Multiphoton ionization of highly excited atomic states

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A quasiclassical approximation is used to obtain simple expressions for cross sections of N-photon ionization of highly excited atomic states. The corrections to these cross sections, which improve their behavior at interresonance minima, are found for the two-photon ionization case. A practically complete agreement between quasiclassical cross sections and corresponding quantum-mechanical values is obtained for the principal quantum number $n = 8$. This agreement is satisfactory even for the ground state of the hydrogen atom.

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Experimental results obtained on multiphoton ionization of atoms are usually explained on the basis of a quantum-mechanical theory of higher order perturbations.[1–3] In the case of ionization of the ground state of the hydrogen atom the calculations have been carried out including even the sixteenth order of perturbation theory.[4] However, the transition from the N-photon to the $(N + 1)$-photon ionization calculations always involves an additional integration (or summation) and the number of matrix elements that have to be calculated rises very rapidly on increase in N and also on transition from the ground state to excited levels. Additional mathematical difficulties arise in calculation of the cross sections of $(N + k)$-photon ionization above the N-photon ionization threshold.[5,6] Recent experimental results[7,8] indicate that up to seven additional photons can be absorbed above the five-photon ionization threshold of Xe. In the case of the experiments carried out by Bayfield's group[9,10] on the ionization of excited states of the hydrogen atom with the principal quantum number n of the order of 50 and with N amounting to a few hundreds, a quantum-mechanical perturbation theory can hardly be applied. Obviously, some simplifying assumptions are needed. Since highly excited states of atoms are hydrogen-like and also since in the case of multiphoton ionization of atoms the energy of the initial state as well as the energies of the intermediate and final states are less (in absolute value) than the energy on the first Bohr orbit, it seems natural to apply a quasiclassical approach. The idea was put partly into practice in Refs. 11 and 12, where the motion of an electron is described simply by classical equations and simple expressions are obtained for the cross sections of N-photon ionization, the expressions being generalizations of the well-known Kramers formula for the usual photoelectric effect. In the case of two-photon ionization above the one-photon ionization threshold the semiclassical cross sections are accurate to within a few percent. However, the cross sections obtained by such calculations are, strictly speaking, valid only above the one-photon ionization threshold because they do not reflect the resonance structure of multiphoton ionization cross sections and in the resonance region they give cross sections averaged over the resonances.

We shall use a quasiclassical approximation[1)] to generalize the cross sections obtained in Refs. 11 and 12 to include the resonance region. In the case of two-photon ionization we shall also find corrections which improve the behavior of the ionization cross sections at interresonance minima.

§1. DERIVATION OF MULTIPHOTON IONIZATION CROSS SECTIONS IN A QUASICLASSICAL APPROXIMATION

We shall begin from the Schrödinger equation for an electron interacting with the Coulomb field of a nucleus and with a periodic radiation field; we shall use the dipole approximation. If the wave function for this problem is expanded as a Fourier series in terms of the radiation field and as a series in terms of spehrical functions, the following system of equations is obtained for the radial functions[13,14]:

$$\left[\frac{d^2}{dr^2}+k_{Nl}^2(r)\right]f_{Nl}(r)=-F_0 r\left\{\left[\frac{l^2-M^2}{(2l-1)(2l+1)}\right]^{1/2}\times\left[f_{N-1,l-1}(r)+f_{N+1,l-1}(r)\right]\right.$$
$$\left.+\left[\frac{(l+1)^2-M^2}{(2l+1)(2l+3)}\right]^{1/2}\left[f_{N-1,l+1}(r)+f_{N+1,l+1}(r)\right]\right\}, \quad (1)$$

$$k_{Nl}^2(r)=2[E+N\omega+Z/r-l(l+1)/2r^2]. \quad (2)$$

Here, E is the electron energy; M is the magnetic quantum number, which is conserved in the case of a linearly polarized radiation field; $l \geqslant |M|$; Z is the nuclear charge; F_0 and ω are the amplitude and frequency of the radiation field (the atomic system of units is used).

