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Electromechanical resonance behavior of suspended single-walled carbon nanotubes under high bias voltages

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Abstract
We characterize the nanoelectromechanical response of suspended individual carbon nanotubes under high voltage biases. An abrupt upshift in the mechanical resonance frequency of approximately 3 MHz is observed at high bias. While several possible mechanisms are discussed, this upshift is attributed to the onset of optical phonon emission, which results in a sudden contraction of the nanotube due to its negative thermal expansion coefficient. This, in turn, causes an increase in the tension in the suspended nanotube, which upshifts its mechanical resonance frequency. This upshift is consistent with Raman spectral measurements, which show a sudden downshift of the optical phonon modes at high bias voltages. Using a simple model for oscillations on a string, we estimate the effective change in the length of the nanotube to be $\Delta L/L \approx -2 \times 10^{-5}$ at a bias voltage of 1 V.

(Some figures in this article are in colour only in the electronic version)

Nanoelectromechanical systems (NEMS) have been realized for many applications, including ultralow mass sensors [1–3], strain detection [4, 5] and pressure sensors [4–6], and mechanical oscillators [2, 7–9]. Carbon nanotubes' (CNT) exceptionally low mass, high tensile strength and stiffness, and possible defect-free nature place them amongst the best possible systems for this application. Room temperature electromechanical resonators have been built using doubly clamped suspended single-walled carbon nanotubes (SWNT), excited through electrical actuation by a gate electrode [10]. Similar systems have been used for atomic scale mass sensing at cryogenic temperatures [3]. However, the high temperature electromechanical response of SWNT NEMS resonators has not been studied. Given the negative thermal expansion coefficient (TEC) [11–13] that graphitic systems have, these suspended devices are expected to behave differently in this regime. In this work, we report anomalous behavior in these resonator systems under high electrical bias. We show that there is a critical voltage, above which optical phonon emission causes the nanotube temperature to increase suddenly, thus causing the nanotube to contract.

Figure 1(*a*) shows a scanning electron microscope (SEM) image of one of the devices used in this study. In the fabrication process, nanotubes are grown by chemical vapor deposition (CVD) over 500–700 nm deep trenches etched in silicon substrates between predefined catalyst islands and metal (Pt/W) electrodes [14–16]. No post-processing was performed after the nanotube growth, minimizing defects and surface contaminants on the nanotubes. The resulting suspended nanotubes are 1–2 μm in length, as defined by the trench width, and between 1 and 3 nm in diameter.

Our electrical and optical measurements are carried out in an optical vacuum chamber. Mechanical vibrations are

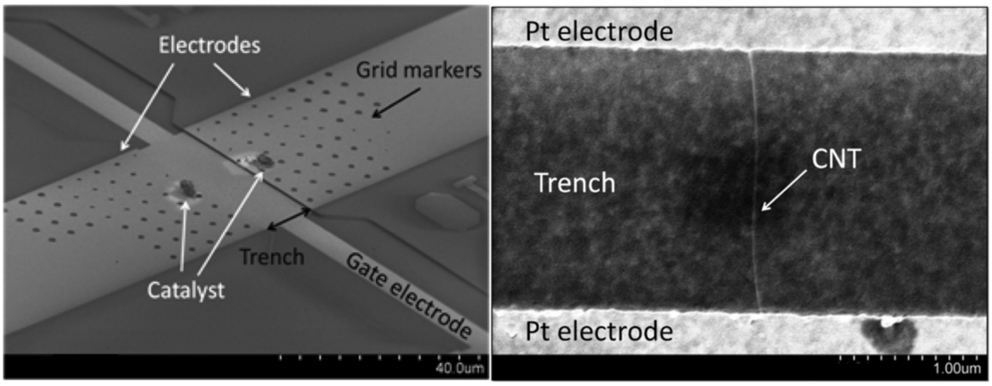

Figure 1. SEM images of a suspended CNT sample device. The right panel shows a high resolution SEM image of an individual carbon nanotube.

