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Symmetry breaking in a mechanical resonator made from a carbon nanotube

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Nanotubes behave as semi-flexible polymers in that they can bend by a sizeable amount. When integrating a nanotube in a mechanical resonator, the bending is expected to break the symmetry of the restoring potential. Here we report on a new detection method that allows us to demonstrate such symmetry breaking. The method probes the motion of the nanotube resonator at nearly zero-frequency; this motion is the low-frequency counterpart of the second overtone of resonantly excited vibrations. We find that symmetry breaking leads to the spectral broadening of mechanical resonances, and to an apparent quality factor that drops below 100 at room temperature. The low quality factor at room temperature is a striking feature of nanotube resonators whose origin has remained elusive for many years. Our results shed light on the role played by symmetry breaking in the mechanics of nanotube resonators.

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carbon nanotube is a unique system that can be seen both as a crystal and as a polymer. Its crystallinity confers excellent mechanical properties to nanotube-based resonators ¹⁻⁶, such as high resonant frequencies ^{7,8} and low dissipation at low temperature ^{9,10}. As a result, these resonators are well suited for ultra-sensitive detection of mass ^{11,12}, charge ^{2,3} and force ¹³. A nanotube has also much in common with a polymer as both can bend by a large amount. In a resonator, the bending can be generated by the mechanical tension that builds in during the fabrication process as well as by the electrostatic force used in most studies. This curvature is expected to have profound consequences on the dynamics of nanotube resonators, as the transverse vibrational modes lack inversion symmetry.

In a bent nanotube, if one thinks of a vibrational mode as an oscillator, its potential is not symmetric with respect to the displacement from the equilibrium position (Fig. 1a). This leads to a nonlinear term in the restoring force that depends quadratically on the displacement, $F_2 = m\beta \times \delta z^2(t)$ with m the effective mass of the resonator, β a constant quantifying the strength of the symmetry breaking effect and $\delta z(t)$ the transverse displacement of the resonator for the given mode. The mechanism underlying the effect can be understood as follows. For a resonator that is curved and clamped at both ends, the length is different for $+\delta z$ and $-\delta z$ and, therefore, the tension induced by the motion is asymmetric with respect to δz (Fig. 1b).

In a potential with broken symmetry, the equilibrium position of the mode depends on its vibrational amplitude z_{vibra} (see Fig. 1a). Indeed, if the resonator vibrates as $\delta z(t) = z_{\text{vibra}} \times \cos(\omega t)$, the quadratic term in the restoring force becomes

$$F_2 = m\beta \frac{z_{\text{vibra}}^2}{2} [1 + \cos(2\omega t)].$$
 (1)

The second term in the bracket leads to the second overtone, that is, motion at 2ω . The first term corresponds to a time-independent force and, therefore, generates a shift of the equilibrium position, $\delta z_{\rm eq}$. In other words, it is possible to move the equilibrium position by varying $z_{\rm vibra}$. The timescale of this motion is characterized by the ring-down time associated to $z_{\rm vibra}$. When considering the thermal motion of such a resonator, the

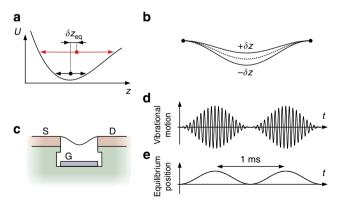


Figure 1 | Effect of curvature in a nanotube resonator. (a) Symmetry breaking of the restoring potential U(z). The equilibrium position depends on the energy of the resonator mode. (b) Schematic of a curved resonator. The dashed line represents the static profile of the resonator, that is, when it does not vibrate. The plain lines show the profiles for displacements $+\delta z$ and $-\delta z$. (c) The resonator studied consists of a carbon nanotube suspended over a trench between source (S) and drain (D) electrodes. A gate electrode (G) is defined at the bottom of the trench. The trench has a width of 1.8 μ m and a depth of \sim 350 nm. (d,e) The vibrational motion is amplitude modulated at a slow angular frequency ω_{AM} . As a result, the equilibrium position is modulated with the same period.

power spectrum of the displacement is expected to feature a peak at zero-frequency, its width being roughly the inverse of the ring-down time 14,15 . To the best of our knowledge, this low-frequency motion of the equilibrium position of oscillators with high quality factor Q has not been observed in nanomechanical resonators or other condensed-matter systems.

Here we report on a new method to measure the motion of the equilibrium position in response to resonant excitation of the vibrations. We use this method to determine the symmetry breaking strength. We further discuss the connections between symmetry breaking and the mechanics of nanotube resonators. Symmetry breaking controls the dependencies of the resonance frequency on the constant voltage applied to the gate electrode and on the amplitude of the driving force. It leads to the dependance of the resonance frequency on the vibrational amplitude; this dependence induces a sizeable spectral broadening of the mechanical resonances at room temperature.

Results

Device. The device consists of a single carbon nanotube that is clamped by two metal electrodes and is suspended over a trench. A gate electrode is defined at the bottom of the trench (Fig. 1c). The fabrication is described elsewhere¹⁰. Briefly, we pattern the three electrodes and the trench using standard electron-beam lithography techniques. We grow the nanotube by chemical vapour deposition in the last fabrication step in order to avoid contamination⁹. All measurements are performed at 65 K to avoid Coulomb blockade^{2,3}. We have studied five nanotube devices in total. We discuss in the following the data for one device. Data for a second device yielding similar results are shown in Supplementary Fig. S1 and discussed in Supplementary Note 1.

