Electrical Transport and Magnetoresistance in Single-Wall Carbon Nanotubes Films

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Electrical transport properties and magnetoresistance of single-wall carbon nanotubes (SWCNT) films were investigated within temperature range (2-300) K and in magnetic fields up to 8 T. A crossover between metallic (dR/dT > 0) and non-metallic (dR/dT < 0) temperature dependence of the resistance as well as low-temperature saturation of the resistance in high bias regime indicated on the diminishing of role of the contact barriers between individual nanotubes essential for the charge transport in SWCNT arrays. The magnetoresistance (MR) data demonstrated influence of weak localization and electron-electron interactions on charge transport properties in SWCNT films. The low-field negative MR with positive upturn was observed at low temperatures. At T > 10 K only negative MR was observed in the whole range of available magnetic fields. The negative MR can be approximated using 1D weak localization (WL) model. The low temperature positive MR is induced by contribution from electron-electron interactions.

Keywords: carbon nanotubes, charge transport, magnetoresistance, weak localization, electron-electron interaction.

1. INTRODUCTION

Among number of excellent properties (physical, chemical, mechanical etc.) of single walled carbon nanotubes (SWCNT) electrical transport in these unique structures is of particular importance both for development of fundamental condensed matter theories and for possible applications. However most of the realistic applications of electrical properties of SWCNT, such as interconnects in integrated circuits, transparent electrodes for solar cells, gas-, bio- and chemical censors etc., assume necessity of fabrication of SWCNT assembles with high conductivity [1]. Charge transport in different types of SWCNT arrays (networks, films, mats, fibers) is intensively investigated as far as verification of conductivity mechanisms in these structures is a crucial task for utilizing of their electron transport properties in devices and sensors. It was found that electrical properties of SWCNT assembles are strongly depended on the quality of contacts between individual nanotubes [2]. Different conductivity mechanisms were observed in SWCNT arrays: metallic conductivity [2], fluctuation induced tunneling [2-4], variable-range hopping [4, 5], weak localization (WL) [5]. Conductivity of SWCNT arrays can be enhanced by alignment of nanotubes inside of assembles and by increasing of length of individual nanotubes. In case, when the role of contacts barriers between nanotubes is diminished, phenomena inherent for individual SWCNT (Luttinger liquid behaviour [6], quantum corrections to conductivity due to WL and electron-electron interactions [7]) are observed in SWCNT assembles.

In this paper we present charge transport properties of SWCNT films in which enhancement of their conductivity is induced by two main factors: functionalization of SWCNT and using of contact plates with long electrodes separated by small distance which is comparable with average length of nanotubes.

2. EXPERIMETAL DETAILS

The samples used in this work were produced from SWCNT dispersed in 1 % solution of sodium dodecylsulfate (SDS) surfactant in deionised water. SWCNT with diameter within range of (1.2-1.5) nm and average length of $(1-3) \mu m$ grown by arc-discharge method in the presence of Ni-Y catalyst (manufacturer "CarboLex, Inc.") were used as pristine material for preparation of carbon nanotubes films. SWCNT were stuck in bundles with diameters (3-7) nm and average length 3.5 µm. Sonication for 4 hours at 44 kHz and at 70 W⁻cm⁻² ultrasonic intensity and subsequent centrifugation for 1.5 h at 900 g was applied for homogeneous SWCNT' suspension preparation. Functionalization of SWCNT in SDS is used for homogeneous distribution of nanotubes in solutions and for preventing of their aggregation due to strong van der Waals binding energies [8]. A droplet of suspension was deposited on the glass substrate with lithographically defined four Pt 3 mm long and 5 µm wide contact electrodes purchased from "ABTECH Scientific Inc.". Distance between contact electrodes was 5 µm. Space outside of contact area was covered by insulating layer Si₃N₄. Substrate was heated at 100 °C during SWCNT' suspension deposition for rapid evaporation of an aqueous solution of sodium-dodecyl-sulfate.

The transport properties of SWCNT films were measured using DC technique. The temperature dependences of the resistance R(T), magnetoresistance R(B) and IV-characteristics were measured in the temperature interval (2–300) K at applied voltage up to 5 V and in magnetic fields up to 8 T at the Physics Department of Belarusian State University using helium closed-cycle refrigerator "Cryogenics Limited Inc.".

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3. RESULTS AND DISCUSSION

The data of the temperature dependence of resistance of the SWCNT films measured at different applied voltage (0.1 V, 0.5 V and 1 V) are plotted in Fig. 1. As one can see from the right inset to Fig. 1, in the high temperature range R(T) dependences show inherent to metallic systems behavior with positive temperature coefficient of the resistance (dR/dT > 0) displaying minimum which is shifted to the lower temperatures as the applied voltage was increased. Negative temperature coefficient of the resistance (dR/dT < 0) was observed for R(T) dependences in the low temperature range. The temperature of the crossover between metallic and non-metallic type of the R(T) dependences was found to be shifted from 213 K at 0.1 V to 158 K at 1 V. Thus, enhanced metallic behavior is clearly observed on the R(T) curves as the applied voltage was increased. In contrast to SWCNT mats [9] lowtemperature saturation of resistance in our SWCNT films was observed even at low bias voltage. In difference from the high bias region, where saturation of RT dependences is induced by decreasing of the height of contact barriers between nanotubes, saturation at low bias can be also explained by low temperature saturation of dephasing time inherent for weakly disordered systems [10].



