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Triple-differential cross-section measurements of electron-impact ionization of argon 3*s* in the backward scattering direction

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The triple-differential cross section for the electron-impact ionization of the 3*s* orbital of argon has been measured over the complete backward scattering direction of the ejected electron, using a magnetic angle changer in a conventional $(e,2e)$ spectrometer. The results reveal the position and relative magnitude of the recoil scattering peak in the cross section for this target, and enable a detailed comparison with recently published calculations of argon 3*s* ionization which have been performed using a hybrid distorted-wave Born approximation—*R*-matrix approach. These theoretical calculations show dramatically improved agreement with the experimental results, in comparison with earlier approaches, but puzzling discrepancies still remain.

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INTRODUCTION

Electron-impact ionization is a process of fundamental importance in a wide range of physical phenomena which occur in areas such as astrophysics, discharge physics, plasma physics, and radiobiology. The measurement of the triple-differential cross section (TDCS) for the electron-impact ionization of atoms and molecules provides the most complete information on the dynamics of this ionization process. In obtaining this cross section, the process under investigation is an $(e,2e)$ process, in which the single ionization of the target by the incident electron produces an outgoing scattered electron and ejected electron, and the energies and momenta of all three electrons are uniquely determined. The TDCS for the electron-impact ionization of the simplest targets, H and He, has been extensively investigated both experimentally and theoretically, and in recent years there has been significant theoretical progress in the description of this three-body process [1–3]. For heavier targets such as argon and krypton, it has not been possible to implement the highly computationally intensive theoretical approaches used so successfully for H and He, and other theoretical approaches must be pursued, with the most widely used being the distorted-wave Born approximation (DWBA) (see [4] for a comprehensive overview of previous work). These heavy targets thus provide a particular challenge for theory, and indeed recent experimental data for electron-impact ionization of the 3*s* [5,6] and 3*p* [7–9] orbitals in argon at low to intermediate incident energies have proven to be surprisingly difficult to describe theoretically. This was particularly true in the "recoil," or backward scattering region. The revelation of these very significant discrepancies between experiment and theory has prompted a considerable amount of theoretical development. In particular, Madison and co-workers [10,11], using the distorted-wave Born approximation approach, considered how the treatment of exchange and postcollision interaction in the final channel might influence the TDCS. More recently, a hybrid DWBA–*R*-matrix approach, originally developed for first-order calculations of total ionization cross sections [12,13], was further extended and applied to angle-differential 3*p* and 3*s* ionizations of argon, treating the projectile-target interaction up to second order [4,14]. These theoretical developments have afforded improvement in the agreement with the experiment, but there are still areas of considerable disagreement, particularly in the case of argon 3*s* ionization, where there are large model-dependent variations. A common theme evinced by theorists has been the need for more experimental results, particularly internormalized data sets, and measurements over a large angular range. Here, we report results that address the second of these requests. We present experimental data for the electron-impact ionization of argon, which have been measured over a previously inaccessible angular range, using a device that is referred to as a magnetic angle changer (MAC) [15]. This device alters the trajectory of the electrons to deflect them from inaccessible to accessible regions, potentially allowing access to the full angular range. The MAC has previously been used by a number of groups to extend elastic and inelastic (excitation) scattering measurements (see, for example, [16–18]), for a range of targets, to encompass scattering angles from 0° to 180°. In the present work, the MAC is used to extend the angular range of our previously measured TDCS for Ar 3*s* ionization [5], into the backward scattering direction. The results provide an opportunity to compare an expanded experimental data set with very recently published theoretical calculations [14].

EXPERIMENTAL APPARATUS

The coincidence spectrometer has been described in detail in a previous publication [5]. An electron gun produces a beam of monoenergetic electrons from a tungsten filament, using two cylindrical three-element electrostatic lenses. The electron beam crosses a target gas beam, produced from a stainless steel capillary, at right angles. Electrons from the ionization event are detected using two hemispherical electron energy analyzers with channel electron multipliers (CEMs) mounted at the hemisphere exit apertures. Conventional fast timing electronics are used in the coincident detection of the two electrons.

