Fully Determined (e, $3e$) Experiments for the Double Ionization of Helium

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Angular distributions of the two ejected electrons following electron impact double ionization of helium have been measured using a multicoincidence multiangle $(e, 3e)$ spectrometer at an incident energy ~5.5 keV and equal outgoing energies $E_b = E_c = 10$ eV. The measured distributions are similar to photodouble ionization $(h\nu, 2e)$ ones, but with evidence of additional nondipolar contributions which tend to fill up the characteristic node at the mutual angle $\theta_{bc} = 180°$. They are also in satisfactory agreement with recent $(e, 3e)$ calculations. [S0031-9007(98)07708-4]

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The study of the double ionization (DI) process under photon or charged particle impact has undergone a recent revival due to the spectacular advances in coincidence detection techniques. In these measurements the energy transfer to the target, the energies and momenta of the final electrons are determined. Such a detailed differential investigation is fundamental to unravel the role of the dynamical electron-electron correlation during the collision. The understanding of correlation in multielectron transitions, where the Coulomb force plays a paramount role, is one of the basic unsolved problems of modern atomic physics. Measurement of the coincidence angular distributions of the DI products should provide an essential insight into this problem. The ideal target for such a study is helium, the simplest two-electron system that yields a pure three-body or four-body problem in the final state under photon or electron impact, respectively. In the last few years $(h\nu, 2e)$ photodouble ionization (PDI) experiments [1] have provided a wealth of new results in several kinematics and energy sharing conditions from near zero up to 50 eV above the threshold of the process [2]. In contrast, electron impact $(e, 3e)$ experiments [3] have proved much more difficult to perform. Detailed $(e, 3e)$ experiments have been recently reported for the outer-shell DI of argon [4] and neon [5], but were unsuccessful for helium. Moreover, because of the low triple coincidence counting rate, the modest energy resolution did not allow us to distinguish among different final states of the doubly charged ion [4,5]. In that sense, these were not kinematically completely determined experiments.

In this paper we report the $(e, 3e)$ cross sections for DI of He. For the first time, a kinematically completely determined experiment (apart from electron spins variables) has been performed, since the He^{2+} ion is a bare nucleus with no relevant internal structure. These experiments were performed in the so-called dipole scattering regime, characterized by high incident energy and small momentum transfer to the target (i.e., small scattering angle), where the electron impact cross section is expected to converge on the photoionization cross section [6].

A complete description of the electron impact $(e, 3e)$ process in a coplanar geometry is given by the variables represented in the vector diagram in Fig. 1. E_j and $\mathbf{k}_j$ ($j = 0, a, b,$ or c) are the electron energies and momenta. The index 0 stands for the incident electron. The outgoing electrons, although indistinguishable, are indexed with an "a" for the fast "scattered" one, while of the two slower "ejected" electrons the fastest is labeled "b" and the slowest "c." The collision plane is defined by $\mathbf{k}_0$ and $\mathbf{k}_a$, and $\mathbf{K} = \mathbf{k}_0 - \mathbf{k}_a$ is the momentum transfer to the target (the $\mathbf{K}$ direction plays the role of the direction of the axis of polarization of the light in the PDI case). The ion recoil momentum $\mathbf{q}_r$ is given by $\mathbf{q}_r = \mathbf{K} - (\mathbf{k}_b + \mathbf{k}_c)$, that is, the momentum transferred to the target minus that one of the center-of-mass of the ejected electron pair. The full determination of all kinematical variables requires that the three final electrons be simultaneously analyzed both in direction and in energy and detected in triple coincidence. This yields a fivefold differential cross section (5DCS) with respect to the three solid angles of the outgoing electrons and the energies of two of them (the third being known from the energy conservation equation). The coplanar kinematics is the natural one, because $(\mathbf{k}_0, \mathbf{k}_a)$ define a plane, and $\mathbf{K}$ lies in this plane; thus no momentum is transferred to the target out of plane. $(e, 2e)$ experiments have shown that the out-of-plane geometry is dominated by high (second) order effects, and this kinematics does not

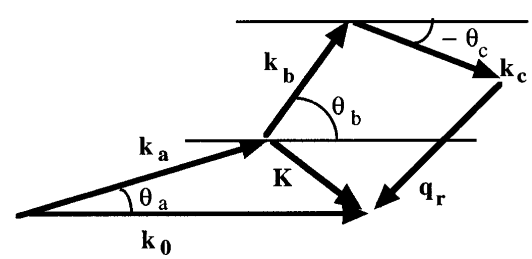

FIG. 1. Schematic momentum vector diagram for a coplanar $(e, 3e)$ electron impact double ionization experiment. See text for explanation of symbols.

give an appreciable contribution to the total cross section. Thus, even though the $(e, 3e)$ is a many-body reaction, and particles can be ejected all over the space provided the perpendicular momentum is zero, for similarity we have restricted our investigation to coplanar kinematics.

