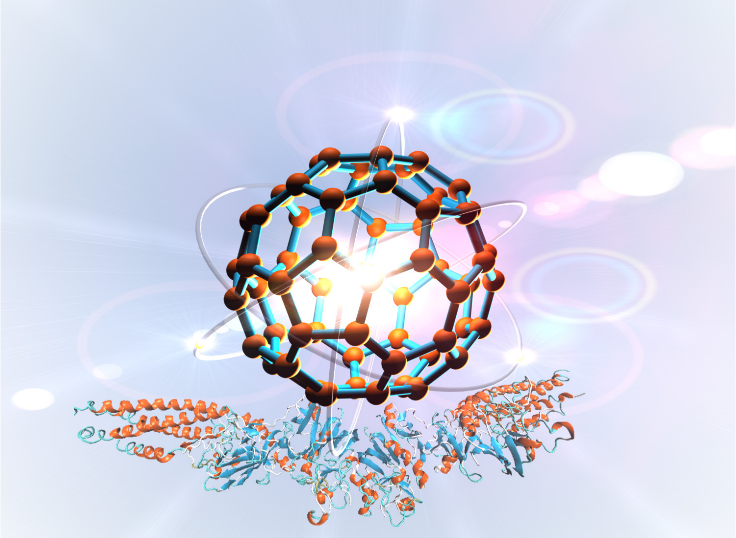

ISACC 2007

International **S**ymposium **A**tomic **C**luster **C**ollisions: structure and dynamics from the nuclear to the biological scale

Darmstadt (Germany), July 19-23, 2007

Editor: Andrey V. Solov'yov

Abstracts

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Preface

The Second International Symposium "Atomic Cluster Collisions: structure and dynamics from the nuclear to the biological scale" (ISACC 2007) is a satellite meeting of the XXVth International Conference on Photonic, Electronic, and Atomic Collisions (ICPEAC 2007, Freiburg, Germany). This symposium is organized by the Frankfurt Institute for Advanced Studies (FIAS) and Gesellschaft für Schwerionenforschung (GSI) and will be held on July 19 - 23, 2007 at GSI, Darmstadt, Germany. ISACC has been recognized by the European Physical Society (EPS) as a Europhysics Conference.

ISACC started as an international symposium on atomic cluster collisions in St. Petersburg, Russia in 2003. The ISACC 2007 will promote the growth and exchange of scientific information on the structure and properties of nuclear, atomic, molecular, biological and complex cluster systems studied by means of photonic, electronic, heavy particle and atomic collisions. In the symposium, particular attention will be devoted to dynamical phenomena, many-body effects taking place in clusters, molecular and biological systems, which include problems of fusion and fission, fragmentation, collective electron excitations, phase transitions and many more. Both experimental and theoretical aspects of cluster physics, uniquely placed between nuclear physics on one hand and atomic, molecular and solid state physics on the other, will be subject of the symposium.

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We also acknowledge assistance from Ms. Stephanie Lo and Ms. Adilah Hussein, FIAS Frankfurt am Main, in the preparation of the book of abstracts.

Conference Program

July 19 - 23, 2007

Thursday, 19 July 2007

9^{00} – 9^{30}		**ISACC-2007 Opening** **S. Hofmann, A. Solov'yov, H. Stöcker**
9^{30} – 11^{00}	**Th-I**	**Clustering phenomena at nuclear and subnuclear scales (part I)** **Yu. Oganessian** – Synthesis and decay properties of the heaviest nuclei **S. Hofmann**– Production and decay of superheavy nuclei **V. Zagrebaev** – Clustering phenomena in fission and fusion processes of heavy nuclei
11^{00} – 11^{20}		**Coffee break**
11^{20} – 12^{50}	**Th-I**	**Clustering phenomena at nuclear and subnuclear scales (part II)** **W. Scheid –** Nuclear molecules **T. Neff –** Shell structure, clustering and halos nuclei in the fermionic molecular dynamics approach **T. Bürvenich –** Properties of heavy and superheavy nuclei in supernova environments
12^{50} - 14^{00}		**Lunch**
14^{00} - 15^{40}	**Th-II**	**Structure and properties of atomic clusters (part I) :** **J.-P. Connerade** – Relating clusters to giant atoms **J. Jellinek –** Metal and semiconductor clusters: size-driven evolution towards metallicity and half-metallicity **A. Lyalin** – Magnetism in clusters **P. Lievens –** Physical and chemical properties of binary clusters
15^{40} – 16^{00}		**Coffee break**
16^{00} – 18^{00}	**Th-II**	**Structure and properties of atomic clusters (part II)** **S. Khanna** – From designer clusters to synthetic cluster assemblies **B. Johnson –** Nanoparticles, nanotubes and related chemistry **M. Moseler –** Electronic structure and thermodynamics of gold clusters **D. Poenaru** – New deformed single-particle shell model **M. Lopez –** Molecular adsorption on carbon nanotubes
18^{00} - 19^{00}	**Th-III**	**Poster session (coffee and snacks are provided during the poster session)**

Friday, 20 July 2007

9^{30} – 11^{00}	**Fr-I**	**Electron & photon cluster collisions (part I)** **M. Broyer –** Photofragmentation and photodetachement of biomolecules and cluster-biomolecules complex **T. Märk** – Electron scattering on clusters and biomolecules embedded in helium droplets **B. von Issendorff –** Angular-resolved photoelectron spectroscopy of cold sodium cluster anions
11^{00} - 11^{20}		**Coffee break**
11^{20} - 12^{50}	**Fr-I**	**Electron & photon cluster collisions (part II)** **A. Korol** – Plasmons and the dynamical screening of endohedral atoms **U. Reinköster** – Photoabsorption and photoionization of fullerenes and clusters **A. Müller –** Photo-ionization and fragmentation of fullerene ions
12^{50} - 14^{00}		**Lunch**
14^{00} – 15^{40}	**Fr-II**	**Ion-cluster collisions** **M. Ichihashi, T. Hanmura, and T. Kondow** – Collision of transition metal cluster ions with simple molecules **B. Huber** – Fragmentation dynamics of charged finite-size systems **H. Zettergren –** Fragmentation of multiply ionized fullerene dimers **M. Alcami –** Structure and fragmentation of fullerenes and fullerene dimmers produced in collisions with ions
15^{40} – 16^{00}		**Coffee break**
16^{00} - 18^{00}	**Fr-III**	**T. Haberer** – The impact of nuclear interactions on ion beam tumor therapy **Group photo, Excursion to GSI**

Saturday, 21 July 2007

9^{30} - 11^{30}	**Sa-I**	**Clusters on surface** **A. Nakajima –** Fine controlled deposition of functional binary clusters onto a designed surface **K.-L. Choy –** Interaction of nanoclusters with various surfaces **M. Pellarin –** Size selected clusters soft landed on a surface: morphology and organization **P. Berke –** Simulation of the nanoindentation procedure on Nickel on the smallest length scale: a simple atomistic level model
11^{30} – 11^{50}		**Coffee break**
11^{50} – 13^{20}	**Sa-II**	**Phase transitions, fusion, fission and fragmentation in finite systems (part I)** **C. Bréchignac** –Nanofractals, selfassembling **F. Gianturco** – Flying cryostats: bosonic helium droplets with molecular dopants. Selective localization and quantum effects. **H. Haberland** – Melting of small metal clusters: thermodynamic on a finite scale
13^{20} - 14^{20}		**Lunch**
14^{20} - 16^{00}	**Sa-II**	**Phase transitions, fusion, fission and fragmentation in finite systems (part II)** **A. Yakubovich –** Conformational changes in polypeptides /phase transition **M. Schmidt –** Experiments on atomic clusters: boiling and sublimation **F. Calvo** – Multiscale approach to cluster fragmentation **J.-M. L'Hermite** – Atom by atom nucleation of sodium and caloric curves
16^{00} – 16^{20}		**Coffee break**
16^{20} – 18^{00}	**Sa-III**	**Clusters in laser fields** **A.W. Castleman** – Ground state ion pair formation and excited state biradical formation in $(NI)(H_2O)_n$ clusters **M. Rost** – Helium embedded rare gas clusters under a strong laser pulse **P.-G. Reinhard –** Large-amplitude dynamics of clusters **A. Lindinger –** Optimal control on alkali systems
19^{30} - 22^{30}		**Conference Banquet at the Restauraunt "Zum Schwarzen Stern",** **Römerberg 6, 60311, Frankfurt am Main**

Sunday, 22 July 2007

Guided tour to the Heavy ion therapy centre in Heidelberg and a boat trip over the Neckar river.

Monday, 23 July 2007

9^{30} - 11^{10}	**Mo-I**	**Clustering phenomenon in systems of various degrees of complexity** **V. Ivanov –** Electron-positron clusters: structure and stability **P. Hvelplund –** Formation and stability of gas phase multiply charged (+/-) clusters of biomolecules **L. Andersen –** Spectroscopy and decay of chromophore ions in the gas phase **D. Wales –** Energy landscapes: structure, dynamics and thermodynamics
11^{10} – 11^{30}		**Coffee break**
11^{30} – 12^{45}	**Mo-II**	**Structure and dynamics of bio-macromolecules** **H. Hotop**–Vibrational Feshbach resonances in low-energy electron collisions with molecules and molecular clusters **O. Obolensky –** Structure of unfolded polypeptides **E. Henriques –** Computational simulations of antibody:antigen unbinding **I. Solovyov –** Cryptochrome magnetoreception
12^{45} – 13^{45}		**Lunch**
13^{45} - 15^{25}	**Mo-III**	**Collision processes involving biomolecules** **T. Schlathölter –** Ion induced fragmentation of nucleobase and amino-acid clusters **T. Elsässer –** Biophysical modelling of fragment distributions of DNA plasmids after photon and heavy ion irradiation **M. Scholz –** The increased biological effectiveness of ion beam radiation: Experimental and theoretical aspects
15^{25} – 15^{45}		**Coffee break**
15^{45}-17^{05}	**Mo-IV**	**From biomolecules to cells and system biology** **I. Pshenichnov –** Towards Monte Carlo calculations of biological dose in heavy-ion therapy: modeling of nuclear fragmentation reactions **E. Surdutovich –** Mechanisms of radiation damage of biomolecules **M. Meyer-Hermann –** Organization of transmembrane molecules in immunological synapses **A.M. Samsonov –** On modelling of gene expression patterns in Drosophila embryo
17^{05} – 17^{30}		**Closing symposium** **Andrey Solov'yov**

Overview of Abstracts

Clustering phenomena at nuclear and subnuclear scale

Structure and properties of atomic clusters

Electron and photon cluster collisions

Ion-cluster collisions

Clusters on surface

Phase transitions, fusion, fission and fragmentation in finite system

Clusters in laser fields

Clustering phenomenon in systems of various degrees of complexity

Structure and dynamics of bio-macromolecules

Collision processes involving biomolecules

From biomolecules to cells and system biology

Posters

Invited Speakers

Clustering phenomena at nuclear and subnuclear scale

Th-I-1

SYNTHESIS AND DECAY PROPERTIES OF THE HEAVIEST NUCLEI

Yu. Oganessian[1]

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Predictions of microscopic models on the limits of nuclear masses and on the existence of "islands of stability" in the region of hypothetical superheavy elements are tested in the experiments on the synthesis of nuclei with Z=104-118 and N=161-177 in the reactions induced by ^{48}Ca. The properties of new nuclides, together with the data for the neighboring isotopes with atomic numbers Z≤113 produced in cold fusion reactions, point to a strong enhancement of nuclear stability in the vicinity of predicted neutron shells N=162 and 184.

Relatively long half-lives of the new nuclides open possibilities for the study of the strucute of superheavy atoms, in particular chemical properties of the new elements. Experiments on the observation of the so-called "relativistic effect" in the electron structure of elements 112 and 114 using express radiochemical methods are now underway at the ^{48}Ca ion beam.

The search for the most long-lived superheavy elements in nature is aimed at the registration of decay of element 108 isotopes. Preliminary results of measurements of spontaneous fission rare events in the underground laboratory at Modane (France) are discussed in context of the maximal nuclear half-lives at the top of the island of stability and in context of setting up new experiments.

The study of superheavy nuclei is carried out using a set-up DGFRS installed at the beam of heavy ion accelerator at the FLNR (JINR, Dubna) in collaboration with the colleagues from LLNL (Livermore) and PSI (Villigen); experiments in the underground laboratory – in collaboration with CSNSM (Orsay).

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PRODUCTION AND DECAY OF SUPERHEAVY NUCLEI

S. Hofmann[1]

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The nuclear shell model predicts that the next doubly magic shell-closure beyond ^{208}Pb is at a proton number Z = 114, 120, or 126 and at a neutron number N = 172 or 184, see figure 1. The outstanding aim of experimental investigations is the exploration of this region of spherical 'SuperHeavy Elements' (SHEs). Experimental methods are described, which allowed for the identification of elements produced on a cross-section level of about 1 pb. The decay data reveal that for the heaviest elements, the dominant decay mode is alpha emission, not fission. Decay properties as well as reaction cross-sections are compared with results of theoretical investigations. Finally, plans are presented for the further development of the experimental set-up and the application of new techniques, as for instance the precise mass measurement of the produced nuclei using ion traps. At increased sensitivity, the detailed exploration of the region of spherical SHEs will start, after first steps on the island were made in recent years.

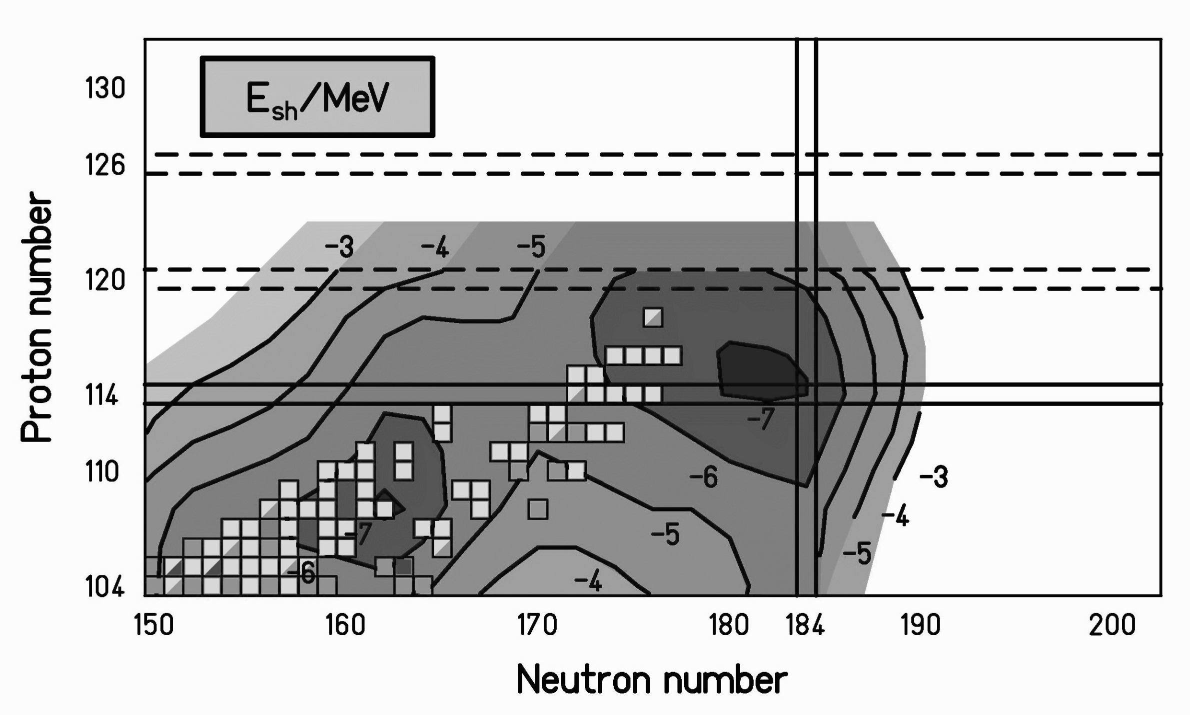

Figure 1: The upper end of the chart of nuclei showing the shell correction energy as calculated by macroscopic-microscopic models and the presently known nuclei up to element 118.

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CLUSTERING PHENOMENA IN FISSION AND FUSION PROCESSES OF HEAVY NUCLEI

Valery Zagrebaev[1] and Walter Greiner[2]

[1]Flerov Laboratory of Nuclear Reactions, JINR, Dubna,Moscow Region, Russia
[2]Frankfurt Institute for Advanced Studies, J.W. Goethe-Universität, Frankfurt, Germany

We applied the Langevin type equation of motions and the modified two-center shell model for description of the strongly coupled processes of compound nucleus formation, fission and quasi-fission. Among other things, the model allows us to perform an analysis both in the space of "$R, \eta, \delta_1, \delta_2$" (elongation, mass asymmetry, deformations of the fragments) and in the space "$a_1, \delta_1; a_2, \delta_2$", because for a given nuclear configuration "$R, \eta, \delta_1, \delta_2$" we may unambiguously determine the two cores a_1 and a_2 (see the figure). This turns out to be extremely important for interpretation of physical meaning of some deep minima on the potential energy surface.

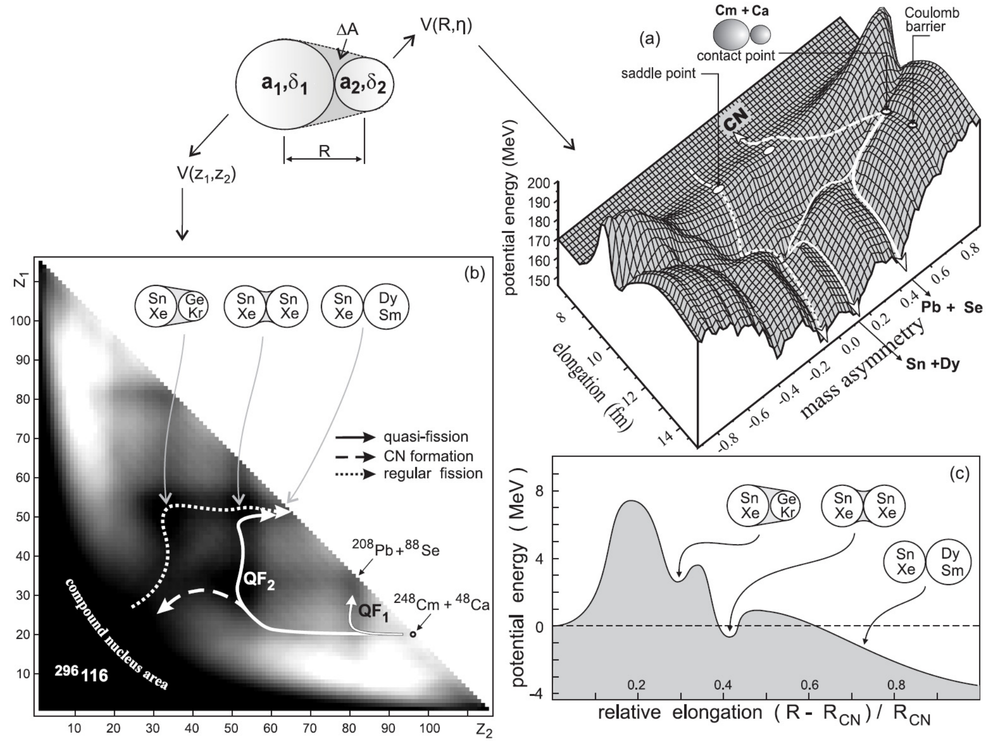

Figure 1: Driving potential of nuclear system $^{296}116 \leftrightarrow {}^{48}\mathrm{Ca} + {}^{248}\mathrm{Cm}$. (a) "Elongation – mass asymmetry" space. (b) Landscape of the driving potential on (z_1, z_2) plane. (c) Three-humped barrier calculated along the fission path (dotted curve).

The shell structure, clearly revealing itself in the contact of two nuclei is also retained at $R < R_{\mathrm{cont}}$ (see the deep minima in the regions of $z_{1,2} \sim 50$ and $z_{1,2} \sim 82$ in Fig. 1b). Following the fission path (dotted curves in Fig. 1a,b) the nuclear system goes through the optimal configurations (with minimal potential energy) and overcomes the multi-humped fission barrier (Fig. 1c). These intermediate minima correspond to the shape isomer states. Now we may definitely conclude that these isomeric states are nothing else but the two-cluster configurations with magic or semi-magic cores surrounded with a certain number of shared nucleons.

Th-I-4

NUCLEAR MOLECULES

G. G. Adamian[1], A. V. Andreev[1], N. V. Antonenko[1], S. P. Ivanova[1],
R. V. Jolos[1], A. K. Nasirov[1], W. Scheid[2], T. M. Shneidman[1] and A. S. Zubov[1]

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A nuclear molecule or a dinuclear system (DNS) is a configuration of two touching nuclei (clusters) which keep their individuality. Such a system occurs in nuclear structure, in heavy ion collisions and in fission. It has two main degrees of freedom which govern its dynamics: (1) *the relative motion* between the nuclei giving rise to molecular resonances in the internuclear potential, observed e. g. in the scattering of ^{12}C on ^{12}C, and to the decay of the dinuclear system which is called quasifission and (2) *the transfer of nucleons* between the nuclei leading to a dependence of the dynamics on the mass and charge asymmetries in fusion and fission reactions. The latter processes are described by the mass and charge asymmetry coordinates $\eta = (A_1 - A_2)/(A_1 + A_2)$ and $\eta_Z = (Z_1 - Z_2)/(Z_1 + Z_2)$. These coordinates can be assumed as continuous or discrete quantities. For $\eta = \eta_Z = 0$ we have a symmetric clusterization with two equal nuclei, and if η approaches the values ± 1 or if A_1 or A_2 is equal to zero, a fused system has been formed.

In this contribution we consider more recent aspects of molecular configurations playing a role in nuclear structure and reactions. The work is based on the DNS model proposed by V. V. Volkov. We give a short and concise review on several applications of the DNS model, namely we discuss hyperdeformed states, normal- and superdeformed bands, the fusion dynamics in producing superheavy nuclei, multi-nucleon transfer between nuclei, and the decay of the dinuclear system in fission and quasifission. The concept of the nuclear molecule gives a simple intuitive explanation of complex nuclear collective dynamics and is valuable for far-reaching predictions in nuclear physics.

Th-I-5

Shell Structure, Clustering and Halos Nuclei in the Fermionic Molecular Dynamics approach

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Clustering has been known to be important for the structure of ground and excited states of nuclei. Especially states close to the corresponding thresholds are expected to show pronounced clustering. In the Ikeda diagram [1] cluster states in α-nuclei built with ^{4}He, ^{12}C, ^{16}O clusters are predicted.

In the Fermionic Molecular Dynamics approach [2] we describe the many-body wave function with Slater determinants of Gaussian wave packets. The wave packets can easily be arranged in phase space such that they describe various kinds of cluster states. On the other hand they also allow the description of shell model like configurations. By superimposing wave packets of different widths we are also able to describe nuclear halos.

We use an effective nucleon-nucleon interaction that is derived from the realistic Argonne V18 interaction by explicitly including short-range central and tensor correlations that are essential in nuclear physics.

We will present a recent study [3] of the ^{12}C system with a special emphasis on the structure of the excited 0^+ and 2^+ states. Even in the most advanced calculations these excited states can not be described in a no-core shell model approach. α-cluster models on the other hand suffer from very simple effective interactions and an unsatisfactory description of the low-lying states where the α-clusters are broken by the spin-orbit force. Our FMD calculations make a quantitative analysis of the degree of α-clustering in the low-lying and excited states of ^{12}C possible and confirm, taking inelastic electron scattering data into account, the conjecture that the Hoyle state has to be understood as a loosely bound system of α particles.

We will also present first results on the application of the FMD model for low-energy nuclear reactions. The FMD basis allows for a consistent description of bound states, resonances and scattering states. Asymptotically, states are described like in microscopic cluster models. A new collective coordinate method was developed to include the boundary conditions. In the interaction region distortion effects are included by additional FMD configurations. The S-factor for the ^{3}He$(\alpha,\gamma)^7$Be reaction is calculated.

References

[1] K. Ikeda, N. Tagigawa, H. Horiuchi, Prog. Theor. Phys. Suppl. Extra number (1968) 464.

[2] R. Roth, T. Neff, H. Hergert, and H. Feldmeier, Nucl. Phys. **A745**, 3 (2004).

[3] M. Chernyk, H. Feldmeier, T. Neff, P. von Neumann-Cosel, A. Richter,
Phys. Rev. Lett. **98**, 032501 (2007).

Th-I-6

Properties of heavy and superheavy nuclei in supernova environments

Thomas Bürvenich[1], Igor N. Mishustin[1,2] and Walter Greiner[1]

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[2] *Kurchatov Institute, Russian Research Center, 123182 Moscow, Russia*

In this talk we present results concerning the structure and stability of nuclei embedded in the hot and dense stellar matter, calculated within a relativistic mean-field model. The emphasis is made on possible formation of heavy and superheavy nuclei in supernova environments. The electron gas is treated as a constant background in the Wigner-Seitz cell approximation. The presence of electrons leads to two important changes in the nuclear properties. First, due to the attractive interaction with electrons the, proton levels shift downwards as compared with the vacuum. Second, the nuclear Coulomb potential is reduced due to the screening effect of electrons. As the result, the proton dripline shifts to more proton-rich systems, and the stability line with respect to β-decay moves to more neutron-rich nuclei, and finally reaches the neutron dripline. This signals appearance of free neutrons in the system (the ”neutronization” process). We investigate the stability of nuclei with respect to α and β decay, as well as to the spontaneous fission. We find that the presence of the electrons leads to stabilizing effects for both α decay and spontaneous fission for high electron densities. Implications for the existence and creation of very heavy systems are discussed. Our main conclusion is that the formation of heavy and superheavy elements (SHE) is favored by the electron screening in dense stellar matter. Therefore, one can expect that these elements can be synthesized in supernova environments and ejected in space in the course of explosions. If long-lived SHE exist, they may be present in cosmic rays.

Structure and properties of atomic clusters

Th-II-1

RELATING CLUSTERS TO GIANT ATOMS

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One of the challenges posed by the demand for clean urban transportation is the compact and cyclically recoverable storage of energy in quantities sufficient for propulsion. Two promising routes are (a) the reversible insertion of Li+ ions inside solids for 'rocking chair' batteries or (b) the reversible storage of hydrogen inside suitable materials. Either way, the requirement is for a deformable host material with no irreversibility, which means no breaking of chemical bonds and (in principle) no change in the number of microstates. Such 'soft' deformations (corresponding to the slow polaronic distortion of a lattice) are in general highly complex, but the compressibility of atoms or larger systems can be studied directly in situations with simpler symmetry. Thus, the search for 'soft' materials leads one to consider certain types of cluster (considered here as a small piece of idealised solid), as well as linear or nearly-spherical structures (chains of metallofullerenes, for example) whose deformations (or elongations) can be computed from the Schrodinger equation. For this purpose, extended or 'giant' atomic models are very useful: the electronic wavefunctions of such systems can be regarded as a compressible working fluid with rather general properties. Such models allow one to construct compression-dilation cycles analogous in a rough sense to the Carnot cycle of classical thermodynamics. This simplified approach approach suggests that, even for idealised systems, there are constraints on the reversible storage and recovery of energy, and that (when applied to realistic structures) modelling based on such principles might help in the selection of appropriate materials.

Th-II-2

METAL AND SEMICONDUCTOR CLUSTERS: SIZE-DRIVEN EVOLUTION TOWARDS METALLICITY AND HALF-METALLICITY[*]

Julius Jellinek

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I will present and discuss results on the phenomena of size-driven evolution towards metallicity and half-metallicity in clusters of nominally metallic and semiconductor elements. The analysis will be based on data obtained within gradient-corrected density functional theory and will include the roles of the cluster material, structure, and charge state. The emphasis will be on the spectra of electron binding energies and dipole polarizabilities as indicators of metallic and half-metallic attributes in finite systems.

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[*] This work was supported by the Office of Basic Energy Scienceas, Division of Chemical Sciences, Geosciences, and Biosciences, U.S. Department of Energy under Contract number DE-AC-02-06CH11357.

Th-II-3

MAGNETISM IN CLUSTERS

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The study of evolution of magnetic properties from atoms to the bulk is important for the development of magnetic nanomaterials with specific properties and for understanding the fundamental principles of spin coupling in finite and low dimensional systems. In this work we have performed a systematic theoretical investigation of optimized ionic structure, electronic and magnetic properties of La clusters within the size range N < 14 [1]. We found a giant enhancement of magnetism in La_4, La_6, and La_{13} clusters. We also found that the ground states of La_2, La_3, La_5, La_7, La_9-La_{11}, and La_{14} clusters possess nonzero magnetic moments, that ranged from 0.1 μ_B to 1.0 μ_B per atom. This clearly indicates that small La clusters display magnetic behavior, even though bulk La has no magnetic ordering. We show that magnetism in La clusters is governed by unpaired valence electrons, in contrast to the local-moment magnetism in clusters of heavy rare-earth elements. In addition to the ground state isomers of La clusters we found an ensemble of energetically low-lying spin isomers. We predict an increase of the average magnetic moments for ensembles of La_2, La_3, La_5, La_8, La_9, La_{11}, La_{12}, and La_{14} clusters with temperature due to the thermal population of the spin isomers. For ensembles of La_4, La_7, and La_{13} clusters, the average magnetic moment decreases with temperature. Such an anomalous behavior of the magnetic moment with temperature can be detected in Stern-Gerlach deflection experiments.

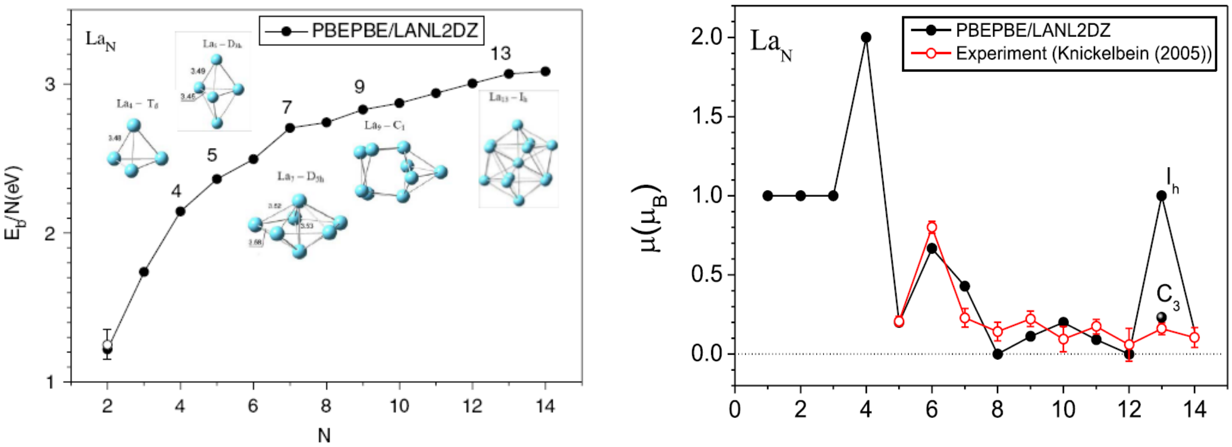

Figure 1. Binding energy per atom for the most stable La clusters (left); magnetic moments per atom for La clusters as a function of cluster size. Open circles present the results of experiment by Knickelbein [2] (right).

We report the results of a systematic theoretical investigation of optimized structure, electronic and magnetic properties of linear chains of Mn atoms and organometallic sandwich clusters $Mn_N(C_6H_6)_{N+1}$ within the size range $N \leq 6$ [3]. The choice of Mn is stipulated by the fact that the Mn atom possesses a large magnetic moment due to the half-filled 3d electron shell, and thus, manganese is a good candidate for strong nanomagnets. Our calculations are based on *ab initio* theoretical methods invoking density-functional theory with the gradient-corrected exchange-correlation functional of Perdew, Burke and Ernzerhof (PBEPBE). The standard LANL2MB basis set of primitive Gaussians have been used to expand the electronic orbitals formed by the $3s^2 3p^6 3d^5 4s^2$ outer electrons of Mn (15 electrons per atom). We show that the finite linear chains of

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Mn atoms exhibit novel magnetic properties that differ from those of the corresponding Mn bulk and one-dimensional infinite chains. Thus, we demonstrate that finite one-dimensional monoatomic chains of Mn atoms possess ferromagnetic order in spite of the fact that the most stable crystal structure of Mn exhibits antiferromagnetic behaviour. We demonstrate that magnetic ordering in finite linear chains depends on its length and therefore can be controlled by the fixing chain's geometry. We also predict enhancement of magnetism in organometallic sandwich clusters $Mn_N(C_6H_6)_{N+1}$.

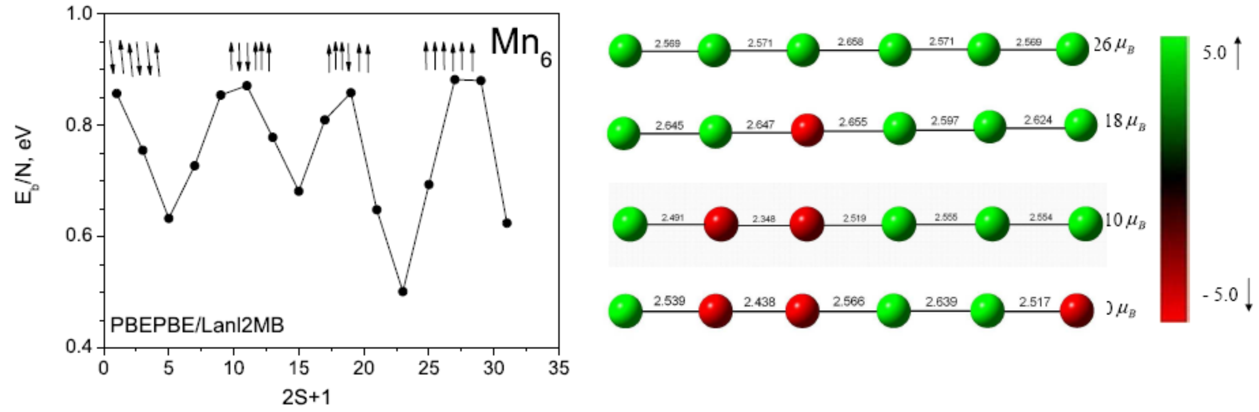

Figure 2: Binding energy per atom for Mn_6 linear chain as a function of multiplicity 2S+1 (right). Mulliken atomic spin densities for different spin isomers of Mn_6 chain (left). Interatomic distances are given in angstroms.

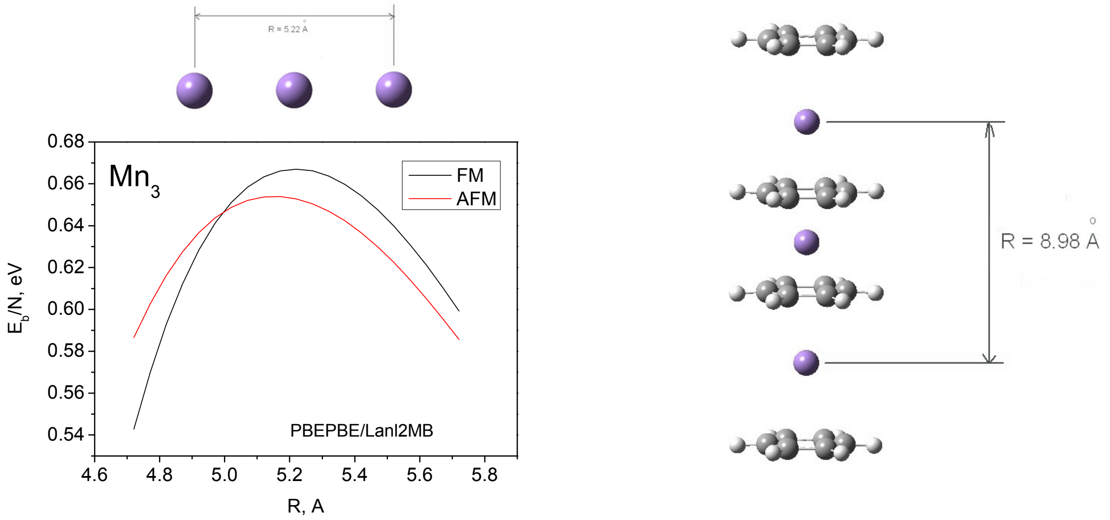

Figure 3: Binding energy of linear chain as a function of length (left); magnetic organometallic sandwich $Mn_3(C_6H_6)_4$ (right).

This work is partially supported by the European Commission within the Network of Excellence project EXCELL, and by INTAS under the Grant No. 03-51-6170.

REFERENCES

[1] A. Lyalin, A. V. Solov'yov and W. Greiner, Phys. Rev. A **74**, 043201 (2006).
[2] M. B. Knickelbein, Phys. Rev. B **71**, 184442 (2005).
[3] A. Lyalin, A.V. Solov'yov and W. Greiner, in preparation for Phys. Rev. A (2007).

Th-II-4

PHYSICAL AND CHEMICAL PROPERTIES OF BINARY CLUSTERS

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Introducing a single transition metal atom into a small noble metal cluster may modify the electronic shell structure significantly. The strength of the interaction between host cluster and dopant atom is strongly determined by the size of the dopant (geometric stabilization) and the number of dopant valence electrons (electronic shell closures). The combined effect of these parameters is reflected in the total binding energy and the differential binding energies or dissociation energies.

We will discuss a series of studies on transition metal doped gold, silver and copper clusters, produced with a dual-target dual-laser vaporization source. Size and composition dependent stability fluctuations are investigated with photofragmentation and mass spectrometry. Dissociation pathways of mass selected doped gold clusters were studied with delayed fragmentation yield measurements. The altered electronic structure of the doped clusters also is reflected in their magnetic properties, e.g., with computational evidence of quenching of the dopant magnetic moments for specific doped silver species. Altered chemical properties are evidenced by reactivity studies of doped gold and silver clusters with carbon monoxide.

Acknowledgements. This work is supported by the Fund for Scientific Research - Flanders (FWO), by the Flemish Concerted Action (GOA), and the Belgian Interuniversity Attraction Poles (IAP) research programs.

Th-II-5

FROM DESIGNER CLUSTERS TO SYNTHETIC CLUSTER ASSEMBLIES

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A promising developments in the field of clusters and nanoscience is the possibility of synthesizing nanoscale materials where size specific clusters serve as the elementary molecular building blocks. As the physical, chemical, electronic, and magnetic properties of clusters can be tuned by size and composition, this may provide an unprecedented ability to design customized materials. The cluster materials, in addition, possess intra-cluster and inter-cluster length scales leading to novel functionalities not available in conventional materials. While this thought has existed for more than two decades, its realization has been delayed by the reality that clusters are usually metastable and coalesce when assembled. Indeed, the only working example is the alkali-doped-fullerides where fullerenes marked by directional C-C bonds serve as the primary unit. Extending the fullerene experience to other metal and semi-conducting systems is likely to emerge as an important frontier in the nanoscience.

The talk will focus on our efforts in this direction. I will first talk about the possibility of forming cluster motifs, based on metals, that are fairly stable and maintain their identity upon further growth. A class of these can be classified as superatoms forming a new dimension to the periodic table [1-3]. I will then present a new three step protocol that combines (1) gas phase investigations to examine feasible units, (2) theoretical investigations of energy landscapes and geometrical shapes of feasible units to identify potential motifs, and (3) synthetic chemical approaches to synthesize and structurally characterize such cluster assemblies in the solid state. Through this approach we have established selected arsenic-alkali cluster as a potential building block via gas phase molecular beam experiments. Employing the idea that the particular species identified in the gas phase is a uniquely stable Zintl entity that could effect self-assembly, we report success in synthesizing and characterizing a lattice of analogous super-cluster assembled material. We demonstrate how the electronic properties of such assemblies can be fine tuned.

The last part of the talk will highlight our recent work on aluminum-hydrogen systems where we have identified $Al_4H_7^-$ as a potential cluster motif that is stable and exhibits the desired attributes needed for transforming from free clusters to clusters assemblies. The talk will also present a novel simple electronic structure model derived from first principal electronic structure studies that accounts for chemical features of metal-hydrogen assemblies and can predict global trends in the observed properties.

REFERENCES

[1] D.E. Bergeron, A.W. Castleman, Jr., T. Morisato, and S.N. Khanna, *Science* **304**, 84 (2004).
[2] D. E. Bergeron, P. J. Roach, A. W. Castleman, Jr., N. O. Jones, and S. N. Khanna, *Science* **307**, 231 (2005).
[3] J. U. Reveles, S. N. Khanna, P. J. Roach, and A. W. Castleman, Jr., *Proc. Nat. Acad. Sci.* **103**, 18405 (2006).

Th-II-6

NANOPARTICLES, NANOTUBES AND RELATED CHEMISTRY

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Improved routes to the synthesis of metallic nano-particles will be described together with their employment in the preparation of a wide range of carbon nanotubes. The preparation of exceptional long nano-wires which consist of linked C_{60} units with a width to length aspect ration as large as 3000 will be described. These have been found to possess a highly unusual morphology and composed of two nano-belts joined along the growth direction to give an L shaped cross-section. Possible models for their formation and growth will be discussed.

Th-II-7

ELECTRONIC STRUCTURE AND THERMODYNAMICS OF GOLD CLUSTERS

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Gold clusters have attracted considerable attention during the last years. This interest has been triggered by several experimental studies of the peculiar nature of nanoscale gold, for instance the discovery of gold based catalysts by Haruta, the pronounced size evolution of reactivity found by Heiz or the discovery of unusually large planar gold clusters by Kappes

.

