Heating of a carbon nanotubes array with few-cycle optical pulses

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Abstract. In this paper, we investigate the heating of carbon nanotubes under the action of few-cycle optical pulses. Within the framework of the hot electron model, an array of carbon nanotubes which is irradiated by an electromagnetic wave with a high emission intensity is considered. The time-dependence of the generated heat is analyzed for various system parameters (duration of the few-cycle laser pulse and relaxation time of the electrons in carbon nanotubes).

1. Introduction
Exceptional properties of carbon nanotubes (CNTs) [1–3] provide a wide range of applications as electrothermal films, composites, sensors, etc. The excellent thermal conductivity of CNT films provides a high heating rate and a uniform temperature distribution with high optical transparency [4], and thus they are excellent heating elements. On the other hand, their highly nonlinear electrodynamic properties attract attention, including the interaction with few-cycle laser pulses (FCP) with durations corresponding to several periods of field oscillations [5], which makes it possible to use them as bolometers [6], as absorbing coatings in protective elements [7, 8], etc.

This work aims to study the heating of CNTs under the few-cycle optical pulse action. The interest in the study of FCP propagation is associated with several factors, such as the high directivity of their radiation, the stability of their shape and resistance to perturbations of the parameters, as well as the peak intensity of their field at which the waveguide material does not break down [9]. It should be noted, that the "light bullets" are of great interest among such few-cycle pulses. They are localized in all directions and propagating in a nonlinear medium at a constant speed [10, 11]. It should be noted, that the successes of modern laser technologies in terms of the formation of powerful electromagnetic radiation with given characteristics, including "light bullets", are an incentive for a comprehensive study of such pulses in various media, including carbon nanotubes [12–14].

We consider an array of CNTs in the framework of the hot electron model, and it is irradiated by an electromagnetic wave with an emission intensity of the order of $2 \, TW/cm^2$. This model allows us to adequately describe the results at strong excitation powers, in contrast to the perturbation theory. Photoexcited electrons absorb the energy of a given wave due to the processes of intraband and interband absorption. In the general case, the absorbed energy is released in the form of Joule heat, which depends only on the current component that is in phase with the electric field, and therefore is determined by the real part of the conductivity.
2. Model

We consider the beam of semi-infinite CNTs (the nanotube density is about $3 \cdot 10^{12}$ cm$^{-2}$ and tube radius $R_0$ is about $5.5 \cdot 10^{-8}$ cm), oriented perpendicular to the front of electromagnetic pulse (Figure 1a).

We take into account that the linear conductivity of carbon nanotubes has three components: intraband electron $\sigma^e$, intraband hole $\sigma^h$, and interband $\sigma_{in}$ [15]. We write down only their real parts:

$$\sigma^e(\omega) = \frac{e^2 g_e g_v}{16 \pi \hbar k_B T} \int_0^\infty \frac{\gamma \varepsilon d\varepsilon}{\varepsilon^2 + \gamma^2} \frac{1}{\cosh^2 \left( \frac{\varepsilon - \mu_e}{2k_B T} \right)},$$

$$\sigma^h(\omega) = \frac{e^2 g_e g_v}{16 \pi \hbar k_B T} \int_0^\infty \frac{\gamma \varepsilon d\varepsilon}{\varepsilon^2 + \gamma^2} \frac{1}{\cosh^2 \left( \frac{\varepsilon + \mu_h}{2k_B T} \right)},$$

$$\sigma_{in}(\omega) = \frac{e^2 g_e g_v (16 \pi \hbar)^{-1} \cdot \sinh \left( \frac{\hbar |\omega| - (\mu_e - \mu_h)}{2k_B T} \right)}{\cosh \left( \frac{\mu_e + \mu_h}{2k_B T} \right) + \cosh \left( \frac{\mu_e - \mu_h}{2k_B T} \right)}$$

(1)

here $\gamma = 1/\tau$, $\tau$ is the electron relaxation time in carbon nanotubes, $e$ is the electron charge, $g_e$, $g_v$ are the spin and valley degeneracies, $\mu_e$, $\mu_h$ are the electron and hole chemical potentials, respectively, $\omega$ is the electromagnetic frequency, $\varepsilon$ is the energy spectrum of carbon nanotubes, $T$ is the temperature, $k_B$ is the Boltzmann constant.

