NANOSYSTEMS: PHYSICS, CHEMISTRY, MATHEMATICS, 2014, 5 (1), P. 91–97

PROPAGATION OF FEMTOSECOND PULSES IN CARBON NANOTUBES

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PACS 73.63.Fg, 78.67.Ch

The generation of higher harmonics of carbon nanotubes interacting with femtosecond laser pulses was investigated. The analysis was conducted on the basis of quantum kinetic equation for the π-electrons involved in the inside of the band and inter-band transitions. The dynamics of the electromagnetic pulse, depending on the parameters of the problem, were studied.

Keywords: Carbon nanotube, femtosecond pulse, electromagnetic field.

1. Introduction

This paper investigates the dynamics of femtosecond pulses propagating in carbon nanotubes (CNTs). Significant progress in the synthesis of new mesoscopic structures and their great promise for use in nanoelectronics and optics has led to increased research in this area. The unusual manifestation of nonlinear optical properties of these materials when interacting with strong electromagnetic fields was expected. Theoretical and experimental studies predict that the strong nonlinear response is inherent in metal clusters [1–3], fullerenes [4–6], carbon nanotubes and composites based on them [7, 8]. In particular, the generation of higher harmonics, previously observed in noble gases [9, 10], has been predicted for mesoscopic structures. Moreover, a distinctive feature of the CNT is a strong selectivity of their high harmonic spectrum. A theoretical study of the nonlinear response of isolated nanotubes by intense laser radiation was studied in this work. A full quantum-theoretical approach to the one-electron approximation in the tight-binding model was used.

2. Statement of a problem

2.1. Kinetic equations

We consider an infinitely long single-walled carbon nanotube oriented along the z axis and irradiated normal to the axis of the ultra-short pulse with a natural frequency w_0, polarized to this axis: $\mathbf{E}(\mathbf{r}) = e_z E_z(x, y)$ (e_z is the unit vector along the z axis). CNTs are considered as a single-layer graphene sheet which is scrolled into a cylinder. We take into account only π-electrons, suggesting that their motion can be described in the tightly-bound approximation [11, 12]. CNT radius is much smaller than the wavelength of the field, which allows us to neglect the spatial inhomogenity of the field in the tubes.

Following [13] in the one-electron approximation, the Schrdinger equation becomes:

$$i\hbar\frac{\partial\psi}{\partial t} = -\frac{\hbar^2}{2m_0}\Delta\psi + [W(\mathbf{r}) - e(\mathbf{E}\cdot\mathbf{r})]\,\psi. \qquad (1)$$

Neglecting the harmonic solution of (1) can be written as:

$$\psi = \sum_{l,\mathbf{p}} C_l(\mathbf{p})\psi_l(\mathbf{p},\mathbf{r}), \tag{2}$$

where l is the set of quantum numbers characterizing the state of π-electrons with a given quasi-momentum. $\psi_l(\mathbf{p},\mathbf{r}) = \hbar^{-0.5}\exp(i\mathbf{pr}/h)\,u_{l,\mathbf{p}}(\mathbf{r})$ are the Bloch functions with an amplitude $u_{l,\mathbf{p}}(\mathbf{r})$, which is periodical to an arbitrary vector lattice $\mathbf{a}$: $u_{l,\mathbf{p}}(\mathbf{r}+\mathbf{a}) = u_{l,\mathbf{p}}(\mathbf{r})$. Here $\mathbf{a} = n_1\mathbf{a}_1 + n_2\mathbf{a}_2$, n_1, n_2 are the integers, $\mathbf{a}_1$, $\mathbf{a}_2$ are the elementary vectors of the graphene hexagonal lattice.

The dispersion law for zig-zag CNTs can be written in the following form [14]:

$$\varepsilon_s(p) = \pm\gamma_0\sqrt{1 + 4\cos(ap)\cos(\pi s/m) + 4\cos^2(\pi s/m)}, \tag{3}$$

where $s = 1, 2, \ldots m$, $(m, 0)$ is type of CNT, γ_0 is the hopping integral (2.7 eV), $a = 3b/2\hbar$, $b = 0.142$ nm is the distance between the adjacent carbon atoms.

