3\sqrt{7}} \vert 20 ; 1,4 ; 1/2 > -$

has been done on the basis of the mass formulae. (Parameters of the mass formulae could be calculated from the mass values of the resonances already assigned.)

Table IV

Name	SU(4) state $\| J^P\ Y\ I\ N\ S\ C_2^{(4)}\rangle$	Mass (exp.) MeV	Mass (theor.) MeV	Width (exp.) MeV	Width (theor.) MeV	Final products	Branching ratios % (exp.)	Branching ratios % (theor.)
$N^*_{1/2}$	$\| 5/2^-\ 1\ 1/2\ 3/2\ 1\ 39/4\rangle$	1670 ± 10	1670*	140	100 ± 20	$N\pi$ ΛK $N\eta$	40 small small	35 0 5
Y^*_1	$\| 5/2^-\ 0\ 1\ 2\ 1/2\ 15\rangle$	1768 ± 4	1768*	89 ± 12	89*	$N\bar{K}$ $\Lambda\pi$ $\Sigma\pi$ $\Sigma\eta$	49 17 < 1 2	45 20 7 0.1
$N^*_{1/2}$	$\| 3/2^-\ 1\ 1/2\ 3/2\ 0\ 55/4\rangle$	1525 ± 8	1525*	105	110 ± 22	$N\pi$ $N\eta$	65 —	65* 1
Y^*_0	$1.5\,\| 3/2^-\ 0\ 0\ 2\ 1/2\ 15\rangle +$ $+\ \| 3/2^-\ 0\ 0\ 1\ 1/2\ 15\rangle$	1518.8 ± 1.5	1520*	16 ± 2	14 ± 3	$N\bar{K}$ $\Sigma\pi$	39 ± 5 51 ± 6	36 ± 8 47 ± 10
Y^*_0	$\| 3/2^-\ 0\ 0\ 1\ 3/2\ 5\rangle$	1700 ± 10	1700*	40 ± 10	60 ± 15	$N\bar{K}$ $\Sigma\pi$	20 seen	15 85
Y^*_1	$\| 3/2^-\ 0\ 1\ 2\ 1/2\ 15\rangle$	1660 ± 2	1670 ± 20	50	65 ± 18	$\Sigma\pi$ $\Lambda\pi$ NK	$\sim$30 (?) $\sim$ 5 (?) small	35 1 4
$N^*_{1/2}$	$\| 1/2^-\ 1\ 1/2\ 1/2\ 0\ 39/4\rangle$	1570 ± 10	1560 ± 20	130	250 ± 40	$N\pi$ $N\eta$	$\sim$30 $\sim$70	99 1
$N^*_{3/2}$	$\| 1/2^-\ 1\ 3/2\ 1/2\ 0\ 55/4\rangle$	1670 ± 10	1730 ± 30	$\sim$180	220 ± 40	$N\pi$	40	40*
$N^*_{1/2}$	$\| 1/2^-\ 1\ 1/2\ 1/2\ 0\ 15/4\rangle$	1700 ± 20	1700 ± 30	240	240*	$N\pi$ ΛK ΣK $N\eta$	100 — — —	88 11 } 1
Y^*_0	$\| 1/2^-\ 0\ 0\ 1\ 1/2\ 15\rangle$	1405 ± 0.5	1465 ± 20	35	210 ± 40	Σ	100	100
? Y^*_0	$\| 1/2^-\ 0\ 0\ 1\ 3/2\ 5\rangle$	1670	1650 ± 20	18	480 ± 80	$\Sigma\pi$ $\Lambda\eta$ NK	— seen seen	$\sim$100 } small
? $\Xi^*_{1/2}$	$\| 3/2^-\ —1\ 1/2\ 1/2\ 1\ 15/4\rangle$	1815	1860 ± 30	16 ± 8	28 ± 6	K ΣK	$\sim$65 (?) $\sim$10 (?) —	$\sim$1 $\sim$60 $\sim$2

Formula (A) contradicted experience. Mass values predicted by formula (B) can be found in Table IV. On the basis of this classification and the vertex function the widths and branching ratios for these resonances could be calculated.

The coupling constant for the resonances decaying in *s* wave has been determined from the width of the $1/2^-$ $N^*_{1/2}$ (1700) (see Table IV). The results for $3/2^-$ Y^*_1 (1660) and $3/2^-$ $\Xi^{-*}_{1/2}$ (1815) do not conflict with experimental values. The predications for the resonances $1/2^-$ $N^*_{1/2}$ (1700) and $1/2^-$ $N^*_{3/2}$ (1670) are also reasonable.

The results for $1/2^-$ $N^*_{1/2}$ (1570), $1/2^-$ Y^*_0 (1450) and $1/2^-$ Y^*_0 (1670) concerning both the widths and the branching ratios strongly contradict the experimental data.

If a state in the U chain does not contain an octet, decuplet or singlet, its two-body decay through strong interaction is forbidden. From Table II we can see that there are 23 states with this property. According to the mass formulae, 17 of them have mass values about 2000 MeV. Therefore, their widths could be large by the three-body decays. For the remaining five states the predicted mass values are about 1500 MeV.

Here three-body decays are either forbidden by conservation of energy and hypercharge, or the probability of the decays is very small. When asking why these resonances did not come up so far, we must bear in mind that the simplified mass formulae may not be too reliable.

As for the N (1570), Y (1405) and Y (1670) particles, it is possible that if we had identified them on the basis of the decay widths and branching ratios we should have obtained better results. These calculations, however, would have been too lengthy so they have been abandoned.

Summing up, we may say that in the case of the $5/2^-$ and $3/2^-$ baryons our classification seems to be quite acceptable, the difficulties connected with the $1/2^-$ and the unabsorbed particles may be ascribed to the inadequacy of the simplified mass formula.

REFERENCES

1. ROSENFELD et al. Rev. Mod. Phys., January 1967.
2. M. A. B. BÉG and V. SINGH, Phys. Rev. Letters, **13**, 509, 1964.
3. A. PAIS, Phys. Rev. Letters, **13**, 175, 1964.
4. R. GATTO, L. MAIANI and G. PREPARATA, Phys. Rev., **142**, 1135, 1966.
5. S. MESHKOV and G. B. YODH, Phys. Rev. Letters, **13**, 1966.
6. H. RUEGG, W. RÜHL and T. S. SANTHANAM, CERN preprint TH. 709, 1966.
7. I. MONTVAY and T. NAGY, Acta Phys. Hung., **21**, 345, 1966.
8. B. BÉG and V. SINGH, Phys. Rev. Letters, **13**, 418, 1964

БАРИОННЫЕ РЕЗОНАНСЫ ОТРИЦАТЕЛЬНОЙ ЧЕТНОСТИ В СИММЕТРИИ SU(6)

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Резюме

Соотношение распада барионных резонансов отрицательной четности сравнивается с предсказанием SU_W (6) путем определения данных резонансов к сверхмультиплетам 540. Исследуются также некоторые «полуэмпирические» формулы. Выявляется, что настоящие экспериментальные данные соответствуют определяемым. Однако, предполагается, что не все барионные резонансы отрицательной четности со значением спина $1/2^-$, $3/2^-$, $5/2^-$ пригодны для 540-плетов без противоречия экспериментальным результатам.

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THE MANY-BODY PROBLEM IN TERMS OF PARTICLE GROUP FUNCTIONS

By

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In the treatment of many-particle systems the particle correlations can be taken into account to a certain extent at the very beginning by choosing particle-group basis functions. However, when these basis functions do not satisfy a certain restrictive condition ("strong orthogonality") some mathematical difficulties arise. Using a previously discovered way of overcoming these difficulties [3] we develop practical methods to solve this problem. The Green's function formalism of Martin and Schwinger is applied using field operators for the particle groups with intrinsic degrees of freedom. Besides the possible computational advantages and the intrinsic interest of the problem this theory yields some possibilities for an ab initio microscopic theory of condensed media. In addition, it can also be useful in throwing light on some questions of relativistic field theory of structured particles.

1. Introduction

The formulation of the many-body problem in terms of particle-group basis functions or equivalently the many-body problem for composite particles has a long history. However, to construct a consistent theory without a restrictive condition for the basis functions ("strong orthogonality" [1]) is not a trivial task at all. The main difficulty of a direct approach lies in the fact that the annihilation and creation operators of the composite particles (particle groups) as constructed from the field operators of the elementary particles turn out to be linearly dependent. This is reflected by the (anti) commutation relations of these operators, also. Hence it is not clear how to make computations with these quantities or how to interpret the results. Following DYSON's treatment for spin waves [2] GIRARDEAU has succeeded [3] in finding a suitable formalism for this problem. The main trick of this formalism is that there exists an H_i "ideal" Hilbert space with the properties that a certain section of H_i is isomorphic with the Hilbert space of physical states, but the annihilation and creation operators as transformed to H_i obey the usual (anti) commutation rules. Hence, after having determined the form of various operators in H_i we can proceed with the computations in the usual way, provided we restrict ourselves to the suitable section of H_i. The restriction to this section of H_i means a super selection rule. GIRARDEAU proposed the use of a generalized grand canonical ensemble in which this superselection rule is satisfied on the average. He proved that in the thermodynamic limit the physical consequence agrees with those that can be derived using the grand canonical ensemble in which the super selection rule is exactly satisfied. Starting from this basis, the present paper is an attempt to construct a practical method of treating the many-body problem in terms of particle group basis functions.

The motivation for such a theory is obvious; most systems have the property that the motion of some particles is strongly correlated, while these clusters of particles are relatively independent. This property manifests itself in superconductivity abruptly in a drastic way but it is more general, e.g. this is the precise meaning of the somewhat intuitive statement that condensed media consist of atoms or molecules. It is quite natural, therefore, to start from the beginning with a set of basis functions that take into account the correlations characteristic for the system, instead of with the usual choice of plane waves.

This method has already been applied in quantum chemistry with special particle group functions [4], as well as in the theory of superconductivity [5]. In the former case it yields a very promising method of treating electron correlations in molecules, while in the latter case it enables us to treat superconductivity despite the "nonperturbative" nature of the problem. Hence we feel it is worth while to investigate this line.

The subject of a paper by SAHLIN and SCHWARTZ [6] is similar to that of this paper. They treated a many-body system of composite particles consisting of two elementary particles. We feel that the method considered in the present paper has some advantages over that of [6]; we can treat any structured particles (particle groups) on an equal footing with the usual methods of the many-body theory.

Besides the intrinsic interest and the possible computational advantages, this theory can be useful in clarifying some questions of relativistic field theory. It is not clear to what extent a composite field theory is equivalent to a canonically quantized phenomenological field theory; it can be shown that the (anti) commutator of a structured field need not vanish outside the light cone, etc. [7]. The difficulties met here are of the same nature, it may be instructive to know how to overcome them in nonrelativistic field theory with the aid of the DYSON transformation.

In addition, this theory yields a possibility for an ab initio microscopic approach to the physics of condensed media on an equal footing.*

It should be remarked that the formulation of GIRARDEAU has some similarity with the pseudo potential theory. In the theory of pseudo potentials

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* Generally, the various excitation modes in condensed media (excitons, phonons, etc.) are treated by methods suited for the specific problems. Sometimes these are derived on she basis of classical considerations. For an example of the insufficiency of such a theory, tee [8].

a model potential is added to the Hamiltonian of the problem in order to impose some conditions on the wave function [9]. However, in pseudo potential theory this trick is always employed within the framework of an approximation method while the formalism of [3] is exact.

In Section 1 the theory of thermodynamic Green's functions [10] is formulated in terms of particle-group field operators. Section 2 is devoted to the boundary conditions and analytic structure of the propagators. In Section 3 we discuss Green's function equations. A formal tool (cf. e.g. [11] will be used in order to gain some further insight into the properties of these equations. A perturbational technique based on diagrams is developed in Section 4. We derive some results, needed for our considerations, of [3] in a more general from in Appendix A. In order to make contact with the macroscopic parameters of the system Appendix B is devoted to the determination of the thermodynamic potential.

2. Green's functions

According to [3] and the results of Appendix A the (generalized) free energy operator is of the form

$$\mathscr{K} = H - \sum_{q=1}^{m} \mu_q N_q + \sum_{l=1}^{L} \sum_{r \leqq q} \gamma_{l;rq} I_{l;rq} = H_0 + V_{\text{eff}}\,, \tag{1}$$

where H is the Hamiltonian of the system; N_q, the number of particle groups of the q-th type; $I_{l;rq}$, the exchange operators introduced in [3]; μ_q and $\gamma_{l;rq}$ are the Lagrange multipliers connected with conditions (A.6) and (A.4). The detailed form of these operators is given in (A.5) and (A.7)—(A.13). The creation and annihilation operators obey the usual (anti)commutation rules, as shown in (A.3). The operator $\mathscr{K}$ can be used to define the Heisenberg picture with the usual prescription, i.e. for any operator O we have the equation ($\hbar = 1$)

$$i \frac{d}{dt} O = [O, \mathscr{K}]_- \,. \tag{2}$$

With the formal solution

$$O(t) = e^{i\mathscr{K}t}\, O\, e^{-i\mathscr{K}t}$$

the operators of Appendix A can be transformed to this picture. In the following we suppose that this has been done. Now let us define Green's functions as averages of time-ordered products of particle group operators. The simplest Green's functions are

$$\begin{aligned} G(r\alpha \,|\, r\alpha_1) &= -\,i\,\langle T a(r\alpha)\, a^+(r\alpha_1)\rangle\,, \\ G(r\alpha q\beta \,|\, r\alpha_1\, q\beta_1) &= (-i)^2 \langle T a(r\alpha)\, a(q\beta)\, a^+(r\alpha_1)\, a^+(q\beta_1)\rangle\,, \end{aligned} \tag{3}$$

and so on. The chronological operator includes change of sign depending on the boson- or fermion-like nature of the commutation rules between the time-ordered operators. In the formulas we have suppressed time variables; in the following this convention will be followed for all dummy variables and indices. Using the standard group theoretical apparatus of quantum theory a lot of information about these Green's functions can be obtained in the usual way. For instance invariance under translation in time and space assures conservation of energy and momentum, etc. The symmetry group of the Hamiltonian can be a lower one than the full Galilean group. However, it is much more interesting when Galilean invariance is spontaneously broken, i.e. the Hamiltonian of the system is invariant under the Galilean group yet the Green's functions (3) have the symmetry of a space group. We met this situation in an ab initio microscopic theory of solids [8]. However, we do not go into details on this point.

The information regarding the physical properties of the system can be obtained from the analytical structure of the Green's functions. The last of this Section is devoted to this question as well as to the problem of boundary conditions. Two auxiliary quantities will be introduced [10] by the definitions

$$\begin{aligned} G^{>}(r\alpha t\,|\,r\alpha_1 t_1) &= (-i)\,\langle a(r\alpha t)\,a^{+}(r\alpha_1 t_1)\rangle, \\ G^{<}(r\alpha t\,|\,r\alpha_1 t_1) &= (\pm)_{rr}(-i)\,\langle a^{+}(r\alpha_1 t_1)\,a(r\alpha t)\rangle\,, \end{aligned} \tag{4}$$

where $(\pm)_{rq}$ means a positive (negative) sign if the operators $a(r\alpha)$ and $a(q\beta)$ obey boson-like (fermion-like) commutation rules. In this symbol the indices will be suppressed when they are evident. From the cyclic invariance of trace

$$G^{<}(r\alpha 0\,|\,r\alpha_1 t_1) = \pm\, G^{>}(r\alpha - i\beta\,|\,r\alpha_1 t_1)\,. \tag{5}$$

Extending the definition of the chronological operator for the imaginary axis in the interval $0 \leqslant it \leqslant \beta$ in the following way

$$\begin{aligned} G(r\alpha t\,|\,r\alpha_1 t_1) &= G^{>}(r\alpha t\,|\,r\alpha_1 t_1)\,, \quad \text{if} \quad it > it_1\,, \\ &= G^{<}(r\alpha t\,|\,r\alpha_1 t_1)\,, \quad \text{if} \quad it < it_1\,, \end{aligned}$$

one can prove that

$$G(r\alpha 0\,|\,r\alpha_1 t_1) = \pm\, G(r\alpha - i\beta\,|\,r\alpha_1 t_1)\,,$$

or taking into account the invariance of the Hamiltonian against translations in time

$$G(r\alpha\,|\,r\alpha_1; t) = \pm\, G(r\alpha\,|\,r\alpha_1; t - i\beta)\,. \tag{6}$$

This invariance implies that eq. (5) can be written as

$$G^{<}(r\alpha\,|\,r\alpha_1; t) = \pm\, G^{>}(r\alpha\,|\,r\alpha; t - i\beta)\,,$$

and taking into account the relations of the Fourier transform

$$G^{<(>)}(r\alpha\,|\,r\alpha_1;\ t) = \int \frac{d\omega}{2\pi}\, e^{-i\omega t}\, G^{<(>)}(r\alpha\,|\,r\alpha_1;\,\omega)\,,$$

$$G^{<(>)}(r\alpha\,|\,r\alpha_1;\,\omega) = \int dt\, e^{i\omega i}\, G^{<(>)}(r\alpha\,|\,r\alpha_1;\,t)\,,$$

one can express this condition as

$$G^{<}(r\alpha\,|\,r\alpha_1;\,\omega) = \pm\, e^{-\beta\omega}\, G^{>}(r\alpha\,|\,r\alpha_1;\,\omega)\,.$$

This implies that both of the definitions (4) can be expressed by a spectra function in the following form

$$\begin{aligned} G^{>}(r\alpha\,|\,r\alpha_1;\,\omega) &= -\,i\,[1 \pm f(\omega)]\; A(r\alpha\,|\,r\alpha_1;\,\omega)\,, \\ G^{<}(r\alpha\,|\,r\alpha_1;\,\omega) &= \mp\, i f(\omega)\; A(r\alpha\,|\,r\alpha_1;\,\omega)\,, \end{aligned} \tag{7}$$

where $f(\omega) = (\exp \beta\omega \mp 1)^{-1}$ is the boson or fermion occupation function. From eq. (7) one can write

$$A(r\alpha\,|\,r\alpha_1;\,\omega) = i\,[G^{>}(r\alpha\,|\,r\alpha_1;\,\omega) - G^{<}(r\alpha\,|\,r\alpha_1;\,\omega)]$$

or taking into account (4) and the definition of the Fourier transform

$$A(r\alpha\,|\,r\alpha_1;\,\omega) = \int\limits_{-\infty}^{\infty} dt\, e^{i\omega t}\,\langle a(r\alpha t)\, a^{+}\,(r\alpha_1\,0) \mp a^{+}(r\alpha_1 0)\, a(r\alpha t)\rangle\,.$$

Hence we have the sum rule

$$\int\limits_{-\infty}^{\infty} \frac{d\omega}{2\pi}\, A(r\alpha\,|\,r\alpha_1;\,\omega) = \delta(\alpha\,|\,\alpha_1)\,. \tag{8}$$

As regards the interpretation of the spectral function the Källen—Lehmann representation of the first of definitions (4) yields

$$\begin{aligned} G^{>}(r\alpha\,|\,r\alpha_1;\,\omega) = -\,2\,\pi i\, Z_g^{-1} \sum_{ij} \langle \Phi_i\,|\,a(r\alpha 0)\,|\,\Phi_j\rangle\langle\Phi_j\,|\,a^{+}(r\alpha_1\,0)\,|\,\Phi_i\rangle \,\times \\ \times\, e^{-\beta E_i}\,\delta(\omega + E_i - E_j) = -\,i\,[1 \pm f(\omega)]\; A(r\alpha\,|\,r\alpha_1;\,\omega)\,, \end{aligned}$$

where $|\,\Phi_i\rangle$ is a complete system of state vectorsin the generalized grand canonical ensemble and $\mathcal{H}\,|\,\Phi_i\rangle = E_i\,|\,\Phi_i\rangle$. Thus, with this construction we have achieved the separation of statistical and dynamical effects. The function

$A(r\alpha \mid r\alpha; \omega)$ gives the energy spectrum of the particle group of the type r in the quantum state α. Of course, it is even a function of temperature (temperature-dependent quasi-particle spectrum).

Because of the periodicity condition (6) we can write for the interval $0 \leqslant it \leqslant \beta$

$$G(r\alpha \mid r\alpha_1; t) = \frac{1}{-i\beta} \sum_\nu e^{-iz_\nu t} G(r\alpha \mid r\alpha_1; z_\nu)\,, \tag{9}$$

where

$$G(r\alpha \mid r\alpha_1; z_\nu) = \int\limits_0^{-i\beta} dt\, e^{iz_\nu t}\, G(r\alpha \mid r\alpha_1; t)\,.$$

$z_\nu = \pi\nu/-i\beta$, where ν is an even (odd) integer if the plus (minus) sign holds in (6). Now, proceeding as in [10], one can prove that

$$G(r\alpha \mid r\alpha_1; z_\nu) = \int\limits_{-\infty}^{\infty} \frac{d\omega}{2\pi}\, \frac{A(r\alpha \mid r\alpha_1; \omega)}{z_\nu - \omega}\,.$$

This relation defines the function $G(r\alpha \mid r\alpha_1; z)$, when inserting any complex z or z_ν. Using the relations

$$(z - \omega \pm i\varepsilon)^{-1} = P(z - \omega)^{-1} \mp i\pi\delta(z - \omega)\,,$$

we can prove that

$$A(r\alpha \mid r\alpha_1; \omega) = i\,[G(r\alpha \mid r\alpha_1; \omega + i\varepsilon) - G(r\alpha \mid r\alpha_1; \omega - i\varepsilon)]\,. \tag{10}$$

Thus, from the Green's function we can compute the energy spectrum of the particle groups in the same way as in conventional many-body theory of quasi-particles. Suppose for a moment that the group functions are eigenfunctions of the Hamiltonian of the particle groups (c.f. (A.10) and (A.11)), i.e.

$$\langle r\alpha \,|H_0|\, r\alpha_1\rangle = \langle r\alpha \,|H_0|\, r\alpha\rangle\, \delta(\alpha \mid \alpha_1)\,.$$

Of course, even under this condition the spectral function will not be a delta function, but because of the interaction of particle groups it will have a shape with finite width. This broadening of spectral lines is dependent on the thermodynamic properties of the system ("quasi molecule" spectrum).

As regards the analytic structure and the spectral representation of higher Green's functions the formulas are rather involved. However, certain special cases can be treated easily. The functions

$$\begin{aligned} D(p\alpha\alpha_1 t_1 \mid q\beta\beta_1 t_2) &= \\ &= -i\,\langle Ta^+(p\alpha r_1 t_1 + 0)\, a\,(p\alpha_1 r_1 t_1)\, a^+(q\beta r_2 t_2 + 0)\, a\,(q\beta_1 r_2 t_2)\rangle \end{aligned} \tag{11}$$

and

$$
\begin{aligned}
&P(p\alpha r_1\, v_1\, t_1 \,|\, q\beta r_2\, v_2\, t_2) = \\
&\qquad = -i\,\langle T a^{+}(p\alpha r_1\, t_1 + 0)\, a(p\alpha r_1 + v_1\, t_1)\, a^{+}(q\beta r_2 + v_2\, t_2 + 0)\, a(q\beta r_2\, t_2)\rangle
\end{aligned}
\tag{12}
$$

will be named as exciton and phonon propagators. In (11) and (12) the coordinate of the center of mass of the particle groups is indicated separately. The interpretation of (11) and (12) is obvious; they describe the propagation of exciton- and phonon-like excitations. The boundary conditions and analytic structure can be treated like those of $G(r\alpha \,|\, r\alpha_1; t)$, but details will not be given here. As a result we get the boson-like nature of these excitations. The connection between the propagators and the spectral functions is the same as eq. (10), but we find a more complicated sum-rule than (8).

This makes it possible to develop a true microscopic theory of excitons and phonons. We hope to set forth this in another place.

3. Green's function equations

Using the expression of the Hamiltonian (1), the (anti) commutation relations (A.3) and the equation of motion in the Heisenberg picture

$$
\begin{aligned}
i\frac{d}{dt}\, a(r\alpha) &= [a(r\alpha), \mathscr{H}]_{-} = \mathsf{S}_{\alpha_1} \langle r\alpha |H_0| r\alpha_1\rangle\, a(r\alpha_1) - \mu_r\, a(r\alpha) + \\
&\quad + \sum_q \mathsf{S}_{\alpha_1 \beta\beta_1} \langle r\alpha q\beta |V_{\mathrm{eff}}| r\alpha_1\, q\beta_1\rangle\, a^{+}(q\beta)\, a(r\alpha_1)\, a(q\beta_1)\,,
\end{aligned}
\tag{13}
$$

where

$$
\langle r\alpha q\beta |V_{\mathrm{eff}}| r\alpha_1\, q\beta_1\rangle = \langle r\alpha q\beta |V| r\alpha_1\, q\beta_1\rangle + 2\sum_l \gamma_{l;rq}\, \langle r\alpha q\beta |I_l| r\alpha_1\, q\beta_1\rangle\,.
$$

A similar equation can be deduced for the adjoint operator. With the aid of (13) we can derive the chain of Green's function equations in the usual way. The simplest one is

$$
\begin{aligned}
\left[i\frac{d}{dt} - e_{r\alpha}\right] G(r\alpha t \,|\, r\alpha_2\, t_2) &= \delta(\alpha t \,|\, \alpha_2 t_2) + \mathsf{S}_{\alpha_1 \neq \alpha} \langle r\alpha |H_0| r\alpha_1\rangle\, G(r\alpha_1 t \,|\, r\alpha_2 t_2) + \\
&\quad + i\sum_q (\pm)_{qq}\, \mathsf{S}_{\alpha_1\beta\beta_1} \langle r\alpha q\beta |V_{\mathrm{eff}}| r\alpha_1\, q\beta_1\rangle\, G(q\beta_1 t\, r\alpha_1 t \,|\, q\beta t + 0 \;\; r\alpha_2\, t_2)\,,
\end{aligned}
\tag{14}
$$

where

$$
e_{r\alpha} = \langle r\alpha |H_0| r\alpha\rangle - \mu_r\,.
$$

This chain of equations should be decoupled somehow in order to solve it. The simplest choice is a "Hartree—Fock" decomposition of the two-particle Green's function. This gives the equation

$$\left[i\frac{d}{dt}-e_{r\alpha}\right]G(r\alpha t\,|\,r\alpha_2 t_2)=\delta(\alpha t\,|\,\alpha_2 t_2)+\mathsf{S}_{\alpha_1\neq\alpha}\langle r\alpha\,|H_0|\,r\alpha_1\rangle\,G(r\alpha_1 t\,|\,r\alpha_2 t)+ \tag{15}$$
$$+\,i\sum_q(\pm)_{qq}(\pm)_{rq}\,\mathsf{S}_{\alpha_1\beta\beta_1}\langle r\alpha q\beta\,|V_{\mathrm{eff}}|\,r\alpha_1 q\beta_1\rangle\,G(q\beta_1 t\,|\,q\beta t+0)\,G(r\alpha_1 t\,|\,r\alpha_2 t_2)+$$
$$+\,i\,(\pm)_{rr}\,\mathsf{S}_{\alpha_1\beta\beta_1}\langle r\alpha r\beta\,|\,V_{\mathrm{eff}}|\,r\alpha_1 r\beta_1\rangle\,G(r\alpha_1 t\,|\,r\beta t+0)\,G(r\beta_1 t\,|\,r\alpha_2 t_2)\,,$$

where the exchange term is included because particle groups of the same type are indistinguishable.

As a formal tool we shall introduce a functional differentiation technique similar to that of [11] in order to gain further insight into the structure of Green's function equations. The definition of Green's functions is modified so that

$$G(r\alpha t\,|\,r\alpha_2 t_2;\,U)=-\,i\,\frac{\langle TSa(r\alpha t)\;a^{+}(r\alpha_2 t_2)\rangle}{\langle S\rangle}\,, \tag{16}$$

where

$$S=\exp\left[-\,i\int_0^{-i\beta}dt\sum_q\mathsf{S}_{\beta\beta_1}\;a^{+}(q\beta t)\;\langle q\beta\;|U(t)|\,q\beta_1\rangle\;a(q\beta_1 t)\right].$$

It is easy to verify that (16) has the same boundary conditions as the first of (3), and obeys the equation

$$\left[i\frac{d}{dt}-e_{r\alpha}\right]G(r\alpha t\,|\,r\alpha_2 t_2)-\mathsf{S}_{\alpha_1}\langle r\alpha\,|U(t)|\,r\alpha_1\rangle\,G(r\alpha_1 t\,|\,r\alpha_2 t_2)= \tag{17}$$
$$=\text{the same as the right-hand side of eq. (14),}$$

where we have suppressed U in the argument of the propagators. This convention will be followed in the following formulae. With the aid of the usual definition of the functional derivation (17) can be written as

$$\left[i\frac{d}{dt}-e_{r\alpha}\right]\,G(r\alpha t\,|\,r\alpha_2 t_2)-\mathsf{S}_{\alpha_1}\langle r\alpha\,|U(t)|\,r\alpha_1\rangle\,G(r\alpha_1 t\,|\,r\alpha_2 t_2)=\delta(\alpha t\,|\,\alpha_2 t_2)+$$
$$+\,\mathsf{S}_{\alpha_1\neq\alpha}\langle r\alpha\,|H_0|\,r\alpha_1\rangle\,G(r\alpha_1 t\,|\,r\alpha_2 t_2)+i\,\sum_q\mathsf{S}_{\alpha_1\beta\beta_1}(\pm)_{rq}\langle r\alpha q\beta\,|V_{\mathrm{eff}}|\,r\alpha_1 q\beta_1\rangle\times$$
$$\times\left[\frac{\delta}{\delta\,\langle q\beta|U(t)|\,q\beta_1\rangle}-i\varrho(q\beta t\,|\,q\beta_1 t)\right]G(r\alpha_1 t\,|\,r\alpha_2 t_2)\,, \tag{18}$$

where

$$\varrho(q\beta t\,|\,q\beta_1 t)=\pm\,iG(q\beta_1 t\,|\,q\beta t+0)\,.$$

The functional differentiation can be replaced by an integral operator, which is more familiar. Let us define the reciprocal Green's function by the equation

$$\int_0^{-i\beta}dt_1\,\mathsf{S}_{\alpha_1}G(r\alpha t\,|\,r\alpha_1 t_1)\,G^{-1}(r\alpha_1 t_1\,|\,r\alpha_2 t_2)=\delta(\alpha t\,|\,\alpha_2 t_2)\,, \tag{19}$$

as well as the quantities

$$\Gamma(r\alpha_1 t_1 \,|\, r\alpha_3 t_3;\, q_1 \beta_2 t_2 \,|\, q_1 \beta_3 t_2) = -\frac{\delta G^{-1}(r\alpha_1 t_1 \,|\, r\alpha_3 t_3)}{\delta\varrho(q_1 \beta_2 t_2 \,|\, q_1 \beta_3 t_2)}, \tag{20}$$

and

$$\Phi(q_1 \beta_2 t_2 \,|\, q_1 \beta_3 t_2;\, q\beta_4 t \,|\, q\beta_1 t) = \frac{\delta\varrho(q_1 \beta_2 t_2 \,|\, q_1 \beta_3 t_2)}{\delta\langle q\beta_4 | U(t) | q\beta_1\rangle}. \tag{21}$$

The quantities (20) and (21) will be named as vertex and fluctuation operators. Using (19) and the rules of functional differentiation [11] eq. (18) can be written as

$$\left[i\frac{d}{dt} - e_{r\alpha}\right] G(r\alpha t \,|\, r\alpha_2 t_2) - \int\limits_0^{-i\beta} dt_3\, \mathsf{S}_{\alpha_3}\, M(r\alpha t \,|\, r\alpha_3 t_3)\, G(r\alpha_3 t_3 \,|\, r\alpha_2 t_2) = \delta(\alpha t \,|\, \alpha_2 t_2), \tag{22}$$

where

$$\begin{aligned} M(r\alpha t \,|\, r\alpha_3 t_3) = {} & \delta(t \,|\, t_3)\,[1 - \delta(\alpha \,|\, \alpha_3)]\, \langle r\alpha | H_0 | r\alpha_3\rangle + \\ & + i \sum_q (\pm)_{rq}\, \mathsf{S}_{\alpha_4\beta_1\beta_4} \langle r\alpha q\beta_4 | V_{\text{eff}} | r\alpha_4 q\beta_1\rangle \left[\sum_{q_1} \mathsf{S}_{\alpha_1\beta_2\beta_3} \int\limits_0^{-i\beta} dt_1\, dt_2\, G(r\alpha_4 t \,|\, r\alpha_1 t_1) \times \right. \\ & + \Gamma(r\alpha_1 t_1 \,|\, r\alpha_3 t_3;\, q_1\beta_2 t_2 \,|\, q_1 \beta_3 t_2)\, \Phi(q_1 \beta_2 t_2 \,|\, q_1 \beta_3 t_2;\, q\beta_4 t \,|\, q\beta_1 t) - \\ & \left. \times\, i\varrho(q\beta_4 t \,|\, q\beta_1 t)\, \delta(\alpha_4 t \,|\, \alpha_3 t_3) \right] \end{aligned} \tag{23}$$

is the mass operator, and the limit $U \to 0$ has been performed after the functional differentiation. The symbol $[1 - \delta(\alpha \,|\, \alpha_3)]$ in the first term on the right-hand side indicates that in a summation the terms $\alpha = \alpha_3$ should be left out. (c.f. (14).) Note that this procedure does not involve any approximation. Eq. (22) is exact, and it expresses a topological property of the Green's function equation (14) (Fig. 1). The DYSON equations cannot be written easily because

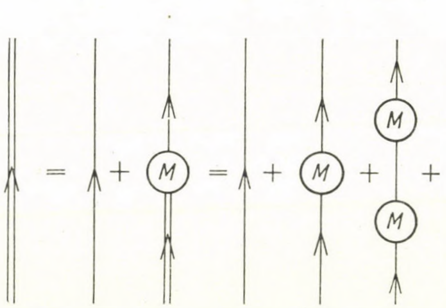

Fig. 1. The topological structure of a particle group propagator

of the nonperturbative definition of operators (20) and (21). However, a direct evaluation of (21) yields ($U = 0$)

$$\begin{aligned} i\Phi(q_1 \beta_2 t_2 | q_1 \beta_3 t_2; q\beta_4 t\, q\, \beta_1 t) = \langle T a^+(q\beta_4 t + 0)\, a(q\beta_1 t)\, a^+(q_1 \beta_2 t_2 + 0)\, a(q_1 \beta_3 t_2)\rangle \\ - \langle a^+(q\beta_4 t)\, a(q\beta_1 t)\rangle \langle a^+(q_1 \beta_2 t_2)\, a(q_1 \beta_3 t_2)\rangle , \end{aligned} \tag{24}$$

whereas taking into account (18) a perturbative expansion gives

$$\begin{aligned} \Gamma(r\alpha_1 t_1 \,|\, r\alpha_3 t_3; q_1 \beta_2 t_2 \,|\, q_1 \beta_3 t_2) \cong (\pm)_{rq_1} \langle r\alpha_1 q_1 \beta_2 |V_{\mathrm{eff}}| r\alpha_3 q_1 \beta_3\rangle \times \\ \times\, \delta(t_1 \,|\, t_3)\, \delta(t_3 \,|\, t_2) + \ldots . \end{aligned} \tag{25}$$

The other contributions to (25) are higher order in the matrix elements of the interaction. Thus (20) and (21) play indeed the role of vertex and density fluctuation operators.

4. Perturbation theory

The formal theory of functional differentiation can be used very advantageously in the derivation of perturbation theory. Define the unperturbed propagator as the solution of the following equation

$$\left[i \frac{d}{dt} - e_{r\alpha}\right] G_0(r\alpha t \,|\, r\alpha_1 t_1) = \delta(\alpha t \,|\, \alpha_1 t_1) . \tag{26}$$

The unperturbed propagator can be a functional of a "test potential" also. Then the left hand side of (26) is the same as that of eq. (17). We can convert the time-differential equations into an integral equation with the aid of the unperturbed propagator, e.g.

$$\begin{aligned} G(r\alpha t \,|\, r\alpha_3 t_3) = G_0(r\alpha t \,|\, r\alpha_3 t_3) + \int_0^{-i\beta} dt_1\, \mathrm{S}_{\alpha_1 \alpha_2 \neq \alpha}\, G_0(r\alpha t \,|\, r\alpha_1 t_1) \times \\ \times \langle r\alpha_1 |H_0| r\alpha_2\rangle\, G(r\alpha_2 t_1 \,|\, r\alpha_3 t_3) + i \int_0^{-i\beta} dt_1 \sum_q \mathrm{S}_{\beta_1 \beta_2 \alpha_1 \alpha_2} (\pm)_{rq} \times \\ \times G_0(r\alpha t \,|\, r\alpha_1 t_1)\, \langle r\alpha_1 q\beta_1 |V_{\mathrm{eff}}| r\alpha_2 q\beta_2\rangle \times \\ \times \left[\frac{\delta}{\delta \langle q\beta_1 |U(t_1)| q\beta_2\rangle} - i\varrho(q\beta_1 t_1 \,|\, q\beta_2 t_1)\right] G(r\alpha_2 t_1 \,|\, r\alpha_3 t_3) , \end{aligned} \tag{27}$$

where after the functional differentiation we effected the limit $U \to 0$. If we are trying to solve the equation (27) by an iterational method the Neumann series generates the perturbational expansion of the propagator in the same way as in [10]. Associating with this procedure a graph-theoretical technique for enumerating various terms the following recipes can be given for the calculation of the particle group propagator.

1. Draw all topologically distinct connected graphs consisting of the elements shown in Fig. 2. Each graph should contain an ingoing and an outgoing particle line.

2. The graphs should be indexed in the manner shown in Fig. 2. The "two particle" and "four particle" elementary vertices are equal time.

3. Associate

a) an unperturbed propagator with each particle line;

b) $\langle r\alpha \mid H_0 \mid r\alpha_1 \rangle$ with each "two particle" vertex;

c) $i(\pm)_{rq} \langle r\alpha q\beta_1 \mid V_{\text{eff}} \mid r\alpha_1 q\beta_2 \rangle$ with each "four particle" vertex;

d) $(\pm)_{rr}$ with each closed particle line.

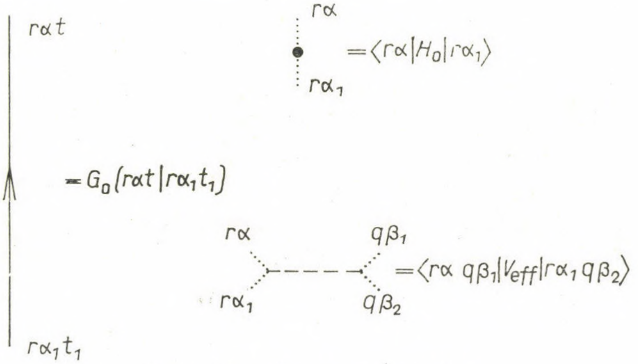

Fig. 2. The elements of graphs

4. Sum (integrate) over all internal variables. The integrations over the time variables should be performed over the interval $[0, -i\beta]$. Do not forget that sums do not contain terms with the diagonal matrix elements $\langle r\alpha \mid H_0 \mid r\alpha \rangle$ (c.f. the first summation in eq. (14).)*

If the system is invariant under translations in space and time it is advantageous to treat the center of mass coordinate separately. Because of energy momentum conservation

$$G(r\alpha k t \mid r\alpha_1 k_1 t_1) = G(r\alpha \mid r\alpha_1; k\ t - t_1)\, \delta(k - k_1)\,.$$

Taking into account the definition of the unperturbed propagator and eq. (9)

$$G_0(r\alpha \mid r\alpha_1; kt) = \frac{1}{-i\beta} \sum_{\nu} \frac{e^{-iz_\nu t}\, \delta(\alpha \mid \alpha_1)}{z_\nu - \left(\dfrac{k^2}{2M_r} + \varepsilon_{r\alpha} - \mu_r\right)}\,, \tag{28}$$

---

* When the group functions are eigenfunctions of the Hamiltonian of particle groups only diagonal matrix elements exist. Hence, in this case there are no graphs with "two particle" vertices.

where M_r is the mass of the r-th group of particles. $\varepsilon_{r\alpha} = \langle r\alpha \mid H_0' \mid r\alpha \rangle$, where H_0' is the Hamiltonian of the r-th particle group in the center of mass frame and for z_ν see the comment after (9). Thus, with (28) we can formulate more explicit recipes for computing the graphs. Rules 2.—3. should be replaced by the following ones.

2.' Only each particle line should be indexed by a set of indices $\{r\alpha k z_\nu\}$.

3.' Associate

a) the propagator

$$G_0(r\alpha k z_\nu) = \frac{1}{z_\nu - \left(\frac{k^2}{2M_r} + \varepsilon_{r\alpha} - \mu_r\right)}$$

with every particle line;

b) $\langle r\alpha \mid H_0 \mid r\alpha_1 \rangle \, (2\pi)^3 \, \delta(k - k_1)\delta_{\nu,\nu_1}$ with each "two particle" vertex;

c) $i(\pm)_{rg} \, \langle r\alpha q\beta_1 \mid V_{\text{eff}} \mid r\alpha_1 \, q\beta_2 \rangle \; (2\pi)^3 \; \delta(\nu + \nu_1 - \nu_2 - \nu_3)\delta_{\nu+\nu_1,\nu_2+\nu_3}$ with each "four particle" vertex;

d) $(\pm)_{rr}$ with each closed particle line.

In addition we should multiply this expression by a factor $[i\beta]^{-p+s}$, where p is the number of propagators and s the number of particle lines for which the summation was carried out. With this procedure we get the Green's function in energy momentum representation from which we can easily proceed to the calculation of the spectral function.

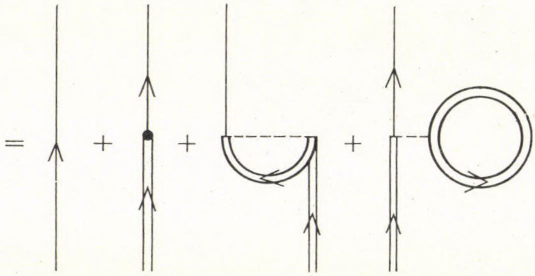

Fig. 3. The Hartree—Fock graphs

Since we hope the bulk of particle correlation is taken into account by the choice of group functions, we can expect that perturbation theory of finite order can give satisfactory results in many cases.* However, it is interesting to see what possibilities there are for a summation of graphs to infinite order. As is always the case this can be carried out by solving an integral

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* This is similar to the Weinberg quasiparticle method, in some respects [12].

equation instead of the simple iterational scheme of the perturbation theory. E.g. the "Hartree—Fock" approximation (15) includes the summation of all Hartree—Fock graphs as shown in Fig. 3. Further possibilities are given when after having determined the mass operator as a sum of proper self-energy graphs we calculate the Green's function self-consistently from (22).

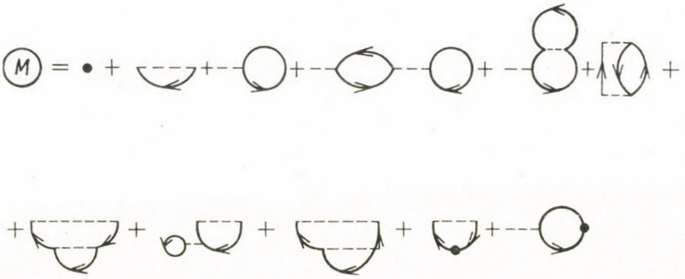

Fig. 4. The mass operator to second order in interaction matrix elements

Because of the "two particle" vertices some unusual graphs arise. In Fig. 4 the mass operator is given to the second order in the interaction matrix elements.

As regards the two-particle Green's function, in practice one is always compelled to sum a selected class or graphs (chain and ladder diagrams). We hope to return to this problem elsewhere discussing applications of this theory.

5. Discussion

In statistical physics it is always easier to understand things if we can separate "statistical" and "dynamical" effects. It is known that non-interacting elementary particles are either bosons of fermions. When treating interacting systems we start from the boson- or fermion-like behaviour of particles. In many cases this is satisfactory, however, sometimes switching on the interaction leads to radical changes in the statistical properties of the system. It is known that attractive interaction can cause the instability of the Fermi surface, etc. In this case, we cannot expect that starting from the dynamical and statistical properties of the non-interacting system good results can be achieved by perturbative methods. However, by choosing particle group basis functions we can take the interaction into account to a certain extent from the beginning. These "composite particles" cannot be regarded rigorously either as fermions or bosons. However, there exists a possibility [3] by which we can treat this problem consistently. The aim of this paper is to present a practical way of treating problems in this theory.

Comparing this theory with the usual methods of many-body physics we see its advantages and disadvantages. In conventional many-body theory we must sum infinite plane waves in order to get the "wave functions" of particle groups (or the t matrix, higher order Green's functions, etc.), whereas this theory takes into account the particle correlations characteristic of the system from the very beginning. A disadvantage is that when working with a plane wave basis all the matrix elements of the kinetic energy, interaction potential, etc. are usually available, whereas in this method we must compute them one after the other. Therefore in a perturbative procedure we can usually use finitely many intermediate states.

As regards the practical applications, this theory could be used to advantage in those fields where a true microscopic approach would be necessary for an adequate treatment of the problem (phonons in quantum solids, exciton modes). Another possibility is to investigate superconductive systems along somewhat similar lines as in [5.]

Acknowledgement

The author is grateful to Prof. P. Gombás for encouragement during this work.

Appendix A

In this Appendix we shall re-derive some results of [3] in a more general form. As regards more detailed treatment the reader should consult [3].

We intend to develop a many-body theory in terms of the sets $\{\varphi_{r\alpha}(\xi^r)\}$, $i = 1, \ldots, m$ of group functions. r denotes the species of the group functions, while α is a complete set of quantum numbers characterizing the group functions within a certain type. The structure of the argument of the r-th group function is

$$\xi^r = \left\{x_{11}, x_{12}, \ldots, x_{1j_1^r}, x_{21}, \ldots, x_{Lj_L^r}\right\}, \tag{A.1}$$

where x is an abbreviated notation for spatial and spin variables. The first index refers to the type of the elementary particle (altogether L species in the system). Thus j_1^r are elementary particles of the first type, j_2^r elementary particles of the second type, etc. and belong to the r-th species of groups. We suppose that these group functions form an orthonormal and complete set with the right (anti) symmetry conditions, i.e.

$$\begin{aligned} \mathsf{S}_{\xi^r} \varphi_{r\alpha}^*(\xi^r)\, \varphi_{r\beta}(\xi^r) &= \delta(\alpha \mid \beta), \\ \mathsf{S}_{\alpha} \varphi_{r\alpha}(\xi^r)\, \varphi_{r\alpha}^*(\xi^{r\prime}) &= \Lambda_r\, \delta(\xi^r \mid \xi^{r\prime}), \end{aligned} \tag{A.2}$$

where summation means integration over the continuous part of the summation indices and $\delta(|)$ means Dirac delta over the continuous and Kronecker delta over the discrete parts of its argument. These notational conventions will be followed throughout this work. In eq. (A.2) Λ_r means the (anti) symmetrizer operator over suitable variables.

These particle-group functions can be used to construct annihilation and creation operators for groups of particles in the usual way. However, because of the mathematical difficulties mentioned in Section 1 it is more advantageous to formulate the problem in the "ideal" Hilbert space H_i. The annihilation and creation operators of particle groups in H_i obey the usual (anti) commutation relations

$$\begin{aligned} [a(q\alpha), a(r\beta)]_{\mp} &= [a^+(q\alpha), a^+(r\beta)]_{\mp} = 0\,, \\ [a(q\alpha), a^+(r\beta)]_{\mp} &= \delta(q\alpha\,|\,r\beta)\,, \end{aligned} \tag{A.3}$$

where $a(q\alpha)$ $(a^+(q\alpha))$ means annihilation (creation) operator for a group of particles characterized by the "wave function" $\varphi_{q\alpha}(\xi^q)$. In eq. (A.3) commutation or anticommutation is understood depending on the "common fermion content" of the particle groups. Generalizing the arguments of [3] it can be shown that the section of H_i in which the state vectors $|\,\Phi\rangle$ satisfy the subsidiary conditions

$$I_{l;rq}|\Phi\rangle = \pm\, n_r\, n_q |\Phi\rangle, \tag{A.4}$$

is isomorphic with the Hilbert space of physical states, where

$$I_{l;rq}\, \mathsf{S}_{\alpha\alpha_1\beta\beta_1}\, a^+(r\alpha)\, a^+(q\beta)\, \langle r\alpha q\beta\, |I_l|\, r\alpha_1\, q\beta_1\rangle\, a(r\alpha_1)\, a(q\beta_1)\,, \tag{A.5}$$

$l = 1, 2, \ldots, L$, and the upper (lower) sign holds, if the l-th elementary particle is a Bose—Einstein (Fermi—Dirac) particle. Furthermore

$$N_r|\Phi\rangle = n_r|\Phi\rangle, \tag{A.6}$$

where

$$N_r = \mathsf{S}_\alpha\, a^+(r\alpha)\, a(r\alpha) \tag{A.7}$$

is the number operator of the r-th particle groups. Now the Hamiltonian in H_i can be written as

$$H = H_0 + V, \tag{A.8}$$

where

$$\begin{aligned} H_0 &= \sum_r \mathsf{S}_{\alpha\alpha_1}\, a^+(r\alpha)\, \langle r\alpha\, |H_0|\, r\alpha_1\rangle\, a(r\alpha_1)\,, \\ V &= \frac{1}{2} \sum_{rq} \mathsf{S}_{\alpha\alpha_1\beta\beta_1}\, a^+(r\alpha)\, a^+(q\beta)\, \langle r\alpha q\beta\, |V|\, r\alpha_1\, q\beta_1\rangle\, a(r\alpha_1)\, a(q\beta_1)\,. \end{aligned} \tag{A.9}$$

If the Hamiltonian of the system is given by the expression

$$H = \sum_{l}^{L} \Bigg[\int dx_l \, \psi_l^{+}(x_l) \, T_l(x_l) \, \psi_l(x_l) + \\ + \frac{1}{2} \int dx_l \, dx_l' \, \psi_l^{+}(x_l) \, \psi_l^{+}(x_l') \, V_{ll}(x_l, x_l') \, \psi_l(x') \, \psi_l(x_l) \Bigg] + \\ + \sum_{k<l}^{L} \int dx_k \, dx_l \, \psi_k^{+}(x_k) \, \psi_l^{+}(x_l) \, V_{kl}(x_k, x_l) \, \psi_l(x_l) \, \psi_k(x_k) \, ,$$

the matrix elements occurring in (A.8) are given by the following expressions

$$\langle r\alpha | H_0 | r\alpha_1 \rangle = \langle r\alpha | T | r\alpha_1 \rangle + \langle r\alpha | V | r\alpha_1 \rangle \, , \tag{A.10}$$

where

$$\langle r\alpha | T | r\alpha_1 \rangle = \mathsf{S}_{\xi^r} \, \varphi_{r\alpha}^{*}(\xi^r) \Bigg[\sum_{l}^{L} j_l^r \, T_l(x_{l1}) \Bigg] \varphi_{r\alpha_1}(\xi^r) \, ,$$

$$\langle r\alpha | V | r\alpha_1 \rangle = \mathsf{S}_{\xi^r} \, \varphi_{r\alpha}^{*}(\xi^r) \Bigg[\frac{1}{2} \sum_{k \neq l}^{L} j_k^r j_r^r \, V_{kl}(x_{k1}, x_{l1}) + \\ + \frac{1}{2} \sum_{l}^{L} j_l^r (j_l^r - 1) \, V_{ll} x_{l1}, x_{l2}) \Bigg] \varphi_{r\alpha_1}(\xi^r) \, . \tag{A.11}$$

Furthermore,

$$\langle r\alpha q\beta | V | r\alpha_1 q\beta_1 \rangle = \mathsf{S}_{\xi^r \xi^q} \, \varphi_{r\alpha}^{*}(\xi^r) \varphi_{q\beta}^{*}(\xi^q) \Bigg[\sum_{kl} j_k^r j_l^q \, V_{kl}(x_{k1}, x_{l1}') \Bigg] \varphi_{r\alpha_1}(\xi^r) \, \varphi_{q\beta_1}(\xi^q) \, , \tag{A.12}$$

and in the last equation $x_{k1} \in \xi^r$, $x_{l1}' \in \xi^q$. The matrix elements of operator $I_{l;rq}$ are given by the equation

$$\langle r\alpha q\beta | I_l | r\alpha_1 q \beta_1 \rangle = \mathsf{S}_{\xi^r \xi^q} \, \varphi_{r\alpha}^{*}(\xi^r) \, \varphi_{q\beta}^{*}(\xi^q) \, \varphi_{r\alpha_1}(\tilde{\xi}^{\,r}) \, \varphi_{q\beta_1}(\tilde{\xi}^{\,q}) \, , \tag{A.13}$$

where the tilde over the arguments means that $x_{l1} \in \xi^r$ and $x_{l1}' \in \xi^q$ should be interchanged. Following [3] the free energy operator of a generalized grand canonical ensemble can be defined as

$$\mathcal{K} = H - \sum_{q=1}^{m} \mu_q N_q + \sum_{l}^{L} \sum_{r \leqslant q} \gamma_{l;rq} \, I_{l;rq} \, , \tag{A.14}$$

where μ_q and $\gamma_{l,rq}$ are the usual Lagrange multipliers associated with the density of the particle groups and conditions (A.4). In order to save space the alternative form of (A.14)

$$\mathcal{K} = H_0 + V_{\text{eff}}$$

will be used, where V_{eff} means the terms of $\mathscr{K}$ containing all fourfold products of annihilation and creation operators. Now one can form the density operator of the generalized grand canonical ensemble as

$$\varrho = Z_g^{-1}\, e^{-\beta \mathscr{K}}, \tag{A.15}$$

where

$$Z_g = tr\, e^{-\beta \mathscr{K}} \tag{A.16}$$

is the grand partition function and $\beta = (kT)^{-1}$, k is Boltzmann's constant. The average of any operator O should be calculated by the expression

$$\langle O \rangle = tr\, \varrho O\,, \tag{A.17}$$

and the thermodynamic potential W of the ensemble can be defined as

$$W = -\, \beta^{-1} \ln Z_g. \tag{A.18}$$

From eqs. (A.4), (A.6) and (A.15)—(A.18) the Lagrange multipliers in (A.14) should be fixed from the conditions

$$\begin{aligned} \frac{\partial W}{\partial \mu_q} &= -\, \mu_q \\ \frac{\partial W}{\partial \gamma_{l;rq}} &= \pm\, n_r\, n_q\,, \end{aligned} \tag{A.19}$$

which point out the suitable subsection of H_i. Any operator O can be expressed in H_i with the aid of the "ideal" annihilation and creation operators. The most general operators occurring in practice are of the form

$$O = O_1 + O_2\,, \tag{A.20}$$

where

$$O_1 = \sum_l^L \int dx_l\, \psi_l^+(x_l)\, O_l(x_l)\, \psi_l(x_l)\,,$$

$$O_2 = \frac{1}{2} \sum_l^L \int dx_l\, dx_l'\, \psi_l^+(x_l)\, \psi_l^+(x_l')\, O_{ll}(x_l, x_l')\, \psi_l(x_l')\, \psi_l(x_l) +$$

$$+ \sum_{l<k}^L \int dx_l\, dx_k\, \psi_l^+(x_l)\, \psi_k^+(x_k)\, O_{lk}(x_l, x_k)\, \psi_k(x_k)\, \psi_l(x_l)\,.$$

In H_i these are expressed as

$$O_1 = \sum_r \mathsf{S}_{\alpha\alpha_1} a^+(r\alpha) \langle r\alpha | O_1 | r\alpha_1 \rangle a(r\alpha_1)$$

$$O_2 = \sum_r \mathsf{S}_{\alpha\alpha_1} a^+(r\alpha) \langle r\alpha | O_2 | r\alpha_1 \rangle a(r\alpha_1) +$$

$$+ \frac{1}{2} \sum_{rq} \mathsf{S}_{\alpha\alpha_1\beta\beta_1} a^+(r\alpha)\, a^+(q\beta) \langle r\alpha q\beta | O_2 | r\alpha_1\, q\beta_1 \rangle a(r\alpha_1) a(q\beta_1),$$

where

$$\langle r\alpha | O_1 | r\alpha_1 \rangle = \mathsf{S}_{\xi^r} \varphi^*_{r\alpha}(\xi^r) \left[\sum_l^L j_l^r \; O_l(x_{l1}) \right] \varphi_{r\alpha_1}(\xi^r)$$

$$\langle r\alpha | O_2 | r\alpha_1 \rangle = \mathsf{S}_{\xi^r} \varphi^*_{r\alpha}(\xi^r) \left[\frac{1}{2} \sum_{l \neq k}^L j_l^r j_k^r O_{lk}(x_{l1}, x_{k1}) + \right.$$

$$\left. + \frac{1}{2} \sum_l^L j_l^r (j_l^r - 1)\, O_{ll}(x_{l1}, x_{l2}) \right] \varphi_{r\alpha_1}(\xi^r)$$

$$\langle r\alpha q\beta | O_2 | r\alpha_1\, q\beta_1 \rangle = \mathsf{S}_{\xi^r \xi^q} \varphi^*_{r\alpha}(\xi^r)\, \varphi^*_{q\beta}(\xi^q) \left[\sum_{lk}^L j_l^q j_k^r O_{lk}(x_{l1}, x'_{k1}) \right] \times$$

$$\times \varphi_{r\alpha_1}(\xi^r)\, \varphi_{q\beta_1}(\xi^q) \,,$$

and in the last section $x_{k1} \in \xi^q$, $x'_{k1} \in \xi^r$.

Appendix B

In this Appendix we determine the connection between the spectral function and the generalized thermodynamic potential. The thermodynamic potential plays a central role in this theory; we select the suitable section of H_i with the aid of the thermodynamic potential, and it is also closely connected with the macroscopic parameters of the system. Introduce a formal coupling constant in the non-diagonal part of the free energy operator

$$\mathcal{K}(\lambda) = \mathcal{H}_d + \lambda \mathcal{V}_n \,,$$

where

$$\mathcal{H}_d = \sum_r \mathsf{S}_\alpha a^+(r\alpha) \left[\langle r\alpha | H_0 | r\alpha \rangle - \mu_r \right] a(r\alpha) \,,$$

and

$$\mathcal{V}_n = \mathcal{V}_n' + \mathcal{V}_n'' = \sum_r \mathsf{S}_{\alpha \neq \alpha_1} a^+(r\alpha) \langle r\alpha | H_0 | r\alpha_1 \rangle a(r\alpha_1) +$$

$$+ \frac{1}{2} \sum_{rq} \mathsf{S}_{\alpha\alpha_1\beta\beta_1} a^+(r\alpha)\, a^+(q\beta) \langle r\alpha q\beta | V_{\mathrm{eff}} | r\alpha_1\, q\beta_1 \rangle a(r\alpha_1)\, a(q\beta_1) \,.$$

Of course $\mathcal{K}(1) = \mathcal{K}$, and $W(1) = W$. From the definition of the thermodynamic potential it follows that

$$\frac{\partial W(\lambda)}{\partial \lambda} = \langle \mathscr{V}_n \rangle_\lambda , \tag{B.1}$$

where the index λ means that the average should be taken with the aid of the density operator

$$\varrho(\lambda) = [e^{-\beta \mathscr{K}(\lambda)}]^{-1} \, e^{-\beta \mathscr{K}(\lambda)} .$$

With the aid of the relations

$$\sum_r \mathsf{S}_\alpha \, a^+(r\alpha) \, [\mathscr{V}_n', a(r\alpha)]_- = - \mathscr{V}_n' ,$$

$$\sum_r \mathsf{S}_\alpha \, a^+(r\alpha) \, [\mathscr{V}_n'', a(r\alpha)]_- = - 2 \mathscr{V}_n'' ,$$

we can write

$$\langle \mathscr{V}_n \rangle_\lambda = \frac{1}{2} \sum_r \{ \mathsf{S}_{\alpha \neq \alpha_1} \langle r\alpha | H_0 | r\alpha_1 \rangle \langle a^+(r\alpha) \, a(r\alpha_1) \rangle_\lambda -$$
$$- \mathsf{S}_{\alpha\alpha_1} \, \delta(\alpha | \alpha_1) \langle a^+(r\alpha) \, [\mathscr{V}_n, a(r\alpha)]_- \rangle_\lambda \} .$$

From this equation

$$\langle \lambda \mathscr{V}_n \rangle_\lambda = \frac{1}{2} \sum_r \mathsf{S}_{\alpha\alpha 1} \{ [\lambda \langle r\alpha | H_0 | r\alpha_1 \rangle - \delta(\alpha | \alpha_1) \left((1 + \lambda) \langle r\alpha | H_0 | r\alpha \rangle - \mu_r \right)] \times$$
$$\times \langle a^+(r\alpha) \, a(r\alpha_1) \rangle_\lambda - \delta(\alpha | \alpha_1) \langle a^+(r\alpha) \left[\mathscr{K}(\lambda), a(r\alpha) \right]_- \rangle_\lambda \} .$$

Using the definition of Green's functions and the equation of motion of operators

$$\langle \lambda \mathscr{V}_n \rangle_\lambda = \frac{1}{2} i \sum_r (\pm)_{rr} \, \mathsf{S}_{\alpha\alpha_1} \left\{ \lambda \langle r\alpha | H_0 | r\alpha_1 \rangle + \delta(\alpha | \alpha_1) \times \right.$$
$$\left. \times \left[i \frac{d}{dt} - (1 + \lambda) \langle r\alpha | H_0 | r\alpha \rangle + \mu_r \right] \right\} G(r\alpha_1 | r\alpha; t = - 0, \lambda) ,$$

where we have indicated explicitly that the propagator is a function of λ. Applying the Fourier transform and taking into account (4) and (7)

$$\langle \lambda \mathscr{V}_n \rangle_\lambda = \frac{1}{2} \sum_r \mathsf{S}_{\alpha\alpha_1} \oint_C \frac{d\omega}{2\pi} \{ \lambda \langle r\alpha | H_0 | r\alpha_1 \rangle + \delta(\alpha | \alpha_1) \times$$
$$\times [\omega - (1 + \lambda) \langle r\alpha | H_0 | r\alpha \rangle + \mu_r] \} f_r(\omega) \, A(r\alpha_1 | r\alpha; \omega, \lambda) ,$$

where C is the real axis closed with a semicircle on the upper half of the ω plane. With the aid of (B.1)

$$W = W(1) = W(0) + \int_0^1 \frac{d\lambda}{\lambda} \langle \lambda \mathscr{V}_n \rangle_\lambda ,$$

where $W(0)$ is the thermodynamic potential belonging to the diagonal part of $\mathscr{K}$, which can be calculated with the usual methods of elementary statistical mechanics.

REFERENCES

1. R. McWeeny, Proc. Roy. Soc. London, **A253,** 242, 1959.
2. F. J. Dyson, Phys. Rev., **102,** 1217, 1956.
3. M. Girardeau, J. Math. Phys., **4,** 1096, 1963.
4. A. C. Hurley, J. E. Lennard-Jones and J. A. Pople, Proc. Roy. Soc. London, **A220,** 446, 1953.
5. J. M. Blatt, "Theory of Superconductivity", Academic Press, New York and London; 1964, Appendix B.
6. H. L. Sahlin and J. L. Schwartz, Phys. Rev., **138B,** 267, 1965.
7. R. Delbourgo, A. Salam and J. Strathee, Phys. Letters, **22,** 680, 1966.
8. D. R. Fredkin and N. R. Werthamer, Phys. Rev., **138A,** 1527, 1965.
9. P. Gombás, Z. Phys., **94,** 473, 1935; H. Hellmann, J. Chem. Phys., **3,** 61, 1935. Acta Physico-chimica URSS, **1,** 913, 1935.
10. P. C. Martin and J. Schwinger, Phys. Rev., **115,** 1342, 1959.
    L. P. Kadanoff and G. Baym, "Quantum Statistical Mechanics" Benjamin, New York, 1962.
11. V. L. Bonch-Bruevich and S. V. Tyablikov, "The Green function method in statistical mechanics" North-Holland, Amsterdam, 1962.
12. S. Weinberg, Phys. Rev., **130,** 776, 1963; **131,** 440, 1963.

ПРОБЛЕМЫ МНОГИХ ТЕЛ ПО ГРУППОВЫМ ФУНКЦИЯМ ЧАСТИЦ

ДЬ. БЮТИ

Резюме

В рассмотрении систем многих частиц в некоторой степени принимается во внимание корреляция частиц выбором групповых базисных функций частиц. Однако, если эти базисные функции не удовлетяоряют некоторым строгим условиям («строгая ортогональность»), возникает несколько математических трудностей. Используя раньше разработанный метод для преодоления данных трудностей, нами даются практические методы для решения этой проблемы. Применяется формализм гриновских функций Мартина и Швингера, используя полевые операторы для групп частиц с внутренними степенями свободы. Наряду с возможным вычислительным преимуществом и действительной важностью проблемы, данная теория дает некоторые возможности для ab initio микроскопической теории конденсированных сред. Наконец, данная теория может быть полезна при уяснении некоторых вопросов релятивистской теории поля структурных частиц.

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DECAY SCHEME OF Pr^{144}

By

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The decay of the 17.5 min Pr^{144} has been investigated with a scintillation spectrometer. The energy of the gamma transitions in the daughter nucleus Nd^{144} are found to be 2.18, 1.49 and 0.70 MeV in agreement with the measurements of other investigators. The ambiguities concerning the previously reported 2.8, 1.67 and 1.16 MeV gamma transitions are removed. A decay scheme is then proposed.

1. Introduction

The beta transitions following the decay of Pr^{144} have been carefully studied by EMMERICH et al. [1] using a magnetic lens spectrometer. They identified three beta groups with energy limits of 2.98 ± 0.02 (95.5%), 2.28 ± 0.2 and 0.80 ± 0.1 MeV, thus confirming the results of ALBURGER et al. [2], JOHN et al. [3] and PORTER et al. [4].

For the gamma transitions, previously published results are more controversial. KELLER et al. [5] and PORTER et al. [4] observed a 60.3 keV internally converted gamma rays which they assigned to Nd^{144}. EMMERICH et al. [1] confirmed its existence but were unable to find a 60 keV free gamma ray. However, CORK et al. [6] settled the question by proving that the conversion line is actually due to the 59 keV γ-ray line in Pr^{144}. By photoelectric and conversion electron measurements ALBURGER et al. [2] observed gamma rays of energies 0.695 ± 0.005, 1.48 ± 0.01 and 2.185 ± 0.015 MeV having relative intensities 1 : 0.4 : 1.1, respectively. These results were confirmed by PORTER et al. [4] who used a similar technique and by CORK et al. [6] and KREGER et al. [7] who employed a scintillation spectrometer. Moreover, CORK et al. [6] were able to observe in the single spectrum of the 2.185 MeV two gamma peaks of energies 1.7 and 1.1 MeV which were interpreted as single and double annihilation escaped quanta. Applying the same technique FIRSOV [8] found gamma rays of energies 2.8, 2.185, 1.7, 1.49, 1.1 and 0.7 MeV with relative intensities 2.5, 100, 2, 30, 2, weak and 150, respectively. He suggested a decay scheme with two new levels at energies of 1.7 and 2.8 MeV. So it was necessary to check the existence of these levels by studying in detail the single gamma spectrum.

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2. Experimental technique

The source applied in the present investigation is made of pure Ce^{144} of 282 days half life in equilibrium with its 17.5 min Pr^{144} daughter activity. It was prepared by extraction from fission fragments of uranium and supplied by the Amersham Radio-chemical Laboratory.

Measurements have been performed with a scintillation spectrometer using 5.1 cm $\times$ 5.1 cm NaI (Tl) crystal coupled to a (10—17) RCA photomultiplier and a transistorized RIDL 200 channel analyser. The resolution of the spectrometer was found to be about 7.5% for the 661 keV gamma quanta of Cs^{137}. The energy calibration has been carried out in various regions of the spectrum by means of standard sources of almost the same strength as Pr^{144}.

The single gamma spectrum was measured, with the source in contact with the crystal and at 10 cms above it (to minimize the intensity of the summing peaks), several times during a period of one month. The relative intensities of the peaks were the same, indicating that the Ce^{144} source did not contain any impurities. After each set of measurements the natural background was subtracted under exactly the same experimental conditions.

