Optical rectification upon self-action of femtosecond laser pulses in air

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The paper analyzes possible mechanism of manifestation of second-order nonlinear effects, in particular, the effect of optical rectification, upon propagation of femtosecond laser pulses in air under self-action conditions. With a Ti:Sapphire laser as an example, it is shown that the laser pulse propagation under such conditions is accompanied by generation of a video pulse, whose shape is determined by the derivative of the spatiotemporal shape of the laser field envelope.

Introduction

Optical rectification, along with the second harmonic generation, falls in the category of the second-order nonlinear effects, in which the nonlinear polarization of the medium depends quadratically on the optical field strength:

$$P_i^{(2)}(\mathbf{r},t) = \sum_{jk} \chi_{ijk}^{(2)} E_j(\mathbf{r},t) E_k(\mathbf{r},t), \qquad (1)$$

where $P_i^{(2)}(\mathbf{r},t)$ and $E_j(\mathbf{r},t)$ stand for the nonlinear polarization of the medium and the optical field strength, respectively; χ is the nonlinear quadratic susceptibility of the medium.

The quadratic nonlinearity, as well as other even nonlinearities, is characteristic only of media without centers of symmetry, for example, for anisotropic crystals (see, for example, Ref. 1). Propagation of high-power femtosecond pulses in such crystals can be accompanied by generation of video solitons of femtosecond duration,[2] keeping their shape during the propagation in a medium of electromagnetic pulses free of a high-frequency carrier as in ordinary optical radiation.

In gas media, due to their isotropy, the lowest nonlinearity is of the third order and is connected with self-action effects, third harmonic generation, etc. Under the self-action conditions in such media, second-order nonlinear effects are principally impossible. These effects become possible in isotropic media only in the presence of additional constant electric field.[1]

Because of the well-known unique properties of femtosecond laser pulses, their interaction with medium is accompanied by a wide variety of different nonlinear phenomena,[3] whose combination can provide for the channels of nonlinear interaction with the medium, which do not occur in traditional nonlinear optics characteristic of nano- and picosecond pulses. Thus, Ref. 4 reports the experimental observation of second harmonic generation in air upon propagation of femtosecond Ti:Sapphire laser pulses. The results from Ref. 4 indicate that the second harmonic signal is proportional to the squared pulse energy. This may be one of explanations for the analogous dependence of a photoacoustic detector signal upon the femtosecond laser pulse propagation in air.[5]

The aim of this work was to analyze possible mechanisms for second-order nonlinear effects in isotropic media.

1. Formulation of the model

One of the technical achievements of femtosecond optics is effective multiphoton and tunnel ionization of air by IR laser radiation, during which the number of "simultaneously" absorbed photons achieves ~ 6–8 [Ref. 6]. The typical model of generation of free electrons in air under the effect of femtosecond laser pulses is determined by the equation[7–9]:

$$\frac{\partial \rho}{\partial t} = \sigma^{(k)} I^k(\mathbf{r},t)(\rho_{\text{at}} - \rho), \qquad (2)$$

where ρ is the charge concentration; $\sigma^{(k)}$ is the coefficient of multiphoton ionization; $k = \text{mod}(U/\hbar\omega + 1)$ is the number of light quanta needed for ionization (in air the oxygen molecule has the lowest ionization potential ($U = 12.1$ eV) and $k = 8$ for Ti:Sapphire laser[10]); $\hbar\omega$ is the quantum of laser radiation; ρ_{at} is the initial concentration of charges in the atmosphere.

Free charges with the time-variable density form an electromagnetic wave in the medium, and the field strength $\mathbf{E}_e$ of this wave can be described by the wave equation:

$$\nabla^2 \mathbf{E}_e - \frac{1}{c^2}\frac{\partial^2 \mathbf{E}_e}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial \mathbf{j}_e}{\partial t} - 4\pi e \nabla \rho. \qquad (3)$$

Here e is the electron charge; $\mathbf{j}_e$ is the density of the free-electron current. To determine this parameter, we can use the equation[9]:

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$$\frac{\partial \mathbf{j}_e}{\partial t} = \frac{e^2}{m}\rho \mathbf{E}_e - \frac{\mathbf{j}_e}{\tau_c}, \quad (4)$$

where τ_c is the time of collisional relaxation of plasma. For air under normal conditions $\tau_c = 3.5 \cdot 10^{-13}$ s [Ref. 8].

Equation (3) ignores the secondary effects connected with the interaction of $\mathbf{E}_e$ with the medium, which are described by the constitutive equations of the following form

$$\mathbf{P} = f(\mathbf{E}_e),$$

where $\mathbf{P}$ is the specific polarization of the medium.

