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# Dynamics of multiple ionization of atoms and molecules by electron, photon, and ion impact—investigated by the COLTRIMS imaging method

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**Abstract**

Fully differential cross-sections in momentum space for multiple ionization processes of atoms and molecules have been investigated by a multi-coincidence imaging technique, called COLTRIMS (cold target recoil ion momentum spectroscopy) (J. Phys. B 30 (1997) 2917; Nucl. Instrum. Methods B 108 (1996) 425; In: Ullrich, J., Shevelko, V.P. (Eds.), Many Particle Quantum Dynamics in Atomic Fragmentation, Series Atomic, Optical, and Plasma Physics, Vol. 35. Springer, Berlin, 2003; Phys. Rep. 330 (2000) 95). This technique is as powerful as the bubble chamber system in high-energy physics. It has opened a new observation window into the hidden world of many-particle dynamics: correlated many-particle dynamics in Coulombic systems can now be experimentally approached with unprecedented completeness and precision.

The principle of the method, namely measuring the momentum of the emitted charged particles from an atomic or molecular 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 be determined precisely. 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.


*Keywords:* Atomic and molecular physics; Momentum imaging; Many-particle dynamics

## 1. Experimental technique

In Fig. 1, the scheme of the new reaction microscope (acronym: COLTRIMS) is presented. In a well-chosen electric field configuration (static, or pulsed, combined with a superimposed magnetic field) the positively and negatively charged fragments are projected onto two

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position-sensitive detectors (typically with $4\pi$ solid angle). Measuring the impact position on the detector (typically $<0.2$ mm resolution) and the time-of-flight of the fragment (TOF) after the fragmentation, the initial momenta of the particles can be determined. To improve the momentum resolution, electrostatic lenses can be incorporated into the projection system, in order to compensate for the influence of the unknown size of the target region, from where the fragments originate (Doerner et al., 2000). Magnetic or pulsed electric fields can be applied in cases where different species of particles that differ largely in kinetic energy and $q/m$-ratio need to be detected in coincidence. Particle detectors based on fast delay-line position read-out allow for multi-hit detection. Even two particles hitting the detector at the "same" moment ($\Delta t<1$ ns) can be detected simultaneously. The number of detected multi-hits is practically only limited by the electronics needed to store all information in event mode. In future even up to 100 particles/μs might be detectable by using fast transient recorder units with channel resolution of about 0.5 ns. Thus, the COLTRIMS method is indeed as powerful as an advanced bubble chamber system or even comparable with modern TPC systems used in high-energy physics (Doerner et al., 2000; Moshammer et al., 1996). Furthermore rates of fragmentation processes of more than 60 kHz are feasible.

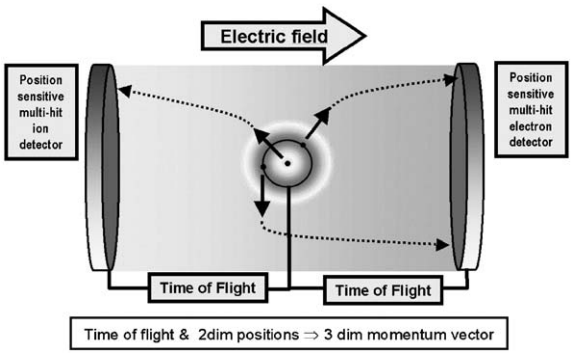


Fig. 1. Schematic diagram of the COLTRIMS imaging system (Doerner et al., 2000).

## 2. Experimental data

In numerous benchmark experiments (Doerner et al., 1996, 2000, 2002, 2003; Dorn et al., 1999, 2001; Dorn, 2003; Jahnke et al., 2002; Knapp et al., 2002; Landers et al., 2001; Ullrich et al., 1997; Ullrich, 2003; Weber et al., 2000a,b, 2001a,b, 2003) quasi-snapshots, with a duration as short an atto-sec, of the correlated dynamics between electrons and nuclei had been made for atomic and molecular objects.