This contribution will present recent advances in the quantum mechanical modelling of gold nanoclusters and their implications for the catalytic, dimensional and thermodynamical peculiarities of free gold clusters.

1) Density functional theory and density funcional based tight-binding calculations [1] have been used for the exploration of phase transitions in small gold cluster anions [2]. Evidence for a novel type of dynamical coexistence namely a liquid-liquid (2D/3D) phase coexistence has been found. The tendency to supercooling of metastable 3D isomers will be discussed.

2) The theoretical explanation of the observed reactivity of gold cluster anion towards adsorption of molecular oxygen provides a second example for the successful application of density functional theory to elucidate catalytic mechanisms [3]. Experimentally an odd-even oscillation in the O_2 take-up of Au_n^- clusters is observed (a pattern which can be easily explained by open spin shells). We found an explanation for anomalies in this odd-even pattern occurring e.g. for Au_{16}^- . The reactive cluster states belong to a partial jellium model of the gold 6s electrons. In this model the hexadecamer anion is close to a shell closing and therefore behaves halogen-like. Consequently, electron donation to the oxygen dimer is strongly reduced resulting in the inertness of the cluster.

REFERENCES

[1] P.Koskinen, H.Häkkinen, S.Sanna, T.Frauenheim, G.Seifert and M.Moseler, Density-functional based tight-binding study of small gold clusters, New J. Phys. **8**, 9 (2006)

[2] P.Koskinen, H.Häkkinen , B.Huber, B. v. Issendorff and M.Moseler, Liquid-Liquid Phase Coexistence in Gold Clusters: 2D or not 2D?, Phys. Rev. Lett. **98**, 015701 (2007)

[3] B.Yoon, P.Koskinen, B.Huber, O.Kostko, B. von Issendorff, H.Häkkinen, M.Moseler and U.Landman, Size-dependent Structural Evolution and Chemical Reactivity of Gold Clusters, Chem.Phys.Chem. **8,** 157 (2007)

Th-II-8

NEW DEFORMED SINGLE-PARTICLE SHELL MODEL

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The spheroidal harmonic oscillator had been used in various branches of Physics, e.g. the famous Nilsson model [1] very successful in Nuclear Physics. With a variant adapted to atomic clusters, Clemenger [2] could explain the spherical magic numbers $N = 2, 8, 20, 40, 58, 92$ which have been found in the mass spectra of sodium clusters.

Aiming at a future description of atomic clusters deposited on a planar surface, we present analytical relationships for the energy-levels of a new single-particle shell model whose equipotential surfaces are semi-spheroids with the symmetry axis z perpendicular to the plane of the circular base, containing the ρ axis:

$$\rho^2 = \begin{cases} (a/c)^2(c^2 - z^2) & z \geq 0 \\ 0 & z < 0 \end{cases}$$

For other kind of shapes (as in the liquid-drop calculations [3]) we shall consider in the future, it is not possible to get analytical formulae; the problem will be solved numerically.

The $Z(z)$ component of the wave function should vanish at $z = 0$ where the height of potential is infinitely high, hence only the negative parity states $(-1)^{n_z} = -1$ are allowed. In units of the shell gap $\hbar\omega_0$, the energy levels vs. the deformatin δ, defined by Clemenger, are given by

$$\epsilon = \frac{2}{(2-\delta)^{1/3}(2+\delta)^{2/3}} \left[n + \frac{3}{2} + \delta \left(n_\perp - \frac{n}{2} + \frac{1}{4} \right) \right]$$

where the integer quantum numbers n and $n_\perp = 0, 1, ..., n$ for $n = 1, 2, 3, ...$ have to give an odd number $n_z = n - n_\perp = 1, 3, ...$ Each level labeled by $n, n_\perp$ may accomodate $2n_\perp + 2$ particles.

The semi-spherical magic numbers are identical with those obtained at the oblate spheroidal superdeformed shape: 2, 6, 14, 26, 44, 68, 100, 140, ... The superdeformed prolate magic numbers of the semi-spheroidal shape are identical with those obtained at the spherical shape of the spheroidal harmonic oscillator: 2, 8, 20, 40, 70, 112, 168 ... This striking kind of symmetry will not hold for a Hamiltonian containing the term [2] proportional to $(\mathbf{l}^2 - \langle \mathbf{l}^2 \rangle_n)$, or a more complex equipotential surface.

References

[1] S.G. Nilsson, Dan. Mat. Fys. Medd. **29**, (1955)

[2] K.L. Clemenger, Phys. Rev. B **32**, 1359 (1985).

[3] V.V. Semenikhina, A.G. Lyalin, A.V. Solov'yov, W. Greiner, to be published.

Th-II-9

MOLECULAR ABSORPTION ON CARBON NANOTUBES

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The high specific surface area of Carbon Nanotubes, CNTs, makes them ideal candidates for applications and processes based on the adsorption of different molecules. We have investigated the basic mechanims for adsorption of several molecules on CNTs and the nature of bonding between the nanotube and the adsorbed molecules. This presentation will focus on two applications exhibiting different bonding properties of the adsorbed molecules: i) the selective removal of the metallic CNTs from the semiconducting ones through the chemisorption of nitronium ions and ii) the storage of Hydrogen for its use as the energy carrier in automotive applications. Hydrogen is weakly physisorbed to the nanotube.

i) After more than 15 years of intense research on CNTs since its discovery, most of the production techniques yield bundles of CNTs of mixed radii and chiralites. However, samples of CNTs of well defined characteristics, specific radius and either metallic or semiconducting character, are required for many applications and also for basic research. Therefore, the selective production and separation of CNTs become key issues for further scientific and technological developments based on CNTs. Recently, the selective removal of the metallic, from the semiconducting, SWCNTs in the presence of nitronium ions (NO_2^+) has been demonstrated [1]. To shed some light into this problem, we have performed Density Functional (DFT) calculations to investigate the adsorption mechanism of nitronium ions and neutral nitrogen dioxide (NO_2) molecules on both metallic and semiconducting nanotubes. The neutral NO_2 molecules bind to the tubes through weak Van der Waals forces which leave the tubes almost unaffected. However, the nitroniun ions bind to the tubes through strong charge transfer interactions (stronger for metallic tubes). Moreover, the structural stability of the metallic tubes (not the semiconducting ones) becomes compromised upon adsorption of nitronium ions. Our results help to understand the selective removal of the metallic tubes found in the experiments.

ii) The hydrogen storage problem is the biggest remaining research problem in the development of fuel cell vehicles powered by hydrogen. Due to their large specific surface area and low weight, CNTs have been proposed as candidates for storing hydrogen based on the adsorption of molecular hydrogen on the tube walls. However, the storage capacity of pure CNTs at room temperature and moderate pressures is much smaller (<1 wt%) than the required (6 wt%) for on board automotive applications. Using DFT we have investigated the enhancement on the storage capacity obtained by the Li-doping of the tubes. Other carbon nanoporous materials (such as Activated Carbons, ACs, and Carbides derived Carbons, CDCs) are being investigated for storaging hydrogen with promising results. We have developed a thermodynamical model for hydrogen storage on slitpores (two parallel graphene layers separated by a given interlayer distance) to model nanoporous materials. This model reproduces the experimental saturation on the gravimetric capacity with increasing external pressure and predicts that the nanopore size should be about 6 Å for obtaining the best storage capacities.

REFERENCES

[1] K.H. An, et al., J. Am. Chem. Soc. **127**, 5196 (2005)

Electron and photon cluster collisions

Fr-I-1

PHOTOFRAGMENTATION AND PHOTODETACHEMENT OF BIOMOLECULES AND CLUSTER-BIOMOLECULES COMPLEX

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In this paper, we present recent photo-dissociation experiments performed on biomolecules stored in a quadrupole ion trap. Information on the molecular structure can be inferred from the fragmentation channels as well as from the depletion absorption spectrum. We will show that the fragmentation channels recorded from photodissociation are specific and different from those recorded from Collision Induced Dissociation (CID). Some fragile bonds may be preserved in photodissociation processes: for example for phosphorylated peptides, the loss of phosphate group is dominant in CID and is restrained in photodissociation. In contrast side chain loss and formation of radicals are observed.

In the case of Tryptophan silver complex (WAg_n^+) the optical absorption spectrum are obtained from depletion and compared to ab initio theoretical calculations. Two classes of structure, zwitterionic and charge solvated ones can be identified. They are correlated not only to different absorption spectra, but also to two different fragmentation channels which may be therefore considered as fingerprint of the structure.

The photodetachement of peptides and DNA polyanions are investigated. For DNA Strands, the measured spectra as a function of the photon energy are base dependent. In both cases, the formation of radical anions and the mechanism of photodetachement are discussed. Moreover for peptides and protein polyanions, the photodetachement spectral dependence may be used as a signature of ionization state of tyrosine.

Fr-I-2

ELECTRON SCATTERING ON CLUSTERS AND BIOMOLECULES EMBEDDED IN HELIUM DROPLETS

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The extensive number of spectroscopy studies with doped helium droplets shows impressively the ability of superfluid helium droplets to be a perfect matrix for the preparation and study of cold targets and the formation of complex fragile species [1]. In contrast, inelastic electron interaction with doped helium droplets has been a much less studied subject.

This is the more surprising as clusters of biomolecules can be formed from the gas phase by embedding successively single biomolecules into a cold droplet. Moreover, it is known since recently that electrons can induce efficient DNA damage. This is important because in cells secondary electrons are produced with high abundance by ionizing radiation. Thus the underlying chemical and physical processes of the inelastic electron interaction with isolated and also solvated biomolecules is of relevance for the investigation of DNA damage by ionizing radiation and moreover, of fundamental interest in physical chemistry.

We have recently constructed a helium cluster source which was initially used to study in detail the properties of electron impact ionization of pure helium clusters [2], as well as metastable decays of helium cluster ions produced [3]. Recently we modified our setup by adding a pick up chamber including molecular beam ovens, a pick up cell and external gas inlets which allow the embedment of various molecules in cold superfluid helium droplets. The neutral mixed clusters thus produced are ionized in a Nier type electron impact ion source (with an energy range from about 0 to 150 eV) and ensuing cations and anions are mass analyzed by a high resolution two sector field mass spectrometer.

First studies of the pick up process have been performed with DNA nucleobases adenine and thymine both of which are well studied in the gas phase. Several interesting phenomena could be observed, e.g. in contrast to the gas phase situation electron attachment in this environment leads to the production of parent anions for adenine and thymine [4]. Moreover, site selectivity in the electron attachment process recently discovered in our laboratory for isolated nucleobases [5] is preserved in this complex environment and in addition a novel two step reaction scheme has been proposed to explain characteristic differences in the attachment spectra.

These pick-up experiments have been recently extended by embedding other systems into the helium (e.g., chloroform, valine and fullerenes) and in some cases even in the additional presence of water molecules.

In addition to these experiments with helium droplets we have recently analyzed the stability and fission dynamics of multiply charged neon cluster ions [6]. The critical sizes for the observation of long-lived ions (see the measured mass spectra in Fig.1) are n_2=284 and n_3=656 for charge states 2 and 3, respectively, a factor 3 to 4 below the predictions of a previously successful liquid-drop model developed by Echt and others. The preferred fragment ions of fission reactions are surprisingly small ($2 \lesssim n \lesssim 5$); their kinetic energy distributions peak at the very small energy of 200 meV or below. The size of these fission fragments and their average kinetic energies are much less than predicted by the previously accepted liquid-drop model.

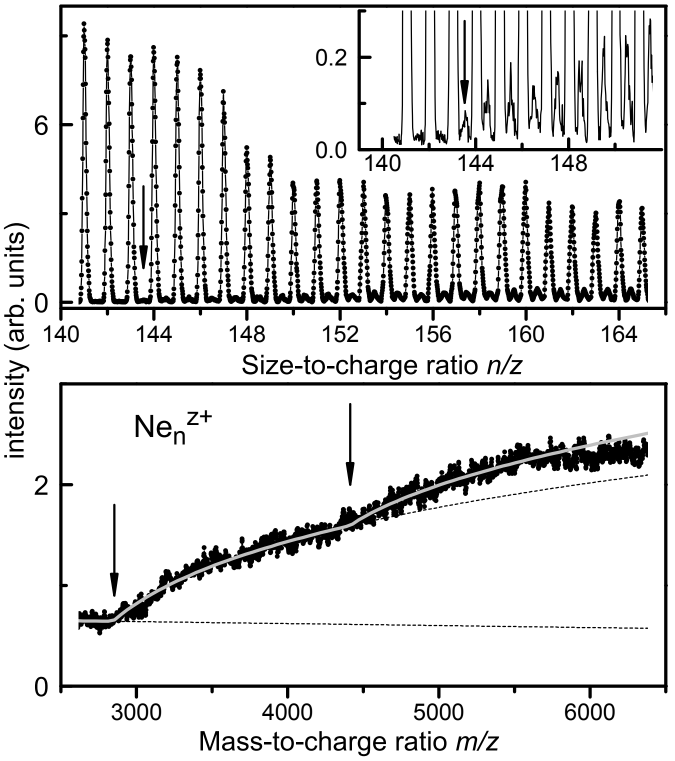

Figure 1: a) Mass spectrum of isotopically pure ^{20}Ne cluster ions. Arrows indicate the smallest observable doubly charged cluster, Ne_{287}^{2+}. b) Cluster ions formed from isotopically mixed neon. The stepwise intensity increases at $m/z \approx 2875$ and 4425 arise from doubly and triply charged cluster ions above their critical sizes $n_2 \approx 284$ and $n_3 \approx 656$, respectively.

This work was partially supported by FWF, Wien, the European Commission, Brussels and the Brazilian agency CNPq.

REFERENCES

[1] J. P. Toennies, A. F. Vilesov, *Angew. Chem. Int. Ed.* **43**, 2622 (2004)
[2] S. Denifl, M. Stano, A. Stamatovic, P. Scheier, T. D. Märk, *J. Chem. Phys.* **124,** 054320 (2006)
[3] S. Feil, K. Gluch, S. Denifl, F. Zappa, O. Echt, P. Scheier, T. D. Märk, *Int. J. Mass Spectr.* **252,** 166 (2006)
[4] S. Denifl, F. Zappa, I. Mähr, J. Lecointre, M. Probst, T. D. Märk, P. Scheier, *Phys. Rev. Letters* **97,** 043201 (2006)
[5] S. Ptasinska et al., *Angew. Chem. Int. Ed.* **44,** 6941 and *Phys. Rev. Letters* **95,** 093201 (2005)
[6] I. Mähr et al., *Phys. Rev. Letters* **98,** 023401 (2007)

Fr-I-3

ANGLE-RESOLVED PHOTOELECTRON SPECTROSCOPY OF COLD NA CLUSTER ANIONS

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Sodium clusters exhibit a strongly discretized density of states in good agreement with the predictions of the free electron model [1]. Nevertheless there is a significant electron lattice interaction, which perturbs the angular momentum eigenstates [2]. In order to study the character of the electron wavefunctions, angle resolved photoelectron spectroscopy has been performed on cold (~20 K) size-selected Na cluster anions in a wide size range using ns laser pulses with wavelengths from UV to IR.

The results clearly demonstrate a different character of different electron shells, and are in qualitative agreement with simple model calculations. Most importantly they indicate that even for the case of a cluster as large as Na_{147} the electron emission is a completely coherent process.

REFERENCES
[1] G. Wrigge, M. Astruc Hoffmann and B. v.Issendorff, Phys. Rev. A **65**, 063201 (2002)
[2] O. Kostko, B. Huber, M. Moseler, and B. v. Issendorff, Phys. Rev. Lett. **98**, 043401 (2007)

Fr-I-4

PLASMONS AND DYNAMICAL SCREENING OF ENDOHEDRAL ATOMS

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We discuss the manifestation of manyelectron excitations (plasmons) in various processes involving fullerenes and nanotubes.

The investigation is based on model within which a fullerene (or a nanotube) is treated as a spherical (a cylindrical) layer of the finite width. The two surfaces of the fullerene lead to the presence of two coupled surface plasmon eigenmodes. In the processes, which involve the interaction of a dipole photon with a fullerene, the two surface plasmon modes manifest themselves as coupled resonances in the cross sections. In this talk a particular emphasis is made on the role of plasmon excitations in the processes of photoionization of fullerenes and on the dynamical screening of an atom confined within a fullerene. We discuss the peculiarities in the cross sections of these processes and analyze their evolution with the size of a fullerene.

The processes, which involve the plasmon excitation of higher multipolarity, allow one to analyze the interplay between the surface plasmons and the volume plasmon modes. This interplay leads to additional peculiarities in the cross sections. We discuss these features in connection with the processes of elastic and inelastic electron scattering from fullerenes.

Fr-I-5

Photoabsorption and Photoionization of Fullerenes and Clusters

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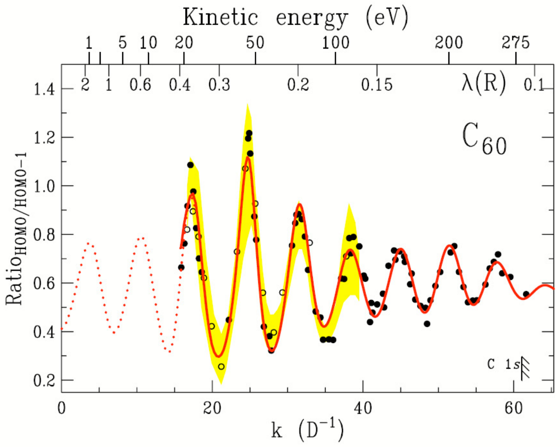
The electronic structure of fullerenes and a particular group of clusters is well described by delocalized electrons in a box-like potential, the so-called Jellium potential. This spherical potential gives rise to very specific photoionization behavior known as oscillatory behavior of the partial photoionization cross sections (see figure). These oscillations are basically due to the periodicity of the solutions of the Schrödinger equation or box-like potentials. One may interpret these solutions as standing waves in the cluster or fullerene. First evidence for this behavior was found in solid fullerene samples and later on in gas phase samples. Theoretical studies for regular clusters predicted similar behavior for a variety of clusters. First experimental effort to prove these predictions for different clusters is under way.



Concerning the fullerenes the oscillatory behavior has proven to be an analytical tool regarding the differences in the electronic structure of adsorbed and free fullerenes, in particular for C_{70}. One may consider this behavior on an even more fundamental level - the existence of strict or approximate point-like reflection symmetry. This symmetry, exhibited most pronounced by homonuclear diatomic molecules, gives rise to phase-coupled photoelectron emission from each point of two reflection-symmetric points. These so called molecular 'double slits' cause an oscillatory behavior with alternating photoelectron intensities for the gerade and ungerade combinations of the molecular states of all homonuclear diatomic moelecules. The same is true for the fullerenes and clusters with one major difference: here, the photoionization of the valence orbitals displays this behavior whereas in homonuclear diatomic molecules it is exhibited basically by the photoemission of core electrons. The reasons for this difference will be discussed, and the similarity in the photoionization behavior will be shown by comparison with the photoelectron emission behavior of 1*s* electrons of H_2 and N_2 in the molecule frame. In addition, it will also be shown that the formation of shape resonance-like features in the partial photoionization cross sections are based on the existence of standing waves at de Broglie wave lengths of $\lambda = 2$ and 4 times the bond lengths of the molecule or are diameter of the cluster. This would give rise to a resonance feature in C_{60} at approximately $E_{\mathrm{kin}} = 0.2$ eV, which has indeed been observed.

Fr-I-6

PHOTO-IONIZATION AND FRAGMENTATION OF FULLERENE IONS

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Mass selected fullerene ions have been exposed to synchrotron radiation at energies ranging from 17 eV to about 300 eV. Absolute cross sections for single and multiple ionization as well as fragmentation were measured as a function of photon energy for ions of C_{60}, C_{70}, C_{80}, C_{82}, and C_{84}. More recently, the first ever experiments with endohedral fullerene ions were conducted with $Sc_3N@C_{80}^+$ and $Ce@C_{82}^+$.

In the single ionization of C_{60}^+ ions [1] the well known surface plasmon resonance near 20 eV was observed. In addition, a higher order resonance was found near 40 eV. The individual contributions of the two resonance features are indicated in Fig. 1 by Lorentz profiles fitted to the experimental data on top of a smooth background. Similar plasmon contributions at almost identical energies were found in single ionization of all other fullerene ions as well.

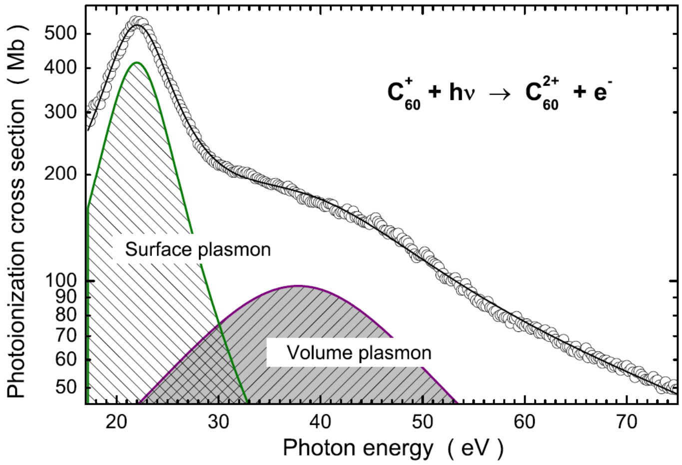

Figure 1: Absolute cross section for single photoionization of C_{60}^+ ions. The thin solid line results from a fit to the measured data of a linear background (not shown) plus two separately displayed Lorentzian curves which represent the plasmon resonances.

Calculations based on time-dependent density functional theory were performed for the photoabsorption of C_{60}^+ ions confirming the collective nature of the broad resonance features. In particular, the theoretical results support the conclusion that the second resonance at 40 eV is due to a dipole-excited volume plasmon made possible by the fullerenes' hollow-sphere geometry [1].

References

[1] S. W. J. Scully, E. D. Emmons, M. F. Gharaibeh, R. A. Phaneuf, A. L. D. Kilcoyne, A. S. Schlachter, S. Schippers, A. Müller, H. S. Chakraborty, M. E. Madjet and J. M. Rost, Phys. Rev. Lett. **94**, 065503 (2005).

Fr-I-6

Ion-cluster collisions

Fr-II-1

COLLISION OF TRANSITION METAL CLUSTER IONS WITH SIMPLE MOLECULES

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The reactivity of iron clusters with hydrogen has at first been related to the electronic structures [1]. In this report, we showed that the dehydrogenation of an ethylene molecule on an iron-group metal cluster (M_n^+ (M = Fe, Co, Ni; $n \leq 30$)) is closely related to the densities of 3d- and 4s-electrons in the valence electronic state through measuring their reaction cross sections.

Mass spectra of the product ions showed that dehydrogenation of an ethylene molecule proceeded exclusively on a metal cluster ion with leaving $M_n^+(C_2H_2)$ (M = Fe, Co, Ni). As the cluster size, n, increases, the dehydrogenation cross section increases rapidly at a threshold size, and reaches as high as ≈50 Å^2, which is close to the cross section estimated from the electrostatic interaction between the cluster ion and an ethylene molecule. The cross section on Fe_n^+ increases at $n \approx 20$, while those on Co_n^+ and Ni_n^+ increase at $n \approx 10$.

Electronic structures of Fe_n^+ (n = 13, 19, 25), Co_{13}^+ and Ni_{13}^+ were calculated. The density-of-states profiles calculated showed that in Co_{13}^+ and Ni_{13}^+ a weak 4s peak is present next to a strong and wide 3d peak, while in Fe_{13}^+ a sharp 4s peak is well separated from a 3d peak. As the cluster size increases in Fe_n^+, the 3d peak is broadened while the 4s peak approaches to the 3d peak. This propensity of the electronic structure agrees with the reported photoelectron spectra of Fe_n^-, Co_n^- and Ni_n^- [2]. These findings indicate that 3d electrons of the iron-group metal clusters play a central role in the ethylene dehydrogenation on the iron-group metal clusters.

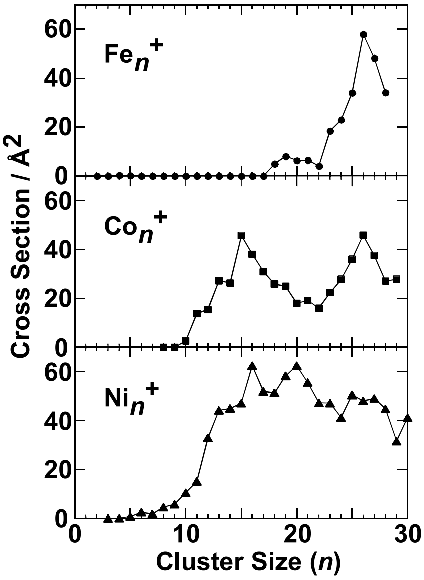

Figure 1: Cross sections for ethylene dehydrogenation on Fe_n^+, Co_n^+ and Ni_n^+. The collision energy was 0.4 eV.

REFERENCES
[1] R. L. Whetten et al., *Phys. Rev. Lett.* **54**, 1494 (1985)
[2] L.-S. Wang et al., *Chem. Phys.* **262**, 53 (2000); S.-R. Liu et al., *Phys. Rev.* B **65**, 113401 (2002)

Fr-II-2

FRAGMENTATION DYNAMICS OF CHARGED FINITE-SIZE SYSTEMS

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In the present contribution we will discuss different processes linked to the instability and fragmentation of small charged systems, in particular of small molecules of biological interest up to large systems like micro-droplets.

Radiation damage of biological tissues has been studied for a long time in order to clarify the mechanisms and the phenomena occurring during the interaction of high-energy radiation with living cells[1]. These studies clearly showed that the most severe consequences of irradiation are due to the damage of DNA, more precisely to the production of single and double strand breaks and in particular to the clustering of these lesions[2]. More recently, studies of low-energy collisions involving isolated molecules of biological interest are performed in the gas phase in order to understand the phenomena on a molecular level. However, the question is raised, to which extend the obtained results can be applied to biomolecular systems in their natural environment, i.e. in an aqueous solution or a chemical environment.

In contrast to the biomolecular case, where the molecular structure plays an important role in the selection of the fragmentation modes, neutral liquid micro-droplets are highly symmetric with a perfectly spherical shape. Thus, many of their basic properties can be described with the well-known liquid-drop model.

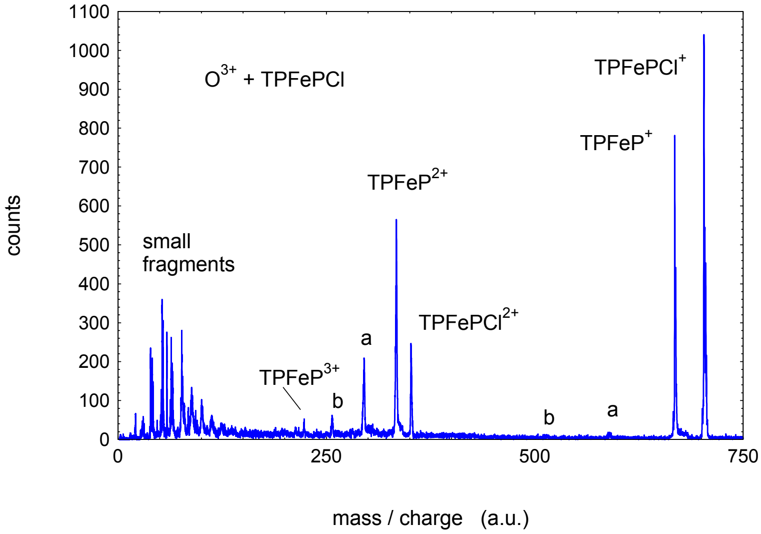


Figure 1: Typical mass spectrum of intact TPFePCl ions and fragments produced in collisions of triply charged oxygen ions with TPFePCl.

Fr-II-2

As an example for an isolated molecule in the gas phase, we will describe the fragmentation pattern obtained in collisions of highly charged ions with a Porphyrin molecule (Tetraphenyl Iron (III) Porphyrin Chloride) (see figure 1). Different processes like the loss of phenyl groups, the emission of the neutral Cl atom or the Cl^- anion occurring as direct processes as well as delayed on a microsecond time scale are observed. Furthermore, strong differences in the charge state distribution of atomic fragments will be reported (see figure 2).

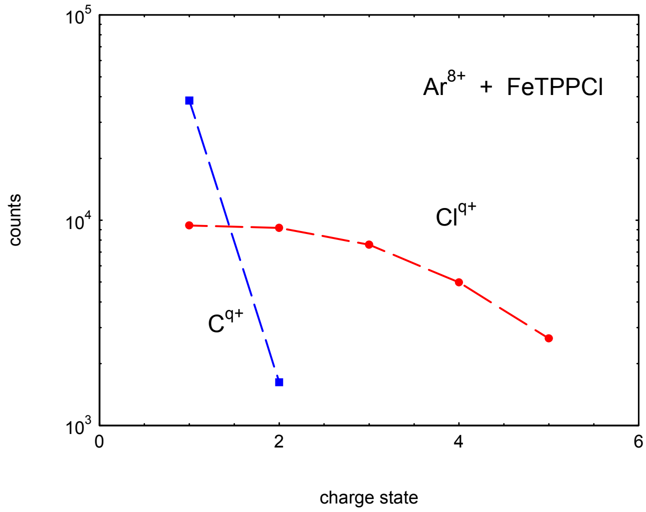
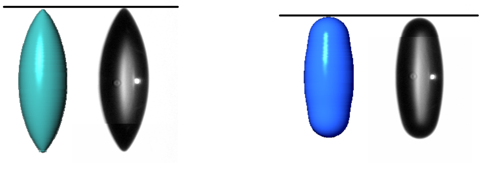


Figure 2: Multi-ionised atomic fragments, produced in collisions of Ar^{8+} ions (80 keV) with TPFePCl.

As a second example, we will discuss the possible protection of biomolecular systems by a surrounding water shell. Thus, it is found, that with increasing number of attached water molecules the fragmentation yield due to collision induced dissociation is reduced as the transferred energy is efficiently dissipated by the evaporation of the water molecules[3]. Different phenomena are observed after electron capture collisions.

Finally, we will discuss the shape deformation of liquid droplets of micrometer size when the systems arrive at the limit of stability. New theoretical results will be compared with experimental data obtained by using levitated droplets [4,5] observed with a fast photo-imaging technique.



before after

Figure 3: Droplet forms before and after Coulomb explosion. The colored forms (left ones) are calculated.

REFERENCES

[1] C. von Sonntag, *Free-Radical-Induced DNA damage and its Repair* (Springer 2006)
[2] K.M. Prise, C.H.L. Pullar, and B.D. Michael, Carcinogenesis **20**, 905 (1999)
[3] B. Liu *et al.,* Phys. Rev. Lett. 97, 133401 (2006)
[4] D. Duft *et al.*, Phys. Rev. Lett. 89, 084503 (2002)
[5] D. Duft *et al.*, Nature 421, 128 (2003)

Fr-II-3

FRAGMENTATION OF MULTIPLY IONIZED FULLERENE DIMERS

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The ionization and fragmentation of isolated fullerene monomers (C_{60} and C_{70}) have been the subject of a vast number of experimental and theoretical studies during the past decades [1]. In contrast there have only been a few studies on the smallest fullerene aggregates (dimers) of van der Waals type, all devoted to neutral and singly charged species.

Here, we have used slow highly charged ions (Xe^{30+}) to multiply ionize fullerene dimers in slow $Xe^{30+} + [C_{60}]_2$ ($[C_{60}C_{70}]$) $\rightarrow \ldots + [C_{60}]_2^{r+}$ ($[C_{60}C_{70}]^{r+}$) + (r-s)e^- electron-transfer collisions at 600 keV ($r \leq 7$) [2,3]. With the aid of multi-coincidence techniques we have registered the intact ions and ions stemming from fragmentation. This allowed us to deduce the relative ionization cross sections as functions of the final charges states of the dimers and the kinetic energy releases for the fragmentation processes. The relative ionization cross sections display even odd variations that are markedly different compared to the typical smooth decreasing behaviour of the cross sections for multiple ionization of fullerene monomers. The kinetic energy releases for the dominant fragmentation processes that yield intact fullerenes are found to be low, indicating that internal excitations of the seperating fullerenes are important. The experimental results are interpreted within the framework of a simple electrostatic model for dimer ionization and fragmentation [2,3].

REFERENCES
[1] E. E. B. Campbell and F. Rohmund, Rep. Prog. Phys. 63, 1061 (2000); E. E. B. Campbell, *Fullerene Collision Reactions* (Kluwer Academic, London, 2003).
[2] H. Zettergren, H. T. Schmidt, P. Reinhed, H. Cederquist, J. Jensen, P. Hvelplund, S. Tomita, B. Manil, J. Rangama, and B. A. Huber, Phys. Rev A., accepted (2007).
[3] H. Zettergren, H. T. Schmidt, P. Reinhed, H. Cederquist, J. Jensen, P. Hvelplund, S. Tomita, B. Manil, J. Rangama, and B. A. Huber, J. Chem. Phys., submitted (2007).

Fr-II-4

STRUCTURE AND FRAGMENTATION OF FULLERENES AND FULLERENE DIMERS PRODUCED IN COLLISIONS WITH IONS

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In order to analyze in detail the fragmentation patterns observed in collisions of fullerenes with ions it is essential to have accurate information about the stability of different isomers and different quantities as dissociation energies, ionization potentials or fission barriers.

In this communication we present the theoretical results obtained using density functional theory (DFT) for a very a large series of fullerenes. Neutral, singly and doubly charged fullerenes from C_{20} up to C_{72} have been studied at exactly the same level of theory (fully optimized geometries at B3LYP/6-31G* level) [1-3] and several isomers have been considered for each fullerene size and charge. Furthermore in the case of C_{58}, C_{60}, C_{68} and C_{70} we have extended the calculations to include highly charged fullerenes (charge up to 14) [4-5]. The analysis of these data has allowed us to better understand the different factors governing the stability of fullerenes, to determine the relative energy of the different possible dissociation channels and to compare our results with the available experimental data. These calculations have been recently extended to the study C_{60} dimers linked by van der Waals forces.

Once the structure, electronic energy and harmonic vibrational levels of the different fragments are well established it is possible to perform the analysis of the fragmentation process by using Microcanonical Metropolis Montecarlo (MMMC) and the Weiskopff theory; both methodologies have been previously applied to predict the fragmentation patterns of small carbon clusters [6,7]

REFERENCES

[1] S. Díaz-Tendero, M. Alcamí and F. Martín J. Chem. Phys. 119, 5545 (2003)
[2] G. Sánchez, S. Díaz-Tendero, M. Alcamí and F. Martín Chem. Int. J. Mass Spectrom. 252, 133 (2006)
[3] M. Alcamí, G. Sánchez, S. Díaz-Tendero, Y. Wang and F. Martín. J. Nanosci. Nanotech. 7, 1329 (2007)
[4] S. Díaz-Tendero, M. Alcamí and F. Martín. Phys. Rev. Lett. 95, 013401 (2005)
[5] S. Díaz-Tendero, M. Alcamí and F. Martín. J. Chem. Phys. 123, 184306 1-12 (2005)
[6] G. Martinet et al. Phys.Rev. Lett 93 063401 (2004)
[7] S. Díaz-Tendero, P.-A. Hervieux, M. Alcamí and F. Martín. Phys. Rev. A 033202 (2005).

Fr-II-5

THE IMPACT OF NUCLEAR INTERACTIONS ON ION BEAM TUMOR THERAPY

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Clusters on surface

Sa-I-1

FINE CONTROLLED DEPOSITION OF FUNCTIONAL BINARY CLUSTERS ONTO A DESIGNED SURFACE

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The adsorption state and thermal stability of organometallic sandwich clusters of vanadium (V) – benzene (Bz), $V(Bz)_2$ and $V_2(Bz)_3$, soft-landed onto a self-assembled monolayer of different chain-length n-alkanethiols (Cn-SAM, n = 8, 12, 16, 18, and 22) were studied by means of infrared reflection absorption spectroscopy (IRAS) and temperature programmed desorption (TPD). The IRAS measurement confirmed that $V(Bz)_2$ clusters are molecularly adsorbed and maintain a sandwich structure on all of the SAM substrates. In addition, the clusters supported on the SAM substrates are oriented with their molecular axes tilted 70–80° off the surface normal. An Arrhenius analysis of the TPD spectra reveals that the activation energy for the desorption of the supported clusters increases linearly with the chain-length of the SAMs. For the longest chain C22-SAM, the activation energy reaches ~150 kJ/mol, and the thermal desorption of the supported clusters can be considerably suppressed near room temperature. The clear chain-length-dependent thermal stability of the supported clusters observed here can be explained well in terms of the cluster penetration into the SAM matrixes. Besides, the vibrational assignment of the IRAS spectrum for the $V_2(Bz)_3$ sandwich was based on a harmonic frequency analysis that employed density functional theory. The close similarity between the experimental and calculated results for the IR absorption frequencies demonstrates that the $V_2(Bz)_3$ complexes have a multidecker sandwich structure on the SAM substrate. Our thermal desorption study established that the sandwich complexes can be isolated on the SAM substrate up to a high temperature of ~350 K.

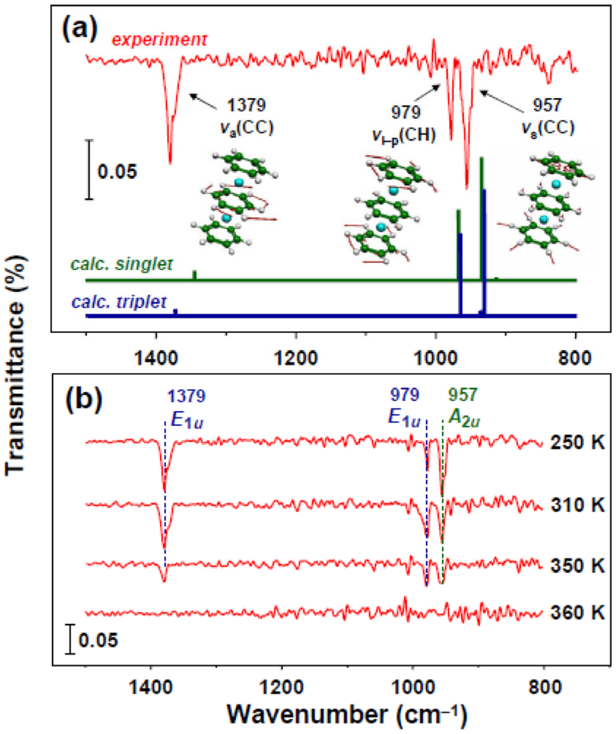

Figure 1: (a) IRAS spectrum of the $V_2(benzene)_3$ complex on C18-SAM, with calculated IR absorption spectra for singlet and triplet states. (b) IRAS spectra showing the temperature-dependent variation of $V_2(benzene)_3$ complex on C18-SAM.

REFERENCES

[1] S.Nagaoka,T.Matsumoto,K.Ikemoto,M.Mitsui,A.Nakajima, *J. Am. Chem. Soc.* **129**, 1528 (2007).

[2] S. Nagaoka, T. Matsumoto, E. Okada, M. Mitsui, A. Nakajima, *J. Phys. Chem. B* **110**, 16008 (2006).

[3] M. Mitsui, S. Nagaoka, T. Matsumoto, A. Nakajima, *J. Phys. Chem. B (Letter)* **110**, 2968 (2006).

Sa-I-2

INTERACTION OF NANOCLUSTERS WITH VARIOUS SURFACES

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The fundamental studies of the interaction of nanoclusters with various surfaces, including metal, ceramic and polymer will be presented. The nanoclusters that are introduced from vapour phase, and their interaction with surfaces, leading to the nucleation and growth of nanostructured films will be described. The effects of physical and chemical properties of nanoclusters on various deposited surfaces will be discussed. The surface energy and properties of the films and their hydrophobic or hydrophilic behaviour towards water molecules will be presented. The potential applications of fundamental studies of the interaction of nanoclusters with various surfaces in the field of engineering and biomedical will be highlighted.

Sa-I-3

SIZE SELECTED CLUSTERS SOFT LANDED ON A SURFACE: MORPHOLOGY AND ORGANIZATION

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The low energy cluster beam deposition technique is an interesting route for synthesizing nanostructured thin films with specific properties, especially when the size of the particles is sharply selected in the nanometer range. For that purpose, we have developed an experimental set-up for depositing mass selected clusters produced in a laser vaporization source [1]. Size selection has first proven to be a tool for understanding some fundamental aspects of cluster physics that will be illustrated here through two examples.

The first one concerns the precise correlation between the size and the morphology of individual particles that are soft-landed onto a surface and further observed by transmission electron microscopy. In the case of platinum clusters, a transition from spherical to strongly ramified structures is detected when the size increases beyond a critical value (Figure1). This shows that the kinetics of growth essentially depends on particle-particle binding collisions and that the coalescence of paired particles must be quenched or at least strongly slowed down above a transition size. The final cluster structure will be ruled by the interplay between coagulation and shape relaxation processes that both take place during the stage of formation, in the cluster source [2]. Such a ramification of nanoparticles *in the gas phase* will be discussed in the light of simple models previously developed to describe the bi-dimensional growth of fractal cluster islands *on surfaces*.

