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Doubly differential cross sections for the electron impact ionization of hydrogen

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We report the first completely *ab initio* calculations of doubly differential cross sections for the electron impact ionization of hydrogen at incident energies of 17.6 eV, 25 eV, and 30 eV. These cross sections have been extracted from wave functions obtained by directly solving a finite difference discretized Schrödinger equation without explicit reference to any assumed asymptotic form. Outgoing wave boundary conditions are assured by the use of the exterior complex scaling method. Our calculations suggest the need for additional experiments to augment the one available measurement.

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Electron-impact ionization of atoms in the low-energy region (within tens of eV of threshold) has proved to be a sensitive probe of electron correlation effects. Even for the simplest three-body system, the development of fully quantal, nonperturbative methods has been beset with formidable theoretical and computational challenges, and it is only within the last few years that significant progress has been made in the ability of theory to produce accurate, fully differential ionization cross sections at low collision energies. Largely ignored in recent theoretical efforts has been the doubly differential cross section (DDCS), which is differential in energy and the scattering angle of only one electron.

For hydrogen targets, the only DDCS measurements are those of Shyn [1]. Shyn integrated these DDCS over angles to obtain the singly differential (or energy sharing) cross section (SDCS). To our knowledge, there have been only two previous calculations published [2,3] that attempt to compare with this data, but they are either perturbative or model calculations. Here we report the DDCS for hydrogen at several energies computed using wave functions previously employed [4,5] in *ab initio* calculations of triply differential cross sections (TDCS) for equal-energy sharing between the two outgoing electrons. Those calculations showed excellent agreement with experiment. The DDCS calculations presented here also probe the case of asymmetric energy sharing and suggest the need for additional measurements.

The fact that the final ionization state involves three particles interacting via Coulomb forces and that the wave function for such a system is known only in the far asymptotic region where the particles are well separated has prevented anyone to date from solving the Schrödinger equation using the asymptotically correct boundary condition for three-body breakup. For this reason, there has been a significant effort directed toward the development of methods that do not rely on the explicit enforcement of asymptotic boundary conditions. These include time-dependent methods [6,7] in which a known initial state is propagated until the collision interaction is complete. We have developed an alternative approach, in which an exterior complex scaling (ECS) transformation is applied to the time-independent Schrödinger equation. ECS significantly simplifies the boundary conditions by transforming the scattered portion of the full wave function into an exponentially damped outgoing wave. This approach has recently been shown to be capable of providing accurate TDCS for the electron-hydrogen system [4,5] in the difficult region below 50-eV incident electron energy.

The ionization problem has also been studied using two-body, coupled-states approaches in which the target continuum is represented by a discrete set of normalized pseudostates that are then treated as part of an expansion basis in conventional close-coupling formalisms [8]. Both the convergent close coupling (CCC) [9,10] and *R*-matrix plus pseudostates (RMPS) [11] methods have been applied to ionization problems, the former more extensively to the calculation of differential ionization cross sections. While the ability of CCC to calculate total ionization cross sections has been convincingly demonstrated [9,10], accurate differential cross sections have been more difficult to obtain. The SDCS, which measure energy sharing between the two electrons in the final state, computed by CCC, oscillate about the correct values, and consequently the angular distributions, though evidently correct in shape, are not accurate in magnitude. In the special case where both final state electrons have the same energy, there is some evidence that the CCC SDCS is converging to 1/4 the correct value [12–14]. There is also evidence to suggest that this behavior is closely related to the

way the cross sections are constructed from the boundary conditions and is not necessarily indicative of any intrinsic inaccuracy in the wave functions computed by CCC or any other close-coupling method. Indeed, we have recently shown that, starting with an accurate wave function computed by ECS, anomalies can be induced by simply matching that function to a sum of discrete two-body terms [12].

