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One- and two-photon ionization of model atoms: The spherical δ-shell potential

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One- and two-photon ionization cross sections for the negative hydrogen ion H^- are evaluated by using a one-electron model where the effective intra-atomic potential energy is assumed to be of a very-short-range character and approximated by a "spherical δ shell." The adjustment of the two parameters involved in the model potential so as to reproduce either the photodetachment energy or the behavior of the one-photon ionization cross section over a substantial range of frequencies beyond the ionization threshold leads to conjecture of a reasonable estimate of the two-photon ionization cross section over the proper range of frequencies of the ionizing field.

I. INTRODUCTION

The advent in the last two decades of intense laser sources has determined the development of a broad variety of processes involving absorption and/or emission of several photons in atoms and molecules and stimulated the consequent demand of an adequate, interpretative, theoretical framework. On the latter side, we are nowadays in a position to assert that there is noticeable evidence in favor of the use of quantum-mechanical perturbative techniques even in the case of systems subjected to rather strong electromagnetic fields;[1] so, for example, we are convinced that perturbation theory at Nth order is adequate for dealing with problems which involve the absorption of $N>1$ light quanta, provided that we are far away from intermediate resonance conditions corresponding to the absorption of a number $n<N$ of photons. Even though simplified conditions like these are met, however, the task of correctly evaluating the quantum-mechanical transition amplitude $T^{[N]}_{f\leftarrow i}$ for a N-photon process is a formidable one for any realistic situation, which cannot but require approximations at various stages; since the approximations introduced are seldom of a completely assessable nature, the final result may reflect the concurrence of interferential pseudoeffects, with hardly predictable exits.[2]

A partial overcoming of the difficulties posed by the complex nature of the multiphoton phenomena is through the development of (usually schematic) models. To remain as adherent as possible to the contents of this paper, we stress the case of atomic and molecular anions, a class of fairly interesting systems which have attracted noticeable attention either at the level of *ab initio* calculations[3] (usually requiring a considerable burden of computational effort) or through an approach founded on the study of appropriate models[4] able to mimic relevant characteristics of such a class of systems. We take sides with the second attitude, our aim being addressed to investigate the reliability of some one-electron models in predicting the multiphoton ionization cross section of atomic anions, as complicating effects related to the target excitation are absent. The interaction between optical electron and neutral inner shell, which is assumed to be of very-short-range character, is modeled by an appropriate effective potential energy. While in some previous publications the representation of such interaction has been mimicked in terms of a Fermi pseudopotential,[5] in the present paper we propose to study a second schematic model potential, which in the literature is frequently referred to as the "spherical δ-shell potential."[6] Such a model is interesting for several reasons: Firstly, it provides us with a simple picture of a short-range potential, endowed with two independent, adjustable parameters (see Secs. II and III); secondly, an explicit (and simple) expression for the quantum-mechanical propagator (in the energy domain) is within our reach; and, thirdly, multiphoton ionization cross sections can be derived in an entirely analytical form, as a consequence of the simplicity of the spectral properties of the Hamiltonian model, which is gratifying in view of drawing conclusions or stressing remarks of behavior.

The plan of the present paper is as follows: The next section is devoted to a short survey of the theoretical problems posed by the study of one- and two-photon ionization processes, along the lines of a procedure recently suggested by two of the present authors (GPA, CG),[5] which avails itself of a reformulation of the relevant cross-section expressions in terms of Fourier transforms of proper time-dependent correlation functions; in Sec. III we move to specifically examine the "spherical δ-shell" model and discuss the results which descend from its use.

II. THEORETICAL APPROACH TO THE EVALUATION OF PHOTOIONIZATION CROSS SECTIONS

In the so-called "length" form,[5,7] ionization cross sections corresponding to the absorption of one and two photons from a linearly polarized radiation field of frequency ω are respectively expressed as follows (atomic units with $e=m_e=\hbar=1$ are used throughout this paper):

$$\sigma^{[1]}(\omega)=(4\pi^2\omega/c)\int_0^\infty dE_f\delta(E_f-E_b-\omega)\,|\langle E_f^-10\,|z\,|E_b00\rangle|^2\,, \tag{1}$$

$$\sigma^{[2]}(\omega)=(8\pi^3\omega^2/c^2)\sum_{L=0,2}\int_0^\infty dE_f\delta(E_f-E_b-2\omega)\,|\langle E_f^-L0\,|zG^+(E_b+\omega)z\,|E_b00\rangle|^2\,, \tag{2}$$

$|E_f^-L0\rangle$ denotes the (energy-normalized) final state, with incoming-wave boundary conditions, appropriate to the channel of angular momentum $(L,M_L=0)$, while $|E_b00\rangle$ represents the spherically symmetric ground state of the atom. $G^+(E)\equiv[H-(E+i0^+)]^{-1}$, with $E=E_b+\omega$, is the propagator in the energy domain, i.e., the Green's operator of the (unperturbed) atom.

