PHYSICAL REVIEW B VOLUME 61, NUMBER 24 15 JUNE 2000-II

Magnetoresistance and differential conductance in multiwalled carbon nanotubes

Jeong-O Lee, Jae-Ryoung Kim, and Ju-Jin Kim
Department of Physics, Chonbuk National University, Chonju 561-756, Korea

Jinhee Kim, Nam Kim, Jong Wan Park, and Kyung-Hwa Yoo
Electricity Group, Korea Research Institute of Standard and Science, Taejon 305-600, Korea

Kang-Ho Park
Telecommunication Basic Research Laboratory, Electronics and Telecommunication Research Institute, Taejon 350-600, Korea

(Received 9 February 2000)

We have measured both the magnetoresistance and differential conductance of multiwalled carbon nanotubes as a function of magnetic field perpendicular to the tube axis. The measured differential conductance showed a large depletion of the density of states near the Fermi level and its magnitude was changed with a magnetic field. It was found that the aperiodic fluctuations and negative magnetoresistance mainly originate from the change of density of the states near the Fermi level with the magnetic field, rather than a quantum interference effect. At a particular magnetic field, the true metallic conduction along the outermost shell was observed, and the conductance approached the theoretical value, $2G_0 = 4e^2/h$, as the temperature was lowered.

Recently, there have been numerous reports on electrical transport properties in carbon nanotubes with the aid of nanofabrication techniques and novel contact methods.[1–5] Most of the magnetotransport studies on carbon nanotubes reported the appearance of negative magnetoresistance (MR) and reproducible resistance fluctuations, which were generally explained in terms of two-dimensional weak localization theory and universal conductance fluctuations based on the ''*diffusive*'' nature of conduction electrons.[2–5] A clear periodic oscillation in the MR with a magnetic field parallel to the tube axis was also observed and explained within the context of the well-known Al'tshuler-Aronov-Spivak effect.[6]

However, carbon nanotubes are predicted theoretically to be ideal ''*ballistic*'' conductors with only two occupied one-dimensional subbands, and the conductance should remain $2G_0(=4e^2/h)$.[7,8] Recently, Frank *et al.*[7] observed that a nanotube can conduct current ballistically with a quantized level of G_0 at room temperature, suggesting *ballistic* transport over micrometer distances. This observation is definitely inconsistent with the weak localization picture based on the *diffusive* nature of electrons. Also, the electronic band structure of the nanotube is predicted to be drastically distorted in the presence of the magnetic field.[9,10] In a magnetic field perpendicular to the tube axis, a Landau level forms at the cross of the valence and conduction bands, thereby increasing the density of states (DOS) near the Fermi level, resulting in a negative MR.[9] In a parallel field, the band gap near the Fermi level exhibits periodic modulations as a function of magnetic flux passing through the nanotube with the period of h/e.[9,10] In spite of intensive studies so far, there has been no clear evidence on the change of the DOS near the Fermi level with a magnetic field. Most of transport measurements up to now ignored such a magnetic-field-induced band effect and were mainly described by the quantum interference effect.[1–5]

In order for the clear understanding in the low-temperature transport properties in this material, the following point has to be elucidated clearly. There is no consensus on whether the nanotube can conduct current ballistically or diffusively. The existing transport measurement results and theoretical predictions are inconsistent with each other, as mentioned above. One insists that the whole low-temperature conduction can be explained reasonably in terms of the weak localization scheme,[2–4] the other insists that nanotubes can conduct electrons ballistically even at room temperature.[7] To resolve this inconsistency, one must measure both the differential conductance (dI/dV) to probe the DOS near the Fermi level and the MR for the same sample.

In this study, we have measured both the MR and dI/dV of multiwalled carbon nanotubes as a function of magnetic field perpendicular to the tube axis. A pronounced negative MR and superimposed reproducible aperiodic fluctuations were observed. The measured dI/dV curves showed a large depletion of the DOS near the Fermi level and its magnitude changed with the magnetic field. It is found that the measured aperiodic fluctuations and MR were critically dependent on the change of the DOS near the Fermi level with the magnetic field. Our results clearly show that the magnetic-field-induced band transport rather than the quantum interference effects mainly govern the low-temperature conduction properties in this system.

