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Photoionization by a bichromatic field: Adiabatic theory

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Atom photoionization by the superposition of a fundamental field and its second harmonic is considered. The finite analytical expressions for the photoionization probability are obtained using the adiabatic approximation. They demonstrate that the photoelectron angular distribution has a polar symmetry when the electrical field strength has a maximal polar asymmetry and the distribution is asymmetrical when the field is symmetrical. A strict proof of the polar symmetry of the photoionization probability in the case of the electrical field with maximal asymmetry is deduced using the Keldysh-Faisal-Reiss theories. The obtained results are in agreement with the experimental data available.

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I. INTRODUCTION

Ionization of atoms and photodecay of negative ions in a bichromatic field have recently received more attention, in both experiment and theory. The polar asymmetry of the photoelectron angular distribution by the two-color laser radiation atom photoionization, observed first in Ref. [1] (see also [2,3]), is of special interest. The linearly polarized laser field consisting of the coherent superposition of the fundamental field and its second harmonic was used in these experiments. The observed effect essentially depends on the phase harmonic shift and is the consequence of quantum interference of several ionization paths. A similar effect was found in Ref. [4] by two-color photocathode irradiation.

The considered effect was analyzed theoretically in the range of validity of the perturbative treatment for laser field in Refs. [4–6]. The strong field region was studied using numerical simulation in Refs. [7,8], but a numerical experiment can not contribute much to the understanding of the behavior of that effect. The analytical expressions for atom ionization probability in the strong bichromatic field were obtained using tunneling approaches: the "imaginary time" approach [9] (before the first experimental results [1] appeared) and the estimation by tunneling probability [10]. The so-called Keldysh-Faisal-Reiss (KFR) theory was employed in [10] as well. The tunneling approaches used in Refs. [9] and [10] are incorrectly founded since they are based only on a qualitative semiclassical picture of the photoionization process. The quantum interference is not included in such approaches.

In the present paper the photoionization probability of a atom by the bichromatic field is obtained using the KFR theory. Nowadays such theories (see Refs. [11–15] and a review in [16]) are the only analytical nonperturbative approach to the qualitative description of the observed photoionization behavior. Using such theories the energy and the angular electron spectra for the atoms ionized by monochromatic field [12,13] and above-threshold photoelectron spectra (see, for example, Refs. [17,18]) are obtained.

The paper is organized as follows. General expressions for the evaluation of the photoionization probability using the Landau-Dykhne adiabatic approximation are given in Sec. II. The photoionization probability in the limiting tunneling case is considered in Sec. III, and the limiting multiphoton case in Sec. IV. It is demonstrated that in both limiting cases the photoelectron angular distribution has a polar symmetry at the phase shift when $\langle E^3\rangle \neq 0$ and $\langle A^3\rangle = 0$ and that this symmetry disappears in the case when $\langle E^3\rangle = 0$ and $\langle A^3\rangle \neq 0$ (the angular brackets denote time averaging). Hence the polar symmetry of the photoionization probability is correlated to the polar symmetry of the vector potential rather than the electrical field symmetry. An interesting analogy to the well-known Aharonov-Bohm effect (see [21]) may be drawn here.

To validate such a paradoxical result, a strict analysis of the particular case of the antisymmetrical vector potential $[\mathbf{A}(-t) = -\mathbf{A}(t)]$ has been performed in Sec. V (the antisymmetrical vector potential corresponds to the maximal polar asymmetry of the electrical field). The polar symmetry of the photoelectron angular distribution by photoionization in this field is proved by applying only the adiabatic approximation without using any additional approximations. This statement is also proved using the Nikishov-Ritus-Faisal-Reiss approach, making no use of the adiabatic approximation.

The obtained result is in contrast with the tunneling approaches [9,10], but it is in agreement with the KFR theory [10] and with the perturbation theory computation [4]. It is also in accordance with the experiment [2], the only experiment where the absolute value of the phase shift was measured. It does not conflict with the experimental data [1,3], where the absolute value for the phase shift was not measured (for more details, see the discussion in Sec. VI).

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As in any KFR theory, the analysis of the present paper is applicable only to the case when the photoelectron interaction with an ion core may be neglected. However, there are reasons to believe that the influence of such an interaction on the considered effect is small (Sec. VI). The atomic system $e = \hbar = m = 1$ is used below.

II. ADIABATIC APPROXIMATION

The present analysis of atom photoionization in the asymmetric field is based on the approach of Ref. [18], where the Landau-Dykhne adiabatic approximation (see [19,20]) was employed to evaluate the atom photoionization probability by the monochromatic field, taking the photoelectron primary kinetic energy into account. The photoionization probability per unit time is expressed by

$$W = \sum_{N \geq N_0} W_N(\mathbf{p}) \delta(\mathbf{p}^2/2 + K + U_p - N\omega), \quad (1)$$

where K is the ionization potential of the atom, $\mathbf{p}$ is the photoelectron canonical momentum, $U_p = \langle \mathbf{A}^2 \rangle / (2c^2)$ is the pounderomotive potential, $\mathbf{A}(t)$ is the vector potential of the electromagnetic field, $N_0 = [K/\omega] + 1$ is the minimal number of photons required for atom ionization, and $[X]$ denotes an integer part of the number X. The photoionization probability with the energy absorption $N\omega$ is

