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Cross sections for the multiphoton ionization of sodium in a linearly polarized radiation field

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A calculation of the N-photon ionization cross sections with $N=2,3,4,5$ in the frequency interval $0.098<\omega<0.165$ (atomic units) for sodium has been performed using a procedure described by Pan [J. Mod. Opt. **36**, 877 (1989)]. The final-state wave function represents the Volkov-like continuum, distorted by the atomic potential, while the initial state is described by the rotating-wave approximation with the inclusion of the ground-state 3s and two excited p states. The cross sections were calculated for two kinds of initial conditions: instantaneous and adiabatic switching of radiation. The results of the calculation with the distorted continuum wave functions representing the final state were compared to those obtained from Coulomb and plane-wave functions.

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I. INTRODUCTION

The development of intense ultra-short-pulse lasers has provided a tool to probe atoms under conditions where the electric-field strength associated with the laser pulse can be comparable to atomic field strengths. Exposure to such large time-dependent electric fields modifies both atomic structure and atomic processes. One such process of considerable importance and interest is multiphoton ionization. A number of theoretical and experimental results [1,2] indicate the richness of the physics and the diversity of the experimental observations, including above-threshold ionization and harmonic generation. Our interest is in the development of a simpler more applicable expression for the multiphoton ionization cross section applied initially to simple atomic systems but extended eventually to more complex atoms.

In the present treatment multiphoton ionization is regarded as a process in which the electron makes a transition between the initial bound state of a generally complex atom and a final continuum state. The final state is represented by an approximate solution of the Schrödinger equation for a free electron in both the radiation field and the field of the ionized atom. Our methodology therefore belongs to a broad category of Keldysh-like models. A majority of these models uses plane-wave Volkov states for the description of the final state. Several authors tried to improve the original Keldysh procedure and replaced the plane-wave final state by the Coulomb-Volkov waves. References are given in the paper by Trombetta, Basile, and Ferrante [3], who discuss and compare the results for the ionization of hydrogen. In the short-wavelength region, the Keldysh approximation appears to overestimate the ionization cross sections, while the results from the Coulomb-Volkov waves are in good agreement with the perturbation theory calculations. For larger wavelengths, both approximations severely underestimate the cross sections due to the neglect of excited bound states. Using a different approach, Pan [4] derived another form of the Coulomb-Volkov waves based on the continuum-continuum coupling. In the case of complex atoms, the potential for the photoelectron should be further modified by the screening effect of the bound electrons, and this effect may be conveniently incorporated into Pan's formalism.

The aim of this work is to develop a method for the evaluation of photoionization cross sections using a procedure similar to Pan's but extended to include the effect of excited states, and to assess the importance of screening by atomic electrons. In our approach, the final state is described by a wave function that already includes the effect of the non-Coulombic atomic potential. In addition, the initial-state wave function used for the calculation of the transition matrix element represents a time evolution of the atom under the influence of the radiation field and contains several bound states. As in all Keldysh-type models, continuum states can be regarded as dressed by photons and the resulting energy sidebands can be excited by one-photon transitions from the lower bound states.

We study the absorption of light by an electron outside closed shells, and we shall assume that in the absence of a radiation field both the bound and the free electron move in the same central potential, and that the charge distribution of core electrons remains unaffected when the radiation field is applied. The electron spin will be ignored in this paper.

In the presence of a radiation field the nonrelativistic equation for the electron in the radiation gauge [without the quadratic term $\frac{1}{2}(A/c)^2$ in the Hamiltonian] is

$$i\frac{d\psi}{dt}=\left[\frac{1}{2}\vec{p}^{\,2}-\frac{1}{c}\vec{A}\cdot\vec{p}+V(r)\right]\psi\ . \tag{1}$$

All quantities in (1) and throughout the paper are given in atomic units ($e=m=h/2\pi=1$) unless explicitly indicated. $\vec{A}$ is the vector potential of the radiation field and V is the potential energy of the electron in the field of the ion. We investigate the case of a monochromatic and linearly polarized radiation field in the dipole approximation and set

$$A_x=0\ ,\quad A_y=0\ ,\quad A_z=A\sin\omega t\ . \tag{2}$$

Two types of initial conditions at time $t=0$ will be investigated: an instantaneous radiation switch-on when the atom is in the ground state (case A), and an adiabatic switch-on condition corresponding to a quasistationary state (case B).

First we derive the expression for a generalized N-photon ionization cross section based on a model of a three-level atom with levels n_1s,n_2p,n_3p. The calculations are then carried out for a sodium atom initially in the $3s$ ground state for photon energies around the $4p$ and $5p$ resonances and for $N=2$, 3, 4, and 5.

II. WAVE FUNCTIONS FOR THE FINAL AND INITIAL STATES

In the absence of the radiation field, the solution of (1) for a free electron with kinetic energy E is

$$\psi_{Elm}(\vec{r})e^{-iEt}=\sqrt{2/\pi}\,r^{-1}F_{El}(r)Y_{lm}e^{-iEt}\ , \tag{3}$$

with

$$F_{El}(r)\underset{r\to\infty}{\sim}p^{-1/2}\sin\left[pr-\frac{\pi l}{2}+Zp^{-1}\ln 2pr+\eta_l\right]\ ,$$
$$p^2=2E\ , \tag{4}$$
$$\eta_l=\arg\Gamma(l+1-iZ/p)+\eta_l^0\ .$$

Z is the asymptotic charge for the free electron and the functions ψ_{Elm} are energy normalized. The phase shift η_l^0 is due to the short-range part of the potential V.

