Double and Single Ionization of He and Other Targets Studied Using Cold Target Recoil Momentum Spectroscopy

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Double ionization of an atom by a single photon is the simplest and most fundamental many-electron process. The ejection of two electrons following the absorption of one photon is strictly prohibited in an independent electron approximation. Thus determining the probability of double photoionization alone is already a challenging test of our understanding of electron-electron correlation. Furthermore, in the slow breakup of a bound system into three charged particles, the final state wave function must represent a high degree of few-body Coulomb correlation involving the simultaneous interaction of all three particles. The case of double photoionization is again particularly well suited to study this problem as the energy and the angular momentum delivered to the system can be very well controlled.

Helium, as the most basic three body system, has been the target of extensive studies over the past decades. The purpose of this project has been to study double and single ionization using cold target recoil ion momentum spectroscopy (COLTRIMS). This technique has been widely applied within the area of ion-atom collisions to study the dynamics of energy and momentum transfer in collisions between few-electron systems [1], and the entire technical machinery has been transferred to photon-atom collisions [2]. The technique uses space- and time-imaging of He^+ and He^{++} recoil ions created in photon-He collisions to measure the full momentum vector of each ion produced. Event-mode recording is used and a solid angle of nearly 4π is realized, allowing an extremely high data-collection efficiency. In order to reduce the initial momentum spread of the He target a precooled supersonic He jet is used.

For the case of single ionization of He, determination of the He^+ recoil momentum is equivalent to determination of the photoelectron momentum vector. Fig. 1 shows an image of a typical momentum distribution obtained at a photon energy of 80 eV. When double ionization occurs, the channel is uniquely identified by the production of a He^{++} ion but a full determination of the final state of the system requires that in addition to the ion mnomentum the momentum vector of one of the photoelectrons also be determined. This is accomplished using a second imaging detector opposite the ion detector which performs the complimentary momentum vector measurement of one electron. The experiment is run in two-bunch mode, requiring a timing pulse from the beam clock to measure the electron and ion flight times.

Two major projects have been completed to date and published in refs. [3] and [4].

Summaries are given here.

Ratio of Single to Double ionization of He between 80 and 400 eV

The ratio of double to single ionization ($R=\sigma^{++}/\sigma^{+}$) for He has been studied extensively in recent years (for a review, see [5]). In spite of this intense work, the various data sets reported over the last 30 years from threshold (79 eV) to a few hundred eV above are in substantial disagreement. We have used COLTRIMS to remeasure this ratio for photons between 85 and 400 eV. The technique allows the elimination of all known systematic errors discussed previously in the literature on this subject. A detailed discussion about systematic errors can be found in [3]. The results are shown in fig. 2, where they are compared to previous experimental work and to current theoretical calculations (refs. 9–12 in [3]). Our results are about 25% lower than most of the earlier data (refs. 4–7, 13–18 and 23 in [3]), but are in excellent agreement with the most recent calculations.

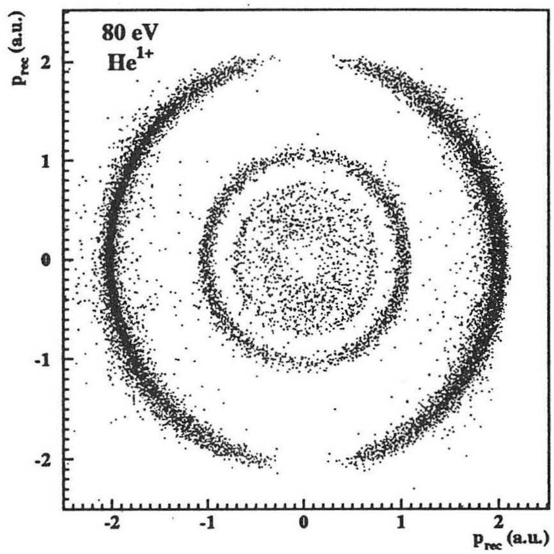

Figure 1: Momentum distribution of He^{+} ions produced by 80 eV photons. The X axis is the direction of the electric field vector of the linearly polarized light. The Y axis is the direction of the gas jet. The data are integrated over a momentum range of ± 0.3 a.u. in the direction of the photon beam, the Z axis.

