Imaging Spectroscopy for Ions and Electrons

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Abstract

Novel imaging techniques allow to determine in coincidence the vector momenta of ions and electrons from ionizing interactions of photons and charged particles with atoms and molecules. These devices combine 4 π solid angle with high resolution in momentum space. They deliver multidimensional images of the multi particle breakup processes. In many cases these fully differential cross sections unveil the physical processes dominating the reaction. We discuss the application of this technique to studies of double photoionization of He by linear and circular polarized light.

Introduction

The investigation of few-particle transitions in atoms or molecules induced by photons or charged particles are a fascinating testground for our understanding of many-body dynamics in quantum mechanics. Such dynamics of Coulomb systems is the governing factor for much of the structure and evolution in our everyday world. Atomic and molecular many-particle reactions are characterized by fully differential cross sections (FDCS), i.e. cross sections differential in all observables of the final state. In an ionization process this typically corresponds to the vector momenta, spins and internal excitation of all reaction products. Such FDCS provide the most stringent test for theory. Any integration over observables often masks important characteristics of the process. In turn, experimental FDCS in the best case directly unveil mechanisms of the many-particle transition. Tremendous progress in measuring such FDCS have been made in the field of (e,2e) collisions (see

Experimental technique

The basic principles for high resolution 4π spectrometers are identical for ion and electron detection. They are based on a small reaction volume (typically below 1 mm^3) from which the fragments are guided by electric and magnetic fields to large area position-sensitive detectors. The momenta of electron and ion can then be calculated from the time-of-flight and the position where the particles hit the detectors. The ion momenta resulting from atomic reactions are typically in the range of a few atomic units (a.u.), their energies in the μeV - meV regime. This is comparable or even smaller than the thermal motion of the atoms at room temperature (4.6 a.u. for He). Thus, one has to provide an internally cold atomic target for the collision. This is presently achieved by using supersonic gas jet targets. A further improvement in resolution is envisaged by the future use of laser cooled targets [15].

