Science AAAS

www.sciencemag.org/cgi/content/full/1174290/DC1

Supporting Online Material for

Coupling Mechanics to Charge Transport in Carbon Nanotube Mechanical Resonators

B. Lassagne, Y. Tarakanov, J. Kinaret, D. Garcia-Sanchez, A. Bachtold*

*To whom correspondence should be addressed. E-mail: adrian.bachtold@cin2.es

Published 23 July 2009 on *Science* Express
DOI: 10.1126/science.1174290

This PDF file includes:

Materials and Methods

Coupling mechanics to charge transport in carbon nanotube mechanical resonators

B. Lassagne[1], Y. Tarakanov[2], J. Kinaret[2], D. Garcia-Sanchez[1] and A. Bachtold[1]
[1]CIN2(CSIC-ICN), campus UAB, E-08193 Bellaterra, Spain
[2] Department of Applied Physics, Chalmers University of Technology, SE-41296 Göteborg, Sweden

A) Mixing technique

The nanotube resonator is actuated by applying an oscillating voltage $V_g^{AC}\cos(2\pi f\,t)$ on the backgate of the wafer. It induces an oscillating electrostatic force F_{drive} on the nanotube, which reads:

$$F_{drive} = C'_g V_g^{DC} V_g^{AC} \cos(2\pi\, ft) \tag{1}$$

with C'_g the derivative of the nanotube-gate capacitance with respect to the tube deflection and V_g^{DC} the static voltage applied on the gate. The induced motion δz of the nanotube modulates C_g. To track this capacitance modulation, we use the nanotube as a frequency mixer. The measurement consists of applying a second oscillating voltage $V_{SD}^{AC}\cos(2\pi\, f\,t + 2\pi\delta f\,t + \alpha)$ between the source and the drain and measuring the mixing current I_{mix} at the frequency δf from the drain. α is the phase between the two voltage sources (δf = 10 kHz). We have:

$$I_{mix} = \frac{1}{2} V_{SD}^{AC} \frac{dG}{dV_g^{DC}} \left(V_g^{AC}\cos(\alpha) + V_g^{DC} \frac{C'_g}{C_g} \delta z \cos(\theta_m - \alpha) \right) \tag{2}$$

with $\frac{dG}{dV_g^{DC}}$ the transconductance of the nanotube and θ_m the phase between the motion and the driving force. We note that the first term in eq. 2 has a purely electrical origin and results in a background signal at any frequencies, while the second term becomes important only when f matches the mechanical resonance frequency of the nanotube.

At weak driving force we can model the nanotube resonator with a classical harmonic oscillator characterized by a mechanical resonance frequency $f_0 = 1/2\pi \cdot \sqrt{k/m_{eff}}$, with k the spring constant and m_{eff} the effective mass. The resonance (δz as a function of f) has a Lorentzian shape:

$$\delta z = \frac{1}{(2\pi)^2} \frac{F_{drive}}{m} \frac{1}{\sqrt{\left(f^2 - f_0^2\right)^2 + \frac{f_0^2 f^2}{Q^2}}} \tag{3}$$

with Q the quality factor. To extract the parameters f_0 and Q, we fit the measured $I_{mix}(f)$ to

$$I_{mix} = A + B\frac{\cos(\theta_m - \alpha)}{\sqrt{\left(f^2 - f_0^2\right)^2 + \frac{f_0^2 f^2}{Q^2}}} \quad (4)$$

The parameter A corresponds to the background current signal, which is extracted from the experimental data far from the resonance. B is a fitting parameter.

B) Coupling mechanical oscillations to conducting electrons in the Coulomb blockade regime

In the Coulomb blockade regime, the electronic charge residing on the dot is $q_{dot} = -Ne$, with N the number of charges. The electrostatic potential of the dot V_{dot} reads

$$V_{dot} = \frac{-Ne}{C_{dot}} + \frac{C_g V_g^{DC}}{C_{dot}} = \frac{q_{dot} - q_c}{C_{dot}} \quad (5)$$

with $q_c = -C_g V_g^{DC}$ the control charge and C_{dot} the dot capacitance.

