Multiphoton ionization in strong fields

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The photoionization of atoms in a strong electromagnetic field leads to the effect of "above-threshold" ionization, displaying a number of peaks in the energy spectrum of the photoelectrons. In the case of a one-electron model atom in a circularly polarized field, we derive expressions that describe the position as well as the intensity of those peaks as a function of the field parameters by means of a nonperturbative method. It is immediately clear from our formulas that the lowest-energy peaks in general disappear with increasing field intensity, for Coulomb-like as well as short-range potentials. For a zero-range potential, a numerical evaluation is easily performed, leading to results that are in every respect qualitatively similar to the experimental results obtained thus far on these kinds of processes.

I. INTRODUCTION

Although multiphoton ionization has been studied in atomic physics for many years, this subject has gotten new impetus with the advent of powerful lasers which are capable of producing an electric field incident on an atom, which is a substantial fraction of the field strength caused by the (screened) nucleus on a valence electron. Indeed many interesting experiments on multiphoton ionization of atoms have been performed in recent years.[1–5] The results of a typical experiment concerning N-photon ionization ($N \geq 11$) of Xe are shown in Fig. 1 (Ref. 5). At the wavelength of the laser used, it takes at least the absorption of 11 photons in order to enable an electron to escape from the remaining ion. At the prevailing field intensities, however, we do not only notice a peak associated with the absorption of 11 photons (the left one in the figure), but also peaks associated with 12, 13, etc., photon absorption. These peaks are not sharp, but exhibit a certain broadening (exceeding the detector bandwidth). Another striking feature is the decrease of the first (11 photon) peak with increasing field strength. In a recent communication[6] we related the latter effect to an increase in the ionization potential due to the presence of the field. There it was also argued that this change in the ionization potential canceled the ponderomotive shift (the phenomenon that a charged particle is pushed out of the field[7]). It would be of the order of several eV for the case at hand. The results of Muller *et al.*[6] were based upon a recent result of one of us[8] concerning atoms in circularly polarized fields, applied to the simple case of a zero-range potential, supporting one bound state. In the present work we give a general discussion, also for circularly polarized fields, applicable to those cases where the interaction of the ionized electron with the remaining ion can be described by means of a spherically symmetric, dilatation analytic effective potential. This means that we neglect the fact that the ionization limit acquires an imaginary part (since the ionic ground state is dynamically Stark broadened), but our formalism is exact for hydrogen. The restriction to circularly polarized fields is important in connection with the fact that in this case the Floquet Hamiltonian (see Sec. II) is explicitly known for spherically symmetric potentials. Recently,[9,10] atoms in fields with different polarization properties (in particular linear polarization), have been studied from an abstract point of view. In these cases, however, a numerical evaluation is bound to be more complicated, even for the case of a zero-range potential. Therefore we address ourselves to

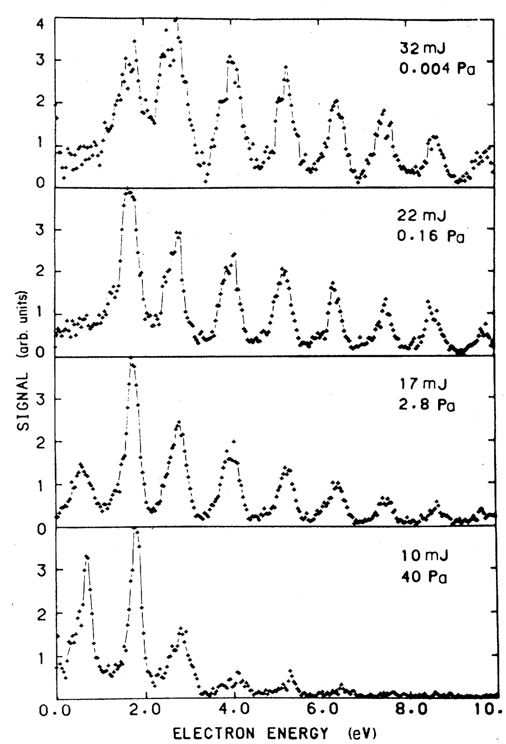

FIG. 1. Electron energy spectrum from ionization of xenon gas by the Nd—yttrium-aluminum-garnet laser (1.064 μm) (from Ref. 5). The first peak corresponds to 11-photon ionization to the $P_{3/2}$, and 12-photon ionization to the $P_{1/2}$ continuum. Estimated intensities are $(2-7)\times 10^{13}$ W/cm^2.

the case where at time $t=0$ the atom is in a given state (typically the ground state), and study its asymptotic time development as $t\rightarrow\infty$ in the presence of a circularly polarized field of constant amplitude. This excludes the transient effects associated with the turning on and off of the field. (The latter problem has been solved recently.[11]) Nevertheless our formalism adequately explains the salient features of the experimental results, such as the decrease of the first electron peak with increasing field intensity. Moreover our approach is nonperturbative (i.e., no series expansion in the field amplitude is made). Since the electromagnetic field turns the atomic eigenvalues into resonances, we have made use of the complex dilatation method which is well adapted for this purpose (the method is due to Aguilar and Combes[12] and Balslev and Combes,[13] for an introduction to the subject, see Reinhardt[14]). The dilatation formalism was used earlier by Chu,[15] who, in particular, considered two-photon ionization in hydrogen. In fact, his starting point and ours coincide, but the further developments in both cases go in different directions.

The present work is organized as follows. In Sec. II we derive general expressions for the energy spectrum of the ejected electrons in terms of probability amplitudes $F_{lm}(k)$. In Sec. III we give approximate expressions for the latter from which the experimentally observed peak structure becomes evident. Section IV is devoted to the case of a zero-range potential for which explicit results are obtained in Sec. V. This paper ends with a discussion section.

II. GENERAL THEORY

We consider an electron bound by a spherically symmetric, dilatation analytic potential $V(r)$ in the presence of a spatially homogeneous circularly polarized external field, represented by the vector potential

$$\vec{A}(t)=(a\cos(\omega t),\, a\sin(\omega t),\, 0)\ . \tag{2.1}$$

In atomic units ($\hbar=m=e=1$) the corresponding Hamiltonian is

$$H(t)=\tfrac{1}{2}[\vec{p}-\vec{A}(t)]^2+V(r)=H_0(t)+V(r) \tag{2.2}$$

acting in $\mathscr{H}=L^2(\mathbb{R}^3)$. Here $\vec{p}$ is the electronic momentum vector, $\vec{x}$ its position vector and $r=|\vec{x}|$. $V(r)$ can be an attractive Coulomb potential (the hydrogen case) or an effective potential felt by a valence electron in an atom or ion. It can have an attractive Coulomb tail (atoms or ions) or be short range (singly charged negative ions such as H^-). In Sec. IV we consider the special case of rank one projector potentials, and their limiting case, the zero-range potential. We let $U(t)$ be the time-evolution operator, associated with $H(t)$, i.e.,

$$\partial_t U(t)=-iH(t)U(t),\quad U(0)=1\ . \tag{2.3}$$

By changing to a rotating reference frame $H(t)$ can be made time independent.[16,17,8] Thus

$$\begin{aligned} &U(t)=\exp(-i\omega l_3 t)\exp(-iH^{\rm Fl}t)\ ,\\ &H^{\rm Fl}=\tfrac{1}{2}(\vec{p}-\vec{a})^2-\omega l_3+V(r)\ , \end{aligned} \tag{2.4}$$

where $l_3=x_1p_2-p_2x_1$ is the third component of the electronic angular momentum vector and $\vec{a}=(a,0,0)$. Similarly we have for the time-evolution operator $U_0(t)$ associated with $H_0(t)$

$$\begin{aligned} &U_0(t)=\exp(-i\omega l_3 t)\exp(-iH_0^{\rm Fl}t)\ ,\\ &H_0^{\rm Fl}=\tfrac{1}{2}(\vec{p}-\vec{a})^2-\omega l_3\ . \end{aligned} \tag{2.5}$$

Also, by direct integration

$$U_0(t)=\exp\left[-i\int_0^t ds\, H_0(s)\right]=\exp\left[-i\left[\tfrac{1}{2}(p^2+a^2)t-\frac{a}{\omega}\{p_1\sin(\omega t)+p_2[1-\cos(\omega t)]\}\right]\right]\ . \tag{2.6}$$

