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Ultrasensitive force detection with a nanotube mechanical resonator

J. Moser[1,2], J. Güttinger[1,2], A. Eichler[1,2], M. J. Esplandiu[2], D. E. Liu[3], M. I. Dykman[3] and A. Bachtold[1,2]*

Since the advent of atomic force microscopy[1], mechanical resonators have been used to study a wide variety of phenomena, including the dynamics of individual electron spins[2], persistent currents in normal metal rings[3] and the Casimir force[4,5]. Key to these experiments is the ability to measure weak forces. Here, we report on force sensing experiments with a sensitivity of 12 zN $Hz^{-1/2}$ at a temperature of 1.2 K using a resonator made of a carbon nanotube. An ultrasensitive method based on cross-correlated electrical noise measurements, in combination with parametric downconversion, is used to detect the low-amplitude vibrations of the nanotube induced by weak forces. The force sensitivity is quantified by applying a known capacitive force. This detection method also allows us to measure the Brownian vibrations of the nanotube down to cryogenic temperatures. Force sensing with nanotube resonators offers new opportunities for detecting and manipulating individual nuclear spins as well as for magnetometry measurements.

Force sensing with a mechanical resonator consists in converting a weak force F into a displacement z that is measurable by electrical or optical means. Advances in microfabrication in the late 1990s made it feasible to reach a force sensitivity of 820 zN $Hz^{-1/2}$ with ultrasoft cantilevers[6,7]. In spite of intensive efforts over the last decade, progress in force sensitivity has been modest. These efforts include using new materials for the resonator (such as diamond[8]), improving the displacement detection[9,10] (which can reach an imprecision below that at the standard quantum limit) and developing novel resonators (such as optically levitated nanospheres[11–13]). Optimizing both the resonator and its readout has led to a record sensitivity of 510 zN $Hz^{-1/2}$ (ref. 9).

A promising strategy for measuring lower forces is to use resonators made of a molecular system, such as a carbon nanotube[14–18]. Nanotube resonators are characterized by an ultralow mass M, which can be up to seven orders of magnitude lower than that of the ultrasoft cantilevers mentioned above[7], whereas their quality factor Q can be high[19] and their spring constant k_0 low. This strategy has a great potential for generating an outstanding force sensitivity, the classical limit for which is given by

$$S_F = 4k_B T\gamma = 4k_B T\sqrt{Mk_0}/Q \quad (1)$$

where $k_B T$ is the thermal energy and γ the mechanical resistance[7]. This limit is set by the fluctuation-dissipation theorem, which associates Langevin fluctuating forces with the irreversible losses existing in a resonator, quantified by Q. Such losses may originate, for instance, from the phononic or electronic thermal bath coupled to the resonator.

Measuring the thermal vibrations (the Brownian motion) of the resonator demonstrates that its actual force sensitivity is limited by the Langevin fluctuating forces. The thermal vibrations of resonators made of single chemically synthesized nanowires have been measured at cryogenic temperatures[20]. Their mass is larger than that of nanotubes, but lower than that of typical microfabricated wires. These chemically synthesized nanowires have been successfully used as force sensors[20].

A challenge with resonators based on nanotubes is to detect their low-amplitude vibrations, because these vibrations are transduced into electrical and optical signals that are small and have to be extracted from an overwhelmingly large noise background. In particular, the thermal vibrations of a nanotube have not yet been detected below room temperature[21]. The best force sensitivity achieved thus far with nanotube resonators[15,18] has been limited by noise in the electrical measurement set-up, and has not surpassed the record sensitivity obtained with other resonators.

