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Editorial

Dynamics of systems on the nanoscale*

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Abstract. Various aspects of the structure formation and dynamics of animate and inanimate matter on the nanoscale is a highly interdisciplinary field of rapidly emerging research interest by both experimentalists and theorists. The International Conference on Dynamics of Systems on the Nanoscale (DySoN) is the premier forum to present cutting-edge research in this field. It was established in 2010 and the most recent conference was held in Bad Ems, Germany in October of 2016. This Topical Issue presents original research results from some of the participants, who attended this conference.

1 Introduction

This Topical Issue aims at highlighting the interdisciplinary research topics that elucidate the structure formation and dynamics of animate and inanimate matter on the nanometre scale. There are many examples of complex many-body systems of micro- and nanometre scale size exhibiting unique features, properties and functions. These systems may have very different nature and origin, e.g. atomic and molecular clusters, nanostructures, ensembles of nanoparticles, nanomaterials, biomolecules, biomolecular and mesoscopic systems. A detailed understanding of the structure and dynamics of these systems on the nanometre scale is a difficult and fundamental task, the solution of which is necessary in numerous applications of nano- and bio-technology, material science and medicine.

Although mesoscopic, nano- and biomolecular systems differ in their nature and origin, a number of fundamental problems are common to all of them: What are the underlying principles of self-organization and self-assembly of matter at the micro- and nanoscale? Are these principles classical or quantum? How does function emerge at the nano- and mesoscale in systems with different origins? Which criteria govern the stability of these systems? How do their properties change as a function of size and composition? How are their properties altered by their environment? Seeking answers to these questions is at the core of a new interdisciplinary field that lies at the intersection of physics, chemistry and biology, a field now entitled Meso-Bio-Nano (MBN) Science.

This research field bundles up several traditional topics in theoretical physics under a common theme. The range of open challenging scientific problems (topical areas) in the field is very broad. They may include:

- structure and dynamics of clusters, nanoparticles, biomolecules and many other nanoscopic and mesoscopic systems;
- clustering, self-organisation, growth and structure-formation processes and their multiscale nature;
- assemblies of clusters/nanoparticles and biomacromolecules, hybrid bio-nano systems, nanostructured materials;
- surface phenomena;
- nanoscale phase and morphological transitions;
- thermal, optical and magnetic properties;
- collective or many-body phenomena;
- electron transport and molecular electronics;
- collisional, fusion, fission and fragmentation processes;
- particle propagation through a medium;
- radiation effects;
- radiobiological effects.

There are many important applications closely linked to the field. The list of topical areas in the field grows rather rapidly facilitating also the development of the relevant theoretical and computational methods.

The articles in this Topical Issue provide a snap shot of the current research activities (experimental, theoretical, applied) in the field of MBN science. Particular attention is be devoted to dynamical phenomena and many-body effects taking place in various MBN systems on the nanoscale, which include problems of structure formation, fusion and fission, collision and fragmentation, surfaces and interfaces, collective electron excitations, reactivity, nanoscale phase and morphological transitions, irradiation driven transformations of complex molecular systems, biodamage, channelling phenomena. The contributions to this issue represent the studies both at the fundamental level of elementary mechanisms and at the

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more applied level which is necessary in numerous applications of nano- and bio-technology, material science and medicine.

The premier conference in this field, the International Conference "Dynamics of Systems on the Nanoscale" (DySoN) started in Rome, Italy in 2010. The DySoN conference was built upon a series of International Symposia "Atomic Cluster Collisions: structure and dynamics from the nuclear to the biological scale (ISACC)" (see www.isacc-portal.org). During these meetings it has become clear that there is a need for an interdisciplinary conference covering a broader range of topics than just atomic cluster collisions, related to the Dynamics of Systems on the Nanoscale. Therefore, in 2010 the ISACC International Advisory Committee decided to launch the DySoN conference series. The second conference was held in St. Petersburg, Russia in 2012 (see Ref. [1] for the collection of contributions); the third one took place in Edinburgh, UK in 2014.

