Experimental and theoretical determination of the triple differential cross section for Kr(4p) electron-impact ionization

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The triple-differential cross section (TDCS) for electron-impact ionization of the krypton $4p$ orbital has been measured at an incident energy of 919.4 eV, scattered electron energy of 880 eV, and ejected electron energy of 25 eV. The measurements were performed at scattering angles ranging from 3° to 20°. The cross sections have been placed on an absolute scale using a technique that relies on a relative normalization against a reliable theoretical TDCS for helium $1s$ ionization. The experimental data are compared with a distorted-wave Born-approximation calculation. There is good shape agreement between the theoretical results and the experimental data, and satisfactory agreement in magnitude (within the experimental uncertainty of 23%) across all scattering angles. [S1050-2947(99)03010-3]

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I. INTRODUCTION

In the field of electron-impact ionization, the $(e,2e)$ technique yields detailed information on the dynamics of the single-ionization process. The process may be represented as

$$e_0+A\rightarrow A^+ +e_a+e_b .$$

The target A is ionized by an incident electron of energy and momentum $E_0,\mathbf{k}_0$, and the two outgoing electrons have energies E_a and E_b, with momenta $\mathbf{k}_a,\mathbf{k}_b$. The resulting cross section is a measure of the probability of detecting these two electrons, in coincidence, in solid angles Ω_a and Ω_b and is called the triple-differential cross section (TDCS). By energy conservation,

$$E_0=E_a+E_b+\varepsilon_i , \tag{1}$$

where ε_i is the binding energy of the orbital in question. A considerable body of experimental data has been accumulated over the last 20 years, much of it on light targets such as hydrogen and helium (see, for example, [1,2]). In recent years, theoretical approaches have been developed that are very successful for these targets [3,4], and considerable progress has been made in extending this success to heavier atoms such as argon, krypton, and xenon [5]. In testing the range of validity of the various theories, it has become increasingly important to obtain absolute measurements of the relevant cross sections [6]. The production of absolute experimental $(e,2e)$ cross sections is a difficult problem, and a number of techniques have been used, with varying degrees of success [7].

Recently, $(e,2e)$ results for Kr($4p$) and Xe($5p$) ionization have been published in which the authors attempted to place the experimental data on an absolute scale [8]. A comparison with theoretical results calculated in the distorted-wave Born approximation (DWBA) revealed major discrepancies (factors of 4–6) in the absolute magnitude, although the shapes of the cross sections were described quite well. It was suggested by the authors that the technique used by them to obtain the absolute experimental cross sections may be inappropriate in the case of heavy atoms, where the strong static potential results in distortion effects that severely restrict the region of applicability of the first Born approximation.

In order to investigate further the apparent disagreement between theory and experiment for such heavy targets, we have performed a series of measurements of the TDCS for Kr($4p$) ionization at an incident electron energy of 919.4 eV, ejected electron energy of 25 eV, and scattered electron energy of 880 eV. The scattering angles used were 3, 5, 8, 10, 15, and 20°. These kinematic conditions include very asymmetric geometries similar to those used in Ref. [8], but also encompass the bound Bethe ridge (see Sec. III) and beyond. The experimental cross sections have been placed on an absolute scale using a technique described in Refs. [9, 10], and are compared with a DWBA calculation.

II. EXPERIMENTAL DETAILS

The experimental measurements were performed in an apparatus incorporating two independently rotatable hemi-

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spherical analyzers and a fixed electron gun, mounted in such a way that the momentum vectors of the incident electrons and the outgoing detected electrons are coplanar. The incident electron beam crosses at right angles a beam of atoms produced by effusion of the target gas from a stainless-steel capillary. The energy analyzers are equipped with five-element input lenses, and channeltrons are used for electron detection at the exit. Further details regarding the apparatus may be found in Ref. [11]. The TDCS's were measured in coplanar asymmetric kinematics, in which the incident and scattered electrons have a much higher energy than the ejected electron. The scattered electron-energy analyzer is fixed at a forward angle, θ_a and the angular position of the ejected electron-energy analyzer θ_b is varied. The coincidence count rate is then measured as a function of θ_b. It is important that both electron-energy analyzers view the entire

