ARTICLES

Secular perturbation theory of long-range interactions

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Solutions of Schrödinger's equation for systems interacting with long-range power-law potentials, $V_{\rm int}(r) \propto r^{-n}$ with $n \geq 2$, are cast in the form of a power series in $V_{\rm int}$. These perturbations lead to secular divergences that are eliminated by renormalizing the angular-momentum quantum number. Long-known perturbation techniques in classical mechanics and quantum-field theory yield modified effective-range formulas, quantum-defect functions, and solutions of close-coupling equations for wave propagation in long-range fields. As an example, we extract in first-order perturbation theory a modified effective-range expansion for the phase shift of an electron interacting with an atomic $1/r^4$ polarization potential. Near thresholds, the method is applicable to all power-law potentials with $n \geq 2$, and to their combinations, as well as to multichannel problems involving anisotropic potentials.

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I. INTRODUCTION

Long-range interactions between quantum systems can drastically affect fragmentation processes, particularly near thresholds where interaction times are enhanced by the low velocity of the receding fragments [1]. We refer here to the multipole interactions, $V(r) \propto r^{-n}$ with $n \geq 2$, which are shown in Ref. [1] to possess an infinite effective range. ("Short-range" interactions, in contrast, decay at least exponentially with distance.) Much effort has been devoted to analytical methods for calculating contributions to phase shifts from such residual multipole interactions between fragmenting systems [2–4]. Only after successfully incorporating these long-range effects can one focus on the difficult problems that accompany the more complicated interactions at short distances.

Historically, long-range fields have been dealt with by rather cumbersome mathematical analyses associated with the special functions appropriate to each type of interaction. Even those few cases where known special functions apply [5,6] require extremely tedious manipulations [7] to extract essential information, such as the density of states and the energy dependent phase accumulated in the long-range field. Furthermore, analytical treatment of these long-range interactions has dealt essentially with single-channel problems, leading to success only where long-range motions are treated by solving a single (uncoupled) radial equation. The analysis of long-range interactions thus remains confined to essentially adiabatic phenomena [8]; the relative motion of the receding fragments is neglected in lowest order.

An alternative to the analytical methods described above is an expansion of the wave function in the form of an oscillatory function, $e^{\pm ikr}$, multiplied by a series in inverse powers of r [9]. Gailitis [10] improved upon these solutions by replacing the oscillatory function with a solution of an "unperturbed" radial equation with a modified angular momentum barrier. The *ad hoc* angular-momentum L of Ref. [10] was selected arbitrarily and, as a matter of convenience, L was generally integral, except in cases of long-range dipole $(1/r^2)$ potentials. These series diverge rapidly close to threshold, where they apply only for very large fragment separations. The use of Padé approximants [11] allows these series to be evaluated numerically, accelerating their convergence, and extending their region of applicability to much smaller distances.

This paper aims at demonstrating how wave functions of fragments moving in long-range fields can often be calculated accurately and analytically by relatively elementary and long-established methods of perturbation theory. The key element of this analysis recognizes that the divergence of a series expansion in powers of the long-range potential stems from *secular* perturbations—that is, from driving terms that resonate with the unperturbed system. The standard procedure for eliminating secular terms renormalizes the "effective frequency" of the system, be that a classical oscillator frequency or a self-energy [12]. In the context of long-range interactions, angular-momentum quantum numbers must be renormalized to shift the response out of resonance with the driving term. The energy-dependent "shift" of the angular momentum is simply related to the phase accumulated in the long-range field.

This paper provides a simple procedure for calculating accurate near-threshold wave functions for general long-range potentials. We focus in Secs. III and V on the example of a polarization potential both for clarity and because this case has received the most attention in the literature. Generalizations to other long-range fields fol-

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low naturally, and need not be given explicitly here—though an example is cited in Sec. VI. The relationship of our analytical solutions with the numerical solutions of Refs. [10] and [11] is discussed in Sec. VI. Wave functions resulting from this construction may serve in the study of ultracold collisions [13], as well as in more traditional applications of effective-range and quantum-defect theories.

