Exchange distortion and postcollision interaction for intermediate-energy electron-impact ionization of argon

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Measurements of fully differential cross sections for electron impact ionization of atoms have been performed for over 30 years. However, only within the last ten years has agreement between experiment and theory been achieved for ionization of hydrogen and helium. For the heavier inert gases, reasonably good agreement between experiment and theory has only been achieved for high incident energies while serious discrepancies are common for intermediate and low incident energies. It is believed that a major source of the problem stems from an improper/inadequate treatment of exchange distortion (ED) and the effects of post-collision interactions (PCIs). In this paper, two different methods for including ED are examined—one based upon the *R* matrix (close-coupling) approach and one originating from the single-configuration Hartree-Fock approach. In general, these two methods predict significant, but different, ED effects. The importance of PCI is studied by including the final-state Coulomb interaction directly in the final-state wave function. This procedure guarantees that PCI effects will be included to all orders of perturbation theory. For intermediate energies, PCI is an important effect and leads to an overall improvement in the agreement between experiment and theory.

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I. INTRODUCTION

Fully differential cross sections for electron impact ionization have been studied since Ehrhardt *et al.* [1] and Amaldi *et al.* [2] published the first experimental cross sections that resolved all continuum electron momenta. In doing so, they began work that continues nearly 35 years later. The first-order distorted-wave Born approximation (DWBA) yielded good agreement with experiment for high energies [3–6]. For low incident-electron energies, it took until the 1990s before experiment and theory were in accord even for the simplest electron-hydrogen scattering problem [7–12]. For the case of electron-hydrogen scattering, the DWBA is relatively good for energies greater than about 200 eV [13–15]. One of the problems with the DWBA lies in the fact that the final-state electron-electron interaction [the so-called "post-collision interaction" (PCI)] is neglected in the formation of the final-state wave function. Brauner *et al.* [16] showed that good agreement could be achieved down to around 100 eV incident energy if PCI was included directly in the final-state wave function. Jones and Madison [15] showed that the good-agreement limit could be lowered to around 50 eV if initial-state projectile-atom interactions were also included in the wave functions. For ionization of heavier atoms, the DWBA gives relatively good agreement with experiment above about 200 eV [17–24]. However, experiment and theory are still not in accord for lower incident-energy ionization of heavy atoms.

Some experiments have been performed around and below the 200 eV limit for heavier atoms (Hong and Beaty [25]: 100 eV Ar; Khajuria *et al.* [26]: 64 eV He and Ar; Haynes and Lohmann [27–29], and Haynes *et al.* [30]: 35.5 eV to 197.5 eV Ar and Kr). Since including PCI in the final-state wave function was important for electron-hydrogen scattering, it seems logical that including PCI in the DWBA should provide better agreement with experiment for lower incident-electron energies. Prideaux and Madison [31] reported the first DWBA calculation with PCI included in the final-state wave function. The model was labeled 3DW since the final-state wavefunction includes two distorted waves plus the electron-electron Coulomb distortion factor. The 3DW results for *s*-shell ionization of argon and krypton by electron impact (Haynes *et al.* [30], Prideaux and Madison [31]) showed improved agreement with experiment for incident energies of 113.5eV. In the present paper, the 3DW approach is extended to ionization of *p* shells. As an example, ionization of the $3p$ shell of argon is examined for 113.5 eV incident electrons [29] and 200 eV incident electrons [32].

A second physical effect that can become important for low-energy electrons results from electron exchange. For atomic ionization, there are two different types of exchange that can take place: (1) the two final-state continuum electrons can exchange with each other and (2) any of the continuum electrons can exchange with an atomic electron. The second type of exchange is sometimes referred to as "exchange distortion" (ED) since this exchange modifies the differential equation for the continuum electron and "distorts" the continuum electron wave function. It has been shown that ED plays a key role in understanding spin-asymmetry

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measurements for atomic ionization [33], [34]. The proper way to include ED in a DWBA calculation would be to perform a Hartree-Fock calculation for the continuum waves. However, such a procedure is sufficiently challenging, so that the nonlocal Hartree-Fock distorting potential is often approximated by the spherically symmetric local potential of Furness and McCarthy [35] (FM). We use this approximation in the 3DW approach.

