# ORAL PRESENTATIONS

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# SHIM2012

IC-T-01

# The stopping of light ions in solids

Néstor R. Arista

*División Colisiones Atómicas*

*Centro Atómico Bariloche and Instituto Balseiro, S.C. Bariloche, Argentina*

This tutorial will include a review of several developments, current directions and questions related the phenomenon of energy loss of light ions in solids. The topics to be discussed include the following: (a) review of various basic models for the electronic stopping mechanisms: linear and perturbative models, self-consistent and non-linear models, Density Functional method, and related experimental results. (b) charge state effects, partial stopping powers and related issues; protons, antiprotons, positrons and muons; experimental evidences. (c) band structure and threshold effects at low energies: metals and insulators; experiments and models. What have we learned in the last years?.

* email: arista@cab.cnea.gov.ar

# Modeling of HCI-surface interaction

Christoph Lemell*

*Institute for Theoretical Physics, Vienna University of Technology, Wiedner Hauptstr. 8-10, A-1040 Vienna, Austria EU*

As a true many particle problem, the simulation of the interaction of highly charged ions with solid surfaces poses big challenges to theory. A large variety of simulation methods ranging from classical to full ab-initio calculations have to be combined to model experiments in this field.

Starting from experimental results I will work out prototypical pathways to the final simulation results and present details about the methods used. Emphasis will be put on understanding the advantages but, even more important, the limitations of the various methods.

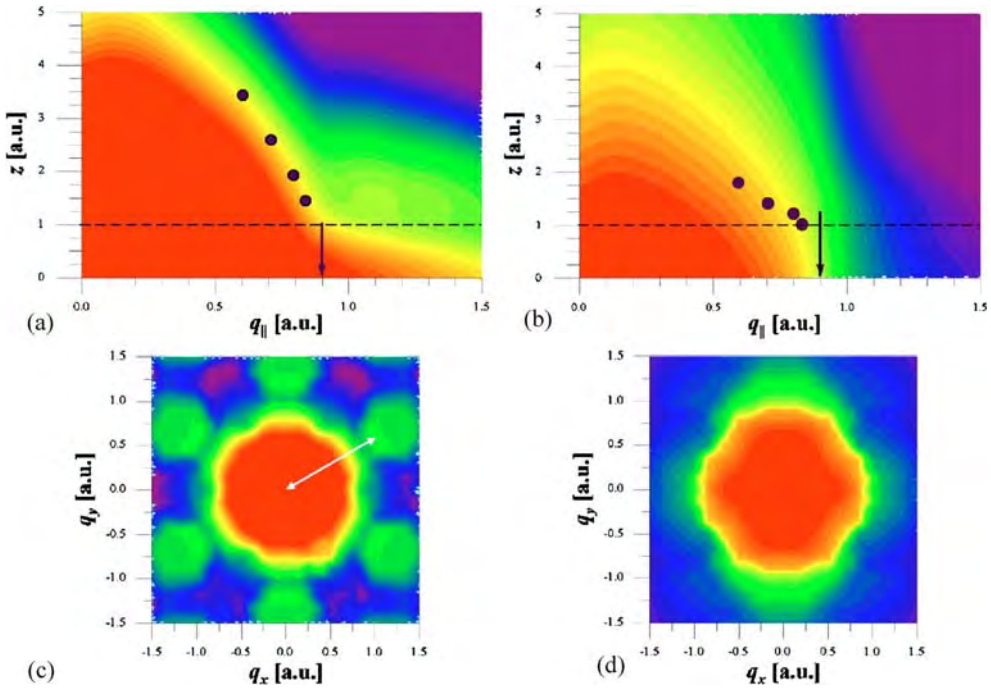


Figure 1: momentum distributions as a function of the distance from the topmost atomic layer of an Al(111) (left panels and an Al(110) surface (right panels). Black dots indicate experimental estimates for local Fermi momenta [1]. Panels c and d show $q_x$-$q_y$ distributions for both faces taken at $z$=1 a.u. above the topmost atomic layer. [from (2)]

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* lemell@concord.itp.tuwien.ac.at

# Scattering of fast atoms and ions from surfaces under channeling conditions

H. Winter

*Institut für Physik der Humboldt-Universität zu Berlin, D-12489 Berlin, Germany*

Scattering of fast atoms and ions from surfaces under glancing angles of incidence proceeds in the regime of “surface channeling” where projectiles are steered in terms of small angle collisions by atoms of the topmost layer of the surface. Collisions with the solid under these conditions are characterized by defined trajectories for the ensemble of scattered projectiles which is the basis for a detailed model description of the interaction mechanisms [1]. An important issue of channeling is the presence of the two vastly different regimes of motion during the scattering process: (1) a “fast” motion parallel to the surface (“planar channeling”) or strings of surface atoms (“axial channeling”) which proceeds with the kinetic energy of the incident projectiles, whereas (2) the motion normal to the surface plane or atomic strings is several orders of magnitude smaller; i.e. amounts for keV projectiles to about eV-energies.

In the talk an overview will be presented concerning the different aspects and techniques which demonstrate the substantial potential of surface channeling for studies on the ion-surface interactions and on the structure of surfaces [2]. As examples will be discussed the production of polarized beams, studies on resonant charge transfer, energy loss phenomena, resonant coherent excitation, interactions with insulators, scattering of clusters, investigation of growth and structure of ultra-thin films, as well as the recently discovered Fast Atom Diffraction for quantum scattering of fast atoms under axial surface channeling conditions [3].

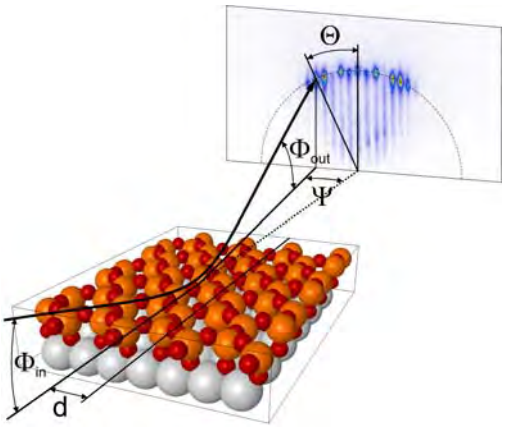


Figure 1. Scattering geometry for (axial) surface channeling experiment.

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# Surface Ion Microscopy (MEIS & SIMS)

# for Nanotechnology & Biotechnology

DaeWon Moon

*Korea Research Institute of Standards and Science*

Energetic ions in the energy range from ~ 100 eV to ~ 100 keV are useful to understand the surface and interface composition and structure of nanothin films and biosystems using scattering and sputtering processes. Two ion beam analysis techniques actively studied for nanotechnology (NT) and biotechnology (BT) applications are medium energy ion scattering (MEIS) and secondary ion mass spectrometry (SIMS)

In this lecture, I'd like to present the basic principles, instruments, and major applications of MEIS and SIMS for semiconductors, displays, nanoparticles, biosurfaces, cells and tissues. Rather than listing up many application results, I will try to discuss the present status and challenges of MEIS and SIMS for practical and useful applications in NT and BT.

MEIS is based on binary scattering and electronic stopping near sub surface region down to ~ 10 nm range. With channeling, MEIS is excellent for compositional and structural depth profiling of nm thin films. However, MEIS has not been widely used due to the lack of imaging or small spot analysis capability, the high cost of analysis, the large instrument size, and etc. Recent progresses to make MEIS more practically useful techniques such as HRRBS and TOF-MEIS will be discussed.

SIMS is still a unique & mandatory technique for dopant depth profiling in semiconductors due to its extremely high sensitivity, nano 3D imaging capability. However, the intrinsic destructive feature and serious matrix effect are the major problems for more reliable and wide applications in NT such as ultrashallow junction profiling and nanostructured materials. For last more than 10 years, the SIMS community has been trying to make SIMS a useful tool for bio-organic analysis with a partial success. Cluster ion beam technology almost solved the ion beam damage issues in polymer and bio specimens. However, the sensitivity of SIMS for high molecular weight molecules in polymer and biospecimen is still poor. Improvement of secondary ionization efficiency of organic and bio-molecues by orders of magnitude will be the major challenge in bio-SIMS in the near future. Recent development of bio-SIMS using cluster ion beam will be discussed.

Finally, visions of various cutting-edge ion beam techniques will be discussed to meet the rapidly increasing demands from NT and BT with other complementary techniques.

# Auger neutralization at metal surfaces

R.C. Monreal
Departamento de Física Teórica de la Materia Condensada, C5, Universidad Autónoma de Madrid, 28049 Madrid (Spain).

In this talk I will review the more recent advances in the theory of Auger neutralization of noble gas ions scattered off metal surfaces. The progress is related to different kinds of crystal effects, such as an influence of the band structure or the surface orientation on the neutralization efficiency, that account for some characteristics of the measured ions yields, either at normal incidence or under grazing scattering conditions. I will also present our latest theoretical and experimental results on the probabilities of $He^+$ ions to survive Auger neutralization when scattered at Cu, Ag and Au surfaces. I emphasize what we understood to be the key quantities to govern Auger neutralization and outline the challenges remaining to be met. This opens new perspectives for the further development of our theoretical and experimental work.

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IC-O-01

# Electron Transfer Processes on Au Nano Clusters and Chains Supported on HOPG and Alumina Films and Size Effects

Jie Shen, Juanjuan Jia, Kirill Bobrov, Laurent Guillemot, and Vladimir Esaulov

Institut des Sciences Moléculaires d'Orsay, UMR 8214, CNRS & Univ. Paris Sud, bat 351, Osay 91405, France

Effects of cluster size in reactivity of supported clusters have been noted in several studies. We investigate here how electron transfer processes are affected as a function of growth of clusters on different surfaces on the example of neutralization of $Li^+$ ions and in particular if the substrate plays some role. This extends an earlier study (1) of Li ion neutralization on semiconducting $TiO_2$ to HOPG and alumina. Previously $Li^+$ neutralization in scattering on Au clusters and thin films supported on $TiO_2$ was found to display significantly more efficient neutralization on small clusters, with a decrease with increasing cluster size. These results closely follow the size-effects observed in the reactivity of these systems. In this study we observed again a much more efficient neutralization on small clusters. In case of HOPG on pristine surface with large planes and few defects, clusters agglomerate along step edges forming nanochains as observed by STM. On the nanochains (or wires) neutralization was found to be even more efficient. In case of Au clusters on alumina films, grown by oxidation of a NiAl crystal, we again observe this effect. We also note differences in the electron transfer probability for small clusters on these different substrates. In case of alumina some differences were noted for differently prepared substrates, presumably due to differences in the nature of the oxide support. These results will be presented and discussed.

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# Size effects on the charge exchange process: Li+ neutralization on Au wires and clusters

J. D. Fuhr and M. L. Martiarena

Consejo Nacional de Investigaciones Cientificas y Técnicas (CONICET) and Instituto Balseiro, Centro Atómico Bariloche, Bariloche, Rio Negro, Argentina

Electron transfer processes on surfaces, that play an important role in surface chemistry and influence charge states of ions or atoms scattered on surfaces, have been quite extensively investigated in the past[1]. Recently a renewed activity has developed in this field involving nano-scaled structures. Recent experimental studies of alkali neutralization on supported Ag and Au clusters, performed in the quest of quantum size effects, show that alkali neutralization occurs much more efficiently on small clusters than on films or bulk metal surfaces[2]. A study of size effects on electron transfer is then interesting and necessary. A prerequisite for this is the knowledge of the characteristic of the neutralization on bulk surfaces.

Recently, to understand the anomalous ion energy dependence of the Li neutral fraction for Au, we presented a theoretical study of such systems using DFT and a linearized rate equation approach [3]. This work allowed us to follow the Li charge state evolution for the (111) surfaces of Ag, Au, and Cu, and for the Ag covered Au(111) surface. Since the understanding of the simpler case of neutralization on bulk surfaces was an essential prerequisite, we have started with investigation into nanostuctured systems.

We present here a theoretical study of the neutralization of the Li charge in front of wires and cluster of Au(111). We have carried out DFT calculations within the supercell approach by using the ab-initio total energy and molecular dynamics program VASP[4]. By considering different Au geometries (perfect surface, wire and cluster), and comparing the Li atom number of valence electrons and of the projected density of state as a function of the Li - Au distances, we analyse the size effects on the charge exchange process.

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# First Principles Theory of Nonadiabatic Forces in Ion-Solid Interactions

Alfredo A. Correa[(1)1], J. Kohanoff[(2)], E. Artacho [(3)], D. Sanchez-Portal[(4)], and A. Caro[(5)]

*[(1)] Lawrence Livermore National Laboratory, USA, [(2)] Queen's University, Belfast, [(3)]Nanogune and DIPC, San Sebastıan, Spain ; Ikerbasque, Bilbao, Spain ; and Cavendish Laboratory, UK [(4)] CCSIC-UPV/EHU and DIPC, San Sebastian, Spain, [(5)] Los Alamos National Laboratory, USA.*

The Born-Oppenheimer (BO) approximation is the keystone for molecular dynamics simulations of radiation damage processes; however, actual materials response involves nonadiabatic energy exchange between nuclei and electrons. Therefore, interatomic forces have components that are history-dependent, and in particular dependent on the velocity of the projectile ion. A first principles atomistic description of the ion-solid interaction that simultaneously goes beyond BO, the dielectric linear response, and homogeneous electron gas approximation was so far an elusive task. In this presentation, we will report recent advances that allows us to have a coherent quantitative description of energy loss mechanism, fully non-adiabatic forces and momentum transfer in a chemically specific way. The approach is based on Time-Dependent Density Functional Theory, ab initio pseudo-potentials and coupled electron-ion dynamics. For example, in the case of a fast proton channeling in aluminum, we find that forces and momentum transfer of host atoms near ion channel deviate significantly from the adiabatic case when velocities approach the maximum of the stopping curve.

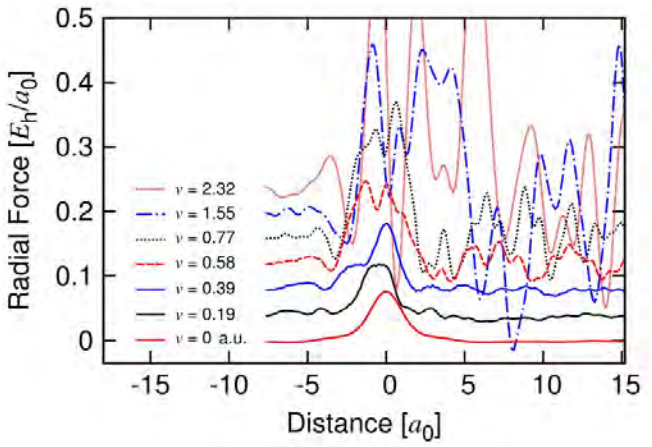


Figure 1. Proton in aluminum: Radial force exerted on host atom (first neighbor to channel trajectory) vs. parallel distance to projectile at different projectile velocities $v$; $x = 0$ is the point of maximum proximity. The nonadiabatic curves have been shifted vertically for visualization purposes.

References

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[1]E-mail: correaa@llnl.gov

# Excitation Thresholds in Electronic Stopping of Low- and Medium-Energy Ions

D. Primetzhofer[(1),(2),*], S. Rund[(2)], D. Roth[(2)], D. Goebl[(2)], and P. Bauer[(2)]

[(1)] *Institutionen för Fysik och Astronomi, Uppsala Universitet, Box 516, S-751 20 Uppsala, Sweden*

[(2)] *Institut für Experimentalphysik, Johannes Kepler Universität Linz, A-4040 Linz, Austria*

The road to success for conventional ion beam analysis is paved by good quantitative predictions and qualitative explanations for the observed electronic stopping cross section $\varepsilon \sim dE/dx$ of the materials for ions at high energies. In contrast, at medium and low ion energies, i.e. at energies below the stopping power maximum, $\varepsilon$ is much less understood, and in many cases even not well known quantitatively, with typically increasing uncertainties towards lower energies. The scatter of data is large, and only model systems have been well characterized. One description commonly employed, and - for metals - expected to be valid to a large degree is a free electron gas (FEG). In a FEG, $\varepsilon$ is expected to be proportional to the ion velocity [1]. Effects like a changing density of the FEG due to a decrease of the maximum possible energy transfer between ion and target electrons lead to a more complicated velocity scaling, which has been observed experimentally [2],[3],[4] and can be understood considering the density of states of the target materials.

Recent experiments, have been devoted to study and compare the velocity scaling of $\varepsilon$ for H and He ions in ultrathin Al, Au and Pt films for low and intermediate ion velocities [5],[6]. For He, a distinct deviation from $\varepsilon \sim v$ is observed for all investigated materials, even if only one of them, Au, features a distinct excitation thresholds for electrons in the conduction band. These findings represent interesting complementary results to earlier studies, and cannot be straightforward explained by considering the unperturbed electronic properties of the investigated target materials. Instead changes induced in the electronic structure of the projectile when passing the material of interest may be used to explain at least qualitatively some of the observed phenomena.

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* daniel.primetzhofer@physics.uu.se

# Wakes, Plasmons and Fermi Gas in the Theory of Stopping of Swift Atomic and Molecular Ions

P. Sigmund[(1)] and A. Schinner[(2)*]

[(1)] *University of Southern Denmark, Odense, Denmark,* [(2)] *Johannes-Kepler University, Linz, Austria*

It is wellknown that the stopping force on a charged particle or aggregate can be expressed by the induced polarization, the wake field. Existing estimates of wake fields most often refer to a Fermi gas, the excitation spectrum of which is known to exhibit a characteristic plasmon peak.

In the stopping literature one may find numerous studies in which wakes, plasmons and Fermi gases play central roles, and others in which those concepts are barely mentioned at all. This does not necessarily imply different predictions concerning measurable quantities.

However, the implied coupling between wake, plasmon and Fermi gas needs caution in the analysis of

1) Stopping of Aggregates and

2) Gas-solid and metal-insulator differences.

We have studied these aspects on the basis of a dielectric function describing a gas of *bound* electrons [1]. Wake effects can be identified by varying the density of the stopping medium. We find that

a) Enhanced stopping of swift molecular compared to atomic ions, found long ago, is predicted to be *increasingly* pronounced as the density *decreases* [2],

b) This implies that the wake field, which has frequently been identified as the *cause* of the enhancement, actually *weakens* the effect [3].

c) Pronounced differences are predicted between conductors and insulators in the charge-state dependence of the wake potential.

d) The common origin of these observations is that over the range of distances relevant to stopping, the primary effect of the wake is to *screen* the ion potential.

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* sigmund@sdu.dk

# Depth profiling of thin films using Coulomb explosion

S.M. Shubeita[(1)], P.L. Grande[(1)*], R.C. Fadanelli [(1)], and J.F. Dias[(1)]

[(1)] *Instituto de Física da Universidade Federal do Rio Grande do Sul, Porto Alegre, RS, Brazil*

Depth profiling measurements of thin films made by backscattering spectrometry expresses depth in units of length assuming the knowledge of the density of the target otherwise "depth" stands as an abbreviation for the number of atoms per unit area (density) over the distance traversed (length) in the target [1]. A method to determine the absolute depth without the knowledge of the density is useful since the density is a physical quantity which can presents different values for the same material.

In this work we explore the Coulomb explosion with occurs when energetic $H_2^+$ ionic clusters interact with thin layers of dielectric materials ($LaScO_3$, $HfO_2$ and $LaAlO_3$, thickness < 100 Å). The Coulomb explosion is connected with the excitation pattern of the target electrons generated by the simultaneous interaction of fragment ions moving in a correlated way within the solid. The molecules dissociate after passing the first monolayer and get stripped of all their electrons. The moving ionic fragments repel each other via mutual quasi-Coulomb forces and excite the electronic medium coherently. The Coulomb explosion leads to a broadening of the energy-loss distribution of the ionic fragments, and can be evaluated through the energy straggling. The information obtained by Coulomb explosion of $H_2^+$ clusters in this approach can provide the *dwell time* of the ionic fragments in the thin layers after the breakup, and thus the *absolute traversed distance.* For this purpose, high-energy resolution backscattering experiments (MEIS: Medium Energy Ion Scattering) were carried out as a function of the incoming projectile energy, covering an energy range between 100 and 200 keV/nucleon. Our results show that the study of the Coulomb explosion and the dwell time of $H_2^+$ molecules traversing ultra-thin films constitute a powerful technique to determine absolute thicknesses and densities of amorphous targets. The experimental values of the Coulomb broadening agree well with calculations considering a screened repulsive potential between the ionic fragments. The thicknesses and densities of different films obtained from the Coulomb broadening are in excellent agreement with those values provided by other techniques like XRR and TEM, thus corroborating the present methodology.

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* grande@if.ufrgs.br

# Surface Structure Determination via Fast Atom Diffraction

J. Seifert* and H. Winter

*Institut für Physik, Humboldt-Universität zu Berlin, Newtonstr. 15, 12489 Berlin, Germany*

We demonstrate the potential of grazing scattering of fast atoms for surface structure determination. Under axial surface channeling conditions, i.e. directions along low indexed strings of surface atoms, projectiles are scattered out of the incident scattering plane normal to the surface. Then, at the maximum of azimuthal deflection an intensity enhancement, the so called "collisional rainbow", is present. For sufficiently small angles of incident ($\Phi_{in} \sim 1°$) and projectile energies ($E \sim 1$ keV), diffraction patterns can be observed in the angular distribution of scattered atoms and molecules [1,2]. In "Fast Atom Diffraction" (FAD), in analogy to established diffraction techniques as LEED, RHEED, HAS, or SXRD, from the splitting of diffraction spots the unit cell size and symmetry and from relative intensities atomic positions in the topmost layer can be deduced [3]. In combination with new triangulation methods based on the transition between axial and planar surface channeling structural models can be tested.

For the system of one monolayer of $SiO_2$/Mo(112) it could be unambiguously distinguished between two competing structural models. The positions of atoms can be determined with an accuracy of a few 0.01 Å [4]. The adsorption of oxygen on a Mo(112) surface was studied in-situ by detection of scattered He-atoms. From the intensity of specularly reflected atoms the formation of well-ordered adsorbate phases is monitored. For scattering along axial channels the diffraction patterns provide information on the evolving surface geometry. For the c(2x4) and pg(2x1) adsorbate structure we demonstrate that not only a periodic lattice but also a separated pair of oxygen strings can be the origin of diffraction. The interference of matter waves from the two equivalent rows within the unit cell can be analyzed in analogy to Young-type interference in a double-slit experiment [5].

