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Nanomechanical vibrational response from electrical mixing measurements

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ABSTRACT

Driven nanomechanical resonators based on low-dimensional materials are routinely and efficiently detected with electrical mixing measurements. However, the measured signal is a non-trivial combination of the mechanical eigenmode displacement and an electrical contribution, which makes the extraction of the driven mechanical response challenging. Here, we report a simple yet reliable method to extract solely the driven mechanical vibrations by eliminating the contribution of pure electrical origin. This enables us to measure the spectral mechanical response as well as the driven quadratures of motion. This method is crucial for nanomechanical vibrations in the nonlinear regime, since the shape of the mechanical response depends on the physics at work. We further show how to calibrate the measured signal into units of displacement. Our method marks a key step forward in the study of nanoelectromechanical resonators based on low-dimensional materials in the nonlinear regime.

Nanomechanical resonators[1] are exquisite sensors of mass adsorption[2–4] and external forces.[5–8] These sensing capabilities enable advances in different research fields, such as mass spectrometry,[9] surface science,[10–12] heat transport,[13,14] *in situ* nanofabrication,[15] magnetic resonance imaging,[16–18] scanning probe microscopy,[19–21] nanomagnetism,[22–25] and probing viscosity in liquids.[26] Many of these studies are carried out with mechanical resonators based on low-dimensional materials, such as carbon nanotubes,[27,28] because of their tiny mass. However, the detection of motion becomes increasingly difficult as resonators get smaller.

The electrical detection of resonators based on low-dimensional materials is usually realized with a mixing-based method,[28,29] where the vibrations are driven near resonance frequency and detected at a low frequency within the *RC* bandwidth of the circuit. The electrical mixing detection has been applied to resonators based on carbon nanotubes,[6,10,12,28,30–47] graphene,[35,48–59] transition metal dichalcogenides (TMDs),[60–65] and semiconducting nanowires.[66–74] Different variants of the mixing method were developed by applying either two signals[28] on the device or one signal that is amplitude[49] or frequency[33] modulated. The transduction from displacement into current can be based on capacitive[28] or piezo-resistive measurements.[75] Methods were also implemented to measure thermal vibrations[39] and ring-downs[43,76] at temperatures down to below 0.1 K. The fundamental detection limit was theoretically investigated in Ref. 77. Despite this large amount of work, the measurement of the spectral response of nanomechanical vibrations to a driving force—the most common method to study mechanical resonators[1]—remains to be demonstrated with the mixing detection.

Here, we report on a simple, yet reliable, method to measure the spectral mechanical response to a driving force using the mixing method with two signals applied to the device. By properly tuning the phase of the measured signal, we are able to separate the signal of the mechanical vibrations from the signal of pure electrical origin inherent to the mixing method. Moreover, we use the pure electrical contribution as a resource to calibrate the displacement. The ability to measure the spectral mechanical response is a key step for exploring the rich physics of nonlinear phenomena.

We produce nanotube mechanical resonators by growing nanotubes using chemical vapor deposition on prepatterned electrodes. The

nanotube is suspended $\simeq 150$ nm above a gate electrode and connected between two metal electrodes[6,78] [Fig. 1(a)]. We clean the nanotube surface from contamination molecules by applying a large current through the device under vacuum at low temperature.[79]

We detect the vibrations of the nanotube resonator by capacitively driving it with an oscillating voltage $V_g^{ac}\cos\omega t$ on the gate electrode, applying the voltage $V_s^{ac}\cos((\omega+\delta\omega)t+\varphi_e)$ on the source electrode, and measuring the current at frequency $\delta\omega$ from the drain electrode with a lock-in amplifier[6] [Fig. 1(a)] where φ_e is the phase difference between the two oscillating voltages. We set $\delta\omega$ within the bandwidth of the circuit, and we sweep ω through the mechanical frequency ω_m ($\delta\omega \ll \omega_m$). All the measurements are carried out with the device in the single-electron tunneling regime[80–84] at the temperature $T = 6$ K.

To detect the vibrations, the nanotube has to behave as a transistor such that the conductance G depends on the charge Q in the nanotube. The application of $V_g^{ac}\cos\omega t$ modulates the charge through two terms $\delta Q = C_g\delta V_g + \delta C_g V_g$. The first term has a pure electrical origin, while the second term is proportional to the driven vibration displacement δz via $\delta C_g = C_g'\delta z$, where C_g' is the spatial derivative of the capacitance. The application of $V_s^{ac}\cos((\omega+\delta\omega)t+\varphi_e)$ enables one to mix down the modulation of G into a current oscillation at the frequency $\delta\omega$ within the circuit bandwidth via Ohm's law $I = GV_s$. The mixing intertwines the two terms of the charge modulation.

