Control of Mid-IR Waveforms Generated During Gas Ionization by Two-Color Laser Pulses

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Abstract. The paper is devoted to further investigation of the generation of tunable mid-infrared radiation during gas ionization by two-color laser pulses. We demonstrate that control of generated mid-infrared frequency can be ensured with the use of linearly chirped laser pulse by changing the group delay between the laser-pulse components.

1. Introduction

The generation of ultrashort pulses in the mid-infrared range with the possibility of frequency detuning is a topical problem, which attracts attention in connection with numerous scientific and practical applications. One of the promising schemes for the generation of such pulses is based on gas ionization by intense two-color laser pulses whose component frequencies are close to irreducible rational fraction, where the sum of the numerator and denominator is a not too large odd integer \cite{1–5}. The duration of the generated pulses, in this case, is determined by the duration of ionization (characteristic time of an increase in the plasma density) and can be very small. The central frequency is determined by the lowest combination frequency that occurs during ionization-induced multiwave mixing in the produced plasma \cite{3, 4, 6}.

The most practically interesting case of implementation of this mechanism corresponds to ionizing pulses containing strong fundamental field and weaker additional second-harmonic field generated by a frequency doubling crystal. Frequency-selective elements \cite{1, 2} were previously considered to create a small frequency detuning of the additional field from the exact doubled fundamental frequency. However, in these schemes, the energy of the additional field rapidly decreases with increasing detuning, which is an obstacle to the tuning of mid-infrared radiation over a wide frequency range.

In this work, we demonstrate another method to control the frequency of mid-infrared pulses generated during gas ionization by two-color laser pulses. This method is based on the use of chirped laser-pulse components and the time shift of the envelopes of these components, which ensures the presence of a frequency detuning of the additional field frequency. We use a quantum-mechanical approach to calculate the low-frequency current density arising from the ionization of argon atoms by a two-color chirped laser pulse and show the control of the central frequency of generated mid-infrared waveforms by changing the group time delay between laser-pulse components.
2. Two-color chirped electric field

Suppose that the electric field of the laser pulse $\mathbf{E}(t) = E(t)\mathbf{\hat{z}}$ is linearly polarized and consists of the fundamental and additional components whose envelopes maxima are shifted by $t_{\text{delay}}$:

$$E(t) = E_0(t) + E_1(t - t_{\text{delay}}),$$

(1)

$$E_0(t) = E_0 e^{-t^2/2\sigma^2} \cos \varphi(t), \quad E_1(t) = E_1 e^{-t^2/2\sigma^2} \cos 2\varphi(t), \quad \varphi(t) = \omega_0 t + c_0 t^2.$$  

(2)

Here, $t$ is time, $E_0$ and $E_1$ are the amplitudes of the fundamental and additional fields, respectively, $\omega_0$ is the fundamental-field frequency, $c_0$ is the value of the linear frequency chirp, $\tau = \tau_p / \sqrt{4 \ln 2}$, $\tau_p$ is the full-width at half maximum (FWHM) duration of the fundamental field. It is assumed that the second harmonic is obtained by passing the fundamental field through a frequency doubling crystal. Therefore, the duration of the additional field is $\sqrt{2}$ times less than $\tau_p$, and instantaneous frequency is two times higher than that of the fundamental field.

As a result of strong ionization nonlinearity, a large set of combination frequencies of a two-color laser pulse arises in the excited current density of free electrons [4, 6]. The smallest of the combination frequencies is determined by the difference between the higher and doubled lower frequencies of the components of the laser pulse [3, 4, 6] and is equal to

$$\Delta \Omega(t_{\text{delay}}) = 2\partial \varphi / \partial t |_{t - t_{\text{delay}}} - 2\partial \varphi / \partial t |_{t} = 4c_0 t_{\text{delay}}.$$  

(3)

Thus, by varying the time delay $t_{\text{delay}}$ and/or the chirp value $c_0$, we can tune the generation frequency $\Omega$ in the mid-infrared range. Below, we confirm this by finding the low-frequency current density from the ab initio numerical solution of the time-dependent Schrödinger equations (TDSE) for argon atoms, which are often used in the experiments on the generation of radiation in hard-to-reach frequency ranges.