The TDCS measures the energy and scattering angles of both outgoing electrons. The TDCS measurements of Röder [15] are the most complete, with some cross sections determined on an absolute scale [16]. The parameter space for the TDCS is large, and the measurements have generally been taken for geometries that contain the incoming and both outgoing electrons in the same plane. For low collision energies (below 50 eV), the most extensive set of measurements are for the case of equal energy sharing between the electrons. There have been many theoretical calculations on electron impact ionization of hydrogen that attempt to reproduce the measured TDCS values. The majority of these studies are perturbative, distorted-wave calculations, some of which attempt to incorporate aspects of the asymptotically correct, three-body boundary conditions into the treatment [17]. These methods are best suited to high energy, asymmetric energy sharing. At collision energies below 50 eV, where exchange, short-range correlation and postcollision interaction are all important, the results are largely mixed and the magnitudes of the calculated cross sections generally unreliable [18].

For the DDCS in the case of hydrogen, Berakdar and Klar [2] use the approximation developed by Brauner, Briggs and Klar [17], based on an asymptotically correct ansatz for the final-state wave function, to compute the ionization amplitudes. At higher energies, they find reasonable agreement with Shyn [1] except in the forward direction. They present the DDCS at energies as low as 25 eV, but at this energy the agreement with experiment is only within a factor of about 2. Das and Seal [3] used a multiple scattering approach to study the problem. They only presented high-energy results (150 and 250 eV) and achieved only qualitative agreement with experiment. We are also aware of DDCS calculated with the CCC method, but to our knowledge they have not been published.

Our approach is to explicitly obtain the radial components of the scattered wave function for the two-electron system in a coupled angular-momentum representation (we treat the nucleus as infinitely massive) on a large two-dimensional grid. We use an exterior complex scaling transformation [19] of the radial coordinates to properly ensure outgoing boundary conditions on the two electrons, without having to explicitly enforce the Coulomb three-body asymptotic form in computing the wave function. The exterior complex scaling transformation

$$r \rightarrow \begin{cases} r, & r < R_0 \\ R_0 + (r - R_0)e^{i\eta}, & r \geqslant R_0 \end{cases} \tag{1}$$

maps each radial electron coordinate onto a complex contour, but only outside a hyperradius, R_0. Any outgoing wave will decay exponentially on the complex part of the exterior scaling contour in Eq. (1), so we partition the total wave function into an unperturbed initial state term and a scattered wave term,

$$\Psi^{(+)} = \Psi_{\rm sc} + \Phi_0, \tag{2}$$

and write the Schrödinger equation with a driving term for $\Psi_{\rm sc}$:

$$[E - H]\Psi_{\rm sc} = [H - E]\Phi_0. \tag{3}$$

To solve this equation, we expand the scattered wave function in partial waves as

$$\Psi_{\rm sc}(\mathbf{r}_1, \mathbf{r}_2) = \sum_{L \ell_1 \ell_2} \mathcal{Y}^{LM}_{\ell_1 \ell_2}(\hat{\Omega}_1, \hat{\Omega}_2)\, \psi^{L}_{\ell_1 \ell_2}(r_1, r_2), \tag{4}$$

where the coupled spherical harmonics $\mathcal{Y}^{LM}_{\ell_1 \ell_2}(\hat{\Omega}_1, \hat{\Omega}_2)$ are defined in standard texts [20]. Substituting this expansion into Eq. (3) gives a set of coupled equations for the radial functions $\psi^{L}_{\ell_1 \ell_2}(r_1, r_2)$, which we solve on a large two-dimensional complex grid using a high-order finite difference approximation for the second derivatives. This procedure results in systems of as many as five million complex linear equations that we solve on distributed memory, parallel supercomputers using specially crafted iterative algorithms. Details of the numerical implementation can be found in Ref. [5].

