

# RECOIL ION MOMENTUM SPECTROSCOPY MOMENTUM SPACE IMAGES OF ATOMIC REACTIONS

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## ABSTRACT

Recoil-ion momentum spectroscopy is a powerful tool for investigating the dynamics of ion, electron or photon impact reactions with atoms or molecules. It allows to measure the three-dimensional momentum vector of the ion from those reactions with high resolution and  $4\pi$  solid angle. In many cases already the recoil-ion momentum distribution alone unveils directly the physical processes dominating the reaction. The most detailed information, however is gained by combining the recoil-ion momentum measurement with the coincident detection of momentum vector of one or more emitted electrons or a measurement of the momentum exchange of the projectile. By such many particle momentum imaging one obtains a fully differential cross section of the reaction, i.e. for each registered event one measures the momenta of all particles and the full final state momentum space is covered in one experiment. Thus the experiment yields the square of the final state wave function of the reaction in momentum space. Such multidimensional data arrays can be sorted in many different ways after the actual experiment. Examples for ion impact ionization are discussed.

## INTRODUCTION

Interaction of charged particles or photons with atoms or molecules are an inter-

esting subject of active research in their own right and are a tool to obtain information about internal dynamics and structure of the target atom or molecule. Such reactions allow to study the dynamics of quantum mechanical coulomb systems, which is the governing factor for much of the structure and evolution in our everyday world. If the fragments of atomic reactions have no relevant internal structure, the final state of the process is fully determined by the momentum vectors of the particles and their spins. To gain understanding of the process and for a detailed check on existing theories it is desirable to determine cross sections as highly differential as possible. Extensive work on such kinematically complete experiments has been carried out by coincident electron detection in  $(\gamma,2e)$ ,  $(e,2e)$  and  $(e,3e)$  reactions. Many examples and references can be found in this book. The multi particle momentum microscopes for atomic collisions which are the subject of this article differ from such coincident electron detection techniques mainly by the fact that they cover the full final state phase space of the reaction and that they can be used for the study of ion atom collisions.

**Cold Target Recoil Ion Momentum Spectroscopy COLTRIMS** is the key technology for such kinematically complete experiments with  $4\pi$  solid angle. For reactions with only two particles in the final state, like single electron capture or photo single ionization, the recoil-ion measurement delivers already the complete momentum information. The momentum of the second particle can be inferred from momentum conservation. For electron capture the energy loss and the scattering angle of the projectile can thus be measured via the recoil-ion, and for photo single ionization the recoil-ion measurement is equivalent to the detection of the photoelectron. For more complex reaction with  $n$  particles in the final state  $(n-1)$  of those have to be detected to yield complete momentum information. Also in principle one is free to choose which of the particles to observe, it is in many respects advantageous to detect the recoil-ion. One reason is that the recoil-ion momentum itself, even if one integrates over all other observables, is already a rich source of information, in particular since the ion measurement yields additionally the information on the charge state and thus the multiplicity of the process. By observing the recoil-ion momentum for example Compton scattering has been separated from photoabsorption [1], electron-electron interaction could be isolated from other interactions in projectile electron loss [2, 3, 4] and in transference-ionization [5], post collision interaction in multiple ionization [6, 7, 8] and  $Q$ -values in multiple capture collision [9, 10] could be studied. We discuss the kinematics of recoil-ion production in ion atom collisions in the second section together with some applications.

A second reason for the benefit of recoil-ion momentum measurements is the high resolution and  $4\pi$  solid angle which can be achieved. The recoil-ions are typically so low energetic, that they can be efficiently collected by weak electrostatic fields. A resolution of 0.05 a.u. for each momentum component together with  $4\pi$  solid angle is routinely achieved. In an ion-atom collision the projectile typically loses only a very small fraction of its total energy, thus for projectile detection one has to observe in the laboratory frame a tiny change of a huge momentum vector. Projectile momentum measurements are therefore limited by the achievable quality of the beams. The recoil-ion momentum measurement as well as any electron momentum measurement are almost unaffected by this problem, since the observed particles emerge from the atom which is at rest in the laboratory. Thus by detecting the ion momentum for pure capture reactions high resolution energy gain and projectile scattering angle measurements can be performed even for fast projectile [11, 12]. The same can be achieved for ionization reactions if the recoil-ion and all electrons are measured [13]. For 1 MeV/u Ne projectiles for example a recoil-ion momentum resolution of 0.05 a.u. corresponds to a projectile scattering

angle resolution of  $2 \times 10^{-7}$  rad and an energy gain resolution of  $\Delta E/E=4 \times 10^{-7}$ .

