

Supporting information for: Graphene Nanoelectromechanical Systems as Stochastic-Frequency Oscillators

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S0: Overview

In this Supporting Information section, we present derivations of the equations used in the main text as well as derivations of background material. Sections S1-S2 derive basic equations for the mechanics of the graphene sheet. Section S3 presents calculations of the magnitude of the expected signal when using the frequency modulated (FM) method for excitation and detection described in the main text.

The remainder of the supplement can be divided into sections concerning the linewidth in the linear damping regime (Sections S4-S5) and phenomena in the nonlinear regime (S6-S7). Specifically, section S4 first presents derivations of the non-dissipative line broadening due to stiffness fluctuations induced by the coupling between the fundamental and the rest of the thermally excited membrane modes. Section S4 then considers dissipative mechanisms. The energy loss rate from

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fast modes due to interactions with the other sheet modes is calculated using perturbation theory for the wave equation. The energy loss rate from the fundamental mode due to interactions with the other sheet modes is also considered, using a model of parametric driving of the fast modes. Section S5 considers how the fundamental mode quality factor is modified by electrostatic forces.

Finally, section S6 derives results for the membrane motion in the nonlinear regime, while S7 is concerned with the magnitude of the parametric drive by thermal expansion in the nonlinear regime.

S1: Resonant Frequency and Deflection of Graphene Membranes under Electrostatic Pressure

To calculate the resonance frequency of an initially tensioned graphene circular membrane with tension T_0 versus gate voltage V_g , the starting point is the elastic energy for a two dimensional (2D) membrane,^{1,2}

$$\mathcal{U} = \frac{1}{2} \kappa \int dx dy (\nabla^2 h)^2 + \frac{1}{2} \int dx dy 2\mu \sum_{i,j} u_{ij}^2 + \lambda \left(\sum_i u_{ii} \right)^2 - P \int dx dy h, \quad (\text{S1})$$

where μ is the shear modulus, λ is Lamé's first constant, h is the vertical height displacement of the membrane, P is the pressure on the membrane, κ is the bending modulus, and u_{ij} is the strain tensor. For graphene, $\lambda \approx 48$ N/m, $\mu \approx 144$ N/m, and $\kappa \sim 1$ eV.³ The strain tensor is given by²

$$u_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} + \frac{\partial h}{\partial x_i} \frac{\partial h}{\partial x_j} \right),$$

A vector in the x - y plane with components $u_i(x,y)$, and a z -component $h(x,y)$ represents the displacement of each point on the membrane from equilibrium. To calculate the deflection and resonance frequency of the fundamental mode, we neglect the bending term.³ The membrane is considered to be initially tensioned by stretching it by an isotropic in-plane strain factor $1 + c$ and then vertically displaced by the pressure. We take a variational approximation to the height to be $h = z_0(1 - r^2/r_0^2)$, where r_0 is the radius of the resonator, r is the cylindrical radius coordinate, and z_0 is a variational parameter equal to the vertical displacement of the center of the membrane. This

gives the strain tensor components

$$\begin{aligned}
u_{xx} &= 2x^2(z_0/r_0^2)^2 + c \\
u_{yy} &= 2y^2(z_0/r_0^2)^2 + c \\
u_{xy} &= 2xy(z_0/r_0^2)^2
\end{aligned} \tag{S2}$$

Putting eq. S2 into eq. S1 gives

$$\mathcal{U} = \frac{2}{3} \pi \frac{z_0^4}{r_0^2} (\lambda + 2\mu) + 2\pi c (\lambda + \mu) z_0^2 - \frac{1}{4} \frac{\epsilon_0 \bar{A}}{d^2} z_0 V_g^2 + 2c^2 (\lambda + \mu) \pi r_0^2 \tag{S3}$$

The term $2c^2(\lambda + \mu)\pi r_0^2$ is a constant arising from the work required to initially strain the membrane, and the electrostatic pressure was expressed as

$$P = \frac{1}{2} \frac{\epsilon_0}{d^2} V_g^2, \tag{S4}$$

where ϵ_0 is the dielectric constant and d is the distance between the gate and the ground plane. Here we neglect the contribution of the first order corrections to the pressure (calculated below) since they are negligible compared to the tension term for typical device parameters. Minimizing the energy with respect to z_0 gives the equation

$$\frac{8}{3} \pi (\lambda + 2\mu) \frac{\bar{z}_0^3}{r_0^2} + 4\pi c (\lambda + \mu) \bar{z}_0 - \frac{1}{4} \frac{\epsilon_0 \bar{A}}{d^2} V_g^2 = 0, \tag{S5}$$

where \bar{A} is the area of the resonator, and \bar{z}_0 is the optimizing value. For sufficiently small V_g we neglect the cubic terms and get

$$\bar{z}_0 = \frac{1}{4\pi c (\lambda + \mu)} \frac{\epsilon_0 \bar{A}}{4d^2} V_g^2. \tag{S6}$$

