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Optimized Contact Configuration for the Study of Transport Phenomena in Ropes of Single-Wall Carbon Nanotubes

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Optimized contact configuration for the study of transport phenomena in ropes of single-wall carbon nanotubes

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The study of the intrinsic transport properties of carbon nanotubes suffers from the difficulties in fabricating noninvasive contacts. Here, we present a scheme for the investigation of transport phenomena in metallic single-wall carbon nanotubes by means of a special four-terminal measurement configuration. To suppress the impact of the contacts on the measured conductance in a tube, we found a combination of top and bottom contacts to the rope of single-wall nanotubes to be most appropriate. Our experimental findings demonstrate that a linear decrease of the sample resistance can be observed under these circumstances without the common increase of resistance for decreasing temperatures. [DOI: 10.1063/1.1373413]

The electrical properties of carbon nanotubes (NTs), a class of macromolecules discovered almost a decade ago,[1] may play a central role in future nanoelectronics. Their unique structure makes them potentially useful as basic elements for generations of highly integrated circuits.[2] While the well known impact of the geometrical structure on charge transport in NTs is responsible for the wide variety of electrical properties found so far,[3] it is this structure, on the other hand, which makes it very difficult to characterize the *intrinsic* transport properties of NTs. The problem is to perform a noninvasive measurement which is essential to avoid effects associated with the contacts.

The impact of contacts on the NT properties are reported by several groups. As pointed out by Bezryadin and co-workers,[4] the bending of a single-wall nanotube (SWNT) dispersed on metal electrodes (bottom contacts) can result in the formation of barriers in the tube, which is ideal for the investigation of coulomb blockade effects but undesired for a transport experiment. The same is true for most top contacts where the tube is placed on a flat substrate and the electrodes are attached to the top of the tube. For such a case, Bockrath and co-workers[5] showed that the leads divide the NTs into segments.[6] Again, barriers are obviously created inside the NT. In addition, Bachtold and co-workers[7] clearly demonstrated the strong impact of electron irradiation on multiwall nanotubes. This implies, that, for top contacts patterned by electron-beam lithography, an influence on the electrical properties of the tube cannot be excluded. All these facts support the aforementioned statement that a noninvasive measurement is difficult to perform. It is not the contact resistance R_C that causes the problems, but the resistance inside the tubes R_B induced by making contacts. Thus, a four-terminal measurement alone is not a solution to the problem.

Electrical transport measurements presented so far (e.g., Refs. 5 and 7–11) have something in common independent of whether a four- or a two-probe configuration is used. Below a critical temperature (between 30 and 300 K), a negative temperature coefficient, i.e., an increase of resistance with decreasing temperature is observed. We attribute this to the fact that even in ropes of SWNTs (most of the aforementioned measurements were done on ropes instead of SWNTs) making contact to the NTs can perturb the electron (or hole) motion. From the results presented in this letter, we believe this to be the reason why Kaiser and co-workers[8] were able to obtain good fits to resistance versus temperature plots $R(T)$ in the low-temperature range with a formula taking into account fluctuation-induced tunneling. However, we do not claim that the resistance increase for decreasing temperature in *all* existing measurements is due to tunneling. In particular, we do not want to exclude Luttinger-liquid (LL) behavior as found by different authors (e.g., Ref. 5) in two-terminal measurement configurations as an explanation for the increase of $R(T)$ for decreasing T. But even in these cases, the contacts play a central role in the measurement since LL behavior cannot be observed without the presence of barriers in the current path.[12] A precursor of what we consider the intrinsic properties of the NTs has been found for higher temperatures, where a linear increase of $R(T)$ with increasing temperature is observed, probably due to electron–phonon interaction in metallic NTs.[10]

Here, we propose a scheme to study the intrinsic electrical properties of metallic single-wall carbon nanotubes down to lowest temperatures. As will be described, a combination of top and bottom contacts to a rope of SWNTs is ideally suited for that purpose.