Technically the multi particle momentum space imaging requires a well localized reaction zone. Since the typical recoil-ion momenta are subthermal an internally cold gas target is necessary to achieve sufficient resolution. Localization and cooling is today achieved by using supersonic gas jet targets. The reaction fragments (electrons and recoil-ions) are guided by electric and magnetic fields towards position sensitive channel plate detectors. The momenta can be calculated from the time-of-flight and the position of impact on the detector. Different designs for field geometries have been use, more details can be found for example in [14, 15, 12, 16]. Figures showing typical spectrometer designs can be found the the articles of Mergel et al and Spielberger et al in this volume. A discussion of magnetic confinement for electrons, which is essential to imaging of higher energetic electrons can be found in [15].

Also **COLTRIMS** is still a young technique it has been already applied to many fields of atomic collision physics such as: single capture [12, 17, 18, 19, 20] and single ionization by ion impact [13, 21, 22, 23, 24, 25], multiple electron processes like double capture [26], transfer ionization [5, 18, 17] and multiple ionization [27, 28, 29, 6, 7, 8, 23], electron impact ionization [30, 31], photon induced double ionization by linear [32, 33, 16, 34, 35] and circular polarized light [36], Compton scattering [1, 37] and electron emission from aligned molecules [38]. The energies of the projectiles range for charged particles from hundred eV electrons to GeV bare Uranium [23] and antiprotons [25] and for photons from threshold to 100 keV. This article can not even cover a subsection if this fruitful work. A more complete review can be found in [14, 38]. Here we discuss only a few selected examples from the subfield of ion atom collisions. Section 2 focuses on the kinematics of recoil-ion production in order to show the wealth of information which can be obtained from the momentum measurement of the recoil-ion, section 3 gives examples for fully differential cross sections for ion impact single ionization.

## KINEMATICS OF ION ATOM COLLISIONS

For heavy ion collisions the energy loss or gain of the projectile is typically small compared to the total energy of the projectile and the scattering angles are in the range of only a few mrad. In this case the momentum components of the recoil-ion longitudinal to the beam ( $p_{\parallel rec}$ ) and perpendicular to the beam ( $p_{\perp rec}$ ) are fully decoupled and carry different information about the collision process.

The longitudinal momentum of the recoil-ion for a complex reaction involving multiple target ionization, projectile ionization (loss), electron capture and excitation, can be calculated from energy and momentum conservation to be:

$$\begin{aligned}
 p_{\parallel rec} &= p_{\parallel rec}^{capture} + p_{\parallel rec}^{ionisation} + p_{\parallel rec}^{loss} + p_{\parallel rec}^{excitation} & (1) \\
 p_{\parallel rec}^{capture} &= -\frac{n_c v_{pro}}{2} + \frac{Q_c}{v_{pro}} \\
 p_{\parallel rec}^{ionization} &= \sum_{k=1}^{n_i} \frac{E_{bind}^k + E_{cont}^k}{v_{pro}} - p_{\parallel}^{e_k} \\
 p_{\parallel rec}^{loss} &= \sum_{j=1}^{n_i} \frac{E_{bind}^j + E_{cont}^j}{v_{pro}}
 \end{aligned}$$

$$p_{\parallel rec}^{excitation} = \frac{E_{exc}}{v_{pro}}$$

Atomic units are used throughout this paper.  $n_c$ ,  $n_i$  and  $n_l$  is the number of captured, ejected target and projectile electrons.  $Q_c$  is the differences in binding energy in the initial and final state summed over all captured electrons (exothermic reactions leading to  $Q_c > 0$ ),  $E_{bind}$  and  $E_{cont}$  are the binding and continuum energies of the target and projectile electron in their parent rest frame and  $p_{\parallel}^{ek}$  is the longitudinal momentum of target electron  $k$  in the final state.  $E_{exc}$  is the sum of the excitation energies of target and projectile (if not already counted in  $Q_c$ ).