In fact the existence of the two representations (2.5) and (2.6) played a major role in the determination of the spectrum of $H^{\rm Fl}$ in the complex dilated case.[8] $H^{\rm Fl}$ and $H_0^{\rm Fl}$ are the full and the free Floquet Hamiltonians. $H^{\rm Fl}$ is only known explicitly for the case of circular polarization with spherically symmetric $V(r)$. We note that in general the bound states of

$$H^{\rm at}=\tfrac{1}{2}\vec{p}^{\,2}+V(r) \tag{2.7}$$

turn into resonances of $H^{\rm Fl}$. In fact the term $-\omega l_3$ shifts the bottom of the m-subcontinuum of $H^{\rm at}$ by an amount $-m\omega$ which brings it below the energy of any bound state for sufficiently large m. Thus the bound states become continuum embedded. Since they are coupled to each other and the various m-continua by the term $-\vec{p}\cdot\vec{a}$, they will change into resonances. Resonances are conveniently described within the framework of the dilatation analytic method. Thus the dilatation analytic properties of $H^{\rm Fl}$ were studied.[8] The dilatated Floquet Hamiltonian is

$$H^{\rm Fl}(\zeta)=\tfrac{1}{2}[\vec{p}\exp(-\zeta)-\vec{a}]^2-\omega l_3+V[r\exp(\zeta)]\ , \tag{2.8}$$

where ζ is the complex dilatation parameter. As shown in Ref. 8 its spectrum consists of a number of half-lines, starting at the points $\frac{1}{2}a^2+m\omega$, $m\in\mathbb{Z}$ and going off under an angle $-2\,{\rm Im}\zeta$, together with a set of eigenvalues (the resonances), see Fig. 2. We drop the superscript Fl from now on. In case $V(r)$ is a sufficiently smooth short-range potential [i.e., $V(r)=O(r^{-1-\epsilon})$, $\epsilon>0$, as $r\rightarrow\infty$] the existence of the wave operators (as strong limits)

$$\Omega_\pm^{\rm SR}=\lim_{t\rightarrow\pm\infty}U(t)^*U_0(t)=\lim_{t\rightarrow\pm\infty}\exp(iHt)\exp(-iH_0t) \tag{2.9}$$

is easily established by means of Cook's method (see, for instance, Reed and Simon[18]) [in (2.9) SR stands for short range]. In case $V(r)$ has a Coulomb tail (without loss of generality we assume this tail to be attractive and of charge one), we have to replace $U_0(t)$ by the asymptotic evolution operator

$$U_{as}(t)=U_0(t)\exp(ip^{-1}\ln|t|)\ , \qquad (2.10)$$

so that (LR means long range, i.e., a Coulomb tail)

$$\begin{aligned}\Omega_\pm^{LR}&=\lim_{t\to\pm\infty}U^*(t)U_{as}(t)\\&=\lim_{t\to\pm\infty}\exp(iHt)\exp[-i(H_0t-p^{-1}\ln|t|)]\ . \qquad (2.11)\end{aligned}$$

Equation (2.11) is a simple adaptation of Dollard's original expression,[19] whose method of proof can be used in this case as well to establish its existence. We now cast it in a form more convenient for applications. Thus we note that

$$\begin{aligned}\exp(-iH_0t)=\exp(-i\vec{h}\cdot\vec{p})&\\ \times\exp\{-i[\tfrac{1}{2}(p^2+a^2)-\omega l_3]t\}&\\ \times\exp(i\vec{h}\cdot\vec{p})\ ,& \qquad (2.12)\end{aligned}$$

where $\vec{h}=(0,a/\omega,0)$ corresponds to the Hertz vector $\vec{Z}(t)$, $\partial_t\vec{Z}(t)=\vec{A}(t)$, in the nonrotating frame. Equation (2.12) is a version of the Kramers transformation[20] often encountered in work on atomic systems in electromagnetic fields. We write

$$\Omega_\pm^{at}(\vec{h})=\exp(-i\vec{h}\cdot\vec{p})\Omega_\pm^{at}\exp(i\vec{h}\cdot\vec{p})\ , \qquad (2.13)$$

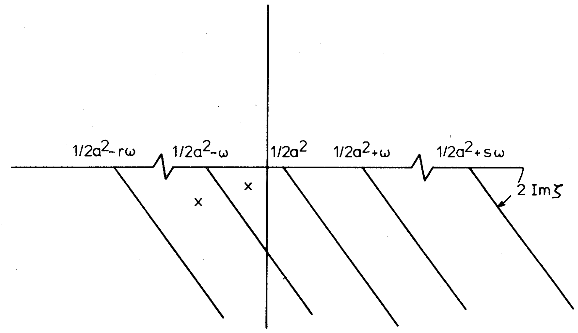

FIG. 2. Spectrum of the complex dilated Hamiltonian $H^{Fl}(\zeta)$; crosses are resonance eigenvalues, solid lines continuous spectrum.

where

$$\Omega_\pm^{at}=\lim_{t\to\pm\infty}\exp(iH^{at}t)\exp[-i(\tfrac{1}{2}p^2t-p^{-1}\ln|t|)] \qquad (2.14)$$

(the two quantities differ in a shift of the coordinate vector in the potential over $-\vec{h}$). Now

$$\begin{aligned}\Omega_\pm^{LR}&=\lim_{t\to\pm\infty}\exp(iHt)\exp(-i\vec{h}\cdot\vec{p})\exp(-iH^{at}t)\exp(iH^{at}t)\exp[-i(\tfrac{1}{2}p^2t-p^{-1}\ln|t|)]\\&\quad\times\exp[-i(\tfrac{1}{2}a^2-\omega l_3)t]\exp(i\vec{h}\cdot\vec{p})\\&=\lim_{t\to\pm\infty}\exp(iHt)\exp(-i\vec{h}\cdot\vec{p})\exp(-iH^{at}t)\Omega_\pm^{at}\exp[-i(\tfrac{1}{2}a^2-\omega l_3)t]\exp(i\vec{h}\cdot\vec{p})\\&=\lim_{t\to\pm\infty}\exp(iHt)\exp(-i\vec{h}\cdot\vec{p})\Omega_\pm^{at}\exp[-i(\tfrac{1}{2}p^2+\tfrac{1}{2}a^2-\omega l_3)t]\exp(i\vec{h}\cdot\vec{p})\\&=\lim_{t\to\pm\infty}\exp(iHt)\Omega_\pm^{at}(\vec{h})\exp(-iH_0t)\ . \qquad (2.15)\end{aligned}$$

Here use was made of the intertwining property

$$\exp(-iH^{at}t)\Omega_\pm^{at}=\Omega_\pm^{at}\exp(-i\tfrac{1}{2}p^2t)\ .$$

With

$$\Omega^{LR}(t)=\exp(iHt)\Omega_\pm^{at}(\vec{h})\exp(-iH_0t)$$

we have

$$\begin{aligned}\Omega_\pm^{LR}&=\Omega_\pm^{at}(\vec{h})+\int_0^{\pm\infty}dt\,\partial_t\Omega^{LR}(t)\\&=\Omega_\pm^{at}(\vec{h})+i\int_0^{\pm\infty}dt\,\exp(iHt)[H\Omega_\pm^{at}(\vec{h})-\Omega_\pm^{at}(\vec{h})H_0]\exp(-iH_0t)\\&=\Omega_\pm^{at}(\vec{h})+i\int_0^{\pm\infty}dt\,\exp(iHt)\{H\Omega_\pm^{at}(\vec{h})-[H_0+V(\vec{x}-\vec{h})]\Omega_\pm^{at}(\vec{h})\}\exp(-iH_0t)\\&=\Omega_\pm^{at}(\vec{h})+i\int_0^{\pm\infty}dt\,\exp(iHt)[V(r)-V(|\vec{x}-\vec{h}|)]\Omega_\pm^{at}(\vec{h})\exp(-iH_0t)\ , \qquad (2.16)\end{aligned}$$

which relation holds when applied to any function in the domain of p^2. Equation (2.16) is a variant of the so-called two-potential formula; note that $V(r)-V(|\vec{x}-\vec{h}|)$ is short range. We note in passing that a general approach to the existence of long-range wave operators in terms of expressions of the type (2.15) has been presented in Refs. 21 and 22. In the short-range case (2.16) is still valid [here $\Omega_\pm^{at}$ is simply $\lim_{t\to\pm\infty}\exp(iH^{at}t)\exp(-ip\tfrac{1}{2}{}^2t)$], but we also have the more simple representation

$$\Omega_{\pm}^{SR} = 1 + i \int_0^{\infty} dt \exp(iHt) V(r) \exp(-iH_0 t) \ . \tag{2.17}$$

We let P be a projector, commuting with $U_0(t)$ and suppose that the system is in the state $|\psi\rangle$ at $t=0$. The probability w to find the system in a state in the P-projected part of $\mathscr{H}$ as $t \to \infty$ is then

$$w = \lim_{t\to\infty} \langle \psi | U^*(t) P U(t) | \psi \rangle = \lim_{t\to\infty} \langle \psi | U^*(t) U_{as}(t) P U_{as}^*(t) U(t) | \psi \rangle = \langle \psi | \Omega_+ P \Omega_+^* | \psi \rangle \tag{2.18}$$

[in the short-range case $U_{as}(t) = U_0(t)$]. In an actual experiment the number of electrons with kinetic energy in a band Δ is measured. This corresponds to a projector P given by the multiplication operator $\chi_\Delta(\frac{1}{2}p^2)$ [$\chi_\Delta(u)$ is the characteristic function of the interval Δ; it is one for u in Δ and zero otherwise]. Thus [if $\Delta = (\frac{1}{2}\delta_1^2, \frac{1}{2}\delta_2^2)$, then $\Delta' = (\delta_1, \delta_2)$]

$$\begin{aligned} w &= \langle \psi | \Omega_+ \chi_\Delta(\tfrac{1}{2}p^2) \Omega_+^* | \psi \rangle = \langle \psi | \Omega_+ \exp(-i\vec{h}\cdot\vec{p}) \chi_\Delta(\tfrac{1}{2}p^2) \exp(i\vec{h}\cdot\vec{p}) \Omega_+^* | \psi \rangle \\ &= \int_{\Delta'} dk \sum_{l,m} \langle \psi | \Omega_+ \exp(-i\vec{h}\cdot\vec{p}) | k,l,m \rangle \langle k,l,m | \exp(i\vec{h}\cdot\vec{p}) \Omega_+^* | \psi \rangle \\ &= \int_{\Delta'} dk \sum_{l,m} | F_{lm}(k) |^2 \ , \end{aligned} \tag{2.19}$$

where the probability amplitude $F_{lm}(k)$ is given by

$$F_{lm}(k) = \langle k,l,m | \exp(i\vec{h}\cdot\vec{p}) \Omega_+^* | \psi \rangle \ . \tag{2.20}$$

