High-order harmonic generation in a bichromatic elliptically polarized laser field

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(Received 11 April 1996)

The quantum theory of high-order harmonic generation by a low-frequency laser field is generalized to the case of a bichromatic elliptically polarized laser field. The quasiclassical cutoff law is analyzed. Numerical results for a linearly polarized bichromatic laser field are presented and analyzed for different laser field frequencies, intensities, and relative phases. Harmonic intensity is many orders of magnitude higher in the bichromatic case than in the monochromatic one, but the cutoff is shifted toward the lower harmonics. The plateau height can be controlled by changing the relative phase of the fields. A qualitative agreement with the recent experiments is shown. [S1050-2947(96)09207-4]

PACS number(s): 42.50.Hz, 42.65.Ky

I. INTRODUCTION

High-order harmonic generation (HOHG) is one of the major topics of multiphoton physics [1,2]. During the interaction with a short-pulse intense laser atoms emit coherent radiation at frequencies that are multiples of the laser frequency ω. The emitted harmonics are characterized by a rapid drop at low orders, followed by a broad plateau where all the harmonics have the same strength and a sharp cutoff at frequency $k\omega \approx I_p + 3U_p$, where I_p is the atomic ionization potential and $U_p = A_0^2/4$ (in a.u.) is the electron ponderomotive energy in the laser field with the vector potential amplitude A_0. In the last few years the possibility of control of HOHG using elliptically polarized laser field or using two independent laser fields has attract special attention [3–11]. Recently, Lewenstein *et al.* [12] proposed a fully quantum theory of HOHG by a low-frequency laser field. Their results confirmed the two-step semiclassical picture of HOHG ([2], pp. 95–110 and [13]) and are valid in the tunneling regime (Keldysh parameter $\gamma = (I_p/2U_p)^{1/2} < 1$ [14]). The HOHG in the multiphoton regime ($\gamma > 1$) was considered by other authors (see, for example, [15] and references therein). Lewenstein *et al.* [12] presented results for a monochromatic linearly polarized laser field. These results can be generalized to the case of a bichromatic elliptically polarized laser field. This is the first aim of our paper. In Sec. II we present general theory. The generalized cutoff law and its classical limit are presented in Sec. III. Numerical results and comparison with experiments for a bichromatic linearly polarized laser field are given in Sec. IV and finally Sec. V contains the conclusions.

II. THEORY

A bichromatic elliptically polarized laser field is described by the electric field vector

$$\vec{E}(t) = \sum_{j=1,2} E_{0j}[\hat{\vec{e}}_x \cos\xi_j \cos(\omega_j t + \varphi_j) + \hat{\vec{e}}_y \sin\xi_j \sin(\omega_j t + \varphi_j)], \quad (1)$$

where ω_j, E_{0j}, and φ_j, $j=1,2$, are the laser field components' frequency, amplitude, and phase, respectively. $\hat{\vec{e}}_x$ and $\hat{\vec{e}}_y$ are the unit polarization vectors and the jth component polarization is characterized by the angle ξ_j (for $\xi_j = 0$ polarization is linear, while for $\xi_j = \pi/4$ it is circular; ellipticity can be defined as the ratio of the electric-field strength of the minor to the major axis of the ellipse, $R_j = E_{yj}/E_{xj} = \tan\xi_j$). The vector potential of the laser field is

$$\vec{A}(t) = -\int_{t_0}^{t} \vec{E}(t')dt' = -A_0 \sum_{j=1,2} a_j[\hat{\vec{e}}_x \cos\xi_j \sin(\omega_j t + \varphi_j) - \hat{\vec{e}}_y \sin\xi_j \cos(\omega_j t + \varphi_j)], \quad (2)$$

where we extract the vector potential amplitude A_0 and introduce relative amplitudes a_j ($A_{0j} = E_{0j}/\omega_j = A_0 a_j$, $j=1,2$). t_0 is the initial time for which $A_0 = 0$. Generalization of the results by Lewenstein *et al.* [12] to the laser field defined by Eqs. (1) and (2) is straightforward and we will not repeat details of derivation. It should be mentioned that their theory (and therefore our generalization of this theory) can be applied when there are no intermediate resonances and in the tunneling regime (Keldysh parameter $\gamma < 1$). They neglected the contribution to the evolution of the system of all bound states except the ground state. The generalization to the presence of other bound states and to the laser field with the time-dependent pulse envelope can also be done and we plan to consider these problems in the near future. We suppose that frequencies ω_1 and ω_2 are commensurate, i.e., $\omega_1 = p\omega$, $\omega_2 = q\omega$ with p,q integers. The harmonic strength D_k is defined as the Fourier component of the time-dependent dipole moment

$$D_k(\hat{\vec{n}}) = \int_0^T \frac{dt}{T} D_{\hat{n}}(t) e^{ik\omega t}, \quad T = 2\pi/\omega. \tag{3}$$

We use the index k instead of $2k+1$ (see [12]) because in the presence of a bichromatic laser field generation of both even and odd harmonic is possible. $D_{\hat{n}}(t)$ is the time-dependent dipole moment along an arbitrary direction $\hat{\vec{n}}$ (in a.u.; c.c. denotes a complex conjugate part)

$$D_{\hat{n}}(t) = i\int_0^{t-t_0} d\tau \int d^3\vec{p} F_{\hat{n}}(t,\tau;\vec{p}) e^{-iS(t,\tau;\vec{p})} + \text{c.c.}, \tag{4}$$

where $F_{\hat{n}}(t,\tau;\vec{p})$ denotes the product of dipole matrix elements

$$F_{\hat{n}}(t,\tau;\vec{p}\,) = \hat{\vec{n}} \cdot \vec{d}^*(\vec{p} - \vec{A}(t)) \vec{E}(t-\tau) \cdot \vec{d}(\vec{p} - \vec{A}(t-\tau)), \tag{5}$$

and $S(t,\tau;\vec{p})$ is the quasiclassical action

$$S(t,\tau;\vec{p}) = \int_{t-\tau}^{t} dt' \{ \tfrac{1}{2}[\vec{p} - \vec{A}(t')]^2 + I_p \}. \tag{6}$$

