Rabi oscillations and saturable absorption effect in single-wall carbon nanotubes

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Abstract. We have studied theoretically a single-wall carbon nanotube (SWNT) interaction with the short high-intensive electromagnetic pulse whose carrier frequency is in the resonance with the frequency of the electron interband transitions in the SWNT. For this purpose kinetic equations describing evolution of the density matrix of the electron sub-system have been solved numerically and spectra of the surface current density induced in the SWNT by the external electromagnetic field have been calculated. We have demonstrated that Rabi oscillations of the population of the SWNT electronic bands occur for the pulse intensity exceeding $10^{10}$ W/cm$^2$.

1. Introduction
Carbon nanotubes are known to possess strongly nonlinear optical response [1]. In particular, it has been demonstrated that single-wall carbon nanotubes (SWNTs) have high values of the third-order nonlinear optical susceptibility [2, 3, 4], may be used for effective generation of the high-order harmonics [5, 6] and as saturable absorbers for passive mode-locking in lasers [7, 8]. Among the different nonlinear processes Rabi oscillations of the inversion that can occur in two-level [9] or two-bands [10] systems illuminated by a high-intensive resonant electromagnetic field are of a great interest. We expect that due to the SWNTs strong optical nonlinearity Rabi oscillations will also occur in SWNTs illuminated by the external electromagnetic field.

2. System under the consideration
Let us consider single-wall carbon nanotube (SWNT) illuminated by an electromagnetic pulse (see Fig. 1) of the form:

$$
E(t) = e_z \begin{cases} 
E_0 \sin(\omega_0 t), & 0 \leq t \leq \sigma_0 \\
0, & t < 0, t > \sigma_0
\end{cases},
$$

where $E_0$ is the pulse amplitude, $\sigma_0$ is the pulse duration, $e_z$ is the unit vector along the $z$-axis. The direction of pulse propagation $k$ is normal to the SWNT axis and the vector of electric field strength $E(t)$ is polarized along the SWNT axis (see Fig. 1a). The pulse carrier frequency $\omega_0$ is in the resonance with the frequency of electron interband transitions $\omega_{cv}(p, s) = (E_c(p, s) - E_v(p, s))/\hbar$ (see Fig. 1b) for the electrons with quasi-momentum $p = 0$ in the sub-band $s = 9$, where

$$
E_{c,v}(p, s) = \pm \gamma_0 \sqrt{1 + 4 \cos \left( \frac{3pb}{2\hbar} \right) \cos \left( \frac{\pi s}{m} \right) + 4 \cos^2 \left( \frac{\pi s}{m} \right)}
$$

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is the electrons dispersion law, $\gamma_0 = 2.7 \text{ eV}$, $p$ is the electron quasi-momentum, indices $c,v$ stand for the conduction and valence bands, respectively. In $(m,0)$ SWNT both conduction and valence bands are split on $2m$ sub-bands [11], designated by index $s = 1, \ldots, 2m$. First Brillouin zone in the SWNT is defined by the condition $-\pi \hbar/3b < p \leq \pi \hbar/3b$, $b = 0.142 \text{ nm}$.

![Figure 1](image)

**Figure 1.** (a) Schematic representation of $(14,0)$ SWNT (black cylinder) illuminated by the electromagnetic pulse $E(t)$ of the form defined by Eq. (1). (b) Band structure of $(14,0)$ SWNT in the vicinity of the Fermi level.

### 3. Evolution of the dynamical inversion

To study the SWNT response on the applied electromagnetic field we solve numerically kinetic equations for the density matrix $\rho_{\alpha\beta}(t,p,s)$ of $\pi$-electrons in SWNT [3, Eq. (1)]

\[
\frac{\partial \rho_{\alpha\beta}(t,p,s)}{\partial t} + e E(t) \frac{\partial \rho_{\alpha\beta}(t,p,s)}{\partial p} = - \frac{\rho_{\alpha\beta}(t,p,s) - \rho^{eq}(p,s)}{T_1} + \frac{2ie}{\hbar} E(t) R_{cv}(p,s)(\rho_{cv}(t,p,s) - \rho_{cv}(t,p,s)),
\]

or

\[
\frac{\partial \rho_{cv}(t,p,s)}{\partial t} + e E(t) \frac{\partial \rho_{cv}(t,p,s)}{\partial p} = - \left( \frac{1}{T_2} + i \omega_{cv}(p,s) \right) \rho_{cv}(p,s) - \frac{i e}{\hbar} E(t) R_{cv}(p,s) \rho_{nv}(t,p,s),
\]

where $T_{1,2} = 40 \text{ fs}$ is the electron relaxation time, $R_{cv}(p,s)$ is the normalized matrix elements of the dipole momentum operator, $\rho_{nv}(t,p,s) = \rho_{cv}(t,p,s) - \rho_{cv}(t,p,s)$ is the dynamical inversion.