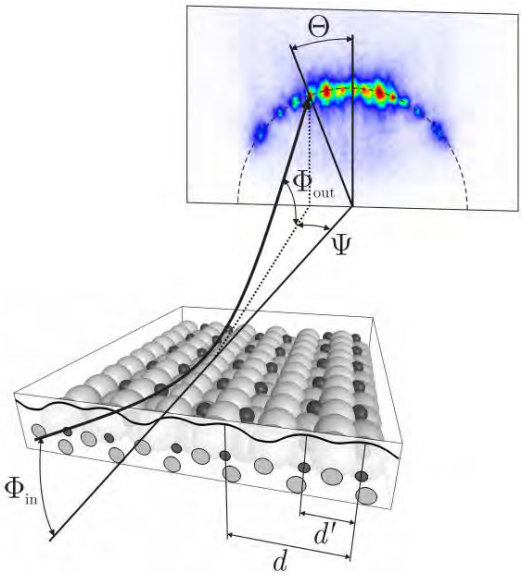


Figure 1. Sketch of scattering geometry for FAD

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* jan.seifert@physik.hu-berlin.de

# Polarized $^4He^+$ ion-surface collisions

T. Suzuki*, Y. Yamauchi, and S. Hishita

*National Institute for Materials Science*

We investigated electron-spin-polarized $^4He^+$ ion scattering on various nonmagnetic surfaces at kinetic energies below 2 keV (spin-polarized ion scattering spectroscopy (SP-ISS) [1]). It was observed that the scattered $He^+$ ion yield depends on the $He^+$ ion spin. We interpret this spin-dependent scattering in terms of the spin-orbit coupling that acts transiently on the $He^+$ 1s electron spin in the $He^+$-target binary collision. This interpretation qualitatively explains the relationship between the spin-dependent scattering and the scattering geometry, incident velocity, and magnetic field arrangement. This is the first study to report spin-orbit coupling caused by projectile electron spin in ion scattering. [2]

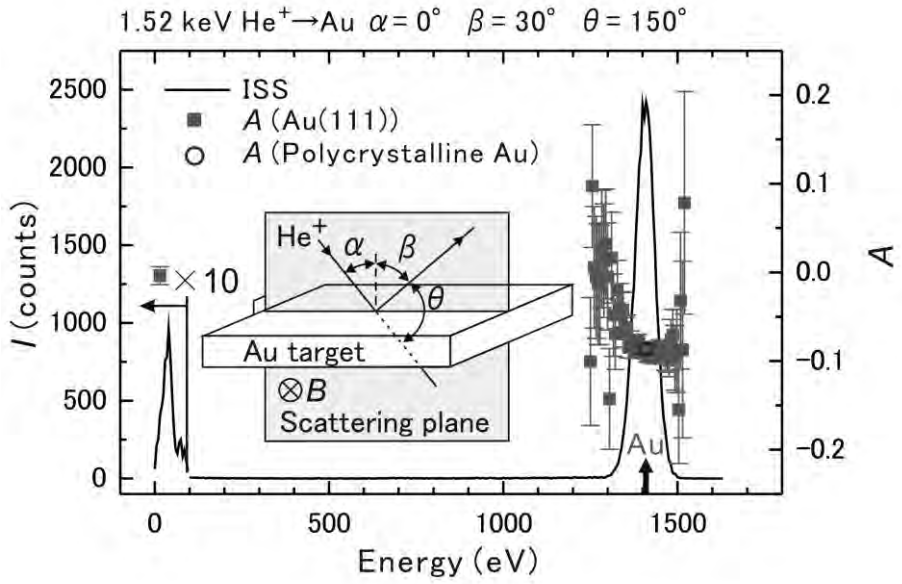


Figure 1. ISS spectrum of the Au(111) surface with the calculated $He^+$-Au binary collision energy (arrow). Filled squares represent the spin asymmetries (*A*) of Au(111); an open circle indicates that of polycrystalline Au. The error of the spin asymmetry corresponds to statistics. The inset shows the scattering geometry. The scattering plane is perpendicular to both the Au surface and the magnetic field ***B***.

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* suzuki.taku@nims.go.jp

# Collisions of 1-10 eV ions at ice surfaces

T. Pradeep

*DST Unit of Nanoscience (DST UNS), Department of Chemistry, Indian Institute of Technology Madras, Chennai - 600036, India*
*Email: pradeep@iitm.ac.in*

Collision of ultra low energy mass selected ions in the range of 1-10 eV on molecular surfaces can lead to a number of interesting phenomena[1]. On water ice, several processes phenomena such as diffusion, phase transition and reaction have been probed.[2-5] The most recent in the category of reactions is the formation of molecular ion of dihydrogen ($H_2^+$) by 1 eV collisions of protons ($H^+$) on amorphous water ice surfaces.[6] The reaction is also observed on crystalline ice surfaces, but with lower efficiency. Collisions of $D^+$ on amorphous $H_2O$ and $D_2O$ ices yield $D_2^+$ on the former, subsequent to isotope exchange on the $H_2O$ surface. Ultra-low-energy collision-induced dihydrogen ion production is also observed from alkanol surfaces, with decreasing efficiency as the alkyl chain length increases. There is no corresponding reaction on solid hexane. This endothermic reaction, with implications for interstellar chemistry and plasma etching processes, is proposed to occur as a result of stabilization of the other reaction product, a hydroxyl radical ($OH^{\bullet}$), on water surfaces through hydrogen-bonding interactions with the surface. These results point to an interesting chemistry involving ultra-low-energy ions on molecular solids. A brief review of our previous work in the area will also be presented.

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IC-O-05

# Thiol Terminated 1,4-Benzenedimethanethiol Films Grown on Au, Ag and Cu Surfaces studied by Direct Recoiling Spectroscopy

L. Salazar Alarcón[1], L. Chen[2], Jie Shen[2], JuanJuan Jia [2], V.A. Esaulov[2], E. A. Sánchez[1], M.L. Martiarena[1],and O. Grizzi[1,*]

*(1) Centro Atómico Bariloche, Instituto Balseiro, Comisión Nacional de Energía Atómica, CONICET, Bariloche, Argentina,*

*(2) Institut des Sciences Moléculaires d'Orsay, Université-Paris Sud and CNRS, Bâtiment 351, 91405 Orsay, France*

Dithiol films deposited on metallic surfaces are promising systems for developing sensors. In most of the cases studied up to now, the dithiol films are grown by dipping an already prepared surface in a solution containing the appropriate dithiol [1]. This has been shown to work well for Au surfaces, however in more reactive surfaces a vacuum approach is desirable to obtain cleaner film-substrate interfaces. In the vacuum approach [2-5], at low exposures the dithiol molecule tends to adsorb with both S atoms bonded to the substrate. The question whether the molecule will stand up or not at higher exposures is open and the adsorption conditions to obtain this Self Assembled Monolayer (thiol exposed film) depend on the substrate-molecule interaction. Direct Recoil Spectroscopy (DRS) [5] has the appropriate surface and element sensitivity to test the thiol termination of the films, and the low damage imparted to the organic film allows carrying on detailed studies of the film stability with surface temperature. In this work we present a TOF-DRS study of 1,4-benzenedimethanethiol films grown in vacuum on Au, Ag and Cu surfaces. We discuss the thiol termination and compare the corresponding spectra to those for the same substrates under pure S exposures. DFT calculations for these systems are used to obtain the shape of the molecule in front of the surface, to study the energetics of its adsorption and to understand the main features of the TOF-DRS spectra. Different implementations of the Van der Waals correction within the VASP[6] code are tested to study how this term affect the system description. Studies performed at temperatures below room temperature suggest formation of a multilayer. This thicker film is identified from the multiple scattering of Ar or Kr projectiles at high incidence angles, i.e., involving collisions with both molecule and substrate. Thermal desorption by TOF-DRS allows the study of the stability of the thiol termination with temperature, determination of the temperature at which the multilayer desorbs, and identification of the final molecule products remaining after annealing.

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Corresponding author: grizzi@cab.cnea.gov.ar

# Okorokov Effect in a Magnetic Lattice for a Slow Atomic Beam

A. Hatakeyama[(1)*]

[(1)] *Tokyo University of Agriculture and Technology*

We study resonance transitions in the internal states of atoms passing through a static periodic field. This motion-induced resonance occurs when the frequency of the field oscillation that the atoms experience equals the transition frequency. The principle of the resonance is the same as that of the "Okorokov effect" [1] or "resonant coherent excitation" (RCE) [2], which has been studied extensively using fast ion beams passing through crystals for resonances at high frequencies up to $10^{18}$ Hz or keV. We are interested in its extension to very low energy experiments, such as resonances in the rf or microwave region. The motion-induced resonance is strongly velocity-dependent in nature, and it is particularly worth noting that the internal excitation is supposed to occur at the expense of the atomic kinetic energy [3]. These features are not attainable with the standard rf or microwave resonance technique, and we expect that the motion-induced resonance will find useful applications as a new type of control method for slow atoms.

Our study started with a proof-of-principle experiment in an energy range of neV or $10^5$ Hz [4], quite different from traditional RCE experiments. Magnetic resonance was induced between the Zeeman sublevels of Rb atoms with a velocity of $v \sim 10^2$ m/s in a periodic magnetic field ("magnetic lattice") with a period of $a \sim 1$ mm. We used a thin cell containing a Rb vapor, to which a periodic field was applied with a pair of arrays of parallel current-carrying wires. In the next experiment, an effusive atomic beam of Rb ($v \sim 10^2$ m/s) passed through a magnetic lattice produced by a stack of planar arrays of parallel current-carrying wires ($a \sim 1$ mm) [5]. This experimental configuration realized a longer coherent interaction of atoms with the periodic field, resulting in sharp resonance lines, whose widths were primarily determined by the transit time through the lattice. We also observed Rb spin nutation, which was a first direct demonstration of coherent population transfer induced by atomic motion through a periodic field [6].

Experiments using magnetic lattices with smaller periods (1 μm ~ $a$ < 1 mm) are currently underway.

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* hatakeya@cc.tuat.ac.jp

# (Heavy) Ion Acceleration by Laser Pulses

J. Schreiber[(1,2)*]

*[(1)] Ludwig-Maximilians-Univsersity Munich, [(2)] Max-Planck-Institute for Quantum Optics Garching*

After more than one decade of successful operation at the Max Planck Institute for Quantum Optics, the Advanced Titanium-Sapphire Laser (ATLAS) system has been dismantled and is currently transferred to its new, temporary home, the Laboratory for Extreme Photonics (LEX). This move will allow us to upgrade the peak power from currently 60 TW to 300 TW before it can take on its final upgrade to 3 PW at the Centre for Advanced Laser Applications (CALA), possibly constituting one of the world's most powerful laser systems. The opportunities are manifold. GeV-electron bunches with a few femtosecond pulse duration will be available routinely and in turn enable for the generation of even shorter light, UV, X- and Gamma-ray pulses. The high laser pulse energy (60J) paired with the short duration (20fs) will allow to access light intensities of up to 1023W/cm2 and address fundamental questions of modern physics.

One major prospect is the generation of ion bunches with energies beyond 100 MeV/u, sufficiently high to approach and investigate their applicability in tumour therapy constituting one of the grand goals of our research. One of the most exciting aspects of laser acceleration is the high bunch density not far from a solid even for heavy ions with energies of a few MeV/u. Such bunches are discussed to serve in nonlinear nuclear effects such as coherent energy loss or even fission-fusion reactions for the production of neutron rich nuclei.

In this talk, I will explain various concepts of laser-driven ion acceleration that have been employed and studied over the past years. At present, the application of nanometer thin foils seems to be most promising both in terms of achieving highest energy and conversion efficiency. The demands on the quality and control of the laser pulses, mainly in terms of the suppression of prepulses, are enormous. Despite of these difficulties, we could demonstrate first biological studies with tumour cells irradiated by laser accelerated proton bunches with single shot doses of several Gray delivered within 1 nanosecond. This demonstration has been a major mile stone of our research. Moreover, the combined efforts in laser, target and detector development disclosed a number of new and partially surprising insights that constitute my excitement for this field of physics and motivate for the future challenges and possibilities that await us.

* Joerg.schreiber@mpq.mpg.de

# Intense, short pulse ion beams for studies of warm dense matter and point defect dynamics in solids

Thomas Schenkel*, Pavel Ni and J. W. Kwan

*Lawrence Berkeley National Laboratory, 1 Cyclotron Rd., 5R121, Berkeley CA 94720, USA*

A novel, induction type, heavy ion linear accelerator, the Neutralized Drift Compression eXperiment (NDCX-II) is scheduled to come online at Berkeley Lab in the Fall of 2012. This new user facility is designed to deliver intense (~1 to 50 nC/pulse), sub-ns pulses of 1.2-3.5 MeV lithium ions at a rate of about 2 pulses per minute [1]. When focused to a mm-diameter spot, the beam can volumetrically heat a few micrometer thick metal foil targets up to ~30,000 K (~2-3 eV), generating hundreds of Mbar of pressure. The duration of the ion beam pulse is shorter then the hydro response time of a typical sample, making NDCX-II an ideal driver for warm-dense-matter studies. For studies of radiation effects in solids, the beam intensity can be controlled by defocussing while the short pulse duration is maintained. Incident lithium ions generate collision cascades when they impinge on solid targets. While collision cascades ensue on a ps time scale, diffusion and recombination of vacancies and interstitials can stretch over much longer time scales. The sub-ns excitation pulse with tunable intensity enables "pump-probe" type experiments for *in situ* studies of point defect dynamics and time resolved studies on the formation dynamics of specific defects such as color centers. Diagnostics capabilities include x-ray diffraction and time resolved detection of transmitted ions and emitted light (e. g. from color centers in diamond). In our presentation we will describe the ion beam user facility concept and capabilities and discuss experimental research opportunities in the fields of warm dense matter and radiation effects in solids.

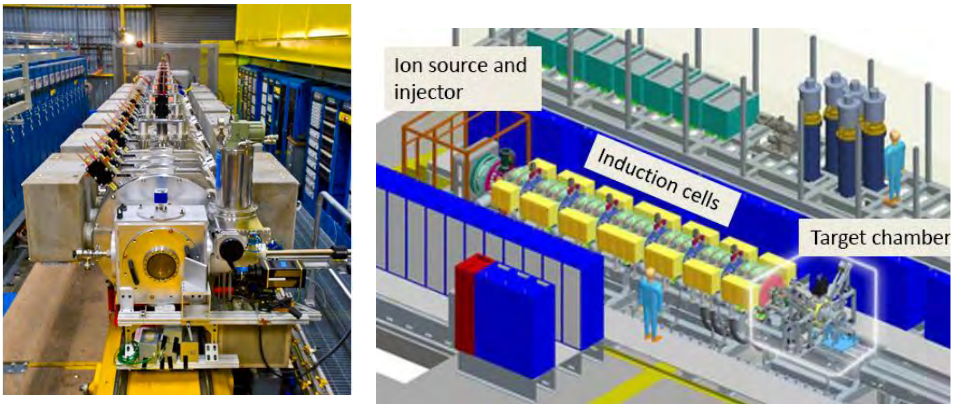


Figure 1 : Photo (left) and schematic (right) of the NDCX-II induction linac at Berkeley Lab.

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Contact email : T_Schenkel@LBL.gov

# Radiobiology with Laser-accelerated Proton Beams

A. Yogo[(1) *], T. Maeda [(2)], H. Sakaki[(1)], Y. Fukuda[(1)], and K. Kondo[(1)]

*[(1)] Japan Atomic Energy Agency, [(2)] Hyogo Ion Beam Medical Center*

Recently, high-intensity laser ion acceleration has been suggested as a potential, cost-saving alternative technology to conventional accelerators for radiotherapy. When a laser pulse of intensity well above $10^{18}$ W/cm$^2$ interacts with a thin foil target, a laser-field-driven force generates a strong accelerating field exceeding 1MV/μm, which exceeds the field in conventional ion accelerators by as much as six orders of magnitude. In this work, investigations of the biological effects of the high bunch current and short bunch duration, that are typical of laser-acceleration, are reported [1,2,3].

The experiment was performed using the J-KAREN laser system at JAEA. Laser pulses of 1 J energy and 45 fs duration are focused to an intensity of $5\times10^{19}$ W/cm$^2$ onto a polyimide foil target of 7.5-μm-thickness which is continuously fed by a servomotor. Laser pulses are delivered at a repetition rate of 1 Hz. The emitted proton spectrum is continuous up to a maximum energy of 4 MeV. Energy selection for transport to cancer cells is determined by four pairs of dipole magnets (DMs), which is about 40 cm in length. Each DM consists of a pair of permanent magnets generating a central magnetic field of 0.78 T. The second and third magnetic fields are oriented antiparallel to the first and fourth ones. Proton energy is selected by a pinhole in the middle plane located between the second and third DMs.

We have successfully obtained 2.25 MeV proton beams with an energy spread of 0.66 MeV (FWHM) and single bunch duration of 20 ns. The beams were extracted from vacuum into air through a thin-foil window and used to irradiate in-vitro cell samples from a human salivary gland tumor (HSG cells). The dose given in a single proton bunch was 0.2 Gy, hence, the single bunch dose rate is estimated to be $10^7$ Gy/s. At the 1 Hz repetition-rate cell samples were irradiated with successive proton bunches with integrated dose levels up to 8 Gy (40 shots).

Using a colony formation assay, we determined the value of relative biological effectiveness (RBE) of this proton irradiation for cell inactivation. X-rays delivered from a 4-MV clinical linac were used as reference radiation. RBE at the 10% surviving rate was evaluated to be 1.20 ± 0.11 for the laser-accelerated protons having a volume-averaged LET of 17.1 ± 2.8 keV/μm. This is the first RBE estimation for laser-accelerated ions. We compare our results with published ion beam RBE using conventional accelerators.

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* yogo.akifumi@jaea.go.jp

# Science of asteroid sample return mission "HAYABUSA"

H. Yurimoto[(1)*]

[(1)] *Hokkaido University*

Of the ~40,000 meteorites we know of, only 14 have had their pre-impact orbits ascertained. The aphelia of these 14 orbits are located within the Main Asteroid Belt between Martian and Jovian orbits, which suggests that meteorites are an asteroidal origin. However, telescopic spectroscopy of asteroidal surfaces had raised serious doubts on the hypothesis. One of the primary scientific purposes of Hayabusa mission was demonstration of this hypothesis. The Hayabusa spacecraft made two touchdowns on the surface of asteroid 25143 Itokawa on November 20th and 26th, 2005. After recoveries from serious accidents on the way home from Itokawa, the spacecraft made the reentry into the terrestrial atmosphere on June 12th, 2010 and the sample capsule was successfully recovered in Australia on June 13th, 2010.

More than 1,500 grains were found in the sample capsule and identified as rocky particles. Although their sizes are mostly less than 10 micrometers, some larger grains of about 100 micrometers or larger were also included. About 50 particles among the larger grains were subjected to one set of preliminary examinations. The preliminary examinations started from January 21st, 2011. The examinations include X-ray CT analysis, X-ray diffraction analysis, petrology, mineral chemistry, oxygen isotope analysis, trace element analysis and noble gas analysis.

In order to determine extraterrestrial origin of the particles in the capsule, isotope analysis of oxygen for individual particles is essential because extraterrestrial materials usually show different isotope ratios from terrestrial materials. However, the degree of difference is less than 1%. A precise analysis technique by SIMS was newly developed and applied.

The SIMS characterization of Itokawa particles certified that Itokawa was composed of similar materials of ordinary chondrites [1]. This is the first direct link showing that S-type asteroids is one of the sources of the ordinary chondrites, which are the most abundant meteorite species fallen into the Earth. From this citification, we confirm that insights based on meteorite research are the right way to study origin of our solar system. New insights for the early solar system are expected by further detail measurements of Itokawa particles.

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* yuri@ep.sci.hokudai.ac.jp

# Ionization at Insulating Surfaces – A Review about a successful Technique to measure Energetic Neutral Atoms (ENA) in Space

P. Wurz[(1)], and J.A. Scheer[(1)*]

[(1)] *University of Bern, Physikalisches Institut, Sidlerstrasse 5, 3012 Bern, Switzerland*

In space science many applications of remote sensing of plasma populations require a high detection efficiency of Energetic Neutral Atoms (ENA) because the sources of these ENAs are very faint [1]. This requires sophisticated detection techniques and not before long measurements of ENAs were quite impossible due to the technical limitations onboard of a spacecraft. The reason is neutral particles have to be ionized before they can be analyzed and ionizing techniques, which work fine in a laboratory on earth, can be very difficult, if not impossible, to implement in an instrument, which shall work on a satellite in space.

In the mid 1990ies reports about unexpected high yields of negative ions upon scattering of positive and neutral particles from various surfaces led to a new type of neutral particle sensing mass spectrometers, which use the effect of surface ionization and work now successfully on several space missions, which are IMAGE [2] and IBEX [3] (NASA) and the CENA instrument (ESA & ISRO) from the Indian mission to moon, i.e. Chandrayaan-1 [4]. Furthermore, another instrument of such kind, called ENA, is part of the scientific payload of the Mercury Magnetospheric Orbiter (MMO) satellite of the BepiColombo mission to Mercury. BepiColombo is a joint mission of ESA and JAXA and the MMO module is lead by JAXA. Launch is scheduled for August 2015.

However, it took many years to find good working so-called Conversion Surfaces (CS) [5] and this report will summarize our tests with possible candidates for conversion surfaces, with special emphasis on ionization efficiency and scattering properties. Furthermore, we will report about the current status and finally explain why the quest for the perfect conversion surface is still going on.

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* jscheer@space.unibe.ch

# Why Is Low Energy $Cs^+$ Efficient For Depth-Profiling Both Organics And Inorganics?

L. Houssiau[(1)*], J. Brison[(1)], N. Mine[(1)], N. Wehbe[(1)]

[(1)] *University of Namur (FUNDP), Research Centre in Physics of Matter and Radiation (PMR), 61 rue de Bruxelles, B-5000 Namur, Belgium*

Depth-profiling a material without modifying its chemistry has always been a challenge. This holds especially true for organic materials, which are very sensitive to etching beams and tend to degrade rapidly. However, new polyatomic ion sources ($C_{60}^+$, $Ar_n^+$) have been developed over the last decade, allowing real molecular depth profiling thanks to their huge sputtering yield. In 2007, our group has also shown that, surprisingly, low energy (~250 eV) $Cs^+$ ions could also be used to depth profile polymers[1]. We have extended the study to many different organic materials, including amino acid thin films, analyzed in the energy range 150-1000 eV. This paper aims at reviewing recent advances in understanding how low energy $Cs^+$ ions interact with organics in such a way that the chemical structure is preserved, contrary to all other monoatomic ions. It is indeed a special case of atomic collisions in solids where atoms are extremely reactive, inducing major chemical effects.

$Cs^+$ beams have been extensively used in SIMS on inorganic materials, thanks to the strong negative ionization enhancement they bring about, but also for the creation of $MCs_n^+$ clusters, where M is a matrix element. We will show that a similar negative ionization enhancement, along with $MCs_n^+$ cluster formation, also occurs on polymers and small organic molecules, making low energy $Cs^+$ a versatile ion source. In both organics and inorganics, the interest of Cs lies in its strong reactivity, being the most electropositive element. Implanted Cs atoms reduce the surrounding molecules, creating anionic sites, therefore enhancing the negative ionization. Moreover, Cs reacts preferentially with free radicals, preventing cross-linking reactions, thus allowing molecular depth-profiling. Our model is supported by recent XPS data, showing changes in the charge state of phenylalanine molecules irradiated by $Cs^+$ (a decrease of the N 1s $NH_3^+$ contribution along with an increase of the O 1s $COO^-$ contribution). XPS also allows measuring the surface Cs concentration (up to 8%). Optical Emission Spectroscopy (OES) data from the surface were also acquired, proving the existence of neutral Cs species at the surface.