Detection method. We use a new technique to detect the motion of nanotube resonators. We capacitively drive the vibrations at $\omega_{\rm drive}$ near the resonant angular frequency ω_0 by applying a constant voltage $V_{\rm g}^{\rm dc}$ and an oscillating voltage of amplitude $V_{\rm g}^{\rm ac}$ on the gate electrode. Central to the technique is that the oscillating voltage is amplitude modulated (AM). The resulting displacement near ω_0 for not too large $V_{\rm g}^{\rm ac}$ is proportional to the driving amplitude,

$$\delta z(t) = z_{\text{vibra}} \times \cos(\omega_{\text{drive}} t - \varphi_{\text{m}}) \times [1 - \cos(\omega_{\text{AM}} t)]$$
 (2)

where the amplitude modulation has a depth of 100% and its angular frequency $\omega_{\rm AM}$ is typically $2\pi \times 1\,{\rm kHz}$ (Fig. 1d; we checked that the measurements do not depend on $\omega_{\rm AM}$ for $\omega_{\rm AM}$ up to $2\pi \times 10\,{\rm kHz}$); $\varphi_{\rm m}$ is the phase difference between the displacement and the driving force. We apply a constant voltage $V_{\rm sd}^{\rm dc}$ to the source electrode and measure from the drain electrode the low-frequency current at $\omega_{\rm AM}$ with amplitude $I_{\rm LF}$ using a lock-in amplifier (Fig. 1e; see Methods for details). We show below that this technique allows us to measure the motion of $\delta z_{\rm eq}$ associated with the symmetry breaking in nanotube resonators.

We observe that $I_{\rm LF}$ features a peak when $\omega_{\rm drive}$ is swept through a mechanical resonance (Fig. 2a); the mechanical resonance is also verified by directly measuring the vibrational motion using the frequency modulation (FM) mixing technique (Fig. 2b)⁴. The height $I_{\rm LF}^{\rm max}$ of the peak in $I_{\rm LF}$ goes linearly to zero as $V_{\rm sd}^{\rm dc}$ is decreased (Fig. 2c,d; black squares). These data show that the detected peak in $I_{\rm LF}$ is related to the modulation of the nanotube conductance $\delta G = I_{\rm LF}/V_{\rm sd}^{\rm dc}$ at $\omega_{\rm AM}$. It rules out an artefact related to the capacitive coupling between the gate and the source electrodes. (This coupling could result in a sizeable AM oscillating voltage at the source electrode, and could thus drive the resonator, but $I_{\rm LF}$ would then be independent of $V_{\rm sd}^{\rm c}$.)

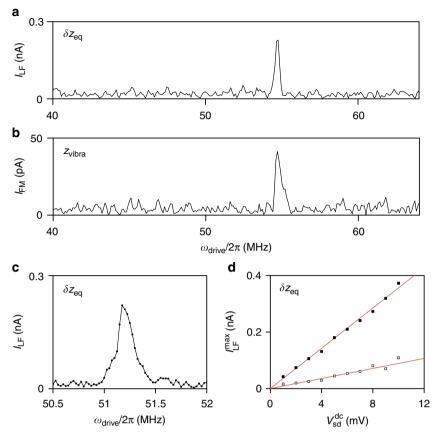


Figure 2 | Characterization of the low-frequency current. (a) Low-frequency current I_{LF} measured at angular frequency ω_{AM} as a function of angular driving frequency ω_{drive} . We use an oscillating gate voltage $V_g^{ac} = 0.53$ mV, a source voltage $V_{sd}^{dc} = 10$ mV and a gate voltage $V_g^{dc} = -0.45$ V. (b) Mechanical vibrations detected with the FM technique⁴. $V_{FM} = 1.1$ mV and $V_g^{dc} = -0.45$ V. (c) I_{LF} versus ω_{drive} with $V_g^{ac} = 1.1$ mV, $V_{sd}^{dc} = 10$ mV and $V_g^{dc} = -0.4$ V. (d) The height I_{LF}^{max} of the peak in I_{LF} as a function of V_{sd}^{dc} measured at ω_{AM} (black squares) and $2\omega_{AM}$ (open squares). $V_g^{ac} = 2.2$ mV and $V_g^{dc} = -0.4$ V.

We also observe a current peak when setting the reference (angular) frequency of the lock-in amplifier to $2\omega_{\rm AM}$. The peaks measured at $\omega_{\rm AM}$ and $2\omega_{\rm AM}$ are similar in that they appear at the same driving frequency and their heights depend linearly on $V_{\rm sd}^{\rm dc}$ (Fig. 2d). However, the height of the peak measured at $2\omega_{\rm AM}$ is four times smaller. These observations suggest that the measured peaks are related to a nonlinearity that scales as $(\delta z(t))^2$ and therefore $z_{\rm vibra}^2$. (Indeed, if $\delta z(t) \propto 1 - \cos(\omega_{\rm AM} t)$, any physical quantity that is proportional to $(\delta z(t))^2$ will be modulated at $\omega_{\rm AM}$ and $2\omega_{\rm AM}$ with a ratio of four between the amplitudes of the two components.)

