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Many-particle dynamics in atomic and molecular physics investigated with the COLTRIMS-technique: New inside into e-e- correlation

H. Schmidt-Böcking[a], R. Dörner[a], O. Jagutzki[a], T. Jahnke[a], V. Mergel[a], L. Ph. H. Schmidt[a], Th. Weber[a], A. Czasch[a], Ch. Wimmer[a], M. Hattass[a], A. Knapp[a], M. Schöffler[a], C. L. Cocke[b], M. Prior[c], A. Kheifets[d], E. Weigold[d] and F. Afaneh[e]

[a]Institut für Kernphysik, Universität Frankfurt, August-Euler-Str. 6, 60486 Frankfurt, Germany

[b] Kansas State University, Manhattan, KS 66506, USA

[c] Lawrence Berkeley National Lab., Berkeley CA 94720

[d] Research School of Physical Sciences and Engineering, Australian National University Canberra ACT 0200, Australia

[e] Physics Department, Hashemite University, Zarqa 13115, Jordan

Correlated many-particle dynamics in Coulomb systems, which is one of the unsolved fundamental problems in AMO-physics, can now be experimentally studied with so far unprecedented completeness and precision of the momenta of all particles involved in the reaction. The recent development of the COLTRIMS technique (COLd Target Recoil Ion Momentum Spectroscopy) provides a coincident multi-fragment imaging technique for eV and sub eV particle detection (1,2). In its completeness, e.g. measuring the fully differential cross section of a reaction by detecting the particles in 4π for all energies up to a couple of 100 eV so far, it is as powerful as the bubble chamber in high energy physics. Based on state-of-the-art cooling techniques (super sonic jets, MOT etc.) and nuclear physics imaging methods, fragmentation processes of atoms, molecules, clusters, as well as the dynamics of electron emission from solid state surfaces which are induced by single photon or multi photon laser absorption, electron or ion impact can be explored completely in momentum space and, for ions, with micro eV resolution. In recent benchmark experiments quasi snapshots (duration as short as an attosec) of the correlated dynamics between electrons and nuclei are taken for atomic and molecular systems.

This new imaging technique has opened a powerful observation window into the hidden world of many-particle dynamics. So far, it was generally assumed that the observation of correlated dynamics of electrons and the nuclei in atoms or molecules is beyond any technical realization, since it requires detection techniques, which can detect the electronic motion in the low attosecond domain. Furthermore it requires the observation of several particles at the same time with high momentum resolution. With the recent development of the COLTRIMS imaging technique this observation window is now opened. Instead of measuring two transition energies in atoms or molecules at two a few femtosec separated moments (e.g. with pump and probe laser technique) here the atom or molecule is very suddenly (a few attosec time duration) fragmented into several free particles. Thus the

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time is too short that the momenta of these ejected particles are changed more that a few percent with respect to the initial momenta. The momenta of these ejected particles carry the information on the initial dynamics of the initial state as the pointers of a complex molecular clock. The angular correlation and the momenta of these fragments contain the information on the correlated dynamics of the initial many-particle state. The information can only be read if the momentum resolution is at least a factor of ten better than the initial momenta, i.e. typically 0.1 a.u.. These momenta correspond to electron energies of about 100 meV or ionic recoil energies of less than 100 micro eV.

The principle of the COLTRIMS-method, namely measuring the momentum of the emitted charged particles from an atomic fragmentation process is as simple as determining the trajectory of a thrown stone. From knowing the position, from where the stone was slung and where it hits the target as well as measuring its time-of-flight, the trajectory of the stone and thus its initial velocity vector can precisely be determined. Furthermore, in order to achieve good precision we have to know whether the person, who throws the stone, was at rest in the frame of observation or with which relative velocity this person was moving. Thus to obtain optimal momentum resolution for the exploding fragments one has to bring the fragmenting object to a complete rest in the frame of measurement before the reaction occurs, i.e. if the object is a gas atom or molecule one has to cool it down to sub milli Kelvin temperatures.

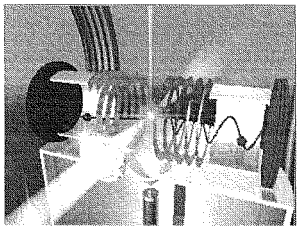

Figure 1. Artist view of the COLTRIMS imaging system (3)

