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FAST TRACK COMMUNICATION

Deep interference minima in non-coplanar triple differential cross sections for the electron-impact ionization of small atoms and molecules

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Abstract
The time-dependent close-coupling method and a distorted-wave approach are used to explore deep minima discovered in the non-coplanar triple differential cross sections for the electron-impact ionization of helium. This phenomenon has been well studied experimentally but so far has not been investigated by a non-perturbative theoretical approach. We find that our time-dependent calculations reproduce very well the experimental minima, and that the distorted-wave calculations also confirm this phenomenon. Further investigations reveal that the minima appear to be due to deep destructive interference between the partial wave contributions which make up the cross sections. We also show that similar minima may be found in triple differential cross sections arising from the electron-impact ionization of atomic and molecular hydrogen.

(Some figures in this article are in colour only in the electronic version)

Investigations of the electron-impact ionization of small atoms and molecules (commonly known as (e, 2e) processes) continue to further our knowledge of the nature of the three-body Coulomb dynamics inherent in the final state of this process [1]. In recent years, significant progress has been made on the theoretical side, with several theories demonstrating excellent agreement with a variety of experimental data yielding multiple differential cross sections for ionization of H [2–7] and He [8–11], for a variety of outgoing electron geometries and kinematics, and from near threshold to relatively high impact energies [12, 13].

Experimental investigations of (e, 2e) processes in small atoms have been underway for several years, since the earliest pioneering measurements (see, e.g., [14]). A variety of multiple coincidence techniques, together with more sophisticated optimization and computer control of the electron spectrometers, have allowed precise measurements to be conducted over a wider range of electron angles and kinematics [12, 15, 16, 17, 18]. Recently, several of these techniques have been extended to investigate (e, 2e) processes in small molecules at low incident energies, with corresponding theoretical progress now showing reasonable agreement with these new measurements [19–22].

An outstanding puzzle in some of the earlier (e, 2e) measurements [16, 17] was the presence of an unexpected deep and sharp minimum in the triple differential cross sections (TDCS) measured from helium out of the coplanar geometry. The minimum was observed in the 'doubly symmetric' geometry pioneered by the Manchester group and shown in figure 1. The doubly symmetric label refers to the equal

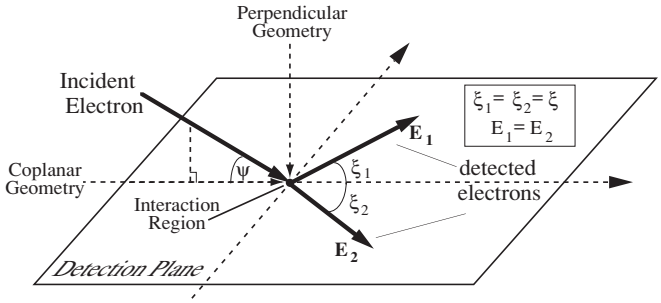

Figure 1. The experimental geometry. The incident electron beam makes an angle ψ with respect to the detection plane defined by the analysers. The analysers rotate through angles ξ_1 and ξ_2 as shown. In the measurements discussed here, $\xi_1 = \xi_2$. A common normalization point exists for all gun angles when $\xi_1 = \xi_2 = 90^\circ$.