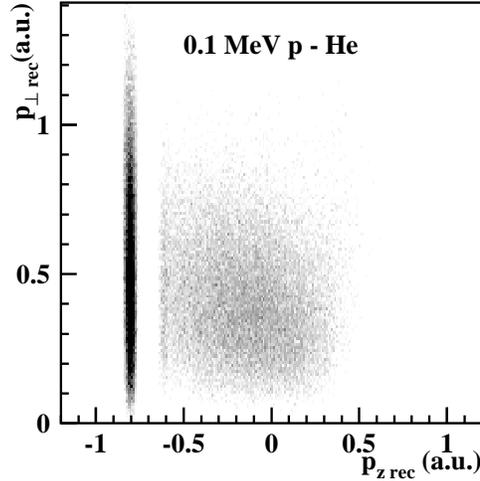


Figure 1: Momentum distribution of  $He^{1+}$  recoil-ions created by 0.1 MeV p impact. The horizontal axis gives the momentum in beam direction, the vertical axis gives the momentum component transverse to the beam. The line at a longitudinal momentum of about -0.8 a.u. results from single electron capture to the ground state of the projectile, the broad distribution mostly from single ionization with a small unresolved contribution due to capture to excited states and capture plus target excitation (from [39]).

Figure 1 shows for illustration of equation 1 the momentum distribution of  $He^{1+}$  recoil-ions from 0.1 MeV p impact on He. The line of at  $p_{\parallel rec} = -0.8$  a.u. results from collisions in which the electron is captured to the ground state of the proton. The discrete values of the energy gain leads to discrete values of longitudinal recoil-ion momentum. The transverse momentum distribution of these recoil-ions mirrors exactly the projectile scattering. The broad distribution around momentum zero results mostly from single ionization. Since the momenta of three particles can be coupled in many ways, single ionization shows a continuous distribution of recoil-ion momenta. There is however a lower threshold to the longitudinal momentum for single ionization of

$$p_{\parallel rec}^{min} = -\frac{v_{pro}}{2} - \frac{E_{bind}}{v_{pro}}, \quad (2)$$

which is related to electrons travelling in forward direction with the velocity of the projectile (cusp electrons) [40]. A more detailed discussion of the momentum exchange in single ionization is given in the next section.

If the projectile carries an electron and is ionized in the collision the recoil-ion receives the additional longitudinal forward momentum  $p_{\parallel rec}^{loss}$ . This is due to the fact that

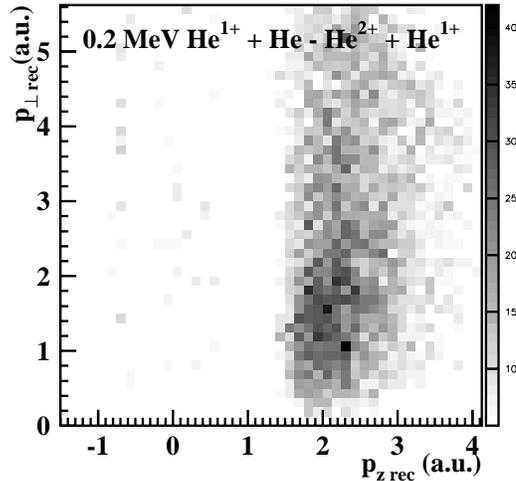


Figure 2: Momentum distribution of recoil-ions for the reaction  $0.2 \text{ MeV He}^{1+} + \text{He} \rightarrow \text{He}^{2+} + \text{He}^{1+} + 2e^{-}$ . The horizontal axis gives the momentum in beam direction, the vertical axis give the momentum component transverse to the beam. The forward emission of the recoil-ions compensates for the momentum loss of the projectile which is necessary to ionize it (compare to data in [3, 2]).

the projectile has to loose at least the momentum according to the binding energy of the emitted electron and the collision partner has to balance this backward momentum transfer to the projectile. Figure 2 shows this forward recoil-ion emission for the reaction



In this reaction the electron loss of the projectile is accompanied by target ionization. Thus the full longitudinal momentum of the recoil-ion is the sum  $p_{\parallel \text{rec}}^{\text{loss}} + p_{\parallel \text{rec}}^{\text{ionization}}$ . At  $0.2 \text{ MeV}$   $p_{\parallel \text{rec}}^{\text{ionization}}$  for reaction 3 is smaller than  $p_{\parallel \text{rec}}^{\text{loss}}$ . The ionization of the projectile proceeds via an interaction of the projectile electron with the target nucleus (ne), throwing the recoil-ion forward. At  $1 \text{ MeV}$  however, an additional reaction mechanism becomes possible. The energy is high enough that an interaction between the target electron and the projectile electron could now lead to projectile ionization ((ee)-mechanism) (see [41, 42] and references therein). In this case the electron is emitted forward, balancing the projectile momentum loss, while the recoil-ion is a spectator receiving only little momentum. The measurement of the recoil-ion momentum thus offers a unique possibility to separate the (ee) and (ne)-mechanism of electron loss. Dörner et al [3] and Wu et al [2, 4] have found two peaks in the recoil-ion momentum distribution from loss reactions which could be attributed to the (ee) and (ne)-mechanism.

