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ULTRASHORT PULSES IN PHOTONIC CRYSTALS MADE OF CARBON NANOTUBES WITH NONLINEAR ABSORPTION

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Abstract. In this paper, we consider the problem of the propagation of three-dimensional ultrashort optical pulses in a photonic crystal made of zig-zag carbon nanotubes under the condition of nonlinear absorption and amplification. The nonlinear absorption is described phenomenologically using data from real experiments. The possibility of stable optical pulse propagation under the conditions of external pump and damping field is shown. We analyze the dependence of the pulse shape on various parameters of the medium, such as the period of carbon nanotubes inhomogeneity in a photonic crystal and the modulation depth of the refractive index.

Key words: ultrashort pulses, carbon nanotubes, photonic crystals, nonlinear absorption.

1. INTRODUCTION

One of the promising tasks of modern nonlinear and coherent optics, both from a theoretical and practical point of view, is the study of interaction of electromagnetic radiation with optical media. Works on the creation of basic devices for optoelectronics, nanoelectronics, and nanophotonics, using materials with desired properties, happen to be of great interest to many researchers. Photonic crystals can be successfully used to create various nanophotonic devices for information processing. A photonic crystal is an optical medium with a spatially periodic refractive index in which a photonic band gap exists. This photonic band gap allows us to consider a photonic crystal as an optical filter that transmits photons with a certain frequency [1, 2]. It is also worth noting that the inhomogeneities in photonic crystals provide an ideal nonlinear medium for the study of electromagnetic solitons, the propagation of ultrashort (few-cycle) optical pulses, and the formation of spatiotemporal optical solitons (*alias* “light bullets”) [3].

In this paper, we consider ultrashort optical pulses, which are femtosecond electric field pulses localized in space with duration of several periods of field oscillation. All the energy of these pulses is concentrated in a finite limited region

of space [4, 5]. Interest in ultrashort pulses is due to the high directivity of their radiation, the stability of their shape, and the resistance to perturbations of the involved parameters. Also, it should be noted the currently achievable high values of the peak intensity of the pulse field, when there is no destruction of the waveguide material, but its nonlinear properties already appear and should be taken into account [6–18].

For the stable propagation of ultrashort pulses, it is necessary that the optical medium possesses nonlinear properties. Carbon nanotubes (CNTs) with unique nonlinear properties in the optical range can be used as such media. Their dispersion law for electrons is non-parabolic and it determines the nonlinear response of nanotubes to moderate electromagnetic stresses – 10^3–10^4 V/cm [19, 20]. However, there exist dissipative effects in CNTs. Therefore, the compensation of energy ("pumping") is necessary. Thus, it becomes relevant to consider dissipative effects, which occur in real devices. Consequently, taking into account all parameters stabilizing and destroying the ultrashort optical pulse at large times is quite nontrivial.

However, it should be noted the prerequisites for the work reported here. Namely, the possibility of the stable propagation of either two-dimensional or three-dimensional ultrashort optical pulses in media with oriented CNTs with a spatially variable refractive index has been reported in Refs. [21–34]. The regularities of the pulse dynamics in media with a varying refractive index under the action of external fields are also revealed [35, 36]. The dependence of the optical pulse evolution on the medium parameters (period and depth of modulation of the refractive index) is shown. Also there is a question about taking into account the real relaxation time obtained from experiments [37, 38]. The experimentally observed relaxation time can be divided into two parts: the linear (in pulse amplitude) and the nonlinear one. The linear component was described in Refs. [39, 40]. The question of taking into account the nonlinear part, namely, the correct numerical specification of nonlinear absorption, has not been addressed in the literature, to the best of our knowledge.

It is the aim of this work to study the problem of the propagation of three-dimensional ultrashort optical pulses in a photonic crystal made of zig-zag CNTs under the condition of nonlinear absorption and amplification. This paper is organized as follows. In Section 2 we present the geometry of the problem under study and the basic equations for the vector potential of the electromagnetic field that are solved numerically. The main results of this study are given in Section 3. Finally, in Section 4 we present our conclusions.

