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LETTER TO THE EDITOR

Comparative theoretical study of (e, 3e) on helium: Coulomb-waves versus close-coupling approach

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Abstract

We discuss common features and differences in the fully differential cross sections (FDCS) for the electron-impact double ionization of He evaluated using the convergent close-coupling (CCC) method and various methods that utilize the three-body Coulomb wavefunctions (3C and C4FS). The calculations are restricted to the first Born approximation with respect to the interaction of the fast projectile with the target. In contrast to the similar (γ, 2e) reactions, where predictions of all the above theories qualitatively agree, there is a systematic difference between the FDCS calculated in the CCC and the 3C-type theories which is amplified as the momentum transfer from the projectile to the target increases. We argue that this fact is an indication that the P partial wave of a two-electron final state wavefunction is well described by the 3C model, but higher partial waves are poorly treated.

The angular and energy-resolved detection of the double ionization fragments following fast electron impact on atomic targets (the so-called (e, 3e) reaction) is a powerful tool to study few-particle dynamics. As compared to the fully resolved double ionization upon the absorption of a single VUV photon (the (γ, 2e) reaction), the momentum q transferred from the projectile to the target in a (e, 3e) process can be varied in a wide range. In the *dipolar regime*, under the condition $q \ll 1$, the incoming electron beam induces predominantly two-electron electric-dipole transitions in the target and the (e, 3e) and (γ, 2e) processes are directly related. In contrast, in the *collisional regime*, where $q \leqslant 1$ is more substantial, higher multipoles provide significant contributions. Ultimately, in the *impulsive regime* $q > 1$, the ionization event can be described as a binary knock-out collision of the projectile with the target.

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The helium atom is an ideal target to study the (e, 3e) reaction since its electronic structure is relatively well understood and the final state of the ion is a bare nucleus. The (e, 3e) experiments on He have been reported by the Orsay group (Taouil *et al* 1998, Lahmam-Bennani *et al* 2001) and by the Freiburg group (Dorn *et al* 1999, 2001a, 2001b). Both experiments involved fast (few keV) incident electrons and low energy (few eV) ejected electrons. In the Orsay experiments the momentum transfer was small ($q < 1$), whereas the use of the cold target recoil ion momentum spectroscopy (COLTRIMS) allowed the Freiburg group to detect simultaneously both the $q < 1$ and $q > 1$ ionization events. In both experiments the fully differential cross section (FDCS) was measured. The absolute FDCS were reported by the Orsay group, whereas only relative data were obtained from the Freiburg experiments.

From a formal theoretical point of view the description of the (e, 3e) reaction entails the knowledge of the scattering states of an interacting system which consists of four charged particles (three electrons in the field of the He^{2+} ion) as well as the evaluation of a correspondingly complicated series of scattering amplitudes (Berakdar 2001b). To render possible the numerical evaluation of (e, 3e) cross sections one considers kinematical situations in which the fast incoming electron transfers a small momentum to the target $q \lesssim 1$ and ejects two slow electrons. In this case the following simplifying assumptions are reasonable. The fast projectile is represented by a plane wave and its interactions with the target can be treated perturbatively by way of the Born series. Most practical computations are restricted to the first non-vanishing term in the Born series (the first Born approximation—FBA). However, the motion of the two slow ejected electrons in the presence of the residual ion has to be treated nonperturbatively. The latter process constitutes a Coulomb three-body problem and a number of approaches have been developed to treat this problem computationally. Differences between various computational methods may result in different FBA results even if the experimental kinematics ensures the applicability of the FBA.

An efficient and (in most cases) accurate way to describe a three-body Coulomb system is provided by the three Coulomb waves (3C) method (Brauner *et al* 1989). This method was applied to the (e, 3e) process on He by Joulakian *et al* (1992), Joulakian and Dal Cappello (1993), and most recently by Muktavat and Srivastava (2001). In the 3C theory the three-body system (the two slow electrons and the He^{2+} ion) is broken down into three decoupled two-body subsystems. The latter are then treated to all orders of perturbation theory, i.e. isolated two-body scattering events are correctly described by the 3C model, whereas the modification of the two-particle scattering due to the presence of a third charged particle is not taken into account. Such a three-body coupling occurs through cross terms of the kinetic energy operator. These terms are most important at lower energies and/or small distances, and hence the 3C theory becomes justifiable for large inter-particle separation *or* at high energies (Berakdar 2001a). We note that the separation of small and large distances is energy dependent here. Similarly, the 3C theory performs well in the high energy regime or when, in a scattering reaction, isolated two-body collisions are dominant.