A general solution of (1) corresponding to a free electron may be written in terms of ψ_{Elm} in the form

$$\Psi_E=\sum_{lm}\int a_{E'lm}(t)\psi_{E'lm}e^{-iE't}dE'\ , \tag{5}$$

and coefficients $a_{E'lm}$ may be found by substituting (5) into (1) and performing the integration over E'. A detailed account of this procedure was provided by Pan [4]. Since the ground state is an s state with $m=0$, then for a linearly polarized radiation field in the z direction, only states with $m=0$ can be coupled to the ground state. Therefore in (5) we may set $m=0$ without loss of generality and omit the subscript $m=0$ in the remainder of the paper. The radiation field causes mixing of ψ_{Elm} with different l and E'. However, the dominant term is produce by "on shell" coupling and we will ignore contributions to Ψ_E corresponding to $E'\neq E$. Such contributions may nevertheless become important near the ionization threshold when E approaches zero, as pointed out by Trombetta, Basile, and Ferrante [3].

In our procedure to find expansion coefficients a_{El} we follow the method of Pan [4]. It requires the evaluation of the free-free dipole matrix elements $\langle\psi_{El}|\partial/\partial z|\psi_{E'l\pm1}\rangle$, which are singular at $E'=E$ and may be written as

$$\left\langle\psi_{El}\left|\frac{\partial}{\partial z}\right|\psi_{E'l\pm1}\right\rangle=\frac{2}{\pi}l_>[(2l_>-1)(2l_>+1)]^{-1/2}\times\int F_{El}\left[\frac{d}{dr}\pm\frac{l_>}{r}\right]F_{E'l\pm1}dr\ , \tag{6}$$

where $l_>=l+1$ for the upper sign and $l_>=l$ for the lower sign, respectively. A general formula for this matrix element was derived by Korol [5]. We use Korol's equation 13 to evaluate the element (6). There are three types of singular terms in the expression for the dipole matrix element: a term with the δ function $\delta(E'-E)$, a term proportional to $(E'-E)^{-1}$, and a term containing $\ln|E'-E|$. The contribution of the logarithmic singular term is negligible in the narrow region around $E'=E$ and consequently, the matrix element in the limit $E'\to E$ may be expressed in the form

$$\left\langle\psi_{El}\left|\frac{\partial}{\partial z}\right|\psi_{E'l\pm1}\right\rangle=\pm l_>[(2l_>-1)(2l_>+1)]^{-1/2}\times\left[p'\cos(\eta'_{l\pm1}-\eta_l)\delta(E'-E)-\frac{p'+p}{2\pi(E'-E)}\sin(\eta'_{l\pm1}-\eta_l)\right]\ , \tag{7}$$

where the phase shifts $\eta'_{l\pm1}$ and η_l are associated with the asymptotic forms of $F_{E'l\pm1}$ and F_{El} according to (4).

The importance of the δ-function term in the free-free dipole matrix element has only been realized in recent years [6–8]. In Pan's [4] expression (B17), which is applicable to continuum-continuum coupling of Coulomb partial waves with $Z=1$, the δ-function term is missing and the first term in her expression corresponds to the second term of our Eq. (7).

By integrating (7) over E' and taking only the residue of the second integral of the right-hand side, we obtain

$$\int\left\langle\psi_{El}\left|\frac{\partial}{\partial z}\right|\psi_{E'l\pm1}\right\rangle dE'=\pm pl_>[(2l_>-1)(2l_>+1)]^{-1/2}\times\exp[-i(\eta_{l\pm1}-\eta_l)]\ . \tag{8}$$

If we define a real symmetric tridiagonal matrix $\mathbf{R}$ such that

$$R_{l,l-1}=R_{l-1,l}=l[(2l-1)(2l+1)]^{-1/2}\ ,$$

then from (8) and (1) we obtain a system of coupled equations for the coefficients a_{El}:

$$\dot{a}_{El}=\frac{pA}{c}\{-R_{l,l-1}a_{El-1}\exp[-i(\eta_{l-1}-\eta_l)]+R_{l,l+1}a_{El+1}\exp[-i(\eta_{l+1}-\eta_l)]\}\sin\omega t\ . \tag{9}$$

The solution may be written in the form

$$a_{El\lambda}=b_{l\lambda}i^{l}\exp\left[i\left[\eta_l-\frac{pA}{\omega c}\xi_\lambda\cos\omega t\right]\right]$$
$$\lambda=0,1,2,\ldots, \tag{10}$$

where $b_{l\lambda}$ are elements of matrix $\mathbf{b}$ such that $\mathbf{b}^{-1}\mathbf{R}\mathbf{b}$ is a diagonal matrix with diagonal elements ξ_λ and $\sum_l b_{l\lambda}^2=1$. $\mathbf{R}$ and $\mathbf{b}$ are infinite matrices and each λ corresponds to an independent approximate solution of (1) for a free electron (with $m=0$) in the form

$$\Psi_{E\lambda}=\sum_{l=0}^{\infty}a_{El\lambda}\psi_l(\vec{r})e^{-iEt}\ . \tag{11}$$

If the electric field associated with the incident radiation is smaller than atomic field intensities, then the time-dependent wave functions of the valence electron may be conveniently expressed in terms of unperturbed functions φ_j. For atomic systems with one electron outside of a closed shell, we limit the expansion to three states, namely the ground state n_1s and two excited states n_2p and n_3p with $m=0$, which directly couple to the ground state by dipole interaction. Solutions of (1) with $\vec{A}=\vec{0}$ are $\varphi_j(\vec{r})\exp(-iE_jt)$, and we set

$$\varphi_1\equiv n_1s\ ,\quad \varphi_2\equiv n_2p\ ,\quad \varphi_3\equiv n_3p\ . \tag{12}$$

