# Impulsive Electron-Impact Double Ionization and the Two-Electron Momentum Density

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Cross sections for electron-impact double ionization of magnesium have been measured in a triple coincidence experiment. When the momentum transfer is large, the double-ionization cross section as a function of the angle of the motion of the center of mass of the two ejected electrons closely resembles the analogous variation of the single-ionization cross section when the electron is ejected in a quasielastic binary collision. Assuming that the similarity in the shape of the cross section implies a similarity in the high-momentum-transfer ionization mechanism, atomic two-electron momentum densities have been derived from the data. Two-electron densities provide information on electron correlation in the atom.

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Electron-impact single ionization is employed to determine the momentum density of atomic and molecular electrons [1]. The purpose of the present research is to use electron-impact double ionization to obtain electron-pair momentum densities. Measured single-electron momentum densities are useful in assigning orbital symmetries and in distinguishing between alternative theoretical wave functions. The distribution of the two-electron density is the physical observable that most directly reflects electron correlation. The single-electron momentum-density research was based on detailed investigation of the kinematics of ionization that resulted in the establishment of the experimental conditions for which the target-electron momentum could be directly related to the measured cross section [2–4]. What is sought here are the conditions for which the double-ionization cross section can be related to the two-electron momentum density.

The angular distribution of ejected electrons from electron-impact single ionization is characteristic of the momentum transfer in the ionizing collision if the impact energy is more than a few times the ionization energy [5]. When the momentum transfer, $K$, is greater than 1 a.u., the angular distribution approaches that of hard-sphere collisions with ionized electrons preferentially ejected in the momentum-transfer direction. Ejected electrons also appear in the backward (antimomentum-transfer direction) as a consequence of backscattering from the residual ion core. These high-momentum-transfer conditions are very much different from low-momentum-transfer electron-impact ionization where the angular distribution of electrons from either single or double ionization is similar to that obtained from photoionization [5–7].

For high-momentum-transfer single ionization, a polar plot of the cross section as a function of the ejection angle relative to the incident-electron direction (an Ehrhardt plot [5]) shows a lobe centered on the momentum transfer direction. Impulsive electron-electron collisions with the ion core participating only as a spectator account for the electrons ejected into this lobe. The shape of the lobe reflects the target-electron momentum distribution at the instant of collision. The realization of an analogous, impulsive, high-momentum-transfer, double-ionization regime from which two-electron momentum densities can be derived is the subject of this paper.

The experiments described here are completely (eightfold) differential cross section measurements of electron-impact double ionization—so-called $(e, 3e)$ measurements. The target is magnesium, a pseudo-two-electron atom with two $3s$ electrons outside a closed-shell doubly charged core. It is the direct ionization of the $3s$ electrons that is being studied. A triple-coincidence measurement is carried out to measure, from each double ionization event, the ejected-electron momenta, $\mathbf{p}_1$ and $\mathbf{p}_2$, and the incident and scattered electron momenta, $\mathbf{p}_0$ and $\mathbf{p}_s$, from which the momentum transfer, $\mathbf{K} = \mathbf{p}_0 - \mathbf{p}_s$, is calculated along with the center-of-mass momentum of the two ejected electrons, $\mathbf{P} = \mathbf{p}_1 + \mathbf{p}_2$. To obtain momentum-transfer values in excess of 1 a.u., the kinematics are restricted to relatively high incident-electron energies and large scattering angles. In our experiments the incident energy is in the range 732 to 1032 eV and the scattering angle $\theta_s$ is 18° giving values of $|\mathbf{K}|$ in the range 2.3 to 2.7 a.u. The energies of the ejected electrons are equal: $E_1 = E_2 = 55$ eV, well in excess of the 22 eV double ionization potential of magnesium. These ejected-electron energies correspond to momenta of magnitude $|\mathbf{p}_1| = |\mathbf{p}_2| = 2.0$ a.u.

In double ionization, the magnitude of the ejected electrons' center-of-mass momentum is a function of the angle, $\omega_{12}$, between $\mathbf{p}_1$ and $\mathbf{p}_2$. We observe events

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for which this angle falls in the range $91° < \omega_{12} < 170°$. The corresponding magnitude of the center-of-mass momentum extends from $|\mathbf{P}| = 0.37$ a.u. to $|\mathbf{P}| = 2.82$ a.u., a value somewhat greater than the magnitude of the momentum transfer. Our apparatus permits a three-dimensional observation of ejected and scattered electrons. For the present experiments, the ejected-electron detectors, although arranged in a 3D array, are positioned so that $\mathbf{p}_0$, $\mathbf{p}_s$, $\mathbf{K}$, and $\mathbf{P}$ lie in a common plane. We are thus able to display the measured cross sections as a function of the angle of orientation of $\mathbf{P}$ in polar plots that are analogous to the Ehrhardt plots of single ionization.