energies of the outgoing electrons and the same angle (ξ) made by the outgoing electrons with respect to the axis defined by the incident electron beam. The minimum in the TDCS was most clearly observed for measurements made with an incident electron energy of 64.6 eV (where the outgoing electrons have equal energies of 20 eV), and for a gun angle of $\psi = 67.5^\circ$ (where the gun angle is the angle between the incident electron beam and the detection plane as shown in figure 1). Although the TDCS often is relatively small between the two usual forward and backward scattering peaks, it is not expected to go to zero, as was observed experimentally. Minima were also found to be present for other incident electron energies, and for a neon target ionized from the 2s shell [23, 24]. These minima are unexpected because their positions do not correspond to the 'usual' dips in the TDCS. For example, there is usually a (near) zero in the TDCS for the geometries where both electrons leave along the same direction (due to Coulomb repulsion between the equal energy electrons), and in double photoionization differential cross sections, zeros in the cross section have long been identified as due to selection rules governing the two-electron ejection. The minima observed in the (e, 2e) experiments did not seem to be associated with any known selection rule, and a selection rule argument seemed even more unlikely since the angular position of the minima changed with the incident energy of the electron. Early distorted-wave approaches to the (e, 2e) problem [25] had difficulty reproducing this minimum, but modified 3C (three-Coulomb product wavefunctions) calculations [26, 27], and later modified distorted-wave approaches [24] showed that the TDCS minimum could be ascribed to interference between the various terms which make up the T-matrix. However, these calculations predicted slightly different minima positions than observed experimentally, and also predicted that no minima should be observed for (e, 2e) ionization measurements from atomic hydrogen targets.

In this communication, we explore the minimum in the TDCS from helium. The time-dependent close-coupling (TDCC) [28] method and a three-body distorted-wave (3DW) approach [29, 30] are used to compute the TDCS for a variety of electron energies. TDCC calculations are seen to predict minima in the TDCS in very good agreement with previous measurements. Further investigations also find that, within our partial wave formulation of the (e, 2e) scattering problem, the minimum is due to deep destructive interference between the various partial waves which contribute to the cross section. We also find that the minimum found for helium can also be seen in TDCS from atomic hydrogen (although this is weaker than in helium) and from molecular hydrogen (when the TDCS is considered from a molecule at a fixed orientation with respect to the scattering geometry).

The time-dependent close-coupling theory as applied to electron-impact ionization has previously been well described [11, 28]. The central idea is the propagation of the time-dependent Schrödinger equation for the two outgoing electrons with the interaction between the two electrons treated in full. The remaining electron (in the case of helium and molecular hydrogen) is frozen, and its interaction with the outgoing electrons is represented through direct and local exchange potential terms.

In the TDCC approach, the triple differential cross section for electron-impact ionization of helium is given by

$$\begin{aligned}\frac{\mathrm{d}^3\sigma}{\mathrm{d}E_1\,\mathrm{d}\Omega_1\,\mathrm{d}\Omega_2} &= \frac{\pi}{4k_0^2}\frac{1}{k_1 k_2}\sum_S (2S+1)\int \mathrm{d}k_1 \int \mathrm{d}k_2 \\ &\times \delta\left(\alpha - \tan^{-1}\left[\frac{k_2}{k_1}\right]\right)\left|\sum_L i^L\sqrt{2L+1}\right. \\ &\times \sum_{l_1 l_2}(-i)^{l_1+l_2}\,\mathrm{e}^{\mathrm{i}(\sigma_{l_1}+\sigma_{l_2})}\,\mathrm{e}^{\mathrm{i}(\delta_{l_1}+\delta_{l_2})}\,P^{LS}_{l_1 l_2}(k_1,k_2) \\ &\times \left.\sum_{m_1,m_2} C^{l_1 l_2 L}_{m_1 m_2 0}\,Y_{l_1 m_1}(\hat{k}_1)\,Y_{l_2 m_2}(\hat{k}_2)\right|^2, \end{aligned} \tag{1}$$

where the incident electron energy is $k_0^2/2$, α is the angle in the hyperspherical plane between the two outgoing momenta vectors k_1 and k_2, $Y_{lm}(\hat{k})$ is a spherical harmonic, $C^{l_1 l_2 l_3}_{m_1 m_2 m_3}$ is a Clebsch–Gordan coefficient, and σ_l and δ_l are Coulomb and distorted-wave phase shifts, respectively. Integration of the TDCS over all electron angles and energies recovers the total ionization cross section, where we remember that multiplication by the initial state occupation number is also required (which is 2 for He and H_2). The function $P^{LS}_{l_1 l_2}(k_1,k_2)$ is formed by projecting the final two-electron radial wavefunction (after propagation to a time T) $P^{LS}_{l_1 l_2}(r_1,r_2,t=T)$ onto the one-electron continuum orbitals via

