Bolometric detection of mechanical bending waves in suspended carbon nanotubes

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Abstract

We show that it is possible to detect mechanical bending modes on 1\,\mu m long ropes of single walled-carbon nanotubes suspended between 2 metallic contacts. This is done by measuring either their dc resistance in a region of strong temperature dependence (in the vicinity of superconducting or metal-insulator transition), or their critical current. The vibrations are excited by a radio-frequency electric field produced by an antenna located in the vicinity of the sample. We analyze the mechanism of detection of the mechanical resonances in terms of heating and phase breaking effects.
Single walled Carbon nanotubes (SWNT) are molecular wires which combine both remarkable electronic and mechanical properties. Depending of its diameter and helicity a SWNT can be either semiconducting or metallic with only two conducting modes at the Fermi energy \([1,2]\). The Young modulus of a rope constituted of small number of aligned SWNT has been estimated from the study of vibrations by electron microscopy \([3,4]\) and more recently by force microscopy experiments \([5]\) to be as high as 1TPa. This result is in agreement with theoretical calculations \([6]\). In the following we show that it is possible to excite and detect stationary mechanical bending modes of a SWNT rope which is suspended between metallic contacts across a slit. The excitation is produced by applying a radio-frequency electromagnetic field to the tube. The energy dissipated in the sample at resonance is detected by measuring the dc resistance of the tube in a range of strong temperature dependence and the critical current for the superconducting ropes.

We use as starting material carbon nanotubes prepared by the electrical arc method. When cobalt is used as a catalyst \([7]\), this method produces single wall tubes whose diameters are all of the order of 1nm. In most cases these tubes are assembled into ropes containing typically 100 nearly ordered parallel tubes.

Isolation of an individual rope and its connection to electrical contacts were performed according to the following procedure: a focused laser beam releases a nanotube from a target onto the sample with a \(Si_3N_4\) membrane coated with a metal film. A submicron width slit about 100\(\mu m\) in length has previously been cut in the membrane using a focused ion beam and has been disposed under the tube on the target using an optical microscope. After deposition, the nanotube connects the edges of the slit and shorts the electric circuit whose resistance was over 1\(G\Omega\) before the nanotube was deposited. This procedure has been shown to produce low resistivity ohmic contacts between the tube and the metal of the electrodes which is melt during the process \([8,9]\). A transmission electron microscopy image of such a sample is shown in fig.1A. Depending on the value of the resistance of the rope at room temperature, different temperature dependences are observed at low temperature, going from a thermally activated behavior at low temperature for resistive samples to a quasi
metallic behavior where the resistance is temperature independent. When low resistive ropes are connected to superconducting contacts they become superconducting due to proximity effect with zero resistance below the transition temperature of the contacts (see fig.1B).

In the following we will restrict our discussion on the two samples $RO_1$ and $RO_2$ mounted on Au/Re contacts superconducting at 1.2K. $R_{01}$ is 1.7µm long and contains approximately 200 SWNT, the room temperature value of its resistance is equal to 65Ω which is only twice the expected value assuming that each tube is a two-channel ballistic wire. $R_{01}$ becomes superconducting courtesy of the proximity effect, below the transition temperature of Au/Re \cite{9}. On the other hand $R_{02}$ (0.6µm long and constituted of 100 SWNT), is much more resistive (10kΩ at room temperature) and exhibits an exponential divergence of its resistance at low temperature (see fig. 1B). The dc transport properties of these ropes are strongly affected by the presence of a radio-frequency electromagnetic field produced by an antenna located in the vicinity of the sample.

The dc voltage drop across the superconducting rope $R_{01}$ is depicted on fig.2 as a function of the rf electric field frequency which amplitude is estimated to be of the order of $E_{rf} = 30mV/cm$. The sample is fed by a dc current of 2µA (below the critical value $i_c = 2.7µA$ \cite{9}). One can clearly identify a succession of resonance peaks which are harmonics of $f_1 = 330MHz$. When the rf power is not too high these can be described by a lorentzian line-shape with a quality factor of the order of 1000 at 100mK and decreasing with temperature like $1/T$ in the superconducting regime (see inset of fig. 2). These resonances are not observed in the normal state where the resistance is temperature independent. Similar type of resonances are also observed on semi-conducting tubes. An example is shown on fig.3B with the semiconducting sample $R_{02}$: the dc resistance function of the RF frequency presents a negative peak at 860MHz. In both cases, we interpret these resonances as stationary mechanical bending modes of the rope. The fundamental mode of a vibrating rod attached on both ends is indeed given by \cite{10}:

$$f_1 = 22.4 \frac{R}{2L^2} \sqrt{\frac{Y}{\rho}}$$  \hspace{1cm} (1)
where $Y$ is the Young modulus estimated to $10^{12} Pa$, $\rho = 1.3 kg/m^3$ is the mass per unit volume, $L$ and $R$ the length and radius of the rod ($L = 1.7 \mu m$, $R = 7 nm$ for $RO_1$ and $L = 0.6 \mu m$, $R = 2.5 nm$ for $RO_2$). This yields to $f_1 = 280 MHz$ for $RO_1$ and $f_1 = 1.2 GHz$ for $R0_2$. Note that for $R0_1$ the resonance frequencies experimentally observed are of the order of $nf_1$ with $n > 1$. The fundamental frequency is not observed, which may be due to very small coupling between the antenna and the tube in this frequency range. On the other hand only the fundamental mode could be observed for $R0_2$. The mechanical nature of the resonances has been confirmed for $R0_1$ by injecting a small amount of nitrogen gas in the vacuum can of the dilution fridge. Adsorption of nitrogen atoms on the tube resulted in a small drift of the resonances down to lower frequency, which is indeed expected when the mass of the tube is slightly increased. A similar frequency shift of the resonance could also be detected on $R0_2$ at $T=4.2K$ when the sample is immersed from helium gas into liquid helium (see fig.3). This measured relative shift ($\delta f/f = -0.012$) is smaller but of the order of the calculated value -0.03 taking an increase of the effective density of the SWNT equal to the density of liquid helium. It is possible that the SWNT already initially contains a certain amount of adsorbed helium atoms.

An important issue for the understanding of these observations is the mechanism of conversion of the electromagnetic field into a transverse ultrasonic wave through the tube. We could not detect any observable change in the resonance spectrum when applying a dc voltage on the antenna superimposed on the ac excitation. So, we suggest that the presence of electrostatic charges on the tube and the Coulomb force produced by the radio-frequency electric field on these charges are the main mechanism of excitation of mechanical vibrations. The existence of charge depletion in a carbon nanotube in close contact with a noble metal such as Au or Pt has been shown to arise from the difference of electronic work functions between the SWNT and the metal by Dekker et al. From this work, the resulting uncompensated charges on a SWNT can be estimated to be of the order of $q = 100e$ where $e$ is the electron charge. It is possible that these charges are not uniformly spread on the tube which could favor the excitation of harmonics compared to the fundamental.
A similar procedure for the excitation of mechanical resonances has been investigated in cantilevered multiwalled nanotubes under an electron microscope \cite{13}.

As we discussed below the resonances can be detected by dc transport only in the regions where the resistance of the tubes depends strongly on temperature. This suggests that the mechanism underlying the detection of the signal is the heating of the electrons, which is maximal at resonance. Nevertheless in that case the line-shape of the resonances should depend strongly on the power of the rf field, since as discussed below their quality factor strongly depends on temperature. This is not what was measured: on the contrary, the quality factor remains constant as the power is varied, until a critical value above which saturation effects appear, as shown on fig. 4. This implies that the suspended tubes are probably poorly thermally connected to the contacts which are not heated by the rf power. Then the central part of the sample is heated up to a temperature $T_{eff}$ whereas the ends remain at $T_0$. Let us try now to estimate $T_{eff}$. The force on the rope can be estimated from the knowledge of the rf electric field $E_{rf}$ applied on the tubes: $F = (1/4\pi\varepsilon_0)NqE_{rf}$. This force yields to a vibration of amplitude $\delta x$ given by $\delta x = QFL^3/\lambda R^4Y$ where $Q$ is the quality factor of the resonance and $\lambda$ a numerical coefficient of the order of 300. The power $P_{diss}$ dissipated at a resonance of frequency $f$ reads $P_{diss} = F\delta x2\pi f/Q$. This power is transferred to the electrons via electron-phonon collisions in the tube which results in increasing the temperature in the center of the sample. The thermal impedance $Z_{th}$ between the tube and the superconducting contacts is dominated by the value of the superconducting part of the rope at $T_0$ and be estimated as follows \cite{14}:

$$Z_{th} = \exp(\Delta/T_0)(T_0/\Delta)^2/Nk_1(T_0) \sim 410^{17} WmK^{-1}$$ (2)

where $N$ is the number of SWNT in the rope, $k_1(T) = k_B^2T/h$ is the thermal conductance of a ballistic 1D wire, $\Delta$ is the superconducting gap for the bilayer $Au/Re$ estimated to be of the order of $2K$ and $T_0 = 100mK$ the temperature of the contacts. From that value the effective temperature of the tube is obtained from the relation: $T_{eff} - T_0 = Z_{th}P_{diss}$ which gives $T_{eff} \sim 0.5K$, to be compared with the range of temperature where the resistance
of $RO_1$ increases from 0 to 50$\Omega$, i.e. $0.8 - 2.3K$. We have neglected the contribution of the phonons to the thermal conductivity. This is justified by the observation of a strong decreasing of $T_{\text{eff}}$ for a given rf power when applying a magnetic field of the order of the critical field of the contacts. A similar calculation can be done on the rope $R0_2$, but for this semiconducting sample one has to take into account the reduced value of $k_1$ in the normal state according to the Wiedemann-Franz law. This yields to an effective temperature of $T_{\text{eff}} = 5K$ (for $T_0 = 4.2K$ and $E_{\text{rf}} = 1V/cm$).