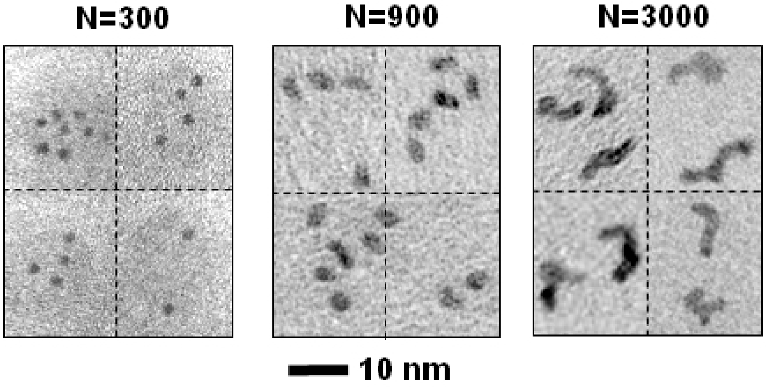

Figure 1: Transmission electron micrographs of size selected platinum clusters deposited on amorphous carbon (N is the number of atoms)

In a second example, we will show that the organization of deposited clusters in large assemblies is reminiscent of their initial size selection. This will be examined through two extreme situations represented by indium and platinum clusters. Because of very different diffusion and coalescence behaviors when they are deposited on a substrate, the former easily coagulate to form large spherical entities with a hierarchy of discretely scaled sizes (Figure 2) when the later do not touch one each other and rather form regular arrays with a sharply defined inter-particle distance. At that time, there is no definitive explanation for this short range effective repulsion between platinum nanoparticles.

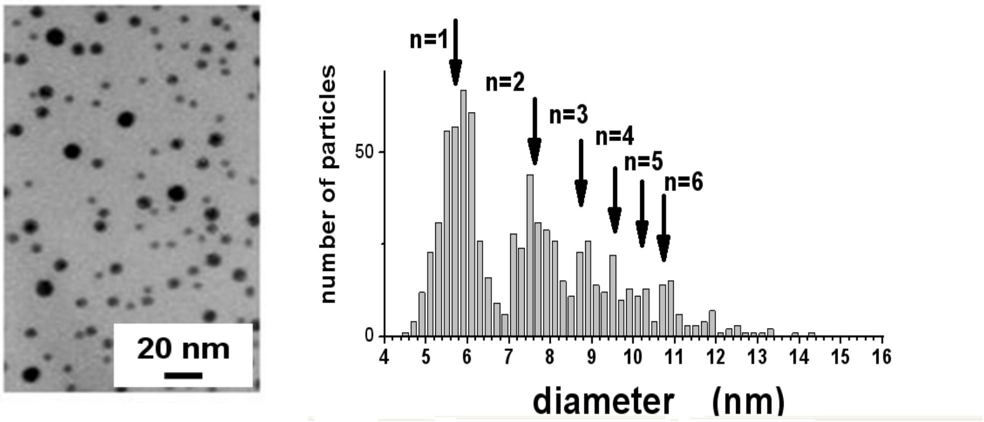

Figure 2: Transmission electron micrograph of size selected indium clusters (5.8 nm in diameter) deposited on amorphous carbon and corresponding size histogram (n is the number of coalesced primary clusters)

REFERENCES

[1] R. Alayan, L; Arnaud, A. Bourgey, M. Broyer, E. Cottancin, G. Guiraud, J.R. Huntzinger, J. Lermé, J. L. Vialle and M. Pellarin, Rev. Sci. Instrum. **75**, 2461 (2004)

[2] R. Alayan, L.Arnaud, M. Broyer, E. Cottancin, J. Lermé, J.L. Vialle, and M. Pellarin, Phys. Rev. B. **73**, 125444 (2006)

Sa-I-4

Simulation of the nanoindentation procedure on Nickel on the smallest length scale: a simple atomistic level model

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A new field of interest which emerged recently from the development of Micro–Electro–Mechanical Systems (MEMS) is the use of (coated) metallic materials for chirurgical applications because of their potential bio-compatibility and interesting mechanical and wear properties.
Nanoindentation is a testing procedure developed in order to characterize the mechanical behavior of materials at nanoscales. The principle of the nanoindentation experiment [1] is similar to the "classical" hardness measurement of materials: a diamond indenter with a defined geometry is pushed into the sample material deforming a small volume of the sample. The output of the experiment is the continuously sensed applied load–indenter displacement curve (Fig 1) which is is considered to be a mechanical fingerprint of a material's response to the deformation. The concept is thus quite simple, but the encountered physics in this measurement procedure are very complex, which require the use of numerical models for their understanding.

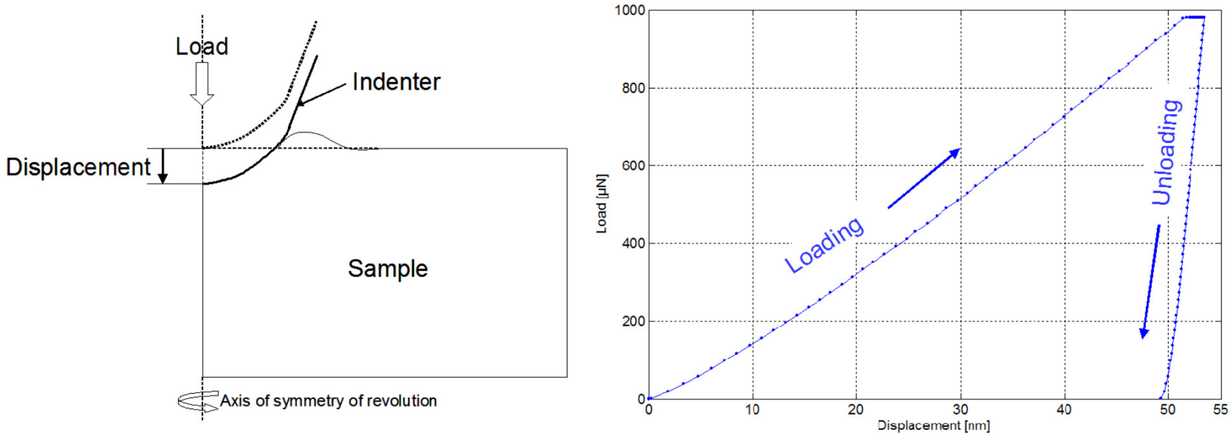

Figure 1: Left: Schema of the nanoindentation with conical indenter; Right: Load–displacement curve for pure Nickel issued from finite element simulation

An atomic scale model has been set up as an alternative to the frequently used continuum descriptions for the problem of characterization of pure Nickel in very shallow indentations depths (used for thin films). In order to circumvent the computational difficulties related to the choice of a discrete atomic level model some simplifications have been considered so that the response can be efficiently calculated numerically even for quite large system sizes. The nanoindentation procedure is modeled as a quasi–static phenomenon reducing the numerical problem to a structural minimization of the total energy of the Nickel cubic close-packed atomic structure in successive increments. The applied interaction potentials are of Lennard–Jones type, they are an approximation of Ni–Ni and Ni–C interaction potentials issued from DFT calculations [2]. A perfect lattice structure of parallelepipedic shape is considered both for the Nickel sample and for the diamond indenter. A program has been written to drive this displacement controlled calculation. The results obtained by this numerical simulation are the positions of the atoms in the deformed configuration and the total energy of the structure (Fig. 2). The total energy–displacement curve issued from this simple atomic level model is consistent with other simulations in the domain [3] and yields physically sound trends. An atomic scale load–displacement curve can also be computed from the above results.

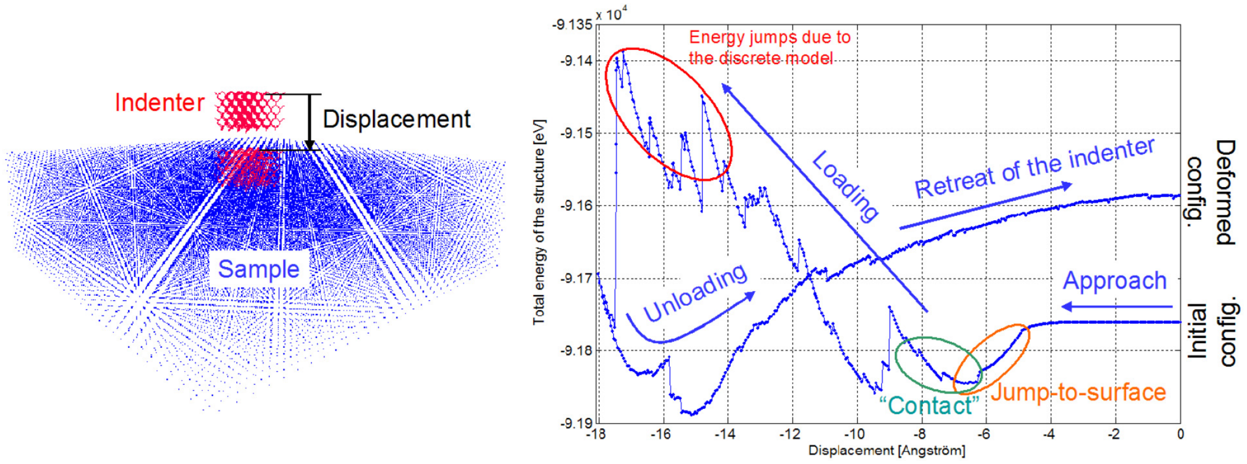

Figure 2: Left: The numerical model; Right: The total energy–displacement curve obtained from the atomic level model

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References

[1] S.J. Bull, J. Phys. D: Appl. Phys. **38**, R393 (2005)

[2] Y. Shibuta1, S. Maruyama, Comp. Mat. Sci., in press (2006)

[3] I. Szlufarska, Materials Today **9**, 42 (2006)

Phase transitions, fusion, fission and fragmentation in finite systems

Sa-II-1

NANOFRACTALS AND SELFASSEMBLING

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Sa-II-2

FLYING CRYOSTATS: BOSONIC HELIUM DROPLETS WITH MOLECULAR DOPANTS. SELECTIVE LOCALIZATION AND QUANTUM EFFECTS

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Our knowledge and understanding of He droplets has witnessed a tremendous growth in recent years since, from the standpoint of picking up possible impurities the clusters containing ^{4}He atoms have shown themselves to be able to do so with any species with which they collide and the attached "dopant" can in turn reside either in the inside of the droplet or at its surface, depending on the strength of the He-dopant interaction with respect to the one among solvent atoms [1].

We have been using variational Monte Carlo and Diffusion Monte Carlo calculations involving atomic and molecular dopant species (e.g. Li^+, K^+, Li_2^+, K_2^+, LiH, LiH^+, etc.) in order to establish as accurately as possible the localization effects induced by the stronger ionic interaction with its immediate surrounding of solvent atoms (snowball features) and the further accumulation of the He atoms in such inner, microsolvation shells (electrostriction). The interaction forces have been obtained from ab initio methods and the spatially selective allocation of solvent atoms is clearly shown within the quantum picture from the calculations [2,3,4].

A pictorially illuminating description of the surrounding wavefunctions and solvent densities also helps to better understand the strongly orientational nature of the ionic dopants and the spatial selectivity of the microsolvation process [5].

REFERENCES

[1]e.g. see: J.P. Toennies and A.F. Vilesov, Angew. Chem. Int. Ed. 43, 2622 (2004).
[2] C. Di Paola and F.A. Gianturco, Eur. Phys. J. D 35, 513 (2005).
[3] E. Bodo, E. Yurtsever, M. Yurtsever and F.A. Gianturco, J. Chem. Phys. 124, 074320 (2006).
[4] C. Coccia et al., J. Chem. Phys. (2007) in press.
[5] F. Marinetti, E. Bodo, F.A. Gianturco, Chem. Phys. Chem. 7, 1 (2006).

MELTING OF SMALL PARTICLES AND CLUSTERS: THERMODYNAMICS ON A FINITE SCALE

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One of the best known phase transition is the melting of a solid or the freezing of a liquid. It has turned out to be very difficult to construct a consistent picture of this prototypical first order phase transition for bulk systems, as the transitions occur in dense, strongly interacting systems. They are collective phenomena of a many particle system, which might be easier to treat for finite systems.

It has become possible in the last ten years to measure[1-3] and calculate[4] melting and freezing for mass selected metal clusters in vacuum. The measured or calculated caloric curves (i.e. the internal energy as a function of temperature) give the three relevant parameters describing this finite size analogue of a fist order phase transition: the melting temperature T_{melt}, and the changes upon melting in entropy ΔS and internal energy ΔE. These are related by $\mathrm{T}_{melt} = \Delta E/\Delta S$. All three values decrease for small particles, in general. Most experimental results are available for sodium clusters and these will be discussed in detail.

Surprisingly, the maxima in the size dependence of T_{melt} cannot be interpreted as electronic magic numbers, while nearly all other properties of Na clusters do allow such an interpretation. Instead, the maxima in the size dependence of ΔE and ΔS (but not of T_{melt}) are due to geometrical shell closings.

The similarities and differences between a canonical and a microcanonical description near a phase transition will be treated, as well as the ensuing negative heat capacity. Finally, recent measurements on oxidised clusters and problems to be attacked in the future will be discussed.

[1] Ch.Ellert et al. *Phys. Rev. Lett.* **75** 1731 (1995)

[2] M.Schmidt et al. *Nature* **393** 238 (1998)

[3] H.Haberland et al. *Phys. Rev. Lett.* **94** 035701 (2005)

[4] A.Aguado, J.M.López, *Phys. Rev. Lett.***94** 233401 (2005)

Sa-II-4

CONFORMATIONAL CHANGES IN POLYPEPTIDES / PHASE TRANSITIONS

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The phase transitions in finite complex molecular systems, i.e. the transition from a stable 3D molecular structure to a random coil state or vice versa (also known as (un)folding process) occur or can be expected in many different complex molecular systems and in nano objects, such as polypeptides, proteins, polymers, DNA, fullerenes, nanotubes.

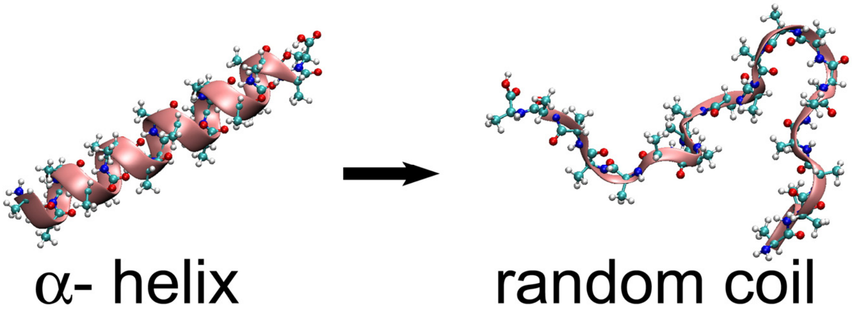

Fig. 1 The characteristic structural change of alanine polypeptide experiencing an α-helix↔random coil phase transition.

We suggest a novel *ab initio* theoretical method [1] for the description of phase transitions in the mentioned molecular systems. In particular, it was demonstrated that in polypeptides (chains of amino acids) one can identify specific, so-called twisting degrees of freedom, responsible for the folding dynamics of the amino acid chain, i.e. for the transition from a random coil state of the chain to its α-helix structure (see Fig. 1). The essential domain of the potential energy surface of polypeptides with respect to these twisting degrees of freedom can be calculated and thoroughly analysed on the basis of *ab initio* methods such as density functional theory (DFT) or Hartree-Fock method. It is shown [1] that this knowledge is sufficient for the construction of the partition function of a polypeptide chain and thus for the development of its complete thermodynamic description, which includes calculation of all essential thermodynamic variables and characteristics, e.g. heat capacity, phase transition temperature, free energy etc. The method has been proved to be applicable for the description of the phase transition in polyalanine of different length by the comparison of the theory predictions with the results of several independent experiments and with the results of molecular dynamics simulations.

REFERENCES

[1] A. Yakubovich, I. Solov'yov, A. Solov'yov, and W. Greiner, Eur. Phys. J. D (Highlight paper), **40**, 363 (2006); Europhys. News, **38**, 10 (2007).

Sa-II-5

EXPERIMENTS ON ATOMIC CLUSTERS: BOILING AND SUBLIMATION

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Unimolecular decays of moderately exited atomic clusters have been studied since the very early days of cluster physics. Less attention has been paid to the close link between those non-equilibrium decays of free clusters in the gas phase and the concepts of boiling or sublimation in equilibrium thermodynamics [1]. Using this link, microcanonical caloric curves across the liquid to gas phase transition can be constructed based on measured caloric curves for solid and liquid clusters [2], on ideal gas theory and on the enthalpies of vaporization that are determined from fragmentation pattern from well defined excitations of mass selected clusters.

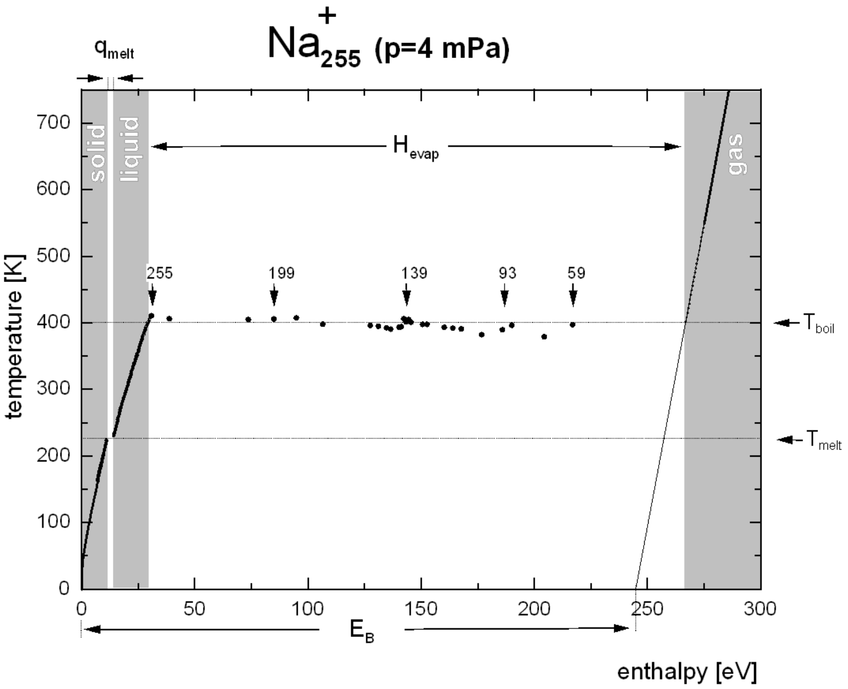

Figure 1: The microcanonical caloric curve of a cluster of 255 sodium atoms including the solid-liquid and the liquid-gas phase transitions. The numbers in the liquid-gas coexistence range indicate specifically stable clusters that appear as magic numbers in the fragmentation pattern.

REFERENCES

[1] M. Schmidt, T. Hippler, J. Donges, W. Kronmüller, B. v. Issendorff, P. Labastie and H. Haberland, Phys. Rev. Lett. **87**, 203402 (2001).

[2] M. Schmidt, R. Kusche, W. Kronmüller, B. v. Issendorff, and H. Haberland, Phys. Rev. Lett. **79**, 99 (1997); M. Schmidt, R. Kusche, B. v. Issendorff, and H. Haberland, Nature **393**, 238 (1998).

Sa-II-6

MULTISCALE APPROACH TO CLUSTER FRAGMENTATION

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A general theoretical framework for describing the thermally induced sequential decay in atomic clusters is presented. Our scheme relies on a full treatment of individual dissociation steps based on phase space theory (PST), built into a kinetic Monte Carlo (kMC) procedure [1]. This combined PST/kMC approach allows us to follow the evolution of several statistical properties such as the size, the temperature or the angular momentum of the cluster over arbitrarily long time scales. Quantitative accuracy is achieved by incorporating anharmonicities of the vibrational densities of states, the rigorous conservation of angular momentum, and a proper calibration of the dissociation rates. After validating the method on selected Lennard-Jones clusters thermally excited, we extend it to describe the decay of argon clusters initially ionized and excited on high electronic states, that we describe with a diatomic-in-molecules quantum Hamiltonian. The multiscale protocol consists of explicit nonadiabatic molecular dynamics with quantum transitions, followed by dynamics on the ground state, and further processed at a statistical level by applying the kinetic Monte Carlo scheme.

REFERENCES

[1] F. Calvo and P. Parneix, J. Chem. Phys. 126, 034309 (2006)

Sa-II-7

ATOM BY ATOM NUCLEATION OF SODIUM AND CALORIC CURVES

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Thermodynamics of small clusters has received a renewed interest since the pioneering work of Haberland *et al*[1] who experimentally demonstrated that very small clusters could undergo a phase transition, by measuring caloric curves. Doing calorimetry (in any system) requires being able to measure the variation of internal energy ΔE corresponding to a variation of temperature ΔT. One possibility is to control the temperature by a heat bath and to observe a physical observable, say S, in one to one correspondence with the internal energy $E^*=f(S)$. In free clusters, the energy is brought either by a laser [1] or by collisions [2]. In the first case, S is characterized by the number of evaporated atoms, in the second by 50% evaporation of the incoming cluster A third method measures the mobility of clusters in a drift tube; the phase transition from liquid to solid is identified thanks to a variation in the collision cross section [3].

We propose here a novel method based on an original experimental setup [4] to measure the caloric curve of clusters. The basic idea is to bring energy to the cluster by sticking atoms. After each sticking, E^* is increased by a known energy. After a given number of sticking collisions $n_{\max}$ the cluster is so heated that it dissociates before it can stick again. $n_{\max}$ is related to the dissociation time, thus is a measure of E^*. As in the first two experiments mentioned above, we can construct the caloric curve by varying T. Differential measurements at two different collision energies makes our results independent on the exact relation $E^*=f(S)$. The method does not require laser excitation, is easily transferable to many systems and it is model free.

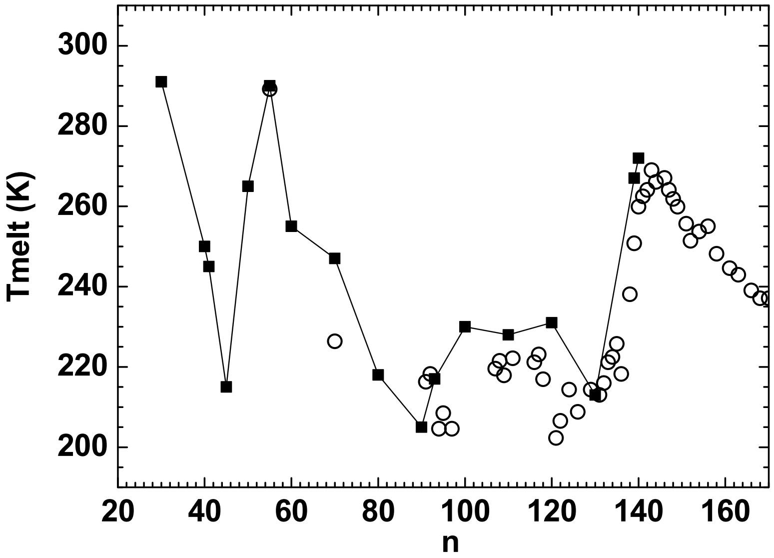

Figure 1: Melting temperature of sodium clusters. Comparison of our results (squares) with the one of Haberland *et al* (circles) [1].

Sa-II-7

REFERENCES
[1] H. Haberland *et al*, *Phys. Rev. Lett.* **94**, 035701 (2005).
[2] G.A. Breaux *et al, Phys. Rev. Lett.* **91**, 215508 (2003).
[3] G.A. Breaux *et al, J. Phys. Chem.* B, **109**, 16575 (2005).
[4] F. Chirot *et al*, *Rev. Sci. Instrum.* **77**, 063108 (2006).

Clusters in laser fields

Sa-III-1

GROUND STATE ION-PAIR FORMATION AND EXCITED STATE BIRADICAL FORMATION IN HI(H_2O)$_n$ CLUSTERS

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Elucidating the process of ground state ion-pair formation in simple acids has been of long standing interest to the community[1, 2] and in our laboratory[3-5]. Ion-pair formation has broad significance in many fields from heterogeneous atmospheric processes to fundamental modeling of ions in solution. On the molecular level a certain number of water molecules are necessary for ion-pair formation to occur in the ground state. Femtosecond pump-probe studies of $HBr(H_2O)_n$ and $HI(H_2O)_n$ clusters have shown that five water molecules are necessary for ground state ion-pair formation in HBr whereupon solvent reorganization takes place around the newly formed ion-pairs. These studies show the importance of the halogen in the reorganization process as the reorganization dynamics of HI are slower than HBr despite similar excitation energies. Recent studies in our laboratory on $HI(H_2O)_n$ have uncovered evidence for a theoretically proposed excited state species, termed a biradical[6], that also has been proposed as an alternative moiety for the hydrated electron. Our studies of $HI(H_2O)_n$ reveal a long-lived state (~ 1 ns) after 6.2 eV excitation for all cluster, n = 1-6, and a likely evaporation of an iodine atom from the $HI(H_2O)_6$ cluster. This evaporation is expected from the predicted structure of the biradical which has an elongated bond between the halogen and the rest of the cluster[7]. The excitation energy (6.2 eV) is also consistent with theoretical predictions. Earlier work[3] at 9.45 eV of excitation also saw evidence of an evaporation of an I atom, suggesting, as predicted, that the biradical structure is the lowest energy excited state structure. Probe power studies reveal a progressive decrease in amount of photons needed for ionization as the number of water molecules increases, suggesting a threshold for the number of water molecules necessary to form the excited state biradical. We propose mechanisms based on the known excitation energy, probe power studies, and bond enthalpies to explain the observed phenomena. These mechanisms allow us to show that though caging could be involved in some of the processes seen in our experiments, other mechanisms are also at work. Our findings as well as hydrogen halide doped ice nanoparticle experiments[8] support the existence of a biradical like species and the concept of hydrated hydronium as an alternative moiety for the hydrated electron.

REFERENCES

1. Al-Halabi, A., R. Bianco, and J.T. Hynes, *Acid dissociation of HBr on a model ice surface.* Journal Of Physical Chemistry A, 2002. **106**(33): p. 7639-7645.
2. Gertner, B.J. and J.T. Hynes, *Molecular dynamics simulation of hydrochloric acid ionization at the surface of stratospheric ice.* Science, 1996. **271**(5255): p. 1563-1566.
3. Dermota, T.E., et al., *Photoinduced ion-pair formation in the (HI)(m)(H2O)(n) cluster system.* Journal Of Chemical Physics, 2005. **123**(21).
4. Hurley, S.M., et al., *The ultrafast dynamics of HBr-water clusters: Influences on ion-pair formation.* Journal Of Chemical Physics, 2003. **118**(20): p. 9272-9277.
5. Hurley, S.M., et al., *Dynamics of hydrogen bromide dissolution in the ground and excited states.* Science, 2002. **298**(5591): p. 202-204.
6. Sobolewski, A.L. and W. Domcke, *Hydrated hydronium: a cluster model of the solvated electron?* Physical Chemistry Chemical Physics, 2002. **4**(1): p. 4-10.

7. Sobolewski, A.L. and W. Domcke, *Photochemistry of HCl(H2O)(4): Cluster model of the photodetachment of the chloride anion in water.* Journal Of Physical Chemistry A, 2003. **107**(10): p. 1557-1562.
8. Poterya, V., et al., *Photodissociation of hydrogen halide molecules on free ice nanoparticles.* Journal Of Chemical Physics, 2007. **126**(7).

HELIUM EMBEDDED RARE GAS CLUSTERS UNDER A STRONG LASER PULSE

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While the mechanism of energy absorption of rare gas clusters from strong laser pulses of different wavelengths has been studied intensively over the last years and there is a good understanding of the basic processes [1], the knowledge of composite clusters is much more scarce. Here, we study theoretically Xe@He, i.e., xenon clusters (size of the order of 100 atoms) embedded in a superfluid droplet of helium (about 1000 atoms) and compare the results with those for isolated Xe clusters. The clusters are exposed to a strong laser pulse of 100 fs halfwidth and a peak intensity of $3 \cdot 10^{14}$ W/cm^2 at 800 nm wavelength.

The study is motivated by experiments (e.g., [2]) performed with helium embedded clusters. Production of clusters by growing them inside a superfluid droplet is an elegant alternative to supersonic expansion of an atomic beam. However, as found in [2] and analyzed here for rare gas cluster, the helium hull has significant influence on the dynamics of the cluster in the strong laser pulse. This is in contrast to the argument that the helium droplet has little influence since the ionization potential of helium is much higher than that of Xe or even metal clusters, and therefore helium is less responsive to the laser than the cluster inside. Yet, it was found experimentally [2] that the embedded cluster explodes *faster* than the isolated one. This is surprising since one may naively expect that, if at all, the influence of the surrounding helium is to delay the cluster explosion.

Our calculations show that there are many more active electrons in the vicinity of the Xe@He cluster ("quasi-free electrons") than in the Xe cluster (see Fig. 1), i.e., helium acts as an electron "source". Theses electrons are attracted to the cluster center due to the background charge generated by the ionized Xe atoms. Their field at the position of the helium atoms outside is much stronger than the laser field and easily overcomes the binding potential.

The high electron density allows for an efficient transfer of energy from the laser pulse to the cluster electrons which is apparent from their temperature in comparison to the electron temperature in the isolated xenon cluster (Fig. 2). More efficient energy transfer leads to higher ionic charge states. One also sees from Fig. 2, that this energy transfer occurs earlier than for the isolated cluster. These two factors give rise to a faster explosion of the embedded cluster. The mechanism of the explosion, including the role of collective excitation [3], will be discussed in detail.

References

[1] U. Saalmann, Ch. Siedschlag and J. M. Rost, TopicalReview, J. Phys. B **39**, R39 (2006)

[2] T. Döppner et al., EPJD **24**, 157 (2003)

[3] U. Saalmann and J. M. Rost, Phys. Rev. Lett. **91**, 223401 (2003).

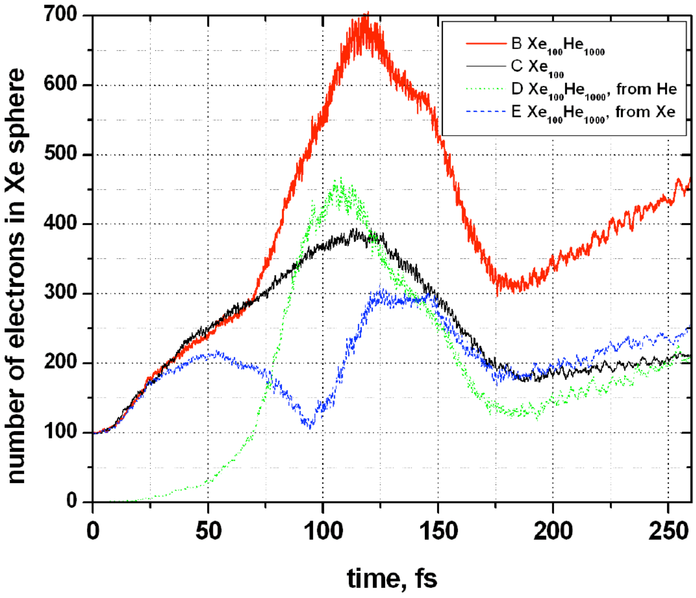

Figure 1: The number of quasi-free electrons as a function of time within the Xe-cluster volume: highest curve at right boundary (red) – Xe_{100}@He_{1000}; second highest curve (blue), electrons from Helium atoms – Xe_{100}@He_{1000}; third curve (black) – Xe_{100}; lowest curve (green), electrons from Xe atoms – Xe_{100}@He_{1000}.

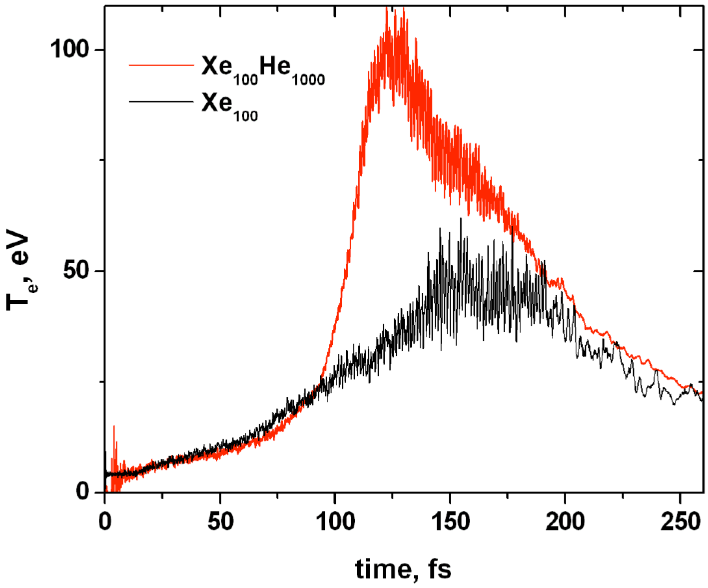

Figure 2: Temperature of quasi-free electrons for Xe_{100}@He_{1000} (higher curve, red) and Xe_{100}.

Sa-III-3

LARGE-AMPLITUDE DYNAMICS OF CLUSTERS

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The talk presents theoretical studies of coupled ionic and electronic dynamics of metal clusters. It considers free clusters as well as clusters in contact with an inert environment (rare gases, insulator substrates as MgO). After a brief introduction of the basic cluster properties, it concentrates on the highly non-linear regime following excitation by strong laser fields or particle impact. The studies resolve the fully detailed electronic dynamics at fs scale and proceed up to several ps to track the consequences on the slow ionic and atomic motion in these complex compounds.

The observable signatures of the various dynamical effects are discussed, e.g., photo-electron spectroscopy, angular distributions, or pump-and-probe analysis. For embedded and deposited clusters, there emerges the particularly interesting question of energy and particle transport from the highly excited cluster (acting as chromophore) to the inert environment. The various transport processes and corresponding time scales will be analyzed. We will also address the remarkable difference in the dynamics of free clusters and clusters embedded in rare-gas matrix. For example, the environment stops the initially strong Coulomb explosion and stabilizes clusters at rather high charge states.

The theoretical tool for these investigations is time-dependent density-functional theory at the level of the local-density approximation. A much similar method, using the Skyrme energy functional, is used in the description of large-amplitude dynamics of nuclei and heavy-ion collisions. Some examples will be shown in comparison.

Sa-III-4

OPTIMAL CONTROL ON ALKALI SYSTEMS

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Control of photo-induced molecular processes has attained considerable success in recent years. It became most exciting when self-learning feedback loop algorithms were employed where tailored laser pulses can be generated, which drive the induced processes at a maximum yield along desired paths [1]. An important issue in this regard is the information coded in the optimized laser pulse shape which supplies insight about the underlying processes. Small alkali systems are suitable for this aim since they exhibit a number of bound states available for resonant transitions with weak fields which aids the theoretical description and hence the interpretation.

Closed loop optimizations on alkali dimers in a supersonic beam are first demonstrated for maximization of the ionization yield. Next, isotopomer selective optimizations are presented to examine the efficiency of the optimization procedure for the weak differences between the isotopic species. Surprisingly large enrichment factors are found and information about the dynamics on the involved vibrational states is extracted from the optimal pulse shapes, which provides a new spectroscopical approach of yielding distinct frequency pattern on fs-time scales [2]. Both cases are compared with optimal control calculations in order to decipher the underlying processes in detail [3,4].

A main aspect of this contribution is the development of novel optimal control methods to extract the most relevant information from the optimized laser field. One approach examines the implementation of genetic pressure within the algorithm for performing control pulse cleaning where extraneous pulse features were removed to expose the most important structures. This leads also to first investigations on multi-criteria optimizations [5]. Another method addresses parametric optimization by introducing physically relevant pulse parameters employing a computer assisted sub pulse encoding in order to narrow the search space and to aid the interpretation. Moreover, novel pulse shaper schemes for combined phase, amplitude, and polarization pulse control were developed and applied on alkali dimers, even in a parametric encoding. The results demonstrate the perspectives of adding a new dimension by including also the polarization and hence all properties of the light field in the pulse modulation [6].

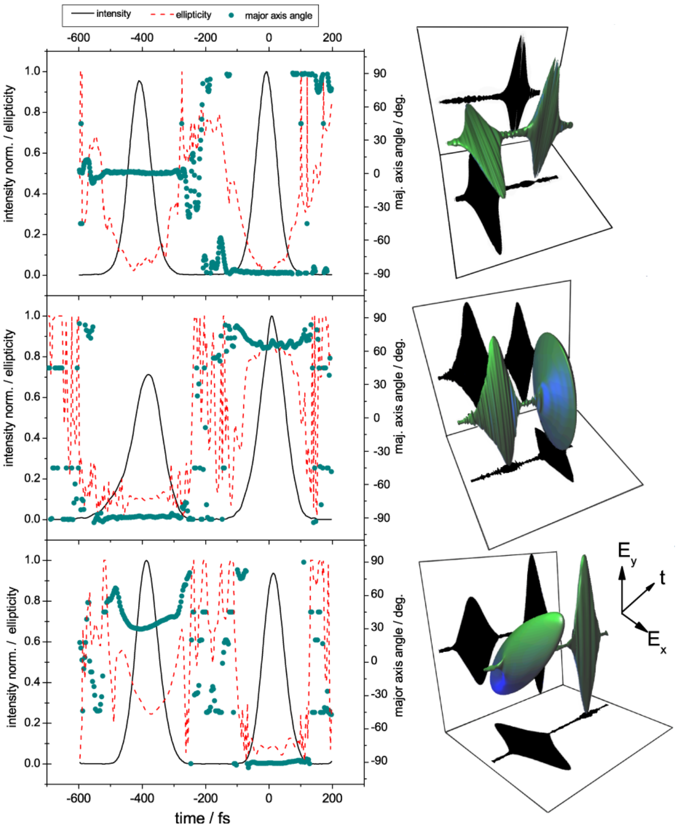

Figure 1: Parametrically shaped pulseforms where the phase, amplitude, and polarization are subject to modulation. On top are two linear polarized perpendicular sub pulses, in the middle a linear and a circular polarized subpulse, and the lowest figure shows two sub pulses with the major axes at 90 and 30 to the horizontal x-direction, respectively.

Currently, coherent control is applied to ultracold trapped ensembles motivated by the perspective to perform photoassociation and photostabilization of alkali systems. First results are received regarding optimized multi-photonic excitation to molecular ions and pump-probe experiments exposing signal oscillations. They provide indications for photoassociation and open the perspective for transitions to lower vibrational levels in the electronic ground state, which would be a first step to an internally cold molecular Bose Einstein condensate.

REFERENCES

[1] R. S. Judson and H. Rabitz, *Phys. Rev. Lett.* **68**, 1500 (1992)

[2] A. Lindinger, C. Lupulescu, M. Plewicki, F. Vetter, S. M. Weber, A. Merli, and L. Wöste, *Phys. Rev. Lett.* **93,** 033001 (2004)

[3] B. Schäfer-Bung, R. Mitric, V. Bonacic-Koutecky, A. Bartelt, C. Lupulescu, A. Lindinger, S. Vajda, S. M. Weber, and L. Wöste, *J. Phys. Chem. A* **108**, 4175 (2004)

[4] B. Schäfer-Bung, V. Bonacic-Koutecky, F. Sauer, S. M. Weber, L. Wöste, and A. Lindinger, *J. Chem. Phys.* **125,** 214310 (2006)

[5] A. Lindinger, S. M. Weber, C. Lupulescu, F. Vetter, M. Plewicki, A. Merli, L. Wöste, A. Bartelt, and H. Rabitz, *Phys. Rev. A* **71,** 013419 (2005)

[6] F. Weise, S. M. Weber, M. Plewicki, and A. Lindinger, *Chem. Phys.* **332,** 313 (2007)

Sa-III-4

Clustering phenomenon in systems of various degrees of complexity

Mo-I-1

ELECTRON-POSITRON CLUSTERS: STRUCTURE AND STABILITY

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In this contribution we discuss the new quantum object of finite size - electron-positron droplet [1]. These systems consist of the number of electrons and positrons which are held together by the attractive Coulomb and mutual polarization forces and form the shell structure similar to metal clusters [2]. Contrary to metal clusters, in electron-positron clusters the motion of negatively and positively charged subsystems is identical and quantized. The main properties of these clusters of different size such as the structure, stability and dynamics are under consideration. The calculated structure is compared with the properties of metal clusters, which were obtained in particular within the ordinary and Optimized Jellium Model (OJM) [3]. In order to prove the existence of electron-positron clusters we analyze the stability of these objects against annihilation process and fragmentation into a number of positroniums. Our analysis of the electron-positron cluster properties is based on both the self-consistent Hartree-Fock (HF) approximation and the formalism of non-relativistic density functional theory. The latter is realized within the Local Density Approximation (LDA) by the self-consistent solution of the Kohn-Sham equations with different parameterizations of the exchange-correlation potential.

The shell structure in the system arises in self-consistent way. Due to the charge symmetry the electron and positron subsystems have the identical shell structure. Numerically, this condition was fulfilled by the iterative solution of the HF and LDA equations. This procedure permits to obtain the single-particle energies and wave functions. The electron-positron droplet structure has been calculated for the clusters with number of electron-positron pairs $N = 2, 8, 18, 20, 40$. These numbers of electrons (positrons) correspond to the cluster with the spherically symmetric density distribution and close $1s^2$, $1p^6$, $1d^{10}$, $2s^2$ $1f^{14}$ and $2p^6$ shells, respectively. Many-body correlations are taken into account within the LDA with both the simplest Gunnarsson - Lundqvist and Perdew and Wang (PW91) parameterizations for the exchange-correlation functional [4]. The exchange-correlation potential includes three parts: the local exchange interaction between equivalent particles and the correlation interaction between equivalent and non-equivalent particles. The total binding energies of electron-positron clusters per one particle are comparable with the binding energy of positronium.

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REFERENCES

[1] A.V. Solovyov, V.K.Ivanov, R.G.Polozkov, *Eur. Phys. J. D* **40**, 313 (2006).
[2] *Metal clusters*, edited by W. Ekardt (Wiley, New York, 1999).
[3] V.K. Ivanov, V.A. Kharchenko, A.N. Ipatov, M.L. Zhizhin, *Pis'ma JETPh* **60**, 345 (1994).
[4] J.P. Perdew, Y. Wang, *Phys. Rev. B* **45**, 13244 (1992).

