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Röder, J.; Ehrhardt, H.; Pan, Cheng; Starace, Anthony F.; Bray, Igor; and Fursa, Dmitry V., "Absolute triply differential (e, 2e) cross sections for He in the intermediate energy region with comparison to theory" (1998). *Anthony F. Starace Publications*. 153.

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Submitted April 15, 1998; published June 14, 1998.

LETTER TO THE EDITOR

Absolute triply differential (e, 2e) cross sections for He in the intermediate energy region with comparison to theory

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Abstract

Absolute triply differential (e, 2e) cross section measurements are presented for He at incident energies $E_0 = 32.6, 44.6, 64.6,$ and 104.6 eV with equal energy sharing and the $\theta_{12} = \pi$ configuration. Results of distorted partial-wave calculations agree with the measurements; those of convergent close-coupling calculations agree with the relative angular distributions, and with absolute values after rescaling by consideration of the singly differential cross section.

The (e, 2e) process in He, in the case in which the He⁺ ion is left unexcited, is a fundamental way of investigating three-body Coulomb dynamics. In the intermediate range of energies from a few eV to the range of 50–100 eV above threshold, the many-body aspects of the process come to the fore, providing a stringent test of electron-impact ionization theories. Interest in triply differential (e, 2e) measurements in both H and He was sparked in the late 1980s by measurements taken at only 4 eV above the ionization thresholds [1, 2]. These showed that the shapes of the angular distributions depend on the target even though the asymptotic Coulomb fields experienced by the three final-state particles are target independent. While theoretical calculations [3–6] were able to replicate reasonably well the experimentally observed (e, 2e) angular distributions, the first absolute measurements, for He [7, 8], agreed with only two of these [4, 6]. The implication was that accurate accounting of target effects on the various electronic partial waves as well as treatment of electron–electron interactions are necessary to obtain agreement with absolute data and that omission of these effects can lead to disagreement with experiment by factors of 2–200 (see table II of the second reference of [4]). In response to these first absolute measurements for He, theorists have since devoted increased attention to providing not only accurate angular distributions but also accurate absolute cross sections [8–10]. In the intermediate energy region, an absolute measurement for He has been obtained by Gélébart and Tweed [11] at an excess energy of ≈ 75.4 eV, and the relative experimental measurements of Murray *et al.* [12] have been put on an absolute basis, in part by normalization to the measurements of [11]. However, these latter mea-

surements have been found to require a reduction by 18% [13]. Pan and Starace [14] extended their calculations [4] to the intermediate energy region in He in order to compare with the absolute data of [11, 12]. Although, in general, the agreement between theory and experiment is reasonably good, there are four energies for which theory did not pass through the experimental error bars.

We report here absolute measurements for the triply differential cross section (TDCS) for the $(e, 2e)$ process in He for excess energies of 8, 20, 40, and 80 eV, thereby covering the “intermediate” energy region. In all of the measurements presented, the two final-state electrons share the excess energy equally and depart in opposite directions (i.e. $\theta_{12} = \pi$). The new measurements are compared here with results of two theoretical approaches: the distorted partial-wave (DPW) approach of Pan and Starace [4] and the convergent close-coupling (CCC) approach of Bray and Fursa [15]. At low energies, the DPW approach [4] has been found to be in excellent agreement with the first absolute TDCS results for He [7, 8]. This theory has been employed recently to put new intermediate energy results for He by Bowring *et al.* [16] on an absolute basis. At high energies, the CCC approach provides accurate cross sections. Indeed, results for electron-He scattering at 100 eV and higher incident energies are in excellent agreement with most experimental features [15]. Comparisons are also made here with the other absolute experimental data for He in the intermediate energy region [11–13].

The measurements have been made with an $(e, 2e)$ spectrometer of crossed-beam type. It consists of an electron gun with a monochromator and two independently rotatable electron analyzers with an angular resolution of about $\pm 3.5^\circ$. Both analyzers use 127° cylindrical condensers for energy resolution, with the entrance slits being horizontally oriented. This has the advantage that no angular correction of the measured data is needed. The electron current produced by the gun is about 300 nA with an energy resolution of about 180 meV.