3. Numerical method

Interaction of argon atoms with the laser pulse is simulated by the TDSE for the single electron wavefunction $\psi$, initially located in $3p_0$ state (most actively interacting with the external linearly-polarized field) of static ion potential $V_{\text{eff}}(r)$ (frozen-core approximation). The latter is found from the solution of stationary Kohn — Sham equations for the argon atom (atomic units are used):

$$\left(-\frac{1}{2} \nabla^2 + V_{\text{eff}}(r)\right) \psi_{n,0}(r) = E_n \psi_{n,0}(r), \quad n = 1, 2, \ldots N.$$  

(4)

Here, $\psi_{n,0}$ are the wavefunctions of Kohn–Sham orbitals, $N$ is total the number of electrons, $E_n$ is the energy of the $n$-th orbital. The potential energy $V_{\text{eff}}(r)$ is connected with Kohn–Sham orbitals by

$$V_{\text{eff}}(r) = -\frac{N}{r} + V_{\text{ee}}[\rho_0(r)],$$  

(5)

where $\rho_0(r) = \sum_{n=1}^{N} |\psi_{n,0}(r)|^2$ is the electron density, which is assumed spherically-symmetric due to complete filling of the outer shell of the argon atom. The electron–electron interaction potential $V_{\text{ee}}[\rho] = V_H[\rho] + V_{\text{xc}}[\rho]$ consists of the Hartree potential

$$V_{H}[\rho(r,t)] = \int d^3 r' \rho(r', t) \frac{\rho(r', t)}{|r - r'|}.$$  

(6)

desccribing the electron repulsion in the framework of the mean field, and the exchange-correlation potential $V_{\text{xc}}$ for which the spin-unpolarized form of LB94 approximation [7] is used. The equations (4) are solved iteratively in a spherical coordinate system as in [8].
The obtained in such way potential $V_{\text{eff}}(r)$ and initial wavefunction $\psi(r, t = 0) = \psi_{n^* 0}(r)$, where $n^*$ corresponds to the $3p_0$ Kohn–Sham orbital, are substituted in the TDSE

$$
i \frac{\partial}{\partial t} \psi(r, t) = \left[ -\frac{1}{2} \nabla^2 + z E(t) + V_{\text{eff}}(r) \right] \psi(r, t).$$

(7)

Since gas consisting of non-interacting atoms is considered, equation (7) is complemented by standard boundary conditions for $\psi$ at infinity, which follow from the finiteness of the integral of $|\psi|^2$ and $|\nabla \psi|^2$ over the entire space. In numerical calculations, the spatial domain is finite; therefore, it is impossible to satisfy the mathematical boundary conditions directly. To find the approximate solution near the nucleus, we introduce an absorbing layer of width $l_{\text{abs}} = 50$ a.u. near the grid boundary to suppress the reflected waves [9]. The spatial domain boundary is set at a distance $r_{\text{max}} = 250$ a.u. For the numerical solution of the TDSE, wavefunction $\psi(r, t)$ is decomposed into spherical harmonics. The evolution of radial component of wavefunction is performed by split-operator method [10] with the combination of matrix diagonalization, Crank–Nicholson approximation for exponential operators, and Numerov approximation for second-order derivative with respect to radial coordinate.

Based on the Eirenfest theorem, the time-dependent dipole acceleration of the atomic system $a(t) = a(t)\hat{z}$ is expressed as

$$a(t) = -E(t) - \int \frac{\partial V_{\text{eff}}}{\partial z} |\psi(r, t)|^2 d^3r.$$  

(8)

The dipole acceleration is proportional to the time derivative of the electron current density $j = j(t)\hat{z}$,

$$\frac{\partial j}{\partial t} = -N_g a(t),$$

(9)

where $N_g$ is the gas density before the start of the ionization process. The dependence of the time derivative of the low-frequency current density on $t$ is obtained from $\partial j/\partial t$ 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 of $\partial j/\partial t$ reaches a local minimum). To study the temporal dynamics of ionization, we calculate the concentration of electrons outside of the sphere of radius $r_0 = 10$ a.u.:

$$n_{\text{free}}(t) = N_g \int_{r > r_0} |\psi(r, t)|^2 d^3r.$$  

(10)

The radius $r_0$ is chosen large enough to exclude most populated excited bound states; therefore, $n_{\text{free}}$ estimates free-electrons concentration.

