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Two-photon detachment cross sections and dynamic polarizability of H^- using a variationally stable, coupled-channel hyperspherical approach

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We present a generalization of the variationally stable method of Gao and Starace [B. Gao and A. F. Starace, *Phys. Rev. Lett.* **61**, 404 (1988); *Phys. Rev. A* **39**, 4550 (1989)] for two-electron atoms and ions that incorporates a coupled-channel adiabatic hyperspherical approach. Using this approach, we report results for two-photon detachment of H^- , in which we have coupled one, two, three, and four adiabatic hyperspherical channels within each term level of the initial, intermediate, and final states. We present results also for the dynamic polarizability of H^- as well as for the one-photon detachment cross section. Comparisons are given with results of prior work.

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I. INTRODUCTION

The hydrogen negative ion, one of the fundamental three-body Coulomb systems, has long served as a testing ground for theoretical methods aimed at the accurate treatment of electron correlation effects. This is true also for the case of multiphoton detachment processes, for which the number of theoretical calculations including at least some electron correlation effects has grown substantially since the mid-1980s [1–11]. However, significant disparities exist even among the results of only these more accurate calculations. According to an analysis of Liu *et al.* [8], a key reason for part of the disparities is the extreme sensitivity of multiphoton cross sections to any errors in the theoretical value of the electron affinity. Correspondingly Liu *et al.* [8] as well as Dörr *et al.* [11] empirically altered their calculations so that the electron affinities obtained agreed with that of an accurate variational calculation [12], resulting in large shifts in their predicted multiphoton detachment cross sections, which then agree with one another. However, for photon energies above about 0.3 eV there are three other calculations [3(d),7,10] which lie 8–10 % below the adjusted results of Refs. [8,11]. Moreover, in the region below 0.3 eV all three of these calculations [3(d),7,10] disagree with one another. Although the number of experimental measurements of multiphoton detachment of H^- has been growing [13–18], so far these either have measured total yields for all multiphoton detachment processes [13,14] or else have focused on frequencies in the excess photon detachment region of the spectrum [15–18]. Thus, for the most fundamental three-body Coulomb system, there does not exist either a consensus among the theoretical predictions or an experimental measurement of the two-photon detachment cross section in the energy region between the two-photon and one-photon thresholds.

Our aim in this paper is to provide benchmark results for the two-photon detachment cross section of H^- for photon energies up to the one photon threshold. For this purpose we have combined the variationally stable method of Gao and Starace [19,20] with a coupled-channel, adiabatic hyperspherical approach [21–25]. The variationally stable method allows one to control errors in calculating perturbation ma-

trix elements of second and higher order by ensuring that the error in the matrix element is of second order in any errors that occur in representing the generally infinite summations over intermediate states. The adiabatic hyperspherical representation is known, on the other hand, to provide an excellent basis for describing correlations in two-electron states [25–27] as well as for describing single- and multiphoton processes in two-electron systems [28]. For single-photon detachment of H^- , coupled-channel adiabatic hyperspherical calculations have obtained excellent results [29]. The single-channel adiabatic hyperspherical representation was first used to calculate two-photon detachment of H^- by Fink and Zoller [2]. However, the single-channel adiabatic hyperspherical results of Liu *et al.* [8] for two- and three-photon detachment of H^- are probably more accurate because they employed the variationally stable method of Refs. [19,20] and also empirically adjusted the well depth of the ground-state adiabatic hyperspherical potential so that the electron affinity agreed with that of Pekeris [12]. Our results, which entail no empirical adjustments, show the effects of including one to four coupled adiabatic hyperspherical channels. A key finding is that our three- and four-coupled-channel results for the two-photon cross section (for both linearly polarized and circularly polarized light) are virtually identical, and have qualitatively the same shape as the results of Refs. [8,11], but are modestly lower in magnitude than those results, thereby agreeing with results of Refs. [3(d),7] above photon energies of 0.3 eV and with the *B*-spline results of van der Hart [10] for all energies.

We have carried out a number of additional calculations to enable us (and readers) to judge the reliability of our approach. The main question to be answered in a coupled-channel calculation is, how many coupled channels are required? For this purpose we have first calculated the single-photon detachment cross section of H^- using one to four coupled channels. In the energy region with which we are concerned in this paper, our three- and four-coupled-channel results for the single-photon detachment cross section are in excellent agreement with the variational results of Stewart [30], which are essentially identical to recent results of Abrashkevich and Shapiro [31], who used the hyperspherical

artificial-channel method. An additional check of our use of four coupled channels as well as of our combination of the variationally stable method with the coupled-channel, adiabatic hyperspherical approach is provided by our results for the dynamic polarizability of H^- , for which there are a number of very accurate results with which to compare. Our results for the dynamic polarizability agree well with the accurate results of Chung [32] and of Pipin and Bishop [33], which in turn lie within the rigorous upper and lower bounds of Glover and Weinhold [34]. Our results for the single-photon detachment cross section as well as for the dynamic polarizability of H^- thus confirm the accuracy of the approach we employ to obtain our results for the two-photon detachment cross section of H^- up to the single-photon detachment threshold.

In Sec. II we outline the theoretical aspects of the present approach, reviewing briefly the variationally stable method of Refs. [19,20], the coupled-channel adiabatic hyperspherical approach [21–25], and the application of these two methods to two-photon detachment of H^- . In Sec. III we present our results, showing first those for the single-photon detachment cross section and for the dynamic polarizability of H^- , and then presenting our results for the two-photon detachment cross section for both linearly and circularly polarized incident light. Finally, in Sec. IV we discuss our results and present some conclusions.