A few examples for COLTRIMS data will now be presented. In Fig. 2, the electron momentum correlation for photon He and $D_2$ double ionization is shown (Weber et al., 2004). The photon energy for both cases is 25 eV above the double ionization threshold. The vector indicates the direction of electron 1 and the contour plot the momentum density distribution of electron 2. The experimental distribution follows nicely the theoretical expectations (not shown). The $D_2$ data are integrated over all molecular orientations. In case of integration no molecular effect is seen. If the data are plotted for selected molecular alignment (see Fig. 3), one observes

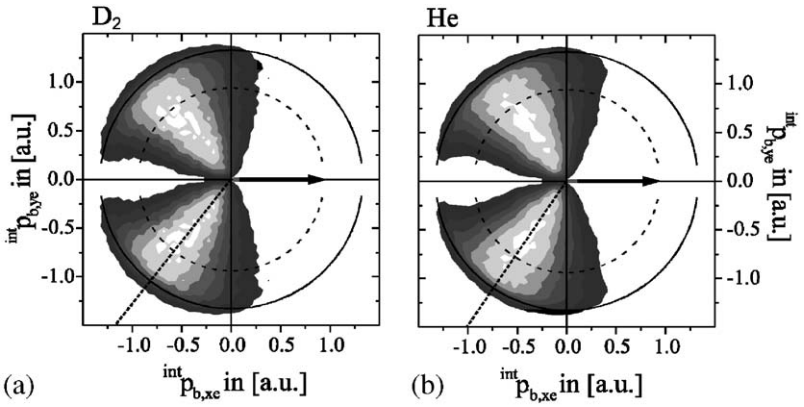


Fig. 2. Momentum distribution of the photo electrons in the internal frame defined by the two outgoing electrons in the photo double ionization of $D_2$ (a) and He (b) with linear polarized light. Both spectra are normalized individually to their maximum intensity and are integrated over the direction of the polarization vector and over the orientation of the molecular axis (case a). One electron is always in the direction of the black arrow, while the momentum distribution of the second electron is shown in linear grey scale. The inner dashed circles represent the events with equal energy sharing. The short dashed lines indicate the most probable emission direction for each target in one quadrant.

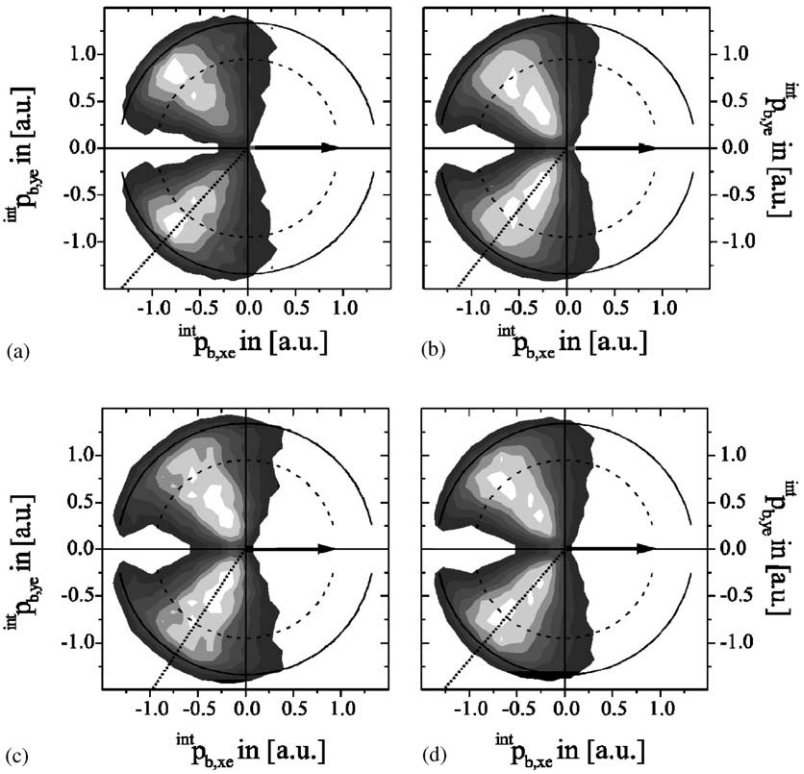


Fig. 3. Electron momentum distribution of $D_2$, as in Fig. 2a, but for selected molecular alignments. (a) The axis is orientated along the first outgoing electron represented by the arrow; (b) the molecular axis is aligned in the plane perpendicular to the black arrow; (c) the molecular axis is orientated perpendicular to the arrow but in the internal electron frame; (d) the molecular axis is aligned perpendicular to the black arrow and comes out of the plane along its perpendicular line.

small but significant differences for the molecule compared to He. These differences get even more pronounced if special molecular alignment is chosen.