It is rather straightforward to cast $\sigma^{[1]}$ into an equivalent, alternative form, which exhibits it as the *expectation value* of an appropriate operator. From the completeness of the set of states supported by the model, we find

$$\sigma^{[1]}(\omega)=(4\pi^2\omega/c)\langle E_b00\,|z\delta[H-(E_b+\omega)]z\,|E_b00\rangle\,. \tag{3}$$

An entirely analogous expression is obtained for $\sigma^{[2]}$:

$$\sigma^{[2]}(\omega)=(8\pi^3\omega^2/c^2)\times\langle\Phi(\omega)\,|z\delta[H-(E_b+2\omega)]z\,|\Phi(\omega)\rangle\,, \tag{4}$$

where we have defined the state ket

$$|\Phi(\omega)\rangle\equiv G^+(E_b+\omega)z\,|E_b00\rangle\,. \tag{5}$$

The operator $\delta(H\text{-}E)$ figuring in Eqs. (3) and (5) may be expressed in several useful ways. From the standard integral representation

$$2\pi\delta(H\text{-}E)=\int_{-\infty}^{+\infty}dt\,\exp[i(H\text{-}E)t]$$

it is quite simple to cast the expressions for $\sigma^{[1]}$ and $\sigma^{[2]}$ into the following forms:

$$\sigma^{[1]}(\omega)=(2\pi\omega/c)\int_{-\infty}^{+\infty}dt\,e^{i\omega t}\langle E_b00\,|z(t)z(0)\,|E_b00\rangle\,, \tag{6}$$

$$\sigma^{[2]}(\omega)=(2\pi\omega/c)^2\int_{-\infty}^{+\infty}dt\,e^{2i\omega t}\langle E_b00\,|\xi_z(t)\xi_z(0)\,|E_b00\rangle\,, \tag{7}$$

respectively. In Eqs. (6) and (7) we have set

$$z(t)=e^{iHt}z(0)e^{-iHt},\quad \xi_z(t)=e^{iHt}\xi_z(0)e^{-iHt}\,, \tag{8}$$

where

$$z(0)\equiv z,\quad \xi_z(0)\equiv zG^+(E_b+\omega)z\,. \tag{9}$$

Equations (6) and (7) display the photoionization cross sections $\sigma^{[1]}$ and $\sigma^{[2]}$ as Fourier transforms of appropriate time-dependent autocorrelation functions.[5] Other equivalent expressions stem easily from Eqs. (6) and (7). So, for example, $\sigma^{[1]}$ can be rewritten as

$$\sigma^{[1]}(\omega)=(2\pi\omega/c)\int_{-\infty}^{+\infty}dt\,e^{i(E_b+\omega)t}\times\langle E_b00\,|ze^{-iHt}z\,|E_b00\rangle\,. \tag{6'}$$

From this point of view $\sigma^{[1]}$ is then essentially the Fourier transform (at the energy $E_b+\omega$) of the overlap between the initial "doorway" state[8] $z\,|E_b00\rangle$ and its temporal evolution $e^{-iHt}z\,|E_b00\rangle$ at t time as dynamically induced by the unperturbed Hamiltonian operator of the atom. $\sigma^{[2]}$ is clearly susceptible to an analogous interpretation, even though in terms of a different (more complicated) doorway state. The results expressed by Eqs. (6), (6′), and (7) are formally similar to analogous expressions recently put forward for photodissociation cross sections of molecular systems.[9]

If viable procedures for calculating time-dependent correlation functions can be set up, the approach just suggested is likely to acquire some computational perspective in photoionization problems; by it, for instance, we could hope to bypass the necessity of explicitly disposing of the continuum final states of the system.