3. Results and discussion

A representative spectrum with the Pr^{144} source in contact with the crystal is shown in Fig. 1. The energies of the observed gamma groups together with the relative intensities after being corrected for crystal efficiencies are also shown in Table I. Since the highest observed transition energy is at 2.18 MeV, attempts have been made to find higher transition energies but none has been observed. It thus seems that the γ-transition of energy 2.8 MeV observed by Firsov [8] does not exist and may be attributed to the piling up of the 0.695 and 2.18 MeV radiation or may be that of the 1160 and 1670 KeV radiation. The former is much more probable. It should be pointed out that the gamma transition of energy 2.18 MeV is found to be partially due to the summing of the 0.7 and 1.48 MeV transitions since its relative intensity decreases when carrying out the same measurements with the source 10 cm above the crystal. The percentage of such a decrease in intensity still supports the partial existence of the 2.18 MeV group as a real transition. It should also be noted that the present intensities of the 700 and 1480 MeV transitions are found to be 50% higher than those reported by Firsov and this could be attributed to the high summing probability of the peak at 2.18 MeV in his measurements. To decide whether the 1.67 and 1.16 MeV transitions are real or due to single and double escaped annihilation radiation from the 2.18 MeV transition or due to real gamma transitions of these energies plus the single and double escaped peaks

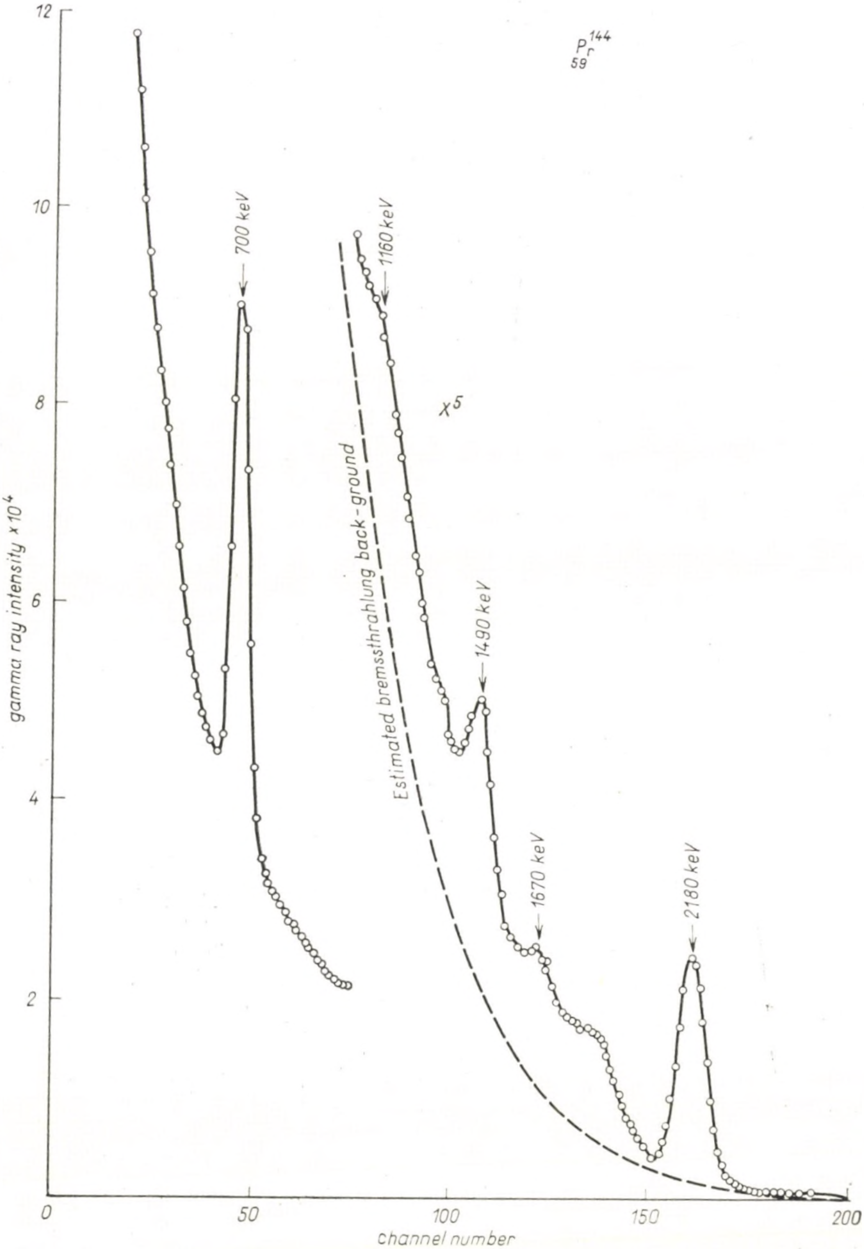

Fig. 1 Single spectrum of Pr^{144}

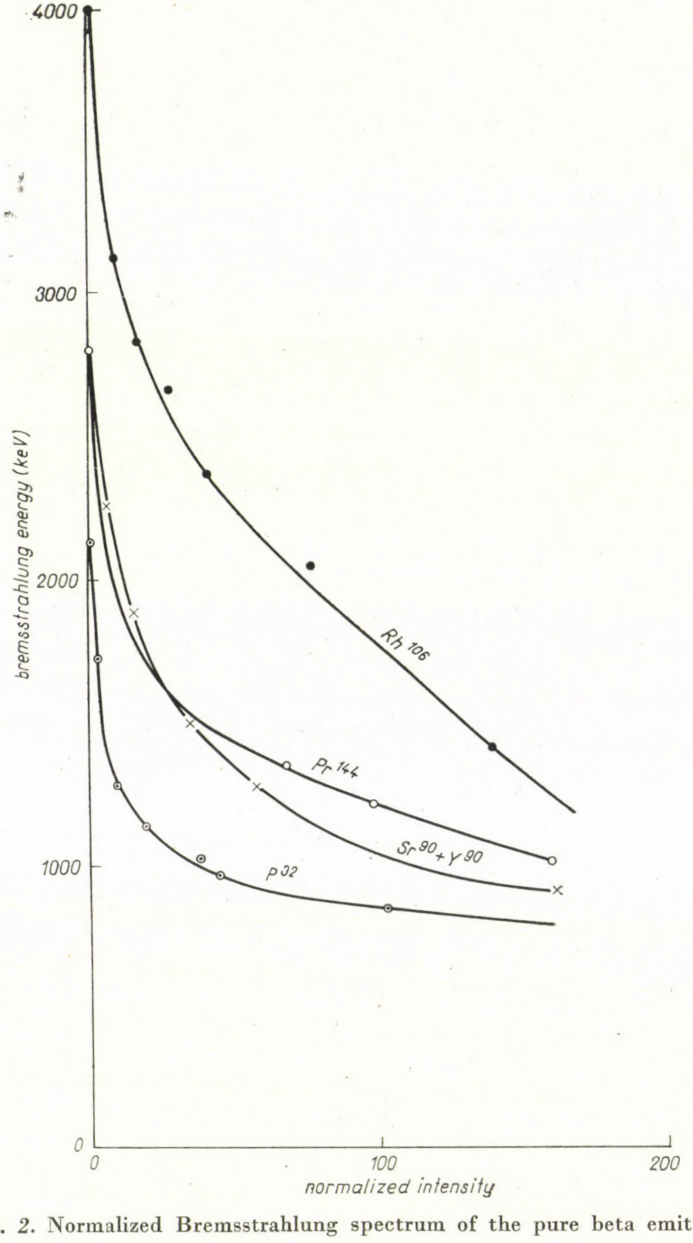

Fig. 2. Normalized Bremsstrahlung spectrum of the pure beta emitters

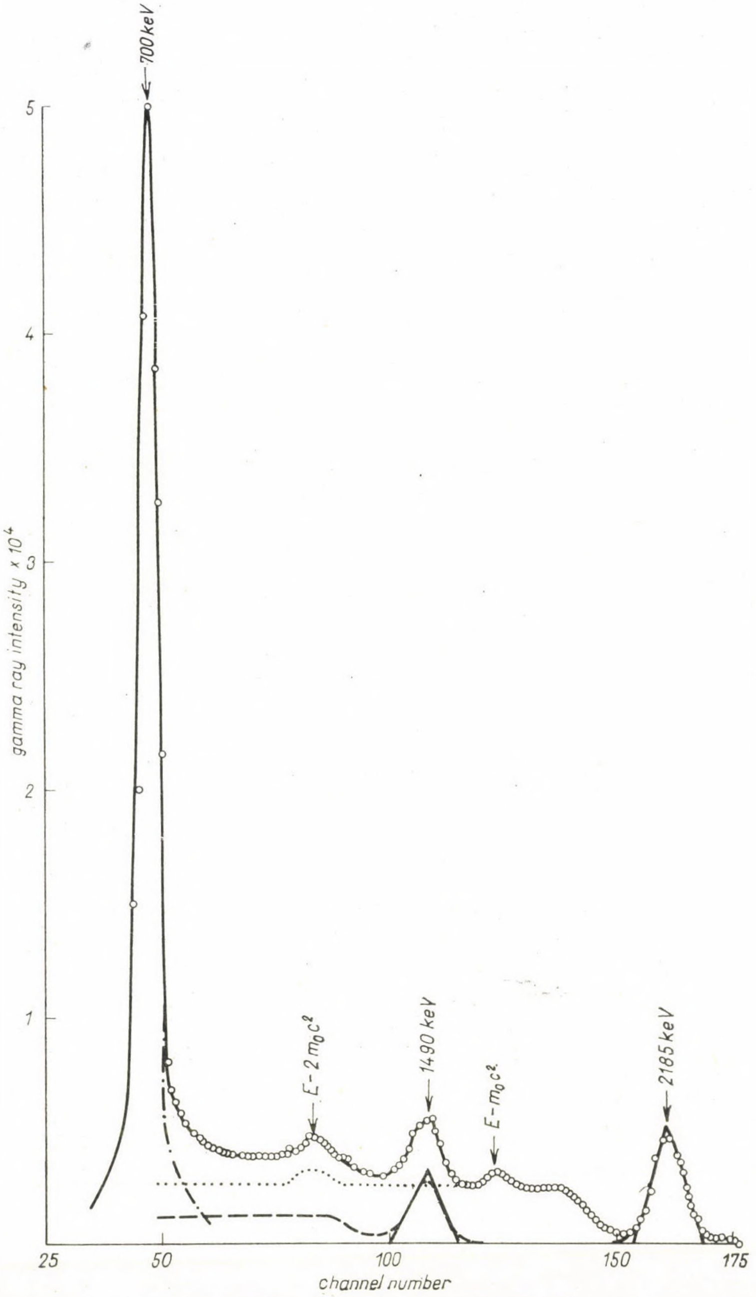

Fig. 3. Single spectrum of Pr^{144} after subtracting the normalized Bremsstrahlung spectrum

of the 2.18 MeV transition, the peeling off method [9] was applied. However, the presence of the intense 2.98 MeV beta group gives rise to a high background of Bremsstrahlung radiation and one has to subtract this from the single spectrum before carrying out the peeling-off method. The exact shape of the Bremsstrahlung radiation has been determined by applying sources of pure beta emitters of nearly the same intensity as the Ce^{144} source. The chosen beta emitters were P^{32}, $Sr^{90} + Y^{90}$ and Rh^{106} and their spectra were measured with the same geometry. The Bremsstrahlung spectrum of each mentioned standard isotope was normalized to the same intensity of the Pr^{144} source at the points corresponding to $E/2$, where E is the beta end-point energy of the measured sources. The normalized spectra of these isotopes are shown in Fig. 2 from which it is possible to calculate the normalized Bremsstrahlung spectrum of Pr^{144}. Fig. 3 shows the single spectrum of Pr^{144} after subtracting the normalized Bremsstrahlung spectrum. Applying the peeling off method the peaks at 1.67 MeV and 1.16 MeV were found to be the single and double escaped annihilation peaks of the 2.18 MeV transition. If these groups do exist the upper limit for their existence was estimated to be 0.01% and 0.016% from the intensity of the 2.18 MeV peak. The remaining gamma peaks at 0.7, 1.48 and 2.18 MeV seem to be real and their relative intensities, after being

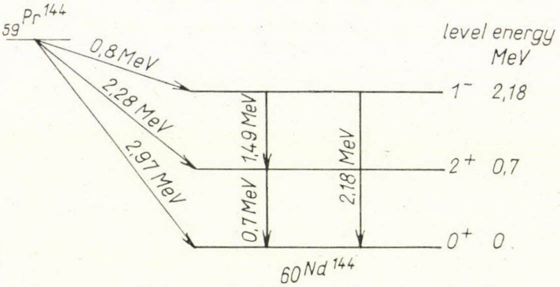

Fig. 4 Present decay scheme of Pr^{144}

Table I

Energies and relative intensities of gamma rays of Pr^{144}

E (keV)	Relative intensity
700 ± 15	240
1160 ± 20	< 0.016
1480 ± 20	48.3
1670 ± 30	< 0.016
2180 ± 30	100

corrected for the crystal efficiencies, are shown in Table I. In accordance with the above mentioned results the decay scheme of Pr^{144} is proposed as hsown in Fig. 4.

REFERENCES

1. W. Emmerich, W. Auth and J. Kurbatov, Phys. Rev., **94**, 110, 794, 1954.
2. D. Alburger and J. Kraushaar, Phys. Rev., **87**, 448, 1952 and **86**, 633, 1952.
3. G. John, L. Cheng and J. Kurbatov, Phys. Rev., **85**, 487, 1952 and **86**, 632, 1952.
4. F. Porter and C. Cook, Phys. Rev., **85**, 733, 1952 and **87**, 464, 1952.
5. H. Keller and J. Cork, Phys. Rev., **84**, 1079, 1951.
6. J. Cork, M. Brice and L. Schmid, Phys. Rev., **96**, 1295, 1954.
7. W. Kreger and C. Cook, Phys. Rev., **96**, 855, 1276, 1954.
8. E. J. Firsov and A. A. Basilov, Tezisy 7—90 Soveschanye po jardernoj spektroskopiji (1957).
9. M. Sc. Thesis by M. E. Morsy, Ain Shams University, Faculty of Science, 1966.

СХЕМА РАСПАДА Pr^{144}

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Резюме

Сцинтилляционным спектрометром был исследован 17,5-минутный распад Pr^{144}. Энергия гамма-перехода в ядре-дочери Nd^{144} составляет 2,18, 1,49 и 0,70 Мэв в соответствии с измерениями других исследователей. Двусмысленность по отношению ранее опубликованных значений 2,8, 1,67 и 1,16 Мэв при гамма-переходах ликвидирована. В последующей части предлагается схема распада.

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DECAY SCHEME OF Pd^{111} AND Pd^{111m}

By

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The gamma spectra accompanying the decay of the 22 min. Pd^{111} and the 5.5h Pd^{111m}, were studied with a scintillation gamma ray spectrometer. Seven new gamma transitions were found to be associated with the decay of Pd^{111}. The energies of these gamma rays are 220, 300, 350, 500, 770 and 1150 keV. A scheme representing the decay of Pd^{111} and Pd^{111m} is proposed.

1. Introduction

The beta and gamma ray spectra associated with the decay of the 5.5h Pd^{111m} and the 22 min Pd^{111} were first studied in 1952 by McGinnis [1] who observed a 2.15 MeV beta ray group and 60 keV conversion electrons in these decays. Additional beta and gamma ray measurements employing the scintillation spectrometer technique were later carried out either by irradiating enriched palladium metal with reactor neutrons (Pratt and Cochran [2]) or by investigating the Pd^{111m} activity from the reaction Pd^{110} (d, p) Pd^{111} as reported by Eccles [3]. These measurements indicated that the association of the beta ray groups to the isomeric and ground states was based on the existence of gamma rays having 22 min and 5.5h half lives. In the decay of the 22 min ground state Pratt and Cochran [2] observed beta rays of energy 2.18 MeV and gamma rays of energies 377, 580, 620, 810, 1380 and 1450 keV but no decay scheme was presented.

In the case of the 5.5h isomeric state these authors were able to associate with it beta rays of energy 2.02 MeV and gamma rays of energies 170 and 1690 keV. Eccles [3] confirmed these gamma ray groups and was able to report 24 additional groups of energies ranging from 67 to 1990 keV among which a number of weak and uncertain groups were listed. However, this author presented a partial level diagram of the daughter nucleus Ag^{111} on the basis of the majority of the observed gamma ray energies together with their relative intensities and a comparison between the low lying levels of the isotopes Ag^{107} and Ag^{109}.

The above review shows that a careful investigation is required for the decay of both the isomeric and ground states of Pd^{111} isotopes in order to check previous data as well as to propose reasonable decay schemes for both isotopes.

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2. Experimental procedure

Samples of spectroscopically pure Pd Cl_2 were irradiated either for 20 minutes or 5 hours in a flux of about 10^{13} thermal neutrons $\cdot$ cm $^{-2}$ $\cdot$ sec $^{-1}$ in the WW—Rl reactor at the UAR Atomic Energy Establishment. The separation of palladium by precipitation with dimethylglyoxime [4] was not found suitable for the study of the 22 min Pd^{111} since a long period was required. The process adopted for separating palladium from chlorine was to dissolve the irradiated sample in NH_4Cl in order to replace the active chlorine ions by inactive ones, and then forming a Cu—Pd amalgam by dropping some solution on copper foils of 0.2 mm thickness. After a few minutes the copper foils were washed with NH_4Cl and then, with distilled water. They were then heated over a direct flame to expel the existing chlorine. For the measurement of gamma ray spectra a scintillation spectrometer with a transistorised RIDL 200 channel pulse height analyser was used after it had been calibrated for energy and intensity. The applied detector is a 5.1 cm × 5.1 cm Nal (Tl) scintillator, optically coupled to a RCA 6810A photomultiplier. The resolution of this detector has been found to be better than 7.5% for the 661 keV gamma ray of Cs^{137}.

Measurement of the low energy region of the gamma ray spectrum was carried out while the source was in contact with the crystal or a few centimetres higher, depending on the source strength, whereas lead absorbers of 2—5 mm thickness were used in investigating the high energy region. The energies and relative intensities of the observed gamma ray groups were determined by applying the peeling off method [5].

In investigating the decay of the 22 min Pd^{111}, measurements started on the 20 min exposed samples within one hour of the end of the irradiation period. A number of successive measurements were then carried out continuously during a period of two hours and the corresponding half-life time of each gamma group was determined. These measurements proved that the entire spectrum was due to the 22 min Pd^{111}, the 5.5h Pd^{111m} and the 13.6h Pd^{109}. After a comparatively long period, the activity of the ground state Pd^{111} became negligible and the spectrum represented the activity of the remaining palladium nuclei. The intensity of the background spectrum due to Pd^{109} was stronger than that of Pd^{111m} as the abundance of Pd^{108} is twice that ofPd^{110} and their neutron capture cross-sections are 11 and 0.4 barns, respectively. However, the contributions of these background spectra measured after a period of 5 hours from the beginging of the experiment were subtracted from the first measurement, carried out with the same geometry, after making the necessary decay corrections. By this process the spectrum of the 22 min Pd^{111} was determined for various samples.

In the case of the Pd^{111m}, samples exposed for 5 hours were used after a few hours from the end of irradiation to secure a negligible contribution of the 22 min activity. In a similar way to the case of Pd^{111m} successive measurements were taken and the background contribution of the 13.6h Pd^{109} was subtracted from the total spectrum to obtain the pure spectrum of the 5.5 Pd^{111}.

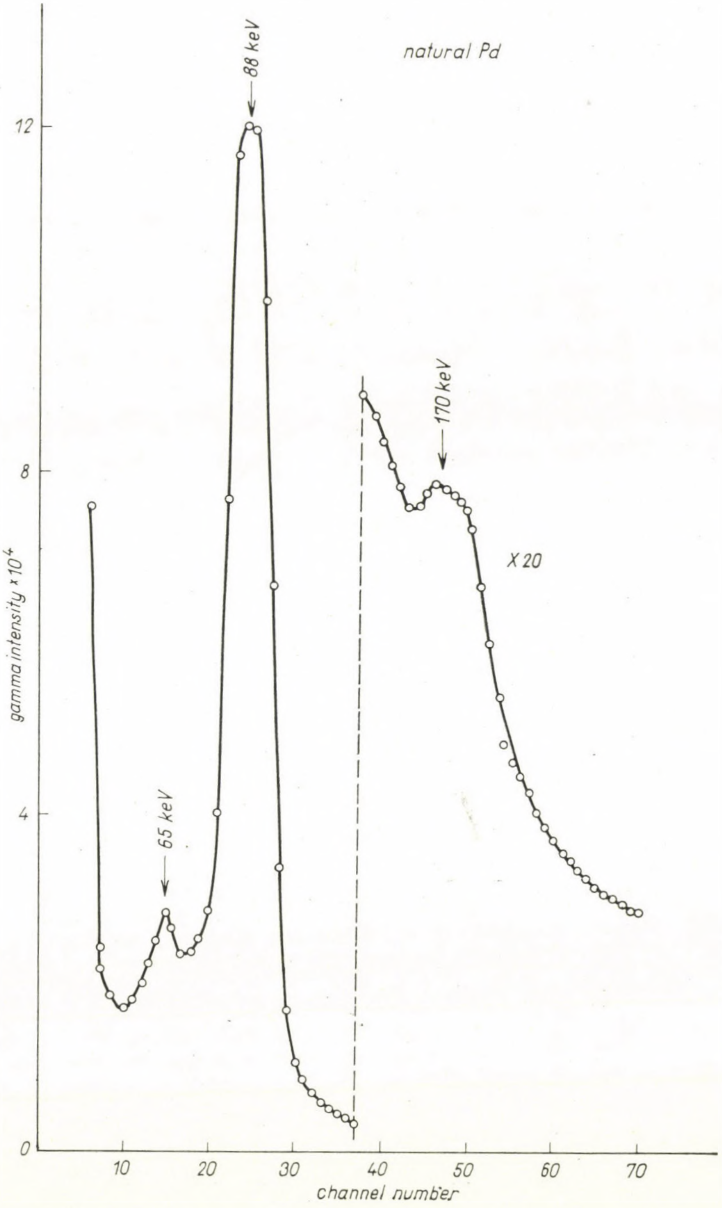

Fig. 1. Single spectrum of the low energy region of activated natural palladium

3. Results and discussion

Representative spectra of Pd^{111} and Pd^{111m} for the low and high gamma ray energy regions are shown in Figs. 1—4. The energies and relative intensities of the gamma transitions are presented in Tables I and II after correcting for crystal efficiencies and absorption, then normalizing the intensities to a value of 100 for the 220 keV gamma transition. A comparison with the energies of gamma transitions observed by previous authors is also presented in these Tables. The error in the relative intensity is estimated to be about 20% (relative error).

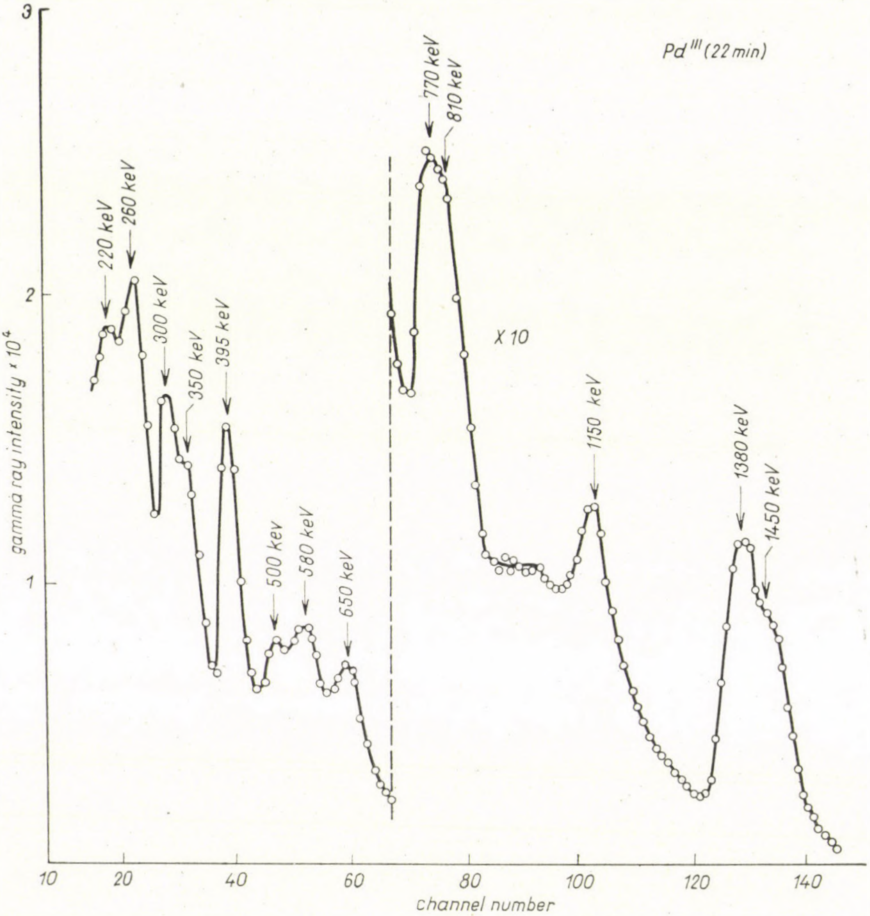

Fig. 2. Single spectrum of the high energy region of Pd^{111} using 2 mm Pd absorber

For the 22 min Pd^{111} seven new transitions of energies 220, 260, 300, 350, 500, 770 and 1150 keV were found to be associated with the decay of this isotope. The other six transitions of energies 395, 580, 650, 810, 1380

Table I

Energies and relative intensities of gamma rays of Pd^{111} (22 min)

Energy of gamma transitions in keV			Present relative intensity
McGinnis [1] 1952	Pratt and Cochran [2] 1960	Present work	
60	—	65 ± 5	
	—	220 ± 10	100
	—	260 ± 10	200
	—	300 ± 10	150
	—	350 ± 10	185
	377	395 ± 10	400
	—	500 ± 20	250
	580	580 ± 20	370
	620	650 ± 20	315
	—	770 ± 20	100
	810	810 ± 20	200
	—	1150 ± 20	85
	1380	1380 ± 30	100
	1450	1450 ± 30	140

and 1450 keV were found to be in agreement with the results of Pratt and Cochran [2].

In the case of the 5.5h Pd^{111m} the transitions of energies 65, 170, 300, 350, 395, 500, 580, 650, 810, 960, 1060, 1240, 1380, 1580, 1700 and 2000 keV were in agreement with the results of Eccles [3]. The transitions of energies 108, 128, 435, 479, 865, 1315 and 1780 and 1780 keV were reported by Eccles [3] in his decay scheme without representation due to their very weak intensity. In the present investigation special care was taken to search for these transitions, but nothing was observed.

Applying the present data of the energies and relative intensities of the gamma transitions, one can attempt to construct a number of decay schemes which may account for a good percentage of the observed transitions. However, the decay scheme proposed in the present work as shown in Fig. 5 is found to be most consistent with the experimental results as well as with the already established data. Moreover, its construction is based on the systematics of the low lying levels of the other silver isotopes. It should be pointed out that in order to have a satisfactory energy level scheme for Ag^{111}, the decay scheme of the 22 min Pd^{111} was first constructed and then the other decay scheme of the isomer was superimposed on it. The beta decay of Pd^{111} is considered to feed mainly (85%) the excited level of the daughter nucleus at 65 keV,

Table II

Energies and relative intensities of gamma rays of Pd^{111m} (5.5h)

Energies of gamma transitions in keV				Present relative intensity
McGinnis [1] 1952	Pratt and Cochran [2] 1960	Eccles [3] 1962	Present work	
		67	65 ± 5	
		108	—	
		128	—	
		148	—	
170	170	170	170 ± 10	8200
		—	220 ± 10	100
		—	260 ± 10	250
		293	300 ± 10	160
		367	350 ± 10	180
		395	395 ± 10	440
		435	—	
		479	—	
		523	500 ± 20	200
		584	580 ± 20	250
		650	650 ± 20	325
		727	685 ± 20	120
		—	770 ± 20	100
		828	810 ± 20	150
		865	—	
		963	960 ± 20	65
		1030	1060 ± 20	75
		1140	1150 ± 20	55
		1220	1240 ± 20	60
		1315	—	
		1400	1380 ± 30	50
		—	1450 ± 30	55
		1600	1580 ± 30	70
	1690	1690	1700 ± 30	65
		1780	—	
		—	1860 ± 30	40
		1990	2000 ± 30	50

while Pd^{111m} decays mainly (80%) to Pd^{111}. This latter value has been estimated from the present relative intensity of the 170 keV transition after correcting for the internal conversion using a value [7] of $\alpha = 1.24$ for this transition.

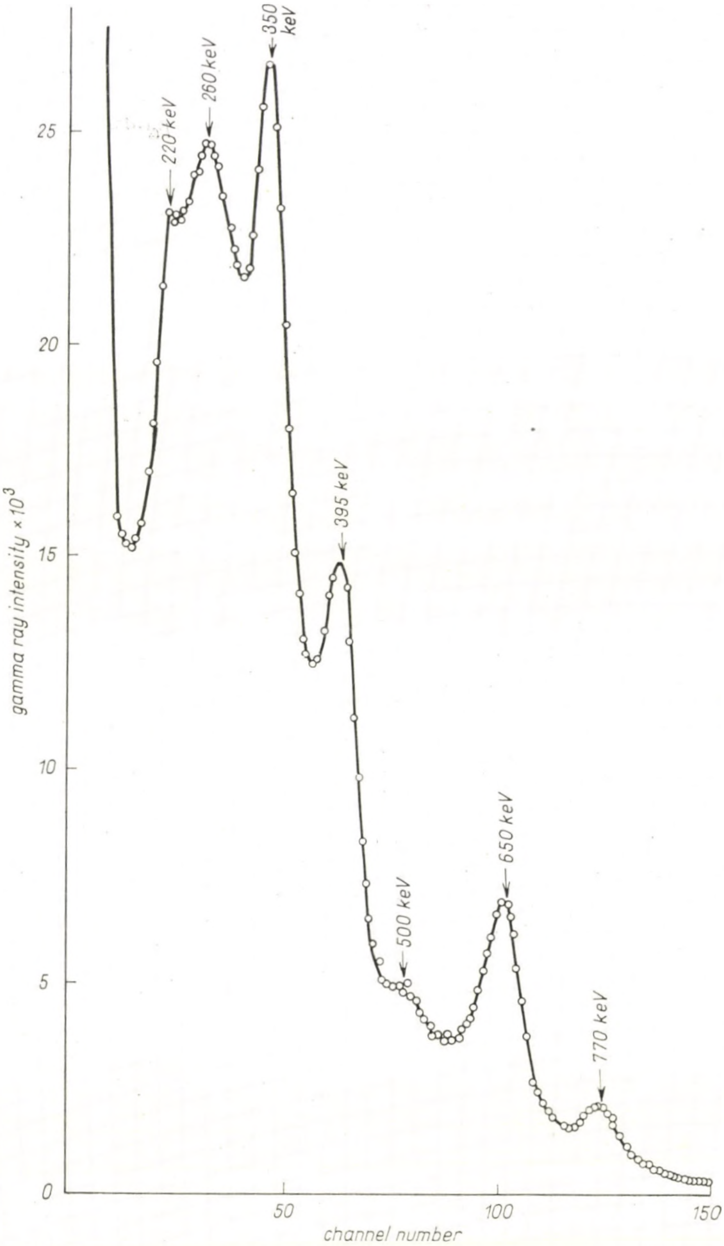

Fig. 3. Single spectrum of the low energy region of Pd^{111m}

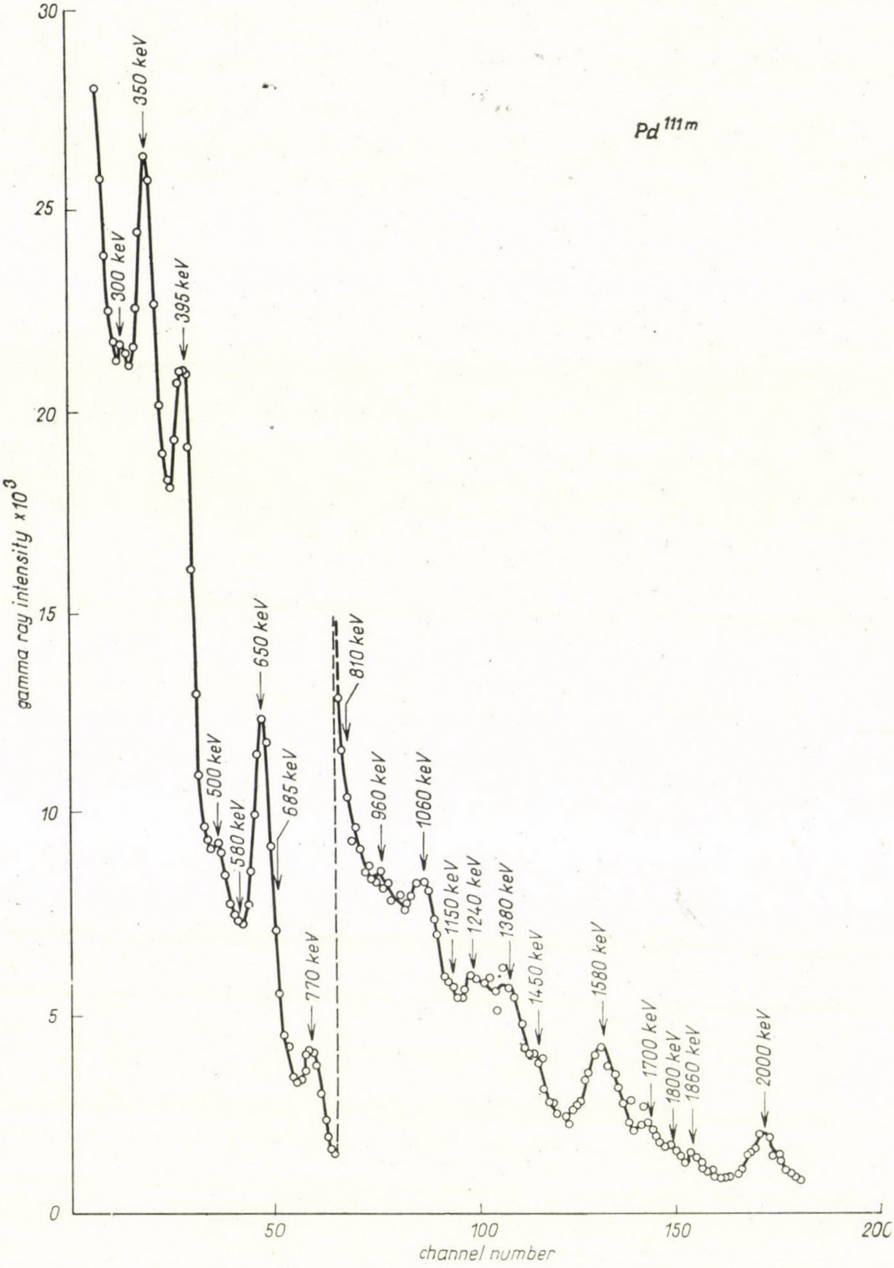

Fig. 4. Single spectrum of the high energy region of Pd^{111m} using 5 mm Pd absorber

Table III

Energies, relative intensities and log ft of different betas accompanying the decay of Pd^{111} and Pd^{111m}

Isotope	E_B keV	Relative intensity	Log ft
22 min Pd^{111}	730	2.9	4.8
	1110	4.6	5.4
	1600	5.6	5.6
	2115	58.0	5
5.5h $Pd^{111}C$	350	2.6	4.8
	490	1.4	5.6
	770	2.1	6.0
	1520	0.9	7.8

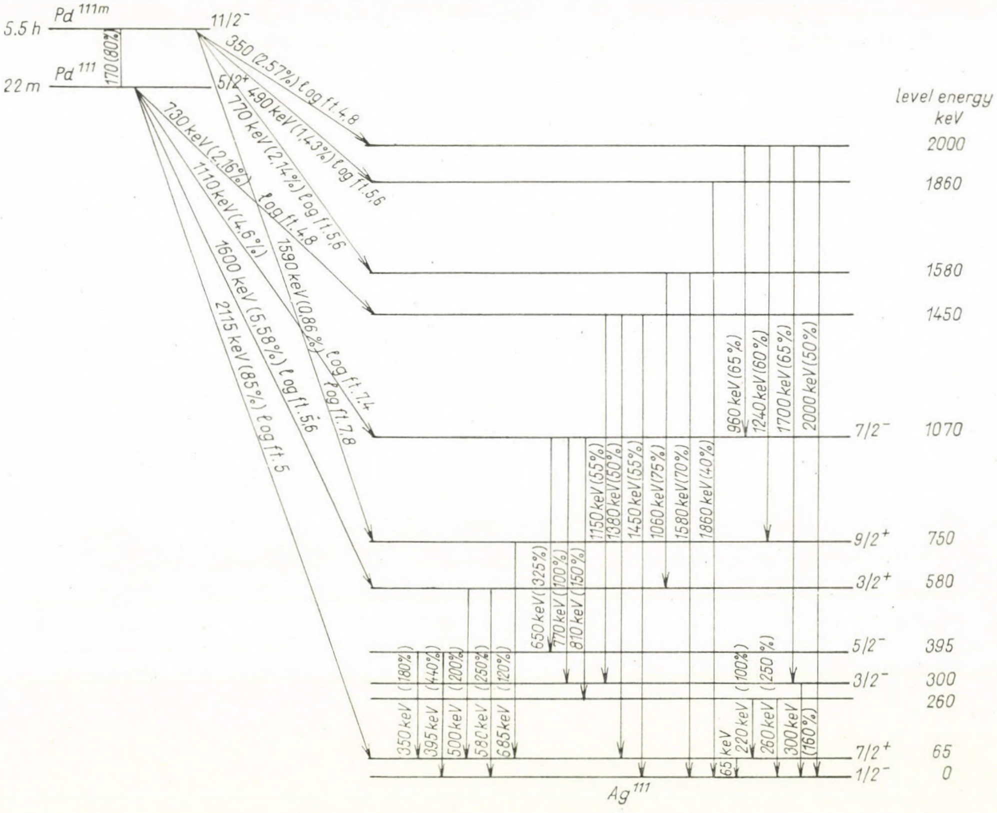

Fig. 5. Present decay scheme of Pd and Pd^{111m}

Assuming the proposed decay scheme to be correct the relative intensities of the beta components feeding the different levels in Ag^{111} were estimated according to the relative gamma intensities. On this basis, the log ft values for each beta group were calculated as presented in Table III. It can be seen that most of the beta transitions whether from the excited state or from the ground state of Pd^{111} appear to be allowed or first forbidden. Thus, a great variety of spins and parities are probable for each of the excited states in Ag^{111}.

The low lying excited states in Ag^{111} at 0, 65, 300 and 395 keV are known to have spins $1^-/2$, $7^+/2$, $3^-/2$ and $5^-/2$, respectively. For higher excited states the present investigation shows that the level at 580 keV can have the assignments $3^+/2$, $5^+/2$ or $7^+/2$ since this level is populated by an allowed beta component. This level is de-excited by emission of the 580 keV gamma transition (relative intensity 250) to the $1^-/2$ ground state and so its probable assignment is $3^+/2$. The level at 750 keV is fed by a first forbidden beta group, so it can have a spin and parity of $9^+/2$, $11^+/2$ or $13^+/2$. This level decays through a 685 keV gamma transition (relative intensity 120) to the $7^+/2$ level at 65 keV which makes its probable assignment to be $9^+/2$.

According to the assumption that the levels at 300 and 395 keV are the first and second rotational levels, the energies of the third and fourth rotational levels, of spins and parities $7^-/2$ and $9^-/2$ will theoretically be at 1070 and 1250 keV, respectively. The existence of the third rotational level is confirmed by the observation of direct transitions of energies 650 keV (relative intensity 315) and 770 keV (relative intensity 100) from the 1070 keV level to the second and first rotational levels at 395 and 300 keV, respectively. Moreover, the beta group follows that a spin value of $7^-/2$ can be assigned to this level.

REFERENCES

1. C. L. McGinnis, Phys. Rev., **87,** 202 A, 1952.
2. W. W. Pratt and R. C. Cochran, Phys. Rev., **118,** 1313, 1960.
3. S. F. Eccles, Physica, **28,** 251, 1962.
4. A. H. Wapstra and W. Vander Eijk, Nuclear Physics, **4,** 325, 1957.
5. R. Van Lieshout et al., Alpha, beta and gamma ray spectroscopy, ed. by K. Seighahn, North-Holland Publ. Co., Amsterdam, 1965.
6. Decay Scheme of Radioactive Nuclei by B. Dzelepov, L. Peker and V. Sergejev, Academy of Science of the USSR Press, Moscow, Leningrad, 1963.
7. L. Sliv and J. Band, Tables of Internal Conversion Coefficients, published in Moscow, Leningrad, 1956—1958.

СХЕМА РАСПАДА Pd^{111} и Pd^{111m}

Ф. А. Эл БЕДЕВИ, З. МИЛИГИ, Н. А. ЭИССА и М. МОРСИ

Резюме

Сцинтилляционным гамма-спектрометром изучался гамма-спектр распада Pd^{111} (22 мин) и Pd^{111m} (5,5 ч). Найдено семь новых гамма-переходов, принадлежащих к распаду Pd^{111}. Значения энергии данных гамма-лучей следующие: 220, 300, 350, 500, 700 и 1150 Кэв. Предлагается схема, представляющая распад Pd^{111} и Pd^{111m}.

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NONLINEAR MODEL IN QUANTUM FIELD THEORY

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The mass spectrum of a nonlinear real scalar field was sought for using Ritz's variational method. In the case of suitable renormalization 1, 2 or 3 finite values of rest mass were found. The different types of these excitations belong to different inequivalent representations of the field operators.

The programme of the nonlinear field theory has a maximal aim of describing a number of elementary particles and their interactions by a single field equation. After the first steps, however, enormous mathematical difficulties arise. Just because of these we do not yet know whether such a programme might prove successful, or whether it is inadequate for describing the world at all. Efforts have been made essentially in two directions. There have been attempts to investigate the content of an equation which has been regarded as a candidate for the full description. This line was followed by **Heisenberg** in establishing his famous equation [1]. On the other hand, the mathematical structure of simple models has been investigated in order to gain experience for the final solution.

This approach was successful in finding simple, exactly solvable models and in giving an approximate solution to other models [2]. These calculations yield some insight into the mathematical structure of nonlinear field theories. The present paper gives an account of such an attempt.

The nonlinear real scalar field, previously investigated by **Goldstone**, **Marx** and **Kuti** [3] has been modified by an additional term in the field equation:

$$\Box\varphi + \mu_0^2\varphi + \varkappa_0^2\varphi^2 - \lambda_0^2\varphi^3 = 0\,, \tag{1}$$

where $\varkappa_0^2\,\varphi$ is the new term, which destroys the $\varphi \to -\varphi$ symmetry of the original model. The solution was sought for within the framework of Lagrangian formalism and canonical quantization.

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§ 1. Canonical quantization

The Lagrangian corresponding to the field equation (1) is:

$$L = \frac{1}{2} \partial_\mu \varphi \, \partial_\mu \varphi + \frac{1}{2} \mu_0^2 \varphi^2 + \frac{1}{3} \varkappa_0^2 \varphi^3 - \frac{1}{4} \lambda_0^2 \varphi^4 ,$$

where $\mu_0^2, \varkappa_0^2, \lambda_0^2$ are real parameters. We use a system of units, in which $h = 1$, $c = 1$.

For the momentum conjugated to φ,

$$\pi = \frac{\partial L}{\partial \dot{\varphi}} = \dot{\varphi}, \tag{2}$$

we require canonical commutation relations:

$$[\varphi(r, t), \varphi(r', t)] = [\pi(r, t), \pi(r', t)] = 0 ,$$
$$[\pi(r, t), \varphi(r' t)] = - i\delta(r - r') .$$

The Hamiltonian and the field momentum is given by

$$H = \int (\pi \dot{\varphi} - L) \, d^3 , \tag{3}$$

$$P = - \frac{1}{2} \int (\pi \nabla \varphi + \nabla \varphi \cdot \pi) \, d^3 r . \tag{4}$$

Supposing that the field is enclosed in a cube of volume Ω we represent φ and π by Fourier expansions:

$$\varphi(r, t) = \frac{1}{\sqrt{\Omega}} \sum_k q_k(t) \, e^{ikr} , \tag{5}$$

$$\pi(r, t) = \frac{1}{\sqrt{\Omega}} \sum_k p_k(t) \, e^{-ikr}, \tag{6}$$

where the vector index $k = (k_x, k_y, k_z)$ runs over the values

$$k_i = 2 \pi \Omega^{-1/3} n_i \qquad n_i = 0, \pm 1, \pm 2, \pm \ldots \qquad i = x, y, z.$$

Eq. (2) is equivalent to the commutation relations

$$[p_k, q_{k'}] = - i\delta_{k,k'} ; \; [q_k, p_{k'}] = [q_k, q_{k'}] = 0 \tag{7}$$

and the hermiticity of φ and π requires

$$q_k^+ = q_{-k} , \qquad p_k^+ = p_{-k} .$$

The relations (3)—(7) define a quantized field: p_k, q_k, H, P are elements of an abstract algebra, and they obey the above relations according to the operations defined in this algebra.

A representation of this field means a homomorphism between the p_k, q_k algebra elements and $\hat{p}_k, \hat{q}_k$ operators acting on a separable Hilbert space $\mathscr{H}$. A representation is called "proper" if the set of linear operators acting on $\mathscr{H}$ contains elements, say $\hat{H}$ and $\hat{P}$, homomorphic to the algebra elements defined by eqs. (3) and (4). For a "physical" representation some further properties (existence of vacuum state, Lorentz invariance) are required.

The basic problem is to find proper representation. This is over and above the customary difficulties of solving the eigenvalue problem. Exact representations have been found so far only for very simple models.

Another difficulty beyond those of quantum mechanics is involved in the following. VON NEUMANN's theorem states [4] that in the case of a finite number of p_k, q_k variables (i.e. quantum mechanics) all irreducible representations are equivalent, i.e. they can be transformed into each other by unitary transformations. So, if one of these representations is proper, all of them are proper, and all representations give the same physical results (e.g. eigenvalues). For an infinite number of variables this does not hold.

§ 2. Generalized coordinate representation

Let us consider the linear space $\tilde{\mathscr{H}}$ of the twice differentiable functions of a numerable infinite number of real variables

$$\psi = \psi(x_0, x_1 \ldots) .$$

We define the following linear operations on $\tilde{\mathscr{H}}$:

$$\hat{X}_j \psi(x_0, x_1 \ldots) = x_j \psi(x_0, x_1 \ldots) ,$$

$$\hat{P}_j \psi(x_0, x_1 \ldots) = - \frac{\partial}{\partial x_j} \psi(x_0, x_1 \ldots) .$$

Since within each Hilbert space $\mathscr{H} \subset \tilde{\mathscr{H}}$, where the scalar product of any two elements of $\mathscr{H}$, say Φ and Φ' is defined in an appropriate way to realize the usual definition $(\Phi, \Phi') = \int \Phi^* \Phi' \, dx_0 dx_1 \ldots)$

$$[\hat{P}_j, \hat{X}_l] = i\delta_{jl} ,$$

$$[\hat{X}_j, \hat{X}_l] = [\hat{P}_j, \hat{P}_l] = 0$$

and $\hat{X}_i$, $\hat{P}_j$ are hermitian, it is easy to construct a representation of the commutation relations (7) using $\hat{X}_j$'s and $\hat{P}_j$'s.

So we identify the labels of the x_j variables with the k vectors of eqs. (5) and (6), and introduce the definitions,

$$\left.\begin{aligned}\hat{q}_k &= \frac{1}{\sqrt{2}}\left(\hat{X}_k + i\hat{X}_{-k}\right)\\ \hat{p}_k &= \frac{1}{\sqrt{2}}\left(\hat{P}_k - i\hat{P}_{-k}\right)\end{aligned}\right\} \text{ for } k > 0, \tag{8}$$

$$\hat{q}_{-k} = \hat{q}_k^+, \qquad \hat{p}_{-k} = \hat{p}_k^+, \qquad \hat{q}_0 = \hat{X}_0, \qquad \hat{p}_0 = \hat{P}_0\,, \tag{9}$$

where $k > 0$ means that the first nonzero coordinate of k is positive. Using the $z_k \in (0, \infty)$, $\Theta_k \in (0, 2\pi)$ $(k>0)$ new real variables defined by the equations

$$x_k = z_k \cos\Theta_k\,, \qquad x_{-k} = z_k \sin\Theta_k\,,$$

we have on the basis of eqs. (3), (4), (8) and (9) the algebraic expressions homomorphic to H and P:

$$\begin{aligned}\hat{H} = &- \frac{1}{2}\frac{\partial^2}{\partial x_0^2} - \frac{\mu_0^2}{2}x_0^2 + \frac{1}{2}\sum_{k>0}\left(-\Delta_k^{(2)} + (k^2 - \mu_0^2)\, z_k^2\right) - \\ &- \frac{\varkappa_0^2}{\sqrt{\Omega}}\left(\frac{1}{6\sqrt{2}}\sum_{k,l,m>0}^{(\pm)} z_k\, z_l\, z_n\, e^{i(\pm\Theta_k \pm \Theta_l \pm \Theta_m)}\,\delta(\pm k \pm l \pm m) + \right.\\ &\left. + x_0 \sum_{k>0} z_k^2 + \frac{1}{3}x_0^3\right) + \\ &+ \frac{\lambda_0^2}{\Omega}\left(\frac{1}{16}\sum_{k,l,m,n>}^{(\pm)} z_k\, z_l\, z_m\, z_n\, e^{i(\pm\Theta_k \pm \Theta_l \pm \Theta_m \pm \Theta_n)}\,\delta(\pm k \pm l \pm m \pm n) + \right.\\ &+ \frac{1}{2\sqrt{2}}x_0 \sum_{k,l,m>0}^{(\pm)} z_k\, z_l\, z_m\, e^{i(\pm\Theta_k \pm \Theta_c \pm \Theta_m)}\,\delta(\pm k \pm l \pm m) + \\ &\left. + \frac{3}{2}x_0^2 \sum_{k>0} z_k^2 + \frac{1}{4}x_0^4\right)\end{aligned} \tag{10}$$

$$\hat{P} = \sum_{k>0} ik\frac{\partial}{\partial\Theta_k}\,,$$

where $\Delta_k^{(2)}$ is the two dimensional Laplace operator in the polar variables z_k, Θ_k and $\Sigma^{(\pm)}$ means summation over all indicated combinations of signs too, and

$$\delta(k) = \begin{cases} 1, & \text{if } k = 0,\\ 0, & \text{if } k \neq 0.\end{cases}$$

§ 3. Variational method

According to SCHIFF [5] we shall apply Ritz's variational method to approximate the proper representations with the following reasoning. In the Hilbert space of a "good" representation the eigenfunctions of $\hat{H}$ realize different "local" or "conditional" minimum values of the expectation value $(\psi, \hat{H}\psi)$. Obviously, if we carry out this minimalization in a "larger" (non-separable) Hilbert space, the resulting functions possess a minimum property in a "smaller" (physically reasonable, i.e. separable) Hilbert space contained in the original larger space.

We use the trial functions

$$\Psi = \Psi_0(x_0) \prod_{k>0} \Psi_k(z_k, \Theta_k) . \tag{11}$$

Thus a single φ_k function contains the variables x_k and x_{-k}. A sufficient normalization condition reads:

$$\begin{aligned} \langle \Psi_0 | \Psi_0 \rangle &= \int_{-\infty}^{+\infty} | \Psi_0(x_0) |^2 \, dx_0 = 1 , \\ \langle \Psi_k | \Psi_k \rangle &= \int_0^{2\pi} d\Theta_k \int_0^{\infty} | \Psi_k(z_k, \Theta_k) |^2 \, z_k \, dz_k . \end{aligned} \tag{12}$$

Since $\hat{H}$ and $\hat{}$ commute with each other, we are looking for their common eigenfunctions. The eigenfunctions of $\hat{P}$ having the form given by eq. (11) are

$$\Psi = \Psi_0(x_0) \prod_{k>0} f_k(z_k) \, e^{-in_k\Theta_k} ,$$

where ψ_0, f_k are arbitrary functions, $n_k = 0, \pm 1, \pm 2, \ldots$. The corresponding eigenvalue is $\vec{K} = \sum_{k>0} kn_k$

For such wave functions

$$\begin{aligned} \langle \Psi | \hat{H} | \Psi \rangle = & \left\langle \Psi_0 \right| - \frac{1}{2} \frac{\partial}{\partial x_0^2} - \frac{\mu_0^2}{2} x_0^2 - \frac{\varkappa_0^2}{3\sqrt{\Omega}} x_0^3 + \frac{\lambda_0^2}{4\Omega} x_0^4 \left| \Psi \right\rangle + \\ & + \sum_{k>0} \left\langle \Psi_k \right| - \frac{1}{2} \Delta_k^{(2)} + \frac{1}{2} (k^2 - \mu_0^2) z_k^2 + \frac{3\lambda_0^2}{8\Omega} z_k^4 \left| \Psi_0 \right\rangle - \\ & - \frac{\varkappa_0^2}{\sqrt{\Omega}} \langle \Psi_0 | x_0 | \Psi_0 \rangle \sum_{k>0} \langle \Psi_k | z_k^2 | \Psi_k \rangle + \frac{3\lambda_0^2}{4\Omega} \sum_{\substack{k \neq l \\ k,l>0}} \langle \Psi_k | z_k^2 | \Psi_k \rangle \langle \Psi_l | z_l^2 | \Psi_l \rangle + \\ & + \frac{3\lambda_0^2}{2\Omega} \langle \Psi_0 | x_0^2 | \Psi_0 \rangle \sum_{k>0} \langle {}_k | z_k^2 | \Psi_k \rangle . \end{aligned} \tag{13}$$

The auxiliary conditions (12) will be inserted into the minimalization with the Lagrange multipliers E_0, E_k $(k > 0)$: These are equivalent to the following system of coupled differential equations:

$$\left(-\frac{1}{2}\frac{\partial^2}{\partial x_0^2} + V(x_0)\right)\Psi_0 = E_0\Psi_0\,, \tag{14}$$

where

$$V(x_0) = \frac{\lambda_0^2}{4\,\Omega}x_0^4 - \frac{\varkappa_0^2}{3\sqrt{\Omega}}x_0^3 - \frac{1}{2}\mu^2 x_0^2 - \nu^2\sqrt{\Omega}\,x^2\,,$$

$$\mu^2 = \mu_0^2 - \frac{3\,\lambda_0^2}{\Omega}\sum_{k>0}\langle\Psi_k|z_k^2|\Psi_k\rangle,$$

$$V^2 = \frac{\varkappa_0^2}{\Omega}\sum_{k>0}\langle\Psi_k|z_k^2|\Psi_k\rangle$$

and

$$\left(-\frac{1}{2}\Delta_k^{(2)} + V_k(z_k)\right)\Psi_k = E_k\Psi_k\,, \tag{15}$$

where

$$V_k(z_k) = \frac{3\,\lambda_0^2}{8\,\Omega}z_k^4 + \frac{1}{2}\Bigg(k^2 - \mu^2 + \frac{3\,\lambda_0^2}{\Omega}\langle\Psi_k|z_k^2|\Psi_k\rangle +$$

$$+ \frac{3\,\lambda_0^2}{\Omega}\langle\Psi_0|x_0^2|\Psi_0\rangle - \frac{2\,\varkappa_0^2}{\sqrt{\Omega}}\langle\Psi_0|x_0|\Psi_0\rangle\Bigg)z_k^2\,.$$

Eqs. (14) and (15) are identical to the Schrödinger equations of the one and two dimensional motion of a free particle of mass unity. $V(x_0)$ may lead to one dimensional anharmonic oscillations around two equilibrium positions x_A and x_B (Fig. 1), whereas $V_k(z_k)$ correspond to two dimensional anharmonic oscillations around $z_k = 0$.

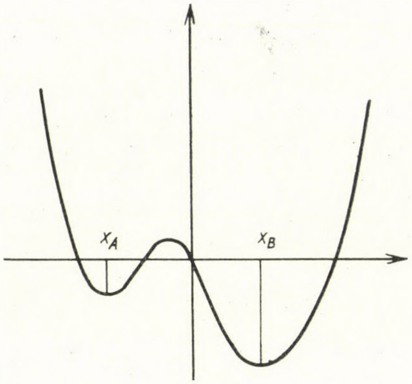

Fig. 1

When solving these equations we must take into account that $\hat{H}$ is defined by a limit $\hat{H} = \lim\limits_{k\to\infty} \hat{H}_K$, where $\hat{H}_k$ is obtained from eq. (10) by summing only over indices $|k|, |l|, |m|, |n| < K$.

This is why the parameters λ_0^2, μ_0^2 and $\varkappa_0^2$ can be considered as functions of K. It is sufficient to require weak convergence, that is:

$$\langle \Psi | \hat{H} | \Phi \rangle = \lim_{K\to\infty} \langle \Psi | H_K \varkappa \,|\big(\lambda_0^2(K), \mu_0^2(K), \varkappa_0^2(K)\big)| \Phi \rangle \tag{16}$$

for all $\Psi, \Phi \in \mathscr{H}$, the Hilbert space of the specified representation. Eq. (16) defines $\hat{H}$. We are interested in large values of Ω, but the $\Omega \to \infty$ limit does not alter our results at all, so we shall omit it. We mention that this is not inconsistent, since all physical measurements are confined to a finite region of space. But, on the other hand, the $\Omega \to \infty$ limit may lead to superfluous complications concerning inequivalent representations [6].

§ 4. Renormalized solution

We approximate the solution of the system (14)—(15) by the ground state eigenfunctions of one dimensional harmonic oscillators for ψ_0 and by those of two dimensional harmonic oscillators for ψ_k. The corresponding frequencies are $\omega_0^{(t)} = m^{(t)}$ and $\omega_k^{(t)}$, where t refers to the type of the solution x_A or x_B according to Fig. 1. (In what follows t means "A or B".)

Thus

$$\begin{aligned} \langle \Psi_0 | x_0 | \Psi_0 \rangle &= x_t\,, \\ \langle \Psi_0 | x_0^2 | \Psi_0 \rangle &= x_t^2 + \frac{1}{2\,\omega_0{}^{(t)}}\,, \\ \langle \Psi_k | z_k^2 | \Psi_k \rangle &= \frac{1}{\omega_k{}^{(t)}}\,. \end{aligned} \tag{17}$$

So we obtain the following relations as conditions for the self-consistency of our approximate solution:

$$V(x_0) = \frac{1}{2}(\omega_0{}^{(t)})^2 (x_0 - x_t)^2 \quad + \text{ terms of higher order,}$$

$$V_k(z_k) = \frac{1}{2}(\omega_k{}^{(t)})^2 z_k^2 \qquad\quad + \text{ terms of higher order.}$$

Using eqs. (14), (15) and (17), we have:

$$\begin{aligned} \omega_0{}^{(t)2} &= 3\,\lambda_0^2 \left(\frac{x_t}{\sqrt{\Omega}}\right)^2 - 2\,\varkappa_0^2 \left(\frac{x_t}{\sqrt{\Omega}}\right) - \mu^2 , \\ \omega_k{}^{(t)2} &= k^2 + 3\,\lambda_0^2 \left(\frac{x_x}{\sqrt{\Omega}}\right)^2 - 2\,\varkappa_0^2 \left(\frac{x_t}{\sqrt{\Omega}}\right) - \mu^2 = \omega_0{}^{(t)2} + k^2 , \end{aligned} \tag{18}$$

and x_t is subjected to the condition $V(x_t) =$ extremum, i.e.

$$\lambda_0^2 \left(\frac{x_t}{\sqrt{\Omega}}\right)^3 - \varkappa_0^2 \left(\frac{x_t}{\sqrt{\Omega}}\right)^2 - \mu^2 \left(\frac{x_t}{\sqrt{\Omega}}\right) - \nu^2 = 0 , \tag{19}$$

where

$$\mu^2 = \mu_0^2 - 3\,\lambda_0^2 Q_t , \qquad \nu^2 = \varkappa_0^2 \cdot Q_t , \tag{20}$$

$$Q_t = \frac{1}{\Omega} \sum_{k>0} \frac{1}{\omega_k^{(t)}} .$$

The system of equations (18)—(20) does not have a finite solution for non-vanishing λ_0^2, $\varkappa_0^2$, since Q is a quadratically divergent function of K:

$$\begin{aligned} Q_t(K) &= \frac{1}{\Omega} \sum_{|k|<K} \frac{1}{\omega_k^{(t)}} \approx \frac{1}{(2\,\Omega)^3} \int_{-\pi/2}^{\pi/2} d\varphi \int_0^{\pi} d\vartheta \int_0^K dk \, \frac{k^2 \sin\vartheta}{\sqrt{k^2 + m_t^2}} = \\ &= \frac{1}{(2\,\pi)^2} \left(\frac{K}{2} \sqrt{K^2 + m_t^2} - \frac{m_t^2}{2} \ln \frac{K + \sqrt{K^2 + m_t^2}}{m_t} \right) . \end{aligned}$$

So we are forced to introduce the renormalized parameters which may be chosen to be real finite numbers:

$$\begin{aligned} \lambda^2 &= \lim_{K\to\infty} \lambda_z^2(K) \ln K , \\ \bar{\mu}^2 &= \lim_{K\to\infty} \left(\mu_0^2(K) - \frac{3\,\lambda_0^4(K)\,K^2}{2\,(2\,\pi)^2} \right) , \\ \varkappa^2 &= \lim_{K\to\infty} \varkappa_0^2(K) \, \frac{K^2}{2\,(2\,\pi)^2 \sqrt{\ln K}} . \end{aligned}$$

Thus eqs. (18)—(20) take the form for the interesting limit $K \to \infty$

$$\begin{aligned} 3\,\lambda^2 \eta_t^2 - \mu^2 &= m_t^2 , \\ \lambda^2 \eta_t^3 - \mu^2 \eta_t - \varkappa^2 &= 0 , \\ \mu^2 &= \bar{\mu}^2 + \frac{3\,\lambda^2}{2\,(2\,\pi)^2} m_t^2 ; \end{aligned} \tag{21}$$

where

$$\eta_t = \lim_{K \to \infty} \left(\frac{x_t(K)}{\sqrt{\Omega}\,\sqrt{\ln K}} \right) .$$

We are interested in the solution m_t of this system. The dependence of m_t on $\lambda^2, \bar{\mu}^2, \varkappa^2$, i.e. on the bare parameters $\lambda_0^2(K), \varkappa_0^2(K), \mu_0^2(K)$ is shown in Fig. 2. λ^2 and $\varkappa^2$ are always taken as positive (in the case of the latter the negative sign corresponds only to the transformation $\varphi \to -\varphi$). On the other hand, for both positive and negative values of $\bar{\mu}^2$ we can get right solutions, which we have on the right and left hand sides of the Figure, respectively. "A" and "B" refer to the type of the solution as denoted in Fig. 1. We could take into account the ignored terms of the Schrödinger eqs. (14), (15) by more accurate calculation of the eigenfunctions. It is easy to see, however, that these corrections do not influence eqs. (18)—(20) at all, even in the case of finite Ω[1]. This means that our renormalized solution to the variational problem is exact.

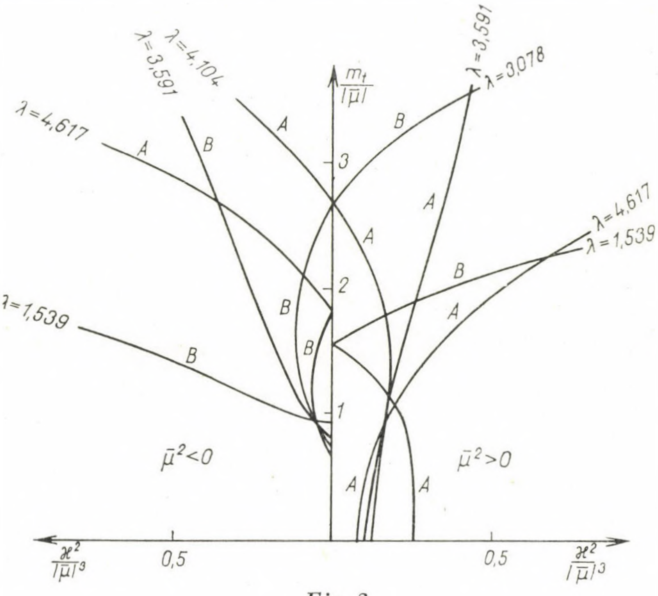

Fig. 2

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[1] E.g. To first order in the perturbation the corrections due to the terms $3\lambda_0^2\, z_k^4\,/8$ are

$$\langle \psi_k \,|z_k^2|\, \psi_k \rangle - \langle \psi_k^0 \,|z_k^2|\, \psi_k^0 \rangle = o\left(\frac{1}{\omega_k^7}\right).$$

The contribution of these to Q_t is:

$$o\left(\lambda_0^2(K) \int_0^k \frac{k^2\, dk}{(\sqrt{k^2 + m_t^2})^7}\right) = \lambda_0^2(K)\, o(m_t).$$

Because of $\lambda_0^2(K) \to 0$ for $K \to \infty$ this equals 0. This holds for higher order corrections too.

§ 5. Mass spectrum

Let us consider the wave functions

$$\Psi_t^{n_0;\ldots;n_k,m_k;\ldots} = \psi^{n_0}(x_0 - x_t) \prod_{0>0} \psi^{n_k,m_k}(z_k, Q_k), \tag{22}$$

where ψ^{n_0} is the energy eigenfunction of the one dimensional harmonic oscillator belonging to the eigenvalue $\omega_0^{(t)}(n_0 + 1/2)$, the $\psi_k^{n_k,m_k}$ are eigenfunctions of two dimensional harmonic oscillators belonging to the energy eigenvalues $\omega_k^{(t)}(n_k + 1)$ and the $\partial/\partial\Theta_k$ eigenvalue m_k ($m_k = -n_k, -n_k + 2 \ldots n_k - 2, n_k$). Let the connection between the ω's be given by $\omega_k^{(t)} = \sqrt{\omega_0^{(t)2} + k_2}$.

Consider those $\psi^{n_0;n_k,m_k;\ldots}$ functions for which only a finite number of n_0 and n_k are different from 0. These functions form a basis for a (separable) Hilbert space $\mathscr{H}_t^{(r)}$, and since $\lambda_0^2(K) \to 0$ if $K \to \infty$, the Q value for each $\psi^{n_0,n_k,m_k,\ldots}$ of this set is exactly the same. Consequently, we obtain from eqs. (21) the same $\omega_0^{(t)} = m_t$ for all these functions. Thus we have a system of exact solutions of the variational equations, for which

$$\hat{P}\psi_t^{n_0;\ldots;n_k,m_k;\ldots} \left(\sum_{k>0} k m_k\right) \varphi_t^{n_0;\ldots;n_k,m_k;\ldots}$$

and

$$\langle \psi_t^{n_0;\ldots;n_k,m_k;} | \hat{H} | \psi^{n_0,\ldots;n_k,m_k;\ldots} \rangle = \omega_0^{(t)} n_0 + \sum_{k>0} \omega_k^{(t)} n_k + E_0^{(t)},$$

where

$$E_0^{(t)} = \frac{1}{2}\omega_0^{(t)} + \sum_{k>0} \omega_k^{(t)} = \frac{3x^2}{4\Omega}\left(\sum_{k>0} \frac{1}{\omega_k^{t)}}\right)^2 +$$
$$+ \Omega\left(-\frac{\mu_0^2}{2}\left(\frac{x_t}{\sqrt{\Omega}}\right)^2 - \frac{x^2}{3}\left(\frac{x_t}{\sqrt{\Omega}}\right)^3 + \frac{\lambda_0^2}{4}\left(\frac{x_t}{\sqrt{\Omega}}\right)^4\right)$$

is the infinite zero point energy.

So we have obtained in our approximation the relativistic mass-momentum spectrum of free particles of mass m_t determined by eqs. (21). We cannot claim, however, that we have found the eigenstates of $\hat{H}$, or, at least, that our representation defined by $\hat{p}_k$, $\hat{q}_k$'s acting on our $H_t^{(r)}$ space is a proper one. Although

$$\langle \psi_t^{n_0;\ldots;n_k,m_k} | \hat{H} | \psi_t^{n_0;\ldots;n_k.m_k;\ldots} \rangle = \delta_{n_0 n_6}; \ldots; \delta_{n_k, n_k}, \delta_{m_k, m_k}; \ldots$$

holds for the basis functions (in the limit $K \to \infty$ by definition), in general $\langle \Psi | H | \Phi \rangle$ does not exist for every $\Psi, \Phi \in \mathscr{H}_t^{(r)}$.[2]

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[2] E.g.: $\lim\limits_{K\to\infty} \| H(K)\psi^{0;\ldots;0,0;\ldots} \|^2 = \langle H\psi^{0;\ldots0,0;\ldots} | H | \psi^{0;\ldots;0,0;\ldots} \rangle = \infty$.

§ 6. Conclusions

To the extent determined by the form (11) of the trial functions we have found the exact solutions (22) of the variational equations (13), which in turn define a representation in the Hilbert space $\mathscr{H}_t^{(r)}$.[3] We have seen that given the bare parameters $\lambda_0^2, \mu_0^2, \varkappa_0^2$ we may get 3, 2, 1 or 0 solutions (see Fig. 2), contrary to the classical case of 2 or 1 solutions. Our different solutions correspond to different inequivalent representations of the field operators φ and π, of course [7]. In the representations of the type A or B, $\langle \psi \mid \varphi \mid \psi \rangle$ equals $x_A \to -\infty$ or $x_B \to +\infty$, respectively. By our approximation, which is not a "too good" one, as mentioned in the previous section, we have obtained particles of different masses described by the same field equation. However, these different particles belong to different inequivalent representations. So, in the framework of the usual quantum theory they cannot exist simultaneously: the model has no solution containing excitations corresponding to different solutions of eqs. (21). An interpretation which would give physical reality to more than one excitation described by different solutions of our single field equation lies outside the realm of the present form of quantum theory.

It is worthwhile mentioning that the self-consistent method of KAMEFUCHI and UMEZAVA [8], based on the Bogoljubov transformation also leads to eqs. (21), while the non-self-consistent approximation of GOLDSTONE [3] does not give all terms in these equations. These facts emphasize the importance of self-consistency.

The author is very much indebted to Prof. G. MARX, who suggested this work and supported his first steps. Grateful thanks are due to Dr. G. KUTI for his continuous help during the whole work, and to B. PÁZMÁNY for his kind help with computer calculations.

REFERENCES

1. H. P. DÜRR and W. HEISENBERG, Z. Naturf., **16a**, 726, 1961.
2. E. M. HENLEY and W. THIRRING, Elementary Quantum Field Theory, New York, 1962.
3. I. GOLDSTONE, Nuovo Cimento, **19**, 154, 1961; G. MARX, Acta Phys. Hung., **14**, 27, 1962; G. KUTI and G. MARX, Acta Phys. Hung., **17**, 125, 1964; G. MARX and G. KUTI, preprint Budapest, 1964; G. KUTI and G. MARX, Acta Phys. Hung., **19**, 67, 1965.
4. J. v. NEUMANN, Math. Ann., **104**, 570, 1931.
5. L. I. SCHIFF, Phys. Rev., **130**, 458, 1963.
6. R. HAAG and D. KASTLER, J. of Math. Phys., **5**, 848, 1964.
7. R. HAAG, Brandeis University Summer Institute in Theoretical Physics, 353, 1960.
8. S. KAMEFUCHI and H. UMEZAVA, Nuovo Cimento, **31**, 429, 1964.

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[3] It is easy to show that our solution to the variational problem is exact only within the separable Hilbert space $\mathscr{H}_t^{(r)}$.

НЕЛИНЕЙНАЯ МОДЕЛЬ В КВАНТОВОЙ ТЕОРИИ ПОЛЯ

Ф. МЕЗЕИ

Резюме

В работе искался массовый спектр нелинейного реального скалярного поля вариационным методом Ритца. В случае соответствующей ренормализации найдены конечные значения массы покоя, равные 1, 2 или 3. Различные типы данных возбуждений принадлежат к различным неэквивалентным представлениям операторов поля.

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COMMUNICATIONES BREVES

Z → 0 LIMITS IN THE LEE MODEL

By

K. L. NAGY

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One of the several conditions for a particle to be composite is that its wave function renormalization constant Z should vanish. It has also been noted that for determining the mass and coupling constant of the particle, a condition for the vertex renormalization constant $Z_1 = 0$, and the behaviour of their ratio should also be used in addition (for an extensive list of references see e.g. [1]). In the Lee model, however, where the vertex function remains unrenormalized, $Z = 0$ was considered as a sole condition [2]. Here we wish to point out a situation where $Z = 0$ can be arranged without changing the elementary V-particle into a composite one. In this case, incidentally, the renormalized coupling constant turns out also to be zero, and we still have scattering and LEVINSON's theorem takes the form

$$\delta_t - \delta_\infty = \left(N_t - N_e + \frac{1}{2}\right)\pi\,, \tag{1}$$

a well known form in potential scattering [3] but not frequently discussed in elementary particle physics. Here N_t means the number of all stable states definitely below the threshold, N_e the number of elementary particle states, δ_t is the limit of the scattering phase at the threshold from above. 1/2 appears if only a bound state pole sits just at the threshold.

In the Lee model all the information for the $N - \theta$ scattering and for the real V-particle stem from an analytic function of the complex z variable $h(z)$ cut along the real axis from m_θ to $+\infty$. Thus, e.g.

$$S = e^{2i\delta(\omega)} = \frac{h(\omega - i\varepsilon)}{h(\omega + i\varepsilon)}\,, \qquad \omega \geq m_\theta\,, \tag{2}$$

$h(m_\theta) = 0$ gives the real mass of the V-particle ($m_n = 0$),

$$Z^{-1} = g_0^2\, h'(z)|_{m_V},$$
$$g^2 = g_0^2\, Z = \frac{1}{h'(z)|_{m_V}}\,, \text{ etc.} \tag{3}$$

For the Lee model with cut off, $h(z)$ expressed by $m_v(< m_\theta)$ and the bare coupling constant g_0 is

$$h(z) = (z - m_\nu)\left[\frac{1}{g_0^2} + \int_\theta^\infty \frac{|f(\omega)|^2\sqrt{\omega^2 - m_\theta^2}\,d\omega}{2\,(\omega - m_\nu)(\omega - z)}\right]. \tag{4}$$

If g_0^2 is finite one gets the elementary V-particle case manifesting in $Z \neq 0$ and also in $\delta(m_\theta) - \delta(\infty) = 0$. $g_0^2 \to +\infty$ gives the solution obtained by Vaughan et al. [2] with $Z = 0$, and $\delta(m_\theta) - \delta_\infty) = \pi$ expressing the compositeness of V. In these cases $Z\delta m = Z(m_{\text{bare}} - m_\nu) \neq 0$. With $|f(m_0)|^2 = 1$ from (3) and (4) we see that Z diminishes also when $m_v \to m_\theta$. Let us take the situation $m_v = m_\theta$. Then (4) remains well defined, $h'(m_\nu) = \infty$, $Z = 0$, $g^2 = 0$, $Z\,\delta\,m = 0$. From (2) and (4) we get a non trivial δ with

$$\delta(m_\theta) - \delta(\infty) = -\frac{\pi}{2}, \quad g_0^2 \text{ finite},$$

$$\delta(m_\theta) - \delta(\infty) = \frac{\pi}{2}, \quad g_0^2 = +\infty.$$

Thus, we have found a case with $Z = 0$, and Levinson's theorem takes the form (1), but only when also $g_0^2 \to \infty$, $N_e = 0$. From some technical points of view we remark that $S(m_\theta) = -1$, meaning that δ_t is an integral multiple of $\pi/2$, and when in proofs we use the

$$\text{``}\frac{1}{2\pi i}\int \frac{D'(z)}{D(z)}\,dz = \text{integer}\text{ ''}$$

method, the infinitesimally small semicircle just at the end of the cut (e.g. in (1)) is of vital importance.

For the Lee model without cut off, i.e. when a ghost is also present, a composite particle appears without finding some $Z = 0$ criteria [4]. Actually both of the Z_i ($i = 1$ for a normal particle, $i = 2$ for a ghost) are infinite. In passing we mention that the det $Z = 0$ criteria [5] which serves for establishing the situation in which one of the two particles with identical quantum numbers is elementary and the other is composite is based on the supposition that from the field theoretical point of view we have two field operators, which is clearly not so in our case.

A limit similar to $g_0^2 \to +\infty$, which could eliminate the elementary particle does not exist, only the ratio of Z_1/Z_2 which is always finite, can be made equal to zero with $m_1 = m_\theta$. Then

$$\delta(m_\theta) - \delta(\infty) = \frac{\pi}{2}.$$

REFERENCES

1. K. Hayashi, M. Hirayama, T. Muta, N. Seto and T. Shirafuji, Fortschr. d. Phys., **15** 625, 1967.
2. M. T. Vaughan, R. Aaron and R. D. Amado, Phys. Rev., **124,** 1258, 1961.
3. N. Levinson, Kgl. Danske Videnskab. Selskab. Mat. Phys. Medd., No. 9, 1949.
4. K. L. Nagy, Acta Phys. Hung., **24**, 433, 1968.
5. K. Kang, Phys. Rev., **152**, 1234, 1966.

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SURFACE RECOMBINATION STUDIES OF GERMANIUM BY SUHL EFFECT APPLYING LIGHT INJECTION

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(Received 21. III. 1968)

Surface recombination on germanium was studied applying the Suhl effect [1, 2] with the modification of using minority carrier injection by light instead of by a probe.

The scheme of the arrangement used for experiments is shown in Fig. 1. A part of the sample was illuminated, the remainder was shielded, and the probe was applied to this dark region at a point about 0,8 mm from the edge of the illuminated area. The light was interrupted at 44 cps by a rotating disc. The photovoltage was measured by a selective measuring receiver. In the experiments, made at room temperature, germanium samples having ohmic contacts were used ($\varrho \sim 20\ \Omega$ cm, $\tau = 550\ \mu$sec). The values of surface recombination velocity were checked by H_2O_2 and CP-4 etching, and by mechanical polishing. The d.c. field was adjusted for each value of the magnetic field because of the magnetoresistance effect. The change in the resistance due to the illumination was about 1%. The peak in the photovoltage vs. magnetic field is independent from the d.c. field in a sufficiently wide interval. The d.c. was chosen to be 10 mA (lower d.c. would produce a low photovoltage, while higher d.c. would cause undesirable heating of the sample).

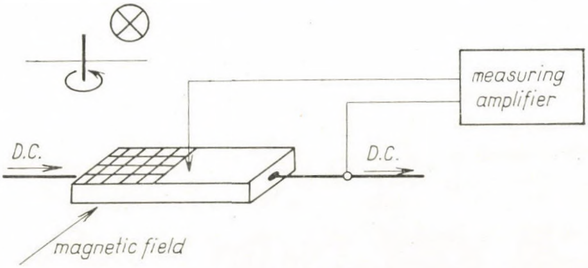

Fig. 1

We observed the shift of the peak by changing the surface recombination velocity. The effect was measured on an etched sample, using H_2O_2. Then the sample was left between the poles of the magnet without lifting the probe.

