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Influence of an alternating electric field on the few-cycle pulses dynamics in a polymer composite with carbon nanotubes

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This paper presents a study of the dynamics of few-cycle optical pulses in a polymer-carbon nanotube composite medium exposed to an external alternating electric field. Numerical simulation of pulse evolution is performed using a modified nonlinear wave equation that takes into account the contributions of both the polymer matrix and the nanotubes to the electric current density. It is established that an external alternating field is an effective tool for controlling the amplitude, shape, and propagation velocity of pulses. The dependence of pulse dynamics on the amplitude and frequency of the external field, as well as the concentration and geometry of carbon nanotubes in the composite, is analyzed in detail. It is shown that varying these parameters allows one to change the pulse characteristics, opening up prospects for the creation of controllable optical elements and media.

Keywords: polymer composite, carbon nanotubes, few-cycle optical pulses, high-frequency electric field.

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Introduction

Today, photonics and optoelectronics place ever-higher demands on the data processing and transmission devices, which stimulates an active search for new nonlinear materials and methods for controlling light signals. Of particular interest in this context are extremely short optical pulses [1], the duration of which is comparable to several periods of oscillations of the electromagnetic field (usually 1−3 oscillations) [2,3]. Such pulses are not only a powerful tool for studying fundamental ultrafast processes [4], but also promising information carriers in next-generation communication systems, allowing record data transfer rates to be achieved [5].

One of the key challenges when working with extremely short pulses is the development of compact and efficient devices for controlling their parameters amplitude, phase, duration, and shape. Conventional approaches based on the nonlinear effects in bulk crystals or optical fibers often require significant powers or interaction lengths. In this regard, the researchers are focused on composite materials, the properties of which can be purposefully controlled. Among them, polymer composites [6,7] filled with carbon nanotubes (CNT) [8,9] are distinguished. Such systems combine the techical availability and flexibility of a polymer matrix with the unique nonlinear optical properties of CNT featuring strong nonlinear polarizability and ultrafast response over a wide spectral range. It is also worth noting that CNT are finding application in the development of key elements of optoelectronic devices, including integrated circuits [10], photonic emitters [11], resonators [12], saturable absorbers in mode-locked lasers [13] and optical limiters [14].

The key benefit of the composite structure is the ability to purposefully control the spatial arrangement of CNT tubes — their geometry, orientation, and packing density. Control over these parameters can be carried out both at the stage of material synthesis and after it. One of the most promising pre-treatment methods is a method of floating catalyst (FCCVD) chemical vapor-phase deposition (CVD) [15,16], in which, unlike conventional CVD, the catalyst is introduced in gaseous or aerosol form into the reaction chamber. Among the post-treatment methods, dry spinning [17,18], wet spinning [19,20], combined spinning [21], chemical cross-linking [22] can be distinguished, which are used to achieve uniaxial alignment and packing of CNT. It is this controlled nanostructure architecture that enables to obtain hybrid materials with excellent functionality, including increased mechanical strength, high electrical conductivity, improved thermal stability, and optimal rheological properties.

An important direction in controlling the optical properties of materials is the use of external electric fields. This approach provides a dynamic control of electromagnetic wave propagation, which is a key factor in fabrication of modern optical devices. For example, the study [23] demonstrated the possibility of using a CNT array with artificially created inhomogeneity for engineering the soliton valves, the permeability of which is controlled by an external electric field which is a perspective sector in terms of development of controlled optical gates and modulators. It is also shown that by changing the amplitude of the external pumping, it is possible to control the shape of an extremely

short optical pulse and stabilize it [24]. This is especially valuable for the tasks of data transmission without distortion and controlling extremely short pulses in nonlinear media.

Unlike a permanent field, which can cause irreversible processes, an alternating electric field is a more flexible tool. It makes it possible to dynamically modulate the electronic structure and, as a result, the nonlinear optical responses of nanofillers without accumulating spatial charge. However, the mechanisms of impact of an alternating electric field on the propagation and transformation of extremely short pulses in such complex dielectric media as polymer composites with CNT have not been fully studied so far. Thus makes this study especially relevant.

