Multiphoton and tunnel ionization by an optical field with polar asymmetry

N. B. Baranova
Technical University, Chelyabinsk, 454080 Russia

H. R. Reiss
Physics Department, The American University, Washington, D.C. 20016-8058

B. Ya. Zel'dovich
Technical University, Chelyabinsk, 454080 Russia

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A simplified form of multiphoton and tunneling ionization is applied to the problem of ionization of atoms by a bichromatic field consisting of the coherent superposition of a fundamental field and its second harmonic: $\mathbf{E}(t)=0.5[\mathbf{E}_1\exp(-i\omega t)+\mathbf{E}_2\exp(-2i\omega t)]+\text{c.c.}$ This superposition possesses a polar asymmetric average of the cube of the field, $\langle E^3\rangle=\frac{3}{8}(E_1^{2*}E_2+E_1^2E_2^*)$. Two limits of the adiabaticity parameter γ are considered, where $\gamma=\omega(2mI)^{1/2}/(|e|E)$ and I is the ionization potential. The case $\gamma\ll1$ corresponds to tunneling ionization by a quasistatic field. Here, the electron is released from the barrier with almost zero velocity at the moment when the magnitude of the field strength $|\mathbf{E}|$ is maximum. Subsequent oscillations of the free electron in the strong but adiabatically decreasing field yield some residual velocity. Polar asymmetry of the distribution of that velocity and $\langle\mathbf{v}\rangle$ are calculated as a function of the phase shift, $\arg(E_1^2E_2^*)$, between the squared fundamental field E_1^2 and its second harmonic E_2. The other case, $\gamma\gg1$, corresponds to multiphoton ionization by the combined fields where $n_1\hbar\omega+n_2 2\hbar\omega\approx I$. Interference of the amplitudes corresponding to opposite parities of the total number of quanta n_1+n_2 with the same energy $(n_1+2n_2)=\text{const}$ $(\approx I/\hbar\omega)$ gives rise to the $\arg(E_1^2E_2^*)$-dependent polar asymmetry of emitted electrons. Recent experiments on $\arg(E_1^2E_2^*)$-sensitive effects in multiphoton ionization are discussed.

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I. INTRODUCTION

Ionization of atoms by a quasimonochromatic field for the case when $I\gg\hbar\omega$ is the subject of a large number of papers. Here I is the ionization potential and ω is the frequency of the field. We will employ the classification of possible processes based on the dependence of the parameters of atom and field established in the paper by Keldysh [1] and discussed also in Sec. 77 of the text by Landau and Lifshitz [2]. Related techniques are treated in other references [3–7]. The main idea of the Keldysh classification is the estimation of the velocity of the electron under the barrier, which is the result of the sum of the atomic potential $U_{\text{at}}(\mathbf{r})$ and that of the external field $eE(t)x$, as shown in Fig. 1. Here and below, e is the absolute magnitude of the electron's charge. The kinetic energy $K=-I-U_{\text{at}}(\mathbf{r})-eE(t)x$ of the electron is negative under the barrier. Therefore the velocity $v=\pm0.5\sqrt{2K/m}\approx\pm0.5i\sqrt{2I/m}$ is purely imaginary there, and that fact corresponds to a rather low probability of ionization. However, a knowledge of the order of magnitude of that velocity, $|\mathbf{v}|$, makes it possible to estimate the time $\tau_{\text{tun}}=L/|\mathbf{v}|=\sqrt{2mI/e^2E^2}$, after which the (exponentially small) steady-state probability of tunneling ionization would be established. Here $L=I/eE$ is the estimate of the length of the barrier. If the field amplitude and (or) its direction change only over a much longer time period, i.e., if the parameter

$$\gamma=\omega\tau_{\text{tun}}=(2mI\omega^2/e^2E^2)^{1/2} \tag{1}$$

is much smaller than unity $(\gamma\ll1)$ and ω represents a low frequency, then at each instant one can deal with the quasistatic tunneling ionization probability

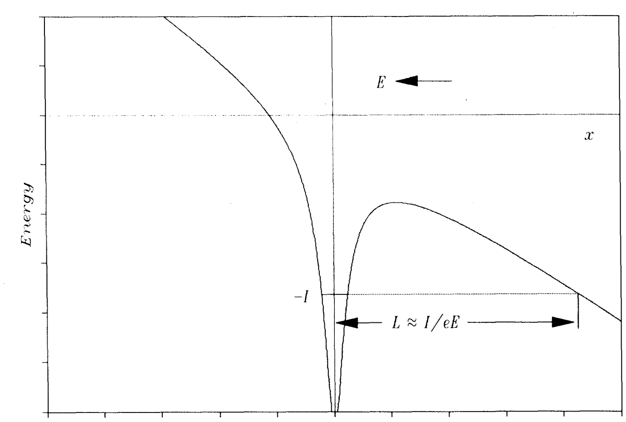

FIG. 1. Instantaneous picture of the superposition of the atomic potential U_{at} binding the electron in the atom and the linear potential of the external light field. The length of the barrier L may be estimated as $L\approx I/eE$, where I is the ionization potential and e (>0) is the absolute value of the electron charge.

$$W_{\text{tun}}(|\mathbf{E}(t)|) \sim \exp[-B/|\mathbf{E}(t)|] ,$$
$$B = \tfrac{4}{3}\sqrt{2mI^3}/\hbar e . \tag{2}$$

For the last expression, see Ref. [2] or Sec. V below. We note that $\gamma \ll 1$ can also occur at high frequencies if E is sufficiently large [8], as is true in much of the work done on atomic stabilization. That situation does not correspond to tunneling, and is not treated here. In the opposite case when the parameter γ is large, $\gamma \gg 1$, the field is rapidly oscillating and the ionization probability $W \sim (E)^{2N}$ corresponds to an N-photon process. $N \approx I/\hbar\omega$ is approximately the minimum number of quanta which are to be absorbed to release the electron. The above view of the ionization mechanism underlies much of the work in this field [1,2,5,9].

In reality, an optical pulse certainly is not monochromatic, as in Fig. 2(a). However, the behavior of the optical frequency carrier $E_x \cos(\omega t + \alpha)$ possesses inversion symmetry: the change $E_x \to -E_x$ corresponds to an insignificant change of the time origin $t \to t + \pi/\omega$ or a phase change $\alpha \to \alpha + \pi$. Therefore the process, whether of tunneling or N-photon nature, exhibits total inversion symmetry in terms of the velocity distribution of the emitted electrons and other properties.