The COLTRIMS technique allows such fully differential measurements to be performed with very high multi-coincidence efficiency. Thus the coincidence rate can easily exceed several kHz. Dorn et al. (1999, 2001) and Dorn (2003) have demonstrated that using COLTRIMS so-called (e, 3e) measurements can be performed with excellent resolution and high coincidence efficiency never feasible with traditional techniques. COLTRIMS is also well suited to the study of electron attachment or fragmentation processes for low-energy electron impact ionization of atoms and molecules. Inserting the electron beam in the axis of the recoil spectrometer, electron impact energies from a few eV to several keV are feasible.

Very recently, COLTRIMS has been applied to study multi-fragmentation processes in strong femtosecond laser pulses. Weber et al. (2000a,b) and Moshammer et al. (2003) measured the correlated electron momentum pattern for double ionization of rare gases. In Fig. 4, the correlated electron momentum pattern of argon double ionization is shown for a 800 nm, 50 fs laser pulse of $3.75 \times 10^{14}\,W/cm^2$. This ionization can only occur if approximately 50 photons are absorbed. Since the events are most likely to be found in those quadrants (Fig. 4), where both electron momenta are either positive or negative, the data prove that both electrons are preferably emitted in the same direction. This is very different to ionization by a single photon: it can be explained by the so-called re-scattering process (Corkum, 1993).

In Fig. 5 for 17.5 keV/u $He^{2+}$ on He the momentum exchange pattern, projected on the nuclear scattering plane, for the one electron transfer ionization channel is shown. In the transverse direction only nuclear momentum exchange is visible, whereas the momentum of the ionized electron is close to zero. In longitudinal direction the projectile is about 1 a.u. slowed down and the recoil ion is slightly pushed forward. Traditional electron spectroscopy would hardly resolve the details of the electron momentum structure, but the COLTRIMS imaging technique demonstrates here its full power. The beauty of the electron momentum pattern (measured in coincidence with the nuclei with respect to the scattering plane) becomes visible with high resolution. The zoomed electron momentum distribution clearly

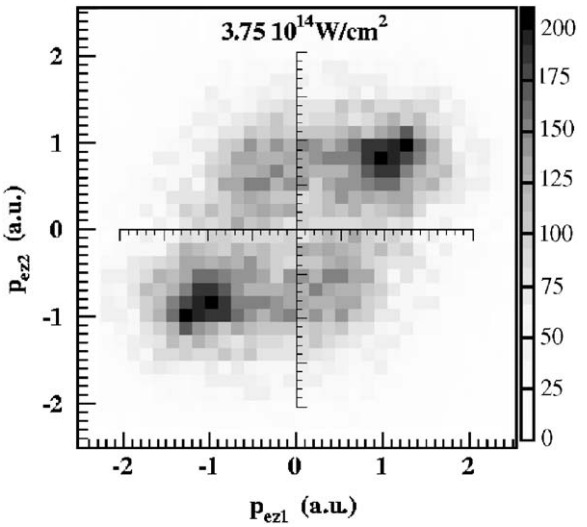


Fig. 4. Momentum correlation between the two emitted electrons when an $Ar^{2+}$ ion is produced in the focus of a 220 fs, 800 nm laser pulse at peak intensities of $3.75 \times 1014$ W/cm$^2$. The horizontal axis shows the momentum component of one electron along the polarization direction of the laser field; the vertical axis shows the same momentum component of the corresponding second electron. The same sign of the momenta for both electrons means emission to the same half sphere. The data are integrated over the momentum components in the direction perpendicular to the polarization (Weber et al., 2000a,b).

shows (Doerner et al., 1996) that the electrons are promoted to the continuum by quasi-molecular electron promotion processes (Ovchinnikov and Macek, 1995) rather then by the saddle point mechanism (Olson et al., 1987).

