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Extraction of energy-differential ionization cross sections in time-dependent calculations

K. Bartschat, S. Riordan, and G. Ver Steeg
Department of Physics and Astronomy, Drake University, Des Moines, Iowa 50311
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We propose an alternative way to extract single-differential ionization cross sections from time-dependent wave-packet calculations. Using the Temkin-Poet *S*-wave model for *e*-H collisions, it is shown that a properly constructed time-dependent wave function ''knows'' the energy of the one-particle bound or continuum orbital it has been projected on. No explicit symmetrization scheme is therefore needed to ensure a single-differential ionization cross section that accounts for the indistinguishability of the two outgoing electrons.

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As the classic three-body breakup problem, electron-impact ionization of atomic hydrogen has been of central interest since the beginning of quantum mechanics. Not until less than ten years ago, however, numerical studies were not even attempting to solve the problem in a ''convergent'' fashion, i.e., by using algorithms that can be expected to converge to the correct answer if sufficient computational resources are allocated. This situation changed with the pioneering work of Bray and Stelbovics [1], who developed the so-called ''convergent close-coupling'' (CCC) method and demonstrated its applicability to the Temkin-Poet (TP) model problem [2,3], in which only the *S* wave is considered and hydrogenic states with angular momentum $L=0$ are accounted for.

In the meantime, many different methods have been tried out to solve this model problem, and many have moved on with great success to solve the full *e*-H collision problem. In addition to the CCC method [1,4] these include close coupling with pseudostates [5], the intermediate-energy *R*-matrix (IERM) [6], eigenchannel *R*-matrix [7], and *R*-matrix with pseudostates (RMPS) approaches [8], hyperspherical close coupling [9,10], the *J*-matrix method [11], exterior complex scaling (ECS) [12], a *T*-matrix method [13], direct finite-difference methods (FDM) [14,15], and time-dependent wave-packet approaches [16,17]. Overall, it was found that the total excitation and ionization cross sections could be predicted with very high accuracy by essentially all these methods, with the only practical limitation being the available computational resources.

As pointed out by Pindzola *et al.* [13], however, the situation is much less satisfactory for the SDCS (with respect to the ejected-electron energy). For example, the close-coupling plus pseudostates methods (CCC, RMPS, IERM) exhibit an unphysical asymmetry in the SDCS with respect to half of the available energy for the two outgoing electrons. The reason for this asymmetry has been discussed in great detail [18–21]; it is effectively due to the unequal treatment of the two positive-energy electrons. For futher discussion, particularly with respect to the way the single-differential cross section (SDCS) is extracted from a numerical wave function, see the work by Rescigno *et al.* [25] and by Madison *et al.* [26]. Although fixes for this problem have been suggested and implemented, the appearance of unphysical oscillations even after an *a posteriori* symmetrization of the ''raw results'' remains of some concern. Time-dependent approaches, on the other hand, as well as the ECS method, often suffer from the need for a very large radial mesh on which the problem has to be solved. Like the *T*-matrix formulation [13], these yield symmetric results of the SDCS for ionization. Note, however, that this is partly due to the fact that the information is extracted using obviously symmetric recipes [see, for example, Eqs. (1) and (8) of Ref. [13]). Finally, Jones and Stelbovics [15] obtained benchmark results for the SDCS of the TP model by pushing a direct finite-difference method essentially to numerical convergence.

In this paper, we suggest another way of extracting the ionization cross section from a time-dependent wave-packet solution of the TP model. The method has several appealing features, namely: (1) it is a straightforward extension of the way cross sections for discrete-state excitation are extracted; (2) it can be implemented easily; (3) it is *not explicitly symmetrized* and, therefore, deviations from the required symmetry can be used to judge the numerical accuracy before a final symmetrization; and (4) it further enhances the possibilities for visualizing the details of the collision process.

To illustrate the basic idea, we will concentrate on the singlet spin channel only. The wave-packet method solves the time-dependent Schrödinger equation (TDSE)

$$i\frac{\partial P(r_1,r_2,t)}{\partial t}=\left[-\frac{1}{2}\left(\frac{\partial^2}{\partial r_1^2}+\frac{\partial^2}{\partial r_2^2}\right)-\frac{1}{r_<}\right]P(r_1,r_2,t) \quad (1)$$

by propagating the initial state

$$P(r_1,r_2,0)=[P_{1s}(r_1)G_{k_0}(r_2)+P_{1s}(r_2)G_{k_0}(r_1)]\sqrt{2} \quad (2)$$

in time. Here P_{1s} and G_{k_0} represent the initial bound orbital and a Gauss packet moving with momentum k_0, respectively, while $r_<$ is the smaller of r_1 and r_2. Total excitation cross sections for discrete states $2s,3s,4s,\ldots,ns$ are then obtained from the probabilities to find one of the electrons in that particular state. After defining