With the scattered wave function in hand, we need some prescription for using it to calculate differential ionization cross sections. The approach used here is the same as the one previously employed to compute the TDCS: we compute the outgoing flux through the surface of a hypersphere that lies within the volume of coordinate space where both coordinates are real, and then extrapolate the result to infinite volume. We begin by expressing the radial components of the scattered wave function in hyperspherical coordinates, $\rho = \sqrt{r_1^2 + r_2^2}$ and $\alpha = \tan^{-1}(r_1/r_2)$, and define a generalized, dimensionless flux as

$$\mathbf{f}_{\rho_0}(\alpha, \hat{\Omega}_1, \hat{\Omega}_2) \equiv \mathrm{Im}\left\{ k_i \rho (r_1 r_2 \Psi_{\rm sc})^* \frac{d}{d\rho}(r_1 r_2 \Psi_{\rm sc}) \right\} \Bigg|_{\rho = \rho_0}. \tag{5}$$

The flux is related to the total ionization cross section in the limit $\rho_0 \rightarrow \infty$ by the formula

$$\sigma_{\rm total} = \frac{1}{k_i^2} \int_0^{\pi/2} \int_{4\pi} \int_{4\pi} \mathbf{f}_{\rho_0}(\alpha, \hat{\Omega}_1, \hat{\Omega}_2)\, d\hat{\Omega}_1 d\hat{\Omega}_2 d\alpha \Bigg|_{\rho_0 \rightarrow \infty}. \tag{6}$$

In Eqs. (5) and (6), k_i refers to the momentum of the incident electron. If we denote the incident energy by $E_i = k_i^2/2$ and the outgoing electron energies by E_1 and E_2, then by energy conservation $E = E_1 + E_2 = E_i + E_b$, where E_b is the target energy.

The asymptotic behavior of the scattered wave function correlates to the energy distribution of the outgoing electrons, so in the limit $\rho_0 \rightarrow \infty$, the angles $\hat{\Omega}_1$ and $\hat{\Omega}_2$ are those

of the electrons in the final state and the hyperspherical angle α parametrizes the momentum distribution between the two electrons,

$$k_1 = K\cos\alpha, \tag{7}$$

$$k_2 = K\sin\alpha,$$

where $E = K^2/2$. To calculate the DDCS, we integrate the generalized flux over one set of angles (say those of electron 2) and take the limit $\rho_0 \to \infty$. Substituting Eq. (4) into Eq. (5) and integrating over the ejection angles of electron 2 gives the expression for the DDCS,

$$\sigma(\alpha,\theta_1) = \lim_{\rho_0\to\infty} \frac{1}{k_i} \sum_{\substack{L \ell_1 \ell_2 \\ L' \ell_1'}} \mathrm{Im}\left[\psi^{L'*}_{\ell_1' \ell_2}(\rho,\alpha)\rho \frac{\partial}{\partial\rho} \psi^{L}_{\ell_1 \ell_2}(\rho,\alpha) \right]$$

$$\times \sum_m Y^*_{\ell_1' m}(\theta_1,\phi_1) Y_{\ell_1 m}(\theta_1,\phi_1)$$

$$\times \langle \ell_1' m \ell_2 - m | L' 0 \rangle \langle \ell_1 m \ell_2 - m | L 0 \rangle |_{\rho=\rho_0}, \tag{8}$$

where $\langle \ell_1 m_1 \ell_2 m_2 | LM \rangle$ is a Clebsch-Gordan coefficient [20]. Note that the DDCS is a function of E_1 and θ_1, but for the 1s initial state, is independent of ϕ_1.

Since the flux can only be calculated within the finite volume where both electron coordinates are real, the extrapolation to infinite hyperradius ρ_0 must be performed numerically. The extrapolation procedure we have employed to reach this limit is described in Ref. [5].

The sums in Eq. (8) go over L, L' from 0 to ∞, and all ℓ_1, ℓ_1', ℓ_2, and m allowed by the triangle and projection rules prescribed by the Clebsch-Gordan coefficients [21]. In practice we have to terminate these sums over L and L' at some maximum value $L_{\max}$. Our previous work showed that total angular momentum up to $L = 9$ was sufficient to converge the TDCS for the most significant directions of the outgoing electrons, and we find similar convergence behavior for the DDCS. Figure 1 shows the convergence in L of the DDCS at 25-eV incident energy and 4-eV observed electron energy (E_1). The results are seen to have converged for all angles greater than five degrees. This behavior is typical for the energies we investigated.

