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Theoretical and experimental investigation of $(e,2e)$ ionization of argon $3p$ in asymmetric kinematics at intermediate energy

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The field of electron-impact ionization of atoms, or $(e,2e)$, has provided significant detailed information about the physics of collisions. For ionization of hydrogen and helium, essentially exact numerical methods have been developed which can correctly predict what will happen. For larger atoms, we do not have theories of comparable accuracy. Considerable attention has been given to ionization of inert gases and, of the inert gases, argon seems to be the most difficult target for theory. There have been several studies comparing experiment and perturbative theoretical approaches over the last few decades, and generally qualitative but not quantitative agreement is found for intermediate energy incident electrons. Recently a nonperturbative method, the B -spline R -matrix (BSR) method, was introduced which appears to be very promising for ionization of heavier atoms. We have recently performed an experimental and theoretical investigation for ionization of argon, and we found that, although the BSR gave reasonably good agreement with experiment, there were also some cases of significant disagreement. The previous study was performed for 200-eV incident electrons and ejected electron energies of 15 and 20 eV. The purpose of the present work is to extend this study to a much larger range of ejected electron energies (15–50 eV) to see if theory gets better with increasing energy as would be expected for a perturbative calculation. The experimental results are compared with both the BSR and two different perturbative calculations.

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I. INTRODUCTION

There has been a long history of interest in the problem of obtaining triple-differential cross sections (TDCS) for electron-impact ionization of atoms [called an $(e,2e)$ process] since the pioneering work of Ehrhardt and his collaborators [1–3]. One of the important reasons that measurements of TDCS have remained of interest for so many years lies in the fact that these experiments represent the most sensitive test of theoretical models since all kinematic parameters are determined (except for the spin). Consequently, accurate experimental measurements remain in demand for testing new theoretical developments.

In the early days of this work, the theoretical calculations were primarily first- or second-order distorted-wave (DWB1 or DWB2) [4–12] or first- or second-order R -matrix (DWB1-RM or DWB2-RM) calculations [13,14]. By the 1990s, computers became powerful enough to be able to perform nonperturbative calculations. Starting at that time the convergent close-coupling (CCC) approach [15–18], the exterior complex-scaling (ECS) approach [19–23], and the time-dependent close-coupling (TDCC) method [24–27] were applied to electron-impact ionization of hydrogen and helium. Excellent agreement was found between experiment and theory, so these two problems can be regarded as “solved.” However, the development of similarly “exact” nonperturbative methods for heavier atoms has proven to be very difficult. The most promising recent development seems to be the B -spline R -matrix (BSR) approach introduced by Zatsarinny and Bartschat [28–32]. Very good agreement between experiment and the BSR results was found for ionization of helium [29,30], and neon [28]. The agreement was not as good for ionization of Ar [32], although the principal problem lay with

the original experimental data, which were recently corrected [33] for 200 eV incident energy. Nevertheless, even after the correction (the same experimental problem occurred for the 71 eV data [32]), agreement between experiment and theory will be far from perfect.

Argon has been studied from the discharge point of view for more than 100 years, and there have been several $(e,2e)$ measurements made for argon over the past few decades. Groups in Australia and Orsay, France, have studied ejection of the $2p$ electrons for incident electron energies greater than 1 keV [34–36]. Lahmam-Bennani *et al.* [37] have presented measurements of absolute TDCS for ionization of the $3p$ electrons and Avaldi *et al.* [38] have shown that distorted-wave impulsive approximations satisfactorily described the TDCS at the Bethe ridge conditions. The Orsay group proposed a high incident energy (~ 720 eV) experiment, in which the incident electron energy loss is large and momentum transfer is small. Under this condition, the two outgoing electrons strongly interact with each other [36–39].

