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Quantum-Mechanical Description of Ionization-Induced Generation of Tunable Mid-Infrared Pulses

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Abstract. The work is devoted to the analytical study of the excitation of low-frequency current density (with frequencies much smaller than the frequency of the optical wave) which radiates in mid-infrared band in the gas ionization by two-color laser pulses. We calculate the low-frequency current density using the analytical solution of time-dependent Schrödinger equation on the basis of strong-field approximation. Closed-form analytical formulas for the low-frequency current density are obtained for laser-pulse parameters corresponding to the tunneling and multiphoton ionization regimes. The features of excitation of low-frequency current density are analyzed in both regimes.

1. Introduction

The process of gas ionization by two-color laser pulses, containing the components with the frequency ratio close to two, is accompanied by excitation of the low-frequency current density (with frequencies much smaller than the frequency of the optical wave) [1–5]. The frequency detuning from the exact ”synchronism” determines the frequency of the low-frequency current, which can be located in mid-infrared band [3–5]. This can be used to generate short pulses of mid-infrared radiation [4]. The low-frequency current density excited during gas ionization by a two-color laser pulse has been previously calculated analytically and numerically with the help of semiclassical and quantum-mechanical approaches [1–9]. The semiclassical approach is based on the solution of the hydrodynamic equation for the electron current density and the equation for the density of free electrons with a quasi-static probability of tunneling ionization per unit time [10]. The quantum-mechanical approach is based on the solution of the three-dimensional time-dependent Schrödinger equation for the electron wave function [9, 11]. The range of applicability of the semiclassical approach is limited by the laser pulse parameters corresponding to the tunneling ionization regime, at which the Keldysh parameter $\gamma = I_p/2U_p$ [12] is much less than unity (here $I_p$ is the ionization potential of an atom, and $U_p$ is the ponderomotive energy of an electron in the laser field) [5, 11, 13]. For $\gamma \gg 1$, the electron release occurs at a time of the order of the field period and greater, and to adequately calculate the low-frequency current density it is necessary to apply the quantum-mechanical approach.

This work is devoted to the derivation of closed-form analytical expressions for the low-frequency current density on the basis quantum-mechanical approach using strong-field
approximation [12, 14]. Previously, such analytic expression was obtained for the laser-pulse parameters corresponding to tunneling regime of ionization [8], where the absolute coincidence with the results of semiclassical approach is achieved [5]. In this paper, we also find an analytic expression for the low-frequency current density for multiphoton ionization regime. The main features of the formation of low-frequency current density in tunneling and multiphoton regimes are discussed.

2. Analytical model

Let the electric field $E(t)$ of an ionizing two-color laser pulse be linearly polarized along the $z$ axis and be consisted of an intense component at the main frequency $\omega_0$ and a weak additional field at a frequency $\omega_1 = 2\omega_0 + \Delta \omega$. The presence of the detuning can be considered as a change in time in the phase shift $\varphi$ between the fields with frequencies $\omega_0$ and $2\omega_0$:

$$E(t) = -\frac{1}{c} \frac{dA}{dt}, \quad A(t) = -\hat{z} \frac{cE_0}{\omega_0} a(t),$$

$$a(t) = f(t) \left( \sin[\omega_0 t] + \frac{\alpha}{2} \sin[2\omega_0 t + \varphi(t)] \right),$$

where

$$\varphi(t) = \varphi_0 + \Delta \omega t.$$  \hspace{1cm} (3)

Here, $\hat{z}$ is the unit vector along the $z$ axis, $E_0$ is the peak amplitude of the main field, $\alpha \ll 1$ is the small ratio of the amplitudes of the additional and main fields, $\omega_0$ is the main frequency, $\varphi_0$ is the constant part of phase shift, $f(t)$ is the pulse envelope, and $c$ is the speed of light in vacuum. We neglect the interaction of atoms with each other assuming that the gas density is sufficiently low. In addition, we do not take into account the polarization response of plasma assuming that the maximum density of plasma is much less than the critical density and plasma frequency is $\omega_p \ll \tau_p^{-1}$. For simplicity, we use atomic units in which $|e| = \hbar = m_e = 1$, where $\hbar$ is the reduced Planck constant, $e$ and $m_e$ are the electron charge and mass, respectively.

The quantum-mechanical approach for calculation of the low-frequency current density is based on the solution of time-dependent Schrödinger equation for the electron wave function $\psi$:

$$i \frac{\partial \psi}{\partial t} = \left( -\frac{1}{2} \nabla^2 + U(r) + zE_z \right) \psi.$$  \hspace{1cm} (4)

Here, $E_z$ is the projection of the electric field strength on the $z$ axis and $U(r)$ is the potential of interaction with the parent ion. For simplicity, we assume that the gas consists of hydrogen atoms. In this case, the potential of the ion is the Coulomb potential, $U(r) = -1/r$. Due to cylindrical symmetry of the system, the excited electron current density is directed along the $z$ axis.

