Slow ion collisions involving molecules of biological interest

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In recent years the interest in studying collision induced processes with biomolecular species has increased considerably, not only motivated by the aim to better understand the fundamental fragmentation processes at the molecular level, but also due the relevance of these processes in radiation damage schemes and their possible importance for cancer therapy applications. Whereas collision studies with fast ions, x-rays and electrons have started out much earlier, the interest in slow ion collisions is more recent as it was recognised that they may play an important role as primary particles when high-energy ions are slowed down due to the interaction with matter to the so-called Bragg Peak energy, where most of the energy is deposited, or as slow recoil particles produced along the ion track formed by the primary irradiation.

For detailed studies many experiments have been performed in the gas phase with isolated biomolecules applying mass spectrometric techniques. The practical relevance of these studies might be questioned due to the absence of a natural environment. Small biomolecular systems are mostly produced by evaporation techniques, whereas larger ones require the use of Electro Spray or ablation techniques. The latter ones allow also for preparing solvated systems and fore studying the influence of the environment on the fragmentation and dissociation patterns. Thus, experiments with isolated systems and those embedded in water clusters or in a 'chemical' environment, like biomolecular clusters, become feasible and the results can be compared.

In the present contribution we will focus on different collision processes and the corresponding induced molecular damage. We will discuss different cases where in the collision either only energy is transferred to the biomolecular system leading to collision induced dissociation processes (CID), or where electron(s) are taken away by multi-electron capture forming unstable multiply charged biomolecules or where electron(s) are added to an ionic system provoking its dissociation (ECID). The results will be discussed for isolated as well as for 'nano-solvated' species.

TESTS OF FUNDAMENTAL THEORIES WITH HEAVY IONS: CURRENT STATUS AND FUTURE PROSPECTS

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During the last few decades a considerable progress was made both in experimental and theoretical studies of the few-electron Highly Charged Ions (HCI). These studies can be considered as the tests of the Quantum Electrodynamics (QED) in strong fields. The Coulomb field of the nucleus in the HCI exceeds by its magnitude all the electric fields available at the moment in the laboratories. The most accurate comparisons of the experimental and theoretical results were performed for the energy levels including the second-order (two-loop) radiative corrections, for the hyperfine splittings and for the bound-electron g-factors. The measurement of g-factors combined with the precise theoretical calculations allowed for the new and most precise determination of the electron mass.

Up to now the tests of QED are the unique tests of the fundamental theories with HCI where the experimental results are available. However, in principle, the HCI provide the possibilities for testing the very broad field of the modern fundamental theories. First, it concerns the tests of the Standard Model (SM) in the low-energy limit via observing the space Parity Nonconservation (PNC) effects in HCI. Up to now the PNC effects were observed only in the heavy neutral atoms where the theoretical calculations required for the extraction of the SM parameters from the experimental data, are very involved. At the moment the agreement with the high-energy results for SM does exist. Still the PNC experiments with the few-electron HCI where the theory is much more clearer would be of essential importance. The most feasible PNC experiments with HCI require the employment of the polarized HCI beams . Suggestions for the ion beam polarization also are discussed in this talk.

The PNC experiments with the polarized HCI beams would allow a direct observation of the anapole moments of the nuclei which originate from the PNC effects inside the nuclei and represent the new nuclear property. Up to now only the indirect observation of the nuclear anapole moment contribution to the PNC effect was registered in the experiments with neutral atoms.

There are suggestions to observe the effects of the space- and time-parity nonconservation (P,T-nonconservation) in HCI. These effects, discovered more than 50 years ago in the meson physics, should allow also for the existence of the Electric Dipole Moment (EDM) of the particles: electrons, protons, neutrons, nuclei, atoms, molecules etc. In spite of the very intensive experimental search for the EDMs of all these particles , initiated by numerous theoretical suggestions, no firm evidence for the existence of EDM is yet found..The HCI physics provides some prominent possibilities for the observation of the EDMs for the nuclei. These possibilities are discussed in the present talk.

The latest developments in the modern fundamental theories include the search for the time variation of the fundamental constants. The first observation of such time dependence for the fine structure constant which follows from the astrophysical data was reported recently. This observation is not yet confirmed neither by the other astrophysical observations nor by the laboratory experiments. The HCI provide a good opportunities for testing the time dependence of the fine structure constant and the electron to proton mass ratio. These opportunities are also discussed in the talk.

Electron-Ion Collision Experiments at Storage Rings: Fundamental Processes and Astrophysical Implications

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Heavy-ion storage rings equipped with electron coolers are an excellent experimental environment for electron-ion collision studies. Some recent studies of dielectronic recombination (DR) focussed on high-resolution spectroscopy of highly-charged ions. Highlights of this research are the measurement of the hyperfine induced decay rate of the $1s^2\,2s\,2p\;{}^3P_0$ state in berylliumlike Ti^{18+} [1] utilizing DR at the storage ring TSR of the Heidelberg Max-Planck-Institute for Nuclear Physics, the observation of the isotope shift in DR of three-electron Nd^{57+} [2] using different isotopes of this ion at GSI's storage ring ESR and the observation of the hyperfine splitting of Sc^{18+} low-energy DR resonances at the TSR high-resolution electron target [3]. The latter experiment resulted in the determination of the $Sc^{18+}(2s_{1/2} - 2p_{3/2})$ energy splitting with an uncertainty of only 4.6 ppm which is less than 1% of the few-body effects on radiative corrections [4].

Another line of research is the determination of absolute photorecombination rate coefficients for astrophysical and other plasma physical applications [5-8]. Storage ring experiments provide particularly valuable information on DR in low-temperature plasmas such as photoionized plasmas that occur e. g. in active galactic nuclei in the vicinity of super-massive black holes. In such plasmas highly charged ions exist at relatively low temperatures. For many ions, the DR rate coefficients, that determine the charge balance in these plasmas, depend sensitively on the low-energy DR resonance structure at relative electron-ion energies below $\lesssim 3$ eV [9]. Even state-of-the-art theoretical methods are not always capable of calculating the low-energy DR resonance structure with sufficient accuracy to satisfy the astrophysical data needs [10]. For the time being, storage ring experiments are the only reliable source for low-temperature DR data and therefore provide valuable benchmarks for the further development of the theoretical tools.

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Spectroscopy of Highly Charged Ions in Solar and Astrophysical Plasmas

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Matter in the Universe is not distributed uniformly; some is very hot, highly ionized, may not be in thermal equilibrium, or even not thermalized. Spectroscopy of lines originating from highly charged ions is surely of crucial importance to understand the characteristics of these hot plasmas in the Universe. One of the fastest ways to understand the characteristics of astrophysical plasmas is to understand those of solar plasma, because it is believed that most of the phenomena happening somewhere in the Universe really take place on the Sun. In addition, the Sun serves as "a laboratory in space," which shows interesting experiments of plasma physics on a gigantic scale. Spectroscopic observation of EUV emission lines in the transition-region and corona provide unique information on physical conditions in these outer atmospheres of the Sun.

The EUV Imaging Spectrometer (EIS) on board Solar-B [1] is capable of observing, for the first time in Solar EUV observations, spectra and monochromatic images of possibly non-ionization-equilibrium plasmas in the solar transition-region and corona at two-wavelength bands of 170 - 210Å and 250 - 290Å, with typical time-resolutions of 1 - 10 seconds. Dynamic plasma accelerations and heating are found to take place in the solar atmospheres, and they are confined in tiny structures.

Time-dependent collisonal-radiative model for the element of iron is developed to diagnose temperatures and densities of those plasmas in the outer atmospheres of the Sun; no systematic models yet exist for iron ions at the ionization stages of L- and M-shells, which are very important for coronal plasma diagnostics. Adopting the best available theoretical calculations of atomic parameters of these iron ions, as well as generating the experimental data, it is one of the aims of our research that the mechanism of coronal heating is addressed via accurate diagnostics information obtained by the EIS instrument.

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EMISSION AND ABSORPTION IN LASER PRODUCED PLASMAS; PROCESSES AND APPLICATIONS

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Laser produced plasmas have been used for many years as intense sources of extreme ultraviolet (EUV) and soft x-ray radiation. Depending on the choice and composition of target the EUV spectra can be dominated by line, unresolved transition array or continuum emission. Nowadays, volume microchip manufacturing is performed using 193 nm excimer laser radiation with which feature sizes of 45 nm can be attained. However Moore's Law requires a doubling of processor speed every eighteen months and is predicated on a 40% reduction in feature size. To begin manufacturing at feature sizes of 32 nm and below requires the introduction of a new technological step, namely EUV Lithography, which is based on the availability of mirrors with high reflectivity in a 2% bandwidth at 13.5 nm wavelength. Much effort is being expended on the development of suitable sources because of the power requirement of ~500 W of in-band radiation with no ions or debris for high volume manufacturing. The UTA emission from laser produced plasmas using tin provide a potential solution [1].

The results of recent experimental measurements of absolute in-band and out of band intensity, ion distribution and debris will be presented. It has been shown that, because of opacity effects, the conversion efficiency is sensitive to ion density and laser wavelength [2, 3]. Various schemes to improve the conversion efficiency will be discussed. The results of recent plasma modelling calculations will also be presented and compared with experiment.

In addition, laser produced plasmas of some high Z elements emit intense line free continua over extensive energy ranges [4]. Some recent results on inner shell photoabsorption spectra of ions obtained using these continua will be discussed. .

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Strong field electron dynamics in intense laser fields

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So-called 'Reaction Microscopes' enable kinematically complete experiments of atomic and molecular break-up reactions with ultimate completeness. We used these 'bubble chambers' of atomic physics to investigate the many-electron quantum-dynamics under the influence of external time-dependent fields in various situations: The response of atoms and molecules on femtosecond (10^{-15} s) time-scales has been probed by exposing them to intense (up to 10^{16} W/cm^2) ultra-short laser pulses. In pump-probe experiments the rotational and vibrational motion of small molecules was followed in real-time and it is hoped that in near future even the formation of new bonds can be traced as function of time. In first experiments with VUV laser pulses from the free-electron laser FLASH in Hamburg the simultaneous absorption of two or three photons has been studied, a regime that is completely unexplored up to now. In very recent experiments at FLASH it was found that the conformational and electronic structure of molecules can be explored in utmost detail with intense VUV laser pulses. Presently, work is in progress to visualize, for the very first time, the breaking of chemical bonds as a function of time and, thus, to realize the dream of producing a "molecular movie".

RESPONSE OF INSULATOR SURFACES TO A VERY SLOWLY APPROACHING HIGHLY CHARGED ION

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Contrary to the interaction of slow highly charged ions with conducting targets, the physical scenario of their impact on insulators is not very well understood by now. A deeper understanding of this interaction is desirable as it would aid their employment as a gentle tool for surface nanostructuring, where they have been recently shown to induce similar defect as swift (≈ GeV) heavy ions, but at a much lower cost and without damaging the deeper layers of the target.