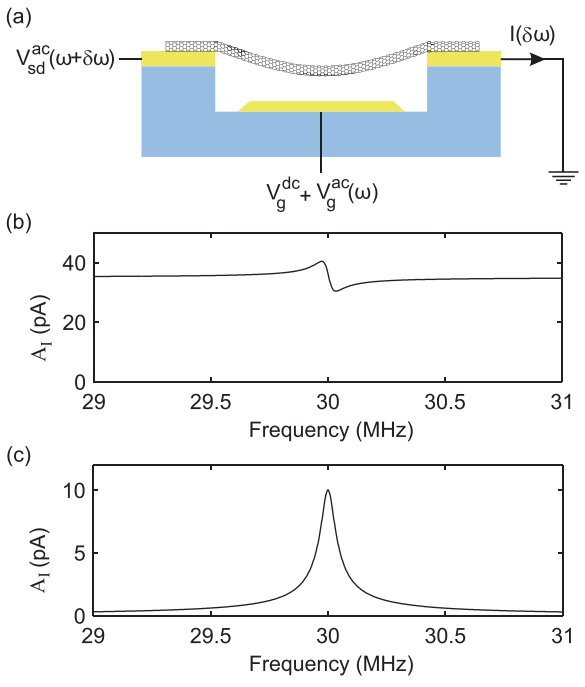

FIG. 1. (a) Schematics of the measured device. The nanotube is suspended over a gate electrode and electrically connected to two metal electrodes. Two oscillating voltage signals are applied to the device. The current I is measured with a *RLC* resonator and a low-temperature amplifier.[6] (b) Calculated response of a underdamped harmonic oscillator electromechanically driven by a capacitive force $F(\omega)\propto V_g^{ac}(\omega)$ expected from the mixing method in the limit where the mechanical displacement is much smaller than $C_gV_g^{ac}/C_g'V_g^{dc}$. (c) Same as panel b but in the opposite limit.

The downside of the mixing method is that the measured current is not directly proportional to the driven vibration displacement, making the extraction of the driven mechanical response complicated. To illustrate this, we consider the amplitude of the current $A_I = \alpha\sqrt{(X_z + C_gV_g^{ac}/C_g'V_g^{dc})^2 + Y_z^2}$, where we express the vibration displacement and the measured current as

$$z = A_z\cos(\omega t+\phi_z) = X_z\cos\omega t + Y_z\sin\omega t, \quad (1)$$

$$I = A_I\cos(\delta\omega t+\phi_I) = X_I\cos\delta\omega t + Y_I\sin\delta\omega t \quad (2)$$

and $\alpha = (\partial G/\partial V_g)V_s^{ac}V_g^{dc}C_g'/2C_g$ is a constant that depends on the transconductance $(\partial G/\partial V_g)$. In the limit where the displacement z is much smaller than $C_gV_g^{ac}/C_g'V_g^{dc}$, the response of A_I consists of a signal proportional to the quadrature X_z together with a large, frequency-independent background that has a pure electrical origin, see Fig. 1(b). In the opposite limit, the responses of A_I and A_z become proportional to each other [Fig. 1(c)]. This shows that while the shape of the mechanical response remains the same in these two limits, the measured shape of the spectral response of A_I is completely different. In practice, the measured response of A_I in the linear regime is usually compared with the predictions based on a Lorentzian line shape using several free parameters.[85] These can include the resonance frequency, the mechanical linewidth, the offset $C_gV_g^{ac}/C_g'V_g^{dc}$, the constant α, and the phase difference φ_e. This analysis usually becomes unreliable in the nonlinear regime, since the mechanical response can take a large range of different line shapes. Therefore, a method to directly extract the mechanical response without any fitting procedure is highly desirable.

We demonstrate a method to separate the signal of the mechanical vibrations from the signal of pure electrical origin. The idea is to put all the current signal of pure electrical origin in the quadrature X_I such that $X_I \propto X_z + C_gV_g^{ac}/C_g'V_g^{dc}$ and $Y_I \propto Y_z$. This is achieved by manipulating the data after the measurements with a rotation of the angle ϕ_I [Eq. (2)] in the plane (X_I, Y_I) (see supplementary material section, Sec. I). To illustrate this method, we proceed with the response of the two quadratures X_I and Y_I of the current directly acquired from the lock-in amplifier [Figs. 2(a) and 2(b)]. The two responses cannot be described by the usual functional forms of driven linear oscillators. We then compute the background offset of Y_I by incrementing the rotation phase ϕ_I by $\delta\phi_I$ from 0 to 2π [Fig. 2(c)]. When this background offset in Y_I is zero, all the current signal of pure electrical origin is in X_I and can be subtracted from the data. The resulting quadrature responses have now the familiar functional form of linear oscillators [Figs. 2(e) and 2(f)], and the spectral response of the displacement is well described by a Lorentzian [Fig. 3(a)].