$$\begin{aligned}P^{LS}_{l_1 l_2}(k_1,k_2) = \iint & P_{k_1 l_1}(r_1)\,P_{k_2 l_2}(r_2) \\ & \times P^{LS}_{l_1 l_2}(r_1,r_2,t=T)\,\mathrm{d}r_1\,\mathrm{d}r_2, \end{aligned} \tag{2}$$

where the $P_{kl}(r)$ are box normalized continuum orbitals. We note that, for this highly symmetric geometry, only singlet terms in the expansion in equation (1) contribute to the TDCS. The TDCC calculations discussed below were performed in a similar manner to previously published calculations [11], where details of the numerics of the calculation may be found.

The three-body distorted-wave (3DW) approach to electron-impact ionization of atoms and molecules has been described in detail previously [22, 29–32]. The scattering amplitude is given by

$$\begin{aligned}T = \langle \chi_a^-(\vec{k}_a,\mathbf{r_1})\chi_b^-(\vec{k}_b,\mathbf{r}_2) \\ \times C_{\text{scat-eject}}\left(r_{12}^{\text{ave}}\right)|V-U_i|\phi_j(\mathbf{r}_2)\chi_i^+(\vec{k}_i,\mathbf{r}_1)\rangle, \end{aligned} \tag{3}$$

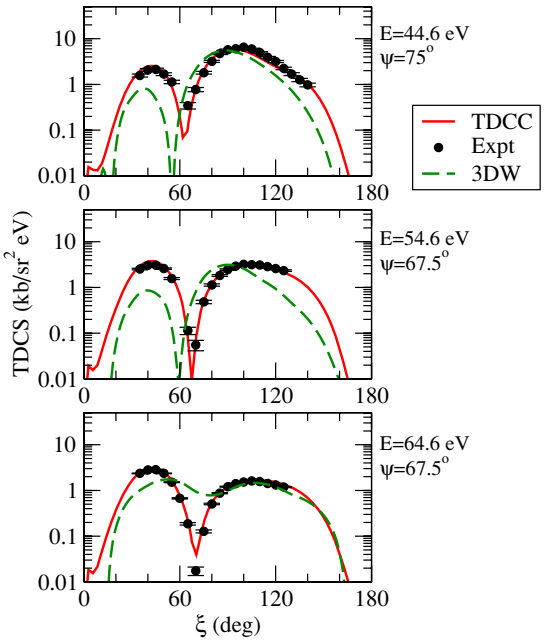

Figure 2. Triple differential cross sections for the electron-impact ionization of helium for three incident electron energies and gun angles as indicated. In all cases, the outgoing electrons have equal energy sharing. The experimental data are compared with TDCC calculations (solid red lines) and with 3DW calculations (dashed green lines) 1.0 kb = 1.0×10^{-21} cm^2.

where distorted-waves χ are used to represent the incident (i), scattered (a) and ejected (b) electrons. The initial bound-state wavefunction is ϕ_j; for He, a Hartree–Fock wavefunction is used, and for H_2, an orientation-averaged molecular wavefunction is used [33]. The factor $C_{\text{scat-eject}}\left(r_{12}^{\text{ave}}\right)$ is the average Coulomb-distortion factor [34], V is the initial state interaction potential between the incident electron and the neutral atom or molecule, and U_i is the initial state distorting potential. In the 3DW calculations presented below for H_2, we present two different sets of calculations, one of which contains a correlation–polarization potential [35, 36] (labelled with CP) and one without this potential (labelled no CP). Inclusion of this potential was found to make a noticeable difference to the H_2 calculations, but made very little difference to the He calculations also presented here.