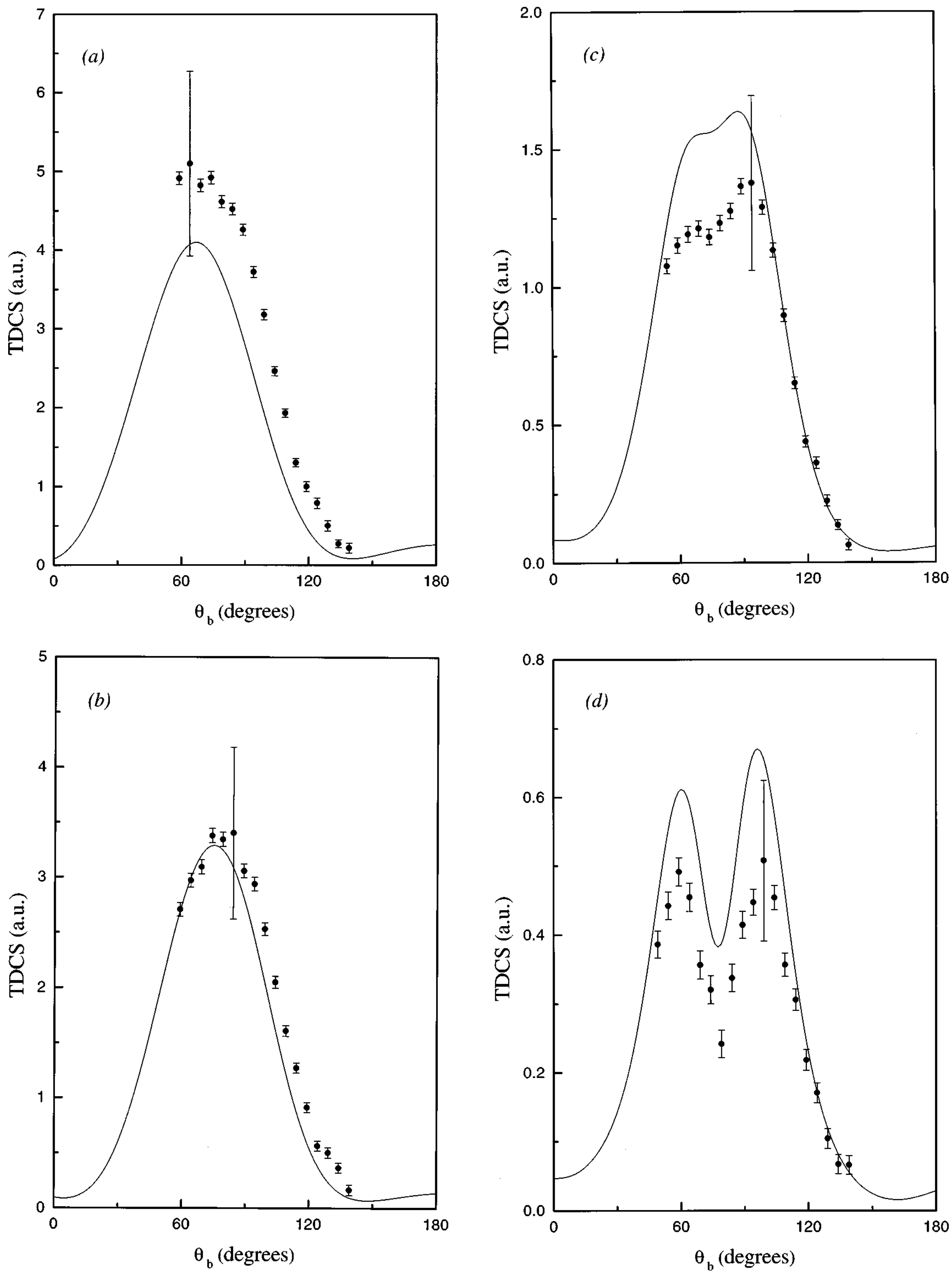

FIG. 1. Absolute TDCS (in atomic units) for Kr($4p$) ionization with $E_0 = 919.4$ eV, $E_a = 880$ eV, and $E_b = 25$ eV and θ_a equal to (a) 3°, (b) 5°, (c) 8°, (d) 10°, (e) 15°, and (f) 20°. The points are the experimental data and the solid curve is the DWBA calculation. The estimated uncertainty in the experimental absolute-scale determination is indicated by the large error bar near the binary-peak maximum.

