Conductance in multiwall carbon nanotubes and semiconductor nanowires: evidence of a universal tunneling barrier.

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Electronic transport in multiwall carbon nanotubes and semiconductor nanowires was compared. In both cases, the non ohmic behavior of the conductance, the so-called zero bias anomaly, shows a temperature dependence that scales with the voltage dependence. This robust scaling law describes the conductance $G(V, T)$ by a single coefficient $\alpha$. A universal behavior as a function of $\alpha$ is found for all samples. Magnetococonductance measurements furthermore show that the conduction regime is weak localization. The observed behavior can be understood in terms of the coulomb blockade theory, providing that a unique tunnel resistance on the order of $2000 \Omega$ and a Thouless energy of about $40 \text{ meV}$ exists for all samples.

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There is intense interest in electronic transport in nanostructures in various contexts, from single electron transistors to carbon nanotubes, semiconductor nanowires, metallic nanoconstrictions or other molecular structures. In the presence of a tunnel junction, a non-Ohmic behavior of the conductance $G$, termed zero-bias anomaly (ZBA), is generally observed at low temperature.

For carbon nanotubes (CNT), the voltage dependence of the ZBA at low temperatures and high bias is a power law $G = G_V \cdot (eV)^\alpha$, and the temperature dependence at low bias is also a power law, with the same power coefficient $\alpha$; $G = G_T \cdot (kT)^\alpha$. Here $e$ is the electronic charge and $k$ is the Boltzmann constant. Under this approximation, the conduction properties $G_p(kT, eV)$ can then be described for each sample (at zero magnetic field) by a single scaling coefficient $\alpha$, and the two prefactors $G_V$ and $G_T$. Beyond this approximation, a more general description is given by a scaling function $f$, such that $G T^{-\alpha} = f(eV/kT)$.

The scaling law is presented in the CNT literature as a manifestation of an underlying physical mechanism. In the presence of a tunnel junction, a Coulomb Blockade (CB) effect is expected. In the case of an ultrasmall junction, CB is described by the environmental impedance $Z(\omega)$. In more extended tunnel junctions with disorder, the field and electrons propagate diffusively within the electrodes, and non perturbative methods should be used. Finally, in the case of 1D systems, Luttinger Liquid states are expected. In all three cases, the conductance takes an identical form (see Eq. 2 below) under a rather general hypothesis [1,10,11]. Furthermore, measurement of the conductivity under applied magnetic field show typical weak localization. Accordingly, it is possible to invoke either Luttinger liquid states [2,3,11,12, CB effects [2,3,13,14], or diffusion effects related to disorder and weak localization [2,13,14].

In order to clarify the situation, the method followed in this work is to correlate systematically the coefficient $\alpha$ to other experimental parameters, by performing a comparative study on a statistical ensemble of samples. In parallel with the CNT we present a systematic study of the scaling law occurring in semiconducting nanowires as a new argument in this debate.

We measured two sets of samples. The first set of about 50 samples is composed of nanotubes obtained by CVD on Ni or Co catalyst in a nanoporous alumina membrane (the process is described elsewhere [6,12]). The nanotubes are well separated (one nanotube per pore) and are connected perpendicularly to a Au, Ni or Co contact. The diameter of the nanotube is calibrated by the diameter of the pore. One or a few nanotubes are contacted in parallel. The anodisation techniques allow the diameter of the pores to be well controlled, from 40 nm down to 5 nm [6,15]. The length of the nanotube (controlled by the length of the catalyst electrodeposited inside the pores) was adjusted between about 1.5 $\mu$m down to 100 nm. The nanotubes are grown inside the pores by standard CVD technique with acetylene at 640 °C, after the electrodeposition of Ni or Co catalyst. The CNT are multiwalled. The top contact is made by sputtering, or evaporation, after the growth of the tubes, and after exposing the samples to air. Different materials and crystallinities have been used for the top contacts.