A method that includes ED properly for the *slower* of the two final-state electrons only is the hybrid distorted-wave plus *R*-matrix approach for atomic ionization. It was originally developed by Bartschat and Burke [36], and a general computer code to calculate angle-integrated cross sections was published by Bartschat [37]. The basic idea is to describe a "fast" projectile electron by a distorted wave, but to treat the initial *N*-electron bound state and the interaction between the residual $(N-1)$-electron ion and the "slow" ejected electron by an *R*-matrix (close-coupling) expansion. Since this expansion uses fully antisymmetrized *N*-electron wave functions, exchange effects between all these *N* electrons are treated in a computationally exact manner, but exchange with the projectile is neglected. During the past decade, the code has been further developed and was applied successfully to angle-differential ionization and ionization-excitation of helium, including even second-order effects between the projectile and the target (see, for example, Refs. [38], [39], and references therein.) Since the most important ED effect between the slow ejected electron and the ion is treated properly, one may expect this method to do well if the excess energy is shared in a highly asymmetric way between the two final-state electrons. We will use both the FM and *R*-matrix approaches to examine the importance of ED.

II. THEORY

A. The 3DW approach

The three distorted wave (3DW) approach has been previously discussed by Haynes *et al.* [30] and Prideaux and Madison [31]. Here we present a brief overview with the key features. The 3DW *T* matrix is given by

$$T_{fi}^{\mathrm{3DW}} = \langle \chi_f \chi_{\mathrm{eject}} C_{\mathrm{proj\text{-}eject}} | V - U | \psi_{\mathrm{active}} \chi_i \rangle. \tag{1}$$

Here ψ_{active} is the initial bound-state wave function for the active electron, $\chi_i(\chi_f)$ is the initial (final) state distorted wave for the projectile electron, χ_{eject} is the final state distorted wave for the ejected electron, $C_{\mathrm{proj\text{-}eject}}$ is the Coulomb interaction between the projectile and ejected electron, V is the initial-state interaction between the projectile and neutral atom, and U is a spherically symmetric approximation for V. Brauner *et al.* [16] demonstrated that it was very important to include the final-state Coulomb interaction between the two continuum electrons $C_{\mathrm{proj\ eject}}$ [the so-called "post-collision interaction"(PCI)] in the approximation for the final-state wave function. The standard distorted wave (DW) approximation does not include this interaction, i.e.,

$$T_{fi}^{\mathrm{DW}} = \langle \chi_f \chi_{\mathrm{eject}} | V - U | \psi_{\mathrm{atom}} \chi_i \rangle. \tag{2}$$

The initial-state distorted wave is a solution of the Schrödinger equation

$$(T_{\mathrm{proj}} + U - k_i^2)\chi_i(r) = 0. \tag{3}$$

Here T_{proj} is the kinetic energy operator for the projectile and k_i^2 is the energy of the incident electron. The distorting potential U is given by

$$U = U_i + U_{\mathrm{ex}}, \tag{4}$$

where U_i is the Hartree-Fock potential for the neutral atom and U_{ex} is a local approximation for the exchange potential for the projectile-electron exchanging with all the atomic electrons [normally refereed to as the exchange distortion (ED) potential]. We use the triplet form of the Furness and McCarthy [35] (labeled FM) approximation for U_{ex} where the charge density is half the full density for the atom (since the projectile can only exchange with an atomic electron of identical spin). This procedure gave good agreement with experiment for *p*-shell ionization of xenon [33,34]. On the other hand, it should be noted that the accuracy of local approximations for exchange distortion for *p* shells has been questioned [40,41].

Both final-state distorted waves are solutions of Schrödinger equations similar to Eq. (3), except that the Hartree-Fock atomic neutral potential U_i is replaced with the Hartree-Fock potential for the final-state ion U_{ion}. The final-state distorted waves are orthogonalized to ψ_{atom} using the Gram-Schmidt procedure. Finally, as described by Prideaux and Madison [31], if the ionization event is treated as a three-body problem, the perturbation can be approximated as

$$V - U = -\frac{1}{r_{ab}} - U_{\mathrm{active}}, \tag{5}$$

where r_{ab} is the distance between the two electrons and U_{active} is the spherically symmetric potential for the electron-electron interaction.