Further, following the Joule-Lenz law, we calculate the amount of heat $Q$ when an electric current flows through an array of carbon nanotubes:

$$Q(T) = \int_0^T I^2 R \, dt$$

(2)

where $I$ is the current density, $R$ is the resistance.

The electric current can be found as:

$$I(t) = \frac{L}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \sigma(\omega) \cdot E(\omega) \cdot e^{i\omega t} \, d\omega$$

(3)

here $\sigma(\omega)$ is determined according to formula (1), $L$ is the nanotube length, $E(\omega)$ is the Fourier transform of the electric field of the pulse, moreover, $E(t) = e^{i\omega t} \partial A / \partial t$, $c$ is the light velocity, $A$ is the vector potential of the electric field. Here we consider the electromagnetic pulse propagating along the axis of the nanotubes and the vector potential having only one non-zero component.

Note that we neglect the conductivity changes due to the CNT temperature changes. This is associated with the typical values of chemical potentials. Namely, the change in the arguments of exponential functions that determine the temperature dependence (1) occurs in the second decimal place.
3. Results

The initial pulse shape is chosen as a Gaussian pulse (4a) and as a "light bullet" (4b):

\[ A(t) = A_0 \exp \left( -\frac{t^2}{\Delta^2} \right) \] (4a)

\[ A(t) = A_0 \exp \left( -\frac{t^2}{\Delta^2} \right) \cdot \cos(\nu t) \] (4b)

here \( A_0 \) is the pulse amplitude, \( \Delta \) is the pulse duration, \( \nu \) is the pulse frequency.

Figure 1b demonstrates the influence of the shape of a few-cycle optical pulse on the amount of heat.

Figure 1. (a) The scheme of the CNT system; (b) The dependence of the amount of heat on the temperature: curve 1 – "light bullet"; curve 2 – Gaussian pulse. The non-dimensional unit of \( Q \) corresponds to 10 kJ, the non-dimensional unit of \( T \) corresponds to 1 K.

Figure 1b shows that the shape of the pulse has a significant effect on the heating of the carbon nanotubes array. We observe the greatest amount of heat when CNTs are irradiated with a Gaussian pulse. This fact is associated with such characteristics of the few-cycle optical pulse as its localization and stability. We can conclude that for the most successful heating of a CNT array, it is compulsory to use pulses with the largest spectral width at some given duration. The central frequency of the latter depends mainly on the chemical potential, which in turn is determined by the structure of the CNT and adsorbed impurities.

Let us consider the amount of heat \( Q \) depending on time using the example of a Gaussian pulse (Figure 2):

\[ Q(t) = \int_{-\infty}^{\infty} \sigma(\omega) \cdot E(\omega)^2 \cdot e^{i\omega} d\omega \] (5)

We use a Gaussian shape because it is easier to get in practice and more stable than a "light bullet".
Figure 2. The amount of heat $Q$ depending on time at $T=200$ K for the Gaussian pulse: (a) at different pulse duration $\Delta$ (curve 1 – $\Delta=2.5\cdot10^{-15}$ s; curve 2 – $\Delta=5.0\cdot10^{-15}$ s; curve 3 – $\Delta=7.5\cdot10^{-15}$ s); (b) at different inverse of the relaxation time $\gamma$ (curve 1 – $\gamma=0.5\cdot10^{-11}$ s$^{-1}$; curve 2 – $\gamma=1.0\cdot10^{-12}$ s$^{-1}$; curve 3 – $\gamma=1.5\cdot10^{-12}$ s$^{-1}$). The amount of heat $Q$ is scaled in kJ, time $t$ is scaled in $10^{-11}$ s.

Figure 2 shows that the heating of the CNT array significantly depends on the pulse duration and the electron relaxation time. This is due to the interband and intraband absorption of laser pulse photons by electrons. In addition, increasing the pulse duration leads to an increase in the amount of radiated heat. An increase in the relaxation time (e.g., due to the inclusion of impurities) leads to decrease in heating, which reflects the fact that at a long relaxation time the pulse weakly interacts with the CNT, which leads to less heat release.

Conclusions
The key results obtained may be summarized as follows:

- The model of hot electrons proposed in the work [15] allows predicting the carbon nanotubes heating dynamics using few-cycle optical pulses.
- Due to the change in the initial shape of the few-cycle laser pulse, it is possible to control the amount of heat in carbon nanotubes.
- The duration of the electromagnetic pulse and the relaxation time of electrons in carbon nanotubes are the key factors affecting the CNT array heating.

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