$$W_N(\mathbf{p}) = Cp \left| \sum_k \exp(iS_k) \right|^2, \quad (2)$$

where S_k is the classical action

$$S_k = \int_0^{t_k} \left\{ \frac{1}{2} \left[\mathbf{p} + \frac{1}{c} \mathbf{A}(t) \right]^2 + K \right\} dt \quad (3)$$

and t_k are transition points in complex time where the initial and final state energies are crossed. These points are the roots of the equation

$$\frac{1}{2} \left[\mathbf{p} + \frac{1}{c} \mathbf{A}(t) \right]^2 + K = 0, \quad -\pi/\omega < \text{Re} t_k < \pi/\omega \quad (4)$$

in the upper half of the complex t plane. It should be noted that in the adiabatic approximation, one fails to determine a preexponential factor for W_N that can depend on various parameters. In the following, we will assume that this dependence is weak and an unknown preexponential factor, together with other variables, will be incorporated into the constant C in (2). The factor p in (2) describes the phase volume expansion with an increase in energy of the translational photoelectron motion. The expressions used in Ref. [10] are identical to Eqs. (1)–(4).

The linearly polarized field $\mathbf{A}(t) = \mathbf{e}A(t)$ ($\mathbf{e}$ is the unit vector by the polarization direction) is considered below. In this case the Eq. (4) can be reduced to the quadratic equation in $A(t)$, which can be solved as

$$A(t_k)/c = -p_\parallel \pm i\sqrt{p_\perp^2 + 2K}, \quad (5)$$

where $p_\parallel = (\mathbf{p} \cdot \mathbf{e})$, $p_\perp^2 = \mathbf{p}^2 - p_\parallel^2$.

Nevertheless, the transcendent equation (5) has to be solved to determine the transition points t_k. Even in the case of the bichromatic field

$$A(t) = -\frac{c}{\omega} \left[E_1 \sin \omega t + \frac{E_2}{2} \sin(2\omega t + \varphi) \right], \quad (6)$$

this equation may be reduced only to an algebraic equation of the fourth degree. Because of this, the two limiting cases (tunneling and multiphoton) will be analyzed further. As shown in [18], the tunneling limiting case takes place when the inequality

$$\mathbf{p}^2/2 + K \ll U_P \quad (7)$$

is satisfied and the multiphoton case occurs when the inverse inequality is satisfied. This classification may be reduced to the traditional classification in according to the Keldysh parameter value by neglecting the photoelectron momentum $\mathbf{p}$ in Eq. (7).

III. TUNNELING LIMIT

Since the vector potential $A(t)$ is a periodic function with no constant component, it has zeros ζ_k on a real axis

$$A(\zeta_k) = 0. \quad (8)$$

In the considered case the transition points t_k are in the vicinity of ζ_k (the closeness criteria will be presented below). Therefore, Eq. (5) may be written in the form

$$(t_k - \zeta_k) E(\zeta_k) = p_\parallel \pm i\sqrt{p_\perp^2 + 2K}, \quad (9)$$

where

$$E(\zeta_k) = -\frac{1}{c} \dot{A}(\zeta_k). \quad (10)$$

It is evident from Eq. (9) that the transition point in upper half complex t plane corresponds to every ζ_k:

$$t_k = \zeta_k + p_\parallel / E(\zeta_k) + i\sqrt{p_\perp^2 + 2K} / |E(\zeta_k)|. \quad (11)$$

The sign on the right-hand side of Eq. (9) was chosen in accordance with the sign of $E(\zeta_k)$. Using the $A(t)$ expansion in terms of $(t_k - \zeta_k)/\omega$, Eq. (3) integrates to the expression

$$S_k = \int_0^{\zeta_k} \left[\frac{1}{2} \left(\mathbf{p} + \frac{1}{c} \mathbf{A}(t) \right)^2 + K \right] dt + \frac{p_\parallel}{2E(\zeta_k)} \left(2K + p^2 - \frac{2}{3} p_\parallel^2 \right) + \frac{i}{3|E(\zeta_k)|} \left(2K + p_\perp^2 \right)^{3/2}. \quad (12)$$

The power series for $A(t)$ is used within the region of size $\sim t_k - \zeta_k$; this size is small in comparison with the field period when the inequality (7) is satisfied.

The adiabatic approximation is applicable when the imaginary part of the classical action is sufficiently large. Therefore, only the terms corresponding to the maximal $|E(\zeta_k)|$ value should be retained in the sum (2); the other terms will be exponentially small in relation to them.

Let us consider the bichromatic field (6). Equation (8) may be solved exactly only for some special φ values. In the case of $\varphi = 0$ and $E_1 < E_2$, four solutions of Eq. (8) may be written:

$$\zeta_2 = 0, \quad \zeta_4 = \pi/\omega, \quad E(\zeta_{2,4}) = E_2 \pm E_1,$$
$$\zeta_{1,3} = \mp \frac{1}{\omega}\left(\pi - \arccos \frac{E_1}{E_2}\right), \quad E(\zeta_{1,3}) = -\frac{E_2^2 - E_1^2}{E_2}. \tag{13}$$

When $\varphi = 0$ and $E_1 \geq E_2$, Eq. (8) has two solutions:

$$\zeta_1 = -\pi/\omega, \quad \zeta_2 = 0, \quad E(\zeta_{1,2}) = E_2 \mp E_1. \tag{14}$$