It is possible to carry out the Fourier transform required by the previous results and pass from time to energy domain. By the usual trick

$$\int_{-\infty}^{+\infty}dt\,e^{i\Omega t}=\lim_{\eta\to0^+}\left[\int_0^\infty dt\,\exp[i(\Omega+i\eta)t]+\text{c.c.}\right]\,,$$

we attain[5]

$$\sigma^{[1]}(\omega)=(4\pi\omega/c)\langle E_b00\,|z[\operatorname{Im}G^+(E_b+\omega)]z\,|E_b00\rangle\,,$$
$$\sigma^{[2]}(\omega)=\tfrac{1}{2}(4\pi\omega/c)^2\times\langle\Phi(\omega)\,|z[\operatorname{Im}G^+(E_b+2\omega)]z\,|\Phi(\omega)\rangle\,, \tag{10}$$

the forms actually used in the present paper.

III. APPLICATION TO THE SPHERICAL δ-SHELL MODEL

For a single (nonrelativistic) electron, the Hamiltonian operator H of the model system is[6]

$$H=\tfrac{1}{2}\nabla^2-V\delta(r-R)\,, \tag{11}$$

where V,R are the two (positive) parameters which characterize the (intra-atomic) interaction potential energy of the spherical shell. The Schrödinger equation associated with the Hamiltonian operator H,

$$H\,|\,ELM\rangle = E\,|\,ELM\rangle\ ,$$

is easily solved for the $L=0$ bound states supported by the model. One finds that, provided $VR>\frac{1}{2}$, there exists one (and only one) spherically symmetric bound state (the ground state),

$$\begin{aligned} u_b(\mathbf{r}) &\equiv \langle \mathbf{r}\,|\,E_b 00\rangle \\ &= N Y_{00}(\hat{\mathbf{r}})[\Theta(R-r)(e^{\gamma r}-e^{-\gamma r}) \\ &\qquad + \Theta(r-R)(e^{2\gamma R}-1)e^{-\gamma r}]/r\ , \\ N &= [\gamma/(e^{2\gamma R}-2\gamma R-1)]^{1/2}\ , \end{aligned} \tag{12}$$

$\Theta(x)$ being the Heaviside step function. The associated eigenvalue, of energy $E_b=-\gamma^2/2$, is determined by the (real) solution of the transcendental equation

$$\gamma/V = 1-e^{-2\gamma R}\ . \tag{13}$$

The propagator $G^+(E)\equiv[H-(E+i0^+)]^{-1}$ in the energy representation is also obtainable in an easy way for the model on issue. One finds (for $E>0$)

$$\begin{aligned} G^+(\mathbf{r},\mathbf{r}';E) &\equiv \langle \mathbf{r}\,|\,G^+(E)\,|\,\mathbf{r}'\rangle \\ &= \sum_{L,M} g_L^+(r,r';E) Y_{LM}(\hat{\mathbf{r}}) Y_{LM}^*(\hat{\mathbf{r}}')\ , \end{aligned} \tag{14}$$

with

$$\begin{aligned} g_L^+(r,r';E) &= g_L^{0+}(r,r';E) \\ &\quad + \frac{VR^2 g_L^{0+}(r,R;E) g_L^{0+}(R,r';E)}{1-VR^2 g_L^{0+}(R,R;E)}\ , \\ g_L^{0+}(x,y;E) &= 2(2E)^{1/2} j_L \\ &\quad \times((2E)^{1/2}x_<) h_L^{(+)}((2E)^{1/2}x_>)\ , \end{aligned} \tag{15}$$

where $g_L^{0+}(E)$ is the contribution of the partial wave with angular momentum L to the Green's function of a free particle of energy E, and $j_L(x)$ and $h_L^{(+)}(x)$ are spherical Bessel functions and spherical Hankel functions of the first kind, respectively.

From Eqs. (10) and (14) one then obtains in a straightforward way the following expression for $\sigma^{[1]}$:

$$\begin{aligned} \sigma^{[1]}(\omega) &= (16\pi^2\omega/3c) \\ &\quad \times \operatorname{Im}\int_0^\infty\int_0^\infty dr\,dr' r^3 (r')^3 u_b(r) u_b(r') \\ &\quad \times g_1^+(r,r';E_b+\omega)\ , \end{aligned} \tag{16}$$

which is directly evaluable making use of the explicit expressions for $u_b(r)$ and $g_1(r,r';E)$, Eqs. (12) and (15), respectively.

