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Extremely short pulses in anisotropic optical media with carbon nanotubes taking into account multiphoton absorption

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In this paper, we study the influence of multiphoton absorption on the propagation of electromagnetic waves in a nonlinear optical anisotropic medium with carbon nanotubes. The effective equation for the vector potential of the electromagnetic field of the pulse is obtained, taking into account the second component of the polarization of the field, as well as two- and three-photon absorption processes. The dependence of the components of the pulse field on the parameters of the problem is revealed.

Keywords: optical anisotropy, extremely short pulse, multiphoton absorption.

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Introduction

In recent years becomes popular the direction of research related to investigation of features of the interaction between high-power electromagnetic radiation and substances, that find a lot of practical applications in most unlikely fields, including biology, astrophysics, and many others [1,2].

It is important to note, that with high radiation intensity the major role is played by multiphoton processes. It means that in the elementary act of interaction between light and a substance atom not one, but several photons are absorbed.

Since the theoretical work by Maria Goeppert-Mayer in 1931, this absorption process has been extensively studied for its significance in both fundamental science and technological applications. For instance, this process can be used for frequency conversion to realize light at a frequency for which no available light source exists. In terms of fundamental science, multiphoton processes can be used to examine and control those quantum states and chemical reactions that cannot be accessed by the one-photon process [3].

Multiphoton absorption is the cause of many interesting effects in different media. First of all, we are interested in media that contain carbon nanotubes (CNT) [4], which proved to be good in terms of stabilization of the pulse propagating in the media with CNTs [5,6]. Here we note just a few of nonlinear optical effects caused by multiphoton processes. For example, photoluminescence from multiwall CNTs and graphite suspensions [7]. Authors of [8] propose an approach to extend performances of optical limiters over broad spectral and temporal ranges due to the association of non-linear scattering from single-wall carbon nanotubes (CNT) and multiphoton absorption from organic chromophores. Also, we should like to note that two-photon absorption is an elegant technique to determine excitonic effects in CNTs [9].

In this study, in addition to the multiphoton absorption, we also take into account optically anisotropic properties of nonlinear medium [10,11], i.e. the effect of the general polarization itself on the propagation of extremely short pulses.

Model and basic equations

Let us consider a dielectric anisotropic medium where CNTs with impurity are placed. Axes of the Cartesian coordinate system are co-directional with axes of the crystal. Axes of the CNTs lie in the XOY plane and make angle α with the OX axis. The electric field is co-directional with the OX axis [12].

The dispersion law for electrons of CNTs is as follows [13]

$$\varepsilon(p,s) = \pm\gamma_0\sqrt{1+4\cos(a\,p)\cos\left(\frac{\pi s}{m}\right)+4\cos^2\left(\frac{\pi s}{m}\right)}, \tag{1}$$

where $s = 1, 2, \ldots, m$, nanotube type is $(m, 0)$, $\gamma_0 \approx 2.7\,\text{eV}$, $a = 3b/2$, b — distance between neighboring carbon atoms.

Vector potential is as follows: $\mathbf{A} = \big(A_x(x,y,z,t), A_y(x,y,z,t), 0\big)$, electric current density is $\mathbf{j} = \big(j_x(x,y,z,t),\ j_y(x,y,z,t), 0\big)$.

By changing to cylindrical coordinate system and taking into account the following calibration: $\mathbf{E} = -c^{-1}\partial\mathbf{A}/\partial t$, we can write a three-dimensional wave equation for two components of the vector potential:

$$\Box A_x + \frac{4\pi}{c}\,j_x(A_x)\,f(t) + \Gamma_x\,\frac{\partial A_x}{\partial t} - F_1\left(\frac{\partial A_x}{\partial t}\right)^{2n_p-1} = 0,$$

$$\Box A_y + \frac{4\pi}{c} j_y(A_y) f(t) + \Gamma_y \frac{\partial A_y}{\partial t} - F_1 \left(\frac{\partial A_y}{\partial t} \right)^{2n_p - 1} = 0, \tag{2}$$

where the first term of sum in equations (2) is the d'Alembertian operator in a cylindrical coordinate system, acting on components of the vector potential (A_x, A_y):

$$\Box A_x = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial A_x}{\partial r} \right) + \frac{\partial^2 A_x}{\partial z^2} + \frac{1}{r^2} \frac{\partial^2 A_x}{\partial \phi^2} - \frac{1}{v_o^2} \frac{\partial^2 A_x}{\partial t^2},$$