From equations (1) and (2) with the spectrum of nanotubes:

$$\hat{H}_0 = \begin{pmatrix} 0 & H_{12}(\mathbf{p}) \\ H_{21}^*(\mathbf{p}) & 0 \end{pmatrix}, \tag{4}$$

which can be rewritten in the tightly-bound approximation as:

$$H_{12}(\mathbf{p}) = -\gamma_0\sum_{j=1}^{3}\exp\left(\frac{i\mathbf{p}\tau_j}{\hbar}\right), \tag{5}$$

where τ_j is the vector connecting the atom to its nearest neighbors. For density matrix elements taking into account $F = \Re(\rho_{cv})$; $\Phi = \Im(\rho_{cv})$; $\rho = \rho_{cc}$, ρ_{cv} is the density matrix, ω is the transition frequency, is the electron charge, we can obtain the following expression:

$$\begin{gathered} \frac{\partial\rho}{\partial t} + eE_z\frac{\partial\rho}{\partial p_z} = 2\frac{e}{\hbar}E_z R_{ab}\Phi, \\ \frac{\partial F}{\partial t} + eE_z\frac{\partial F}{\partial p_z} = \omega\Phi, \\ \frac{\partial\Phi}{\partial t} + eE_z\frac{\partial\Phi}{\partial p_z} = \frac{e}{\hbar}E_z R_{ab}(2\rho - 1) - \omega F, \\ R_{ll',\mathbf{p}} = \frac{i\hbar}{2}\int\limits_{\Omega}\left(u_{l,\mathbf{p}}^*\frac{\partial u_{l',\mathbf{p}}}{\partial p_z} - \frac{\partial u_{l,\mathbf{p}}^*}{\partial p_z}u_{l,\mathbf{p}}\right)d^2\mathbf{r}. \end{gathered} \tag{6}$$

The integration domain is the volume Ω of two-dimensional unit cell.

The initial conditions are following:

$$\rho_{t=0} = F_0\left(\varepsilon_c(p_z, s)\right);\ F|_{t=0} = \Phi|_{t=0} = 0. \tag{7}$$

This means that at room temperature, the electrons are distributed according to the equilibrium Fermi distribution with zero chemical potential ($\mu = 0$).

The boundary conditions reflect the periodicity of solutions in the space of quasi-momentum (similar for F and Φ):

$$\rho\left(t, \frac{\sqrt{3}\pi}{\omega_{cn}a}\right) = \rho\left(t, -\frac{\sqrt{3}\pi}{\omega_{cn}a}\right). \tag{8}$$

3. Electric current in nanotube

A quantum-mechanical operator of current density can be written in the following form [13]:

$$\hat{j}_z(r) = -\frac{ie\hbar}{2m_0}\left(\frac{\partial}{\partial z'}\delta(r-r') + \delta(r-r')\frac{\partial}{\partial z'}\right). \tag{9}$$

Total current density was decomposed into two components: $\hat{j}_z = j_z^{(1)} + j_z^{(2)}$, where intraband transitions is responsible for:

$$j_z^{(1)} = \frac{4e}{(2\pi\hbar)^2}\int\limits_{1ZB}\frac{\partial\varepsilon_c(\mathbf{p})}{\partial p_z}\rho(t,\mathbf{p})\,d^2\mathbf{p}, \tag{10}$$

and inter-band:

$$j_z^{(2)} = \frac{8e}{(2\pi\hbar)^2\hbar}\int\limits_{1ZB}\varepsilon_c(\mathbf{p})\,R_{cv}(p_z,s)\,\Phi(t,\mathbf{p})\,d^2\mathbf{p}. \tag{11}$$

We take into account that $\rho_{vv} + \rho_{cc} = 1$ and $\varepsilon_v = -\varepsilon_c$.

