# ATOMKI ANNUAL REPORT 1997





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



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NNUAL REPOR

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#### Preface

The year 1997 has seen vital decisions for the institutes of the Hungarian Academy of Sciences in the course of the 'consolidation' procedure started a year before. 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 financial measures are to be effected by setting up a guaranteed level of funding for each surviving institute, which is mostly higher than the funding during the 1995–1997 fast, and this level is to be reached by 1999. The extra funds contained in the state budget for stabilization are to be spent also on removals and other prompt costs implied by the ruling of the Stabilization Committee chaired by the President of the Academy.

To quote a few items of this ruling, it should be mentioned that, among the institutes of physics, the Research Institute for Technical Physics and the Research Institute for Materials Science have been merged to form a Research Institute for Technical Physics and Materials Science, and two of the four research institutes of natural sciences situated outside the capital have been handed over to universities, and a third one has lost its independence through merger with two Budapest institutes. This leaves ATOMKI as the only research institute for natural sciences outside the capital. (Life sciences are counted separately.)

The viability of this Institute as an independent institution has thus been acknowledged. It is also entirely in line with our own wish that the basic document of the consolidation emphasizes the importance of our close link with the universities of Debrecen. The document also states that we should coordinate our activity in particle physics and nuclear physics with that of the Research Institute for Nuclear and Particle Physics, Budapest. In fact, none of these imposes any restriction on our activity since in particle physics we co-operate with the Research Institute for Nuclear and Particle Physics at any rate, while in nuclear physics there is virtually no overlap between our fields of activity.

Our financial support has been very tight in 1997 as well; a first, though tiny, instalment of the budget increase, owing to the stabilization, came as a relief towards the end of the year. In our shortage of budgetary support and project funding it helped us to survive that we received a commission from IAEA to manufacture an irradiation and target transport system and an automated system for the radio-chemical separation of <sup>123</sup>I produced from <sup>123</sup>TeO<sub>2</sub> via (p,n) reaction. These devices are to be installed at a cyclotron of the Nuclear Research Centre of the Atomic Energy Authority, Egypt. We count as a success that, in a highly competitive scheme, we got a grant from OMFB (National Committee for Technological Development) to buy an escape-suppressed Ge "clover" detector for  $\gamma$ -ray spectroscopy for nuclear physics, nuclear astrophysics and analytical studies. This is the first substantial fund that can be used to buy research facilities for several years.

It belongs to the chronicle of the year that there were two international conferences in Debrecen organized by members of ATOMKI: the International Symposium on Exotic Nuclear Shapes, 12–17 May, and the 7th Joint Vacuum Conference of Hungary, Austria, Croatia and Slovenia, 26–27 May. It is also worth mentioning that this year the main subject of the Physics Week, which is organized by ATOMKI every March for the broad public, jointly with the local organization of the Eötvös Loránd Physical Society and the Kölcsey Centre of Culture, was the possibility of extraterrestrial life. Needless to say, the interest of the public was enormous.

We commemorated two anniversaries in 1997. At the end of June we celebrated the 80th birthday of the oldest surviving member of the Debrecen nuclear research school, Dr. László Medveczky, who was deputy director of the Institute in the sixties and seventies. In October we commemorated the 10th anniversary of the death of the founder of this Institute, Prof. Sándor Szalay. To remind the public of Debrecen of this remarkable scientist, we have put a memorial tablet on the outside wall of the main building of the Institute.

Last year a new nation-wide day of celebration was introduced by Government Decree, the Day of Hungarian Science, on 3rd November. The idea of such a festival may have been conceived in Debrecen, or at least a push was given to it here, by Ede Teller, who made such a proposal upon receiving an honorary degree at Kossuth University last year. The Institute celebrated the Day of Hungarian Science by an open lecture given by Prof. József Cseh, entitled "Jenő Wigner and sixty years of nuclear symmetries".

To mention some of the scientific feats, I now single out just one, which I think to be the most outstanding result. It was last year that our hunt for bands of hyperdeformed states in nuclear fission has concluded for the first nucleus considered,  $^{236}$ U. The nucleus  $^{235}$ U was bombarded by 10-MeV deuterons at the Debrecen cyclotron, and the reaction  $^{235}$ U(d,p) $^{236}$ U was generated. Outgoing protons belonging to fissioning final states were observed in coincidence with the fission fragments. The observed states of  $^{236}$ U form unresolved broad bumps, and they can be attributed to rotational bands. The moments of inertia extracted belong to axis ratio 3:1, which means hyperdeformation. The hyperdeformed states that can be reached as intermediate steps in fission have low spins, alternating parities, and can be interpreted as metastable states in the third dip of the potential acting between the fission fragments. The positions of the observed bands are fully consistent with the theoretically predicted states in the interfragment potential energy brought about by the nucleons in an adiabatic picture.

As a final remark, I should mention that, like last year, this Annual Report will be fully accessible through the WorldWide Web at http://www.atomki.hu. Its computerized version has been prepared on the basis of  $ET_EX$ , and one can get access to it in HTML, DVI, PDF and PS(.GZ) formats.

Debrecen, 9 April 1998

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#### **Organizational structure of ATOMKI**



#### Data on ATOMKI

#### Personnel

At present the Institute employs 214 persons. The affiliation of personnel to units of organization and the composition of personnel are given below.





#### Finance

In 1997 the total budget of the Institute was 435 million Hungarian for ints. The composition of the budget and the share of personnel expenditure within the budget are shown below.  $\ \cdot$ 



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

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Personnal Expenditure stec Non-Personne Expenditure 29%

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### Three-potential picture for the three-body Coulomb problem

#### (G. Leval, Papp J. Papp

During a scattering process the quantum mechanical system evolves from a state governed by the asymptotic Hamiltonian to the state described by the total Hamiltonian. In the two-potential formalism we define an intermediate Hamiltonian and connect the asymptotic Hamiltonian first to the intermediate Hamiltonians, and then to the total Hamiltonian. In the "three-potential" picture [1] a three-body Coulomb scattering process can be viewed as three consecutive scattering processes starting from the asymptotic channel Hamiltonian, via two intermediate Hamiltonians, to the total Hamiltonian. If we formulate this picture in terms of integral equations we get a set of the Faddeev and Lippmann-Schwinger integral equations. The integral equations have been solved for some cases in Coulomb-Sturmian space representation. The method performed well for bound states with repulsive [2] and with attractive [3] Coulomb interactions, and, in Ref. [1], excellent agreements with existing below-breakup p - d scattering calculations were reported.

In this "three-potential" formalism we have to split the Coulomb-like subsystem interaction into a sum of long-range and short-range terms, and the Faddeev procedure is applied only for the short range term. If the splitting has been performed in such a way that the long-range term does not support any bound states the solution for the auxiliary Hamiltonian which contains all the long-range terms can be given by one Lippmann-Schwinger equation. In a recent work [4] we have shown that the "three-potential" equations possess compact kernels for all energies, including above breakup energies, thus the equations are suitable for further numerical approximations, like separable expansion of the potential.

The derivation of this result is based on the assumption that the long-range term does not support any bound states. This kind of splitting can be reached easily for repulsive Coulomb interaction, one just should take the Coulomb interaction as the long-range term and the nuclear potential as the short-range one. The situation is more involved if some of the Coulomb interactions are attractive. Here the splitting into local terms is not possible. It seems now that one should split the Coulomb interaction into nonlocal terms.

- [1] Z. Papp, Phys. Rev. C 55, 1080 (1997). ggs [3] bus synold a isvel [2]
- [2] Z. Papp, and W. Plessas, *Phys. Rev.* C **54**, 50 (1996).
- [3] Z. Papp, *Few-Body Systems*, accepted for publication.
- [4] Z. Papp, Compactness of the set of Faddeev and Lippmann-Schwinger equations for the three-body Coulomb problem, to be published

### Exact analytical calculations for Natanzon-class potentials

G. Lévai, B. Kónya and Z. Papp

Local potentials have been used to describe the interactions of the subatomic world ever since the beginnings of quantum mechanics. Potential based quantum mechanical methods utilize analytical calculations, numerical techniques or the combination of these. Besides being an interesting area of mathematical physics, exactly solvable potentials thus form an important tool in the arsenal of quantum mechanical methods. Although the range of exactly solvable potentials has increased considerably, these new potentials are not used in realistic calculations.

Here we wish to advocate for the practical application of these potentials. The reason for this is that a large number of analytical results obtained for the simplest (shape-invariant) potentials (like the Coulomb, harmonic oscillator, Pöschl–Teller potentials etc.) can be generalized to the much wider class of Natanzon potentials. The example we consider is the generalized Coulomb potential, which is a simultaneous generalization of the Coulomb and harmonic oscillator potentials. Our work concerns the generalization of analytical results in the following areas [1]: S-matrix, Coulomb–Sturmian basis, matrix elements of the Green's operator and an SU(1,1) algebra associated with the problem. All these results have their equivalents for the Coulomb and the harmonic oscillator potentials, which can be recovered as special limiting cases.

The applications of this potential can be envisaged in fully or partly analytical calculations related to the Coulomb field of extended objects. Examples for this may be mesic atoms or atoms in strong magnetic field. Besides these practical aspects the generalized Coulomb potential allows reformulation of the Coulomb–oscillator connection and also helps the understanding of the peculiarities of the one-dimensional Coulomb potential.

There are also other examples confirming the expectations that seemingly complicated formulae can be evaluated analytically even for highly non-trivial potentials. In Ref. [2] for example, potentials phase equivalent with the Ginocchio potential have been determined analytically, using transformations of supersymmetric quantum mechanics.

- [1] G. Lévai, B. Kónya and Z. Papp, to be published.
- [2] G. Lévai, D. Baye and J.-M. Sparenberg, J. Phys. A 30 (1997) 8257.

### Green's matrix from Jacobi-matrix Hamiltonian

B. Kónya, G. Lévai and Z. Papp

Green's operators play a central role in theoretical physics, especially in quantum mechanics, since the fundamental equations are formulated as integral equations containing Green's operators in their kernels. The analytic calculation of the Green's operator in a particular basis representation, i.e. the Green's matrix, is of essential for asymptotically correct description of few-body scattering problems.

We have shown that, if the Hamiltonian in some basis representation appears in a symmetric infinite tridiagonal form with analytically known matrix elements, i.e. Jacobi-matrix form, then the corresponding Green's matrix can be constructed. We have found that in a certain domain of the complex energy plane the Green's matrix can be given by a continued fraction, which can be continued analytically to the whole complex plane. Thus, for constructing the Green's matrix only the Jacobi-matrix is required.

The procedure is illustrated with the non-trivial but calculable examples of the D-dimensional Coulomb Green's matrix and the D-dimensional harmonic oscillator.

[1] B. Kónya, G. Lévai, Z. Papp, J. Math. Phys. 38 (1997) 4833

tharmes (QCD) analysis group. Based on recent theoretical work of trocsanyl er of (see this Annual Report) we have started analysing multijet OPAL events in order to measure the strong coupling and test the symmetry structure of the underlying theory simultamenusly. These two analyses will be the topics of the PbD theses of Gabriella Pásztor and Beatrix Dienes.

The maintenance and operation of such an enormous detector as OPAL needs a lot of bandware and software work as well. Every participating institute is responsible for a subdetector in 1997 we participated in maintaining the so called Pressurple: Endeup drift chambers and also in building a scintifisting tile-wall to measure the timing of charged particles when they enter the lead glass electromagnetic calorimeter of the OPAL Endeup.

In 1997 we co-authored 27 journal publications and presented OPAL results at three international conferences. More information (with a tour of the OPAL detector and spectacular event pictures) can be found in the OPAL home page [1].

http://www.cern.ch/Opal

#### High energy physics studies with the OPAL detector at CERN

Beatrix Dienes,<sup>†</sup> Csaba Hajdu,<sup>‡</sup> Dezső Horváth, József Pálinkás, Gabriella Pásztor<sup>‡</sup> and Zoltán Trócsányi <sup>†</sup> Dept. Experimental Physics, Lajos Kossuth University, Debrecen, Hungary

<sup>‡</sup> Research Institute for Particle and Nuclear Physics, Budapest, Hungary

The OPAL (Omni-Purpose Apparatus for LEP, the Large Electron Positron collider at CERN) calorimeter is operated by a collaboration of 34 institutes of 7 countries. Our group joined the collaboration in 1995. Since then LEP increased the collision energy from 91 GeV, the Z<sup>0</sup> production peak, to 183 GeV where the pair production of two weak bosons,  $e^+ e^- \rightarrow W^+ W^-$  and Z<sup>0</sup> Z<sup>0</sup>, becomes energetically possible.

We participate in the analysis work of two OPAL working groups. The search for Higgs bosons, the last missing pieces of the menagerie of the Standard Model of electroweak interactions, is one of the most important task of present-day particle physics. As Higgs bosons were not observed yet, so far the result of our work was exclusion of mass ranges for Higgs bosons and thus limiting the parameter space available for the various models. The OPAL search for charged Higgs bosons is mainly based on the work of our group; with this technique we contributed to several other search efforts as well. In 1997 we joined the work of the Quantum Chromodynamics (QCD) analysis group. Based on recent theoretical work of Trócsányi *et al.* (see this Annual Report) we have started analysing multijet OPAL events in order to measure the strong coupling and test the symmetry structure of the underlying theory simultaneously. These two analyses will be the topics of the PhD theses of Gabriella Pásztor and Beatrix Dienes.

The maintenance and operation of such an enormous detector as OPAL needs a lot of hardware and software work as well. Every participating institute is responsible for a subdetector; in 1997 we participated in maintaining the so-called Presampler Endcup drift chambers and also in building a scintillating tile wall to measure the timing of charged particles when they enter the lead glass electromagnetic calorimeter of the OPAL Endcup.

In 1997 we co-authored 27 journal publications and presented OPAL results at three international conferences. More information (with a tour of the OPAL detector and spectacular event pictures) can be found in the OPAL home page [1].

[1] http://www.cern.ch/Opal

#### Perturbative quantum chromodynamics

Zsolt Molnár,<sup>†</sup> Zoltán Nagy<sup>†</sup> and Zoltán Trócsányi <sup>†</sup> Dept. Theoretical Physics, Lajos Kossuth University, Debrecen, Hungary

#### <sup>†</sup> Department of Physics, Niigata University, Japan

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 meaurement 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 made important progress in calculating the higher order corrections to this process. We

- presented a group indpendent color decomposition of the four-parton one-loop amplitudes in QCD and QCD + light gluino theory [1];
- 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 [2, 3];
- calculated the NLO corrections to five different four-jet event shape variables. In general we found large (more than 100%) higher order corrections [4, 5];
- calculated four-jet rates at NLO for various jet clustering algorithms. We found that the NLO prediction when matched to the next-to-leading logarithmic approximation describes the data obtained at LEP very well [4];
- calculated various angle distributions at NLO and found that the normalized angle distributions did not change their shape when going from leading to NLO [6];
- showed that the existence of light gluinos can be excluded comparing the matched NLO and next-to-leading logarithmic predictions to the four-jet data obtained at LEP using Durham clustering in the 0.002–0.004  $y_{cut}$  region [7];
- [1] Z. Nagy and Z. Trócsányi, Phys. Lett. B414 (1997) 187.
- [2] Z. Nagy and Z. Trócsányi, http://dtp.atomki.hu/HEP/pQCD.
- [3] Zsolt Molnár, diplom work, KLTE, 1997.
- [4] Z. Nagy and Z. Trócsányi, Phys. Rev. Lett. 79 (1997) 3604.
- [5] Z. Nagy and Z. Trócsányi, E-print hep-ph/9708344, to appear in Nucl. Phys.
- [6] Z. Nagy and Z. Trócsányi, E-print hep-ph/9712385.
- [7] Z. Nagy and Z. Trócsányi, E-print hep-ph/9707343.

## Structure and reactions of light exotic nuclei

## Y. Suzuki,<sup>†</sup> R.G. Lovas, Y. Yabana<sup>†</sup> and K. Varga<sup>†</sup> Department of Physics, Niigata University, Japan

With the advent of radioactive beams, light exotic nuclei have come to the focus of a great number of investigations. The most prominent phenomena found are the neutron halos: in some of these nuclei one or two of the most loosely bound neutrons occupy orbits that are much more extensive than the other orbits. The behaviour of these nuclei question the validity of all traditional basic concepts of nuclear structure: mean field, single-particle orbits, shape etc., which calls for an unrestrictive fully microscopic description.

We have developed a fundamental and yet feasible microscopic approach to the description of light exotic nuclei [1, 2], which has been successfully applied to quite a number of cases. It is a multicluster model combined with a stochastic variational method and an analytical calculational scheme, and, therefore, it may be called the correlated Gaussian stochastic variational approach. "Good" clusters are only included ( $\alpha$ -, triton, helion and "single-nucleon" clusters).

For light exotic nuclei the nuclear structure studies are inextricable from reaction studies. Some members of the cooperating team have contributed to the implementation of reaction theory to exotic nuclei. The basic tool is the Glauber approach. We are working on a comprehensive and didactically balanced review of the structure and reaction theory of light exotic nuclei. Our nuclear-structure approach is a good starting point for this purpose since in its most general form it is a general theory of bound many-body systems, so that any models used so far to describe these nuclei can be considered limiting cases of our multicluster approach or its macroscopic equivalent.

For the time being, this general structure theory has been written up. This is followed by a brief review of cluster models, and that paves the way before introducing the the multicluster approximation. That is the apt point to clarify the relationship of the microscopic multicluster approach to other microscopic approaches and to macroscopic models. The structure theory part is then completed with a review of calculations for actual cases.

K. Varga, Y. Suzuki and R.G. Lovas, Nucl. Phys. A571 (1994) 447–466
K. Varga and Y. Suzuki, Phys. Rev. C52 (1995) 2885–2905

### Numerically exact description of multicluster bound states

R.G. Lovas, K. Arai,<sup>†</sup>Y. Suzuki<sup>†</sup> and K. Varga

 $^\dagger$  Department of Physics, Niigata University, Japan

The most realistic picture of some of the light nuclei is a multicluster picture. A model based on such a picture involves a few-body problem with composite bodies. The few-body nature of the problem may be especially important for p-shell nuclei near the drip lines, where clustering may be strong and s.p. orbits ill-defined.

The solution of the few-body problem with composite bodies becomes feasible in a stochastic variational method with a generalized (so-called "correlated") Gaussian trial function [1]. Each such Gaussian may represent a partition of the particles.

We tested five models of <sup>6</sup>He with this approach: (i) a pure  $\alpha$ +n+n model with the  $\alpha$ -cluster in a 0s configuration; (ii) an  $\alpha$ +n+n model with breathing distortions of  $\alpha$ ; (iii) an { $\alpha$ +n+n; t+t} model, with breathing distortions of  $\alpha$ ; (iv) an { $\alpha$ (t+p)+n+n;  $\alpha$ (h+n)+n+n} model with breathing distortions of t and h; (v) an { $\alpha$ (t+p)+n+n;  $\alpha$ (h+n)+n+n; t+t} model [2]. The results are as shown below.

Mode	clusters, w	Editio	opic struct	6 concretes	account t	Ra	dii	plicitly
	Force I	Force II	Force I	Force II	Force	N	р	n n
(i)	-25.349	-24.909	-0.660	-0.220	I Band still	2.52	1.83	2.81
(ii)	-26.333	-25.914	-0.738	-0.319	I	2.49	1.82	2.77
(iii)	-27.013	-26.557	-1.418	-0.961	II	2.42	1.81	2.68
(iv)	-28.127	-27.699	-1.578	-1.151	II	2.35	1.76	2.60
(v)	-28.222	-27.794	-1.673	-1.245	II	2.34	1.76	2.58
Exp.	-29	.271	-0	.975	gate thes	2.33	1.72	2.59

<sup>6</sup>He binding energies E,  $\alpha$ +n+n separation energies  $\varepsilon$  (in MeV) and point nucleon (N), proton (p) and neutron (n) rms radii (in fm)

The  $\alpha$ -breakup deepens the energy much more than the  $\alpha$ -breathing. The overall underbinding is mostly due to the treatment of the t and h clusters. The extra state space included via the t+t partition is much the same as that included via the (t+p)+n+n and (h+n)+n+n partitions. Even the simplest model is satisfactory.

