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Electronic Transport in Multiwalled Carbon Nanotubes Contacted with Patterned Electrodes

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The electrical conductance of 0.8 ~ 5-μm-long multiwalled carbon nanotubes (MWCNT) was measured at room temperature in a multiprobe scanning tunneling microscope (STM)-scanning electron microscope (SEM) system and a conventional prober system, by bringing the MWCNTs into contact with patterned metal electrodes. The contact resistance between the CNTs and metal electrodes was sufficiently small. The conductance was proportional to A/L (and also to B/L, within our experimental error), where A, B, and L are the cross section, circumference, and length of CNTs. This indicates the occurrence of diffusive transport. A nonlinear current-voltage characteristic was obtained; the conductance increased steeply with current. A multiprobe STM-SEM system was very useful for measuring individual CNTs. [DOI: 10.1143/JJAP.43.L1081]

KEYWORDS: carbon nanotube, multiprobe STM, electrode, conductance, *I-V* characteristic

The electronic transport properties of cabon nanotubes (CNTs) have received much attention from the points of view of one-dimensional transport physics as well as device applications.[1)] Though a number of test devices such as field-effect transistors,[2–4)] single electron transistors,[5,6)] and nanosensors[7)] have been created with CNTs, the fundamental transport property is still controversial,[8)] probably because of their poorly defined quality and structure. Some groups have reported that the multiwalled carbon nanotubes (MWCNTs) exhibit ballistic conduction,[9–11)] while other groups have reported diffusive conduction characteristics.[12–14)] Another interesting issue is the current path; Collins *et al.* suggested that the current flows preferentially through the outermost shell of MWCNTs, rather than through the entire cross section.[15)] The third point to be addressed is that of the nonlinear current-voltage characteristics; since they have been reported only very recently, their origin has not yet been identified.[16,17)]

In this Letter, we show systematic measurements of the conductance of MWCNTs prepared by bringing them into contact with metal electrodes at both ends of the MWCNTs in two different ways. One is that the CNTs were dispersed on patterned Ta electrodes, and the conductance of CNTs bridging the two electrodes was measured with a four-tip-scanning tunneling microscope (STM) combined with a scanning electron microscope (SEM) in ultrahigh vacuum.[18,19)] With this system, we could identify the CNTs bridging the electrodes and measure the conductance selectively by touching two STM tips to the electrodes. This method was very effective for identifying and measuring many CNTs on a substrate at one time. Another method involved depositing Ti electrodes over the CNTs at both ends. While this technique is expected to be more effective for future device application of CNTs, the contact resistance is said to be problematic in some cases.[9)]

We found that the conductance g is proportional to A/L, where A is the cross section and L is the length of CNTs, for both methods. The data also appeared to be proportional to B/L within our experimental error, where B is the circumference of CNTs. In any case, the results indicates the occurrence of diffusive transport through the CNTs longer than 0.8 μm. But we could not resolve whether the conduction is through the outermost shell ($g \propto B/L$) or though the entire cross section ($g \propto A/L$), because of the limited CNT diameter range we investigated. We also found nonlinear current-voltage (*I-V*) characteristics which were similar to recently reported ones.[16)] This phenomenon is, therefore, a property which is intrinsic to MWCNTs, irrespective of the synthesis and measurement methods used. Since our samples exhibited low contact resistance and consistent results, we can say that the contacts at the electrodes were highly reproducible.

The experiments were performed in two ways. In the first method (Method I), we used a SiO_2/Si wafer as the substrate, with patterned square Ta pads separated by various distances (0.8–5 μm).[20)] Commercial CNTs having various diameters (0.05–0.2 μm), fabricated by the arc discharge method, were randomly dispersed on the substrate using ethanol or dimetylformamide. We investigated the structure and quality of the CNTs separately by means of transmission electron microscopy (TEM), and confirmed they were of good quality with few defects (Fig. 1(b)). A sample was placed in the four-tip STM-SEM vacuum chamber.[18,19)] We chose CNTs which bridged two pads under the SEM, and measured their *I-V* curves by touching two STM W-tips onto the two pads (Fig. 1(a)). In the case with no CNT between the pads, the resistance was larger than 10 MΩ, and thus was much larger than that between two pads bridged by a CNT. We also confirmed, by touching the two tips onto the same pad, that the resistance between tip and pad was sufficiently small (~1 Ω). A typical *I-V* curve is shown in Fig. 1(c). Though it shows slight non-linearity, we took the slope near the origin as the resistance.