In Eq. (4) the integration is over the return time τ. $\vec{d}(\vec{p}) = \langle \vec{p} | \vec{x} | 0 \rangle$ denotes the atomic dipole matrix element for the bound-free transition. The explicit form of $\vec{d}(\vec{p})$ for the Gaussian model and for hydrogenlike atoms is given in [12]. In the general case the integral over $\vec{p}$ can be solved using the saddle-point method. If the exponent of the subintegral function (in our case it is the action S) is proportional to a large parameter x, then we search for a stationary point of S and obtain a power series in $1/x$. In [12] it was supposed that the ponderomotive potential U_p, ionization potential I_p, and velocities are large. In that paper there was no explicit expansion parameter and no correction to the zeroth-order term is obtained. We will present this here. If one does not introduce the atomic system of units, then, instead of S, one obtains $S/\hbar$, where $\hbar$ is the Planck constant devided by 2π. If we suppose that $\hbar$ is small (this is a variant of the WBK method) and use $1/\hbar$ as a large parameter, then the solution for the triple integral $\int d^3\vec{p} = \int_{-\infty}^{\infty} dp_x \int_{-\infty}^{\infty} dp_y \int_{-\infty}^{\infty} dp_z$ takes the form,

$$e^{-iS(t,\tau;\vec{p})/\hbar} \sum_{n_x,n_y,n_z=0}^{\infty} \frac{e^{-i(n_x+n_y+n_z)\pi/2}}{(2\tau/\hbar)^{n_x+n_y+n_z} n_x! n_y! n_z!} F_{\hat{n}}^{(2n_x,2n_y,2n_z)}(t,\tau;\vec{p})_{\vec{p}=\vec{p}_s}, \tag{7}$$

where index $(2n_x,2n_y,2n_z)$ denotes partial derivatives over p_x, p_y, and p_z, respectively. The zeroth-order term and the first-order corrections are

$$e^{-iS(t,\tau;\vec{p})/\hbar} \left(1 - i\frac{\hbar}{2\tau} \frac{\partial^2}{\partial \vec{p}^2} \right) F_{\hat{n}}(t,\tau;\vec{p})_{\vec{p}=\vec{p}_s}. \tag{8}$$

The $\vec{p}_s$ is the solution of the equation $\nabla_{\vec{p}} S(t,\tau;\vec{p}) = 0$, which is

$$\vec{p}_s(t,\tau) = \frac{1}{\tau} \int_{t-\tau}^{t} dt' \vec{A}(t'). \tag{9}$$

In this case the stationary action is (in a.u.)

$$S_s(t,\tau) \equiv S(t,\tau;\vec{p}_s) = I_p \tau - \frac{\tau}{2} \vec{p}_s^{\,2}(t,\tau) + \frac{1}{2} \int_{t-\tau}^{t} dt' \vec{A}^2(t'), \tag{10}$$

and $D_{\hat{n}}(t)$ is given by ($\epsilon \to 0^+$, $t_0 \to -\infty$)

$$D_{\hat{n}}(t) = i\int_0^{\infty} d\tau \left(\frac{\pi}{\epsilon + i\tau/2} \right)^{3/2} e^{-iS_s(t,\tau)} \left(1 - \frac{i}{2\tau} \frac{\partial^2}{\partial \vec{p}^2} \right) F_{\hat{n}}(t,\tau;\vec{p})_{\vec{p}=\vec{p}_s} + \text{c.c.} \tag{11}$$

Let us introduce the following notation:

$$\xi_\pm = \xi_1 \pm \xi_2, \quad \omega_\pm = \omega_1 \pm \omega_2, \quad \varphi_\pm = \varphi_1 \pm \varphi_2,$$

$$\psi_n = \omega_n \left(t - \frac{\tau}{2} \right) + \varphi_n, \quad n = 1,2,\pm. \tag{12}$$

After some elementary calculations we obtain the following expression for the quasiclassical action:

$$S_s(t,\tau)=S_0(\tau)+\sum_{j=1,2}S_j(\tau)\cos 2\psi_j(t,\tau)+\sum_{n=\pm}S_n(\tau)\cos\psi_n(t,\tau), \tag{13}$$

where

$$S_0(\tau)=I_p\tau+U_p\sum_{j=1,2}\frac{a_j^2}{\omega_j}\left[\omega_j\tau-\frac{2(1-\cos\omega_j\tau)}{\omega_j\tau}\right],$$

$$S_j(\tau)=U_p\cos 2\xi_j\frac{a_j^2}{\omega_j}\left[\frac{2(1-\cos\omega_j\tau)}{\omega_j\tau}-\sin\omega_j\tau\right],\quad j=1,2,$$

$$S_n(\tau)=-4nU_pa_1a_2\cos\xi_n\left(\frac{1}{\omega_n}\sin\frac{\omega_n\tau}{2}+\frac{1}{\omega_1\omega_2\tau}\sum_{n'=\pm}n'\cos\frac{\omega_{n'}\tau}{2}\right),\quad n=\pm. \tag{14}$$

In the case of a linearly polarized monochromatic laser field the interference terms $S_\pm$ disappear and the result of paper [12] can be easily reproduced. In the general case the final result for the harmonic strength is

$$D_k(\hat{\vec{n}})=I_k(\hat{\vec{n}})+I^*_{-k}(\hat{\vec{n}}), \tag{15}$$

where

$$I_k(\hat{\vec{n}})=i\int_0^\infty d\tau\left(\frac{\pi}{\epsilon+i\tau/2}\right)^{3/2}e^{-iS_0(\tau)}\int_0^T\frac{dt}{T}\left(1-\frac{i}{2\tau}\frac{\partial^2}{\partial\vec{p}^2}\right)F_{\hat{n}}(t,\tau;\vec{p})_{\vec{p}=\vec{p}_s}$$
$$\times\exp\left\{i\left[k\omega t-\sum_{j=1,2}S_j(\tau)\cos 2\psi_j(t,\tau)-\sum_{n=\pm}S_n(\tau)\cos\psi_n(t,\tau)\right]\right\}. \tag{16}$$

According to Eq. (5) terms $\vec{p}_s-\vec{A}(t)$ and $\vec{p}_s-\vec{A}(t-\tau)$ appear in $F_{\hat{n}}(t,\tau;\vec{p}_s)$. They can be written as

$$\vec{\pi}(t')\equiv\vec{p}_s(t,\tau)-\vec{A}(t')=A_0\sum_j a_j\{\hat{\vec{e}}_x\cos\xi_j[\sin(\omega_jt'+\varphi_j)-b_j(\tau)\sin\psi_j(t,\tau)]+\hat{\vec{e}}_y\sin\xi_j[-\cos(\omega_jt'+\varphi_j)+b_j(\tau)\cos\psi_j(t,\tau)]\}, \tag{17}$$

with $b_j(\tau)=(2/\omega_j\tau)\sin(\omega_j\tau/2)$. The explicit form of $F_{\hat{n}}(t,\tau;\vec{p}_s)$ can be obtained using the expression for $\vec{d}(\vec{p})$ from [12], but we will not present it here. The conclusion is that the corrections to the result for $D_k(\hat{\vec{n}})$ depend on the chosen model. We computed numerically $D_k(\hat{\vec{n}})$ for the Gaussian model and for the hydrogenlike atoms. In the Gaussian case the corrections to the zeroth-order term are really small. Furthermore, for the Gaussian model we computed the integral over $\vec{p}$ analytically and the comparison of these results with the saddle-point method results also shows a very good agreement. In the case of hydrogenlike atoms our results show that the corrections cannot be neglected (the difference is about 30%). Nevertheless, the qualitative behavior of the harmonic strength as a function of the harmonic order is completely determined by the quasiclassical action S_s, which does not depend on the chosen model.

