Double- and triple-differential cross sections for the low-energy electron-impact ionization of hydrogen

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Double- and triple-differential cross sections are presented for the electron-impact ionization of ground-state hydrogen at incident electron energies of 15.6 and 17.6 eV. The time-dependent close-coupling method is used to calculate the differential ionization cross sections, and comparisons are made with previous theoretical calculations and with experimental measurements. Excellent agreement is obtained between our calculations and previous work.

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I. INTRODUCTION

Recent intense efforts in studies of all measurable quantities from the electron-impact ionization of atomic hydrogen have produced spectacular agreement between theory and experiment for almost all possible cross sections. This topic, which has focused on this simplest ionization process because it is one of the cleanest examples of the classic three-body Coulomb problem, has seen this rapid development in only the last 10–15 years. The total cross section for the electron-impact ionization of hydrogen was measured by Shah *et al.* [1], and soon after several theoretical techniques were developed which produced very good agreement with these absolute experimental measurements. These included the convergent close-coupling (CCC) technique [2], the hyperspherical close-coupling technique [3], the *R*-matrix with pseudostates method [4], and the time-dependent close-coupling (TDCC) method [5]. In the last 10 years the CCC [6], exterior-complex-scaling (ECS) [7–10], and TDCC methods [11] have all demonstrated that accurate triple-differential cross sections can also be produced using these nonperturbative techniques. In the last few years, the CCC and ECS methods have been shown to produce double- [12,13] and triple- [14–16] differential cross sections in very good agreement with each other and with new experimental measurements for electron-impact energies quite close to threshold. We note that nonperturbative techniques are necessary to accurately calculate triple-differential cross sections, since it is known that perturbative distorted-wave methods typically overestimate the total cross section for electron-impact ionization of hydrogen [5] and can also produce unphysical shapes for the triple-differential cross sections [11]. These problems will be exacerbated as the impact energy is lowered.

The purpose of this paper is to show that the time-dependent close-coupling method can also be straightforwardly used to calculate double- and triple-differential cross sections for the electron-impact ionization of hydrogen for impact energies within a few eV of the ionization threshold. Very good agreement is found between our TDCC calculations and the CCC and ECS methods, as well as with the latest experimental measurements. These results are further confirmation of the ability of the TDCC method to accurately solve the three-body Coulomb problem inherent in this, the simplest electron-impact ionization process. Our paper proceeds as follows. In Sec. II we give a brief overview of the theoretical details. In Sec. III we present our results for the electron-impact ionization of hydrogen for impact energies of 15.6 and 17.6 eV, and compare these with results from the CCC and ECS methods, as well as with experiment. We end with a short conclusion.

II. THEORY

The time-dependent close-coupling theory for the electron-impact ionization of hydrogen has previously been presented in some detail [11]. Here we only discuss the main points of our method.

The time-dependent Schrödinger equation for electron scattering from hydrogen can be written as

$$i\frac{\partial\Psi(\vec{r}_1,\vec{r}_2,t)}{\partial t} = H(\vec{r}_1,\vec{r}_2)\Psi(\vec{r}_1,\vec{r}_2,t), \tag{1}$$

where the time-independent Hamiltonian is given by

$$H(\vec{r}_1,\vec{r}_2) = -\frac{1}{2}\nabla_1^2 - \frac{1}{2}\nabla_2^2 - \frac{1}{r_1} - \frac{1}{r_2} + \frac{1}{|\vec{r}_1-\vec{r}_2|}. \tag{2}$$