The essential feature of the $(e, 3e)$ spectrometer used here [4,7] is the multicoincidence and multiangle detection of both ejected electrons using two twin double toroidal analyzers equipped with position sensitive detectors (PSD). Briefly, the incident electron beam ($E_0 = 5599$ eV) crosses the target gas beam at a right angle. The fast scattered electrons ($E_a = 5500$ eV) are angle selected, then energy analyzed in a cylindrical analyzer, and detected on a scintillator-photomultiplier arrangement. The scattering angle ($\theta_a = +0.45° \pm 0.10°$), and hence the momentum transfer vector is fixed. A high momentum transfer resolution is achieved, $\Delta K = \pm 0.02$ a.u., while the resolution for the total momentum of the recoiling ion is $\Delta q_r \sim 0.1$ a.u. The ejected electrons ($E_b = E_c = 10 \pm 2$ eV) are collected by the toroidal analyzers in the collision plane $(\mathbf{k}_0, \mathbf{k}_a)$ over the useful angular ranges $20° < \theta_b < 160°$ and $200° < \theta_c < 340°$. (Throughout this paper, positive scattering and ejection angles are measured counterclockwise, starting at the incident beam direction, and are allowed to vary between 0 and 360°.) Electrons arriving at any position on one of the two PSDs can be correlated with electrons simultaneously arriving anywhere on the other PSD. A triple multicoincidence is thus performed between the scattered electron, on the one hand, and any pair of ejected electrons, on the other hand. This allows one during the off-line analysis to sort the data in a variety of modes: (i) the so-called "θ-variable mode" either at fixed θ_b and varying θ_c or vice versa; (ii) the "fixed mutual angle mode" at varying θ_b and θ_c but keeping fixed the mutual angle θ_{bc}; (iii) the so-called "symmetric geometry mode" with varying but equal emission angles (modulus 2π), $\theta_b = 2\pi - \theta_c$. This results in the simultaneous production of a large number of angular distributions.

The obtained three-dimensional triple coincidence time spectrum is shown in Fig. 2. In this case, the data from all θ_b and θ_c angles have been combined. The peak at the center corresponds to the triple coincidence DI signal, superimposed on a background due to the fully accidental coincidences, where the three electrons e_a, e_b, and e_c are uncorrelated, and the semirandom ones are due to two correlated electrons, the third one being random [8]. These produce walls or ridges in the time spectrum. Two run parallel to the time axes (e_a-e_b and e_a-e_c walls), and the third one runs along the diagonal (e_b-e_c wall).

Because of the very small value of the 5DCS, a long accumulation time (~32 days) was needed to reach a reasonable statistical accuracy. Correspondingly, a very high stability of all the experimental parameters is desired and was achieved [7]. Before starting the $(e, 3e)$ acquisition, $(e, 2e)$ angular distributions for single ionization of He were measured under the same kinematical parameters as in the $(e, 3e)$ experiments. The good agreement found with well-established results [9] excludes systematic distortion of the measured angular distributions due to experimental artifacts. Moreover, a daily measurement of the same $(e, 2e)$ angular distributions enabled us to correct in the off-line analysis for any local changes in the collection efficiency of the multichannel plates [7].

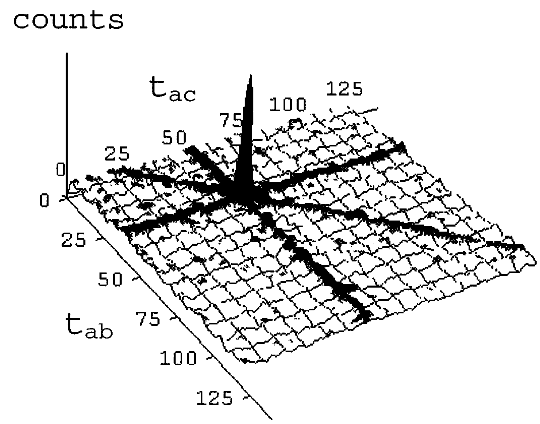

FIG. 2. 3D coincidence time spectrum: The number of coincidences is plotted versus the electrons arrival time differences, t_{ab} and t_{ac}. Shown are the triple coincidence peak and the three semirandom "walls" lying on top of the fully accidental contribution.

