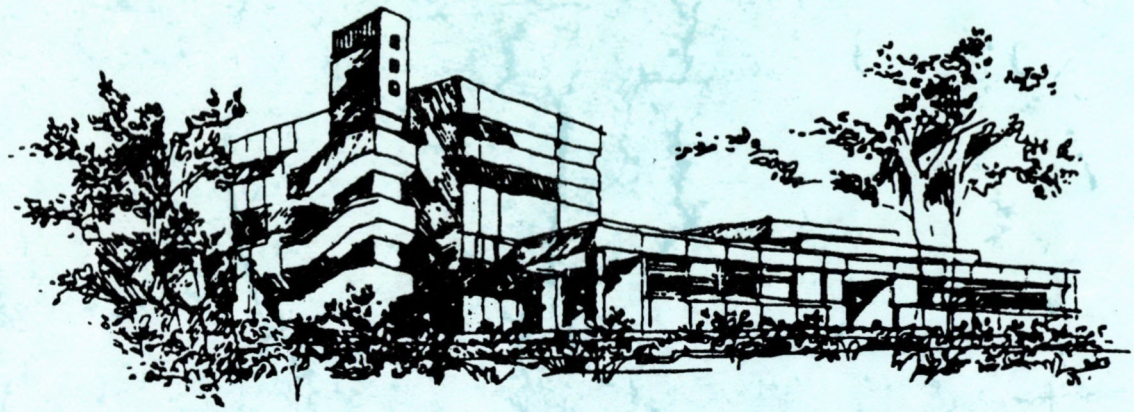
ATOMKI
ANNUAL REPORT
1998



INSTITUTE OF NUCLEAR RESEARCH
OF THE HUNGARIAN ACADEMY OF SCIENCES
DEBRECEN, HUNGARY

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Preface

In 1998 the process of "consolidation" of the institutes of the Hungarian Academy of Sciences was going on as planned beforehand. The declared objective of the consolidation has been to streamline the research network, to unite institutes of thematically related fields, to revise and modernize the scopes of their research, to stop parallel (but unrelated) research programs, to transfer some institutes to universities or to other organizations and to allot a firmer financial support to more concentrated research. The consolidation has preserved ATOMKI as the only research institute for natural sciences outside the capital. (Life sciences are counted separately.) The basic target prescribed for us is to reduce the number of employees to 190, which is a substantial but not vital reduction. This number includes only those employed at the cost of the Institute's budget granted by the state through the Academy; thus, e.g., it does not include PhD students, who all receive their fellowships from other sources. (The numbers contained in the pie diagrams to follow do include all members without regard to the source of their remuneration.) By the end of 1998 we were 16 fewer than a year before. Parallel to this cut, the budget from the Academy was substantially increased: from Ft 253,000,000 (1997) to Ft 334,000,000 (1998). The rate of inflation in 1997 was about 17%, and this budget increase is the first one in real terms for many years. It has become the routine in recent years (perhaps since 1993) that the Academy budget is only enough for keeping the Institute open and keeping its facilities in a state of readiness; all other research cost has had to be raised from research grants or from industrial contracts. In 1998, however, we managed to spend a small portion of the Academy budget on actual research projects. At the same time, the OTKA grants also tend to be higher than before. It has also improved our welfare that the irradiation and target transport system manufactured in ATOMKI for Egypt to facilitate ^{123}I production has been completed and delivered.

Early last year the Debrecen University Federation was established with the participation of all institutions of higher education in Debrecen. This is an interim organization, which will have existed until the end of 1999, and its sole role is to pave the way for the (re)unification of the universities. Informally, we have been taking part in the shaping of this Federation from the very beginning, and our participation was formalized in autumn by signing an associate membership treaty. We have two seats in the forty-strong Senate of the Federation and one seat in each of its committees. We have kept our affiliation with the Academy of Sciences; with the associate member's status we hope to preserve the close links with the member institutions of the University Federation as well.

Last year there was an international meeting on physics in Debrecen: a Japanese–Hungarian Joint Symposium on Physics in Modern Science and Technology, from 23–28 March. On this occasion, with Prof. H. Ejiri, then Director of the Research Centre for Nuclear Physics, Osaka University, I have signed a Cooperation Agreement between the two laboratories. Last year I have signed another significant

document of similar nature: a Memorandum of Understanding, which expresses our commitment to take part in the EXOGAM project, which is a project of γ-ray spectroscopy to be implemented at the radioactive beam facility of the SPIRAL (Systeme de Production d'Ions Radioactifs Accélérés en Ligne) facility at GANIL (Grand Accélérateur National d'Ions Lourds) at Caen.

The most notable scientific feat to mention of last year is the completion of a monograph with the co-authorship of a colleague, Dr. Kálmán Varga: *Stochastic Variational Approach to Quamtum-Mechanical Few-Body Problems*, by Y. Suzuki and K. Varga (Springer, 1998). A variational approach may be stochastic via the procedure applied in the construction of its trial function. The voluminous volume gives an introduction to the stochastic variational approaches to bound-state problems, and presents the intricate formalism implied by generalized Gaussian trial functions. Kálmán worked out this formalism in the early nineties in Debrecen, and implemented, refined and applied it at Niigata University and at the Institute of Physical and Chemical Research (RIKEN), Wako-shi, together with Professor Suzuki and his students, keeping intensive interaction with his mother institution, ATOMKI. The approach was developed to facilitate the microscopic description of light multicluster nuclei, but it turned out to be a very useful tool for any few-body systems. This approach and its copious applications have earned recognition in several fields. The monograph reviews only its applications to systems of structureless particles (as contrasted with clusters): systems of electrons and positrons, small normal and mesonic atoms and molecules, baryons as systems of three quarks, excitonic complexes, quantum dots and few-nucleon systems.

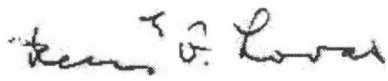
We celebrated the 70th birthday of our former director, Dénes Berényi, who is now President of the Debrecen Committee of the Hungarian Academy of Sciences in 1998. To mark this occasion, the 7th Workshop on Fast Ion–Atom Collisions, held in Debrecen on 9–11 September, was dedicated to him. This series of international workshops was initiated by him in 1981. This time he was honoured by several addresses of personal appreciation, and there was a lecture summarizing his work in this field by Prof. K.-O. Groeneveld (Johann-Wolfgang-Goethe-Universität, Frankfurt).

It belongs to the chronicle of the year that my immediate predecessor as Director, Prof. József Pálinkás, has been appointed Secretary of State at the Ministry of Education. Dr. Ede Hertelendi was awarded the prestigious Dénes Gábor Prize for innovative activity in scientific and technological fields. It is also my duty to report that I was awarded a "Prize of the Academy". The Sándor Szalay Prize of this Institute was now issued on the Day of Hungarian Science, on 3rd November: Dr. Endre Somorjai was awarded this prize for his research in nuclear astrophysics.

The story of the year is not complete without mentioning the Physics Week held in each March to address the general public. This time its subject has been symmetry in nature. As an experiment, we deliberately chose this esoteric subject, in an attempt to show something we are really fascinated ourselves. There were talks on the role of symmetry principles in physics in general, in particle physics, in mineralogy, in biology and on the teaching of symmetry principles in secondary schools. These talks were perhaps not as popular as if we had talked on ufos, but they attracted quite a lot of people, who, I hope, enjoyed them.

As a final remark, I should mention that last year we have re-organized our web-site at the address http://www.atomki.hu. This work is never really complete, but I can certainly affirm that you will have more valuable and more up-to-date information now than last year. This Annual Report will also be fully accessible through the web. Its computerized version has been prepared on the basis of LaTeX, and one can get access to it in HTML, DVI, PDF and PS(.GZ) formats.

Debrecen, 5 May 1999



Rezső G. Lovas
Director

Organizational structure of ATOMKI

DIRECTOR: Prof. R.G. Lovas

DEPUTY DIRECTORS:
Prof. Á.Z. Kiss
Dr. S. Mészáros

FINANCE OFFICER:
Dr. M. Pálinkás

SECRETARIAT
scientific secretary: Dr. Á. Kovách

INFORMATION SERVICES
chief librarian: Mrs. M. Nagy

ACCOUNTING
Headed by Mrs. J. Sas

BASIC SERVICES
& MAINTENANCE
headed by J. Bagi

SCIENTIFIC SECTIONS

DIVISION OF NUCLEAR PHYSICS
headed by Prof. J. Cseh

SECTION OF NUCLEAR SPECTROSCOPY
headed by Dr. A. Krasznahorkay
SECTION OF
ELECTROSTATIC ACCELERATORS
headed by Dr. E. Somorjai
SECTION OF THEORETICAL PHYSICS
headed by Dr. T. Vertse

DIVISION OF ATOMIC PHYSICS
headed by Prof. J. Pálinkás

SECTION OF ATOMIC COLLISIONS
headed by Dr. L. Sarkadi
SECTION OF ELECTRON SPECTROSCOPY
headed by Dr. D. Varga

CYCLOTRON SECTION
headed by Dr. F. Tárkányi

SECTION OF ENVIRONMENTAL
& EARTH SCIENCES
headed by Dr. E. Hertelendi

SECTION OF ELECTRONICS
headed by Dr. J. Gál

MECHANICAL WORKSHOP
headed by I. Gál

Data on ATOMKI

Personnel

At present the Institute employs 198 persons. The affiliation of personnel to units of organization and the composition of personnel are goven below.

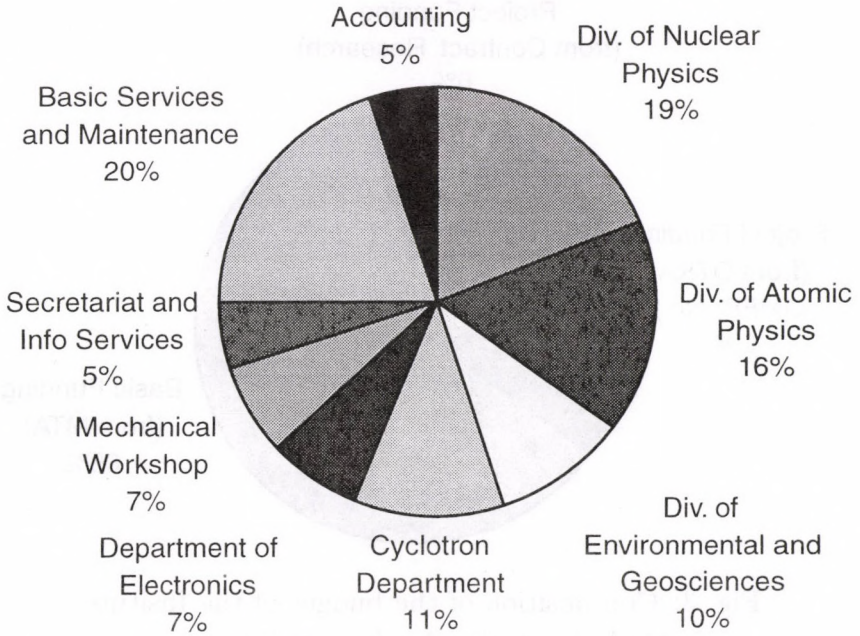


Fig. 1: Affiliation of personnel to units of organization

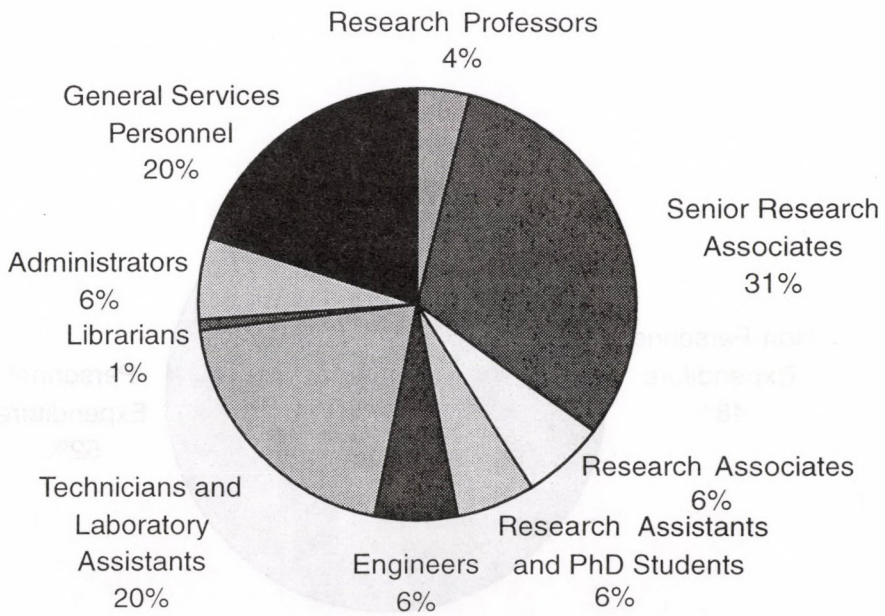


Fig. 2: Composition of personnel

Finance

In 1998 the total budget of the Institute was 512 million Hungarian Forints. The composition of the budget and the share of personnel expenditure within the budget are shown below.

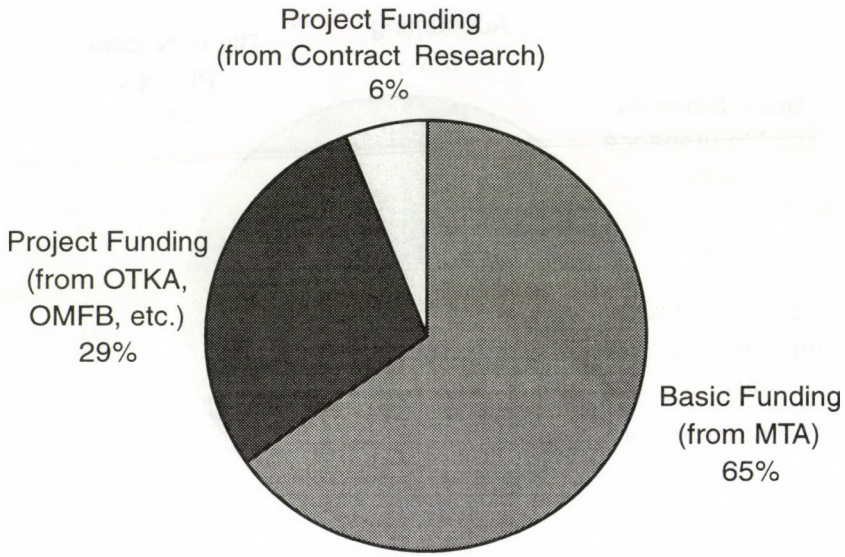


Fig. 3: Composition of the budget of the Insitute
MTA: Hungarian Academy of Sciences
OTKA: National Fund for Scientific Research
OMFB: National Committee for Technological Development

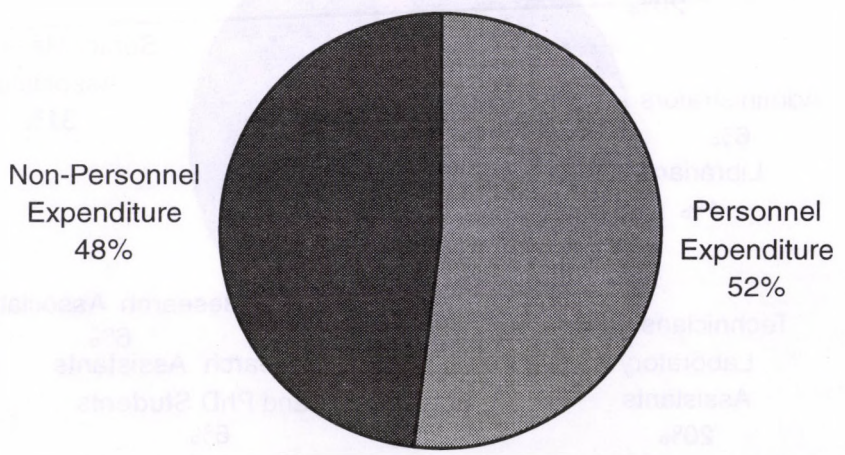


Fig. 4: Breakdown of expenditure into personnel and non-personnel expenditures

Contents

Reports

Publications and Seminars

General Physics

On the Coulomb Sturmian matrix elements of relativistic Coulomb Green's operators*

Balázs Kónya and Zoltán Papp

In quantum mechanics the knowledge of the Green's operator is equivalent to the complete knowledge of the system. An analytic basis representation of the Green's operator can tremendously simplify the actual calculations. If we know the Green's operator only of the asymptotic part of the Hamiltonian we can treat the remaining terms as perturbations and approximate them by finite matrices.

In a recent publication, Ref. [1], we have proposed a method for calculating matrix representation of Green's operators. If, in some basis representation, the Hamiltonian possesses an infinite symmetric tridiagonal (Jacobi) matrix structure the corresponding Green's operator can be given in terms of continued fractions. In Ref. [1], this theorem were exemplified with the Green's operator of the non-relativistic Coulomb and harmonic oscillator Hamiltonian.

The aim of our work is to extend this result for relativistic Coulomb Green's operators, i.e. for the Coulomb Green's operator of the Klein–Gordon and of the second order Dirac equations. This later is equivalent to the conventional Dirac equation and seems to have several advantages. For details see Ref. [2] and references therein. The Coulomb Sturmian matrix elements of the second order Dirac equation has already been obtained by Hostler [2] via evaluating complicated contour integrals. Our derivation, however, is much simpler, it relies only on the Jacobi-matrix structure of the Hamiltonian, and the result obtained is also better suited for numerical calculations. In Ref. [2] the result appears in terms of Γ and hypergeometric functions, while our procedure results in an easily computable and analytically continuable continued fraction.

*Accepted for publication in Journal of Mathematical Physics.

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Factorization of spin-dependent Hamiltonians

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Isospectral quantum mechanical systems can be described in a variety of ways, such as factorization techniques, algebraic constructions and supersymmetry schemes. In a rather general class of problems the isospectral Hamiltonians are constructed as $H_1 = QR$, $H_2 = RQ$, where the Q and R operators are written in terms of differential operators and/or matrices. We have made a systematic survey of quantum mechanical systems admitting the above factorization scheme by writing the Q and R operators in the form

$$Q = \sigma \cdot (\mathbf{p} + \mathbf{a}(r)) + C(r) \ , \qquad R = \sigma \cdot (\mathbf{p} + \mathbf{b}(r)) + D(r) \ .$$

This construction recovers Hamiltonians containing the $\mathbf{p}^2$ term, so it naturally leads to Schrödinger-type equations.

Making further prescriptions for the properties of Q, R and the Hamiltonians various conditions follow for the general functions $\mathbf{a}(r)$, $\mathbf{b}(r)$, $C(r)$ and $D(r)$. Central potentials are obtained, for example, with the choice $\mathbf{a}(r) = f(r)\mathbf{r}$ and $\mathbf{b}(r) = g(r)\mathbf{r}$. A rather general class of problems arises from the $g(r) = -f(r)$, $C(r) = D(r) = 0$ choice: the Hamiltonians obtained this way have strictly non-negative energy eigenvalues, and are free from pseudoscalar and explicitly linear derivative terms:

$$H_1 = \mathbf{p}^2 + 2\mathrm{i}f\sigma \cdot \mathbf{L} + \mathrm{i}f'r + 3\mathrm{i}f - f^2r^2 \ , \quad H_2 = \mathbf{p}^2 - 2\mathrm{i}f\sigma \cdot \mathbf{L} - \mathrm{i}f'r - 3\mathrm{i}f - f^2r^2 \ .$$

This construction naturally introduces a spin-orbit term in the Hamiltonians.