The total wave function for the two electrons may be expanded in coupled spherical harmonics and projected onto the time-dependent Schrödinger equation to obtain the following set of partial differential equations for each *LS* symmetry:

$$i\frac{\partial P^{LS}_{l_1l_2}(r_1,r_2,t)}{\partial t} = T_{l_1l_2}(r_1,r_2)P^{LS}_{l_1l_2}(r_1,r_2,t) + \sum_{l_1'l_2'} U^{L}_{l_1l_2,l_1'l_2'}(r_1,r_2)P^{LS}_{l_1'l_2'}(r_1,r_2,t), \tag{3}$$

where

$$T_{l_1l_2}(r_1,r_2) = -\frac{1}{2}\frac{\partial^2}{\partial r_1^2} - \frac{1}{2}\frac{\partial^2}{\partial r_2^2} + \frac{l_1(l_1+1)}{2r_1^2} + \frac{l_2(l_2+1)}{2r_2^2} - \frac{1}{r_1} - \frac{1}{r_2} \tag{4}$$

and the coupling operator is given by

$$U^{L}_{l_1 l_2, l_1' l_2'}(r_1, r_2) = (-1)^{L+l_2+l_2'} \times \sqrt{(2l_1+1)(2l_1'+1)(2l_2+1)(2l_2'+1)} \times \sum_{\lambda} \frac{r^{\lambda}_{<}}{r^{\lambda+1}_{>}} \begin{pmatrix} l_1 & \lambda & l_1' \\ 0 & 0 & 0 \end{pmatrix} \times \begin{pmatrix} l_2 & \lambda & l_2' \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} L & l_2' & l_1' \\ \lambda & l_1 & l_2 \end{Bmatrix}. \quad (5)$$

The two-electron radial functions of Eq. (3) at time $t=0$ are constructed as

$$P^{LS}_{l_1 l_2}(r_1, r_2, t=0) = \sqrt{\frac{1}{2}} [G_{k_1 l_1}(r_1)\delta_{0,l_2} P_{1s}(r_2) + (-1)^{S} \delta_{0,l_1} P_{1s}(r_1) G_{k_2 l_2}(r_2)], \quad (6)$$

where k is the linear momentum and $G_{kl}(r)$ is a radial wave packet. The coupled equations (3) are then propagated according to the usual time-dependent close-coupling prescription, for each LS symmetry. At an appropriate time $t=T$ after the collision, in which only outgoing waves are present in each channel, the wave function in momentum space for each LS symmetry is given by

$$P^{LS}_{l_1 l_2}(k_1, k_2) = \int \int P_{k_1 l_1}(r_1) P_{k_2 l_2}(r_2) P^{LS}_{l_1 l_2}(r_1, r_2, t=T) dr_1 dr_2, \quad (7)$$

where $P_{kl}(r)$ are single-particle continuum channels that are appropriately normalized as

$$P_{kl}(r) \to \sqrt{\Delta k} \sin\left(kr + \frac{q}{k} \ln(2kr) - \frac{l\pi}{2} + \sigma_l \right), \quad (8)$$

where σ_l is the Coulomb phase shift, Δk is the momentum mesh spacing, and q is the asymptotic charge.

This momentum-space wave function allows us to simply define the differential cross sections. The triple-differential cross section is given by

$$\frac{d^3\sigma}{d\alpha d\Omega_1 d\Omega_2} = \frac{\pi}{4k^2} \sum_{S} (2S+1) \frac{2}{\pi} \int dk_1 \frac{2}{\pi} \times \int dk_2 \delta\left(\alpha - \tan^{-1}\left[\frac{k_2}{k_1} \right] \right) \times \left| \sum_{L} i^{L} \sqrt{2L+1} \sum_{l_1 l_2} (-i)^{l_1+l_2} e^{i(\sigma_{l_1}+\sigma_{l_2})} \times P^{LS}_{l_1 l_2}(k_1, k_2) \sum_{m_1, m_2} C^{l_1 l_2 L}_{m_1 m_2 0} Y_{l_1 m_1}(\hat{k}_1) Y_{l_2 m_2}(\hat{k}_2) \right|^2, \quad (9)$$

where in this case 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, and $C^{l_1 l_2 l_3}_{m_1 m_2 m_3}$ is a Clebsch-Gordan coefficient. The double-differential cross section in angle is obtained by integrating the triple-differential cross section over one of the outgoing electron angles Ω.

