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Ejected-energy differential cross sections for the near-threshold electron-impact ionization of hydrogen

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We have calculated, independently, the ^1S singly differential cross section for the electron-impact ionization of hydrogen at an incident energy of 4.0 eV above threshold using two different methods: time-dependent close coupling and exterior complex scaling. The absolute value of the ^1S cross section for equal energy sharing is critical in assessing recent theoretical and experimental results for coplanar triply differential cross sections at low energies. Convergence of the cross section is studied as a function of the number of coupled channels. We find that at this energy the singly differential cross section is relatively flat over the entire ejected-energy range as predicted by semiclassical calculations.

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Nonperturbative methods developed to treat the electron-impact ionization of simple atoms are generally in excellent agreement for total ionization cross sections over a wide range of incident energies. For hydrogen, the convergent close-coupling [1], the hyperspherical close-coupling [2], the *R*-matrix pseudostates [3], and the time-dependent close-coupling [4] methods yield results that are all within the error bars of the total cross-section measurements of Shah *et al.* [5]. For helium, the convergent close-coupling [6], the *R*-matrix pseudostates [7], and the time-dependent close-coupling [8] methods also yield results that are all in excellent agreement with the total cross-section measurements of Montague *et al.* [9].

However, nonperturbative calculations for differential cross sections in ejected-energy and electron emission angles may sometimes yield surprising differences. Recently, the exterior complex scaling (ECS) [10,11] and the convergent close-coupling (CCC) [12,13] methods were used to analyze the coplanar triply differential cross-section measurements of Röder *et al.* [14,15] for the electron-impact ionization of hydrogen at low incident energies. The measurements were made for equal-energy sharing between the ejected electrons. The ECS calculations [11] are in excellent agreement with the relative triply differential cross-section (TDCS) measurements of Röder *et al.* [14] at incident electron energies of 17.6, 20, 25, and 30 eV, over the full range of scattering angles. The CCC calculations of Bray [12] yield TDCS values that agree reasonably well with the ECS results in shape, but not in absolute magnitude. In particular, for the dominant ''Wannier'' geometries, where the electrons are ejected in nearly opposite directions, the CCC results are roughly a factor of 2 smaller than the ECS results at 17.6 eV; at 30 eV, the ratio is $\sim$1.5. Unfortunately, the absolute measurements of Röder *et al.* [15] at 17.6 eV cannot be used to settle the discrepancy because, as Bray [12] has pointed out, there are internal inconsistencies in the 17.6 eV data that cast doubt on its absolute normalization.

Differences between ECS and CCC can be traced to values of the singly differential cross sections (SDCS's) calculated at equal-energy sharing, where CCC predicts smaller values than ECS, the differences again approaching a factor of 2 at 17.6 eV. This discrepancy is particularly troubling since it has been claimed that, for equal-energy-sharing kinematics, CCC should converge uniformly to 1/4 the true cross section [12,13]. Thus the ''raw'' CCC values are always multiplied by 4 before comparing to experiment. For unequal-energy sharing, close-coupling methods produce oscillatory SDCS values that are not symmetric about half the total energy [16], so smoothing of the data is required. It is noteworthy that the evidence suggesting convergence of CCC at $E/2$ is largely empirical since formal work on this subject has been carried out only for short-range potentials [17] and a simplified, *s*-wave model of e-H ionization [18].

The purpose of this Rapid Communication is to help resolve the question of which theoretical method has produced the correct low-energy results by using independent methods to calculate the ejected-energy SDCS for electron-impact ionization of hydrogen at 17.6 eV incident energy. To focus the comparison, we study only the $L=0$, $S=0$ component of the full e-H problem. (Incidentally, Rost [19] has shown that for $E\rightarrow 0$ the classical dynamics for all fixed values of total angular momentum L collapse to the same effective Hamil-

tonian.) Here we compare two independent calculations. One uses a time-dependent close-coupling (TDCC) approach and the other uses the ECS method. These two methods are quite different, but they do share one significant common feature. Neither method relies on detailed specification of asymptotic boundary conditions in computing the quantities from which the SDCS is ultimately extracted.