To efficiently convert weak forces into sizable displacements, we have designed nanotube resonators endowed with spring constants as low as ~10 μN m^{-1}. This is achieved by fabricating the longest possible single-walled nanotube resonators. The fabrication process starts with the growth of nanotubes by chemical vapour deposition onto a doped silicon substrate coated with silicon oxide. Using atomic force microscopy (AFM), we select nanotubes that are straight over a distance of several micrometres, so that they do not touch the underlying substrate once they are released[22]. We use electron-beam lithography to pattern a source and a drain electrode that electrically contact and mechanically clamp the nanotube. We suspend the nanotube using hydrofluoric acid and a critical point dryer. Figure 1a shows a 4-μm-long nanotube resonator. We characterize its resonant frequencies by driving it electrostatically and using a mixing detection method[18,23]. The lowest resonant frequency is 4.2 MHz (Fig. 1c). This gives a spring constant of 7 μN m^{-1} using an effective mass of 1×10^{-20} kg, estimated from the size of the nanotube measured by AFM (Supplementary Section S2.1). This spring constant is comparable to that of the softest cantilevers fabricated to date[6]. When changing the gate voltage V_g^{dc} applied to the silicon substrate, the resonant frequency splits into two branches (Fig. 1c). These two branches correspond to the two fundamental modes, which vibrate in directions perpendicular to one another (Fig. 1c, inset).

We have developed an ultrasensitive detection method based on parametric downconversion that (i) uses a cross-correlation measurement scheme to reduce the electrical noise in the set-up and (ii) takes advantage of the high transconductance of the nanotube in the Coulomb blockade regime to convert motion into a sizable electron current. Our detection scheme, summarized in Fig. 2a, proceeds as follows. The oscillating displacement of the nanotube, induced by the Langevin fluctuating forces, modulates the capacitance C_g between the nanotube and the gate, which in turn yields a modulation δG of the conductance of the nanotube. We apply a weak oscillating voltage of amplitude V_{sd}^{ac} on the source electrode at a frequency f_{sd} a few tens of kilohertz away from the resonant frequency f_0. (We verify that the amplitude of the thermal vibrations does not change upon varying V_{sd}^{ac};

[1]ICFO-Institut de Ciencies Fotoniques, Mediterranean Technology Park, 08860 Castelldefels (Barcelona), Spain, [2]ICN, CIN2-CSIC, Campus UAB, 08193 Barcelona, Spain, [3]Department of Physics and Astronomy, Michigan State University, East Lansing, Michigan 48824, USA. *e-mail: adrian.bachtold@icfo.es

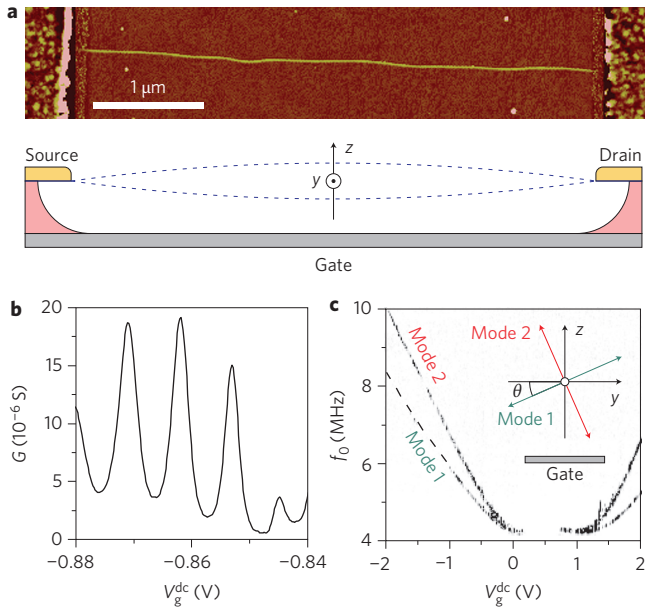

Figure 1 | Nanotube resonator with low spring constant. a, AFM image of a 4-μm-long nanotube before removing the silicon oxide (top) and schematic of the device (bottom). **b**, Conductance G of the nanotube as a function of gate bias V_g^{dc} at 1.2 K. **c**, Resonant frequency f_0 as a function of V_g^{dc} in the presence of a driving force (data obtained by measuring the mixing current with the frequency-modulation technique[18,23]). The two lowest frequency modes are shown. For values of V_g^{dc} ranging from −2 to −1 V, we indicate the resonant frequency of mode 1 with dashes because the mixing current is weak and difficult to see in the figure. The resonant frequency is highly tunable, and can change by 100% when varying V_g^{dc} by only 1.5 V. Inset: modes 1 and 2 vibrate along directions perpendicular to one another; mode 1 vibrates at an angle θ with respect to the y-direction, which runs parallel to the gate.