DySoN 2016, the fourth conference in the series, was held in Bad Ems, Germany in October 2016 bringing together a number of scientists from around the world. Also, the conference highlighted the breakthroughs achieved within the currently running COST Action CM1301 CELINA "Chemistry for ELectron-Initiated Nanolithography" and the project FP7-ITN-ARGENT-608163 – "Advanced Radiotherapy, Generated by Exploiting Nanoprocesses and Technologies". The latter project inherited and extended the scopes of the recently ended COST Action "Nanoscale insights into ion-beam cancer therapy" (Nano-IBCT) towards the understanding of nanoparticle impacts on biological systems and related biomedical applications. Therefore, DySoN 2016 continued traditions of the earlier Nano-IBCT Conference series. Also, the mini-workshop "Periodically bent crystals for crystalline undulators" held within the HORIZON 2020 RISE-PEARL-690991 project was linked to DySoN 2016. The research areas represented by the mentioned European projects overlap strongly with the Topical Areas of the DySoN Conference.

Below we briefly characterize the contributions to this Topical Issue made by the participants of the DySoN 2016 Conference. When doing this, we thematically attribute the contributions to some of the Topical Areas listed above.

1.1 Structure and dynamics of clusters, nanoparticles, biomolecules.

Several papers in the Topical Issue are devoted to recent advances in the understanding of structure and essential dynamic properties of selected cluster systems, nanostructures and biological systems. Both theoretical and experimental aspects are presented.

Results of experimental study of multi-photon ionization of thymine-water clusters in the gas phase are presented and discussed in reference [2]. Metastable H_2O loss from $T^+(H_2O)_n$ was observed at $n \geq 3$ only. Ab initio quantum-chemical calculations of a large range of optimized $T^+(H_2O)_n$ conformers have been performed, enabling binding energies of water to be derived. These decrease smoothly with n, consistent with the general trend of increasing metastable H_2O loss in the experimental data. The lowest-energy conformers of $T^+(H_2O)_3$ and $T^+(H_2O)_4$ feature intermolecular bonding via charge–dipole interactions, in contrast with the purely hydrogen-bonded neutrals. We found no evidence for a closed hydration shell at $n = 4$, also contrasting with studies of neutral clusters.

The impact of dissipation on the energy balance in the electron dynamics of metal clusters excited by strong electro-magnetic pulses is investigated theoretically in reference [3] by means of Time-Dependent Density-Functional Theory (TDDFT). Separate contributions to the total excitation energy, namely, (i) energy exported by electron emission, (ii) potential energy due to changing charge state, (iii) intrinsic kinetic and potential energy, and (iv) collective flow energy are evaluated. The balance of these energies is studied as function of the laser parameters (frequency, intensity, pulse length) and of the system size and charge. For collisions with a highly charged ion the dependence on the impact parameter has also been considered. Dissipation turns out to be small where direct electron emission prevails namely for laser frequencies above any ionization threshold and for slow electron extraction in distant collisions. Dissipation is large for fast collisions and at low laser frequencies, particularly at resonances.

The use of an extrinsic fluorophore (rhodamine B isothiocyanate) as a versatile probe to measure rotational motions of proteins is discussed in reference [4]. To illustrate the usefulness of this probe, the authors describe the fluorescence anisotropy values of this fluorophore covalently linked to myoglobin protein measured in aqueous solutions of increased methanol content. Methanol-induced unfolding is revealed by the transition from constrained to free rotation of the covalently attached rhodamine B fluorophore.

Reference [5] discussed the scenario when a UV-induced damage in DNA is repaired by a specific enzyme, called (64) photolyase, which binds itself to the damaged sites. By means of molecular dynamics simulations the binding between photolyase and the DNA have been described revealing the importance of several charged amino acid residues in the enzyme, such as arginines and lysines. Especially R421 is crucial, as it keeps the DNA strands at the damaged site inside the repair pocket of the enzyme separated. DNA photolyase is structurally highly homologous to a protein called cryptochrome. Both proteins are biologically activated similarly, namely through flavin co-factor photoexcitation. It is, however, striking that cryptochrome cannot repair UV-damaged DNA. The present investigation allowed authors to conclude on the small but, apparently, critical differences between photolyase and cryptochrome.

