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Multiwalled carbon nanotubes are ballistic conductors at room temperature

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Abstract. Following the experiments of Frank et al. [1], which demonstrated quantum transport in multiwalled carbon nanotubes, there have been several experiments that appear to contradict the main conclusion of that paper, which is that the transport of a MWNT at room temperature is ballistic. Here we demonstrate that the intrinsic resistance of clean-arc-produced carbon nanotubes is at most $200\,\Omega/\mu\text{m}$, which implies that the momentum mean free path is greater than $30\,\mu\text{m}$, which in turn is much larger than the tube length. This implies that these tubes are ballistic, according to the standard definition of ballistic transport. We also show that the contact resistance with mercury is quite large: a nanotube in contact with Hg over 100 nm of its length still represents a $3000\,\Omega$ resistance.

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Transport in carbon nanotubes is both fascinating and controversial. Nanotubes have been shown to be (depending on the conditions and environment) both ballistic [1] and diffusive [2–5], and SWNTs appear to be p-doped due to the contact [6] or from atmospheric gasses [7, 8]. They are either metallic or semiconducting depending on the helicity [9].

Frank et al. [1] demonstrated several important transport properties in arc-produced MWNTs in a very simple experiment that involved contacting the nanotubes by dipping them into a liquid metal [1, 10]. The tubes protruded from a nanotube fiber, which was recuperated from nanotube arc deposits [11]. The tubes were not processed in any way in order to avoid contamination or damage. The conductance of the nanotubes was measured as a function of the depth L into the liquid metal. The resistance of these tubes was found to demonstrate flat quantized conductance plateaus (i.e. sample-length-independent resistance) and the tubes could sustain very large currents [1]. The independence of the conductance values on the tube diameter (which were close to G_0; values near $1/2G_0$ were also observed), the flatness of the plateaus and the very large currents (up to 1 mA through a tube) led Frank et al. to conclude that the transport was confined to the outer layer of the tube and that the transport was ballistic [1]. It appeared that the conductance of the outer layer was G_0 rather than $2G_0$, as expected on theoretical grounds [9, 12]. At the time of publication, it was known that nanotubes were conductors (from numerous experiments), however ballistic transport had not been observed under any conditions.

Subsequent experiments confirmed that only the surface of MWNTs transport the current and that very high current densities can be applied [2, 13]. Aharonov Bohm experiments [13] showed that at most only the outer two layers conduct. Furthermore, high currents will destroy the outer layers only. Since only one in three layers is expected to be metallic, it is expected statistically that only a few layers at the top can participate in the transport. Our present experiments strongly suggest that only the outer layer participates, with only the expected conductance value. However, the key result, that the transport is ballistic was contradicted in at least two experiments [2, 3]. In the experiments by Schonenberger et al. [2], it was found that the resistance per unit length of MWNTs is of the order of $5\,\text{k}\Omega/\mu\text{m}$ while the experiments of Bachtold et al. show even higher values (of the order of $10\,\text{k}\Omega/\mu\text{m}$) [3].

Very recently, Tinkham's group conclusively demonstrated that SWNTs are ballistic conductors by showing the quantum interference effect in the current between two electrodes [13]. The fact that SWNTs are ballistic and MWNTs are not is counter to general arguments which all tend to favor the MWNTs to have longer mean free paths [2, 15]. Here, we focus on direct measurements of ϱ, the resistance per unit length of MWNTs which can be converted to the momentum mean free paths. We concentrate on the shape of the conductance steps. It is important to realize that the mean free path is the critical parameter that determines the ballistic aspects of the conductance [16] and not the total resistance of the system, which also includes spurious contact resistances, as

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shown below. (Note that in the standard definition, ballistic conduction occurs if the momentum mean free path is longer than the length of the conductor [16].)

We have found in our investigations that some arc deposits are much richer in nanotubes presenting quantized plateaus (G_0 steps) than other deposits made in the same apparatus [17]. We believe that the discrepancies are due to the amounts of graphitic debris, which in turn determines the contact of the nanotube with the fiber. However, we never observed step heights substantially greater than G_0. Moreover, the plateaus were very flat in all cases (independent of the plateau values).

Here we demonstrate from detailed analysis of the conductance plateaus that the resistances per unit length are less than 200 Ω/μm.