Specifying $f(r)$ as $\mathrm{i}\omega/2$ and $\mathrm{i}c/r$ leads to analytically solvable problems built on the harmonic oscillator and the Coulomb potentials, respectively. The corresponding energy spectra exhibit large-scale degeneracy patterns [1]. In particular, in both cases H_1 has a zero-energy ground state, which is infinitely degenerate, while the corresponding level is missing from the spectrum of H_2. This feature is also shared by various quasi-exactly solvable problems, for which analytic solutions can be obtained for a part of the spectrum only.

We have also discussed the relation of this construction with know factorization techniques and supersymmerty schemes [1].

[1] G. Lévai and F. Cannata, to be published.

Subatomic Physics

A key reaction of the solar neutrino puzzle: ^{7}Be(p,γ)^{8}B

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The ^{7}Be(p,γ)^{8}B reaction determines the branching of the pp-chain III and thus the predicted flux of the high-energy neutrinos from the ^{8}B-decay[1]. The understanding of this reaction is hampered by the difficulty in producing either a ^{7}Be beam or a reliably produced and analysed ^{7}Be solid state target.

The target preparation involves hot chemistry and evaporation of the radioactive ^{7}Be material on a solid backing of selected low activity material. Finally the ^{7}Be atoms ($T_{1/2}$ = 53 d) are transformed into ^{7}Be oxide. The initial ^{7}Be mater ial is produced at the cyclotron of the ATOMKI via the ^{7}Li(p,n)^{7}Be reaction. In previous experiments this kind of target has proven to be very stable, even when subjected to intense ion beams [2]. The stoichiometry and the areal density of the target has to be measured via γ-ray activity, nuclear reaction methods, Rutherford backscattering and microbeam analysis.

Using this kind of radioactive ^{7}Be-target, the $\sigma(E)$ measurement via the direct detection of the capture γ-rays or of the residual ^{8}B nucleides is extremely difficult and has not yet been carried out. Instead, the cross section was derived from the β-delayed α-activity of ^{8}B, where the lowest data point is at $E_{\rm cm}$ = 117 keV [3], while the solar Gamow peak is at $E_{\rm cm}$ = 19 keV with a width of 9 keV.

In our experiment in Bochum (Germany), the target was mounted on a rotating arm. The target irradiated by the beam is moved in front of the particle detector in order to measure the β-delayed α-decay of ^{8}B and then transfered back to the beam position. This procedure was repeated many times. The particle detector must have a low intrinsic activity.

The preliminary measurements have been carried out and the first results are promising. However, the quality of the target has to be improved further. The work is in progress.

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Loss of ^{8}Li recoil nuclei in ^{7}Li(d,p)^{8}Li and implications on the ^{7}Be(p,γ)^{8}B cross section

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The absolute cross section $\sigma_{17}(E)$ of the ^{7}Be(p,γ)^{8}B reaction influences sensitively the calculated flux of high energy neutrinos from the sun. Due to its importance for the solar-neutrino-puzzle, the cross section $\sigma_{17}(E)$ should be known with adequate precision. In all direct measurements this cross section was determined from the yield of the of the ^{8}B recoils. The absolute cross section $\sigma_{27}(E)$ of the ^{7}Li(d,p)^{8}Li reaction is also of interest, since it serves for the normalisation of the ^{7}Be(p,γ)^{8}B cross section. The ^{8}Li recoils exhibit similar kinematics in their β-delayed α-decay via ^{8}Be.

The experiments were carried out [1] with the 4 MV Dynamitron tandem accelerator at the Ruhr-Universität Bochum with a deuteron beam at $E_d = 0.4$ to 1.8 MeV. The experimental setup was the same as used for the ^{7}Be(p,γ)^{8}B measurements [2].

The loss of ^{8}Li recoil nuclei in ^{7}Li(d,p)^{8}Li has been measured using different backings and ^{7}LiF target thicknesses. The results confirm essentially the recent TRIM calculations. The losses are large (about 13%) for the combination of thin targets and heavy backings at $E_d = 0.80$ MeV and increase with decreasing deuteron energy. Unfortunately the exact ^{7}Be target composition of the earlier individual ^{7}Be(p,γ)^{8}B experiments are not known hampering thus a precise correction of reported values for the recoil effect. One might suggest therefore that the reported absolute $\sigma_{17}(E)$ values should include an additional systematic uncertainty of the order of 15%. The energy dependence of $\sigma_{17}(E)$ might be affected by the recoil losses even stronger at low energies and might thus influence severely the extrapolation of the data to the solar Gamow energy E_0 producing a nonnegligible influence on the solar-neutrino-puzzle. In order to reach the goal of 5% precision, new measurements of $\sigma_{17}(E)$ must include quantitative in-situ determination of the ^{8}B losses nearly at each energy.

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Excitation of isovector spin-dipole resonance and neutron skin of nuclei

A. Krasznahorkay, M. Fujiwara,* P. van Aarle,* H. Akimune,* I. Daito,* H. Fujimura,* Y. Fujita,* M.N. Harakeh,* T. Inomata,* J. Jänecke,* S. Nakayama,* A. Tamii,* M. Tanaka,* H. Toyokawa,* W. Uijen* and M. Yosoi*

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+ 12 other institutes

Radioactive nuclear beams have opened up new research opportunities because a new generation of bombarding particles has become available to study the structure of nuclei far from stability. Nuclear properties which are not observed in nuclei near the stability line like the neutron halo and the neutron skin have been revealed [1].

In our previous work on inelastic alpha scattering, the giant dipole resonance (GDR) excitation was used to extract the neutron-skin thickness of nuclei [2]. The cross section of this process depends strongly on $\Delta R_{\rm np}/R$, the radial difference of the neutron and proton densities.

Another possible tool for extracting the neutron-skin thickness, the excitation of the spin-dipole resonance (SDR), is proposed in the present work. The total $L = 1$ strength of the SDR is sensitive to the neutron-skin thickness [3]. In order to demonstrate the neutron-skin thickness dependence of the SDR strength, the even-even Sn targets were chosen. It has also been shown that intermediate energy (^{3}He,t) reactions can be effectively used to excite the spin-isospin resonance in nuclei [4]. The experiment was carried out at the Research Center for Nuclear Physics (RCNP), Osaka University at $E_{^3\rm He} = 450$ MeV. The energy of the tritons was measured with the magnetic spectrometer "Grand Raiden" [5]. The spectrometer was set at 0° with an opening angle of ±20 mrad horizontally and ±20 mrad vertically. The ejectile tritons were detected with two multi-wire drift chambers (MWDC's) placed along the focal plane.

The spectra taken at 0° and 1° using the reactions ^{114,116,118,120,122,124}Sn(^{3}He,t) were fitted simultaneously with the same parameters for the excitation energies and the widths of the SDR, Gamow–Teller resonance (GTR) and isobaric analog state (IAS). Although the fitting procedure has been done very carefully, the assumption of the Breit–Wigner shapes for describing the shapes of the different resonances may not be justified. In order to overcome this dilemma, a "spectrum-difference" method has been used to check the intensity of the SDR.

The difference of the neutron–proton rms radii has been determined by using the following simple expression derived from sum-rules [6].

$$\langle r^2\rangle_n^{1/2} - \langle r^2\rangle_p^{1/2} = \frac{\alpha\sigma_{exp} - (N-Z)\langle r^2\rangle_p - 2.80A}{2N\langle r^2\rangle_p^{1/2}}\,, \qquad (1)$$

where α is a normalization constant determined by accepting the theoretical result

of Ref. [9] for the difference $\langle r^2 \rangle_n^{1/2} - \langle r^2 \rangle_p^{1/2}$ in ^{120}Sn and the values of $\langle r^2 \rangle_p^{1/2}$ were taken from Ref. [7]. The results obtained are shown in Fig. 1.

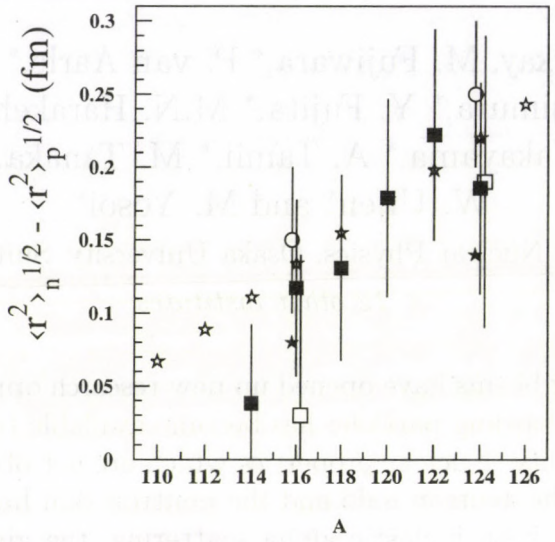

Fig. 1: The difference of the neutron and proton root-mean-square radii as a function of the mass number of the Sn isotopes. The full squares with error bars show the present results. The previous experimental results measured in (p,p′) reaction [8] and by using the GDR method [2] are shown as open circles and squares with error bars, respectively. The open and full stars show the theoretical results of Angeli *et al.* [9] and Dechargé *et al.* [10], respectively.

With the above experiment we have demonstrated that the precise determination of the SDR cross sections provides a novel method to determine the difference of the rms radii of the neutron and proton distributions. This method can also be used in (p,n) reactions in inverse kinematics with radioactive ion beams using, e.g. polyethylene targets offering a new possibility for the determination of the neutron-skin thicknesses in radioactive nuclear beams.

One of us (A.K.) would like to express gratitude to the RCNP COE program for the financial support of his stay at RCNP.

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Absolute measurement of the $5/2^+$ resonance of ^{36}Ar(p,γ)^{37}K at $E_p = 918$ keV

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[2] Institut für Kernphysik, Techn. Univ. Wien, Austria

The low energy resonance of the ^{36}Ar(p,γ)^{37}K reaction at $E_p = 321$ keV is important for nuclear astrophysics because this resonance determines the the reaction rate of the ^{36}Ar(p,γ)^{37}K reaction in hot and explosive hydrogen burning. However, this resonance strength has been determined relative to the $5/2^+$ resonance at $E_p = 918$ keV (corresponding to $E_x = 2750.3$ keV in ^{37}K) which was measured absolutely by Goosman and Kavanagh in 1967 [1, 2]. In our work this resonance strength was remeasured [3]. The new result is consistent with previous results. Since this resonance was used in several experiments as a calibration standard, our experimental uncertainties were carefully analyzed and the work was compared to earlier experiments. The new result is consistent with previous ones.

The experiment was performed at the Van-de-Graaff accelerator, ATOMKI, Debrecen. The ^{36}Ar targets consisted of implanted ^{36}Ar into tantalum backings. The targets were produced at the isotope separator of the University of Helsinki. The number of implanted argon atoms was measured after the (p,γ) experiment by proton-induced X-ray emission (PIXE) at the PIXE setup of ATOMKI. The γ rays from the ^{36}Ar(p,γ)^{37}K reaction were detected in a high-purity germanium (HPGe) detector with 40% relative efficiency.

The resonance strength was determined from two different procedures: (i) from the shape and the amplitude of the yield curve and (ii) from the maximum of the yield curve.

Our final result for the resonance strength is the weighted average of both determinations taking into account that the uncertainties of both determinations are mainly independent: $\omega\gamma = (258 \pm 24)$ meV. This result is slightly higher than the previous result: $\omega\gamma = (208 \pm 30)$ meV [1, 2].

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Production of ^{44}Ti at the RIKEN Radioactive Ion Beam Facility

Zs. Fülöp, E. Somorjai, M. Golovkov,[1] T. Kato,[1] K. Kimura,[2] H. Kudo,[3] Y. Mochizuki,[1] H. Otsu,[1] A. Ozawa,[1] H. Petrascu,[1] H. Sakurai,[1] T. Suzuki,[3] I. Tanihata,[1] Y. Wakasaya,[1] K. Yoshida[1] and R.N. Boyd[4]

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In addition to studies of exotic nuclei, radioactive ion beams also proved to be a useful tool to determine the half-life of isotopes with $T_{1/2}$ in the region of 100 years [1]. The observation of the $E_\gamma = 1157$ keV line — characteristic to the decay of ^{44}Ti — in the Cassiopeia A [2] supernova called for the accurate determination of the half-life of ^{44}Ti ($T_{1/2}$ ~60y). In lack of reliable half-life data the initial amount of ^{44}Ti ejected by the Cassiopeia A cannot be given with a precision needed to test the various supernova models. In fact, since ^{44}Ti can also be produced in meteorites by means of cosmic-ray interactions, the half-life of ^{44}Ti also has impact on the galactic cosmic ray flux and its modulation by solar activity [3]. The abundance of ^{44}Ca in the solar system also may depend on the half-life of ^{44}Ti [4].

In our study we have tested the feasibility of the RIKEN RIPS radioactive beam facility [5] to produce ^{44}Ti beam. During our test measurement the ^{44}Ti was produced by fragmentation of 2 particle nA, 90 A MeV ^{46}Ti on 200 mg/cm^2 thick Beryllium target. The projectile fragments were separated by the RIPS fragment separator with a 221 mg/cm^2 thick Aluminium wedge degrader placed between the two analyser dipole magnets. Through appropriate settings of the fragment separator we achieved high secondary beam intensity (2×10^5/s) with high ratio of ^{44}Ti nuclei without implanting isotopes contributing to the gamma activity of the sample. Also, we kept the content of ^{42}Sc (same A/Z as ^{44}Ti, i.e. time of flight determination is not enough to separate this isotope from ^{44}Ti) to minimum.

The γ-activity of the implanted sample has been investigated by a well-shielded high-efficiency HPGE gamma detector. There is no contamination observed in the gamma spectra of the irradiated sample with the exception of the ^{44}Sc^m isomeric state decay ($E_\gamma = 271$ keV, $T_{1/2} = 58.6$ h).

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Terminating high-spin bands in $A \approx 100$ Ru, Rh and Pd nuclei

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In many nuclei collective and single particle excitation modes are competing with each other in the experimentally observable spin region. Such a competition is involved in the phenomenon of band termination when the rotating nucleus changes its shape from collective deformed to near-spherical or non-collective deformed. In these latter shapes the nuclear spin is built up solely from the spin contributions of the individual nucleons and consequently it has a maximum value for a given single-particle configuration, at which spin the corresponding band terminates. In the $A \approx 100$ mass region it was predicted [1] that in some Ru and Pd nuclei terminating high-spin bands should exist and remain yrast from the low-spin (prolate) rotational states up to the termination.

An experiment has been carried out with the aim to search for terminating bands in ^{102}Pd using the ^{70}Zn + ^{36}S reaction at a bombarding energy of 130 MeV. The beam was provided by the Vivitron accelerator at IReS, Strasbourg. The target was made of two stacked self-supporting foils of isotopically enriched Zn. γ-rays were detected with the EUROGAM2 spectrometer. Level schemes have been deduced using the RADWARE program package as well as total and gated $E_{\gamma 1} - E_{\gamma 2} - E_{\gamma 3}$ coincidence cubes that have been constructed from the measured events. The spins and parities of levels were deduced from DCO ratios and linear polarisation values.

We have obtained several terminating bands in ^{102}Pd which were interpreted as valence space and core excited configurations [2]. The success of this analysis motivated a systematic search for terminating bands in the neighbouring ^{98,99,100}Ru ^{101,102}Rh and ^{100,101,103}Pd nuclei as well.

The high-spin bands found in the studied nuclei have been interpreted using Nilsson–Strutinsky cranking calculations. On the basis of the good agreement ob-

tained between the calculated and observed level energies as a function of spin (for spin larger than $\sim (15-20)\hbar$), many high-spin bands have been assigned as terminating configurations. Most of them were observed up to the predicted terminating state [3, 4, 5]. These assignments enabled us to identify the single-particle configurations of the observed bands in terms of the number of protons in the $g_{9/2}$ and $h_{11/2}$ orbitals and in the low-j, $N = 3$ orbitals, as well as the number of neutrons in the $h_{11/2}$ and in the $d_{5/2}g_{7/2}$ orbitals.

Negative parity $\pi(g_{9/2})^p\nu(d_{5/2}g_{7/2})^{n-1}(h_{11/2})^1$ configuration and positive parity $\pi(g_{9/2})^p\nu(d_{5/2}g_{7/2})^{n-2}(h_{11/2})^2$ configuration have been found in each studied nucleus (p and n denotes, respectively, the number of valence protons and neutrons in the given nucleus with respect to the ^{90}Zr core). These configurations are systematically the yrast ones at the highest observed spins. In the Ru nuclei the $\pi(N = 3)^{-1}(g_{9/2})^{p+1}\nu(d_{5/2}g_{7/2})^n$ and the $\pi(N = 3)^{-1}(g_{9/2})^{p+1}\nu(d_{5/2}g_{7/2})^{n-1}(h_{11/2})^1$ configurations are also close to yrast at the highest spins and even become yrast at lower spins. In the Rh nuclei these configurations are predicted to lie higher in energy. In agreement with the predictions, they were not observed in ^{101}Rh, while a band was tentatively assigned as $\pi(N = 3)^{-1}(g_{9/2})^6\nu(d_{5/2}g_{7/2})^6(h_{11/2})^1$ in the ^{102}Rh, but it was not observed up to the predicted termination. In the Pd nuclei these configurations are predicted to lie high above yrast and were not observed experimentally. In the ^{102,103}Pd nuclei, however, we have observed high-energy bands that were tentatively assigned as core-exited configurations in which one proton from the $N = 3$ shell is lifted to the $h_{11/2}$ shell or two protons are lifted to higher shells.

As most of the $\pi(g_{9/2})^p\nu(d_{5/2}g_{7/2})^{n-1}(h_{11/2})^1$ and $\pi(g_{9/2})^p\nu(d_{5/2}g_{7/2})^{n-2}(h_{11/2})^2$ bands were observed up to the terminating state there is a possibility to study in these states the filling order of the different Nilsson orbitals originating from the $d_{5/2}g_{7/2}$ subshell as summarized in the table.

k	Terminating state	Spin contribution	Increment	Occupied orbital
3	^{98}Ru: 25^-; ^{99}Ru: $59/2^+$ ^{100}Pd: 25^-	7.5 $\hbar$	3/2 $\hbar$	$d_{5/2}$ Ω=3/2
4	^{99}Ru: $55/2^-$; ^{100}Ru: 32^+ ^{101}Rh: $65/2^+$ ^{102}Pd: 32^+	10 $\hbar$	5/2 $\hbar$	$g_{7/2}$ Ω=5/2
5	^{100}Ru: 28^- ^{101}Rh: $57/2^-$; ^{102}Rh: 33^+ ^{102}Pd: 28^-; ^{103}Pd: $65/2^+$	10.5 $\hbar$	1/2 $\hbar$	$d_{5/2}$ Ω=1/2

It shows the spin contribution of the neutrons in this subshell to the total terminating spin as a function of the number (k) of these neutrons. The spin increment caused by adding the last neutron is deduced from the obtained spin contribution and the orbital occupied by this neutron is proposed. It has been found that the obtained filling order, and that it does not depend on the actual nucleus or configuration is in agreement with the Nilsson cranking model predictions.

