Practical calculations of quantum breakup cross sections

C. W. McCurdy[1,3] and T. N. Rescigno[1,2]
[1]*Lawrence Berkeley National Laboratory, Computing Sciences, Berkeley, California 94720*
[2]*Lawrence Livermore National Laboratory, Physics Directorate, Livermore, California 94551*
[3]*Department of Applied Science, University of California, Davis, Livermore, California 94551*
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The Schrödinger equation is solved numerically using the method of exterior complex scaling for several models of the breakup of an atom by electron impact. Using the accurate wave functions thereby obtained for these model problems, several well-known integral expressions for quantum-mechanical breakup amplitudes are tested. It is shown that some formally correct integral expressions for the breakup amplitudes can yield numerically unstable or poorly convergent results. Calculations are presented for a case with simple exponential potentials and a case in which a metastable state of the target, analogous to an autoionizing state, can decay into the breakup channel. For cases involving only short-range (non-Coulomb) interactions, alternative expressions can be found that are stable in calculations of practical scale.

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I. INTRODUCTION

The collisional breakup problem in quantum mechanics poses both formal and practical difficulties that appear only in quantum scattering problems involving the fragmentation of the colliding partners. Given an accurate solution of the Schrödinger equation for the scattering wave function, the extraction of the probabilities for breakup processes can be both computationally and formally challenging. The subject of this paper is the exploration of various approaches to computing breakup cross sections and amplitudes, given the scattering wave function as the starting point.

The question of how to extract the breakup amplitude in practice from the scattering wave function makes sense only in a context where the wave function can be computed by some method that is independent of the asymptotic matching condition that defines the scattering amplitudes. We have shown recently [1–4] that it is possible in breakup problems, without appealing to any explicit asymptotic form, to solve directly for the scattered portion Ψ_{sc} of the wave function $\Psi^{(+)}$,

$$\Psi^{(+)}=\Phi_0+\Psi_{sc}, \tag{1}$$

where Φ_0 is the initial, unperturbed state. Since the initial state is defined so that the scattered wave Ψ_{sc} contains only outgoing waves, exterior complex scaling [5] of the coordinates of the particles can be used to construct it without matching to asymptotic boundary conditions. As will be explained in the following section, in place of the scattering boundary conditions, exterior complex scaling allows one to solve for the same wave function using the asymptotic condition that $\Psi_{sc}\rightarrow 0$ $(r\rightarrow\infty)$ as the coordinates of any particle go to infinity.

Once the driven equation for the scattered wave,

$$[E-H]\Psi_{sc}=[H-E]\Phi_0, \tag{2}$$

has been solved, the cross section for any process that is energetically allowed can in principle be calculated from the asymptotic behavior of the quantum-mechanical flux,

$$\mathbf{F}=\frac{\hbar}{2i\mu}[\Psi_{sc}^{*}\boldsymbol{\nabla}\Psi_{sc}-\Psi_{sc}\boldsymbol{\nabla}\Psi_{sc}^{*}]. \tag{3}$$

Thus the calculation of cross sections in this approach is a two-step process: first the calculation of the scattered wave via Eq. (2) with exterior scaling of the coordinates, and second the interrogation of the wave function to compute the cross sections from the appropriate quantum-mechanical flux, using Eq. (3) with correctly specified directions for the outgoing particles, or by projection onto final target states [1]. To compute the electron-impact ionization cross section in collisions of electrons with hydrogen, we resorted to this fundamental definition of the cross section, because the Coulomb interactions between the three separating particles prevented matching to the asymptotic form in practice [4].

For purely geometrical reasons, the calculation of the asymptotic flux can require calculations well beyond the range of the interaction potentials, even in the case of short-range interactions. That fact is sometimes discussed in introductions to scattering theory, but often forgotten because it is irrelevant to the more common integral expressions for scattering amplitudes. Dirac [6] touches on this point when deriving the expression for the scattering amplitude (or ''scattering coefficient'' as it was then called). In part for this reason, formal scattering theory makes use of expressions for amplitudes that involve matrix elements of the appropriate interaction potential between the full scattering wave function and an unperturbed asymptotic state.

For example, in a simple case involving only short-range interactions and two particles departing from a third, infinitely massive particle in the final state, the breakup amplitude can formally be written

$$f=\langle \mathbf{p}_1,\mathbf{p}_2|V|\Psi^{(+)}\rangle, \tag{4}$$

where V is the appropriate interaction potential, and the final state is just a product of plane waves

$$|\mathbf{p}_1,\mathbf{p}_2\rangle = \frac{1}{(2\pi)^{3/2}} \exp[i(\mathbf{p}_1\cdot\mathbf{r}_1+\mathbf{p}_2\cdot\mathbf{r}_2)], \tag{5}$$

with two particles exiting with momenta $\mathbf{p}_1$ and $\mathbf{p}_2$. Equation (4) is an example of an integral expression for a breakup amplitude. One of the conclusions of this paper concerns the utility of Eq. (4) in numerical calculations. Even given a numerically exact representation of $\Psi^{(+)}$, Eq. (4) does not provide a practical way to compute the breakup amplitude, because, for any finite-sized integration volume, the discrete inelastic channel components of $\Psi^{(+)}$ make large spurious contributions. We will demonstrate that Eq. (4) has this practical limitation, even though it is formally correct and quite useful for analytically deriving approximations to the breakup amplitude from assumed forms of $\Psi^{(+)}$.

In this paper we test the formal definitions of breakup amplitudes derived from the theory of rearrangement collisions for several model problems by first computing the scattered wave portion of the wave function and then using it in both flux and integral expressions for the cross section or amplitude. There are some considerable surprises in this exercise, even (especially) for short-range interactions. Having access to accurate wave functions for breakup collisions uncovers some unexpected numerical problems with common integral expressions for the amplitudes, while showing others to be entirely stable and practical. From this study we conclude that expressions based on ''two-potential'' formulas, which are known to be formally equivalent to Eq. (4), can avoid numerical problems associated with discrete inelastic channels, as well as more insidious numerical problems associated with the presence of competing breakup processes having different time scales.

For example, in a problem where a metastable state is formed that can decay into the breakup channel, the lifetime of the state can be long enough for the originally incident particle to leave the interaction region before the target breaks up. The two processes of direct and resonant breakup can interfere quantum mechanically. Other difficulties can arise when a short lifetime leads to postcollision interactions, in which the three (or more) fragments of the collision system still interact while separating. If all the interactions are short range in nature, it would seem that an expression like Eq. (4) should be entirely appropriate and practical. However, as we will show, neither Eq. (4) nor the flux approach using the appropriate version of Eq. (3) is an effective way to calculate the cross section in such cases.