- K. Varga, Y. Suzuki and R.G. Lovas, Nucl. Phys. A571 (1994) 447–466; K. Varga and Y. Suzuki, Phys. Rev. C52 (1995) 2885–2905
- [2] R.G. Lovas, K. Arai, Y. Suzuki and K. Varga, Nuovo Cimento A, 1998

### Dynamic and discrete symmetries of nuclear cluster systems

G. Lévai and J. Cseh

Department of Physics, Niigata University, Jap

Nuclear clusterization can be analyzed in terms of various symmetry schemes, such as those based on dynamic and discrete symmetries. In the first case the interaction (i.e. the Hamiltonian) is thought to have symmetric nature, while in the second one the geometric arrangement of the cluster system is supposed to reflect some kind of discrete symmetry. In both cases the existence of the symmetry manifests itself in characteristic spectral patterns (band structure, etc.), which can be compared with observations.

We considered both symmetry-based approaches to interpret the gross features of the spectra of the <sup>12</sup>C and the <sup>16</sup>O nuclei as  $3\alpha$  and  $4\alpha$  systems [1]. Assuming that the <sup>4</sup>He clusters are in their ground states the excitations of the unified nucleus can be assigned to various rotational-vibrational modes of the  $\alpha$ -particles. These excitations can naturally be treated in terms of both the Semimicroscopic Algebraic Cluster Model (SACM) [2] and point groups [3]. The geometric configuration of the  $3\alpha$  and  $4\alpha$  systems can be identified as an equilateral triangle and a regular tetrahedron, corresponding to the point groups  $D_{3h}$  and  $T_d$ , respectively. The SACM implicitly takes into account the microscopic structure of the clusters, while in the discrete symmetry case they are considered structureless.

We found that the band structure predicted for <sup>12</sup>C by the dynamic symmetry of the SACM is fairly similar to that resulting from the geometric considerations. The situation is, however, quite different for <sup>16</sup>O: here the cluster bands have essentially different composition in the two schemes, and there are experimental data supporting both band assignments.

We plan to investigate these nuclei in terms of other algebraic models developed for the 3- and 4-body systems [4]. In these models further observables (like electromagnetic form factors) can be calculated, which might help our understanding of the structure of these nuclei.

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## Coexistence of cluster configurations in <sup>32</sup>S

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The energy spectrum and the electromagnetic transitions of the <sup>32</sup>S nucleus are described in terms of the Semimicroscopic Algebraic Cluster Model [1]. <sup>28</sup>Si +  $\alpha$  and <sup>16</sup>O + <sup>16</sup>O configurations are considered for the low-lying bands [2] and quasi-molecular resonances [3], respectively.

The cluster configurations, including their Hamiltonians, are treated in a unified framework. The densities of high-lying core-plus-alpha particle states are *predicted* in those energy windows where good-resolution experiments have been carried out [4]. The prediction is based on the concept of multichannel dynamic symmetry, which connects different channels, and consequently, different cluster configurations [5]. The multichannel symmetry requires an invariance with respect to the transformations from one cluster configuration to another one, i.e. from one set of Jacobian coordinates to another.

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## A symmetry inspired extension of the Weizsäcker mass formula to $\Lambda$ hypernuclei

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We introduced an extension of Weizsäcker's mass formula in order to give simultaneous description of the binding energies of normal and  $\Lambda$  hypernuclei [1, 2]. It is based on the symmetries of the nuclear many-body system, formulated in a unified way in Wigner's SU(4) spin-isospin scheme and in the SU(6) spin-flavour scheme of Gürsey and Radicati [3]. The symmetry considerations are built in through the use of the Majorana operator: this has minimal value in the ground state, in which the spatial symmetry of the nucleus is maximal due to the short-range attractive nature of the residual nuclear interaction. The mass formula containing also a strangeness term is then

$$B(N, Z, \Lambda) = a_{\rm v}A - a_{\rm s}A^{2/3} - a_{\rm c}\frac{Z^2}{A^{1/3}} - a_{\rm a}\frac{(N-Z)^2}{A} + a_{\rm y}\frac{\mathcal{S}}{A^{\gamma_{\rm m}}} + a_{\rm m}\frac{\langle M \rangle}{A^{\gamma_{\rm m}}} \,.$$

When applied to normal nuclei this formula reproduces the location of the valley of stability, the maximum in the binding energy per nucleon in the Fe–Ni region and nucleon separation energies. It also gives a smaller rms deviation for the 1909 known nuclear masses than the traditional Weizsäcker formula. We also used least-square fitting of the parameters to analyze the binding energies of 35 single and 3 double  $\Lambda$ hypernuclei separately and jointly with normal nuclei in various mass regions. We found that the parameters are rather close to those determined for normal nuclei [1]. Except for the lightest nuclei (for which the mass formula is not expected to work) the largest deviations in the binding energies occured near the shell closures both for normal and for hypernuclei. Shell effects, however, essentially cancelled out in the  $\Lambda$ separation energy  $B_{\Lambda} \equiv B(N, Z, 1) - B(N, Z, 0)$ . Our results also indicate that the Majorana term can be considered a sophisticated replacement of the pairing term.

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#### Binding energies of light nuclei

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The solution of the few-nucleon problem with a realistic nucleon-nucleon interaction including spin-orbit, tensor, etc. potentials is a real challenge in nuclear physics. In this work we extend the application of the stochastic variational method to noncentral interactions and we attempt to calculate the binding energies and other physical properties of the light nuclei. In our variational approach the trial function is a correlated gaussian multiplied by appropriate orbital and spin-isospin functions. The most adequate variational parameters and angular momentum channels are determined by gambling: random configurations are probed and the most suitable functions are chosen as basis functions [1, 2].

To test this approach we compared our results to those of the exact Greenfunction Monte-Carlo method (GFMC) [3]. The agreement, as is shown in the Table, is excellent. This agreement and the fact that the computational load is not too heavy, encourage the further applications of the stochastic variational method to light nuclei beyond A = 4.

Table 1: Energies (in MeV) and radii (in fm) of the alpha particle by different methods and with the AV8 interaction

$^{4}\mathrm{H}$	SVM	GFMC
$\langle T \rangle$	98.8	1)
$\langle V \rangle$	-124.4	-124.20(1.0)
$\langle V_{LS} \rangle$	-2.9	(6.6)
$\langle r^2 \rangle^{\frac{1}{2}}$	1.50	1.51(0.01)
E	-25.62	-25.75(0.05)

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## Exploration of resonances by analytic continuation in the coupling constant

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The energies and widths of resonance states are determined by analytic continuation of bound-state energies as functions of the coupling constants (potential strengths) [1]. The advantage of the method is that existing bound-state techniques can be used, without any modifications, to determine the positions of resonances. Various numerical examples show the applicability of the method to three-body (or three-cluster) systems, including the excited states of the <sup>6</sup>He and <sup>6</sup>Li [2]. The example in Table 1 compares the results of microscopic calculations using the analytic continuation of the coupling constant (ACCC) with those using the complex scaling method (CSM).

$^{6}\text{He}(2^{+},\text{T}=$	1) $(M, N)$	$E  [{\rm MeV}]$	$\Gamma [MeV]$
ACCC	(9,9)	0.73	0.07
$\rm CSM^{a)}$		0.74	0.06
Exp		$0.82 \pm 0.025$	$0.133 \pm 0.020$
$^{6}\text{Li}(0^{+},\text{T}=1)$	L)	8.86	12) 201
ACCC	(9,9)	0.21	0.003
$CSM^{a)}$		0.22	0.001
Exp	(10.0)16.1	-0.137	(*1)
$^{6}\text{Li}(2^{+}, T=1)$	l)	20.62	
ACCC	(9,9)	1.61	0.27
$CSM^{a)}$		1.59	0.28
Exp		$1.696 \pm 0.015$	$0.54 \pm 0.020$

Table 1: Comparison of resonance energy and width between ACCC and CSM. The energy is counted from the three-body threshold.

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### Unified description of light- and strange-baryon spectra

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There is a delicate question in low-energy QCD concerning the effective degrees of freedom that govern the properties of light and strange baryons. Beyond the limit of spontaneous breaking of chiral symmetry (SB $\chi$ S), the original QCD degrees of freedom—*current* quarks q and gluons g—are no longer adequate. Rather, as a consequence of the SB $\chi$ S, *constituent* quarks Q and Goldstone Bosons G appear and furnish the effective degrees of freedom in this energy domain. The constituent quarks bear dynamical masses related to the  $\langle \bar{q}q \rangle$  condensate and couple directly to the Goldstone bosons. Thus, light and strange baryons are to be considered as systems of three constituent quarks that interact by Goldstone-boson exchange (GBE) and are subject to confinement. We describe the properties of light and strange baryons in a semirelativistic constituent quark model whose hyperfine interaction relies on Goldstone-boson-exchange dynamics.

From the results [1] it is immediately evident that quite satisfactory description of the spectra of all low-lying light and strange baryons is achieved in a unified framework. In particular, the level orderings of the lowest positive- and negativeparity states in the nucleon spectrum are reproduced correctly, with the  $\frac{1}{2}^+$  Roper resonance N(1440) falling well below the negative-parity  $\frac{1}{2}^-$  and  $\frac{3}{2}^-$  states N(1535)and N(1520), respectively.

Likewise, in the  $\Lambda$  and  $\Sigma$  spectra the positive-parity  $\frac{1}{2}^+$  excitations  $\Lambda(1600)$  and  $\Sigma(1660)$  fall below the negative-parity  $\frac{1}{2}^--\frac{3}{2}^-$  states  $\Lambda(1670)-\Lambda(1690)$  and the  $\frac{1}{2}^-$  state  $\Sigma(1750)$ , respectively. In the  $\Lambda$  spectrum, at the same time, the negative-parity  $\frac{1}{2}^--\frac{3}{2}^-$  states  $\Lambda(1405)-\Lambda(1520)$  remain the lowest excitations above the  $\Lambda$  ground state. By the correct level orderings of the positive- and negative-parity states a long-standing problem of baryon spectroscopy has been resolved.

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#### Structure of the beryllium isotopes

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Nuclear mean field plays a basic role in nuclear structure by generating regular shell structure, but in light nuclei near the drip-lines it may not be stable enough to play such a role. Instead, correlation between the nucleons, particularly clustering, plays an important role in determining their structure. In this respect the light Be isotopes show various interesting properties, e.g., in binding mechanism, E1 transition and parity inversion. We attempt to describe the nuclei  $^{9-12}$ Be in a unified framework of two  $\alpha$ -particles and extra neutrons to see how well the structure of these isotopes can be understood. The dynamical treatment of these 3–6-cluster systems in this microscopic multicluster model is very challenging.

First the nucleus <sup>9</sup>Be was studied in an  $\alpha + \alpha + n$  three-cluster model. Both the ground state and resonance states were obtained fairly accurately. The data available for the magnetic and quadrupole moments and electron-scattering observables were reproduced well. The enhancement of the strong E1 transition was well accounted for by a drastic change of the valence neutron motion between the ground state and excited states. The analysis of the <sup>9</sup>Li  $\rightarrow$  <sup>9</sup>Be  $\beta$ -decay has demonstrated the distortion of the  $\alpha$ -particle. Its mirror nucleus, <sup>9</sup>B, which has no bound state, has also been studied in a similar  $\alpha + \alpha + p$  model, and the spins and parities of several states have been predicted.

Next we investigated the energy spectrum and other physical properties of <sup>10</sup>Be in a four-cluster model employing the same two-nucleon potential as in the A =9 system. The energy spectrum and the electromagnetic properties are in good agreement with experiment. It is very satisfactory that the second 0<sup>+</sup> state at 6.18 MeV can be reproduced very well. It naturally comes out that one neutron added to <sup>9</sup>Be makes <sup>10</sup>Be tightly bound in its ground state but allows the second 0<sup>+</sup> state to form a loose, spatially extended density distribution. The proton density at the centre decreases to almost half of the ground-state density. Since the second 0<sup>+</sup> state is thought to have a well-developed cluster structure of the type of <sup>6</sup>He +  $\alpha$ , we are calculating the amount of clustering and the weights of the components belonging to definite numbers of harmonic-oscillator excitation quanta to check this assumption.

The interest in <sup>11</sup>Be includes the anomalous level sequence of the ground state and first excited state and the strong E1 transition between them. These are related to the single-neutron halo structure of the system. No theory has yet been able to reproduce these features. We are currently investigating this nucleus in a microscopic  $\alpha + \alpha + n + n + n$  model. The reaction <sup>10</sup>Be $(n, \gamma)^{11}$ Be is interesting for astrophysics, and can also be studied by an extension of our model.

#### Isospin splittings of baryons

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We discuss the isospin-breaking mass differences between baryons, with particular attention, in the charm sector, to the  $\Sigma_c^+ - \Sigma_c^0$ ,  $\Sigma_c^{++} - \Sigma_c^0$ , and  $\Xi_c^+ - \Xi_c^0$  splittings.

A successful phenomenological description of the hadron spectrum has been obtained using non-relativistic potential models which tentatively simulate the lowenergy limit of QCD. Among the observables of interest, isospin-violating mass differences have received much attention. In general, the n - p,  $\Sigma^- - \Sigma^0$ ,  $\Sigma^+ - \Sigma^+$ ,  $\Xi^- - \Xi^0$  splittings of the nucleon,  $\Sigma$  and  $\Xi$  multiplets are well reproduced. These isospin splittings arise from several contributions, which largely cancel each other (kinetic energy, mass, potential, Coulomb interaction, etc.), so that each effect should be carefully computed, and even small terms should be incorporated.

We find that (see the Table), albeit there is a good agreement for light-quark baryons (including spin excitations), even an accurate variational treatment implementing all the realistic interactions does not permit to explain the data on  $\Sigma_c^+ - \Sigma_c^0$ in the framework of potential models based on one-gluon-exchange. A large  $\Xi_c^+ - \Xi_c^0$ is not really reproducible either. More precise data are, however, required before drawing conclusions.

Table 1: Comparison of calculated isospin-breaking splittings (in MeV) with experimental results.

Splitting	Exp.	BCN	AL1	AP1
n-p	1.293318	1.38	1.16	1.29
$\Delta^0 - \Delta^{++}$	$2.7\pm0.3$	3.21	2.20	6.10
$\Sigma^{-} - \Sigma^{0}$	$4.88\pm0.08$	6.62	5.16	6.07
$\Sigma^{-} - \Sigma^{+}$	$8.09\pm0.16$	11.96	8.25	10.57
$\Sigma^{*0} - \Sigma^{*+}$	-4 to 4	4.10	1.82	2.65
$\Sigma^{*-} - \Sigma^{*0}$	$2.0 \pm 2.4$	6.34	3.85	4.40
$\Xi^ \Xi^0$	$6.4 \pm 0.6$	10.62	7.12	9.19
$\Xi^{*-} - \Xi^{*0}$	$3.2\pm0.6$	5.87	3.68	3.58
$\Sigma_c^{++} - \Sigma_c^0$	$0.8 \pm 0.4$	0.12	1.06	2.91
$\Sigma_c^+ - \Sigma_c^0$	$1.4 \pm 0.6$	-0.96	-0.55	0.55
$\Xi_{c}^{0} - \Xi_{c}^{+}$	$2.5 \pm 1.7$	4.67	2.58	2.90
$\Xi'^{0}_{c} - \Xi'^{+}_{c}$	$1.7\pm4.6$	1.04	0.47	-0.22
$\Xi_{c}^{*0} - \Xi_{c}^{*+}$	$6.3 \pm 2.6$	0.40	0.44	-0.85

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#### Global vector representation

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The aim of this work [1] is to explore a new representation for the orbital part of the wave function of few-particle systems. Let us denote a set of Jacobi coordinate vectors of N particles by  $\mathbf{x} = (\mathbf{x}_1, \mathbf{x}_2, \dots, \mathbf{x}_{N-1})$ . One may assume that the spatial part of a general wave function can be expressed as a linear combination of products  $g(\mathbf{x})\theta(\mathbf{x})$  of a spherically symmetric  $g(\mathbf{x})$  and an orbital factor  $\theta(\mathbf{x})$ . The orbital part  $\theta(\mathbf{x})$  describes the angular motion of the particles. One can introduce a linear combination of the relative coordinate vectors, a "global vector":

$$\mathbf{v} = \sum_{i=1}^{N-1} u_i \mathbf{x}_i. \tag{1}$$

The orbital motion will have orbital angular momentum L if  $\theta(\underline{\mathbf{x}})$  is chosen to be

$$\theta_{KL}(\mathbf{v}) = v^{2K+L} Y_{LM}(\hat{\mathbf{v}}). \tag{2}$$

Any complicated angular motion can be described by a linear combination of such terms containing different vectors  $\mathbf{v}$  whose parameters  $u_i$  are to be determined variationally. The above form is simple and convenient in evaluating matrix elements.

The Table shows that one can attain the same accuracy with the global vector representation (GVR) as with a complicated partial wave expansion (PWE). This result encourages applications of the GVR for nuclear and atomic systems.

Table 1: Total energies (in MeV) of the ground states of the triton and of the alpha particle with the AV6 interaction. Only natural parity states have been taken into account for the orbital motion.

1.82	method	K	energy
triton	P6.0	2.4	± 0.8
21-2	GVR	100	-6.80
	PWE	100	-6.80
alpha	21.0	0.4	± 8.0
66.0-	GVR	300	-23.29
	PWE	300	-23.27
15.0	PO 1	C.P	王 \.1

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#### Shell corrections at the particle drip lines

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Continuum states of a single-particle potential affect significantly calculated shell corrections for nuclei close to the neutron or proton drip lines where the Fermi level approaches zero. In order to test the reliability of the standard shell correction method with continuum, a systematic comparison was carried out between the results of the Strutinsky procedure and that of the semi-classical Wigner-Kirkwood method for the spherical nuclei both along the  $\beta$ -stability valley and close to the particle drip lines. In the Strutinsky method the resonance contribution can be taken into account in a similar way as it is done for the bound states. Therefore it is extremely useful to separate the discrete contribution of the bound and resonant terms from that of the remaining continuum. This was done by determining a path of the complex energy plane which passes far from the singularities of the S-matrix. The smoothed level density  $\tilde{g}(E)$  is calculated by the aid of the Cauchy theorem as the sum of the residues of the poles of S(E) at the resonances encircled by the contour and a smooth background along the complex path. Since the plateau condition for the smoothed single-particle energy seldom holds, a new recipe is suggested for the definition of the shell correction. The new recipe uses the observed linear behavior of the Wigner-Kirkwood level density in a certain energy region and can be considered as a generalization of the Strutinsky smoothing procedure. The results of the generalized Strutinsky method are compared with the results of the semi-classical Wigner-Kirkwood expansion. A good agreement has been found for weakly bound nuclei in the vicinity of the proton drip line. However, some deviations remain for extremely neutron-rich systems due to the pathological behavior of the semi-classical level density around the particle threshold.

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## Influence of the continuum on the continuum on the continuum on two-particle two-hole excitations

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In this work we study 2p plus 2p2h excitations within the shell model (TDA approximation) by using the multi-step shell model method (MSM) [1] and including the continuum by means of a truncated Berggren representation [2] which includes all bound and certain resonant single particle states of a spherically symmetric Woods-Saxon potential. Resonant states (Gamow states) having larger imaginary parts than 200 keV are neglected together with the scattering states lying on the complex path.

High lying excitations consisting of particle-hole plus two-particle two-hole configurations coupled to the continuum are analyzed using this representation in <sup>208</sup>Pb. Partial decay widths are calculated and the effect of the two-particle two-hole excitations upon the widths are assessed. It is found that double giant resonance excitations are not mixed with other degrees of freedom and that double monopole giant resonances are likely to be detected in <sup>208</sup>Pb by measuring dilepton (E0) emission.

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semi-classical Wigner–Kirkwood expansion. A good agreement has been found for weakly bound nuclei in the vicinity of the proton drip line. However, some devia tions remain for extremely neutron-rich systems due to the pathological behavior of the semi-classical level density around the particle threshold.

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### Halo effect in the reaction ${}^{36}\mathrm{Ar}(\mathrm{p},\gamma){}^{37}\mathrm{K}$ and the astrophysical consequences

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The aim of this work is the exprimental determination of the halo properties of the  $1/2^+$  first excited state in  ${}^{37}$ K from the  ${}^{36}$ Ar(p, $\gamma$ ) ${}^{37}$ K reaction. The energy dependence of the branching ratio between the ground state  $(3/2^+)$  transition and the transition to the  $1/2^+$  first excited state is an experimental signature for these halo properties. Because of the small binding energy of the  $1/2^+$  state the bound state wave function has a very long tail, and at small energies the direct capture (DC) process can happen far outside the classical nuclear radius. This leads to an enhancement of the astrophysical S-factor at low energies, and therefore the branching ratio changes at low energies. The first experimental evidence of the halo effect was shown in a recent work [1].

