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Unexpected Higher-Order Effects in Charged Particle Impact Ionization at High Energies

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Most of the experimental and theoretical studies of electron-impact ionization of atoms, referred to as (e, $2e$), have concentrated on the scattering plane. The assumption has been that all the important physical effects will be observable in the scattering plane. However, very recently it has been shown that, for C^{6+}-helium ionization, experiment and theory are in nice agreement in the scattering plane and in very bad agreement out of the scattering plane. This lack of agreement between experiment and theory has been explained in terms of higher-order scattering effects between the projectile and target ion. We have examined electron-impact ionization of magnesium and have observed similar higher-order effects. The results of the electron-impact ionization of magnesium indicate the possible deficiencies in the calculation of fully differential cross sections in previous heavy particle ionization work.

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Remarkable progress has been made in the field of atomic collisions due to the development of cold-target recoil-ion momentum spectroscopy (COLTRIMS) [1]. COLTRIMS spectrometers (also called reaction microscopes) allow experimentalists to analyze the results of complex collisions in full detail by determining the final-state momenta of all collision fragments. Processes currently being studied fully differentially range from charged particle impact ionization of atoms [2] to fragmentation of large molecular targets [3]. Ionization of light atoms by heavy particle impact, which was once considered ''understood'' (at least for large collision energies), has seen a resurgence of interest and excitement resulting from the unveiling of new fundamental physics that challenges even the most sophisticated theoretical approaches.

One of the puzzling developments in recent work lies in the observance of an unexpected structure in the fully differential cross sections (FDCS) out of the scattering plane for heavy particle impact ionization of helium [2]. This structure is not predicted by theoretical approaches previously thought to be accurate. The comparison between experiment and theory at the measured kinematics were particularly surprising because a first-Born approximation (FBA) was expected to provide an adequate description of the ionization process (i.e. where higher-order contributions should have been negligible). The heavy particle work sparked questions about whether similar structures would also be seen for electron-impact ionization [normally called (e, $2e$)] and, Dürr *et al.* [4] have reported FDCS for electron-impact ionization of helium which were similar to the heavy particle FDCS.

The unexpected structure out of the scattering plane was attributed to a higher-order mechanism involving an interaction between the projectile and the residual target ion [projectile-ion (PI) interaction] [2]. However, such mechanisms are conceptually accounted for by the three-distorted wave (3DW) approach [2], where the PI interaction is described in the final-state wave function and interactions contained in the wave function are automatically contained to all orders of perturbation theory. Nevertheless, this model was not able to reproduce this structure. As a qualitative explanation for this failure of 3DW, it was argued that the final-state 3DW wave function is not accurate if all collision fragments are close to each other [5]. However, until now it was not clear how this shortcoming of the 3DW approach could be addressed.

More recently, van Boeyen *et al.* reported new data for high-energy electron-impact ionization of Mg [6]. As we will demonstrate in this Letter, a theoretical analysis of these data can shed some light on why the 3DW model failed to predict the experimentally observed out-of-plane structure. van Boeyen *et al.* measured the FDCS for a cone centered on the momentum transfer vector $\mathbf{Q} = \mathbf{k}_0 - \mathbf{k}_f$, where $\mathbf{k_0}$ ($\mathbf{k_f}$) is the initial (final) momentum of the projectile. The fast scattered electron is detected at a fixed scattering angle of 20° and 8 detectors are positioned on a cone of half angle 45° centered on $\mathbf{Q}$ as shown in Fig. 1. Two of the eight detectors (0° and 180° detectors) are in the scattering plane and the remaining detectors are above and below the scattering plane as can be seen in Fig. 1.

We calculated the FDCS for the geometry studied by van Boeyen *et al.* using the first order distorted wave Born

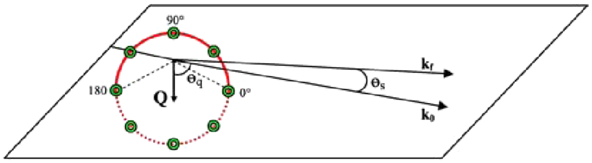

FIG. 1 (color online). Experimental scattering geometry with a view of detectors around the momentum transfer vector, **Q** (direction of **Q** ~ 70° from $\mathbf{k_0}$).

approximation (DWBA), the second-order distorted wave Born approximation (DWB2), and the 3DW approximation. The details of these calculations may be found in Chen and Madison [7] (DWBA and DWB2) and Prideaux *et al.* [8] (3DW). Here we only note that the DWB2 calculation does not use the closure, or other approximations to simplify the evaluation of the second-order term. Instead, a full set of intermediate pseudo states was summed over in the second-order term.