The time-dependent wave-packet expression for the electron-impact ionization of hydrogen for ^1S scattering ($L=0$, $S=0$), is given by [4,8] (in atomic units)

$$\sigma=\frac{\pi}{k_i^2}\int_0^E\frac{d\epsilon_e}{k_e k_f}\mathcal{P}(l_i,l_e,l_f,k_i,k_e,k_f), \tag{1}$$

where the linear momenta (k_i,k_e,k_f) and angular-momentum quantum numbers (l_i,l_e,l_f) correspond to the incoming, ejected, and outgoing electrons, respectively. Note that the spin-averaged cross section would include a factor of 1/4 in this expression. The total energy is $E=\epsilon_i+I=\epsilon_e+\epsilon_f$, where I is the ionization energy of hydrogen and $\epsilon=k^2/2$. The scattering probability is given by a projection of the wave packet onto continuum orbitals $\overline{P}_{kl}$:

$$\begin{aligned}\mathcal{P}(l_i,l_e,l_f,k_i,k_e,k_f)&\\ =\left|\int_0^\infty dr_1\int_0^\infty dr_2\overline{P}_{k_e l_e}(r_1)\overline{P}_{k_f l_f}(r_2)\right.&\\ \left.\times P^{l_i}_{l_e l_f}(r_1,r_2,t=T)\right|^2.&\end{aligned} \tag{2}$$

The two-dimensional radial wave function $P^{l_i}_{l_1 l_2}(r_1,r_2,t)$ is a solution to the time-dependent radial Schrödinger equation, which is given by

$$\begin{aligned}i\frac{\partial P^{l_i}_{l_1 l_2}(r_1,r_2,t)}{\partial t}&=T_{l_1 l_2}(r_1,r_2)P^{l_i}_{l_1 l_2}(r_1,r_2,t)\\ &+\sum_{l_1',l_2'}U_{l_1 l_2,l_1' l_2'}(r_1,r_2)P^{l_i}_{l_1' l_2'}(r_1,r_2,t),\end{aligned} \tag{3}$$

where

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

and the coupling operator is given by

$$\begin{aligned}U_{l_1 l_2,l_1' l_2'}(r_1,r_2)=&(-1)^{l_1+l_1'}\delta_{l_1,l_2}\delta_{l_1',l_2'}\\ &\times\sqrt{(2l_1+1)(2l_1'+1)}\sum_\lambda\frac{r_<^\lambda}{r_>^{\lambda+1}}\\ &\times\begin{pmatrix}l_1&\lambda&l_1'\\0&0&0\end{pmatrix},\end{aligned} \tag{5}$$

where we note that this expression has been greatly simplified for the ^1S case. The initial condition in this case is given by

$$\begin{aligned}P^0_{l_1 l_2}(r_1,r_2,t=0)=\delta_{l_1,0}\delta_{l_2,0}\sqrt{\frac{1}{2}}[&\overline{P}_{1s}(r_1)G_{k,s}(r_2)\\ &+G_{k_i s}(r_1)\overline{P}_{1s}(r_2)],\end{aligned} \tag{6}$$

where $G_{k_i s}(r)$ is a radial wave packet defined by

$$G_{ks}(r)=Ce^{-ikr}\exp\left[-\frac{(r-r_0)^4}{w^4}\right], \tag{7}$$

and C is a normalization constant. The wave function at time $t=T$ following the collision is obtained by propagating the Schrödinger equation on a two-dimensional spatial lattice. The bound $\overline{P}_{ns}(r)$ and continuum $\overline{P}_{ks}(r)$ orbitals needed in the above equations are obtained by diagonalizing the single particle Hamiltonian,

$$h(r)=-\frac{1}{2}\frac{\partial^2}{\partial r^2}-\frac{1}{r}, \tag{8}$$

on a one-dimensional spatial lattice.

