Fragmentation of Atoms in Intense Femto and Attosecond Fields

R. Moshammer[1], H. Kollmus[1,2], B. Feuerstein[1], W. Schmitt[1,2], M. Schulz[4], R. Mann[2], S. Hagmann[3], A. Dorn[1], P. Fainstein[5], B. Bapat[1], H. Rottke[6], C. Trump[6], W. Sandner[6], R.E. Olson[4], J. Ullrich[1].

[1]Uni Freiburg, [2]GSI Darmstadt, [3]KSU Manhattan, [4]UMR Missouri, [5]Barriloche Argentina, [6]MBI Berlin.

The most intense electromagnetic fields of up to 10^{21} W/cm^2 which are extended over more than atomic dimensions can either be generated by fast heavy ions or by ultra short high intensity lasers. There is increasing demand to come to a better understanding of the interaction of these femto- to attosecond pulses with matter in general and with single atoms in particular. Interest is fuelled by unsolved fundamental questions on the dynamics of many-particle quantum systems on the one hand as well as by long range perspectives for potential applications ranging from inertial fusion driven by heavy ion or laser beams, laser driven particle accelerators to new techniques in medical treatment.

The main channels of energy conversion from the field to matter or to single atoms are single and multiple ionization reactions. Two principle complications arise in the description of the interaction: First, the field strength is typically on the order of or even exceeds the nuclear Coulomb field in atoms (10^{11} V/cm) so that any perturbative treatment of the interaction is prohibited. Second, many-particle effects are known to play a decisive role in the energy transfer: More than fifty percent of the total energy transfer in ion-atom collisions is due to simultaneous multiple ionization. The electron-electron correlation has been demonstrated to enhance multiple ionization rates of atoms in intense laser fields by up to forty (!) orders of magnitude. The mechanisms leading to these drastic effects widely remained in the dark since even the simplest atomic reactions involving only two active electrons cannot be fully described theoretically. Just recently after decades of theoretical efforts a rigorously new approach led to a correct description for the most simple time dependent many-particle quantum system, namely single ionization of hydrogen by low energy electron impact [1].

Urgently required in this situation are kinematically complete data sets guiding theoretical efforts to isolate the dominant dynamic mechanisms (matrix elements), to explore the role of the correlated electronic initial state as well as to elucidate the importance of the dynamical correlation of electrons during and after the pulse. Using the "Reaction Microscope" [2] such data have been collected for the first time for single and multiple ionization of Neon in collisions with 3.6 MeV/u Au^{53+} at GSI and PW/cm^2, 30 fs laser-pulse impact at the MBI in Berlin. Seemingly very different, both situations turn out to be indeed very similar in that the passing ion generates an attosecond (10^{-18} s) electromagnetic pulse of similar power density as the femtosecond (10^{-15} s) laser pulse. Three major questions were addressed:

First: How does the low energy electron continuum in single ionization of simple atoms depend on the electronic structure of the target? How and to what extend does the momentum distribution of the ionized electron reflect the atomic initial state momentum distribution?

Second: What is the dynamics in multiple ionization of atoms induced by attosecond ion pulses in the regime of large perturbation (i.e. for large projectile charge to velocity ratio q/v > 1)? What is the structure of the many electron continuum and what are the signatures of electron-electron correlation?

Third: What are the mechanisms leading to ionization in oscillating electromagnetic laser fields lasting for a few femtoseconds? In particular the question about possible double (multiple) ionization mechanisms was furiously debated since more than ten years.

Snapshots of Bound State Momentum Distributions

Still under discussion is the old question whether the correlated ground state wave function of many-electron atoms or molecules is directly accessible in any practicable experiment [3]. Whereas the mapping of effective one-electron initial state momentum distributions (Compton profiles) by impulsive binary collisions with fast electrons has become a well established technique, such measurements are a hopeless task for correlated many-electron states. The simultaneous detection of several low-energy electrons emitted after interaction of fast ion generated extremely strong (10^{19} W/cm^2), sub-attosecond ($\Delta t < 10^{-18}$ s) electromagnetic pulses with atoms or molecules has been proposed as an alternative approach [4]. However, up to now it has not even been conclusively demonstrated for single ionization that the low-energy electron continuum depends on the initial state wave function in a characteristic way. More than 15 years ago it was predicted that the shape of the low-energy electron continuum mirrors the nodal structure of the initial state and the alignment of the target with respect to the beam axis [5]. These calculations have never been verified experimentally because of large uncertainties and restricted electron momentum resolution. The recent development of novel and extremely efficient electron spectrometers combined with recoil-ion momentum spectroscopy made such experiments feasible.