Each of the terms with S_1, S_2, S_+, and S_- in Eq. (16) can be expanded in a series of the Bessel functions and, after introducing the Fourier components of the product of dipole moments,

$$\sum_{m=-\infty}^{\infty}B_m(\tau)e^{-imt}=\left(1-\frac{i}{2\tau}\frac{\partial^2}{\partial\vec{p}^2}\right)F_{\hat{n}}(t,\tau;\vec{p})_{\vec{p}=\vec{p}_s}, \tag{18}$$

the integral over t can be computed analytically. The final expression is cumbersome. It contains a fourfold sum with a product of the four Bessel functions (for a similar approach see [11], where a zero-range potential model was considered). It is simpler to compute the double integral in Eq. (16) numerically. In the special case of a monochromatic elliptically polarized laser field, similarly as in [12], one obtains

$$I_k(\hat{\vec{n}})_{\omega_1=\omega_2=1}=i\sum_{n=-\infty}^{\infty}\int_0^\infty d\tau\left(\frac{\pi}{\epsilon+i\tau/2}\right)^{3/2}e^{i[n\tau-S_0(\tau)]}i^nJ_n(U_p\cos 2\xi C(\tau))B_{k-2n}(\tau), \tag{19}$$

where now

$$S_0(\tau)=(I_p+U_p)\tau-2U_p(1-\cos\tau)/\tau,$$

$$C(\tau)=\sin\tau-2(1-\cos\tau)/\tau. \tag{20}$$

It is important that the argument of the Bessel function J_n depends on the polarization through a simple factor $\cos 2\xi$. For the linear polarization it is $\cos 2\xi=1$ and the only difference between the zeroth-order term of our result (15) with (18)–(20) and the result (18) from [12] is that we have the term I^*_{-k} [which originates from the c.c. part in Eq. (11)] and that the sum over n goes from $-\infty$ to $+\infty$, while the result of Ref. [12] contains only the non-negative n. For $U_p\gg 1$ and $n<0$ the factor $\exp\{i[n\tau-S_0(\tau)]\}$ introduces a fast oscillation in the integral over τ and the contribution to the result (19) from the terms with $n<0$ is small. For $n\geqslant 0$ and $k<0$ the Fourier components $B_{k-2n}(\tau)=B_{-|k|-2n}(\tau)$ are small and therefore the contribution I_{-k} to the final result is small. The conclusion is that for a high-intensity laser field ($U_p\gg 1$) and for high-order harmonics ($k\gg 1$) the result of Lewenstein *et al.* (Eq. (20) in [12]) is recovered. Our exact numerical computations [using the double integral, Eqs. (15) and (16)] confirm this conclusion. We compute the integral over τ from zero to infinity as a Padé series of integrals from $2\pi m$ to $2\pi(m+1)$, $m=0,1,2,\ldots$. The series shows fast convergence and the main contribution to the integral comes from the first return of the ionized electron to the nucleus ($m=0$). For an elliptically polarized laser field with a high ellipticity (close to circular) one has $\cos 2\xi=0$, $J_n(0)=\delta_{n,0}$, and the sum over n in Eq. (19) is canceled. The Fourier components' B_k are small for high-order harmonics and function $\exp[-iS_0(\tau)]$ has fast oscillations. Therefore, for such an elliptically polarized laser field the HOHG is suppressed, which is in agreement with the experiments [3,8]. In the circular polarization case it can be explicitly shown that $B_k(\tau)=0$, $k\neq\pm 1$, and therefore no harmonics are emitted, which is in accordance with the angular momentum conservation.

The case of an elliptically polarized laser field was considered recently [16]. The result similar to (19) was obtained in the case of hydrogenlike atoms and transition from the s state. The argument of the Bessel function contains the same factor $U_p\cos 2\xi=U_p(1-\varepsilon^2)/(1+\varepsilon^2)$ (parameter $\varepsilon=\tan\xi$ defines polarization in Ref. [16]). In Appendix A of [16] an analytic form of the Fourier coefficients for the hydrogenlike atoms is presented. In the circular polarization case these coefficients are equal to zero and no harmonic is emitted. Numerical results presented in [16] confirm the conclusions of our analysis: (i) a strong suppression of the harmonic strength with the increase of the ellipticity and (ii) an increase of this suppression with the increase of the harmonic order. In [16] the propagation effects are also taken into account. This is not considered in our paper, but we expect that the main influence of the propagation will be in the smoothing of the quantum interference.

III. CUTOFF LAW

The cutoff law can be obtained by applying the saddle-point method for computing the remaining integrals in (16),

$$D_k\sim\int d\tau\int dt\exp\{i[k\omega t-S_s(t,\tau)]\}\times F_{\hat{n}}(t,\tau)(\epsilon+i\tau/2)^{-3/2}. \tag{21}$$

One obtains $\partial S_s(t,\tau)/\partial\tau=0$ and $\partial S_s(t,\tau)/\partial t-k\omega=0$, which, according to Eqs. (10) and (17), gives

$$\vec{\pi}^2(t)=-2I_p,\quad \vec{\pi}^2(t)-\vec{\pi}^2(t-\tau)=2k\omega. \tag{22}$$

An expanded form of (22) is

$$\left[\sum_{j=1,2}a_j\cos\xi_j(c_j\sin\psi_j-s_j\cos\psi_j)\right]^2+\left[\sum_{j=1,2}a_j\sin\xi_j(c_j\cos\psi_j+s_j\sin\psi_j)\right]^2=-I_p/(2U_p), \tag{23}$$

$$\frac{k\omega}{8U_p}=\sum_{i=1,2}a_i\cos\xi_i s_i\cos\psi_i\sum_{j=1,2}a_j\cos\xi_j c_j\sin\psi_j-\sum_{i=1,2}a_i\sin\xi_i s_i\sin\psi_i\sum_{j=1,2}a_j\sin\xi_j c_j\cos\psi_j, \tag{24}$$

where

$$c_j(\tau)=\cos\frac{\omega_j\tau}{2}-b_j(\tau),\quad s_j(\tau)=\sin\frac{\omega_j\tau}{2}. \tag{25}$$

Equations (23) and (24) depend on t through the functions $\sin\psi_j(t,\tau)$ and $\cos\psi_j(t,\tau)$ [see Eq. (12)], which also should satisfy the condition $\sin^2\psi_j+\cos^2\psi_j=1$. The right-hand side of Eq. (23) is negative, so that the solutions for t and τ are complex. In the general case the cutoff law can be obtained by finding the maximum of the function $\mathrm{Re}(k\omega/8U_p)$ under the constraint $\mathrm{Im}(k\omega/8U_p)=0$ (see [12] for a monochromatic linearly polarized laser field). This is not an easy task. For our purpose we consider only a special case of a linearly polarized bichromatic laser field. In the limit $I_p\to 0$, Eq. (23) gives

$$\sum_{j=1,2}a_j(c_j\sin\psi_j-s_j\cos\psi_j)=0, \tag{26}$$

so that

$$k\omega=8U_p\left(\sum_{j=1,2}a_j s_j\cos\psi_j\right)^2. \tag{27}$$

In this case one should find the real t and τ for which $k\omega$ is maximal. We suppose that at some time $t=t_t$ (i.e., $\tau_t=t-t_t=0$) the electron tunnels into the continuum and moves in the presence of the laser field only. The electron is most likely to emit the harmonic photon when it is back at the atomic binding potential origin. This happens at some time $t=t_e$, i.e., after the return time τ_e. The electron can tunnel at different instants $t_t=t-\tau$. In order to find for which t_t $k\omega$ has maximum, we start from the condition