Upon impact on a solid surface the potential energy stored in slow highly charged ions (HCI) is primarily deposited into the electronic system of the target. With respect to electron emission, the charge mobility on these targets is limited; therefore the electron holes created during the impact of a HCI might interfere with the incoming projectile and therefore alter the interaction process ("trampoline-effect"?). Electron emission experiments were performed on insulating KBr, LiF and CaF_2 single crystal surfaces bombarded by slow highly charged xenon ions. We have recently shown [1] that for somewhat faster ions (≈ keV/amu) impinging on insulators, the "hollow-atom" decay process is by far not complete at the time of impact, as a strong sub-surface contribution to the electron emission yield was found. More recent experiments with very slow (down to 30 eV/amu) Xe HCI show a velocity dependence of the electron yield on the impact velocity that clearly deviates from the case of metallic surfaces.

CaF_2(111) surfaces that have been irradiated by slow highly charged ions have been analyzed by atomic force microscopy (AFM). We have observed hillock-like topographic nanostructures which are stable in air and non erasable by AFM scanning [2]. The number density of surface structures is identical to the applied fluence, thus every individual ion creates one nano-hillock. A sharp and well-defined threshold of potential energy is required for the onset of hillock formation but neither the threshold nor the size of the structures strongly depend on the kinetic energy of the projectiles [2,3]. We show that similar to the swift heavy ion case, the emission of energetic electrons into the solid and the conversion of these electrons into lattice vibrations give rise to a local melting of the impact region. Simulations of the dissipation of potential energy into the target material on the basis of an extended classical over-the-barrier model have been performed to facilitate the interpretation of the experimental findings [4].

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NON-STATISTICAL POPULATION OF 1s2s2p ^{4}P QUARTET STATES BY ELECTRON TRANSFER INTO MULTIPLY CHARGED IONS

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A mechanism for the *selective* population of 1s2s2p ^4P_J states by electron capture in energetic collisions of F^{7+}(1s2s ^{3}S) ions with H_2 and He has recently been elucidated [1]. Detailed single electron capture calculations within the Continuous Distorted Wave (CDW) [2] and Classical Trajectory Monte Carlo (CTMC) [3] frameworks both indicate (1s2s ^{3}S) nl ^{2}L doublet and ^{4}L quartet levels to be approximately evenly populated for n=2-5. Following electron capture into the (1s2s ^{3}S) metastable fraction of the ion beam, the populated (1s2s ^{3}S) nl ^{2}L doublet states are found to Auger decay strongly to the $1s^2$ ground state, and thus only negligible feeding to other lower lying doublets by radiative transitions is possible. The (1s2s ^{3}S) nl ^{4}L quartet states, however, find both Auger or radiative decay to the $1s^2$ ground state blocked by spin conservation. Instead they can only radiatively cascade through lower lying *quartets*, eventually strongly populating the lowest-lying 1s2s2p ^4P_J levels [4]. These theoretical results based on Hartree-Fock calculations using the Cowan code and a time dependence cascade analysis are in agreement with existing experimental zero-degree Auger projectile measurements by Lee *et al.* [5] for collision energies above 0.7 MeV/u and also with the recent results of Tanis *et al.* [6]. However, for collision energies below 0.7 MeV/u the present theory seems to underestimate the populations of both 1s2s2p ^{2}P doublets and 1s2s2p ^4P_J quartets for reasons not yet understood. The present status of our results will be presented together with a discussion of alternative scenarios such as the recently proposed Pauli exchange interaction [6, 7].

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FRAGMENTATION AND DESORPTION IN LOW-ENERGY HIGHLY CHARGED ION COLLISIONS WITH MOLECULES AND SURFACES

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Electron capture, which is one of the most fundamental and characteristic phenomenon in low-energy highly charged ion (HCI) collisions, often induces fragmentation of molecules and desorption from or sputtering of surfaces. The reactions have significant selectivity, since the excitation processes have quasi-resonant features and very large cross sections. Roughly speaking, electron-capture cross section increases linearly with an increase in the charge state of incoming HCIs. [1] Some electronic transitions can only participate in the reaction when nuclear motion does not affect the collision. Therefore, HCI excitation makes it possible to induce state-selective reactions more effectively than excitation with any other charged particles such as electrons, singly charged ions, and cluster ions. Electron-capture collisions with small scattering angles or large impact parameters, in other words, offer the possibility of filtering a specific reaction channel, because nuclear motion does not affect the reaction much. [2, 3]

Coincidence detection of multiple emitted particles is a powerful tool in the spectroscopic studies of reactions. Momentum imaging offers a new method of analysis when combined with translational energy spectroscopy or energy-gain spectroscopy of scattered HCIs. [2-4]

This technique was successful in specifying the reaction pathways of the electronic transitions of molecules and following the dissociation processes in CF_4 and N_2. [2, 3] Recently, we had success in secondary ion mass spectroscopy (SIMS) of the topmost layer of both GaN (0001) and $(000\bar{1})$ surfaces. [5] The SIMS technique allows us to develop an *in situ* quantitative lattice-polarity analysis of compound crystals. We are developing SIMS with atomic-depth resolution, [6] and have already obtained preliminary results of proton desorption with atomic-depth resolution. [7] This HCI-SIMS technique makes it possible to measure depth profiles of hydrogen adsorbed in interstitial sites very sensitively. [8] The technique will be useful for the surface analysis of various hydrogen storage capacitors, as well as those of typical semiconductors and alloys.

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Regular Nanostructures by Swift Heavy Ions

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In the last few years numerous experiments on surface modifications studied by irradiation under grazing angles of insulators such as $SrTiO_2$, TiO_2 and Al_2O_3 have been performed. As shown in [1], irradiation under grazing angles leads under certain conditions to almost periodic hillock chains at the surface.

The focus of our present studies is on the surface modifications of typical ionic materials such as CaF_2, LiF as well as KBr and SrF. Although some of these materials have been studied intensively in the past (see e.g. [2]) , the irradiation under grazing angles provides new insights into these systems.

Scanning probe microscopy (SPM) measurements of CaF_2 and LiF surfaces after ion bombardment with swift heavy ions have clearly shown chains of nanodots with different features. While the main part of these chains is nearly regularly arranged, the initial region exhibits randomly distributed nanodots. Depending on the angle of incidence the overall length of the chains varies between 200 nm and over 1000 nm. Moreover, each of these ion-induced tracks is completely enclosed by a flat region (see Fig. 1) with a height of 1 nm.

Freshly cleaved LiF and polished CaF_2 samples with (111) surface orientation have been irradiated without prior surface treatment at the ion beam facility IRRSUD of the GANIL, France. The irradiation was performed using of Pb and Xe ions with kinetic energies of 130 MeV and 93 MeV, respectively. This energy corresponds to a stopping power of 20 keV/nm and 15 keV/nm respectively as calculated with SRIM [4]. The fluences were typically chosen to yield between 10 and 15 tracks on a surface area of $1\mu m^2$. After irradiation all samples were analyzed by means of SPM.

Although neither the chains of nanodots nor the plateaus are completely understood, one can assume that the spatial electron density plays a key role in the track production as has recently been demonstrated for $SrTiO_3$ [1,4,5].

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PROGRESS OF THE SPECTROSCOPY RESEARCH PLATFORM AT THE SHANGHAI EBIT.

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The Shanghai Electron Beam Ion Trap (EBIT) has been in operation for a short time now and is slowly approaching the initial design parameters, see [1]. At this stage in its development the Shanghai EBIT is dedicated to studies using photon spectrometers and detectors. In the near future an ion-extraction line will be constructed along with a recoil-ion momentum microscopy. However in this report we will focus on spectrometer development, spectroscopic studies and a few other recent developments at the Shanghai EBIT laboratory. Currently the Shanghai EBIT has three spectrometers covering totally the wavelength region of 1 to 10000 Å. Two of these instruments are home made. A flat crystal spectrometer covers the wavelength range of around 1 – 20 Å while a flat field instrument covers the range of around 20 – 400 Å. The 3rd instrument is a commercial McPherson 225 normal incidence spectrometer. All spectrometers employ CCD cameras for photon detection. The Shanghai EBIT is also equipped with high purity Germanium detectors for amongst other things dielectronic recombination studies [2] and time evolution studies of ion distributions[3]. To back up these experimental studies computer codes have been developed for calculation of charge state balances etc. Parallel to the experimental program we have also developed experience at running a number of atomic structure codes (MCHF, MCDF, FAC) for various systems, e.g. the M3 decay of the $3d^9 4s\ ^3D_3$ for Ni-like ions [4].

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HIGH ACCURACY TEST OF QED AT THE HEIDELBERG EBIT

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An experimental cornerstone of atomic theory and quantum electrodynamics (QED) is the precise determination ($\delta\lambda/\lambda = 2 \times 10^{-14}$) of the $1S$-$2S$ transition wavelength in hydrogen by the group of Hänsch [1]. Here, the experimental precision exceeds the theoretical by far due to the large uncertainty in the proton radius. This makes experimental results suitable for predicting other transitions in hydrogen and getting relative theoretical uncertainties smaller than that of the Rydberg constant.
For highly charged ions (HCI) the ($Z\alpha$)-perturbation expansion (α being the fine structure constant and Z the nuclear charge) used for the description of low Z systems is no longer applicable, and HCIs are therefore well suited for testing QED in its non-pertubative limit. Theoretical precision in this regime, such as in all-order non-pertubative calculations, still exceeds experimental precision by a factor 10 in case of U^{91+}.
On the other hand theory of few-electron ions has difficulties including interelectronic correlation into calculations. For He-like ions the unified method (UM) [2], all-order method (AO)[3] and recent calculation by Artemyev *et al.* [4] are different theoretical approaches of taking electron-electron interaction into account. Even the most accurate experiment ($\Delta\lambda/\lambda$ =12 ppm) [5] is by a factor of 5 not precise enough to distinguish among the various calculations.
We report about our recent absolute and relative high-precision wavelength measurements of HCI at the Heidelberg Electron Beam Ion Trap (EBIT) performed in order to overcome this situation. The results were obtained with a novel flat crystal (Si-111) x-ray spectrometer [6] using two light fiducial beams to determine the incoming direction of the x-rays, and thus the Bragg angle. Absolute wavelength measurements were carried out using the Bond method [7]. Experiments on the Lyman-α_1 transition in S^{15+}, measured absolutely for the first time, resulted in a transition energy of $2622.692(27)$ eV with an experimental uncertainty of only ($\delta\lambda/\lambda = 10$ ppm). This points at the possibility of establishing absolute x-ray wavelength standards using Lyman-α_1 transitions in the future. For the $1s2p\ ^1P_1 \rightarrow 1s^2\ ^1S_0$ resonance line in He-like Ar^{16+} an uncertainty of $\delta\lambda/\lambda = 2 \times 10^{-6}$ was achieved [8]. This is the most precise wavelength reported for highly charged ions up to now and allows to test recent predictions on QED two-electron and two-photon radiative corrections for He-like ions.