2. BASIC EQUATIONS

The study of the electronic structure of CNTs, as a rule, is carried out in the tight-binding approximation as part of an analysis of the π-electrons dynamics. The

general dispersion relation for the structure of semiconductor-type nanotubes is the following [41, 42]:

$$\varepsilon_s(p) = \pm\gamma_0\sqrt{1+4\cos(ap)\cos(\pi s/m)+4\cos^2(\pi s/m)}, \quad (1)$$

where $\gamma_0 \approx 2.7$ eV, $s = 1, 2\ldots, m$, $a = 3b / 2\hbar$, $b = 0.142$ nm is the distance between the adjacent carbon atoms, and p is the component of the momentum along the CNT axis. We note that m stands for the hexagons number along the nanotube circumference.

The geometry of the problem (Fig. 1) assumes that the current, the applied electric field, and the pulse electric field are directed along the CNT axis, and the pulse moves perpendicular to the array of nanotubes. Since the typical size of CNTs and the distance between them are much smaller than the typical size of the spatial region in which the ultrashort pulse is localized, we can use the continuous medium approximation and consider the current distributed throughout the volume.

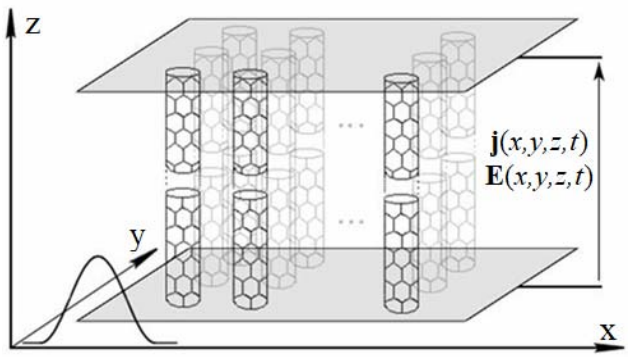

Fig. 1 – The problem's geometry.

The vector potential has the form: $\mathbf{A} = (0, 0, A(x, y, z, t))$, the current density is $\mathbf{j} = (0, 0, j(x, y, z, t))$.

Since the typical relaxation time for electrons in CNTs can be estimated as $3 \cdot 10^{-13}$ s [43], then the electron ensemble at 10^{-14} s (typical for ultrashort optical pulse duration) can be described by the Boltzmann collisionless kinetic equation [44]:

$$\frac{\partial f}{\partial t} - \frac{q}{c}\frac{\partial A_z}{\partial t}\frac{\partial f}{\partial p} = 0, \quad (2)$$

where $f = f(p, s, t)$ is the distribution function implicitly dependent on the coordinate (due to the dependence of the vector potential $\boldsymbol{A}$ on the coordinate). Moreover, the distribution function f at the initial instant of time coincides with the function F_0 – the Fermi equilibrium distribution function.

To calculate the current density, we use the standard expression [45]:

$$j = 2e\sum_{s=1}^{m}\int_{ZB} v_s(p)\cdot f(p,s)\mathrm{d}p\,, \tag{3}$$

where the group electron velocity is introduced: $v_s(p) = \partial\varepsilon_s(p) / \partial p$, $\varepsilon_s(p)$ is the dispersion law describing the electronic properties of CNTs (formula (1)), and e is the electron charge. The integration is carried out in the first Brillouin zone.