In the case of $\varphi = \pi/2$, Eq. (8) can also be solved exactly. When $E_1 < E_2/2$, the solutions have the form

$$\zeta_1 = -\pi/\omega - \zeta_2, \zeta_2 = -\frac{1}{\omega}\arcsin\left(\frac{\sqrt{E_1^2 + 2E_2^2} - E_1}{2E_2}\right),$$

$$E(\zeta_{1,2}) = \mp\sqrt{\frac{E_1^2 + 2E_2^2}{2E_2^2}\left(E_2^2 - E_1^2 + E_1\sqrt{E_1^2 + 2E_2^2}\right)},$$

$$\zeta_3 = \frac{1}{\omega}\arcsin\left(\frac{\sqrt{E_1^2 + 2E_2^2} + E_1}{2E_2}\right), \quad \zeta_1 = \pi/\omega - \zeta_3,$$

$$E(\zeta_{3,4}) = \mp\sqrt{\frac{E_1^2 + 2E_2^2}{2E_2^2}\left(E_2^2 - E_1^2 - E_1\sqrt{E_1^2 + 2E_2^2}\right)}. \tag{15}$$

When $E_1 \geq E_2/2$, only two of these solutions ($\zeta_{1,2}$) are retained.

Only a numerical solution of Eq. (8) may be obtained at other φ values. Obtained in such a manner, the plots of $E(\zeta_k)/\sqrt{E_1^2 + E_2^2}$ versus φ are shown in Fig. 1 for several values of the parameter $\xi = E_1^2/(E_1^2 + E_2^2)$ (the ratio between intensities of the fundamental frequency and the total field). As follows from Eqs. (6) and (10), $E(\zeta_k)$ has period π and even symmetry in φ. Thus only the interval $0 \leq \varphi \leq \pi/2$ has to be considered in the following. As may be seen from Fig. 1, the $|E(\zeta_2)|$ value is maximal from all $|E(\zeta_k)|$ for all φ and ξ values, and $|E(\zeta_1)| = |E(\zeta_2)|$ at $\varphi = \pi/2$ only. Thus only one term may be retained in the sum (2) for all φ values, except for those close to $\pi/2$. Therefore, at $\varphi \neq \pi/2$ the photoionization probability takes the form

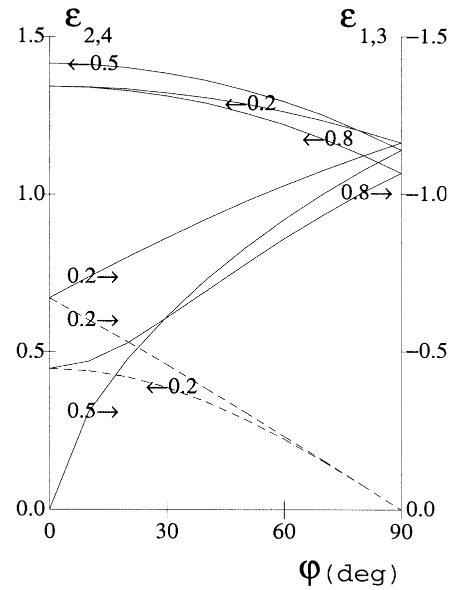

FIG. 1. Plots of $\epsilon_{1,2} = E(\zeta_{1,2})/\sqrt{E_1^2 + E_2^2}$ (solid line) and $\epsilon_{3,4} = E(\zeta_{3,4})/\sqrt{E_1^2 + E_2^2}$ (dashed line) versus phase harmonic shift. Numbers on the curves are values of the parameter $\xi = E_1^2/(E_1^2 + E_2^2)$.

$$W_N(\mathbf{p}) = Cp \exp\left(-\frac{2}{3E_{\text{eff}}}\left(2K + p_\perp^2\right)^{3/2}\right), \tag{16}$$

where $E_{\text{eff}} = |E(\zeta_2)|$. The numerical calculations of E_{eff} are presented in Fig. 2. From this figure we see that E_{eff} and hence W_N have a maximum at $\varphi = 0$. They decrease as φ varies from 0 to $\pi/2$. Notice that E_{eff} and hence $W_N(\mathbf{p})$ have even symmetry and period π in φ.

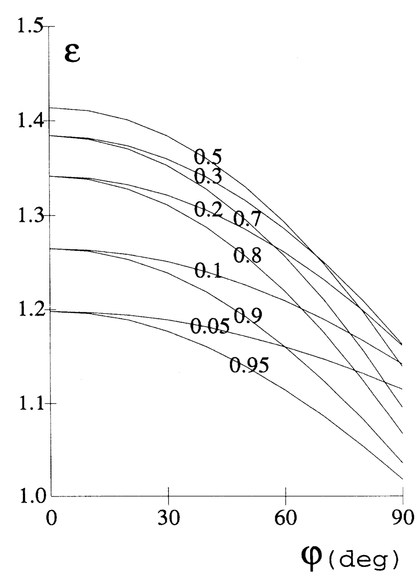

FIG. 2. Plots of $\epsilon = E_{\text{eff}}/\sqrt{E_1^2 + E_2^2}$ versus phase harmonic shift. Numbers on the curves are values of the parameter $\xi = E_1^2/(E_1^2 + E_2^2)$ (the ratio between intensities of the fundamental frequency and total the field).