The full set of experimental data is shown in Fig. 3 as a 3D plot of the 5DCS versus both the ejection angles θ_b and θ_c. The slow electrons are preferentially ejected in two groups which result in the two hills of the 3D plot: the backward hill at about ($\theta_b = 110°; \theta_c = 230°$) and the forward hill at about ($\theta_b = 45°; \theta_c = 275°$). The two hills are separated by a valley, and it is interesting to observe that the Bethe ridge condition, which corresponds

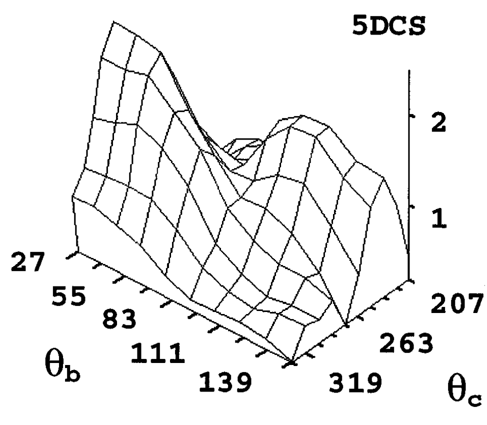

FIG. 3. 3D plot of the $(e, 3e)$ fivefold differential cross section for He for two 10 eV ejected electrons versus both the ejection angles θ_b and θ_c. $E_0 = 5599$ eV, $E_a = 5500$ eV, $\theta_a = +0.45°$.

to the case where all momentum transfer is given to the two atomic electrons ($\mathbf{K} = \mathbf{k}_b + \mathbf{k}_c$), and hence no momentum is imparted to the ion ($q_r = 0$), occurs at ($\theta_b = 41°; \theta_c = 237°$), i.e., exactly at the bottom of the valley, where the coincidence intensity is at a minimum. Considering that (i) the collisions leading to events in the backward (forward) hill correspond to a rather large ion recoil momentum, $q_r \sim 1.2$ a.u. ($\sim$0.6 a.u.), and (ii) the ones with zero or minimum momentum imparted to the ion are unlikely; these data show that the DI mechanism, at least in the present kinematics, requires a large active participation of the ion and illustrates the complexity of the process. It also means that, unlike the He $(e, 2e)$ case, where binary electron-electron interactions are predominant, the $(e, 3e)$ "binary" interaction of the incident particle with the "pair of ejected electrons" is less probable.

Among all the possible cuts of Fig. 3, we discuss the following two examples that we believe to be of particular significance. The conditions of our experiments were chosen to closely mimic existing PDI data [10] at $E_1 = E_2 = 9.3$ eV, where the indexes 1 and 2 refer to the two photoelectrons. The He coincidence angular distribution for equal energy sharing is given by [10]

$$\sigma_{\mathrm{PDI}} = |a_g(E_1, \theta_{12})|^2 (\cos\theta_1 + \cos\theta_2)^2, \qquad (1)$$

where the amplitude a_g is the angular correlation function, θ_1 and θ_2 are the electron emission angles with respect to the direction of the polarization of the light, and θ_{12} is their mutual angle. The effects of the electron-electron and electron-residual ion interactions are included in the energy and θ_{12} dependence of a_g. The σ_{PDI} distribution shows two nodes: The first one at $\theta_{12} = 0$, where a_g approaches zero, corresponds to the forbidden situation, where the two electrons fly out in the same direction with the same energy; the second node at $\theta_{12} = \pi$, where the cosine terms in (1) cancel each other, is characteristic of the $^1P^o$ symmetry for the pair of outgoing electrons in ionization of He [11]. These nodes depend only on θ_{12} and not on the emission directions θ_1 and θ_2 themselves. Hence, our $(e, 3e)$ data are sorted as a function of the mutual angle θ_{bc} in Fig. 4, adding up all the pairs that make the same θ_{bc}, irrespective of the direction of emission θ_b. The main features of the PDI distributions are also found here with a vanishing intensity in the $\theta_{bc} = 0$ direction and a minimum at the mutual angle θ_{bc} of 180°. However, differences are also observed: (i) The two lobes although symmetrical are wider than in the PDI case, and (ii) the PDI characteristic node at 180° is here "filled up." Neither of these effects can be explained by the finite angular resolution ($\pm 7°$) of the spectrometer. A plausible explanation is that nondipolar contributions are always present in the electron impact DI, and hence several electron final states are accessible. Thus, the 180° mutual angle is no longer forbidden, and the minimum is less pronounced. A similar observation was previously reported [12] in $e, (3-1)e$ experiments on He, where the 5DCS was integrated over all possible momentum transfers.