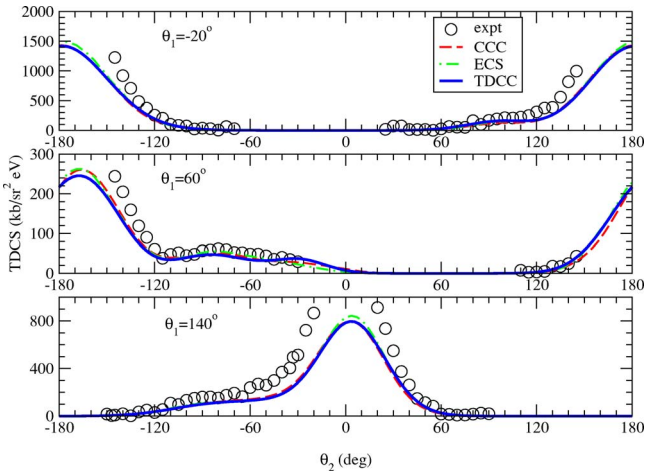

FIG. 1. (Color online) Triple-differential cross sections for hydrogen at an incident electron energy of 17.6 eV, for various fixed angles of one of the ejected electrons, as indicated. The excess energy is equally shared between the two electrons—i.e., $E_1=E_2=2$ eV. Our time-dependent close-coupling calculations are compared with the experimental measurements of Röder *et al.*, convergent close-coupling calculations, and exterior-complex-scaling calculations (all from Ref. [14]) (1.0 kb$=1.0\times10^{-21}$ cm^2).

III. RESULTS

Our time-dependent close-coupling calculations were carried out using a mesh spacing of $\Delta r=0.2$ a.u. and with a box size of 192 a.u. Such a large box size was found to be necessary to fully converge the triple-differential cross sections, which, for certain electron geometrical arrangements, were found to be slowly convergent. The number of LS symmetries used was $L=7$, which was found to be sufficient to fully converge the triple-differential cross sections at these low incident electron energies. The convergence of the differential cross sections for individual LS symmetries was also monitored as a function of the number of $l_1 l_2$ pairs included in the calculation. The number of $l_1 l_2$ pairs used for the TDCC calculations presented here ranged from 6 for $L=0$ to 36 for $L=7$. For the low impact energies presented here, long propagation times were also required to fully converge the cross sections.

We present calculations using our TDCC method of triple-differential cross sections for the electron-impact ionization of hydrogen at 15.6 and 17.6 eV incident electron energies, for cases where the excess energy is equally shared between the outgoing electrons. At an incident electron energy of 17.6 eV, our TDCC calculations are compared with experimental measurements and CCC and ECS calculations from Röder *et al.* [14] in Figs. 1–3. Figure 1 shows the comparison between our calculations and previous work for the case where the angle of the first electron is held fixed and the cross section measured as a function of the second electron. The agreement between all three theoretical techniques is excellent for all three θ_1 values considered, and for the most part, very good agreement is also seen with experiment. In Fig. 2 we present similar comparisons for cases where the angle between

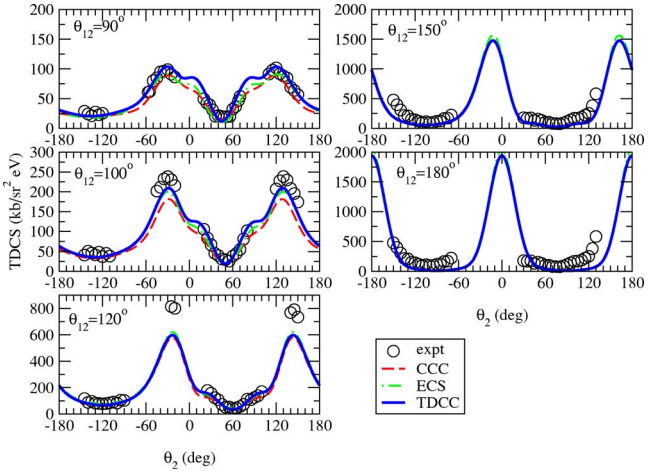

FIG. 2. (Color online) Same as Fig. 1, except that for these cases the cross section is measured as the angle between the ejected electrons (θ_{12}) is held fixed.

