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Unconventional Magnetotransport Phenomena in Individual Carbon Nanotubes

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Abstract. We investigate the quantum transport in different individual carbon nanotubes in the light of magneto-transport experiments in intense (60T pulsed) magnetic field. Large magnetic fields are required to probe field dependent gap modulation and quantum interference effects along the circumference of the tube. Such experiments along with a control of the electrostatic doping of the tube by a back-gate voltage constitute an unique tool to explore the exceptional electronic properties of this material. We bring evidence that the field dependence of the conductivity is a fingerprint of the electronic conduction modes and their interplay with the band structure (helicity), the static disorder and the location of the Fermi level of the tube. We infer the characteristic lengths of the electronic transport (the electronic mean free path and the phase coherence length) which are differently modified by the Fermi level location, depending on the disorder.

1. Introduction

Carbon nanotubes (CNTs) discovered in 1991 show remarkable unconventional mesoscopic behaviors. Among them, they can be either metallic or semiconducting depending on their

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diameter and chirality [1]. Intense debates are currently carry out to understand the relation between the electronic properties and the different conduction regimes. Up to now, transport measurements have shown that CNTs can act as a ballistic system with Fabry-Perot interferences [2], a quantum dot [3] or a diffusive conductor in the Weak Localization (WL) regime [4]. Deviations from the expected 1D conduction of a pristine tube are somehow connected to native defects which affect the electronic transport in a complex way. Several studies using different calculation technics reveal that introducing disorder (impurity, vacancy) yields to a very energy dependent electronic mean free path (l_e) [5] and resonant scattering on defect [6]. As a consequence, we can pass from a ballistic behavior to a diffusive regime with quantum interference effects, only by tuning the Fermi level (E_F) [7]. One way to explore these different regimes is to combine the tuning of E_F using a back-gate voltage with a high magnetic field. Applying a large magnetic field allows to probe quantum dephasing, either in WL regime in determining the phase coherence length (L_φ) or in Ballistic regime, where modulations of the energy gap [8] due to the Aharonov-Bohm effect (AB) are observed. Moreover, it remains an experimental challenge to observe AB effects in electronic transport properties. Gap modulation has been recently observed in optical experiments on solution of CNTs [9]. An other group observe them in a quantum dot regime, but in ballistic or diffusive mode, some confusions persist due to uncertainty on E_F location, therefore the tuning of E_F is necessary.

We present in this paper, several magnetotransport measurements in different CNTs. We bring evidence that magnetotransport along with an electrostatic controlled doping of the tube unveil the conductance regime and the associated transport characteristic lengths. Experiments in a isolated Double Wall Carbon Nanotube (DWCNT) in a semi-diffusive regime [10] demonstrate a complex interplay between WL and AB depending on E_F. Others in strongly diffusive Multi Wall Carbon Nanotubes (MWCNT) [11] reveal the strongly energy dependence of the characteristic lengths l_e and L_φ.

2. Experimental results

2.1. Experimental set-ups

CNTs are deposited on a Si/SiO_2 ($100nn$) substrate using molecular combing [12] or using a micropipette with a micrometer control. Electronic beam lithography is used to make evaporation metallic electrodes (Pd or Au) on the top of isolated CNTs. A back-gate on the *Si* allows to control electrostatic doping of the tube. Samples are plugged into a He cryostat where special care is taken to ensure high pulsed magnetic field [13].

2.2. Semi-diffusive Double Wall Carbon Nanotube

A DWCNT with 2.8nm diameter from CIRIMAT-LCMIET was connected with two gold electrodes spaced by 2.5μm. Magnetotransport combined with gate tuning of E_F for different temperatures were investigated up to 50T and down to 2K. Two probes resistance increases from 150$k\Omega$ at room temperature to 550$k\Omega$ at 2K. The differential conductance versus bias voltage exhibits a zero bias anomaly (ZBA) enhanced at low temperature.

