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Probing the electronic properties of individual carbon nanotube in 35 T pulsed magnetic field

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Abstract

After optimization of the alignment and the nano-contact processes of isolated single wall and double-walls carbon nanotube, we investigate the high magnetic field effects on the electronic transport properties of an individual metallic CNT. We develop pioneer multi-probes magneto-transport experiments under a 35 T pulsed field which reveal an unexpected oscillatory behavior of $R(H)$ inconsistent with existing theories.

1. Introduction

The combination of an exceptional tubular structure and the semi-metallic behavior of the graphene sheet gives to the carbon nanotube (CNT) fascinating electronic properties [1]. Recent transport experiments on individual metallic CNT reveal the richness of the interplay between the one-dimensional structure, the electronic correlations and the invasive nature of the contacts [2–8]. It is well known that applying a magnetic field on an electronic system is an unique efficient tool to probe the band structure near the Fermi level and

the quantum behaviors of the conductivity. In case of CNT, new phenomena are predicted under a magnetic field. We shall cite, in a the parallel configuration, giant magneto-resistance oscillations due to field-induced periodic band gap modulation and Aharonov–Bohm type oscillations of the magneto-conductance [9–11]. In a the transverse geometry, field-induced electronic backscattering [10,12], Landau level formation [9] and complex changes of the density of state at E_F due to field-induced shift of the van Hove singularities are expected to induce complex magneto-resistance spectra [13–16]. Experimental evidence of these effects require magnetic fields whose characteristic length ($l_B = \sqrt{\hbar c/eB}$) is comparable to the CNT diameter. This is a challenging area as the high field behavior of the conductivity is a

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signature of the intimate nature of the electronic transport through the contacted CNT, i.e., ballistic [17], quasi-ballistic [18] or diffusive [19]. Up to now, first magneto-transport results on individual CNT focus on large multi-wall CNT with diameters ranging from 19 to 40 nm and under a maximum static field of 12 T [3,19–23]. In a parallel configuration, oscillatory behaviors of the conductance have been observed; although they demonstrate the field effects on the electronic wave function along the CNT circumference, both periods and magnitudes of these oscillations disagree to a large extent to current predictions [3,20,21]. Attempts to explain these discrepancies have involved the multi-wall contributions to the electronic transport [20] or the possible existence of chiral currents along the tube [3]. Clarification requires new experimental results which combine much higher fields to unambiguously define the oscillatory periods and measurements on smaller CNTs. The experimental physics under high magnetic field of a single-wall CNT is still to be explored.

In the following, we first present a new approach allowing the alignment and the electrical nano-connection of isolated CNTs. Next, we address the feasibility of pulsed field magneto-transport experiments on an individual single and bi-walls small diameter CNT up to 35 T and more. We present magneto-transport experiments on an individual metallic 2 nm CNT under a 35 T pulsed field in the longitudinal configuration. Quantum oscillations of the magneto-resistance are observed at low temperature and their origin is discussed in the frame of existing theories.

2. Experimental

For this study, we work on CNTs synthesized by chemical vapor deposition of H_2/CH_4 at 1000 °C using a solid Co–MgO powder. The control of the process allows the synthesis of CNTs being that mainly single and double-walled with a length within the μm range and a diameter centered around 2 nm [24]. After extraction of the CNTs from the catalyst by a mild acidic treatment, a stable aqueous suspension of CNTs is prepared by

addition of a surfactant, sodium dodecyl sulfate (SDS).

The nanocontacting process we present is based on a novel combination of proven techniques: it consists in the deposition of aligned CNTs over a Si/SiO₂ substrate and the fabrication of nanoelectrodes by high resolution electron beam lithography (HREBL) that selectively contact a single CNT to predefined microelectrodes. A precise control of the alignment of the CNTs on the substrate is important because it facilitates both the design of the nanoelectrodes and the orientation of the sample in the magnetic field.