The electrostatic force applied on the nanotube F_{el} is:

$$\begin{aligned} F_{el} &= \frac{1}{2} C'_g \left(V_g^{DC} - V_{dot}\right)^2 \\ F_{el} &\approx \frac{1}{2} C'_g \left(V_g^{DC\,2} - 2V_g^{DC} V_{dot}\right) \end{aligned} \quad (6)$$

The first term in eq. 6 is constant. Hence, the relevant force F due to Coulomb blockade can be written as

$$F = -\frac{C'_g V_g^{DC}}{C_{dot}}\left(q_{dot} - q_c\right) \quad (7)$$

We note that this force vanishes at high temperature. Indeed, we have $q_{dot} = q_c$ when the thermal energy $k_B T$ is larger than the charging energy $E_c = e^2/C_{dot}$.

We now assume that the nanotube is mechanically oscillating as a harmonic oscillator

$$\delta z = \delta z_0 e^{i2\pi f t} \quad (8)$$

with δz_0 the amplitude of the motion and f the frequency. As a result, the tube-gate capacitance and the control charge oscillate as

$$\delta C_g = C_g{}' \delta z_0 e^{i2\pi f t} \quad (9)$$

$$\delta q_c = -\delta C_g V_g^{DC} \quad (10)$$

when δz_0 is small. Consequently, the dot charge is also oscillating

$$\delta q_{dot} = e\delta P = e\frac{dP}{dq_c}\delta q_c \tag{11}$$

with P the occupation probability of the dot. We note that P is related to the conductance G of the dot in a simple way in the quantum regime of Coulomb blockade when the level spacing $\Delta E >> k_B T$ (C.W.J. Beenakker, *Phys. Rev. B* **44**, 1646 (1991))

$$G = \frac{\Gamma}{2}eC_{dot}\frac{dP}{dq_c} \tag{12}$$

Γ being the tunnel rate at the contacts. Thus, δq_{dot} is given by

$$\delta q_{dot} = \frac{2G}{C_{dot}\Gamma}\delta q_c \tag{13}$$

Turning back to the force F, we have

$$\delta F = -\frac{C'_g V_g^{DC}}{C_{dot}}\left(\delta q_{dot} - \delta q_c\right) = \frac{\left(C_g{}' V_g^{DC}\right)^2}{C_{dot}}\left(\frac{2G}{C_{dot}\Gamma} - 1\right)\delta z_0 e^{i2\pi f t} \tag{14}$$

This force is proportional to δz as $\delta F = -\delta k\,\delta z$ so it is equivalent to a spring force and it changes the resonance frequency as

$$\delta f = -\frac{f_0}{2}\frac{C_g{}'^2}{k}\frac{\left(V_g^{DC}\right)^2}{C_{dot}}\left(\frac{2G}{C_{dot}\Gamma} - 1\right) \tag{15}$$

We now look at dissipation. When the nanotube position is oscillating, the charge on the nanotube oscillates as δq_{dot} and has to flow through the tunnel resistance at the nanotube-electrode interface with the current

$$\delta I_{dot} = 2\pi i f\,\frac{2G}{C_{dot}\Gamma}V_g C'\delta z_0 e^{i2\pi f t} \tag{16}$$

The energy loss E_{diss} during one oscillation cycle is given by

$$E_{diss} = \int_0^{1/f}\frac{1}{G}\delta I_{dot}^2 dt = \left(\frac{2G}{C_{dot}\Gamma}V_g^{DC}C_g{}' z_0\right)^2\frac{2\pi^2 f}{G}$$

(17)

The energy stored by the mechanical resonator is $E_m = \frac{1}{2}k\delta z_0^2$. Hence, the quality factor Q quantifying the losses reads

$$1/Q = \frac{1}{2\pi}\frac{E_{diss}}{E_m} = 2\pi f\,\frac{C_g{}'^2}{k}\left(\frac{2V_g^{DC}}{C_{dot}\Gamma}\right)^2 G \tag{18}$$

We note that δf and Q could be expressed as functions of $\frac{dP}{dq_c}$ (and not G). However, the measured Coulomb blockade peaks are not fully periodic in V_g^{DC} and the peak heights are different (Fig. 3A). This reflects that the nanotube dot is not perfect and is to some extent disordered. To include these irregularities in the model, we choose to express δf and Q as functions of G. This also allows us to obtain a better agreement between the model and measurements for the oscillations of δf and Q.