Since most aspects of the problem of collisional breakup can be viewed in terms of the formalism for rearrangement collisions [7] in quantum scattering theory, integral expressions for scattering amplitudes like that in Eq. (4) are generally derived in that context. That approach requires fundamental modifications in the case that the separating fragments interact via Coulomb or other long-range forces. While those modifications have been the subject of a considerable literature, beginning with Rudge [8] and Peterkop [9] and most recently extended by Alt and Mukhamedzhanov [10], a formulation that is both theoretically sound and computationally practical has yet to emerge. Even in cases where only short-range forces are involved, much of the relevant theory of rearrangement scattering has been tested primarily in the context of distorted-wave or other perturbative approximations.

The problem of how to construct the breakup cross sections is a central one for electron-impact ionization of atoms, where it is compounded both by long-range Coulomb interactions and by the presence of autoionizing states of the target atom. In the following sections we attempt to shed some light on that question, although the results we present here address only short-range interactions. For the Coulomb case, which is of course the most interesting physical problem, we must currently resort to cutting off the Coulomb interactions, or to the flux approach, which may be impractical when long-lived autoionizing states are present.

The outline of this paper is as follows. Section II describes the explicit connection between the asymptotic quantum-mechanical flux and breakup cross sections. Section III poses two model problems for breakup, both of which involve only exponentially bounded forces, and in one of which a metastable state of the target can be excited leading to breakup. In Sec. IV we explore two different forms of integral expressions for the breakup amplitudes and compare with the flux approach. For these problems we are able to identify an accurate and efficient way to calculate the breakup cross sections. Finally, in Sec. V we offer some speculations on how the work presented here might be extended to Coulomb interactions to treat electron-impact ionization, and point out a key difference between the integral expressions used here and those of the formal theory of the Coulomb breakup problem.

II. BREAKUP CROSS SECTIONS FROM DIRECT CALCULATION OF QUANTUM-MECHANICAL FLUX

Because our motivation is to develop practical methods for treating electron- and positron-impact ionization of atoms and molecules, we will restrict the model systems we consider here to involve particles of the mass of an electron. Furthermore, we restrict our discussion to two-dimensional models in the spirit of the Temkin-Poet model for electron-hydrogen atom scattering [11]. We will also specialize the discussion of quantum-mechanical flux to these cases, but obviously all the formalism generalizes easily. We employ atomic units $(m=\hbar=e=1)$ throughout.

Consider a system of two particles interacting with each other and a center of force. All the model problems in this paper are described by a Hamiltonian of the form

$$\begin{aligned} H &= -\frac{1}{2}\frac{\partial^2}{\partial r_1^2} - \frac{1}{2}\frac{\partial^2}{\partial r_2^2} + v(r_1) + v(r_2) + V_{\rm int}(r_1,r_2) \\ &\equiv T + v(r_1) + v(r_2) + V_{\rm int}(r_1,r_2), \end{aligned} \tag{6}$$

where r_1 and r_2 are restricted to the interval $(0, \infty)$. The initial state in Eqs. (1) and (2) is defined with permutational symmetry that depends on whether singlet or triplet spin coupling is employed,

$$\Phi_0 = \frac{1}{\sqrt{k_0}}[\sin(k_0 r_1)\varphi_0(r_2) \pm \sin(k_0 r_2)\varphi_0(r_1)], \qquad (7)$$

where the upper (lower) sign corresponds to singlets (triplets) and $\varphi_0(r)$ is a bound state of the one-particle Hamiltonian with potential $v(r)$. These equations completely define the problem of solving Eq. (2) for the scattered wave, subject to a boundary condition that it is purely outgoing.

Assuming that only short-range potentials appear in Eq. (6), the asymptotic form of the scattered wave function appropriate to this case is

$$\Psi_{\rm sc}(r_1,r_2) \underset{r_1 \text{ or } r_2 \to \infty}{\sim} \sum_n f_n(e^{ik_n r_1}\varphi_n(r_2) + e^{ik_n r_2}\varphi_n(r_1))/k_n^{1/2} + F(\alpha)e^{iK\rho}/(K\rho)^{1/2}. \qquad (8)$$

The sum over n corresponds to elastic and inelastic scattering leaving the target in the discrete state $\varphi_n(r)$, and f_n is the corresponding scattering amplitude. The second term corresponds to breakup, expressed in terms of the hyperradius $\rho = (r_1^2 + r_2^2)^{1/2}$ and hyperangle $\alpha = \arctan(r_2/r_1)$, with $F(\alpha)$ denoting the breakup amplitude. The two outgoing momenta satisfy the energy conservation relation

$$k_1^2 + k_2^2 = K^2 = 2(E - \varepsilon_b). \qquad (9)$$

In the asymptotic region they also correspond to the hyperangle in $F(\alpha)$, with $\alpha = \arctan(k_2/k_1)$, so we will sometimes use the notation $F(k_1,k_2)$ for the breakup amplitude.

Conceptually, the simplest way to extract the breakup cross section is via the direct evaluation of the flux. For a two-dimensional problem the flux vector $\mathbf{F}_\rho$ evaluated at a particular value of the hyperradius is given by

$$\mathbf{F}_\rho = \frac{1}{2i}\left[\Psi_{\rm sc}^*\begin{pmatrix}\partial/\partial r_1\\ \partial/\partial r_2\end{pmatrix}\Psi_{\rm sc} - \Psi_{\rm sc}\begin{pmatrix}\partial/\partial r_1\\ \partial/\partial r_2\end{pmatrix}\Psi_{\rm sc}^*\right]_\rho. \qquad (10)$$

This is the total outgoing flux associated with the scattered wave and contains contributions for all channels, both discrete and breakup. To get the flux associated with a particular discrete channel a, we can apply a projection operator for that state,

$$\mathbf{P}_a = P_a(r_1) + P_a(r_2) - P_a(r_1)P_a(r_2), \qquad (11)$$

where the individual projection operators are defined as

$$P_a(r_1) = \varphi_a(r_1)\int dr_1' \varphi_a^*(r_1'). \qquad (12)$$

Inserting $\mathbf{P}_a\Psi_{\rm sc}$ into the flux expression in Eq. (10) yields the flux into channel a evaluated at a particular (large) value of ρ. The cross section for any discrete two-body channel is then given by

$$\sigma_a = \lim_{\rho\to\infty}\frac{4\pi}{k_0^2}\int_0^{\pi/2}\mathbf{F}_\rho^{(a)}\cdot\begin{pmatrix}\cos\alpha\\ \sin\alpha\end{pmatrix}d\alpha. \qquad (13)$$