Here the $|k,l,m\rangle$'s are free spherical waves and the mathematically strict interpretation of $F_{lm}(k)$ is that of a Bessel transform. The insertion of $\exp(\pm i\vec{h}\cdot\vec{p})$ in (2.19) is done in order to convert $|k,l,m\rangle$ into a continuum eigenstate of H_0, i.e., formally

$$H_0 \exp(-i\vec{h}\cdot\vec{p}) | k,l,m \rangle = [\tfrac{1}{2}(k^2+a^2) - m\omega] \exp(-i\vec{h}\cdot\vec{p}) | k,l,m \rangle \ . \tag{2.21}$$

Equation (2.18) refers to an electron that, although asymptotically free from the remaining ion, is still in the spatially homogeneous field, represented by $\vec{A}(t)$. In an actual experiment, ionization takes place in a laser focus, and the detector is outside the field. On its way out of the field the electron then gains an additional amount $\frac{1}{2}a^2$ of kinetic energy, due to the so-called ponderomotive acceleration effect[7] (Pinard *et al.*[23] have found experimental evidence for this effect). In fact more can happen to the electron on its way out, but it can be shown that for fields $\vec{A}(x,t)$, which decrease slowly for large $\vec{x}$, that this is the only remaining effect. Thus $F_{lm}(k)$ is not the amplitude associated with electrons with energy $\frac{1}{2}k^2$ at the detector (which is outside the field), but with energy $\frac{1}{2}(k^2+a^2)$ instead, i.e., their energy is always $\geq \frac{1}{2}a^2$. This, however, is an artifact of the model considered. Actual fields have a finite spatial extension, and this can be taken into account by localizing the field, $a \to a(r)$, where $a(r)$ vanishes for large r. [Within the present context we cannot allow general $a(\vec{x})$ since then (2.4) would break down.] Now the ponderomotive potential $\frac{1}{2}a^2(r)$ vanishes for large r and this brings back the various thresholds from $\frac{1}{2}a^2 + m\omega$ to $m\omega$ [due to the relative compactness of $\frac{1}{2}a^2(r)$]. Combining $V(r)$ with $\frac{1}{2}a^2(r)$, see Fig. 3, we now encounter a potential with a barrier of macroscopic dimensions (the laser focus), and although electrons with arbitrary low, positive energy can be produced in principle, they have to tunnel through this barrier. This makes the competitive process of picking up one or more additional photons (the electron is still close to the nucleus in these circumstances), and thus gaining a sufficient amount of energy to pass over the barrier, much more likely. Thus our model can be expected to be accurate, only the very small tunneling probability has been neglected. Deviations may occur, however, for electrons with energy only slightly above the barrier height. In that case other small nonuniformity effects in the fields will also become important.

III. EXPRESSIONS FOR THE MULTIPHOTON IONIZATION AMPLITUDES

A. Short-range potentials

We start with the short-range case. Since

$$\begin{aligned} \Omega_+^{SR} &= 1 + i \int_0^{\infty} dt \exp(iHt) V(r) \exp(-iH_0 t) \\ &= 1 + i \lim_{\epsilon \searrow 0} \int_0^{\infty} dt \exp(-\epsilon t) \exp(iHt) V(r) \\ &\quad \times \exp(-iH_0 t) \ , \end{aligned} \tag{3.1}$$

we have (the limit $\epsilon \searrow 0$ being understood)

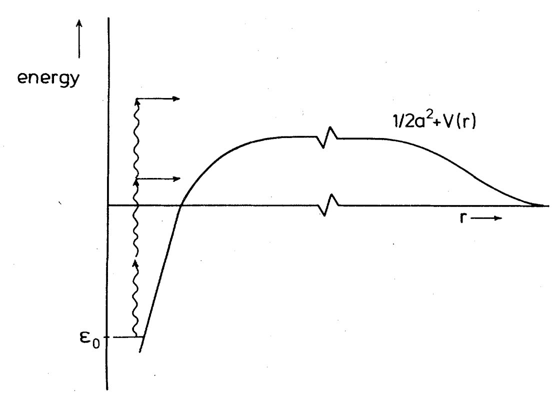

FIG. 3. Typical situation where an electron that has gained twice the photon energy still has to penetrate the barrier in order to escape. After the absorption of an additional photon it can pass over the barrier.

$$F_{lm}(k)=\langle k,l,m\,|\exp(i\vec{h}\cdot\vec{p})\,|\,\psi\rangle + \langle k,l,m\,|\exp(i\vec{h}\cdot\vec{p}) \times V[\tfrac{1}{2}(k^2+a^2)-m\omega+i\epsilon-H]^{-1}\,|\,\psi\rangle = F^B_{lm}(k)+\Phi_{lm}(k)\,, \quad (3.2)$$

where we have split $F_{lm}(k)$ in a Born part F^B (i.e., a part not containing the resolvent of H) and a second part Φ. [Note that the $\exp(\pm i\vec{h}\cdot\vec{p})$ cancel if $|F^B_{lm}(k)|^2$ is summed over l and m.] In order to get some idea about the behavior of Φ let us suppose that $|\psi\rangle$ is the "atomic" ground state $|\phi_0\rangle$ with associated eigenvalue $\epsilon_0<0$. We suppose $|\phi_0\rangle$ to be an s state. Then, in the absence of the symmetry-breaking term $-\vec{p}\cdot\vec{a}$ in H,

$$[\tfrac{1}{2}(k^2+a^2)-m\omega+i\epsilon-H]^{-1}\,|\,\phi_0\rangle$$

changes into (we now set $\epsilon=0$)

$$[\tfrac{1}{2}(k^2+a^2)-m\omega-\epsilon_0]^{-1}\,|\,\phi_0\rangle$$

so that we encounter poles at $\frac{1}{2}k^2=\epsilon_0+m\omega-\frac{1}{2}a^2$. In view of what has been said in Sec. II this corresponds to poles at $\epsilon_0+m\omega$ in terms of the energy outside the field. In the presence of $-\vec{p}\cdot\vec{a}$ the poles will move away from the real axis, but still we expect Φ to peak at such values of $\frac{1}{2}k^2$. The physical interpretation is clear: the peak associated with $m\omega$ corresponds to the absorption of m photons. Negative m cannot occur, since $\frac{1}{2}k^2$ must be positive. In fact m must be such that $\epsilon_0+m\omega-\frac{1}{2}a^2\geq 0$. In order to obtain the precise form of the peaks in Φ we have to continue analytically into the nonphysical sheet where the resonances of H are located. A direct dilatation of the expression for Φ as given in (3.2) may not always be possible. The point is that

$$f(\vec{x})=\langle\vec{x}\,|\,V\exp(-i\vec{h}\cdot\vec{p})\,|\,k,l,m\rangle = V(\vec{x})\langle\vec{x}+\vec{h}\,|\,k,l,m\rangle \quad (3.3)$$

must be square integrable and

$$[U_d(\theta)\vec{f}](\vec{x})=\{\exp[\tfrac{1}{2}i\theta(\vec{x}\cdot\vec{p}+\vec{p}\cdot\vec{x})]f\}(\vec{x}),\quad \theta\in\mathbb{R}\,, \quad (3.4)$$

which is the real dilated function, must have an analytic continuation for $\theta\to\zeta\in\mathbb{C}$, i.e., f is a dilatation analytic vector. Now the shift over $\vec{h}$ leads to complications with branch points. This problem can be circumvented by applying the exterior scaling variant of the dilatation method,[24] but even so

$$\langle\vec{x}+\vec{h}\,|\,k,l,m\rangle=(2\pi)^{-3/2}4\pi k i^l j_l(k\,|\,\vec{x}+\vec{h}\,|\,)Y_l^{\overline{m}}(\Omega_{\vec{x}+\vec{h}})\,, \quad (3.5)$$

where j_l is a spherical Bessel function and $\Omega_{\vec{x}+\vec{h}}$, the solid angle associated with $\vec{x}+\vec{h}$, is an unpleasant quantity to deal with. The reason is that j_l contains sines and cosines. Upon complex dilatation one of the exponentials in the latter will be decaying but the other one blows up, so that we are able to proceed only if V has sufficient (exponential) decay. In the long-range case the situation becomes even more complicated, since then we encounter spherical Coulomb waves. The way out of this problem is to apply the Feshbach formula. Thus we let $P=|\,\phi_0\rangle\langle\phi_0\,|$. Then ($Q=1-P$, $H_{PQ}=PHQ$, etc.)