Estimation of vibrational amplitude. We estimate $z_{\rm vibra} \simeq 2.1$ nm at resonant frequency for the driving force used in Fig. 2c. For this, we use the 2-source mixing technique with a driving voltage of 1.1 mV (Fig. 3a,b). The value of $z_{\rm vibra}$ is inferred by comparing the signals on and away from the resonance 16 (see Methods).

Driving different eigenmodes. The peak in $I_{\rm LF}$ is detected only for a fraction of the mechanical eigenmodes. In the resonator discussed thus far, the peak is observed for the second eigenmode but not for the first one (by comparing $I_{\rm LF}$ in Fig. 4a and the current in Fig. 4b obtained with the FM mixing technique). In all the five studied resonators, we find that about half of the eigenmodes feature a peak in $I_{\rm LF}$.

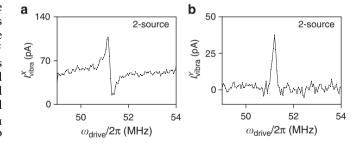


Figure 3 | Vibrational motion measured with the 2-source mixing technique. (a) X quadrature (l_{vibra}^{Y}) and (b) Y quadrature (l_{vibra}^{Y}) of the current measured with the lock-in amplifier. l_{vibra}^{X} consists of a current proportional to the in-phase component of the vibrational displacement in addition to a purely electrical background current. l_{vibra}^{Y} is proportional to the quadrature component of the vibrational displacement. The oscillating gate voltage is $V_{\rm sd}^{\rm ac}=1.1\,{\rm mV}$, the oscillating source voltage $V_{\rm sd}^{\rm ac}=0.3\,{\rm mV}$ and the constant gate voltage $V_{\rm gd}^{\rm dc}=-0.4\,{\rm V}$.

Discussion

We now discuss different possible origins of the peak in $I_{\rm LF}$. It could be related to the nonlinear capacitive coupling between the nanotube and the gate electrode, which leads to a $(\delta z(t))^2$ nonlinearity in the conductance of the nanotube. However, we estimate that the current associated to this effect is $I_{\rm LF}^{\rm capa}=10\,{\rm pA}$, which is 20 times smaller than the measured value in Fig. 2c

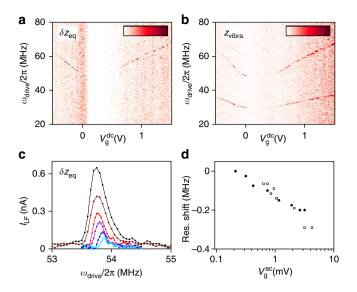


Figure 4 | Response of the low-frequency current to static and oscillating forces. (a) Low-frequency current I_{LF} measured at angular frequency ω_{AM} as a function of angular driving frequency $\omega_{
m drive}$ and constant gate voltage $V_{\rm g}^{\rm dc}.$ The oscillating gate voltage is $V_{\rm g}^{\rm ac}=0.53\,\rm mV$, and the source voltage $V_{sd}^{dc} = 10 \text{ mV}$. Colour bar: 0 (white) to 280 pA (dark red). The background signal varies with $V_{\rm g}^{\rm dc}$; this variation likely has a purely electrical origin. The number of measurement points is kept as low as possible so that resonances are captured with about three points along the frequency axis. (**b**) Current as a function of $\omega_{\rm drive}$ and $V_{\rm g}^{\rm dc}$ measured with the FM technique. $V_{FM} = 2.2 \text{ mV}$. Colour bar: 0 (white) to 170 pA (dark red). (c) Measured lineshapes of I_{LF} as a function of ω_{drive} for different V_g^{ac} . $V_g^{ac} = 4.2$, 3.5, 2.5, 1.6, 1 and 0.6 mV, from top to bottom. (d) Resonance shift extracted from the data in **c** (black dots) and from FM measurements (open squares).

(Methods). Thus, we reject the $(\delta z(t))^2$ nonlinearity induced by the capacitive coupling as the physical origin of the peak in I_{LF} . Neither is the I_{LF} peak attributed to the nonlinearity of the conductance in gate voltage9, as it leads to a current that is three orders of magnitude lower than that measured in Fig. 2c (Methods). Another mechanism for the I_{LF} peak could be the piezoresistance of the nanotube, whose dependence on the displacement is quadratic to a good approximation 17. The piezoresistance effect in nanotubes is by far strongest for positive gate voltages (where electrons tunnel from the p-doped regions of the nanotube near the metal electrodes into the n-doped region of the suspended part of the nanotube 18,19). However, the observed height of the peak in $I_{\rm LF}$ can be as large for negative as for positive gate voltages (see Fig. 4a). We can thus rule out the piezoresistive effect.