Equation (8), defining ζ_i, as well as Eq. (10), defining $E(\zeta_i)$, lacks the photoelectron momentum. Therefore, $E_{\rm eff}$ is $\mathbf{p}$ independent and the photoionization probability (16) has polar symmetry in $\mathbf{p}$. The polar asymmetry may be associated only with the terms omitted in the sum (2); consequently it is exponentially small. This result seems to be paradoxical since the electrical field strength has polar asymmetry at all $\varphi \neq \pi/2$, just in the range of applicability of Eq. (16).

Now let us consider the case when $\varphi = \pi/2 - \delta$ ($\delta \ll 1$) and the contributions from both transitions points t_1 and t_2 must be taken into account in Eq. (2). In this case the $A(t)$ roots corresponding to the maximal electrical field strength may be written in the form

$$\zeta'_{1,2} = \zeta_{1,2} + \frac{1}{2\omega}\left(1 - \frac{E_1}{\sqrt{E_1^2 + 2E_2^2}}\right)\delta. \quad (17)$$

The corresponding electrical field strength values are expressed as:

$$E(\zeta'_{1,2}) = E(\zeta_{1,2}) + \frac{E_1}{2E_2}\frac{4E_1(E_1 - \sqrt{E_1^2+2E_2^2}) + 5E_2^2}{\sqrt{E_1^2+2E_2^2}}\delta, \quad (18)$$

where $\zeta_{1,2}$ and $E(\zeta_{1,2})$ are given in Eq. (15). The integral in (12) is evaluated using the upper limit given in Eq. (17). Substituting Eq. (12) into Eq. (2), one may obtain the following expression for the photoionization probability:

$$W_N(\mathbf{p}) = Cp\exp\left(-\frac{2}{3E_{\rm eff}}(2K + p_\perp^2)^{3/2}\right) \times \left[\cosh \mathrm{Im}(S_2 - S_1) + (-1)^N \cos(ap_\parallel - b)\right], \quad (19)$$

where $E_{\rm eff} = E(\zeta_2)$ [see Eq. (15)], and $ap_\parallel - b = \mathrm{Re}(S_1 - S_2) + N\pi$. The relative polar asymmetry of the photoelectron angular distribution may be evaluated as

$$\frac{W_N(\mathbf{p}) - W_N(-\mathbf{p})}{W_N(\mathbf{p}) + W_N(-\mathbf{p})} = \frac{(-1)^N \sin b \sin ap_\parallel}{\cosh \mathrm{Im}(S_1 - S_2) + (-1)^N \cos b \cos ap_\parallel}. \quad (20)$$

In the first nonvanishing order with respect to δ,

$$\mathrm{Im}(S_2 - S_1) = \frac{4}{3}(2K + p_\perp^2)^{3/2} E_{\rm eff}^{-2}\frac{\partial E_{\rm eff}}{\partial \delta}\delta. \quad (21)$$

When this quantity becomes large, the polar asymmetry of the photoionization probability becomes exponentially small [see Eq. (20)]. In the case when $E_1 \sim E_2$, Eq. (18) gives $\partial E_{\rm eff}/\partial\delta \approx E_{\rm eff}$, which is why the polar asymmetry may be observed only at

$$\delta \leq E_{\rm eff}/(2K + p_\perp^2)^{3/2} \approx |\ln W|^{-1}.$$

Without presenting elaborate expressions for a and b, we note that for small δ and $E_1 \sim E_2$ they may be estimated as $b \sim N \gg 1$, and $a \sim E_{\rm eff}/\omega^2$. Thus even minor variations in the field intensities cause the value of b to vary by π, resulting in the change in sign in Eq. (20). Hence, in the considered limiting case at $E_1 \sim E_2$, the polar asymmetry of the photoelectron angular distribution may be observed only within a narrow range of the harmonics phase shift and for high laser intensity stability.

A different situation occurs when the intensity of one harmonic is sufficiently larger than that of another. In this case, as Eq. (18) shows, $\partial E_{\rm eff}/\partial\delta \ll E_1 + E_2$ and, according to Eqs. (20) and (21), the polar asymmetry does not vanish at small δ. Let us consider a particular case of $E_2 \ll E_1$. This case is interesting in view of the interpretation of the experimental results [1], where $E_2 \ll E_1$ in the fringe area of laser focus. In this case the solutions of Eq. (8) and $E(\zeta_k)$ have the form

$$\zeta_k \approx \frac{1}{\omega}[(k-2)\pi - (-1)^k E_2/(2E_1)\sin\varphi], \quad k = 1,2$$

$$E(\zeta_k) \approx (-1)^k E_1 + E_2\cos\varphi\ . \quad (22)$$

Provided

$$\mathrm{Im}(S_1 - S_2) = \frac{2}{3E_1}(2K + p_\perp^2)^{3/2}\frac{E_2}{E_1}\cos\varphi \ll 1$$

is satisfied, using Eqs. (12) and (2), one can obtain the following expression for the photoionization probability:

$$W_N(\mathbf{p}) = 2Cp\exp\left(-\frac{2}{3E_1}(2K + p_\perp^2)^{3/2}\right) \times [1 + (-1)^N\cos(ap_\parallel - b)],$$

$$a = 2E_1/\omega^2 - (2K + p^2 - 2p_\parallel^2/3)/E_1, \quad (23)$$

$$b = \frac{E_2}{E_1}\left(N + \frac{E_1^2}{4\omega^3}\right)\sin\varphi.$$

The analysis of these expressions shows that W_N has extremes at $\varphi = \pm\pi/2 + 2n\pi$ (n is an arbitrary integer). The extreme can be maximum or minimum, depending on the N parity and the $p_\parallel$ value. If $\frac{E_2}{E_1}\left(N + \frac{E_1^2}{4\omega^3}\right) > 3/2\pi$, an additional extreme of W_N in the range $-\pi < \varphi < \pi$ appear. This effect was observed experimentally [1] at some N values. The polar asymmetry of the photoelectron angular distribution vanishes [see (20) and (23)] at $\varphi = 0$, when $\langle E^2\rangle$ is maximal and $\langle A^3\rangle = 0$, and peaks at $\varphi = \pi/2$, when $\langle E^3\rangle = 0$ and $\langle A^3\rangle$ is maximal. The direction of the preferential photoelectron emission depends on the N parity and the values of E_1, E_2, and $p_\parallel$.