The DWBA model accounts for the PI interaction to all orders of perturbation theory. However, the projectile-ejected-electron (PE) interaction is contained only to first order. The DWB2 also accounts for the PI interaction to all orders of perturbation theory and additionally accounts for the PE interaction to second order. Finally, the 3DW approach treats both the PI and the final-state PE interaction (often called postcollision interaction between the projectile and ionized electron—PCI) to all orders. However, in the 3DW model, the PI interaction is described in terms of a Coulomb wave in the final state with an effective target charge of +1, independent of distance between the projectile and the residual target ion, whereas the DWBA solves a numerical Schrödinger equation for a Hartree-Fock potential which corresponds to a variable effective charge ranging from the nuclear charge at close distances to a charge of +1 asymptotically.

In Fig. 2, the experimental and theoretical FDCS are compared for single ionization of the 3*s* shell ($I_{3s} =$ 7.646 eV) of Mg, where the solid line represents the DWBA calculation, the dotted line the DWB2 results, and the dashed line the 3DW calculation. The six different panels correspond to the six different incident electron energies measured by van Boeyen *et al.* [6]. For this geometry, the FDCS above and below the scattering plane are symmetric so only the cross sections from the scattering plane and above are shown.

Although the experimental data are not absolute, the relative cross sections for different energies are absolute, such that a single normalization is required for all the data sets and we have chosen to normalize the experiment to the DWBA calculation at 90° for 1.0 keV incident energy. The overall agreement between the DWBA and experiment is quite good for all the energies except the lowest. The DWB2 results are included to test higher-order contributions from PE. We see very little difference between DWBA and DWB2 and the two calculations are essentially the same by 1.0 keV indicating that second-order PE interactions are not very important for these energies. As expected, the biggest difference between the two theories is seen for 400 eV and (for this normalization) the DWBA is unexpectedly closer to the experiment.

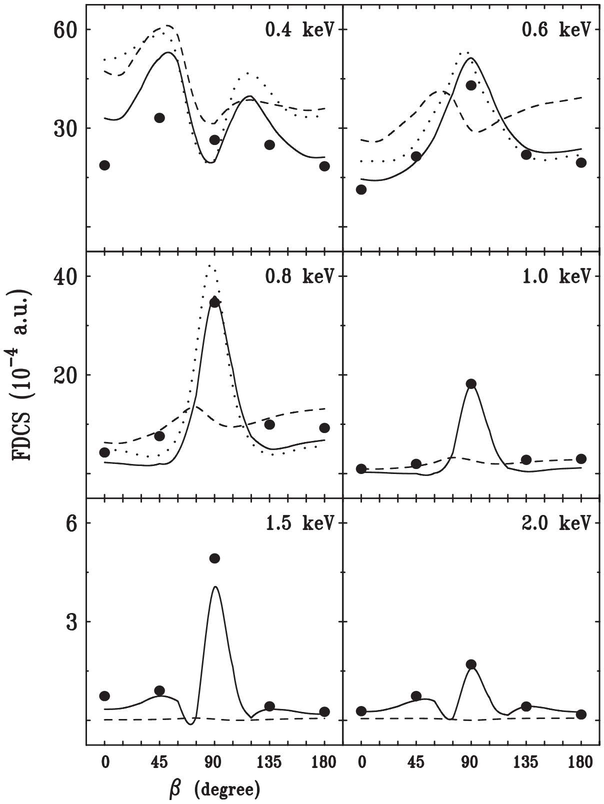

FIG. 2. Fully differential cross section as a function of position on the cone of Fig. 1 for (*e*, 2*e*) ionization of Mg (3*s*). The energy of the incident electron is noted in the upper right corner of each figure. The fast electron is observed at 20° and the energy of the ejected electron is 62 eV. The solid circles are the experimental data of van Boeyen *et al.* [6]. The theoretical curves are solid—DWBA, dotted—DWB2, and dashed—3DW.

In the FBA, the FDCS must be cylindrically symmetric about **Q** which means that the cross section should be constant on a cone centered on **Q**(corresponding to a flat curve in Fig. 2). One of the intriguing aspects of Fig. 2 is the fact that the experimental cross sections are not flat and that the largest cross sections are found at 90°, which lie in a plane perpendicular to the scattering plane (i.e., here the departure of the ejected electron from the scattering plane maximizes). This peak structure is reproduced by both the DWBA and the DWB2 calculations, but not by the 3DW results.