The response of a three-level atom to the radiation field was studied by several authors (see Shore, Ref. [9]). More recently, Jönsson [10] presented a diagrammatic solution to the three-level problem. We employ the rotating-wave approximation to solve Eq. (1) for the bound electron. The general solution then takes the form

$$\Phi(t)=\sum_{k=1}^{3}[C_{1k}\varphi_1+(C_{2k}\varphi_2+C_{3k}\varphi_3)\exp(-i\omega t)]$$
$$\times\exp[-i(E_1-\alpha_k)t]\ . \tag{13}$$

The energy parameters α_k satisfy the equation

$$\begin{vmatrix}\alpha_k & W_a & W_c\\ W_a & \alpha_k+E_2-E_1-\omega & 0\\ W_c & 0 & \alpha_k+E_3-E_1-\omega\end{vmatrix}=0\ , \tag{14}$$

with W_a and W_c given by

$$W_a=\frac{1}{2}\left\langle\varphi_1\left|\frac{A}{c}\frac{\partial}{\partial z}\right|\varphi_2\right\rangle\ ,\quad W_c=\frac{1}{2}\left\langle\varphi_1\left|\frac{A}{c}\frac{\partial}{\partial z}\right|\varphi_3\right\rangle\ . \tag{15}$$

For a linearly polarized wave in the z direction, both W_a and W_c are real. The expansion coefficients C_{jk} may be written as $C_{jk}=C'_{jk}K_k$. The ratio $C'_{1k}:C'_{2k}:C'_{3k}$ for each k is determined by the value of α_k, and the coefficients K_k depend on specific boundary conditions.

We investigate two possibilities: in case A, the atom at $t=0$ is in the ground state so that $\Phi(t=0)=\varphi_1(\vec{r})$ and the radiation field is switched on instantaneously. In this case, generally none of the K_k values is zero. In case B the sum over k in (13) has only one term with $k=k_0$ and $\Phi(t=0)=\sum_j C_{jk}\varphi_j$. The probability of finding the atom in any of the three states is time independent; the atom has adjusted to the radiation field. This situation corresponds to the adiabatic switch-on condition. For each value of the radiation intensity and frequency ω we choose k_0 such that it is associated with the largest probability of finding the atom in the ground state.

In this paper we have applied our method to the ionization of an isolated sodium atom. The central potential $V(r)$ has been constructed using wave functions of Clementi and Roetti [11] for the core electrons and a Slater-type [12] exchange-correlation potential with an adjustable factor F. The $3s$ radial wave function was then self-consistently generated and F determined from the condition that the eigenvalue $E_1=E_{3s}$ agrees with the observed ionization energy of sodium. The resulting potential V was used to obtain all functions φ_j and F_{El}. The calculated values of the excitation energies for the $3p$, $4p$, and $5p$ levels agree with the observed values to within 1%.

III. CALCULATION OF THE PHOTOIONIZATION CROSS SECTIONS

The total cross section for the photoionizing transition from the initial state to the continuum states in which the ejected electron is described by wave functions $\Psi_{E\lambda}$ is the sum of partial cross sections corresponding to all values of λ for a given energy E. In analogy to the first-order perturbation theory, the transition-matrix element T_λ connecting the initial and final states of the ionization process is taken as

$$T_\lambda=\left\langle\Psi_{E\lambda}\left|i\frac{A}{c}\sin\omega t\frac{\partial}{\partial z}\right|\Phi(t)\right\rangle\ . \tag{16}$$

However, as the functions $\Psi_{E\lambda}$ and $\Phi(t)$ already contain the effects of the radiation field, there will be contributions to (16) corresponding to higher orders of the vector potential $\vec{A}$. For convenience we introduce the following notation:

$$G_\lambda=-\frac{p\xi_\lambda}{c\omega}\ ,$$
$$X_N=2N(-i)^{N-1}(G_\lambda A)^{-1}J_N(G_\lambda A)\ , \tag{17}$$

where J_N is the Bessel function,

$$M_{j\lambda}=\left\langle\Psi_{E\lambda}\left|\frac{\partial}{\partial z}\right|\varphi_j\right\rangle\ , \tag{18}$$

$$\Theta_{kN\lambda}=\frac{A}{2c}[C_{1k}M_{1\lambda}X_N+(C_{2k}M_{2\lambda}+C_{3k}M_{3\lambda})X_{N-1}]\ , \tag{19}$$

$$\vartheta_{kN}=E-E_1-N\omega+\alpha_k\ . \tag{20}$$

In evaluating (16) we use the relationship

$$\exp(ix\cos\beta)=\sum_{\mu=-\infty}^{+\infty}i^{\mu}J_\mu(x)\exp(i\mu\beta)\ .$$

Then T_λ is reduced to a sum of terms with factors $\exp[(E-E_1-\mu\omega+\alpha_k)t]$ corresponding to different μ.

For each value of the exponent there are two terms in the expansion with $\mu=\mu_1$ and $\mu=\mu_2$ such that $\mu_1+1=\mu_2-1=N$, and the matrix element (16) can be cast in the form

$$T_\lambda=\sum_{kN}\Theta_{kN\lambda}e^{i\vartheta_{kN}t} . \tag{21}$$

The probability P of ejection of the bound electron into any of the $\Psi_{E\lambda}$ states during the time from $t=0$ to t_1 with kinetic energy in the interval dE is

$$P=\sum_\lambda \left| \int_0^{t_1} T_\lambda dt \right|^2 dE . \tag{22}$$

In (22) there are terms with factors ϑ_{kN}^{-2}, and for high values of t_1 their contributions are sharply peaked around the values of $E\equiv E_k$ for which $\vartheta_{kN}=0$. The contributions of other terms with factors $\vartheta_{kN}^{-1}\vartheta_{k'N'}^{-1}$ for which $kN\neq k'N'$ are small and may be neglected. If the radiation pulse is long enough, the contributions from different values of N and k are well separated in energy and each contribution may be calculated by taking the limit $t_1\to\infty$. N can be interpreted as the number of photons absorbed by the bound electron in the ionization process.