This paper gives an insight on theoretical and numerical analysis of the effect imposed by the alternating electric field on the dynamics of extremely short IR pulses in a polymer composite with semiconductor CNT. Unlike previous studies of dynamics in similar systems [25–27], where the effects of an external field were neglected, the present study is aimed at analyzing new effects caused by exposure to a high-frequency electric field.

1. Model and basic equations

The passage of an extremely short optical pulse through a composite medium is modeled under the following conditions. The array of vertically aligned CNT in the polymer is oriented so that the wave vector of the laser pulse is perpendicular to it and directed along Oz axis (Fig. 1). The distance between CNT is considered a way larger than their radius. The electric field strength of $E = (E(y, z, t), 0, 0)$ pulse is oriented along Ox axis, which causes an electric current with a surface density of $j = (j(y, z, t), 0, 0)$ in the CNT. Such an array orientation can be obtained, for example, using a polymer matrix. Another option is catalytic chemical vapor-phase deposition (CCVD), in which nanoparticles of iron, cobalt or nickel [28,29] are usually used as catalysts. In addition to the classical approach, the improved CCVD technique, first proposed in 2004 by a scientific team led by K. Khaty [30], is of great interest. This method, often referred to as „stimulated by steam“, has proven to be extremely effective in the synthesis of high-quality single-layer CNT. The key difference from CCVD is the controlled introduction of a small amount of water vapor into the reaction chamber along with the main carbon-containing precursor gases. This allows to deal with rapid deactivation of the catalyst, while maintaining the catalytic activity of the nanoparticles for several minutes rather than seconds compared to a simple CCVD. Due to its efficiency and simplicity, this method has become one of the standard and widely reproducible approaches in CNT synthesis for the basic research and advanced technological applications.

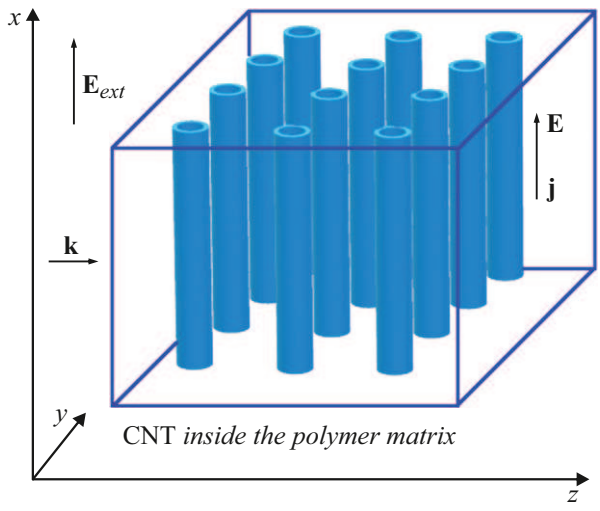

Figure 1. Schematic diagram of the system geometry: CNT are placed in the polymer matrix.

The wave equation for x-component of the vector potential $A = (A(y, z, t), 0, 0)$, taking into account calibration: $cE = -\partial A/\partial t$ can be written as follows:

$$\frac{\varepsilon}{c}\frac{\partial^2 A}{\partial t^2} = \frac{\partial^2 A}{\partial y^2} + \frac{\partial^2 A}{\partial z^2} + \frac{4\pi}{c} j(A), \tag{1}$$

where ε — permittivity of a medium, the density of electric current is composed of two constituents — j_{CNT} (CNT contribution) and j_{pol} (polymer contribution). Note that a simple model is used here to determine the effective permittivity, which is based on Maxwell-Garnett approximation [31] and provides only an estimate ε.

