Transport in ropes of carbon nanotubes: Contact barriers and Luttinger liquid theory

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In this article we report on the conductance of bundles of single-wall carbon nanotubes as a function of temperature and bias voltage. We focus on the nonlinearity in the IV curves near zero bias, i.e., a pronounced conductance dip. While we observe that all temperature dependent data scale onto a single curve, we also find that the characteristic scaling exponent α is widespread between 0.1 and 0.9. This behavior cannot be easily understood within the context of a Luttinger liquid. On the other hand, our data reveal a correlation between α and a possible interfacial barrier between the metal contacts and the nanotubes. We point out the importance to gain a better understanding of the interfacial properties between three-dimensional and one-dimensional metals and to include contact effects into the currently existing theory on Luttinger liquid.

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Carbon nanotubes (CN's) have attracted an increasing amount of interest over the past years. This is a consequence of their unique mechanical, thermal and electrical properties. For example they represent one of the rare cases of an ideal one-dimensional conductor. Due to this fact they are promising candidates for the study of Luttinger liquid (LL) theory.[1] Recently, a number of experiments have been published confirming the presence of a LL state in metallic CN's.[2–4] All of these measurements rely on the electrical transport properties of a carbon nanotube in contact with two electron reservoirs—metal contacts attached to the two ends of the CN. While on one hand high quality contacts are mandatory for a reliable electrical transport experiment, it is impossible to observe LL behavior in a clean and ''completely open'' system—one with no barriers at the metal contact/nanotube interface.[5] For the quantitative analysis of the LL state it is thus desirable to correlate experimental observations on Luttinger liquids with the type and strength of the contact barriers.

In this article the influence of barriers on the transport properties of ropes consisting of single-wall nanotubes is investigated. Transport in CN's bundles which is dominated by metallic tubes is studied within the framework of LL theory. We present evidence for the existence of barriers at the metallic nanotube/metal contact interface and relate those experimental findings with a LL-type scaling behavior of our transport data. We also point out similarities in this study with the recent observation that Schottky barriers are easily formed between metal contacts and semiconducting carbon nanotubes.[6,7]

Carbon nanotube bundles grown at Rice University were dissolved and sonicated in dichloroethane. Then, they were deposited on a $SiO_2(300\ \mathrm{nm})/Si(n++)$ substrate. Ropes were located by scanning electron microscopy relative to prepatterned alignment marks. A variety of different contact configurations have been fabricated using electron beam lithography and subsequent liftoff. Titanium is used to create top-lying electrodes and gold is used to contact tubes from the bottom, meaning that tubes are spun on predefined gold pads. Annealing of the sample at 800 °C for 30 s in argon gas below atmospheric pressure was used to reduce the two terminal resistance at room temperature.[7,8] Bundle diameters as characterized by atomic force microscopy were typically found to be approximately 10 nm. Electrical transport measurements were performed in a standard lock-in technique as well as by employing conventional current-voltage (IV) measurements. The sample resistance does not show any significant monotonic variations when a back gate voltage applied to the silicon substrate is swept at any temperature. This behavior is consistent with the notion that metallic tubes dominate electrical transport inside the ropes.[9] In fact, for zero gate voltage and drain voltages V_{SD} as applied in our experiments, semiconducting nanotubes, which are likely to be also present in the ropes, behave like insulators within the temperature range considered here[10] and do not contribute to the measured conductance.

Figure 1 shows the sample resistance at room temperature of devices exclusively contacted by titanium electrodes in the linear response regime for small applied voltages. The measured resistance R is normalized by the number N of current carrying tubes in the respective rope (R is multiplied by N). The value N is estimated from the measured current according to the current saturation model for every rope individually.[11,12] The normalized experimental resistance $N \times R$ varies from 20 to 120 kΩ. Because of the presence of many metallic tubes inside every rope, the measured resistance can be viewed as an effective average resistance per tube. There is no obvious correlation between the rope source-drain separation given by the distance between the annealed contacts and the device resistance. Accordingly, the

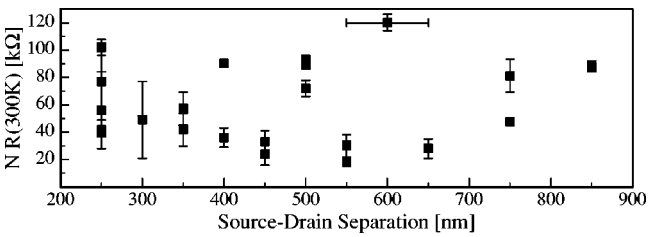

FIG. 1. Room-temperature device resistance as a function of source-drain separation for samples made with annealed titanium contacts.