In recent years, a common type of bichromatic optical field,

$$\mathbf{E}(t) = \tfrac{1}{2}[\mathbf{E}_1 \exp(-i\omega t) + \mathbf{E}_2 \exp(-2i\omega t)] + \text{c.c.} \tag{3}$$

(i.e., a fundamental wave and its second harmonic) has attracted much attention due to the polar asymmetry of such a field. Figure 2(b) shows an example of the field $E(t) = \cos(\omega t) + \cos(2\omega t)$, and Fig. 2(c) shows the field $E(t) = \cos(\omega t) + \cos(2\omega t + \pi/2)$. There is no dc component in these fields: $\langle E \rangle = 0$. However, they possess a certain polar asymmetry which, in particular, may be characterized by third-order moments, e.g.,

$$\langle E_x^3(t) \rangle = \tfrac{3}{8}(E_{1x}^2 E_{2x}^* + E_{1x}^{2*} E_{2x}) . \tag{4}$$

Novel physical effects which arise in fields with $\langle E^3 \rangle \neq 0$ include the recording of holograms of second-order polarizability $\sim \chi^{(2)}(\mathbf{r})$ in fused silica fibers [10,11], ponderomotive forces $\sim \nabla \langle E^3 \rangle$, interference between one- and two-photon ionization [9,11–14], multiphoton ionization [9,13], recording of a static photorefractive hologram by a running interference pattern [15], coherent photovoltaic effect [16], and others.

In this paper we plan to discuss both multiphoton and tunneling ionization of an atom by the bichromatic field of Eq. (3), with emphasis on the effects of polar asymmetry. We do not intend to cover the full variety of conditions relating to the pulse duration, focal waist size, and other such matters. We do, however, take full account of the second-order ponderomotive potential (or quiver energy); see Eq. (13) or (14) below. No attempt is made to treat its spatial gradient, i.e., the ponderomotive force. (Detailed consideration and review of the effects of that force are given in Ref. [17].) This means that for the tunneling regime we are restricted to the case of an ultrashort pulse and rather wide focal waists. Such a restriction allows us to emphasize the peculiarity of optical fields with $\langle E^3 \rangle \neq 0$ and the effects of polar asymmetry. The advantage of our approach consists of using the universal starting expression for the ionization amplitude, given in Eq. (9) below, which makes possible a qualitative description of both the tunneling and the multiphoton regimes.

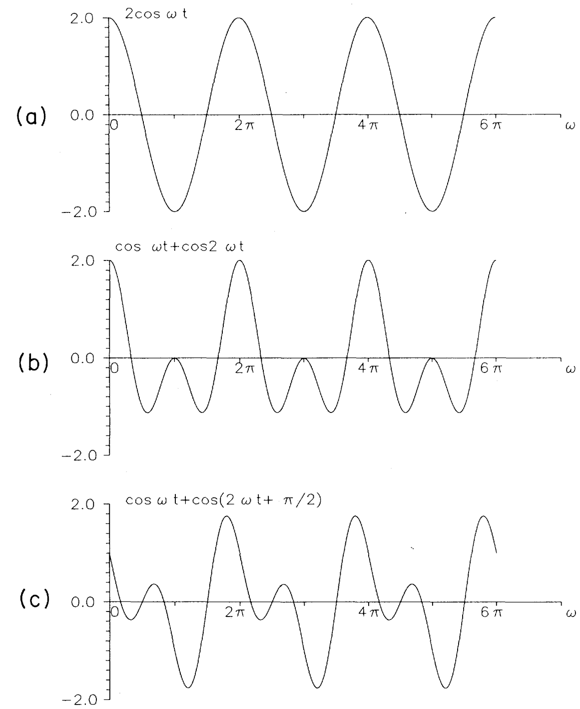

FIG. 2. (a) The monochromatic field $E = 2\cos\omega t$ possesses inversion symmetry. (b) The bichromatic field $E = \cos\omega t + \cos 2\omega t$ is "directed" upwards, $\langle E^3 \rangle = \frac{3}{4}$. (c) The bichromatic field with a relative phase shift, $E = \cos\omega t + \cos(\omega t + \pi/2)$, also possesses polar asymmetry, which may be characterized by a third-order time-correlation function.

II. SIMPLE APPROXIMATION FOR THE TRANSITION AMPLITUDE

An approximation which makes possible a simple analytical theory of multiphoton ionization is based on the following idea [1]. The atomic potential acts mainly on the initial bound state of the electron. Suppose that the initial state is described without any effects of the external field,

$$\Psi_{\text{init}}(\mathbf{r}, t) = \Psi_0(\mathbf{r}) \exp(iIt/\hbar) . \tag{5}$$

On the contrary, suppose that one can neglect the atomic potential while describing the final state of an almost free electron oscillating in the external field $\mathbf{E}(t)$,

$$\Psi_{\rm final}(\mathbf{r},t)={\rm const}\times\exp\left\{i(\mathbf{p}\cdot\mathbf{r}/\hbar)-(i/2\hbar m)\int_{-\infty}^{t}[\mathbf{p}+(e/c)\mathbf{A}(t')]^2dt'\right\} . \tag{6}$$

These are the conditions posited by Keldysh [1], but they are in fact the proper selections that arise in a "time-reversed" or "prior" form of a strong-field transition amplitude stated with boundary conditions appropriate to photoionization experiments [4,6,18]. Here $e>0$ is the absolute value of the electron charge, and $\mathbf{p}$ is the conserved value of its generalized momentum. We assume the following Hamiltonian of an electron in the homogeneous time-varying field $\mathbf{E}(t)$:

$$\hat{H}=(1/2m)[\hat{\mathbf{p}}+(e/c)\mathbf{A}(t)]^2 , \quad \hat{\mathbf{p}}=-i\hbar\frac{\partial}{\partial\mathbf{r}} , \tag{7}$$

where $\mathbf{A}(t)$ is the vector potential of the field,

$$\mathbf{A}(t)=-c\int_{-\infty}^{t}\mathbf{E}(t')dt' , \quad \mathbf{E}(t)=-(1/c)\partial\mathbf{A}(t)/\partial t . \tag{8}$$

If $I\gg\hbar\omega$, and we are not interested in the exact preexponential coefficient, then the transition probability $W\sim|a|^2$ is proportional to the squared modulus of the transition amplitude a, which is given approximately by

$$\begin{aligned} a(\mathbf{p})&\approx\int_{-\infty}^{+\infty}dt\,\Psi^*_{\rm final}(t)\Psi_{\rm init}(t)\\ &\approx\int_{-\infty}^{+\infty}\exp\left\{(i/\hbar)\left[It+(1/2m)\int_{-\infty}^{t}[\mathbf{p}+(e/c)\mathbf{A}(t')]^2dt'\right]\right\}dt . \end{aligned} \tag{9}$$

In this approximation, the details of atomic structure do not appear. All atomic information is contained in $\Psi_{\rm init}$ and in the ionization potential I. Hence the approximation is expected to have good applicability to negative ions, and to atoms under certain conditions [6]. It is not necessary to solve either ordinary or partial differential equations. All that is necessary is to calculate with reasonable precision the one-dimensional time integral (9).