Let us assume that the pulse duration is sufficiently great, so that the concentration of free electrons increases during many periods of the electric field. This makes it possible to present the low-frequency current density as the integral $\bar{j}(t) = \int_{-\infty}^{t} (\partial \overline{j}/\partial t)dt$, where time derivative of the low-frequency current density is

$$\frac{\partial \overline{j}}{\partial t} = -(N_g - \overline{N}) \int p_z \overline{W}(p, t) d^3 p.$$  \hspace{1cm} (5)

Here, $\overline{W}(p, t)$ is the time-averaged momentum distribution of the ionization probability per unit time, $N_g$ is the initial concentration of gas atoms, and $\overline{N}$ is the time-averaged concentration of electrons, which satisfies the approximate equation

$$\frac{\partial \overline{N}}{\partial t} = (N_g - \overline{N}) \int \overline{W}(p, t) d^3 p.$$  \hspace{1cm} (6)
Since the characteristic time of variation of \( W(p, t) \) is much larger than the field period, in order to calculate \( W(p, t) \) we can assume that the electric field has constant envelope and phase shift [8] and that the depletion of the ground state of an atom for the period of the laser pulse field is negligible. The latter assumption is valid even at a very high peak intensity, because in this case, the depletion of the ground state of the atom takes place mainly at the leading edge of the laser pulse at an intensity that is close to the breakdown threshold. Within the framework of the strong-field approximation [12], in which we neglect the interaction of photoelectrons with the parent ion, \( W(p, t) \) is represented as the sum of the probabilities of \( n \)-photon processes:

\[
W(p, t) = \frac{\omega_0^2}{2\pi} |L(p)|^2 \sum_{n=m}^{\infty} \delta (\Delta E - n\omega_0).
\] (7)

Here, \( m = (\hat{I}_p/\omega_0 + 1) \) is the minimum number of photons required for ionization (the expression \( \langle x \rangle \) denotes the integer part of the number \( x \)), \( \hat{I}_p = I_p + U_p, U_p = U_{p0}(1 + \alpha^2/4) \) is the ponderomotive energy in the two-color field, \( U_{p0} = E_{f0}^2/4\omega_0^2 \) is the ponderomotive energy in the field of main field, \( E_{f0}(t) = E_0 f(t) \) is the main field amplitude, \( \Delta E = p^2/2 + \hat{I}_p \) is the energy for ionization and acceleration of the electron, and \( \delta(x) \) is Dirac delta function. Thus, the momentum distribution of photoelectrons is concentrated near spheres with radii corresponding to the final momentum of the photoelectrons which satisfies the energy conservation law, \( \Delta E = n\omega_0 \), where \( n \) is natural number.

The function \( L(p) \) in (8) describes the envelope of the momentum distribution of the ionization probability per unit time. Assuming that the photon energy is much less than the ionization energy (i.e. \( n_0 = I_p/\omega_0 \gg 1 \)), the function \( L(p) \) is

\[
L(p) = \sum_{t_s} e^{iS(p,t_s)+iS_C(p,t_s)} \left( \pi \frac{\partial^2 S}{\partial t^2} |_{t_s} \right)^{-1/2}.
\] (8)

Here,

\[
S(p, t) = \int_0^t \left( \frac{(p + A/c)^2}{2} + I_p \right) dt'
\] (9)

is the part of the action of a free electron which does not depend on the coordinates, \( t_s \) are the stationary points of \( S(p, t) \) and \( s \) is an index numbering the stationary points. The \( t_s \) values satisfy the equation

\[
\frac{\partial S(p, t) / \partial t|_{t_s}}{t_s} = 0
\] (10)

and have a positive imaginary part and a real part which lies in the range \([0, 2\pi/\omega_0]\). \( S_C \) is correction of action associated with the interaction of photoelectrons with the parent ion [15].

