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G. Fedorov, Benjamin Lassagne, Mathieu Sagnes, Bertrand Raquet, Jean-Marc Broto, et al.. Gate-dependent magnetoresistance phenomena in carbon nanotubes. *Physical Review Letters*, 2005, 9, pp.066801.1-066801.4. 10.1103/PhysRevLett.94.066801 . hal-03601122

HAL Id: hal-03601122

<https://hal.science/hal-03601122>

Submitted on 6 Nov 2023

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Gate-dependent magnetoresistance phenomena in carbon nanotubes

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We report on the first experimental study of the magnetoresistance of double-walled carbon nanotubes under magnetic field as large as 50 Tesla. By varying the field orientation with respect to the tube axis, or by gate-mediated shifting the Fermi level position, evidences for unconventional magnetoresistance are presented and interpreted by means of theoretical calculations.

Since their discovery, carbon nanotubes have been the focus of numerous studies and intense debate has been carried out concerning the nature of their electronic transport properties. The initial prediction of their electronic properties [1] and the possibility to obtain ideal 1D-ballistic conductors [2] have been confirmed experimentally by means of the scanning tunnelling microscopies[3] or transport measurements of nanotube-based devices [4].

More precisely, depending on their helical symmetries, the electronic properties manifest either a metallic or a semiconducting character. The origin of such behavior comes from the distribution of available states in the reciprocal space, as a consequence of quantization conditions (in relation with the circumference of the tube, and periodic boundary conditions), and in function of symmetry considerations. The application of a magnetic field is able to modulate the distribution of such states, resulting in strong modifications of the electronic spectrum [5]. Recently, such prediction of Aharonov-Bohm effects on the density of states was challenged experimentally on both large multiwalled nanotubes in the Coulomb blockade regime [7] and on SWNT by magneto-optical spectroscopy [6].

On the other side, while the first magnetotransport experiments performed on large diameter multiwalled nanotubes have clearly evidenced signatures of Weak Localization (WL) phenomena with negative magnetoresistance effects and $\phi_0/2$ -periodic Aharonov-Bohm oscillations [8], others works have proposed contradictory interpretations of magnetotransport data [9], as mainly stemming from the magnetic-dependence of the density of states (DOS) of a clean system in the ballistic regime [9]. To unravel the possible competing contributions enclosed within magnetofingerprints, it has been theoretically proposed to explore the gate-dependent variations

of the magnetoresistance patterns [10]. A gate-dependent magnetotransport study at low field has recently enabled to discuss the Luttinger liquid scenario in multiwalled nanotubes [11].

In this Letter, the magnetoresistance (MR) of double-walled carbon nanotubes with 3 nm diameter is investigated in large magnetic field up to 50 Tesla, and as a function of applied gate voltage, temperature, and magnetic field orientation with respect to the tube axis. Unconventional fingerprints are unveiled such as a switching mechanism from negative to positive magnetoresistance as a function of applied gate voltage. The analysis of the experimental data is consistent with the scenario of a strong contribution from magnetic-field modulated density of states in conjunction with weak localization effects. Numerical computation of the Landauer-Büttiker magnetoconductance also confirms such phenomenon, and a consistent theoretical explanation of experiments is proposed assuming an initial upshift of the Fermi level with respect to the charge neutrality point, attributed to unavoidable doping from the contacts or contaminants.

Our experiments are performed on small, 2.5 μm long, bundles of DWNTs deposited on the top of two gold nano-electrodes, and with typical diameter of 8 nm (the bundle contains thus less than nine nanotubes). Synthesis of DWNTs by chemical vapor deposition and the nano-manipulation for positioning the bundles have been presented elsewhere [12–14].

The individual DWNTs present a well defined diameter, centered around 2.5-3 nm. Given the expected proportion of metallic tubes (1/3), and owing to some device geometry considerations, electronic transport should be statistically dominated by a single metallic tube. This statement seems to be consistent with our (magneto)-transport data as detailed hereafter.