The remaining problem is that for $I\gg\hbar\omega$ the integrand is an extremely rapidly oscillating function. Therefore, the natural method of calculation is the steepest-descent method. Up to a preexponential factor, that gives

$$a(\mathbf{p})=\sum_i\exp\left\{(i/\hbar)\left[It_i+(1/2m)\int_{-\infty}^{t_i}[\mathbf{p}+(e/c)\mathbf{A}(t')]^2dt'\right]\right\} , \tag{10}$$

where the t_i are the complex roots of the equation

$$I+(1/2m)[\mathbf{p}+(e/c)\mathbf{A}(t_i)]^2=0 , \tag{11}$$

which determines the positions of the saddle points in the complex plane $t=t'+it''$. It is necessary to select only steepest descent points, and those which lie along a continuous path of integration in the complex space deformed from the original path along the real t axis.

For the optical field with period $T=2\pi/\omega$, there are many equivalent solutions of Eq. (11) which differ from each other by the shift $t_i\rightarrow t_i+T$. In that case the sum (10) may be expressed as

$$\begin{aligned} a(\mathbf{p})=\exp(iM\xi/2)&\left\{\sum_i{}'\exp[iS(t_i)/\hbar]\right\}\\ &\times\sin[(M+1)\xi]/\sin\xi . \end{aligned} \tag{12}$$

Here $\xi=\Delta S/2\hbar$, the prime on the sum means that the summation is to be over one period only, $M\gg1$ is the number of periods which are taken into account, and

$$\Delta S/\hbar=(2\pi/\hbar\omega)[I+\mathbf{p}^2/2m+e^2\langle\,\mathbf{A}\,\rangle^2/2mc^2] \tag{13}$$

is the increment of action during one period. Angular brackets denote averaging over the period, and the value of $e^2\langle\,\mathbf{A}\,\rangle^2/2mc^2$ corresponds to the time-averaged oscillation (or quiver) energy of the electron in the field. The factor $\sin(M+1)\xi/\sin\xi$ is large only for $\xi=N_1\pi$, so that

$$I+\frac{\mathbf{p}^2}{2m}+\frac{e^2\mathbf{E}_1\cdot\mathbf{E}_1^*}{4m\omega^2}+\frac{e^2\mathbf{E}_2\cdot\mathbf{E}_2^*}{4m(2\omega)^2}=N_1\hbar\omega , \tag{14}$$

where N_1 is an integer. That corresponds to absorption of field energy in small discrete portions $\hbar\omega$. Increments of $2\hbar\omega$ are also possible. Equation (14) shows which discrete values of $\mathbf{p}^2/2m$ are allowed due to interference of contributions from different periods, or, in other words, due to energy conservation in the stationary "atom + field" system.

If $\gamma\ll1$, then the elementary quantum $\hbar\omega$ is small, and the quiver energy is much larger than even the ionization potential. In that case the nominally discrete energy spectrum of the emitted electrons is almost continuous. If we are not interested in that fine structure, we can neglect the interference between contributions of different steepest descent points t_i.

On the contrary, for $\gamma\gg1$, the interference of terms with different t_i plays a very important role, as will be seen below.

In a technical sense, the remainder of the present paper deals with the search for the roots of Eq. (11), substitution of these roots into Eq. (10), and calculation of the sum (10) and its square modulus. The particular field $\mathbf{E}(t)$ will be taken to be of the type of Eq. (3) with some smooth envelope $f(t)$, i.e.,

$$\mathbf{A}(t)=[cf(t)/2][(\mathbf{E}_1/i\omega)\exp(-i\omega t)+(\mathbf{E}_2/2i\omega)\exp(-2i\omega t)]+\text{c.c.}\ , \qquad (15)$$

where $f(t)\approx 1$ near the peak of the pulse. Even for this very simplified approximation, the resulting ionization probability $W\sim|a|^2$ still depends on a large number (formally 17) of real variables $W=W(\omega,I,\mathbf{p},\text{Re}\mathbf{E}_1,\text{Im}\mathbf{E}_1,\text{Re}\mathbf{E}_2,\text{Im}\mathbf{E}_2)$ and constants e, $\hbar$, m. [We consider the nonrelativistic problem, so the light velocity c appears in the definition of $\mathbf{A}$ in Eq. (8) in the esu system. There will be no c in the final answer if expressed via $\mathbf{E}_1$ and $\mathbf{E}_2$.]

We hope, however, that there is physical content in our paper which leads to an understanding of the types of processes which correspond to one or another particular domain of the space of variables. We will try also to elucidate the physical effects which are the manifestation of the intrinsic polar asymmetry of the field, Eq. (3).

III. QUASISTATIC TUNNELING LIMIT, OSCILLATIONS OF A FREE ELECTRON, AND RESIDUAL VELOCITY

Consider the motion of a free electron in a spatially homogeneous oscillating electric field $\mathbf{E}(t)$, described by the vector potential (8). The homogeneity condition ($\langle \mathbf{A}^2\rangle$ does not depend on the spatial coordinates) means that we take into account the ponderomotive potential, but not its spatial gradient. The validity of such a consideration is limited to the case of ultrashort pulses and of laser beams with rather broad focal waists [17]. Newton's second law, $md\mathbf{v}/dt=-e\mathbf{E}=(e/c)(\partial\mathbf{A}/\partial t)$ has the evident solution $(e>0)$

$$\mathbf{v}(t)=(e/mc)[\mathbf{A}(t)-\mathbf{A}(t_0)]+\mathbf{v}(t_0)\ . \qquad (16)$$

For an optical pulse with no dc component of the field, one has $\mathbf{A}(t=-\infty)=\mathbf{A}(t=+\infty)=0$, and therefore the velocity of an electron which remains after the action of the light pulse is equal to

$$\mathbf{u}\equiv\mathbf{v}(t=+\infty)=\mathbf{v}(t_0)-(e/mc)\mathbf{A}(t_0)\ . \qquad (17)$$

Thus the residual velocity $\mathbf{u}$ is determined by $\mathbf{A}(t_0)$ and $\mathbf{v}(t_0)$ at the moment t_0 of the electron's emergence. This presumes that the electron may be considered as free starting at t_0. (See Fig. 3.)

