

Quantum Transport in a Multiwalled Carbon Nanotube

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We report on electrical resistance measurements of an individual carbon nanotube down to a temperature $T = 20$ mK. The conductance exhibits a $\ln T$ dependence and saturates at low temperature. A magnetic field applied perpendicular to the tube axis increases the conductance and produces aperiodic fluctuations. The data find a global and coherent interpretation in terms of two-dimensional weak localization and universal conductance fluctuations in mesoscopic conductors. The dimensionality of the electronic system is discussed in terms of the peculiar structure of carbon nanotubes.

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With the discovery by Iijima [1] of carbon nanotube (CN) structures, a new class of materials with a reduced dimensionality has been introduced. The nanotubes are made of coaxial graphitic cylinders. Each cylinder can be visualized as the conformal mapping of a two-dimensional (2D) honeycomb lattice onto its surface. Theoretical calculations predict that structural parameters, i.e., diameter, helicity, and number of concentric cylinders, strongly influence the band structure and hence the electronic properties of CNs [2]. This implies that a CN can either be a metal, a semimetal, or an insulator, depending on structural parameters. It has also been predicted that the presence of a magnetic field will strongly affect the band structure near the Fermi level [3].

Up to now no electrical resistance measurements on individual nanotubes have been reported. Our group succeeded to measure a microbundle (total diameter around 50 nm) of multiwalled CNs [4]. At higher temperature ($T > 1$ K) we observed a typical semimetallic behavior, consistent with the simple two-band model for semimetallic graphite. Song *et al.* reported on a large CN bundle exhibiting a behavior similar to that of disordered semimetallic graphite [5].

In this Letter we report on the first electrical resistance measurements performed on an individual CN. In zero magnetic field we observe a logarithmic decrease of the conductance with decreasing temperature at high temperature, followed by a saturation below $T \approx 0.3$ K. In the presence of a magnetic field, applied perpendicular to the tube axis, a pronounced and positive magnetoconductance (MC), i.e., an increase of conductance with increasing field, is observed. The temperature dependence of the conductance in a magnetic field can be described consistently within the framework of the theory for 2D weak localiza-

tion (WL) [6]. The effect of the magnetic field on the density of states predicted by Ajiki and Ando [3] is also observed. The mesoscopic dimensions of the CN result in the appearance of reproducible fluctuations of the MC with respect to magnetic field. These fluctuations show an amplitude and a temperature dependence consistent with the presence of universal conductance fluctuations (UCF) [7]. Our conductance measurements strongly support the idea that isolated multiwalled nanotubes behave as disordered mesoscopic 2D graphite sheets.

The carbon nanotubes were synthesized using the standard carbon arc-discharge technique [8]. High resolution transmission electron microscopy (TEM) images of our CN materials reveal a few nanotube bundles and many single tubes with an average diameter around 18 nm.

In order to contact a nanotube, the growth material is dispersed on an oxidized Si wafer covered with an array of large square gold pads. After evaporation of a thin gold film, a few layers of a negative electron resist, ω -tricosenoic acid, is deposited using the Langmuir-Blodgett (LB) technique. The scanning tunneling microscope (STM) is then used to locally expose the resist from the CN towards the predefined gold pads [9,10]. The unexposed parts of the LB film are dissolved in ethanol and the unprotected parts of the gold film are removed by Ar ion milling. The result of the STM lithography is an electrically connected CN. High resolution TEM investigations of CNs with comparable diameter were used to check that the CNs are not damaged by the ion milling process.

ac electrical conductance measurements at 15 Hz were performed in the mixing chamber of a dilution refrigerator in a magnetic field B perpendicular to the tube axis. We have been able to attach electrical contacts to four similar multiwalled CNs. All samples reveal the presence of

a pronounced positive MC on which reproducible fluctuations are superimposed. Here, we focus on the results for one particular CN, having a length $L \approx 800$ nm between the two voltage probes and a diameter $d \approx 20$ nm, as determined by atomic force microscopy. Three contacts were attached to our CN sample, which allowed us to check directly that the contact resistances were negligible compared to the total longitudinal resistance. The observed impedance was in the Ohmic regime for currents ranging from 10^{-11} to 10^{-9} A, and did not present any capacitive component.