Every ion source has its pros and cons. $Ar_n^+$ clusters sources are extremely efficient on organic materials but are of limited use on inorganics. On the other hand, $Cs^+$ ions allow depth profiling on all types of materials, although with a much lower sputtering yield.

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* laurent.houssiau@fundp.ac.be

# Hydrogen and Oxygen Trapping and Retention in Metals and Graphite Materials Irradiated in Plasma

L. Begrambekov

*National Research Nuclear University (MEPhI), Moscow, Russia*

The plasma facing materials are subjected to irradiation with ions and neutrals in a wide energy range including practically zeroth energies. Last years the features of trapping of low energy hydrogen and oxygen in plasma irradiated metals (NI, W, stainless steel, etc.) and carbon materials (pirolytic graphite, fine grain graphite, carbon fiber composite (CFC)) have been studded in details. Special attention was paid to hydrogen and oxygen interaction with stainless steel (SS) representing the metals (alloys) with oxide layer on the surface.

The report presents the newly discovered features and mechanisms of hydrogen and oxygen trapping and desorption from carbon materials and metals under plasma irradiation.

Carbon materials. Dependence of hydrogen trapping on material structure, charge state of impinging particles, plasma ion flux and energy, irradiation dose, presence of oxygen impurity in plasma, specimen temperature are discussed. Experimental results allowed elaboration of "potential" mechanism of hydrogen trapping. This mechanism considers trapping as the process proceeded at the expense of energy of inelastic interaction of particles with the surface. It plays the main role in trapping of low energy hydrogen particles and explains features of hydrogen trapping in carbon materials irradiated in plasma.

Conditions of methane formation in carbon materials irradiated in the hydrogen plasma, its thermal desorption and interrelations between hydrogen trapping and "chemical" sputtering are described. Oxygen trapping in carbon materials irradiated in $H_2$+$O_2$ plasma and its removal due to hydrogen plasma irradiation are the subject of special consideration.

Metals. The processes on oxidized surface of SS are described. It is shown; in particular, that irradiation with hydrogen atoms of not annealed SS submersed in $H_2$+$O_2$ gas mixture initiates formation of $H_2O$ molecules on SS surface. Hydrogen released from SS as well as hydrogen of surrounding atmosphere take part in $H_2O$ formation. The similar result was observed when SS was irradiated with ions of ($H_2$+$O_2$) plasma.

The parallel measurements of hydrogen and oxygen trapping in SS and Ni exposed in oxygen contaminated hydrogen plasma were performed to clarify the role of oxide surface layer in this process. Among them there were measurements of dependence of trapping on ion energy, irradiation dose, oxygen concentration in plasma. The results demonstrate that oxide layer appears to be the reason of significant increase of the rate of hydrogen trapping and of amount of retained hydrogen in SS. The main fraction of hydrogen trapping in SS is provided by the "water" molecules sorbed on irradiated surface through the mechanism of "potential" trapping. Oxygen atoms trapped in SS are practically fully concentrated within oxide surface layer and bond with chromium atoms.

Oxygen concentration in plasma influenced on hydrogen trapping in SS. Hydrogen trapping in Ni proceeded by the kinetic energy of implanting particles ("kinetic" mechanism.) and did not depend on oxygen concentration in plasma.

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lbb@plasma.mephi.ru

# $H^-$ production by surface ionization on carbon materials in $H_2$ plasma

J.M. Layet[(1)*], A. Ahmad[(1)], C. Pardanaud[(1)], A. Gicquel[(2)], and G. Cartry[(1)]

*[(1)] PIIM, Aix-Marseille Univ, CNRS, France, [(2)] LSPM, CNRS, Paris 13 Univ, France*

The ITER (International Thermonuclear Experimental Reactor) project aims at demonstrating power production by magnetic confinement nuclear fusion. In such a device, neutral beam injection (NBI), which uses high power beams of fast neutral atoms of hydrogen isotopes to heat the plasma, is necessary to reach high performances. The ITER NBI system requires beam energy at 1 MeV where the neutralization of positive ions becomes very inefficient and only the negative ions can be neutralized with sufficient yield. Hence the development of high current negative ion sources (50A D- beam for ITER) is crucial. The use of cesium (Cs) injection to enhance surface ionization in the negative-ion source is nowadays the only way to meet ITER requirements in terms of D- current. However, cesium injection has severe drawbacks, and the development of Cs-free negative-ion sources would be highly valuable. The present work deals with negative-ion production by surface ionization on carbon materials in Cs-free hydrogen plasma.

In beam experiments, surface ionization mechanisms are most often studied by measuring angle and energy distribution functions of all particles (positive and negative ions, electrons, neutrals) emitted or scattered by a well characterized surface. In plasma experiments, such analyses cannot be performed easily due to the presence of multiple charged and neutral species, to the presence of electric fields modifying angle and energy distributions and to strong surface modifications induced by particle bombardment. In the present works, energy distributions of $H^-$ negative ions emitted by a carbon sample in hydrogen plasma are measured by an energy-resolved mass spectrometer. In order to get information on the energy distributions on the surface, the trajectories of the negative ions between the sample and the spectrometer, as well as the negative-ion transmission inside the spectrometer are computed. The measured distribution functions are then modeled using a test surface-distribution-function. It appears that the distribution functions given by SRIM and arising from the impact of $H^+$ onto a-CH material are appropriate to reproduce measured negative-ion distribution functions, suggesting few dependence of the electron capture probability with angle and energy of the outgoing particle.

A comparative study between HOPG (Highly Oriented Pyrolitic Graphite) and BDD (Boron Doped Diamond) has revealed the possible influence of the electro-affinity of diamond on the electron capture. Although HOPG and BDD behaves similarly at low surface temperature, the negative-ion yield is completely different at high temperature (400°C) where it increases by a factor 5 on diamond and decreases by a factor 3 on graphite. Surface modifications after plasma treatment at low and high temperature have been studied by Raman spectroscopy, allowing giving a possible interpretation of this result.

* jean-marc.layet@univ-amu.fr

# Atomic relocation in ion-irradiated ultra-thin magnetic films visualized with sub-nm spatial resolution

H. Gnaser[(1),(2),*], R. Schiller[(2)], M. Wahl[(2)], B. Reuscher[(2)], A. Zeuner[(2)], M. Kopnarski[(2)], R. Neb[(1)], B. Hillebrands[(1)]

*[(1)] Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, D-67663 Kaiserslautern, Germany*
*[(2)] Institute for Surface and Thin-Film Analysis (IFOS), Trippstadter Str. 120, D-67663 Kaiserslautern, Germany*

The relocation of atoms in solids caused by ion irradiation has been studied by 3D Atom Probe Tomography (APT) [1]. This technique enables the determination of atomic positions with sub-nm spatial resolution. This is illustrated using a MBE-grown trilayer system of Fe(10 nm)/Cr(0.7 nm)/Fe(10 nm) [2]. It is well documented [3] that the magnetic properties of such films can be modified by ion-beam exposure. The specimen was bombarded by 30 keV $Ga^+$ ions at low fluences ($\leq 3\times10^{15}$ $Ga^+/cm^2$), and the compositional changes induced thereby were monitored by APT. In the pristine specimen the presence of the 0.7-nm Cr layer could be verified. Upon irradiation, this layer is broadened to ~ 1.7 nm at a fluence of $3\times10^{14}$ $Ga^+/cm^2$. Fig. 1 illustrates these results by means of a representation of the positions of the Cr and Fe atoms before and after ion bombardment.

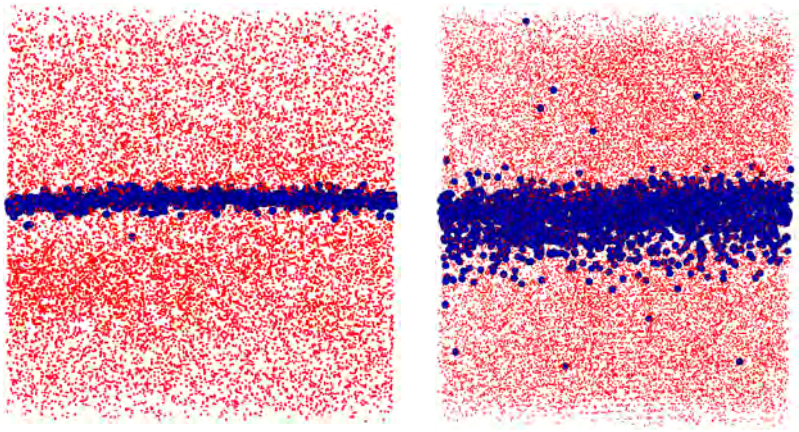

Fig. 1. The distribution of Cr (blue spheres) and Fe (red dots) atoms in a Fe/Cr/Fe layer system in the pristine state (left panel) and upon irradiation with 30 keV $Ga^+$ ions at a fluence of $3\times10^{14}$ $Ga^+/cm^2$ (right panel), obtained by 3D atom probe tomography. The width of the displayed region is 10 nm.

These data indicate that 3D APT is a promising tool for the examination, with sub-nm resolution, of atomic relocation processes in solids induced by ion bombardment. Future experiments and possible limitations are discussed.

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*gnaser@rhrk.uni-kl.de

# Anisotropic Mass Transport on Ion-Bombarded Titanium Dioxide Surfaces

M. Kolmer [(1)], A. A. Ahmad Zebari [(1)], D. Suszalski [(1)], A. Polit [(1)], F. Krok [(1)], F. Buatier de Mongeot [(2)], F. Zasada [(3)], W. Piskorz [(3)], Z. Sojka [(3)], and M. Szymonski [(1)]

[(1)] Research Center for Nanometer-Scale Science and Advanced Materials (NANOSAM), Faculty of Physics, Astronomy and Applied Computer Science, Jagiellonian University, Reymonta 4, 30-059 Krakow, Poland, [(2)] Dipartimento di Fisica, Universita di Genova, Via Dodecaneso 33, 16146 Genova, Italy, [(3)] Faculty of Chemistry, Jagiellonian University, Ingardena 3, 30-060 Krakow, Poland

Basic processes responsible for nanoscale morphology changes of single crystal anisotropic $TiO_2$ (110) surfaces irradiated by keV energy ion beams ($Ar^+$, $Xe^+$) have been investigated by means of the scanning tunnelling microscopy. We have found that the ripple morphology pattern developed on $TiO_2$ (110) surface under ion irradiation shows a remarkable temperature-dependent rotation. At 300 K the ripples are oriented perpendicular to the ion beam direction whereas at the other studied temperatures (150 K, 620 K and 720 K) they are aligned along the beam direction. We have demonstrated that the formation of the nanostructures is determined by the interplay between the effects of preferential erosion of the monatomic step edges at grazing incidence and directional surface diffusion along the favoured crystallographic orientation. Density-functional theory (DFT) calculations reveal that the highly mobile surface species responsible for the development of the nanoripples are TiO dimers. The observations highlight a unique character of the anisotropic mass transport on irradiated oxide surfaces invoking dimer (metal oxide) mediated diffusion.

# Anisotropic dewetting of ion irradiated solid films

L. Repetto[(1) *], B. Šetina Batič [(1), (2)], G. Firpo[(1)], E. Piano[(1)], and U. Valbusa[(1)]

[(1)] *Dipartimento di fisica, Università di Genova, Via Dodecaneso 33, 16146 Genova, Italy ,*
[(2)] *Inštitut Za Kovinske Materiale in Tehnologije, Lepi pot 11,1000 Ljublijana, Slovenia*

When solid matter is brought to a molten state, forces acting on a molecular scale can induce self-organization processes of scientific and technological relevance. In the case of thin films, the concurring action of surface tension and Van der Waals forces can be responsible for the amplification of surface fluctuations and produce the mechanism of spinodal dewetting. We have recently demonstrated that a spinodal like phenomenon, modified by the presence of sputtering, can be induced also when the melting agent is ion bombardment [1]. Here we show that by recognizing the real forces acting on the system we are able to perform a modified experiment were the symmetry is broken and deterministic structures are created thus showing that the process can be exploited for nanofabrication purposes.

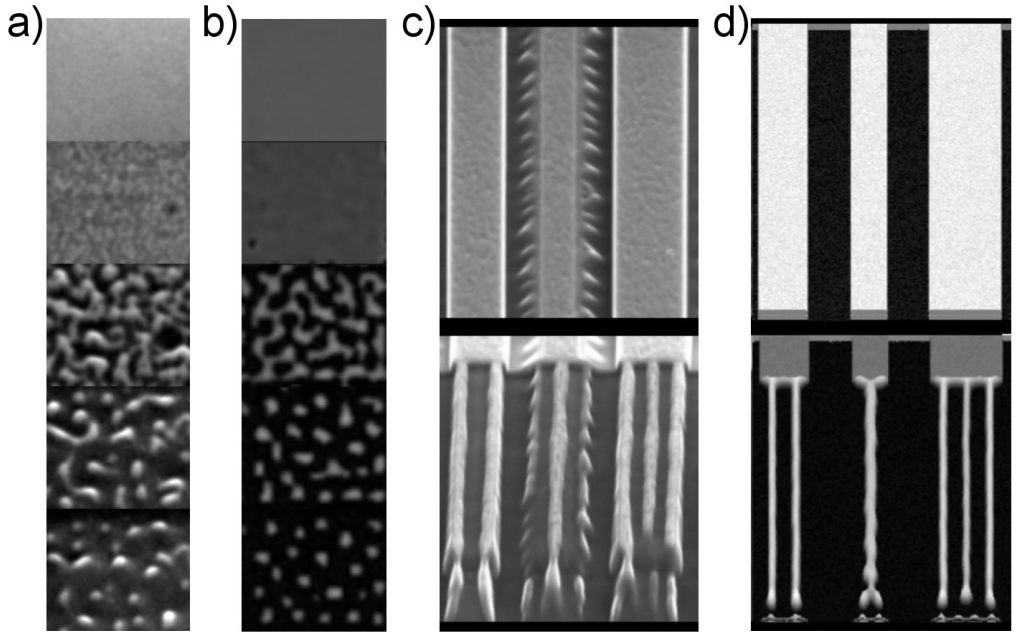


Figure 1. 30 keV Ga ions irradiation of Cr films. a) Experimental sequence of isotropic dewetting. b) Simulation sequence of isotropic dewetting. c) Experimental sequence of anisotropic dewetting. d) Simulation sequence of anisotrpic dewetting.

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* Luca.Repetto@unige.it

# Tailoring of keV-HCI Beams by Transmission through Insulating Nanocapillaries

N. Akram[1], I. L. Soroka[1†], C. Trautmann[2], HQ. Zhang[1]*, and R. Schuch[1]

[1]*Physics Department, Stockholm University, Stockholm, Sweden*

[2]*GSI Helmholtzzentrum für Schwerionenforschung, D-64291 Darmstadt, Germany*

Charged particles, in particular slow highly charged ions, have shown to be guided through nanocapillaries in various insulating materials by self-organized charge patches on the inner walls of the channels due to initial ion impact [1-3]. The time dependent formation of the guiding potential has been demonstrated by measurements[2,3]. In contrast to previous guiding experiments, we are utilizing capillaries of rhombic and rectangular cross-sections (Fig.1). The capillaries were fabricated by chemical track etching of the irradiated mica sheets. The irradiation was performed at the GSI UNILAC with 11.1 MeV/u Pb- and Xe-ions for muscovite and phlogopite mica, respectively. Ion-beam transmission measurements were performed at the ECR and S-EBIT sources of Stockholm University using $Ne^{7+}$-ions of kinetic energy between 1-10 keV/q.

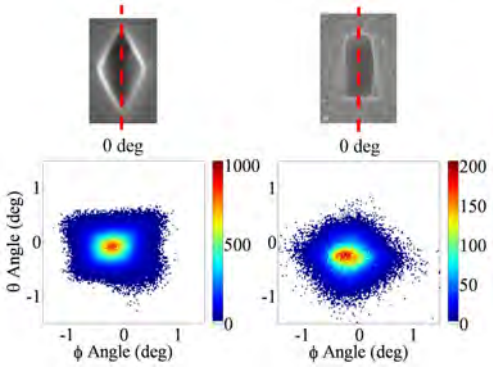


Figure 1: Cross sections of rhombic (muscovite) and rectangular (phlogopite) nanocapillaries and the corresponding angular distributions of 7-keV-$Ne^{7+}$ ions.

Surprisingly, we found that the ion beam profiles are tailored by the geometrical shape of the nanocapillaries: rectangular cross sections produce rhombic shaped ion transmission profiles with the long axis aligned along the short sides of the rectangles (Fig. 1) and, vice versa, rhombic cross sections produce rectangular shaped ion transmission profiles with the long side aligned along the short axes of the rhombi (Fig. 1). More details on this novel beam shaping mechanism are described in this contribution and in publications [4-6].

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*Present address: School of Nuclear Science and Technology, Lanzhou University, Lanzhou 730000, China
†Present address: Nuclear Chemistry, Royal Institute of Technology, SE-100 44 Stockholm, Sweden

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schuch@fysik.su.se

# MeV ion micro-beams shaped by glass capillaries

A. Cassimi[a], C. L. Zhou[a], T. Ikeda[b], M. Simon[c], I. Monnet[a], C. Grygiel[a], A. Mueller[c], A. Méry[a], S. Guillous[a], H. Lebius[a], A. Benyagoub[a], H. Shiromaru[d], M. Doebeli[c]

[a]CIMAP CEA/CNRS/ENSICAEN/UCBN, BP5133, F-14070 Caen cedex 5, France
[b]Atomic Physics Laboratory, Riken, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan
[c]ETH Swiss Federal Institute of Technology, Rämistrasse 101, 8092 Zurich, Switzerland
[d]Tokyo Metropolitan University, Hachioji, Tokyo 192-0397, Japan

The application of microscopic ion beams focused by tapered glass capillary received extensive attention in recent years [1], due to a surprisingly high increase of the beam current density, so called focusing factor, with reported values up to ~$10^4$ [2]. The well accepted scenario for Slow Highly Charged Ion (SHCI) is a self-organization charge-up guiding, which leads to charge patches on the walls of tapered glass capillary [3, 4]. However, the physics mechanism for fast heavy ions transmission in glass capillary cannot be the same and is still under discussion. Nevertheless, as it is a convenient method for producing micro-beam, capillaries are applied in more and more fields [5]. In order to improve our understanding of the transmission features, we performed experiments on the IRRSUD beam line of GANIL facility (Caen France) for 71 MeV $Xe^{19+}$ and 26.5 MeV $Kr^{10+}$ projectiles transmitted through 2 different tapered glass capillariy shapes, in association with a 2-D PSD for beam cross section measurement. In contrast to the charge-up guiding in the SHCI [5], the transmission for high energy ions has been observed to be dominated by collimation of the beam through the exit aperture (20 μm). A scattered contribution was also clearly visible as a diffuse ring around the central part of the beam. A simple simulation based of Rutherford scattering reproduces quite well the experimental observations. Furthermore, energy loss spectra measurements have shown the correlation between the scattering angle and the ion energy loss (Fig. 1).

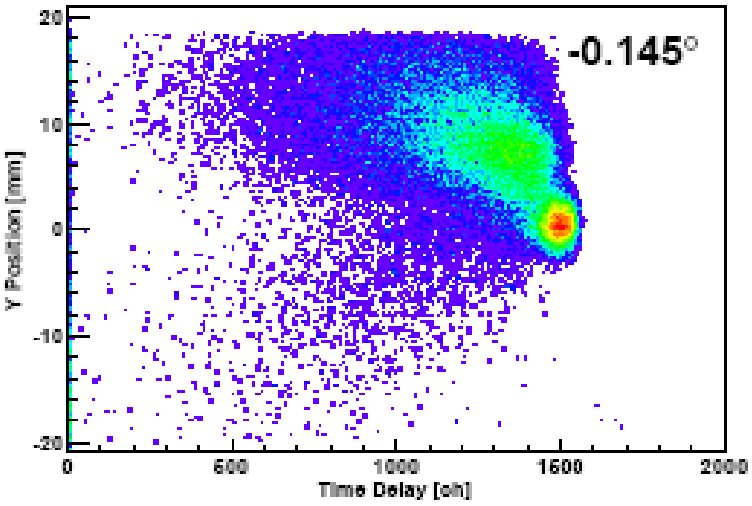


Figure 1. Density plot showing how the energy loss (Time delay) depend on the scattering angle (Y position) for the 71 MeV $Xe^{19+}$ transmitted ions.

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# Material Deposition by Ion Irradiation in Liquid

T. Kobayashi*, T. Ikeda, K. Ogiwara and Y. Yamazaki

*RIKEN, Atomic Physics Laboratory*

Ion irradiation in liquid or at liquid-solid interface is quite promising because it can be applied to surface modification processes [1], novel material production, surface corrosion tests in radiation fields and radiation tests of living subject [2]. In this study, we deposited metals on insulating substrates by ion irradiation in liquid using tapered glass capillaries aiming for a fast and fine patterning process.

The capillaries were produced by pulling both ends of heated glass tubes. They have a lid of glass or plastic film so that the irradiation can be performed in liquid (Fig.1). 3MeV $H^+$ ion beam of a tandem accelerator is focused and guided by small angle scatterings with inner wall of tapered glass capillaries with inserted in a liquid container. The substrates were set perpendicular to the beam at the distance of 100 μm. The irradiation was performed in electroless nickel plating solution containing nickel sulfate and hypophosphorous acid as a reducing agent. Usually, heat-activation (>350K) and palladium catalyst coating are required for nickel deposition.

At the irradiated area, metallic nanoparticles were deposited on the surface without heat-activation or catalysts (Fig.2). The EDX analysis showed the particles consist of nickel containing about 10% of phosphor. The deposited layer had a poor adhesion to the substrates and was easily removed by scratching. The nanoparticles spread on substrates broader than the irradiated area especially toward the downward direction. They diffused over the substrate or grew up at the outer side of the irradiated area and then stuck. The mechanisms of particle growth will be investigated by changing the irradiation conditions.