Note however that all this analysis relies on the assumption that it is possible to define a temperature of the electrons. This is only true when the inelastic electron-electron scattering length is short compared to the total length of the tube. It is also possible that the vibrations of the tube are at the origin of dephasing processes, like electron-phonon scattering. This results in a reduction of the phase coherence length of the electrons below the length of the tube. The existence of such a phase breaking mechanism more efficient at low rf power than the simple heating of the tube for the destruction of proximity induced superconductivity is corroborated by the study of the critical current through $R0_1$ versus excitation power. As a matter of fact we observed an exponential decay of the critical current, whereas its temperature dependence follows a BCS type of behavior [9], very flat below $T_c/2$ (see fig. 4A). The critical current through an SNS junction is indeed known to vary like $I_c \propto \exp(-L/L_\phi)$ where $L_\phi$ denotes the phase coherence length, related to the phase coherence time $\tau_\phi$ by $L_\phi = v_F \tau_\phi$ for a ballistic junction. Thus our result can be understood as a contribution of the rf phonons to the inverse coherence time as $\tau_\phi^{-1} \propto P_{\text{rf}}$. Note that the dephasing is not directly related to the rf electric field but to the induced vibrations at resonance. The same rf power applied out of resonance has indeed no effect on the critical current.

The mechanical resonances on $RO_1$ are detected by measuring the critical current for very low excitation power. For higher power the critical current vanishes whereas a finite resistance shows up. As shown in fig. 4B, there is a narrow range of rf power for which the resistance exhibits a lorentzian shape as a function of frequency. At even higher power the
shape is no longer lorentzian but presents a plateau in its center. This can be understood considering that the ends of the rope are no longer at $T_0$. When they become normal their thermal conductivity increases drastically, resulting in a much better thermalization of the center of the sample, whose temperature does not increase anymore and even can decrease, as observed on fig. 4B.

We have shown that suspended nanotube molecules work as a passive radio set tuned to definite resonant frequencies which correspond to the mechanical transverse eigen-modes of the tube. The detection of these modes is done courtesy of the very low thermal conductivity of the tubes which behave as extremely sensitive bolometers when their resistance is temperature dependent. Moreover we have shown for the superconducting sample that a more effective mechanism is involved, implying phase breaking effects. The next step would be to investigate the influence of these low energy phonons on transport properties by systematic comparison of the proximity effect on deposited and suspended tubes. It has indeed been shown [15] that coupling with low energy phonons can turn repulsive interactions in a Luttinger liquid into attractive ones and drive the system towards a superconducting phase.
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FIGURES

FIG. 1. A. Transmission electronic micrograph of a rope of single wall nanotubes, suspended across a slit between 2 metallic pads. From these pictures it is possible to confirm that the metal of the contacts neither covers nor penetrates the tubes and to estimate the length and diameter of the samples. B. Temperature dependence of the resistance of different ropes mounted on Au/Re contacts measured for zero magnetic field. The arrow indicates the transition of the Au/Re bilayer. The sample $R_{01}$ becomes superconducting below 1K.

FIG. 2. A. Effect of a radio-frequency electromagnetic radiation on the dc voltage across the rope $R_{01}$ when it is run through by a dc current below the critical current. Inset: Evolution of the resonance line shapes of the 6th harmonics on $R_{01}$ with the temperature of the contacts.

FIG. 3. A. Effect of the deposition of solid nitrogen onto the rope $R_{01}$ detected on the dc resistance versus frequency of the electromagnetic radiation. B. Effect of the immersion in liquid helium of the rope $RO_2$ detected on the dc resistance versus frequency of the electromagnetic radiation.

FIG. 4. A. Critical current measured on the rope $RO_1$, on one hand as a function of the rf power at resonance of the 6th harmonics (lower curve), and on the other hand as a function of the temperature (upper curve). B. Resistance of the rope $RO_1$ versus rf frequency near the 6th harmonics, for different applied powers.
A

\[ R (\Omega) \]

\[ T = 115 \text{mK} \]

after \( N_2 \) deposition

B

\[ \delta R/R \]

\[ T = 4.2K \]

\[ P_{\text{He}} = 10^{-4} \text{bar} \]

liquid He →
A

I_c (T) without RF

I_c (P_rf) at T=110mK

B

R (Ω)

P=-14dB

P=-15dB

T=110mK

P=-16dB

T (K)

RF power (a.u.)

0.0 0.1 0.2 0.3 0.4 0.5 0.6

0.0 0.2 0.4 0.6 0.8 1.0

0.0 0.2 0.4 0.6 0.8 1.0

0.0 0.2 0.4 0.6 0.8 1.0

0.0 0.2 0.4 0.6 0.8 1.0

0.0 0.2 0.4 0.6 0.8 1.0

frequency (MHz)