$$(z-H)^{-1}=(z-H_Q)^{-1}Q+[(z-H_Q)^{-1}H_{QP}+P] \times G_p(z)[P+H_{PQ}(z-H_Q)^{-1}]\,,$$
$$G_p(z)=P(z-H)^{-1}P = [z-H_P-H_{PQ}(z-H_Q)^{-1}H_{QP}]^{-1}P\,, \quad (3.6)$$

which expression is valid for z outside the spectra of H and H_Q. Now

$$(z-H)^{-1}\,|\,\phi_0\rangle=[(z-H_Q)^{-1}H_{QP}+P]G_p(z)\,|\,\phi_0\rangle\,, \quad (3.7)$$

so that

$$\Phi=\langle k,l,m\,|\exp(i\vec{h}\cdot\vec{p})V[(z-H_Q)^{-1}H_{QP}+1]\,|\,\phi_0\rangle \times\langle\phi_0\,|\,(z-H)^{-1}\,|\,\phi_0\rangle\,, \quad (3.8)$$
$$z=z_m(k)=\tfrac{1}{2}(k^2+a^2)-m\omega+i\epsilon\,.$$

Q removes the ground state from H^{at} and consequently the corresponding resonance will be absent in H_Q. We therefore expect the first factor in Φ to vary smoothly for z close to this resonance. The second factor can be dilated straightforwardly

$$g(z)=\langle\phi_0\,|\,(z-H)^{-1}\,|\,\phi_0\rangle = \langle\phi_0(\bar{\zeta})\,|\,[z-H(\zeta)]^{-1}\,|\,\phi_0(\zeta)\rangle\,, \quad (3.9)$$

where ζ is the complex dilatation parameter. In case there is another resonance close to the one associated with ϵ_0 (this happens if the field frequency is such that the difference of ϵ_0 and another atomic eigenvalue is nearly a multiple of ω, resonant multiphoton ionization), we simply enlarge P so that Q also projects away this second eigenvalue. Then $P(z-H)^{-1}P$ can again be dilated. Suppose now that $\epsilon_0(a)$ is the eigenvalue of $H(\zeta)$, corresponding to ϵ_0 for $\vec{a}=0$ and we let

$$P(\zeta,\vec{a})=|\,\phi_0(\zeta,\vec{a})\rangle\langle\phi_0(\bar{\zeta},\vec{a})\,|$$

be the corresponding eigenprojector. [We suppose that the dilatation angle $\psi=2\,\text{Im}\zeta$ is such that $\epsilon_0(a)$ is uncovered indeed.] We assume now that no other resonances are near $\epsilon_0(\vec{a})$. Then

$$g(z)=[z-\epsilon_0(\vec{a})]^{-1}\langle\phi_0(\bar{\zeta})\,|\,\phi_0(\zeta,a)\rangle \times\langle\phi_0(\bar{\zeta},\vec{a})\,|\,\phi_0(\zeta)\rangle+g_{\text{bg}}(z)\,, \quad (3.10)$$

where $[Q(\zeta,\vec{a})=1-P(\zeta,\vec{a})]$

$$g_{\text{bg}}(z)=\langle\phi_0(\bar{\zeta})\,|\,Q(\zeta,a)[z-H(\zeta)]^{-1}\,|\,\phi_0(\zeta)\rangle \quad (3.11)$$

is a background contribution as compared to the first term in $g(z)$ which peaks in $z=\text{Re}\epsilon_0(\vec{a})$. The peaking will be the more pronounced the smaller $\Gamma=\text{Im}\epsilon_0(\vec{a})$. Indeed Γ will be small for $\vec{a}$ close to zero, in which case the overlap factor

$$\langle \phi_0(\bar{\zeta}) \mid \phi_0(\zeta,\vec{a}) \rangle \langle \phi_0(\bar{\zeta},\vec{a}) \mid \phi_0(\zeta) \rangle$$

will be close to one. As to what happens for very large $\vec{a}$, not much is known. Present day experiments[5] concerning 11 and more photoionization of Xe still show a set of well-resolved "N-photon" peaks; $N \geq 11$. We finally note that due to the dilatation analyticity the $\epsilon \searrow 0$ limit in $g(z)$ exists trivially, except for the thresholds $\frac{1}{2}a^2 + m\omega$, $m \in \mathbb{Z}$. In the remaining term in (3.8) we also expect no difficulties with this limit provided $V(r)$ has sufficient decay ($H_{PQ} \mid \phi_0 \rangle = \vec{p}\cdot\vec{a} \mid \phi_0 \rangle$ has exponential decay, along with ϕ_0). In summary we have

$$\Phi = [z(k) - \epsilon_0(\vec{a})]^{-1} \eta(k) + \Phi^{\rm bg}(k) , \qquad (3.12)$$

where the background part $\Phi^{\rm bg}$ originates from the contribution containing $g_{\rm bg}(z)$. We can make the detector bandwidth Δ' so large that it covers the peaks completely. Thus we let m be sufficiently large positive, fixed. The corresponding $w = w_m$ (see 2.19) is then obtained by integrating $|F_{lm}(k)|^2$ over Δ' and summing over the appropriate l

$$w_m = \int_{\Delta'} dk \sum_l |F_{lm}(k)|^2$$
$$= \sum_l \int_{\Delta'} dk \, | F^B_{lm}(k) + \Phi^{\rm bg}_{lm}(k) + [z_m(k) - \epsilon_0(\vec{a})]^{-1} \eta_{lm}(k) |^2 . \qquad (3.13)$$

Since $F^B_{lm}(k)$, $\Phi^{\rm bg}_{lm}(k)$, and $\eta_{lm}(k)$ are supposed to vary slowly across a peak, it makes sense to evaluate them at the value k_m of k for which $|z_m(k) - \epsilon_0(\vec{a})|^{-1}$ peaks. Then

$$w_m \approx \sum_l |F^B_{lm}(k_m) + \Phi^{\rm bg}_{lm}(k_m)|^2 \Delta' + \sum_l \{ [F^B_{lm}(k_m) + \Phi^{\rm bg}_{lm}(k_m)]^* \int_{\Delta'} dk [z_m(k) - \epsilon_0(\vec{a})]^{-1} \eta_{lm}(k_m) + \text{c.c.} \}$$
$$+ \sum_l \int_{\Delta'} dk \, |z_m(k) - \epsilon_0(\vec{a})|^{-2} |\eta_{lm}(k_m)|^2 . \qquad (3.14)$$

We now make a further approximation by extending the k integration over the full k range, $0 \leq k \leq \infty$. The integrals are easily evaluated with the result

$$w_m \approx \sum_l |F^B_{lm}(k_m) + \Phi^{\rm bg}_{lm}(k_m)|^2 \Delta'$$
$$+ \sum_l \{ [F^B_{lm}(k_m) + \Phi^{\rm bg}_{lm}(k_m)]^* (\pi i/\sqrt{2}) [\epsilon_0(\vec{a}) + m\omega - \tfrac{1}{2}a^2]^{-1/2} \eta_{lm}(k_m) + \text{c.c.} \}$$
$$+ \sum_l \Gamma^{-1} (\pi/\sqrt{2}) |\epsilon_0(\vec{a}) + m\omega - \tfrac{1}{2}a^2|^{-1} \text{Re}[\epsilon_0(\vec{a}) + m\omega - \tfrac{1}{2}a^2]^{1/2} |\eta_{lm}(k_m)|^2 . \qquad (3.15)$$

Note that w_m refers to the peak in the ejected electron spectrum located at $\text{Re}\epsilon_0(\vec{a}) + m\omega$ outside the field (see the remark made in Sec. II). For small Γ the last term in (3.15) is the leading one. Here the factor $\text{Re}[\epsilon_0(\vec{a}) + m\omega - \frac{1}{2}a^2]^{1/2}$ is interesting since it decreases rapidly in magnitude once $\text{Re}[\epsilon_0(\vec{a}) + m\omega - \frac{1}{2}a^2]$ comes close to zero. Experimentally this situation is realized by increasing the laser intensity (i.e., $\frac{1}{2}a^2$). Indeed it is found that the 11 photon peak in the Xe photoionization experiment[5] decreases rapidly under these circumstances. One might argue that $\text{Re}\epsilon_0(\vec{a})$ is also dependent on the field intensity through $\vec{a}$. In fact the eventual shift in $\epsilon_0(\vec{a})$ is precisely the dynamic Stark shift of the ground-state energy. For sufficiently small ω this shift is much smaller than $\frac{1}{2}a^2$, as can be seen by considering its perturbation expansion in the electric dipole representation where the field dependence of the Hamiltonian is through $\vec{x}\cdot\vec{E}$. The strong localization of the ground state leads to a small shift only (in fact no convergent perturbation expansion for the perturbed eigenvalue exists in this representation, but still we get some idea about what to expect).

