Ultrasensitive force detection with a nanotube mechanical resonator

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Abstract

Since the advent of atomic force microscopy [1], mechanical resonators have been used to study a wide variety of phenomena, such as 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 at a temperature of 1.2 K using a resonator made of a carbon nanotube. An ultra-sensitive method based on cross-correlated electrical noise measurements, in combination with parametric down-conversion, 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.

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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 1990’s made it feasible to reach a force sensitivity of 820 $\text{zN}/\sqrt{\text{Hz}}$ with ultra-soft cantilevers ($1 \text{zN} = 10^{-21} \text{N}$) \cite{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 \cite{8}; improving the displacement detection \cite{9, 10}, which can reach an imprecision below that at the standard quantum limit; and developing novel resonators, such as optically levitated nanospheres \cite{11–13}. Optimizing both the resonator and its readout have led to a record sensitivity of 510 $\text{zN}/\sqrt{\text{Hz}}$ \cite{9}.

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

$$S_F = 4k_B T \gamma = 4k_B T \sqrt{M k_0 / Q}.$$  \hspace{1cm} (1)

Here $k_B T$ is the thermal energy and $\gamma$ the mechanical resistance \cite{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 the electronic thermal bath coupled to the resonator. Measuring the thermal vibrations, i.e. the Brownian motion, of the resonator demonstrates that its actual force sensitivity is limited by the Langevin fluctuating forces.

A challenge with resonators based on nanotubes is to detect their low-amplitude vibrations, since 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 been detected below room temperature \cite{20}. The best force sensitivity achieved thus far with nanotube resonators \cite{15, 18} has been limited by noise in the electrical measurement setup, and has not surpassed the record sensitivity obtained with other resonators.

To efficiently convert weak forces into sizable displacements, we design nanotube resonators endowed with spring constants as low as $\sim 10 \mu \text{N/m}$. This is achieved by fabri-
cating the longest possible single-wall nanotube resonators. The fabrication process starts
with the growth of nanotubes by chemical vapor 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 micrometers, so that they do not touch the underlying
substrate once they are released [21]. 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
nanotube resonator that is 4 µm long. We characterize its resonant frequencies by driving
it electrostatically and using a mixing detection method [18, 22]. The lowest resonant fre-
quency is 4.2 MHz (Fig. 1c). This gives a spring constant of 7 µN/m using an effective mass
of 10^{-20} kg, estimated from the size of the nanotube measured by AFM (supplementary
information). This spring constant is comparable to that of the softest cantilevers fabri-
cated so far [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; they vibrate in perpendicular directions (inset to Fig. 1c).

We have developed an ultrasensitive detection method based on parametric down-
conversion, which (i) employs a cross-correlation measurement scheme to reduce the elec-
trical noise in the setup 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, which is 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 $\delta G$ 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 kHz away from the resonant frequency
$f_0$. (We verify that the amplitude of the thermal vibrations does not change upon vary-
ing $V_{sd}^{AC}$; see supplementary information.) The resulting current fluctuations at the drain
electrode at frequency $\sim |f_{sd} - f_0|$ are described by

$$\delta I = V_{sd}^{AC} \delta G = V_{sd}^{AC} \frac{dG}{dV_g} V_{g}^{DC} \frac{C_g}{\delta z(t)} \cos(2\pi f_{sd} t),$$

(2)

where $dG/dV_g$ is the static transconductance of the nanotube, and $\delta z$ the fluctuational
displacement along the $z$ axis (Fig. 1c). In order to enhance $\delta I$, 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 $R = 2 \, \text{k}\Omega$. 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 analyzer \cite{23–25}. As a result, the voltage noise of the amplifiers cancels out, while the weak signal of the thermal vibrations can be extracted from the noise background (see the supplementary information for details). This procedure allows us to measure the power spectral density of current fluctuations through the nanotube, which reads $S_I = \langle \delta I^2 \rangle / \text{rbw}$. Here, 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 $\sim |f_{sd} - f_0|$. Figures 2b,c show the resonance of the thermal vibrations at 1.2 K for the two modes characterized above, which are hereafter labeled mode 1 and mode 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 (Figs. 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) while this dependence is monotonous 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 Figs. 2b,c, where we measure $Q = 13,000$ for mode 2 and $Q = 48,000$ for mode 1. To understand why mode 1 and mode 2 exhibit distinct features, we recall that Coulomb blockade enhances the coupling between vibrations and electrons in the nanotube \cite{16, 17, 26}, 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 behaviors measured for modes 1 and 2 indicate that mode 1 essentially vibrates parallel to the direction of the gate while mode 2 vibrates perpendicularly to it (inset to Fig. 1c). The angle $\theta$ 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 results in $\theta = 19.5^\circ \pm 2^\circ$ in the studied range of $V_g^{DC}$ (see the supplementary information).