The second set of samples is composed of tellurium semiconductors (Te) obtained by electrodeposition in nanoporous polycarbonate or alumina membranes [12] of diameters $d=40$ nm and $d=200$ nm. At 200 nm, the nanowires should no more be 1D with respect to electronic transport (because the energy separation between quantum levels $\Delta E = (\pi \hbar)^2/(2m^* d^2)$, where $m^*$ is the effective mass, should be above the thermal energy). With the electrodeposition technique, a single nanowire can be contacted in situ with a feedback loop on the inter-
membrane electric potential $\eta$. Both contacts are free of oxides, due to the chemical reduction at the Te interfaces during the electrodeposition, and due to in the in situ contacts. The Te are contacted with Au or Ni: $\text{Au/Te/Au or Ni/Te/Ni}$.

The dynamical resistance measurements were performed with a lock-in detection bridge LR700 (using AC current of 0.3 nA for most samples to 10 nA for low resistive samples) and a DC current. DC resistance measurements were also made with a nanovoltmeter. The temperature is ranged between 4K and 200K. A superconducting coil gives a perpendicular magnetic field, ranging between $\pm 1.2$ T. This experimental protocol allows us to measure dynamical resistance as a function of DC current amplitude, perpendicular magnetic fields, and temperature.

The typical profile of the ZBA is plotted in Fig 1 for a Te semiconducting wire (a and c) and for a CNT sample ((b) and (d)). Two well defined conductance regimes are observed: the voltage dependence of the ZBA at low temperature and high bias $G = G_\ell (eV)^\alpha$ and the temperature dependence at low bias, which is also a power law with the same power coefficient $\alpha$: $G = G_T (kT)^\alpha$. The ZBA vanishes above 50K, but the temperature dependence is also valid at high temperature. A more general description (which shows the deviation to the simple power law approximation) is presented in the form: $GT^{-\alpha} = f(eV/kT)$ (Fig 1 (c) and (d)). A very large majority of samples exhibit the scaling law (48 CNT over 55 with enough length [6] and 13 Te nanowires over 14). This scaling law is very robust since samples are different from the point of view of the nature of the contacts, and the quality and nature of the nanowires or nanotubes. The CNT are contacted with Ni or Co catalysts [16] on the bottom. The top of the wire is contacted either with amorphous Ni, or with highly disordered Co (mixed hcp and cfc nanocrystallites) or with single crystalline cfc Co layer [16]. The coefficient $\alpha$ for Co electrodes is statistically larger than that for Ni (Fig 1 (f)).

Most of the resistances at room temperature are distributed from about 300 to 40 000 Ω (Fig 1 (e)). There are no statistical correlations between the resistances at room temperature and the coefficient $\alpha$. The conductance variations from one sample to the other are not due to the nature of the contact. Fig 1 (f) shows the corresponding histogram for $\alpha$. The first peak near $\alpha = 0$ is due to short CNTs, with a length $L \leq 300$ nm of the order of the thermal length (i.e. the CNT are screened by the contacts).

There are no statistical correlations between the resistance and the length or the diameter of the CNTs (not shown). We define a ratio $\eta = R(50K)/R(300K)$ as the resistance at 50K divided by the resistance at room temperature. The coefficient $\eta$ is a measure of the contribution of the electrodes and interfaces at high temperatures (i.e. with the exclusion of the contribution of the physical mechanism responsible for the ZBA). The parameter $\eta$ is correlated to the coefficient $\alpha$ (Fig 2 (a)), but the correlation depends strongly on the nature of the electrodes. A tendency is sketched by the straight lines in Fig. 2 (a), and the most important deviation is seen for CNT with single crystalline cfc Co. This shows that $\alpha$ is related to the contacts, and is not exclusively defined by the states of the wires or tubes. The coefficient $\alpha$ is hence dependent on extrinsic parameters. On the other hand, the coefficient $\alpha$ is correlated to the length of the CNT (as shown in reference [6]), which indicates that $\alpha$ depends also on intrinsic parameters (including defects).

