Ensemble averaging of conductance fluctuations in multiwall carbon nanotubes

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New Journal of Physics 6 (2004) 27
Received 30 July 2003
Published 27 February 2004
Online at http://www.njp.org/ (DOI: 10.1088/1367-2630/6/1/027)

Abstract. We report resistance measurements for a single multiwall carbon nanotube as a function of gate voltage and perpendicular magnetic field. The tubes were trapped onto pre-patterned Al electrodes by means of an ac electric field. Magnetoresistance traces measured for various values of the gate voltage were averaged, which corresponds to an ensemble averaging of conductance fluctuations induced by quantum interference. The ensemble averaging decreases the conductance fluctuations, while leaving the weak localization contribution to the resistance unchanged. Our data can be consistently interpreted in terms quantum transport in the presence of a weak disorder.

Carbon nanotubes (CNTs) have attracted strong interest from technological and fundamental points of view. They have proven to be very promising for many applications, such as long-time stable-field electron emitters \cite{1, 2}, and can serve as the basic elements of future molecular electronic devices \cite{3, 4}. Therefore, it is highly desirable to have a good understanding of the electronic properties of CNTs as well as a method for constructing more complex devices from them. The latter would necessarily involve the controlled placement of individual tubes.

The specific electronic properties of CNTs are closely linked to their tubular topology. Their conduction characteristics vary between those of quantum wires and quantum dots depending on the contact resistance to the leads, which are made from classical metals. A wide range of quantum dot phenomena ranging from full Coulomb blockade to Kondo-like behaviour has been explored \cite{5}--\cite{7}. With highly transparent contacts, single-wall nanotubes (SWNTs) show pronounced signatures of ballistic transport and electron–electron interaction effects \cite{8}. Semiconducting SWNTs have successfully been used as molecular transistors. In contrast, the
properties of multiwall carbon nanotubes (MWNTs) with transparent contacts are still disputed. Ballistic transport [9] as well as signatures of diffusive quantum transport have been reported [10, 11]. It is not yet clear how the scenario of strictly one-dimensional (1D) transport and that of quantum interference in a magnetic field can be combined into a consistent picture. A strictly 1D wire has no phase space for trajectories enclosing magnetic flux. The quantum interference observed in a transverse magnetic field is a strong indication of the existence of 2D quasi-classical trajectories within the graphene cylinders. This requires the presence of several 1D subbands at the Fermi energy, which allow propagation around the tube circumference and intersubband scattering. Hence, the Fermi energy of the MWNTs must be shifted significantly away from the charge neutrality point, where maximally two degenerate subbands with propagation along the tube axis exist. Indeed, evidence for hole doping of MWNTs in air has been reported [12].

In this paper, we investigate whether a consistent description of the conduction properties of MWNTs, in terms of diffusive quantum transport, is possible. The fundamental signatures of this type of transport are universal conductance fluctuations (UCFs) and weak localization (WL). We use a very efficient gating of the MWNTs to generate a statistical ensemble of magnetoresistance (MR) traces and to check whether ensemble averaging of the MR traces occurs in the way expected for diffusive quantum transport. Ensemble averaging also allows us to separate the WL contribution from the conductance fluctuations. The results can indeed be consistently explained by the interplay of 1D WL (see [13] and references therein) and UCFs. In addition, we observe the other contributions to MR and their origin requires further investigation.

The MWNTs investigated were grown by arc discharge [14]. To obtain single tubes, the starting material was suspended in isopropanol using supersonic vibration. A very dilute suspension was used for the experiment to keep the deposition rate low. For a controlled placement of MWNTs on top of the gate electrodes, we used trapping by an ac electric field, similar to the method of Krupke et al [15]. On top of an oxidized Si wafer (oxide thickness, 400 nm), we prepared a grid of Au alignment marks as well as Al-trapping electrodes, together with Al gate fingers, by means of electron beam lithography. The thickness of the Al layer was 25 nm, the width of the gate electrode was 800 nm and the trapping electrodes were spaced by 1.8 \( \mu \)m. Owing to the small thickness of the native oxide layer of Al, a strong electrostatic coupling between the electrodes and the nanotube was expected.