The measurements of van Boeyen *et al.* [6] are reminiscent of the COLTRIMS experiments for ion impact ionization which have produced similar out-of-plane structures (although in different geometry) that so far have not been theoretically explained [2]. Since the COLTRIMS measurements produce the full 4π angular distribution for the ejected electron, it is possible to extract cross sections for the same cone geometry measured in the van

Boeyen *et al.* experiment. Figure 3 contains the FDCS results for 24 MeV and 1.2 GeV C^{6+} ionization of helium on the van Boeyen cone. Indeed, for both projectile energies, a peak structure at 90° similar to that seen by van Boeyen *et al.* [6] is found.

For heavy particle scattering, a DWBA calculation has never been done due to the numerical difficulties associated with calculating distorted waves for a heavy projectile with extremely short wavelengths. Consequently, analytic Coulomb waves are typically used instead of distorted waves for the projectile. Nonetheless, the 3DW approach [5,9,10] for heavy particles has been considered to be the equivalent of the DWBA for (e, $2e$).

There are two important differences between the DWBA and 3DW. Analytic Coulomb waves are used for the final state of the projectile in the 3DW while distorted waves are used in the DWBA. Second, the 3DW contains PCI to all orders of perturbation theory while PCI is only included to lowest order in the DWBA. However, for the kinematics of these experiments, PCI is not very important, which is also seen by the very small differences we find between the DWBA and DWB2 calculations (see above). The most important difference between the DWBA and 3DW lies in the treatment of the projectile wave function. The 3DW is the fully quantum mechanical equivalent of the standard CDW (continuum distorted wave) approach which has been successfully used for many years for heavy particle scattering [see Refs. [5,10]].

From Fig. 3, it is seen that, similar to the case of electron-impact ionization, the 3DW results do not reproduce the structure at 90° and, in fact, are almost constant like the FBA would predict. Since the important difference between the DWBA and 3DW lies in the treatment of the projectile wave function, this suggests that the failure of the theoretical models for out-of-plane heavy particle scattering can be attributed to a poor representation of the heavy projectile wave function.

It is surprising that this change in the projectile wave function can have such a dramatic effect on the results. As mentioned above, the effective charge in the 3DW Coulomb wave for the projectile is unity at all distances from the ion. A distorted wave, on the other hand, can be regarded as a solution of Schrödinger's equation for a radially dependent effective charge that is determined by the screening of the nucleus by the electron charge cloud. These effective charges are shown in Fig. 4 for Mg. The solid line is the effective charge obtained from the Hartree-Fock charge density for a magnesium ion and the long-dashed–short-dashed line is for a single positive charge. From the figure, it is seen that the two different effective charges are identical beyond approximately $4a_0$. Consequently the structure in the perpendicular plane for electron-Mg scattering must be determined by the projectile penetrating closer than 3–$4a_0$ from the nucleus.

For heavy particle scattering, the 3DW results are generally in reasonably good agreement with absolute experiment in the scattering plane. Although van Boeyen *et al.* [6] did not measure complete angular distributions for the scattering plane; it is instructive to examine DWBA and 3DW results in the scattering plane for electron-impact ionization. Figure 5 contains in-plane DWBA and 3DW results for (e, $2e$) ionization of magnesium ($3s$) for the same incident energies as measured in the cone geometry experiment.

The large peak in Fig. 5 is called the binary peak since it corresponds to a classical collision between the projectile and an electron at rest. For the lower energies, the two theories yield very similar results. However, there is a drastic drop in the 3DW binary peak starting at 1.5 keV (i.e., the 3DW becomes an incorrect approximation). Classically, for a projectile to be scattered to the same angle with increasing energy requires decreasing impact parameters. If we use Rutherford scattering to estimate the classical impact parameter, we find an impact parameter of approximately $3.5a_0$ for 1 keV and approximately $1.5a_0$ for 1.5 keV. From Fig. 4, we see that the 3DW fails when a classical particle would penetrate into the region where the Hartree-Fock effective charge deviates from

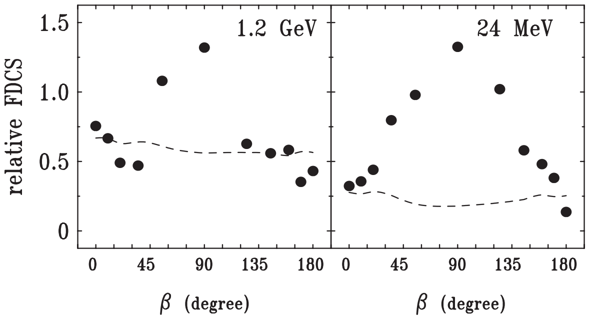

FIG. 3. Fully differential cross section as a function of position on the cone of Fig. 1 for 24 MeV and 1.2 GeV C^{6+} ionization of He. The solid circles are the experimental data of Refs. [9,10]. The energy of the ejected electron is 62 eV, the momentum transfer is 0.75 a.u. (left panel) and 1.5 a.u. (right panel) and the theoretical curves are 3DW results.