To get the singly differential breakup cross section $\sigma(k_1,k_2)$, which gives the dependence of breakup on the final values of the momenta of the two outgoing particles, we must project out all of the target bound-state contributions using the projection operator

$$Q = 1 - \sum_a^{N_{\rm pound}} \mathbf{P}_a. \qquad (14)$$

The flux for breakup, $\mathbf{F}_{\rho_0}^{(\rm ion)}$, evaluated at a particular value of the hyperradius, is given by Eq. (10) with $Q\Psi_{\rm sc}$ replacing $\Psi_{\rm sc}$. The flux is related to the breakup cross section for particular values of k_1 and k_2 by its dot product with the outward normal in the direction determined by k_1 and k_2,

$$\sigma(k_1,k_2) = \frac{1}{k_1 k_2}\frac{4\pi}{k_0^2}\mathbf{F}_{\rho_0}^{(\rm ion)}\cdot\boldsymbol{\rho}_0 + O(1/\rho_0), \qquad (15)$$

where the vector $\boldsymbol{\rho}_0$ is given by

$$\boldsymbol{\rho}_0 = \begin{pmatrix}k_1\\ k_2\end{pmatrix}\rho_0/K. \qquad (16)$$

The residual term of order $1/\rho_0$ in Eq. (15) can easily be derived from the asymptotic form in Eq. (8) and is purely geometrical; that is, it does not depend on the range of the potential. As we will see below, terms of this type, as well as other slowly decaying terms having to do with how the wave function reaches the asymptotic form in Eq. (8), can be avoided by using integral expressions for the amplitude.

We next explore some simple examples, for which we demonstrate the complex exterior scaling approach, the asymptotic form of the wave function, and the asymptotic behavior of the scattered flux.

III. TWO BREAKUP PROBLEMS INVOLVING ONLY SHORT-RANGE POTENTIALS

To solve the Schrödinger equation for the scattered wave, we apply the exterior complex scaling transformation, and then use either ordinary polynomial finite elements, as we have for other breakup problems [1], or the discrete-variable representation/finite-element method we developed recently [12].

Under the exterior scaling transformation, a real scalar distance r is transformed as

$$R(r) = \begin{cases} r, & r < R_0 \\ R_0 + (r - R_0)e^{i\eta}, & r \geqslant R_0, \end{cases} \qquad (17)$$

where R_0 is a large real number and η is a positive number between 0 and π. This transformation is applied to the radial coordinates of both electrons. Exterior complex scaling of coordinates was invented by Simon [5] in 1979 to extend the formal mathematical theorems in scattering theory associated with complex scaling of coordinates [13]. The crucial fact is that under this transformation the scattered wave tends to zero exponentially at large distances because it is purely outgoing.

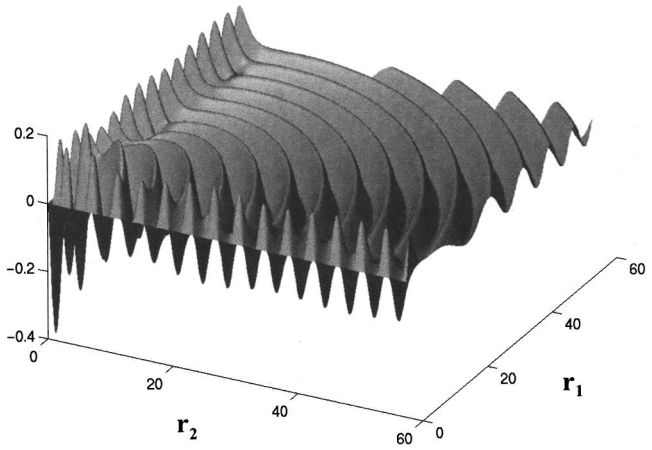

FIG. 1. Scattered wave for the model of Sec. III A for 35 eV incident energy.

Thus we can formulate the driven Schrödinger equation

$$\left[-\frac{1}{2}\frac{\partial^2}{\partial r_1^2}-\frac{1}{2}\frac{\partial^2}{\partial r_2^2}+v(r_2)+v(r_1)+V_{\rm int}(r_1,r_2)-E\right]\Psi_{\rm sc}(r_1,r_2)$$

$$=\frac{1}{\sqrt{k_0}}\{[v(r_1)+V_{\rm int}(r_1,r_2)]\sin(k_0 r_1)\varphi_0(r_2)\pm[v(r_2)+V_{\rm int}(r_1,r_2)]\sin(k_0 r_2)\varphi_0(r_1)\} \qquad (18)$$

as a set of linear equations with the boundary conditions that the scattered wave $\Psi_{\rm sc}(r_1,r_2)$ vanish at the edges of the finite-element grid. For values of r_1 and r_2 both less than R_0, the scattered wave is the physical wave function. Thus we can apply the flux expressions above in that region to extract the cross sections.

A. Simple exponential interactions

The first and simplest of our examples is one we have considered before [12,14]. The one-body potentials are attractive exponentials that bind only one state at $\varepsilon_b = -0.411\,45$ hartree,

$$v(r)=-3\exp(-r), \qquad (19)$$

and the total interaction potential is of the form

$$V(r_1,r_2)=v(r_1)+v(r_2)+10\exp(-r_1-r_2). \qquad (20)$$

This example allows only two processes, elastic scattering and breakup.

The scattered wave function $\Psi_{\rm sc}(r_1,r_2)$ for an incident energy of 35 eV where both channels are open is shown in Fig. 1. Its features correspond to the two processes that are allowed in this example. The peaks near the axes correspond to the elastic flux from the first term in Eq. (8), and the wave fronts in the shape of arcs correspond to breakup from the second term.

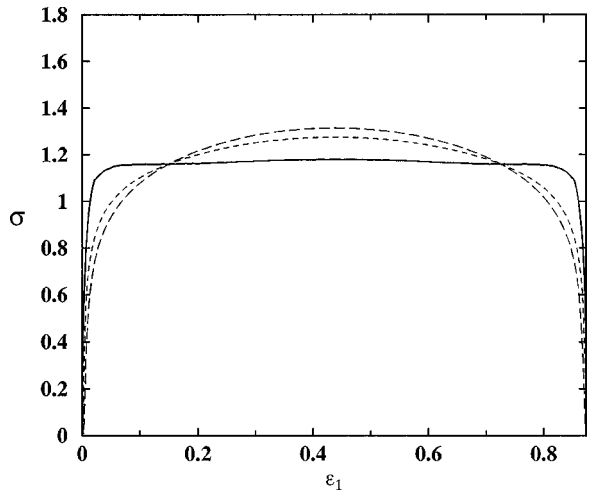

FIG. 2. Singly differential cross section (a_0^2/hartree) vs energy (hartrees) of electron 1 from projected flux evaluation for model of Sec. III A at 35 eV incident energy; long-dashed line, flux at $\rho = 40a_0$, short-dashed line, flux at $\rho = 56a_0$, solid line, extrapolation of projected flux to $\rho \to \infty$.

In Fig. 2 we show the singly differential cross section for breakup evaluated at several values of ρ_0. To obtain the physical cross section we extrapolate to $\rho_0 \to \infty$, which is easily done because the dependence on ρ_0 of the cross section evaluated at finite hyperradii is known via Eq. (15). The total breakup cross section as a function of incident energy is shown in Ref. [14].

