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Magnetotransport of carbon nanotubes: *Magnetic-field-induced metal-insulator transition*

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We have measured magnetotransport of an individual multi-walled carbon nanotube. Though the resistance without magnetic field increases with decreasing temperature (non-metallic) from room temperature down to 2 K, it becomes metallic below 30 K by applying a magnetic field ($H \fallingdotseq 4$ T) perpendicular to the nanotube axis. The transverse magnetoresistance (TMR) below 30 K decreases with increasing magnetic field, and after reaching minimum value around 4 T, it increases. On the other hand, under the intense magnetic field above 6 T the TMR does not show continuous increase but tends to be saturated. The results can be explained by the theoretical prediction of the magnetic-field-induced metal-insulator transition of semiconducting carbon nanotubes.

keywords Carbon nanotubes, magnetotransport, metal-insulator transition

Introduction

Since the discovery of the carbon nanotube (CNT) [1], it has attracted great attention as a very interesting electronic material because of the one-dimensional (1D) structure and the tubular honeycomb-network in the nanometer scale[2-4]. Theoretical studies of the CNT [5-8] predict some novel electronic properties such as the 1D band-structure characteristic due to the periodic boundary conditions in the circumferential direction and the magnetic quantum effect. The band structure of the CNT is either metallic or semiconductive depending on the chirality and the diameter of the tube; the energy gap (E_g) of semiconductor phase is inversely proportional to the diameter. Some experimental works confirm these electronic structures [9-11] and the magnetic quantum effect[12,13].

On the other hand, in many transport measurement results CNTs do not show typical metallic or semiconducting behavior. In particular, diameter or circumference of multi-walled carbon nanotubes (MWCNTs) is comparable to the typical length in mesoscopic physics, *i.e.*, the mean free path, the phase coherence length and the magnetic length. Therefore, it is reasonable that transport properties of MWCNTs do not show ideal 1D conducting behavior, and a wide variety of physical properties is expected.

In this study, we have measured the transverse magnetoresistance (TMR) of a MWCNT. The positive temperature dependence of resistance appeared under magnetic field of $H = 4$ T below 30 K, whereas the negative temperature dependence of resistance were observed under low magnetic fields $H < 1$ T and intense magnetic fields H $\fallingdotseq$ 7 T. The observed behavior in TMR is ascribed to the successive metal-insulator transition predicted by band structure calculation with taking account of elastic/inelastic scattering of electrons[14].

Experimental

MWCNTs were obtained from carbon soot (Type I, Vacuum Metallurgical Co. Ltd., Japan) by repetition of physical purification processes, that is, the centrifugation at 5000 r.p.m. for 20 min. and filtration; we did not use any chemical process such as oxidation treatment, because these processes may lead serious damages on the surface of MWCNTs which act as electronic scattering centers.

In this work, we measured the resistance of a single MWCNT by using directly attached electric contacts, as shown in a SEM image (Fig. 1). The contacts were made by the electron-beam-lithography technique in the following processes. MWCNTs, which were ultrasonically dispersed in methyl alcohol, were placed onto a substrate of oxidized Si-wafer. After spin-coated with a positive photoresist, the lead pattern of contacts with 1.25 µm pitch and 10 µm in length was drawn by an electron beam. The exposed pattern was removed by a solvent, on which gold was deposited.

The d.c. MR measurements were carried out by using

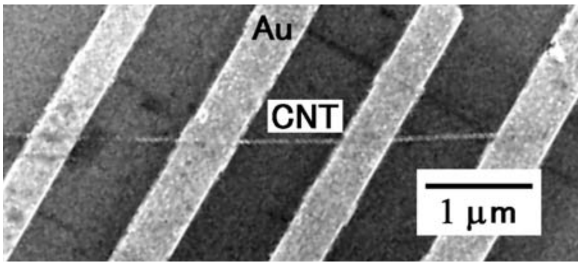

Fig.1 SEM image of a multi-walled carbon nanotube and gold contacts for transport measurements. The stripe shaped gray areas are gold leads and a fine line presents a nanotube.

Quantum Design PPMS system with a rotating sample-mount. The measurements were performed in the ohmic region, usually in the region from 5 to 30 nA. We will present the experimental results for a sample with 190 Å in outer diameter and 1.0 μm in voltage-contact distance of the sample. The inner diameter is estimated as about 30 Å from the TEM observation of samples in the same lot. In the present case we have not found out any mechanical stress coming from bending, defects or flexure reported previously[4,15,16], although MR occasionally jumps irreversibly in the region of very high fields, which might arise from a displacement of sample due to a strong galvanomagnetic force.