Coplanar asymmetric kinematics were used for these measurements. In the coplanar geometry, the incident and two outgoing electrons from the ionization event lie in the same plane. The electron energy analyzers are mounted in this plane on rotatable turntables, independently driven by stepper motors, while the electron gun is fixed. The two outgoing electrons from the ionization event are detected with different kinetic energies (asymmetric energy sharing), with the faster electron (referred to as the scattered electron) being detected at a fixed forward angle of −15°, while the detection angle of the (slow) ejected electron is varied in the scattering plane. The emission angles of the ejected electrons are presented as positive angles. Normally, in such "conventional" coincidence spectrometers (as opposed to multiparameter spectrometers based on toroidal analyzers or "reaction microscopes" [19,20]) the ejected electron can only be detected over a limited angular range due to the physical constraints imposed by the relative positions of the electron gun and electron energy analyzers. For example, in the present apparatus the accessible angular regions are 40°–135° and 225°–285°. This restriction has been overcome in the present work by the incorporation of the magnetic angle changing device developed by Read and Channing [15]. The MAC utilizes a system of solenoids to produce a uniform localized magnetic field at the interaction region. It consists of two coaxial pairs of solenoids, an inner pair and an outer pair, the upper and lower coils of each pair being separated by a gap through which electrons enter and exit the interaction region. The device used in our measurements is based on a previous design [18] which incorporates mu-metal pole pieces that surround the solenoids. With a suitable geometry and careful selection of the outer to inner solenoid current ratio, I_0/I_I [14], the MAC generates a highly localized field in which low order multipoles of the B field are canceled out and only short-range higher order multipoles remain. A highly localized field is required in order to ensure that the performance of the electron gun and analyzers remains unaltered. Electrons passing through the interaction region are deflected by an amount

$$\theta = a \arcsin\left(b\frac{I_0}{\sqrt{E}}\right), \tag{1}$$

where E is the kinetic energy of the electron and a and b are empirically determined constants. The mu-metal pole pieces assist in reducing stray magnetic flux beyond the physical extent of the MAC and also enable the generation of higher fields in the interaction region for a given value of I_0. This enables larger deflections to be achieved (without heat dissipation problems), but has the disadvantage of an increased background of secondary electrons at low energies, as observed in [18]. We note that this adds significantly to the difficulty of the $(e,2e)$ experiments with ejected electron energies below 5 eV.

During the $(e,2e)$ process, the incident, scattered, and ejected electrons all undergo deflection by different amounts ($-\Delta\theta_{\text{inc}}$, $-\Delta\theta_{\text{scat}}$, and $\Delta\theta_{\text{ej}}$, respectively) depending on their energies. In the asymmetric energy scheme, the incident and "fast" scattered electrons will be deflected by a small amount compared to the "slow" ejected electrons. If the field direction is chosen to rotate the ejected electron distribution anticlockwise with respect to the incident beam, the direction of the scattered beam is also rotated anticlockwise. As the lower angular range of the ejected electron analyzer is typically limited by the position of the scattered electron analyzer, this has the advantage of increasing the angular range of the ejected electron analyzer by an amount $\Delta\theta_{\text{inc}}+\Delta\theta_{\text{scat}}$. However, if the deflection is chosen to be clockwise, this decreases the angular range in a similar way. Figure 1 schematically illustrates the effect of the magnetic angle changer on the electron trajectories.

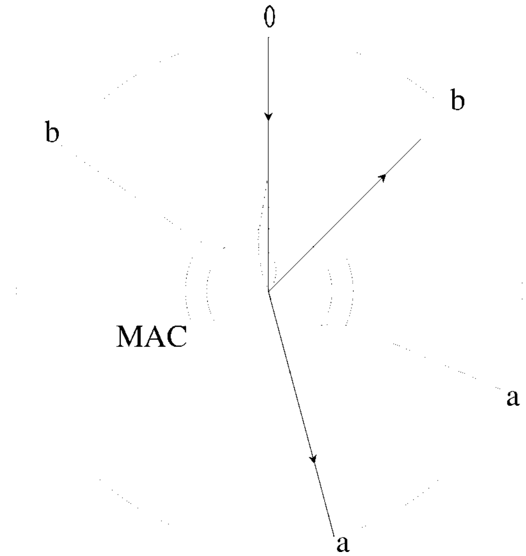

FIG. 1. Schematic diagram of the trajectories of the incident (*0*), scattered (*a*), and ejected (*b*) electrons, for emission of the ejected electrons into the recoil direction. The solid lines are the unperturbed trajectories and the dashed lines are the trajectories with the magnetic angle changer.

The angular deflection of the elastically scattered incident electrons, and of the scattered electrons from the ionization event, can be experimentally determined by using the symmetry of the elastic scattering cross section, or of the double-differential cross section (DDCS) for the scattered electrons, to obtain the new zero position. This approach is not applicable to the low energy ejected electrons, as the DDCS in this case is rather isotropic. However, measurements of the deflection of elastically scattered electrons at a number of energies allows one to empirically obtain the a and b parameters in Eq. (1), and thereafter apply this equation to the low energy ejected electrons. Using this procedure, we estimate an uncertainty of ±5° in the final angular scale.