The multiwalled carbon nanotubes synthesized by an arc discharge method were prepared on a Si substrate with a 500-nm-thick thermally grown SiO_2 layer. The patterns for electrical leads were generated using *e*-beam lithography onto the selected carbon nanotube, and then 20 nm of Ti and 50 nm of Au were deposited successively on the contact area by thermal evaporation. Figure 1 shows the scanning electron microscopy (SEM) image of one of the samples (S1) with the four electrodes being denoted. The separation between voltage probes was about 520 nm (110 nm) for the

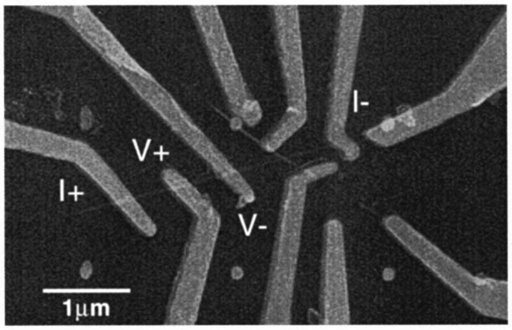

FIG. 1. Scanning electron microscope image of sample $S1$ used in this study. The four electrodes used in the measurement are denoted in the figure.

sample $S1$ ($S2$) and the diameter of the selected carbon nanotube about 25–30 nm. To form ohmic contacts between the nanotube and the Ti/Au electrodes, we have performed rapid thermal annealing at 600–800 C for 30 s.[11] Each contact resistance was below 3.5 kΩ (0.5 kΩ) for the sample $S1$ ($S2$) in the whole measured temperature range.

Figure 2 shows the four-probe MR curves of the sample $S1$ (Fig. 2) and $S2$ (inset of Fig. 2) in a perpendicular magnetic field at different temperatures. Reproducible and aperiodic fluctuation features with relatively large amplitudes can be seen with a superimposed negative MR. The shape of the MR curves is quite similar to those of Langer *et al.*[2] and Schönenberger *et al.*[5] This kind of aperiodic fluctuation has commonly been observed in carbon nanotubes and is reasonably well explained by the well-known ''universal conductance fluctuations'' mechanism. The negative MR was also interpreted as a Landau level effect by Langer *et al.*[2] and as a one-dimensional weak localization effect by Schönenberger *et al.*[5] As temperature is lowered, the overall structures become clearer and the amplitude of fluctuation becomes larger. One interesting observation is that the amplitude of the MR was not changed at the particular field at $H \approx 7.0$ T (4.0 T) for the sample $S1$ ($S2$), and its value remains around 6.45 kΩ ($=h/4e^2$) as the temperature was

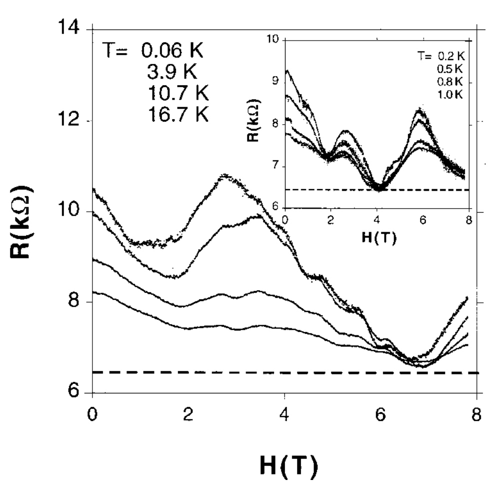

FIG. 2. The four-probe MR curves for sample $S1$ in a perpendicular magnetic field with a bias current of 50 nA at various temperatures. Inset: the four-probe MR curves for sample $S2$ in a perpendicular magnetic field with a bias current of 0.5 nA. The dotted line represents the resistance value of 6.45 kΩ ($=h/4e^2$).

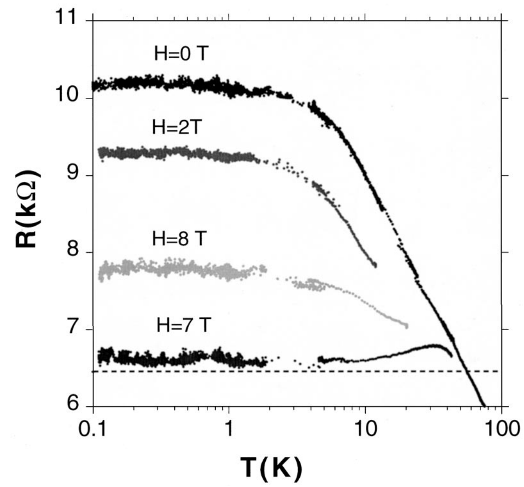

FIG. 3. The temperature dependence of the four-probe resistance for sample $S1$ in the presence of a perpendicular magnetic field. The dotted line represents a resistance value of 6.45 kΩ ($=h/4e^2$).

lowered. In the case of single multiwalled nanotubes with the tube diameter of 20–35 nm, we found that the resistances in the metallic state were around $2G_0$ at sufficiently low temperatures.