To find the current density j_{CNT}, the kinetic Boltzmann equation is used, from which, taking into account the substitution of $p \to p - |e|A/c$ and the collisionless approximation (the pulse duration is much shorter than the electron relaxation time), the following formula is obtained [32]:

$$j_{\text{CNT}} = -\frac{|e|N}{\pi\hbar}\sum_{s=1}^{m}\int_{\text{BZ}} dp \cdot v\left(p - \frac{|e|A}{c}, s\right) f(\varepsilon_s(p) - \mu), \tag{2}$$

where e — electron charge, p — electron quasi-pulse, N — nanotube placement density, $f(p, s, \mu)$ — Fermi-Dirac distribution function, μ — chemical potential, BZ — boundaries of the first Brillouin zone, $v(p, s) = \partial\varepsilon_s(p, s)/\partial p$ — electron velocity in CNT, the law of electron dispersion $\varepsilon_s(p, s)$ of zig-zag CNT $(m, 0)$ has the form [33]:

$$\varepsilon_s = \gamma_0\sqrt{1 + 4\cos(a\,p)\cos\left(\frac{\pi s}{m}\right) + 4\cos^2\left(\frac{\pi s}{m}\right)}, \tag{3}$$

$s = 1, 2, \ldots, m$, $\gamma_0 = 2.7\,\text{eV}$ — hopping integral, $a = 3b/(2\hbar)$, b — distance between the neighbouring carbon atoms in CNT.

Further, expanding the law of dispersion (3) in the Fourier series, we obtain the following expressions for the density of electric current in nanotubes:

$$j_{\text{CNT}} = -\frac{|e|N\gamma_0}{\pi\hbar}\sum_{r=1}^{\infty} rG_r \sin\left(\frac{r|e|aA}{c}\right),$$

$$G_r = -\sum_{s=1}^{m} \frac{B_{r,s}}{\gamma_0} \int_{-\pi}^{\pi} \frac{\cos\left(\frac{ra\tilde{p}}{c}\right)}{1+\exp\left(\sum\limits_{r=1}^{\infty} B_{r,s}\cos\left(\frac{ra\tilde{p}}{c}\right)-\mu\right)} d(a\tilde{p}), \tag{4}$$

where $B_{q,s}$ — expansion coefficients (3).

Note that in zig-zag CNT, which have a number m that is not a multiple of three, there is a slit in the energy spectrum. Moreover, the larger the diameter of the CNT, the smaller the band gap (E_g). Thus, for instance, for the CNT of type (7,0) $E_g = 1.33$ eV. For pulses with a wavelength of 1.5 μm, the photon energy will be approximately 0.83 eV, i.e. it is not enough for the electrons to cross the slit and appear in the conduction band. Therefore, we use electrostatic doping [34–36], which will lead to a shift in the Fermi level closer to the conduction band and corresponds to the nonzero chemical potential μ (formula (4)), which determines the concentration of electrons in the CNT conduction band $n(T,\mu)$:

$$n(T,\mu) = \sum_{s=1}^{m} \int_{\mathrm{BZ}} \frac{N}{\pi\hbar} f(\varepsilon_s(p)-\mu)dp. \tag{5}$$

It should be underscored that electrostatic doping is one of the methods that make it possible to control the concentration of charge carriers without physically introducing foreign atoms (as in the classical doping of [37–39]), but by applying an external electric field. The principle of operation is easy to understand from the device of a field effect transistor [40,41]. In this case, the type and concentration of charge carriers in the material are dynamically controlled by the magnitude and sign of the applied voltage. There are other ways that help to tackle the problem of low concentration of electrons in the conduction band. These include: thermal excitation — heating of the material increases the thermal energy of electrons in the valence band, which becomes sufficient to overcome the band gap [42], carrier injection [43], multiphoton absorption during high-intensity laser radiation, when an electron simultaneously absorbs several photons, the total energy of which is higher or equal to the band gap [44,45].

The density of current in polymer j_{pol} was calculated similar to the calculation of component j_{CNT}. We selected polyacetylene in the trans-$(CH)_x$ configuration as the polymer. The law of electron dispersion in polymer [46] can be written as follows:

$$\varepsilon(q) = -t_0\cos(qa_0),$$
$$qa_0 = \frac{2\pi n}{N_0}\left(n = 0, \pm1, \ldots, \pm\frac{N_0}{2}\right), \tag{6}$$

where N_0 — number of atoms in the lattice, a_0 — coupling length between the carbon atoms in polymer (0.14 nm), t_0 — overlap integral in the electron clouds in polymer (2.5 eV). Finally, the expression for the current density in the polymer takes the form:

$$j_{pol} = -\frac{|e|}{\pi\hbar} R\cdot t_0\cdot\chi\cdot\sin\left(\frac{a_0|e|A}{c}\right),$$

$$R = \int_{-\pi}^{\pi} \frac{\cos(a_0q)}{1+\exp(\varepsilon(q)/k_BT)}\, d(a_0q), \tag{7}$$

where χ determines the concentration of electrons in a polymer, k_B —Boltzmann constant, T — temperature.