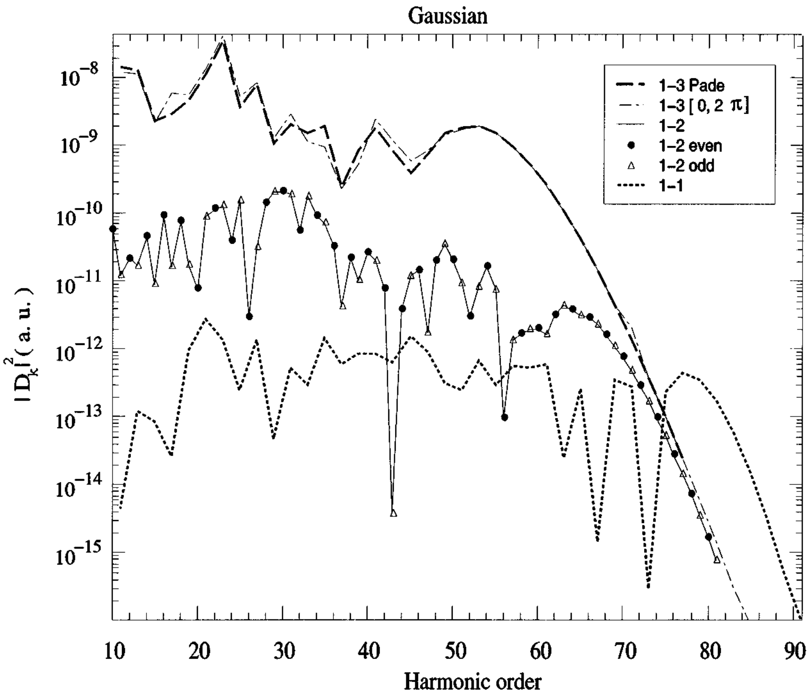

FIG. 1. Harmonic intensity as a function of the harmonic order for the Gaussian model and for $\omega_1=\omega_2$ (short-dashed line: curve 1-1), $\omega_2=2\omega_1$ (solid line: curve 1-2, even harmonics: filled circles, odd harmonics: open triangles) and $\omega_2=3\omega_1$ [curve 1-3: long-dashed line, result obtained using Padé method (see text); dot-dashed line, result obtained by integrating over τ from zero to 2π]. The other parameters are $\varphi_1=\varphi_2=0$, $a_1=a_2=0.5$, $U_p=20\omega_1$, $I_p=13.6\omega_1$ ($\omega_1=1$ eV).

$\partial k\omega(t_e,t_t)/\partial t_t=0$. In order to apply this condition one has to express $k\omega$, Eq. (27), through the variable $t_t=t-\tau$. The result is

$$k\omega(t_e,t_t)=2U_p\left\{\sum_{j=1,2}a_j[\sin(\omega_j t_e+\varphi_j)-\sin(\omega_j t_t+\varphi_j)]\right\}^2. \tag{28}$$

Similarly, the condition (26) can be expressed as

$$\sum_{j=1,2}a_j[\cos(\omega_j t_e+\varphi_j)-\cos(\omega_j t_t+\varphi_j)+\omega_j(t_e-t_t)\sin(\omega_j t_t+\varphi_j)]/\omega_j=0. \tag{29}$$

Differentiating this equation with respect to t_t, we find the condition for $\partial t_e/\partial t_t$, which we will use to compute $\partial(k\omega)/\partial t_t=0$. The last equation gives the following condition (written again through our variables ψ and τ):

$$\sum_{j=1,2}a_j\omega_j(s_j\sin\psi_j-c_j\cos\psi_j)=0. \tag{30}$$

Equations (26) and (30) represent a system of two equations for two real variables t and τ, which can be easily solved. Introducing these solutions into Eq. (27) one obtains the looked-for $(k\omega)_{\max}$. We will use this method in the next section in order to predict the cutoff position. From Eqs. (26) and (30) it follows that the cutoff position depends on the laser field components' intensities (a_1,a_2), frequencies (ω_1,ω_2), and phases (φ_1,φ_2).

It should be mentioned that Eqs. (26)–(30) can be obtained using the classical model developed by Kulander and co-workers [18] (see also [19]). In this case we start from the classical equation of motion $m\ddot{x}(t)=eE(t)$ and the model assumption that at $t=t_t$ the electron is set free in the continuum with the velocity zero $[\dot{x}(t_t)=0]$. We obtain an equation equivalent to (26). The harmonic photon energy $k\omega$ corresponds to the kinetic energy E_k and the condition (30) can be obtained from the condition $\partial E_k/\partial t_t=0$. As a conclusion we can say that the result for the cutoff law obtained using the classical model is equivalent to the semiclassical result in the limit when the atomic ionization potential tends to zero.

IV. NUMERICAL RESULTS

Harmonic strength depends on many parameters: frequencies ω_1 and ω_2, vector potential amplitude A_0 (i.e., the ponderomotive potential $U_p=A_0^2/4$), relative amplitudes a_1 and a_2, phases φ_1 and φ_2 (usually it is enough to analyze the dependence on a_1/a_2 and relative phase $\varphi=\varphi_1-\varphi_2$), polarization parameters ξ_1 and ξ_2, direction $\hat{\vec{n}}$ on which one projects dipole moment (parameters $n_x=\hat{\vec{e}}_x\cdot\hat{\vec{n}}$ and $n_y=\hat{\vec{e}}_y\cdot\hat{\vec{n}}$) and ionization potential I_p (or other parameters that characterize the model chosen to describe the atom). Experiments [3,8] and theory (see factor $\cos 2\xi$ in Eq. (19) and [16]) showed that the harmonic strength is the highest for a linearly polarized laser field. Taking this into account and in order to decrease the number of parameters we analyze numerically only the case of a linearly polarized bichromatic laser field. We compute $D_k(\hat{\vec{e}}_x)$ by applying formulas (15)–(17), which are now simplified $(\xi_j=0)$. We first analyze the results presented in Fig. 1. The harmonic intensity as a function of the harmonic order is shown for three cases: $\omega_1=\omega_2=\omega$ (short-dashed line: curve 1-1), $\omega_1=\omega$, $\omega_2=2\omega$ (solid line: curve 1-2), and $\omega_1=\omega$, $\omega_2=3\omega$ (long-dashed line: curve 1-3). In all three cases the phases are zero $(\varphi_1=\varphi_2=0)$, relative amplitudes are $a_1=a_2=0.5$, $U_p=20\omega$, $I_p=13.6\omega$, and a Gaussian model [12] is chosen. The result for $\omega_1=\omega_2$ is in an agreement with the previous result [12], except for the low-order harmonics [see comments below Eq. (20)]. For $\omega_2=2\omega_1$ we observe even and