Absolute values are obtained with the assistance of accurate reference data for total ionization cross sections, σ_{ion} , rather than by attempting to measure all aspects of the scattering process. The dependence of the absolute values on the product of the target density n_{He} , the scattering length ℓ , and the rate of primary electrons N_e may be inferred by measuring the ion count rate N_{ion} via

$$N_{\text{ion}} = n_{\text{He}} N_e \ell \sigma_{\text{ion}}. \quad (1)$$

The ion rate was determined with a movable ion collector before and after the measurement of the coincident count rate. For the determination of the viewing angles and the detection efficiencies of the analyzers, the double differential cross sections (DDCS) of Müller-Fiedler *et al.* [27] have been used. Details about the spectrometer and the method used for the absolute normalization are given in the first reference of [7].

The theoretical approaches whose results are compared to the experimental measurements reported here have been described in detail elsewhere. Thus we merely summarize briefly their main features. For infinite nuclear mass, the differential cross section for electron-impact ionization becomes (in au) [17]:

$$\frac{d\sigma}{dk_1 dk_2} = \frac{(2\pi)^4}{k} |\langle \Psi_f^- | V | \Phi_i^+ \rangle|^2 \delta(E_f - E_i). \quad (2)$$

In equation (2), k is the magnitude of the momentum of the incident electron, k_1 and k_2 are the momenta of the two continuum electrons in the final state, and E_i and E_f are the energies of the initial and final states. The perturbation V is the difference between the exact Hamiltonian and the approximate Hamiltonian used to construct Φ_i^+ , the distorted wave used to describe approximately the initial state [18]. In the DPW approach, the incident electron is expanded in LS -cou-

pled partial waves in which each radial wavefunction is calculated in the Hartree-Fock (HF) potential V_{HF}^{N+1} describing the interaction of the incident electron with the target electrons. These HF radial functions are used to construct Φ_i^+ . In equation (2), V is defined approximately by

$$V = \sum_{i=1}^N \frac{1}{|\mathbf{r}_{N+1} - \mathbf{r}_i|} - V_{\text{HF}}^{N+1}(\mathbf{r}_{N+1}) \quad (3)$$

where the first term on the right-hand side of equation (3) is the Coulomb interaction between the incident electron and the N target electrons. Equation (3) is approximate for He, since we omit corrections to our ground-state HF description of the target. The final-state wavefunction Ψ_f^- in equation (2) should, in principle, be the exact solution to the full Hamiltonian satisfying the exact boundary conditions [19] for two continuum electrons moving in the Coulomb field of the ionized target. We have expanded our final-state wavefunction in independent-electron states for the two continuum electrons and have coupled their orbital and spin angular momenta to partial waves characterized by L and S . For He, S must be coupled to the spin of the target electron to form the system's spin, which equals $1/2$. Thus, in He, the target electron couples singlet and triplet states of the continuum-electron pair. However, we have ignored such interchannel coupling and treat the channels designated by L and S as uncoupled. The major approximation to Ψ_f^- in our calculation is our replacement of the exact Coulomb interaction between the two continuum electrons by a variationally determined screening potential [20–22]. Further details on the DPW approach are presented in [4, 23]. In summary, this approach treats distortion, non-local exchange interactions, both singlet and triplet partial waves, and mutual screening interactions using effective charges which satisfy proper asymptotic boundary conditions.

In the CCC approach (see [15] for details) the total wavefunction is expanded in a set of square-integrable (L^2) states, with the resultant coupled equations for the T -matrix solved in momentum space. The ionization amplitudes are constructed directly from the amplitudes corresponding to the excitation of the positive-energy pseudostates. The number of states N is increased until the CCC(N) results converge to a desired accuracy. The usage of the L^2 expansion leads to the final channels being a product of an asymptotically plane wave for one (projectile-space) electron and a Coulomb wave for the other (target space). The CCC results are independent of whether the projectile-space electron is represented by a distorted or a plane wave. One may expect that such an asymmetric treatment of the outgoing electrons would yield poor angular distributions in the case of equal energy-sharing kinematics. However, the CCC theory has already obtained excellent agreement with the experimental profiles for all coplanar geometries in the case of 64.6 eV e-He ionization with 20 eV outgoing electrons, though a factor of 1.8 less in magnitude [24]. Similarly, excellent angular distributions were obtained in the case of 32.6 eV incident energy with 4 eV outgoing electrons [25]. Here the initial CCC results were also substantially lower than experiment. However, using the explanation suggested by Bray [26] for the cause of the problem and how it may be remedied, the single factor by which all of the CCC TDCS need to be multiplied may be obtained from the ratio of the CCC singly differential cross section (SDCS), and the true SDCS. For this we require the knowledge of the true SDCS. Using the detailed measurements of Röder *et al.* [28] as a check, we do this by assuming that the true SDCS is well described by a quadratic with the three coefficients given by (i) the required symmetry about $E/2$, (ii) the integral (from 0 to $E/2$) yields the CCC total ionization cross section (agrees with experiment to within a few per cent) and (iii) the SDCS starts at the same value ($2.5 \times 10^{-18} \text{ cm}^2 \text{ eV}^{-1}$) irrespective of the total energy E [25]. The latter condition ap-