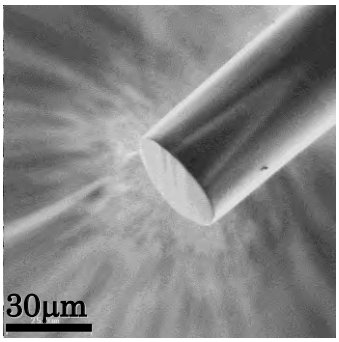


Figure 1. A tip of the glass capillary with a lid shaped by FIB（internal diameter:10μm）

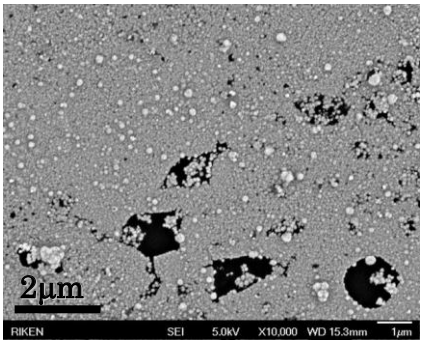


Figure 2. Ni-P nanoparticles covering polyimide substrate formed by proton irradiation

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* t-koba@riken.jp

# Secondary ion emission with energetic cluster beam

Jiro Matsuo[(1,2)*]

*[(1)] Graduate School of Engineering, Kyoto University , [(2)] JST-CREST*

Secondary particle emission provides unique opportunities for further insight on ion collisions with matter. In particular, molecular ion emission from organic or biological molecules is of interest, not only for fundamental studies on excitation of molecules, but also for practical applications. For example, secondary ion mass spectrometry (SIMS) has been widely used for organic and biological material analysis. However, secondary ion yields for organic molecules are very low because of collisions with incident ions. It has been reported that cluster ions can enhance the yields of secondary ions, because of the high-density energy deposition and multiple collisions near surfaces. Clusters such as $SF_5$, $C_{60}$, $Au_3$, $Bi_3$ and $Ar_n$ were found to be quite useful for SIMS of organic materials [1]. Moreover, the velocity of cluster ions is quite low, because the kinetic energy of ion is shared with constituent atoms in the cluster, and therefore cluster ion beam is actually an equivalent low-energy ion beam. This also provides a unique opportunity to sputter materials with little damage formation on the surface.

We have proposed to use large (N>500, where N is the number of atoms) cluster ion beams, which are generated with adiabatic expansion through a small nozzle. Shallow junction formation, surface smoothing, nanofabrication and thin-film formation have been demonstrated with various gas cluster beams. SIMS analysis and molecular depth profiling of soft materials were demonstrated using large Ar cluster ion beams [2–4]. However, there are still many open questions. One of the fundamental issues are cluster-size effects, because the cluster size distribution can range from a few hundred to several tens of thousands atoms. However, measuring the cluster size effects is very difficult because after size selection significant degradation in beam intensity occurs. Recently, we found a way to overcome the difficulties and successfully measured the effect of cluster size on sputtering and secondary ion emission using size-selected cluster ion beams and observed significant lowering of the sputtering threshold energy.

Both experimental and molecular dynamic (MD) simulation results will be presented to propose a possible collision model for large cluster ions, together with the enhancement effect on the yield and lowering of the threshold energy for sputtering.

This work was partially supported by JST, CREST.

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* matsuo.jiro.7s@kyoto-u.ac.jp.

# Soft-sputtering of protein molecules using large cluster ion beams

K. Moritani[(1)*], K. Goto[(1)], I. Ihara[(1)], N. Inui[(1)], and K. Mochiji[(1)]

[(1)] *Department of mechanical and system engineering, Graduate School of Engineering, University of Hyogo, 2167 Shosha, Himeji, Hyogo 671-2201, Japan*

A gas cluster ion beam (GCIB) provides an extremely low-energy projectile compared to conventional polyatomic projectiles, since each gas cluster ion typically contains several thousands atoms, and the energy per atom ($E_{atom}$) should be only several eV. Therefore, the damage of organic molecules can be substantially suppressed, causing the soft sputtering of the protein molecules.

We have investigated the damage formation on highly orientated pyrolytic graphite (HOPG) and the sputtering of DNA molecules adsorbed on HOPG by Ar-GCIB.[1] This study has suggested that $E_{atom}$ was the most important variable to the damage formation on the sample. Moreover, it has been indicated that adjusting $E_{atom}$ of the GCIB can suppress damage on the sample. Motivated by these experimental results, we have developed a size-selected gas-cluster SIMS apparatus[2,3], where $E_{atom}$ can be controlled from several tens eV to below 1 eV by selecting the cluster size and the acceleration voltage of the gas-cluster ion. On applying this instrumentation to several protein and peptide molecules, we demonstrated that the fragmentation is substantially suppressed when $E_{atom}$ was decreased below ~5 eV and intact ions of some protein molecules, for example, insulin (molecular weight: 5,808) and chymotripsin (molecular weight: ~25,000), were detected without using any matrix.[4,5] We have focused on the dependence of the emission of intact ions and fragment ions on the incident direction of the cluster ions as well as accumulated dosage of the cluster ions.[6] The bombardment at a lower angle of incidence significantly enhanced the emission of intact ions. However, the yields of intact ion should be enhanced still more for practical use. In this presentation, results of the biomolecule measurement in SIMS by various gas cluster ion projectiles will be presented.

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* moritani@eng.u-hyogo.ac.jp

# Sputtering and reflection under cluster bombardment of solids

Christian Anders[(1)] and Herbert M. Urbassek [(1)*]

[(1)] *Fachbereich Physik und Forschungszentrum OPTIMAS, University Kaiserslautern, Erwin-Schroedinger-Straße, D-67663 Kaiserslautern, Germany*

Using molecular-dynamics simulation, we study the sputtering induced by energetic impacts of projectile clusters containing up to N = 10000 atoms. Both a metallic target and a van-der-Waals-bonded material are studied. We focus on self-bombardment at perpendicular incidence. The total emission yield is composed of the sputter yield of the target material and the reflection yield of reflected projectile cluster atoms. The dependence of these yields on the impact energy shows a slow transition from the reflection- to the sputter-dominated emission. Similarities and differences between the emission yield of metals and of van-der-Waals-bonded materials are discussed.

* urbassek@rhrk.uni-kl.de

# Cluster Effect on Damage Accumulation in a Si Crystal Bombarded with 10-540-keV $C_{60}$ Ions

K. Narumi[(1), (2) *], H. Naramoto[(2)], K. Yamada[(1)], A. Chiba[(1)], Y. Saitoh[(1)], Y. Morita[(3)], K. Nakajima[(3)], K. Kimura[(3)], Y. Maeda[(2), (4)]

[(1)] *Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency*
[(2)] *Advanced Science Research Center, Japan Atomic Energy Agency*
[(3)] *Department of Micro Engineering, Kyoto University*
[(4)] *Department of Energy Science and Technology, Kyoto University*

When a solid target is bombarded with a molecular/cluster ion in the velocity region where nuclear collisions are dominant, cluster effect on sputtering, displacement of target atoms, *etc.* is observed [1, 2]. The effect on displacement of lattice atoms by molecular-ion bombardment has been reported by many groups: most of them investigated the effect in connection with nonlinear effect on the collision cascade or spike effect. Impact of large cluster such as $C_{60}$ would lead to huge cluster effect. On the other hand, the recent development of application studies using keV $C_{60}$-ion bombardment such as surface-sensitive analyses and secondary-ion mass spectroscopy (SIMS) of high-polymer materials and/or biomaterial has been based on the belief that little damage in the target is induced by its bombardment. However, there have been few studies on the damage accumulation by $C_{60}$ impact. In this study we have investigated damage accumulation in bombardment of a Si crystal with 10-540-keV $C_{60}$ ions.

Pieces of Si(100) wafer treated with an RCA method were irradiated with 10-540-keV $C_{60}$ ions at room temperature. The areal density of displaced lattice atoms was determined from the area of a surface damage peak of backscattering yields of 2-MeV $He^+$ ions. The number of displaced Si atoms per $C_{60}$ ion, $N_{D60}$, has been derived from the fluence dependence of the areal density of displaced Si atoms. Figure 1 shows the result as a function of the energy of $C_{60}$ ions. Comparing with that by C-ion bombardment calculated with SRIM2008, $N_{D1}$, large nonlinear effect on the displacement of Si atoms has been found. The effect is the maximum around 100 keV, where the number ratio, $N_{D60}/(60\times N_{D1})$ is more than 50. Together with result obtained from irradiation at liquid-nitrogen temperature, cluster effect on damage accumulation will be discussed in connection with how dense the collision cascade is.

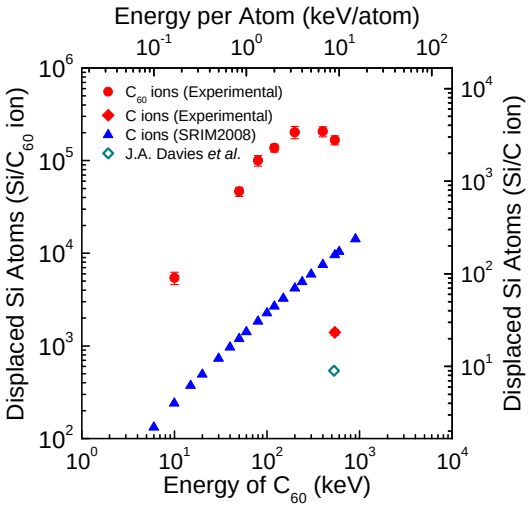


Figure 1. Energy dependence of the number of displaced Si atoms per $C_{60}$ ion. For comparison, those by a monatomic C ion calculated with SRIM2008 and measured in the present study and reference [2] are shown.

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* narumi.kazumasa@jaea.go.jp

# Postcollision Multifragmentation in keV cluster-surface impact

E.Kolodney[1*], A.Kaplan[2], V.Bernstein[1], M.Fleischer[1] A.Bekkerman[1] and B.Tsipinyuk[1]

[1]*Schulich Faculty of Chemistry , Technion-Israel Institute of Technology, Haifa 32000, ISRAEL*

[2] *School of Physics and Astronomy, University of Birmingham, Birmingham B15 2TT UK*

When a large molecule or cluster is impacting a solid target at high velocity (typically a few hundreds of eV collision energy) it experiences a strong deformation which eventually leads to a variety of fragmentation phenomena , manifested by the abundances pattern of the outgoing fragments ,their kinetic energy distributions and angle distributions . Up to a certain impact energy and vibrational excitation (well below its total cohesion energy) the energized cluster is assumed to recoil intact and undergo a statistical unimolecular delayed emission of its subunits. At higher impact and internal energies, multifragmentation (multiparticle break-up) dynamics will start to dominate, eventually reaching complete disintegration of the system into its smallest units (the shattering limit). We have observed and characterized different intermediate multifragmentation modes by scattering $C_{60}^{-}$ ions from surfaces (gold and nickel) at a few hundreds of eV impact energies [1-3]. By analyzing kinetic energy and angle distributions of outgoing $C_n^{-}$ (2=15) fragments for different incidence angles and impact energies we distinguish between multifragmentation events which occur at the surface ("during-collision') and events which occur away from the surface ("post-collision") [1,2]. By measuring and analyzing also positively charged fragments, we show that the multifragmentation dynamics is independent of the charge transfer dynamics [3]. The postcollision multifragmentation is characterized by velocity correlated emission of all outgoing fragments and gradual narrowing of incidence angle dependences of the $C_n^{-}$ fragment yield as a function of the fragment size *n*. A predicted $1/\sqrt{n}$ narrowing law is found to be in good agreement with the experimental results [2]. Molecular dynamics (MD) simulations will also be reported, providing microscopic insight into the nature of the postcollision multifragmentation process [4]. Finally, we will present results related with the collisional formation of the larger $C_{60-2n}^{+}$ (n=1-5) ionic cage fragments [5].

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*eliko@tx.technion.ac.il

JO-I-01

Particle Interactions with Matter at TeV energies and above: the cosmic-ray experience

Spencer Klein
Lawrence Berkeley National Laboratory, and
University of California, Berkeley

Cosmic-rays are the highest energy particles observed by man; particles with energies up to $3*10^{20}$ eV have been observed. Cosmic neutrinos have been observed with energies up to about 1 PeV. In this talk, I will discuss electron and photon and muon interactions with matter, focusing on the special features that become important at very high energies: Landau-Pomeranchuk-Migdal and dielectric suppression of bremsstrahlung and pair production, the growing importance of photonuclear and electronuclear interactions, and, ultimately, at the highest energies, coherent photonuclear interactions with bulk matter. Then, I will discuss muon energy loss, and present some of the ways that large Cherenkov telescopes use these interactions to measure neutrino energy. Finally, I will briefly discuss data from the Auger observatory which sheds light on proton interactions at EeV (above $10^{18}$ eV) interactions.

# Direct measurement of the formation length of photons

K.K. Andersen*, and U.I. Uggerhøj[(1)]

[(1)] *Department of Physics and Astronomi, Aarhus University, Denmark*

We report the first observation of a shoulder in the radiation spectrum from GeV electrons in a structured target consisting of two thin and closely spaced foils. The position of the shoulder depends on the target spacing and is directly connected to the finite formation length of a low-energy photon emitted by an ultrarelativistic electron. The formation length corresponds to the distance it takes for the emitted photon to be separated by a reduced wavelength from the emitting electron. With the present setup it is possible to control the separation of the foils on a micrometer scale and hence measure interference effects caused by the macroscopic dimensions of the formation length. Several theoretical groups have predicted this effect using different methods. Our observations have a preference for the modified theory by Blankenbecler but disagree with the results of Baier and Katkov.

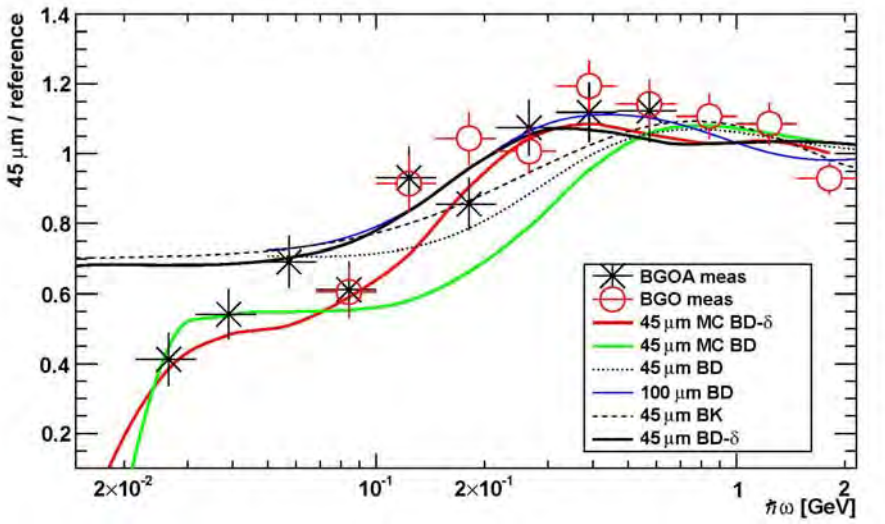


Figure 1. The ratio between the radiation spectra of a 45 micrometer structured target and a reference target.

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* kka@phys.au.dk

# Influence of Stopping on Transition Radiation of Relativistic Heavy Ions Crossing a Target

E.I. Fiks [(1)], Yu.L. Pivovarov [(1)*]

[(1)] *National Research Tomsk Polytechnic University, Tomsk, Russia*

When the relativistic heavy ions (RHI) penetrate through the thin solid amorphous target with a constant velocity, two types of electromagnetic radiation may appear: Cherenkov radiation (CR) (optically transparent target) and transition radiation (TR). The bremsstrahlung is strongly suppressed (compared to relativistic electrons) due to large mass of RHI. In fact, the velocity of RHI slightly decreases due to ionization energy loss (stopping) and it changes the spectral-angular distributions both of CR and TR.

The influence of the stopping on the spectral-angular properties of CR has been investigated recently in [1-4] (see, also our Abstract to this Conference, “Stopping of Relativistic Heavy Ions and its Influence on Angular Distributions of Cherenkov Radiation”).The results of calculations show that the stopping of RHI in radiator leads to additional broadening of CR ring and forming of new CR angular distribution which is different compared with Tamm-Frank distribution.

Here, we present theoretical analysis and results of calculations of spectral-angular properties of TR taking into account RHI stopping in a radiator. The physical reason for appearance of new peculiarities is connected with interference of two waves emitted at entrance and exit of the radiator of finite thickness. These waves are emitted by RHI crossing the boundary vacuum-target and target-vacuum with slightly different velocities, which may change the condition of constructive interference compared to a case of relativistic electrons, see, e.g. [5].

The key parameters here are the plasma frequency, photon energy, attenuation length and thickness of the target, and stopping of RHI, which in turn is a complicated function of the energy, charge and mass of RHI.

The possible applications of the considered effect of stopping on TR from RHI are discussed.

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* pivovarov@tpu.ru

# Effects of Nuclear and Electronic Energy Loss on Damage Formation and Recovery

M. Backman[(1), (2)], F. Djurabekova [(2)], K. Nordlund[(2)], Y. Zhang[(3), (1)], M. Toulemonde[(4)], A. Debelle[(5)], and W. J. Weber[(1), (3)*]

[(1)] *Department of Materials Science and Engineering, University of Tennessee, USA,*
[(2)] *Helsinki Institute of Physics and Department of Physics, University of Helsinki, Finland,*
[(3)] *Materials Science and Technology Division, Oak Ridge National Laboratory,USA,*
[(4)] *CIMAP-CEA-CNRS-ENSICAEN, University of Caen, France,*
[(5)] *Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse, University Paris-Sud, France*

The interaction of ions with solids results in energy loss to both atomic nuclei and electrons. At low energies, nuclear energy loss dominates, and irradiation damage occurs primarily by ballistic collisions. At high energies for swift heavy ions, electronic energy loss dominates and can lead to latent track formation or recovery of existing irradiation damage. At intermediate energies, both nuclear and electronic energy losses are significant and can lead to synergistic or competitive processes that that affect irradiation damage. We have integrated experimental and computational approaches to investigate the separate and combined effects of nuclear and electronic energy loss on damage formation and recovery in several materials. Experimentally, it has been shown that that there is a synergy between the nuclear and electronic energy loss on damage formation in amorphous $SiO_2$ at intermediate ion energies [1]. Large scale molecular dynamics simulations, which include ballistic collisions and/or local electronic heating based on the inelastic thermal spike model, have been employed to investigate the separate and combined effects of nuclear and electronic energy loss on damage production; these simulations demonstrate conclusively the additive effect on nuclear and electronic energy loss on damage production. On the other hand, electronic energy loss in $Ca_2La_8(SiO_4)_6O_2$ leads to competitive damage recovery processes that decrease damage production [2]. In SiC, irradiation with intermediate energy ions leads to defect formation and amorphization [3]; however, it has been shown that swift heavy ions can induce some recovery of such irradiation damage [4]. New experimental results and molecular dynamics simulations reveal that swift heavy ions induce defect recovery and recrystallization in SiC that are well described by an inelastic thermal spike phenomenon.

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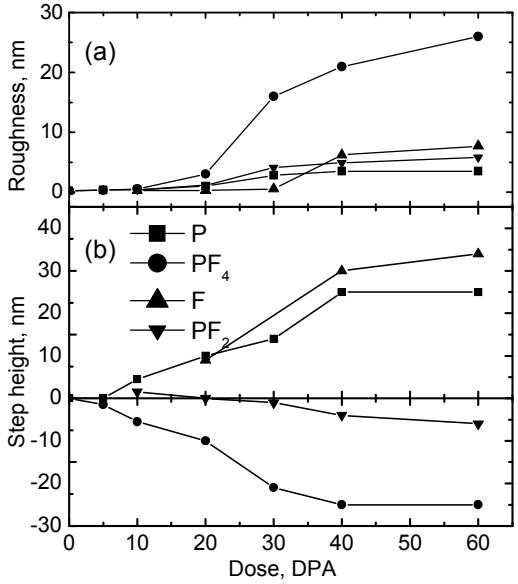
* wjweber@utk.edu

# Molecular Effect on Swelling and Surface Topography of GaN Irradiated by $PF_n^+$ Ions

A.I. Titov,* P.A. Karaseov, V.S. Belyakov, K.V. Karabeshkin, A.V. Arkhipov, and L.M. Nikulina

*St Petersburg State Polytechnic University, Polytechnicheskaya 29, 195251, St.Petersburg, Russia*

Ion-implantation-induced amorphization of GaN is accompanied by material decomposition with the formation of nanoscale $N_2$ bubbles and concomitant changes in the material density and surface topography [1]. As a result, swelling and increase in surface roughness takes place. Under molecular ion irradiation, collision cascades created by components of the cluster ion overlap in the near-surface region, creating combined individual collision cascades with larger volumetric densities of atomic displacements than in the case of irradiation with atomic ions of the same velocity. It results in a significant increase in the level of stable radiation damage in the near-surface region [2] and, accordingly, in change of surface roughness and swelling [3]. An additional effect during molecular ion bombardment is an increase of ion sputtering [4].




In the present report, results of room temperature irradiation of GaN by 1.3 keV/amu $PF_n^+$ (n = 0, 2, 4) and $F^+$ ions will be discussed. Roughness of the surface and height of the step between implanted and masked regions of the sample surface irradiated with different ions (see the legend) are shown in the figure as a function of dose in DPA. It is clearly seen that increase of ion mass (and hence the density of collision cascade) leads to roughness enhancement. Behavior of step height as a function of dose and mass of ions is a result of competition between swelling and sputtering. For $F^+$ irradiation, accumulation of implanted gas atoms has to be taken into account.

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* andrei.titov@rphf.spbstu.ru

# A possible new mechanism for defect formation in irradiated $UO_2$

Desgranges L.[(1)*], Guimbretière G.[(2)], Simon P.[(2)], Jegou C.[(1)], and Caraballo R.[(1)]

[(1)] *CEA/DEN*, [(2)] *CNRS/CEMHTI*

In a previous study we observed the formation of irradiation induced defects in $UO_2$ by Raman spectroscopy [1]. 25 MeV $He^{2+}$ ions were used and should have only induced ballistic defects around the implantation area at 120 µm depth. However Raman damage was observed from the surface to the implantation depth suggesting that the corresponding defects would be formed during electronic stopping of $He^{2+}$ even though their dE/dx is less than the threshold for track formation.

In this paper we examine what could be the mechanism for the formation of these defects observed by Raman spectroscopy. For that purpose, we consider the $UO_2$ intrinsic defect named polaron. The polaron corresponds to an electron-hole pair formation associated to a lattice distortion. It was recently proposed that polaron could act as a supplementary channel for energy dissipation during electronic stopping of swift heavy ions [2]. For dE/dx lower than the track formation threshold, polarons can be formed because their formation energy is only 2eV. We then make the assumption that polaron formation would lead to positively and negatively charged domains. When the charged domains are large enough they could induce charged driven oxygen migration leading to the formation of hypo and hyper stoichiometric domains. This scenario is consistent with the three observed defect Raman modes ($U_1$, $U_2$ and $U_3$) interpretation given in [1]: $U_2$ is due to the activation of the Raman-forbidden LO mode, $U_3$ is due to an over-stoichiometric structural defect of cuboctahedral symmetry and $U_1$ may be the signature of sub-stoichiometric structural defect.