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Study of the proton drip line nucleus ^{106}Sb

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As a continuation of our study of the nuclei far from the stability line in the $A = 100$ mass region the yrast states of ^{106}Sb, the last proton stable Sb nucleus, were investigated via the ^{54}Fe(^{58}Ni,αpn) and ^{50}Cr(^{58}Ni,pn) reactions. On the basis of $\gamma\gamma$-coincidence analysis, the level scheme shown in Fig. 1 was proposed. It contains a new isomeric state with $t_{1/2} = 232(21)$ ns half live at 103 keV excitation energy. The multipolarities of the observed transitions were determined by means of angular distribution ratios. The spins of the states were assigned assuming that all observed transitions are stretched and that the spins increase with increasing excitation energy. The experimental results were compared with a shell model calculation performed in a limited model space using MSDI effective interaction. The low lying states could be assigned to the $\pi d_{5/2}\nu g_{7/2}$ and $\pi d_{5/2}\nu d_{5/2}$ configurations, while the higher lying states are expected to arise from broken pair configurations. The calculated yrast states are compared with the experimental data in Fig. 1.

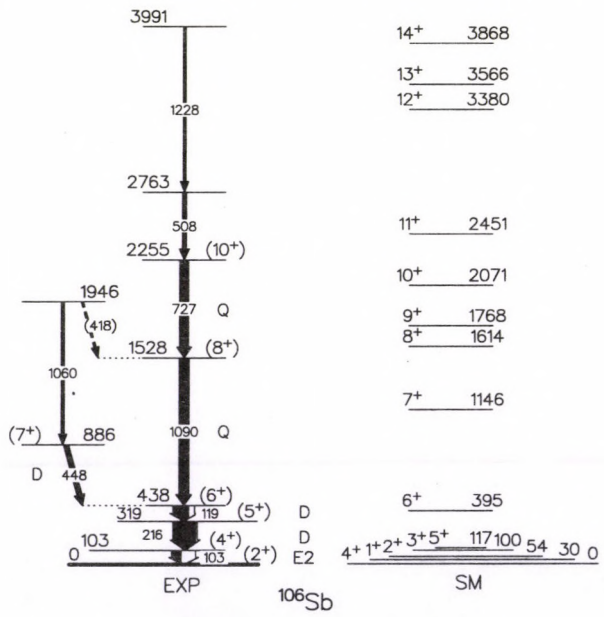

Fig. 1: Experimental and theoretical level schemes of ^{106}Sb. The multipolarities of the transitions are also given. D marks dipole transitions, and Q marks quadrupole transitions. All the spin values are tentative. They are given relative to the assumed ground state spin $I^\pi = 2^+$. The theoretical level scheme is calculated in the framework of the shell model.

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Spectroscopy of the neutron deficient ^{108}Te

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Structure of the neutron deficient nucleus ^{108}Te was reinvestigated via the ^{54}Fe(^{58}Ni,$2p2n$) reaction in a pre-Euroball experiment at the Niels Bohr Institute. A detector system consisting of 4 high efficiency Euroball cluster detectors, a Si charged-particle detector ball and a neutron multiplicity filter was used to detect the emitted particles and γ rays. The level scheme of ^{108}Te, shown in Fig. 1 a), was extended up to 4.7 MeV excitation energy on the basis of the new coincidence relations. The multipolarities of the transitions were deduced from their angular distribution ratios. The DCO ratios were determined from particle gated γ-ray spectra, obtained from three different rings of the detectors lying approximately at the same angles relative to the beam direction. According to the two-dimensional plot shown in Fig. 1 b), stretched quadrupole character was assigned to the 625, 664, 758, 803, 897 keV γ rays, while the 662, 830, 940, 1038 keV transitions were accepted as stretched dipole ones.

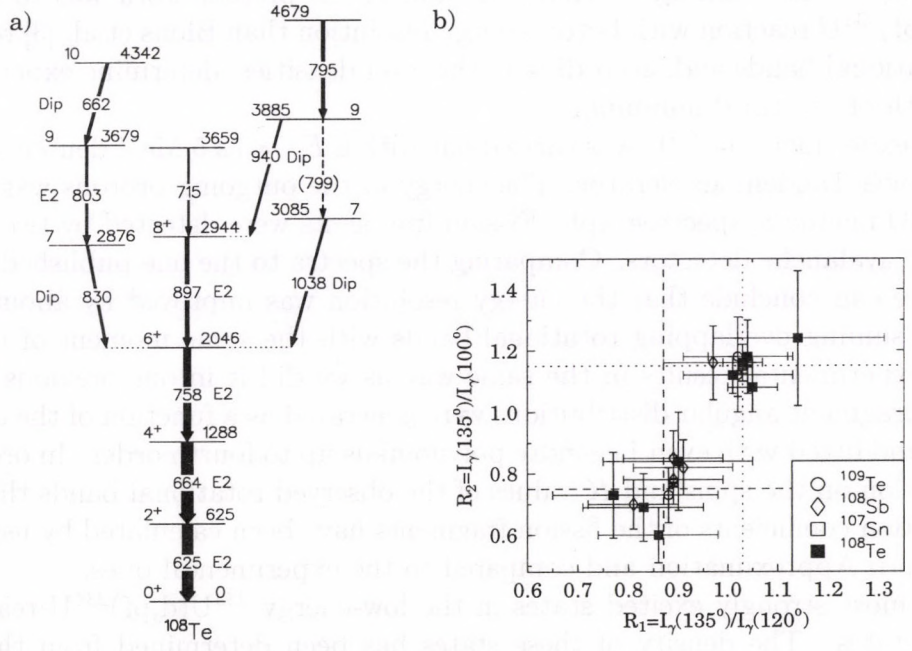

Fig. 1: a) Proposed experimental level scheme of $^{108}_{52}$Te$_{56}$. The multipolarities of the transitions are also given. *Dip* marks dipole transitions. b) Two-dimensional plot of the angular distribution ratios. The two kinds of dashed lines indicate the weighted average values for dipole and quadrupole transitions obtained from DCO ratios of γ rays with known multipolarities.

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On the excitation energy of the ground state in the third minimum of ^{234}U

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In the actinide region a third minimum in the potential energy was predicted already more than twenty years ago by Möller et al. However, according to the recent calculations, the depth is predicted to be much larger ($\Delta E \approx 3$ MeV) than it was believed earlier [1].

In our previous work [2] we reanalyzed the fission resonances measured by Blons et al. [3] concerning ^{234}U and showed that the unresolved peaks around $E^* = 4.9$ MeV could be interpreted as being the consequences of HD states lying in the third well of the potential barrier. The aim of the present work was to study the ^{233}U(d,pf)^{234}U reaction with better energy resolution than Blons et al. [3] resolve the HD rotational bands and, according to the level densities, determine experimentally the depth of the third minimum.

The experiment on ^{234}U was carried out with a $E_d = 12.5$ MeV deuteron beam of the Munich Tandem accelerator. The energy of the outgoing protons was analyzed by a Q3D magnetic spectrograph. Fission fragments were detected by two position-sensitive avalanche detectors. Comparing the spectra to the one published by Blons et al., we can conclude that the energy resolution was improved by about a factor of 3. Assuming overlapping rotational bands with the same moment of inertia we fit the experimental results in the same way as we did it in our previous work [2]. Fission-fragment angular distributions were generated as a function of the excitation energy and fitted with even Legendre polynomials up to fourth order. In order to get information on the spins and K values of the observed rotational bands the angular distribution coefficients of the fission fragments have been calculated by using Plane Wave Born Approximation and compared to the experimental ones.

The most strongly excited states in the low-energy ^{233}U(d,pf)^{234}U reaction are the 3^- states. The density of these states has been determined from the experimental data and compared to the results obtained with the back-shifted Fermi-gas description with parameters determined by Rauscher et al. [4] in order to determine the depth of the third well [5]. The obtained energy of the ground state in the third minimum has been deduced to be $E_{\mathrm{III}} = 2.35 \pm 0.3$ MeV, which value agrees well with the predicted one [1].

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Spontaneous fission and multicluster configurations

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Due to the multiple gamma-coincidence experiments [1] these days we can get very detailed information on spontaneous fission. This situation calls for a description which includes also the microscopic aspects of the nuclear structure. Recently we have proposed an approach of this kind, based on the cluster picture and on the effective U(3) symmetry of the participating nuclei [2]. So far these considerations were applied for the description of the structural aspects of the bimodal fission, i.e. zero-neutron channels exclusively [2]. Here we generalize the method for fission channels including (several) neutrons as well; i.e. from the structure viewpoint we consider multicluster configurations.

As an example we take the Ba-Mo fissions of the ^{252}Cf nucleus. The effective U(3) symmetries of the parent and daughter nuclei are determined as before [2]. The neutrons are considered to be evaporated from the heavier fragments, according to the indication of the experiments [1]. The relative preference of the different channels are calculated on the basis of the similarities between the effective U(3) symmetry of the parent nucleus and those of the cluster configurations [2], the latter ones include now both the possible two-cluster and multicluster fragmentations.

What we find is the following. For a specific Mo isotope the more neutrons are evaporated, the more favoured is the fission. However, the relative preference of the Ba isotopes, when they are summed-up from various neutron channels, show a single-bumped distribution, like their relative yieds in the experiment, except that the maximum shows up at slightly lighter isotopes. We stress here, that the result is a consequence of the microscopic structure, i.e. independent from the penetrability effects, which are obviously very important as well.

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Reaction anomalies as signatures of structure effects?

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The clusterization of the constituent nucleons in the atomic nuclei is a very interesting phenomenon of nuclear structure. The most direct experimental evidences for the clusterization effect are provided by the spontaneous emission of composite light nuclei, like e.g. ^{4}He, ^{14}C, etc either i) from the ground states of heavy nuclei, or ii) from the highly excited (resonant) states of light nuclei. Structure theories predict clusterization also in the ground state region of light nuclei, however, the direct cluster emission is energetically forbidden from these states. Therefore, their experimental observation is a highly nontrivial and complicated task.

Recent scattering studies of heavy ions [1, 2] show very different results, depending on the fact whether or not the target nucleus is expected to have an (exotic) cluster configuration defined by the projectile. Experiments were carried out for ^{12}C scattering on ^{24}Mg on the one hand side, and on ^{28}Si on the other. The ground state of the ^{24}Mg nucleus is expected to have a core-plus-^{12}C cluster configuration [3, 4], while that of the ^{28}Si is not [3, 5]. Both the primer experimental data and their description in terms of the potential picture have fairly different characteristics in the two cases. This means different oscillatory pattern of the angular distributions, and weak or strong absorption of the corresponding potentials, as well as the existence or nonexistence of dispersive potential, etc. [6]. The explanation for the different behaviour of the two systems may very well be related to the appearance or nonappearance of the interference between the elastic scattering and elastic transfer of the ^{12}C nucleus.

If the reaction anomaly is really due to the cluster effect of the target nucleus, then this phenomenon, i.e. the elastic transfer effect can serve as a very nice and powerful tool for the search of (exotic) cluster effects in the ground states of light and medium heavy nuclei. Further experimental and theoretical investigations are needed in order to clarify the situation.

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Dynamic symmetry breaking in the SACM: The role of the internal cluster structure

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The semimicroscopic algebraic cluster model (SACM) [1] utilizes group theoretical techniques to formulate the dynamics of cluster systems and this fact reduces the complexity of calculations considerably. In particular, the dynamic group and its relevant subgroups for an even-even core+α-particle type cluster system are

$$\mathrm{SU}_C(3) \times \mathrm{U}_R(4) \supset \mathrm{SU}_C(3) \times \mathrm{SU}_R(3) \supset \mathrm{SU}(3) \supset \mathrm{O}(3) \ .$$

In the dynamic symmetry approximation the Hamiltonian consists exclusively of terms related to (Casimir) invariants of the groups appearing in the above group chain. This approach has the advantages that it results in realistic energy spectra in many applications, and the energy eigenvalues can still be determined analytically.

We considered a departure from this crude approximation by allowing symmetry breaking terms in the Hamiltonian. The simplest such terms are those identified with Casimir invariants of the *subgroups* of $\mathrm{SU}_C(3)$ and $\mathrm{U}_R(4)$ assigned to the internal structure (C) and the relative motion (R) of the clusters, respectively. In the first such application we considered Casimir invariant of the $\mathrm{O}_C(3)$ group, which is the square of the orbital angular momentum operator assigned to the core nucleus. This operator lifts the degeneracy of the core states belonging to the same representation of the $\mathrm{SU}_C(3)$ group.

As the first test example we studied the ^{12}C+α system, where the internal structure of the ^{12}C nucleus was represented by the $J^\pi = 0_1^+$, 2_1^+ and 4_1^+ states assigned to the $(\lambda_C, \mu_C) = (0, 4)$ $\mathrm{SU}_C(3)$ representation. These states form a rotational band built on the 0_1^+ ground state, a pattern which can be simulated by the introduction of the L_C^2 symmetry-breaking term. The effect of this term resulted in the following developments. *i)* The composition of the low-lying ^{16}O states has changed in the sense that the contribution of the $J^\pi = 2^+$ and 4^+ states of the ^{12}C cluster was reduced. *ii)* The degeneracy of those ^{16}O levels which have the same J^π value and belong to the same SU(3) multiplets has been lifted. This is a possible realization of the K-band splitting, which we have taken into account previously in the case of unbroken dynamical symmetry by a phenomenologic K^2 operator. Now the role of this somewhat ad hoc term can be taken over by the L_C^2 term, which has more solid foundations in our model.

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Unbound states by analytic continuation in the coupling constant

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Nuclei have a large number of discrete unbound states (viz. resonances and virtual states). Such unbound states may be put on the same footing as bound states by the concept of the S-matrix pole. The unbound-state poles may be located by an analytic continuation in the "coupling constant" [1], i.e. in a strength parameter of the potential. In this way the solution of the unbound-state problem is substituted by solutions of a number of bound-state problems, and the pole position is determined by extrapolation. In the present work [2] the objective was to study controversial unbound states of the most typical two-cluster and three-cluster nuclei. In this way we broadened the applicability of the analytic continuation method, and explored the nature of the states considered.

The general results are the following. First, we found that, with a slight generalization, the method performs well for $l = 0$ states as well, where the pole trajectory passes through the virtual-state region. Second, we have demonstrated that the three-body resonances of systems that interact via purely attractive forces behave like two-body resonances within a potential barrier. In this way we confirmed the appearance of an effective three-body barrier. Third, we have demonstrated that the analytic continuation is reliable even if the two-body thresholds, as functions of the coupling constant, cross the three-body threshold.

Turning to more specific results, the position of the $1/2^-$ resonance of the α+nucleon system has been confirmed to be much lower than obtained in conventional phenomenological analyses. For the $1/2^+$ states of ^{5}He and ^{5}Li, the long-standing contradiction between the bound-state and unbound-state methods is now resolved. The $1/2^+$ states do exist but lie very far from where they were predicted to be by the bound-state methods.

For the three-cluster systems ^{9}Be=α+α+n and ^{9}B=α+α+p, the $1/2^+$ states have been found.

From the pole trajectories it is apparent that a ^{8}Be+nucleon structure is more prominent in ^{9}Be than in ^{9}B. The agreement with experiment is very satisfactory, apart from the widths of the $1/2^+$ states.

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Structure of ^{6}He with an extended three-cluster model

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The standard cluster model of the neutron-halo nucleus ^{6}He [1] assumes an α+n+n structure. The aim of this work [2] has been to examine effect of the dynamical breakup of the α-particle in ^{6}He. The extended model used treats the α-particle as a $3N$+N two-cluster system. As an alternative model, a t+t component was added to the α+n+n component. The breakup and the t+t channel have much the same effect: they deepen the intercluster binding substantially and enhance the probability of the t+t component, especially in the asymptotic region. When the two improvements were combined, no further substantial change was found.

These results can be explained as follows. The simple $(0s)^4$ wave function used to represent the α-cluster in the pure α+n+n model has a too rapidly decreasing tail. The two halo neutrons can form a triton with a proton of the α-cluster in a region where the distribution of the halo neutrons overlaps with that of the α core. Since the halo neutrons fill a large region around the core, the spatial extension of this overlap is determined by the tail of the core, and that is why the pure α+n+n model has a tendency to underestimate the t+t probability for well-separated triton clusters. This is remedied by the inclusion of the t+t channel. On the other hand, the $3N$+N model of the α-particle produces a realistic tail and also reproduces the second 0^+ state, which guarantees that in the [$3N$+N]+n+n model of ^{6}He the dynamical core breakup is also taken into account realistically. Thus the $3N$+N treatment of the core allows for more or less the same effect as the t+t channel, and no wonder that both the binding energies and the t+t probabilities are similar. That is why, when added to the [$3N$+N]+n+n model, the extra effect of the t+t channel is not appreciable. Thus the t+t effect is rather a core breakup effect than an exotic clustering effect. Almost all t+t clustering is already included in a [$3N$+N]+n+n model. The [$3N$+N]+n+n model is consistent with a core plus halo picture.

We have learned that the precise value of the binding energy as well as other specific properties of a halo nucleus (the t+t clustering here) do require a realistic treatment of the core. The core breakup effect is appreciable even for the most tightly bound α core. This result suggests that a similar breakup effect must be much more pronounced in other neutron-halo nuclei, such as ^{11}Li, ^{11}Be and ^{10}Li.

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Mechanism of parity inversion in ^{11}Be with a microscopic cluster model

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It is well known that in the nucleus ^{11}Be the sequence of the $1/2^{\pm}$ states is inverted with respect to the shell-model prediction, and the $1/2^{+}$ ground state has a spatially extended density distribution attributable to a single neutron, that is, a single-neutron halo. The aim of this work is to understand the mechanism of this parity inversion. We describe the nucleus by a microscopic multicluster model with fully antisymmetrized 11-nucleon wave functions.

As a previous step, the nucleus ^{10}Be was studied in an α+α+n+n four-cluster model [1], and all states below the ^{9}Be+n threshold have been obtained. The bases were built up from generalized ("correlated") Gaussians optimized by random sampling ("stochastic variational method") [2]. By analogy, it would be straightforward to describe ^{11}Be as an α+α+n+n+n five-cluster system, but the number of configurations required would be extremely large. However, the optimized bases yieldig the ^{10}Be states provide a framework to describe ^{11}Be as a superposition of ^{10}Be+n configurations. This is a reasonable model, at least as a first step, since ^{11}Be is most likely to get partitioned as ^{10}Be+n. This is so because the ^{10}Be+n threshold is at 0.503 MeV, whereas the other partition channels, ^{9}Be+2n, ^{6}He+n+α, 2α+3n and ^{8}Li+t open above 7 MeV.

The effective two-nucleon interaction employed contains central and spin-oribit components as well as the Coulomb potential. We calculate the two ^{11}Be energy levels of spin-parity $1/2^{\pm}$ gradually extending the basis. After the simple shell-model limit, we employed a ^{10}Be+n basis with the realistic ground state of ^{10}Be, then we include the second 0^{+} state of ^{10}Be, the the first 2^{+} state and so on. We would like to see in which step the parity inversion occurs, which will tell us what kinds of core polarization are responsible for the parity inversion. Together with the inversion, we hope to be able to reproduce the neutron halo as well.