For the determination of the branching of the DC process one has to measure the non-resonant DC cross section between the resonances of the reaction  ${}^{36}\text{Ar}(p,\gamma){}^{37}\text{K}$ . It is necessary to know the resonance properties very accurately for finding an energy region where the non-resonant DC process is dominating. In the first experiment we tried to measure the low-energy resonances.

The experiment was carried out at the Van de Graaff accelerator in ATOMKI, Debrecen. Implanted <sup>36</sup>Ar (into Ta-backing) target has been used. The known resonances [2, 3, 4] at  $E_{p,lab} = 321 \text{ keV} (3/2^{-})$  and  $E_{p,lab} = 918 \text{ keV} (5/2^{+})$  in the <sup>36</sup>Ar( $p,\gamma$ )<sup>37</sup>K reaction have been found. However, the 7/2<sup>+</sup> state in <sup>37</sup>K which corresponds to a resonance energy of  $E_{p,lab} = 440 \text{ keV}$  could not be found, and we can give only an upper limit for the yield of this resonance. According to the preliminary analysis, our upper limit is better than the existing value [4]. Higher number of implanted atoms in the target, a low background  $\gamma$ -ray detector and higher beam intensities are needed for getting measurable resonance (at  $E_{p,lab} = 440 \text{ keV}$ ) and DC yields. The work for fulfilling these demands as well as the analysis of the experimental data are in progress.

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#### Lifetimes of <sup>32</sup>S levels<sup>\*</sup>

Zs. Fülöp, Gy. Gyürky, Á.Z. Kiss, E. Somorjai, A. Kangasmäki,<sup>1</sup> P. Tikkanen,<sup>1</sup> J. Keinonen,<sup>1</sup> W.E. Ormand<sup>2</sup> and S. Raman<sup>3</sup>

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Mean lifetimes of 20 out of 31 bound levels in <sup>32</sup>S below an excitation energy of 8.0 MeV are deduced from the Doppler-broadened  $\gamma$ -ray line shapes produced in the reactions  ${}^{2}\text{H}({}^{31}\text{P}, n\gamma){}^{32}\text{S}$ ,  ${}^{28}\text{Si}({}^{6}\text{Li}, pn\gamma){}^{32}\text{S}$ , and  ${}^{31}\text{P}(p, \gamma){}^{32}\text{S}$ . The experiments with heavy ions and protons were carried out at the tandem accelerator in Helsinki and at the 5 MV Van de Graaff accelerator in Debrecen, respectively.

Of the 20 levels, lifetimes for 4 are reported here for the first time. The results based on reliable stopping powers in the DSA analysis and realistic MC simulations of the experimental conditions remove the large uncertainty of the lifetime values of excited <sup>32</sup>S levels reported in the literature [1]. The low-lying portion of the level scheme, level lifetimes,  $\gamma$ -ray branchings, E2/M1 mixing ratios, and reduced transition probabilities are compared with shell-model calculations.

. The overall agreement is good. A one-to-one correspondence between 33 experimental and predicted states is established up to 8.4 MeV for both positive- and negative-parity states. Shell-model calculations are able to reproduce the measured lifetimes to a reasonable degree. Also, the electromagnetic transition strengths are in most cases predicted correctly.

A more detailed comparison requires additional information such as unambiguous  $J^{\pi}$  and T assignments and data on E2/M1 mixing ratios.

\* Submitted for publication to Phys. Rev. C

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#### Spectroscopic study of <sup>65</sup>Ga

I. Dankó, D. Sohler, Zs. Dombrádi, B.M. Nyakó and the NORDBALL collaboration

The neutron-deficient <sup>65</sup>Ga nucleus was investigated via the <sup>12</sup>C(<sup>58</sup>Ni, $\alpha$ p)<sup>65</sup>Ga heavy-ion reaction. The experiment was performed at the Tandem Accelerator of the Niels Bohr Institute in Riso, Denmark. For detection of the evaporated particles and  $\gamma$  rays we used the NORDBALL detector array equipped with charged-particle and neutron detectors to select different reaction channels. Numerous new  $\gamma$  rays were assigned to <sup>65</sup>Ga. The angular distributions of the transitions were analyzed to obtain multipolarity information. The <sup>65</sup>Ga  $\gamma$  rays are marked in the spectrum shown in Fig. 1, which was obtained by subtracting contaminating reaction channels.





 D. Sohler et al., in Proc. 9<sup>th</sup> Int. Symp. on Capture Gamma-Ray Spectroscopy, eds. G. Molnár, T. Belgya and Zs. Révai (Springer, Budapest, 1997) p. 262.

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#### Study of structure of <sup>67</sup>Ga

#### I. Dankó, D. Sohler, Zs. Dombrádi, B.M. Nyakó and the NORDBALL collaboration

The structure of the <sup>67</sup>Ga nucleus was studied via <sup>12</sup>C(<sup>58</sup>Ni,3p $\gamma$ )<sup>68</sup>As reaction with the NORDBALL detector array. The level scheme constructed on the basis of the  $\gamma\gamma$ -coincidence relations was extended up to 10 MeV excitation energy. In order to assign the spin and parity values to the excited states the angular correlation ratios of the stronger transitions were analyzed. The highest spin parity values obtained were J<sup> $\pi$ </sup> = 33/2<sup>+</sup> and (27/2<sup>-</sup>) for positive and negative parity levels, respectively. The proposed level scheme of <sup>67</sup>Ga is shown in Fig. 1.

67Ga



Fig. 1: Proposed level scheme for <sup>67</sup>Ga

D. Sohler et al., in Proc. 9<sup>th</sup> Int. Symp. on Capture Gamma-Ray Spectroscopy, eds. G. Molnár, T. Belgya and Zs. Révai (Springer, Budapest, 1997) p. 262.
### Structure of <sup>67</sup>Ge

#### D. Sohler, Zs. Dombrádi, B.M. Nyakó and the NORDBALL collaboration

As a continuation of our study of the Ge isotopes [1] the level scheme of <sup>67</sup>Ge have been significantly extended up to about 9 MeV and to  $J^{\pi} = 31/2^{(-)}$  for negative parity and  $(29/2^+)$  for positive parity levels. The structure of the studied nucleus was discussed in the framework of the interacting boson-fermion model extended by inclusion of a broken pair (IBFBPM). In Fig. 1 we present the high-spin states close to the yrast calculated in IBFBPM in comparison to available experimental data shown by full circles. The IBFBPM states having largely one-quasineutron coupled to bosons (referred to as  $1\nu$ -states), threequasineutron coupled to bosons (referred to as  $1\nu 2\pi$ -states) are presented by squares and triangles, respectively.



Fig. 1: Comparison of the experimental high-spin states of <sup>67</sup>Ge with the theoretical energy spectra calculated in the framework of the interacting boson-fermion model extended by inclusion of one broken pair

 D. Sohler et al., in Proc. 9<sup>th</sup> Int. Symp. on Capture Gamma-Ray Spectroscopy, eds. G. Molnár, T. Belgya and Zs. Révai (Springer, Budapest, 1997) p. 262.

### Structure of <sup>72</sup>As

#### D. Sohler, Zs. Dombrádi, Zs. Podolyák, A. Algora and the NORDBALL collaboration

The <sup>72</sup>As nucleus was studied through  $(\alpha, n\gamma)$  reaction at 14.2 MeV bombarding energy [1]. A new level scheme was constructed on the basis of  $\gamma\gamma$ -coincidence measurement. Several new internal conversion coefficients of <sup>72</sup>As transitions were determined from internal conversion electron spectra, making possible unambiguous parity assignments also for higher-spin levels, resulting in definite parity assignment to the bands previously observed.

Using the new spin-parity values and the  $\gamma$ -ray branching ratios, the states could be grouped into proton-neutron multiplets. The multiplet assignments are presented in Fig. 1.



Fig. 1: Experimental proton-neutron multiplet states in <sup>72</sup>As. The left-side and the right-side columns are the negative and positive parity levels, respectively. The abscissa is scaled according to J(J+1), where J is the spin of the state.

 D. Sohler et al., in Proc. 9<sup>th</sup> Int. Symp. on Capture Gamma-Ray Spectroscopy, eds. G. Molnár, T. Belgya and Zs. Révai (Springer, Budapest, 1997) p. 270.

# $^{144}$ Sm $(\alpha, \gamma)$ cross section at astrophysically relevant energies

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As a stringent test of statistical model calculations used in extended reaction networks to follow the synthesis of p-nuclei we have measured the cross sections of  $^{144}Sm(\alpha, \gamma)^{148}Gd$  reaction at 10.5 MeV  $< E_{\alpha} < 13.4$  MeV. The resulting cross sections and astrophysical S-factors are compared to new statistical model calculations. The measured data on  $^{144}Sm$  at low energies reflect the strong energy dependence of the optical potential which is found to affect significantly our predictions of the  $\alpha$ -capture rate [1].



Fig. 1: The astrophysical S-factor for  $^{144}Sm(\alpha, \gamma)^{148}Gd$ 

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- [2] P. Mohr et al., Phys. Rev. C55 1523 (1997).

### Fission fragment mass distribution of the hyperdeformed states in <sup>236</sup>U

#### M. Hunyadi, A. Krasznahorkay, M. Csatlós, J. Gulyás and Z. Máté

Recently we have identified some hyperdeformed bands in  $^{236}$ U [1] by their large moments of inertia. Another signature of these states could be their highly characteristic fission fragment mass distribution predicted by Ćwiok et al. [2]. In order to demonstrate an enhanced mass yield for masses around 100 and 134 we performed a preliminary time-of-flight correlation measurement for the fission fragments using  $^{235}$ U(d,pf)<sup>236</sup>U reaction.

The experimental set-up consisted of two  $166 \times 166 \text{ mm}^2$  area position sensitive fission counters equipped with delay-line read-out. They were placed opposite each other at  $\Theta_1 = 64^\circ$  and  $\Theta_2 = 116^\circ$  to the beam line facing opposite sides of the target at a distance of 32 cm. A four-fold Si-detector array measured the energy of the outcoming protons, and was placed at a backward angle of  $\Theta = 150^\circ$ .

The data acquisition system recorded the TDC signals of the time-correlated fragments and the ADC signals of the protons. The TDC data measures the TOF between the proton and fragment events. The excitation energy of the prefission nucleus was determined by the energy of the protons. The mass distribution can be deduced from the TOF-data of the (p-f-f) triple coincidence events using the linear momentum conservation and the assumption of negligible neutron evaporation. The Fig. 1.a shows the TOF-correlation between the fragments of the same fission event. In Fig. 1.b preliminary mass distributions of both the whole and a sharper  $E^* = 5.2 - 5.4$  MeV excitation energy range around the hyperdeformed resonances are represented. On the basis of these results we are going to greatly improve the determination of the mass distribution, and thereby prove the expected sharpening of the distribution relative to that of prompt fission.



Fig. 1: a) TOF-correlation of the fission fragments; b) Mass distributions of the whole and a gated ( $E^* = 5.2 - 5.4$  MeV) excitation energy range.

- [1] A. Krasznahorkay et al., Phys. Rev. Lett. 80 (1998) 2073
- [2] S. Ćwiok et al., Phys. Lett. B322 (1994) 304

## The study of <sup>238</sup>U beta-band structure using enhanced E0 transitions

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Both the  $\beta$  band and the ground state band of a rotation-vibration nucleus have quantum number K=0, and hence the E0 transitions between states of the same J will be strongly enhanced, especially in nuclei with high Z [1]. This fact has been used to study the structure of two potential  $\beta$ -bands in the nucleus <sup>238</sup>U [2]. The internal conversion electrons corresponding to the E0 transitions were detected with a super-conducting electron spectrometer in the ATOMKI laboratory following excitation of the nuclei in a thin <sup>238</sup>U target by incident 19 MeV alpha particles. A spectrum of these electrons is shown in Fig. 1. The large continuum background is



Fig. 1: An electron spectrum from the excitation of  $^{238}$ U nuclei by 19 MeV  $\alpha$  particles

due to internal pair production resulting from the decay of the 6.05 MeV 0<sup>+</sup> state in an oxygen contaminant in the target. Nevertheless, the strong E0 transitions from <sup>238</sup>U stand out clearly in the spectrum, and are labeled by the spin. The well-defined shoulder due to the 0<sup>+</sup>  $\rightarrow$  0<sup>+</sup> transition verifies the suspected existence of the 0<sup>+</sup><sub>3</sub> level at 997 keV, and gives a very good measure of the level energy, which is needed to determine the moment of inertia. The relative weakness ( $\approx \times 0.1$ ) of transitions from the suspected  $\beta$  band headed by the 0<sup>+</sup><sub>2</sub> level at 926 KeV is an indication of the very different character of this band, which is at present not understood. Further measurements need to be made with a target having a much smaller oxygen contamination than was present in the specially prepared target used in these measurements [3], to set a confident upper limit on the E0 transition strength of the members of the 926 keV band, as well as to look for enhanced transitions from higher lying bands.

This work has been supported by the Hungarian OTKA Foundation, No. T23163 and the Nederlandse Organisatie voor Wetenschapelijk Onderzoek (NWO).

- [1] E.L. Church and J. Weneser, Phys. Rev. 103 (1956) 1035
- [2] J.M. Hoogduin et al., Phys. Lett. B34 (1996) 43
- [3] Oxygen free target supplied by the target lab at GSI.

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### Search for two-phonon superdeformed states in <sup>240</sup>Pu

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The study of multiphonon states has become the center of interest again. In this work, states predominantly of  $\beta$  vibrational type (i.e. the stretching mode, which leads to fission) have been studied by measuring the fission probability as a function of the excitation energy. The levels in the first well were populated by the <sup>239</sup>Pu(d,p) reaction. We expected enhanced fission probability (transmission resonances) for those excitation energies in the first well which coincide with vibrational states in the second well.

The experiment has been carried out at the Munich Tandem Accelerator at  $E_d = 13$  MeV. The enriched (99.9 %) <sup>239</sup>Pu target was produced in the recently finished radioactive target laboratory in Garching. The energy of the protons was analyzed by a Q3D magnetic spectrometer ( $\Theta_{LAB} = 130 \text{ deg}$ ) equipped with a 1.8 m light-ion focal-plane detector, in coincidence with the fission fragments detected by two position-sensitive avalanche detectors (PSAD) using two wire planes (with delay-line read-out) corresponding to horizontal and vertical directions. A preliminary proton energy spectrum (containing only a small fraction of the whole statistics) is shown in Fig. 1. as a function of excitation energy.



Fig. 1: a) Proton energy spectrum in coincidence with the fission fragments as a function of the excitation energy, b) potential energy curve for <sup>240</sup>Pu as a function of the quadrupole deformation parameter  $\beta_2$  [1]. The shadowed excitation energy region was presently investigated.

The two highly damped vibrational resonances with centroids of 4.5 and 5.1 MeV, which were previously known [2], have been nicely resolved into substates corresponding to SD states in the second well of the potential barrier. Information on the spins of the resonances will be obtained by comparing the measured fission fragment angular distributions with the results of PWBA calculations.

This work has been supported by the Hungarian OTKA Foundation No. T23163, and the DFG under IIC4-Gr 894/2.

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## **Atomic Physics**

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### Electron capture to the continuum induced by dipolar force

L. Sarkadi and K. Tőkési berez I. 1968

The process of *electron capture to the continuum* (ECC) leading to cusp (a singularity in the energy spectrum of the forward ejected electrons in atomic collisions) is well understood for the case when the interaction between the outgoing projectile and the electron is the long-range Coulomb force. In the past few years we carried out systematic investigations to understand the cusp electron production for collisions where the outgoing projectile is a *neutral* atom [1, 2], i.e. the interaction is characterized by short range.

One of our recent experiments convincingly proved that for He<sup>0</sup> projectile the ECC cusp is formed via the excitation of a low-lying virtual state of the intermediate He<sup>-</sup> ion at the 2<sup>1</sup>S He threshold [3]. There is no such explanation for the simplest neutral atomic projectile, H<sup>0</sup>. In this case one may assume that the ECC cusp is induced by a *dipolar* force between the outgoing H<sup>0</sup> and the ejected target electron. Dipolar interaction may arise if the outgoing H<sup>0</sup> is in an excited state. For example, for the n = 2 excitation the collisional mixing of the degenerated 2s and 2p states in H<sup>0</sup> may lead to a permanent electric dipole moment. An indication for the existence of the dipolar cusp was found in studies of the single-electron detachment from H<sup>-</sup> [4].

In the present work we made classical trajectory Monte Carlo (CTMC) calculations for the ECC cusp assuming dipolar interaction between the projectile and the target electron. Our question was whether the range of the interaction is long enough for cusp production in this case. Particularly, our aim was to explain the surprising experimental observation that the ECC cusp obtained by  $H^0$  impact was more intense than that obtained by proton impact [2].

The calculations were made in the three-body approximation. The target was hydrogen, the collision energy was 25 keV. The projectile consisted of a proton and an electron separated by a fixed distance, d. The orientation of the dipole was also fixed. The cusp formation was studied as a function of the dipole length and the angle of the orientation. A very intense cusp — in agreement with the experiment — was observed when the proton was the leading particle in the dipole. The maximum cusp intensity was found at  $d \approx 3$  a.u. From the analysis of the trajectories we concluded that the main cusp production mechanism is not the "Coulomb focusing" of the ejected electron by the projectile, as it was expected. Typical collision events leading to cusp are such that the target electron is captured into very weakly bound states around the dipole. Most of these states have negative energies because of the proximity of the ionized target (two-center orbits). When the collision partners separate from each other, the electron becomes free. Such electrons have very small kinetic energy in the projectile reference system, therefore in the laboratory system they appear as a cusp.

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is well understood for the case when the interaction between the outgoing projectile and the electron is the long-range Coulomb force. In the past few years we carried out systematic investigations to understand the cusp electron production for collisions where the outgoing projectile is a *neutral* atom [1, 2], i.e. the interaction is characterized by short range.

One of our recent experiments conviacingly proved that for  $\mathbb{N}^{0}$  projectile the ECC cusp is formed via the excitation of a low-lying virtual state of the intermediate  $\mathbb{H}^{-1}$  ion at the 2<sup>1</sup>S He threshold [3]. There is no such explanation for the simplex neutral atomic projectile,  $\mathbb{H}^{-1}$ . In this case one may assume that the ECC cusp is induced by a *dipolar* force between the outgoing  $\mathbb{H}^{0}$  and the globed ruget electron. Divolar interaction may arise if the outgoing  $\mathbb{H}^{0}$  is in an excited state. For example,  $\mathbb{H}^{0}$  the n = 2 excitation the collisional mixing of the aggenerated  $\mathbb{H}^{0}$  exites is of the outgoing  $\mathbb{H}^{0}$  is in an excited state. For example, for the n = 2 excitation the collisional mixing of the aggenerated  $\mathbb{H}^{0}$  is the existence  $\mathbb{H}^{0}$  may be detection for the collisional mixing of the single-electron for the existence of the dipolar cusp was found in studies of the single-electron detacliment from  $\mathbb{H}^{0}$ .

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The calculations were made in the three-body approximation. The target was hydrogen, the collision energy was 25 keV. The projectile consisted of a proton and an electron generated by a fixed distance, d. The orientation of the dipole was also fixed. The cusp formation was studied as a function of the dipole length and the angle of the orientation. A very intense cusp — in agreement with the experiment was observed when the proton was the leading particle in the dipole. The maximum cusp intensity was found at  $d \approx 3$  a.u. From the analysis of the trajectories we concluded that the main cusp production mechanism is not the "Coulomb focusing" of the ejected electron by the projectile, as it was expected. Typical collision events tates around the dipole. Most of these states have negative energies because of the proximity of the ionized target (two-center orbits). When the collision partners the proximity of the ionized target (two-center orbits). When the collision partners the proximity of the ionized target (two-center orbits). When the collision partners here in each other, the clectron becomes free. Such electrons have very small the proximity of the ionized target (two-center orbits). When the collision partners apparate from each other, the clectron becomes free. Such electrons have very small they appear as a cusp.

