Acoustoelectric Effects in Carbon Nanotubes

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We report observations of acoustoelectric effects in carbon nanotubes. We excite sound in μm long ropes of single walled carbon nanotubes suspended between two metallic contacts by applying radio-frequency electric field. The sound is detected by measuring either the dc resistance of the tubes in a region of strong temperature dependence (in the vicinity of superconducting or metal-insulator transition), or their critical current. We show that, depending on the excitation power, the vibrations produce either electron heating or phase coherence breaking.

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Single walled carbon nanotubes (SWNT) are molecular wires with both remarkable electronic and mechanical properties. Depending on 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 made of a 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 10^{12} Pa. This result is in agreement with theoretical calculations [6]. However, acoustoelectric effects which connect the electric and mechanical properties of these systems have not been investigated so far. In the following we show that it is possible to excite and detect stationary sound waves in a SWNT rope which forms a suspended bridge between two metallic contacts. The excitation is produced by applying a radio-frequency (rf) electromagnetic field to the tube. We show that at high rf power the mechanical energy dissipated at resonance causes electron heating which can be detected, in certain temperature dependence conditions, via a change of the dc resistance of the tube. At lower rf power the vibrations induce a loss of phase coherence which is detected via a reduction of the critical current for a sample made superconducting by the proximity effect.

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 1 nm. 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 procedure described previously [8,9]. A transmission electron microscopy image of such a sample suspended across a slit 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 for resistive samples to a quasimetallic behavior where the resistance is temperature independent (Fig. 1A). When low resistance ropes are connected to superconducting contacts they become superconducting due to the proximity effect, with zero resistance below the transition temperature of the contacts [9].

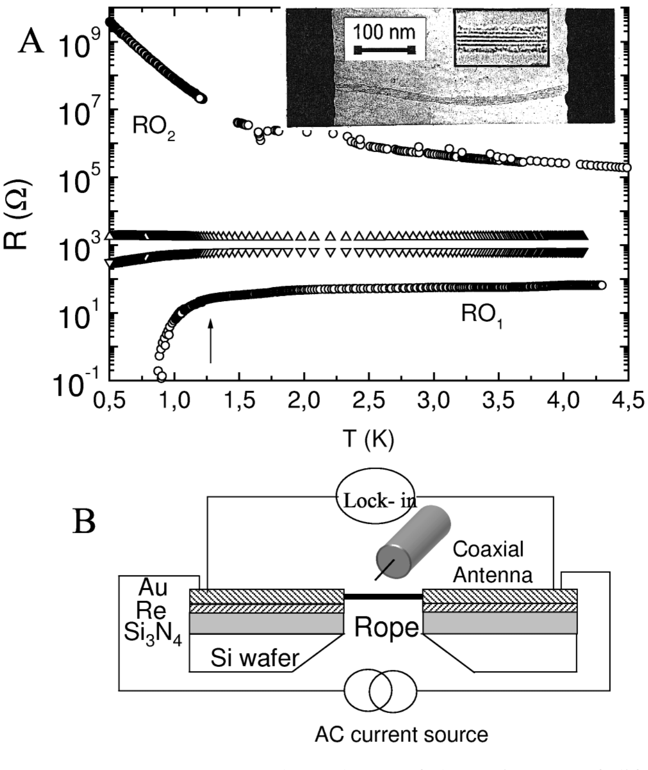

FIG. 1. (A) Temperature dependence of the resistance of different ropes mounted on Au/Re contacts. The arrow indicates the transition of the Au/Re bilayer. The sample RO_1 becomes superconducting below 1 K. Inset: Transmission electronic micrograph of a rope of SWNT, suspended across a slit between two metallic pads. From this picture it is possible to confirm that the metal of the contacts neither covers nor penetrates the tube and to estimate the length and diameter of the samples. (B) Schematic drawing of the experimental setup.

In the following we restrict our discussion to two samples, RO_1 and RO_2, mounted on superconducting ($T_c = 1.2$ K) Au/Re contacts. RO_1 is 1.7 μm long and contains approximately 200 SWNT [10]. Its room temperature resistance (65 Ω) is only twice the value expected assuming that each tube is a two-channel ballistic wire [1]. RO_1 becomes superconducting below the transition temperature of Au/Re [9]. On the other hand RO_2 (constituted of 100 0.6 μm long SWNT), is much more resistive (10 kΩ at room temperature) and exhibits an exponential divergence of its resistance at low temperature.