B. Long-range potentials

In the long-range case we have to start from (2.16) instead of (3.1) with the result

$$F_{lm}(k) = \langle k,l,m \mid (\Omega^{\rm at}_+)^* \exp(i\vec{h}\cdot\vec{p}) \mid \psi \rangle$$
$$+ \langle k,l,m \mid (\Omega^{\rm at}_+)^* \exp(i\vec{h}\cdot\vec{p}) [V(r) - V(|\vec{x} + \vec{h}|)] [\tfrac{1}{2}(k^2 + a^2) - m\omega + i\epsilon - H]^{-1} \mid \psi \rangle , \qquad (3.16)$$

where, in the case of a pure Coulomb potential (hydrogen), $\Omega^{\rm at}_+ \mid klm \rangle$ is an outgoing spherical Coulomb wave. Apart from this difference the same procedure can be followed as in the short-range situation, and with corresponding results

In a numerical evaluation it is important to know the values of the quantities

$$K_1(k) = \langle k,l,m \mid \exp(i\vec{h}\cdot\vec{p}) V \mid \phi_0 \rangle$$
$$= \langle k,l,m \mid \exp(i\vec{h}\cdot\vec{p}) (H^{\rm at} - \tfrac{1}{2}p^2) \mid \phi_0 \rangle$$
$$= (\epsilon_0 - \tfrac{1}{2}k^2) \langle k,l,m \mid \exp(i\vec{h}\cdot\vec{p}) \mid \phi_0 \rangle \qquad (3.17)$$

and

$$K_2(k)=\langle k,l,m \mid \exp(i\vec{h}\cdot\vec{p})V(z-H_Q)^{-1}H_{QP} \mid \phi_0\rangle \ . \tag{3.18}$$

Since $H_{QP}\mid\phi_0\rangle=-\vec{p}\cdot\vec{a}\mid\phi_0\rangle$, we have

$$K_2(k)=-\langle k,l,m \mid V[\tfrac{1}{2}k^2-m\omega+i\epsilon -(H^{\rm at}-\omega l_3)_Q]^{-1}\vec{p}\cdot\vec{a}\mid\phi_0\rangle +O(a^2)\ , \tag{3.19}$$

but a convergent expansion in powers of a does not seem to be possible. Still one might try to obtain an Agmon-type estimate for $K_2(k)$ (see Reed and Simon,[25] Chap. 9), but this involves at least a great deal of work. It would of course be pleasant if one could show $K_2(k)$ to be much smaller than $K_1(k)$ for $\frac{1}{2}k^2\approx\epsilon_0+m\omega$, i.e., in the m-photon peak. The alternative is a direct numerical evaluation of the continuum eigenstate $\Omega_+\exp(i\vec{h}\cdot\vec{p})\mid k,l,m\rangle$ of H. There is, however, one exception, namely the case where V is a separable potential (finite-dimensional projector), including the limiting case of a zero-range potential. A numerical evaluation based on the latter has already been performed.[6] In the next section we turn to such potentials in some detail.

We further note that it is possible to give an expression for $F_{lm}(k)$ without a Born term, but at the expense of introducing the atomic wave operator $\Omega_+^{\rm at}$. Starting from (3.16) it follows after a couple of formal manipulations and the introduction of the Feshbach decomposition for the resolvent (P and Q as before) that

$$\begin{aligned}F_{lm}(k)=&-\langle\phi_0\mid(z-H)^{-1}\mid\phi_0\rangle\\ &\times\{\langle k,l,m\mid(\Omega_+^{\rm at})^*\exp(i\vec{h}\cdot\vec{p})\vec{p}\cdot\vec{a}\mid\phi_0\rangle\\ &+\langle k,l,m\mid(\Omega_+^{\rm at})^*\exp(i\vec{h}\cdot\vec{p})\mid\phi_0\rangle\langle\phi_0\mid\vec{p}\cdot\vec{a}(z-H_Q)^{-1}\vec{p}\cdot\vec{a}\mid\phi_0\rangle\\ &+\langle k,l,m\mid(\Omega_+^{\rm at})^*\exp(i\vec{h}\cdot\vec{p})[V(r)-V(\mid\vec{x}-\vec{h}\mid)](z-H_Q)^{-1}\vec{p}\cdot\vec{a}\mid\phi_0\rangle\}\ ,\end{aligned} \tag{3.20}$$

valid for both short- and long-range $V(r)$. In this connection we note that

$$\exp(-i\vec{h}\cdot\vec{p})\Omega_+^{\rm at}\mid k,l,m\rangle$$

is simply an atomic continuum state with energy $\frac{1}{2}k^2$, but with the shifted potential $V(\mid\vec{x}-\vec{h}\mid)$.

We finally comment upon the choice $\mid\psi\rangle=\mid\phi_0\rangle$ made in this section. This choice corresponds to a sudden switching on of the vector potential at $t=0$, whereas it increases gradually (on a time scale associated with the period of the field) in actual experimental situations. In order to be sure that the electron spectrum measured in such an experiment results from photoionization at some definite intensity, ionization in the transient region during which the field is switched on must be negligible. So, in general, $\mid\psi\rangle$ will be a mixture of bound states of the atom. If, however, none of the higher bound states is resonant with the ground state, their contribution will be very small due to energy mismatch. Even if the field intensity passes through a value for which a higher state can be resonantly excited from the ground state (this depends on the field intensity because the levels are ac-Stark shifted), it will not be populated appreciably provided the time during which the resonance condition is fulfilled is short enough.[26] In most instances this is automatically the case if the switching on is fast enough to give negligible ionization.

IV. SEPARABLE POTENTIALS

Instead of a multiplicative potential $V(r)$, we now consider a potential of the type

$$V=-\lambda\mid\phi\rangle\langle\phi\mid,\ \ \lambda>0\ , \tag{4.1}$$

where $\mid\phi\rangle$ is spherically symmetric $[\langle\vec{x}\mid\phi\rangle=\phi(\mid\vec{x}\mid)]$ and dilatation analytic. Then $H^{\rm at}=\frac{1}{2}\vec{p}^{\,2}-\lambda\mid\phi\rangle\langle\phi\mid$ has, for sufficiently large λ, a bound state $\mid\phi_0\rangle$ given by (unnormalized)

$$\mid\phi_0\rangle=(\epsilon_0-\tfrac{1}{2}p^2)^{-1}\mid\phi\rangle\ , \tag{4.2}$$

where $\epsilon_0<0$, the corresponding eigenvalue, is given by the implicit equation

$$\langle\phi\mid(\epsilon_0-\tfrac{1}{2}p^2)^{-1}\mid\phi\rangle=-\lambda^{-1}\ . \tag{4.3}$$

The formalism in the preceding section still applies, except that, due to the nonlocal nature of V, we now have

$$\begin{aligned}H&=\tfrac{1}{2}(\vec{p}-\vec{a})^2-\omega l_3+\exp(i\vec{a}\cdot\vec{x})V\exp(-i\vec{a}\cdot\vec{x})\\ &=\tfrac{1}{2}(\vec{p}-\vec{a})^2-\omega l_3+V(\vec{a})\ .\end{aligned} \tag{4.4}$$

This follows by starting from the representation, where the field term is $-\vec{x}\cdot\vec{E}(t)$ $[\vec{E}(t)=-\partial_t\vec{A}(t)$ is the electric field vector], the so-called $\vec{x}$ form of the formalism. Since $\vec{E}(t)$ is a physically observable quantity we must have V as given by (4.1). Referring to the representation introduced in Sec. I (the so-called $\vec{p}$ form), we then obtain (4.4) for the Floquet Hamiltonian. In the limiting case of a zero-range potential, V becomes local once more so that the $\exp(\pm i\vec{a}\cdot\vec{x})$ cancel.[27]

Since V and $V(\vec{a})$ are rank-one operators, the wave operators exist in the standard sense (i.e., short-range case), so that taking again $\mid\psi\rangle=\mid\phi_0\rangle$,

$$\begin{aligned}&\Phi_{lm}(k)=\langle k,l,m\mid\exp(i\vec{h}\cdot\vec{p})V(\vec{a})(z-H)^{-1}\mid\phi_0\rangle\ ,\\ &z=\tfrac{1}{2}(k^2+a^2)-m\omega+i\epsilon\ .\end{aligned} \tag{4.5}$$

$V(\vec{a})$ in (4.4) is still a rank-one operator,

$$V(\vec{a}) = -\lambda \exp(i\vec{a}\cdot\vec{x})\,|\,\phi\rangle\langle\phi\,|\exp(-i\vec{a}\cdot\vec{x})$$
$$= -\lambda\,|\,\phi(\vec{a})\rangle\langle\phi(\vec{a})\,| \;, \tag{4.6}$$

so that

$$\Phi_{lm}(k) = -\lambda\langle k,l,m\,|\exp(i\vec{h}\cdot\vec{p})\,|\,\phi(\vec{a})\rangle \times\langle\phi(\vec{a})\,|\,(z-H)^{-1}\,|\,\phi_0\rangle \;. \tag{4.7}$$

Since for rank-one V we have

$$(z-H)^{-1} = (z-H_0)^{-1} - \frac{(z-H_0)^{-1}\,|\,\phi(\vec{a})\rangle\langle\phi(\vec{a})\,|\,(z-H_0)^{-1}}{\lambda^{-1}+\langle\phi(\vec{a})\,|\,(z-H_0)^{-1}\,|\,\phi(\vec{a})\rangle} \;, \tag{4.8}$$

it follows that

$$-\lambda\,|\,\phi(\vec{a})\rangle\langle\phi(\vec{a})\,|\,(z-H)^{-1} = \frac{-\,|\,\phi(\vec{a})\rangle\langle\phi(\vec{a})\,|\,(z-H_0)^{-1}}{\lambda^{-1}+\langle\phi(\vec{a})\,|\,(z-H_0)^{-1}\,|\,\phi(\vec{a})\rangle} \;. \tag{4.9}$$