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Perturbative Quantum Chromodynamics

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Four-jet final states at LEP play an important role in testing quantum chromodynamics (QCD), the theory of strong interactions. This process makes possible the simultaneous precision measurement of the only free parameter of perturbative QCD (the strong coupling) and the color charge factors (the eigenvalues of the quadratic Casimir operator of the underlying Lie group), provided the next-to-leading order (NLO) perturbative prediction is known. During the last year, we finished the calculation of the higher order corrections to this process and performed extensive analysis of four-jet data measured at the Large Electron Positron Collider (LEP). We

- constructed a partonic Monte Carlo event generator that calculates NLO corrections to the group independent kinematical functions of four-jet observables in electron-positron annihilation [1];
- calculated the NLO corrections to four different four-jet event shape variables, the C parameter for $C > 0.75$, $T_{\min}$, the y_{34} distribution for Durham and Cambridge jet algorithms. We found large radiative corrections [1];
- calculated four-jet rates at NLO for two jet clustering algorithms the Durham and the Cambridge algorithms. We found that the NLO prediction when matched to the next-to-leading logarithmic approximation describes the data obtained at LEP very well [1, 2];
- based upon the fitting of the theoretical prediction of multijet rates to the jet rates measured by the ALEPH collaboration at LEP, we measured the strong coupling α_s with high precision and in agreement with the world average [2];
- calculated various angle distributions at NLO and found that the normalized angle distributions did not change their shape when going from leading to NLO [3]. This information can be utilized in measuring two fundamental constants, the color charges in QCD.

For further information we refer to our web site [4].

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Atomic Physics

A new electron spectrometer for electron–electron coincidence studies

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The low energy electron spectroscopy is widely used in different fields of physics. In the last 30 years different type of electrostatic analyzers were developed and applied to determine the energy distribution of electrons. The traditional way to measure the angular distributions of emitted photo- and Auger electrons is to rotate the analyzer around the interaction region. It is very time consuming and causes serious calibration problems. In order to overcome these problems and to perform electron–electron coincidence measurements, a new electron spectrometer (ESA-22) was developed in ATOMKI, Debrecen.

The ESA-22 is a modified version of ESA-21 analyzer [1], it consists of a spherical and a cylindrical part. Comparing with ESA-21 the main differences is that the focal ring can be set different diameters thus either a series of channel detectors can be used to detect the electrons at different angles or a position sensitive channel plate can be applied for simultaneous angular recording of electrons. Furthermore, the outer sphere and cylinder are cut into two parts so the spectrometer is capable to analyze two independent angularly resolved electron spectra (in the $0° - 180°$ region) at different energy regions, simultaneously. A special electronic control and data handling electronics and software was worked out to control the analyzer.

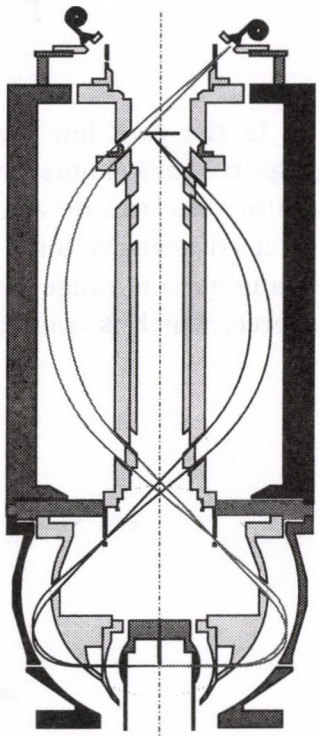

Fig. 1: The coincidence spectrometer ESA-22

Recently we are testing the performances of the analyzer. The achieved relative energy resolution is ($\Delta E/E = 4 \times 10^{-3}$, slit size 1.4 mm) which can be improved by using a retardation lens. The position sensitive detector not only records the angular distribution of electrons simultaneously but simultaneously detects the energy distribution, too in an energy region which is about 1% of the nominal energy value.

These advanced properties of the spectrometer offer a new way to investigate the angular distributions of electrons with high efficiency and to perform completely new coincidence measurements e.g. coincidence between photo- and Auger electrons, etc.

This work is supported by the Hungarian Science Research Found: OTKA No. T025325.

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Modification of the DANFYSIK 911A ion source for production of negative ions

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In the past few years we carried out ion–atom collision experiments involving negative ions, namely we have measured electron spectra following the single- and double electron detachment from H^- and He^- ions [1, 2]. For such measurements we need negative ion beams with current in the nA range. To produce negative ion beams in this range we replanned and modified the DANFYSIK 911A positive ion source, which is connected to the beamline of the VdG-1.

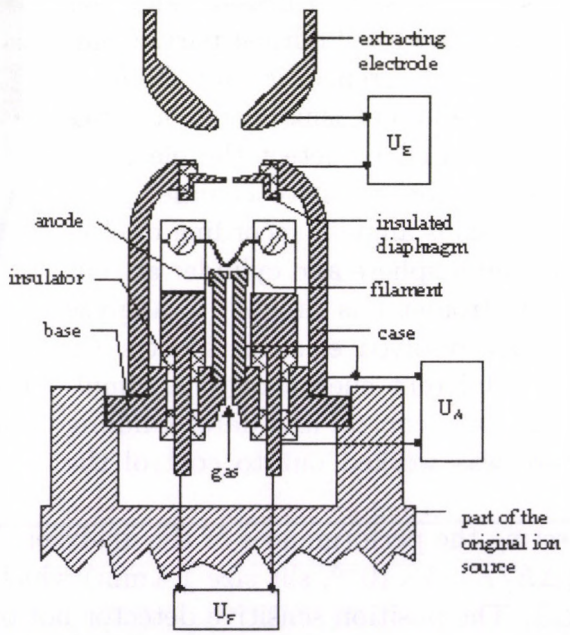

Fig. 1: The latest version of the modified ion source

During the modification several electrode configurations were made, here we present the latest version. The main parts of the new ion source are shown in Figure 1. In the development of the presented configuration we tried to confine the gas discharge to a minimal volume in order to achieve high plasma density, similarly as in the hollow-cathode ion source. For this purpose, the electrodes are arranged in that way that the discharge takes place close to the point of the largest gas pressure.

The new configuration is capable to produce both negative and positive ions. With this configuration we could produce 10 nA current of H^- ions and 10 μA proton current, for ion energy of 20 keV. These values were measured after a 90° analyzing magnet. Depending on the operational conditions, the lifetime of the filament is approximately 50 hours.

We emphasize that most factory-made components of the DANFISYK ion source remained unchanged. An advantage of the modified ion source is that simply changing the polarity of the extraction voltage one can produce also positive ions, with an intensity comparable with that of the original ion source. The new ion source is easy to dissemble, and operated in the same way as the old one.

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Multiple scattering of the emitted electrons in 150 keV/u C^+ + He, Ne, Ar collisions

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We measured the energy and angular distribution of electrons ejected in collisions of intermediate velocity C^+ ions with various noble gas targets. The experiments were carried out using the ESA-21 electron spectrometer at the beam line of the 5 MV Van de Graaff accelerator of ATOMKI.

In the experiments we observed an unusual increase of the cross section data at backward emission angles at energies corresponding approximately to the zero degree binary encounter (BE) energy $E_{BE} = \frac{1}{2}m_e(2v_p)^2$, where v_p is the projectile velocity. This behaviour is similar to that observed by Bechthold et al. [1] in collisions of high energy (5.9MeV/u) U^{29+} ions impacting on noble gas and molecular targets. Our explanation of this finding is also similar: the electron which first suffered a head-on binary collision with the projectile can be scattered later in all directions by the target nucleus. So it is possible to observe electrons with BE energy in other directions than those predicted by theories considering only one scattering of the active electron.

A classical trajectory Monte Carlo (CTMC) calculation, which takes into account the interaction of the active electron with both the projectile and target nuclei, supports our idea that the enhancement in the measured cross sections at electron energies $\frac{1}{2}m_e(nv_p)^2$, $(n = 1, 2, 3, \dots)$ can be associated with multiple scattering of the ejected electron on the heavier collision partners ("Fermi shuttle" mechanism [2]).

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Electron detachment above the H($n = 3$) threshold in H^-/rare gas collisions

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In this work we continued our studies on the collisional electron detachment from H^-. A particularly interesting feature of this fundamentally important process is the non-zero electron emission cross section at the H($n \geq 2$) thresholds, and consequently the occurrence of cusp in the energy spectrum of the detached electrons emitted in the direction of the incoming H^- beam. The cusp is produced in this case by dipolar force due to the collision-induced permanent electric dipole moment of the outgoing excited neutral H atom. Unlike the cusp produced by the Coulomb force, the properties of the dipolar cusp are not so well known. Another exciting motivation of these studies is to observe the theoretically predicted oscillation of the cross section [1].

Applying the method of zero-degree electron spectroscopy, in our former works we observed the dipolar cusp. In one of our experiments the electrons were detected in coincidence with the Lyman-α photons emitted from H($2p$) formed in the collisions of H^- with rare gas atoms at 4 keV impact energy [2]. In another experiment carried out for 85 keV H on He collisions, the contribution from the *double* electron detachment was eliminated detecting the electrons in coincidence with the outgoing neutral H atoms [3]. Unfortunately, in the obtained spectra the cusp was strongly masked by the peaks resulting from the decay of the $(2s2p)1P^o$ shape resonance lying above the H($n = 2$) threshold by 20 meV, therefore our cusp shape analysis was rather uncertain.

In the present experiment [4] we obtained the first results on coincident detection of 0° electrons with Balmer-α photons emitted in H^-/rare gas (Ne and Ar) atom collisions at 5 keV. With this experiment our goal was not only to determine the contribution of the H($n = 3$) + e^- detachment channel to the total dipolar cusp, but also to observe some shape resonances predicted to exist above H($n = 3$). For example, an interesting question is the existence of the $3D^o$ resonance that lies above the H($n = 3$) threshold by 1.5 meV according to the calculations of Bhatia and Ho [5]. The statistics of the obtained spectrum does not allow to establish the existence of such resonances. At the same time, the spectrum has a pronounced cusp shape, which means that this is the first observation of a "clean" dipolar cusp produced by the dipolar interaction between a low-energy (0–100 meV) electron and an excited ($n = 3$) hydrogen atom.

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Two- and three-body effects in the single ionization of Li by fast Ar^{18+} ions: Analogies with photoionization

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In the present work [1, 2], cross sections for single electron emission have been measured in collisions of 95 MeV/u Ar^{18+} projectiles with atomic Li for electron energies ranging from $3 - 1000$ eV and angles ranging from $25° - 155°$. Models based on the Born approximation are introduced to separate two- and three-body effects in the angular distributions of the ejected electrons. Both experiment and theory provide information about the separability of the two- and three body effects. The high projectile velocity and the use of the Li target are shown to be essential for the present analysis. The emission of the $1s$ electron is attributed mainly to three-body effects. The cross section for three-body collisions rapidly decreases with the electronic energy transfer involving a power law with an exponents of -3.5. Consequently, two-body effects dominate at high electron emission energies. Remarkably large contributions from two-body collisions were also observed for the low-energy emission of the $2s$ electrons. Demonstrating the analogy in ionization by photons and ions, the two- and three-body processes are associated with Compton scattering and photoabsorbtion, respectively.

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Single and double K-shell vacancy production in N^{7+} + Ti collisions

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Double vacancy production on the target K-shell of $Z \geq 10$ elements in the high impact energy region is a special case of double transitions, since here the role of electron correlation in the ground state is relatively small. Experimental study of such processes [1, 2] can be important for mapping the limits of the independent electron approximation (IEA) [2].

In this work [3] the vacancy production for the K-shell of titanium is studied. The velocities of the nitrogen projectiles are in the range of 12 to 30 a.u. Single- and double-vacancy production cross sections are calculated by means of nonperturbative classical models and classical trajectory Monte-Carlo methods within and beyond the independent electron approximation. Their ratio is compared with our published and preliminary reported experimental data [1]. In all cases, the measurements were performed by the method of X-ray spectroscopy, and the experimental double/single vacancy production ratios were determined from the measured intensity ratios of the K-hypersatellite and K-satellite line groups [1].

It is demonstrated that a simple method for accounting the final state electron–electron correlation in the double capture and the capture + ionization channels significantly improves the agreement between experiment and theory compared to the independent electron approximation.

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Phys. Rev. **A 33** 914 (1986)

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K. Tőkési et al., accepted for publication in Physica Scripta

Single ionization of Ar($2p$) by electron and antiproton impact

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One interesting and fundamental part of the collision physics is the understanding of the ionization process in ion-atom collisions. Study of the energy and angular distribution of the ejected electrons and scattered projectiles gives a basic information about the collision dynamics. Moreover, it is a sensitive test of various theories when the calculated cross sections are compared with each other. In recent decades both classical and quantum-mechanical theories have been extensively used to calculate the ionization cross sections of atoms and molecules for various projectiles. The main difficulty of calculations is that long-range character of Coulomb interactions have to be taken into account for accurate description of the cross sections. The classical trajectory Monte Carlo (CTMC) method has been quite successful in dealing with ionization processes in ion-atom collisions. The CTMC simulation is a non-perturbative method. One of its main advantages is that many-body interactions are exactly taken into account during the collision. Recently, the continuum-distorted-with-eikonal-initial-state (CDW-EIS) model has also successfully been applied to study the ionization process by proton impact.

In the present work CTMC theory is used to calculate ionization cross sections in electron and argon($2p$) atom collisions. The energies of the projectile electrons investigated were 300 eV, 500 eV and 2 keV. We also made model calculations using a CTMC and CDW-EIS approximation for the collisions between antiproton and argon(2p) atoms at impact energy having equivalent velocity to 2 keV electron. It is shown that the calculated cross sections are in relatively good agreement. The best agreement can be found at 90 degree.

Post-collision interaction at the beam direction

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Auger-line energy shift induced by post-collision interaction is studied for proton–argon collisions. Proton beam of 340 keV energy was obtained by 1.5 MV van de Graaff accelerator. The energy shift of the Ar L_3-$M_{2,3}M_{2,3}$ ($^1D_2$) is determined for the Auger electrons emitted at 0°, for four values of acceptance angles of the electron electrostatic spectrometer ESA-13 [1]: ±1.9°, ±1.1°, ±0.3° and ±0.2°. The ±0.3° and ±0.2° acceptance angles are obtained by use of the spectrometer in combination with an electrostatic lens, constructed specially for purposes of 0° electron spectroscopy [2].

The energy of the Auger electrons is 203.5 eV, which corresponds to the velocity slightly larger than the projectile velocity ($v_p/v_A = 0.95$). The experimentally obtained energy shift decreases with decrease of the acceptance angle, and becomes negative for acceptance angles ±0.3° and ±0.2°. The energy shift is also calculated for the same projectile energy, as a function of observation (acceptance) angle. Two theoretical methods are used: semi-classical [3], and quantum-mechanical [4, 5]. In the range of observation angles from ±0.1° to ±2.4° the semi-classical method gives that energy shift increases with decrease of the angle. The energy shift obtained by the quantum-mechanical method shows the same tendency as the experimentally obtained shift: it decreases with decrease of observation angle. However, the theory gives much higher values for the shift. The reason of the disagreement could be that the quantum-mechanical method does not take into account the post-collision effects of cusp electrons [7].

In our earlier works [6, 7], for the above matching velocity region ($v_p/v_A = 1.008$) in proton–neon collisions, we found a very good agreement between the values of energy shift calculated by the two theories and obtained by the experiment.

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Interaction of highly charged ions with microcapillaries

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Studies of the interactions of highly charged ions (HCI) with surfaces are of growing interest. From a number of experimental as well as theoretical studies [1, 2, 3, 4] the following picture of the HCI-surface interaction has emerged: When a highly charged ion approaches a solid surface, one or more electrons are resonantly captured at a characteristic distance d_c into Rydberg states of the projectile [1]. As a result, a multiply excited Rydberg atom with inner shell vacancies, a so-called hollow atom [2], is created.

Recently, a novel technique has been introduced to produce long-lived hollow atoms by interaction of highly charged ions with internal surfaces of microcapillaries [3, 4]. When an ion travels approximately parallel to the capillary axis, it is attracted toward the capillary wall by its self-image potential. Some projectiles will approach the capillary surface to within the critical distance d_c in the interior of the capillary. For projectiles hitting the capillary wall, destructive close collisions will erase the memory on the hollow-atom formation. These collisions closely resemble an ion-surface collision at grazing incidence leading to complete relaxation to the neutral ground state. However, if the location of the capture process is close enough to the exit hole of the capillary, hollow ions or atoms can escape the capillary before hitting the wall and are extracted into vacuum. It was found experimentally that a fraction of the extracted hollow ions from the capillary have extremely long lifetimes of the order of ns indicating the formation of spin-aligned states.

We analyze the transmission of HCI's through microcapillaries by a classical trajectory Monte Carlo simulation. The interaction of an HCI with the internal surface of the capillary is treated within the framework of linear response. For Ne^{q+} projectiles transmitted through a Ni capillary we find the fraction of transmitted projectiles which are partially neutralized in good agreement with recent experiments.

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The energy of the $K_{\alpha 5,6}$ x-ray satellites in Ca and Ti

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Above $Z = 19$ in the literature one can not find data on the energies of the $K_{\alpha 5}$ and $K_{\alpha 6}$ multiple ionization satellite lines (only an average $K_\alpha L^2$ energy was measured in most cases). We determined by different ways (graphically or by computer evaluation, using symmetric or asymmetric Voigt peak shapes) these satellite energies for Ca and Ti. The K x-ray spectra induced by 3.2 MeV He ion bombardment of Ca and Ti solid targets were recorded by means of a flat crystal spectrometer. An example spectrum is given as Fig. 1. The results of these measurements are given in Table 1.

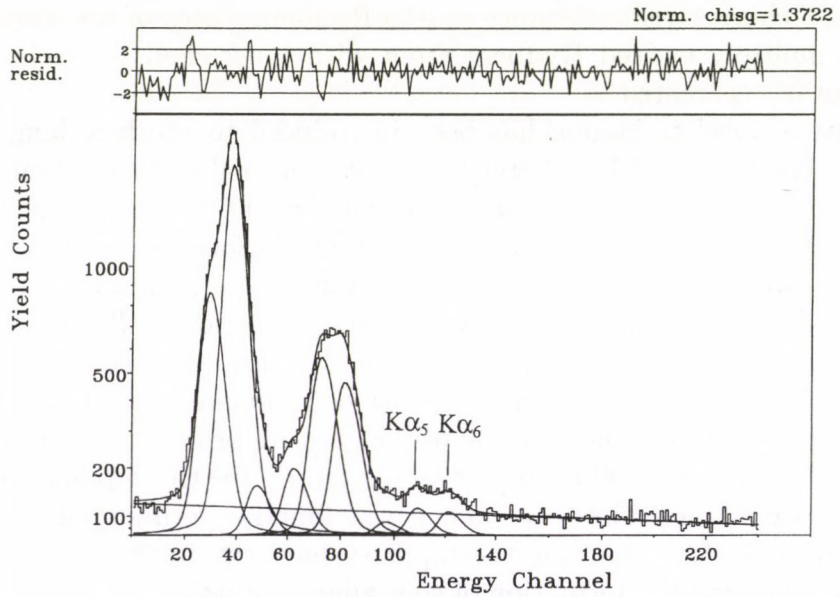

Fig. 1: The K_α x-ray spectrum of calcium

Table 1: Ti and Ca $K_{\alpha 5}$ and $K_{\alpha 6}$ satellite energies obtained by different evaluation methods

Sample	Origin and method	$K_{\alpha 5}$ [eV]	$K_{\alpha 6}$ [eV]
Ti	graphically	4562.0±3.0	4572.0±3.0
Ti	computer, sym.	4562.7±2.6	4570.1±1.0
Ti	computer, asym.	4561.6±3.0	4569.4±3.0
Ca.	computer, asym.	3732.2±3.0	3739.5±3.0

This work was presented at the FIAC-7 workshop as a poster [1], and a paper is in print in the proceedings of the workshop (Nucl. Instr. Meth. B). The work was supported by OTKA No. 3011 and T016636.