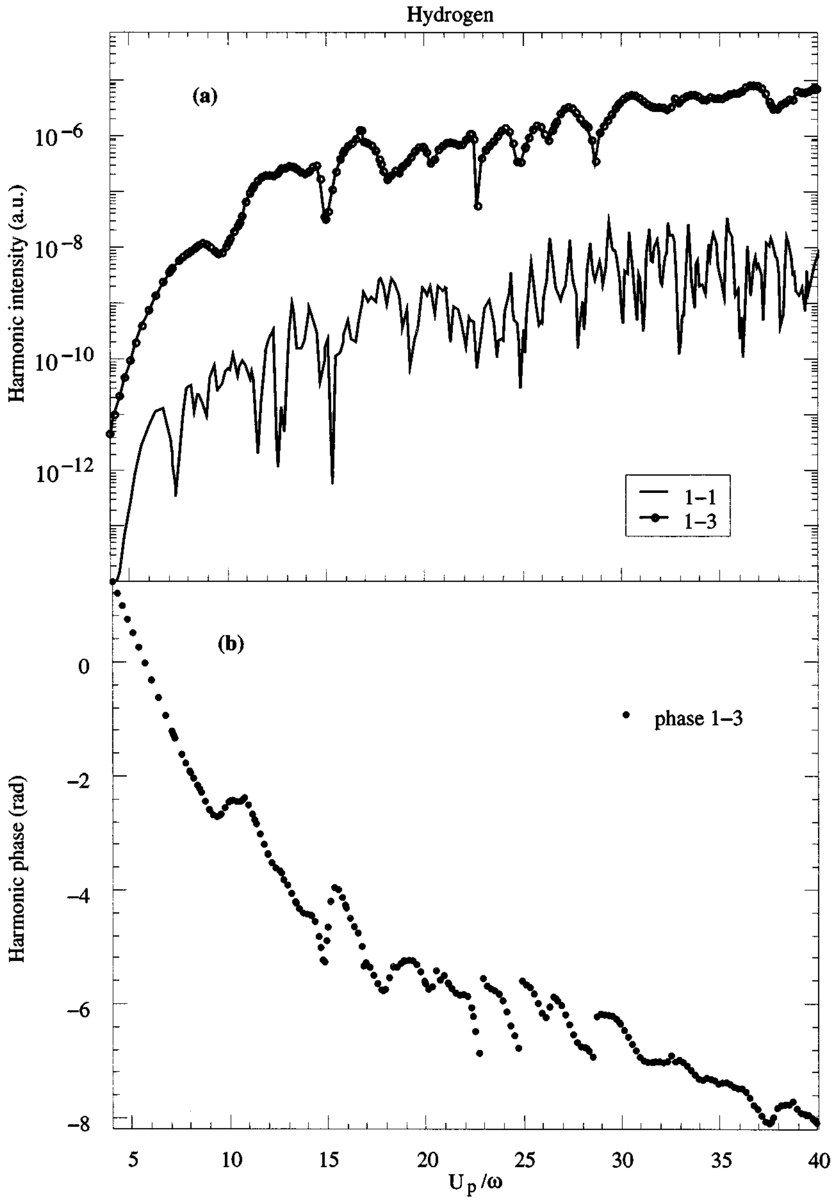

FIG. 2. Harmonic intensity (a) (curves 1–3) and phase (b) as functions of the ponderomotive potential U_p for the $\omega_2 = 3\omega_1$ case. Harmonic intensity for the monochromatic case [lower curve: 1-1 at (a)] is also shown. The hydrogenlike atom model is used and $\varphi_1 = \varphi_2 = 0$, $a_1 = a_2 = 0.5$, $I_p = 13.6\omega_1$ ($\omega_1 = 1$ eV).

odd harmonics as one can expect from the symmetry breaking [2,5]. The intensities of the even harmonics (filled circles) are of the same magnitude as the intensity of the odd harmonics (open triangles) and they follow the same curve. The harmonic intensity in the plateau region is about two orders of magnitude greater in the $\omega_2 = 2\omega_1$ than in the $\omega_1 = \omega_2$ case. The cutoff is shifted towards the lower harmonics. This is expected because, as the solution of Eqs. (26) and (30), we obtained $\max(k\omega)_\omega = 3.17U_p + I_p = 77\omega$ and $\max(k\omega)_{\omega-2\omega} = 2.52U_p + I_p = 64\omega$. In the $\omega_2 = 3\omega_1$ case the symmetry is conserved and we have only odd harmonics. The harmonic intensity is four orders of magnitude greater than in the $\omega_1 = \omega_2$ case and the position of the cutoff is shifted toward the lower harmonics [$\max(k\omega)_{\omega-3\omega} = 2.24U_p + I_p = 58.4\omega \rightarrow k = 57$]. We compared the exact result (integration over τ from zero to infinity is done using the Padé method, long-dashed line) and the result obtained by integrating over the first return ($\tau \in [0,2\pi]$, dot-dashed line). The difference between these two results is small, which shows that the effect of the electron rescattering (see Ref. [12]) is small in this case.

As the next result we chose to present the dependence of the harmonic intensity and phase (for a fixed harmonic) on the ponderomotive potential U_p. These results are presented in Fig. 2 and correspond to hydrogenlike atoms with $I_p = 13.6\omega$, $\omega_2 = 3\omega_1$, $\varphi_1 = \varphi_2 = 0$, $a_1 = a_2 = 0.5$, and $k = 35$. The upper harmonic intensity curve at Fig. 2(a) corresponds to the $\omega_2 = 3\omega_1$ case, while the lower one corresponds to the $\omega_1 = \omega_2$ case. With the increase of U_p the harmonic intensity first rapidly increases. After this cutoff region there is a plateau region in which the harmonic intensity saturates and is dominated by the quantum interference effect. The interference oscillations are more rapid in the $\omega_1 = \omega_2$ case. Figure 2(b) shows the dependence of the 35th harmonic phase on U_p for the $\omega_2 = 3\omega_1$ case. In the cutoff region the phase decreases linearly with the increase of U_p. The slope of the curve is much smaller than in the monochromatic case where it is ≈ -3.2 [17]. The explanation of this slope in the monochromatic case is connected with the approximate value of the action acquired along the most relevant saddle-point trajectory, that is,