Mo-I-2

FORMATION AND STABILITY OF GAS PHASE MULTIPLY CHARGED (+/-) CLUSTERS OF BIOMOLECULES

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We have studied multiply-charged noncovalent cluster anions of adenosine-5'-monophosphate (AMP) formed by electrospray ionization (ESI). Ions in higher charge states were observed when the ions were accumulated in an ion trap with helium buffer gas. We have determined the smallest size (n_a) or appearance size as a function of charge state (q), i.e., $n_a = 4$ for $q = 2$, $n_a = 8$ for $q = 3$, and $n_a = 13$ for $q = 4$. The relation between n_a and q (see Fig.1) can be described by a charged droplet model. When the size is larger than n_a for a given q, the fragmentation pathway of an anion cluster is dominated by loss of neutral fragments. In contrast, when the size approaches the appearance size, only charged fragments are formed [1].

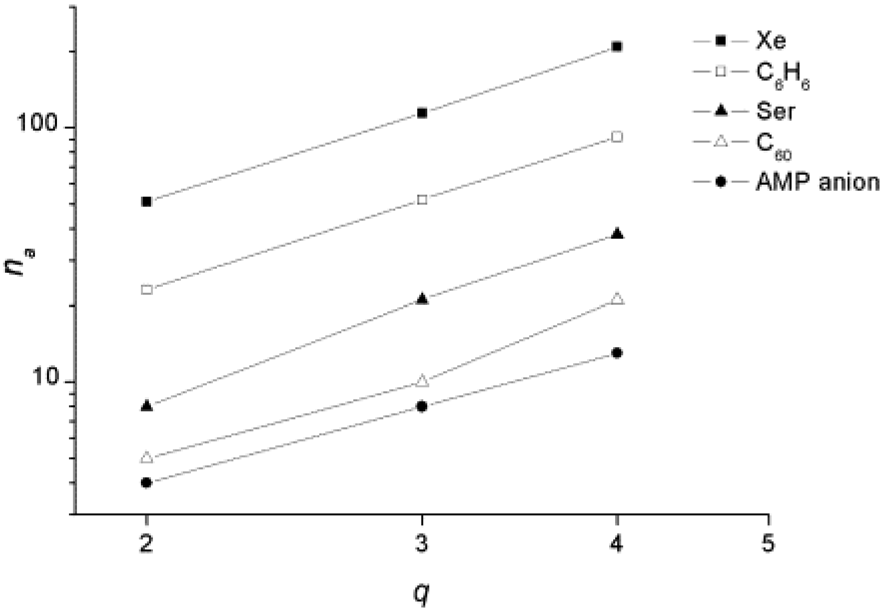

Figure 1: The log−log plot shows that n_a scales approximately with q^2 for a selection of positive clusters ions as well as for negative (AMP) cluster ions.

Positively charged amino acid clusters have been studied by several groups and most notably magic number clusters and chiral recognition have been reported. We have studied the formation of amino acid clusters by electrospray ionization (ESI) and their stability by high-energy collision-induced dissociation (CID) [2]. Appearance sizes were determined for multiply charged clusters where the charge is either due to protons or to sodium ions. We found that chiral selectivity plays an important role in cluster formation but seems to be of minor importance for the fragmentation of mixed clusters.

In a resent series of experiments we have studied how nucleotide or peptide ions embedded in water clusters react in high energy collisions with atoms [3]. We found that loss of water becomes the dominating fragmentation channel for large clusters of this type. The embedded biomolecule in other words seems to be well protected when surrounded by a large number of water molecules.

Mo-I-2

REFERENCES
[1] B. Liu, P. Hvelplund, S.Brøndsted Nielsen. *J Am Soc Mass Spectrom*, **16**,1845 (2005)
[2] B. Concina, P. Hvelplund, A.B. Nielsen, S.Brøndsted Nielsen, J. Rangama, B. Liu, S.Tomita. *J Am Soc Mass Sprectrom*, vol. **17**, 275 (2006)
[3] B. Liu, S.Brøndsted Nielsen, P. Hvelplund, H. Zettergren, H. Cederquist, B. Manil, B.A. Huber, *Phys. Rev. Lett* **97**, 133401.(2006)

Mo-I-3

SPECTROSCOPY AND DECAY OF CHROMOPHORE IONS IN THE GAS PHASE

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Absorption of light in the visible part of the spectrum normally results in excitation of chromophores from the electronic ground state to the first or second electronically excited states. When chromophores and their ions (protonated/deprotonated) are found in biological environments there will typically be interactions that may change the absorption wavelength as well as the speed and quantum yields of the molecular response. To have a reference for such interactions we are studying the absorption properties and the molecular response times of chromophores in the gas phase, i.e. in the completely isolated form devoid of external perturbations from charges, dipoles and hydrogen bonding [1-13]. It appears that in some cases (like GFP), almost vacuum-like conditions exist inside proteins whereas in other cases (like retinal) there are significant protein perturbations.

REFERENCES

[1] L. Lammich *et al.*, Biophysical J. **92** 201 (2007)
[2] I. B. Nielsen *et al.*, J. Phys. Chem. A **110** 12592 (2006)
[3] I. B. Nielsen, L. Lammich and L.H. Andersen. Phys. Rev. Lett. **96** 018304 (2006)
[4] I. B. Nielsen *et al.*, Biophys. J., **89** 2597-2604 (2005)
[5] L. H. Andersen *et al.*, J. Am. Chem. Soc. **127** 12347-12350 (2005)
[6] L. H. Andersen *et al.*, Phys. Chem. Chem. Phys., **6** 2617 – 2627 (2004)
[7] S. Boyé *et al.*, Phys. Chem. Chem. Phys. **5**, 3021-3026 (2003)
[8] S. Boyé *et al.*, J. Chem. Phys **119**, 338-345 (2003)
[9] S. Boyé, *et al.*, Phys. Rev. Lett. **90** 118103-1 (2003)
[11] L. H. Andersen, S.B. Nielsen, S.U. Pedersen. Eur. Phys. J. D. **20** 597-600 (2002)
[12] L. H. Andersen *et al.*, Nucl. Phys. Review. **19** 176 (2002)
[13] S. B. Nielsen *et al.*, Phys. Rev. Lett. **87** 2281021 (2001)

Mo-I-4

ENERGY LANDSCAPES: STRUCTURE, DYNAMICS AND THERMODYNAMICS

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Coarse-graining the potential energy surface into the basins of attraction of local minima [1, 2], provides a powerful framework for global optimisation via basin-hopping [3, 4] and for calculating thermodynamic properties using the superposition approach [1, 5] and basin-sampling [6]. To admit dynamics we must include transition states of the potential energy surface, which link local minima via steepest-descent paths. This framework leads to the discrete path sampling method, which provides access to rate constants for rare events [7–9]. In large systems the paths between different morphologies may involve hundreds of stationary points of the potential energy surface. New algorithms have been developed for both geometry optimisation [10] and making connections between distant local minima [11], which allow us to treat such systems. Kinetic analysis of large stationary point databases is facilitated by a new graph transformation approach [12].

Applications will be presented for a wide variety of atomic and molecular clusters, including systems bound by isotropic interatomic potentials such as the Lennard-Jones and Morse forms, clusters of dipolar spheres [13], and molecular clusters such as nanodroplets of water [14–16].

REFERENCES

[1] D. J. Wales, Energy Landscapes, Cambridge University Press, Cambridge (2003).
[2] D. J. Wales and T. V. Bogdan, J. Phys. Chem. B 110, 20765 (2006).
[3] D. J. Wales and J. P. K. Doye, J. Phys. Chem. A 101, 5111 (1997).
[4] D. J. Wales and H. A. Scheraga, Science 285, 1368 (1999).
[5] D. J. Wales, Mol. Phys. 78, 151 (1993).
[6] T. V. Bogdan, D. J. Wales and F. Calvo, J. Chem. Phys. 124, 044102 (2006).
[7] D. J. Wales, Mol. Phys. 100, 3285 (2002).
[8] D. J. Wales, Mol. Phys. 102, 891 (2004).
[9] D. J. Wales, Int. Rev. Phys. Chem. 25, 237 (2006).
[10] S. A. Trygubenko and D. J. Wales, J. Chem. Phys. 120, 2082 (2004).
[11] J. M. Carr, S. A. Trygubenko and D. J. Wales, J. Chem. Phys. 122, 234903 (2005).
[12] S. A. Trygubenko and D. J. Wales, J. Chem. Phys. 124, 234110 (2006).
[13] M. A. Miller and D. J. Wales, J. Phys. Chem. B 109, 23109 (2005).
[14] D. J. Wales, M. A. Miller and T. R. Walsh, Nature 394, 758 (1998).
[15] D. J. Wales and M. P. Hodges, Chem. Phys. Lett. 286, 65 (1998).
[16] M. P. Hodges and D. J. Wales, Chem. Phys. Lett. 324, 279 (2000).

Mo-I-4

Structure and dynamics of bio-macromolecules

Mo-II-1

VIBRATIONAL FESHBACH RESONANCES IN LOW-ENERGY ELECTRON COLLISIONS WITH MOLECULES AND MOLECULAR CLUSTERS

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Temporary negative ion states XY^-, formed in low energy electron collisions with molecules XY, are crucial for vibrational excitation as well as for formation of anions by dissociative electron attachment (DEA) [1,2]. Both shape resonances (electrons trapped within a centrifugal barrier) and Feshbach resonances (electrons attached to electronically excited states XY^* of the neutral molecule) are representatives of such temporary negative ion states, typically located at incident electron energies of 1 to 10 eV. At very low energies (in the range of vibrational onsets) another type of resonance can occur which is mediated by attractive long-range interactions in the electron-target system and may exist without the supporting influence of a centrifugal barrier, i.e. for s-wave electrons. These anion resonances are found just below vibrational onsets of the molecule in its electronic ground state and are therefore addressed as vibrational Feshbach resonances (VFR) [2]. In cross sections for elastic and vibrationally inelastic scattering, VFRs show up as narrow features (typically dips), as experimentally observed, e.g., for XY = HF [3,4] and CH_3I [2,5]. In DEA to molecules and molecular clusters, VFRs may lead to a prominent enhancement of anion formation, as observed, e.g., for the molecules CH_3I [2,6], N_2O [2,7], CH_3Br [8], the biomolecules uracil and thymine [9], and for clusters composed of N_2O [2,10], O_2 [11], CO_2 [2,12], OCS [2,13]. VFRs with widths as narrow as 2 meV have been detected with the Laser Photoelectron Attachment method [2,10]. Depending on the nature of the electron–molecule interaction at long and short range, the VFRs are more or less red-shifted from the vibrational levels of the neutral precursor, as reproduced in model calculations for VFRs of N_2O and CO_2 clusters [2,12]. Recent calculations of VFR structure in electron attachment to CO_2 clusters [14,15] show qualitative agreement with the experimental observations and predict VFRs to play an important role for vibrational excitation of CO_2 clusters [14,15].

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REFERENCES

1. E. Illenberger, in *Photoionization and Photodetachment* Part II (Adv. Ser. Phys. Chem., Vol. 10 B, Ed. C.-Y. Ng), p. 1063 ff., World Scientific, Singapore (2000)
2. H. Hotop, M.-W. Ruf, M. Allan, I. I. Fabrikant, Adv. At. Mol. Opt. Phys. **49** (2003) 85
3. G. Knoth et al., Phys. Rev. Lett. **62** (1989) 173
4. M. Cizek, J. Horacek, M. Allan, I. I. Fabrikant, W. Domcke, J. Phys. B **36** (2003) 2837
5. M. Allan, I. I. Fabrikant, J. Phys. B **35** (2002) 1025
6. A. Schramm, I. I. Fabrikant, J. M. Weber, E. Leber, M.-W. Ruf, H. Hotop, J. Phys. B **32** (1999) 2153
7. M. Allan, and T. Skalicky, J. Phys. B **36** (2003) 3397
8. M. Braun, I. I. Fabrikant, M.-W. Ruf, H. Hotop, J. Phys. B **40** (2007) 659
9. P. D. Burrow, G. A. Gallup, A. M. Scheer, S. Denifl, S. Ptasinska, T. Märk, P. Scheier, J. Chem. Phys. **124** (2006) 124310
10. J. M. Weber, E. Leber, M.-W. Ruf, H. Hotop, Phys. Rev. Lett. **82** (1999) 516
11. S. Matejcik, A. Kiendler, P. Stampfli, A. Stamatovic, T. D. Märk, J. Chem. Phys. **111** (1999) 3548
12. E. Leber, S. Barsotti, I. I. Fabrikant, J. M. Weber, M.-W. Ruf, H. Hotop, Eur. Phys. J. D **12** (2000) 125
13. S. Barsotti, T. Sommerfeld, M.-W. Ruf, H. Hotop, Int. J. Mass Spectrom. **233** (2004) 181
14. I. I. Fabrikant, H. Hotop, Phys. Rev. Lett. **94** (2005) 063201
15. I. I. Fabrikant, J. Phys. B **38** (2005) 1745

Mo-II-2

STRUCTURE OF UNFOLDED POLYPEPTIDES

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Unfolded proteins have recently attracted a lot of attention when it became evident that the intrinsically unstructured proteins comprise a large part of the proteins being encoded in eukaryotic genomes [1]. The open question, which is important, e.g., for the protein folding studies, is how much of residual structure is actually present in the unfolded proteins. High-resolution, liquid-state nuclear magnetic resonance (NMR) spectroscopy is an ideal tool for answering this question. Several experimental observables (e.g., chemical shifts) can be used for this purpose. One of such observables is the direct dipole-dipole interactions between nuclear spins (the so-called residual dipolar couplings, RDCs). The RDCs are expressed via the average orientation of internuclear vectors in an anisotropic environment (created by dissolving the polypeptide in a liquid crystal medium). As RDCs throughout the polypeptide chain are correlated, NMR spectroscopy makes it possible to deduce the persistent residual structure of the chain [2].

We have developed a theoretical framework [3] suitable for describing various characteristics of unfolded polypeptides including the prediction of RDCs. The framework is rather general and can serve as a basis for determining RDCs in unfolded polypeptide chains under a wide spectrum of experimental conditions. The framework allows one to employ various models of polypeptide chains and of aligning media for finding RDCs with the desired degree of accuracy. Using the framework we showed that within a simple model which approximates the alignment media as infinite planes and in which unfolded polypeptides are described within the random walk formalism, it is possible to obtain a closed-form analytical result for the RDCs. The two general features predicted by the model are (i), RDCs in the center of the chain are larger than RDCs at the ends and (ii), RDCs are larger for shorter chains than for longer chains at a given liquid crystal medium concentration. We demonstrate how these features can be explained from the general physical considerations. Experimental data available from the literature confirm the first prediction of the model, providing, therefore, a tool for recognizing fully unfolded polypeptide chains. With less certainty experimental data appear to support the second prediction as well.

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REFERENCES

[1] A. L. Fink, *Curr. Opin. Struct. Biol.* **15**, 35 (2005)
[2] M. Blackledge, *Progr. Nucl. Magn. Reson. Spectrosc* **46**, 23 (2005)
[3] O. I. Obolensky, K. Schlepckow, H. Schwalbe, A. V. Solov'yov, accepted for *J. Biomol. NMR* (2007)

Mo-II-3

COMPUTATIONAL SIMULATIONS OF ANTIBODY: ANTIGEN UNBINDING

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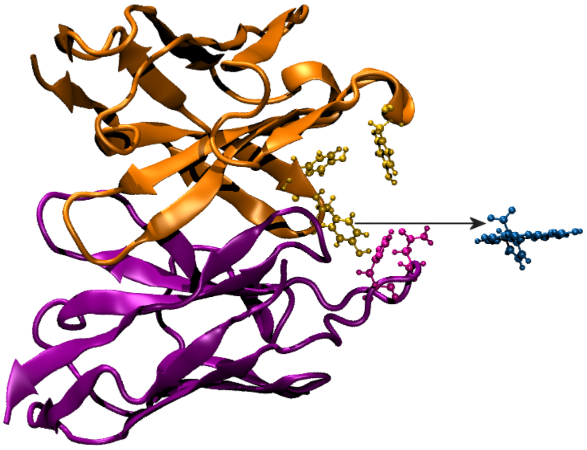
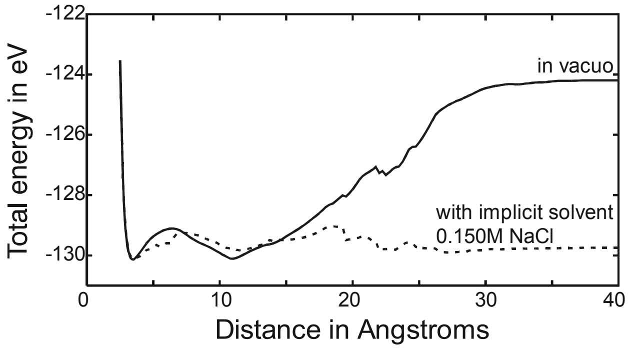
Life is driven by interactions between the molecular components of cellular machinery. Most cellular processes portray a cascade of association/dissociation events between proteins and their target molecules. The dissociation events are, in essence, a fragmentation of complex multi-atomic aggregates.

The fragmentation of multi-atomic aggregates has been extensively studied in atomic cluster physics, the emerging key idea being that such processes can be successfully described in terms of a few generalized coordinates that define the overall configuration of the escaping and parent fragments [1]. This concept also holds for similar events in more complex systems [2], and it prompted us to now address the dissociation process of an aggregate of higher complexity, a biological one.

Antibody–antigen complexes are involved in fundamental recognition processes during the body immune response. A reference example is the anti–fluorescein system, and the present work reports a rational computational approach to probe its unbinding at the atomistic classical-mechanical level. For the wild type antibody Fab4-4-20 and related mutations, the complex problem has been reduced to a low-dimensional scanning along a few selected coordinates (exemplified in figure a) [3]. Positional and orientational coordinates of the escaping fluorescein (the antigen) have been assessed in order to fully characterize the unbinding. Solvent effects have been accounted for, primarily by means of a continuum model, though the explicit inclusion of water has been also addressed. The dissociation times of the complexes have been derived from the calculated barrier heights (see figure b), in compliance with the experimentally reported Arrhenius-like behavior. The computed results are discussed and compared with the available experimental data.



(a)



(b)

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REFERENCES
[1] A. V. Solov'yov, J.-P. Connerade, W. Greiner, in *Latest Advances in Atomic Cluster Collisions*, Imperial College Press, London (2004).
[2] I. A. Solov'yov, A. Yakubovich, A. V. Solov'yov, W. Greiner, *JETP*, **103**, 463 (2006).
[3] E. S. Henriques, A. V. Solov'yov, *arXiv: 0704.3193v1* (2007).

Mo-II-4

CRYPTOCHROME-1 MAGNETORECEPTION

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The ability of some animals, most notably migratory birds, to sense magnetic fields is still poorly understood. It has been suggested [1,2] that animal magnetoreception is mediated by the blue light receptor protein cryptochrome, which is known to be localized in the retinas of migratory birds. Recent experiments [3] have shown that the activity of cryptochrome-1 in *Arabidopsis thaliana* is enhanced by the presence of a weak external magnetic field, confirming the ability of cryptochrome to mediate magnetic field responses.

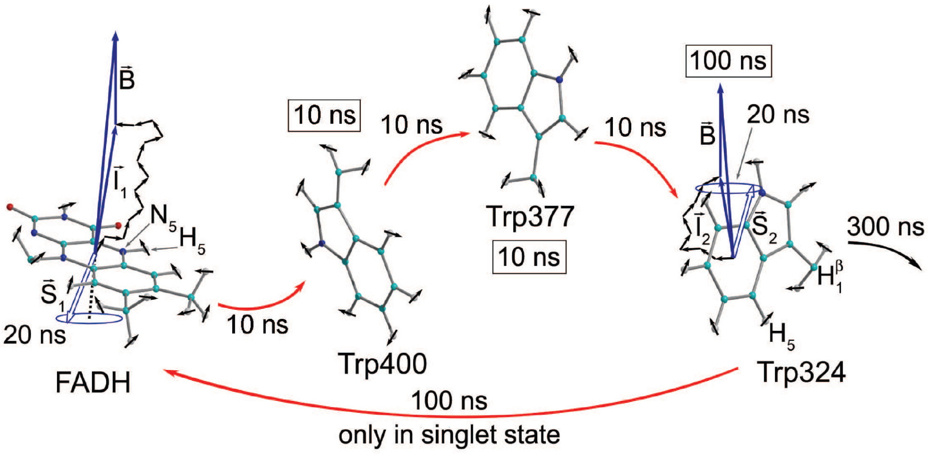

Figure 1: A semi-classical description of the magnetic field effect on the radical pairs between FADH and tryptophan in cryptochrome.

Cryptochrome's signaling is tied to the photoreduction of an internally bound chromophore, flavin adenine dinucleotide (FAD). The spin chemistry of this photoreduction process is schematically shown in Fig. 1. It involves electron transfer from a chain of three tryptophans, and can be modulated by the presence of a magnetic field through the so-called radical pair mechanism based on Zeeman and hyperfine coupling. We present calculations showing that the radical pair mechanism in cryptochrome can produce an increase in the protein's signaling activity of approximately 10% for magnetic fields on the order of 5 Gauss, which is consistent with experimental results [3]. These calculations, in view of the similarity between bird and plant cryptochromes, provide further support for a cryptochrome-based model of avian magnetoreception.

REFERENCES

[1] Ritz T., Adem S., and Schulten K., *Biophysical Journal*, **78**, 707 (2007)
[2] Solov‘yov I., Chandler D., and Schulten K., *Biophysical Journal*, **92**, 2711 (2007)
[3] Ahmad M., Galland P., Ritz T., Wiltschko R. and Wiltschko W., *Planta*, in press. DOI 10.1007/s00425-006-0383-0 (2007).

Collision processes involving biomolecules

Mo-III-1

ION INDUCED FRAGMENTATION OF NUCLEOBASE AND AMINO-ACID CLUSTERS

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Recently, a large number of studies have been devoted to the investigation of molecular ionization and fragmentation dynamics underlying biological radiation damage. Most of these studies were based on gas phase collisions with isolated DNA building blocks [1]. The radiobiological significance of these studies is often questioned because of the lack of a chemical environment.

Radiation damage in living cells always involves the condensed phase where the affected molecules are surrounded by a medium. Ion irradiation studies are therefore often performed on nucleobases in the solid phase [2] and on DNA deposited on solid surfaces [3]. Such studies are hampered by the complexity of the systems under study.

A natural solution to avoid these difficulties is the investigation of finite systems, which still allow intermolecular interactions, i.e. clusters of DNA building blocks or mixed clusters containing biomolecules and water. Spectroscopic techniques, which proved their value in gas phase studies, can still be used to study the interaction and dissociation dynamics of such clusters. We studied interactions of keV ions with isolated nucleobases/amino-acids and their clusters by means of coincidence time-of-flight spectrometry. Significant changes show up in molecular fragmentation for very small cluster sizes already, which we attribute to the influence of intermolecular hydrogen bonding.

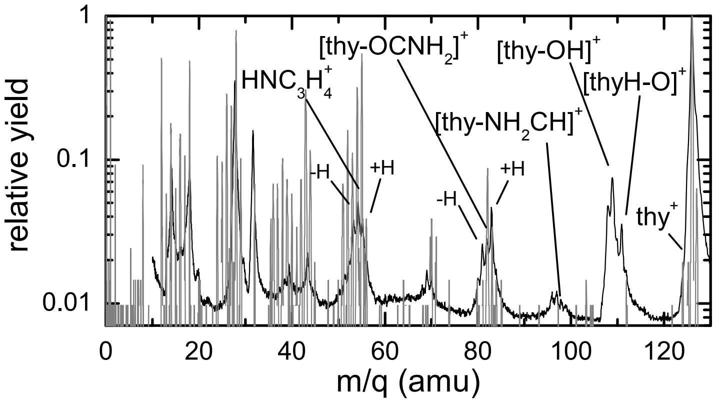

Figure 1 **Mass spectra of fragmentation products for collisions of 60 keV C5+ with isolated thymine (grey) and for 50 keV O5+ with thymine.**

Fig.1 shows that for instance the loss of O and OH is only observed in the presence of a chemical environment. Similar results are observed for other nucleobase clusters, as well. Note, that these channels are also seen ion induced desorption studies on thin nucleobase films [2] and can thus be regarded as fingerprints of the condensed phase. Currently, experiments on trapped biomolecular cluster ions are in progress where biologically more realistic systems will be investigated

REFERENCES

[1]J. de Vries et al., PRL **91** (2003) 053401
[2] Z. Deng et al., PRL **95** (2005) 153201
[3] S. Lacombe et al., Phys. Med. Biol. **49** (2004) N65
[4] T.Schlathölter et al., ChemPhysChem **7** (2006) 2339

Mo-III-2

BIOPHYSICAL MODELLING OF FRAGMENT DISTRIBUTIONS OF DNA PLASMIDS AFTER PHOTON AND HEAVY ION IRRADIATION

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The production of double strand breaks (DSB) is considered to be the most severe single radiation-induced damage for numerous types of biological endpoints like cell inactivation or chromosomal aberrations. It has become apparent that not the mere number of DSB(s) represents the observed radiation damage, but the spatial distribution of DSB(s) seems to be crucial for understanding the biological response. In order to investigate the impact of localized energy deposition generated by ion irradiation on biological effects, it is desirable to exploit a simple model system which allows us to clearly deduce the influence of track structure on the induction of radiation damage. For this purpose, we decided to use plasmid DNA with its simple geometrical conformation for our investigations elucidating the influence of localized energy distribution on the induction of DNA damage, particularly the clustering of double strand breaks. After X-ray or ion irradiation DSB(s) are induced in the plasmid molecule leading to DNA fragmentation. The resulting fragment length distribution is analyzed by carefully measuring the fragment length with Atomic Force Microscopy (AFM) directly with a precision of a few tens of nanometers[1].

We present a model that describes fragment length distributions of linear and supercoiled plasmid DNA after irradiation with heavy ions by combining the Local Effect Model [1] originally developed to predict cell survival with the widely used broken stick approach for X-rays [2]. With this concept, it is possible to rapidly calculate fragment distributions for a broad range of ions and energies. These simulations use the amorphous track structure model of the LEM to determine the local dose delivered to the plasmid by the ions. For the small plasmid volume, the local dose is averaged and the probabilistic considerations of the broken stick model are applied to determine the fragmentation pattern.

For a reasonable comparison of simulated results with experimental data, we need to take experimental constraints, such as the minimum detection length of plasmid DNA, the limited resolution of the AFM measurement and the selection method of analyzable fragments into account. The integration of these experimental constraints into the model calculations changes the resulting distributions strongly. We find a good agreement of our simulations with experimental fragment distributions using atomic force microscopy. The model calculations reveal that for very high-LET irradiation using nickel or uranium ions no difference in the fragment distribution can be expected for the same dose level, since the higher fragmentation power of uranium ions is compensated for by the higher hit probability of nickel ions. However, for carbons ions with an intermediate LET, the fragmentation pattern is almost fully resolved and the influence of track structure may be clearly demonstrated. By comparing the fragmentation in terms of average hits per DNA molecule, we can uniquely distinguish the effect of different radiation qualities. In order to better discriminate between fractions of long, most probably intact molecules and short fragments, we propose a fragmentation parameter η describing the radiation effect of the non-random spatial distribution of DSB(s). Finally, we simulate different ion species to understand the degree of fragmentation in relation to LET as well as to find the most appropriate ion species to visualize the difference between photon and heavy ion irradiation.

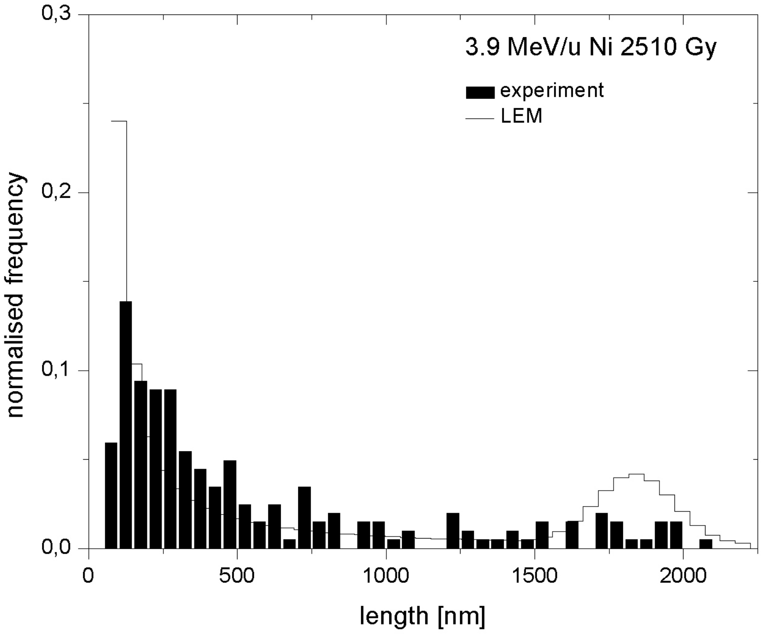

Figure 1: Comparison of measured (AFM) and simulated fragment distributions of supercoiled plasmid DNA following irradiation with 3.9 MeV/u Ni ions with a dose of 2510 Gy taking into account experimental constraints.

REFERENCES

[1] K. Psonka, E. Gudowska-Nowak, S. Brons, Th. Elsässer, M. Heiss, G. Taucher-Scholz, DNA fragmentation induced by ionizing radiation – Atomic Force Microscopy and Biophysical Modeling, Adv. Space. Res., doi:10.1016/j.asr.2007.02.089 (2007)

[2] M. Scholz and G. Kraft, Calculation of heavy ion inactivation probability based on track structure, x-ray sensitivity and target size. Radiat. Prot. Dosim. 52, 29-33 (1994).

[3] T. Radivoyevitch and B. Cedervall, Mathematical analysis of DNA fragment distribution models used with pulsed-field gel electrophoresis for DNA double-strand break calculations. Electrophoresis 17, 1087-1093 (1996).

Mo-III-3

THE INCREASED BIOLOGICAL EFFECTIVENESS OF ION BEAM RADIATION: EXPERIMENTAL AND THEORETICAL ASPECTS

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The effect of cell killing after ion beam irradiation in general is enhanced as compared to conventional photon beam irradiation. This difference constitutes one of the major rationales for the application of ion beams in tumour therapy. However, the precise knowledge of the biological action of charged particle radiation is not only relevant for ion beam therapy, but for radiation protection purposes as well, since also for other biological endpoints like cell transformation an increased effectiveness of charged particle beams is observed. Furthermore, it is a relevant issue in space research, because heavy charged particles are responsible for a significant fraction of radiation damage induced by cosmic rays.

The increased effectiveness is usually described in terms of the Relative Biological Effectiveness (RBE), defined by the ratio of doses which have to be applied to achieve the same biological effect with conventional photon and ion beam radiation, respectively. However, the RBE cannot be represented by a single number, but instead depends on several physical and biological parameters, which include e.g. the dose level, the ion species, beam energy and the cell or tissue type under consideration. These systematic dependencies will be explained using experimental data for cell inactivation, representing an important endpoint with respect to tumor cell killing and thus tumor control probability.

Because of the complex dependencies of RBE as described above, biophysical modeling is required for applications in ion beam therapy or risk estimates of space radiation. Photon and ion beam radiation mainly differ with respect to their microscopic energy deposition pattern; therefore, the spatial energy deposition pattern on a subcellular scale is a major determinant of the RBE. But even for the same physical radiation field, the response of different biological objects might differ as well. Therefore, biological parameters also play a crucial role. A model approach will be presented, which combines both physical and biological aspects and allows us to predict the response of a biological system to particle radiation on the basis of the response to conventional photon radiation.

From biomolecules to cells and system biology

TOWARDS MONTE CARLO CALCULATIONS OF BIOLOGICAL DOSE IN HEAVY-ION THERAPY: MODELING OF NUCLEAR FRAGMENTATION REACTIONS

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We study the propagation of carbon nuclei in tissue-like media within a Monte Carlo Model for Heavy-Ion Therapy (MCHIT) [1, 2]. The model is based on the GEANT4 toolkit [3] and properly takes into account fragmentation of projectile nuclei and interactions of secondary particles in water and PMMA phantoms. Electromagnetic interaction of charged particles is described within a set of models called “standard electromagnetic physics” [3]. The binary cascade model [3] is used to describe the collisions of energetic nuclei with protons and nuclei from the phantom material. Following the cascade stage of interaction, the decay of excited nuclear remnants is described within the statistical evaporation model or within the Fermi break-up model [4], depending on the excitation energy of the nuclear system.

Model predictions are compared with available experimental data for water and PMMA phantoms irradiated by ^{12}C, ^{16}O and ^{14}N beams. The MCHIT model describes well (1) the depth-dose distributions in water and PMMA, (2) the yields of positron emitting nuclear fragments produced in fragmentation of projectile and target nuclei, (3) the attenuation of primary beam due to nuclear fragmentation reactions. There is still room for model improvement with respect to the description of specific fragment yields at different depths and angles.

The MCHIT capability of calculating three-dimensional dose distributions is demonstrated. Such doses obtained separately for each kind of secondary fragments provide input for evaluation of biological dose distributions, as the biological effectiveness of heavy ions essentially depends on their charge, mass and energy.

References

[1] I. Pshenichnov, I. Mishustin, and W. Greiner, Phys. Med. Biol. **50**, 5493 (2005)

[2] I. Pshenichnov, I. Mishustin, and W. Greiner, Phys. Med. Biol. **51**, 6099 (2006)

[3] S. Agostinelli *et al.*, (GEANT4 Collaboration) Nucl. Instrum. Meth. A **506**, 250 (2003)

[4] J. P. Bondorf, A. S. Botvina, A. S. Iljinov, I. N. Mishustin and K. Sneppen, Phys. Rept. **257**, 133 (1995)

Mo-IV-2

PHYSICAL BASES OF HEAVY-ION CANCER THERAPY

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When a charged projectile enters a medium it loses its energy in inelastic processes such as excitation of host atoms and molecules and their ionization resulting in a shower of secondary electrons. In living matter the projectile and the secondary electrons interact with the DNA molecules which may lead to damages beyond repair capabilities of cellular machinery.

We present here a review of state-of-the-art in investigation of the general mechanisms leading to the DNA damage and consequent cell death caused by exposure of tissue to a heavy ion beam. To increase efficiency and reduce side effects of the heavy ion cancer therapy one needs to be able to predict very accurately the distribution of the damages in the tissue and the minimum effective dose.

One of the most difficult questions in this study concerns the molecular mechanisms of DNA damage. Does a single ionization destroy the DNA molecule or a multiple ionization is needed? What are the cross sections of interaction of DNA molecules with charged par-ticles, especially at low energies? What are the mean paths of electrons in the tissue-like medium and which particles are most efficient in producing non-repairable DNA dam-ages? Some estimates can be made on this account based on recent calculations that rely on the number density of electrons in the vicinity of the DNA and their energy spectrum [1,2].

The depth of penetration of projectiles depends on the medium and properties of the projectile such as mass, energy, and charge. The rate of energy loss by the projectile typically increases as its energy decreases and the cross section of interaction increases corre-spondingly; thus, the most of energy is lost on the last stretch of the trajectory. This gives rise to the so-called Bragg peak in dependence of energy losses on the depth of penetra-tion. The Bragg peak's position is a very important quantity to be predicted, especially for the deeply seated tumors in brain. Therefore, one needs to address the problem of choos-ing the properties and characteristics of projectiles so that the desired location in the tu-mor is the subject of maximum radiation damage.

REFERENCES
[1] I. Pshenichnov, I. Mishustin, W. Greiner, *Phys. Med. Biol.* **50**, 5493 (2005)
[2] I. Pshenichnov, I. Mishustin, W. Greiner, *Phys. Med. Biol.* **51**, 6099 (2006)

Mo-IV-3

Organization of transmembrane molecules in immunological synapses

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The immunological synapse is a dynamical structure that emerges from cell-cell-interactions during immune responses. The mechanisms underlying this structure are related to the reorganisation of transmembrane molecules into specific patterns which are often called bull's eye pattern. The type of molecules involved are adhesion molecules and antigen-presenting molecules (pMHC) and their receptors (TCR). The different length of the molecules that mediate the cellular contact leads to short-range repulsive forces. At the same time the cytoskeleton induces long-range attractive forces. The organisation is overlayed by ongoing diffusion of molecules as well as binding and unbinding of receptor and ligand pairs following chemical reaction kinetics.

The formation of immunological synapses is modeled with the help of an agent-based approach using hybrid cellular automata [1]. It is possible with such a technique to disentangle the underlying mechanisms that drive the specific bull's eyes pattern and to pin down which mechanisms are necessary for the formation. Also it can be investigated how the synapse pattern may be actively changed. As the formation of immunological synapses are considered to be an essential step towards lymphocyte activation, thus, mediating immune activity, the control of a proper formation of synapses and the possible disregulations are likely to act as suitable targets for drug design, with the aim to regulate and control immune responses.

In the talk an introduction to the immune system with special attention to the immunological synapse will be given. The concept of agent-based modelling is explained and the approach is justified with respect to alternative methods used in theoretical Biology. The method is illustrated for the example of macro-molecular organisation and dynamics as realised in immunological synapses. Experimental constraints will be used to interprete the results of the agent-based simulation.

References

[1] M.T. Figge, M. Meyer-Hermann, 2006: Geometrically repatterned immunological synapses uncover formation mechanisms PLoS Comput. Biol. 2 (11) e171

ON MODELLING OF GENE EXPRESSION PATTERNS IN DROSOPHILA EMBRYO

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In general, biological systems possess spatial structure at all scales, from organs down to the molecular level that complicates construction of theoretical models of spatially controlled development. It is in cogent contrast with classical reaction-diffusion (RD) model, having a large gap in scale between the molecular mean free path and the scale of structures formed due to the balance between reaction and diffusion. It is thus unclear if continuous non-linear RD equations can be used in a biological system, because borders within and between cells by membranes and other structures invalidate the physics on which the classical RD theory is based. Moreover, the RD equation has an unrealistic feature: motion has an infinite velocity.

It is common knowledge in developmental biology that pattern formation implies a necessity of cell (or nuclear) division, and we decided to examine it. This basic question is refractory to experimental analysis because the nuclear structure of the embryo cannot be disrupted in the living state. We study the relationship between pattern formation in Drosophila segmentation and nuclear structure by replacing the nuclei by a homogeneous continuum, that cannot be performed experimentally. Mathematical simulation is performed for a model based on partial differential equations, see [1], [2] for details. Parameter values are found for continuum models by means of the simulated annealing method supplied with a new optimal steepest descent algorithm, [3]. Each of these models contains a different mathematical representation of nuclear divisions (mitoses), see [4].

We obtained correct pattern dynamics from all of them, as well as from the model with explicit nuclear structure. This allows to conclude that nuclear divisions are not coupled to pattern formation and serve only to populate the blastoderm with nuclei. The implications of our results for models of biological pattern formation based on partial differential equations are discussed.

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References

[1] Gursky et al. , Physica D, **197**, 3-4, 286-302 (2004).

[2] Gursky et al. , Physica D, **218,** 1, 70–76 (2006).

[3] K.N.Kozlov, A.M.Samsonov, Techn.Physics J., **48**, 11, 1364-1371. (2003).

[4] M.G.Samsonova et al., In: “ Multiple aspects of DNA and RNA: from biophysics to bioinformatics”, Chatenay, Cocco, Monasson, Thieffry, Dalibard eds., Elsevier, 2005, pp. 305-323. (2005)

Posters

PS-01

INFLUENCE OF ALKALI METAL IONS ON INTERNAL DYNAMICS OF DNA

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Unger natural conditions the negatively charged phosphate groups of the DNA backbone are neutralized by alkali metal counterions. Interacted DNA phosphate groups and counterions form a regular structure that in present work is considered as ion lattice. The dynamics of ion-phosphate lattice is studied to determine the mode of ion-phosphate vibrations, and counterion influence on DNA internal vibrations. To determine the frequency of the DNA ion-phosphate vibrations the constant of ion-phosphate interaction was estimated in framework of ion crystal theory [1]. Long-range electrostatic interactions are considered by the Madelung constant of the DNA ion-phosphate lattice. The environment is taken into account by the dielectric constant considering electronic and ionic polarizability mechanisms of DNA ion-hydrate shell.

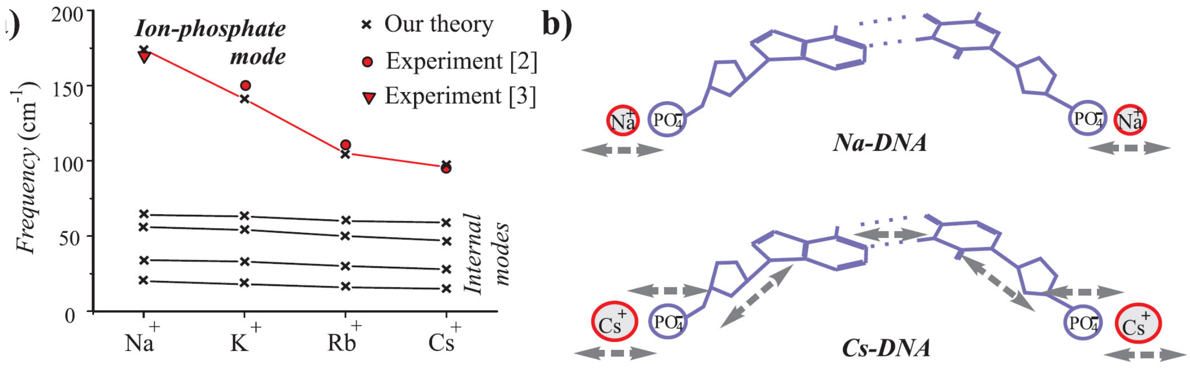

Figure 1: a) Dependence of the DNA low-frequency vibrations on counterion type. b) Displacements of Na- and Cs-DNA atomic groups on the mode of ion-phosphate vibrations.