Consequently

$$\Phi_{lm}(k) = \frac{\langle k,l,m\,|\exp(i\vec{h}\cdot\vec{p})\,|\,\phi(\vec{a})\rangle\langle\phi(\vec{a})\,|\,(z-H_0)^{-1}\,|\,\phi_0\rangle}{\lambda^{-1}+\langle\phi(\vec{a})\,|\,(z-H_0)^{-1}\,|\,\phi(\vec{a})\rangle} \;. \tag{4.10}$$

Suppose now that $|\,\phi(\vec{a})\rangle$ is a dilatation analytic vector [in view of the $\exp(i\vec{a}\cdot\vec{x})$ this implies that $\phi(\vec{x}) = \langle\vec{x}\,|\,\phi\rangle$ must decay sufficiently fast]. Then $H(\zeta)$ is dilatation analytic and its (complex) eigenvalues $\epsilon(\vec{a})$ follow from the equation

$$\langle\phi(\vec{a},\bar{\zeta})\,|\,[\epsilon(\vec{a})-H_0(\zeta)]^{-1}\,|\,\phi(\vec{a},\zeta)\rangle = -\lambda^{-1} \tag{4.11}$$

of which (4.3) is a special case. Taking $\mathrm{Im}\zeta > 0$ sufficiently large, we expect to find one eigenvalue $\epsilon(\vec{a})$ that coincides with ϵ_0 for vanishing a and is analytic in a. Thus the denominator in (4.10) vanishes for $z = \epsilon(\vec{a})$ and $\Phi_{lm}(k)$ has a resonance peak for $z \approx \mathrm{Re}\epsilon(\vec{a})$. For small $\mathrm{Im}\epsilon(\vec{a})$ we have for z close to the resonance value

$$[\lambda^{-1}+\langle\phi(\vec{a})\,|\,(z-H_0)^{-1}\,|\,\phi(\vec{a})\rangle]^{-1} = \langle\phi(\vec{a},\bar{\zeta})\,|\,\{-[\epsilon(\vec{a})-H_0(\zeta)]-[z-H_0(\zeta)]^{-1}\}\,|\,\phi(\vec{a},\zeta)\rangle^{-1}$$
$$\approx [z-\epsilon(\vec{a})]^{-1}\langle\phi(\vec{a},\bar{\zeta})\,|\,[\epsilon(\vec{a})-H_0(\zeta)]^{-2}\,|\,\phi(\vec{a},\zeta)\rangle^{-1} \;, \tag{4.12}$$

so that

$$\Phi_{lm}(k) \approx [z-\epsilon(\vec{a})]^{-1}\frac{\langle k,l,m\,|\exp(i\vec{h}\cdot\vec{p})\,|\,\phi(\vec{a})\rangle\langle\phi(\vec{a})\,|\,(z-H_0)^{-1}\,|\,\phi_0\rangle}{\langle\phi(\vec{a},\bar{\zeta})\,|\,[\epsilon(\vec{a})-H_0(\zeta)]^{-2}\,|\,\phi(\vec{a},\zeta)\rangle}$$
$$\approx [z-\epsilon(\vec{a})]^{-1}\frac{\langle k,l,m\,|\exp(i\vec{h}\cdot\vec{p})\,|\,\phi(\vec{a})\rangle\langle\phi(\vec{a},\zeta)\,|\,[\epsilon(\vec{a})-H_0(\zeta)]^{-1}\,|\,\phi_0(\zeta)\rangle}{\langle\phi(\vec{a},\bar{\zeta})\,|\,[\epsilon(\vec{a})-H_0(\zeta)]^{-2}\,|\,\phi(\vec{a},\zeta)\rangle}$$
$$= [z-\epsilon(\vec{a})]^{-1}\langle k,l,m\,|\exp(i\vec{h}\cdot\vec{p})\,|\,\phi(\vec{a})\rangle\frac{\langle\phi_0(\vec{a},\bar{\zeta})\,|\,\phi_0(\zeta)\rangle}{\langle\phi_0(\vec{a},\bar{\zeta})\,|\,\phi_0(\vec{a},\zeta)\rangle} \;, \tag{4.13}$$

where $|\,\phi_0(\vec{a},\bar{\zeta})\rangle = [\epsilon(\vec{a})-H_0(\zeta)]^{-1}\,|\,\phi(\vec{a}),\zeta)\rangle$ is the (unnormalized) eigenvector of $H(\zeta)$, associated with $\epsilon(\vec{a})$.

We now turn to the case of the zero-range potential. Although this potential cannot be presented in the form (4.1) in a strict sense, it is possible to give it a rigorous meaning in terms of the resolvent.[27] Here we encounter a slight generalization, due to the presence of the field-dependent terms. Thus we let $\phi_\delta(p) = \langle\vec{p}\,|\,\phi_\delta\rangle = (2\pi)^{-3/2}\rho_\delta(p)$, where $\rho_\delta(p)\to 1$ as $\delta\searrow 0$. Examples are $\rho_\delta(p) = \exp(-\delta p^2)$ and $\rho_\delta(p) = (1+\delta p^2)^{-1}$. Then, for absolutely integrable f

$$\langle\phi_\delta(\vec{a})\,|\,f\rangle = (2\pi)^{-3/2}\int d\vec{p}\,\langle\phi_\delta\,|\exp(-i\vec{a}\cdot\vec{x})\,|\,f\rangle$$
$$= (2\pi)^{-3/2}\int d\vec{p}\,\rho_\delta(p)f(\vec{p}+\vec{a}) \to (2\pi)^{-3/2}\int d\vec{p}\,f(p) \quad \text{as } \delta\searrow 0$$
$$= \langle\vec{x}\,|\,f\rangle\,|_{\vec{x}=\vec{0}} \equiv \langle\vec{0}\,|\,f\rangle \;. \tag{4.14}$$

Thus, formally, $|\,\phi_\delta> \to |\,\vec{0}\rangle$. $(z-H_0)^{-1}\,|\,\phi_\delta(\vec{a})\rangle$ has a limit in $\mathscr{H}$ as $\delta\searrow 0$. Indeed, using the resolvent equation $(z-H_0)^{-1} = (z-H_1)^{-1}+(z-H_0)^{-1}\vec{p}\cdot\vec{a}(z-H_1)^{-1}$, where $H_1 = \frac{1}{2}(p^2+a^2)-\omega l_3$, we have ($\mathrm{Im}z \neq 0$)

$$(z-H_0)^{-1}\,|\,\vec{0}\rangle = (z-H_1)^{-1}\,|\,\vec{0}\rangle + (z-H_1)^{-1}\vec{p}\cdot\vec{a}(z-H_1)^{-1}\,|\,\vec{0}\rangle + (z-H_0)^{-1}[\vec{p}\cdot\vec{a}(z-H_1)^{-1}]^2\,|\,\vec{0}\rangle \;. \tag{4.15}$$

Now $(z-H_1)^{-1}\,|\,\vec{0}\rangle = [z-\frac{1}{2}(p^2+a^2)]^{-1}\,|\,\vec{0}\rangle \in \mathscr{H}$ ($|\,\vec{0}\rangle$ is rotational invariant) and, with $P_m = |\,m\rangle\langle m\,|$,

$$(z-H_1)^{-1}\vec{p}\cdot\vec{a}(z-H_1)^{-1}\,|\,\vec{0}\rangle = \sum_{j=\pm 1}[z-\tfrac{1}{2}(p^2+a^2)+j\omega]^{-1}[z-\tfrac{1}{2}(p^2+a^2)]^{-1}P_j\vec{p}\cdot\vec{a}\,|\,\vec{0}\rangle \;, \tag{4.16}$$

which is also square integrable; the same being true for $[\vec{p}\cdot\vec{a}(z-H_1)^{-1}]^2\,|\,\vec{0}\rangle$. Consequently $(z-H_0)^{-1}\,|\,\vec{0}\rangle \in \mathscr{H}$ and, in fact, is a dilatation analytic vector, as can also be seen from (4.15). Keeping ϵ_0 fixed and letting $\delta\searrow 0$, we find that λ^{-1} in (4.3) blows up. Since λ^{-1} occurs in the denominator

$$D_\delta=\lambda^{-1}+\langle\phi_\delta(\vec{a})\,|\,(z-H_0)^{-1}\,|\,\phi_\delta(\vec{a})\rangle\ , \tag{4.17}$$

we have to check that D_δ has a limit D as $\delta\searrow 0$. Writing

$$\begin{aligned}D_\delta=&\ \langle\phi_\delta(\vec{a})\,|\,(z-H_0)^{-1}-(z-H_1)^{-1}\,|\,\phi_\delta(\vec{a})\rangle\\&+\langle\phi_\delta\,|\,\exp(-i\vec{a}\cdot\vec{x})(z-H_1)^{-1}\exp(i\vec{a}\cdot\vec{x})-(\epsilon_0-\tfrac{1}{2}p^2)^{-1}\,|\,\phi_\delta\rangle\\&\to\langle\vec{0}\,|\,(z-H_0)^{-1}-(z-H_1)^{-1}\,|\,\vec{0}\rangle+\langle\vec{0}\,|\,(z-\tfrac{1}{2}a^2-\tfrac{1}{2}p^2)^{-1}-(\epsilon_0-\tfrac{1}{2}p^2)^{-1}\,|\,\vec{0}\rangle\quad\text{as }\delta\searrow 0\\&=D_1+D_2=D(z)\ ,\end{aligned} \tag{4.18}$$

where we substituted (4.3) for λ^{-1}, we have to show that D_1 and D_2 are finite. Defining $\frac{1}{2}k_0^2=-\epsilon_0$ and $\frac{1}{2}\hat{k}^2=-z+\frac{1}{2}a^2$, we have for $\text{Im}z>0$,

$$D_2=(2\pi)^{-3}\int d\vec{p}\{[\tfrac{1}{2}(p^2+k_0^2)]^{-1}-[\tfrac{1}{2}(p^2+\hat{k}^2)]^{-1}\}=(2\pi)^{-1}(\hat{k}-k_0)\ , \tag{4.19}$$

which is finite. Turning to D_1 we note that

$$(z-H_j)^{-1}=-i\int_0^\infty dt\,\exp[i(z-H_j)t],\quad \text{Im}z>0,\ j=0,1\ . \tag{4.20}$$