II. REVIEW OF SECULAR PERTURBATIONS

Elementary textbooks in classical mechanics cite the example of an oscillator containing a small anharmonic component, ϵ [14]:

$$\ddot{x}+\omega_0^2 x=\epsilon x^3 . \quad (1)$$

Attempting a solution as a series in powers of ϵ,

$$x(t)\approx x_0(t)+\epsilon x_1(t)+\cdots , \quad (2)$$

leads to a first-order inhomogeneous equation with a component of the driving force proportional to a solution of the homogeneous or unperturbed equation. This component is termed "secular" since it drives the system at resonance, leading to a divergence of the amplitude with time. A perturbative approach is still possible by renormalizing the frequency,

$$\omega_0^2=\omega^2+\epsilon\lambda_1+\epsilon^2\lambda_2+\cdots , \quad (3)$$

and adjusting the constants λ_n to eliminate secular terms. Equation (3) is then inverted to determine the renormalized frequency ω.

III. THE POLARIZATION POTENTIAL

The analysis of long-range forces between atomic or molecular fragments proceeds in close analogy with the above example. Consider a long-range polarization potential that varies inversely with the fourth power of the distance between the fragments, yielding Schrödinger's radial equation

$$\left[-\frac{1}{2r^2}\frac{d}{dr}r^2\frac{d}{dr}+\frac{l(l+1)}{2r^2}-\frac{\beta^2}{2r^4}-E\right]\Psi(r)=0 , \quad r>r_0 , \quad (4)$$

in units with $e=\hbar=\mu=1$, where μ represents the reduced mass of the fragments. This equation can be transformed into Mathieu's equation [15], which possesses thoroughly studied, but quite complicated, solutions [7]. We instead rewrite Eq. (4) (for $E>0$) in terms of $z=kr$, $\Psi(r)=\sqrt{k/z}\,M(z)$, and $k=\sqrt{2E}$, yielding

$$\left[z^2\frac{d^2}{dz^2}+z\frac{d}{dz}+z^2-(l+\tfrac{1}{2})^2\right]M(z)=-\frac{\Delta}{z^2}M(z) . \quad (5)$$

The factor $\Delta=2E\beta^2$ will index the order of our perturbation series.

To see how Eq. (5) leads to secular perturbations, consider a series expansion of the solution in the form

$$M(z)=\sum_{n=0}^{\infty}\Delta^n M^{(n)}(z) . \quad (6)$$

The resulting zeroth-order solution reduces to the Bessel function, $M^{(0)}(z)=J_{l+1/2}(z)$, representing free motion of the receding fragments. The first-order equation is then

$$\left[z^2\frac{d^2}{dz^2}+z\frac{d}{dz}+z^2-(l+\tfrac{1}{2})^2\right]M^{(1)}(z)=-\frac{1}{z^2}J_{l+1/2}(z) . \quad (7)$$

Its driving term expands into a Bessel series through the recurrence relation

$$\frac{1}{z^2}J_{l+1/2}(z)=\frac{J_{l-3/2}(z)}{4(l+\frac{1}{2})(l-\frac{1}{2})}+\frac{J_{l+1/2}(z)}{2(l-\frac{1}{2})(l+\frac{3}{2})}+\frac{J_{l+5/2}(z)}{4(l+\frac{1}{2})(l+\frac{3}{2})} , \quad (8)$$

whose second term on the right-hand side solves the homogeneous equation—thus constituting a secular perturbation and resulting in a divergence of the first-order solution. All higher equations in the series also include secular terms.

Elimination of the secular terms is readily accomplished, following Eq. (3), by adjusting the angular-momentum quantum number:

$$(l+\tfrac{1}{2})^2=(\gamma+\tfrac{1}{2})^2+\sum_{n=1}^{\infty}\Delta^n\Gamma^{(n)} . \quad (9)$$

The renormalized angular momentum γ will be determined below. Substituting Eqs. (6) and (9) into Eq. (5), and equating coefficients of equal powers of Δ, yields for the lowest three orders of Δ

$$\left[z^2\frac{d^2}{dz^2}+z\frac{d}{dz}+z^2-(\gamma+\tfrac{1}{2})^2\right]M_\gamma^{(0)}(z)=0 ,$$
$$\left[z^2\frac{d^2}{dz^2}+z\frac{d}{dz}+z^2-(\gamma+\tfrac{1}{2})^2\right]M_\gamma^{(1)}(z)=\left\{\Gamma^{(1)}-\frac{1}{z^2}\right\}M_\gamma^{(0)}(z) , \quad (10)$$
$$\left[z^2\frac{d^2}{dz^2}+z\frac{d}{dz}+z^2-(\gamma+\tfrac{1}{2})^2\right]M_\gamma^{(2)}(z)=\Gamma^{(2)}M_\gamma^{(0)}(z)+\left\{\Gamma^{(1)}-\frac{1}{z^2}\right\}M_\gamma^{(1)}(z) .$$