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An improved description of the multiple ionization $K_\alpha L^i$ satellite energy spacings

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In analytical works, and in basic atomic physics research one needs the data of the satellite lines, therefore we began to make a new compilation of the energy shifts of the $K_\alpha L^i M^m N^n ...$, $K_\beta L^i M^m N^n ...$ satellites, and $K^2_\alpha L^i M^m N^n ...$, $K^2_\beta L^i M^m N^n ...$ hypersatellites. A byproduct of this compilation is a test of the "classic" description of their energy spacings [1]. The larger database we have now made it possible, to obtain a better description. The old version gave equidistant spacing, using an average effective Z seen by the electrons: $\Delta(K_\alpha L) = 1.66 Z_L = 1.66(Z - 4.15)$, our version uses an effective Z changing with the number of spectator L vacancies:

$$\Delta E(K_\alpha L^i) = i \times 1.530[Z + (i-1)0.5 - 6.828], \qquad i = 1, 2, 3, ..., 7. \qquad (1)$$

Figure 1 compares the two descriptions graphically. Full account is given in Nucl. Instrum. Meth. B (Proc. of 8th PIXE conf., Lund, in print).

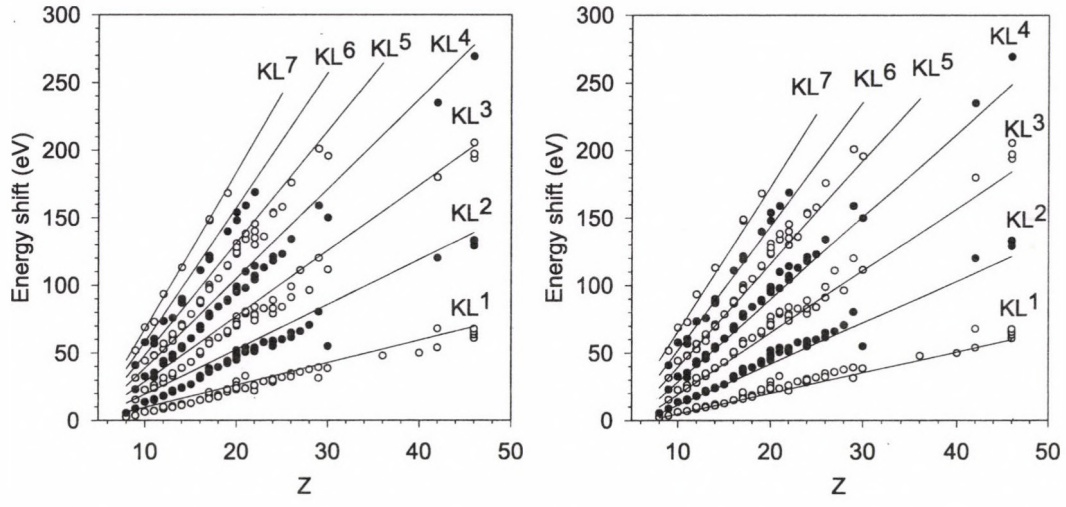

Fig. 1: The energy spacings of the $K_\alpha L$ satellites: a) after Burch et al. [1] and b) according to Eq. (1).

The work was supported by OTKA No. T016636.

* ORNL is managed by Lockheed Martin Energy Research Corporation under contract No. DE-AC05-96OR22464 with the U.S. Department of Energy.

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Materials Science and Analysis

KLL Auger transition energies of 3d transition metals Cu and Ni

L. Kövér, D. Varga, I. Cserny, J. Tóth and Zs. Kovács

KLL Auger spectra of Cu and Ni provide important information on the electronic structure of these metals. Earlier experimental studies on X-ray excited Cu KLL transitions [1], performed in a moderate vacuum, showed the presence of a satellite of the main $^1D_2$ line, identifying as electron loss contribution.

Cu and Ni KLL spectra were excited by Cu X-rays from Cu and Ni layers of 10 nm thickness. Following in situ Ar^+ sputter cleaning, the first high resolution measurements of these spectra were performed using a hemispherical electron spectrometer [2] with an energy resolution of 1.3 eV. In the high energy range the energy calibration of the spectra is based on the measurement of the Cu 2p lines excited by Cu Kα X-rays [3, 4]. The cleanness of sample surfaces was monitored by Al Kα excited XPS at a vacuum level better than 10^{-9} mbar. Evaluating the spectra, Voigtian lineshapes and a modified Tougaard-type background correction [5] were used.

From the measured KLL spectra the presence of the satellites is obvious. In our recent paper [6] we provide evidences that the satellites are similar in these metals and in their alloys and they can be attributed to 3d shake-up processes. Due to the good energy resolution, the $KL_3L_3(^3P_0)$ lines can also be identified in our spectra, in contrast with the previous measurement [1].

A part of the results of the evaluation of our spectra is summarized below. While the Cu KLL and Ni KLL relative energies are in a rather good agreement with both the earlier experimental [1] and theoretical [7, 8] data, the absolute Auger transition energies differ considerably.

Table 1: Cu and Ni KLL relative energies (eV)

Line	Present work (E)		[1] (E)	[7] (T)		[8] (T)
	Cu	Ni	Cu	Cu	Ni	Cu
Satellite	−11.4(0.7)	−7.1(1.5)	–	–	–	–
$KL_2L_2(^1S_0)$	−25.8(1.3)	−19.4(1.8)	−29(0.3)	−24.2	−21.6	−29
$KL_2L_3(^1D_2)$	0	0	0	0	0	0
$KL_3L_3(^3P_0)$	15.9(2.3)	16.3(2.3)	–	16.3	14.5	–
$KL_3L_3(^3P_2)$	28.5(0.6)	25.1(1.0)	28(0.3)	26.6	24.8	26
$E(KL_2L_3\ ^1D_2)$	7038.3(0.5)	6559.1(0.5)	7034.5(1.0)	7031.1	6542.1	–

The authors are indebted to Dr. G. Langer for the preparation of the thin film samples. This work was supported by the research projects OTKA T026514 (Hungarian) and ERBIC15CT960800 (EU).

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Rutherford backscattering measurement (RBS) of interdiffusion in amorphous Si/Ge multilayers

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Multilayers and superlattices are of considerable industrial interest because of their specific properties and many promising areas of applications, such as: electronic or optical applications [1], metallic multilayers for X-ray and UV mirrors [2], GMR and magnetic recording [3] etc. Thus investigation of the thermal stability and understanding of the factors controlling structural changes of these multilayers is very important for the interpretation of their operation and prediction of their lifetime.

The Si/Ge multilayer samples were prepared at room temperature by DC magnetron sputtering on Si(100) substrate. The modulation wavelength of the samples was 16.6 nm. The total thickness of the films was approximately 200 nm. For annealing the specimens were placed in Ar-atmospheric furnace, using 99.999% purity argon gas. In order to prevent the crystallisation, the annealing temperature was chosen definitely lower (between 400 °C and 480 °C) than the phase-transformation temperature of amorphous Si.

To study the thermal stability of Si/Ge amorphous multilayers Rutherford backscattering spectrometry (RBS) measurements with 1 MeV He^+ beam have been

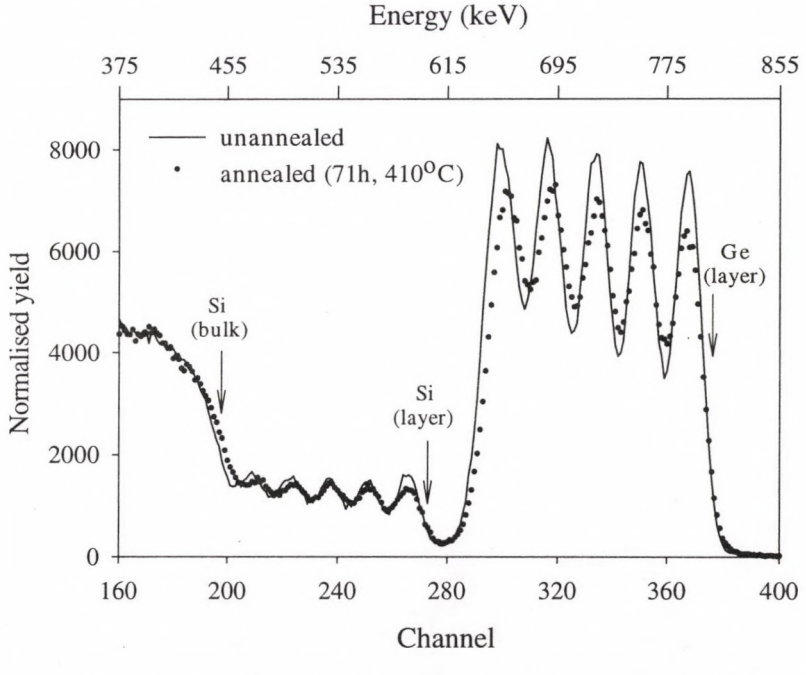

Fig. 1: RBS spectra of a Si/Ge multilayer sample

started. For increasing the depth resolution grazing incidence was applied. Figure 1 shows the measured RBS spectrum of a sample before and after annealing. The peaks belonging to the Si and Ge layers are well separated. By comparing the normalised spectra the decrease of the modulation amplitudes is determinable. Then the interdiffusion coefficient can be calculated [4]. In our results the calculated interdiffusion coefficient is in good agreement with the expected value. By RBS simulations we definitely showed the mechanism of the interdiffusion process, among the possible ones. Further heat treatments, calculations and Rutherford backscattering measurements with improved resolution are in progress in the frame of international collaboration supported by the International Atomic Energy Agency.

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3-dimensional scanning of ion-implanted porous silicon

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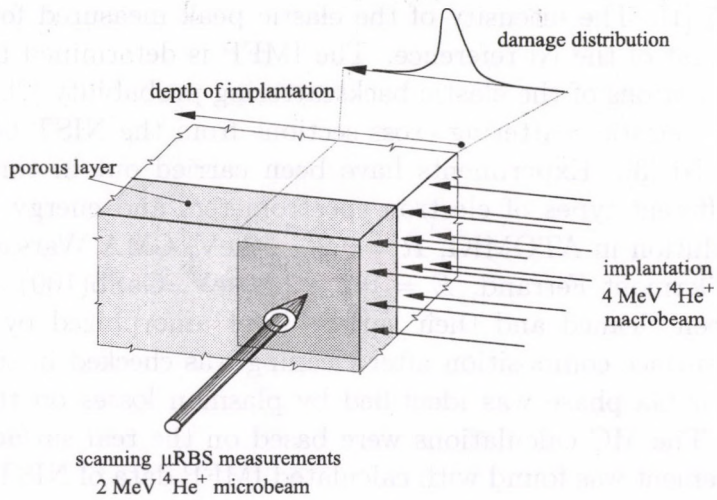
As it was recently shown, the porosity of porous Si gradually decreases under ion implantation, until the sample completely transforms into a compact material [1]. To determine the underlying elementary process, i.e. to decide whether the deposited total energy or the collision cascades are responsible for the above phenomenon, it is necessary to measure the degree of densification along the ion track.

For this purpose, different types of porous Si layers were implanted by 4.0 MeV $^4He^+$ ions from the side of the samples parallel to the surface (see the figure below).



This way, the depth profile of the implanted ions was transferred into lateral distribution. To transform the porosity changes into variations in the atomic composition, after implantation the pores were filled with hydrocarbon molecules. The implanted lateral spots were scanned 3-dimensionally by RBS (Rutherford backscattering spectrometry) microprobe using 2 MeV $^4He^+$ ions of 1.5×1.5 mm^2 beam size.

Results obtained for columnar and spongy type porous Si samples clearly demonstrate that the new method is indeed suitable to survey the irradiation induced densification along the ion track in porous samples. It was observed that the densification is much more effective around the penetration depth of the ions than elsewhere, indicating that this process is caused by the collision cascades and not by the deposited energy. Present results also suggest, that production of sharp buried compact layers in porous materials via ion implantation might be feasible [2].

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Experimental determination of the inelastic mean free path of electrons in GaSb and InSb

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Experimental determination of IMFP is based on elastic peak electron spectroscopy (EPES) [1]. The intensity of the elastic peak measured for the sample is compared with that of the Ni reference. The IMFP is determined from the Monte Carlo (MC) calculations of the elastic backscattering probability [2]. The MC algorithm is based on elastic scattering cross sections from the NIST 64 database and IMFP values of Ni [3]. Experiments have been carried out in three laboratories working with different types of electron spectrometers and energy range: HSA of high energy resolution in ATOMKI, $R = 0.2 - 5$ keV, CMA Warsaw, $E = 0.2 - 2$ keV and RFA Clermont Ferrand, $E = 0.2 - 1.5$ keV. GaSb(100) and InSb (100) samples have been cleaned and their surface layer amorphized by Ar^+ ion bombardment. The surface composition after cleaning was checked in situ by XPS. No metallic In, Sb or Ga phase was identified by plasmon losses on the surface after Ar^+ treatment. The MC calculations were based on the real surface composition. Reasonable agreement was found with calculated IMFP data of NIST [3] and others.

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Determination of the transmission and correction of electron spectrometers based on backscattering and elastic reflection of electrons

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The material parameter elastic reflection coefficient $r_e(E, Z, O)$ of a surface appears in the elastic peak intensity [1]. Absolute values of $r_e(E, Z, O)$ have been published by several authors, mainly working with RFA. Koch [2] published the angular distribution $r_e(E, Z, O)$ covering the $E = 400 - 2400$ eV range. Goto developed a CMA for elastic current measurements and published results on graphite, Ne, Au and Si [3]. The transmission of CMA and DESA 100 electron spectrometer of Staib Ltd was determined from the backscattering and quantitative elastic peak spectra of standard samples. Comparing experimental $r_e(E, Z, 137.7°)$ data of Koch with those of Goto, their ratios exhibited nearly constant values close to 5 at low energies and slowly decreasing with E. The elastic peak data are affected by the spectrometer energy resolution integrating the loss spectrum adjacent to the elastic peak. Spectrometer correction was needed; it was based on highly resolved loss spectra, measured with a HSA. The transmission (response) of the DESA 100 was determined. Experimental results of Goto have been compared with $r_e(E, Z, O)$ by Monte Carlo calculations based on NIST IMFP data.

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The paper was presented at QSA-10 International Conference, Birmingham, UK, 31 Aug. – 4 Sep. 1998. Abstract Book: AS.ThA.5 p. 47. The full paper to be published in Applied Surface Science.

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Installation of a new PGT Si(Li) detector and analytical application of a new clover-Ge-BGO detector system at the scanning nuclear microprobe of ATOMKI

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In 1998 the research possibilities have been improved significantly at the scanning nuclear microprobe of ATOMKI, in that way we could purchase and install a new Si(Li) detector (Princeton Gamma Tech. type) and a Clover-Ge-BGO detector system. In case of the Si(Li) detector our department received almost full financial support from the management of ATOMKI in the framework of an internal application. The Clover-Ge-BGO system has been shared by many teams in ATOMKI and partly installed for analytical purposes at our department in this year. The reader will find detailed information about its characteristics and installation in a separate paper in this volume.

A new experimental set-up for microPIXE

There is a continuous and growing interest for ion beam techniques all around the world particularly for the analysis of light elements. For example, in geological or biological research the measurement of C or Na, Mg and Al may be of vital importance. The installation of the new PGT Si(Li) detector with light element window at the nuclear microprobe beside our Canberra-type detector (25 μm Be window) have expanded the analytical range down to boron in our microPIXE measurements. This way light elements ($Z < 14$) and heavier ones ($Z \geq 14$) can be measured simultaneously in the same run without the need for repositioning samples, with fairly low limit of detection values and at a low cost/sample. The installation procedure involved a significant development of the measuring chamber. Special mechanical interfaces were developed for the detectors making it possible to hermetically separate them from the volume of the chamber when it is necessary (e.g. during RBS measurements, etc.). The optical microscope was placed on the top of the measuring chamber; a deflector magnet was designed for the protection of the PGT detector from the backscattered protons and the vacuum system was upgraded, as well. Furthermore, a beam-chopper was also elaborated for precise current measurement in the pA range and the capabilities of PIXYKLM program package extended to handle light elements.

A new system for gamma ray detection at the microprobe chamber

With the Clover-Ge-BGO system particle induced gamma ray emission (PIGE) measurements have been carried out on various types of samples at the nuclear microprobe. Artificial materials and geological specimens with known composition

were used to test the ability of the system. It turned out that at common condition of PIGE experiments remarkable low detection limits could be achieved for elements in the atomic number region of $3 \leq Z \leq 20$. These results can be derived from the excellent properties of this complex system. The Clover germanium detector in add-back mode has very high photopeak efficiency with only a slight deterioration of the energy resolution of peaks and using it with the BGO veto detector high values of Compton suppression factor can be obtained. These experiments were presented on the 6th International Conference on Nuclear Microprobe Technology and Applications.

Analysis of archaeological ceramics and glass samples by microPIXE and LA-ICP-MS methods

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Global chemical characterisation of archaeological ceramics or glass materials is commonly used to determine their provenance and also the workshops where they were manufactured. In case of ceramics, besides bulk analysis, the chemical characterisation of the different mineral inclusions in the samples may also be indispensable for provenance determination. However, their analysis is a rather difficult task as inclusions of 10–100 mm size are not easily identifiable on thick sections of ceramics, thus point methods have to be applied.

Laser ablation — inductively coupled plasma — mass spectrometry (LA-ICP-MS), proton induced x-ray fluorescence (microPIXE), and analytical scanning electron microscopy (SEM) are suitable and, in many respects, complementary techniques for the above tasks. The availability of a proton microprobe at ATOMKI and LA-ICP-MS and SEM techniques at the French institutes made it possible to start a joint research of archaeological ceramics and glass samples in the framework of COST-G1 program about one year ago.

Concerning ceramics the task has been the determination of provenance of some roman amphorae samples from Pian di Spille in the Latium and Mondragone on the Campanian coast, as well as some ones from the wreck of Le Grand Congloué (submerged near Marseilles). Because the trace element content of minerals found in different volcanic areas may be characteristic for their provenance, as a first step, we have started to analyse natural silicates such as olivines, augites, orthoses, sanidines and garnets from known sources. Though the obtained concentrations show non uniform distributions for some elements, nevertheless, further extensive research is needed before final conclusion.

In another project we have analysed ~ 30 Hungarian medieval (14th–17th centuries) glass samples found in the royal palaces of Buda and Visegrád. The results allowed us to discriminate between the soda-lime glasses (imported from Venice)

and the potash-lime ones (made locally). Furthermore, our results are in good accordance with those of the Orléans group for the origin of cobalt blue pigments in glass, as well. Namely, we concluded that from the 14th till the 17th centuries three different types of cobalt ores were used as colouring agents in three consecutive periods and they are characterised by different chemical (minor and trace element) composition. These results may be utilized both in exact dating and determination of provenance of glass artefacts of unknown origin. Publication in British Archaeological Reports is in progress.