$$F_{ns}(r_2,t)\equiv\int_0^\infty dr_1 P_{ns}(r_1)P(r_1,r_2,t), \quad (3)$$

the cross section (in a_0^2) is given by

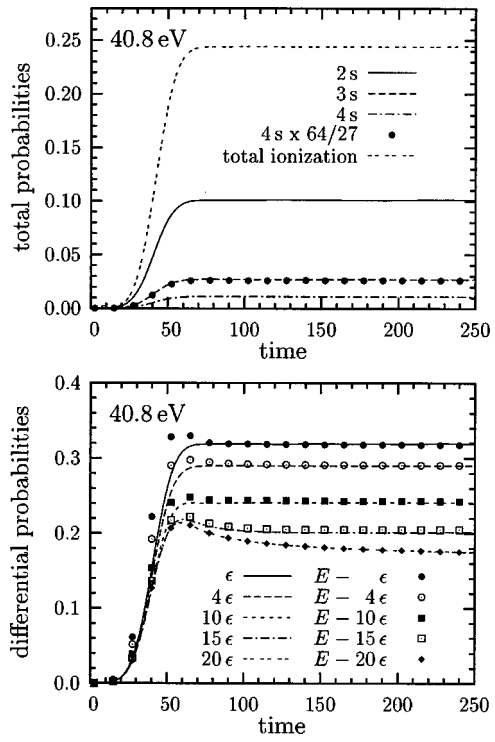

FIG. 1. Time-dependent probabilities for discrete excitation and total ionization (top) and energy-differential ionization (bottom) for the singlet spin channel of the Temkin-Poet model at an incident energy of 40.8 eV. The energy ϵ is 1/40 of the available excess energy.

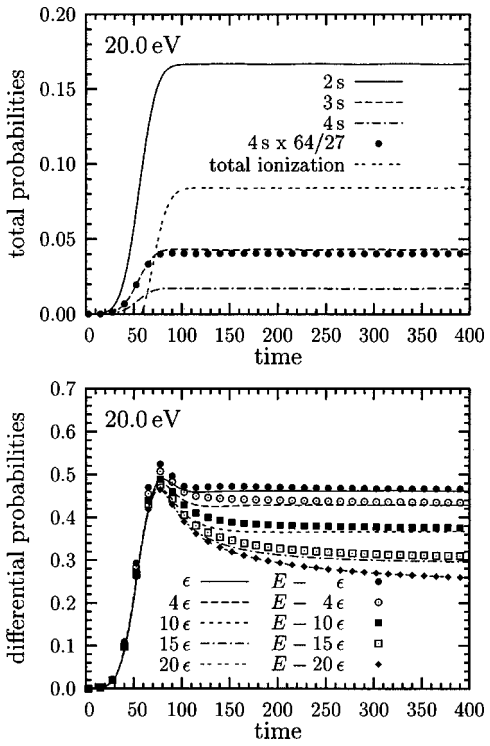

FIG. 2. Same as Fig. 1 for an incident energy of 20.0 eV.

$$\sigma_{ns} \equiv \lim_{t\to\infty} \frac{\pi}{4k_0^2}\, 2 \int_0^\infty dr_2 |F_{ns}(r_2,t)|^2. \tag{4}$$

Note that the factor of 4 in the denominator reflects the spin weight of the singlet channel, while the 2 in the numerator comes from the fact that projection to $P_{ns}(r_1)$ and $P_{ns}(r_2)$ give the same result. Finally, the total ionization cross section can be obtained as

$$\sigma_{ion} = \frac{\pi}{4k_0^2}\left(1 - 2 \lim_{t\to\infty} \sum_{n=1}^{\infty} \int_0^\infty dr_2 |F_{ns}(r_2,t)|^2 \right), \tag{5}$$

i.e., by subtracting the probabilities for elastic scattering and discrete-state excitation from unity. [For numerical reasons, it may be advisable to also subtract the probabilities of having both electrons end up in bound states, but for simplicity we will omit these details here.]