These simple considerations have important consequences for the tunneling ionization of an atom. Namely, one can assume that just after emergence from the barrier the electron has almost zero velocity, $\mathbf{v}(t_0)\approx 0$. In that case, the probability distribution $W(\mathbf{u})$ of the residual velocity is given by

$$W(\mathbf{u})=\text{const}\times\int dt\,\exp[-B/|\mathbf{E}(t)|]\times\delta^{(3)}(\mathbf{u}+(e/mc)\mathbf{A}(t))\ . \qquad (18)$$

The most important contributions to the ionization probability arise from the vicinity of the points t_α, where $|\mathbf{E}(t_\alpha)|$ has local maxima (Fig. 3). One can use the expansion for t near t_α,

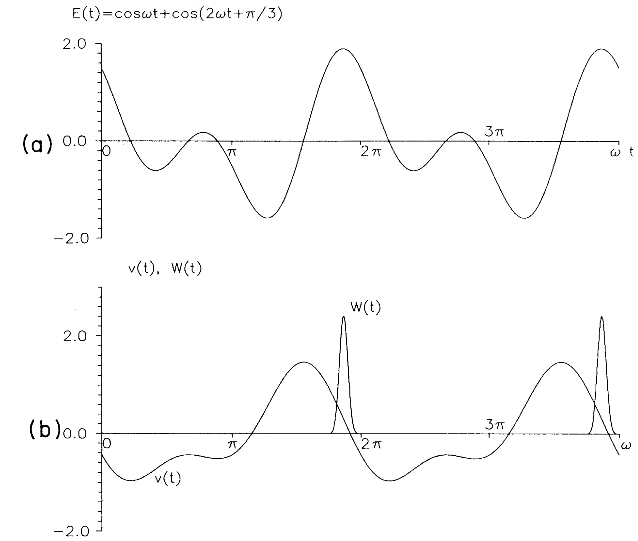

FIG. 3. (a) Superposition of the field of frequency ω and its second harmonic with a $\pi/3$ phase shift. (b) The solid line gives the value $v=(-\omega/E_{\text{at}})\int_t^\infty E(t')dt'$ proportional to the residual velocity of an electron produced by the field shown in (a) as a function of the time t of the electron's appearance. The dotted curve shows the quasistatic probability of tunneling ionization $W=\exp(-2E_{\text{at}}/3|E|)$ for the field in (a), with $E_{\text{at}}=10^2$.

$$-\frac{B}{|\mathbf{E}(t)|}\approx-\frac{B}{|\mathbf{E}(t_\alpha)|}-\frac{B}{|\mathbf{E}(t_\alpha)|}\frac{D^2}{4}(t-t_\alpha)^2+\cdots$$
$$D^2=-\frac{1}{\mathbf{E}^2}\frac{d^2}{dt^2}(\mathbf{E}^2)\ , \qquad (19)$$

where D has the dimension sec^{-1} (i.e., frequency). The value $e\,\mathbf{A}(t)/mc$ in the δ function in Eq. (18) may be approximately replaced by the expansion $(e/mc)\mathbf{A}(t_\alpha)-(e/m)\mathbf{E}(t_\alpha)(t-t_\alpha)$, and then the time integration can be done to yield the contribution of the αth maximum,

$$W^{(\alpha)}(\mathbf{u})\sim\delta^{(2)}\left[\mathbf{u}_\perp+\frac{e}{mc}\mathbf{A}_\perp(t_\alpha)\right]\times\exp\left\{-\frac{[\mathbf{u}_\parallel+(e/mc)\mathbf{A}_\parallel(t_\alpha)]^2}{2\Delta^2}\right\}\ , \qquad (20)$$

where

$$\Delta=[2|\mathbf{E}(t_\alpha)|/B]^{1/2}[e|\mathbf{E}(t_\alpha)|/Dm]\ .$$

We have introduced in Eq. (20) the subscripts $\parallel$ and $\perp$, which denote the components of vectors along ($\parallel$) the direction of $\mathbf{E}(t_\alpha)$ and perpendicular ($\perp$) to it. We see that the residual velocity $\mathbf{u}$ has the definite value $\mathbf{u}_\perp=(e/mc)\mathbf{A}_\perp(t_\alpha)$ in the direction perpendicular to $\mathbf{E}(t_\alpha)$. D is about ω, the frequency of the field, so that the factor $e|\mathbf{E}(t_\alpha)|/Dm$ in the definition of Δ in Eq. (20) is essentially the quiver velocity $e\mathbf{E}(t_\alpha)/m\omega$. Hence there can be a considerable longitudinal velocity dispersion, of the order of the quiver velocity diminished by the factor $[2|\mathbf{E}(t)|/B]^{1/2}<1$.

IV. POLAR ASYMMETRY OF RESIDUAL VELOCITY FOR TUNNELING IONIZATION

Since the field in Eq. (3) or (15) possesses polar asymmetry, it is plausible that the distribution of the residual velocity will possess it also. We shall show that this is so in fact.

It is easy to calculate the velocity distributions $\langle v_i \rangle$, $\langle v_i v_k \rangle$, etc. using the instantaneous probability (20). Moreover, it is not difficult to take into account the change of the pulse envelope in time and in space near the focal waist. That is the reason we will not make any attempt in this section to present analytical expressions for the asymmetry effects. It is evident that polar asymmetry vanishes for a monochromatic field, i.e., in cases when either $\mathbf{E}_1=0$ or $\mathbf{E}_2=0$. Contrary to intuition, the field

$$\mathbf{E}(t)=\mathbf{e}_x(\cos\omega t+\cos 2\omega t) \tag{21}$$

also gives no polar asymmetry in the approximation of $W(t)$ given in Eqs. (18) and (20). Moreover, for $|\mathbf{E}| \ll E_{\rm at}$ the residual velocity will be almost zero for the field (21).

It is curious that the field with orthogonal linear polarizations

$$\mathbf{E}(t)=\mathbf{e}_x\cos\omega t+\mathbf{e}_y\cos(2\omega t+\phi) \tag{22}$$

produces a distribution of residual velocities with $\langle u_x \rangle=0$, but $\langle u_y \rangle \neq 0$.

The most characteristic feature of the polar asymmetry under discussion is its periodic dependence on the phase shift ϕ between the fundamental field and its second harmonic. Figure 4 shows the dependence of $[\langle u_x^2 \rangle]^{1/2}$ and $\langle u_x \rangle$ on the phase ϕ for the field $\mathbf{E}(t)=\mathbf{e}_x[\cos\omega t+\cos(2\omega t+\phi)]$ and $B=(\frac{2}{3})10^2$.

It is worth mentioning that, contrary to common intuition, polar asymmetry appears even in the case when both waves $\mathbf{E}_1$ and $\mathbf{E}_2$ are circularly polarized (see Fig. 5).

The last comment is connected with the possibilities which arise with intersecting optical beams. For monochromatic radiation, that results in an interference (or possibly speckle) pattern. For the ideally coherent monochromatic field, the polarization at any given point is of an elliptical type with some certain plane of that ellipse.

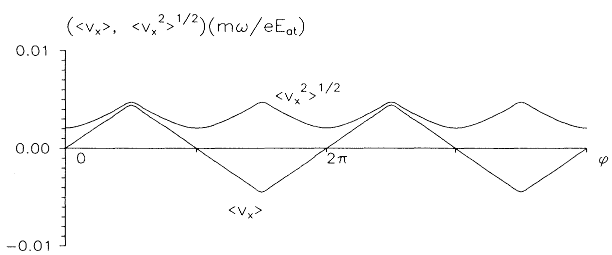

FIG. 4. Dependence of the average residual velocity of electrons $\langle u_x \rangle$ and $[\langle u_x^2 \rangle]^{1/2}$ on the phase shift ϕ for the field $\mathbf{E}=\mathbf{e}_x[\cos\omega t+\cos(2\omega t+\phi)]$ in units of $E_{\rm at}e/m\omega$, calculated via the quasistatic probability of tunneling ionization $W=\exp(-2E_{\rm at}/3|E|)$, with $E_{\rm at}=10^2$.