II. THEORY

A. Variationally stable method

The N th-order transition matrix element between an initial state $|i\rangle$ and a final state $|f\rangle$ is written as

$$T_{i \rightarrow f}^{(N)} = \langle f | D \frac{1}{E_{N-1} - H} D \cdots D \frac{1}{E_2 - H} D \frac{1}{E_1 - H} D | i \rangle, \quad (1)$$

where D is a transition operator, E_n are intermediate state energies, and H is the unperturbed Hamiltonian. For example, for an N th-order multiphoton process, D is the electric dipole operator, and $E_n \equiv E_i + n\hbar\omega$, where ω is the photon frequency and E_i is the initial state energy. Similarly to the Dalgarno-Lewis technique [35], we define two intermediate states,

$$|\lambda\rangle = \frac{1}{E_{N-1} - H} D \cdots D \frac{1}{E_2 - H} D \frac{1}{E_1 - H} D | i \rangle \quad (2)$$

and

$$\langle \lambda' | = \langle f | D \frac{1}{E_{N-1} - H} D \cdots D \frac{1}{E_2 - H} D \frac{1}{E_1 - H}. \quad (3)$$

With these two new states, the matrix element (1) may be rewritten in three different but completely equivalent ways,

$$T_{i \rightarrow f}^{(N)} = \langle f | D | \lambda \rangle, \quad (4)$$

$$T_{i \rightarrow f}^{(N)} = \langle \lambda' | D | i \rangle, \quad (5)$$

and

$$T_{i \rightarrow f}^{(N)} = \langle \lambda' | [E_1 - H] D^{-1} [E_2 - H] D^{-1} \cdots \times D^{-1} [E_{N-1} - H] | \lambda \rangle, \quad (6)$$

where D^{-1} is the inverse of the interaction operator. Using Eqs. (4)–(6), we may thus write the transition matrix element as

$$T_{i \rightarrow f}^{(N)} = \langle f | D | \lambda \rangle + \langle \lambda' | D | i \rangle - \langle \lambda' | [E_1 - H] D^{-1} \times [E_2 - H] D^{-1} \cdots D^{-1} [E_{N-1} - H] | \lambda \rangle. \quad (7)$$

This is a key equation since this special combination makes the transition matrix element variationally stable to second order in any deviations of λ and λ' from their exact values, λ_{ex} and λ'_{ex} [20]. That is, if $\lambda \equiv \lambda_{\text{ex}} + \delta\lambda$ and $\lambda' \equiv \lambda'_{\text{ex}} + \delta\lambda'$, it is easy to show that

$$T_{i \rightarrow f}^{(N)}(\lambda_{\text{ex}} + \delta\lambda, \lambda'_{\text{ex}} + \delta\lambda') = T_{i \rightarrow f}^{(N)}(\lambda_{\text{ex}}, \lambda'_{\text{ex}}) + O(\delta\lambda \delta\lambda'). \quad (8)$$

B. Coupled-channel hyperspherical approach

The use of hyperspherical coordinates for the treatment of the three-body problem has a long tradition in atomic and molecular physics [21,22,24–29,31,36], as well as in nuclear physics [37–42]. Applications to solid state physics have also been made [43–46]. The use of the hyperspherical formalism has many motivations, such as its universality, which permits its application to any few-body problem in an intuitive and elegant way. Another reason is that it provides a description of physical states in terms of potential curves and their couplings, which are independent of the energy, similarly to the Born-Oppenheimer [47] method. For a two-electron problem, such as that considered here for H^- , in which the nucleus may be regarded as infinitely massive, one rewrites the Schrödinger equation in terms of the new variables R and α , where $R = (r_1^2 + r_2^2)^{1/2}$ is the hyperspherical radius and $\alpha = \tan^{-1}(r_1/r_2)$ is the hyperangle, to obtain

$$\left(\frac{d^2}{dR^2} + \frac{U(R, \Omega) + 1/4}{R^2} + 2E \right) \Psi(R, \Omega) = 0. \quad (9)$$

The symbol Ω denotes the set $\{\alpha, \theta_1, \phi_1, \theta_2, \phi_2\}$ of all angular variables, where θ_i and ϕ_i ($i=1,2$) are the usual spherical coordinate angles for each of the electrons. The angular operator is given by

$$U(R, \Omega) = \frac{\partial^2}{\partial \alpha^2} - \frac{L_1^2}{\sin^2 \alpha} - \frac{L_2^2}{\cos^2 \alpha} + \frac{2ZR}{\sin \alpha} + \frac{2ZR}{\cos \alpha} - \frac{2R}{\sqrt{1 - \sin(2\alpha)\cos \theta_{12}}}. \quad (10)$$

In this equation, L_1^2 and L_2^2 are the usual angular momentum operators of the electrons and θ_{12} is the angle between their position vectors. The angular operator depends parametri-

cally on the hyperspherical radius R and is independent of the energy. By solving the eigenvalue problem for the angular operator [21],

$$U(R, \Omega) \Phi_{\mu}(R; \Omega) = U_{\mu}(R) \Phi_{\mu}(R; \Omega), \quad (11)$$

one obtains a set of angular functions $\Phi_{\mu}(R; \Omega)$, known as channel functions, for fixed values of R , taken as a parameter. One also obtains the set of corresponding potential curves $U_{\mu}(R)$, where the index μ is a collective label for all relevant quantum numbers. The total two-electron wave function is then expanded using the channel functions as basis [21],

$$\Psi(R, \Omega) = (R^{5/2} \sin \alpha \cos \alpha)^{-1} \sum_{\mu} F_{\mu}(R) \Phi_{\mu}(R; \Omega), \quad (12)$$

where the expansion coefficients at each R , $F_{\mu}(R)$, satisfy the coupled radial equations

$$\begin{aligned} \left(\frac{d^2}{dR^2} + \frac{U_{\mu}(R) + 1/4}{R^2} + 2E \right) F_{\mu}(R) \\ + \sum_{\nu} \left(2P_{\mu\nu}(R) \frac{d}{dR} + Q_{\mu\nu}(R) \right) F_{\nu}(R) = 0. \end{aligned} \quad (13)$$

In order to solve the coupled radial equations one must first calculate the nonadiabatic couplings, namely, $P_{\mu\nu}(R) = \langle \Phi_{\mu} | \partial / \partial R | \Phi_{\nu} \rangle$ and $Q_{\mu\nu}(R) = \langle \Phi_{\mu} | \partial^2 / \partial R^2 | \Phi_{\nu} \rangle$. Solving the coupled system of radial equations permits a controlled inclusion of electron correlation corrections via the nonadiabatic couplings, starting from the extreme adiabatic approximation (EAA), in which all couplings are neglected, up to the desired level of precision in a coupled adiabatic approximation (CAA), where all couplings are taken into account for a given number of channels [24,25]. The inclusion of only diagonal couplings furnishes an upper bound for the eigenvalue and is called the uncoupled adiabatic approximation (UAA). It has been demonstrated that the exact ground-state energy lies between the EAA value and the UAA and CAA values [48], where the following inequality holds for any nonrelativistic quantum problem:

$$E_{\text{EAA}} \leq E_{\text{exact}} \leq E_{\text{CAA}} \leq E_{\text{UAA}}. \quad (14)$$

This inequality is used for monitoring the convergence of the ground-state energy as increasing numbers of channels are taken into account, as seen in Table I.