In kinematically complete experiments the doubly differential cross section (DDCS) for low energy electron production has been measured for single and multiple ionization of He, Ne and Ar with 3.6 MeV/u Au^{53+} [6]. This are the first experimental data on electron emission for defined degrees of ionization and with sufficient resolution for soft electrons. Unexpected and target specific structures were observed (see fig. 1). These structures turned out to be signatures of the initial state momentum distribution. For very fast encounters and in the limit of minimal momentum transfer the transition matrix element for ionization by charged particles becomes identical to that of photoionization which is known to sensitively depend on the Compton profile. Moreover, a comparison with advanced CDW (Continuum Distorted Wave) calculations [7] proved that the observed structures in the electron spectra are attributable to the nodal structure of the initial bound state momentum distribution. Some other observed features like e.g. the dominant forward emission and the sharp peak at zero electron velocity (the so called “target cusp”) are independent of the target. The first is a remnant of the projectile-electron interaction dragging electrons after the projectile (post collision interaction PCI) and the target cusp arises because of pure phase space arguments. Thus, low energy electrons emitted in fast ion-atom collisions are sensitive to both, the many particle collision dynamics and the multi electron initial state. In a simple physical picture ionization by fast ions can be viewed as weighted “projection” of the bound state to the low-lying continuum essentially conserving the initial state electron momentum. These results underpin the recent statement that double (multiple) ionization with fast charged particles delivers a unique tool to study the short time correlation of many electron bound states on a time scale which is short compared to the electron revolution time in atoms, molecules and cluster.

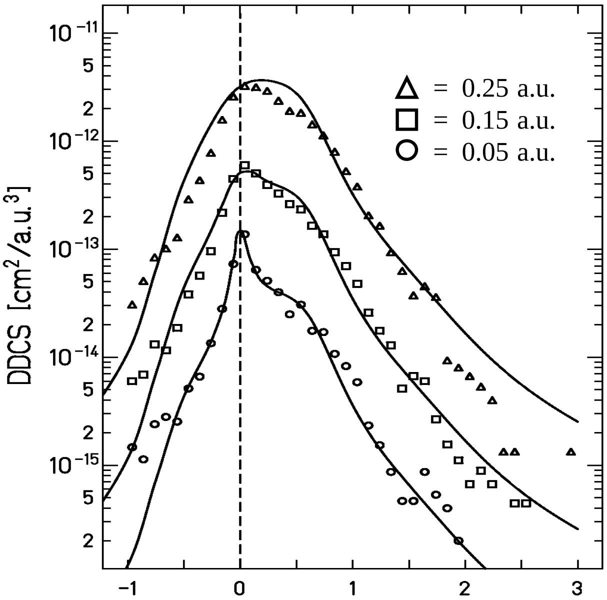

Fig. 1: Doubly differential cross sections DDCS = $d^2\sigma/(dv_{\|}dv_{\perp}\cdot 2\pi p_{\perp})$ as function of the longitudinal electron velocity for certain transverse velocity cuts in singly ionizing 3.6 MeV/u Au^{53+} on Ar collisions. DDCS at different $v_{\perp}$ are multiplied by factors of ten, respectively. Lines: theoretical CDW results.