The dc transport properties of these ropes are strongly affected by the presence of a rf electromagnetic field produced by an antenna located in the vicinity of the sample (Fig. 1B). The dc voltage drop across the superconducting rope RO_1 is shown in Fig. 2A as a function of the frequency of the rf electric field, which amplitude is estimated to be of the order of $E_{\rm rf} = 1$ V/cm for an injected power of $+13$ dB. The dc current (2 μA) flowing through the samples is smaller than the critical current $i_c = 2.7$ μA [9]. One can clearly identify a succession of resonance peaks which are nearly harmonics of $f_1 = 330$ MHz. When the rf power is not too high, the width of the peaks is well defined and grows linearly with temperature in the superconducting regime (Fig. 2B). At 100 mK the quality factor of all the peaks is of the order of 1000. At higher power we observe a striking change of the resonance line shapes (Fig. 2C). The resonances are not detected in the normal state where the resistance is temperature independent. We observe similar resonances in semi-conducting tubes as shown on Fig. 3B where the dc resistance of RO_2 exhibits a negative peak at 860 MHz. In both cases, we interpret these resonances as stationary mechanical bending modes of the rope. Indeed, the frequency of the fundamental transverse [11] vibration mode of an unstretched rod clamped on both ends is given by [12]

$$f_1 = \frac{22.4}{2\pi} \frac{R}{2L^2} \sqrt{\frac{Y}{\rho}}, \qquad (1)$$

where Y is the Young modulus estimated to be 10^{12} Pa [5,6], $\rho = 1.3\ 10^3$ kg/m^3 is the mass of a rope per unit volume [6], L and R are, respectively, the length and radius of the rod ($L = 1.7$ μm, $R = 12$ nm for RO_1 and $L = 0.6$ μm, $R = 5$ nm for RO_2). This yields $f_1 = 240$ MHz for RO_1 and $f_1 = 1.2$ GHz for RO_2. The higher modes of the unstretched rod are not harmonics and occur at $2.7f_1$, $5.4f_1$, $8.9f_1$, etc. On the other hand, if the rod is stretched all modes are harmonics of a frequency higher than f_1.

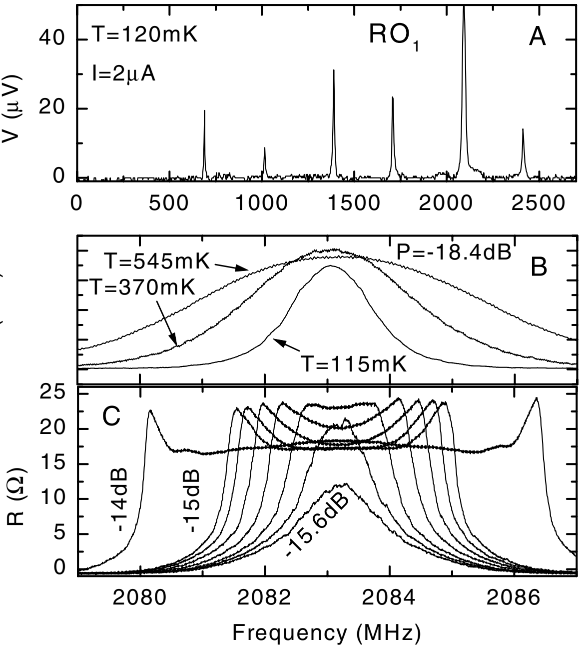

FIG. 2. (A) Effect of a rf electromagnetic radiation on the dc voltage across the rope RO_1 when it is run through by a dc current below the critical current. (B) Evolution of the resonance line shapes of the sixth harmonics on RO_1 with the temperature of the contacts. (C) Resistance of RO_1 versus frequency near the sixth harmonics, at 110 mK and for different applied rf powers.

For RO_1 the experimentally observed resonance frequencies are approximately multiples of 300 MHz as expected for a slightly stretched rope. The fundamental frequency is, however, not observed. This absence may have intrinsic origin (like the nonhomogeneity of the strain along this long rope, leading to weak coupling of the fundamental mode to a homogeneous excitation), or simply due to the vanishing coupling to the antenna in this frequency range. In contrast only one mode is observed for the much shorter RO_2 rope, since higher modes were outside of our experimental detection range. The mechanical origin of the resonances was confirmed for RO_1 by injecting a small amount of nitrogen gas in the vacuum can of the dilution refrigerator. Adsorption of nitrogen atoms on the tube resulted in a small drift of the resonances to lower frequency, as expected when the mass of the tube is slightly increased (Fig. 3A). Adsorption also resulted in an increased sensitivity of the sample resistance to rf excitation at resonance. A similar frequency shift of the resonance was also detected on RO_2 at $T = 4.2$ K when the sample was immersed from helium gas into liquid helium (Fig. 3B). The measured relative shift ($\delta f/f = -0.012$) is smaller but of the order of the value -0.03 expected, assuming an increase of the effective density of the SWNT equal to the density of liquid helium. The SWNT might already contain some adsorbed helium atoms.