The equation for the vector potential of the electromagnetic field with taking into account the Coulomb calibration ($\mathbf{E} = -\partial\mathbf{A}/c\partial t$) has the following form:

$$\frac{\partial^2 \boldsymbol{A}}{\partial x^2}+\frac{\partial^2 \boldsymbol{A}}{\partial y^2}+\frac{\partial^2 \boldsymbol{A}}{\partial z^2}-\frac{n^2(x,y,z)}{c^2}\frac{\partial^2 \boldsymbol{A}}{\partial t^2}+\Gamma\frac{\partial \boldsymbol{A}}{\partial t}+\frac{4\pi}{c}\boldsymbol{j}- \\ -F_1\left(\frac{\partial \boldsymbol{A}}{\partial t}\right)^3-\frac{F_2\dfrac{\partial \boldsymbol{A}}{\partial t}}{1+\rho\left(\dfrac{\partial \boldsymbol{A}}{\partial t}\right)^2}=0, \tag{4}$$

where $n(x,y,z)$ is the spatially modulated refractive index, Γ is the electric field pump rate (excluding linear absorption) [46], F_1 and F_2 are the nonlinear absorption coefficients [47], the forms and values of which are taken from experimental works.

Here we assume that $\partial/\partial\varphi \to 0$ due to cylindrical symmetry. The inhomogeneity of the field along a certain axis leads to the inhomogeneity of the current. As a result, charge accumulates in some region. Earlier calculations [44] showed that the charge accumulation can be neglected, thus we can assume that cylindrical symmetry in the field distribution is preserved. Thus, the final equation for the vector potential in a cylindrical coordinate system takes the form:

$$\frac{\partial^2 \boldsymbol{A}}{\partial z^2}+\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial \boldsymbol{A}}{\partial r}\right)-\frac{n^2(z,r)}{c^2}\frac{\partial^2 \boldsymbol{A}}{\partial t^2}+\Gamma\frac{\partial \boldsymbol{A}}{\partial t}+\frac{4en_0}{c}\sum_{q=1}^{\infty}b_q\cos\left(\frac{aeqA}{c}\right)\cdot \\ \cdot\frac{aeq}{c}f(t)-F_1\left(\frac{\partial \boldsymbol{A}}{\partial t}\right)^3-\frac{F_2\dfrac{\partial \boldsymbol{A}}{\partial t}}{1+\rho\left(\dfrac{\partial \boldsymbol{A}}{\partial t}\right)^2}=0,\quad r=\sqrt{x^2+y^2} \tag{5}$$

The term, which depends on the rotation angle, can be neglected [48], n_0 is the electron concentration,

$$f(t)=\begin{cases}0, & t<t_0(z)\\ \exp(-t/t_{rel}), & t\ge t_0(z)\end{cases},$$

$t_0(z) \cong (z - z_0)/v$ is the time at which the pulse intensity at its leading edge, measured at the point with coordinate z, is e times less than the peak pulse intensity; z_0 is the initial coordinate of the "mass center" of the pulse at $t = 0$, $v \cong c/\sqrt{k_0}$ is the approximate pulse velocity, k_0 is the average relative dielectric constant of the medium (CNTs array), and t_{rel} is the relaxation time of CNTs subsystem [48]. The term $f(t)$ is the empirical correction for the current density in the collisionless approximation. The current arising in a photonic crystal during the ultrashort pulse propagation decays exponentially, taking into account the relaxation processes.

The coefficients b_q in Eq. (5) have the following form:

$$b_q = \sum_s a_{sq} \int_{ZB} \mathrm{d}p \cdot \cos(pq) \frac{\exp\left(-\varepsilon_s(p)/k_b T\right)}{1 + \exp\left(-\varepsilon_s(p)/k_b T\right)}, \tag{6}$$

where a_{sq} are the coefficients in the expansion of the electron dispersion law (1) as a Fourier series:

$$\varepsilon_s(p) = \frac{1}{2\pi} \sum_{s=1}^{m} \sum_{q=1}^{\infty} a_{sq} \cos(\mathrm{pq}), \tag{7}$$

$$a_{sq} = \int_{ZB} \mathrm{d}p \cdot \cos(pq) \varepsilon_s(p). \tag{8}$$

Due to a decrease in the coefficients b_q (6) with an increase in q in the sum, we can restrict ourselves to the first 15 nonvanishing terms [45] and obtain the generalized sine-Gordon equation [33].