B. Hybrid distorted-wave+*R*-matrix approach

This method is based upon the formalism outlined by Bartschat and Burke [36] and the computer program RMATRX-ION of Bartschat [37]. As mentioned previously, the basic idea is to describe a "fast" projectile electron by a distorted wave and then calculate the initial bound state and the interaction between the residual ion and the "slow" ejected electron by an *R*-matrix (close-coupling) expansion.

In the present work, we used a first-order distorted-wave representation for the projectile and a two-state close-coupling approximation for electron scattering from Ar^+, coupling only the ionic ground state $(3s^2 3p^5)^2P^0$ and the first excited state $(3s3p^6)^2S$. This hybrid model will be referred to as RM2 below. The ionic target description is the one used first by Burke and Taylor [42] for the corresponding photoionization problem and later by Bartschat and Burke [43] in the calculation of single-differential (with respect to energy loss) and total ionization cross sections of argon by electron impact.

The distortion potential for the fast projectile was chosen as the static ground-state potential of neutral argon. Exchange distortion between the projectile and the target was ignored. In addition to the exact treatment of exchange be-

tween the ejected electron and the residual ion, this model accounts for a small amount of channel coupling, and it also yields an accurate description of the ionic structure and the initial atomic bound state. While the latter improvements over a single-configuration Hartree-Fock (SCHF) description are not unimportant, test calculations with a one-state R-matrix expansion to describe e-Ar^+ collisions and an SCHF Hartree-Fock representation of the $Ar^+(3s^2 3p^5)^2P^0$ final state produced very similar results to the ones shown below.

Partial waves up to orbital angular momenta of 90 for the fast projectile guaranteed the convergence of its partial wave expansion. Furthermore, the Coulomb interaction between the fast projectile and the target was accounted for through multipole components $\lambda = 0-5$.

III. RESULTS

A. Exchange distortion

As discussed in the previous section, the RM2 model is similar to the DWBA approach with the slow ejected electron being represented by a close-coupling wave function. The additional physics contained in the close-coupling wave function is channel coupling and exchange distortion. However, test calculations revealed that the channel-coupling effects are small for the problem dealt with in the present paper, namely, $3p$ ionization of argon. As a result, RM2 is primarily a DWBA calculation with exact exchange distortion (ED) for the ejected electron. Although RM2 does not contain ED for the fast projectile, these effects are expected to be small for the initial and final projectile energies under consideration. This was verified by running the DWBA code with FM exchange for all three continuum electrons and FM exchange just for the ejected electron. Very little difference was found in the results.

In Fig. 1, the experimental data of Haynes and Lohmann [29] are compared with first-order DWBA (labeled DW), DWBA plus FM exchange distortion (labeled DW-FM) and RM2 predictions. Since the experimental data are not absolute, we chose to normalize experiment and all theories to the DW results at the first maximum (the so-called "binary peak"). These results are in the scattering plane with the z-axis parallel to the incident beam direction. The faster final-state electron is measured at 15° counterclockwise when viewed from above the scattering plane while the observation angle θ_b for the slower electron is measured clockwise. In this reference frame, the detector for the faster electron is located at $\theta_b = 345°$. The energy of the ejected electron is noted in each panel.

The most striking aspect of Fig. 1 is the fact that the two different ED methods produce very different types of changes when compared to DW. At the highest ejected-electron energy of 10 eV, FM yields an improved binary peak, which is in very good agreement with experiment—both in shape and location of the maximum. With decreasing energy, the FM results for the binary peak become increasingly worse while the RM2 results become increasingly better. At the lowest energy, there is a double binary peak which is not predicted by FM. For the recoil peak, the original DW