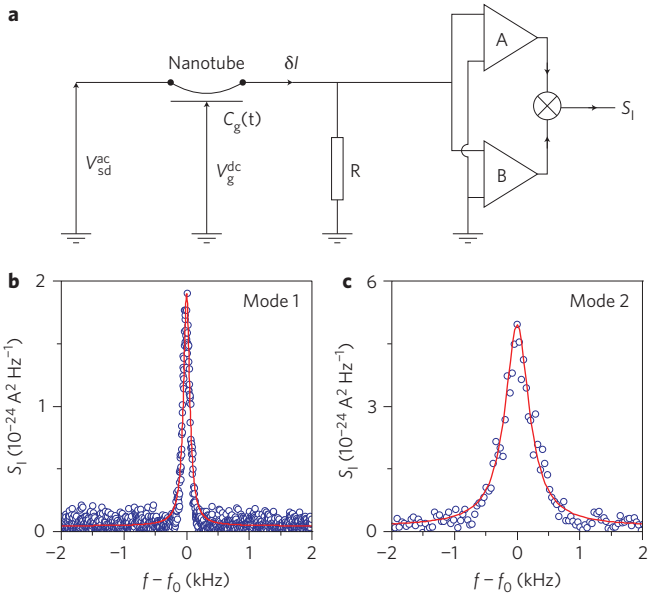

Figure 2 | Measuring thermal vibrations. a, Schematic of the cross-correlation measurement set-up. **b**,**c**, Power spectral density S_I of current fluctuations for modes 1 and 2 at 1.2 K, centred at the mode's resonant frequency. We apply $V_g^{dc} = -0.854$ V and $V_{sd}^{ac} = 89\,\mu$V. The mechanical quality factors are $Q = 48{,}000$ for mode 1 and $Q = 13{,}000$ for mode 2. We find that the quality factor of mode 2 oscillates as a function of V_g^{dc} between 8,000 and 20,000. Because the signal is weaker for mode 1, the resonance can be clearly resolved only over a limited range of V_g^{dc}.

Supplementary Section S2.5). The resulting current fluctuations at the drain electrode at frequency $\sim|f_{sd} - f_0|$ are described by

$$\delta I = V_{sd}^{ac}\delta G = V_{sd}^{ac}\frac{dG}{dV_g}V_g^{dc}\frac{C_g'}{C_g}\delta z(t)\cos(2\pi f_{sd}t) \qquad (2)$$

where dG/dV_g is the static transconductance of the nanotube, δz is the fluctuational displacement along the z-axis (Fig. 1c), and C_g' is the derivative of C_g with respect to z. To enhance δI, we select a nanotube that features sharp Coulomb blockade peaks (Fig. 1b), so that dG/dV_g is high for certain values of V_g^{dc}. We then convert current fluctuations into voltage fluctuations across a resistor ($R = 2\,k\Omega$). This voltage signal is amplified by two independent low-noise, high-impedance amplifiers. We perform the cross-correlation of the output of the two amplifiers using a fast Fourier transform signal analyser[24–26]. As a result, the voltage noise of the amplifiers cancels out, and the weak signal of the thermal vibrations can be extracted from the noise background (Supplementary Section S2.3). This procedure allows us to measure the power spectral density of current fluctuations through the nanotube, which is given by $S_I = \langle\delta I^2\rangle/\mathrm{rbw}$, where rbw is the resolution bandwidth of the measurement and $\langle\delta I^2\rangle$ is the mean square Fourier component of the time-averaged current cross-correlation at frequency $\sim|f_{sd} - f_0|$. Figure 2b,c shows the resonance of the thermal vibrations at 1.2 K for the two modes characterized above, which are hereafter labelled modes 1 and 2. The lineshapes are well described by a Lorentzian function.

