

Laser-plasma generation of tunable ultrashort pulses in terahertz and mid-infrared ranges

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The generation of tunable ultrashort pulses in the terahertz (THz) and mid-infrared (mid-IR) ranges is an important problem of both fundamental interest and for many applications. In particular, such sources are required for the observation of ultrafast electronic and vibrational dynamics in a wide class of materials [1]. Such sources are also of interest because of the possibilities of spectroscopy and diagnostics of various media [2]. Apart from this, the use of intense ultrashort pulses in the mid-IR range with controlled phase for the generation of high harmonics makes it possible to reach the short-wave region of the soft X-ray range and to obtain ultrashort attosecond pulses [3].

In this paper we report on analytical and numerical investigation of generation of tunable ultrashort pulses in THz and mid-IR ranges based on the ionization-induced multiwave mixing of fields of intense two-color femtosecond pulses. If the ratio of the frequencies $\omega_{1,2}$ of quasi-monochromatic components of the two-color ionizing field is close to an irreducible rational fraction $\omega_2 : \omega_1 = a : b$, where $a + b$ is a not too large odd integer, the ionization-induced multiwave mixing in the produced plasma can result in the efficient excitation of the current of free electrons at a low (compared to the frequencies of the ionizing field) combination frequency responsible for generation of tunable ultrashort pulses in THz and mid-IR ranges [4, 5]. A strong dependence of the ionization rate on the field strength is responsible for a very short duration of generated pulses, which is determined by the ionization duration (characteristic time of increase in the plasma density). As a result, the generated pulses can be few-cycle or even subcycle, and the phase difference between the carrier and envelope in these pulses is determined by the phase shift between single-color components in the ionizing field.

As the most important and practically interesting example of ionization-induced generation of ultrashort pulses in the mid-IR range, we consider here the two-color ionizing pulses containing quasi-monochromatic components whose frequency ratio $\omega_2 : \omega_1$ is close to 2. Such two-color pulses usually consist of the pump field and an additional field, which is obtained using either frequency doubling crystals and frequency-selective elements creating detuning frequency [6] or an optical parametric amplifier generating radiation near half the frequency [7]. We assume that the time-dependent linearly polarized two-color ionizing field is given by

$$\mathbf{E}(t) = [F_1(t) \cos(\omega_1 t) + F_2(t) \cos(\omega_2 t + \varphi)] \mathbf{x}_0, \quad (1)$$

where $F_{1,2}(t)$ are the slow envelopes of single-color quasimonochromatic components, φ is the phase shift, and \mathbf{x}_0 is the unit vector. We consider Gaussian envelopes $F_{1,2} = (8\pi I_{1,2}/c)^{1/2} \exp(-t^2/2\tau^2)$ with the maximum intensities $I_{1,2}$, where $\tau = \tau_p/(4\ln 2)^{1/2}$, τ_p is the intensity full width at half-maximum, and c is the speed of light. Let ω_1 be the lowest of two frequencies, $\omega_2 = 2\omega_1 + \Delta\omega$, and $|\Delta\omega| \ll \omega_1$. We also assume that the duration of the generated pulse is much smaller than the characteristic time of current relaxation in plasma and the period of plasma natural oscillations. As a result, the field (1) can be considered as given and all dissipative terms in equations for low-frequency (at frequencies much lower than ω_1) current density \mathbf{j}_{LF} can be neglected.

The performed *ab initio* simulations of the excitation of the electron current are based on the solution of the three-dimensional time-dependent Schrödinger equation (3D TDSE) [4, 5]. This equation allows describing all stages of the dynamics of the electron wavefunction $\psi(\mathbf{r}, t)$ of the hydrogen atom in a strong external field $\mathbf{E}(t)$. The initial condition corresponds to the ground ($1s$) state. The time derivative of the electron current density is expressed in terms of as

$$\frac{\partial \mathbf{j}}{\partial t} = \frac{e^2 N_m}{m} \left[\mathbf{E} + \left\langle \psi \left| \frac{e\mathbf{r}}{r^3} \right| \psi \right\rangle \right], \quad (3)$$

where N_m is the density of neutral atoms before the ionization, e and m are electron charge and mass, respectively. The temporal profile of $\partial \mathbf{j}_{\text{LF}}/\partial t$ is obtained from $\partial \mathbf{j}/\partial t$ by means of the ideal low-pass filter (the cutoff frequency of the filter corresponds to the lowest frequency above $|\Delta\omega|$ at which the spectral density $\partial \mathbf{j}/\partial t$ reaches a local minimum).

Spectra and time profiles of $\partial \mathbf{j}_{\text{LF}}/\partial t$ calculated from the numerical solution of Eqs. (2) and (3) are shown in Figs. 1 and 2, respectively, for various values of the detuning frequency $\Delta f = \Delta\omega/2\pi$. As is seen in Fig. 1, the spectrum contains a pronounced bell-shaped peak near Δf . The width of the peak is much larger than the widths of the spectra of quasi-monochromatic components of the ionizing field and depends only slightly on Δf , whereas the height of the peak depends on both the magnitude and sign of Δf . Pulses generated at negative detuning are more intense than those generated at positive detuning.