Since Eqs. (2.5) and (2.6) give

$$\exp(-iH_0t)=\exp(i\omega l_3t)\exp[-i\tfrac{1}{2}(p^2+a^2)t]\exp\{i(a/\omega)[p_1\sin\omega t+p_2(1-\cos\omega t)]\} \tag{4.21}$$

and $\langle\vec{0}\,|\exp(i\omega l_3t)=\langle\vec{0}\,|$, we have, integrating over the angle of $\vec{p}$,

$$\begin{aligned}D_1&=-i\left\langle\vec{0}\,\middle|\int_0^\infty dt\,\exp\{i[z-\tfrac{1}{2}(p^2+a^2)]t\}\left[\exp\left[i\frac{a}{\omega}\{p_1\sin(\omega t)+p_2[1-\cos(\omega t)]\}\right]-1\right]\middle|\vec{0}\right\rangle\\&=-i(2\pi)^{-3}\int d\vec{p}\int_0^\infty dt\,\exp[-\tfrac{1}{2}i(p^2+\hat{k}^2)]t\left[\exp\left[i\frac{a}{\omega}\{p_1\sin(\omega t)+p_2[1-\cos(\omega t)]\}\right]-1\right]\\&=-i(2\pi)^{-2}\int_{-\infty}^{+\infty}dp\,p^2\int_0^\infty dt\,\exp[-\tfrac{1}{2}i(p^2+\hat{k}^2)t]\left\{\left[\frac{2ap}{\omega}\sin\left[\frac{\omega t}{2}\right]\right]^{-1}\sin\left[\frac{2ap}{\omega}\sin\left[\frac{\omega t}{2}\right]\right]-1\right\}\\&=-i(2\pi)^{-2}\int_{-\infty}^{+\infty}dp\,p^2\int_0^\infty dt\,\exp(-\zeta t)\left\{\left[\alpha\sin\left[\frac{\omega t}{2}\right]\right]^{-1}\sin\left[\alpha\sin\left[\frac{\omega t}{2}\right]\right]-1\right\}\\&=-i(2\pi)^{-2}\frac{2}{\omega}\int_{-\infty}^{+\infty}dp\,p^2\left[1-\exp\left[-\frac{2\pi\zeta}{\omega}\right]\right]^{-1}\int_0^\pi ds\,\exp(-2\zeta s/\omega)[(\alpha\sin s)^{-1}\sin(\alpha\sin s)-1]\ ,\end{aligned} \tag{4.22}$$

where $\zeta=i\frac{1}{2}(p^2+\hat{k}^2)$ and $\alpha=2ap/\omega$. With $f(s)=(\alpha\sin s)^{-1}\sin(\alpha\sin s)-1$ we find after a few partial integrations that

$$\int_0^\pi ds\,\exp\left[-\frac{2\zeta s}{\omega}\right]f(s)=\frac{\omega a^2p^2}{3\zeta^3}\left[\exp\left[-\frac{2\pi\zeta}{\omega}\right]-1\right]+\left[\frac{\omega}{2\zeta}\right]^4\int_0^\pi ds\,\exp\left[-\frac{2\zeta s}{\omega}\right](\partial_s^4f)(s)\ , \tag{4.23}$$

so that

$$\begin{aligned}D_1=i(2\pi)^{-2}\tfrac{2}{3}a^2\int_{-\infty}^{+\infty}dp\,p^4/\zeta^3-i(2\pi)^{-2}\left[\frac{\omega}{2}\right]^3\int_{-\infty}^{+\infty}dp\,p^2\left[1-\exp\left[-\frac{2\pi\zeta}{\omega}\right]\right]^{-1}\zeta^{-4}\\\times\int_0^\pi ds\,\exp\left[-\frac{2\pi\zeta s}{\omega}\right](\partial_s^4f)(s)\ .\end{aligned} \tag{4.24}$$

The integral over s in (4.24) can in absolute value be bounded by p^2 times a positive number, depending on z but not on p and which is finite for finite z. It thus follows that (4.24) is finite and, moreover, can be continued analytically to z in the negative half-plane in an infinite number of ways, the points $\hat{k}^2=n\omega$, $n\in\mathbb{Z}$ being the branch points. We can cast D_1 in a different form by noting that for $\text{Re}\zeta>0$

$$\int_0^\infty dt \exp(-\zeta t)\left[\sin\left[\alpha\sin\frac{\omega t}{2}\right]\Big/\left[\alpha\sin\frac{\omega t}{2}\right]-1\right]$$

$$=(2/\omega)\int_0^\infty ds \exp(-2\zeta s/\omega)\sum_{n=1}^{\infty}(-1)^n(\alpha\sin s)^{2n}/(2n+1)!$$

$$=\zeta^{-1}\sum_{n=1}^{\infty}(-1)^n(2n+1)^{-1}(\alpha\omega/2)^{2n}(\zeta^2+\omega^2)^{-1}\cdots(\zeta^2+n^2\omega^2)^{-1}, \tag{4.25}$$

so that

$$D_1=-(2\pi^2)^{-1}\sum_{n=1}^{\infty}\frac{(2a)^{2n}}{2n+1}\int_{-\infty}^{+\infty}dp\frac{p^{2n+2}}{p^2+\hat{k}^2}[(p^2+\hat{k}^2)^2-4\omega^2]^{-1}\cdots[(p^2+\hat{k}^2)^2-4n^2\omega^2]^{-1}, \tag{4.26}$$

where the possibility of analytic continuation is also evident. Equations (4.26) and (4.19) are a convenient starting point for the calculation of $\epsilon(\vec{a})$. [The analytic properties of $\epsilon(\vec{a})$ are discussed in Ref. 28.] By summarizing our results we have for the case of zero-range potential

$$[z-H(\zeta)]^{-1}=[z-H_0(\zeta)]^{-1}-D^{-1}(z)[z-H_0(\zeta)]^{-1}\,|\,\vec{0}\rangle\langle\vec{0}\,|\,[z-H_0(\zeta)]^{-1}, \tag{4.27}$$

$$\Phi_{lm}(k)=D^{-1}(z)\langle k,l,m\,|\exp(i\vec{h}\cdot\vec{p})\,|\,\vec{0}\rangle\langle\vec{0}\,|\,(z-H_0)^{-1}\,|\,\phi_0\rangle . \tag{4.28}$$

We note further that in the approximate expression (4.13) we now have (j_l is a spherical Bessel function)

$$\langle k,l,m\,|\exp(i\vec{h}\cdot\vec{p})\,|\,\phi_\delta(\vec{a})\rangle\to\langle k,l,m\,|\exp(i\vec{h}\cdot\vec{p})\,|\,\vec{0}\rangle \quad\text{as } \delta\searrow 0$$

$$=\langle k,l,m\,|\,\vec{h}\rangle=(2\pi)^{-3/2}4\pi k(-i)^l j_l(kh)Y_l^m(\Omega_{\vec{h}}) . \tag{4.29}$$

It follows from (4.27) that $H(\zeta)=H(\zeta,\vec{a})$ is dilatation analytic. In addition, $H(\zeta,\vec{a})$ and $H(\zeta,-\vec{a})$ are unitarily equivalent and $H(i\pi,\vec{a})=H(0,-\vec{a})$ is self-adjoint. In concluding the formal part of this section we finally note that for a general $|\phi\rangle\in\mathscr{H}$ we do not need dilatation analyticity, since it is in principle possible to calculate $\Phi_{lm}(k)$ from (4.10), for instance by means of (4.20) and (4.21).