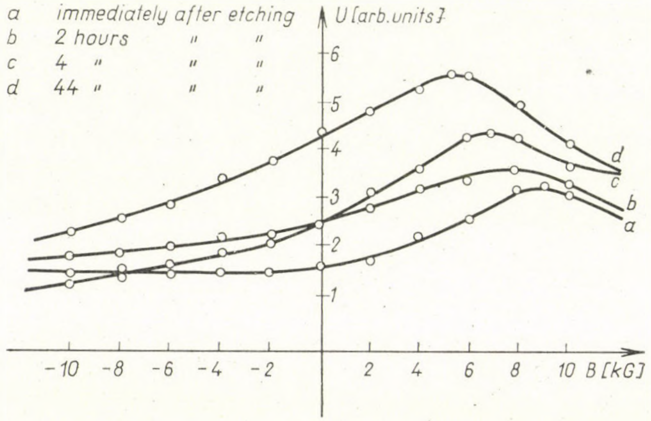

Fig. 2

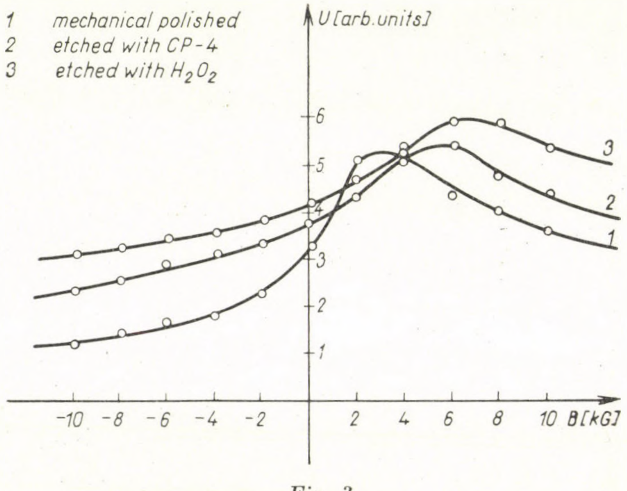

Fig. 3

After 2, 4, and 44 hours the measurements were repeated. The results are shown in Fig. 2 (the measurements were performed also using a water-filter, and germanium-filters. The position of the peak did not change, but the photovoltage decreased).

It is known that the surface recombination velocity in germanium increases in an air ambient [3, 4], because an oxide forms on the surface after etching. At higher surface recombination velocities the carriers recombine more rapidly on the surface. Therefore, the maximum value can be obtained with a lower magnetic field.

The SUHL effect measurements were performed on a sample 3 hours after a refreshing etching, followed by a measurement on the sample treated with 20% $K_2Cr_2O_7$ and dried. The oxidation of the surface was increased by the $K_2Cr_2O_7$ treatment, and the peak shifted in a direction identical to that observed previously. The photovoltages for surfaces treated by various methods are shown in Fig. 3.

The numerical estimation of the peak position based on the fundamental equations of the photoelectromagnetic (PME) effect [5] agreed qualitatively with the shifts obtained experimentally for several surface recombination velocities and magnetic field values. The differential equation of the effect could, however, not be solved exactly owing to the existing complex boundary conditions.

A change in the surface recombination velocity of about one order of magnitude could be found. The surface recombination velocity can easily be determined by calibration of peak position with samples of known surface recombination velocities. The measurements were well reproducible contrary to those made by point contact injection.

The author is indebted to G. PATAKI for suggesting the subject and for valuable discussions, and to F. BELEZNAY for making the computer programme.

LITERATURE

1. H. SUHL and W. SHOCKLEY, Phys. Rev., **75**, 1617, 1949.
2. H. SUHL and W. SHOCKLEY, Phys. Rev., **76**, 180, 1949.
3. CHROBOCZEK et al,. Phys. Stat. Sol., **6**, K137, 1964.
4. U. HARTEEN, Phil. Res. Repts., **14**, 346, 1959.
5. W. ROOSBROECK, Phys. Rev., **101**, 1713, 1956.

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ON THE FREQUENCY DEPENDENCE OF ZnS ELECTROLUMINESCENCE SPECTRA

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It is well known that in the electroluminescence spectra of ZnS : Cu two main emission bands with maxima near 2.35 and 2.62 eV can be observed. The same emission bands have been found with other excitations as well [1, 2]. Recently the atomic structure of the Cu green centre has been investigated extensively [3, 4] and it has been established that this centre is a sort of associated centrum where pairs of different associations take part in the emission process resulting in a spectral shift in the luminescence afterglow and with a change in the excitation intensity. Fischer and co-workers [5] have stated in their review on the luminescence of solids that the spectrum of an electroluminophor shows a gradual shift of emission maximum with increasing frequency from 2.38 eV to 2.5 eV. A similar, although smaller shift was observed by Kawashima [6]. These shifts could be regarded as the counterpart of the effect found by Shionoya [4].

A gradual shift of the maximum of the green Cu band with frequency would mean — according to the explanation of the shift found in the photoluminescence decay — that in the second half-period of the delayed recombination the carriers are available in large quantities and the emission is determined by the recombination rate and not, for instance, by the detrapping of the carriers excited in the previous half cycle. However, it must be considered that a small contribution of the blue emission band (getting stronger at high frequencies) can shift the green maximum.

The two effects, a real shift of the maximum and an apparent one owing to the contribution of a second band, can be distinguished by a thorough investigation of the band shapes.

Fig. 1 shows the spectral energy distribution of a ZnS: Cu,Cl electroluminescence cell at different frequencies. Between 20 c/s (curve 1) and 200 c/s (curve 2) a wavelength shift seems to occur. At higher frequencies (curve 3: 500 c/s and curve 4: 2500 c/s) the gradual build-up of the blue band is apparent. It is an old practice to find the band maxima of secondary bands by the help of band analysis supposing the band shape to be — for instance — a Gaussian one [7]. It was ascertained that the 20 c/s curve can be well approximated by a Gauss curve. Now, we supposed that at the low energy half-width of this band (2.15 eV) the emission of the 200 c/s electroluminescence also originates only from this same band. By fitting a Gauss curve corresponding to the 20 c/s distribution to the 200 c/s curve and subtracting it from the original one, curve 5 (Fig. 1) was derived. This curve corresponds well to the ZnS : Cu blue band. Fig. 2 shows the spectral distribution of the green band in the "Treptow representation" (curve 1). In the same Figure the points of

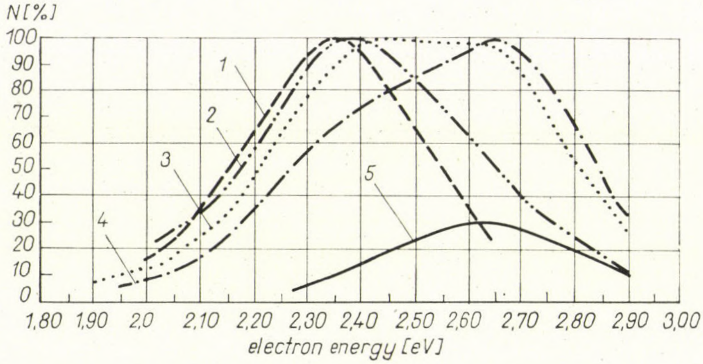

Fig. 1. Spectral energy distribution of a ZnS : Cu, Cl electroluminescence cell. Curve 1: 20 c/s Curve 2: 200 c/s; Curve 3: 500 c/s; Curve 4: 2500 c/s; Curve 5: blue band, calculated

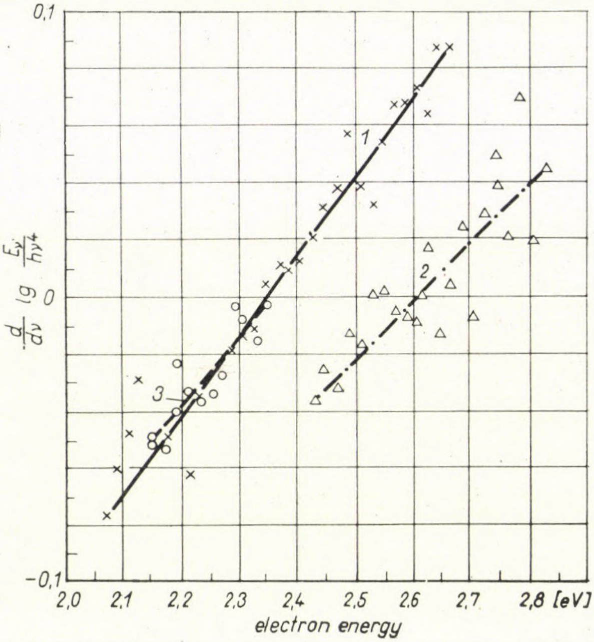

Fig. 2. Spectral distribution of the green (curves 1 and 3) and blue (curve 2) band in Gauss-curve representation

the derived 200 c/s blue band (curve 2) and the 2500 c/s green band, derived by the same method as well as the corresponding straight lines calculated by the least squares method have been plotted. The calculated band maximum of the "200 c/s" blue band lies at 2.615 eV, that of the "2500 c/s" green one at 2.365 eV, both of which correspond well with the positions of the maxima found in phosphors where that band is predominant (2.62 eV and 2.35 eV, resp.). Similar results were obtained by starting from spectra measured at other frequencies.

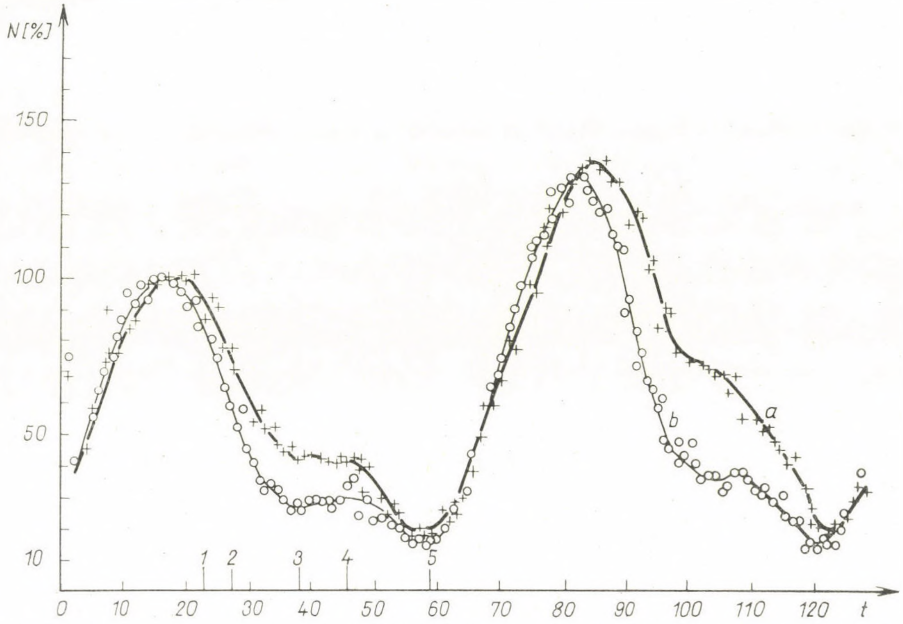

Fig. 3. Brightness-waves measured at 2.30 eV (curve a) and 2.60 eV (curve b). Points 1—5 define time marks (see Fig. 4)

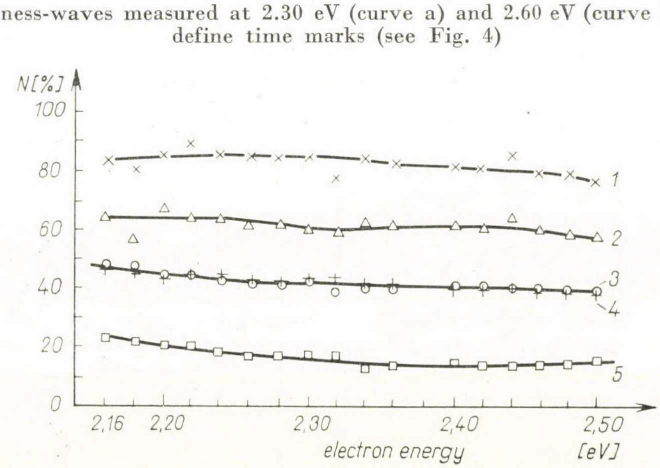

Fig. 4. Five relative spectral energy distribution curves (see time marks in Fig. 3), normalised to the first maximum of the brightness wave

The measurements prove that the spectral shift observed between 20 c/s and 200 c/s is only a virtual one. Further on, even at 2.500 c/s, only a very slight displacement occurs, if any. Thus, no counterpart of the effect observed by SHIONOYA in photoluminescence could be observed in the electroluminescence measurements, although the frequency dependence of electroluminescence emission shows in many respects, especially in the counteraction of the green and blue bands, an analogy with the intensity dependence of photoluminescence.

As the spectral distribution of the average electroluminescence is mainly due to the emission of the primary maximum, the spectral changes during the brightness-wave have also been investigated. Fig. 3 shows the brightness-wave at 2.30 eV (curve a) and 2.60 eV (curve b). The phase-shift between the green and blue band is apparent from this Figure. A probable occurrence of a spectral shift of the green band has been checked in the following way: Spectral brightness-waves were measured between 2.16 and 2.60 eV, and normalized at the first primary maximum. This normalized spectral distribution (showing only the time-dependent changes of the spectra) has been plotted in Fig. 4 at 5 different phase-angles, corresponding to the time marks 1—5 in Fig. 3. No shift — higher than the statistical error of measurement — is observed during the afterglow of the primary as well as the secondary maximum.

Summarizing our results, it can be concluded that neither the green nor the blue electroluminescence emission band of ZnS : Cu shows a frequency-dependent wavelength shift. Postulating a recombination mechanism of the green band taking place via "waiting states" of an associated centrum the rate determining factor of recombination is not the recombination process itself.

The author is indebted to Prof. G. SZIGETI for continuous interest and to Miss M. POWER for performing the brightness-wave measurements.

REFERENCES

1. L. BALÁZS, J. PUSKÁS, J. WEISZBURG and J. SCHANDA, Festkörp. phys. Balatonfüred, 1959. Akad. Verl. 1961, pp. 317—22.
2. R. DITTMANN, H. GOERSCH and D. HAHN, ICOL 10/31, 1966.
3. S. SHIONOYA and T. KODA, J. Phys. Soc. Jap., **19**, 1157, 1964.
4. S. SHIONOYA, K. ERA and Y. WASHIZAWA, ICOL 6/112, 1966.
5. A. G. FISCHER, R. E. SHRADER and S. LARACH, Luminescence of Solids, in Photoelectronic Materials and Devices, ed.: S. Larach, Van Nostrand, Princeton, 1965.
6. S. KAWASHIMA, Jap. J. Appl. Phys., **5**, 1161, 1966.
7. GY. SZIGETI, MTA Mat. Term. tud. Oszt. Közl., **1**, 30, 1950.
8. H. TREPTOW, phys. stat. sol., **6**, 555, 1964.

RECENSIONES

DAVID PARK: Introduction to Strong Interactions

(A Lecture-Note Volume)
W. A. Benjamin, Inc. New York, Amsterdam, 1966. XI chapters, 250 pages

Parallel with the substantial increase of particle physics research the literature on this subject acquired dimensions rendering almost impossible to keep each publication in evidence, not to speak of the unattainable objective of studying them thoroughly. This is the reason why the intention of the Publishers to commence a low-priced series for beginning researchers is most welcome. The series discusses the newest research methods and results pertaining to this subject in introductory volumes written in an unsophisticated style, filling up thus the gap which existed so far between university studies and monographs on recent achievements. This series undoubtedly offers a great assistance to young, would-be researchers.

DAVID PARK was successful in realizing the aims of the Publishers in his work of 250 pages on strong interactions. The discussion of the most modern methods of this branch of physics is based on the assumed knowledge of the material of university lectures. Emphasis is laid on teaching the techniques how to treat theoretically experimental data through a simplified model. Models reducing problems to their essence help understanding and are of great value in more intricate discussions of actual problems. At the end of each chapter a collection of questions is included to foster independent thinking and to awake creativeness. Compilation and sequence of the questions prove the gift of teaching and excellent expert knowledge of the author. The rich and well-selected bibliography attached to each chapter is also of assistance in finding one's way among monographs dealing with questions of detail.

The volume is divided into the following chapters:

1. Introduction, 2. Quantized Fields, 3. Interactions, 4. Potential Scattering, Bound States and Resonances, 5. Formal Scattering Theory, 6. Relativistic Scattering Amplitudes, 7. Calculation of Scattering Amplitudes, 8. Dispersion Relations, 9. Invariance and Conservation Laws, 10. Symmetries of Strong Interactions, 11. The Eightfold Way.

This excellent introductory work may be recommended in good faith to researchers wishing to study particle physics.

K. NAGY

GEOFFROY F. CHEW: The Analytic S Matrix

(A Basis for Nuclear Democracy)
W. A. Benjamin, Inc. New York and Amsterdam, 1966; XIV chapters, 100 pages

The book discusses systematically the dynamics of strongly interacting particles on basis of the *S*-matrix. Its most important part deals with the maximal analyticity of the second degree being equivalent with nuclear democracy and constituting the basis for bootstrap dynamics. The lecture of the book requires a knowledge of the principles of non-relativistic quantum mechanics (including scattering theory) and of the Lorentz-group. Such requirements, however, do not exist in relation to the quantum field theory; the author even states in the Preface: "Indeed, as pointed out in the preface to my 1961 lecture notes, lengthy experience with Lagrangian field theory appears to constitute a disadvantage when attempting to learn *S*-matrix theory".

The volume is divided into the following chapters:

1. Analyticity as a Fundamental Principle in Physics, 2. General *S*-matrix Principles, Excluding Unitarity, 3. Unitarity and Discontinuities, 4. Maximal Analyticity of the First

Degree: Landau Singularities, 5. The Four-Line Connected Part with One Channel Invariant Fixed, 6. Angular Momentum Decomposition, 7. Analytic Continuation in Angular Momentum and Asymptotic Behavior, 8. Maximal Analyticity of the Second Degree, Bootstrap Dynamics, 9. The Nonrelativistic Potential Scattering Model, 10. The Strip Model of the Four-Line Connected Part, 11. Dynamic Equations for the Strip Model, 12. Regge Pole Approximation to the Strip Model, 13. The Nuclear Bootstrap: General Discussion, Continued, 14. Conclusion.

K. NAGY

Particle Interactions at High Energies

(Scottish Universities' Summer School, 1966)
Edited by
T. W. PREIST and L. L. J. VICK, Oliver and Boyd, Edinburgh and London,
VIII chapters, 405 pages, 1967

The seventh Summer School of the Scottish Universities was held in August 1966. Subjects of the lectures embraced the most up-to-date questions of high energy physics.

Lectures were delivered and discussion conducted by distinguished physicists. The volume prints the lectures given at the Summer School including the newest achievements of this field of science in a more complete and detailed form than scientific publications usually do.

Titles of the different chapters are as follows: 1. Properties of Scattering Amplitudes at High Energy (Dr. R. J. EDEN), 2. Very High Energy Scattering of Strongly Interacting Particles (Prof. L. VAN HOVE), 3. Some Features of Electromagnetic Interactions (Prof. R. WILSON), 4. Precision Test of Quantum Electrodynamics at Low Energies (Prof. S. D. DRELL), 5. The Symmetries of the Weak Interaction (Dr. P. K. KABIR), 6. The Algebra Associated with the Vector and Axial-Vector Current (Prof. L. A. RADICATI), 7. Pion-Nucleon Phase Shift Analysis (Dr. A. DONNACHIE), 8. Quark Models (Prof. E. J. SQUIRES).

Director of the School was Prof. N. KEMMER who also prefaced the Volume, while Dr. T. W. PREIST and Dr. L. L. J. VICK acted as editors.

K. NAGY

PÁL GOMBÁS: **Pseudopotentiale**

Springer Verlag, Wien, New York, 1967, pp. 186

The laws of motion of atomic particles have been formulated in quantum theory in a mathematically elegant and exact way, by means of the operator calculus. However, the actual solution of the non-linear operator equations of quantum field theory or the Schrödinger equation of the many dimensional configuration space presents almost insurmountable difficulties.

The behaviour of a complex system can most easily be grasped by the physicist if he can pick out individual atomic particles one at a time and describe their three dimensional motion by means of forces or potentials. Force and potential energy are not exact concepts; modern physics repeatedly pointed out the approximate nature of these mechanistic conceptions. In actual calculations in classical and quantum mechanics, however, the relation of a micro-particle to its neighbourhood is almost always described in terms of these concepts, the many-body problem is thus reduced to a one-body problem.

This new monograph by Professor GOMBÁS gives a survey of the problem fundamental from the point of view of the application of modern physics: How is it possible to reduce the non-relativistic Schrödinger equation (restricted with the Pauli principle) in configuration space to the Schrödinger equation of a single particle in which the effect of the neighbourhood is described by potential energy-like terms, the pseudopotentials. This problem is not at all trivial. The wave function, which is symmetrical or antisymmetrical, depends on the coordinates of all particles. This means that the behaviour of the particle under considerations is influenced not only by its own position with respect to the complex system but also by the simultaneous position of the other particles. There is correlation in the motion of the particles and these are just the quantum phenomena, originating in the operator algebra, that mostly make the understanding of atomic motion difficult. Professor GOMBÁS shows in this book that for a single electron all this can be taken into account by means of an exchange, an exclusion and a correlation potential in a surprisingly good approximation. As a result, the gain can hardly be overstimated: The manybody problem, difficult to handle even by computer, is simplified to a differential equation soluble by conventional methods and even more important: pseudopotentials help to develop the ability of the physicist to visualise the behaviour of particles. The loss is rather insignificant: The author shows on the example of the heavy atoms that the behaviour of electrons is described by pseudopotentials to an accuracy of 1—2% which is an astonishingly good result.

The book is made especially interesting by the fact that the method of reducing the many-body problem to a one-body problem corrected with pseudopotentials has developed into an efficient approximation method at the hands of Professor GOMBÁS and his school. The method has been initiated independently by him and HELLMANN and was afterwards developed by GOMBÁS into a method working with a 1% precision in which also some results of HARTREE, FOCK, SLATER and WIGNER have been used. We also see with pleasure the priority of GOMBÁS and his coworkers documented concerning some results which some people prefer these days to associate with other names.

While in his other well-known books Professor GOMBÁS studies the behaviour of the quantum system as a whole, this new book complements these by studying, as it does, the behaviour of individual particles putting an emphasis on the one-electron wave function and the calculation of the energy levels. Without the knowledge of these a number of phenomena could not be understood.

Even the reader well versed with quantum-mechanics discovers with excitement how much of the possibilities given by the many-dimensional configuration space can be condensed into potential-like expressions. The exclusion and the exchange potentials have already been with us for some time. Especially interesting is the introduction of the correlation potential; an improvement on the Hartree-Fock approximation is indispensable today. We can only regret that the author gave so relatively little space, for the sake of simplicity of presentation, to this problem.

The author applies his method to atomic electrons. I am quite sure that pseudopotentials incorporating the influence of other delocalized electrons which are also easily treated by computer will play a major role in future developments of quantum-chemistry and molecular biology. I am sure the author had in mind the needs of these future developments when he decided to publish this clear, concisely presented monograph.

G. MARX

C. H. BAILEY: The Electromagnetic Spectrum and Sound
(50 modern experiments)

Pergamon Press, Oxford 1967. pp IX + 135, 88 figs., price 25 s.

The laboratory experiments detailed in this book can be connected with the subject matter of instruction in physics of the two upper classes of secondary schools in Hungary. Their level too is similar, though it sometimes is higher than that of the Hungarian subject matter. The author deserves special appreciation for applying the most up-to-date technological means (transitions, clystrons, etc.) in his experiments and for linking up in his book vibration phenomena over the whole field of physics.

The six parts of the book are: I, Sound; II, Light; III, Infrared; IV, Ultraviolet; V, Radio, VI, 3 cm-Radio (microwaves). In all parts stress in laid primarily on measuring wavelengths but the measurement of speed (in sound) and absorption (in light) is also taken into consideration. In most cases even setting up the measuring equipment is left to the student by a precise enumeration of the parts. Much practical advice is given, too (e.g. soldering transistors). It is peculiar, that beside the really modern concept of all other chapters, that dealing with acoustics is somewhat classical. For example, not a single experiment concerning ultrasonics can be found among exercises.

The subject matter and lay-out of this book make it very useful for teachers leading study circles in physics.

T. TARNÓCZY

G. v. Békésy: Sensory Inhibition

Princeton University Press, Princeton 1967, X + 265 pp, 188 figs., price $ 8.50

This new book by Békésy includes the material of his 1965 Langfeld Memorial Lectures at Princeton University. Here, the author gives a summary of those of his results which can be connected with the extraordinarily complicated group of problems of sensory inhibition. For the sake of clarity should be mentioned that Békésy — as he states in his preface — has recently discovered a general similarity in inhibition processes occurring in various sense organs.

His experiments which must be considered as important landmarks in modern biophysics refer mainly to the funneling of wide range simultaneous stimuli as well as the mechanism of sensory localization. Sensory perception of the skin seems to be investigated most thoroughly, but the most particular experimental techniques are applied in the investigation of taste perception. In the Spring of 1965 the reviewer had the opportunity of seeing the apparatus — although dismantled — at Harvard University for the investigation of taste. The premixing of water and solution reaching the tongue can be continuously altered, while the change of concentration is recorded as a function of time by an automatic recorder. The concentration of fluid on the surface of the tongue was checked by the change of electrical resistance or by observing the colour change by means of fiber-optics. In order to eliminate errors due to fluid eddies the peristaltic pumps supplying the mixing fluid are fixed on a concrete block floating on compressed air. The whole equipment is capable of indicating a change of stimuli with an accuracy of 1 ms.

The most important discovery of Békésy since winning the Nobel-Prize (1961) is in connection with the ability to localize and separate taste stimuli. He has proved that two stimuli occurring on two points of the tongue with a time delay of 1—2 ms cause a directional sensation. This magnitude of time delay is also valid for smelling, hearing and vibration sensation. At the same time, time constants for the sensation of amplitude or intensity lie between 200 and 1000 ms for all sense organs. Thus, two components of neural activity with different times the ratio of which is at least 1 : 200 can be distinguished. By the discovery of short time neural activity our complete modern conception on the biophysics of sense organs may be reformulated.

This small book contains so much valuable material, raises so many new ideas, introduces such a great number of connections, and initiates so many new ideas, that we can only be grateful to all those (above all Professor E. G. Wever) who have made it possible for these lectures to be made available to the public.

The chapters of the book are: I. Adaptation and inhibition as a means of suppressing an excess of information. II. The inhibition of simultaneous stimuli. III. Inhibition as a result of time delay. IV. Stimulus localization as a method of investigating neural activity. V. Funnelling and inhibition in hearing. VI. The role of inhibition in various fields of sensory perception.

The book is completed by a detailed bibliography. Hereby most of minor errors of the bibliography in Experiments in Hearing are corrected so that the 122 works mentioned up to 1966 cover almost the whole literary output of the author.

With pleasure we acknowledge that Békésy makes appreciative mention of his working places in Hungary, the Post-Research Institute, the Physical Institute of Budapest University and the Széchenyi Foundation, which, while supporting his activities for long years "never wanted him to give a progress report".

T. Tarnóczy

E. Zwicker and R. Feldtkeller: Das Ohr als Nachrichtenempfänger

II. Auflage, Hirzel Vlg., Stuttgart 1967. XVI + 232 S, 216 Abb.

In durchgearbeiteter und in grossem Masse erweiterter Form ist das auch bei uns wohlbekannte Werk 11 Jahre nach der ersten Auflage wieder erschienen. Das Buch kann sogar als Neuerscheinung betrachtet werden, was dadurch gerechtfertigt ist, dass obwohl der Titel derselbe geblieben ist, die Reihenfolge der Namen der Verfasser umgekehrt wurde.

Die Kapitel sind die Folgenden: I. Schallschwingungen, II. Schallfelder, III. Reiz und Empfindung, IV. Hörschwelle und Mithörschwellen von Dauerschall, V. Eben wahrnehmbare Frequenzänderungen, VI. Frequenzgruppen, VII. Skalen der Tonhöhe, VIII. Eben wahrnehmbare Schallpegeländerungen, IX. Erregung, X. Skalen der Schallstärke, XI. Funktionsschema für eben wahrnehmbare langsame Schalländerungen, XII. Hörschwelle und Mithörschwelle von Impulsen, XIII. Schwellen für schnelle Schalländerungen, XIV. Eben wahrnehmbare Phasenänderungen, XV. Tonhöhenunschärfe, XVI. Lautstärke von Impulsen, XVII. Berechnung und Messung der Lautheit, XVIII. Wirkung von Störimpulsanfang und Störimpulsende auf die Mithörschwelle, XIX. Knacke und XX. Nichtlinearität des Gehörs.

Gegenüber der ersten Ausgabe können folgende, bedeutende Unterschiede vermerkt werden. Der Umfang des Buches hat sich um das zweieinhalb-fache erweitert. Die Gliederung geht vielmehr auf Einzelheiten ein: das Werk besteht gegenüber den vorherigen fünf, aus 20 Kapiteln. Die eigentliche Behandlung des Gegenstandes beruht hierbei auf einer viel ausführlicheren Grundlage. Die Grundbegriffe der Kapitel I—III, VII. und X. machen das Buch auch für diejenige lesbar, die nicht über die notwendigen akustischen Grundkenntnisse verfügen. Selbstverständlich wurden die neuen Resultate der inzwischen vergangenen 10 Jahre auch in die neue Auflage aufgenommen. In diesem Zusammenhang sind besonders die Kapitel XIV., XVIII. und XIX. hervorzuheben, aber im wesentlichen enthält jeder einzelne der Kapitel XI. bis XX. etwas wesentlich Neues. Ausserdem sind in das Buch die 10 Schablonen der in der Zwischenzeit normalisierten ZWICKER'schen Lautheitsberechnungsmethode aufgenommen worden.

Der Aufbau, die Behandlungsweise und der Stil des Buches können nur mit Anerkennung erwähnt werden. Wir sind aber nicht einverstanden mit der Numerierung der Abbildungen innerhalb von einzelnen Paragraphen, weil dadurch das Nachschlagen sehr erschwert wird. Auch hätte diese, in jeder Hinsicht wertvolle Monographie eine ausführliche Bibliographie verdient. Dabei denken wir nicht an solche allgemein bekannte und umfassende Werke, die zu den ersten zwei Kapiteln erwähnt sind. Diese hätte man sogar teilweise weglassen, oder im allgemeinen bibliographischen Teil anführen können. Auf anderen Gebieten hingegen, wo vielleicht von historischem Standpunkt aus, oder zur besseren Übersicht über die in der Literatur vertretenen verschiedenen Meinungen und schliesslich zum gründlicheren Studium irgendeiner Frage eine Orientierung benötigt würde, bleibt man sozusagen auf sich selbst verlassen. Als Beispiel sei der Kapitel über die Lautstärke von Impulsen angeführt. Die drei Literaturangaben stammen aus 1959, 1963 und 1966. In der internationalen Literatur sind aber ähnliche Ergebnisse schon viel früher erreicht worden (GARNER, MISKOLCZY—FODOR u. a.) und diese werden selbst in den angegebenen Referenzen nicht erwähnt.

Das neue Werk von ZWICKER—FELDTKELLER bedeutet eine wertvolle Hilfe in der Arbeit von Akustikern und Nachrichtentechnikern. Unserer Meinung nach, kann es auch ausserhalb des deutschen Sprachgebietes auf einen ausgedehnten Leserkreis rechnen.

T. TARNÓCZY

Proceedings of the 1966 Heat Transfer and Fluid Mechanics Institute

Edited by M. A. Saad and J. A. Miller. Published by Stanford University Press, 1966.

The Heat Transfer and Fluid Mechanics Institute was established to provide an opportunity for the presentation and discussion of current basic research in heat transfer and fluid mechanics, and related fields. The nineteenth meeting of the Heat Transfer and Fluid Mechanics Institute was sponsored jointly by the University of Santa Clara and the U.S. Naval Postgraduate School and was held at the University of Santa Clara at Santa Clara, California, from 22 to 24 June, 1966.

Five main sessions were planned in the following topics:

1. boundary layer flows including stability, non-newtonian flow, and shock-boundary layer interactions;
2. two-phase-flows and change of phase;
3. free and forced convective heat and mass transfer;
4. wakes and separated regions and radiation gas dynamics;
5. special problems, such as vortex flows, fluid motion in modified force fields, fluid dynamics of biological systems and heat transfer in constricted *arcs.*

In addition, the Institute presented a session of invited lectures on new and unusual areas related to fluid mechanics. One of the most interesting guest papers deals with a simplified procedure for opacity calculations in radiation gas dynamics, by S. S. **Penner** who is the co-author of the well known monograph entitled: "Radiative Transfer and Quantitative Spectroscopy Relating to Re-entry Phenomena" (Academic Press, New York, 1966). **Penner**, in his lecture, summarizes the type of opacity data that is customarily used in the solution of problems in radiation gas dynamics. He briefly describes the physical ideas involved in developing simplified relations for estimating Planck and Rosseland means for continuum radiation in plasmas, for line radiation in pure rotation spectra, in vibration-rotation band systems belonging to diatomic emitters, for line radiation in the spectra of polyatomic molecules and for bound-free transitions in electronic band spectra.

In conclusion the author comments on the present status of numerical computation programmes for estimating air opacities.

A large number of contributed papers gives theoretical models and solutions — applying the computer technique — for phenomena of heat transfer, supersonic flow and shock waves. For example, the phenomena of a shock wave propagating into a supersonic crossflow is described mathematically by **Tyler** and **Zumwalt** using a finite difference technique. In this case the transient shock discontinuity was modelled by an artificial "dissipative" scheme in the difference technique. The results of three special cases for the shock-crossflow interaction are presented, and show, that numerical schemes using artificial "dissipative" techniques seem to provide a means of solving more complex interaction phenomena.

The "Proceedings" of the Heat Transfer and Fluid Mechanics Institute contains 24 selected papers relating to the above topics. The "Proceedings" is a well known internationally recognised publication, characterizing the continuous striving for technical excellence of the selected papers.

J. F. Bitó

Proceedings of the 1967 Heat Transfer and Fluid Mechanics Institute

Edited by Paul A. Libby, Daniel B. Olfe, Charles W. van Atta
Published by Stanford University Press, 1967.

The twentieth meeting of the Heat Transfer and Fluid Mechanics Institute was held at the University of California at San Diego, La Jolla, California, on June 19, 20, 21, 1967.

New and basic advances in fluid mechanics, heat transfer, and related fields are contained in this collection of papers published by Stanford University Press. Roughly two-thirds of the papers presented deal with Post-Apollo re-entry phenomena and propulsion fluid mechanics and heat transfer. The papers are selected on the basis of their fundamental character and the extent to which they enlarge the understanding of heat transfer and fluid mechanics. Emphasis is given to topics which have broad implications and which, therefore, may not readily fall into any one of the conventional fields of aeronautical, chemical, civil, or mechanical engineering. Selection of papers is made by engineers and scientists who are engaged in related work and who are particularly qualified to judge fundamental research.

There were three main sessions: Post-Apollo re-entry phenomena, with eight papers; propulsion fluid mechanics and heat transfer, with seven papers; and vortices, wakes and boundary layers, with eight papers.

The meeting began with an invited paper by **Leonard Roberts**, Director of Mission Analysis of NASA, entitled "Atmospheric Entry in the Post-Apollo Era". In this lecture a summary was made of possible future mission requirements in the Post-Apollo period, particularly as they relate to the entry of vehicles into planetary atmospheres. The missions considered involve unmanned probes that enter the atmosphere of Mars, Venus and Jupiter, manned vehicles that aerobrake into orbit about Mars and Venus, or land on the surface of Mars, and manned vehicles that return and enter the Earth's atmosphere at high speed. Such manned vehicles may require lifting and manoeuvering capabilities greater than those used in the past and may lead to configurations that differ from the current family of semi-ballistic vehicles. The various problems in aerothermodynamics that must be solved in order to permit the optimum design of these advanced vehicles were discussed and conclusions drawn regarding the need for improvement in our understanding of entry phenomena, particularly in the interaction of the configuration and its thermal protection system with the flow field in which it is immersed. The most interesting contributed paper dealt with the aerothermodynamics of the Apollo experience (by R. B. ERB et al.). In this paper a complete thermal protection system is constructed theoretically by the authors. The verification of these results was obtained on Scout, Flight Investigation of Re-entry Environment (FIRE), and Pacemaker missions.

The greatest part of the published papers contain information on the work performed under contracts with NASA.

J. F. BITÓ

A. I. AKHIEZER, I. A. AKHIEZER, R. V. POLOVIN, A. G. SITENKO and K. N. STEPANOV: **Collective Oscillations in a Plasma**

(International Series of Monographs in Natural Philosophy, Vol. 7), p. X+190,
translated from the original in Russian by R. J. Taylor,
Pergamon Press, Oxford etc., 1967, Price: 45 s

The book covers the theory of linear oscillations in a collisionless plasma, i.e. in a plasma in which binary collisions have no significant effect on its oscillatory properties. The book deals with the following basic problems: the spectra of natural oscillations, the stability and instability of various particle distributions, and fluctuations in a homogeneous plasma.

The authors are experts in this field and have contributed greatly with their research to its development. In this valuable book they deal with many aspects of physical and technical problems. The study of collective oscillations in a plasma is of great interest in the propagation of radio-waves in the ionosphere, the radio emission of stars, the amplification and generation of micro-waves in a plasma, plasma diagnostics, etc.

This excellent book is warmly recommended to the specialist as well as to the young researcher who wants to enter this field of study.

P. GOMBÁS

D. A. KIRZHNITS: **Field Theoretical Methods in Many-Body Systems**

(International Series of Monographs in Natural Philosophy, Vol. 8) XVI+394, translated from
the original Russian by A. T. Meadows,
Pergamon Press, Oxford etc., 1967. Price £ 5. 10 s

Dies Buch gibt eine sehr gut gelungene Einführung in die Anwendung moderner feldtheoretischer Methoden auf Mehrteilchensysteme. Der Autor der dieses Gebiet durch mehrere bedeutende Arbeiten bereichert hat, hat mit diesem Buch ein sehr nützliches Werk geschaffen, das vielen Physikern, die mit den feldtheoretischen Methoden nicht ganz vertraut sind, ein Vordringen in dieses Gebiet sehr erleichtern wird.

Nach einer kurzen Zusammenfassung der Grundlagen, wird die Hartree—Focksche und die statistische Thomas—Fermische Methode besprochen und insbesondere auf Materie unter hohem Druck und auf Atomkerne angewendet. Danach folgt eine ausführliche Darstellung der Störungsrechnung mit Anwendungen. In den darauf folgenden beiden Kapiteln behandelt der Verfasser die Methode der Green-Funktion in der Quantenmechanik und in der Quantenstatistik.

Man kann dem Verfasser zu der klaren und übersichtlichen Darstellung dieses Gebietes bestens beglückwünschen; dem Buch möchten wir besten Erfolg wünschen.

P. GOMBÁS

ENRICO FERMI: **Molecules, Crystals and Quantumstatistics**

übersetzt aus dem italienischen von M. Ferro-Luzzi, herausgegeben von L. Motz,
W. A. Benjamin, Inc., New York, Amsterdam, XIV+300, 1966. Preis: $ 13,50

Obwohl dieses Buch in 1934 geschrieben wurde und die Forschung auf den behandelten Gebieten sehr grosse Fortschritte gemacht hat, besitzt dieses Buch von FERMI einen ganz besonderen Reiz. Es legt nämlich die Gedankweise eines der grössten Physikers unseres Zeitalters dar, der nicht nur als Forscher Überragendes geleistet hat, sondern auch die seltene didaktische Begabung besass, seinen Schülern die schwierigsten Probleme restlos verständig zu machen. Dies macht das Buch auch heute noch gerade so aktuell wie vor 33 Jahren.

Das Buch zergliedert sich in 3 Teile. Im ersten werden die chemische Bindung von Molekülen, die Spektren zweiatomiger Moleküle, thermische Eigenschaften und mehratomige Moleküle behandelt. Der zweite Teil befasst sich mit Kristallgittern, mit deren Geometrie, sowie den physikalischen Eigenschaften. So unter anderen mit der spezifischen Wärme, Gitterschwingungen, Raman Effekt in Kristallen, Wärmeausdehnung, Theorie der Ionenkristalle, elektromagnetische Wellen in Kristallen, optische Eigenschaften von Kristallen. Im dritten Teil gibt FERMI einen Überblick über die statistische Mechanik und deren Anwendungen. Im Anschluss hieran werden die Quantenstatistiken begründet und hergeleitet. Am Ende der einzelnen Kapitel wurden vom Herausgeber in Anhängen einige neuste Forschungsergebnisse hinzugefügt.

Die englische Übersetzung eröffnet dem Buch einen grossen Leserkreis, der durch dieses Buch mit dem Genius dieses grossen Physikers in Kontakt gebracht wird, was besonders für die heranwachsende junge Physikergeneration von unschätzbarem Wert ist.

P. GOMBÁS

D. H. MARTIN: Magnetism in Solids

452, London Illife Books Ltd., London, 1967. Price: 120 s

This book is divided into the following chapters: Chapter 1: The characteristic properties of magnetically ordered solids. Chapter 2: The origins of magnetism in atoms and ions. Chapter 3: Diamagnetism and paramagnetism in solids. Chapter 4: The molecular field models of ordered magnetic solids. Chapter 5: Exchange interactions between two electrons. Chapter 6: Exchange interactions in solids. Chapter 7: The excited states and the statistical mechanics of ordered magnetic solids.

The problem of the field covered and the aim of the book are best explained by the author in the Preface:

"Magnetism in solids is a subject which continues to demand attention because of its technological importance and also it challenges the ingenuity of both experimental and theoretical physicists. Several recently developed experimental techniques have found their most fruitful application in magnetism. They have revealed a far greater variety of magnetic structure in solids than could have been anticipated a few years ago and detailed and subtle information can now be obtained about the structures of selected materials. In searching for satisfactory microscopic theories for this wide spectrum of magnetic behaviour the theoretician becomes involved in several of the most fundamental and difficult problems posed by the solid state — among them, for example, are critical point phenomena and correlations in the motions of electrons. While answers to many basic questions are still wanting the subject has been remarkably well served by the interpretive frameworks provided by phenomenological models, from the simple molecular-field theories to more sophisticated calculations based on the Heisenberg Hamiltonian.

All this leaves a confusing picture for anybody coming to the subject for the first time or wishing to develop or maintain a broad interest in magnetism while working in a specialised branch. This is the difficulty I had in mind when writing this book. In it I have aimed, not to teach techniques, whether experimental or theoretical, but to present a broad account of the subject which takes the discussion of major topics to the points of current investigation. A researcher in a particular field may need to acquire skills in special techniques to facilitate the solution of problems in that field but the essential aims and difficulties can normally be appreciated in more widely familiar theoretical terms and this I have attempted to do. There is frequently a wide gap between the generalities of the opening paragraphs of research papers and the technicalities which normally abruptly follow. It is hoped that this book will help to bridge such gaps."

In my opinion the author has fully accomplished his aims. A particular value of the book is that the exposition is clear and always easy to follow. It can be warmly be recommended to everybody who is interested in the magnetic behaviour of solids.

P. GOMBÁS

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QUENCHING OF FLUORESCENCE OF EOSIN IN SOLUTIONS

By

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Absorption and fluorescence spectra and the relative quantum yield of fluorescence of solutions of eosin-blue in various solvents have been studied and the data interpreted in terms of the non-radiative processes occurring in the solution, dimer formation and the dipole moment of eosin. The results seem to suggest a lower value for the dipole moment of the eosin molecule in the excited state than in the ground state.

Experimental

The relative quantum yields have been measured by the null method using photoelectric cells [1]. The fluorescence spectra are recorded with a constant-deviation spectrograph and the intensities measured using a recording microphotometer. Absorption studies are carried out using a spectrophotometer and the transmittance recorded as a function of the wave-length. The concentrations of the solutions used are of the order of 10^{-5} g of eosin-blue/c.c. of the solvent, so that the results are not vitiated by self-absorption to any appreciable extent.

Introduction

Various theories have been put forward to explain the non-radiative transfer of excitation energy of molecules of dyestuffs in solutions and the consequent quenching of the fluorescence of these solutions [2—6]. The effect of the viscosity of the solvent on the quantum yield of fluorescence has been investigated by various authors [7, 8] and the experimental results sought to be explained on the basis of the phenomena of collisional and statistical quenching, as suggested by VAVILOV [2] and PERIN [3]. In discussing these results, the effect of the interaction of the solvent and the solute molecules, affecting the term-values of the latter, is not taken into consideration. This solvent-effect should manifest itself not only in the quantum yield of fluorescence, as shown by BOWEN et al. [6], but also in the spectral nature of the absorption and the fluorescence of the solutions of the dye-stuff in different solvents. Therefore, a study of these — the absorption and the fluorescence spectra and the quantum yield of fluorescence in different solvents — may be expected to throw light on the mechanism of the interaction of the solvent with the molecules of the dye-stuff which affects the term-values and the energy-transitions in the molecules.

The positions of the absorption and the fluorescence maxima, $\bar{\nu}_a$ and $\bar{\nu}_f$, respectively, are related to the macroscopic properties of the solvent, such as its dielectric constant, D, and the refractive index, n [9, 10]. For polar molecules in less-polar solvents, the relationship between $\bar{\nu}_a$ and $\bar{\nu}_f$ and the solvent properties D and n can be expressed, to a first approximation [11] as

$$\bar{\nu}_a - \bar{\nu}_f \propto \left(\frac{D-1}{2\,D+1} - \frac{n^2-1}{2\,n^2+1}\right) = \Phi(D,n), \text{ say}. \qquad (1)$$

Marked deviations from the above relationship have been observed by **Zelinski** et al. [12] who showed that a better representation of the solvent-effect may be obtained in terms of the spectroscopic solvent scale, based on the idea of universal interactions developed by **Lippert** [13], **Kosower** [14] and **Zelinski** et al. [15, 16].

Results

Table 1 presents the experimental data for $\bar{\nu}_a$, $\bar{\nu}_f$ and the relative quantum yield Q_r of the fluorescence of solutions of eosin-blue in various solvents. Also included in the Table are the calculated values of $\Phi(D, n)$ and $(\bar{\nu}_a - \bar{\nu}_f)$.

Fig. 1 is a plot of $(\bar{\nu}_a - \bar{\nu}_f)$ against $\Phi(D, n)$. In Fig. 2, the relative quantum yields and the S values of the solvents on the spectroscopic solvent scale [12] have been plotted against $\bar{\nu}_f$.

Fig. 3 shows a plot of the **Kosower**'s Z values [14] against $\bar{\nu}_a$.

The solvent number in the Figures corresponds to that in Table 1.

Table 1

No.	Solvent	$\bar{\nu}_a$ cm^{-1}	$\bar{}_f$ cm^{-1}	$(\bar{\nu}_a - \bar{\nu}_f)$ cm^{-1}	$\Phi(D, n)$	Q_r
1	Water	19 680	17 760	1920	0.321	0.1
2	Ethanol	19 340	17 690	1650	0.291	0.42
3	Methanol	19 340	17 670	1670	0.306	0.40
4	Butanol	19 050	17 600	1450	0.267	0.37
5	Glycerol	19 460	17 630	1830	0.265	—
6	Acetone	18 800	17 510	1290	0.284	0.44
7	Pyridine	—	17 500	—	—	0.33

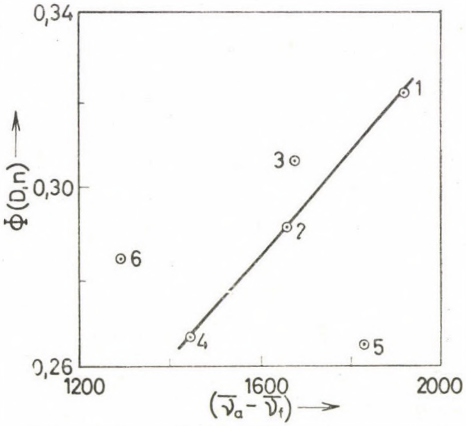

Fig. 1. $(\bar{\nu}_a - \bar{\nu}_f)$ plotted against $\Phi(D, n)$

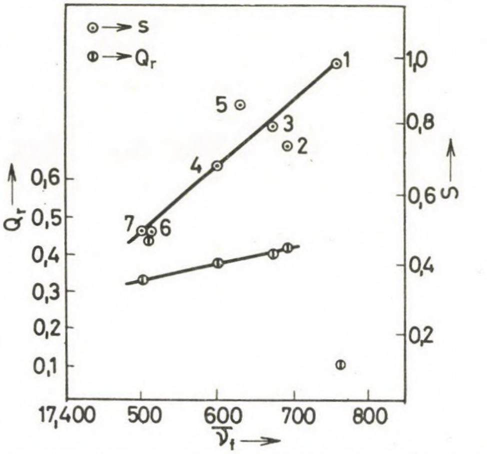

Fig. 2. Relative quantum yield, Qr and S values plotted against $\bar{\nu}_f$

Discussion

For a number of fluorescent substances, ZELINSKI [12] has shown that the plot "$\bar{\nu}_f$ vs S values" is a straight line and that the plot "$\bar{\nu}_a$ vs S values" is a smooth curve. In the case of eosin also, it is seen that the anomalous behaviour of acetone and glycerol which appears when relationship (1) is used (Fig. 1), is absent when the solvent-effect on the dye-molecule is represented on the spectroscopic solvent scale, (Fig. 2).

Coming to the relative quantum yield of fluorescence of the solutions of eosin in different solvents, it may be mentioned that the "quantum-yield of fluorescence vs $\bar{\nu}_f$" plot for a number of organic substances has been shown [17, 18] to consist, in general, of two linear parts. When solutions of a dye-stuff in different solvents (each with its characteristic value of $\bar{\nu}_f$) are used, it is found that the quantum yield of fluorescence at first increases linearly with $\bar{\nu}_f$, reaches an optimum value for a certain $\bar{\nu}_f$ and then decreases linearly with further increase in $\bar{\nu}_f$. This behaviour suggests the existence of two independent processes through which the excited dye-molecule may become deactivated. Fig. 2 shows that the fluorescence of solutions of eosin in the solvents studied follows this pattern, with water probably falling on the descending portion of the "fluorescence yield vs $\bar{\nu}_f$" plot.

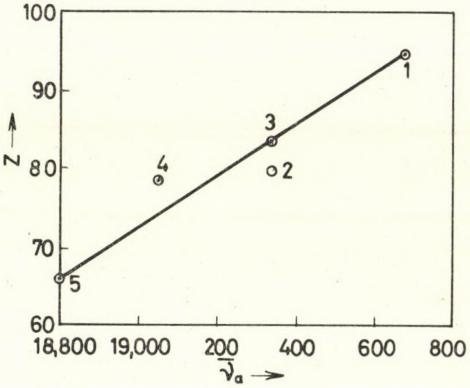

Fig. 3. KOSOWER's Z values plotted against $\bar{\nu}_a$

The term-values of the molecules in the ground state and the excited state and, consequently, $\bar{\nu}_a$ and $\bar{\nu}_f$ will be determined by the solvent—solute interaction in the ground state and the excited state, respectively. The linear relationship of the S and the Q_r values with $\bar{\nu}_f$ suggests that the efficiency of fluorescence — or the complementary process, viz., the non-radiative transfer of the excitation energy of the dye molecules — is determined by the interaction of the solvent with the solute-molecules in the excited state.

Coming to Fig. 3, the Z values give, according to KOSOWER, a measure of the solvent polarity. KOSOWER further contends that the Z value of a solvent is a microscopic measure of the solvent—solute interaction, while D is a macroscopic measure of the solvent behaviour. The slope of the line in Fig. 3 may be taken to indicate a smaller dipole moment of the eosin molecule in the excited state than in the ground state, as is inferred for fluorescein by ROHATGI [11] and for the pyridinium iodide complex by KOSOWER [14].

With a solvent change, the variations observed for eosin solutions, in the frequency maxima of their absorption and fluorescence spectra which reflect the variations in the dipole moment of the solute-molecule in the ground and the excited state, respectively, seem to confirm the above conclusion. The trend in the differences in $\bar{\nu}_a$ and $\bar{\nu}_f$, as we go from one solvent to another, indicates that the solvent interacts with the solute-molecule more strongly in the ground state than in the excited state. This may be attributed to a lower value of the dipole moment of the solute-molecule in the excited state than in the ground state.

We have shown qualitatively [19], from a study of the fluorescence spectrum and the relative quantum yield, that fluorescent dimers are formed in eosin as in erythrosin and rose bengale [20]. It is shown below (a) that evaluation of the frequency of the dimer peak, on KASHA's lines [21], gives a quantitative proof of dimer formation and (b) that the observed values of the term-differences in the monomer and the dimer levels in the ground and the excited state of the dye-molecule also suggest a decrease in the dipole moment of the eosin molecule on excitation.

For a parallel long-chain-dimer, resulting from dipole—dipole interaction, the energy-level shift from monomer is given [21] by

$$\mathscr{E} = \frac{M^2}{r^3}, \tag{2}$$

where M is the transition moment in e.s.u. and r the point-dipole, point-dipole distance may be taken as the distance between the centres of gravity of the two molecules.

Using the relation between the oscillator strength, f, and the transition moment, M

$$f = 4 \cdot 704 \times 10^{29} \cdot \bar{\nu}_\alpha \cdot M^2, \tag{3}$$

where $\bar{\nu}_a$ is the wave number of the maximum of monomer absorption, (2) becomes

$$\mathscr{E} = \frac{f}{4 \cdot 704 \times 10^{29} \times \bar{\nu}_a \times r^3}. \tag{4}$$

The oscillator strength, f, can be calculated from

$$f = 4 \cdot 315 \times 10^{-9} \int \epsilon_{\bar{\nu}} \cdot d\bar{\nu}, \tag{5}$$

where $\int \epsilon_{\bar{\nu}} \cdot d\bar{\nu}$ is the area under the graph "absorption coefficient, ϵ, vs $\bar{\nu}$" for the first absorption band between the two limits determined from the plot "polarization of fluorescence vs the exciting wave-length" [22a].

For eosin-blue in aqueous solution, we get $f \sim 0.50$ and $\bar{\nu}_a \sim 19\,700$ cm^{-1}. The value of r may be taken as 6Å [23]. The difference, $\Delta\bar{\nu}$, between the monomer and the dimer peaks, as calculated from (4), comes out to be 1250 cm^{-1}. The experimental values of $\Delta\bar{\nu}$ are $\sim$1300 cm^{-1} and 1020 cm^{-1} from observation on the absorption [22b, 24] and the fluorescence [19] spectra, respectively; these values are of the same order as the predicted value.

However, it is seen that the observed energy — difference in the monomer and the dimer peaks of eosin is smaller in fluorescence than in absorption. This may be due either to the dimers in the ground state being in the card-packed form, with somewhat oblique moments of the two monomers in the excited state of the dimer, or to the dipole moment of the dye-molecule in the excited state being less than that in the ground state.

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REFERENCES

1. M. K. Machwe, J. Kishore, K. Gopalakrishnan and K. D. Chaudhuri, Curr. Sci. **33,** 301, 1964.
2. S. I. Vavilov, Z. Phys., **53,** 665, 1929.
3. F. Perrin, Ann. de Phys., **17,** 283, 1932.
4. E. Rabinowitch, J. Am. Chem. Soc., **63,** 69, 1941.
5. W. L. Levshin, Z. Phys., **43,** 230, 1927.
6. E. J. Bowen and D. Seaman, Luminescence of Organic and Inorganic Materials, H. P. Kallmann et al. Ed., p. 153. John Wiley and Sons, Inc., New York, 1962.
7. B. S. Veshnikov, Acta Physichim. USSR., **4,** 453, 1936.
8. B. Ya. Sveshnikov and G. A. Tishchenko, Opt. i Spektr., **1,** 155, 1956.
9. N. Mataga, Y. Kaifu and M. Koizumi, Bull. Chem. Soc. Japan, **29,** 115, 465, 1956.
10. N. G. Bakhshiev, Opt. Spectry., **13,** 24, 104, 1962.
11. K. K. Rohatgi, J. Indian Chem. Soc., **42,** 737, 1965.
12. V. V. Zelinski and V. P. Kolobkov, Opt. i Spect. **16,** 186, 1964.
13. E. Lippert, Z. Electrochem., **61,** 962, 1957.
14. E. M. Kosower, J. Am. Chem. Soc., **80,** 3253, 1958.
15. V. V. Zelinski, V. P. Kolobkov and L. G. Pikulik, Opt. i Spekt. **2,** 402, 1957.
16. I. A. Zhmyreva, V. V. Zelinsky, V. P. Koblokov, A. S. Kochemironsku and I. I. Reznikova, Opt. Spectry., **8,** 214, 1960.
17. V. V. Zelinsky, V. P. Kolobkov and L. G. Pikulik, Opt. i Spektr., **1,** 161, 1956.
    V. V. Zelinski, V. P. Kolobkov and H. D. Krasnitskaia, Opt. Spectry., **6,** 261, 1959.
18. E. N. Viktorova, Opt. Spectry., **10,** 141, 1961.
19. S. S. Rathi and M. K. Machwe, Acta Phys. Hung., **23,** 449, 1967.
20. M. K. Machwe, J. Kishore, K. Gopalakrishnan and S. S. Rathi, Curr. Sci., **36,** 261, 1967.
21. E. G. McRae and M. Kasha, Physical Processes in Radiation Biology, Leroy Augenstein et al. Ed., p. 23, Academic Press, Inc. 1964.
22. P. Pringsheim, Fluorescence and Phosphorescence, (a) p. 381, (b) p. 354, Interscience Publishers, Inc., New York, 1949.
23. S. E. Sheppard, Rev. Mod. Phys., **14,** 303, 1942.
24. John P. Philips, Spectra-Structure Correlation, p. 156, Academic Press, Inc., New York, 1964.

ГАШЕНИЕ ФЛУОРЕСЦЕНЦИИ ЭОЗИНА В РАСТВОРЕ

Ш. Ш. РАТИ, К. ГОПАЛАКРИШНАН и Ю. КИШОРЕ

Резюме

Исследовались спектры поглощения и флуоресценции и относительный квантовый выход флуоресценции раствора эозина-синего в различных растворителях. Данные толкуются с точки зрения неизлучающих процессов, происходящих в растворе, образования димера и дипольного момента эозина. На основе результатов показывается, что молекула эозина в возбужденном состоянии имеет более низкий дипольной момент, чем в основном

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CALCULATIONS ON THE APPLICATION OF ELECTRON SCATTERING TO THE DETERMINATION OF PRESSURE

By

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Electron scattering is suggested to measure low pressures. The relation between the scattered electron current and the pressure of the gases is calculated using the Born approximation to obtain the differential cross-section of fast electrons. The method described by us uses the excess information deriving from elastic collisions. A further advantage over ionisation gauges is that the relation between the pressure and the measurable parameters can be computed. The calculations are carried out for three gases at different electron energies.

Introduction

The pressure of gases at small values of pressure is defined by the relation $p = nkT$, where p is the pressure, n is the number of molecules per unit volume; k is the Boltzmann constant and T is the absolute temperature. In the region of the validity of the perfect gas model this relation expresses the proportionality of the pressure to the concentration of molecules at constant temperature. In the case of vacuum systems of low pressures this relation is applied so widely that the operation of the ionisation gauges is founded upon the measurement of the concentration of molecules. A common characteristic of ionisation gauges of quite different constructions is the fact that the particles (generally electrons) interacting with the gas molecules produce ions proportional in number to the concentration of molecules, so the ratio between the ionising particle current and the measured ion current characterises the pressure. So, for the measurement of low pressures a problem is to increase the efficiency of ionisation and for this purpose the ionising electrons are constrained to the longest possible path by application of electric and magnetic fields. Since the produced electrons as well as the ionising electrons also must be collected, the presence of a fairly strong electric field in the measuring region is unavoidable. The complicated electron paths and the sensitive dependence of ionisation efficiency on the energies of the electrons and on the nature of the gases prevent us from calculating practically the relation between the ionising electron—ion current ratio and the concentration of gas molecules. So ionisation gauges used in the high vacuum and ultra high vacuum region must be calibrated and, naturally, the individual calibration for every gas gives a further source of errors, in addition to the difficulties of calibration. The other common characteristic of ionisation gauges is the fact that among the collisions of atoms and electrons only collisions causing ionisation provide information about the pressure. The numerous elastic collisions and those causing only excitation of molecules are unusable interactions for pressure measurement.

A new measuring principle

A few years ago the idea was raised that one could determine the concentration of gas molecules, i.e. the pressure, by the measurement of the current of the electrons scattered by elastic collisions. Over and above the greater information deriving from numerous elastic collisions a further advantage is given by the possibility of the realization of an equipotential field in the measuring region, where it is unnecessary to collect the ions. So the paths of the relevant electrons will be more simple physically and more suitable for a mathematical description. ANTAL and KŐNIG [1] suggested an arrangement where the electrons move in a homogeneous magnetic field on helical paths and a detector measures the intensity of the current of the non colliding electrons. This paper emphasizes the difference in principle between the method which takes the scattered electrons into account and the usual ionisation gauges, with regard to the possibility of mathematical treatment, but it suggests no detailed method for the calculations. For a possible method of calculation FALCKENBERG and SCHUMACHER [2] suggested a treatment, by which they determined the collision cross-section using the Born approximation. In their arrangement, used for calculations, a detector with a large surface is placed opposite the electron source, and a hole in the middle of the detector permits the non scattered electrons to pass through. In this way, the detector measures the intensity of the current of those scattered electrons that deviate with an angle larger than ϑ_0 as a consequence of their collision with the gas molecules along their straight paths in the measuring region. The electrons scattering through an angle ϑ_0, just reach the detector and the angle ϑ_0 depends on the place of the collision and on the geometrical conditions. The electron beam was supposed to be well focussed and monoenergetic having an energy of a few keV. It was possible to use the Born approximation for electrons of energies as large as these, and this enabled the current of the electrons scattered along the path of a given length to be determined from data on the differential cross-section. The authors published the results of their calculations for different gases at several electron energies. The results obtained give in logarithmic scale a linear relation between the pressure and the intensity of the scattered electron current in the region of low pressures but the values of the intensities are very small, detectable only by a particle counter.

The increase in the probability of collisions

The small intensity of the scattered electrons, a consequence of the straight line paths of the incident electrons, is the main disadvantage of the arrangement described in [2]. The only way to increase the intensity is to enlarge the size of the instrument, but this possibility is very limited in practice. The application of a magnetic field increases the probability of collisions and lengthens the paths of the electrons under unchanged geometrical conditions. On the basis of this let us investigate an arrangement as in [1] where a cylindrical measuring chamber is placed in a homogeneous magnetic field, parallel with the symmetry axis of the cylinder. Thus the focussed and monoenergetic electron beam entering the lower part of the cylinder will be constrained to a helical path. The length of this path depends on the angle α between the magnetic field and the entering electron beam. The detector is placed opposite the electron source to measure the current of the non-scattered electrons. If the distance between the electron source and the detector is l, then the length of the path of the unscattered electrons entering through an angle α is $l^* = l/\cos\alpha$.

It seems to be simplest with respect to both measurement and calculation to determine the pressure dependence of the decrease in intensity. But the decrease in intensity must not be too large, because it is then impossible to assume only simple collisions and one ought to take into account the space charge caused by the scattered electrons, all of which makes the calculations too complicated with the geometry described above. In the case of small decreases in intensity it is possible to measure the change in this decrease with satisfactory accuracy only within a small region of pressure. If the ratio between the currents of the entering electrons and those measured is equal to e.g. 0.9, then, when the pressure changes by one order, the proportion of the scattered electrons decreases from 0.1 to 0.01 approximately, i.e. the preceding ratio becomes 0.99, and so when the uncertainty of the measurement is 3% in the first case, this increases to 30%, other conditions remaining the same.

Therefore, instead of measuring the decrease in intensity, the variable is taken to be the angle between the entering electron beam and the direction of the magnetic field, and its dependence on the pressure will be determined, while the decrease in intensity will be assumed to be constant (Fig. 1). The measurement of the angle presents some technical difficulties, particularly as the position of the detector has to be changed depending on the angle α owing to the change of the helical path. But the measurement of the change of energy of the electron beam entails even more difficulties. The advantages expected from the considerable increase in length of the electron path suggests that the relation between the pressure and the angle α should be calculated.

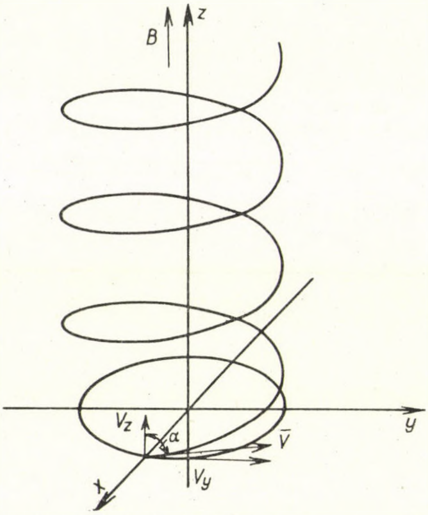

Fig. 1. Helical path in homogeneous magnetic field

Calculation of the scattered electron current

In the case of slow electrons the scattering varies so greatly for different scattering atoms and the relative intensity of the scattered electrons depends so sensitively on the velocities of the incoming electrons, that the calculation of the scattering cannot give a useful result suitable for the required purpose. The calculation of the scattering will be made for electrons with an energy of a few keV as this is also more suitable for technical reasons. The basis of the calculations is the determination of the differential cross-section using the Born approximation.

In our problem the scattering centres are the gas molecules and the scattered particles the electrons. dN, the number of the particles scattered per unit time in the solid angle $d\Omega$ is proportional to N, the current of the incoming particles, and to the solid angle $d\Omega$:

$$dN = \sigma(\vartheta)\, N d\Omega\,, \tag{1}$$

where $\sigma(\vartheta)$ is the differential cross-section, which depends on the plane angle ϑ only, because the solid angle between cones with vertical angles ϑ and $\vartheta + d\vartheta$ is $d\,\Omega = 2\,\pi \sin\vartheta\, d\,\vartheta$. Here $\sigma(\vartheta)$ characterises both the elastic and inelastic collisions. The lengths of the electron paths allow the assumption of straight motion in the region of the interaction with the molecules i.e. the magnetic field does not affect the collision process.

In the case of the scattering of fast electrons **Morse** [3] gave the differential cross-section for elastic scattering in the form

$$\sigma = \frac{4}{a_H^2 q^4} \left[(Z - F)^2 + S \right], \tag{2}$$

where a_H is the Hydrogen Bohr-radius; q is the absolute value of the difference of the momenta of the incident and scattered electrons; Z is the atomic number, $F = F(\vartheta)$ is the form factor of the scattering atoms characterizing the elastic collisions; S describes the inelastic scattering and it is determined by the sum of the squares of the form factors of every electron of the scattering atoms. Evidently, for the determination of the differential cross-section the form factors of the atoms must be known. The values of atom form factors are known for numerous atoms from calculations by **Wyrwich** and **Lenz** [4]. Their calculations were performed by using the radial density functions, determined by the self-consistent field method. Let dN be the number of the electrons scattered into the solid angle $d\Omega$ characterized by the angle ϑ when the number of the incident electrons passing over the gas layer of very small thickness l' is N_0, so

$$dN = \varrho \cdot l' \cdot N_0 \cdot \sigma \cdot d\Omega , \tag{3}$$

where ϱ is the density of the gas. Evidently, the number of the scattered electrons is proportional to the density of the gas, the thickness of the gas layer and the number of the incident electrons.

The problem is to find the ratio between the number of the electrons scattered through angles larger than a minimum ϑ_0, and the number of the incident electrons. To do this it is *necessary* to integrate equation (3) from the given angle ϑ_0 to the maximum angle ϑ_1, where the values of F and S are not negligible within the accuracy of the calculations. Using (2) we obtain

$$\frac{N'}{N_0} = \frac{8 \pi l' \sigma}{a_H^2} \int_{\vartheta_0}^{\vartheta_1} \frac{\left[(Z - F)^2 + S \right]}{q^4} \sin \vartheta \, d\vartheta . \tag{4}$$

The problem is to find ϑ_0 as a function of the given geometry, (detector surface was assumed to be 2 mm^2) because the scattering takes place mainly in the region of small angles. In (4) the values of ϑ_0 and ϑ_1, change continually along the length of the gas layer also in the case of straight line paths of the electrons since the position of the detector determinates ϑ_0 and ϑ_1. In the case of the electrons moving in a helical path the use of formula (4) is indirect, for, because of the fixed position of the detector, it is not irrelevant in which of the points of the path the collision takes place. First, it is necessary to calculate an average which depends on the vertical position of the helical path, then in every circle depending on the relative position between the detector and the place of the collision. Since the probability of scattering is independent of the angle φ between the plane yz and the plane uv but the different directions mean different minimum angles ϑ_0, it is also necessary to calculate the average of the angle φ (Fig. 2). The determination of the maximum angle ϑ_1 is also carried out in a similar manner, but since the scattering leading the electrons to some path directing to the detector has a very small probability, it is easy to take into account this correction. The calculations were carried out by numerical integration using the data of WYRWICH and LENZ. The pressure values belonging to all angles α are determined from the condition $N/N_0 = 0.1$, (here N is the number of the scattered electrons, the sum of N' quantities), and using (4) we can calculate the gas density ϱ, i.e. the pressure at a given temperature.

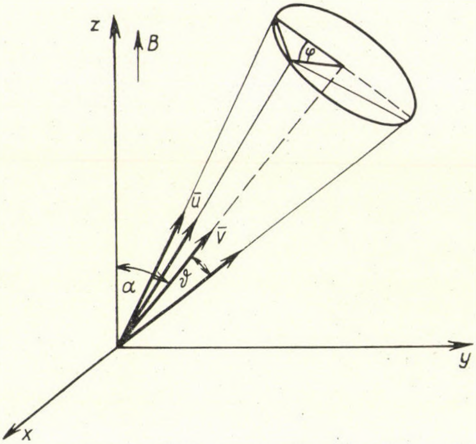

Fig. 2. The characterising of the scattering by the angles α, ϑ and φ

The results of the calculation

The calculations were carried out for three different gases at three electron energies. We calculated the intensity decrease caused by scattering as a function of the pressure in the case of electron energies of 3 keV, 10 keV and 50 keV for nitrogen, oxygen and neon. The results of the calculations are shown in Table I and for nitrogen in Fig. 3. From Table I we see the similarity of the connection between the pressure and the angle α in the case of every gas at different energies. We can find the decrease of the degree of the change with the increase of energy only for large values of α, which is probably a result of the increase of q under unchanged ϑ_0. The comparison of the different electron energies also shows well numerically that an electron beam of lower energy is suitable for the measurement of low pressures, but the limits of validity of the Born approximation used in the calculation of scattering restrict the decrease of the energy. It seems, too, that in the case of oxygen and nitrogen the difference is less then 5 per cent and so it is possible to calculate the pressure values for air.

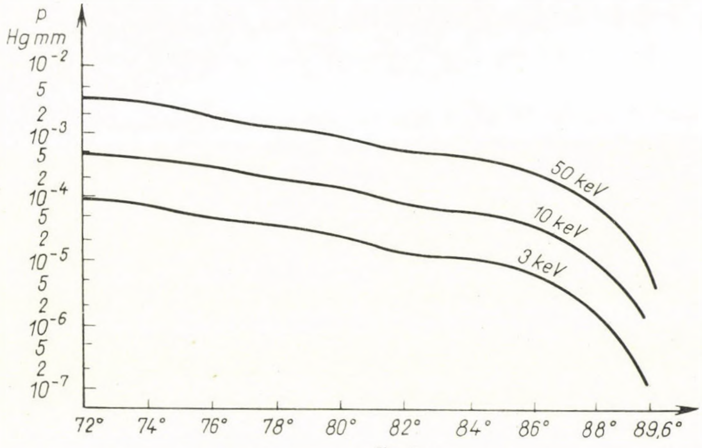

Fig. 3. The relation between the pressure and the angle α at different electron energies in the case of nitrogen

For the calculations leading to these results we must make the following remarks: At the basis of the results is the supposition that the electron scattering is simple. Under the given intensity decrease since the probability of double scattering of the electrons is roughly equal to the square of the simple scattering, the effect of double or manifold scattering is negligible. A further reason is the fact that a further scattering of an electron scattered from the entering electron beam, which takes it back to the original helical path, has a very small probability. All other scatterings make no difference to the detected electron intensity.

In the case of an inelastic collision we must take into account the new electrons produced by the ionisation besides the incident electrons. But the velocities of the electrons produced in such a way are very small — electrons do not accelerate in a field of constant potential — and so the paths of these electrons deviate from the paths of the ionising electrons. This conclusion is also valid for heavy ions. Further, the ratio between the elastic and inelastic scattering shows that, above all, the elastic scattering is responsible for the intensity decrease. (In the case of electrons of 10 keV this ratio is 10 : 1 when $\alpha = 85°$.)

Table I

Pressure values (in 10^{-6} Hgmm) of nitrogen, oxygen and neon gases at different electron energies

α	Nitrogen			Oxygen			Neon		
	3 keV	10 keV	50 keV	3 keV	10 keV	50 keV	3 keV	10 keV	50 keV
72°	99.5	501	3490	97.3	482	3370	234	1042	6730
74°	76.8	407	2960	75.0	391	2630	181	794	5410
76°	49.3	268	1750	48.6	256	1680	124	521	3520
78°	37.6	203	1290	36.7	197	1240	91.2	392	2570
80°	28.4	149	927	25.5	143	882	62.0	287	1830
81°	17.7	92.3	631	17.2	89.5	613	47.5	209	1370
82°	15.3	78.4	550	14.7	76.2	536	40.2	189	1060
83°	13.2	69.1	491	12.9	67.4	472	34.2	154	911
84°	11.4	60.2	389	11.2	58.6	381	29.4	121	784
85°	9.24	50.4	324	9.23	48.2	314	23.2	98.7	652
86°	5.71	31.2	202	5.50	30.1	195	14.1	61.0	461
87°	3.43	20.3	140	3.30	19.8	137	9.00	34.7	271
87.5°	2.24	13.9	101	2.23	13.5	99.6	5.31	25.2	179
88°	1.46	9.75	72.4	1.42	9.47	70.8	3.51	18.8	121
88.4°	0.892	6.70	50.4	0.873	6.51	49.1	2.13	11.1	72.4
88.8°	0.560	4.21	28.9	0.546	4.12	28.4	1.47	6.81	43.5
89°	0.411	3.27	22.6	0.394	3.21	22.3	1.12	5.16	31.7
89.2°	0.286	2.26	17.4	0.275	2.23	17.1	0.671	3.70	21.8
89.4°	0.190	1.63	12.3	0.183	1.59	12.0	0.491	2.17	15.9
89.6°	0.101	0.87	9.4	0.097	0.83	9.1	0.261	1.21	8.20

Discussion

The expected limit of the low pressure where the treatment is useful is at the value 10^{-7} Hg mm (See Table I). This limit can be extended to the region of lower pressures by the increase of geometrical sizes or the more accurate measurement of the intensity of the current of non-scattered electrons (the decrease of the ratio N/N_0), but these possibilities have narrow limits in practice. Otherwise, the increase of the pressure region towards the lower pressures is possible only by the application of an electron beam of lower energy, but the method used is unsuitable for the calculation of the scattering of slow electrons. Naturally, it is possible to calculate the scattering of slow electrons, but the application of this calculation to the chosen geometry (or any kind of geometry) is impracticable because of the sensitive change of the dependence on the velocities of the differential cross-section and the complicated distribution of the intensities of the electrons, scattered into the different solid angles. This difficulty does not impede the application of slow electrons and so the increase of the pressure region towards low pressures, but it is impossible to calculate the relation between the pressure and electron scattering. On the basis of the experimental results of SCHUMACHER [5] we can estimate the expected numerical values: using electrons of 300 eV the measurable pressure region can be extended to 10^{-10} Hg mm in the case of nitrogen.