A quantum-mechanical perturbation theory for the system of equations (1) is based on the use of the Green functions. In the quasiclassical approximation the Green functions for the open and closed channels are as follows:

$$G_{Nl}(r,r')=-[k_{Nl}(r)k_{Nl}(r')]^{-1/2}\sin[S_{Nl}(r_<)+\pi/4]\times\exp[i(S_{Nl}(r_>)+\pi/4)], \quad E_N>0, \quad (3)$$

$$G_{Nl}(r,r')=-[k_{Nl}(r)k_{Nl}(r')]^{-1/2}\{\sin[S_{Nl}(r_<)+\pi/4]\times\cos[S_{Nl}(r_>)+\pi/4]$$
$$+\operatorname{tg}[S_{Nl}(r_2)]\sin[S_{Nl}(r)+\pi/4]\sin[S_{Nl}(r')+\pi/4]\}, \quad E_N<0,$$

$$S_{Nl}(r)=\int_{r_1}^{r}dr'k_{Nl}(r'), \quad (4)$$

$$E_N=E+N\omega. \quad (5)$$

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Here, $r_<$ and $r_>$ are, correspondingly, the smaller and larger of the quantities r and r'; r_1 and r_2 are the turning points (for simplicity, we shall omit the indices N and l of these quantities). Using a quasiclassical wave function of the initial state[15] with quantum numbers n and l_0

$$f_{0l_0}(r)=a_0 k_{0l_0}^{-1/2}(r)\sin[S_{0l_0}(r)+\pi/4], \quad (6)$$

$$a_0=(2Z^2/\pi n^3)^{1/2}, \quad (7)$$

we can develop a quasiclassical perturbation theory for the specific case of the system (1), which would have reduced the procedure to a calculation of quasiclassical multiphoton matrix elements. However, in view of strong oscillations of the quasiclassical functions $f_{Nl}(r)$ and of the Green functions of Eq. (3), it is difficult to calculate them directly. We shall show later that over a major range of r the strongly oscillating part of the radial functions is the same for all the channels so that it is convenient to separate it in advance and to adopt[14,16] compelx, and slowly varying with r, amplitudes $a^{\pm}{}_{Nl}(r)$ using the representation

$$f_{Nl}(r)=-{}^1/_2 ik_{Nl}^{-1/2}\{a_{Nl}{}^+(r)\exp[i(S_{Nl}(r)+\pi/4)] - a_{Nl}{}^-(r)\exp[-i(S_{Nl}(r)+\pi/4)]\}. \quad (8)$$

Introduction of variable amplitudes $a^{\pm}{}_{Nl}(r)$ is analogous to the method of variation of constants in the solution of inhomogeneous differential equations. If we now substitute Eq. (8) into Eq. (1) and employ the usual quasiclassical approximations, dropping the rapidly oscillating terms from the argument of the exponential function $S_{N'l'}+S_{Nl}$, we obtain[14,16] the following system of equations for the slow amplitudes:

$$\pm i\frac{da_{Nl}{}^{\pm}(r)}{dr}=-\frac{F_0 r}{2}\sum\left[\frac{l_m{}^2-M^2}{(2l_m-1)(2l_m+1)}\right]^{1/2} \times\frac{a^{\pm}_{N'l'}(r)}{(k_{Nl}k_{N'l'})^{1/2}}\exp[\pm i(S_{N'l'}-S_{Nl})], \quad (9)$$

where the summation is carried out over all values of $N'=N\pm 1$ and $l'=l\pm 1$, and l_m is the greater of the two values l and l'. We can see that the change in the amplitudes $a^{\pm}{}_{Nl}(r)$ considered as a function of r is entirely due to the coupling between the channels.

We shall now consider the boundary conditions for the functions $a^+{}_{Nl}(r)$ and $a^-{}_{Nl}(r)$. Firstly, the function $f_{Nl}(r)$ should decrease exponentially to the left of the turning point r_1. This gives rise to the condition[17]

$$a_{Nl}{}^+(r_1)=a_{Nl}{}^-(r_1). \quad (10)$$

In the case of open channels in the ionization problem a wave converging at infinity should be absent, i.e.,

$$a_{Nl}{}^-(\infty)=0, \quad E_N>0. \quad (11)$$

In the case of closed channels the function of Eq. (8) should also decrease exponentially to the right of the second turning point r_2. This occurs only on condition that

$$a_{Nl}{}^+(r_2)\exp[iS_{Nl}(r_2)]+a_{Nl}{}^-(r_2)\exp[-iS_{Nl}(r_2)]=0, \quad E_N<0. \quad (12)$$