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RELATIVISTIC AND MANY–BODY EFFECTS IN RADIATIVE RECOMBINATION OF HEAVY IONS

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Studies on the capture of electrons by heavy, highly-charged ions have a long tradition [1]. In the *radiative recombination*, for example, a free (or quasi-free) electron is transferred from the target to the ion under simultaneous emission of a photon. During the last decade this capture process has been found to provide a unique tool for improving our understanding of the electron-electron and electron-photon interactions in the presence of strong electromagnetic fields [2]. Therefore, a large number of experiments have been performed in order to investigate the properties of the *recombination* radiation. Owing to the recent progress in the detector design, these experiments have dealt not only with spectra and angular distributions of emitted x-ray photons but also with their polarization properties [3].

In this contribution, we present the recent theoretical advances in describing the electron capture into highly-charged hydrogen- and lithium-like ions. Emphasis will be placed on the angular and polarization properties of the recombination photons [4] as well as the (subsequent) characteristic Kα radiation [5,6]. It is shown in particular, that the angle- and polarization resolved x-ray spectroscopy provides an efficient way for studying the many-body and relativistic effects on the structure and dynamics of high-Z, relativistic ions. To illustrate these effects, detailed calculations will be presented for the recombination of both, decelerated and ultrarelativistic heavy projectiles, and the results will be compared with available experimental data.

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Momentum distribution of highly charged ions formed by strong laser fields

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Technological advances in laser and in atomic collision physics have opened new pathways for measurements beyond the ion yield production. Foremost, the momentum distribution of ionized atoms produced by linearly polarized lasers with moderate intensity has been intensively analyzed employing COLTRIMS. The measured distributions of the doubly charged ion momentum parallel to the polarization of the laser show for Ne and Ar two symmetrical peaks with a dip at zero momentum. This is known as an indication of the electron-electron interaction in the system, i.e. non-sequential ionization of two electrons [1]. In the strong field regime, on the other hand, ions with higher charges can be produced and the momentum distributions of each individual ion show Gaussian-like distributions centered at zero momentum [2]. This suggests that the interaction between electrons is relatively weak and each electron is ionized sequentially. For ionization of multiply charged ions by intense multi-cycle (~ 200 fs) laser field with a maximum intensity of $\sim$ (50-70) PW/cm^2, ions with different charge states are produced during a single laser shot due to a spatial variation of the laser intensity within the beam focus. The measurements show approximately a simple linear relation between the width of the momentum distributions and the ionization potential of the ions. Such a power law scaling appears to be universal, *i.e.*, largely independent of the target atoms used. The underlying ionization dynamics is analyzed using a quasi-classical tunneling theory for a single active electron model assuming that the interaction between electrons is negligible in such a strong-field limit. Due to the laser envelope, ionization saturates even before the peak intensity of the envelope is reached. This saturation intensity, rather than the peak intensity, determines the width of the final momentum distribution [3].

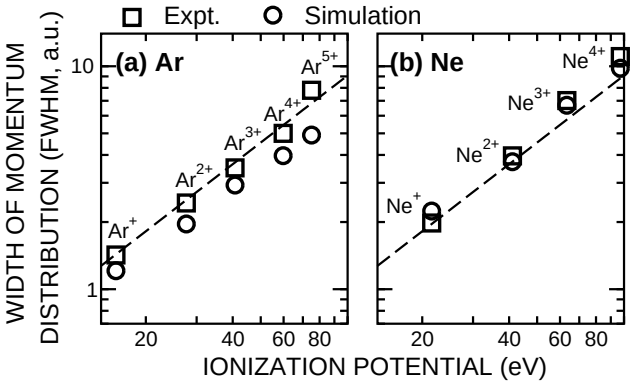

Figure 1: Width (FWHM) of the ion momentum distribution as a function of ionization potential. The atoms (Ar or Ne) are subject to a 200 fs laser pulse with a wave length of 775 nm and a peak intensity of 50 - 70 PW/cm^2. The measured results (squares) are compared with the calculated widths (circles) using the classical trajectory Monte Carlo method including tunneling.

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CONSTRUCTION AND OPERATION OF THE TOKYO EBIT

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1. History

In late 1970s, the collaborative research group was organized in the Institute of Plasma Physics, Nagoya, by Professors Y. Kaneko and T. Iwai to study atomic processes of slow highly charged ions (HCIs) , in which N. Kobayashi, K. Okuno, H. Tawara, S. Ohtani and others participated as young collaborators. This activity was called "NICE project", where a few EBISes were constructed with different characteristics, and by using them, the systematic study of state-resolved electron capture processes of slow HCIs was developed. Thereafter, in early1990s, the inter-University research program, named "Atomic Physics of HCIs" began to be operated as a small national project. In this activity, Kobayashi and Ohtani planned that various HCI-sources would be designed and installed at several research facilities to perform the collaborative study. The Tokyo EBIT was considered to be constructed as one of major HCI-sources in the Institute for Laser Science (ILS), Tokyo.

2. Tokyo EBIT

In 1990, the high energy Super EBIT was constructed at the Lawrence Livermore National Laboratory, California, which has produced a lot of pioneering works by observing high q/Z ions trapped ions in the drift tube. However, the structure seems to be somewhat complicated, that is, the Super EBIT basically consists of an original EBIT and the additionally connected floating electron gun and collector. Therefore, we considered that the Tokyo EBIT should have a more simple shape, from which trapped ions can be easily extracted. Although the conceptual design and the whole rough structure were considered by Ohtani in 1992,the most important parts such as the optimum structures of the electrodes and the magnets were determined by graduate students (N. Nakamura, H. Watanabe et al.) through the elaborated computer simulation of electron beam trajectories under the various shapes of electro-magnetic fields, where the maximum design parameters were Ee=300 keV, Ie=300mA, B=4.5T. F. Currell, M. Sakurai and other young collaborators also participated in the detailed design of the apparatus. Since the contribution by above young colleagues was essentially important for design and construction, the Tokyo EBIT team is named "YEBISU (Young Electron Beam Ion Source Unit)" After the preliminary experiments with Ee=10-80keV, we have made the collaborative study from 1997 for five years on the HCI-spectroscopy with Oxford University where the 20keV EBIT has been operated. During this activity, we have prepared two extraction beam lines from the trap. In 2000s, the resonance interaction of trapped HCIs with the beam electrons has been intensively investigated by observing the charge state variation of extracted HCIs as a function of Ee. In addition, by using the extracted ions, the HCI-surface interaction has been also studied eagerly in relation to the HCI-based nano-science.

3. The near future plan

Since the Tokyo EBIT has been operated continuously for ten years and accumulated several wrong parts, we would like to dismantle, overhaul and improve the apparatus in the near future. It is also expected that the Tokyo EBIT will be provided to the international collaborations.

COMPACT X-RAY FREE ELECTRON LASER IN JAPAN

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Japan started construction of an x-ray free electron laser (XFEL) as one of the five Key Technologies of National Importance. The XFEL is a self-amplified spontaneous emission (SASE) based light source using an 8 GeV electron linear accelerator and a long undulator. Assembly of innovative technologies developed in Japan enabled us to make the facility size to be 1/3-1/4 of the corresponding US and European ones. All the newly developed technologies have been verified by constructing and operating a smaller-size prototype free electron laser operating at 50-60 nm ultraviolet regions with 0.25 GeV electrons. The 8 GeV XFEL will be completed in 2010 and emit up to 20 keV laser pulse with 60 Hz repetition. As an intense and coherent x-ray source, the XFEL will be applied to advanced material researches and life science researches. The present status of the construction project as well as the envisaged scientific cases will be discussed.

Fragmentation of Small biomolecules induced by highly charged ion impact

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The fragmentation is a characteristic behaviour of complex systems such as large molecules and clusters upon excitation. A large number of experimental investigations have been performed on C_{60} and small molecules of DNA bases during the last years using photon excitation (laser and synchrotron source) and electron or ion beam impact excitation. Collision induced fragmentation using highly charged ions from ECR source has been demonstrated to be a powerful method providing complementary information comparing to other experimental methods. The specificity of experiments with an ECR source is related to the large choice of the charge and the atom of beams, the easy scan and high selectivity of the kinetic energy and the possibility to perform multi-coincidence measurements in event by event mode.

In this poster, we present experimental results on the fragmentation of DNA bases induced by impact of highly charged ions, Kr^{17+} (13 kV) and Xe^{23+} (10 kV). Multicharged ion beams delivered by the ARIBE facility allowed us to prepare charged molecules at low internal energy via electron capture process at large impact distances. By coincidence detection of the scattered projectile, the charged fragments and the ejected electron number, we were able to study the fragmentation dynamics and delayed dissociation processes. These results will be compared with the fragmentation patterns of DNA bases induced by singly charged ion beams using a novel experimental method, the so-called CIDEC (collision induced dissociation under energy control) [1-3].

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Formation of Nano Pits on the KBr (001) Surface Induced by Single Impact of Slow Highly Charged Ions

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In recent years investigations of the potential energy dissipation of slow highly charged ions (HCI) on solid surfaces have brought out many new interesting phenomena, e.g. hollow atom formation, enhanced sputter yields, and potential electron emission [1].
The rapid release of the potential energy of a HCI produces a highly excited electronic system in the surface comparable to the excitation by high power ultra-short laser pulses or swift heavy ions [2]. This far from equilibrium state is known to induce various changes in the topography of insulating surfaces on a nanometer scale. Hence, HCIs are considered to be a promising tool for nano structuring and analysis [3]. However, the underlying detailed microscopic mechanisms, especially on atomically flat surfaces, are less investigated so far.
Therefore, we have studied the interaction of slow (300 eV/amu) highly charged Xe ions with the KBr (001) surface. The individual impact of such projectiles induces nanometer size pit-like structures on the surface. Thereby, the pit formation ability and the pit volume are found to depend strongly on the projectiles initial charge state and their kinetic energy. From complementary high fluence irradiations evidence is found that the pit formation is associated with the agglomeration of electronic defects induced by the potential energy dissipation into complex centers (X-centers).

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LOW ENERGY CARBON ION IRRADIATION OF WATER ICES

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Ion processing plays an important role in the chemical and physical modification of ice surfaces in astrophysical environments. Magnetospheric ions surrounding the Gas Giants in the outer Solar System impinge upon and modify the icy satellite surfaces creating new chemical species, incorporating elements not originally present in the local ice composition.

$^{13}C^{+}$ and $^{13}C^{2+}$ ions were produced by an Electron Cyclotron Resonance ion source, accelerated by a low energy accelerator and were incident upon pure water ice samples. Modifications to the ice were measured using an FTIR spectrometer. The most significant modifications observed within the IR spectra were the growth of features corresponding to $^{13}CO_2$ and H_2O_2 and the decrease of features corresponding to H_2O. Additionally, a feature corresponding to 'dangling OH' bonds was seen to decrease, indicating a change in the porosity of the ice being irradiated. It was also interesting to note that no signature for ^{13}CO was observed at any stage during the experiments.