It is compelling to assume that the peak in I_{LF} is due to symmetry breaking of the vibrations. For the AM modulation $\propto [1 - \cos(\omega_{AM}t)]$, I_{LF} depends on the maximal displacement of the equilibrium position $\delta z_{\rm eq}^0$ as

$$I_{\rm LF} = V_{\rm sd}^{\rm dc} V_{\rm g}^{\rm dc} \partial_{V_{\rm g}} G \times \frac{\partial_z C_{\rm g}}{C_{\sigma}} \delta z_{\rm eq}^0 \tag{3}$$

where $\delta z_{\rm eq}^0$ is proportional to $z_{\rm vibra}^2$. Here $\partial_z C_{\rm g}$ is the derivative of the nanotube-gate capacitance $C_{\rm g}$ with respect to displacement; it is determined from Coulomb blockade measurements at helium temperature (see Supplementary Fig. S2). From the measured $I_{\rm LF}^{\rm max}$ in Fig. 2c, we get that $\delta z_{\rm eq}^0 = 0.18$ nm and $\beta = 4.3 \times 10^{24}$ m $^{-1}$ s $^{-2}$ using the relation $\beta = \omega_0^2 \delta z_{\rm eq}^0 / z_{\rm vibra}^2$ (Methods). This value for the symmetry breaking strength can be compared

with the one estimated from the measurement of ω_0 as a function

of the oscillating driving force. Figure 4c,d Shows that the peak in I_{LF} shifts to lower frequency upon increasing the driving force. Disregarding the cubic restoring force (which in nanotubes leads to the shift in the opposite direction, see ref. 20), we obtain from the shift in ω_0 that $\beta = 4.1 \times 10^{24} \,\mathrm{m}^{-1} \,\mathrm{s}^{-2}$ (Methods). This value agrees with the one estimated from I_{1F}^{max} , demonstrating that the peak in I_{LF} is due to symmetry breaking of

The strength of symmetry breaking can be made large in nanotubes, as it scales as $\beta \simeq \frac{E}{\rho} z_s \left(\frac{\pi}{L}\right)^4$ and the length L can be as short as 100 nm (refs 7,8). This expression is derived for the fundamental mode of a rod (Supplementary Equation S6 in the Supplementary Information of ref. 20), and z_s is the characteristic static displacement induced by the bending. Assuming that z_s ranges from 1 to 10 nm, and using $L = 1.8 \,\mu\text{m}$ and the graphite density $\rho = 2{,}300 \,\mathrm{kg} \,\mathrm{m}^{-3}$ and Young modulus E = 1 TP, we obtain $\beta = 3 - 30 \times 10^{24} \,\mathrm{m}^{-1} \,\mathrm{s}^{-2}$, which is consistent with the value obtained from our measurements. The quadratic nonlinear force associated to symmetry breaking is three orders of magnitude larger than the quadratic electrostatic force, $-\partial_z^3 C_{\rm g} \left(V_{\rm g}^{\rm dc}\right)^2/2m \times \delta z^2(t)$. The observed decrease of ω_0 with the increasing resonant driving in Fig. 4c,d indicates that the cubic nonlinear (Duffing) force has no substantial effect on the dynamics of the resonator. This points out that the actual static deformation z_s is large compared with the vibration amplitude (because the dynamical cubic restoring force scales as $F_3 \simeq F_2 \cdot \delta z(t)/z_s$, and thus supporting our above assumption that $z_{\rm s} = 1 - 10 \, \rm nm$.

The observation of a peak in I_{LF} for only about half of the mechanical eigenmodes indicates that β varies from one eigenmode to the next. This is something expected from the interplay between the shapes of the vibrational eigenmodes and the static deformation along the nanotube if the static displacement is primarily in one plane. Our data suggest that the static displacement is essentially perpendicular to the gate electrode. In such a geometry, the lowest-frequency eigenmode detected in Fig. 4b corresponds to the lowest-energy mode vibrating (essentially) parallel to the surface of the gate electrode, as shown in ref. 20. A comparatively small static deformation of the nanotube towards the gate electrode does not break the vibration symmetry of this mode (because the elastic tension inside the nanotube is equal for $+\delta z$ and $-\delta z$). As a result, I_{LF} should be weak, in agreement with the measurements. The second eigenmode in Fig. 4b is assigned to the lowest-energy mode vibrating in a direction (essentially) perpendicular to the gate electrode²⁰. In the presence of a static deformation towards the gate electrode, this mode experiences symmetry breaking of vibrations. A peak shows up in I_{LF} , as observed in Fig. 4a.

Having shown that symmetry breaking leads to motion at (nearly) zero-frequency, we demonstrate other connections between symmetry breaking and the mechanics of nanotube resonators. A hallmark of nanotube resonators is that the resonance frequency can be widely tuned with V_{σ}^{dc} . Symmetry breaking is expected to control this tunability in ω_0 by an amount

$$\Delta\omega_0 = \frac{\beta \partial_z C_g}{2m\omega_0^3} \left(V_g^{\rm dc}\right)^2 \tag{4}$$

as shown in Supplementary Discussion (V_g^{dc} is here offset so that $V_{\rm g}^{\rm dc} = 0$ when ω_0 is minimum). We estimate that $m \simeq 4$ ag assuming that the length of the nanotube is equal to the trench width (1.8 µm) and using the typical radius (1.5 nm) obtained with our chemical vapour deposition recipe. Using the curvature of $\Delta\omega_0(V_{\rm g}^{\rm dc})$ near the minimum of ω_0 , we get that $\beta = 3(\pm 1) \times 10^{24} \, {\rm m}^{-1} \, {\rm s}^{-2}$, which is close to the value estimated above. This result underscores that symmetry breaking is connected to the response of the resonance frequency to $V_{\rm g}^{\rm dc}$. We emphasize that Equation (4) is only valid for not too large $V_{\rm g}^{\rm dc}$, as this is the leading-order term of the expansion of ω_0 in $V_{\rm g}^{\rm dc}$. Our results indicate that β and the bending of the nanotube both remain essentially constant over the whole range of $V_{\rm g}^{\rm dc}$ that we studied. This suggests that the bending is a consequence of the mechanical tension built in during the fabrication process. In the future, it will be interesting to measure β as a function of $V_{\rm g}^{\rm dc}$ for other nanotube resonators.