The rate of absorption of N photons with energy ω is then

$$W_N=\lim_{t_1\to\infty}\frac{1}{t_1}\sum_\lambda\int \left| \int_0^{t_1} T_\lambda dt \right|^2 dE =2\pi\sum_{k\lambda}|\Theta_{kN\lambda}|^2 \tag{23}$$

and the corresponding generalized ionization cross section [13] is given by

$$\sigma_N=W_NF_\omega^{-N} , \tag{24}$$

where F_ω is the photon flux. All quantities in (23) depending on E should be evaluated for values $E\equiv E_k=E_1+N\omega-\alpha_k$.

If $N\omega$ is close to the ionization energy of the atom, it may happen that the condition $E-E_1-N\omega+\alpha_k=0$ is satisfied by negative values of E for certain values of ω. In this case the ionization channel Nk is closed, but it may open by increasing ω.

For very short radiation pulses the formula (23) may not be generally applicable, because if t_1 is small, the contributions corresponding to each k and N are not limited to a very narrow interval of energies and they may overlap. This may happen if t_1 is comparable to the period of the radiation field, or at very low radiation intensity because then the difference of α_k values is small. If t_1 is the duration of the pulse and $\Delta\alpha_k$ the difference of two adjacent α_k values, the approximate condition of validity of (23) is

$$|\Delta\alpha_k|\rangle\pi t_1^{-1} . \tag{25}$$

If the intensity of radiation is small, so that $(G_\lambda A)^2/4(N+1)\ll 1$, only the first term in the power expansion of Bessel functions in(17) may be taken into account and from (24) we obtain the generalized ionization cross section in the form

$$\sigma_N=(2\pi)^{N+1}\omega^{-N}c^{N-2}[(N-1)!]^{-2} \times\sum_{k\lambda}|G_\lambda^{N-2}[-iG_\lambda M_{1\lambda}C_{1k} +2(N-1)A^{-1}(M_{2\lambda}C_{2k} +M_{3\lambda}C_{3k})]|^2 . \tag{26}$$

Values of G_λ and $M_{j\lambda}$ depend on energy E and should be taken at $E=E_k$. The term with C_{1k} represents a direct transition from the ground state n_1s to the continuum due to absorption of N photons. The terms with C_{2k} and C_{3k} correspond to the absorption of one photon accompanied by a transition to the n_2p or n_3p state, respectively, and to the subsequent absorption of $N-1$ photons and transition to the continuum. Contributions from the excited states n_2p and n_3p exhibit resonance behavior and are sharply peaked at photon energies ω equal or close to E_2-E_1 and E_3-E_1. In the region between the two resonances the cross section may be strongly affected by interference effects, as can be seen from (26). The widths of individual resonances in the cross section are determined by parameters W_a and W_c defined by (15). For A approaching zero and ω far away from any resonance, C_{1k} approaches unity and both C_{2k} and C_{3k} become proportional to A so that σ_N becomes independent of the field intensity and the N-photon absorption rate follows the power law $W_N\propto F_\omega^N$. Near the resonance, σ_N behaves like A^{-2}. When the field strength is so large that the Bessel function in (17) can no longer be approximated by the first term of the series expansion, deviations from the power law for the absorption rate will occur. Expression (26) is valid even if the radiation field is given by $A_z=A\sin(\omega t+\varepsilon)$ with arbitrary phase ε.

Our calculation of the multiphoton ionization has been done for photon energies ω from 0.098 to 0.165 (2.67 to 4.49 eV) and the generalized cross section σ_N for neutral sodium has been evaluated according to formula (26) for $N=2,3,4,5$ and for three values of the field strength $E_z=0.002$, 0.01, and 0.03. ($E_z=A\omega/c$ and the atomic unit of E_z corresponds to the field strength 5.14×10^9 V/cm and to the intensity of 3.5×10^{16} W/cm^2.) Values of $|\xi_\lambda|$ in our calculation are smaller than 1.0, and the inaccuracy caused by the replacement of Bessel functions by the first term of the expansion is less than 10% at $\omega=0.098$ and $E_z=0.03$, when it has its maximum value.

The coefficients C_{jk} depend only on ω and A, the phase shifts η_l and the dipole radial integrals connecting the bound states and continuum states are smoothly varying functions of E, and for each N they were calculated for five different energies E of the ejected electron and then interpolated for values of E associated with any given ω by the condition $E=E_1+N\omega-\alpha_k$. The summation over l in (5) was truncated at $l_{\max}=34$. No significant difference was found by comparing the result with calculations using $l_{\max}=20$.

In the interval of photon energies studied in this paper, there are two low levels that may be excited directly from the 3s ground state by dipole interaction with the radiation field, namely the 4p and 5p states. The excitation energy of the 3p level in sodium at 0.077 lies outside this interval, but the effect of the 3p excited state in this interval is not negligible due to the large value of the parameter W_a defined by (15) with $\varphi_2 \equiv 3p$, and near the $5p$ resonance it is even larger than the effect of the $4p$ level. Therefore, in order to account for resonances in the cross sections due to both the $4p$ and $5p$ levels using formula (26), which contains only two p levels, we performed two sets of calculations, one with $n_2p \equiv 3p$, $n_3p \equiv 4p$ for $\omega < 0.155$, and the other one with $n_2p \equiv 3p$, $n_3p \equiv 5p$ for $\omega > 0.155$. In the narrow region between the $4p$ and $5p$ resonances, the effects of all three levels $3p$, $4p$, and $5p$ are of the same order of magnitude and (26) is not applicable.