To control the direction of the CNT on the substrate, we use the so-called ‘molecular combing’ method initially developed by one of us [25]. The affinity of the CNTs with the surface of the substrate drives the CNTs density and their degree of alignment with respect to extraction direction. A silanization of the SiO₂ surface with perfluorodecyltrichlorosilane is chosen to optimize the deposition. It gives a perfect alignment of the nanotube and a density of 0.03 CNT/ μm^2 very well adapted to our geometry (only a few CNT in the active area of our device). The molecular combing is achieved by dipping the substrate into the CNTs suspension for 15 min. The sample is then pulled out of the suspension through the liquid meniscus at low speed (200 $\mu m/s$) and rinsed a few seconds in milli-Q water to remove the SDS layer using the same velocity.

The multi-probes nano-connection of an isolated CNT using the HREBL is helped by the control of its orientation on the substrate. First, CNTs are pinpointed and selected by AFM observations. Their coordinates are then introduced in the HREBL software for alignment of 4 (at least) nanoelectrodes contacting the CNT perpendicularly to its axis. HREBL is performed at an acceleration voltage of 200 keV and the dose for exposure of the nanoelectrodes is typically 2200 $\mu C/cm$. This dose is higher than the one required on a bare Si wafer due to the thick SiO₂ insulating layer (620 nm), however it remains below the damaging threshold for CNTs [26]. The nanoelectrodes are fabricated by lift-off of a Ti_{20 nm}/Au_{5 nm} metallic layer deposited in a high vacuum thermal deposition chamber. A typical

result of an individual CNT nanoconnection is presented in Fig. 1. In this case, 6 nanoelectrodes of 200 nm width connect a 7 μm CNT whose diameter is estimated to 2 nm.

The magnetic fields necessary to probe the electronic transport in small diameter CNT are far above the accessible limit in static field. We propose experiments in a 40 T pulsed magnetic field (soon to be increased up to 60 T) obtained by a capacitor bank discharge in a resistive coil. Some uncertainties reside in how a CNT may react to a noisy electromagnetic environment due to the high capacitor charge voltage, the intrinsic inductive current due coming from to a large dH/dt and the Lorenz forces applied on the CNT during the pulse. Inductive currents are strongly reduced by minimizing the effective area perpendicular to the applied field. However, selfic components in the measuring circuit are expected to induce voltages proportional to the second derivative of the field which is quite large at the beginning. A straightforward calculation of the CNT inductance L gives $L \approx \mu_0 \pi r^2 l^2 / a(l^2 + 2r^2)$, where l and r are, respectively, the CNT length and radius and a is the carbon-carbon distance. For the CNTs we work

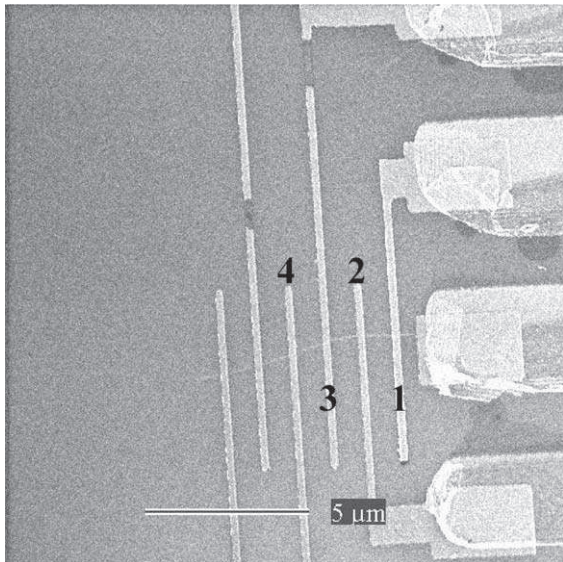


Fig. 1. Scanning electron microscopy observation of a 2 nm diameter CNT after nano-connection. The nanoelectrodes ($\text{Ti}_{20 \text{ nm}}/\text{Au}_{5 \text{ nm}}$) are 200 nm wide and 1 μm spaced.

on, we infer a very small inductance, $L \approx 10^{-13}$ H. Therefore, the resulting voltage peak at the beginning of the field due to the tubular structure of the CNT should be negligible. On the other hand, we estimate the Laplace force applied on the CNT during the measurement: for a 40 T pulsed field and a sensing current of few tens of nA, the magnetic force is in the range of the pN. The van der Waals adhesive forces of a the CNT lying on the silane layer are thought to be several decades higher [27]. So the strength of the mechanical contact of the CNT on its substrate plus its intrinsic rigidity should prevent from any field-induced vibrations. With a suitable electromagnetic shielding of the sample, magneto-transport experiments in long durative pulsed magnets are therefore conceivable.