The computation of $\sigma^{[2]}$ according to Eq. (10) suggests that we preliminarily get the state $|\,\Phi(\omega)\rangle$, Eq. (5). In the coordinate representation,

$$\begin{aligned} \Phi(\mathbf{r};\omega) &\equiv \langle \mathbf{r}\,|\,\Phi(\omega)\rangle = \int d\mathbf{r}' G^+(\mathbf{r},\mathbf{r}';E_b+\omega) z' u_b(\mathbf{r}') \\ &= (4\pi/3)^{1/2} Y_{10}(\hat{\mathbf{r}}) \int_0^\infty dr' (r')^3 g_1^+(r,r';E_b+\omega) u_b(r') \equiv Y_{10}(\hat{\mathbf{r}}) \varphi(r;\omega)\ . \end{aligned} \tag{17}$$

If we limit ourselves to consider two-photon absorption processes which do not invade the so-called "above-threshold ionization" region,[10] so that $E_b+\omega<0$, $g_1^+(r,r';E_b+\omega)$, which is obtained by analytic continuation from Eq. (15), is a real quantity along with $\varphi(r;\omega)$, Eq. (17), and one is led to the result

$$\begin{aligned} \sigma^{[2]}(\omega) &= \tfrac{1}{6}(4\pi\omega/c)^2 \operatorname{Im}\int_0^\infty\int_0^\infty dr\,dr' r^3 (r')^3 \varphi(r;\omega)\varphi(r';\omega) \\ &\quad \times[g_0^+(r,r';E_b+\omega) \\ &\quad + \tfrac{4}{5} g_2^+(r,r';E_b+\omega)]\ . \end{aligned} \tag{18}$$

It is rather simple, although very annoying, to get $\varphi(r;\omega)$ from Eqs. (15) and (17) and then evaluate $\sigma^{[2]}$, Eq. (18), in a completely analytical form. In view of the lengthy formulas which yield $\sigma^{[1]}$ and $\sigma^{[2]}$ (especially the latter one), however, we shall forbear giving them in explicit form and limit ourselves to presenting the results graphically.

To be as concrete as possible, we have considered the case of the negative hydrogen ion H^-. As far as the theoretical determination of $\sigma^{[1]}$ is concerned, one may document for such a system a fairly abundant literature in terms of variously accurate wave functions for both ground and final ionized state (for a recent paper, with appropriate references, see Ref. 11); even the two-photon ionization cross section $\sigma^{[2]}$ of H^- has been the object of more or less sophisticated calculations,[12] but it is likely that a comparison curve which is reliable over the full range of frequencies beyond the ionization threshold, is not yet available.

According to the approach pursued in this paper, the parameter $\gamma=(-2E_b)^{1/2}$ has been fixed on the basis of the known photodetachment energy of the ion, which leads to $\gamma=0.235\,588$ a.u. From Eq. (13) one then gets a simple relation between radius R of the spherical shell and strength V of the intra-atomic potential energy, $V=\gamma[1-\exp(-2\gamma R)]^{-1}$. In Fig. 1 we have drawn various curves for the photoionization cross section $\sigma^{[1]}$ as a function of the momentum $k_f=[2(E_b+\omega)]^{1/2}$ of the ejected electron, each curve being associated with a given R value. In any case the drawings have been interrupted at $k_f=\gamma$, corresponding to a frequency $\omega=\gamma^2=2\,|\,E_b\,|$. The agreement of the results from the model investigated in this paper with Stewart's[13] (to be considered very reliable) is considerably good, in view of the noticeable simplicity of our model. As R varies in the range 0.3—0.95, the curves drawn for $\sigma^{[1]}$ sandwich the reference points, hinting at optimal R values around 0.6—0.7 a.u. if one intends to sample the whole range of k_f values starting from the threshold. Figure 1 also shows the behavior of $\sigma^{[1]}$ as derived from a like model, where the interaction potential energy between optical electron and atomic neutral core is assumed in the form of the so-called Fermi pseudopotential,[5] i.e.,

$$^{(F)}V = 2\pi a \delta(\mathbf{r})(\partial/\partial r)r \ . \tag{19}$$

It is possible to show that such a potential is able to support a single bound state of S symmetry, with energy $E_b = -\frac{1}{2}/a^2$, whose eigenstate has the simple form

$$^{(F)}u_b(r) = (2\pi a)^{-1/2}\exp(-r/a)/r \ , \tag{20}$$

while the corresponding Green's function $^{(F)}G^+(\mathbf{r},\mathbf{r}';E)$ is expressed in the same way as Eq. (14), with

$$^{(F)}g_L^+(r,r';E) = g_L^{0+}(r,r';E) - \frac{2a\exp[ik_f(r+r')]}{(1+ik_f a)rr'}\delta_{L0} \ . \tag{21}$$