The straightforward extension of the above method to excitation of continuum states, i.e., ionization, is to replace the bound orbital $P_{ns}(r_1)$ in the above formulas by a Coulomb function $P_{ks}(r_1)$, where $k^2/2 \equiv \epsilon$ denotes the energy of the electron represented by this function. Consequently, we define

$$F_{ks}(r_2,t) \equiv \int_0^\infty dr_1 P_{ks}(r_1) P(r_1,r_2,t), \tag{6}$$

and calculate

$$\overline{\sigma}_{ks} \equiv \lim_{t\to\infty} \frac{\pi}{4k_0^2}\, 2 \int_0^\infty dr_2 |F_{ks}(r_2,t)|^2. \tag{7}$$

If the Coulomb function $P_{ks}(r_1)$ is normalized to behave as $k^{-1/2}$ times a sine function for large r_1, then $\overline{\sigma}_{ks}$ *is proportional to the SDCS at the energy* ϵ. Note that this normalization by unit energy brings the continuum functions at par with the bound orbitals [22]. The proportionality constant is not known immediately, but it is easy to determine since the area under the SDCS curve must equal the total ionization cross section. Since the latter can be determined with high accuracy, $\overline{\sigma}_{ks}$ can be normalized to give the absolute values of $d\sigma/d\epsilon$. It is worth noting that this method does *not* require to generate quasicontinuum states through diagonalization of the Hamiltonian on the lattice, nor is it restricted to perform projections only at the resulting energies of the diagonalization. Note that box-normalized continuum functions have also been used recently by Colgan *et al.* [23].

Figures 1–4 illustrate our method for incident energies of 40.8 and 20.0 eV in the Temkin-Poet model of *e*-H collisions. These calculations were performed on a single-CPU Alpha workstation and could run over a period of a few days on a personal computer. We used the standard leapfrog time-propagation method on a 2400×2400 radial mesh with a stepsize of $h=0.2$, i.e., the maximum radius was $480a_0$. Such a large mesh is not really necessary for the 40.8 eV case, but it seems needed to get converged results at 20.0 eV [24]. The total probabilities for excitation of the $2s$, $3s$, and $4s$ states, as well as for ionization, are shown in the top parts of Figs. 1 and 2 as a function of the propagation time, while

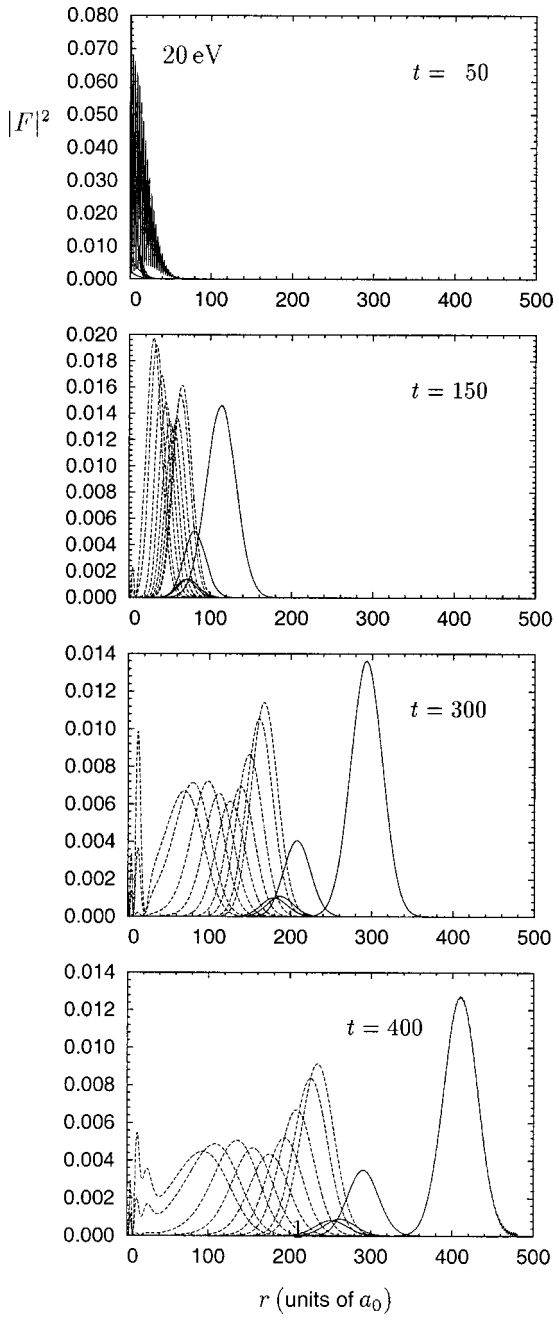

FIG. 3. The functions $|F_{ns}(r_2,t)|^2$ (solid lines) and $|F_{ks}(r_2,t)|^2$ (dashed lines) for the TP model at various times t for an incident electron energy of 20.0 eV. For better visibility, $|F_{4s}(r_2,t)|^2$ has been multiplied by 64/27. The functions plotted correspond to the cases shown in the bottom part of Fig. 2, with the energy loss increasing from right to left.

the normalized differential probabilities for exciting 39 Coulomb states in equal-energy steps are shown in the bottom parts.