4. Numerical results

To demonstrate the method, we consider a two-color chirped laser pulse with the FWHM duration of the fundamental field $\tau_p = 200$ fs, the fundamental frequency corresponding to the wavelength $\lambda = 2\pi c/\omega_0 = 0.8\mu\text{m}$ (where $c$ is the speed of light), the peak intensity of the fundamental and additional fields $I_0 = 2 \times 10^{14}$ W/cm$^2$ and $I_1 = 2 \times 10^{13}$ W/cm$^2$, respectively. We fix the chirp value $c_0 = \omega_0/20\tau_p$ (which corresponds to a change in frequency by 10 % on the FWHM) and vary the group time delay $t_{\text{delay}}$ between the components.

Figure 1 shows the Fourier spectra of the dipole acceleration for two time delays $t_{\text{delay}} = -\tau_p/4$ and $-\tau_p/2$. It is seen that the dipole acceleration contains the low-frequency part with nearly-Gaussian envelope and the maximum at frequency coinciding with the prediction of the formula (3). With an increase in the time delay $t_{\text{delay}}$, the generation frequency continuously increases, which makes it possible to generate pulses of mid-infrared radiation with an arbitrary
Figure 1. Fourier spectra of the dipole acceleration $a(t)$ for the Ar atom in two-color chirped laser pulse with two different group time delays between components: $t_{\text{delay}} = -\tau_p/4$ (blue solid line) and $-\tau_p/2$ (red dashed line), where $\tau_p = 200$ fs is the fundamental FWHM pulse duration. Fundamental-field intensity is $I_0 = 2 \times 10^{14}$ W/cm$^2$, additional-field intensity is $I_1 = 2 \times 10^{13}$ W/cm$^2$, the wavelength of fundamental field is $\lambda_0 = 0.8$ µm, chirp value $c_0 = \omega_0/20\tau_p$. The vertical lines denote the smallest of the laser-pulse combination frequency (3) $|\Delta\Omega(-\tau_p/4)| = 0.05\omega_0$ (solid line), $|\Delta\Omega(-\tau_p/2)| = 0.1\omega_0$ (dashed line).

Figure 2. The waveforms of the time derivative of the low-frequency current density (blue solid lines) and the time dependences of the normalized free-electrons concentration $n_{\text{free}}(t)$ (red dashed lines) for the group time delays $t_{\text{delay}} = -\tau_p/4$ (a) and $-\tau_p/2$ (b).
carrier frequency within limits determined by the spectral width of the fundamental and additional components of the laser pulse. Figure 2 shows the waveforms of the time derivative of the low-frequency current, corresponding to the two considered values of the detuning time $t_{\text{delay}}$.

As follows from the figure, the amplitudes and temporal envelopes of the generated waveforms approximately coincide with each other. The temporal envelopes have maxima near the moment of free-electrons concentration fastest growth, which indicates that the dominant mechanism of the low-frequency current generation is the ionization-induced multiwave mixing [3, 4, 6]. By controlling the intensity of the components of the laser pulse, one can control the duration of the generated radiation, which opens up the possibility of generating few-cycle and even sub-cycle pulses in the mid-infrared range [3, 4].

5. Conclusions
Based on the numerical solution of the time-dependent Schrödinger equation for the argon atom, it is shown that the use of a linear chirp of components of a two-color laser pulse with a frequency ratio close to two allows effective control of the generated mid-infrared frequency. By changing the group time delay between the components of a two-color laser pulse, it is possible to tune the spectrum of the generated radiation in a wide frequency range determined by the spectral widths of the laser-pulse components.

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