It is this last condition that gives rise to a tangent in the Green function (3) and, in the final analysis, is responsible for resonances of the ionization cross sections. For a Coulomb field, we obtain

$$S_{Nl}(r_2)=\pi[Z(-2E_N)^{-1/2}-l-{}^1/_2]. \quad (13)$$

Before applying perturbation theory to the system (9), we note that two limiting cases[17] may be encountered in the calculation of quasiclassical matrix elements. In one case the matrix elements are calculated between the states whose energies are close to one another and, therefore, the matrix elements convege to the Fourier components of the corresponding classical quantity. In the other limiting case the energies of states differ considerably and the matrix elements are exponentially small. However, in the problem under discussion the values of the matrix elements are influenced not only by the energy but also by the orbital quantum number l. In the multiphonon ionization case the matrix elements are calculated between the states whose energies differ by ω. We shall assume that ω is much greater than the separation between the neighboring levels, amounting to n^{-3}, where n is the principal quantum number of the initial state. Since in the N-photon ionization case the value of $N\omega$ is of the order of n^{-2}, it follows that further steps should be restricted by the conditions

$$1\gg\omega\gg n^{-3}, \quad N\ll n. \quad (14)$$

In fact, it follows from a comparison of the quasiclassical cross sections with the corresponding quantum-mechanical values (see below) that the quasiclassical cross sections are sufficiently accurate even subject to conditions less stringent than those given by Eq. (14).

If $\omega\gg n^{-3}$, a direct calculation based on Eqs. (4) and (2) can be used to show that the differences $S_{Nl}-S_{N'l'}$ are generally of the order of $n-n'$, i.e., these differences are large, so that the matrix elements are exponentially small with just one exception. This exception occurs when $l\ll n$ so that the differences $S_{Nl}-S_{N'l'}$ become small near the turning point r_1. It is this range of variation of r that we shall be interested in the subsequent treatment. If $l\ll n$, the tuning point

$$r_1=(n^2/Z)\{1-[1-(l+{}^1/_2)^2/n^2]^{1/2}\}\approx(l+{}^1/_2)^2/2Z \quad (15)$$

is independent of the electron energy and it is approximately the same for all the channels (including open ones), because we are assuming that the usual quasiclassical conditions $l-l'\ll l$ are obeyed. In the case of values of l close to n the differences $S_{Nl}-S_{N'l'}$ again decrease, but there is also a reduction in the classically accessible range of electron motion (the turning points r_1 and r_2 approach one another), so that once again the matrix elements are small.

Expanding the quantities k_{Nl} as a series near the point r_1, we find that

$$k_{Nl}=\left[\frac{2Z}{r}-\frac{L^2}{r^2}+2E_N+\frac{L^2-(l+{}^1/_2)^2}{r^2}\right]^{1/2}\approx\left[\frac{2Z}{r}-\frac{L^2}{r^2}\right]^{1/2} +\left[E_N+\frac{L^2-(l+{}^1/_2)^2}{2r^2}\right]\left[\frac{2Z}{r}-\frac{L^2}{r^2}\right]^{-1/2}, \quad (16)$$

$$S_{Nl}-S_{N'l'}\approx(N-N')\omega t-(m-m')\varphi, \quad (17)$$

$$r=(L^2/2Z)(1+u^2), \quad t=(L^3/2Z^2)(u+u^3/3), \quad \varphi=2\tan^{-1}u, \quad (18)$$

$$L=l_0+{}^1/_2, \quad m=l-l_0, \quad l-l_0\ll l\ll n. \quad (19)$$

The approximations (16)–(18) imply that when the condition (19) is obeyed, the electron motion near r_1 is the same for all the channels and follows a parabola. Here, t and φ are the classical time and angle. The range far from r_1 makes an exponentially small contribution to the matrix elements and, therefore, in the calculation of these elements we can extend the variable r (or u) to infinity. It follows from Eq. (17) that in our approximation the matrix elements again reduce to the Fourier components. As is usual in the calculation of quasiclassical matrix elements, we have to use a slightly ambiguous procedure of introduction of certain average quantities.[18] In our case such a quantity is the orbital momentum L. For simplicity, we shall equate it to the orbital momentum of the initial state.

Ignoring the difference between the pre-exponential factors for the various channels in the system (9), allowing for Eq. (17) and adopting the variable t in accordance with Eq. (18), we finally obtain the following impact-parameter type equations instead of the system (9):

$$a_{Nm}(t)=a_{Nm}(-\infty)+\frac{iF_0}{4}\left(1-\frac{M^2}{l_0^2}\right)^{1/2}\int_{-\infty}^{t}dt'r(t')$$

$$\times\sum a_{N'l'}(t')\exp[i(N'-N)\omega t'+i(m-m')\varphi(t')], \quad (20)$$

where the summation is carried out over all the values $N'=N\pm1$ and $l'=l\pm1$, and the function $a_{Nm}(t)$ is equal to $a^{+}{}_{Nm}(t)$ and $a^{-}{}_{Nm}(t)$, for $t>0$ and $t<0$, respectively. The constant $a_{Nm}(-\infty)$ vanishes for $E_N<0$, whereas if $E_N<0$, we find from Eqs. (12) and (13) tht is determined by the condition

$$a_{Nm}(-\infty)=a_{Nm}(\infty)\exp[2i\pi Z(-2E_N)^{-1/2}]. \quad (21)$$

We shall now introduce functions

$$h_{\pm}(t)=1/4iF_0(1-M^2/l_0^2)^{1/2}\int_{-\infty}^{t}dt'r(t')\exp[-i\omega t'\pm i\varphi(t')],$$
$$H_{\pm}=h_{\pm}(\infty). \quad (22)$$