We observe the coupling between thermal vibrations and electrons in the Coulomb blockade regime by collecting S_I spectra as a function of V_g^{dc} for these two modes (Fig. 3a,b). The resonant frequency of mode 2 oscillates as a function of V_g^{dc} with the same period as the conductance oscillations (Fig. 3c), but this dependence is monotonic for mode 1. As for damping, the resonance lineshape of mode 2 is much wider than the resonance lineshape of mode 1. This is readily seen in Fig. 2b,c, where we measure $Q = 13{,}000$ for mode 2 and $Q = 48{,}000$ for mode 1. To understand why modes 1 and 2 exhibit distinct features, we recall that Coulomb blockade enhances the coupling between vibrations and electrons in the nanotube[16,17,27], causing oscillations in resonant frequency as well as additional dissipation. The magnitude of both effects scales with the modulation of C_g induced by the nanotube vibrations, that is, with the nanotube displacement projected onto the z-direction perpendicular to the gate. The distinct behaviours measured for modes 1 and 2 indicate that mode 1 essentially vibrates parallel to the direction of the gate and mode 2 perpendicular to it (Fig. 1c, inset). Angle θ between the vibrations of mode 1 and the direction parallel to the gate can be estimated by comparing the integrated areas of the measured spectra of modes 1 and 2, which also depend on C_g. This gives $\theta = 19.5 \pm 2^\circ$ in the studied range of V_g^{dc} (Supplementary Section S2.4).

We then measure the force sensitivity of the resonator using a calibrating force. For this, we apply a capacitive force on mode 1 of amplitude $F_d = C_g' V_g^{dc} V_g^{ac} \sin\theta$, with $V_g^{ac}(t)$ a small oscillating gate voltage at the resonant frequency of mode 1. We perform the calibration with this mode because its high Q leads to higher force sensitivity. As a result, the driven vibrations appear as a sharp peak superimposed on the thermal resonance in the $\langle\delta I^2\rangle$ spectrum of mode 1 (Fig. 4a). The square root of the height of this peak, I_{peak}, scales linearly with V_g^{ac}, as expected (Fig. 4b). By comparing the height of the driven peak with that of the thermal resonance using

$$S_F = \frac{\text{thermal resonance height}}{\text{driven peak height}} \times F_d^2/\mathrm{rbw} \qquad (3)$$

we obtain $\sqrt{S_F} = 12 \pm 8\,\mathrm{zN\,Hz^{-1/2}}$ at $T = 1.2$ K. Here, we use

