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Modeling of the dynamics of extremely short optical pulses in carbon nanotubes with random impurities taking into account multiphoton absorption

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In this work, we study the evolution of an extremely short pulse in a dielectric crystal with carbon nanotubes containing an impurity with random parameters (energy level, electron hybridization energy). The dependence of the spatial characteristics of the pulse on the impurity parameters and the number of photons taken into account in the model is analyzed.

Keywords: carbon nanotubes, impurities, optical pulse, multiphoton absorption.

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Introduction

It is known that in the synthesis of carbon nanotubes (CNTs) [1] one often has to deal with the presence of various kinds of impurities, the removal of which is very important in solving many applied problems, since they can significantly change the properties of CNTs. At the same time, purification from impurities is the most time-consuming step [2–4]. However, in some cases, the presence of an impurity may not have a significant effect on the nature of the process under study. Note that the impurity parameters (energy level, electron hybridization energy) can turn out to be random and vary within rather wide limits, due to the different environment of the impurity, its different random position relative to the nanotube hexagons (in the center of the hexagon, on the C−C-bond, above the carbon atom), as well as the presence of other nearby impurities or adsorbed atoms. Therefore, in this paper, we study the effect of such impurity on the dynamics of extremely short optical pulse as it propagates in a nonlinear medium with CNTs, taking into account multiphoton absorption [5] according to the model given in the paper [6].

Note that the solution of the problem stated is important, since CNTs are often used in the development of optoelectronic devices [7–9], including waveguide structures [10,11] and ultrashort pulse lasers [12].

1. Model and basic equations

Let us consider a $3D$ electromagnetic pulse passed through a dielectric matrix containing an array of impurity CNTs. Note that impurities in CNTs are randomly distributed, which is often observed when nanotubes are obtained industrially.

According to the paper [13], we write the dispersion law for CNTs containing impurities (ε_{imp}):

$$\varepsilon_{imp}(p,s)=0.5\Big(2B+\sqrt{-4(D(f+f^*)-\varepsilon(p,s)^2-D^2)}\Big), \tag{1}$$

where $\varepsilon(p,s)=|f|$ — dispersion law for zigzag CNT electrons without impurity [14], $s=1,2,\ldots,m$, p — electron quasi-pulse, B — parameter characterizing the processes associated with electron transitions from impurity levels to one of the nanotube sublattices,

$$B=-\sum_{j=1}^{4}\frac{|\alpha_j|^2}{W_j}, \tag{2}$$

where W_j — is the energy of an electron localized at the j-th impurity level, α_j — are quantities equal to the hopping integrals related to the impurity concentration between sublattice of CNTs and j-th impurity level, D — determines electron transitions between two CNT sublattices:

$$D=\sum_{j=1}^{4}\frac{\alpha_{1,j}\alpha_{2,j}^*}{W_j}. \tag{3}$$

Note that we consider transitions between impurity levels and the first and second CNT sublattices to be equivalent; therefore, in formula (2), the indices 1 and 2 of the hopping integral are omitted.

As can be seen from formulas (2) and (3), we consider only 4 impurity levels, since it was found that with level number increasing its influence weakens [13].

Since the electric field is aligned with the axis of the nanotubes OZ, the transverse components are equal to zero, and we deal only with the z-component of the field only — $E(x,y,z,t)$. The electric current density is set in a similar way: $|bfj=(0,0,j(x,y,z,t))$.

Next, we write the three-dimensional wave equation of the nonzero component of the vector potential in a cylindrical coordinate system, and taking into account the calibration $\mathbf{E} = -c^{-1}\partial\mathbf{A}/\partial t$:

$$\Box\mathbf{A} + \frac{4\pi}{c}\mathbf{j}(\mathbf{A}) + \Gamma\frac{\partial\mathbf{A}}{\partial t} - K_P\left(\frac{\partial\mathbf{A}}{\partial t}\right)^{2n_p-1} = 0, \quad (4)$$

where c — speed of light, n_p — number of absorbed photons (single-photon absorption coefficient is combined with pumping coefficient), K_p — photon absorption coefficient jcite15, $\Box$ — Dalembertian, Γ — determines the pumping of the electric field, which was chosen as a sixth-order supergaussian function with amplitude Q_Γ:

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

Here l_Γ determines the width of the amplifying medium in the direction perpendicular to the direction of pulse propagation.

Note that in the case of extremely short pulses, amplification in a two-level medium is either proportional to the distance traveled ($\sim z$) [16], or is described by processes like negative diffusion [17,18]. Here we use a simplified model and assume that the amplification is constant at each point z [19]. In this paper the attention is focused on the effect of randomly distributed impurities in CNTs and multiphoton absorption on the pulse dynamics. Accounting for more complex amplification models is beyond the scope of this study and will be carried out separately.

