Electron-Impact Double Ionization of Argon Studied by Double and Triple Coincidence Techniques: The First (e,3e) Experiment

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Angular correlations between three outgoing electrons in the double ionization of argon by electron impact have been measured for the first time. Double and triple coincidence techniques are used to determine simultaneously all final electron energies and angles, and provide fourfold- and fivefold-differential cross sections, respectively. The angular distributions are consistent with a two-step model for the double-ionization mechanism. The present results clearly show the feasibility and potentiality of $(e,3e)$ experiments.

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The study of ionization by particle impact is of fundamental importance to many fields of physics. Considerable knowledge of the single-ionization process of atoms and molecules has been gained using a wide variety of projectiles. On the other hand, studies of double-ionization processes are by far less abundant in the literature, because of the smallness of the corresponding sections. Almost all investigators measured *integrated* cross sections as a function of the incoming particle energy;[1-6] *differential* cross sections with respect to the energies and/or solid angles of emission of the final particles[7-10] were very seldom reported.

The present work was motivated by several unanswered questions: (a) What is the energy partitioning between the two "atomic" electrons escaping into the continuum? (b) Does this energy partitioning depend on the respective directions of emission? (c) What is the angular distribution of the electrons as well as their angular correlation? (d) How do the above energy and angular distributions vary with the amount of momentum transferred to the target? (e) What is the main process responsible for outer-shell double ionization under given kinematical conditions (i.e., is it a direct double-ionization process or does it proceed via correlation between the electrons)?

Clearly, many problems in the field of electron-impact double ionization are widely open.

In order to tackle these problems, we have undertaken electron-impact double-ionization experiments that completely determine the kinematics: *All three final electrons are simultaneously analyzed both in direction and in energy and are detected in coincidence.* These are the so-called $(e,3e)$ experiments,[11] analog to the pioneering $(e,2e)$ experiments for single ionization[12] (see Ref. 13 for recent reviews). Though triple coincidence experiments have been recently[14] reported, to our knowledge, *full*-differential triple coincidence experiments have never been reported so far in atomic physics.

The process under investigation obeys the energy- and momentum-conservation equations $E_0 = E_a + E_b + E_c + E_i^{2+}$ and $\mathbf{k}_0 = \mathbf{k}_a + \mathbf{k}_b + \mathbf{k}_c + \mathbf{q}_r$. Here E_i^{2+} stands for the double-ionization threshold energy and $\mathbf{q}_r$ is the recoil momentum of the Ar^{2+} ion. Electron energies and momenta are E_j and $\mathbf{k}_j$ ($j = 0$, a, b, or c) with index 0 representing the incident electron. Following the terminology used in $(e,2e)$ experiments,[15] the three outgoing electrons, though indistinguishable, are indexed with an a for the fast "scattered" one, while of the two slower electrons the fastest is labeled b and the slowest c. In the experiments reported below, all three electrons are observed in the collision plane defined by $\mathbf{k}_0$ and $\mathbf{k}_a$ at angles θ_a, θ_b, and θ_c with respect to the incident direction. Finally, $\mathbf{K} = \mathbf{k}_0 - \mathbf{k}_a$ stands for the momentum transfer to the target after the collision. The quantity that is measured in the present $(e,3e)$ experiments at a given impact energy is a fivefold-differential cross section, 5DCS, or $d^5\sigma/dE_a\,dE_b\,d\Omega_a\,d\Omega_b\,d\Omega_c$.

The apparatus is a modified version of our previously described crossed-beam $(e,2e)$ spectrometer.[16] The fast scattered electrons are energy selected in a stationary cylindrical analyzer, while twin hemispherical analyzers are used for the two slow ejected electrons. One of them is fixed so that the corresponding ejection angle is 255° with respect to the incident beam direction. The second ejection angle as well as the scattering angle θ_a are varied by rotating the second hemispherical analyzer and the electron gun, respectively, around the gas jet axis.