To perform electrical transport measurements, an individual rope of SWNTs is contacted as shown in Fig. 1. Cross-like patterns consisting of Ti/Au are defined on a Si/SiO_2 substrate prior to the dispersion of the NT ropes.[13] Afterwards, the scanning electron microscope (SEM) is used to identify a rope of sufficient length (around 10 μm) touching three metal crosses and a large metal pad (1). Four additional metal leads are then attached from the top to the rope using standard electron-beam lithography and lift-off technique. The cross-like structures which have been electrically connected in the same process step serve in the following as bottom contacts. A schematic of the positions of the contacts relative to the rope is shown in the lower part of Fig. 1. The

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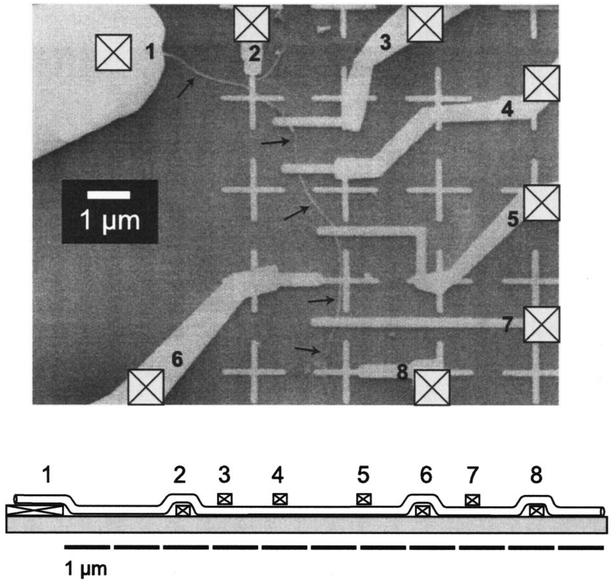

FIG. 1. Top: SEM image of the eight contacts attached to a 10 μm rope (indicated by arrows). The lower part shows the contact positions relative to the rope with four top contacts (3, 4, 5, and 7) and four bottom contacts (1, 2, 6, and 8).

configuration allows one to study the impact of the different contact types and—as will be shown—offers the possibility to eliminate the contact effects.

Electrical transport measurements are performed in a ^{4}He continuous flow cryostat between 2.5 and 300 K using a standard lock-in technique. Since the intertube coupling is weak,[14,15] the current will flow in the lower part of the well-ordered rope if the bottom current contacts are used, and in the upper part of the rope if top contacts are used accordingly. The main idea is that the top contacts only disturb the transport in NTs in the upper part of the rope while the bottom contacts only have an impact on the tubes in the bottom part of the rope.[16] Therefore, contributions to the resistance from the voltage probes are avoided since the current is driven through the tubes close to the substrate using the bottom contacts while the voltage drop is measured with two top contacts that are weakly coupled to the current carrying NTs.

To prove that this is the case and to show that a standard four-terminal measurement does not strictly probe the intrinsic electrical properties of the tube, we performed two types of measurements. First, as usually done, we determined the temperature dependent resistance $R(T)$ using a set of four top *or* four bottom contacts as plotted in Fig. 2(a). In the other measurement configurations, we used two bottom contacts (1 and 8) to drive the current through the sample while measuring the voltage drop across the rope with two adjacent top contacts (3 and 4) [Fig. 2(b)]. For simplicity, we assumed at this point that the whole current (500 pA) is carried by a single tube in the rope. The difference between the two types of measurements—although *both* excluding contributions from the contact resistances R_C—is striking. While the standard configuration results in an increase of $R(T)$ with decreasing T, very similar to the results commonly obtained,[5,7–11] in Fig. 2(b) a clear linear decrease of resistance with decreasing temperature is visible. Even more important is the extremely small resistance value of around 1