The expression for the current density along the CNT axis can be written as [20]:

$$j = 2e\sum_{s=1}^{m}\int_{-\pi/a}^{\pi/a}\frac{\partial\varepsilon_{imp}(p,s)}{\partial p}F(p,s)dp, \quad (6)$$

where e — electron charge, $F(p,s)$ — Fermi distribution function.

Thus, taking into account the symmetry by the angle $(\partial/\partial\varphi \to 0)$ due to the small accumulated charge because the field is inhomogeneous [21], the effective equation can be written as

$$\Box A + \frac{4en_0\gamma_0 a}{c}\sum_{q=1}^{\infty} b_q \sin\left(\frac{aeqA}{c}\right)\exp\left(-\frac{t}{\tau}\right)$$

$$+\Gamma\frac{\partial A}{\partial t} - K_P\left(\frac{\partial A}{\partial t}\right)^{2n_p-1} = 0, \quad (7)$$

where n_0 — is the electron concentration, $\gamma_0 \approx 2.7\,\text{eV}$, $a = 3b/2\hbar$, $b = 0.142\,\text{nm}$, exponent here takes into account the attenuation of the pulse field at times when the pulse intensity at its leading edge is by e times less than the peak intensity of the pulse, τ — relaxation time of the CNT electronic subsystem,

$$n_q = \sum_s \frac{q}{\gamma_0} a_{sq}\int_{-\pi/a}^{\pi/a} F(p',s)dp',$$

$$a_{sq} = \int_{-\pi/a}^{\pi/a}\int_0^{\gamma_0}\frac{\cos(pq)}{\sqrt{2\pi}\Delta}$$

$$\times\exp\left(-\frac{(D-D_0)^2}{2\Delta^2}\right)\varepsilon_{imp}(p,s,D)dDdp, \quad (8)$$

where a_{sq} — coefficients in the decomposition of the electron dispersion law (1) in Fourier series, taking into account the random distribution of impurity parameters according to the normal law, D_0 — median, Δ — dispersion of this distribution. Since we believe that the impurity is distributed uniformly over the bulk of the CNT array, and the impurity parameters are affected by a large number of random factors, the Gaussian distribution is a good model in this case. Note that we explicitly indicated the argument D in the dispersion law under the integral sign, since it also depends on the integration variable.

In the sum from equation (7) we take into account the first 10 terms, since the coefficients b_q decrease strongly with q increasing [22].

2. Numerical simulation and results

The effective equation (7) after reduction to a dimensionless form was solved numerically taking into account the initial conditions for the field vector potential in the form of a Gauss function (9a) and Bessel function (9b):

$$A = Q\exp\left(-\frac{z^2}{l_z^2}\right)\exp\left(-\frac{r^2}{l_r^2}\right),$$

$$\frac{dA}{dt} = \frac{2\nu_0 zQ}{l_z^2}\exp\left(-\frac{z^2}{l_z^2}\right)\exp\left(-\frac{r^2}{l_r^2}\right), \quad (9a)$$

$$A(r,z,0) = QJ_0\left(\frac{r}{l_r}\right)\exp\left(-\frac{(z-z_0)^2}{l_z^2}\right)\exp\left(-\frac{r}{\gamma}\right),$$

$$\frac{dA(r,z,0)}{dt} = \frac{Q\nu_0(z-z_0)}{l_z^2}J_0\left(\frac{r}{l_r}\right)$$

$$\times\exp\left(-\frac{(z-z_0)^2}{l_z^2}\right)\exp\left(-\frac{r}{\gamma}\right), \quad (9b)$$

where Q — the amplitude of the electromagnetic pulse at the initial moment of time, l_z, l_r — the pulse width along the corresponding directions, ν_0 — the initial pulse speed along the nanotube axis, z_0 — initial shift of the pulse center along axis OZ, γ — cutoff parameter for the Bessel function.

The evolution of the field strength of extremely short optical pulse during its propagation in a dielectric medium with CNTs taking into account the processes of two-photon absorption is shown in Fig. 1.

The Figure demonstrates the localized propagation of the pulse, which is facilitated by the balance of pumping and damping.

A comparison of the cases considering two-photon and three-photon absorption is shown in Fig. 2.