## Collisional excitation of Feshbach resonances in H<sup>-</sup> below the H(n = 3) and H(n = 4) thresholds

### F. Penent,\* J.-P. Grouard,\* J.-L. Montmagnon\* and L. Sarkadi \* D.I.A.M., Université P.&M. Curie, Paris, France

[K. Bhatia and Y.K. Ho, Phys. Rev. A 50 (1994) 4880

The collisional electron detachment from  $H^-$  is a fundamental process. The study of the resonant and non-resonant electron emission from the simplest negative ion yields important information about the dynamics of the collision and the structure of  $H^-$ . In our former works [1] we studied the process applying zero-degree electron spectroscopy. This method allowed us to get information on ejection of extremely low-energy electrons (in the projectile reference frame). We could resolve the dominant structures characterizing the spectrum of the detached electrons: the lines from the  $(2s2p)^1P^0$  shape resonance and the cusp peak. However, we could not exclude the contribution of other unresolved shape resonances, like the theoretically predicted  $^3D^0$  resonance that lies above the H(n = 3) threshold by 1.5 meV [2].

The question about the existence of the above  ${}^{3}D^{o}$  resonance has motivated us to look also for other resonances of H<sup>-</sup> near the H(n = 3) threshold. Most of the known resonances of H<sup>-</sup> were observed in electron on hydrogen collisions as well as in photodetachment of  $H^-$ . In these experiments mainly the *excitation* of the various resonance states was established without any further information on the decay of the resonances. In the present experiment we detected the auto-detached electrons, i.e. the decay products of the resonances. This allowed us, in many cases for the first time, to determine branching ratios of the decay of the resonances towards all opened continua. Significant is the finding, for example, that we observed the decay of the  $(2s2p)^1 P^o$  shape resonance (lying above the H(n=2) threshold by 20 meV) to the ground state of the hydrogen atom. Another important feature of the present experiment is that the spectrum of the auto-detached electrons is superimposed on a large background due to the directly detached electrons. The *interference* between the amplitudes of the resonant and non-resonant electron detachment leads to rapidly changing Fano-profile shape of the peaks in the spectrum as a function of the parameters of the collision. We found a strong dependence of the interference on the target in collisions of 6 keV H<sup>-</sup> ions with rare gas atoms. The peaks belonging to the <sup>1</sup>S, <sup>3</sup>P, <sup>1</sup>D, <sup>1</sup>P, <sup>3</sup>F Feshbach resonances below the H(n = 3) threshold showed very pronounced interference effects for He and Ne target, but not for Ar target. We should mention that due to the very small signal-to-noise ratio of these measurements, good statistics could be achieved only applying position sensitive electron detection technique. This explains why these resonances were not observed before. The improved detection technique made it possible to see even the very weak Feshbach resonances lying below the H(n = 4) threshold.

To get more detailed information on the excitation of the H(n = 3) Feshbach resonances, in 3 keV H<sup>-</sup> on Ne collisions we detected the electrons coming from the decay to H(n = 2) in coincidence with the Lyman- $\alpha$  photons emitted from H(2p). In this way we could separate the H(2s) and H(2p) decay channels, as well as suppress the background associated with the direct electron detachment leading to H(1s).

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To get more detailed information on the excitation of the H(n = 3) resubach resonances in 3 keV H<sup>-</sup> on Ne collisions we detected the electrons coming from the

# Existence of the electron capture to continuum process at positron impact

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The electron capture to continuum process (ECC) is well-known in ion-atom collisions. It produces a cusp like peak in the electron spectrum at zero degree at the energy where the velocity of the electron matches the velocity of the projectile. For positron impact this process has not been observed until now.

In the last few years we have investigated the double differential electron spectrum ejected from 100 eV e<sup>+</sup> – Ar collisions at different ejection angles (0°, 30°, 45°, 60°) to find the ECC cusp. However, the spectra around 42 eV were smooth inside the error bars, at every angle in good agreement with the results presented with the CTMC method [1, 2, 3].

Recently we developed an experimental set-up to detect the ionised electron in coincidence with the scattered positron resulting from 100 eV  $e^+$  – H<sub>2</sub> collision system into 0°, in order to determine the triple differential cross section. The time spectra (the intensity as a function of the time difference between the detected electron and positron) were recorded for several electron energies around the ECC cusp energy (42 eV). The figure shows the existence of the ECC process at positron impact. In the time spectrum measured at 41 eV a definite coincidence peak appears. The TDCS spectrum has been investigated in the 25–55 eV electron energy range [4].



Fig. 1: ide kene irni

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### The O and C K-VV Auger-electron spectra from CO<sub>2</sub> molecule induced by 1.0 MeV proton impact

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The angular distribution of the KVV Auger-lines give useful information on the wave functions of the molecules and the orientation of the processes in ionmolecule collisions. In our previous work [1] we observed significant anisotropy of the molecular Auger-line in proton $-N_2$  collisions, indicating that the ionization probability of the molecular core depends on the direction of the internuclear axis, relative to the ion beam. Recently we extend our study to determine the angular distribution of Auger transition for  $CO_2$  target, which is a linear heteronuclear molecule.

The Auger-electron spectra were collected at 13 different angles (from  $0^{\circ}$  to 180°), simultaneously with a triple pass high-resolution electrostatic electronspectrometer [2].



Fig. 1: Oxygen K-LL Auger spectrum of carbon dioxide. The points are the measurement and the solid line is the result of the fit.

Fig. 1 shows the measured oxygen KVV Auger spectrum at  $135^{\circ}$  relative to the direction of the proton beam. The identification of the Auger-transition were based

on the assignment of Moddeman et al. [3] and Agren [4] papers. The detailed analysis of the Auger spectrum at different angles are in progress. The preliminary data show a slight anisotropy for the oxygen KVV Auger emission of the carbon dioxide molecule.

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 $^{\ast}$  On leave from the Institute of Physics, P.O. Box 57, 11001 Belgrade, Yugoslavia.

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were investigated by incashing in antoionate and position in Berlin. A unique 90 eV [1]. This work was done at the lact that both single and double K excitation, feature of using Li as a target is the fact that both single and double K excitation, and the ratio of their cross sections, can be obtained from a single ejected-electron spectrum permitting an accurate determination of the double-to-single K excitation ratio.



Fig. 1: Differential electron emission cross sections for 10.6 MeV/u N'  $\pm$  Li collisions

Typical electron emission spectra for several ejection angles are shown in Fig. 1. Super-imposed on a monotonically decreasing distribution of continuum electrons. Auter lines corresponding to single K excitation are observed in the large peak

### Double K excitation of Li by 10.6 MeV/u N<sup>7+</sup> projectiles

J.A. Tanis,<sup>1</sup> J.-Y. Chesnel,<sup>2</sup> F. Frémont,<sup>2</sup> M. Grether,<sup>3</sup> B. Skogvall,<sup>4</sup> B. Sulik, M. Tschersich<sup>4</sup> and N. Stolterfoht<sup>3</sup> <sup>1</sup> Western Michigan University, Kalamazoo, Michigan 49008 USA <sup>2</sup> Laboratoire de Spectroscopie Atomique, F-14050 Caen Cedex, France <sup>3</sup> Hahn-Meitner Institut, Glienickerstrasse 100, D-14109 Berlin, Germany

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Single and double K excitation of Li by 10.6 MeV/u N<sup>7+</sup> (v/c = 0.15) projectiles were investigated by measuring Li autoionization spectra in the energy range 50– 90 eV [1]. This work was done at the Ionenstrahl Labor (ISL) in Berlin. A unique feature of using Li as a target is the fact that both single and double K excitation, and the ratio of their cross sections, can be obtained from a single ejected-electron spectrum permitting an accurate determination of the double-to-single K excitation ratio.



Fig. 1: Differential electron emission cross sections for 10.6 MeV/u  $N^{7+}$  + Li collisions

Typical electron emission spectra for several ejection angles are shown in Fig. 1. Super-imposed on a monotonically decreasing distribution of continuum electrons, Auger lines corresponding to single K excitation are observed in the large peak centered near 55 eV, while the Auger peaks in the range 65-90 eV are due to the doubly-K-excited states.

From these data, the ratio of double-to-single K excitation was determined to be 0.36% independent of ejection angle. This value is six times larger than the calculated ratio based on independent interactions between the projectile nucleus and each of the Li target electrons, i.e., TS2. This result suggests that the electron– electron interaction dominates double K excitation in this charge and velocity regime (Z/v = 0.34). To give further insight into the underlying excitation mechanisms, high-resolution measurements of the double K excitation region are planned to identify which intermediate configurations populate the double-K-vacancy states.

This work was supported by the Hungarian–German Intergovernmental Collaboration in Science and Technology (No. B/129), by the Hungarian National Science Fund (OTKA-3011) and by the U.S. Department of Energy.

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Several experimental and theoretical results are available for contributed ionatom collisions in the range usually referred to as the low-energy range [2, 3, 4, 5]. For example, for Ne<sup>10+</sup> tons, low projectile energies are typically a tew tens of keV to a few hundred keV. Only little is known about collisions involving multicharged ions at energies lower than 10 keV [1]. In many applied fields such as astrophysics, there is a considerable need for data in this very low energy range, however. Moreover, for basic research, the investigation of charge transfer mechanisms for double electron capture at very few energies is essential.

In the present work [12], the method of high-resolution Auger spectroscopy was used to study mechanisms for double electron capture producing the projectile contigurations  $3\ln^n$  and  $4\ln^l$  ( $n \ge 4$ ) in Ne<sup>(0+)</sup> + He collisions. Emphasis was given to slow collisions with projectile categies as low as 1 keV. At low collision cutrgits the production of the configurations  $3\ln^l$  ( $n \ge 4$ ) of non-equivalent electrons is found to become dominant. It has been shown that dielectronic processes in dared by the electron electron interaction play a major role in the creation of the Ne<sup>3+</sup>  $3\ln^l$  states. The measurements were performed in Hahn-Meitner Institute Berlin, Germany and in the Laboratoire de Spectroscopic Atomique, Caen, France.

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T. Watanabe et al., in Molecular Processes in Space (Plenum, New York, 1990)
 M. Boudjema et al., J. Phys. B 24, 1713 (1991);

## Enhancement of dielectronic processes in $Ne^{10+}$ + He collisions at energies as low as 1 keV

J.-Y. Chesnel,<sup>1</sup> B. Sulik, H. Merabet,<sup>2</sup> M. Grether,<sup>1</sup> A. Spieler,<sup>1</sup> C. Bedouet,<sup>2</sup> F. Frémont,<sup>2</sup> X. Husson<sup>2</sup> and N. Stolterfoht<sup>1</sup>

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The study of mechanisms responsible for charge exchange in ion-atom collisions is essential for understanding various phenomena such as synthesis of molecules in interstellar space, astrophysical plasmas and laboratory plasmas for fusion research [1]. Much attention has been devoted to double electron capture in slow collisions of multiply-charged ions with neutral atomic targets [2, 3, 4, 5]. Mechanisms for double capture have been extensively studied by means of translational energy gain spectroscopy [6, 7], photon spectroscopy [8, 9, 10], and Auger electron spectroscopy [2, 3, 11].

Several experimental and theoretical results are available for multicharged ionatom collisions in the range usually referred to as the low-energy range [2, 3, 4, 5]. For example, for Ne<sup>10+</sup> ions, low projectile energies are typically a few tens of keV to a few hundred keV. Only little is known about collisions involving multicharged ions at energies lower than 10 keV [1]. In many applied fields such as astrophysics, there is a considerable need for data in this very low energy range, however. Moreover, for basic research, the investigation of charge transfer mechanisms for double electron capture at very low energies is essential.

In the present work [12], the method of high-resolution Auger spectroscopy was used to study mechanisms for double electron capture producing the projectile configurations 3lnl' and 4lnl'  $(n \ge 4)$  in Ne<sup>10+</sup> + He collisions. Emphasis was given to slow collisions with projectile energies as low as 1 keV. At low collision energies the production of the configurations 3lnl'  $(n \ge 4)$  of non-equivalent electrons is found to become dominant. It has been shown that dielectronic processes produced by the electron-electron interaction play a major role in the creation of the Ne<sup>8+</sup> 3lnl' states. The measurements were performed in Hahn-Meitner Institute, Berlin, Germany, and in the Laboratoire de Spectroscopie Atomique, Caen, France.

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frequently cited in the open literature is Ref. [1], which is restricted to electron and photon excitation. In the last quarter of a century many ion induced satellite measurements were performed, pushing further the Z, spectator vacancy number and energy shift limits of [1]. Fig.1 shows the Z and energy shift limits for the KeL satellites graphically. 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  $K_{\alpha}L^{4}M^{m}N^{n}$ ...,  $K_{\beta}L^{4}M^{m}N^{n}$ ... satellites, and  $K_{\alpha}^{2}L^{4}M^{m}N^{n}$ ...



Fig. 1: Past and present atomic number, and energy shift limits of data in Ref. [1] and data from the present work, for KoL satellites. Light columns: status 1978, dark columns: present status.

\* 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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## Multiple ionization $K_{\alpha}L$ satellite energy systematics

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Wide range of information stretching from analytical work to neutrino mass from star evolution to plasma temperature can be derived from x-ray spectra. To achieve the possible accuracy all the details of the x-ray spectra need to be understood. The presence of x-ray satellites are common in x-ray spectra. The last compilation frequently cited in the open literature is Ref. [1], which is restricted to electron and photon excitation. In the last quarter of a century many ion induced satellite measurements were performed, pushing further the Z, spectator vacancy number and energy-shift limits of [1]. Fig.1 shows the Z and energy shift limits for the K $\alpha$ L satellites graphically. 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<sup>i</sup>M<sup>m</sup>N<sup>n</sup>..., K $_{\beta}$ L<sup>i</sup>M<sup>m</sup>N<sup>n</sup>... satellites, and K $_{\alpha}^{2}$ L<sup>i</sup>M<sup>m</sup>N<sup>n</sup>..., K $_{\beta}^{2}$ L<sup>i</sup>M<sup>m</sup>N<sup>n</sup>... hypersatellites.



Fig. 1: Past and present atomic number, and energy shift limits of data in Ref. [1] and data from the present work, for K $\alpha$ L satellites. Light columns: status 1978, dark columns: present status.

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## Multiple ionization $K_{\beta}L$ satellite energy systematics

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Similarly to the  $K_{\alpha}L$  satellites (see the previous report), we made a comparison of atomic number and energy shift regions of data in Ref. [1], and data from our present compilation of KL satellite lines, also for the  $K_{\beta}L$  case, for ion (even heavyion) induced satellite measurements. Fig. 1 shows these limits for the  $K_{\beta}L$  satellites graphically.

In many cases the spectator L shell vacancy number increased from 2–3 up to 7, mainly in the  $K_{\alpha}L$  case.



Fig. 1: Past and present atomic number, and energy shift limits of data from Ref. [1] and data from the present work, for  $K_{\beta}L$  satellites. Lightly columns: status 1978, dark columns: present status.

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# High resolution study of the K x-ray spectra from low-Z elements

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The K x-ray spectra induced by 1.5 MeV proton bombardment of Ca, Ti and Fe solid targets were recorded by means of a flat crystal spectrometer. (An example spectrum is given as Fig. 1.) The  $K_{\beta_5}$  line intensity, the KL satellite intensities and the KMM RAE intensities were extracted from the measured spectra. The self-absorption significantly influences the measured spectra (the K absorption edge is situated just above the  $K_{\beta_{1,3}}$  line). The importance of absorption corrections in accurate intensity determination of the weak lines near to K absorption edge is emphasized. The need of inclusion of the whole K x-ray spectrum region in the fitting process for the determination of weak line intensities is discussed. The work was presented at the IBA-13 conference [1], and a paper is in print in the proceedings of the conference (Nucl. Instr. Meth. B).



Fig. 1: The K x-ray spectrum of titanium

The work was supported by OTKA No. 3011 and T016636, and by the Slovenian-Hungarian Intergovernmental Science and Technology Program No. SLO-9/95.

 M. Kavčič, M. Budnar, I. Török, High resolution study of the K x-ray spectra from low-Z elements. Program and Abstracts IBA-13, Lisbon, July 27 – August 1, 1997. Abstract 15/A/P1/MON.

### Post-collision interaction in electron-argon collision processes

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Following the rearrangement processes in collisions among particles (photons, charged particles) and neutral atoms, the influence of the outgoing charged particles on the Auger-electron emission is known as post-collision interaction (PCI), which changes the line-shape, energy [1], and intensity of the Auger-lines [2]. In the last 20 years, these phenomena have been widely studied by a lot of theoretical and experimental groups. In the last few years, the attention was concentrated on the PCI effect of ionized electrons. The existing theories could describe the coincident measurements with high precision, but in the case of the single measurements the problem is to estimate the double-differential cross-section of ionized electrons.

To check the existing theoretical calculation in the case of electron-atom collisions [1], we measured the Ar  $L_{2,3}-M_{2,3}M_{2,3}$  diagram Auger-lines [3] for seven different projectile-electron energies, from 275 to 3000 eV. For the measured impact energies, fitting the collected spectra, we determined the energy shift of the Auger-lines, as a function of the impact energy. The measured energy shifts were compared with the calculated values using the eikonal-approximation of Kuchiev et al. [1]. Theoretical values underestimate the measured data, therefore, to get better agreement between the theoretical and experimental data, we are going to make new approximation of the process [4]. For this purpose, we use the classical-trajectory Monte-Carlo (CTMC) theory, to calculate the double-differential ionization cross section of the Ar  $L_3$  sub-shell in the 0.3–2.0 keV impact energy region. For the largest impact velocities there is no difference between the cross sections induced with light and heavy projectiles. Recently the continuum-distorted-with-eikonalinitial-state (CDW-EIS) model has successfully been applied to study the ionization of Ar by proton impact [5]. So in the present study we also performed a CDW-EIS calculation for the antiproton-Ar collision, at impact energy having equialent velocity to 2 keV electron, to check the agreement with the CTMC calculation. For smaller energies (500 and 300 eV impact energies), only the CTMC method is used [6], where DDCS was calculated for projectiles, and ejected electrons as well. The calculated DDCS (of the projectile, and the ejected electron) is used to determine the energy-shift of the Ar Auger-lines, due to PCI. The applied technique is already used in the case of ion-atom collision [7].

We expect that the agreement between the new calculated PCI energy-shift, using a numerical method, and the measured data will be better because in the former approximation [1] the DDCS was described by an isotropic Rudd-distribution, while measuring the energy shift due to PCI, we found a slight anisotropy. <sup>†</sup> On leave from the Institute of Physics, P.O. Box 57, 11001 Belgrade, Yugoslavia

electron-argon collision processes

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The sheets the exaction theorem calculation in the case of electron-atom collisions [4], we measured the At  $1_{-3}$  M<sub>1,3</sub>AL<sub>23</sub> diagram Angue-films [3] for seven deferrent protecties beet on enciptes. From 275 to 3000 eV, i on the measured unpart mergies, fitting the collected spectra, we determined the energy shifts were compared with the calculated values using the elematrapproximation of Kuchiev *et al.* [1]. Theoretical values and reactions using the elemand data, therefore, to get better agreement between the theoretical and experimental data, therefore, to get better agreement between the theoretical and experimental data, we are going to make new agreement between the theoretical and experimental data, we are going to make new house Carlo (C1MC) theory, to calculate the double-differential ionization cross action of the Ar L, sub-shell in the 0.3-2.0 keV impact energy region. For the initial-state (CDMC) theory projectiles. Recently the continuum-distorted-with-eikonalwith light and heavy projectiles. Recently the continuum-distorted-with-eikonalinitial-state (CDM-EIS) model has successfully been applied to study the ionization of Ar by proton impact [5]. So in the present study we also performed a CDW-EIS house the DDCS was calculated for projectiles, and ejected electrons are well. The calculation for the antipoten-Ar collision, at impact energy having equalent vecalculated DDCS (of the projectiles, and ejected electrons as well. The smaller energies (500 and 300 eV impact energies) only the CTMC calculation. For the congry-shift of the Ar Arger-lines, due to PCI. The applied technique is already calculated DDCS (of the projectile, and the ejected electrons as well. The section the case of ion-atom collision [7].

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# Examination of initial and final state interactions in p + H collisions by CTMC

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It is a continuously interesting field of physics to study the angular and/or energy distributions of the ejected electrons in ion-atom collisions. These kinds of experimental and theoretical studies give basic information about the collision dynamics. It is especially interesting and difficult to define the doubly differential cross-sections (DDCS; differential in energy and the angle of ejected electrons) for low-energy electrons. From the experimental point of view, it is difficult to measure yields of slow energy electrons in a few eV range. From the theoretical point of view, the main difficulty arises when taking into account the many-body interactions during the collision. The contribution of the low-energy electrons to the total ionization cross sections are dominant, therefore the precise knowledge of the behavior of the low-energy electrons is very important. Another interesting question is which region of the projectile's path contributes to the ionization during the incoming projectile's path, and how does it relates to the 'ionization fraction' due to the outgoing half-path of the projectile ('ejectile')?