In figure 1 the principle of a COLTRIMS microscope is presented. In a well designed electric field configuration (a static or pulsed as well as with a superimposed magnetic field (1,2,4)) the positively as well as the negatively charged fragments are projected (typically with 4π solid angle) on two position-sensitive detectors. Measuring the impact position on the detector (typically < 0.2 mm resolution) and the time-of-flight (TOF) of the fragment after the moment of fragmentation till hitting the detector, the momentum vector of the charged particle can be determined. To improve the momentum resolution electrostatic lenses can be incorporated into the imaging system, thus the influence of the unknown size of the target region, from where the fragments originate, can completely be eliminated (1,5,6). To detect low energetic and simultaneously also the higher energetic fragments which come from the same fragmentation process magnetic fields and a pulsed electric fields can be used to improve multi-coincidence efficiency. For multi-hit detection it is

possible to use particle detectors based on fast delay-line position read-out (7). Even two particles hitting the detector at the "same" time ($\Delta t < 1$ ns) can be detected individually. The number of detected multi-hits is practically only limited by the electronics needed to store in event mode all information. In future even up to 100 particles per microsec might be detectable if fast transient recorder units with channel resolution of about 0.1 nanosec become available. Thus the COLTRIMS method is indeed powerful like an advanced bubble chamber system or even comparable with modern TPC systems used in high energy physics. Furthermore the rate of fragmentation processes per sec can exceed several 100 kHz.

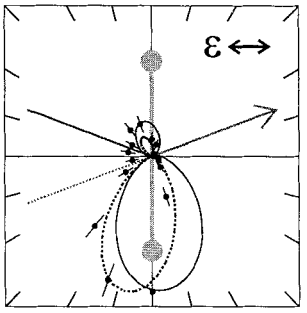

Figure 2. Electron distribution for the fragmentation process $D_2+\gamma \rightarrow D^+ + D^+ + 2e$ induced by a single photon (25 eV above the threshold)(8)

In figure 2 as an example for COLTRIMS data the measured electron distribution for the complete fragmentation photon induced process $D_2+\gamma \rightarrow D^+ + D^+ + 2e$ is shown (8). The direction of the fixed electron 1 is given by the arrow and the angular distribution of the complementary electron 2 is plotted. Additionally, the direction of the electric field vector of the photon ϵ is indicated in the upper right corner of the figure, the measured orientation of the D_2 molecule is given by the bar-bell. The solid curve represents the corresponding He double ionization data and the dashed line calculations by Kheifets et al.(8). The figure shows a small but clear difference between the D_2 and the He target.

In figure 3 for the reaction He+$\gamma \rightarrow$He**(m,m') $\rightarrow He^{1+}(n) + e_{Aug}$ the electron momentum distributions in x and y direction (polarization axis is perpendicular to x and y, light propagation is parallel to x) for autoionizing processes close to the double-ionization-threshold are plotted (9). For the first time the absolute angular differential cross sections could be measured even up to $He^+(n = 16)$ states, where the autoionizing electrons have energies below 100 meV. An overall view of the so far only pre analyzed data shows a β-parameter between β=-0.6 and β=-0.7 for these electrons, a more detailed look indicates clear fluctuations of the β-parameter for different photon energies beyond the statistical error limits. Such doubly excited states are already macroscopic objects with a diameter

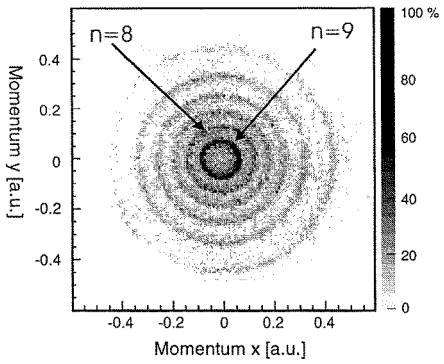

Figure 3. electron momentum distributions for autoionization processes close to the double-ionization-threshold for the reaction He+$\gamma \rightarrow$ He**(m,m') $\rightarrow He^{1+}(n) + e_{Aug}$ (9). x and y are oriented perpendicular to the polarization vector. In order to reveal the rings that are visible in the plot a narrow cut in z-direction at z=0 was applied.

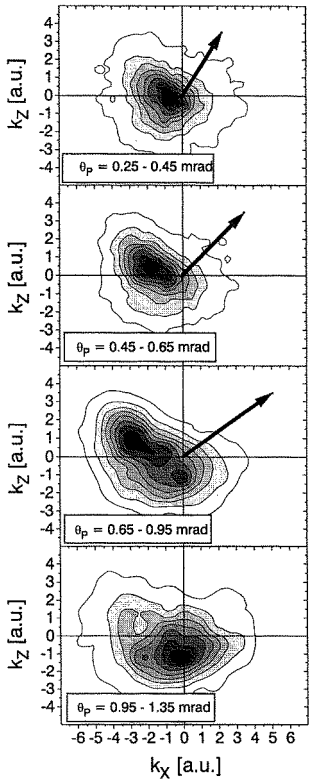

Figure 4. Electron momentum pattern for 630 keV p+He $\rightarrow$ He^{2+}+ H^o + e at different H scattering regimes (10)