2. Effect of external alternating electric

Taking into account the effect of external alternating electric results in chaning of the vector potential which is expressed as the following substitution in the formula for the current density: $A \to A + A_1$. Since $E_{ext} = (E_1, 0, 0)$ and $cE_1 = -\partial A_1/\partial t$,

$$A \to A - E_{ext}\frac{c}{\omega_{ext}}\sin(\omega_{ext}t+\alpha). \tag{8}$$

Here E_1, ω_{ext} and α — amplitude, frequency and phase of external electric field whereas $E_1 = E_{ext}\cos(\omega_{ext}t)$.

Next, we substitute the expression (8) in (4) and apply the formula „sine of the difference“:

$$j_{\mathrm{CNT}} = -\frac{|e|N\gamma_0}{\pi\hbar}\sum_{r=1}^{\infty} rG_r \times\left\{\sin\left(\frac{r|e|a}{c}A\right)\cos\left(\frac{r|e|aE_{ext}}{\omega_{ext}}\sin(\omega_1t+\alpha)\right) - \cos\left(\frac{r|e|a}{c}A\right)\sin\left(\frac{r|e|aE_{ext}}{\omega_{ext}}\sin(\omega_1t+\alpha)\right)\right\}. \tag{9}$$

Given the known formulae [47]:

$$\cos(a\cdot\sin b) = J_0(a) + 2\sum_{k=1}^{\infty} J_{2k}(a)\cos(2kb),$$
$$\sin(a\cdot\sin b) = 2\sum_{k=1}^{\infty} J_{2k-1}(a)\cos((2k-1)b), \tag{10}$$

where J_k is the Bessel function of k-th order, we average over the period of the high-frequency field and finally obtain the following expression for the current density in CNT:

$$j_{\mathrm{CNT}} = -\frac{|e|N\gamma_0}{\pi\hbar}\sum_{r=1}^{\infty} rG_r\sin\left(\frac{r|e|a}{c}A\right)J_0\left(\frac{r|e|aE_{ext}}{\omega_{ext}}\right). \tag{11}$$

Acting in a similar way, we obtain an expression for the current density in the polymer, taking into account the external alternating field:

$$j_{pol} = -\frac{|e|Rt_0\chi}{\pi\hbar}\sin\left(\frac{a_0|e|A}{c}\right)J_0\left(\frac{|e|aE_{ext}}{\omega_{ext}}\right). \tag{12}$$

Note that an external high-frequency field generates a longitudinal surface current and a surface wave for the longitudinal Hertz vector. In this case, it is also necessary to consider the Leontovich-Levin equation, which describes