The classical trajectory Monte Carlo (CTMC) method is quite successful in treating the ionization processes in ion-atom collisions. The CTMC calculation is a nonperturbative method as in it all the interactions between the colliding partners can be taken into account exactly during the collision. This makes the CTMC method a useful tool for studying the two- or more center effects. In the classical picture, it is straightforward to switch on and off the interaction potentials between the individual particles. In this way the effects of the Coulomb interaction between the projectile and the target system can easily be studied along its path segments.

For the first investigation, the simplest collision system was chosen, namely the collision between the proton and the hydrogen atom. The projectile energy was 95 keV, where experimental data for DDCS of low-energy electrons are available [1]. In the present work, based on CTMC, the double differential cross sections of the ionization as a function of the electron emission angle, when the ejected electron energy is 1.5 eV were calculated. In forward observation angles using the full 3-body approximation, the agreement between CTMC calculations and the experimental results seems to be closer than for the quantum mechanical predictions [2]. Moreover, it is seen that the interaction during the incoming half-path gives a significant contribution to the ionization cross-section ('projectile ionization'). At backward observation angles the incoming projectiles ionizations are dominant, while at forward angles that of the outgoing ones ('ejectile ionization').

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- [2] K. Tőkési and G. Hock, Nucl. Instrum. Methods B124 (1997) 398

### The structure of exotic atoms containing positrons and positronium

G.G. Ryzhikh,<sup>†</sup> J. Mitroy<sup>†</sup> and K. Varga

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is a continuously interesting field of physics to study the angular and/or o

The structure of exotic atoms with an attached positron or positronium atom is studied with the use of a large-scale variational expansion in terms of explicitly correlated Gaussian basis functions. The six exotic atoms studied, Lie<sup>+</sup>, LiPs, Bee<sup>+</sup>, Nae<sup>+</sup>, NaPs and Mge<sup>+</sup> were found to have ground states that are stable againt electron emission (see the Table). Two of them, Lie<sup>+</sup> and Nae<sup>+</sup>, are seen to be best described as slightly stretched positronium atoms orbiting around the positively charged cores Li<sup>+</sup> or Na<sup>+</sup>, respectively.

While the stochastic variational method (SVM) can be used and has been used to compute the binding energies of systems with 4 or 5 active particles, the computational effort rapidly increases with the number of particles. Given the existence of a stable positronic lithium, it would be interesting to examine whether heavier alkali atoms, such as sodium and potassium, can bind a positron. However, a full N-body calculation on a system containing 11 electrons and 1 positron (for sodium) is out of the question. The way forward is, obviously, to introduce a fixed-core approximation. In the Fixed-Core Stochastic Variational Method (FCSVM) only a few valence electrons are active around a fixed inert core ion. The Table shows that this is a rather good approximation.

Table 1: Binding energies (in Hartree) of atoms with an attached positron or positronium atom.

	Atom	SVM	FCSVM	For the first investigat
le projectile energy	HPs	0.039190	on and the	Illision between the prot
electrons are availa	Lie <sup>+</sup>	0.002351	0.002177	V, where experimental
ential cross sections	LiPs	0.01105	0.011011	
nara nara electre en tra	Bee <sup>+</sup>	0.002	0.002011	
	Nae <sup>+</sup>		0.000178	
	NaPs		0.007777	
all shou and the star	$Mge^+$	nuanaup 90	0.011111	sults seems to be closer
	a Sumoo	m am Zum	D HORDERAG	er, if is seen that the in

contribution to the ionization cross-section ('projectile ionization'). At backward observation angles the incoming projectiles ionizations are dominant, while at forward angles that of the outgoing ones ('ejectile ionization').

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[2] K. Tökési and G. Hock, Nucl. Instrum. Methods B124 (1997) 398

# The excited state of the Positronium molecule

#### K. Varga

While in the case of the  $H_2$  molecule many bound excited states have been observed and later theoretically studied, in the case of the  $Ps_2$  molecule only the ground state has been predicted. Unlike the  $H_2$  molecule,  $Ps_2$  is a complicated nonadiabatic four-body system, for which the L = 0 state has only been investigated until now. The aim of this work is to explore the spectra of  $Ps_2$  and the biexciton molecules and to look for their possible bound excited states. The four-body problem is solved by the stochastic variational method [1].

We have applied our method to all possible combinations of states with orbital angular momenta L = 0, 1, 2, 3 and spins S = 0, 1, 2. No bound excited states have been found except for one case. In the case of L = 1 (with negative parity) and S = 0 our calculation predicts the existence of a second bound state of the Ps<sub>2</sub> molecule. In that system, the spins of the positronium atoms are coupled to zero. In this spin state, the Ps<sub>2</sub> molecule can dissociate into two Ps atoms (bosons) only if the relative orbital angular momentum is even. Consequently, the Ps<sub>2</sub> molecule with L = 1 and negative parity cannot decay into the ground states of two Ps atoms (Ps(L = 0)+Ps(L = 0)). The energy of this Ps<sub>2</sub>(L = 1) state (E = -0.334408 a.u.) is lower than the energy of the relevant threshold (-0.3125 a.u.), and this state is therefore stable against autodissociation into Ps(L = 0)+Ps(L = 1). The binding energy of this state is 0.5961 eV, which is about 40% stronger than that of the ground state of Ps<sub>2</sub> (0.4355 eV).

This system can annihilate by emitting photons. The lifetime against the annihilation is estimated to be 1.8 ns for the excited state and 0.9 ns for the ground state. Some other properties are listed in the Table.

Expectation value	$\mathrm{Ps}_2(L=0)$	$\operatorname{Ps}_2(L=1)$
$\langle r_{e^-e^-}^2 \rangle$	46.371	96.047
$\langle r_{e^+e^-}^2 \rangle$	29.111	80.152
$\langle r_{e^-e^-} \rangle$	6.033	8.856
$\langle r_{e^+e^-} \rangle$	4.487	7.568
$\langle \delta(r_{e^-e^-}) \rangle$	0.00063	0.00015
$\langle \delta(r_{e^+e^-}) \rangle$	0.022	0.011
$-\langle V \rangle / (2 \langle T \rangle)$	0.999999970	0.9999984

Table 1: Ground-state and excited state expectation values of observables (in a.u.)

[1] K. Varga, J. Usukura and Y. Suzuki, Phys. Rev. Lett., in press.

### Few-electron quantum dots

K. Varga

With the rapid advances in semiconductor technology it has recently become possible to manufacture "zero-dimensional" structures called quantum dots. They are essentially little two-dimensional islands populated by a few electrons, which are laterally confined by an artificial potential. Alternatively, they can be regarded (and called) as artificial atoms, where the confining potential replaces the Coulomb potential of the nucleus.

The example considered here is a two-dimensional (2D) problem of electrons in a quadratic confining potential  $(\sum_{i=1}^{N} \frac{1}{2}m\omega^2 \mathbf{r}_i^2)$ . The novelty of our approach with respect to conventional treatments of quantum dots is that we use explicitly correlated basis functions [1]. The correlation between the electrons is expected to be important and it cannot be appropriately described by mean field approaches. We present calculations for N = 2, 3, 4 electrons as an example (see the Table). Not surprisingly, if the harmonic confinement is strong, the spectrum is close to that of a harmonic oscillator. In the practical cases ( $\omega = 1$  in atomic units), however, the eigenvalues strongly differ from those of the harmonic oscillator. This example illustrates an important feature of these systems: by changing the confinement, one can "tune" the properties of the artificial atoms.

As a second example, we consider a system of N = 3 polarized (S = 3/2) electrons with harmonic confinement  $(\omega = 1)$ , subjected to a magnetic field perpendicular to the 2D plane. The strength of the magnetic field is characterized by the cyclotron frequency  $\omega_c$ . The calculations show that the orbital angular momentum of the ground state of this system is L = 3 for  $\omega_c/\omega \leq 5$ , it is L = 6 for  $5 < \omega_c/\omega \leq 9$ , and then it jumps to L = 9, reproducing the experimentally observed "magic number" sequence (L = 3, 6, 9, ...).

Table 1: Energies of electrons in 2D (in atomic units). The orbital momentum and the spin are zero. The numbers in parentheses are the eigenvalues without the Coulomb interaction.

N	$\omega = 1$	$\omega = 100$
2	3.000(2.000)	212.198 (200.00)
3	7.220(5.000)	525.12 (500.00)
4	10.600(6.000)	654.45 (600.00)

[1] K. Varga and Y. Suzuki, Phys. Rev. A53 (1996) 1907.

### Two-dimensional excitonic complexes

K. Varga

The excitonic complexes have considerable importance in the development of semiconductor physics and spectroscopy. The bound state of a positively charged hole with an effective mass  $m_h$  and an electron with an effective mass  $m_e$  is called an exciton. The mass ratio  $\sigma = m_e/m_h$  can change between  $\sigma = 0$  (hydrogenic limit) and  $\sigma = 1$  (positronic limit) depending on the material and on other factors. Much the same as electron-proton systems, where not only the atom H but the molecule H<sub>2</sub> and the ions H<sub>2</sub><sup>+</sup>, H<sup>-</sup>, H<sub>3</sub><sup>+</sup> are bound, a system of  $n_e$  electrons and  $n_h$  holes can also be bound. Such systems are called excitonic complexes.

Modern semiconductor technology is able to produce artificial nanostructures with diameters comparable to atomic distances. In a particular direction, the system may be frozen to its ground state, to move in a restricted geometry as it were. Such systems are the two-dimensional (2D) excitonic complexes.

The binding energies of these 2D systems have been calculated (see the Table). The most intriguing property of the results is a large (about a factor of 8) increase in the binding energy with respect to the three-dimensional cases. This binding-energy enhancement has been observed experimentally as well.

Table 1: Energies  $(E(\sigma))$  and binding energies with respect to the relevant thresholds  $(B(\sigma))$  of the 2D exciton complexes (in a.u.). The asterisk expresses that the state is found to be unbound.

System	$E(\sigma = 0)$	$E(\sigma = 1)$	$B(\sigma = 0)$	$B(\sigma = 1)$
eh	-2.00	-1.00	-2.00	-1.00
eeh	-2.24	-1.12	-0.24	-0.12
ehh	-2.82	-1.12	-0.82	-0.12
eehh	-5.33	-2.19	-1.33	-0.19
eeehh	*	*	0.00	0.00
eehhh	-6.41	*	-1.08	0.00

#### [1] K. Varga, Phys. Rev. B, accepted for publication.

### Two-dimensional excitonic complexes

K. Varga

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1] K. Varga, Phys. Rev. B. accepted for publication.

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## Energy loss functions for electron energy loss spectroscopy

T. Nagatomi,<sup>a</sup> K. Tőkési, R. Shimizu<sup>a</sup> and R.H. Ritchie<sup>b</sup>

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Detailed interpretation of surface electron spectroscopy data must rely on models for relevant scattering processes. X-ray photoelectron spectroscopy, Auger-electron spectroscopy, and reflection-electron-energy-loss spectroscopy, for example, involve low-energy electrons (<10 keV) for which basic experimental cross section data may be scarce. Because such spectra may contain valuable information about the composition and chemical bonding of atoms in the surface vicinity, it is important to evaluate these models.

The question of how losses to surface plasmons affect experiments has been considered in detail by several different groups of workers [1, 2, 3, 4]. Various models have been invoked in order to study surface effects in the different spectroscopies.

We consider simple electron source configurations and material compositions in order to emphasize aspects that may have received little attention before. We have studied the effect of choosing different response functions in the context of the hydrodynamic model and the specular reflection model [5]. The latter has been invoked recently by several groups in various contexts.

Of some conceptual interest is the spatial dependence of the local energy loss by a charge approaching the boundary of a solid. We will compare commonly assumed response functions with the most reliable that are presently available. The begrenzung (boundary) effect [6], originating in the orthogonality of the surface and bulk eigenmodes, manifests itself in interesting interference effects in the loss functions. We make a simple generalization of the Landau multiple loss function that is much-used in interpretation of experimental data. Errors associated with various commonly assumed response functions will be estimated.

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### Energy losses of swift protons to hydrogen atoms

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Collisions among energetic particles are fundamental process of nature having importance in many different areas of research and technology. Since the beginning of this century one of the exciting problems in collision theory is the calculation of the energy loss of ions passing through different stopping media. In recent decades both classical and quantum-mechanical theories have been used to calculate the stopping power of atoms, molecules and solids for various projectiles [1, 2, 3, 4, ]5, 6]. The main difficulties of calculations for atomic and molecular media is that many-body interactions have to be taken into account. Therefore, the success of various approaches to the calculation of stopping power depends in part on how far a given theory is capable of describing the many-body character of the collision. The theory of the electronic stopping power for charged particles developed by Bethe [7] is frequently used in the interpretation. This theory is based on the first Born approximation, when the stopping is caused by Coulomb excitation and ionization and the result is proportional to the square of the projectile charge. This fact is in contradiction with the observation of Barkas [8] in which higher order terms of the projectile charge have to be taken into account for accurate description of electronic stopping power at lower energies.

The classical trajectory Monte Carlo (CTMC) method has been proved to be successful to deal with the projectile energy loss processes in ion-atom collisions. The CTMC simulation is a non-perturbative method and one of its main advantages is that many-body interactions are exactly taken into account during the collision. Moreover in the classical picture it is straightforward to switch on and off the interaction potentials between the individual particles using switching parameters and thereby to study the effects of single particle interactions. This makes the CTMC method useful for the study of two- or more center effects.

In the present work a classical trajectory Monte Carlo method is used to determine the stopping power for protons inpinging on hydrogen atoms. The energies of the projectiles investigated were in the range of 10 and 1000 keV/amu. The impact parameter dependence of the energy loss is determined along with partial contributions among excitation, ionization and capture. Contributions of the projectile-electron and projectile-target nucleus interactions to projectile loss are also investigated. Our results are compared with experimental data and other theo-
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Elastic peak electron spectroscopy (EPES) is increasingly used [2] for dotermining the inelastic mean free path (IMFP) of electrons in solids. EPES spectra however, especially in the lower (<2 keV) energy region of the primary electron beam, can be influenced considerably by scattering in the surface layers. Therefore, comparing elastic peak intensities obtained from experiments, to those derived from model calculations, more realistic models (accounting for surface effects) are needed. The magnitude of the surface effects on the IMFP  $\lambda$  of the electrons in the energy region 20 eV = 5 keV for ÅI, Co, Ni, Si and Au te demonstrated calculating the surface and bulk  $\lambda$  values from experimental optical data using the defective function model. The depth dependence of  $\lambda$  has been approximated by an exponential function:

 $1/\lambda = 1/\lambda_{\rm B} + (1/\lambda_{\rm S} - 1/\lambda_{\rm B}) \exp(-z\lambda_{\rm B})$  if  $\lambda_{\rm S} > \lambda_{\rm B}$  and

AS = 1/AS + (1/AB + 1/AS)(1 - 3AB) = 1/AS < AB

where  $\lambda_{3}$  and  $\lambda_{41}$  are the surface and bulk (MPP-s, respectively, and z is the depth measured perpendicularly from the surface. A Monee Carlo simulation was applied to calculate the intensity of elastically backscattered electrons from solid using 1) the surface, 2) bulk IMFP only and 3) the new combination of the surface and bulk IMFP, as a function of the primary energy and compared to the available experimental data [3]. The partial expansion method [4] was applied for describing the elastic scattering cross sections in the 200 eV  $\pm$  5 keV energy region.

As a consequence of the surface effects on  $\lambda_i$  considerable changes in the intensit of the elastically scattered electrons (up to 30%) are obtained, as a function of electron energy.

This work was supported by the research projects OTKA F022059, OTKA T007274 and COST/D5/0001/94.

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### Effects of surface excitations on the yield of elastically backscattered electrons

K. Tőkési, L. Kövér and D. Varga

Quantitative applications of electron spectroscopy rely on the knowledge of the elastic and inelastic electron scattering processes in the surface and near surface region of the analysed samples. It has been recognized earlier [1] that for a quantitative description of the energy loss processes the surface excitations should be included and due to the non negligible electron momentum transfer and to the interference between the field created by the incoming and outgoing electron, the simple separation of the cross section into pure surface and bulk contributions is not possible.

Elastic peak electron spectroscopy (EPES) is increasingly used [2] for determining the inelastic mean free path (IMFP) of electrons in solids. EPES spectra, however, especially in the lower (<2 keV) energy region of the primary electron beam, can be influenced considerably by scattering in the surface layers. Therefore, comparing elastic peak intensities obtained from experiments, to those derived from model calculations, more realistic models (accounting for surface effects) are needed. The magnitude of the surface effects on the IMFP  $\lambda$  of the electrons in the energy region 20 eV – 5 keV for Al, Cu, Ni, Si and Au is demonstrated calculating the surface and bulk  $\lambda$  values from experimental optical data using the dielectric function model. The depth dependence of  $\lambda$  has been approximated by an exponential function:

 $1/\lambda = 1/\lambda_B + (1/\lambda_S - 1/\lambda_B) \exp(-z\lambda_B)$  if  $\lambda_S > \lambda_B$  and

 $1/\lambda = 1/\lambda_S + (1/\lambda_B - 1/\lambda_S) (1 - \exp(-z\lambda_B))$  if  $\lambda_S < \lambda_B$ ,

where  $\lambda_S$  and  $\lambda_B$  are the surface and bulk IMFP-s, respectively, and z is the depth measured perpendicularly from the surface. A Monte Carlo simulation was applied to calculate the intensity of elastically backscattered electrons from solid using 1) the surface, 2) bulk IMFP only and 3) the new combination of the surface and bulk IMFP, as a function of the primary energy and compared to the available experimental data [3]. The partial expansion method [4] was applied for describing the elastic scattering cross sections in the 200 eV – 5 keV energy region.

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#### Production of Fe and Cu nanocrystalline particles by thermal decomposition of ferro- and copper-cyanides

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Nanocrystalline Fe and Cu particles have been produced at 653 K in an open vertical tube (filled with pure argon) by thermal decomposition of  $Fe_2Fe(CN)_6$  and  $Cu_2Fe(CN)_6$ , prepared from the precipitates of the solution  $K_4Fe(CN)$  +  $(NH_4)_2Fe(SO_4)_2$  as well as  $K_4Fe(CN)_6+CuSO_4$ , respectively. Depending on the time of the thermal processing (between 0 and 96 hours) small metallic particles with diameter of d = 2 - 100 nm were observed by Transmission Electron Microscopy (TEM). The X-ray diffraction patterns also showed that in the case of  $Cu_2Fe(CN)_6$ the initial grain size was about 20-25 nm and after the heat treatment this decomposed to a mixture of microcrystalline  $Cu_2Fe(CN)_6$ ,  $K_2CuFe(CN)_6$ , pure copper (up to about d = 100 nm) and iron (with d < 20 nm) particles. In the case of the pure ferrocyanide the X-ray diffraction indicated a probable mixture of  $Fe_4(Fe(CN)_6)_3$ and  $K_4 Fe(CN)_6$  in the initial state with grain size of about 16 nm and 40 nm, respectively. This can be due to the fact that the  $Fe_2Fe(CN)_6$  is amorphous and there are no X-ray data for the  $(CN)_n$ , as well, which are expected to be the initial and the final matrix before and after the thermal processing. After the heat treatments there is still a mixture of the above two cyanides with slightly larger grain sizes and with a relatively less amount of  $Fe_4(Fe(CN)_6)_3$  and there are also small iron particles (d < 50 nm) present. The above conclusions were also confirmed by energy dispersive X-ray analysis in TEM and by the magnetic hysteresis behaviour. It was shown that the initial normal paramagnetic behaviour changed to be a mixture of superparamagnetic (due to the small iron particles) and normal paramagnetic components (due to the remaining matrix) at room temperature.

the low angle X-ray diffraction spectrum but they play important role in the strong porosity formation in the silicon side. Furthermore it was illustrated that the initial curvature on the  $\ln(I/I_{c})$  vs. t curve is due to the strong concentration dependence of the intrinsic diffusion coefficients. In epitaxial Mo-V multilayers the thermal behaviour is different. There is a fast transformation of the layered structure into a heterogeneous, partly reacted structure which consists of regions of a reacted Mo(V) alloy and the termaining Mo-V layers. This can not be interpreted by bulk diffusion but rather by the formation of grain-boundaries inside the Mo layer and probably

#### Thermal stability of amorphous and crystalline multilayers produced by magnetron sputtering

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The huge activity in production and investigation of structures with controlled modulation of chemical composition is initiated by many promising areas of application and basic research. For both the applications and basic studies it is necessary to have a very sophisticated technique for the production of good multilayers as well as their structural characterisation and thermal stability is also very important. To demonstrate the high diversity of different applications of magnetron sputtering technique in the fabrication of multilayers the production, characterisation and studies on thermal stability of epitaxial metallic Mo-V and amorphous Si-Ge semiconductor multilayers have been performed. The investigation of the stability of different bi- or multilayers is essential because of the understanding of their behaviour under thermal cycling and/or stress conditions.