$$\Box A_y = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial A_y}{\partial r} \right) + \frac{\partial^2 A_y}{\partial z^2} + \frac{1}{r^2} \frac{\partial^2 A_y}{\partial \phi^2} - \frac{1}{v_o^2} \frac{\partial^2 A_y}{\partial t^2},$$

$$v_o = c/n_x, \quad v_e = c/n_y, \tag{3}$$

where r, z, φ — coordinates in the cylindrical coordinate system, n_x, n_y — refraction indices in directions of x and y, respectively, c — speed of light, n_p — number of photons, F_1 — coefficient of multiphoton absorption [13]. Parameters Γ_x, Γ_y describe the pumping of electric field in the direction of x and y,

$$f(t) = \begin{cases} 0, & t < t_0(z), \\ \exp\left(-\frac{t}{t_{\text{rel}}}\right), & t \geq t_0(z), \end{cases} \tag{4}$$

where $t_0(z) \cong (z - z_0)/v$ is the time at which the intensity of the pulse at its leading edge, measured at the point with the z coordinate, is e times less than the peak intensity of the pulse; z_0 is the initial coordinate of the „center of mass" of the pulse at the initial time $t = 0$, $v \cong c/\sqrt{k_0}$ is the approximate pulse velocity by the order of magnitude; k_0 is the average relative dielectric permittivity of the medium (array of nanotubes); t_{rel} is the relaxation time of the electron subsystem with CNTs [5].

Note that $f(t)$ is a correction factor to the expression for current density in the collisionless approximation, that makes it possible to take into account the exponential current attenuation under the action of relaxation. This approach is demonstrated in [14] for a supergrating and allows us to take into consideration the effect of electron collisions. Since current densities in the supergrating and in systems with CNTs are similar to each other, it allows us to apply this approach to our problem as well.

In previous studies devoted to birefringence a strong spreading of the pulse was reported due to the presence of the second component of field polarization [10,11]. To stabilize the pulse, it is proposed to use external field pumping as an option to alleviate the unavoidable dissipative effects that lead to the damping of the pulse. Thus, in [15] we have shown that propagation of three-dimensional extremely short optical pulses in principle can be maintained in CNTs by energy pumping into the pulse through an external electromagnetic field. Let us select the parameter Γ, which is responsible for the pumping of the electric field with the super-Gaussian profile:

$$\Gamma = Q_\Gamma \exp\left(-\frac{r^6}{l_\Gamma} \right). \tag{5}$$

Here l_Γ determines the width of the amplifying medium in the direction perpendicular to the direction of propagation of the electric field pulse component x or y, Q_Γ is a phenomenologically introduced factor that depends on properties of the amplifying medium. Note, that the choice of the gain in the form of (5) is caused by the need to compensate the diffraction spreading of the pulse.

Let us write a standard equation for current density along the CNT axis [16]:

$$j = 2e \sum_{s=1}^{m} \int v(p, s) F(p, s) dp, \tag{6}$$

where e is the electron charge, the integration is carried out over the first Brillouin zone, p is component of the quasimomentum of the conduction electron along the axis of the nanotube, $v(p, s) = \partial \varepsilon(p, s)/\partial p$ is the electron velocity, $F(p, s)$ is the Fermi distribution function.

In [17] it is shown that due to field inhomogeneity, the accumulation of charge does not yield a significant contribution for extremely short pulses. Therefore, the cylindrical symmetry of the field distribution is preserved and we can neglect the derivative with respect to angle. In this case we obtain the following system of effective equations for components of the vector potential:

$$\begin{cases} \Box A_x + \dfrac{4en_0\gamma_0 a \cos\alpha}{c} \displaystyle\sum_{q=1}^{\infty} b_q \sin\left(\frac{aeq(A_x \cos\alpha + A_y \sin\alpha)}{c} \right) \\ \quad \times f(t) + \Gamma_x - F_1 \left(\dfrac{\partial A_x}{\partial t} \right)^{2n_p - 1} = 0, \\ \Box A_y + \dfrac{4en_0\gamma_0 a \cos\alpha}{c} \displaystyle\sum_{q=1}^{\infty} b_q \sin\left(\frac{aeq(A_x \cos\alpha + A_y \sin\alpha)}{c} \right) \\ \quad \times f(t) + \Gamma_y - F_1 \left(\dfrac{\partial A_y}{\partial t} \right)^{2n_p - 1} = 0, \end{cases} \tag{7}$$

where n_0 is the concentration of electrons,

$$b_q = \sum_s \frac{q}{\gamma_0} a_{sq} \int_{1Bz} dp' \cos(p'q) \frac{\exp\left(-\varepsilon(p', s)/k_B T\right)}{1 + \exp\left(-\varepsilon(p', s)/k_B T\right)}, \tag{8}$$

where k_B is the Boltzmann constant, T is the temperature, a_{sq} are coefficients in the expansion of the electron dispersion law (1) as a Fourier series.