In the presence of thermal vibrations, symmetry breaking leads to spectral broadening²¹. Because the amplitude of thermal vibrations fluctuates in time, the nonlinearity-induced shift in ω_0 (Fig. 4d) also fluctuates and, therefore, broadens the mechanical resonance. The broadening in ω_0 reads

$$\overline{\delta\omega} = 5\beta^2 k_{\rm B} T / 6m\omega_0^5 \tag{5}$$

when the cubic restoring force is negligible compared with the quadratic one (Supplementary Discussion). Using $\beta=4.3\times10^{24}\,\mathrm{m^{-1}\,s^{-2}}$, we get $\delta\omega=2\pi\times7.5\times10^5\,\mathrm{Hz}$ at room temperature. This corresponds to an apparent quality factor of 67, which is comparable to the value of $\simeq50$ measured with the FM technique (Supplementary Fig. S3). We emphasize that this broadening is analogous to dephasing of two-level systems and qubits, which sets the characteristic time T_2 . The measured broadening is not related to dissipation, so that the energy relaxation time could be much longer than $1/\delta\omega$ (in fact, it is in this case that Equation (5) gives the spectral broadening). For eigenmodes with a small β , the broadening can be due to the cubic restoring force²¹. Mechanical resonances might be further broadened by the coupling between eigenmodes²¹, as shown by recent simulations of nanotube resonators²².

We assumed in our analysis of $I_{\rm LF}$ that the response of the amplitude of the vibrational motion is linear with the driving force. When the response becomes nonlinear at large driving forces due to the restoring force nonlinearity, the ratio of $I_{\rm LF}$ at $\omega_{\rm AM}$ and $2\omega_{\rm AM}$ is expected to deviate from four. Calculations show that the width of the peak in $I_{\rm LF}$ remains nearly constant upon varying the driving force, in contrast to the measurements in Fig. 4c. A general theory that incorporates nonlinearities in both the restoring force and damping $^{10,23-26}$ as well as thermal vibrations is beyond the scope of this article. We note that our new technique to measure the motion of the equilibrium position allows one to study the response of the resonator over a broad parameter range in driving force.

In conclusion, we demonstrate that symmetry breaking leads to motion at nearly zero-frequency in response to resonant excitation of the vibrations. Our results indicate that symmetry breaking of vibrational modes also leads to such important dynamical properties as the apparent low quality factor of nanotube resonators at 300 K, and the shift of the vibration frequency in response to both (i) the static gate voltage and (ii) the amplitude of the oscillating driving force. A future strategy to improve the apparent Q at 300 K is to tune β with the gate voltage in order to compensate the spectral broadening due to symmetry breaking with that due to the Duffing nonlinearity. Symmetry breaking is important for other vibrational systems of current interest, such as graphene resonators 10,27-31 and levitating particles 32-34. Our new technique may help to reveal this effect in such systems. Symmetry breaking also leads to mode mixing and to parametric resonance in response to additive driving. This holds promise for a number of applications, such as controlled mode mixing $^{35-37}$ and phase noise cancellation $^{38-40}$.

Methods

Electrical characterization of the nanotube device. In Supplementary Fig. S2a, we show the electrical conductance G of the nanotube device as a function of the gate voltage $V_{\rm g}^{\rm dc}$ at 65 K. We find this measurement to be reproducible over a timescale of weeks (a current annealing procedure is performed every day to counter the effects of contamination with residual gas particles).

The capacitance $C_{\rm g}$ between the gate and the nanotube is determined as follows. In the Coulomb blockade regime, the separation between two conductance peaks is given by $\delta V_{\rm g} = e C_{\rm g}$ where e is the electron charge. From the measurement in Supplementary Fig. S2b, we get $C_{\rm g} = 12\,{\rm aF}$, which is in agreement with an estimation based on the device geometry:

$$C_{\rm g} = \frac{2\pi\epsilon_0 L}{\ln(2d/r)}\tag{6}$$

Here ϵ_0 is the vacuum permittivity, $L=1.8\,\mu\mathrm{m}$ is the nanotube length and $d=350\,\mathrm{nm}$ is the equilibrium distance between the nanotube and the gate electrode. As we cannot measure the diameter of the nanotube due to the large surface roughness of the electrodes in the studied device, we use a typical value for the radius $(r=1.5\,\mathrm{nm})$. We determine $\partial_z C_g$ and $\partial_z^2 C_g$ by differentiating Equation (6) and get $\partial_z C_g = 5.6\,\mathrm{pF}\,\mathrm{m}^{-1}$ and $\partial_z^2 C_g = 21\,\mathrm{\mu F}\,\mathrm{m}^{-2}$.