In the exterior complex scaling method we solve the time-independent Schrödinger equation by calculating Ψ_{sc}, the scattered portion of the full wave function $\Psi^{(+)}$,

$$\Psi^{(+)}=\Psi_{sc}+\Phi_0, \tag{9}$$

where Φ_0 is the initial, unperturbed state,

$$\Phi_0=\frac{1}{\sqrt{k_i}}[e^{i\mathbf{k}_i\cdot\mathbf{r}_1}\overline{P}_{1s}(r_2)+e^{i\mathbf{k}_i\cdot\mathbf{r}_2}\overline{P}_{1s}(r_1)]. \tag{10}$$

The scattered wave satisfies the driven Schrödinger equation,

$$[E-H]\Psi_{sc}=[H-E]\Phi_0. \tag{11}$$

Under ECS the radial coordinates for both electrons are transformed so that they become complex beyond a certain distance R_0,

$$r\rightarrow\begin{cases}r, & r<R_0,\\ R_0+(r-R_0)e^{i\eta}, & r\geq R_0.\end{cases} \tag{12}$$

Under the transformation in Eq. (12), all outgoing waves become exponentially damped functions beyond R_0. Thus, solving Eq. (11) with ECS ensures that the purely outgoing wave solution will be calculated. Just as in the TDCC

method, the angular variables are eliminated from Eq. (11) by expanding Ψ_{sc} in coupled spherical harmonics. The result is a set of coupled, two-dimensional radial equations that were solved on a numerical grid using high-order finite difference.

Once the scattered wave is in hand, there are several options available for computing the SDCS. In our original implementations of ECS [10,11], we relied on a direct evaluation of the quantum-mechanical flux through the finite (hyper)sphere that bounds the physical region where the electron radial coordinates are real, followed by numerical extrapolation of the calculated flux to infinite hyperradius. In this work, we compute the SDCS from an integral expression for the ionization amplitude that was recently described and tested on the Temkin-Poet and colinear models of *e*-H ionization [20]. Using standard rearrangement theory, we express the ionization amplitude as

$$f(k_e,k_f)=\left\langle \phi_{k_e}^{(-)}\phi_{k_f}^{(-)}\left|E-T-\frac{1}{r_1}-\frac{1}{r_2}\right|\Psi_{sc}\right\rangle, \qquad (13)$$

where $\phi_k^{(-)}$ is an incoming Coulomb function and the integral is over the finite volume where the coordinates are real. Application of Green's theorem to Eq. (13) gives an equivalent surface integral form that is more convenient in numerical calculations:

$$f(k_e,k_f)=\tfrac{1}{2}\int_S(\phi_{k_e}^{(+)}\phi_{k_f}^{(+)}\nabla\Psi_{sc}-\Psi_{sc}\nabla\phi_{k_e}^{(+)}\phi_{k_f}^{(+)})\cdot\hat{\mathbf{n}}dS. \qquad (14)$$

Since the Coulomb functions are orthogonal to target bound states, this amplitude formulation is not affected by the ''contamination'' from discrete, two-body channels that limited the flux-based method [11]. This allows the SDCS to be calculated over the full range of the outgoing electron energy.

In Fig. 1 we compare the TDCC and ECS calculations of SDCS for the electron-impact ionization of hydrogen at an incident energy of 17.6 eV, for the case where $L=0$, $S=0$. The time-dependent close-coupling equations for the two-electron radial wave functions were solved on a numerical lattice with a uniform mesh spacing $\Delta r=0.20a_0$. Two sets of results are shown, for grids that extended to $200a_0$ and $400a_0$, respectively. The time propagation of the radial wave functions is determined by the convergence of the collision probabilities. Because of the low energy of the incident electron the total time propagation was several hundred atomic units for these calculations. The lower set of lines is from a calculation that included only the *ss* pair from the l_1l_2 expansion, which is the Temkin-Poet model [21,22]. The middle set of lines coupled the $ss+pp$ pairs and the upper set of lines added *dd* to the l_1l_2 expansion. It was found that including another l_1l_2 pair (*ff*) makes a very small difference of no more than 2% to the differential cross section. The total cross section for the ^1S partial wave was found to be 8.0 Mb calculated using a box size of $200a_0$, and 8.1 Mb when a box size of $400a_0$ was used. The much larger calculations carried out at $400a_0$ show that the differential cross sections are well converged to within 2% at $200a_0$.