Earth and Cosmic Sciences, Environmental Research

Geochronological studies with the K/Ar method in 1998

K. Balogh and Z. Pécskay

In 1998 continued the research of Tertiary magmatic rocks in Bulgaria (Cooperation: Geol. Inst. Bulg. Acad. Sci.). Papers appeared on the investigation of the Yabalkovo [1] and Briastovo [2] volcanic structure in the Eastern Rhodopes; Middle Eocene as well as Lower Oligocene ages were reported. A review paper summarizing earlier and recent studies of the Rhodopes has been also published [3]. A complex research has been performed on the alkaline basaltic magmatism SE of Krumovgrad, SE Bulgaria (C: Geol. Inst. Bulg. Acad. Sci.; Birkbeck College, London; Univ. of Florence) [4].

Improvements in experimental techniques of conventionel K/Ar and Ar/Ar methods have been published (C: KFKI, Atomic Energy Res. Inst.) [5].

A complex mineralogical and geochemical study of Devonian ultramafic intrusions of the southern Kola Peninsula, Russia, has been extended with K/Ar dating, which resulted ages similar to those of the Kola Alkaline Province (C: Birkbeck College, London; Univ of Tübingen; TsNIGRI, Moscow; Kola Science Center, Murmansk) [6].

In 1998 continued dating of Miocene volcanic rocks in Transcarpathia, Ukraine; Sarmatian – Pannonian ages were determined (C: Birkbeck College; ELGI; Geochem. Res. Lab., MTA; Inst. Geol. Romania).

Alkali basalts were dated from different areas of Serbia. Pan African ages were indicated by Ar–Ar method on an amphibolite from the Serb–Macedonian Massif. Ages were obtained both for the opening and closure of the Vardar ocean (C: Univ. of Belgrade).

Dating of ignimbrites in the Bükk Foreland has been continued and the laboratory participated in the complex (volcanology, geochemistry, paleomagnetism, chronology) research of the Tokaj Mts. (C: Geol. Surv. Hung.; ELGI; Geochem. Res. Lab., MTA; Inst. Geol. Romania).

Eocene-Oligocene-Miocene ages were measured on magmatic rocks from the Czech Middle Mts. (C: Geol. Inst. Czech Acad. Sci.).

In N Hungary Ar–Ar dating was performed on metamorphic rocks and gabbros. White micas from metamorphic rocks of the Vepor Unit resulted Lower and Upper Cretaceous ages and proved the Hercynian metamorphic age of the Vilyvitány crystalline schists. Gabbros from Szarvaskő and the Bódva valley resulted Middle Jurassic as well as Lower Triassic plateau ages.

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Radon flux density measurements on soil surfaces

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A method is developed to measure ^{222}Rn exhalation rate on soil surfaces using an ionization chamber radon monitor (AlphaGUARD PQ2000, Genitron Instruments GmbH, Frankfurt, Germany). First, the transient response function of the radon monitor to a linear change of the external radon concentration was measured using an 0.14 m^3 ^{222}Rn calibration chamber. A compartment mathematical model is developed to describe the time variation of ^{222}Rn concentration in the ionization chamber and to calculate the average ^{222}Rn concentration over the 10 minutes integrating periods. Comparison with experiments showed, that the model predicted the ^{222}Rn concentration values within the accuracy of the ^{222}Rn measurement of the instrument (Fig. 1).

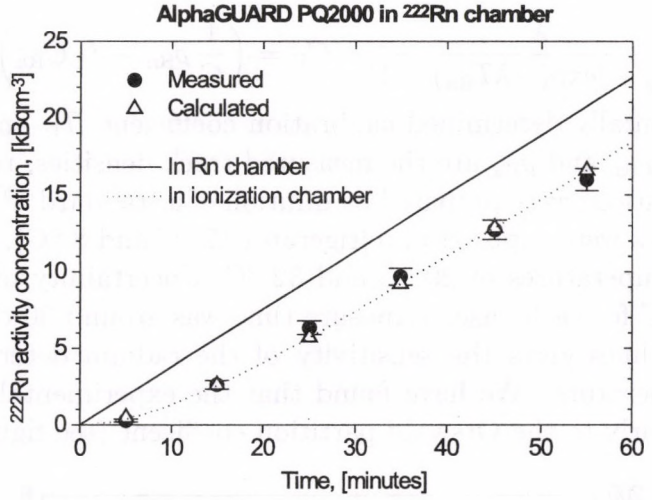

Fig. 1: Response of AlphaGUARD ionization chamber ^{222}Rn monitor to linearly increasing ^{222}Rn concentration. Comparison with compartment model description.

Then, the model was applied to measurements of ^{222}Rn concentration in a 60 cm diameter and 30 cm height exhalation vessel. A formula was found to extract free radon exhalation rate from measured time series. The lower limit of detection was found to be in the order of 1 mBqm^{-2}s^{-1}. Relative error of measurement was found to be about 10% at 25 mBqm^{-2}s^{-1} ^{222}Rn flux density. As an application of the method the effects of meteorological parameters on ^{222}Rn exhalation rate on an undisturbed soil surface were studied over 1 year period. Lower ^{222}Rn flux density were found in winter time. Effects of wind, rain, temperature and pressure variation were also observed.

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Radium determination in water samples by SSNTD

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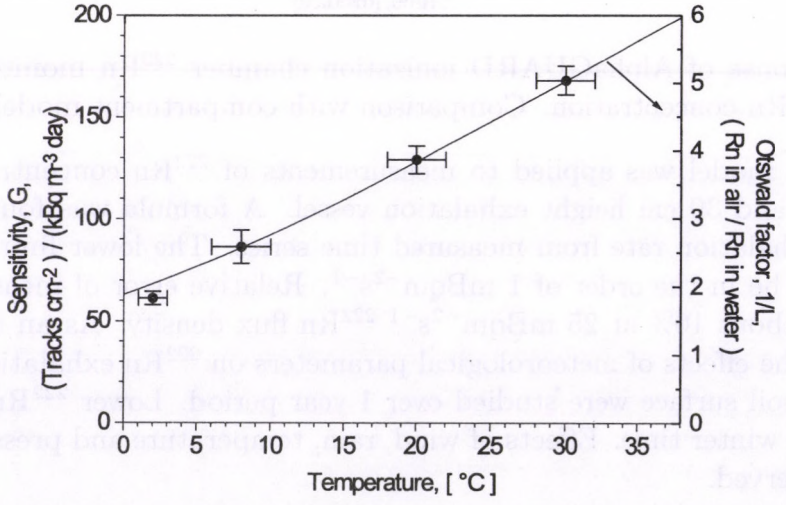
We developed a method to determine radon and radium content of water samples using an etched track radon detector (Radamon) packed in rubber bag, and immersed in the water. Radon activity content of natural water (C_W) consist of a radium bounded (C_{Ra}) and a radium unsupported (C_U) parts. Sealing the sample in field initial ^{222}Rn content of the water sample is $C_{W0} = C_{Ra} + C_U$. Initial ^{222}Rn content of degassed sample is 0. In case of complete mixing of water and small detector to glass volume ratio the problem can be solved analytically. Temporal variation of ^{222}Rn in a not degassed water sample is $C_W(t) = C_{Ra} + C_U \exp(-\lambda t)$, while in a degassed sample it is $C_W(t) = C_{Ra}[1 - \exp(-\lambda t)]$. After integrating we obtain:

$$C_{Ra} = \frac{1}{G}\rho_{Ra}\frac{\lambda}{\lambda T_{Ra} + [\exp(-\lambda T_{Ra}) - 1]}, \quad C_U = \left(\frac{1}{G}\rho_{Rn} - T_U C_{Ra}\right)\frac{\lambda}{1 - \exp(-\lambda T_U)}.$$

G is the experimentally determined calibration coefficient, T_U and T_{Ra} are the exposure times and ρ_{Rn} and ρ_{Ra} are the measured track densities, respectively.

Four ^{226}Ra solutions were prepared by dilution of a standard ^{226}Ra solution. Two concentration series were exposed in refrigerator (2 °C and 8 °C), and two in rooms with regulated temperatures of 20 °C and 32 °C. Uncertainity of the temperature was around 1–2 °C for each case. Exposure time was around 30 days for each case. The slope of the lines gives the sensitivity of the radium determination method at the given temperature. We have found that the experimental values vary with temperature similarly to the Otswald partition coefficent (see figure).



This work was supported by the Nat. Scientific Research Fund, No. 22985 and 16558.

Diffusion of ^{3}H, ^{99}Tc, ^{125}I, ^{36}Cl and ^{85}Sr in granite and concrete

Zs. Szántó, E. Hertelendi, M. Molnár and L. Palcsu

Understanding the characteristics of diffusion is essential in the assessment of radionuclide release through the backfill of waste repository. The diffusion behavior of ^{3}H, ^{99}Tc, ^{125}I, ^{36}Cl and ^{85}Sr in granite and concrete was studied in a specially designed diffusion cell. Diffusion coefficients (D) and time lag (t_D) were measured, break-trough curves were plotted and mechanism governing radionuclide diffusion was discussed, to interpret experimental data or to predict diffusion rates.

The results of the measurements in terms of calculated diffusivities are given in Table 1.

Table 1: Diffusion coefficients for ^{3}H, ^{99}Tc, ^{125}I, ^{36}Cl and ^{85}Sr in granite and concrete

Radionuclide	Backfill material	D ($\times 10^{-12}$ m^2/s)
^{3}H	Granite	15.0
	Concrete	54.9
^{99}Tc	Granite	4.6
	Concrete	–
^{125}I	Granite	0.06
	Concrete	26.0
^{36}Cl	Granite	0.023
	Concrete	–
^{85}Sr	Granite	0.1
	Concrete	–

The results showed that tritium, iodine and chlorine could be considered as non-sorbing tracers, while technetium and strontium were weakly sorbing (no break-trough was observed during the period of the experiments). The diffusion time of these experiments was 2 months. To obtain further information about the diffusion of chlorine and strontium in concrete we have to extend the experiments for longer times (90–120 days).Technetium was introduced into the system as pertechnetate (TcO_4^-) and normally has little tendency to be sorbed on geologic materials, but at pH = 12 technetium would be tetravalent (as TcO_2 or $Tc(OH)_4$) and highly sorbing on concrete.

The interaction of trace levels of ^{3}H, ^{99}Tc, ^{63}Ni, and ^{14}C with granite, concrete, carbonate, chlorite, quartz and Na-bentonite

Zs. Szántó, E. Hertelendi, M. Molnár and L. Palcsu

Using the batch technique, the interaction of ^{3}H, ^{99}Tc, ^{63}Ni, and ^{14}C with granite, concrete, carbonate, chlorite, quartz and Na-bentonite suspended in solution of synthetic groundwater was examined. These materials are being considered as possible components of barrier materials for nuclear waste disposal sites because of their high sorptivity, low permeability and long-term structural stability.

Table 1: Distribution coefficients for ^{63}Ni, ^{99}Tc, and ^{14}C in the investigated geological materials

Isotope	Geological material	K_d (m^3/kg)
^{63}Ni	Granite	0.76
	Carbonate	0.90
	Chlorite	1.55
	Na-bentonite	2.54
	Quartz	3.24
	Concrete	7.84
^{99}Tc	Granite	4.2×10^{-3}
	Carbonate	4.6×10^{-2}
	Chlorite	2.1×10^{-2}
	Na-bentonite	1.9×10^{-2}
	Quartz	2.8×10^{-2}
	Concrete	2.1×10^{-3}
^{14}C	Granite	2.4×10^{-3}
	Carbonate	4.4×10^{-3}
	Chlorite	2.6×10^{-3}
	Na-bentonite	1.0×10^{-3}
	Quartz	2.3×10^{-3}
	Concrete	4.0×10^{-3}

The sorption behavior including the change of distribution coefficients with decreasing the solution-to-solid ratios was investigated and sorption dynamic curves were plotted. The distribution coefficients (K_d) were calculated from

$$K_d = \frac{(A_0 - A_1)/W}{A_1/V},$$

where A_0 is the initial activity of solution spiked with radionuclide, A_1 the activity of solution after sorption equilibrum, W the weight of solid and V the volume of solution.

The results of the measurements in terms of calculated distribution coefficients are given in Table 1.

For tritium, there was negligible sorption onto the studied geological materials, and it could be considered as a conservative tracer (non-sorbing isotope).

Determination of ^{59}Ni, ^{63}Ni and ^{41}Ca in nuclear waste

Zs. Szántó, E. Hertelendi, M. Molnár and L. Palcsu

Two radiochemical separation methods were developed for determination of ^{59}Ni + ^{63}Ni and ^{41}Ca in the nuclear waste coming from the Paks NPP (used ion exchange resins and evaporator concentrates).

1 mg of Ni carrier was added to 100 ml evaporator concentrates or 10 g used ion exchange resin. After an acidic digestion in which 50 ml cc. HCl, 50 ml cc. HNO_3 and 30 ml cc. HNO_3 + 1 ml cc. $HClO_4$ were added to the samples, the residues were diluted with 50 ml bidistilled water and neutralized with cc. NH_4OH (pH = 7 – 8) to precipitate $Fe(OH)_3$. The solutions were filtrated, 3 ml DMG were added to the filtrates, when Ni-DMG were obtained. (DMG = dimethyl-glyoxime).

The cleaning procedure of the Ni-DMG includes 5 steps:

1. dilution of the Ni-DMG in 10 ml HCl.
2. evaporation to dry
3. dilution in 25 ml bidistilled water
4. neutralization with NH_4OH (pH = 7 – 8)
5. reprecipitation with 3 ml DMG

The reprecipitated Ni-DMG was extracted afterthat in chloroform, which was evaporated and the residue digested with HCl. After dilution in bidistilled water the samples were filtrated and the (^{59}Ni + ^{63}Ni)-DMG precipitate remaining on the cellulose nitrate filter paper was dryed and mounted into a metallic target support.

^{59}Ni was measured with a Si(Li) detector. ^{55}Fe standard was used for the calibration of the measurement.

^{63}Ni was measured by LSC techniques after dilution in HCl, digestion with 3 × 2 ml cc. HNO_3, evaporation to dry and dilution of the sample in 1 ml 0.5 M HCl. 10 ml Insta Gel Plus scintillation cocktail was added to the samples. Calibration was made with a known activity ^{63}Ni standard.

The XR measurement of ^{41}Ca was performed with a Si(Li) detector, after a radiochemical separation which includes several acidic digestions, evaporations and precipitation of the samples.

10 mg calcium carrier was added to 1 l evaporator concentrate or 100 g used ion exchange resin and incinerated during 24 h in an oven at 600 °C. After an acidic digestion step in which 50 ml cc. HCl, 50 ml cc. HNO_3 and 30 ml cc. HNO_3 + 1 ml cc. $HClO_4$ were added to the samples, the residues were evaporated to dry, diluted with 50 ml bidistilled water and neutralized with cc. NH_4OH (pH = 7 – 8). The filtration of the samples were followed by the precipitation of ^{41}Ca as calcium 8-hydroxy quinolinate in the presence of 50 ml 2n NH_4OH and 50 ml 2n NH_4Cl.

The precipitate was diluted in acetic acid, evaporated to dry and digested with 3×2 ml cc. HNO_3 and 2 ml H_2O_2. The residue was diluted with bidistilled water, neutralized with cc. NH_4OH to pH $= 7-8$ and afterthat ^{41}Ca was precipitated with 5–10 ml $(NH_4)_2CO_3$.

The ^{59}Ni activity concentration in evaporator concentrates was found about 103 Bq/l and in used ion exchange resin about 103–104 Bq/kg, while the ^{63}Ni activity concentration was somewhat higher (104–105 Bq/l in evaporator concentrates and 105 Bq/kg in used ion exchange resin).

The ^{41}Ca activity concentration was low, < 5 Bq/l in evaporator concentrates and about 80–100 Bq/kg in used ion exchange resin.

The role of Tritium–Helium method in sitting radioactive waste disposal facilities

László Palcsu and Ede Hertelendi

The performance of a mass spectrometric system for measurement of tritium by the ^{3}He ingrowth method is described. The system consists of a noble gas mass spectrometer (VG 5400) and units for quantitative extraction of the dissolved gases from water samples, purification of the extracted gases. A routine tritium measurement takes about 25 minutes. Calibration of the measurement is done using dry air standards, which are treated in the same way as a sample. The ^{3}He detection limit of the system is about 2×10^{-16} ccSTP corresponding to a tritium detection limit of 0.003 TU using 3000 g water sample stored for 2 months.

The measurement of tritium concentration of the water includes four steps. The first step is to collect water samples in the field and store them in 3 l glass bottles. The second step is the distillation of the all sample. After this approximately 3000 cm^3 of water is poured into a 6 l metal bulb provided with metal valve. The water sample is degassed by shaking the metal bulb, because the gases are transported out of the bulb by water vapour and pumped away. After gas extraction the metal bulb is closed and stored for typically 2 to 4 months to allow ^{3}He ingrowth from tritium decay. In the last step the sample containers are fitted to the inlet system of the mass spectrometer for measurement. Here the water vapour is removed in a cold trap cooled by liquid nitrogen and all gases except helium and neon are adsorbed on a charcoal held at liquid nitrogen temperature. After the purification the helium fraction is admitted to a dual collector noble gas mass spectrometer and ^{3}He and ^{4}He are measured simultaneously applying the peak height method.

The tritium concentration of a sample can be calculated from the measured tritiogenic helium as follows:

$$c_{\mathrm{trit}} = \frac{^3\mathrm{He}_{\mathrm{trit}}}{\mathrm{C}} \frac{\exp(\lambda t_{\mathrm{se}})}{1-\exp(-\lambda t_{\mathrm{em}})} \frac{1}{W} \left[1-(\alpha-1)\frac{W_0-W}{W_0}\right],$$

c_{trit} - tritium concentration in TU,
$^3\mathrm{He}_{\mathrm{trit}}$ - measured tritiogenic 3He in ccSTP,
C - conversion factor from ccSTP to TU (2.4889×10^{-15} ccSTP/g/TU),
λ - reciprocal of mean lifetime of tritium.
t_{se} - time from sampling to extraction in day,
t_{em} - time from extraction to measurement in day,
W_0, W - weights of sample before and after the extraction in gram.
α - correction for the T/H fractionation due to loss of distilled water during gas extraction. $\alpha = 1.15$ (ratio of tritium concentration in the liquid phase to tritium concentration in the water vapour).

The method was used to estimate the infiltration rate of the three-phase zone and mean residence times of water in the saturated zone in the Tolna hill area. The

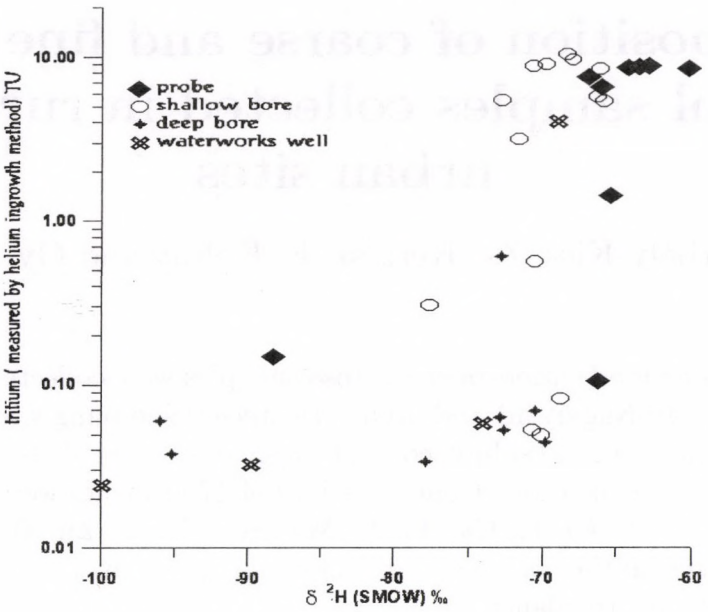


Fig. 1: Tritium concentration plotted against of $\delta^2 H$ in different samples

method played an essential role in the decision making process of site selection for final disposal of low and intermediate level radioactive waste. Results show that even water coming from deep boreholes contain tritium indicating mixing of fresh water with older water bodies. The correlation between tritium content and δD values was satisfactory (Fig. 1). The detection limit of T–He method proved much lower than the detection limit of conventional tritium method using electrolytic enrichment and liquid scintillation counting technique (Fig. 2).