A more recent non-perturbative method to treat the Coulomb three-body problem was developed by Bray and Stelbovics (1992) and applied to the (e, 3e) on He by Kheifets *et al* (1999a, 1999b). In this fully numerical convergent close-coupling (CCC) method only one of the two slow electrons is described by a Coulomb wave whereas the other electron is treated as a pseudo-state of the He^{+} ion. Inelastic scattering of the Coulomb wave on the He^{+} ion results in promotion of the bound electron to the positive energy pseudo-state thus creating a doubly ionized final state. A similar approach was taken independently by Knyr *et al* (2001).

Up to now, due to numerical complexity, the close-coupling calculations of the (e, 3e) on He were limited to the FBA. The second Born correction was estimated only on the basis of the 3C theory (El Mkhanter and Dal Cappello 1998, Grin *et al* 2000). It was found that

the first Born term was clearly dominant. However, the second Born correction was sizeable even at large incident electron energies. A further non-perturbative method that goes beyond the FBA was suggested by Berakdar (1997) and applied to the (e, 3e) on He by Lahmam-Bennani *et al* (1999). This method is a generalisation of the 3C approach to a four-body system, however the interaction of the scattered projectile electron with the rest of the system is described on the basis of effective charges yielding thus a correlated four-body final state (C4FS). Recently, an incremental many-body Green function technique has been utilized by Berakdar (2000) to derive the four-body Green function and applied to the calculation of the (e, 3e) cross section. However, due to the numerical complexity only a limited set of calculations is available (Berakdar 2000).

Comparison of the experimental and theoretical FDCS produced mixed results. The first experiment of the Orsay group performed at a very high incident energy of 5.5 keV in the collisional $q \lesssim 1$ regime (Taouil *et al* 1998) was qualitatively understood by applying the dipole selection rules (Lahmam-Bennani *et al* 1999). However, detailed comparison of the shape of the measured and calculated FDCS showed strong deviation at some angles (Kheifets *et al* 1999). Comparison was much worse for the magnitude of the FDCS which varied greatly from one calculation to another (Kheifets *et al* 1999, Lahmam-Bennani *et al* 1999, Knyr *et al* 2001) and none of them agreed with the absolute measurements of the Orsay group. Recently, the Orsay group reported the experimental FDCS at the incident energy of 1 keV (Lahmam-Bennani *et al* 2001). These results have been compared with the 3C, C4FS and the CCC calculations. Considerable deviations between theory and experiment have been observed which have been assigned to contributions of scattering processes whose description goes beyond the FBA. This proposition seems reasonable in view of the relatively low energy of the incident electron. On the other hand, results of the Freiburg experiments at the incident energy of 2 keV, both in the dipole regime (Dorn *et al* 2001a) and the impulsive regime (Dorn *et al* 2001b), have been compared with the FBA–CCC calculations. The theory explained well the main features of the experimental FDCS. Only minor deviations of the experiment from the symmetry expected in an FBA regime could not be reproduced by the FBA–CCC theory.

Clearly, incident electron energy in (e, 3e) reactions can always be taken sufficiently low so that there is a need for higher Born corrections. However, at this stage, even the FBA calculations from different models strongly disagree. This could be seen in a recent work by Lahmam-Bennani *et al* (2001) where the 3C and C4FS calculations, while very close to each other, were far apart from the CCC calculation. In contrast, similar (γ, 2e) calculations performed with the 3C and CCC models produced very close, if not identical, results (Cvejanović *et al* 2000, Bolognesi *et al* 2001, Dawson *et al* 2001).