The energy difference of the $3p$ and $3d$ levels in sodium is equal to 0.0556 and consequently for $\omega < 0.098$ the effect of the $3d$ level which is not included in (26) becomes important. Therefore, the present calculation has not been extended to lower frequencies.

IV. NUMERICAL RESULTS AND DISCUSSION

The generalized cross section σ_N for the absorption of N photons calculated from the expression (26) for $N = 2,3,4,5$ and for three values of electric-field intensity E_Z of radiation is shown in Fig. 1 for case A (instantaneous switch-on) and in Fig. 2 for case B (adiabatic switch-on). In the narrow interval of frequencies between the $4p$ and $5p$ resonances, where the three-level formula (26) is not applicable, values of σ_N are not shown. For low values of E_Z, and ω not close to resonance, the cross sections become independent of the field strength.

In case A (Fig. 1), the widths of individual resonances increase with increasing field strength, the values of σ_N decrease, and eventually individual resonances merge into a continuous background. There is also a small redshift

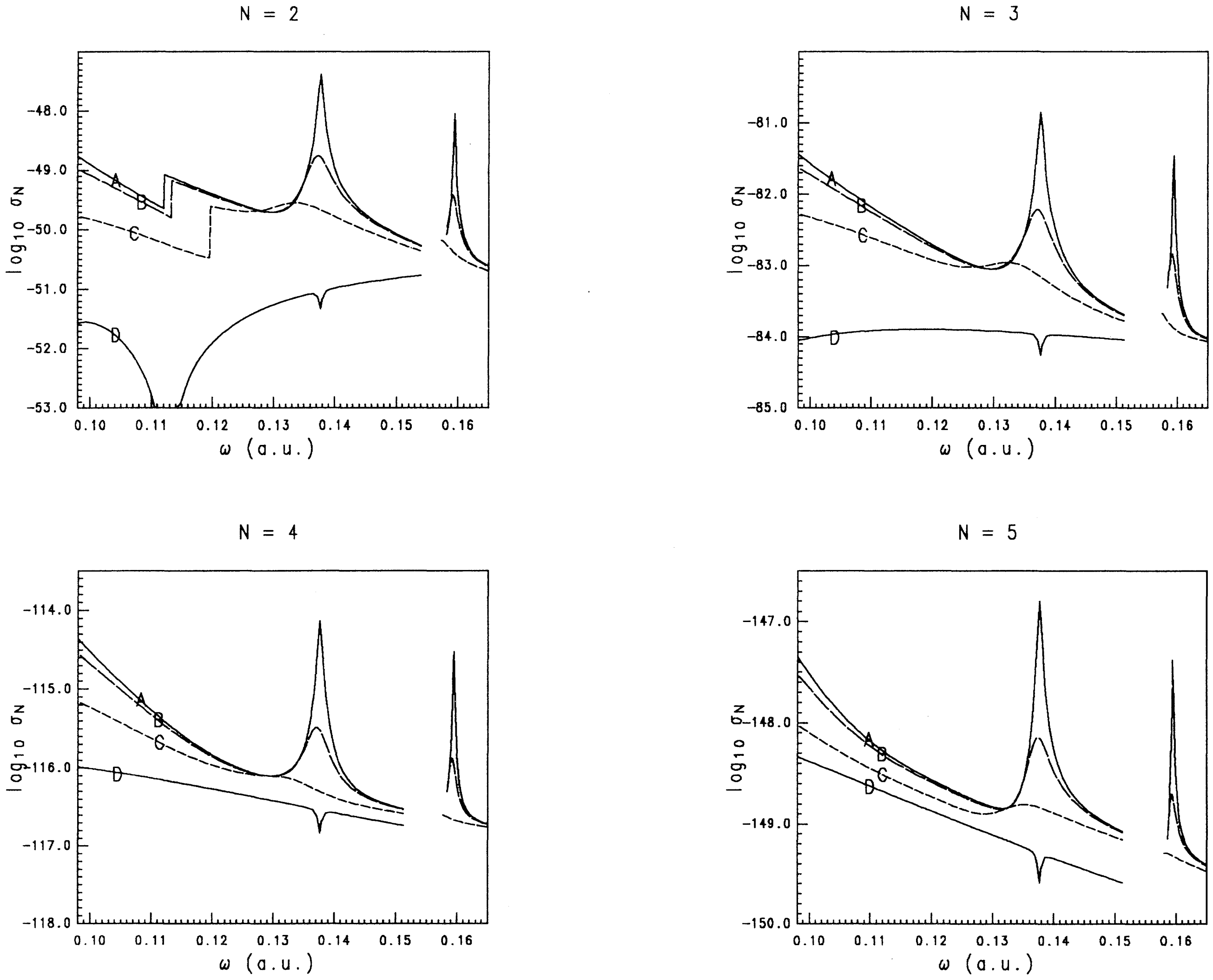

FIG. 1. Generalized N-photon ionization cross section σ_N (in $\mathrm{cm}^{2N}\mathrm{s}^{N-1}$) for sodium. Linearly polarized field, $E_z = 0.002$ (radiation intensity $= 1.4 \times 10^{11}$ W/cm^{-2}, curve A), $E_z = 0.01$ (curve B), $E_z = 0.03$ (curve C). $\omega =$ photon energy (atomic units). Instantaneous switch-on condition (case A). The peaks of the curve A at $\omega = 0.138$ and 0.159 coincide with the excitation energies of the $4p$ and $5p$ levels. Curve D represents the contribution from the direct $3s$-Ep transition at $E_z = 0.002$. (For $N = 2$, $D = 10\times$ this contribution.)