We now turn to the discussion of the electroacoustical mechanisms responsible for the excitation and detection of these resonant sound waves. The first important issue is the conversion of an electromagnetic field into a transverse ultrasonic wave through the tube. Since we could not detect any change in the resonance spectrum when a dc voltage was added to the antenna's rf excitation, we suggest that the presence of electrostatic charges on the tube and the Coulomb force produced by the rf electric field on these charges is the main mechanism of excitation of vibrations [13]. The existence of charge depletion in a

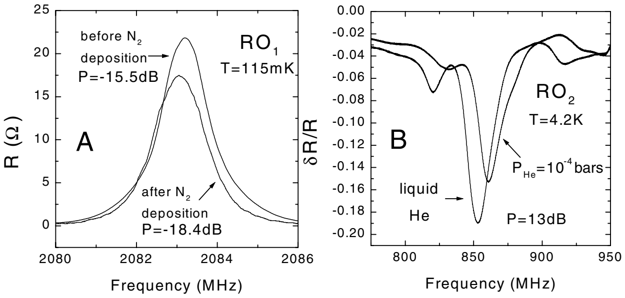

FIG. 3. (A) Effect of deposition of solid nitrogen onto the rope RO_1 detected on its dc resistance versus frequency of the electromagnetic radiation. (B) Effect of the immersion of the rope RO_2 in liquid helium, detected on its dc resistance versus frequency of the electromagnetic radiation.

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 [14]. From that 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. If these charges are not uniformly distributed on the tube, the excitation of harmonics is favored compared to the fundamental. A similar mechanism for the excitation of mechanical resonances has been demonstrated in cantilevered multiwalled nanotubes under an electron microscope [15].

In the following we discuss the two detection mechanisms depending on the level of excitation: electron heating and phase coherence breaking. As mentioned above, 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 allowing the detection of the signal is the heating of the electrons, which is maximal at resonance. This implies that the suspended tubes must be 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 to estimate T_{eff}. The force on the rope can be deduced from the knowledge of the rf electric field E_{rf} applied on the tubes: $F = NqE_{\text{rf}}$, where N is the number of SWNT in the rope. This force produces a vibration of amplitude δx given by $\delta x = QFL^3/\lambda R^4 Y$, where Q is the quality factor of the resonance and $\lambda = \pi(11.2)^2$ according to expression (1). The power dissipated at a resonance of frequency f reads $P_{\text{diss}} = F\delta x 2\pi f/Q$. This power transferred to the electrons results in an increase of the temperature in the center of the sample. The thermal impedance between the tube and the superconducting contacts is dominated by the superconducting part of the rope at T_0 and can be estimated to be [16]

$$Z_{\text{th}} = \frac{1}{Nk_1}\left(\frac{k_B T_0}{\Delta}\right)^2 \exp\left(\frac{\Delta}{k_B T_0}\right) \sim 4.10^{17}\ \text{K W}^{-1}, \tag{2}$$

where $k_1 = k_B^2 T_0/h$ is the thermal conductance of a ballistic 1D wire, $\Delta \sim 2$ K is the superconducting gap for the bilayer Au/Re, and $T_0 = 100$ mK is the temperature of the contacts. The effective temperature of the tube is then obtained using $T_{\text{eff}} = T_0 + Z_{\text{th}} P_{\text{diss}} \sim 0.5$ K for $E_{\text{rf}} = 30$ mV/cm, to be compared with the range of temperature where the resistance of RO_1 increases from 0 to 50 Ω, i.e., 0.8–2.3 K. In this estimate the contribution of phonons to $k_1(T)$ was neglected. A similar calculation can be done on the rope RO_2, but for this semiconducting sample, we estimate k_1 from the resistance of the sample and the Wiedemann-Franz law. This yields an effective temperature $T_{\text{eff}} = 5$ K (for $T_0 = 4.2$ K and $E_{\text{rf}} = 1$ V/cm). These estimations show that suspended nanotubes behave as extremely sensitive bolometers when their resistance is temperature dependent. Note that this interpretation in terms of a temperature profile distributed along the tube somehow assumes that the thermalization time of the electrons is very short compared to the electron transit time through the tube. This hypothesis is valid only in a situation of strong electron-electron inelastic scattering which is probably achieved at high rf excitation power.