Triple Ionization: Structure of the 3 Electron Continuum

In the non- perturbative regime (q/v > 1) classical trajectory Monte-Carlo calculations (CTMC) are the only theoretical approach to predict differential cross sections for double and multiple ionization [8]. Whereas certain global quantities like the total kinetic energy of all ejected electrons for a given degree of ionization are in accord with CTMC results [9], the relative energy sharing among the electrons or their mutual emission angles are not reproducible. The reason for this discrepancy might be the inadequate inclusion of e-e correlation which turned out to be important for a proper description of multiple ionization [4]. This correlation can be separated from the interaction with the projectile like e.g. PCI by inspection of the relative momenta and energies of emitted electrons. The result of such an analysis is shown in fig. 2 where the energy sharing between the three electrons ejected in triple ionization of Ne with 3.6 MeV/u Au^{53+} is presented in a modified “ Dalitz-Plot” [9]. There the electron energy sharing is plotted in an equilateral triangle where the distance from each individual side is proportional to the relative energy of the corresponding electron $\varepsilon_i = E_i/\Sigma E_j$ as indicated in the figure. Numbering of electrons is achieved exploiting the information about the electron emission angles. Electron 1 is the one with the smallest angle relative to the projectile direction, electron 3 the one with the largest angle. Obviously, the electron energies are not independent of each other: the many electron continuum explored for the first time experimentally is found to be strongly correlated. There is an increased probability that electron 1 and 3 have large energies compared to electron 2. Performing CTMC calculations with the e-e interaction not included beyond an effective potential in the initial state these structures can not be reproduced. Such calculations have been demonstrated to reliably describe many aspects of multiple ionization reactions. Some

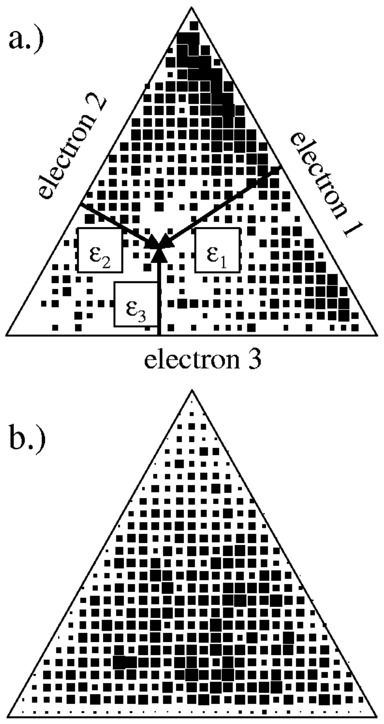

Fig. 2: Dalitz-plots representing the energy sharing of three electrons emitted in triply ionizing 3.6 MeV/u Au^{53+} on Ne collisions: a.) experimental data, b.) CTMC-calculation.

features however, are only predictable when the e-e interaction is included to a certain level of completeness.

With increasing degree of ionization and therefore increasing complexity the question arises whether a description based on any collective model like e.g. a thermodynamic ansatz might be more adequate to describe fragmentation of atoms into many particles. To establish the border from only a few to really many electron processes will be a subject of future experiments.

Multiple Ionization in Intense Laser Fields

Similar to charged particle induced ionization the very intense light pulse delivered by short pulse lasers can be interpreted as a strong time-dependent electric field. But, in contrast to fast ions, the time scale of the acting field is much longer (femtoseconds) than the characteristic time scale of bound electrons (attoseconds). Nevertheless, many experimental results suggested a strongly correlated behavior of electrons resulting in unexpected large yields of doubly and multiply ionized atoms (for a review see [11]). This enhancement, termed non-sequential ionization, which can amount to several orders of magnitude, appears at medium intensities and was subject of controversial discussions.

Three different dynamical mechanisms have been proposed for non-sequential double ionization. First, the ejection of one electron leads to a sudden change of the screened ionic potential seen by another electron which can cause an instantaneous “shake–off” to the continuum. Second, a “ rescattering” process was proposed within a semi-classical model where the ejected electron is driven back by the external laser field to its parent ion after about half an optical cycle ionizing the second electron in a (e,2e)-like collision. Third, instantaneous collective multi-electron tunneling has been considered as a possible contribution in the regime of non-sequential ionization. The complete lack of differential data sets for double (or multiple) ionization in intense laser fields made a definitive decision about the dominant ionization mechanism impossible.