interaction region at all angles used in the measurements. To check that the ejected electron analyser views the whole interaction region we have measured the TDCS for He ionization at an incident energy of 1024.6 eV, a scattered electron energy of 950 eV, and 50-eV ejected electron energy; the scattering angle was 13.5°. Our results are in excellent agreement with those in Ref. [12], generally within 5%, and at all angles within the combined error bars of the two data sets. We have also measured the Kr$(4p)$ TDCS under the same conditions used in Ref. [8], that is, $E_0=1034.5$ eV, $E_a=1000$ eV, $E_b=20$ eV, and $\theta_a=2°$. Our results agree with the latter within the combined error bars. For the scattered electron-energy analyzer, calculated geometrical angular acceptances indicate that at the forward angles considered here the analyzer views the entire interaction region. Additionally, angular distribution measurements of the intensity of the isotropic $L_2-M_{23}M_{23}$ (^3P) Auger line in argon showed that within 10% both analyzers viewed the entire interaction region, over the range of angles accessed in these measurements. Angular calibration of the ejected electron analyzer is performed by measuring the position of the sharp minimum in the elastic-scattering cross section for argon at an incident energy of 100 eV [13]. Zero degrees for the scattered electron-energy analyzer is determined from the symmetry of the double-differential cross section (DDCS). Relative normalizations between the cross sections measured at different scattering angles are determined in a separate experiment in which the ejected electron analyzer is fixed at an angle near the maximum of the binary peak in each case; a constant counting time is used at each scattering angle, and gas pressure and gun current are monitored to ensure no variations. The coincidence energy resolution is given by $\Delta E=(\Delta E_a^2+\Delta E_b^2+\Delta E_0^2)^{1/2}=1.8$ eV, where $\Delta E_a=\Delta E_b=1.2$ eV and $\Delta E_0\sim 0.5$ eV. The angular acceptances are 0.9° for the scattered electron analyzer and 3.5° for the ejected electron analyzer.

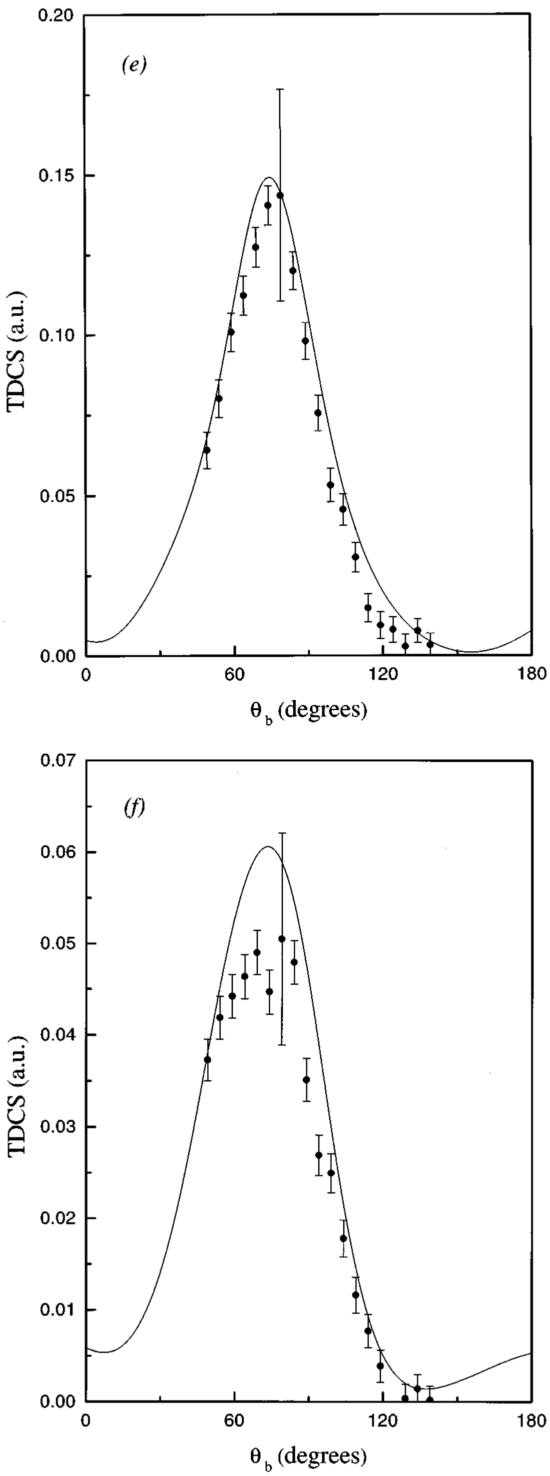

FIG. 1 (*Continued*).