For this problem the flux approach to computing the breakup cross section is both practical and adequate. Surprisingly, that is not the case for all short-range potentials.

B. Resonance problem with exponentially bounded potentials

This example is constructed to illustrate some of the dynamics involved when the breakup process proceeds through autoionization. The potential also involves only exponentially bounded interactions,

$$V(r_1,r_2)=v(r_1)+v(r_2)+4\exp(-r_1-r_2), \qquad (21)$$

with a one-body potential chosen to bind a shape resonance,

$$v(r)=-3\exp(-r)+0.4\exp[-0.5(r-5)^2]. \qquad (22)$$

The one-body potential again binds only one state, at $\varepsilon_b = -0.403\,26$ hartree, but it also has a shape resonance corresponding to the complex resonance energy $\varepsilon_{\rm res}=0.3114 - 0.0277i$ hartree. The potential is shown Fig. 3, where the barrier through which the resonance tunnels is visible, separating the entrance and exit wells from the breakup region.

It is useful to understand from the outset that a resonance such as the one in this example, like a physical autoionizing state of an atom, does not correspond to an isolated pole of the *full* Hamiltonian, even though it does correspond to an isolated pole of the *target* Hamiltonian. Instead, since a free

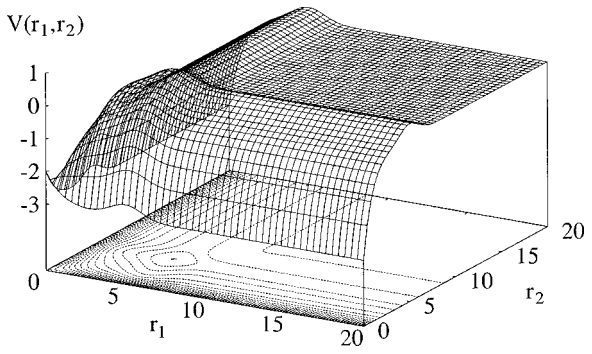

FIG. 3. Potential for the model problem of Sec. III B, which supports an autodetaching resonance.

electron of any momentum can be associated asymptotically with the discrete, complex pole of the target Hamiltonian, the resonance corresponds to a branch cut of the Green's function for the full Hamiltonian. That branch cut has its origin at the complex resonance energy of the autoionizing or autodetaching state.

In Fig. 4 we show the eigenvalue spectrum of the full Hamiltonian, but for a case with a somewhat broader resonance so that the origin of the resonance branch cut is more apparent in the graph, namely, $v(r)=-3\exp(-r)+0.4\exp[-0.5(r-4)^2]$ [a Gaussian barrier moved inward slightly from that in Eq. (22)]. This graph shows the discrete eigenvalues from an ordinary complex scaling calculation [$R_0=0$ in Eq. (17)] using finite elements. The branch cuts correspond to rows of poles in this discrete representation. In this figure, the bound state, the elastic scattering cut, and the ionization cut are clearly visible. Also, above the ionization threshold, the resonance branch cut appears with its origin at the complex autodetaching resonance energy. Incidentally, also visible in this figure are two discrete poles corresponding to discrete states of the full Hamiltonian in which *both* electrons, in the simplest interpretation, are in the autodetaching resonances. Those states correspond to a discrete state of the anion that can decay emitting *two* electrons.

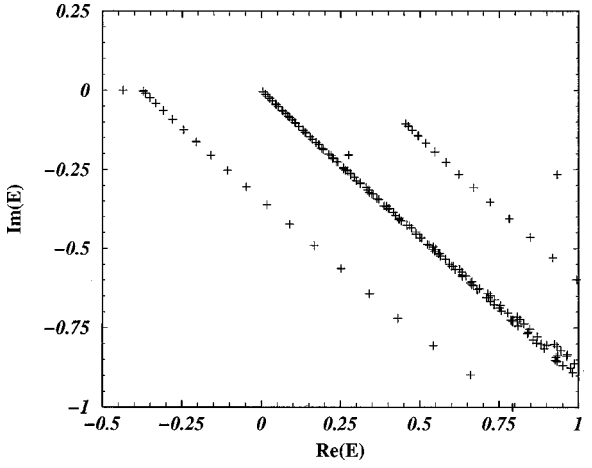

FIG. 4. Eigenvalue spectrum in the complex plane of the complex-scaled Hamiltonian for the two-electron model problem of Sec. III B with $v(r)=-3\exp(-r)+0.4\exp[-0.5(r-4)^2]$.

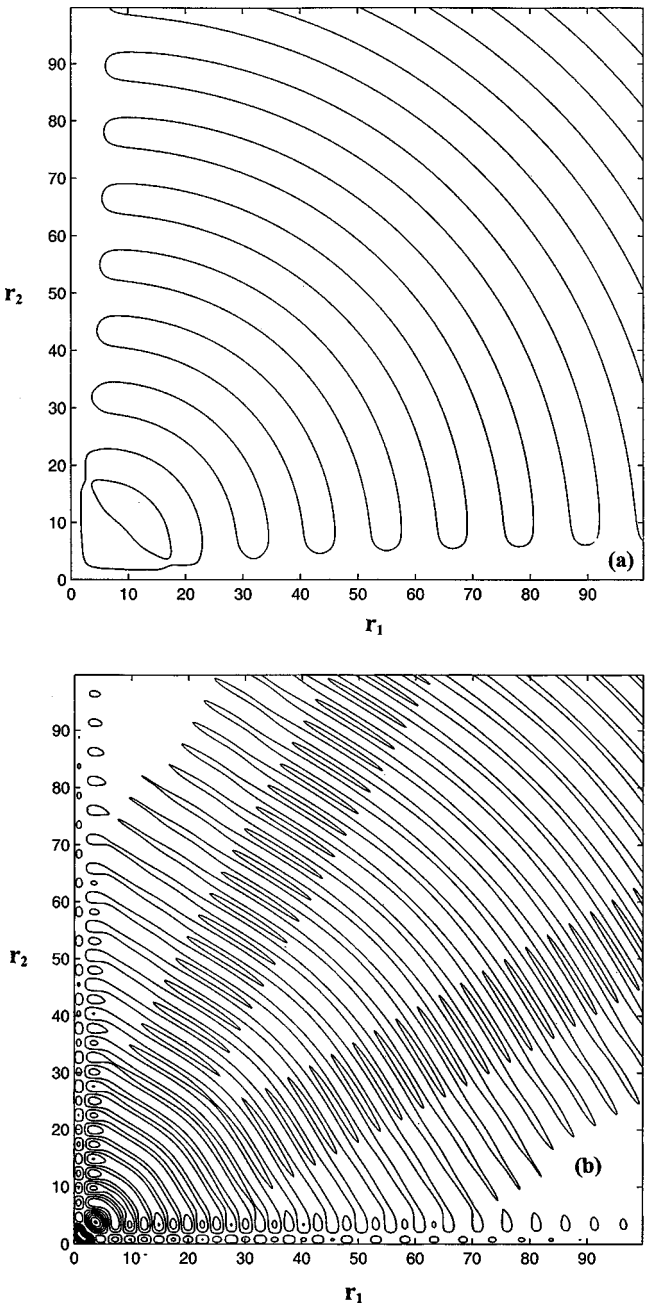

FIG. 5. Scattered wave function for the model problem of Sec. III B (a) below the resonance threshold (15 eV incident energy) and (b) above the resonance threshold (40 eV incident energy).