Results and discussion

Figure 2 shows the temperature dependence of resistance $R(T)$ of a single MWCNT. At the zero magnetic field, resistance increases with decreasing temperature. This temperature dependence is often observed in measurement on individual MWCNTs, and is interpreted as *ln* T dependence resulting from the weakly localization effect of metallic nanotubes[13,17]. On the other hand, under magnetic fields perpendicular to the nanotube axis, the degree of negative slope of $R(T)$ decreases below about 30 K. At the magnetic field of 4 T, resistance shows metallic behavior, $dR(T)/dT > 0$, from about 30 K down to 2 K. By applying further magnetic fields, metallic behavior is weakened and the degree of negative slope of $R(T)$ is enhanced again. This non-monotonic dependence of $R(T)$ on the magnetic field cannot be explained only by the suppression of the localization.

Effect of the magnetic fields on the transport property can be clearly seen in the transverse magnetoresistance as shown in Fig. 3(a), in which

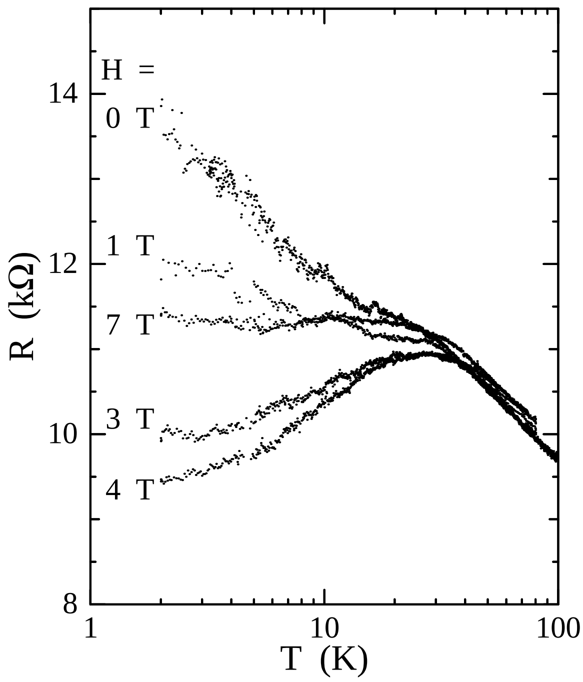

Fig. 2 Temperature dependence of resistance of a single multi-walled carbon nanotube under magnetic fields perpendicular to the nanotube axis.

data are normalized by the resistance at zero magnetic field. Below 30 K, resistance decreases with increasing magnetic field up to $H \fallingdotseq 4$ T, followed by the increasing. The minimum values of TMR at $H = 4$ T correspond to metallic behavior observed in the temperature dependence of resistance in Fig. 2. More noteworthy is that the TMR tends to be saturated at intense magnetic fields at $H \fallingdotseq 7$ T. This tendency can be confirmed by the derivative of TMR data at 2 K as shown in Fig. 3(b); $dR(H)/dH$ shows maximum value around $H = 6$ T, and then decreases toward zero.

In the band structure calculation in which CNT is considered to be ideal ballistic conductor, a Landau level forms at the Fermi level and then the density of states (DOS) increases under magnetic fields perpendicular to the nanotube axis[8]. In this picture, conductance decreases significantly because the effective mass increases owing to the formation of the flat band of the Landau level. On the other hand, Roche and Saito[14] predict that the metal-insulator transition under magnetic fields perpendicular to the nanotube axis by taking account of elastic/inelastic scattering of electrons. The DOS at the Fermi level is predicted to have finite value both in metallic and