The data presented above show that COLTRIMS is a powerful technique to study the details of practically all the multiple ionization processes of atoms and molecules. It can reveal details of ionization mechanisms never seen before. It has provided new insights into such ionization processes, which might be of crucial importance for understanding the damage processes in biological tissues particularly for low-energy photon or electron impact.

That ionization processes that often do not follow simple Born-approximation predictions can be deduced from the following ionization data for fast heavy ion impact presented below. Ionization cross-sections are often analyzed in terms of an effective charge. Thus, one expects that, independent of the nuclear charge, ions with the same ionic charge show the same ionization strength and the electron emission pattern should be similar for all such collision systems. The Born approximation predicts that for fast, completely stripped ion impact the electron emission peaks near zero energy (Lab system) and most of these slow electrons (named TBE electrons in Fig. 6) are emitted towards 90° with respect to the ion impact. As characteristic features we observe: (1) in the forward direction (0°) the CUSP (ECC electron emission peak in Fig. 6) is seen centered

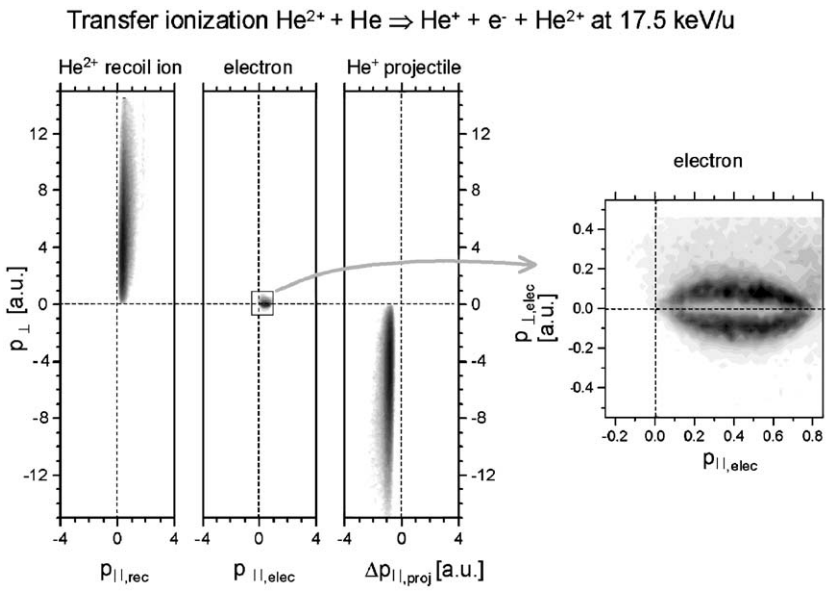


Fig. 5. Momentum exchange pattern projected on the nuclear scattering plane for the transfer ionization channel at 17.5 keV/u (u is the atomic mass unit) $He^{2+}$ on He collisions (Doerner et al., 1996).

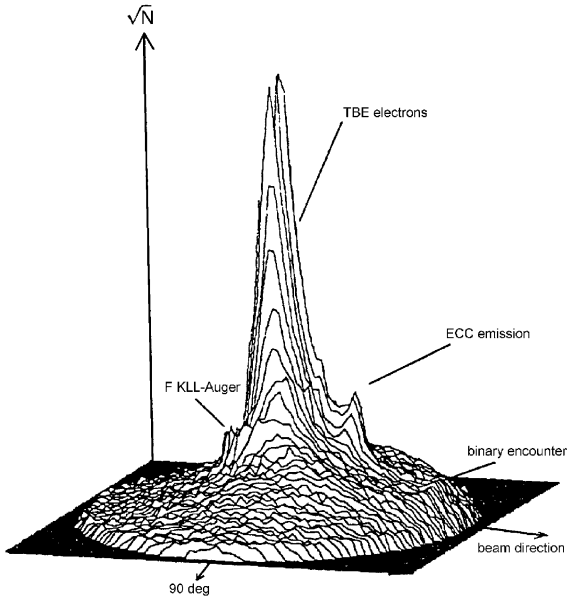


Fig. 6. Electron momentum distribution for 3.6 MeV/u for $Ne^{10+}$ impact on $SF_6$ (see text) (Bechthold et al., 1997).