3. MAIN RESULTS

The initial conditions for the vector potential correspond to a Gaussian pulse profile for a single oscillation of the electric field (see Eq. (9) bellow) and the refractive index of the photonic crystal made of CNTs (10) is specified by Eq. (10) below:

$$\begin{aligned} A_{t=0} &= A_0 \cdot \exp\left(\frac{r^2}{\gamma^2}\right) \exp\left(-\frac{(z - z_0{}^2)}{\beta^2}\right), \\ \left.\frac{\mathrm{d}A}{\mathrm{d}t}\right|_{t=0} &= \frac{2 \cdot v \cdot A_0 (z - z_0)}{\gamma^2} \exp\left(-\frac{r^2}{\gamma^2}\right) \exp\left(-\frac{(z - z_0)^2}{\beta^2}\right) \end{aligned} \tag{9}$$

$$n(z, r) = n_0 (1 + \alpha \cdot \cos(2\pi z / \chi)). \tag{10}$$

Here β and γ are the parameters determining the pulse width along the z and r axes, respectively, t_0 is the initial time point, v is the initial pulse velocity upon

entry into the medium, α is the refractive index modulation depth, and χ is the refractive index modulation period.

The equation (5) is solved numerically using an explicit difference scheme of the "cross" type [49].

The evolution of the electromagnetic field pulse during its propagation in the CNT medium with a spatially variable refractive index (that is, in a photonic crystal) and with nonlinear absorption and amplification, in the case of a single oscillating electric field, is shown in Fig. 2.

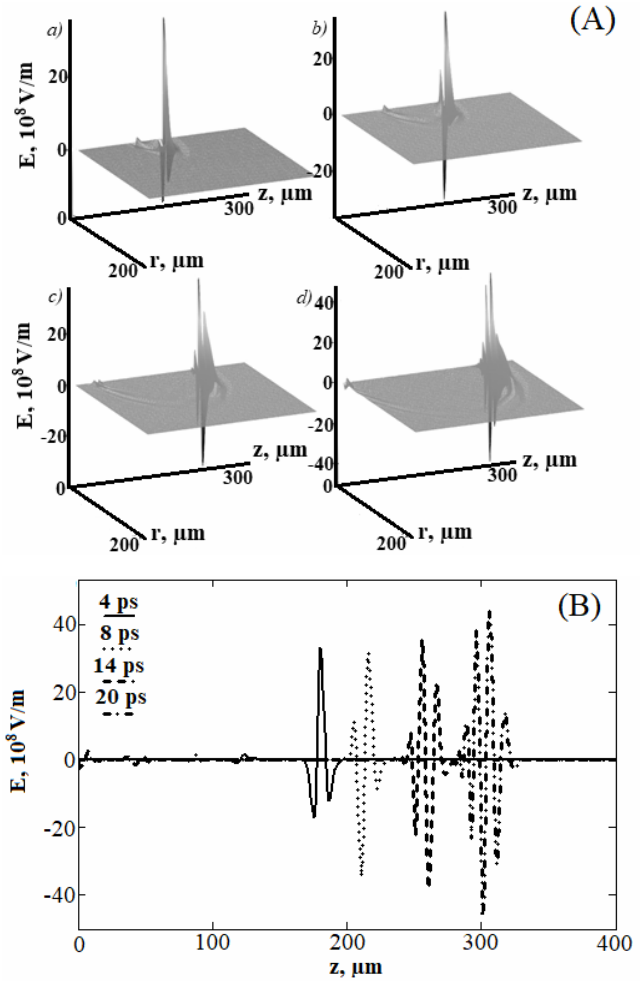

Fig. 2 – A) Dynamics of the three-dimensional ultrashort optical pulse in the photonic crystal made of CNTs with nonlinear absorption and amplification at different instants of time: a) 4 ps, b) 8 ps, c) 14 ps, d) 20 ps; B) Slices of a three-dimensional extremely short optical pulse at $r = 0$. The refractive index modulation period is $\chi = 2.5$ µm and the refractive index modulation depth is $\alpha = 0.25$. The nonlinear absorption rate is $F_1 = F_2 = 0.01$.