The increase of the pressure region towards pressures higher than 10^{-3} Hg mm is possible by the increase of the electron energies but it is more convenient to measure the change of the intensity decrease as a function of the increase of the pressure under a given electron energy and a fixed angle α. In this case the physical circumstances are also very complicated because of the numerous scattered electrons, and so we cannot calculate the necessary relations.

Since experimental results concerning the arrangement are not at our disposal we cannot conclude much from the comparison with ionisation gauges. But it is expected that effects disturbing the measurement which are caused by the ions will decrease since the majority of the electron scatterings are elastic and we can expect smaller intensities because the measuring current of non-scattered electrons is about equal to the current of the incident electrons.

The principle of measurement reviewed here is basically different from that of ionisation gauges because it uses elastic collisions as well as ionising collisions. Since the ratio between elastic and inelastic collisions is 10 : 1 in the case of electron energies of a few keV, this fact increases the sensitivity of the measurement in constructions of similar arrangement. Further, the computable geometry and the physical simplicity of the principle allow the calculation of the relation between the pressure and the measured quantities in a measuring region, whereas with other instruments it is much more difficult.

REFERENCES

1. J. ANTAL and A. KŐNIG, Vacuum, **14/15,** 318, 1962.
2. H. R. FALCKENBERG and B. W. SCHUMACHER, Canad. J. Phys., **40,** 1521, 1962.
3. P. M. MORSE, Z. Phys., **33,** 443, 1932.
4. H. WYRWICH and F. LENZ, Z. Naturforsch., **13,** 515, 1958.
5. B. W. SCHUMACHER, Ann. Physik, **13,** 404, 1953.

РАСЧЕТЫ О ПРИМЕНЕНИИ РАССЕЯНИЯ ДЛЯ ОПРЕДЕЛЕНИЯ ДАВЛЕНИЯ

Л. ФЮШТЭШ

Резюме

Для измерения низких давлений предлагается использовать рассеяние электронов. Соотношение между током рассеянных электронов и давлением газов определяется применением Борновского приближения с целью получения дифференциального сечения быстрых электронов. Описанный нами метод использует информации, происходящие от упругих столкновений. Дальнейшее преимущество ионизационных измерителей заключается в том, что имеется возможность для определения соотношения между давлением и измеряемыми параметрами. Вычисления проводились для трех газов при различных энергиях электронов.

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SMALL ANGLE ELASTIC SCATTERING OF 14 MeV NEUTRONS BY ^{238}U

By

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The theories proposed by various authors for the explanation of the apparently anomalous small-angle elastic scattering of fast neutrons by ^{238}U nuclei [1] are reviewed. The problem is then considered by questioning the validity of the model and the assumptions which give rise to the anomalous discrepancy between the predictions and the experimental data. The measurement of the angular distribution of 14.7 MeV neutrons scattered elastically by uranium is reported. In the evaluation of the experimental data the inelastic contribution is taken into account as a consequence of bad energy resolution. The analysis of the scattering data in terms of the optical model does not show the occurrence of any anomaly.

Introduction

The anomalous elastic scattering of neutrons on ^{238}U has been studied by several authors in the last few years [1—8]. The problem was raised by Y. A. ALEKSANDROV et al. [1] who were the first to observe, using bombarding neutron energies of about 2 MeV, that the differential cross-section for elastic scattering at small angles ($\vartheta < 15°$) differs from the value predicted with the assumption of a "black nucleus". Their observation was confirmed by the experiments of Y. V. DUKAREVICH et al. [2] who used 14 MeV neutrons and had better experimental conditions. Similar deviations from the predictions have also been reported by other authors 6, 5, 3 for bombarding energies of 7, 3, 0.8 and 0.6 MeV. The theories proposed by various authors for the explanation of this effect may be summarized as follows.

Contributions from long-range nuclear interactions to elastic scattering

It seemed obvious to attribute the increased small-angle scattering of neutrons to the effect of long-range interactions between the neutron and nucleus.

The interaction potential of the nuclear Coulomb field with the magnetic moment of the neutron has the form

$$V_s(r) = |\mu_n| \frac{e\hbar}{2\, m^2\, c^2} \sigma \cdot E \times p = |\mu_n|\, (Ze^2\, \hbar^2/2\, m^2\, c^2)\, \Phi(r)\, \sigma \cdot l, \tag{1}$$

where μ_n is the magnetic moment of the neutron and $Z \cdot e$ is the charge of the scattering nucleus.

The radial dependence is described by the terms

$$\begin{aligned} \Phi(r) &= r_c^{-3} \quad \text{if} \quad r \leq r_c\,, \\ \Phi(r) &= r^{-3} \quad \text{if} \quad r > r_c\,, \end{aligned} \tag{2}$$

where r_c is the "charge radius" characterizing the charge distribution of the scattering nucleus, σ is the Pauli spin vector.

The Schwinger scattering, described by this potential [9], has a predominant forward character. Using the Born approximation the scattering amplitude is given by

$$f_s(\vartheta) = i\gamma \cot \frac{\vartheta}{2} (\sigma \cdot n) \tag{3}$$

for ^{238}U $\gamma = 1.35 \cdot 10^{-14}$ cm, n is the normal vector of the scattering plane.

As is well-known, the contribution from Schwinger scattering is not sufficient to account for the experimental data.

Y. A. ALEKSANDROV and I. I. BONDARENKO [3] suggested that on the basis of fast neutron scattering on Coulomb fields consequences may be drawn covering the electric polarizability of the neutron.

They assumed that in the nuclear Coulomb field E an induced electric dipole moment $p = \alpha E$ is to be attributed to the neutron, where α is the electric polarizability of the neutron. The interaction potential has the form

$$\begin{aligned} V_p(r) &= -\frac{1}{2}\alpha E^2 = -\frac{1}{2}\alpha Z^2 e^2 \psi(r)\,, \\ \psi(r) &= r_c^{-4} \text{ if } r \leq r_c\,, \\ \psi(r) &= r^{-4} \text{ if } r > r_c\,. \end{aligned} \tag{4}$$

This interaction may increase the small angle scattering ($\vartheta < 15°$) and its extent depends on the electric polarizability of the neutron.

Supposing that the observed anomaly is connected with the polarizability ALEKSANDROV deduced a value of $\alpha = (8 \pm 3.5) \cdot 10^{-41}$ cm^3 for $E_n' = 2$ MeV. However, the meson theory [7] and deuteron-photon scattering experiments [9], show that the value of α is actually less by an order of magnitude.

M. WALT and D. B. FOSSAN [8] also studied the interference between nuclear scattering and polarization for the interpretation of the scattering of 0.57 MeV neutrons. To describe the short range nuclear interaction, they used the optical potential

$$V_n(r) = VF(r) - iVG(r) + V_{s0}\left(\frac{\hbar}{m_\pi c}\right)^2 \left(\frac{1}{r}\frac{d}{dr}F(r)\right) \sigma \cdot l\,, \tag{5}$$

where the real and imaginary parts of the potential are described by the form functions $F(r)$ and $G(r)$, with depths V and W, respectively, while V_{s0} is the strength of the spin-orbit term and m_π is the mass of the π meson.

Using the Born approximation, the scattering amplitude also contains two terms in the form

$$f_n(\vartheta) = f(\vartheta) + g(\vartheta)\,\sigma \cdot n \tag{6}$$

corresponding to the spin-dependent parts and to those which show only radial dependence.

If the interference term is taken into account in the differential cross-section expressed as

$$\frac{d\sigma}{d\Omega} = |f_n|^2 + |f_p|^2 + 2 f_n f_p\,, \tag{7}$$

then, for the scattering of neutrons with energy $E_n = 0.57$ MeV, the lower limit of polarizability is

$$\alpha \lesssim 4 \cdot 10^{-40}\,\mathrm{cm}^3,$$

while from the data obtained with 14 MeV energy neutrons [2] the value

$$\alpha > 4 \cdot 10^{-40}\,\mathrm{cm}^3$$

was deduced. These values are inconsistent with those obtained for α in other types of experiments. R. F. REDMOND [4] attempted to explain the anomaly by introducing an extraordinarily strong spin-orbit term V_{s0} into the nuclear interaction potential. The long-range electromagnetic interaction (Schwinger scattering) is also of the spin-orbit type. Using the differential cross-section

$$\frac{d\sigma(\vartheta)}{d\Omega} = |g_n(\vartheta) + f_s(\vartheta)|^2\,, \tag{8}$$

where g_n is the spin-orbit term of the nuclear scattering amplitude and f_s is the amplitude of Schwinger scattering, REDMOND found that for sufficiently strong V_{s0} the interference is constructive at small angles and destructive at larger ones.

Though the character of the interference would permit such an explanation of the anomaly, if we take into account the analysis of A. J. ELWYN et al. [5] we have to abandon this assumption. They measured the elastic scattering of 0.83 MeV neutrons and the polarization of the scattered neutrons. The anomaly, though it was observed, did not give any contribution to the polarization of the scattered neutrons. The latter can be explained without the assumption of a very strong spin-orbit interaction term in the optical potential. Thus, the suggestion of REDMOND cannot be accepted.

One of the present authors, P. Hraskó, who estimated the interference between the spin-orbit terms at the small angle elastic scattering of 14 MeV neutrons in terms of the "black-nucleus" model, found the contribution from interference to be negligible, if usual values of V_{s0} were assumed. Thus, the interference suggested by Redmond does not lead to any increase in the scattering cross-section either in the case of 870 keV or at 14 MeV.

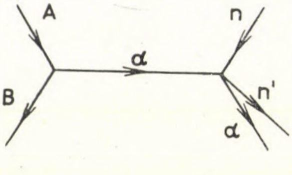
These facts allow us to state that the theories which attribute the anomaly to the effect of long-range interactions have to be rejected.

Contribution from special nuclear reactions to elastic scattering

If the energy balance of a nuclear reaction happens to be such that the neutron emitted in the forward direction has an energy equivalent to that of the elastically scattered neutron, the nuclear reaction can be made responsible for the excess forward scattering observed.

Considering the type of reactions which could satisfy this condition, it is of interest to note that the anomaly was observed by Dukarevich et al. only in elastic scattering experiments on U, Th and Pu nuclei. Thus, it seems reasonable to connect the anomaly with the α-decay of these nuclei.

In our investigations we assumed the induced α-decay, i.e. the possibility of $(n, \alpha\, n')$-type reaction [11]. The amplitude of the reaction is evaluated by P. Hraskó and Zs. Kövesy, using the pole approximation of the dispersion model proposed by Shapiro, i.e., from the infinite set of terms describing the amplitude they used only that which is represented in the pole graph by



expressed as

$$M \sim \frac{M(A \to B + \alpha) \cdot M^*(\alpha + n \to \alpha' + n')}{p_a^2 - 2\, m_a E_\alpha} . \tag{9}$$

P_a and E_a are functions of the kinetic energy available in the initial and final state, respectively. In the process $A \to B + \alpha$, if the scatterer is ^{238}U, we have $Q > 0$. It follows from the kinematic relations involved in the momentum transfer between the "decay" and the $\alpha + n \to \alpha' + n'$ scattering process that the neutrons are scattered with very high probability in the direction of the bombarding neutrons and then their energy is equal to the energy of the neutrons scattered elastically by uranium. This effect can lead to an increase in the elastic scattering cross-section. If it is actually responsible for the anomaly, it should account for the excess value of $\sigma = 15$ mb obtained for the differential cross-section from the experimental data [2].

In the experiment performed by the present authors [12] in order to check this theory, the value of the U$(n, \alpha\, n')$ cross-section was found to be $\sigma < 0.5$ mb, which was too small to explain the anomaly.

Considering that the anomalous behaviour increases with increasing bombarding energies and is observed only for U, Pu and Th scatterers, A. J. ELWYN et al. [5] tried to relate the anomaly to the fission of these nuclei. The energy width of the bombarding neutrons is such that even in the relatively low range of the neutron energies applied, it may attain 0.6 MeV, which is the approximate threshold energy for the fission of ^{238}U. In this approach the variation of the anomaly with the bombarding energies is thought to reflect the energy-dependence of the fission cross-section.

In the experiments performed by the present authors for the investigation of the $(n, \alpha\, n')$ reaction the measuring technique enabled us to get the sum of the cross-sections for (n, fn')-type reactions where f stands for any charged particle with an energy equivalent to that of an α-particle of minimum 1 MeV [13]. The small value of the integral cross-section obtained in the measurement contradicts the theory of ELWYN, the more so if one realizes that at the 14 MeV bombarding energies used in the experiment, the fission contribution is expected to be much more pronounced.

Considerations of the existence of an anomaly in the ^{238}U(n,n)

In the light of the above results, it can be questioned whether the anomaly exists at all. Let us see, therefore, if the experimental data have been compared with the proper theoretical predictions. The calculations are based on several assumptions. To ascertain the validity of the approximations in the case of ^{238}U, we shall consider successively each of the factors which may affect the differential cross-section for elastic scattering.

Effect of nuclear deformation

The ^{238}U nucleus is known to be strongly deformed. Thus, an interaction potential which includes the contribution from the collection nuclear deformation will give a more accurate description of the interaction mechanism.

We consider, therefore, the small angle scattering behaviour without the assumption of spherical symmetry. Recalling also that the energy resolution in the experiment of **Dukarevich** et al. was less than 1.5 MeV, it seems necessary to perform a new evaluation of the experimental data.

It is apparent from the known level scheme of ^{238}U (Fig. 1) that the cross-section data contain both the elastic and inelastic scattering contributions to the cross-section. These inelastic scatterings excite the rotational band of the ground state and other collective states of a non-rotational nature.

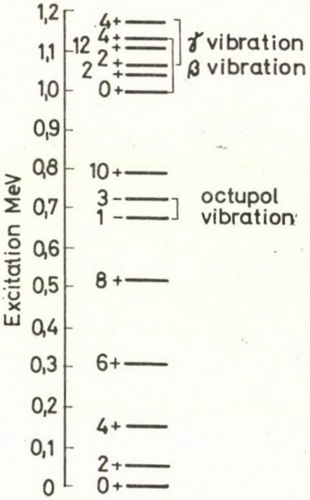


Fig. 1. Level scheme of ^{238}U

Consequently, the experimental data can be better described by a sum of the form

$$\frac{d\bar{\sigma}(\vartheta)}{d\Omega} = \frac{d}{d\Omega}\sigma_{0^+\to 0^+}(\vartheta) + \sum_{J'M'} \frac{d}{d\Omega}\sigma_{0^+\to J'M'}(\vartheta), \tag{10}$$

where $J'M'$ represents the states summed over the rotational band. The $d/d\,\Omega\bar{\sigma}(\vartheta)$ was evaluated in the adiabatic approximation by making use of the generalized diffraction model of **Blair** [15]. The amplitude of the inelastic scattering for transitions from the O^+ state to those characterized by quantum number JM is given by

$$f_{0^+\to JM} = < JM\,|f_{0^+\to 0^+}|\,00 >, \tag{11}$$

where $f_{0^+\to 0^+}$ is the amplitude of the elastic scattering associated with the given value of nuclear deformation. The ^{238}U nucleus rotates 1° if a 14 MeV neutron passes by. Thus, the adiabatic approximation can be applied. In the scattering process the nucleus is regarded as a rigid ellipsoid fixed at the origin.

The cross-section for elastic scattering to the total rotational band, as defined in (10), now has to be written in the form

$$\frac{d\bar{\sigma}(\vartheta)}{d\Omega} = \frac{1}{4\pi}\int |f_{0^+\to 0^+}|^2 \, d\Omega \,, \tag{12}$$

where the elastic scattering is averaged over the positions of the ellipsis.

The amplitude $f_{0^+\to 0^+}$ is evaluated in the diffraction approximation in terms of a Fraunhofer diffraction pattern resulting from an ellipsis with symmetry axes α, β in a fixed position (ω, ψ), normal to the incident neutron beam. In the direction ϑ, φ

$$f_{0^+\to 0^+} = f_F(\vartheta, \varphi, \omega, \psi) = ik\,\frac{1+\cos\vartheta}{2}\, w\cdot\beta\cdot J_1(x)\,,$$

where

$$x^2 = \left[w^2\cos^2(\varphi-\psi) + \beta^2\sin^2(\varphi-\psi)\right] k^2 \sin^2\vartheta\,, \tag{13}$$

$$w^2 = \alpha^2\sin^2\omega + \beta^2\cos^2\omega\,.$$

k is the wave number of the incident neutron, $J_1(x)$ is the first order Bessel function and the deformation parameter is defined as

$$\delta^2 = \frac{\alpha^2-\beta^2}{\alpha^2}\,.$$

The measured data obviously do not contain the inelastic contribution leading to the excitation of the total rotational band, but it is obvious that for higher values of J the inelastic contribution to $\bar{\sigma}(\vartheta)$ becomes increasingly smaller.

From (12) and (13) we get the expression

$$\frac{\bar{\sigma}(\vartheta)}{\cos^4\dfrac{\vartheta}{2}} = \bar{\sigma}(\vartheta) + \gamma\sin^2(\vartheta) \tag{14}$$

for small angles, where the deformation of the target nucleus determines the values of $\bar{\sigma}(0)$ and γ. The dependence of these values on δ is plotted in Fig. 2. For electromagnetic transitions the measured values of B(E2) yield $\delta_{EM} = +0.446 \pm 0.008$. The electromagnetic transition data are obviously sensitive to the nuclear charge distribution and consequently to the nuclear deformation of this type. This may differ from the mass deformation also indicated by nuclear spectroscopic data. The values of γ and $\bar{\sigma}(0)$ are, however, practi-

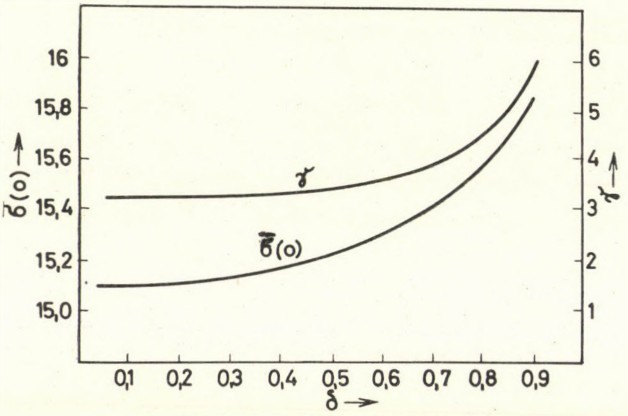


Fig. 2. δ-dependence of $\bar{\sigma}(0)$ and γ

cally insensitive to the deformation over a fairly wide range around δ_{EM}. Thus, it cannot be expected that an explanation of the problem of small angle scattering should be found in this way.

Analysis in terms of the optical model

A further assumption used in the evaluation of Dukarevich's experiment was the assumption of a "black nucleus" which implies the absorption of every incident neutron. The use of the optical model seems to be more realistic. However, the analysis and getting to satisfactory conclusions concerning the small angle anomaly would require experimental data with a much smaller error and a better energy resolution than those obtained in the reported expe-

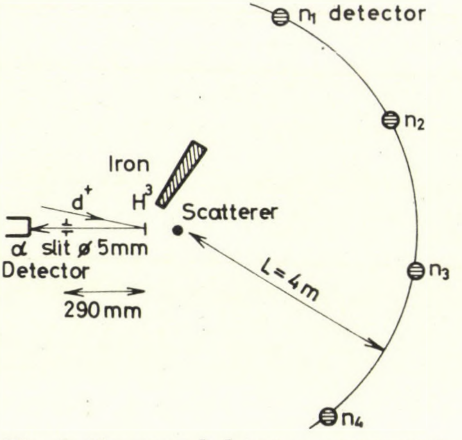


Fig. 3. Target and detector arrangement

riments [2, 12]. For a proper comparison with the values calculated, the elastic scattering in terms of the optical model as well as for the unambiguous determination of the optical parameters, it is necessary to give a reasonable estimate of the inelastic contribution to the measured value of the elastic scattering cross-section. As a consequence of the collective nature of the inelastically excited levels, the direct interactions are expected to dominate, and the compound processes can be neglected.

This problem was investigated by the present authors who simultaneously measured the angular distributions of the elastically scattered 14.7 MeV energy neutrons and of the inelastically scattered part, exciting the first 2^+ and 4^+ levels in ^{238}U.

Experimental

The differential cross-sections for the above types of scattering processes were evaluated by detecting the neutrons scattered by a uranium target and analyzed with a four-channel fast neutron time-of-flight spectrometer [17]. The neutrons were observed simultaneously at four scattering angles, over a flight path of 4 m, with an energy resolution of 0.5 MeV. The arrangement of the target and detector is shown in Fig. 3. The radius of the cylindrical target was chosen to be $\lambda_{att}/4$, where λ_{att} is the mean free path of attenuation for the bombarding neutrons.

The recoil particle technique was used which in the case of neutrons from $H^3(d, n)He^4$ reaction provides a well collimated bombarding neutron beam. The deuterium beam was led through a slit of 0.1 mm by 0.5 mm placed in front of the H^3 target. The solid angle covered by the α-detector was $2.7 \cdot 10^{-5}$ sterad. The divergence of the neutron beam "collimated" by means of the detected α-particles is determined by the solid angle of the α-detector and the width of the deuterium energies used for the production of the neutrons.

To limit the size of the neutron source, the deuteron beam was collimated by two semi-cylindrical iron blocks, 5 cm in radius and 50 cm long each, placed in front of the incident deuterium beam, with 0.5 mm spacing between them. In this way an intensive self-target was moved further from the tritium target and the detectors were shielded against self-target neutrons. The direct neutrons were prevented from reaching the detectors at higher scattering angles by a 50 cm long cylindrical iron shield. The neutrons were detected by cylindrical plastic scintillators, 5 cm in diameter and 10 cm in length. The detector axis was kept parallel to the direction of the flight path of the neutrons. The total angular resolution was $\Delta\vartheta = \pm 1.4°$. The measurements were taken in the angular range between $5° < \vartheta < 18°$ in steps of 2° and between $20° < \vartheta < 100°$ in steps of 15°. The threshold energy for neutron detection was equivalent to 3 MeV proton energy.

A typical time-of-flight spectrum of neutrons scattered at $\vartheta = 5°$ is shown in Fig. 5.

The random coincidence background varying with the square of the bombarding neutron intensity was measured simultaneously in a given range of the spectrum, suppressing with appropriate delays the systematic events in this region.

The 1 mm thick tungsten backing of the tritium target acts as a scatterer in the solid angle of incident neutrons and contributes to the background counts in the time-of-flight spectrum. To evaluate this effect the measurements were performed both with and without a scatterer.

The differential cross-section was evaluated from the expression

$$\frac{d\sigma(\vartheta)}{d\Omega} = \frac{N_k - \dfrac{N_\alpha}{N_{\alpha b}} N_{kb}\, e^{-\frac{l}{\lambda_{ntt}}}}{N_\alpha\, \Omega_n\, \varepsilon(E_n)} \, \frac{\sigma_{att}}{1 - e^{-l/\lambda_{att}}}, \tag{15}$$

where N_k and N_{kb} are the counts falling into the area below the elastic scattering peak in the time-of-flight spectrum after subtraction of the random coincidence counts distributed uniformly in time. N_a stands for the counts of the α-detector. The suffix b denotes the counts taken without a scatterer, l is its length in the direction of the incident neutrons, σ_{att} is the attenuation cross-section of the scatterer, Ω is the solid angle and ε_n the counting efficiency of the neutron detector. Expression (15) holds whenever the solid angle of the scatterer is larger than Ω_a, as was the case in our measurements.

The differential cross-section for elastic scattering was evaluated in terms of the optical model, assuming the complex potential to have the usual form

$$U(r) = Vf(r) + iWg(r) + V_{s0}\, h(r)\, \sigma \cdot l, \tag{16}$$

where

$$f(r) = \left[1 + \exp\frac{r - R_v}{a_v}\right]^{-1},$$

$$g(r) = \frac{\exp\dfrac{r - R_w}{a_w}}{\left[1 + \exp\dfrac{r - R_w}{a_w}\right]^2},$$

$$h(r) = -\left(\frac{\hbar}{m_\pi c}\right)^2 \frac{1}{2} \frac{d}{dr} f(r).$$

The parameters $R_v = R_w = 1.25\, A^{1/3} f$, $a_v = 0.65\, f$, $a_w = 0.55\, f$, where V and W have the values given by the expressions of BJORKLUND and FERNBACH [18]. The values of the potential are in agreement with those reported on the neutron scattering analysis on uranium [16, 19].

As for the inelastic contribution within the given energy resolution, it is sufficient to consider the excitation to the 2^+ state, the excitation to the second rotational level seems to be of low probability. The theoretical values for this correction were taken from [19].

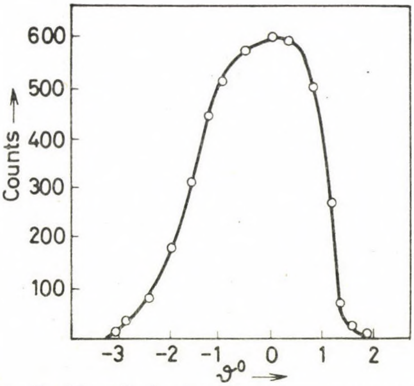


Fig. 4. Profile of the bombarding neutron beam

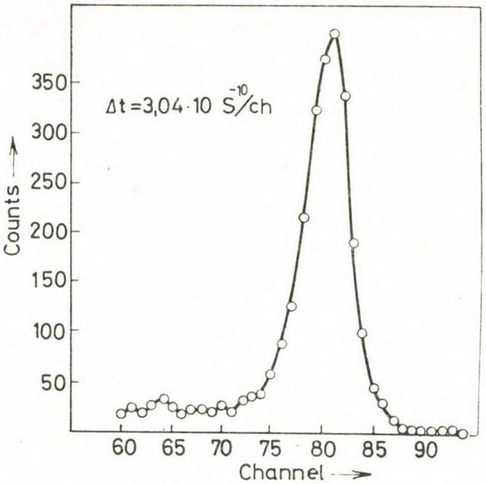


Fig. 5. Time-of-flight spectrum of the elastically scattered neutrons at a flight distance of 4 m at angle $\tau = 5°$

In Fig. 6 we have plotted the measured and calculated angular distribution in the range $10° < \vartheta < 100°$. As is to be expected, the inelastic contribution in the forward direction can be neglected and may become appreciable only in the diffraction minima. Nevertheless, if it is taken into account, it considerably changes the agreement between the experimental values and the predictions. The contribution from the compound elastic scattering can be ignored at this energy.

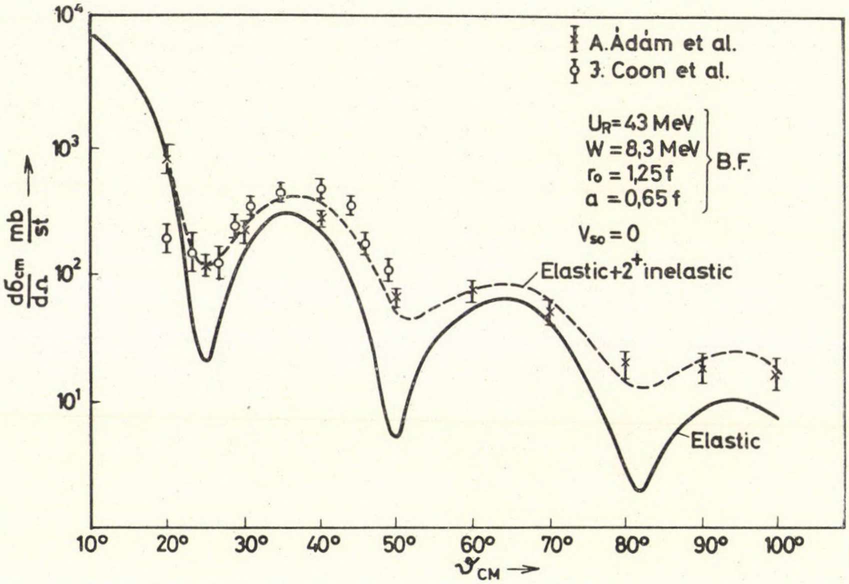


Fig. 6. Angular distribution (measured data, solid curve: predicted differential elastic scattering cross-section, broken line: the former plus the inelastic excitation to 2^+ level)

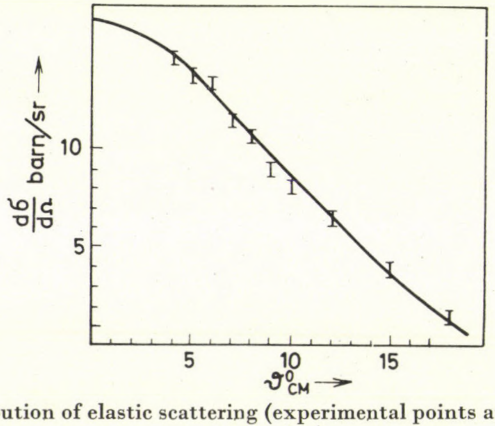


Fig. 7. Angular distribution of elastic scattering (experimental points and predicted differential cross-section)

In Fig. 7 the calculated and measured values of the differential cross-section at small angles are shown. The agreement is fairly good.

Discussion

It can be concluded, at least at neutron energies of 14 MeV, that the small angle anomaly actually does not appear and it seems that the neutron scattering by uranium can be well described by using the optical model if one takes into account the possibility of an appreciable inelastic contribution to the experimental data. A similar analysis was performed simultaneously with the present experiments by G. V. GORLOV et al. [20] for neutron energies $E_n = 4$ MeV which similarly did not show any increased small angle scattering compared with the predictions of the optical model.

Finally, it is of interest to note that a comparison of ELWYN's data with BALDONI's theoretical curve for the differential scattering curve suggests that the small angle anomaly results from a direct inelastic contribution.

REFERENCES

1. Y. A. ALEKSANDROV, G. V. ANIKIN and A. S. SOLDATOV, Zh. Eksper. i Teor. Fiz., **40,** 1878, 1961.
   Y. A. ALEKSANDROV, Zh. Eksper. i Teor. Fiz., **33,** 294, 1957.
2. Y. V. DUKAREVICH and A. N. DYUMIN, Zh. Eksper. i. Teor. Fiz., **44,** 130, 1963.
3. Y. A. ALEKSANDROV and I. I. BONDARENKO, Zh. Eksper. i Teor. Fiz., **31,** 726, 1956.
4. R. F. REDMOND, Phys. Rev., **140,** B. 1265, 1965; Proc. of Int. Conference Study of Nuclear Structure with Neutrons, Antwerpen, 1965.
5. A. J. ELWYN, J. E. MONAHAN, R. O. LANE, A. LANGSDORF, Jr. and F. P. MOORING, Phys. Rev., **142,** 3758, 1966; Proc. of Int. Conference Study of Nuclear Structure with Neutrons, Antwerpen, 1965.
6. G. V. ANIKIN, Y. A. ALEKSANDROV and A. S. SOLDATOV, Proc. of Int. Conference Study of Nuclear Structure with Neutrons, Antwerpen, 1965.
7. V. S. BARASENKOV, B. M. BARABASKOV, Nucl. Phys., **9,** 426, 1958.
8. M. WALT and D. B. FOSSAN, Phys. Rev., **137,** B629, 1965.
9. J. SCHWINGER, Phys. Rev., **73,** 407, 1948.
10. G. BREIT, M. L. RUSTGI, Phys. Rev., **114,** 830, 1959.
11. P. HRASKÓ and Zs. KÖVESY, Acta Phys. Hung., **20,** 285, 1966.
12. A. ÁDÁM, G. PÁLLA and P. QUITTNER, Acta Phys. Hung., **20,** 227, 1966.
13. A. ÁDÁM, P. HRASKÓ and G. PÁLLA, Phys. Letters, **22,** 475, 1966.
14. A. ÁDÁM, F. DEÁK, P. HRASKÓ, A. KISS, Zs. KÖVESY, L. JÉKI and G. PÁLLA (to be published); G. PÁLLA, Summer School, Sárospatak, 1967.
15. N. AUSTERN and J. S. BLAIR, Annals of Physics, **33,** 1564, 1965; J. S. BLAIR, S. SHARP and L. WILETS, Phys. Rev., **125,** 1625, 1962.
16. J. H. COON and J. R. BEYSTER, Phys. Rev., **111,** 250, 1958.
17. A. ÁDÁM, GY. KOZMA, F. DEÁK, L. JÉKI, G. PÁLLA and L. SZABÓ (to be published).
18. F. BJORKLUND and S. FERNBACH, UCRL-report, 4926, 1957.
19. B. BALDONI and A. M. SARUIS, Il Nuovo Cimento, **33,** 1145, 1964.
20. G. V. GORLOV, H. S. LEBEDEVA and B. M. MOROSOV, Phys. Letters, Vol. **25B,** 197, 1967.

УПРУГОЕ РАССЕЯНИЕ НЕЙТРОНОВ С ЭНЕРГИЕЙ 14 МЭВ ПОД МАЛЫМИ УГЛАМИ НА ЯДРАХ U^{238}

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Ж. КЁВЭШИ и Г. ПАЛЛА

Резюме

Подытоживаются попытки истолкования различными авторами аномалии упругого рассеяния быстрых нейтронов под малыми углами на ядрах U^{238} [1]. Далее, проблема выдвигается с соображения, относительно чего и при каких предположениях данная аномалия появляется.

Измерялось угловое распределение упругого рассеяния нейтронов с энергией 14,7 Мэв. При измерении принимался во внимание неупругий вклад в результатах измерений, обусловленный разрешением по энергии. Анализируя оптическую модель рассеяния, малоугловая аномалия не обнаружена.

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REMARK TO THE INTERPRETATION OF THE KENNEDY—THORNDIKE EXPERIMENT

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(Received 29. II. 1968)

It is shown that the result of the Kennedy—Thorndike experiment can be understood in terms of the Lorentz contraction only and it is not necessary — as it is generally supposed — to make use of the relativistic behaviour of clocks for the interpretation of the result.

I

R. J. KENNEDY and E. M. THORNDIKE [1] carried out a remarkable modification of the Michelson—Morley experiment; the authors used an interferometer with arms of different lengths and showed that just like in the case of the original Michelson—Morley experiment no fringe shift occurs when the arrangement is turned round.

The experiment is one of the fundamental experiments supporting the theory of relativity. It seems, however, that the authors have come to partly incorrect conclusions regarding the theoretical implications which follow from the experiment. The arguments of KENNEDY and THORNDIKE have been generally accepted (see e.g. H. DINGLE [2]), therefore it seems worthwhile to re-analyse the experiment.

The Michelson—Morley experiment can be interpreted in terms of the Lorentz contraction, i.e. supposing if a solid is made to move with a velocity $\mathbf{v}$ relative to the carrier of light, then its dimensions parallel to $\mathbf{v}$ contract by a factor $\sqrt{1 - v^2/c^2}$. Alternatively, if a solid is moving with a velocity $\mathbf{v}$ relative to the carrier of light and is turned round, then it suffers deformations in such a manner, that the dimensions turned into the direction of $\mathbf{v}$ contract, while those turned away from the direction of $\mathbf{v}$ expand.

The claim is made — which in our opinion is incorrect — that the KENNEDY—THORNDIKE experiment cannot be fully interpreted in terms of the Lorentz contraction of arms of the interferometer alone, but it is claimed to be necessary to consider also the relativistic effects clocks suffer when accelerated.

To prove our point of view we show that the result of the experiment can be explained in terms of the Lorentz contraction of arms of the interferometer.

II

Consider a system of reference K with measures such that the propagation of light appears isotropic relative to K. Consider further two points A, B both moving with a constant velocity $\mathbf{v}$ relative to K. The coordinate vectors of the points are thus

$$\mathbf{r}_A(t) = \mathbf{r}_A + \mathbf{v}t\,, \qquad \mathbf{r}_B(t) = \mathbf{r}_B + \mathbf{v}t\,, \tag{1}$$

where $\mathbf{r}_A$, $\mathbf{r}_B$ are constant vectors. A signal of light starting from A at $t = 0$ will arrive in B at a time $t = t_1$ such that

$$\big(\mathbf{r}_B(t_1) - \mathbf{r}_A(0)\big)^2 = c^2 t_1^2\,. \tag{2}$$

Similarly a signal will travel from B to A in a time t_2 such that

$$\big(\mathbf{r}_A(t_2) - \mathbf{r}_B(0)\big)^2 = c^2 t_2^2\,. \tag{3}$$

Introducing (1) into (2) and (3) we find that

$$\left.\begin{aligned} & a^2 + 2\,\mathbf{av}t_1 - (c^2 - v^2)\,t_1^2 = 0 \\ & a^2 - 2\,\mathbf{av}t_2 - (c^2 - v^2)\,t_2^2 = 0 \\ \text{with} \qquad & \\ & \mathbf{a} = \mathbf{r}_B - \mathbf{r}_A\,. \end{aligned}\right\} \tag{4}$$

Solving equations (4) and remembering that $t_1, t_2 > 0$ we find

$$t_1 + t_2 = \frac{2\sqrt{(\mathbf{av})^2 + a^2(c^2 - v^2)}}{c^2 - v^2}$$

or putting

$$\mathbf{av} = av\cos\vartheta\,,$$

$$t_1 + t_2 = \frac{2\,ac}{c^2 - v^2}\sqrt{1 - \frac{v^2}{c^2}\sin^2\vartheta}\,. \tag{5}$$

Expression (5) gives the time a signal of light — or a surface of constant phase — takes for the to and return flight along a rod the ends of which are connected by the vector $\mathbf{a}$ and which is moving with a velocity $\mathbf{v}$.

Consider a rod with length a_0 when it is placed perpendicular to $\mathbf{v}$. Let us turn the rod until it stands with an angle ϑ inclined towards $\mathbf{v}$. Supposing its projection upon $\mathbf{v}$ contracts we find

$$a^2(\vartheta) = a_1^2 + a_2^2\left(1 - \frac{v^2}{c^2}\right),$$

where

$$a_1^2 + a_2^2 = a_0^2$$

and

$$a_1 = a(\vartheta) \sin \vartheta \,,$$

thus a_1 is the component of **a** perpendicular to **v** (see Fig. 1). We find thus

$$a^2(\vartheta) = a^2(\vartheta) \sin^2 \vartheta + \left(a_0^2 - a^2(\vartheta) \sin^2 \vartheta\right) \left(1 - \frac{v^2}{c^2}\right)$$

or

$$a(\vartheta) = \frac{a_0 \sqrt{1 - \frac{v^2}{c^2}}}{\sqrt{1 - \frac{v^2}{c^2} \sin^2 \vartheta}} \,. \tag{6}$$

Replacing a by $a(\vartheta)$ in (5) we find

$$t_1 + t_2 = \frac{2\, a_0}{\sqrt{c^2 - v^2}} \,. \tag{7}$$

We conclude thus as follows. Consider a rod of length a_0 when orientated in a direction perpendicular to **v**. If the rod is turned so that its direction stands at an angle ϑ to **v** its length changes to $a(\vartheta)$ as given in (6), provided it suffers the ordinary Lorentz contraction.

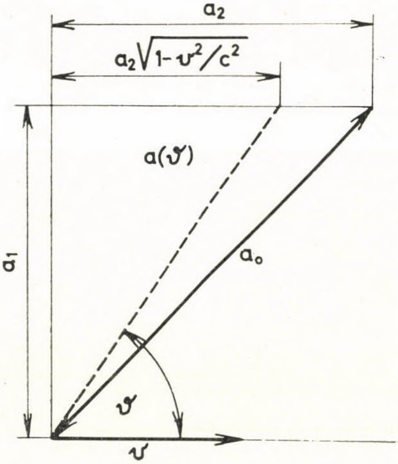

Fig. 1

From (7) it follows therefore that turning round a solid rod the time $t_1 + t_2$ a light signal takes to move along the rod and back again remains the same independent of the orientation of the rod.

III

Using a Michelson type of interferometer the observed interference pattern is determined by the difference ΔT of phase in which the two coherent beams arrive. Writing

$$\Delta T = (t_1^{(1)} + t_2^{(1)}) - (t_1^{(2)} + t_2^{(2)}) ,$$

where $t_1^{(1)} + t_2^{(1)}$ is the time the phase fronts travel to and fro along the one arm, while $t_1^{(2)} + t_2^{(2)}$ is the time the to and fro travel takes along the other arm.

Turning round the interferometer it follows from (7) that neither $t_1^{(1)} + t_2^{(1)}$ nor $t_1^{(2)} + t_2^{(2)}$ changes its value, therefore ΔT remains constant as was established by experiment. For the above consideration it is immaterial whether the length of the arms of the interferometer are equal or not. Thus we conclude that *the result of the Kennedy—Thorndike experiment can be understood in terms of the Lorentz contractions of the arms of the interferometer.*

REFERENCES

1. R. J. Kennedy and E. M. Thorndike, Rev. Phys., **42,** 400, 1932.
2. H. Dingle, "The special Theory of Relativity". London—New York, Methuen-Wiley, 1940, pp. 18—19.

ЗАМЕЧАНИЕ К ИСТОЛКОВАНИЮ ЭКСПЕРИМЕНТА КЕННЕДИ–СОРНДАЙК

Л. ЯНОШИ

Резюме

Показывается, что результат эксперимента Кеннеди—Сорндайк можно интерпретировать только на основе сокращения Лоренца и не является необходимым — как это обычно предполагается — использовать релятивистическое поведение часов при истолковании результата.

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PRECIPITATION OF FISSION PRODUCTS FROM THE ATMOSPHERE IN DEBRECEN, HUNGARY, DURING 1966 AND 1967

By

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(Received 28. III. 1968)

Results of the measurements relating to the beta activity of fission products in atmospheric precipitation during 1966 and 1967 are discussed. It can be ascertained from the summarized data of recent years that the fission product content of the precipitation decreased rapidly with an average decontamination half period of 0.7 year. With respect to the quarterly summarized data of the last two years a nearly constant activity level can be observed which is due to sporadic atmospheric nuclear tests.

Fresh fission products with a short half period (originating from the Chinese nuclear test of Oct. 27, 1966) were detected in the precipitation by means of the gamma spectrometric method. Be-7 produced by cosmic rays is separated chemically from the precipitation and the Be-7 activity of the precipitation is determined by a gamma spectrometer. It amounts to 88 dpm/litre.

Introduction

Beta-activity of fission products in the atmospheric precipitation has been measured systematically in this Institute since 1952 [1—8].

The precipitation is collected daily by means of an ombrometer with a 40 cm diameter PVC funnel. In the absence of precipitation the deposited dust is washed into the collecting vessel and measured together with the next precipitation. In this manner practically the total fallout is collected because 90% of the fission product content of the troposphere is carried down by precipitation [9]. The water, dust and soot content of the ombrometer is concentrated by evaporation into standardized glass cups of 25 mm diameter. The beta-activity is measured by an end-window GM counter. A detailed description of the techniques is given in our preceding papers [4].

Results of measurements

Fig. 1 demonstrates the results of the measurements for the years 1966 and 1967 in the same form as in our previous papers. Determined beta-activities are plotted on the ordinate upwards in cpm and 10^{-11} Curie units as a function of calendar time. The amount of daily precipitation is shown on the ordinate downwards in mm and ml units. All data are reduced to $1/50\ m^2$

surface of the standard meteorological ombrometer. The type of precipitation is denoted by the conventional meteorological key. Data of some published nuclear tests are marked but without responsibility for the completeness and exactness of such information gathered from various public sources (press, radio, etc.).

It is preferable to summarize the fallout activities for longer periods (one month or year) because of the statistical nature of precipitation in time and place. In this way we get a more realistic mean value for comparison.

Table I

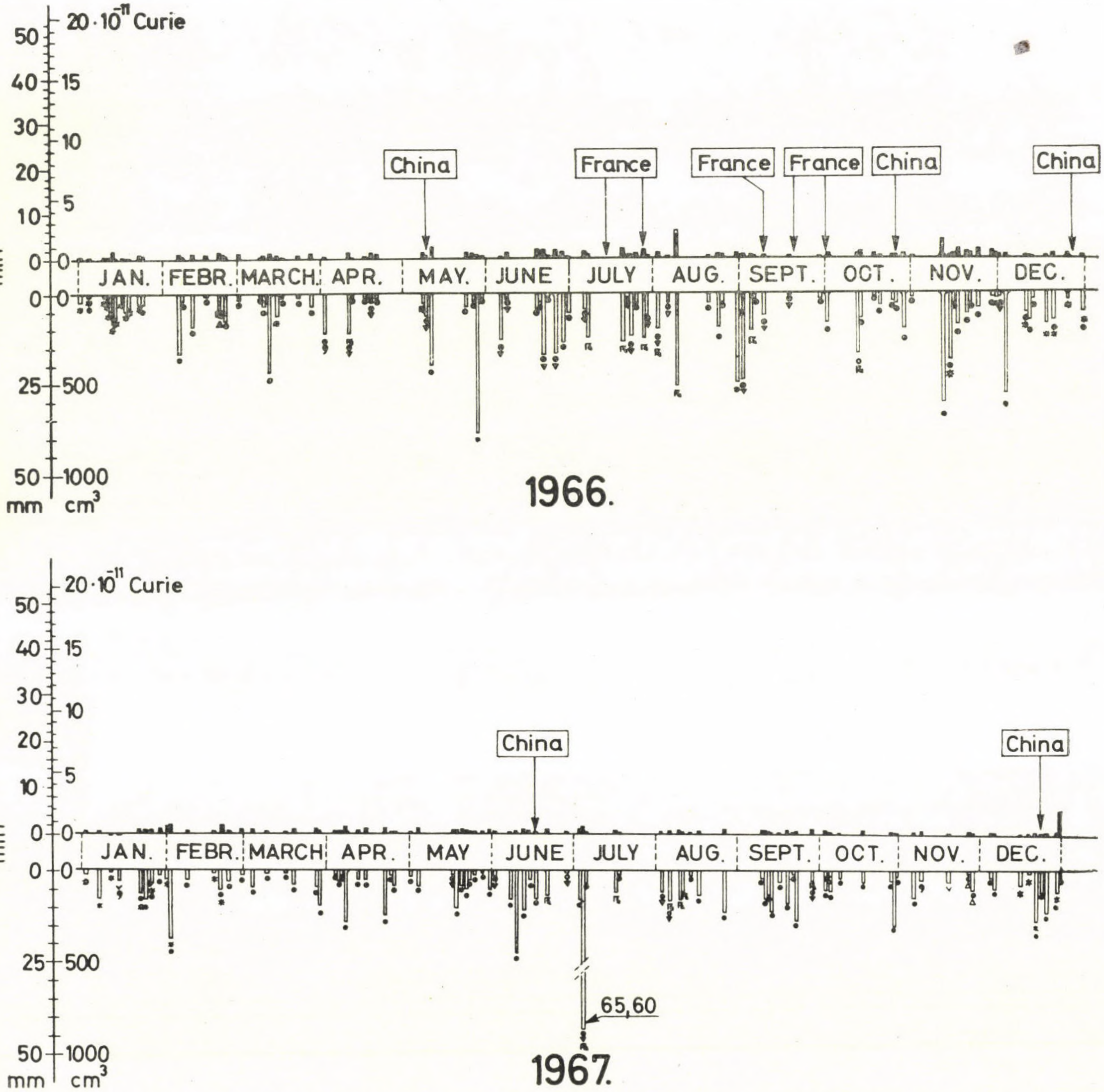
Monthly and yearly sum of beta activity measured in the precipitation in Debrecen

Month	1966	1967
	mCi/km²	mCi/km²
January	0.546	0.642
February	0.735	1.32
March	0.701	0.684
April	0.534	1.014
May	1.311	0.897
June	1.509	0.483
July	1.242	0.684
August	1.53	0.534
September	0.402	0.609
October	0.546	0.399
November	2.334	0.309
December	0.456	1.299
Yearly sum	11.846	8.874

In Table I the monthly and yearly sums of beta-activity are shown in mCi/km² units, measured about 48 hours after precipitation.

Some comparatively high values in this Table can be correlated with the Chinese nuclear tests on May 9, 1966; October 27, 1966; Dec. 28, 1966 and Dec. 24, 1967. The increased activities appeared in the precipitation usually with 10—30 days delay, in some cases with 7 days delay. Effects of the June 17, 1967 Chinese test and the five French tests in 1966 were not significant. According to the press this was the first Chinese thermonuclear test. The fact that this test was not observable in tropospheric fallout in Debrecen demonstrated that the fission products emerged into the stratosphere. The five French tests were carried out in the South Pacific Ocean. Nuclear tests in the southern hemisphere are usually not observable in the northern hemisphere.

The variation in the average specific activity of the precipitation verifies our statements above. In Table II the monthly average specific activity is shown. (The values of the specific activity are generally not so characteristic as is the monthly sum of beta activity because the amount of precipitation



Note added in proof: In June 1967 there was a new French test series in the South Pacific Ocean.

Fig. 1. Beta activity of fission products in the atmospheric precipitation in Debrecen, Hungary, during 1966 and 1967. Ordinate upwards, right: activity in 10^{-11} Curie units, corrected for the geometry of the counting equipment. Ordinate upwards, left: activity observed in cpm, both reduced to 1/50 m^2 ombrometer surface. Ordinate downwards, right: one day's rainfall in ml water volume collected by an exposed area of 1/50 m^2. Ordinate downwards left: one day's rainfall in mm. Abscissa: calendar time

fluctuates and the specific activity — especially when the amount of precipitation is small — depends strongly on the amount of precipitation.)

If the data of the last two years are compared with those for previous years it can be stated that the yearly sum of beta activity increased in 1961—1962 according to the large scale nuclear tests, and decreased continually after a maximum in the year 1962 (Fig. 2 and Fig. 3). Fig. 2 demonstrates the yearly sum of beta activity measured in the precipitation between 1960 and

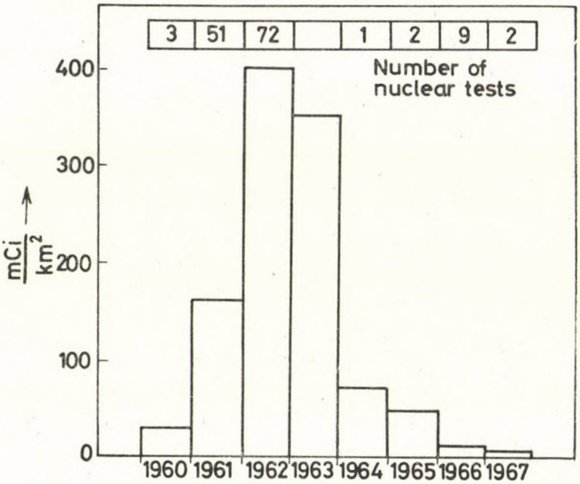


Fig. 2. Yearly sum of beta activity measured in the precipitation in Debrecen, Hungary between 1960 and 1967. Similar data between 1952 and 1960 are in [6]

Table II

Monthly average specific activity of precipitation in Debrecen between 1966 and 1967

Month	1966	1967
	pCi/ml	pCi/ml
January	0.0139	0.0174
February	0.0166	0.0403
March	0.0198	0.0282
April	0.0186	0.0241
May	0.0181	0.0251
June	0.0209	0.0082
July	0.0192	0.0083
August	0.0200	0.0118
September	0.0086	0.0112
October	0.0108	0.0120
November	0.0308	0.0174
December	0.0064	0.0265

1967, Fig. 3 shows the quarterly sums of the same data. A previous report [6] contains our results from the years 1952—1960 in a similar graphical summary.

From these data of the yearly decrease of activity an average decontamination half period of 0.7 ± 0.1 year was determined which is similar to the value stated earlier [6]. Recent sporadic nuclear tests caused an approximately constant level of activity in the last two years (see Fig. 3).

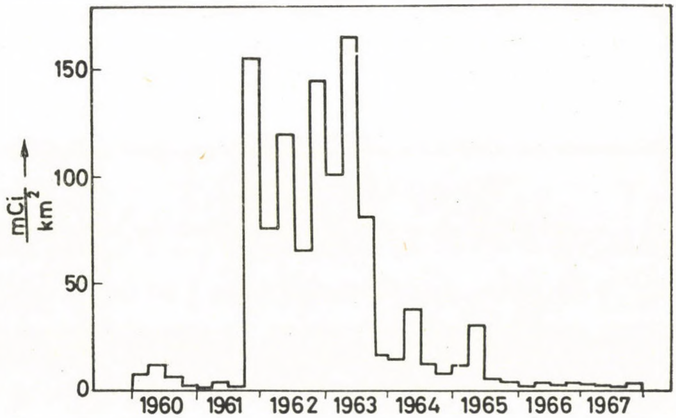


Fig. 3. Quarterly sum of beta activity measured in the precipitation in Debrecen, Hungary, between 1960 and 1967. Similar data between 1952 and 1960 are in [6]

Fig. 3 demonstrates strikingly how sharp the seasonal variation of the activity is in the years 1960, 1963, 1964 and 1965. These are the well-known spring maxima which are the consequences of the exchanges of air masses between stratosphere and troposphere. Tropospheric fallout of sporadic contemporary nuclear tests obscured this periodicity in 1961, 1962, 1966 and 1967.

Investigation of Be-7 activity of the precipitation

It is preferable to use a gamma-spectrometer for the analysis of the composition of fission products in the precipitation but in this case a larger amount of rainwater is required.

Between Oct. 29, and Nov. 17, 1966 171 l of precipitation were collected by means of a polyethylene foil with a surface area of about 10 m^2. A smaller fraction of the precipitation (25 l) was evaporated and the gamma spectrum of the sample was measured on the 115th, 143rd, 151st and 171st day after beginning the collection. Fig. 4 shows the result of the measurement on the 115th day. A NaI scintillator (5 cm × 3.2 cm) and a 400 channel TMC pulse height analyser were used for gamma counting.

The radioactive isotopes in the precipitation can be identified partly by the photopeaks in the gamma spectrum, partly by the half periods of the radioactive decay. Gamma emitting isotopes with a half life shorter than 14 days can be neglected because our first measurements started 117 days after the Chinese nuclear test of Oct. 27, 1966. The main constants of the gamma emitting isotopes ($T_{1/2} > 14$ days) of fission products are shown in Table III.

A comparison of the gamma spectrum in Fig. 4 with the data of Table III indicates unambiguously the presence of fission products having a half life shorter than 3 months. Certain isotopes are correlated to some photopeaks

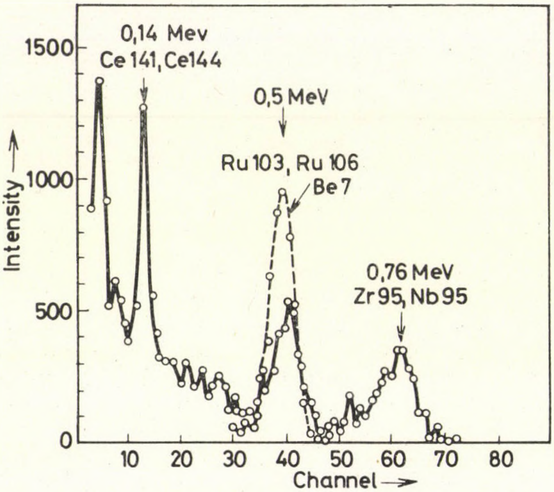


Fig. 4. Gamma ray scintillation spectra of radioactive content of the precipitation collected between Oct. 29, and Nov. 17, 1966. Curve 1 gained from 25 l precipitation, shows the presence of fresh fission products ^{141}Ce, ^{103}Ru, ^{95}Zr, ^{95}Nb originating from the Chinese nuclear test on Oct. 27, 1966. Curve 2 shows the photopeak of Be—7 due to cosmic rays. This sample was prepared by chemical separation of Be—7 from 146 l of the same precipitation

Table III

Gamma emitting isotopes ($T_{1/2} > 14$ days) of fission products

	Yield % [10]	Half period	E_γ MeV
Zr 95	6.2	65 d	0.72; 0.76
Nb 95	6.2	35 d	0.77
Ru 103	3.0	40 d	0.5
Ru 106	0.38	1.0 a	0.51; 0.62
Sb 125	0.02	2.7 a	0.43; 0.6
Cs 137	6.15	30 a	0.66
Ce 141	6.0	32.5 d	0.14
Ce 144	6.0	285 d	0.13

in Fig. 4. These identifications are confirmed by the half periods. The measured half periods are: 36 days for the 0.14 MeV photopeak, 50.5 days for the 0.5 MeV photopeak and 98 days for the 0.76 MeV photopeak, belonging to ^{131}Ce, ^{104}Ru, ^{95}Zr isotopes.

The larger part of the collected rainwater (146 l) was used to determine the Be-7 content of the precipitation. Be-7 is produced by cosmic rays through spallation of atmospheric O_2 and N_2 molecules. Be-7 is precipitated in the form of beryllium oxide adhering to atmospheric dust particles. The Be-7 decays by electron capture with a half life of 53 days, emitting gamma rays of 0.479 MeV with a branching ratio of 12%.

There is another possible source of atmospheric Be-7 from the Li-6 content of a thermonuclear device [15].

The gamma energy and the half period of Be-7 are close to the characteristic features of the gamma rays of ^{103}Ru, so chemical separation is required for the determination of the Be-7 content of the precipitation. The procedure used in [11] was adopted for the separation but in a modified form because purification was necessary only from the heavy metal Ru isotopes.

After chemical separation, the gamma spectrum was measured. The Be-7 photopeak was present at 0.48 MeV (Fig. 4. curve *b*). The intensity was measured on the 151st, 170th and 181st days after the beginning of the collection of the precipitation. The results were evaluated by means of the least square method and amounted to 52.5 days for the half period. These data prove the presence of Be-7.

Using the gamma spectrum, the Be-7 activity of the precipitation was determined and corrected to the datum of the collection of the rainwater and an activity of 88 dpm/litre was obtained. This result is in good agreement with the data measured by several authors [12—14] amounting to 10—120 dpm/litre. This Be-7 activity is explained by cosmic rays origin and there is no reason to assume a thermonuclear origin.

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REFERENCES

1. A. Szalay and D. Berényi sen., Acta Phys. Hung., **5**, 1, 1955.
2. A. Szalay and D. Berényi sen., MTA III. Oszt. Közl., **5**, 89, 1955.
3. A. Szalay and D. Berényi sen., MTA III. Oszt. Közl., **9**, 175, 1959.
4. A. Szalay and D. Berényi sen., Proc. 2nd Genova Conf., **18**, 570, 1958.
5. A. Kovách and A. Szalay, ATOMKI Közl., **2**, 229, 1960.
6. A. Szalay and A. Kovách, Acta Phys. Hung., **13**, 281, 1961.
7. A. Szalay and A. Kovách, Acta Phys. Hung., **16**, 321, 1964.
8. A. Szalay and A. Kovách, Acta Phys. Hung., **23**, 137, 1967.
9. R. Björnerstedt and K. Edvarson, Ann. Rev. Nucl. Sci., **13**, 505, 1963.
10. S. Katcoff, Nucleonics, **18**, 201, 1960.

11. J. A. **Arnold** and H. A. **Al-Salih**, Science, **121**, 451, 1955.
12. D. H. **Peirson**, J. Geophys. Res., **68**, 3831, 1963.
13. G. **Schumann** and M. **Stoeppler**, J. Geophys. Res., **68**, 3827, 1963.
14. Y. **Miyake** and Y. **Ohtsuka**, Papers in Met. Geophys., **15**, 89, 1964.
15. O. C. **Allkofer**, J. M. **Fox**, M. **Simon**, Atomkernenergie, **11**, 515, 1966.

ПРОДУКТЫ ДЕЛЕНИЯ В АТМОСФЕРНЫХ ОСАДКАХ В Г. ДЕБРЕЦЕН (ВЕНГРИЯ) В 1966 И 1967 ГГ.

А. САЛАИ и Е. ЧОНГОР

Резюме

Обсуждаются результаты измерений относительно бета-активности продуктов деления в атмосферных осадках в 1966 и 1967 гг. На основании суммированных данных последних лет устанавливается, что количество продуктов деления в осадках быстро уменьшилось со средним периодом полувыпадения 0,7 года. Что касается суммированных данных по кварталам двух последних лет, наблюдался близко постоянный уровень активности, обусловленный единичными атмосферными ядерными испытаниями.

Свежие продукты деления с коротким периодом полураспада (происходящие от ядерного испытания в Китае в 27 октября 1966 г.) были обнаружены в осадках с помощью гамма-спектрометрического метода. Be-7, созданный космическим излучением, отделялся химически от осадков. Активность Be-7 осадков определялась гамма-спектрометром. Для данной активности получено значение 88 распадов в минуту на 1 литр.

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ENERGY TRANSFER PROCESSES IN LUMINESCENT MIXED SOLUTIONS

By

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A relation between fluorescence spectra of mixed solutions and those of the component solutions is given, by which the radiative energy transfer is taken into account more precisely than by earlier formulas. In this relation the resonance transfer of energy is characterized only by two quantities; by using the latter as well as fluorescence yields and decay times of the components, the probability of molecular processes connected with emission and energy transfer is determined. Experimental results are in good agreement with **Jabłonski**'s, **Förster**'s and **Ketskeméty**'s investigations and support their theories concerning the mechanism of energy transfer. **Rozman**'s method is also discussed and some corrections are suggested.

Since **Franck** and **Cario** [1] first observed sensitized fluorescence, numerous investigators have tried to clarify the mechanism of intermolecular energy transfer. Earlier relations based on phenomenological considerations were difficult to apply because of the relatively numerous parameters involved; furthermore, most of these theories (e.g. [2—5]) account only for resonance transfer of energy. Relatively fewer investigations take radiative energy transfer also into account and so allow, as does the present paper, a theoretically more precise control of the different theories and a verification of their results (e.g. [6—8]).

I. Radiative energy transfer in mixed solutions

If the emission spectra and absorption spectra of luminescent solutions overlap, the primary luminescent light will excite secondary luminescence, which, in turn, excites tertiary luminescence, and so on, the intensity of these depending on the overlap of the spectra, on the wavelengths of the exciting light and of the luminescent light observed, on the layer thickness of the samples, and on other experimental conditions. In mixed solutions the overlap of absorption and emission spectra is considerable and therefore the effect of secondary, tertiary, etc. fluorescent light produced by radiative energy transfer can be very important.

By generalizing the phenomenological theory concerning radiative energy transfer in solutions of a single component [9], and taking into account only secondary fluorescence, **Ketskeméty** [6] found the following connection between the directly measured fluorescence spectrum $B(\lambda')$ of the mixed solution and the true, normalized fluorescence quantum-spectra $f_1(\lambda'), f_2(\lambda)$ of the solutions of the components:

$$\begin{gathered} B(\lambda')/C(\lambda,\lambda') = [(1+\varkappa_{11})\,\eta_1'(\lambda) + \varkappa_{21}\,\eta_2'(\lambda)]\,f_1(\lambda') + \\ + [(1+\varkappa_{22})\,\eta_2'(\lambda) + \varkappa_{12}\,\eta_1'(\lambda)]\,f_2(\lambda')\,, \\ C(\lambda,\lambda') = \frac{\varrho}{4\pi n^2} E_{\lambda 0} \frac{\alpha}{\alpha+\beta}\left(1 - e^{-(\alpha+\beta)}\right), \end{gathered} \tag{1}$$

where λ and λ' are the wavelengths of the exciting light and of the observed luminescent light, respectively, $\eta_i'(\lambda)$ (or $\eta_{i\lambda}'$) means the apparent yield of the ith component of the mixed solutions, *i.e.* the quotient of the number of photons emitted by the ith substance contained in a unit volume of the mixed solution and of those absorbed by the mixed solution in the same time interval, $k_i(\lambda)$ and $k(\lambda)$ are the absorption coefficients of the ith substance and the mixed solution, respectively, $k(\lambda) = k_1(\lambda) + k_2(\lambda)$, $\alpha = k(\lambda)\,l$, $\beta = k(\lambda')\,l$, l being the layer thickness of the solution, $E_{\lambda 0}$ the quantum density of the exciting light,[1] n the refractive index of the solution, ϱ and $\varkappa_{ik}$ quantities taking into account losses by reflection and radiative energy transfer,[2] respectively. $\varkappa_{ik}$ can be determined from the relation

$$\varkappa_{ik} = \int_0^\infty \eta_k'(\lambda'')\,f_i(\lambda'')\,M(\lambda'')\,d\lambda'' \qquad (i,k = 1,2)\,; \tag{2}$$

or the definition of the function $M(\lambda'')$ see Eq. (26) in [9][3] ·

In mixed solutions of low concentration

$$\eta_i'(\lambda) = \eta_{0i}'(\lambda) \equiv \eta_i(\lambda)\,k_i(\lambda)/k(\lambda), \tag{2a}$$

where $\eta_i(\lambda)$ (or $\eta_{i\lambda}$) is the absolute quantum yield of the ith substance measured at the wavelength λ (see Eq. (8,14) in [13]). For solutions of higher concentrations, the spectra $B(\lambda')/C(\lambda, \lambda')$ calculated on the basis of Eqs. (1) and (2) using

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[1] The above quantities are expressed in the following units:

$$B(\lambda') : \frac{\text{number of photons}}{\text{cm}^2\,\text{sec sterad}}\,; \qquad f_i(\lambda') : \frac{\text{number of photons}}{\text{nm}}\,;$$

$$k(\lambda) : \text{cm}^{-1};\ l\text{: cm};\ \lambda\text{: nm}; \qquad E_{\lambda 0} : \frac{\text{number of photons}}{\text{cm}^2\ \text{sec}}\,;$$

the others being dimensionless numbers.

[2] The physical meaning of the quotient $\varkappa_{ik} f_k(\lambda')/f_i(\lambda')$ is the observed intensity of the secondary fluorescence excited in the kth substance by the primary fluorescence of the ith substance, divided by the observed intensity of the primary fluorescence of the ith substance [6].

[3] Detailed tables concerning the function $M(\lambda'')$ can be found in [10].

the relation $\eta'_i(\lambda) = \eta'_{0i}(\lambda)$ considerably differ from the measured spectra (see Fig. 1); this fact, the spectra of the components being practically independent from concentration, can only be explained by supposing that the apparent yield functions are dependent on concentration. This dependence, as can be seen from the following, is to be ascribed to, and explained by, resonance transfer of energy.

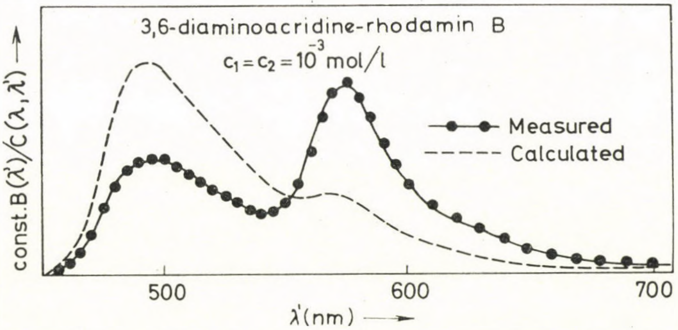

Fig. 1

II. Non-radiative energy transfer in mixed solutions

In taking into account the effect of the resonance transfer of energy, let us start from considerations given in [6], which, with some generalizations, may be summarized as follows.

Let n_i mean the number of the excited molecules of the ith substance in unit volume of the solution; a_i, b_i the probabilities of the spontaneous emission and the inner quenching of an excited molecule of the ith substance, respectively; w_k the probability of its quenching by a molecule of the kth substance, and a_k the probability of nonradiative energy transfer to a molecule of the kth substance. When exciting by light of wavelength λ and quantum density $E_{\lambda 0}$, the number of molecules excited by absorbed photons in a volume element $\mathrm{d}V$ during the time interval $\mathrm{d}t$ will be $E_{\lambda 0}\, \eta^*_{i\lambda}\, k_{i\lambda}\, \mathrm{d}V\, \mathrm{d}t$; $\eta^*_{i\lambda}$ is the probability of a molecule getting into the excited state by absorption of a photon of wavelength λ. In the case of steady state excitation the following relations will hold:

$$\begin{aligned}(E_{\lambda 0}\, \eta^*_{1\lambda}\, k_{1\lambda} + a_1 n_2 - s_1 n_1 - a_2 n_1 - w_2 n_1 - b_1 n_1)\, \mathrm{d}V\, \mathrm{d}t = 0\,,\\ (E_{\lambda 0}\, \eta^*_{2\lambda}\, k_{2\lambda} + a_2 n_1 - s_2 n_2 - a_1 n_2 - w_1 n_2 - b_2 n_2)\, \mathrm{d}V\, \mathrm{d}t = 0\,.\end{aligned} \tag{3}$$

During the time interval $\mathrm{d}t$, the ith substance in a volume element $\mathrm{d}V$ emits $s_i\, n_i\, \mathrm{d}V\, \mathrm{d}t$ photons, and $E_{\lambda 0}\, k_\lambda\, \mathrm{d}V\, \mathrm{d}t$ photons will be absorbed in $\mathrm{d}V$ in the same time. Substituting the values n_i expressed from Eq. (3) into the quotient of the number of emitted and absorbed photons, this quotient gives $\eta'_i(\lambda)$ according to the definition of the apparent yield. Thus, the following relations between $\eta'_i(\lambda)$ and the probabilities s_i, b_i, w_i, a_i can be obtained:

$$\begin{aligned}\eta'_{1\lambda} &= \frac{s_1 n_1}{E_{\lambda 0} k_\lambda} = \eta^*_{1\lambda} \frac{k_{1\lambda}}{k_\lambda} \frac{s_1 e_2}{e_1 e_2 - a_1 a_2} + \eta^*_{2\lambda} \frac{k_{2\lambda}}{k_\lambda} \frac{s_1 a_1}{e_1 e_2 - a_1 a_2}\,,\\ \eta'_{2\lambda} &= \frac{s_2 n_2}{E_{\lambda 0} k_\lambda} = \eta^*_{1\lambda} \frac{k_{1\lambda}}{k_\lambda} \frac{s_2 a_2}{e_1 e_2 - a_1 a_2} + \eta^*_{2\lambda} \frac{k_{2\lambda}}{k_\lambda} \frac{s_2 e_1}{e_1 e_2 - a_1 a_2}\,,\end{aligned} \tag{4}$$

where

$$e_i = s_i + b_i + a_k + w_k \qquad (i, k = 1, 2\,;\ i \neq k)\,.$$

If the concentration of one of the components (e.g. c_k) is equal to zero, then $\eta'_{i\lambda} = \eta_{i\lambda}$ and $a_k = w_k = 0$. In both limiting cases $c_1 = 0$ and $c_2 = 0$, $\eta^*_{i\lambda}$ can be expressed by the quantities $\eta_{i\lambda}$, s_i and b_i, and thus Eq. (4) obtains the following forms

$$\begin{aligned}\eta'_{1\lambda} &= \eta_{1\lambda}\frac{k_{1\lambda}}{k_\lambda}\,\frac{(s_1+b_1)\,e_2}{e_1\,e_2-a_1\,a_2}+\eta_{2\lambda}\frac{k_{2\lambda}}{k_\lambda}\,\frac{(s_2+b_2)\,s_1\,a_1}{e_i\,e_2-a_1\,a_2},\\ \eta'_{2\lambda} &= \eta_{1\lambda}\frac{k_{1\lambda}}{k_\lambda}\,\frac{(s_1+b_1)\,s_2\,a_2}{e_1\,e_2-a_1 a_2}+\eta_{2\lambda}\frac{k_{2\lambda}}{k_\lambda}\,\frac{(s_2+b_2)\,e_1}{e_1\,e_2-a\,a_2},\end{aligned} \tag{5}$$

with new symbols

$$\begin{aligned}\eta'_1(\lambda) &= \eta_{1\lambda}\frac{k_{1\lambda}}{k_\lambda}S_1+\eta_{2\lambda}\frac{k_{2\lambda}}{k_\lambda}A_1,\\ \eta'_{2\lambda} &= \eta_{1\lambda}\frac{k_{1\lambda}}{k_\lambda}A_2+\eta_{2\lambda}\frac{k_{2\lambda}}{k_\lambda}S_2,\end{aligned} \tag{6}$$

where the meaning of S_i and A_i is clear from the comparison of Eqs. (5) and (6). In mixed solutions of very low concentration $a_i = w_i = 0$, consequently $S_i = 1$, $A_i = 0$. Only in this case the value of $\eta'_i(\lambda)$ resulting from Eq. (6) will be equal to $\eta'_{0i}(\lambda)$ defined in Eq. (2a), as is easily seen. Eq. (6), with the symbol $\eta'_{0i}(\lambda)$, can be written as follows:

$$\begin{aligned}\eta'_1(\lambda) &= S_1\,\eta'_{01}(\lambda)+A_1\,\eta'_{02}(\lambda),\\ \eta'_2(\lambda) &= A_2\,\eta'_{01}(\lambda)+S_2\,\eta'_{02}(\lambda).\end{aligned} \tag{7}$$

Using the apparent yields η'_{0i}, which also take into account non-radiative energy transfer, to calculate $\varkappa_{ik}$ according to Eq. (2), we have

$$\begin{aligned}\varkappa_{11} &= S_1\,\varkappa^\circ_{11}+A_1\,\varkappa^\circ_{12}, & \varkappa_{12} &= A_2\,\varkappa^\circ_{11}+S_2\,\varkappa^\circ_{12},\\ \varkappa_{21} &= S_1\,\varkappa^\circ_{21}+A_1\,\varkappa^\circ_{22}, & \varkappa_{22} &= A_2\,\varkappa^\circ_{21}+S_2\,\varkappa^\circ_{22},\end{aligned} \tag{8}$$

$$\varkappa^\circ_{ik} = \int_0^\infty \eta'_{0k}(\lambda'')f_i(\lambda'')\,M(\lambda'')\,\mathrm{d}\lambda''.$$

By substituting the quantities η'_i and $\varkappa_{ik}$ given in Eqs. (7) and (8) into Eq. (1), we obtain a relation between the directly observed quantum density $B(\lambda')$ of the fluorescence of the mixed solution and fluorescence spectra of the components.