As a result of calculations the frequencies and amplitudes of vibrations for DNA with metal cations Na$^+$, K$^+$, Rb$^+$ and Cs$^+$ were obtained. It was shown that the frequency of DNA ion-phosphate vibrations decreases from 170 to 90 cm^{-1} as counterion mass increases (Fig. 1a). The calculated frequencies agree well with the experemental data [2, 3]. The amplitudes of vibrations show that light counterions (Na$^+$) not disturb internal dynamics of DNA, but heavy counterions (Cs$^+$) make move all structure elements of the double helix (Fig.1b). The founded difference in the dynamics of double helix with light and heavy counterions may explain the negative effect of heavy metal cations on the DNA biological functioning.

References

[1] S.M. Perepelytsya, S.N. Volkov, Ukr. J. Phys. **49**, 1072 (2004).
[2] J.W. Powell *et al.* Phys. Rev. A **35**, 3929 (1987).
[3] T. Weidlich *et al.* Biopolymers **30**, 477 (1990).

PS-02

TO THE UNDERSTANDING OF PHASE DIAGRAM OF DNA DOUBLE HELIX UNZIPPING

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The transition of DNA macromolecule from the double-stranded (ds) state to the single-stranded (ss) configuration occurs under a temperature increase, change of ion concentration in solution, interaction with proteins or action of some external forces. Under such processes the H-bonds in complementary pairs of dsDNA break out and the nucleic bases become open. The opening of the pairs in the double helix is the cooperative process that induces the separation of two DNA strands. In the native conditions the DNA opening is a key element of important biological processes, such as DNA transcription and replication.

During the last years there appeared a number of new experimental techniques for probing single-molecule micromechanics which have been used for studying the process of DNA double helix opening. Thus, there was developed a special system for the investigation of mechanical separation of the DNA strands (unzipping) [1]. The unzipping of the DNA helix occurs under the action of external force (optical or magnetic tweezers) applied to one of the strands of the double helix. Recently, this experiment was supplemented with measurements of the temperature dependence of DNA unzipping within the temperature interval from 15 up to 50°C [2]. The results obtained were presented as the force-temperature phase diagram of DNA unzipping. The experiment shows that the base pair opening by external force depends on temperature in a nontrivial manner [2]. The unzipping force decreases steadily with the temperature growth only within the range of temperatures 24-35°C. Beyond this range, i.e. at 18-24°C and at 35-40°C, the force-temperature diagram of DNA unzipping possesses the plateaus. In these temperature ranges the unzipping force drops without significant temperature rise [2]. The observed plateaus cannot be explained within the frame of traditional thermodynamic approach [2], and are not seen in the Monte-Carlo simulations based on a simple DNA model [3].

Present contribution gives a new understanding of the mechanism of DNA unzipping taking into account possible conformational changes in dsDNA and formation of metastable states of base pairs under temperature increase.

The DNA unzipping is considered as a process of probing of dsDNA state by applied force at definite temperature. It is taken into account that the DNA double helix is relatively soft structure with a set of metastable states, which are populated under temperature increase. Using the normal mode analysis of dsDNA dynamics in the frame of 4-mass model approach [4] and considering the possible pathways of DNA unzipping, a new model for base pair stretching by external force in dsDNA is suggested. The suggested model is close to the Peyrard-Bishop model for DNA melting [5], but it also accounts for the disposition of base pairs in the DNA helix and for the dependence of the model parameters on pathways of base pair opening. The constructed model supports the common view that the observed process of DNA unzipping is the first order phase transition induced by applied force. However, at some stages this process may have a different character. The developed model leads to linear force-temperature dependence of unzipping at the temperature interval 24-35^{o}C as described in Refs. [2,3]. In the frame of the model an explanation of the plateaus appearance on the force-temperature diagram of DNA unzipping can also be done.

It is taken into account that the dsDNA macromolecule does not obtain any significant change in its configuration of complementary pairs in the double helix within the temperature range 15-24^{o}C. For this temperature range, it is known that the effect of double helix unwinding under temperature increase takes place [6,7]. This effect may lead to appearance of torque for dsDNA. As mentioned in [8], the torque changes an effective unzipping force during the construction of the experimental system for DNA unzipping. Taking into account the necessity of pair rotation before stretching by external force, as well as the DNA unwinding and unwrapping, the torque is included in the effective unzipping force. It is shown that accounting for the DNA torque in unzipping force is sufficient for understanding the plateau existing below 24^{o}C.

A mechanism of DNA unzipping at the range above 35^{o}C is treated as two-stage process. On the first stage, due to temperature increase the closed base pairs in dsDNA transmit to preopened state. This state of base pairs is known as intermediate between close and open states. It is characterized by partial destruction of hydrogen bonds and by insertion of a water molecule between the bases in the pair [9,10]. The considered stage of DNA unzipping has all features of the second order phase transition induced by temperature. Really, the analysis of the normal modes of 4-mass model shows the possible instability of the lowest optical type phonon mode (near 20 cm^{-1}) under temperature increase. The transition of base pair in the preopened state occurs as the displacement of the pair center of mass with the order parameter of about 0.5Å. As shown, the corresponding transition barrier has a low value along the pathway of pair “opening”. The second stage of DNA unzipping at the temperatures above 35^{o}C takes place as the phase transition induced by force along the pathway of pair “stretching”. On this stage, the base pair unzipping happens from the preopen state, and thus it requires a smaller force. It is taken into account also that the base pair transition along “opening” pathway involves the double helix deformation (bending and unwinding), which also leads to the unzipping force drop. The effect of DNA preopening explains the plateau appearance on force-temperature phase diagram of DNA unzipping at 35^{o}C.

REFERENCES

[1] U. Bockelmann, Ph. Thomen, B. Essevaz-Roulet, V. Visnoff, F. Heslot, *Biophys. J.* **82**, 1537 (2002)
[2] C. Danilowicz, Y. Kafri, R.S. Conroy, V.W. Coljee, J. Weeks, M. Prentiss, *Phys. Rev. Lett*. **93**, 078101 (2004)
[3] N.K. Voulgarakis, A. Redondo, A.R. Bishop, K.Ø. Rasmussen, *Phys. Rev. Lett*. **96**, 248101 (2006)
[4] S.N. Volkov, A.M. Kosevich, *J. Biomolec. Struct. Dynamics* **8**, 1069 (1991)
[5] M. Peyrard, A.R. Bishop, *Phys. Rev. Lett.* **62**, 2755 (1989)
[6] R.B. Genis, C.R. Cantor, *J. Mol. Biol.* **69**, 381 (1973)
[7] A.K. Schyolkina, L.E. Minchenkova, V.I. Ivanov, *Mol. Biol. (Moscow)* **11**, 466 (1977)
[8] S. Cocco, R. Monasson, J.F. Marko, *Phys. Rev.* E **65**, 041907 (2002)
[9] E.S. Kryachko, S.N. Volkov, *Inter. J. Quant. Chem.* **82**, 193 (2001)
[10] E. Giudice, P. Várnai, R. Lavery, *Nucleic Acids Research* **31**, 1434 (2003)

PS-03

MODELLING OF STIMULATED DEPOLYMERISATION AND AUTOREPAIRING OF DNA FRAGMENTS AT SEPARATED AND COMBINED ACTION OF IONIZING IRRADIATION

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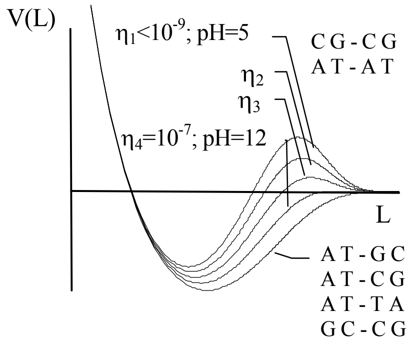
The creation of double breaks and depolymerisation of complementary stands of DNA macromolecules are the most harmful of all actions of ionizing irradiation and free radicals to living organisms. The investigations have been aimed at determining the conditions under which double breakages of DNA chains (owing to detrimental action of irradiation) become self-eliminable. Strict electrodynamic considerations suggest that the energy V(L) of electromagnetic interaction between the two end-pairs of DNA nucleotides on the opposite sites of a double break are totally under the control of: 1) the width of the break L; 2) Coloumb electromagnetic interaction $V(r_{ni,mj})$ of charges, distributed on a anisotropic surface of these nucleotides; 3) dispersion features of dielectric permeabilities $\varepsilon_i(\omega)$ and $\varepsilon_j(\omega)$ of the nucleotides (**A**denine, **T**hymine, **G**uanine**, C**ytosine) and inter-molecular salt-aqueous medium $\varepsilon_3(\omega)$ through all the range of frequencies ω (Kasimier-like force) [1]. The formation of hydrated electrons and heavy ions influences the energy of interaction in two main ways — changing the frequency structure of dispersion features of dielectric permeability of intermolecular salt-aqueous medium $\varepsilon_3(\omega)$ and changing the Coloumb interaction between nucleotides. This may result in a sharp change of the equilibrium of the forces of interaction between the nucleotides pair on the opposite sides of a DNA breakage.



It has been for the first time established that at the low concentration η of hydrated electrons (see Fig. at $\eta \leq 10^{-9}$) or at high acidity pH of intermolecular salt-aqueous medium (see Fig. at pH=5) the energy of the mutual interaction of the some end-pairs of nucleotides (AT-AT, CG-CG) has a repelling barrier with value $V(L_0,\eta) \approx (1\text{-}3)$ kT at $L_0 \approx 7\text{-}8$ A and corresponds to attraction between them at $L<L_0$. At $\eta \geq 10^{-7}$ or at pH=12 the barrier disappears. All other transversal end-pairs of DNA nucleotides experience only attraction [1].

The time-dependent dynamics of the process of depolymerisation, autorepairing, degradation and radiation curing of DNA at combined action of two kinds J_1 and J_2 of ionizing radiation and free radicals action is described by equations

$$dn/dt = b_1J_1 + b_2J_2 + W - n/\tau_{rel}; \quad dn_1/dt = n/T_1.$$

Here: n and n_1 are concentrations of reversible and irreversible DNA double breaks; $1/\tau_{rel} \equiv (1/\tau_d)\exp\{-V_0(1-c_1J_1- c_2J_2)/KT\}+1/T_1$ is the total probability of radiation DNA double breaks relaxation at temperature T; τ_d and T_1 are the times of reversible and irreversible double breaks relaxation; W is the probability of non-radiation (e.g. by free radicals action) double breaks generation; $b_{1,2}$ and $c_{1,2}$ are parameters, that describing interaction of ionizing radiation J_1 and J_2 with both DNA and intercellular salt-aqueous medium. The total concentration of DNA double breaks equals:

$$N = n(t) + n_1(t) = (b_1J_1 + b_2J_2 + W)\, \tau_{rel}\{1-\exp(-t/\tau_{rel}) +t/T_1 - (\tau_{rel} /T_1)\, [1-\exp(-t/\tau_{rel})]\}$$

There are some main results of this consideration.

1). For the case of short-time irradiation we have the usual linear (additive) radiation effect: the number of double breaks is proportional to radiation doses and total action of free radicals.

2). For the case of long-time action of weak intensity radiation we have the phenomenon of “hormesis” - decreasing of concentration of double breaks of DNA caused by free radicals action with grows of intensity of ionizing radiation.

3). For the case of combined action of two kinds of irradiation the phenomenon of radiation antagonism take place: the number of DNA double breaks caused by radiation J_1 is decreasing with growing the intensity of radiation J_2.

REFERENCES

[1]. Vysotskii V.I., Smirnov I.V., Kornilova A.A. Introduction to the Biophysics of Activated Water, Universal Publishiers, 2005

PS-04

MECHANISMS OF ANOMALOUS DNA DEFORMATIONS AT NANO SCALE

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Functioning of biological molecules depends mainly on regulatory DNA fragments of a nano scale length (A-tract, TATA-box, etc.). In many cases certain DNA fragments undergo anomalous large – from the viewpoint of the elastic theories - deformations of the double helix. For example TATA-box 2 nm long in the complex with proteins bends on roughly 80° and unwinds on 110°. Double stranded DNA (dsDNA) passing through nano pores undergoes overstretching transition, which have may be observed by means of laser tweezers direct manipulation of a single DNA molecule. Another example of dsDNA stretching (in approximately 1.5 times) occurs when DNA forms a complex with proteins molecule. Usually such deformations reveal the changes of the double helix conformation. Furthermore the experiments with high-resolution AFM [1] have been demonstrated that theory of DNA double strand deformation on nano scales should be more complicated than elastic one (for example model a worm like chain - WLC). Therefore this work dedicates theoretical investigation of mechanisms of anomalous deformability dsDNA.

We have advanced phenomenological approach in relevant nonlinear models [2,3] allowing studying the influence of the conformational transformations of DNA on the shape of the double helix chain (intrinsically induced mechanism of DNA deformation). In addition to the deformation like that of an elastic rod, those models make allowance for the internal conformational degree of freedom, as well as for the interrelation between its conformational transformations of the macromolecule and its deformation. For bistable DNA fragments (e.g. in heteronomouse conformation as it is for TATA-box) the nonlinear models with two and tree components of conformational transformations have been constructed. The strained fragment shape and the energy of macromolecule deformation have been found. It is argued, that the significant bending and (or) untwisting of short fragments of bistable dsDNA may be realized with energy-cost of thermal fluctuations order, on the assumption of realizing of definite boundary conditions (different conformational states) and of decreasing of DNA resistance in comparison with that predicted by the WLC. Confronting our theoretical results with experimental ones obtained for the anomalous deformation of the TATA-box DNA fragment testifies that the mechanism of the intrinsically induced deformation may explain the observed effects for a certain DNA fragments.

This work presents also the evolution of our approach of intrinsically induced deformation consideration for explaining DNA overstretching. In contrast to intrinsically induced bending and (or) untwisting deformations the case of intrinsically induced overstretching is realized under the influence of external force, which switch the system into bistable state of normal and overstretching conformations. It have been suggested that external force reduces the stretching rigidity of dsDNA molecule and at some critical point the macromolecule reach the bistable conformational state and can undergo kink-like conformational transition into overstretched state with energy-cost of thermal fluctuations order. The boundary conditions and the value of critical force for DNA B-form – overstretched transition are determined. We argue that the conformational transition can propagate along the molecule with some certain velocity value. The value-range for the propagation velocities given by the model concurs with the value-range for puling velocities of DNA molecule in the overstretching experiment.

PS-04

REFERENCES
[1] . P.A. Wiggins, T. Heijden, F.M. Herrero, A. Spakowitz, J. Widom, C. Dekker and P.C. Nelson, *Nature Nanotechnology* **1**(2), 137, (2006)
[2] S.N.Volkov, *J.Biol.Phys* **31**, N 3-4, 323, (2005)
[3]P.P.Kanevska, S.N. Volkov, *Ukr.J.Phys.* **51**, N10, 1003, (2006)

PS-05

THEORETICAL STUDIES OF STRUCTURE AND SPECTRAL PROPERTIES OF BIOLOGICAL CHROMOPHORES IN THE GAS PHASE, SOLUTION AND INSIDE THE PROTEIN

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A new effective Hamiltonian technique based on the multiconfigurational quasi-degenerate perturbation theory (MCQDPT2) [1] is introduced for modeling vertical excitation energies of large biological chromophores in different media. The use of augmented effective Hamiltonians enables to correct deficiencies of the standard state-specific and multistate approaches and to obtain the stable saturated solution for the target low-lying excited states. High accuracy of the new methodology (within 0.07 eV from the experimental absorption maxima) is illustrated for representative examples of photoactive protein chromophores in the gas phase. The method is also extended to the modeling of excitation energies of biological chromophores in solution and protein. The strategy used here has two major steps: (1) the ground-state geometry optimization with an explicit solvation model for solvents and combined quantum mechanical/molecular mechanical approach for proteins and (2) evaluation of transition energies of chromophores treated at the augmented MCQDPT2 level in the external electrostatic field originated from the distributed multipoles of surrounding media.

The methodology is applied to the modeling of photoabsorption of visual pigment Rhodopsin utilizing 11-cis retinal as a chromophore covalently bound to the protein via a protonated Schiff base linkage and fluorophores related to the ageing process in the human lens. The key questions addressed in the present theoretical study are the sensitivity of excitation energies to changes in the micro-environment of chromophores and factors contributing to solvent and protein shifts.

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References

[1] H. Nakano J. Chem. Phys. **99**, 7983 (1993)

PS-06

QUANTUM BUILDING BLOCKS CONCEPT FOR BIOMACROMOLECULES

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The idea of representing complex molecular structures as sets of building blocks is not new. It allows one to reduce drastically the volume of the conformational space, since fine details are integrated out producing renormalized effective interactions between the surviving collective degrees of freedom (the building blocks). The number of the degrees of freedom and accuracy of the effective interactions range widely depending on the system under consideration and on simplicity of the model. This strategy has led to significant breakthroughs in understanding structural properties of complex systems. For example, in the protein folding problem such phenomena as nucleation of the hydrophobic core or existence of a molten globule state can be explained with the building blocks concept [1]. The shortcoming of this approach is in the lack of predictive power for the *de novo* structure determination stemming from the excessive simplicity of the interaction potentials used.

We present here the formalism and the first results of quantum building blocks calculations in which the interaction potentials are calculated at an *ab initio* quantum mechanical level. We have performed density functional theory calculations of multidimensional potential energy surface for alanine-alanine interactions. In our approach the potential energy of the interaction depends on six variables, three of which describe the relative position and the other three describe the mutual orientation of the coordinate systems associated with each amino acid, see the figure. Using the quantum building blocks approach we found the global energy minimum structures of several Ala oligopeptides. The found structures are in a good agreement with our previous complete quantum mechanical analysis [2].

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REFERENCES

[1] A. Kolinski, J. Skolnick, *Polymer* **45**, 511 (2004)
[2] I. A. Solov'yov, A. V. Yakubovich, A. V. Solov'yov, W. Greiner, *Phys. Rev. E* **73**, 021916 (2006)

PS-07

COMPUTATIONAL MODELING STRUCTURE AND SPECTROSCOPIC PROPERTIES OF BIOGENE AMINES (SEROTONINE, HISTAMINE AND γ-AMINO OIL ACID) INCLUDING THE LASER AND NEUTRON CAPTURE EFFECTS

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Paper is devoted to the Monte-Carlo computational studying the structural, spectroscopic and biochemical properties for the biogene amines: the serotonine (ST), histamine (HM), γ-amino oil acid (AC) and the laser and neutron capture action on these peoperties for cited biomolecules. The ST (or 5-hydroxitriptamine, 5-HT) is produced by means of the hydroxciliration of essencial amine acid of the triptophane [1]. Under normal conditions an absolute majority of the triptophane (~90%) is used for the protein and nicotinamide synthesis and only a rest of the ST is used for synthesis.. Probably, ST is the mediator for transition of the nervous pulses in some branch of the brain. HM is produced in cells (mastocides) from the histidine amino acid and plays a role of the nervous transfer mediator in some regions of nervous system. The γ-amino oil acid AC is produced in the brain substance and probably plays a role of the mediator or inhibitor of pulses. Many biomolecules (BM) are composed not only by hydrophilic, but also by hydrophobic groups, in the vicinity of which the water-water (or blood plasma) interaction is expected to be present even in the zeroth approximation. We present results of the Monte-Carlo calculating the cluster consisting of the ST (HM) molecules and 100 molecules of water. All relevant interaction potentials are obtained by means of the quantum mechanical calculation [2-5]. The water-water interaction potential was found by Matsouka et al by the CI method [5]. The BM-water interaction potential was obtained in the SCF approximation Calculation is carried out at T=300K; All molecules are treated as rigid. The results for interaction energies are given below in table:

Potential, $kJmol^{-1}$	Neutral molecule	zwitterion
water-water	-27.7 ± 0.8	-27.2 ± 0.7
ST-water	-59.5 ±2.0	-348.5 ± 15.0
HM-water	-37,8 ±2.0	-178,4 ± 15.0

Analysis shows that the zwitterion appears as expected to be strongly favoured with respect to the neutral BM. This result is similar to one for system: glycine-water [5]. Analysis shows that the HM in the “zwitterion” more intensively (on the order) catalyses the gastric juice secretion and secretion from other endocrine glands. We at first consider the possibilities of the laser and neutron capture action on the structure and spectral properties of the clusters, including an analysis of the Szilard-Chalmers (n,γ), (n,n), Mössbauer and GM [3] effects. Some new features are discovered.

REFERENCES
[1] V. Schreiber , Pathophysiology of Endocrine Glands, Avicenum; Praga (1987); Theoretical and
[2] Computational Chemistry: Vol.8, J. Leszczynski (Ed.), Elsevier; Amsterdam (1999).
[3] A.V. Glushkov, Journ. Struct. Chem. **34** (1993) 3-10; **31** (1990) 9-15; 32 (1992) 11-16; Rus. J. Phys. Chem. **62** (1988) 100-106; **65** (1991) 2970-78; **66** (1992) 1259-76; **66** (1992) 589-596.
[4] A.V. Glushkov, S.V. Malinovskaya, Int. J. Quant. Chem. **99** (2004) 889-896; **99** (2004) 936-942; **104** (2005) 562-570; A.V. Glushkov et al, J.Phys.CS. **35** (2006) 420-425; Recent Advances in Theory of Phys. and Chem. Systems. Eds J.-P.Julien, J. Maruani (Springer), **15** (2006) 285-300.
[5] E. Clementi etal, Gazz.Chim.Ital. **108** (1978) 157-174; J. Am.Chem. Soc. **99** (1977) 5531-5542.

PS-08

Photoabsorption of biological chromophores in the gas phase: the case of Schiff-base retinal chromophore

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Photoactive proteins play crucial role in the spectral tuning of biological chromophores. Visual pigments, utilizing the retinal molecule as a chromophore, absorb light in the wide range of UV/Visible spectra: absorption maxima of cone pigments vary from 360 to 635 nm. The chromophore photoexcitation is the primary event of vision, followed by the light-induced isomerization of retinal and conformational changes of protein, eventually leading to the visual transduction. Photoabsorption spectra of isolated biological chromophores provide an important reference data for studying the influence of environment on the optical properties of chromophores.

We present here a multistate multiconfigurational perturbation theory study of vertical excitation energies of biochromophores in the gas phase. For this purpose we used recently developed aug-MCQDPT2 technique based on the extension of the multiconfigurational quasidegenerate perturbation theory (MCQDPT2) [1] . The suggested technique allowed us to attribute the main features of the observed S0-S1 gas phase absorption spectra [2] of the N-dimethyl-11-cis- and N-n-butyl-all-trans- retinal chromophores to the different conformers. It was shown that the farther red absorption maximum position (∼600 nm) could be ascribed to the 6-s-cis conformer and the higher energy shoulder (∼540 nm) of the S0-S1 absorption to the 6-s-trans conformer. Furthermore, on the basis of the aug-MCQDPT2 excitation energies estimations it was also demonstrated that retinal conformations arising from 6-s-, 8-s-, 12-s- cis/trans single bond configurations inside the polyene chain could account for the widening of the S0-S1 absorption peak in the 540–550 nm and 580–590 nm spectral range.

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References

[1] H. Nakano J. Chem. Phys. **99**, 7983 (1993)

[2] L.H. Andersen, I. B. Nielsen, M. B. Kristensen, M. O. A. El Ghazaly, S. Haacke, M. B. Nielsen, M. A. Petersen J. Am. Chem. Soc. **127**, 12347, (2005)

PS-09

PHOTOLYSIS OF BIOLOGICALLY RELEVANT PYRROLE MOLECULES IN CLUSTERS

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Study of pyrrole molecule is important for its biological relevance [1]. Pyrrole is a prototype for the heteroatomic molecules which form the molecular basis of life, i.e. nucleobases. Photochemical processes in nature, however, do not occur in the gas phase but in a confined environment. Study of the pyrrole photochemistry in molecular clusters can therefore be very relevant in this context for the study of photochemical stability of molecular building blocks of life.

The photolysis of pyrrole in clusters was investigated in a molecular beam experiment [2]. The clusters were produced by supersonic expansions. The neutral cluster size distributions under different expansion conditions were determined in a scattering experiment. Therefore clusters of different known size distributions could be studied by the photodissociation experiment. The kinetic energy distributions (KED) of H-fragments after photolysis have been measured by a time-of-flight spectrometer in low field mode. The clusters were photolysed with a 243 nm laser and the H-fragments were subsequently ionized by a (2+1) REMPI process within the same laser pulse. Alternatively the clusters were photolysed with a 193 nm laser followed by the 243 nm laser ionization.

The measured H-fragment KED exhibits a bimodal character observed previously for the pyrrole molecule [3–5]. A narrow maximum of *fast* H-fragments was assigned to a direct N-H bond fission in the excited $\pi\sigma^*$ state, while a less intense broad distribution of *slow* fragments corresponds to the internal conversion to the electronic ground state (S_0) followed by unimolecular decomposition of the vibrationally hot molecule [3–5]. Theoretically, the photodissociation dynamics of the molecule has been rationalized in terms of the predicted conical intersections between $\pi\pi^*$, $\pi\sigma^*$, and S_0 potential energy surfaces [6].

Figure 1 shows a comparison of the H-fragment KED from photolysis of a single pyrrole molecule (a), with the photolysis in a cluster of the mean cluster size $\bar{n} = 3$ (b), and $\bar{n} \geq 5$ (c). Clearly the significance of the *slow* channel increases with the cluster size. This dependence will be discussed and shown that the cluster environment effectively blocks the direct N-H bond fission channel.

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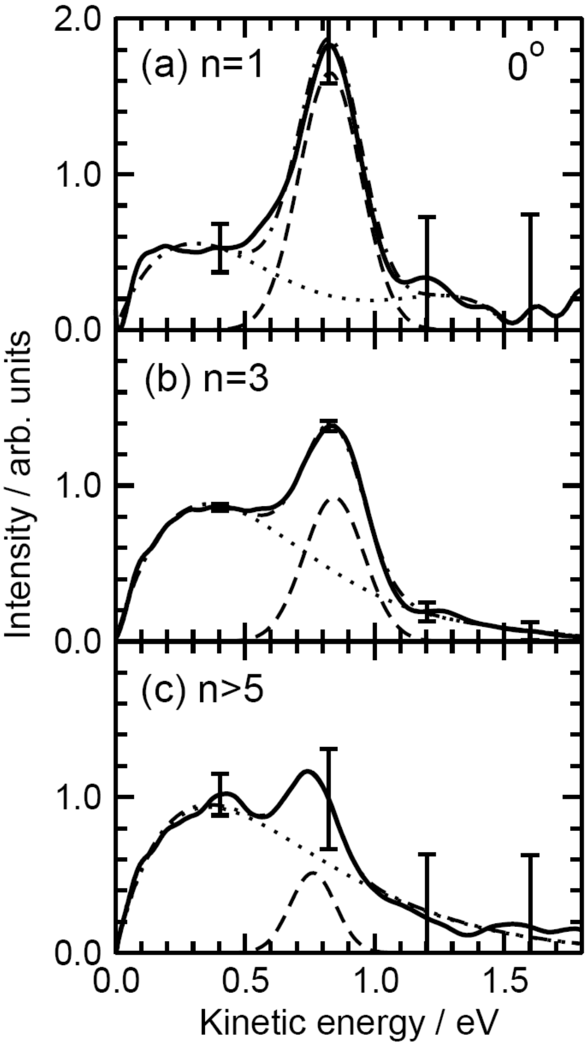

Figure 1: Comparison of H-fragment KEDs from photolysis of pyrrole molecule at 243 nm (a), clusters with $\bar{n} = 3$ (b), and clusters with $\bar{n} \geq 5$ (c).

References

[1] M.N.R. Ashfold, B. Cronin, A.L. Devine, R.N. Dixon, M.G.D. Nix, Science **312**, 1637 (2006).

[2] U. Buck, J. Phys. Chem. A **106**, 10049 (2002).

[3] H. Lippert, H.-H. Ritze, I.V. Hertel, W. Radloff, Chem. Phys. Chem. **5**, 1423 (2004).

[4] B. Cronin, M.G.D. Nix, R.H. Qadiri, M.N.R. Ashfold, Phys. Chem. Chem. Phys. **6**, 5031 (2004).

[5] J. Wei, J. Riedel, A. Kuczmann, F. Renth, F. Temps, Faraday Discuss. **127**, 267 (2004).

[6] M. Barbatti, M. Vazdar, A.J.A. Aquino, M. Eckert-Maksić, H. Lischka, J. Chem. Phys. **125**, 164323 (2006).

PS-10

PHOTOLYSIS OF MOLECULES IN CLUSTERS: FROM RARE GAS CLUSTERS TO WATER NANOPARTICLES

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The challenges in studies of photolysis in condensed media stems from the many-body nature of the interactions and from the solvent influence on the unimolecular fragmentation mechanism [1]. Studies of such processes in the finite cluster environments can provide a detailed insight on the molecular level [2]. Besides the fundamental scientific interest, photodissociation of molecules on clusters is of practical importance in atmospheric chemistry. By the way of example, photolysis of hydrogen halide molecules on ice nanoparticles in polar stratospheric clouds can play a key role in the ozone depletion process [3]

The photolysis of molecules in clusters was investigated in a molecular beam experiment [2], which has recently been moved from the Max-Planck Institute in Göttingen to the J. Heyrovský Institute of Physical Chemistry in Prague. The clusters of various sizes were produced in supersonic expansions. The clusters could be doped with foreign molecules in a pick-up cell. Then the cluster beam entered a vacuum chamber hosting a time-of-flight (TOF) spectrometer. Here the molecules were photolysed with a 193 nm laser and the H-fragments were subsequently ionized by a (2+1) REMPI process with 243 nm laser. Alternatively the molecules could be photolysed and the fragments ionized within the same laser pulse of 243 nm wavelength. The TOF spectra of the H-fragment atoms measured in the so called "low field mode" of the TOF spectrometer provided detailed information about the photodissociation process.

By the way of example, the photolysis of hydrogen halide (HX) molecules in various cluster environments will be presented in this contribution. Rare gas clusters with HX molecules weakly bound by the van der Waals interactions will serve as model systems. On their example the experimental procedure will be illustrated, and at the same time interesting phenomena will be shown such as *cage exit* and *caging* of the H-fragments after photolysis in the cluster. Further examples, e.g. photolysis of molecules in the hydrogen bonded networks of $(HX)_n$ clusters will be discussed. Finally, a very recent study [4] of a system of real atmospheric relevance will be presented: photolysis of HX molecules on large water clusters.

In the later system real chemistry including acid dissociation can occur. Our experimental evidence suggests, that the the acid dissociation leads to generation of a rather immobile contact ion pair $X^- \cdots H_3O^+$, which is then optically excited, leading to the neutral hydronium H_3O molecule. This molecule can relax into its ground state and subsequently dissociate to H_2O+H. The H-fragment of this decay is detected in our experiment. An alternative, though less probable, reaction channel starts with a UV excitation of an intact HX molecule leading again to the hydronium. Both reaction channels, depicted in Figure 1, will be discussed in detail and conclusions will be drawn in this contribution.

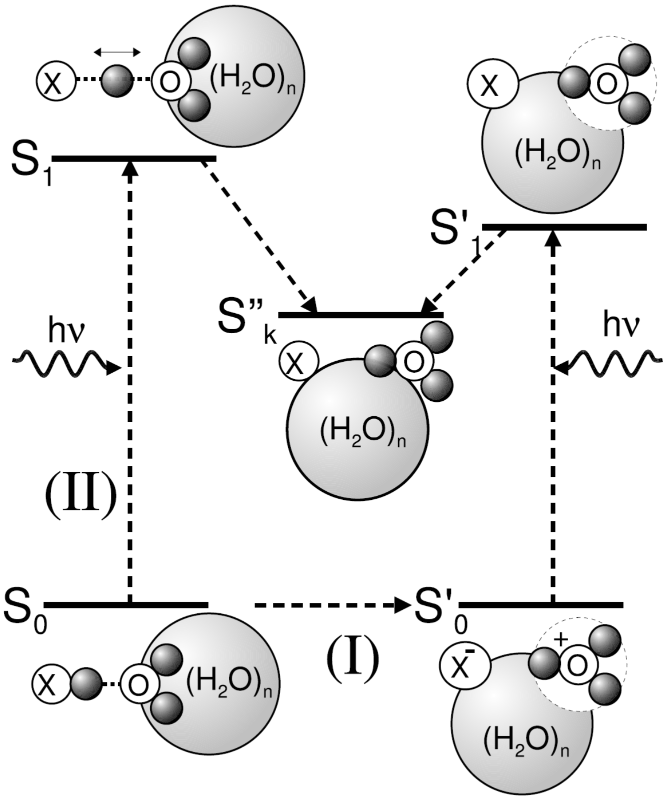

Figure 1: Schematic picture of $HX(H_2O)_n$ clusters photochemistry.

Acnowledgement This work has been partially supported by the program "Nanotechnology for society" of the Czech Academy of Sciences via grant Nr. KAN400400461 and by the Grant Agency of the Czech Republic via grant Nr. 203/06/1290. Theoretical interpretation by P. Slavíček is gratefully acknowledged.

References

[1] V.A. Apkarian, N. Schwentner, Chem. Rev. **99**, 1481 (1999).

[2] U. Buck, J. Phys. Chem. A **106**, 10049 (2002).

[3] T. Huthwelker, M. Ammann, T. Peter, Chem. Rev. **106**, 1375 (2006).

[4] V. Poterya, M. Fárník, P. Slavíček, U. Buck, V. Kresin, J. Chem. Phys. **126**, 071101 (2007).

PS-11

MID-INFRARED SEMICONDUCTOR WGM LASERS

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Whispering gallery modes (WGM) are universal linear excitations of circular and annular resonators. They have been first observed in 1910 in the form of sound wave travelling along the outer wall of a walk way in the circular dome of St. Paul's Cathedral in London and were investigated by Lord Raylegh. Referring to the acoustic phenomenon the name “whispering gallery modes” was also coined for electromagnetic eigenmodes of circular resonators. Dielectric rings, disks or spheres can be used as passive or optically pumped active resonators with integrated active media. This paper is devoted to the development of electrically pumped WGM semiconductor lasers for the mid-infrared waverange.

The spectral range of 3-5 μm is extremely important for applications based on molecular spectroscopy. This range corresponds to a window of atmospheric transparency and, on the other hand, it contains strong absorption lines of hazardous or explosive gases. Spectroscopic systems based on tunable semiconductor lasers can be used for high accuracy and selective monitoring of these substances and in trace concentrations.

At present, there are no semiconductor lasers exhibiting high performance in the 3-5 μm range near room temperature, which makes difficult the development of corresponding systems for gas analysis. There are several types of semiconductor lasers able to operate in this spectral region such as type I double heterostructure or quantum well lasers, type II “W” lasers, interband cascade lasers or quantum cascade lasers. For different reasons all these lasers exhibit low gain decreasing with temperature and, consequently, require low loss waveguides to achieve high temperature operation. The Q-factor of the micro-disk resonator can exceed the factor of 10^6. Thus, it is possible to obtain laser emission form the structures with relatively small optical gain. The use of the WGM resonators makes it possible to reduce lasing thresholds and consequently to increase operating temperatures of mid-infrared semiconductor lasers.

InAs based interband WGM lasers emitting in the 3-4 μm range have been reported by the Ioffe Institute and the Lancaster University. This investigation was awarded as the best work of the Ioffe Institute for 2005 year. These devices exhibit higher operation temperature than Fabry-Perot lasers fabricated from the same material but room temperature operation has not yet been achieved. Many particular tasks were solved:

- The mathematical modeling of the processes in the active region was performed including the processes of the radiative and non–radiative recombination, current distribution, optical gain-loss and optical mode distribution in the disk resonator for particular resonator geometries;
- MBE and MOVPE growth of above mentioned heterostructures for WGM lasers was developed;
- Post-growth processing was developed for all types of active media;
- Ring laser structures with various diameter of the active area, and with different deformation degree, was fabricated and studied;
- Extraction of output emission from WGM resonators was studied;
- Optimized design of mid-IR WGM lasers was proposed;

The physical processes due to the specifics of the WGM concept are to be studied, which will open the way for different applications. A distinctive feature of WGM devices lies in the fact that it is the optical modes are degenerate in frequency and waves propagate in opposite directions. This results in phase-sensitive mode interaction and the space grid produced by the joint action of both modes. This interaction leads to the complicated dynamics of the ring laser, which can reveal experimentally a number of effects. For example, the sensitivity of the laser spectrum to rotation or magnetic field can be used to fabricate the corresponding sensors. Also the WGM lasers are good candidates for laser spectroscopy, where the single frequency operation is more important than high output power.

Conclusion: We have obtained laser emission in the mid-IR at 3 mkm from a ring resonator DH diode fabricated by LPE. Coherent emission was observed at temperatures up to 125 K and was thought to originate from a whispering gallery mode. The results obtained earlier will be combined to realise room temperature mid-infrared WGM lasers.

We wish to thanks Russian Foundation for Basic Research and FASIE for support of this work.

PS-12

QED THEORY OF LASER-ATOM (MOLECULE, CLUSTER) INTERACTION. GENERATION OF ULTRA-SHORT VUV AND X-RAY PULSES IN CLUSTERS IN A STRONG LASER FIELD

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1. Unified QED theory is developed for studying interaction of atoms (nuclei, molecules, atomic clusters) with an intense and superintense laser field and analysis of the multi-photon phenomena in atomis systems in a field. Method is based on a description of system in a field by the k- photon emission and absorption lines [1]. The lines are described by their QED moments of different orders, which are calculated within Gell-Mann & Low adiabatic formalism (energy approach). We have studied the cases of single-, multi-mode, coherent, stochastic laser pulse shape. Results of calculation for the multi-photon resonance and ionization profile in Na,Cs, Yb, Gd atoms are presented. New effect of giant Rydberg, autoionization resonances broadening in Yb, Gd in a weak electromagnetic field is discovered. Possibilities of realization of the same effects in similar molecular and cluster systems are considered. As example in fig.1 we present results of calculation for multi-photon resonance width for transition 6S-6F in Cs atom (wavelength 1059nm) in dependence on the laser intensity. We use the denotations: S- for single-mode Lorentz laser pulse; M_1, M_3, M_4- for multi-mode Gauss laser pulse respectively with line band 0.03cm^{-1}, 0.08cm^{-1}, 0.15cm^{-1}; M_2,M_5-for multi-mode soliton- type with line band 0.03 cm^{-1} and 0.15cm^{-1}; dots- experimental data by Lompre-Mainfrau-Manus-Marinier.

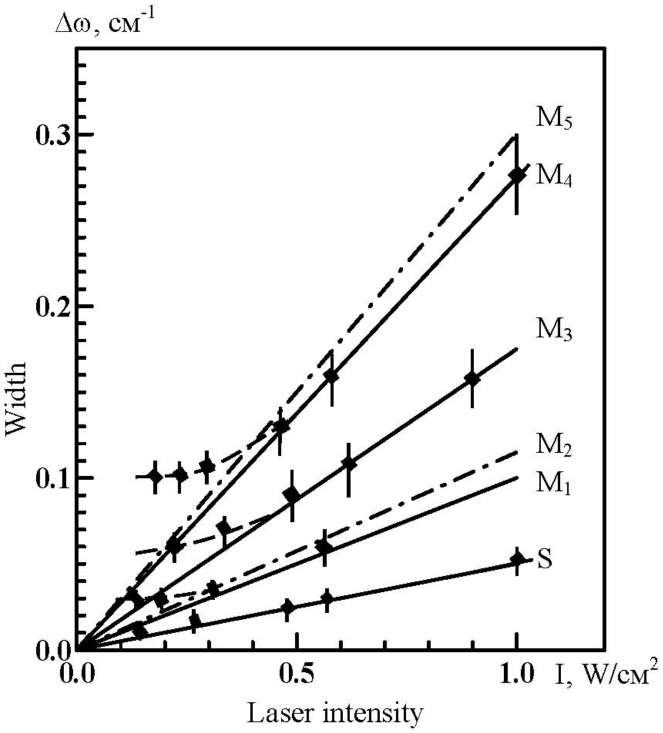

Figure 1: The multi-photon resonance width for transition 6S-6F in Cs atom (wavelength 1059nm) in dependence on the laser intensity (see text)

2. Further we study a behaviour of diatomic molecules and cluster systems in an intense laser field that is of a great interest in the last years [1-3].. We present the preliminary results of modeling and

predict an effective generation of the atto-second VUV and X-ray pulses under ionization of the hydrogen molecular ion and sodium cluster system in a strong laser field. For molecules we used the time dependent non-relativistic scheme of the complex rotation method and non-Hermitian Floquet formalism [3]. More effective (in comparison with standard scheme) method of the eigen-values definition and functions problem solution, which is based on the operator perturbation theory and Runge-Cutt integration procedure [1], is further used in the time-dependent problem. For all systems the final action includes the following steps: i). generalized complex scaling pseudospectral discretization of wave function and hamiltonian in the prolate spheroidal coordinates (methodics [3]); ii). construction and diagonalization of time evolution operator for one optical cycle, which yields the complex quasienergy eigenvalues and Floquet eigenstates; iii). time propagation of the selected Floquet states and evaluation of the time-dependent expectation values of dipole moment, momentum with further Fourier analysis of the above cited quantities to produce the harmonic generation rates. A consideration of the cluster system in a laser field is carried out within the relativistic scheme of the X∀- scattered waves method [4]. We present the results of modelling and first predicting the generation of the atto-second VUV and X-ray pulses under ionization of the cluster system N10 in a intense laser field. In ref. [3] the results of modelling the generation of the atto-second VUV and X-ray pulses under ionization of the molecular system 2D H2+ by femto-second optical pulse have been presented. We carried out the detailed comparison of the cluster Na10 response (our result), the molecular H2+ response for different internuclear distances 2.5, 3.5, 7.4, 16a.u. (our data and the coresponding data from refs.[3]) with smoothed Coulomb potential and atomic (H) response (spectral dependence) under ionization of the system by femto-second optical pulse. Our calculation shows that the generation of the atto-second pulses in cluster system is more effective and profitable (as minimum the 2-3 orders) than in similar molecular atomic one. Correspondingly, it has been shown earlier and confirmed by us that the generation of the atto-second pulses in molecular system is more profitable too (as minimum the 1-2 orders) than in similar atomic one [3]. It is demonstrated by comparison of the corresponding spectral dependences of the atomic (atom of H) and molecular responses. Last experimental achievements in field of generating high harmonics of optical radiation during atomic ionization by powerful femtosecond laser pulses demonstrate a possibility of construction of the compact sources of VUV radiation.