There are a few $(e,2e)$ experimental studies for argon in asymmetric geometry for intermediate energies. In this geometry, the postcollision interaction (PCI) and exchange effects can be very important, especially for slow ejected electrons. The first experimental study of argon at 100 and 250 eV incident energies was performed by Ehrhardt *et al.* [3] at asymmetric kinematics. The Australian group has presented a series of experimental studies on $3s$ and $3p$ ionization of argon at low to intermediate energies [40–43]. They have focused generally on an incident energy of 113.5 eV and low electron ejection energies, and they have compared the experimental data with the distorted-wave Born approximation (DWBA). Using the same kinematic conditions, a comparative study was made by Stevenson and Lohmann over an extended angular range using

a magnetic angle changer [44]. Recently, both experimental and theoretical investigations have concentrated on ionization of the outer $3p$ orbital of argon at an intermediate incident energy (200 eV) for asymmetric kinematics. These kinematics were chosen due to the anticipation that multiple competing interactions (such as PCI and exchange effects) will be important. Stevenson *et al.* [45] compared their measurements with the DWBA, the DWB1-RM, and DWB2-RM predictions, and they found good agreement with experiment for the high ejection energies, and large discrepancies for lower ejection energies. More recently, Ren *et al.* [32,33,46] reported measurements for incident energies of 195 and 70.8 eV. They found good agreement with the RM calculations even for lower incident energy of 70.8 eV. Finally, Hargreaves *et al.* [47] examined argon ($3p$) ionization, and they also found significant discrepancies between experiment and theory.

Last year, we reported an experimental and theoretical study of the ionization of the argon $3p$ orbital at 200 eV incident energy for asymmetric coplanar geometry, ejected electron energies of 15 and 20 eV, and three fixed scattered electron angles of 10° , 15° , and 20° [48]. DWB1-RM, DWB2-RM, and nonperturbative B -spline R -matrix (BSR) results were compared with experiment. Surprisingly, good agreement between the BSR calculation and experiment was found only for the smallest scattering angle (10°) with very significant differences for 20° . In fact, the DWB2-RM results gave better shape agreement with experiment than the BSR for the larger scattering angles. This is surprising since one would expect a perturbative approach to become less accurate with increasing angles (decreasing cross section) while a nonperturbative approach should not have this problem.

The purpose of the present paper is to further investigate this situation. The previous work represented an angular scan for two fixed ejected electron energies. Here we report an energy scan for two fixed scattering angles. Comparing results from perturbative and nonperturbative calculations, one would expect a perturbative calculation to get better with increasing energy while a nonperturbative calculation should not be affected by energy (as long as it is converged). The highest energy considered in the previous work was 20 eV and here we examine energies ranging from 15 to 50 eV for scattering angles of 10° and 15° . As mentioned above, it is expected that PCI and exchange effects are probably important for these energies. Consequently, in addition to the DWBA-RM and BSR calculations, we also compare with a three-body distorted-wave (3DW) calculation that includes PCI to all orders of perturbation theory.

II. EXPERIMENTAL APPARATUS

The experiments described here were performed using an electron spectrometer especially designed for electron-electron coincidence experiments in the e-COL laboratory, Afyon. A detailed description of the apparatus and its applications to ionization of He [49,50], Ar [48], and H₂ [51,52] targets is given in the references. As described previously [53], the electron spectrometer is comprised of an electron gun, two hemispherical electron analyzers, and a Faraday cup. A schematic diagram of the electron spectrometer and electron pulse handling system is shown in Fig. 1. The