From the measurements of the resonance frequency as a function of $V_{\rm g}^{\rm dc}$ in Fig. 4b, we obtain a voltage offset of 0.45 V, which corresponds to the work function difference between the nanotube and the gate electrode. This offset in $V_{\rm g}^{\rm dc}$ is included in all the estimates. However, the values of $V_{\rm g}^{\rm dc}$ that we indicate in the text are always the voltages that are applied to the gate electrode.

Measurements of vibrational motion. We discuss first the frequency modulation (FM) technique⁴. A driving voltage is applied to the source electrode taking the form $V^{\rm FM} \times \cos[\omega t + \omega_\delta/\omega_{\rm L} \times \sin(\omega_{\rm L} t)]$, where ω is the carrier angular frequency, ω_δ is the angular frequency deviation (typically $2\pi \times 100$ kHz) and $\omega_{\rm L}$ is the modulation angular frequency (typically $2\pi \times 671$ Hz). It results in a current ($I_{\rm FM}$) at $\omega_{\rm L}$ at the drain electrode. This technique has a low current background and is typically more sensitive than the 2-source technique, so we preferentially use it to detect the eigenmodes of a nanotube resonator. The main drawback of the FM technique is that the measured signal is not proportional to $z_{\rm vibra}$ but to the derivative with respect to the frequency of the in-phase component of the displacement.

In the 2-source technique¹, we apply a driving voltage $V_{\rm g}^{\rm ac}{\rm cos}\omega t$ to the gate electrode in addition to a DC voltage $V_{\rm g}^{\rm dc}$. The motion of the nanotube is detected by applying a second voltage $V_{\rm s}^{\rm ac}$ to the source electrode. The two oscillating voltages are slightly detuned, and the mixing current $I_{\rm mix}$ is measured at the detuning frequency (typically $\delta\omega/2\pi=10\,{\rm kHz}$). When the displacement is written as $z(t)={\rm Re}[\tilde{z}(\omega)]\cos(\omega t)+{\rm Im}[\tilde{z}(\omega)]\sin(\omega t)$, the mixing current $I_{\rm mix}$ measured with the 2-source technique has the form 16

$$\begin{split} I_{\rm mix} &= \frac{1}{2} \, V_{\rm sd}^{\rm ac} \partial_{V_{\rm g}} G \bigg(V_{\rm g}^{\rm ac} \cos(\delta \omega t - \varphi_{\rm E}) + V_{\rm g}^{\rm dc} \frac{\partial_z C_{\rm g}}{C_{\rm g}} \, {\rm Re}[\bar{z}(\omega)] \cos(\delta \omega t - \varphi_{\rm E}) \\ &+ V_{\rm g}^{\rm dc} \frac{\partial_z C_{\rm g}}{C_{\rm g}} \, {\rm Im}[\bar{z}(\omega)] \sin(\delta \omega t - \varphi_{\rm E}) \bigg) \end{split} \tag{7}$$

where G is the conductance of the nanotube, and $\varphi_{\rm E}$ is the phase difference between the voltages applied to source and gate. For a properly tuned phase of the lock-in amplifier, the out-of-phase component of the lock-in amplifier output, Y, corresponds to the quadrature of the resonant displacement (third term in Equation (7)), whereas the in-phase component, X, corresponds to the in-phase component of the resonant displacement (second term in Equation (7)) added to a background (first term in Equation 7) that weakly depends on the frequency near resonance with a given mode (we note that it can have contributions from other modes). When the driving frequency matches the resonant frequency, we get for the Y-component of the mixing current $I_{\rm mix}$, which we denote as $I_{\rm vibra}^{Y}$,

$$I_{\text{vibra}}^{\text{Y}} = \frac{1}{2} \times V_{\text{sd}}^{\text{ac}} V_{\text{g}}^{\text{dc}} \times \partial_{V_{\text{g}}} G \times \frac{\partial_{z} C_{\text{g}}}{C_{\text{g}}} \times z_{\text{vibra}}, \tag{8}$$

where $z_{
m vibra}$ is the amplitude of resonant forced vibrations. For the considered small $V_a^{
m gc}$, $z_{
m vibra}$ is proportional to the amplitude of $V_a^{
m ac}$.

Estimation of vibration amplitude. Equation (7) allows estimating the vibration amplitude of the resonator for resonant driving by comparing the out-of-phase current on resonance, I_{vibra}^{χ} , with the background far from resonance, I_{offres}^{χ} 1,16. Using Equation (6), we get

$$z_{
m vibra} \simeq d \times \ln \left(\frac{2d}{r} \right) \frac{I_{
m vibra}^{
m V}}{I_{
m offres}^{
m V}} \frac{V_{
m g}^{
m ac}}{V_{
m g}^{
m dc}}.$$
 (9)

If we use the distance to the gate electrode $d=350\,\mathrm{nm}$ and a nanotube radius $r=1.5\,\mathrm{nm}$, the measurement in Fig. 3 yields a value of $z_{\mathrm{vibra}}=2.1\,\mathrm{nm}$ for the modulation amplitude $V_{\mathrm{g}}^{\mathrm{ac}}=1.1\,\mathrm{mV}$.

