Appearance and disappearance of the second Born effects in the $(e,3e)$ reaction on He

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We demonstrate, both experimentally and theoretically, clear manifestation of the second Born effects in the angular distributions of two ejected electrons produced by a 500 eV electron impact on the He atom in the so-called $(e,3e)$ reaction. The second Born contribution, due to subsequent interaction of the projectile with the target, is most prominent for glancing collisions with small momentum transfer. However, these effects are absent for hard knock-out collisions with large momentum transfer.

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Complete fragmentation of the helium atom under electron impact is one of the clearest examples of the Coulomb four-body break-up process. Understanding of such processes still remains a challenging task [1]. In conrast, a related but simpler process of He fragmentation under photon impact is much better understood [2]. The double photoionization proceeds along the following two pathways, or processes. In the ''shake-off'' process, simultaneous ejection of the second photoelectron takes place because the departure of the first electron suddenly changes the effective atomic field. In the rescattering, or ''two-step-one'' process, the first ejected electron knocks the second electron on its way out of the atom. Relative contributions of these two processes depend on the photon energy. As the photon energy increases, the shake-off process becomes gradually dominant over the two-step-one process. The signature of this take-over is the energy-independent ratio of the double-to-single photoionization cross sections [3].

Physics is more complicated for double ionization by fast electron impact. In addition to the shake-off and two-step-one processes, there is the possibility of the projectile colliding with the target repeatedly, ejecting two electrons in sequence (the so-called two-step-two process [4]). It is customary to consider this repeated interaction within perturbation theory and to label it as a higher-order Born process, as opposed to the first-order Born process in which the projectile interacts with the target only once. The perturbation theory parameter Z/v is the ratio of the projectile charge to its velocity. For very small perturbations, double ionization proceeds predominantly through processes involving a single interaction of the projectile with only one target electron. For large perturbations, the dominant double ionization mechanism involves two independent interactions of the projectile with both electrons.

This separation of the first and higher Born regimes, i.e. the crossover from single to multiple projectile-target interaction, is based on the analysis of the ratio of the double-to-single ionization cross sections [5,6]. This ratio, however, is a fairly rough indicator of the relative contributions of different ionization mechanisms. In a recent study of the double photoionization of He [7], a much more detailed separation was achieved by investigating the angular and energy correlations between two ejected photoelectrons, detected in time coincidence [the so-called $(\gamma,2e)$ reaction]. Similar coincidence studies can now be performed for electron impact ionization of He [the $(e,3e)$ reaction] [8–14]. In these experiments, the energy of the projectile has varied from 5.5 keV [8] down to 0.6 keV [13,14]. The latter is likely to be in the domain of the significant non-first-Born contributions. The presence of higher-order Born effects was identified in Ref. [13] and, more clearly, in Ref. [14]. Indeed, the experimental results of Ref. [14] were completely inconsistent with a first Born calculation based on the convergent close-coupling (CCC) theory [15]. The CCC model treated the interaction of the two ejected electrons exactly. Therefore, its deviation from experiment could only be attributed to higher Born processes which were not included in this implementation of the CCC model. The authors of Ref. [14] also applied a second Born model [16] based on the asymptotic three-body Coulomb wave functions, known in the literature as BBK. This model, however, is inadequate for the description of the low incident energy $(e,3e)$ reaction as it gives inaccurate results already in the first Born term [17]. Götz *et al.* [18] extended the BBK model to the four-particle continuum, thus incorporating the first and higher Born terms. However, this extension inherited all the problems of the BBK which were already manifested clearly in the first Born term. In addition, the calculated results were at variance with the experiment by Lahmam-Bennani *et al.* [13]. Another theoretical attempt to go beyond the first Born model was made by Berakdar [19], who employed an incremental approach to the four-body Coulomb problem. The use of this method, however, was limited by numerical difficulties.