Choosing the gauge field, $\mathbf{E} = -\frac{1}{c}\frac{\partial\mathbf{A}}{\partial t}$, (where is the light velocity in a vacuum). We need to change the momentum to a generalized momentum: $\mathbf{p} \to \mathbf{p} - e\mathbf{A}/c$, so:

$$\left\{\begin{array}{c}\Phi\\F\\\rho\end{array}\right\} = \hat{x}\left(\mathbf{p} - \frac{e\mathbf{A}}{c}\right)\left\{\begin{array}{c}\Phi\\F\\\rho\end{array}\right\}.$$

Then, equation (5) takes the following form with the replacement $\rho - 0.5 \to \rho$:

$$\left\{\begin{array}{l}\rho_t = -\frac{2}{c}A_t R\Phi,\\F_t = \omega\Phi,\\\Phi_t = -\frac{2}{c}A_t R\rho - \omega F.\end{array}\right. \tag{12}$$

The matrix of the system coefficients can be written as:

$$\hat{x} = \left[\begin{array}{ccc}0 & 0 & A\\0 & 0 & \omega\\A & -\omega & 0\end{array}\right].$$

Where the notation: $A = -2c^{-1}A_t R$ and according to the approximation described in [15–19]:

$$e^{\hat{x}} = \hat{I} + \hat{x}\left(1 + \frac{(A^2-\omega^2)}{3!} + \frac{(A^2-\omega^2)^2}{5!} + \ldots\right) + \hat{x}^2\left(\frac{1}{2!} + \frac{(A^2-\omega^2)}{4!} + \frac{(A^2-\omega^2)^2}{6!} + \ldots\right),$$

$$j_z^{(1)} = \frac{4e}{(2\pi\hbar)^2}\int\limits_{1ZB}\frac{\partial\varepsilon_c(\mathbf{p}-e\mathbf{A}/c)}{\partial p_z}A(t)^2\sum_{k=0}^{\infty}\frac{\left(A(t)^2-\omega(\mathbf{p})^2\right)^k}{(2k+2)!}\frac{1}{(1+\exp(-\varepsilon_c(\mathbf{p})/k_BT))}d^2\mathbf{p},$$

$$j_z^{(2)} = \frac{8e}{(2\pi\hbar)^2\hbar}\int\limits_{1ZB}\frac{-2A(t)\,\varepsilon_c(\mathbf{p}-e\mathbf{A}/c)\,R_{cv}(\mathbf{p})}{(1+\exp(-\varepsilon_c(\mathbf{p})/k_BT))}\sum_{k=0}^{\infty}\frac{\left(A(t)^2-\omega(\mathbf{p})^2\right)^k}{(2k+1)!}d^2\mathbf{p},$$

where k_B is the Boltzmann constant, T is the temperature.

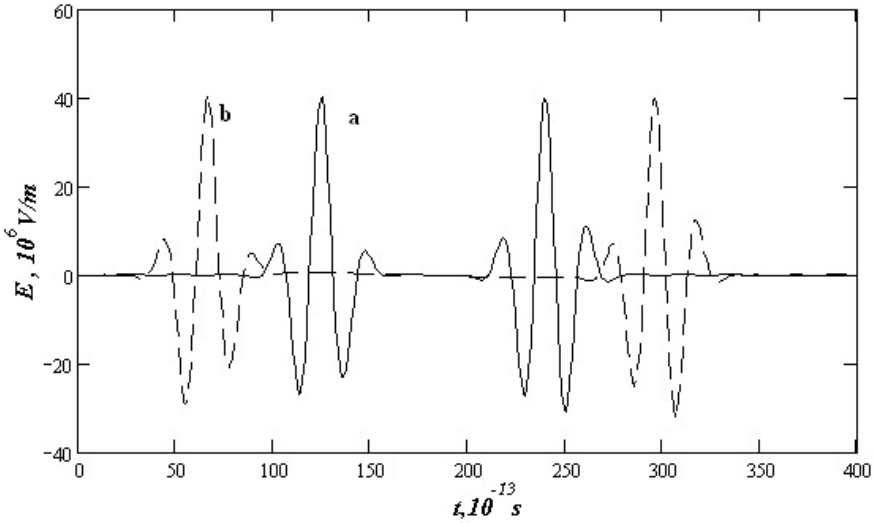
The Maxwell equations for the dielectric non-magnetic medium can be written as following [16]:

$$\frac{\partial^2 \mathbf{A}}{\partial x^2}+\frac{2N_0}{c}a\frac{\partial^4 \mathbf{A}}{\partial t^4}-\frac{2N_0}{c}b\mathbf{A}+\frac{4\pi}{c}\left(j_1+j_2\right)-\frac{1}{c^2}\left(1+4\pi\alpha\right)\frac{\partial^2 \mathbf{A}}{\partial t^2}-\frac{12\pi\eta}{c^4}\frac{\partial^2 \mathbf{A}}{\partial t^2}\left(\frac{\partial \mathbf{A}}{\partial t}\right)^2=0, \quad (13)$$

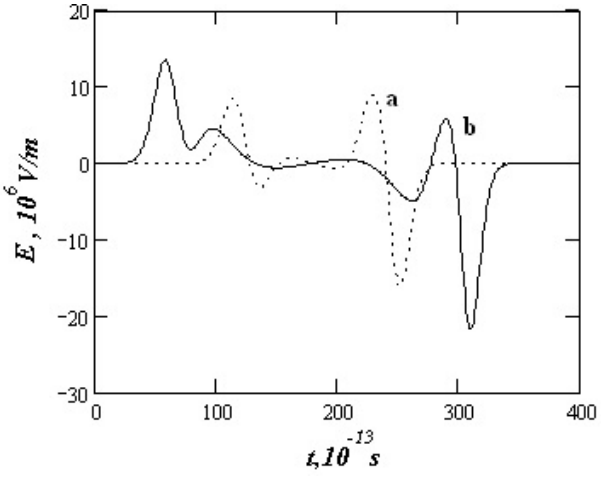
where $\mathbf{A}$ is the vector-potential, $\mathbf{P}_L=\alpha\mathbf{E}$ is the polarization, $\mathbf{P}_{NL}=\eta\left|\mathbf{E}\right|^2\mathbf{E}$ is the nonlinear part of polarization, t is the time. We consider a simple model, where the polarization vector is parallel to the vector $\mathbf{E}$ is the electric field of light wave, $\mathbf{N}_0$, a, a_1, $\ldots$, b, b_1, $\ldots$ are the empirical constants of medium dispersion [17].

4. Numerical analysis and results

Equation (13) was solved numerically [20]. Boundary conditions took on a Gaussian form with one field- (14a) and two field oscillations (14b):



(A)



(B)

FIG. 1. Time dependence of the electric field at various space points a) $x=0.1\cdot10^{-5}$ m; b) $x=0.2\cdot10^{-5}$ m. (A) two field oscillations; (B) one field oscillation

$$A(0,t) = Q \cdot e^{-(ut)^2/\gamma},$$
$$\frac{dA(0,t)}{dx} = \frac{2Qut}{\gamma} e^{-(ut)^2/\gamma}, \tag{14a}$$

$$A(0,t) = Q \cdot e^{-(ut)^2/\gamma} \sin(ut),$$
$$\frac{dA(0,t)}{dx} = \frac{2Qut}{\gamma} e^{-(ut)^2/\gamma} \sin(-ut) + Q \cdot e^{-(ut)^2/\gamma} \cos(ut), \tag{14b}$$

where Q is the pulse amplitude, u is the pulse velocity, $\gamma^2 = 1/(1 - u^2/A^2)$.

The evolution of the electric field during its propagation in the sample is shown in Fig. 1.

It can be seen from this dependence that when the propagation distance increases, pulses drift away from each other. Moreover, in the case of two field oscillations, the amplitude of the main peak is stable, and in the case of one oscillation there is a decrease in the main pulse amplitude and an increase in the trailing pulse. This can be attributed to the dispersion characteristics of the medium.

The dependence of the electric field on the initial pulse amplitude is shown in Fig. 2.

As expected, low-amplitude pulses propagate almost without distortion and exhibit only the dispersion spread. However, pulses with high amplitudes exhibit a larger distortion caused by both the interference of the edge pulse and the specific form of the medium's nonlinearity.