The $(e, 3e)$ cross section is small in comparison to single ionization cross sections and decreases rapidly with increasing incident energy and with increasing momentum transfer. Moreover, the statistical exigencies of triple-coincidence measurements place severe limits on the incident-beam current and target density required for optimum signal-to-noise. Of necessity, $(e, 3e)$ spectrometers are multiplexed devices with arrays of detectors so that many triple-coincidence measurements can be carried out at the same time [8,9]. The virtue of making many simultaneous measurements is quite evident in the present experiments involving high incident-electron energies and large momentum transfers—the triple-coincidence rate for each set of three detectors is a few tens of events per week.

Our $(e, 3e)$ apparatus has been described in detail elsewhere [9,10]; the electron optics and the magnesium-vapor source will not be specifically discussed, however, it is important to understand the detector arrangement employed in these experiments. A vector diagram of the $(e, 3e)$ experiment is shown in Fig. 1 and superimposed on it are the apertures of the scattered-electron detector and ejected-electron detectors. The scattered-electron detector aperture is a segment of an annular aperture with a polar angle width $\Delta\theta_s = \pm 2°$ and an azimuthal extent $\Delta\phi_s = \pm 20°$. Sixteen ejected-electron detectors are positioned at 45° to a line perpendicular to the incident-electron direction with $\pm 7°$ apertures. 64 coincidence measurements are performed simultaneously, each corresponding to a triple coincidence between an electron arriving at the scattered-electron detector, an ejected electron arriving at one of the eight "upper" ejected-electron detectors, and a second ejected electron arriving at one of the eight "lower" ejected-electron detectors. The incident- and scattered-electron momentum vectors define a horizontal scattering plane as shown in the figure. The momentum transfer vector $\mathbf{K}$ lies in this plane. With the ejected-electron detectors arranged as shown, and choosing equal energies for the ejected electrons, the ejected-electron center-of-mass momentum vector $\mathbf{P}$ lies in the plane of $\mathbf{p}_0$, $\mathbf{p}_s$, and $\mathbf{K}$.

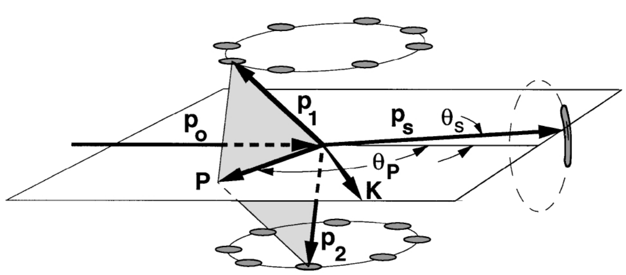


FIG. 1. A vector diagram of the $(e, 3e)$ experiment superimposed upon a diagram showing the location and effective apertures of the scattered-electron detector and the array of ejected-electron detectors. $\mathbf{p}_0$ and $\mathbf{p}_s$ are the momentum vectors of the projectile electron before and after scattering, $\mathbf{p}_1$ and $\mathbf{p}_2$ are the momentum vectors of the ejected electrons, $\mathbf{P}$ represents the ejected electrons' center-of-mass momentum, and $\mathbf{K}$ the momentum transfer.

Cross section data for the double ionization of the outer two $3s$ electrons of magnesium by electron impact at an incident electron energy of 1032 eV are shown in Fig. 2. The cross sections are measured for eight values of $|\mathbf{P}|$. These data are binned in four groups with $|\mathbf{P}| =$ (2.75 to 2.82 a.u.), (2.26 to 2.46 a.u.), (1.42 to 1.73 a.u.), and (0.37 to 0.74 a.u.) and presented as polar plots of cross section as a function of $\theta_P$, the angle of the ejected-electron center-of-mass momentum vector, $\mathbf{P}$, with respect to the incident electron direction, $\mathbf{p}_0$. The direction and magnitude of the scattered-electron momentum and the momentum transfer are indicated. When the magnitude of $\mathbf{P}$ is similar to that of the momentum transfer, $\mathbf{P}$ tends to be oriented in the momentum-transfer direction. In the top panels a lobe centered on the momentum transfer direction appears that is similar in appearance to that seen for single ionization in the impulsive regime. For small values of $|\mathbf{P}|$, the cross sections are essentially isotropic in $\theta_P$.