We use the subtracted background current X_I^e of pure electrical origin to calibrate the displacement of the nanotube resonator in units of meters [Fig. 3(a)]. This background current is given by $X_I^e = (\partial G/\partial V_g)V_s^{ac}V_g^{ac}/2$; we verify that it depends linearly on V_g^{ac} [Fig. 2(d)]. The two quadratures then read

$$X_z = \frac{C_gV_g^{ac}}{C_g'V_g^{dc}}\frac{X_I - X_I^e}{X_I^e}, \quad Y_z = \frac{C_gV_g^{ac}}{C_g'V_g^{dc}}\frac{Y_I}{X_I^e}. \quad (3)$$

The calibration of the displacement is subject to the uncertainty in the estimation of C_g/C_g' (see below).

This method enables us to obtain the mechanical response in the nonlinear regime, see Fig. 3(c). While the response can be described by

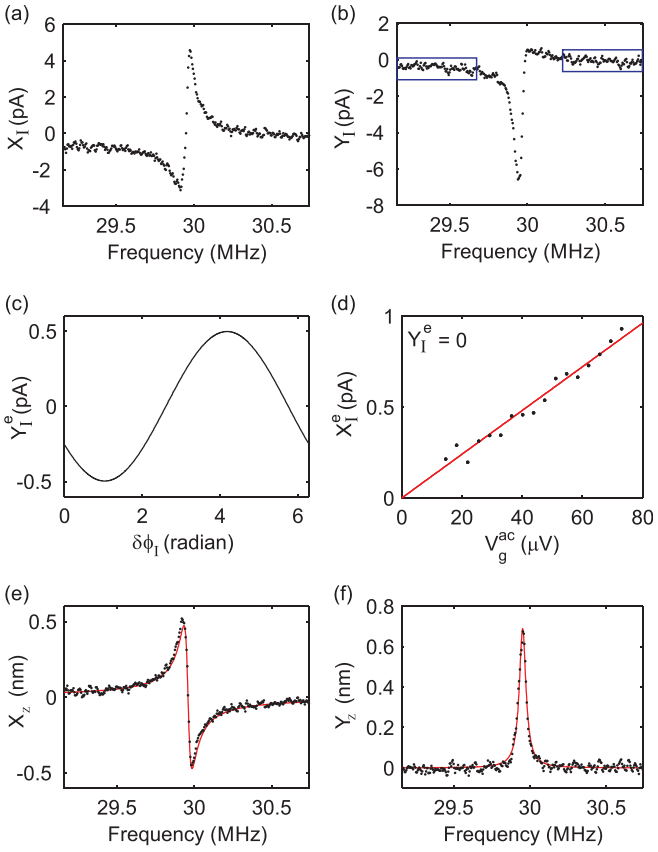

FIG. 2. (a) and (b) Spectral response of the current quadratures X_I and Y_I to the driven capacitive force. The two blue boxes indicate the Y_I values used to compute the background offset Y_I^e. (c) Estimated background offset Y_I^e from the data in a and b by incrementing the phase ϕ_I in Eq. (1) by $\delta\phi_I$. (d) Background current X_I^e with pure electrical origin with $\delta\phi_I$ set so that $Y_I^e = 0$. The red line is a linear fit of the data. (e) and (f) Spectral response of the displacement quadratures X_z and Y_z to the driven capacitive force after having subtracted X_I^e from X_I. The data are compared to the quadratures expected for a linear oscillator (red lines).