III. ABSOLUTE-VALUE DETERMINATION

In order to place their Kr$(4p)$ and Xe$(5p)$ measurements on an absolute scale, Rasch *et al.* [8] employed a method used by Jung *et al.* [14] to normalize their helium data, taken at an incident energy of 600 eV. The measurements of Rasch *et al.* were taken at incident energies of around 1 keV, ejected electron energy of 20 eV, and scattering angles of 2 and 8°. The normalization procedure relies on the fact [15] that in the limit of zero-momentum transfer, the triple-differential generalized oscillator strength (TDGOS) is proportional to the dipole (optical) oscillator strength. This is true for both the binary collisions and the recoil collisions, and relies on the fact that as the momentum transfer $K\to 0$, a first Born treatment becomes justified, since higher-order effects tend to zero, regardless of the incident energy.

Thus, for a helium target one obtains [7]

$$\lim_{K\to 0} f^{(3)}(K,E,\alpha=0)=\lim_{K\to 0} f^{(3)}(K,E,\alpha=\pi)$$
$$=\frac{1}{4\pi}f_0(E)[1+\beta P_2(\cos\alpha)]$$
$$=\frac{3}{4\pi}f_0(E), \tag{2}$$

where $f^{(3)}(K,E,\alpha)$ is the TDGOS, $f_0(E)$ is the dipole oscillator strength, E is the energy loss, α is the angle between the ejected electron direction and the vector $\mathbf{K}=\mathbf{K}_0-\mathbf{K}_a$ and $\beta=2$ is the asymmetry parameter for helium. The TDCS is related to the TDGOS by

$$\frac{d^3\sigma}{d\Omega_a d\Omega_b dE}=\left(\frac{2k_a}{k_0}\right)\frac{1}{EK^2}f^{(3)}(K,E,\alpha). \tag{3}$$

The dipole oscillator strength may be obtained from photoionization transition probabilities [16]. Experimental measurements of the TDCS may then be placed on an absolute scale by plotting the associated TDGOS [from Eq. (3)] versus K. A polynomial fitting procedure is used to extrapolate the measured TDGOS to $K=0$, where the absolute scale is set by reference to Eq. (2). Rasch *et al.* [8] identified two problems in using this procedure for their Kr and Xe results. Reliable extrapolation of the measured data requires closely spaced measurements down to as low a value of K as possible. In fact, there is a region of K close to zero (the unphysical region) where the cross section cannot be measured (this is due to the inelasticity of the process, which means that θ_a reaches zero before $K=0$) [17]. Additionally, experimental constraints often restrict the lowest K value, which can be reached. The first problem identified by Rasch *et al.* [8] was the unreliability of the fitting procedure, with different polynomial fits yielding widely different limiting values. The second problem emerged from an examination of the behavior of first Born and DWBA calculations as K approaches zero. The authors found that the two calculations converged only very close to or even inside the unphysical region, suggesting that even with a more extensive data set, the extrapolation approach is fundamentally flawed for these targets, in these kinematics. The authors emphasized, however, that the approach appears to be valid for helium.

Given the above conclusions, it is appropriate to investigate the suitability of other techniques for placing experimental TDCS results for heavy atoms on an absolute scale. The technique we have employed is based on that described in Refs. [9,10]. It relies on (i) normalization against a reliable absolute TDCS for helium and (ii) the proportionality between the double-differential cross section and the Compton profile.

As part of the normalization process $(e,2e)$ experiments were performed, under identical conditions, for krypton and helium. This means that the energies of the outgoing electrons and the scattering angle were kept the same, as were the analyzer efficiencies. The small change in incident energy required to meet the energy balance in the case of helium (24.5 eV binding energy compared with 14.4 eV for krypton) does not affect the electron-beam profile. Following [10], the coincidence count rates may be written

$$\dot{N}_{ab}^{(\mathrm{Kr})}=\sigma_{ab}^{(\mathrm{Kr})}(nLI)^{(\mathrm{Kr})}(\varepsilon_a\Delta\Omega_a)(\varepsilon_b\Delta\Omega_b)\Delta E_{ab}, \qquad (4)$$

$$\dot{N}_{ab}^{(\mathrm{He})}=\sigma_{ab}^{(\mathrm{He})}(nLI)^{(\mathrm{He})}(\varepsilon_a\Delta\Omega_a)(\varepsilon_b\Delta\Omega_b)\Delta E_{ab}. \qquad (5)$$

