Analytical study of residual-current excitation during gas ionization by two-color laser pulse

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Abstract. This work is focused on the analytical investigation of the excitation of low-frequency residual current during gas ionization by two-color laser pulses. Such excitation is of great interest since it may be used to convert efficiently laser pulses into low-frequency radiation, in particular, into radiation of the terahertz frequency band. We derive analytical expressions for the residual current density for the cases when two-color laser pulse contains the main strong fundamental-frequency field and additional frequency-tunable field with the frequency close to the double or half frequency of the main field. Derived formulas are used to analyze and compare main features of residual-current excitation in the cases when additional field is second-or half-harmonic of the main field.

1. Introduction

At present, a great attention is given to the terahertz (THz) generation methods based on the use of laser-induced plasmas. These methods are usually realized under the ionization of different gases, including the ambient air, by femtosecond laser pulses. Great interest in these methods is due to the capabilities of obtaining coherent, intense, broadband, and highly directional THz pulses [1]. The THz spectrum usually contains a low-frequency core in the range on few THz (typically, 1–3 THz), where the main energy of the THz pulse is concentrated, and a long high-frequency tail, which can spread to tens of THz [2–7]. The latter can be attributed to the currents varying rapidly under the laser pulse action. The main contribution to the low-frequency THz radiation is made by the plasma currents in the long wakefield of the laser pulse. The amplitude of the low-frequency currents is determined by the free-electron residual current density (RCD) [8–15], which is equal to the zero-frequency spectrum of the time derivative of the current density excited during ionization.

The most popular and efficient laser-plasma schemes of THz generation are the so-called two-color schemes, in which the laser pulse at the fundamental frequency
is supplemented by its second harmonic obtained using a nonlinear crystal \[2, 4, 16-18\]. In the recent works \[19, 20\] THz radiation was generated in the inverted scheme, where two-color femtosecond laser pulse contain, besides the fundamental-frequency main field, a weak additional field tunable near the frequency of the half-harmonic. In order to explain the experimental results, the analytical approach to RCD calculation was developed. The approach is based on calculation of the ionization-induced electron-density harmonics at the frequencies of the main and additional fields \[19\].

In this work we apply the developed analytical approach to derive formulas for the time derivative of low-frequency current and residual current density excited during gas ionization by two-color laser pulses containing the strong fundamental-frequency main field and weak frequency-tunable second-harmonic field \((\omega + 2\omega)\) case. For this case, some analytical expressions were derived in \[21, 22\]. However in the present paper, we for the first time take into account the finiteness of the laser pulse duration, and consider the laser pulses with arbitrary angles between the main and additional field vectors and nonzero frequency detuning of the additional field. The obtained formulas are used to determine the main differences of RCD excitation in commonly used \((\omega + 2\omega)\) case and inverted \(\omega + \omega/2\) case. It is also shown that the frequency detuning of the additional field from the doubled or halved fundamental frequency results in upshift of the low-frequency current spectrum.

2. Analytical model

In order to calculate the density \(N(t)\) of free electrons and their current density \(j(t)\), we use the semiclassical approach, which is valid in the tunneling regime of ionization \[11, 12\]. This approach consists in the solution of the following equations \[4, 7-12, 23\]:

\[
\frac{\partial j}{\partial t} + \nu j = \frac{e^2}{m} N E, \quad \frac{\partial N}{\partial t} = (N_g - N) w(E).
\]

Here, \(N_g\) is the undisturbed gas density, \(\nu\) is the electron collision frequency, \(e\) and \(m\) are the electron charge and mass, respectively, and \(w(E)\) is the tunneling ionization probability per unit time. We assume here for simplicity that ionization degree and the collision frequency are small enough, \(N \ll N_g\) and \(\nu \ll \tau_p^{-1}\), where \(\tau_p\) is the laser pulse duration. Under these assumptions, the RCD \(j_{RCD}\) can be found from the solution of equations (1) for \(\nu = 0\) and \(t \to \infty\),

\[
j_{RCD} = \frac{e^2}{m} \int_{-\infty}^{\infty} N E \, dt, \quad N(t) = N_g \int_{-\infty}^{t} w \, dt.
\]

In what follows, we use equations (2) to find approximate analytical formulas for RCD by using the approach based on calculation of the harmonics of \(N\) at the frequencies of the main and additional fields.