Figure 1 shows the formation of $^{13}CO_2$ as a function of ion fluence at kinetic energies of 2 (left) and 4 keV (right). It is clear that, at both energies, significantly different yields of $^{13}CO_2$ were observed with the two different ion charge states. This difference is believed to be a result of the additional potential energy depostited by the doubly charged ion during an electron capture event.

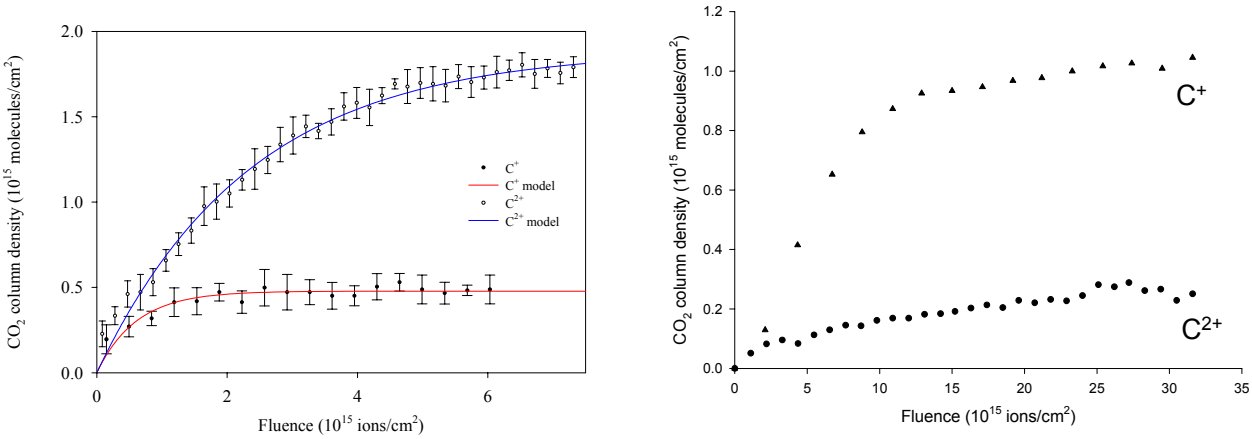

***Figure 1**: CO_2 growth at 2 (left) and 4 (right) keV. In both cases, the yield of CO_2 is significantly different for the two charge states.*

[1] Anita Dawes, Adam Hunniford, Philip D Holtom, Robin J Mukerji, Robert W McCullough and Nigel J Mason *Phys. Chem. Chem. Phys.* **9** 2886-2893 DOI: 10.1039/b618026d (2007)

POTENTIAL SPUTTERING OF IONIC SPECIES FROM RARE GAS SOLIDS BY MULTIPLY CHARGED ION IMPACT

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Interaction of rare gas solids (RGS) with low energy photons and electrons were well studied in these two decades [1]. In these studies, creation of excitons and ions is found to play an important role in the desorption processes. Sputtering of ions from RGS by singly charged ion impact has been investigated by some groups [2, 3], however little report can be found by multiply charged ion impact. Here we report the results for the potential sputtering yields of ions from RGS by multiply charged ion impact.

Figure 1 shows mass spectra of ions sputtered by the impact of 1 keV Ar^+ and Ar^{7+} from solid Ne. Very large cluster ions up to cluster size n ~100 are observed (not shown in the figure). One can find that the intensity and size distribution for small clusters ($n \leqslant 3$) strongly depend on the charge state q of the incident ion, while almost no dependence on q has been observed for the large clusters ($n \geqslant 7$), suggesting that the kinetic sputtering is dominant for the desorption of large cluster ions.

Considering that the potential energy of Ar^+ (15.76 eV) is less than the creation energy of an exciton (17.1 eV) and an ion (21.6 eV) in solid Ne, we assume that only the kinetic sputtering occurs by Ar^+ impact, and estimate the potential sputtering yield Y_{PS}. The results (Fig. 2) show that Y_{PS} is proportional to the potential energy of the projectile ion, which suggests that all of the potential energy is consumed to create excitons and ions in the solid. Detailed discussion will be given at the conference.

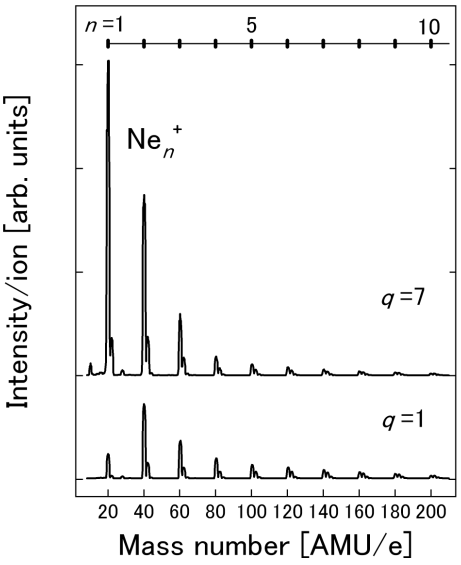

Fig. 1. Mass spectra of sputtered ions from the surface of the solid Ne by 1keV Ar^{q+} (q = 1, 7) impact. Thickness of the solid Ne is about 600ML.

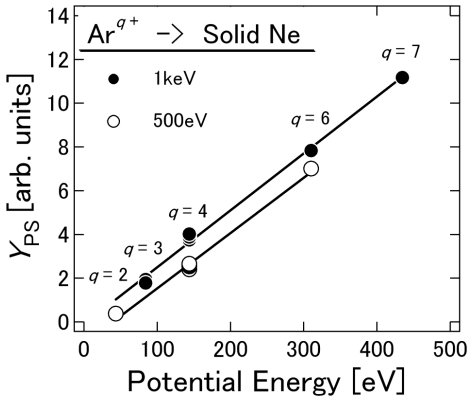

Fig. 2. Potential sputtering yields of ions sputtered by Ar^{q+} (q = 2, 3, 4, 6, 7) from the surface of solid Ne. Full and open circles correspond to 1 keV and 500 eV impact energy of incident ions, respectively.

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Re-trapping and Cooling Highly-Charged Ions

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Presently, a trapping system for cooling highly-charged ions that were extracted from the new Stockholm electron-beam ion trap (S-EBIT) is being set up at AlbaNova (Fig.1). The experiment aims at production of low temperature (emittance) highly-charged ions at very low energy for injection into the precision trap of SMILETRAP II. As a first step the Penning-type cooling trap with deceleration and acceleration system, which facilitates the injection and extraction, was brought into position and successfully tested. Ions can be created either externally, i.e., in the ion sources S-EBIT or SMILIS, or internally by transmitting a 4 kV electron beam through the trap to ionize rest gas. The segmented centre electrode allows for rf-excitation for cleaning the cooling trap from unwanted ions. Currently, a pepper-pot emittance meter is being installed to monitor the emittance of trapped ions in real-time.

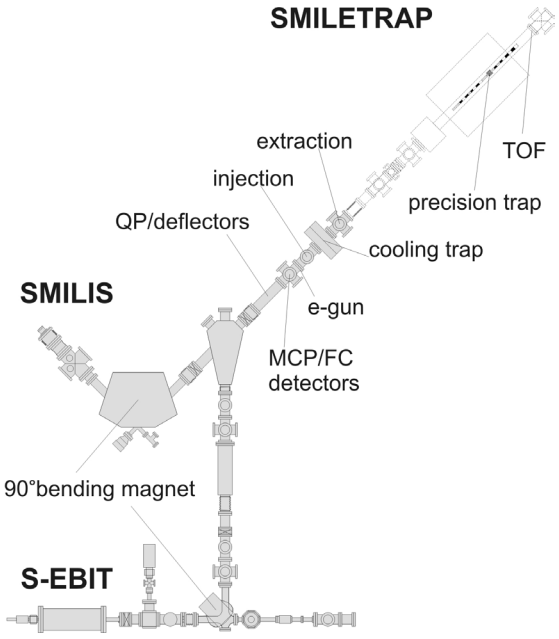

Fig.1 Layout of the HCI facility at AlbaNova.

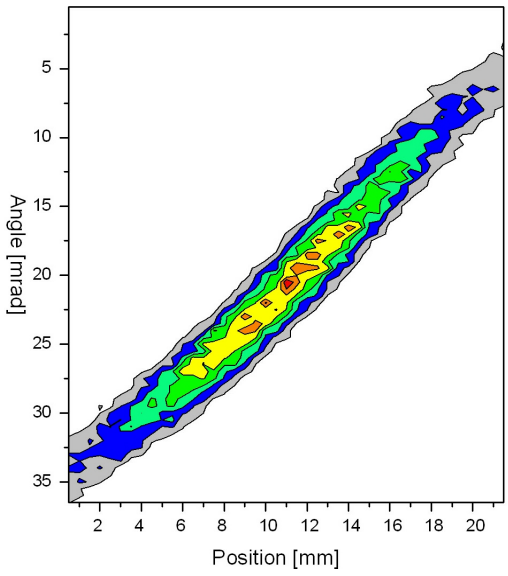

Fig.2 Emittance of Ar^{16+} ions trapped for 100 us.

From S-EBIT and SMILIS two ion species, highly-charged and low charged ions (e.g. S^{14+} or Ar^{16+} and He^{+}) could be injected sequentially and stored simultaneously in the cooling trap. The emittance of the trapped ions was measured (Fig.2), which will be used for optimizing the evaporation of the light ions to cool the highly-charged ions in the cooler trap. For the primary ions, emittances in the order of a few mm·mrad and energy spreads of few qeV were measured. It is expected that these values will be reduced and highly charged ions of 0.1 qeV energy spread can be extracted as pure ion beam.

THE FIRST TEST EXPERIMENT PERFORMED AT THE ELECTRON COOLER OF STORAGE RINGS IN LANZHOU

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The cooler storage ring CSR project was launched in 2000 at the Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou. The project consists of two rings, the main cooler storage ring CSRm and the experimental cooler storage ring CSRe. Both CSR rings are equipped with electron cooling devices [1]. In 2007, the installation was completed and the commissioning of CSRs gained great success, a new highly precise generation of collision experiments will become accessible even for the heaviest ion species. The CSR provides unique and unprecedented conditions for experiments based on the use of highly charged ions and in particular for research in the realm of atomic and nuclear physics. The interaction of the brilliant beams of cooled high-Z ions with low-dense gaseous matter as well as with electrons can be now studied under almost completely background free experimental conditions and with highest luminosity [2,3].