in the position of the peak value of each resonance associated with the increasing radiation intensity. A similar behavior at low intensities was found by Chu and Reinhardt [14] for hydrogen. The increase of σ_N towards small ω for all N is caused by the presence of the $3p$ resonance at $\omega=0.077$. According to (26), the total ionization cross section σ_N may be written as a sum of partial contributions σ_{Nk} corresponding to different roots α_k of Eq. (14) as described above. α_k and σ_{2k} for $E_Z=0.03$ are displayed in Fig. 3. In the energy spectrum of photoelectrons, each N is associated with three peaks whose position is given by $E=E_1+N\omega-\alpha_k$. For low field strength one of the α_k values is always close to zero. The sudden increase of the total two-photon cross section at small values of ω (Fig. 1) is a consequence of the opening of a new ionization channel corresponding to σ_{21}. The sharp minimum of σ_{23} at $\omega=0.126$ for $E_Z=0.03$ [Fig. 3(b)] is caused by zeros of the expression $C_{23}M_{2\lambda}+C_{33}M_{3\lambda}$ in formula (26) near this frequency. They appear at slightly different positions depending on λ. Curve D (Fig. 1) illustrates the relative importance of the term with C_{1k} in (26), which corresponds to the transition $3s$-Ep. The values of σ_N represented by curve D were obtained from (26) by ignoring terms with C_{2k} and C_{3k}. The minimum near $\omega=0.113$ for $N=2$ is caused by zero of the dipole matrix element connecting the $3s$ and Ep states, and a dip at $\omega=0.1376$ is a consequence of the decreased $3s$ population at the $4p$ resonance.

In case B (Fig. 2), the wave function (13) of the bound electron has only one term with $k=k_0$ corresponding to a particular value α_k, and in the energy spectrum of photoelectrons each N is associated with only one peak. The behavior of the total cross section σ_N is very similar to the behavior of the partial cross section σ_{N3} in case A for $\omega<\omega'$, and to σ_{N2} for $\omega>\omega'$. The frequency ω' corresponds to the situation when the probability of finding the atom in the ground state is the same for both $k=2$ and 3. At small values of E_Z, ω' practically coincides with the peak of a resonance and $\sigma_{N2}\approx\sigma_{N3}$ at ω'. However, with increasing E_Z this equality is no longer valid

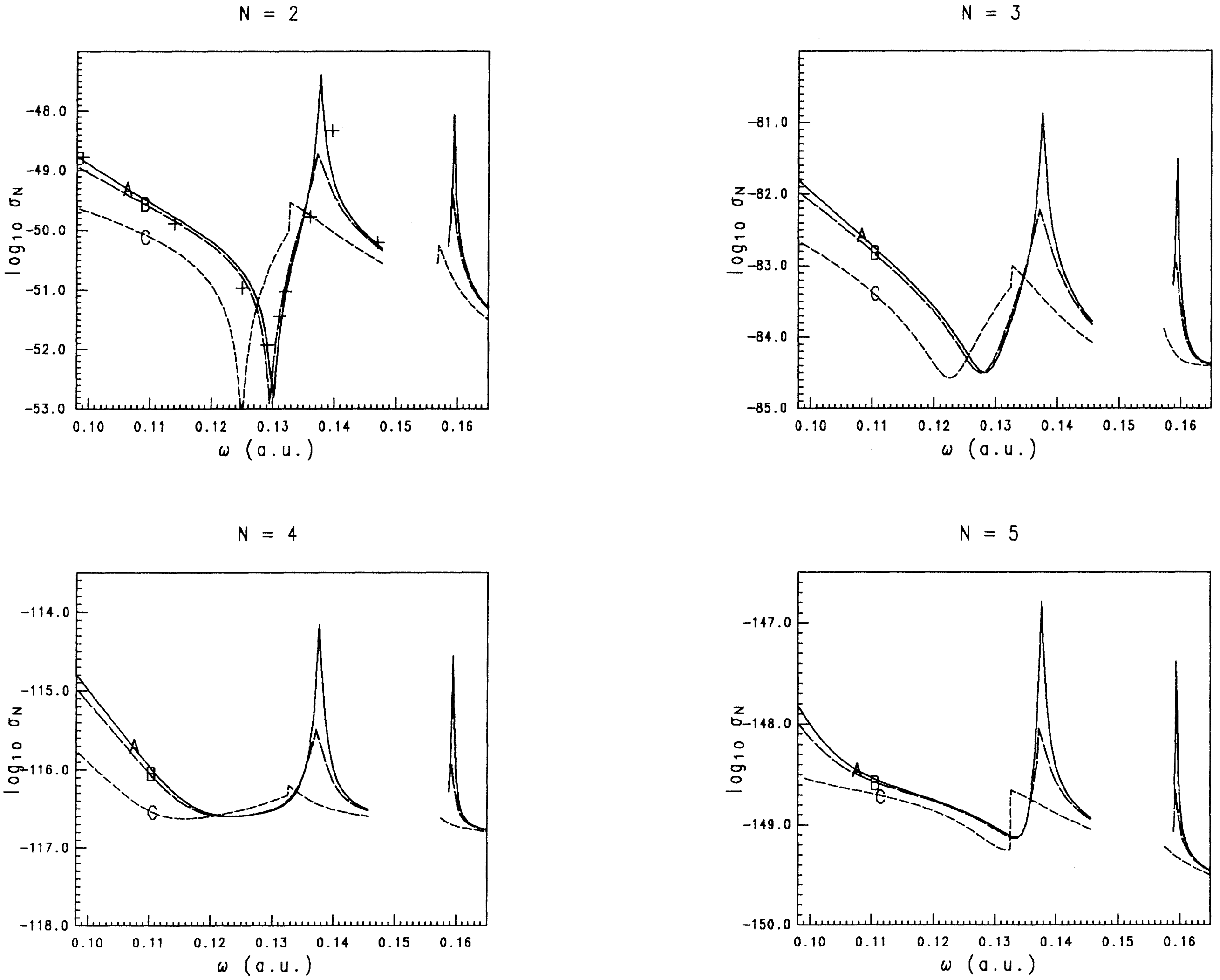

FIG. 2. Same as Fig. 1. Adiabatic switch-on condition (case B). Crosses: Mizuno [15] (limit of low radiation intensity).