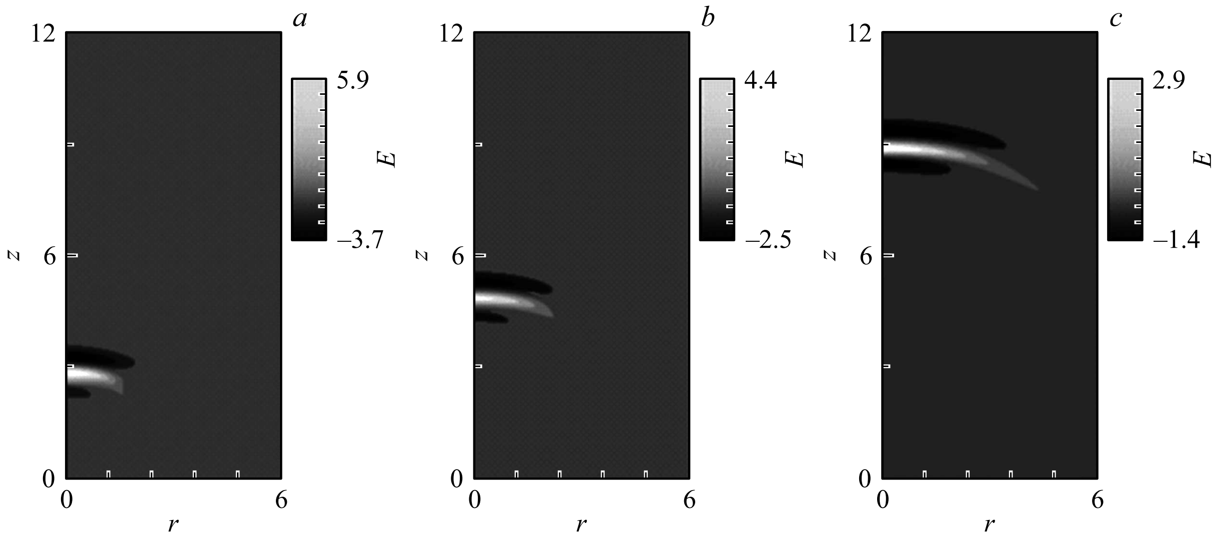

Figure 1. Electric field strength vs. coordinates ($D_0 = -B = 0.1\gamma_0$): t, s: a — $2 \cdot 10^{-14}$, b — $6 \cdot 10^{-14}$, c — $8 \cdot 10^{-14}$. Unit along axis E — 10^7 V/m.

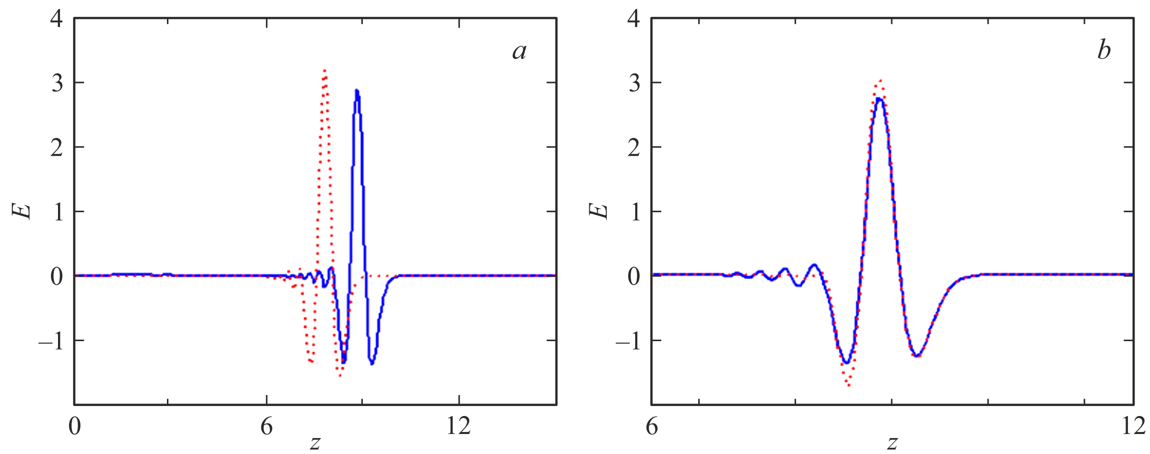

Figure 2. Longitudinal sections of the field E at $r = 0$ depending on the coordinate z for different number of photons ($t = 8 \cdot 10^{-14}$ s): a — initial conditions in the form (7a); b — initial conditions in the form (7b). The solid curve corresponds to $n_p = 2$, the dashed curve corresponds to $n_p = 3$. The unit along axis E is taken to be 10^7 V/m.