However, the most important result of this study is the unique relation existing between the prefactors $G_\ell$, $G_T$ and $\alpha$ (Fig 2 (b)), whatever the nature of the samples. For each sample, the extrapolation at 1K gives the conductance $G_T/k^\alpha$ (plotted in Fig 2 (b)), and the extrapolation at 1V gives the coefficient $G_\ell/(eV)^\alpha$ (plotted in the inset of Fig 2 (b), same scale). All points align on the same curve.

The correlation between $\alpha$ and the prefactors is a priori not expected because there is no correlations between $G$ and $\alpha$ at room temperature. The function appearing in fig 2 (b) is a new universality exhibited by all measured samples, providing that the scaling law is measured. The discussion of the observed relation in terms of CB (curves fitted in Fig 2 (b)) follows.

In Fig 2(b), note that the difference between the fit in the main figure and the fit in the inset is about $(e/k)^\alpha \approx 10^{4\alpha}$, so that the two prefactors $G_\ell$ and $G_T$ are approximately equal. This means that the deviation from the approximation of the function $G_\alpha (eV, kT)$ in the two power laws is small even for intermediate regimes.

More information about the system, and especially about disorder and quantum diffusion, can be obtained by applying a magnetic field $H$ perpendicular to the wire or tube axis [2, 13, 14]. Only the magnetoconductance
localization is also strongly correlated to the coefficient α. However, the Fig 3 (c) shows that, surprisingly, the weak magnetic field (MC) of CNTs (1.5 μm) and Te wires (about 5 μm) of fixed length are presented. As plotted in Fig 3(a) a positive MC is present, but depends on the bias regime, low or high. At the high bias regime, the MC is destroyed and this effect is not due to Joule heating, as seen in the Fig 3 (a) by comparing two temperatures. This observation is observed in all samples (including semiconductor nanowires), and has not been reported previously. In the low bias regime, the MC exhibits all characteristics of weak localization. The MC curves at zero bias are fitted (Fig 3 (b)) with the 1D weak localization formula \[ \Delta G_{WL} = -\frac{e^2}{\pi \hbar L} \left( l_\Phi^2 + W^2/3l_m^4 \right)^{-1/2} \] \[ \text{(1)} \]

where \( l_\Phi \) is the coherence length, \( l_m = \sqrt{\hbar/eB} \), \( L \) is the length and \( W \) is the radius of the wire. The fit is valid for all samples, except for the Te samples of diameter 200 nm (the large wires are no longer 1D with respect to the coherence length). The parameter \( l_\Phi \), ranged between 50 and 300 nm, is greater than the diameter of CNTs and wires, and follows the expected temperature dependence \( T^{-1/3} \) (inset of Fig 3 (b)). The decrease in the amplitude of MC with increases in the wire length and diameter has been observed. The presence of weak localization confirms the diffusive nature of the transport, and confirms the high degree of disorder. The diffusion coefficient obtained with \( \alpha \approx 100 \text{nm} \) is around \( D_\Phi \approx 100 \text{ cm}^2/\text{sec} \), confirming previous results about CNT. However, the Fig 3 (c) shows that, surprisingly, the weak localization is also strongly correlated to the coefficient α. In contrast to the universal law plotted in Fig 2 (b), the relation between the MC and the coefficient α depends on the nature of the contacts for CNTs. Two different curves are present for Ni and Co contacts to CNTs. A linear relation is observed for the Te of 40 nm diameter (the Au or Ni electrodes cannot be differentiated). Accordingly, α accounts also for the diffusion mechanisms, and these mechanisms depend on the nature of the interface. The coherence length is plotted as a function of α in the inset of Fig 3 (c).

We now discuss the data in terms of CB theory. In the CB regime, the coefficient α is defined by the action of the electromagnetic environment on the charge carriers, or in terms of transmission lines, by the impedance \( Z \) of the circuit to which the junction is contacted. The coefficient α depends on the diffusion constant of both the electromagnetic field and the charge carriers (some expressions are given in \[ \text{[2]} \text{[11]} \]). The scaling is obtained if the spectral density of electromagnetic modes \( I(\omega) \) is finite at low energy down to zero frequency modes \( \alpha = I(\omega \to 0) = Z(\omega \to 0)^{-1} \).