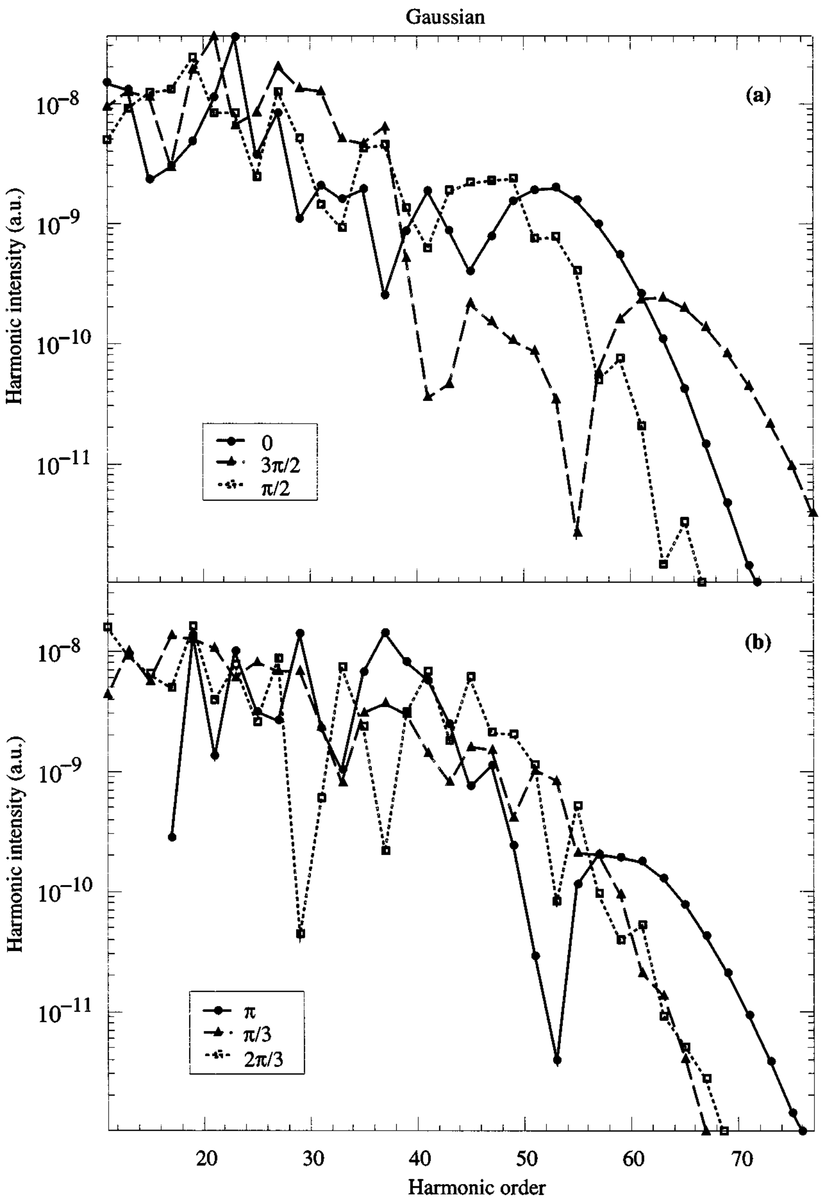

FIG. 3. Harmonic intensity as a function of the harmonic order for the Gaussian model, $\omega_2=3\omega_1$, $a_1=a_2=0.5$, $U_p=20\omega_1$, $I_p=13.6\omega_1$, $\omega_1=1$ eV, and for the different values of the relative phase $\varphi=\varphi_1-\varphi_2$: (a) $\varphi=\{0,3\pi/2,\pi/2\}$ and (b) $\varphi=\{\pi,\pi/3,2\pi/3\}$.

$S_s\approx U_p\tau_s+\cdots$, so that the harmonic phase $\phi=-S_s \approx -4.086U_p/\omega$ (see [17] and our Eq. (20) with $\omega\tau_s=4.086$). An even better estimation of the harmonic phase can be obtained if one takes into account the complete monochromatic stationary action

$$S_s\approx U_p\tau_s-\frac{2U_p}{\omega}\frac{1-\cos\omega\tau_s}{\omega\tau_s}$$

$$+\frac{U_p}{\omega}\left[\frac{2(1-\cos\omega\tau_s)}{\omega\tau_s}-\sin\omega\tau_s\right]\cos\omega(2t_s-\tau_s)$$

$$\approx 3.31U_p/\omega,$$

where $\cos\omega(2t_s-\tau_s)=0$. In the bichromatic $\omega_2=3\omega_1$ case, according to Eqs. (13) and (14), it is

$$S_s(t_s,\tau_s)\approx S_0(\tau_s)+\sum_{j=1,2}S_j(\tau_s)\cos 2\psi_j(t_s,\tau_s)$$

$$+\sum_{n=\pm}S_n(\tau_s)\cos\psi_n(t_s,\tau_s)$$

$$\approx 1.226U_p/\omega,$$

where we used $\omega\tau_s=3.540$ and $\omega(2t_s-\tau_s)=3.8466$ (obtained by solving the cutoff law equations). The line $\phi=-1.226U_p/\omega$ is in poorer agreement with our numerical results ($\phi_{\rm num}\approx-0.84U_p/\omega$) than it was in the monochromatic case. This indicates that the method of the harmonic phase estimation from [17] is less accurate in the bichromatic $\omega_2=3\omega_1$ case. In the plateau region, at each interference oscillation of the harmonic intensity, the phase also has oscillation. Moreover, the phase changes more rapidly and it is difficult to determine the slope.

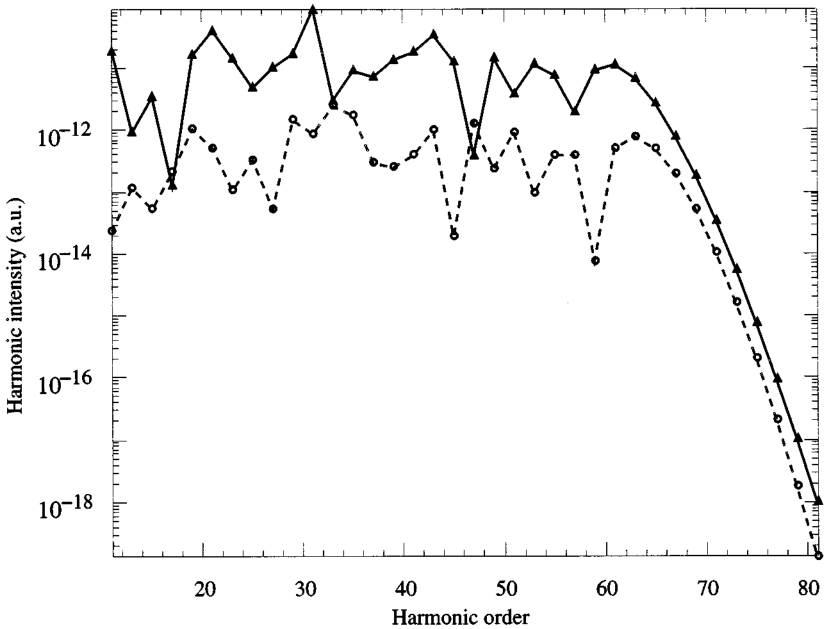

FIG. 4. Harmonic intensity as a function of the harmonic order for monochromatic (ω=1.664 eV, dashed line, open circles) and bichromatic $\omega-3\omega$ (full line, filled triangles) laser fields. Other parameters correspond to the experiment of Kondo *et al.* (see text).