Results of calculations are presented in Fig. 2 for $(14,0)$ SWNT illuminated by the electromagnetic field defined by Eq. (1). We restrict our consideration to sub-band $s = 9$ and only small part ($-0.3 \leq p \leq 0.3$) of first Brillouin zone of the SWNT corresponding to the electron interband transitions frequencies $\omega_{cv}(p,s)$ that are close to the carrier frequency $\omega_0$ of the pulse. As one can see, for low intensities of the driving field (Fig. 2a,b) the dynamical inversion increases while the pulse is switched on and decreases exponentially after the pulse passage due to the relaxation effects. The slight inversion oscillations with the frequency equals to the carrier frequency of the pulse is due to the intraband electrons motion. With the pulse intensity increase (Fig. 2c,d) we observe that high frequency oscillations with the period $T_0$ are superimposed by the low frequency oscillations with the oscillations period $T_R$. These are so-called Rabi oscillations of the inversion that are due to the electron transitions between the valence and conduction bands.
current density spectra with the frequencies increase of the intensity leads to the appearance of the additional spectral lines in the induced field intensity increase, i.e. the SWNT-electromagnetic field interaction is saturated. Further between the SWNT and external electromagnetic field occurs in the saturation regime (Fig. 3a) the value of the induced current is approximately proportional to the electric field strength, i.e. \( |j(\omega_0)| \sim E_0 \). For the driving field intensities exceeding \( 10^9 \) W/cm\(^2\) the interaction between the SWNT and external electromagnetic field occurs in the saturation regime (Fig. 3b). In this regime the value of the induced electric \(|j(\omega_0)|\) varies very slow with the electric field intensity increase, i.e. the SWNT-electromagnetic field interaction is saturated. Further increase of the intensity leads to the appearance of the additional spectral lines in the induced current density spectra with the frequencies \( \omega_0 \pm \Omega_R \). The origin of these lines are the Rabi oscillations of the dynamical inversion presented on Fig. 2c,d and \( \Omega_R \) is the Rabi frequency.

4. Surface current density

We also calculated the surface current density induced in the SWNT by the external electromagnetic field defined by Eq. (1)

\[
j(t) = \frac{e}{2\pi^2 h R_{cn}} \sum_{k=1}^{2m} \left( \frac{\partial \mathcal{E}_k(p, s)}{\partial p} \rho_{\text{inv}}(t, p, s) + i \omega_{\text{exc}}(p, s) R_{\text{exc}}(p, s)(\rho_{\text{exc}}(t, p, s) - \rho_{\text{exc}}(t, p, s)) \right) dp, \tag{5}
\]

where integration is performed over the first Brillouin zone of the SWNT. The spectra of the induced current density \( j(t) \) are presented on Fig. 3.

5. Conclusions

Concluding, we have studied the single-wall carbon nanotube (SWNT) interaction with the short high-intensive electromagnetic pulse whose carrier frequency is in the resonance with the frequency of the electron interband transitions in the SWNT. To describe the dynamics of the electron sub-system in the SWNT we use kinetic equations for the electron density matrix. These
Figure 3. (a)-(c) Spectra $|j(\omega)|$ of the surface current density $j(t)$ induced in (14,0) SWNT by electromagnetic field defined by Eq. (1); $\Omega_R$ is the Rabi frequency. (d) Intensity dependence of the spectral component $|j(\omega_0)|$ of the induced current.

equations have been solved numerically and the density of the surface current induced on the SWNT surface by the external electromagnetic field has been calculated. We have demonstrated that for the pulse intensities of order $10^9 \text{ W/cm}^2$ SWNTs interaction with pulse occur in the saturation regime. However, for the pulse intensities higher that $10^{10} \text{ W/cm}^2$ Rabi oscillations of the inversion have been predicted. The triplet Mollow in the spectra of the induced current density has been demonstrated.

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