Generation of sub cycle terahertz pulses via coherent control of nonlinear medium by femtosecond pulses

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Abstract. In this paper, we revise our recent advances in study of high-efficient methods of sub cycle THz pulse generation and control of their wave shape. These methods are based on coherent control of low frequency oscillations in nonlinear medium excited by infrared femtosecond pulses. Our results showed the possibility of sub and few-cycle pulse formation of controllable wave shapes: half-cycle, rectangular and triangular and single-cycle ones in THz, XUV and optical ranges.

1. Introduction

Generation of broadband sub cycle terahertz (THz) waveforms with controllable wave shapes became of great interest in the past decades because of huge amount of possibilities for their applications in ultrafast spectroscopy, imaging, and charges acceleration, see reviews [1-3] and references therein. Methods of THz pulses generation developed so far are based on nonlinear frequency conversion of high power femtosecond pulses to THz frequencies [1-3]. Although high efficiencies of THz generation are nowadays achievable, looking for methods which are highly efficient yet allowing precise pulse shape control at the sub cycle scale is an actual problem.

In this paper, we revise our recent advances in study of high-efficient methods of sub cycle THz pulse generation and control of their wave shape [4-12]. These methods are based on coherent control of low frequency oscillations in nonlinear medium (in particular, Raman Active Medium) excited by infrared femtosecond pulses. We show the possibility of THz pulse formation of controllable wave shapes in THz and XUV ranges.

2. Physical idea of coherently controlled generation of sub cycle THz pulses

Let us consider a thin layer of nonlinear medium located along \( z = z_1 \) and \( z = z_2 \), and which has resonances at THz frequency. This layer is irradiated by a pair of infrared femtosecond pulses with a distance between them of \( T_d = T_0/2 \) equal to the half period of proper oscillations. In this case, the first pulse starts the low-frequency oscillations of medium polarization, whereas the second pulse stops them. As a result medium polarization has a shape of half-wave (shown in the inset to Fig.1). This polarization can be a source of sub cycle THz wave form. Pulse duration and delay between them is smaller than polarization relaxation of the medium \( T_2 \). Since, coherent interaction of the pulses with the medium takes place and THz wave emission mechanism is based on free induction decay mechanism [13].
To describe nonlinear medium polarization a simplest oscillator model is used. In this model, medium polarization is governed by the equation [8]

\[ \ddot{P}(t) + \gamma \dot{P}(t) + \omega_0^2 P(t) = g_0 E^2(t), \]  

where \( g_0 \) is the coupling to the external field, \( \gamma \) the damping rate, \( \omega_0 \) the eigen frequency, which is assumed to be in THz range, and \( E(t) \) is the electric field of the pumping infrared femtosecond pulses. It is seen from the right side of Eq. (1) that medium has quadratic response to the external field. This response is typical for Raman Active Media (RAM) [13]. A more detailed molecular RAM model is based on two-coupled nonlinear high frequency (electron) and low-frequency (nucleous) oscillators. This system of equations for displacement of high frequency oscillator \( x(t) \) and low frequency one has the form

\[ \ddot{x}(t) + \gamma_e \dot{x}(t) + \Omega_0^2 x(t) = \frac{q}{m} E(t) - \frac{k}{2M} x^2(t), \]  

\[ \ddot{y}(t) + \gamma_n \dot{y}(t) + \omega_0^2 x(t) = -\frac{k}{2M} x^2(t), \]  

here \( m \) is the electron mass, \( M \) is the nucleus mass, \( k \) is the coupling coefficient between two oscillators, \( q \) is the electron charge. It is seen that in the right sight of the Eq. (3) there is a term \( x^2(t) \), i.e. the displacement of low frequency oscillator is proportional to the square of pump field as in Eq.(1). The medium is excited by a pair of Gaussian pulses

\[ E(t) = E_0 e^{-\frac{t^2}{\tau_p^2}} \sin(\omega_p t + \phi) + E_0 e^{-\frac{(t-T_d)^2}{\tau_p^2}} \sin(\omega_p (t - T_d) + \phi). \]  

Here \( \phi \) is the carries envelope phase of the pulses (CEP). The results of the generated field calculations is presented in the following sections below. The generated field of THz wave is calculated using the wave equation

\[ \Delta E(r, t) - \frac{1}{c^2} \frac{\partial^2 E(r, t)}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial^2 p(r, t)}{\partial t^2}. \]  

Numerical simulations were performed with both models of the medium response given by Eq. (1) and Eqs. (2)-(3). Using the both models led approximately to same results [5]. Below we will see that the result of THz wave shape calculation depends on the problem dimension and setup configuration.