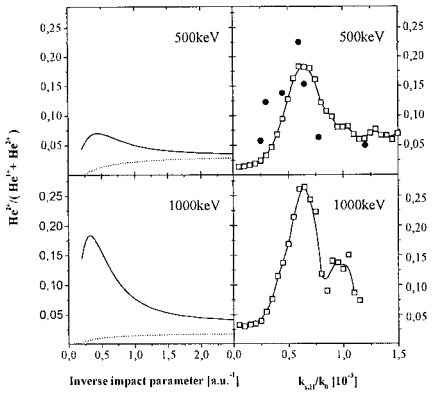

Figure 5. Ratios of transfer ionization to capture probabilities as function of the H transverse momentum. (10)

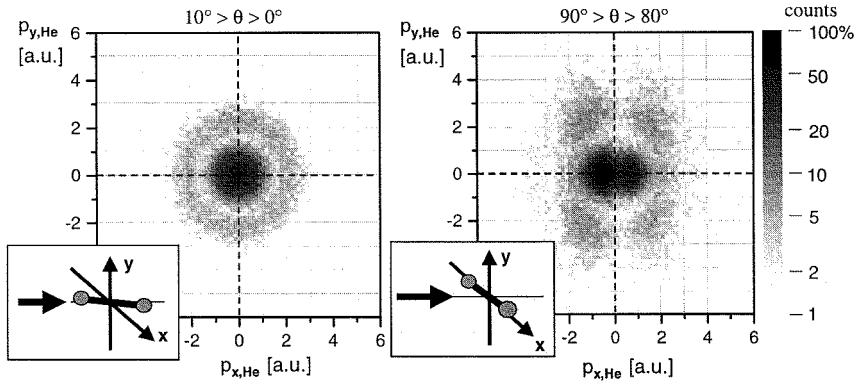

Figure 6. He transversal momentum distribution for the 5 keV/u dissociative electron transfer process $He + H_2^+ \rightarrow He^+(1s) + H_2(1s\sigma_g, 2p\sigma_u) \rightarrow He^+(1s) + H(1s) + H(1s)$: Within the axial recoil approximation the molecular orientation during the collision was determined by the relative motion of the two H fragments. The angle θ between the initial beam direction (z) and the inter nuclear axis was fixed as visualized at the sketches to a) $0° < \theta < 10°$ and b) $80° < \theta < 90°$ with the molecular axis oriented at the x, z plane.

close to a tenth of a micron. This autoionization process is a very interesting subject for testing the transition to quantum chaos.

Through its superb multi-coincidence detection power and high momentum resolution the COLTRIMS technique enables one to investigate even tiny fractions of the correlated wave function, such small contributions which can never be seen even by highest resolution spectroscopy, e.g. the investigation of the non-s^2 off-shell contributions in the asymptotic part of the He ground state momentum wave function. Like the sun corona is visible in a solar eclipse event one can view the non-s^2 contributions of the momentum wave function in the differential cross section of the transfer ionization reaction in fast p+He $\rightarrow$ He^{2+}+ H^o + e collisions. The nuclear p - He^{2+} deflection is an approximate measure of the nuclear impact parameter. The H^o deflection and the momentum of the emitted electron yield in their momentum pattern the electron-electron correlation in the He ground state wave function. In figure 4 the measured electron momentum pattern for four different H scattering angles are shown. The arrows indicate the momentum vector of the captured electron deduced from the H kinematics (10).

In figure 5 the ratios of transfer ionization to capture probabilities are plotted as function of the H transverse momentum. The theoretical predictions (left column) prove that towards smaller transverse momentum (i.e. larger impact parameter) this ratio increases and reaches a pronounced maxima, which is due to the relative increase of non-s^2 contributions with increasing impact parameter.

In figure 6 the He^+ transverse momentum distribution for the reaction channel 5keV/u He on H_2^+ $\rightarrow$ He^{1+}(1s) + H(1s) + H(1s) is shown. The arrow indicates the impact direction of the He projectile. The bar-bell shows the orientation of the molecule (see angular range of molecule orientation above each figure). The transverse momentum distribution shows interesting structure indicating that only in certain orientations and for certain impact parameter (transverse momentum) the electron can be transferred from a He orbital to the molecule resulting in instantaneous fragmentation.

Thus COLTRIMS provides a powerful method to study chemical reactions in full kinematical detail. By varying the projectile velocity it allows to control the chemical reaction time in the atto- and femtosecond time domain. Few- and many-body dynamics in atomic and molecular collisions can be explored with so far unprecedented completeness and precision.

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