The multilayers were prepared using dc planar magnetron sputtering in a highvacuum system. The Si-Ge films were deposited onto Si wafer substrates. Epitaxial (001) oriented Mo-V superlattices were grown on MgO (001) substrates kept at 973 K. Combination of low- and high angle X-ray diffraction as well as transmission electron microscopy were used for characterisation of the modulated structures and to study their behaviour upon thermal treatment. These properties are basically determined by the interdiffusion coefficient in the given system, but because of the complexity of the solid state intermixing the details can be very different for different layers.

The experimental results were compared with numerical calculations made by finite element method for the diffusional homogenisation controlled by bulk diffusion. It was shown that in the case of the amorphous Si-Ge system the intermixing is possible in amorphous state and diffusional stresses do not cause a significant curvature on the thermal decay curves  $(\ln(I/I_o) \text{ vs. } t)$  of the first order Bragg-peak in the low angle X-ray diffraction spectrum but they play important role in the strong porosity formation in the silicon side. Furthermore it was illustrated that the initial curvature on the  $\ln(I/I_o)$  vs. t curve is due to the strong concentration dependence of the intrinsic diffusion coefficients. In epitaxial Mo-V multilayers the thermal behaviour is different. There is a fast transformation of the layered structure into a heterogeneous, partly reacted structure which consists of regions of a reacted Mo(V) alloy and the remaining Mo-V layers. This can not be interpreted by bulk diffusion, but rather by the formation of grain-boundaries inside the Mo layer and probably by a grain-boundary motion induced alloying.

### Intrinsic and domain magnetism of nanocrystalline Fe, Fe(Si) and Ni(Fe) alloys produced by ball-milling

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Experimental results in ball-milled nanocrystalline Fe, Fe(Si) and Ni(Fe) alloys, based on our measurements of the temperature dependence of magnetization, Barkhausen-noise, hysteresis loops and Mössbauer-effect are reviewed and compared with results of measurements obtained on nanocrystalline samples produced by other techniques.

It has been shown that the intrinsic magnetic properties (the saturation magnetization  $(M_s)$ , the hyperfine magnetic field in the Mössbauer spectra, and the Curietemperature  $(T_c)$ ) are almost independent of the grain size (d) down to about 6 nm. This is in accordance with the newest experimental results obtained on nanocrystalline Fe produced by inert gas evaporation [1] and on Ni produced by severe plastic deformation consolidation of ball-milled powders [2] and by electrodeposition [3].

The grain size dependence of coercivity  $(H_c)$  has been found to be similar to what can be expected from the classical and random anisotropy model. The differences observed between the  $H_c$  vs grain size curves can be explained by different sample preparation techniques and to different materials.

A definite correlation between the magnetic Barkhausen-noise (MBN) and the grain shape has been found in Fe which can be attributed to the formation of lamellar and textured grain structure. In Ni — where no texture formation was observed — there was a maximum on the MBN curve versus grain size at the same d where the sharp drop in  $H_c$  at small d values was obtained. Furthermore it was also shown — by the separation of the effect of grain size and the residual strain (by relaxing it in an appropriate heat treatment) — that in nanocrystalline Ni the coercivity is practically independent of the residual strain.

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#### Charge transfer in Cu-Pd alloy systems

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The knowledge of charge transfer between constituents and of changes in local electronic configuration is important in understanding the chemistry of alloying and the catalytic processes at alloy surfaces. Determination of charge transfer in Cu-Pd alloys has become a controversial issue recently [1]. Here an estimated value of the transferred charge in  $Cu_{50}Pd_{50}$  (100) single crystal alloy is presented as determined from measurements of deep core alloy-metal Auger parameter shifts using the linear potential model [1].

Core electron spectra were excited from the alloy and pure metal samples. Cu KLL+2p; Pd 2s, 2p,  $L_{23}MM$  spectra induced by Cu X-rays were measured by using a home-built electron spectrometer [2]. High resolution, Al K $\alpha$  excited spectra (Pd 3d; Cu 2p; VB) were obtained using a SCIENTA ESCA-300 spectrometer [3]. Alloymetal Auger parameter shifts were calculated from the measured Auger kinetic and photoelectron binding energies using the definition given by Williams and Lang [4]. For estimating charge transfer from the analysis of the initial and final state Auger parameter shifts the method of Thomas and Weightman [5] was used, with parameters from atomic structure calculations for free atoms [6].

For CuPd the initial state Auger parameter showed more sensitivity to the changes in the valence charge than the final state one. On the basis of our measurements of deep core transitions in Cu and Pd using the linear potential model we can conclude that a very small,  $\sim 0.05e$  overall charge is transferred from the Pd to the Cu site and the results for Cu and Pd are consistent with the charge neutrality in the alloy. The direction and the value of the transferred charge is confirmed by the calculations using the relativistic DV-X $\alpha$  cluster MO theory for estimating ground state charges. In Pd, the increase of the d occupancy is overcompensated by the loss of s charge, however, the population analysis of the charges indicates a larger loss of d electrons than increase of sp electrons, therefore further, spin-selective calculations are needed.

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#### Observation of surface topography by RBS microbeam

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Nuclear microprobes are able to scan  $\sim 1 \text{ mm}^2$  areas with ion beams focused to a few microns, and have been successfully used in measuring 2 dimensional (2D, or lateral) elemental distributions. The combination of a microbeam with traditional Rutherford backscattering (RBS) techniques can provide 3D distributions for the investigated sample, where the depth as the third dimension is characterised by the energy loss during the incoming and outgoing paths of the scattering ions. Conventionally the ions enter the sample perpendicular to the surface and, due to the nearly 180° scattering angle, the incident and backscattered ions follow almost the same trajectory but in opposite directions. The surface of the samples generally are not flat (they have elevations, depressions, craters, etc., i.e. have a surface topography), but conventional RBS is not sensitive for such details.

In 1997 we upgraded the above technique by equipping our sample chamber with two well-collimated silicon surface barrier detectors movable in a wide range of scattering angle at opposite sides of the beam. By moving the detector from the traditional scattering angle of ~ 170° into grazing exit angle position (around 110°), it becomes sensitive also to the surface topography. Using this arrangement and the scanning capability of the microprobe it became possible to observe the topographical details. For example, by mapping with two detectors simultaneously we could distinguish between effects of elemental distribution and that of surface topography [1, 2]. Furthermore a combination of RBS mapping images with tomographic images could provide quantitative three-dimensional information on the sample [3].

The method is capable of handling even micronsized features and the results obtained in some cases can be evaluated quite easily. Further study, including a simulation program, is needed for the full exploitation of the method in more complex cases.

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# Analysis of spherules and inclusions by $\mu$ PIXE method

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In the framework of the international project IGCP 384 [1] the analysis of cosmic and terrestrial spherules has been continued in 1997. Spherules are microscopic  $(1 - 1000 \ \mu\text{m})$  spheroid or deformed spheroid objects, which can be found in many geological formations such as stones, deep-sea sediments, polar ice and even in the stratosphere. The aim of our investigations is to gain more information about the origin, structure and composition of these objects.

This year the authors focused their efforts chiefly on the study of glassy spherules collected from three Mesozoic (Cretaceous, 67-137 Ma BP.; Jurassic, 137 – 195 Ma BP.; Triassic 195 – 225 Ma BP.) stratigraphical layers in Hungary: Bakonyjákó (Cretaceous); Aszófö (Triassic, limestone) and Vérhalom (Triassic, limestone). The  $\mu$ PIXE analyses of 24 glassy spherules revealed surprisingly high Ca and Ba concentrations (CaO ~ 40%, BaO ~ 2.5%) [2] to which similar results have never been reported before. From geological arguments it may be inferred that these spherules are terrestrial impact products. The main problem, which remains to be explained, is the perfectly uniform composition of samples from various stratigraphical layers separated by 80 Ma.

As a continuation of the previous measurements the analysis of metallic spherules has also been started. Metallic spherules were collected partly from roofs of houses in Hungary (~30 samples) and partly from Permian/Triassic stratigraphical layers (5 samples). The  $\mu$ PIXE analyses has clearly shown that samples collected from roofs are of industrial origin while those from P/T layers should be of cosmic origin concluded from their high Ni, Cu and Zn concentrations (~2%, ~0.6% and ~2%).

In the framework of a Japanese Hungarian co-operation (IGCP 384) the elemental composition of inclusions (< 100  $\mu$ m) in impact samples have been investigated. In four different rocks collected in the neighbourhood of impact craters in Japan (Si-rich rocks of granite, sandstone, etc.) 13 inclusions were analysed by  $\mu$ PIXE. The evaluation of measurements is in progress.

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### The inelastic mean free path (IMFP) and the inelastic scattering cross section of electrons in GaAs determined from highly resolved electron energy spectra

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GaAs samples have been studied with hemispherical analyser of high resolution (type ESA-31). The analyser covers the energy range 50 - 5000 eV with controlled energy resolution. Prior to measurements, sample surfaces have been exposed to Ar<sup>+</sup> ions in order to amorphise the surface layers. This procedure resulted in Ga enrichment (70 - 85 at. % Ga as determined by XPS). The elastic peak and EELS spectra were measured in the loss range  $E_p$ - $E_l$  of 50 eV. The elastic peak intensity ratios of GaAs and Ni reference were used to determine the IMFP in GaAs. The relations between these ratios and the IMFP have been determined from Monte Carlo simulation of the elastic scattering effect. The values of the IMFP resulting from this procedure are reasonable agreement with literature data. The inelastic scattering cross sections have been determined using the Tougaard procedure. The energy loss distributions  $\lambda$  K are presented in the 200 - 5000 eV range.

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*Note:* The paper was presented at ECOSS-17 (European Conf. on Surface Science) Sep. 16–19, 1997, Enschede, NL., (Europhysics Conference Abstracts Vol. 21E TuP33 pp. 55; Eds: G.A.M. Kip and H. Wormeester) and accepted for publication in Surface Science.

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# **Resolution correction in EELS**

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Correction for inelastically scattered electrons is important in determining intensities in elastic peak electron spectroscopy (EPES), e.g. for obtaining information on inelastic mean free path in the material, for quantitative applications of electron spectroscopy. Spectral shape of electron energy loss spectra depends more strongly on the spectrometer function in the elastic peak region, than in the continuous energy part. Therefore the resolution correction of the EELS spectra results in a considerable change of the spectral shape. The importance and the role of this correction is demonstrated in the case of EPES of Ni sample comparing the shapes of REELS spectra measured at different energy resolutions using different type spectrometers: 1. home-made HSA (ESA-31), Debrecen; 2. home-made HSA (Clermont-Ferrand); 3. CMA MAC2 by CAMECA-Riber and a HSA (Lyon); 4. CMA OPC103 type by Riber (Budapest). The resolution correction was made by the ratio type deconvolution method.

*Note:* the paper was presented at the int. conf. of JVC-7 (7th Joint Vacuum Conference of Austria, Croatia, Slovenia and Hungary) 26–29 May 1997, Debrecen. The paper accepted for publication in the int. journ. of *Vacuum*.

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### Experimental determination of the inelastic mean free path (IMFP) of electrons in GaAs and InP

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The inelastic mean free path (IMFP),  $\lambda_i$ , is an important parameter determining the electron emission from solids excited by electrons or photons. The knowledge on the IMFP is indispensable for quantitative surface analysis using AES, XPS, UPS or EELS. Recently the IMFP values for elements and compounds have been published in the energy range 0.1 - 2.0 keV. In the case of semiconducting III-V compounds Kwei and Chen, and Gries have reported theoretical calculations of  $\lambda_i$ . Presently EPES (elastic peak electron spectroscopy) proved to be an efficient experimental method using clean solid surfaces. The method is based on the ratio of Auger elastic peak intensities arising from a sample and from a reference sample,  $I_E/I_E^s$ . The Ni sample proved to be a good standard. Systematic research resulted in experimental IMFP data for a great number of elements. The retarding field analyser (RFA) was also used for determining the absolute values of elastic reflection coefficient  $r_c$ (%) of the surface. Experiments have been evaluated by Monte Carlo calculations. Very few experimental  $\lambda_i$  data have been published on compounds. Apart from our early paper, authors are not aware of  $\lambda_i$  of III-V compounds. The paper is reporting on round robin measurements of GaAs and InP performed in four laboratories using the Ni reference and different electron spectrometers, covering various energy ranges.

Note: The paper was presented at ECASIA-97 in Göteborg (Abstracts: QA-22, pp. 268.) and published in the peer reviewed Conf. Proc. of ECASIA-97 pp. 836–839. Eds: I. Olefjord, L. Nyborg, D. Briggs; John Wiley & Sons, Chichester, 1997. ISBN 0 471 97827 2

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## Low frequency inverse AC Josephson effect in granular high- $T_c$ Bi(Pb)SrCaCuO films

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Some special features of current-voltage characteristics of Josephson junction arrays which are formed by grain boundary junctions in granular high- $T_c$  Bi(Pb)SrCa-CuO superconducting films were investigated. We observed an AC to DC conversion effect in a relatively low frequency (10 - 500 MHz) AC field in these films without DC bias current. The dependence of DC voltage on temperature, magnetic field, AC field amplitude and film width were investigated. We gave an interpretation to this non-zero voltage and through a simulation study showed that there exists a new type of inverse AC Josephson effect in high- $T_c$  screen printed films at frequencies well below the plasma frequency. The plasma oscillation in Josephson junctions causes a special phase lock mechanism of the oscillating Josephson current to the AC field, which results in a DC voltage at zero transport current. We calculated the stability areas of this inverse AC Josephson effect for a single junction as a function of frequency. The results are shown in the Fig. 1 for two different  $\beta_c$  parameters. The stable lock-in ranges where there is a phase lock between the external AC excitation and Josephson oscillation are those frequency ranges where the voltage curves are drawn on the broken lines.



Fig. 1: The stable lock-in ranges as a function of frequency

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#### Seasonal variation of the composition of urban and rural atmospheric aerosol

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Aerosol sampling has been continued in urban and rural sampling sites. The characterization of regional atmospheric aerosol in and around Debrecen was performed through the elemental analysis on total, coarse and fine fraction aerosol samples.

During the evaluation of elemental concentration data together with yearly average values subdivision to seasonal winter and summer averages have been also made according to the winter heating period. Seasonal average concentration of elements of predominantly natural origin (Al, Si, Ca, Ti, Fe, Ba with enrichment factors less than 6) in the coarse mode are obviously lowered in the winter period by frozen soil and snow, while summer increase can be explained by increased erosion of the ground during summer period. Elements of anthropogenic origin (S, Cl, K, V, Mn, Zn, Br, Pb with high enrichment factors) in the fine mode are present more abundantly during the winter heating period.

The time distributions of monthly average concentration values over a year's period show much the same regularity: in total Debrecen values as well as in coarse and fine fractions from both the urban and rural locations Al, Si, Ca, Ti, Fe exhibit well expressed summer maxima, while S, Cl, K, Cu, Zn, Br and Pb show maxima in winter. To some extent P belongs to the first group, too. (The two size fractions for V and Mn behave differently, in the cases of these elements coarse and fine fractions are characterized with summer and winter maxima, respectively).

The summer maxima appearing on the concentrations of soil-derived elements indicate that increased wash-out during the broad summer maxima of monthly amount of precipitation observed over multiannual periods for the location can not fully compensate for the effect of the increased summer erosion. However, the wash-out effect is clearly indicated by a negative correlation between concentrations and precipitation data as well as by the clean up of the atmosphere in the immediate vicinity of rains.

Additional to the determination of elemental concentrations in PIXE analysis the total suspended particular masses (TSP) on single stage samples as well as particle masses of the  $2\mu m < EAD < 10\mu m$  and  $EAD < 2\mu m$  fractions denoted as PM10 and PM2, respectively, on two stage samples have been measured by the use of a Sartorius microbalance. "Black Carbon" component (BC) collected on the fine stage samples has been measured by a Smoke stain reflectometer on an absolute ng/m<sup>3</sup> scale.

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### Dependence of the composition of urban atmospheric aerosol on wind direction

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Data on elemental composition of atmospheric aerosol at a given location are widely used for the qualification of local air conditions. The determination of correlations with local meteorological conditions makes the qualification more complete.

In the present work the Hungarian urban site Debrecen have been used as sampling station for collecting atmospheric aerosol samples. Proton induced X-ray emission analytical method has been applied for deducing multielemental absolute concentration data on elemental constituents of the samples. A PIXE computer package named PIXYKLM has been used for the evaluation of spectra and elemental concentrations.

The actual shape of concentration curves are to a smaller extent influenced by wind distribution data. A correlation analysis performed on concentration and wind speed data indicate negative correlation in all the data sets.

The dependence of the elemental concentrations on local wind direction has been investigated by the help of concentrations averaged for separate sectors of local wind distribution over a period of five years. The results clearly indicate the similarity of the wind rose distributions of concentrations for soil-derived elements. There appears a well expressed asymmetry with respect to an axis of north-south direction. This behaviour closely reflects the geological structure of the region: Debrecen is situated over a line of north-south direction which separetes a western loess covered table from an eastern slightly undulated sandy terrain with spots of loose forest. Elements of anthropogenic origin partly arising from long range transport processes exhibit more uniform wind rose distributions with slight indications of local sources and main traffic roads crossing the town. While the above mentioned regional effects can be detected by the help of local wind distribution data, the influence of long range transport effects is better represented in trajectory sector data analysis based on calculated backward air trajectories. Such type of evaluation can be treated, too.

The results of the work presented above gave new information on the aerosol load of the Hungarian atmosphere over the Debrecen urban site. The nature of the contributing sources has been investigated and some attempts have been made for indicating the role played by neighbouring regions in determining the air quality at the site mentioned.

This work was supported in part by the Hungarian National Foundation for Scientific Research (OTKA No. T17040) and by the International Atomic Energy Agency (IAEA CRP-7525/RB).

### Characterisation of Carpathian obsidians by LA-ICP-MS, $(\mu)$ PIXE and PIGE methods

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The last comprehensive summary on the study of Carpathian obsidians has been published recently by Williams-Thorpe (1995) [1], indicating that all open questions in this field has been answered. However, it seems that there are still problems to be solved. Even in the above mentioned review article the current lack of routine application of the provenancing methods was mentioned and the need for further non-destructive methods of analysis and characterisation was emphasised.

Hypothetical sources in south-west Ukraine still remained unexplored and claims for Romanian sources not adequately tested. Provenance of some artefacts found in Eastern Hungarian museums is still unknown and could not be matched with known Hungarian and Slovakian sources. The characterisation of further sources and artefacts by sensitive analytical methods could reveal further existing connections. The comparative raw material collection of the Hungarian National Museum (LITHOTHECA) contains a good collection of samples which we intend to complete with new samples and analyses.

The aim of our study was to compare three different analytical methods which can be applied for obsidian characterisation, such as LA-ICP-MS (laser ablation – inductively coupled plasma – mass spectrometry technique), non-destructive PIXE (particle-induced x-ray emission) and a complementary method, PIGE (protoninduced gamma-ray emission). The analyses were carried out mainly on Hungarian source materials and artefacts. For comparison, some samples from Armenia, Iceland, Mexico, Slovakia and Turkey were included as well.

Up to now we have analysed more than 25 obsidian samples. The evaluation of measurements will be completed in the near future. Preliminary results indicate that based on trace elemental concentration data the provenance of numerous Carpathian obsidian artefacts can be determined.

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#### Ion beam analysis of the meteorite from Kaba, Hungary

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On the 15th of April 1857 a stony meteorite (mass  $\sim 2940$  g, max. diam.  $\sim 15$  cm) fell into the neighbourhood of the village Kaba, Eastern Hungary. Shortly after it was recovered, the meteorite was deposited in the collection of the College of the Reformed Church at Debrecen where it is still preserved. Since that time several chemical analyses was performed. The last one was completed in 1961 in Hungary [1].