Recombination between electrons and ions is one of the most fundamental atomic collision processes for all kinds of plasmas in the universe. For such investigations, the electron cooler of the storage rings can be used as an electron target for ion-electron interaction studies. In the electron cooler environment, two processes compete: dielectronic recombination (DR) and radiative recombination (RR). The rate coefficient, most important parameters in plasma modelling, can be obtained for various electronic configurations and detailed information on the atomic structure can be deduced in addition. Theoretical investigations of the DR process show that DR might be a powerful tool for the investigation of influence of nuclear effects on the atomic structure and may even be used to obtained model independent information on the nuclear structure, e.g. the nuclear charge radius. This has just been confirmed experimentally [4]. Because the experimental storage ring CSRe can accumulate and store radioactive ions produced by nuclear fragmentation, DR experiments open a novel way for studying the ground-state properties of nuclei far from stability.

A commissioning RR experiment was performed at the electron cooler for Ar^{18+} ions, the results were presently under evaluation. A further RR experiment is being under preparation for krypton ions. A program is planned to be completed before the end of this year for fine detuning the electron beam energy in order to perform high precision DR experiments.

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OBSERVATION OF HIGHER ORDER RESONANT ELECTRON RECOMBINATION PROCESSES BY HIGHLY CHARGED KRYPTON IONS

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By measuring resonant electron capture processes by highly charged ions, where the kinetic energy of the captured electron is transferred to bound electrons, atomic structure calculations can be tested stringently. These data are also relevant for plasma diagnostics applications. Many measurements of dielectronic recombination (DR) have been reported in recent years, see e. g. [1,2]. More detailed benchmarking of theory would results from the measurement of higher order resonant electron capture processes In the so called "trielectronic recombination" (TR) or "quadruelectronic recombination" (QR) resonances, two respectively three bound electrons are excited by the captured electron. Among the few reported related experiments, there exists an upper limit determination of the cross section for the TR-process in krypton [3] and also an observation of TR resonances in outer shells of Be-like chlorine at the Test Storage Ring [4]. We present the first observation of TR resonances including the K shell (*KL-LLL* TR) in highly charged krypton ions. Signatures of QR resonances

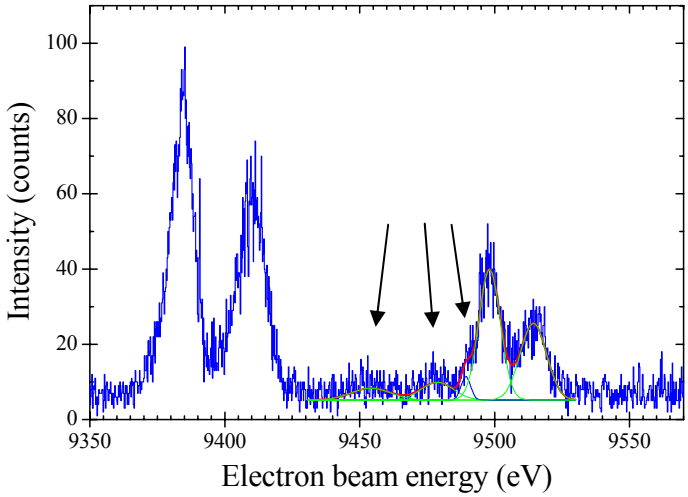

Fig. 1. Recombination of Kr HCI as a function of the electron beam energy. Mean peaks are due to C-like and N-like Kr. The weak features indicated by the two leftmost arrows were reproduced in several measurements and can be identified as the predicted TR of C-like krypton. The rightmost arrow appears at the position predicte for Be-like QR. The energy scale is preliminary.

have also been found for the first time (*KLL-LLL* QR). The excellent resolution achieved, has yielded more accurate data also for the DR resonances. These results have brought a further reduction of the uncertainty in the determination of the absolute resonance energies, and show good agreement with newest predictions of their values.

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X-RAY SIGNATURES OF CHARGE EXCHANGE IN L-SHELL IONS

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The X-ray signature of L-shell charge exchange in Sulfur was studied in the laboratory. Charge states from S^{11+} to S^{14+} were created in the LLNL Electron Beam Ion Trap and were left to interact via charge exchange with neutral SF_6 gas. The measurements were monitored with the EBIT Calorimeter Spectrometer. Comparison of these charge exchange (CX) spectra with those obtained under electron-impact excitation showed marked differences. In the CX spectra, an enhancement was observed in the high-n transitions; $n = 4,5,6 \rightarrow n = 2$, in comparison to the $n = 3 \rightarrow n = 2$ transitions that dominate the direct excitation spectra. An even greater enhancement was recorded in transitions from the levels of electron capture to the ground states; $n_c = 7,8,9 \rightarrow n = 2$. The spectra mainly consist of S^{13+} lines, but lower charge states such as S^{12+}, S^{11+} and S^{10+} are also present. These are the first high-resolution results of L-shell charge exchange. The spectra have been compared to low-resolution data on charge exchange in L-shell Iron, and showed a similar spectral structure. However, the high-resolution spectra from Sulfur exhibit a significant enhancement in transitions from the electron capture-levels n_c=7,8,9, whereas the low-resolution spectra of Iron showed the greatest enhancement in the transitions from n=4,5 levels.

Part of this work was performed under the auspices of the U. S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344 and NASA grants to LLNL, SAO, and GSFC.

STATE-SELECTED DIFFERENTIAL CROSS SECTION MEASUREMENTS FOR THE ONE-ELECTON CAPTURE PROCESSES IN THE F^{4+} - He, Ne, Ar SYSTEMS AT E_{lab} = 45 eV

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Using a crossed-beam apparatus[1], we are measuring systematically the relative state-selected differential cross sections for the charge-transfer processes at very low energy region. A small electron-beam ion-source is employed to produce multiply charged ions. The mass- and energy-selected primary ion-beam was crossed at a right angle with a supersonic nozzle-beam. The scattered ions were energy-analyzed by a one-dimensional position-sensitive detector. The energy spectra of the scattered ions at different angles were recorded by rotating the analyzer.

Typical energy spectrum of the scattered ions observed in F^{4+} - He collisions at E_{lab} = 44 eV is shown in figure 1. The peak (a) corresponds to the elastically scattered ions, and (b) is due to the one-electron capture process. This peak is assigned to the following reactions:

$F^{4+}(2s^2 2p\ ^2P) + He$ ---> $F^{3+}(2s^2 2p3s\ ^1P) + He^+ + 10.0$ eV,
---> $F^{3+}(2s^2 2p3s\ ^3P) + He^+ + 10.9$ eV.

The relative differential cross section, $d\sigma/d\Omega$, for the reaction is shown in figure 2.

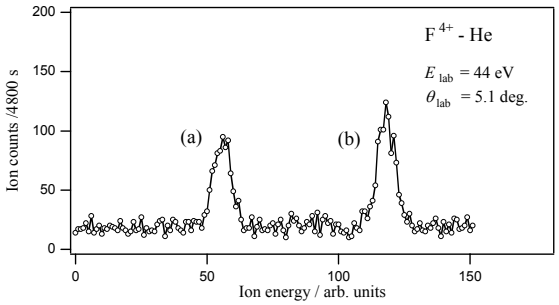

Figure 1: Energy spectrum of the scattered ions.

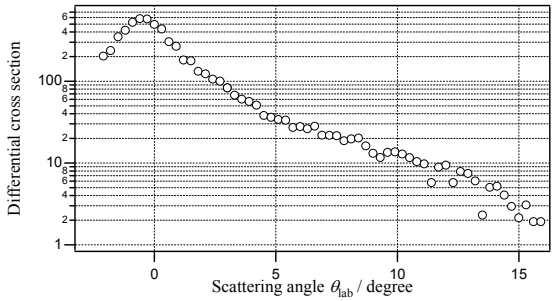

Figure 2: Relative differential cross sections.

The cross section shows a peak around 0° and decreases monotonically with the increase of the scattering angle, while the differential cross sections obtained in F^{4+} - Ne and Ar collisions at the same collision energy show clear angular thresholds. The reaction channels observed in these systems are

$F^{4+}(2s^2 2p\ ^2P) + Ne$ ---> $F^{3+}(2s^2 2p3p\ ^3D) + Ne^+ + 9.6$ eV,
---> $F^{3+}(2s^2 2p3s\ ^1P) + Ne^+ + 13.1$ eV,

and

$F^{4+}(2s^2 2p\ ^2P) + Ar$ ---> $F^{3+}(2s^2 2p3d\ ) + Ar^+ + \sim 9$ eV.

As the differential cross sections are very sensitive to the shape of the interaction potentials, it can be considered that the interaction potentials for the F^{4+} - He system is much different from those for the F^{4+} - Ne and Ar systems.

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STRONG FORWARD-BACKWARD ASYMMETRY OF H_2O IONIC FRAGMENTS BY SLOW HIGHLY CHARGED IONS IMPACT

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Fragmentation of molecules subsequent to the interaction with ions has been studied intensively in the last decade [1]. If the projectile is fast that the interaction time is orders of magnitude smaller than the dissociation time, the collision can be considered as a two-step process [2]. On the other hand, for slow ($v < 1$ a. u.), highly charged projectiles the energy and angular distribution of fragments is determined by the energy and momentum transferred from the projectile, in addition to the energy gained by the repulsive dissociation of the molecule [3].

We present the results of the fragment ion spectroscopy studies of water molecules subsequent to impact of highly charged 1-220 keV Xe^{q+} ions (q=10, 15 and 22). We observed a strong forward-backward asymmetry in the emission of the ionic fragments. For example, the ion yield in the forward direction is almost completely suppressed for the impact of below 10 keV Xe^{22+} ions. These results are compared with our previous studies of water fragmentation induced by $He^{1,2+}$ and $Ne^{(3-9)+}$ ions [1, 4, 5]. For projectiles with low charge state, the final kinetic energy is apparently determined by the related Franck-Condon transition. In order to guide the interpretation of the experimental data, we performed a classical trajectory simulation within the framework of a Coulomb Explosion model wherein a satisfactory agreement is achieved. Finally, the integrated cross sections over all fragmentation channels are compared with results of the semiempirical scaling law deduced from a multi-electron capture model [6].

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COLLISIONS OF IONS WITH INSULATING SURFACES: CHARGING AND DISCHARGING DYNAMICS.

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Guiding and focussing of keV to MeV ions by insulator micro-capillaries offers exciting perspectives for the production of low divergence micro-sized beams and a fine spatial control over the irradiated zone. Such high quality beams may find applications in surface nano- structuring. A spectacular demonstration of the effective use of a tapered capillary is the direct injection of ions inside a living cell [1]. The guiding effect results from the local charging of the capillary inner wall, so this effect depends only on the charging dynamics of the insulating material.
Finding the best material for these applications requires that the charge induced processes are well understood. For that purpose, we have studied the dynamical behaviour of charge deposition and beam deflection on well defined planar geometry where the exact number of charges left on the surface as well as the time dependant beam deflection can be monitored. Depending on surface composition, temperature and structure, discharging time constant vary by orders of magnitude giving rise to different characteristic behaviour.
We have also performed numerical simulations that, besides giving access to the field distribution above the surface plane, point to the importance of the intensity distribution within the incident beam.