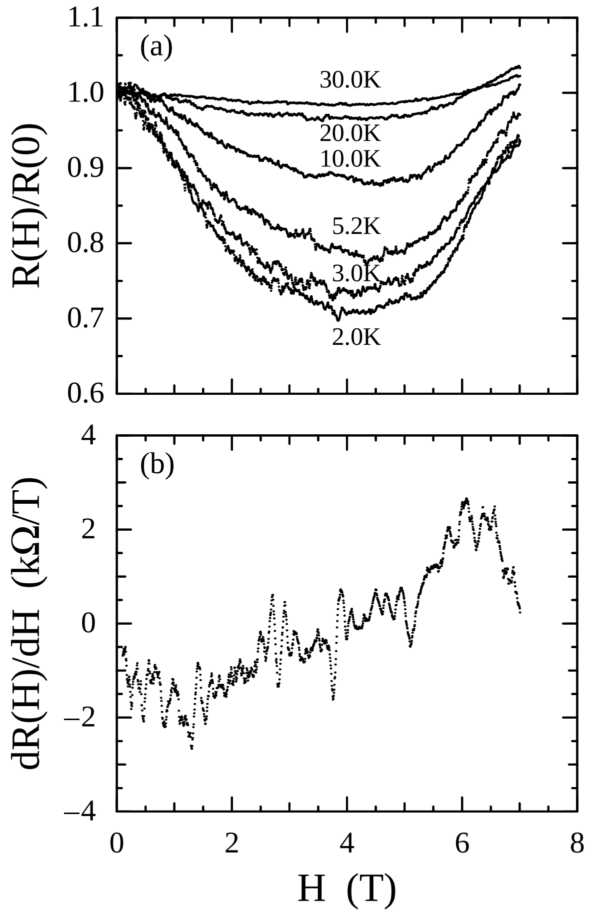

Fig.3 (a)Transverse magnetoresistance (TMR) of a single multi-walled carbon nanotube at various temperatures. (b)Derivative of TMR data $dR(H)/dH$ at 2 K.

"semiconducting" nanotubes and change oscillatory between metallic and semiconducting electronic states as a function of $\nu = L/2\pi l$, where L is the circumference of the nanotube, l is the magnetic length $(h/2\pi eH)^{1/2}$. In the case of metallic nanotubes, with increasing ν the DOS at the Fermi level decreases to the value which is almost the same to that of "semiconducting" nanotube at zero magnetic field. On the other hand, as ν increases the DOS at the Fermi level of "semiconducting" nanotubes increases to a maximum value around $\nu \fallingdotseq 0.7$ and decreases to the value being similar to that at $\nu = 0$ around $\nu = 1.2$. In these cases, the Landau level is not formed, and then effective mass of electrons hardly changes. Therefore we can expect that the conductance corresponds to the DOS at the Fermi level and that the metal-insulator transition and insulator-metal-insulator transition occur in metallic and "semiconducting" nanotubes, respectively.

Because many experimental results suggest diffusive transport for MWCNTs[12,13,17], it is meaningful to apply the theory by Roche and Saito to observation in this work. In the case of the MWCNT in this work, the value of $\nu = 0.7$ corresponds to $H = 3.6$ T, which is very close to the magnetic field at which the minimum value of TMR and the metallic behavior were observed ($H = 4$ T). The observed TMR is consistent with the behavior of "semiconducting" nanotube. The negative temperature dependence of resistance under low and intense magnetic fields corresponds to the "semiconducting" nanotubes with very small value of DOS at the Fermi level, which is considered to be situated near the metal-insulator transition. The metallic behavior at $H \fallingdotseq 4$ T corresponds to the magnetic field induced metallic phase of nanotube caused by the increase in DOS at the Fermi level. As a result, observed variations of $R(T)$ and TMR can be explained by magnetic field electronic phase transition taking account of diffusive conductance of CNTs.

According to this interpretation, the feature of CNT originating from the periodic boundary conditions vanishes above about 30 K, because the change in TMR due to the metal-insulator transition of CNTs is eliminated. This tendency is also observed in the Aharonov-Bohm effect of MWCNTs in the previous paper; the periodic oscillation in magnetoresistance under the magnetic fields parallel to the nanotube axis disappears above about 30 K[13]. These behaviors can be attributed to dimensional crossover with the competition between the coherence length and the circumference of the CNT. When the coherence length, which decreases with increasing temperature, becomes much shorter than the circumference of the CNT, the periodic boundary condition cannot contribute to the electronic structure of CNTs and the 1D feature of CNTs disappears.

In the viewpoint of this model, because even in the "semiconducting" nanotubes have finite value of DOS, the reason that the activation-type typical semiconducting $R(T)$ have been rarely observed can be explained. If this interpretation is correct, it is necessary to reconsider the transport properties of MWCNTs for true understanding. In addition, the tuning of carrier density by the magnetic fields will be very interesting subject to study. On the other hand, in spite of these advantages for explaining the experimental results by this theory, one question arises and remains to be solved. The observation in this work can be explained by the existence of only semiconducting nanotubes, whereas a MWCNT is consist of only metallic nanotubes or mixture of metallic and semiconducting nanotubes in the model explaining reported results so far.

Summary and conclusions

In transport measurements of a single multi wall-carbon-nanotube (MWCNT) with 190 Å in diameter, we have observed metallic behavior in transverse magnetoresistance around $H \fallingdotseq 4$ T below 30 K, whereas semiconducting-like behavior were observed for $H < 1$ T and $H \fallingdotseq 7$ T. The observation is ascribed to the successive metal insulator transition taking account of the diffusive nature of MWCNTs predicted theoretically by Roche and Saito. On the other hand, our results show further studies are still required for a clear understanding of the transport properties of MWCNTs.

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