The energy of our SEM electron beam was 10 keV, and we exposed the beam on CNTs or STM tips for from about 10 minutes to 12 hours, resulting in an exposure of about 20 C/cm^2 at most under usual conditions. The SEM observations showed that the irradiation did not make any visible contaminations or defects on CNT or STM tips (even by 100 times larger exposures than usual), probably because of the ultrahigh vacuum environment in our chamber and room temperature condition. The measured *I-V* curves and resistance looked unaffected by such electron beam irradiations. Some groups have reported[21,22)] that CNTs are

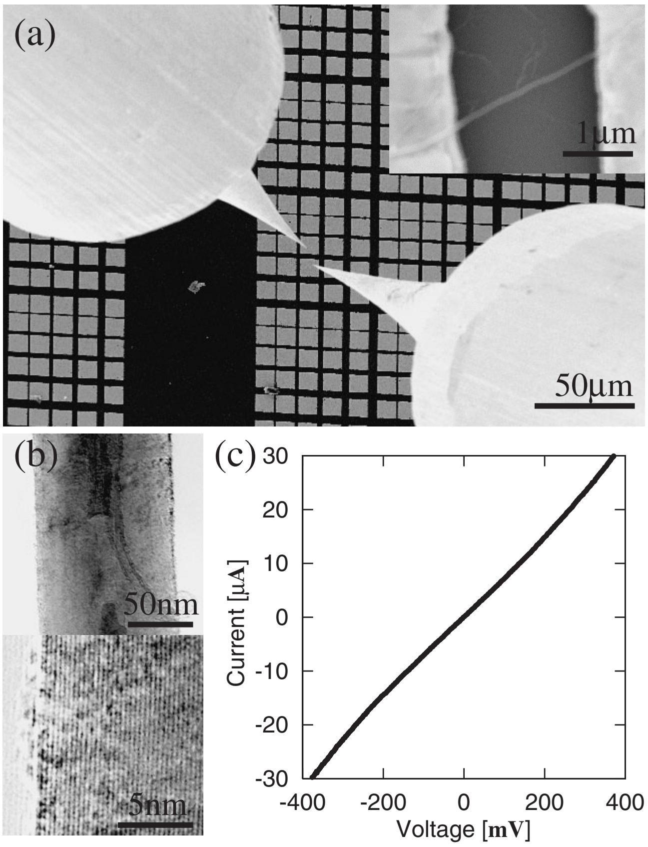

Fig. 1. (a) A SEM image of two STM tips approaching the Ta pads. Inset is a typical SEM image of a MWCNT bridging two pads. (b) TEM image of a MWCNT used in Method I. (c) Typical *I-V* characteristic.

damaged by the electron beam when its energy is above some critical value (80–100 keV), which is much higher than the energy of our electron beam. Therefore was can safely say that the electron beam of our SEM does not affect the results presented here.

In the second method (Method II), CNTs were dispersed on a SiO_2/Si wafer, and then patterned Ti pads were deposited on top of them (Fig. 2(a)). To reduce the contact resistance, annealing was performed at 800 °C for 30 min.[23] Without annealing, the measured resistance was very high (>100 kΩ) due to poor contact. After identifying CNTs bridging two electrodes under an SEM (Fig. 2(b)), the conductance was measured by touching the probes to the Ti pads in a macroscopic prober under an optical microscope in air. The quality of the CNTs used was evaluated by TEM (Fig. 2(c)). The CNTs in this method were also commercial ones obtained from a different company, fabricated by the arc discharge method, and their diameters ranged from 20 nm to 100 nm; thus they were smaller than those in Method I.

The diameter and length of the measured CNTs were determined *in situ* from SEM images, whose resolution was about 5 nm. All measurements were performed at room temperature. We defined the CNT length as the length of the CNT portion spanning the gap between the electrodes.

Figure 3 shows the measured conductance as a function of A/L, as obtained by Method I. The conductance is proportional to A/L up to $A/L \sim 30$ (nm). We plotted the measured conductance separately as a function of diameter and of length, but did not find any correlations of the conductance with either diameter or length.