First, at zero magnetic field, the two probes resistance

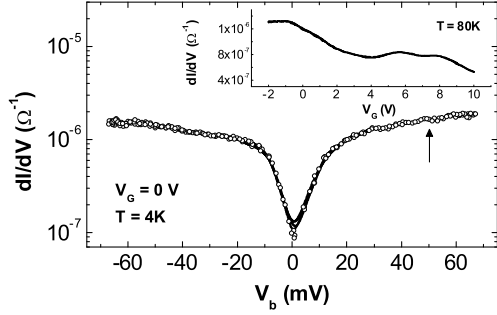


FIG. 1: Differential conductance versus applied bias voltage at 4 K for $V_G = 0$ V. The arrow indicates the high energy regime studied under 50 T at 4K. In inset, the gate voltage effects on the differential conductance for $V_b = 20$ mV at 80 K.

is typically in the order of 150 k Ω at room temperature, and it increases with lowering the temperature to reach 550 k Ω at 2 K, for a 50 mV applied bias voltage. The differential conductance $G = dI/dV$ exhibits a Zero Bias Anomaly (ZBA) which is markedly enhanced at low temperatures (Fig. 1). Whereas the appearance of ZBA and a power law behavior of $G(T, V_b)$ is usually interpreted in terms of Luttinger Liquid (LL) behavior for SWNT [16], the situation is more controversial for MWNT since LL-like ZBA can also be achieved from other considerations [11, 17]. In our experiment, gate effects on the differential conductance $G(V_G)$ exhibit relevant structures depending on the temperature and the bias voltage (inset Fig. 1). For small bias voltage, small modulations of $G(V_G)$ already appear at 80 K, where charging effects are excluded. This seemingly suggests some energy dependent structures of the DOS. Indeed, electronic resonant backscattering on quasi-bound states due to structural defects and/or impurities along the tube are known to modify the local DOS close to the charge neutrality point [18].

The impact of large magnetic fields on the transport properties is studied in the high kinetic energy regime: the voltage drop across the sample is fixed well above the ZBA as shown by the arrow on Fig. 1, for experiments at 4 K. In this regime, it is likely that the measured field dependence of the conductance is an intrinsic property of the tube. For small polarization, within the ZBA, the total resistance is widely dominated by the tunnel resistance at the contacts and measurements of the magneto-conductance (MC) give a negligible field dependence whatever the gate voltage.

Fig. 2 shows the conductance variation $\Delta G(B) = G(B) - G(0)$ at 80K for magnetic fields parallel (B_{\parallel}) and perpendicular (B_{\perp}) to the tube.

In both cases, the applied field increases the conductance of the NT and the saturation is almost reached below 50 T. The corresponding MC is respectively 10% and 6% at 50 T for B_{\parallel} and B_{\perp} , whereas minor gate voltage

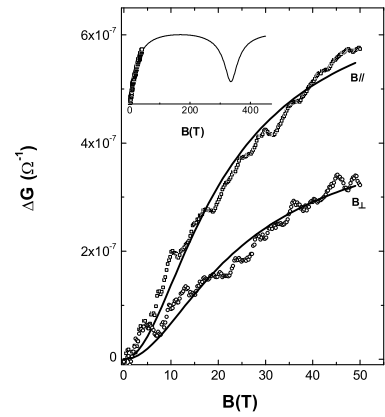


FIG. 2: Magneto-conductance $\Delta G(B)$ up to 50 T for B_{\parallel} and B_{\perp} with a bias voltage $V_b = 300$ mV and zero gate voltage, at 80 K. Solid lines represent the quasi-2D WL fits for both configurations. Inset: the predicted $\Delta G(B_{\parallel})$ from Eq. (1) in very high field, superimposed to our measurement.

dependences are observed on $\Delta G(B)$ at this temperature.

The origin of the positive MC is elaborated in terms of field induced dephasing effect of the electron waves interference in the WL regime. Indeed, let us consider a hollow cylinder of diameter $2R = 2.8$ nm with a wall thickness $a = 0.14$ nm. Based on Altshuler, Aronov and Spivak (AAS) calculations [21], the conductance variation $\Delta G_{WL}(B)$ as a function of a magnetic field applied along the tube axis can be expressed as follows:

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

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 and $W = a$ (the wall thickness) in parallel configuration. The A factor accounts for the contact transparency of the interface in the two probes measurements. Using Eq. (1) with two fitting parameters (L_{φ} and A), a convincing agreement (solid line, Fig. 2) to the experimental data for B_{\parallel} is obtained from zero to 50 T, with L_{φ} equal to 30 nm at 80K and $A \simeq 0.7$. Extrapolation to very high field shows that the WL contribution is already strongly reduced at 50 T, whereas AAS oscillations with a $\phi_0/2$ period corresponding to 336 T are expected (inset Fig. 2). Note that the A value corresponds to rather transparent contacts in the high bias voltage regime. The same value is used to fit all the data at other temperatures, for both B_{\parallel} and B_{\perp} .