The high-resolution vacuum ultraviolet photoabsorption spectrum of isobutyl acetate, $C_6H_{12}O_2$, measured over the energy range 4.3–10.8 eV, is presented in [6]. Valence and Rydberg transitions with their associated vibronic series have been observed and are assigned in accordance with new ab initio calculations of the excitation energies and oscillator strengths. The measured

photoabsorption cross sections have been used to calculate the photolysis lifetime of this ester in the Earth's upper atmosphere (20–50 km). Calculations have also been carried out to determine the ionization energies and fine structure of the lowest ionic state of isobutyl acetate and are compared with a photoelectron spectrum (from 9.5 to 16.7 eV), recorded for the first time.

1.2 Collisional, fusion, fission and fragmentation processes

Several papers in this issue are devoted to theoretical, computational and/or experimental studies of various phenomena occurring in collision processes involving complex systems.

The results of classical molecular dynamics simulations (MD) of collision-induced fusion and fragmentation of C_{60} fullerenes are presented and discussed in reference [7]. The simulations were performed by means of the MBN Explorer software package [8,9]. The analysis of fragmentation dynamics at different initial conditions shows that the size distributions of produced molecular fragments are peaked for dimers, which is in agreement with a well-established mechanism of C_{60} fragmentation via preferential C_2 emission. The results are compared with experimental time-of-flight distributions of molecular fragments and with earlier theoretical studies. Considering the widely explored case study of C_{60}–C_{60} collisions, broad capabilities of the MBN Explorer software were demonstrated, which can be utilized for studying collisions of a broad variety of nanoscale and biomolecular systems by means of classical MD.

The framework of classical MD with reactive potential was applied in reference [10] to model the process of water molecules evaporation from high-velocity argon atoms impinging on protonated water clusters. The analysis of the simulation data revealed the conditions under which a specific number of molecular evaporation events is found one nanosecond after impact. These simulations provide velocity distributions that exhibit two main features, with a major statistical component arising from a global redistribution of the collision energy into intermolecular degrees of freedom, and another minor but non-ergodic feature at high velocities. These discoveries are consistent with recent experimental measurements and confirm that electronic processes are not explicitly needed to explain the observed non-ergodic behavior.

An instability of a liquid droplet traversed by an energetic ion was explored in reference [11]. This instability is brought about by the predicted shock wave induced by the ion [12]. An observation of multifragmentation of small droplets traversed by ions with high linear energy transfer is suggested to demonstrate the existence of shock waves. A number of effects are analysed in effort to find the conditions for such an experiment to be signifying. The presence of shock waves crucially affects the scenario of radiation damage with ions since the shock waves significantly contribute to the thermomechanical damage of biomolecules as well as the transport of reactive species.

A computational analysis for the interaction between the vibrating/rotating molecular ions H_2^+, HD^+, D_2^+ colliding with He atoms is presented in reference [13]. The work describes the calculation of the 3D interaction potentials and of the ionic vibrational levels needed to obtain the vibrational coupling potential matrix elements which are needed in the multichannel treatment of the rovibrationally inelastic collision dynamics. The general features of such coupling potential terms are discussed in view of their employment within a quantum dynamical modeling of the relaxation processes, as well as in connection with their dependence on the initial and final vibrational levels which are directly coupled by the present potentials. As a preliminary test of the potential effects on the observables, calculations are performed for the collision $H_2^+ + He$ at the energies of an ion-trap by using either the rigid rotor approximation or a more accurate vibrationally averaged description for the $v = 0$ state of the target.

1.3 Radiobiological effects

Papers [14–17] are devoted to the analysis of the structural changes and damage of biological objects exposed to ionizing radiation.

Reference [14] addresses the problem of variable cell survival probability along the spread-out Bragg peak which is highly important for planning and optimisation of ion-beam therapy. In the cited paper the problem is considered using the multiscale approach [18,19] to the physics of ion-beam therapy. The physical reasons for this problem are analysed and understood on a quantitative level. A recipe of solution to this problem is suggested which can be used in the design of a novel treatment planning and optimisation based on fundamental science.

Paper [15] presents the advances in quantitative description of the damage caused by the shock wave propagating radially from the ion path [12]. In contrast to earlier works, which were based on the assumption of a uniform energy deposition in thin cylinders around the path, the present work uses the radial dose distributions, calculated by solving the diffusion equation for the low energy electrons and complemented with a semi-empirical inclusion of more energetic δ-electrons. The effect of these energy distributions vs. stepwise energy distributions in tracks on the strength of shock waves induced by carbon ions both in the Bragg peak region and out of it is studied by molecular dynamics simulations.