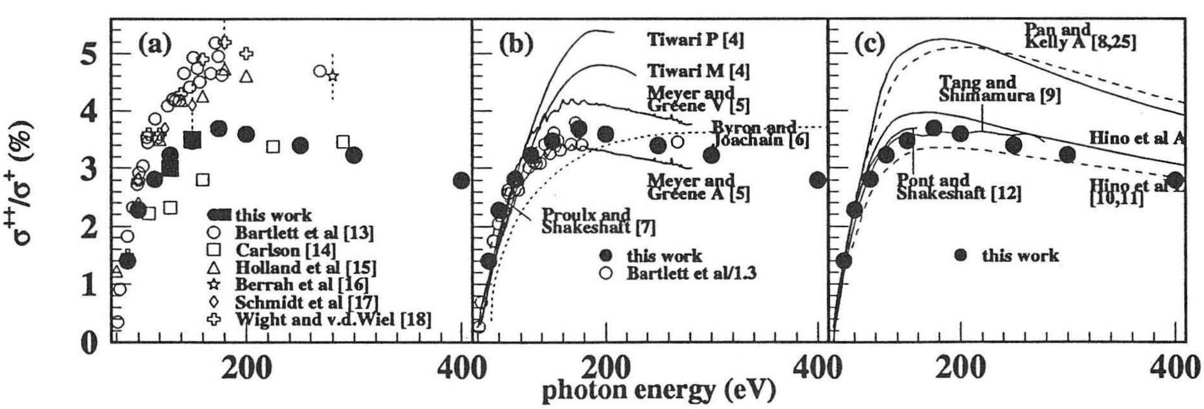

Figure 2: R as a function of photon energy. Full circles: this work (ALS). The open circles in (b) are the same data as in (a) but scaled down by 1.3. V, A, L stand for results obtained in the velocity, acceleration or length form, respectively. The references for the theories and all the older experiments can be found in [3].

Fully differential cross sections for double photoionization of He measured be recoil ion momentum spectroscopy
Several previous experiments have reported differential cross sections for coplanar geometry and for selected cases of equal and unequal energy sharing (refs. 1–6 in [4]). In our work we have for the first time determined fully differential cross sections for double ionization of He near threshold. Here we understand the word fully to mean that, apart from electron spins, the final state of the system is completely kinematically determined and we sample the complete momentum space (4π solid angle) for the ion and one of the electrons. This means that the final momenta of all three particles are determined for *every double ionization event*, with no necessity to choose a priori a particular angle or energy for either electron. Thus the entire final five-dimensional momentum space of the escaping three particles is sampled without prejudice, and the physical process itself determines which parts of this space are the most important.

The results (fig. 3) show that, at an energy of only 1 eV above threshold, the "collective" behavior of the outgoing system is evident in the correlated momenta of the outgoing electrons. The left and right parts of the figure show the data in terms of the Jacobi coordinates $\mathbf{k_r} = \mathbf{k_1} + \mathbf{k_2}$ and $\mathbf{k_R} = \frac{1}{2}(\mathbf{k_1} - \mathbf{k_2})$, where $\mathbf{k_1}$ and $\mathbf{k_2}$ are the momentum vectors of the two electrons. The left figure shows that the recoil ion momentum, the negative of $\mathbf{k_r}$, has a dipole-like distribution, reflecting the dipole-dominated interaction operator. Departure from a pure dipole shape is caused by the absorption of angular momentum into the internal motion of the electron pair, the $\mathbf{k_R}$ motion. The right figure shows the corresponding $\mathbf{k_R}$ distribution. We emphasize that this shows the correlation between the motion of the two electrons, not the motion of the individual electrons. The data reveal that the electrons are preferentially emitted in a direction perpendicular to the recoil momentum. This can be intuitively explained as the result of the electrons trying to move in such a way that neither falls into the He^+ potential, thereby producing single rather than double ionization. These data were found to be in good agreement with a fourth-order Wannier calculation and give an experimental basis for the choice of quantum numbers of the Wannier state populated in the photoionization. In our most recent experiment we obtained fully differential cross sections at 20 eV above threshold. These data show that the Wannier description is still in good agreement with the experiment at this energy. Further detail is given in the publication.