If no momentum is transferred to the ionic core by the collision, the initial momentum of the target electrons can be determined from a measurement of their momentum following knockout from the atom. This can be demonstrated in a simple accounting for momentum conservation in a (double) ionizing collision.

The total momentum before the collision is equal to that following the collision:

$$\mathbf{p}_0 + \mathbf{p}^i_{\mathrm{ion}} + \mathbf{q}_1 + \mathbf{q}_2 = \mathbf{p}_s + \mathbf{p}_1 + \mathbf{p}_2 + \mathbf{p}^f_{\mathrm{ion}}, \quad (1)$$

where $\mathbf{q}_1$ and $\mathbf{q}_2$ are the momenta of the target electrons before the collision, $\mathbf{p}^i_{\mathrm{ion}}$ is the momentum of the doubly charged atomic core before the collision, and $\mathbf{p}^f_{\mathrm{ion}}$ is the momentum of the core after the collision. If the target atom is stationary, $\mathbf{p}^i_{\mathrm{ion}} + \mathbf{q}_1 + \mathbf{q}_2 = 0$, or $\mathbf{q}_1 + \mathbf{q}_2 = -\mathbf{p}^i_{\mathrm{ion}}$. In other words, $\mathbf{p}^i_{\mathrm{ion}}$ is the momentum with which the residual-ion core would recoil following the removal of two electrons in a collision in which the ion is a spectator. The difference between the final and initial core momenta, $\mathbf{p}^f_{\mathrm{ion}} - \mathbf{p}^i_{\mathrm{ion}}$, is the momentum imparted to the core by the collision.

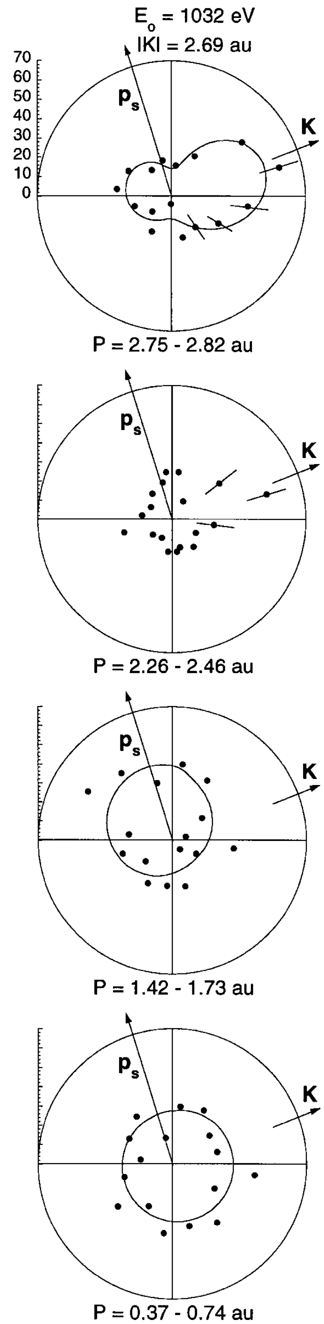


FIG. 2. Polar plots of relative cross section as a function of the angle, $\theta_P$, of the center-of-mass momentum of the two ejected electrons from the double ionization of the $3s$ electrons of magnesium by electron impact at an incident-electron energy of 1032 eV with momentum transfer of 2.69 a.u. From top to bottom are shown data for which the magnitude of **P** falls in the range (2.75 to 2.82 a.u.), (2.26 to 2.46 a.u.), (1.42 to 1.73 a.u.), and (0.37 to 0.74 a.u.). The incident-electron direction is vertically upward. The scattered-electron momentum vector, $\mathbf{p}_s$, and the momentum-transfer vector, **K**, are shown to scale on each plot. The radial scale is the same on all plots and corresponds to the true triple coincidence rate per week (over a background of accidental coincidences). Error bars represent statistical uncertainty. The curves shown are polynomial fits to the data intended only to guide the eye.

For a stationary target atom, the momentum transfer is

$$\mathbf{K} = \mathbf{p}_0 - \mathbf{p}_s = \mathbf{p}_1 + \mathbf{p}_2 + \mathbf{p}_{\mathrm{ion}}^f = \mathbf{P} + \mathbf{p}_{\mathrm{ion}}^f . \quad (2)$$

Since **K** and **P** are measured in the experiment, the residual ion momentum can be determined:

$$\mathbf{p}_{\mathrm{ion}}^f = \mathbf{K} - \mathbf{P} . \quad (3)$$

For collisions where the ion is a spectator and no momentum is imparted to the core by the incident electron,

$$\mathbf{p}_{\mathrm{ion}}^f = \mathbf{p}_{\mathrm{ion}}^i = -(\mathbf{q}_1 + \mathbf{q}_2) = -\mathbf{q} = \mathbf{K} - \mathbf{P} . \quad (4)$$

In this case, **q**, the net momentum of the target pair in the atom at the moment of collision, can be determined from the measurement of **K** and **P**.