In a recent experiment performed at the Max-Born Institut in Berlin the first differential data on double and triple ionization of neon in ultra-short (30 fs FWHM) laser pulses at intensities of 10^{15} W/cm^2 have been collected. Cold Target Recoil-Ion Momentum Spectroscopy (COLTRIMS) [12] has been used to measure the momentum vectors of ejected Ne ions. Since the momentum transferred by the light pulse is negligible small the ion momentum reflects the sum momentum of emitted electrons. Thus, the ion momentum distributions are a sensitive measure of the many-electron dynamics. Moreover, each proposed multiple ionization mechanism leads to distinct momentum patterns of the ions and a comparison with the observed distributions gives, for the first time, decisive information about the importance of different mechanisms. If electrons were released in an independent manner one by one the shape of the final ion momentum distribution should be similar to the shape observed for single ionization with a maximum at zero momentum. Moreover, any mechanism based on an instantaneous release of two (or more) electrons should result in an ion momentum distribution peaking at zero momentum.

Based on the experimental momentum distributions for multiply charged Ne ions (fig. 3) we were able to definitely rule out several mechanisms proposed to explain non-sequential ionization in intense light pulses. Our data are in accord only with the kinematics of the rescattering mechanism where the tunnel ionized first electron is pushed back by the external laser field and upon inelastic scattering a second or even more electrons are removed from Ne$^+$ by electron impact ionization. This is a unique example on how electron correlation determines the response of a many electron systems on a time dependent external force. The release of the second electron in a double ionization event depend on the help of the first electron.

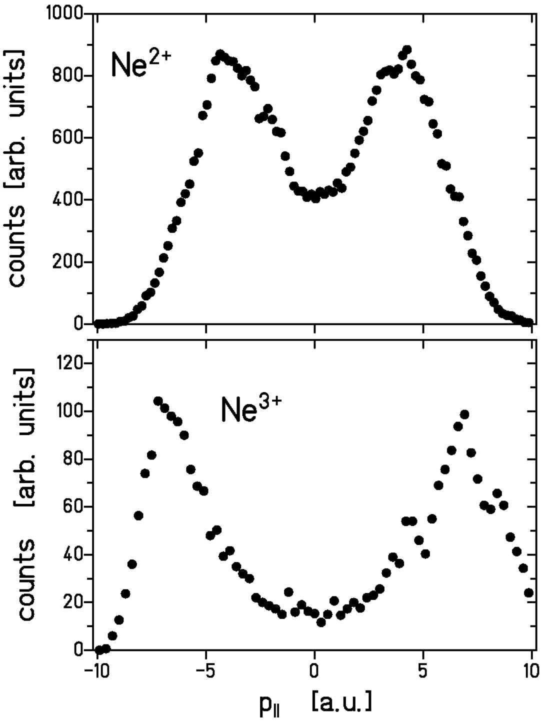

Fig. 3: Momentum distributions of Ne^{2+} and Ne^{3+} ions generated in an intense laser field (wave length 800 nm, pulse length 30 fs, intensity 10^{15} W/cm^2). Plotted are the momenta parallel ($p_{||}$) to the laser light polarization axis.

In future, kinematically complete experiments similar to those performed for ion, electron and single photon impact are planed using our reaction microscope to gain further information about the emission characteristics of electrons ionized by intense laser fields.

References

[1] T.N. Rescigno et al. Science 286, 2474 (1999).
[2] J. Ullrich et al., J. Phys. B 30, 2917 (1997).
[3] W. Heisenberg, Z. Phys. 43, 172 (1927).
[4] R. Moshammer et. al., Phys. Rev. Lett. 77, 1242 (1996).
[5] J. Burgdörfer, Phys. Rev. Lett. 51, 374 (1983).
[6] R. Moshammer et al., Phys. Rev. Lett. 83, 4721 (1999).
[7] P.D. Fainstein et al., Phys. Rev A 53, 3243 (1996).
[8] C.J. Wood et al., Phys. Rev. A 56, 3764 (1997).
[9] M. Schulz et al., J. Phys. B 32, L557 (1999).
[10] M. Schulz et al., Phys. Rev. A 61, 22703 (2000).
[11] L.F. DiMauro et al.,
Adv. At. Mol. Opt. Phys. 35, 79 (1995).
[12] R. Moshammer et al., Phys. Rev. Lett. 84, 447 (2000).