This relation, which takes into account both radiative and non-radiative energy transfer, becomes considerably less complicated if there is no overlap between the absorption spectrum $k_1(\lambda)$ of substance 1 and the emission spectrum $f_2(\lambda)$ of substance 2, which frequently occurs. In this case $A_1 = 0$, $S_2 = 1$ and $\varkappa_{21} = 0$. The accuracy of this simpler expression of $B(\lambda')$ can be increased by also taking into account excited centres of higher order. Considering that the intensities of primary, secondary, etc. fluorescences decrease according to a geometrical progression, generally with good approximation [9], it is easy to obtain

$$\begin{aligned}B(\lambda')/C(\lambda,\lambda') \equiv B^*(\lambda') &= \frac{S\eta'_{01}(\lambda)}{1-S\varkappa^\circ_{11}}f_1(\lambda')+\\ &+\left[\frac{\eta'_{02}(\lambda)+A\eta'_{01}(\lambda)}{1-\varkappa_{22}}+\frac{S\eta'_{01}(\lambda)(\varkappa^\circ_{12}+A\varkappa^\circ_{11})}{(1-S\varkappa^\circ_{11})(1-\varkappa^\circ_{22})}\right]f_2(\lambda');\end{aligned} \tag{9}$$

with the symbols $S_1 = S$ and $A_2 = A$. These quantities have a clear physical meaning, $\eta_{1m}S$ and $A\eta_{1m}/\eta_{2m}$ being the probabilities for an excited molecule of the substance 1, to lose its excitation energy by spontaneous emission, and to transmit its energy to substance 2 by non-radiative transfer, respectively [6].

III. Determination of the quantities characterizing the non-radiative energy transfer on the basis of the emission spectra

With a knowledge of the emission spectrum $B(\lambda')$ of a mixed solution, relation Eq. (9) enables us to determine the quantities S and A characterizing the non-radiative energy transfer.

a) Determination of S

Let λ_1' be a wavelength of observation at which substance 2, the acceptor, shows no emission, *i.e.* $f_2(\lambda_1') = 0$; then we obtain from Eq. (9) the following expression for S:

$$S = \frac{B^*(\lambda_1')}{\eta_{01}'(\lambda)\, f_1(\lambda_1') + \varkappa_{11}^{\circ}\, B^*(\lambda_1')}\,, \tag{10}$$

in which all quantities can be determined experimentally. This method is, however, cumbersome to apply because of the relatively complicated measurement of the intensity $E_{\lambda 0}$ of the exciting light in the formula of $B^*(\lambda_1')$ (see [15]).

In the case of equimolar mixed solutions, the dependence of S on concentration can also be determined by measuring at the wavelength λ_1' the photocurrents $I_c(\lambda_1')$ produced by the fluorescent light of solutions of different concentration c, with constant intensity of excitation, and holding the product of concentration and layer thickness at a constant value. It is easy to see from Eq. (10), that the relation

$$I_c(\lambda_1') = C_1 \frac{S}{1 - S\varkappa_{11}^{\circ}} \tag{11}$$

holds for the intensities of these photocurrents. Let the lowest concentration applied, c_0, be so low that the non-radiative energy transfer may be neglected, i.e. $S = 1$. (The fulfilment of this condition can be controlled on the basis of Eq. (10).) By measuring the photocurrent $I_0(\lambda_1')$ at c_0, C_1 can be determined from Eq. (11), and with the symbol $I_c^*(\lambda_1') = I_c(\lambda_1')/I_0(\lambda_1')$ we obtain:

$$S = \frac{I_c^*(\lambda_1')}{1 - \varkappa_{11}^{\circ} + I_c^*(\lambda_1')\, \varkappa_{11}^{\circ}}\,. \tag{12}$$

In the case of non-equimolar mixed solutions, it is not possible to choose the layer thicknesses so that $\varkappa_{11}^{\circ}$ and the factor $\alpha(1 - e^{-(\alpha+\beta)})/(\alpha + \beta)$ in $C(\lambda, \lambda')$ can be held at a constant value for different concentrations of the acceptor. In this case, dividing the intensities of the photocurrents by the above factor and designating these quotients by $I_0(\lambda_1')$ and $I_c(\lambda_1')$ instead of the intensities, we obtain, on the basis of our considerations for equimolar solutions:

$$S = \frac{I_c^*(\lambda_1')}{1 - \varkappa_{11}^\circ(c_0) + I_c^*(\lambda_1')\, \varkappa_{11}^\circ(c)}\,, \tag{13}$$

where $\varkappa_{11}^\circ(c_0)$ and $\varkappa_{11}^\circ(c)$ are the values of $\varkappa_{11}^\circ$ corresponding to the acceptor concentrations c_0 and c, respectively.

b) Determination of A

Knowing S and the fluorescence characteristics of the solutions of the components, A can also be determined with the aid of Eq. (9). In order to determine exactly the energy of the exciting light absorbed by the sample, it is convenient to perform the measurements with greater layer thicknesses, which absorb the exciting light as far as possible; therefore we used the following experimental method, also expounded by ROZMAN et al. [7, 11], but using the relation Eq. (9) for the evaluation of the results.

Let the mixed solution be excited first with a wavelength λ_1, absorbed mainly by the donor (substance 1), then with a wavelength λ_2, absorbed by the acceptor (substance 2) alone, and the fluorescent light observed at a wavelength λ_0' for which the absorption coefficient of the solution — and therefore also β — is practically equal to zero, and the measured photocurrents $I_{\lambda_1}(\lambda_0')$ and $I_{\lambda_2}(\lambda_0')$ are predominantly due to the emission of substance 2. Then if the intensities of the fluorescence, excited with the two wavelengths mentioned, but with the same exciting energy, are designated by I_{21} and I_{22}, respectively, these can be determined from the relations

$$I_{21} = \frac{I_{\lambda_1}(\lambda_0')\, Q(\lambda_0')}{\left[\int_0^\infty I_e(\lambda) Q(\lambda)\, \mathrm{d}\lambda\right]_{\lambda_1}}\,, \quad I_{22} = \frac{I_{\lambda_2}(\lambda_0')\, Q(\lambda_0')}{\left[\int_0^\infty I_e(\lambda) Q(\lambda)\, \mathrm{d}\lambda\right]_{\lambda_2}}\,, \tag{14}$$

where $I_e(\lambda)$ means the photocurrent produced by the exciting light of bandwidth λ to $\lambda + d\,\lambda$, and $Q(\lambda_0')$ and $Q(\lambda)$ the sensibility of the photomultiplier for the corresponding wavelengths. Applying Eq. (9) to both exciting wavelengths, we obtain for A

$$A = \frac{\dfrac{1}{\alpha_{11}}\left(\dfrac{I_{21}}{I_{22}} \cdot \dfrac{1 - e^{-\alpha_1}}{1 - e^{-\alpha_2}} - \alpha_{21}\dfrac{\eta_{02}(\lambda_1)}{\eta_{02}(\lambda_2)}\right)\dfrac{\eta_{02}(\lambda_2)}{\eta_{01}(\lambda_1)} - S\left(\dfrac{1 - \varkappa_{22}^\circ(\alpha_2)}{1 - S\varkappa_{11}^\circ(\alpha_1)}\,\dfrac{f_1(\lambda_0')}{f_2(\lambda_0')} + \dfrac{\varkappa_{12}^\circ(\alpha_1)}{1 - S\varkappa_{11}^\circ(\alpha_1)}\right)}{1 + S\dfrac{\varkappa_{11}^\circ(\alpha_1)}{1 - S\varkappa_{11}^\circ(\alpha_1)}} \tag{15}$$

with the symbols $\alpha_1 = \alpha(\lambda_1)$, $\alpha_2 = \alpha(\lambda_2)$, $\alpha_{11} = k_1(\lambda_1)/k(\lambda_1)$, $\alpha_{21} = k_2(\lambda_1)/k(\lambda_1)$; in the functions $\varkappa^{\circ}_{ik}$, depending on α and β, only the dependence on α has been designated, in our case $\beta = 0$; furthermore, it is supposed that $\varkappa^{\circ}_{22}(\alpha_1) = \varkappa^{\circ}_{22}(\alpha_2)$, the dependence of $\varkappa^{\circ}_{22}$ on α being very slight for large values of α. It is to be emphasized, that in determining A from Eq. (15), the greatest care is necessary in measuring the fluorescence characteristics, particularly the yield values, because they exert a sensible influence on the value of A, especially if the difference in the numerator is small.

c) Determination of the probability of the elementary processes in the molecules

The definition of quantities S and A, which can be determined experimentally, gives two simple relations between these quantities and the probabilities s_1, s_2, b_1, b_2, a_2 and w_2 of the elementary processes. Using further the relations

$$\eta_{im} = \frac{s_i}{s_i + b_i} \quad \text{and} \quad \tau_i = \frac{1}{s_i + b_i}, \tag{16}$$

which give the maximum quantum yields of the solutions of the components η_{im} and their decay times [12, 13], we easily obtain from these six equations:

$$\begin{aligned} s_1 &= \frac{\eta_{1m}}{\tau_1}, \quad s_2 = \frac{\eta_{2m}}{\tau_2}, \quad b_1 = \frac{1-\eta_{1m}}{\tau_1}, \quad b_2 = \frac{1-\eta_{2m}}{\tau_2}, \\ a_2 &= \frac{1}{\tau_1}\frac{A}{S}\frac{\eta_{1m}}{\eta_{2m}}, \quad w_2 = \frac{1}{\tau_1}\left(\frac{1}{S} - \frac{A}{S}\frac{\eta_{1m}}{\eta_{2m}} - 1\right), \end{aligned} \tag{17}$$

which enable us to determine experimentally the quantities $s_1, \ldots, w_2$.

IV. Experimental

a) *Absorption spectra* have been measured with autocollimating grating spectrophotometers *Optica Milano*, type CF 4 and CF 4 DR.

b) For the measurement of *fluorescence spectra* an apparatus has been constructed, with which it is possible to eliminate the errors due to fluctuations in the exciting light and to obtain the conditions of excitation and observation on which the evaluation of the measurement is based (see e.g. [9]). High pressure Hg- and X-bulbs Osram type HBO 500 and XBO 450, respectively, were used as a light source, and the exciting band, which could be considered as approximately monochromatic, was selected by an interference filter or a monochromator. In order to eliminate errors of measurement due to fluctuations in the intensity of the exciting light, the following method was used. In the monochromator M resolving the luminescent light, a thin glass plate G was placed immediately behind the entrance slit (Fig. 2). This plate projected a small fraction I of the unresolved luminescent light by means of the mirror T and through the crossed filter F on the photomultiplier Ph_2 placed inside the monochromator.

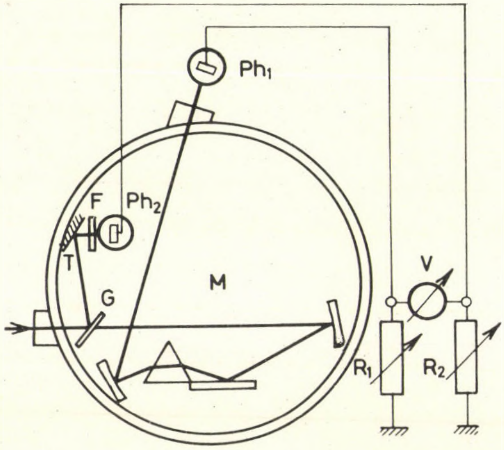

Fig. 2

whereas a fraction of the luminescent light passing the plate G fell on the multiplier Ph_1 o-known spectral sensitivity, the width $d\lambda'$ of this band of the luminescence spectrum $I(\lambda')$ being determined by the exit slit. If the resistances R_1 and R_2 of the resistors in the circuits of both multipliers are chosen so that the voltage drop on both is the same, then the quotient of the photocurrents will be equal to R_1/R_2 and independent from the intensity of the exciting light. Thus we obtain for the spectral distribution of the fluorescent light: $I(\lambda') = \text{const.}\, R_1/R_2$.

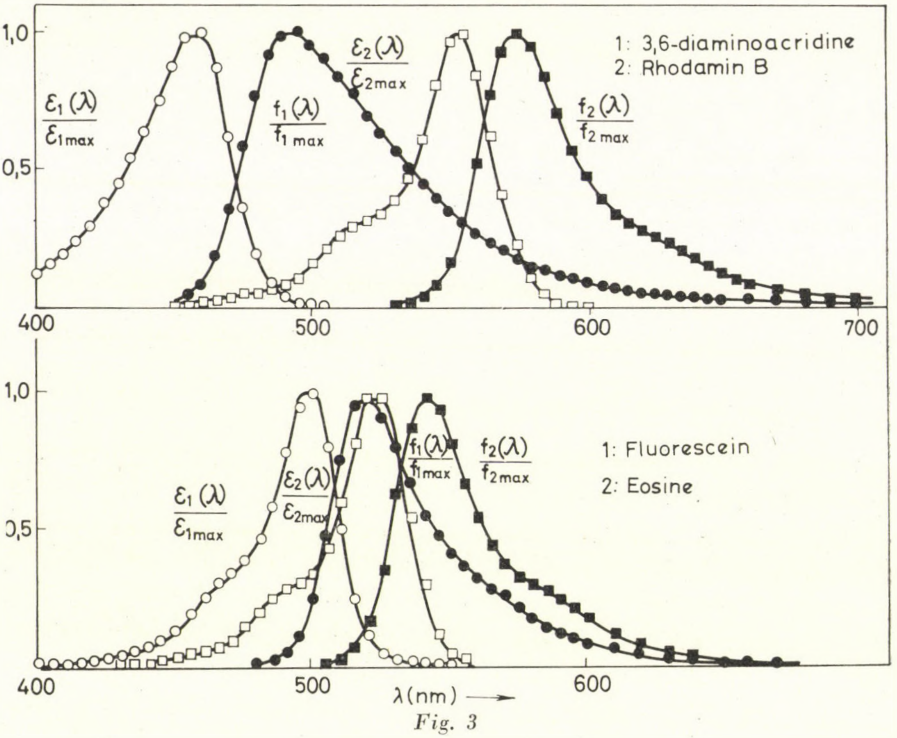

Fig. 3

The two-cells method described above, owing to the position of Ph_2 behind the entrance slit, not only also eliminates errors originating from fluctuations of the intensity of the exciting light, but also those due to the varying position of the arc projected on the sample. The linearity of the multipliers was checked separately.

c) *The true quantum yield* has been determined with our method published in [14] and [15], using the apparatus described above. In measuring the distribution of the exciting band the two cells-method has been used. The intensity of the exciting light scattered from a MgO plate and weakened by a rotating-sector disc and that of the luminescence emitted by the sample were measured with Ph_1 at wavelengths corresponding to the maxima of the exciting band and of the luminescence spectrum, respectively. The yield function $\eta(\lambda)$ was determined according to the method given in [16].

d) *The decay time* of fluorescent light was measured with a phase-fluorimeter built in our Institute, based essentially on the same principle as that described by BAUER and ROZWADOWSKI [17].

In determining the fluorescence characteristics from the measurements, the influence of secondary processes and those of higher order has always been taken into account; therefore the results obtained can be considered as true fluorescence characteristics.

e) *Dye-stuffs and solvents.* For our investigations we used mixed solutions of 3,6-diaminoacridine and rhodamin B, and of fluorescein and eosine, respectively, because the emission spectra $f_1(\lambda)$ of substance 1 and the absorption spectra $\varepsilon_2(\lambda)$ of substance 2 in these mixtures show a considerable overlap (Fig. 3). The dye-stuffs were purified by the usual chemical processes and the grade of purity checked by chromatography and absorption measurements. As solvent 96% ethanol with 1 mole/litre acetic acid was used for 3,6-diaminoacridine and rhodamin B; a mixture of 85% ethanol and 15% water with 10^{-2} mole/litre NaOH for fluorescein and eosine. Fluorescence characteristics of the components as well as of the mixed solutions were measured at the following concentrations: $1 \cdot 10^{-5}$, $2.5 \cdot 10^{-5}$, $5 \cdot 10^{-5}$, $1 \cdot 10^{-4}$, $2.5 \cdot 10^{-4}$, $5 \cdot 10^{-4}$, $1 \cdot 10^{-3}$, $2.5 \cdot 10^{-3}$ mole/litre. In non-equimolar mixed solutions the concentration of the acceptor varied between the above limits, the concentration of the donor being held at the constant value of 10^{-4} mole/litre.

V. Results and discussion

a) The absorption measurements demonstrated the validity of the Bouguer—Beer—Lambert law, both for the solutions of the components and the mixed solutions, in the concentration interval investigated. The additivity of the absorption coefficients $k_1(\lambda)$, $k_2(\lambda)$ of the solutions of the components and that of the mixed solutions, $k(\lambda) = k_1(\lambda) + k_2(\lambda)$, was well fulfilled, which shows that there was no chemical interaction between the components in the mixed solution.

b) The shape of the true fluorescence spectra of the four investigated substances, presented in Fig. 3, was independent of the concentration of the solutions. Emission spectra of equimolar and non-equimolar mixed solutions, however, showed a strong dependence on concentration. As examples, the $B^*(\lambda')$ spectra of non-equimolar solutions of 3,6-diaminoacridine and rhodamin B and those of fluorescein and eosine are presented in Figs. 4 and 5, respectively.

c) The relative quantum yields $\eta(\lambda)/\eta_{\max}$ of the solutions of the components and their absolute quantum yields measured at the exciting wavelengths λ_e used in the measurements of the spectra $B(\lambda')$ are given in Fig. 6 and Table I.

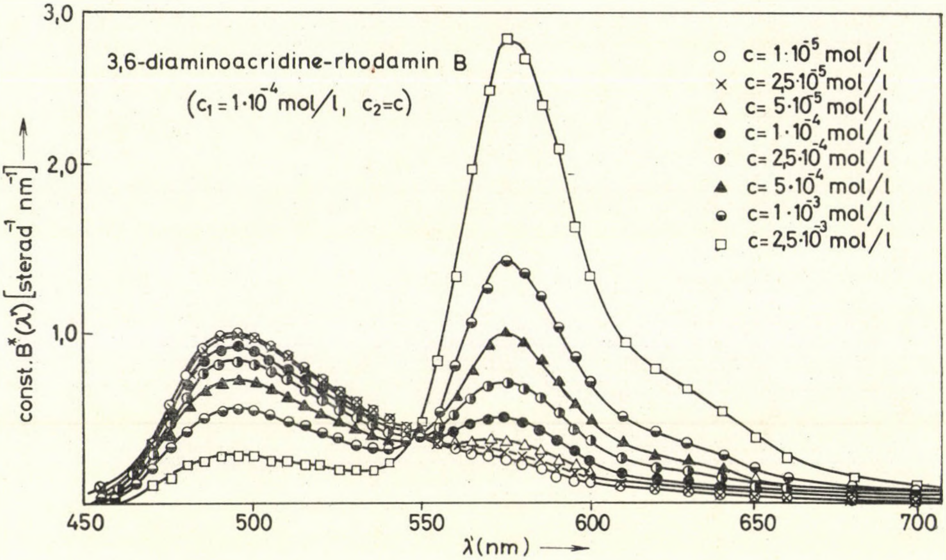

Fig. 4

d) The fluorescence characteristics of the components mentioned above being known, we determined the quantities $\varkappa_{ik}$, which take into account the radiative energy transfer. If the condition $[\beta(\lambda')]_{\max} = 1$ is fulfilled, $\varkappa_{ik}$ is

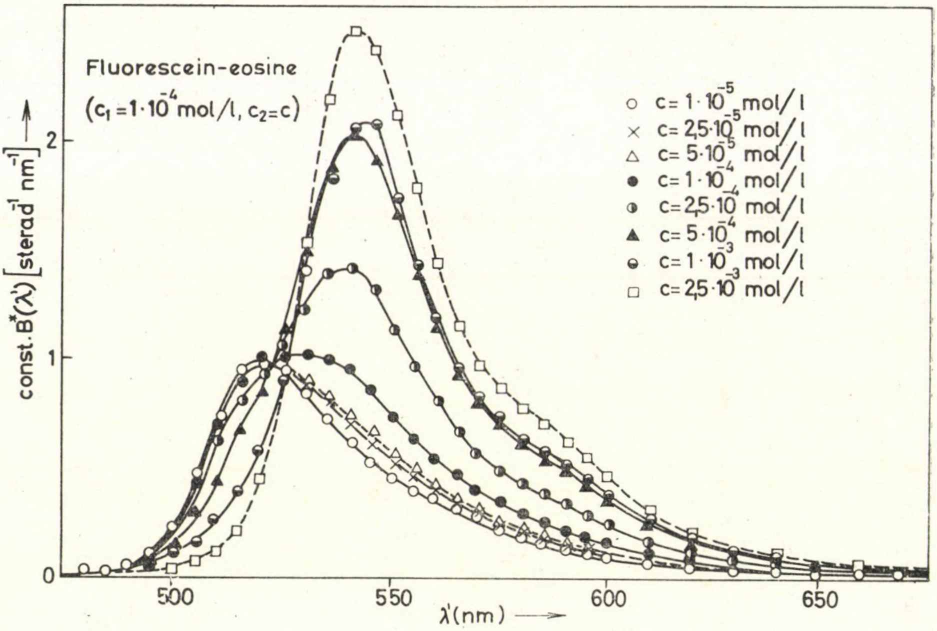

Fig. 5

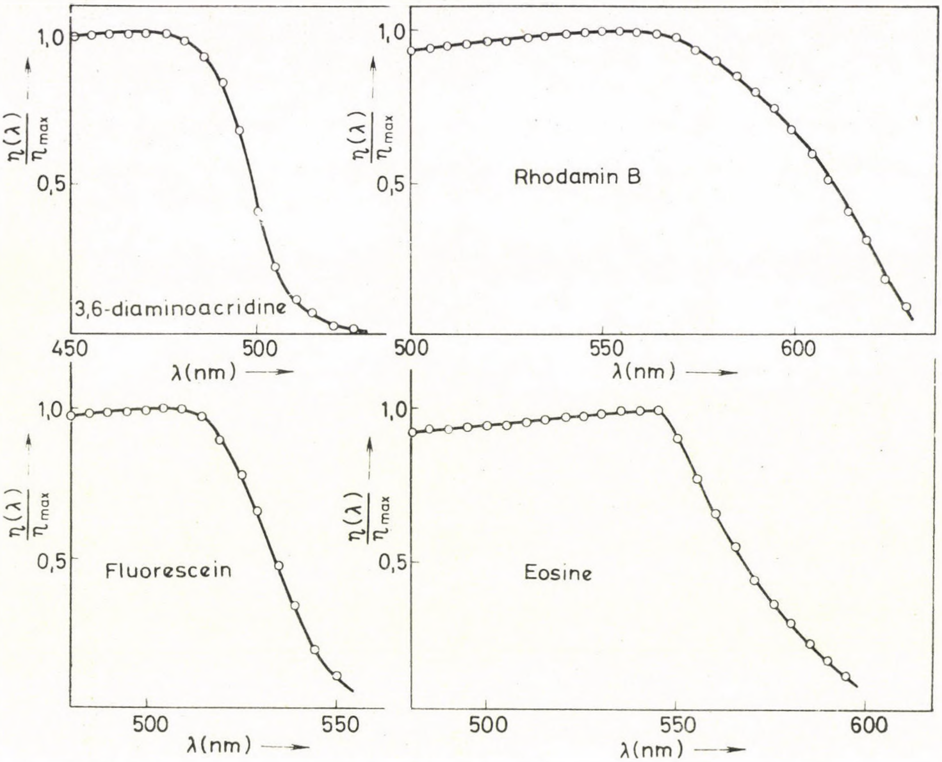

Fig. 6

independent of the wavelength λ' of the observation, and depends only on the maximum value of $\beta(\lambda')$ (Fig. 7). If this condition is not fulfilled, the dependence of $\varkappa_{ik}$ on λ' (and thus on $\beta(\lambda')$) is the stronger, the greater the value of $[\beta(\lambda')]_{\max}$ (Fig. 8; which, as well as Fig. 7, refers to equimolar mixed solu-

Table I

Absolute quantum yields

c (mole/l) substance	$\eta(\lambda_e)$							
	$1 \cdot 10^{-5}$	$2.5 \cdot 10^{-5}$	$5 \cdot 10^{-5}$	$1 \cdot 10^{-4}$	$2.5 \cdot 10^{-4}$	$5 \cdot 10^{-4}$	$1 \cdot 10^{-3}$ $2.5 \cdot$	$\cdot 10^{-3}$
3.6-diaminoacridine ($\lambda_e = 436$ nm)	0.61	0.60	0.61	0.58	0.60	0.60	0.56	0.51
rhodamin B ($\lambda_e = 546$ nm)	0.50	0.50	0.51	0.49	0.49	0.50	0.49	0.45
fluroescein ($\lambda_e = 436$ nm)	0.87	0.87	0.89	0.88	0.88	0.89	0.85	0.79
eosine ($\lambda_e = 436$ nm)	0.49	0.49	0.50	0.48	0.49	0.49	0.48	0.45

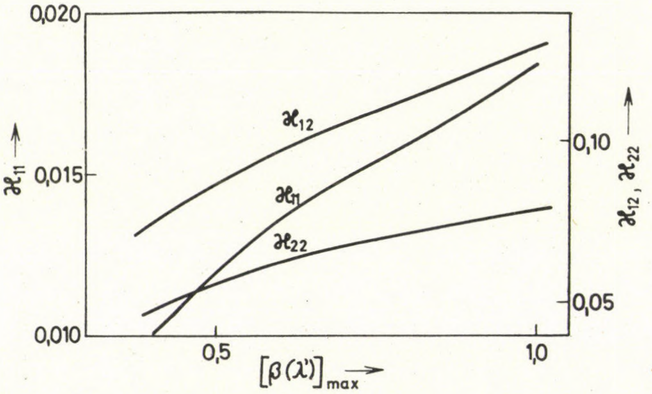

Fig. 7

tions of 3,6-diaminoacridine and rhodamin B). It follows from the above results that the intensity of the fluorescence emitted by a mixed solution is substantially affected by radiative energy transfer. Furthermore, the emission spectrum $B^*(\lambda')$ can be expressed as a linear combination

$$B^*(\lambda') = b_1 f_1(\lambda') + b_2 f_2(\lambda') \tag{18}$$

of the spectra of the components only when the product of the concentration and layer thickness of the solution is low enough; namely, in this case the factors b_1 and b_2 are independent of λ', as can be seen from the comparison of Eq. (9) with Eq. (18) and from the statements in c).

e) The dependence of S on c has been determined for both equimolar ($c_1 = c_2 = c$) and non-equimolar ($c_1 = 10^{-4}$ mole/l, $c_2 = c$) mixed solutions of 3,6-diaminoacridine-rhodamin B and of fluorescein and eosine with the formulas (10), (12) and (13) for excitation with two different wavelengths (436 nm, 460 nm and 436 nm, 490 nm, respectively). For a given acceptor concentration the same values of S were obtained at different exciting wavelengths both for equimolar and non-equimolar solutions. Thus S practically

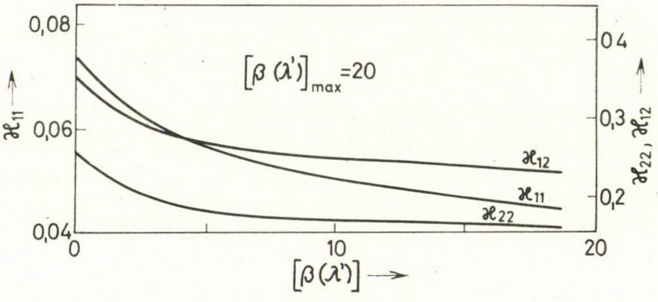

Fig. 8

depends only on the concentration of the acceptor. These results support the fundamental supposition concerning the non-radiative character of the energy transfer (see chapter II), which had not been checked carefully enough earlier.

The experimental results obtained for S render possible to control the relations resulting from JABŁONSKI's [20], FÖRSTER's and GALANIN's [18, 19], and KETSKEMÉTY's [21] theories, which are the following:

$$S = \frac{1-e^{-\nu}}{\nu} \quad \left[\nu = \frac{4\pi}{3}(1{,}33\,R_0)^3\,n_2\right], \tag{19}$$

$$S = 1 - 2\,qe^{q^2}\int_0^q e^{-x^2}\,\mathrm{d}x \quad \left[q = 2{,}74\,(2\pi n)^{-2}\left(\frac{\overline{\lambda^4}\,\tau_0}{L\tau_e}\right)^{1/2} n_2\right], \tag{20}$$

$$S = \int_0^\infty f_1(\lambda)\exp\left(-k_2(\lambda)/k^4a^3\right)\mathrm{d}\lambda \quad \left[k = 2\pi n/\lambda;\ a = (1/2\pi n_2)^{1/3}\right], \tag{21}$$

where τ_0 and τ_e are the main and the natural decay times, respectively, L Loschmidt's constant, $\overline{\lambda^4} = \int_0^\infty f_1(\lambda)\,\varepsilon_2(\lambda)\,\lambda^4\,d\lambda$, n_2 the number of molecules in a cm^3, R_0 the critical distance to be calculated from the fluorescence characteristics (R_0 is the distance between a donor and an acceptor molecule at which the probability of spontaneous emission from the donor is equal to the probability of non-radiative transfer of the exciting energy). In Figs. 9 and 10, curves 1, 2 and 3 show results calculated from JABŁONSKI's (Eq. (19)), FÖRSTER's and GALANIN's (Eq. (20)), and KECSKEMÉTY's (Eq. (21)) formulas, whereas the circles give the experimental results. As can be seen, the dependence S from concentration c is well described by all three curves. In curves 1 and 2 the deviation from the experiments increases with increasing acceptor concentration c and is somewhat greater for 1 than for 2. Curve 3 fits well to

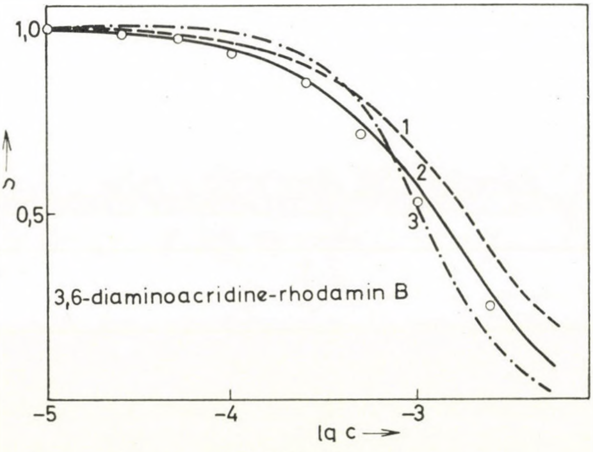

Fig. 9

the experimental results also in the region of high concentration, showing some deviation only in the region of medium concentration.

According to our experimental results the quantity *a* in Eq. (21), which, according to the theory, means the radius of the greatest sphere containing no acceptor molecule around a donor molecule, is to be substituted by $0.72\ (1/2\ \pi\ n_2)^{1/3}$; curve 3 has been calculated with this value. It can be stated on the basis of the results obtained that the dependence on concentration of the quantity S, characteristic for the relative yield of the donor molecules, can be well described with the formulas deduced on the basis of the supposed non-radiative mechanism of energy transfer.

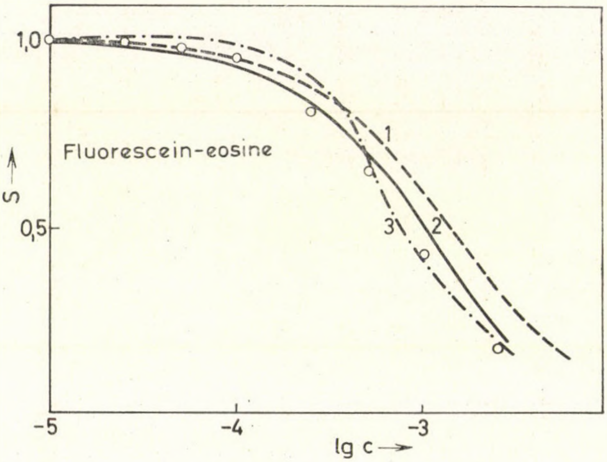

Fig. 10

The dependence of A on acceptor concentration is shown in Figs. 11 and 12. These results are used in the following calculations.

f) The probability of the elementary processes occurring in the molecules has been determined on the basis of the formulas given in Eq. (17). For the decay time τ of 3,6-diaminoacridine, rhodamin B, fluorescein and eosine, up to the limit of concentration quenching ($5 \cdot 10^{-4}$ mole/l), the values 4.0, 2.4, 3.5 and 2.4 nsec were obtained in turn. For the two higher concentrations ($1 \cdot 10^{-3}$ and $2.5 \cdot 10^{-3}$ mole/l) the corresponding values of τ were calculated from the relation $\tau/\tau_0 = \eta/\eta_0$ (see [13] p. 207), this relation being well fulfilled in the beginning of the region of concentration quenching. In the above sequence of the dye-stuffs, and expressed in 10^8 sec^{-1} as units, we obtained for s_i 1.51, 2.05, 2.64 and 2.68, for b_i 0.96, 2.05, 0.23, and 1.84 for all concentrations; higher values for b_i were obtained only for the concentrations 10^{-3} and $2.5 \cdot 10^{-3}$ mole/l, as a consequence of the decreasing yields of the dye-stuffs.

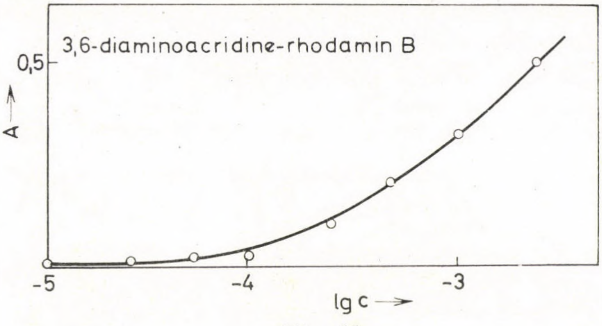

Fig. 11

While s_i and b_i are independent of the concentrations of the solutions below the region of concentration quenching, a_2 depends strongly on the acceptor concentration. According to the assumption of VAVILOV and other authors, the following relation between the probability a_2 and the molecule concentration n_2 corresponding to c should be valid:

$$a_2 = \frac{1}{k_2} n_2 , \tag{22}$$

where k_2 is a constant independent of the concentration n_2 (number of molecules/cm^3). On the contrary, when plotting the values lg a_2 obtained from our measurements performed in a wider region of concentration as a function of lg c (see Figs. 13 and 14), we obtained straight lines with slopes of 1.1 for mixed solutions of 3,6-diaminoacridine and rhodamin B, and 1.4 for fluorescein and eosine; thus a_2 is not a linear function of c (and therefore not of n_2).

For w_2 we obtained values near zero, which are low compared with the other probabilities in the concentration interval 10^{-5}—$5 \cdot 10^{-4}$ mole/l. In the case of the two highest concentrations, however, w_2 differs markedly from zero (e.g. at $2.5 \cdot 10^{-3}$ mole/l w_2 amounts to about 10% of a_2): so the quenching

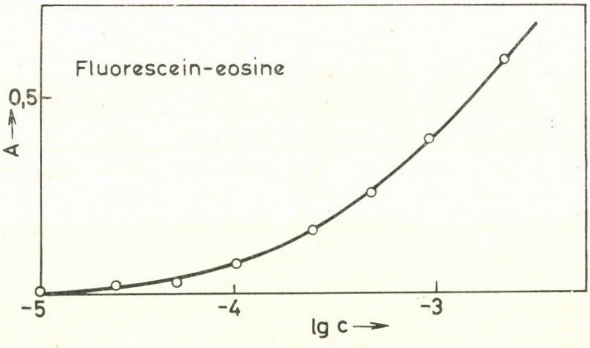

Fig. 12

effect of the acceptor on the fluorescence of the donor cannot be neglected at higher concentrations, in contradiction with ROZMAN's assumption [7].

g) In our investigations (see chapter IVb) we have used, with some modifications, a method of ROZMAN et al. to determine the probability A [7, 11]. As to the very ingenious original method of ROZMAN et al. and related considerations, we should like to make the following correcting remarks.

ROZMAN introduced the "quantum yield η_t of non-radiative energy transfer", and the "efficiency T_{12}" of the energy transfer, determinable by measurements; according to his considerations the following relation should exist between these quantities:

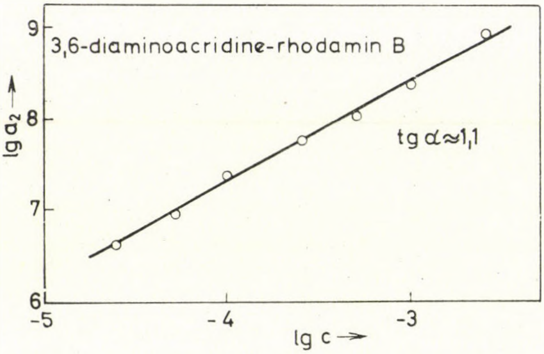

Fig. 13

$$T_{12} = \eta_t + \eta_1 R_{12}^* \tag{23}$$

the definition of which (also using our symbols) is

$$T_{12} = \frac{\frac{I_{21}}{I_{22}} - \alpha_{11}}{\alpha_{11}}, \qquad \eta_t = 1 - \frac{\eta_1}{\eta_{01}} = 1 - S. \qquad (24), (25)$$

The meaning of the symbols in Eq. (24) is given in connection with Eq. (15); η_{10} and η_1 are the quantum yields of the donor if $c_2 = 0$ and $c_2 \neq 0$, respectively. The term $\eta_1 R_{12}^*$ in Eq. (23), determinable on the basis of experimental data, takes into account the radiative energy transfer. In order to verify the adequacy of FÖRSTER's and GALANIN's theory, ROZMAN determined η_t experimentally from Eq. (23) and compared the dependence on concentration of this value with the dependence on concentration of η_t calculated from Eqs. (25) and (20).

Closer examination of the physical meaning of T_{12} and η_t shows that the definition of η_t has to be modified to ensure the validity of Eq. (23). Namely, let n be the number of exciting photons impinging on the sample in unit time,

n_e^0 and n_e^1 the number of the photons emitted by the donor in the same time, if $c_2 = 0$ and $c_2 \neq 0$, respectively. In case of complete absorption ($\eta_{10} = n_e^0/n$ and $\eta_1 = n_e^1/n\,\alpha_{11}$)

$$\eta t = \frac{\dfrac{n_e^0}{n} - \dfrac{n_e^1}{n\alpha_{11}}}{\dfrac{n_e^0}{n}} . \tag{26}$$

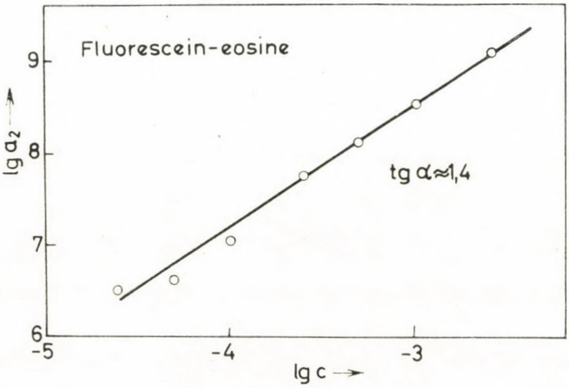

Fig. 14

Assuming that the decrease in yield of the donor is due to the efficient non-radiative energy transfer alone (i.e. $w_2 = 0$), Eq. (26) gives the number of photons transferred to the acceptor by non-radiative processes, divided by the number of the photons emitted by the "pure" donor solution under the same excitation.

Let the fluorescence intensities I_{21} and I_{22} in the definition of T_{12} be excited by n photons in unit time, of wavelengths λ_1 and λ_2, respectively, and completely absorbed by the sample; then the number of the acceptor molecules getting into the excited state will evidently be $\alpha_{21}\,n + n_s + n_r$ and n for the wavelengths λ_1 and λ_2, respectively, if n_s and n_r mean the number of the exciting photons transferred from the donor by radiative and non-radiative processes. If the quantum yield of the acceptor is η_A for both λ_1 and λ_2, then

$$I_{21} = (\alpha_{21}\,n + n_s + n_r)\,\eta_A\,, \quad I_{22} = n\eta_A\,. \tag{27}$$

From this and Eq. (24) we obtain

$$I_{12} = \frac{n_s + n_r}{n\alpha_{11}} . \tag{28}$$

That is, T_{12} gives the number of the transferred photons not in relation to the number of photons emitted by the donor, but to the number of the exciting quanta absorbed by the donor. Considering that the effect of the radiative energy transfer decreases with decreasing layer thickness, and in the limiting case $R_{12}^* = 0$, and therefore $\eta_t = T_{12}$, the relation (23) between T_{12} and η_t holds evidently only in the case, when in the definition of η_t the number of quanta transferred by non-radiative processes is given also in relation to the number of the exciting quanta absorbed by the donor, i.e. if η_t is defined as follows:

$$\eta_t = \eta_{10} - \eta_1 \equiv \eta_{10}(1 - S). \tag{29}$$

According to the above, the considerations of ROZMAN et al. will be valid even for $w_2 = 0$ only, if the true absolute quantum yield of the pure donor solution $\eta_{10} = 1$, which condition is generally not fulfilled, and further if the quantum yield of the acceptor is equal for both wavelengths λ_1 and λ_2, which differ markedly.

Now, according to the more precise definition given in Eq. (29), and if $w_2 = 0$ (i.e. for low acceptor concentrations), it is easy to obtain from Eq. (17), with the acceptable supposition that $\eta_{10} = \eta_{1m}$, the following relation for η_t:

$$\eta_t = \frac{\eta_{1m}^2}{\eta_{2m}} A, \tag{30}$$

which our measurements proved to be relatively well fulfilled in the region of not too high concentrations.

The author wishes to express his sincere thanks to Prof. A. BUDÓ, Director of the Institute, and to Prof. I. KETSKEMÉTY for their valuable advice during the investigations.

REFERENCES

1. G. CARIO and J. FRANCK, Z. Phys., **11**, 161, 1922.
2. S. I. VAVILOV, M. D. GALANIN and F. M. PEKERMAN, Abhandlungen aus der sowjetischen Physik, Folge I, p. 9, Verlag Kultur und Fortschritt, Berlin, 1951.
3. A. F. WATSON and R. LIVINGSTON, J. Chem. Phys., **18**, 802, 1950.
4. E. J. BOWEN and R. LIVINGSTON, J. Amer. Chem. Soc., **76**, 6300, 1954.
5. J. B. BIRKS and J. W. CAMERON, Proc. Phys. Soc., **72**, 53, 1958.
6. I. KETSKEMÉTY, Acta Phys. Hung., **10**, 429, 1959.
7. I. M. ROZMAN, Optics and Spectrosc., **10**, 354, 1961.
8. Á. BUDÓ and I. KETSKEMÉTY, Acta Phys. Hung., **14**, 167, 1962.
9. Á. BUDÓ and I. KETSKEMÉTY, Acta Phys. Hung., **7**, 207, 1957.
10. M. TÖRÖK, Doctoral dissertation, Szeged, 1967.
11. E. A. ANDREESTHEV, C. F. KILIN, I. M. ROZMAN and B. I. SHIROKOV, Izv. Akad. Nauk. SSSR, Ser. Fiz., **27**, 533, 1963.
12. S. I. VAVILOV, A fény mikrostruktúrája, III. rész, Akadémiai Kiadó, Budapest, 1955
13. TH. FÖRSTER, Fluoreszenz organischer Verbindungen, Vandenhoeck and Ruprecht Göttingen, 1951.
14. I. KETSKEMÉTY, J. DOMBI, R. HORVAI, J. HEVESI and L. KOZMA, Acta Phys. et Chem. Szeged, **7**, 17, 1961.

15. Á. Budó, J. Dombi and L. Szöllősy, Acta Phys. et Chem. Szeged, **2,** 18, 1956.
16. I. Ketskeméty, J. Dombi, J. Hevesi, R. Horvai and L. Kozma, Acta Phys. et Chem. Szeged, **7,** 88, 1961.
17. R. Bauer and M. Rozwadowski, Bull. Acad. Polon. Sci., Cl. III, **7,** 365, 1959.
18. Th. Förster, Z. Naturforsch., **4a,** 321, 1949.
19. M. D. Galanin, Zh. Eksper. Teor. Fiz., **28,** 485, 1955.
20. A. Jabłonski, Bull. Acad. Polon. Sci., Cl. III, **6,** 663, 1958.
21. I. Ketskeméty, Z. Naturforsch., **20a,** 82, 1965.

ПРОЦЕССЫ ПЕРЕДАЧИ ЭНЕРГИИ В ЛЮМИНЕСЦИРУЮЩИХ РАСТВОРАХ СМЕСИ

Й. ДОМБИ

Резюме

В работе дается зависимость между спектрами флуоресценции растворов смеси и компонентных растворов, которая принимает во внимание передачу излучаемой энергии с точностью, не достигнутой до настоящего времени. Резонансная передача энергии в данном выражении охарактеризуется всего двумя величинами. При помощи этих величин, далее использованием коэффициентов полезного действия и времен затухания компонентных веществ определяется частота молекулярных процессов, связанных с излучением и передачей энергии. Результаты экспериментов согласуются с данными исследований Яблонски, Фёрстера и Кечкемети, и подтверждают выдвинутую ими теорию о механизме перехода энергии. Проводится анализ метода Розмана, относящегося к переходу энергии, предлагаются коррекции к данному методу.

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AN EXACTLY SOLUBLE MODEL FOR RESONANCE SCATTERING

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A model system is constructed consisting of a single particle potential and two interacting particles. The potential has two bound states so that four configurations exist for the two particle system. One of these lies in the continuum giving rise to a resonance of "compound" type. Both the potential and the two particle interaction are of non-local and separable types. The exact transition amplitudes are obtained by means of FADDEEV's method. The model is designed for checking the "microscopic" nuclear reaction theories.

I. Introduction

In the course of the development of nuclear theory the description of nuclear reactions has been attempted mainly by two approaches. The first of these is characterized by a high degree of generality and exactness which is achieved without going into the details of the internal dynamics of the many-nucleon system. The main aim of these general theories [1] is to clarify the structure and the analytical properties of the collision matrix. The other, so-called microscopic approach starts from a more or less detailed dynamic description of the interacting many-nucleon system and tries to derive the S-matrix explicitly in terms of wave functions and Hamiltonians. As a rule, these microscopic theories of nuclear reactions [2] contain ab ovo some approximations, since at the present stage our ability to tackle the many-nucleon problem is very limited. In nuclear structure calculations concerning the low-lying states of nuclei, the various microscopic approximations used in the shell model, in the pairing model, in the RPA, etc. have been extensively studied and their validity and accuracy have often been tested.

As far as the microscopic nuclear reaction theories are concerned the situation is quite different. First, the actual calculations performed up to now are rather few in number mainly because of the cumbersome numerical computations involved. Second, the comparison of the calculated results with the experimental data does not allow definite conclusions to be drawn concerning

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the reliability of the approximations, because in addition to the approximations characteristic of these theories we always have to introduce further assumptions of a more general type, e.g. about the form of the interaction potentials, about the restriction of the configuration space, etc. In the final results of a detailed calculation designed to reproduce some experimental data the effect of these different approximations are completely mixed up and it is very difficult to decide which of them is responsible for the success of failure of the attempt. Therefore, it seems to us that the effect of the specific approximations used in microscopic reaction theories can be studied most conveniently on a model system for which an exact solution can be evaluated and which can then be compared with the results given by the different approximations [3].

In this paper our aim is to construct such a model and to work out an exact solution for it. The results of the numerical calculations and the comparison of this exact solution with the solutions obtained in different approximations will be reported in a subsequent paper. Now, the question arises: what kind of model system should be chosen? Of course, it must consist of an incoming particle and a target. If we want to go beyond the case of simple potential scattering we have to choose a target with an excitable internal structure. The simplest target that meets this requirement is a particle which has at least two bound states in a potential. This system is appropriate for the investigation of resonance effects in elastic and inelastic scattering and pick-up reactions. With a slight modification stripping reactions and composite particle elastic scattering can be studied as well. The two particles are taken to be equal mass fermions, but their spins will not enter explicitly since all the interactions will be taken as spin-independent and thus the fermion character of the particles will show up only in the symmetry properties of the spatial wave functions. In fact, the calculation can be carried out as if the particles were spinless and distinguishable, and the two possible spin-states (triplet and singlet) and the Pauli-principle (if necessary) can be taken into account afterwards in a simple way. Solving the Schrödinger equation of the above described system we are confronted essentially with a three-body problem: mathematically, because the Hamiltonian contains three interaction terms (the two single-particle potentials and the two-particle interaction) and physically, because the single particle potential, acting on both particles can be considered as a third particle with infinite mass. As is well known the three-body problem can be solved by FADDEEV's method leading to a system of coupled integral equations. FADDEEV's [4] method solves all the principal difficulties associated with the three-body problem, its practical application, however, is rather difficult, since it leads to coupled, two-variable integral equations. It was realized many years ago that the use of non-local, separable potentials greatly simplifies the problem [5]. Since the main guide in the choice of the model was mathematical simplicity, all the interactions entering the problem were taken to be spin-independent, non-local and separable acting only in relative s-states. The form and the parameters of the potentials were chosen to yield two single-particle bound states and one bound state in the two-particle system.

In Section II we specify our model in detail, in Section III we derive the integral equations to be solved numerically and in Section IV the calculation of the transition matrix elements is discussed.

II. The model

The investigated model consists of two particles: the first is bound in a potential well and the second is scattered on the first one. This system should be treated essentially as a three-body system since the potential which acts on both particles can be considered as a third particle with infinite mass. The Hamiltonian is the following:

$$H(1,2) = t(1) + t(2) + u(1) + u(2) + v(1,2)\,, \tag{1}$$

where t is the kinetic energy operator, u is the single particle potential (which is assumed to be the same for both particles) and v (1,2) is the interaction between the two particles. Since the aim of the present work is not a comparison between certain theoretical calculations and experimental data but rather a study of the reliability and accuracy of some approximations used in nuclear reaction theories, the choice of the potentials u and v was governed mainly by mathematical simplicity. Therefore both u and v were taken to be non-local and separable. It is the separability which simplified the calculation. Also, for the sake of simplicity, in both cases we restricted ourselves to the s-state interactions only. As we want to study not only direct, but also inelastic and resonance scattering, the potential u must have at least two bound states. The general form of the non-local separable potential which acts only in relative s-states is given in momentum representation by

$$\langle k_1\, k_2\, |v|\, k_1'\, k_2'\rangle = \delta(p-p') \sum_i \varrho_i\, \mathrm{v}_i(k)\, \mathrm{v}_i(k')\,, \tag{2}$$

where the total and relative momenta are denoted by p and k, respectively. It has been proved [6] that the number of bound states of such a potential is limited by the number of terms having the negative strength parameter (ϱ_i). In our model the two-particle system has only one bound state. Therefore, it is enough to retain one attractive term:

$$\langle k_1\, k_2\, |v(1,2)|\, k_1'\, k_2'\rangle = -\,\lambda^2\, \delta(p-p')\, \mathrm{v}(k)\, \mathrm{v}(k')\,. \tag{3}$$

The form factor $v(k)$ is defined as

$$v(k) = \frac{1}{\beta^2 + k^2} \tag{4}$$

which was first introduced by YAMAGOUCHI [5]. The wave function of the bound state can be obtained very easily and is given by

$$\varphi(k) = \frac{\sqrt{\alpha\beta(\alpha+\beta)}}{\pi} \cdot \frac{v(k)}{k^2 + \alpha^2} \tag{5}$$

or in coordinate representation:

$$\varphi(r) = \sqrt{\frac{\alpha\beta(\alpha+\beta)}{2\pi}} \cdot \frac{1}{\alpha - \beta} \left(\frac{e^{-\alpha r} - e^{-\beta r}}{r} \right). \tag{6}$$

The binding energy

$$\varepsilon = -\alpha^2 \frac{\hbar^2}{m}$$

is determined by the range (β) and strength (λ^2) parameters:

$$\alpha = -\beta + \sqrt{\frac{m\pi^2 \lambda^2}{\hbar^2 \beta}}\,. \tag{7}$$

Turning to the single particle potential u, first of all we note that in expression (2) the factor $\delta(p - p')$ expressing the translational invariance should be omitted. Thus

$$\langle k_1 | u(1) | k_1' \rangle = \sum_i \varrho_i \, v_i(k_1) \, v_i(k_1')\,. \tag{8}$$

Later on, we shall need the matrix elements of the single particle potential in the space of two particle states, and these can be expressed as

$$\begin{aligned} \langle k_1 k_2 | u(1) | k_1' k_2' \rangle &= \delta(k_2 - k_2') \sum_i \varrho_i \, v_i(k_1) \, v_i(k_1')\,, \\ \langle k_1 k_2 | u(2) | k_1' k_2' \rangle &= \delta(k_1 - k_1') \sum_i \varrho_i \, v_i(k_2) \, v_i(k_2')\,. \end{aligned} \tag{9}$$

Here the δ-functions clearly express the one-body character of u. Since in our model we want to have two single particle bound states, the potential must contain at least two terms:

$$\langle k_1 k_2 | u(1) | k_1' k_2' \rangle = -\delta(k_2 - k_2') \sum_i^2 \lambda_i^2 \, v_i(k_1) \, v_i(k_1')\,. \tag{10}$$

For bound states the Schrödinger equation reads

$$k_1^2\varphi_\nu(k_1) - \sum_{i=1}^{2} \Lambda_i\, v_i(k_1) \int \Lambda_i\, v_i(k_1')\, \varphi_\nu(k_1')\, \mathrm{d}k_1' = -\, \alpha_\nu^2\, \varphi_\nu(k_1)\,, \tag{11}$$

where

$$E_\nu = -\, \frac{2\, m}{\hbar^2}\, \alpha_\nu^2$$

is the ν — the nergy eigenvalue ($\nu = 0{,}1$) and v_i is defined as

$$\sqrt{\frac{2\, m}{\hbar^2}}\, \lambda_i\,.$$

The wave function can be obtained directly as

$$\varphi_\nu(k_1) = \frac{1}{k_1^2 + \alpha_\nu^2} \sum_{i=1}^{2} \Lambda_i\, v_i(k_1)\, N_i(\alpha_\nu)\ , \tag{12}$$

$$N_i(\alpha_\nu) = \int \Lambda_i\, v_i(k_1)\, \varphi_\nu(k_1)\, \mathrm{d}k_1\,. \tag{13}$$

Thus the coefficients $N_i(\alpha_\nu)$ are the solutions of the following homogeneous equations:

$$\sum_{i=1}^{2} M_{ji}(\alpha_\nu)\, N_i(\alpha_\nu) = 0\ , \tag{14}$$

where the symmetrical matrix $M_{ji}(\alpha_\nu)$ is defined as

$$M_{ji}(\alpha_\nu) = \delta_{ji} - 4\,\pi\, \Lambda_i\, \Lambda_j \int \frac{v_j(k_1)\, v_i(k_1)}{k_1^2 + \alpha_\nu^2}\, k_1^2\, \mathrm{d}k_1\,. \tag{15}$$

Equation (14) has non-trivial solutions only if

$$\det\big(M_{ji}(\alpha_\nu)\big) = 0\ . \tag{16}$$

This is an algebraic equation for the determination of the energy eigenvalues. The model is completely specified by the explicit definition of the form factors which are given by:

$$v_1(k) = \frac{1}{k^2 + \beta_1^2}\,, \tag{17}$$

$$v_2(k) = \frac{1}{k^2 + \beta_2^2} - \frac{\Gamma}{k^2 + \gamma_2^2}\,(\Gamma > 0)\,, \tag{18}$$

In this case the matrix $M_{ji}(\alpha_\nu)$ can be expressed explicitly in terms of the potential parameters as follows:

$$M_{11}(\alpha_\nu) = 1 - 2\pi^2 \Lambda_1^2 \frac{1}{2\beta_1(\beta_1+\alpha_\nu)^2} \tag{19a}$$

$$\begin{aligned} M_{22}(\alpha_\nu) = 1 - 2\pi^2 \Lambda_2^2 \Bigg\{ & \frac{1}{2\beta_2(\beta_2+\alpha_\nu)^2} - \\ & - \frac{2\Gamma}{(\gamma_2+\beta_2)(\gamma_2+\alpha_\nu)(\beta_2+\alpha_\nu)} + \frac{\Gamma^2}{2\gamma_2(\gamma_2+\alpha_\nu)} \Bigg\}, \end{aligned} \tag{19b}$$

$$\begin{aligned} M_{12}(\alpha_\nu) = - 2\pi^2 \Lambda_1 \Lambda_2 \Bigg\{ & \frac{1}{(\beta_1+\beta_2)(\beta_1+\alpha_\nu)(\beta_2+\alpha_\nu)} - \\ & - \frac{\Gamma}{(\beta_1+\gamma_2)(\beta_1+\alpha_\nu)(\gamma_2+\alpha_\nu)} \Bigg\}. \end{aligned} \tag{19c}$$

This choice of the form factors can be motivated by the following considerations. The bound states are orthogonal to each other and both have zero orbital angular momentum. Consequently, they must have a different number of nodes. A separable non-local potential characterized by the form factors (17) and (18) leads to a bound 1*s* state if $\Lambda_2 = 0$ and to a 2*s* state if $\Lambda_1 = 0$. Therefore it is expected that in the case when both v_1 and v_2 are different from zero we get two bound states. It is worth while mentioning that in the special case when both $v_1(k)$ and $v_2(k)$ are of YAMAGOUCHI type, that is, $\Gamma = 0$, we get two solutions with negative energy eigenvalues but these cannot be interpreted as proper bound states, since one of the wave functions has wrong asymptotic behaviour, namely, it decays more slowly in the asymptotic region than $e^{-\alpha_\nu r}$. This difficulty, however, does not arise if $\Gamma \neq 0$, and in this case it is possible to prove that with a proper choice of the parameters Λ_1, Λ_2, Γ, β_1, β_2, γ_2 the eigenvalue equation (16) has two positive roots α_1 and α_2 so that

$$\max(\alpha_1, \alpha_2) < \min(\beta_1, \beta_2, \gamma_2).$$

This condition guarantees the correct asymptotic behaviour of the bound state wave functions. Besides the roots α_1 and α_2 there exist two negative roots and a pair of complex roots which can give rise to a "potential resonance".

The main features of the model are summarized in Fig. 1, which shows the energy spectrum of the system and the thresholds of the various processes.

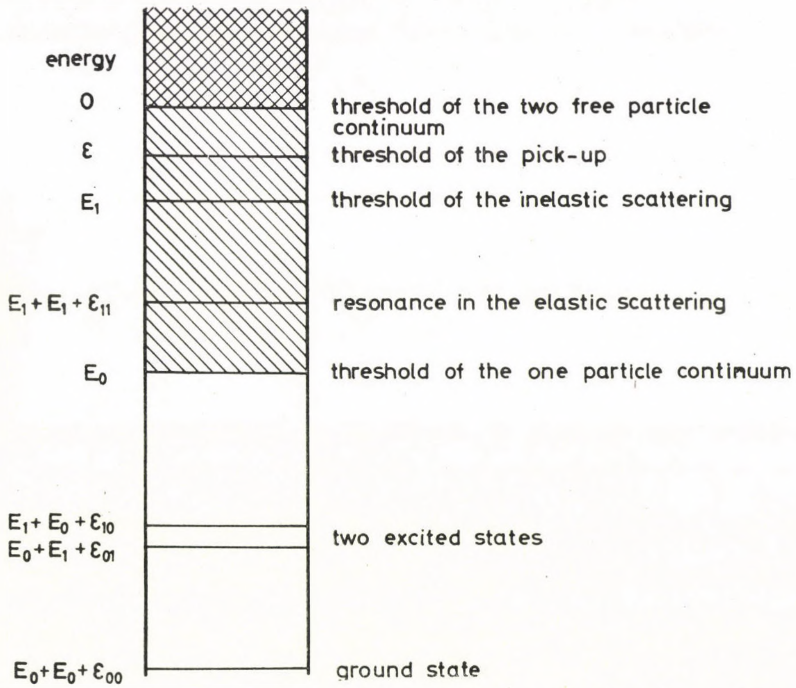

Fig. 1. The possible states of the model system. E_0 and E_1 are the bound state energies in the single particle potential, ε is the binding energy of the two particle system, ε_{00}, ε_{10}, ε_{01} and ε_{11} are the energy corrections to the "unperturbed" energies of u produced by the interaction $v(12)$

III. Integral equations for the wave function

The Schrödinger equation in momentum space is the following:

$$\left(k_1^2 + k_2^2 + \tilde{u}(1) + \tilde{u}(2) + v(1,2) - e\right) \Psi(k_1 k_2) = 0\,, \tag{20}$$

where $e = 2m/\hbar^2 E$ and E is the total energy, and the tilde means multiplication by $2m/\hbar^2$. In order to solve this equation the FADDEEV [4] method should be used in which the wave function Ψ is written as a sum of three terms and a system of coupled integral equations is obtained for these terms:

$$\Psi(k_1, k_2) = \Psi^1(k_1, k_2) + \Psi^2 + (k_1, k_2) + \Psi^{12}(k_1, k_2)\,, \tag{21a}$$

$$\begin{pmatrix} \Psi_1 \\ \Psi_2 \\ \Psi^{12} \end{pmatrix} = \begin{pmatrix} \Phi_{0ka} \\ 0 \\ 0 \end{pmatrix} + G_0 \begin{pmatrix} 0 & T_1 & T_1 \\ T_2 & 0 & T_2 \\ T_{12} & T_{12} & 0 \end{pmatrix} \begin{pmatrix} \Psi^1 \\ \Psi^2 \\ \Psi^{12} \end{pmatrix}. \tag{21b}$$

Here G_0 is the Green-operator of the non-interacting system; in the momentum representation:

$$\langle k_1\, k_2\, |G_0|\, k_1'\, k_2'\rangle = \frac{\delta(k_1 - k_1')\;\delta(k_2 - k_2')}{e - k_1^2 - k_2^2 + i\eta}\,. \tag{22}$$

The Φ^1_{0ka} is the wave function of the initial state, in which the first particle is in the bound ground state (indicated by the superscript 1 and subscript 0) and the second particle is described by an incident plane wave with momentum k_a. The operators T_1, T_2 and T_{12} are the T-operators of two-body problems and satisfy the equations:

$$\begin{aligned} T_1 &= \tilde{u}(1) + \tilde{u}(1)\, G_0\, T_1\,, \\ T_2 &= \tilde{u}(2) + \tilde{u}(2)\, G_0\, T_2\,, \\ T_{12} &= \tilde{v}(1,2) + \tilde{v}(1,2)\, G_0\, T_{12}\,. \end{aligned} \tag{23}$$

The first task is to solve equations (23). Using formulae (3) and (10) after some straight-forward calculations we obtain the matrix elements of the T-operators in momentum representation:

$$\langle k_1\, k_2\, |T_1|\, k_1'\, k_2'\rangle = \delta(k_2 - k_2') \sum_{i,j} \Lambda_i\, \Lambda_j\, v_i(k_1)\, v_i(k_1')\, M^{-1}_{ij}\left(\sqrt{k_0^2 + k_2^2}\right), \tag{24}$$

where the notations

$$\Lambda_i = \sqrt{\frac{2\,m}{\hbar^2}}\,\lambda_i \quad \text{and} \quad e = -\,k_0^2$$

were used and M^{-1} denotes the inverse of the matrix M defined in (16). A similar expression holds for the matrix element of T_2 except that the indices 1 and 2 must be interchanged on the right hand side of (24). The matrix element of T_{12} is as follows:

$$\langle k_1\, k_2\, |T_{12}|\, k_1'\, k_2'\rangle = -\,\Lambda^2\, \delta(p - p')\, v(k)\, v(k')\, \left[M\left(\sqrt{p^2 + 2\,k_0^2}\right)\right]^{-1}, \tag{25}$$

where again

$$\Lambda = \sqrt{\frac{2\,m}{\hbar^2}}\,\lambda, \quad e = -\,k_0^2$$

and the total and relative momenta were used. The quantity $M(x)$ is

$$M(x) = 1 - 2\,\pi\Lambda^2 \int_0^\infty \frac{v^2(k)\, k^2\, \mathrm{d}k}{k^2 + \left(\dfrac{x}{2}\right)^2} = 1 - \frac{2\,\pi^2\,\Lambda^2}{\beta}\;\frac{1}{(x + 2\beta)^2}\,. \tag{26}$$

Substituting (24) and (25) into (21b) we get the following expressions:

$$\Psi^1(k_1 k_2) = \Phi^1_{0ka}(k_1 k_2) + G_0 \sum_i \Lambda_i \, v_i(k_1) \, F'_i(k_2) \,, \tag{27a}$$

$$\Psi^2(k_1 k_2) = G_0 \sum_i \Lambda_i \, v_i(k_2) \, F^2_i(k_1) \,, \tag{27b}$$

$$\Psi^{12}(k_1 k_2) = G_0 \, \Lambda v(k) \, F^{12}(p) \,. \tag{27c}$$

On the right hand side of (27c) the total and relative momenta are introduced. The unknown quantities $F^1_i(x)$, $F^2_i(x)$, $F(x)$ satisfy the following equations:

$$\begin{aligned} M(\sqrt{x^2 + 2\,k_0^2}) \; F^{12}(x) &= \Lambda v \left(\frac{1}{2} x - k_a\right) \varphi_0 (x - k_a) + \\ &\quad + \Lambda \sum_i \int B_i(y, x) \left[F^1_i(y) + F^2_i(y)\right] \mathrm{d}y \end{aligned} \tag{28a}$$

$$\begin{aligned} \sum_i M_{ij} (\sqrt{x^2 + k_0^2}) F^1_j(x) &= \Lambda \int B_i(x, y) \, F^{12}(y) \, \mathrm{d}y + \\ &\quad + \sum_i \Lambda_j v_j(x) \int A_i(x, y) \, F^2_j(y) \, \mathrm{d}y \,, \end{aligned} \tag{28b}$$

$$\begin{aligned} \sum M_{ij}(\sqrt{x^2 + k_0^2}) \, F^2_j(x) &= \Lambda_i \, v_i(k_a) \, \psi_0(x) + \int B_i(x, y) \, F^{12}(y) \, dy + \\ &\quad + \sum_i \Lambda_j v_j(x) \int A_i(x, y) \, F^1_j(y) \, \mathrm{d}y \,, \end{aligned} \tag{28c}$$

where k_a is the momentum vector of the incident particle and the following energy equation holds:

$$E = E_0 + \frac{\hbar^2 k_a^2}{2\,m} \,;$$

$\varphi_0(x)$ is the ground state wave function of the bound particle. Further, the notations:

$$B_i(x, y) = \Lambda_i \, \frac{v_i(y - x) \, v \left(x - \frac{1}{2} y\right)}{k_0^2 + x^2 + (y - x)^2} \,, \tag{29}$$

$$A_i(x, y) = \Lambda_i \, \frac{v_i(y)}{k_0^1 + x^2 + y^2}$$

are used. Thus, we obtained a system of coupled integral equations for the 5 unknown functions $F_i^1(x)$, $F_i^2(x)$, $F^{12}(x)$. Each of these depends on a vector variable and, therefore, all the integrals in (14) are three-fold. In order to get one-variable integral equations we expand the F as follows:

$$F(x) = \sum_{lm} F_{lm}(x)\, Y_{lm}(\Omega_x)\,. \tag{30}$$

The kernel $B_i(x, y)$ can also be expanded making use of the fact that it depends only on the relative angle between x and y:

$$B_i(x, y) = \sum_{lm} B_i^l(x, y)\, Y_{lm}^*(\Omega_x)\, Y_{lm}(\Omega_y)\,. \tag{31}$$

The $A_i(x, y)$ need not be expanded because it is independent of the angles of x and y. To complete the expansion procedure we have to expand the first term on the right hand side of (28a):

$$v\left(\frac{1}{2}x - k_a\right) \varphi_\nu(x - k_a) = \sum_{lm} \varkappa_\nu^l(x, k_a)\, Y_{lm}(\Omega_x)\, Y_{lm}^*(\Omega_{xa})\,. \tag{32}$$

Here the index ν can be either 0 or 1 according to the two bound states of the potential u. In the initial wave function the particle 1. is in the ground state, therefore $\nu \neq 0$. If we assume — and this can be done without loss of generality — that the momentum of the incident particle is directed along the z-axis, then expansion (32) takes the following form:

$$v\left(\frac{1}{2}x - k_a\right) \varphi_\nu(x - k_a) = \sum_{l} \chi_\nu^l(x, k_a)\; Y_{l0}(\Omega_x) \sqrt{\frac{2l+1}{4\pi}}\,. \tag{32a}$$

Substituting the expressions (30), (31) and (32a) into equations (28), performing the integrations over the solid angle, and using the orthonormality of the spherical harmonics, we finally get the following set of one-variable integral equations:

$$\begin{aligned} M(\sqrt{x^2 + 2k_0^2})\, F_{lm}^{12}(x) = {} & \sqrt{\frac{2l+1}{4\pi}}\, \delta_{m0}\, \chi_0^l(x, k_a) + \\ & + \Lambda \sum_j \int B_j^l(y, x) \left[F_{jlm}^1(y) + F_{jlm}^2(y)\right] y^2\, dy, \end{aligned} \tag{33a}$$

$$\begin{aligned} \sum_j M_{ij}(\sqrt{x^2 + k_0^2})\, F_{jlm}^1(x) = {} & \Lambda \int B_i^l(x, y)\, F_{lm}^{12}(y)\, y^2\, dy + \\ & + 4\pi\, \delta_{l0}\, \delta_{m0} \sum_j \Lambda_j\, v_j(x) \int A_i(x, y)\, F_{jlm}^2(y)\, y^2\, dy, \end{aligned} \tag{33b}$$

$$\begin{aligned} \sum_j M_{ij}(\sqrt{x^2 + k_0^2})\, F_{jlm}^2(x) = {} & \Lambda \int B_i^l(x, y)\, F_{lm}^{12}(y)\, dy + \\ & + 4\pi\, \delta_{l0}\, \delta_{m0} \sum_j \Lambda_j\, v_j(x) \int A_i(x, y)\, F_{jlm}^1(y)\, y^2\, dy + \\ & + \sqrt{4\pi}\, \delta_{l0}\, \delta_{m0}\, v_i(k_a)\, \varphi_0(x)\,. \end{aligned} \tag{33c}$$

The equations (33) are uncoupled in the quantum numbers l and m This is due to the separability of the potentials; that is, to the fact that in expansions (16) we could introduce l and m in such a way that in the total wave function they turn out to be the total orbital angular momentum quantum numbers. This can be seen immediately if one substitutes the expressions (27) and (30) into (21a). The total wave function can be written as a sum over terms with different angular momentum and each term consists of three parts: in the first part particle 1 has zero angular momentum and particle 2. carries the whole angular momentum l; the second part has the same structure as the first, only the roles of particles 1. and 2. are interchanged; in the third part the relative motion of the two particles has zero angular momentum, while the centre-of-mass motion carries the total angular momentum l.

There is another feature of the solutions of eq. (33) that we can discover on the basis of symmetry considerations. The whole problem possesses an axial symmetry around the z-axis since the momentum vector of the incident particle is directed along the z-axis. This means that the wave function cannot depend on the azimuthal angle. Thus, in expansions (30) only the term with $m \neq 0$ should be retained; that is, the quantities F_{lm} are proportional to δ_{m0}.

IV. Transition matrix elements

In the preceding Section an exact solution was obtained for the wave function $\psi_a(k_1 k_2)$ of the scattering problem (there the subscript a indicates the initial state, described by Φ^1_{0ka}; see (21)). Our next task is to calculate the transition matrix elements

$$T_{ba} = \langle \Phi_b \, |v_b| \, \Psi_a \rangle, \tag{34}$$

which are connected with the observable cross-sections in a simple way. In (34) the Φ_b is the final state wave function and V_b is that part of the total interaction which is "not contained" in Φ_b. More precisely, this means that if satisfies the equation

$$H \Psi_a = E_a \Psi_a , \tag{35}$$

then Φ_b must satisfy

$$(H - V_b) \, \Phi_b = E_a \Phi_b . \tag{36}$$

To calculate the matrix elements (34) we have to construct, first of all, the possible final states Φ_b. There are three basic types of final states:

a) Simple scattering: particle 1. remains in one of the bound states while particle 2. is scattered into another free state, characterized by momentum k_b. The wave function of such a final state is:

$$\Phi_b(k_1 \, k_2) = \Phi^1_{\nu k b}(k, k_2) = \varphi_\nu(k_1) \, \delta k_2 - k_b) \; , \tag{37}$$

where the subscript ν refers to the bound state, and the superscript 1. indicates that in this wave function the particle 1. is bound. The vector k_b must satisfy the energy equation:

$$k_b^2 = \frac{2\,m}{\hbar^2}\,(E - E_\nu) > 0\,, \tag{38}$$

where E_ν and E are the energy of the ν-th bound state and the total energy, respectively. In this case:

$$V_b = u(2) + v(1{,}2)\,. \tag{39}$$

b) Exchange scattering: particle 1. flies out with momentum k_b and particle 2. remains bound. The wave function:

$$\Phi_b(k_1\,k_2) = \Phi^2_{\nu kb}\,(k_1\,k_2) = \varphi_\nu(k_2)\delta(k_1 - k_b)\,. \tag{40}$$

The energy equation coincides with (38). The remaining interaction has the form:

$$V_b = u(1) + v(1{,}2) \tag{41}$$

c) Pick-up reaction: particles 1. and 2. form a bound state and together fly out with total momentum k_b. The wave function:

$$\Phi_b(k_1\,k_2) = \Phi^{12}_{kb}(k_1\,k_2) = \varphi(k)\,\delta(p - k_b)\,, \tag{42}$$

where p and k are the total and relative momenta and $\varphi(k)$ is the wave function of the bound state of potential $v(12)$.