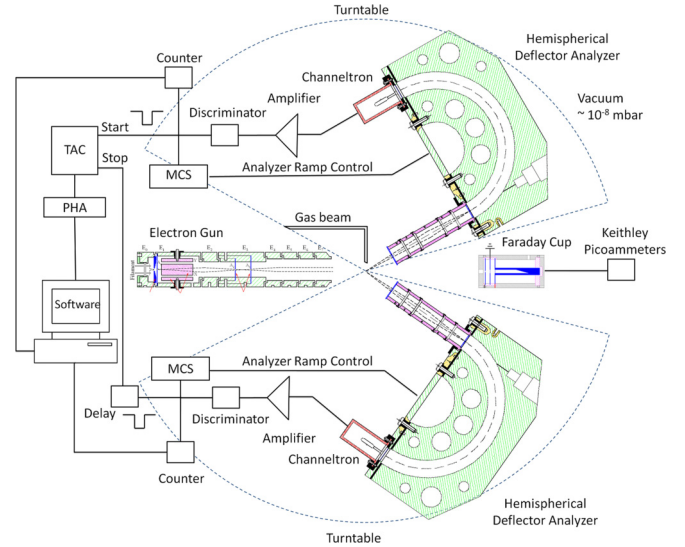


FIG. 1. (Color online) Schematic diagram of the coincidence electronics used to accumulate a coincidence timing spectrum at each kinematics.

spectrometer is contained in a cylindrical stainless steel vacuum chamber. The pressure in the chamber was maintained at $\sim 5.0 \times 10^{-6}$ mbar during data handling. This spectrometer operated at an electron current of $\sim 1 \mu\text{A}$ with a resolution of ~ 0.6 eV. The $(e,2e)$ technique is used to detect two outgoing electrons in coincidence after ionization of the target atom. The two electrons produced by single ionization of an atom are energy analyzed by hemispherical electron energy analyzers and detected by channel electron multipliers (CEMs), which are mounted on the hemispherical electron energy analyzers. This technique has an advantage for obtaining single ionization events for which the outgoing electrons have originated from the same ionization event. To do this, time correlation between the detected electrons is taken into account. The time delay between the electrons is converted to a signal that is recorded by computer, and a narrow coincidence peak in the timing spectrum is observed.

III. THEORY

We have used three different numerical methods to describe the process of interest. Each of them has been described previously. Hence we will only summarize them briefly to the extent necessary for the present discussion, but provide references where interested readers can find more information.

A. 3DW

The three-body distorted-wave (3DW) approach has been described in previous works, so we will just present the aspects of the theory necessary for the present discussion [54]. The T matrix can be written as

$$T_{fi}^{3DW} = \langle \Phi_f | W | \Phi_i \rangle, \quad (1)$$

where Φ_i and Φ_f are the initial- and final-state wave functions, respectively, and W is the perturbation.

In the 3DW approximation, the initial-state wave function Φ_i is approximated as a product of the initial bound state of the

atom (ψ_A) times a distorted-wave function χ_i for the incoming electron (the projectile),

$$\Phi_i = \Psi_A \chi_i. \quad (2)$$

For atoms, we use the Hartree-Fock bound-state wave function (ψ_{HF}) for the target. The perturbation (W) is given by

$$W = V - U_i. \quad (3)$$

Here V is the interaction between the incident electron and the atom, and U_i is the initial-state spherically symmetric static approximation for V , which is asymptotically equal to zero.

The final-state wave function Φ_f is approximated as a product of two final-state continuum electron distorted waves (χ_{scat} and χ_{eject}), and the Coulomb interaction between the outgoing electrons ($C_{\text{ele-ele}}$), normally called the postcollision interaction (PCI),

$$\Phi_f = \chi_{\text{scat}} \chi_{\text{eject}} C_{\text{ele-ele}}. \quad (4)$$

We use the exact postcollision Coulomb interaction between the two electrons ($C_{\text{ele-ele}}$), which is equal to a Gamov factor times a hypergeometric function,

$$C_{\text{ele-ele}}(\mathbf{r}_{12}, \mathbf{k}_{12}) = \Gamma\left(1 - \frac{i}{k_{12}}\right) e^{-\frac{2\pi}{k_{12}}} {}_1F_1(\mathbf{r}_{12}, \mathbf{k}_{12}). \quad (5)$$

Here \mathbf{r}_{12} is the relative distance between the two electrons and \mathbf{k}_{12} are the relative momenta.