Using the definitions of Eq. (18) and the integral representations[19] of the Airy function Ai(x) and its derivative Ai'(x), we readily find that

$$H_{\pm}=i\pi F_0(Z/2\omega^5)^{1/3}(1-M^2/l_0^2)^{1/2}[-\mathrm{Ai}'(x)\pm x^{1/2}\,\mathrm{Ai}\,(x)], \quad (23)$$
$$x=(\omega l^3/2Z^2)^{2/3}. \quad (24)$$

Since the initial state is described by the function (6), we can use Eq. (20) and the iteration procedure to obtain the following expressions for the first few amplitudes a_{Nm}:

$$a_{11}(t)=(h_++\gamma_1H_+)a_0, \qquad a_{1,-1}(t)=(h_-+\gamma_1H_-)a_0,$$
$$a_{22}(t)=\frac{1}{2!}[h_+^2+2\gamma_1h_+H_++\gamma_2(1+2\gamma_1)H_+^2]a_0,$$
$$a_{20}(t)=[h_+h_-+\gamma_1(h_+H_-+h_-H_+)+\gamma_2(1+2\gamma_1)H_+H_-]a_0,$$

where

$$\gamma_k=0, \quad k\omega>Z^2/2n^2,$$
$$\gamma_k=-1/2(1-i\,\mathrm{ctg}\,\pi\nu_k), \quad k\omega<Z^2/2n^2, \quad (25)$$
$$\nu_k=Z(Z^2/n^2-2k\omega)^{-1/2}.$$

The induction method can be used to demonstrate the validity of the general formula

$$a_{Nm}(t)=\frac{a_0}{((N+m)/2)!((N-m)/2)!}$$
$$\times\sum_{k=0}^{(N+m)/2}\sum_{s=0}^{(N-m)/2}\binom{(N+m)/2}{k}\binom{(N-m)/2}{s}$$
$$\times h_+^{(N+m)/2-k}(t)H_+{}^kh_-^{(N-m)/2-s}(t)H_-{}^s\gamma_{k+s}R_{k+s}. \quad (26)$$

The quantities R_s satisfy the recurrence relationship

$$R_s=\sum_{k=0}^{s-1}\binom{s}{k}\gamma_kR_k, \quad \gamma_0R_0=1. \quad (27)$$

We are interested in the probability of the N-photon ionization process, which is governed by the amplitudes $a_{Nm}(\infty)$ when $E_N>0$. It follows from Eq. (26) that these amplitudes are

$$a_{Nm}(\infty)=a_0H_+^{(N+m)/2}H_-^{(N-m)/2}R_N/((N+m)/2)!((N-m)/2)!. \quad (28)$$

Calculating the flux of electrons of energy E_N across an element of the surface area, dividing it by the photon flux $F_0^2/8\pi\alpha\omega$, and averaging over the magnetic quantum number M, we obtain the differential cross section of N-photon ionization of a state with the quantum numbers n and l_0:

$$d\sigma_N(nl_0,\vartheta)=\frac{2\pi\alpha\omega}{F_0^2(2l_0+1)}\sum_{M=-l_0}^{l_0}\left|\sum_m a_{Nm}(\infty)Y_{l_0+m,M}(\vartheta,\varphi)\right|^2d\Omega. \quad (29)$$

Here, α is the fine structure constant, ϑ is the angle between the direction of emission of an electron and direction of polarization of an electromagnetic field, and m assumes the values $-N,-N+2,\ldots,N$ (naturally, $l_0+m\geqslant|M|$). Since M in the inner sum is the same for all the spherical functions, the cross section is independent of the angle φ. It follows from Eqs. (28) and (23) that the cross section (29) contains the factor $(1-M^2/l_0^2)$ raised to the power N.