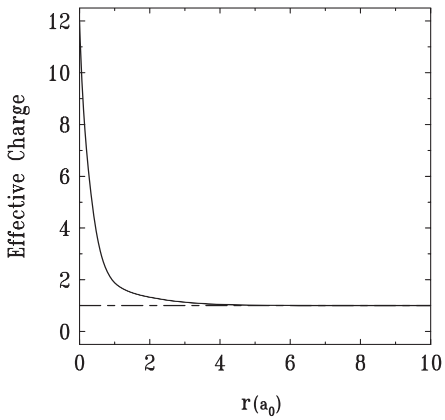

FIG. 4. Effective charges seen by the projectile for ionization of magnesium.

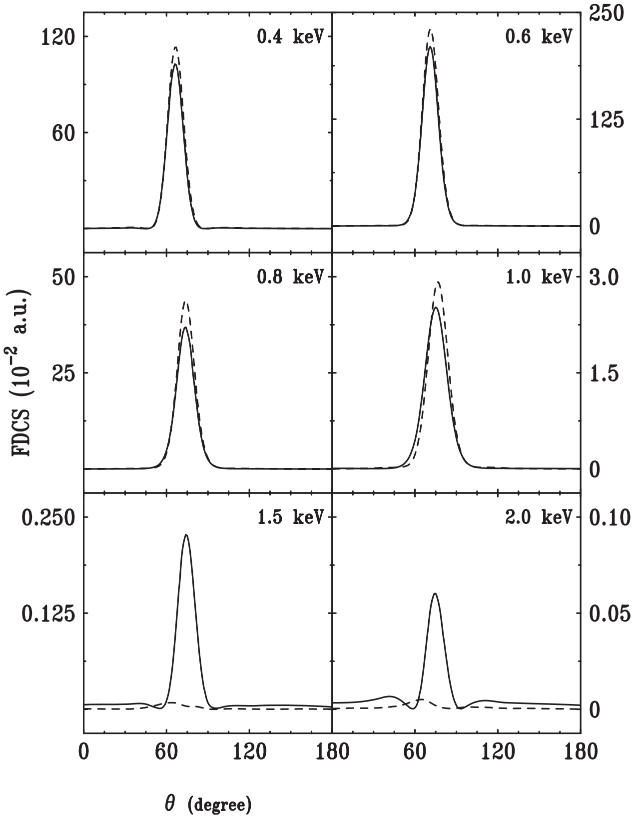

FIG. 5. Fully differential cross section for $(e, 2e)$ on Mg $(3s)$ in the scattering plane. The theoretical results are DWBA (solid) and 3DW (dashed).

unity. Consequently, we would conclude that a theoretical approach using Coulomb potentials will fail if the projectile penetrates closer than about $2a_0$ to the nucleus. These results are also consistent with the conclusions obtained by Madison *et al.* [5].

In conclusion, we have examined out-of-plane FDCS for electron-impact ionization of magnesium. The overall agreement between the first order DWBA and experiment was very satisfactory. When results for the same geometry were examined for heavy particle ionization, the experimental results were similar, but the agreement between theory and experiment was bad. This disagreement was attributed to a poor treatment of small impact parameter collisions for heavy projectiles. The results of this work show why the PI interaction is so dramatically underestimated by the 3DW model for any structure resulting from close collisions with the nucleus. By describing that interaction in terms of a Coulomb wave, the effective target charge is increasingly underestimated with increasing projectile energy. This also explains why the observed out-of-plane structures, which are due to higher-order effects, become more pronounced with increasing energy while it is generally assumed that higher-order effects should become less important with increasing energy.

In summary, a solid understanding of the electron-impact ionization process has emerged in recent years, at least for ionization of hydrogen and helium. For low-energy incident electrons, experimental data are well reproduced by nonperturbative methods [11,12] and at large energies by perturbative distorted wave approaches. For ion impact, the situation is much less satisfactory and experiment and theory are not in accord even for very high incident energies. In recent years, experimental evidence has accumulated which suggests that the problem with the theoretical treatment for ion impact lies in the description of the PI interaction [13]. Here, we demonstrated that a distorted wave treatment of this interaction is needed. Although a distorted wave treatment for heavy ions has not been accomplished yet, extrapolating the rapidly increasing computer capabilities to the future would suggest that it should not be too long before such calculations become possible.

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