We perform magnetoconductance (MC) measurements at 80K in parallel and perpendicular configuration (fig1, left panel) in high bias regime far away from the ZBA. In both

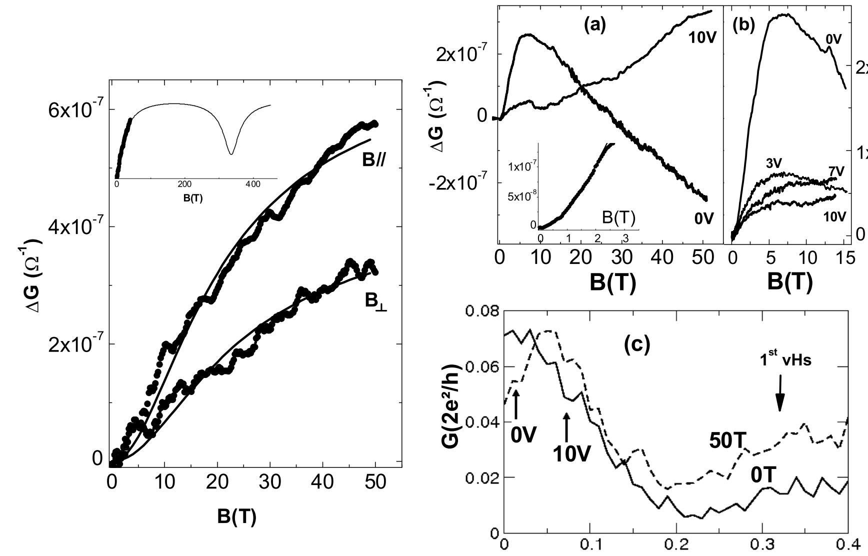

Figure 1. *Left panel:* Magneto-conductance $\Delta G(B)$ up to 50 T for $B_{\parallel}$ and $B_{\perp}$ with a bias voltage $V_b = 300$ mV and zero gate voltage, at 80 K. Solid lines represent the quasi-2D WL fits for both configurations. Inset: the predicted $\Delta G(B_{\parallel})$ from Eq. (1) in very high field, superimposed to our measurement. *Right panel:* (a) Magneto-conductance $\Delta G(B_{\parallel})$ up to 50 T with a bias voltage $V_b = 50$ mV and for $V_G = 0$ V and 10 V at 4 K. Inset: low magnetic field fit with the quasi 2D WL model (solid line), using Eq. (1) up to $B_{\parallel} = 3$ T. (b) the low field variations of $\Delta G(B_{\parallel})$ for different gate voltages from 0 to 10 V. (c) Conductance calculation versus Fermi level position, for a disordered (22,22) nanotube, and two values of a parallel magnetic field : $B_{\parallel} = 0$ T (solid line) and $B_{\parallel} = 50$ T (dashed line).

configurations, positive MC is observed up to 50T without significant gate voltage effect. Positive MC is interpreted in term of induced wave dephasing by a magnetic field. We used the expression of WL in a hollow cylinder of diameter $2R = 2.8nm$ and wall thickness a=0.14nm :

$$\Delta G_{WL}(B) = -AN\frac{e^2}{\pi\hbar}\frac{2\pi R}{L}[\ln(\frac{L_\varphi(B)}{L_\varphi}) + 2\sum_n (K_0(\frac{n2\pi R}{L_\varphi(B)}) \times \cos(2\pi n\frac{2\phi}{\phi_0}) - K_0(\frac{n2\pi R}{L_\varphi}))] \qquad (1)$$

where $K_0(x)$ is the Macdonald function and $\phi = \pi R^2 B$. $L_\varphi(B)$ is the phase coherence length defined by: $1/L_\varphi{}^2(B) = 1/L_\varphi{}^2 + (WeB/\hbar)^2/3$, where L_φ is the phase coherence length at zero field, $W = a$ (the wall thickness) in parallel configuration and N, the number of conduction channels, equals to 2. The A factor accounts for the contacts transparency in the two probes measurements. A very good agreement is obtained with A=0.7, which emphasizes the good quality of the contacts, and for $L_\varphi = 28nm$. We demonstrate that at 80K and up to 50T, MC is completely dominated by WL phenomena. It is the first time that WL is observed in DWCNT even if it remains a standard mesoscopic behavior.

Magnetotransport performed at 4K is rather different (fig1, top right panel). At this temperature, the gate has a preponderant effect on MC. For a zero-gate, the behavior is non monotonous with a change of the slope near 7T, whereas for 10V, the positive slope remains unchanged up to 50T. We can treat once again the low field regime with WL and for $V_g = 0V$, we get $L_\varphi = 84nm$ (inset fig1a right panel). The ratio $L_\varphi(80K)/L_\varphi(4K)$ agrees with a $T^{-1/3}$ dependence which is consistent with a dephasing by inelastic mechanism dominated by quasi electron-electron scattering [14].