RESULTS AND DISCUSSION

The new data together with the results of [5] can be seen in Fig. 2. Triple-differential cross sections were measured with an incident electron energy of 113.5 eV, for ejected electron energies of 10, 5, and 2 eV. The scattered electron energy is determined by energy conservation. Thus

$$E_0 = E_a + E_b + \varepsilon_i, \tag{2}$$

where E_0 is the incident electron energy, E_a and E_b are the scattered and ejected electron energies respectively, and ϵ_i is

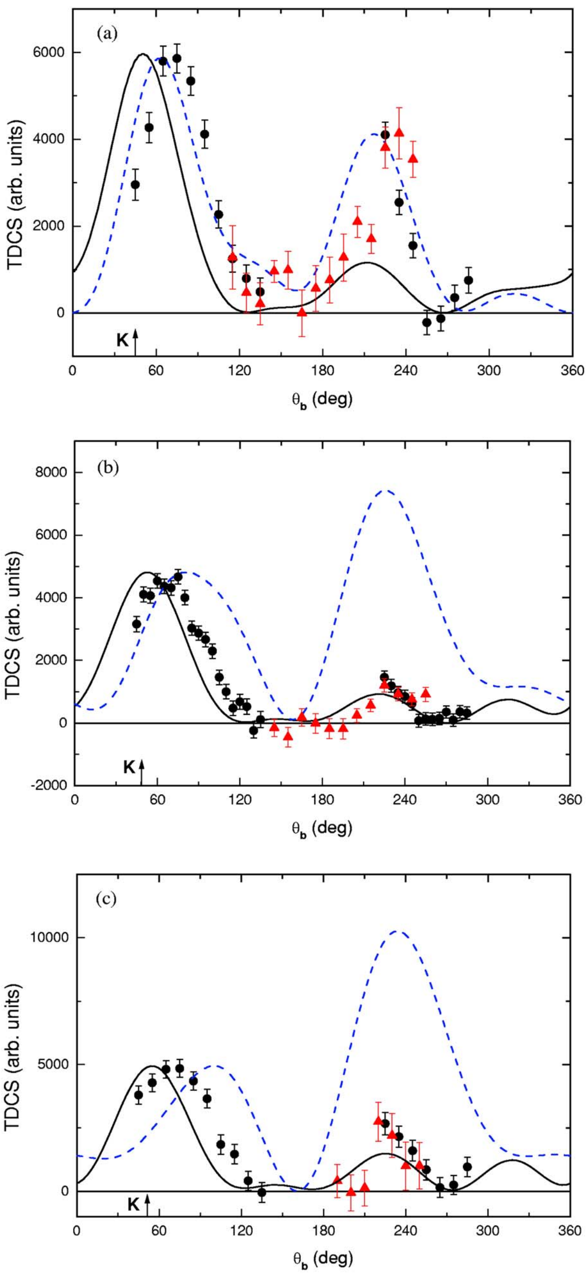

FIG. 2. (Color online) Measured and calculated triple-differential cross sections for the electron impact ionization of the 3*s* orbital of argon. The kinematic conditions were $E_0=113.5$ eV, $\theta_a=-15^\circ$, and (a) $E_b=10$ eV, (b) $E_b=5$ eV, and (c) $E_b=2$ eV. The solid circles are the original experimental data from Ref. [5], and the solid triangles are the results obtained with the magnetic angle changer. The dashed curve is the 3DW calculation, and the solid curve is the DWB2-2st-MC (see text for details). The direction of the momentum transfer, **K**, is indicated by the arrow on the *x* axis.