C. Application to two-photon detachment of H^-

Our main interest in this paper is the two-photon detachment of H^- with linearly and circularly polarized light, where the final hydrogen atom is left in its ground state, i.e.,

$$2\gamma_{L,C} + \text{H}^- \rightarrow \text{H}(1s) + e^-. \quad (15)$$

TABLE I. Ground-state energy convergence of H^- as a function of the number N_c of coupled adiabatic hyperspherical channels in the radial equations. The first row corresponds to the calculation in which all couplings are neglected [EAA result, cf. Eq. (14)]; the second row corresponds to the one in which only the diagonal coupling matrix element was taken into account [UAA result, cf. Eq. (14)]. The variational result is that of Pekeris [12].

N_c	Energy (a.u.)	$(E_{\text{var}} - E) / E_{\text{var}}$ (ppm)
1	-0.536 904 592	-17 344.501
1	-0.526 032 797	3 255.735
2	-0.527 152 939	1 133.251
3	-0.527 703 141	90.712
4	-0.527 710 579	76.617
var.	-0.527 751 014	

For the two-photon case, the transition matrix element in Eq. (1) is written as

$$T_{i \rightarrow f}^{(N=2)}(\omega) = \langle f | D \frac{1}{E_i + \omega - H} D | i \rangle, \quad (16)$$

where $D = \boldsymbol{\epsilon} \cdot (\mathbf{r}_1 + \mathbf{r}_2)$ is the length form of the electric dipole operator, E_i is the energy of the initial state, $\boldsymbol{\epsilon}$ is the light polarization vector, and ω is the photon energy. The variationally stable form of Eq. (16), according to Eq. (7), is

$$T_{i \rightarrow f}^{(N=2)}(\omega) = \langle f | D | \lambda \rangle + \langle \lambda' | D | i \rangle - \langle \lambda' | E_i + \omega - H | \lambda \rangle, \quad (17)$$

where the Hamiltonian in hyperspherical coordinates is given by

$$H = -\frac{1}{2} \left(\frac{\partial^2}{\partial R^2} + \frac{U(R, \Omega) + 1/4}{R^2} \right). \quad (18)$$

At this point we expand the initial and final wave functions as well as the functions λ and λ' in adiabatic hyperspherical channel functions (12):

$$|i\rangle = (R^{5/2} \sin \alpha \cos \alpha)^{-1} \sum_{\mu_i} F_{\mu_i}(R) \Phi_{\mu_i}(R; \Omega), \quad (19)$$

$$|f\rangle = (R^{5/2} \sin \alpha \cos \alpha)^{-1} \sum_{\mu_f} F_{\mu_f}(R) \Phi_{\mu_f}(R; \Omega), \quad (20)$$

$$|\lambda\rangle = (R^{5/2} \sin \alpha \cos \alpha)^{-1} \sum_{\nu} \lambda_{\nu}(R) \Phi_{\nu}(R; \Omega), \quad (21)$$

$$|\lambda'\rangle = (R^{5/2} \sin \alpha \cos \alpha)^{-1} \sum_{\mu} \lambda'_{\mu}(R) \Phi_{\mu}(R; \Omega). \quad (22)$$

Using these functions, the three matrix elements in expression (17) take the forms

$$\langle f | D | \lambda \rangle = \sum_{\mu_f, \nu} \int_0^{\infty} I_{\mu_f \nu}^L(R) R F_{\mu_f}(R) \lambda_{\nu}(R) dR, \quad (23)$$

$$\langle \lambda' | D | i \rangle = \sum_{\mu, \mu_i} \int_0^\infty I_{\mu\mu_i}^L(R) R \lambda'_{\mu'}(R) F_{\mu_i}(R) dR, \quad (24)$$

and

$$\begin{aligned} \langle \lambda' | E_i + \omega - H | \lambda \rangle = & \sum_{\mu, \nu} \left\langle \lambda'_{\mu'} (E_i + \omega) \delta_{\mu\nu} \right. \\ & + \frac{1}{2} \left[\delta_{\mu\nu} \left(\frac{d^2}{dR^2} + \frac{U_\nu(R) + 1/4}{R^2} \right) \right. \\ & \left. \left. + 2P_{\mu\nu}(R) \frac{d}{dR} + Q_{\mu\nu}(R) \right] \right| \lambda_\nu \rangle, \end{aligned} \quad (25)$$

respectively, where $I_{\mu\mu'}^L(R)$ accounts for the electric dipole angular integrals between channel functions for channels μ and μ' in dipole length (L) approximation [49]. In order to evaluate the radial integrals, we expand the unknown radial functions as

$$\lambda_\nu(R) = \sum_j a_j^\nu \phi_j^\nu(R), \quad (26)$$

$$\lambda'_{\mu'}(R) = \sum_{j'} b_{j'}^\mu \theta_{j'}^\mu(R), \quad (27)$$

where $\phi_j^\nu(R)$ and $\theta_{j'}^\mu(R)$ are chosen to be Slater orbitals:

$$\phi_j^\nu(R) = N_j^\nu R^{m_\nu + 1/2 + j} e^{-\beta_\nu R}, \quad (28)$$

$$\theta_{j'}^\mu(R) = N_{j'}^\mu R^{m_\mu + 1/2 + j'} e^{-\beta_\mu R}, \quad (29)$$

where $m_\mu = \sqrt{-U_\mu(R=0)}$ (cf. Ref. [22]) and β_μ is an arbitrary positive constant, which may be complex. N_j^μ is a normalization constant for each of the basis functions, given by

$$\frac{1}{(N_j^\mu)^2} = \frac{\Gamma(2m_\mu + 2j + 2)}{(2\beta_\mu)^{2m_\mu + 2j + 2}}. \quad (30)$$