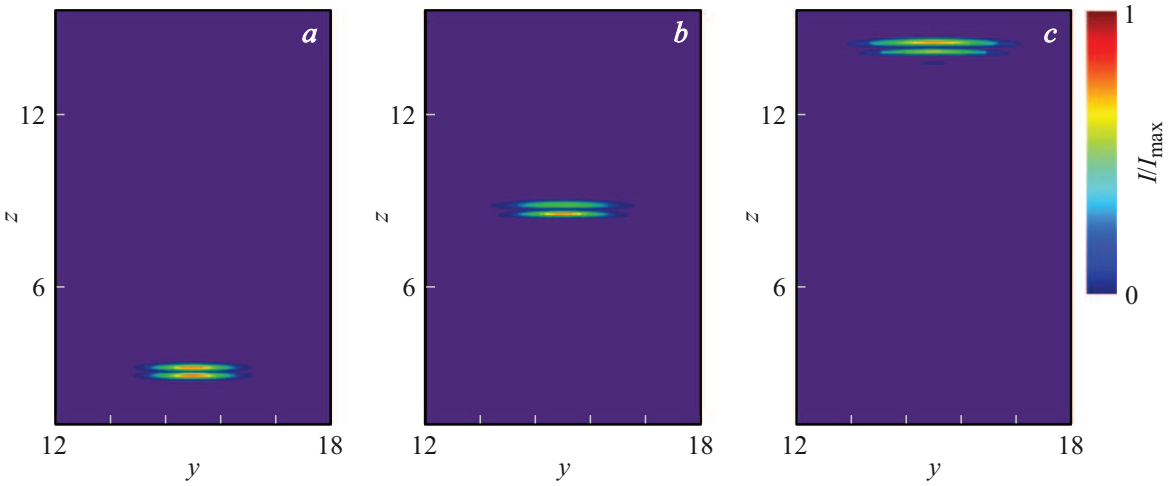

Figure 2. The intensity of the pulse electric field in the absence of an external electric field at various points in time: a — $t = 0$; b — $t = 1.4 \cdot 10^{-14}$ s; c — $t = 2.8 \cdot 10^{-13}$ s. The coordinate scale units correspond to $4 \cdot 10^{-6}$ m. The color scale is given for the intensity assigned to the maximum intensity shown in all figures (a–c).

the behavior of the field on the surface of an object and allows to detect unusual resonances and the wave deceleration effect. As seen in [48], meeting the condition: $L/d < 0.1$ (d — distance between the nearest CNT, L — nanotube length) provides that there's no any adverse effect of resonances on the pulse ($L < 200\,\text{nm} \cdot 0.1$). In this paper, the CNT array ($L < 20\,\text{nm}$) is considered as a homogeneous layer with some effective thickness and dielectric permittivity. Going beyond this approximation and taking into account the Leontovich-Levin equation was not the purpose of this work and will be carried out by the authors in the future.

3. Results of numerical modeling

The equation (1), taking into account (11) and (12), after being reduced to a dimensionless form, was solved using the finite difference scheme [49] using standard stability conditions. Since the equation for the vector potential has the form equivalent to the sine-Gordon equation, it is necessary to fulfill the Courant–Friedrichs–Lewy condition. Moreover, the steps of the difference scheme were reduced sequentially by half until the solution changed in the fifth decimal place. The initial condition for the vector potential of an extremely short pulse was set using the Gauss function:

$$A(y, z, 0) = A_0 \cdot \exp\left(-\frac{z^2}{\gamma_z^2}\right) \exp\left(-\frac{y^2}{\gamma_y^2}\right),$$

$$\frac{dA(y, z, 0)}{dt} = \frac{2A_0 \cdot z \cdot v_z}{\gamma_z^2} \cdot \exp\left(-\frac{z^2}{\gamma_z^2}\right) \exp\left(-\frac{y^2}{\gamma_y^2}\right), \quad (13)$$

where A_0 — the initial amplitude of the pulse, $v_z = 0.95$ — its velocity at the moment of time $t = 0$ (in units of the speed of light), γ_x, γ_z — determine the initial width of the extremely short pulse along the specified directions y and z.

Computational modeling was made given the following parameters: CNT of type (7,0), plymer composite had the following dimensions: $1 \times 1 \times 1$ cm, $z = 0$ corresponds to its location (beginning). According to our estimates, the electrons concentration in CNT is equal $5.5 \cdot 10^{14}\,\text{cm}^{-3}$ for $\mu = 0.65$ eV and temperature $T = 77$ K (found by the formula (5) for the CNT array (7,0) with a diameter of $d_{\text{CNT}} = 0.55$ nm, nanotubes spacing was $d \approx 200$ nm, CNT array on an area of 1×1 cm, which corresponds to the package density $N = 2.5 \cdot 10^9$ per square centimeter). Note that the transition across the boundary is not considered, since we believe that at $t = 0$ the pulse is inside the sample. The electric field strength of the pulse at $t = 0$ (maximum value) is $5 \cdot 10^7$ V/m.

The intensity of the electric field of an extremely short pulse is determined by the formula

$$I \propto E^2. \quad (14)$$

Figure 2 shows the evolution of the intensity of a two-dimensional extremely short optical pulse during its passage through a polymer composite with CNT in the absence of an external high-frequency field.