The next example that we present concerns the dependence of the harmonic intensity on the relative phase $\varphi=\varphi_1-\varphi_2$ for the $\omega_2=3\omega_1$ case. For a fixed harmonic order the harmonic intensity changes rapidly with the change of the relative phase φ over the interval $[0,2\pi]$. We will not present these results, but it should be mentioned that the general conclusion of our analysis is that the behavior of the harmonic intensity with the change of the relative phase does not depend on the chosen atomic model (hydrogenlike or Gaussian). This behavior is completely determined by the quasiclassical action S_s. In Fig. 3 we present the dependence of the harmonic intensity on the harmonic order for the fixed relative phases: $\varphi=\{0,3\pi/2,\pi/2\}$ [Fig. 3(a)] and $\varphi=\{\pi,\pi/3,2\pi/3\}$ [Fig. 3(b)]. From these figures one can see that both the height of the plateau and the position of the cutoff change with the change of φ. The cutoff position is slightly shifted toward the higher harmonics for $\varphi=3\pi/2$ and $\varphi=\pi$ in comparison with the $\varphi=0$ case, while for $\varphi=\pi/3$, $\pi/2$, and $2\pi/3$ the shift is toward the lower harmonics. This is in agreement with the solution of Eqs. (26) and (30), which gives the following factors in the cutoff law equation:

$$(k\omega)_{\max}=cU_p+I_p:$$

$$c=\{2.24,2.716,2.07,2.55,1.86,2.23\}$$

for $\varphi=\{0,3\pi/2,\pi/2,\pi,\pi/3,2\pi/3\}$. From Fig. 3 it follows that by changing the relative phase it is possible to control the height of the plateau. For example, the intensity of the harmonics of order 21–39 is the highest for $\varphi=3\pi/2$ and $\varphi=\pi$, while for $k\approx 41-51$ it is the highest for $\varphi=\pi/2$ and $\varphi=2\pi/3$.

In order to check the validity of our theory we compute the harmonic spectrum for recent experiments with the bichromatic laser field [5,7]. The comparison can be only qualitative because we are not able to include the propagation effects [16,17]. In the first experiment Kondo *et al.* [5] used a Ti:sapphire laser ($\lambda=745$ nm, $\omega=1.664$ eV) and its third harmonics. The laser field intensity was $I_\omega=5\times10^{14}$ W/cm^2 (so that $U_p=15.59\omega$), the ratio of the electric-field strengths was $E_{3\omega}/E_\omega=0.3$ (and therefore our parameters $a_{3\omega}=0.1a_\omega$), and Ne atoms were used ($I_p=21.6$ eV$=12.98\omega$, so that the Keldysh parameter $\gamma=0.645<1$). The cutoff law for the laser field with only one frequency ω and $a_1=a_2=0.5$ is $\max(k\omega)_\omega=I_p+3.17U_p=62.4\omega$, while the bichromatic cutoff law for the mentioned experimental parameters is $\max(k\omega)_{\omega-3\omega}=I_p+3.102U_p=61.34\omega$. The harmonic spectrum (harmonic intensity as a function of the harmonic order) for these two cases is shown in Fig. 4. One can observe that (i) the cutoff for both spectra is approximately at the same position $\approx 63\omega$ and (ii) the height of the plateau in the $\omega-3\omega$ case is one order of magnitude higher. Such an enhancement of the intensity is really observed in the experiment. Besides, the position of the cutoff in the experiment is the same for ω and $\omega-3\omega$ cases, but the cutoff is relatively low ($\approx I_p+1.8U_p$; as explained in [5], this is probably due to the tight focusing).

In the experiment by Eichmann *et al.* [7], a high-power Ti:sapphire laser (ω=1.6 eV, $I_\omega=1.33\times10^{14}$ W/cm^2) and its second harmonic ($I_{2\omega}=0.58\times10^{14}$ W/cm^2, $a_2=0.33a_1$) were used. The ionization potential of Ar is $I_p=15.76$ eV$=9.85\omega$ ($U_{p_\omega}=4.664\omega$, $\gamma\approx1$). For the cutoff laws we obtained $\max(k\omega)_\omega=I_p+3.17U_{p_\omega}=24.63\omega$, $\max(k\omega)_{\omega-2\omega}=I_p+4.508U_{p_\omega}=30.88\omega$. Numerical results presented in Fig. 5 are in agreement with these positions. The plateau height for the $\omega-2\omega$ case is almost two orders of magnitude higher and the intensities of the even and the odd harmonics are of the same order. In [7] the experimental results were compared with the numerical calculations using a zero-range potential model (see also [11]), but without taking into account the propagation of the harmonics through the medium. We presented our results only as an illustration, with no intention for quantitative comparison with this theory and experiment.

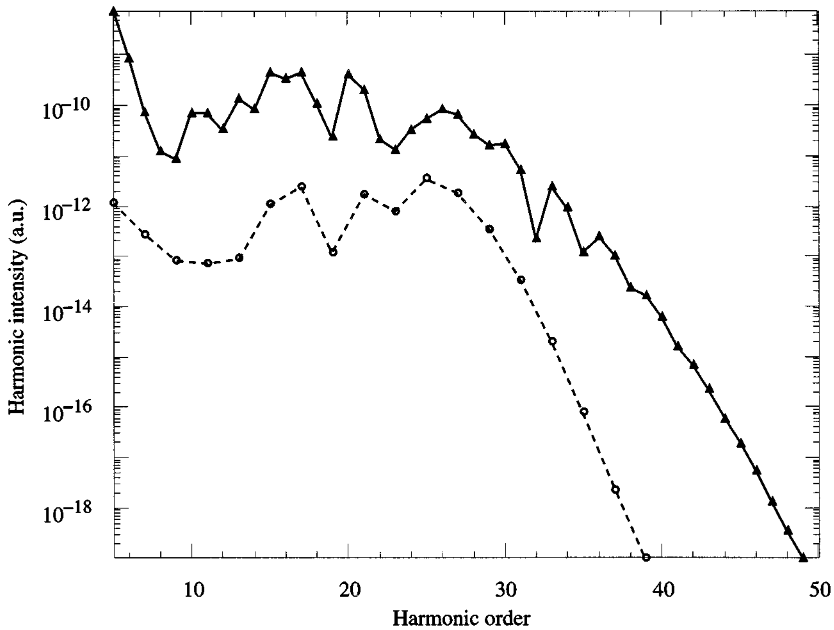

FIG. 5. Harmonic intensity as a function of the harmonic order for monochromatic (ω=1.6 eV, dashed line, open circles) and bichromatic $\omega-2\omega$ (full line, filled triangles) laser fields. Other parameters correspond to the experiment of Eichmann *et al.* (see text).