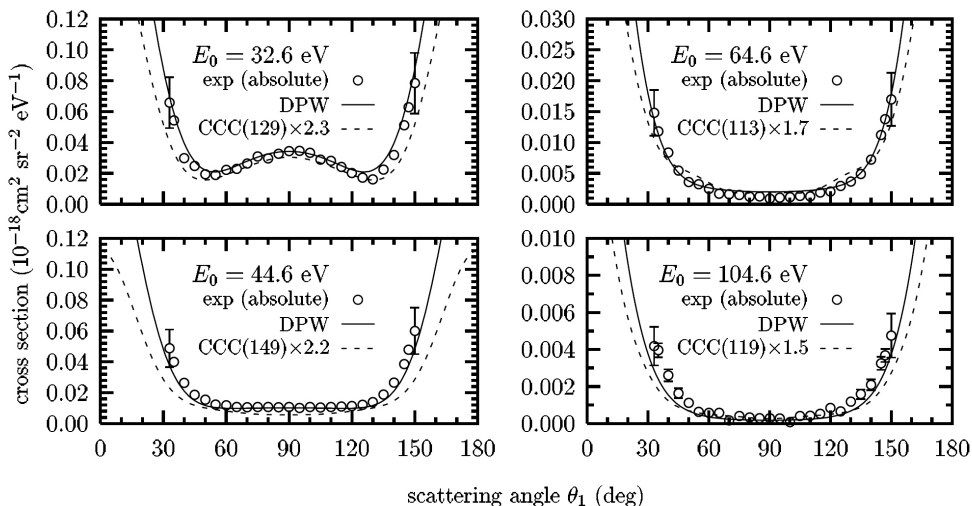


Figure 1. Equal energy sharing $\theta_{12} = \pi$ e-He ($e, 2e$) triply differential cross sections at the indicated projectile energies. See the text for details of the theory and experiment.

appears to work well from near threshold to several hundred eV [15, 28]. The assumption of a quadratic description of the SDCS is only appropriate to low and intermediate energies. The rescaling procedure does not affect the angular distributions, or depend on the geometry of detectors. If, after rescaling, agreement with experiment is found, then it suggests that the CCC theory obtains accurate angular distributions for all possible in- and out-of-plane positioning of detectors. Here we concern ourselves with just the coplanar $\theta_{12} = \pi$ geometry.

Figure 1 shows $\theta_{12} = \pi$ equal energy-sharing e-He TDCS* measurements as well as both DPW and CCC results for incident energies of 32.6, 44.6, 64.6, and 104.6 eV. (The measured and CCC results for 64.6 and 32.6 eV have been presented elsewhere [24, 25]; they are presented here for completeness and to compare with the DPW results.) The measured results are absolute, with statistical (<10%) and absolute ($\approx 25\%$) error bars shown. For all energies, both DPW and CCC results accurately describe the shape of the measured angular distribution. However, whereas the DPW results are in essentially perfect agreement with the measured absolute values (at least for the lowest three energies), the CCC results are in reasonable agreement only after multiplication by the specified factors. At the lowest three energies the factors are obtained by assuming the true SDCS may be described by a quadratic function as discussed above. At 104.6 eV this approximation is insufficiently accurate, so instead we use the doubly differential cross section (DDCS) measurements of Müller-Fiedler *et al.* [27] at the secondary energy of 40 eV. The multiplicative factor of 1.5 yields complete agreement of the CCC and the absolute DDCS measurements and so may also be used to rescale the CCC TDCS. Note that the rescaling procedure only makes sense if the CCC theory obtains correct angular distributions in the full two-electron phase space. Thus far detailed comparison with experiment has suggested that this is indeed the case [24, 25].