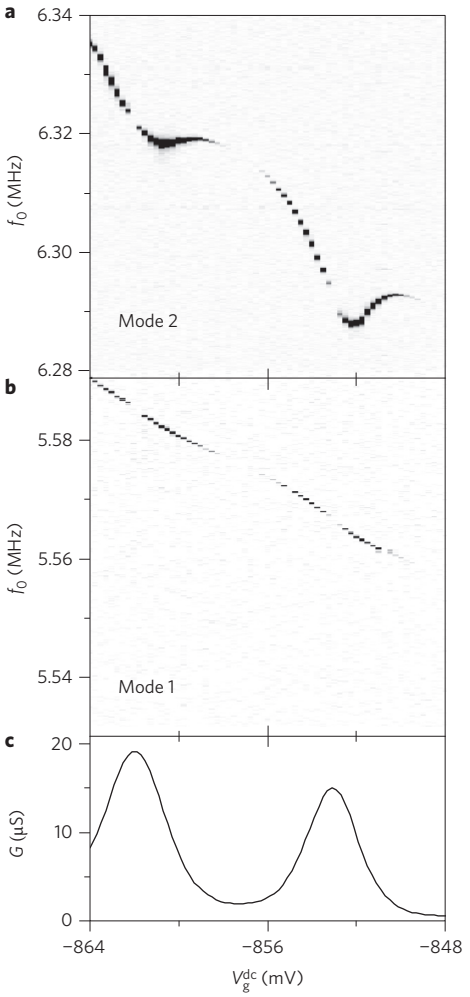

Figure 3 | Electron–vibration coupling in the Coulomb blockade regime. **a**,**b**, S_I spectra showing the resonant frequency f_0 as a function of V_g^{dc} at 1.2 K for modes 1 and 2. **c**, Conductance G as a function of V_g^{dc} at 1.2 K. S_I in **a** and **b** strongly depends on dG/dV_g, as expected from equation (2).

$C_g' = 1.2(\pm 0.4) \times 10^{-12}$ F m^{-1}, estimated from the spacing in gate voltage between the Coulomb blockade peaks and the effective distance between the nanotube and the gate. The uncertainties in $\sqrt{S_F}$ reflect imprecisions in C_g', θ and the heights of the driven peak and the thermal resonance. See Supplementary Section S1.4 for details on the measurement of the force sensitivity. Note that the voltage V_g^{ac} also induces a current of purely electrical origin (Supplementary Section S1.1), the contribution of which is negligible. This electrical contribution, which can be measured with a drive off resonance, can be detected only for exceedingly large V_g^{ac}.

We have assumed thus far that the fluctuational displacement δz is thermal in origin; here, we show that this assumption is correct. First, within the experimental uncertainties, the force sensitivity measured at 1.2 K is in agreement with the value expected from the fluctuation-dissipation theorem (equation (1)), which is 23 ± 5 zN Hz$^{-1/2}$. The latter uncertainties reflect imprecisions in the effective mass of the nanotube (Supplementary Section S2.1) and the temperature. Second, by raising the temperature to 3 K, the measured fluctuating forces increase by an amount that agrees with the fluctuation-dissipation theorem. At 3 K, we obtain a force sensitivity of 38 zN Hz$^{-1/2}$ (Fig. 4c,d). This is equal to the value expected at 3 K from

$$S_F(3\,\mathrm{K}) = S_F(1.2\,\mathrm{K})\frac{Q(1.2\,\mathrm{K})}{Q(3\,\mathrm{K})}\frac{3\,\mathrm{K}}{1.2\,\mathrm{K}}$$