The only parameter (a) contained in this model can be fixed by forcing the photodetachment energy $-E_b$ to be equal to the experimental datum for such quantity, which leads to the choice $a = (-2E_b)^{1/2} \equiv \gamma^{-1}$. Although the qualitative behavior of $\sigma^{[1]}$ as a function of k_f is the correct one, the Fermi pseudopotential model does not appear sufficiently accurate at the quantitative level, its curve being a bit too depressed with respect to the reference points over a substantial range of k_f values. Such a curve is actually coincident with the limit curve obtained for the spherical δ-shell model as the radius R drops to zero. Under such conditions, in fact, the ground state $u_b(r)$ of the spherical shell, Eq. (12), tends to the eigenfunction $^{(F)}u_b(r)$, Eq. (20), while the difference $g_1^+(r,r';E) - g_1^{0+}(r,r';E) = O(R^3)$; from Eqs. (16) and (21) one is thus easily led to rationalize the behavior above stressed.

The evaluation of the two-photon ionization cross section $\sigma^{[2]}$ has been carried out according to Eqs. (17) and (18) just using the same values of the parameters adopted for $\sigma^{[1]}$. Even in this case all of the curves (see Fig. 2) have been interrupted at $k_f = (4\omega - \gamma^2)^{1/2} = \gamma$, in correspondence to the threshold where ionization induced by a single photon becomes possible ("above-threshold ionization" region)[10] and still refer to the case of the ion H^-, with $\gamma = 0.235\,588$ a.u. As R decreases, the curves change their shape appreciably in the region encompassing the maximum and simultaneously shift downward in a uniform way, tending as $R \rightarrow 0$ to a limit curve, which is

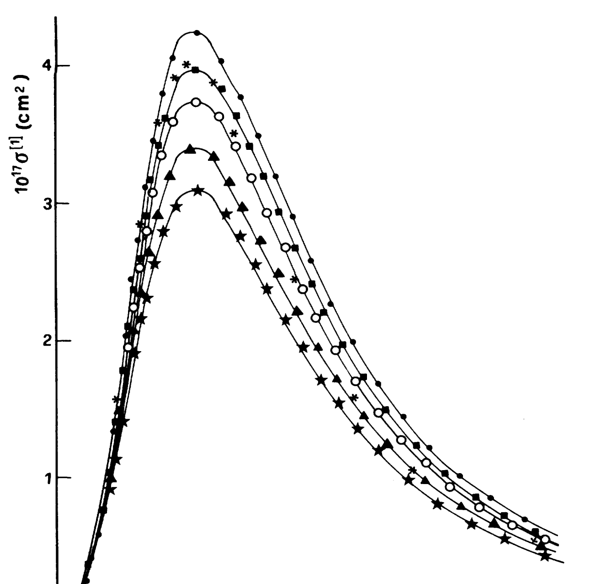

FIG. 1. Behavior of the one-photon ionization cross section for H^- as a function of the momentum k_f of the ejected electron. The various curves refer to different values of the radius R of the spherical δ-shell potential: (-▲-▲-) $R = 0.3$ a.u., (-0-0-) $R = 0.6$ a.u., (-■-■-) $R = 0.775$ a.u., (-●-●-) $R = 0.95$ a.u. The curve (-★-★-) corresponds to the results arising from the Fermi pseudopotential model (or, equivalently, $R \rightarrow 0$), while the unconnected points indicated by * represent Stewart's accurate estimates.