This observation can be confirmed further by examining the temperature dependence of the resistance as a function of magnetic field. Figure 3 shows the resistance (R) vs temperature (T) data of the sample $S1$ in the temperature range 0.1 K $<T<$80 K at various magnetic fields, with a 50 nA bias current. The zero field R vs T data shows a ln(T) dependence followed by a saturation below 2–3 K. However, an unusual temperature dependence of the resistance was observed at $H \approx 7.0$ T, at which the MR value was not changed as the temperature was lowered and approached around 6.45 kΩ ($=h/4e^2$). The resistance at 7.0 T shows a true metallic temperature dependence of resistance without any weak localization effect below 30 K, commonly found in the crystalline materials, that is, the resistance decreases as the temperature is lowered. At lower temperatures, resistance saturates around 6.45 kΩ ($=h/4e^2$), the minimum resistance for one graphene cylinder. At sufficiently low temperatures, it is known that the electrical conduction through the outermost shell would be dominant in a multiwalled nanotube. Neighboring shells of multiwalled nanotubes in general have different chirality, so that it can be considered as a series of tunnel junctions along the radial direction.[12,13] Thus, thermally activated layer-to-layer hopping is known to be only possible at high enough temperatures above 10 K due to the relatively large hopping energy. It is believed that the turning point of the resistance at 7.0 T near 35 K shown in Fig. 3 would be a starting temperature for the outermost shell conduction, so that the electric conduction can mostly occur along the outermost graphene sheet below 35 K. Thus, one graphene sheet would have a metallic temperature dependence of resistance without any weak localization correction, found in the crystalline materials.

To clarify the origin of the unusual temperature dependence of the resistance and the aperiodic resistance fluctuation, we have measured the current-voltage (I-V) characteristics with varying magnetic fields and obtained dI/dV

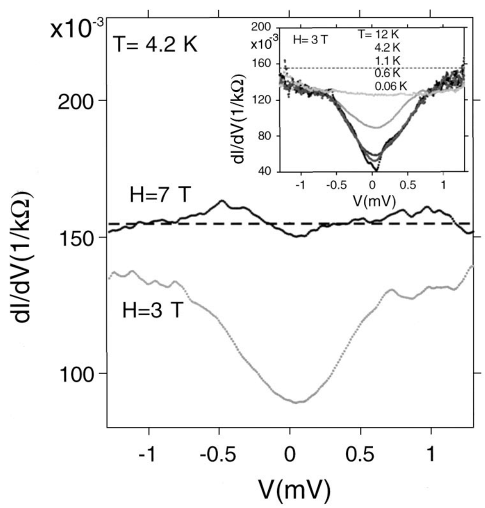

FIG. 4. The dI/dV curves for sample $S1$ near the resistance maximum ($H = 7.0$ T) and minimum ($H = 3.0$ T) points at 4.2 K. Inset: the temperature dependence of the dI/dV curves at $H = 3.0$ T. The dotted line represents the resistance value of 6.45 kΩ ($= h/4e^2$).

curves. To avoid the contact effect, we have employed a four-probe technique in measuring the *I*-*V* characteristics. We have also measured the two probe *I*-*V* characteristics and found no differences in the curves except the addition of a magnetic-field-independent contact resistance. Nonlinear *I*-*V* characteristics were observed in the low bias region.

Figure 4 shows the dI/dV curves near the resistance maximum ($H = 3.0$ T) and minimum ($H = 7.0$ T) points at 4.2 K, respectively. The shape of the dI/dV curve at the resistance minimum is quite different from that at the resistance maximum. The dI/dV curves for $H = 3.0$ T showed a large depletion of the DOS with a size of about 1.8 meV on both sides of the Fermi level. Since the *I*-*V* characteristic is highly nonlinear at the depletion region, the MR changed considerably by varying the bias current level and temperature. We found that the whole transport properties are critically dependent on the change of the DOS near the Fermi level.