In this letter we trace the emerging difference between the close-coupling and Coulomb-waves based theories using the cross-over of the (e, 3e) reaction from the dipolar regime in which it is identical to the (γ, 2e) reaction to the collisional regime. The kinematics of the Orsay experiment lie in the latter regime. The full set of theoretical FDCS is presented here of which only a small sub-set have been reported previously in (Lahmam-Bennani *et al* 2001).

The evolution of the FDCS from the dipolar to the collisional regime is illustrated in figure 1. Here we show results of the CCC calculations for the (γ, 2e) and (e, 3e) reactions at ejected electron energies of $E_1 = E_2 = 10$ eV. Geometries of the (γ, 2e) and (e, 3e) reactions are depicted at the bottom panels. In the case of the (e, 3e) reaction the kinematics of the Orsay experiment (Lahmam-Bennani *et al* 2001) are used, where the incident electron (momentum vector $\boldsymbol{k}_0$ along the z direction) has 1 keV energy. In the case of the (γ, 2e) reaction the coplanar geometry is chosen in which both photoelectrons (momentum vectors $\boldsymbol{k}_1$, $\boldsymbol{k}_2$) are detected in the polarisation plane of the linearly polarized light. The escape angles of the two photoelectrons θ_1 and θ_2 are measured with respect to the electric field vector $\boldsymbol{E}$ which is

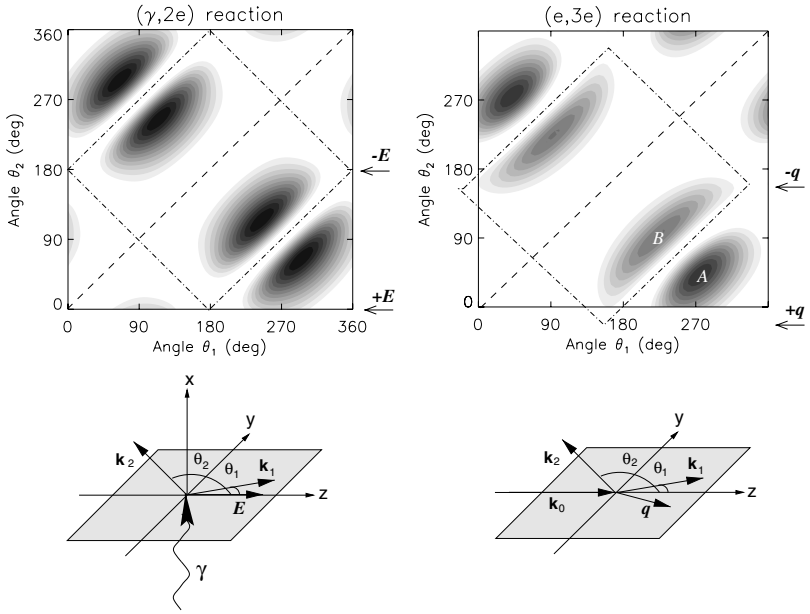

Figure 1. The FBA–CCC calculations of the He (γ, 2e) and (e, 3e) fully differential cross sections at ejected electron energies of $E_1 = E_2 = 10$ eV (top left and right panels, respectively). Geometries of the (γ, 2e) and (e, 3e) reactions are illustrated in the bottom panels. Cross sections are exhibited in contour plots as functions of the escape angles θ_1 and θ_2. The areas of large cross section are indicated by darker shades of grey. The straight lines indicate the areas of vanishing cross section according to the dipole selection rules (dash-dotted lines) and Pauli exclusion principle (dashed line). The arrows show the directions of the electric field vector $\pm\boldsymbol{E}$ and the momentum transfer vector $\pm\boldsymbol{q}$ for the (γ, 2e) and (e, 3e) reactions, respectively. The angular range accessible in the Orsay experiment contains the peaks denoted by A and B in the plots, see text.

directed along the z axis. We use contour plots to exhibit FDCS as a function of the escape angles θ_1 and θ_2. The areas of larger cross section are indicated by darker shades of grey.