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, since 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, $I_{\text{peak}}$, 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

$$S_F = \frac{\text{thermal resonance height}}{\text{driven peak height}} \times F_d^2/\text{rbw},$$

we obtain $\sqrt{S_F} = 12 \pm 8 \text{ zN/}\sqrt{\text{Hz}}$ at $T = 1.2$ K. Here, we use $C_g' = 1.2(\pm 0.4) \times 10^{-12} \text{ F/m}$, 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 $C_g'$, $\theta$, and the heights of the driven peak and the thermal resonance. See the supplementary information for details on the measurement of the force sensitivity. Within the experimental uncertainties, the measured force sensitivity is in agreement with the value expected from the fluctuation-dissipation theorem (Eq. 1), which is $23 \pm 5 \text{ zN/}\sqrt{\text{Hz}}$. The latter uncertainties reflect imprecisions in the effective mass of the nanotube (supplementary information) and the temperature.

By raising the temperature to 3 K, the Langevin fluctuating forces increase. Using the measurement method employed at 1.2 K, we obtain a force sensitivity of $38 \text{ zN/}\sqrt{\text{Hz}}$ (Figs. 4c,d). This is in agreement with the value expected at 3 K from $S_F(3K) = S_F(1.2K) \frac{Q(1.2K)}{Q(3K)} \frac{3K}{1.2K}$ according to Eq. 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.

Measuring the thermal vibrations of nanotube resonators sheds new light on their dynamics. Different sources of noise in nanomechanical resonators were discussed in Ref. [27]. 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 [28].

Carbon nanotube resonators enable an unprecedented force sensitivity on the scale of $10 \text{ zN/}\sqrt{\text{Hz}}$ 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, 29–31]. If our nanotube resonator can be implemented
in the experimental setups described in Ref. \cite{2, 30} without degrading the force sensitivity achieved in the present work, it should be feasible to detect a single nuclear spin \cite{8}. A first step in this direction will be to manipulate the nuclear spins of $^{13}$C atoms naturally present in nanotubes using the protocol reported in Ref. \cite{31}. These resonators may also be used for ultra-sensitive magnetometery 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 setup, and carried out the measurements. J.G. and A.E. provided support with the experimental setup. J.G. participated in the measurements. J.G. and J.M. analyzed 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.
FIG. 1: Nanotube resonator with low spring constant. (a) Atomic force microscope image of a 4 μm long nanotube prior to 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, 22]). The two lowest frequency modes are shown. We indicate the resonant frequency of mode 1 with dashes for $V_g^{DC}$ ranging from -2 to -1 V, because the mixing current is weak and is difficult to see in the figure. The resonant frequency is highly tunable, as it can be changed by 100% when varying $V_g^{DC}$ by only 1.5 V. Inset: modes 1 and 2 vibrate along perpendicular directions; mode 1 vibrates at an angle $\theta$ with respect to the $y$ direction, which runs parallel to the gate.
FIG. 2: Measuring thermal vibrations. (a) Schematic of the cross-correlation measurement setup. (b,c) Power spectral density $S_I$ of current fluctuations for modes 1 and 2 at 1.2 K, centered at the mode’s resonant frequency. We apply $V_{g}^{DC} = -0.854$ V and $V_{sd}^{AC} = 89$ $\mu$V. 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 with the same period as the conductance oscillations. Since the signal is weaker for mode 1, the resonance can be clearly resolved only over a limited range of $V_{g}^{DC}$. 
FIG. 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,b) strongly depends on $dG/dV_g$, as expected from Eq. 2. We estimate the variance of the displacement of the thermal vibrations to be $\simeq (1.1 \text{ nm})^2$ 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 the supplementary information.
FIG. 4: **Force sensing experiment.** (a) The 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 ($\langle \delta I^2 \rangle = S_I \times \text{rbw}$). The driven vibration signal is indicated in red while the thermal vibration signal is indicated in blue. We apply $V_{sd}^{AC} = 89 \, \mu \text{V}$, $V_{g}^{DC} = -0.854 \, \text{V}$, and $V_{g}^{AC} = 70 \, \text{nV}_{\text{rms}}$, and set rbw = 4.69 Hz. (b) The square root of the driven resonance height in (a), measured as a function of oscillating voltage $V_{g}^{AC}$ applied to the gate. Also shown is the driving force estimated from $V_{g}^{AC}$. (c) and (d) are analogous to (a) and (b) at 3 K. The voltage $V_{g}^{AC}$ induces a current of purely electrical origin (supplementary information), whose contribution is negligible. This electrical contribution, which can be measured with a drive off resonance, can be detected only for exceedingly large $V_{g}^{AC}$.