By requiring Eq. (17) to be variationally stable with respect to the coefficients a_j^ν and $b_{j'}^\mu$, that is,

$$\frac{\partial T_{i \rightarrow f}^{(N=2)}}{\partial a_j^\nu} = \frac{\partial T_{i \rightarrow f}^{(N=2)}}{\partial b_{j'}^\mu} = 0, \quad (31)$$

we finally obtain two linear systems of equations, which permit the calculation of the unknown coefficients a_j^ν and $b_{j'}^\mu$, i.e.,

$$\sum_\nu \sum_j A_{j'j}^{\mu\nu} a_j^\nu = c_{j'}^\mu, \quad (32)$$

$$\sum_\mu \sum_{j'} b_{j'}^\mu A_{j'j}^{\mu\nu} = d_j^\nu, \quad (33)$$

where

$$c_{j'}^\mu = N_{j'}^\mu \sum_{\mu_i} \int_0^\infty I_{\mu\mu_i}^L(R) R^{m_\mu + 1/2 + j' + 1} F_{\mu_i}(R) e^{-\beta_\mu R} dR, \quad (34)$$

$$d_j^\nu = N_j^\nu \sum_{\mu_f} \int_0^\infty I_{\mu_f\nu}^L(R) R^{m_\nu + 1/2 + j + 1} F_{\mu_f}(R) e^{-\beta_\nu R} dR, \quad (35)$$

$$A_{j'j}^{\mu\nu} = N_{j'}^\mu N_j^\nu \left[J_{j'j}^{\mu\nu} \delta_{\mu\nu} + \frac{1}{2} K_{j'j}^{\mu\nu} (1 - \delta_{\mu\nu}) \right], \quad (36)$$

and where $J_{j'j}^{\mu\nu}$ and $K_{j'j}^{\mu\nu}$ are, respectively, the integrals for the diagonal and nondiagonal terms in Eq. (36),

$$\begin{aligned} J_{j'j}^{\mu\nu} = & \frac{1}{2} [2(E_i + \omega) + \beta_\mu^2] \frac{\bar{l}!}{(2\beta_\mu)^{\bar{l}+1}} \\ & - \left(\frac{m_\nu + 1/2 + j}{2} \right) \frac{(\bar{l}-1)!}{(2\beta_\mu)^{\bar{l}-1}} \\ & + \frac{1}{2} (m_\nu + 1/2 + j)(m_\nu + 1/2 + j - 1) \frac{(\bar{l}-2)!}{(2\beta_\mu)^{\bar{l}-1}} \\ & + \frac{1}{2} \int_0^\infty R^{\bar{l}} e^{-2\beta_\mu R} \left(\frac{U_\mu(R) + 1/4}{R^2} + Q_{\mu\mu}(R) \right) dR \end{aligned} \quad (37)$$

and

$$\begin{aligned} K_{j'j}^{\mu\nu} = & \int_0^\infty R^{\bar{l}} e^{-(\beta_\nu + \beta_\mu)R} \\ & \times \left[2P_{\mu\nu}(R) \left(\frac{m_\nu + 1/2 + j}{R} - \beta_\nu \right) + Q_{\mu\nu}(R) \right] dR, \end{aligned} \quad (38)$$

where $\bar{l} = m_\mu + m_\nu + j + j' + 1$.

The total generalized two-photon cross section [50] is then calculated according to

$$\sigma^{(N=2)} = 8\pi^3 \alpha^2 \omega^2 |T_{i \rightarrow f}^{(N=2)}|^2, \quad (39)$$

where α is the fine structure constant. To convert the cross section we obtain in atomic units to $\text{cm}^4 \text{s}$ units, one multiplies Eq. (39) by the numerical conversion factor $[L]^4 [T] = 1.896791616 \times 10^{-50}$.

III. RESULTS

We present in this section our results for the single-photon detachment cross section of H^- , for the dynamic polarizability of the H^- ground state, and for the generalized two-photon detachment cross section of H^- by linearly and circularly polarized light. First, however, we discuss briefly some general computational aspects. All calculations have

been done using the length gauge. (The velocity gauge matrix elements within a hyperspherical coordinate basis are much more complicated to evaluate numerically, as may be seen, e.g., from Appendix A of Ref. [49].) Atomic units are used throughout the paper. Where conversion between units was necessary, we have used $a_0 = 0.529\,177 \times 10^{-8}$ cm for the Bohr radius, $t_0 = 2.418\,884 \times 10^{-17}$ s for the atomic unit of time, and $\alpha = 1/137.0360$ for the fine structure constant. In each numerical calculation, we have used in our codes REAL*16 (quadruple) precision to minimize numerical error propagation and to deal accurately with large numbers. The potential curves $U_\mu(R)$ and the channel functions $\Phi_\mu(R; \Omega)$ have been obtained by a power series expansion in a new variable $x = \tan(\alpha/2)$. This change of variable has been suggested by a careful analysis of the topological properties of the angular equation, discussed in detail in Ref. [22]. It enables one to generate numerically accurate solutions. Also we have used for the individual angular momenta the maximum value $l_1^{\max} = l_2^{\max} = 9$ for $^1S^e$ and $^1P^o$ states and $l_1^{\max} = l_2^{\max} = 6$ for $^1D^e$ states. (Note that if we were to use $l_1^{\max} = l_2^{\max} = 6$ for all states, our results would be essentially unchanged; we did not use $l_1^{\max} = l_2^{\max} = 9$ for the $^1D^e$ states, therefore, in order not to increase the computation time further.) In the calculation of the initial and final hyperradial wave functions $F_\mu(R)$ we have used up to $N_c = 4$ coupled equations. Table I lists the calculated energies for the initial ground state for $N_c = 1, \dots, 4$. The first row corresponds to the calculation in which all nonadiabatic couplings were neglected [EAA result, cf. Eq. (14)]. The second row is also an uncoupled calculation, but in which the diagonal matrix element coupling was included [UAA result, cf. Eq. (14)]. This table shows the difference in parts per million (ppm) between our results and the variationally calculated value of Pekeris [12], listed in the last row. By reducing the difference between our *ab initio* prediction for the electron affinity and the accurate value obtained by Pekeris [12] to about 0.15%, we have reduced this source of error in the two-photon cross section to below 1% for the case of linearly polarized light [8].