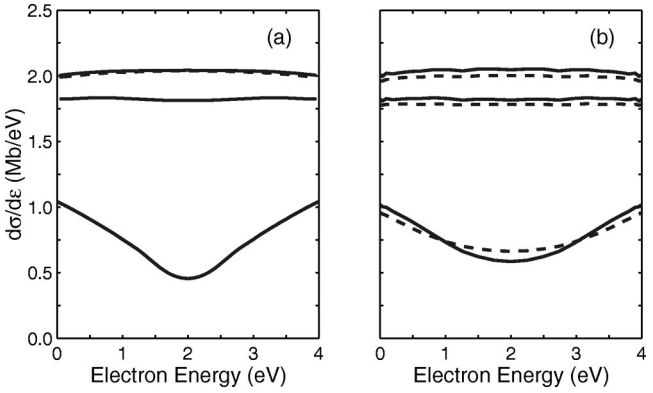

FIG. 1. Singly differential cross sections for electron-impact ionization of hydrogen at 4 eV above threshold, for $L=0$, $S=0$. Statistical weights are not included and the SDCS is defined to give the total ionization cross section when integrated from 0 to 4 eV. (a) Results computed using exterior complex scaling. The lowest curve is the result with only the *ss* partial wave in the calculations and the middle curve couples $ss+pp$. The upper set includes up to *dd* (solid) and *ff* (dashed). (b) Results computed using the time-dependent close-coupling method. The dashed lines are calculations using a box size of $200a_0$, while the solid lines use a box size of $400a_0$. The ordering is as in panel (a), with results shown up to the case using $ss+pp+dd$ pairs ($1.0\,\mathrm{Mb}=1.0\times10^{-18}\,\mathrm{cm}^2$).

For the ECS calculations, the partial wave components of the scattered wave were obtained on a grid whose real part extended to $200a_0$ in each radial dimension. The SDCS were computed from the surface integral expression for the ionization amplitudes given in Eq. (14). Convergence was tested by computing the surface integrals over a range of hyperradii from $150a_0$ to $200a_0$. Over the entire range of ejected electron energy, the computed SDCS, as a function of hyperradius, were found to exhibit only small-scale oscillations (1–2%) about a constant value, so that a simple numerical average produced very stable results. The exception is the Temkin-Poet model, which has slightly different behavior. In this case, the computed SDCS exhibits small oscillations about a slowly varying function that reaches its asymptotic value inversely proportional to the hyperradius. The SDCS's computed here from the integral expression for the ionization amplitudes show the same dependence on ejected electron energy as our earlier results obtained from flux extrapolation [11], with differences in magnitude on the order of 5–8%.

The ejected-energy differential cross sections are, of course, symmetric about $E/2$, where E is the total energy available to the scattered and ejected electrons. We find that the SDCS in the Temkin-Poet model exhibits a V shape, and that adding the *pp* terms in the expansion increases the magnitude of the cross section substantially, showing the critical importance of the dipole coupling in the $L=0$ cross section. The most striking effect of coupling higher angular-momentum terms to the *ss* pair is to flatten the overall differential cross section, an effect predicted by semiclassical calculations [23] and consistent with Wannier theory.

We note that the cross sections computed by TDCC and

ECS are in excellent agreement. The largest differences are seen in the Temkin-Poet model. For this case, TDCC is not fully converged even at a box radius of $400a_0$. We estimate that it would take a box radius of up to $800a_0$ to achieve full convergence, which would result in an *ss* only cross section slightly lower at equal energy sharing and higher at the edges, in better agreement with the ECS result as well as other recent accurate calculations of the Temkin-Poet model [24–26]. This slow rate of convergence is also consistent with the observed behavior in the ECS calculations and is due to the fact that the electrons interact more strongly when higher-angular-momentum components are omitted.

In conclusion, we report ejected-energy differential cross sections for the electron-impact ionization of hydrogen at an impact energy of 17.6 eV for the ^1S partial wave. We find convergence is obtained for a box size of $200a_0$ and three coupled channels. The final differential cross section is virtually flat with a value of 2.0 Mb/eV at equal energy sharing between the electrons, and a total cross section of 8.0 Mb. Recent ECS results [11] find that the total SDCS is also nearly flat at this energy, in contrast to CCC results [13] that are much more V shaped. The same difference between ECS and CCC results persists in the simplified case of the ^1S partial wave [27] and is a major source of discrepancies between previous calculations of both singly and triply differential cross sections for the electron-impact ionization of hydrogen [10–12]. We hope that these results will shed some light on the differences between the ECS and CCC methods and stimulate further theoretical work on this fundamental problem. Efforts are underway to calculate the complete singly differential cross section, as well as triply differential cross sections, at low energies using nonperturbative time-dependent close-coupling theory in order to compare more closely with other theoretical calculations.

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