* The TDCS is differential in the momenta k_1 and k_2 of the two continuum electrons in the final state. All our results focus on the geometry $\hat{k}_1 = -\hat{k}_2$, in which the electrons depart in opposite directions. Thus $\hat{k}_1 \cdot \hat{k}_2 \equiv \cos \theta_{12} = -1$, from which arises the terminology “ $\theta_{12} = \pi$ geometry.” With respect to the incident electron direction (taken as the z -axis), one may define $\hat{k}_1 \equiv (\theta_1, \varphi_1)$ and $\hat{k}_2 \equiv (\theta_2, \varphi_2)$ in the usual way. In the $\theta_{12} = \pi$ geometry, $\theta_1 + \theta_2 = 180^\circ$ and $\varphi_2 = \varphi_1 + 180^\circ$.

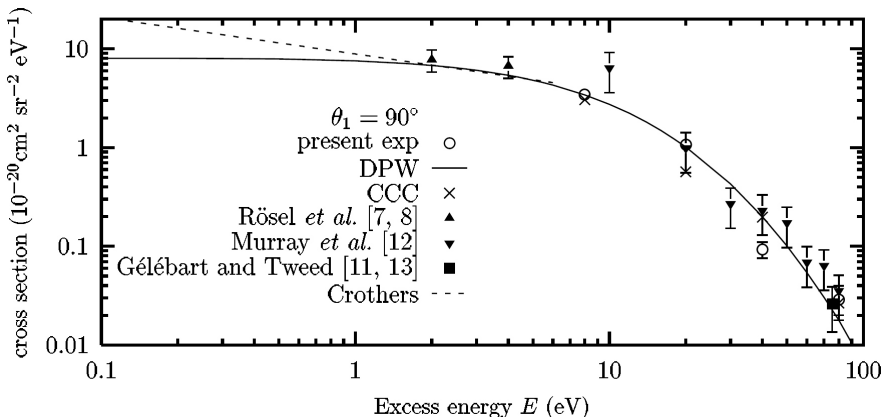


Figure 2. Equal energy sharing e-He ($e, 2e$) triply differential cross sections at $\theta_1 = 90^\circ$ in the Wannier ($\theta_{12} = \pi$) geometry as a function of excess (total) energy. The CCC and experimental results are from Figure 1. Note that the data point of Gélébart and Tweed [11] has been reduced by 18% according to [13].

We note that the measurements at 64.6 eV incident energy lie in the energy region of the doubly excited states of He [29] and that those at 104.6 eV lie in the double-ionization continuum of He. The good agreement of the absolute measurements at these latter two energies with the presented results is rather remarkable since neither theory includes the effects of He doubly excited states or of the influence of ($e, 3e$) processes on the ($e, 2e$) TDCS.

Figure 2 presents the corresponding results over a broad energy range for completeness and to exhibit the trends both near threshold and at intermediate energies. We discuss the latter first. Our new absolute measurements for He in the intermediate energy region are compared with absolute measurement of Gélébart and Tweed [11] at an incident energy of 100 eV ($E_{\text{ex}} = 75.4$ eV) corrected by the 18% reduction specified in [13]. We also make comparisons to the absolute experimental results obtained in perpendicular plane measurements by Murray *et al.* [12] for the configuration $\theta_1 = \theta_2 = 90^\circ$ and equal energy sharing. (We emphasize that the Murray *et al.* [12] results were put on an absolute basis in part by normalizing to the (incorrect) data point of [11] and hence require revision. We nevertheless present these here because they are the only published absolute data of which we are aware in the range of the current measurements for the $\theta_{12} = \pi$ configuration.) These absolute experimental results are compared with our DPW and CCC theoretical results. Throughout the intermediate energy region shown, the DPW results agree on the whole very well with the absolute data. The CCC results, after rescaling, reproduce the experimental trend, with the exception of the point at 20 eV. Note, in particular, that the newly measured point at $E_{\text{ex}} = 8$ eV is in excellent agreement with the DPW prediction, in contrast to the measured point of [12] at $E_{\text{ex}} = 10$ eV. Note also that for excess energies above about 35 eV, doubly excited states of He may play a role in the obviously increased scatter of the measured points. Likewise, for excess energies above about 54 eV, the influence of ($e, 3e$) processes on the ($e, 2e$) TDCS may contribute to the values measured by experiment. Neither doubly excited states nor ($e, 3e$) processes are taken into account in the present calculations.