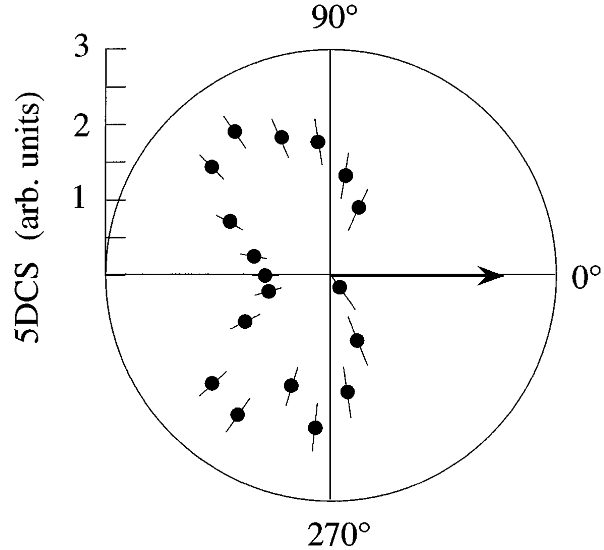

FIG. 4. $(e, 3e)$ 5DCS for He plotted as a function of the mutual angle θ_{bc}, irrespective of the direction of emission θ_b. Kinematical parameters as in Fig. 3. The error bars are 1 standard deviation statistical error after the addition of all of the pairs that make the same θ_{bc}.

As an example of the θ-variable mode, we represent in Fig. 5 the 5DCS distribution measured when one electron is detected along $\mathbf{K}$ ($\theta_c = \theta_K = 319°$), and the other one is mapped in the opposite half plane. The results are compared on a relative scale with the only existing $(e, 3e)$ calculations [13] performed within the Born approximation. Among the different mechanisms proposed for DI by electron impact [14], only the shakeoff and the two-step-2 (TS2) contributions were considered in the calculations. No major differences, at least in the part of the plane accessed by the present measurements,

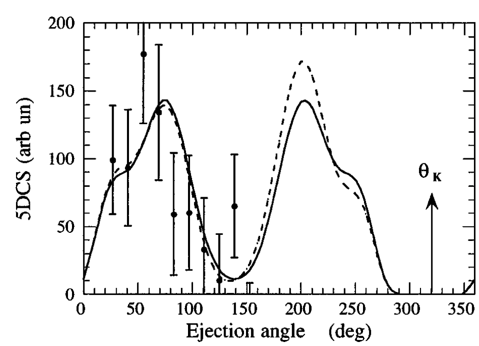

FIG. 5. $(e, 3e)$ 5DCS for He versus ejection angle for one "atomic" electron. The other ejected electron is observed along the $\mathbf{K}$ direction, shown by the arrow. Full circles, experiments. Full and broken lines, shakeoff and shakeoff plus two-step-2 contributions, respectively (Ref. [13]). Kinematical parameters as in Fig. 3.

are predicted by the theory when the TS2 mechanism is included or not included in the calculations. In spite of the large statistical error bars, the agreement between experiment and theory is satisfactory, especially as far as the positions of the maximum and minimum intensity are concerned.

The high sensitivity and efficiency of an angular multichannel experimental setup have enabled us to achieve the first fully determined measurement of the DI of He by electron impact. The richness and, at the same time, complexity of the $(e,3e)$ data are illustrated by few selected results. Because of the chosen dipolar kinematics, a subset of the obtained data has been compared with PDI results at the same excess energy above the DI threshold. The comparison gives clear evidence for the presence of nondipolar contributions in the $(e,3e)$ data. A second subset of the data was found in satisfactory agreement as to the shape with available Born calculations. Binary collisions of the incident particle with the "pair of ejected electrons" are shown to be unlikely, whereas events implying a large participation of the ion are more probable. The full set of the data obtained have been fully analyzed, and their representation in the θ-variable mode, as shown in a forthcoming longer paper [15], is clearly the best suited testing ground for theoretical models that aim to unravel the mechanisms of electron impact DI. The understanding of the DI mechanism in He will help in selecting the base model and in disentangling the contribution of the "target structure" from the pure dynamical effects. At present, because of the low energy resolution that did not allow the separation of the different ionic states in the heavier rare gases [4,5], only a qualitative observation can be done [16] on the relevance of the second and higher order DI mechanisms which are of increasing importance when going from He to Ne and to Ar.

Finally, as previous $(e,2e)$ experiments had provided unique information on the dynamics of the three-body Coulomb problem, this work shows that $(e,3e)$ experiments are quickly approaching the state where similar results are obtainable for the case of the four-body Coulomb problem. Moreover, the combination of the $(e,3e)$ and $(h\nu,2e)$ results provides detailed and complementary pictures of the electron-electron correlations that dominate the DI process.

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Note added in proof.—A. Dörn and co-workers in Freiburg have recently succeeded in observing $(e,3e)$ coincidence events from He, using the COLTRIMS technique.

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The full set of data may be obtained on request from this author.
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