## SINGLE IONIZATION BY CHARGED PARTICLES

The mechanism leading to single ionization in ion-atom collisions strongly depend on the velocity of the projectile and the strength of the perturbation it causes to the target atom. The dominant physical processes underlying such reactions can be unveiled by looking at the momentum exchange pattern among the three particles involved. Also in general single ionization is a three-body process one can discuss three extreme scenarios in which one of the three possible two-body momentum exchanges, electron-recoil-ion

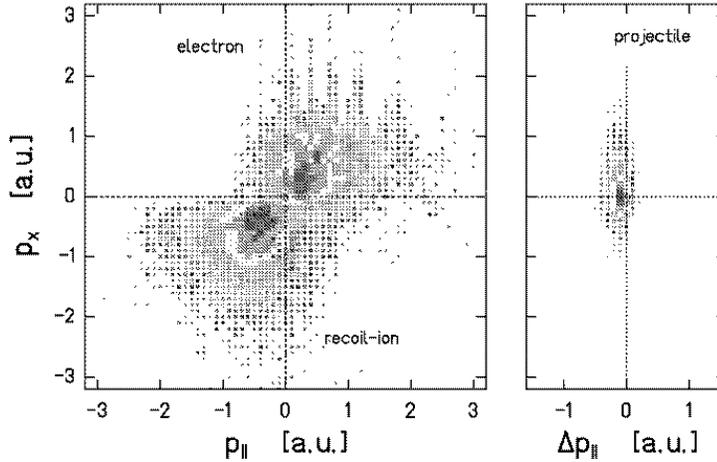
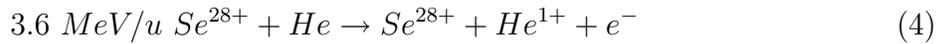


Figure 3: 3.6 MeV/u  $Se^{28+} + He \rightarrow He^{1+} + e^{-} + Se^{28+}$ . Projection in momentum space of all particles in the final state onto the plane defined by beam (horizontal axis) and the recoil-ion transverse momentum (vertical axis). The cluster size represents the corresponding doubly differential cross section  $d^2\sigma/(dp_x dp_{\parallel})$  on logarithmic scale (from [22]) (compare to figure 4.)

Dominance of (er) interaction is characteristic for ionization by photo absorption. Since the photon momentum is small compared to the momentum of the emitted electron, i.e. the photon delivers energy but (almost) no momentum to the system, the electron momentum must be compensated by the recoil-ion momentum. Such dominance of (er) momentum exchange has been found for ionization of Helium by 3.6MeV/u  $Ni^{24+}$  3.6 MeV/u  $Se^{28+}$  and 1GeV/u  $U^{92}$  [13, 43, 44, 22]. Figure shows the momentum exchange for the reaction



The momenta are projected onto a plane defined by the beam direction (beam from left to right) and the momentum vector of the recoil-ion. The electron and recoil-ion are clearly found to be emitted opposite compensating their momenta while the projectile suffers only small momentum exchange. In addition the electron emission is found to be forward directed while the recoil-ions are backward emitted. This polarization is caused by the postcollision interaction with the long range positive charge of the emerging highly charged projectile. This effect is well described by CTMC calculations [13] as well as by CDW calculations [45, 46, 40]. The common nature of fast charged particle and photons interacting with matter was discussed already by Fermi, Weizsäcker and Williams [47, 48]. In their approach ionization of an atom by charged particles is modeled as photoionization by a field of equivalent photons of various energies (equivalent photon method). The photon field is obtained by a Fourier transformation of the time and impact parameter dependent electromagnetic field of the passing projectile. At 1 GeV/u  $U^{92+}$  ion generates a sub attosecond ( $10^{-18}$ s) superintense ( $I > 10^{19} \text{ W/cm}^2$ ) field of virtual photons, shorter and more intense than any laser. At 3.6MeV/u the Weizsäcker-Williams approach is generally not expected to be applicable for a reliable

calculation, figure 3 shows, however, that already at 3.6 MeV the momentum exchange with the projectile is very small.