The conductance at zero temperatures \[ \text{[2]} \text{, Chap 2 formula (113) and (19), formula (19)} \], is given by Eq. \[ \text{2} \] for the prefactor \( G_V \) (below). It has also been predicted that the value at finite temperature and low bias coincides \[ \text{[8] chap 3, p25 (3.63)} \] with the expression of \( G_V \); the bias voltage energy and the thermal energy \( eV \leftrightarrow kT \).
can be permuted:

\[ G_T \approx G_V = \frac{1}{R} \frac{e^{-\gamma \alpha}}{\Gamma(2+\alpha)} \left( \frac{\pi \alpha}{e V_0} \right) \alpha \]

where \( \gamma = 0.577 \ldots \) is the Euler constant and \( \Gamma \) the Gamma function. The resistance of the tunnel barrier is \( R \), and the energy \( eV_0 \) is, in the case of ultra-small tunnel junctions, the Coulomb energy \( E_C = e^2/2C \), where \( C \) is the capacitance of the tunnel barrier. In a diffusion regime, the relevant energy is the Thouless energy \( eV_0 = E_T = kT_D/a^2 \) where \( D_T \) is the diffusion constant for the electromagnetic field in the electrodes or for the charges, and \( a \) the relevant length (the capacitance is now included in the coefficient \( \alpha \) [2][4].

As already mentioned, the power law is observed in Fig 1 (a) and (c). However, it is very surprising that Eq. 2 also fits the data plotted in Fig 2 (b) as a function of the coefficient \( \alpha \). The only fitting parameters are now the tunnel resistance \( R \) and the energy \( eV_0 \). This means that all samples have the same tunnel barrier (within the tolerance of one order of magnitude over nine). The fit with Eq. 2 of the data \( G_T \) plotted as a function of \( \alpha \) (in Fig 2 (b), after correction of the ratio \( 1/k^\alpha \)) gives a tunnel resistance on the order of \( R = 2.5 \Omega \), an energy of about 40 meV (which corresponds to a capacitance of about \( C = 2 \times 10^{-18} \) F). The fit of the data \( G_T \) is less convincing, but gives, however, the same tunnel resistance and an energy of about 100 meV. The relation \( G_V \approx G_T \) is confirmed within the approximation of a scaling function \( f \) composed of two power laws. For a typical length of a few nanometers, the diffusion constant coincides with the diffusion obtained from the weak localization \( D_T \approx D_0 \approx 100 \) cm²/sec. In contrast, the diffusion constant \( D_T \) deduced from the coherent length \( a = l_\Phi \) is about \( D_T = 1000 \) cm²/sec. This value is one order of magnitude larger than \( D_0 \) measured under magnetic field.

Why, despite the huge dispersion of intrinsic and extrinsic parameters (and especially the typical sizes and disorder of the electrode/wire junction), the parameters of the "tunneling junction" deduced from the scaling law are universal? In other terms, why is it not possible to differentiate between the samples from the point of view of the CB? These results suggest that disorder and quantum diffusion, together with relatively low dimensionality, impose a universal value to the relevant Thouless energy and resistance involved. The origin of this universality is not known.

In conclusion, a comparative study of electronic transport between multwall carbon nanotubes and Te nanowires has been performed. The samples are defined by a single scaling coefficient \( \alpha \) describing the ZBA. A universal relation is observed between \( \alpha \) and the conductance, valid whatever the nature of the electrodes, the lengths (\( \mu \)m range), and the diameters, ranged between 5 to 200 nm. All samples, except the 200 nm diameter Te, exhibit a typical 1D weak localization behavior from which the coefficient \( \alpha \) is also correlated. This study shows that the scaling law of the ZBA originates from a quantum diffusive process together with coulomb blockade with a universal tunnel barrier. An interpretation of the scaling law in terms of Luttinger liquid states can hardly be maintained.

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