Eq. (1) is easily rewritten in the B_{\perp} case considering the surface of the tube perpendicular to the field. The field dependence of the phase coherence length varies

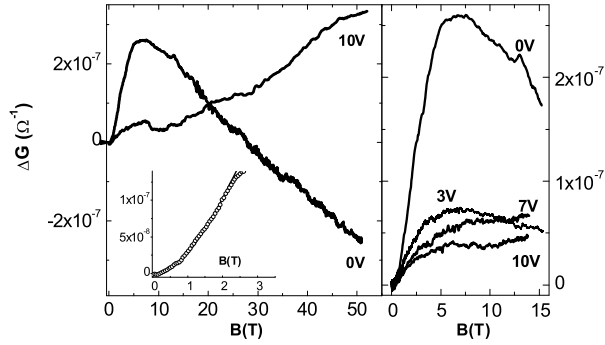


FIG. 3: Left panel: Magneto-conductance $\Delta G(B_{\parallel})$ up to 50 T with a bias voltage $V_b = 50$ mV and for $V_G = 0$ V and 10 V at 4 K. Inset: low magnetic field fit with the quasi 2D WL model (solid line), using Eq. (1) up to $B_{\parallel} = 3$ T. Right panel: the low field variations of $\Delta G(B_{\parallel})$ for different gate voltages from 0 to 10 V.

within the diameter of the tube instead of the wall thickness for the parallel case. For a width $W \approx 2R$ and $\phi = 0$, we straightforwardly derive a new expression for $\Delta G_{WL}(B)$, which yields to a very good agreement with our data in the perpendicular configuration with $L_{\varphi} \approx 28$ nm (solid line, Fig. 2). The consistency between B_{\parallel} and B_{\perp} up to 50 T clearly demonstrates that quantum interferences in the WL regime dominate the DWNT conductivity at 80K with a phase coherence length a few times larger than the circumference.

The situation at 4.2 K gains in complexity. From hereon, we focus on the B_{\parallel} case. At zero gate voltage, a positive MC is observed up to $B \approx 7$ T, above which a large negative MC dominates with no hint of saturation (Fig. 3 - left panel). Applying a 10 V gate voltage astonishingly suppresses the high negative MC, and $\Delta G(B)$ remains positive with no sign of saturation up to 50 T. The electrostatic doping under 50 T tunes the MC from negative (-10%) to positive (+35%). Changes of MC for different V_G are detailed in Fig. 3 - right panel. An increase of the gate voltage gradually decreases the low field positive MC. Simultaneously, the high field MC changes from a negative slope to a positive one.

These unconventional magneto-fingerprints cannot be strictly interpreted from quantum interferences effects. The non-monotonous MC in B_{\parallel} , at 4 K and zero gate voltage, clearly results from the superposition of a very sensitive positive MC in low field and a large negative MC which dominates the high field regime. A likely origin of the low field positive MC is, once again, WL around the circumference of the tube. Fitting the low field MC with Eq. (1) and $L_{\varphi}(4K)$ as a unique parameter gives a reasonable agreement with a phase coherence length equal to 84 nm (solid line, inset Fig. 3 - left panel). The ratio $L_{\varphi}(78K)/L_{\varphi}(4K)$ agrees remarkably well with a $T^{-1/3}$ dependence, a fact consistent with a dephasing by quasi-elastic electron-electron scattering [21].

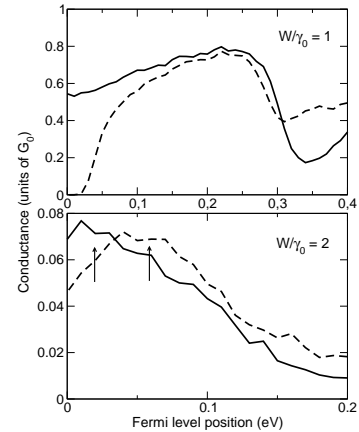


FIG. 4: Conductance versus Fermi level position, for a disordered (22,22) nanotube, and two values of the parallel magnetic field: $B_{\parallel} = 0$ T (solid line) and $B_{\parallel} = 50$ T (dashed line). Two disorder strengths are considered: $W/\gamma_0 = 1$ (upper panel) and $W/\gamma_0 = 2$ (lower panel). In the strong disorder case, the two arrows indicate possible Fermi level positions for $V_G = 0$ V (left) and $V_G = 10$ V (right).