2.1. Strong fundamental and weak second-harmonic field

In this section, we consider that the two-color laser pulse is a superposition of the strong field on the fundamental frequency \(\omega_1\) and weak additional second-harmonic field on the
frequency \( \omega_2 = 2\omega_1 + \Delta \omega \) and is given as

\[
E(t) = E_1(t) \cos(\omega_1 t) + E_2(t) \cos(\omega_2 t + \varphi).
\]

Here, \( \varphi \) is the constant part of phase shift between carriers of fundamental and additional field, \( \Delta \omega \) is the small detuning of the second-harmonic frequency, \( E_1(t) = E_1(t) \hat{x} \) and \( E_2(t) = E_2(t)(\cos \theta \hat{x} + \sin \theta \hat{y}) \) are the slowly time-dependent amplitudes of the fundamental- and second-harmonic fields, respectively, \( \theta \) is the angle between the fundamental- and second-harmonic field vectors, \( \hat{x} \) and \( \hat{y} \) are unit vectors along the x-axis and y-axis respectively.

For small enough frequency detuning, \( \Delta \omega \ll \omega_1 \), the spectrum of free-electron density is localized near the multiples of the fundamental frequency \( \omega_1 \). The change of \( \Delta \omega \) results in shifts of peaks in this spectrum. Thus, one may present free-electron density as

\[
N(t) = [N_1(t)e^{i\omega_1 t} + N_2(t)e^{2i\omega_1 t} + \text{c.c.}] + \delta N(t).
\]

Here, \( N_1(t) \) and \( N_2(t) \) are the complex amplitudes (slow envelopes) of the density oscillations at the fundamental and second-harmonic frequencies, \( \delta N(t) \) is the rest part of \( N(t) \) which includes low-frequency part located near the zero frequency, and high-frequency part localized near the frequencies \( k\omega_1 \) with integer \( k \). In what follows we are interested only in fundamental and second-harmonics of \( N(t) \) since mixing of these harmonics with the laser field leads to the generation of the low-frequency current density. Assuming that the characteristic time \( \tau_e \) of density rise is much greater than the fundamental period \( T = 2\pi/\omega_1 \), we can represent the RCD as

\[
\mathbf{j}_{\text{RCD}} \approx \frac{e^2}{m} \int_{-\infty}^{\infty} \overline{N \mathbf{E}} \, dt,
\]

where the slowly time-dependent period-averaged product of \( N \) and \( \mathbf{E} \) is

\[
\overline{N \mathbf{E}} = \text{Re} \left[ N_1(t)E_1(t) + N_2(t)E_2(t)e^{-i\varphi - i\Delta \omega t} \right].
\]

In order to find approximate expressions for \( N_1(t) \) and \( N_2(t) \), one can use the method presented in [19]. In this method the density harmonics are calculated under the assumption that the pulse envelope is constant and the frequency detuning is zero. We assume that \( w(E) \) is a sharp function, i.e., \( Ew'(E)/w(E) \gg 1 \), where the prime denotes a derivative with respect to the argument. In this case, on the time interval \( T \), the function \( w(E(t)) \) has two sharp maxima which are located near the maxima of \( |\cos(\omega_1 t)| \) (for small enough \( E_2 \)). This allows one to use Laplace’s method and find that

\[
N_1(t) \approx 2N_g \frac{\overline{w(E_1)}}{\omega_1} \frac{(E_1, E_2)}{E_1^2} \sin(\varphi + \Delta \omega t),
\]

\[
N_2(t) \approx -\frac{iN_g \overline{w(E_1)}}{2} \frac{\omega_1}{\omega_1},
\]

where \( \overline{w(E_1)} = [2w^3(E_1)/\pi E_1 w'(E_1)]^{1/2} \) is the average ionization rate for sharp function \( w(E) \). Using (7) and (8), we obtain the expression for the low-frequency current \( \mathbf{j} \),

\[
\frac{\partial \mathbf{j}}{\partial t} \approx \frac{1}{2} \frac{\overline{\omega(E_1)}}{f_1(t)} \frac{\sin(\varphi + \Delta \omega t)}{4 \frac{(E_1, E_2)}{E_1^3} E_1 - \frac{E_2}{E_1}}.
\]
Here, $j_{osc} = e^2 N_g E_{01}/m \omega_1$ is the maximum oscillatory current density in the main field, where $E_{01}$ is the maximum fundamental-harmonic amplitude, $f_1(t)$ is the slowly varying envelope of the fundamental field, i.e., $E_1(t) = E_{01} f_1(t)$.