Detection of the motion of the equilibrium position. In the following, we explain in more detail the technique we develop to detect the motion of the equilibrium position of the nanotube resonator due to symmetry breaking. We drive the resonator with an AM-driving force, which causes an AM vibrational motion (Equation 2). The amplitude change of the vibration is quasi-adiabatic from the point of view of the resonator (we checked that the result is independent of the modulation period $2\pi/\omega_{\rm AM}$ down to 0.1 ms.) In the presence of AM modulation the quadratic nonlinear force $F_2 \propto \delta z(t)^2$ leads to the oscillation of the nanotube equilibrium position, $\delta z_{\rm eq}$, as illustrated in Fig. 1e.

The conductance G of the nanotube is a function of the total charge q of the nanotube. Using Supplementary Equations (S1–S3), the linear dependance of G on the displacement is

$$\delta G(t) = \partial_{V_g} G \times V_g^{\text{dc}} \frac{\partial_z C_g}{C_a} \delta z(t). \tag{10}$$

The slow oscillation of the conductance is caused by the motion of the equilibrium position due to symmetry breaking. As shown in Fig. 1e, for comparatively weak resonant modulation, where the AM vibrational motion is of the form of $\delta z(t) = z_{\rm vibra} \times \cos(\omega_{\rm drive} t) \times [1-\cos(\omega_{\rm AM} t)]$, the equilibrium position $\delta z_{\rm eq} \propto \left\lceil \overline{\delta z(t)^2} \right\rceil$ oscillates with period $2\pi/\omega_{\rm AM}$. The vibrations are nonsinusoidal,

$$\delta z_{\rm eq}(t) = \delta z_{\rm eq}^0 \left[-\cos\omega_{\rm AM} t + \frac{3}{4} + \frac{1}{4}\cos 2\omega_{\rm AM} t \right]. \tag{11}$$

The amplitude of the low-frequency current oscillation measured at $\omega_{\rm AM}$ is then simply

$$I_{\rm LF} = V_{\rm sd}^{\rm dc} \times \delta G = V_{\rm sd}^{\rm dc} V_{\rm g}^{\rm dc} \left| \partial_{V_g} G \times \frac{\partial_z C_{\rm g}}{C_{\sigma}} \right| \delta z_{\rm eq}^0. \tag{12}$$

This current corresponds to the second term in the bracket of Supplementary Equation (S11). By comparing Equation (12) to Equation (8) and recalling that $I_{\rm LF}^{\rm max}$ is defined as the low-frequency current when $\omega_{\rm drive}$ matches the resonant frequency, we get

$$I_{\rm LF}^{\rm max} = 2I_{\rm vibra}^{\rm Y} \times \frac{\delta z_{\rm eq}^{\rm 0}}{z_{\rm vibra}} \times \frac{V_{\rm sd}^{\rm dc}}{V_{\rm ed}^{\rm sd}}$$
 (13)

provided that $I_{\rm LF}^{\rm max}$ and $I_{\rm vibta}^{\gamma}$ are measured with the same driving force and $I_{\rm vibta}^{\gamma}$ is taken at the resonant frequency. (The driving force comes from the gate voltage oscillation at frequency ω ; however, in the 2-source technique used to measure $I_{\rm vibta}^{\gamma}$ the voltage amplitude is not modulated; we note again that we study the regime where $V_{\rm g}^{\rm ac}$ is comparatively small.)

Estimation of I_{LF} **due to capacitive nonlinearity.** In this section, we estimate the current that is expected due to the nonlinearity of the capacitance with respect to displacement, $\partial_z^2 C_g$ (see Supplementary Equations (S2, S3 and S11)). From Supplementary Equations (S2 and S3), the current at $\omega_{\rm AM}$ that we expect due to the capacitive nonlinearity in the presence of a DC bias voltage ($V_{\rm sd}^{\rm cc}$) has the form

$$I_{\rm LF}^{\rm capa} = V_{\rm sd}^{\rm dc} \times \delta G = \frac{1}{2} V_{\rm sd}^{\rm dc} V_{\rm g}^{\rm dc} \times \partial_{V_g} G \times \frac{\partial_z^2 C_{\rm g}}{C_{\rm c}} z_{\rm vibra}^2. \tag{14}$$

Comparing Equation (14) with Equation (8), we can express this current in terms of the 2-source mixing current measured on resonance in Fig. 3. We get

$$I_{\rm LF}^{\rm capa} = I_{\rm vibra}^{\rm Y} \times \frac{V_{\rm sd}^{\rm dc}}{V_{\rm ed}^{\rm ac}} \times \frac{\partial_z^2 C_{\rm g}}{\partial_z C_{\rm g}} z_{\rm vibra}, \tag{15}$$

where the currents are taken at the resonance frequency and for the same amplitude $V_a^{\rm ac}$. This relation is useful, because it depends on a small number of parameters. Using $I_{\rm vipta}^{\rm Y}=37~{\rm pA}$ from Fig. 3b, $V_{\rm sd}^{\rm dc}=10~{\rm mV},~V_{\rm sd}^{\rm ac}=0.3~{\rm mV},~0_zC_{\rm g}=5.6~{\rm pF}~{\rm m}^{-1},~\partial_z^2C_{\rm g}=21~{\rm \mu F}~{\rm m}^{-2}$ and $z_{\rm vibra}=2.1~{\rm nm},$ we get that $I_{\rm LF}^{\rm cap}=9.7~{\rm pA}\pm5~{\rm pA},$ which is far below $I_{\rm LF}^{\rm max}=219~{\rm pA}$ that we measured in Fig. 2c.