In the future we plan to probe the capabilities of COLTRIMS for the study of other systems. Initial projects include:

1) *double photoionization of Ne* very near the double ionization threshold (62.5–64.5 eV photon energy). In this energy range only the ground state of the Ne^{++} ion can be populated, so the situation is similar to that in He. (Three J values can be populated, however). We wish to investigate to what extent the Wannier patterns observed for He remain for a system in which two of the three potentials are no longer pure Coulomb in nature.

2) *fully differential cross sections for the three body break up of* $H_2 \rightarrow p + e^- + H$ close to threshold. Our experimental technique allows us to measure the energy and angular distribution of the photoelectron with respect to the alignment of the molecular axis with 4π solid angle, delivering the most complete picture of the physical process possible. Such fully differential cross sections will show the details of the electronic motion from the bound to the

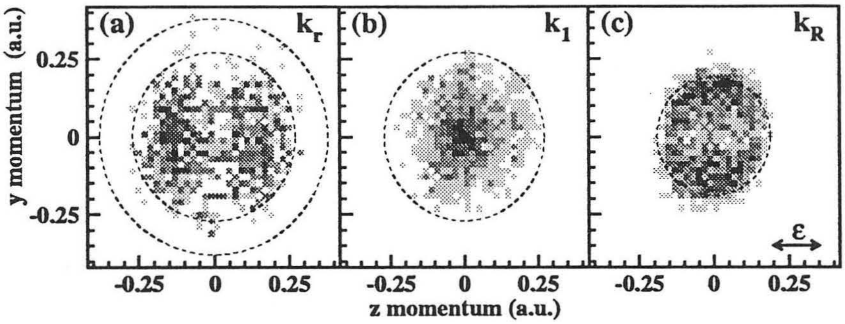

Figure 3: Density plots of projections of the momentum spectra from double ionization of He by 80.1 eV photons. The z and y components of the momentum are plotted on the horizontal and vertical axes, respectively, the polarization vector of the photon is in the z direction and the photon propagates in the x direction. Only events with -0.1< k_{rx} <0.1 are projected onto the plane. (a) The recoil (or $-\mathbf{k_r}$) momentum distribution. The outer circle indicates the maximum calculated recoil momentum, and the inner circle is the locus of events for which the $\mathbf{k_r}$ motion has half of the excess energy. (b) The distribution of single electron momenta ($\mathbf{k_1}$ or $\mathbf{k_2}$). The circle locates the momentum of an electron which carries the full excess energy. (c) The relative electron momentum (or $\mathbf{k_R}$) distribution. The circle identifies the maximum possible value for $\mathbf{k_R}$.

continuum state in the aligned molecular environment like the secondary scattering of the electron at the nuclei of the molecule. Our very high resolution for the ion momentum measurement allows to see the effects of the electron recoil on the nucleus. This will identify from which of the two centers the electron has originally been emitted.

3) *ratio of single to double ionization probabilities for Mg.* This is an extension of the He results discussed above to other systems. No other analogous two-electron system has been studied. We choose Mg because of the convenience of constructing an appropriate oven for producing the atomic beam target. We give up the very high momentum resolution in the direction of motion of the target beam which we have with the cold supersonic He jet, but believe the basic COLTRIMS geometry will remain useful, allowing control of such problems as scattered light, secondary processes, etc. in the same way as for He.

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