In Fig. 1 we present the conductance G as a function of temperature for the CN. The conductance clearly shows a $\ln T$ dependence followed by a saturation at low temperature, while the MC is positive at all temperatures. The saturation of G occurs at higher temperatures in the presence of a magnetic field. A $\ln T$ dependence either can be indicative of weak localization [11] or disorder enhanced electron-electron interactions [12] in 2D systems or can be the signature of a Kondo anomaly due to the presence of magnetic impurities [13]. The presence of a Kondo effect is very unlikely, since this effect has never been observed in graphitic materials even containing detectable amounts of magnetic impurities [14]. Moreover, spectrographic analysis revealed that our CN material contains less than 10 ppm (detection limit) of magnetic impurities [15]. The data in Fig. 1 also rule out the presence of predominant disorder enhanced electron-electron interactions since they should exhibit an opposite behavior, i.e., a positive MC and a saturation of the low-temperature conductance which is independent of the magnetic field [6].

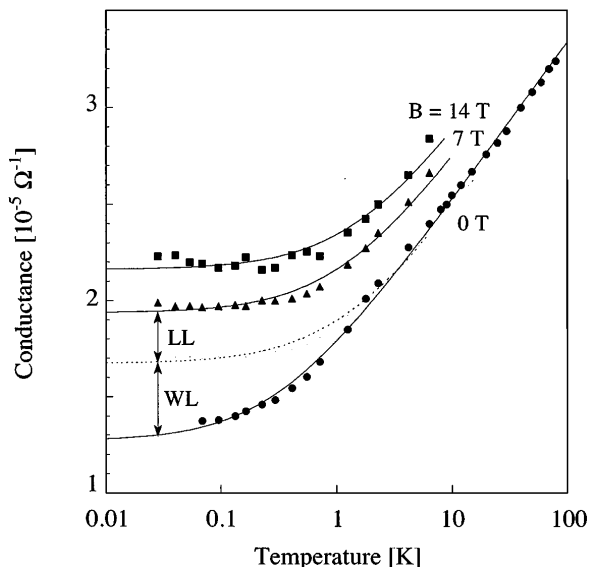


FIG. 1. Electrical conductance as a function of temperature at the indicated magnetic fields. The solid lines are fits by Eq. (1) with parameters $p = 1$, $n = 4$, and $T_c = 0.3, 1.1$, and 1.5 K at $B = 0, 7$, and 14 T, respectively. The dashed line separates the contributions to the magnetoconductance of the Landau levels and the weak localization.

We will now show that our results are consistent with the presence of 2D WL effects in the concentric graphite cylinders forming the CN. The WL effects result from the enhanced backscattering probability for the electrons due to the constructive interference between the partial electron waves traveling back to their original position along time-reversed paths [6]. Since the constructive interference is destroyed by inelastic scattering (by other electrons or phonons), the backscattering becomes much more pronounced at lower temperatures, where the inelastic scattering time $\tau_{\text{in}}(T) \propto T^{-p}$, with p depending on the dominant inelastic scattering mechanism.

At the lowest temperatures, the spin scattering at magnetic impurities, with a characteristic scattering time τ_s , will also contribute to the destruction of the interference effects [6]. The spatial extent of the interference effects is limited to the phase coherence length $L_\phi = \sqrt{D\tau_\phi}$, where D is the elastic diffusion constant and the phase coherence time τ_ϕ describes the combined effect of the inelastic and magnetic scattering: $\tau_\phi^{-1}(T) = \tau_{\text{in}}^{-1}(T) + 2\tau_s^{-1}$. For 2D materials the WL results in a reduction of the conductance which depends logarithmically on the relevant length scale $L_\phi(T)$. This implies that the WL causes a logarithmic decrease of G in the highest temperature range, where $\tau_{\text{in}}(T) \ll \tau_s$, and a saturation of G at the lowest temperatures, where $\tau_{\text{in}}(T) \gg \tau_s$. This is exactly the behavior which is observed in Fig. 1 at $B = 0$ [16].

Assuming that the spin-orbit scattering is weak [14], the WL theory can also account for the pronounced positive MC in the presence of a perpendicular magnetic field. Indeed, the magnetic field also contributes to the dephasing of the interference effects when the Landau orbit size $L_B = \sqrt{\hbar/eB}$ becomes smaller than the phase coherence length $L_\phi(T)$. For a sufficiently high magnetic field, i.e., when $L_B \ll L_s = \sqrt{D\tau_s}$, the applied magnetic field governs the saturation of the conductance at lower temperatures. This is in agreement with the observation in Fig. 1 that the saturation of G occurs at higher temperatures when increasing the magnetic field.