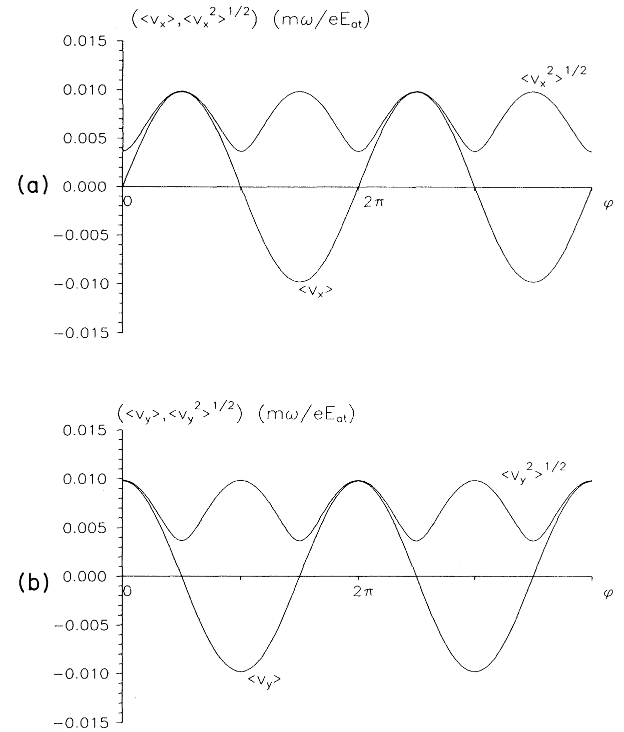

FIG. 5. Dependence of the average residual velocity of electrons on the phase shift ϕ for the bichromatic field with circular polarization $E=\mathrm{Re}[(\mathbf{e}_x+i\mathbf{e}_y)(e^{-i\omega t}+e^{-2i\omega t-1\phi})]/2^{1/2}$ and quasistatic probability $W(t)=\exp(-2E_{\rm at}/3|E|)$, with $E_{\rm at}=10^2$. It is seen that the direction of electron emission is governed by the phase ϕ. (a) $\langle u_x \rangle$, $[\langle u_x^2 \rangle]^{1/2}$; (b) $\langle u_y \rangle$, $[\langle u_y^2 \rangle]^{1/2}$.

However, for a bichromatic field, the planes of the corresponding ellipses may not be coincident. It is in this case where the three-dimensional nature of the electric field vector in the optical wave may be revealed.

Thus, in this section we have shown that the residual velocity for tunneling ionization by a field with $\langle E^3 \rangle \neq 0$ possesses polar asymmetry. Contrary to intuition, the maximum of the velocity asymmetry does not coincide with the maximum of $\langle E^3 \rangle$, but with the maximum of something like $\langle E^2(t)\int E(t')dt' \rangle$. The physical reason for this is the time asymmetry of the detachment process as opposed to the attachment process.

V. DERIVATION OF THE TUNNEL LIMIT FORMULA FROM SIMPLIFIED TIME INTEGRATION MODEL

Suppose that the parameter $\gamma=\omega(2mI)^{1/2}/e|\mathbf{E}|$ is small. In that case, the roots t_i of Eq. (11) for the saddle points in the complex plane $t=t'+it''$ are very near the real points t_α of the local maxima of $|\mathbf{E}(t)|$. Denoting $t_i=t_\alpha+f+ig$, one can express $\mathbf{A}(t_i)$ in Eq. (11) as $\mathbf{A}(t_i)\approx \mathbf{A}(t_\alpha)-c\mathbf{E}(t_\alpha)(f+ig)+\cdots$. Then Eq. (11) has the simple solution

$$f=m(\mathbf{E}\cdot\mathbf{u})/(e\mathbf{E}^2)\ ,$$
$$g=[2m(I+m\mathbf{u}_\perp^2/2)/e^2\mathbf{E}^2]^{1/2}\ . \tag{23}$$

Here and below in this section, $\mathbf{E}$ denotes the vector $\mathbf{E}(t_\alpha)$ at the real maximum of the field strength, and we have introduced the notations

$$m\mathbf{u}=[\mathbf{p}+(e/c)\mathbf{A}(t_\alpha)] \ , \quad \mathbf{u}_\perp=\mathbf{u}-(\mathbf{u}\cdot\mathbf{E})\mathbf{E}/\mathbf{E}^2 \ . \tag{24}$$

Substitution of these values into the "action" in Eq. (10) yields

$$\begin{aligned} a^{(j)}&=\exp[iS(t_j)/\hbar] \\ &=\exp[(i/\hbar)\mathrm{Re}S(t_j)]\exp\{-\widetilde{B}/[2|\mathbf{E}(t_\alpha)|]\} \ , \\ B&=(2/e\hbar)[2m(I+m\mathbf{u}_\perp^2/2)^3]^{1/2} \ . \end{aligned} \tag{25}$$

Since for $\gamma \ll 1$ the ionization potential $I=\gamma^2E^2e^2/2m\omega^2$ is much smaller than the oscillation energy of the free electron $E^2e^2/2m\omega^2$, any considerable (about $eE/m\omega$) deviation of the perpendicular residual velocity $\mathbf{u}_\perp$ from the value given by Eq. (24) results in an extremely strong decrease of the probability $|a^{(j)}|^2$. That is just the $\delta^{(2)}(\mathbf{u}_\perp+e\,\mathbf{A}_\perp/mc)$ from the preceding section. In this simplest approximation, the value of $\mathbf{u}_\parallel$ may take any value with the same probability. To get the result (20) for $\mathbf{u}_\parallel$, one should calculate the corrections $f+ig$ to the root position with one step higher precision. We will not display these calculations here.

Let us justify now the assumption that it is possible to use the lowest-order Taylor expansion for $\mathbf{A}(t)$ in the complex plane. If $\mathbf{u}_\parallel$ is about $(eE/m\omega)(2E/B)^{1/2}$, then

$$f\sim\omega^{-1}(2E/B)^{1/2} \ , \quad g\sim\omega^{-1}\gamma \ . \tag{26}$$

These corrections (especially the imaginary part g) of the dimension sec, should be compared with the characteristic time scale $T/2\pi=\omega^{-1}$ of the field. We see that we need a very moderate ($g \ll T$) analytical continuation and first-Taylor-term estimation of the potential $\mathbf{A}(t)$ into the complex plane from the real axis.

It is worth mentioning that none of the derivations of this section make use of any time periodicity of the field, and hence may be applied to any radiation, even a broadband one.