V. NUMERICAL RESULTS FOR THE ZERO-RANGE POTENTIAL

In order to obtain numerical data on the area of the peaks in the photoelectron energy spectra we use the peaking (third) term from (3.15) together with (3.12), (4.13), and (4.29). This leads to

$$w_m=\sum_l\Gamma^{-1}\frac{\pi}{\sqrt{2}}\,|\,(2\pi)^{-3/2}4\pi k_m(-i)^l j_l(k_m h)Y_l^m(\Omega_{\vec{h}})\,|^2$$
$$\times|\,\epsilon_0(\vec{a})+m\omega-\tfrac{1}{2}a^2\,|^{-1}$$
$$\times\mathrm{Re}[\epsilon_0(\vec{a})+m\omega-\tfrac{1}{2}a^2]^{1/2} . \tag{5.1}$$

The ionization rates R_m are proportional to this, but normalized in a different way so that $\sum R_m=2\Gamma$ ($\sum w_m=1$ since in the limit $t\to\infty$ every atom eventually becomes ionized). Noting that k_m^2 cancels $|\,\epsilon_0(\vec{a})+m\omega-\frac{1}{2}a^2\,|^{-1}$, and dropping all m-independent factors, we get the proportionality ($\vec{h}$ falls along the x_2 axis)

$$R_m\propto\sum_l[j_l(k_m h)Y_l^m(0,\pi/2)]^2\mathrm{Re}[\epsilon_0(\vec{a})+m\omega-\tfrac{1}{2}a^2]^{1/2} . \tag{5.2}$$

This equation can be summed easily since the $j_l(k_m h)$ decrease rapidly once l becomes large with respect to $k_m h$. k_m is calculated from $\epsilon_0(\vec{a})$ given by (4.11), which in the zero-range limit (4.18) leads to

$$D(z)=0 . \tag{5.3}$$

This implicit equation is solved by means of the secant-Newton method, where the function D is calculated according to (4.19) and (4.26). The integration in the latter is performed by summing all the relevant pole contributions. The way of analytic continuation is chosen such that for $\vec{a}\to\vec{0}$ the energy converges to ϵ_0, i.e., only those poles are included which are in the correct half plane in the $\vec{a}=\vec{0}$ case, and these poles are retained during the entire calculation, no matter where they are shifted to.

The result for the case with $\omega=0.24$ can be seen in Fig. 4, where the various R_m are plotted as a function of field strength $\vec{a}$. The minimum number of photons needed to overcome the ionization potential ($\epsilon_0=-1$) at $\vec{a}=\vec{0}$ is five. For small $\vec{a}$ we note that the R_m have an intensity dependence that can be described by a simple power law, the exponent being the one expected from perturbation theory. The real part of $\epsilon_0(\vec{a})$ is plotted in Fig. 5, where it can be seen that the shift in ionization potential is proportional to a^2 at low $\vec{a}$. This increase of ionization potential eventually leads to the collapse of R_5, and, later on, R_6, as can be seen from the dips in the corresponding curves. The values of $\vec{a}$ at which the ionization potential is exactly equal to m times the photon energy (as can be seen from Fig. 5) are marked along the horizontal axis in Fig. 4. The behavior on the low-field side of these points is dominated by the factor in (5.2) containing the real part, and therefore drops very rapidly as $\vec{a}$ approaches the

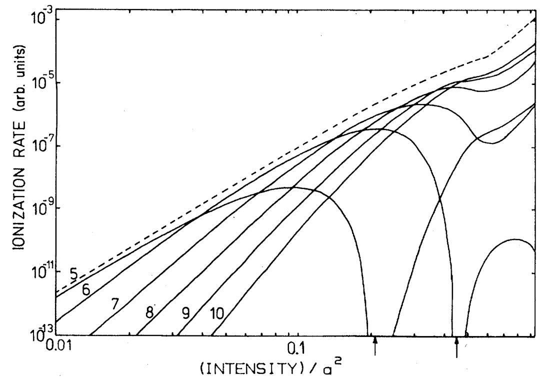

FIG. 4. Rates for absorption of various number of photons by the zero-range model atom, as a function of the field strength a. The minimum number of photons necessary to ionize is 5 in this case. Note the minimum in the rates at the points (marked by arrows) where the argument of the square root in (5.2) acquires a negative real part.

critical value. After going through a minimum, however, this factor increases again as the imaginary part Γ of $\epsilon_0(\vec{a})$ (and therefore the real part of the square root) grows larger. Since the order of the intensity dependence of Γ is much larger than that of the real part, the decrease of R_m accompanying the growing negativeness of the square-root argument is more than offset. Note that at the high-field side of the disappearance points the $F_{lm}(k)$ no longer has a peak at real k. The integration leading to (3.15) is in this case entirely over a tail of the lifetime broadened profile of the resonance, so that the considerations leading to the choice of the third term of (3.15) as the physically interesting one, in fact, become invalid. Although the increase of the R_m seems rather dramatic on the logarithmic scale of Fig. 4, the total contribution of these "subthreshold" electrons is always very small. The behavior of R_7 becomes understandable from Fig. 5 also; after a

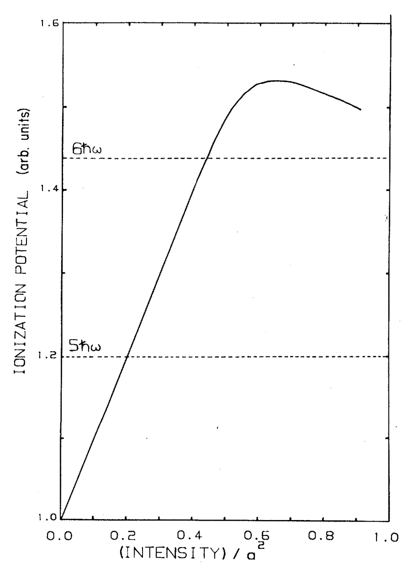

FIG. 5. Real part of the ionization potential of zero-range model atom as a function of the field strength a. The dashed lines indicate the thresholds for 5- and 6-photon ionization.

decrease caused by the approach of the 7-photon threshold the ac-Stark shift of the ground state (which lowers the energy if the first photon does not bring us above any excited states) reverses sign, and eventually grows larger than the continuum shift $\frac{1}{2}a^2$, thereby increasing R_7 again. For a Coulomb potential the cross section has a finite value immediately above the threshold, so in real atoms the collapse of the R_m might be more abrupt.

As a final remark we draw attention to the way in which the total ionization rate 2Γ depends on $\vec{a}$. For low $\vec{a}$ the ionization process is dominated by R_5, as expected from perturbation theory, and the order of nonlinearity of the ionization process is therefore equal to five. As the various peaks disappear, however, the higher R_m start to dominate the electron spectrum, but the order of nonlinearity for their total stays more or less the same. This is not at all what is expected from perturbation theory, and the fact that the order of nonlinearity for the photoionization of Xe by the yttrium-aluminum-garnet—laser turns out to be[26] 11 even in an intensity region where 11-photon ionization is completely negligible compared to the total ionization rate,[5] suggests that a similar result holds for long-range potentials.

VI. DISCUSSION

In the preceding sections we obtained a reasonably simple description of the energy spectrum of the ejected electron in a multiphoton ionization experiment with a circularly polarized field. The disappearance of the first electron peak with increased intensity is accounted for and it is seen, for instance from (3.12), that the broadening of the electron peaks is basically due to the dynamic Stark broadening of the ground state [through $\epsilon(\vec{a})$]. (In experiments employing very short laser pulses an additional source of broadening occurs due to the finite duration of the pulse, because then the intensity at which the electron leaves the laser focus might be different from the one at which it was created, so ponderomotive force and ionization potential shift no longer cancel.) In case the continuum is reached through an intermediate resonant or nearly resonant state, our formalism can readily be adapted by increasing the projector P in Sec. III. Under these circumstances the peak width will depend on the ac-Stark width of all states involved. The general picture will not change in the case of an n-electron atom. We did not consider that case here since there still exists a gap in the underlying mathematics (see Ref. 8, discussion section) so that we do not know whether or not the corresponding Floquet Hamiltonian is dilatation analytic. In the n-electron case the term $-\omega l_3$ in H^{Fl} is replaced by $-\omega J_3$, $\vec{J}$ being the total angular momentum vector (including spin). If we put a cutoff on this operator, i.e.,

$$-\omega J_3 = -\sum_{n=-\infty}^{+\infty} m\omega P_m \to -\sum_{m=-\infty}^{M} m\omega P_m ,$$

M finite, the corresponding Floquet Hamiltonian is dilatation analytic, whereas the physical consequences of the cutoff are minimal for M sufficiently large. This would allow us to discuss the consequences of the fact that the ionization threshold becomes complex in an n-electron

$(n \geq 2)$ atom (since the ionic ground state is ac-Stark broadened). In connection with the above cutoff it would be interesting if we could show that the amplitude Φ (the generalization of Φ in Sec. III) can be written as the sum of the amplitude, containing the resolvent of the cutoff Hamiltonian, and a second term which can be chosen to be arbitrarily small. Since this can be done before the dilatation it can probably be justified on the basis of strong resolvent convergence. It should be noted that an appropriate description of an n-electron, $n \geq 2$, atom through a real or complex effective potential, that vanishes at large distances, never gives rise to a complex ionization threshold (Weyl's theorem on the invariance of the essential spectrum of an operator under relatively compact perturbations).

In actual experiments the field polarization is usually linear. It is possible to treat this case as well, for instance by Howland's[10] approach. The trade-off is that we have to work with an enlarged Hilbert space leading to a more involved bookkeeping. The final formulas will be of the type encountered in Sec. III, the basic difference being a change in the "selection rules." An essentially equivalent description can also be obtained by means of the second quantized formalism of Grossmann and Tip.[29] This formalism is in fact valid for any many-electron atom, due to the semiboundedness of the Hamiltonian. The results of Sec. IV are specific for the circularly polarized case, since the rank of the potential becomes infinite dimensional in the enlarged space mentioned above.

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