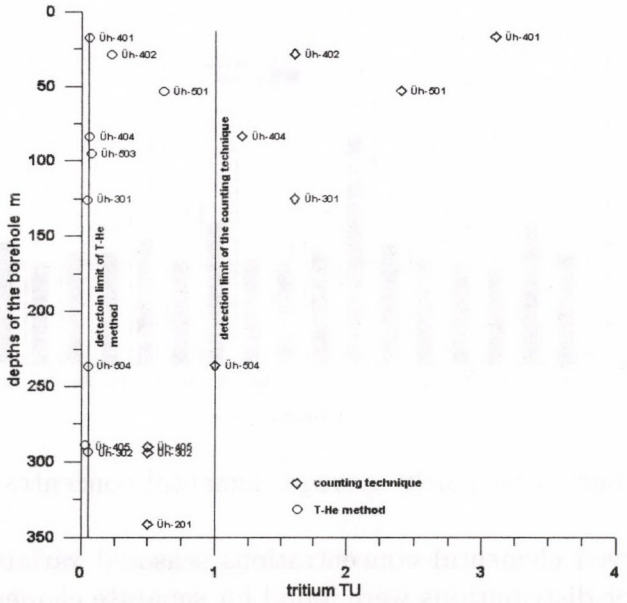


Fig. 2: Tritium contents and detection limits measured by two different techniques

Composition of coarse and fine mode aerosol samples collected in rural and urban sites

I. Borbély-Kiss, Zs. Kertész, E. Koltay and Gy. Szabó

Coarse and fine mode atmospheric aerosol samples were collected simultaneously in rural (Hortobágy-Nagyiván) and urban (Debrecen) sampling sites with two-stage stacked filter units, and absolute concentration data were deduced on elemental constituents by PIXE method. Concentrations of 17 elements were detected on the samples: Al, Si, P, S, Cl, K, Ca, Ti, V, Mn, Fe, Ni, Cu, Zn, Br, Ba, Pb. Total particulate masses on the coarse and fine mode samples have been measured by the use of a Sartorius microbalance.

The yearly average of the total mass of the urban particulate matter in coarse mode was equal to 16 $\mu g/m^3$. Rural particulate matter in coarse mode, was 10 $\mu g/m^3$. Urban particulate matter in fine mode was 21 $\mu g/m^3$, and rural particulate matter in fine mode was 17 $\mu g/m^3$.

In general, remarkable similarities are present in the respective urban and rural concentration values. Rural values are as a rule lower (or equal), than urban values. Differences are higher in coarse mode than in fine mode. It can be clearly seen in Fig. 1.

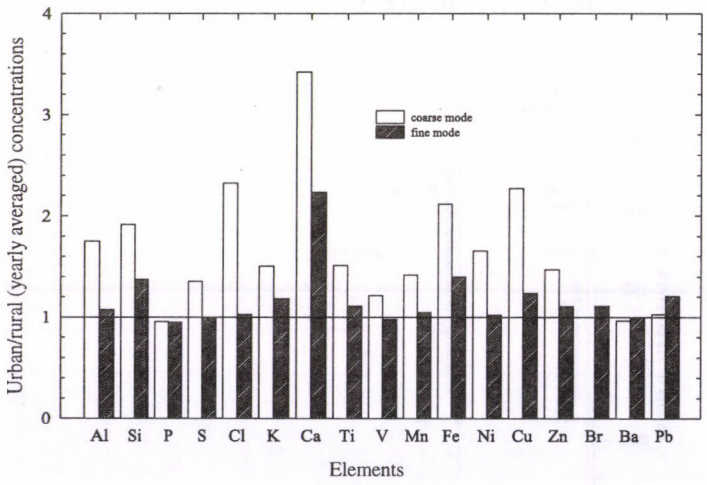


Fig. 1: Urban/rural yearly average elemental concentration ratios

From the observed elemental concentrations seasonal variations, and variation with the wind sector distributions were found for separate elements. Principal component analyses gave four factors in urban aerosol, and three factors in the rural one. The order of the factors in the coarse mode aerosol is different from the order, found in the fine mode. Results in detail are published in [1].

The present work was supported by the Hungarian National Foundation for Scientific Research (OTKA No. T17040).

[1] Borbély-Kiss, I., Kertész Zs., Koltay, E., Szabó, Gy., Tar K., Nucl. Instr. Meth. in Phys. Res. B (accepted for publication)

Elemental composition study of aerosols collected in a speleotherapeutic cave situated below Budapest

Zs. Kertész, I. Borbély-Kiss and I. Hunyadi

The Szemlőhegy-cave is one of the well-known hydrothermal caves of the densely poppulated Rózsadomb area of Budapest, which have been used for speleotherapy of respiratory diseases for years. It is known from the periodically changing airborne radon activity concentration data, that airflow of seasonally reversed direction are formed along the cave passages and fissures due to the temperature difference between the surface and cave air. This means that an intensive interaction takes place between the cave and its environment. The pollution of nearby waters and the urban atmospheric air represents a real danger for these caves below Buda, which are candidates for the UNESCO World Heritage. The study of cave aerosols should be very important from the point of view of either the control possibilities of environmental protection or speleotherapy, and probably helps in getting acquainted with the cave-forming processes. We applied our standard aerosol sampling method to the high-humidity environment of the caves, and we studied the elemental composition, size fractionation as well as the spatial distribution and the seasonal variation of cave aerosols.

Aerosol sampling was performed in October 1995, when the summer air circulation was dominant, and in January 1996 and in March 1998, when the winter air circulation took place in the cave. Six portable membrane pumps with 2-stage Nuclepore filter holders were placed along the cave fissures to obtain aerosol spatial distribution. In March 1998 a 7-stage cascade impactor was also deployed in the speleotherapeutic sites, with the help of which we sudied the size fractionation. Temperature and relative humidity were also measured during the sampling.

Concentrations of the following elements were determined by PIXE: Al, Si, P, S, Cl, K, Ca, Ti, V, Mn, Fe, Ni, Cu, Zn, Ba, and Pb.

We pointed out that the external polluted air penetrate into the cave, down to its bottom due to cave air motions. However aerosol concentration diminish with increasing distance from the entrance, and are minimal in the site of the therapy. Statement could be made also for the most probable air passages. With the help of the cascade impactor the cleaning effect of the cave was also showed.

Though traces of the anthropogenic pollution of the Budapest air is shown in the Szemlőhegy-cave, every case the measured elemental concentrations remained less than one-tenth the air quality standards valid for the increasingly protected areas. The elements S, Cl, Zn, Br or Pb as tracers indicate the interaction between the cave air and the urban atmosphere, and also the direction and the strength of air movements in the cave.

This work was supported by the National Scientific Research Found, Budapest (Contract No. OTKA-T017040 and T016558), and the results are accepted for publication in NIMB (Ref. No. 2101): Zs. Kertész, I. Borbély-Kiss and I. Hunyadi: Study of Aerosols Collected in a Speleotherapeutic Cave Situated below Budapest, Hungary.

Influence of surface properties on the diffusion profiles obtained by Rutherford backscattering spectrometry

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It is commonly known that surface roughness, such as grains, can make the interpretation of RBS spectra difficult, as they can cause diffusion-like broadening. Recently Metzner et al. [1] have proposed a model which can determine the surface topography from the measured RBS spectra. This model cannot however be used in cases where both surface roughness and diffusion are involved and the signal caused by the surface properties overlaps the signal from the diffused atoms. For this purpose we have developed a method which combines RBS with atomic force microscopy (AFM).

The gold is widely used as a contact material on compound semiconductors and the electrical properties of the Au/ZnSe system have been extensively studied, therefore we chose this material for our investigations [2]. The ZnSe were grown on GaAs substrate by molecular beam epitaxy, and the gold was deposited on top of it in e-beam vacuum evaporator. After the preparation the sample was annealed in argon atmosphere. During this process besides the diffusion, grain formation also occurred on the surface.

RBS spectra of the samples were measured with 2 MeV He^+, both macro- and microbeam with a beam size of 2×2 mm^2. The scanning nuclear microprobe measurements on grains and grain free areas showed that the grains on the surface consisted of mainly gold and confirmed the diffusion of this element. The topographical information obtained from the AFM study were used to calculate the resulting theoretical macro-RBS spectrum. From the AFM results we calculated the area of a given grain thickness. RBS spectra of gold layers on top of Au/ZnSe heterostructure were then calculated with the same thickness as in the AFM calculations. The diffusion profile was also included in the calculated spectra. These spectra were then weighted with the relative area of the specific grain thickness and summed to get the final spectrum.

We can conclude that the final spectrum fits well the measured points. The diffusion coefficient calculated by the micro-RBS was in good agreement with the combined macro-RBS result, which proved the correctness of the new method.

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[2] J. Slotte, A. Laakso, T. Ahlgren, E. Rauhala, R. Salonen, J. Räisänen, A. Simon, I. Uzonyi, Á.Z. Kiss and E. Somorjai, submitted to Journal of Applied Physics Letters

Biological and Medical Researches

One column method to prepare ^{11}C-labelled methyl iodide

Z. Kovács, É. Pribóczki and G. Horváth*

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The generally used method for production of [^{11}C]methyl iodide starts from $^{11}CO_2$. The $^{11}CO_2$ produced is recovered usually in an empty spiral tube cooled down to -160 °C [1, 2]. The $^{11}CO_2$ is then released by warming and led into a reaction vessel to form radio-methylate complex with lithium aluminium hydride. This radiocomplex is converted either to [^{11}C]CH_3I with HI under reflux conditions [3] or first hydrolyzed into radiomethanol. The hydrolysis is carried out in the same reaction vessel and the radiomethanol is distilled out at approximately 160 °C [4]. The [^{11}C]methanol is converted into [^{11}C]methyl iodide with different iodinating agents [4, 5] or simply on an alumina column mixed with HI [2].

We report a different method in which the [^{11}C]methyl iodide is prepared on one alumina column. Instead of using a separate low temperature unit for trapping of the $^{11}CO_2$ and a reaction vessel for reduction with $LiAlH_4$, a high specific surface alumina column, previously impregnated with lithium aluminium hydride solution, was used for direct trapping from the target gas and reduction into radiocomplex. The complex was then reacted on this column with HI to form [^{11}C]methyl iodide. The use of one alumina column, instead of a freezing trap, reaction vessel and separate unit for iodination, simplifies the apparatus, shortens the synthesis time and is well suitable for automation.

The irradiated N_2 target gas was transported with 1 l/min flow rate into an alumina column containing 300 mg neutral aluminium oxide in a glass tube with 4 mm i.d. The alumina was degassed and impregnated with 250 μl 0.2 M ethereal $LiAlH_4$ solution before use [6]. The target gas was led through the column at room temperature to get [^{11}C]methylate complex.

After finishing the trapping, the diethylether was eliminated from the column under 30 ml/min He flow with raising the temperature up to 160 °C followed by addition of HI (200–400 ml, 57%) to form [^{11}C]CH_3I. The product was transported into cooled acetonitrile for HPLC analysis [6]. In the iodination step the radiomethanol partly escapes (~40%) without forming [^{11}C]CH_3I, therefore the amount and the concentration of HI, the length of the column and the volume speed of the carrier gas still have to be optimized.

Combining our former separate column methods for the synthesis of [^{11}C]CH_3OH [6] and the iodination of radiomethanol with HI to produce [^{11}C]CH_3I [2] into a single column synthesis, the preparation time was shortened to be 10 minutes which means about 30% increase in the overall yield.

This work was financially supported by the Hungarian Scientific Research Fund No. T 019079. The authors thank the crew of the cyclotron for irradiation.

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[6] Sarkadi, É., Kovács, Z., Horváth, G., Radiochimica Acta **83** (1998) 49

Development of Methods and Instruments

Installation of a Ge + BGO γ-ray spectrometer in ATOMKI

A. Krasznahorkay, Á.Z. Kiss, M. Csatlós, Z. Gácsi, J. Gulyás, Z. Máté and J. Molnár

A significant improvement of the research possibilities in γ-ray spectroscopy were made by the installation of a Clover Ge + BGO spectrometer in 1998. The spectrometer was purchased in a long standing and fruitful cooperation between KVI Groningen and ATOMKI and supported by OMFB and several OTKA programs due to the considerable cost.

The so called "clover type" composite HPGe detector includes four separate coaxial n-type Ge crystals packed together in a four-leaf clover arrangement. It has been developed by CRN Strasbourg and Eurisys Mesures (France). The surrounding bismuth-germanate (BGO) anti-Compton shield, which was manufactured by Cyberstar S.A. (France) contains sixteen independent crystals. 120% relative efficency (at 1332 keV) and 0.55 peak/total ratio have been obtained in "adding-back" mode with the Ge crystals and using the usual anti-Compton suppression technique.

The installation and the first experiment using the clover detector were performed at KVI Groningen. Using the possibilities of a newly built superconducting cyclotron AGOR, we studied the γ-decay of the Spin Dipole Resonance (SDR). We have demonstrated that the strength of the SDR is related to the neutron-skin thickness of nuclei [1]. By measuring the γ-decay one can get additional information to the microscopic structure of this giant resonance.

The spectrometer was moved to Debrecen after the experiment and installed at the Nuclear Spectroscopy Department. In ATOMKI we are using the data taking system developed previously for the split-pole magnetic spectrometer [2]. The Clover + BGO spectrometer plays an important role in a program which aims at the γ and conversion electron spectroscopic investigation of uranium isotopes in which hyperdeformed states were found recently [3].

Another application in elemental analysis of this excellent device is also reported in this Annual Report [4].

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Research and Development at the ECR ion source

S. Biri, A. Valek and L. Kenéz

A gradually growing interest could be detected in 1998 toward the Electron Cyclotron Resonance (ECR) Ion Source Laboratory from foreign research institutes and from internal education organisations. The parameters of the heavy ion beams available for research, education and applications are continously submitted in the homepage of the ECR Laboratory (http://www.atomki.hu/atomki/ECR). In the ECR laboratory several students perform research and have the opportunity to write their diploma work and Ph.D. dissertation. In the fields of research and development the main results in 1998 are followed.

Instrument development. During the year new heavy ion beams were produced both from gaseous and solids samples (Kr, C, Ni). To ionize from solids we developed a new method to regulate the metal particle flow (see figure and [1]). This way the generated charge state distribution of the extracted beam could be modified. We also solved the problem of the 14.5 GHz microwave power regulation resulted in an intensity increase of factor of 2–3 for the higher charge states.

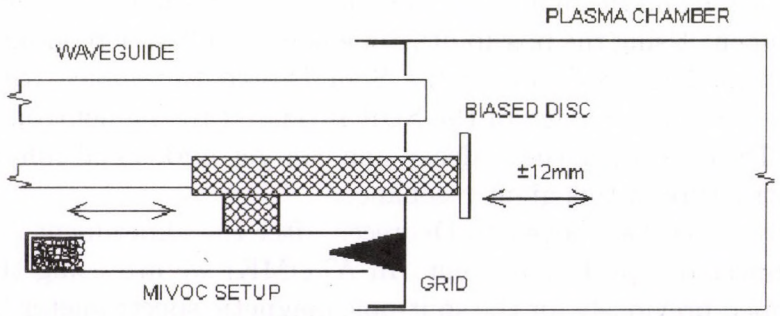

Fig. 1: The solid sample holder (MIVOC setup) and the electron donor electrode (biased disc) form a new unit to regulate gas flow and charge state distribution.

Plasma physics research. The investigation of an electron donor electrode (which was built into the ECRIS in 1997) led to a new approach: the injected external electron current will not increase directly the plasma density, but it modifies the trapping and extracting conditions by means of changing the plasma potential [2]. We also measured numerous Kr X-ray spectra using a Si(Li) detector at different ion source tunings. Analysing these spectra and the simoultaneosly measured extracted ion beam intensities we studied the connection between the properties of the internal plasma and the charge state distribution of the external highly charged particle beam.

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Production of highly charged ions in ECR ion sources using an electrode in two modes

S. Biri and T. Nakagawa*

*The Institute of Physical and Chemical Research (RIKEN), Japan

We placed an axially movable isolated electrode into the plasma chambers of the 18 GHz RIKEN and 14.5 GHz ATOMKI Electron Cyclotron Resonance Ion Source (ECRIS). The goal was to produce a higher intensity of multiply charged ions and to study how the electrode affects the highly charged ion production. We found that the effect of the electrode strongly depends on the local plasma parameters. At lower floating potential we need to increase the plasma density by means of biasing the electrode and injecting electrons into the plasma. The electrode operates as an electron source (Biased or Electron Donor mode, Fig. 1, left). At higher floating potential the electrode works by changing the plasma potential. The best result is obtained when the electrode remains at floating potential (Floating or Plasma Tuner mode, Fig. 1, right). These two modes were checked and successfully found both in continuos and in pulsed mode operation. In both (ED and PT) modes we generated higher highly charged ion currents than without the electrode. In PT mode we successfully obtained 300 eμA of Ar^{11+} at the RIKEN-ECRIS. This high current means a milestone in the ECRIS beam developments. We continue these investigations to understand the exact mechanism of electrode-plasma interaction in the ECR ion sources and traps [1].

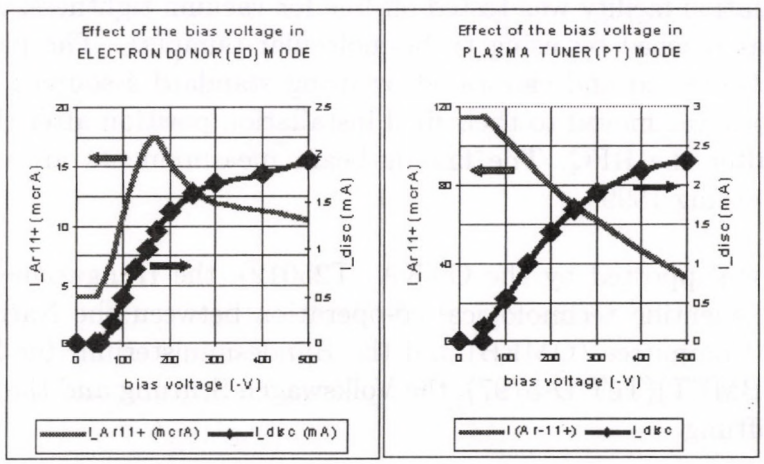

Fig. 1: The two operating modes of the auxiliary electrode in ECR ion sources

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Scattering chamber on the ECR-RFQ beam for RBS/channeling measurements

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In the framework of a bilateral cooperation between the ATOMKI and the above German institute on the field of application of highly charged heavy ions for the purposes of material science instrumental development has been made both in ATOMKI and in the Frankfurt institute. In both laboratories 14.5 GHz ECR Ion Sources (ECRIS) were established several years ago. These sources can accelerate highly charged heavy ions with an extraction voltage of up to 30 kV [1] (60 kV [2]). In the Frankfurt institute an RFQ post/accelerator stage was also established after the extracted beam of the ECR with an output energy of 100–200 keV/amu [3].