Further we take into account the zero-order correction of action which is calculated neglecting the influence of additional-field component. Accounting of this correction in the exponent in (8) leads to the well-known Coulomb factor [15, 16], which is valid for arbitrary values of Keldysh parameter\( \gamma = \sqrt{I_p/2U_{p0}} \),

\[
Q_C = |e^{iS_C}|^2 = (2/E_{f0})^2(1 + 2\gamma/c)^{-2}.
\] (11)

In order to calculate the sum in Eq. (8) we will take into account the action of the second harmonic field by considering terms linear with \( \alpha \). Under the conditions \( \alpha \ll \gamma, \gamma^{-1} \) the stationary points of action are found in the absence of additional field:

\[
t_s^0 = \omega_0^{-1} \arcsin \left( \gamma p_x + i\gamma \sqrt{1 + p_\perp^2} \right),
\] (12)

\[
t_s^1 = \pi/\omega_0 - (t_s^0)^*.
\] (13)
Here \( p_z \) is the projection of momentum on the \( z \) axis, \( p_\perp \) is the module of the transverse momentum. Following the work [12] we assume that the main contribution in \( \mathcal{W}(p, t) \) is given by the small values of final electron momentum, \( p^2 \ll 1 \). It makes possible to neglect the momentum dependence in the pre-exponential factor and use a Taylor series expansion of \( p \) up to the quadratic terms in the exponential factor:

\[
\mathcal{W}(p) \approx \frac{\omega_0 \gamma Q_C}{\pi^2 \sqrt{1 + \gamma^2}} \exp(2n_0b(p)) [\cosh(2n_0a(p)) + \varepsilon(p, \gamma)] \sum_{n=-\infty}^{\infty} \delta(\Delta E - n\omega_0). \tag{14}
\]

Here,

\[
a(p) = \alpha \gamma \cos \varphi \left( \frac{2}{3} + p_\perp^2 \right), \tag{15}
\]

\[
b(p) = \frac{\gamma p_z}{\sqrt{\gamma^2 + 1}} (p_z - \alpha \gamma \sin \varphi) + \frac{\sqrt{\gamma^2 + 1}}{2\gamma} - \left( p^2 + 1 + \frac{1}{2\gamma^2} \right) \arcsinh \gamma. \tag{16}
\]

The term \( \varepsilon(p, \gamma) \) in Eq. (14) is associated with the intercycle interference of two electron trajectories originating from electron ionization from neighboring half-cycles [17]. Therefore, when calculating the integral characteristics such as average ionization probability and current density, one can neglect the term \( \varepsilon \). The rest of the function \( \mathcal{W}(p) \) is a product of the smooth envelope and the sum of delta functions corresponding the spheres defined by the energy conservation law. The maximum of the smooth envelope corresponds to some optimal momentum

\[
p_{\text{opt}} = \frac{\alpha \gamma \sin \varphi}{2 (1 - \sqrt{1 + \gamma^{-2} \arcsinh \gamma})}. \tag{17}
\]

It can be seen that for \( \alpha = 0 \) the optimal momentum is \( p_{\text{opt}} = 0 \) and the function \( \mathcal{W}(p) \) is symmetric in the longitudinal momentum. Therefore, in the absence of the second harmonic the average photoelectron momentum is zero. The addition of a small field at the doubled frequency breaks the symmetry of the momentum distribution of the ionization probability. It leads to the excitation of non-zero low-frequency current density.

Substituting Eq. (14) in Eq. (5) we obtain that the low-frequency current and free-electron density can be represented as a sum over \( n \)-photon processes:

\[
\frac{\partial j}{\partial t} = -C_1 \sum_{n=m}^{\infty} \int_{-p_n}^{p_n} F_n(p_n, p_z) p_z \, dp_z, \tag{18}
\]

\[
\frac{\partial N}{\partial t} = C_1 \sum_{n=m}^{\infty} \int_{-p_n}^{p_n} F_n(p_n, p_z) \, dp_z, \tag{19}
\]

\[
C_1 = (N_g - N), \quad \mathcal{W}(p) = \frac{2\omega_0 \gamma Q_C}{\pi \sqrt{1 + \gamma^2}} \exp \left[ -\frac{2I_0}{\omega_0} \arcsinh \gamma - \sqrt{1 + \frac{\gamma^2 + 1}{2\gamma^2}} \right]. \tag{20}
\]

Here, \( p_n = \left[ n/n_0 - (1 + 1/2\gamma^2) \right]^{1/2} \) is the momentum corresponding to \( n \)-photon absorption, and

\[
F_n = \exp(-2n_0p_n^2 \arcsinh \gamma) \cosh \left[ 2n_0 \alpha \gamma \cos \varphi (2/3 + p_n^2 - p_z^2) \right] \exp \left[ \frac{2n_0 \gamma}{\sqrt{\gamma^2 + 1}} \left( p_z^2 - \alpha \gamma \sin \varphi p_z \right) \right]. \tag{21}
\]

The obtained expression is rather complicated. However, it can be significantly simplified in two limiting cases: in the tunneling regime of ionization (\( \gamma \ll 1 \)) and multiphoton regime of ionization (\( \gamma \gg 1 \)).
2.1. Tunneling limit