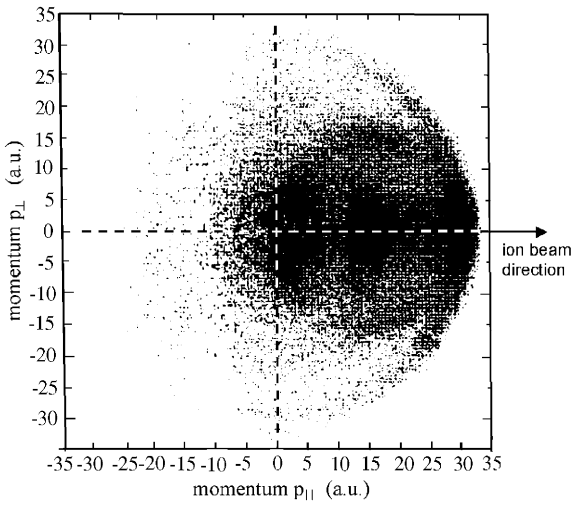


Fig. 7. Electron momentum distribution for 5.88 MeV/u for $U^{29+}$ impact on $C_3F_8$ (see text) (Bechthold et al., 1997).

at the ion velocity $v_p$ and (2) a binary encounter ridge (circle with radius $v_p$) centered at the CUSP position.

As seen in Fig. 7, the electron emission pattern can deviate quite significantly from such simple predictions (Born approximation) and vary strongly between collision systems. In Fig. 7, the TBE peak has nearly disappeared and suddenly the binary encounter ridge and CUSP are the dominating structures in the electron

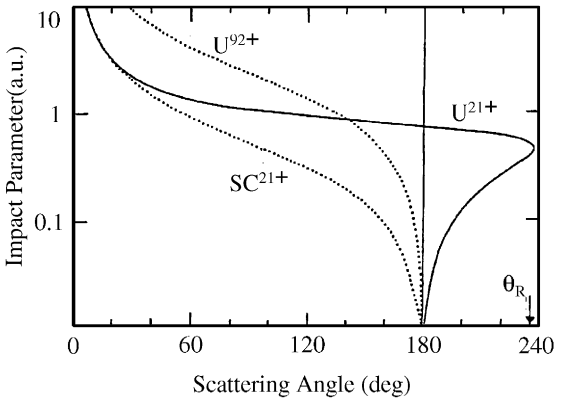


Fig. 8. Relation between electron impact parameter and electron scattering angle (projectile system, here 180° projectile system correspond to 0° Lab system).

momentum distribution: the ridge shows even clear interference structures. The reason for these variations is quite simple, but it is often neglected in theoretical interpretations of ionization processes in fast ion-atom/molecule collisions.

The final electron momentum depends strongly on the interacting ionic force (derivative of the potential) but not on the strength of the potential (i.e. a huge but constant potential does not ionize at all). In case of electron screening, like in $U^{29+}$ ions, the effective nuclear Coulomb force is huge and it bends the ionized target electrons much stronger in $U^{29+}$ than in $U^{92+}$. Thus, as seen in Fig. 8, the relation between impact parameter and electron scattering (emission angle) depends strongly on that screening. $U^{21+}$ bends thus many more electrons towards 180° (0° in Lab system) than $U^{92+}$ or $Sc^{21+}$. Since also several trajectories (impact parameters) contribute to the same final scattering angle, the ionization amplitudes do interfere, creating the strong interference structures. Note that the 0° binary encounter peak corresponds to electrons with a momentum of about 30 a.u., i.e. a kinetic energy of about 13 keV. It is interesting to notice that $U^{0+}$ compared to $U^{92+}$ (moving with the same velocity) emits about 100 times more electrons at 0° with a velocity twice the ion velocity.