The (γ, 2e) FDCS vanishes along the dipole nodal lines $\theta_1 \pm \theta_2 = \pm 180^\circ$. In the case of the low-q (e, 3e) reaction the cross section also remains small along the lines $\theta_1 - \theta_2 = \pm 180^\circ$ and $\theta_1 + \theta_2 = \pm 180^\circ - 2\theta_q$, where $\theta_q = 21^\circ$ is the momentum transfer angle for the chosen kinematics. Both cross sections are symmetric with respect to the electron interchange $\theta_1 \leftrightarrow \theta_2$ since $E_1 = E_2$.

The angular range accessible in the Orsay experiments is restricted to the angles containing the two peaks, marked A and B. These peaks are identical in the (γ, 2e) reaction because the directions $+\boldsymbol{E}$ and $-\boldsymbol{E}$ are fully equivalent due to a negligible momentum of the photon $k = \omega/c \ll 1$. This is not so in the (e, 3e) reaction since the momentum transfer q is finite. As we observe in the right plot of figure 1 the peak A acquires more intensity relative to the weaker peak B. The peak A is aligned closer to the direction of the momentum transfer $+\boldsymbol{q}$ and it is convenient to label it as a binary peak. Conversely, the peak B is positioned closer to the vector $-\boldsymbol{q}$ and we shall call it the recoil peak. It is natural that the asymmetry between the binary and recoil peaks increases as the momentum transfer q grows.

In figure 2 we present the CCC calculated (e, 3e) FDCS restricted to the bottom-right corner of figure 1, where experiment exists. We also show the corresponding FDCS from the 3C and C4FS calculations as well as the experiment of Lahmam-Bennani *et al* (2001). We see that the results of the 3C and C4FS calculations are quite close. This is expected, as for higher

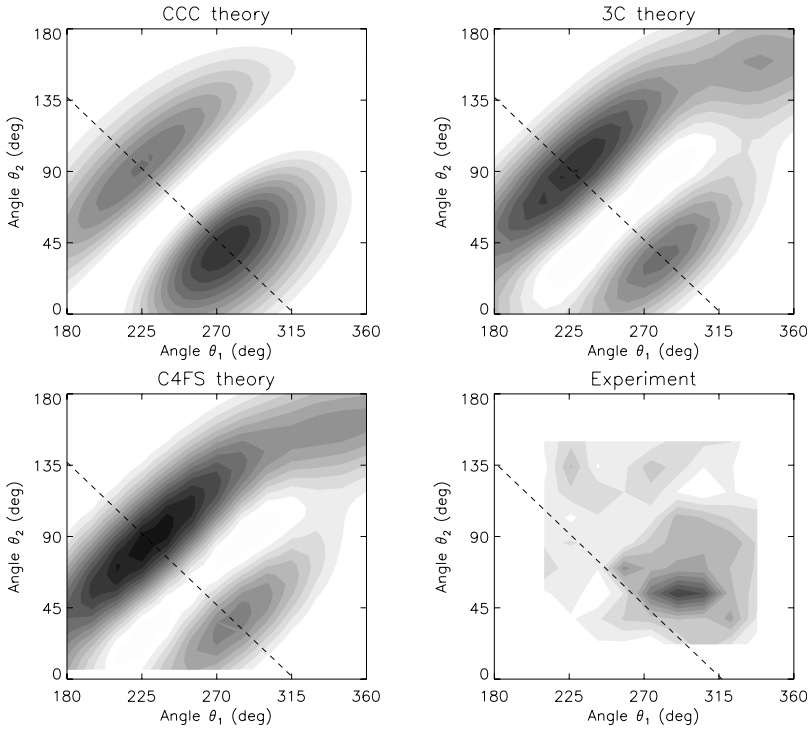

Figure 2. Contour plots of the (e, 3e) FDCS at the kinematical conditions of the experiment by Lahmam-Bennani *et al* (2001). The escape angles of the two slow ejected electrons θ_1 and θ_2 are shown on the axes. Only the quadrant of the experimentally accessible range of angles is displayed. Top left—CCC calculation, top right—3C calculation, bottom left—C4FS calculation, bottom right—experiment. The dashed line indicates the symmetry axis of the FBA model.