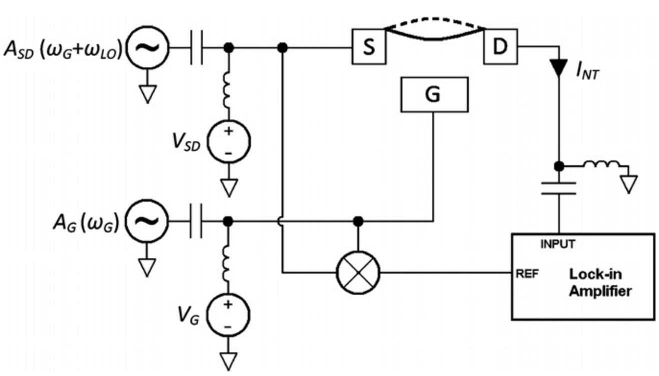

Figure 2. Circuit diagram of the electromechanical resonator setup.

detected by applying two ac voltages to the drain and gate voltages, as described by Sazonova *et al* [10]. An additional dc bias voltage is applied across the nanotube device using a 'Bias-Tee', as shown in figure 2. This provides a dc offset, $v_{SD}(t) = A_{SD}\cos(\omega_G + \omega_{LO})t + V_{SD}$. Another 'Bias-Tee' is connected to the input of the lock-in amplifier to ensure that the dc current does not saturate the input by creating a path to the ground terminal. By separating the dc path and ac path on the source and detection side of the device, the noise was significantly reduced. An ac gate voltage, $v_G(t) = A_G\cos\omega_G t + V_G$, is applied to the gate electrode on the bottom of the trench. Nominal values for the applied voltages are $A_{SD} = 30$ mV, $A_G = 40$ mV, $f_{LO} = \omega_{LO}/2\pi = 20$ kHz. The current at the input of the lock-in amplifier (I_{NT}) is the sum of all frequency components from mixing in the nanotube device ($\omega_G + \omega_{LO}$, $2\omega_G + \omega_{LO}$, ω_G, ω_{LO}). The high frequency components are filtered by the lock-in amplifier's low pass filter. By tuning ω_G through a mechanical resonance, we observed a change in current at the local oscillator frequency (ω_{LO}). The mechanical resonance frequency of the nanotube (f_R) is distinguished from other resonances due to the passive elements in the circuit by varying V_G, and observing whether or not a shift occurs in the mechanical resonance frequency.

Figure 3(*a*) shows the RF current through a suspended CNT at $f = f_{LO}$ as the gate frequency (ω_G) is swept through the mechanical resonance frequency. The different datasets in figure 3(*a*) correspond to different applied gate voltages. The shifting peak (or dip) appearing between 34 and 38 MHz corresponds to a mechanical resonance, while the stationary peaks correspond to other resonances due to the passive elements in the circuit. The shift in the resonance peak is caused by increased tension in the nanotube due to the electrostatic force between the nanotube and the dc gate electrode. Figure 3(*b*) shows the electric current plotted

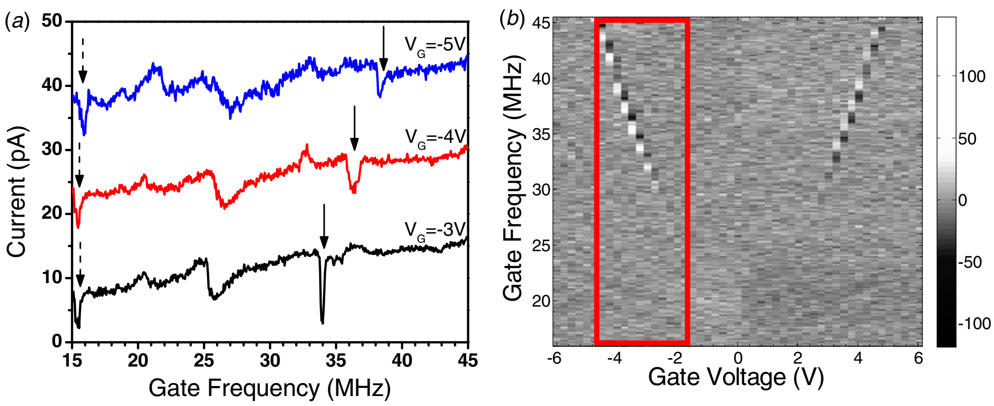

Figure 3. (*a*) Electric current plotted as a function of gate frequency. The solid arrows show the resonance due to the nanotube and dashed arrows point to resonances due to passive circuitry elements. (*b*) Electric current plotted as a function of the gate frequency (f_G) and dc gate voltage. The red box corresponds to the experimental regime shown in figure 4(*b*).