Estimation of I_{LF} **due to conductance nonlinearity.** We estimate the current that is expected due to the nonlinearity of the conductance with respect to the gate voltage, $\partial_{V_g}^2 G$, which is described by the last term in Supplementary Equation (S1). From Supplementary Equations (S2 and S3), the current at ω_{AM} is

$$I_{\rm LF}^{\rm cond} = \frac{1}{2} V_{\rm sd}^{\rm dc} (V_{\rm g}^{\rm dc})^2 \times \partial_{V_{\rm g}}^2 G(\partial_z C_{\rm g}/C_{\rm g})^2 z_{\rm vibra}^2. \tag{16}$$

Using $V_{\rm sd}^{\rm dc}=10\,{\rm mV}$, $V_{\rm g}^{\rm dc}=-0.4\,{\rm V}$, $\partial_z C_{\rm g}=5.6\,{\rm pF\,m^{-1}}$, $C_{\rm g}=12\,{\rm aF}$, $z_{\rm vibra}=2.1\,{\rm nm}$ and $\partial_{V_{\rm g}}^2 G=54\,{\rm \mu S\,V^{-2}}$, we get that $I_{\rm LF}^{\rm cond}=0.19\,{\rm pA}$. We determine $\partial_{V_{\rm g}}^2 G$ by twice differentiating the conductance in Supplementary Fig. S2a with respect to the gate voltage.

Estimation of the symmetry breaking strength. In the presence of the AM vibrational motion of the form of $\delta z(t) = z_{\mathrm{vibra}} \times \cos(\omega_{\mathrm{drive}} t) \times [1 - \cos(\omega_{\mathrm{AM}} t)]$ and in the limit of small δz_{eq}^0 (such that the leading-order term in the restoring force is $m\omega_0^2\delta z$), δz_{eq}^0 is related to the parameter β as

$$\omega_0^2 \delta z_{\rm eq}^0 = |\beta| z_{\rm vibra}^2 \tag{17}$$

Using Equation (17) together with Equation (13), we arrive at the relation

$$I_{\mathrm{LF}}^{\mathrm{max}} = 2I_{\mathrm{vibra}}^{\mathrm{Y}} \times \frac{V_{\mathrm{sd}}^{\mathrm{dc}}}{V_{\mathrm{vac}}^{\mathrm{qc}}} \times \frac{\mid \beta \mid}{\omega_{0}^{0}} \times z_{\mathrm{vibra}}.$$
 (18)

Here again the currents are taken at the resonance frequency and for the same amplitude $V_{\rm sc}^{\rm gc}$. This relation also depends on a small number of parameters that, in addition, are well characterized. With $I_{\rm vibra}^{\gamma}=37\,{\rm pA}$ from Fig. 3b, $V_{\rm sd}^{\rm dc}=10\,{\rm mV}$, $V_{\rm sd}^{\rm ac}=0.3\,{\rm mV}$, $\omega_0=2\pi\times51\,{\rm MHz}$, $z_{\rm vibra}=2.1\,{\rm nm}$ and $I_{\rm LF}^{\rm max}=219\,{\rm pA}$ measured in Fig. 2c, we get $\delta z_{\rm eq}^0=0.18\,{\rm nm}$ and $\beta=4.3\times10^{24}\,{\rm m}^{-1}\,{\rm s}^{-2}$. An alternative estimation of β is possible using the fact that the symmetry

An alternative estimation of β is possible using the fact that the symmetry breaking force leads to a shift of the resonance frequency with driving force. We have measured the shift of the resonance frequency as a function of the vibrational amplitude with the FM technique as well as with our new detection technique. The values measured with the two methods agree well and are plotted in Fig. 4d. We use the relation⁴¹

$$\Delta\omega_0 = \frac{3}{8} \frac{\gamma_{\text{eff}}}{\omega_0} z_{\text{vibra}}^2, \tag{19}$$

where $\Delta\omega_0$ is the shift of the resonance frequency and

$$\gamma_{\text{eff}} = \gamma - \frac{10}{9}\omega_0^{-2}\beta^2. \tag{20}$$

(γ is the coefficient of the Duffing nonlinearity, see Supplementary Equation (S8)). We estimate $\gamma_{\rm eff}=-1.8\times10^{32}\,{\rm m}^{-2}\,{\rm s}^{-2}$ from Equation (19) by inserting $z_{\rm vibra}=2.1\,{\rm nm},\,\omega_0=2\pi\times51\,{\rm MHz}$ and $\Delta\omega_0\simeq-2\pi\times150\,{\rm kHz}$ from Fig. 4d at the driving voltage $V_{\rm s}^{\rm ac}=1.1\,{\rm mV}$. We then get $\beta=4.1\times10^{24}\,{\rm m}^{-1}\,{\rm s}^{-2}$ from Equation (20) by assuming that γ is negligible. A nonzero value of $\gamma>0$ would slightly increase the estimate for β . See Supplementary Discussion for yet another method to estimate β .

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Author contributions

A.E. fabricated the devices and carried out the measurements. J.M. participated in the measurements. M.I.D. provided support with the theory and wrote the theoretical part of Supplementary Information. All the authors contributed to writing the manuscript. M.I.D. and A.B. conceived the experiment. A.B. supervised the work.

Additional information

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