This asymmetry with respect to the sign of detuning can be very large, and the spectral peak at positive detuning can be an order of magnitude lower than the peak at negative detuning. The peaks have

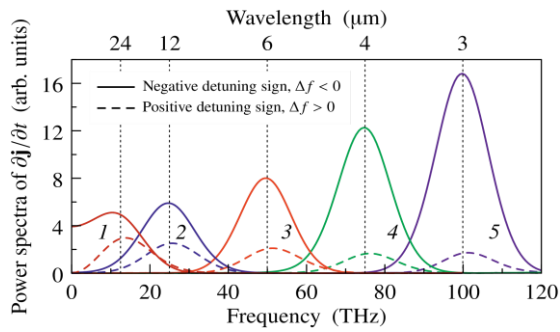


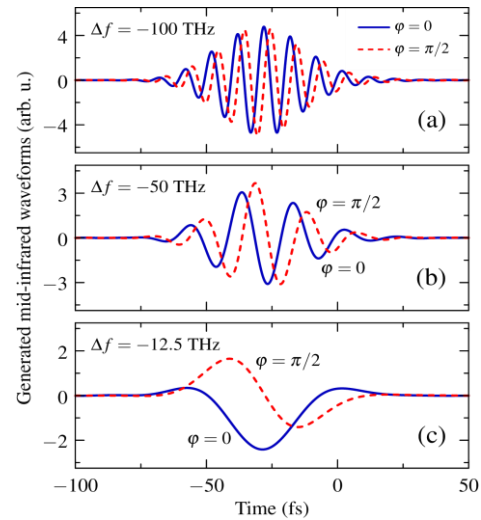
Fig. 1. Power spectra of $\partial\mathbf{j}/\partial t$ found from the numerical solution of the 3D TDSE (2) for the two-color ionizing field (1) at various (dashed lines) positive and (solid lines) negative detuning frequencies $\Delta f = \Delta\omega/2\pi = (1) \pm 12.5$, (2) ± 25 , (3) ± 50 , (4) ± 75 , and (5) ± 100 THz. The frequency ω_1 corresponds to the wavelength 800 nm, components intensities $I_1 = 2 \cdot 10^{14}$ W/cm², $I_2 = I_1/10$, pulse duration $\tau_p = 100$ fs, and phase shift $\varphi = 0$.

smooth flanks, and the corresponding time dependences also have fairly smooth envelopes, as is seen in Fig. 2 plotted for two values of the phase shift. Changing φ by $\pi/2$ results in a phase shift of $\pi/2$ in the generated pulse. The durations of pulses at different detuning values are approximately equal and are much smaller than the duration of the initial ionizing field, which corresponds to the generation of few-cycle or subcycle pulses (depending on the detuning frequency).

For the interpretation of the results of quantum-mechanical simulation, we use an analytical approach based on the equation for the plasma density $N(t)$ and the classical equation for the current density $\mathbf{j}(t)$ of free electrons in a cold collisionless plasma with a varying number of particles

$$\frac{\partial N}{\partial t} = (N_m - N)w(E), \quad \frac{\partial \mathbf{j}}{\partial t} = \frac{e^2 N}{m} \mathbf{E} \quad (4)$$

with the initial condition $N = 0$ (at $t \rightarrow -\infty$) [4, 5, 7]. Here, $w(E)$ is the probability of tunnel ionization per unit time, and it is assumed that the ponderomotive energy of the electron in this field is much higher than the ionization energy. The use of perturbation theory with respect to the corresponding weak field (F_1 or F_2) allows to obtain closed-form analytical expression for $\partial\mathbf{j}_{LF}/\partial t$ and explain the origin of the asymmetry and other characteristic features of the excitation of low-frequency current. Particularly, it is shown that the duration of the excited current is determined by the ionization duration τ_i (the characteristic time of plasma formation), which is much shorter than the duration of the ionizing field because of the steep dependence of the ionization rate on the field strength. At quite large detuning frequencies, $|\Delta f|\tau_i \geq 1$, a pulse is generated at the frequency Δf , and its phase is determined by φ . At smaller detunings $|\Delta f|\tau_i < 1$, a subcycle pulse is generated, and φ affects both the phase and amplitude of the generated pulse. The central frequency of the generated pulse is about $1/\tau_i$, and this value specifies the lower limit of the possible frequency tuning range. In this case, a significant residual current density, i.e., a static component (zeroth harmonic) of $\partial\mathbf{j}_{LF}/\partial t$



Waveforms of generated pulses of the low-frequency current $\mathbf{x}_0(-\partial\mathbf{j}_{LF}/\partial t)$ found from the numerical solution of 3D TDSE at the same parameters as in Fig. 1, but at two phase shifts $\varphi = 0$ and $\pi/2$ and three negative detuning frequencies $\Delta f =$ (a) -100 , (b) -50 , and (c) -12.5 THz.

depending on φ can be excited. The presence of this component can be seen in the results of numerical calculations presented in Fig. 1 (solid line 1) and in Fig. 2c. This residual current density is responsible for the generation of THz radiation [4, 6, 7], and Eqs. (10)–(12) determine the high-frequency part of the spectrum of this THz radiation, which extends to frequencies about $1/\tau_i$.

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