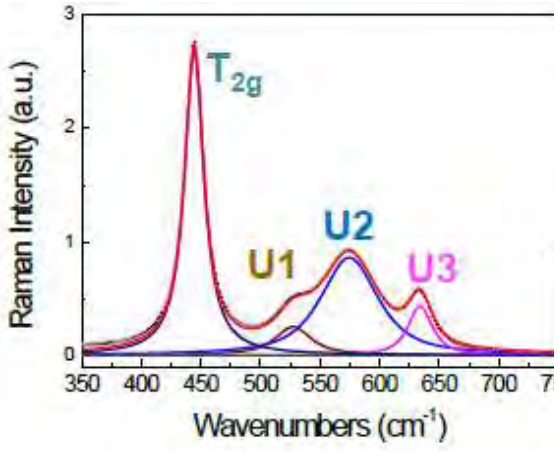


Figure 1. Raman spectra of He irradiated $UO_2$ evidencing UO2 Raman mode (T2g) and three defects modes $U_1$, $U_2$ and $U_3$

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* Lionel.desgranges@cea.fr

# SEM Analysis of Ion Implanted SiC

Johan B Malherbe[1 *], NG van der Berg[1], AJ Botha[1], EKH Friedland[1], TT Hlatshwayo[1], RJ Kuhudzai[1], E Wendler[2], WE Wesch[2] & EF da Silveira

*[(1)] Department of Physics, University of Pretoria, Pretoria, 0002, South Africa, [(2)] Institut für Festkörperphysik, Friedrich-Schiller-Universität, 07743 Jena, Germany, [(3)] Physics Department, Pontifícia Universidade Católica do Rio de Janeiro, Rio de Janeiro, Brazil*

SiC is a material used in two future energy production technologies: As a photovoltaic layer to harness the UV spectrum in high power solar cells, and as a diffusion barrier material for radioactive fission products in the fuel elements of the next generation of nuclear power plants. For both applications, there is interest in the implantation of reactive and non-reactive ions into SiC and their effects on the properties of the SiC.

In this study 360 keV $Ag^+$, $I^+$ and $Xe^+$ ions were separately implanted into 6H-SiC and in polycrystalline SiC at various substrate temperatures. The implanted samples were also annealed in vacuum at temperatures ranging from 900°C to 1500°C for various times.

In recent years there had been significant advances in scanning electron microscopy (SEM) with the introduction of an in-lens detector. This allows SEM images to be taken with very low voltages resulting in more surface sensitive images to be obtained. Examples will be shown of the effect of the accelerating voltage on the SEM image. Examples on cross-sectional samples will be shown of how radiation damage created by the implanted ions can be detected with SEM. The implanted depths and distributions of the ions can also be detected from such samples. Examples of void formation and of conglomeration of the implanted Ag into droplets will be shown. The different ions also led to differences in the recrystallization of the bombardment-induced amorphous SiC.

* johan.malherbe@up.ac.za

# Ultrashort electron dynamics in solids irradiated with a laser or swift heavy ion

B. Rethfeld[1]

*University of Kaiserslautern, Germany*

High-energy laser pulses of subpicosecond duration irradiating metals or dielectrics are primarily absorbed by electrons within the solid. Also swift heavy ions deposit energy mainly in the electronic system of the irradiated material. A common way to describe energy dissipation due to subsequent energy transport and electron-lattice heating is the two-temperature model [1], which was - before the advent of lasers - motivated to describe the interaction of charged particles with metals [2] and is known today in this context as the thermal spike model [3]. The application of the two-temperature model on ultrashort timescales is, however, questionable. For a nonequilibrium electron energy distribution, which does not resemble a Fermi- or Maxwell-distribution, respectively, the concept of a temperature looses its meaning [4].

In this talk I present how kinetic approaches as Boltzmann collision integrals or Monte Carlo simulations identify distinct effects of nonequilibrium energy distributions. Special attention is paid to the influence of the electronic energy distribution on the electron-phonon coupling parameter. Examples shown in the figure are under study [5,6].

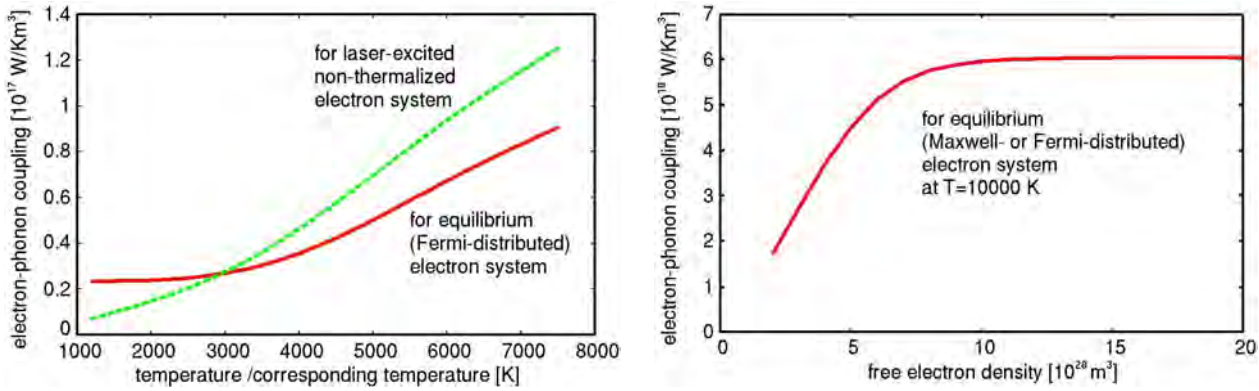


Figure 1. Electron-phonon coupling strength for laser-excited gold in dependence on excitation strength (left, [5]) and for fused silica in dependence on excited electron density (right, [6]).

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[1] rethfeld@physik.uni-kl.de

# Ion tracks: new insights using small-angle x-ray scattering experiments

P. Kluth[(1)]*, B. Afra [(1)], M. D. Rodriguez[(1)], M. Lang[(2)], T. Bierschenk[(1)], M. C. Ridgway[(1)], O. H. Pakarinen[(3)], F. Djurabekova[(3)], K. Nordlund[(3)], C. Trautmann[(4,5)], R. C. Ewing[(2)], M. Toulemonde[(6)], N. Kirby[(7)]

*[(1)] Department of Electronic Materials Engineering, The Australian National University, Canberra, Australia*
*[(2)] Department of Geological Sciences, University of Michigan, Michigan, USA*
*[(3)] Department of Physics and Helsinki Institute of Physics, University of Helsinki, Finland*
*[(4)] GSI Helmholtz Centre for Heavy Ion Research, Darmstadt, Germany*
*[(5)] Technische Universität Darmstadt, Germany*
*[(6)] Centre Interdisciplinaire de Recherche sur les Ions, les Matériaux et la Photonique, Caen, France*
*[(7)] Australian Synchrotron, 800 Blackburn Road, Clayton VIC, Australia*

Although discovered more than five decades ago, there are still many unanswered questions concerning ion track formation, structure and thermal stability. In recent years, we have demonstrated that synchrotron small angle x-ray scattering (SAXS) provides a powerful non-destructive tool for investigating the morphology of ion tracks. SAXS is sensitive to small density changes that often exist in the damaged regions and is well suited for studying tracks in amorphous materials, where most conventional techniques fail. Furthermore, due to the short acquisition times, time resolved measurements enable determination of the annealing kinetics and the stability of ion tracks in environments such as high temperature and high pressure using *in situ* experiments.

This presentation will give an overview of our recent results on ion tracks, including the morphology and annealing kinetics of ion tracks in natural apatite, which are important for geo- and thermochronology. According to our SAXS results, tracks are amorphous cylinders, which recover by a two-step process upon annealing through structural relaxation followed by recrystallization [1]. Tracks in amorphous $SiO_2$ (a-$SiO_2$) reveal a core-shell structure [2] and show an overlap behaviour consistent with a track-core annihilation process [3]. In α-quartz, *in situ* annealing of ion tracks below 600 °C reveals an interesting evolution of the ion track radius that can be well explained by an elastic model using the temperature dependent elastic behavior of quartz and a-$SiO_2$ for the matrix and track material, respectively. SAXS also provides evidence for ion track formation in amorphous silicon, germanium and several amorphous metallic alloys [4]. Results from the SAXS measurements are complemented by molecular dynamics simulations and calculations using an inelastic thermal spike model.

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* patrick.kluth@anu.edu.au

# Nanopores in 1 nm thick carbon-nanomembranes drilled by slow highly charged ions

R. Ritter [(1)*], R. A. Wilhelm[(2)], M. Stöger-Pollach[(3)], A. Mücklich[(2)], U. Werner[(4)], A. Beyer[(4)], S. Facsko[(2)], A. Gölzhäuser[(4)], and F. Aumayr[(1)]

[(1)] Institute of Applied Physics, TU Wien, 1040 Vienna, Austria, EU, [(2)] Helmholtz-Zentrum Dresden Rossendorf, 01314 Dresden, Germany, EU, [(3)] USTEM, TU Wien, 1040 Vienna, Austria, EU, [(4)] Fakultät für Physik, Universität Bielefeld, 33615 Bielefeld, Germany, EU

Nanostructures produced by slow highly charged ion (HCI) impact on surfaces have been a hot topic recently (see [1 - 4] and refs. therein). In this contribution we present first investigations on the effect of individual slow HCI bombardment of freestanding carbon nano-membranes (CNMs). The CNMs are produced by cross-linking of an aromatic self-assembled monolayer of biphenyl units with low-energy electrons [5]. The substrate is then subsequently removed and the resulting nanosheet (1 nm thickness) transferred onto a holey carbon TEM grid. CNMs produced in such a way are irradiated by slow $Xe^{q+}$ ions of various charge states ($20 \leq q \leq 40$) and kinetic energies ($4\ keV \leq E \leq 180\ keV$). After irradiation the CNMs are inspected by high resolution imaging techniques, e.g. TEM, SEM and AFM. On the irradiated CNMs we find nanoscopic pores (3 - 30 nm in diameter, see fig. 1), whose number density corresponds well with the incident ion fluence, indicating that about every HCI produces a nano-hole in the CNM.

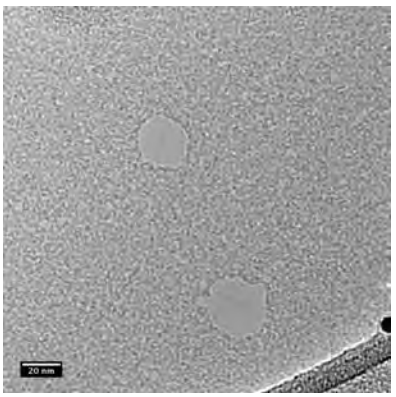


Figure 1: TEM image of 2 holes in a carbon nanomembrane induced by impact of two $Xe^{40+}$ ions ($E_{kin}$ = 40 keV, $E_{pot}$ = 38.5 keV).

First evaluations of the size distribution of the created pores indicate that the average diameter of a hole induced by a given ion depends strongly on the potential energy of the projectile ion, but is also influenced by the kinetic energy.

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* ritter@iap.tuwien.ac.at

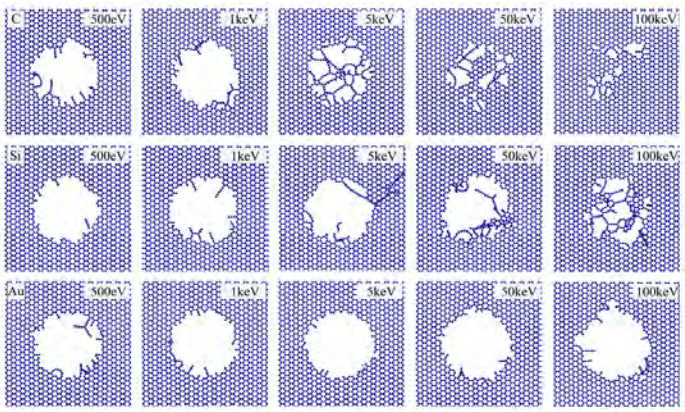
# NanoscaleEngineering of Graphene with Heavy Ion Beams

Shijun Zhao[(1)], Li Liang[(1)], JianmingXue[(1), (2)*], andYugang Wang[(1), (2)]

[(1)]*State Key Laboratory of Nuclear Physics and Technology, School of Physics, Peking University, Beijing 100871, P. R. China*

[(2)]*Center for Applied Physics and Technology, Peking University, Beijing 100871, P. R. China*

Graphene is a two-dimensional carbon nanostructure consisting of $sp^2$-hybridized carbon atoms arranged in a honeycomb-like lattice. Since techniques were developed to exfoliate it from graphite in the form of single layers or multilayers, it has been the focus of intense research due to its unique electronic properties. In view of its wide applications, controllable modification of the properties of graphene is crucial to achieve its optimum performance. In this context, ion beam technique as an effective way has been used to introduce various defects or dope foreign species in graphene in order to engineer its properties.




In this presentation, we have used various energetic ions including Au, C and Si to irradiate the graphene in order to fabricate nanopores in graphene, which holds promise for DNA sequencing, sea water desalination and biosensors, etc. Molecular dynamics simulations based on empirical potentials show that a nanopore with desired size and morphology could be created by carefully choosing the parameter of incident ions. The typical nanopores fabricated with this method are demonstrated in the above Figure, in which we give the shape of the nanopores as a result of different incident ions with various incident energies. We also performed experiments to irradiate the graphene supported by Cu substrate with various ions. It is found that nanopores with predetermined parameters was successfully created, in accordance with the simulation results.

*Author to whom correspondence should be addressed. Electronic mail:jmxue@pku.edu.cn.

# X-ray spectroscopy of the heavy ion interaction with matter

O. N. Rosmej

*GSI Helmholtzzentrum für Schwerionenforschung, Darmstadt, Germany*

Methods of X-ray spectroscopy were applied in investigations of the heavy ion stopping process in cold matter and the response of the stopping media on the deposited ion energy. The evolution of the ion charge and velocity was measured along 80% of the ion stopping path in gaseous and solid targets using the K-shell projectile radiation. The stopping dynamics of heavy ions (Mg, Ar, Ca, Ti, Ni) penetrating solid and gaseous targets with over-range thickness was measured using relativistic Doppler Effect. In order to resolve the ion stopping dynamics in solid targets, low density nanostructures (silicagels) have been used providing stretching of the ion stopping length in condense matter up to 100 times. The method was approved by comparing the ion charge state distribution defined from the X-ray spectra before emerging from the solid target with those measured by means of dipole magnets.

K-shell target radiation measured with a spatial resolution along the ion stopping path was used to investigate the early stage of the ion track formation – the excitation of the target electrons by the dynamical projectile Coulomb field. The application of the K-shell target radiation which occurs in the femtosecond time scale after the Coulomb excitation allowed for the first time investigations of the early stage of the material modification, which supplies the initial conditions for the next stages of the radiation damage process.

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# Inner shell processes in the collisions of highly charged ions near Bohr velocity*

Y. Zhao(1), J. Ren(1), X. Zhou(1), R. Cheng(1), Y. Wang(1), X. Wang(1, 3), Y. Lei(1), Y. Yu(1), Y. Li(1), X. Zhang(2),C. Liang(2), Z. Xu(3), F. Li(3), and G. Xiao(1)

(1) *Institute of Modern Physics, Chinese Academy of Science, Lanzhou730000 China,*

(2) *Xianyang Normal University, Xiangyang, 713000 China*

*(3)Xi,an Jiaotong University, Xi,an 710049 China*

Since the charge state equilibration time is typically in order of femto-second for a highly charged ions impacting into a solid, the initial charge state effect on inner shell processes (ionization and exchanging) between HCIs and target atoms will be significant in collision near Bohr velocity ($0.1\upsilon_{Bohr}<\upsilon<10\upsilon_{Bohr}$, $\upsilon_{Bohr}$=2.19*10^6m/s), where the interaction length before charge equilibrium is around tens to hundreds atomic layers, which makes the initial charge effect possible to be observed, on the other hand, the HCIs will not only have enough kinetic energy to approach a target atom to a certain distance for inner shell ionization, but also have enough time to form a quasi-molecular and exchange the inner-shell electrons or vacancies. The initial charge state effect on inner shell processes will not be so significant, if the projectile velocity is too slow (much slower than $\upsilon_{Bohr}$), the HCIs will be neutralized before a strong inner-shell process happens between projectiles and target atoms, while if the velocity is too fast (much faster than $\upsilon_{Bohr}$, in order of MeV/u), only Coulomb ionization dominates the inner shell processes.

During this conference, we would like to report our preliminary results in measurements of X ray emission induced in the collisions of highly charge ions near Bohr velocity. The experiments were carried out recently at the 320 kV highly charged ion physics platform at IMP-Lanzhou, China. It was found that both the initial charge state and the initial kinetic energy strongly affect the cross-sections of the characteristic X-ray emission from the projectile or from the target atoms. Theories of quasi-molecular can give a reasonable description to those phenomena, while future experiments with very thin foil targets will probably confirm our deductions.

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* zhaoyt@impcas.ac.cn

# X-ray Emission in Fast Collisions of Heavy Ions with Solids

M. Czarnota[(1)], D. Banaś[(1)], J.-Cl. Dousse[(2)], J. Hoszowska[(2)], Y.-P. Maillard[(2)], M. Polasik[(3)], K. Słabkowska[(3)], and M. Pajek[(1)*]

*[(1)] Institute of Physics, Jan Kochanowski University, 25-406 Kielce, Poland, [(2)] Department of Physics, University of Fribourg, CH-1700 Fribourg, Switzerland, [(3)] Faculty of Chemistry, Nicolaus Copernicus University, 87-100 Toruń, Poland*

The x-rays excited by fast heavy ions in solids have a complex structure due to the multiple ionization of atoms leading to emission of x-ray satellites. Generally, such x-ray spectra carry information on the *dynamics* of the collision process as well as the *structure* and *relaxation* of multiply ionized atoms. In particular, the vacancy rearrangement processes taking place between the moments of collision and x-ray emission have to be accounted for in order to describe the measured x-ray spectra. For this reason the understanding of x-ray emission in fast collisions of heavy ions with solids needs adequate theoretical description of the multiple ionization process as well as the extensive calculations of the electronic structure and radiative/nonradiative decay rates for multiple ionized atoms.

In this report we summarize the results of systematic high-resolution studies of x-ray emission from metallic foils (Zr, Mo, Pd and Th) bombarded by energetic (10-25 MeV/amu) oxygen and neon ions. In particular, the x-ray satellites and hypersatellites for $L\alpha_{1,2}$ x-ray transitions in Zr, Mo and Pd [1] and $M\alpha_{1,2}$ x-ray transitions in Th excited by fast oxygen and neon ions were measured using a high-resolution diffraction von Hamos spectrometer [2] having an instrumental energy resolution below 1 eV. The x-ray hypersatellites for the L- and M-shells were observed for the first time. The measured x-ray spectra, including x-ray satellites and hypersatellites, were interpreted in terms of the relativistic Multi-Configuration Dirac-Fock (MCDF) calculations [3] which were performed for multi-vacancy configurations ($L^{-l,2}M^{-m}N^{-n}$ and $M^{-m}N^{-n}$) ($n,m \leq 4$) expected to contribute to the observed x-ray spectra. For higher number of vacancies the developed model of averaged <MCDF> calculations [4] was used. The performed MCDF calculations allowed to interpret the complex structure of x-ray satellites for $L\alpha_{1,2}$ and $M\alpha_{1,2}$ transitions taking into account the vacancy rearrangement processes. Finally, the ionization probabilities per electron for the moment of collision were extracted from the measured spectra of x-ray satellites. The ionization probabilities were compared with the predictions of the semiclassical approximation (SCA) using the relativistic hydrogenic (HYD) and selfconsistent Dirac-Hartre-Fock (DHF) [5] wave functions. Generally, it was found that realistic selfconsistent (DHF) description of the wave functions for the L- and M-shell electrons was necessary in order to interpret the measured ionization probabilities.

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* pajek@ujk.edu.pl

# The History of Swift Heavy Ions in Matter

R. Neumann[1] and M. Toulemonde[2]

*(1) GSI Helmholtz Centre for Heavy Ion Research, Darmstadt, Germany, (2) Centre Interdisciplinaire de Recherche sur les Ions, les Matériaux et la Photonique (CIMAP), Caen, France*

The first SHIM Symposium was held in 1989, promoted by J. C. Jousset from the CIMAP laboratory in Caen (France) and P. Armbruster from GSI Helmholtz Centre for Heavy Ion Research, Darmstadt, (Germany). With this initiative, they pursued the aim of developing a better understanding of material transformations under dense and nanometric excitations induced by heavy ions in the electronic energy loss regime. Since the excitations are mainly reached with ions at high velocities, the attribute "swift" was chosen to characterize this field of research. We present an overview of the SHIM Conferences [1], considering their major contributions to topics such as fundamental processes, materials, nanosystems, and applications.

**Part I: Interactions of swift heavy ions with matter**

Swift heavy ions create in many solids straight, cylindrical, and nanometric damage trails, called ion tracks. These tracks result from a three-step process: (1) MeV to GeV projectiles generate excitations and ionizations in the electronic system of the atoms; (2) this energy is transferred from the electrons to the atoms; (3) the energy is dissipated in the material. The SHIM community has combined atomic physics, solid state physics, and materials science to study the track creation process and its final results. In this part, it will be shown how the understanding has improved over the last two decades and an attempt will be offered to clarify the present status of our knowledge.

**Part II: Applications on the nanoscale**

Ion tracks represent fascinating nanostructures that can be transformed via etching into open nanochannels. These channels can be modified specifically to control small species and act, e.g., as sensors and transmitters. Depositing a material in the channels provides nanowires that also exhibit various novel effects. In this part, microtechnological achievements with swift heavy ions will be briefly recollected that began already in the 1970ties, preparing the ground for gradual size decrease down to the nanoscopic objects now under study. Various examples illustrating recent results and emerging developments will be displayed, comprising material modifications on the nanoscale, plasmonic and thermoelectrical effects in nanostructures, and novel applications of nanochannel- and nanowire-based devices.

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E-mail: r.neumann@gsi.de and toulemonde@ganil.fr

# Lindhard Lecture:
# Short-time Processes Triggered by Fast Ions in Solids

Gregor Schiwietz [(1)*]

[(1)] *Helmholtz-Zentrum Berlin f. Materialien u. Energie, Institute G-I2, Hahn-Meitner-Platz 1, 14109 Berlin, Germany*

During the passage of solid or liquid matter, ions transfer their kinetic energy into different electronic and atomic excitation modes. Notwithstanding the complication that arises due to the large variety of ion-solid interaction processes, the primary interaction is extremely localized (typically on an atomic scale) and also very rapid (for fast ions). The time duration of individual energy-transfer processes is about $10^{-16}$ s or less, far below typical relaxation times. Since our current knowledge on energy-loss processes has reached a high level of sophistication [1], fast ions are ideal candidates for short-time investigations specifically of electronic relaxation processes in solids.

In this lecture, fast ions will be compared to pulsed photon sources in their ability as triggers and probes of short-time processes. Different detection schemes for short-time dynamics driven by fast ions in solids will be discussed. Special emphasis will be put on high-resolution Auger spectroscopy [2] (see also Fig. 1) for the investigation of rapid electronic and atomic [3] processes. Our current knowledge will be sketched and open questions will be high-lighted, specifically those regarding our understanding of atomic reactions triggered by dense electronic excitations.