The pulse propagates localized, which is due to the balance of two competing processes — dispersion and nonlinearity of the medium. While it tends to broaden with time.

Figure 3 shows an extremely short optical pulse depending on the amplitude of an external alternating field.

According to Fig. 3, it can be concluded that application of an external alternating electric field leads to a decrease in intensity and occurrence of additional peaks following the main pulse. The observed phenomena are a direct consequence of changes in electrodynamic properties of a composite exposed to the action of external alternating electric field. The external field changes the dielectric

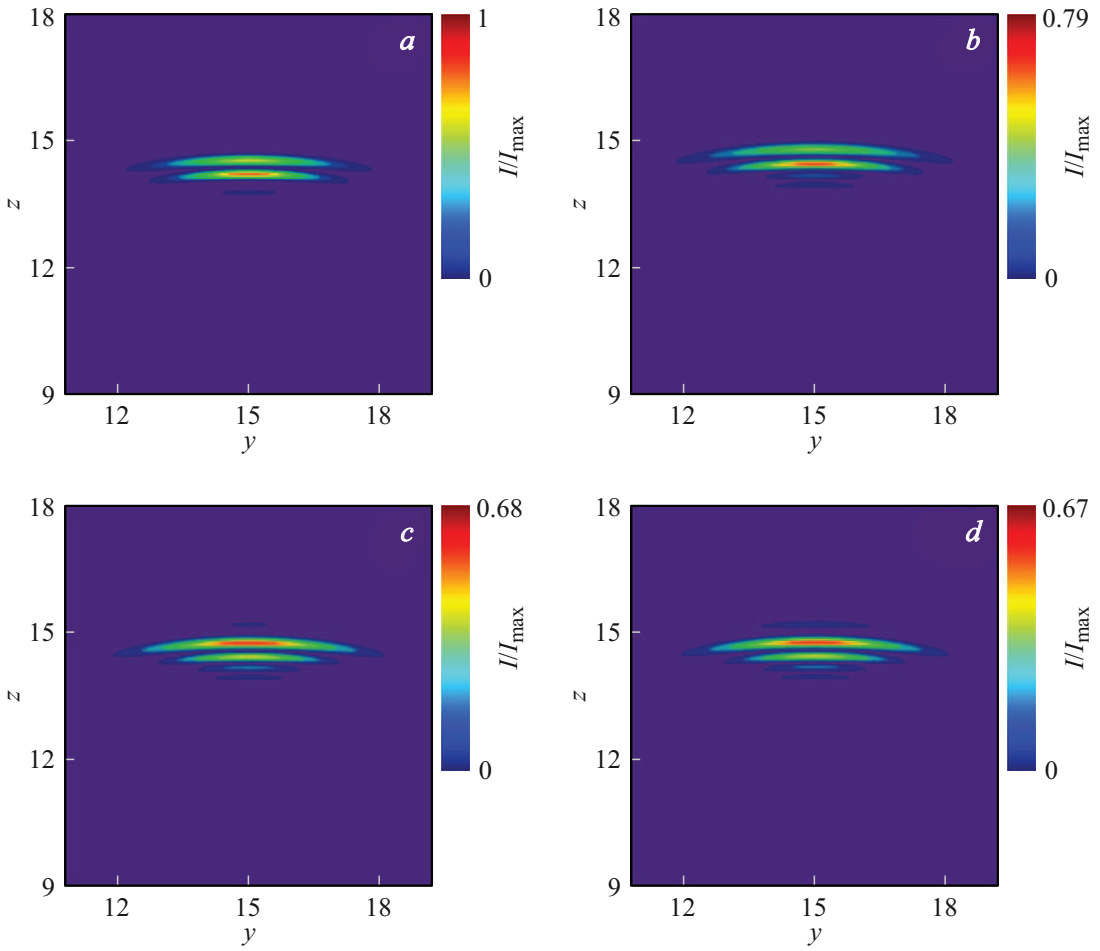

Figure 3. Electromagnetic pulse intensity at a moment of time $t = 2.8 \cdot 10^{-13}$ s for different values of the amplitude of the high-frequency field $E_{ext}(\omega_{ext} = 10^{14}\,\mathrm{s}^{-1})$: a — $E_{ext} = 0$; b — $E_{ext} = 1.5 \cdot 10^8$ V/m; c — $E_{ext} = 3 \cdot 10^8$ V/m; d — $E_{ext} = 6 \cdot 10^8$ V/m. The coordinate scale units correspond to $4 \cdot 10^{-6}$ m. The color scale displays the intensity brought to the maximum of all the figures (a–d).