The energy equation is now:

$$\frac{1}{2}\,k_b^2 = \frac{2\,m}{\hbar^2}\,(E - \varepsilon) > 0\,. \tag{43}$$

In this case

$$V_b = u(1) + u(2)\,. \tag{44}$$

A final state with both particles 1. and 2. in free states cannot occur since we have restricted ourselves to negative E values only. By forming matrix elements (34) with the final state wave functions of these three types we can evaluate the cross-sections for any energetically possible process. Up to this point we have treated particles 1. and 2. as distinguishable, but it can be shown that in a symmetrized treatment the cross-sections can be expressed in terms of the same matrix elements. In order to calculate matrix elements (1) it is convenient to use the following identities:

$$\begin{aligned} u(1)\,\Psi_a &= (E - H_0)\,\Psi^1, \\ u(2)\,\Psi_a &= (E - H^0)\,\Psi^2\,, \\ v(1{,}2)\Psi_a &= (E - H_0)\,\Psi^{12}\,. \end{aligned} \tag{45}$$

These formulae can be obtained from equations (21) and (23) using an operator algebra. With the help of these equations the three types of matrix elements (34) can be easily evaluated. The results are the following:

Simple scattering

$$T^D(\nu k_b, o k_a) = \sum_l T_l^D(\nu k_b, 0 k_a)\, P_l(\cos\Theta)\,, \tag{46}$$

where the arguments of the transition matrix elements indicate the initial and final states, and Θ is the scattering angle (the angle between vectors k_a and k_b). The meaning of T_l^D is the following:

$$T_l^D(\nu k_c, 0 k_a) = -\frac{\hbar^2}{2m}\sqrt{2l+1}\left\{\sqrt{4\pi}\,\delta_{l0}\sum_i \Lambda_i\, v_i(k_b)\, I_{i\nu}^2 + \Lambda I_{l\nu}^{12}\right\} \tag{47}$$

with

$$I_{i\nu}^{\alpha} = \int \varphi_{\nu}(x)\, F_{i0}^{\alpha}(x)\, x^2\, dx\,, \qquad \alpha = 1,2 \tag{48}$$

and

$$I_{l\nu}^{12} = \int \chi_{\nu}^{l}(x,(k_b)\, F_l^{12}(x)\, x^2\, dx\,. \tag{49}$$

Exchange scattering

$$T^E(\nu k_b, 0 k_a) = \sum_l T_l^E(\nu k_b, 0 k_a)\, P_l(\cos\Theta)\,. \tag{50}$$

Here the notations are the same as in the preceding case.

$$\begin{aligned} T_l^E(\nu k_b, 0 k_a) = -\frac{\hbar^2}{2m}\sqrt{2l+1}\,\{\delta_{l0}\big[(k_0^2 + k_a^2 + k_b^2)\,\varphi_\nu(k_a)\,\varphi_0(k_b) &+ \\ + \sqrt{4\pi}\sum_i \Lambda_i\, v_i(k_b)\, I'_{i\nu}\big] + \Lambda\, I_{l\nu}^{12}\}&\,. \end{aligned} \tag{51}$$

Pick-up reaction

$$T^P(k_b, 0 k_a) = \sum_l T_l^P(k_b, 0 k_a)\, P_l(\cos\Theta)\,, \tag{52}$$

$$T_l^P(k_b, 0 k_a) = -\frac{\hbar^2}{2m}\left\{\omega_l(k_b, k_a) + \sqrt{\frac{2l+1}{4\pi}}\sum_i \Lambda_i\,(R_{il}^1 + R_{il}^2)\right\}, \tag{53}$$

where the quantities $\omega_l(k_b, k_a)$ and R_{il}^{a} are defined as follows:

$$\varphi\left(\frac{1}{2}k_b - k_a\right)\varphi_0(k_b - k_a)\left(k_0^2 + k_a^2 + (k_b - k_a)^2\right) = \sum_l \omega_l(k_a, k_b)\, P_l(\cos\Theta) \tag{54}$$

and

$$R_{il}^{\alpha} = \int \eta_l^i(k_b, x)\, F_{il}^{\alpha}(x)\, x^2\, dx\,. \tag{55}$$

The definition of $\eta_l(x, y)$ is the following:

$$\varphi\left(\frac{1}{2}x - y\right) u_i(x - y) = \sum_{l_m} \eta_l^i(x, y)\, Y_{lm}(\Omega_x)\, Y_{l_m}^*(\Omega_y). \tag{56}$$

In eqs. (54) and (56) $\varphi(x)$ denotes the wave function of the bound state of the potential $v(12)$.

It is well known that the transition matrix is diagonal in total angular momentum quantum number, but since we have used as initial and final states not angular momentum eigenstates, our transition matrix elements (46a), (50) and (52) do not correspond to a definite angular momentum. But the diagonal matrix elements of the transition operator, corresponding to a definite l can be extracted immediately just from equations (46), (50) and (52): they are the coefficients of $P_l(\cos \Theta)$ in those equations.

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REFERENCES

1. E. P. Wigner and L. Eisenbund, Phys. Rev., **72**, 29, 1947; J. Humblet and L. Rosenfeld, Nucl. Phys., **26**, 529, 1961.
2. H. Feshbach, Ann. of Phys. (N. Y.), **5**, 137, 1958; **19**, 287, 1962; C. Bloch and V. Gillet, Phys. Lett., **16**, 62, 1965; H. A. Weidenmüller, Nucl. Phys., **75**, 189, 1966.
3. R. Aaron and R. D. Amado, Phys. Rev., **150**, 857, 1966; A. I. Baz, V. F. Demin and I. I. Kuzmin, Jadernaja Fizika, **4**, 1131, 1966; P. E. Shanley and R. Aaron, Ann. of Phys., (N. Y.) **44**, 363, 1967.
4. L. D. Faddeev, ZETF, **39**, 1459, 1960; C. Lovelace, Phys. Rev., **135**, B 1225, 1964.
5. Y. Yamagouchi, Phys. Rev., **95**, 1628, 1954; A. N. Mitra and V. L. Narasimhan, Nucl. Phys., **14**, 407, 1959; G. A. Chilashvili, Bull. Acad. Sci. Georgian SSSR, **32**, 43, 1963; **33**, 35, 1964.
6. G. C. Ghirardi and A. Rimini, J. Math. Phys., **6**, 40, 1965.

ТОЧНО РАЗРЕШИМАЯ МОДЕЛЬ ДЛЯ РЕЗОНАНСНОГО РАССЕЯНИЯ

И. ЛОВАШ и Я. РЕВАИ

Резюме

Составляется модельная система, состоящая из потенциала одинокой частицы и из двух взаимодействующих частиц. Потенциал имеет два связанных состояния, так как в случае системы двух частиц существует четыре конфигурации. Одно из состояний лежит в континууме, вызывающее резонанс «составного» типа. Как потенциал, так и двучастичное взаимодействие относятся к нелокальным и отделимым типам. Точные переходные амплитуды получены посредством метода Фадеева. Модель разрабатывалась для проверки теорий «микроскопических» ядерных реакций.

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COMMUNICATIO BREVIS

SIMPLE DERIVATION OF EQUAL SPACING RULE IN MASSES IN DECUPLET BY CURRENT COMMUTATORS

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In SU(3) scheme, the decuplet belongs to the 10 dimensional irreducible representation of it and the masses of all the particles in decuplet are equal which we know is not true. This gives birth to the belief that there must be some force which breaks this symmetry. When the symmetry breaking mechanism is so chosen that a preferred direction F_8 is taken in the space of the generator algebra of SU(3), the masses of the particles in decuplet split.

The total Lagrangian for the decuplet system is

$$L = - \bar{B}_i^*(\gamma_\mu \delta_\mu + M_{B^*}) B_i^* \qquad (i = 1,2, \ldots 10) \tag{1}$$

where B_i^* is a Rarita—Schwinger field and stands for a particle in decuplet and $M_{B_i^*}$ its bare mass. B_i^* transforms under the set of infinitesimal transformations generated by the eight independent Hermitian operators F_i belonging to the eight dimensional algebra of SU(3) the following way

$$B_i^* \to (1 + i\boldsymbol{u} \cdot \boldsymbol{F}) B_i^* ,$$

and

$$\bar{B}_i^* \to \bar{B}_i^*(1 - i\boldsymbol{u} \cdot \boldsymbol{F}) , \quad (i = 1,2, \ldots, 10) \tag{2}$$

where u_i-s are infinitesimal vectors in F-space. The F-spin currents generated by F_i transformations are given by

$$J_\mu = \frac{\partial L}{\partial(\partial_\mu \boldsymbol{u})} . \tag{3}$$

When the SU(3) scheme is exact, the Lagrangian is invariant with respect to these transformations and we have

$$\partial_\mu J_\mu^i = 0 \qquad (i = 1,2, \ldots, 8) \tag{4}$$

and also the unitary spin densities J_0^i satisfy the relation

$$[J_0^i(\vec{x}, t), J_0^j(\vec{x}', t)] = -f_{ijk} J_0^k(\vec{x}, t)\, \delta(\vec{x} - \vec{x}')\,. \tag{5}$$

If we take into account the particular symmetry breaking mechanism that chooses the preferred direction F_8 in the space of the generator algebra the relation (4) is no more true but

$$\int [\partial_\mu J_\mu^A(\vec{x}', t)\, J_0^A(\vec{x}', t)]\, d^3x\, d^3x' = 0, \tag{6}$$

where $A = K^\pm, L^\pm$ holds good. We will use this identity to find the equal spacing rule in decuplet.

The eight components of the total unitary F-spin currents of the decuplet system using relation (3) is

$$J_\mu^i = i\bar{B}^* \gamma_\mu F_i B^*\,. \tag{7}$$

The F-spin current density J_μ^{K+} with the transformation property K^+ in V-spin space is given by

$$\begin{aligned} J_\mu^{K+} = {} & \frac{1}{\sqrt{6}} \bar{N}^{*\circ} \gamma_\mu Y^{*-} + \frac{1}{\sqrt{3}} \bar{N}^{*+} \gamma_\mu Y^{*\circ} + \frac{1}{\sqrt{3}} \bar{Y}^{*\circ} \gamma_\mu \Xi^{*-} + \\ & + \frac{1}{\sqrt{2}} \bar{N}^{*++} \gamma_\mu Y^{*+} + \frac{1}{\sqrt{2}} \bar{\Xi}^{*\circ} \gamma_\mu \Omega^- + \sqrt{\frac{2}{3}}\, \bar{Y}^{*+} \gamma_\mu \Xi^{*\circ}. \end{aligned} \tag{8}$$

Differentiating J_μ^{K+} with respect to x_μ and using field equations of $\bar{B}_i^*$, B_i^* we get

$$\begin{aligned} \partial_\mu J_\mu^{K+} = {} & \frac{1}{\sqrt{6}} (M_{N^{*\circ}} - M_{Y^{*-}})\, \bar{N}^{*\circ} Y^{*-} + \frac{1}{\sqrt{3}} (M_{N^{*+}} - M_{Y^{*\circ}})\, \bar{N}^{*+} Y^{*\circ} + \\ & + \frac{1}{\sqrt{3}} (M_{Y^{*\circ}} - M_{\Xi^{*-}})\, \bar{Y}^{*\circ} \Xi^{*-} + \frac{1}{\sqrt{2}} (M_{N^{*++}} - M_{Y^{*+}})\, \bar{N}^{*++} Y^{*+} + \\ & + \frac{1}{\sqrt{2}} (M_{\Xi^{*\circ}} - M_{\Omega^-})\, \bar{\Xi}^{*\circ} \Omega^- + \sqrt{\frac{2}{3}} (M_{Y^{*+}} - M_{\Xi^{*\circ}})\, \bar{Y}^{*+} \bar{\Xi}^{*\circ}. \end{aligned} \tag{9}$$

Now using the anticommutation relations

$$\begin{aligned} \{B_{a\mu}^*(\vec{x}, t), B_{a'\mu'}^*(\vec{x}', t)\} &= 0, \\ \{\bar{B}_{a\mu}^*(\vec{x}, t), B_{a'\mu'}(\vec{x}', t)\} &= \gamma_0\, \delta_{\mu\mu}\, \delta_{aa'}\, \delta(\vec{x} - \vec{x}') \end{aligned} \tag{10}$$

we get

$$\begin{aligned}
&\int d^{3'}x\, d^3x' \left[\partial_\mu J_\mu^{k+}(\vec{x},t), J_0^{k+}(\vec{x}',t)\right] = \\
&\int d^3x \frac{1}{3}\left\{(M_{N^{*++}} - M_{Y^{*\circ}}) - (M_{Y^{*\circ}} - M_{\Xi^{*-}})\right\}\bar{N}^{*+}\,\Xi^{*-} + \\
&+ \frac{1}{\sqrt{3}}\left\{(M_{N^{*++}} - M_{Y^{*+}}) - (M_{Y^{*+}} - M_{\Xi^{*\circ}})\right\}\bar{N}^{*++}\,\bar{\Xi}^{*\circ} + \\
&+ \frac{1}{\sqrt{3}}\left\{(M_{Y^{*+}} - M_{\Xi^{*\circ}}) - (M_{\Xi^{*\circ}} - M_{\Omega^-})\right\}\bar{Y}^{*+}\,\Omega_- = 0
\end{aligned} \tag{11}$$

which gives us

$$\begin{aligned}
&M_{N^{*+}} - M_{Y^{*\circ}} = M_{Y^{*\circ}} - M_{\Xi^{*-}}, \\
&M_{N^{*++}} - M_{Y^{*+}} = M_{Y^{*+}} - M_{\Xi^{*\circ}} = M_{\Xi^{*\circ}} - M_{\Omega^-}.
\end{aligned} \tag{12}$$

If we neglect electromagnetic splittings compared to splittings between the I-spin multiplets, this becomes the equal spacing rule

$$M_{N^*} - M_{Y^*} = M_{Y^*} - M_{\Xi^*} = M_{\Xi}{}^* - M_{\Omega^-} \tag{13}$$

which is in excellent agreement with experiments.

REFERENCES

1. M. GELL-MANN, CTSL—20 Report (1961) unpublished.
2. J. W. MOFFAT, Phys. Rev., **145**, 1177, 1966.
3. S. K. BOSE, to be published in Nuovo Cimento.

RECENSIONES

Symbole, Einheiten und Nomenklatur in der Physik

Friedrich Vieweg & Sohn Verlag, Braunschweig, 52 Seiten

Das Heft ist die deutschsprachige Ausgabe des in englischer und französischer Sprache schon erschienenen Dokumentes "Symbols, Units and Nomenclature in Physics" (Document UIP 11—SUN 65—3) der Kommission für Symbole, Einheiten und Nomenklatur der Internationalen Union für reine und angewandte Physik (IUPAP).

Das Dokument enthält in neun Teilen Empfehlungen über Formelzeichen, Bezeichnungen und Einheiten. Die empfohlenen Bezeichnungen sind ausser der deutschen Sprache auch in englischer und französischer Sprache mitgeteilt und so kann das Heft in seinem wesentlichen Teil auch als dreisprachiges Nomenklatur-Wörterbuch angewendet werden.

Als Anhang finden wir im zweckdienlichen Hefte die Vergleichung der verschiedenen Einheitensysteme und eine Tabelle der im internationalen und CGS-System angegebenen Werte der Konstanten der Physik.

Die Bearbeitung des übersichtlich zusammengestellten Dokumentes von geschicktem Format, besorgte im Auftrage der Deutschen Physikalischen Gesellschaft deren Kommission für Grössen, Einheiten und Symbole unter der Leitung von Prof. Dr. U. STILLE.

J. ANTAL

G. H. A. COLE:

An Introduction to the Statistical Theory of Classical Simple Dense Fluids

XXI + 284, Pergamon Press, Oxford, London, Edinburgh, New York, Sydney, Paris, Braunschweig, 1967, Price 75s.

The strong continuous interactions of the molecules in a liquid or in a dense gas make the theoretical description of such a system a very difficult problem. A restricted theory has been made in terms of phenomenological and semi-empirical theories and more recently a remarkable formal completeness for both classical and quantum systems has been achieved in the case of matter in equilibrium. However, no theory of comparable generality has yet been devised for systems not in equilibrium, although certain theories of restricted scope are now well established and other general approaches are being actively explored especially for dilute gases and crystalline solids, based on molecular models characteristic of these two states of matter. Only more recently have quantitative studies of dense fluids proved feasible containing much of interest to physicists, chemists and engineers. These studies have developed from the recognition of the importance of the reduced distribution function of small groupings of particles in space. The exact form of these functions is determined by the nature of the interaction forces between the details of the particle spatial correlation effects. Having in mind that the literature of the subject is now both wide and varied, and that the study of this problem still remains a specialist field having its own formalism, the present monograph is an attempt to provide non-specialists with a short readable introductory report of certain aspects of the study of dense fluids, based on the analysis of the correlation effects between representative small groupings of molecules.

In the introductory chapters the semi-empirical results, the methods of the microscopic representations and the fundamental concepts of fluid statistical thermodynamics are summarized. Then the theory of equilibrium short-range order, the so-called closure and total correlation approximations as well as the consequences of the equilibrium theory are treated in a very attractive presentation. Finally, the problem of irreversibility and the theories of non-equilibrium classical gases and liquids are discussed. As the emphasis is on simplicity and lucidity of these rather complicated methods, only insulating and single-component classical fluids are investigated, but a wide range of comments and references is included, in order to set the arguments of the book within the framework of the wider literature.

J. I. HORVÁTH

Electrons, Ions and Waves. Selected Works of William Phelps Allis

Edited by S. C. Brown, p. X + 442, The M.I.T. Press, Cambridge (Massachusetts) and London (England), 1967.

The selected papers of W. P. Allis, whose name is linked with the development of plasma physics, are collected in this volume in celebration of his promotion from Professor to Professor Emeritus of Physics at the Massachusetts Institute of Technology. The papers are grouped in the following parts: electrons and their interactions with atoms (1927—1933), diffusion and mobility (1937—1965), motions of electrons and ions (1935—1959), microwave discharges (1950—1952), waves and oscillations (1958—1962), plasma properties (1958—1962) and special types of discharge (1941—1942). The dates in parentheses refer to the periods when they were published. The collected papers should not be considered as a look at the past, i.e., they are not of historical interest only, but should be regarded as a compilation of fundamental insights upon which to built future progress. Therefore, this volume will be very useful for the beginner who intends to specialize in this up-to-date field.

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ZOLTÁN GYULAI
1887—1968

In the eighty-first year of his life ZOLTÁN GYULAI died on the thirteenth of July 1968. He had been a Full Member of the Hungarian Academy of Sciences, President of the Roland Eötvös Physical Society and Kossuth and State prize winner, as well as the holder of numerous home and foreign decorations.

He started his career under the guidance of Professor K. TANGL at the University of Kolozsvár in 1912. His work, after a promising start, was interrupted by the first world war. As a soldier he spent seven years in captivity, and returned home in 1922. Worn out but with undiminished energy he began a new life at the Institute of Experimental Physics of the University of Szeged where he worked as the assistant of Professor P. FRÖHLICH till 1935, although from 1924—1926 he was working in Göttingen with Professor R. W. POHL. Those years made a deep impression on him, and he always remembered them with affection. He developed warm friendships with Professor POHL and numerous other members of the Göttingen school which lasted during subsequent years too. He was appointed professor in 1935, first at the University of Debrecen till 1940, then at the University of Kolozsvár till 1947 and at the Technical University of Budapest till 1962, the year of his retirement. His scientific activities, however, did not stop; he remained the leader of the Research Laboratory on Crystal Growth of the Hungarian Academy of Sciences.

The first major field of his scientific activity included the problems of colour centres which came to light in the twenties. In the developmental period of quantum physics he enunciated the important fact that the photoconductivity produced by F light in X-irradiated NaCl crystals shows the quantum nature of light. He also found that the F bands observed in alkali halide crystals coloured additively and photochemically originate from centres of identical nature. His result has served as the basis of the F centre model. He was the first to describe the changes found in the absorption and photoconductivity of coloured crystals, which are now explained by F → F′ and F′ → F transformations, respectively. He succeeded in giving an explanation of the colouration of the blue and violet coloured rock-salt occurring in nature by examining their absorption and photoconduction spectra.

In the middle of the twenties the first large alkali halide monocrystals were grown from the melt in the Göttingen Institute (Kyropoulos method). GYULAI was the first to measure the dispersion (200—600 nm) of some alkali halides, and his figures serve even today as the basis for comparison.

Similarly in the twenties the investigation of crystal imperfections, a fundamental problem of modern solid state physics began to develop. GYULAI approached this question in a simple, ingenious way. His investigations, in collaboration with D. HARTLY in Szeged, into the increase of ionic conductivity in NaCl monocrystals under pressure, have remained fundamental. Their work is the first deliberate experiment to detect the existence of crystal imperfections, and the phenomenon known as the GYULAI—HARTLY effect is the subject of a wide variety of investigations even today.

In the thirties, his interest was attracted to the mechanism of crystal growth, and he remained faithful to this field to the end of his life. He was especially interested in KOSSEL's theory of crystal growth, and was the first to describe the appearance and movement of layers of microscopic thickness in the growth process of ionic crystals. He found that the rules given by the theory were in agreement with the observable mechanism of growth, though the theory works with single ions, while microscopic observation refers to layers of several thousand Å. Thus, the agreement is formal, and there is a fact behind it which is left out of consideration by the theory. To surmount this difficulty he assumed the existence of a semi-ordered boundary layer in the liquid phase at the crystal surface. In reality, according to him, not single ions but precrystalline formations in the boundary layer take part in the growth in a similar way as KOSSEL had suggested. — He was among the first to produce whiskers and to observe their physical properties. His examinations played an important part in arousing an interest in whiskers both from the theoretical and technical aspects. He observed interesting phenomena and processes in the appearance of crystallites in supersaturated alkali halide solutions. The fundamental phenomenon is that owing to mechanical effects an avalanche-like process of crystallization starts spreading over the whole volume of the solution consisting of several phases well separated in time. The appearing crystal forms are determined by the degree of supersaturation, temperature, impurities, etc.

GYULAI had always sought for the simple manifestations of principles covering wide ranging phenomena of Nature. He liked to employ simple methods and attempted to follow the processes by describing them with expressive models. His educational activity was also characterized by the same qualities. His demonstrative experiments during lectures were always of primary importance and were convincing owing to their clear-cut structure. He took an active part himself in their preparation, and constantly developed his experiments and supplemented them with new ones. Several of his ingenious demonstrative experiments have been published.

He dealt creatively with questions of methodology of education, and his students could learn from him in this respect, too.

His interests were exceedingly diverse. He was occupied with philosophy, analysed phenomena of biology and society, was fond of literature and the arts. However, he was not content only with gathering knowledge and enjoying spiritual values, but was always looking for fundamental connections, principles and laws in them.

GYULAY was not well treated by fate. He spent the greater part of his life in a period where objective and psychic conditions were equally missing for quiet and profound scientific work. Besides external circumstances, working conditions within his institute were generally not satisfactory. His infinite love for science, his great enthusiasm and inexhaustible energy rendered him capable of recommencing everything even several times in reduced circumstances with neither space for work nor sufficient financial means.

With GYULAI's death we have lost not only the great scientist but the Master has left us, from whom many of us had learned, and who had been our paragon. He started the study of solid state physics in our country, and the scientific atmosphere in which he educated his students has created a school. He has left us, but the results of his work live on in science, and the memory of his personality is cherished by the great number of his devoted and loving colleagues, friends and students.

I. TARJÁN

Acta Physica Academiae Scientiarum Hungaricae, Tomus 25(4), pp. 331—340 (1968)

THE INFORMATION GAIN BY LOCALIZING A PARTICLE

By

V. MAJERNIK

INSTITUTE OF PHYSICS, SLOVAK ACADEMY OF SCIENCES, BRATISLAVA, ČSSR*

(Received 19. III. 1968)

The paper is dealing with the application of information theory in quantum physics by using the mathematical term-information gain, which does not diverge for the continuous probability distribution. It is shown that the information gain by localizing a particle is connected with its energy change. An equation for the wave function, which maximalizes the effectiveness of information gain about the coordinate of a particle with respect to its energy change is given.

Introduction

A quantitative measure has been found recently for a fundamental concept of science information. This is the merit of a new mathematical iscience, the theory of information, the foundations of which were laid by C. E. SHANNON in 1948 [1, 2]. It is possible to define some concepts of information theory if we limit ourselves to simple probability distributions.

The information entropy for a discrete probability distribution $\mathscr{P}$, determined on a complete set of stochastical events **A**, according to the scheme:

$$\left|\begin{array}{c|c|c|c|c} \mathbf{A} & A_1 & A_2 & \cdots & A_n \\ \hline \mathscr{P} & P_1 & P_2 & \cdots & P_n \end{array}\right| \qquad \Sigma_{i=1}^{n} P_i = 1$$

is defined by the equation [3]

$$H = - \sum_{i=1}^{n} P_i \log_2 P_i \,. \tag{1}$$

In the expression (1) for the information entropy of the probability distribution of a stochastical variable ξ_d, $P_1, P_2, \ldots P_n$ represent the probabilities of the occurrence of its possible values $x_1, x_2, \ldots x_n$. The information entropy of a continuous stochastical variable ξ_c, the probability distribution of which is determined by a function of probability density $p(x)$, is given as follows [4]

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$$H = -(S_1 + S_2), \tag{2}$$

where

$$S_1 = -\int p(x) \log_2 p(x)\, dx$$

and

$$S_2 = \lim_{\Delta x \to 0} \log_2 \Delta x\,.$$

Let us consider two discrete probability distributions $\mathscr{P}\,[P_1, P_2, \ldots, P_n]$ and $\mathscr{W}\,[W_1, W_2, \ldots, W_n]$ determined on the same complete set of stochastical events **A**, according to a scheme

A	A_1	A_2	. . .	A_n
$\mathscr{P}$	P_1	P_2	. . .	P_n
$\mathscr{W}$	W_1	W_2	. . .	W_n

$$\sum_{i=1}^{n} P_i = \sum_{i=1}^{n} W_i = 1.$$

The information gain, when replacing the probability distribution $\mathscr{W}$ by probability distribution $\mathscr{P}$, is mathematically defined by the expression [5]

$$I(\mathscr{P} | \mathscr{W}) = \sum_{i=1}^{n} P_i \log_2 (P_i/W_i)\,. \tag{3}$$

For continuous probability distribution $\mathscr{P}_c$ and $\mathscr{W}_c$, determined by the functions of probability density $p(x)$ and $w(x)$, the expression (3) takes the form [5]

$$I(\mathscr{P}_c \,|\, \mathscr{W}_c) = \int p(x) \log_2 \big(p(x)/w(x)\big)\, dx\,. \tag{4}$$

Though at the beginning no concrete application of information theory appeared in physics, in the course of time, the conceptions introduced in the framework of information theory found more and more applications in science, mainly thanks to JAYNES' work in statistical mechanics [6, 7] and thanks to BRILLOUIN, who proved the negentropic character of information [8]. Information-theoretical quantities are determined by probability or statistical characteristics of a definite physical system. As certain physical quantities are also being defined by statistical averages of physical system as e.g. thermodynamic entropy, it is possible in statistical mechanics to discover the required correlations between the information-theoretical and physical quantities of the statistical system. If we consider a single particle only, the physical situation of such a physical system is somewhat complicated, as the statistical quantities of the system are unknown. In this case, however, we have in the framework of quantum mechanics the means for discovering the probability distributions of physical quantities, which so become physical stochastical variables, often continuous with certain functions of probability density [9].

A complication arises because the expression for the information entropy (2), diverges for continuous probability distributions and consequently it is impossible to ascribe a final value of the information entropy to the continuous physical stochastical variable. This difficulty may be overcome by using another information-theoretical quantity instead of the information entropy, namely the information gain, defined according to (3). In the framework of physics, the information gain may be interpreted as an alternation of "knowledge" level of a definite physical quantity of a physical system before and after a definite physical process. In order to determine the physical parameters of a physical system, it must be observed, i.e. affected in such a manner that it assumes the state in which the value of these parameters can be determined. The observation product can be evaluated qualitatively by means of the information gain. To do this, we need, of course, probability characteristics of physical stochastical variables of this system before and after observation. It can be said that each physical observation has a communication aspect, determined in terms of information theory. This aspect may sometimes be insubstantial; in other cases it determines the limit of measuring possibilities, as e. g. in the microworld. In this connection some questions of physical interest arise:

a) What are the relations between the information gain and other physical parameters of the observed physical system?

b) How can an observation be planned so that at a maximum information gain of a certain physical stochastical variable the other physical quantities of the physical system are altered by a minimum amount?

c) Is there any sense in considering physical quantities if there is no possibility of gaining any information about them?

In the following article we are going to consider some of these questions in a concrete physical system, namely in a system with a localized particle, described by the Schrödinger equation.

1. Particle localization and information gain

In a system containing one particle in a stationary case, we are going to find out the answer to some of the above questions. We shall try, at the same time, to find the correlations between the information gain connected with alternation of probability distributions of the coordinate of a particle, as a physical stochastical variable, and its corresponding energy change. Such a quantum mechanical system is described by means of the Schrödinger time-independent equation

$$-\frac{\hbar^e}{2m}\Delta\psi = E\psi\,, \tag{5}$$

where m is the particle mass. For the sake of simplicity we shall limit ourselves to the one-dimensional case of equation (5), i.e. to

$$-\frac{\hbar^2 \partial^2 \psi}{2m\, \partial x^2} = E\psi\,. \tag{6}$$

The particle coordinate in this case becomes a continuous stochastical variable, probability density $f(x)$ which is given by the relation

$$f(x) = \psi(x) \cdot \psi^*(x)\,,$$

where $\psi(x)$ is the solution of equation (6). If $f(x) = \text{const.}$, the probability of the occurrence of particles is equal in the whole space and, therefore, there is no localization. In order to localize the particle, we have to impose certain boundary conditions on the function of probability density and, therefore, also on the wave function. If the particle is to be localized in some domain only, having a radius a (localization radius), the conditions imposed upon the wave function are as follows:

$$\begin{aligned} \psi(x) &= 0 \qquad \text{for} \qquad |x| > a\,, \\ 0 \leqq \psi(x) &\leqq M \qquad \text{for} \qquad |x| \leq a, \end{aligned} \tag{7}$$

where M is a final value according to one condition which is required for the solution of Schrödinger equation.

To localize the particle for the space domain with a localization radius a means to find the solution of equation (6) with the boundary conditions (7). Such a solution can easily be found and has the form

$$\begin{aligned} \psi_n(x) &= B_n \cos\frac{n\pi x}{2a} \quad \text{for } n \text{ odd}, \\ \psi_n(x) &= A_n \sin\frac{nx\pi}{2a} \quad \text{for } n \text{ even}, \\ E_n &= \frac{\pi^2 \hbar^2 n^2}{8ma^2} \qquad n = 1, 2, 3, \ldots. \end{aligned} \tag{8}$$

The particle localized to localization radius a, has the lowest energy level for $n = 1$

$$E_1 = \frac{\hbar^2 \pi^2}{8m\, a^2}\,. \tag{8a}$$

If $a \neq \infty$ then $E_n \neq 0$. Thus, each localized particle has a certain energy, the lowest value of which is E_1. Altering the localization radius a by δa, also alters the minimum energy of the particle according to

$$\Delta E(a) = \frac{\hbar^2 \pi^2}{8m}\left(\frac{1}{a^2} - \frac{1}{(a+\delta a)^2}\right). \tag{9}$$

The information gain when altering the localization radius of the particle in case of $n = 1$ according to (4) is given by

$$I(\psi_1 | \psi_1') = \int_{-a}^{a} \frac{1}{a}\cos^2\frac{\pi x}{2a}\log_2\left[\frac{\frac{1}{a}\cos^2\frac{\pi x}{2a}}{\frac{1}{a'}\cos^2\frac{\pi x}{2a'}}\right] dx, \tag{10}$$

where a and a' are the localization radii, whereby

$$a < a'.$$

After modification, we get from (10) these integrals

$$I(\psi_1 | \psi_1') = J_1 + J_2 + J_3,$$

where

$$\begin{aligned} J_1 &= \int_{-a}^{a} \frac{1}{a}\cos^2\frac{\pi x}{2a}\log_2\left(\frac{a'}{a}\right) dx, \\ J_2 &= \int_{-a}^{a} \frac{1}{a}\cos^2\frac{\pi x}{2a}\log_2\left(\cos^2\frac{\pi x}{2a}\right) dx, \\ J_3 &= -\int_{-a}^{a} \frac{1}{a}\cos^2\frac{\pi x}{2a}\log_2\left(\cos^2\frac{\pi x}{2a'}\right) dx. \end{aligned} \tag{11}$$

By integrating, we obtain from (11)

$$\begin{aligned} I(\psi_1 | \psi_1') = \log_2(a'/a) + \frac{1}{\ln 2}\Bigg[1 - \frac{4}{\pi}\Phi_1(1) - 2\ln\left(\cos\frac{\pi a}{2a'}\right) - \\ - \frac{4}{\pi}\left(\frac{a}{a'}\right)\Phi_2(\pi/2) - \frac{2}{\pi}\left(\frac{a}{a'}\right)\Phi_3(\pi/2)\Bigg], \end{aligned} \tag{12}$$

where

$$\Phi_1(x) = \int_0^x \frac{\arcsin t}{t}\, dt,$$

$$\Phi_2(\pi/2) = \left(\frac{a}{a'}\right)\frac{\pi}{24} + \left(\frac{a}{a'}\right)^3\frac{\pi^3}{480} + \left(\frac{a}{a'}\right)^5\frac{\pi^5}{6720} + \ldots,$$

$$\Phi_3(\pi/2) = \left(\frac{a}{a'}\right)\frac{\pi}{4} + \left(\frac{a}{a'}\right)^3\frac{\pi^3 - 6\pi}{3\cdot 2^4} + \left(\frac{a}{a'}\right)^5\left(\frac{\pi^5 + 20\pi^3 - 12\pi}{15\cdot 2^6}\right) + \ldots.$$

For the ratio of information gain and energy change $q(a, a')$, which represents a measure for effectiveness of information gain about the particle coordinate with respect to the energy, we get, according to (12) and (9),

$$q(a, a') = \frac{\log_2\left(\frac{a}{a'}\right) + \frac{1}{\ln 2}\left[1 - \frac{4}{\pi}\Phi_1(1) - 2\ln\left(\cos\frac{\pi a}{2a'}\right) - \frac{2a}{\pi a'}\left(2\Phi_2(\pi/2) + \Phi_3(\pi/2)\right)\right]}{\frac{\pi^2 \hbar}{8m}\left(\frac{1}{a^2} - \frac{1}{a'^2}\right)} . \tag{13}$$

The expression (13) gives one required relation between the information-theoretical quantity, i.e. information gain and physical quantities, i.e. coordinate and energy of a particle.

For $a \ll a'$ (12) becomes

$$I(\psi_1 | \psi_1') = \log_2(a'/a) + \frac{1}{\ln 2}\left[1 - \frac{4}{\pi}\Phi_1(1)\right]$$

and (9) due to the inequality $1/a'^2 \ll 1/a^2$ transists to

$$\Delta E \doteq \frac{\pi^2 \hbar^2}{8ma^2} ,$$

so that we obtain for

$$q(a, a') = \frac{8ma^2\left(\log_2\left(\frac{a'}{a}\right) + C\right)}{\pi^2 \hbar^2} , \tag{13a}$$

where $C = 1/\ln 2\,(1 - 4/\pi\,\Phi_1(1))$. As $C \approx 1$, we can, regarding the condition $a < a'$, neglect the constant C, when comparing it with $\log_2(a'/a)$. Thus, we get

$$q(a, a') = Ka^2 \log_2(a'/a) , \tag{13b}$$

where $K = 8\,m/\pi^2\hbar^2$. According to (13b) the effectiveness of information gain is zero when $a \to 0$.

Similarly, for the three-dimensional case, we get for a localization radius a by boundary conditions:

$$\psi(r) = 0 \qquad \text{for} \qquad |r| > a ,$$
$$0 \leq \psi(r) < M \qquad \text{for} \qquad |r| \leq a ,$$

a solution of (5) (which is, moreover, dependent upon the orbital quantum number l) in the form (for $l = 0$)

$$\psi(r) = \frac{1}{\sqrt{2\pi a}} \, \frac{\sin \dfrac{n\pi r}{a}}{r}$$

and energy

$$E_n = \frac{n^2 \pi^2 \hbar^2}{2ma^2} \qquad n = 1, 2, 3, \ldots .$$

For the information gain, we get an integral ($n = 1$) similar to (10)

$$I(\psi(r) \mid \psi'(r)) = \int_{-a}^{a} \frac{2}{a} \sin^2 \frac{\pi x}{a} \log_2 \left[\left(\frac{a'}{a} \right) \frac{\sin^2 \left(\dfrac{\pi x}{a} \right)}{\sin^2 \left(\dfrac{\pi n}{a'} \right)} \right] dr ,$$

so that we can also determine the required ratio between the information gain and energy change for a change in the localisation radius of a particle.

It should be noted that when $a \to 0$, the wave function has an infinite value which, however, with regard to the boundary conditions imposed on the solution of the Schrödinger equation, is not permitted. Therefore, it is not possible within the framework of quantum mechanics to determine the particle coordinate exactly. From the above mentioned it may be assumed:

1) When localizing the particle, its energy changes, the minimum value of which is given by expression (8*a*).

2) The information gain is connected with the change of the localization radius of a particle according to (12).

3) The energy of a localized particle must be infinitely increased for the particle coordinate to be determined precisely.

2. Extreme value for the effectiveness of information gain

In the previous paragraph we have examined the probability distribution of the coordinate of a particle which is localized in the localization radius a and as the lowest energy state, so that the function of the probability density has been given in the form

$$f(x, a) = \psi_1^2 (x, a) = \frac{1}{a} \cos^2 \frac{\pi x}{2a} .$$

However, the solution (8) forms an orthonormal system of wave functions by means of which it is possible to expand a continuous real valued function

$f(x)$ defined in the domain $|\, x \,| < a$. If $f(x, a)$ is the function of probability density of the coordinate of a particle, then the required wave function is given by the relation

$$\psi(x, a) = \sqrt{f(x, a)}\,.$$

The wave function $\psi(x, a)$ can be expanded on the eigenfunctions $\psi_1(x, a)'$ $\psi_2(x, a), \ldots, \psi_n(x, a)$ given for the localization radius a by (8)

$$\psi(x, a) = \sum_{i=1}^{\infty} \tau_i\,(a)\; \psi_i\,(x, a)\,,$$

where

$$\tau_i\,(a) = \int_{-a}^{a} \psi(x, a)\, \psi_i\,(x, a)\, dx\,.$$

The information gain when changing the localization radius is, according to (4), given by the equation

$$I\big(\psi(x, a)\,|\,\psi'\,(x, a')\big) = \int_{-a}^{a} \psi^2\,(x, a) \log_2 \left[\frac{\psi^2\,(x, a)}{\psi'^2\,(x, a')}\right] dx\,. \tag{14}$$

For the mean energy change ΔE we get

$$\Delta E = \sum_{i=1}^{\infty} (E_i\,|\,\tau_i^2| - E_i'\,|\tau_i'|^2\,,$$

where

$$\tau_i'\,(a') = \int_{-a'}^{a'} \psi'\,(x, a')\, \psi_i'\,(x, a')\, dx$$

and E_i and E_i' are eigenvalues of energy for localization radii a and a'. In this case the ratio $q(a, a$') takes the form

$$q(a, a') = \frac{\displaystyle\int_{-a}^{a} \psi^2\,(x, a) \log_2 \left[\frac{\psi^2\,(x, a)}{\psi'^2\,(x, a)}\right] dx}{\displaystyle\sum_{i=1}^{\infty} \big(E_i\,|\tau_i|^2 - E_i'\,|\tau_i'|^2\big)}\,. \tag{15}$$

As $\tau_i(a)$ and $\tau_i'(a')$ are functional-bound with $\psi(x, a)$ and $\psi(x, a')$ represents expression (15) a functional defined on the set of continuous functions, by means of which it is possible to find a class of functions for which the expression (15) gets an extreme value. In our case a maximum value is required. In order to find the extreme values of $q(a, a')$, we can also proceed so that the functions $\psi(x, a)$ and $\psi'(x, a')$ are expanded into series of eigenfunctions and find out the extreme value of $q(a, a')$, if τ_i and τ_i' are considered as the parameters. We look for the extreme value of the expression:

$$q(a, a') = \frac{\int_{-a}^{a} \left(\sum_{i=1}^{\infty} \tau_i \, \psi_i\right)^2 \log_2 \left[\frac{\sum_{i=1}^{\infty} \tau_i \, \psi_i \,(x, a)}{\sum_{i=1}^{\infty} \tau_1' \, \psi_i' \,(a', x)}\right]^2 dx}{\sum_{i=1}^{\infty} \left(E_i \, |\tau_i|^2 - E_i' \, |\tau_i'|^2\right)} \, .$$

The necessary condition for acquiring the extreme value of $q(a, a')$ is by satisfying the equations:

$$\begin{aligned} \frac{\partial q}{\partial \tau_i} &= 0 \qquad i = 1, 2, 3, \ldots, \\ \frac{\partial q}{\partial \tau_i'} &= 0 \qquad i = 1, 2, 3 \; \ldots . \end{aligned} \tag{16}$$

By means of equations (16) τ_i and τ_i' are, in principle, determined and thus also the maximizing functions $\psi(x, a)$ and $\psi'(x, a')$. The solution of equations (16), with respect to their number, is a rather complicated matter but, nevertheless, it can be done numerically to a definite approximation.

To sum up:

1) The information gain is necessarily combined with the energy change of a particle.

2) The effectiveness of the information gain regarding the energy about a stochastical variable-coordinate of a particle is given by the ratio defined as:

$$q(a, a') = \frac{I\big(\psi(x, a) \,|\psi' \,(\psi'(x, a'))\big)}{\Delta E(a, a')} \, .$$

3) For $a \to 0, q(a, a')$ is zero and the wave function obtains an infinite value that is not permitted in quantum mechanics.

4) Functions exist which maximize the expression for effectiveness of information gain by localizing a particle. Their forms can be determined by means of equation (16).

REFERENCES

1. C. E. SHANNON, A Mathematical Theory of Communication. BSTJ **27**, 379, 623, 1948.
2. C. E. SHANNON and W. WEAVER, The Mathematical Theory of Communication, Univ. of Illinois Press, Urbana, 1949.
3. A. J. CHINTSCHIN, Der Begriff der Entropie in der Wahrscheinlichkeitsrechnung. Arbeiten zur Informationstheorie I. VEB Deutscher Verlag d. Wissenschaften, Berlin, 1957.
4. A. KATZ, Principles of statistical mechanics. The information theory approach. Preprint 1965
5. A. RÉNYI, Wahrscheinlichkeitsrechnung, VEB Deutscher Verlag d. Wissenschaften, Berlin, p. 482, 1962.
6. E. T. JAYNES, Phys. Rev., **106**, 620, 1957.
7. E. T. JAYNES, Phys. Rev., **108**, 108, 1957.
8. L. BRILLOUIN, J. of Applied Physics, **24**, 1152, 1953.
9. V. MAJERNIK, Acta Physica Austriaca, **25**, 243, 1967.

УВЕЛИЧЕНИЕ ИНФОРМАЦИИ ПУТЕМ ЛОКАЛИЗАЦИИ ЧАСТИЦЫ

В. МАЙЕРНИК

Резюме

В работе рассматривается вопрос применения теории информации в квантовой физике с использованием математического терм-информационного увеличения, которое не расходится для непрерывного распределения вероятности. Показывается, что увеличение информации путем локализации частицы связано с изменением ее энергии. Дается уравнение для волновой функции, которое доводит до максимума эффективность увеличения информации по координате частицы относительно изменения ее энергии.

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APPLICATION OF THE WAVE-MECHANICAL REPULSIVE POTENTIAL IN THE CALCULATION OF THE ENERGY OF THE Li ATOM

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The first ionization energy and the (2*s*) eigenfunctions of Li are calculated in the Hartree approximation by using a quantum-mechanical repulsive potential. The usefulness of the pseudopotential method is demonstrated.

We know from the theory of the pseudopotentials [1] that the calculation of the energy in the statistical theory of atoms is greatly simplified if we replace the Pauli principle by a repulsive potential. Using this potential we can construct a simplified "self-consistent field" method [1—2]. With this method we can obtain, without the use of computers, energy and eigenfunctions comparable in accuracy to those obtained with the Hartree or Hartree—Fock method, if by arranging the electrons of the atom into shells according to the principal quantum number we calculate the energy of the K-, L-, M-shells in such a way that each energy expression is complemented with the potential energies connected with F_l or G_l, or terms related to the operators Φ_n or Φ_{nl}, which can be interpreted only wave-mechanically (for the definition of these expressions see [1]). This procedure makes it possible that following simple Slater type function can be chosen for radial wave function of principal quantum number n

$$f_n = A_n r^{\varkappa_n} e^{-\lambda_n r} . \tag{1}$$

Depending on the accuracy required we can choose in this Ansatz $\varkappa_n = n$ (in this paper f_n denotes the radial wave function multiplied by r) and in this case the results can be regarded as first approximation. The parameters $\varkappa_n$ and λ_n, the energy and the eigenfunction can then be determined by the variation of the parameters.

The aim of the present paper is the application of the theory of repulsive potentials to the determination of the ionization potential of the Li atom. The calculations could be carried through both by the use of the repulsive potentials F_l and G_l or of the operator Φ_{nl}. We here chose this latter possibility.

The wave-mechanical repulsive potential of the electron in the (n, l) state can be written as [3]

$$-\frac{1}{2}\Phi_{nl}(r,r') = -\frac{1}{e}\sum_{n'=l+1}^{n-1}(\varepsilon-\varepsilon_{n'l})\,\varphi_{n'l}(r)\,\varphi_{n'l}(r')\,. \tag{2}$$

Here ε is the energy of the valence electron to be computed, $\varepsilon_{n'l}$ and $\varphi_{n'l}(r)$ are energy and the orthonormal radial wave function multiplied by r, respectively, of the core-electrons lying lower than(n, l) to be calculated.

As is known, the Li atom has in the ground state two $(1s)$ electrons and one $(2s)$ valence electron. The energy ε_2 in terms of the operator above has the form [1—2]

$$\varepsilon_2 = \int_0^\infty f_2^*(r')\,H_l f_2(r')\,dr' + \frac{|(f_1,f_2)|^2}{1-|(f_1,f_2)|^2}\left[\int_0^\infty f_2^*(r')\,H_l\,f_2(r')\,dr' - \varepsilon_1\right]. \tag{3}$$

The Hamilton operator H_l does not contain the exchange and correlation energy, in accordance with the Hartree approximation and is of the form

$$H_l = -\frac{1}{2}e^2 a_0 \frac{d^2}{dr^2} - eV + \frac{1}{2}e^2 a_0 \frac{l(l+1)}{r^2}\,, \tag{4}$$

where l is the angular momentum quantum number, which is zero for all three electrons in Li, and V is the following interaction potential of valence electron with the nucleus and the two core electrons

$$V = -\frac{Ze}{r} + 2V_e(r)\,, \tag{5}$$

where $Z = 3$ and $V_e(r)$ is the potential of a $(1s)$ electron.

In ε_2 we also find the overlap integral

$$(f_1, f_2) = \int_0^\infty f_1^* f_2\,dr\,. \tag{6}$$

As we mentioned we chose the radial eigenfunctions (multiplied by r) to be of the Slater type $f_n = A_n r^{\varkappa_n} e^{-\lambda_n r}$ and we determined $\varkappa_n$ and λ_n by the variation of the energy. We obtained the parameters of the radial eigenfunctions of the $(1s)$ electrons by the variation of the energy of Li$^+$

$$E_1 = \int_0^\infty f_1^*(r')\left[-e^2 a\frac{d^2}{dr^2} - \frac{2Ze^2}{r'} + eV_e(r')\right] f_1(r')\,dr'\,, \tag{7}$$

from which we get $\varkappa_1 = 1$ and $\lambda_1 = 2.6875\ 1/a_0$. The energy minimum coincides with the energy of the Li$^+$ ion computed by HYLLERAAS [4]. We get ε_1

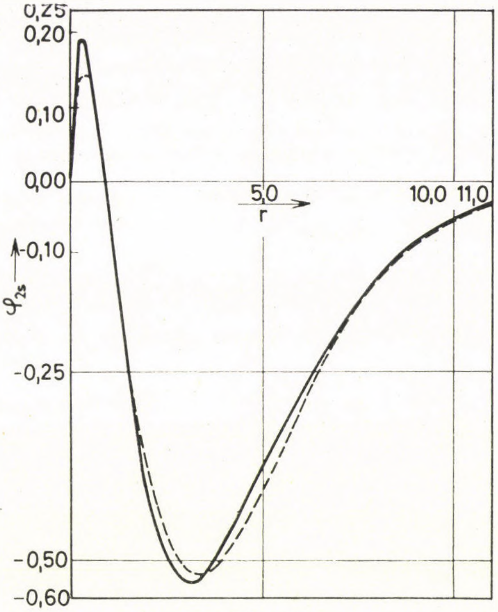

Fig. 1

in the energy expression (3) by adding $1/2\ e \int_0^\infty f_1^*(r')\ V_e(r')\ f_1(r')\ dr'$ to $1/2\ E_1$, therefore

$$\varepsilon_1 = \frac{1}{2}\ E_1 + \frac{1}{2}\ e \int_0^\infty f_1^*(r')\, V_e(r') f_1(r')\, dr' \,. \tag{8}$$

With the energy expression (3) we obtained the value —5.24 eV for the ionization energy of the Li atom which, taking into account the fact that our energy does not contain the exchange and correlation energy, is in good agreement with the experimental —5.40 eV [5]. We give our results in the Table:

Table I

$n,\ l$	$\varkappa_n$	λ_n	ε_n eV	ε_n eV exp.
1*s*	1.	2.6875	—75.47	—75.7
2*s*	2.5	0.764	— 5.24	— 5.40

We proceed to the second approximation by orthogonalizing the eigenfunctions $f_n(r)$ according to SCHMIDT [1—2]. Let us denote the orthogonalized radial eigenfunction multiplied by r with φ_{nl}; then

$$\varphi_{nl} = C_{nl}\left[f_n - \sum_{n'=l+1}^{n-1} (\varphi_{n'l}, f_n)\, \varphi_{n'l}\right], \tag{9}$$

where

$$C_{nl} = \frac{1}{\left[1 - \sum\limits_{n'=l+1}^{n-1} \left|\varphi_{n'l}, f_n)\right|\right]^{1/2}} . \tag{10}$$

The orthonormalized Li(2s) eigenfunction is given in the Figure. Since the Li(2s) Hartree eigenfunction is not tabulated we compared our function with that of Hartree—Fock with which it can only agree to the extent demonstrated in the Figure because the Hartree—Fock calculations contain also exchange energy [6].

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REFERENCES

1. P. Gombás, Pseudopotentiale, Springer, Wien, 1967.
2. P. Gombás, Theor. Chim. Acta (Berlin) **5**, 112, 1966.
3. See [1] p. 109.
4. See e.g. P. Gombás: Theorie und Lösungsmethoden des Mehrteilchenproblems der Wellenmechanik, Birkhäuser, Basel, 1950, p. 163.
5. Landolt-Börnstein, Atom- und Molekularphysik I. Band, 1. Teil, Springer, Berlin—Göttingen—Heidelberg, 1950. p. 211.
6. F. Herman and Sh. Skillman: Atomic Structure Calculations, Prentice-Hall, Inc. Englewood Cliffs, New Jersey, 1963.

ПРИМЕНЕНИЕ ВОЛНОВО-МЕХАНИЧЕСКОГО ОТТАЛКИВАЮЩЕГО ПОТЕНЦИАЛА ПРИ ОПРЕДЕЛЕНИИ ЭНЕРГИИ АТОМА Li

О. КУНВАРИ

Резюме

Первая ионизационная энергия и собственные функции (2s) атома Li определяются в приближении Хартри применением квантово-механического отталкивающего потенциала. Демонстрируется полезность псевдопотенциалъного метода.

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LÖSUNGEN DES VEREINFACHTEN SELF-CONSISTENT FIELD IN ERSTER NÄHERUNG FÜR DIE ATOME $Z=2$ BIS $Z=92$ DES PERIODISCHEN SYSTEMS DER ELEMENTE I.

Von

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FORSCHUNGSGRUPPE FÜR THEORETISCHE PHYSIK DER UNGARISCHEN AKADEMIE DER WISSENSCHAFTEN, BUDAPEST

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Es werden die Lösungen des von einem der beiden Verfasser entwickelten vereinfachten self-consistent field in erster Näherung, d. h. mit einfachen Slater-Funktionen für die Atome von $Z = 2$ bis $Z = 92$ angegeben, die durch simultane Variation aller Variationsparameter berechnet wurden. Durch Orthogonalisierung dieser Eigenfunktionen nach Schmidt erhält man gute analytische Näherungen der exakten Hartreeschen Eigenfunktionen.

The solutions in the first approximation of the simplified self-consistent field developed by one of the authors have been calculated, with simultaneous variation of all parameters in the Slater-type eigenfunctions, for the atoms from $Z = 2$ to $Z = 92$. When we orthogonalize these eigenfunctions according to Schmidt we get good analytical approximations to the exact Hartree eigenfunctions.

In zwei vorangehenden Arbeiten [1, 2] wurde von einem von uns beiden ein vereinfachtes self-consistent field für Atome entwickelt, in welchem in erster Näherung alle Elektronen einer Schale mit der Hauptquantenzahl n, unabhängig vom Wert der Nebenquantenzahl l, durch eine einheitliche radiale Eigenfunktion vom Slaterschen Typ

$$f_n(r) = A_n r^{\varkappa_n} e^{\lambda_n r} \tag{1}$$

beschrieben werden, wo $\varkappa_n$ und λ_n Variationsparameter bedeuten, die durch die Minimumsforderung der Energie festzulegen sind; r ist die Entfernung vom Kern und A_n eine Normierungskonstante, die aus der Forderung

$$\int_0^\infty f_n^2(r)\,dr = 1 \qquad (n = 1, 2, \ldots) \tag{2}$$

bestimmt wird. Das Besetzungsverbot der Elektronen der energetisch tiefer liegenden Elektronenzustände bei vorgegebener Nebenquantenzahl l wird statt der Orthogonalisierung auf die radialen Eigenfunktionen dieser Zustände durch ein Pseudopotential in Betracht gezogen.*

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* In dem in den vorliegenden Berechnungen verwendeten Pseudopotential G_l wurde das azimutale Restglied statt mit dem in [1] und [2] angegebenen Ausdruck $-ea_0/(8r^2)$ mit dem früher gebrauchten Ausdruck $-ea_0/(4r^2)$ gleichgesetzt. Der Unterschied, der sich nur für s-Zustände bemerkbar macht, ist im Falle der vorliegenden Berechnungen unbedeutend.

Für die Energie wurde der Ausdruck (17) von [1] bzw. (16, 17) von [2] zugrunde gelegt, in dem die Eigenfunktionen (1) nur durch die radialen Dichten $D_1, D_2, \ldots$ der einzelnen Elektronenschalen eingehen. In der ersten Näherung, von der wir hier ausgehen, ist die radiale Elektronendichte D_n der n-ten Elektronenschale folgendermassen definiert

$$D_n = N_n f_n^2 = N_n A_n^2 r^{2\varkappa_n} e^{-2\lambda_n r}, \tag{3}$$

wo N_n die Anzahl aller Elektronen in der n-ten Schale bedeutet.

In der vorliegenden Arbeit haben wir die Methode in dieser ersten Näherung auf alle Atome von He bis U angewendet. Ausgehend vom Energieausdruck (17) von [1] oder (16, 17) von [2] wurden die Variationsparameter $\varkappa_1, \varkappa_2, \ldots; \lambda_1, \lambda_2, \ldots$ durch *simultane* Variation aller Variationsparameter aus der Minimumsforderung der Energie bestimmt.** Die so erhaltenen Parameterwerte, und zwar das Doppelte dieser Werte, sind in der nachstehenden Tabelle zusammengestellt. $2\lambda_n$ ist in $1/a_0$-Einheiten angegeben, wo a_0 den ersten Bohrschen Wasserstoffradius bezeichnet. In der Tabelle ist auch die gemäss Formel (17) von [1] berechnete Gesamtenergie E der Atome in e^2/a_0-Einheiten angeführt; e bezeichnet die positive Elementarladung. In der Spalte EK der Tabelle steht die Elektronen-Konfiguration der betreffenden Schale.

Eine zweite Näherung und zugleich eine sehr gute Übereinstimmung mit den Hartreeschen exakten Eigenfunktionen erhält man, wenn man die hier bestimmten Eigenfunktionen f_n nach Schmidt orthogonalisiert und die so gewonnenen Eigenfunktionen, die auch von l abhängig sind, den Zuständen (n, l) zuordnet. Diesbezüglich verweisen wir auf [1] und [2], wo dies ausführlich besprochen wurde und wo auch Vergleiche einiger so gewonnenen Eigenfunktionen mit den exakten Hartreeschen zu finden sind.

Eine ausführliche Berechnung der orthonormierten Eigenfunktionen für alle Atome mit weiteren Daten folgt in einem demnächst erscheinenden II. Teil dieser Arbeit.

LITERATUR

1. P. Gombás, Theoretica Chimica Acta (Berl.) **5**, 112, 1966.
2. P. Gombás, Pseudopotentiale, S. 90—107, Springer, Wien, New-York, 1967.

** Bei der Variation von $\varkappa_1 \varkappa_2, \ldots$ wurden nur ganz- und halbzahlige Werte zugelassen.

РЕШЕНИЕ УПРОЩЕННОГО САМОСОГЛАСОВАННОГО ПОЛЯ В ПЕРВОМ ПРИБЛИЖЕНИИ ДЛЯ АТОМОВ ОТ Z=2 ДО Z=92 В ПЕРИОДИЧЕСКОЙ СИСТЕМЕ ЭЛЕМЕНТОВ I

П. ГОМБАШ и Т. СОНДИ

Резюме

В работе определяется решение упрощенного самосогласованного поля в первом приближении, разработанного одним из авторов, с одновременной вариацией всех параметров в собственных функциях типа Слейтера для атомов от Z=2 до Z=92. Ортогонализация этих собственных функций методом Шмидта результирует хорошее приближение точных собственных функций Хартри.

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Tabelle

Die doppelten Werte der Variationsparameter $\varkappa_n$ und λ_n sowie der Wert der Energie E. Die Werte für $2\lambda_n$ sind in $1/a_0$- und die für E in e^2/a_0-Einheiten angegeben

	Z	n	EK	$2\varkappa_n$	$2\lambda_n$	E
He	2	1	$(1s)^2$	2	3,375	−2,8477
Li	3	1	$(1s)^2$	2	5,396	−7,4110
		2	2s	5	1,594	
Be	4	1	$(1s)^2$	2	7,464	−14,5360
		2	$(2s)^2$	5	2,389	
B	5	1	$(1s)^2$	2	9,505	−24,3457
		2	$(2s)^2$ 2p	4	2,414	
C	6	1	$(1s)^2$	2	11,55	−37,2400
		2	$(2s)^2$ $(2p)^2$	4	3,010	
N	7	1	$(1s)^2$	2	13,60	−53,5093
		2	$(2s)^2$ $(2p)^3$	4	3,617	
O	8	1	$(1s)^2$	2	15,65	−73,4569
		2	$(2s)^2$ $(2p)^4$	4	4,233	
F	9	1	$(1s)^2$	2	17,69	−97,3865
		2	$(2s)^2$ $(2p)^5$	4	4,850	
Ne	10	1	$(1s)^2$	2	19,74	−125,633
		2	$(2s)^2$ $(2p)^6$	3	4,012	
Na	11	1	$(1s)^2$	2	21,90	−158,450
		2	$(2s)^2$ $(2p)^6$	4	6,389	
		3	3s	6	1,681	
Mg	12	1	$(1s)^2$	2	24,05	−195,684
		2	$(2s)^2$ $(2p)^6$	4	7,307	
		3	$(3s)^2$	6	2,229	
Al	13	1	$(1s)^2$	2	26,20	−237,263
		2	$(2s)^2$ $(2p)^6$	4	8,218	
		3	$(3s)^2$ 3p	6	2,538	
Si	14	1	$(1s)^2$	2	28,35	−283,445
		2	$(2s)^2$ $(2p)^6$	4	9,135	
		3	$(3s)^2$ $(3p)^2$	6	2,899	
P	15	1	$(1s)^2$	2	30,54	−334,377
		2	$(2s)^2$ $(2p)^6$	4	10,06	
		3	$(3s)^2$ $(3p)^3$	6	3,274	

	Z	n	EK	$2\varkappa_n$	$2\lambda_n$	E
S	16	1	$(1s)^2$	2	32,72	$-390{,}206$
		2	$(2s)^2$ $(2p)^6$	4	10,98	
		3	$(3s)^2$ $(3p)^4$	6	3,652	
Cl	17	1	$(1s)^2$	2	34,91	$-451{,}081$
		2	$(2s)^2$ $(2p)^6$	4	11,92	
		3	$(3s)^2$ $(3p)^5$	6	4,034	
Ar	18	1	$(1s)^2$	2	37,09	$-517{,}152$
		2	$(2s)^2$ $(2p)^6$	4	12,86	
		3	$(3s)^2$ $(3p)^6$	6	4,417	
K	19	1	$(1s)^2$	2	39,31	$-588{,}423$
		2	$(2s)^2$ $(2p)^6$	4	13,82	
		3	$(3s)^2$ $(3p)^6$	6	4,963	
		4	4s	7	1,428	
Ca	20	1	$(1s)^2$	2	41,49	$-664{,}846$
		2	$(2s)^2$ $(2p)^6$	4	14,78	
		3	$(3s)^2$ $(3p)^6$	7	6,487	
		4	$(4s)^2$	7	1,817	
Sc	21	1	$(1s)^2$	2	43,70	$-746{,}055$
		2	$(2s)^2$ $(2p)^6$	4	15,69	
		3	$(3s)^2$ $(3p)^6$ 3d	6	5,862	
		4	$(4s)^2$	7	1,877	
Ti	22	1	$(1s)^2$	2	45,87	$-832{,}770$
		2	$(2s)^2$ $(2p)^6$	4	16,61	
		3	$(3s)^2$ $(3p)^6$ $(3d)^2$	6	6,225	
		4	$(4s)^2$	7	1,953	
V	23	1	$(1s)^2$	2	48,08	$-925{,}098$
		2	$(2s)^2$ $(2p)^6$	4	17,51	
		3	$(3s)^2$ $(3p)^6$ $(3d)^3$	6	6,594	
		4	$(4s)^2$	7	2,033	
Cr	24	1	$(1s)^2$	2	50,22	$-1\,022{,}55$
		2	$(2s)^2$ $(2p)^6$	4	18,37	
		3	$(3s)^2$ $(3p)^6$ $(3d)^5$	5	5,603	
		4	4s	6	1,450	
Mn	25	1	$(1s)^2$	2	52,44	$-1\,127{,}27$
		2	$(2s)^2$ $(2p)^6$	4	19,32	
		3	$(3s)^2$ $(3p)^6$ $(3d)^5$	5	6,071	
		4	$(4s)^2$	6	1,830	

	Z	n	EK	$2\varkappa_n$	$2\lambda_n$	E
Fe	26	1	$(1s)^2$	2	54,63	—1 237,40
		2	$(2s)^2$ $(2p)^6$	4	20,25	
		3	$(3s)^2$ $(3p)^6$ $(3d)^6$	5	6,391	
		4	$(4s)^2$	6	1,893	
Co	27	1	$(1s)^2$	2	56,83	—1 353,67
		2	$(2s)^2$ $(2p)^6$	4	21,16	
		3	$(3s)^2$ $(3p)^6$ $(3d)^7$	5	6,715	
		4	$(4s)^2$	6	1,954	
Ni	28	1	$(1s)^2$	2	59,03	—1 476,23
		2	$(2s)^2$ $(2p)^6$	4	22,08	
		3	$(3s)^2$ $(3p)^6$ $(3d)^8$	5	7,039	
		4	$(4s)^2$	6	2,013	
Cu	29	1	$(1s)^2$	2	61,20	—1 604,14
		2	$(2s)^2$ $(2p)^6$	4	22,94	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	5	7,212	
		4	4s	6	1,676	
Zn	30	1	$(1s)^2$	2	63,45	—1 740,69
		2	$(2s)^2$ $(2p)^6$	4	23,91	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	5	7,694	
		4	$(4s)^2$	6	2,129	
Ga	31	1	$(1s)^2$	2	65,67	—1 883,46
		2	$(2s)^2$ $(2p)^6$	4	24,87	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	5	8,168	
		4	$(4s)^2$ 4p	6	2,347	
Ge	32	1	$(1s)^2$	2	67,89	—2 032,67
		2	$(2s)^2$ $(2p)^6$	4	25,85	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	5	8,649	
		4	$(4s)^2$ $(4p)^2$	6	2,606	
As	33	1	$(1s)^2$	2	70,12	—2 188,41
		2	$(2s)^2$ $(2p)^6$	4	26,82	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	5	9,128	
		4	$(4s)^2$ $(4p)^3$	6	2,868	
Se	34	1	$(1s)^2$	2	72,36	—2 350,77
		2	$(2s)^2$ $(2p)^6$	4	27,80	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	5	9,610	
		4	$(4s)^2$ $(4p)^4$	6	3,124	

	Z	n	EK	$2\varkappa_n$	$2\lambda_n$	E
Br	35	1	$(1s)^2$	2	74,55	−2 519,86
		2	$(2s)^2$ $(2p)^6$	4	28,79	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	5	10,09	
		4	$(4s)^2$ $(4p)^5$	6	3,378	
Kr	36	1	$(1s)^2$	2	76,80	−2 695,76
		2	$(2s)^2$ $(2p)^6$	4	29,76	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	5	10,57	
		4	$(4s)^2$ $(4p)^6$	6	3,628	
Rb	37	1	$(1s)^2$	2	79,04	−2 878,33
		2	$(2s)^2$ $(2p)^6$	4	30,75	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	5	11,06	
		4	$(4s)^2$ $(4p)^6$	6	3,997	
		5	5s	7	1,253	
Sr	38	1	$(1s)^2$	2	81,30	−3 067,79
		2	$(2s)^2$ $(2p)^6$	4	31,82	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	6	13,88	
		4	$(4s)^2$ $(4p)^6$	7	5,121	
		5	$(5s)^2$	8	1,803	
Y	39	1	$(1s)^2$	2	83,56	−3 263,60
		2	$(2s)^2$ $(2p)^6$	4	32,82	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	6	14,45	
		4	$(4s)^2$ $(4p)^6$ 4d	7	5,290	
		5	$(5s)^2$	8	1,851	
Zr	40	1	$(1s)^2$	2	85,77	−3 466,39
		2	$(2s)^2$ $(2p)^6$	4	33,80	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	6	15,03	
		4	$(4s)^2$ $(4p)^6$ $(4d)^2$	7	5,473	
		5	$(5s)^2$	7	1,646	
Nb	41	1	$(1s)^2$	2	87,99	− 3 675,62
		2	$(2s)^2$ $(2p)^6$	4	34,81	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	6	15,59	
		4	$(4s)^2$ $(4p)^6$ $(4d)^4$	7	5,496	
		5	5s	7	1,417	
Mo	42	1	$(1s)^2$	2	90,26	−3 892,65
		2	$(2s)^2$ $(2p)^6$	5	44,75	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	6	16,28	
		4	$(4s)^2$ $(4p)^6$ $(4d)^5$	7	5,743	
		5	5s	7	1,457	

	Z	n	EK	$2\varkappa_n$	$2\lambda_n$	E
Tc	43	1	$(1s)^2$	2	92,72	—4 117,20
		2	$(2s)^2$ $(2p)^6$	5	45,87	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	6	16,84	
		4	$(4s)^2$ $(4p)^6$ $(4d)^6$	7	5,959	
		5	5s	7	1,497	
Ru	44	1	$(1s)^2$	2	95,01	—4 349,00
		2	$(2s)^2$ $(2p)^6$	5	47,14	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	6	17,42	
		4	$(4s)^2$ $(4p)^6$ $(4d)^7$	7	6,183	
		5	5s	7	1,534	
Rh	45	1	$(1s)^2$	2	97,24	—4 588,17
		2	$(2s)^2$ $(2p)^6$	5	48,39	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	6	18,00	
		4	$(4s)^2$ $(4p)^6$ $(4d)^8$	7	6,410	
		5	5s	7	1,569	
Pd	46	1	$(1s)^2$	2	99,54	—4 833,90
		2	$(2s)^2$ $(2p)^6$	5	49,63	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	6	18,57	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$	7	6,502	
Ag	47	1	$(1s)^2$	2	101,8	—5 088,94
		2	$(2s)^2$ $(2p)^6$	5	50,88	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	6	19,17	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$	7	6,869	
		5	5s	6	1,382	
Cd	48	1	$(1s)^2$	2	104,0	—5 351,32
		2	$(2s)^2$ $(2p)^6$	5	52,17	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	6	19,77	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$	7	7,227	
		5	$(5s)^2$	6	1,710	
In	49	1	$(1s)^2$	2	106,3	—5 621,59
		2	$(2s)^2$ $(2p)^6$	5	53,49	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	23,81	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$	8	8,801	
		5	$(5s)^2$ 5p	7	2,241	
Sn	50	1	$(1s)^2$	2	108,7	—5 899,30
		2	$(2s)^2$ $(2p)^6$	5	54,78	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	24,52	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$	8	9,204	
		5	$(5s)^2$ $(5p)^2$	7	2,463	

	Z	n	EK	$2\varkappa_n$	$2\lambda_n$	E
Sb	51	1	$(1s)^2$	2	110,9	$-6\,184{,}56$
		2	$(2s)^2$ $(2p)^6$	5	56,05	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	25,21	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$	8	9,604	
		5	$(5s)^2$ $(5p)^3$	7	2,683	
Te	52	1	$(1s)^2$	2	113,2	$-6\,477{,}50$
		2	$(2s)^2$ $(2p)^6$	5	57,31	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	25,92	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$	9	11,34	
		5	$(5s)^2$ $(5p)^4$	8	3,353	
J	53	1	$(1s)^2$	2	115,5	$-6\,778{,}21$
		2	$(2s)^2$ $(2p)^6$	5	58,62	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	26,62	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$	9	11,78	
		5	$(5s)^2$ $(5p)^5$	8	3,584	
Xe	54	1	$(1s)^2$	2	117,8	$-7\,086{,}67$
		2	$(2s)^2$ $(2p)^6$	5	59,90	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	27,35	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$	9	12,24	
		5	$(5s)^2$ $(5p)^6$	8	3,815	
Cs	55	1	$(1s)^2$	2	120,0	$-7\,402{,}71$
		2	$(2s)^2$ $(2p)^6$	5	61,17	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	28,05	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$	9	12,68	
		5	$(5s)^2$ $(5p)^6$	8	4,136	
		6	6s	8	1,249	
Ba	56	1	$(1s)^2$	2	122,3	$-7\,726{,}39$
		2	$(2s)^2$ $(2p)^6$	5	62,49	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	28,76	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$	9	13,14	
		5	$(5s)^2$ $(5p)^6$	9	5,028	
		6	$(6s)^2$	9	1,724	
La	57	1	$(1s)^2$	2	124,7	$-8\,057{,}58$
		2	$(2s)^2$ $(2p)^6$	5	63,78	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	29,48	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$	9	13,58	
		5	$(5s)^2$ $(5p)^6$ 5d	9	5,196	
		6	$(6s)^2$	9	1,770	

	Z	n	EK	$2\varkappa_n$	$2\lambda_n$	E
Ce	58	1	$(1s)^2$	2	126,9	—8 395,47
		2	$(2s)^2$ $(2p)^6$	5	65,03	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	30,14	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ 4f	9	13,88	
		5	$(5s)^2$ $(5p)^6$ 5d	9	5,280	
		6	$(6s)^2$	9	1,789	
Pr	59	1	$(1s)^2$	2	129,1	—8 741,44
		2	$(2s)^2$ $(2p)^6$	5	66,27	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	30,78	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^2$	9	14,18	
		5	$(5s)^2$ $(5p)^6$ 5d	9	5,356	
		6	$(6s)^2$	9	1,809	
Nd	60	1	$(1s)^2$	2	131,4	—9 095,59
		2	$(2s)^2$ $(2p)^6$	5	67,52	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	31,45	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^3$	9	14,48	
		5	$(5s)^2$ $(5p)^6$ 5d	9	5,440	
		6	$(6s)^2$	9	1,826	
Pm	61	1	$(1s)^2$	2	133,7	—9 458,13
		2	$(2s)^2$ $(2p)^6$	5	68,77	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	32,09	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^4$	8	13,08	
		5	$(5s)^2$ $(5p)^6$ 5d	8	4,870	
		6	$(6s)^2$	8	1,613	
Sm	62	1	$(1s)^2$	2	136,0	—9 829,18
		2	$(2s)^2$ $(2p)^6$	5	70,07	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	32,74	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^5$	8	13,36	
		5	$(5s)^2$ $(5p)^6$ 5d	8	4,941	
		6	$(6s)^2$	8	1,628	
Eu	63	1	$(1s)^2$	2	138,3	—10 208,6
		2	$(2s)^2$ $(2p)^6$	5	71,33	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	33,39	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^6$	8	13,63	
		5	$(5s)^2$ $(5p)^6$ 5d	8	5,016	
		6	$(6s)^2$	8	1,642	

	Z	n	EK	$2\varkappa_n$	$2\lambda_n$	E
Gd	64	1	$(1s)^2$	2	140,5	—10 596,5
		2	$(2s)^2$ $(2p)^6$	5	72,58	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	34,04	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^7$	8	13,91	
		5	$(5s)^2$ $(5p)^6$ 5d	8	5,082	
		6	$(6s)^2$	8	1,656	
Tb	65	1	$(1s)^2$	2	142,8	—10 993,0
		2	$(2s)^2$ $(2p)^6$	5	73,84	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	34,73	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^8$	8	14,19	
		5	$(5s)^2$ $(5p)^6$ 5d	8	5,154	
		6	$(6s)^2$	8	1,673	
Dy	66	1	$(1s)^2$	2	145,1	—11 398,1
		2	$(2s)^2$ $(2p)^6$	5	75,05	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	35,33	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^9$	7	12,60	
		5	$(5s)^2$ $(5p)^6$ 5d	6	3,921	
		6	$(6s)^2$	7	1,462	
Ho	67	1	$(1s)^2$	2	147,4	—11 812,3
		2	$(2s)^2$ $(2p)^6$	5	76,36	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	35,97	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{10}$	7	12,84	
		5	$(5s)^2$ $(5p)^6$ 5d	6	3,971	
		6	$(6s)^2$	7	1,470	
Er	68	1	$(1s)^2$	2	149,5	—12 235,2
		2	$(2s)^2$ $(2p)^6$	5	77,62	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	36,64	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{11}$	7	13,09	
		5	$(5s)^2$ $(5p)^6$ 5d	6	4,022	
		6	$(6s)^2$	7	1,482	
Tu	69	1	$(1s)^2$	2	151,9	—12 666,9
		2	$(2s)^2$ $(2p)^6$	5	78,88	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	37,30	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{12}$	7	13,34	
		5	$(5s)^2$ $(5p)^6$ 5d	6	4,072	
		6	$(6s)^2$	7	1,495	

	Z	n	EK	$2\varkappa_n$	$2\lambda_n$	E
Yb	70	1	$(1s)^2$	2	154,1	—13 107,6
		2	$(2s)^2$ $(2p)^6$	5	80,10	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	37,94	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{13}$	7	13,59	
		5	$(5s)^2$ $(5p)^6$ 5d	6	4,121	
		6	$(6s)^2$	7	1,510	
Lu	71	1	$(1s)^2$	2	156,4	—13 557,2
		2	$(2s)^2$ $(2p)^6$	5	81,42	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	38,61	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	7	13,84	
		5	$(5s)^2$ $(5p)^6$ 5d	6	4,167	
		6	$(6s)^2$	7	1,519	
Hf	72	1	$(1s)^2$	2	158,7	—14 017,2
		2	$(2s)^2$ $(2p)^6$	5	82,68	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	39,32	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	7	14,21	
		5	$(5s)^2$ $(5p)^6$ $(5d)^2$	7	4,984	
		6	$(6s)^2$	7	1,549	
Ta	73	1	$(1s)^2$	2	161,1	—14 486,1
		2	$(2s)^2$ $(2p)^6$	5	83,95	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	40,03	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	7	14,58	
		5	$(5s)^2$ $(5p)^6$ $(5d)^3$	7	5,127	
		6	$(6s)^2$	7	1,586	
W	74	1	$(1s)^2$	2	163,3	—14 963,8
		2	$(2s)^2$ $(2p)^6$	5	85,28	
		3	$(3d)^2$ $(3p)^6$ $(3d)^{10}$	7	40,74	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	7	14,95	
		5	$(5s)^2$ $(5p)^6$ $(5d)^4$	7	5,277	
		6	$(6s)^2$	7	1,624	
Re	75	1	$(1s)^2$	2	165,7	—15 450,5
		2	$(2s)^2$ $(2p)^6$	5	86,61	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	41,45	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	7	15,31	
		5	$(5s)^2$ $(5p)^6$ $(5d)^5$	7	5,427	
		6	$(6s)^2$	7	1,663	

	Z	n	EK	$2\varkappa_n$	$2\lambda_n$	E
Os	76	1	$(1s)^2$	2	167,9	−15 946,2
		2	$(2s)^2$ $(2p)^6$	5	87,88	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	42,17	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	7	15,68	
		5	$(5s)^2$ $(5p)^6$ $(5d)^6$	7	5,583	
		6	$(6s)^2$	7	1,699	
Ir	77	1	$(1s)^2$	2	170,2	−16 451,0
		2	$(2s)^2$ $(2p)^6$	5	89,16	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	42,90	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	7	16,05	
		5	$(5s)^2$ $(5p)^6$ $(5d)^7$	7	5,744	
		6	$(6s)^2$	7	1,733	
Pt	78	1	$(1s)^2$	2	172,5	−16 964,8
		2	$(2s)^2$ $(2p)^6$	5	90,50	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	43,59	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	7	16,41	
		5	$(5s)^2$ $(5p)^6$ $(5d)^9$	7	5,905	
		6	6s	7	1,771	
Au	79	1	$(1s)^2$	2	174,9	−17 487,6
		2	$(2s)^2$ $(2p)^6$	5	91,78	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	44,31	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	7	16,77	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	7	5,957	
		6	6s	6	1,283	
Hg	80	1	$(1s)^2$	2	177,1	−18 020,1
		2	$(2s)^2$ $(2p)^6$	5	93,06	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	45,05	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	7	17,15	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	7	6,228	
		6	$(6s)^2$	6	1,570	
Tl	81	1	$(1s)^2$	2	179,4	−18 561,9
		2	$(2s)^2$ $(2p)^6$	5	94,46	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	45,86	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	8	20,06	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	8	7,442	
		6	$(6s)^2$ 6p	7	2,030	

	Z	n	EK	$2\varkappa_n$	$2\lambda_n$	E
Pb	82	1	$(1s)^2$	2	181,6	−19 112,7
		2	$(2s)^2$ $(2p)^6$	5	95,75	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	46,60	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	8	20,48	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	8	7,729	
		6	$(6s)^2$ $(6p)^2$	7	2,220	
Bi	83	1	$(1s)^2$	2	184,1	−19 672,6
		2	$(2s)^2$ $(2p)^6$	5	97,04	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	47,33	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	8	20,92	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	8	8,018	
		6	$(6s)^2$ $(6p)^3$	7	2,406	
Po	84	1	$(1s)^2$	2	186,3	−20 241,8
		2	$(2s)^2$ $(2p)^6$	5	98,39	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	48,05	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	8	21,36	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	9	9,373	
		6	$(6s)^2$ $(6p)^4$	8	2,967	
At	85	1	$(1s)^2$	2	188,7	−20 820,1
		2	$(2s)^2$ $(2p)^6$	5	99,69	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	48,77	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	8	21,78	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	9	9,685	
		6	$(6s)^2$ $(6p)^5$	8	3,158	
Rn	86	1	$(1s)^2$	2	191,0	−21 407,6
		2	$(2s)^2$ $(2p)^6$	5	101,0	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	49,50	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	8	22,20	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	9	9,996	
		6	$(6s)^2$ $(6p)^6$	8	3,343	
Fr	87	1	$(1s)^2$	2	193,4	−22 004,2
		2	$(2s)^2$ $(2p)^6$	5	102,3	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	50,22	
		4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	8	22,63	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	9	10,31	
		6	$(6s)^2$ $(6p)^6$	8	3,610	
		7	7s	9	1,296	

	Z	n	EK	$2\varkappa_n$	$2\lambda_n$	E
		1	$(1s)^2$	2	195,6	$-22\,609{,}8$
		2	$(2s)^2$ $(2p)^6$	5	103,6	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	50,95	
Ra	88	4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	8	23,06	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	9	10,63	
		6	$(6s)^2$ $(6p)^6$	9	4,353	
		7	$(7s)^2$	9	1,557	
		1	$(1s)^2$	2	197,8	$-23\,224{,}6$
		2	$(2s)^2$ $(2p)^6$	5	104,9	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	51,71	
Ac	89	4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	8	23,51	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	10	12,17	
		6	$(6s)^2$ $(6p)^6$ 6d	10	5,002	
		7	$(7s)^2$	9	1,603	
		1	$(1s)^2$	2	200,3	$-23\,848{,}8$
		2	$(2s)^2$ $(2p)^6$	5	106,2	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	52,44	
Th	90	4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	8	23,94	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	10	12,51	
		6	$(6s)^2$ $(6p)^6$ $(6d)^2$	10	5,165	
		7	$(7s)^2$	9	1,651	
		1	$(1s)^2$	2	202,5	$-24\,482{,}3$
		2	$(2s)^2$ $(2p)^6$	5	107,5	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	53,17	
Pa	91	4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	8	24,37	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	10	12,85	
		6	$(6s)^2$ $(6p)^6$ $(6d)^3$	10	5,327	
		7	$(7s)^2$	9	1,691	
		1	$(1s)^2$	2	204,9	$-25\,125{,}2$
		2	$(2s)^2$ $(2p)^6$	5	108,9	
		3	$(3s)^2$ $(3p)^6$ $(3d)^{10}$	7	53,90	
U	92	4	$(4s)^2$ $(4p)^6$ $(4d)^{10}$ $(4f)^{14}$	8	24,79	
		5	$(5s)^2$ $(5p)^6$ $(5d)^{10}$	10	13,18	
		6	$(6s)^2$ $(6p)^6$ $(6d)^4$	9	4,830	
		7	$(7s)^2$	8	1,309	

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ÜBER DIE WEIZSÄCKERSCHE KINETISCHE ENERGIEKORREKTION

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Es wird für den Fall einer Inhomogenitätskorrektion die statistische Berechnung der kinetischen Energie eines Elektronengases untersucht und gezeigt, dass man bei einer einfachen Addition des Fermischen Anteils und des Weizsäckerschen Inhomogenitätsanteils der kinetischen Energie einen Fehler begeht. Dieser wird für einen einfachen Fall berechnet. Weiterhin wird gezeigt, dass einige bisherige von mehreren Autoren unternommene Versuche diesen Fehler durch den an dem Weizsäckerschen Anteil angebrachten Korrektionsfaktor 1/9 zu korrigieren zu unbefriedigenden Energiewerten der Atome führt: die mit diesem Korrektionsfaktor berechneten Energien liegen für schwere Atome um $\sim 8\%$, für mittelschwere um $\sim 12\%$ und für leichte um $\sim 23\%$ zu tief.