In figure 2, we compare previous experimental measurements [17] with TDCC and 3DW calculations, where the measurements are normalized to the absolute TDCC calculations. The measurements presented here were all made using the Manchester computer-controlled and computer optimized apparatus, have been fully described previously [15–18] and so will not be discussed again here. We note that, for some of these energies, convergent close-coupling (CCC) calculations also show good agreement with these experimental data [10]. The TDCC calculations are in excellent agreement with the measurements, and the TDCC calculations and experiment clearly display the strong minima in the TDCS. The 3DW calculations for incident energies of 44.6 and 54.6 eV also find the strong minima in the TDCS, although this method predicts the minima position at a slightly different angle than experiment or the TDCC calculations. Puzzlingly, the 3DW calculations at 64.6 eV do not show a minimum, although the cross section does dip in the region of the experimental minimum. The ψ value for which the minima are deepest appears to decrease as the incident energy is increased (subsequent TDCC calculations for $E = 64.6$ eV find a deeper minimum when $\psi = 61.5°$, although there are no measurements at this angle). The ξ angle at which the minima appear also increases as the incident energy increases. The TDCC calculations and experiment also agree extremely well as to the position and heights of the forward and backward peaks in the TDCS.

To further analyse the source of the minima in the TDCS, in figure 3 we present TDCC calculations at $\psi = 61.5°$ (the angle at which the minimum was found to be deepest). The full TDCC calculations in this energy range include partial waves up to $L = 9$ in the expansion in equation (1). Figure 3 shows TDCC calculations which have been truncated at various L values as indicated. Interestingly, it is not until partial waves $L = 2$ and higher are included that the TDCS minimum begins to appear. We also note that by $L = 6$ the cross section is already well converged at this energy.

We can further examine the TDCC calculations by considering the contributions from individual partial waves, as shown by the dashed lines in each panel of figure 4. The black dashed lines show the contributions from each partial wave from $L = 0$ to $L = 8$. The red solid lines show the contribution from the cross terms (or interference terms) which arise in the coherent sum over L in equation (1). For example, the red solid line in the upper middle panel shows the cross term contribution between the partial waves $L = 0$ and $L = 1$. The total cross section can be recovered from this figure by summing each individual partial wave contribution (all black dashed lines) and the final red solid line (lower right panel), which represents the cross term contribution between all partial waves. It is immediately obvious that it is the *interference* between the partial waves which causes the minima in the TDCS (the dotted vertical line indicates the position of the minima in the total TDCS). The individual (or direct) partial wave contributions are significant for $L = 0$–2 and contribute strongly in the region from $\xi \sim 60$ to 120°. The interference terms, however, *destructively* contribute to the TDCS in this region, and as more partial waves are included, the destructive term is enhanced. This explains the findings of figure 3; when contributions from $L = 0, 1$ only are included, the positive contribution from the direct terms is much larger than the destructive interference term, so that no minima is found. However, as more partial waves are included, the destructive contribution becomes larger, leading eventually to the deep minima observed in the full calculations and as found experimentally.

Similar investigations for the other incident energies presented in figure 2 reach the same conclusion: that it is the deep destructive interference between the contributing partial waves in the sum in equation (1) which causes the observed minima in the TDCS. Our findings are consistent with the conclusions of previous 3C calculations [26, 27] that destructive interference is the cause of the TDCS minima.