the outgoing electrons θ_{12} is held fixed. Again, excellent agreement is observed between all three theoretical calculations. Very good agreement with experiment is also found. The worst agreement is perhaps for the case of $\theta_{12}=90°$, where even the largest measured cross sections are relatively small compared to those in other geometries. Even in this case, the experimental measurements do not favor any of the theoretical results. In Fig. 3 we show similar triple-differential cross sections, this time for the case where the cross section is measured such that the angle of one ejected electron is equal to the angle of the other ejected electron, but on opposite sides of the scattering plane—i.e., $\theta_1=-\theta_2$. In this case the agreement is still very good between all theoretical methods, although perhaps not as spectacular as in Figs. 1 and 2. All three calculations are somewhat below the peaks of the experimental measurements, which, for all cases, have an uncertainty of 40%. This uncertainty arises from the determination of the absolute value of the cross sections [14,17].

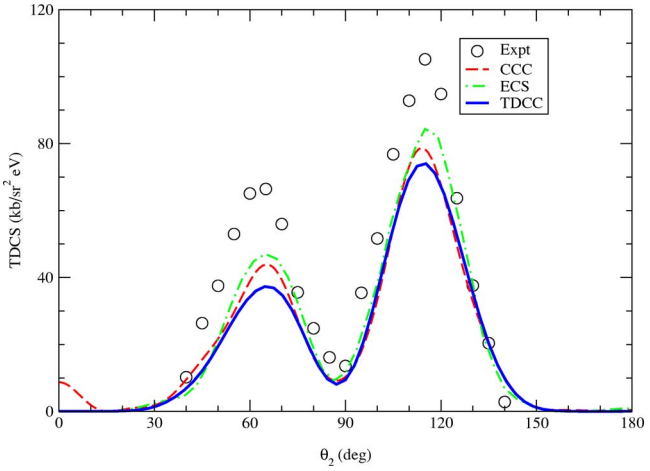

FIG. 3. (Color online) Same as Fig. 1, except that in this case the cross section is measured such that angle of one of the ejected electrons is equal to the angle of the other ejected electron, but on a different side of the scattering plane ($\theta_1=-\theta_2$).

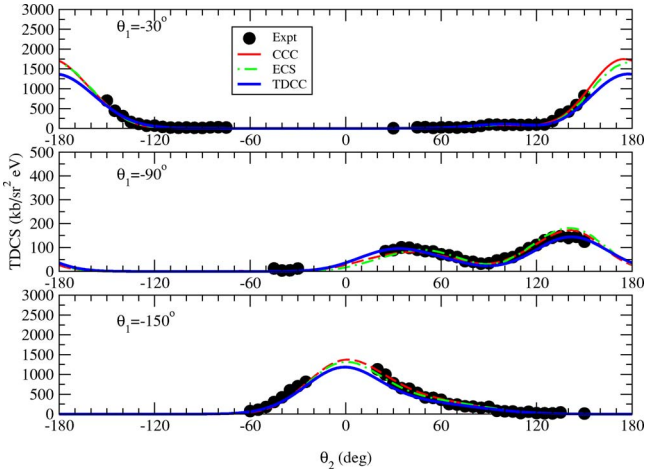

FIG. 4. (Color online) Triple-differential cross sections for hydrogen at an incident electron energy of 15.6 eV, for various fixed angles of one of the ejected electrons, as indicated. The excess energy is equally shared between the two electrons—i.e., $E_1=E_2=1$ eV. Our time-dependent close-coupling calculations are compared with the experimental measurements of Röder *et al.* (Ref. [16,17]), convergent close-coupling calculations (Ref. [15]), and exterior-complex-scaling calculations (Ref. [18]) (1.0 kb=1.0 $\times 10^{-21}$ cm^2).