In the case $\gamma \ll 1$, the characteristic scale of the envelope distribution of photoelectrons $W(p)$ is much wider than the distance between the spheres of location of function $W(p)$. This allows one to replace the summation by integration in Eq. (18). As a result, time derivative of low-frequency current density is represented as a product of the averaged ionization rate and the most probable momentum

$$\frac{\partial j}{\partial t} = -p_{\text{opt}} \frac{\partial N}{\partial t},$$

(22)

where

$$p_{\text{opt}} = -\frac{3}{2} \frac{\alpha E_0}{\omega_0} \sin \varphi.$$  

(23)

The equation for the averaged electron density becomes

$$\frac{\partial N}{\partial t} = (N_g - \overline{N}) \overline{\omega_0}(E_0) \cosh \left( \frac{2\alpha}{3E_0} \cos \varphi \right),$$

(24)

where

$$\overline{\omega_0}(E) = 4\sqrt{\frac{3}{\pi E}} \exp \left( -\frac{2}{3E} \right)$$

(25)

is the averaged tunneling ionization probability in the main field of the laser pulse.

2.2. Multiphoton limit

In the multiphoton regime of ionization the distance between the spheres (at which the momentum distribution is concentrated) is comparable with the characteristic scale of the distribution. As a result, the main contribution to the low-frequency current density is provided by the electrons which absorb the minimum possible number of photons $n = m$:

$$\frac{\partial j}{\partial t} = \left[ C_2 \sqrt{\frac{2}{\pi n_0}} \sinh \left( \alpha p_m E_0^{-1} \sin \varphi \right) - \frac{1}{2} \alpha \gamma \sin \varphi \frac{\partial N}{\partial t} \right],$$

(26)

$$\frac{\partial N}{\partial t} = C_2 \sum_{s=\pm 1} \exp \left[ so p_m E_0^{-1} \sin \varphi \right] F \left[ \sqrt{\frac{2}{n_0}} \left(p_m + \alpha \gamma \sin \varphi \right)/2 \right],$$

(27)

$$C_2 = (N_g - \overline{N}) 2^{1-2m} \pi^{-1} e^{m+2} \omega_0^{-2m-1/2} E_0^{-2m} \cosh \left( \frac{2\alpha}{3E_0} \cos \varphi \right).$$

(28)

Here $F(x) = \int_0^\infty \exp(y^2 - x^2)dy$ is the Dawson integral.

3. Discussions and conclusions

The obtained expressions reveal that in the tunneling regime of ionization the time derivative of the low-frequency current density is a sine function of the phase shift $\varphi$ between the carriers of the laser-pulse harmonics. Therefore, when the frequency of additional field is detuned from $2\omega_0$, the maximum of low-frequency current spectrum corresponds to the detuning frequency $\Delta \omega$ (in accordance with the results of the semiclassical model [3–5]). As can be seen from the formula (26), in the multiphoton regime $\partial j/\partial t$ is a complex function of $\varphi$. The form of this function depends on the relationship between the parameters $\alpha$, $\gamma$, and $n_0$. In particular, in the multiphoton regime the factor $\cosh \left( 2\alpha \cos \varphi / 3E_0 \right)$ becomes significant. It is due to the fact that the probability of atom ionization dramatically increases when peaks of the laser-pulse components are superimposed (it is implemented for the phase shift $\varphi = 0$). As a result, the form of the spectrum of low-frequency current can differ from the corresponding spectrum in
tunneling regime. In particular, the maximum of spectrum can be shifted from the detuning frequency. However, the strong-field approximation used in this paper may give inaccurate results in finding the dependence of low-frequency current density on the phase. It is due to the fact that the Coulomb correction to the action depends essentially on the final momentum of the photoelectron in multiphoton regime. The account of this dependence can be made on the basis of imaginary time method used in [7, 9] for the calculation of residual current density excited by two-color laser pulse.

Unlike the phase dependences, the analytical result for maximum of time derivative of low-frequency current density (corresponding to the optimum phase) is, apparently, accurate throughout the full range of the Keldysh parameter. This is associated with the fact that accounting for the zero-order Coulomb correction to the electron action leads to a quantitatively correct values of the probability of ionization per unit time [16]. As can be seen from the formula (26), the dependence of the maximum $\frac{\partial j}{\partial t}$ on the intensity of the main field is non-monotonic function for $\gamma \gg 1$. It has minima close to certain intensity values corresponding to the thresholds of closing of ionization channels. When the intensity increases, the distance between the minima is reduced, and they become less pronounced. The investigation of threshold effects in the formation of low-frequency current density can become interesting topic of research, since it is important for understanding the physical bases of the ionization-induced transformation of two-color laser pulses into low-frequency radiation.

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