Fig. 1. The temperature dependence of the resistance R(T) of the SWCNT films in semilogarithmic scale at different applied voltage (1 - 0.1 V, 2 - 0.5 V, 3 - 1 V). Solid line show fitting results of the R(T) dependence of the SWCNT films in the range (10-125) K from the equation (1) for the fluctuation-induced tunneling model. The right inset figure: R(T) dependence of the SWCNT films in the range (130-300) K. The left inset figure: schematical image of contact electrodes' configuration

In the intermediate temperature range (10 K - 125 K) at low bias voltage R(T) dependences can be approximated by following equation inherent for thermal fluctuation-induced tunneling (FIT) model [11]:

$$R = R_0 \exp(T_1 / T + T_0), \qquad (1)$$

where T_1 according [11] is the temperature below which the conduction is dominated by the charge carrier tunneling through the barrier and T_0 is the temperature above which fluctuation effects become significant.

The current-voltage characteristics measured within temperature range (2-300) K are plotted in Fig. 2. At high temperatures non-linear *IV*-curves are well fitted by the equation obtained by Kaiser et al. [12] by means of

$$I = G_0 V \exp(V / V_0), \qquad (2)$$

where parameter G_0 is the low-field conductance, V_0 is the scale parameter. At T < 75 K discrepancy between experimental data and fitting curves was observed. Analysis of non-linear electrical properties of SWCNT films in strong electric fields within frame of Luttinger liquid behavior will be reported elsewhere.



Fig. 2. The current-voltage characteristics of the SWCNT films measured at different temperatures. Solid lines are fitting results of the *IV*-characteristics from the eq. (2)

The dependences of the normalized change of the resistance of SWCNT films on the magnetic field (R(B) - R(B=0)/R(B=0))measured at different temperatures and reconstructed from them for higher temperatures (15 K - 50 K) dependences of the normalized change of the conductance (G(B) - G(B=0)/G(B=0)) on magnetic field are shown in Fig. 3, a, and Fig. 3, b, respectively. At low fields, MR is negative. Positive upturn was observed on the MR curves, which shifted to the high field's values with temperature increase. The upturn field of the MR effect was shifted from 5.6 T at 2 K to value of about 6.8 T at 8 K. At T > 10 K only negative MR was observed in the whole range of experimentally available magnetic fields (0-8) T. As one can see from Fig. 3, b, positive magnetoconductance (corresponding to negative MR) of SWCNT films follows typical for 1D WL (case $L_{\varphi} > w$) behavior [13]:

$$G(B) = G_0 - \frac{e^2}{\pi \hbar L} \left(\frac{1}{L^2_{\varphi}} + \frac{(eBw)^2}{3\hbar^2}\right)^{-1/2},$$
(3)

where G_0 is the conductance in zero magnetic field, \hbar stands for Planck constant, *e* being the electronic charge, *L* is the contact spasing, *w* labels diameter of the system, L_{φ} denotes phase coherence length of charge carriers. Assuming that *w* corresponds to the diameter of nanotubes, we found that L_{φ} varies from 16.6 nm (at 50 K) to 29.8 nm (at 15 K).

The positive MR (increase of the resistance with the rising of magnetic field) observed at very low temperatures and at high magnetic fields can be explained by influence of electron-electron interactions on charge transport. This contribution to MR data is usually observed in case when the energy difference between the Zeeman split levels is much more than the thermal energy $(g\mu_B H >> k_B T)$. As a result the electron diffusion becomes much slower and electron-electron interactions, that induces lowering of the conductivity, should be taken into account [7, 14]. For the detailed analysis of positive magnetoresistance of SWCNT films measurements in higher magnetic fields are requested.



Fig. 3. The dependence of the normalized change of the resistance R - R(B = 0)/R(B = 0) (a) and of the normalized change of the conductance G - G(B = 0)/G(B = 0) (b) of the SWCNT films on the magnetic field measured at different temperatures. Solid lines are fitting results of the G(B) dependence from the equation (3) for 1D WL

As one can see from the analysis of electrical and magnetotransport properties of SWCNT films, both mechanism of fluctuation-induced tunneling and quantum corrections to the conductivity due to weak localization and electron-electron interactions were observed in our samples. Geometry of contact structures (length of the contact strips is several orders of magnitude higher than the distance between them: 3 mm vs 5 μ m), functionalization of nanotubes in SDS solution and fact that average length of individual tubes (1 μ m-3 μ m) is comparable on the value with the distance between contacts assumes that in our samples a large number of SWCNTs connected in parallel. Therefore ifluence of contact barriers between nanotubes connected in series is diminished.

4. CONCLUSIONS

It is shown that electrical properties and magnetoresistance of SWCNT films can be explained both by thermally activated tunnelling of charge carriers between individual nanotubes and by quantum corrections to conductivity due to 1D weak localization and electron-electron interactions.

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