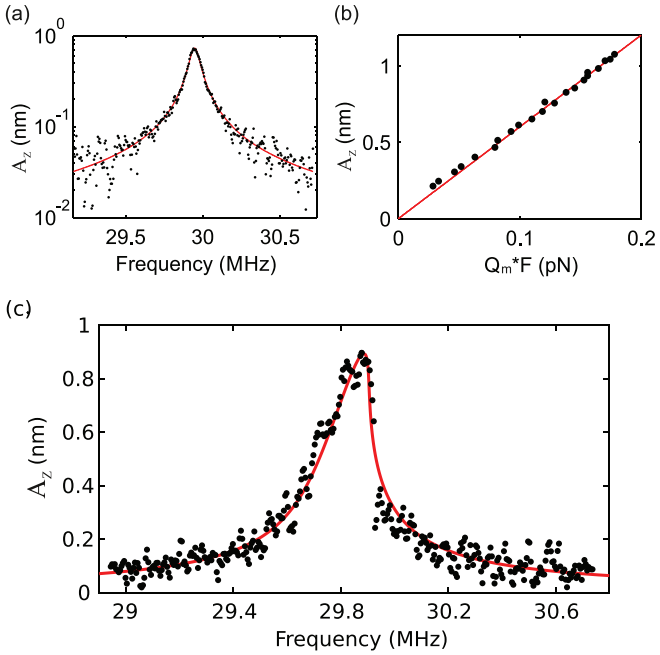

FIG. 3. (a) Spectral response of the displacement amplitude A_z to the driven capacitive force in the linear regime after having subtracted X_I^e from X_I. The data are compared to a Lorentzian peak (red line). (b) Force response of the displacement amplitude A_z at the mechanical resonance frequency. The red line is a linear fit of the data. The force is multiplied by the quality factor, since the latter varies when increasing the driving force.[35] (c) Nonlinear mechanical response of the displacement amplitude A_z to the driven capacitive force. The spectral response is extracted using the method described in Fig. 2. The red line is the fit to the usual Duffing equation.[86] The Duffing constant is much larger than that in panel (a) due to the coupling of vibrations and single-electron tunneling, see Ref. 87; concomitantly, the resonance frequency is lower.

the usual Duffing equation without nonlinear damping [red line in Fig. 3(c)], a detailed study of this device where vibrations are coupled to single-electron indicates that the nonlinear mechanical response to a driving force is significantly modified by thermal fluctuations (see Ref. 87). This results in a decreasing responsivity for an increasing drive and the absence of hysteresis when the driving frequency is swept back and forth.[87] When taking into account the effect of thermal fluctuations, the Duffing constant is 1.9×10^{31} m^{-2} s^{-2}. Note that while the thermal fluctuations modify the response of the driven vibrations, they are barely visible in the measurement shown in Fig. 3(c) as they are averaged out with the lock-in amplifier.

The current X_I^e of pure electrical origin also enables quantifying the mass of the mechanical mode in a way that is simple and reliable. In Fig. 3(b), we compute the force response of the displacement amplitude at resonance frequency ω_m in the linear regime using

$$A_z = \frac{C_g}{C_g'}\frac{V_g^{ac}}{V_g^{dc}}\frac{Y_I}{X_I^e}, \quad F = \beta C_g' V_g^{dc} V_g^{ac}, \tag{4}$$

where Y_I corresponds to the current amplitude at resonance frequency after having separated the signal of pure electrical origin. The constant β can be different from one for electron transport in the single-electron tunneling regime [Eq. (5) and supplementary material, Sec. II]. The mass m is determined from the slope of the force-displacement response using $A_z = (Q_m/m\omega_m^2)F$ with Q_m the quality factor. The slope depends on the current terms Y_I and X_I^e measured from the lock-in amplifier but is independent of V_g^{ac}, V_s^{ac}, and $\partial G/\partial V_g$ that enter the prefactor α in the current-displacement conversion and whose values could be somewhat altered by the amplification chain and the losses along the coaxial cables. We determine $m = 4.5 \pm 1.5$ ag from the mass measured at different V_g^{dc} values [Fig. 4(b)]. This value is consistent with the length of the suspended nanotube measured by scanning electron microscopy and assuming a 1.5 nm radius single-wall nanotube.

The uncertainty in the mass measurement and the displacement calibration comes from the uncertainty in the estimation of the nanotube-gate separation d and the mass fluctuations in Fig. 4(b). The separation $d = 150 \pm 20$ nm measured by atomic force microscopy

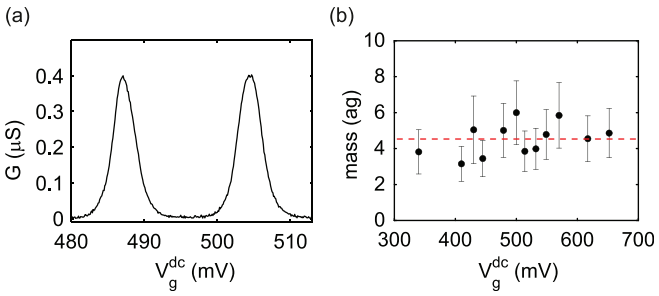

FIG. 4. (a) Electrical conductance of the nanotube device as a function of gate voltage. (b) Mass of the eigenmode measured at different gate voltage values. The red dashed line indicates the average mass of 4.5 ag.