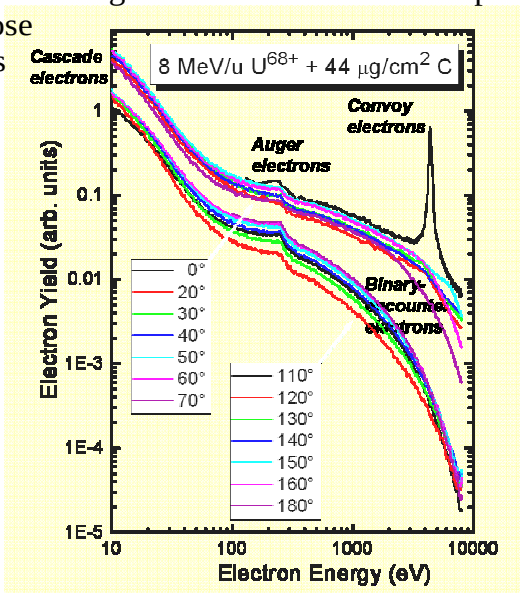


Figure 1.:
Electron-energy distribution for emission angles between 0 and 180 degrees with respect to the ion beam. Measured for highly charged uranium ions at normal incidence on a thin amorphous carbon foil [experimental data taken from D.Schneider, G.Schiwietz, and D.DeWitt, Phys. Rev. A 47, 3945 (1993)].

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* e-mail address for correspondence: schiwietz(AT)helmholtz-berlin.de .

# Beyond the ion-beam shaping mechanism: toward plasmonic applications

**G.Rizza[1)*],** P.E. Coulon[1)], A.Slablab[1)], J. Amici[1)], S.Perruchas[2)], T.Gacoin[2)], I.Monnet[3)], C.Dufour[3)], V. Khomenkov[3)], J.Cardin[3)], A.Fafin[3)], M.Kociak[4)], A. Losquin[4)], D.Mailly[5)], C.Ulysse[5)], X. Lafosse[5)]

*1) LSI, Ecole Polytechnique, 91128 Palaiseau Cedex, France, 2) LPMC, Ecole Polytechnique, 91128 Palaiseau Cedex, France, 3) CIMAP, 14070 Caen Cedex 5, France, 4) LPS, Bâtiment 510, Université Paris Sud XI, F 91405 Orsay, France, 5) LPN, 91460 Marcoussis, France*

In the last years, ion-shaping technique has been proposed as an innovative and powerful tool to manipulate matter at the nanometer scale [1-3]. Deformation can be indirectly induced by embedding metallic NPs into an ion-deformable amorphous host matrix.

A model system composed of nearly monodisperse metallic nanoparticles (NPs) (8-100nm) confined between two silica layers is used to investigate the ion-shaping mechanism. We show that this phenomenon is not limited to the transformation into prolate nanorods and/or nanowires, but that depending on the initial size of the NPs, several new classes of ion-shaped NPs can also be obtained: i) facetted-like NPs, ii) nanowires growing from a facetted core or iii) chromosome-like NPs. In parallel, the evolution of the temperature profile within the nanoparticle is simulated by implementing the thermal-spike model for three-dimensional anisotropic and composite media [4]. In this way, a straight correlation is found between the fraction of the nanoparticle that is molten(vaporized) and the deformation path followed by the nanoparticles during the irradiation. This allows the construction of a size-vs-shape diagram relating the initial nanoparticle size to its final morphology. This diagram is used to give a rational description of the ion-beam shaping process for all the nanoparticle dimensions [5].

Besides the fundamental aspects related to the ion-matter interaction, ion-shaping can also be used to give new insights into the plasmonic properties of metallic nanorods and nanowires. Here, Electron Energy Loss Spectroscopy (EELS) is used to study Localized Surface Plasmon Resonances (LPSR) in ion-shaped metallic nanoparticles with a nanometer-scale spatial resolution, [6]. LSPR are generated through electron excitation is a Scanning Transmission Electron Microscope (STEM), equipped with a High Angle Annular Dark Field (HAADF) detector. As the NPs are continuously deformed under irradiation, we investigate the LPSRs dependence on both the geometry and aspect ratio of the nanostructure. Finally, the experimental results are simulated using a specifically developed Auxiliary Differential Equations-Finite Difference Time Domain (ADE-FTDT) code, [7].

This work demonstrates the possibility to use ion irradiation as tool for the controllable fabrication of a whole family of plasmonic nanostructures with topologically tunable optical properties. These ion-beam shaped composite media have potential applications spanning from plasmonic photovoltaics, to bio-sensing, SERS and SEIRA spectroscopies.

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**Keywords**: ion-beam shaping, metallic nanoparticles, plasmonics
**Corresponding author***: giancarlo.rizza@polytechnique.edu

# Synthesis of Semi-Embedded Au Nanostructures by Ion Irradiation: Combined Effect of Sputtering, Viscous Flow and Ion Recoil Implantation

Udai B. Singh [(1)*], D. C. Agarwal[(1)], S. A. Khan[(1)], A. Tripathi[(1)], H. Amekura[(2)], D. P. Datta[(2)], Ajay Kumar[(3)], R. K. Choudhury[(3)], B.K. Panigrahi[(4)], T. Osipowicz[(5)], D. K. Avasthi[(1)]

*[(1)] Inter-University Accelerator Centre, New Delhi, India, [(2)] National Institute for Materials Science, Tsukuba, Japan, [(3)] Nuclear Physics Division, Bhabha Atomic Research Centre, Mumbai, India, [(4)] Materials Science Group, Indira Gandhi Centre for Atomic Research, Kalpakkam, India, [(5)] National University of Singapore, Singapore*

Ion beam synthesis is one of the techniques to synthesize the nanostructures [1-3]. Noble metal nanostructures are of great interest due to their high surface to volume ratio, surface plasmon resonance and applications in different fields. Formation of surface nanostructures by ion irradiation of thin films occurs mainly due to interplay between the surface instability and surface diffusion [2]. Semi embedded Au nanostructures were synthesized by 50 keV Si ion irradiation of 5 nm Au thin films on glass. High resolution Rutherford backscattering spectroscopy (HRBS) provided evidence of embedding of Au atoms into substrate irradiated sample at fluence of $3\times10^{16}$ ions/cm$^2$. The formation of semi embedded nanoparticles having an average diameter of 16 nm was revealed from the cross sectional transmission electron microscopy (XTEM) characterization of irradiated sample at fluence of $3\times10^{16}$ ions/cm$^2$. The mechanism of synthesis is explained by ion beam induced sputtering, thermodynamically dominated capillary forces [4] and recoil-implantation of Au [1].

Similar results were also obtained for 50 keV Ar ion irradiation of thin Au film from the fluence of $1\times10^{15}$ ions/cm$^2$ to $3\times10^{16}$ ions/cm$^2$. RBS spectra of pristine and irradiated samples reveals that Au peak shifts to the lower energy with decrease in the intensity of peak along with tailing in the lower energy edge with irradiation which indicates that sputtering and recoil-implantation occur during irradiation. The absorbance spectra of pristine and irradiated samples show that ion irradiation leads to the formation of plasmonic structures. Semi-embedded nanostructures have great potential for use as Surface Enhanced Raman spectroscopy (SERS) substrate. The test for SERS will be performed in near future.

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* udaibhansingh123@gmail.com

JO-I-07

# Heavy Ion Sputtering of LiF, astrophysical ices and silicates

P. Boduch[(1)] and H. Rothard [(1)*]

[(1)]*Centre de Recherche sur les Ion, les Matériaux et la Photonique (CEA/CNRS/ENSICAEN/UCBN), CIMAP-CIRIL-Ganil, BP5133, 14070 Caen Cedex 05, France*

Ion-surface collisions involving swift heavy ions (MeV/u), where the electronic energy loss $S_e$ is dominant, lead to "electronic sputtering". The microscopic mechanisms are still under debate (coulomb explosion, thermal spike, excitonic mechanisms, etc.). The measurement of yields and of energy- and angular distributions of sputtered particles contribute to the understanding of the initial microscopic processes of defect creation in materials. We present recent results on heavy ion sputtering of different materials. A new UHV set-up allows measuring the velocity vector of secondary ions from controlled surfaces including thin layers [1] by means of the imaging technique XY-TOF-SIMS ("time of flight-secondary ion mass spectroscopy"). In addition to mass distributions of emitted particles, their energy- and angular distributions can be deduced. Results obtained with LiF crystals [1,2] and evaporated thin layers [3] show an important contribution of $Li^+(LiF)_n$ cluster emission. As first application to materials relevant for astrophysics (exposure of surfaces to cosmic rays or solar wind) we show sputtering yields and energy distributions for nepheline, a silicate being a model for mercury's surface [4]. We also present results obtained with astrophysical ices at 15K by FTIR infrared absorption spectroscopy. Although protons are orders of magnitude more abundant in the cosmic ray spectrum, the high electronic energy loss of heavy ions can compensate for the lower flux. This results in non-negligible contributions of heavy ion induced physico-chemistry. In particular, we observed a strong increase of sputtering yields Y with deposited energy $Y \sim S_e^2$ in CO and $CO_2$ ices bombarded with heavy ions [5,6]. Clearly, the contribution of cosmic ray heavy ion induced desorption from dust grains must be taken into account for understanding the presence of gas phase molecules in dense interstellar clouds [6].

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[*]rothard@ganil.fr

# Microscopic analysis of particles sputtered with heavy ions in the electronic stopping regime

Christina Trautmann[(1,2)*], Walter Assmann[(3)], Arndt Mücklich[(4)], Marcel Toulemonde[(5)], Andreas Welzmüller[(3)]

*[(1)] GSI Helmholtzzentrum für Schwerionenforschung, Darmstadt, Germany, [(2)] Technische Universität Darmstadt, Darmstadt, Germany, [(3)] Ludwig Maximilians Universität München, Garching, Germany, [(4)] Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany, [(5)] CIMAP laboratoire, CEA, CNRS, ENSICAEN, Univ. Caen, Caen, France*

For a variety of ionic crystals in particular LiF and $CaF_2$, sputtering processes with swift heavy ions have been studied in detail. In most cases, the total number of sputtered particles and their angular distribution were quantified by using the catcher technique in combination with elastic recoil detection analysis (ERDA) [1,2]. The most prominent phenomena are sputtering at rates much larger than values known from elastic sputtering and unexpected angular distributions characterized by a sharp jet-like component which is symmetric around the surface normal and superimposed on a cosine distribution.This contribution presents new transmission electron microscopy (TEM) investigations of fluorite particles sputtered onto TEM grids as catchers (Fig. 1). The analysis at different angular positions reveals that the jet-like component is correlated with the ejection of large nanocrystals of similar size (up to 10 nm), whereas outside the jet-like component, the size distribution of the particles is broad.

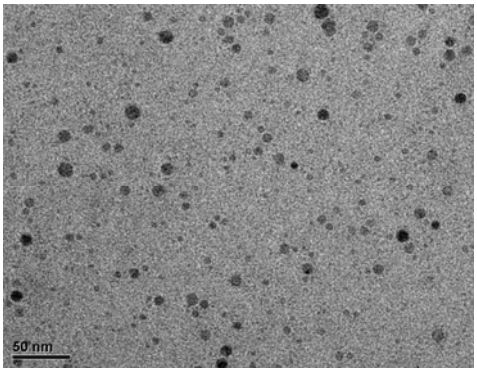


Figure 1. TEM image of catcher showing nanoparticles sputtered from a $CaF_2$ single crystal when irradiated with 180-MeV Au ions under 45º beam incidence. The sputtering experiments were performed at the Munich tandem.

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* c.trautmann@gsi.de

# Ion Tracks in Non-amorphizable Non-radiolytic Materials – a Revision of the Thermal Spike Concept

S. Klaumunzer[(1)]

[(1)] *Helmholtz-Center for Materials and Energy, Department G-A1, Berlin, Germany*

The basis for any thermal spike model is a combination of irreversible thermodynamics and continuum mechanics to fulfill conservation of mass, momentum, angular momentum, energy, and the second law of thermodynamics. Starting from this basis the rudimentary form of the presently used thermal spike models for swift heavy ions becomes obvious. By means of a fictive material with simplified material properties, quantitative differences between those models and a more advanced one will be outlined. It will be shown that a more complete model provides a natural basis to distinguish materials showing no tracks, amorphous tracks or more complicated track effects like hollow tracks or tracks with reduced mass density in the core. Particular emphasis will be put on the recently discovered phenomenon of ion hammering in originally single crystalline NiO. It will be argued that dislocation emission is inevitable if the thermal spike is sufficiently intense. Three electronic energy loss regimes can be distinguished. At low energy losses a non-radiolytic single crystal can remain essentially undamaged if transient melting is followed by epitaxial recrystallization. At intermediate energy losses essentially shear dislocation loops will be generated, and at very high energy losses prismatic loops will dominate. The latter are the starting point for the formation of dislocation networks, dislocation cells and, finally, to small-angle boundaries leading to an increase in crystal mosaicity or, depending on irradiation geometry, to the formation of nano-crystals and crystallite rotation.

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klaumuenzer@helmholtz-berlin.de

# Computer Simulation of SHI Effects in Materials

Z. Insepov[(1)*], Norman G.E.[(2)], Pisarev V.V.[(2)], Starikov S.V.[(2)], Stegailov V.V.[(2)], Yanilkin A.V.[(2)], and Terasawa M.[(3)]

*[(1)] Argonne National Laboratory, IL, USA, [(2)] Joint Institute for High Temperature of RAS, Moscow, Russia, [(3)] LASTI, University of Hyogo, Hyogo, Japan*

Interactions of swift heavy ions (SHI) with matter are of fundamental interests since they advance our understanding of high-energy accelerators, muon cooling, particle detectors by giving us an insight into the nature and properties of warm dense matter. In the present paper, the following phenomena are of interests such as (multiple-charged) plasma formation at SHI interaction with materials, charge screening and neutralization, shock wave generation, ion track and crater formation, and sputtering by SHI [1-14]. Sputtering of solids by SHI is interesting from both fundamental and potential application viewpoints. Experimental sputtering data obtained by SHIM were not explainable by a simple linear cascade theory [1]. Computer simulations of sputtering by SHI interaction with solids were capable of confirming at least some of the experimental observations [10,11]. Several new phenomena were discovered recently such as phase transitions by irradiation with SHI [2], atomic mixing in thin films by SHIs [3], crater formation on the surfaces [4], desorption from amorphous carbon [5], sputtering of large organic molecules from the surfaces by shock waves generated by SHIM [6] that are in the interests of our paper. We also discuss a recent development of computer simulation methods that were significant tools for understanding of the above fundamental and applied topics related to SHIM, including two-temperature molecular dynamics (TTM) [15,16] capable of studying the dynamics of hot electrons in the solids irradiated with SHI by introducing an electron-phonon coupling into classical molecular dynamics [9]; and coupling of TTM with the plasma model [8].

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* Corresponding author: insepov@anl.gov.

# Role of Ion Track Density Evolution in Void Formation in Amorphous Ge and in Metal Nanoparticle Shaping

O. H. Pakarinen[(1)1], A. A. Leino[(1)], R. Giulian[(2)],T. Bierschenk[(2)], M. C. Ridgway[(2)], P. Kluth[(2)], O. Osmani[(3)], N. Medvedev[(4)], M. Schleberger[(3)], B. Rethfeld[(4)], F. Djurabekova[(1)], and K. Nordlund[(1)]

*[(1)] University of Helsinki, Finland [(2)] Australian National University, Canberra, Australia*
*[(3)] University of Duisburg-Essen, Germany [(4)] TU Kaiserslautern, Germany*

Combination of molecular dynamics (MD) simulations and experiments reveals nanoscale structure and time evolution in latent ion tracks. SHI irradiation induces porosity in amorphous Ge. As precursors, unusual bow-tie shaped voids are observed in transmission electron microscopy (TEM). MD and analytical models show how the density changes at phase transitions govern both the initial void formation in the molten ion track and its shape change out of the minimum surface energy spheroidal shape upon solidification.

In a comparison of small-angle x-ray scattering experiments and MD simulations, a previously unresolved fine structure with a low-density core and a high-density shell in ion tracks in amorphous silica was found [1]. The origin of the fine structure is a picosecond time scale pressure wave out of the track center that freezes in. We present large-scale MD simulations of SHI irradiation into Au nanoclusters in silica, compared to experiments where nanoparticles elongate into long nanorods. Simulations show how the track density evolution and thermal expansion of gold govern the shape transformation of nanoparticles [2].

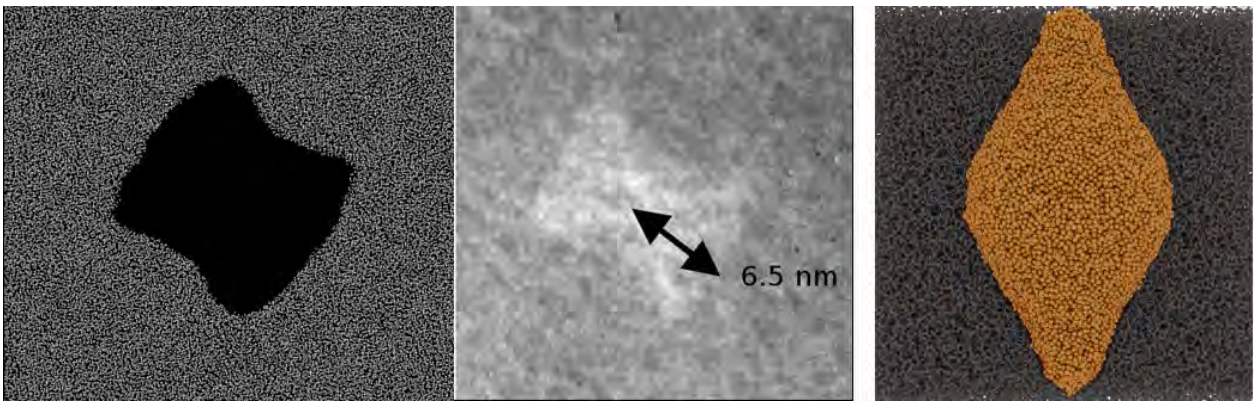


Figure 1. (left) Void in amorphous Ge ion track, MD simulation
(center) Bow-tie shaped void in aGe, TEM experiment
(right) Elongation of originally spherical gold nanoparticle in $SiO_2$ after multiple ion impacts in MD simulation

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[1] olli.pakarinen@helsinki.fi

SH-I-02

# SWIFT HEAVY ION INDUCED THERMAL SPIKE ENGINEERING OF THIN FILMS: MODIFICATION AND EVOLUTION OF NANOSTRUCTURES

*S. Ghosh, H.Kumar, S. P. Singh and P. Srivastava*
*Nanostech laboratory, Indian Institute of Technology Delhi, New Delhi-110 016, India.*

*D. Kabiraj, S. A. Khan, D. Kanjilal and D. K. Avasthi*

*Inter University Accelerator Centre, Aruna Asaf Ali Marg, New Delhi-110067, India*

High energy heavy ions popularly known as swift heavy ion (SHI) can be used as a unique tool to engineer the properties of materials down to nanoscale because of large energy deposition in the material. The energy deposited in the electronic subsystem eventually leads to a high temperature spike (in (subnanosecond time scale) within a narrow spatially confined (few nm in dimension) zone of the lattice. After a brief introduction to the basic mechanism of SHI-material interaction, the following three studies will be described: (i) elongation of nickel (Ni) and Au nanoparticles in thin $SiO_2$ film matrix and associated modification in optical and magnetic properties, (ii) evolution of Si nanostructures in Si rich silicon nitride films and (iii) evolution of nanodimensional structures and nanometallic lines on copper nitride thin films under SHI irradiation. The possible mechanism based on thermal spike mechanism and simulation explaining these effects will be discussed. Finally, future research direction and possible applications will be highlighted.

**Key words:** (i) Swift heavy ion, (ii) Thermal spike, (iii) Nanocomposite films, (iv) Silicon nitride, (v) Copper nitride.

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Author's contact: e mail: ghoshsantanu1@yahoo.co.in, Phone: 91-11-26591348, Fax: 91-11-26581114.

# Effects of 2.6 GeV U Irradiation in Co-doped $BaFe_2As_2$

T. Tamegai[(1)*], H. Yagyuda[(1)], T. Taen[(1)], Y. Nakajima[(1)], and T. Kambara[(2)]

*[(1)] Department of Applied Physics, The University of Tokyo, [(2)] Nishina Center, RIKEN*

The discovery of superconductivity in LaFeAs(O,F) has revived the research activity on superconductors [1]. Following this discovery, various iron-based superconductors (IBS) with layered structures have been found. For the applications of IBS, critical current density, $J_c$, is one of the most important parameters. Earlier studies on polycrystalline samples of IBS clarified that $J_c$ of the order of $10^5$-$10^6$ A/cm$^2$ flows within the grain, while intergranular $J_c$ is orders of magnitude smaller [2]. This fact has been later confirmed by studies on single crystals [3]. Introduction of disorder in a controlled way offers an excellent opportunity for enhancing $J_c$. Such an improvement of $J_c$ in IBS by defects created by swift ions has been demonstrated [4]. Depending on the energy and mass of particles, created defects are either point defects or continuous columnar defects [5].

In the present study, we explore the effect of 2.6 GeV U irradiation on $J_c$ and vortex dynamics in $Ba(Fe_{0.925}Co_{0.075})_2As_2$ [5]. We focus on the origin of the novel low-field dip feature in *M-H* hysteresis loop as shown in Fig.1. This dip feature is wiped away by misaligning the field direction ($\theta_H$) from the direction of columnar defects ($\theta_{CD}$). Such a suppression of $J_c$ near zero field can be caused by the curvature of vortices in flat superconductors close to zero field as shown in Fig. 2(a). When magnetic field is increased to the self-field ($H_{sf}$), vortices are straightens up as shown in Fig. 2(b) and the efficiency of columnar defects becomes higher, giving higher $J_c$.

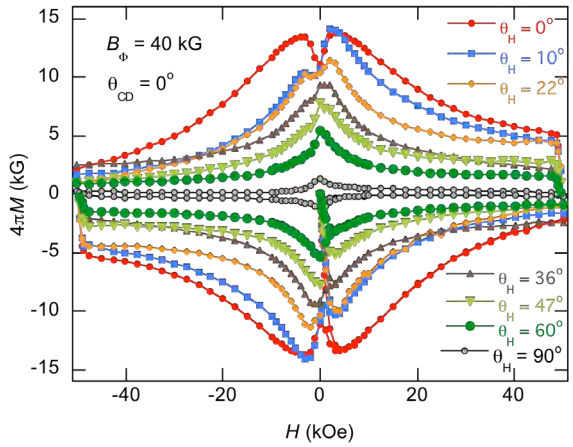


Figure 1. *M-H* hysteresis loops at 5 K in 2.6 GeV U irradiated $Ba(Fe_{0.925}Co_{0.075})_2As_2$ at several field angles.