We shall replace the summation over M by integration with respect to the variable β, which is related to M by $M=l_0\cos\beta$. After integration of Eq. (29) with respect to the angular variables and β, and after summation over m, we obtain the total cross section of N-photon ionization of a state with the quantum numbers n and l_0:

$$\sigma_N(nl_0)=\frac{4\alpha\omega Z^2(2\pi F_0)^{2N}}{n^3F_0{}^2(2N+1)!}$$
$$\times\left(\frac{Z}{2\omega^5}\right)^{\frac{2N}{3}}|R_N|^2[\mathrm{Ai}'(x)-x^{1/2}\,\mathrm{Ai}\,(x)]^{2N}$$
$$\times F[-N,-N;1;[\mathrm{Ai}'(x)$$
$$+x^{1/2}\,\mathrm{Ai}\,(x)]^2/[\mathrm{Ai}'(x)-x^{1/2}\,\mathrm{Ai}\,(x)]^2]. \quad (30)$$

The last factor in the above expression is a polynomial which can be expressed in terms of a hypergeometric function characterized by an argument which varies from 1 to 0 when x is varied from zero to infinity.

We shall also determine the cross section for N-photon ionization of an atom for a shell with the principal quantum number n:

$$\sigma_N(n)=\frac{1}{n^2}\sum_{l_0=1}^{n-1}(2l_0+1)\sigma_N(nl_0)\approx\frac{2}{n^2}\int_0^\infty dL\,L\sigma_N(nl_0). \tag{31}$$

Using the relationship for hypergeometric functions[19]

$$(1+x)^{2N}F\left[-N,-N;1;\left(\frac{1-x}{1+x}\right)^2\right]$$
$$=\frac{(2N)!}{(N!)^2}F[-N,{}^1/_2;-N+{}^1/_2;x^2], \tag{32}$$

we can rewrite Eq. (31) in the form

$$\sigma_N(n)=\frac{\alpha F_0^{2N-2}Z^{(2N+10)/3}T_N|R_N|^2}{n^5(N!)^2\omega^{(10N-1)/3}}, \tag{33}$$

where the numbers T_N are given by the integral

$$T_N=\frac{2^{(4N+8)/3}\pi^{2N}}{2N+1}$$
$$\times\int_0^\infty dx[\mathrm{Ai}'(x)]^{2N}F[-N,{}^1/_2;-N+{}^1/_2;x\,\mathrm{Ai}^2(x)/\mathrm{Ai}'^2(x)]. \tag{34}$$

These numbers were calculated in Ref. 11. In the case of circularly polarized radiation we have the same expression (33), but the numbers T_N should be replaced with others also given in Ref. 11. The cross section $\sigma_N(n)$ calculated in Refs. 11 and 12 differs from Eq. (32) only by the absence of the resonance factor $|R_N|^2$. The quantities R_N are defined by Eqs. (27) and (25) and for $N=1$, 2, and 3, they are

$$R_1=1,\quad R_2=\begin{cases}1, & \omega>Z^2/2n^2,\\ i\operatorname{ctg}\pi\nu_1, & \omega<Z^2/2n^2,\end{cases}$$
$$R_3=\begin{cases}1, & \omega>Z^2/2n^2,\\ -{}^1/_2(1-3i\operatorname{ctg}\pi\nu_1), & Z^2/4n^2<\omega<Z^2/2n^2,\\ -{}^1/_2(1+3\operatorname{ctg}\pi\nu_1\operatorname{ctg}\pi\nu_2), & \omega<Z^2/4n^2.\end{cases}$$

The different form of R_N at different frequencies is due to the fact that intermediate states may be in discrete or continuous spectra. It is clear from the above formulas that on transition of an intermediate state from a discrete to a continuous spectrum the corresponding $\cot\pi\nu_k$ should be, in accordance with Eq. (25), replaced simply with $-i$. In the $N=1$ case the cross section (33) reduces to the well-known Kramers formula for the usual photoelectric effect.

Before comparing the cross sections of Eq. (33) with the corresponding cross sections found completely by the quantum-mechanical approach, it should be noted that in the resonance region the cross sections (33) should be sufficiently exact where they are large. However, at interresonance minima, where the cross sections are much smaller than the average quasiclassical values ($R_N=1$), Eq. (33) cannot yield all the fine features of the quantum-mechanical interference. Therefore, we shall find the corrections to the cross sections of Eq. (33) in the two-photon ionization case.