IV. MULTIPHOTON LIMIT

Let us consider the case when the transition points t_k are a large distance from the real axis, so that $\omega \mathrm{Im} t_k \gg 1$. The following expressions may be written in this case:

$$\sin\omega t_k \approx iz_k/2, \quad \sin(2\omega t_k + \varphi) \approx iz_k^2 e^{-i\varphi}/2, \quad (24)$$

where

$$z_k = \exp(-i\omega t_k), \quad |z_k| \gg 1. \tag{25}$$

According to Eq. (24), Eq. (5) with the vector potential (6) takes the form of the quadratic equation

$$z_k^2 + 2\frac{E_1}{E_2}e^{i\varphi}z_k - \sigma_1 y \frac{E_1^2}{E_2^2}e^{i(\varphi-\sigma_1\vartheta)} = 0, \quad \sigma_1 = \pm 1, \tag{26}$$

where

$$y = E_2E_0/E_1^2, \quad E_0 = 4\omega\sqrt{p^2+2K},$$
$$\vartheta = \arcsin(p_\parallel/\sqrt{p^2+2K}). \tag{27}$$

The solution of Eq. (26) has the form

$$z_k = -\frac{E_1}{E_2}e^{i\varphi}\left[1 - \sigma_2\sqrt{1+\sigma_1 y e^{-i(\varphi+\sigma_1\vartheta)}}\right], \quad \sigma_2 = \pm 1. \tag{28}$$

In the case of $y \ll 1$,

$$z_k = (\sigma_2 - 1)\frac{E_1}{E_2}e^{i\varphi} + \sigma_1\sigma_2\frac{yE_1}{2E_2}e^{-i\sigma_1\vartheta}.$$

Absolute values of the roots z_k corresponding to $\sigma_2 = -1$ far exceed those corresponding to $\sigma_2 = 1$. Therefore, the points t_k at $\sigma_2 = -1$ are more distant from the real axis and may be neglected in the evaluation of the transition probability. The absolute values of the roots taken into account may be written in the form

$$|z_k| = 2\omega\sqrt{p^2+2K}/E_1. \tag{29}$$

In the case of $y \gg 1$, all the roots

$$z_k = \sigma_2\frac{E_1}{E_2}\sqrt{\sigma_1 y e^{-i\sigma_1\vartheta}}e^{i\varphi/2},$$
$$|z_k| = (4\omega/E_2)^{1/2}(p^2+2K)^{1/4} \tag{30}$$

have been taken into account.

As seen from Eqs. (29) and (30), the validity conditions of the applied approximation may be written in the form

$$p^2/2 + K \approx N\omega \gg E_1^2/(8\omega^2) + E_2^2/(32\omega^2) = U_P/2. \tag{31}$$

When this condition is satisfied, from Eq. (3) one can obtain

$$S_k \approx N\omega t_k \tag{32}$$

and write the photoionization probability in the form

$$W_N(\mathbf{p}) = Cp\left|\sum_k z_k^{-N}\right|^2 = Cp(E_1/E_0)^{2N}f_N(y,\vartheta,\varphi),$$
$$f_N(y,\vartheta,\varphi) = \left|\sum_{\sigma_1=\pm1}\left[(1+\sqrt{1+\sigma_1 y e^{-i(\varphi+\sigma_1\vartheta)}})^N + (1-\sqrt{1+\sigma_1 y e^{-i(\varphi+\sigma_1\vartheta)}})^N\right] \times e^{iN(\varphi+\sigma_1\vartheta)}\right|^2. \tag{33}$$

The parameters $y, E_i/E_0, \vartheta$ and the condition (31) may be written using ordinary units

$$y = 10^7\sqrt{\frac{NI_2}{\lambda^3 I_1^2}},$$
$$E_i/E_0 \approx E_i/(\sqrt{32N\omega^3}) = 10^{-7}\sqrt{I_i\lambda^3/N}$$
$$\vartheta = p_\parallel/\sqrt{N} \tag{34}$$
$$= \arcsin\left(\sqrt{1 - \frac{0.8K\lambda + 0.75\times10^{-13}\left(I_1+\frac{I_2}{4}\right)\lambda^3}{N}} \times(\hat{\mathbf{p}}\cdot\hat{\mathbf{e}})\right), \quad I_1 + I_2/4 \ll 2.1\times10^{13}K/\lambda^2,$$

where I_1 and I_2 are the harmonic intensities in W/cm^2, λ is the fundamental harmonic wavelength in μm, and K is ionization potential of the atom in eV.