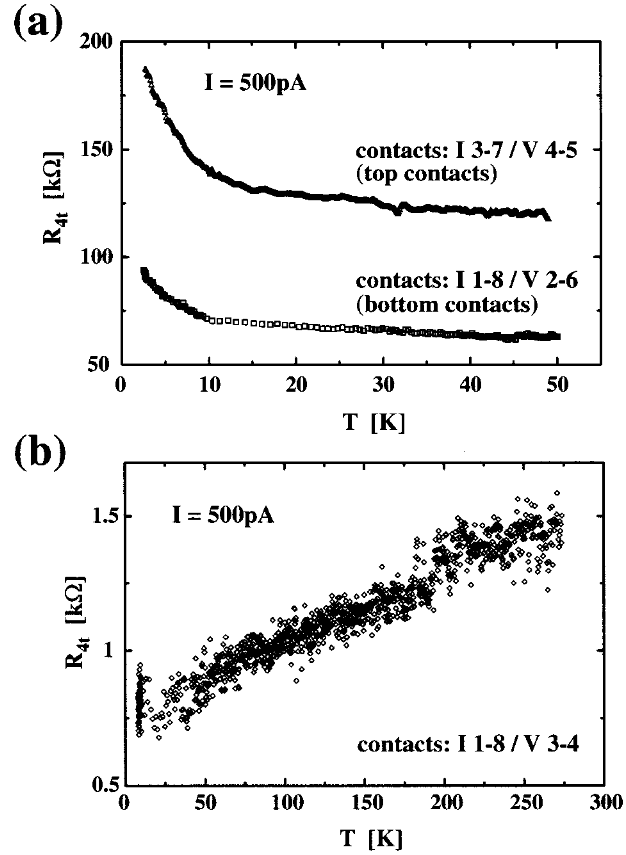

FIG. 2. Four-terminal measurements on a rope of SWNTs. In case (a), both curves were taken with a set of either four top *or* four bottom contacts. The characteristic shows a monotonic decrease of resistance up to room temperature (not shown). Measurement (b) is performed with two top contacts as voltage probes (3 and 4) and two bottom contacts (1 and 8) serving as current source and drain.

kΩ in Fig. 2(b) measured over the entire temperature range for a voltage probe separation of 1.1($\pm$0.2) μm. On the other hand, in Fig. 2(a), we observe resistances approximately 100 times larger although the voltage probe separation only differs by a factor of ~2 and ~4, respectively.

Our interpretation of the results is that current transport for the two cases in Fig. 2(a) is perturbed by both types of voltage probes. It only makes a quantitative difference whether top or bottom contacts are used. In Fig. 2(a), the barriers introduced by the top contacts are causing somewhat more backscattering than the bottom contacts. The resulting average barrier resistance R_B at a few Kelvin is between 35 and 80 kΩ arguing that two barriers are present in each of the two measurements. Because of these barriers in the current paths, the *intrinsic* electrical properties of the NT cannot be resolved. Another argument supporting our analysis is that the data obtained for the standard measurement configuration do not scale with the voltage probe separation L_V. In particular, the measurement for the larger L_V (bottom contact) shows the smaller four-terminal resistance value which would imply a—very unlikely—pronounced difference of scattering inside different metallic tubes in a rope. On the other hand, top contacts—although damaging the upper part of the rope—do not alter the situation inside a NT in the bottom part of the rope. Thus, the combination of top and bottom contacts as shown in Fig. 2(b) makes it possible to study the intrinsic transport properties of metallic NTs.

Before evaluating in detail the data from Fig. 2(b), the first issue to discuss is the rather high noise level of approximately 15%. This noise results from the very small voltage difference of only a few hundred nanovolts detected with the weakly coupled voltage probes. Nonetheless, the linear shape in the measurement can easily be identified.