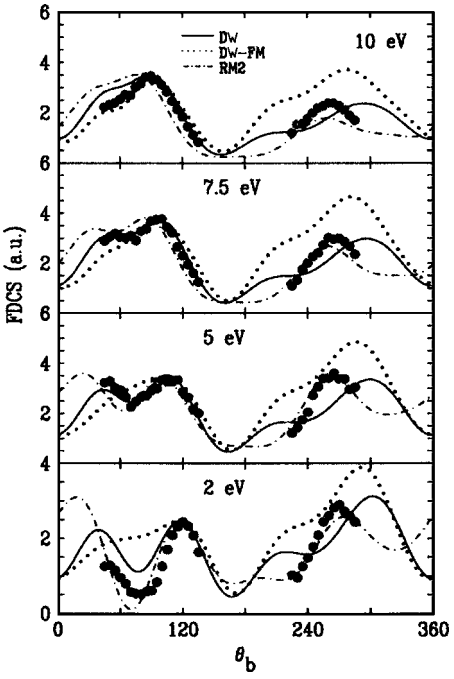

FIG. 1. Fully differential cross sections (in atomic units) for 113.5 eV electron impact ionization of the $3p$ shell of argon in the scattering plane. The experimental data of Haynes and Lohmann [29] and all theories are normalized to the DW results at the first maximum. The horizontal axis θ_b (in degrees) corresponds to the slower electron observation angle while the faster final-state electron is observed at $-15°$ ($\theta_b = 345°$). The energy of the ejected electron is noted in each panel. The normalization factors, by which the various results have been multiplied, are as follows: DW-FM [0.5 (2 eV); 1.12 (5 eV); 1.8 (7.5 eV); 2.4 (10 eV)] and RM2 [2.0 (2 eV); 2.1 (5 eV); 1.65 (7.5 eV); 1.22 (10 eV)].

results have about the correct magnitude (relative to the binary peak) for all energies. The only problem is that the maximum is shifted to higher angles by about 30°. FM ED shifts the recoil peak to smaller angles by only about 10° while also increasing the magnitude of the recoil peak. RM2 ED effects, on the other hand, shift the recoil peak by the proper amount while generally decreasing the magnitude slightly. Consequently, the RM2 results for the recoil peak are in quite good agreement with experiment. Overall the RM2 results are better for the lower energies.

Figure 2 contains a similar comparison between experiment and theory for the higher incident energy of 200 eV. Here the experimental data are those of Stevenson *et al.* [32]. These data are not absolute either, and hence we have used the same normalization procedure as before. Similar to Fig. 1, DW-FM yields the best agreement with the shape of the binary peak for the highest ejected-electron energy (20 eV) but the worst agreement for the lowest energy (2 eV). RM2 is in very good agreement with the double binary peak for 10 and 2 eV, but strangely not as good for 5 eV. In contrast to Fig. 1, RM2 is now in good agreement with the experimental recoil peak for the higher energies but not the lower energies. Somewhat surprisingly, the relative DW-FM peak heights are now closer to experiment for the lower energies. Consequently, a general rule of thumb for what one might expect

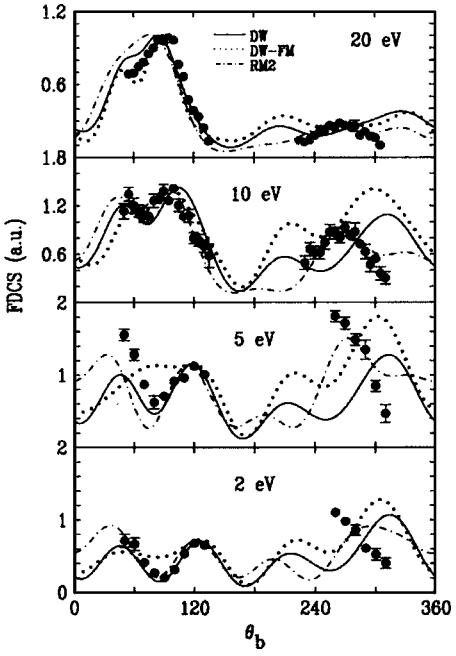

FIG. 2. Fully differential cross sections (in atomic units) for 200 eV electron impact ionization of the $3p$ shell of argon in the scattering plane. The experimental data of Stevenson *et al.* [32] and all theories are normalized to the DW results at the first maximum. The horizontal axis θ_b (in degrees) corresponds to the slower electron observation angle while the faster final-state electron is observed at $-15°$ (θ_b=345°). The energy of the ejected electron is noted in each panel. The normalization factors are as follows: DW-FM [0.43 (2 eV); 1.0 (5 eV); 1.85 (10 eV); 1.9 (20 eV)] and RM2 [1.85 (2 eV); 2.2 (5 eV); 1.2 (10 eV); 0.74 (20 eV)].