Equations (33) and (34) are rather simple and may be used to estimate the photoionization probability for the multiphoton limiting case. The plots W_N versus φ are shown in Fig. 3 at several values of y and ϑ. It is evident that the location of the photoionization probability

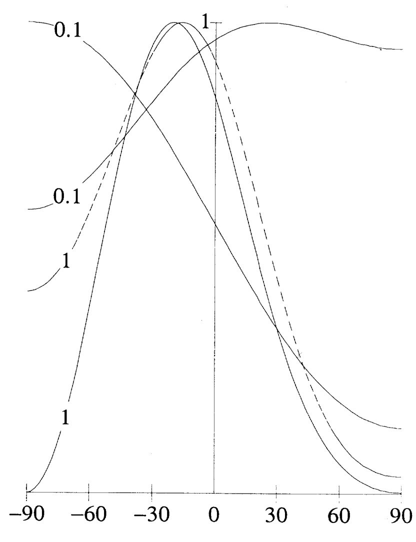

FIG. 3. Plots of photoionization probability W_{14} (in arbitrary units) versus phase harmonic shift for $\vartheta = \pi/8$ (solid line) and $\vartheta = \pi/12$ (dashed line). Numbers on the curves are values of the parameter y.

maximum varies with variation of y and ϑ.

For $\varphi = 0$ the inversion of the photoelectron emission direction leads to a change in sign of ϑ and to the complex conjugation of the sum terms in (33). Therefore, $f_N(y, -\vartheta, 0) = f_N(y, \vartheta, 0)$ and the photoelectron distribution has polar symmetry at $\varphi = 0$. This statement may be proved in terms of more general assumptions as well (see Sec. V).

Let us expand the sums in the Nth powers in Eq. (34) and represent f_N as the square of modulus of polynomial in y. This allows us to obtain the following representation for W_N:

$$W_N(\mathbf{p}) = 4Cp\left|\sum_{j=0}^{[N/2]} D_N^j e^{i(N-j)(\varphi+\pi/2)} \times \cos(N-j)(\vartheta - \pi/2)\left(\frac{E_1}{E_0}\right)^{N-2j}\left(\frac{E_2}{E_0}\right)^j\right|^2, \tag{35}$$

where

$$D_N^j = 2\sum_{i=j}^{[N/2]} C_N^{2i} C_i^j$$

and C_i^j are binomial coefficients. Equation (35) has the following physical meaning: the total transition amplitude is a sum of the transition amplitudes with absorption of $N - 2j$ quanta of frequency ω and j quanta of frequency 2ω. The φ dependence of the photoionization probability is a result of the interference of these transitions.

Simpler analytical expressions for W_N may be obtained for some limiting cases. Provided the condition

$$E_2 \ll E_1^2/(NE_0) \tag{36}$$

is satisfied, the polynomial in E_2 in Eq. (35) may be truncated to two terms and the following expression may be obtained:

$$W_N(\mathbf{p}) = 2Cp\left\{\left(\frac{2E_1}{E_0}\right)^{2N}[1 + (-1)^N \cos 2N\vartheta] - N\left(\frac{2E_1}{E_0}\right)^{2(N-1)}\frac{2E_2}{E_0}[\sin\vartheta - (-1)^N \times \sin(2N-1)\vartheta]\sin\varphi\right\}. \tag{37}$$

In the case of

$$E_1^2 \ll E_0 E_2/N \tag{38}$$

we could retain two terms with minimal E_2 powers in Eq. (35) and write W_N in the form

$$W_{2N_2}(\mathbf{p}) = 8Cp\left\{\left(\frac{E_2}{E_0}\right)^{2N_2}[1 + (-1)^{N_2}\cos 2N_2\vartheta] - 4N_2^2\left(\frac{E_2}{E_0}\right)^{2N_2-1}\frac{E_1^2}{E_0^2}[\sin\vartheta + (-1)^{N_2} \times \sin(2N_2+1)\vartheta]\sin\varphi\right\}, \tag{39}$$

$$W_{2N_2+1}(\mathbf{p}) = 8CpN^2\left\{\left(\frac{E_2}{E_0}\right)^{2N_2} \times \frac{E_1^2}{E_0^2}[1 - (-1)^{N_2}\cos(2N_2+1)\vartheta] - \frac{N_2^2 - 1}{3}\left(\frac{E_2}{E_0}\right)^{2N_2-1}\frac{E_1^4}{E_0^4}[\sin\vartheta - (-1)^{N_2} \times \sin(2N_2+3)\vartheta]\sin\varphi\right\}. \tag{40}$$

V. THE ELECTRICAL FIELD WITH MAXIMAL POLAR ASYMMETRY

As shown in the previous sections, the photoelectron angular distribution has polar symmetry when $\varphi = 0$ and polar asymmetry of the electrical field strength is at a maximum. To check this paradoxical result, a strict analysis of the photoionization by such a field is carried out in the present section. The approximations employed in previous sections for considering the tunneling and multiphoton limiting cases are not used now.

Hereafter let us consider such $A(t)$:

$$A(t^*) = A^*(t), \quad A(-t) = -A(t). \tag{41}$$

The first of these conditions expresses the function $A(t)$ analytically; the second one is valid for

$$A(t) = -c\sum_{n\geq 1}\frac{E_n}{n\omega}\sin n\omega t\ , \tag{42}$$

which leads to the time dependence of the electrical field given by the expression

$$E(t) = \sum_{n\geq 1} E_n \cos n\omega t. \tag{43}$$

Maxima of the electrical field strength for all harmonics coincide in time with each other for this dependence. Such a choice corresponds to the largest electrical field asymmetry. For example, in the case of two-color field (6) the asymmetry index $\langle E^3\rangle$ as a function of phase shift peaks at the phase shift, corresponding to the dependence (42).