It can be seen from Fig. 2 that the number of absorbed photons affects both the field intensity and the distance traveled by the pulse. In the case of three-photon absorption, the amplitude of the extremely short pulse is greater than for two photons. But at the same time, the pulse experiences a delay. This is due to nonlinear absorption and further interference, which have a different nature with respect to two and three-photon absorption. Namely, three-photon absorption is more sensitive to processes occurring at the pulse leading and trailing edges, and less sensitive to processes near its maximum.

The effect of the energy of transitions between impurity levels and CNT sublattices on the shape of the extremely short pulse is shown in Fig. 3.

According to Fig. 3 we can conclude that the integrals of the transition between impurity levels and CNT sublattices have significant effect on the pulse shape in the case of three-photon absorption. As before, we attribute this to the different character of the parity of the term responsible for the nonlinear absorption under time reversion. Namely, if for two-photon absorption the contribution of the nonlinearity responsible for absorption „works“ smaller at the pulse leading and trailing edges, then for three-photon absorption its contribution is greater.

Note that in the case of pulse with a transverse Bessel profile, the impurity parameters B and D do not affect the shape of the pulse.

Next, we studied the features of the pulse propagation upon parameters change of impurity responsible for its random distribution (D_0, Δ). The performed calculations showed that the parameters of the random distribution of the impurity do not have a significant effect on the

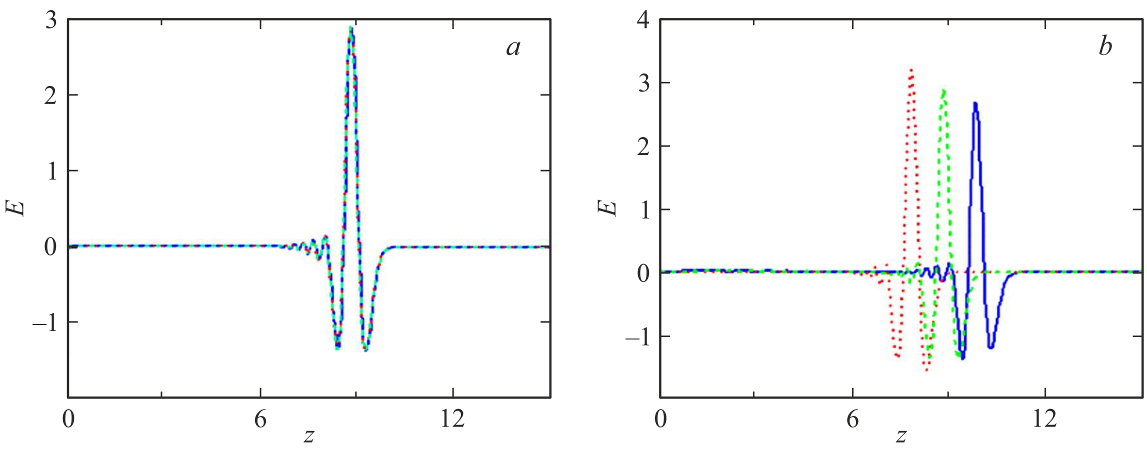

Figure 3. Longitudinal sections of the field E vs. coordinate z for different number of photons ($t = 8 \cdot 10^{-14}$ s, initial conditions (7a)); n_p: a — 2; b — 3. Solid curve corresponds to $D_0 = -B = 0.1\gamma_0$, dashed line — $D_0 = -B = 0.5\gamma_0$, dashed line — $D_0 = -B = 1.0\gamma_0$. The unit along axis E is taken to be 10^7 V/m.

pulse dynamics. So, in the case of pulse with transverse Bessel profile only the influence of the D_0 parameter, which determines the median of the random distribution, is observed, it manifests itself as change in the pulse amplitude by about 10%. Thus, even in the presence of impurity in CNTs, the latter can be used as elements placed in the dielectric medium for the localized propagation of the electromagnetic field. For pulse with a Gaussian profile in the case of three-photon absorption, the number of control parameters of randomly distributed impurity is larger (transition integrals B and D, median D_0 and dispersion Δ). In this case the main effect is associated with change in the propagation speed of the extremely short pulse during three-photon absorption.

Conclusion

The main conclusions can be formulated as follows.

1. A model is made for the interaction of electromagnetic pulse with nonlinear medium containing CNTs with random impurities, taking into account the processes of multiphoton absorption and the pumping field.

2. It is shown that the presence of impurity with randomly distributed parameters in CNTs does not affect the stable propagation of the extremely short pulse with initial profile in the form of Gaussian and Bessel function.

3. It is found that in the case of model that takes into account three-photon absorption, the influence of the impurity parameters manifests itself as decrease in the propagation speed of the extremely short optical pulse.

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Conflict of interest

The authors declare that they have no conflict of interest.

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