At the resistance minimum ($H = 7.0$ T), on the other hand, the dI/dV curve becomes rather flat without a depletion of DOS and shows true ohmic behavior. Also note that the conductance value at this field approaches at the theoretical value, $2G_0 = 4e^2/h$, regardless of temperatures below 4 K. Also, MR did not change with varying the bias current indicating the ohmic behavior. The appearance of a metallic and a flat DOS is quite consistent with the temperature dependence results. The flat and metallic DOS without a depletion could give a true metallic temperature dependence of the resistance, that is, the resistance decreases as the temperature is lowered, as mentioned above. In this case, true metallic conduction along the outermost graphene sheet starts below 35 K, and the conductance increases and finally saturates to the maximum theoretical value, $2G_0 = 4e^2/h$, as shown in Fig. 3.

The change of the DOS near the Fermi level with varying magnetic field was confirmed by observing the perspective view of $dI/dV(V,H)$ curves at 4.2 K as a function of bias voltage V and magnetic field H, as shown in Fig. 5(a). The

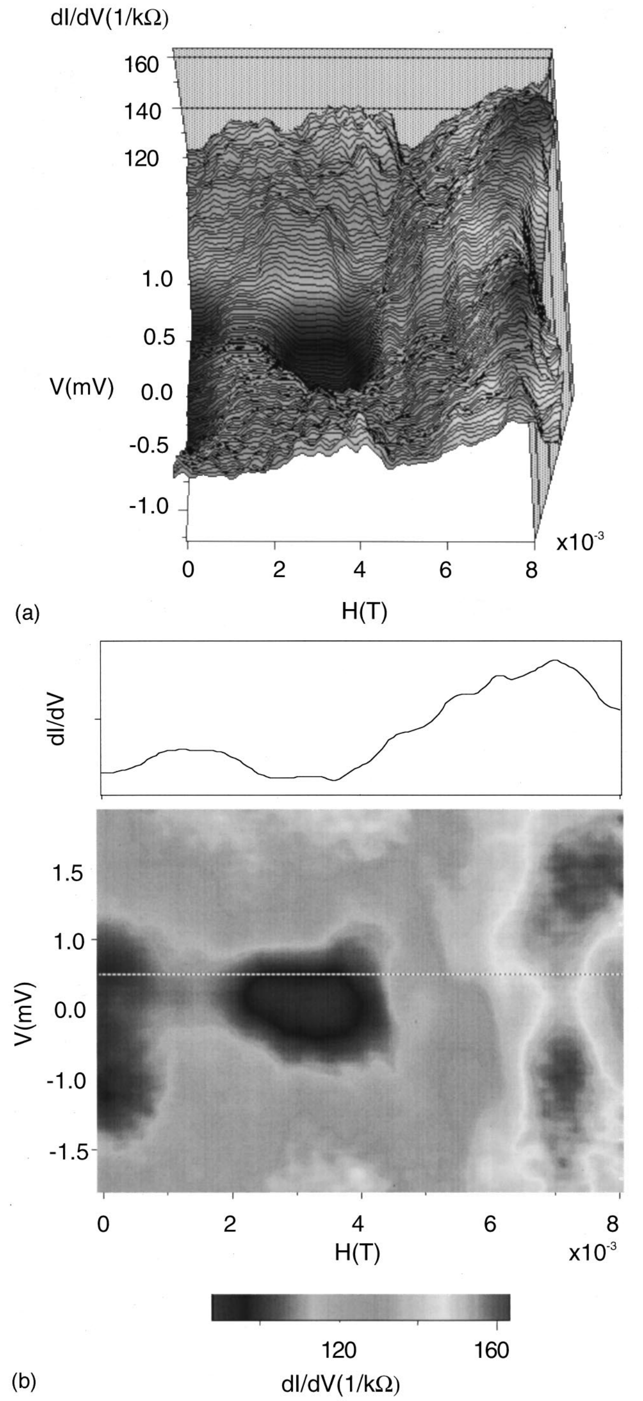

FIG. 5. (a) A perspective view of the $dI/dV(V,H)$ curves for sample $S1$ at 4.2 K as a function of bias voltage V and magnetic field H. (b) A top view of the $dI/dV(V,H)$ curves for sample $S1$ and the cross-sectional plot along the dotted line.