The presence of the additional field $\mathbf{E}_e$ in the medium in combination with the powerful laser field $\mathbf{E}$ can leads to nonlinear polarization of air, which, in particular, has the following term:

$$\mathbf{P}^{(3)}(\mathbf{r},t) = \chi^{(3)} \mid E(\mathbf{r},t) \mid^2 \mathbf{E}_e(\mathbf{r},t) = n_2 I(\mathbf{r},t)\mathbf{E}_e(\mathbf{r},t), \quad (5)$$

where n_2 characterizes the nonlinear change of the refractive index of the medium.

If $\mathbf{E}_e$ has a quasistationary character – at least, does not change with the frequency close to the frequency of the laser field, the nonlinear polarization of the medium (5) can lead to the effect analogous to optical rectification.

The calculations were performed for the following values of the parameters: Ti:Sa laser with the pulse duration $\tau_p = 50$ fs and energy 10 mJ, beam diameter of 3 mm; $\sigma^{(k)} = 3.7 \cdot 10^{-96}\ \mathrm{s}^{-1} \cdot \mathrm{cm}^{16} \cdot \mathrm{W}^{-8}$ [Ref. 13], $\rho_{at} = 2.7 \cdot 10^{19}\ \mathrm{cm}^{-3}$ [Ref. 13]. Note that the available literature data for the nonlinear response of the medium correspond to the pulsed radiation with the high-frequency carrier falling within the region of visible or UV radiation. Strictly speaking, the polarization field (5) is not of this kind, but the frequency dependence of n_2 is not very strong, and therefore the value for the UV region: $n_2 = 8 \cdot 10^{-19}\ \mathrm{s}^2 \cdot \mathrm{W}^{-1}$ [Ref. 7] was used for estimates.

2. Analysis of the free charge field

Let us analyze the properties of the free charge field arising due to the multiphoton ionization of air by a powerful laser field. To do this, decrease the order of Eq. (3).

As a rule, to decrease the order of a wave equation describing the propagation of optical pulses, in particular, those of femtosecond duration, the method of slowly varying amplitudes (SVA) is used.[3,11] Because the field of free charges cannot be believed *a priori* as having a high-frequency carrier, the SVA method is inapplicable in this case. Therefore, to decrease the order, we use the approximation of unidirectional propagation.[12] Equation (3) in the approximations of unidirectional propagation and quasioptics takes the form

$$\frac{\partial \mathbf{E}_e}{\partial z} + \frac{1}{c}\frac{\partial \mathbf{E}_e}{\partial t} = \frac{c}{2}\int_0^t \nabla_\perp^2 \mathbf{E}_e \mathrm{d}t' - \frac{2\pi}{c}\mathbf{j}_e - 2\pi c \int_0^t \nabla \rho \, \mathrm{d}t'. \quad (6)$$

Assume, for simplicity, that the laser field has a form of a slit beam. Then, in the scalar approximation, the transversal component of the field $E_e = (\mathbf{E}_e)_x$ is described by the equation

$$\frac{\partial E_e}{\partial z} + \frac{1}{c}\frac{\partial E_e}{\partial t} = \frac{c}{2}\frac{\partial^2}{\partial x^2}\int_0^t E_e \mathrm{d}t' - \frac{2\pi}{c} j_e - 2\pi c \int_0^t \frac{\partial \rho}{\partial x}\mathrm{d}t'. \quad (7)$$

The propagation of the free charge field, described by Eqs. (2), (4), and (7), was analyzed by numerical methods, taking into account the following conditions:

$$\frac{\rho}{\rho_{at}} \ll 1, \quad \frac{\tau_p}{\tau_c} \ll 1. \quad (8)$$

The laser pulse field at the entrance into the medium has the form

$$E(x,z,t)\mid_{z=0} = \begin{cases} E_0 \sin(\pi t/\tau_p)\exp(-x^2/x_0^2)\exp(i\omega t), & 0 \le t \le \tau_p \\ 0, \quad t > \tau_p,\ t < 0 \end{cases}, \quad (9)$$

where x_0 is the initial beam radius. For simplicity, the approximation of the given pump field, which is quite typical for problems of nonlinear parametric interaction, was used, that is, the field of laser pulse in the medium was believed to have the form (9).

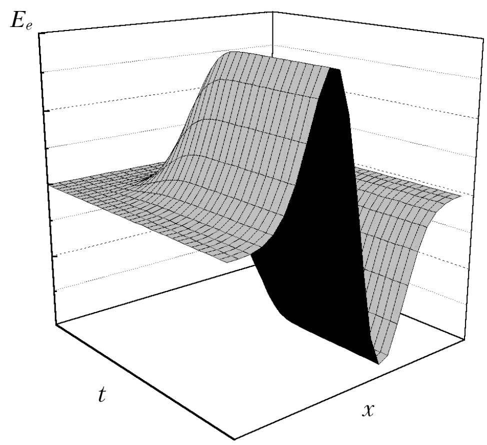

Fig. 1. Spatiotemporal structure of the field of free charges generated by a femtosecond Ti:Sa laser pulse in air.