The samples were bonded into a chip carrier and connected in series with a 350 M\( \Omega \) resistor. A droplet of nanotube suspension was put onto the chip, and a voltage of 10 V\(_{\text{rms}}\) at a frequency of 3 MHz was applied. After 3 min, the suspension was removed and the voltage was turned off. Subsequently, the exact position of the tube relative to the alignment marks was determined by scanning electron microscopy (SEM) inspection, which was used in the fabrication of 80-nm-thick ohmic gold contacts in a final lithographic step.

In agreement with Krupke et al [15], we also observed that trapping works best both at high electrical fields and at high frequencies. The native oxide layer on the Al electrodes prevents electric contact and, thus, the nanotube deposition cannot be monitored by using electrical current. Nevertheless, the trapping seems to be self-stopping, i.e. usually only one tube at maximum is trapped at the same pair of electrodes. We assume that the tube causes a capacitative shortcut, which leads to a breakdown of the trapping field and, in this way, prevents other tubes from attaching.

A typical sample, processed as described above, is shown in figure 1. A single MWNT is trapped across the Al gate electrode and is contacted by Au contacts patterned in a further lithographic step. In contrast with Al, the gold electrodes on top of the tube provide a good
Figure 1. Scanning-electron microscopy image of a typical sample. A single multiwall carbon nanotube is trapped between two Al electrodes and is contacted with 400-nm-spaced Au fingers from above. The Al finger under the tube serves as a backgate.

electric contact, with a total resistance of 6 kΩ at room temperature. The length of the tube was 2.1 µm, and the contact spacing was chosen to be 400 nm. The distance between the tube and the gate electrode was determined by the thickness of the native Al₂O₃ layer, which is typically of the order of 3 nm. The gate efficiency at such a small distance is higher compared with conventional techniques, such as degenerately doped Si backgates having a typical distance of 100 nm.

Electronic transport measurements at low temperatures have been performed for a nanotube with a diameter ≈28 nm. Figure 2 shows the linear response resistance (R) as a function of the Al backgate voltage for temperatures in the range 40–1.7 K in zero magnetic field and for gate voltages between −3 and 2 V. An aperiodic fluctuation pattern in R arises with decreasing temperature. This pattern has been interpreted previously as UCFs [16], which are thermally averaged as the temperature is increased. UCFs as a variation of gate voltage arise from the shift of the Fermi wavelength in a static scattering potential. Figure 3 shows a gate sweep of the same sample as in figure 2, under similar conditions, but measured 1 day later. Thermal cycling to about 80 K causes significant changes in the gate characteristics when compared with figure 2, which is a typical signature of UCF. This result indicates a partial scrambling of the interference pattern by the thermally activated motion of scatterers between two cool downs.

For a further substantiation of the presence of diffusive quantum transport, we performed magnetotransport measurements with a magnetic field transverse to the tube axis. MR curves have been taken for various fixed-gate voltages and fields from −10 to 10 T at a temperature of 1.7 K. The values of the gate voltage are marked by arrows in figure 3. Note that the field sweeps have been performed at gate voltages corresponding to UCF peaks and dips as well as at intermediate points. A plot of the resulting set of MR curves is shown in figure 4. Each curve reveals a symmetric peak in the MR located at B = 0 T. This negative MR can be well explained in terms of WL [10, 16]. WL originates from the destructive interference of pairs of time-reversed quasi-classical paths, which is cancelled in an external magnetic field of sufficient
Figure 2. Two-terminal resistance of a MWNT as a function of gate voltage for 1.7, 5, 10, 15, 20 and 40 K (from top to bottom). The curves are offset for clarity.

Figure 3. Gate sweep after thermal cycling to 300 K of the same MWNT as in figure 2. The temperature was $T = 1.7$ K. The arrows indicate the positions of the magnetic field sweeps shown in figure 4.

strength. The characteristic field for the suppression of WL has the same value of $\approx 1$ T for all the curves in figure 4. For higher fields, aperiodic fluctuations appear, which can again be identified as UCFs. The UCF amplitude at this temperature is comparable with the zero field WL peak, which indicates that the phase coherence length is less than the tube length, but roughly of the
same magnitude, since $\delta G_{WL} \propto (l/\Phi_1/L)$, whereas $\delta G_{UCF} \propto (l/\Phi_1/L)^{3/2}$ [17]. In some traces, the UCFs also affect the shape of the MR peak at zero magnetic field.