Width of an extremely short pulse

Value of eaE_{ext}/ω_{ext}	Width of extremely short pulse pulse, $4 \cdot 10^{-6}$ m
0	1.08
0.5	1.14
1	1.32
2	1.35
4	1.5

permittivity of the medium. The key mechanism of this change is the acceleration of free charge carriers, which interact more effectively with passing radiation, which leads to increased energy dissipation on them. In other words, a combination of amplification of dissipative processes and activation of nonlinear optical phenomena in the composite under the action of a controlling electric field is observed.

It is worth noting that the pulse behavior is influenced by a value proportional to the ratio of the amplitude of external alternating field E_{ext} to its frequency ω_{ext}. The dependence of the extremely short pulse on the value of eaE_{ext}/ω_{ext} is shown in the table. The width of the pulse was taken to be the distance from its center, at which the maximum intensity is halved. At $eaE_{ext}/\omega_{ext} = 4.0$, the diffraction blurring of the pulse is significantly more pronounced than at $eaE_{ext}/\omega_{ext} = 0$. This is because the controlling alternating field causes dynamic changes in the electronic subsystem of the medium, which, in turn, weakens its self-focusing properties and leads to a greater blurring of the pulse.

The effect of polymer concentration on the parameters of an extremely short pulse is shown in Figs. 4 and 5.

Spatial modulation of the refractive index caused by the dependence of the dielectric properties on the polymer concentration (parameter χ) leads to a redistribution of the pulse energy in the longitudinal and transverse directions. Nevertheless, this does not entail a qualitative change in the dynamics of the pulse (Fig. 4). Figure 5 shows a significant