Let us consider the photoionization probabilities for two photoelectron momentum values $\mathbf{p}$ and $\mathbf{p}' = -\mathbf{p}$. From Eqs. (5) and (41) it follows that the transition points t_k and t_k', corresponding to the above-mentioned momentum values, are related by the equation

$$t_k' = -t_k^*. \tag{44}$$

The integrals of analytical even $F_+(t)$ and odd $F_-(t)$ functions have the properties

$$\int_0^{-t^*} F_+(\zeta)d\zeta = -\left[\int_0^{t} F_+(\zeta)d\zeta\right]^*,$$
$$\int_0^{-t^*} F_-(\zeta)d\zeta = \left[\int_0^{t} F_-(\zeta)d\zeta\right]^*. \tag{45}$$

This may be proved by substituting the power series for $F_\pm(t)$ into the integrals. Hence

$$\begin{aligned} S'_k &= \int_0^{t'_k}\left[\frac{1}{2}\mathbf{p}'^2+\frac{1}{2c^2}\mathbf{A}^2(t)+K+\frac{1}{c}\mathbf{A}(t)\cdot\mathbf{p}'\right]dt \\ &= -\left\{\int_0^{t_k}\left[\frac{1}{2}\mathbf{p}^2+\frac{1}{2c^2}\mathbf{A}^2(t)+K\right]dt\right\}^* \\ &\quad +\mathbf{p}\cdot\left[\int_0^{t_k}\frac{1}{c}\mathbf{A}(t)dt\right]^* \\ &= -S_k^*. \end{aligned} \tag{46}$$

The expression (2) for the photoionization probability can be transformed to the form

$$\begin{aligned} W_N(\mathbf{p}) = Cp\Bigg\{&\sum_k \exp(-2\mathrm{Im}S_k) \\ &+2\sum_{k_1>k_2}\exp(-\mathrm{Im}S_{k_1}-\mathrm{Im}S_{k_2}) \\ &\times\cos(\mathrm{Re}S_{k_1}-\mathrm{Re}S_{k_2})\Bigg\}. \end{aligned} \tag{47}$$

From (46) we see that $\mathrm{Im}S_k$ is unchangeable and $\mathrm{Re}S_k$ changes its sign upon replacement of $\mathbf{p}$ by $-\mathbf{p}$. Thus, according to (47), $W_N(-\mathbf{p}) = W_N(\mathbf{p})$, which is what we set out to prove.

One could suppose that this result is a consequence of the inaccuracy of the adiabatic approximation. To demonstrate the invalidity of this supposition let us employ the Nikishov-Ritus-Faisal-Reiss approach [12, 14, 15] where the adiabatic approximation is not used. As shown in these works, the transition probability per unit time with $N\omega$ energy absorption is given by the expression

$$W_N(\mathbf{p}) = 2\pi(N\omega-U_P)^2|\tilde{\varphi}_0(\mathbf{p})|^2|a_N(\mathbf{p})|^2, \tag{48}$$

where $\tilde{\varphi}_0(\mathbf{p})$ is the wave function of the discrete state in the momentum representation

$$a_N(\mathbf{p}) = \frac{1}{2\pi}\int_0^{2\pi/\omega} dt\exp\{iN\omega t+if(\mathbf{p},t)\} \tag{49}$$

and

$$f(\mathbf{p},t) = \int_{-\infty}^{t} dt'\left[\frac{1}{c}\mathbf{A}(t')\mathbf{p}+\frac{1}{2c^2}\mathbf{A}^2(t')-U_P\right]. \tag{50}$$

Equation (2) may be derived from (49) by using the steepest-descent method for the calculation of the integral (see [10]).

A strict analysis may be carried out for the case of the odd time dependence of the vector potential

$$\mathbf{A}(-t) = -\mathbf{A}(t). \tag{51}$$

The above considered field (42), as well as the superposition of linearly polarized harmonics with arbitrary polarization direction, satisfies this condition.

Let us consider the change in $a(\mathbf{p})$ on the $\mathbf{p}$ inversion. Changing the integration variable in Eq. (50) from t' to $-t'$ and taking into account that the integrand in Eq. (50) has a zero time average, we obtain $f(-\mathbf{p},-t) = f(\mathbf{p},t)$. This leads to the relation

$$a_N(-\mathbf{p}) = a_N^*(\mathbf{p}). \tag{52}$$

The free-atom Hamiltonian has spherical symmetry and its wave function $\tilde{\varphi}_0(\mathbf{p})$ has a certain parity. As a result

$$|\tilde{\varphi}_0(-\mathbf{p})|^2 = |\tilde{\varphi}_0(\mathbf{p})|^2 \tag{53}$$

and, according to Eq. (48), again $W_N(-\mathbf{p}) = W_N(\mathbf{p})$. Thus the absence of the polar asymmetry of photoelectron angular distribution for ionization by the field with maximal asymmetry is now supported by the Nikishov-Ritus-Faisal-Reiss approach.