VI. ANTIADIABATIC LIMIT FOR BICHROMATIC FIELD

For the field (3), Eq. (11) may be written with the notation $z=\exp(-i\omega t_i)$ as

$$\begin{aligned} I+\frac{e^2\mathbf{E}_1\cdot\mathbf{E}_1^*}{4m\omega^2}+\frac{e^2\mathbf{E}_2\cdot\mathbf{E}_2^*}{4m(2\omega)^2}+\frac{\mathbf{p}^2}{2m}-\frac{ie}{4m\omega}[2(\mathbf{p}\cdot\mathbf{E}_1)z+(\mathbf{p}\cdot\mathbf{E}_2)z^2-2(\mathbf{p}\cdot\mathbf{E}_1^*)z^{-1}-(\mathbf{p}\cdot\mathbf{E}_2^*)z^{-2}] \\ -\frac{e^2}{32m\omega^2}[4(\mathbf{E}_1\cdot\mathbf{E}_1)z^2+(\mathbf{E}_2\cdot\mathbf{E}_2)z^4+4(\mathbf{E}_1^*\cdot\mathbf{E}_1^*)z^{-2}+(\mathbf{E}_2^*\cdot\mathbf{E}_2^*)z^{-4}+4(\mathbf{E}_1\cdot\mathbf{E}_2)z^3 \\ +4(\mathbf{E}_1^*\cdot\mathbf{E}_2^*)z^{-3}-(\mathbf{E}_1^*\cdot\mathbf{E}_2)z-(\mathbf{E}_1\cdot\mathbf{E}_2^*)z^{-1}]=0 \ . \end{aligned} \tag{27}$$

This is an equation of eighth power in z. If $\mathbf{E}_2=\mathbf{0}$, the equation is reduced to the fourth power in z, and if $\mathbf{E}_1=\mathbf{0}$, it is reduced to the fourth power in $y=z^2$.

We expect that the integral, Eq. (9), of an analytical function which has very rapid oscillations on the real axis of t will be exponentially small. Analysis shows that the saddle points are given by the solutions of Eq. (9) or (27) with $\mathrm{Im}t_i>0$, i.e., with $|z|>1$. For the case when $\gamma \gg 1$, the actual value of $|z|$ for the roots of (27) is much larger than one, $|z|\gg 1$. Now we shall discuss the physical meaning of these roots.

For the case $\gamma \gg 1$ one should expect the straightforward multiphoton picture of ionization. If we denote

$$\widetilde{I}=I+\frac{e^2\mathbf{E}_1\cdot\mathbf{E}_1^*}{4m\omega^2}+\frac{e^2\mathbf{E}_2\cdot\mathbf{E}_2^*}{4m(2\omega)^2} \ , \tag{28}$$

then the allowed values of $\mathbf{p}^2/2m=K$ are determined by the interference of contributions from several time periods, so that $K=\mathbf{p}^2/2m=-\widetilde{I}+N_1\hbar\omega$, where N_1 is an integer. See Eq. (14). When $\gamma \gg 1$, the kinetic energy is much smaller than I. Therefore a crude estimate for N_1 is $N_1\approx\widetilde{I}/\hbar\omega$. The possible processes are classified by diagrams of the type shown in Fig. 6, so that

$$n_1\hbar\omega+n_2\hbar(2\omega)=N_1\hbar\omega=\widetilde{I}+\mathbf{p}^2/2m \ . \tag{29}$$

Here we shall not go into details of the very interesting matter of above-threshold ionization and the meaning of the oscillation (quiver) energy. That subject has been widely discussed for monochromatic light and for bichromatic radiation with $\omega_1 \ll \omega_2$. See, for example, Ref. [7].

Calculation of the roots of Eq. (7) is simplified by the assumption $|z|\gg 1$. For a monochromatic field $\mathbf{E}_1\neq\mathbf{0}$, $\mathbf{E}_2=\mathbf{0}$, one gets from Eq. (27), when terms $\sim z^{-1}$, z^{-2} are omitted,

$$z_\pm^{(0)}=2\Gamma(-iQ\pm 1) \ . \tag{30}$$

The following notation has been introduced:

$$\begin{aligned} \Gamma&=\frac{\omega}{e}\left\{\frac{2mI+\mathbf{p}^2+e^2\mathbf{E}_1\cdot\mathbf{E}_1^*/2\omega^2-(\mathbf{p}\cdot\mathbf{a})^2}{E_1\cdot E_1}\right\}^{1/2} , \\ Q&=(\mathbf{p}\cdot\mathbf{a})[2mI+\mathbf{p}^2+e^2\mathbf{E}_1\cdot\mathbf{E}_1^*/2\omega^2-(\mathbf{p}\cdot\mathbf{a})^2]^{-1/2} \ , \\ \mathbf{a}&=\mathbf{E}_1/(\mathbf{E}_1\cdot\mathbf{E}_1)^{1/2} \ . \end{aligned} \tag{31}$$

It should be noted that for general elliptical polarization we have $\mathbf{a}\cdot\mathbf{a}^*>1$. We wish to note here the special case of almost-circular polarization, when $\mathbf{E}\cdot\mathbf{E}\to 0$. We shall see that in that case our expressions give almost vanishing ionization probability. That is known to be attributable to the repelling action of the centrifugal barrier for the final state, since the latter should have an angular momentum equal to the number of absorbed quanta.

For a linearly polarized field $\mathbf{E}_1$, the parameter Γ turns out to be real, and in the case $\mathbf{p}=0$ and the substitution $\tilde{I}\to I$, the value of Γ coincides with the usual Keldysh parameter γ of Eq. (1). If for some reason the momentum $\mathbf{p}$ is negligibly small, then the real parts of the two roots $\mathrm{Re}(\omega t_1)$ and $\mathrm{Re}(\omega t_2)$ correspond to the local maxima of the field on the real axis. It should be emphasized, however, that for $\gamma \gg 1$ the imaginary part $\mathrm{Im}(\omega t_j)$ of the root is much larger than 2π. That means that one cannot attribute the N-photon ionization to a definite moment of time within the period, quite in accord with the energy versus time uncertainty principle.

For $\mathbf{p}\neq 0$, the substitution of the roots (30) into the sum within one $2\pi/\omega$ period yields

$$a^{(0)}(\mathbf{p})=C(\mathbf{p})\{[1+(-1)^{N_1}]\cos(N_1\Phi)+i[1-(-1)^{N_1}]\sin(N_1\Phi)\}\ ,$$

$$C(\mathbf{p})=\exp\left[\frac{N_1}{2}+\frac{(\mathbf{p}\cdot\mathbf{a})^2}{2m\hbar\omega}\right](2\Gamma)^{-N_1}\ ,\quad \Phi=Q+\arctan Q\ . \tag{32}$$

It is important for subsequent work that Q (and Φ) are odd functions of the momentum $\mathbf{p}$.