According to Fig. 2, we can conclude that the pulse energy remains localized in a limited spatial region and the pulse propagates stably. However, due to diffraction effects, a damping effect takes place, and a “tail” also appears at the trailing edge of the pulse (see also Figs. 3 and 4).

The dependence of a three-dimensional ultrashort optical pulse at 8 ps with various parameters of the photonic crystal is shown in Figs. 3 and 4. These plots show that the pulse shape depends on the modulation depth of the refractive index, but its energy remains in a limited spatial region when this index changes. In turn, varying the period of the refractive index, one can control the change in the group velocity of the wave packet of the ultrashort optical pulse. The longer the period, the faster the pulse propagates, because reflection and interference processes occur less frequently.

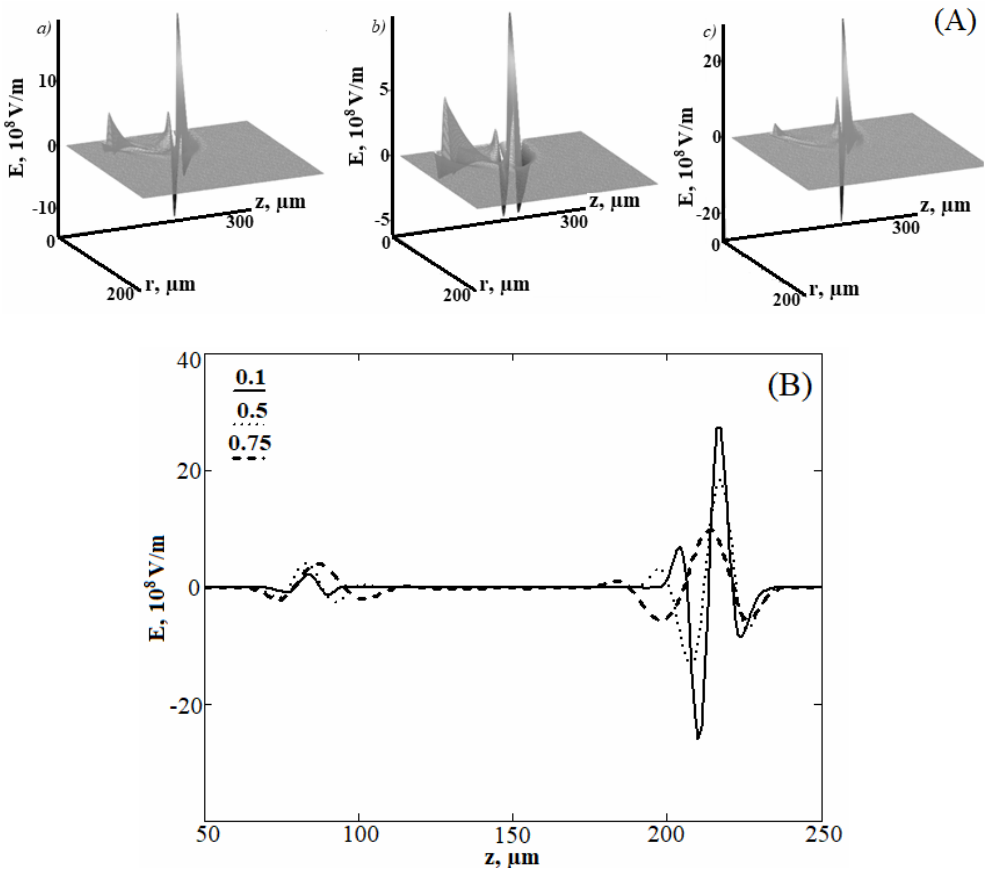

Fig. 3 – A) Dependence of a three-dimensional ultrashort optical pulse in the photonic crystal made of CNTs with nonlinear absorption and amplification at different refractive index modulation depths (α) for t = 8 ps: a) 0.1, b) 0.5, c) 0.75; B) Slices of a three-dimensional extremely short optical pulse at r = 0. The refractive index modulation period is χ = 2.5 μm.