Those states have no analogy in a two-electron Coulomb system.

In Fig. 5 we show two wave functions for the example in Eqs. (21) and (22). The first, below the resonance energy, is similar in appearance to the nonresonant case in Fig. 1, and its features can be interpreted in exactly the same way. The second is for an energy above the resonance threshold. This wave function is strikingly different. Even though it arises from the same short-range potentials, it shows no indication at similar radial distances of the arcs corresponding to outgoing flux in the breakup channel. Instead it appears, out to quite large distances, to be the superposition of products of

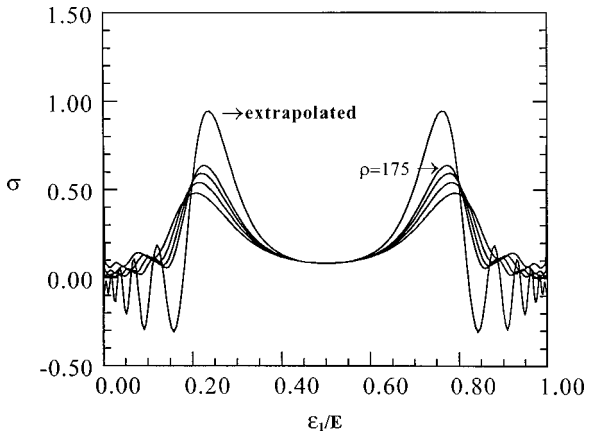

FIG. 6. Singly differential cross section (a_0^2/hartree) vs energy fraction for electron 1 from projected flux evaluation for the model problem of Sec. III B at 40 eV incident energy. Cross section is shown at $\rho=100a_0$, $125a_0$, $150a_0$, and $175a_0$, along with the extrapolated result for $\rho\to\infty$.

two terms corresponding to an outgoing electron with a real momentum and one with a complex momentum. If $\varepsilon_{\rm res}=\varepsilon_r - i\Gamma/2$ is the resonance energy, the two momenta that appear to be determining the form of the wave function in this region are

$$k_{\rm res}=\sqrt{\varepsilon_{\rm res}}, \tag{23}$$

$$k_{\rm free}=\sqrt{2(E-\varepsilon_r)}. \tag{24}$$

Since the wave function is symmetric under interchange of the electronic coordinates, the scattered wave has the appearance of a superposition of two ''plane waves''

$$\Psi_{\rm sc}\propto\exp i\begin{pmatrix}k_{\rm res}\\ k_{\rm free}\end{pmatrix}\cdot\begin{pmatrix}r_1\\ r_2\end{pmatrix}+\exp i\begin{pmatrix}k_{\rm free}\\ k_{\rm res}\end{pmatrix}\cdot\begin{pmatrix}r_1\\ r_2\end{pmatrix}. \tag{25}$$

This is the form one would deduce from the assumption that the incident electron excites an autodetaching state, which, once excited, emits an outgoing wave at the resonance energy as it decays. Near the axes, for small r_1, for example, the distance (in a_0) over which this behavior is evident is roughly $\Delta r_2\approx k_{\rm free}/\Gamma$. The range over which the interference pattern from Eq. (25) qualitatively describes the dynamics is considerably larger, however. In this region we see wave fronts perpendicular to the wave vectors $\binom{k_{\rm res}}{k_{\rm free}}$ and $\binom{k_{\rm free}}{k_{\rm res}}$. The distance over which this nonasymptotic behavior persists is determined by the dynamics of autodetachment and not by the range of the interaction potential, which in this case is obviously of much shorter range.

Figure 6 shows the singly differential cross section computed from the flux at an incident energy above the resonance energy. The flux has obviously not reached its asymptotic behavior, and does not extrapolate to reasonable values. Here, then, is a practical dilemma: although the flux approach works in principle, it will require unrealistically large grids in the case that breakup can proceed through decay of an autoionizing target state. This model problem restricts the interaction between the two electrons to the region where *both* electrons are at short distances from the origin. So the behavior of the scattered flux might seem to be at odds with the usual expectation of collision theory that only calculations within the range of the interaction potential should be required to extract the cross section for *any* energetically allowed process. Using the appropriate integral expression for the breakup amplitude, as we will see below, resolves this apparent contradiction.

IV. INTEGRAL EXPRESSIONS FOR BREAKUP AMPLITUDES

Starting with the formal definition of the breakup amplitude, we can derive both a working expression in terms of $\Psi_{\rm sc}$ and an equivalent two-potential formula involving distorted waves for the final state. Afterward we will apply those working expressions to the two examples of the previous section.

A. Derivation of various matrix elements for breakup amplitudes

The standard theory of rearrangement scattering, discussed by Newton [7], can be applied to the case of breakup, at least so long as the potentials contain only short-range interactions. Such a formulation was specialized to the case of electron scattering by McCartor and Nuttall [15], who took as their starting point the expression for what they called the ''Faddeev amplitude,''

$$f=\langle\mathbf{k}_1,\mathbf{k}_2|V+V(E-H+i\varepsilon)^{-1}V_1|\mathbf{k}_0\alpha\rangle. \tag{26}$$

The final state is simply a product of plane waves, and the initial state is a product of a plane wave and the initial target state,

$$|\mathbf{k}_0\alpha\rangle=\frac{e^{i\mathbf{k}_0\cdot\mathbf{r}_1}}{(2\pi)^{3/2}}\varphi_\alpha(\mathbf{r}_2). \tag{27}$$

For the discussion that follows we will call f the ''breakup amplitude'' to avoid confusion with Faddeev theory in which three-body breakup is treated in terms of two-body T operators. The potential V in Eq. (26) is the full potential, i.e., $H=T+V$, while the potential V_1 is the interaction appropriate to the initial channel. Its operation on the initial state can be replaced by

$$V_1|\mathbf{k}_0\alpha\rangle=(H-E)|\mathbf{k}_0\alpha\rangle \tag{28}$$

so that

$$\begin{aligned}f&=\langle\mathbf{k}_1,\mathbf{k}_2|V+V(E-H+i\varepsilon)^{-1}(H-E)|\mathbf{k}_0\alpha\rangle\\&=\langle\mathbf{k}_1,\mathbf{k}_2|V|\Psi^{(+)}\rangle\\&=\langle\mathbf{k}_1,\mathbf{k}_2|E-T|\Psi^{(+)}\rangle,\end{aligned} \tag{29}$$

where T is the kinetic-energy operator and $\Psi^{(+)}$ is the full scattering wave function in Eq. (1).