We see that terms of even power in $\mathbf{p}$ in the amplitude (32) require the absorption of even numbers of photons N_1, and correspondingly the terms which are odd in $\mathbf{p}$ need an odd number N_1 of quanta $\hbar\omega$. That means that for the given number of absorbed photons $N_1\hbar\omega$, and hence for the given kinetic energy K, the amplitude $a(\mathbf{p})$ is either an odd or even function of $\mathbf{p}$ (has definite parity). Therefore the probability distribution $W(\mathbf{p})\sim|a(\mathbf{p})|^2$ possesses inversion symmetry $W(\mathbf{p})=W(-\mathbf{p})$ for the monochromatic radiation $\mathbf{E}_1$.

To elucidate the polar symmetry effects, we shall consider first the case when $\gamma\gg 1$, but $|\mathbf{E}_2|$ much smaller than $|\mathbf{E}_1|$. Then the main contribution to the process is due to N_1-photon ionization by the $\mathbf{E}_1$ field, i.e., $n_1\approx I_1/\hbar\omega$, $n_2=0$. The influence of the 2ω field will be taken into account as a small perturbation to the roots (30), (31), and amplitudes (32).

The calculations, which are very simple in principle but somewhat lengthy, give the modified values $z_\pm$ of two roots with terms of the first order in $\mathbf{E}_2$:

$$z_\pm=z_\pm^{(0)}\left\{1\pm\frac{\omega}{e(\mathbf{E}_1\cdot\mathbf{E}_1)\Gamma}\left[-i\mathbf{p}\cdot\mathbf{E}_2 z_\pm^{(0)}+\frac{e}{2\omega}(\mathbf{E}_1^*\cdot\mathbf{E}_2)-\frac{e}{2\omega}(\mathbf{E}_1\cdot\mathbf{E}_2)(z_\pm^{(0)})^2\right]\right\}\ . \tag{33}$$

The next step is calculation of the sum $\sum'$ for the amplitude $a(\mathbf{p})$ in Eq. (12) within a single period $T_1=2\pi/\omega$, up to terms of the first order in $\mathbf{E}_2$. When expressed in terms of $K=N_1\hbar\omega-\tilde{I}$, one gets more felicitous expressions when written for even N_1 and odd N_1 separately. For even N_1 one obtains

$$a(\mathbf{p})=2C(\mathbf{p})[\cos(N_1\Phi)+iN_1\lambda\sin(N_1\Phi)]\ , \tag{34a}$$

and for odd values of N_1

$$a(\mathbf{p})=2C(\mathbf{p})[i\sin(N_1\Phi)+N_1\lambda\cos(N_1\Phi)]\ , \tag{34b}$$

where we have introduced the variable λ proportional to the first power of the field $\mathbf{E}_2$,

$$\lambda=(2m\tilde{\tilde{I}})^{-1/2}\left[\frac{2\omega}{e}\frac{(\mathbf{p}\cdot\mathbf{E}_2)(\mathbf{p}\cdot\mathbf{E}_1)}{(\mathbf{E}_1\cdot\mathbf{E}_1)^{3/2}}+\frac{2\omega}{3e}\frac{\mathbf{E}_1\cdot\mathbf{E}_2}{(\mathbf{E}_1\cdot\mathbf{E}_1)^{3/2}}(2m\tilde{\tilde{I}})-\frac{e}{2\omega}\frac{\mathbf{E}_1^*\cdot\mathbf{E}_2}{(\mathbf{E}_1\cdot\mathbf{E}_1)^{1/2}}\right]\ , \tag{35}$$

$$\tilde{\tilde{I}}=I+\mathbf{p}^2/2m+e^2\mathbf{E}_1\cdot\mathbf{E}_1^*/4m\omega^2-(\mathbf{p}\cdot\mathbf{a})^2/2m\ .$$

Now we can attribute different terms in the expression (35) for λ to particular processes depicted in Fig. 6. Namely, the first and second terms correspond to ionization by the combination of photons $(N_1-2)\hbar\omega+1(2\hbar\omega)$; the third term corresponds to the absorption of $(N_1-1)\hbar\omega+1(2\hbar\omega)$ photons with the subsequent stimulated emission of one $\hbar\omega$ photon. That third term, after substitution into $a(\mathbf{p})$, has the relatively small amplitude $\sim(E_1/E_0)^{N_1}(E_2/E_0)$, while the second term is of the order of $\sim(E_1/E_0)^{N_1-2}(E_2/E_0)$, and the first is about $\sim(E_1/E_0)^{N_1-2}(E_2/E_0)(\mathbf{p}^2/2mI)$. Here $E_0=(\omega/e)(2m\tilde{\tilde{I}})^{1/2}$ is the field which is used to make the powers of E_1 or E_2 dimensionless. When $E_1\sim E_0$ the quiver energy is about the ionization potential, and for such high fields we would have $\gamma\sim 1$. It should be remembered that we consider here the case $\gamma\gg 1$, the zeroth term is of the order $\sim(E_1/E_0)^{N_1}$, and the E_1 dependence just corresponds to N_1-photon ionization by the E_1 field.

We conjecture that the description of the process with two and more 2ω photons included can also be obtained with first-order precision for the roots $z_\pm$ in Eq. (33), but with higher-order Taylor terms for the amplitude $a(\mathbf{p})$.

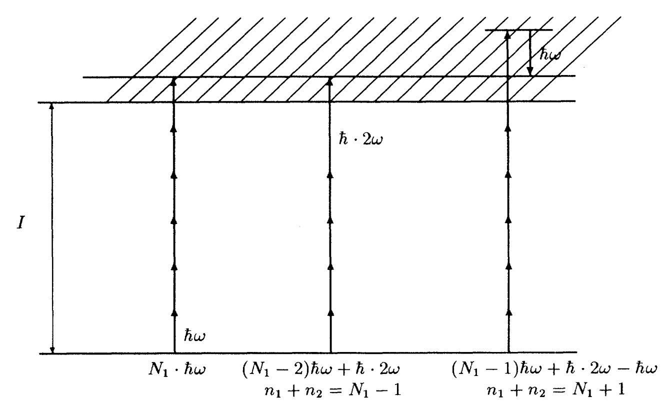

FIG. 6. Possible channels of multiphoton ionization by the bichromatic field $\mathbf{E}=\frac{1}{2}[\mathbf{E}_1e^{-i\omega t}+\mathbf{E}_2e^{-2i\omega t}]+\mathrm{c.c.}$ Any scheme includes all possible permutations of photons. We consider the case $|\mathbf{E}_1|\gg|\mathbf{E}_2|$, when the $N_1\hbar\omega$ process is the strongest one and the processes in which a $2\hbar\omega$ photon participate are given by first-order perturbation theory.

However, this question needs more accurate consideration, which we shall postpone to the future. We should also like to note that the condition stating the weakness of the contribution of the E_2 field to the ionization probability in comparison with that of the E_1 field is $E_2 \le E_1(E_1/E_0) \sim E_1/\gamma_1$, where γ_1 is the parameter of Eq. (1) calculated for the E_1 field.