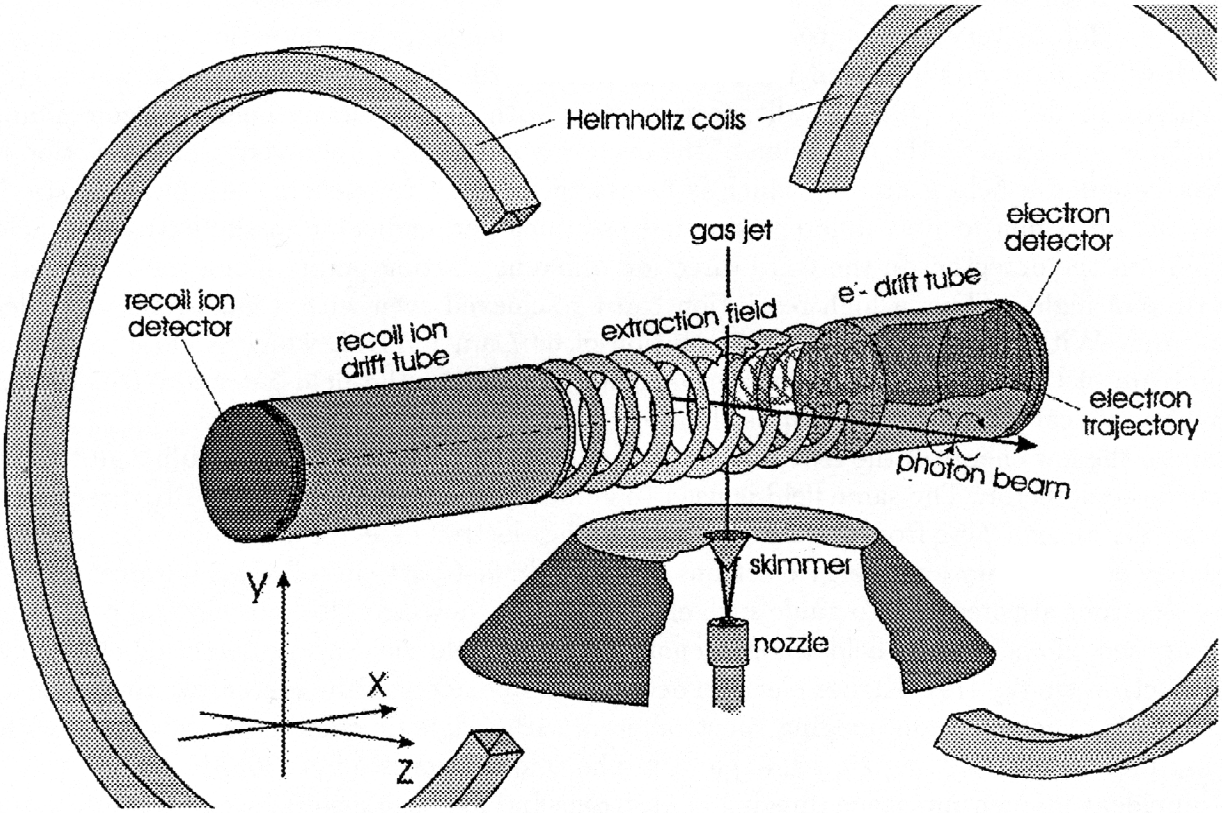

Figure 2: Typical COLTRIMS setup. The gas nozzle is cooled to 15-30 K, the super sonic gas jet has a diameter of 1.1 mm at the intersection with the photon (or charged particle) beam. The electron detector is located on the right side of the spectrometer and the ion detector on the left side. A set of Helmholtzcoils provide a homogeneous field for guiding the electrons towards the electron detectors (from [16])

single photon. This process is a detailed probe of the effects of dynamic electron correlation, one of the hottest topics in todays atomic collision physics. The most simple and widely studied quantity for this reaction is the ratio of total cross sections for double to single ionization (see e.g. the recent experimental papers [40, 13, 41, 42, 43]).

To illuminate how one photon can couple to two electrons, it is instructive to compare the final state momentum distributions of the He^{2+} ions and the electrons created by photoabsorption. Fully equivalent to the case of single ionization shown in figure 1a , figure 3 shows the momentum distribution of the ions and electrons for double ionization by 80 eV photons, 1 eV above the He double ionization threshold. Since the two electrons in the continuum can share the excess photon energy and emerge with various relative angles, the ion momenta are no longer restricted to spheres in momentum space. The maximum ion momentum at a given photon energy is

$$k_{ion} = 2\sqrt{2(E_\gamma - E_{bind})} \tag{1}$$

where E_{bind} is the sum of the ionization potentials for both electrons. This momentum, which is indicated by the outer circle, corresponds to the situation where both electrons escape with equal energy in the same direction. As already shown in [4] this is inhibited by the electron electron repulsion, thus the cross section falls to zero towards the circular line. The cross section has also a node at ion momentum zero. This corresponds to the case of both electrons emerging with equal energy in opposite directions. As has been shown by several authors this is prohibited by a selection rule [4, 44, 45].

For comparison with the ionic momentum distributions we have displayed the electronic distribution in cartesian momentum coordinates for 1 eV excess energy (3a). The striking difference between the electronic and ionic distributions invites speculation on the mechanisms of photo double ionization. The photon acts upon a charge dipole in the atom. This dipole might be thought of as consisting of the positive ion on one pole and either the center of charge of the electron pair or one of the electrons on the other pole. In either case the first step of the absorbtion of the photon will imprint the dipolar characteristics of the linear polarized photon on the distribution of the fragments of a charge dipole. The experiment indicates that the momentum distribution of the nucleus shows a memory of this absorbtion of the photon, while it is completely smeared out in the electron momentum distribution. If one favours the electron pair as the ion's counterpart in the photon-absorbing charge dipole, the subsequent breakup motion of the electron pair is mainly responsible for the electron distribution. The direction of this breakup given by the electron-pair relative momentum $\mathbf{k}_R = \frac{1}{2}(\mathbf{k}_1 - \mathbf{k}_2)$ has been found for 1 eV excess energy to be mainly perpendicular to the photon polarization axis. For additional discussion see [46, 47]. The picture of such a collective motion of the electron pair is most plausible close to threshold. At higher photon energy it seems more appropriate to think of a (single-electron + ion dipole) absorbing the photon. Of course, then electron correlation is indispensable to double ionization. One concludes that it is this electron-electron interaction which smears out the observed recoil-ion dipole pattern . This point of view is in qualitative agreement with the model of Samson [48, 49] which views photo double ionization as photoabsorption by one electron followed by internal electron impact ionization.

An overview of the three-body continuum in the momenta of the two electrons is given in figure 4. It shows the momentum of one electron with respect to the other at 1 and 20 eV above the double ionization threshold. All three particles are necessarily in one plane (following from momentum conservation). This internal plane of the breakup has some orientation to the electric field vector ϵ of the linear polarized photon beam.