A. One-photon detachment cross section

For the one-photon detachment cross section, the transition matrix element (1) is reduced to the simplest form,

$$T_{i \rightarrow f}^{(N=1)} = \langle f | D | i \rangle. \quad (40)$$

This calculation is a good test for the *continuum* wave functions since there are reliable calculations in the literature for comparison, including those that employ a hyperspherical approach. We first performed our calculation using the quasiseparable approximation ($N_c = 1$) and then successively coupled two, three, and four radial equations. In Fig. 1 we show the one-photon cross section as a function of the kinetic energy of the detached electron for $N_c = 1, \dots, 4$. We also show the variational values of Stewart [30] for comparison. Note that there are a large number of accurate results for photodetachment of H^- below the $n = 2$ excitation threshold [29–31, 51–55]. Most of these agree within about 5%, as

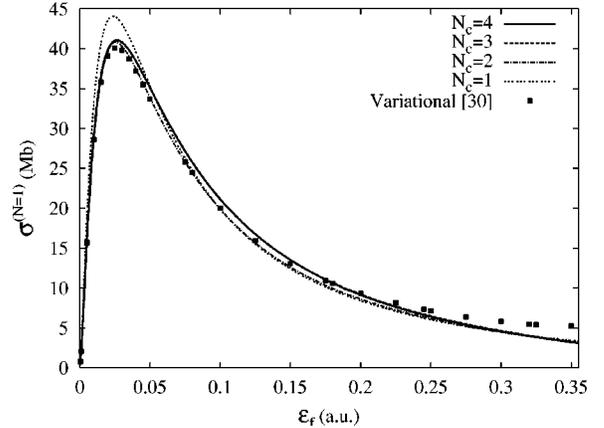


FIG. 1. One-photon detachment cross section of H^- using linearly polarized light as a function of the photoelectron kinetic energy ϵ_f . N_c is the number of coupled channels within each term level of the initial, intermediate, and final states. Also shown are the variational results from Ref. [30].

shown in Table VI of Ref. [31]. As our purpose in computing the single-photon detachment cross section of H^- is simply to verify and demonstrate the validity of using four coupled channels, we compare here only with the variational results of Stewart [30]. We notice that with only four coupled channels we have achieved very good agreement in general with the variational results of Stewart [30], particularly in the photon energy region from threshold up to ≈ 0.15 a.u. (≈ 4.1 eV). We also see that the results with $N_c = 3$ and $N_c = 4$ are very close together, indicating that our results have good convergence as a function of the number N_c of coupled channels. Above $\hbar\omega \approx 0.15$ a.u. Stewart's values become higher than ours, indicating a possible need for a greater number of coupled channels in this energy region. However, as this energy region is much higher than the one with which we are concerned in calculating the two-photon detachment cross section [i.e., $\hbar\omega < 0.0277\,51$ a.u. (0.755 eV)], and as the single-photon cross section is small in this energy region, we can conclude that using $N_c = 4$ coupled channels is sufficient to obtain accurate two-photon cross sections.

B. Dynamic polarizability

The dynamic polarizability $\alpha(\omega)$ is a two-photon process that involves only the ground-state wave function. This calculation is useful not only for testing the variational procedure itself but also for assuring the quality of the ground-state wave function, since it is well known that a good calculated energy, as in Table I, is not a guarantee of an equally good wave function. In order to obtain $\alpha(\omega)$, one substitutes for the final state $|f\rangle$ the initial state $|i\rangle$ in Eq. (16). Thus, the frequency-dependent polarizability is calculated as follows:

$$\alpha(\omega) = -[T_{i \rightarrow i}^{(N=2)}(+\omega) + T_{i \rightarrow i}^{(N=2)}(-\omega)]. \quad (41)$$

In the numerical calculation, we have chosen $\beta_\mu = 0.65$ and 60 basis functions to achieve very good convergence. Figure 2 shows the dynamic polarizability α of H^- as a function of

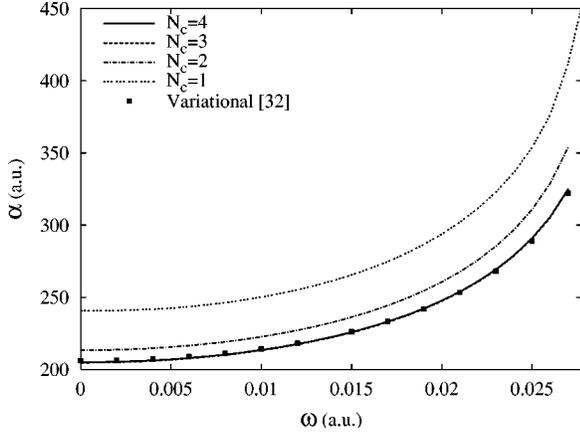


FIG. 2. Dynamic polarizability of H^- as a function of the photon energy. N_c is the number of coupled channels within each term level of the initial and intermediate states. The curve labeled $N_c = 1$ is the uncoupled-channel result. Also shown are the variational results from Ref. [32].

the photon energy, for $1 \leq N_c \leq 4$ coupled adiabatic hyperspherical channels within each term level, compared with the variation-perturbation results of Chung [32]. The three- and four-coupled-channel calculations give converged results and the agreement with the variational values is excellent. Our best value for the static polarizability ($\omega=0$) is $\alpha(0) = 205.014\,509$, which compares with the variational value of 206.0 [32]. For the uncoupled calculation ($N_c=1$), we also included a semiempirical shift of the electron affinity to the Pekeris [12] value of 0.027751014 a.u. The result can be seen in Fig. 3, where we compare our best, four-coupled-channel calculation for the dynamic polarizability of H^- with the one-channel shifted result, which is labeled $N_c = 1$ (adj). [Note that this shift is produced by a very slight *ad hoc* lowering of the minimum of the $\mu=1$ $^1S^e$ hyperspherical

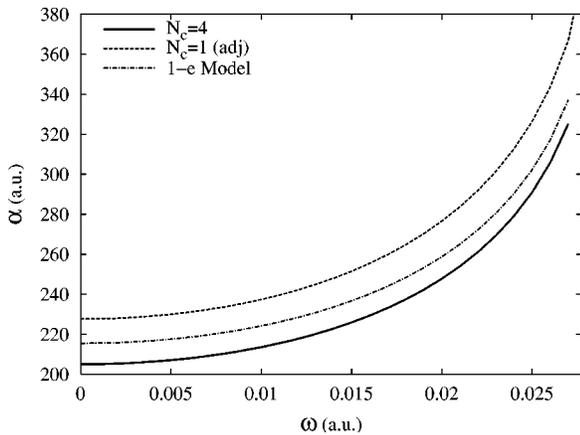


FIG. 3. Dynamic polarizability of H^- as a function of the photon energy. N_c is the number of coupled channels within each term level of the initial and intermediate states. The curve labeled $N_c = 1$ (adj) is the result employing a semiempirical adjustment of the lowest $^1S^e$ adiabatic hyperspherical potential so that the predicted electron affinity agrees with the variational result of Pekeris [12]. Also shown is the result using a single-electron (1- e), zero-range potential model.