REFERENCES

[1] .V.Glushkov, L.N.Ivanov, *J.Phys.B.* **26**, L379 (1993); E.Ivanova, L.Ivanov, A.Glushkov, A.Kramida, *Phys.Scr.***32**,524 (1985); A.Glushkov,L.Ivanov, *Phys. Lett. A*,**170**, 33 (1992); L.Ivanov, E.Ivanova, L.V.Knight, *Phys.Rev.A* **48**, 4365 (1993); A. Glushkov et al, In: *New projects and new lines of research in nuclear physics.*Eds. G.Fazio, F.Hanappe, Singapore : World Sci., 126 (2003) ; *Nucl.Phys.A.* **734**, e21 (2004); *Int. J. Quant. Chem.* **99**, 889 (2004); **99**, 936 (2004); **104**, 496 (2005); **104**, 512 (2005); **104**,562 (2005); A.Glushkov et al, *J.Phys.CS.* **35**,420 (2006); **35**, 426 (2006); *Recent Advances in Theory of Phys. and Chem. Systems*. Eds J.-P.Julien, J. Maruani (Springer), **15**, 301 (2006); A.V.Glushkov, In: *Low Energy Antiproton Phys*., Eds.D.Grzonka, W.Oelert et al, AIP Serie.**796**, 206 (2005).

[2]. A.Jaron-Becker, A.Becker, F.H.M.Faisal, *Phys.Rev.A.* **69**, 023410 (2004); S.Fomichev, D.Zaretsky, D.Bauer, W.Becker, *Phys.Rev. A.* **71**, 013201 (2005);

[3]. M.Emelin, M.Ryabikin, A.Sergeev etal, *JETP Lett.* **77**, 254 (2003); D.Telnov, I.Chu Shih, *Phys.Rev A*.**71,** 013408 (2005).

[4] V.Gedasimov, A.Zelenkov, V.Kulakov, et al, *JETP.* **86** , 1169 (1984); A.Soldatov, Preprint of I.V.Kurchatov Institute for Atomic Energy IAE-3916, Moscow, (1983); N.March, *J.Mol.Str.* **300**, 479 (1993);

PS-13

SUPER INTENSE LASER FIELD ACTION ON SURFACE AND FORMING THE FEMTO-SECOND LASER PLASMA IN THE NANO-STRUCTURED POROUS CLUSTERS

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Using the super short light pulses changes principally a character of interaction of the laser radiation with substance (surface, cluster) [1] under laser radiation intensities more than 101515 Wt/cm2. During the laser pulse electrons get energy of 100-1000eV and it is realized a process of forming the femto-second laser plasma.(FLP) In this paper we consider possibilities of governing by processes, which are taken a place in the FLP in nano-structured porous materials (Si). Nano-structured porous materials consists of the separated clusters set, that forms the fractal structure or regular net. The main mechanism of the hot electrons generation in plasma is provided by oscillation of electron on the border "plasma-vacuum" or resonant absorption of laser radiation. For porous materials one may wait for the sharp increasing the hot electrons generation and X-ray radiation. Under large intensity of laser field it is observed a new type of the hydrodynamic ablation, which has the explosion character. Experimental estimates show that a velocity of the plasma flying of the strongly porous samples Si (I~1016 W/cm2) is ~108 cm/s, that is corresponding to energy ~2 MeV. We carried out the modelling of FLP forming in the porous materials on the basis of the energy balance equations and Green's function formalism for non-ordered materials [2,3]. Special attention is devoted to the modelling the system: nano-structured porous material with clusters, on surface of which there is a great number of bonds with H and OH groups. In a case of the D-and OD group's one can wait for realization of the cluster explosion process and reaction D+D-alpha+n (3,8MeV). In the high density plasma there is possible an excitation of the low lying isomers (level energy less 20 keV) by means of the following channels: photo excitation by own X-ray plasma radiation, the electron impact excitation, electron conversion etc. Atomic-nuclear numerical code [3] is then used for calculating the stable and long lived nuclei low lying isomers characteristics (time of life, spin, energy, decay channels etc.).

REFERENCES

[1]. Superstrong Fields in Plasmas. Eds. M.Lontano, AIP Proc. V.**426**, N.-Y. (1998); V.Gordienko, A. Saveliev, *Soviet Uspekhi-UFN* **169**, 78 (1999);

[2]. L.N.Ivanov, A.V.Glushkov, S..Malinovskaya, S.Ambrosov, *Preprint ISAN,* №AS-5, Moscow,1991; A.V.Glushkov, L.N.Ivanov, *J.Phys.B.* **26**, L379 (1993); E.Ivanova, L.Ivanov, A.Glushkov, A.Kramida, *Phys.Scr.***32**,524 (1985); A.Glushkov,L.Ivanov, *Phys. Lett. A*,**170**, 33 (1992); L.Ivanov, E.Ivanova, L.V.Knight, *Phys.Rev.A* **48**, 4365 (1993); A.V.Glushkov, E.P.Ivanova, *J.Quant.Spectr.Rad.Tr.(US)* **36**,127 (1986);

[3] A. Glushkov et al, In: *New projects and new lines of research in nuclear physics.*Eds. G.Fazio, F.Hanappe, Singapore : World Sci., 126 (2003) ; *Nucl.Phys.A.* **734**, e21 (2004); *Int. J. Quant. Chem.* **99**, 889 (2004); **99**, 936 (2004); **104**, 496 (2005); **104**, 512 (2005); **104**,562 (2005); A.Glushkov et al, *J.Phys.CS.* **35**,420 (2006); **35**, 426 (2006); *Recent Advances in Theory of Phys. and Chem. Systems*. Eds J.-P.Julien, J. Maruani (Springer), **15**, 301 (2006); A.V.Glushkov, In: *Low Energy Antiproton Phys.*, Eds.D.Grzonka, W.Oelert et al, AIP Serie.**796**, 206 (2005).

PS-14

AUGER SPECTROSCOPY OF CLUSTERS AND SOLIDS

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The Auger electron spectroscopy is an effective method to study the chemical composition of solid surfaces and near-surface layers [1]. When considering the method principles, the main attention is given as a rule to the models for drawing chemical information from the Auger spectra and to the surface composition determination methods by the Auger spectrum decoding. It is just the two-step model that is used most widely when calculating the Auger decay characteristics. Since the vacancy lifetime in an inner atomic shell is rather long (about 10^{-17} to 10^{-14}s), the atom ionization and the Auger emission are considered to be two independent processes. In the more correct dynamic theory of the Auger effect [1,2] the processes are not believed to be independent from one another.. In fact, a consistent Auger decay theory has to take into account correctly a number of correlation effects [1]. Here we use our approach to studying the Auger decay characteristics for complex atomic systems, clusters and solids, based on the S-matrix formalism by Gell-Mann and Low and QED perturbation theory (PT) [2]. Within the frame of QED PT approach, the Auger transition probability and, accordingly, the Auger line intensity are defined by the square of an electron interaction matrix element having the form:

$$V^{\omega}_{1234} = [(j_1)(j_2)(j_3)(j_4)]^{1/2} \sum_{\lambda\mu} (-1)^{\mu} \begin{pmatrix} j_1 j_3 & \lambda \\ m_1 - m_3 & \mu \end{pmatrix} \times \operatorname{Re} Q_{\lambda}(1234),$$

$$Q_{\lambda} = Q^{\text{Qul}}_{\lambda} + Q^{\text{Br}}_{\lambda}. \tag{1}$$

The terms Q^{Qul}_{λ} and Q^{Br}_{λ} correspond to subdivision of the potential into Coulomb part $\cos|\omega|r_{12}/r_{12}$ and Breat one, $\cos|\omega|r_{12}\boldsymbol{\alpha}_1\boldsymbol{\alpha}_2/r_{12}$. The real part of the electron interaction matrix element is determined using expansion in terms of Bessel functions: :

$$\frac{\cos|\omega|r_{12}}{r_{12}} = \frac{\pi}{2\sqrt{r_1 r_2}} \sum_{\lambda=0} (\lambda) J_{\lambda+1/2}(|\omega|r_<) J_{-\lambda-1/2}(|\omega|r_>) P_{\lambda}(\widehat{\cos \mathbf{r}_1\mathbf{r}_2}). \tag{2}$$

where J is the 1st order Bessel function, $(\lambda)=2\lambda+1$. The Coulomb part Q^{Qul}_{λ} is expressed in terms of radial integrals R_{λ}, angular coefficients S_{λ} [2]:

$$\operatorname{Re} Q^{\text{Qul}}_{\lambda} = \frac{1}{Z} \operatorname{Re}\{R_l(1243) S_{\lambda}(1243) + R_{\lambda}(\tilde{1}24\tilde{3}) S_{\lambda}(\tilde{1}24\tilde{3}) +$$
$$+ R_{\lambda}(1\tilde{2}\tilde{4}3) S_{\lambda}(1\tilde{2}\tilde{4}3) + R_{\lambda}(\tilde{1}\tilde{2}\tilde{4}\tilde{3}) S_{\lambda}(\tilde{1}\tilde{2}\tilde{4}\tilde{3})\}.$$

As a result, the decay probability is expressed in terms of $\operatorname{Re}Q_{\lambda}(1243)$ matrix elements:

$$\mathbf{Re}\, R_{\lambda}(1243) = \iint dr_1 r_1^2 r_2^2 f_1(r_1) f_3(r_1) f_2(r_2) f_4(r_2) Z^{(1)}_{\lambda}(r_<) Z^{(1)}_{\lambda}(r_>). \tag{4}$$

where f is the large component of radial part of single electron state Dirac function; function Z is: :

$$Z^{(1)}_{\lambda} = \left[\frac{2}{|\omega_{13}|\alpha Z}\right]^{\lambda+1/2} \frac{J_{\lambda+1/2}(\alpha|\omega_{13}|r)}{r^{\lambda}\Gamma(\lambda+3/2)}.$$

The Auger width is obtained from the adiabatic Gell-Mann and Low formula for the energy shift [2]. The basis of electron state functions was defined by the solution of Dirac equation (integrated numerically using the Runge-Cutt method). The energy of an electron formed due to a transition jkl is defined by the difference between energies of an atom with a hole at the j level and double-ionized atom at kl levels in the final state:

$$E_A(jkl,{}^{2S+1}L_J) = E_A^{+}(j) - E_A^{2+}(kl,{}^{2S+1}L_J) \tag{5}$$

To single out the above-mentioned correlation effects, the equation (5) can be presented as:

$$E_A(jkl,{}^{2S+1}L_J) = E(j) - E(k) - E(l) - \Delta(k,l;{}^{2S+1}L_J) \tag{6}$$

where the item Δ takes into account the dynamic correlation effects (relaxation due to hole screening with electrons etc.) To take these effects into account, the set of procedures elaborated in the atomic theory [1,2] is used. For solid phase, the more precise form of equation (6) is:

$$E^S{}_A(jkl,{}^{2S+1}L_J) = E_A(jkl,{}^{2S+1}L_J) + \Delta E^S + R_{rel} + e\Phi \tag{7}$$

where ΔE^S is a correction for the binding energy change in the solid; R_{rel}, the same for out-of-atom relaxation; $e\Phi$ takes into account the work of exit. As example, in table 1 we present the data on Auger electron energy for some solids calculated using our method (column B), the semi-empirical method under Larkins' equivalent core approximation [1] (column A) as well as experimental data. The calculation accuracy using the Larkins method is within about 2 eV as an average. Our approach provides more accurate results that is due to a considerable extent to more correct accounting for complex electron interaction.

Table 1.

Theoretical and experimental data for Auger electron energy for solids and calculated values (A, semi-empirical Larkins method [1]; B, present paper)

Element	Auger line	Experiment	Theory: A	Theory: B
Na	*KL2,3L2,3 1D2*	994,2	993,3	994,3
Si	*KL2,3L2,3 1D2*	1616,4	1614,0	1616,3
Ge	*L3M4,5M4,5 1G4*	1146,2	1147,2	1146,2
Ag	*M5N4,5N4,5 1G4*	353,4	358,8	353,5

To conclude, note that the use of the Auger electron spectroscopy in analysis of the surface chemical composition requires consideration of Auger spectra and the corresponding characteristics of the Auger transitions, interpretation of effects like the shape transformations of the valence Auger spectra due to appearance of new lines, position and intensity changes of individual lines caused by the redistribution in the electron state density of the valence band. The correct theoretical estimations of the spectral characteristics are of critical importance for their full understanding.

REFERENCES

[1]. T.Aberg, G.Hewat G. Theory of Auger effect. Springer-Verlag: Berlin (1979); M.Amusia, Atomic photoeffect. Acad.Press: N.-Y., 1988.

[2]. A.Glushkov,L.Ivanov, *Phys. Lett. A*,**170**, 33 (1992); L.Ivanov, E.Ivanova, L.V.Knight, *Phys.Rev.A* **48**, 4365 (1993); A. Glushkov et al, *Int. J. Quant. Chem.* **99**, 936 (2004); **104**, 512 (2005); A.Glushkov et al, *J.Phys.CS*. **178**, 188 (2005); **178**, 199 (2005).

PS-15

SEMI-SPHEROIDAL ATOMIC CLUSTER

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The liquid drop model (LDM) was for many years the main theory of the nuclear fission processes. Since the delocalized electrons of a metallic cluster may be considered to form a Fermi liquid like the atomic nucleus, it was also adapted to the atomic cluster physics [1]. The Strutinsky's macroscopic-microscopic approach [2] is well suited to consider both the phenomenological liquid drop properties and the quantum shell effects particularly manifested in the ground state.

As a first approximation to the complex problem of cluster deposition on planar surfaces, we assume a simple shape of semi-spheroidal, neutral, free cluster, for which it was possible to develop a new analytical single-particle shell model [3]. Different shapes and interaction energy with the surface have been considered elsewhere [4]. We use dimensionless (units of the semi-sphere radius $R_s = 2^{1/3} r_s N^{1/3}$) cylindrical coordinates ρ, z and semi-axes a, z. The Wigner-Seitz radius of Na is $r_s = 2.117$ Å [5]. Volume conservation leads to $a^2 c = 1$. The Clemenger deformation parameter δ is defined by $a = [(2-\delta)/(2+\delta)]^{2/3}$.

We derived analytical relationships for the surface and curvature deformation energies of spheroidal clusters. Then, by using the energy levels of the semi-spheroidal quantum harmonic oscillator [3], the shell corrections δU were calculated and added to the liquid drop energy, so that the total deformation energy is $E = E_{LD} + \delta U$, as shown in Fig. 1 for Na$_{56}$ cluster.

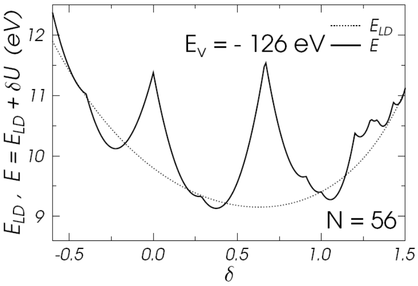

Fig. 1 Deformation energies (LD compared to LD plus shell corrections) for a semi-spheroidal Na$_{56}$ cluster.

The example illustrates the typical results we obtained by sudying the potential energy surfaces of deformation versus δ and the number of atoms in the cluster N. Unlike for spheroidal shapes for which the minimum of E_{LD} is always at $\delta = 0$, for semi-spheroids the minimum is at a superdeformed prolate shape, in this case $\delta = 0.63$. By adding the shell corrections the ground state minimum is shifted to the minimum of E, e.g. $\delta = 0.37$.

References

[1] W.A. Saunders, Phys. Rev. Lett. **64**, 3046 (1990).

[2] V.M. Strutinsky, Nucl. Phys. A **95**, 420 (1990).

[3] D. Poenaru, R.A. Gherghescu, A.V. Solov'yov, W. Greiner, this Symposium.

[4] V.V. Semenikhina, A.G. Lyalin, A.V. Solov'yov, W. Greiner, to be published.

[5] C. Yannouleas, U. Landman, Phys. Rev. B **51**, 1902 (1995).

PS-16

LIQUID DROP MODEL FOR SUPPORTED CLUSTERS

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We have performed a theoretical exploration of structure formation and stability of atomic clusters deposited on a surface. In order to describe energetics, stability and shape of an atomic cluster that interacts with a surface we adapt the simple Liquid Drop Model (LDM). In particular, we investigate the role of cluster – surface interaction in the stability and deformation of Ar clusters with number of particles N (ranging from 4 to 150 particles) deposited on a graphite surface (001). The proposed model takes into account the cluster–surface interaction as well as the cluster deformation due to the interaction with the surface. We proceed from the fact that the total energy of free clusters of noble gases can be predicted within LDM with high accuracy [1,2]. In the present work we elucidate the applicability of LDM to the description of properties of deposited clusters of noble gases. We obtain a good agreement between the results of LDM and the results of numerical simulations based on the dynamical search of the most stable isomers forming in the cluster growth process. The numerical simulations were performed with the use of cluster fusion algorithm (CFA) [1,2]. We demonstrate that LDM can be used for simple estimation of the shape, stability and energetics of deposited atomic clusters.

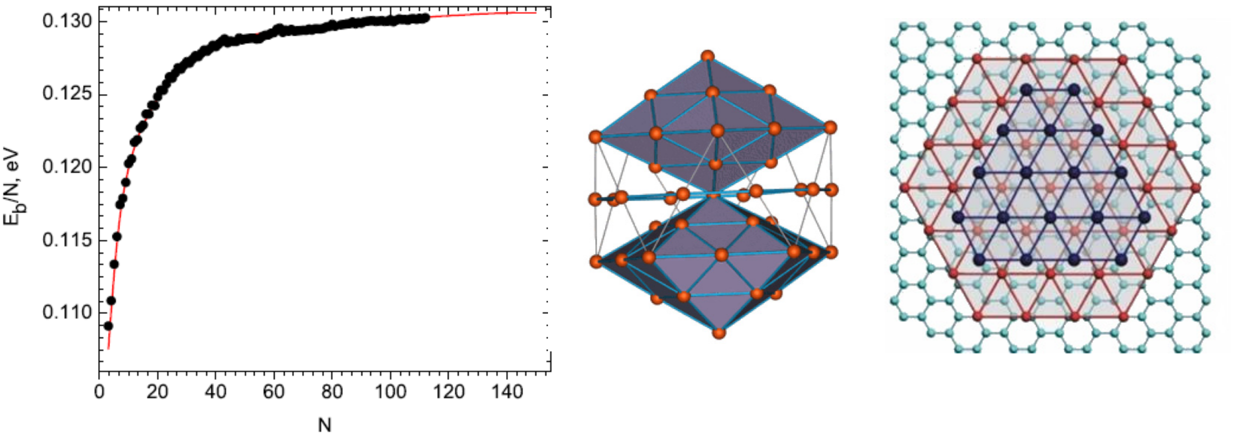

Figure 1: Left: binding energy per atom for Ar clusters deposited on a graphite surface (001) calculated within LDM (line) and CFA (dots); middle: free Ar_{55} cluster; right: Ar_{55} deposited on a graphite (001) surface.

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REFERENCES

[1] I.A.Solov'yov, A.V.Solov'yov, W.Greiner *et al.*, Phys. Rev. Lett. **90**, 053401 (2003).
[2] I.A.Solov'yov, A.V.Solov'yov, W.Greiner, Int. J. Mod. Phys. **13**, 697 (2004).
[3] V. Semenikhina, A. Lyalin, A. Solov'yov, and W. Greiner in preparation for JETP (2007).

PS-17

APPEARANCE SIZES OF MULTIPLY-CHARGED RARE-GAS CLUSTERS

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We report theoretical predictions on the appearance sizes of multiply-charged rare gas clusters based on the results of recent accurate calculations for the global minimum energies of Lenard-Jones clusters [1]. Comparisons are made with experimental results, including very recent ones by Maehr et al. [2] and a previous calculation [3]. We discuss also the role of geometrical shell effects on the stability of multiply-charged rare gas clusters.

A multiply-charged cluster is stable if its size is larger than the *appearance size*, which is obtained from the condition that an energy barrier exists for every fission channel. About 20 years ago, Echt et al. [3] calculated the appearance sizes for multiply-charged van der Waals clusters using the liquid drop model (LDM) with an assumption that the two fragment clusters are in contact at the transition state (*contact sphere model*). Their calculations have been in good agreement with the experimental measurements performed on many multiply-charged clusters. However, still some discrepancies were observed for some clusters such as those of Ar. Very recently, the appearance sizes of doubly- and triply- charged Ne clusters have been measured [2]. They are found to be much smaller than those estimated by [3]. In order to reveal these discrepancies, we have recalculated the appearance size of multiply-charged van der Waals clusters.

In the previous work [3], the parameters of the LDM were those given by Brian and Burton [4]. These parameters were determined through a molecular dynamics simulation for Ar clusters at the temperature $T = 40$ K. Recently, two groups [1] have calculated the energy of the global minimum of Lenard-Jones clusters up to size $n = 1000$. This situation corresponds to $T = 0$ K. Besides, we propose a new set of parameters for the LDM at $T = 0$ K which reproduce these recent calculations [3]. We calculate the appearance sizes using these global minimum energies and using the liquid drop energy at $T = 0$ K. The comparison of these two types of calculations, allows us to study the role of *geometrical shell effects* on the fragmentation process. Figure 1 shows the lowest energy barrier for the fission of Ar_n^{2+} as a function of n. The appearance size is given from the figure as the size at which the line (solid, dashed or dotted) crosses the horizontal axis. The LDM leads a smooth variation to the barrier energy as functions of n while shell oscillations are observed if the energies of [1] are used in the calculation.

We calculate the appearance sizes for multiply-charged rare gas clusters at $T = 0$ K. Our predictions are listed in Table I and are compared with available experimental data and with previous LDM ones obtained at $T = 40$ K. [3]. In general the appearance sizes at $T = 0$ K are smaller than those at $T = 40$ K since the surface energy at $T = 0$ K is approximatively 3/2 times larger than the one at $T = 40$ K. With respect to the results obtained in [3], the present calculation are in better agreement with the experimental measurements for Ar clusters. For Ne clusters, the discrepancy is diminished although it is still insufficient to explain the new experimental findings. In most cases, the LDM gives nearly the same appearance sizes as

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Rg	z	Experiment	Calculation using [1]	LDM (T = 0 K)	LDM (T = 40 K [3])
Ne	2	288	649	657	868
	3	656	—	2203	2950
Ar	2	91	97	94	122
	3	226	255	256	333
Kr	2	71	54	55	71
	3	156	134	137	177
Xe	2	46	40	36	51
	3	107	87	84	114
	4	208	139	153	196

Table 1: Appearance sizes $n_c(z)$ for $(\mathrm{Rg})_n^{z+}$, experimental measurements, calculation using global minimum energies and using the LDM at T= 0 K and 40 K [3].

those obtained from the global minimum energies. However, shell effects slightly modify the appearance sizes. Such a tendency has been also observed in the fission of multiply-charged clusters of fullerenes [5]. This work has been financially supported by Matsuo Foundation.

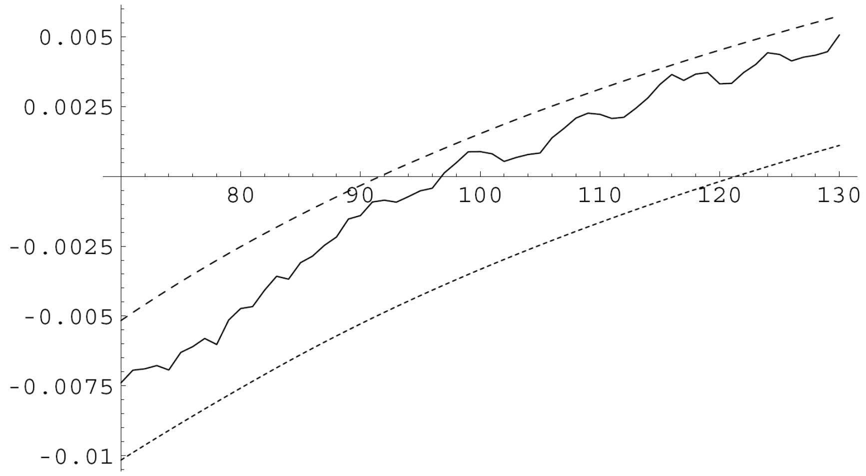

Figure 1: Fission barriers (in hartree) for Ar_n^{2+} as functions of parent size n. Solid line: model including shell corrections; dashed line: LDM at T = 0 K; dotted line: LDM at T = 40 K.

[1] D. Wales and J. Doye, J. Phys. Chem. A **101**, 5111 (1997); Y. Xiang et al., J. Phys. Chem. A, **108**, 3586 (2004), *ibid* **108** , 9516 (2004)

[2] I. Maehr et al., Phys. Rev. Lett. **98**, 023401 (2007).

[3] O. Echt et al., Phys. Rev. A **38**, 3236 (1988).

[4] C. L. Briant and J. J. Burton, J. Chem. Phys. **63**, 2045 (1975).

[5] M. Nakamura and P -A. Hervieux, Chem. Phys. Lett. **428**, 138 (2006).

PS-18

STRUCTURE OF CORONENE CLUSTERS

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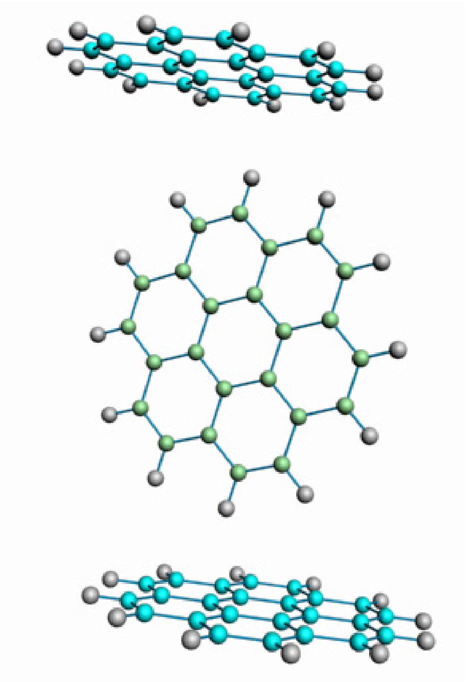
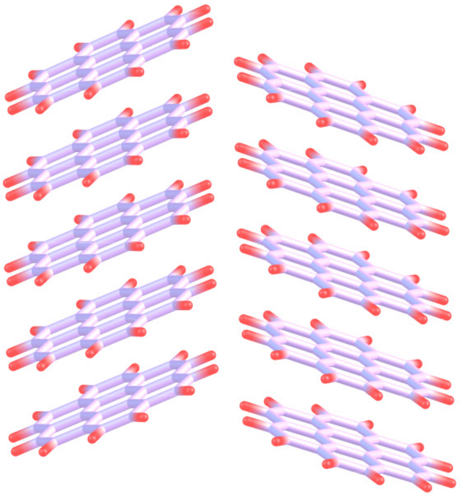
Clusters of polycyclic aromatic hydrocarbons (PAH) and their ions are believed to be one of the most probable species in the interstellar space responsible for converting the UV radiation of the stars into the intensive IR radiation, known as unidentified infrared bands [1,2]. The current knowledge of structure and properties of PAH clusters is very limited. Laboratory studies have just started to appear while reliability of the theoretical results is limited due to the subtle interplay between the van der Waals polarization forces and the electrostatic interaction [3].



Using the methods of density functional theory (DFT) we have demonstrated [3] that there are two energetically competitive geometries of the coronene dimer. These are a T-shaped configuration and a parallel-displaced stack which are promoted by electrostatic and van der Waals forces, respectively. Based on these results we found the global energy minimum structures of the coronene clusters in the situation when the electrostatic interaction dominates and in the situation when the van der Waals forces prevail (also studied previously [4]). The dominance of electrostatics results in T-shaped motifs, exemplified on the left for (coronene)$_3$ cluster, while van der Waals interaction favors stack-based configurations, shown below for (coronene)$_{10}$.



Rotational constants and the normal modes in the obtained sets of structures are distinctively different making possible to distinguish experimentally between the two situations.

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REFERENCES
[1] W. W. Duley, S. Lazarev, *Astrophysical Journal* **612**, L33 (2004)
[2] M. Rapicioli, C. Joblin, P. Boissel, *Astronomy and Astrophysics* **429**, 193 (2005)
[3] O. I. Obolensky, V. Semenikhina, A. V. Solov'yov, W. Greiner, *Int. J. of Quantum Chemistry* **107**, 1335 (2007)
[4] M. Rapacioli, F. Calvo, F. Spiegelman, C. Joblin, D. Wales, *J. Phys. Chem.* A **109**, 2487 (2005)

PS-19

INDUCED-DISPERSION MECHANISM OF VELOCITY-DEPENDENT INTERACTION OF ATOMS AND CLUSTERS WITH SURFACE

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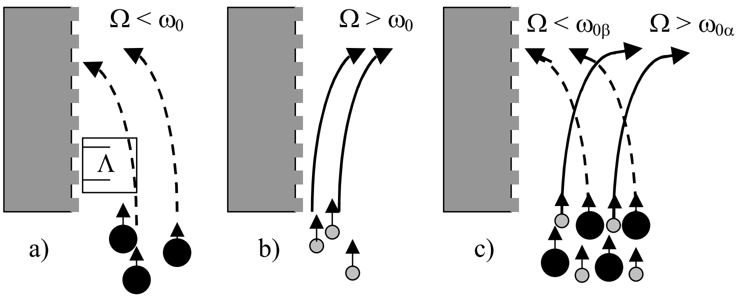
The collective induced-dispersion mechanism of selective atom-surface interaction - non-diffraction velocity and frequency-dependent grazing collision and interaction between the beam of neutral atoms (molecules or clusters) with longitudinal velocity $\vec{v} = \vec{e}_z v$ and the target surface having a periodical structure (e.g., a crystal lattice with periodical electric field $\vec{E}(x,z) = \vec{E}(x,z \pm \Lambda)$, outside the crystal in the immediate vicinity of its surface) is discussed. The same phenomenon takes place for the modulated surface moving in the motionless cold gas of neutral atoms or molecules. The regarded effect takes place at certain combination of relative (particle to surface) velocity $\vec{v}$, longitudinal period Λ of the near-surface field and the frequency ω_0 of one of the strong electron resonance in a moving particle. At longitudinal relative motion of atoms in the near-surface periodic electric field $\vec{E}(x,z)$ they are influenced by alternate excitation with frequency $\Omega = 2\pi v/\Lambda$. In atoms with polarizability $\alpha(\Omega,\omega_0)$ the field induces electric moments $\vec{p}(t,x) = \alpha(\Omega,\omega_0)\vec{E}(x,t)$, which oscillate with the same frequency Ω and phase φ dependent upon the relation between Ω and ω_0. Each of these moments (1) in turn interacts with the field $\vec{E}(x, z = vt)$ it's caused by. The interaction energy is $W(x) = \alpha(\Omega,\omega_0)|E(x,t)|^2$.

It was shown [1] that the values of atom-surface interaction energy and the force of interaction

$$W(x) \approx W_0 \exp(-2\pi x/\Lambda), \quad F_x \approx [2\pi W_0/\Lambda] \exp(-2\pi x/\Lambda)$$

depends on the distance x from the surface to a moving atom. For oriented Ge or Si crystal surface (e.g., at $\Lambda \approx 2$Å) we have $|W_0| \approx 0{,}5$ eV. The sign of this energy depends on the dispersion of internal electron atom polarizability $\alpha(\Omega,\omega_0) \equiv \alpha_0/[(\Omega-\omega_0)+i\Gamma/2]$ and is determined by the relation between frequencies Ω and ω_0.

If $\Omega < \omega_0$ (see case a) on Fig.), the energy of atom-surface interaction W(x) is negative, which means the atoms are attracted to the surface. Such combination of Ω, v, Λ and ω_0 causes the existence of a potential hole $W(x)<0$ and negative force F_x near the surface and the increase of near-surface concentration of longitudinally moving atoms. In another case b) (at $\Omega > \omega_0$) the energy $W(x)>0$ (potential barrier) and force $F_x>0$ and the atoms are repelled from the surface to the distance $x > \Lambda$. For the case c) of multi-component moving atom (molecule) beam (with presence of different types of atoms A_α, A_β,.., A_γ with resonant frequencies $\omega_{0\alpha}$, $\omega_{0\beta}$,..., $\omega_{0\gamma}$) it is possible to achieve strong attraction for certain type A_α of atoms (at $\Omega < \omega_{0\alpha}$ and $(\omega_{0\alpha}-\Omega) \leq \Gamma_\alpha$) and repulsion regime of motion for other types A_β,.., A_γ of atoms (at $\Omega > \omega_{0\beta}$,..., $\omega_{0\gamma}$).



This mechanism of controlled interaction between the neutral atom (molecules) and the crystal surface was investigated for the case of natural crystal (plane) surface and at motion of different clusters (e.g. C_{60} fulleren) along the periodical 2-dimensional magnetic domain surface.

REFERENCES

[1]. V.I. Vysotskii, R.N. Kuzmin // Soviet Phys. - Uspekhi, v.35 (1992) 725-746

PS-20

LEAD ISLANDS CRYOFORMATION: EXPERIMENT AND SIMULATION

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The microstructure of the material can essentially influence its properties. This phenomenon becomes more pronounced when going down to nanoscale. The present work deals with the preparation of nanostructured thin Pb films by vapor deposition technique. It was found that such films near the percolation threshold exhibit gas sensitivity towards ammonia vapors [1]. These films were deposited on glass and alumina at 80 K and then annealed to room temperature. The film structure was strongly influenced by the deposition conditions, namely the flux. By changing the flux from $4 \cdot 10^{13}$ atoms/cm^2 to $16 \cdot 10^{13}$ atoms/cm^2 one was able to produce sensitive and insensitive films with the same amount of lead and very different microstructure [2]. The conductance of the deposit was measured during the experiment using Keithley 6517A electrometer. It was found that the resistance changed during annealing nonmonotonically during heating from 80 to 300 K. The columnar growth model, explaining the observed changes is proposed.

In order to study the atomistic processes, taking place during the cryocondensation, the combined Molecular Dynamics (MD) – Temperature Accelerated Dynamics (TAD [3]) simulation was carried out. Using TAD allowed us to simulate the deposition of Pb with typical rates (0.05 ML/s), used in our experiments. The dependence of film roughness and percolation threshold on deposition conditions are discussed.

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REFERENCES

[1] V. Bochenkov, V. Zagorsky, and G. Sergeev, *Sensor. Act. B*, 103 (2004) 375.
[2] V.E. Bochenkov, G.B. Sergeev *Adv. Coll. Int. Sci.,* 116 (2005), 245-254.
[3] F. Montalenti and A. Voter, *Phys. Rev. B*, 64 (2001) 081401.

PS-21

ELECTRON IMPACT IONIZATION OF DIMERS AND CLUSTERS

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Introducing a single transition metal atom into a small gold cluster can modify the electronic shell structure significantly. The strength of the interaction between host cluster and dopant atom is strongly determined by the size of the dopant (geometric stabilization) and the number of dopant valence electrons (electronic shell closures). The combined effect of these parameters is reflected in the total binding energy, B_n, and the differential binding energies or dissociation energies, $D_n = B_n - B_{n-1}$. In our experiment, the clusters are excited by high fluence laser pulses to promote fragmentation. After photoexcitation the internal energy is statistically distributed over the vibrational modes of the system and the cluster does not instantaneously dissociate. This concept of delayed fragmentation can be described by statistical models and the recorded fragmentation yields after a well defined time can be converted to dissociation energies [1].

The dissociation pathways and size-dependent stability of cationic Au_nX^+ (X = Y, Er, and Nb, n = 5-20) clusters produced in a laser vaporization source are investigated. The photo-fragmentation experiments were performed with a reflectron time-of-flight mass spectrometer incorporating a wire-type mass gate. Statistical models based on the detailed balance theory are presented and applied to determine the relative dissociation energies from both the recorded abundances and the fraction of mass selected clusters that undergo delayed fragmentation in the time window of the experiment. The dopant elements Y, Er, and Nb have been chosen to extend our earlier work using 3*d* dopant atoms towards 4*d* transition metal atoms (Y, Nb) and lanthanide (Er) dopants [2], [3], [4]. Gold monomer evaporation turns out to be the most likely fragmentation channel for doped Au_nX^+, with the exception of $\mathrm{Au}_{18}\mathrm{Y}^+$ and $\mathrm{Au}_{20}\mathrm{Y}^+$ for which gold dimer evaporation is a competing channel. The extracted dissociation energies show an odd-even staggering and enhanced stabilities for certain cluster sizes, in agreement with simple electronic shell model predictions.

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REFERENCES

[1] K. Hansen, A. Herlert, L. Schweikhard and M. Vogel, *Phys. Rev.* A **73**, 063202 (2006)
[2] W. Bouwen, F. Vanhoutte, F. Despa, S. Bouckaert, S. Neukermans, L. Theil Kuhn, H. Weidele, P. Lievens, R.E. Silverans, *Chem. Phys. Lett.* **314**, 227 (1999)
[3] S. Neukermans, E. Janssens, H. Tanaka, R.E. Silverans, P. Lievens, *Phys. Rev. Lett.* **90**, 033401 (2003)
[4] E. Janssens, H. Tanaka, S. Neukermans, R.E. Silverans, and P. Lievens, *Phys. Rev.* B **69**, 085402 (2004)

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T-SHAPED CORONENE DIMER

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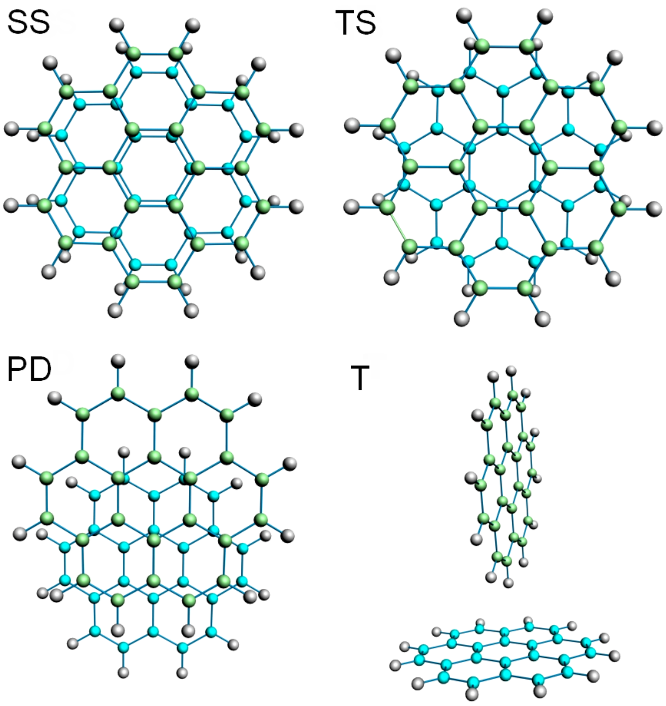
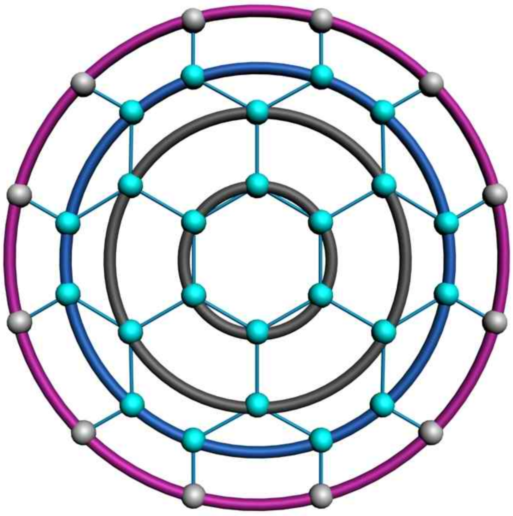


We have performed density functional theory (DFT) calculations of energies for four plausible configurations of dimer of coronene ($C_{24}H_{12}$) [1] (shown on the left). The stack configurations (superimposed stack (SS), twisted stack (TS) and parallel-displaced (PD) stack) are favoured by the van der Waals forces while the T-shaped configuration is favoured by the electrostatic interaction. In order to get a better description of the van der Waals interaction, the DFT results were corrected by a phenomenological London-type $1/r^6$ term. Depending on the magnitude of this term, both the T-shaped and the PD stack configurations can be the global minimum configuration of the system. It is quite surprising that the T-shaped configuration is energetically competitive for such a large system for which a graphite-like stacked structure had been expected *a priori*.