There is only a narrow range of rf power for which the resistance of RO_1 exhibits a simple resonance shape as a function of frequency. At higher power the peak height saturates. 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 can even decrease (Fig. 2C).

We now discuss the behavior of the superconducting rope RO_1 for low rf power such that it remains in a phase coherent state with zero linear resistance. In this regime the critical current (deduced from nonlinear transport [9]) decays exponentially with the rf power at resonance, whereas it exhibits a BCS-like temperature dependence, very flat below $T_c/2$ (Fig. 4A). This suggests that when the sample is superconducting a much more efficient mechanism than simple heating is responsible for the breaking of Cooper pairs. The difference between the $V(I)$ curves at low temperature in the presence of rf and at higher temperature without rf (but for the same critical current) corroborates this point (Fig. 4B). The critical current through a superconductor-normal metal-superconductor junction is indeed known to vary like $I_c \propto \exp(-L/L_\phi)$, where L_ϕ denotes the phase coherence length, related to the phase coherence time τ_ϕ by $L_\phi = v_F \tau_\phi$ for a ballistic junction. Thus the exponential behavior indicates that the excited sound contributes to the inverse coherence time as $\tau_\phi^{-1} \propto P_{\text{rf}}$. Since the same rf power applied out of resonance has no effect on the critical current we conclude that the dephasing is not directly related to the rf electric field but rather to the induced vibrations at resonance.

What is the nature of the acoustoelectric coupling responsible for electron heating and phase coherence breaking? It is unlikely that the very low energy phonons created by these vibrations can directly exchange energy

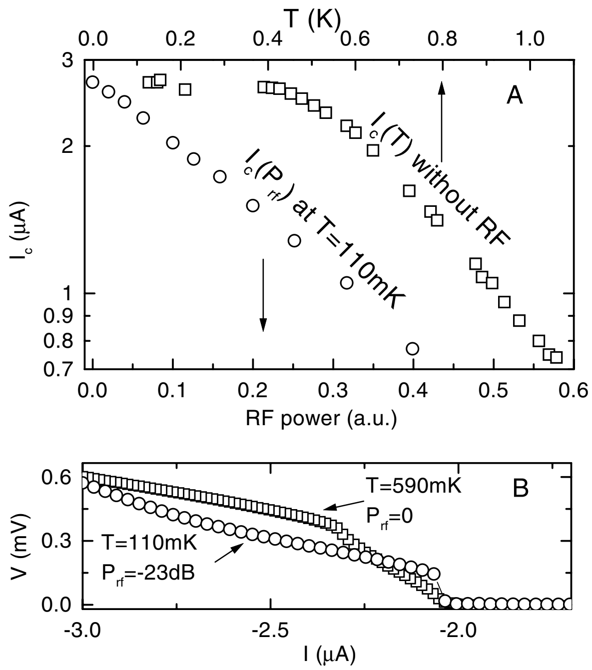

FIG. 4. (A) Critical current of the rope RO_1, as a function of the rf power at resonance of the sixth harmonics at 110 mK, and as a function of temperature without rf field. (B) Voltage across RO_1 as a function of the dc current.

with electrons. Here we argue that the sound attenuation in the nanotubes below 1 K is due to two level systems (TLS) as has been seen is most solids (both metallic and insulator). The linear increase of the resonance line width at very low temperature in RO_1 (Fig. 2B) is similar to the temperature dependence of sound attenuation observed down to $T \sim \hbar\omega/k_B$ in many systems [17]. It has also been shown that the interaction between TLS and electrons is at the origin of the telegraphic and $1/f$ noise on the resistance generally observed in mesoscopic conductors [18] and also observed in carbon nanotubes [19]. More recently, the possibility of low temperature dephasing due to TLS has also been emphasized [20]. Thus the presence of TLS in carbon nanotubes may provide an indirect electron-sound coupling mechanism which is far more efficient in terms of energy and phase relaxation than the direct electron-phonon coupling. A likely origin of these TLS in carbon nanotubes is adsorbed gas molecules on the surface of the tubes. This hypothesis is corroborated by the observed increase of the resonance intensity with the injection N_2 molecules: before injection the resonance starts to saturate at −15.5 dB (Fig. 2C) while after injection the saturation threshold is lowered to −18.4 dB (Fig. 3A).

This work opens a new field of investigation of acoustical properties in carbon nanotubes and their interplay with coherent electronic transport. 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 [21] 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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