3. Results and discussion

Electronic transport measurements are performed on the 2 nm diameter CNT shown on Fig. 1, in AC current using the 2 and 4 probes configuration, between contacts 1 and 4. At room temperature, the contact resistance is around 15 k Ω and it increases by one order of magnitude at 4 K. The 4 probes CNT resistance conductance is $(25 \text{ k}\Omega)^{-1}$ at 300 K and its temperature dependence clearly exhibits a power law dependence ($G \propto T^\alpha$) from 300 K down to 20 K (Fig. 2). The α exponent equals 0.34; this strongly suggests a metallic CNT with a conductivity dominated by the electronic tunneling into a Luttinger liquid. At lower temperatures, a departure towards a thermally activated regime is observed with an effective energy gap of 0.6 meV (inset, Fig. 2). Note that the same temperature laws behavior over the whole temperature range and roughly identical energy gaps (between 0.4 and 0.6 meV) are measured for various locations along the CNT. The physical origin of the activation energy may be due to structural defects like a twisted tubule which is theoretical expected to localize quantum particles [28], as well as the intrinsic curvature of the CNT [29]. We may also involve electrostatic charge effects in the CNT. For a 7 μm long CNT with tunnel contacts, we expect a Coulomb gap of 0.65 meV [4], which is consistent with the experimental value.

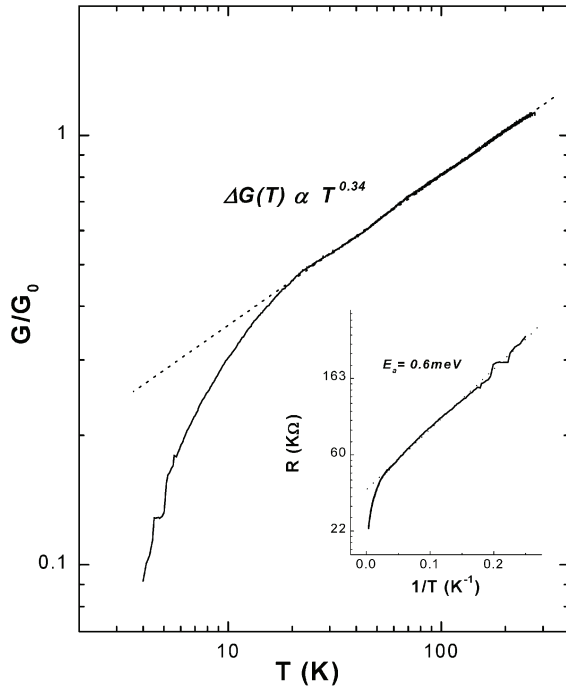


Fig. 2. Conductance versus temperature of the 2 nm diameter CNT measured in AC current in the 4 probes configuration with the current applied between contact (1) and (4), and the voltage measurement between (2) and (3) and with $f_0 = 30$ Hz and $I = 0.5$ nA. From 300 K down to 20 K, $G(T)$ follows a power law T^α . The inset illustrates a thermally activated regime below 20 K.

Fig. 3 represents the 2 probes magneto-resistance (MR) measurement on the same 2 nm diameter CNT at 2 K, in the longitudinal configuration and for several 35 T magnetic shots with different applied currents. An oscillatory behavior is clearly visible with a first period of 18 T and an amplitude of the order of 15–20 k Ω ($\Delta R/R \approx 2\%$). We do not observe any significant bias voltage dependence on the MR curves and similar results are obtained for various locations of the CNT. The same magnitude of $\Delta R(H)$ are measured for the 2 and 4 probes configuration, although the oscillatory behavior at 4 probes is barely visible for low bias voltages. These remarks insure that the quantum oscillations are reproducible and not related to any localized defects in the CNT or at the interface nano-contact/CNT.