In this paper, we employ a new set of experimental and theoretical tools and present clear evidence of sequential double ionization of He at 500 eV electron impact. In addition, we observe a new, somewhat unexpected effect. Deviation from the first Born regime depends strongly on the momentum transfer from the projectile to the target. We investigate two qualitatively different reaction kinematics, corresponding to glancing incidence of the projectile (small momentum transfer of 0.7–0.9 a.u.) and heavy knock-out on the target (large momentum transfer of 2 a.u.). At glancing incidence, the projectile bounces off the target and impinges on it again in a very strong deviation from the first Born regime. In stark contrast, for heavy knock-out collisions the projectile makes a very close encounter with the target, ejecting two electrons at once, with the first Born contribution becoming dominant. These two markedly different regimes are observed at the same impact energy of the projectile, i.e., the same perturbation Z/v, which is at odds with the conventional perturbation theory. We corroborate these experimental findings by carrying out first and second Born calculations based on the CCC model. The second Born implementation of the CCC theory is reported here for the first time, to our knowledge.

The experiment was performed with the same combined multielectron recoil-ion spectrometer that has been used for earlier high-energy experiments [11,12]. In brief, ions and slow electrons produced in the intersection point of the He target and a pulsed electron beam were extracted in opposite directions by means of static electric and magnetic fields and detected by two position-sensitive multichannel-plate detectors. From the measured positions and times of flight, the momentum vectors of two slow electrons $\boldsymbol{k}_b$ and $\boldsymbol{k}_c$ and the recoiling ion $\boldsymbol{k}_{\mathrm{He}^{2+}}$ were determined. The kinematics of the fast scattered electron, as well as the momentum $\boldsymbol{q}$ transferred by the scattered projectile, follow from momentum conservation: $\boldsymbol{k}_0-\boldsymbol{k}_a=\boldsymbol{q}=\boldsymbol{k}_b+\boldsymbol{k}_c+\boldsymbol{k}_{\mathrm{He}^{2+}}$, $\boldsymbol{k}_0$ and $\boldsymbol{k}_a$ being the momentum vectors of the incoming and the scattered projectile, respectively. The electron detector was equipped with a fast delay-line readout and a multihit time-to-digital converter. Whereas the complete final-state momentum space is mapped for all ions, the detector dead-time results in a small loss of the total momentum space for the second electron hitting the detector.

We performed the first and second Born calculations with the same highly correlated ground-state wave function and CCC representation of the final state of the He atom with two continuum electrons. The first Born model only allows a single interaction of the projectile with the target. This model has been described in detail in Ref. [15]. The second Born model differs from these earlier calculations by allowing ejection of the two electrons in two subsequent knock-outs. All the intermediate states of the target between two subsequent interactions with the projectile are weighted equally with an average energy denominator $\Delta=\overline{k_n^2}-k^2$, where $\boldsymbol{k}$ is the momentum of the projectile between the collisions (the so-called closure approximation [20]). We follow Ref. [21] and choose, somewhat arbitrarily, $\overline{k_n^2}=(k_0k_1)^{1/2}$. Other choices according to Refs. [22,23] were also tried but the second Born amplitude proved to be rather insensitive to Δ.

To simplify the second Born model further, we note that the Born amplitude decreases rapidly with the momentum transfer and that, most likely, the projectile imparts a small amount of momentum in each encounter with the target. Therefore, we follow Ref. [22] and restrict the interaction of the projectile with the target to the leading dipole term. We note, however, that the decrease of the Born amplitude with the momentum transfer is not rapid enough to substitute the spherical Bessel function in the dipole Born operator by its optical limit $j_1(qr)\rightarrow qr/3$, as attempted in Ref. [22]. In the present second Born calculation, we treated the dipole operator $j_1(qr)$ fully.

In each of the two dipole interactions, the projectile exchanges one unit of angular momentum with the target, resulting in a total angular momentum transfer of 0 or 2 in the second Born process. Restriction of the angular momentum exchange allows us to perform an analytical angular integration over all the possible directions of the projectile between the collisions with the target. This speeds up the computation considerably compared with fully numerical integration employed in earlier second Born calculations [23,24]. As a test of our second Born model, we calculated the fully differential cross section of a related process of the electron impact ionization of helium with simultaneous excitation to the ion $n=2$ state. We used the kinematics of the experiments of Refs. [25,26] for which the second Born results are well established [23,24]. The difference between our calculations and these test results did not exceed 20%.