In contrast to crystalline graphite, the random stacking of graphene sheets in turbostratic graphite induces a 2D behavior of the electron system with respect to the WL effects [14]. The cylindrical structure of CNs produces a similar situation since the relative positions of carbon atoms in adjacent cylinders of a CN are randomized due to their specific geometry. Consequently, each individual graphite cylinder behaves as an individual intrinsic 2D system. In the presence of 2D WL effects, the conductance of the CN, contributed by n cylinders in parallel, is related to the 2D conductivity σ via $G = \sigma \pi n d / L$ and is given by [6]

$$G(T) = G_0 + \frac{e^2}{2\pi^2\hbar} \frac{n\pi d}{L} \ln \left[1 + \left(\frac{T}{T_c(B, \tau_s)} \right)^p \right]. \quad (1)$$

The quantitative analysis of the conductance using Eq. (1) provides us with values $p = 1$, $n = 4$, and T_c at different B (Fig. 1). The value $p = 1$ is in agreement with previous experiments [14] and indicates that the dominating inelastic scattering mechanism is likely to be disorder enhanced electron-electron scattering in 2D systems [17]. The value for n is likely to be smaller than the total number of concentric cylinders which are present in the multiwalled CN. Indeed, we make electrical contacts mainly to the outer cylinder and the coupling with the inner cylinders is very weak due to the very large anisotropy of the graphitic material. Moreover, nonconducting cylinders may also be present.

The observation of weak localization requires that elastic scattering dominates inelastic scattering. This generally limits the observation of weak localization as the temperature increases. However, contrary to common metals, weak localization was observed up to relatively high temperatures in 2D turbostratic graphite [14]. This comes from the basic differences between common metals and graphitic materials, including CNs, where important parameters such as screening, Debye temperature, and Fermi energy are drastically different.

The WL theory predicts that when $T \gg T_c$ the conductance becomes independent of B [6]. The data in Fig. 1 show that this is not the case and that there is an additional contribution to the MC of the CN which is independent of temperature. Ajiki and Ando [3] have predicted the formation of Landau states when a magnetic field is applied perpendicular to the nanotube axis. In particular, a Landau level should form at the crossing of the valence and conduction bands, thereby increasing the density of states at the Fermi level, and hence increasing the conductance. The resulting positive MC is expected to be temperature independent as long as $k_B T$ remains smaller than the width of the Landau level. Previous results obtained on a CN bundle [4] revealed a behavior consistent with these predictions. In the present work, we attribute the additional temperature independent positive MC to the same mechanism. Both 2D WL and "Landau level" (LL) contributions to the MC can be separated as illustrated in Fig. 1 for $B = 7$ T.

The temperature T_c , which is obtained from Eq. (1) corresponds to $L_s \approx L_{in}(T_c)$ for low magnetic fields and to $L_B \approx L_{in}(T_c)$ for sufficiently high fields. The data for $B = 14$ T allow us to determine that $L_{in} \approx L_B \approx 7$ nm at $T = T_c \approx 1.5$ K. Since $L_{in} = \sqrt{D\tau_{in}} \propto T^{-p/2}$, the zero-field data give $L_s \approx L_{in} \approx 20$ nm at $T = T_c \approx 0.3$ K. Thus, at the lowest temperatures L_ϕ is smaller than but comparable to L , implying that we may observe interference phenomena related to the mesoscopic dimensions of the CN.

In Fig. 2 we show the details of the MC behavior at different temperatures. At low temperature, reproducible, aperiodic fluctuations of the conductance appear, which are superimposed on the positive background caused by the

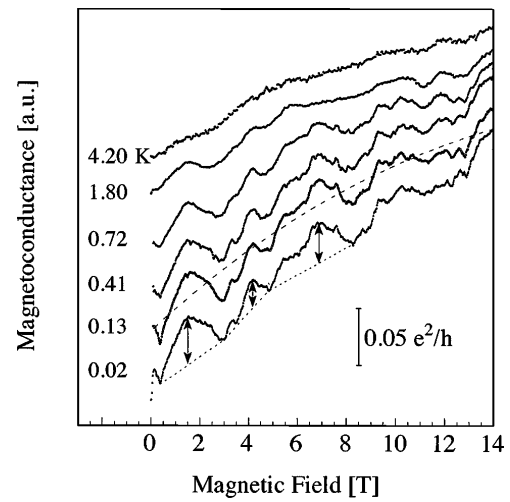


FIG. 2. Magnetic field dependence of the magnetoconductance at different temperatures.

