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Phonon-assisted tunnelling in electrical conductivity of individual carbon nanotubes and networks ones

P. Pipinys, A. Kiveris *

Department of Physics, Vilnius Pedagogical University, Studentų 39, LT-08106 Vilnius, Lithuania

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ABSTRACT

Current–voltage (I–V) characteristics of single-wall carbon nanotubes (SWCNT), measured in the low temperatures by Tang et al. [Science 292 (2001) 2462] and transparent SWCNT networks presented by Jaiswal et al. [J. Phys.: Condens. Matter 19 (2007) 446006], are reinterpreted in the framework of phonon-assisted tunnelling theory as a free charge carriers generation mechanism in the strong electrical field. The good fit of the temperature-dependent I–V data in low temperature region (i.e., $T<25$ K) has been achieved using the phonons of energy <1 meV.

1. Introduction

In the last decade, extensive studies have been carried out on the electrical conductivity of carbon nanostructures or individual nanotubes [1–17]. Some peculiarities, such as nonlinear behaviour of electrical conductivity, temperature variation of current–voltage (I–V) characteristics, and a crossover of conductivity from semiconducting-like to metallic-like temperature dependence, have been observed. Kaiser et al. [2] pointed out similarity between the resistivity temperature behaviour $\rho(T)$ observed in SWCNTs and that of highly conducting polymers, in particular the change in sign of the $\rho(T)$ dependence from metallic to non-metallic as the temperature was decreased.

Several models have been invoked for explanation of observed peculiarities of conductivity in carbon nanotubes. Frequently, the model of variable range hopping (VRH) is used to describe the temperature dependence of the conductivity [3,5,8,13,15,16].

According to the VRH mechanism, the dependence of $\rho(T)$ is described by the formula

$$\rho(T) = \rho_0 \exp(T_0/T)^{1/(n+1)}, \quad (1)$$

where n is the dimension of the system and T_0 is a fitting parameter. The reports of different authors vary in their findings on the dimensionality of the VRH, ranging from one to three dimensions. There are a number of publications in which the Luttinger liquid (LL) model is used for explanation of temperature behaviour of the conductivity in SWCNT [7,11,12,14,17]. However, the discrepancy between predicted values of the power-law exponents in equations for current and conductivity from the theory and derived from the experimental data is often observed [12,14].

Moreover, the authors of Ref. [8], upon varying the temperature from 4.2 to 295 K, envisaged four transport mechanisms in self-organized carbon networks.

A plenty of mechanisms used for explanation of the temperature dependence of current peculiarities in SWCNT networks imply that the conduction mechanism in these materials is not fully understood. In our previous publication [18], some results of current dependence on temperature, including the crossover from a semiconducting-like temperature dependence conductivity to a metallic-like one as temperature increases, has been interpreted based on the phonon-assisted tunnelling (PhAT) model, which has been previously used for explanation of temperature-dependent I–V data in some polymers [19,20].

The main purpose of this paper is to show that nonlinear I–V characteristics and their temperature variation, measured for

* Corresponding author. Tel.: +370 5 279 0036; fax: +370 5 279 0548.
E-mail addresses: pipiniai@takas.lt (P. Pipinys), studsk@vpu.lt, akiveris@vpu.lt (A. Kiveris).

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SWCNT networks and single nanotubes in the region of low temperatures, as well as conductivity dependence on temperature in semiconducting SWCNT extracted from recent publications [4,15,16], are explicable in the framework of the field-induced PhAT model. Since the experimental data under analysis have been obtained in the low-temperature region (4–25 K) to describe the results, the phonons of low energy (about 1 meV) were requested.

2. PhAT model and comparison with experimental data

It is assumed that a source of charge carriers is the local electronic states in the nanotube–electrode interface layer, electrons from which emerged to the conduction band of the nanotube crystal due to the PhAT from these centres.

If electrons released from these centres originate the current I in the nanotube, it will be proportional to the electron release rate W and the density of the centres N, i.e. $I \propto NW$. Whereas PhAT rate is a function of field strength E and temperature T, the current through the sample may be the same function of these parameters. This factor enables to compare the temperature and field dependence of current with the tunnelling rate W dependence on these parameters. For the calculation of the $W(E, T)$, the equation derived within the effective mass approximation on the assumption that the centre absorption band is described by the Gaussian law is used [21]:

$$W = \frac{eE}{(8m^*\varepsilon_T)^{1/2}}[(1+\gamma^2)^{1/2}-\gamma]^{1/2}[1+\gamma^2]^{-1/4} \times \exp\left\{-\frac{4}{3}\frac{(2m^*)^{1/2}}{eE\hbar}\varepsilon_T^{3/2}\left[(1+\gamma^2)^{1/2}-\gamma\right]^2[(1+\gamma^2)^{1/2}]+\frac{1}{2}\gamma\right]\right\}$$
$$\gamma = \frac{(2m^*)^{1/2}\Gamma^2}{8e\hbar E\varepsilon_T^{1/2}}. \quad (2)$$

Here $\Gamma^2 = 8a(\hbar\omega)^2(2n+1)$ is the width of the centre absorption band, $n = [\exp(\hbar\omega/k_BT)-1]^{-1}$, where $\hbar\omega$ is the phonon energy, ε_T is the energetic depth of the centre, e is electron charge unit and a is the strength of the electron–phonon interaction ($a = \Gamma^2_{T=0}/8(\hbar\omega)^2$). Consequently, we will use this equation to explain the peculiarities of the current dependence on temperature and field strength.