Since coefficients defined by equation (8) decrease with growth of number q, we can take into account only the first 10 terms of sum in equations (7) [18].

Results of the numerical modelling

System of equations (7) was solved numerically with the following initial conditions

$$A_x = U \exp\left(-\left(\frac{z}{l_z} \right)^2 \right) \exp\left(-\frac{x^2 + y^2}{l_r^2} \right),$$

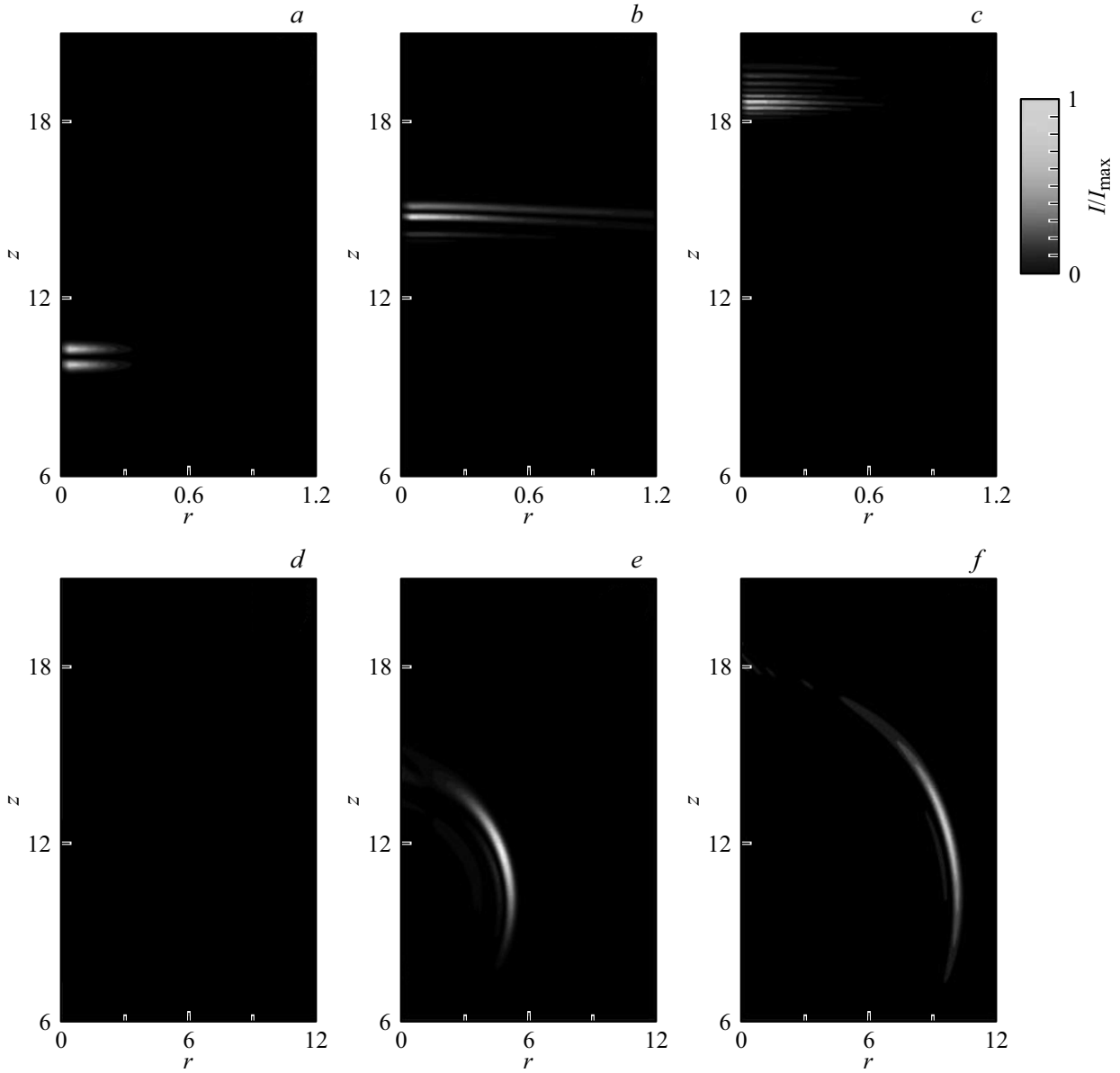

Figure 1. The intensity for x-component of the field (a–c) and for y-component (d–f) as function of coordinates: (a, d) $t = 0$; (b, e) $t = 5 \cdot 10^{-14}$ s; (c, f) $t = 10^{-13}$ s. $I_{\max}$ is the maximum intensity for each moment of time. Units along r and z axes correspond to $2 \cdot 10^{-5}$ m.