1. Einleitung

In der vorliegenden Arbeit setzen wir uns zum Ziel zu zeigen, dass bei einer statistischen Behandlungsweise eines Elektronengases die Fermische Nullpunktsenergie und die aus der Inhomogenität der Dichte resultierende Weizsäckersche kinetische Inhomogenitätskorrektion nicht einfach addiert werden können, da ein Teil der Energie in beiden Energieanteilen enthalten ist. Die Fermische kinetische Energie enthält nämlich nicht nur den aus dem Paulischen Besetzungsverbot resultierenden Anteil (demzufolge die Endpunkte der Impulsvektoren der verschiedenen Bahnzustände in verschiedene Impulsraumzellen fallen müssen), sondern auch den aus der endlichen Breite der Impulsraumzellen resultierenden Anteil, der dem Weizsäckerschen Anteil entspricht. Wenn man also den Fermischen und den Weizsäckerschen Anteil einfach addiert, wie dies häufig geschah und geschieht, so zählt man diesen Anteil doppelt.

Dass der Fermische und Weizsäckersche kinetische Energieanteil nicht einfach addiert werden können, tritt am bezeichnendsten im extremen Fall von nur zweier Elektronen im Grundzustand, z. B. der beiden Elektronen im Grundzustand des He-Atoms, zu Tage. In diesem Fall müsste nämlich der Fermische kinetische Energieanteil verschwinden und der exakte kinetische Energieausdruck wird durch den Weizsäckerschen Anteil allein dargestellt.

Der Fehler, den man bei der Berechnung der kinetischen Energie durch die einfache Addition des Fermischen und Weizsäckerschen Energieanteils infolge der eben erläuterten Ursachen begeht, wurde von mehreren Autoren korrigiert, jedoch meistens so, dass sie an den Weizsäckerschen Anteil einen Faktor <1 angebracht haben. Von mehreren Autoren wurde für diesen Faktor der Wert 1/9 hergeleitet ohne aber, dass kontrolliert wurde, zu welchem Resultat dieser Faktor führt.[1] Mit dem einfachen, jedoch sehr flexiblen Ansatz für die Elektronendichte $\varrho = Ae^{-(\lambda r)^{1/n}}$, wo λ und n Variationsparameter sind und A eine Normierungskonstante bezeichnet, erhält man mit diesem Faktor z. B. für die Atome Ne($Z = 10$), Kr($Z = 36$) und Hg($Z = 80$) der Reihe nach die folgenden Energiewerte $-160{,}7\ e^2/a_0$, $-3050\ e^2/a_0$ und $-19\,500\ e^2/a_0$ wo e die positive Elementarladung und a_0 den ersten Bohrschen Wasserstoffradius bezeichnen, während die empirischen bzw. halbempirischen Werte der Reihe nach $-129{,}5\ e^2/a_0$, $-2704\ e^2/a_0$ und $-18\,130\ e^2/a_0$ sind. Die mit dem Korrektionsfaktor 1/9 erhaltenen Energien liegen also besonders für mittlere und kleinere Z-Werte viel zu tief; dieser Korrektionsfaktor ist also unbrauchbar.

Diese Art der Korrektion, durch einen an dem Weizsäckerschen Energieanteil angebrachten Korrektionsfaktor, ist jedoch meiner Ansicht nach auch aus folgendem Grunde nicht befriedigend. Aus der Variation des Energieausdruckes hinsichtlich der Teilchendichte ϱ gelangt man nämlich zu einem Zusammenhang zwischen ϱ und dem Potential. Dieser ist der grundlegende Zusammenhang der statistischen Theorie und muss — wenn man $\psi = \sqrt{\varrho}$ als Eigenfunktion betrachtet — mit einer erweiterten Schrödingerschen Gleichung für das Elektron mit höchster Energie identisch sein, falls man statt dem elektrostatischen Potential V das mit dem Besetzungsverbotpotential $-(5\,\varkappa_k/3\,e)\,\psi^{4/3}$ und dem Austauschpotential $(4\varkappa_a/3e)\,\psi^{2/3}$ ergänzte sogenannte modifizierte Potential

$$\Phi = V - \frac{5}{3}\,\frac{\varkappa_k}{e}\,\psi^{4/3} + \frac{4}{3}\,\frac{\varkappa_a}{e}\,\psi^{2/3} \tag{1}$$

setzt, wo $\varkappa_k$ und $\varkappa_a$ die folgenden universellen Konstanten

$$\varkappa_k = \frac{3}{10}\,(3\,\pi^2)^{2/3}\,e^2\,a_0 \qquad \text{und} \qquad \varkappa_a = \frac{3}{4}\left(\frac{3}{\pi}\right)^{1/3} e^2 \tag{2}$$

bedeuten. Die erweiterte Schrödinger-Gleichung hat die Form

$$\frac{1}{2}\,e^2\,a_0\,\Delta\psi + (\varepsilon + \Phi e)\,\psi = 0\,, \tag{3}$$

wo ε den Energieparameter bezeichnet. Der Energieeigenwert, der sich mit der Normierungsbedingung für ϱ aus dieser Gleichung ergibt, ist die Energie des Elektrons im höchsten Energiezustand.

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[1] D. A. KIRZHNITS, Journ. exp. theoret. Phys. URSS **32**, 115, 1957; K. J. LE COUTEUR, Proc. Phys. Soc. **84**, 837, 1964.

Falls man den Weizsäckerschen Energieanteil nach dem weiter oben erwähnten Verfahren durch einen Korrektionsfaktor korrigiert, so tritt dieser auch im ersten Glied auf der linken Seite in (3) auf und die Grundgleichung (1) kann dann nur mehr schwerlich als die erweiterte Schrödinger-Gleichung des Elektrons mit der höchsten Energie des Elektrons interpretiert werden.

Hauptsächlich aus diesem Grunde war ich immer der Ansicht,[2] dass man den oben erwähnten Fehler, der bei der einfachen Addition des Fermischen und Weizsäckerschen Energieanteils entsteht, nicht einfach so korrigieren kann, dass man an dem Weizsäckerschen Energieanteil einen Zahlenfaktor <1 anbringt, sondern man sollte vielmehr den Fermischen Anteil korrigieren, wodurch die Schrödinger-Gleichung (3) erhalten bleibt und eine Änderung nur im Besetzungsverbotpotential, d. h. im zweiten Glied auf der rechten Seite von (1) entsteht. Auf diese Weise ist es mir tatsächlich gelungen eine Korrektion einzuführen, die zwar von den leichtesten bis zu den schwersten Atomen die Energien der Atome mit einem Fehler $<2\%$ wiedergibt, und nicht nur für Atome sondern auch für Kerne zu Dichteverteilungen führt, die mit der Erfahrung gut übereinstimmen,[3] die jedoch auf der wenig befriedigenden Annahme beruht, dass die radiale Impulsbreite einer Elementarzelle im Impulsraum dieselbe ist wie die azimutale.

Zur Herleitung der Korrektion wurde meinerseits in 1953 auch noch ein anderer Weg eingeschlagen,[4] der ebenfalls zur Korrektion des Fermischen Anteils führt. Das erhaltene Resultat, das sich auf einen vereinfachten Fall bezieht, schien mir jedoch damals für rechnerische Zwecke als wenig verwertbar, konnte aber in letzterer Zeit für Berechnungen verwendet werden. Übrigens weist die damals gegebene Herleitung dieser Korrektion einige beträchtliche Mängel auf, wodurch jedoch glücklicherweise das Endresultat nicht beeinflusst wird. Es sei daher die Herleitung in einwandfreier Form für denselben vereinfachten Fall hier nochmals wiedergegeben. Das Endresultat wollen wir in einer demnächst folgenden Arbeit in eine für rechnerische Zwecke brauchbare Form bringen und auf Atome anwenden.

2. Berechnung der kinetischen Energie für einen vereinfachten Fall

Unsere folgenden Betrachtungen beziehen sich auf ein Atom mit vielen Elektronen, die wir im späteren Verlauf statistisch behandeln wollen. Hierzu befassen wir uns zunächst mit den Elektronen des Atoms in einer Kugelschale und gehen von der Schrödinger-Gleichung für den radialen Teil der Eigenfunktion eines Elektrons mit der Nebenquantenzahl l aus, die folgendermassen lautet

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[2] P. GOMBÁS, Acta Phys. Hung. **3**, 106, 1953.
[3] M. TISZA, Acta Phys. Hung. im Erscheinen.
[4] P. GOMBÁS, Acta Phys. Hung. **3**, 106, 1953.

$$\frac{1}{2}e^2 a_0\left[\frac{d^2 f}{dr^2} - \frac{l(l+1)}{r^2}f\right] + (\varepsilon + Ve)f = 0, \tag{4}$$

wo f die radiale Wellenfunktion (d. h. das r-fache des radialen Anteils der üblichen Wellenfunktion), und ε den Energieparameter bezeichnen, V ist das elektrostatische Potential, das im Atom auf das in Betracht gezogene Elektron wirkt.

Wir unterteilen nun die Elektronenwolke des Atoms durch konzentrische Kugelflächen, deren gemeinsames Zentrum sich im Kern befindet, in Kugelschalen von der Dicke s. Diese seien so bemessen, dass in den Kugelschalen V und $1/r^2$ als »fast« konstant betrachtet werden können. Abweichungen vom konstanten Wert seien von erster Ordnung klein. Für ein Elektron in der k-ten Kugelschale kann man dann die Lösungen von (4), die den Randbedingungen — nach welchen f an den beiden Berandungsflächen der Kugelschale verschwinden muss — genügen, folgendermassen ansetzen

$$f_\lambda = A \sin\frac{\lambda\pi}{s}(r - r_k) = A \sin \lambda x, \tag{5}$$

wobei wir zur Abkürzung die Variable

$$x = \frac{\pi}{s}(r - r_k) \tag{6}$$

einführten und wo r_k den Radius der inneren Kugelfläche und λ eine ganze Zahl

$$\lambda = 1, 2, \ldots, n \tag{7}$$

bedeuten. Für die Amplitude A müssen wir — wegen der nicht vollkommenen Konstanz von V und $1/r^2$ — noch eine schwache Abhängigkeit von r voraussetzen. Wir setzen in der k-ten Kugelschale eine Entwicklung von A nach Potenzen vom $r - r_k$ an, die wir nach dem dritten Glied abbrechen, es wird also

$$\begin{aligned} A(r) &= A_0 + \left(\frac{\partial A}{\partial r}\right)_0 (r - r_k) + \frac{1}{2}\left(\frac{\partial^2 A}{\partial r^2}\right)_0 (r - r_k)^2 = \\ &= A_0 + (A')_0\, x + \frac{1}{2}(A'')_0\, x^2, \end{aligned} \tag{8}$$

wo A' und A'' die erste und zweite Ableitung von A nach x bedeuten und der Index 0 sich auf die innere Berandungsfläche ($r = r_k$, $x = 0$) bezieht.

Die radiale kinetische Energie η_λ eines Elektrons der k-ten Kugelschale im Zustand (λ, l) können wir aus der folgenden Formel berechnen

$$\eta_\lambda = \frac{1}{2} e^2 a_0 \frac{\int_{r_k}^{r_k+s} \left(\frac{\partial f_\lambda}{\partial r} \right)^2 dr}{\int_{r_k}^{r_k+s} f_\lambda^2 \, dr} = \frac{\pi^2}{2s^2} e^2 a_0 \frac{\int_0^\pi (f'_\lambda)^2 \, dx}{\int_0^\pi f_\lambda^2 \, dx}, \tag{9}$$

wo f'_λ die erste Ableitung von f_λ nach x bezeichnet, für die sich der Ausdruck

$$f'_\lambda = A' \sin \lambda x + A \lambda \cos \lambda x \tag{10}$$

ergibt.

Nach Einsetzen dieses Ausdruckes in (9) erhält man

$$\eta_\lambda = I_1 + I_2 + I_3 \tag{11}$$

mit

$$I_1 = \frac{\pi^2}{2s^2} e^2 a_0 \frac{\int_0^\pi (A')^2 \sin^2 \lambda x \, dx}{\int_0^\pi A^2 \sin^2 \lambda x \, dx} \tag{12}$$

$$I_2 = \frac{\pi^2}{2s^2} e^2 a_0 \lambda \frac{\int_0^\pi 2 \, A A' \sin \lambda x \cos \lambda x \, dx}{\int_0^\pi A^2 \sin^2 \lambda x \, dx} = \frac{\pi^2}{2s^2} e^2 a_0 \lambda \frac{\int_0^\pi A A' \sin 2 \lambda x \, dx}{\int_0^\pi A^2 \sin^2 \lambda x \, dx} \tag{13}$$

und

$$I_3 = \frac{\pi^2}{2\,s^2} e^2 a_0 \lambda^2 \frac{\int_0^\pi A^2 \cos^2 \lambda x \, dx}{\int_0^\pi A^2 \sin^2 \lambda x \, dx} . \tag{14}$$

In den bisherigen Arbeiten[5] ist man immer so vorgegangen, dass man im Integrationsbereich A und A' als konstant betrachtet hat, wodurch man mit Rücksicht darauf, dass das Integral von sin $2\lambda x$ von 0 bis π verschwindet, die folgenden Resultate erhält: $I_1 = \frac{\pi^2}{2s^2} e^2 a_0 (A')^2/A^2$, $I_2 = 0$, $I_3 = \frac{\pi^2}{2s^2} e^2 a_0 \lambda^2$. Für die Gesamtenergie η_λ ergibt sich so

$$\eta_\lambda = \frac{\pi^2}{2s^2} e^2 a_0 \left[\lambda^2 + \left(\frac{A'}{A} \right)_0^2 \right] \cdot \tag{15}$$

Obwohl dieses Resultat richtig ist, ist die Herleitung dieses Resultates unrichtig. Zur richtigen Herleitung müssen wir zunächst folgendes klarstellen. Wir befassen uns hier mit dem Fall, dass die Amplitude A nur wenig veränderlich ist. Man wird also A' als eine im Verhältnis zu A von erster Ordnung kleine Grösse betrachten. Da jedoch das Resultat zu $(A')^2$ proportional, d. h.

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[5] H. HELLMANN, Acta Physicochimica URSS **4**, 225, 1936; P. GOMBÁS, Die statistische Theorie d. Atoms u. ihre Anwendungen, S. 110ff, Springer, Wien, 1949.

eine von zweiter Ordnung kleine Grösse ist, hat man bei der Entwicklung von A in (8) auch noch das von zweiter Ordnung kleine Glied A'' zu berücksichtigen. Es ist also keinesfalls zulässig, dass man in den Integralen in den Ausdrücken I_1, I_2 und I_3 A und A' als konstant betrachtet, wie dies in den bisherigen Herleitungen geschah, sondern man hat die Entwicklung (8) zugrunde zu legen und die Integrale bis zu Grössen zu berechnen, die von zweiter Ordnung klein sind, man hat also z. B. in A^2 auch noch das zu $A_0(A'')_0\,x^2$ proportionale Glied zu berücksichtigen.

Nach diesen Festsetzungen bleibt nur noch die Berechnung der Integrale in I_1, I_2 und I_3, die ganz elementar ist und zu folgendem Resultat führt:

$$I_1 = \frac{\pi^2}{2\,s^2}\,e^2 a_0 \left(\frac{A'}{A}\right)_0^2, \tag{16}$$

$$I_2 = -\frac{\pi^2}{2\,s^2}\,e^2 a_0 \left[\left(\frac{A'}{A}\right)_0^2 + \left(\frac{A''}{A}\right)_0\right], \tag{17}$$

$$I_3 = \frac{\pi^2}{2\,s^2}\,e^2 a_0 \left[\lambda^2 + \left(\frac{A'}{A}\right)_0^2 + \left(\frac{A''}{A}\right)_0\right], \tag{18}$$

womit für die Gesamtenergie der Ausdruck

$$\eta_\lambda = \frac{\pi^2}{2\,s^2}\,e^2 a_0 \left[\lambda^2 + \left(\frac{A'}{A}\right)_0^2\right] \tag{19}$$

folgt, der überraschenderweise mit dem Ausdruck (15) identisch ist, den wir mit einer nicht einwandfreien Herleitung erhalten haben.

Wenn wir in (19) $a_0 = h^2/(4\,\pi^2\,me^2)$ setzen, wo h die Plancksche Konstante und m die Masse des Elektrons bezeichnen, so ergibt sich der Ausdruck

$$\eta_\lambda = \frac{p_r^2}{2\,m} + \frac{h^2}{8\,s^2\,m}\left(\frac{A'}{A}\right)_0^2, \tag{20}$$

in welchem $p_r = \lambda\,h/(2s)$ den radialen Impuls des Elektrons bezeichnet. Hieraus ist unmittelbar ersichtlich, dass das erste Glied auf der rechten Seite in (20) die radiale Fermische kinetische Energie und das zweite die Weizsäckersche Korrektion darstellt.

Unsere Aufgabe ist nun $(A'/A)_0$ mit der mittleren radialen Dichte D_l der Elektronen mit der Nebenquantenzahl l und mit $\mathrm{grad}_r\,D_l = \partial D_l/\partial r$ in Zusammenhang zu bringen, dann mit diesem Zusammenhang $(A'/A)_0$ in (20) durch $\mathrm{grad}_r D_l$ und D_l auszudrücken und den so erhaltenen Ausdruck für η_l über alle Zustände der k-ten Kugelschale zu summieren.

3. Der Zusammenhang von $(A'/A)_0$ mit $\mathrm{grad}_r D_l$ und D_l

Zur Herleitung des Zusammenhanges zwischen $(A'/A)_0$ und $\mathrm{grad}_r D_l$ sowie D_l gehen wir von der Definitionsgleichung der mittleren radialen Dichte der Elektronen mit der Nebenquantenzahl l in der in Betracht gezogenen Kugelschale aus. Mit Rücksicht darauf, dass die Zustände $\lambda = 1, 2, \ldots, n$ alle $\mu = 2(2l+1)$ — fach besetzt sind, hat man

$$D_l = \mu \sum_{\lambda=1}^{n} \frac{f_\lambda^2}{\int_{r_k}^{r_{k+s}} f_\lambda^2 \, dr} \, . \tag{21}$$

Lange Zeit (bis 1953) war irrtümlicherweise die Annahme verbreitet, dass dies folgendermassen geschehen kann.[6] Man hat zur Definition eines mittleren Dichtegradienten $\mathrm{grad}_r D_l$ die Definitionsgleichung (21) der mittleren radialen Dichte D_l zugrunde gelegt und gefordert, dass die Summe der über das in Betracht gezogene Intervall von r ($r_k \leqq r \leqq r_{k+s}$) hinweggemittelten Dichtegradienten der einzelnen Elektronen mit dem Gradienten der mittleren Elektronendichte D_l, d. h. mit $\mathrm{grad}_r D_l$ übereinstimme, also dass

$$\frac{\mu}{s} \sum_{\lambda=1}^{n} \frac{\int_{r_k}^{r_k+s} \mathrm{grad}_r (f_\lambda^2) \, dr}{\int_{r_k}^{r_k+s} f_\lambda^2 \, dr} = \frac{\mu\pi}{s^2} \sum_{\lambda=1}^{n} \frac{\int_0^\pi \mathrm{grad}_x (f_\lambda^2) \, dx}{\int_0^\pi f_\lambda^2 \, dx} = \mathrm{grad}_r \, D_l \tag{22}$$

sei. Hier hat man auf der linken Seite für f_λ die Eigenfunktionen (5) eingesetzt und A sowie A' wieder fälschlicherweise als konstant betrachtet, also dieselbe unrichtige Näherung zugrunde gelegt, die wir schon auf S.365 erwähnten. Mit Rücksicht darauf, dass μn in der Kugelschale die Anzahl ν_l der Elektronen mit der Nebenquantenzahl l darstellt, dass also $\mu n/s = \nu_l/s = D_l$ ist, ergibt sich so

$$\left(\frac{A'}{A} \right)_0 = \frac{1}{2} \frac{\mathrm{grad}_r D_l}{D_l} \, . \tag{23}$$

Dieser Zusammenhang, der jedoch unrichtig ist, würde in (19) oder (20) gerade das Weizsäckersche Korrektionsglied liefern.

Dass der Zusammenhang (23) unrichtig ist, hat zwei Ursachen. Erstens ist die Definition des mittleren radialen Dichtegradienten von D_l durch (22) grundsätzlich unrichtig und zweitens kann man nach Einsetzen der Eigenfunktionen (5) in (22) A und A' nicht durch eine mittlere Konstante ersetzen, sondern man hat für A die Entwicklung (8) zugrunde zu legen, womit sich

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[6] H. Hellmann, Acta Physicochimica URSS **4**, 225, 1936; P. Gombás, Die statistische Theorie d. Atoms u. ihre Anwendungen, S. 113, Springer, Wien, 1949.

statt (23) eine ganz andere Beziehung ergeben würde: es würde dann auf der linken Seite bei der Mittelung über s nicht nur das von erster Ordnung kleine Glied $(A'/A)_0$, sondern auch das von zweiter Ordnung kleine Glied $(A'/A)_0^2$ verschwinden.

Das Verschwinden dieser Glieder ist eine natürliche Folge dessen, dass die Beziehung (22) zur Definition eines Mittelwertes von $\mathrm{grad}_r D_l$ grundsätzlich unbrauchbar ist. Diese entsteht nämlich aus (21) in der Weise, dass man auf beiden Seiten den Gradienten bildet und in (22) die linke Seite — und zwar nur die linke — über das in Betracht gezogene Intervall von r hinwegmittelt. Hierdurch begeht man einen Fehler, denn auf der linken Seite heben sich durch die Mittelbildung die von dem periodischen Anteil (d. h. vom Faktor $\sin^2 \lambda x$) der Teildichten f_λ^2 herrührenden Gradienten gerade weg. Die periodische Funktion $\sin^2 \lambda x$ hat nämlich im Intervall $0 - \pi \lambda$ Maxima, deren ansteigende und abfallende Äste symmetrisch verlaufen, für die sich also bei der Mittelbildung die Gradienten wegheben. Hierdurch erweist sich die Beziehung (22), aus der durch unzulässige Annahmen das falsche Resultat (23) hergeleitet wurde, als grundsätzlich ungeeignet.

Zur Vermeidung dieses Fehlers hat man nicht die Gradienten, sondern — unter Beibehaltung der Definitionsgleichung (21) — die Quadrate der Gradienten in Beziehung zu setzen und die Mittelung über das Gradientenquadrat durchzuführen.

Hierzu gehen wir wieder von der Grundgleichung (21) aus und definieren eine mittlere radiale Eigenfunktion f der in Betracht gezogenen Elektronen folgendermassen

$$\nu_l f^2 = D_l\,, \tag{24}$$

wo $\nu_l = \mu n = 2(2l+1)\,n$ die Anzahl der Elektronen mit der Nebenquantenzahl l in der in Betracht gezogenen k-ten Kugelschale bezeichnet.

Mit Hilfe von $f = (D_l/\nu_l)^{1/2}$ können wir $(A'/A)_0$ durch folgende Gleichung

$$\frac{\nu_l}{s}\int_{r_k}^{r_k+s} (\mathrm{grad}_r f)^2\, dr = \frac{1}{4}\,\frac{(\mathrm{grad}_r D_l)^2}{D_l}\,, \tag{25}$$

$$\int_{r_k}^{r_k+s} (\mathrm{grad}_r f)^2\, dr = \frac{1}{4}\left(\frac{\mathrm{grad}_r D_l}{D_l}\right)^2 \tag{26}$$

bestimmen, wo für ν_l/s wieder die mittlere radiale Elektronendichte gesetzt wurde. Wir bilden also jetzt den Mittelwert eines Gradientenquadraten und nicht wie vorher den eines Gradienten.

Zur Berechnung des auf der linken Seite in (26) stehenden Integrals hat man zunächst f zu berechnen. Für f erhält man mit (21) aus (24)

$$f=\left[\frac{1}{n}\sum_{\lambda=1}^{n}\frac{f_\lambda^2}{\int_{r_k}^{r_k+s}f_\lambda^2\,dr}\right]^{1/2}. \tag{27}$$

Wenn wir der Kürze halber für das Normierungsintegral in den Nennern der Summenglieder die Bezeichnung K^2 einführen, so erhält man mit der Entwicklung (8)

$$\begin{aligned}K^2&=\int_{r_k}^{r_k+s}f_\lambda^2(r)\,dr=\frac{s}{\pi}\int_0^\pi f_\lambda^2(x)\,dx=\\&=\frac{s}{2}A_0^2\left\{1+\pi\left(\frac{A'}{A}\right)_0+\left(\frac{\pi^2}{3}-\frac{1}{2\lambda^2}\right)\left[\left(\frac{A'}{A}\right)_0^2+\left(\frac{A''}{A}\right)_0\right]\right\}.\end{aligned} \tag{28}$$

Da das Glied $1/(2\lambda^2)$ im Verhältnis zu $\pi^2/3$ besonders für grössere λ sehr klein ist, kann man es hier neben $\pi^2/3$ vernachlässigen, wodurch man für das Normierungsintegral von f_λ den von λ unabhängigen Ausdruck

$$K^2=\frac{s}{2}A_0^2\left\{1+\pi\left(\frac{A'}{A}\right)_0+\frac{\pi^2}{3}\left[\left(\frac{A'}{A}\right)_0^2+\left(\frac{A''}{A}\right)_0\right]\right\} \tag{29}$$

erhält.

Diesen kann man in (27) vor die Summe setzen, wodurch sich die Summation leicht durchführen lässt. Mit Rücksicht darauf, dass wir A von λ als unabhängig betrachten, ergibt sich

$$\begin{aligned}f&=\left(\frac{1}{nK^2}\sum_{\lambda=1}^{n}f_\lambda^2\right)^{1/2}=\frac{A}{K}\left(\frac{1}{n}\sum_{\lambda=1}^{n}\sin^2\lambda x\right)^{1/2}=\\&=\frac{A}{K}\left[\frac{2n+1}{4n}-\frac{1}{4n}\,\frac{\sin(2n+1)x}{\sin x}\right]^{1/2}=\frac{A}{K}g(x),\end{aligned} \tag{30}$$

wobei zu beachten ist, dass A noch eine schwache Abhängigkeit von r aufweist.

Wir befassen uns zunächst mit der Funktion

$$g(x)=\left[\frac{2n+1}{4n}-\frac{1}{4n}\,\frac{\sin(2n+1)x}{\sin x}\right]^{1/2}. \tag{31}$$

Man sieht sofort, dass $g(x)$ bei $x=0$ und $x=\pi$ gleich 0 ist, d. h. dass f und somit auch D_l an den beiden Berandungsflächen der Kugelschale verschwinden. Der Verlauf von $g(x)$ für den Fall $n=10$ ist in Fig. 1 dargestellt. Aus dieser ist zu sehen, dass $g(x)$ aus einem Wellenzug in der Höhe $g_0=\left(\frac{2n+1}{4n}\right)^{1/2}$ und einem steil ansteigenden Ast am Anfang des Wellenzuges bei $x=0$, sowie einem zu diesem symmetrischen steil abfallenden Ast am Ende des Wellenzuges bei $x = \pi$ besteht. Bei der Berechnung von $\operatorname{grad} f$ ziehen wir den Wellenzug und die beiden Äste gesondert in Betracht. Hierzu wollen wir zunächst die Ausdrücke feststellen, durch die wir den Wellenzug und die beiden Äste von g möglichst einfach darstellen können.

Zunächst ist unmittelbar zu sehen, dass man g für grosse n in folgender Form schreiben kann

$$g = \left(\frac{1}{2} - \frac{1}{4n} \frac{\sin 2nx}{\sin x} \right)^{1/2} . \tag{32}$$

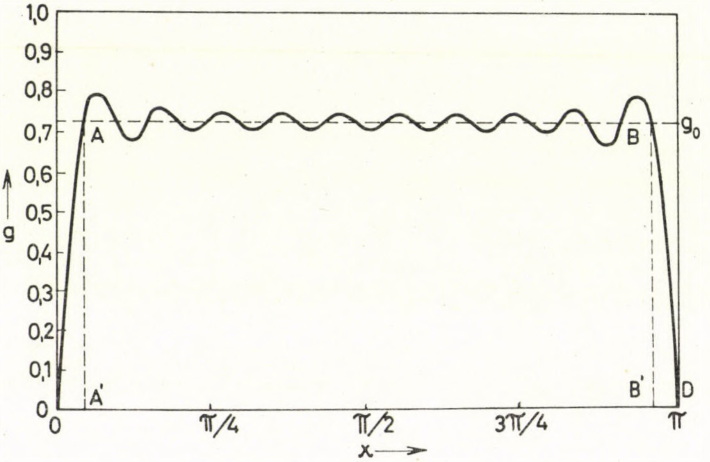

Fig. 1. Verlauf der Funktion $g(x)$ für $n = 10$

Für den Wellenzug können wir im Falle grosser n im zweiten Glied in der Klammer im Nenner für die im Verhältnis zum Zähler langsam veränderliche Funktion $\sin x$, im Intervall $0 \leq x \leq \pi$, deren Mittelwert $1/2$ setzen. Da weiterhin für grosse n das zweite Glied in der Klammer im Verhältnis zum ersten klein ist, kann man die rechte Seite in (32) in eine Reihe entwickeln, die man nach dem zweiten Glied abbricht. Man kann also für grosse n den Wellenzug durch folgenden Ausdruck approximieren

$$g_w = \frac{1}{2^{1/2}} \left(1 - \frac{1}{2n} \sin 2nx \right) . \tag{33}$$

Der ansteigende Ast g_a von g lässt sich durch die durch den Koordinatenursprung und den Punkt A hindurchgehende Gerade und zwar durch das Geradestück OA approximieren. Die Koordinaten des Punktes A sind $x_A = \pi/2\, n$ und $g_A \cong 1/2^{1/2}$. Der Anstieg dieser Gerade beträgt also

$$\frac{g_A}{x_A} = \frac{1}{2^{1/2}}\,\frac{2\,n}{\pi} = \frac{2^{1/2}}{\pi}\,n\,. \tag{34}$$

Wir können also den ansteigenden steilen Ast näherungsweise durch folgenden einfachen Ausdruck darstellen

$$g_s = \frac{2^{1/2}}{\pi}\,nx. \tag{35}$$

Für den abfallenden Ast g_a zwischen dem Punkten B und D gilt bei Zugrundelegung derselben Näherung der Näherungsausdruck

$$g_a = \frac{2^{1/2}}{\pi}\,n(x-\pi)\,. \tag{36}$$

Mit diesen Näherungsfunktionen können wir nun für die linke Seite von (26) einen Näherungswert berechnen. Wenn wir statt r wieder die Variable x einführen, so hat man für die linke Seite von (26)

$$L = \int_{r_k}^{r_k+s} (\mathrm{grad}_r f)^2\,dr = \frac{\pi}{s}\int_0^\pi (\mathrm{grad}_x f)^2\,dx = \frac{\pi}{s}\,\frac{1}{K^2}\int_0^\pi (A'g + Ag')^2\,dx\,, \tag{37}$$

wo g' die Ableitung von g nach x bezeichnet und K die durch (29) definierte Normierungskonstante ist.

Wir wollen zunächst den Anteil L_2 zum Integral L berechnen, der vom Wellenzug g_w resultiert, der für das Intervall $A'B'$ gültig ist. Wenn man in Betracht zieht, dass man für grosse n das Intervall $A'B'$ durch das gesamte Intervall OD ersetzen kann, so folgt mit der Näherungsfunktion (33)

$$\begin{aligned} L_2 = \frac{\pi}{s}\,\frac{1}{K^2}\,\frac{1}{2}\int_0^\pi \Bigg[(A')^2 \left(1 - \frac{1}{2n}\sin 2\,nx\right)^2 - \\ - 2\,AA'\left(1 - \frac{1}{2\,n}\sin 2\,nx\right)\cos 2\,nx + A^2\cos^2 2\,nx\Bigg]\,dx. \end{aligned} \tag{38}$$

Wenn wir für A und K die Ausdrücke (8) bzw. (29) heranziehen und alle Glieder, die von der Ordnung $1/n$ oder von höherer Ordnung klein sind, vernachlässigen, so erhalten wir

$$L_2 = \frac{\pi^2}{s^2}\left[\left(\frac{A'}{A}\right)_0^2 + \frac{1}{2}\right]. \tag{39}$$

Für den vom steil ansteigenden Ast g_s resultierenden Anteil L_1 des Integrals L ergibt sich mit denselben Annahmen und in derselben Näherung wie bei L_2

$$L_1 = \frac{\pi}{s}\frac{1}{K^2}\frac{2\,n^2}{\pi^2}\int_0^{\pi/2n}[(A')^2 x^2 + 2\,AA'\,x + A^2]\,dx = \\ = \frac{\pi^2}{s^2}\left[\frac{1}{\pi}\left(\frac{A'}{A}\right)_0 + \frac{2\,n}{\pi^2}\right]. \tag{40}$$

Auf dieselbe Weise erhält man für den vom steil abfallenden Ast g_a resultierenden Anteil L_3 des Integrals L

$$L_3 = \frac{\pi^2}{s^2}\left[-\frac{1}{\pi}\left(\frac{A'}{A}\right)_0 + \frac{2\,n}{\pi^2}\right]. \tag{41}$$

Für das ganze Integral L hat man also

$$L = L_1 + L_2 + L_3 = \frac{\pi^2}{s^2}\left[\left(\frac{A'}{A}\right)_0^2 + \frac{1}{2} + \frac{4n}{\pi^2}\right], \tag{42}$$

womit mit Rücksicht auf die Definitionsgleichung (26) der Zusammenhang

$$\left(\frac{A'}{A}\right)_0^2 = \frac{1}{4}\frac{s^2}{\pi^2}\left(\frac{\mathrm{grad}_r\,D_l}{D_l}\right)^2 - \frac{1}{2} - \frac{4\,n}{\pi^2} \tag{43}$$

folgt.

Da wir diesen Zusammenhang nicht nur für den Fall grosser n-Werte anwenden wollen, berechnen wir wie sich dieser Zusammenhang für den kleinsten n-Wert, $n = 1$, gestaltet. Man erhält dann aus (30)

$$f = \frac{A}{K}\sin x\,, \tag{44}$$

wo wir jetzt wegen $\lambda = 1$ für K statt (29) den genaueren Ausdruck (28) und für A wieder den Ausdruck (8) setzen. Mit diesen ergibt sich aus (26)

$$\left(\frac{A'}{A}\right)_0^2 = \frac{1}{4}\frac{s^2}{\pi^2}\left(\frac{\mathrm{grad}_r\,D_l}{D_l}\right)^2 - 1\,, \tag{45}$$

womit man aus (19) für die radiale kinetische Energie der Elektronen im Zustand $n = 1$

$$\eta_1 = \frac{1}{8}\,e^2 a_0\left(\frac{\mathrm{grad}_r\,D_l}{D_l}\right)^2 \tag{46}$$

erhält. Dies ist der richtige Ausdruck für die radiale kinetische Energie der Elektronen im tiefsten Zustand, da in diesem Zustand, wie schon erwähnt wurde, die gesamte radiale kinetische Energie der Elektronen durch den Weizsäckerschen Anteil dargestellt wird. Dies gibt also zugleich auch einen Beweis dafür, dass die Gleichung (26) die richtige Basis zur Herleitung des Zusammenhanges zwischen A'/A einerseits und grad D_l, sowie D_l andererseits gibt.

Wir können nun den für $n = 1$ exakt gültigen Zusammenhang (45) mit unserem für grosse n-Werte hergeleiteten Zusammenhang (43) vergleichen, wenn wir in diesem versuchsweise $n = 1$ setzen. Es ergibt sich dann das Resultat

$$\left(\frac{A'}{A}\right)_0^2 = \frac{1}{4}\frac{s^2}{\pi^2}\left(\frac{\text{grad}_r D_l}{D_l}\right)^2 - \frac{1}{2} - \frac{4}{\pi^2} = \frac{1}{4}\frac{s^2}{\pi^2}\left(\frac{\text{grad}_r D_l}{D_l}\right)^2 - 0{,}9 \quad (47)$$

das sich überraschenderweise von (45) nur darin unterscheidet, dass auf der rechten Seite $-0{,}9$ statt -1 steht.

4. Zusammensetzung der mittleren radialen kinetischen Energie der Elektronen aus dem Fermischen und Weizsäckerschen Anteil

Mit dem Zusammenhang (43) erhält man für die radiale kinetische Energie η aller Elektronen der in Betracht gezogenen Kugelschale

$$\eta = \mu \sum_{\lambda=1}^{n} \eta_\lambda = \frac{1}{2} e^2 a_0 \frac{\pi^2}{s^2} \mu \sum_{\lambda=1}^{n} \left(\lambda^2 - \frac{1}{2} - \frac{4}{\pi^2} n\right) + \frac{1}{8} e^2 a_0 \left(\frac{\text{grad}_r D_l}{D_l}\right)^2 \nu_l . \quad (48)$$

Hieraus folgt mit Rücksicht auf die Beziehung

$$\sum_{\lambda=1}^{n} \lambda^2 = \frac{1}{6} n(n+1)(2n+1) \quad (49)$$

$$\eta = \frac{1}{2} e^2 a_0 \frac{\pi^2}{s^2} \mu \left[\frac{1}{6} n(n+1)(2n+1) - \right. \\ \left. - \frac{1}{2} n - \frac{4}{\pi^2} n^2\right] + \frac{1}{8} e^2 a_0 \left(\frac{\text{grad}_r D_l}{D_l}\right)^2 \nu_l . \quad (50)$$

Wenn wir hier noch $\mu n = \nu_l$ setzen, so erhält man das Resultat

$$\eta = \frac{1}{2} e^2 a_0 \frac{\pi^2}{s^2} \left[\frac{1}{6}(n+1)(2n+1) - \right. \\ \left. - \frac{1}{2} - \frac{4}{\pi^2} n\right] \nu_l + \frac{1}{8} e^2 a_0 \left(\frac{\text{grad}_r D_l}{D_l}\right)^2 \nu_l . \quad (51)$$

Für $n = 1$ sollte die eckige Klammer im ersten Glied auf der rechten Seite verschwinden, das infolge dessen, dass die Formel (51) bzw. (43) für grosse n hergeleitet wurden, nicht der Fall ist. Der Ausdruck in der eckigen Klammer hat für $n = 1$ den Wert 0,1; die Korrektion gibt also auch noch für $n = 1$ einen sehr gut brauchbaren Wert, denn es hebt in diesem Fall 0,9 des unkorrigierten Gliedes $1/6\,(n+1)\,(2n+1)$ weg. Wir können unser Resultat für $n = 1$ dem tatsächlichen Sachverhalt vollkommen anpassen, wenn wir im letzten Glied der eckigen Klammer statt $4/\pi^2$ die etwas grössere Zahl 1/2 setzen, was für grosse n unwesentlich ist und was dadurch unterstützt werden kann, dass es — wie man aus Fig. 1 sieht — gerechtfertigt ist, für die aus den steil ansteigenden und abfallenden Äste von f resultierenden Anteile der kinetischen Energie die Integration, statt auf die Intervalle OA' und $B'D$, auf etwas grössere Intervalle auszudehnen. Man erhält so

$$\eta = \frac{1}{2} e^2 a_0 \frac{\pi^2}{s^2} \frac{1}{6} (n+1)(2n-2)\, \nu_l + \frac{1}{8} e^2 a_0 \left(\frac{\operatorname{grad}_r D_l}{D_l}\right)^2 \nu_l . \tag{52}$$

Das erste, dem Fermischen Anteil entsprechende Glied des auf der rechten Seite stehenden Ausdruckes ist also durch die Korrektion im Vergleich zum entsprechenden nicht korrigierten im Verhältnis

$$\gamma = \frac{2n-2}{2n+1} \tag{53}$$

verkleinert.

Wenn wir für die mittlere nicht-korrigierte radiale Fermische Energie pro Elektron die Bezeichnung ε_F und für den radialen Weizsäckerschen Anteil pro Elektron die Bezeichnung ε_W einführen, also

$$\varepsilon_F = \frac{1}{2} e^2 a_0 \frac{\pi^2}{s^2} \frac{1}{6} (n+1)(2n+1) \tag{54}$$

und

$$\varepsilon_W = \frac{1}{8} e^2 a_0 \frac{(\operatorname{grad} D_l)^2}{D_l} \tag{55}$$

setzen, so können wir unser Resultat folgendermassen formulieren

$$\varepsilon_R = \gamma \varepsilon_F + \varepsilon_W , \tag{56}$$

wo ε_R die gesamte radiale kinetische Energie pro Elektron bezeichnet.

Wenn man also bei der statistischen Berechnung der radialen kinetischen Energie der Elektronen neben dem Fermischen Anteil auch den Weizsäckerschen in Betracht zieht, kann man die beiden Anteile nicht einfach addieren, sondern man hat hierbei eine Korrektion anzubringen und zwar — wie wir eingangs erwartet haben — nicht am Weizsäckerschen, sondern am Fermischen Anteil.

Der Korrektionsfaktor γ erweist sich besonders für kleine n-Werte als beträchtlich; es ist für

$$n = 1,\ 2,\ 3,\ 4,\ 5,\ \ldots,\ \infty$$
$$\gamma = 0,\ 2/5,\ 4/7,\ 6/9,\ 8/11,\ \ldots,\ 1.$$

Der Korrektionsfaktor wächst also mit wachsendem n von 0 an und erreicht für sehr grosse n-Werte den Wert 1.

Wie man die hier erhaltenen Resultate auf ein statistisch behandeltes Atom übertragen kann, folgt in einer demnächst erscheinenden Arbeit.

О КОРРЕКЦИИ КИНЕТИЧЕСКОЙ ЭНЕРГИИ ВЕЙЦЗЕККЕРА

П. ГОМБАШ

Резюме

Исследуется статистический расчет кинетической энергии электронного газа для случая коррекции неоднородности и показывается, что при простом сложении фермиевой части и части неоднородности Вейцзеккера получается погрешность. Последняя рассчитывается для простого случая. Далее показывается, что некоторые прежние попытки-предпринятые несколькими авторами исправить эту погрешность через уместный фактор, коррекции равный 1/9 привели к неудовлетворительным значениям энергии; в то же время расчетные энергии с данным фактором коррекции лежат ниже на 8% у тяжелых, на 12% у промежуточных и на 23% у легких атомов.

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VELOCITY AND CURRENT DETERMINATION FOR STEADILY TRAVELLING DOMAINS IN SEMICONDUCTORS. II

WATKINS—GUNN EFFECT

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The velocity and current determination method proposed in Part I is applied for the Watkins—Gunn effect.

Two cases are considered: a) the case of quasi-equilibrium between high- and low-mobility states (instantaneous transitions) and b) the case of non-instantaneous transitions. For the former case the total current j_S and the domain velocity u_0 are determined separately also including field-dependent diffusion coefficients. For the latter case, an algebraic equation system is derived for j_S and u_0 in terms of "domain shape parameters" S_i which characterize the domain shape around the neutrality point.

The analysis of the resulting velocity expressions shows that the Gunn domains, in contrast to the slowly moving recombination domains, are insensitive to the domain shape, at least in GaAs.

I. Introduction

It is well known that in semiconductors there are various current instabilities which are connected with the negative differential conductivity of the sample. As was proposed by Ridley and Watkins [1] and Hilsum [2], in GaAs and in other materials with similar conduction band structure the negative differential conductivity is due to the field-dependent electron transfer from the high-mobility central valley to the low-mobility upper valleys. As a result, a high field domain forms which can travel steadily from the cathode to the anode and causes oscillations in the external circuit. It is interesting to note that it was proposed in an early paper by Böer [3], without referring to the electron transfer mechanism, that high field domains would occur if the mobility decreased stronger than linearly with increasing field. Firstly, Gunn has discovered such current instability in GaAs [4a]. Detailed investigations were given in [4b], [5], [6] and later, a similar effect was observed in other materials [7].

The problem of stable domain propagation requires a solution of an essentially non-linear differential equation system. In other words, at the onset of the instability, a linear treatment may be adequate [8]—[11] but the stable domain propagation, when non-linear effects are predominant, needs different treatment. The numerical solution of the non-linear differential equation system [12]—[13], or analytical investigations [14]—[21] give equally important information on the behaviours of high field domains.

In the present paper a method different from the previous ones is given to determine the domain velocity (u_0) and the total current (j_s) for steadily travelling domains, accepting a two-state model. The method is a generalization of that used in paper [22] (hereafter referred to as I) for velocity and current determination of slowly moving recombination domains*. Namely, starting from the wavelike (Riemann-type) solution of the non-linear problem [15], [17], the charge densities in the low- and high-mobility states ϱ_i ($i = 1, 2$) are expressed in terms of electrical field E and its derivatives, or more precisely $\varrho_i = \varrho_i(E, E', E'')$, where the $'$ means differentiation with respect to $z = x - u_0 t$. Then an algebraic equation system is derived for u_0 and j_s in terms of "domain shape parameters" defined by

$$S_i \equiv \frac{\varepsilon}{4\pi} \left(\frac{d^i E}{dz^i} \right)_{z_0} .$$

The parameters S_i characterize the domain shape in the neutrality point z_0.

A numerical analysis of quantities entering into the algebraic equation system shows that inside the domain a quasi-equilibrium between the high- and low-mobility states might be supposed (short intervalley relaxation time). In this case the calculations are simple: the domain velocity and the current can be determined separately and u_0 turns out to be insensitive to the domain shape. In this case the peak field dependence of u_0 can be predicted. Using the theoretical data given by Butcher *et al.* [18], [19b] it is shown that the diffusion and the field dependence of the diffusion coefficients give only small contributions to u_0, which is determined entirely by the *average* drift velocity at the *peak field.*

Neglecting the diffusion terms in the current j_s, an algebraic equation of third degree is derived for u_0, which can be solved exactly. The resulting velocity expressions, which can be considered as a generalization of previously obtained formulae for the two-state model, make it possible to determine the shape dependence of domain velocity.

2. Basic equations

Let ϱ_1 and ϱ_2 denote the charge densities in the high- and low-mobility states, respectively, and let the mobilities $\mu_i(E)$ and the diffusion constants $D_i(E)$ depend on the electrical field strength E. Following papers [9] and [24], besides the continuity and Poisson's equations, we shall use the equations describing the transitions between the states "1" and "2":

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* A preliminary report on the method was published in [23].

$$\frac{\partial \varrho_i}{\partial t} - \frac{\partial j_i}{\partial x} = \left(\frac{\partial \varrho_i}{\partial t}\right)_{\text{gen.}}, \tag{1}$$

$$j_i = \varrho_i \mu_i E + \frac{\partial D_i \varrho_i}{\partial x}, \tag{2}$$

$$\frac{\partial E}{\partial x} = \frac{4\pi}{\varepsilon}(\varrho_0 - \varrho_1 - \varrho_2) \equiv \frac{4\pi}{\varepsilon}\varrho, \tag{3}$$

where ϱ_0 is the compensating positive charge density, assumed uniformly distributed in the whole sample, ϱ is the space charge density, ε is the dielectric constant. For the transitions $1 \leftrightarrows 2$ we can write:

$$\left(\frac{\partial \varrho_i}{\partial t}\right)_{\text{gen.}} = (-1)^i \left[\frac{1}{\tau_r(E)}\varrho_1 - \frac{1}{\tau_g}\varrho_2\right] \equiv (-1)^i \Psi(\varrho_1, \varrho_2, E), \tag{4}$$

where $\tau_r(E)$ is the characteristic time for transition $1 \rightarrow 2$, τ_g is the same for reverse transition $2 \rightarrow 1$ which is assumed to be independent from the electrical field E. This assumption is based on a result of CONWELL and VASSELL [25], where it is shown that τ_g (with the notation of [25] τ_{21}) is independent from the electron energy.

It is convenient to introduce, besides the space charge density ϱ, the current $j = j_1 + j_2$. From the continuity equations follows:

$$\frac{\partial \varrho}{\partial t} + \frac{\partial j}{\partial x} = 0 \tag{5}$$

since it was supposed, that only transitions $1 \rightarrow 2$ exist, and thus

$$\sum_{i=1}^{2} \left(\frac{\partial \varrho_i}{\partial t}\right)_{\text{gen.}} = 0.^*$$

Let us assume that for the stable domain propagation a wave-like solution is valid. In other words, all quantities (ϱ_i, E) are assumed to be dependent on the variable $z = x - u_0 t$. In this case the total current defined by

$$j_s \equiv j + \frac{\varepsilon}{4\pi}\frac{\partial E}{\partial t}$$

remains constant [26], [16]. From the basic equations and also from Eq. (5) the following differential equation system is obtained:

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* In principle it is not difficult to include also the traps, as well as the external generation of the carriers.

$$j' = u_0 \varrho' = \frac{u_0 \varepsilon E''}{4\pi}, \tag{6}$$

$$j = \frac{u_0 \varepsilon E'}{4\pi} + j_s, \tag{7}$$

$$E' = \frac{4\pi}{\varepsilon} (\varrho_0 - \varrho_1 - \varrho_2), \tag{8}$$

$$u_0 \varrho_1' + j_1' = \Psi, \tag{9}$$

$$u_0 \varrho_2' + j_2' = - \psi. \tag{10}$$

The full description of the stable domain propagation requires the solution of the above equation system, taking into account the proper boundary conditions; namely the condition of electrical neutrality of the whole sample and Kirchoff's law [15]:

$$\begin{aligned} \int_0^L \varrho \, dx &= 0, \\ \int_0^L E \, dx &= \mathcal{E} - \mathcal{J} R, \end{aligned} \tag{11}$$

where L is the length of the sample, $\mathcal{J}$ is the current in the circuit of resistivity R, and $\mathcal{E}$ is the e.m.f. of the battery. Instead of finding an explicit solution for $E(z)$, the field distribution is assumed to be known, say, around the neutrality point z_0 (where $E'(z_0) = 0$) and then an algebraic equation system for u_0 and j_s can be derived in terms of parameters S_i. For this purpose, it is necessary to express ϱ_i in terms of E and its derivatives. Two cases will be considered: a) the case of instantaneous transitions between two states (the "quasi-equilibrium" case) and b) the case when the intervalley relaxation times are commensurable with the other characteristic times of the problem ("non-instantaneous" transitions).

3. Determination of the function $\varrho_i(E, E', \ldots)$ and the differential equation for the field $E(z)$

The functions $\varrho_i(z)$, $E(z)$ and their derivatives are connected with the differential equation system derived in the previous Section. In principle, any of these can be used as a "basic function" and then the two others can be expressed with the aid of this basic function and its derivatives. As only the measurement of $E(z)$ is relatively simple, the space charge density ϱ and its components ϱ_1, ϱ_2 will be expressed in terms of E and its derivatives. In what follows, the previously mentioned cases a) and b) will be treated separately.

a) *Quasi-equilibrium between the valleys*

Knowing the orders of magnitude of the time constants τ_r and τ_g (see e.g. [25] or Section 5 of the present paper) it seems to be quite reasonable to assume that inside the domain a quasi-equilibrium exists between the high- and low-mobility states. In other words, we shall assume that the transitions between the valleys are instantaneous. This assumption is, on the other hand, very characteristic for previous papers dealing with Gunn domain dynamics. This is especially true for the analytical investigations [14]—[21]. The conditions for this assumption will be given in Section 5, using some characteristic quantities containing the domain shape parameters also. Now, according to the assumption of instantaneous transitions, we can write Eq. (4) in the form:

$$\frac{\varrho_2}{\varrho_1} = \frac{\tau_g}{\tau_r(E)} \equiv r(E)\,, \tag{12}$$

where the function $r(E)$ can be determined from the experimental data [27], [28] or from a microscopic theory [18], [25]. Such relation between the high- and low-mobility densities was assumed first by Kroemer [29], [30]. From the Poisson's equation and Eq. (12), ϱ_1 and ϱ_2 might be calculated *separately*. In fact, the simple calculation gives:

$$\varrho_1 = \frac{T}{1 + r(E)}\;; \qquad \varrho_2 = \frac{r(E)\,T}{1 + r(E)}\,, \tag{13}$$

where $T \equiv \varrho_0 - \varepsilon E'/4\pi$ is the total negative charge density. Comparing Eqs. (12) and (13) we see that while the ratio ϱ_2/ϱ_1 depends only on the field E, the components ϱ_1/ϱ_2 both depend on E and E'. Finally, with the aid of Eq. (13), it is easy to write the differential equation for $E(z)$. Substituting (13) in (7):

$$F(E, E', E'') \equiv j_s + \frac{u_0\,\varepsilon E'}{4\pi} - \\ - \sum_{i=1}^{2} \left[G_i\,\mu_i\,E + \dot{D}_i\,E'\,G_i + D_i \sum_{k=0}^{1} \frac{\partial G_i}{\partial E^{(k)}}\,E^{(k+1)} \right] = 0\,,$$

where $G_i \equiv \varrho_i(E, E')$ and $\dot{D}_i \equiv (dD_i)/(dE)$.

b) *Large intervalley relaxation times*

The determination of functions $G_i \equiv \varrho_i(E, E', \ldots)$ is more complicated now, since Eqs. (9)—(10) should be taken into account, too. For simplicity, we shall assume that μ_i and D_i are independent from the field E. In case a) we did not make such a simplification.

Firstly, the functions ϱ_i' will be expressed by differentiating the Poisson's equation and using Eq. (17). We obtain:

$$\varrho_i' = (-1)^i (B - A - C/D_i)\,; \quad (i = 1, 2)\,, \tag{14}$$

where

$$A(D_1 - D_2) = (j_s + \varepsilon u_0 E'/4\pi)\,; \; B(D_1 - D_2) = (\varrho_1 \mu_1 + \varrho_2 \mu_2) E\,;$$
$$C(D_1 - D_2) = D_1 D_2 \varepsilon E''/4\pi\,.$$

Now, using Eqs. (9), (10) and differentiating the Poisson's equation repeatedly, one can express ϱ_i'' with the aid of E, its derivatives and ϱ_i', then substituting ϱ_i' from Eq. (14) we obtain the following equation system for ϱ_1 and ϱ_2:

$$\varrho_1 L - \varrho_2 M = R\,,$$
$$\varrho_1 + \varrho_2 = T\,,$$

thus

$$\varrho_1 = \frac{R + TM}{L + M}\,; \; \varrho_2 = \frac{-R + TL}{L + M}\,, \tag{15}$$

where

$$R = \frac{D_1 D_2 \varepsilon E'''}{4\pi} + \left(j_s + \frac{\varepsilon E' u_0}{4\pi}\right)\left(u_0 + \frac{\eta E'}{D_1 - D_2}\right) + \left[u_0 (D_1 + D_2) + \frac{E (D_1^2 \mu_2 - D_2^2 \mu_1)}{D_1 - D_2}\right] \cdot \frac{\varepsilon E''}{4\pi}\,;$$

$$L = D_2 \mu_1 E' + u_0 \mu_1 E + (D_1 - D_2)\frac{1}{\tau_r} + \frac{\eta}{D_1 - D_2} \mu_1 E^2, \tag{15'}$$

$$M = - D_1 \mu_2 E' - u_0 \mu_2 E + (D_1 - D_2)\frac{1}{\tau_g} - \frac{\eta}{D_1 - D_2} \mu_2 E^2,$$

$$T = \varrho_0 - \frac{\varepsilon E'}{4\pi}\,; \; \eta = D_1 \mu_2 - D_2 \mu_1\,.$$

Thus, the first part of the calculation is completed; the functions $G_i \equiv \varrho_i(E, E', E'', E''')$ are determined, provided μ_i and D_i to be constant.

For $E(z)$ we have the following nonlinear differential equation, containing u_0 and j_s as parameters:

$$F(E, E', \ldots, E^{\mathrm{IV}}) \equiv j_s + \frac{u_0 \varepsilon E'}{4\pi} - \sum_{i=1}^{2} \left[G_i \mu_i E + D_i \sum_{k=0}^{3} \frac{\partial G_i}{\partial E^{(k)}} E^{(k+1)} \right] = 0 .$$

4. Determination of current and domain velocity for arbitrary domain shape

In the further calculations we shall use the expressions ϱ_1 and ϱ_2 obtained in the previous Section. If the field distribution is known around any point of the domain (say, around the neutrality point z_0), then, based on Eqs. (6), (7), an algebraic equation system could be derived for u_0 and j_s in terms of S_i and other characteristic quantities. In the neutrality point z_0, ϱ_i may have singularities for a possible value of u_0 and so for velocity determination another point should be chosen. It will be shown *a posteriori*, however, that for the *exact* velocity values, the space charge components ϱ_i have no singularities. Similar problems do not arise in case a), where only a single domain velocity exists, and the ϱ_i $(i = 1, 2)$ do not contain the domain velocity explicitly.

a) *Quasi-equilibrium between the valleys*

The calculation is very simple now (including field dependent mobilities and diffusion coefficients as well), because both the current and the velocity can be obtained separately. As we shall see, in the general case, u_0 and j_s must be determined from a nonlinear algebraic equation system. Assuming $S_2 \neq 0$ we obtain:

$$j_s = v_{a0} \varrho_0 - D_{a0} S_2 , \tag{16}$$

$$- u_0 = v_{a0} + \frac{\gamma r_0 (D_{10} - D_{20})}{\tau_{M0} v_{10} (1 + r_0)^2} - \frac{\dot{D}_{a0} E_0}{\tau_{M0} v_{10}} + \frac{D_{a0} S_3}{S_2} , \tag{17}$$

where

$$v_{a0} = \frac{v_{10} + r_0 v_{20}}{1 + r_0}$$

is the average drift velocity,

$$D_{a0} = \frac{D_{10} + r_0 D_{20}}{1 + r_0}$$

is the average diffusion coefficient and

$$\dot{D}_{a0} = \frac{\dot{D}_{10} + r_0 \dot{D}_{20}}{1 + r_0}$$

is the average of derivatives of D_i with respect to the field E. The index "0" refers to the neutrality point. In Eq. (17) the Maxwell relaxation times,

$$\tau_{M0} = \varepsilon/4\pi\,\varrho_0\,\mu_{10} \text{ and } \gamma \equiv \frac{E_0}{r_0}\left(\frac{dr}{dE}\right)_0 = -\frac{E_0}{\tau_{r0}}\left(\frac{d\tau_r}{dE}\right)_0$$

were introduced.

As we shall see, all terms containing the diffusion coefficients are generally negligible compared to the drift term and so we obtain the well known relation [26], [14], [16]:

$$j_s = -\,u_0\,\varrho_0\,. \tag{18}$$

It is interesting to remark that this relation was derived here as an approximation, and the deviation from it can be calculated from Eqs. (16), (17). The most important consequence of the quasi-equilibrium assumption is that the domain velocity is slightly dependent on the domain shape: in Eqs. (16) and (17) $D_{a0}S_2$ and $D_{a0}S_3/S_2$ can be neglected. The numerical analysis of the general case justifies this result and thus, based on Eq. (17), the peak field dependence of u_0 can be calculated.

b) *Large intervalley relaxation times*

To avoid unnecessary complications we shall assume that μ_i and D_i are constants and also the Einstein relations are valid for both valleys in the form $\mu_i/D_i = f_0$ $(i = 1, 2)$. Then $\eta = 0$. We have to determine the following quantities:

$$L_0;\ M_0;\ L_0 + M_0;\ L_0';\ M_0';\ (L_0' + M_0' = 0);\ L_0'' + M_0'';\ R_0;\ R_0';\ R_0''$$

and H_i defined by

$$D_1\,M_0^{(i)} + D_2\,L_0^{(i)} \equiv (D_1 - D_2)\,H_i\,.$$

Eqs. (6) and (7) lead to the following equation system for u_0 and j_s:

$$\begin{aligned} j_s = v_{a0}\,\varrho_0 - D_{a0}\,S_2 + S_2\,\tau_0\left[(v_{10} + v_{20})\,u_0 + v_{10}\,v_{20} - D_2/\tau_{M0}\right] + \\ + S_3\,\tau_0\left[2\,v_{10}\,D_2 + u_0\,(D_1 + D_2)\right] + S_4\,\tau_0\,D_1\,D_2\,, \end{aligned} \tag{19}$$

$$b_3\,u_0^3 + b_2\,u_0^2 + b_1\,u_0 + b_0 = 0\,, \tag{20}$$

where the coefficients are

$$b_3 = f_0\,E_0\,S_3\,;\ b_2 = \frac{S_3}{\tau_0} + f_0\,E_0\,P - \frac{4\pi\,S_2\,f_0}{\varepsilon}\left[j_s + (D_1 + D_2)\,S_2\right];$$

$$b_1 = \frac{P}{\tau_0} + f_0 E_0 K - \frac{4\pi S_2 f_0}{\varepsilon} G - \frac{4\pi S_2 \gamma}{\varepsilon \tau_{r0} E_0} [j_s + (D_1 + D_2) S_2];$$

$$b_0 = \frac{K}{\tau_0} - \frac{4\pi S_2 \gamma}{\varepsilon \tau_{r0} E_0} G,$$

and here

$$G = \varrho_0 H_0 + v_{10} D_2 S_2 + D_1 D_2 S_3; \; P = -\frac{S_2}{\tau_0} + (v_{10} + v_{20}) S_3 + (D_1 + D_2) S_4;$$

$$K = -(S_2 E_0 f_0 + S_3) H_0 + (\varrho_0 E_0 f_0 - 2 S_2) H_1 + \varrho_0 H_2 + \\ + \frac{4\pi S_2^2}{\varepsilon} D_2 \mu_1 + v_{10} v_{20} S_3 + 2 v_{10} D_2 S_4 + D_1 D_2 S_5.$$

As b_1 and b_2 contain the current j_s, the velocity u_0 will obey an algebraic equation of fourth degree. It is convenient to introduce the following dimensionless quantities (if $S_2 \neq 0$):*

$$\begin{aligned} & x = u_0/v_{10}; \; B = D_2/D_1 = \mu_2/\mu_1; \; \alpha_r = \tau_0/\tau_{r0}; \\ & \alpha_g = \tau_0/\tau_g; \; \alpha_M = \tau_0/\tau_{M0}; \; r_0 = \tau_g/\tau_{r0}; \; (1/\tau_0 \equiv 1/\tau_{r0} + 1/\tau_g); \\ & \alpha_0 = D_1 \tau_0 E_0''/E_0; \; \alpha_1 = D_1/v_{10}^2 \tau_0; \; \alpha_2 = v_{10} \tau_0 S_3/S_2; \\ & \alpha_3 = D_1 S_3/v_{10} S_2; \; (\alpha_1 \alpha_2 = \alpha_3); \; \alpha_4 = \tau_0 D_1 S_4/S_2; \\ & \alpha_5 = D_1^2 \tau_0 S_5/v_{10} S_2. \end{aligned} \tag{21}$$

Introducing the above parameters into Eq. (19) we have

$$j_s/\varrho_0 v_{10} = C_0 + C_1 x + C_2 x^2, \tag{22}$$

where

$$C_0 = \alpha_g (1 + r_0 B) \left(1 - \frac{\alpha_0}{\alpha_M}\right) + \frac{\alpha_0 B}{\alpha_1 \alpha_M} (1 + 2\alpha_3 + \alpha_1 \alpha_4 - \alpha_1 \alpha_M);$$

$$C_1 = \frac{\alpha_0 (1 + B)}{\alpha_1 \alpha_M} (1 + \alpha_3); \; C_2 = \frac{\alpha_0}{\alpha_1 \alpha_M}.$$

For the dimensionless velocity x we obtain:

$$b_3' x^3 + b_2' x^2 + b_1' x + b_0' = 0, \tag{23}$$

where the coefficients are

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* To compare the formulae of the present paper with those of paper I one has to remember that in I: $x = u_0/w_0$; $(w_0 = D/v_0 \tau_0)$; $\alpha_0 = E_0'' D^2/E_0 v_0^2$ which differ from the present notations.

$$\begin{aligned} b_3' &= \alpha_2 ; \\ b_2' &= -P' - \alpha_0 (1+B) + \alpha_3 - \alpha_M \frac{j_s}{\varrho_0 v_{10}} ; \\ b_1' &= -\alpha_1 P' - K' - K'' - \gamma \alpha_1 \alpha_r \alpha_0 (1+B) - \gamma \alpha_1 \alpha_r \alpha_M \frac{j_s}{\varrho_0 v_{10}} , \\ b_0' &= -\alpha_1 K' - \gamma \alpha_1 \alpha_r K'' , \end{aligned} \qquad (23')$$

and here

$$\begin{aligned} P' &= 1 - (\alpha_2 + \alpha_4)(1+B) ; \\ K' &= \alpha_g (1 + r_0 B)(1 + \alpha_3) + B\left[\alpha_M (1+\alpha_3) - \gamma \alpha_1 \alpha_r \alpha_M - 3\alpha_0 - \alpha_2 - 2\alpha_4 - \alpha_5\right]; \\ K'' &= \alpha_1 \alpha_g \alpha_M (1 + r_0 B) + \alpha_0 B (1 + \alpha_3) . \end{aligned} \qquad (23'')$$

Thus the current j_s and the possible domain velocities u_0 can be determined from Eqs. (22) and (23) if the parameters containing the S_i are known as well. A numerical solution of this nonlinear algebraic equation system can yield the exact velocity values. To obtain a general insight into the domain dynamics, however, it is more convenient to give some approximative expressions for j_s and u_0 using special assumptions. If the possible domain velocities are well separated, a simple approximation is valid, according to Eq. (20) of I. The other possibility is to neglect the diffusion terms $D_i \varrho_i'$ in the current $j = j_1 + j_2$, and to investigate the resulting equation for u_0 (see I, Eq. (22)). Although it might be expected that diffusion can play an important role in the determination of the charge- and field-distribution, it makes only a small contribution to the current. A simple numerical estimate proves this statement. Neglecting the terms $D_i \varrho_i'$ in Eq. (7) we obtain:

$$j_s / \varrho_0 v_{10} = \bar{C}_0 + \bar{C}_1 x , \qquad (24)$$

where

$$\begin{aligned} \bar{C}_0 &= \alpha_g (1 + r_0 B) + \alpha_0 B (1 + \alpha_3) / \alpha_1 \alpha_M \equiv \frac{K''}{\alpha_1 \alpha_M} , \\ \bar{C}_1 &= \alpha_0 (1+B) / \alpha_1 \alpha_M . \end{aligned} \qquad (24')$$

As can be seen, the neglect of the terms $D_i \varrho_i'$ is not equivalent with the limit $D_i \to 0$, since the term x^2 has disappeared, too, which does not contain the diffusion coefficients at all. This term might be neglected only if the condition $| x | \ll 1$ is valid, i.e. if the domain velocity u_0 is much less then the drift-velocity of the high-mobility component at the peak field ($| u_0 | \ll v_{10}$). This assumption is a good one, since for usual values of μ_1 and E_0, $v_{10} = 4 \cdot 10^9$ cm/sec, while the experimentally observed domain velocity is about 10^7 cm/sec [4b], [5]. Substituting (24) in (23) we obtain an algebraic equation of third degree for u_0:

$$a_3 x^3 + a_2 x^2 + a_1 x + a_0 = 0 , \qquad (25)$$

where the coefficients are

$$\begin{aligned} a_3 &= K_2/\alpha_1 ; & K_2 &= \alpha_3 - \alpha_0(1+B) ; \\ a_2 &= -K_1/\alpha_1 + K_2 ; & K_1 &= \alpha_1 P' + K'' + \gamma\alpha_0\alpha_1\alpha_r(1+B) ; \\ a_1 &= -K_1 - K_0 ; & K_0 &= K' + \gamma\alpha_r K'' ; \\ a_0 &= -\alpha_1 K_0 . \end{aligned}$$

Now, the following remark is necessary. The above approximation led to the neglect of some terms in j_s which still appear in K'. This inconsequence is not essential because the further calculations are independent of the neglect of these small terms in K'. The most important results of this approximation are the neglect of x^2 term in (22) and the connection between $\bar{C}_0$ and K'' given by (24'). One can see immediately that $x = -\alpha_1$ is a root of (25), and thus:

$$u_0^{(\mathrm{I})} = -\frac{D_1}{v_{10}\,\tau_0} . \tag{26}$$

The two other roots can be determined from the following simple equation

$$K_2 x^2 - K_1 x - \alpha_1 K_0 = 0 . \tag{27}$$

If the condition

$$\frac{|4\alpha_1 K_0 K_2|}{K_1^2} \ll 1$$

is valid, the approximate roots are:

$$-x^{(\mathrm{II})} \approx \alpha_1 K_0/K_1 ; \quad x^{(\mathrm{III})} \approx K_1/K_2 . \tag{28}$$

It is important to emphasize that the slowest domain velocity is an exact root of the approximate equation (25) but not that of the original one (23). This fact is very important because it shows, that for the exact u_0 the ϱ_i $(i = 1, 2)$ have no singularities in the neutrality point. Thus if we use the approximate domain velocity (26) we have to make an appropriate approximation also in the numerator of ϱ_i in order to eliminate the singularities originating from the approximation itself.