By applying the first and second Born models to the $(e,3e)$ process and making comparison with experiment, we can clearly identify the kinematics where repeated interactions of the projectile with the target are significant. An example of such an analysis is shown in Fig. 1, where the fully differential cross section (FDCS) is presented for ejection of two equal energy electrons $E_b=E_c=5$ eV at a small momentum transfer of $q=0.7-0.9$ a.u. which is close to the kinematical limit of $q=0.55$ a.u. We present our data by using two-dimensional (2D) graphs in which the ejection angles of two electrons in the projectile scattering plane are plotted on the axes and the cross section is coded by different shades of gray. The experimental cross section Fig. 1(a) consists of four main peaks. Both peaks in the upper left are equivalent to the peaks in the lower right of the diagram (marked A and B), since for symmetric energy sharing both ejected electrons are interchangeable. The pattern of the experimental cross section is dominated by peak B, corresponding to one electron being emitted with an angle slightly larger than 180° ($\theta_b=180°-210°$) and the second electron going in the forward direction ($\theta_c=0°$). A second, much weaker peak A is observed for one electron emitted at 90° and the second electron at small negative angles of $\theta_c=-30°-0°$. In Fig. 1(b) we present the theoretical FDCS obtained within the first Born model. A similar calculation for a higher incident energy of 2 keV was found in fair agreement with experiment [11] with only a minor displacement of peak B. In the present case, however, the first Born model fails completely. Neither the positions nor the relative heights of the experimentally observed peaks are reproduced by the calculation. There are two underlying symmetries of the first Born FDCS which are violated strongly by the experimental data, thus indicating multiple projectile-target interaction. First, a dipole selection rule gives rise to a cross-section minimum for back-to-back emission of the ejected electrons at equal energies. The corresponding angular combinations are marked by dashed lines in Fig. 1. The first Born cross section in Fig. 1(b) largely obeys this selection rule with only a small intensity for back-to-back emission with one electron going along the momentum transfer direction. On the contrary, the experimental cross section, which violates this selection rule maximally since peak B, the strongest feature of the observed pattern, corresponds to back-to-back emission of the ejected electrons. Assuming dipolar collisions to be most important, this can only be the result of at least two collisions