Taking into account that $\varpi(E_1(t))$ is a sharp function of $t$ near its maximum with time width of $\tau_i$, we can write that $\varpi(E_1(t)) \approx \varpi(E_{01}) \exp(-2t^2/\tau_i^2)$, where $\tau_i = 2[d^2E_1/dt^2]_{t=0} \varpi^2(E_{01})/\varpi(E_{01})]}^{-1/2}$. Since $\tau_i \ll \tau_p$, the calculation of the integral (9) gives

$$J_{RCD} \approx \frac{\sqrt{2\pi}}{4} j_{osc} \varpi(E_{01}) \tau_i e^{-(\tau_i \Delta \omega)^2/8} \frac{E_{02}}{E_{01}} \Phi(\varphi, \theta),$$

(10)

$$\Phi(\varphi, \theta) = \sin \varphi [3 \cos \theta \hat{x} - \sin \theta \hat{y}],$$

(11)

where $E_{02}$ is the maximum second-harmonic amplitude.

2.2. Strong fundamental and weak half-harmonic field

Let us consider now the case when the field on the higher frequency $\omega_2$ has stronger intensity [19]. In this case the field with the frequency $\omega_2 = 2\omega_1 + \Delta \omega$ plays the role of the main field, and the field with the frequency $\omega_1$ is the additional half-harmonic field. The electric field $E(t)$ is specified as follows:

$$E(t) = E_1(t) \cos(\omega_1 t + \varphi) + E_2(t) \cos(\omega_2 t).$$

(12)

We assume that the fundamental field is polarized along the $x$ axis, $E_2(t) = E_2(t) \hat{x}$ and the half-harmonic field is $E_1(t) = E_1(t) (\cos \theta \hat{x} + \sin \theta \hat{y})$, where $\theta$ is the angle between the fundamental- and half-harmonic fields. In this case the free-electron density can be written as

$$N(t) = [N_1(t) e^{i\omega_2 t/2} + N_2(t) e^{i\omega_2 t} + \text{c.c.}] + \delta N(t).$$

(13)

Here, $N_1(t)$ and $N_2(t)$ are the complex amplitudes of the density oscillations at the half-harmonic and fundamental-harmonic frequencies and $\delta N(t)$ is the rest part of $N(t)$. The excitation of low-frequency current density corresponds to mixing the laser field and the fundamental- and half-harmonics of $N(t)$. The low-frequency part of product $N(t)E(t)$ is

$$\overline{NE} = \text{Re} \left[ N_1(t) E_1(t) e^{-i\varphi + i\Delta \omega t/2} + N_2(t) E_2(t) \right].$$

(14)

The envelopes $N_1(t)$ and $N_2(t)$ are [19]

$$N_1(t) \approx -iN_g \frac{\varpi(E_2)}{\omega_2} \frac{E_{\omega_1}}{E_{\omega_2}} e^{-i\varphi + i\Delta \omega t/2},$$

(15)

$$\text{Re}[N_2(t)] \approx \frac{N_g}{4} \frac{\varpi(E_2)}{\omega_2} \frac{E_{\omega_1}^2}{E_{\omega_2}^2} \sin(2\varphi - \Delta \omega t),$$

(16)

and the time derivative of low-frequency current density is

$$\frac{\partial j}{\partial t} = j_{osc} f_2(t) \frac{\varpi(E_2)}{4} \sin(2\varphi - \Delta \omega t) \left[ E_2 \frac{(E_1, E_2)^2}{E_2^2} - 4E_1 \frac{(E_1, E_2)}{E_2^2} \right],$$

(17)
where \( j_{osc} = e^2 N g E_0^2 / m \omega_2 \) is the maximum oscillatory current density in the field of fundamental harmonic, and \( f_2(t) \) is the envelope of fundamental field, \( E_2(t) = E_{02} f_2(t) \). The residual current density is

\[
\mathbf{j}_{\text{RCD}} \approx j_{osc} \frac{\sqrt{2 \pi}}{8} w'(E_0^2) E_{02} \tau_i \left( \frac{E_{01}}{E_{02}} \right)^2 e^{-(\Delta \omega \tau_i)^2/8} \Phi(\varphi, \theta),
\]

where

\[
\Phi(\varphi, \theta) = -\sin(2\varphi)(3 \cos^2 \theta \hat{x} + 2 \sin(2\theta) \hat{y}).
\]