σ_{ab} is the TDCS for emission of electrons of energy E_a and E_b into solid angles Ω_a and Ω_b. n is the target gas number density, L is the effective interaction length, I is the incident current in electrons per second, ΔE_{ab} is the effective coincidence energy resolution [18], and $\varepsilon_a,\varepsilon_b$ are the transmission efficiencies of the analyzers.

The singles count rate in the scattered electron channel is related to the DDCS by

$$\dot{N}_{a}^{(\mathrm{Kr})}=\sigma_{a}^{(\mathrm{Kr})}(nLI)^{(\mathrm{Kr})}(\varepsilon_a\Delta\Omega_a)\Delta E_{ab}, \qquad (6)$$

$$\dot{N}_{a}^{(\mathrm{He})}=\sigma_{a}^{(\mathrm{He})}(nLI)^{(\mathrm{He})}(\varepsilon_a\Delta\Omega_a)\Delta E_{ab}. \qquad (7)$$

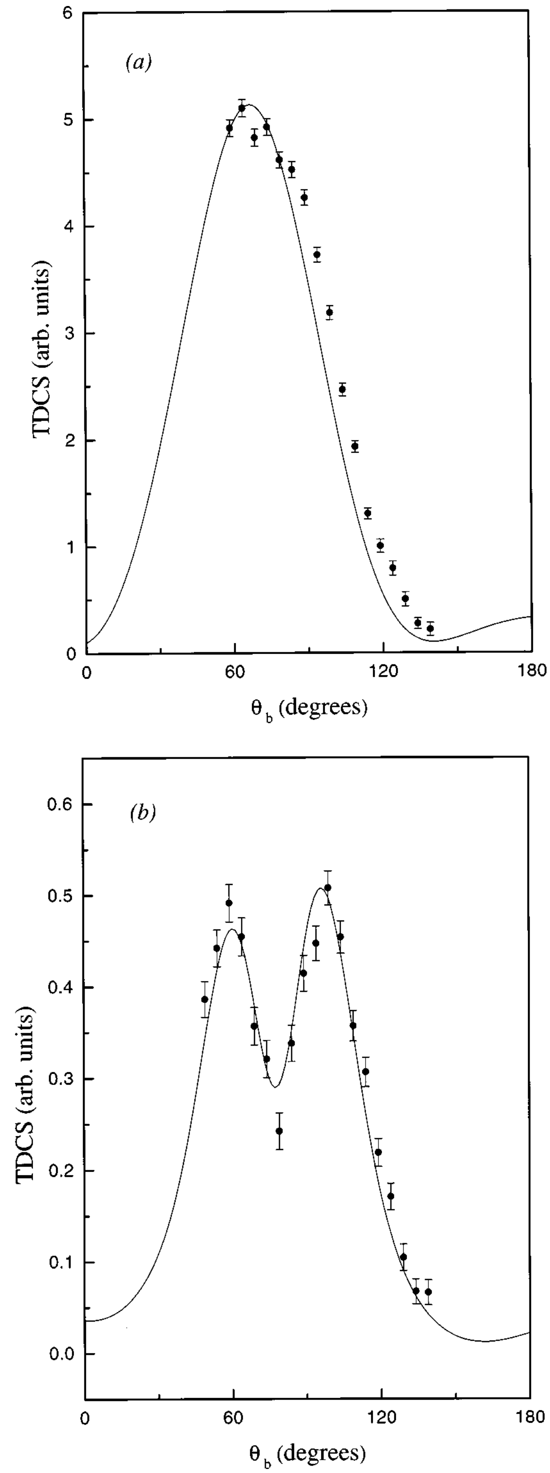

FIG. 2. TDCS for Kr$(4p)$ ionization with $E_0=919.4$ eV, $E_a=880$ eV, and $E_b=25$ eV and (a) $\theta_a=3°$, (b) $\theta_a=10°$. The points are the experimental data and the solid curve is the DWBA calculation normalized to the experimental data at the maximum of the binary peak.