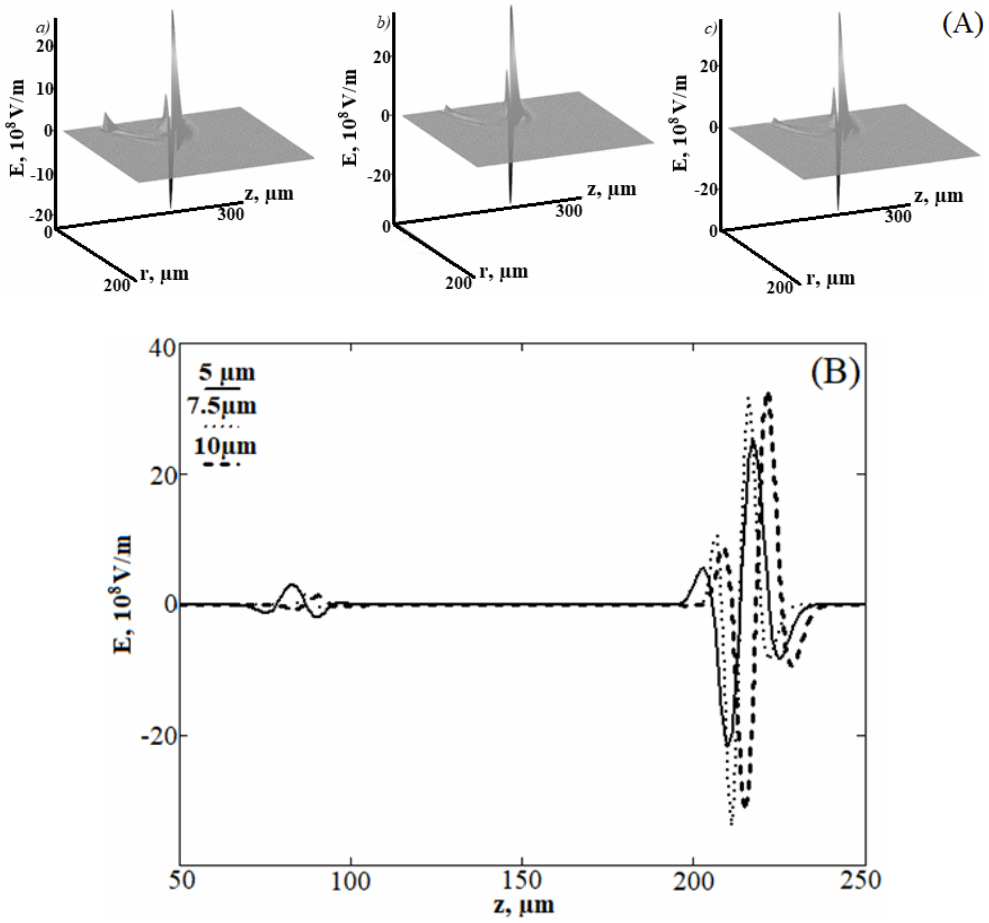

Fig. 4 – A) Dependence of a three-dimensional ultrashort optical pulse in the photonic crystal made of CNTs with nonlinear absorption and amplification at different refractive index modulation periods (χ) for $t = 8$ ps: a) 5 μm, b) 7.5 μm, c) 10 μm; B) Slices of a three-dimensional extremely short optical pulse at $r = 0$. The refractive index modulation depth is $\alpha = 0.25$.