the binding energy of the 3*s* orbital in argon (29.3 eV). The scattered electrons were detected at a scattering angle of $\theta_a=-15^\circ$ (345°). Error bars are statistical and represent one standard deviation. As only relative, and not absolute cross sections are measured, the new data have been measured over an angular range which overlaps that of the previous data set. This enables the magnitude of the new data to be normalized against the previous data, in such a way as to provide the best visual fit. The cross sections show the typical behavior that has been observed for the ionization in the coplanar asymmetric kinematics, i.e., a forward angle (binary) peak, roughly in the momentum transfer direction, which is due to an impulsive electron-electron collision, and a backward angle (recoil) peak that can be explained by a further recoil scattering of the ejected electron from the nucleus. An examination of the earlier data illustrates the problem encountered when the full angular emission range of the electrons cannot be accessed. In each case (10, 5 and 2 eV ejected electron energy) the earlier measurements were not able to ascertain the position of the maximum of the recoil peak, or how large this peak might be in relation to the binary peak. As will be seen from the calculations, the theoretically predicted position and relative magnitude of this peak are critically dependent on the particular model. We compare the experimental data with the 3DW (with exchange) calculation of Ref. [11] (referred to as 3DW-EX in [11]), and with a hybrid DWBA–*R*-matrix calculation [14]. To our knowledge, the 3DW calculation of Prideaux and Madison[11] was the first DWBA calculation with post collision interaction (PCI) included in the final-state wave function. In this model, the final-state wave function includes two distorted waves plus the electron-electron Coulomb distortion factor. When the Coulomb distortion factor is contained in the final state wave function, PCI is included to all orders of perturbation theory. The hybrid DWBA–*R*-matrix calculation uses a second-order distorted-wave Born approximation (DWB2) to describe the incident and scattered electrons. The ejected electron-residual ion scattering process is modeled by a close coupling expansion, where the inner 3*s* and outer 3*p* shell ionization channels are coupled (2 states=2st). The initial bound state and final ionic state are modeled by a multiconfiguration (MC) expansion to account for configuration mixing. The calculation is referred to in Ref. [14] as DWB2-2st-MC. In each case in Fig. 2, the theoretical calculations are normalized to the experimental data at the maximum in the binary peak. This is necessary as the experimental cross sections are not absolute, but disguises the large variations in magnitude between the different calculations.

We note that in Ref. [14], the authors commented on the large discrepancy between the measured and the theoretically predicted magnitude of the recoil peak relative to the binary peak for an ejected electron energy of 10 eV. In light of this large discrepancy, and with regard to the additional information on the position and shape of the recoil peak, we have remeasured this ratio. The ratio was measured without employing the magnetic angle changer, using the same procedure outlined in [5], that is, by moving the scattered electron energy analyzer from -15° to $+15^\circ$, and measuring the ejected electron signal at the peak in the binary and recoil regions. The ratio is somewhat smaller than our previous value (recoil /binary binary$=0.7\pm0.1$ as opposed to 0.9 as given in [5]), but the measured recoil peak is still substantially larger than that predicted by the DWB2-2st-MC model. The agreement with the 3DW is, however, considerably better. This is not the trend for the other ejected electron energies, where the DWB2-2st-MC tends to be in a much better agreement than the 3DW. At 5 and 2 eV ejected electron energy, the latter calculation is in very good agreement with the experimental cross section in the recoil region, including the experimentally determined binary/recoil ratio. One may contrast this with the 3DW which at these lower ejected electron energies grossly overestimates the size of the recoil peak. Both calculations have difficulty in correctly predicting the angular position of the binary peak, but the position of the recoil peak (which has now been experimentally verified by the new data) is predicted quite well. Post collision interaction between the two outgoing electrons is not included in the DWB2-2st-MC and may explain the incorrect binary peak position. (Measurements of argon $3p$ ionization over a greatly extended angular range are currently underway in our group—the results suggest that there is a strong influence due to PCI at smaller ejected electron emission angles.) Comparison of our results with the latest theoretical calculations supports the suggestion [14] that consideration of channel coupling in the ejected electron-residual ion scattering process and use of a multiconfiguration target description are very important at these energies, particularly for the argon $3s$ ionization. However, a serious discrepancy between the DWB2-2st-MC calculation and the experimental results still exists at 10 eV ejected electron energy.

CONCLUSION

The magnetic angle changing technique has been employed to measure triple-differential cross sections, in the backward scattering direction, for the inner valence shell ionization of argon. The new experimental data have resolved the uncertainty with regard to the magnitude and position of the recoil peak in the angular distribution. Comparison with very recent calculations which include channel coupling effects, and multiconfiguration effects in the target description, indicate that these calculations significantly improve the agreement with experiment. Despite this, it is clear that significant physical effects are still missing from the calculations.

The experimental results also demonstrate the efficacy of the magnetic angle changing technique in $(e,2e)$ experiments. Under certain energy conditions, the MAC can facilitate measurement of triple-differential cross sections over the full angular range. This introduces the possibility of accessing particularly interesting angular regions of the TDCS, such as the region where the scattered electron and ejected electron emerge from the collision at the same angle, a region where PCI effects should be most significant. Although such measurements are possible in principle with reaction microscopes, the use of a conventional $(e,2e)$ spectrometer equipped with a MAC would allow one to perform experiments concentrating on this small region of parameter space. Experiments to explore such effects are underway.

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