Figure 2 shows the comparison between the calculated DDCS and the measurements of Shyn [1] at the lowest incident energy measured, 25-eV, and several values of E_1. The agreement is largely satisfactory, except for some values of E_1 at high and low θ_1. Figure 3 shows the DDCS for 17.6-eV incident energy at several values of E_1. Since the three particles in the final state are structureless, we expect the DDCS to have no sharp features. This is borne out in the calculations, and the trends in the cross sections are smooth and monotonic: at lower E_1 the DDCS increase at high angles, and at higher E_1 they are more forward peaked. Figure 4 presents the DDCS at an incident energy of 30-eV. The same trends seen at lower energies are repeated here, with even stronger forward peaks for larger E_1. As can be seen in

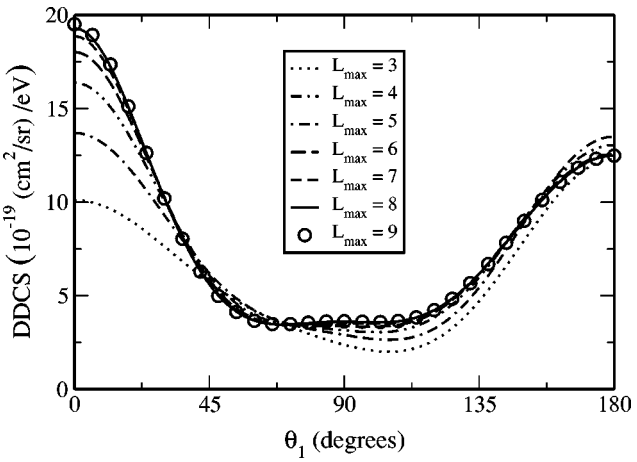

FIG. 1. Convergence in total angular momentum L of the DDCS. The incident energy is 25-eV and the observed electron energy is 4 eV.

Fig. 2, at 25-eV and large or small θ_1, the measurements of Shyn [1] do not follow these smooth trends. For example, the points at the smallest value (12°) of θ_1 are not monotonically increasing with E_1 as one would expect, although the error bars on the measured points do not rule this out.

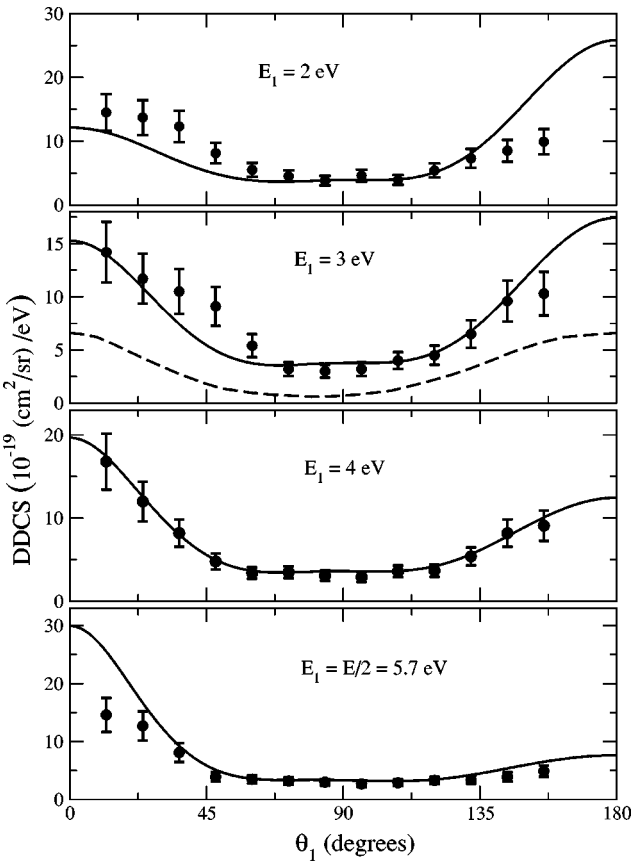

FIG. 2. Doubly differential cross sections for electron impact ionization of hydrogen at an incident energy of 25-eV and several values of the observed electron energy E_1. The circles represent the data of Shyn [1], and the dashed line is the calculation of Berakdar and Klar [2].