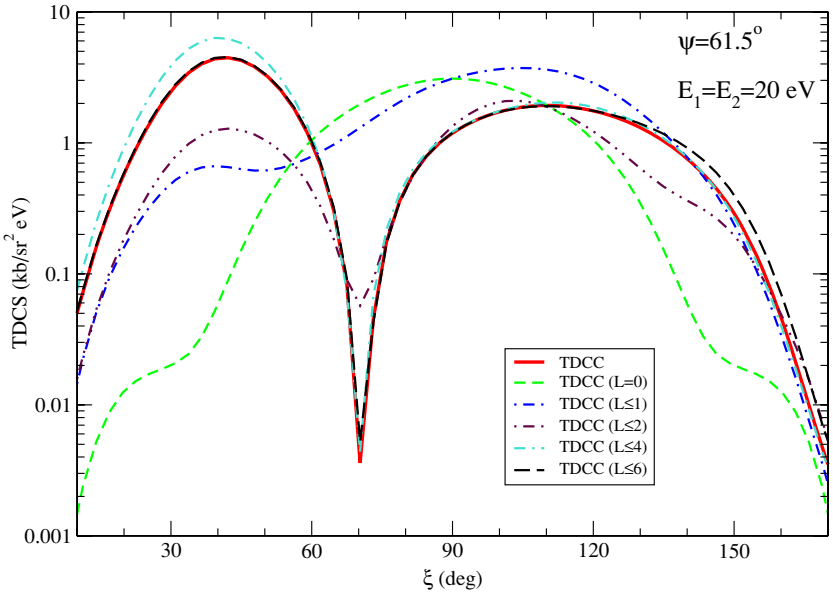

Figure 3. TDCC calculations for the triple differential cross sections for the electron-impact ionization of helium for an incident electron energy of 64.6 eV, at a gun angle of $\psi = 61.5^\circ$, and for equal energy sharing outgoing electrons. A complete TDCC calculation (including $L = 0$–9) is indicated by the solid red line. The various dashed lines show TDCC calculations which include fewer partial wave contributions, as indicated in the caption.

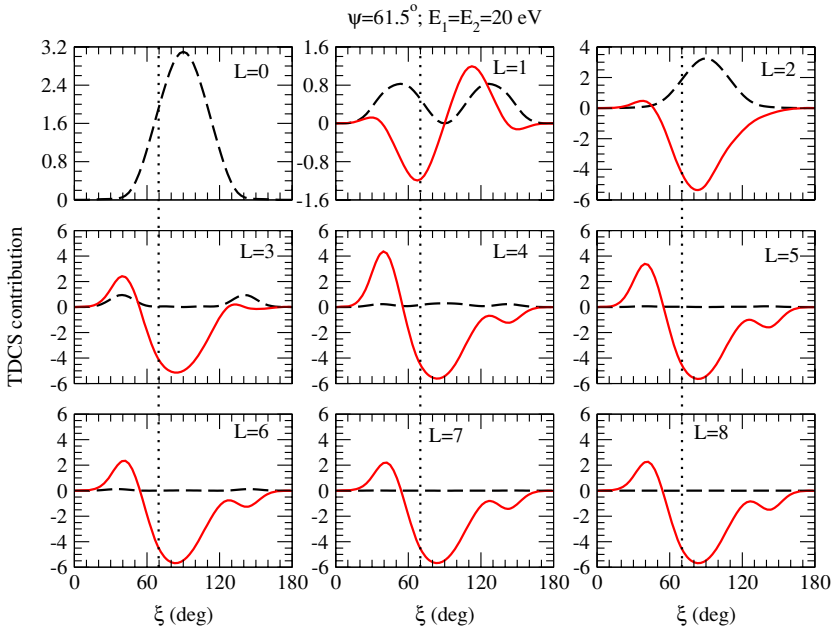

Figure 4. TDCC calculations for the same case as figure 2. Here, we show the contributions from individual partial waves (dashed black lines) in each panel up to $L = 8$. The solid red lines signify the contribution from the interference (cross) terms inherent in the coherent sum in equation (1). For example, the red line in the upper right panel shows the contribution from the $L = 0, 1, 2$ cross terms. The vertical dotted lines indicate the position of the minimum in the TDCS in figure 3.