Figure 3 demonstrates that the dispersion constants have a great influence on the electric field (and the effect of the b constant is manifested much more strongly compared to the constant a). In our opinion, this effect is due to this type of dispersion reducing the linear response of the system to an external field.

Thus, based on the results of numerical calculations, one can draw the conclusion that the pulse propagation is determined primarily by the dispersion characteristics of the medium, as well as the process of establishing a balance between the dispersion and interference effects of the wave front.

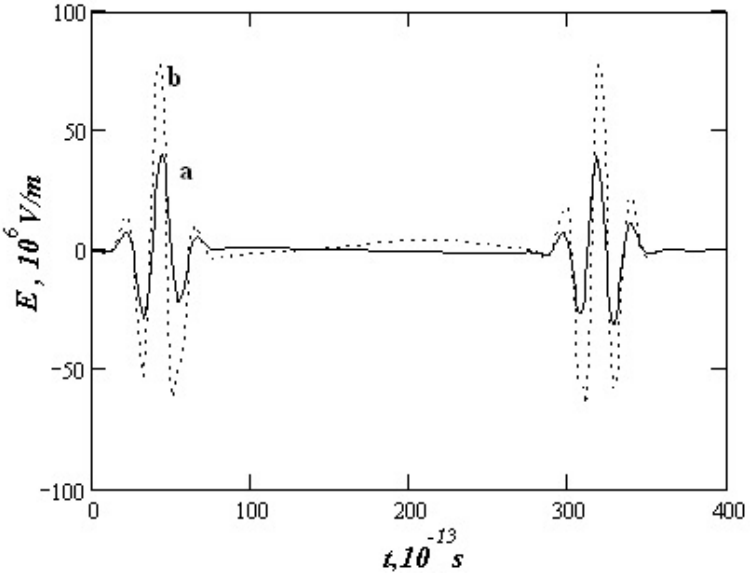


FIG. 2. Time dependence of the electric field at various initial amplitudes of the pulse ($x = 0.2 \cdot 10^{-5}$ m): a) $Q = 1$ a.u.; b) $Q = 2$ a.u. (the case of two field oscillations)

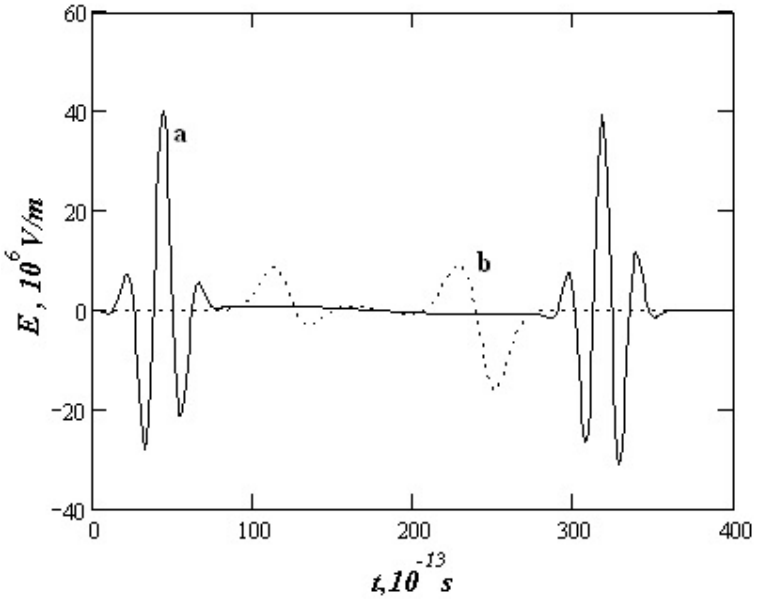


FIG. 3. Time dependence of the electric field at various dispersion constants ($x = 0.1 \cdot 10^{-5}$ m): a) $b = 0.001$ a.u; b) $b = 0.005$ a.u. (the case of two field oscillations)

Acknowledgments

This study was supported by the Russian Foundation for Basic Research (project No. 12-02-31654).

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