Previously published total-cross-section data for electron-impact double ionization[3] indicate that at the incident energy of ~ 5.5 keV the ratio σ^{2+}/σ^{+} for Ar is roughly 5%. Therefore, most electrons entering each detector at a given energy correspond to a single ionizing event and thereby generate a high accidental coincidence rate. This signal-to-noise ratio problem, together with the intrinsic smallness of triple coincidence cross sections, led us to perform, in the first instance, a set of measurements standing halfway between $(e,2e)$ and $(e,3e)$ experiments. In these experiments, an arbitrary pair of electrons is detected in coincidence, irrespective of the direction of the third unobserved one. Referring to the energy-conservation law, electron pairs $(e_a e_b)$, $(e_a e_c)$, or $(e_b e_c)$ were selected corresponding to a double-ionizing event. We, therefore, measured fourfold-differential cross sections, 4DCS, $d^4\sigma/dE_i\,dE_j\,d\Omega_i\,d\Omega_j$, for each $(e_i e_j)$ pair. Yet, due to the low coincidence rates, it was necessary to operate at modest energy and angular resolutions. The overall $(e,2e)$ energy resolution was set to 9 eV, while the acceptance solid angles were $\Delta\Omega_a \sim 5\times10^{-5}$, $\Delta\Omega_b$ and $\Delta\Omega_c \sim 5\times10^{-2}$ sr, respectively. However, the angular information is preserved, as shown by measurements made on well-known angular distributions (Fig. 1).

A sample 4DCS result for the pair $(e_a e_c)$ is shown in Fig. 1. The incident energy ($E_0 = 5623$ eV) is chosen in such a way that the unobserved b electron carries out an energy $E_b = 75$ eV, while the c electron is detected at $E_c = 5$ eV. The modest energy resolution does not allow us to discriminate among various final Ar^{2+}-ion states, mainly the $3p^{-2}\,{}^3P$ ground state ($E_i^{2+} = 43$ eV) and the ${}^1D_2$ and ${}^1S_0$ metastable states with excitation energies of 1.7 and 4.2 eV, but also the $3s^{-1}3p^{-1}$ state whose threshold is $E_i^{2+} = 58$ eV. However, from observations made at low impact energy,[17] we may reasonably expect the Ar^{2+} $3p^{-2}\,{}^3P$ ground state to be the dominant contribution. Moreover, due to the tail of the resolution function, the data in Fig. 1 may also be contaminated by single-ionizing events leading to Ar^+ ions with a $3p^{-1}$ or $3s^{-1}$ hole in their ground or excited (the so-called "satellites") states. The resolution function was carefully measured on helium under identical conditions. From this and known Ar-satellite spectra,[18] we estimated the single-ionization contribution to amount to 10% of the double-ionization signal at $E_c = 5$ eV.

Finally, the 4DCS shown in Fig. 1, hereafter denoted by σ_{ac}^{2+}, have been normalized to an absolute scale by comparison with the corresponding $(e,2e)$ triple-differential cross sections, σ_{ac}^{+}, for Ar-$3p$ single ionization, also shown in Fig. 1. The latter and the former are measured for the same E_c value, under identical conditions except for the incident energy which is adjusted to meet the single- or the double-ionization energy balance. Therefore, both differential cross sections are obtained on the same relative scale. The absolute scale for σ_{ac}^{+} is inferred by reference to previously published works.[15,19] The overall estimated uncertainty on the absolute scale for σ_{ac}^{2+} is about 15%.

Also shown in Fig. 1 are the $\pm\mathbf{K}$ directions, corresponding to single ($\mathbf{K}^+$) or double ($\mathbf{K}^{2+}$) ionization of the target. As is well known, the binary and recoil lobes of the σ_{ac}^{+} distribution are found to peak in the $\pm\mathbf{K}^+$ directions, hence ruling out any angular problem in our experiments. The σ_{ac}^{2+} distribution shows no such marked structure. This different angular behavior first indicates that the σ_{ac}^{2+} data are not appreciably contaminated by single ionization, and second that the momentum-transfer direction has lost its significance as a symmetry axis. This point is discussed in more detail later on, along with the results of Fig. 3.