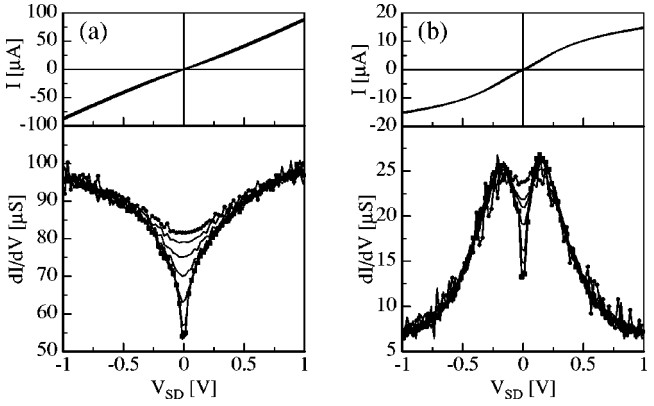

FIG. 2. Upper panels: Typical temperature dependent IV curves taken at 2.5, 10, 25, 50, 100, 200, and 250 K, respectively. Lower panels: Differential conductance calculated from the IV curves. A temperature dependent zero bias anomaly occurs. Here, sample (a) is made with bottom-lying gold electrodes and (b) with top-positioned annealed titanium contacts. The total number of contributing tubes is $N \geqslant 4$ in case (a) and $N=1$ in (b).

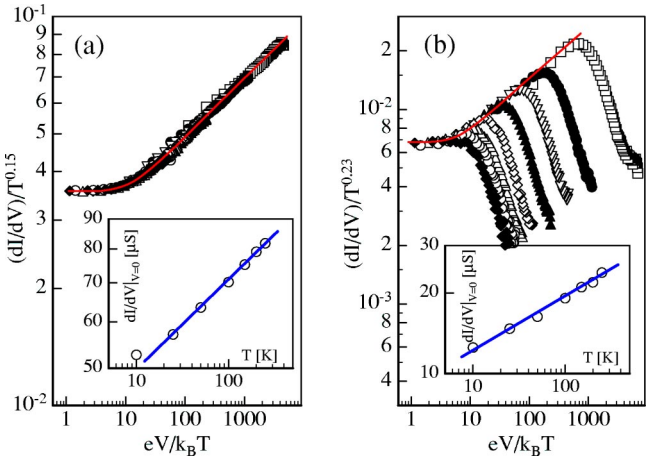

FIG. 3. Scaling plot of IV curves from Fig. 2 (same temperature range). (a) All data fall onto one scaling curve (highlighted as guide to the eye). (b) Scaling is only found for the dip region (V_{SD} <0.2 V), at larger bias current saturation suppresses the scaling behavior. Insets: The temperature dependence of the zero bias conductance showing power law behavior.

observed variations cannot be attributed to scattering inside the tube which should show a distinct length dependence. It is rather a consequence of the formation of contacts of non-reproducible quality. As in the case of other molecular systems (for a review see, e.g., Ref. 13) making a reliable contact to a carbon nanotube is challenging and often masks the observation of the intrinsic transport properties.

More information can be gathered from the temperature dependent IV curves as displayed for two samples in the upper part of Fig. 2. We find that the two types of characteristics are representative for *all* examined samples (in total 12) under investigation.[14]

The bottom part of Fig. 2 shows the corresponding differential sample conductance dI/dV_{SD}. While both measurements are sufficiently different in terms of conductance values and overall shape, one can easily identify a common feature—a dip in the dI/dV_{SD} characteristics—for small V_{SD}. Increasing the temperature or the bias increases the sample conductance thus suppressing the dip independent of the type of sample under investigation. Such dips called zero bias anomalies (ZBA's) are frequently found in carbon nanotubes and we will focus our attention on the ZBA as a measure of the formation of a Luttinger liquid (LL).