In Figs. 4–6 we show similar comparisons of our TDCC calculations, again with experimental measurements [16] and with CCC [15] and ECS calculations [18], for an incident energy of 15.6 eV. The triple-differential cross sections shown here are again for the equal energy sharing case of $E_1=E_2=1$ eV. As first pointed out by Bray [19] and further discussed subsequently [15,18], we divide the experimental measurements [16] by a factor of 2 to account for improper experimental normalization. Figure 4 shows the case where the angle of the first electron is held fixed. Again very good agreement is observed between our TDCC calculations and with experiment and with the previous CCC and ECS calculations. Only for the most extreme angles of the first case shown

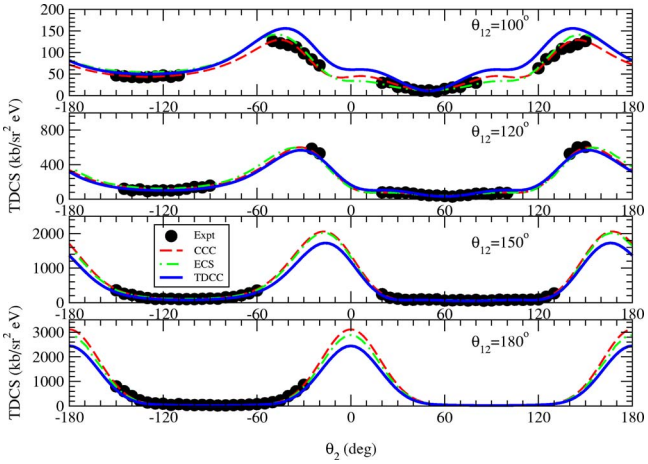

FIG. 5. (Color online) Same as Fig. 4, except that for these cases the cross section is measured as the angle between the ejected electrons (θ_{12}) is held fixed.

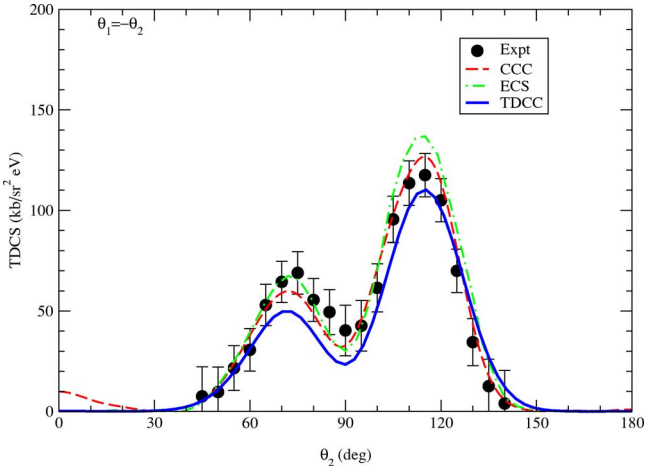

FIG. 6. (Color online) Same as Fig. 4, except that in this case the cross section is measured such that the angle of one of the ejected electrons is equal to the angle of the other ejected electron, but on a different side of the scattering plane ($\theta_1=-\theta_2$).

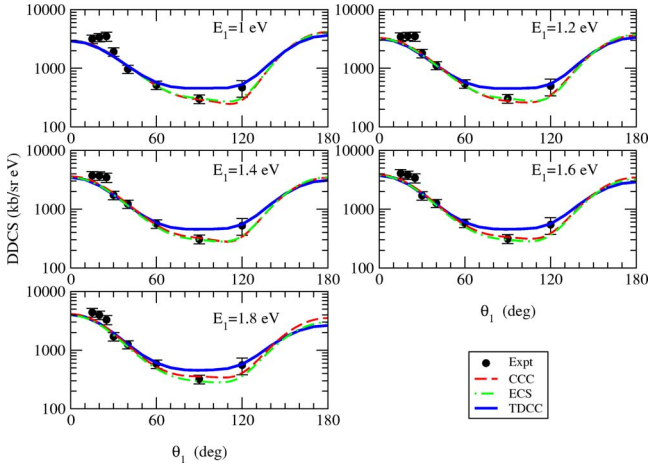

FIG. 7. (Color online) Double-differential cross sections for the electron-impact ionization of hydrogen at an incident electron energy of 15.6 eV, for various energy sharings between the electrons, as indicated. Our time-dependent close-coupling calculations are compared with the experimental measurements of Childers *et al.* (Ref. [13]), as well as convergent close-coupling and exterior-complex-scaling calculations (Ref. [13]).