If $E_2 \ge E_1/\gamma_1$, then the main contribution to the ionization probability is due to n_2-photon ionization by the 2ω field, with $n_2 \approx \tilde{I}/2\hbar\omega$, and $n_1 = 0$ or $n_1 = 1$. In that case, the zeroth approximation for solving Eq. (31) corresponds to omission of all the terms $\sim E_1$ and $\sim E_1^2$. The resulting equation is identical to the previous case with the substitution $z \to y = z^2$, $E_1 \to E_2$, $\omega \to 2\omega$. That means that in the zeroth approximation in E_1, we now have four roots for z at a "large" period $T = 2\pi/\omega$, i.e., two equivalent pairs shifted by $T/2$ from each other. Switching on the field E_1 will result in splitting of these roots, making all of the four roots nonequivalent. We hope to give the detailed analysis of that limiting case in the future.

VII. POLAR ASYMMETRY OF VELOCITY DISTRIBUTION FOR ANTIADIABATIC LIMIT

In this section we will discuss the effects of polar asymmetry as manifested by the interference of N- and $(N \pm 1)$-photon ionization. A large variety of interference effects have been discussed in the literature, including interference effects in multiphoton ionization [5,9,11–14,17–25]. However, the most widely considered case is the superposition of the fundamental frequency field $E_1 \cos(\omega t + \phi_1)$ and its third harmonic $E_3(\cos\omega t + \phi_3)$. A specific feature of the field is that it possesses true polar symmetry: all the time-averaged odd momenta are zero, $\langle E^{2k+1} \rangle = 0$. Then a state of given energy I_c in a continuum may be excited by the next combination of quanta: $I_c = n(3\hbar\omega) + (N - 3n)\hbar\omega$; $N = \text{const}$. Therefore for any value of n the number of absorbed quanta $N - 2n$ is even if N is even, and conversely, it is odd if N is odd. Due to dipole selection rules it means that all the components of the wave function of the outgoing electron have the same parity, e.g., $s, d, \ldots$ states or $p, f, \ldots$ states. Interference of processes with different n leads to very interesting effects both for the total probability and for the angular distribution of the emitted electrons. However, no polar asymmetry may appear due to interference of states with the same parity.

On the contrary, the field $E(\omega) + E(2\omega)$, Eq. (3), has polar asymmetry and can mix states of opposite parity. For example, polar asymmetry emitted electrons resulting from the interference of single-photon absorption of 2ω-radiation and two-photon absorption of ω radiation was predicted theoretically [25,11] and observed experimentally [25,13,14] for both solid-state and atomic gas media. Interference of seven- and eight-photon ionization of krypton gas was observed in Ref. [9].

The aim of this section is to show that even the extremely simplified model of Sec. II makes possible a discussion of polar asymmetry effects in the antiadiabatic regime. That model, since it is written down without any preexponential factors, is not intended to give a quantitative description of experimental data.

From Eq. (35) we see that λ is an even function of the momentum $\mathbf{p}$. Therefore, for N_1 even [Eq. (34a)], the λ contributions of the processes $(N_1 - 2)\hbar\omega + 1(2\hbar\omega)$ and $(N_1 - 1)\hbar\omega + 1(2\hbar\omega) - 1(\hbar\omega)$ have odd parity in $\mathbf{p}$, i.e., this parity just coincides with the parity of the total number of photons taking part in the process. An analogous situation is for odd N_1, where the zeroth-order amplitude [see Eq. (34b)] is an odd function of $\mathbf{p}$ and the E_2 correction is an even one. Hence the λ terms $\sim E_2$ have $\mathbf{p}$-parity opposite to the zeroth $N_1\hbar\omega$ term of the amplitude. It means that the probability acquires the polar asymmetric term due to interference which sinusoidally depends on the phase difference $\arg(E_1^2 E_2^*)$,

$$W(\mathbf{p}) = W^{(0)}(\mathbf{p})[1 + F(\mathbf{p})E_2/E_1^2 + F^*(\mathbf{p})E_2^*/E_1^{*2}] , \tag{36}$$

where $F(\mathbf{p})$ is an odd and $W^{(0)}(\mathbf{p})$ is an even function of the momentum $\mathbf{p}$. If the polarizations of $\mathbf{E}_1$ and $\mathbf{E}_2$ are linear and parallel to each other, then the values of Q, Φ, and $\mathbf{a}$ are real. In that case, the function $F(\mathbf{p})$ is purely imaginary, and we come to an important conclusion: the maximum polar asymmetry of the emitted electron's velocity distribution is achieved for $\arg(E_1^2 E_2^*) = \pm\pi/2$.

The probability of emission of an electron must be invariant under the time-reversal operation if we neglect the interaction of the electron with the atom in the final state [see Eq. (6)]. The reversal corresponds to the substitution $\mathbf{E} \to \mathbf{E}^*$, $\mathbf{p} \to -\mathbf{p}$. Therefore the terms $\sim G\mathbf{p}E_1^2 E_2^* + G^*\mathbf{p}E_1^{*2}E_2$ must have a purely imaginary coefficient G if we want them to be invariant under the $t \to -t$ reversal operation. Just such a situation arises in the case of interference between single-photon ionization by the field E_2 and two-photon ionization by the field E_1 for $\hbar\omega < I < 2\hbar\omega$ if we can neglect the interaction of the liberated electron with the atomic residue. See Ref. [7]. However, if the influence of the atomic potential on the final state is taken into account, the $t \to -t$ invariance of the expression for $W(\mathbf{p})$ is violated, and a quantum-mechanical scattering phase is added to the corresponding phase of the coefficient G. See Refs. [7,11,24].

VIII. DISCUSSION AND CONCLUSIONS

Recently, polar asymmetry of ionization has been observed in experiments with a photomultiplier [25,12], with krypton gas [9], with a sodium atomic beam [13], and with rubidium [14]. In the experiments of Refs. [25,12,9,13] the quanta $\hbar\omega$ and $2\hbar\omega$ corresponded to the Nd:YAG wavelength $\lambda = 1.06$ μm (where YAG denotes yttrium aluminum garnet) and its second harmonic $\lambda = 0.53$ μm utilized with picosecond pulses. In Ref. [14], the quanta $\hbar\omega$ and $2\hbar\omega$ were obtained from a tunable laser. For the case of sodium atoms [13], the initial state was $4S$, and there was virtual resonance for the transition $4S \to 5P$ ($\lambda = 1.069$ μm). That fortuitous circumstance led to essential enhancement of the two-photon amplitude.

In this paper we have tried to show that polar asymmetry of the angular distribution of photons emitted either by a tunnelinglike process or by a multiphoton process must appear as a result of polar asymmetry of the field (3) with nonzero cubic moment $\langle E_1^2 E_2^* \rangle$.

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