Simulations based on Landauer-Buttiker formalism are performed to investigate the conductance at 4K [10]. The conductance in a (22,22) disorder tube in a coherent regime ($L_\varphi = 100nm$) is calculated versus energy (fig1, bottom left panel). In order to see WL phenomena, intensity of disorder is chosen to get l_e near the charge neutrality point (CNP) on the order of the circumference of the tube. The calculated conductance is found to be very energy dependent. We observe at 0T a decrease of the conductance once we move from the CNP to the first van-Hove singularity (vHs). Under 50T, a negative MC is observed near CNP likely due to the opening of a pseudo-energy gap (AB effect), whereas by shifting the Fermi level under 10V, a positive MC appears driven by the WL contribution. This competitive trend between DOS effect and WL effect explain quantitatively our result. At 0V, E_F is near the CNP as a consequence we observe for the first time AB effect in semi-diffusive regime. For a gate of 10V, we estimated a shift of 50meV (see [10]) as a consequence positive MC appears due to WL phenomena.

2.3. Strongly diffusive Multi-wall carbon nanotubes

A MWCNT of 10nm diameter (AFM estimation) from Nanocyl company is connected with two Pd electrodes spaced by 250nm. Magnetotransport up to 35T betwenn 300K and 4K and Raman spectroscopy are performed on the same isolated CNT. Between 300K and 70K, the resistance slightly increases from $150k\Omega$ to $180k\Omega$ without any gate effects. At 4K the conductance ranges from $500k\Omega$ to $1.5M\Omega$, depending on the gate voltage (between $\pm 6V$). This increasing does not scale with a thermo-activated behavior which rules out a semiconducting behavior or Coulomb blockade effects. Therefore, we probe a disorder metallic system which is confirmed by Raman spectroscopy (fig2). Raman spectroscopy shows two peaks, one from D-band due to presence of disorder and the other corresponding to the addition of two peaks related to tangential phonon mode, G^- and G^+. Their relatively close intensity means that the MWCNT is mainly metallic.

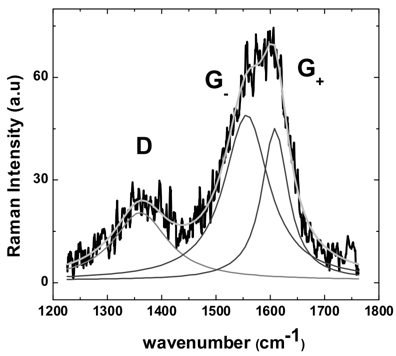

Figure 2. Raman spectroscopy performed on the tube. Three fits in grey are shown, the lorentzian D is a contribution of the double resonant process, G_- and G_+ are contribution of the tangential phonon modes. Light gray curve is the sum of the three lorentzian curves.

Differential conductance measurements versus gate voltage ($G(V_g)$) are performed at low temperature. We observe two kinds of modulations (fig3, left panel): fast and non periodic oscillations superimposed to a slow modulation with 2 minima around $\pm 2V$.

MC at 4K is measured in parallel configuration (fig3, right panel) for different gate voltages. We observe whatever the gate value, a positive MC whose the magnitude increases when the zero field conductance decreases. Both fast oscillations versus V_g and the positive MC at 4K can be interpreted in terms of quantum interferences inducing universal conductance fluctuations and WL phenomena. We use Eq(1) to fit experimental curves (fig3, right panel). We fix N equal to 2 (see below), we find A close to unity which underlines the good quality of the contacts. An original result is that L_φ is found to be significantly gate dependent: L_φ equals 6.2nm, 7.3nm and 10nm for V_g corresponding to 0, 1 and 2 V respectively. To complete our analysis, we determinate l_e. In the WL regime, the conductance is the