($\theta_1=30°$) do our TDCC calculations fall slightly below the peaks predicted by the CCC and ECS calculations. Figure 5 shows again the case where the angle between the outgoing electrons θ_{12} is held fixed. Very good agreement is again found, for the most part, although the TDCC calculations predict a smaller peak in the triple-differential cross section for $\theta_{12}=150°$ and $\theta_{12}=180°$. Also, the TDCC calculations are slightly higher than experiment and the CCC and ECS calculations for $\theta_{12}=100°$, although we again note that for this geometry even the largest measured cross section is relatively small compared with other geometries. Finally, in Fig. 6 we present the triple-differential cross sections for the case where the cross section is measured such that the angle of one ejected electron is equal to the angle of the other ejected electron, but on opposite sides of the scattering plane. Here we again find that our TDCC calculations are somewhat lower than the previous ECS and CCC calculations. However, the difference between the TDCC and CCC calculations is roughly the same as the difference between the CCC and ECS calculations. For the most part, the TDCC calculations fall within the error bars of the experimental measurements, with the exception of the first peak near $\theta_2=75°$, where the TDCC calculations are just below the error bars.

In Figs. 7 and 8 we present comparisons of TDCC calculations for double-differential cross sections at the same incident electron energies of 15.6 and 17.6 eV. Again we compare with previous CCC and ECS calculations [13] and also recent experimental measurements [13]. These results are presented for both equal- and unequal-energy sharing between the electrons, where the various values for the energies of the first electron are indiated in the figure. The energy of the second electron can of course be simply found from $E_2=E-E_1$ where E is the excess energy available to the outgoing electrons. Again, for this particular quantity, we find the agreement between our TDCC calculations and the previous work to be very good. The only small difference between our TDCC calculations and the ECS and CCC calculations is that the TDCC calculations appear to overestimate the double-differential cross section for values of θ_1 around 90°. However, for all other angles and for all energy sharings, the TDCC calculations typically go right through the experimental measurements and are in very good agreement with the previous theoretical calculations.

IV. SUMMARY

In this paper we have presented time-dependent close-coupling calculations for double- and triple-differential cross sections arising from the electron-impact ionization of atomic hydrogen at low impact electron energies. We find that the time-dependent close-coupling method is able to accurately predict the double- and triple-differential cross sec-

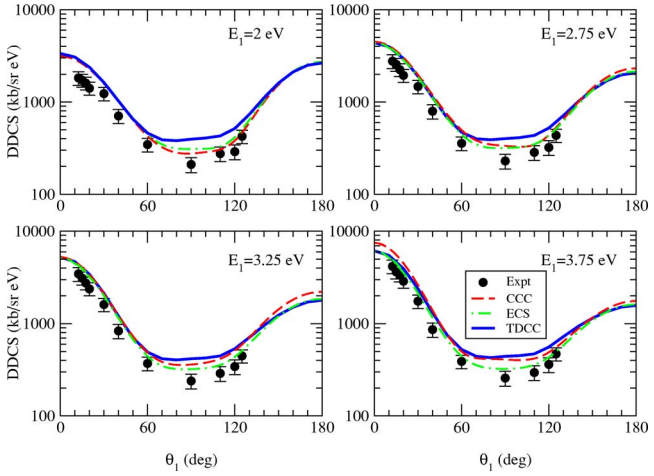

FIG. 8. (Color online) Same as Fig. 7, for an incident electron energy of 17.6 eV.

tions, strongly indicating that this method, along with other nonperturbative convergent close-coupling and exterior-complex-scaling techniques, properly treats the electron-electron interactions between the two outgoing electrons. The good agreement between three methods and experiment shows that the Coulomb three-body problem in atomic scattering is very close to being a completely solved problem by reduction to numerical computation. We finally remark that similarly good agreement between the TDCC and CCC methods has been recently found for all differential cross sections resulting from the electron-impact single ionization of helium [20]. We are now using the TDCC method to focus on the few-body Coulomb dynamics found in even more complex scattering problems, such as the electron-impact double ionization of atoms [21,22] and the electron-impact single ionization of small molecules [23,24].

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