The zeroth-order solution of Eq. (10) is again a Bessel function, $J_{\gamma+1/2}(z)$, albeit of a shifted order. The first-order equation is similar to Eq. (7), but includes the parameter $\Gamma^{(1)}$, whose value is chosen to eliminate the secular term in Eq. (8):

$$\Gamma^{(1)}=\frac{1}{2(\gamma-\frac{1}{2})(\gamma+\frac{3}{2})}=\frac{1}{2[(\gamma+\frac{1}{2})^2-1]} . \quad (11)$$

This choice of $\Gamma^{(1)}$ allows one to write a first-order solution of Eq. (10) by inspection:

$$M_\gamma(z)=M_\gamma^{(0)}(z)+\Delta M_\gamma^{(1)}(z)+O(\Delta^2)$$
$$=J_{\gamma+1/2}(z)+\frac{\Delta}{16(\gamma+\frac{1}{2})}\left[\frac{J_{\gamma-3/2}(z)}{(\gamma-\frac{1}{2})^2}-\frac{J_{\gamma+5/2}(z)}{(\gamma+\frac{3}{2})^2}\right]+O(\Delta^2)\,, \qquad (12)$$

with the value of γ also set to second order in Δ,

$$(\gamma+\tfrac{1}{2})^2=(l+\tfrac{1}{2})^2-\frac{\Delta}{2[(\gamma+\frac{1}{2})^2-1]}+O(\Delta^2)\,. \qquad (13)$$

Note that no homogeneous contribution has been added to the particular solution, $M_\gamma^{(1)}(z)$. This would amount to a Δ-dependent renormalization of the *amplitude* of the zeroth-order solution—and, therefore, to an amplitude renormalization of the entire function $M_\gamma(z)$. The "first-order" solution Eq. (12) manifestly includes terms in Δ to *all orders* through the implicit dependence of γ on Δ, Eq. (13). Solving Eq. (13) for γ yields a solution that reduces to l in the limit $E\to 0$, namely,

$$\gamma=l-\frac{4\beta^2E}{(2l+1)(2l+3)(2l-1)}+O(\Delta^2)\,. \qquad (14)$$

The asymptotic form of Eq. (12) as $z\to\infty$ shows the phase of $M_\gamma(z)$ to depart from that of a free wave of angular momentum l by the amount

$$\frac{\pi}{2}(l-\gamma)\approx\frac{2\pi\beta^2E}{(2l+1)(2l+3)(2l-1)}\equiv\tan(\delta_l)\,, \qquad (15)$$

which coincides with the well-known threshold phase shift for polarization potentials [16].

Proceeding to second order, the secular perturbation on the third line of Eq. (10) is eliminated by setting

$$\Gamma^{(2)}=\frac{5(\gamma+\frac{1}{2})^2+7}{32[(\gamma+\frac{1}{2})^2-4][(\gamma+\frac{1}{2})^2-1]^3}\,, \qquad (16)$$

whereby

$$M_\gamma^{(2)}(z)=\frac{1}{32(\gamma+\frac{1}{2})(\gamma-\frac{1}{2})(\gamma+\frac{3}{2})}\left[\frac{J_{\gamma-3/2}(z)}{(\gamma-\frac{1}{2})^2(\gamma-\frac{5}{2})}-\frac{J_{\gamma+5/2}(z)}{(\gamma+\frac{3}{2})^2(\gamma+\frac{7}{2})}\right]+\frac{1}{512(\gamma+\frac{1}{2})}\left[\frac{J_{\gamma-7/2}(z)}{(\gamma-\frac{1}{2})^2(\gamma-\frac{3}{2})^2(\gamma-\frac{5}{2})}+\frac{J_{\gamma+9/2}(z)}{(\gamma+\frac{3}{2})^2(\gamma+\frac{5}{2})^2(\gamma+\frac{7}{2})}\right]\,. \qquad (17)$$

Equation (16) implies that each successive order of this perturbation theory requires solving an algebraic equation for γ, Eq. (9), in terms of Δ and l.