A closer look at the UCFs reveals that peaks appear primarily in the MR corresponding to enhanced backscattering, whereas comparable dips are absent from the investigated ranges of magnetic fields and gate voltages. Each value of the gate voltage, and hence of the Fermi level of the tube, corresponds to a different Fermi wavelength and, thus, a change of the phase shifts between different scatterers. If the change in gate voltage, and hence in $E_F$, is sufficiently large, a complete scrambling of the interference pattern can be achieved. Adjacent peaks and dips sometimes show a similar magnetofingerprint, differing mainly in the average resistance. The latter seems to be more sensitive to small changes in the gate voltage compared with the pattern of the magnetofingerprint itself. The ensemble average over all MR curves in the upper panel of figure 4 is plotted in the lower panel. The UCF amplitude is substantially decreased, whereas the zero field peak remains after the averaging procedure. This can be taken as additional evidence that the latter is indeed caused by WL. Besides the WL peak, a rather wide plateau of enhanced resistance remains between 4 and 8 T. This behaviour may be caused by the effect of a transversal magnetic field on the density of states and hence on the MR, as discussed in [18, 19]: in SWNTs of small diameter, peaks in the density of states as a function of the transversal magnetic field are predicted, which are strongly damped as the diameter of the tube increases. Nevertheless, structures in the MR are expected when the cyclotron radius becomes

Figure 4. Upper panel: MR of the same sample and cool down as in figure 3. Different voltages were applied to the Al gate and the magnetic field was applied perpendicular to the tube axis. The curves are offset for clarity. Lower panel: ensemble average of all MR traces.
comparable to the tube diameter; this may be related to the broad resistance maximum around 6 T of the averaged curve in figure 4. In a smaller magnetic-field interval, the ensemble averaging has also been performed at 20 and 40 K to explore the temperature dependence of the WL peak. Figure 5 shows the averaged MR curves obtained this way from $-1.5$ to $1.5$ T. With increasing temperature, the WL peak broadens and its height decreases, as expected from the temperature dependence of the electronic phase coherence length $l_{\Phi1}$. If the $l_{\Phi1}$ value is comparable to the tube diameter but lower than the tube length, 1D WL theory is appropriate for interpretation. This result is in agreement with the findings of other groups [20]. The conductance variation $\delta G_{\text{WL}}$ for a wire of width $W$ caused by WL is given as

$$\delta G_{\text{WL}} = -\frac{e^2}{\pi \hbar L} \left[ \frac{1}{l_{\Phi1}^2} + \frac{W^2}{3 l_{m}^4} \right]^{-1/2},$$

where $l_{m} = (\hbar/eB)^{1/2}$ denotes the magnetic length and $L$ the length of the nanotube [27]. Since carbon is a very light element, we have neglected spin–orbit scattering here. We have performed fits of equation (1) to our averaged MR curves using $L = 400$ nm. Using an effective width, $W$, of the tube as an additional fitting parameter yields the best results for $W = D/2 = 14$ nm, where $D$ is the diameter of the tube. This occurs quite probably due to flux cancellation effects [17] and is in accordance with Schönenberger et al [16]. The fitted curves reproduce the measured curves quite well, and the phase coherence lengths obtained this way are 150, 80 and 50 nm for 1.7, 20 and 40 K, respectively. These values are plotted in figure 6. We note that the use of $W = D$ instead of $D/2$ results in similar values of $l_{\Phi1}$ if the fit is restricted to the magnetic field range with $l_{m}^2/W \ll l_{\Phi1}$, where the WL expression is independent of $W$. Thus our results for $l_{\Phi1}$ are only weakly dependent on the precise values of $W$.