enters in the estimation of $C_g' = C_g/d\ln(2d/r)$ in Eqs. (3) and (4) when considering the capacitance between a tube with radius r separated from a plate by the distance d. We estimate $C_g = e/\Delta V_g = 9.7$ aF from the separation ΔV_g in gate voltage between two conductance peaks associated with single-electron tunneling [Fig. 4(a)]. This capacitance is consistent with $C_g = 12.9$ aF obtained from the device geometry measured by scanning electron microscopy and atomic force microscopy. The fluctuations of m in Fig. 4(b) are partly due to the error in the estimation of the average charge occupation f, which varies between 0 and 1 when sweeping V_g^{dc} through the conductance peaks [Fig. 4(a)], since f enters in the prefactor β of the driven force in Eq. (4) as

$$\beta = 1 - \frac{C_g}{C_\Sigma} + f(1-f)\frac{C_g}{C_\Sigma}\frac{e^2/C_\Sigma}{k_B T} \tag{5}$$

in the incoherent single-electron tunneling regime. Here, C_Σ is the total capacitance of the single-electron transistor and varies gradually from 19.9 to 26.5 aF when sweeping V_g^{dc} over multiple conductance peaks. The fluctuations of m are also attributed to the slow increase in the contamination on the nanotube surface at 6 K; the three largest m values in Fig. 4(b) are obtained from force-displacement measurements carried out one month after the first measurements.

We now discuss the limits of our method to measure the mechanical response. The method is expected to be reliable for any device as long as the current contributions with mechanical, and pure electrical origins are both well resolved. This means that the signal of both contributions should be large compared to the electrical noise floor of the measurement circuit. This is the case in our measurements, see Figs. 2 and 3. The noise from the electrical circuit at 6 K is given by the Johnson–Nyquist noise of the 7.5 kΩ impedance of the RLC resonator.[6] Using the current noise $S_I = 210$ fA $/\sqrt{\text{Hz}}$ at 6 K, the conversion α between current and displacement, and the 1.5 Hz measurement bandwidth, we get a 22 pm standard deviation in the displacement noise, which is consistent with the noise observed in Fig. 3(a). The contribution to the measured signal from the thermal mechanical fluctuations is significantly smaller than that from the electrical noise. The vibrations can be driven to large amplitudes, but it is better to have displacements much smaller than the resonator-gate separation to avoid nonlinearity in the detection. In this way, the capacitance C_g and its derivative C_g' remain constant resulting in a linear relation between the measured quadratures $X_I - X_I^e$ and Y_I and the displacement quadratures X_z and Y_z in Eq. (3). The nonlinearity in the detection is related to $C_g/C_g' = (d - \delta z)\ln(2(d-\delta z)/r)$ in our device. For large vibrations, the nonlinearity in the detection has to be taken into account when estimating A_z. In Fig. 3(c), the nonlinearity in the detection modifies A_z by about 1% at the resonance.

In summary, we show how to measure the spectral mechanical response using electrical mixing measurements. Our method enables us to calibrate the displacement in meters. Another asset of this method is the determination of the mass of the measured mechanical eigenmode, which is a key parameter of the mechanical resonators when used in sensor applications. This work opens the possibility to quantitatively study nanoelectromechanical resonators in the nonlinear regime, where interesting mesoscopic phenomena emerge.[1] The driven nonlinear mechanical response can be described by a large range of different shapes depending on, for example, the strength of the thermal noise, the presence of frequency noise, the nonlinear resonant coupling to a high-frequency mode, and whether the restoring force is described by only the cubic nonlinear term in the displacement or also higher order terms. Our method enables us to determine such driven response spectra in an unambiguous way from electrical mixing measurements. Another important aspect of the method is the calibration of the displacement, which enables quantifying the strength of nonlinear forces by determining for instance the Duffing constant from the measurement of the resonance frequency shift as a function of displacement.[1]

See the supplementary material for theoretical description of the two-source mixing technique and the methodology used to derive the driving force in the single electron tunneling domain.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Chandan Samanta: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Methodology (equal); Writing – original draft (equal). **David Czaplewski:** Conceptualization (equal); Data curation (equal); Formal analysis (supporting); Funding acquisition (equal); Methodology (equal); Writing – original draft (equal). **Sergio Lucio**