Both the results of the quasi-equilibrium approximation, and the numerical analysis suggest the identification of the root $u_0^{(\mathrm{II})}$ with the actually measurable domain velocity, and in this case the problem of singularity does not arise at all. From Eq. (28) we obtain (if $K_1 \neq 0$):

$$-u_0^{(\mathrm{II})} \approx \frac{v_{10}\alpha_1 K_0}{K_1} = \frac{v_{10}\alpha_1 (K' + \gamma\alpha_r K'')}{\alpha_1 P' + K'' + \gamma\alpha_0\alpha_1\alpha_r(1+B)} . \tag{29}$$

As will be shown by estimating the quantities in Eq. (29), the **Gunn** domains are generally not sensitive to the domain shape. In other words, one can obtain a shape-independent domain velocity from (29), for many practically important cases. In fact, taking into account the following approximations:

$$K' \approx \alpha_g(1 + r_0 B) + B(\alpha_M - \gamma\alpha_1 \alpha_r \alpha_M) \,;$$
$$P' \approx 1 \,; \;\; K'' \approx \alpha_1 \alpha_g \alpha_M (1 + r_0 B) \,,$$

and making some transformations, using e.g. the relation $\alpha_g = 1/(1 + r_0)$, we have:

$$- u_0^{(\mathrm{II})} \approx v_{10} \frac{\alpha_g(1 + r_0 B) + \gamma\alpha_1 \alpha_g \alpha_M \alpha_r (1 - B) + B\alpha_M}{1 + \alpha_g \alpha_M (1 + r_0 B)} \,. \tag{30}$$

This expression is a generalisation of results given in I, [31] and [32] for the case $B \neq 0$. For $B \equiv 0$ we obtain from Eq. (29) the velocity of recombination domains which was first given in [22]:

$$- u_{0r}^{(\mathrm{II})} \approx \frac{\alpha_g v_0 (1 + \alpha_3 + \gamma\alpha_1 \alpha_r \alpha_M)}{1 - \alpha_2 - \alpha_4 + \gamma\alpha_0 \alpha_r + \alpha_M \alpha_g} \,, \tag{31}$$

where the notation $v_{10} \equiv v_0$ was used. (In Eq. (25) of I the term $\gamma\alpha_0\alpha_r$, being generally small, was neglected.)

5. Comparison with experiments and discussions

Before discussing the results of the calculations it is worthwhile to repeat the most important assumptions. These are: (i) the sample is originally homogeneous; (ii) steadily propagating high field domains exist which can be described by a wave-like (Riemann-type) solution; (iii) the domain formation is due to the field dependent electron transfer from the high-mobility states (subscript "1") to low-mobility states* (subscript "2"); (iv) only the transition 1 → 2 depends on the field; (v) the effect of traps are negligible.

Now, it would be possible, similarly to I, to discuss the propagation of high field domains of various shapes. A short glance at the available experimental and theoretical works on the domain shape shows, however, that both the flat and almost flat domains are very far from the actually observable domain shapes. The experimental results of papers [4a] and [4b] show that

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* It is supposed that μ_i and D_i are local functions of the electrical field E, the conditions of which were examined in a recent paper by **Bonch-Bruevich** and **Gulyaev** [33].

for not too high peak fields ($E_0 < 40$ kVcm^{-1}) a symmetrical (quadratic) domain gives a good approximation. For higher peak fields ($E_0 \approx 60 - 70$ kVcm^{-1}) the domain becomes asymmetrical and for extremely high values of E_0 ($E_0 \approx 100$ kVcm^{-1}) the domains present a triangular shape [5], [6]. These experimental results are consistent with the relevant theoretical works. In fact, in papers [19a, b] both quadratic and asymmetrical domain shapes were found. The numerical integration of the nonlinear differential equation system led to asymmetrical but never to flat or almost flat domains (see Fig. 6. in [19b]).

A rigorous analytical examination given by **Böer** and **Dussel** [21] has also shown that "flat domains" cannot exist at all.* The asymmetry of the domain, i.e. the difference between the leading and trailing edge slopes is connected with the fact, as was pointed out by **Allen** et al. [26], that the maximum charge depletion has a limit ϱ_0, whereas the maximum charge accumulation has no such limit.

The mentioned experimental and theoretical results on the domain asymmetry are very important from the point of view of the velocity determination. In fact, the quasi-equilibrium assumption led to a velocity expression which is practically independent from the domain shape (the term $D_{a_0} S_3/S_2$ is very small in Eq. (17)). On the other hand, the coefficients b_i or a_i contain the domain shape parameters in other combinations too, and it cannot be expected *a priori* that their role is negligible for all practically important cases.

For an order of magnitude estimate, the following parameter values seem to be suitable: $\mu_1 = 8.10^3$ cm^2/Vsec, $D_1 = 240$ cm^2/sec, $B = 0.025$, $\varepsilon = 12.5$, $\varrho_0/e = 10^{14}$ or 10^{15} cm^{-3}. Then the Maxwell's relaxation times are $\tau_{M0} = 8.62 \cdot 10^{-12}$ sec and $8.62 \cdot 10^{13}$ sec, resp. In order to determine the characteristic times τ_r, τ_g we shall assume that the function $r(E)$ has the form $r(E) = (E/E_c)^K$, where E_c and K are parameters. This analytical form of $r(E)$ was proposed by **Kroemer** [29] and recent experimental results confirm this assumption. In fact, the drift velocity-field characteristics of GaAs measured by **Ruch** and **Kino** [28] up to 11 kV/cm, can be well approximated with $E_c = 4.3$ kV/cm, $K = 4$.

Strictly speaking, such an explicit form of $r(E)$ is valid only for fields $E_0 \lesssim 11$ kV/cm $\approx 2\, E_c$. However, we shall use this function up to field $E \approx 10\, E_c - 20\, E_c$. According to our assumption τ_g is independent from the field and thus if $\tau_g = 5.10^{-10}$ sec, and $E_0 = 10\, E_c$, then $r_0 = 10^4$; $\tau_0 \approx \tau_{r0} = 5 \cdot 10^{-14}$ sec, $v_{10} = 3.44 \cdot 10^9$ cm/sec.

Now, we have to make some assumptions on the domain shape to determine the ratios S_{i+2}/S_2 ($i = 1, 2, 3$) and S_2/S_0, where $S_0 \equiv \varepsilon/4\, \pi\, E_0$, which

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* In [26], it was assumed *a priori* that the domain shape is flat.

appear in $\alpha_2, \alpha_3, \alpha_4, \alpha_5$ and α_0. Since for low peak field values ($E_0 < 40$ kV/cm) the domain shape is almost *quadratic* (see Fig. 3. of [4a], [4b]), the only shape-dependent parameter which differs from zero is α_0. In this case $E = E_0(1 - (z/d)^2)$ and thus $\alpha_0 = -2 D_1 \tau_0/d^2$. One can see immediately that for not too small domains the velocity expression (30) is applicable (if $d = 35\,\mu$; $\alpha_0 = -2 \cdot 10^{-6}$). Increasing the peak field E_0, the domain becomes gradually asymmetrical and we have to estimate all shape-dependent dimensionless parameters. It is convenient sometimes to use a concrete domain shape to determine the quantities in question. For an order of magnitude estimate, however, it is sufficient to assume that $| S_{i+2}/S_2 | \approx 1/d^i$; $| S_2/S_0 | \approx \approx 1/d^2$ (d is the "half width" of the domain).

Now, it is easy to see that for practically important cases (if d is not too small) $\alpha_0, \alpha_3, \alpha_4, \alpha_5$ can be neglected. E.g. α_3 appears in b_i or C_i only in the combination $1 + \alpha_3$ and thus its effect is negligible being $| \alpha_3 | \ll 1(| \alpha_3 | = = 2 \cdot 10^{-5})$. The numerical estimate shows that the only parameter which might influence the domain velocity is the parameter $\alpha_2 = \tau_0\, v_{10} S_3/S_2$. It might be called "asymmetry-parameter" as it differs from zero only for asymmetrical domains.

For the previously used data $| \alpha_2 | = 0.05$. At first sight, it might appear that the above values of α_2 can change the domain velocity by 5%. A more careful examination shows that the effect is smaller because both the numerator and denominator of Eq. (29) contain α_2 with the same signs (α_2 appears both in K' and P'). Thus, in the case of WATKINS—GUNN effect, in contrast to the recombination instability, the domain shape plays generally a negligible role in the velocity determination.

Based on these results we can expect that the shape independent velocity expressions (17), (30) give good approximations.

From Eqs. (17) and (30) we can conclude that the domain velocity is equal in a good approximation to the average drift-velocity at the peak field. This means that if the average drift velocity does not change with increasing electrical field and E_0 falls on this part of the $v - E$ curve, the domain velocity also remains constant. This is in agreement with experimental observations [4b], [5].

BUTCHER et al. have pointed out [19b] that the domain velocity becomes greater than the electron drift velocity *outside the domain* if the diffusion coefficients are decreasing with increasing electrical field. Based on Eq. (17) we obtain a similar result: the domain velocity is greater than the average drift velocity *at the peak field* if the diffusion coefficients depend on the field E. The difference is given by the term $\dot{D}_{a0} E_0/\tau_{M0} v_{10}$. The derivative $\dot{D}_{a0}$ can be calculated from the theoretical curve given by BUTCHER and FAWCETT [19b], [18] and the result is that we have significant correction only for samples of small resistivity (this corresponds to the small relaxation time τ_{M0}).

The domain velocity was as far considered to be equal to the *average* drift velocity at the peak field. On the basis of (17), however, we can conclude that if the conditions $r_0 \ll 1$ and also $\mu_{20} r_0 \ll \mu_{10}$ are fulfilled (if $r_0 \ll 1$, all electrons are practically in low-mobility states), the domain velocity is equal to the drift velocity of the *low-mobility* component at the peak field. This conclusion may be very useful if we are interested in the high-field properties of electrons. In fact, measuring the peak field dependence of domain velocity one can get information on the mobility and the diffusion coefficient of electrons in the upper valleys. If E_0 falls on the decreasing part of the $v(E)$ curve, instead of the saturated one, then we can obtain information on the transfer mechanism since from the $u_0(E_0)$ curve the function $r(E_0) = r_0$ can be also determined in principle.

Finally, it is possible to draw an interesting conclusion on the analytical properties of ϱ_i, $(i = 1, 2)$, namely, for the exact velocity values, i.e. for the solutions of Eqs. (22), (23) these functions have no singularities. This point requires, however, further investigations.

Finally, we may summarize the main results of the paper as follows: (i) both for a) instantaneous and b) non-instantaneous transitions the total current j_s and domain velocity u_0 are given in terms of domain shape parameters S_i and other characteristic quantities of the problem; (ii) in contrast to the recombination instability the GUNN domains propagate practically independently from the domain shape; (iii) the diffusion and the field dependence of diffusion coefficients make only a small contribution to u_0, which is determined entirely by the average drift mobility at the peak field; (iv) for sufficiently high values of E_0, the domain velocity equals the drift velocity of the low-mobility component at the peak field; (v) the deviation from the relation $j_s = -u_0 \varrho_0$ is given explicitly, (vi) from the peak field dependence of u_0, one can obtain valuable information on the high field properties of conduction electrons and the transfer mechanism between the valleys using velocity expression (17).

The present calculations show that at least for GaAs, the GUNN domain propagation is insensitive to domain shape. In other materials this may not be the case. Thus the value of the asymmetry parameter α_2 should be estimated for each case. On the other hand if the space charge density has immobile components as well, e.g. electrons in traps, the domain velocity might again depend on the domain shape similarly to the recombination instability.

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REFERENCES

1. B. K. Ridley and T. B. Watkins, Proc. Phys. Soc. (London), **78**, 293, 1961.
2. C. Hilsum, Proc. IRE, **50**, 185, 1962.
3. K. W. Böer, Monatsber. DAW., **1**, 325, 1959.

4a. J. B. Gunn, IBM J. Res. and Develop., **8**, 141, 1964 and Proc. Intern. Conf. Phys. Semicond. Paris, 1964, p. 199.

4b. J. B. Gunn, Proc. Int. Conf. Semiconductor Physics, Kyoto, 1966, p. 505.

5. J. S. Heeks, Trans. IEEE, **ED—13**, 68, 1966.
6. P. Guetin, Trans. IEEE, **ED—14**, 552, 1967 and I. Kuru, P. N. Robson and G. S. Kino, Trans. IEEE, **ED—15**, 21, 1968.
7. J. W. Allen, M. Shyam and G. L. Pearson, Appl. Phys. Letters, **9**, 39, 1966 and A. G. Foyt and A. L. McWhorter, Trans. IEEE, **ED—13**, 79, 1966.
8. V. L. Bonch-Bruevich and Sh. M. Kogan, Soviet Phys. Solid State, **7**, 15, 1965.
9. B. K. Ridley, Trans IEEE, **ED—13**, 41, 1966.
10. R. W. Engelmann and C. F. Quate, Trans. IEEE, **ED—13**, 44, 1966.
11. B. W. Hakki, J. Appl. Phys., **38**, 808, 1967.
12. D. E. McCumber and A. G. Cynoweth, Trans IEEE, **ED—13**, 4, 1966 and Proc. Int. Conf. Semiconductor Physics, Kyoto, 1966. p. 514 and also p. 522.
13. J. A. Copeland, Trans. IEEE, **ED—13**, 185, 1966 and J. Appl. Phys., **37**, 3602, 1966.
14. B. W. Knight and G. A. Peterson, Phys. Rev., **147**, 617, 1966 and Phys. Rev., **155**, 393, 1967.
15. V. L. Bonch-Bruevich, Phys. Letters, **20**, 249, 1966 and Fiz. Tverd. Tela, **8**, 1753, 1966.
16. V. L. Bonch-Bruevich, phys. stat. sol., **22**, 267, 1967.
17. P. N. Butcher, Phys. Letters, **19**, 546, 1965.
18. P. N. Butcher and W. Fawcett, Phys. Letters, **21**, 489, 1966 and Brit. J. Appl., **17**, 1425, 1966.

19a. P. N. Butcher, W. Fawcett and C. Hilsum, Brit. J. Appl. Phys., **17**, 841, 1966.

19b. P. N. Butcher, W. Fawcett and N. R. Ogg, Brit. J. Appl. Phys., **18**, 755, 1967.

20. A. F. Volkov, Fiz. Tverd. Tela, **8**, 3187, 1966 and Phys. Lett. **20**, 598, 1966.
21. K. W. Böer and G. A. Dussel, Phys. Rev., **154**, 292, 1967.
22. G. Pataki, Acta Phys. Hung. **24**, 119, 1968.
23. G. Pataki, Phys. Letters, **26A**, 24, 1967.
24. G. Pataki, Fiz. i Tekn. Polupr., **2**, 206, 1968.
25. E. M. Conwell and M. O. Vassell, Trans. IEEE, **ED—13**, 22, 1966 and Appl. Phys. Letters, **9**, 411, 1966.
26. J. V. Allen, W. Shockley and G. L. Pearson, J. Appl. Phys., **37**, 3191, 1966.
27. D. M. Chang and J. L. Moll, Appl. Phys. Letters, **9**, 283, 1966.
28. J. G. Ruch and G. S. Kino, Appl. Phys. Letters, **10**, 40, 1967.
29. H. Kroemer, Trans. IEEE, **ED—13**, 27, 1966.
30. H. Kroemer, Trans. IEEE, **ED—14**, 476, 1967.
31. B. K. Ridley, Proc. Phys. Soc. (London) **86**, 637, 1965.
32. S. G. Kalashnikov and V. L. Bonch-Bruevich, phys. stat. sol., **16**, 197, 1966.
33. V. L. Bonch-Bruevich and Yu. V. Gulyaev, Preprint of IRE, No. T18111, Moscow, 1967.

ОПРЕДЕЛЕНИЕ СКОРОСТИ И ТОКА ДЛЯ СТАЦИОНАРНО ДВИЖУЩИХСЯ ДОМЕНОВ В ПОЛУПРОВОДНИКАХ. II

ЭФФЕКТ УАТКИНСА-ГАННА

Г. ПАТАКИ

Резюме

Метод, предложенный в части I. для определения скорости домена и полного тока применён к случаю эффекта Уаткинса-Ганна.

Рассматриваются два случая: а) случай квази-равновесия между состояниями с различной подвижностью («моментальные» переходы) и б) случай медленных переходов. В первом случае скорость домена (u_0) и полный ток (j_s) определяются независимо друг от друга включая случай зависящихся от поля диффузионных коэффициентов. Во втором случае получена система алгебраических уравнений для u_0 и j_s с помощью «параметров формы домена» S_i, которые характеризуют форму домена около точки электрической нейтральности. Анализ полученных выражений для скорости показывает, что ганновские домены, в отличии от медленно движущихся рекомбинационных доменов, нечувствительны к форме домена, в крайней мере в GaAs.

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SOME PROPERTIES OF CYLINDRICAL ELECTROVAC FIELDS

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It is shown that the static cylindrical electric and magnetic fields are in a certain sense linearly connected. Furthermore, the known properties of the stationary case are summarized: an extension of Weyl's theorem is proved and the field equations obtained by this theorem are presented in a special coordinate system.

I. Introduction

In this paper we shall present a "classical" treatment of cylindrical stationary electrovac fields (That is, fields consisting of empty space-time and electromagnetism [1]). This means that we shall not use such modern tools of relativists as the spinor calculus or others. For the static case in Section II we shall show that the electrical and magnetic fields obey a strong restriction, especially that they are in a certain sense linearly connected.

In Sections III and IV we summarize the results scattered in the literature about the stationary field: we present the extension of a nice theorem of Weyl and others. This theorem holds originally for static fields and will be extended to stationary fields having the properties described in Section III. The theorem states in both cases that the number of nonvanishing independent metrical field variables can be reduced by one. Finally, we present the field equations obtained from the more general metric by use of the proved theorem.

II. On the static field

A particular exact solution of the Einstein—Maxwell equations*

$$R^{\mu}_{\nu} = \varkappa T^{\mu}_{\nu}\,; \tag{1}$$

$$T^{\mu}_{\nu} = (1/4\,\pi)\left[F^{\mu\alpha}\, F_{\nu\alpha} - (1/4)\,\delta^{\mu}_{\nu}\, F^{\alpha\beta}\, F_{\alpha\beta}\right]; \tag{2}$$

$$(g^{1/2}\, F^{\mu\alpha})_{\alpha} = 0\,; \tag{3}$$

$$F_{\mu\nu} = A_{\nu,\mu} - A_{\mu,\nu} \tag{4}$$

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* Greek indices run from 1 to 4; $x^1 = r$, $x^2 = z$, $x^3 = \varphi$, $x^4 = t$. In this work it will always be assumed that the field variables do not depend on φ and t.

was found by G. Tauber [2] in the static cylindrical case. This solution allows both nonvanishing electric and magnetic fields. The electromagnetic potential vector has the form $A_\mu = (0, 0, \Phi, \Psi)$, where Φ generates the magnetic and Ψ the electrical strain components. The line element is taken

$$ds^2 = - e^{\lambda} (dr^2 + dz^2) - e^{-\varrho} r^2 d\varphi^2 + e^{\varrho} dt^2 . \tag{5}$$

The equations (3) permit us the following substitution [2]:*

$$\Phi_1 e^{\varrho}/r = \Phi_2 ; \qquad \Phi_2 e^{\varrho}/r = - \Phi_1 . \tag{6}$$

Then we get from (3):

$$\left.\begin{aligned} \Delta\Phi &= \varrho_1 \Phi_1 + \varrho_2 \Phi_2 ; \\ \Delta\Psi &= \varrho_1 \Psi_1 + \varrho_2 \Psi_2 . \end{aligned}\right\} \tag{7}$$

Here Δ is the Laplace operator in a cylindrical coordinate system: $\Delta f \equiv f_{11} + f_{22} + (1/r) f_1$. In the following we shall use the equation arising from $R_4^3 = \varkappa T_4^3$:

$$\Phi_1 \Psi_2 = \Phi_2 \Psi_1 . \tag{8}$$

In the particular case found by Tauber, the new electromagnetic potential components are linearly dependent: $\Phi = a \Psi + b$, where a and b are real constants.

Now we shall prove the following.

Theorem: In a continuous field domain there exist only such static cylindrical electromagnetic fields that

$$\Phi = a\Psi + b \tag{9}$$,

(a, b are constants) be fulfilled. The $\Phi = 0$ case corresponds to $a = b = 0$ the $\Psi = 0$ case to the limit $a \to \infty$.

It is clear that this theorem imposes a strong restriction on the shape of static cylindrical electromagnetic fields.

Proof. First we solve (7) for ϱ_1, ϱ_2 as a linear algebraic equation system:

$$\left.\begin{aligned} \varrho_1 &= (\Psi_2 \Delta\Phi - \Phi_2 \Delta\Psi)/(\Phi_1 \Psi_2 - \Phi_2 \Psi_1) ; \\ \varrho_2 &= (\Phi_1 \Delta\Psi - \Psi_1 \Delta\Phi)/(\Phi_1 \Psi_2 - \Phi_2 \Psi_1) . \end{aligned}\right\} \tag{10}$$

Glancing at eq. (8) we see that the denominators vanish. So the continuity of ϱ requires:

$$\Psi_2 \Delta\Phi = \Phi_2 \Delta\Psi ; \qquad \Phi_1 \Delta\Psi = \Psi_1 \Delta\Phi , \tag{11}$$

---

* From now on in this Section the lower indices denote partial derivatives with respect to $x^1 = r$ and $x^2 = z$.

from which we get by use of (8):

$$\Psi_1 (\Phi_{11} + \Phi_{22}) = \Phi_1 (\Psi_{11} + \Psi_{22}) ; \tag{12a}$$

$$\Psi_2 (\Phi_{11} + \Phi_{22}) = \Phi_2 (\Psi_{11} + \Psi_{22}) . \tag{12b}$$

Partial differentiation of (8) with respect to r and z and elimination of the mixed derivatives yields the equations

$$\Psi_1 \left(\Phi_{22} - \frac{\Phi_1 \Psi_1}{\Phi_2 \Psi_2} \Phi_{11}\right) = \Phi_1 \left(\Psi_{22} - \frac{\Phi_1 \Psi_1}{\Phi_2 \Psi_2} \Psi_{11}\right) \tag{13a}$$

and

$$\Psi_2 \left(\Phi_{11} - \frac{\Phi_2 \Psi_2}{\Phi_1 \Psi_1} \Phi_{22}\right) = \Phi_2 \left(\Psi_{11} - \frac{\Phi_2 \Psi_2}{\Phi_1 \Psi_1} \Psi_{22}\right) . \tag{13b}$$

We get from (12) and (13) simply by taking the difference of Eqs. (a) and then of Eqs. (b):

$$\Psi_1 \Phi_{11} \left(1 + \frac{\Phi_1 \Psi_1}{\Phi_2 \Psi_2}\right) = \Phi_1 \Psi_{11} \left(1 + \frac{\Phi_1 \Psi_1}{\Phi_2 \Psi_2}\right) ; \tag{14a}$$

$$\Psi_2 \Phi_{22} \left(1 + \frac{\Phi_2 \Psi_2}{\Phi_1 \Psi_1}\right) = \Phi_2 \Psi_{22} \left(1 + \frac{\Phi_2 \Psi_2}{\Phi_1 \Psi_1}\right) . \tag{14b}$$

Now there are two cases:

a) If $\Phi_1 \Psi_1 / \Phi_2 \Psi_2 \neq -1$ then

$$\Psi_i \Phi_{ii} = \Phi_i \Psi_{ii} ; \quad i = 1, 2 \tag{15}$$

must hold. Integrating these equations we have:

$$\ln \Phi_i = \ln a \Psi_i ; \qquad a = \text{const} ., \tag{16}$$

and further integration yields $\Phi = a \Psi + b$, where $b = \text{const}$.

b) If $\Phi_1 \Psi_1 / \Phi_2 \Psi_2 = -1$, then with (8) we get

$$\Phi_1^2 + \Phi_2^2 = \Psi_1^2 + \Psi_2^2 = 0 , \tag{17}$$

i.e. all the potential components vanish. In this case eq. (9) is satisfied with $b = 0$.

III. The stationary case*

Here we shall make use of the fact that in the cylindrical stationary case it is always true that

$$F^{12} = 0\,; \tag{18}$$

$$F_{34} = 0\,. \tag{19}$$

(19) immediately follows from (4). In order to obtain (18) we integrate the first two Maxwell equations

$$(g^{1/2}F^{21})_{,1} = 0\,; \qquad (g^{1/2}F^{12})_{,2} = 0 \tag{20}$$

and so we obtain $g^{1/2}F^{12} = \text{const.}$ We shall assume that at infinity $F^{12} = 0$. From this we have const. $= 0$ and so (if the metric is regular) everywhere in the space $F^{12} = 0$. Then we may choose the vector potential to have the following form:

$$A_\mu = (0, 0, \Phi, \Psi)\,. \tag{21}$$

Furthermore, from (18) and (19) we may conclude that the cylindrical stationary electromagnetic field has the property

$$T^1_1 + T^2_2 = T^3_3 + T^4_4 = 0\,, \tag{22}$$

the advantage of which fact will be taken in the proof of the following theorem.

In the presence of an origin-centred nonrelativistic electric monopole and magnetic dipole there is a flow of impulse in the $d\varphi$ direction. Now we shall consider the analogue relativistic fields. Then we have $T_{14} = T^{14} = T_{24} = T^{24} = 0$, from which it can be shown that $g_{14} = g^{14} = g_{24} = g^{24} = 0$. If we use canonical co-ordinates, the metric has the form

$$[g_{\mu\nu}] = \begin{bmatrix} \alpha^2 & & & \\ & \alpha^2 & & \\ & & \beta^2 & \varepsilon \\ & & \varepsilon & -\gamma^2 \end{bmatrix}. \tag{23}$$

We stress that the use of (23) is not necessary, exact solutions of the electrovac equations are known which have a very simple form in other co-ordinate systems [3]. Nevertheless, it can be shown [4] that any cylindrical stationary metric can be put into the form (23). (23) can always be diagonalized to have the signature $(+\,+\,+\,-)$.

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* The results presented in Sections III and IV are not new, but have been found independently by a number of authors.

The computation of Christoffel symbols yields the following nonvanishing components*

$$\begin{aligned}
&\Gamma^{1}_{11} = \Gamma^{2}_{12} = -\Gamma^{1}_{22} = \frac{\alpha_1}{\alpha}\,; \qquad \Gamma^{2}_{22} = \Gamma^{1}_{12} = -\Gamma^{2}_{11} = \frac{\alpha_2}{\alpha}\,;\\
&\Gamma^{3}_{i3} = (\gamma^2 \beta\beta_i + \varepsilon_i\, \varepsilon/2)/(\beta^2\gamma^2 + \varepsilon^2)\,;\\
&\Gamma^{4}_{i3} = (\varepsilon\beta\beta_i - \varepsilon_i\, \beta^2/2)/(\beta^2\gamma^2 + \varepsilon^2)\,; \qquad \Gamma^{i}_{33} = -\beta\,\beta_i/\alpha^2\,;\\
&\Gamma^{3}_{i4} = (\varepsilon_i\, \gamma^2/2 - \varepsilon\gamma\gamma_i)/(\beta^2\gamma^2 + \varepsilon^2); \qquad \Gamma^{i}_{34} = -\varepsilon_i/(2\alpha^2)\,;\\
&\Gamma^{4}_{i4} = (\varepsilon_i\, \varepsilon/2 + \beta^2\gamma\gamma_i)/(\beta^2\gamma^2 + \varepsilon^2)\,; \qquad \Gamma^{i}_{44} = \gamma\gamma_i/\alpha^2\,;\\
&\Gamma^{\mu}_{i\mu} = 2\alpha_i/\alpha + (\gamma^2\beta\beta_i + \beta^2\gamma\gamma_i + \varepsilon\varepsilon_i)/(\beta^2\gamma^2 + \varepsilon^2)\,.
\end{aligned} \tag{24}$$

Finally by very tedious calculation we get the components of the Ricci tensor. For later use we put down the proper linear combinations of these components:

$$R_{11} = \sum_i \left[(\ln\alpha)_{ii} + (-1)^i \frac{1}{2}\,\frac{\alpha_i N_i}{\alpha N}\right] + \frac{(\sqrt{N})_{11}}{\sqrt{N}} - \frac{1}{2N}\,[(\beta^2)_1 (\gamma^2)_1 + (\varepsilon_1)^2]\,; \tag{25}$$

$$R_{22} = \sum_i \left[(\ln\alpha)_{ii} - (-1)^i \frac{1}{2}\,\frac{\alpha_i N_i}{\alpha N}\right] + \frac{(\sqrt{N})_{22}}{\sqrt{N}} - \frac{1}{2N}\,[(\beta^2)_2 (\gamma^2)_2 + (\varepsilon_2)^2]\,; \tag{26}$$

$$R^3_3 + R^4_4 = \frac{1}{\alpha^2} \sum_i \frac{(\sqrt{N})_{ii}}{\sqrt{N}}\,; \tag{27}$$

$$\begin{aligned}
R^3_3 - R^4_4 = \frac{1}{\alpha^2 N^2} \sum_i \Bigg[\beta^4\gamma^4 \left(\frac{\beta_{ii}}{\beta} - \frac{\gamma_{ii}}{\gamma}\right) + \\
+ \beta^2\gamma^2\,\varepsilon^2 \left(\frac{\beta_{ii}}{\beta} - \frac{\gamma_{ii}}{\gamma} + \frac{\beta_i^2}{\beta^2} - \frac{\gamma_i^2}{\gamma\varepsilon} - \frac{\beta_i\,\varepsilon_i}{\beta\varepsilon} - \frac{\gamma_i\,\varepsilon_i}{\gamma^2}\right)\Bigg]\,;
\end{aligned} \tag{28}$$

$$\begin{aligned}
R_{12} = -\frac{1}{2}\,\frac{\alpha_1 N_2}{\alpha N} - \frac{1}{2}\,\frac{\alpha_2 N_1}{\alpha N} + \frac{(\sqrt{N})_{12}}{\sqrt{N}} - \\
- \frac{1}{2N}\left[\frac{(\beta^2)_1(\gamma^2)_2 + (\beta^2)_2\,(\gamma^2)_1)}{2} + \varepsilon_1\,\varepsilon_2\right];
\end{aligned} \tag{29}$$

$$R_{34} = \frac{\varepsilon}{2\alpha^2 N} \sum_i \left[\beta^2\gamma^2 \left(\frac{\varepsilon_{ii}}{\varepsilon} - \frac{\beta_i\,\varepsilon_i}{\beta\varepsilon} - \frac{\gamma_i\,\varepsilon_i}{\gamma\varepsilon} + 4\,\frac{\beta_i\,\gamma_i}{\beta\gamma}\right) + \varepsilon^2\,\frac{\varepsilon_{ii}}{\varepsilon}\right], \tag{30}$$

where

$$N \equiv \beta^2\gamma^2 + \varepsilon^2\,.$$

---

* Here and in what follows it is assumed to have $i = 1,2$ and that the summation convention holds for i. Moreover, see footnote on page 394.

IV. Extension of Weyl's theorem

Theorem. In a stationary cylindrical universe having the metric form (23) the number of independent nonvanishing components of the metric tensor can be reduced from four to three. Either of the unknown functions in (23) except α can be eliminated.

Proof. We shall use the consequence of (22):

$$R_3^3 + R_4^4 = 0. \tag{31}$$

(27) can be written by means of this equation in the remarkably simple form

$$\sum_i \left[(\beta^2\gamma^2 + \varepsilon^2)^{1/2}\right]_{ii} = 0\,. \tag{32}$$

From now on we may follow the order of ideas found by WEYL and others [5]. Eq. (32) means that $r' = (\beta^2\gamma^2 + \varepsilon^2)^{1/2}$ is a harmonic function of r and z, furthermore, there exists a conjugate harmonic function $z'(r, z)$ such that $r' + iz' = f(r + iz)$. Making use of the conformal transformation $(r, z) \rightarrow (r', z')$ one can eliminate one of the functions β, γ or ε.

Finally we shall present the field equations in a case when they take a very simple form. The components of the metric are chosen in the following manner:

$$\begin{aligned} \alpha^2 &= f^2 \\ \beta^2 &= h^2 r^2 \\ \varepsilon &= -h^2 r^2 \omega \end{aligned} \tag{33}$$

and so

$$\gamma^2 = 1/h^2 - h^2 r^2 \omega^2\,.$$

Then the line element has the form:

$$ds^2 = f^2(dr^2 + dz^2) + r^2 h^2 (d\varphi - \omega dt)^2 - (1/h^2)dt^2\,. \tag{34}$$

The Maxwell equations remaining to be solved are:

$$\left\{ r\left[\left(\frac{1}{h^2 r^2} - h^2\omega^2\right)\Phi_i - h^2\omega\Psi_i\right]\right\}_{,i} = 0\,; \tag{35}$$

$$\left\{ r\left[h^2\omega\Phi_i + h^2\Psi_i\right]\right\}_{,i} = 0\,. \tag{36}$$

(36) is equivalent to the statement

$$\left.\begin{aligned} h^2 r\omega\Phi_1 + h^2 r\Psi_1 &= \chi_2\,; \\ h^2 r\omega\Phi_2 + h^2 r\Psi_2 &= -\chi_1 \end{aligned}\right\} \tag{37}$$

Now, the Maxwell equations may be written

$$\left(\frac{\Phi_i}{h^2 r}\right)_i = \omega_1 \chi_2 - \omega_2 \chi_1 ; \tag{38}$$

$$\left(\frac{\chi_i}{h^2 r}\right)_i = - \omega_1 \Phi_2 + \omega_2 \Phi_1 . \tag{39}$$

(39) is the consequence of the identity $\Psi_{12} = \Psi_{21}$.

The Einstein equations are in this case:

$$\begin{aligned} \Delta \ln f - \frac{2}{r}\frac{f_1}{f} + \frac{2}{r}\frac{h_1}{h} + 2\frac{h_1^2}{h^2} - \frac{1}{2} h^4 r^2 \omega_1^2 = \\ = \frac{K}{h^2 r^2} (\Phi_1^2 - \Phi_2^2 + \chi_1^2 - \chi_2^2) ; \end{aligned} \tag{40}$$

$$\Delta \ln f + 2\frac{h_2^2}{h^2} - \frac{1}{2} h^4 r^2 \omega_2^2 = - \frac{K}{h^2 r^2} (\Phi_1^2 - \Phi_2^2 + \chi_1^2 - \chi_2^2) ; \tag{41}$$

$$- \frac{1}{r}\frac{f_2}{f} + \frac{1}{r}\frac{h_2}{h} + 2\frac{h_1 h_2}{h^2} - \frac{1}{2} h^4 r^2 \omega_1 \omega_2 = \frac{2K}{h^2 r^2} (\Phi_1 \Phi_2 + \chi_1 \chi_2) ; \tag{42}$$

$$\Delta \ln h + \frac{1}{2} h^4 r^2 \omega_i^2 = \frac{K}{h^2 r^2} (\Phi_i^2 + \Psi_i^2) ; \tag{43}$$

$$(\omega_i h^4 r^3)_i = - 4K (\Phi_1 \chi_2 - \Phi_2 \chi_1) , \tag{44}$$

where K denotes $\varkappa/8\pi$. These equations demonstrate the possibility of a significant simplification of the stationary field equations in consequence of the proved theorem. The only known solution of these equations with non-vanishing Φ, Ψ and ω is obtained by the methods of gravitational radiation theory and the transformation of it to our coordinate system yields very complicated expressions.

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REFERENCES

1. J. L. Synge, The General Relativity, North-Holland Publishing Co., p. 367, 1960.
2. G. Tauber, Canad. Journal of Phys., **35**, 477, 1957.
3. E. T. Newman, E. Couch, K. Chinnapared, A. Exton, A. Prakash and R. Torrence, Journal of Math. Phys., **6**, 918, 1965.
4. A. Papapetrou, Ann. Inst. Henri Poincaré, IV. 83, 1966.
5. See [1], p. 311.

НЕКОТОРЫЕ СВОЙСТВА ЦИЛИНДРИЧЕСКИХ ЭЛЕКРОВАКУУМНЫХ ПОЛЕЙ

З. ПЕРЬЕШ

Резюме

Показывается, что статические цилиндрические электрические и магнитные поля в некотором смысле линейно связаны. Далее суммируются известные свойства статического случая: доказывается распространение теоремы Вейла и представляются уравнения поля, полученные посредством этой теоремы в специальной системе координат.

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COMMUNICATIONES BREVES

THE GROUND-STATE GAMMA DECAY OF THE ^{49}Ca GROUND-STATE ANALOGUE IN ^{49}Sc*

By

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The most convenient target nuclei for the study of the isobaric analogue resonances seem to be the double closed-shell nuclei ^{48}Ca and ^{208}Pb. Unfortunately, inconsistent results have been reported ($\Gamma_n = 2.5$ keV and $\Gamma_n = 0.08$ keV [2]) for the parameters of the ^{49}Ca ground-state analogue in ^{49}Sc, occurring as a resonance at $E_p = 1978$ keV in the ^{48}Ca + p reactions [1]: **Chasman** et al. [3] have measured the strength of the 11.5 MeV ground-state transition relative to the yield of the ^{48}Ca(p, n, γ) reaction, and with $\Gamma_n = 2.5$ keV they give for the partial width of the 11.5 MeV ground-state transition $\Gamma_{\gamma 0} = 3.5$ eV. The aim of the present work is to clarify this situation by the help of an independent measurement of $\Gamma_{\gamma 0}$.

The experiment was performed with the proton beam of a 2.5 MeV Van de Graaff generator with an energy spread and calibration uncertainty of about 1 keV. The excitation function of the $E_\gamma = 11.5$ MeV ground-state transition (dots in Fig. 1) was measured with a 12.7 cm×12.7 cm scintillation counter placed at $\vartheta_\gamma = 90°$ at a distance of 28 cm from the target to avoid pile-up effects. (Distinct measurements have shown that the pile-up effects are negligible at this distance.) The excitation function for all γ-rays with $E_\gamma > 3.5$ MeV (Fig. 1, solid curve) was obtained simultaneously with a 7.5 cm×7.5 cm NaI(Tl) detector placed close to the target at $\vartheta_\gamma = 0°$. The 2 keV thick $CaCO_3$ target, enriched to 60% in $^{48}CaCO_3$, was evaporated onto a thick Au backing. The absolute value of the (p, γ) cross section has been determined with a different experimental set up. The $CaCO_3$ was evaporated onto a thin Au foil. The number of ^{48}Ca atoms in this target was determined from a measurement of the proton elastic-scattering cross section at $\vartheta_p = 167°$ and $E_p = 2300$ keV, to avoid the nuclear scattering contribution to the Rutherford cross section.

The yield of ground-state γ-rays per incident proton was measured at the top of the resonance ($E_p = 1978$ keV) with the γ-ray counter placed at

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23 cm from the target. The cross section obtained in this way is $\sigma_{p\gamma}(E_\gamma = 11.5$ MeV, $E_p = 1978$ keV, $\vartheta_\gamma = 90°) = 55 \pm 14$ μb/sr.

The error given here is the estimated uncertainty in the calculation of the γ-ray counter efficiency. If one assumes an isotropic angular distribution, this numerical value for the cross section yields $\Gamma_p\Gamma_\gamma/\Gamma_{\text{tot}} = 0.07 \pm 0.02$ eV. If one assumes [2] that $\Gamma_p \approx \Gamma_{\text{tot}}$, the partial width for the ground-state γ-transition is obtained as $\Gamma_{\gamma 0} = 0.07 \pm 0.02$ eV in disagreement with the value 3.5 eV given in [3]. This large value for $\Gamma_{\gamma 0}$ comes from the overestimation of Γ_n. Working with $\Gamma_n = 0.08 \pm 0.06$ keV [2] (this Γ_n gives for the (p, n) cross section a value in agreement with the measured [4] 22 mb) one obtains from the data of [3] the value $\Gamma_{\gamma 0} = 0.11 \pm 0.08$ eV, in accord with the present result.

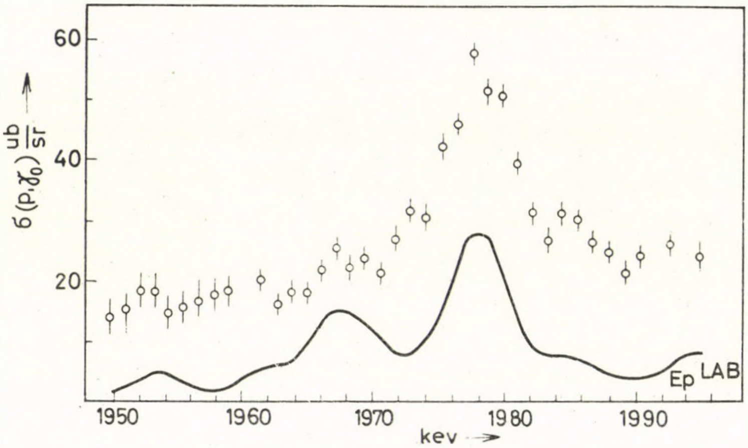

Fig. 1. Excitation functions for the ^{48}Ca(p, γ)^{49}Sc reaction. The solid line shows the excitation of all the γ-rays of energy > 3.5 MeV (in arbitrary units); the dots indicate the yield of 11.5 MeV γ-rays in μb/sr at 90°

It is interesting to note that this ground-state transition is found to be extremely weak in comparison with the single-particle estimate $\Gamma_{\gamma 0} = 1.74$ eV. Nevertheless a two-particle-one-hole model [5] gives for this resonance $\Gamma_{\text{tot}} = 2$ keV and $\Gamma_{\gamma 0} = 0.03$ eV, i.e. it gives the correct order of magnitude for the strength of this transition.

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REFERENCES

1. K. W. JONES, J. P. SCHIFFER, L. L. LEE, A. MARINOV and J. L. LERNER, Phys. Rev., **145**, 894, 1966.
2. R. A. RICCI, G. CHILOSI and G. B. VINGIANI, Int. Conf. on Nuclear Structure, Tokyo, 1967.
3. C. CHASMAN, K. W. JONES, R. A. RISTININ and J. T. SAMPLE, Phys. Rev. Lett., **18**, 219, 1967
4. L. M. EL NADI, OMAR, E. BADAWY, A. EL-SOUROGY, D. A. E. DARWISH and V. Y. GONTCHAR, Nucl. Phys., **64**, 449, 1965.
5. J. ZIMÁNYI, to be published.

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AVERAGE SPACING OF EXCITED LEVELS IN ^{49}V

By

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The average level spacing D in ^{49}V produced by ^{48}Ti(p, γ) ^{49}V reaction at proton energies $E_p = 970 - 1375$ keV was evaluated by DUBOIS [1] from the measured excitation function of a (p, γ) reaction on a thin target, using the formula

$$N = N_0 \, e^{-S/D},$$

where N is the number of resonances (excited levels) with a larger spacing than S keV. The result was $D = 4.0$ keV.

We have made a careful study of the excitation function of ^{48}Ti(p, γ) ^{49}V reaction in order to find isobar analogue resonances at proton energies $E_p = 1330 - 2140$ keV [2]. It seemed of interest to evaluate D from our data as well, to see the energy dependence of D.

In the measurement, a natural Ti target on Ta backing (relative abundance of $^{48}\mathrm{Ti} = 74\%$) of $\sim$2 keV thickness for 1 MeV protons was bombarded with the analyzed proton beam from the 2.5 MeV Van de Graaff generator of the Institute. The energy spread of the proton beam was $\sim$1 keV. The energy calibration of the generator was performed with ^{19}F(p, α, γ) ^{16}O and ^{58}Ni(p, γ) ^{59}Co reactions. The absolute value of proton energy was found to be accurate to ± 2 keV. The γ-radiation with energies from 4 MeV to 8 MeV was measured with a 12.7 cm by 12.7 cm NaI(Tl) scintillation counter at 90° to the proton beam.

The measured excitation function, in which $\approx$85 distinct resonances could be identified, is plotted in Fig. 1. The analysis by eq. (1), shown in Fig. 2, proves that many close lying resonances ($S < 5$ keV) cannot be identified because of the finite resolution. Nevertheless, the average level density, as determined from the slope of the logarithmic curve (Fig. 2), gives the value of $D = 3.0$ keV.

This result compared with that of DUBOIS shows that D decreases with increasing excitation energies of ^{49}V, as is to be expected from the general trend observed in level densities.

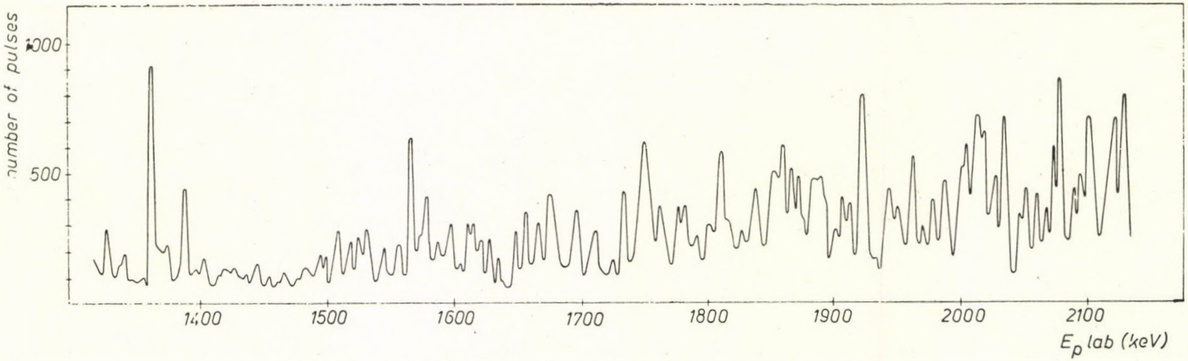

Fig. 1. Excitation function of the ^{48}Ti(p, γ)^{49}V reaction

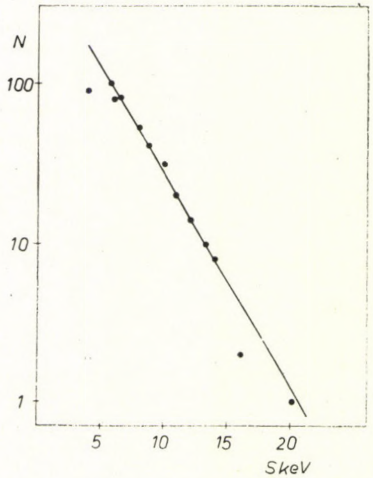

Fig. 2. Determination of the average level spacing

For the evaluation of the results, use was made of the LANG—LE COUTEUR formulae [3], as discussed in [4]. In this formulation two parameters, namely, the pair energy Δ and the single particle level density g, are used in the evaluation of the function $\varrho(U)$ which gives the level density at any excitation energy U. If the relationship between the value of Δ and the atomic number A is assumed to be true, only the value g remains to be fitted to the experimental value of $\varrho(U) = 1/D(U)$. Calculating with $U = 9.3$ MeV, that is the average excitation energy covered by the measurement of DUBOIS [1, 4], we obtained $g = 5.2$ MeV and for the excitation energy $U = 9.95$ MeV in the present measurement $g = 5.1$ MeV.

REFERENCES

1. J. DUBOIS, Nuclear Physics, **23**, 537, 1961.
2. I. FODOR, I. DEMETER, L. KESZTHELYI, I. SZENTPÉTERY, Z. SZŐKEFALVI-NAGY, J. SZÜCS, L. VARGA and J. ZIMÁNYI, Nuclear Physics, **A 116**, 167, 1968.
3. D. W. LANG and K. J. LE COUTEUR, Nuclear Physics, **14**, 21, 1959.
4. A. ASPINALL, G. BROWN and J. G. B. HAIGH, Nuclear Physics, **52**, 630, 1964.

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A REMARK ON THE ALGEBRA OF SPACE-COMPONENTS OF CURRENT DENSITIES

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Current commutation relations supplemented with pole dominance assumptions (such as PDDAC or ϱ-meson dominance, etc.) lead to a number of successful predictions in strong as well as weak or electromagnetic interactions of hadrons. The information contained in an algebra is of course the richer, the larger is the set of specified commutators. For the algebra of vector and axial-vector current densities the quark model gives the chiral $U(6)\otimes \otimes U(6)$ [1]. The simple commutators derived from the quark model, however, can be modified by Schwinger-terms proportional to the derivatives of δ-function [2]. Another proposal for the density algebra is based on gauge fields [3]. As is known, the gauge field algebra (GFA) coincides with the quark current algebra (QCA) in time-component time-component commutators, but has an additional C-number Schwinger-term in time—space component commutators and differs from chiral $U(6)\otimes U(6)$ giving zero for the space—space component commutators.

The purpose of this note is to show that under certain assumptions (accepted in most cases) the GFA leads to a consistent system of sum rules, whereas in QCA the equations coming from space—space component commutators contradict those coming from time—space component commutators. Our assumptions are as follows: 1. the sum rules can be saturated by the known one particle states (and the disconnected part of two particle intermediate states [4]); 2. all the form factors obey a once subtracted dispersion relation; 3. the dispersion relations for the form factors are dominated bypoles. We exploit the commutation relations making use of the r-sum-rules for vertex functions as derived by Halliday and Landshoff [5].

Let us first consider

$$t^A_{\mu\nu}(k,p)_{rst} = \frac{1}{2i}\int d^4 x e^{ikx} \langle 0| \left[A_\mu(x)_r, V_\nu(0)_s\right] |A1(p)_t\rangle \tag{1}$$

$$= \varepsilon_{rst}\, \varepsilon^\varrho \left\{a_1 p_\mu k_\nu k_\varrho + a_2 k_\mu p_\nu k_\varrho + a_3 p_\mu g_{\nu\varrho} + a_4 g_{\mu\varrho} p_\nu + \right.$$

$$\left. + b p_\mu p_\nu k_\varrho + c_1 g_{\mu\nu} k_\varrho + c_2 k_\mu g_{\nu\varrho} + c_3 g_{\mu\varrho} k_\nu + d k_\mu k_\nu k_\varrho \right\}$$

$$a_1 = a_1(k^2, \Delta^2), \ldots ; \quad \Delta = p - k\,.$$

Here $A_\mu(x)_r$ and $V_\nu(x)_s$ denote the vector and axial-vector current densities (or gauge fields), respectively, $A1(p)_t$ is an $A1$-meson state of momentum p, isospin index t $(r, s, t = 1, 2, 3)$ and polarisation vector ε. From the equal time commutator of space-components $(\nu, \mu = 1, 2, 3)$ we can derive in the usual way [5] the sum rules [6]:

$$\frac{1}{\pi}\int dk_0\, p_0\, a_{1,2,3,4} = \frac{1}{\pi}\int dk_0\, p_0\, b = \frac{1}{\pi}\int dk_0\, p_0\, c_{1,2,3} = \frac{1}{\pi}\int dk_0\, p_0\, d = 0\,, \tag{2a}$$

$$\frac{1}{\pi}\int dk_0\, k_0\, a_{1,2} = \frac{1}{\pi}\int dk_0\, k_0\, b = \frac{1}{\pi}\int dk_0\, k_0\, d = 0\,, \tag{2b}$$

$$\frac{1}{\pi}\int dk_0\, k_0\, c_1 = \begin{cases} 2\dfrac{M_A^2}{G_A\sqrt{2(2\pi)^3}} & \text{in QCA}\,, \\ 0 & \text{in GFA}\,. \end{cases} \tag{2c}$$

Setting $\boldsymbol{q}\cdot\boldsymbol{k} = rq_0^2$ and $\boldsymbol{k}^2 = r^2q_0^2 - R$ $(0 < r < 1,\ R \text{ real})$ in the $q_0 \to \infty$ limit we get from eqs. (2*a*—*c*) the r-sum-rules:

$$\frac{1}{2\pi}\int dn a_{1,2,3,4} + A_{1,2,3,4}(\infty) = \frac{1}{2\pi}\int dn b + B(\infty) =$$

$$= \frac{1}{2\pi}\int dn c_{1,2,3} + C_{1.2,3}(\infty) = \frac{1}{2\pi}\int dn\, d + D(\infty) = 0\,, \tag{3a}$$

$$\frac{1}{2\pi}\int dnra_{1,2} + A^r_{1,2}(\infty) = 0 \quad \frac{1}{2\pi}\int dnrb + B^r(\infty) = 0 \quad \frac{1}{2\pi}\int dnrd + D^r(\infty) = 0 \tag{3b}$$

$$\frac{1}{2\pi}\int dnrc_1 + c_1^r(\infty) = \begin{cases} \dfrac{2M_A^2}{G_A\sqrt{2(2\pi)^3}} & \text{in QCA}\,, \\ 0 & \text{in GFA}\,, \end{cases} \tag{3c}$$

$$k^2 = -rn + R\,, \quad \Delta^2 = (1-r)\,n + R + M_A^2\,.$$

Here $A_1(\infty), \ldots, D^r(\infty)$ denote the infinite mass contributions. The sum-rules can be evaluated under assumptions 1—3. As a result we get linear algebraic equations for the coupling constants and subtraction constants in form factors [7].

It can be easily shown that the equations resulting from eq. (3*a*) are trivial in the sense that they follow from divergence conditions (CVC and PCAC) and from the standard reduction technique applied to the different form factors. Eqs. (3*b*) are nontrivial and can be compared with the following equations resulting from time-component space-component commutators [9]:

$$\frac{1}{2\pi}\int dn\,(a_1 + rd) + A_1(\infty) + D^r(\infty) = \frac{1}{2\pi}\int dn\,(b + ra_2) +$$

$$+ B(\infty) + A_2^r(\infty) = \frac{1}{2\pi}\int dn\,(a_2 + rd) + A_2(\infty) +$$

$$+ D^r(\infty) = \frac{1}{2\pi}\int dn\,(b + ra_1) + B(\infty) + A_1^r(\infty) = 0\,. \tag{4}$$

Thus we see that the only new information in (3*b*) is the second equation.

Eq. (3*c*) is also new and gives in pole dominance approximation (in the notations of [9]):

$$K_1' - H_5' = \begin{cases} \dfrac{2M_A^4}{G_A^2} & \text{in QCA}\,, \\ 0 & \text{in GFA}\,. \end{cases} \tag{5}$$

Exactly the same procedure can be applied to

$$t_{\mu\nu}^V(k, p)_{rst} = \frac{1}{2i}\int d^4x e^{ikx} \langle 0|\left[A_\mu(x)_r, A_\nu(0)_s\right]|\varrho(p)_t\rangle \tag{6}$$

instead of $t_{\mu\nu}^A$ defined in eq. (1). The r-sum-rules are the same as in eq. (3*a*—*c*), the only difference ist hat now M_A^2/G_A is replaced by M_V^2/G_V. The new information here is also the second of eq. (3*b*) and eq. (3*c*). From eq. (3*c*) we have now:

$$2H_1' = \begin{cases} -\dfrac{2M_V^4}{G_V^2} & \text{in QCA,} \\ 0 & \text{in GFA}\,. \end{cases} \tag{7}$$

Comparing eqs. (5) and (7) with the results following from time-component space-component commutators (see in [9]):

$$H_1' = K_1' = H_5' = 0 \tag{8}$$

we see that only the GFA gives a consistent system of equations. In fact, it can be shown that all the equations resulting in pole dominance approximation from (3*a*—*c*) follow from the results of [9] if we consider GFA. (As a C-number the Schwinger term does not give any contribution to $t_{\mu\nu}^{A,\,V}$ the predictions of QCA and GFA coincide in the time-component space-component case.)

Finally, the same conclusions can be reached in the case considered in [8] (axial-vector vector commutator taken between the vacuum and one pion state). Here also only the GFA gives consistent equations [10].

In conclusion, under assumptions 1—3. only GFA leads to a consistent system of equations for coupling constants (and subtraction constants in form factors). As the assumptions 1—3. are, in fact, far from being justified (although very often made) we cannot conclude from this that QCA is wrong. In any case, a positive statement which can be made is that in the frame of these approximations the GFA gives good results.

The author thanks Drs. F. Csikor and G. Pócsik for valuable discussions.

REFERENCES

1. M. Gell-Mann, Phys. Rev., **125,** 1062, 1962; Physics, I. 63, 1964.
2. J. Schwinger, Phys. Rev. Letters, **3,** 296, 1959; see also F. Csikor, G. Pócsik, On the operator Schwinger terms in current algebras, ITP-Budapest Report No. 200, 1967 and the work of Buccella, Veneziano, Gatto and Okubo quoted in [10].
3. T. D. Lee, S. Weinberg and B. Zumino, Phys. Rev. Letters, **18,** 1029, 1967.
4. D. Amati, R. Jengo and E. Remiddi, Nuovo Cim., **51,** 999, 1967.
5. I. G. Halliday and P. V. Landshoff, Nuovo Cim., **51,** 980, 1967.
6. Here and in what follows we use the notations of [9], e.g.:

$$\langle 0 |\, A_{\mu}(0)_r \,| A1(k)_r \rangle = - M_A^2\, \varepsilon_{\mu} / [G_A \sqrt{2(2\pi)^3}]$$

7. Besides the more stringent restrictions on intermediate states the advantage of considering sum rules for vertex functions lies in the linearity of the resulting algebraic equations.
8. F. Csikor and I. Montvay, Nuclear Physics, B5, 492, 1968.
9. F. Csikor and I. Montvay, Nuclear Physics, B7, 268, 1968. In this work the width of ϱ- and A1-mesons as well as the charge radius of pions are calculated in terms of F_π and G_v in good agreement with experiment. Note that, besides eqs. (4), there are also other equations coming from time-component space-component commutators.
10. It can be shown that the situation is unchanged in both cases if we saturate the r-sum-rules with an arbitrary (finite) number of (ϱ, A1- and π-like) poles. We also note that in GFA the Jacobi identity considered by F. Buccella, G. Veneziano, R. Gatto and S. Okubo, Phys. Rev., **149,** 1268, 1966 holds trivially. Thus, the necessity for additional q-number Schwinger terms does not emerge. The GFA gives zero also for the r.h.s. of eq. (25) of this work, in coincidence with the possible (ad hoc) modification proposed by these authors.

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A SIMPLE HARTREE—FOCK CALCULATION FOR He^4

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(Received 24. VII. 1968)

One of the usual ways to calculate the nuclear Hartree—Fock energy is to express the unknown radial parts of the single-particle wave-functions $\psi_i(x)$ (x denotes all the variables including the spin and isospin) as sums of oscillator functions. By letting the unknown expansion-coefficients vary one gets energy-minima. (See e.g. [1].) The agreement between the experimental data and the calculated energy-minima is rather poor.

Our simple ansatz in the case of He^4 is that we cut the above mentioned expansion of ψ_i at the first term, i.e.: the radial part for all the four nucleons will be the same and can be written as follows

$$f(r) = N \exp\left(-\frac{1}{2}\alpha r^2\right), \tag{1}$$

where N is a normalizing constant and α is the only unknown parameter. With this restriction the general energy-expression for He^4 ($\hbar^2/M = 41.47$ MeVfm^2 is taken as unit),

$$E = \frac{1}{2}\sum_{i=1}^{4}\langle\psi_i|\hat{p}_i^2|\psi_i\rangle + \sum_{i<j=1}^{4}\langle\Psi_{ij}|V_{ij}|\Psi_{ij}\rangle - \frac{1}{8}\langle\Psi|\sum_{i<j=1}^{4}\hat{p}_i\hat{p}_j|\Psi\rangle, \tag{2}$$

will depend on only one parameter, i.e. on α independently of the form of the effective potential chosen for the interaction. In Eq. (2) ψ_i-s are the single-particle functions, Ψ_{ij}, Ψ are the two-, four-particle determinants constructed from ψ_i-s and the third term gives the center-of-mass correction due to the motion of the nucleus.

Our simplifying condition for the radial function seems to be quite reasonable considering the fact that in the ground-state of He^4 all four particles are in $1\ s_{1/2}$ state.

The calculation was carried out with the Tabakin potential [2]. We got for the energy minimum, $E_{\min} = -18.98$ MeV at $\alpha = 0.8679$ fm^{-2}. The experimental binding energy is -28.296 MeV [3]. Bassichis et al. [1] got $E = -12.78$ MeV after varying the expansion-coefficients at fixed α.

This example shows that our not too arbitrary ansatz on the radial part gives rather good results compared with a more complicated method. Obviously, with this simple method one cannot expect good results in the case of nuclei with higher mass-number, because the present method relies heavily on the simple properties of He^4.

Hence further investigations are needed with less restricted wavefunctions for He^4, and calculations have to be made for nuclei with higher mass number.

The author wishes to thank Prof. P. GOMBÁS for his interest in this work. Thanks are also due to Dr. D. KISDI for helpful advice and discussions.

REFERENCES

1. W. H. BASSICHIS, A. K. KERMAN and J. P. SVENNE, Phys. Rev., **160**, 746, 1967.
2. F. TABAKIN, Annals of Physics, **30**, 51, 1964.
3. A. H. WAPSTRA, Handb. d. Phys., **38/1**, 1, 1958.

Acta Physica Academiae Scientiarum Hungaricae, Tomus 25 (4) pp. 413—415 (1968)

INVESTIGATIONS OF THE I—V CHARACTERISTICS OF ZnS SINGLE CRYSTALS

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We investigated a ZnS single crystal of large resistivity (about $10^{10}-10^{13}$ Ωcm). The measurements were made at room temperature in air or vacuum ($10^{-4}-10^{-5}$/Hg mm) in the dark. We used crystals grown by transport reaction at 850—900°C (B5, B6 cubic) and at 1200—1300°C (mainly hexagonal). The contacts (In or SnO_2) were deposited by the "sandwich" method and we found no difference between the two kinds of contacts. Because of their high resistivity the samples were placed on teflon or sulphur holders. The d.c. measurements were carried out in the current region $10^{-14}-10^{-4}$ A and in the voltage region $10^{-2}-10^{-3}$ V. The thickness of samples was 100—500 μ.

Owing to the fact that at switching on a particular voltage at the beginning the current very strongly changed, the particular points of I—V characteristics were read after ten minutes when the equilibrium state was reached. We succeeded in finding series of both cubic and mainly hexagonal crystals on which the results could be reproduced rather well. In the case of mainly hexagonal crystals the time change of current induced by the applied voltage is more significant than in the case of cubic crystals but in both cases there are differences between the two characteristics taken consecutively. According to our experience this problem can be solved by keeping our samples at 80—150°C. From the differences found in one and the same crystal and from the fact that in particular cases after switching off the applied voltage a reverse and well measurable current flows, it can be concluded that as a result of the applied voltage a reverse electric field builds up in the crystal. When the applied voltage is switched off, one part of the reverse electric field decays (reverse current flows) but a certain part remains causing differences at a new measurement of the characteristics.

From the efficiency of heating to relatively low temperatures (80—150°C) we can conclude that crystal defects of low activation energy play a significant role in the formation of a built-in field. Gersun et al. [1] obtained similar results in connection with both heating and reverse current in the case of CdS of high resistivity of about $10^{11}-10^{12}$ Ω cm.

According to our observations the evacuation of the vessel containing the crystals ensures more reproducible circumstances. From the fact that the resistance values measured in air were 1—2 orders lower than those measured in vacuo, we, too, succeeded in eliminating the surface conduction as did RUSS [2].

The measured, mainly hexagonal crystals activated with Cu and the unactivated cubic and mainly hexagonal ones followed more or less the ideal characteristics obtained in the presence of space charge limited (SCL) currents, i.e. the first region follows Ohm's law (linear); in the second region $I = V_n$ where $n > 2$, and in the third region $I \sim V^2$ (Fig. 1). For some crystals the first or the second region was missing, the square law is not always exact and at great field strength we observed occasionally random deviations from the SCLC behaviour. The first region of the curve described above is missing owing to the high resistivity of the samples; we got measurable current only in the non-Ohmic region. The lack of the second region cannot be interpreted in such an obvious way, because according to our other observations it could not be supposed that our crystals approached the ideal trap-free case. Similar results were obtained by ENGLISH et al., who dealt with SiC single crystals [4]. The departures observed after the square region cannot be explained by means of SCLC as shown by VIŜCAKAS et al. [5].

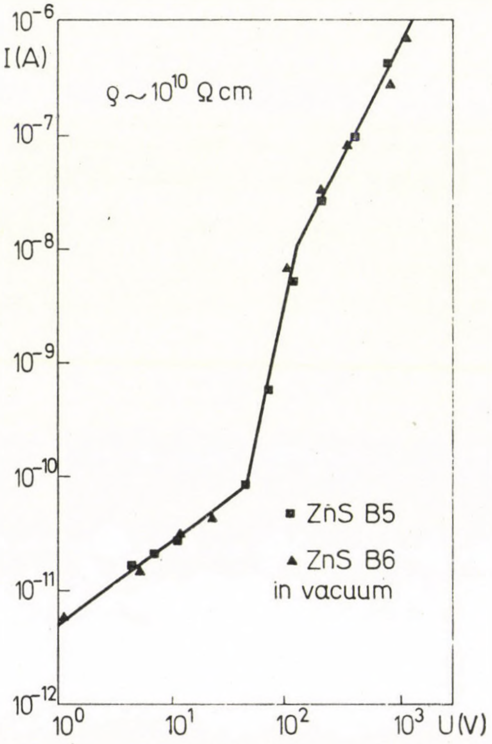

Fig. 1

Our measurements prove that the I—V characteristics follow the ideal SCLC ones rather well. The departures observed with a high field, the time change of current and the efficiency of heating require further detailed investigations.

The author wishes to thank Dr. Z. Bodó for many helpful discussions and advice.

REFERENCES

1. A. S. Gersun and B. L. Timan, Fiz. i Techn. Poluprovodnyikov, **2**, 488, 1968.
2. M. J. Russ, J. of Appl. Phys., **34**, 1831, 1963.
3. M. A. Lampert, Rept. Progr. Phys., **27**, 329, 1964.
4. A. C. English and R. E. Drews, Sci. Electr., **9**, 1, 1963.
5. J. Viŝcakas, P. Mackus and A. Smilga, phys. stat. sol., **25**, 331, 1968.

RECENSIONES

RICHARD SCHLEGEL: **Completeness in Science**

Appleton — Century — Crofts, New York, 1967

It happened a number of times in the course of history that man believed himself to be in the possession of something that could be called complete science. Aristotelian physics and Ptolemaic astronomy were thought of as complete in the Middle Ages. So, or nearly so, appeared Euclidean geometry and classical physics in the 19th century. Since then many things happened and perhaps man had never before felt himself so far removed from a complete and self-consistent world picture as in the 20th century. This negative feeling gives actuality to the problem dealt with in this volume. The author, Professor of Physics at the Michigan State University, puts forward the question: Is there any hope for science to reach completeness at all. The first three chapters of the book treat the problem from the logical and gnosiological point of view. Then the author studies those disciplines which, apparently, nearly reached their own boundaries: the Gödel theory of axiomatic systems, the paradoxes of set theory, cosmological models, the uncertainty relation of quantum theory and the smallest building blocks of matter. The discussion of each of these subjects suggests that the big open questions cannot be answered in the way they had been asked. Instead, it mostly happens that the paradoxes are resolved by the broadening framework from which they are looked at. The ultimate fate of this world is a problem that has always excited human imagination. This problem has not been solved but rather replaced by the much more difficult problem of modern cosmology. The final chapters contain the personal views of the author about the consequences of the incompleteness of science. Here Professor SCHLEGEL puts physics and metaphysics side by side but, in these chapters, he cannot attain the crystal-clear beauty of HERMANN WEYL's "Open World".

G. MARX

ROBERT KEMP ADAIR, EARLE CABELL FOWLER: **Strange Particles**

Interscience Publishers, New York—London, 1963

The unexpected flood of unstable elementary particles began with the strange particles. One of the great achievements of the 1950's was the discovery and study of particles with lifetimes of 10^{-10} sec. The authors were active participants of this research and their present booklet revives the excitement of those days. It is true, particles with lifetimes shorter than 10^{-20} sec have been discovered in the last five years or so, nevertheless the best way to follow for young physicists still seems to be the study of strange particles after the non-strange stable ones. To do this one could hardly offer a better guide than this volume which makes an easy reading but gives ample references to the literature.

G. MARX

BRIAN R. JUDD: **Second Quantization and Atomic Spectroscopy**

The John Hopkins Press, Baltimore, 1967

It is a cruel joke of Nature that quantum field theory, which has been worked out with enormous effort to describe elementary particles, did not succeed just in this domain while it helped much in clarifying the basic questions of the many-body problem. The physics

of fluids and of solid states have been completely reformulated by the quasi-particle method. The technique of creation and annihilation operators, progators and Feynman-graphs have been borrowed from this field to describe heavy nuclei. The aim of BRIAN R. JUDD, Professor at John Hopkins, in this book is to treat heavy atoms by the technique of creation and annihilation operators. His foremost aim is not completeness and precision but the presentation of the technique. Because of this there is not much said about concrete computations, but rather the emphasis is on some aspects of the calculational technique. We therefore recommend the book to those who know the elements of quantum field theory (e.g. from radiation theory) and want a short introduction to its applications in atomic and nuclear physics.

G. MARX

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