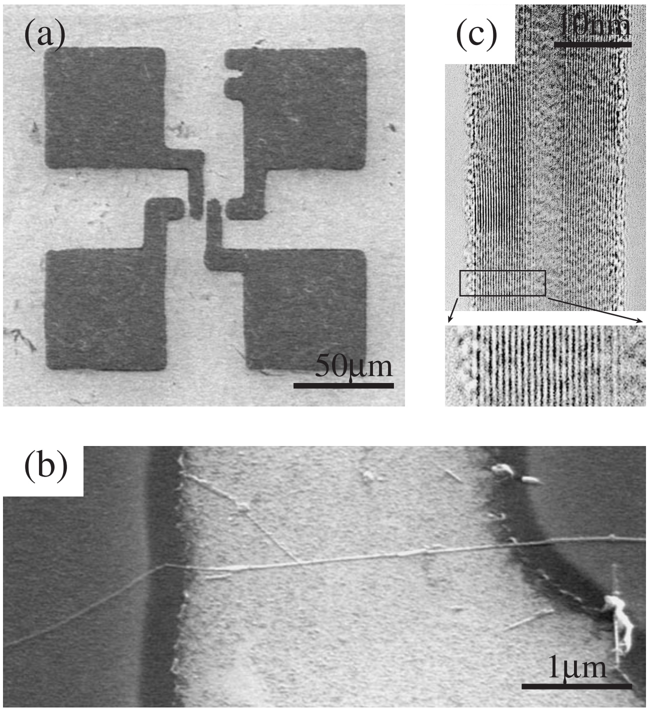

Fig. 2. (a) SEM image of Ti electrodes. (b) Close up view of dispersed CNT covered by the deposited electrodes. (c) TEM image of the CNT used in Method II.

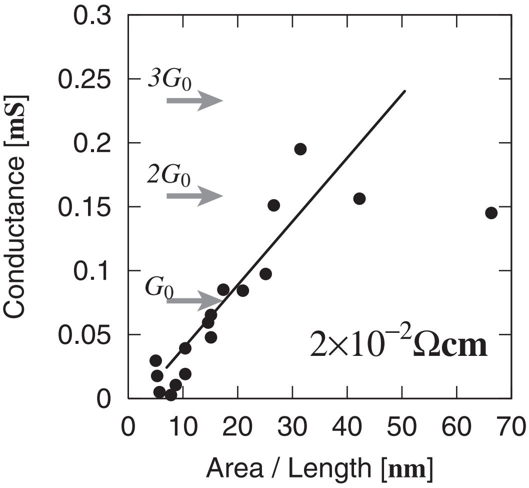

Fig. 3. Plots of measured conductance versus A/L (cross section/length) of MWCNTs in Method I. The data points show a linear dependence up to $A/L \sim 30$ nm. Arrows indicate the quantum conductance ($G_0 = 2e^2/h$).

The measured conductance was also plotted as a function of B/L (not shown here). Due to the limited range of B in our samples and data scattering, the resistance appeared to be proportional to B/L as well as A/L. If the current is restricted to the outer shells of the CNTs,[15] the resistance should be proportional to B/L. However, if the conduction is through the entire cross section of the CNTs, the conductance should be proportional to A/L. We could not determine which possibility fits the experimental data better, within our experimental error.

The linear relation of conductance with A/L (or B/L) in

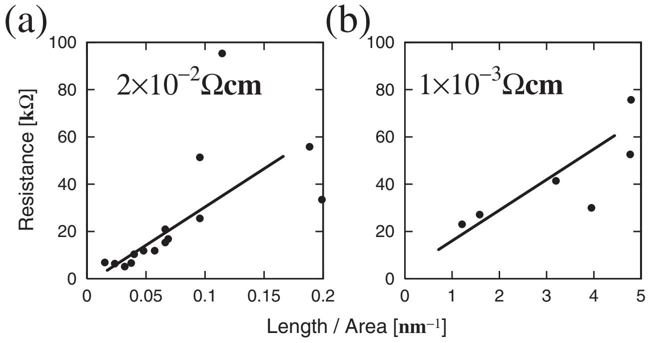

Fig. 4. Plots of resistance as a function of L/A (length per cross section) in (a) Method I and (b) Method II. Both graphs show linear dependence, whose gradient indicates the resistivity.

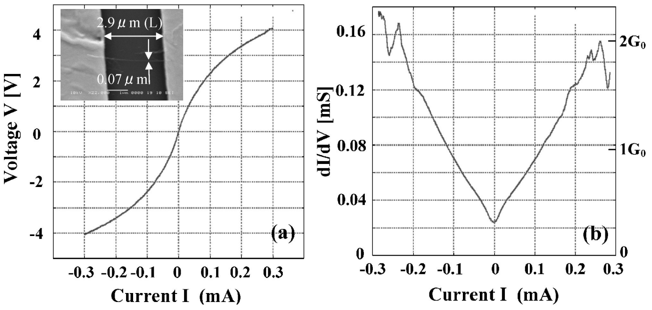

Fig. 5. (a) Nonlinear *I-V* curve of a single MWCNT in Method I. Inset shows a SEM image of the CNT bridging electrodes. (b) Conductance of the single MWCNT as a function of current.