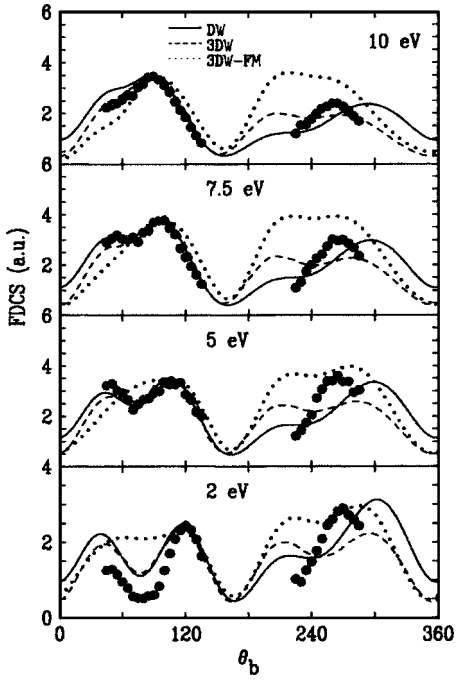

FIG. 3. Fully differential cross sections (in atomic units) for 113.5 eV electron impact ionization of the $3p$ shell of argon in the scattering plane. The experimental data of Haynes and Lohmann [29] and all theories are normalized to the DW results at the first maximum. The horizontal axis θ_b (in degrees) corresponds to the slower electron observation angle while the faster final-state electron is observed at $-15°$ (θ_b=345°). The energy of the ejected electron is noted in each panel. The normalization factors are as follows: 3DW [1.5 (2 eV); 1.5 (5 eV); 1.8 (7.5 eV); 1.7 (10 eV)] and 3DW-FM [0.75 (2 eV); 2.0 (5 eV); 3.2 (7.5 eV); 4.0 (10 eV)].

for the different treatments of ED is elusive. However, these results do indicate that the FM approach is not sufficiently accurate for p vacancies, in spite of the fact that it does get some of the physics at least qualitatively correct.

B. Post-collision interaction (PCI)

One of the problems associated with the standard DWBA approach lies in the fact that the post-collision repulsion between the two continuum electrons is contained only to first order in perturbation theory. Brauner *et al.* [16] demonstrated that this interaction is important for intermediate-energy electrons ionizing hydrogen. When the electron-electron repulsion is included directly in the final-state wave function as described in the theory section, this interaction is automatically contained to all orders of perturbation theory. We label the distorted-wave results with the Coulomb interaction included in the final-state wave function as 3DW. We can also add ED to this model in the same manner (FM) as discussed above for the standard DW approach. Unfortunately, we cannot add PCI to the RM2 treatment at this time.

In Figs. 3 and 4, DW, 3DW, and 3DW-FM results are compared with experiment for 113.5 and 200 eV ionization of the $3p$ shell of argon. Logically, electron-electron repulsion should move the binary peak to the right and the recoil peak to the left. For the case of $3s$ ionization of argon, Prideaux and Madison [31] found that the DW binary peak was at too small an ejection angle. In that case, PCI shifted the binary peak to the right such that very nice agreement with experiment was achieved. The recoil peak was at too large an ejection angle. Although PCI shifted the peak to the left, it was not enough to achieve agreement with experiment. For the case of p-shell ionization shown in Figs. 3 and 4, the binary peak positions are not shifted to the right as one might have expected. However, this is good since the original DW binary peak positions were already correct. For the higher ejected-electron energies, PCI slightly changes the shape of the binary peaks and thereby improves agreement with experiment. Similar to Figs. 1 and 2, FM ED produces incorrect shapes for the binary peak with decreasing ejected-electron energy. In the recoil region, PCI shifts the recoil peak to the left. Again, this is a good trend, but the shift is not large enough to achieve agreement with experiment (similar to s-state ionization). It is encouraging to note that FM ED produces a further left shift in the recoil peak. This brings the peak position closer to experiment, and it also tends to produce magnitudes qualitatively similar to experiment. This suggests that improved agreement with experiment might be achieved by combining a proper treatment of PCI with an improved treatment of ED.