V. CONCLUSIONS

We generalized the quantum theory of HOHG of Lewenstein *et al.* [12] by a low-frequency laser field to the case of a bichromatic elliptically polarized laser field. By applying the saddle-point method for integration over the impulse $\vec{p}$ we expressed the harmonic strength as a double integral over the return time τ and the time t [Eqs. (15)–(17)]. Our numerical results show that the behavior of the harmonic strength depends mainly on the quasiclassical action $S_s(t,\tau)$, Eqs. (13) and (14). In the bichromatic case S_s contains interference terms that depend on the frequency sum $\omega_1+\omega_2$ and the frequency difference $\omega_1-\omega_2$. The polarization parameters come into the action only through the factors $\cos 2\xi_j$, $\cos(\xi_1+\xi_2)$, and $\cos(\xi_1-\xi_2)$. This is important because, in the special case of a laser field with the time-dependent ellipticity [6], which is constructed using two linearly polarized perpendicular fields, one has $\xi_1=0$ and $\xi_2=\pi/2$. This gives $S_\pm=0$ and the interference part of the action disappears, which simplifies the problem. The laser field with the time-dependent ellipticity is important for obtaining subfemtosecond laser pulses [6]. We will consider this problem elsewhere. For a monochromatic laser field factor $\cos 2\xi$ is in the argument of the Bessel function in Eq. (19), which implies that with the increase of the ellipticity the harmonic strength decreases. This is in agreement with experiments [3,8] and theory [16].

By further applying the saddle-point method to the remaining integrals over τ and t we obtained the cutoff law, Eqs. (23) and (24). In the special case of a bichromatic linearly polarized laser field and in the limit of the vanishing ionization potential the cutoff law can be obtained as the solution of Eqs. (26) and (30) with (27). This solution can also be obtained using the classical model [18,19].

The conclusions of our numerical analysis are the following: (i) the harmonic intensity is much greater in the bichromatic case (for $\omega_2=2\omega_1$ it is greater by about two orders and for $\omega_2=3\omega_1$ by four orders of magnitude); (ii) the cutoff position is shifted toward the lower harmonics; (iii) in the $\omega_2=2\omega_1$ case there are both even and odd harmonics and they are of the same intensity; (iv) the harmonic plateau in the bichromatic case is not constant as in the monochromatic case, but has its own structure (see Fig. 1). In the $\omega_2=2\omega_1$ case it looks like a double plateau, the first part of which is higher (for low harmonics). For the $\omega_2=3\omega_1$ case the conversion efficiency is much higher for low-energy harmonics (in the first third of the plateau); (v) the effect of the electron rescattering is small in the bichromatic case; (vi) the harmonic intensity as the function of the ponderomotive potential has similar behavior to the monochromatic case, but with weaker interference oscillations; (vii) the phase of the induced atomic dipole moment exhibits a quasilinear dependence on the ponderomotive potential, similar to the monochromatic case, but with a lower slope; (viii) the behavior of the harmonic intensity with the change of the relative phase does not depend on the chosen atomic model; (ix) the position of the cutoff is slightly shifted in the case when the relative phase is different from zero (in comparison with the position for $\varphi=0$); (x) by changing the relative phase it is possible to control the height of the harmonic intensity plateau. Finally, the qualitative predictions of our theory are in agreement with recent experiments. For a quantitative analysis one should take into account phase matching and propagation effects.

ACKNOWLEDGMENTS

D.M. thanks the Laboratoire de Physique Atomique et Moléculaire, Université Catholique de Louvain for hospitality and financial support.

[1] A. L'Huillier, K. J. Schafer, and K. C. Kulander, J. Phys. B **24**, 3315 (1991).

[2] *Super-Intense Laser-Atom Physics*, Vol. 316 of *NATO Advanced Study Institute, Series B: Physics*, edited by B. Piraux, A. L'Huillier, and K. Rzążewski (Plenum, New York, 1993).

[3] K. S. Budil, P. Salières, A. L'Huillier, T. Ditmire, and M. D. Perry, Phys. Rev. A **48**, R3437 (1993).

[4] M. D. Perry and J. K. Crane, Phys. Rev. A **48**, R4051 (1993).

[5] K. Kondo, Y. Nabekawa, and S. Watanabe, in *High Field Interactions and Short Wavelength Generation*, OSA Technical Digest Series Vol. 16 (Optical Society of America, Washington, DC, 1994), pp. 236–8; S. Watanabe, K. Kondo, Y. Nabekawa, A. Sagisaka, and Y. Kobayashi, Phys. Rev. Lett. **73**, 2692 (1994).

[6] P. B. Corcum, N. H. Burnett, and M. Y. Ivanov, Opt. Lett. **19**, 1870 (1994); M. Ivanov, P. B. Corcum, T. Zuo, and A. Bandrauk, Phys. Rev. Lett. **74**, 2993 (1995).

[7] H. Eichmann, A. Egbert, S. Nolte, C. Momma, B. Wellegehausen, W. Becker, S. Long, and J. K. McIver, Phys. Rev. A **51**, R3414 (1995).

[8] N. H. Burnett, C. Kan, and P. B. Corcum, Phys. Rev. A **51**, R3418 (1995).

[9] F. A. Weihe, S. K. Dutta, G. Korn, D. Du, P. H. Bucksbaum, and P. L. Shkolnikov, Phys. Rev. A **51**, R3433 (1995).

[10] T. Zuo, A. D. Bandrauk, M. Ivanov, and P. B. Corcum, Phys. Rev. A **51**, 3991 (1995).

[11] S. Long, W. Becker, and J. K. McIver, Phys. Rev. A **52**, 2262 (1995).

[12] M. Lewenstein, Ph. Balcou, M. Yu. Ivanov, A. L'Huillier, and P. B. Corcum, Phys. Rev. A **49**, 2117 (1994); A. L'Huillier, M. Lewenstein, P. Salières, Ph. Balcou, M. Yu. Ivanov, J. Larsson, and C. G. Wahlstrom, *ibid*. **48**, R3433 (1993).

[13] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).

[14] L. V. Keldysh, Zh. Éksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)].

[15] Y. Liang, S. Augst, M. V. Ammosov, S. Lazarescu, and S. L. Chin, J. Phys. B **28**, 2757 (1995).

[16] P. Antoine, A. L'Huillier, M. Lewenstein, P. Salières, and B. Carré, Phys. Rev. A **53**, 1725 (1995).

[17] M. Lewenstein, P. Salières, and A. L'Huillier, Phys. Rev. Lett. **74**, 3776 (1995); Phys. Rev. A **52**, 4747 (1995).

[18] J. L. Krause, K. J. Schafer, and K. C. Kulander, Phys. Rev. Lett. **68**, 3535 (1992).

[19] W. Becker, S. Long, and J. K. McIver, Phys. Rev. A **50**, 1540 (1994).