As has been pointed out[2,3] an indication of LL behavior of a system is the scaling behavior of the differential sample conductance. If one observes experimentally a relationship $[dI(V,T)/dV]\cdot T^{-\alpha} \sim F(e_0 V/k_B T)$ a Luttinger liquid state is inferred. F is the scaling functional according to Ref. 2 and α an exponent depending on the interaction strength of the electrons as well as on the contact geometry.[15] To study the origin of the observed ZBA in our experiment Fig. 3 shows the scaling behavior for the two types of samples in Fig. 2. While the actual values of α found for the two cases under consideration vary, both samples—as representative for the entire set of samples characterized—clearly show the expected LL scaling behavior as a function of temperature and bias voltage. Note that curves roll-off in case of sample (b) since optical phonon scattering[11] dominates already at low V_{SD} values. Accordingly, the roll-off occurs at higher values of $e_0 V/k_B T$ for lower temperatures where the ZBA is more pronounced. The envelope is marked and highlights the relevant scaling behavior. A pronounced power law is also visible in the temperature dependent linear response conductance shown in the inset of Fig. 3 as predicted by the universal scaling.

From these data one may conclude that our samples are yet another set of data supporting the existence of a LL state in case of carbon nanotubes and in agreement with very recent results by photoemission experiments that confirm the existence of a LL state in isolated single-wall nanotubes.[16] However, it is interesting to take a closer look at the parameter α. α describes the interaction strength between the one-dimensional system (the carbon nanotube) and the electron reservoirs. In the limit of ideal coupling α is expected to be zero. The maximum expected value of α is the so-called end exponent,[17] in case of a weakly coupled system consisting of a single single-wall carbon nanotube. Taking into account the fact that we are dealing with bundles of tubes, we calculate the maximum expected value of α in our experiment not to exceed $\alpha \approx 0.3$ using the theory by Egger.[18] However, experimentally we find α to range from 0.1 to 0.9. While there are so-called exponent addition rules[3] that could in principle explain larger exponents, their origin—kinks in nanotubes—could not be found to be present in our samples.

At this point it is important to reassess the assumptions that enter into the theoretical predictions on Luttinger liquid formation. Among other things, it is assumed that the transmission probability for charge injection from the reservoirs into the tube is *energy independent*. Only under this condition can the entire temperature and bias voltage dependence as observed in the ZBA be attributed to the LL state. An ohmic contact obviously fulfils the above criteria. However, there is an ongoing debate about the actual interface between a three-dimensional metal contact and a one-dimensional object as a carbon nanotube. In case of a semiconducting car-

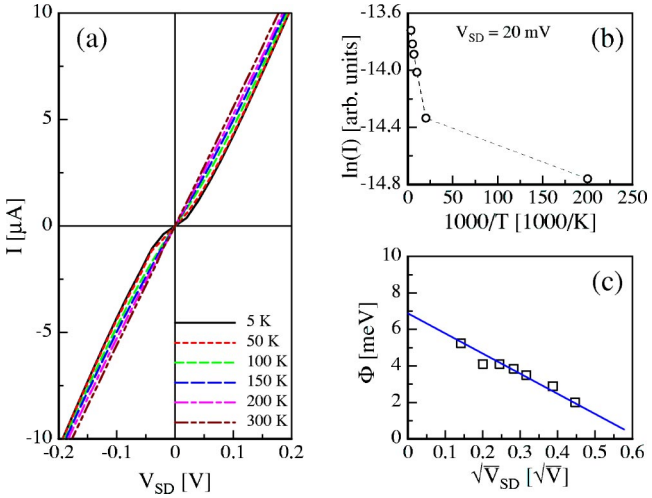

FIG. 4. (a) Temperature dependent IV curve for a sample with annealed titanium electrodes. (b) Current I taken at bias $V_{SD}=20$ mV and different temperatures. The linear slope for the data at temperatures $T>100$ K is used to calculate the barrier height. (c) Effective barrier height as a function of the bias voltage V_{SD}.