The field strength of free charges in the medium is shown in Fig. 1. It can be seen that the field, indeed, has not a high-frequency carrier, that is, has a quasistationary character. Note that, taking into account the collisional relaxation τ_c after the beginning of interaction with a laser pulse, the free charge field decays with time.

3. Analysis of optical rectification

Let us analyze the field generated by the cubic nonlinearity of the form (5). The calculations use the following form of the wave equation, describing the distribution of the strength E_d of the field in the medium in the approximation of unidirectional propagation:

$$\frac{\partial E_d}{\partial z} + \frac{1}{c}\frac{\partial E_d}{\partial t} = -\frac{2\pi}{c}\frac{\partial P^{(3)}}{\partial t}, \qquad (10)$$

where the cubic polarization of the medium $P^{(3)}$ is determined by Eq. (5). Note that the diffraction blooming is neglected here for simplicity, because the extent of the field along the direction of propagation is $c\tau_p \ll x_0$.

Figure 2 depicts E_d calculated by Eq. (10).

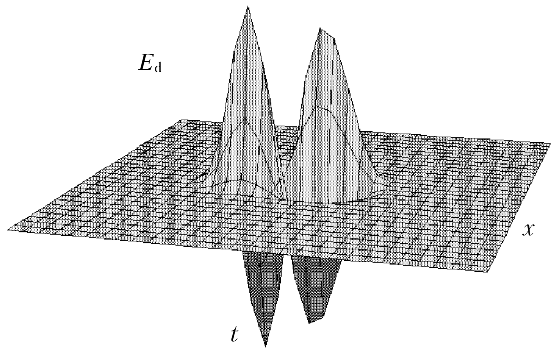

Fig. 2. Spatiotemporal structure of the video pulse field.

In addition, Fig. 3 shows the lines of equal strength of E_d in relative units E_d/E_0, where E_0 is the maximal strength of the laser field after propagation of a distance of about 0.4 cm long in the medium.

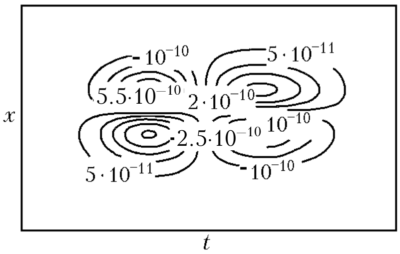

Fig. 3. Lines of equal strength of the video pulse field.

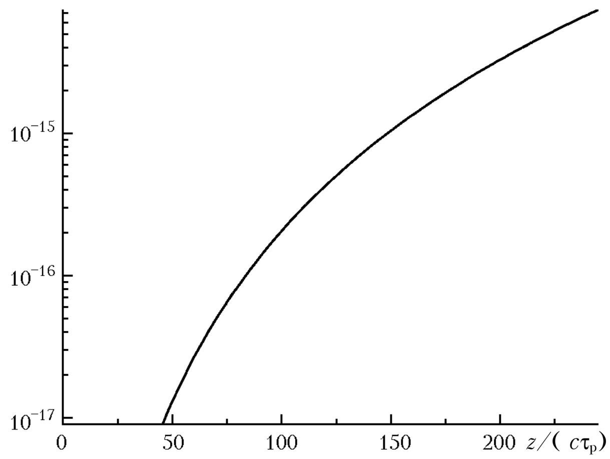

Fig. 4. Dynamics of increase of the relative energy of a video pulse W_d/W upon the propagation of femtosecond Ti:Sa laser pulse in air; W_d is the video pulse energy; W is the laser pulse energy.

It is seen from Figs. 2–3 that, in contrast to the effect of optical rectification in a constant electric field,[1] in this case the structure of the formed video pulse is determined by the derivative of the spatiotemporal form of the laser field envelope.

Note that, within the framework of the approximations used, the rate of energy transfer from a laser pulse to the wave of the video pulse is close to exponential (Fig. 4).

Conclusions

This paper has considered a possible mechanism for manifestation of second-order nonlinear effects, in particular, the effect of optical rectification, during the propagation of femtosecond laser pulses in air under self-action conditions. It has been shown that in this case the laser pulse propagation is accompanied by generation of a video pulse, whose shape is determined by the derivative of the spatiotemporal form of the laser field envelope. This phenomenon can be used for spectral conversion of femtosecond laser pulses.

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