To determine the relevant scattering length in the NT accurately from this slope and the absolute value of $R(T)$, we have to reconsider our previous assumption that only a single metallic NT carries the current in the rope. Indeed, it is in principle possible that more than one metallic tube is involved in current transport from source to drain. This implies, that $R(T)$ as calculated in Fig. 2(b) may underestimate the *true* resistance depending on the number N of metallic tubes involved. To estimate the upper limit of N, we argue as follows. The rope consists of roughly 100 tubes, with approximately 30 of them being metallic. Since current is injected by two bottom contacts, simple geometrical considerations yield no more than $N=5$ to 10 metallic tubes in direct contact with the current leads.

In addition, there is another potential source to explain why the NT resistance may be effectively higher than shown in Fig. 2(b). Let us consider the case of a metallic NT which is closer to the voltage probes relative to the main current carrying tube. The voltage probes would measure the voltage drop across this tube. Because of the weak coupling to the ''main tube''—the one connected directly to the current contacts—only part of the total current would flow through this top tube. As a result, the detected voltage drop would be smaller than in the case of no tube–tube coupling. To estimate the potential error made when plotting the resistance in Fig. 2(b) assuming a current of 500 pA, we measured the two-terminal resistance between contacts 3 and 4. At low temperatures, the resistance including contact resistances and barrier resistances, R_B, as well as the intrinsic resistance add up to 60 kΩ. This relatively small value implies an intrinsic resistance of the metallic NT certainly no larger than 10 kΩ. Using the variable N just introduced to account for the uncertainty in the true tube resistance, this means again $N \leqslant 10$.[17]

Having all this in mind, we find $d\varrho/dT \times 1/N = 2.4 \times 10^6$ Ω/K$\times$m) from the linear slope in Fig. 2(b). For electron–phonon interaction in metallic NTs, a linear temperature dependence of $R(T)$ is theoretically expected[10,18] with a slope of $d\varrho/dT \approx 2\times 10^7$ Ω/K$\times$m). On the other hand, Hertel and Moos[19] found experimentally $d\varrho/dT \approx 5 \times 10^6$ Ω/K$\times$m) using time-resolved photoemission spectroscopy. From the considerations on N, our data rather support the measurements from Hertel and Moos[19] than the theoretical predictions. The characteristic length for electron–phonon scattering from our experiment is determined to be $L_{\mathrm{el-ph}} \times N = 11$ μm at $T=250$ K. Although electron–electron interaction, in principle, can result in a linear increase of $R(T)$ with temperature, as pointed out by Balents and Fisher,[20] it does not dominate transport in Fig. 2(b). We conclude this from the fact that no charge gap is observed in our measurement down to 8 K, which implies that $d\varrho/dT_{\mathrm{el-el}} \leqslant 2\times 10^5$ Ω/K$\times$m) (according to Ref. 20) is much smaller than our experimental findings even for $N=1$. Furthermore, we deduce from our data that residual impurity scattering is responsible for an additional temperature independent resistance contribution. Our data reveal an elastic mean free path of $L_{\mathrm{imp}} \times N = 10$ μm from extrapolating $R(T)$ to zero temperature.

It is important to emphasize that our measurement is only sensitive to those scattering mechanisms that alter the electron momentum. This implies, that the electron–phonon scattering found described does not automatically imply a similar coherence length at room temperature. In particular, electron–electron interaction without a change of electron velocity will not be detected.

In conclusion, we have identified an approach to study the intrinsic electronic properties of metallic single-wall carbon NTs by means of a combination of top and bottom electrodes attached to a rope of SWNTs. Our experiments clearly show that the same contacts in a different configuration (a set of four top *or* four bottom contacts) cannot be used for the study of scattering mechanisms in NTs down to the lowest temperatures. A consistent picture emerges if we assume that barriers are introduced inside the rope by both types of contacts and a suitable measurement setup is necessary to exclude their influence. Our electrical measurements show that a combination of electron–phonon and impurity scattering dominates the four-terminal resistance of NT ropes between 8 and 300 K.

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