§2. CORRECTIONS TO CROSS SECTIONS AT INTERRESONANCE MINIMA

The main error in the quasiclassical cross sections given above originates not from the quasiclassical approximation itself but because of transition from the system (9) to the system (20). This involves neglect of the difference k_{Nl} between the various channels in determination of the preexponential factors and the arguments of the exponential functions allow for the difference in accordance with the expansions (16) and (17), i.e., only in the first order with respect to the difference of the energies and of the angular momentum. Expanding k_{Nl} near the turning point r_1 in the same way as in Eq. (16), we shall allow for the next order. Then,

$$k_{Nm}\approx\frac{2Z}{L}\left\{\frac{u}{1+u^2}+\frac{1}{Lu}\left[\frac{\xi_N}{2}(1+u^2)-\frac{m}{1+u^2}\right]\right.$$
$$\left.+\frac{1}{2L^2u^3}\left[-\frac{\xi_N^2}{4}(1+u^2)^3+m\xi_N(1+u^2)-m^2\right]\right\}, \tag{35}$$

$$(k_{N_1m_1}k_{N_2m_2})^{1/2}\approx\frac{L(1+u^2)}{2Zu}$$
$$\times\left\{1+\frac{1}{2Lu^2}\left[\frac{\xi_{N_1}+\xi_{N_2}}{2}(1+u^2)^2-m_1-m_2\right]\right\}, \tag{36}$$

$$S_{N_1m_1}-S_{N_2m_2}\approx(N_1-N_2)\,\omega t-(m_1-m_2)\,\varphi$$
$$+\frac{1}{L}\left[{}^1/_4(\xi_{N_2}^2-\xi_{N_1}^2)\left(\frac{u^5}{5}+u^3+3u-\frac{1}{u}\right)\right.$$
$$\left.+(m_1\xi_{N_1}-m_2\xi_{N_2})\left(u-\frac{1}{u}\right)+\frac{m_1^2-m_2^2}{u}\right], \tag{37}$$

$$\xi_N=(L^3/2Z^2)\,(E+N\omega),\quad m=l-l_0. \tag{38}$$

We can see that the corrections allowed for in the present section are small quantities of the order of L^{-1}.

We shall first consider the amplitude a_{22}. In this case a solution of the system (9) by the iteration method has only one term with $N'=N-1$ and $l'=l-1$ on the right-hand side. Substituting th expansions (36) and (37) into Eq. (9) and retaining only terms of the order of L^{-1}, we obtain the following expression for the amplitude $a_{22}(\infty)$ below the one-photon ionization threshold

$$a_{22}(\infty)=-\frac{ia_0F_0^2}{8}\left(1-\frac{M^2}{l_0^2}\right)\left\{\left(\frac{Z}{\omega^5}\right)^{2/3}M_+^2\operatorname{ctg}\pi\nu_1-\omega^{-3}I_{22}\right\}, \tag{39}$$

$$M_\pm=2^{2/3}\pi[-\mathrm{Ai}'(x)\pm x^{1/2}\mathrm{Ai}(x)]. \tag{40}$$

The first term in Eq. (39) containing the cotangent corresponds to the approximation of the preceding section, and the correction I_{22} to this term obtained allowing for the integral identity

$$\int_{-\infty}^{\infty}dtf(t)\int_{-\infty}^{t}dt'g(t')=\int_{-\infty}^{\infty}dtg(t)\int_{t}^{\infty}dt'f(t') \tag{41}$$

can be expressed in the form

$$I_{22}=\frac{\xi^3}{8}\int_{-\infty}^{\infty}dug_+(u)\,G_+(u)\left\{i\left[1-\frac{\xi}{2}(1+u^2)^2\right]\right.$$
$$\left.\times\left[u^2-\frac{1}{L}\left(\frac{L^3}{4n^2}-\xi+2\right)\right]^{-1}-z(u)+2\xi\left(\frac{1}{u}-u\right)\right\}, \tag{42}$$

where

$$g_\pm(u)=(1+u^2)\,(1-u^2\pm 2iu)\exp[-i\xi(u+u^3/3)],$$

$$G_\pm(u)=\int_{-\infty}^{u} du'g_\pm(u')-\int_{u}^{\infty} du'g_\pm(u'), \qquad (43)$$

$$z(u)=\frac{\xi^2}{2}\left(\frac{u^5}{5}+u^3+3u-\frac{1}{u}\right)+\frac{2}{u}, \quad \xi=\frac{\omega L^3}{2Z^2}.$$

Near the resonance curve minima where the cotangent vanishes, the term I_{22} predominates. Equation (39) is derived ignoring factors of the order of L^{-1} in front of the cotangent and the corrections of the same order of magnitude to the amplitudes in the nonresonance region, i.e., to the amplitudes above the one-photon ionization threshold.