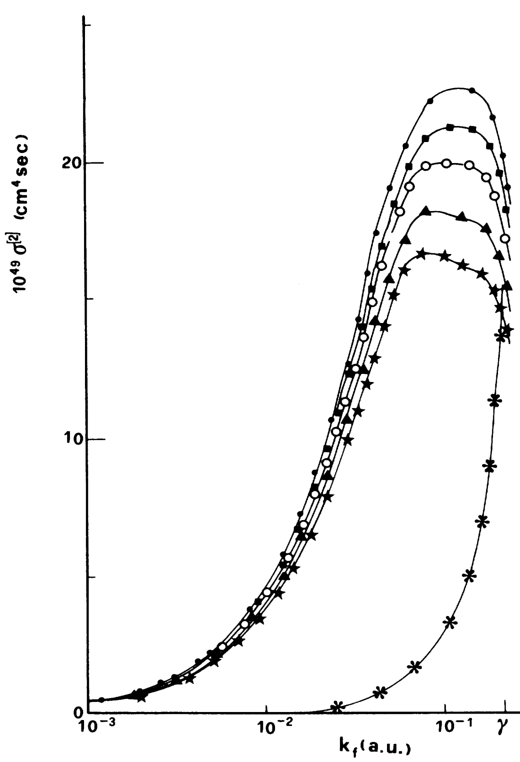

FIG. 2. Behavior of the two-photon ionization cross section for H^- as a function of the momentum k_f of the ejected electron. For the proper identification of the various curves, see the preceding figure. The additional curve (-*-*-) corresponds to values obtained by Crance *et al.* [Ref. 12(c)], that we have extracted from a figure reported in that reference.

the same yielded by the Fermi pseudopotential model. As in the case of $\sigma^{[1]}$, this type of behavior is explained by making reference to Eq. (18) and the properties of $\varphi(r;\omega)$, $g_0^+(r,r';E)$, and $g_2^+(r,r';E)$ as the shell radius $R\to 0$. It is also worthwhile to point out that the inflections exhibited by the various curves represent real peculiarities of the models. In view of the results for $\sigma^{[1]}$ and their good comparison with Stewart, we conjecture that the $\sigma^{[2]}$ curves corresponding to $R\simeq 0.6$—0.7 a.u. could be fairly reliable representations of the two-photon ionization cross section over the range of k_f values considered. The comparison curve in Fig. 2 corresponds to values extracted by us from a figure reported in Ref. 12(c). Such values were deduced by using an approach based on complex dilation techniques and complex square integrable function expansion.

It is also instructive to consider the partition of $\sigma^{[2]}$ in S- and D-partial-wave contributions, both of which in principle play a role in the case here assumed to be of a linearly polarized ionizing radiation [see Eq. (18)]. In Fig. 3 we report the behavior, as a function of k_f, of the S-partial-wave contribution $\sigma_S^{[2]}$ to $\sigma^{[2]}$, at $R=0.775$ a.u. (solid curve); the dashed curve represents the result of a calculation of $\sigma_S^{[2]}$ entirely using the Born approximation as far as both $\varphi(r;\omega)$ and $g_0^+(r,r';E_b+\omega)$ are concerned, while the dot-dashed curve yields the complementary aliquot $\sigma_S^{[2]}-\sigma_{S,\mathrm{Born}}^{[2]}$ which takes into account globally the effects arising from the presence of the intra-atomic interaction potential. The importance of the distortion induced by the potential in both "doorway" state $z\Phi(\mathbf{r};\omega)$ of the ionized electron and imaginary part $\mathrm{Im}g_0^+$ of the propagator is quite evident from the inspection of the figure; this is of course nothing but a different way of asserting the generally important role played by the inclusion of the correct phase shifts in the photoelectron S-wave final wave function. In Fig. 4 we report as a function of k_f the analogous behavior of the D-partial-wave contribution $\sigma_D^{[2]}$ to $\sigma^{[2]}$ (—■■— line), which is not discernible from the result one obtains by a calculation performed entirely with the Born approximation; the dot-dashed curve (which has the same meaning already put in evidence) is obviously totally negligible over the whole range of the k_f values explored (ignorable D-wave phase shifts). The comparison curves refer to values obtained either by other authors or a different model (Fermi pseudopotential). In particular, the full-line curve corresponds to calculations carried out by Fink and Zoller [Ref. 12(b)] by using a hyperspherical adiabatic approach. Figures 3 and 4 emphasize that at sufficiently low energies the angular distribution of the emitted photoelectrons is expected to be