The rms amplitude $\delta G_{\text{rms}}$ of the fluctuations allows us to extract the phase coherence length $l_{\Phi1}$ in an independent way. From the WL measurements, we derive that the $l_{\Phi1}$ value is lower than the tube length $L$. In this situation, the $l_{\Phi1}$ value can be less than or greater than the thermal length $l_T = (D\hbar/k_BT)^{1/2}$, where $D$ is the diffusion constant. If $l_{\Phi1} \ll L$, $l_T$ is valid, then $\delta G_{\text{rms}}$ depends

Figure 5. Averaged MR curves for 1.7, 20 and 40 K (from top to bottom). Lines are fits to 1D WL theory.
Figure 6. Temperature dependence of the phase coherence length derived from WL measurements (△) and from the amplitude of the UCFs (●). The line is a fit to $T^{-1/3}$ power law.

on $l_{\Phi}$ by the relation (see [21, 22])

$$\delta G_{\text{rms}} = \sqrt{12} \frac{e^2}{h} \left( \frac{l_{\Phi}}{L} \right)^{3/2}.$$  \hspace{1cm} (2)

In figure 6, the temperature dependence of $l_{\Phi}$ using equation (2) is included. The values of $l_{\Phi}$ extracted from UCF and WL agree within 30%, which is reasonable given that $l_T$ is still relatively close to $l_{\Phi}$ (see below). For 1 K and 20 K, the value predicted by WL is slightly higher. If we assume that $l_T \ll l_{\Phi} \ll L$, the relation between $\delta G_{\text{rms}}$ and $l_{\Phi}$ is given by [22, 26]

$$\delta G_{\text{rms}} = \left( \frac{8\pi}{3} \right)^{1/2} \frac{e^2}{h} l_{\Phi}^{1/2} \frac{l_T}{L} l_{\Phi}^{3/2}.$$  \hspace{1cm} (3)

It follows that $l_{\Phi} \propto \delta G^2 T$. If we substitute the measured values for $\delta G(T)$, derived from figure 2, into this relation, we obtain an increase in $l_{\Phi}$ with increasing temperature. Such an increase contradicts the WL results and, hence, this regime can be ruled out.

On the other hand, the ratio of $l_{\Phi}$ and $l_T$ can be estimated directly from the measurements. It is possible to estimate the diffusion constant, and hence the thermal wavelength $l_T$, in the following way: fitting a $T^{-1/3}$ power law to the data yields $l_{\Phi} = 164 \text{ nm} K^{1/3} T^{-1/3}$. The good agreement with the data suggests that the dephasing predominantly occurs by quasi-elastic electron–electron scattering as observed previously in similar systems [20, 23, 24]. This implies that the diffusion constant can be extracted from the prefactor of $l_{\Phi}(T)$:

$$l_{\Phi}(T) = \left( \frac{GDLh^2}{2e^2k_B} \right)^{1/3} T^{-1/3},$$  \hspace{1cm} (4)

where $G$ is the conductance of the tube [25]. In this way, we estimate that $D \approx 70 \text{ cm}^2 \text{s}^{-1}$ and, using the Fermi velocity $v_F = 10^6 \text{ m} \text{s}^{-1}$ of graphite, the elastic mean free path $l_e \approx 14 \text{ nm}$. The thermal wavelength $l_T$ then turns out to be of the order of 180 nm at 1.7 K and 40 nm at 40 K. Thus
we find that $l_T \approx \frac{l}{\Phi_1}$; this implies that equations (2) and (3) agree within 20% and demonstrates the consistency of our analysis.

In conclusion, we have contacted individual MWNTs with strong electrostatic coupling to Al gate electrodes. Electronic transport measurements at low temperatures, combining gate sweeps with magnetic field sweeps, were used to generate a statistical ensemble of magnetofingerprints. The amplitude of the UCFs can be combined with the ensemble-averaged low-field MR for a consistent determination of the phase coherence length. This provides additional evidence that transport in MWNTs is governed by quantum interference in a random potential. The origin of the disorder requires further investigation.

**Acknowledgment**

The authors thank G Cuniberti, R Schäfer and D Babić for fruitful discussions. We also thank M Buitelaar and C Schönenberger for experimental support and discussions. Funding by the Deutsche Forschungsgemeinschaft within the Graduiertenkolleg GRK 638 is acknowledged.

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