CHARGE TRANSPORT MECHANISMS IN THE ARRAYS OF MULTI-WALLED CARBON NANOTUBES

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INTRODUCTION

Electronic and magnetotransport properties of carbon nanotubes have attracted much attention due to they importance in verification of existing theories of modern condenced matter physics [1, 2] and number of possible applications [3, 4]. Single-walled carbon nanotubes (SWCNTs) have unique structure and show metallic or semiconducting properties in dependence of their diameter and chirality. Multi-walled carbon nanotubes (MWCNTs) are more complicated systems. They consist of a several shells of different diameter and chirality. Due to weak coupling between the shells the conductivity in a bulk-contacted MWCNTs is defined mostly by the outermost shells. For possibility of commercial manufacturing of carbon nanotubes based sensors and devices the development of fabrication methods of carbon nanotube arrays is of great importance. The intertubes barriers and defects play an essential role in the electrical transport properties of the carbon nanotube arrays. Therefore different charge transport mechanisms can be observed in the arrays of nanotubes: metallic conductivity, variable range hopping (VRH), weak localization (WL), fluctuation induced tunneling. Combination of various mechanisms is possible as well. Different types of morphology of carbon nanotubes arrays are proposed: bundles, networks, fibers, mats etc. We present here magnetotransport properties of the thing layers of MWCNTs.

EXPERIMENTAL DETAILS

MWCNTs were produced by Chemical Vapour Deposition method. Langmuir-Blodgett technique was used for deposition of MWCNTs arrays on the substrates with electrodes for the measurements of magnetotransport properties. Previous chemical functionalization procedure of pristine carbon nanotubes was applied that gives us possibility to obtain dense arrays of MWCNTs.

The magnetotransport properties of the MWCNTs arrays were measured using standard four-probe lock-in technique. The temperature dependencies of the resistance were measured in the temperature range of 4.2–300 K. Magnetoresistance (MR) measurements were carried out in pulsed magnetic fields at the *Laboratoire National des Champs Magnetiques Pulses de Toulouse (LNCMP)* in the wide temperature range and up to 40 T magnitude fields.

RESULTS AND DISCUSSION

The temperature dependence of the resistance shows the negative temperature coefficient of the resistance ($dR/dT<0$) in the whole investigated temperature range (4.2-300 K) as one can see in the Fig.1. Assuming possibility of the VRH conduction in our system due to structural defects and impurities, we fitted $R(T)$ dependence by classical law for VRH [5]:

$$R = R_0 \exp(T_0 / T)^{1/n} , \quad (1)$$

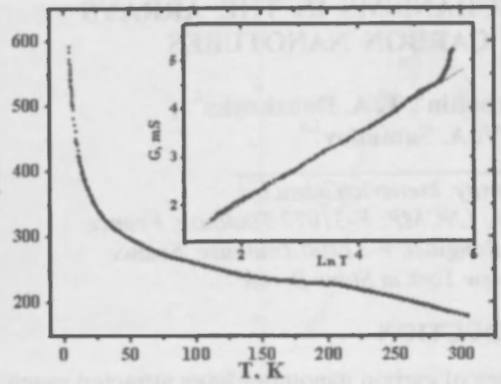

Fig.1. The temperature dependence of the resistance of the MWCNTs arrays. The inset shows the *R(T)* dependence in *G-LnT* scale

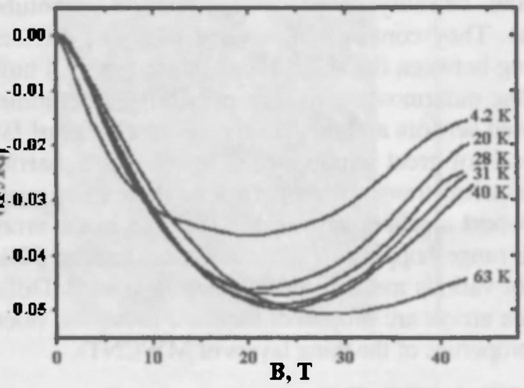

Fig.2. The dependence of the normalized change in the resistance *dR/R(B=0)* f the MWCNTs arrays on the magnetic field in the temperature range 4.2–63 K

where T_0 is a constant, depending on the density of states and localization length of the system, $n=d+1=2,3,4$ (d is the dimensionality of the system). In order to find parameter n we used both linearization in the scale $logR\text{-}T^{1/n}$ and method proposed by Zabrodskii [6]. We found that *R(T)* dependence can be approximated by Eq. (1) in the temperature range T=4.2~60 K and the best fitting results were obtained at T below 60 K with parameter $n=3$, indicating 2D VRH.