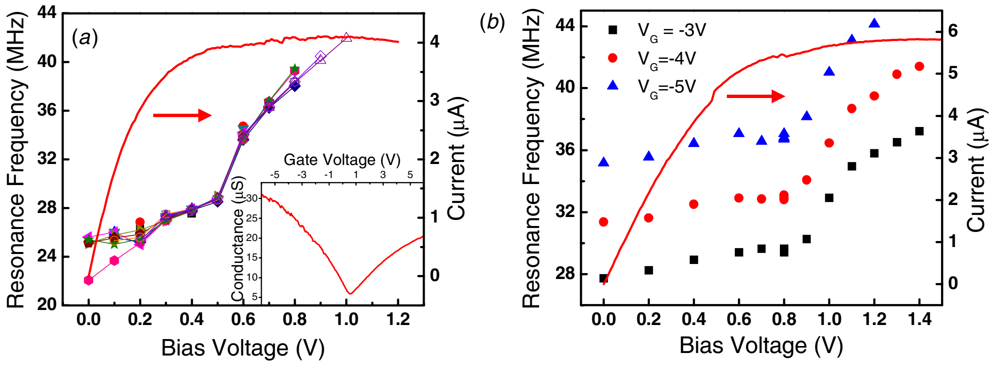

Figure 4. Bias voltage dependence of the mechanical resonance frequency of two different suspended nanotube devices. (*a*) The different datasets were taken on different days at a constant gate voltage of $V_G = -3$ V, demonstrating the repeatability of the observed effect. The inset in (*a*) shows the *I*–*V* characteristics of this device, which exhibit metallic behavior. The red solid lines represent the I–V_{bias} and conductance–V_{gate} characteristics of the device.

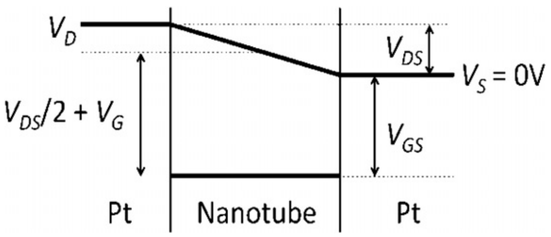

Figure 5. Effective gating of the CNT due to asymmetric source–drain bias voltage.

as a function of gate frequency ($f_G = \omega_G/2\pi$) and gate voltage (V_G) measured at a pressure of 1×10^{-4} Torr. The fundamental resonance frequencies are consistent with the 2 μm length and 1–3 nm diameter of this nanotube. The diameters of the nanotubes in this study were confirmed by Raman spectroscopy. While the SWNT devices in figure 4 are metallic, other semiconducting devices showed the same behavior. As the pressure in the chamber is increased, damping of the nanotube increases and the resonance peak becomes unobservable, as shown in earlier studies [10, 17].

Once a nanotube with low contact resistance and a strong mechanical resonance is identified, the bias voltage is varied from 0 to 1.5 V across the source and drain electrodes. Figure 4 shows the mechanical resonance frequency (f_R) plotted as a function of the dc bias voltage V_{SD} at $P = 1 \times 10^{-4}$ Torr. The small positive slope for the resonance frequency with increasing bias voltage is caused by the effective gating of the nanotube device due to the increased bias voltage. As shown schematically in figure 5, increasing the bias voltage increases the electrostatic potential at the middle of the nanotube by $V_{\text{SD}}/2$. This effective increase in gate voltage increases the electrostatic force between the nanotube and gate electrode, which in turn increases the axial tension in the nanotube [18]. Therefore, by increasing the bias voltage, the nanotube is effectively gated, thus causing the resonance frequency to change. This behavior is shown in figures 4(*a*) and (*b*), where the resonance frequency increases almost linearly with bias voltage.