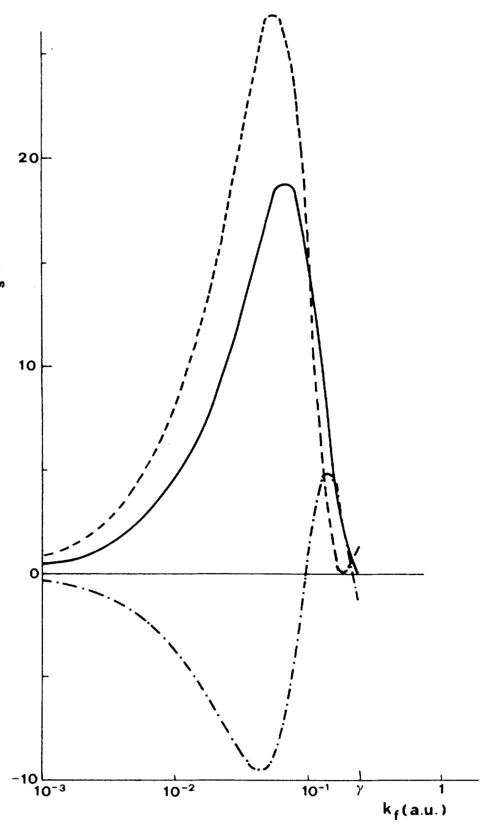

FIG. 3. S-partial-wave contribution to the two-photon ionization cross section for H^- as a function of the momentum k_f of the ejected electron. The case examined corresponds to $R=0.775$ a.u. The solid curve represents the rigorous result $\sigma_S^{[2]}$ predicted by the model, the dashed curve refers to a calculation entirely using the Born approximation, while the dot-dashed curve yields the complement $\sigma_S^{[2]}-\sigma_{S,\mathrm{Born}}^{[2]}$ (see text).

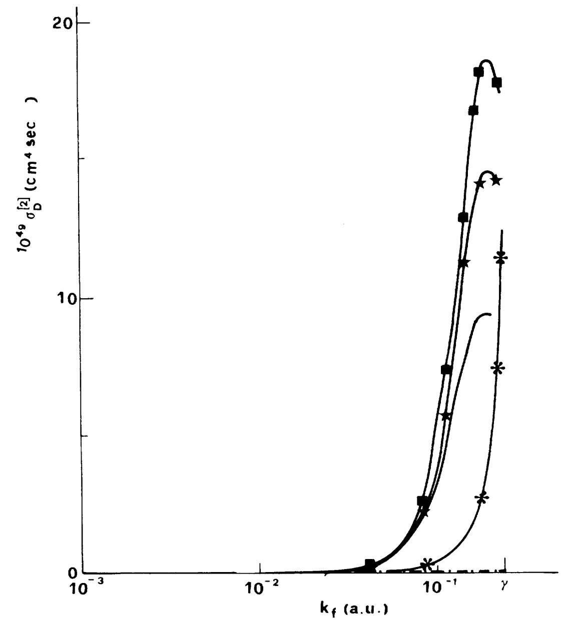

FIG. 4. D-partial-wave contribution to the two-photon ionization cross section for H^- as a function of the momentum k_f of the ejected electron. The -■-■- curve represents the exact result $\sigma_D^{[2]}$ corresponding to $R=0.775$ a.u. It is actually superimposed on the curve for $\sigma_{D,\mathrm{Born}}^{[2]}$, the dot-dashed curve refers to the quantity $\sigma_D^{[2]}-\sigma_{D,\mathrm{Born}}^{[2]}$. The other curves reported provide comparison with previous results: The full-line corresponds to values obtained by Fink *et al.* [Ref. 12(b)], the curve -*-*- to Crance *et al.* [Ref. 12(c)], while -★-★- refers to the Fermi pseudopotential model.