Considering the numerous requests applied to the Board of Directors of the College since the previous sampling they permitted to carry out a new analysis which was inspired by developments in analytical techniques during the past 35 years. This new study has been co-ordinated by I. Kubovics at the Department of Petrology, Eötvös Lóránd University, Budapest. Samples (total mass 14 g) were cut in ATOMKI and were used in different kind of investigations in several laboratories throughout Hungary [2].

At the Department of Electrostatic Accelerators of ATOMKI preliminary ion beam analytical studies of the meteorite Kaba was begun in 1997. The authors used non-destructive PIGE and  $\mu$ PIXE methods which are capable to determine both major and trace elements simultaneously with high sensitivity.

We have analysed an arbitrarily selected  $\sim 5 \text{ mm}^2$  area on the surface of a thin  $(\sim 40 \ \mu m)$  section made from the meteorite using PIGE method at a proton energy of 3.8 MeV. PIGE is sensitive especially for the light elements and in accordance with previous chemical analyses [1] we found O, Na, Mg, Al, Si, and P in the sample. For trace and major elements complementary measurements were carried out at the scanning proton microprobe (SPM) facility of ATOMKI.  $\mu$ PIXE method can provide analytical data for a wide range (Z > 12) of elements with  $\mu m$  spatial resolution (~ 2 × 2  $\mu$ m<sup>2</sup> in our case). As a first step we made elemental maps from a relatively larger part  $(750 \times 750 \ \mu m^2)$  of the sample in order to determine the elemental distributions. The proton energy was 2 MeV. Utilizing the excellent spatial resolution of the SPM we could choose a few homogeneous small areas of the sample and we analysed them separately (see Fig. 1). Rich regions in Si, Ca, Fe and Fe+S were found, which indicates that even a small part of the sample contains many different minerals. Although previous measurements claimed that Zn was not present in the meteorite we could detect it in the Fe+S-rich region. In the near future this work will be extended by joint  $\mu$ PIXE, PIGE and EPMA measurements.

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Fig. 1: Optical image and elemental mapping of a thin ( $\sim 40 \ \mu m$ ) section made from the meteorite Kaba. (These pictures are black and white variants of the original coloured ones.) The elemental maps for Si, S, Ca, Fe and Ni show high inhomogeneity and correlations between elements indicating the presence of many different minerals in very small sizes.

#### Airborne <sup>222</sup>Rn concentrations in karst caves

#### J. Hakl, I. Csige and I. Hunyadi

In the last two decades the development and the widespread application of nuclear techniques have found their place also in radon measurement projects aimed at studying the cave environment. Compiling all the globally available radon concentration data from 220 different caves world-wide we have found that the distribution of <sup>222</sup>Rn concentrations is lognormal (GM = 1130 Bq·m<sup>-3</sup>, GSD = 6.3, see Fig. 1), which is in good accordance with the awaited distribution for a geochemical element. The lower end of the scale is associated either with big cave chamber volumes or high ventilation rates; the upper end is characterised by closed, badly ventilated places and uranium-rich sediments. For the 31 examined Hungarian caves we have determined the cave average annual mean radon activity concentrations, 0.3 - 20 $kBq \cdot m^{-3}$ , the characteristic annual maximum/minimum ratios, 2-50, and the periodicity, which was typically one or half a year. In the majority of cases the marked seasonal variations can be characterised with high radon concentration values in summer and low radon concentration values in winter. In a few cases, reversibly, summer minima and winter maxima were observed. We have observed a long-term variation of the annual mean radon activity concentration in all the studied caves.





This work was supported in part by the National Scientific Research Fund contract Nos. T 016558 and T 017560.

#### Geochronological studies with the K/Ar method in 1997

K. Balogh, E. Árva-Sós and Z. Pécskay

A review paper on the Mesozoic igneous rocks in Hungary appeared in 1997 (Cooperation: Dept. of Petrol. Miner., Eötvös Univ., Budapest) [1]. It has been proved, that in the Nebrodi Mts. (Sicily, Italy) volcanic pebbles in the Reitano-flish, in contrast to the expectations, were not formed during the Tertiary subduction, but they are connected to the rifting when the Tethys has been opened in the Jurassic (C: Univ. of Cagliary) [2]. Age of the Glarus overthrust was successfully dated by measuring  $< \mu m$  clay minerals (C: Lab. Geochem. Res. Hung. Acad. Sci., Budapest; Univ. of Basel) [3]. Correlation in time of the Neogene magmatism in the PANCARDI region has been established (C: Birkbeck College, London) [4]. A set of K/Ar ages on Miocene volcanic rocks has been published from the Apuseni Mts. (Romania) and compared with other volcanic areas of the Carpatho-Pannonian realm (C: Romanian Geol. Survey, Bucharest) [5]. Upper Cretaceous ages have been measured for magmatism and ore mineralization of the Ridanj-Krepojin Belt, E-Serbia (C: Univ. Belgrade) [6]. Dating of bostonite from the Mórágy-Hill (Mecsek Mts., S-Hungary) shows, that consolidation of these rocks happened during the Austrian phase of Alpine orogeny (C: Dept. Miner. Geochem. and Petrol., József Univ., Szeged) [7]. Early Rupelian ages (32.3–30.0 Ma) have been obtained on the magmatic rocks from Ruen magmato-tectonic zone, W-Bulgaria (C: Geol. Inst. Bulg. Acad. Sci., Sofia) [8].

In the Bükk Foreland eruptive sequences of Miocene volcanism have been differentiated in the frame of a complex volcanological, geochemical and paleomagnetic study (C: Lab. Geochem. Res. Hung. Acad. Sci., Budapest; Eötvös Inst. of Geophys., Budapest; Romanian Geol. Survey, Bucharest).

A review paper has been prepared on the basaltic magmatism of the Graz basin (C.: Univ. Florence, Birkbeck College, Univ. Leoben), chronological study of Tertiary basaltic magmatism in the Bohemian Middle Mts. has been started (C.: Geol. Inst. Czech. Acad. Sci.) and high aluminum basalts have been dated from Central Slovakia (C.: Slovakian Geol. Survey, Bratislava).

Upper Cretaceous overthrust ages have been measured with the Ar-Ar method in the Sopron Mts. and in the southern part of the Tisza Unit.

Metamorphic and magmatic rocks were dated from S-Transdanubia for the Hungarian Oil Company and clay minerals for the Mecsek Ore Mining Company.

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#### A realization of Ar-Ar dating in Hungary

K. Balogh and A. Simonits<sup>\*</sup> \* MTA KFKI, Atomic Energy Research Institute . H-1121 Budapest, Konkoly-Thege út 29-33

The  ${}^{40}\text{Ar}/{}^{39}\text{Ar}$  technique for dating geological samples has been introduced in order to study complex thermal history of tectonic units in Hungary. The Budapest Research Reactor (tank type, 10 MW nominal power) of the Atomic Energy Research Institute, was used for irradiation [1], while measurement of Ar isotopes was performed in the geochronological laboratory of the Institute of Nuclear Research, Debrecen [2].

Irradiations were planned so, that without decreasing the accuracy of ages, the radioactivity of the samples be kept at a minimum. This required the application of Cd shield and irradiation of samples with the lowest fluence sufficient for the accurate measurement of the <sup>39</sup>Ar isotope. In our case a <sup>40</sup>Ar(rad)/<sup>39</sup>Ar ratio of 50 was sufficient for the accurate measurement and this was achieved with a fluence of about  $5 \times 10^{17}$  n/cm<sup>2</sup> for Hercynian minerals in 7–8 hours. Channel No. 229/3, positioned within the core, but far from its center, was selected for irradiation. This is a compromise, the fast neutron flux is sufficient here, but the danger of overheating the samples is avoided and the fast/epithermal neutron flux ratio is also favorably high at this position. As a result of a Cd shield, the low fluence and selection of channel 229/3 for irradiation, the activity of the samples remained sufficiently low for further processing while allowing simultaneous neutron activation analysis for elements activating mainly for epithermal neutrons.

Samples and standards were wrapped in 0.1 mm aluminum foil and compressed to cylinders of 6 mm diameter. The cylinders were placed in Al tubes of 50 mm length and 6.5 mm inner diameter. Neutron flux was monitored with Ni and Fe foils along the Al tube. 4 Al tubes were placed in a cylindrical Cd box of 0.5 mm wall thickness, the Cd box was closed hermetically into an Al "bomb" by cold welding. The Al "bomb" was placed in the irradiation can. The monitors indicated a flux variation of about 15% necessitating the use of a lot of monitors for accurate measurement of  $^{40}$ Ar/ $^{39}$ Ar ages. Instead, a simple solution has been found to rotate the Al "bomb" within the irradiation can by fixing the "bomb" in the can by axes equipped with a propeller. This way, the scatter of fluence at a fixed vertical position of the Al tubes decreased to below 0.3%. Typical production ratios are  $2.3 - 3.2 \times 10^{-6}$ cc STP  $^{39}$ Ar/gK×hour and  $1.0 - 1.8 \times 10^{-6}$  cc STP  $^{37}$ Ar/gCa×hour. The total activity of irradiated samples after 70 days cooling was  $< 25 \ \mu$ C. Stepwise degassing was performed in a furnace attached to the Ar extraction line. The furnace was constructed of 2 concentric resistance heated cylinders prepared from molybdenum sheet of 0.07 mm thickness. Samples were degassed in a quartz tube placed within the inner cylinder.

This work was supported by the OTKA Foundation Grant No. 014961.

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### Determination of <sup>36</sup>Cl in nuclear waste

Zs. Szántó, E. Hertelendi, J. Csongor and J. Gulyás

<sup>36</sup>Cl is a soft  $\beta$  emitter ( $E_{\text{max}} = 709 \text{ keV}$ ) with a half-life of  $3.01 \times 10^5$  years. <sup>36</sup>Cl is produced mainly by spallation of <sup>40</sup>Ar by cosmic rays in the atmosphere. In nuclear power plants <sup>36</sup>Cl is formed via neutron activation of <sup>35</sup>Cl in the cooling water system. Due to its long half-life and geochemical behaviour (no retardation or retention due to interaction with the rock) the application of this nuclide has grown extensively and include groundwater dating, use as a geothermal or infiltration tracer or may play a prominent role in the inventory of a radioactive waste repository. The significance of this radionuclide is dependent on its inventory which, currently, is based on rough estimates.

Our aim was to develop a radiochemical separation method of <sup>36</sup>Cl from nuclear waste samples of Paks NPP. The <sup>36</sup>Cl activity was measured with LSC (liquid scintillation counting) method.

l v l	Samples	<sup>36</sup> Cl [Bq/l]	$^{137}\mathrm{Cs}\;[\mathrm{Bq/l}]$	$^{36}{\rm Cl}/^{137}{\rm Cs}$	<sup>60</sup> Co [Bq/l]	$^{36}\mathrm{Cl}/^{60}\mathrm{Co}$
1	01TW30B002	1.5	$2.97 \times 10^4$	$5.0 \times 10^{-5}$	$2.43 \times 10^3$	$6.1 \times 10^{-4}$
2	01TW30B005	3.5	$7.71 \times 10^5$	$4.5  imes 10^{-6}$	$1.03 \times 10^6$	$3.4 \times 10^{-6}$
3	01TW10B001	DL	692	and an and a h	515	antor not
4	01TW30B005	3.4	$4.55 \times 10^5$	$7.4 \times 10^{-6}$	$3.09 \times 10^6$	$1.1 \times 10^{-6}$
5	01TW30B003	4.7	$6.82 \times 10^5$	$6.9 \times 10^{-6}$	$3.18 \times 10^4$	$1.5  imes 10^{-4}$
6	02TW10B002	0.4	$3.26  imes 10^5$	$1.3 \times 10^{-6}$	$1.31 \times 10^4$	$3.0 \times 10^{-5}$
7*	02TW30B004	0.1	$6.85 \times 10^3$	$1.4 \times 10^{-5}$	$2.06 \times 10^4$	$4.8 \times 10^{-6}$
8	02TW30B001	1.3	$1.78 \times 10^{6}$	$7.2 \times 10^{-7}$	$1.40 \times 10^{5}$	$9.2 \times 10^{-?}$
9	02TW30B003	1.8	$1.48 \times 10^{6}$	$1.2 \times 10^{-6}$	$4.21 \times 10^5$	$4.3 \times 10^{-6}$
	Ion exchange resin 10TE01N001	1.0 Bq/g	$2.43  imes 10^4$ Bq/g	$4.0  imes 10^{-5}$	$5.48 \times 10^3$ Bq/g	$1.8  imes 10^{-4}$

Table 1: <sup>36</sup>Cl activity in nuclear waste

#### \* radioactive sludge

In order to separate Cl<sup>-</sup> in the form of HCl a distillation method was developed. The distillation was performed in the following way: 10g of ion exchange resin or 11 of evaporator concentrate were weighed into the reaction vessel. Concentrated  $H_2SO_4$  (250–300 ml) was added slowly to the sample until all the materials were dissolved. The solution was magnetically stirred. NaCl was applied as a carrier and chemical recovery was used as a measure of radiochemical recovery. The distillation temperature was regulated between 200–250 °C. N<sub>2</sub> with an appropriate flow-rate was introduced into the reaction flask to carry the HCl to an absorption trap containing 100 ml of NaOH solution. Prior to the HCl trap, the gas was bubbled through an  $H_2SO_4$  trap to absorb trace amounts of  $SO_2$  and reduce the amount of sulfuric species in the final chloride samples. The HCl collected in the NaOH trap was washed with nitric acid in order to remove carbon from the solution in the form of  $CO_2$ .

Acidified silver nitrate solution ( $\approx 10$  ml) was added to precipitate AgCl. The precipitate was dispergated in diethyleneamine, PICOFLUOR scintillation cocktail was added and <sup>36</sup>Cl activity was determined by LSC.

The developed radiochemical method was used for <sup>36</sup>Cl measurement of samples coming from various waste streams of Paks NPP (used ion exchange resins, evaporator concentrates). The results obtained are presented in Table 1.

Key nuclides <sup>137</sup>Cs and <sup>60</sup>Co were also measured, which allowed us to determine the scaling factors for <sup>36</sup>Cl relative to <sup>137</sup>Cs and <sup>60</sup>Co. The resulting scaling factors for the evaporator concentrates are respectively <sup>36</sup>Cl/<sup>137</sup>Cs:  $5.0 \times 10^{-5}$ – $7.2 \times 10^{-7}$ and <sup>36</sup>Cl/<sup>60</sup>Co:  $1.5 \times 10^{-4}$ – $4.3 \times 10^{-6}$ . For resin sample <sup>36</sup>Cl/<sup>137</sup>Cs:  $4 \times 10^{-5}$  and <sup>36</sup>Cl/<sup>60</sup>Co:  $2.7 \times 10^{-4}$  values were obtained, in reasonable agreement with the values given by the Nagra Technical Report [2].

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the pumped amounts leads to the mining of water resources. The maintenance of the existing water-supply systems are feasible only under a strict control of pumping rates. In that case, the water-supply development of the group of islands considered could be achieved by linking the insular water-supply system with that one from the Kvarnet mainland areas in which the available water resources are already limited.

#### Interpretation of the isotope results

Three profiles of the lake water (1, 2, 3) were sampled at various depths for  $\delta^{13}$ O.  $\delta^{13}$ C. tritium and  ${}^{14}$ C measurements. The measurements were carried out in the Institute of Nuclear Research of the HAS.

From the strong enrichment of <sup>2</sup>H and <sup>18</sup>O (see Table 1) in the lake water (composition close to sea water) one can conclude that there does not exist any considerable outflow from the lake

The inflow of factst water into the lake can be only of minor importance since the amount of precipitation is about the same as that of evaporation from the surface of the lake (about 1000 mm per year) and there is no considerable outflow.

From the existing isotope data there is no evidence of an inflow of karst water in the deepest part of the lake. If there exists an inflow of water which cannot be detected by isotope measurements, this can only be explained by the following mechanism: During periods with high water level in the lake, lake water is flowing into the neighbouring karst water system. If the water level in the lake is below that in the karst aquifer, this water may flow back into the lake again. In this case the isotope ratios of the flowing water do not significantly differ from those of the lake water

#### The origin of the water in Vrana Lake

Ede Hertelendi, Éva Svingor, István Futó and Zsuzsa Szántó

#### The hydrogeological problem

The Lake of Vrana, situated in the central part of Cres Island, has, as a water phenomenon, a dominant position and significance. The size of the lake is fascinating for the islands of so elongated shapes as are the Cres and Losinj. The total area of the lake is  $5 \text{ km}^2$ , maximum depth is 70 m (60 m below sea level) and it accumulates 220 million m<sup>3</sup> of fresh water. (Hertelendi et al., 1995; 1997).

The basic problem of the Lake of Vrana is the origin of its water. The present exploration and plans for further explorations are aimed at solving this problem. Most of this exploration suggests the concept of the water origin from both the local and regional drainage areas. However, for the time being we still have to take into account the earlier concept of the lake water origin from only the local drainage area, i.e. from Cres Island only.

If only the local drainage areas is concerned, substantial hydrological changes should not be expected at the lake but, anyway, the rate of total lake water pumping should be studied carefully. The existing water budget analyses show that the rate of pumping already approaches the upper admissible limit and any increase in the pumped amounts leads to the mining of water resources. The maintenance of the existing water-supply systems are feasible only under a strict control of pumping rates. In that case, the water-supply development of the group of islands considered could be achieved by linking the insular water-supply system with that one from the Kvarner mainland areas in which the available water resources are already limited.

#### Interpretation of the isotope results

Three profiles of the lake water (1, 2, 3) were sampled at various depths for  $\delta^{18}$ O,  $\delta$ D,  $\delta^{13}$ C, tritium and <sup>14</sup>C measurements. The measurements were carried out in the Institute of Nuclear Research of the HAS.

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Profile/depth	$\delta^{13}C$ [% <sub>0</sub> ]	$\delta^{18}O$ [% <sub>0</sub> ]	δD [% <sub>0</sub> ]	pMC	<sup>3</sup> H	Temp
r romoj dopom	(PDB)	(SMOW)	(SMOW)	[%]	(TU)	°C
1/0	-1.29	-0.41	-8.55	109.5	20.8	19.5
1/10	-1.09	-0.48	-10.25	110.1	20.3	16.5
1/20	-1.42	-0.56	-9.12	109.0	21.4	13.0
1/30	-1.28	-0.46	-9.30	110.8	19.7	12.5
1/40	-1.71	-0.49	-9.09	110.2	18.0	10.5
1/50	-2.19	-0.56	-8.70	109.8	20.6	11.0
1/60	-1.69	-0.40	-9.09	107.7	20.4	10.0
2/0	-1.27	-0.33	-10.76	110.0	21.2	22.0
2/10	-1.28	-0.23	-9.41	109.4	20.6	16.6
2/20	-1.25	-0.22	-9.58	109.1	20.9	13.0
2/30	-1.06	-0.27	-9.78	110.6	23.3	11.5
2/40	-1.54	-0.39	-10.97	110.4	21.9	10.5
2/50	-2.02	-0.52	-10.03	109.4	22.0	9.5
3/0	-1.58	-0.70	-10.44	110.3	20.6	1.1.1
3/10	-1.19	-0.45	-8.41	110.1	20.0	
3/20	-1.32	-0.35	-10.14	110.0	20.1	
3/30	-1.38	-0.45	-10.93	109.7	23.1	

Table 1: Radiocarbon and tritium concentrations and isotope hydrological data as a function of depth in the Vrana Lake

From the strong enrichment of <sup>2</sup>H and <sup>18</sup>O in the lake water it can be concluded that also <sup>3</sup>H becomes considerably enriched in the lake water (about 10%). If the enrichment of tritium is taken into account a mean residence time (MRT) of 30–40 years was obtained from the measured tritium values. More precise value of MRT can not be given because of the lack of systematic tritium measurements of the local precipitation. In this calculation we used the <sup>3</sup>H data of precipitation collected in Genoa IAEA Network Station.

The MRT can be given for this lake as the ratio of annual inflow divided by the total volume of the lake. The average depth of the lake is approximately 40 m, the precipitation rate is 1 m/year. The obtained 40 year MRT is in good agreement with that of obtained from the isotope data. This suggests that the lake water originates from the local drainage area i.e. Cres Island only. Therefore the existing water supply system are feasible only under a strict control of pumping rates because water budget analyses show that the rate of pumping already approaches the upper admissible limit.