With these approximations, the 3DW T matrix becomes

$$T_{fi}^{3\text{DW}} = \langle \chi_{\text{scat}} \chi_{\text{eject}} C_{\text{ele-ele}} | V - U_i | \Psi_A \chi_i \rangle. \quad (6)$$

Finally, the triple-differential cross section (TDCS) can be written in atomic units as

$$\frac{d^3\sigma}{d\Omega_f d\Omega_e dE_e} = \frac{1}{(2\pi)^5} \frac{k_f k_e}{k_i} (|T|^2). \quad (7)$$

B. DWB2-RM

As mentioned above, a partially successful theory for electron-impact ionization has been a hybrid approach, in which the interaction of a “fast” projectile electron with the target is described by a first-order or second-order distorted-wave approach, while the initial bound state and the scattering of a “slow” ejected electron from the residual ion is treated by an R -matrix (RM) approach. These DWB1-RM [1,55] and DWB2-RM [56] models were formulated for highly asymmetric kinematics and small energy losses compared to the incident energy.

Details of the hybrid approach can be found in many previous publications, e.g., [14,33,55]. Given that emission of the $3p$ electron is generally the dominant ionization process in the kinematical regime considered here, it is not surprising that using either a first-order or an approximate second-order treatment of the projectile produced very similar results. Also, coupling only the two final ionic states ($3s^2 3p^5$) $^2P^o$ and ($3s 3p^6$) 2S , rather than employing a much larger R -matrix with pseudostates (RMPS) expansion for the ejected-electron-residual-ion problem, is generally sufficient. A key issue, on the other hand, is the description of the initial bound state and the final ionic states included in the close-coupling expansion for the electron scattering from the residual ion. In

the hybrid method, we use the multiconfiguration expansions developed by Burke and Taylor [57] for the corresponding photoionization problem.

C. BSR

The BSR method is based on two steps: (1) the treatment of electron collisions with neutral argon using an extensive close-coupling expansion that contains both physical and pseudostates, with the latter being used to approximate the effect of high-lying discrete Rydberg states as well as the coupling to various (depending on the final ionic states) ionization continua; and (2) the construction of the *ionization* amplitude by combining the scattering amplitudes for *excitation* of the pseudostates using coefficients obtained by direct projection of the wave function to the various scattering channels associated with a particular final ionic state. For the case at hand, we performed a nonrelativistic RMPS calculation for e -Ar collisions with a total of 482 states in the close-coupling expansion. The atomic wave functions for neutral Ar were obtained by the B -spline box-based close-coupling method [58]. Altogether, we generated 482 physical and pseudo-target states with coupled orbital angular momenta $L = 0-5$ and energies reaching up to 80 eV. In the first step, we obtained the scattering amplitudes for excitation of all pseudostates using our suite of BSR codes [59] for electron collisions.

The last, and most crucial, step in the process is the generation of the ionization amplitudes. This is done by summing up the amplitudes for excitation of all energetically accessible pseudostates, with the weight factors given by the overlap of the pseudostates and the true continuum functions [28]. At this stage in the calculation, consistency between the models for the bound states (physical and pseudo) and the physical continuum scattering channels is critical. We ensure this consistency by employing the same expansions coupling the three ionic states ($3s^2 3p^5$) $^2P^o$, ($3s 3p^6$) 2S , and ($3s^2 3p^4 3d$) 2S states. More details can be found in [28,32,33].