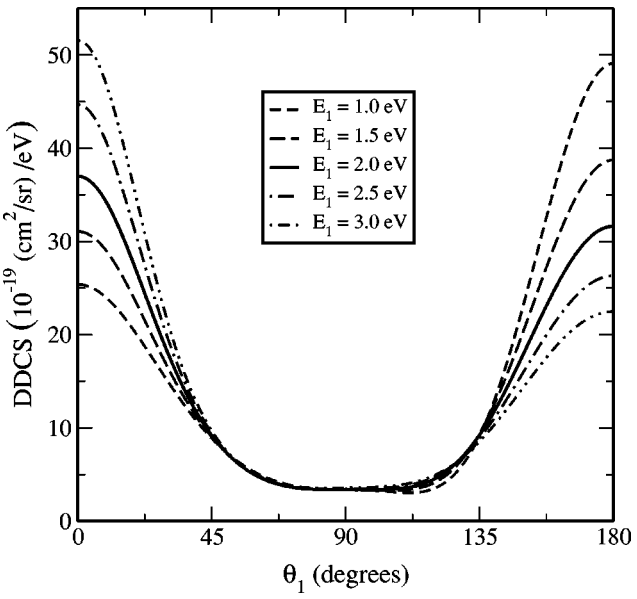

FIG. 3. Doubly differential cross sections for electron impact ionization of hydrogen at 17.6-eV incident energy.

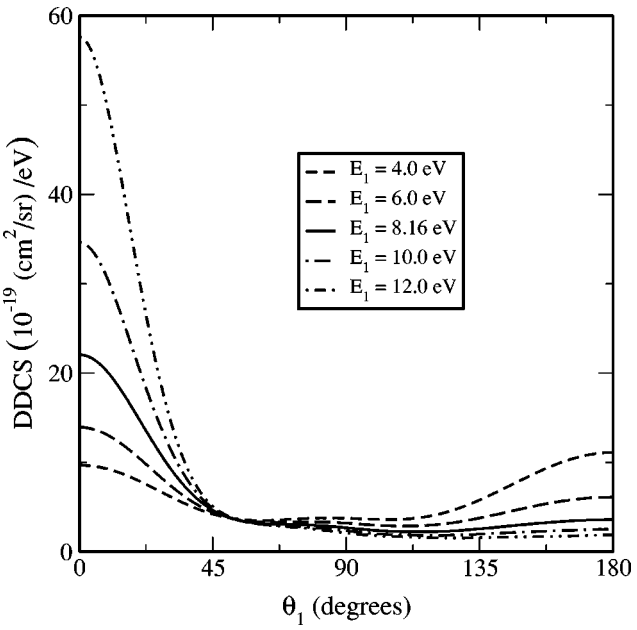

FIG. 4. As in Fig. 3, for 30-eV.

Based on the excellent agreement we have obtained between TDCS extracted from these wave functions and the absolute measurements of Röder, we feel confident in suggesting that the data of Shyn are least accurate at small and large θ_1. The general agreement between those measurements and our calculations is encouraging, but the comparison also suggests that further experiments are called for. The primary goal of Shyn's measurement seems to have been the SDCS. Since the SDCS is symmetric about the equal energy sharing point, Shyn only measured $E_1 \leqslant E/2$, integrating (for each E_1) the DDCS over θ_1 to obtain the SDCS. Since the DDCS does not share this symmetry property, future DDCS measurements should also measure $E_1 > E/2$ for completeness and as a check on the measured SDCS symmetry.

Note added in proof. Recently the CCC calculations of the DDCS were published [21]. The equal-energy sharing DDCS at 25 eV compares favorably to results shown here.

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