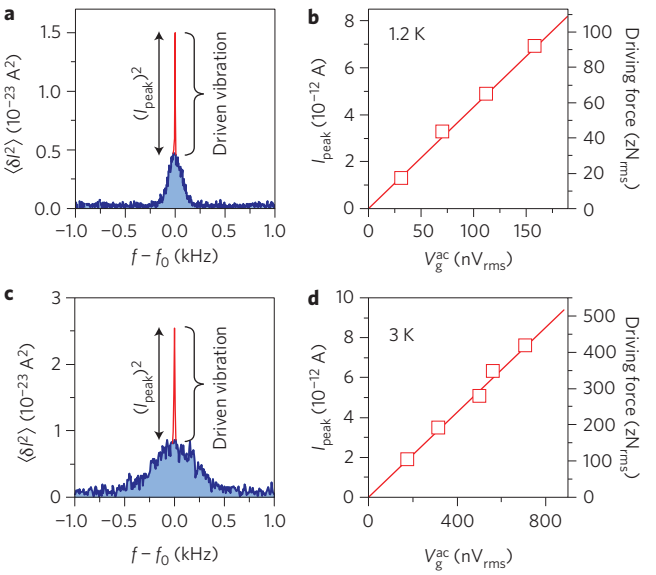

Figure 4 | Force sensing experiment. **a**, Square amplitude $\langle \delta I^2 \rangle$ of the Fourier transform of the current cross-correlation at 1.2 K in the presence of a driving force at the resonant frequency of mode 1 ($\langle \delta I^2 \rangle = S_I \times \mathrm{rbw}$). The driven vibration signal is indicated in red while the thermal vibration signal is indicated in blue. We apply $V_{sd}^{ac} = 89$ μV, $V_g^{dc} = -0.854$ V and $V_g^{ac} = 70$ nV$_{rms}$, and set rbw = 4.69 Hz. **b**, Square root of the driven resonance height in **a**, measured as a function of oscillating voltage V_g^{ac} applied to the gate. The straight line corresponds to a linear regression. Also shown is the driving force estimated from V_g^{ac}. **c**,**d**, As in **a** and **b**, but at 3 K.

according to equation (1), where we use the force sensitivity measured at 1.2 K and the quality factors extracted from the resonances at 1.2 and 3 K. Third, we estimate that non-thermal fluctuating forces do not exceed 1 zN Hz$^{-1/2}$ at 1.2 K. Such non-thermal fluctuating forces could be the electrostatic forces induced by either the Johnson–Nyquist noise of the electrical circuit or the single-electron charging–discharging process in the Coulomb blockade regime. These two non-thermal fluctuating forces are evaluated in Supplementary Section S4. Note that the force sensitivity estimated for our nanotube resonator (12 zN Hz$^{-1/2}$) does not depend on whether the fluctuating forces are thermal or non-thermal in origin; this is because the derivation of equation (3) solely requires a fluctuating force noise that is constant over the bandwidth of the mechanical resonator.

We estimate the variance of the displacement of the thermal vibrations to be $\sim(1.1\,\mathrm{nm})^2$ from the equipartition theorem. We obtain a similar variance by converting the S_I spectra into displacement fluctuations. This conversion, which depends on various parameters obtained separately, is discussed in Supplementary Section S3.

Measuring the thermal vibrations of nanotube resonators sheds new light on their dynamics. Different sources of noise in nanomechanical resonators are discussed in ref. 28. Our finding that the resonance lineshape is well described by a Lorentzian function at low temperature implies that nonlinear damping is negligible[18], the Duffing nonlinearity is weak, and the frequency noise is Gaussian and white[29].

Carbon nanotube resonators enable an unprecedented force sensitivity on the scale of 10 zN Hz$^{-1/2}$ at 1.2 K. We anticipate that the sensitivity will improve by at least a factor of 10 by operating the resonator at millikelvin temperatures. Indeed, the quality factor of nanotube resonators is enhanced at these temperatures[19], so that both low T and high Q reduce S_F. Nanotube resonators hold promise for resonant magnetic imaging with single nuclear spin

resolution[2,20,30,31]. If our nanotube resonator can be integrated into the experimental set-ups described in refs 2 and 31 without degrading the force sensitivity achieved in the present work, it should be feasible to detect a single nuclear spin[8]. A first step in this direction will be to manipulate the nuclear spins of ^{13}C atoms naturally present in nanotubes; following ref. 20, a current-carrying wire microfabricated near the nanotube resonator could be used to generate both the magnetic field gradient and the radiofrequency magnetic field pulses (Supplementary Section S5). These resonators may also be used for ultrasensitive magnetometry measurements of individual magnetic nanoparticles and molecular magnets attached to the nanotube.

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Author contributions

J.M. fabricated the device, developed the experimental set-up, and carried out the measurements. J.G. and A.E. provided support with the experimental set-up. J.G. participated in the measurements. J.G. and J.M. analysed the data. M.J.E. grew the nanotubes. D.E.L. and M.I.D. provided support with the theory and wrote the theoretical part of the Supplementary Information. J.M. and A.B. wrote the manuscript with critical comments from J.G. and M.I.D. A.B. conceived the experiment and supervised the work.

Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to A.B.

Competing financial interests

The authors declare no competing financial interests.