IV. RESULTS AND DISCUSSION

The TDCS for electron-impact ionization of Ar($3p$) as a function of the ejected electron angle (θ_2) are presented in Figs. 2 and 3 for two different scattering angles ($\theta_1 = 10^\circ$ and 15°). (Looking at the scattering plane from above, the ejected electron observation angles θ_2 are measured clockwise and the projectile scattering angles θ_1 are measured counterclockwise.) Results are presented for ejected-electron energies ranging between 15 and 50 eV. The present experimental data are compared with our earlier measurements [48] as well as the measurements of Stevenson *et al.* [45] and Ren *et al.* [46]. The experimental data are also compared with the 3DW (three-body distorted-wave) model and the DWB2-RM (second-order distorted-wave Born R -matrix) model, as well as the nonperturbative BSR (B -spline R -matrix) approach. Since the measurements are not absolute, all experimental data and theoretical calculations have been normalized to unity at their peak, thereby allowing for a shape comparison. The primary difference between the two perturbative calculations (3DW and DWB2-RM) is the fact that the 3DW model contains one “collision” between the projectile and target and PCI is

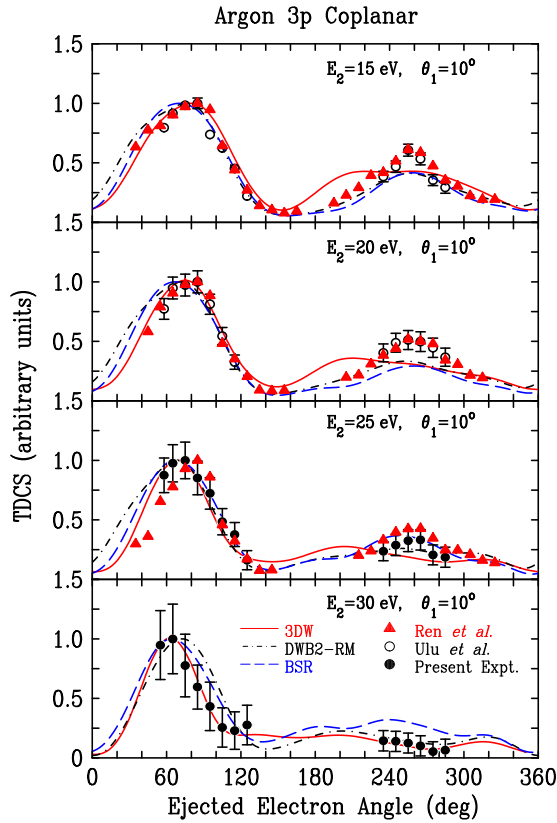


FIG. 2. (Color online) Experimental and theoretical TDCS for 200 eV electron-impact ionization of argon. The projectile scattering angle is 10° and the ejected electron energies are noted in each subsection of the figure. The theoretical calculations are 3DW: solid line; dash-dot: DWB2-RM; and dashed: BSR. The experimental data are triangles: Ren *et al.* [46]; open circles: Ulu *et al.* [48]; and solid circles: present results. All theories and experiment were normalized to 1.0 at the maximum of the binary peak (see text).

included to all orders of perturbation theory while DWB2-RM accounts for up to two collisions between the projectile and target with PCI contained to second order within the R -matrix box. In addition, DWB2-RM contains exchange between the ejected electron and target to numerical accuracy while the 3DW uses the Furness-McCarthy approximation [60] for this exchange effect.

From Figs. 2 and 3, it is seen that overall there is excellent agreement between four different sets of measurements taken at different times and in different laboratories. The only noticeable difference occurs for 25 eV and 10° where it appears that there is a small shift in the location of the binary peak between the present measurements and those of Ren *et al.* [46]. The overall good agreement between the various measurements indicates the accuracy of the present measurements of the TDCS.

Looking in detail first at Fig. 2, it is seen that the BSR and DWB2-RM results are in reasonably good agreement with experiment for all four of the measured energies. For the binary peak, all three theories are in very good agreement with each other and experiment. For 25 eV, all three theories predict the same binary peak angle, in excellent agreement with the present data. For 30 eV, the DWB2-RM binary peak