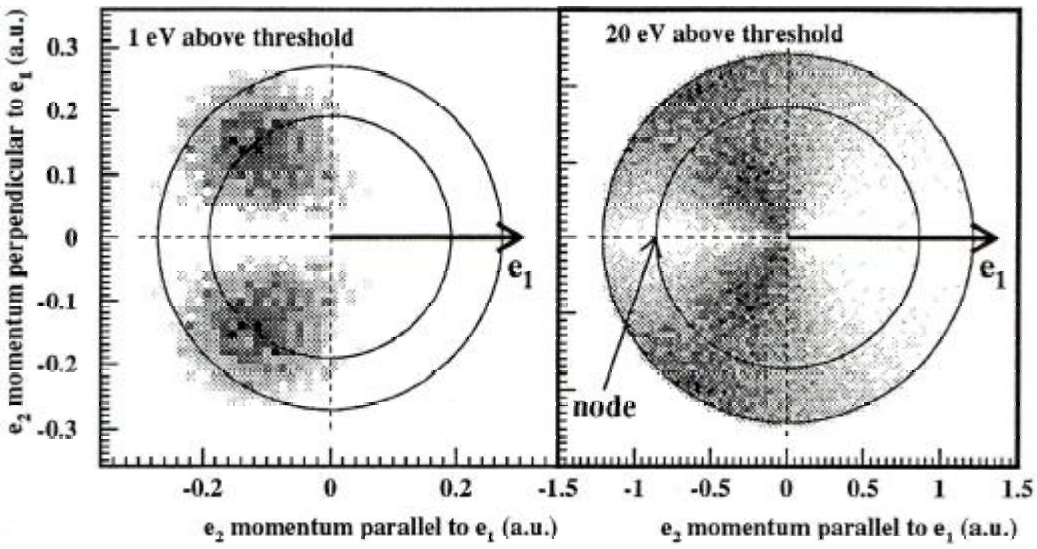

Figure 4: Photo double ionization of He at 1 and 20 eV above threshold by linear polarized light. Shown is the momentum distribution of electron 2 for fixed direction of electron 1 as indicated. The plane of the figure is the internal momentum plane of the three particles. The data are integrated over all orientations of the polarization axis with respect to this plane. The figure thus samples the full cross sections, all angular and energy distributions of the fragments. The outer circle corresponds to the maximum possible electron momentum, the inner one to the case of equal energy sharing. (see text for details)

emission is suppressed at all energy sharings, even so this is not a strict selection rule (see also [9]). At 20 eV the node is really centered at $\mathbf{k_1} = -\mathbf{k_2}$ (indicated by the arrow). This presentation shows strikingly that this node is internal to the three-body system and has nothing to do with ϵ, since the data are integrated over all orientations of ϵ.

An interesting twist is added to this three-body breakup if one introduces a chirality in the initial state by inducing the transition with circular instead of linear polarized light. The question arises how or if at all the chirality of the photon is transferred to the three-body continuum. It has been first pointed out by Berakdar and Klar [51] that such an effect, termed dichroism, might exist even for He double ionization. Viefhaus and coworkers [11] found the first experimental evidence for this effect. The two electrons and the photon axis can span a tripod which might have a handedness if its two legs defined by the electron momenta are distinguishable, i.e. the electrons have unequal energy. This shows up strongest if the three body plane (as it is shown in figure 4) is held fix perpendicular to the photon axis. At 20 eV above threshold, figure 5 shows the momentum distribution of the ion and electron 2 in this plane. The momentum of electron 1,which is chosen to be the faster one, is fixed along the horizontal axis.

Comparison with figure 4 as well as between left and right circular polarized light visualizes that dichroism is a huge effect in this system. While for linear polarized light upper and lower half of the figure are necessarily symmetric, this symmetry is broken for circular polarized light. A detailed

Momentum space imaging provides new spectacular views on many body breakup of coulombic systems. It combines high resolution in momentum space (typically $<< 0.1 a.u.$) with 4π solid angle for all fragments. In many cases such kinematical complete pictures directly 'display' the processes responsible for the breakup of the atom or molecule. Thus some long standing puzzles in atomic collision physics were solved recently using this approach and many new questions and challenges to theory were raised. A great amount of important results have been obtained in particular for ion atom collisions. Since they are not subject of this overview we only name a few of these: Momentum space of single and double ionization by relativistic heavy ions showed the explosion of the atom in the light of an attosecond pulse of virtual photons, stronger than any available laser [30, 29, 27]. A scattering of two electrons inside an atom in a transfer ionization process could be directly seen [58]. Two-center electron-electron interaction has been separated experimentally from nuclear-electron scattering [59, 24]. At slow collisions the promotion of one electron into the continuum via the saddle point mechanism has been imaged [60, 61, 62]. Besides this rapidly increasing progress in atomic and molecular physics such imaging techniques can be expected to be particularly useful in material research and solid state physics in the future. On can envision even the imaging of the many electron emission from surfaces which will give insight in the correlated electron motion in solids.

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