impact energies and small momentum transfer the C4FS tends to the 3C case. Unlike the CCC theory, the 3C and C4FS place more intensity on the recoil peak as compared to a weaker binary peak. In addition, FDCS is significant at $\theta_1 \sim 360^\circ$ and $\theta_2 \sim 180^\circ$ in the 3C and C4FS calculations whereas it is strongly suppressed by the remnant dipole selection rule in the CCC calculation. The measurement of Lahmam-Bennani *et al* (2001) indicates a somewhat larger intensity in the area of the binary peak. This peak is displaced relative to the symmetry axis of the FBA model indicated by the dotted line. All the presented calculations are symmetric with respect to this line.

A similar divergence of the CCC and 3C calculations, and the 3C theory with experiment, was also encountered when comparing the cross sections of the (γ, 2e) and (e, 2e) reactions. While the main structures in the measured (e, 2e) cross sections were present in the 3C calculations, the relative intensities, the absolute magnitudes, and the positions of the peaks were not well predicted at lower energies. The terms in the Hamiltonian that are responsible for this shortcoming of the 3C method have been identified (Berakdar 2001a), however till now there are no numerical calculations that estimate correctly their relative importance in various regions of the phase space. The two-electron continuum states of the (e, 2e) and the (γ, 2e) processes are identical. Nonetheless, at low incident energies (few times the ionization potential), the 3C theory describes well the angular correlation in the two-electron continuum of the (γ, 2e) reaction (Bolognesi *et al* 2001, Cvejanović *et al* 2000, Dawson *et al* 2001) but

is clearly at variance with the experimental (e, 2e) angular distributions (Berakdar *et al* 1999). This situation can be understood by recalling the fact that with increasing q the number of relevant partial waves of the final state wavefunction increases. The 3C theory fails to represent these partial waves adequately.

In conclusion, we studied the (e, 3e) FDCS by means of the accurate fully numerical CCC method and by use of the analytical 3C model. Both approaches produce similar results in the dipolar $q \ll 1$ regime when the (e, 3e) reaction is equivalent to the (γ, 2e) reaction but notably disagree in the collisional $q \lesssim 1$ regime. From this comparison we conclude that the 3C model treats the dipole component of the two-electron continuum adequately but has serious difficulties in describing other partial waves whose contributions grow with increasing q. It should be noted that the region of the configuration space which is relevant for the calculations of the cross sections of the (e, 3e) and (γ, 2e) reactions is limited by the extent of the helium atom ground state wavefunction. Therefore the asymptotic properties of the 3C final state wavefunction play a minor role when it comes to evaluating the FDCS. In essence, the asymptotic behaviour of the 3C wavefunction is a byproduct of the fact that two-body collisions are treated accurately and multiple higher-order collisions are negligible asymptotically. The decisive shortcoming of the 3C is the neglect of the short-range kinetic energy coupling terms which seem to gain more importance with increasing q. Although the impulsive $q > 1$ regime is not explicitly studied here, results of Dorn (2001) at $q = 2$ indicate that the 3C theory deviates very strongly from the experiment. On the other hand, the CCC theory qualitatively agrees with the experimental data.

Depending on the value of the momentum transfer the (e, 3e) reaction can be performed in very different dipolar, collisional and impulsive regimes. This is in contrast to the (γ, 2e) reaction which corresponds to the (e, 3e) reaction only in one particular dipolar regime. This dynamical richness of the (e, 3e) reaction makes it a useful tool for a stringent test for newly developed theories of the atomic ionization (Pindzola *et al* 1999, Rescigno *et al* 1999, Malegat *et al* 2000).

Generally, higher Born corrections have to be included into theoretical models of the (e, 3e) reaction. However, before this is attempted, convergent FBA results have to be determined. This convergence between different FBA models has already been achieved in a related process of the electron impact single ionization (e, 2e reaction) which leaves the ion in an excited state (Kheifets *et al* 1999a, Marchalant *et al* 1999). The present work is aimed to stimulate the establishment of such convergence in (e, 3e) collisions.

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