For the polarization potential, this perturbation procedure is equivalent to a series expansion in Δ of the *exact* Mathieu function solution, as demonstrated by noting that the perturbation series, if taken to all orders, has the form

$$M_\gamma(z)=\sum_{n=-\infty}^{\infty}a_\gamma^{(n)}J_{\gamma+1/2+2n}(z)\,,\quad a_\gamma^{(0)}=1\,, \qquad (18)$$

which is equivalent to a Laurent series [17] for the Mathieu function. (In this context, γ is referred to as a *characteristic exponent;* it identifies the solution according to its analytic continuation around the irregular singularity at $z=\infty$ [18].) Substitution of Eq. (18) into Eq. (5) and use of Eq. (8) yields a three-term recurrence relation for $a_\gamma^{(n)}$, which can be written in terms of a pair of infinite continued fractions. The Laurent series converges for all values of z between the two irregular singular points of Eq. (5), at $z=0$ and ∞ [7], as verified by showing that

$$\lim_{N\to\infty}\frac{a_\gamma^{(\pm|N|)}}{a_\gamma^{(\pm|N-1|)}}\propto\frac{1}{N^2}\to 0\,. \qquad (19)$$

The perturbative approach outlined above results from an expansion of the continued fractions in powers of Δ. In this example, the elimination of secular perturbations amounts, therefore, to determining the renormalized angular momentum γ by *enforcing* convergence of the series.

IV. CALCULATION OF THE PHASE SHIFTS

The perturbation series described above serves to generate a pair of linearly independent functions outside the reaction zone, $r>r_0$, for quantum-defect theory or R-matrix applications. The perturbative solution $M_\gamma(z)$ was generated from a zeroth-order function $J_{\gamma+1/2}(z)$. A second linearly-independent solution, $M_{-\gamma-1}(z)$, arises from the alternative zeroth-order function $M^{(0)}(z)=J_{-\gamma-1/2}(z)$. This pair of independent solutions serves then as a base pair to be joined to an interior solution, $\psi_{\rm int}(r)$, at a matching radius, $r=r_0$. At $r<r_0$, Schrödinger's equation does not have the simple form of Eq. (4), but its solution $\psi_{\rm int}(r)$ may be recast at large r according to

$$r\psi_{\rm int}(r)\underset{r>r_0}{\sim}\sqrt{r}\,A\,[M_\gamma(kr)+\tilde{K}M_{-\gamma-1}(kr)]\,. \qquad (20)$$

The wave function in the reaction zone $r<r_0$, generated independently, is characterized at each $r>r_0$ by its logarithmic derivative,

$$\frac{d}{dr}[\ln r\psi_{\rm int}(r)]_{r=r_0}\equiv b(E)+\frac{1}{2r_0}\,, \qquad (21)$$

an analytic function of the energy. The detailed matching, Eq. (20), reduces to the equation

$$b(E)+\frac{1}{2r_0}=\frac{d}{dr}[\ln\{\sqrt{r}M_\gamma(kr)+\tilde{K}\sqrt{r}M_{-\gamma-1}(kr)\}]_{r=r_0}\,, \qquad (22)$$

or

$$\tilde{K} = -\frac{M_\gamma(kr_0)}{M_{-\gamma-1}(kr_0)} \left[\frac{b(E) - \frac{d}{dr}\ln M_\gamma(kr)}{b(E) - \frac{d}{dr}\ln M_{-\gamma-1}(kr)} \right]_{r=r_0}, \tag{23}$$

whose term in square brackets is an analytic function of the energy.

The base pair functions M_γ and $M_{-\gamma-1}$ oscillate with a phase difference not equal to 90° as $r \to \infty$ (as verified from the asymptotic form of the Bessel functions) whereby arctan $(\tilde{K})$ does not represent the asymptotic phase shift of $\Psi(r)$. Expressing the asymptotic form of Eq. (20) as $\sin(kr - l\pi/2 + \delta)$ yields the correct phase shift in the form

$$K \equiv \tan(\delta) = \frac{\tan[\pi(l-\gamma)/2] + K'}{1 - K' \tan[\pi(l-\gamma)/2]}, \tag{24}$$

where

$$K' \equiv \frac{\tilde{K}\cos\pi\gamma}{1 - \tilde{K}\sin\pi\gamma}. \tag{25}$$

Equations (24) and (25) hold to any order in Δ.