Let us now turn to the *triple coincidence* measurements. The experimental arrangement used for detecting triple coincidences is essentially based on two identical time-to-amplitude converters (TAC), started by the a-electron pulses and stopped by the b- and c-electron pulses, respectively. The data are displayed in two ways as indicated in Figs. 2(a) and 2(b): either as a three-dimensional histogram of the arrival time coincidences [Fig. 2(a)], or as a false color map of those coincidences [Fig. 2(b)]. Cuts along the t_b and t_c axes are also

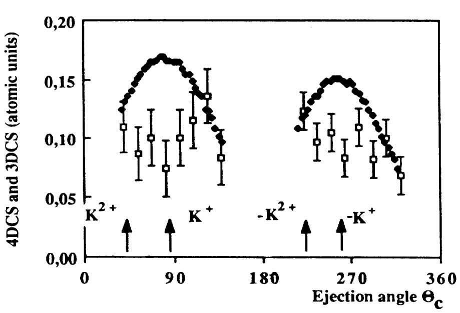

FIG. 1. Absolute fourfold (open symbols) and triple (closed symbols) cross sections for the coincidence detection of the pair $(e_a e_c)$ issued from double and single ionization of Ar $3p$, respectively, plotted against θ_c. $E_a = 5500$ eV, $E_c = 5$ eV, and $\theta_a = 0.55°$. Energy of the unobserved double-ionized b electron is $E_b = 75$ eV. Single-ionization cross sections (i.e., 3DCS) are divided by 1500.

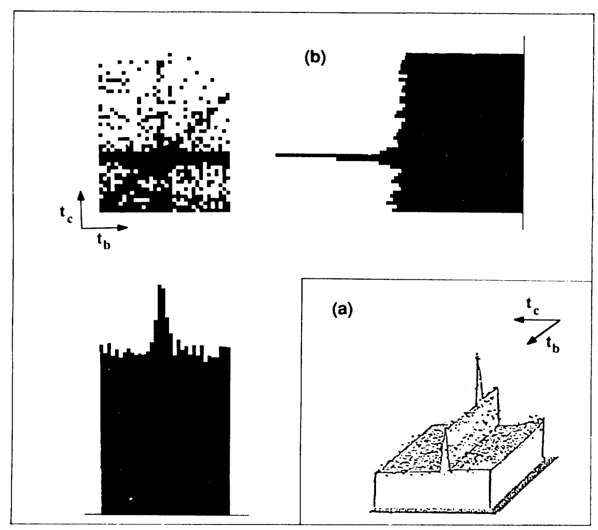

FIG. 2. Time spectrum for triple coincidences. t_b and t_c are the arrival times of the slow b and c electrons with respect to the fast one. (a) 3D perspective view. (b) False color view and cuts along t_b and t_c axes at the position of the triple coincidence peak.

displayed. The accidental coincidences lying at the bottom of the time coincidence peak are due to four different contributions: (a) a fully accidental contribution where the three electrons a, b, and c are uncorrelated, thereby giving rise to counts at any position of the view of Fig. 2(b), (b) three contributions each of which is due to two correlated electrons, the third one being random. This gives rise to so-called "walls" of accidental coincidences superimposed upon the previous fully random contribution (clearly seen in Fig. 2). At the intersection of these three walls stands the triple coincidence peak, superimposed upon the four accidental contributions.

Figure 3 shows the 5DCS for double-electron ejection from Ar at the indicated energies and angles. Because of geometrical constraints, only a limited angular range could be covered. In this range, and within statistical uncertainties, the data show no θ_c angular dependence. In particular, no preference is found for the $\mathbf{K}^{2+}$ direction as it is usually observed for $(e,2e)$ angular distributions for single ionization. This observation is consistent with the results of Fig. 1. If this isotropy of the 5DCS can be assumed to hold in all directions, it would indicate that at the incoming and outgoing considered energies, the three emitted electrons are uncorrelated. Such a result could be interpreted in terms of the dynamics of the double-ionization process, as discussed below.