cross-sectional view of Fig. 5(b) at $V \approx 0.5$ mV retrieves the MR curve exactly in Fig. 2. They clearly demonstrate the change of the DOS near the Fermi level as a function of the magnetic field. They also show that the measured MR and aperiodic oscillations originate from the change of DOS near the Fermi level with the magnetic field. At the resistance minimum ($H \approx 7.0$ T), on the other hand, the dI/dV curve becomes rather flat and the MR become insensitive to the bias level as mentioned. More careful analysis shows that there exist two peak structures in dI/dV curves near $+1$ mV and -0.5 mV around 7.0 T, which seems to be related to two $1d$ subbands originating from the π and π^* bands of a graphene sheet.[8] Due to the two available subbands at the Fermi level, the nanotube should ideally have a conductance of $2G_0=4e^2/h$. At the resistance maximum ($H=3.0$ T), a relatively large depletion of the DOS (dark regions in Fig. 5) can be seen. As the temperature is lowered, the depletion near the Fermi level becomes more and more pronounced, as shown in the inset of Fig. 4. It is also found that the logarithmic temperature dependence of the resistance below ≈ 30 K also mainly originates from the temperature dependence of the DOS, not from two-dimensional weak localization. Thus, the electrical transport in this system must reflect the temperature and bias dependencies of the DOS near the Fermi level, which varies considerably as a function of magnetic field and temperature. Our data can be explained by the magnetic field– and temperature-dependent depletion of the DOS without introducing weak localization effects.[9]

As for the origin of the magnetic-field-modulated DOS, Ajiki and Ando[9] predict that in a magnetic field perpendicular to the tube axis, the band gap is strongly reduced due to a formation of 2D Landau states, and the energy spectra approach those of a graphene sheet. These result in a monotonic decrease of the MR. Although our experimental results show the predicted gradual decrease of MR with increasing magnetic field, the size of the depletion of the DOS and the modulation features of MR are far from the theoretical predictions.[9] The measured energy gap (~ 1.7 meV) is one order of magnitude smaller than the theoretically predicted value. The most unusual observation is the existence of aperiodic fluctuations of the MR in the perpendicular field totally absent in the theoretical predictions.[9] Since the nanotube has chirality, the electrons can move along the helical trajectories and pick up the Aharonov-Bohm–type phase even in the presence of a magnetic field perpendicular to the tube axis, resulting in the modulation of the MR, as in a parallel field. We believe that the unique structural characteristics of the nanotube can give rise to a modulation of the MR with the magnetic field, even in a perpendicular direction, just as in the parallel field case. Further theoretical and experimental studies are required for a clear understanding of the observed MR and the depletion of the DOS.

This work was supported by a Korea Research Foundation Grant (KRF-99-015-DP0128), the BK21 project, KRISS Project No. 00-0502-100, and also by the Nano Structure Project supported by the Ministry of Science and Technology, Korea.

[1] S. N. Song, X. K. Wang, R. P. H. Chang, and J. B. Ketterson, Phys. Rev. Lett. **72**, 697 (1994).
[2] L. Langer *et al.*, Phys. Rev. Lett. **76**, 479 (1996).
[3] M. Baxendale, V. Z. Mordkovich, and S. Yosimura, Phys. Rev. B **56**, 2161 (1997).
[4] A. Bachtold, M. Henny, C. Terrier, C. Strunck, and C. Schönenberger, Appl. Phys. Lett. **73**, 274 (1998).
[5] C. Schönenberger, A. Bachtold, C. Strunk, J.-P. Salvetat, and L. Forro, Appl. Phys. A: Mater. Sci. Process. **69A**, 283 (1999).
[6] A. Bachtold, C. Strunk, J.-P. Salvetat, J.-M. Bonard, I. Forro, T. Nussbaumer, and C. Schönenberger, Nature (London) **397**, 673 (1999).
[7] S. Frank, P. Poncharal, Z. L. Wang, and W. A. de Heer, Science **280**, 1744 (1998).
[8] R. Saito, M. Fujita, G. Dresselhaus, and M. S. Dresselhaus, Appl. Phys. Lett. **60**, 2204 (1992).
[9] H. Ajiki and T. Ando, J. Phys. Soc. Jpn. **62**, 1255 (1993).
[10] W. Tian and S. Datta, Phys. Rev. B **49**, 5097 (1994).
[11] Y. Zhang, T. Ichihashi, E. Landree, F. Nihey, and S. Iijima, Science **285**, 1719 (1999).
[12] J. C. Charlier and J. P. Michenaud Phys. Rev. Lett. **70**, 1858 (1993).
[13] R. Saito, G. Dresselhaus, and M. S. Dresselhaus, J. Appl. Phys. **73**, 494 (1993).