potential $-U_\mu(R)/R^2$.] Also shown is the result obtained using a one-electron, short-range potential model, in which the electronic states have the momentum-space wave function,

$$\psi_i(\mathbf{k}) = 2^{3/2} B \frac{Y_{00}(\hat{\mathbf{k}})}{k^2 - 2E_i}, \quad (42)$$

where $B=0.315\,52$ [56] is a normalization constant. This form of the initial-state wave function represents the solution of an attractive spherical δ -function potential [57]. Using this same model, Adelman [58] calculated the frequency-dependent polarizability. Our one-electron, short-range potential model results are in agreement with his. As pointed out by Glover and Weinhold [34], the results of Chung [32] are in good agreement with their rigorous lower and upper limits at all frequencies, but the results provided by the one-electron model of Adelman [58] fail to lie within these limits, especially at longer wavelengths. Our coupled-channel calculation, as seen in Fig. 2, is in good agreement with the bounds of Glover and Weinhold [34], thereby indicating again that our variationally stable, coupled-channel approach with four coupled channels is reliable over the energy region below the single-photon threshold. Table II presents a detailed comparison of our results with those of Refs. [32–34].

C. Two-photon detachment cross section

For the two-photon detachment process the initial state is $^1S^e$ and the intermediate states have symmetry $^1P^o$. For linearly polarized light the final states can be either $^1S^e$ or $^1D^e$, while for circularly polarized light only $^1D^e$ final states are allowed. The calculation of the two-photon detachment cross section using the method described in Sec. II A proved to be very stable and in all calculations excellent convergence was achieved. The three matrix elements given by Eqs. (4), (5), and (6), which are combined in Eq. (7), are equal (when convergence is achieved) to within at least 20 digits in all our calculations. [Note nevertheless that our Eq. (31), which determines the coefficients in Eqs. (26) and (27) of the unknown functions λ_ν and λ'_μ , can be solved only if each of the different forms for the transition matrix element is included in Eq. (17).] We have used a single value for the arbitrary constant β_μ [cf. Eqs. (28) and (29)] since this is sufficient for the two-photon case. We choose $\beta_\mu=0.45$ for the one-channel calculation and $\beta_\mu=0.85$ for each of the three coupled-channel calculations ($N_c=2,3,4$). We also used 70 basis functions for each ν and μ in the expansions (26) and (27), since this was found to provide a convergence in the summations to five digits in the worst case, and to seven or eight digits in general. The calculation was done in the energy region where a single photon is not sufficient to ionize the system. Above the one-photon ionization threshold it is necessary to make a complex rotation or to choose a complex β_μ [59].

Figure 4 shows the results for the generalized two-photon detachment cross section of H^- as a function of the photoelectron energy. The cross section is shown for $1 \leq N_c \leq 4$ coupled adiabatic hyperspherical channels within each term

TABLE II. Comparison of the present ($N_c=4$) variationally stable dynamic polarizability results (in a.u.) for the ground state of H^- as a function of the photon frequency ω (in a.u.) with results of Chung [32], Pipin and Bishop [33], and Glover and Weinhold [34].

ω (a.u.)	Present	Chung [32]	% Diff. ^a	Pipin	% Diff. ^a	Glover and Weinhold [34]	
	results ($N_c=4$)			and Bishop [33]		Lower bound	Upper bound
0.000	205.0145	206.0	-0.478	206.165	-0.558	203.94	208.84
0.010	205.0947			206.245	-0.557	204.02	208.94
0.020	205.3360	206.3	-0.467	206.486	-0.556	204.25	209.21
0.030	205.7399			206.889	-0.555	204.65	209.65
0.040	206.3095	207.3	-0.477	207.458	-0.553	205.20	210.28
0.050	207.0488			208.197	-0.551	205.92	211.10
0.060	207.9633	208.9	-0.448	209.110	-0.548	205.82	211.12
0.070	209.0598			210.205	-0.544	207.88	213.36
0.080	210.3468	211.2	-0.403	211.491	-0.541	209.14	214.82
0.090	211.8348			212.976	-0.535	210.56	216.50
0.010	213.5362	214.2	-0.309	214.675	-0.530	212.24	218.44
0.011	215.4659			216.602	-0.524	214.12	220.68
0.012	217.6419	218.2	-0.255	218.774	-0.517	216.24	223.22
0.013	220.0854			221.213	-0.509	218.61	226.11
0.014	222.8222			223.944	-0.500	221.27	229.41
0.015	225.8833	226.2	-0.140	226.998	-0.491	224.23	233.15
0.016	229.3064			230.412	-0.479	227.55	237.47
0.017	233.1378	233.2	-0.026			231.25	242.43
0.018	237.4350					235.40	248.22
0.019	242.2707	242.0	0.111			240.06	255.04
0.020	247.7379					245.31	263.23
0.021	253.9592	253.3	0.260			251.27	273.33
0.022	261.0999					258.07	286.19
0.023	269.3920	268.1	0.481			265.93	303.39
0.024	279.1756					275.12	328.54
0.025	290.9832	288.9	0.721			336.07	418.67
0.026	305.7242					249.45	400.77
0.027	325.1676	322.1	0.952			316.39	739.37

^a% Diff. is the percentage difference of the present results from those of Refs. [32,33].