We developed a model in which each coronene molecule is represented by a set of four charged rings [1] (shown on the right). Adjusting the parameters of these rings one can reproduce the binding energies and potential energy surfaces obtained with the *ab initio* DFT calculations. In particular, one can analyze the interplay between van der Waals polarization forces and the electrostatic interaction and determine ranges of parameters which make the T-shaped or the PD stack energetically preferable geometries.



We acknowledge partial financial support from European Commission (project EXCELL).

REFERENCES

[1] O. I. Obolensky, V. Semenikhina, A. V. Solov’yov, W. Greiner, *Int. J. of Quantum Chemistry* **107**, 1335 (2007)

PS-23

X-RAY PHOTOELECTRON SPECTROSCOPIC STUDIES ON COBALT AND NICKEL POLYNUCLEAR COMPLEXES

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X-ray photoelectron spectra (XPS) and electronic structure of some cobalt(II), cobalt(III), cobalt(II/III) and nickel(II) complexes have been investigated on the Kratos Axis Ultra spectrometer with monochromatic excitation by AlK_α radiation. XPS has been widely applied to study the transition metals complexes [1]. In general, each electronic state demonstrates rather characteristic and distinguishing features in the XPS spectrum. For the first time, we have used the XPS method to study the electronic structure of polynuclear cobalt and nickel trimethylacetates. Polynuclear transition metal complexes with bridging carboxyl ligands have unusual structures and demonstrate unique chemical and physical properties. The complexes refer to magneto-active clusters and may be considered as models for metallo-ferments [2]. Polynuclear systems with bridging ligands are among the best to create conditions for the exchange interactions between paramagnetic centers. 3d transition metal (M) complexes are of special importance in this context: high electron-density delocalisation in M-OCO-M moieties generates good exchange channels in them [2,3]. The Me2p, Me3p, Me3s, N1s, O1s and C1s spectra of the cobalt/nickel complexes have been fitted with different states in accordance their structures obtained by XSA [4]. It has been found that composition obtained by using atomic sensitivity factors and reference sample are far different. The second way gives data that are in good agreement with XSA data. Taking into account correlation between $Me2p_{3/2}$-$Me2p_{1/2}$ separations, both relative satellite intensities and energies, and magnetic characteristics the complexes have been identified as Me(II), Co(III) and Co(II/III) complexes or other words as high-spin, diamagnetic and high-spin/diamagnetic ones. It also supported by peak widths and binding energies. It has been found that the most sensitive parameter of the state of the cobalt atom is $\Sigma sat/I\ Co2p_{3/2}$, which differs by a factor of two for the high-spin Co(II) and the low-spin Co(III) complexes. The XPS data suggest that the nonanuclear cobalt complex can be described as a high-spin Co(II) complex and the hexanuclear cobalt complex contains both the diamagnetic cobalt(III) and the high-spin cobalt(II). Thus structural relationships of the newly synthesized complexes are established on the basis of XSA and XPS results.

REFERENCES

[1] C. Frost, C. F. McDowell, and I. S. Woolsey, Mol. Phys. 27, 1473 (1974)

[2] V.V. Pavlishchuk, Teor. Eksp. Khim.v.33, 6, 341 (1997)

[3] V.I. Ovcharenko and R.Z. Sagdeev, Usp. Khim., v.68, 5, 381 (1999)

[4] I.G. Fomina, A.A. Sidorov, G.G. Aleksandrov, V.N. Ikorskii, V.M. Novotortsev, S.E. Nefedov, I.L. Eremenko, Russ. Chem. Bull. Int. Ed. 51, 1423 (2002).

PS-24

INTERPLAY OF ELECTRONIC AND GEOMETRY SHELL CLOSURES FOR NEUTRAL AND CHARGED STRONTIUM CLUSTERS

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The optimized structure and electronic properties of neutral, singly and doubly charged strontium clusters have been investigated using *ab initio* theoretical methods based on density-functional theory. We have systematically calculated the optimized geometries of those clusters consisting of up to 14 atoms, their average bonding distances, electronic shell closures, binding energies per atom, dissociation energies and the gap between the highest occupied and the lowest unoccupied molecular orbitals [1]. It is demonstrated that the size-evolution of structural and electronic properties of strontium clusters is governed by an interplay of the electronic and geometry shell closures. It is shown that the excessive charge affects the optimized geometry of strontium clusters. Ionization of small strontium clusters results in the alteration of the magic numbers. Stability of positively charged strontium clusters towards fission is analysed [1,2]. The obtained results are compared with the available experimental and theoretical data.

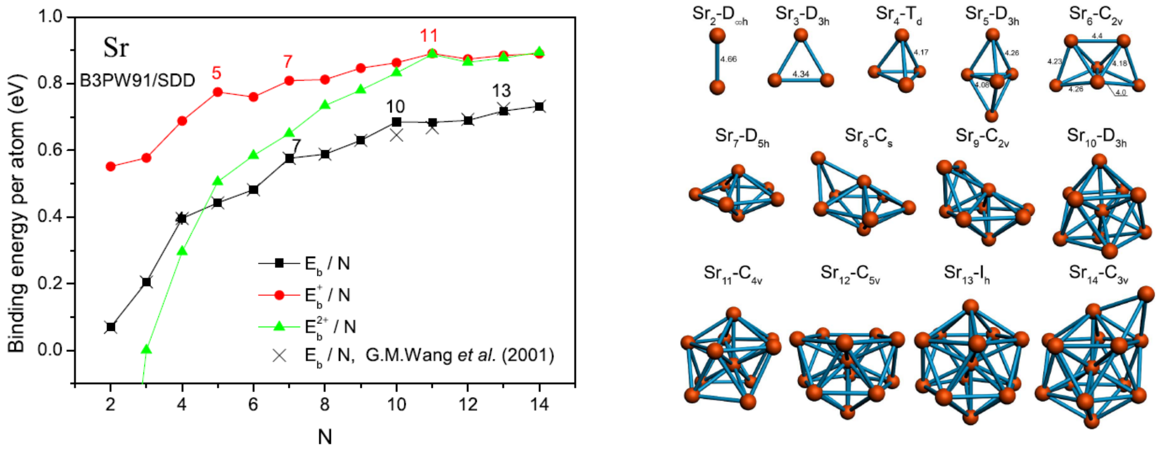

Figure 1: Left: binding energy per atom for the most stable neutral (filled squares), singly charged (filled circles) and doubly charged (filled triangles) Sr clusters; right: optimized geometries of neutral Sr clusters.

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REFERENCES

[1] A. Lyalin, I. A. Solov'yov , A. V. Solov'yov and W. Greiner, Phys. Rev. A **75**, 053201 (2007).
[2] A. Lyalin, A.V. Solov'yov, C. Bréchignac, and W. Greiner, J. Phys. B: At. Mol. Opt. Phys., **38**, L129 (2005).
[3] G.M. Wang, E. Blaisten-Barojas, A.E. Roitberg, and T.P. Martin, J. Chem. Phys. **115**, 3640 (2001).

PS-25

Temperature Dependent Electric Response of Isolated Tin Cluster

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In a molecular beam experiment we probe the deviation of neutral tin clusters in an inhomogeneous electric field. This deviation is traced back to the interaction of the electric dipole moment and static polarizability of the isolated clusters with the electric field.

The tin clusters are produced with a laser ablation source and subsequently expanded through a nozzle into a vacuum apparatus to form a molecular beam. By thermalizing the expansion nozzle between 40 and 310 K the internal temperature of the clusters can be changed. The deviation of the cluster beam in the inhomogeneous field is size-selectively monitored with a position-sensitive mass spectrometer.

In the case of nozzle temperatures above 50 K the experimental results can be well described by modelling the tin clusters as classical, symmetric rigid rotors with a body fixed, effective electric dipole moment and static polarizability tensor and the rotational temperature as fitting parameters [1]. Especially the pronounced size-dependent variation of the effective dipole moment can be directly compared to quantum chemical results [2], which offers a promising route to experimentally validate theoretically predicted cluster structures.

For nozzle temperatures below 50 K the simple rigid rotor model fails for certain cluster sizes as was already pointed out by [3, 4] in the case of niob clusters. Possible reasons for this failure are discussed.

References

[1] P. Dugourd, I. Compagnon, F. Lepine, R. Antoine, D. Rayane, M. Broyer, *Chem. Phys. Lett.*, **336**, 511 (2001).

[2] S. Schäfer, B. Assadollahzadeh, R. Schäfer, P. Schwerdtfeger, *to be published.*

[3] R. Moro, X. S. Xu, S. Y. Yin, W. A. de Heer, *Science*, **300**, 1265 (2003).

[4] X. S. Xu, S. Y. Yin, R. Moro, A. Liang, J. Bowlan, W. A. de Heer, *Phys. Rev. B.* **75**, 085429 (2007).

PS-26

LOW-ENERGY ELECTRON CAPTURE BY MOLECULAR CLUSTER. EVIDENCE OF POLARIZATION AND DIPOLE MECHANISMS

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We report measurements of the absolute cross sections σ^s of low-energy electron attachment to $(CO_2)_n$ and $(H_2O)_n$ molecular clusters. The experiments were performed using crossed beam technique for electron–cluster single collisions and variable cluster sizes n and electron energies E [1, 2]. The clusters were formed by gas (or steam) condensation in a supersonic jet downstream of a sonic nozzle [3].

A sharp increase in the attachment cross section has been observed at $E \to 0$ (see Figure). The effect is most pronounced for water cluster: at $n \approx 1\ 000$ and $E \approx 0.2$ eV (that is the minimal average energy of electron beam used in our research) the attachment cross section is one order of magnitude higher than the geometrical cross-sections of solid cluster.

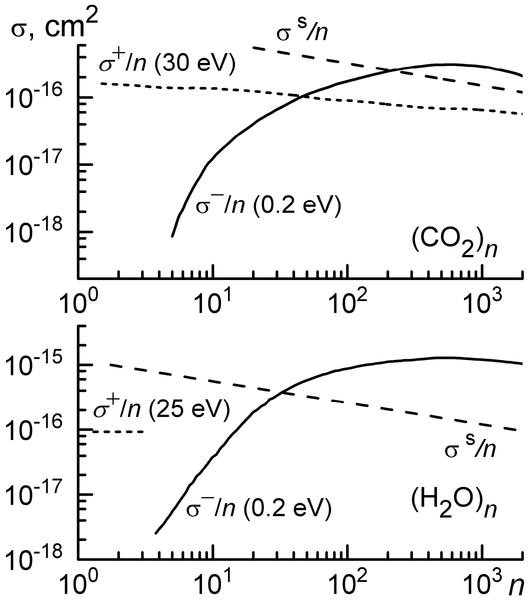

Figure 1: Specific (per molecule) cross sections of electron attachment (solid lines), of electron impact ionization (dotted lines) and geometrical cross-sections (dashed lines) for clusters of carbon dioxide and water vs. cluster size n.

The possible pathways of attacment resulting in σ^s as large as these are discussed in the report. The long-range polarization interaction between electron and cluster is found to be responsible for electron capture by $(CO_2)_n$, but fails to account for larger σ^s for water cluster. In latter case the most likely mechanism is the electron capture in dipolar electric field of cluster. The origin of large permanent dipole moment of water cluster is discussed and the approximate magnitude of the moment is estimated.

REFERENCES

[1] Anatoli A. Vostrikov, Dmitri Yu. Dubov, *Tech. Phys.* **51**, 540 (2006)
[2] Anatoli A. Vostrikov, Dmitri Yu. Dubov, *Tech. Phys.* **51**, 1537 (2006)
[3] Anatoli A. Vostrikov, Dmitri Yu. Dubov, *JETP* **98**, 197 (2004)

PS-27

MODELS OF CLUSTERS COLLISIONS AT DIRECT MONTE-CARLO SIMULATION OF NANOCLUSTERS FORMATION IN EXPANDING VAPOR

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The details of cluster formation from vapor are of fundamental interest to the study of gas–condensed phase transitions laying in the basis of many modern technologies of nanoclusters and nanocluster materials production. One of the main methods modeling these processes is the Direct Simulation Monte-Carlo (DSMC) method [1]. The realization DSMC method requires the description of particles collision at a microscopic level. However in many cases the needed information is missing even for ordinary transition metals and semi-conductors (as silicon or carbon).

In this work the problem of the particles collision model selection was considered by the example of DSMC modeling of condensation of silicon (or carbon) vapor expanding into vacuum or inert gas. The important feature of this problem is the wide range of vapor temperature change (300-7000 K) at expansion. In these conditions using the simple collision models (as hard sphere) leads to the low accuracy. The more complete model of atoms and clusters collisions was created in the context of the common model of clusters formation processes [2] in the mixture of silicon (or carbon) vapor and helium. The model takes into consideration the dependence of the collision cross section from the collision energy. The model is based on the calculations executed partially by the scheme described in the work [3] using the potential obtained by the density functional theory (DFT) calculations. The non-additivity of potential of nanocluster's atoms is shown.

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REFERENCES

[1] G. A. Bird. *Molecular Gas Dynamics and Direct Simulation of Gas Flows*. Clarendon Press, Oxford. 1994.
[2] N. Y. Bykov, G. A. Lukyanov. *Thermophysics and Aeromechanics*. 13, 569 (2006).
[3] I. A. Sokolova, S. A. Vasilevskiy, A.V. Adriatis, Computation of transport factor for multicomponent gas and plasma. On-line journal «*Physicochemical kinetics in gas dynamics*» 3, (2005), www.chemphys.edu.ru/pdf/2005-06-12-001.pdf.

PS-28

ELECTRON STEREODYANAMICS IN MULTIPLE IONIZATION OF RARE GAS DIMERES WITH SLOW HIGHLY CHARGED IONS

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Much attention has been called to the multiple ionization dynamics of molecules by impact of ions. For diatomic molecules, the angular and charge state distributions of Coulomb explosion fragments have been intensively measured for ‘fixed-in-space’ molecules with the momentum imaging technique. In contrast to covalent molecules, however, rare gas dimers have been little investigated, where the dynamics thereby is far different due to electron localization character. In the present contributions, we address the multiple ionization (actually electron transfer) by slow (v <<1 au) highly charged ions in the three-center Coulombic over-the-barrier model developed by the present authors [1-3]; in particular, we clarify the electron stereodynamics peculiar to rare gas dimers (e.g. Ne_2) in comparison to covalent molecules (e.g. N_2) and analyze how it manifests in the ionized fragment distributions.

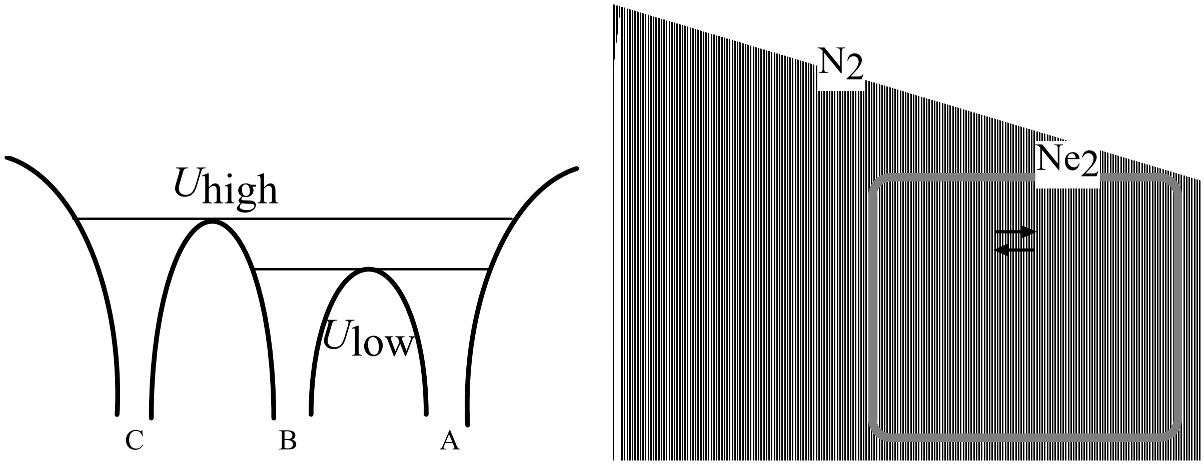

Figure: In the three-center Coulombic over-the-barrier model [1], single electron dynamics is related to two saddle points U_{high} and U_{low} generated by effective charges at three sites A, B and C for collisions of A (ion) + BC (covalent molecule or rare gas dimer).

REFERENCES
[1] A. Ichimura and T. Ohyama-Yamaguchi, Physica Scripta. T 80 (1999) 190.
[2] T. Ohyama-Yamaguchi and A. Ichimura, Nucl. Instr. and Meth. B 205 (2003) 620.
[3] T. Ohyama-Yamaguchi and A. Ichimura, Journl of Physics: Conference Series 58 (2007)247.

PS-29

DYNAMICS OF ELECTRON-ION INTERACTION IN ALCALINE CLUSTERS

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Fission always remains a process of great interest and practical importance. This gives an example of a collective process with large amplitude. A recent conference [1] confirms the importance of studying fission in cluster physics.

Clusters play an outstanding role in the modern nanotechnology. Carbon tubes and fullerens, clusters of alkali elements find most important applications. Original method of calculation of the collective motion is developed in papers [2–7]. Large attention is paid to the collective phonon mode of the ion core, and to its interaction with the plasmon mode of the collective motion of valence electrons. Plasmons find application in construction of computers of future generations. In [2–7], specifically, the role of the phonons has been noted in the thermalization of the plasmon energy, excited by laser. It was specifically noted that relaxation time of the plasmon energy coincides with the phonon period. We show that this result helps us to better understand mechanism of cluster fission. On the basis of the developed method, ternary fission of the $^{27}\mathrm{Na}^{3+}$ sodium clusters into three equal fragments is discussed. We also develop an analytical model of the plasmon-phonon interaction [6,7]. The model properly takes into account the compressibility of the cluster matter. Comparison with experiment is discussed. Its application to description of the Raman scattering is considered.

References

[1] Fission Dynamics of Atomic Clusters and Nuclei. Proc. of the Int. Workshop, Luso, Portugal, 15–19 May 2000. Ed. J. da Providencia, D.M. Brink, F. Karpechine and F.B. Malik. Singapore: World Scientific, 2001.

[2] F.F. Karpeshin. Nuclear Fission in Muonic Atoms and Resonance Conversion. St.-Petersburg: “Nauka”, 2006.

[3] F.F. Karpeshin, A. Vieira, C. Fiolhais, J. da Providencia jr., Europhys. Lett. **42**, 149 (1998).

[4] F.F. Karpeshin, J. da Providencia, C. Providencia and J. da Providencia jr., European Phys. J. **D18**, 319 (2002).

[5] F.F. Karpeshin, J. da Providencia, C. Providencia and J. da Providencia jr., Europhys. Lett. **67**, 760 (2004)

[6] F.F. Karpeshin, J. da Providencia, Vestn. St.-Petersburg Fin.-Eng. Univ. P. 2345 (2001).

[7] F.F. Karpeshin, J. da Providencia, Izv. RAN, Ser. Fiz. **70**, 757 (2006)

PS-30

A NOVEL METHOD FOR MEASURING CLUSTERS CALORIC CURVES

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Melting of small clusters has received a considerable interest since the pioneer experimental work of Haberland *et al* who found that the melting temperature of very small particle show a non-monotonical behaviour. This temperature is found either lower (for the majority of clusters, for instance sodium [1]) or higher (for tin and gallium [2]) than in the bulk. Moreover, both the melting temperature and the latent heat of fusion can vary considerably from one size to another.

We propose here a novel method based on an original experimental setup [3] to measure the caloric curve of clusters. The basic idea is to count the number of atoms that can be stuck onto a cluster as a function of its initial temperature. At each sticking, the internal energy of the cluster M_n is increased by a known energy. After a given number of sticking collisions n_{max} the cluster is so heated that it dissociates before it can stick again. n_{max} is related to the dissociation time, thus is a measure of the internal energy. Our method does not require laser excitation, it is easily transferable to many systems and it is model free.

We present here our first experimental results obtained with sodium clusters. They confirm the ones of Haberland *et al* and extend them towards small sizes.

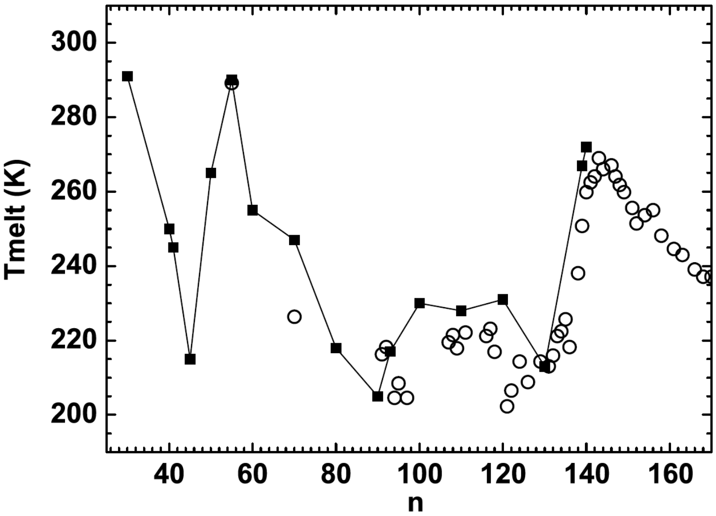

Figure 1: Comparison of our results (squares) with the one of Haberland *et al* [1] (circles).

References

[1] H. Haberland *et al*, Phys. Rev. Lett. **94**, 035701 (2005)

[2] G.A. Breaux *et al*, Phys. Rev. Lett. **91**, 215508 (2003)

[3] F. Chirot *et al*, Rev. Sci. Instrum. **77**, 063108 (2006).

PS-31

Stability, vibration and thermodynamics of clusters of polycyclic aromatic hydrocarbon molecules

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The vibrational spectra of clusters of neutral coronene ($C_{24}H_{12}$) molecules have been calculated using a mixed quantum/classical scheme, each molecule being described by a Tight-Binding Hamiltonian [1], the intermolecular forces being provided by interatomic repulsion and exchange-dispersion contributions complemented by Coulomb interactions between atomic point charges to describe multipolar interactions [2]. Couplings between intramolecular and intermolecular vibrational modes have been investigated as a function of cluster size for different isomeric forms, particularly the stacked conformations [3]. We show that, in large clusters, intra- and intermolecular modes mixing tends to fill the mid-infrared range.

The heat capacity of the $(C_{24}H_{12})_8$ cluster has been also calculated as a function of the temperature, emphasizing the isomerizations that take place during melting [3]. From the Pitzer-Gwinn semi-classical approximation, the effects of vibrational delocalization have been investigated. We show that these effects are important enough to wash out all signatures of the structural transitions, including the melting phase change, on the caloric curve.

Finally, the stability of such polycyclic aromatic hydrocarbon clusters in the interstellar medium will be discussed from a theoretical perspective, by looking at the growth and destruction mechanisms and considering the competition between nucleation, evaporation and IR emission processes [1].

References

[1] M. Rapacioli, F. Calvo, C. Joblin, P. Parneix, D. Toublanc, and F. Spiegelman, Astron. Astrophys. **460**, 519 (2006)

[2] M. Rapacioli, F. Calvo, F. Spiegelman, C. Joblin, and D. J. Wales, J. Phys. Chem. A **109**, 2487 (2005)

[3] M. Rapacioli, F. Calvo, C. Joblin, P. Parneix, and F. Spiegelman, J. Phys. Chem. A (in press, 2007).

PS-32

MOLECULAR DYNAMICS FOR FISSION PROCESSES

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Fission is a ubiquitous process playing important roles in many physical phenomena. In fission, the initial and final states of a system are separated by a potential barrier, the so-called fission barrier. The height and the shape of this barrier determine most of the experimental observables of the process, such as characteristic fission times and branching ratios between different fission channels.

We discuss relative advantages and disadvantages of two general approaches which are used for carrying out a molecular dynamics analysis of fission processes [1]. The goal of both approaches consists in finding the optimum fission pathway which minimizes the fission barrier. The optimum fission pathway frequently involves complicated rearrangements of the ionic structure including formation of intermediate metastable configurations [2] which makes this problem rather difficult.

The first approach to the molecular dynamics analysis is the conventional molecular dynamics simulations which emulate the evolution of a system with time [3,4]. This approach relies on the assumption that the system explores sufficiently large volumes of its phase space (thus covering sufficiently large areas of the potential energy surface) if a sufficiently large number of runs of the simulations are made and each run is sufficiently long. In order to shorten the required CPU time one usually assigns rather high initial temperatures to the system. In such a case, however, the system tends to fission along the shortest path rather than along the path with the lowest potential barrier. Therefore, one has to find the optimum balance between the reliability and the feasibility of simulations.

The second approach consists in a direct (manually- or algorithmically-driven) exploration of the potential energy surface of the system [5]. This approach allows one to perform a systematic analysis of the various fission scenarios, reducing the chance of an accidental omission of an important fission pathway. We review one of the possible algorithms for exploration of the potential energy surface [3,5] and give examples of its applications to fission processes in atomic clusters, bio- and macro-molecules.

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REFERENCES

[1] A. Lyalin, O.I. Obolensky, A.V. Solov'yov, W. Greiner, *Int. J. Mod. Phys. E* **15**, 153 (2006)
[2] O.I. Obolensky, A.G. Lyalin, A.V. Solov'yov, W. Greiner, *Physical Review B* **72**, 085433 (2005) and references therein
[3] P.-G. Reinhard and E. Suraud, Introduction to Cluster Dynamics (Wiley-VCH, Weinheim, 2004)
[4] F. Baletto and R. Ferrando, *Rev. Mod. Phys.* **77**, 371 (2005)
[5] O.I. Obolensky, I.A. Solov'yov, A.V. Solov'yov, W. Greiner, *Computing Letters* **1**, 313 (2005)

PS-33

ION PAIRS FORMATION AT WATER CLUSTER FRAGMENTATION

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Using the nozzle molecular beam (MB) technique and the molecular dynamics (MD) method, the processes of both the polar dissociation (PD) of molecules in $(H_2O)_n$ clusters and the polar fragmentation (PF) of clusters

$$(H_2O)_n \to H_2O^*(H_2O)_{n-1} \xrightarrow{PD} H^+(H_2O)_i \cdot HO^-(H_2O)_j \xrightarrow{PF} H^+(H_2O)_i + HO^-(H_2O)_j \quad (1)$$

at cluster-surface and cluster-cluster collisions are studied.

Molecular beam of water clusters was formed from a supersonic jet of water vapor [1]. In MB experiments, the scattering indicatrix of neutral and charged fragments of $(H_2O)_n$ clusters at varying the mean cluster size in the beam ($n<5\cdot10^4$) and the angle α of the cluster incidence on the target were determined. The targets were made as flat plates from different materials (metals, dielectrics, semiconductors). The kinetic energy of water clusters in the beam was determined by the temperature of superheated vapor in the nozzle source and its value per molecule did not exceed 0.17 eV. It has been found that the scattering is of lobed character, and the ion current increases with n at $n>100$ and α. The values and relation between ion currents of different signs depend on the target material and the angle α.

The MD calculations of stable geometries and energy parameters of neutral $(H_2O)_n$ ($n \le 100$), positively $H^+(H_2O)_n$ and negatively $HO^-(H_2O)_n$ ($n \le 15$) charged clusters have been carried out within the framework of the Stillinger-David polarization model [2,3], which assumes that a H_2O molecule consist of two protons and doubly charged negative oxygen ion having a scalar polarizability. The interaction potential is constructed as a sum of central potential responsible to the polarization energy, which appears due to induced dipole moment of the oxygen ion in the field of remaining charges.

The polar dissociation energy of an isolated water molecule E_{PD}^{M} equals 16.9 eV, whereas the threshold energy E_{PD}^{aq} of the formation of the ionic pair H^+_{aq} and OH^-_{aq} in liquid water is much smaller ($E_{PD} \approx 0.58\text{eV}$) because of the ion hydration. The values of both E_{PD} for molecules in $(H_2O)_n$ clusters and the threshold energy of the clusters polar fragmentation E_{PF}, obviously, depend on n. The energy E_{PD} for the process (1) at the constant temperature can be determined using energy parameters of neutral and charged clusters. For this purpose, we shall schematically present the process (1) in the form

$$(H_2O)_n \to nH_2O \to (n-1)(H_2O) + H^+ + OH^- \to H^+(H_2O)_i + OH^-(H_2O)_j. \quad (2)$$

The threshold energy of this process is

$$E_{PF}(n) = [U(n) + E_{PD}^{M}] - [U^+(i) + U^-(j)], \quad (3)$$

where $U(n), U^+(i)$ and $U^-(j)$ are the breakage energies of intramolecular bonds in neutral $(H_2O)_n$, positively $H^+(H_2O)_i$ and negatively $OH^-(H_2O)_j$ charged clusters, respectively.

In this work, the size dependences of quantities included in (3) were determined through the MD simulation of neutral and charged water clusters.

The dynamics of the cluster fragmentation and ion pairs formation during cluster – cluster (n=27 and 100) and cluster – surface (n=27, 64 and 100) collisions are investigated through the MD simulation for relative collision velocities of 1, 3, and 10 km/s. It has been obtained that ion pairs are formed due excitation at compression of the finite number of molecules followed by their polar dissociation and ion hydratation.

The formation of charged particles was observed only for *V* = 10 km/s. At this velocity the kinetic energy per molecule in cluster is 2.33 eV that is less than E_{PD}^{M} by a factor 7.25. The substantial decrease in E_{PD} for clusters qualitatively corresponds to the size dependences of $E_{PF}(n)$.

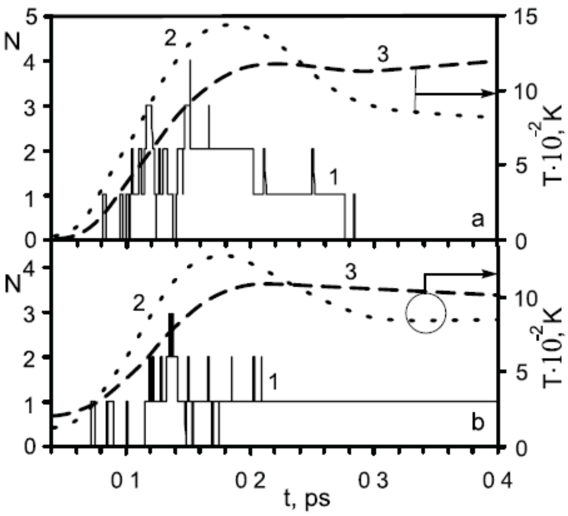

Figure 1: Time variation of the number of ion pairs N, rotational T_R and vibrational T_V temperatures of molecules in colliding $(H_2O)_{27}$ clusters at V=10 km/s and T=20 K (a) T=130 K (b)

Figure 1 shows the time variation of the number of ion pairs N, rotational T_R and vibrational T_V temperatures of molecules in colliding $(H_2O)_{27}$ clusters at V=10 km/s and temperature of clusters 130 K. It should be pointed out that the maximal lifetime of ion pairs (the time up to recombination) was detected at T = 130 K, the maximal number of ion pairs (N = 4) was at T = 70 K. Despite the continuous (classical) energy distribution over the vibrational degrees of freedom in the model adopted, the collision process exhibits a nonequilibrium character. T_R increases faster than T_V. The reason is that at the hydration of ions H^+ and OH^- formed on impact, the first stage of the dissipation of the hydration energy is bound up with the rotational excitation of molecules surrounding the ions. The maximal value of N was observed when t ranges from 0.11 to 0.13 ps that correspond to the most compression of clusters.

This work has been supported by the Russian Foundation for Basic Research (grants 06-03-32751 and 07-02-00825).

REFERENCES

[1] A.A. Vostrikov, D.Yu. Dubov, Journal of Experimental and Theoretical Physics **98**, 197 (2004).
[2] F.H. Stillinger, C.W. David, Journal of Chemical Physics **69**, 1473 (1978)
[3] A.A. Vostrikov, S.V. Drozdov, V.S. Rudnev, L.I. Kurkina, Computational Materials Science, **35**, 254 (2006).

PS-34

FRAGMENTATION OF NUCLEI AND CLUSTERS

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Emission of mono- and dimers from clusters, similar to alpha decay of nuclei, can be described within statistical theory in terms of the inverse cross-section. Such an approach assuming time reversibility of the process leaves out of the scope a possibility of its experimental check. This is in contrast with a number of indications that violations of the reversibility may arise due to back-transparency of the inner slope of the potential barrier in the ingoing channel, or different response of the nuclear surface on the interaction with the emitted and the same incoming particle, or due to temperature effects on the barrier distribution. Moreover, traditional decay theory deals with tunnelling through a barrier of a particle which is in the quasistationary state. This does not involve important cases when a cluster is emitted with an energy which is between the quasistationary eigenvalues, as e.g. in nuclear alpha decay from compound systems formed in fusion reaction.

Our approach [1] allows one to calculate the decay width at any energy of the emitted particle. Strong resonance effects are, specifically, predicted in alpha spectra from compound systems produced in heavy-ion collisions. Both experimental and theoretical aspects of cluster physics, uniquely placed between nuclear physics on one hand and atomic, including problems of fusion and fission, fragmentation, can be better understood on the basis of the discussed approach.

References

[1] F.F. Karpeshin, G. LaRana, E. Vardaci, A. Brondi, R. Moro, S.N. Abramovich, V.I. Serov, J. Phys. B: G: Nucl. Part. Phys. **34**, 587 (2007)

PS-35

FRAGMENTATION OF FULLERENES AS A PHASE TRANSITION PROCESS

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We investigate the various fragmentation and formation channels of fullerenes using a statistical description first applied to polypeptide folding[1]. In this formalism, the growth and defragmentation of fullerenes are treated as phase transitions, and we present a scheme of constructing a parameter-free partition function describing such a process. Thus, we develop a theory describing all essential thermodynamical properties such as heat capactiy, free energy and the phase transition temperature. The predictions of our theory are compared with available experimental measurements on the melting of fullerenes[2] and with results from molecular dynamics simulations[3].

Two examples of possible defragmentation channels of C_{60} are shown in Figure 1. Channel (a) corresponds to the transition of C_{60} in the gas phase of 30 C_2 molecules, while channel (b) corresponds to the polymerisation of the fullerene.

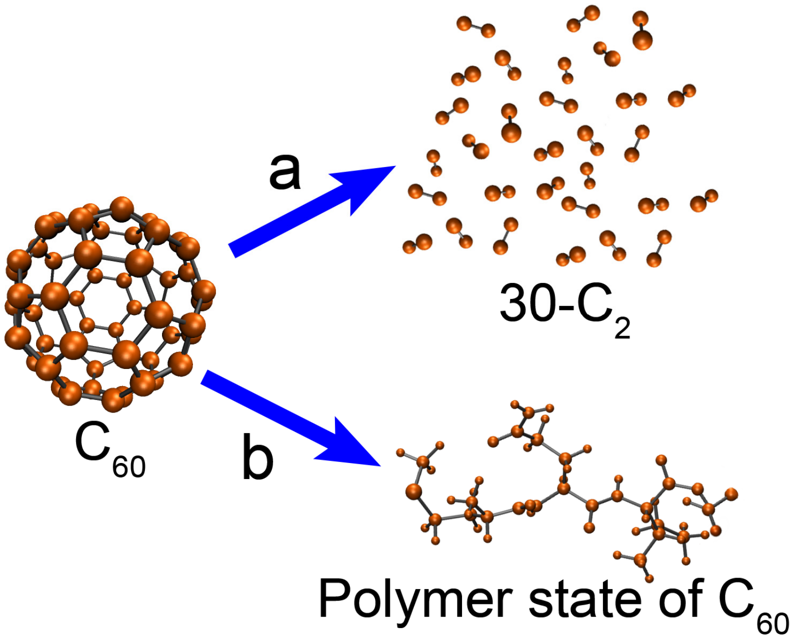

Figure 1. Possible fragmentation channels of C_{60} into a gas or polymer phase.

References
[1] A. V. Yakubovich, I. A. Solov'yov, A. V. Solov'yov and W. Greiner, Eur. Phys. J. D (Highlight paper) 40, 363 (2006); Europhysics News, in print (2007)
[2] Y. Jin, J. Cheng, M. Varma-Nai, G. Liang, Y. Fu, B. Wunderlich, X. Xiang, R. Mostovoy and A. Zetti, J. Phys. Chem. 96, 5151 (1992)
[3] S. G. Kim and D. Tománek, Phys. Rev. Lett. 72, 2418 (1994)

PS-36

Multiple ionisation and fragmentation of C_{60} following excitation with FEL radiation

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The (multiple) ionisation and the subsequent fragmentation of C_{60} was studied with synchrotron radiation and at the new light-source FLASH (**F**ree-**E**lectron **LAS**er in **H**amburg).

The ionisation of C_{60} molecules in the gas phase was studied under single photon excitation condition covering a large photon energy range (26–130 eV) [1, 2]. In these valence ionisation processes C_{60}^{q+} ions with charge states up to 3+ and several smaller C_{60-2n}^{q+} fragments have been observed. Our intention was especially focused on the dynamics of the subsequent fragmentation, which probably proceeds via a subsequent emission of neutral C_2 particles.

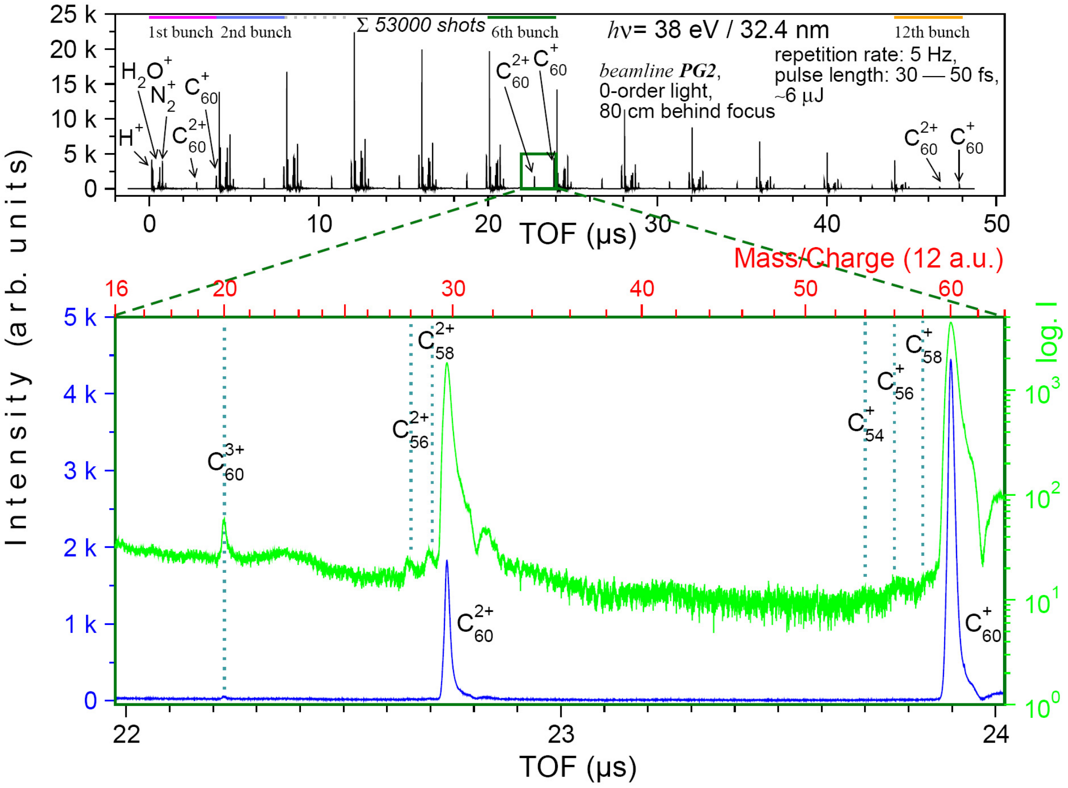

Figure 1: Time-of-flight spectrum recorded at the beamline PG2 at the FEL with a photon energy of 38 eV. The upper part shows a sum over 53000 FEL shots where, besides contributions from rest gases, C_{60} ions can be found. The lower part gives a part of the 6th bunch on a linear and logarithmic scale. Signatures of two-photon processes – C_{60-2n}^{q+} (q=1,2; n=1,2,3) and C_{60}^{3+} – are revealed.

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In addition to 'normal' (non-coincident) electron and ion time-of-flight spectroscopy we investigated this topic in more detail with the help of an electron–ion-coincidence measurement.

Recently experiments were performed at the VUV-FEL's monochromator beamline PG2. Here ions are generated by the FEL radiation ($h\nu$=38 eV, pulse length <50 fs) and analogue signals of micro channel plates from the ion detector were acquired simultaneously by an oscilloscope, each trace shows the full FEL pulse train (Fig. 1). The observation of C_{60} ions can give hints about two-photon processes, since one would expect just intact C_{60}^{q+} ions (q = 1–3) for photon energies of 38 eV under single photon absorption conditions, whereas other traces of fragments, like C_{58}^{q+} or C_{56}^{q+}, require higher energies [1,2]. Indeed the fragments can be found, as shown in Fig. 1, giving evidence for two-photon processes. Moreover, the ions exhibit specific power law dependences on the light intensity, as shown in Fig. 2. An exponent should be higher, if the creation requires more energy and the exponent of ≈2 for C_{60}^{3+} points also to a 2-photon process.