Investigation of crystalline material (semiconductors) has been performed in cooperation in the two institutes for several years [4]. As a new problem the study of charge state modification in channeling circumstances has been emerged. For this reason a scattering chamber assembly was built at the ECR-RFQ beam line of the Frankfurt institute. This irradiation equipment contains independent high-vacuum system, beam monitoring devices, a large-volume target chamber, detector ports with semiconductor particle detectors (PIPS), sample manipulator (goniometer) and beam-current measurement facility. The system is also furnished with two adjustable collimators about 2 m apart from each other in front of the sample position. This collimating path is of vital importance in channeling for producing highly parallel beams.

This irradiation facility was tested off-line for vacuum tightness. Approximately 10^{-7} mbar was reached by using turbo-molecular pump(s). The particle detector system was also tested and calibrated by using standard á-sources. The chamber and beam-tubes was moved to their final installation position after the first switching magnet after the RFQ. The first in-beam measurements are expected to be performed in spring 1999.

This project is supported by the OTKA (T25012), the Hungarian–German intergovernmental scientific–technological co-operation between the National Technical Development Committee (OMFB) and the Bundesministerium für Forschung und Technologie (BMFT)(TéT D-5/97), the Volkswagen Stiftung and the Alexander von Humboldt Stiftung.

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[3] http://hsbpc1.ikf.physik.uni-frankfurt.de/ezr/rfq.htm
[4] http://www.atomki.hu/~ditroi/

Escape-reabsorption effect in scintillation detectors with photodiode readout

G. Kalinka, J. Gál and B.M. Nyakó

Escape of characteristic X-rays of energy E_x from the detector material following photoelectric effects when detecting X- or γ-rays of energy E_0 results in a satellite peak displaced by E_x from E_0. This phenomenon is common to scintillation (SC) detectors with PMT readout, as well. Using photodiode (PD), itself being sensitive to ionizing radiation, instead of PMT, another interaction can also take place: a reabsorption of escaping X-rays in the PD. This effect is pronounced in thin SCs, where the escape-to-photopeak ratio is high: $r_{esc} \approx J\omega/2$, for one side, J, ω being the jump factor and fluorescent yield respectively. In the above process energy E_x is detected directly in the PD, and the remaining $E_0 - E_x$ energy "packet" via scintillation simultaneously. The resulting amplitude of the related escape-reabsorption peak in the PD energy scale, therefore is

$$E_{era} = E_x + (E_0 - E_x)K_{sc}.$$

The K_{sc} factor, which represents the scintillation/direct detection efficiency ratio can be expressed as

$$K_{sc} = \frac{\varepsilon_{pd}}{\varepsilon_{sc}}\, \eta_L^e\, \eta_C\, (1 - B^e) \sum \psi(E_{ei})\, \eta_{sc}^e(E_{ei})\, \frac{E_{ei}}{E_0 - E_x},$$

where ε_{pd} and ε_{sc} are the e-h pair creation energy in the PD and SC, respectively; η_C is the charge collection efficiency of the PD; the η_L^e light collection efficiency and B^e ballistic deficit for electrons are considered as independent of energy; $\psi(E_{ei})$ is the energy distribution of energetic electrons after electron cascade following emission of an E_x quantum, when $\sum E_{ei} = E_0 - E_x$, and $\eta_{sc}^e(E_{ei})$ is the energy dependent scintillation efficiency for electrons. Knowing the $\psi(E_{ei})$ distribution versus E_0 relationship for a given scintillator material it is possible to determine the $\eta_{sc}^e(E_{ei})$ electron response as a fundamental property of the scintillator. Although this method is not as elegant and straightforward as the Compton Coincidence Technique [1], the counting efficiency is much higher, especially at lower energies, and, due to the E_x biased energy scale no lower limit on E_0 is imposed.

A preliminary experiment with a $10 \times 10 \times 0.3$ mm^2 CsI(Tl) SC coupled to a same size Si pin PD clearly showed the effect. An accurate determination of the energy shift of I and Cs $K_{\alpha,\beta}$ lines was, however impossible even by a spectrum deconvolution due to the strong overlap of these lines as a consequence of the $\sim$3 keV noise level. The improvement of resolution by using low noise readout device, as e.g. a CCD or SDD [2], is therefore indispensable.

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Intrinsic energy resolution of CsI(Tl) scintillation detectors

G. Kalinka, J. Gál and B.M. Nyakó

Early experiments for the determination of intrinsic energy resolution as a measure of the dispersion of photons created in scintillation detectors were strongly influenced by poor statistics due to low photoelectric yield of PMTs [1]. CsI(Tl) is among the scintillators for which the scintillation photon to e-h pair conversion efficiency with Si photodiodes (PDs) is practically unity, thus providing much better statistics. Contrary to PMTs, however, the limiting factor in this combination is the much higher electrical noise, which should therefore be taken into correction for the determination of the true intrinsic resolution.

Our detector, developed for light charged particle detection [2], was composed of a $15 \times 15 \times 3$ mm^3 CsI(Tl) crystal coupled to a 10×10 mm^2 Si PD via a trapesoidal plexiglass lightguide. A low noise preamplifier and time variant signal processing enabled < 3 keV$_{Si}$ noise level. Spectra taken with 60–2600 keV γ-rays and 5.15–8.78 MeV α particles from radioisotopes were processed with the EWA spectrum evaluation code [3]. γ photopeaks could be fitted with a single Gaussian, whereas α peaks showed a more complex lineshape due to increased light collection nonuniformity and could best be described by the Doniach–Sunjič model [4, 5], from which the Gaussian component was considered to be related to the intrinsic resolution. Noise subtracted FWHM peak widths $\Delta^y_{d,Si}$ ($y = \gamma, \alpha$) were plotted as a function of the corresponding amplitudes E^y_{Si} (keV$_{Si}$) rather than their source energies E_y to enable direct comparison of the variance in the number of γ and α created photons. Both set of intrinsic resolution data can be approximated by $\Delta^y_{d,Si} = [K_y\, E^y_{Si}]^{1/2}$, with $K_\gamma = 0.25 \pm 0.01$ keV and $K_\alpha = 0.34 \pm 0.02$ keV. For this particular detector these coefficients are 12 and 16 times higher, respectively, than expected for pure Poissonian photon statistics. In the light of recent results [6] the discrepancy for γ-rays is, at least partially, related to the nonproportional electron response of CsI(Tl). The extra deviation for α-particles awaits further study.

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[3] J. Végh, ECASIA '95, John Wiley & Sons, 1996, p. 680
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Charge sensitive preamplifier with low power consumption

J. Gál, G. Hegyesi, G. Kalinka and B.M. Nyakó

In nuclear radiation spectroscopy the interconnection between the detector and the preamplifier should be as short as possible. A long interconnection increases the stray capacitance at the preamplifier input causing enhanced series noise contribution and emphasised microphonic effect. To diminish these effects the best solution is building together the detector and the preamplifier. If the detector is placed in a vacuum chamber, e.g. for the detection of light charged particles, this solution — because of cooling problem — is acceptable only if the preamplifier has very low power dissipation.

During the development of the low power consumption preamplifier we strove not to sacrifice either the noise or the speed parameters of the amplifier. Fig. 1 shows the circuit diagram of such an amplifier, which is a variant of the circuit published in [1]. It applies a low power fast (300 MHz) current feedback operational amplifier [2] which has only 1mA quiescent current and operates from ±5 V supply. To assure the appropriate drain-source voltage even in this low voltage operation the source of the FET is connected to the negative supply through the R6, C6 low pass filter. The drain voltage is zero, therefore the FET current is set approximately to 1mA by R2, R7. Since the gate must be more negative than the source an additional biasing network is required. This network is realised using a current source (T2, R8, R9) and a shifting resistor R3 bypassed with C3. The output DC level of the preamplifier can be set to zero by adjusting the current with R2.

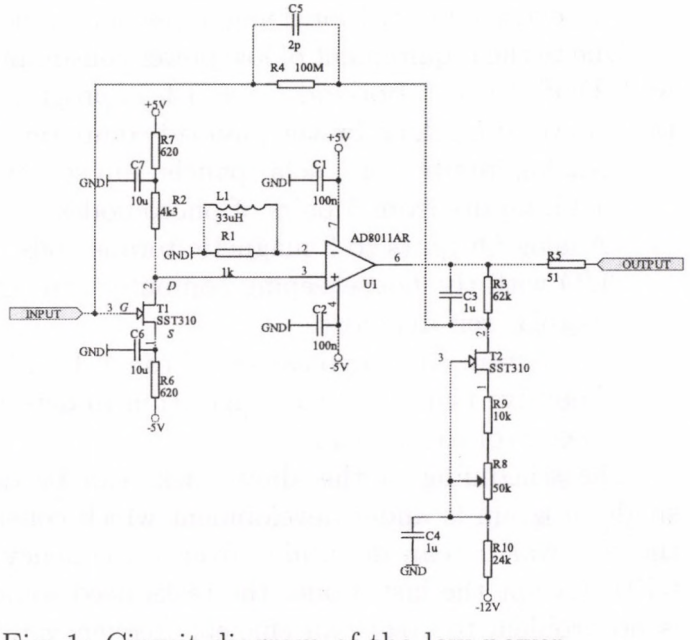

Fig. 1: Circuit diagram of the low power consumption preamplifier

The total power dissipation of the preamplifier is approximately 22 mW, and its geometrical size is 12×15 mm, with a maximum height of 3 mm. The noise and speed parameters are the same as given in [1].

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Preparing the control of a nano satellite

G. Székely and J. Molnár

In collaboration with the Instrumentation group at Physics department of Royal Institute of Technology in Stockholm we are taking part in a project for launching a satellite (see http://www.particle.kth.se/~fmi/hugin/huginstart.html). Its name is HUGIN (name of a god in Swedish mythology) and it is for evaluation of magnetic attitude control and neural networks.

For controlling the satellite [1] a housekeeping and a payload computers will be built in. The authors of this paper will do the programming of the payload computer and will construct an extra I/O card to it The payload computer is a PC/104 system consisting of the following components:

· CPU card with 20 MB RAM disk (AMPRO - CoreModule/3SXi)
· 32 channel AD/DA card (AJECO - DSP104/C31)
· an extra I/O card for doing accessory tasks (see below)

Due to the requirement of low power consumption a 9.8 MHz 8088 CPU is applied with DOS 3.1 and Borland C++ 3.1 as programming environment. The following tasks have to be done by the payload computer:

· Analog inputs from: solar panels, sun sensor, magnetometer and thermometers
· TTL inputs from 5 pairs of photodiodes
· Analog Outputs to 3 magnetic torque coils
· I/O with the housekeeping computer through COM1
· Regular self diagnostics
· Input from the video camera through LPT1
· Adaptive neural network algorithm to determine the optimum output values to control the attitude.

The scheduling of the above tasks can be done in several ways. Presently a single program is under development which consists of a main loop. By examining the real time it can determine from a frequency table which task has to own the CPU. Except the last 3 ones the tasks need some milliseconds CPU time, so there is no problem to create an efficient frequency table as the base of the scheduling. Scheduling the last 3 tasks is not so easy because they require several seconds. A whole picture input from the video camera e.g. needs 5–6 seconds due to the slow CPU and bus. Hence these tasks have to be segmented in order not to leave the system without proper control for longer periods.

The tasks of the extra PC/104 card:

· To hold the sun sensor
· To convert $[-3, 3]$ V from the 3 outputs of the magnetometer to [0,2.5] V
· Handle the signals from the photodiodes
· Adjust control signals to the coils

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Renewal of the cyclotron vacuum system

Z. Kormány, P. Kovács and I. Szűcs

The modernization project* for the vacuum system of the cyclotron was started in 1997. During 1998 significant efforts were focused on the implementation of the project and at the end of the year it got close to the completion. The expected date for accomplishing this work is at the end of the winter maintenance period in February 1999. The scope of the project and the achieved status are detailed in the following:

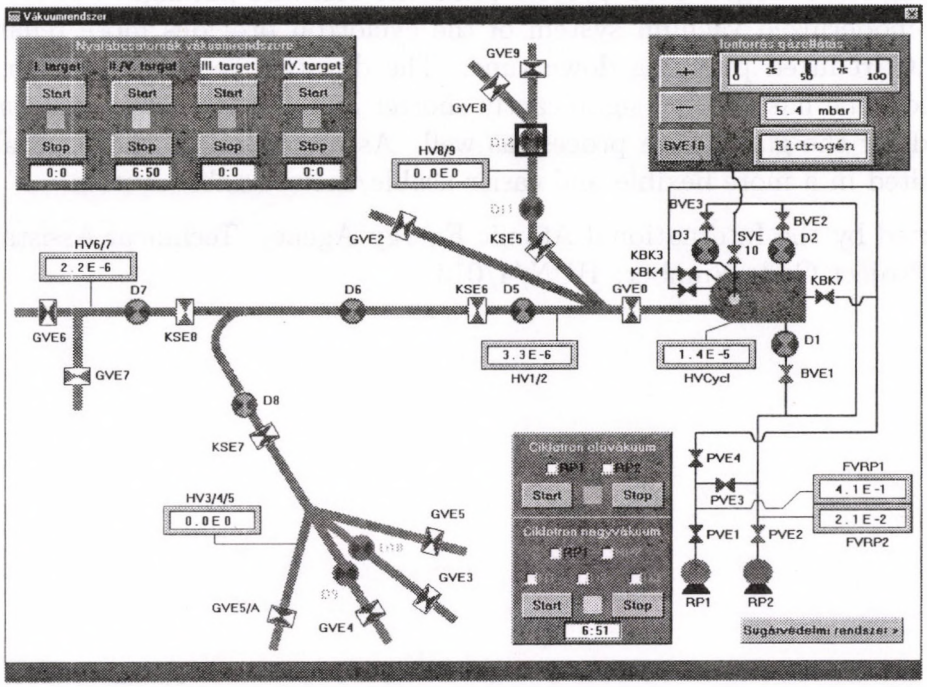

Fig. 1: The graphical user interface of the vacuum control system (the control panel text is — intentionally — in Hungarian)

1. Vacuum measuring system

The outdated measuring units and heads have been replaced by Balzers TPG-300 measurement units and new Pirani and cold-cathode gauges. They were put into operation in the middle of 1997 and have been working very reliably since then.

2. Beam transport system

All the ion-getter pumps were dismantled and eight new Edwards 100/300P diff-stak pumps have been installed. The manually operated gate valves in the transport channels have been replaced by pneumatic ones.

3. Cyclotron vacuum chamber

The fore-vacuum system of the cyclotron chamber has been renewed by installing two high performance Edwards E2M80 rotary pumps and replacing all the manual valves in the system by pneumatic ones.

4. Gas supply system

The manual valves will be replaced by high pressure solenoid valves and the gas flow into the ion source will be controlled by a Tylan FC-280A mass flow controller. This work will be carried out in the winter maintenance period of 1999.

5. Control system

All the newly installed devices — and some of the old ones, like the diffusion pumps of the cyclotron chamber — have been connected to the programmable logic controller (PLC) system based on the Mitsubishi Melsec A family. The control code was written in the Melsec logic symbolic language. It provides automatic and stand-alone operation of the vacuum system and the task of the operator has been simplified to just start and look after the required processes. A highly informative and easy-to-use graphical user interface (Fig. 1) has been developed to support this task, using the Paragon TNT process controller software package.

The modernized vacuum system of the cyclotron provides more reliable operation with reduced pumping down time. The duration required for changing the irradiation line has become significantly shorter and the same improvement can be expected for the gas change process as well. As a conclusion, the renewal project has resulted in a more flexible and easier usable/maintainable system.

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Activities at the Van de Graaff accelerator laboratory

L. Bartha and E. Somorjai

During 1998 the beam time of the VdG-1 machine amounted to 58 hours. The accelerator delivered proton beam on the 90° beamline for nuclear astrophysics (84%) and analytical studies (16%). The hollow-cathode ionsource produced H^- and H^+ ions on the main beamline in 80% and 20% of its 500 hours estimated beam time, respectively. These particles were used for low energy atomic physics experiments.

The 5 MV Van de Graaff machine was operating for 1607 hours during this period. Protons (67%), $^4He^+$ (26%) and $^{12}C^+$ (7%) were accelerated.

The beam time was distributed among different research subjects and education (laboratory practices for undergraduate and PhD students) as it is shown in Table 1.

Table 1: Time distribution amoung different research activities at VdG-5

	Hours	%
Atomic physics	162	10
Nuclear physics	43	3
Analytical studies	549	34
Analytics on the microprobe	692	43
Education	43	3
Machine tests	118	7
Total	1607	100

Improvements

A Bremsstrahlung-level measuring system has been put into operation close to the coronatriode of the VdG-5 machine for better observation of conditioning procedure of the acceleration tube.

The installation of the 90° astrophysical beamline of the VdG-1 machine has been completed.

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January 22
Does the principle of locality break down in quantum mechanics?
Gy. Bene (ELTE, Budapest)

January 29
Investigation of crystallic materials with combined channeling method
F. Ditrói

February 5
Test of QCD in high energy elementary particle collisions
Z. Trócsányi

February 13
Measurements with quadrupole mass spectrometers
S. Bohátka

February 19
Everyday quantum mechanics
Z. Máté

February 26
Systematics of energy shifts of multiple ionization KL X-ray satellites
I. Török

March 12
Dynamical symmetry breaking in weakly coupled superconducting systems
S. Mészáros

March 19
Dynamics of magnetic whirlpool system in superconducting BiSrCaCuO single crystalls
K. Vad

April 9
Transport of radon in fragmentary porous media - experimental studies in caves
J. Hakl

April 16
Anomalous intensity of the $K\beta_5$ X-ray line
I. Török

April 23
Modelling of valence shell photo electron spectrum of tetrahedral oxianions by the DV-Xalpha cluster molecule orbit method
I. Cserny

April 30
ASACUSA: Atomic physics at the CERN antiproton decelerator
D. Horváth

May 7
Antiprotonic atoms as probes of fundamental principles
J. Eades (CERN, Geneva)

May 14
Ionization processes in atomic collisions
Á. Kövér

May 21
The ^{40}Ar – ^{39}Ar method of geological dating
K. Balogh

May 28
Dimensional sampling and PIXE-analysis of atmospheric aerosoles
Zs. Kertész

September 8
Glueball spectrum from an effective Hamiltonian
P.O. Hess (UNAM Mexico – JLU Giessen)

September 17
Determination of SE2 for the $^{12}C(\alpha,\gamma)^{16}O$ reaction for carbon-burning in stars
J.L. Weil (University of Kentucky – Atomki)

October 29
Low energy nucleon–nucleon scattering using non-linear $SU(2) \times SU(2)$ Lagrangian
V. Lengyel (Uzhgorod University)
and
Relativistic approach for the problem of bound states of two-quark structures
S. Spenik (Uzhgorod University)

November 12
Low-energy scattering for four-body systems
S. Yakovlev (St. Peterbourgh State University)

November 19
Results and problems with scintillation detectors
G. Kalinka

November 26
Mineral waters of Hargita
Z. Makfalvi and P. Somay (Csíkszereda, Romania)
and
Study of radium content of waters of depth
I. Hunyadi

December 3
Hundred years of radium (Memory of Maria Skłodowska-Curie)
Sz. Mózsa (SOTE, Budapest)

December 10
New colleagues in Atomki

Author Index

(Surnames with comma: authors from other institutions)

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