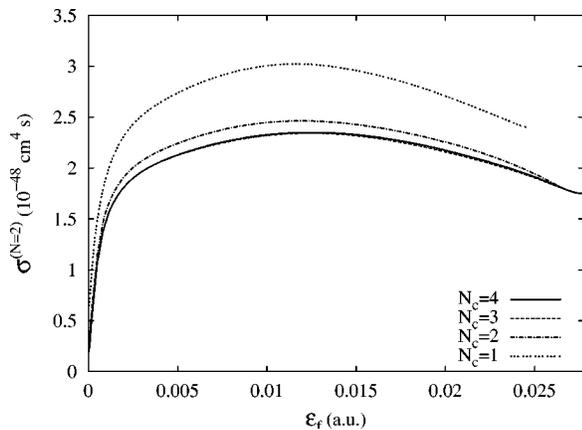


FIG. 4. Generalized two-photon detachment cross section of H^- using linearly polarized light as a function of the photoelectron kinetic energy ϵ_f . N_c is the number of coupled channels within each term level of the initial, intermediate, and final states.

level of the initial, intermediate, and final states. Once more, with only four coupled channels we have already achieved good convergence for the total two-photon detachment cross section, since the $N_c=3$ and $N_c=4$ results are almost indistinguishable from each other. Higher channels $N_c \geq 5$ involve excitations converging to the $H(n)$ threshold, with $n \geq 3$ for $^1S^e$ and $^1P^o$ and $n \geq 2$ for $^1D^e$. The greater radial extent of each of the potentials $-U_\mu(R)/R^2$ for these neglected channels compared to the ones that have been included is expected to make their contributions of minor importance in the energy region of interest for the two-photon detachment cross section.

In Fig. 5 we present a comparison between our best ($N_c=4$) coupled-channel calculation and results of others. We compare with the variationally stable, uncoupled adiabatic hyperspherical channel result of Liu *et al.* [8] in which they semiempirically shifted the electron affinity. The same type of adjustment was utilized by Dörr *et al.* [11], whose R -matrix Floquet results are also shown in Fig. 5. These two results [8,11] lie 8–10% higher than our present results in

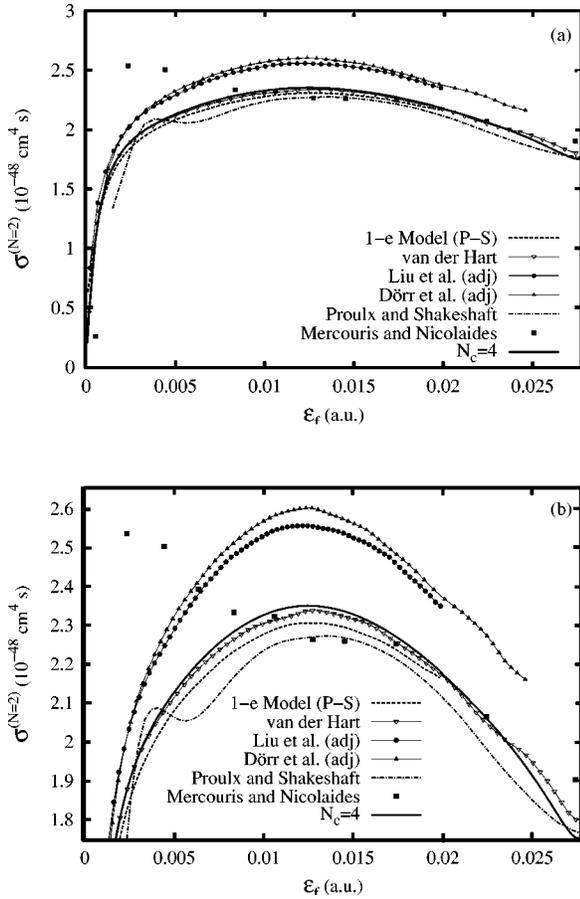


FIG. 5. (a) Generalized two-photon detachment cross section of H^- for the case of linearly polarized light as a function of the photoelectron kinetic energy ϵ_f . Solid curve: present ($N_c=4$) coupled-channel, variationally stable result; solid squares: MEMPT result of Mercouris and Nicolaides [3(d)]; dash-dotted line: complex Sturmian result of Proulx and Shakeshaft [7]; solid circles: variationally stable, uncoupled-channel result of Liu *et al.* [8] with adjusted electron affinity; dashed line: phase-shifted (P-S) single-electron, short-range potential model result of Liu *et al.* [8]; open triangles: B -spline result of van der Hart [10]; solid triangles: R -matrix Floquet result of Dörr *et al.* [11] with adjusted electron affinity. (b) Same as in (a) but on an expanded scale.

the energy range of the cross section plateau. For photon energies $\hbar\omega \geq 0.0175$ a.u. (≈ 0.48 eV) our results are in excellent agreement with the many-electron, many-photon theory (MEMPT) results of Mercouris and Nicolaides [3(d)], the complex Sturmian function results of Proulx and Shakeshaft [7], and the B -spline results of van der Hart [10]. However, for $\hbar\omega < 0.0175$ a.u., our results agree only with those of van der Hart [10]. Figure 5(b) shows our results and all of the other results discussed above on an expanded scale.

Finally, in Fig. 5 we have included the zero-range potential model prediction of Liu *et al.* [8] in which the s -wave phase shift is taken into account. Geltman [6] has shown numerically that inclusion of the s -wave phase shift can significantly improve the two-photon detachment cross section for the case of linearly polarized light. Liu *et al.* [8] formulated the problem analytically and also used the more accurate normalization of the initial-state wave function stem-

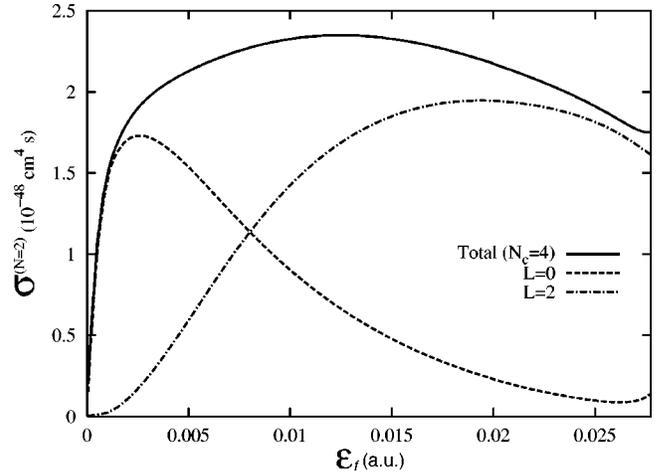


FIG. 6. Contributions to the total two-photon detachment generalized cross section from the $L=0$ and $L=2$ partial waves with four coupled channels.

ming from effective-range theory. One sees that the phase-shifted zero-range potential model results are in excellent agreement with our *ab initio*, $N_c=4$ coupled-channel result as well as with the B -spline results of van der Hart [10].