We can further specialize Eq. (29) to our two-dimensional examples as

$$f=\langle k_1,k_2|E-T|\Psi^{(+)}\rangle$$
$$=\left(\frac{2}{\pi}\right)\int_0^\infty dr_1\int_0^\infty dr_2\sin(k_1r_1)\sin(k_2r_2)(E-T)$$
$$\times\Psi^{(+)}(r_1,r_2), \tag{30}$$

and we can divide $\Psi^{(+)}$ into an incident and a scattered wave as in Eq. (2) with the incident wave function defined as in Eq. (7) to have permutational symmetry reflecting the spin coupling. Doing so simplifies Eq. (30) further and yields the first of our working expressions for the calculations we will describe below:

$$f=\left(\frac{2}{\pi}\right)\int_0^\infty dr_1\int_0^\infty dr_2\sin(k_1r_1)$$
$$\times\sin(k_2r_2)(E-T)\Psi_{\rm sc}(r_1,r_2). \tag{31}$$

The contribution from Φ_0 vanishes since it is proportional to δ functions between the final and incident momenta, and they cannot be the same.

Equation (31) has a property that the other working expressions below will have, and that is that it depends only on the asymptotic form of the scattering wave function. To see that this is the case, we can use Green's theorem to express the amplitude in terms of a matrix element with the operators operating to the left. The product of free waves in the final state is an eigenfunction of the kinetic energy at the total energy of the system, so only a surface term is left,

$$f=\frac{1}{\pi}\int_S[\sin(k_1r_1)\sin(k_2r_2)\boldsymbol{\nabla}\Psi_{\rm sc}$$
$$-\Psi_{\rm sc}\boldsymbol{\nabla}\sin(k_1r_1)\sin(k_2r_2)]\cdot\hat{\mathbf{n}}\,dS, \tag{32}$$

where it is understood that the surface is to be limited to enclose an infinite volume, and $\hat{\mathbf{n}}$ is the outward unit vector normal to the surface. The gradient in this equation is

$$\boldsymbol{\nabla}=(\partial/\partial r_1,\partial/\partial r_2)=\hat{\boldsymbol{\rho}}\partial/\partial\rho+\hat{\boldsymbol{\alpha}}\frac{1}{\rho}\partial/\partial\alpha. \tag{33}$$

If we insert just the breakup term in Eq. (8) into Eq. (32), it gives the connection with the breakup amplitude in Eq. (8). First the surface integral Eq. (32) is converted to an integral over α, which is done with the stationary-phase approximation using the identity

$$\lim_{\rho\to\infty}\int_0^{\pi/2}F(\alpha)\sin(k_1\rho\cos\alpha)\sin(k_2\rho\sin\alpha)d\alpha$$
$$=-\tfrac{1}{2}F(\alpha_0)\sqrt{2\pi/K\rho}\cos\left(K\rho-\frac{\pi}{4}\right), \tag{34}$$

where $\tan\alpha=k_2/k_1$ is the stationary-phase point. Both Eq. (34) and its derivatives with respect to ρ are used. This sort of manipulation is by now standard in the breakup problem, and we find the relation between the ''Faddeev amplitude'' and the breakup amplitude in the asymptotic form of the wave function to be

$$f=-e^{i3\pi/4}F(k_1,k_2)/\sqrt{2\pi}. \tag{35}$$

The contribution of the discrete channel terms in Eq. (8) to Eq. (32) is a more subtle issue, and it is somewhat awkward to see how it limits to zero using the surface integral representation. Returning to the equivalent volume integral in Eq. (31), we can see that their contribution becomes proportional, for infinite volumes, to δ functions between the final and initial momenta and therefore must vanish. For any finite-sized region of integration, however, they still contribute, and that fact has serious practical consequences, as we will see. It turns out that the contribution of discrete channel terms at any finite value of ρ can render Eqs. (31) and (32) computationally unstable.

We can get other forms of the integral expression for the breakup amplitude in Eq. (31) in which the final states are scattering states of some distorting potential. Expressions of this type have recently been explored extensively [16] in the context of formulating ''ansatz'' approximations for ionization amplitudes. Here, we will investigate their properties in accurate numerical calculations. To begin, write Eq. (31) as

$$f=\left(\frac{2}{\pi}\right)\int_0^\infty dr_1\int_0^\infty dr_2\sin(k_1r_1)\sin(k_2r_2)(E-T)G^{(+)}(E)$$
$$\times(H-E)\Phi_0(r_1,r_2), \tag{36}$$

where $G^{(+)}(E)$ is the full Green's function $(E-H+i\eta)^{-1}$. We then write the potential as the sum of two terms,

$$V=V_1+V_2, \tag{37}$$

where

$$V_1=V_{1a}(r_1)+V_{1b}(r_2) \tag{38}$$

is the sum of two one-body potentials. Note that V_1 can be constructed from any short-ranged one-body potentials, and this derivation does not assume that V_1 is the same as the one-body part of the true interaction potential given in Eq. (6).

The full Green's function satisfies the identity

$$G^{(+)}(E)=g_1^{(+)}(E)+g_1^{(+)}(E)V_2G^{(+)}(E), \tag{39}$$

where

$$g_1^{(+)}(E)=(E-T-V_1+i\eta)^{-1}. \tag{40}$$

With only these relationships and the identity

$$(E-T)G^{(+)}(E)=1+VG^{(+)}(E), \tag{41}$$

we can rearrange Eq. (36) into the form

$$f=\langle k_1,k_2|(E-T)G^{(+)}(E)|(H-E)\Phi_0\rangle$$
$$=\langle k_1,k_2|[1+V_1g_1^{(+)}(E)][1+V_2G^{(+)}(E)](H-E)\Phi_0\rangle. \quad (42)$$

In this equation we can operate to the left with the Hermitian conjugate of the factor $[1+V_1g_1^{(+)}(E)]$, which is the wave operator for the potential V_1. Because this potential is the sum of two one-body operators, the wave operator converts the free waves of the final state into distorted waves. The result is a ''two-potential'' form of the breakup amplitude expression,

$$f=\langle\varphi_{k_1}^{(-)}\varphi_{k_2}^{(-)}|(T+V_1-E)|\Phi_0\rangle+\varphi_{k_1}^{(-)}\varphi_{k_2}^{(-)}|V_2|\Psi^+\rangle \quad (43a)$$

$$=\langle\varphi_{k_1}^{(-)}\varphi_{k_2}^{(-)}|(H-E)|\Phi_0\rangle+\langle\varphi_{k_1}^{(-)}\varphi_{k_2}^{(-)}|V_2|\Psi_{sc}\rangle. \quad (43b)$$

The distorted waves in Eq. (43) satisfy one-body Schrödinger equations. For example,

$$[T_1+V_{1a}(r_1)]|\varphi_{k_1}^{(+)}\rangle=\frac{k_1^2}{2}|\varphi_{k_1}^{(+)}\rangle \quad (44)$$

with the normalization that the incoming wave is $(2/\pi)^{1/2}\sin(k_1r_1)$.