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## References


Bechthold, U., Hagmann, S., Bhalla, C., Ullrich, J., Bathelt, B., Bohris, A., Moshammer, R., Kraft, G., Ramm, U., Schmidt-Böcking, H., 1997. Intra-atomic double scattering of binary encounter electrons in collisions of fast, heavy-ions with atoms and molecules. Phys. Rev. Lett. 79, 2034.

Corkum, P., 1993. Plasma perspective on strong field multiphoton ionization. Phys. Rev. Lett. 71, 1994.

Dorn, A., 2003. Helium double ionization in collisions with electrons. In: Ullrich, J., Shevelko, V.P. (Eds.), Many Particle Quantum Dynamics in Atomic Fragmentation, Series Atomic, Optical, and Plasma Physics, Vol. 35. Springer, Berlin, pp. 339–350.

Dorn, A., Moshammer, R., Schröter, C.D., Zouros, T.J.M., Schmitt, W., Kollmus, H., Mann, R., Ullrich, J., 1999. Double ionization of helium by fast electron impact. Phys. Rev. Lett. 82, 2496.

Dorn, A., Kheifets, A., Schröter, C.D., Najjari, B., Höhr, C., Moshammer, R., Ullrich, J., 2001. Double ionization of helium by electron-impact: complete pictures of the four-body breakup dynamics. Phys. Rev. Lett. 86, 3755.

Doerner, R., Khemliche, H., Prior, M.H., Cocke, C.L., Gary, J.A., Olson, R.E., Mergel, V., Ullrich, J., Schmidt-Böcking, H., 1996. Imaging of saddle point electron emission in slow p–he collisions. Phys. Rev. Lett. 77, 4520.

Doerner, R., Mergel, V., Jagutzki, O., Spielberger, L., Ullrich, J., Moshammer, R., Schmidt-Böcking, H., 2000. Cold target recoil ion momentum spectroscopy: a 'momentum microscope' to view atomic collision dynamics. Phys. Rep. 330, 95–192.

Doerner, R., Weber, Th., Weckenbrock, M., Staudte, A., Hattass, M., Moshammer, R., Ullrich, J., Schmidt-Böcking, H., 2002. Multiple ionization in strong laser fields. In: Bederson, B., Walther, H. (Eds.), Advances in Atomic and Molecular Physics, Vol. 48. Academic Press, New York, pp. 1–36.

Doerner, R., Schmidt-Böcking, H., Mergel, V., Weber, T., Spielberger, L., Jagutzki, O., Knapp, A., Bräuning, H.P., 2003. From atoms to molecules. In: Ullrich, J., Shevelko, V.P. (Eds.), Many Particle Quantum Dynamics in Atomic Fragmentation, Series Atomic,Optical, and Plasma Physics, Vol. 35. Springer, Berlin, pp. 245–257.

Jahnke, T., Weber, Th., Landers, A.L., Knapp, A., Schössler, S., Nickles, J., Kammer, S., Jagutzki, O., Schmidt, L., Czasch, A., Osipov, T., Arenholz, E., Young, A.T., Muiño, R.D., Rolles, D., García de Abajo, F.J., Fadley, C.S., Van Hove, M.A., Semenov, S.K., Cherepkov, N.A., Rösch, J., Prior, M.H., Schmidt-Böcking, H., Cocke, C.L., Dörner, R., 2002. Circular dichroism in k-shell ionization from fixed-in-space CO and $N_2$ molecules. Phys. Rev. Lett. 88, 073002.

Knapp, A., Kheifets, A., Bray, I., Weber, Th., Landers, A.L., Schössler, S., Jahnke, T., Nickles, J., Kammer, S., Jagutzki, O., Schmidt, L.Ph.H., Osipov, T., Rösch, J., Prior, M.H., Schmidt-Böcking, H., Cocke, C.L., Dörner, R., 2002. Mechanisms of photo double ionization of helium by 530 eV photons. Phys. Rev. Lett. 89, 033004.

Landers, A., Weber, Th., Ali, I., Cassimi, A., Hattass, M., Jagutzki, O., Nauert, A., Osipov, T., Staudte, A., Prior, M.H., Schmidt-Böcking, H., Cocke, C.L., Dörner, R., 2001. Photoelectron diffraction mapping: molecules illuminated from within. Phys. Rev. Lett. 87, 013002.