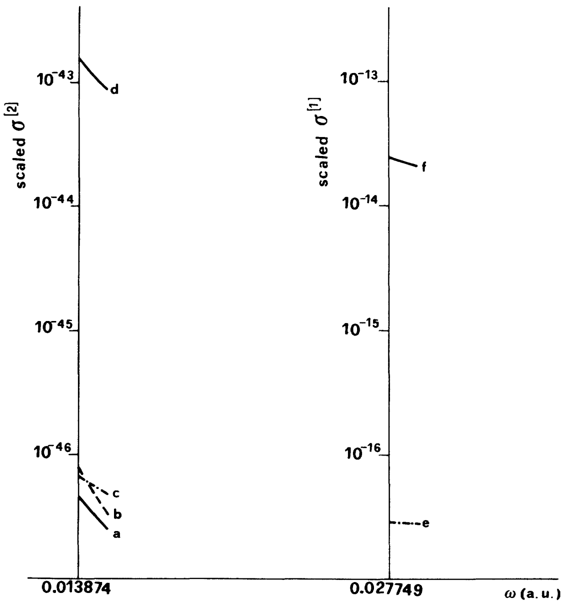

FIG. 5. Test of validity of Wigner threshold laws for the spherical δ-shell model. The two vertical lines at $\omega=0.013\,874$ a.u. and $\omega=0.027\,749$ a.u. mark the threshold for two-photon and one-photon ionization, respectively, and refer to the case $R=0.775$ a.u. The threshold behavior of the relevant cross sections, expressed in terms of scaled quantities (see text), conforms to the following conventions: (a) $\sigma_S^{[2]}/k_f$, (b) $\sigma_{S,\mathrm{Born}}^{[2]}/k_f$, (c) $\sigma_D^{[2]'}/k_f^5$, (d) $\sigma_D^{[2]}/k_f^5$ ($\simeq\sigma_{D,\mathrm{Born}}^{[2]}/k_f^5$), (e) $\sigma^{[1]'}/k_f^3$, (f) $\sigma_{\mathrm{Born}}^{[1]}/k_f^3$ ($\simeq\sigma^{[1]}/k_f^3$).

essentially isotropic, while at the threshold where ionization induced by a single photon is open, the model predicts that the angular distribution will tend to become strongly anisotropic (peaked along the polarization vector of the ionizing field).[12]

It is appropriate to add at this point some remarks concerning the gauge invariance of the results obtainable for $\sigma^{[1]}$ and $\sigma^{[2]}$. As a matter of fact, the expressions deduced for the photoionization cross sections, Eqs. (6) and (7), refer to a definite choice of the gauge (so-called "length" form). Entirely equivalent formulas could, in principle, be written down by using different gauge choices—for example, those leading to the "velocity" or "acceleration" forms[14] for $\sigma^{[1]}$ and $\sigma^{[2]}$. However, one verifies these formally distinct expressions arising from different gauges in a simple way to yield the same observable quantities, provided that *exact* calculations are carried out, which in general involve either using *correct* eigenstates to the Hamiltonian operator H or employing truly *complete* sets of states whenever they are requested (equivalently, the correct propagator). Approximations of any sort, in principle, cannot but result in an invariance breakdown of the evaluated observables, with possibly very erroneous predictions depending on "unappropriately" chosen gauges. Although we have not explicitly verified this point for the spherical δ-shell model, we wish to further stress its general significance, either on the basis of already available elements of knowledge in literature (especially concerning one-photon processes)[15] or thanks to more recent investigations on the behavior of $\sigma^{[2]}$ for a model of atomic anions founded on the use of the Fermi pseudopotential.[5]

As a final topic of the present paper, we would like to examine how the calculated $\sigma^{[1]}$ and $\sigma^{[2]}$ behave at their respective thresholds, so as to put in explicit evidence the validity of Wigner threshold laws[16] for the spherical δ-shell model. In Fig. 5 we report the behavior of the scaled photoionization cross sections $\sigma^{[1]}/k_f^3$, $\sigma_S^{[2]}/k_f$, and $\sigma_D^{[2]}/k_f^5$, the momentum k_f of the ejected electron being raised to the proper exponent $(2l+1)$ expected on the basis of the theory. The two vertical lines at $\omega=\frac{1}{2}\,|E_b|=0.013\,874$ a.u. and $\omega=|E_b|=0.027\,749$ a.u. mark the threshold for two-photon and one-photon ionization, respectively, and correspond to the choice $R=0.775$ a.u. In Fig. 5 we have also represented the behavior of $\sigma_{\mathrm{Born}}^{[1]}/k_f^3$, $\sigma^{[1]'}/k_f^3$, $\sigma_{D,\mathrm{Born}}^{[2]}/k_f^5$, $\sigma_D^{[2]'}/k_f^5$, and $\sigma_{S,\mathrm{Born}}^{[2]}/k_f$ so as to allow possible comparisons between exact results and approximations by the Born method for the model. As expected, the main discrepancy at the threshold appears in $\sigma_S^{[2]}/k_f$, the S contribution to the two-photon ionization cross section, which is appreciably different from its Born approximation $\sigma_{S,\mathrm{Born}}^{[2]}/k_f$. It should be stressed that intense-field effects can cause noticeable deviations from the low-field behavior predicted by perturbative results,[17] particularly as a consequence of the change of the ionization thresholds induced by ac Stark shifts. The implementation of nonperturbative techniques for taking into account nonlinear effects produced by high-intensity sources is a stimulating field of research, which has not yet been sufficiently explored.

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