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Evolution of X-ray Calorimeter Spectrometers at the Lawrence Livermore Electron Beam Ion Trap

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High-resolution broadband, non-dispersive x-ray spectrometers have been under development for spaceflight since 1984. As an offshoot of the significant NASA investment in this technology, we have developed a series of calorimeter instruments for laboratory use and installed them at the Electron Beam Ion Trap (EBIT) facility at the Lawrence Livermore National Laboratory. Coupled with dispersive instruments at the facility, the calorimeter instruments have made significant contributions to our laboratory astrophysics program. Our laboratory astrophysics program involves benchmarking the spectral synthesis codes and the underlying atomic physics calculations that are used to model high-resolution x-ray spectra obtained with current and future x-ray observatories. The calorimeter instruments at EBIT have significantly enhanced our capabilities to study the physics of highly charge ions including broad band measurements of emission from charge exchange recombination and absolute cross sections for collisional excitation.

The first GSFC calorimeter instrument was installed at the EBIT facility in July of 2000 and has seen two major and a number of minor revisions since then. The performance of the instrument has significantly improved with time from the initial instrument that had a resolving power of ~500 at 6 keV, and essentially no quantum efficiency at energies above 20 keV, to the current instrument that has a resolving power of 1350 and 95% quantum efficiency at 6 keV, and a resolving power of 1800 and 32% quantum efficiency at 60 keV. The advances in resolution are especially apparent at lower energy, where, for example, O VII Kβ at 665 eV evolved from a distinguishable “shoulder” on the high-energy side of O VIII Lyα, to a well resolved line. These improvements have significantly increased the scientific yield of the calorimeter instrument at both high and low energies. We discuss the improvements in the instrument performance and the significant impact on the science yield at EBIT.

Part of this work was performed under the auspices of the U. S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344 and is also supported by NASA grants to NASA/GSFC and LLNL.

QED CALCULATION OF INTERELECTRON INTERACTION CORRECTIONS FOR TRANSITION PROBABILITIES IN TWO-ELECTRON IONS

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We present *ab initio* QED calculation of the transition probabilities for two-electron ions with nuclear charge numbers $Z = 10 - 92$. Employing the line profile approach [1], higher orders of interelectron interaction corrections are taken into account. The radiative corrections were not considered. In particular, calculations are performed for nondegenerate levels $(1s2s)\,^3S_1$, $(1s2p_{3/2})\,^3P_2$ ($M1$ and $M2$ transitions, respectively) and for quasidegenerate levels $(1s2p)\,^1P_1$, $(1s2p)\,^3P_1$ ($E1$ transitions), decaying to the ground state $(1s1s)\,^1S_0$. Both the "velocity" and "length" gauges for describing the emitted photons are considered.

This is the first exact QED calculation of the transition probabilities for the quasidegenerate levels. In the case of quasidegenerate levels the standard QED perturbation theory has a slow convergence for ions with the nuclear charge $Z < 50$. There is a necessity to take into account the higher-order interelectron interaction corrections. Within the framework of the line profile approach we developed a special technique to evaluate the higher-order interelectron interaction corrections to the transition probabilities. One- and two-photon exchange Feynman graphs are considered. The QED perturbation theory applied here is eligible for further improvement of the accuracy of calculation order by order. Calculating the contribution of the interelectron interaction, the QED radiative corrections to the transition probabilities for the quasidegenerate levels become also relevant.

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OBSERVATION AND MODELING OF HOLLOW MULTICHARGED IONS X-RAY SPECTRA RADIATED BY LASER PRODUCED PLASMA.

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Plasma of multicharged ions (Z =10 – 20) with the electron temperature 10 – 100 eV and the electr on density 10^{22} – 10^{23} cm^{-3} is produced usually by interaction of a high-contrast femtosecond laser pulses with solids or clusters. The same plasma parameters are also realized when nanosecond shor t-wavelength laser pulses (KrF or XeCl or 3d, 4th harmonics of Nd glass lasers) interact with the sol ids. Highly charged ions plasma with such parameters is already weakly- coupled and it is very inte resting to investigate radiation property of it. It has been shown in [1-5] that the X-Ray emission sp ectra of such plasma contain some exotic spectral lines caused by radiative transitions in the so call ed "hollow ions", that are the highly charged ions with an empty inner K-shell.

In the present work the role of hollow highly charged ions to the X-Ray emission spectra is investigated for 2 cases: 1) plasma obtained under irradiation of Ar clusters by ultrashort laser pulses and 2) Mg-plasma heated by a short-wavelength long-pulse (nanosecond) laser.

For the first case, cluster-gas targets were irradiated by short laser pulses with various intensities, durations and contrasts. Calculations in support of these measurements have been performed using a detailed atomic kinetics model with the ion distributions found from solution of the time-dependent rate equations. The calculations are in reasonably good agreement with the measurements and the role of hollow highly charged ions in the resulting complicated spectra is analyzed. It is demonstrated that, although the presence of hollow atoms is estimated to add only around 2% to the total line emission, signatures of hollow atom spectra can be identified in the calculations, which are qualitatively supported by the experimental measurements.

In the case of long-lived plasma, produced by XeCl laser irradiation of solid Mg target, clear signatures of transitions from hollow ions are observed in the experimental spectrum. Spectra were identified from large-scale atomic kinetics calculations using the recently developed mixed-UTA (MUTA) model [6]. The relative strength of hollow ion spectral lines is explored and the temperature and density regions in which they are produced are analyzed. Large density and temperature gradients are required to simulate the observations, and additional influence to the hollow atom spectra were simulated with using a hot electron component in the electron distribution function

The kinetic simulations were made with collisional rates calculated in the isolated-atom approximation. The relatively good agreement between theoretical and experimental spectra means that use of such an approximation for the description of the collisional processes is justified for weakly-coupled plasma with the electron-ion coupling factor $\Gamma_{ei} < 1$.

This work was partly performed under the auspices of the US Department of Energy through the Los Alamos National Laboratory and with the support of the RFBR (Projects No. 06-02-16174 and 06-02-72005-MNTIa) and by the RAS Presidium Program of basic researches No. 9.

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THEORETICAL INVESTIGATION ON QUANTUM INTERFERENCE EFFECTS BETWEEN DIELECTRONIC RECOMBINATION AND RADIATIVE RECOMBINATION FOR HIGHLY CHARGED IONS

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The photorecombination (PR) process of a multiply charged ion with a free electron is traditionally described in terms of two distinct recombination mechanisms. The first is nonresonant or direct radiative recombination (RR), which is the inverse of the ordinary photoionization process, and the second corresponds to the two-step, resonant dielectronic recombination (DR) process. These two recombination mechanisms are usually treated as independent processes. It has been recognized, however, that the traditional description of RR and DR, as two independent, noninterfering processes, is not strictly permissible within the framework of a rigorous quantum-mechanical theory [1].

Indeed, more than 40 years ago, Fano [2] predicted the interference between transition amplitudes leading directly into the ionization continuum and those indirectly proceeding via a discrete intermediate resonant state. Several theoretical investigations have been carried out in pursuit of prominent manifestations of the quantum-mechanical interference between RR and DR. Asymmetrical PR cross-section profiles, which are characteristic spectral signatures of a prominent quantum-interference effect, have been observed firstly by Knapp *et al.* [3] for the PR of very highly charged uranium ions, and also have been detected for highly charged mercury ions by González Martínez *et al.* [4] recently.

Using a projection-operator and resolvent-operator approach [1], total cross section of electron-ion photorecombination processes are calculated for highly charged Hg^{75+} ion. This approach provides a unfied quantum-mechanical description of the combined electron-ion PR process, including radiative and dielectronic recombination as coherent, interfering components. The related energy levels, wavefunctions and transition data for B-like Hg ion have been obtained using GRASP92 package [5], component program REOSS99 [6] and AUGER [7], which is based on multiconfiguration Dirac-Fock method. The RR cross section calculated using our recent developed program RERR06 [8]. The present theoretical results show evident asymmetrical profile in the $KL_{12}L_3$ DR resonant region.

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PRECISE EXPERIMENTAL AND THEORETICAL STUDIES ON RESONANT ENERGIES OF THE KLL DIELECTRONIC RECOMBINATION PROCESES FOR HE- UP TO O-LIKE XENON

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Dielectronic recombination (DR) is a resonant process, in which a free electron is captured by an ion, and at the same time a bound electron of the ion is excited, followed by radiative stablization. Studies of DR processes are very important, not only for researches relevant to hot plasmas, but also for atomic structure and collision theory, as DR processes carry information on quantum electrodynamics, relativistic effects, many body interactions and so on.

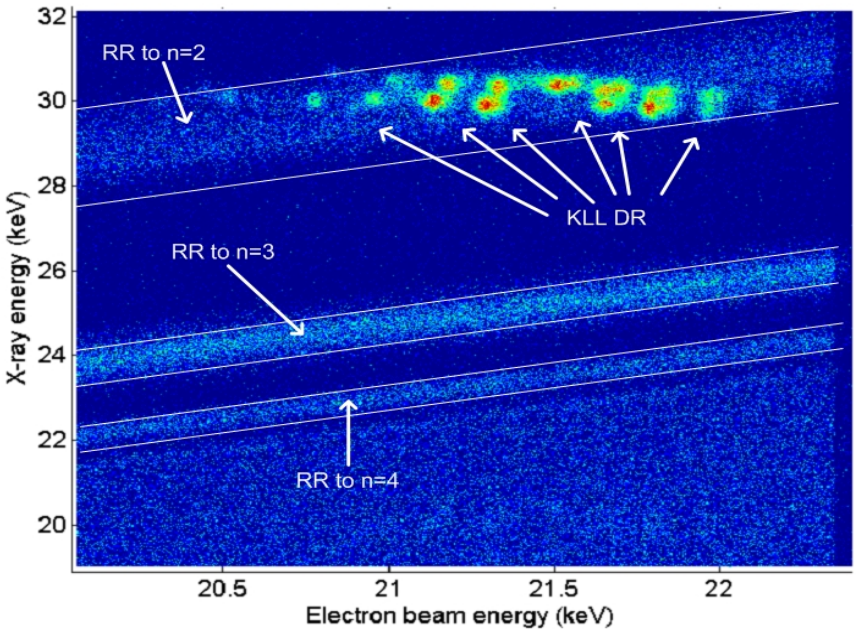

Fig. 1: Scatter plot of X-ray counts over electron beam energy and photon energy, taken at electron beam current of 62 mA. The KLL DR resonant events and radiative recombination to n=2, 3, 4 are shown.