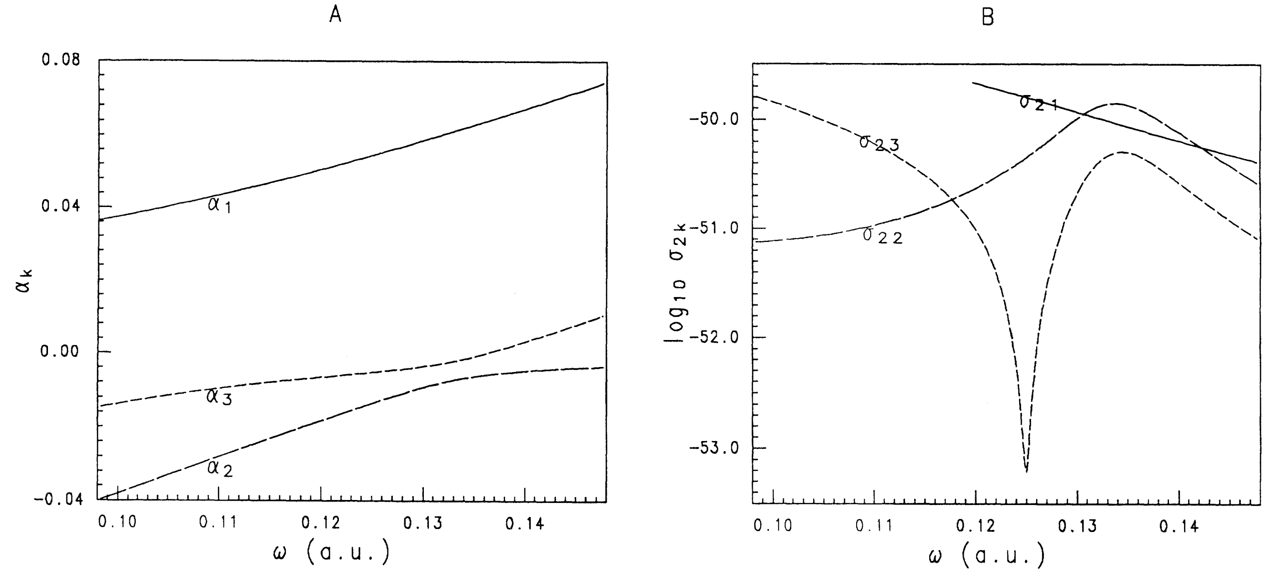

FIG. 3. Parameters α_k and partial cross sections σ_{Nk} (in $\text{cm}^{2N}\text{s}^{N-1}$) for $N=2$, $E_z=0.03$, case A (instantaneous switch-on).

and there is a discontinuity in the total σ_N in case B at ω'. This can be seen by comparing σ_{22} and σ_{23} in Fig. 3 with σ_2 for $E_Z=0.03$ from Fig. 2, where $\omega'=0.133$. The discontinuity is a consequence of our choice of initial conditions in case B, which applies to the idealized case of an isolated atom and coherent excitation. The deep minimum for $N=2$ near $\omega=0.013$ becomes shallower as N increases because of the larger spread of G_λ values so that zeros of the expression $M_{2\lambda}C_{23}+M_{3\lambda}C_{33}$ in (26) appear at different values of ω for different λ.

The adiabatic switch-on condition has been used by several authors for the calculation of σ_2 for sodium in linearly polarized light using various methods. Some of the values obtained by Mizuno [15] based on the perturbation theory are shown in Fig. 2. They should be compared to our curve A. There is an overall agreement between the two results. Similar results were found by McGuire [16], Bebb [17], and Manakov *et al.* [18], although the position of the minimum of σ_2 near $\omega=0.13$ does not always agree. This may be due to different electron wave functions used in the calculations. Consequently the uncertainty of the cross-section value for a particular photon energy in the region of steep increase with ω may be large, as is demonstrated in Table I.

As stated in Sec. I, in our procedure it was assumed that both the bound and the free electrons move in the same potential $V(r)$. For comparison we performed additional calculations in which the distorted wave functions F_{El} were replaced either by corresponding Coulomb wave functions or by functions representing plane waves. In both cases we used formula (26). For Coulomb wave functions we have $\eta_l=\arg\Gamma(l+1-iZp^{-1})$ and for plane waves $\eta_l=0$. The comparison is shown in Fig. 4 for cases A and B of initial conditions and for $N=2$, $E_z=0.002$. The value of σ_2 is only slightly increased by using Coulomb wave functions, except in the region of the minimum near $\omega=0.13$ in case B, but there is a large increase of σ_2 at large ω if plane waves are used for the description of the ejected electron (curve C). Note also a large contribution of the term, with C_{1k} in (26) representing the direct transition $3s$-Ep in the plane-wave calculations (curve D). In the region below $\omega=0.125$, the contributions of the $3s$-Ep and $3p$-El transitions have the tendency to mutually cancel, and the result is a sharp decrease of the total cross section (C). The difference between values of σ_N from distorted waves, Coulomb waves, and plane waves is expected to decrease with increasing N as the energy of the ejected electron increases.