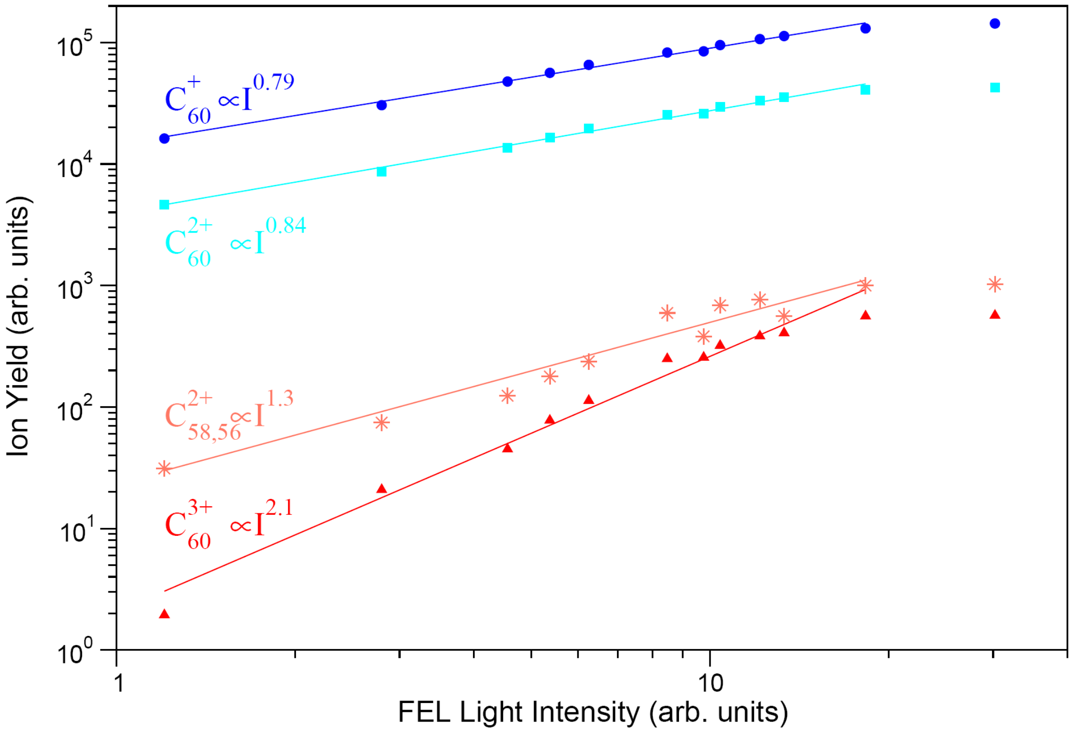

Figure 2: Yield of C_{60} ions and fragments for different FEL light intensities shown on a log.-log. plot. The curves exhibit a power-law dependency before saturation is reached. The exponent of ≈**2** for C_{60}^{3+} underlines the **2**-photon creation process.

References

[1] A. Reinköster, S. Korica, G. Prümper, J. Viefhaus, K. Godehusen, O. Schwarzkopf, M. Mast, U. Becker, *J. Phys. B: At. Mol. Opt. Phys.* **37**, 2135 (2004).

[2] A. Reinköster, S. Korica, B. Langer, G. Prümper, R. Rolles, J. Viefhaus, S. Cvejanović, U. Becker, *J. Electron Spectrosc. and Relat. Phenom.* **144-147**, 151 (2005).

PS-37

DYNAMICAL SCREENING OF AN ATOM INSIDE A FULLERENE OF FINITE WIDTH

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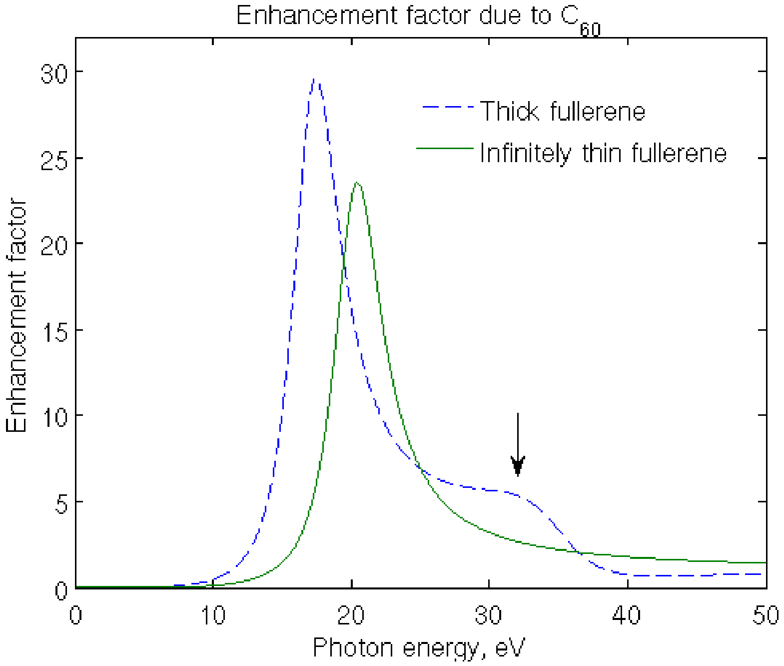
An atom confined within a fullerene is dynamically screened from the external electromagnetic field - it experiences a field that is amplified - leading to an enhancement of the photoabsorption of the confined atom. The amplification occurs in a certain frequency range determined by the plasmon excitation in the confining fullerene. This enhancement factor was discussed by Connerade and Solov'yov in [1] for the case of the infinitely thin fullerene.

The dynamical enhancement factor due to a fullerene of finite thickness is presented here. The existence of a second surface of this fullerene is significant as this allows for the presence of two surface plasmon modes [2], in contrast to the single plasmon considered in [1]. The figure is a plot of the enhancement factor for the infinitely thin fullerene and the fullerene of finite thickness. The profile of the enhancement factor for the thick fullerene has an additional feature [3], which is due to the manifestation of the second plasmon mode, as indicated by the arrow in the figure.



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REFERENCES

[1] J.-P. Connerade & A.V. Solov'yov, *J. Phys. B: At. Mol. Opt. Phys*., **38**, 807 (2005)

[2] A.V. Korol & A.V. Solov’yov, *Phys. Rev. Lett*., **98**, 179601 (2007)

[3] S. Lo, A.V. Korol & A.V. Solov’yov, submitted to *J. Phys. B: At. Mol. Opt. Phys* (2007)

PS-38

EVIDENCE OF TWO PLASMON EXCITATIONS IN FULLERENE PHOTOABSORPTION

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The results of renewed calculations of fullerene plasmon excitations within the jellium model and the Local Density Approximation (LDA) are presented. The account of finite thickness of spherical fullerene layer within the jellium model is shown to lead to the appearance of two surface plasmon resonances in photoabsorption spectrum. Our previous calculations [1] within the LDA, in which the core potential for 240 electrons was created by the uniformly charged thin sphere, revealed only one giant plasmon resonance located at 20 eV.

The renovated fullerene calculations are performed within the self-consistent LDA with the following effective potential:

$$V_{eff}(\vec{r}) = V_{core}(\vec{r}) + \int \frac{n(\vec{r}\,')}{|\vec{r}-\vec{r}\,'|} d\vec{r}\,' + V_{xc}(\vec{r}) + V_{ps}(\vec{r}) \qquad (1)$$

Here $V_{core}(\vec{r})$ is the jellium potential created by 60 positively charged C^{+4} ions on a sphere with radius R=6.66 a.u., the second term describes the Coulomb interaction between electrons. The exchange-correlation potential $V_{xc}(\vec{r})$ is presented by the Perdew and Wang parameterization [2] for density functional. The additional pseudopotential $V_{ps}(r)$ is added here to take into account the finite thickness of fullerene sphere with the following parameterization [3]:

$$V_{ps}(r) = \begin{cases} -V_0, & R-\delta R/2 \le r \le R+\delta R/2 \\ 0, & r < R-\delta R/2;\; r > R+\delta R/2 \end{cases} \qquad (2)$$

The parameters V_0 and δR of this potential are chosen to achieve the agreement with experimental ionization potential and calculated width of valence zone [3]. The inclusion of the potential $V_{ps}(r)$ leads to the stronger mixture of σ- and π- orbitals in the valence zone that increases the role of many-electron correlations.

The obtained single-electron wave functions are used in the calculations of photoionization amplitudes and cross sections within the Random Phase Approximation (RPA). Within the RPA calculations we include the interaction between electrons of 10 outer shells simultaneously. Due to the potential $V_{ps}(r)$ the cross section reveals the double giant resonance structure which may be associated with the electron excitation modes on the external and internal shell surfaces. The first main maximum located at the excitation energy ω ~ 20 eV is the giant plasmon resonance which is well-known and has been found both experimentally and theoretically within different models (see for example [4] and references therein). The second resonance at ~35 eV being smaller than the first one has also the collective nature and appears due to the account of many-electron interactions and double-surface spherical layer.

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The main contribution to the total photoabsorption cross section, in particular to the second resonance, comes from the $l \to l+1$ partial transitions. Fig. 1 shows the typical behavior of number of partial cross sections for σ- orbitals with orbital momentum $l = 7, 8, 9$. Note that when we neglect the pseudopotential (2) in calculations the σ- orbitals do not give the essential contribution to the total cross section.

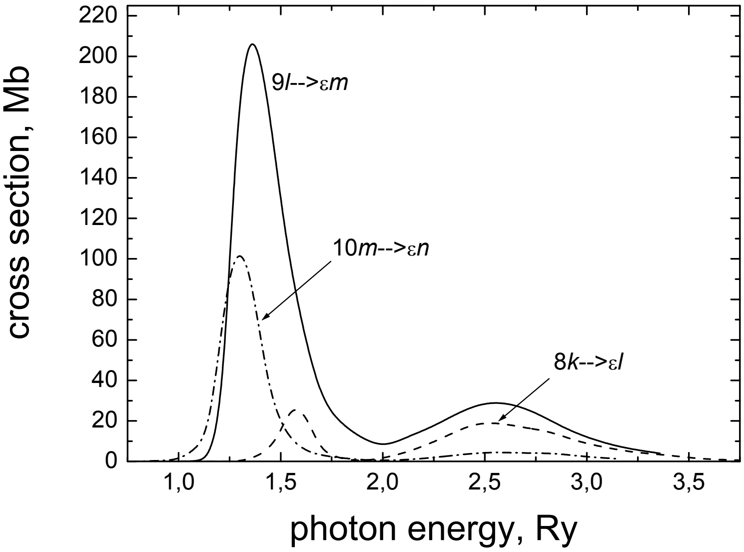

Figure 1: The partial cross sections for $l \to l+1$ transitions from the σ - orbitals of fullerene.

The results of the calculations of fullerene total photoionization cross section will be presented at the conference in comparison with other calculations and experimental photoabsorption cross section.

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REFERENCES

[1] V.K. Ivanov, G.Yu. Kashenock, R.G. Polozkov, A.V. Solov'yov, *J. Phys B* **34**, L669 (2001).
[2] J.P. Perdew, Y. Wang, *Phys. Rev. B* **45**, 13244 (1992).
[3] K. Yabana, G.F. Bertsch, *Phys. Scr.* **48**, 633 (1993).
[4] A.V. Solov'yov, *Int. Journal of Modern Physics B* **19**, 4143 (2005).

ROLE OF SINGLE AND DOUBLE PLASMON EXCITATION IN FAST ION INDUCED MULTIPLE IONIZATION OF C_{60}

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Fullerenes due to its mesoscopic sizes have been proved to be a non-conventional but attractive target for collision studies intermediate between atoms and surfaces. Collision studies of C_{60} with intense lasers, fast beams of electrons and heavy ions have provided greater insights into the structural and collisional aspects. It is known that the C_{60} molecule exhibits a collective mode of excitation of its loosely bound de-localized pi-electrons, called giant dipole plasmon resonance (GDPR). The effect of the GDPR has been predicted in theory and observed in photo absorption studies and intense laser induced ionization of C_{60} [1]. The evidence of the collective excitation and wake-field induced Stark mixing effect on heavy ion-induced x-ray emission in collisions with C_{60} was found [2] which is similar to that observed in ion-solid collisions. The ionization and fragmentation of fullerene in collisions with fast heavy ions have been investigated only in a few cases [3,4,5,6]. The collective excitation is expected to play a major role in the ionization of C_{60} which was first addressed in [3] and a model based on the GDPR excitation was then proposed. The model predicts very weak energy dependence but quite sensitive to the projectile charge state *q*. In order to explore the effect of the GDPR we have, therefore, investigated the single and multiple ionization of C_{60} in collisions with 3.125 MeV/u O ions. The charge state dependence of the ionization cross sections are investigated along with a gaseous target, such as, Ne. The fast heavy ion beams of O-ions were obtained from the 14 MV Pelletron accelerator facility at TIFR, Mumbai. A Wiley-Mclaren type time-of-flight (ToF) mass spectrometer was used to mass and charge state analyze the reaction products. A micro channel plate was used to detect the recoil ions whereas a channel electron multiplier to detect the electrons. We plot the ratios (see Fig. 1) of yields of various recoil C_{60}-ions as a function of recoil ion charge states (R+) and all of these are normalized to single ionization yields (i.e R=1+). It can seen that the ratios of double-to-single ionization yields is about 28.5%. This fraction for triple and quadruple ionizations are 6.5 % and 1.5 %, respectively. These fractions are same for q=7+ and 8+. The observed ratios are compared with the GDPR model prediction for the ratios of double-to-single and triple-to-single plasmon excitation

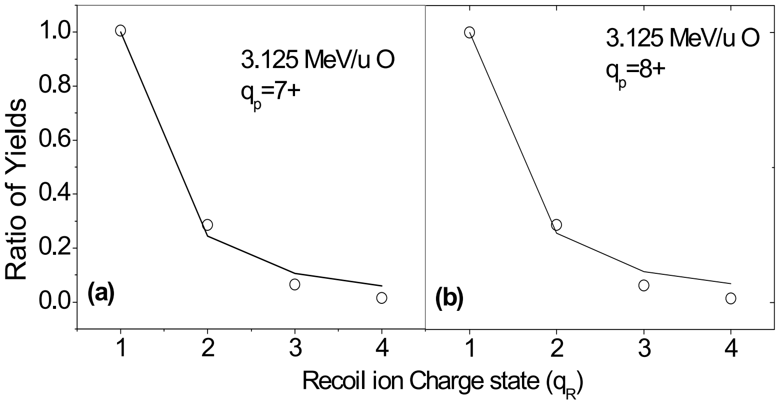

Figure 1: Recoil ion charge state distribution of relative yields of C_{60} ions, normalized to the yields for C_{60}^{1+}. The lines are the GDPR model predictions.

cross sections and so on. Such an agreement is in line with the observations of intense laser based experiment in which one claims that the single and double ionizations are entirely dominated by the single and double plasmon excitation.

Next we compare the yields of the single and double ionization with the prediction of the model. In figure 2, we show the q-dependence of single and double ionization yields. Along with the data we also plot predictions of GDPR model independently normalized to data in each case. The single and double ionizations yields show excellent qualitative agreement with the predictions of GDPR model based on single and double plasmon excitation, respectively. It is to be noted that ionization yields increase linearly with q for both the cases contrary to the expected behaviour for a gaseous target. The figure also shows the comparison with the ionization yields for Ne target. It is obvious that the ionization yields increase with q much faster that that for C_{60}. The triple and quadruple ionization cross sections are found to be almost independent on projectile charge state and that the GDPR model is not adequate to explain the qualitative behavior. This could be due to the complication of the multiple ionization process due to the coupling other low impact parameter collision processes such as fragmentation, evaporation, dynamical screening. However, more recent experiment with Si projectiles (q =6-14) show again a similar linear q-dependence. It is therefore shown that the single and double ionization process is largely dominated by the single and double plasmon excitation. The details can be found in our recent work [9,10] and will be presented.

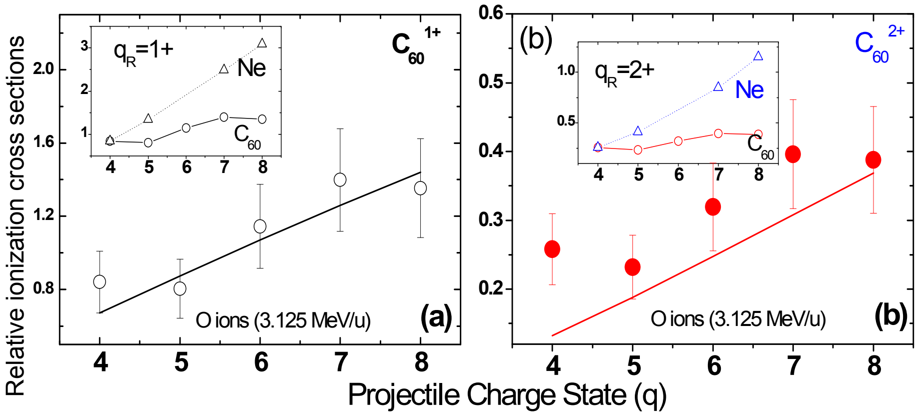

Figure 1: Projectile q dependence for single (a) and double (b) ionizations of C_{60} for 3.125 MeV/u O^{q+} projectile. Solid lines are GDPR cross sections normalized independently to experimental data. Insets in (a) and (b) show single and double ionization for Ne and C_{60} (taken from [9]).

REFERENCES

[1] Hunsche S, *Phys. Rev. Lett.* **77,** 1966 (1996).
[2] U. Kadhane, D. Misra, Y.P. Singh and L.C. Tribedi *Phys. Rev. Lett.* **90** 093401 (2003)
[3] S. Cheng et al. *Phys. Rev. A* **54,** 3182 (1996).
[4] H. Tsuchida et al. *J. Phys. B* **31,** 5383 (1998).
[5] A. Itoh et al. *Phys. Rev. A* **59,** 4428 (1999).
[6]A. Reinkoester et al. *Phys. Rev. A* **64** 023201 (2001).
[9] U. Kadhane, A. Kelkar, D. Misra, A.Kumar, L.C. Tibedi *Phys. Rev. A* **75**, 041201**(R)** (2007).
[10] A. Kelkar, U. Kadhane, D.Misra, A.Kumar, L.C. Tribedi *Nucl. Instr. Meth.* B **256**, 114 (2007).

PS-40

ELECTRON EMISSION SPECTRUM FROM C_{60} IN COLLISIONS WITH 4 MeV/u BARE F IONS AND PLASMON EXCITATION

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Heavy ion atomic collision technique has been proved to be a powerful tool to study the structural and collisional properties of atoms, molecules, solids, surfaces and clusters including fullerenes. The collective excitation, commonly known, as the giant dipole plasmon resonance (GDPR) has been found to play a major role in photo-ionization [1], intense laser based multiphoton ionization [2] and fast-ion ionization [3-7] of C_{60}-fullerene. The fullerenes are ``small" particles with diameter ~10Å over which a large electron density similar to that of a solid foil, is sampled while the single collision condition is satisfied. The GDPR is known to have excitation energy ~20 eV with a width of ~10 eV and integrated oscillator strength 71 [1]. The giant dipole resonance in nuclear reaction is a well known phenomenon. In ion-solid collisions the collective electronic response is known to give rise to a dynamic screening and a wake of electron density fluctuations trailing the ions and a resulting Stark splitting has also been observed in Ly□ x-ray emission yields in e-capture by fast bare ions in solids [8] as well as in fullerene [9]. However, a direct evidence of this process in fast ion-collisions is awaited which could be done by detecting the electrons emitted following the GDPR excitation.

In a typical ion-atom collision the low energy electron emission spectrum is known to carry the signature of different ionization mechanisms such as soft collision, binary collisions, electron capture in continuum, two center effect and newly investigated Young type interference phenomenon [10-12]. In addition to these the collective excitation may also play a major role if the collision partner is solid or cluster including fullerene. In case of collisions involving C_{60}-fullerene the mechanisms of electron emission are governed by even more complicated processes. The double differential electron emission spectrum do have the signatures of single particle behaviour, collective response and inner-shell ionization (apart from characteristic molecular processes). One has find a proper procedure to extract the GDPR contribution from such electron spectrum. We have initiated such experiments in which we detect electrons of energies 1-40 eV for various angles between 20^0 and160^0 in collision of fast (4 MeV/u) bare F-ions with C_{60}-vapour. The ion-beams were obtained from 14 MV tandem Pelletron accelerator and was made to interact with an effusive jet of C_{60} vapour which was produced by heating the C_{60} powder to a temperature of about 450^0-500^0C. The electrons were detected by using an hemispherical electrostatic analyzer which can be rotated between 20^0 and 160^0. The typical DDCS spectrum falls sharply with emitted electron energy. To compare the spectrum with a typical electron spectrum in ion-atom collisions we have also studied the electron emission from He atoms. In addition, we have compared the data with the theoretical spectrum for C-atom in collisions with the same target. The theoretical calculations for C atoms are obtained using the CDW-EIS model. From such a comparison initial results are obtained which indicate that the DDCS spectrum shows broad peak-like structure in the low energy part of the spectrum (between 1-20 eV) arising from the GDPR process. The angular distribution of electrons of different energies were plotted and compared with the similar experimental data for He atoms as well as for the CDW-EIS calculations for the C-atom. The distributions clearly show the trends which are different from that in ion-atom collisions. However a theoretical model explaining the distributions on the basis of collective plasmon excitation is awaited. The details will be provided.

REFERENCES

[1] George F. Bertsch et al. Phys. Rev. Letts. **67**, (1991) 2690.
[2] Hunsche S, *Phys. Rev. Lett.* **77** (1996) 1966.
[3] S. Cheng et al. *Phys. Rev. A* **54** (1996) 3182.
[4] A. Itoh et al. *Phys. Rev. A* **59,** 4428 (1999).
[5] A. Reinkoester et al. *Phys. Rev. A* **64** 023201 (2001).
[6] U. Kadhane, A. Kelkar, D. Misra, A.Kumar, L.C. Tibedi, *Phys. Rev. A,* **75** (2007) 041201**(R)**.
[7] A. Kelkar, U. Kadhane, D.Misra, A.Kumar, L.C. Tribedi *Nucl. Instr. Meth.* B **256**, 114 (2007).
[8] J. P. Rozet et al., Phys. Rev. Lett. **58**, (1987) 337.
[9] U. Kadhane, D. Misra, Y.P. Singh and L.C. Tribedi, *Phys. Rev. Lett.* **90** (2003) 093401.

[10] N. Stolterfoht et al., Phys. Rev. Lett., **87** 023201 (2001).
[11] D. Misra, U. Kadhane, Y.P. Singh, L. C. Tribedi, P.D. Fainstein and P. Richard, *Phys. Rev. Lett*. 92, 153201 (2004).
[12] D. Misra, A. Kelkar, U. Kadhane, Ajay Kumar, L. C. Tribedi, and P.D. Fainstein, *Phys. Rev. A*, Phys. Rev. A **74**, 060701(R) (2006).

PS-41

PHOTOIONIZATION OF THE FULLERENE POSITIVE IONS

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The photoionization cross sections of the fullerene positive ions C_{60}^{q+} q = 1, 2, 3 are calculated within the photon energy range from the ionization thresholds up to 80 eV and compared with the recent experimental data and other calculations for C_{60}^{+} [1], [2]. The main goal is to study the features and changes in giant plasmon excitations along the series from neutral fullerene to its ions. The theoretical framework used in the present work has been developed earlier and applied for the description of photoionzation of C_{60} and C_{60}^{+} [3]. In the present work the theoretical approach is improved by means of using the Perdew-Wang parameterization [4] for the exchange-correlation energy functional.

The total photoionization cross section is calculated as a sum of partial cross sections for each orbital. The ionization amplitude of each orbital is calculated both within the single-electron approximation and also by taking into account many-electron correlations. At the first stage, the frozen core model and the Local Density Approximation (LDA) are used assuming that there is a single electron transition during the process. The correlations between the transitions from different states are taken into account within the Random Phase Approximation (RPA). Single-electronic states of a fullerene ion are calculated within the spherical jellium model with the self-consistent potential [3] determined within LDA. The basic Kohn-Sham equations are used for determining of the single-electron wave functions for both the sphere and the spherical layer. The RPA calculations include the interaction between electrons of the 10 outer shells (six π- orbitals and four σ- orbitals).

The calculated photoionization cross sections for the neutral fullerene, as well as for its ions C_{60}^{+}, C_{60}^{2+} and C_{60}^{3+} indicate the well-known giant plasmon resonances with positions of ~20 eV for all fullerenes. This is in a good agreement with the classical Mie theory and the experimental data [1] for the ions C_{60}^{+}, C_{60}^{2+} and C_{60}^{3+}. Fig. 1 shows the calculated total photoabsorption cross sections for the ions C_{60}^{+} and C_{60}^{2+}. The analysis shows that the dominant contribution in the giant resonance for the total cross sections is due to the ionization from the 6*g* (nearly 50 per cent of the total cross section) and the 5*f* (about 30 per cent) shells with some contribution from the 4*d* and 2*s* shells. Note that the most outer shells (10*m* σ- and 6*h* π- orbitals) practically do not contribute into the giant resonance cross section. The physical nature of the resonances is collective mode of electron excitation and identical for all fullerenes: these giant resonances are due to correlations between σ- and π- orbitals. When the contribution of σ- orbitals is excluded in the calculations then resonance disappears, so the appearance of this resonance is related to σ- electron densities. The photoionization cross sections calculated in the present work fulfill the sum rule for all fullerenes under the consideration, so in contrast to the previous calculations [2] they are not scaled by any factor.

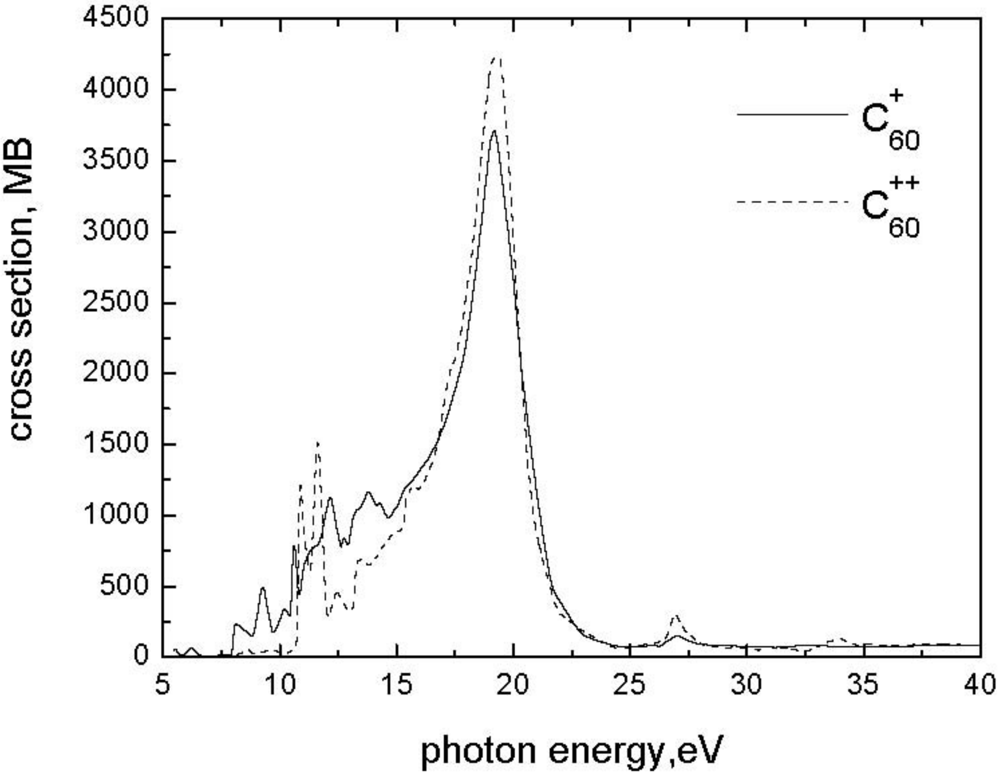

Figure 1: The calculated total photoionization cross sections of the fullerene ions C_{60}^{+} (solid line) and C_{60}^{++} (dashed line).

The comparison with experimental data [1] shows that the position and shape of resonance are in rather good agreement with experiment while the absolute value of cross section differs very significantly. The latter is related to the point that the present calculations give the total photoionization cross section while the experiment measured only one single photoionization channel.

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REFERENCES

[1] S.W.J. Scully et al, *Phys. Rev. Let.t* **94**, 065503 (2005).
[2] R.G. Polozkov, V.K. Ivanov, A.V. Solov'yov, *J. Phys B* **38**, 4341 (2005).
[3] V.K. Ivanov, G.Yu. Kashenock, R.G. Polozkov, A.V. Solov'yov, *J. Phys B* **34**, L669 (2001).
[4] J.P. Perdew, Y. Wang, *Phys. Rev. B* **45**, 13244 (1992).

PS-42

MONO EXCITATION SPECTRA OF CORRELATED C_{60} FULLERENE

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The results of the numerical analysis of inverse spectral problem for mono-photonic electronic spectra of C_{60} fullerene obtained with realistic electron correlation potentials are reported. The spectra of C_{60} are calculated in the framework of extended Hubbard model by a symmetry adapted configuration interaction method [1] using the set of about 200 mono-excitations.

Electron correlation potential in the approach is parameterized by Ohno functional modified by Harigaya and Abe [2] where dielectric screening of the long-distance electron–electron interaction of Coulomb type is introduced via independent parameter. It allows to scale the Hamiltonian persisting only two optimized values: the Hubbard one-center parameter determining the magnitude of overall electron–electron interaction, and screening parameter that defines the contribution of long–range interaction. The both values are optimized for C_{60} fullerene with respect to the consistency of calculated dipole-active transitions with experimental absorption spectra [3] in the spectroscopic range of 2 – 6 eV.

It is shown that simultaneous consistence of transition energies and intensities is of contradiction character. A quantitative criterion and original optimization procedure are proposed taking into account relative importance of both spectral characteristics. From the viewpoints of the obtained results the origin of the low-frequency peak in the C_{60} absorption spectra is discussed. The possible directions of yielding a better consistency are: (i) using more suitable functional form of the electron correlation potential; (ii) including double or higher excitations in the configuration interaction schema; (iii) modification of one-electron part of the model Hamiltonian.

REFERENCES

[1] V.A. Kuprievich, O.L. Kapitanchuk, O.V. Shramko, *Mol. Cryst. and Liq. Cryst.* **427**, 23/[335] (2005).
[2] K. Harigaya, S. Abe, *Phys. Rev. B.* **49**, 16746 (1994).
[3] S.L.Ren, Y.Wang, A.M.Rao *et al.*, *Appl. Phys. Lett.* **59**, 2678 (1991).

PS-43

PHASE DEPENDENT DIFFERENCES IN THE PARTIAL CROSS SECTION OSCILLATIONS OF C_{70}

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We have performed high resolution measurements of emitted photoelectrons from the valence shell of C_{70}, in the gas phase but also in an amorphous solid formation, in order to derive branching ratio and partial cross sections of the two highest molecular orbitals HOMO and HOMO-1. The experiments have been performed at the beamline BW3 at HASYLAB (DESY) using monochromatized synchrotron radiation and time-of-flight (TOF) detection of photoelectrons. The comparison between the two phases shows an interesting and unexpected difference. The ratio between the corresponding HOMO and HOMO-1 levels, which exhibits photon energy dependent oscillations in both cases, shows an offset of 0.5 for the gas phase measurements compared to the solid state data. In contrast to the case of C_{60}[1], the latter one are constantly lower for all energies between 25 and 270 eV. Theoretical calculations performed within the LCAO approximation show good agreement for both data sets, if one assumes, that in the gas phase the (+16a′)-orbital is unoccupied. This behavior is attributed to the small orbital shift upon adsorption.

REFERENCES

[1] Sanja Korica *et al.*, *Physical Review* A **71**, 13203 (2005)

PS-44

JAHN-TELLER SPLITTING OF MULTI-ELECTRONIC STATES IN C60 FULLERENE ANIONS

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Fullerene C_{60} is considered as an ideal building block for novel molecular devices due to its nearly spherical symmetry and high affinity. Strong electron–lattice interaction and high icosahedral symmetry are thought to play a key role in the formation of the physical properties of alkali metal fullerides A_NC_{60}, which range from insulating to metallic and even to superconducting [1]. In these compounds the fullerenes are in multi-charged anionic forms C_{60}^{-N} with N extra electrons, which partially fill the threefold degenerate lowest vacant molecular orbital t_{1u}. The coupling of the degenerate electronic states with certain vibrational modes of the molecule leads to a lowering of the total energy because of spontaneous molecular distortion appearing in Jahn-Teller effect [2].

We report on the results of theoretical study of static Jahn-Teller ground state splittings in icosahedral C_{60}^{-N} (N=1, 2, 3, 4) fullerene anions with different parameterizations of electron-electron interaction [3]. Multi-electronic states of anions were determined within a quasy-π-electron model by the symmetry adapted configuration interaction method for two active spaces of t_{1u} and t_{1u}+t_{1g} molecular orbitals [4]. Electron–lattice interaction is taken into account in a framework of Su–Schrieffer–Heeger model by linear dependence of the neighbor site resonance integrals on the corresponding length change. The calculations are performed with electron correlation potentials of two types: long-range Ohno and short-range Hubbard potentials. Contrary to the case of short-range electron repulsion the inclusion of long-range correlation can essentially change the features of the Jahn–Teller splittings. The influence of the active space expansion to t_{1u}+t_{1g} on long-range electron repulsion is shown as a change of ground state multiplicity in distorted fullerene dianion. The correlation effectively damps electron–lattice interaction favoring the electron delocalization thus destabilizing deformations. The obtained results confirm the Jahn–Teller nature of anions with charges of –2 and –4 providing good agreement with known theoretical [2] and experimental [5] data.

REFERENCES

[1] O. Gunnarsson, *Rev. Mod. Phys.* **69**, 575 (1997).
[2] C.C. Chancey, M.C.M. O'Brien, The Jahn-Teller effect in C_{60} and other icosahedral complexes. New Jersey: Princeton univ. press, (1997).
[3] O.L. Kapitanchuk, V.A. Kuprievich, *Mol. Cryst. and Liq. Cryst.* **426**, 89 (2005).
[4] V.A. Kuprievich, O.L. Kapitanchuk, O.V. Shramko, *Mol. Cryst. and Liq. Cryst.* **427**, 23/[335] (2005).
[5] A. Wachowiak, R. Yamachika, K.H. Khoo *et al.*, *Science* **310**, 468 (2005).

PS-45

KINETIC MODEL FOR NANOTUBE GROWTH

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Size selected nanoclusters of such metals as Ni, Fe, Co are widely used as catalysts in the chemical vapor deposition (CVD) techniques of production of carbon nanotubes [1]. The characteristics of the nanotubes as well as the parameters of their growth are closely related to the size and properties of the catalytic particles [1]. Despite of intensive studies the physical mechanism responsible for the catalytic action of the nanoclusters leading to the nanotubes growth is not yet known.

We have developed a kinetic model describing the process of catalytically assisted growth of carbon nanotubes. The model describes the flow of carbon atoms starting from the catalytic decomposition of the feedstock molecules at the surface of the nanocluster and ending at their embedding into the nanotube. In the figure we show a cross section of the nanocluster and the nanotube with the schematic representation of the carbon flows which are accounted for by the model.

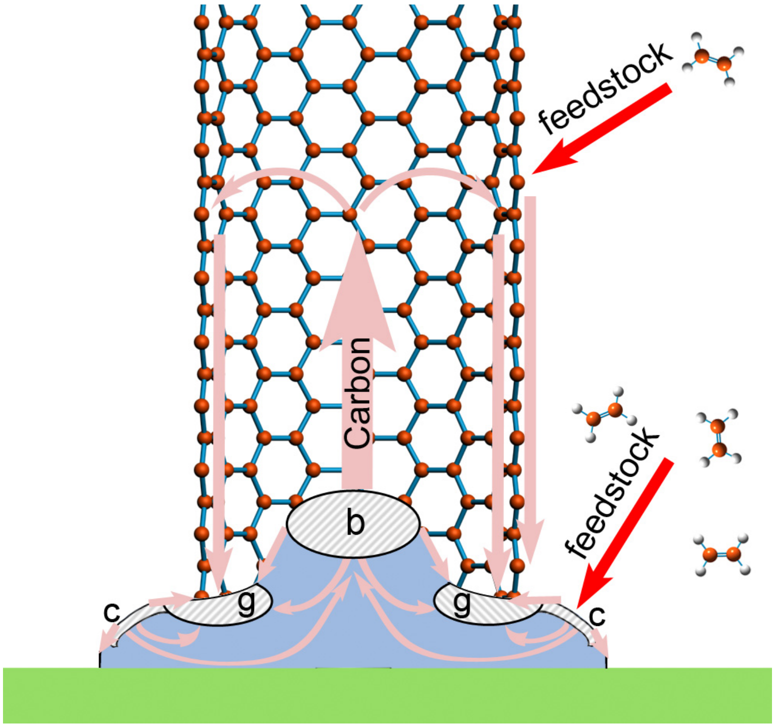

Figure 1: A cross section of the catalytic nanoparticle and the nanotube with the schematic representation of the carbon flows.

Feedstock molecules are decomposed on the catalytically active part of the nanocluster surface, which is exposed to the environment and not screened by any additional inactive layers (region “c”). The carbon atoms from this region diffuse (either along the surface or through the bulk of the cluster) into the other parts of the cluster. We single out the growth region from which the atoms embed themselves into the nanotube walls (region “g”) and the near-surface region inside the nanotube (region “b”). From the region “b” carbon atoms can reach the growth region by surface or bulk diffusion or they can evaporate. Some of the evaporated atoms are deposited on the nanotube walls and reach the growth region by surface diffusion. We also account for the possibility of decomposition of the feedstock molecules directly by the nanotube walls.

Thus, we present a model comprehensively describing the kinetics of the nanotube growth. The model allows one to predict with a reasonable accuracy the distribution of carbon within the catalytic nanoparticle, the relative importance of various carbon fluxes, the nanotube growth rate, etc. The diffusion coefficients, activation energies for embedding into the nanotube, etc. are the input parameters for the model thus allowing one to test various ideas on mechanisms and scenarios of nanotube growth.

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REFERENCES

[1] M. Chhowalla et al., *J. Appl. Phys.* **90**, 5308 (2001) and references therein

PS-46

SYSTEMATIC STUDY ON THE STRUCTURE AND ENERGETICS OF SINGLE WALLED CARBON NANOTUBES OF DIFFERENT CHIRALITIES

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We systematically study carbon nanotubes of various chiralities and their structures and stabilities to understand how the chirality influences the binding energy of nanotubes as well as the structural details like average bond length. Both *ab initio* and molecular mechanics methods are used for the investigation. The *ab initio* density functional theory (DFT) calculations are performed with the use of Gaussian 03 software package [1] and the molecular mechanics calculations are performed with the use of NAMD software package [2]. The results obtained through these studies are used for a better understanding of the growth mechanism of carbon nanotubes [3, 4]. The mechanism which controls the growth of a nanotube of desired chirality is poorly understood. Our studies can be used for the theoretical investigation of the dependence of growth of carbon nanotube on its chirality.

The following figure explains the concept of the chirality of a nanotube by depicting how a nanotube of certain chirality, characterized by two integer numbers n and m, can be constructed from a graphite sheet. The figure shows the structure of a chiral nanotube, corresponding to n=7 and m=4, being a typical conductive nanotube.

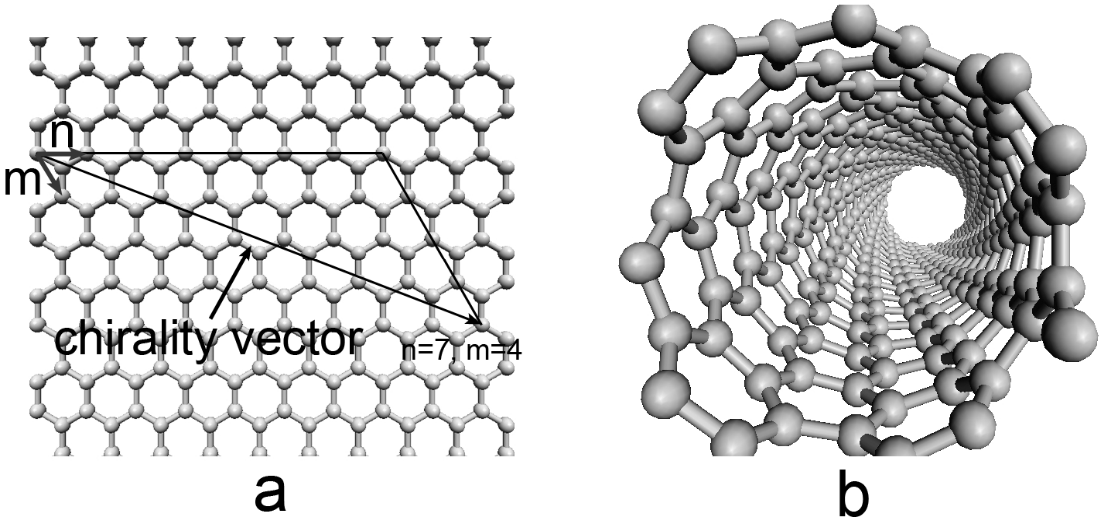

Figure 1: (a) Chirality vector shown in a graphite sheet; (b) The nanotube formed after folding the graphite sheet in figure-(a) along the chirality vector.

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REFERENCES

[1] Gaussian 03, Revision C.02, M. J. Frisch, *et al*, Gaussian, Inc., Wallingford CT (2004)
[2] J.C. Phillips, *et al*, J. of Comp. Chem., 26, 1781 (2005)
[3] Feng Ding, Kim Bolton and Arne Rosen, J. Phys. Chem. B 108, 17369 (2004)
[4] J. Gavillet, *et al*, Phys. Rev. Lett., 87, 27 (2001)

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