IV. CONCLUSIONS

We have investigated the importance of exchange distortion (ED) and post-collision interactions (PCI) for electron

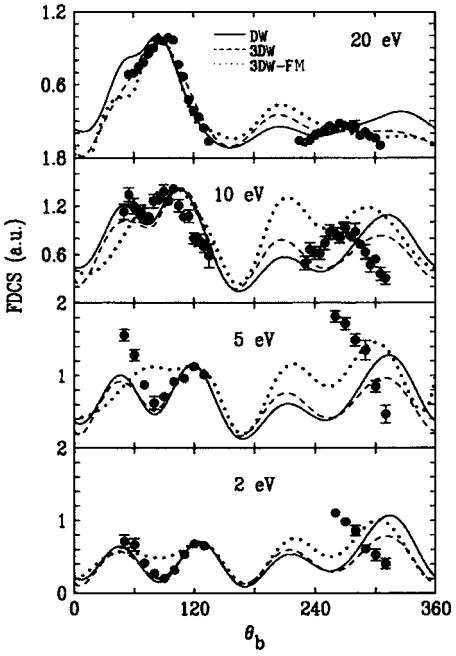

FIG. 4. Fully differential cross sections (in atomic units) for 200 eV electron impact ionization of the 3*p* shell of argon in the scattering plane. The experimental data of Stevenson *et al.* [32] and all theories are normalized to the DW results at the first maximum. The horizontal axis θ_b (in degrees) corresponds to the slower electron observation angle and the faster final-state electron is observed at $-15°$ ($\theta_b=345°$). The energy of the ejected electron is noted in each panel. The normalization factors are as follows: 3DW [1.13 (2 eV); 1.17 (5 eV); 1.33 (10 eV); 1.4 (20 eV)] and 3DW-FM [0.5 (2 eV); 1.21 (5 eV); 2.6 (10 eV); 2.6 (20 eV)].

impact ionization of the 3*p* shell of argon. For ED, we used two different approaches—the local potential approximation of Furness and McCarthy [35] and the *R*-matrix approach, which uses a close-coupling wave function for the ejected electron. Although it was anticipated that the two different ED approaches should yield similar results, this was not the case. For the ejected-electron energies considered here, the exact RM2 treatment of ED was far superior to the approximate treatment via the FM approach. However, RM2 seems to have some difficulties with the shape and location of the binary peak for high-energy ejected electrons and with the recoil peak for low-energy ejected electrons for the case of 200 eV incident electrons. These problems are probably related to the fact that PCI is neglected in the RM2 approach. In any case, it seems clear that the FM approach is not sufficiently accurate for *p*-shell ionization. On the other hand, this approximation does produce results that tend to be at least qualitatively correct. Interestingly, the FM approach produced much improved results for spin-asymmetry measurements for ionization of the *p* shell of xenon [33,34].

For *p*-shell ionization, PCI did not produce the expected shift to the right in the binary peak (which was good). However, it did slightly change its shape and thus produced better accord with experiment for ejected electrons with relatively high energy. PCI had a larger effect on the recoil peak than the binary peak. In general, PCI tended to reduce the magnitude of the recoil peak and slightly shifted the peak position to the left. Interestingly, if PCI had the same effect on the RM2 results, worse agreement with experiment would be produced. Overall, PCI produced an improved agreement with experiment. This suggests that good agreement with experiment can probably be achieved when proper treatments of PCI and ED are combined. Finally, the theoretical calculations predict a double-peak structure for both the binary and recoil peaks for low-energy *p*-shell ionization. However, the currently available experimental results do not cover a sufficiently large angular range in the recoil region to check the double-peak prediction.

The results of the present study clearly indicate the need for more experimental data for this kind of collision systems, in order to enable computational approaches to be developed that might ultimately match the recent success stories regarding ionization of atomic hydrogen and helium. In light of the richer structure predicted theoretically and already partially observed experimentally for *p*-shell ionization of heavy noble gases, it would be highly desirable to have experimental data over the entire angular range. Finally, *absolute* data would help to further discriminate between various theoretical approaches, since often significant differences are found in the predicted magnitudes of the various peaks.

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