We shall now consider calculation of the integral (42). Firstly, we note that this integral is converging (this applies to the principal value), although it does contain negative powers of the variable u in the integrand. Allowing for the fact that $n\gg L\gg 1$, we can integrate Eq. (42) by parts to avoid completely the negative powers of the variable u. The same integration by parts can reduce the powers of the variables u and u'. The resultant single integrals then reduce to the Airy functions. If the variables u and u' are replaced with $v=u+u'$ and $v'=u-u'$, the remaining double integral can be reduced to a double integral with constant limits, and, in the final analysis, to an integral of the Airy function. The result of fairly lengthy calculations is the following expression for the integral (42):

$$I_{22}=\frac{\pi}{64}\Big\{(296y-4y^{5/2}+y^4)\,\mathrm{Ai}(y) -\left(\frac{1608}{5}y^{1/2}-2y^2+4y^{7/2}-y^5\right)\mathrm{Ai}'(y) +\left[\frac{64}{5}(7-2y^{3/2})-4y^{5/2}+y^6\right]\int_y^\infty dv\,\mathrm{Ai}(v)\Big\}, \qquad (44)$$

$$y=(\omega L^3/Z^2)^{2/3}=2^{2/3}x.$$

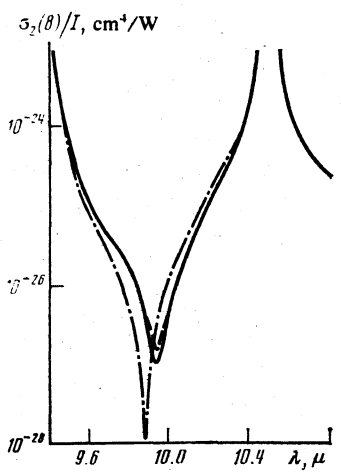

FIG. 1. Cross sections $\sigma_2(8)$ of two-photon ionization of the hydrogen atom from the $n=8$ shell, divided by the intensity of the radiation field I, plotted as a function of the radiation wavelength λ (μ). The continuous curve represents the quantum-mechanical calculations of Ref. 20, the dashed curve gives the values calculated from Eq. (50), and the chain curve is a calculation made using Eq. (50) without the last two correction terms.

It is interesting to note that the argument of the function I_{22} can be obtained from the argument of the function $M_\pm$ by replacing ω with 2ω in the latter.

The amplitudes $a_{2,-2}$ are calculated in a similar manner. The resultant integral $I_{2,-2}$ differs from Eq. (44) only by the opposite signs at fractional powers of y. The amplitude a_{20} is determined by two matrix elements in accordance with two possible values of the orbital momentum $l'=l_0\pm 1$ of an intermediate state and can be represented in the form

$$a_{20}=-\frac{ia_0F_0^2}{4}\left\{\left(1-\frac{M^2}{l_0^2}\right)\times\left[\left(\frac{Z}{\omega^5}\right)^{2/3}M_+M_-\operatorname{ctg}\pi\nu_1-\omega^{-3}I_{20}\right]+\frac{M^2}{l_0^2}\omega^{-3}P\right\}, \qquad (45)$$

$$I_{20}=\frac{\xi^3}{16}\int_{-\infty}^{\infty} du g_-(u)G_+(u)\left\{i\left[1-\frac{\xi}{2}(1+u^2)^2\right]\times\left[u^2-\frac{1}{L}\left(\frac{L^3}{4n^2}-\xi-1\right)\right]^{-1}-z(u)\right\} -\frac{\xi^3}{16}\int_{-\infty}^{\infty} du g_+(u)G_-(u)\left\{i\left[1+\frac{\xi}{2}(1+u^2)^2\right]\times\left[u^2-\frac{1}{L}\left(\frac{L^3}{4n^2}-\xi+1\right)\right]^{-1}+z(u)\right\}, \qquad (46)$$

$$P=-\frac{i\xi^3}{8}\int_{-\infty}^{\infty} du g_-(u)G_+(u). \qquad (47)$$

The last term in the amplitude (45) appears because of the difference between the factors $1-M^2/l'^2$ for these two matrix elements. Calculations similar to those described above yield the following values of the integrals I_{20} and P:

$$I_{20}=\frac{\pi}{320}\left\{(296y+5y^4)\,\mathrm{Ai}(y)+(226y^2+5y^5)\,\mathrm{Ai}'(y) +(-192+256y^3+5y^6)\int_y^\infty dv\,\mathrm{Ai}(v)\right\}, \qquad (48)$$

$$P=2\pi\int_y^\infty dv\,\mathrm{Ai}(v). \qquad (49)$$

Knowing the amplitudes $a_{2m}(\infty)$ and applying the same procedure as in the derivation of Eq. (33), we obtain the two-photon ionization cross section below the threshold of one-photon ionization for an atomic shell with the principal quantum number n:

$$\sigma_2(n)=\frac{\alpha F_0^2Z^{14/3}}{n^5\omega^{19/3}}\left[c_0\operatorname{ctg}^2\pi\nu_1-c_1\frac{\omega^{1/3}}{Z^{2/3}}\operatorname{ctg}\pi\nu_1+c_2\frac{\omega^{2/3}}{Z^{4/3}}\right]. \qquad (50)$$