Equation (43) can be simplified further if we use the original driven Schrödinger equation for Ψ_{sc},

$$f=\langle\varphi_{k_1}^{(-)}\varphi_{k_2}^{(-)}|E-T-V_1|\Psi_{sc}\rangle. \quad (45)$$

This two-potential form for the breakup amplitude also has an equivalent surface integral representation, analogous to Eq. (32), that reveals it to depend only on the asymptotic form of Ψ_{sc},

$$f=\tfrac{1}{2}\int_S(\varphi_{k_1}^{(+)}\varphi_{k_2}^{(+)}\nabla\Psi_{sc}-\Psi_{sc}\nabla\varphi_{k_1}^{(+)}\varphi_{k_2}^{(+)})\cdot\hat{\mathbf{n}}\,dS, \quad (46)$$

where the gradient is defined as in Eq. (33). The volume necessary to converge a numerical evaluation of Eq. (45) or (46) can be deduced by going back to Eq. (43). As we will see below, if the distorting potential V_1 is chosen correctly, the potential V_2 can control the numerical properties of these expressions.

B. Application to example breakup problems

Armed with Eqs. (31) and (32) from the original definition of the breakup amplitude, and with Eqs. (45) and (46) expressing the ''two-potential'' form, we can explore their application to the examples of Sec. III. In general the numerical calculations shown here made use of the surface integral forms of these expressions (which are precisely equivalent to the volume integral forms).

First we turn to the exponential potential problem of Sec. III A. Figure 7 shows the singly differential cross section that results from applying Eq. (32) to calculating the breakup amplitude for this case with the radius ρ_0 of the surface

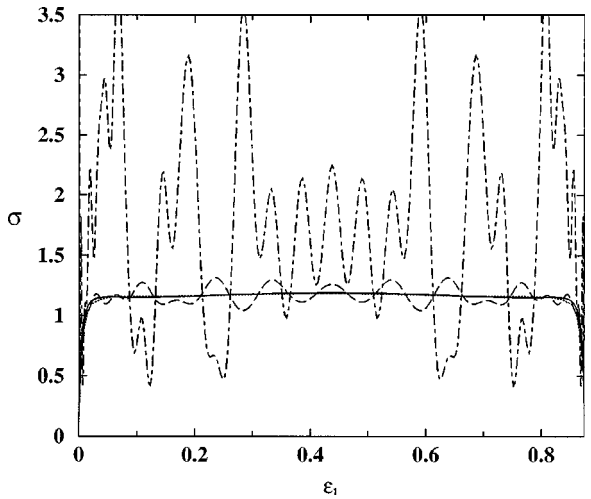

FIG. 7. Singly differential cross section (a_0^2/hartree) vs energy (hartrees) of electron 1 from projected flux evaluation for model of Sec. III A at 35 eV incident energy. Dot-dashed line, results from Eq. (32); dashed line, results from Eq. (32) with projected function $Q\Psi_{sc}$; solid line, results from Eq. (46); dotted line, the nearly indistinguishable results of the projected flux approach after extrapolation.

integral equal to $56a_0$. In this classic case of short-range potentials, it is at first surprising to see that the results are wildly oscillatory. These oscillations evidently arise from the contributions of the discrete channels of the problem (in this case only the elastic channel). To show that this is the case we projected out the elastic channel from Ψ_{sc} using the projection operator $1-\mathbf{P}_0$ constructed using Eq. (11) and recomputed the amplitude from Eq. (32), replacing Ψ_{sc} with $(1-\mathbf{P}_0)\Psi_{sc}$. The resulting cross section oscillates with much decreased amplitude and now oscillates around the correct result. Although it is not shown in Fig. 7, it can be verified that, if the integration volume is increased, the projected form of Eq. (32) eventually converges to the correct result with increasing ρ_0, albeit very slowly.

In contrast, the two-potential formula Eq. (46), using the one-body potentials $v(r_j)$ as the distorting potentials, produces the correct cross section without any oscillations, as is also shown in Fig. 7. The distorted waves in the two-potential formula are, of course, orthogonal to the bound states of the same potential, and their use in Eq. (45) or (46) eliminates the contributions of the discrete channels.

These results lead us to our first assertion regarding the utility of the integral forms of the breakup amplitudes. Equations (26), (29), (31), and (32), while formally correct, are of limited practical utility in calculations that begin with the physical scattering wave function, containing all final channels. Only if the discrete inelastic channel contributions are projected out can these equations give correct results when applied on a finite volume, as they must be in a numerical calculation. They are therefore of limited practical utility, except to formulate distorted-wave-style approximations to the breakup amplitudes.

The two-potential form of the breakup amplitude in Eqs. (45) and (46), on the other hand, suffers from no such limitation, and can be used in practical calculations, provided the

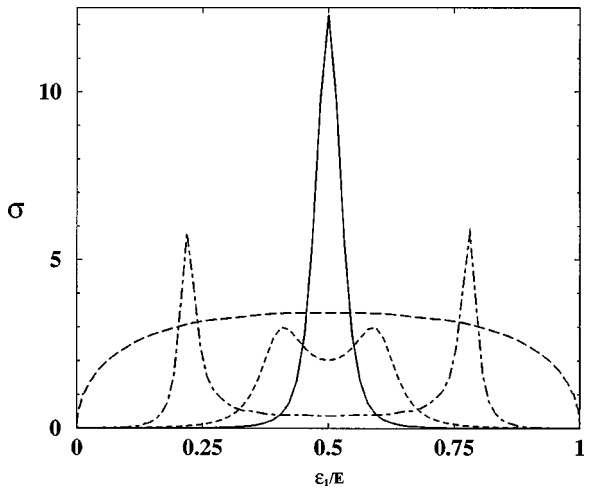

FIG. 8. Singly differential cross section (a_0^2/hartree) vs energy fraction for electron 1 from the two-potential formula for the model problem of Sec. III B; long-dashed line, 15 eV cross section $\times$ 5000; dashed line, 25 eV$\times$5; solid line, 27.91 eV (twice resonance energy); and dot-dashed line, 50 eV$\times$5.