Double ionization may occur mainly via three mechanisms.[5,20] (1) In the shake-off mechanism the projectile electron interacts only once with a single target electron which is ejected without further interaction with other target electrons. Subsequent electronic relaxation leads to the ejection of a second electron. When this process is described in the sudden approximation[21] (valid for fast incident and ejected electrons) electron correlation plays no role. Actually in our experiments the ejected electrons are not fast (10 and 20 eV) and are both removed from the same $3p$ outer shell; therefore correlations should be important[5] and the observed isotropy would rule out such a shakeoff process. (2) In the two-step mechanism termed TS1, the incident electron interacts with one target electron which subsequently collides with a second electron leading to ejection of the pair. This second process should reflect the correlation between the two atomic electrons that have interacted. For this reason, the observed isotropy also rules out this process. (3) In the two-step mechanism termed TS2, the incident electron collides with two target electrons, resulting in their double ejection. This process where no correlation is expected in the final state is compatible with our observations. Moreover, the *total* momentum $\mathbf{K}^{2+}$ lost *in two independent steps* by the incident electron is no longer an $(e,2e)$-like preferential direction of ejection for any of the b or c electrons.

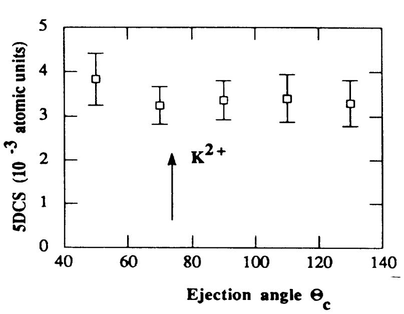

FIG. 3. Absolute fivefold $(e,3e)$ differential cross sections for double ionization of argon vs θ_c. $E_a = 5480$ eV, $E_b = 20$ eV, $E_c = 10$ eV, $\theta_a = 0.45°$, and $\theta_b = 255°$.

However, it must be emphasized that such conclusions cannot be drawn unambiguously. First, because the modest energy resolution does not discriminate among various ion final states; second, because our data are taken over a limited angular range and that the assumption of isotropy might well not be justified. Obviously, the experiments are to be repeated (and are planned to) over an extended angular range and with a better energy resolution. This will then enable us to investigate the influence of each parameter (incident energy, excess energy, energy partitioning, momentum transfer, etc.). For instance, at low impact energies, electron correlations are expected to be particularly important and should result in a large anisotropy of the 5DCS.

Under the hypothesis of isotropy of the 5DCS, the data in Fig. 3 have been made absolute as follows: The integral of the 5DCS over $d\Omega_c$ is simply 4π times this quantity, and must be equal to the corresponding 4DCS, σ_{ab}^{2+}, measured under identical conditions. The latter is in turn normalized as described above. Though the accuracy of the absolute scale strongly depends on the assumed isotropy, we felt it important to determine at least an approximate absolute scale for these first full-differential cross sections.

In conclusion, the present experiments were originally thought of as preliminary ones whose main aim was to demonstrate the feasibility of $(e,3e)$ experiments, to show up all the problems that may arise in these experiments, and to help master this new technique. In this respect we have fully achieved our goal. However, even with the present energy and angular resolutions, the triple coincidence count rate is small, typically 2×10^{-3} counts/s. Increasing the beam current, I_B, or the target gas density, n_G, would result in unacceptable degradation of the signal-to-noise ratio, proportional to $(n_G I_B)^{-2}$. We, therefore, have started building up a new setup including energy and/or angle multidetection of the three outgoing electrons, where about 200 experiments such as the ones described here will be simultaneously performed.

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