3. Generation of sub-cycle THz pulses in 1D geometry

In 1D geometry valid for broad beams at the distances much smaller than the diffraction length the reflected field \( E_g(t) \) in the situation showed in Fig.1 is given by [4]

\[ E_g(t) = -\frac{1}{2\epsilon_0 c} \int_{x_1}^{x_2} \frac{\partial}{\partial t} P(x', t - \frac{x - x'}{c}) dx'. \]  

Eq. (6) is the analytical solution of wave equation (5) in 1D. According to Eq. (6) the generated THz pulse shape is proportional to the derivative of the polarization half-wave shown in Fig.1. This
derivative contains one-cycle of oscillation and the generated THz pulse has single-cycle shape. Fig.2 shows the example of the excitation field time dependence (a), and its spectra (c) as well as generated THz pulse time dependence (b) with corresponding spectra (d) [5].

Figure 2. The driving field (a), its spectrum (b); the spectra (c) and the temporal profiles (d) of the single-cycle THz pulses. Layer length is 8 μkm.

The medium response was calculated using Eq. (1). The field was calculated numerically by solving the 1D wave equation Eq. (5) with Eq. (1). It is seen from Fig.2b that generation of single-cycle THz pulse is possible. This method possesses very high efficiency of generation ~ 10^{-4}-10^{-3} [5].

Similar idea of coherent control of medium oscillations can be used for attosecond pulse generation in the XUV and optical range [6]. The situation is similar to that shown in Fig.1. The gas medium is excited by half-cycle X-ray pulses. As the medium example in Ref. [6] neon gas was considered. It has a strong resonant transition from ground state to the first excited state with wavelength 60 nm. The remaining levels of helium are relative close to the first excited state. The distance between other levels decreases very rapidly. In this sense, two-level approximation to the helium atom can be applied with high accuracy.

Furthermore direct numerical simulations of the time dependent Schrödinger equations have shown that the ionization probability of neon in this case is relatively small ~ 10^{-11} [6]. This allows us to neglect ionization of neon. Example of calculated single-cycle XUV attosecond pulses wave forms are shown in Fig.3. Neon medium response is governed by system of density matrix equations for two-level medium. Generated attosecond field time profiles were calculated using Eq. (5) optically thin layer of two-level Ne atoms, see Fig.3.

Figure 3. The calculated single-cycle XUV attosecond pulses time dependence at different values of X-Ray pumping field CEP. A thin layer of neon gas is excited by a pair of X-Ray half-cycle pulses (3) with E_0=3\times10^7 V/cm, \tau_p = 3 as. Layer thickness is h=40 nm. Other parameters can be found in Ref. [6].

4. Generation of few-cycle THz pulses in 3D case

In 3D case generated field is proportional to the second derivative of the medium polarization half-wave. This derivative has a shape of the half-wave and a high frequency oscillation tail of opposite polarity. These tails can be almost cut off by appropriate spectral filter, hence, the influence of this tail can be
neglected [7-12]. To control the shape of generated THz pulses it is necessary to introduce in the setup delay in the arriving of these half-waves to the observation point.

Figure 4. (a): Scheme of the setup, (b): example of generated THz pulses time dependence obtained using SPP and axicon.

This delay can arise, for example, via superluminal excitation of the medium by ultra-short pulses with wave front obliquely incident to the medium [7-8, 12] or when the medium is excited by spot of light rotating at high superluminal speed [9-10]. More practical way proposed in Ref. [11] to use spiral phase plate (SPP). SPP, which thickness or radius depend on polar angle, can be used to create such a delay, see Fig.4a. Theoretical analysis has shown the possibility of rectangular and triangular unipolar-like THz pulse generation, see Fig.4b and Ref. [11].

5. Conclusions

In this paper we revised our recent results on sub-cycle THz and XUV electromagnetic pulse generation. Physical idea of the method is based on coherent control of resonant transition in the medium excited by ultra-short pulses with appropriate delay. By the appropriate choice of the setup it is possible to control the shape of generated pulses.

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