There are several conditions that restrict the applicability of expression (26). First, the strength E_z of the radiation field should be small compared to atomic field strengths. Second, E_Z should be small enough that the omission of the $\frac{1}{2}(A/c)^2$ term in the Hamiltonian may be justified. Third, the field strength E_Z should satisfy the condition $(G_\lambda A)^2/4(N+1)\ll 1$, so that the Bessel functions in (17) may be replaced by the first term of the power-series expansion. For higher fields the cross section may be obtained again from (17), (18), (19), (23), and (24), keeping the expression for X_N in its original form (17). Fourth, the frequency ω should be such that the ratio $|(E_j-E_1+\omega)/(E_j-E_1-\omega)|$ for $j=2,3$ is much larger than 1, otherwise the rotating-wave approximation may not be valid. In our case, the minimum value of this ratio is 2.74 for $E_j=E_{3p}$ and $\omega=0.165$. Fifth, the duration t_1 of the radiation pulse has to be long enough, i.e., $t_1>\pi/\omega$ and $t_1>\pi/|\Delta\alpha_k|$ so that a pole approximation

TABLE I. Values of $\log_{10}\sigma_2$ (σ_2 in cm^4s) for $\omega=0.13125$ (second harmonic of the ruby laser).

Present result (case B)	Bebb [17]	Mizuno [15]	Manakov *et al.* [18]	McGuire [16]
−52.05	−52.22	−51.44	−51.26	−50.62

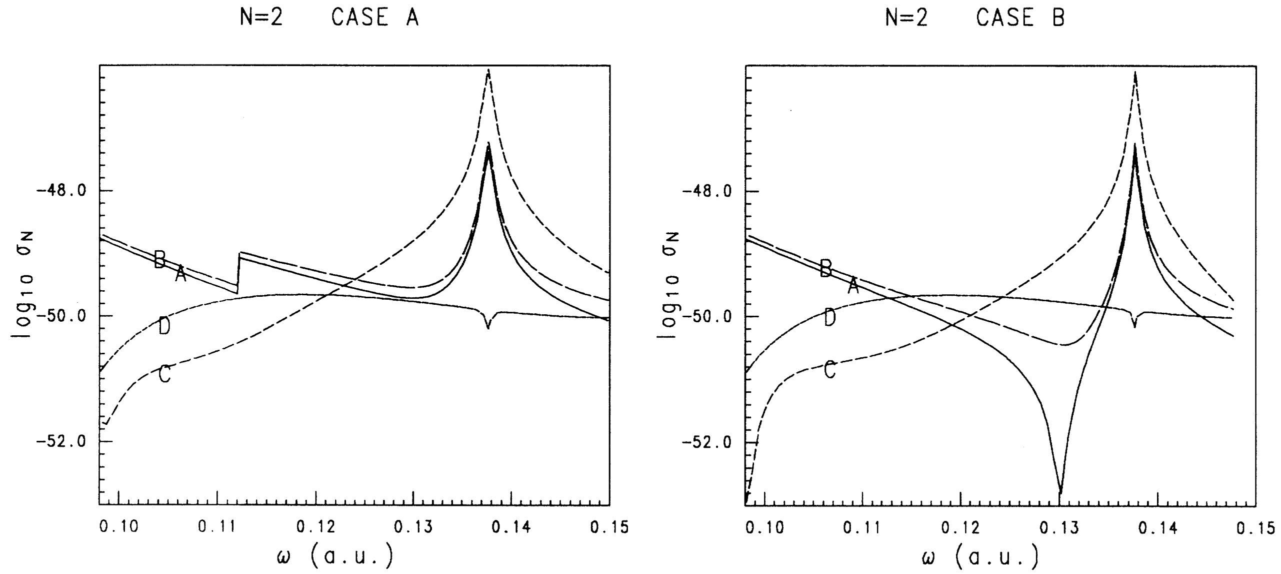

FIG. 4. Comparison of two-photon ionization cross sections calculated from distorted-wave functions (curve *A*), from Coulomb wave functions (curve *B*), and from plane-wave functions (curve *C*). The contributions from the direct 3*s*-*Ep* transition corresponding to calculations with plane-wave functions are shown as *D*. $E_z=0.002$. Case A: instantaneous switch-on. Case B: adiabatic switch-on.

may be used in evaluating separately individual contributions from different N and k in expression (22). For example, the smallest value of $|\alpha_2\text{-}\alpha_3|$ for $E_Z=0.002$, near the vicinity of the 4*p* resonance at $\omega=0.1376$, is 3.4×10^{-4}, which leads to the condition $t_1>0.22$ ps. For higher fields the values $|\Delta\alpha_k|$ increase and condition (25) becomes valid even for shorter pulses.

In summary, the present method represents a relatively simple way to calculate the multiphoton ionization, even in the frequency region where the ionization process is strongly influenced by the presence of excited levels. The procedure has been applied to sodium and it may be easily generalized to be applicable to complex atomic systems. The motion of the photoelectron in a non-Coulomb atomic potential is taken into account by representing the continuum wave functions in terms of distorted waves. The time evolution of the initial state is described by the rotating-wave approximation, including two excited levels, and the cross sections σ_N given by (26) also contain higher than N-order contributions. Two kinds of initial conditions have been investigated, and the results are compared with calculations performed with Coulomb waves and plane waves. Cross sections obtained from distorted-wave functions are generally lower than those from the other two methods. The application of the present method is limited by the validity of the rotating-wave approximation, intensity of radiation, and by the duration of the pulse. For larger values of field intensity E_Z, when (26) may no longer be valid, a more general expression (24) together with (23) and (19) should be used.

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