I–V characteristics variations with temperature in the low-temperature region of carbon nanotube networks have been presented in Refs. [4,15,16] and for individual nanotubes in Refs. [3,4,14]. The main peculiarities of these dependences are their nonlinearity at higher applied voltages and strong dependence on temperature. Such behaviour of I–V data is explicable in the framework of phonon-assisted model. In order to prove this proposition at first, we present comparison of I–V characteristics measured by Tang et al. [4] for single-walled carbon nanotubes embedded in zeolite crystals. The measurement was carried out in the low-temperature region from 0.33 to 18.5 K [4]. The fit of the experimental data from [4, Fig. 2 (inset)] with the theoretical $W(E, T)$ dependences computed using Eq. (2) are shown in Fig. 1. The calculation was performed using the value of $0.2m_e$ for the electron effective mass [22]; for the phonon energy the value of 0.8 meV was chosen. We want to note that there exist a grand variety of SWCNT vibrational modes in the energy range from about 1 to 200 meV [22–24,29,30], and that the phonons of various energies may take part in the tunnelling process. However, at low temperatures only phonons of small energy can be effective because a population of phonons of higher energy would be negligible. As was pointed out in Ref. [24], the low-energy phonons (i.e. 0–1.5 meV) may play an important role in the temperature dependence of the electronic conductivity. The

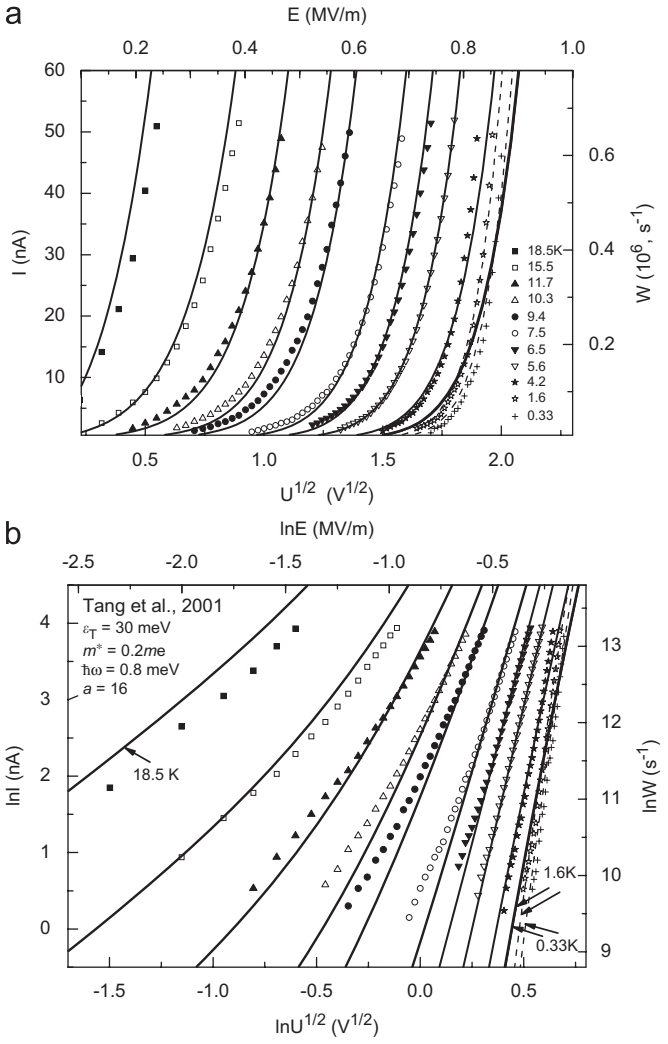

Fig. 1. The I–V data for single SWCNTs measured by Tang et al. [4] in the temperature range from 0.33 to 18.5 K (symbols). In the temperature range of $4.2\,K < T < 18.5\,K$ the best fit gives $W(E, T)$ curves, calculated for the phonon energy of 0.8 meV ($a = 16$) (solid lines), while for the curves of 0.33 and 1.6 K the phonons of 0.2 meV energy are more suitable (dashed lines), $\varepsilon_T = 30$ meV. (a) In the scale of I vs. $\sqrt{V}$ plot; (b) in the scale of $\ln I$ vs. $\ln\sqrt{V}$ plot.

electron–phonon coupling constant a was chosen so that the best fit of the experimental data with the calculated dependences should be received. It is known that in the case of a semiconducting carbon nanotube the Schottky barriers are easily formed between metal contacts and carbon nanotubes [12,25–28]. Therefore, the fit of experimental data with $W(E, T)$ dependences was done on the assumption that the field strength at the junction is proportional to the square root of the applied voltage, i.e. the tunnelling occurs in the high-field region of the Schottky barrier.

As is seen in Fig. 1, a good match of experimental data with the computed $W(E, T)$ dependences (solid lines) is observed in the temperature range from 4.2 to 18.5 K. The dependences of I–V measured at lower temperatures, 0.33 and 1.6 K (symbols), are more steeper then the theoretical dependences and are deflected towards higher-field region. A reason for this discrepancy between computed curves and experimental data may be the factor that at lower temperature region the phonons of lesser energy could take part in the process of tunnelling. Indeed, the theoretical dependences (dashed lines) calculated for the phonons of lesser energy (i.e. 0.2 meV) successfully describe these experimental curves.