In the near-threshold energy region, Figure 2 shows the absolute measurements of Rösel *et al.* [7, 8] as well as the calculations for this geometry of Crothers [30].* The DPW results are in excellent agreement with the measured points as well as with results of Crothers' calculations in the energy region between 2 and 4 eV. Below 2 eV, the DPW and Crothers' results disagree owing to their differing dependences on E_{ex} as $E_{\text{ex}} \rightarrow 0$. According to the Wannier-Peterkop-Rau (WPR) theory [31–34] of the threshold law for fragmentation of three-body Coulomb systems, the triply differential cross sections for (e, 2e) processes should vary with energy as $E_{\text{ex}}^{-0.373}$ [1, 33] where E_{ex} is the energy "excess" above threshold. This energy dependence occurs in the WPR theory owing to the predicted rapid narrowing of the width of the distribution with respect to θ_{12} in the region of $\theta_{12} = \pi$. Crothers' calculations are consistent with this WPR prediction. The DPW results, however, which employ an effective screening approximation [20–22], are independent of E_{ex} as $E_{\text{ex}} \rightarrow 0$. Hence, as shown in Figure 2, Crothers' results have a slope of -0.373 , and the DPW results have a slope of 0 as $E_{\text{ex}} \rightarrow 0$. Experimental tests of the WPR threshold law must thus be made in the energy region between threshold and 2 eV above, over which the DPW results and those of Crothers disagree. Clearly the available absolute measurements for He are not yet sufficiently close to threshold to verify the WPR predicted energy dependence.

In summary, we have reported new absolute measurements of the TDCS for (e, 2e) processes for equal energy sharing and the $\theta_{12} = \pi$ geometry in the intermediate energy region for He and compared them with other absolute measurements in the region from threshold to about 100 eV above. As was the case for the first absolute measurements for such processes in He [7, 8], the DPW theory, which we have restricted to the geometry $\theta_{12} = \pi$, owing to the screening approximation it employs, gives accurate predictions for both the angular distributions and the absolute TDCSs for this geometry over the entire energy region from 2–80 eV above threshold. This success underlines the importance of treating distortion and non-local exchange effects in each partial wave for the (e, 2e) process in addition to mutual screening effects. The CCC theory is shown to provide relatively accurate absolute values after rescaling by the difference of the true and the CCC estimated SDCS. This is consistent with the notion that the CCC theory obtains accurate angular distributions for all possible in- and out-of-plane geometries of the two electron detectors. Given the asymmetric treatment of the two equal-energy electrons this would indeed be truly remarkable. The nature of the rescaling is such that there must be some uncertainty associated with the rescaled absolute CCC values. This varies depending on the uncertainty associated with the estimated true or experimentally obtained SDCS. At 64.6 eV, we have the rescaling factor of 1.7, which compares well with the visual fit factor of 1.8 given by Bray *et al.* [24]. The discrepancy at 44.6 eV is somewhat disappointing and invites further detailed investigation.

The available experimental and theoretical results hint also at interesting new physics in two contrasting energy regions: first, in the energy region between about 40 and 100 eV above threshold, further absolute measurements over a finely spaced set of energies may be able to detect influences of doubly excited states and of the double ionization continuum on the (e, 2e) process in He. Second, absolute measurements below 2 eV above threshold are necessary if the predictions of the WPR threshold law for (e, 2e) TDCSs are to be confirmed.

* Note that TDCS results are given here only for 1 and 2 eV excess energies. We have extracted results for other energies using Crothers' figure 1 and equations (74) and (86): the TDCS for $\theta_{12} = \pi$ and $\theta_1 = 90^\circ$ equals $8.9(E_{\text{ex}}/\text{eV})^{-0.373} \times 10^{-20} \text{ cm}^2 \text{ sr}^{-2} \text{ eV}^{-1}$.

Acknowledgments

The work of Drs. Pan and Starace has been supported in part by US National Science Foundation grant no PHY-9722110. Drs. Röder and Ehrhardt have been supported in part by the Deutsche Forschungsgemeinschaft. Drs. Bray and Fursa acknowledge the support of the Australian Research Council, the South Australian Centre for High Performance Computing and Communications, and the Phillips Laboratory, Air Force Materiel Command, USAF, under cooperative agreement number F29601-93-2-0001.

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