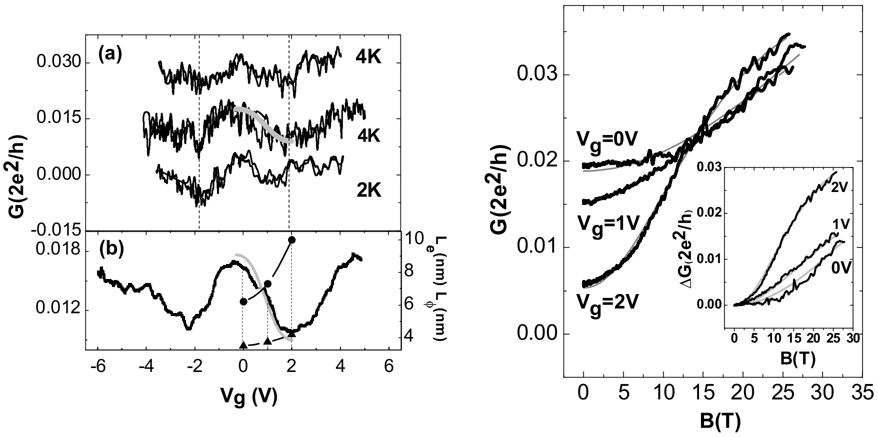

Figure 3. *Right panel*: Differential conductance dI/dV versus V_g at 4K and 2K after different thermal cycling. (a) Top curve shifted by $0.01G_0$, dI/dV at $4K$ and for $V_{bias} = 1mV$. Middle curve, dI/dV at $4K$ and for $V_{bias} = 0.1mV$. Bottom curve shifted by $-0.015G_0$, dI/dV at 2K and for $V_{bias} = 0.1mV$. Grey curve represents the fit of $G(V_g)$. (b) Smooth of the dI/dV measurements at 4K. Black circles and triangles are respectively L_ϕ and L_e versus V_g and represent gate voltage used for magnetoconductance measurement. Grey curve represents fit of $G(V_g)$ (see text). *Left panel*: Conductance versus $B_\parallel$ at 4K and for different gate voltages. Grey lines represent the theoretical curves of the conductance in the WL regime (see text). Inset, zoom on $\Delta G(B) = G(B) - G(0)$ for different V_g and the corresponding fits in grey lines using Eq.(1).

sum of G_{loc}, the WL contribution and $G_{cl} = \frac{2e^2}{h}\frac{Nl_e}{L}$, the classical part. From the experimental zero field conductances, we deduce l_e which is slightly gate voltage dependent: l_e equals 3.5nm, 3.73nm, 4.2nm for V_g equals to 0, 1 and 2V respectively. From the extracted values of l_e and L_φ and introducing them in Eq(1) we can check that the calculated $G(V_g)$ curves matches reasonably well with the experimental data (fig3, left panel).

In this study, we assume N=2. But, $G(V_g)$ presents slow modulation which could be caused by entering in vHs and yielding to a change of N. An estimation of our gate coupling [10, 11] gives : $\Delta E_F(meV) = 7.V^{-1}$. So to reach the first vHs at $\Delta E = 150meV$, we would have to apply $20V$. However, our back-gate is not sufficiently efficient to dope the tube up to the vHs. The slow modulations can not be due to the entering into them.

Theoretical studies [7, 6] show that in coherent regime with L_φ no energy dependent, defects are responsible for drop of the conductance when E_F is in the vicinity of the scatter energy. In our study, the slow conductance modulation are attributed to resonant scattering which modifies l_e and L_φ. A slight decrease of l_e at the resonance around 0V comes along with a large decrease of L_φ and an *increase* of the conductance. This can not be predicted by conductance calculations based on coherent transport. So, we give experimental evidence that in disordered regime, L_φ appears to be strongly energy dependent in the vicinity of resonant scattering at 0V, which is a new phenomena. As a consequence, the WL modulation dominates the classical part of the conductance. One remarks that when the WL contribution is destroyed under high magnetic field, the standard behavior of the conductance is recovered : the highest conductance curves comes with the largest l_e value.

3. Conclusion

We have probed under high magnetic field and with a gate tuning of E_F two kinds of CNTs. We can observe how disorder affects the conduction regime in a complex way. For a relatively low disorder system (DWCNT), we observe a strong interplay between density of states modulation (AB effect) and electronic diffusion effect (WL) on the conduction regime. The electronic transport moves from a quasi ballistic regime near the CNP, where AB effect is observed to a diffusive regime where WL dominates the MC. Whereas in a strong diffusive system (MWCNT from Nanocyl), only electronic diffusive effect are observed. WL contribution completely dominates the electronic transport. We do not observe AB effect due to the relatively weak value of L_{φ}, smaller than the circumference. Despite this fact, we observe indirect DOS effect by via resonant scattering which yields to a strong modulation of the characteristic transport lengths (l_e and L_{φ}). This study shows that it is important to control both, doping and disorder which, in turn, drive conduction regime.

4. Acknowledgments

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