The factor $c(\lambda + \mu)$, which is a strain times an elastic constant, is related to the initial tension. As the constant term in the energy, $2c^2(\lambda + \mu)\pi r_0^2$ represents the work required to strain the membrane we use this to determine the tension by considering the work required for an infinitesimal

expansion. This yields $T_0 = 2c(\lambda + \mu)$. Thus equation S6 is then rewritten as

$$\bar{z}_0 = \frac{1}{8\pi T_0} \frac{\epsilon_0 \bar{A}}{d^2} V_g^2. \quad (\text{S7})$$

For large V_g the cubic term in equation S5 dominates, and we expect $\bar{z}_0 \sim V_g^{2/3}$.

S2: Effective spring constant, mass, and Duffing term

The resonance frequency ω_0 is given by $\omega_0 = \sqrt{k_{eff}/m_{eff}}$ with an effective resonator spring constant k_{eff} and mass m_{eff} . The effective spring constant is given by

$$k_{eff} = \frac{\partial^2 \mathcal{U}}{\partial z_0^2},$$

where \mathcal{U} is the total energy including electrostatic terms from the gate voltage. To account for electrostatic effects on ω_0 , it is required to approximate the electrostatic energy to 2nd order in z_0 to determine the force gradient. The electrostatic energy is $\frac{1}{2}CV^2$, where C is the resonator capacitance, and V is the voltage on the capacitor. Assuming slow variation of the displacement laterally, we approximate the capacitance as

$$C = \int \frac{\epsilon_0}{d - z_0(1 - r^2/r_0^2)} r dr d\theta$$

Expanding this in a power series in z_0 , the maximum deflection, so that $C = C_0 + C_1 + C_2 \dots$, where C_i is a term in the i th power of z_0 , gives

$$\begin{aligned} C_0 &= \frac{\epsilon_0 \bar{A}}{d} \\ C_1 &= \frac{\epsilon_0 \bar{A} z_0}{2d^2} \\ C_2 &= \frac{\epsilon_0 \bar{A}}{3d^3} z_0^2 \end{aligned}$$

The contribution to the energy from C_2 produces a force gradient and alters the effective spring

constant, yielding

$$k_{eff} = \frac{\partial^2 \mathcal{U}}{\partial z_0^2} = 8\pi(\lambda + 2\mu) \frac{z_0^2}{r_0^2} + 4\pi c(\lambda + \mu) - \frac{\varepsilon_0 \bar{A}}{3d^3} V_g^2 \quad (\text{S8})$$

Using the relation $T_0 = 2c(\lambda + \mu)$ derived above and eq. S7, eq. S8 can be rewritten as

$$\omega_{res} = 2\pi f_{res} = \sqrt{\frac{2\pi T_0 - \frac{\varepsilon_0 \pi r_0^2}{3d^3} V_g^2 + aV_g^4}{m_{eff}}}, \quad (\text{S9})$$

with a a constant.

Duffing term

The Duffing term is represented by a force term $F = -\alpha z_0^3$. Using eq. S3 and $\alpha = -\frac{1}{3!} \partial^3 F / \partial z_0^3 = \frac{1}{3!} \partial^4 \mathcal{U} / \partial z_0^4$, yields

$$\alpha = 8\pi(\lambda + 2\mu) / 3r_0^2. \quad (\text{S10})$$

Effective mass

Here we compute the effective mass and resonance frequency of the vibrating membrane using a variant of Rayleigh's method.⁴ For the vibrating graphene (or few layer) sheet, we have

$$z_0(t) = \bar{z}_0 + |\delta z| \sin \omega t,$$

where δz is the complex amplitude of the oscillation, and t is the time. The maximum kinetic energy when $z_0 = \bar{z}_0$ and the potential energy is minimal is then

$$KE = \int \frac{1}{2} \sigma \omega^2 |\delta z|^2 (1 - r^2/r_0^2)^2 r dr d\theta,$$

where σ is the resonator areal mass density. This gives

$$KE = \frac{1}{6} \pi \sigma |\delta z|^2 r_0^2 \omega^2.$$

Comparing this expression to the result for the ordinary harmonic oscillator, we get

$$m_{eff} = \frac{1}{3}\pi\sigma r_0^2. \quad (\text{S11})$$

For the resonance frequency when $V_g = 0$, $k_{eff} = 2\pi T_0$ and using the expression for m_{eff} , we get

$$\omega = \sqrt{\frac{2\pi T_0}{\frac{1}{3}\pi\sigma r_0^2}} = \frac{\sqrt{6}}{r_0} \sqrt{\frac{T_0}{\sigma}},$$

which is within a few percent of the exact result.⁵

S3: Magnitude of FM signal in drum resonator

The FM technique described in the text will produce a signal with geometric factors arising from the drumhead geometry as compared to a parallel plate model. The signal is expected to be smaller both because the average deflection is less due to the pinned boundary conditions and because the average potential is less because of the voltage drop along the sample. Moreover, upon the transduction of the motion into an electrical signal, because of the shape of the deflection, the resistance change will also be less.