A conductance plateau [1] is shown in Fig. 1a, which shows the conductance of a nanotube as a function of L, which is the depth that the nanotube is submerged in the Hg. This is a typical example, and one of a series of 60 measurements of this plateau, which are quite reproducible. The structure shows a jump just after contact followed by a convex rounding, followed by an extended flat plateau. For a classical diffusive system the step would be modeled as:

$$R_{\mathrm{tube}} = (L_0 - L)\varrho = {G_{\mathrm{tube}}}^{-1} \qquad (1)$$

where L_0 is the length of the tube and ϱ is the resistance/length. Figure 1c shows $R(L)$ for a step and what is predicted from (1) using the values of ϱ from Shonenberger [2]and from Bachtold [3]. It is clear that ϱ for this step is much lower; in fact, the slope at the end of this plateau is 0.18 kΩ/μm. It is clear that (1) describes the step very poorly. A much better fit is obtained by taking the metal–nanotube contact into account. The metal–nanotube contact conductance is modeled as:

$$G_{\mathrm{con}} = (G_{\mathrm{tip}} + \gamma L) \qquad (2)$$

where G_{tip} represents the tip conductance which causes the jump in the conductance just after the contact is established. Hence, the total conductance is:

$$G^{-1} = {G_{\mathrm{con}}}^{-1} + {G_{\mathrm{tube}}}^{-1} + {G_{\mathrm{flat}}}^{-1} \qquad (3)$$

where G_{flat} includes remaining L-independent contributions like the nanotube-fiber contact, or the ballistic contribution (see below).

From Fig. 1a and b it is very clear that the rounded shape of the step is indeed due to G_{con}, which produces the convex shape. From a full analysis of the shapes for this step we find that $\varrho = -36 \pm 50\ \Omega/\mu\mathrm{m}$, and $\gamma = (200\ \Omega\ \mu\mathrm{m})^{-1}$. These are typical values. However, in order to be as conservative as possible we will ignore the contribution due to the contact (which clearly dominates, as evidenced by the curvature amply demonstrated above), and hence take the resistivity to be $\varrho < 200\ \Omega/\mu\mathrm{m}$. Nevertheless, this upper bound for ϱ is as much as two orders of magnitude lower than that found by Schönenberger and Bachtold.

The mean free path is found from these resistances from the Einstein relation [18]. From the Fermi velocity (8×10^7 cm/s) [14] and the theoretical density of states, we derive that the momentum mean free path:

$$\lambda_{\mathrm{m}} = 30\ \mu\mathrm{m}$$

Note that it can be shown [17] that the mean free path is related to ϱ by

$$\lambda_{\mathrm{m}} = 1/(nG_0\varrho) \qquad (4)$$

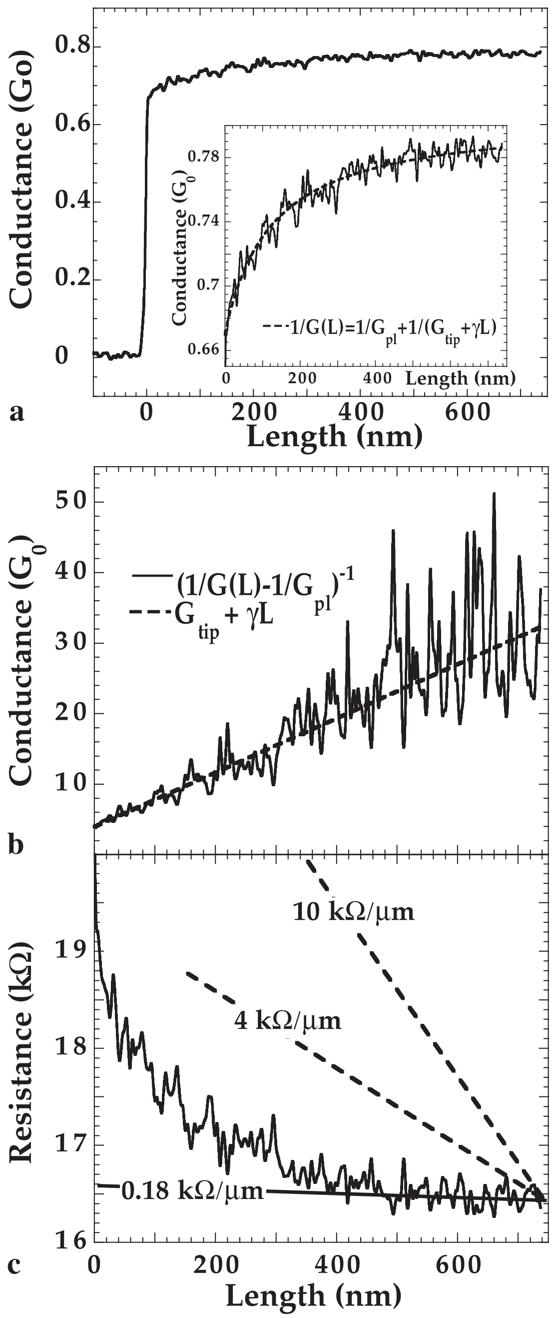

Fig. 1. Typical conductance step showing the conductance of a nanotube as a function of the distance L as it is lowered into the liquid Hg drop ($L = 0$ corresponds to the position of the conductance jump). **a** The conductance rises, at first abruptly, followed by a gradual increase that asymptotically approaches the plateau value G_{pl}. *Insert*, The shape is well approximated by the semi–classical resistance equation $G(L) = \{{G_{\mathrm{pl}}}^{-1} + (G_{\mathrm{tip}} + \gamma L)^{-1}\}^{-1}$, as shown in **b**, which clearly demonstrates the linear dependence of $(1/G(L) - 1/G_{\mathrm{pl}})^{-1}$ on L. **c** The resistance $R(L) = G(L)^{-1}$ of the same step. The obvious non-linear dependence on L indicates that the rounding is primarily due to γ and not to ϱ. Hence the extrapolated line with $\varrho = 0.18\ \mathrm{k}\Omega/\mu\mathrm{m}$ is an upper limit to the intrinsic resistance per unit length ϱ; the lines with $\varrho = 4\ \mathrm{k}\Omega/\mu\mathrm{m}$ and $\varrho = 10\ \mathrm{k}\Omega/\mu\mathrm{m}$ correspond to the intrinsic resistance per unit length reported in [2, 3] respectively

where n is the number of conducting channels (i.e. 2), which gives the same result as above. It is hence clear that nanotubes of micron lengths are ballistic conductors at room temperature. Even if as many as 10 layers were involved in the transport (so that $n = 20$), then the mean free path is still larger than 1 μm and the tube would still be ballistic on the micron length scale. We emphasize that this is really the worst possible case: all layers contribute to the transport (in contrast to several experimental findings) and the resistivity is over estimated (that is, the contact contribution the length-dependent resistivity is ignored: all the dependence is ascribed to the intrinsic nanotube resistivity). Hence, on the basis of this analysis, these free-standing clean nanotubes are indeed ballistic conductors at room temperature.

From the above analysis it is also clear that very large contact between the nanotube and the metal is required in order to ensure high transmission coefficients (i.e. low contact resistances). From Fig. 1b, a nanotube in contact with Hg over 100 nm of its length still represents a 3000 Ω resistance. This effect will be important if nanotubes are incorporated into electronic circuits.

We point out that similar relations as those above can be derived in the Landauer formalism [16], where the series resistance is replaced by sequences of scatterers with their related transmission and reflection coefficients [16, 17]. The conclusions remain the same from this analysis as those from the analysis above, however an additional term in the conductance (1) appears, which relates to the ballistic conductance of the system.

The nanotube–fiber contact is not well characterized and consists of a nanotube which makes contacts with a compact bundle of packed nanotubes. This complex system can be modeled. If we assume that each contact of the nanotube with a tube in the fiber is very good, which means that electrons at each nanotube–nanotube contact are transmitted (and reflected) into all the possible available channels with equal probabilities, then the total transmission coefficient of this contact is found [17] to be at most 0.6. Hence it is quite likely that the 'missing' conductance channel is due to the reflections at the nanotube–fiber contact. This also explains the observed variations in the plateau values and that the maximum observed plateau value is slightly greater than G_0. Lower plateau values (with otherwise flat plateaus) are due to poorer contacts with the fiber. Significantly sloping plateaus are only found when the nanotubes are visibly contaminated with graphitic debris, as observed in in situ electron microscopy experiments [17].

The discrepancy of our measurements with those of Schonenberger [2] and Bachtold [3] still needs to be explained. It should first of all be pointed out that our measurements directly address the variation of the resistance as a function of the length of the nanotube, and hence can directly be related to mean free paths without the use of arguments involving temperature dependencies and magnetic field dependencies. This difference in approach could point to an explanation of the discrepancies. However, we believe that the fundamental difference is in the processing of the nanotubes [19]. For example, surfactants were used to purify the nanotubes [19, 20] and if the surfactant is subsequently not removed (by extreme temperatures in vacuum or by burning off in air [21]) then this coating is very likely to profoundly affect the transport by causing significant scattering, since it is now known that adsorbed molecules on the surfaces affect the resistance [7, 8, 17].

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