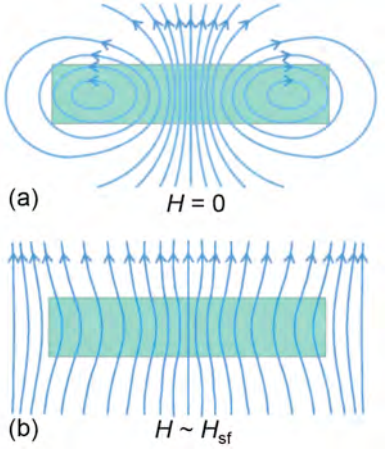


Figure 2. Schematic magnetic field lines in a flat superconductor at (a) zero field and (b) $H \sim H_{sf}$.

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* tamegai@ap.t.u-tokyo.ac.jp

# Swift Heavy Ion Induced Sputtering in $BaF_2$ Thin Films

Ratnesh K. Pandey[(1)*], M. Kumar[(1)], Saif A. Khan[(2)], D. K. Avasthi[(2)], Avinash C. Pandey[(1)]

[(1)]*Nanotechnology Application Centre, University of Allahabad, Allahabad, India 211002*
[(2)]*Inter University Accelerator Centre, New Delhi, India 110067*

The effect of swift heavy ion bombardment on thin films have a great variety of applications in understanding the theoretical modelling of the basic electronic sputtering phenomenon which includes the observation of change in sputter yield with the change in film parameters and of the sputtered species ejected from the film surface due to ion bombardment. This is what we have done in our present experiment with a series of barium fluoride thin films of different thicknesses and deposited by electron beam evaporation technique at room temperature on silicon substrates. The effect of film thickness on the electronic sputter yield of polycrystalline $BaF_2$ thin films has been reported in the present work. The thin films of $BaF_2$ have have been deposited with three different thicknesses 20 nm, 50 nm and 150 nm on Si substrates and sputtering has been performed by 100 MeV $Au^{+25}$ ions. Glancing angle X-ray diffraction (GAXRD) measurements show that the pristine films are polycrystalline in nature and the grain size increases with increase in film thickness. Rutherford backscattering spectrometry (RBS) of pristine as well as irradiated films was done to determine the areal concentration of $BaF_2$. A reduction in the sputter yield of $BaF_2$ films with the increase in film thickness has been observed from the RBS spectra. The thickness dependence sputtering is explained on the basis of thermal spike and the energy confinement of the ions in the smaller grains.

*Email- pandeyratneshk@gmail.com

# From Grain Fragmentation to Grain Rotation:

# Swift Heavy Ion Bombardment of Single Crystalline NiO

D. Severin[(1)*], M. Bender[(1)], A. Delgado[(2)], S. Klaumünzer[(3)]

*[(1)] GSI Helmholtz Centre, Darmstadt, Germany; [(2)] USP, Sao Paulo, Brazil; [(3)] Helmholtz Centre for Materials and Energy, Berlin, Germany*

In insulators with complex crystal structures, e.g. silicates or garnets, ion tracks are often amorphous and can easily be identified by high resolution electron microscopy. At high ion fluences, track overlap leads to complete amorphization. In insulators with simpler crystal structures or composition, tracks can result in a change in crystal structure without amorphization at high fluences. Under ion bombardment, binary fcc insulators do neither amorphize nor change their crystal structure. Some of them, e.g. alkali halides or $CaF_2$, are destroyed because they are easily cleavable and decompose by radiolytic processes, while others, e.g. MgO and $UO_2$ are considered as very radiation resistant.

Single crystals of NiO at room temperature were bombarded with 600 and 940 MeV gold ions. Crystal modifications were monitored by means of on-line x-ray diffractometry with a four-circle diffractometer. We observed pronounced effects depending on the angle of incidence $\alpha$ between the impinging ions and the sample surface normal. During normal beam incidence, i.e. $\alpha = 0$, a (111)-oriented crystal remains an intact single crystal with an increase in mosaicity saturating at about $3\times10^{13}$ Au/cm$^2$. At fluences $>10^{14}$ Au/cm$^2$ target destruction starts from the sample rims, a phenomenon which indicates large internal mechanical stresses. For $\alpha \geq 20°$, single crystals rapidly ($\sim10^{13}$ Au/cm$^2$) transform into well-aligned nanocrystals which change their orientation by crystallite rotation proportional to the ion fluence. In highly symmetric cases, i.e. when the projection of the ion beam onto the sample surface is parallel to one of the cubic crystal axis, the rotation axis coincides also with a crystal axis. In this case the rotation can reach more than 90° at sufficiently large fluences. The process is almost perfectly reversible, because changing the sign of $\alpha$ reverses the direction of rotation. In less symmetric cases there is an additional side motion which hampers the pursuit of the x-ray signal and destroys the reversibility.

Similar effects were also seen in MgO, UO2, TiN [1], and ω-Ti [2]. The occurrence of grain fragmentation and grain rotation seems to be a rather universal phenomenon but requires very high electronic stopping powers. On the other hand, a similar phenomenon was also reported for nanocrystalline gold bombarded with 7 MeV gold ions [3].

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* d.severin@gsi.de

# PARAMAGNETIC TRIGONAL CENTER PRODUCTION IN YTTRIA-STABILIZED ZIRCONIA BY ELECTRONIC EXCITATIONS

**Jean-Marc Costantini*,**

**CEA, SRMA, 91191 Gif-sur Yvette Cedex, France**

**and François Beuneu,**

**LSI, CEA-CNRS-Ecole Polytechnique, 91128 Palaiseau Cedex, France.**

**ABSTRACT**

Electron paramagnetic resonance (EPR) spectroscopy is used to study the point defect production in yttria-stabilized zirconia (YSZ) (100) single crystals by swift heavy ion, electron and X-ray irradiations. A common color center known as the “T center” (for “trigonal” center) with an axial <111> symmetry is produced in all cases. We show that this defect is likely an intrinsic one with concentrations much larger than the major impurities. The growth curves of this point defect versus fluence for ion and electron irradiations can be on the whole rescaled as a function of the absorbed dose. This confirms that T centers are produced by the electronic excitations, either at low density with X-rays and electrons, or at high density with heavy ions. The production rate increases almost linearly versus the average volume density of electron-hole pairs that are generated by ions and electrons in the irradiated volume.

* **Corresponding author, Email address : jean-marc.costantini@cea.fr**

# High Energy Heavy Ion Therapy -Recent Activities at HIMAC NIRS-

T. Kamada*(1)

(1) *Research Center for Charged Particle Therapy, National Institute of Radiological Sciences, Anagawa 4-9-1, Inage-Ku, Chiba 263-8555, Japan*

In 1994, carbon ion radiotherapy was begun at the NIRS (National Institute of Radiological Sciences)using HIMAC (Heavy-Ion Medical Accelerator in Chiba), which was the world's first heavy ion accelerator complex dedicated to medical use in a hospital environment. Among several types of ion species, carbon ions were selected for cancer therapy because they presumably had the most optimal properties in terms of biologically effective dose-localization. The purpose of the clinical study of carbon ion radiotherapy was to investigate the efficacy of carbon ions against a variety of tumors as well as to develop effective and safe techniques for delivering the efficient dose to the tumor. As of February 2012, a total of 6,512 patients were treated with carbon ion radiotherapy. The results have shown that carbon ion radiotherapy has the ability to provide a sufficient dose to the tumor without unacceptable morbidity in the surrounding normal tissues. Tumors that appear to respond favorably to carbon ions include locally advanced tumors and those with histologically non-squamous cell type of tumors such as adenocarcinoma, adenoid cystic carcinoma, malignant melanoma, hepatoma, and bone and soft tissue sarcomas. By using biological and physical properties of the high-LET carbon ion beams, the efficacy of treatment regimen with small fractions in shorter treatment course has been confirmed for almost all type of tumors in our carbon ion radiotherapy experience. Based on these experiences, we embarked on the research and development of a new generation beam delivery facilities such as a 3D-scanning method with a pencil beam and a compact rotating gantry. (Figure 1) A clinical research using the pencil beam scanning was in operation since May 2011.

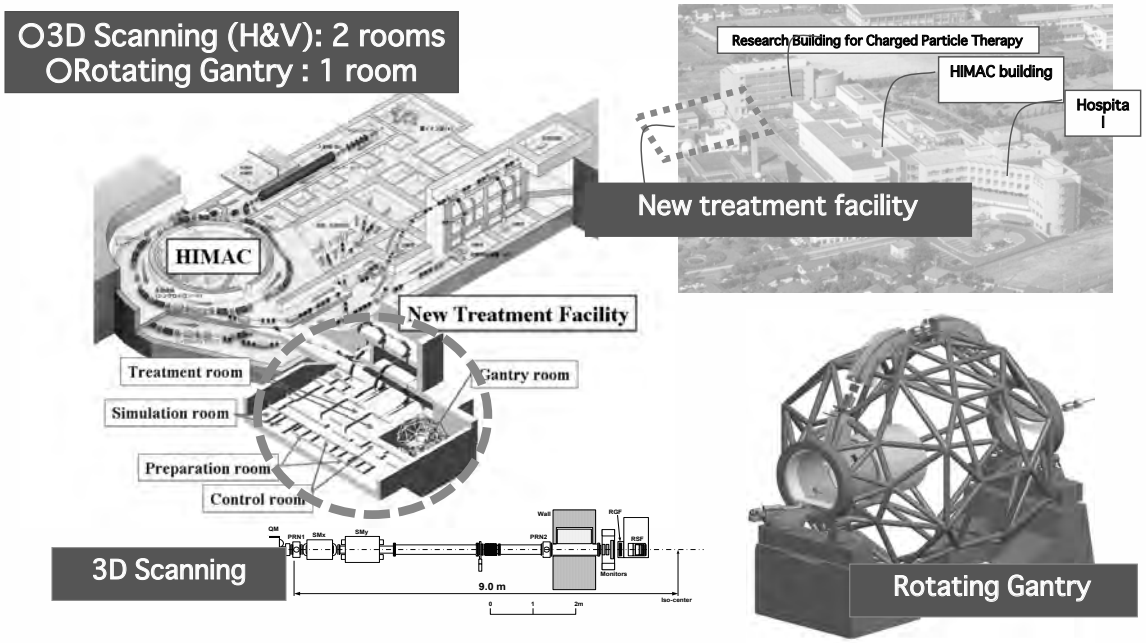


Figure 1. 3D-scanning and rotating gantry at the new facility.

*t_kamada@nirs.go.jp

# DNA Damage Due to Thermomechanical Effects Caused by Heavy Ions Propagating in Tissue

E. Surdutovich[(1)] *, A.V. Yakubovich[(2)], and A.V. Solov'yov[(2)]

[(1)] *Oakland University, Rochester, MI, USA*

[(2)] *Frankfurt Institute for Advanced Studies, Frankfurt am Main, Germany*

Biological effects of ions incident on tissue are a subject of the intense discussion since carbon ion beams are successfully used for therapy and the assessment of radiation damage by heavier ions is important for radiation protection in space [1, 2]. While numerous suggested pathways of DNA damage are due to chemical actions of secondary particles such as secondary electrons, radicals, solvated electrons, etc. on covalent bonds of a DNA molecule, we are considering thermomechanical effects in the medium as a part of the phenomenon-based multiscale approach to the physics of radiation damage with ions [3].

The scenario of these effects starts from the ion's passage through the tissue. The largest fraction of the ion's energy is transferred to secondary electrons produced as a result of ionization of the medium. These electrons lose their energy to the molecules of the medium and it was shown that this energy transfer leads to a thermal spike in a liquid water environment [4]. Due to a transient rise of temperature in a limited volume, the pressure also increases and this leads to a cylindrical shock wave propagating away from the ion's path [5]. A high pressure on the front and its rapid decrease in the wake of the wave result in formidable forces acting on a DNA molecule surrounded with liquid water. Further analysis using Molecular Dynamic simulations allowed us to calculate the energies deposited to covalent bonds of DNA molecules located near the ion's path [6]. This investigation leads to the assessment of DNA damage due to thermomechanical effects. In addition, the shock wave serves as an alternative to diffusion effective mechanism of transport of chemically active species such as free radicals away from the place of their formation in the vicinity of the ion's path. This changes the chemical environment of DNA molecules and may add to the damage effects.

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* surdutov@oakland.edu

# Online monitoring of the ion range during ion therapy by means of prompt secondary radiations

E. Testa[(1)], D. Dauvergne[(1)], M. De Rydt[(1),(2)], G. Dedes[(1), (3)], N. Freud[(3)], J. Krimmer[(1)], J.M. Létang[(3)], M. Pinto[(1)], C. Ray[(1)], M.-H. Richard[(1),(3)], V. Reithinger[(1)], F. Roehlinghoff[(1),(3)]

*[(1)] IPNL,Université de Lyon, Université Claude Bernard Lyon 1,CNRS/IN2P3 France, [(2)] Instituut voor Kern- en Stralingsfysica, KU Leuven, Celestijnenlaan 200D, B-3001 Leuven,Belgium [(3)] Université de Lyon, CREATIS ; CNRS UMR5220 ; Inserm U1044 ; INSA-Lyon ; Université Lyon 1 ; CLB , France.*

The determination in real time of the Bragg peak position during high energy ion irradiation is a challenging objective for the Quality Assurance of ion therapy. Indeed, such a control could allow therapists to stop a treatment in case any deviation from the treatment plan is observed. Since primary particles (carbon ion or proton) are stopped inside the patient, information on the ion range has to be extracted from prompt secondary radiations produced by nuclear reactions.

- Prompt-gamma imaging: during an irradiation, photons in the range 1-10 MeV are emitted almost isotropically within much less than a picosecond after nuclear reactions. Therefore, a dedicated collimated detection setup may provide real-time information on the location of fragmentation. This can be done with a collimated gamma camera or a Compton camera, provided time of flight is used to discriminate between direct photons and background scattered particles like neutrons [1];

- In carbon therapy, secondary protons are – more surprisingly – also helpful for real-time determination of the ion range, even for deep-seated tumors where the residual range for protons to escape the patient in forward direction reaches 10 cm. A tracking system is used to image the interaction vertices that are shown to be correlated to the carbon range [2].

- Simulations are performed with Geant4 to reproduce both the dose profile (including the non-localized dose by secondaries) and the emission of secondaries. A proper tuning of the hadronic models was necessary to reproduce satisfactorily the secondaries yields in order to optimize the detection setup in view of quality assurance instrumentation.

We will present a review of the ongoing activities for prompt radiation imaging, in terms of detector developments and physical issues concerning the correlation between the physical dose and hadronic processes.

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# Water versus DNA: A theoretical description of the ionizing processes induced by proton impact

C. Champion[(1)*], M. E. Galassi[(2)], P. F. Weck[(3)], P. Abufager[(2)], O. A. Fojón[(2)], J. Hanssen[(1)], and R. D. Rivarola[(2)]

[(1)] *Laboratoire de Physique Moléculaire et des Collisions, UMR CNRS 7565, 57078 Metz Cedex 3, France,*
[(2)] *Laboratorio de Colisiones Atómicas (FCEIA, UNR) and Instituto de Física Rosario (CONICET-UNR), Avenida Pellegrini 250, 2000 Rosario, Argentina*
[(3)] *Department of Chemistry, University of Nevada Las Vegas, 4505 Maryland Parkway, Las Vegas, NV 89154, USA*

Ion-induced collisions on DNA bases have been up to now rarely investigated on the experimental as well as the theoretical side. Among the existing theoretical approaches, we find the semi-classical study proposed by Bacchus-Montabonel *et al.* on $C^{q+}$ ($q$ = 2-4) induced collisions with uracil [1] and our recent classical-trajectory Monte Carlo (CTMC) description of single electron loss processes (capture and ionization) induced by $H^+$, $He^{2+}$ and $C^{6+}$ ions on DNA bases. To the best of our knowledge, quantum descriptions of such collisional systems remain scarce and only represented by two series of works, namely, that given by Dal Cappello *et al.* [3] where differential and total ionization cross sections have been reported for protons impinging on cytosine molecules and our recent first Born and CDW-EIS approaches [4-5] in which differential and total cross sections for protons impinging on DNA/RNA bases were reported and compared to experimental data [6-7]. Similarly, we have proposed two theoretical models based on the continuum distorted wave approach for describing the electronic capture induced by heavy charged particles in water and DNA/RNA and provided total cross sections in good agreement with the experiment [8].

In the present work, we describe the different quantum-mechanical approximations also developed to model the ionization and the capture processes for DNA/RNA components, namely, the bases and the sugar-phosphate backbone as well as for water molecule impacted by heavy charged particles. Comparisons with the existing measurements in terms of differential (in angle and in energy) and total cross sections show that the theoretical approaches give a very good description of the studied ionizing processes.

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* champion@univ-metz.fr

# Innovative technology in flower breeding using heavy-ion beams

T. Abe[(1)], and K. Suzuki [(2)*]

*[(1)] RIKEN Nishina Cent., [(2)] Suntory Flowers Ltd.*

Induced mutations are highly effective in enhancing genetic variation in plants and have been successfully applied to the development of improved are new cultivars. Heavy-ion beam irradiation, which has been developed in Japan, is one of the most remarkable mutation technologies. At RIKEN, we have developed a unique technology for mutation induction by using heavy-ion beams from particle accelerators at the RI Beam Factory. This development was achieved through an efficient synergistic link between agricultural science and accelerator physics. The use of ion beams for mutagenesis has a number of advantages: the approach has low exposure levels and high survival rates with high mutation rates, and it creates a wide variety of different mutations. Because heavy-ion beams provide a very high amount of energy, even a single ion is enough to significantly damage a gene. The technique is also very useful in producing mutants that lack just a single gene; multiple propagation technology can be used to convert these mutants into new cultivars. We have put 20 new cultivars in the market in Japan, USA, Canada and EU since 2001. Flower color, long blooming period and a large number of flowers are the important characteristics of floricultural crops. Examples of such breeds include new flower color of 'Surfinia Rose' (petunia), longer blooming period of 'Temari Bright Pink' (vervena), 'Temari Sakura Pink' and 'Temari Momo' (Fig.1). The development period for producing new varieties is only 2 years.

Figure 1. A flower bed in front of the RIKEN cafeteria. These flowers were created by ion-beam irradiation. "Temari Momo", "Temari Bright Pink", "Temari Sakura Pink", and "Safinia Rose Veind" from the center.

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* Kenichi_Suzuki@suntoryflowers.co.jp

# A large scale mutant panel of wheat developed by heavy-ion beam mutagenesis and its application for genetic research

K. Murai[(1)*], Y. Kazama[(2)], and T. Abe[(2, 3)]

*[(1)] Fukui Pref. Univ., [(2)]RIKEN Innovation Cent., [(3)] RIKEN Nishina Cent.*

Mutation analysis is a powerful tool for investigation of gene function. Heavy-ion beam mutagenesis has been recognized to be an effective method of producing mutations because of its high linear energy transfer (LET). High-LET radiation effectively induces DNA double-strand breaks than other mutagenic methods. We have been constructing a large scale mutant panel of diploid einkorn wheat (*Triticum monococcum*) using heavy-ion beam mutagenesis for 12 years. Seeds of the einkorn wheat strain KU104-1, KU104-2 or DV92 were treated with 50-58 Gy of N or C ion beam with LET of 30 keV$\mu m^{-1}$, and then sown in the field. The spikes of $M_1$ plants were bagged and the harvested selfed seeds of each spike were used to produce the $M_2$ lines. Every year, we obtained about 1,000 $M_2$ lines, eventually developing a mutant panel with a sum of 10,000 $M_2$ lines (Fig. 1). We are using this mutant panel for screening mutation of reproductive growth, especially for flowering-time mutants. We have identified several flowering-time mutants of great interest; non-flowering mutants (*mvp*: *maintained vegetative phase*) [1], late-flowering mutants, and early-flowering mutants. The *mvp* mutant is powerful tool for investigation of genetic mechanism of flowering in wheat [2].

Figure 1. Experimental field for screening of $M_2$ lines in Fukui Prefectural University.

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* murai@fpu.ac.jp

# The study of Energy transfers towards radiation-induced defects in polymers submitted to ionizing radiation

Y. Ngono-Ravache[(1) *], A. Ventura[(1)], D. Levavasseur[(1)], E. Balanzat[(1)], T. Chenal[(2)], M. Visseaux[(2)], F. Bonnet[(2)], and P. Zinck[(2)]

*[(1)] CIMAP; BVd Becquerel BP 5133 F-14070 Caen Cedex5, France, [(2)] UCCS-ENSCL; Bât. C7 BP 90108 F-59652 Villeneuve d'Ascq Cedex, France,*

The radiation chemical yields of hydrogen, $G(H_2)$, in polyethylene irradiated under vacuum tend to decrease with increasing dose. This is a known, although partially studied, phenomenon. Theoretical studies [1,2], as well as recent experimental results [3,4] on the evolution of $G(H_2)$ with the irradiation dose, suggest that this yield decrease is linked to the modification of energy transfers in the polymer due to defects created at lower doses and acting as energy sinks.

For a thorough study of this phenomenon, the action of each type of defect, among the major defects created under ionizing radiation, should be specified. But this is made difficult by the variety of defects created and the simultaneity of their creation. To overcome this obstacle, we adopted an innovative strategy based on the insertion, in a regioselective manner, of a given defect type at a given concentration, in a polyethylene backbone. Due to the specifications on the sink concentration and the regioselective introduction sough, these polymers were tailor-made. We focused here on unsaturations of the *trans*-vinylene type which are, with crosslinking, major defects created in the PE backbone.

After optimization, we have managed to synthesize long-chain 1,4-*trans*-polybutadiene whose hydrogenation leads to the regioselective insertion of *trans*-vinylene unsaturations, at molar contents ranged from 0.001% up to 25%, in polyethylene. These PEs were irradiated with $^{20}Ne$ ion beam at very low doses for $G_0(H_2)$ measurements. Evolution of $G_0(H_2)$ as a function of the chemically inserted C=C concentration was compared to the evolution of $G(H_2)$ as a function of radiation-induced C=C concentration in pure PE. Elsewhere, the *trans*-vinylene protection radius and protection factor were determined.

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* ngono@ganil.fr

SH-O-10

# Response of Nuclear Ceramics to Heavy Ion impact

T. Wiss[(1)*], H. Thiele[(1)], A. Janssen[(1)], T. Sonoda[(2)], M. Toulemonde[(3)],

C. Trautmann[(4, 5)], W. J. Weber[(6)], V.V. Rondinella[(1)], R. J. M. Konings[(1)]

*[(1)] European Commission , Joint Research Centre, Institute for Transuranium Elements,P.O. Box 2340, 76125 Karlsruhe, Germany*

*[(2)] CRIEPI, 2-11-1 Iwado-Kita, Komae-Shi, Tokyo 201-8511, Japan*

*[(3)] CIMAP-GANIL, Bd H. Becquerel, F-14070 Caen-cedex 5, France*

*[(4)] Gesellschaft für Schwerionenforschung, Materialforschung, Planckstrasse 1, D-64291 Darmstadt, Germany*

*[(5)] Technische Universität Darmstadt, FB11 Material-und Geowissenschaften, Petersenstraße 23, 64287 Darmstadt, Germany*

*[(6)] Department of Materials Science & Engineering, The University of Tennessee, Knoxville, Tennessee 37996, USA*

Nuclear materials whether used as reactor fuel, inert matrix for transmutation or as a waste form are subjected to radiation damage. The more severe damaging sources are the high energy ( ~ 50 to 100 MeV) fission products generated during fission of uranium or plutonium in fuels or from other actinides in transmutation targets (providing the neutron spectrum is appropriate), and also in waste conditioning matrices or nuclear waste glasses from spontaneous fission of minor actinides.