Not surprisingly, the total probabilities converge very well with time, and accounting for discrete excitations to states with $n \geqslant 5$ via an $1/n^3$ scaling law seems justified. More interesting, therefore, are the bottom parts of the figures. For sufficiently long times after the collision, we see that projecting to a single Coulomb wave representing a fraction ϵ of the

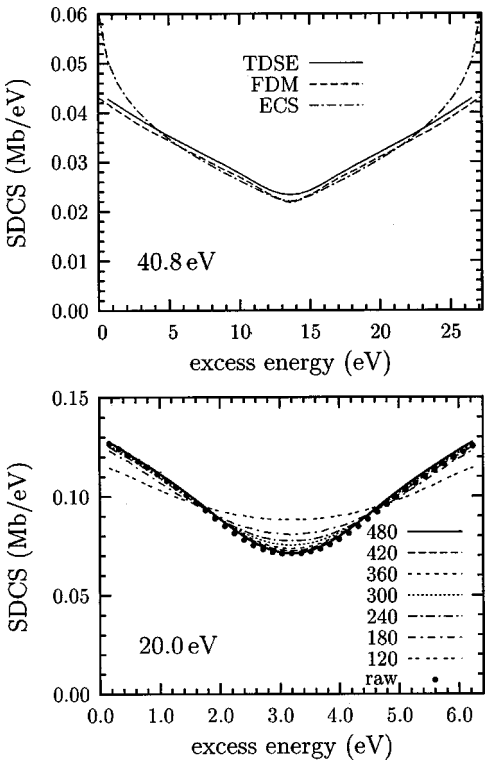

FIG. 4. Energy-differential ionization cross section for the TP model at 40.8 eV (top) and 20.0 eV (bottom). The TDSE results for 40.8 eV are compared with FDM [15] and ECS [12] predictions. The labels in the curves for 20.0 eV indicate the maximum radius (in a_0), and the dots are the results for $480a_0$ before any symmetrization.

excess energy E or, in turn, $E-\epsilon$, essentially gives the same probabilities. In other words, the time-dependent wave function $P(r_1,r_2,t)$ "knows" how to behave when it is projected to either one of these functions. Although the symmetry is not perfect, especially if one does not wait long enough, it is very satisfactory overall. We also note that the results for the equal-energy-sharing situation, while symmetric by construction, are the slowest to converge with time. In fact, for the 20.0-eV case they take longer than we could afford to wait given the finite mesh size. The mesh boundaries result in a reflection of the elastically scattered wave function and, consequently, lead to difficulties in extracting the total ionization cross section.

The response of $P(r_1,r_2,t)$ after projection to bound or continuum states is shown even clearer in Fig. 3, which exhibits $|F_{ns}(r_2,t)|^2$ and $|F_{ks}(r_2,t)|^2$ for the 20.0-eV case at four different times. We see that these functions represent Gauss-type packages as well, with their centers located exactly at the positions one would expect for wave packets that were created at the collision time $(70a_0/k_0)$ and move away from the target with the energy left over after excitation or ionization. Looking, in particular, at the results for very asymmetric energy sharing, it is remarkable that the integrals under two corresponding curves, e.g., the two dashed lines furthest to the right and left, are essentially the same. A color movie of this figure (TP-20p0-proj.mov.sit) can be down-

loaded via anonymous ftp from bartschat2.drake.edu:/pub

Finally, Fig. 4 shows the differential ionization cross section. For 40.8 eV, the agreement in shape with the FDM calculation of Jones and Stelbovics [15] is excellent, but there is a small difference in the absolute value. The reason for this disagreement can be traced back to the predictions for the total ionization cross section—the FDM value of $0.197\pi a_0^2$ [15] is, indeed, approximately 3% smaller than our time-dependent result of $0.202\pi a_0^2$.

For 20.0-eV incident energy, we show how our results converge with the meshsize. For the largest range of $480a_0$, we also present the ''raw data'' to demonstrate that the final symmetrization is essentially optional. For the smaller ranges, the SDCS results are well known to come out too ''flat'' [13], i.e., the results for the symmetric and asymmetric energy sharing are very similar and, therefore, still allow for a satisfactory prediction of the total ionization cross section. A comparison with unpublished FDM results for this case [24], however, shows that the TDSE method most likely does not reach a sufficiently deep minimum near half the excess energy ($E/2$). Additional tests with a broader (in coordinate space) initial wave packet, corresponding to a smaller energy width, showed that the energy resolution is indeed part of the problem. As mentioned above, however, another reason is the slow convergence of the differential ionization probability with time for the equal-energy sharing condition (cf. Fig. 2).

In summary, we have suggested an alternative way to extract energy-differential ionization cross sections from a time-dependent wave-packet model. Using a recipe that is not explicitly symmetrized allows for qualitative tests of the numerical accuracy by checking the results against the symmetry requirements of the underlying physical problem.

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