Moshammer, R., Unverzagt, M., Schmitt, W., Ullrich, J., Schmidt-Böcking, H., 1996. A 4 ô recoil-ion electron momentum analyser: a high-resolution 'microscope' for the investigation of the dynamics of atomic, molecular and nuclear reactions. Nucl. Instrum Methods B 108, 425.

Moshammer, R., Fischer, D., Kollmus, H., 2003. Recoil-ion momentum spectroscopy and "reaction microscopes". In: Ullrich, J., Shevelko, V.P. (Eds.), Many Particle Quantum Dynamics in Atomic Fragmentation, Series Atomic,Optical, and Plasma Physics., Vol. 35. Springer, Berlin, pp. 33–57.

Olson, R.E., Gay, T.J., Berry, H.G., Hale, E.B., Irby, V.D., 1987. Saddle-point electrons in ionizing ion-atom collisions. Phys. Rev. Lett. 59, 36.

Ovchinnikov, S.Yu., Macek, J.H., 1995. Electron-energy and angular-distribution theory for low-energy ion-atom collisions. Phys. Rev. Lett. 75, 2474.

Ullrich, J., 2003. Single and multiple ionization in strong ion-induced fields. In: Ullrich, J., Shevelko, V.P. (Eds.), Many Particle Quantum Dynamics in Atomic Fragmentation, Series Atomic,Optical, and Plasma Physics, Vol. 35. Springer, Berlin, pp. 379–405.

Ullrich, J., Moshammer, R., Dörner, R., Jagutzki, O., Mergel, V., Schmidt-Böcking, H., Spielberger, L., 1997. Recoil-ion momentum spectroscopy. J. Phys. B 30, 2917 (topical review).

Weber, Th., Weckenbrock, M., Staudte, A., Spielberger, L., Jagutzki, O., Mergel, V., Afaneh, F., Urbasch, G., Vollmer, M., Giessen, H., Dörner, R., 2000a. Recoil-ion momentum distributions for single and double ionization of helium in strong laser fields. Phys. Rev. Lett. 84, 443.

Weber, Th., Giessen, H., Weckenbrock, M., Urbasch, G., Staudte, A., Spielberger, L., Jagutzki, O., Mergel, V., Vollmer, M., Dörner, R., 2000b. Correlated electron emission in multiphoton double ionization. Nature 405, 658.

Weber, Th., Khayyat, Kh., Dörner, R., Rodríguez, V.D., Mergel, V., Jagutzki, O., Schmidt, L., Müller, K.A., Afaneh, F., Gonzalez, A., Schmidt-Böcking, H., 2001a. Abrupt rise of the longitudinal recoil ion momentum distribution for ionizing collisions. Phys. Rev. Lett. 86, 224.

Weber, Th., Weckenbrock, M., Staudte, A., Hattass, M., Spielberger, L., Jagutzki, O., Mergel, V., Schmidt-Böcking, H., Urbasch, G., Giessen, H.W., Bräuning, H., Cocke, C.L., Prior, M.H., Dörner, R., 2001b. Atomic dynamics in single and multiphoton double ionization: an experimental comparison. Opt. Express 8, 368–376.

Weber, Th., Weckenbrock, M., Balser, M., Schmidt, L., Jagutzki, O., Arnold, W., Hohn, O., Schöffler, M., Arenholz, E., Young, T., Osipov, T., Foucar, L., De Fanis, A., Muiño, R.D., Schmidt-Böcking, H., Cocke, C.L., Prior, M.H., Dörner, R., 2003. Auger electron emission from fixed-in-space CO. Phys. Rev. Lett. 90, 153003.

Weber, Th., Czasch, A., Jagutzki, O., Müller, A., Mergel, V., Kheifets, A., Jeagin, J., Rotenberg, E., Meigs, G., Prior, M.H., Daveau, S., Landers, A.L., Cocke, C.L., Osipov, T., Schmidt-Böcking, H., Dörner, R., 2004. Fully differential cross sections for photo double ionization of fixed-in-space $D_2$. Phys. Rev. Lett. submitted for publication. Phys. Rev. Lett. 92, 163001–1.