Combining the above equations yields

$$\sigma_{ab}^{(\mathrm{Kr})}=\frac{\dot{N}_{ab}^{(\mathrm{Kr})}}{\dot{N}_{ab}^{(\mathrm{He})}}\frac{\dot{N}_{a}^{(\mathrm{He})}}{\dot{N}_{a}^{(\mathrm{Kr})}}\frac{\sigma_{a}^{(\mathrm{Kr})}}{\sigma_{a}^{(\mathrm{He})}}\sigma_{ab}^{(\mathrm{He})}. \qquad (8)$$

The theoretical DDCS is given by

$$\sigma_a=\frac{4k_a}{k_0K^5}J(0), \tag{9}$$

where $J(q)$ is a Compton profile that may be obtained from tables [19], and

$$q=\frac{(E_0-E_a)-\dfrac{K^2}{2}}{K}. \tag{10}$$

The case $q=0$ corresponds to Bethe-ridge conditions, $E_0-E_a=K^2/2$ (the bound Bethe ridge occurs when $E_b=K^2/2$). The peak in the Compton profile is at $q=0$, and this is normally where the DDCS measurements are performed. We have used a combination of measurements to fix the absolute scale: (i) $\dot{N}_{ab}^{(\mathrm{Kr})}$ and $\dot{N}_{ab}^{(\mathrm{He})}$ measured at a scattering angle of 10°, with DDCS measurements at the same angle. This requires using values of $q\neq 0$ in the tables of $J(q)$. (ii) $\dot{N}_{ab}^{(\mathrm{Kr})}$ and $\dot{N}_{ab}^{(\mathrm{He})}$ measured at a scattering angle of 10°, with DDCS measurements at scattering angles corresponding to Bethe-ridge conditions. This corresponds to $\theta_a=11.9°$ for krypton and 13.5° for helium. (iii) $\dot{N}_{ab}^{(\mathrm{Kr})}$ and $\dot{N}_{ab}^{(\mathrm{He})}$ measured at a scattering angle of 11.9°, with DDCS measurements performed at scattering angles corresponding to Bethe-ridge conditions for each target. A relative normalization is then obtained between the krypton TDCS at $\theta_a=11.9$ and 10°. All three approaches yield the same result, within their respective error bars. The overall error in the absolute values is estimated by adding in quadrature the error in the measured coincidence count rates for Kr and He (5 and 2%), the errors in the measured singles count rates (less than 1%) and the error in determining the DDCS from the Compton profiles. The latter is the most difficult to determine, as it depends on how applicable the impulse approximation is under the kinematic conditions employed in the above Kr and He measurements ($K\sim 1.4$ a.u.). Referring to the discussion in [20] and the estimated error used in [10] we have conservatively estimated an error of 15% in using this procedure. This yields an overall uncertainty of 23%. The theoretical cross section $\sigma_{ab}^{(\mathrm{He})}$ is obtained from a DWBA calculation, and is assumed to have no error. As a check, we also used the optical limit approach [Eqs. (2) and (3)] to obtain an absolute experimental value for this He cross section, for comparison with the theoretical value. The result was within 2% of the theoretical value for the cross section, but it is clear that the extrapolation procedure can be quite unreliable if the experimental data set does not extend to very low values of K.

IV. THEORY

The form of the TDCS for closed-shell atoms is [2]

$$\frac{d^3\sigma}{d\Omega_a d\Omega_b dE}=(2\pi)^4\frac{k_1k_2}{k_0}\sum_{m_b}\,[|f_{m_b}-g_{m_b}|^2+|g_{m_b}|^2+|f_{m_b}|^2], \tag{11}$$

where

$$f_{m_b}=\int\int \chi_a^-(\mathbf{k}_1,\mathbf{r}_1)\chi_b^-(\mathbf{k}_2,\mathbf{r}_2)\frac{1}{|\mathbf{r}_1-\mathbf{r}_2|}\times\chi_0^+(\mathbf{k}_0,\mathbf{r}_1)\phi_{m_b}(\mathbf{r}_2)d\mathbf{r}_1d\mathbf{r}_2 \tag{12}$$

and

$$g_{m_b}=\int\int \chi_a^-(\mathbf{k}_1,\mathbf{r}_2)\chi_b^-(\mathbf{k}_2,\mathbf{r}_1)\frac{1}{|\mathbf{r}_1-\mathbf{r}_2|}\times\chi_0^+(\mathbf{k}_0,\mathbf{r}_2)\phi_{m_b}(\mathbf{r}_1)d\mathbf{r}_1d\mathbf{r}_2. \tag{13}$$