The constants c_0, c_1, and c_2 are given by the integrals

$$c_0=\frac{1}{15\cdot 2^{1/3}}\int_0^\infty dx[M_+^4+M_-^4+4M_+^2M_-^2],$$

$$c_1=\frac{2^{2/3}}{15}\int_0^\infty dx[M_+^2I_{22}+M_-^2I_{2,-2}+M_+M_-(4I_{20}-P)], \qquad (51)$$

$$c_2=\frac{1}{15\cdot 2^{1/3}}\int_0^\infty dx\left[I_{22}^2+I_{2,-2}^2+4I_{20}^2-2I_{20}P+\frac{3}{2}P^2\right].$$

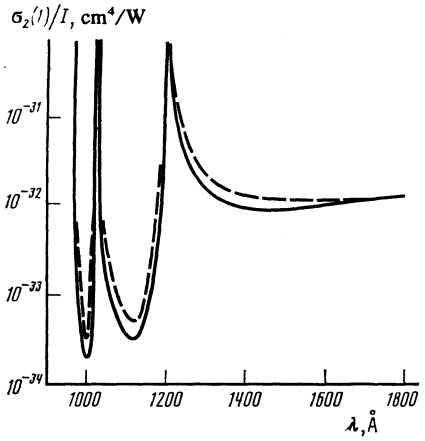

FIG. 2. Cross section $\sigma_2(1)$ of two-photon ionization of the ground state of the hydrogen atom, divided by the intensity of the radiation field I, plotted as a function of the radiation wavelength λ. The continuous curve represents the quantum-mechanical calculation given in Ref. 4 and the dashed cuve is the calculation made using Eq. (50).

Numerical integration yields the following values of these constants:

$$c_0=0.8058, \quad c_1=1.612, \quad c_2=1.299. \tag{52}$$

The last two terms in the cross section (50) are the corrections calculated above to the two-photon ionization cross section and we can see they are the first terms of the expansion in terms of a small quantity $\omega^{1/3}$.

§3. COMPARISON WITH QUANTUM-MECHANICAL CALCULATIONS

We shall now compare the simple expressions for the cross sections given by Eqs. (33) and (50) with the corresponding quantum-mechanical expressions. In the case of two-photon ionization above the one-photon ionization threshold the results given by Eq. (33) are accurate to within a few percent[11] even in the case of low values of n. Unfortunately, only one quantum-mechanical calculation[20] of the two-photon ionization cross sections for large values of n has been published so far. The results of these calculations are compared in Fig. 1 with Eq. (50). We can see that just the first term of Eq. (50) ensures a satisfactory agreement with the quantum-mechanical calculations, excluding the regions of interresonance minima. Allowance for the last two terms in Eq. (50) ensures a practically complete agreement with the quantum-mechanical calculations. A similar agreement is observed also for $n = 9$ and 10. Figure 2 makes the same comparison for the ground state of the hydrogen atom. Even in this case Eq. (50) gives a satisfactory result. Naturally, we can expect that in the case of the differential cross sections (29) and (30) the differences from the corresponding quantum-mechanical cross sections will be larger than for the cross section $\sigma_N(n)$ because in the latter case the finer quantum-mechanical effects may disappear as a result of averaging of Eq. (31). However, more detailed quantum-mechanical calculations are needed to determine this question.

The cross sections (33) and (50) apply, strictly speaking, only to the hydrogen atom. Although the highly excited states of complex atoms are hydrogen-like, it follows from the above analysis that the probability of multiphoton ionization of highly exicted states of atoms is governed mainly by the small distances r and states with low values of l, i.e., precisely by those values of r and l at which the deviation of the wave functions of complex atoms from the hydrogen wave functions is greatest. An allowance for this deviation can be made in the quasiclassical approximation by applying, for example, the quantum defect method.[1]

[1)] The main results of the work reported here were presented at the Eighth All-Union Conference on Physics of Electronic and Atomic Collisions, Leningrad, 1981.

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