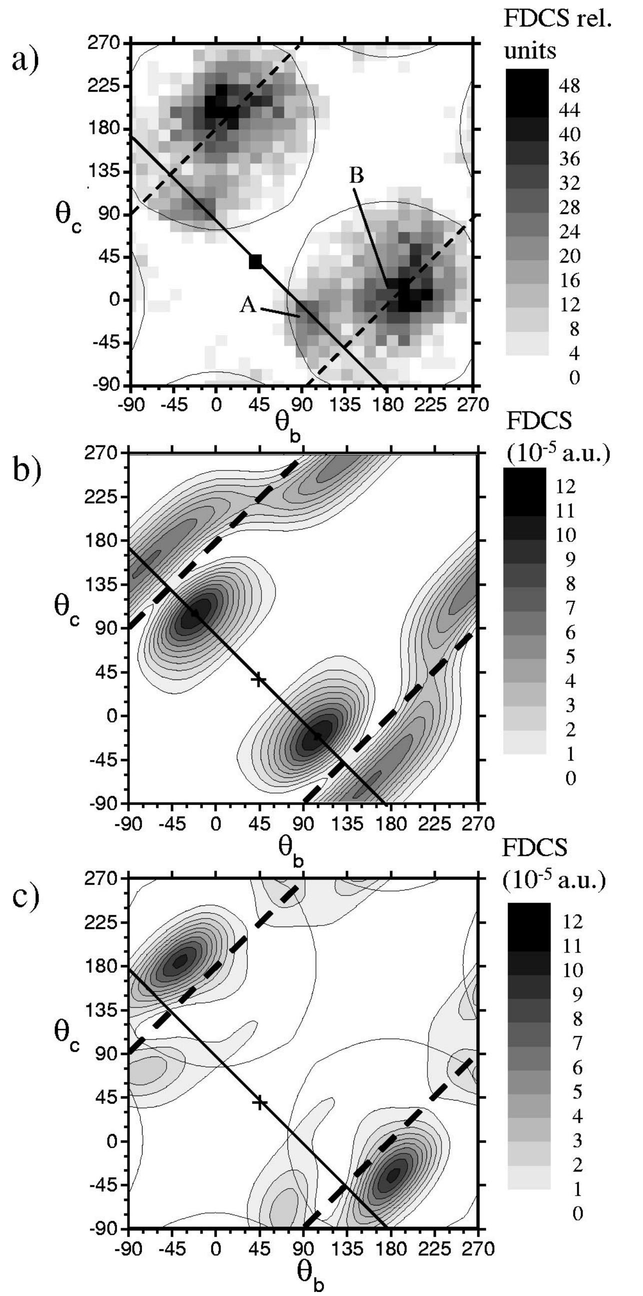

FIG. 1. Fivefold differential cross section (FDCS) for $E_0=500$ eV in coplanar scattering geometry as a function of the ejected electrons emission angles θ_b and θ_c relative to the primary beam forward direction. (a) Experimental cross section for $q=0.8\pm0.1$ a.u. and $E_b=E_c=5\pm2$ eV. The direction of the momentum transfer $\boldsymbol{q}$ ($\theta_q=45°$) is marked by the black square in the diagram; its size indicates the uncertainty in the direction of $\boldsymbol{q}$ resulting from the finite integration interval of $|\boldsymbol{q}|$. The angular range which is not affected by the detector dead-time is encircled by solid lines. (b) First Born CCC calculation. (c) Second Born CCC calculation.

giving rise to monopole or quadrupole transitions for which emission in opposite directions is allowed.

Second, a clear signature of multiple projectile-target interactions is the broken symmetry of the experimental cross section with respect to the momentum transfer direction. The invariance of the cross section for simultaneous inversion of

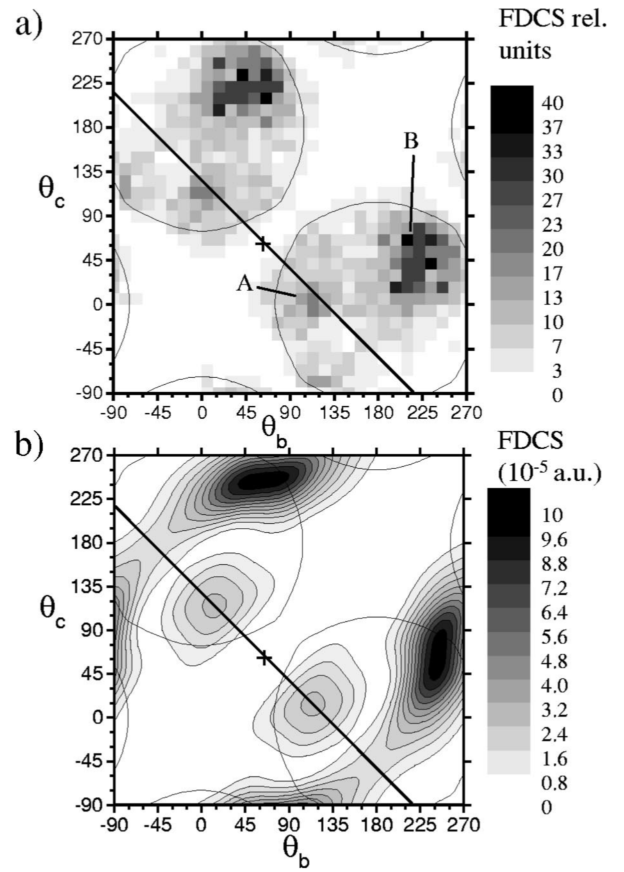

FIG. 2. As for Fig. 1 except $q=2\pm0.2$ a.u. (a) Experimental FDCS. (b) First Born CCC calculation.