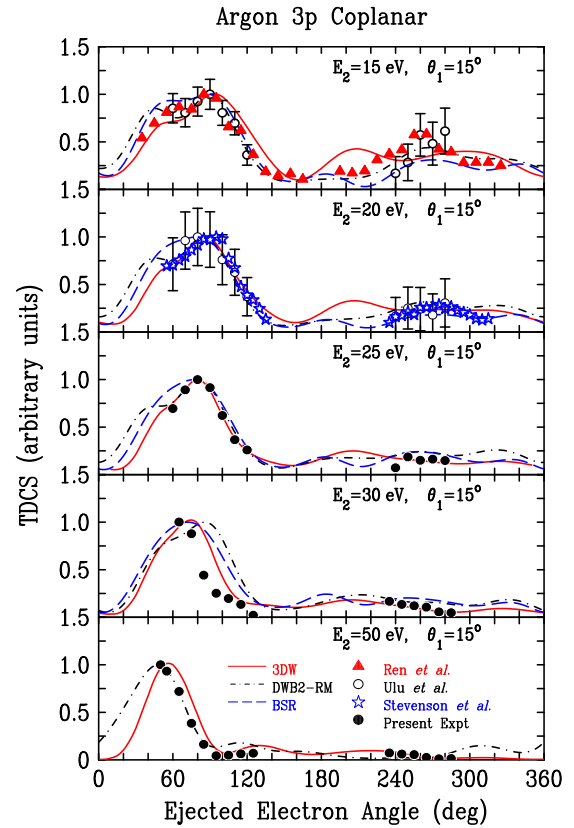


FIG. 3. (Color online) Experimental and theoretical TDCS for 200 eV electron-impact ionization of argon. The projectile scattering angle is 15° and the ejected electron energies are noted in each subsection of the figure. The theoretical calculations are 3DW: solid line; dash-dot: DWB2-RM; and dashed: BSR. The experimental data are triangles: Ren *et al.* [46]; open circles: Ulu *et al.* [48]; stars: Stevenson *et al.* [45]; and solid circles: present results.

is slightly shifted to higher angles as compared to the other two theories and experiment. Overall, the 3DW calculation appears to give the best prediction for the width of the binary peak. On the other hand, the 3DW provides the worst agreement with experiment for the recoil peak, except for the highest energy where the 3DW is in excellent agreement with data. The BSR calculation, which one would expect should give the best agreement with experiment, is in excellent agreement with the data for 25 eV, and very good for the other energies, except for the height of the recoil peak (too small for low energies and too high for large energies). The DWB2-RM results are very similar to the BSR.

It is interesting to note that both perturbative calculations exhibit improved agreement with experiment with increasing ejected electron energy as one would expect. The fact that the 3DW results agree better with the binary peak than the DWB2-RM for 30 eV indicates that PCI is more important than higher-order interactions between the projectile and target. For the recoil peak, the second interaction with the target is clearly much more important than PCI. The fact that the agreement between experiment and theory for the BSR does not exhibit any noticeable energy dependence would also be expected for a converged nonperturbative calculation.

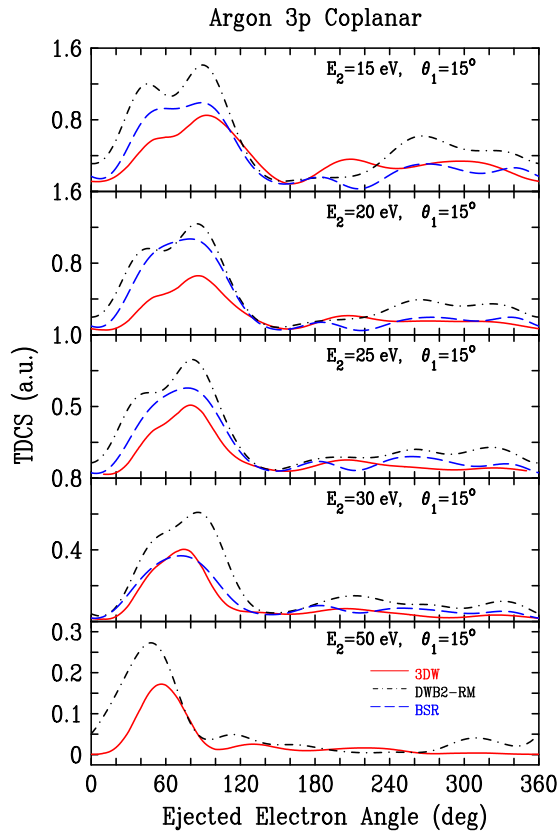


FIG. 4. (Color online) Same as Fig. 3 except that absolute values of the theories are shown in atomic units.