$$\frac{d}{dt} A_x = \frac{2 v_o z U}{l_z^2} \exp\left(-\left(\frac{z}{l_z}\right)^2\right) \exp\left(-\frac{x^2 + y^2}{l_r^2}\right),$$

$$A_y = 0, \quad \frac{d}{dt} A_y = 0, \tag{9}$$

where U is amplitude of the electromagnetic pulse at the inlet to the medium with CNTs, l_z, l_r is width of the pulse along corresponding directions.

Let us demonstrate graphs of the electromagnetic field evolution during its propagation over the specimen for the case of two-photon absorption in Fig. 1.

It can be seen from Fig. 1 that the E_x field component is broadened, however the broadening decreases with time and the pulse is localized in the direction of propagation, which is caused by the balance between amplification and attenuation of the pulse. The E_y component behaves as a radiation from the pulse defined at the initial moment of time, and in the course of propagation of this pulse it also moves in the initial transverse direction. This is caused by the absence of electric field component along the OY axis at the initial moment of time.

In contrast to the case of anisotropic medium without pulse amplification, when a substantial dispersion spreading of the pulse was observed, the situation is different in this case. The pulse becomes more localized despite the presence of the second component of the field polarization due to pumping in the system.

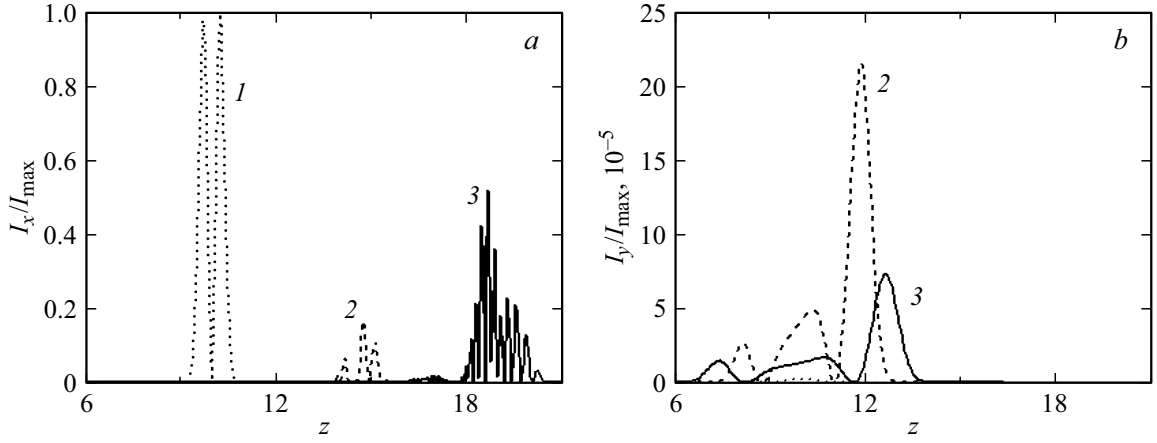

Figure 2. Longitudinal sections of intensity: (a) for the E_x component of electric field; (b) for E_y component as function of coordinate z: curve *1* — $t = 0$; curve *2* — $t = 5 \cdot 10^{-14}$ s; curve *3* — $t = 10^{-13}$ s. $I_{\max}$ is maximum intensity for three moments of time.

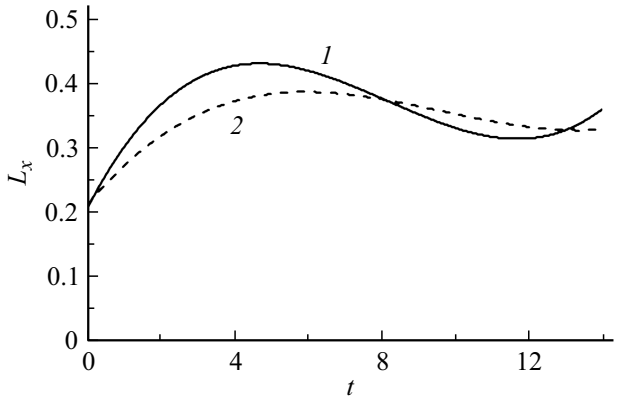

Figure 3. Pulse width as a function of time for photon number $n_p = 2$ (curve *1*) and $n_p = 3$ (curve *2*).