As the energy increases from threshold, the value of γ determined from Eq. (9) can become complex. The extraction of the phase shift proceeds in this case as above, except for replacing the base pair M_γ and $M_{-\gamma-1}$ with their real parts; the phase shifts and wave functions obtained in this manner depend on energy smoothly. While the convergence of our perturbation series far from threshold is slow, the long-range potentials become less important as the kinetic energy of the separating fragments increases. Methods of evaluating the exact Mathieu functions far from threshold are available for the polarization potential [19].

V. A MODIFIED EFFECTIVE-RANGE EXPANSION

In order to illustrate the utility of our perturbed wave functions, we now show that the modified effective-range expansion can be obtained directly from Eq. (23) and from the first order results, Eqs. (12) and (14). The concept of an "effective range" of fragment interactions emerged from expanding the phase shift of an escaping fragment's wave function to lowest order in its escape energy or velocity [20]. Here we illustrate the flexibility of our wave functions $M_\gamma(kr)$ by extracting an effective-range parameter $\tilde{K}$ from Eq. (23) and the first order results Eqs. (12) and (14).

Substituting Eq. (14) into Eqs. (24) and (25), and retaining terms of order E, gives

$$\tan(\delta) = \tan(\delta_t) + (-1)^l \tilde{K} \tag{26}$$

in terms of the threshold phase shift δ_t from Eq. (15). Evaluating the first-order wave functions at $r = r_0$ for $kr_0 \ll 1$ and neglecting terms of order β^4 yields, from Eq. (23),

$$\begin{aligned}\tilde{K} = -(-1)^l \left[\frac{kr_0}{2}\right]^{2l+1} & \frac{\pi(l+\frac{1}{2})}{[\Gamma(l+\frac{3}{2})]^2} \\ \times & \left[1 - \frac{4}{\pi}\tan(\delta_t)\ln\left[\frac{kr_0}{2}\right]\right] \left[\frac{r_0 b(0) - l - \frac{1}{2}}{r_0 b(0) + l + \frac{1}{2}}\right] \\ \times & \left[1 + \frac{\beta^2(l+\frac{1}{2})}{2r_0^2(l-\frac{1}{2})(l+\frac{3}{2})} \right. \\ & \left. \times \left[1 + \frac{2r_0 b(0) + 2}{r_0^2 b^2(0) - (l+\frac{1}{2})^2}\right]\right].\end{aligned} \tag{27}$$

[Note that the product $\tan(\delta_t)\ln(kr_0/2)$, while nonanalytic, vanishes at threshold.] Equations (26) and (27) represent a modified effective-range expansion, including nonanalytic terms [16,21]. They parametrize the near-threshold energy dependence of the phase shift in terms of the threshold value of the logarithmic derivative of the inner wave function at a matching radius r_0. Similar expansions for any inverse power-law potential ($\propto r^{-n}$ for $n \geq 2$), or for any combination of such potentials, result from straightforward application of the secular perturbation method.

VI. EXAMPLES AND DISCUSSION

While the modified effective-range expansions hold only very near threshold ($kr_0 \ll 1$), our perturbative wave functions hold over a much larger energy range. Even the zeroth-order wave function, including the first-order correction to the angular momentum, often constitutes an effective approximation to asymptotic wave functions. Figure 1 presents the long-range or "polarization" phase shift

$$\delta_l(\Delta) \equiv \frac{\pi}{2}(l - \gamma) \tag{28}$$

for an $l = 1$ partial wave. The two curves shown were obtained by expanding the renormalization Eq. (9) to first order (dashed curve) and second order (solid curve) in Δ. The second-order equation was solved by expressing Eq.

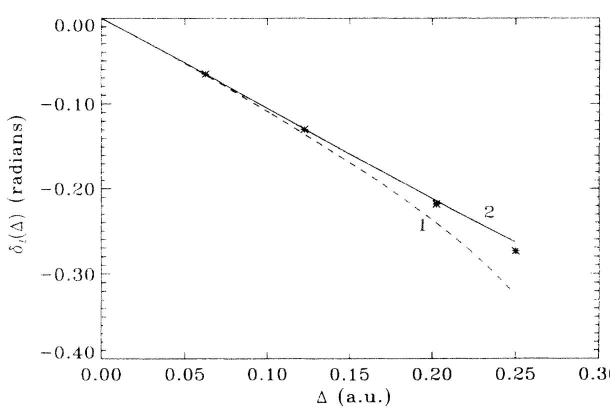

FIG. 1. The long-range phase shift for an $l = 1$ partial wave in a polarization potential, with polarizability β^2. The phase shift is plotted versus $\Delta = 2E\beta^2$. Dashed curve: first order. Solid curve: second order. Asterisks: exact results of Ref. [6].