While single-electron momentum densities have been extensively studied in $(e, 2e)$ experiments, only a few theoretical studies of electron pair momentum densities have been carried out [11,12] and there have been no comparable experiments. The single-electron momentum density is given directly by the impulsive $(e, 2e)$ cross section as a function of the target electron momentum (or, equivalently, the ion recoil momentum). The analogous $(e, 3e)$ measurement of the two-electron density requires the cross section be measured as a function of variables that depend jointly on the coordinates of two electrons. In momentum space, the coordinates for the two-electron problem have been chosen to be the relative momentum, $\mathbf{Q} \equiv \mathbf{q}_1 - \mathbf{q}_2$, and the net momentum, **q**, defined above. The effect of electron correlation on the calculated two-electron density in each of these coordinates has been clearly demonstrated [11]. The relative and net momentum vectors are the obvious choice of independent variables for the experimental study of two-electron momentum densities and, as demonstrated above, it is a straightforward matter to determine **q** in the requisite impulsive $(e, 3e)$ measurement.

From a kinematic point of view, our experiments are analogous to the argon $3s$ and $3p$ single-ionization experiments of Avaldi, McCarthy, and Stefani carried out at a similar incident energy (1 keV) and over a similar range of momentum transfers (1.3 to 3.0 a.u.) [13]. The angular distributions of ejected electrons seen in these experiments display both a forward and backward lobe. The residual ion momentum for ejection in the momentum-transfer direction is just the recoil momentum and is therefore small. The cross section in the forward lobe, plotted against the residual-ion momentum, clearly reflects the momentum distribution of the target electron in the atom. For double ionization with no momentum transfer to the core, the residual-ion momentum can be anticipated to be similarly small—less than about 1 a.u. For example, the measured mean momentum of a single uncorrelated $3s$ electron is less than 0.5 a.u. [14] and hence the mean net momentum of a pair of $3s$ electrons will be less than 1 a.u. The available calculations

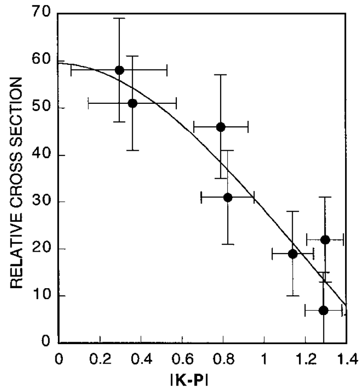


FIG. 3. The cross section for double ionization of the $3s$ electrons of magnesium by 1032-eV electron impact in collisions for which the residual ion momentum, $|\mathbf{p}_{\mathrm{ion}}^{f}| = |\mathbf{K} - \mathbf{P}|$, is relatively small. The vertical error bars represent the statistical uncertainty in the cross section measurement; the horizontal error bars correspond to the finite extent of the detector apertures. The curve shown is polynomial fit intended only to guide the eye.

of two-electron momentum distributions support the same conclusion. We therefore focus on doubly ionizing collisions where $\mathbf{p}_{\mathrm{ion}}^{f}$ is small.

In the top panels of Fig. 2, for the lobes centered on the momentum-transfer direction, **P** and **K** are of similar magnitude and oriented in roughly the same direction, $\mathbf{p}_{\mathrm{ion}}^{f}$ is small, and the condition of small momentum transfer to the core is a reasonable assumption supported by the resemblance of the large-**P** double-ionization scattering pattern to that observed in single ionization under similar conditions of high momentum transfer with the ion as a spectator. By contrast, large values of $\mathbf{p}_{\mathrm{ion}}^{f}$ correspond to **P** opposite to **K** (backscattering), and to small values of $|\mathbf{P}|$ ($\mathbf{p}_1$ opposite $\mathbf{p}_2$). In Fig. 3 we have plotted the 1032-eV double-ionization cross section as a function of $|\mathbf{K} - \mathbf{P}|$ for values of $|\mathbf{K} - \mathbf{P}|$ less than 1.4 a.u. If one concludes that double ionization contributing to these data is the result of impulsive collisions with no involvement of the ion, then the variation of the cross section with $|\mathbf{K} - \mathbf{P}|$ is a measure of the two-electron momentum distribution of the magnesium $3s$ electrons at the instant of knockout.

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