To investigate the origin of the drastic change of MC in high parallel field versus V_G , a numerical study of the Landauer-Büttiker conductance of a nanotube-based heterojunction is performed, using a standard procedure [23]. The whole system is made from two semi-infinite disorder-free (22,22) metallic tubes with diameter 3 nm, that describe the external leads, whereas the central part consists of the same metallic nanotube with length 100 nm, and in which an elastic disorder (Anderson-type) together with a magnetic field are introduced. The common tight-binding Hamiltonian is used with constant integral overlap γ_0 between first neighbors, whereas site energies ε_n are randomly distributed within an interval $[-W/2, W/2]$ (uniform density) to simulate an elastic disorder. Under the application of a magnetic field, standard procedures are used to implement the Peierls phase factors contribution to the quantum phases [5, 10]. In this description, a useful analytical estimation of the elastic mean free path [2] can be derived, enabling to discriminate between ballistic, diffusive and localized regimes. Two values of disorder are chosen to make the mean free path in the vicinity of the charge neutrality point, either much larger ($W = 1$, in units of γ_0) or in the order of the nanotube circumference ($W = 2$), for which the conventional weak localization framework strictly applies [8]. The main results are reported on Fig. 4 where the conductance is plotted for two values of disorder, at zero magnetic field and for $B_{\parallel} = 50$ T, to cope with the experimental range.

The quantum conductance at zero field is first found to be strongly energy dependent, in agreement with prior results [24]. One notes in particular that it decreases in the vicinity of the van Hove singularities due to a de-

crease of the relaxation time. More interesting is the role of the magnetic field that induces two competing trends. First, the WL contribution yields positive MC. For moderate disorder ($W = 1$), it only dominates within the semi-conducting subbands, where the mean free path becomes of the order of the tube circumference. For sufficiently strong disorder ($W = 2$), the WL contribution is important and yields positive MC for a large part of the spectrum. Second, the magnetic field also affects the electronic structure, effect that is more predominant in the vicinity of the charge neutrality point, given the opening of the energy gap, followed by its enlargement, and resulting in a negative contribution to the MC. One may thus interpret experimental results reported on Fig. 3 at zero gate voltage as a signature of such competing contributions of quantum interferences and density of states effects which are expected to dominate in high field. A consistent interpretation of the experimental results is possible in the strong disorder case $W = 2$. An initial position of the Fermi level is proposed in Fig. 4 (left arrow). For this energy, positive MC due to WL dominates at low field (numerical result not shown here) and negative MC due the energy gap dominates at high field. The application of a positive gate voltage, by up-shifting the initial Fermi level, consistently reduces the contribution from DOS effects, and yields a positive MC as reported in Fig. 3. From the differential conductance measurements versus V_G within the ZBA [20], we estimate the back-gate capacitance C_g in the order of a few pF per meter. The expected Fermi level up-shift, given by $\Delta E_f = eV_g C_g / C_{NT}$, (C_{NT} , the electrochemical capacitance) under 10 V is thus around 50 meV. Its position at $V_G = 10$ V is represented by the right arrow in Fig. 4. The reduction of the zero-field conductance at higher gate voltage is also consistently found in theoretical and experimental results (Fig. 1). Interestingly, if one sketches the eventual contribution of an additional semi-conducting tube parallel to the metallic one, one notices that its bandgap reduction due to the magnetic field will be much too small to affect the MC. This states that a single metallic NT is likely to account for the measured gate dependent MC. The assumption of a diffusive regime allows us to rescale the computed zero-field conductance obtained for $W = 2$ and for tube length 100 nm. The numerical estimate for the same device but with length of $2.5 \mu\text{m}$ gives a conductance value comparable but slightly below the experimental measure. This may originate from the fact that the numerical calculations are performed in the fully coherent regime for a tube length of 100 nm, which is larger than the coherence length estimated experimentally (≤ 80 nm). Finally, our gate dependent MC measurements reveal that character-

istic lengths in diffusive DWNT make possible a non-trivial coexistence between quantum interference along the circumference and electronic band structure modulation. Results are significantly different in case of NT in the Coulomb blockade regime [7], for which gate and magnetic field effects give a spectroscopy of the electronic energy levels.

The authors thank Th. Ondarçuhu for experimental assistance on DWNT deposition. Substrates preparation have been performed in the LAAS technological platform. This work has been supported by the french ministry of research under program ACI "Nanosciences 2004" number NR044 "NOCIEL". F.T and S.R acknowledge support from the Micro and Nanotechnology program from French ministry of research under the grant RTB: Post CMOS moleculaire 200mm and the research grant from French ministry of research under ACI project TransNanoFils.

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