Precise experimental studies of the resonant energies of KLL DR processes of He-, Li-, Be-, B-, C-, N-, and O-like xenon ions were performed at the Shanghai Electron Beam Ion Trap, by monitoring the electron beam energies over the KLL DR resonance region, see Fig. 1, employing home developed high precision, high stability high voltage dividers. Effects of the space charge of the electron beam and the ion neutralization, retardation by the capacitor of experimental setup, contact potential as well as fringing field effects were taken into account. Fifteen experimental resonant energies were obtained at an average uncertainty level of 0.03%.

At the same time, calculations using relativistic configuration interaction (RCI) theory, relativistic many-body perturbation (RMBPT) theory were also performed for the above-mentioned KLL DR resonant energies. Comparisons of experimental and theoretical results were made. 13 out of the 15 predictions by RMBPT are in good agreement with our experimental results, while only 5 out of the 15 predictions by RCI calculation agree with our experiments. Three literature available DR resonant energies by multi-configuration Dirac-Fock calculation agree very well with our experiments.

Hyperfine dependent lifetimes in Neon like ions

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As part of our ongoing investigations of hyperfine dependent lifetimes of metastable levels [1-4] we report on a theoretical investigation of hyperfine quenching in Neon-like ions. The studies were done along the iso-electronic sequence ranging from neutral Ne (Z=10) to Au^{69+} (Z=79). $2p^5 3s\ ^3P_2$ is the first excited level in Ne-like systems and it can only decay to the ground state, $2p^6\ ^1S_0$, through a magnetic quadrupole (M2) transition. For the ions up to $Z \approx 51$, were there is an energy level crossing between the $2p^5 3s\ ^3P_0$ and the $2p^5 3d\ ^3P_0$ level [5], the former is the third excited level and it can only decay to $2p^5 3s\ ^3P_1$ through a magnetic dipole (M1) transition and to $2p^5 3s\ ^3P_0$ through an electric quadrupole (E2) transition. Also it has no possibility to decay to the ground state except through a two-photon decay. In the presence of a nuclear spin though, the hyperfine interaction introduce a small mixing of $2p^5 3s\ ^3P_1$ and $^1P_1$ into $2p^5 3s\ ^3P_2$ and $^3P_0$ respectively, opening up hyperfine induced electric dipole (hpf-E1) transition channels to the ground state for both of the metastable levels.

Extensive multiconfiguration Dirac-Hartree-Fock calculations were performed to calculate the transition probabilities for the various transitions from the two metastable levels as well as the transition matrix elements of the $2p^6\ ^1S_0 - 2p^5 3s^3P_1$ and the $2p^6\ ^1S_0 - 2p^5 3s^1P_1$ E1 transitions. Also the off-diagonal hyperfine interaction constants between the metastable levels and the $2p^5 3s\ ^3P_1$ and the $^1P_1$ levels were calculated. First order perturbation calculation were used to calculate the $2p^6\ ^1S_0 - 2p^5 3s\ ^3P_2$ and the $2p^6\ ^1S_0 - 2p^5 3s\ ^3P_0$ hpf-E1 transition rates and the hyperfine dependent lifetimes of the hyperfine levels of the metastable levels. It is shown that the $2p^5 3s\ ^3P_2$ level is sensitive to hyperfine interaction all along the iso-electronic sequence ranging from Z=10 to Z=79. It is also shown that the $2p^5 3s\ ^3P_0$ level is very sensitive to the hyperfine interaction in the beginning of the iso-electronic sequence where the hpf-E1 transition, in many cases, are orders of magnitude larger than the M1 and E2 transition rates. This sensitivity decreases with Z since both the M1 and the E2 transition channels have a higher order of Z dependence compared to the hpf-E1 transition channel. However even for highly charged ions, if the nuclear magnetic dipole moment is large, the hyperfine quenching can have a substantial influence on the lifetime of the $^3P_0$ level. An example is the hyperfine quenching of $^3P_0$ in In^{39+} which is lowering the lifetime by about 25%.

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SELECTIVE FORMATION OF MULTIPLY EXCITED STATES BY RESONANT COHERENT EXCITATION

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Multiply excited states attract great interests for the test of theories involving the strong electron correlations. Especially, investigation of triply excited Li or Li-like ions has progressed significantly in recent years. However, the triply excited Li-like system of heavier ions can be formed only by the atomic collisional processes such as electron capture to the outer shell since the synchrotron radiation is not applicable. Here we demonstrate a novel method for the formation of selective multiply-excited states of heavy ions.

We utilized three-dimensional resonant coherent excitation (3D-RCE) [1] of energetic heavy ions in a crystal, in which the traveling ions are resonantly excited through the interaction with the periodic crystal fields. With the use of ions at relativistic velocity, the available transition energy of RCE rises up to the x-ray region which enables us to excite the inner shell electron of highly-charged heavy ions. Since the oscillating field consists of numerous frequency components, the double resonance of RCE can be realized by adopting two of them simultaneously to the resonance.

We excited the two $1s$-electrons of He-like Ar^{16+} and Li-like Ar^{15+} into the $n = 2$ orbitals by successive 3D-RCEs of different harmonics. The energy diagrams are shown in Fig. 1(a) and (b). The double resonance conditions are satisfied by independently adjusting two rotating angles of the thin Si crystal keeping the ion velocity constant, which is a significant advantage over the previous 2D-RCE method [2]. The experiment was performed at the heavy ion medical accelerator in Chiba (HIMAC) with the $\sim$ 400MeV/u ions. We observed the charge state distribution of the ions emerging from the crystal together with the emitted x-ray yields and their polarization. In both cases, our result provided clear evidence that the multiply excited states are formed in the crystal.

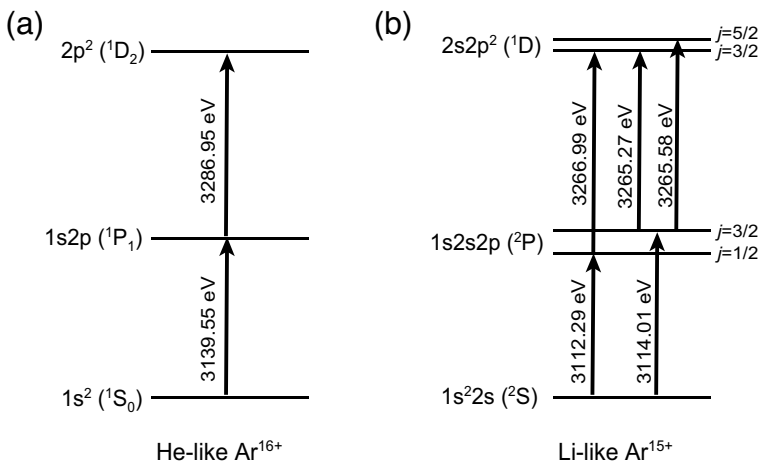

Figure 1: The level diagram of double electron excitation of (a) He-like Ar^{16+} and (b) Li-like Ar^{15+}.

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Density matrix description of resonant coherent excitation of swift highly charged ions in oriented crystals

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Experimental and theoretical studies on resonant coherent excitation (RCE) of channeled ions in crystals started about 40 years ago from pioneering works of V.Okorokov [1]. Unambiguous observation of *the Okorokov effect* by S.Datz group at Oak-Ridge and wide variety of RCE measurements performed recently by Tokyo collaboration have made main contribution to the present-day image of the RCE process [2].

Density matrix description [3,4] considers RCE ion as an open quantum system involved into coherent and incoherent interactions with the crystal medium. Systematic numerical calculations based on the generalized Master equation for density matrix show it as a good instrument for unified approach to charge state distribution of the RCE ions and the yield and angular distirbution of their characteristic X-ray radiation. Other RCE observables including metastable ion production in a usual RCE process [5] and Auger electron production in a doubly resonant process (dRCE) [6] were considered in the same theoretical approach and suggested for experimental observation. Following our long-time interest in the problem of alignment of excited ions in the RCE process we extend previous density matrix calculations [3,7,8] on this point by theoretical predictions for Stokes parameters of linear and circular polarization of the X-ray radiation from relativistic resonant coherently excited channeled ions.

Our current theoretical studies in the field [9] being closely correlated with main trends in the latest RCE experiments by the Tokyo group [10-13] concern also such novel aspects of the RCE process as its trajectory resolved characteristics and resonant coherent excitation of highly charged ions in non-channeling conditions. Corresponding calculation results and our seeing of general perspectives of further development of the density matrix approach in the RCE studies will be presented at the conference.

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ATOMIC PROCESSES OF DAMDE ON BIO-MOLECULES IRRADIATED BY XFEL

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The analysis of three-dimensional structure of single bio-molecules has lately attracted considerable attention for the application of x-ray free electron lasers (XFEL) [1, 2]. This analysis comes from diffraction patterns, which are produced from the irradiation of XFEL onto the bio-molecules. However, the x-ray flux required for this analysis is so large that the bio-molecules are damaged, that is, the atoms in the bio-molecules are more often ionized. This damage appears as noise for the analysis of the three dimensional structure. Therefore, it is indispensable to estimate the damage.

We treat C, N, O atoms which are the main elements of bio-molecules and some atomic processes such as photo-ionization, Compton scattering, Auger, electron impact ionization, and radiative transitions. The atomic data of Auger and radiarive transitions are shown in Ref. [3]. By the application of the atomic data of these processes to rate equations, we have calculated the change of electronic states or charge numbers as a function of times for various parameters such as x-ray flux, x-ray pulses, and wavelength of XFEL the size of bio-molecules .

Figures 1 (a) and (b) show the change of charge number of C as a function of time for the wavelength of 0.1 nm and 0.1 nm of XFEL, respectively. The x-ray flux, pulse of XFEL and the size of bio-molecules are 10^{22}/pulse/mm^2, 10 fs, and 20 nm, respectively. We have found that shorter wavelength produce smaller damage. In our presentation, we will show the results for various parameters and suitable parameters for the experiment of diffraction pattern of bio-molecules.