Figure 4(*a*) shows an abrupt upshift in the mechanical resonance frequency of approximately 3 MHz at high bias, which is well within the experimental precision of this measurement. Figure 4(*b*) shows a similar discrete change in the slope of the resonance frequency with bias voltage. Effective gating by itself is not enough to describe this anomalous behavior at high bias. These abrupt changes in the mechanical resonance frequency occur when the bias voltage is above the threshold for emission of optical phonons [19]. The onset of this optical phonon scattering process corresponds to an abrupt increase in the phonon populations and lattice temperature of the nanotube with bias voltage. Like graphite and graphene [20], carbon nanotubes have a negative thermal expansion coefficient (TEC) [11–13, 21]. Therefore, as the suspended nanotubes become hot, they contract, thus increasing the tension in the nanotube and its mechanical resonance frequency. The onset voltage of OP emission by electron–phonon scattering can also be seen in the I_{DS} versus V_{DS} plot in figure 4(*a*). This upshift is consistent with Raman spectral measurements, which have shown a sudden downshift of the optical phonon frequencies under high bias voltages [22]. It should be noted that, while the effect of gate voltage-induced tension on the resonance frequency of a suspended nanotube is nonlinear, there are no abrupt changes in this gate-induced tension. In fact, figure 3(*b*) shows the parabolic dependence of the mechanical resonance frequency on gate voltage. The red box in figure 3(*b*) indicates the region corresponding to the three datasets in figure 4(*b*). In this regime, the resonance frequency has a linear dependence on gate voltage (at low bias). Therefore, the abrupt changes observed in figures 4(*a*) and (*b*) at high bias must originate from a different underlying mechanism.

Modeling the nanotube as a doubly clamped string [23], equation (1) gives the relation between the tension on the nanotube (T) and the fundamental resonance frequency (f_0):

$$f_0 = \frac{\sqrt{\frac{T}{m/L}}}{2L}. \tag{1}$$

Here, L and m are the suspended length and total mass of the CNT, which are 2 μm and 6.35×10^{-21} kg, respectively. Young's modulus (E) can be expressed as

$$E = \frac{\sigma}{s} = \frac{T/A}{\Delta L/L}, \quad (2)$$

where A and ΔL are the cross-sectional area and change in length due to lattice contraction. From these equations, we estimate the change in length due to thermal contraction to be $\Delta L/L \approx -2 \times 10^{-5}$ at a bias voltage of 1 V for the device in figure 4(*b*). There are currently no measured values for the TEC of individual suspended carbon nanotubes, and simulated models report values that vary significantly [24–26]. In principle, we should be able to compare our results with the TEC values from the literature. However, these suspended nanotubes are known to be in a state of extreme thermal non-equilibrium under these high-applied bias voltages [27]. We, therefore, cannot make a meaningful comparison with TEC values established under thermal equilibrium.

The desorption of gas molecules from the surface of the nanotube [28–30] is another possible explanation for the anomalous upshift in mechanical resonance frequency. For the gases present in the atmosphere, the binding energies of gases present in the atmosphere to the nanotube surface vary from approximately 82 meV (Ar) to 300 meV (O_2) [29]. While these measurements were all carried out under a vacuum of 1×10^{-4} Torr, some of the gases present in the atmosphere (namely H_2O) have binding energies (130 meV) that are considerably larger than $k_B T$ at room temperature [28, 29]. Therefore, it is possible that, as the nanotube temperature increases under high bias voltages, gas molecules desorb from the nanotube surface, thus reducing the mass density of the nanotube and increasing the mechanical oscillation frequency. It should be noted, however, that at room temperature and at a pressure of 1×10^{-4} Torr, as in this experiment, the fraction of potential binding cites occupied by H_2O molecules is <0.0042. The desorption of these molecules corresponds to a frequency shift of only 0.01 MHz, which is considerably lower than the 3 MHz that we observe in this study.

In conclusion, we have observed an anomalous shift in the mechanical resonance frequency of individual carbon nanotubes under high bias voltages. This shift is attributed to the onset of optical phonon emission by hot electrons under high bias voltages, which in turn causes lattice contraction and an associated increase in the mechanical resonance frequency. Based on a simple string under the tension model, we estimate a change in the length of the nanotube of $\Delta L/L \approx -2 \times 10^{-5}$ at a bias voltage of 1 V. Gas desorption has been ruled out due to the low pressures in the experimental setup and lower occupation ratio of the gases on the nanotube.

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