C) Alternative description based on rate equation

An alternative way to describe the coupling between mechanical oscillations and the charge transport is to employ the rate equation. Assuming that the charging energy E_c is the dominant energy, we consider that only two charge states are available close to a conductance peak. They are labeled N and $N+1$ with probability P and $1\text{-}P$, respectively. The rate equation reads

$$\frac{d}{dt}P = -\Gamma_{on} \times P + \Gamma_{off} \times (1-P) \tag{19}$$

with Γ_{on} (Γ_{off}) the rate for electrons to tunnel onto (off) the dot. In addition, we have

$$\begin{aligned}
\Gamma_{on} &= \sum_{S,D} \Gamma_0 \psi(\mu_{el} - eV_{S(D)} - E_F) \\
\Gamma_{off} &= \sum_{S,D} \Gamma_0 \left(1 - \psi(\mu_{el} - eV_{S(D)} - E_F)\right) \\
\mu_{el} &= \mu_c + U_{el}(N+1) - U_{el}(N) \\
U_{el}(N) &= \frac{q_{dot}^2}{2C_{dot}} - \frac{q_{dot}}{C_{dot}} q_c
\end{aligned} \tag{20}$$

with Γ_0 the tunnelling rate at the two contacts (barriers are considered to be symmetric), ψ the Fermi-Dirac distribution, μ_{el} the electrochemical potential of the dot, $V_{S(D)}$ the voltage of the source (drain), E_F the Fermi Energy at the contacts, μ_c the chemical potential of the dot and $U_{el}(N)$ the electrostatic energy of the dot with N electrons. Here, $q_c = -C_S V_S - C_D V_D - C_g V_g$ is the generalized control charge, with C_s and C_d the source and drain capacitances of the quantum dot.

Mechanical oscillations and charge transport are coupled via the position dependent rates Γ_{on} and Γ_{off}. Considering small δz_0 (and $V_{S(D)} = 0$) we have from eq. 8, 9 and 10

$$\mu_{el} = \mu_{el}^{eq} + \frac{e}{C_{dot}} \delta q_c \tag{21}$$

so that

$$\Gamma_{on} = \Gamma_{on}^{eq} + \Gamma_{on}' \frac{e}{C_{dot}} \delta q_c$$
$$\Gamma_{off} = \Gamma_{off}^{eq} + \Gamma_{off}' \frac{e}{C_{dot}} \delta q_c \quad (22)$$

where μ_{el}^{eq}, $\Gamma_{on(off)}^{eq}$ and $\Gamma'_{on(off)}$ are the electrochemical potential in the absence of mechanical oscillations, the rate in the absence of mechanical oscillations and the derivative of the rate with respect to μ_{el}. It is straightforward to show that *P* oscillates with the amplitude δP

$$\delta P = -\frac{e}{C_{dot}} \frac{\psi'(\mu_{el}^{eq})}{1 + i\frac{\omega}{\Gamma_\Sigma}} \delta q_c \quad (23)$$

with $\Gamma_\Sigma = 2\,\Gamma_0$ and Ψ' the derivative of the Fermi-Dirac distribution with respect to energy.

Using eq 7, eq 23 and $\delta q_{dot} = -e\delta P$, we get

$$\delta F = \frac{\left(C'_g V_g^{DC}\right)^2}{C_{dot}} \left(\frac{e^2}{C_{dot}} \frac{\psi'(\mu_{el}^{eq})}{1 + \left(\frac{\omega}{\Gamma_\Sigma}\right)^2} - 1 \right) \delta z - \frac{i\omega\delta z}{\Gamma_\Sigma} \frac{\left(C'_g V_g^{DC}\right)^2}{C_{dot}} \frac{e^2}{C_{dot}} \frac{\psi'(\mu_{el}^{eq})}{1 + \left(\frac{\omega}{\Gamma_\Sigma}\right)^2} \quad (24)$$

The first term is real so it is in phase with the motion. It is responsible of the shift of the resonance frequency. The second term in eq. 2 (imaginary part) is out of phase with the motion and leads to energy dissipation. In the quantum regime of Coulomb blockade, the shift of the resonance frequency and the quality factor become similar to eq. 15 and eq. 18, respectively, provided that $\left(\frac{\omega}{\Gamma_\Sigma}\right) << 1$.