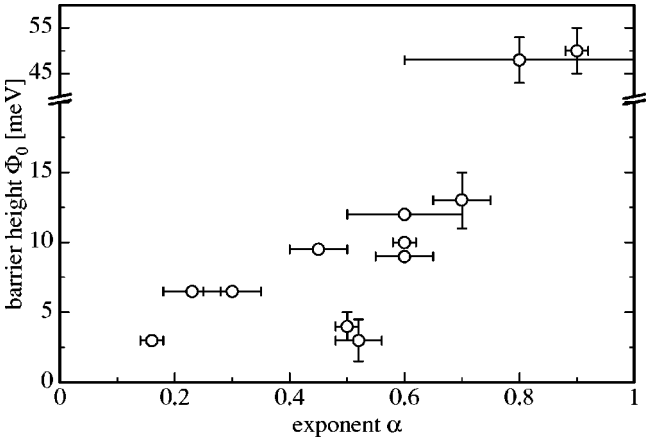

FIG. 5. Barrier height taken at zero bias versus scaling exponent α. The same trend is found for all samples regardless of contact type: The higher the barrier the larger the exponent.

bon nanotube it was pointed out that Schottky barriers can exist at this interface[6,19] and that these barriers can in itself result into a zero bias anomaly.[7] Since charge screening in one-dimensional (1D) metals is much less efficient than in their three-dimensional (3D) counterparts,[20] it may not be justified to describe a 3D/1D metal interface as a barrier free transition region. In fact, the ''resistivity dipole''[21] that occurs at a 3D/3D metal interface for metals with different work functions just over a few angstroms may not be negligible any more in case of a 3D/1D metal contact. Accordingly, one may attempt to characterize backscattering at this interface in terms of a barrier (similar to a Schottky barrier) with a characteristic height Φ_0 and a width given by the screening length in the 1D metal. In this case, charge injection from the 3D metal into the 1D region would *not* be independent of temperature or applied bias.

In order to explore this hypothesis, we have performed an analysis similar to what is typically done in case of a semiconductor/metal interface to characterize a Schottky barrier. Figure 4 summarizes the approach we have used. ''Arrhenius-type'' plots as shown in Fig. 4(b) were employed to extract a V_{SD} dependent barrier height Φ [see Fig. 4(c)], that can be used to extract the actual barrier height Φ_0 for zero applied bias. Interestingly, (i) I shows a clear exponential dependence on the inverse temperature for high enough T and (ii) Φ is indeed proportional to $\sqrt{V_{SD}}$. Both of these findings are typical for barrier dominated transport.[22] While the term ''Schottky barrier'' is certainly inadequate to describe the metal/metal interface, it is curious to note that our experimental data can be equally well understood in terms of barrier dominated transport as in the context of Luttinger liquid theory. In one case the entire temperature and voltage dependence is attributed to the contacts and in the other case it is the formation of a correlated electron system inside the tube which gives rise to the observed zero bias anomaly.

Last, we want to illustrate how the two descriptions relate to each other. Within the framework of LL theory we have extracted the exponent α for the 12 samples under investigation. Those samples differ in terms of contact type, number of contributing tubes, and source-drain separation. Then we used the same samples to extract a hypothetical barrier at the nanotube/metal interface. The result is displayed in Fig. 5. There is a clear correlation between α and Φ without any apparent impact of the actual sample layout. α scales with Φ and both values cover a significant range.

Currently it is unclear whether our observations imply that the zero bias anomaly observed in metallic nanotubes can be entirely explained as a result of barrier dominated current injection or if the theory on LL state has to be extended to explain larger values of α as found in this article. In any case it seems to be relevant to establish a more detailed understanding of the impact of contacts in one-dimensional structures.

In conclusion, we presented temperature and bias dependent transport measurements on ropes of single-wall carbon nanotubes. A pronounced zero bias anomaly is found as the common feature in the IV characteristics for all samples regardless of the actual contact positions and materials. The data were analyzed according to the well established scaling behavior within the Luttinger liquid theory. Reasonable agreement was achieved except for the absolute value and the variation of the scaling exponent which could not be explained in the framework of existing theories. Another approach based on transport properties of barriers at the nanotube/metal contact-interface was used to examine the ZBA. Interestingly, our data could also be described within this context. The clear correlation between the two findings shows that there is a need for a deeper understanding of the impact of contacts on the transport properties of nanotubes.

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