In Fig. 6 we show the contributions to the total two-photon detachment cross section from the $L=0$ and $L=2$ partial waves within the four-coupled-channels calculation. The cross section for detachment to final $^1S^e$ states is dominant for small values of the photoelectron energy, but for higher energies the final $^1D^e$ states become increasingly important and eventually become dominant, while the final $^1S^e$ states become increasingly less important.

For the calculation using circularly polarized light we have used the same set of parameters as in the linearly polarized light calculation. Figure 7 shows the convergence of the generalized two-photon cross section as we systematically increase the coupling between the radial components. As for the linearly polarized calculation, the results are very stable, with high precision and accuracy. In Fig. 8 we show a comparison between our four-coupled-channel result and the

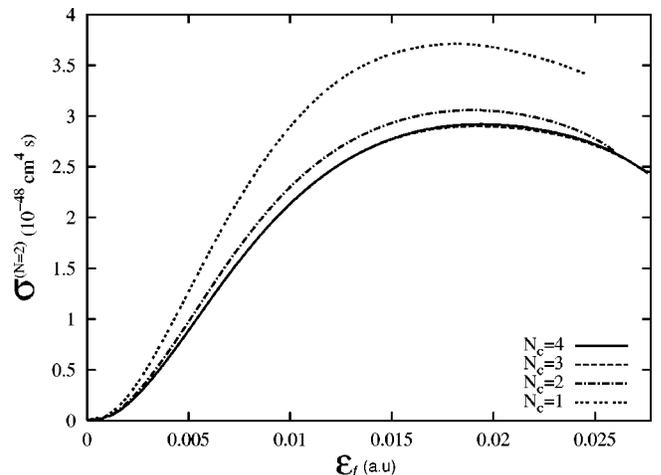


FIG. 7. Same as in Fig. 4 for the case of circularly polarized light.

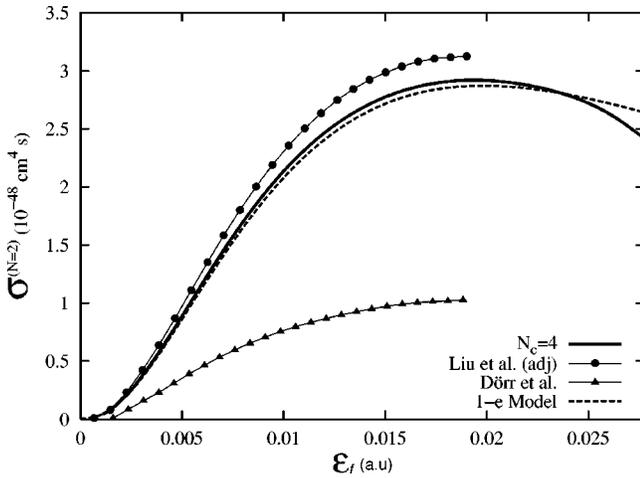


FIG. 8. Generalized two-photon detachment cross section of H^- for the case of circularly polarized light as a function of the photoelectron kinetic energy ϵ_f . Solid curve: present ($N_c=4$) coupled-channel, variationally stable result; solid triangles: two-electron Keldysh theory result of Dörr *et al.* [5]; solid circles: variationally stable, uncoupled-channel result of Liu *et al.* [8] with adjusted electron affinity; dashed line: single-electron, short-range potential model result of Liu *et al.* [8].

variationally stable, semiempirically shifted, single-channel result of Liu *et al.* [8], the “two-electron Keldysh theory” result of Dörr *et al.* [5], and the zero-range potential result of Liu *et al.* [8]. Remarkably, our variationally stable, $N_c=4$ coupled-channel result is in best agreement with the zero-range potential model result [8]. This fact implies that in Fig. 5 for the case of linearly polarized photons it is the $^1S^e$ partial wave (whose contribution is shown in Fig. 6) that is primarily responsible for the differences between the results of various authors.

IV. DISCUSSION AND CONCLUSIONS

We have presented variationally stable, coupled adiabatic hyperspherical channel results for both the dynamic polarizability and the two-photon detachment cross section of H^- by linearly and circularly polarized light for photon energies up to the single-photon detachment threshold. We have also presented results of a coupled adiabatic hyperspherical channel calculation for the single-photon detachment cross section for photon energies below the $H(n=2)$ excitation threshold. As compared with results of Liu *et al.* [8], the advance in this work is the calculation of all quantities beyond the adiabatic (quasiseparable) approximation without

any sort of semiempirical adjustment. By employing an *ab initio*, coupled-channel approach we are able to obtain very accurate cross sections for one- and two-photon detachment and for the dynamic polarizability. Our treatment for the latter two physical quantities employed the variationally stable procedure of Refs. [19,20,23,8] in order to minimize the effects of wave function errors on the observables calculated. Because our dynamic polarizability and two-photon detachment cross section results were obtained in a variationally stable way, we have presented the results in detail. However, the main goal of this work has been to provide accurate two-photon detachment cross section results; our dynamic polarizability and single-photon detachment results were carried out solely to test the various components of our approach for processes for which some consensus among prior theoretical results has already been achieved.

Our variationally stable, $N_c=4$ coupled adiabatic hyperspherical channel results for two-photon detachment of H^- for the case of linearly polarized light are in excellent agreement with the correlated B -spline results of van der Hart [10] over the entire energy region between the two-photon detachment threshold and the single-photon detachment threshold. That two such independent *ab initio* calculations agree so well indicates that a theoretical consensus on the two-photon detachment cross section of H^- has at last been achieved, whereas there has been no such consensus before among *ab initio* calculations. That these two theoretical results agree well also with an approximate zero-range potential model result (i.e., one in which the *continuum* state s -wave phase shift is taken into account and also an appropriate normalization is used for the initial-state wave function) indicates that in this energy range electron-correlation effects are of short range. We expect that this agreement with the phase-shifted, short-range potential model predictions may no longer hold at higher photon energies near the $H(n=2)$ excitation threshold. Extension of the present approach to this higher energy region is possible, but will require use of complex β coefficients in the Slater-type basis functions or else the use of complex rotation techniques, since the intermediate state functions λ have oscillating components above the single-photon detachment threshold.

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