However, in the previous 3C formulation [27], it was stated that the interference is manifested between the various terms which make up the T-matrix, and that the decisive contribution which gives rise to the minima was the interference due to the T_3 term (which represents the initial scattering off the passive (frozen) electron). In the quite different TDCC approach, in which the initial wavefunction is expanded over partial waves, the destructive contribution arises from interference between the partial wave contributions. However, our 3DW approach, which only includes the T_1 term (scattering off the ionized electron), also finds a clear minimum in the TDCS (the 3DW approach uses orthogonal wavefunctions and so the T_2 term, representing the scattering off the nucleus, is automatically zero). Yet the minimum in the

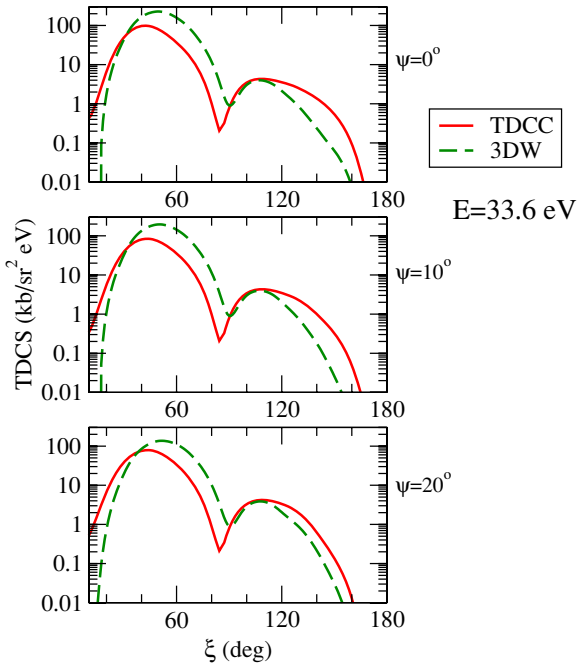

Figure 5. TDCC and 3DW calculations of the triple differential cross sections for the electron-impact ionization of atomic hydrogen at an incident energy of 33.6 eV, for various gun angles as indicated. The $\psi = 0^\circ$ case corresponds to the coplanar geometry.

TDCS found in the 3DW calculations clearly *cannot* be due to interference between the various terms which constitute the T-matrix, since only one term (T_1) exists in our 3DW approach. Berakdar and Briggs [27] also stated that, since in (e, 2e) experiments from atomic hydrogen no T_3 term exists (since there is no passive electron), the deep minima should not be observed. Their 3C calculations of the TDCS for (e, 2e) from atomic hydrogen found a weak minimum at low ψ values. We have now also undertaken TDCC and 3DW calculations of the TDCS for (e, 2e) on atomic hydrogen, as shown in figure 5, although at a different incident energy than [27]. We find that a fairly sharp minimum does exist, which is also most evident for low ψ values close to the coplanar geometry. At this energy sharing ($E_1 = E_2 = 10$ eV), the minimum TDCS does not reach zero, but does reach the lowest value of the TDCS near $\xi = 0$, where the TDCS is very small due to electron–electron repulsion. Similar investigations as for the helium case reveal that the minimum in this atomic hydrogen case is also due to deep destructive interference between the various partial-wave contributions.

As a final demonstration of the minimum in the TDCS, we consider in figure 6 the TDCS from electron-impact ionization of molecular hydrogen at a gun angle of 67.5°, again for equal energy sharing outgoing electrons with $E_1 = E_2 = 10$ eV. Comparisons with experiment (where the molecular orientation is unknown) and theoretical TDCC and 3DW calculations (averaged over all molecular orientations) have previously been published [21, 22]. In figure 6, it is again seen that the agreement between the averaged TDCC calculations and the measurements is excellent, and that the 3DW calculations also are in reasonable agreement with experiment. No minima are observed in the experimental data for randomly oriented molecules. However, by analysing