### 3. Discussion

According to analytical formulas (10) and (18) the RCD depends on the intensities \( I_1 \propto E_{01}^2 \) and \( I_2 \propto E_{02}^2 \) of the field components, the angle \( \theta \), and the phase shift \( \varphi \) as the product of separate functions. Moreover, the dependence of the RCD on the intensity of the additional field, the angle \( \theta \) and phase shift \( \varphi \) are independent of a specific form of the function \( w(E) \). This allows one to explain very clearly the results of experiments on the generation of terahertz radiation using two-color laser pulses. In the recent work [19] analytical formula (18) was used to explain the results of some experiment where fundamental and frequency-tunable half-harmonic field were used to generate THz radiation.

Further we discuss some peculiarities of two mechanisms of RCD excitation in which additional field is the second- and half-harmonics of fundamental field. First of all, it should be noted that formulas (10) and (18) describe the high sensitivity of the RCD to a frequency detuning in both the \( \omega + 2 \omega \) and \( \omega + \omega/2 \) cases. With the detuning, the phase shift between the carriers of fundamental and additional fields becomes time dependent. This leads to destructive superposition of the contributions to average drift velocity from the electrons born at different values of the phase shift. Equations (9) and (17) are exactly an indications that if this shift changes drastically over the ionization time \( \tau_i \), the total RCD is significantly attenuated. The dependence of RCD on \( \Delta \omega \) has the form of a resonantlike curve with FWHM \( \omega_d = 4 \sqrt{\ln 4} / \tau_i \) which is equal in both the \( \omega + 2 \omega \) and \( \omega + \omega/2 \) cases. Thus, for incommensurate two-color pulses, the key parameter that determines the efficiency of low-frequency THz generation is \( \Delta \omega \tau_i \).

The derived expression (18) for RCD for the \( \omega + \omega/2 \) case predicts that RCD is equal to zero at \( \theta = \pi/2 \). It can be explained by the disappearance of the density harmonics \( N_1 \) and \( N_2 \), since they are proportional to the dot product \( \langle E_1, E_2 \rangle \) and its square, respectively, due to the Taylor approximation of \( w(\|E(t)\|) \) when complex amplitudes of density harmonics are calculated. In the \( \omega + 2 \omega \) case the RCD does not vanish for any angle \( \theta \). It is connected by the fact that in the \( \omega + 2 \omega \) case the excitation of RCD is due to appearance of first and second harmonics of free-electron density. The second harmonic of density does not depend on \( \theta \) and the term \( N_2 E_2 \) gives the contribution to RCD at any angle \( \theta \).

The derived analytical formulas show that RCD dependence on the intensity \( I_{\text{add}} \) of additional field is different in the \( \omega + 2 \omega \) and \( \omega + \omega/2 \) cases. For the \( \omega + 2 \omega \) case,
\[ |\mathbf{j}_{\text{RCD}}| \propto \sqrt{|I_{\text{add}}|}, \text{ while in the } \omega + \omega/2 \text{ case, } |\mathbf{j}_{\text{RCD}}| \propto I_{\text{add}}. \] This gives the opportunity to get sufficiently high efficiency of generation of RCD in the \(\omega + \omega/2\) case, which efficiency can be as high as in the \(\omega + 2\omega\) case at some laser pulse parameters.

The equations (9), (17) show that the introduction of the frequency detuning \(\Delta \omega\) results in upshift of the low-frequency spectrum of current density. At sufficiently high \(\Delta \omega\) the low-frequency component of free-electron current can have central frequency in the mid-infrared range. This gives an opportunity to use such incommensurate two-color pulses to generate ultrashort pulses in mid-infrared range with controlled carrier-envelope phase.

4. Conclusions

In this work we have investigated analytically the excitation of residual current density (RCD) during gas ionization by two-color laser pulses. We derived simple closed-form analytical formulas for RCD in cases when two-color pulse contains the strong fundamental field and weak frequency-tunable second- or half-harmonic field. The obtained analytical expressions (10) and (18) gives the dependences of RCD on the parameters of two-color laser pulses. It is shown that introduction of the frequency detuning of the additional field from the resonance value (which is doubled- or half-fundamental frequency) results in upshift of the low-frequency spectrum of free-electron current density. This gives an opportunity to use incommensurate two-color pulses to generate ultrashort pulses in mid-infrared range.

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