Fig. 3 means that *the conduction is diffusive*. If the current flows in a ballistic manner, the resistance should be constant, irrespective of the CNT length and diameter. Or if the number of conduction channels changes depending on the length or diameter, the conductance should be quantized. However, our data shows no such quantization tendency, and also it shows no systematic relation with diameter or length. The conductance appears to be bounded around 2G$_0$ at $A/L > 30$ nm in Fig. 3. This maximum conductance (minimum resistance) appears to be due to the contact resistance at the two ends (ca. 5 kΩ) as estimated below. Some groups have reported that MWCNTs exhibit ballistic conduction at room temperature.[4,10,11] As described above, however, we found no ballistic feature for CNTs longer than 0.8 μm in our experiments, though shorter CNTs (or larger A/L) with fewer defects may effect ballistic conduction.

Figure 4 shows the measured resistances as a function of L/A obtained by the two methods. The resistivity is approximately $2 \times 10^{-2}\ \Omega\cdot$cm in Method I and $1 \times 10^{-3}\ \Omega\cdot$cm in Method II. The values are similar to that given in the previous report,[24] or are an order of magnitude higher than that given in ref. 12. Since the resistance depends on L/A (or L/B), we can say that these resistivities are those of tubes, not the contact resistance. Furthermore, since the contact resistance should depend on the contact area, the resistance should be inversely proportional to the diameter of the CNTs, which it is not in our case. By extrapolating the resistance at $L/A = 0$ in Fig. 4, the contact resistance of $1 \sim 5$ kΩ is yielded, which is sufficiently small compared with the resistance of CNTs.

The resistivities obtained in our two experiments differ by one order of magnitude. Though the samples in the two methods were confirmed to have similarly good crystal quality by means of TEM, it is reasonable to consider that there are differences in defect density and contamination between them. Moreover, as shown in Figs. 1 and 2, the samples used in Method I are much thicker than those used in Method II. If the current flows preferentially in the outer shells in the thicker CNTs, the effective cross section A for the current flow should be smaller than the entire cross section assumed for calculating the resistivity, which may lead to an overestimation of the resistivity in Method I.

We have found a nonlinear *I-V* characteristic as shown in Figs. 5(a) and 1(c). Though the details of the curve differ from tube to tube, the overall features were similar for all MWCNTs (see also Fig. 1(c)). As shown in Fig. 5(b), the conductance has a minimum value of approximately 0.02 mS at zero current (at zero bias voltage) and steeply increases with increasing the current. A similar *I-V* characteristic has been reported previously for MWCNTs.[16] However, this type of nonlinear *I-V* characteristic is significantly different from that of single-walled CNTs.[25] Although the origin for this nonlinearity has not yet been clarified, it can be interpreted as being due to a change of the current distribution in the cross section of the MWCNTs; the outermost shell mainly contributes to the conduction at lower current, while the inner shells gradually begin to contribute with increasing current, so that the total conductance increases with current. A single MWCNT could carry a current of more than 0.3 mA with large bias. This is an indication of very small carrier scattering in the tubes.

We could measure a large number of samples at one time owing to the four-tip STM-SEM system. In this system, we can bring the two tips as close together as 150 nm, which enables us to measure shorter CNTs; this may reveal a shift from diffusive to ballistic conduction depending on the length. Four-point probe measurements will also be available with this machine, which will be reported elsewhere.

In summary, by measuring the resistance of many MWCNTs having various diameters and lengths, we confirmed that MWCNTs longer than 0.8 μm exhibited diffusive conduction. The resistance was proportional to the cross section divided by the length. The resistivity of the CNTs was approximately $1 \times 10^{-3} \sim 2 \times 10^{-2}\ \Omega\cdot$cm. The contact resistance between the CNT and the electrodes was found to be sufficiently small for measuring the CNT resistance in our two methods. We have also confirmed an intrinsic nonlinear *I-V* characteristic; the conductance increases with bias voltage, and a single MWCNT could carry a current of more than 0.3 mA. The four-tip STM was found to be very useful for measuring the individual CNTs.

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