Swift heavy ion irradiations have been performed in $UO_2$ [1, 2], $MgAl_2O_4$ [3-5], $CeO_2$ [6], $Nd_2Zr_2O_4$ [7] to study their behavior in the high electronic stopping power regime. Some materials have also been exposed to spontaneous fission from $^{244}Cm$ used as self-damaging source [8]. Energy loss thresholds have been determined for the formation of visible ion-tracks in several materials (see for example Fig. 1). A thermal spike model [9] has been applied to calculate the temperature along the ions trajectory. The microstructure evolution of some materials during reactor irradiation e.g. $UO_2$ [2]or $MgAl_2O_4$ [10] has been correlated to the damage from a single fission spike.

Transmission Electron Microscopy results from ion-irradiated samples (single effect study) or from $^{244}Cm$ doped samples will be presented together with some thermal spike calculations as well as the ultimate evolution of nuclear materials after cumulating high fission dose.

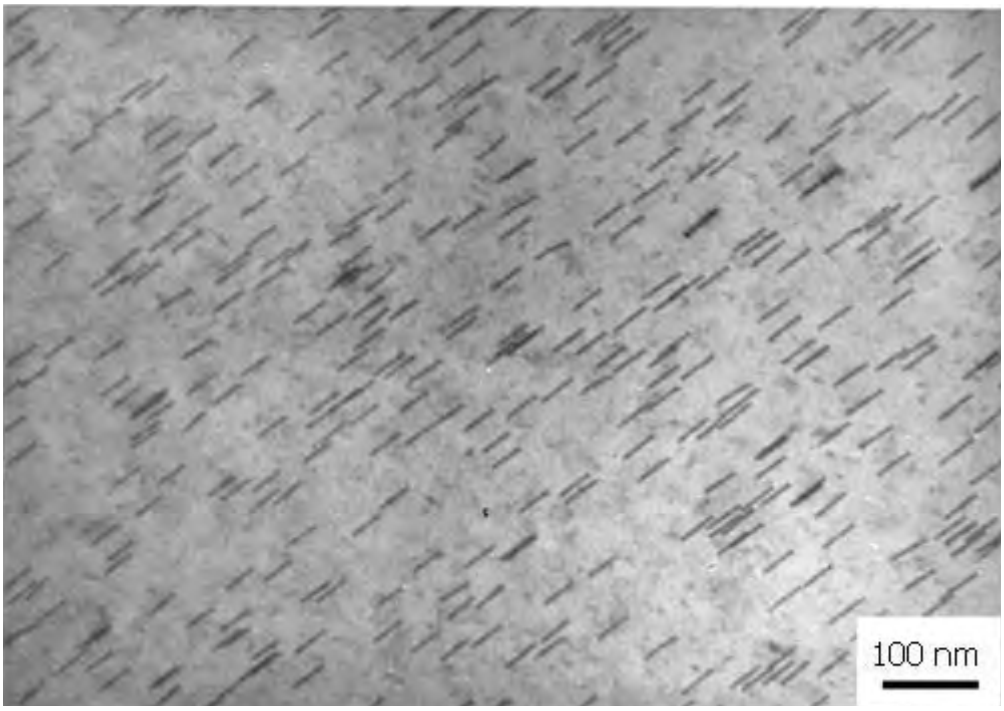


Figure 1: TEM bright field image showing 70 MeV Iodine ion-tracks in a <110> $MgAl_2O_4$ spinel single crystal.

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* thierry.wiss@ec.europa.eu

# Thermoelectric and plasmonic nanowires fabricated by ion-track technology and electrodeposition

I. Alber[(1)], S. Mueller[(1)], R. Neumann[(1)], O. Picht[(1)], C. Trautmann[(1,2)], and M.E. Toimil-Molares[(1)*]

[(1)] *Materials Research Department, GSI Helmholtz Centre for Heavy Ion Research, Darmstadt, Germany*
[(2)] *Department of Material- and Geo-Sciences, Technische Universität Darmstadt, Darmstadt, Germany*

We will present recent results on thermoelectric and plasmonic nanowires produced by electrodeposition in ion-track etched polymer templates.
Large efforts are being currently devoted to synthesize and design materials with higher thermoelectric efficiency, expressed via the figure of merit $ZT = S^2\sigma T/\kappa$, $S$ being the Seebeck coefficient, and $\sigma$ and $\kappa$ the electrical and thermal conductivity, respectively. Nanomaterials are particularly interesting due to their reduced dimensions and controllable morphology. Finite- and quantum-size effects are expected to enable reductions in $\kappa$ and increases in $S^2\sigma$ that are not possible in conventional bulk materials.
We show that $Bi_2Te_3$ and $Bi_{1-x}Sb_x$ nanowires with controlled composition between x = 0.05 and x = 0.40 are electrochemically synthesized in etched ion-track membranes over a wide range of diameters (20–100 nm) interesting for the investigation of the enhancement of thermoelectric efficiency via quantum size effects.[1,2] In the case of $Bi_{1-x}Sb_x$, the nanowire composition is controlled by the relative concentration of Bi and Sb ions in the electrolyte, together with the deposition potential. Two different templates were fabricated to enable the synthesis of thermoelectric nanowires with smooth and rough contour, which enables to influence the phonon scattering and thus the thermal conductivity of the nanowires.
In addition we will discuss the synthesis of AuAg nanowire dimers separated by nanogaps, which is undertaken in two-steps: (i) electrochemical deposition of segmented Au/Ag/Au nanowires from a single electrolyte containing $[Au(CN)_2]^-$ and $[Ag(CN)_2]^-$ ions, and (ii) etching of the silver segment.[3] We will discuss the localized surface plasmon resonances (LSPR) in these metallic nanostructures, which are relevant for applications such as surface-enhanced Raman spectroscopy (SERS), surface-enhanced infrared spectroscopy, and waveguiding below the diffraction limit of light.

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* m.e.toimilmolares@gsi.de

# Non-Graphitizing Carbon Phase Induced by Swift Heavy Ion Irradiation of Graphite

*M. Tomut*[1,2], *M. Krause*[3], *C. Trautmann*[1,3]

[(1)]*GSI, Darmstadt, Germany,* [(2)]*NIMP, Bucharest, Romania,* [(3)]*TU Darmstadt, Germany*

Information on radiation damage in graphite is mainly available from neutron irradiation in nuclear reactors. The present work assesses damage in the nuclear and electronic stopping regimes in graphite by means of Raman spectroscopy. Samples of HOPG (highly oriented pyrolitic graphite) and polycrystalline isotropic graphite (SGL-R 6650 grade) were irradiated at room temperature at the UNILAC accelerator at GSI using GeV heavy ions of ranges of a few tens of micrometers.. The depth evolution of the damage was monitored by performing micro-Raman spectroscopy with successive cleaving for HOPG. For polycrystalline graphite, a Raman depth profiling of damage was done on the cross-section of the sample starting from the surface down to the end of the irradiated layer. The Raman spectra within the irradiated graphite layer show that the graphitic structure evolves toward glassy carbon in the case of electronic stopping regime and towards nanocrystalline carbon in the elastic collisions regime. We have used two parameters deduced from Raman spectra to characterize the structure of the irradiated samples. One is the intensity of a broad peak at about 1460 cm-1, $I_{am}$, that can be fitted between the defect peak, D, and the graphitic peak, G. The other parameter takes into account the tortuosity of the basal planes. The intensity of the broad peak $I_{am}$ in the Raman spectra scales with the electronic energy loss. Higher values at the surface of the sample indicate increased sensitivity of the surface to damage. The evolution of the second graphitic parameter shows a strong bending of the graphitic planes for the electronic stopping regime. The formation of a glassy carbon structure on the surface of the irradiated graphite is confirmed by the presence of the characteristic broad peak in the X-ray diffraction pattern and the strong hardening of the irradiated layer.

# Detecting swift heavy ion modification with graphene

O. Ochedowski[1]*, T. Bollmann[2], S. Akcöltekin[1], B. Ban d´Etat[3], H. Lebius[3], M. Reichling[2] and M. Schleberger[1]

*(1) Universität Duisburg-Essen, Germany, (2) Universität Osnabrück, Germany*
*(2) CIMAP/GANIL, Caen, France.*

It is very well known that swift heavy ion irradiation causes structural modification in many insulating materials [1]. These modifications can be brought to the surface by irradiating the material under glancing incidence conditions which results in a track of hillocks that has been termed “surface track” [2]. However, some insulators such as sodium chloride (NaCl) or silicon carbide (SiC) do not show protrusions. Instead, either no modification at all (NaCl) or trenches (SiC) are observed. In the first case, the modification may be only of transient nature, in the second case the material is obviously ejected from the track. The question arises, whether these effects can be investigated in more detail if the ejected particles or the transient modification could be somehow trapped. Here, we show that graphene, an atomically thin layer of carbon atoms, can indeed be used to trap the emitted material which renders further analysis possible.

For this, we have exfoliated graphene conditions on 6H-SiC samples under ambient conditions. The sample was heated under ultra high vacuum conditions to remove the intercalated water film between graphene and SiC. The irradiation took place at the IRRSUD beamline of the GANIL with a kinetic energy of about 100 MeV at a glancing incidence angle of about 1.5°. We show that the removal of the water film is crucial for this experiment because without it, the irradiation causes graphene to tear [3] and no surface tracks can be observed.  A similar effect is observed on graphene on NaCl.

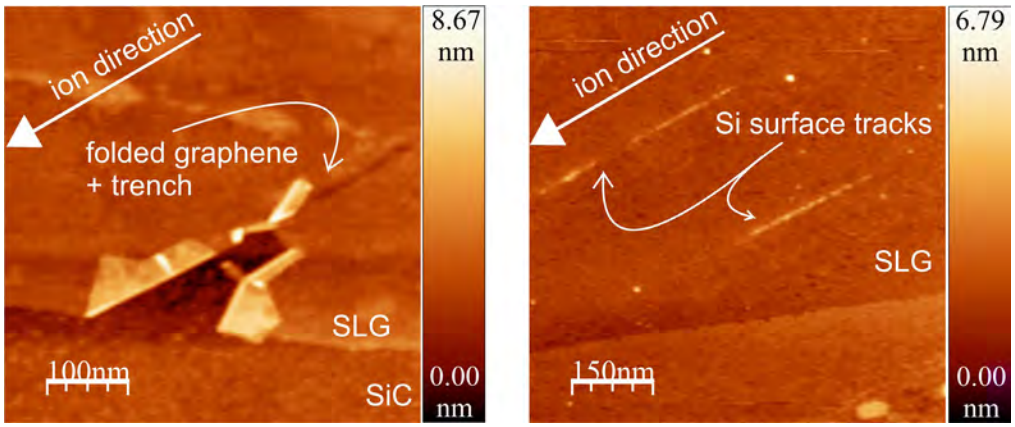


Fig. 1 – left: AFM image (non-contact) of SiC irradiated under a glancing angle of ~ 1.5°. White arrow indicates the direction of the incoming ion beam. A trench in the SiC substrate can be observed as well as the ion induced folded single layer graphene (SLG). Right: SLG flakes that are free of water do not fold and show surface tracks consisting of silicon (Si).

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*oliver.ochedowski@uni-due.de

# Nanoscale Manipulation of the Properties of Solids at High Pressures and Irradiated by Swift Heavy Ions

M. Lang*

*Department of Earth and Environmental Sciences, University of Michigan, Ann Arbor, MI, USA*

Relativistic heavy ions provide a unique opportunity to access a physical regime quite far from the conditions of thermodynamic equilibrium [1]. These projectiles deposit exceptional amounts of kinetic energy (GeV) within an exceedingly short interaction time (less than fs) into nanometer-sized volumes of the material, resulting in extremely high energy densities (up to tens of eV/atom). In this presentation, I will describe a new strategy of combining high-energy ion beams with high-pressure techniques by injecting relativistic ions from one of the world's largest accelerator facilities (GSI – Helmholtz Center for Heavy Ion Research – Darmstadt, Germany) through a mm-thick diamond anvil of a high-pressure cell into a target under pressure [2]. This experimental approach allows for the investigation of the behavior of materials at extreme conditions and opens up unprecedented possibilities for the synthesis of new materials.

We have investigated a number of different oxide compositions and structures at high pressures during irradiation experiments. Radiation-induced energy deposition into highly compressed materials (several tens of GPa) can dramatically modify phase-transformation pathways. The combined use of advanced *in situ* (synchrotron X-ray diffraction and Raman spectroscopy) and *ex situ* (transmission electron microscopy) characterization techniques in experiments up to 65 GPa have revealed: (*i*) the stabilization of a new metastable high-pressure phase of gadolinium-zirconate pyrochlore ($Gd_2Zr_2O_7$) [2] and (*ii*) the transformation into high-pressure and high-temperature phases of ceria ($CeO_2$) and zirconia ($ZrO_2$) at unexpectedly low pressures and/or radiation fluences [3].

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* mklang@umich.edu .

# Annealing of radiation damage in graphite during swift heavy ion irradiation at high temperature

Sandrina Fernandes[1, *], Mikhail Avilov[1], Markus Bender[2], Marine Boulesteix[1], Rodney Ewing[3], Markus Krause[2], Maik Lang[3], Wolfgang Mittig[1,4], Frederique Pellemoine[1], Mike Schein[1], Daniel Severin[2], Marilena Tomut[2,5], Christina Trautmann[2,6]

*[(1)]Facility for Rare Isotope Beams FRIB, Michigan State University, 1 Cyclotron Lab, East Lansing MI 48824-1321, United States*
*[(2)] Dept. of Materials Research, GSI Helmholtzzentrum für Schwerionenforschung, Planckstr. 1, Darmstadt 64291, Germany*
*[(3)] Dept. of Geological Sciences, The University of Michigan, 1100 N. University Ave., Ann Arbor, MI 48109-100, United States*
*[(4)]National Superconducting Cyclotron Lab NSCL, Michigan State University, 1 Cyclotron Lab, East Lansing MI 48824-1321, United States*
*[(5)] Laboratory of Magnetism and Superconductivity, National Institute for Materials Physics NIMP, Bucharest, Romania*
*[(6)] Dept. of Materials Science, Technische Universität Darmstadt, Darmstadt, Germany*

For studying the feasibility of using graphite as a high-power target material for secondary nuclear beam production for the Facility for Rare Isotope Beams FRIB (USA) and the Facility for Antiproton and Ion Research FAIR (Germany) polycrystalline graphite thin foils were irradiated at GSI with a 8.6MeV/u 197Au beam with external electric ohmic heating up to 1600°C. The graphite radiation damage was monitored by in-situ electrical resistance measurements during and after irradiation until its stabilization and cooling to room temperature. The change of electrical resistance provided an assessment of electrical resistivity change and of the accumulation and recovery of irradiation-produced defects in graphite as a function of ion fluence and irradiation temperature. The results show that the saturation of defects occurs faster at low irradiation temperatures due to a less efficient recombination of the irradiation produced defects. The visual inspection of the irradiated graphite foils showed that the higher the irradiation temperature is, the less the observed swelling and dimensional change in the irradiated region. At irradiation temperatures higher than 1200°C no visible foil deformation was observed after irradiation. The annealing of radiation damage at high irradiation temperatures was also confirmed by measuring the crystallographic structure by small-angle X-ray diffraction and other mechanical properties at room temperature. All these measurements showed a nearly complete recovering of the pristine graphite properties at high irradiation temperatures.

[*] *fernandes@frib.msu.edu.*

# Heavy Ion Beam Factory for Material Science based on the KEK Digital Accelerator

K.Takayama[(1,2,3)*], T.Adachi[(1,2)], T.Arai[(1)], D.Arakawa[(1)], H.Asao[(4)], Y.Barata[(1,5)], S.Harada[(5)], K.Horioka[(3)], T.Iwashita[(7)], E.Kadokura[(1)], T.Kawakubo[(1)], T.Kubo[(1)], LIU Xingguang [(1,3)], H.Nakanishi[(1)], Y.Okada[(4)], K.Okamura[(1,2)], M. Okamura[(6)], K.Okazaki[(7)], H.Someya[(1)], K.Takahashi[(3)], Leo Kwee Wah[(2)], M.Wake[(1)], and T.Yoshimoto[(1,3)]

*[(1)]High Energy Accelerator Research Organization (KEK), [(2)]Graduate University for Advanced Studies, [(3)]Tokyo Institute of Technolog, [(4)]NEC Network-Sensor, [(5)]Tokyo City University, [(6)]Broohaven National Laboratory, [(7)]Nippon Advanced Technology Co. Ltd. (NAT)*

The KEK digital accelerator (DA) is an alternative of conventional cyclotron or synchrotron, which are much popular as swift heavy ion beam drivers. The KEK-DA has a capability of delivering a wider variety of ion species with various energy that results from an intrisic nature of this noble accelrator. It is going to serve as a heavy ion beam factory to material science society.

The DA, which is a small-scale induction synchrotron [1] requiring no high-energy injector accelerator and capable of providing ions from hydrogen ion to gold ion, has been constructed at KEK [2]. Since the last year beam commissioning has been carried out. The KEK-DA consists of a 200 kV high voltage terminal, in which a permanent magnet x-band ECRIS is embedded, 15 m long LEBT, electro-static injection kicker, and a 10 Hz rapid cycle synchrotron equipped with the induction acceleration system. An ion pulse chopped in 5 micro-sec by the newly developed Marx generator driven Einzel lens chopper [3] was guided through the LEBT and injected by the electrostatic kicker in one turn. The 3 micro-sec ion pulse was successfully captured with a pair of barrier voltage-pulses of 2 kV and accelerated up to 12-30 MeV with another flat induction-acceleration voltage-pulse through a full acceleration period of 50 msec. Beam commissioning has been started with a He1+ ion beam of 100 μA. Details of fully digital-controlled barrier bucket trapping and induction acceleration are described, although the acceleration/extraction is still at a preliminary stage. The irradiation beam-line has been completed and set-up for the irradiation is under way.

Unique application programs, such as laboratory space science using virtual cosmic rays [4], heavy ion mutation of microorganisms [5], deep ion implantation, modification of materials [6], that may be categorized in systematic studies of a spacial and temporal evolution of the locally and highly excited state of materials [7], are planned.

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takayama@post.kek.jp

# Acoustic Wave Excited in Materials by Swift Heavy Ion Irradiation

Kong-Fang Wei [(1)], Ming-Huan Cui[(1),(2)], Yan-Bin Sheng[(1)], Cun-Feng Yao[(1)],Ya-Bin Zhu[(1)],Li-Long Pang[(1)],Jian-Rong Sun[(1)], Shi-Xu Zhang[(1)],Hai-Long Chang[(1)],Yuan-Fei Li[(1),(2)],Tie-Long Shen[(1),(2)], Hong-Peng Zhang[(1)],Zhi-Guang Wang[(1)*]

[(1)] *Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China,*

[(2)] *Graduate School of Chinese Academy of Sciences, Beijing 100049, China*

High energy density in matter is of fundamental interest for various fields of science, including plasma physics, astrophysics and applications. Intense heavy ion beams could produce high energy density in matter that resulting high pressure to generate acoustic waves and some of them can be detected with sensors [1]. The main subject of the present work is an experimental study of acoustic waves generated by heavy ions in materials.

The experiments were performed at the main electron-cooler storage ring of the Heavy Ion Research Facility in Lanzhou (HIRFL-CSRm). In experiments (Al or water) targets were bombarded using pulsed high energy carbon beams. The bunch width of the beam is 500 ns and the energy of the beam ranged from 650 MeV to 1.4 GeV for solid targets and 1.6 GeV to 2.19 GeV for water. Two acoustic emission sensors were used to detect the acoustic signals and the relative position of the sensors could be altered. For the solid targets, one sensor was fixed on the side of the sample, and the other was fixed on the back of the sample; for water, the sensors were directly soaked in water.

The waveforms of acoustic signals were analyzed and the results show that on the Al sample, for the sensors on both the back and side of sample, the amplitudes increased with the increase of the energy. However, in water, no regular results could be found.

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* E-mail address for correspondence: zhgwang@impcas.ac.cn

# On-line Mössbauer Study of $^{57}$Mn/$^{57}$Fe in Si Materials After Nuclear Projectile Fragmentation and Implantation at RIBF

Y. Yoshida[1], K. Suzuki[3], Y. Kobayashi[2], T. Nagatomo[2], K. Yukihira[1], K. Hayakawa[1], H. Ueno[2], A. Yoshimi[2], D. Nagae[3], K. Asahi[3] and G. Langouche[4]

*(1)Shizuoka Institute of Science and Technology, (2) RIKEN Nishina Center,*
*(3) Tokyo Institute of Technology, (4) Leuven University, Belgium*

A set-up for on-line Mössbauer spectroscopy has been developed and applied for the studies of iron impurities in silicon materials at RIKEN-RIBF [1-4]. We implant GeV-nuclear probes of $^{57}$Mn/$^{57}$Fe into silicon material immediately after a nuclear projectile fragmentation. Electron-hole pairs will be created along the nuclear track, and subsequently, the probe will come to rest on different lattice sites at the end of the track. We study the carrier trapping process on the $^{57}$Fe probes in a p-type Si wafer and in a pn-junction, i.e., a solar cell. $^{57}$Fe probes form deep levels in Si band gap, so that the excess carriers will be effectively trapped. Immediately after each implantation, it is, therefore, possible to observe directly the charge states of $^{57}$Fe probes in terms of the isomer shifts of the Mössbauer spectral components.

$^{57}$Mn nuclei were produced by the nuclear projectile fragmentation of a $^{58}Fe^{21+}$ primary beam (E = 63 MeV/nucleon) with a Be target, and subsequently separated by an on-line isotope separator, RIPS. Mössbauer spectra of $^{57}$Mn/$^{57}$Fe in different Si wafers and Si-solar cells were measured between 300 and 1200 K immediately after the implantation of the radioactive isotope of $^{57}$Mn ($\tau_{1/2}$=1.45m). The implantation was performed through an aluminum foil with a thickness of 200 μm, so that the $^{57}$Mn probes can be stopped at around 100 to 200 μm from the surface of samples. The total fluence of $^{57}$Mn was $2\times10^{12}$ $^{57}$Mn/cm$^2$ typically for one spectrum. Immediately after the GeV-implantation of $^{57}$Mn nuclei $^{57}$Mn/$^{57}$Fe Mössbauer spectra in Si-solar cells are measured with/without light illumination. Comparing the spectrum of p-type multi-crystalline-Si with that of Si solar cell, the broad spectra of the solar cell can be analyzed as a superposition of interstitial and substitutional Fe components with different charge states. The charge states of Fe impurities are created by the directional carrier flow in the p-n junction. The present results provide us a possibility to clarify the carrier trapping process at the Fe impurities in Si-solar cells.

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