both angles θ_b, θ_c with respect to the momentum transfer direction is characteristic of a single interaction of the projectile with the target. This invariance is maintained for the first Born calculation [Fig. 1(b)]. In the experiment [Fig. 1(a)] the peak B, the dominant structure of the cross section, violates strongly this symmetry with respect to the momentum transfer direction. Only the relatively weak peak A obeys this symmetry. It lies perfectly on the off-diagonal line (marked by the continuous line) which indicates configurations where both electrons are emitted at equal angles but to opposite sides with respect to the momentum transfer direction. It is obvious, therefore, from the shape of the experimental FDCS and its relation to the first Born calculation, that the second Born process is important for the present kinematics.

Indeed, including the second Born corrections into the calculation [Fig. 1(c)] modifies the FDCS radically. Agreement with experiment is improved concerning the relative intensity and the angular position of the peaks. The peak B is displaced away from the symmetry line, where it is located in the first Born calculation, into the direction of the experiment, albeit not far enough to be in perfect agreement. As for the peak A, it is shifted to an angular position not accessible experimentally. Nevertheless, the tail of the peak A, which is within the experimental acceptance, is consistent with the experimentally observed cross-section pattern.

In Fig. 2, we show the experimental (a) and theoretical (b)

FDCS for the same kinematics as in Fig. 1, but for $q=2$ a.u. As our second Born model is restricted to two low-q dipole interactions, it is not applicable to the present kinematics. However, there is no limitation to the first Born model. Examining the experimental FDCS, we observe that the relative peak intensities are largely maintained. On the other hand, peak B is more elongated in direction of θ_c and it is shifted by more than 40° to larger values of θ_c while the kinematical shift of the q direction is only 20°. The resulting pattern is consistent with the FDCS being fully symmetric with respect to the momentum transfer direction. This becomes obvious when making comparison with the first Born calculation, which obtains strong peaks for emission of one electron along the momentum transfer direction and the second going in the opposite way. Within the angular limits of the detector acceptance, the calculation is in very good agreement with the experiment. Despite the fact that peak B lies partially outside the experimental acceptance, a strong indication that it has the same elongation as the theoretical result is the observation of its tail for $\theta_b=-75°$, $\theta_c=120°$. Thus, from the good agreement with the first Born calculation, it can be concluded that for the impulsive high-q kinematics, single binary collisions of the projectile with the target are the main mechanism for two-electron ejection.

In summary, we have investigated the relative contribution of the first and second Born processes leading to the double ionization of He by electron impact at 500 eV. In the first Born process, the projectile interacts with the target only once and ejection of the two target electrons is possible solely due to electron correlations in the target before and after collision. In contrast, in the second Born process, ejection of the two target electrons happens sequentially as a result of two subsequent knock-outs of the projectile on the target. In general, both the first and second Born processes contribute to double ionization of He at an incident energy of 500 eV. However, the second Born contribution is insignificant for ionizing collisions with a large momentum transfer. This is because the Born amplitude drops off quickly with increasing momentum transfer, making two sequential collisions with large momentum transfer unlikely.

It is interesting to compare the role of the sequential processes in double and single electron impact ionization of He. In single ionization, simplification of the reaction dynamics at large q to a binary knock-out collision is well documented [27]. It is exploited in electron momentum spectroscopy (EMS) to extract the clearest information on the electronic structure of the target which is not obscured by a complicated reaction mechanism. Outside the EMS regime, at very large scattering angles and momentum transfer $q \gg 1$ the second Born effects may again become noticeable [28]. It is also well known that the generalized oscillator strengths for single ionization by electron impact at low momentum transfer converge towards the optical limit [29]. In this sense, photoionization can be viewed as a zero momentum transfer limit of electron-impact ionization. This is clearly not so for the double ionization as the electron impact ionization at low q is very likely to be affected by higher-order Born processes which are absent for double photoionization.

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