The theoretical oscillations period for an Aharonov–Bohm effect along the 2 nm diameter CNT

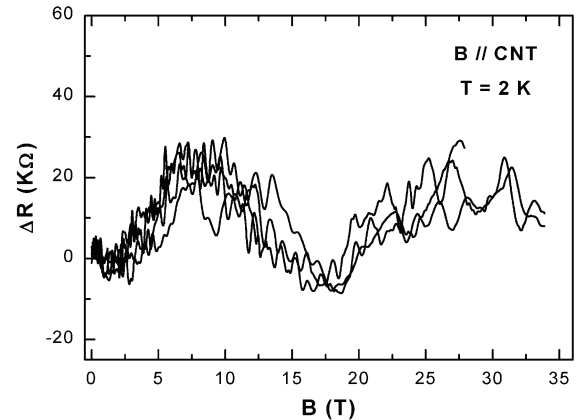


Fig. 3. Two probes longitudinal magneto-resistance measured on a 2 nm CNT at 2 K measured between contacts (1) and (2). The different curves represent successive pulsed field measurements with different applied current ranging from 0.5 to 6 nA and giving evidence for an oscillatory behavior of $R(H)$.

is h/e [30]. If we envisage a diffusive transport with phase coherent scattering along the circumference, Al'tshuler Aronov Spivak (AAS) oscillations are also expected with a period of $h/2e$ [31]. In any case, this gives an inaccessible field periods of 1317 and 658 T, respectively. Therefore, a drastic discrepancy exists between our faster oscillations and theoretical predictions inherent to the tubular structure. Smaller field periods have already been observed in large MWNT [3,20]. Multi-wall contributions with phase shifts related to different chiralities have been invoked to explain oscillations with a period close to $h/3e$ [20]. In our case, as we deal with single or double-walled CNT, this scenario is ruled out. An other possible explanation is the existence of chiral diffusive electron trajectories surrounding the tube due to mechanical distortions of the CNT [32]. Matching the predicted period with our experimental value would require almost an hundred of electron turns before interfering, which is quite questionable.

If one assumes the CNT as an ideal conductor, an applied magnetic field along the tubular axis is expected to modulate the energy gap with an h/e period [9]. Both the period and the magnitude of the oscillations shown in Fig. 3 are inconsistent with a gap modulation. The field-induced opening of the gap should be of the order of hundreds of

meV considering the CNT diameter [30]; the expected $\Delta R(H)$ variations should be much larger than those we experimentally observe. The existence of a finer structure in the energy dispersion remains an open question.

The different theories mentioned above in the diffusive and the ballistic regimes have been recently merged in an exact numerical computation of the Kubo conductivity [16]. Roche et al. [13,14,16] have demonstrated that the magneto-conductance results from the superposition of the field effects on both the density of state $\rho(E_F)$ and on the diffusion coefficient, D . Changes in the chemical potential and the disorder are thought to strongly influence $\rho(E_F)$ and D , respectively, giving rise to complex MR spectra. In any case, oscillatory behaviors in parallel field are expected to scale with the quantum of flux which is in strong disagreement with our results.

It is interesting to note that contrary to [3], we do not observe a negative MR in low field, interpreted in terms of a weak localization regime. We recall that our $G(T)$ measurement strongly suggests a Luttinger liquid behavior of the CNT. We therefore arise the question of quantum interferences in a Luttinger liquid. Electron–electron correlations and AB effects have been theoretically dealt in mesoscopic rings by Jagla and Balseiro [33]. They demonstrated changes in the fundamental AB periodicity of the transmittance originating from the charge and spin separation. Due to different velocities, v_s and v_c , spin and charge excitations make a different number of loops in the ring before arriving at the same time to the opposite contact. This induces a much smaller fundamental AB period depending on the v_s/v_c ratio. Applying this concept to CNT is a lead to understand the origin of the fast periodic oscillations. This appeals for new theoretical calculations of this effect in the frame of the a one-dimensional tubular structure.

4. Conclusion

The alignment procedure and the nano-contacting steps we develop allow for the selection of an isolated CNT and the control of its orientation

for magneto-transport experiments. We demonstrate that conductivity measurements in high pulsed magnetic field on an individual small diameter CNT are feasible. We unambiguously observe unexpected quantum oscillations of the magneto-resistance with respect to the existing theories. This issue brings up fundamental questions of the real nature of the conductivity in a connected CNT. It should stimulate new calculations and further high field experimental studies.

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