Figure 3 presents a similar comparison for a larger projectile scattering angle. There are no BSR results shown for 50 eV, due to excessive computer demands that would have been required to achieve convergence for this energy. Again the BSR model yields overall reasonably good agreement with the data. However, for the smaller electron ejection energies, the experimental binary peak has a noticeable small-angle shoulder that is predicted very nicely by both perturbative calculations. The BSR results exhibit a small shoulder for the lowest energy but not for the higher ones. The largest discrepancy between experiment and theory for the binary peak was found for the 30 eV case. For the recoil peak, there is relatively good agreement between experiment and all three theories for all the measured cases, except for the smallest energy where the 3DW exhibits some unobserved structure. Surprisingly, overall the perturbative approaches appear to yield a little better agreement with experiment than the BSR for this case. Probably the DWB2-RM yields the best overall agreement with experiment. This indicates that, for larger scattering angles, multiple interactions with the target are more important than PCI.

Since the experimental data were not determined on an absolute scale, we have normalized experiment and theory to unity for the binary peak. It is, however, also of significant interest to look at the relative absolute values predicted by the theories. Figure 4 shows the same theoretical cross sections presented in Fig. 3, but now on an absolute scale. In general, the 3DW results tend to predict the smallest binary peak and the DWB2-RM results the largest, with the difference being

a factor of nearly 2. Obviously absolute, or at least cross-normalized measurements, would be highly desirable.

V. CONCLUSION

While there are very accurate nonperturbative numerical calculations available for electron-impact ionization of hydrogen and helium, more study is needed to report comparable accurate calculations for ionization of heavier atoms such as the inert gases. Recently, Zatsarinny and Bartschat introduced the nonperturbative *B*-spline *R*-matrix (BSR) approach for ionization of inert gases, which had some significant success but without resolving all remaining discrepancies between experiment and theory. There have been several previous studies comparing experiment with perturbative theoretical calculations for electron-impact ionization of argon, and significant discrepancies have been found. We recently compared experiment and theory for 200 eV electron-impact ionization of Ar for three projectile scattering angles and ejected electron energies of 15 and 20 eV. The current study revealed a qualitative agreement between experiment and both the perturbative and nonperturbative calculations but there were still significant differences.

The purpose of the present work was to extend this comparison to a much larger energy range (10–50 eV) to see if any general trends could be found. The study was limited to two projectile scattering angles, 10° and 15°. Overall the BSR results were in reasonably good agreement with experiment—but not as good as has been found for hydrogen and helium. For 10°, the BSR width of the binary peak was broader than experiment for the higher energies, and the magnitude of the recoil peak was too small for small energies and too large for the highest energy. The width of the binary peak predicted by the 3DW was closest to experiment, and the agreement between experiment and the 3DW improved dramatically with increasing ejection energy and excellent agreement was found for the highest energy measured. For 15°, again the BSR results were in reasonably good agreement with experiment, particularly for the recoil peak. For the binary peak, the BSR predicted a wider peak than found by experiment and the detailed shape of a low-angle shoulder was better predicted by both the perturbative calculations. Again the 3DW results for the recoil peak gave better agreement with experiment with increasing energy with excellent agreement being achieved already by 20 eV. In summary, the BSR was reasonably good for all measured cases but did not predict all the detailed structure that the perturbative approaches did predict. All calculations showed some good points and some weak points, and hence it would be difficult to pick the “best” one.

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