WL and Landau level effects. Similar fluctuations have often been observed [7] in disordered metals and semiconductor structures and have been related to the tuning of sample-specific electron interference processes by the perpendicular magnetic field (Aharonov-Bohm effect) [18]. These fluctuations are well known as "universal conductance fluctuations." The rms amplitude of the fluctuations δG reaches the "universal" value $\text{rms}[\delta G] \approx e^2/h$ as soon as the sample size L becomes smaller than both the phase-coherence length L_ϕ and the thermal diffusion length $L_T = \sqrt{\hbar D/k_B T}$ [19]. When either L_ϕ or L_T becomes smaller than L , the shorter of these two length scales governs the amplitude of the fluctuations.

We will now show that the results presented in Fig. 2 are consistent with the stochastic self-averaging of the UCF which occurs in samples with $L \gg L_\phi$. Figure 3

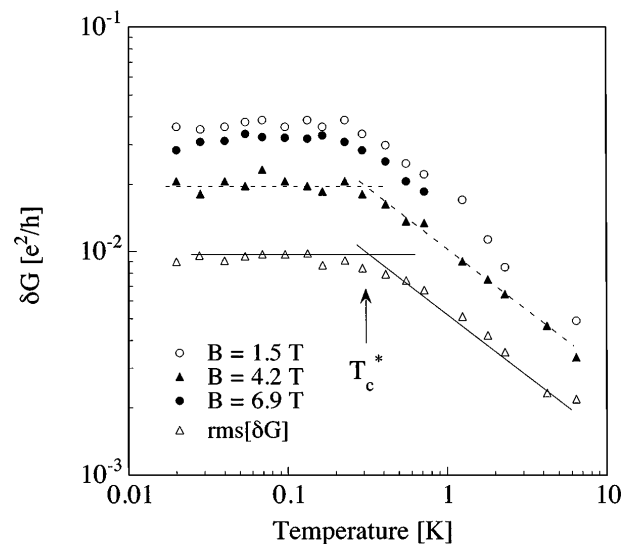


FIG. 3. Temperature dependence of the amplitude of δG for three selected peaks (see arrows in Fig. 2) as well as $\text{rms}[\delta G]$.

shows the temperature dependence of $\text{rms}[\delta G]$ which reaches a constant amplitude at low temperature and decreases following a $T^{-1/2}$ power law at higher temperature. In order to calculate $\text{rms}[\delta G]$, a smooth background G_B (dashed line in Fig. 2) has been subtracted from the MC data so that $\langle \delta G \rangle = \langle G - G_B \rangle = 0$ (the brackets refer to an average over the magnetic field). The saturation in the temperature dependence of the fluctuations occurs at a temperature $T_c^* \approx 0.3$ K (Fig. 3) which, in contrast to the T_c values discussed above, is independent of B . Since T_c^* coincides with the temperature $T_c(B = 0)$ at which L_ϕ saturates, we conclude that L_ϕ is the relevant length scale for both 2D WL and UCF.

Since for our CN sample $L_\phi < \pi d < L$, it can be divided into L/L_ϕ conductors in series and $n\pi d/L_\phi$ conductors in parallel, each of them producing fluctuations of the order e^2/h . The statistical self-averaging of the UCF results in fluctuations with rms amplitude [20]:

$$\text{rms}[\delta G] = 0.61 \frac{e^2}{h} \left(\frac{n\pi d}{L_\phi} \right)^{1/2} \left(\frac{L_\phi}{L} \right)^{3/2}. \quad (2)$$

With the geometrical parameters for our CN, Eq. (2) predicts $\text{rms}[\delta G] \approx 8.5 \times 10^{-3} e^2/h$ below T_c^* , in good agreement with the observed saturation value $\text{rms}[\delta G] \approx 9 \times 10^{-3} e^2/h$. Equation (2) also predicts that above T_c^* $\text{rms}[\delta G] \propto L_\phi \propto T^{-1/2}$ which is indeed observed up to $T = 10$ K.

We conclude that the electrical transport in a multi-walled carbon nanotube is governed by typical electron interference effects occurring in disordered conductors with a reduced dimensionality. The fact that nanotubes behave as disordered conductors is consistent with the study of annealing effects on the conduction electron spin resonance [21]. Despite the very small diameter of the nanotube, the cylindrical structure of the honeycomb lattice gives rise to a two-dimensional electrical conduction process. The temperature and magnetic field dependences of both the weak localization effects and the universal conductance fluctuations can be explained consistently in terms of the existing theoretical models, provided the typical cylindrical geometry of the carbon nanotube is taken into account.

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