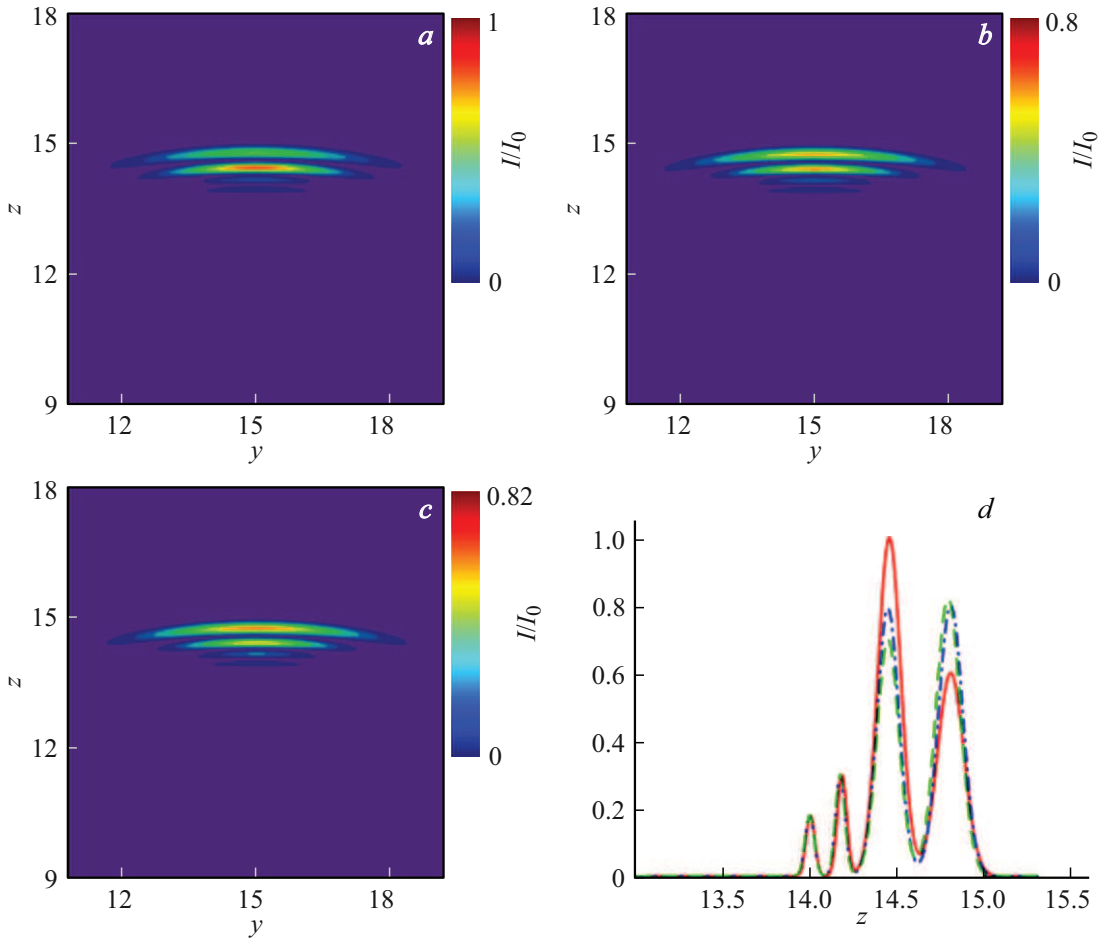

Figure 4. Intensity of the electromagnetic pulse at time $t = 2.8 \cdot 10^{-13}$ s at $eaE_{ext}/\omega_{ext} = 1$ for different polymer concentrations: a — $\chi = 0$; b — $\chi = 0.5$; c — $\chi = 1.0$; d — sections at $y = 15$: red line — for the figure (a), blue line — for the figure (b), green line — for the figure (c). The coordinate scale units correspond to $4 \cdot 10^{-6}$ m. The color scale displays the intensity adjusted to the maximum intensity for the value $\chi = 0$.

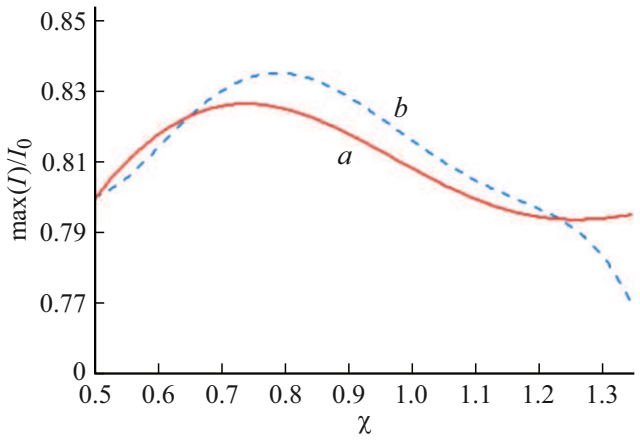

Figure 5. Maximum intensity of the electromagnetic pulse at the moment of time $t = 2.8 \cdot 10^{-13}$ s depending on the polymer concentration: a — at $eaE_{ext}/\omega_{ext} = 1$; b — $eaE_{ext}/\omega_{ext} = 2$. I_0 corresponds to the maximum of intensity at $\chi = 0$.

dependence of the intensity of the electromagnetic pulse on the polymer concentration in the composite. Thus, minimal relative deviation of $\max(I)$ makes 17 %.

Conclusion

1. A model has been developed describing the propagation of an extremely short pulse in a nonlinear medium with a polymer composite containing CNT, taking into account the modulation of its electro-optical properties by an external alternating electric field.

2. By selecting the amplitude and frequency of the external field, it is possible to control the parameters of the electromagnetic pulse (intensity and width) by changing the nonlinear response of the composite material.

3. The effect of the external alternating field leads to the occurrence of additional pulse peaks, which is important for generating higher harmonics, minimizing damage to materials, and controlling nonlinear optical processes.

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Conflict of interest

The author declares that he has no conflict of interest.

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