Experimental data accompanied by quantitative analysis of chromosome aberrations in human lymphocytes exposed to different doses of particle radiation are presented in reference [16]. Distributions of aberration frequency and the shape of dose–response curves for the total aberration yield as well as for exchange and non-exchange aberrations were investigated. Applying the linear-quadratic model, a relation was derived between the fitted parameters and the ion track radius which could explain experimentally observed curvature of the dose–response curves.

In reference [17] the authors investigate the impact of radiation-induced damages on the change the electrical impedance of a DNA molecule. The damage due to ionising radiation is shown to have a direct effect on the

electrical transport properties of DNA. Impedance measurements of DNA samples were carried out by an AC impedance spectrometer before, during and after irradiation. The impedance of all DNA samples exhibited rising capacitive behaviour with increasing absorbed dose.

1.4 Channeling and radiation effects

Contributions [20] and [21] report on the recent advances in theoretical and experimental studies of the radiation produced by ultra-relativistic electrons in periodically bent crystals, known as crystalline undulators (CU) [22]. This field of research is very promising from the viewpoint of creating a new light source in the sub-Angstrom range of radiation wavelength.

Results of numerical simulations of the channeling properties and emission spectra by 855 MeV electrons in periodically bent diamond crystal are presented and discussed in reference [20]. The periodicity of bending profiles enhances the rate of re-channeling events and, as a result, leads to distinct lines in the spectra. The results obtained are analyzed and matched to the properties of the planar channeling in straight and uniformly bent crystals. Suggestions are made towards the experimental studies of the predicted phenomena.

Reference [21] reports on experimental evidence of the crystalline undulator radiation which was detected in channeling experiments with 855 MeV electrons carried out at the MAinzer MIcrotron (MAMI) facility. A significant increase in enhancement of the undulator peak was achieved by measuring the radiation emitted in the nearly forward direction, as it was predicted by earlier calculations.

Another type of phenomenon related to the light propagation in matter is discussed in reference [23]. The effect predicted is in a giant asymmetry in the transmission of light propagating through a linear nonmagnetic optical system consisting of a nanohole in metal film deposited on the surface of a one-dimensional photonic crystal. The reasons for the asymmetry are analyzed and discussed. It is shown that by means of this effect it is possible to realize an optical diode whose reverse transmission can be suppressed by a factor of more than 30 000.

1.5 Hybrid bio-nano systems, nanostructured materials

Reference [24] reviews the results obtained so far in experimental and theoretical studies of the spontelectric state of matter. The latter is exemplified by the presence of static, spontaneous electric fields extending throughout thin films of dipolar solids. The spontelectric state was discovered using a low energy electron beam technique, using the ASTRID storage ring at Aarhus University. Following a resume of the characteristics and of a model for the spontelectric effect, a description is given of the counter-intuitive behaviour of fields in films of methyl formate as a function of deposition temperature, T. It is found that films for $T \leq 77.5$ K show the expected decrease in the field with increasing T but, for $T \geq 77.5$ K, an increase in the field for higher T is revealed. Analysis of these results illustrates the non-linear and non-local characteristics of the spontelectric state. Recently it has been shown that Reflection-Absorption Infrared Spectroscopy (RAIRS) provides a new and independent technique for the detection of the spontelectric effect, through the observation of vibrational Stark shifts in spectra of films. Stark shifts for nitrous oxide are demonstrated to be in harmony with electric fields measured using the electron beam technique. The method is then applied to carbon monoxide, showing that this material displays the spontelectric effect between deposition temperatures of 20 K and 26 K.

In reference [25], the electronic structure and core-to-valence transitions in bone tissue are examined within the framework of the morphological 3DSL model. This model takes into account (i) structural and functional organization of the skeleton in the normal and pathological conditions, and (ii) peculiarities of electron wave propagation in a 3D superlattice of "black-nanocrystallites-in-muddy-waters". The research is focused on the HAP-to-bone red shifts of core-to-valence transitions near Ca and P $2p$ and O $1s$ edges in single-crystal hydroxyapatite (HAP) $Ca_{10}(PO_4)_6(OH)_2$. The origin of the shift is discussed and the extended comparative analysis of the experimental data is performed. This hierarchy effect on the core-to-valence transition energies is regarded as a promising tool for medical imaging and perspective pathway for nanodiagnostics of mineralized bone.