We evaluate each component of the electric field according to Fig. 2, where we show sections along the z axis through the maximum.

The following notes must be made. There is no curve *1* in Fig. 2, b because at the initial moment of time the field component $E_y = 0$. It can be seen that field intensity for the y component is by 5 orders of magnitude lower than the x component of the field. Its attenuation with time is observed (I_y).

Also, we investigated the dependence of pulse width (the distance where its intensity decreases by 2 times) on time (Fig. 3).

It was found that in the case of multiphoton absorption (for two and free photons) pulses are steadily propagated in terms of pulse width with time.

According to Fig. 1, c and Fig. 2, a (curve *3*) in the region of large times ($t = 10^{-13}$ s) a typical „bounce“is observed that corresponds to the mode of higher harmonics generation. Let us use the profile of Fourier spectrum of the pulse at a fixed moment of time (Fig. 4) to determine parameters at which the detected effect is manifested.

Note that in Fig. 4, b the dimensionless frequency w is plotted on the abscissa axis, i.e. a frequency divided by the characteristic angular frequency of CNT electron subsystem in the conductivity zone, so-called „plasma frequency“ ω_0 of electrons in CNT [14,19], which is similar to the „plasma frequency“ of electrons in supergratings. According to Fig. 4, the biggest effect is provided by the processes of two-photon absorption, causing the mode of higher harmonics generation. The cases of electron absorption and three-photon absorption are qualitatively coincided.

Conclusion

In conclusion, we present the main findings of this study.

1. The model of extremely short optical pulses propagation in an optically anisotropic crystal with carbon nanotubes is built taking into account the multiphoton absorption.

2. The substantial transverse dispersion of the pulse related to the presence of the second component of electric field vector can be suppressed by introducing pumping in the system. This allows localization of an extremely short optical pulse in a limited region of space.

3. The effect of higher harmonics generation in the optically anisotropic medium with CNTs with multiphoton absorption and amplification is identified. It is shown that this mode can be controlled by selecting parameters of the multiphoton absorption.

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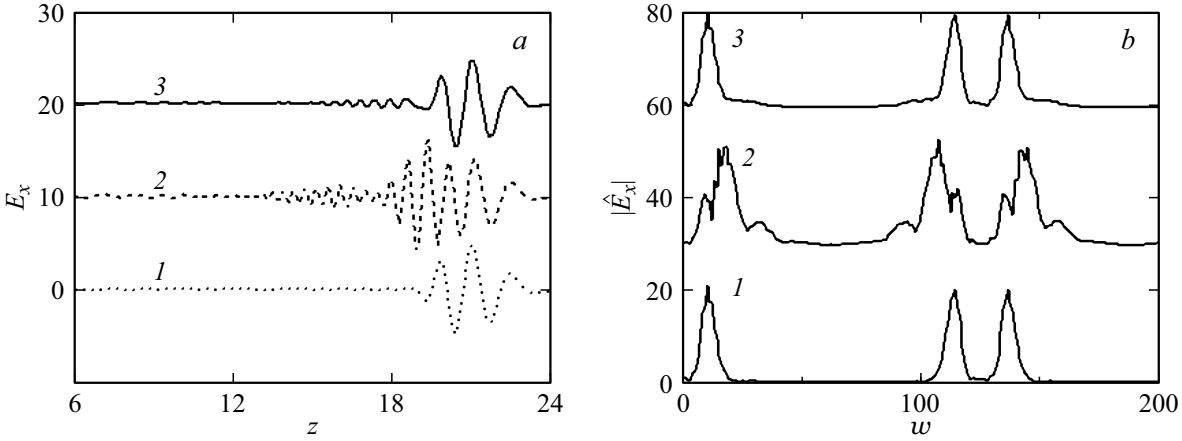

Figure 4. (*a*) Dependence of electric field strength of the pulse E_x on coordinate at the moment of time $t = 10^{-13}$ s for different number of photons *n*: curve *1* — normal nonlinear absorption; curve *2* — $n_p = 2$; curve *3* — $n_p = 3$. Figure (*b*) — Fourier spectra. Unit along E_x corresponds to 10^7 V/m, unit along w axis is $0.1\omega_0$. For illustrative purposes curves *2* and *3* in Figure (*a*) are shifted upwards by 10 and 20 units, respectively, in Figure (*b*) curves *2* and *3* are shifted upwards by 30 and 60 units, respectively.

Conflict of interest

The authors declare that they have no conflict of interest.

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