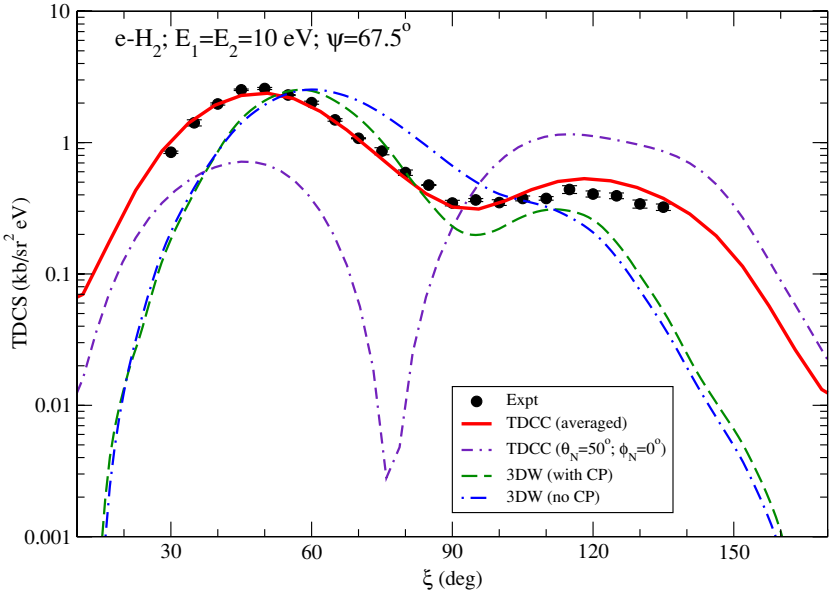

Figure 6. Triple differential cross sections for the electron-impact ionization of molecular hydrogen at an incident energy of 35.4 eV, for equal energy sharing outgoing electrons. Experimental data are compared with TDCC calculations (thick red line) which are averaged over all molecular orientations and two sets of 3DW calculations. The calculations labelled 3DW (with CP) include a correlation-polarization term, while the calculation labelled 3DW (no CP) omits the correlation-polarization term. Both sets of 3DW calculations are divided by 6.3 to allow a better comparison with other results. The double-dashed purple line indicates the TDCC calculation for a specific molecular orientation ($\theta_N = 50^\circ$; $\phi_N = 0^\circ$), where it can be seen that a deep interference minimum is predicted.

TDCC calculations at various fixed molecular orientations, minima in the TDCS *can* be observed. For example, in figure 6, TDCC calculations at a molecular orientation of $\theta_N = 50°$; $\phi_N = 0°$ (where the angles refer to the orientation of the molecule with respect to the z-axis, where the z-axis is defined by the incoming electron beam) find a sharp minimum at a scattering angle ξ around 75°. Since measurements of the TDCS from molecules in which the orientation is known have not yet been made, these calculations represent a prediction for which experimental verification would be highly desirable.

In summary, we have explored the minima in the TDCS first found by (e, 2e) experiments on helium. TDCC and 3DW calculations are in excellent agreement with experiment for helium and reproduce the experimental minima position and depth very well. Analysis of the TDCC calculations finds that the minima appear due to deep destructive interference between the various contributing partial waves which are included in the TDCC calculations. We also demonstrate that a minimum can be observed in the TDCS from atomic hydrogen (although the minima are somewhat weaker than in the helium case). We also find, for the first time, that a minimum is predicted in the TDCS from molecular hydrogen, although only if consideration is made of the TDCS from a molecule with a specific orientation.

Some outstanding questions still remain with regard to the deep minima phenomena. In particular, there is no theoretical guideline as to *where* the minima in the TDCS will occur, i.e. at which specific geometry, such as the selection rules derived for zeros in the TDCS for double photoionization [37]. Although it seems clear that the minima are due to interference effects, there is as yet no obvious physical argument as to why this interference should occur for these particular geometries. However, we have demonstrated that the TDCS minima first observed over 15 years ago appear to be a *general* feature of (e, 2e) studies for the specialized symmetric geometries under consideration, and so it would be extremely interesting to investigate this phenomenon further in more complicated systems [23].

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