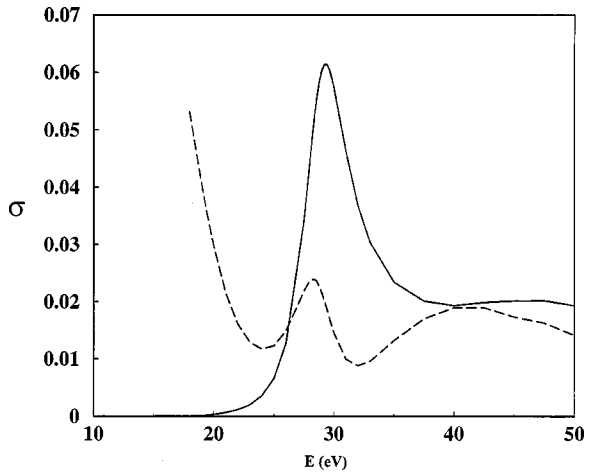

FIG. 9. Total cross sections ($a_0^2/4\pi$) for the model problem of Sec. III B; solid line, breakup; dashed line, elastic scattering.

distorting potential is chosen to be the same as that which supports the discrete states of the target. The first term in Eq. (43a) then vanishes and the two-potential form produces the correct result when applied on a volume that encloses the residual potential V_2. That property of the two-potential form is more dramatically demonstrated in the next example.

Projecting discrete channel contributions from the scattered wave function does not alter the rate at which it reaches its asymptotic form. In the second numerical example, that of Sec. III B, which has an autodetaching resonance that dominates the dynamics, projecting the discrete channel(s) out of $\Psi_{\rm sc}$ does not redeem the utility of Eqs. (31) and (32). In such a case the dynamics of the decay of the resonance causes the wave function to approach its asymptotic form so slowly that Eq. (31) must be applied over impractically large volumes to produce the correct result. However, the two-potential form works perfectly, as shown in Fig. 8 for a range of incident energies, both above and below the resonance energy. Again using the one-body potentials, which support the autodetaching resonance, as the distorting potential, we see that Eqs. (45) and (46) produce the correct breakup amplitude as soon as the volume of the integration region encloses the residual potential. In this case the residual potential is just $V_2(r_1 r_2) = 4\exp(-r_1-r_2)$, and so a very small integration volume, by comparison to the volumes shown in Fig. 5, will suffice.

The singly differential cross sections for the resonance case are interesting in themselves. The cross section must satisfy the symmetry condition

$$\sigma(k_1,k_2)=\sigma(k_2,k_1). \tag{47}$$

We display this symmetry in an obvious way by plotting the cross section as a function of the energy of one of the two outgoing electrons,

$$\sigma_{\rm SDCS}(\varepsilon)\equiv\sigma(\sqrt{2\varepsilon},\sqrt{K^2-2\varepsilon}), \tag{48}$$

as a function of the fraction of the total energy available to the exiting electrons, $\varepsilon/(K^2/2)$. These plots are symmetric about the midpoint for any incident energy. The resonance feature corresponding to electron 1 having the resonance energy $\varepsilon=\varepsilon_{\rm res}$ must also appear when the other electron has that energy, or at $\varepsilon=K_2/2-\varepsilon_{\rm res}$. The singly differential cross section therefore has two ''teeth'' at any total energy above the resonance energy, and those are clearly seen in Fig. 8.

There is one energy above the resonance energy where there is only one ''tooth'' and that is at the total energy where the energy available to the two exiting electrons is exactly twice the resonance energy of the autodetaching state. In that case the two peaks merge into a more intense peak at $\varepsilon=K^2/4$ because, in the singlet case, the two resonance features interfere constructively. That case is also shown in Fig. 8. Not shown in Fig. 8 is the triplet case, in which the two features interfere destructively. All these observations must, of course, be modified to take into account the finite width of the autodetaching (or autoionizing) resonance. The peaks can overlap, and in the triplet case the destructive interference is only at exactly the point $\varepsilon=K^2/4$.

In the singlet case, one might expect that the total cross section for breakup, which with our normalizations is defined by

$$\sigma_{\rm tot}^{\rm ion}=\int_0^{k^2/2}\sigma_{\rm SDCS}(\varepsilon)\,d\varepsilon, \tag{49}$$

might have its peak at the energy that corresponds to having twice the resonance energy available to the exiting electrons. Figure 9 shows that, at least in this example, that is the case.

In the case of electron-impact ionization via autoionizing states, there have already been some experiments done to begin to explore the interference effects in the angular distributions that these calculations hint at [17–19]. Some theoretical efforts have been made along the lines of distorted-wave approximations [20] and exploration of the formal energy dependence of the breakup amplitudes in the pres-

ence of autoionizing states [21,22]. Accurate theoretical predictions of the interference effects associated with this process will almost certainly have to be done using an approach similar to the two-potential formula described here for extracting the breakup amplitudes.

V. CONCLUSION

We have explored various methods for calculating the breakup cross section when presented with an effectively exact numerical representation of the scattered wave function on a finite region of space. We have found that the simple prescription of extrapolating the quantum-mechanical flux becomes impractical when breakup can proceed through excitation of resonance states of the target. In such cases, the component of the scattered wave that describes breakup approaches its asymptotic form very slowly, and indeed well beyond the range of the interaction potentials, even in the case of short-range interactions. In such cases integral expressions for the breakup amplitude offer a more practical method for evaluating the required scattering information, but can yield numerically unstable results unless proper steps are taken to eliminate spurious contributions arising from discrete two-body channels. We have shown that formal rearrangement theory allows one to express the breakup amplitude as an integral involving target continuum states as distorted waves. We have demonstrated that such an expression has the desirable properties of being numerically stable and not requiring the wave function beyond the range of the interaction potentials, even when resonance states are involved in the scattering dynamics.

While the numerical examples we have studied reveal features that we feel are likely to be present in real ionization problems, it is important to bear in mind that the results we have obtained apply only to collisions involving short-ranged interactions. In the formal theory of ionization in which the Coulomb potentials are treated exactly to infinity, the integral expression for the breakup amplitude involves a product of distorted waves for the scattered and ejected electrons in the final state which are Coulomb functions with effective charges Z_1 and Z_2. These effective charges are not entirely arbitrary since they must be chosen to satisfy the Peterkop relation [8,9]

$$\frac{Z_1}{k_1}+\frac{Z_2}{k_2}=\frac{1}{k_1}+\frac{1}{k_2}-\frac{1}{|\mathbf{k}_1-\mathbf{k}_1|}. \tag{50}$$

Thus, in the formal theory, the distorted waves are not the continuum states of the target atom and thus cannot be chosen so as to eliminate the one-body operators from the integral expression for the breakup amplitude. The formal theory, however, has not provided a path to practical calculations and has, to date, never been implemented in an *ab initio* calculation. If the true Coulomb interactions can be truncated at large distances and the cross sections then extrapolated to infinity (and our recent work on electron-H ionization [4] argues that they can), then the two-potential formalism discussed here for short-ranged potentials should be applicable. This subject will be pursued in future studies.

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