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### Concentration of <sup>85</sup>Kr in primary water and stack air of Paks Nuclear Power Plant

#### E. Hertelendi, Zs. Szántó, É. Svingor and M. Molnár

Radionuclide  ${}^{85}$ Kr is a fission product, with half life of 10.78 years, and fuel reprocessing plants are the dominant ones among its sources. (Recently more than 97% of  ${}^{85}$ Kr emissions originated from reprocessing plants). That is why  ${}^{85}$ Kr atmospheric activity is closely connected with nuclear fuel cycle following its strategy in a global scale.

Vacuum extraction was applied for collection of <sup>85</sup>Kr from primary water. Water sample was introduced into an evacuated bulb via capillary. A glass ampoule containing activated charcoal was used for collection of the gases. Noble gases were separated by titanium getter material. Radioactive noble gases other than krypton have short half life, therefore after 1 month of storage time they have negligible activity. Summary of the <sup>85</sup>Kr analysis of the reactor water is shown in Table 1. Several water samples were degassed from the primary cooling system of the four reactors of Paks NPP.

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Block	$^{85}$ Kr act. conc.[Bq/l]	$^{85}$ Kr act. conc.[Bq/l]
ecipitat	$9.85  imes 10^2$	$3.94  imes 10^2$
II	$4.22 \times 10^2$	$9.49 \times 10^{2}$
III	$1.30  imes 10^2$	$2.82  imes 10^3$
IV	$2.55  imes 10^2$	$1.07  imes 10^3$
Ι	$4.52 \times 10^2$	$1.12 \times 10^{3}$
II	$1.21  imes 10^3$	$2.03 \times 10^4$ ·
III	$3.03 imes10^2$	$1.65 \times 10^{3}$
IV	$7.80  imes 10^1$	$1.00 \times 10^{3}$
I	$6.35  imes 10^1$	$6.07 \times 10^2$ three distributions and the second s
II	_	$1.67  imes 10^3$
III	$9.81  imes 10^2$	$3.23  imes 10^4$
IV	$1.42 \times 10^2$	$1.88 \times 10^{3}$
I	$5.82 \times 10^1$	$2.20 \times 10^2$
II	$1.01  imes 10^2$	$6.02 \times 10^{2}$
III	$5.52  imes 10^2$	$1.38 \times 10^{3}$
IV	$1.01 \times 10^3$	11 (1997) 651-655 3

Table 1: <sup>85</sup>Kr activity concentration of the primary water

The average value was  $2.49 \text{ kBql}^{-1}$ .

20 l of air was collected in Plastigas. bags from the stacks of Paks NPP during each month with a dedicated system. The enrichment of krypton was done by performing adsorption at 196 °C and desorption (above 60 °C) in a charcoal trap with a volume of 10 cm<sup>3</sup>. The residual gases 2–3 ml were cleaned by titanium getter. Proportional counting technique was used for activity measurement. The activity concentrations are shown in Table 2.

Block I–II	Block III–IV	Block I–II	Block III–IV
<sup>85</sup> Kr activit	y conc. $[Bqm^{-3}]$	<sup>85</sup> Kr activit	by conc. $[Bqm^{-3}]$
6.16	6.88	72.27	_
_	69.66	65.66	65.70
6.43	9.84	75.19	77.41
3.87	24.58	69.99	71.15
14.44	52.03	49.37	56.11
16.00	20.92		

Table 2: The activity concentrations of <sup>85</sup>Kr in the stack air

The average <sup>85</sup>Kr activity concentration in the stacks of Paks NPP was 42 Bqm<sup>-3</sup>.

#### Conclusions

The normalised <sup>85</sup>Kr release of the plant is 0.3 TBq/GWey. The fission yields lead to <sup>85</sup>Kr production rates of  $1.7 \times 10^4$  TBq/GWey which means that a fraction of  $10^{-5}$  of the <sup>85</sup>Kr gas generated annually is released.

 $^{85}$ Kr is a very good atmospheric tracer for which sources should be known very precisely. Paks data show that a fraction of  $10^{-5}$  of the total inventory of  $^{85}$ Kr is released at the nuclear power plant which is much lower than the predicted rate.

[1] Zs. Szántó, É. Svingor, T. Pintér, Concentration of beta emitters in primary water, stack air, nuclear waste and environment of Middle European Nuclear Power Plants (in press) 204 of air was collected in Plastigas, bags from the stacks of Paks NPP during each month with a dedicated system. The enrichment of krypton was done by performing adsorption at 196 °C and desorption (above 60 °C) in a charcoal trap with a volume of 10 cm<sup>3</sup>. The residual gases 2–3 ml were cleaned by titanium getter. Proportional counting technique was used for activity measurement. The activity concentrations are shown in Table 2.

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<sup>25</sup> for is a very good strategiberic tracer for which sources should be known very precisely. Paks data show that a fraction of  $10^{-5}$  of the total inventory of  $^{16}$  Kr is released at the nuclear power plant which is much lower than the predicted rate.

[7] Zs. Szántó, E. Svingor, T. Pinter, Concentration of Letn emitters in primary water, stack air, nuclear waste and environment of Middle European Nuclear Power Plants (in press).

# **Biological and Medical Researches**



# Preparation of <sup>11</sup>C-labelled methanol on alumina column

É. Sarkadi, Z. Kovács, P. Lehikoinen<sup>\*</sup> and G. Horváth<sup>\*\*</sup> <sup>\*</sup> Åbo Academy, Turku, Finland

\*\* PET Center, University School of Medicine, Debrecen

The [<sup>11</sup>C]methyl iodide is an important intermedier to synthesize <sup>11</sup>C-labelled radiopharmaceuticals for medical diagnostics in positron emission tomography.

As a first step of the  $[^{11}C]$  methyl iodide preparation [1, 2] the classical radiomethanol production has been applied in our laboratory earlier [3]. Recently we have developed a new method to produce  $[^{11}C]$  methanol intermedier.

 $[^{11}C]$ carbon dioxide was produced by the  $^{14}N(p,\alpha)^{11}C$  nuclear reaction, irradiating nitrogen gas (containing oxigen in ppm level) with 15 MeV protons at the Debrecen and Turku compact MGC-20 cyclotrons. Trapping of the  $[^{11}C]$ carbon dioxide, produced and previously concentrated in a freezing unit, was carried out at room temperature on an alumina column properly impregnated with lithium aluminum hydride dissolved in tetrahydofuran. The whole alumina column had to be impregnated, otherwise the dry part of the alumina would adsorb the  $[^{11}C]$ carbon dioxide without complex formation (lithium aluminum  $[^{11}C]$ methylate) and, after heating the column up to 180 °C in order to eliminate the solvent, the unreacted  $[^{11}C]$ carbon dioxide would leave with the helium carrier gas.

After the elimination of tetrahydrofuran the  $[^{11}C]$  methanol was produced by hydrolyzing of the complex (with introduction of water) at 180 °C. Conversion of the released  $[^{11}C]$  methanol into  $[^{11}C]$  methyl iodide was carried out by the hydrogen iodide – alumina method described in [4].

The advantage of this method of radiomethanol preparation is the application of an alumina column at room temperature instead of a complicated cooling unit used with the conventional reaction vessel. The big surface of the alumina wetted with lithium aluminum hydride assures the shorter reaction time. The construction of this synthesis system is simple and more reliable. The yield and purity of radiomethanol was the same as in the previous methods.

 D. Comar, M. Maziere, Radiopharm. Lab. Comp. Vol. 1, IAEA, Vienna (1974) 461

- [2] G. Berger, M. Maziere, Appl. Radiat. Isot. 30 (1979) 393
- [3] É. Sarkadi, Z. Kovács, ATOMKI Annual Report (1994) 78
- [4] É. Sarkadi, Z. Kovács, Radiochimica Acta 76 (1997)

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### Development of Methods and Instruments

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## Application of the combined channeling method on cyclotron<sup>\*</sup>

F. Ditrói, J.D. Meyer,<sup>†</sup> R.W. Michelmann<sup>†</sup> and K. Bethge<sup>†</sup> <sup>†</sup> Institut für Kernphysik, Universität Frankfurt, FRG

The method of ion channeling combined with other analytical methods is widely used for investigation of highly ordered materials by using the low energy linear accelerators. The energy of the bombarding particles (p, d and He ions) extends to several MeV/nucleon. The use of a cyclotron with the channeling method makes it possible to apply higher bombarding energies (20 MeV maximum energy for protons in our case).

The scattering chamber to be used for the channeling measurements is situated after an analyzing magnet and a pair of slits. The beam path is calculated electronoptically so, that practically no collimation is necessary after the last slit of the analyzing magnet (approx. 5 m from the target position) to produce a spot of 1 mm<sup>2</sup> on the sample surface. This can lower the scattering-background and enhance the beam quality for channeling purposes.

The detectors are placed on a ring around the center of the chamber, and their distance from the target is also variable. The detectors can be moved on the ring by remote-controlled stepping motors. For positioning and rotating the samples a FISONS two-axis goniometer was used. The Z-axis and the primary and secondary rotations are also motorized (stepping-motor) and remote controlled. For testing and calibrating the irradiation and measuring equipment simple Si and GaAs crystals were used. Their surfaces were cut and polished perpendicular to main crystallographic directions ( $\langle 110 \rangle$ ,  $\langle 100 \rangle$ ).

The channeling-dip curve was taken around the main axes, and their FWHM was compared with the calculated values to study the conditions of the applicability of the channeling method at higher energies. The energy-loss formula for channeled particles can also be checked at the cyclotron-energies.

\* Supported by the Volkswagen Foundation and the Humboldt Foundation

Fig. 1: Typical charge state distributions without and with biased disk. This latter (dark columns) nowadays is typical in the ATOMKI-BCRIS and, generally, in 14 GHz sources. The source potential and the microwave power were 10 kV and 500 W, respectively. The ion source was optimised for Ar-8+ at both tests.

2. Beam development. The following ion beams were tosted and accelerated in the ECR ion source (in brackets the highest detected charge states with the total number of removable electrons are shown): N(7/7), O(8/8), Ar(13/18), Fe(15/26) and Xe(15/54). More details on the available ion beams (and generally on the

## The first year of operation of the ECR ion source

#### F. Ditroi, J.D. Meyer, R. Virid . Snelmann<sup>†</sup> and K. Bethge

Institut für Kernphysik, Universität I

1997 was the first full year of operation of the Electron Cyclotron Resonance (ECR) Ion Source of ATOMKI. The activity of the laboratory concentrated on three main fields.

1. Device development. The extraction optics was upgraded and now we can optimise the position of the extracting and focusing group of lens (on-line and independently) which is crucial when different ion species have to be extracted. The beam diagnostic block was modified and now all the highly charged ion beam intensity measurements became more reliable and reproducible. An axially movable, isolated metal electrode (called biased disk) was placed into the plasma chamber to produce secondary electrons and, consequently, to increase the plasma density. The result was dramatic: the highly charged ion beam intensities increased by a factor of 3–20 (depending on the charge state) and the charge state distribution shifted towards the higher charges.





2. Beam development. The following ion beams were tested and accelerated in the ECR ion source (in brackets the highest detected charge states with the total number of removable electrons are shown): N(7/7), O(8/8), Ar(13/18), Fe(15/26) and Xe(15/54). More details on the available ion beams (and, generally on the

whole ECR project) can be found on the home page of the laboratory [1]. The first generation and acceleration of highly charged ions from solid materials was also successfully performed [2].

3. Plasma research. The investigations followed three ways: visual observations of the plasma, X-ray measurements and visible light spectroscopy (both spectra analysis and total intensity studies). In every field promising first results were achieved.

#### [1] http://www.atomki.hu/atomki/ECR/

[2] see this Annual Report and AOR and AOR and the periodical system do not have unworking ass. However, most elements of the periodical system do not have unbarrent estimated in a components. Several techniques have been developed in the ECRUS community to ionize from solid materials. The most known methods are sputtering electrode heated over laser ablation and the volatile method. In our ECR ion source the volatile method was tested and successfully modified in the framework of a Hardenic finish Swedish collabor nor.



Fig. 1: A typical analysed Fe spectrum. The plasma was generated by nitrogen gas. The numbered peaks without names are different charge states of Fe-56. The relatively high carbon content comes from the ferrocene molecule. The satellite peaks of the Fe-54 isotope show the difficulty of analysing such spectrums.

# Highly charged Fe ions from the ECR ion source

S. Biri, A. Valek, H. Koivisto<sup>\*</sup> and U. Rosengard<sup>\*\*</sup> <sup>\*</sup> University of Jyväskylä, P.O.Box 35, FIN-40351 Jyväskylä, Finland <sup>\*\*</sup> Manne Siegbahn Laboratory, Freskativagen 24, 10405 Stockholm, Sweden

The easiest way to get plasma in ECR ion sources is to use gas substance as working gas. However, most elements of the periodical system do not have unharmful gas compounds. Several techniques have been developed in the ECRIS community to ionize from solid materials. The most known methods are: sputtering electrode, heated oven, laser ablation and the volatile method. In our ECR ion source the volatile method was tested and successfully modified in the framework of a Hungarian–Finish–Swedish collaboration.



Fig. 1: A typical analysed Fe spectrum. The plasma was generated by nitrogen gas. The numbered peaks without names are different charge states of Fe-56. The relatively high carbon content comes from the ferrocene molecule. The 'satellite' peaks of the Fe-54 isotope show the difficulty of analysing such spectrums.

The volatile method mostly known as the MIVOC-method [1] (Metal Ions from VOlatile Compounds). The vapours of volatile compound (having metal atoms in their molecular structure) are used to release metallic elements. The compound placed in a small, separated vacuum chamber is allowed to diffuse into the plasma which is usually generated by a 'common' gas (e.g. nitrogen). Decomposition and ionization of the compound then take place in the plasma.

In our case ferrocene (Fe( $C_5H_5$ )<sub>2</sub>) powder was used to get iron ions. Several ways of compound injection into the plasma were tested and highly charged (up to 15+) Fe-ions were successfully generated, extracted and analysed. The achieved beam charges and intensities can be found in the home page of the laboratory [2].

### [1] http://arje.jyu.fi/ECRIS/MIVOC/aa1.html

[2] http://www.atomki.hu/atomki/ECR/beams.htm

#### Improvements

Three turbomolecular pump stands have been installed as attended to our our diffusion pumps originally used for pumping of the beam transport tubes. I be ended turbopump stands were received as a present of Eahr University Bochum.

To avoid vibration caused by one of the old throe-printips an emicror transport tube dumping element has been placed between the pump and the beam transport tube of the microrredue

An oil-diffusion pump with a pumping speed of 2000 l/s has been connected parallel to the main-vacuum system of the  $\forall dG$ -5 machine, in order to decrease the residual gas concentration in the acceleration tube.

Aiming at lower bremsstrahlung, a new order of divider-chain resistors has been applied to the electron traps of the acceleration tube. Now the earlier used trapping electrodes are working on a decelerating lens-principle.

## Activities at the Van de Graaff accelerator laboratory

L. Bartha and E. Somorjai

In 1997 there was no request for the beam of the 1 MV machine. Its main vacuum system was also out of use. A modified hollow-cathode ionsource was running on the main beamline. It provided  $H^-$  ions for atomic physics purposes, in 200 hours estimated beam time.

The 5 MV Van de Graaff machine was operated for 1712 hours during this period. Protons (63%), <sup>4</sup>He<sup>+</sup> (25%), H<sub>3</sub><sup>+</sup> (6%) and <sup>12</sup>C<sup>+</sup> (6%) were accelerated. The last one was produced from 8% He + CO<sub>2</sub> source-gas mixture.

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.

Field	Hours	%
Atomic physics	590	34.7
Nuclear physics	55	3.2
Analytical studies	194	11.5
Analytics on the microprobe	598	35.2
Education	233	13.7
Machine tests	29	1.7
Total	1699	100.0

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

#### Improvements

Three turbomolecular pump stands have been installed as alternatives of oildiffusion pumps originally used for pumping of the beam transport tubes. These old turbopump stands were received as a present of Ruhr-University, Bochum.

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## Status Report on the Cyclotron

#### Z. Kormány

The operation of the cyclotron in 1997 was concentrated again to 9 months; January, July and August were reserved for maintenance and holidays. The overall working time of the accelerator was 3946 hours with break down periods amounted to 87 hours. The cyclotron was available for users during 3409 hours, the effectively used beam time is summarized in Table 1. (FERMI: Front-End Readout Microsystems, Radiation hardness measurements, CERN RD-16). The time used for the beam preparation and waiting for the start of an irradiation totalled to 588 hours.

Parallel with the routine operation a large-scale modernization project has been in progress in the frame of a Technical Assistance Program (project code number HUN/4/013) of the International Atomic Energy Agency, which involves the renewal of the vacuum and control systems of the cyclotron. During the summer maintenance period the vacuum measurement subsystem was completely renewed and the control of the safety interlock system was successfully removed from the old fixed-wired electronics to the programmable logic controller.

Projects	Time (hours)	%
Nuclear spectroscopy	314	27
Nuclear astrophysics	161	14
FERMI	114	10
Nuclear data	25	2
Isotope production	554	47
Total	1168	100

Table 1: Distribution of the effectively used beam time

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The operation of the cyclotron in 1997 was concentrated again to 9 months: fannary, fuir and August were reserved for maintenance and holidays. The overall working time of the accelerator was 3946 bours with break down periods amounted to 37 hours. The cyclotron was available for users during 3409 hours, the effectively used beam time is summarized in Table 1. (RERMI: Front-End Readout Microsystrons, Radiation hardness measurements, CERM, RD-16). The time used for the beam periodic and waiting for the start of an irrediation to allected to 38 terms

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Table I: Distribution of the off or ively used brane time

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R<sub>b</sub> using a double tagging method. Zeitschrift für Physik "C" 74 (1997) 1. sepral diw straves zinotodd :.ls ts, L sàknilåd ,.d dtàvroll ,.M flatsrahA .S **fêt** <sup>A</sup>B<sup>(e</sup>srähhL szlzvHP<sup>(t</sup>V aD fð1<sup>(ink</sup> k) ts etnoisillo3<sup>a</sup> at <sup>i</sup> versis finskim<sup>tales</sup> and the photon structure function F<sup>2</sup> gamma in deep inelastic .0<sup>f</sup>2<sup>(f</sup>(7001)<sup>oton</sup> scattering at LEP. Zeitschrift für Physik "C" 74 (1997) 33.

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# **Theses Completed**

## **Doctoral Degree (PhD)**

- Borbély-Kiss I.: A PIXE módszer és alkalmazásai a légköri aeroszol kutatásában és más interdiszciplínáris területeken. Doktori (PhD) értekezés. Debrecen, Kossuth Lajos Tudományegyetem (1997) 75.
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# Seminars in 1997

March 27

Activity concentration of beta-decaying isotopes in the primary circle of the nuclear power plant, in the air of its chimney, in radioactive waste and in the environment of the power plant E. Hertelendi

### April 3

The concept of effective minimal basis and the theoretical interpretation of the Gillespie-VSEP rules I. Mayer (KKKI, Budapest)

#### April 17

After collision interactions in fast ion-atom collision processes Gy. Víkor

April 24

Interaction of high intensity cyclotron beams with gas targets F. Tárkányi

May 8

On the proteins by the eyes of a mathematician G. Tusnadi (KLTE, Debrecen)

May 28

Accelerator based R&D programs at Chiang Mai T. Vilaithong (Chiang Mai, Thailand)

May 29

Transillumination of transillumination (Bottom-view of scientometrics), Part I L. Zolnai

#### June 5

Transillumination of transillumination (Bottom-view of scientometrics), Part II L. Zolnai

June 19

Dynamical symmetries: from molecules to quarks F. Iachello (Yale University, USA)

June 12 Fight against the standard model D. Horváth August 28

Developments in electron spectroscopy and applications in surface analysis J. Tóth

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September 25

Radon and uranium survey and microseismic studies in the N-W Himalaya H. S. Virk (Amritsar, India)

October 2

Radon and space radiation safety measurements I. Csige

October 14

Development of short lived radio tracers H.H. Coenen (Jülich)

October 30 X-ray diffraction structure analysis at the Lajos Kossuth University A. Bényei (KLTE, Debrecen)

November 6 Eugene Wigner and the sixty years of nuclear symmetries J. Cseh

November 20 EXOGAM: a way in the future with radioactive beams B. Nyakó

November 27 Nuclear physics with radioactive beams destructions of the server additional and the server additional additional and the server additional additio

December 11 *RBS-measurements on nuclear micro probe* A. Simon

December 18 The dynamics of atomic collisions reflected by the angular and energy distribution of outgoing electrons B. Sulik

> Dynamical symmetries: from molecules to quarks F. Iachello (Yale University, USA)

> > June 12 Fight against the standard model D. Horváth

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