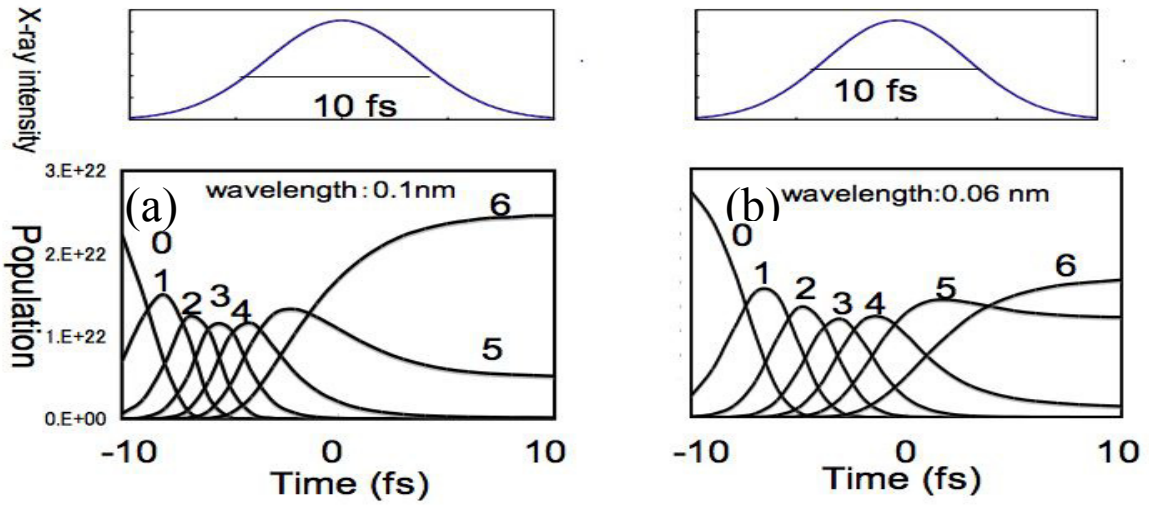

Figure 1 Population of charge number of C vs. time for the wavelengths of (a) 0.1 nm and (b) 0.06 nm, respectively.Upper figure shows the x-ray intensity. The pulse, flux of x-rays, and the size of bio-molecules are 10 fs, 10^{22} /pulse/mm^2, and 20 nm, respectively. The figures shown here are charge numbers.

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EFFECTS OF DEBYE PLASMAS ON THE RESONANCE STATES OF HIGHLY STRIPPED TWO-ELECTRON IONS USING THE STABILIZATION METHOD*

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The effects of screened Coulomb potentials in atomic or molecular processes have become an active and relevant search topic in the recent years ([1-4], references therein). Recently, we have initiated resonance state calculations ([3-4], references therein) on different atomic and molecular systems under the influence of screened Coulomb potentials. In the present work, we investigate the resonance states of two-electron atoms, Mg^{10+} and Si^{12+} interacting with screened Coulomb potentials of the form : exp(-μr)/r, where μ is called the Debye screening parameter (μ=1/ λ_d, λ_d the Debye length). We employ highly correlated exponential basis functions, supported by a widely used quasirandom process [1,3] of the form $\varphi_i = r_1^L P_L(\cos\vartheta)\exp[-(\alpha_i r_1 + \beta_i r_2 + \gamma_i r_{12})] \pm$ exchange, for S-, P- states calculations, whereas for D-wave resonance calculations, we employ CI-type basis functions with certain approximations [4]. The stabilization method [5], a simple and powerful technique that needs only L^2 type basis functions, is used to extract resonance energies (E_r) and widths (Γ). We have obtained several doubly-excited *S*-, *P*- and *D*- waves resonance states of the proposed systems for each μ, below the *n*=2 thresholds of the respective two-body subsystems. In Fig.1, we present lowest S- and D-wave resonance widths of Mg^{10+} and Si^{12+} as functions of μ.

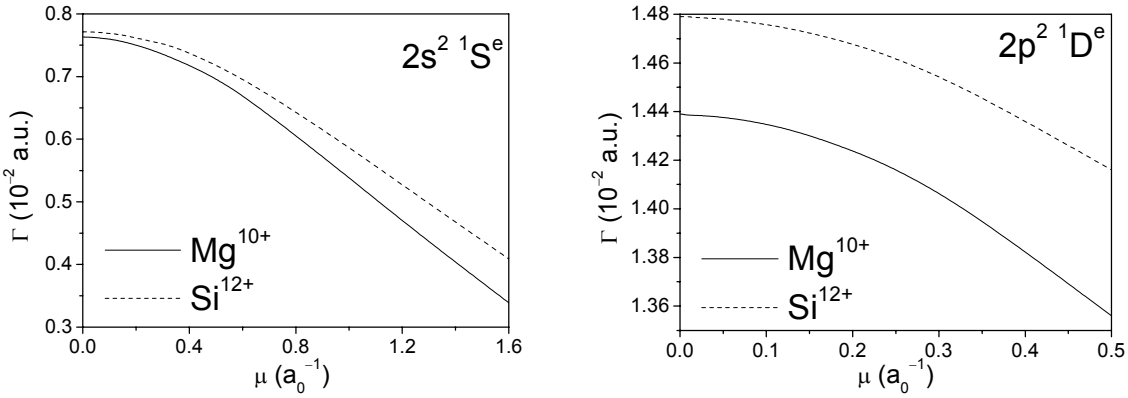

Fig. 1. Lowest S- and D- waves resonance widths of Mg^{10+} and Si^{12+} in terms of μ.

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OPTIMAL PULSE DURATION IN LASER-CLUSTER INTERACTIONS

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The study of the interaction of intense short laser pulses with clusters has received much attention during the last decade [1]. The ions and quasi-free electrons in the cluster form a "nano-plasma" of solid density, where the electrons are efficiently heated by the combined fields of the laser and the surrounding particles [2,3]. Electron-impact ionization produces highly charged ions as well as inner-shell vacancies, which are at the origin of X-ray radiation. As a fraction of the electrons leaves the cluster, a net positive charge is left behind and the cluster begins to expand before disintegrating completely in a Coulomb explosion.

The size of the system and the multitude of mechanisms at play provide a challenge for the theoretical description of the interaction. Due to the large number of atoms ($N > 10000$) in a cluster, a full ab-initio simulation still seems impractical. We therefore opt for an open effective mean-field approach, in which many-particle effects are included via stochastic processes. The roles played by different effects such as cluster polarization, surface disintegration, and microscopic atomic dynamics (elastic electron-ion scattering, electron-impact ionization etc.) can thus be studied.

The measurement of the 3.1 keV characteristic K-shell X-ray radiation emitted from argon clusters [4] provides an excellent experimental tool to gain insight into the dynamics of the interaction on the short time-scale of the irradiating laser pulse ($\tau > 60\,\mathrm{fs}$). High resolution X-ray spectroscopy gives access to the charge state distribution of the highly charged cluster ions Ar^{q+} with $q > 12$.We achieve good quantitative agreement between the simulated and experimental absolute X-ray yields, and the ionic charge-state distributions are also well reproduced at high laser intensities.

By varying the pulse duration, additional information on the interplay between ionic and electronic dynamics during the cluster explosion process can be gained. One highlight is the existence of an optimum pulse duration that maximizes the X-ray yield at constant laser pulse energy. We give an interpretation of this effect different from the previously invoked nanoplasma resonance [5].

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EVIDENCE OF GIANT DIPOLE PLASMON RESONANCE IN ELECTRON SPECTRUM OF C_{60} AND BEAM INDUCED POLARIZATION

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Mechanisms of electron emission in collisions with clusters involving C_{60}-fullerene are governed by more complicated processes than in ion-atom collisions. The fullerenes are "small" particles with diameter ~10Å over which a large electron density similar to that of a solid is sampled. Fullerenes are known to exhibit collective excitation: *giant dipole plasmon resonance* (GDPR). Effect of GDPR on x-ray emission from fullerene and multiple ionization under heavy ion impact has been reported earlier [1-5]. However, a direct evidence of this process in fast ion-collisions was awaited. The plasmon excitation de-excite through emission of electrons of particular energy, which is characteristic of the plasmon frequency. This presents a unique possibility of observing the GDPR peak in the low energy electron spectrum of C_{60}. The electron energy due to the decay of GDPR-excited state is expected at ~10 eV (i.e. 20 eV- I_P, I_P=ionization potential). We measured the low energy (1-300 eV) e^- - DDCS spectrum from C_{60} in collisions with 4 MeV/u bare F ions at various angles. We observe a broad hump like structure near the expected GDPR peak position in all the electron spectra. The C_{60} DDCS spectra are different from the DDCS spectra of other atomic gaseous targets like Ne [Fig.1]. The low energy spectrum was collected with lot of precaution and spectrometer performance was verified by measuring expected spectral shapes from atomic targets.

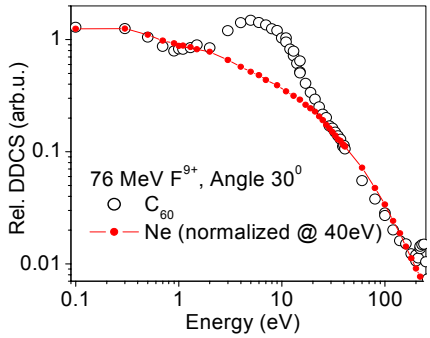

Fig.1 : Typical electron DDCS spectrum from C_{60} (o) and Ne.

We have also measured the angular distributions of the electrons. The electron emission (at ~10 eV i.e. at GDPR peak) is maximum in forward and backward direction w.r.t. to the projectile beam and a dip is observed at 90^0**.** This distribution is just opposite to the expected behaviour for atomic targets. The angular distribution suggests that the dipole oscillations are induced preferably along the projectile beam direction. This is for the first time that such an angular distribution of e-emission in plasmon decay in C_{60} or any type of clusters has been measured.

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STABILITY OF HIGHLY CHARGED FULLERENE CATIONS AND ANIONS

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The most abundant fullerenes, C_{60} and C_{70}, and all the pure-carbon fullerenes larger than C_{70} synthesized so far follow, with no exception, the isolated-pentagon rule (IPR)[1-2]. Fullerenes containing adjacent pentagons (APs) are less stable due to the additional strain. Surprisingly, recent experiments have shown that a few endohedral fullerenes[3], for which IPR structures are possible, hence expected to be the most stable ones, are stabilized in non-IPR cages. These cages are either positively or negatively charged, depending on the character of electron acceptor or electron donor of the encapsulated species. It has been argued that this unexpected stability of charged non-IPR fullerenes is associated with electronic properties of the carbon cage, such as unusually large HOMO-LUMO gaps or bond resonance energies[4,5]. These properties might be related to a reduction of strain induced by the encapsulated species, but the ultimate reasons remain unclear.

By performing density functional theory calculations[6-8] on a large number of C_{60} and C_{70} derivatives with both IPR and non-IPR cages, we show that, apart from strain, the physical property that governs the relative stability of highly charged fullerenes is the charge distribution in the cage[9]. This charge distribution is controlled by the number and location of two different structural motifs, one electrofilic (the pentalene motif, i.e., the pairs of APs,) and the other one electrophobic (the pyrene motifs). APs are the preferential sites to host additional electrons, either by adding explicitly those electrons or by making the AP bonds to react (or both). On the contrary, when one electron is removed from the fullerene cage, the resulting positive charge locates in the pyrene motifs, not in the pyrene bonds but in the more aromatic bonds surrounding the latter. Thus by playing with the number and position of pyrene bonds it is possible to generate non-IPR fullerenes with strongly non uniform positive charge distributions. We show that, when AP and pyrene motifs are uniformly distributed in the cage and well separated from each other, stabilization of non-IPR endohedral and exohedral fullerenes, as well as pure-carbon fullerene anions and cations, is more the rule than the exception. This suggests that non-IPR charged fullerenes might be even more common than IPR ones, which can be relevant to interpret recent experiments in which highly charged fullerenes are produced in collisions with ions or in storage rings.

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