(9) as $x=f(x)$ with $x=(\gamma+\frac{1}{2})^2$ and iterating $x_n=f(x_{n-1})$ from the first-order solution. This procedure converged in a very few iterations over the energy range shown. Our results compare favorably with the exact results of Ref. [6], obtained using the Mathieu functions.

Applications of this method to other potentials of interest is illustrated here by comparing our perturbation expansion with results of a variable-phase calculation [2] of long-range phase shifts for positron scattering by atomic helium. In this example, both the dipole ($\alpha_1=\beta^2=1.322$ a.u.) and quadrupole ($\alpha_2=2.328$ a.u.) polarizabilities contribute to the long-range motion:

$$V(r) \underset{r>r_0}{\sim} -\frac{\alpha_1}{2r^4}-\frac{\alpha_2}{2r^6} . \tag{29}$$

The secular perturbation method recasts Eq. (9), to order α_1 and α_2, as

$$(l+\tfrac{1}{2})^2=(\gamma+\tfrac{1}{2})^2+\frac{E\alpha_1}{[(\gamma+\frac{1}{2})^2-1]} + \frac{E^2}{[(\gamma+\frac{1}{2})^2-4][(\gamma+\frac{1}{2})^2-1]} \times\left[\frac{3\alpha_2}{2}+\frac{\alpha_1^2[5(\gamma+\frac{1}{2})^2+7]}{8[(\gamma+\frac{1}{2})^2-1]^2}\right] . \tag{30}$$

The long-range phase shifts resulting from Eqs. (28) and (30) are shown in Table I for an $l=2$ partial wave and are compared with those of Ref. [2]. Results of Ref. [2] for other partial waves have been similarly tested with good agreement.

The above procedure extends to solving close-coupled radial equations for waves propagating in anisotropic fields. In this context, it is closely related to the asymptotic expansions developed in Ref. [10] and implemented by Noble and Nesbet [11]. While a detailed comparison of our methods is beyond the scope of this manuscript, we note, as discussed above, that a modified angular momentum (denoted by L) is also used in Ref. [10], though no criteria for the choice of L was specified. In fact, it is not difficult to show that convergence of the Gailitis series of arbitrary r values hinges on the proper choice of this variable, for the recurrence relations relating coefficients of the power series are equivalent to an infinite set of homogeneous linear equations whose determinant will vanish only for particular values of L. The secular perturbation theory described here may simply correspond to an expansion of that determinant in powers of Δ, though this connection remains to be investigated in detail.

TABLE I. Long-range phase shifts for positrons scattering from helium atoms at low energy. The $l=2$ phase shifts given by Eqs. (28) and (30) are compared to those of Table 2 of Ali and Fraser [2].

$k=\sqrt{2E}$	Ref. [2]	This work
0.025	0.000 025	0.000 025
0.050	0.000 099	0.000 099
0.075	0.000 223	0.000 223
0.100	0.000 398	0.000 398
0.200	0.001 625	0.001 625
0.300	0.003 774	0.003 776
0.400	0.007 005	0.007 017
0.500	0.011 538	0.011 591
0.600	0.017 658	0.017 830
0.700	0.025 710	0.026 183
0.800	0.036 103	0.037 260
0.900	0.049 303	0.051 914
1.000	0.065 831	0.071 414

Finally, we note that a renormalized angular momentum occurs quite naturally in problems involving long-range dipole potentials ($n=2$), since a permanent dipole moment modifies the effective centrifugal barrier directly [22,23]. In this case, the value of l in Eq. (4) need not be an integer. Effects of octupole and higher-order moments on electron scattering from polar systems [24] therefore fall within the scope of our analysis. Such an application to electron scattering from polar systems is in progress and will be reported elsewhere.

In conclusion, we have demonstrated that near-threshold wave functions for atomic or molecular fragments may be generated using elementary methods of perturbation theory, provided that secular divergences are eliminated. This is accomplished by renormalizing the effective centrifugal barrier for radial motion.

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