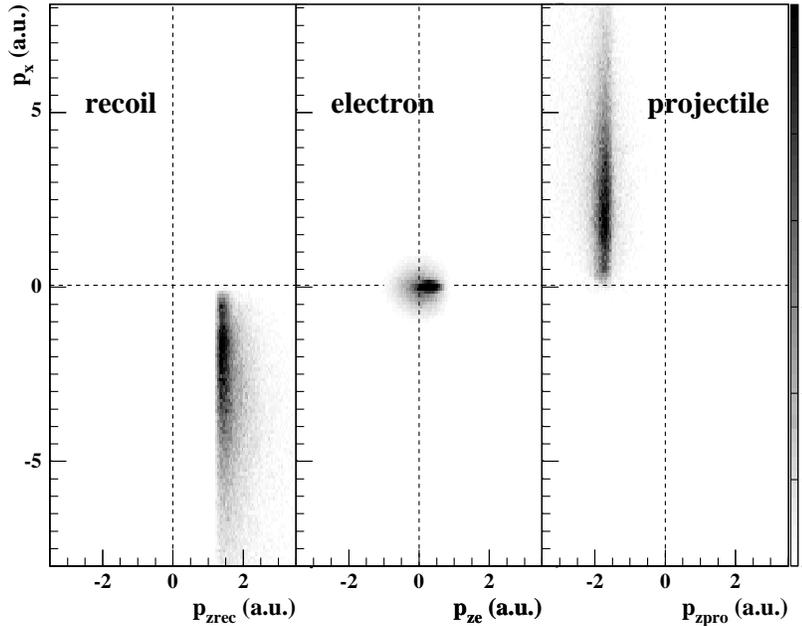


Figure 4: Projection of the momentum transfer vectors of recoil-ion, electron and projectile in the final state onto the plane defined by the projectile beam and the momentum vector of the recoil-ion for  $10 \text{ keV/u } p + He \rightarrow He^{1+} + e^- + p$ . The  $+p_z$  axis is parallel to the incoming projectile direction, the  $-p_y$  axis is parallel to the final transverse momentum component of the recoil-ion. The grey scale represents the corresponding doubly differential cross section  $d^2\sigma/(dp_x dp_z)$  on linear scale (similar to [49]).

The equivalent presentation to figure 3 for the reaction



is shown in figure 4. At these low velocities the (pr) momentum exchange dominates by far. The electrons receive only very little momentum and in the transverse direction the projectile is scattered at the target nucleus. Therefore in this case an impact parameter can be inferred from the transverse momentum exchange. Contrary to figure 3 the recoil-ions are emitted in the forward direction. This is a direct consequence of the  $E_{bind}^k/v_{pro}$  term in equation 1. In terms of reaction mechanism this momentum exchange pattern indicates that for such slow collisions an intermediate quasimolecule is formed. The electron in this quasimolecule acts as a glue which attaches the recoil nucleus for a short time to the forward moving projectile, allowing to transmit forward momentum. When the two nuclei separate the electron relaxes in most cases to a bound state of the projectile or the target. Ionization is a weak channel at these velocities. In the unlikely case that the electron is promoted to the continuum (which is selected in figure 4) it is stranded with small momenta in between the two centers of projectile and recoil-ion.

The mechanism for the promotion of the electron to the continuum have been studied intensely theoretically [50, 51, 52, 53, 54, 55]. Experimentally this question can