In order to see suitability of the WL theory for our system, we tried to fit *R(T)* dependence with one-, two- and three-dimensional WL formulas. We found that *R(T)* dependence follows the 2D weak localization (WL) behavior (*G(T)~lnT*) [7] in the temperature range 4.2~202 K as one can see from the inset to Fig.1. In the temperature range 202-300 K conductivity increases approximately linearly with temperature. MR data measured at various temperatures are plotted in Fig.2. The negative MR in the low magnetic fields range was observed. In the low-temperature range upturn of negative MR was observed. The minimum position of negative MR shifts to higher fields as the temperature rises. Both low-field negative MR due to changing of phase between alternate hopping paths enclosing a magnetic flux [8, 9] and high-field positive MR due to electronic orbit shrinkage [5] are predicted for systems with hopping conductivity mechanism. However, monotonic decrease of relative value of negative MR with the temperature rising are usually observed. In our samples the opposite situation is clearly seen: relative value of negative MR increased with the temperature rising in the low temperature range where VRH can be responsible for the charge transport mechanism. From the other side negative MR is inherent for the systems where conductivity can be described in the frame of WL theory [7].

Therefore we made assumption that MR data are the sum of the positive and negative contribution due to MR effects in the VRH and WL regimes, respectively. We found that high-field positive part of MR can be approximated in frame of Kamimura model for spin-dependent VRH conductivity:

$$\frac{\Delta G}{G} = -A_{KK}\,\frac{H^2}{H_{KK}^2 - H^2}, \qquad (2)$$

where H_{KK} is the characteristic field for spin alignment, A_{KK} the saturation value of the magnetoconductance. Using formula (2) and values of parameters H_{KK} and A_{KK} obtained from the approximation data of high field MR data, we calculated positive magnetoconductance for low-field region. The fitting curves for positive MR are shown in Fig.3, *a*. Pure negative contribution to MR according our assumption was calculated by subtracting positive MR from the experimental data and shown in Fig.3, *b*. These data can be fitted reasonably well by the Eq. (3) for 2D WL:

$$\Delta G = \frac{e^2}{\pi h}[\psi(\frac{1}{2}+x)+ln(x)], \quad (3)$$

where ψ is the digamma function, $x=L_{Th}^2\ 8\pi EH/hc$, $L_{Th} \sim T^{p/2}$ is the Thouless length.

With this assumption we can explain positive upturn observed on MR curves by adding (*a*) and (*b*) from the Fig. 3. Positive MR may come from the spin-dependent VRH among the defects on the surface of nanotubes. The negative contribution to MR can originate from the WL effects.

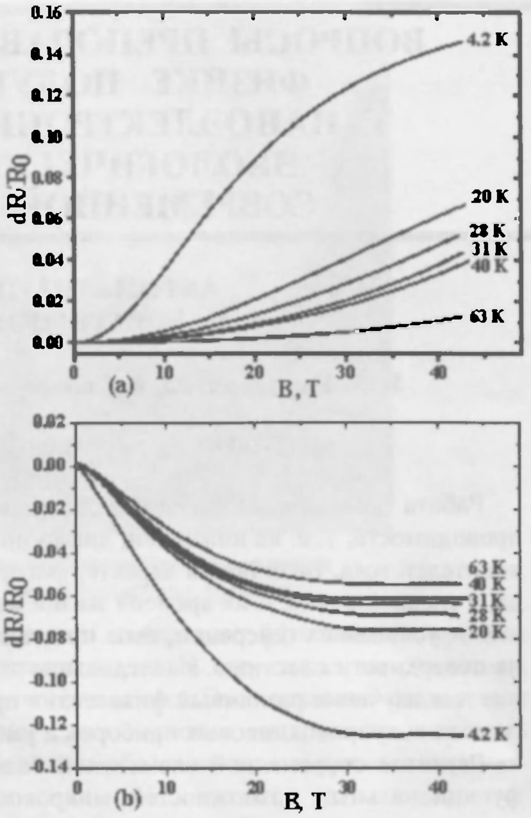

Fig.3. (*a*) Calculated value of positive MR obtained by fitting positive part of MR data to formula (2). (*b*) The negative MR obtained by subtracting the calculated positive MR (*a*) from the experimental MR data

CONCLUSIONS

We have measured magnetotransport properties of MWCNTs arrays in a wide temperature range and up to 40 T. The temperature dependence of resistance was found to follow Mott law for 2D VRH in the temperature range 4.2~60 K and typical law for 2D WL in the temperature range 4.2~202 K. The magnetotransport properties were explained in assumption of positive and negative contribution to MR originating from spin-dependent VRH and 2D WL, respectively.

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