$\chi_i^+(\mathbf{k}_0,\mathbf{r}_1)$, $\chi_a^-(\mathbf{k}_a,\mathbf{r}_1)$, and $\chi_b^-(\mathbf{k}_b,\mathbf{r}_2)$ are the distorted waves representing the incident, fast and slow electrons, respectively. $\chi_i^+(\mathbf{k}_0,\mathbf{r}_1)$ and $\chi_a^-(\mathbf{k}_a,\mathbf{r}_1)$ are calculated in the triplet exchange potential of the atom, while $\chi_b^-(\mathbf{k}_b,\mathbf{r}_2)$ is calculated in the triplet exchange potential of the ion. The exchange potential is of the Furness-McCarthy type [21].

The mathematical problems encountered in achieving convergence of the TDCS in very asymmetric geometry have been discussed in considerable detail in Ref. [8]. As shown there, many partial waves (up to 400) may be required to achieve convergence for this geometry. Ensuring convergence is achieved is very important since an unconverged calculation may yield the same shape for the cross section as the converged calculation, but the absolute value will generally be incorrect.

V. RESULTS

The absolute experimental and theoretical results are shown in Figs. 1(a)–1(f). The solid line is the DWBA calculation. Only the binary region, corresponding to ejected electron angles between 0 and 180°, is shown in each case. The error bars on the data points represent the statistical error (one standard deviation) in the coincidence data, while the estimated uncertainty in the absolute scale of the measurements is indicated by the large error bar near the maximum of the binary peak in each plot. Note that the absolute scale was determined for the experimental data at a scattering angle of 10°. The cross sections at other scattering angles were then scaled appropriately, using the experimentally obtained relative normalizations. In terms of the absolute magnitude of the cross section, there is very satisfactory agreement of theory with the experiment across all scattering angles, generally within the error assigned to the absolute measurements.

If the experimental data are renormalized so that the maximum value in the binary peak coincides with the maximum value in the theory, the DWBA calculation is generally in good shape agreement with the experimental data. Representative plots illustrating the level of agreement are shown in Fig. 2(a) ($\theta_a=3°$) and Fig. 2(b) ($\theta_a=10°$). There are some discrepancies; for example, at smaller scattering angles (3, 5, and 8°), the theory appears to be shifted by a few degrees from the data. Additionally, at $\theta_a=8°$ the structure at the maximum of the binary peak is not quite reproduced by the theory. The angular shift seen at lower scattering angles is also present in the data in Ref. [8], measured at a scattering angle of 2°. The authors noted that this shift is reminiscent of a postcollision interaction (PCI) effect; however they found that the shift could not be reproduced theoretically by inclusion of PCI via the M_{ee} factor of Ward and Macek [22].

At higher scattering angles there is very good agreement in shape between the calculation and the experimental data, generally within the statistical error. The cross section for $\theta_a = 10°$ corresponds closely to bound Bethe-ridge kinematics (the condition $E_b = K^2/2$ is satisfied when $\theta_a = 9.5°$). The dominant feature in the cross section, the strong minimum in the center of the binary peak, is characteristic of ionization of p orbitals under these kinematic conditions.

VI. CONCLUSIONS

TDCS's for Kr$(4p)$ ionization have been measured at a number of different scattering angles and compared with DWBA calculations. The experimental data have been placed on an absolute scale using a method that does not require use of the extrapolation technique [14], which was previously found to be inadequate or invalid for larger atoms [8]. The method used in this paper requires knowledge of an accurate theoretical or absolute experimental cross section for helium. The method is readily implemented, with an accuracy that is dictated by the reliability of the reference cross section and by the applicability of Eq. (9), which relates the DDCS to the Compton profile. The results show very satisfactory agreement between the theoretical calculation and the experimental data across all scattering angles. As noted in [8], the DWBA can correctly describe ionization of helium in these very asymmetric geometries, at a quantitative as well as qualitative level. The results presented here suggest that it is also quite successful in dealing with a heavier atom like krypton, at least in the binary region of the cross section.

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