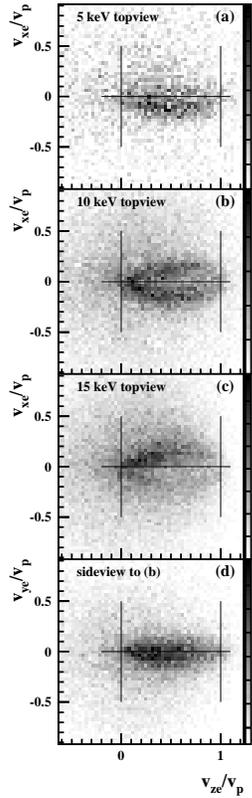


Figure 5: Projection of the velocity distribution of electrons for single ionization in 5 (a), 10 (b) and 15 (c) keV p-He collisions onto the scattering plane, defined by the incoming projectile axis ( $z$ ) and the final momentum vector of the recoil-ion, emerging to the  $-x$  direction. The target center is at  $(0,0)$  the projectile at  $(1,0)$  and the saddle at  $(0.5,0)$ . The data for 10 keV are for a transverse momentum transfer in the interval  $k_{\perp rec} = 1-5$  a.u.. For the other energies this momentum range is scale by  $1/v_{pro}$  in order to sample approximately the same range of impact parameters. (d) sideview to (b), i.e. projection onto the  $y-z$  plane perpendicular to the  $x-z$  scattering plane (from [24]).

be addressed by examining the momentum distribution of the electrons in more detail. Figure shows a blow up of the electron distribution similar to that from figure 4 for various impact energies. The beam direction is horizontal, the recoil-ion is emitted downwards. The electron velocity is shown in units of the projectile velocity. Thus electrons centered at the target are found at  $(0,0)$  those travelling with the projectile at  $(1,0)$  and the saddle point of the two-center potential is located at  $(0.5,0)$ . At 10 keV a symmetric horseshoe like emission pattern is found. It has a minimum on the saddle point with two maxima below and above. This has been interpreted as the characteristics of a p-wave on the saddle [50, 52]. At 5 and 15 keV the symmetry is broken (see [52] for an interpretation). The electron emission also shows a strong impact parameter dependence. The electron emission pattern as well as the impact parameter dependence of the ionization process is well reproduced by CTMC calculation using a Wigner initial state distribution.

At higher proton velocities the situation changes completely. For 0.2-1.3 MeV p impact on helium Weber and coworkers found a significant contribution of the (pe) interaction to the momentum exchange. Figure 6 shows momentum images for 1 MeV p impact to be compared with figures 3 and . Contrary to figure 3 the momenta are projected onto the plane defined by the beam and the scattered projectile. (In figure

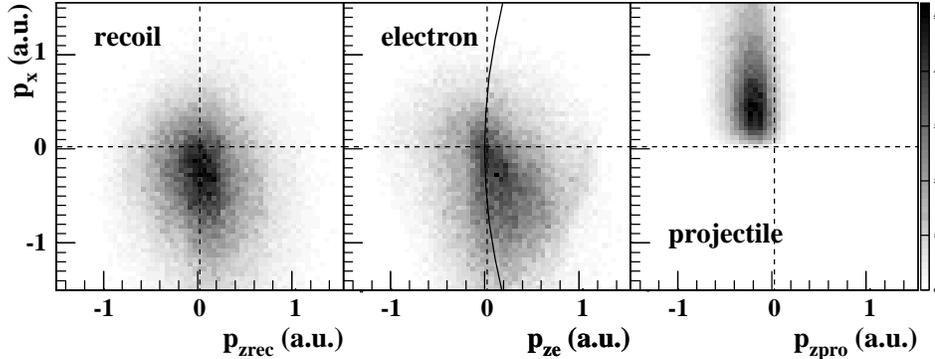


Figure 6: 1 MeV/u  $p + He \rightarrow He^{1+} + e^- + p$ . Projection of the momentum transfer vectors of recoil-ion (upper) electron (middle) and projectile (lower) in the final state onto the plane defined by the projectile beam and the scattered projectile (not by the recoil-ion as figure 4 and 3). The  $+p_z$  axis is parallel to the incoming projectile direction, the  $+p_y$  axis point in the direction of the scattered projectile. The grey scale represents the corresponding doubly differential cross section  $d^2\sigma/(dp_x dp_z)$  on linear scale. The circular arc in the middle figure shows the location of the binary encounter ridge for electrons (from [39]).

4 the plane of the scattered projectile and recoil-ion coincide). This shows that for such fast p collision the projectile is deflected at least partly at the electron leading to electron emission opposite to the projectile.

In summary we have discussed the power of the COLTRIMS technique to provide detailed inside in the physical mechanism of the interaction of ionizing radiation with atoms. Multi particle momentum space imaging for which COLTRIMS is one of the key technologies have already been used for a variety of studies of the dynamics of many particle reactions. The unprecedented resolution and completeness of many of those investigations allowed to resolve some long standing puzzles in atomic collision physics but at the same time raised even more fundamental questions. Similar impact of such imaging techniques can be expected for the future for other fields in physics, chemistry and related areas.

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