As expected, when the lattice period increases, ultrashort pulse propagates faster. Obviously, with the infinite lattice period, due to the absence of interference processes, the pulse propagates at maximum speed. Numerical calculations confirmed this fact. We also note a significant distortion in the pulse shape. Dispersion spreading along the sample axis is compensated by the nonlinearity of the medium of CNTs, due to the nonparabolic dispersion law of electrons in the conduction band.

Another key task is to verify the stability of the obtained solutions of Eq. (5) depending on the electric field perturbation, which for small perturbations δA has the form:

$$\frac{\partial^2 \delta A}{\partial z^2}+\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial \delta A}{\partial r}\right)+\frac{1}{r^2}\frac{\partial^2 \delta A}{\partial \varphi^2}-\frac{n^2(z,r)}{c^2}\frac{\partial^2 \delta A}{\partial t^2}+$$
$$+\Gamma\frac{\partial \delta A}{\partial t}+\frac{4en_0}{c}\sum_{q=1}^{\infty}b_q\cos\left(\frac{aeqA}{c}\right)\cdot\frac{\delta A\cdot aeq\cdot f(t)}{c}- \tag{11}$$
$$-3F_1\left(\frac{\partial A}{\partial t}\right)^2\cdot\delta A-\frac{F_2\cdot\delta A}{1+\Delta\left(\frac{\partial A}{\partial t}\right)^2}+\frac{2\Delta F_2\left(\frac{\partial A}{\partial t}\right)^2\delta A}{\left(1+\Delta\left(\frac{\partial A}{\partial t}\right)^2\right)^2}=0.$$

Due to the linearity of (11), δA can be found as:

$$\delta A\propto\delta A(r,z,t)\exp(i\cdot n\cdot\varphi), \tag{12}$$

where n is the azimuthal number. Next, we can calculate the corresponding corrections to the electric field: $\delta E=-\partial\delta A/c\partial t$.

The dependence of the electric field strength of the ultrashort pulse, in the photonic crystal made of CNTs, on the azimuthal quantum number n that varied in the range from 0 to 9, is shown in Fig. 5.

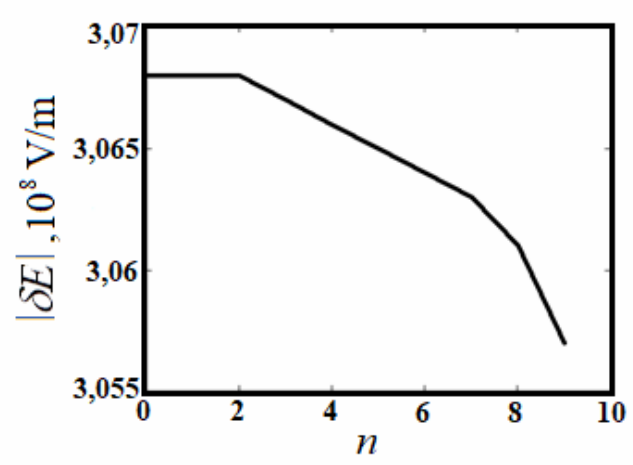

Fig. 5 – The dependence of maximum $|\delta E|$ on n (t = 8 ps).

Figure 5 shows that perturbations decrease over time. Moreover, the faster the perturbation decreases, the larger the azimuthal quantum number is. So, the obtained solutions are stable with respect to perturbations in angle φ.

4. CONCLUSIONS

The main results of this paper are summarized as follows:

1. A physical model, which describes the dynamics of three-dimensional ultrashort optical pulses in photonic crystals made of carbon nanotubes with nonlinear absorption and pumping, is proposed.

2. The stable pulse propagation in such optical media with conserving pulse energy in a limited spatial region is found. The pulse shape slightly changes, and after its passage a "tail" is formed.

3. The period and the depth of modulation of the refractive index have an influence on the shape and the group velocity of the ultrashort optical pulse.

4. The electric field perturbations decrease faster with the increase of the azimuthal number.

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