VI. DISCUSSION

The probability of atom multiphoton ionization by the bichromatic laser field consisting of the fundamental field and its second harmonic as a function of the harmonics phase shift has been obtained in the present paper. The results are applicable to the case of the strong field, when the perturbation theory is inapplicable. Only one experimental research of multiphoton ionization in the strong bichromatic field [1] is known to us; the subject of study in [2] and [3] was the two-photon processes in a rather weak field. It is evident from estimations that the expressions for the tunneling limit from Sec. III may be used under the conditions of experiment [1]. The period 2π in the φ dependence of the photoionization probability was observed in that experiment with approximately equal intensities of two colors. At a first glance our theory disagrees with that experimental result because it predicts the period π in the φ dependence of the photoionization probability at $E_1 \approx E_2$ [see Eq. (16)]. But one should take into account that the intensity used in that experiment greatly exceeds the saturation intensity and the contribution to the signal from the fringe areas of large volume, where $E_1 \ll E_2$, may exceed the contribution from the central area of small volume. Therefore, Eq. (23) rather than Eq. (16) should be used to interpret the experimental results [1]. Equation (23) predicts not only the period 2π, but also faster oscillations experimentally observed for some peaks of the energy photoelectron spectrum. To compare quantitatively the theoretical dependences with the experimental data the averaging over the spatially inhomogeneous laser focus should be performed, taking the saturation effects into account. Unfortunately, the lack of information on the focus size and shape gives no way for such a comparison.

It seems natural to assume that the polar asymmetry of the photoelectron angular distribution is the consequence of the polar asymmetry of the electrical field strength; that is, the polar asymmetry of photoionization probability and $\langle E^3 \rangle$ are at maxima for the same φ values and the photoionization probability has polar symmetry when $\langle E^3 \rangle = 0$. Nevertheless, as it is demonstrated in the present paper that the photoionization probability has polar symmetry when $\langle E^3 \rangle$ is at maximum and the polar asymmetry of the photoionization probability is at maximum when $\langle E^3 \rangle = 0$. Although this result seems paradoxical, it is in accordance with experimental data. The matter is that the absolute difference of the harmonic phase shift was not measured in the experiments [1, 3] and the assumption that the photoionization probability peaks at $\varphi = 0$ was used. The absolute difference of the harmonic phase shift was measured in the experiment [2]; the photoelectron signal plot versus φ, presented in this work, shows that the photoionization probability is at maximum for $\varphi \approx \pi/2$ and at minimum for $\varphi \approx 3\pi/2$ (this means that for $\varphi = 3\pi/2 - \pi = \pi/2$ the photoionization probability with the photoelectron emission in the direction opposite a detector is at minimum). The photoelectron signal is for $\varphi = 0$ and $\varphi = \pi$ have the same value if a systematic error is taken into account; i.e., for $\varphi = 0$ the asymmetry of the photoelectron angular distribution is small.

The polar symmetry of the photoelectron angular distribution for $\varphi = 0$ results not only from the analysis of the tunneling (Sec. III) and multiphoton (Sec. IV) limiting cases. A strict proof of such symmetry was deduced in Sec. V using two KFR theories: the Nikishov-Ritus-Faisal-Reiss approach and the adiabatic approximation. We believe that this result is the consequence of neglecting the photoelectron-ion core interaction in any KFR theory. This interaction was taken into account using the zero-radius potential approximation in the computation of the negative ion photodecay [4], where the perturbation theory for the interaction between the electron and bichromatic radiation was applied. It is evident from Ref. [4] that the photodecay probability becomes symmetric for some φ value that does not exceed 0.15 rad in the case of linearly polarized radiation. In the case of strong radiation the influence of the photoelectron-ion core interaction is suppressed [16]; thus one can expect that taking this interaction into account leaves the present paper's results unchanged.

It is interesting to note that there is a correlation between the polar symmetry of the photoionization probability and that of the vector potential. As noted above, polar asymmetry of the photoionization probability is the consequence of the quantum interference of transition amplitudes. The analogy with the well-known Aharonov-Bohm effect [21], conditioned by the quantum interference of potential-dependent amplitudes as well, can be drawn.

Quantum interference is not included when the photoionization probability is evaluated as the tunneling probability in Refs. [9] and [10]. That fact seems to be a cause of the discordance between our results and the predictions of photoionization probability asymmetry at $\varphi = 0$ in Refs. [9] and [10]. Note that this result, obtained in [9] and [10], disagrees with the experimental data [2] and perturbation theory calculations [4]. The photoionization probability calculation in the multiphoton limiting case [10] is also in agreement with the present paper's result rather than that of the tunneling approaches.

VII. CONCLUSIONS

The Landau-Dykhne adiabatic approximation is applied to the problem of atom ionization by the bichromatic radiation consisting of the fundamental field and its second harmonic. The finite expressions for the photoionization probability versus harmonic phase shift are obtained in the tunneling and multiphoton limiting cases. The case when one of harmonics is weak is considered as well. It is demonstrated that the photoelectron angular distribution has polar asymmetry in the case of symmetric electrical field strength, whereas it has polar symmetry in the case of the field with maximal polar asymmetry. This result is in agreement with the experimental data.

A strict proof of the polar symmetry of the photoionization probability in the case of the time-antisymmetric vector potential (corresponding to the electrical field strength with maximal asymmetry) is derived using the adiabatic approximation as well as the Nikishov-Ritus-Faisal-Reiss approach.

The results are applicable to the case when the photoelectron-ion core interaction may be neglected. Note that there are reasons to believe that this interaction has little influence over the dependence of the photoelectron angular distribution on the phase harmonic shift in the case of the strong laser field considered. This justifies the approximation we have done.

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