D) Numerical simulations

We assume, in our analytical analysis and for the numerical modeling, that the lowest vibrational eigenmode of the doubly-clamped nanotube is excited. We describe the nanotube deflection by a displacement *z*, which is well described by the mass-spring equation

$$\ddot{z} = -(2\pi f_0)^2 z - \frac{\gamma}{m_{eff}} \dot{z} + \frac{F_{el}}{m_{eff}} \quad (25)$$

where $\gamma = 2\pi m_{eff} f_0 / Q_0$ is the damping term and Q_0 is the quality factor arising from the loss mechanisms different from the electron-vibration coupling. F_{el} is given by

$$F_{el} = \frac{1}{2} C'_g \left(V_g - \frac{\langle q_{dot} \rangle - q_c}{C_{dot}} \right)^2 \quad (26)$$

where $\langle q_{dot} \rangle$ is the charge on the dot averaged over a long time compared to the electron tunneling time (the average charge can be introduced if the electron tunneling time is much shorter than the oscillation period). We define V_{g0}^{DC} as the gate voltage for which the energy is at the middle of the Coulomb gap. Moreover, $\langle q_{dot} \rangle = -eN - eP$ (assuming N electrons on the dot). The probability *P* is evaluated from eq. 19. The electric current at the drain electrode is given by

$$I(t) = eP \times \Gamma_{on}^D - e(1 - P)\Gamma_{off}^D \quad (27)$$

Γ_{on}^D and Γ_{off}^D are given by

$$\begin{aligned} \Gamma_{on}^D &= \Gamma_0 \psi(\mu_{el} - eV_D - E_F) \\ \Gamma_{off}^D &= \Gamma_0 (1 - \psi(\mu_{el} - eV_D - E_F)) \end{aligned} \quad (28)$$

In the simulation, we reproduce the mixing experiment by assuming that the applied gate voltage consists of a constant part δV_g^{DC}, which gives the voltage offset from V_{g0}^{DC}, and an alternative AC voltage with amplitude V_g^{AC} modulated at frequency *f*, $V_g^{AC} \cos(2\pi f t)$. We consider $V_D = 0$ and $V_S = V_S^{AC} \cos(2\pi f t + 2\pi \delta f t)$. The simulation comprises iterative numerical solution of equations 19, 25 and 27 for the nanotube deflection δz and the probability *P*. After many iteration steps, the response of the system becomes periodic (*I(t)*, *z(t)* and *P* comprise only harmonics of types $2\pi(nf + m\delta f)$ where *n,m* = 0,1,2). The current amplitude at δf, which is the experimentally measured quantity, is obtained by Fourier-transforming the current

$$I_{\delta f} = \frac{2}{T_f - T_i} \left| \int_{T_i}^{T_f} I(t) e^{i 2\pi \delta f t} dt \right| \quad (29)$$

where T_i is the time at which the transient effects have disappeared and the system response is periodic, and T_f is the time at which simulation stops.
The simulation is repeated for a range of driving frequencies, after which the resonant frequency and the quality factor are extracted from the sampled function $I_{\delta f}(f)$ in the same way as from experimental data.
The following parameters have given the best agreement between calculations and experiments at T =1.5K: $V_S^{AC} = 0.02 mV$, e/C_{dot} = *3meV*, e/C_g = *34 meV*, f_0 = 50.3MHz, Q_0 = *180*, $C'^2_g / k = 1.16.10^{-20} F^2 / Nm$, Γ_0 = 1.28.10^{10} s^{-1}.

E) Dissipation at high temperature due to the electron current

Here we consider damping at high temperature that arises from the current oscillation. When the nanotube is mechanically oscillating, the charge on the nanotube is

$$\delta q_{tube} = -C_g{'} V_g^{DC} \delta z_0 e^{i2\pi f\,t} \tag{30}$$

The corresponding current is

$$\delta I_{tube} = -2\pi i f C_g{'} V_g^{DC} \delta z_0 e^{i2\pi f\,t} \tag{31}$$

The energy loss E_{diss} during one oscillation cycle is given by

$$E_{diss} = \int_0^{1/f} \frac{1}{G} \delta I_{tube}^2 dt = \left(V_g^{DC} C_g{'} \delta z_0\right)^2 \frac{2\pi^2 f}{G} \tag{32}$$

The energy stored by the mechanical resonator is $E_m = \frac{1}{2} k \delta z_0^2$. Hence, the quality factor Q quantifying the losses reads

$$1/Q = \frac{1}{2\pi} \frac{E_{diss}}{E_m} = 2\pi f \frac{C_g{'}^2}{k} \frac{V_g^{DC\,2}}{G} \tag{33}$$

Using $C_g{'}^2 / k = 6 \cdot 10^{-22}$ F^2/Nm and G=1/(20 kΩ) for the sample discussed in the paper, we obtain $Q = 7 \cdot 10^6$.