1.6 Surface phenomena

Results of experimental and theoretical studies of the structures formation and stability on the surface and of the surface diffusion processes are presented in contributions [26] and [27].

Thermal behavior of Pt_{10} and Pt_1 bound to a silicon substrate prepared by the impact of size-selected Pt cluster ions at 1 eV per Pt atom is investigated in reference [26]. The clusters height and diameter were obtained by statistical analysis of their images by means of scanning-tunneling microscopy. The Pt_{10} are stably bound to the Si surface as monatomic-layered $Pt_{10}Si_x$ disks with insertion of silicon atoms into the clusters at the moment of the impact. The thermal stability of the $Pt_{10}Si_x$ disks is comparable to that of a platinum thin film on a silicon substrate, but inferior to that of the Pt_{30} disks. It was found that more Si atoms start to diffuse into a $Pt_{10}Si_x$ disk between 623 and 673 K, but not into a Pt_{30} disk due to a higher barrier for the Si insertion into this close-packed cluster disk.

The surface diffusion processes of Ni, Pd and Au structures with diameters in the range of a few nanometers is studied in reference [27] by means of the developed kinetic Monte Carlo approach on a coarse-grained lattice. Intensity information obtained via 2D transmission electron microscopy imaging techniques is used to create three-dimensional structure models as input for a cellular automaton. A series of update rules based on reaction kinetics is defined to allow for a stepwise evolution in time with the aim to simulate surface diffusion phenomena. The

material flow, represented by the hopping of discrete portions of metal on a given grid, is driven by the attempt to minimize the surface energy.

2 Conference discussion

The MBN science is highly interdisciplinary field of research studying structure-formation and dynamics of animate and inanimate matter on the nano- and the mesoscales. Strictly speaking, the mesoscale is a challenge to define, because it is not an exact size range, as there is no "mesometer". Often, mesoscopic systems are considered as being larger than atomic, molecular and nanosystems, but smaller than a bulk, and thus having a finite size. During the DySoN 2016 Conference the Discussion Session was organized aimed at exchanging ideas, opinions, concerns, etc. expressed by specialists from different fields on the further development of the most important directions in MBN science.

Contribution [28] to this Topical Issue reflects the spirit and the atmosphere of this Session. It seeks for the answer to a seemingly simple question: what are new size-dependent effects characteristic of the nanoscale range which justify considering dynamics on the nanoscale' as a scientific area in its own right rather than merely an extension of existing knowledge to a different parameter range. A preliminary list of such effects is drawn up. It is shown to include many challenging unsolved problems as well as a number of fundamental investigations which are more or less advanced in development at the present time.

3 Concluding remarks

To conclude the Editorial, let us mention a very important issue which concerns tools needed for a systematic approach to field of research embraced by MBN Science. These tools, apart from theoretical models and experimental studies, also include computational aspects. No surprise that MBN Science, as a research field, emerged only recently from, primarily, atomic, molecular and cluster physics together with development of powerful computers and advanced computational techniques. In particular, the multiscale modeling of MBN systems is one of the hot topics of the modern theoretical and computational research. To fully understand and exploit all the richness and complexity of the MBN-world, especially its all-atom dynamics, one needs to consult many disciplines ranging from physics and chemistry to material and life science, exploiting technologies from software engineering and high performance computing. This general trend brought up the idea and then development of MBN Explorer [8,9] and MBN Studio [29]. These software packages have been designed to become a powerful and universal instrument of computational research in the field of MBN Science, which should play a role of a "virtual microscope" and a "camera" capable to explore, simulate, record and visualise both structure and dynamics of the MBN-world at the atomistic level of accuraca, reproduce its known features and predict the new ones. In this Topical Issue, the contributions [7,11,15,20] provide several case studies conducted with these packages. To be noted that these universal and powerful software packages are fully applicable to a broad variety of the topical areas of the MBN Science listed above in the introductory part of the Editorial.

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