This yields a total attenuation factor of 16. This can be understood as one factor of 1/2 in the drive force from the shape of the distortion not being planar, and another factor of 1/2 since the average potential over the sheet is 1/2 of the applied potential. The signal transduction has a relative factor 1/4 because the resistance change is only concentrated near the center where the deflection is largest, and it is probed by a potential near the center which is $\sim 1/2$ the potential applied. In the following we show the detailed computation of the expected total signal reduction from these sources.

From equation S1 the electrostatic energy term is given by

$$\mathcal{U}_{el} = - \int dx dy h(x,y) P(x,y),$$

where here the pressure P is no longer considered constant within the $x - y$ plane since the electro-

static potential V is now assumed to drop linearly between the electrodes. Supporting Information Fig. S1 shows a schematic diagram of the device geometry and potential drop. As before variational expression for the height is given by

$$h = z_0(1 - r^2/r_0^2). \quad (\text{S12})$$

The local pressure is given by

$$P = \frac{\epsilon_0}{2d^2}(V - V_g)^2,$$

with

$$V = V_{sd} \frac{L - x - r_0}{L},$$

where V_{sd} is the source-drain voltage, $x = 0, y = 0$ corresponds to the center of the circular drum-head and $L = 2r_0$. We then have

$$\mathcal{U}_{el} = - \int dx dy \frac{\epsilon_0}{2d^2} \left(-V_g + V_{sd} \frac{L - x - r_0}{L} \right)^2 z_0 [1 - (x^2 + y^2)/r_0^2].$$

If $V_{sd} \ll V_g$ then

$$\mathcal{U}_{el} \cong \mathcal{U}_{DC} + \int dx dy \frac{\epsilon_0}{d^2} V_g V_{sd} \frac{L - x - r_0}{L} z_0 [1 - (x^2 + y^2)/r_0^2] = \mathcal{U}_{DC} + \mathcal{U}_{AC},$$

where \mathcal{U}_{DC} is the electrostatic energy arising from the DC gate voltage V_g . Performing the integral gives

$$\mathcal{U}_{AC} = \frac{\epsilon_0 \bar{A}}{4d^2} V_g V_{sd} z_0,$$

Giving a force magnitude

$$F_{AC} = \frac{\epsilon_0 \bar{A}}{4d^2} V_g V_{sd} \quad (\text{S13})$$

As discussed above, compared to the usual expression for a parallel plate capacitance and a spa-

tially constant electrostatic potential,

$$\begin{aligned} F &= \frac{1}{2}C'V_g^2 \\ &= \frac{\epsilon_0\bar{A}}{d^2}V_gV_{sd}, \end{aligned}$$

the effective force is 1/4 as much.

Current change caused by a membrane displacement

The total measured FM signal depends on the change in current caused by the membrane deflection discussed above. The current change caused by the membrane displacement in our experiment is expected to be smaller than the parallel plate result. This is because under the actual parabolic displacement the center has the most change in conductance while the edge, which is pinned by the boundary, has zero. To determine the change in the current, we use the result that if an inhomogeneous sheet resistance exists then the 2D Laplace equation is modified to⁶

$$\nabla^2V = \frac{\nabla R \cdot \nabla V}{R}, \quad (\text{S14})$$

where R is the local sheet resistance and V the electrostatic potential within the sheet. To solve this, we use perturbation theory by assuming the right hand side is small. The correction to lowest order will be

$$\nabla^2V_{in} = \frac{\nabla R \cdot \nabla V_{hom}}{R}, \quad (\text{S15})$$

where V_{in} is the inhomogeneous potential produced by the forcing term on the right hand side of eq. S15, and V_{hom} is the unperturbed (homogeneous) potential. The total potential will be $V = V_{hom} + V_{in}$. Assuming small resistance changes, we replace the denominator of the right hand side of eq. S15 by the unperturbed value of R which we label R_0 . To get an expression for ∇R , we

start with an expression for R due to small changes in the charge density ρ .

$$\begin{aligned} R &= R_0 + \frac{\partial R}{\partial \rho} \delta \rho \\ \nabla R &= \frac{\partial R}{\partial \rho} \nabla(\delta \rho) \end{aligned} \quad (\text{S16})$$

using eq. S12 $\delta \rho$ can be expressed as

$$\delta \rho = \frac{\partial \rho}{\partial z_0} z_0 [1 - (x^2 + y^2)/r_0^2] \quad (\text{S17})$$

Eqs. S16 and S17 taken together yield

$$\nabla R = \frac{\partial R}{\partial \rho} \frac{\partial \rho}{\partial z_0} \frac{z_0}{r_0^2} (-2r\hat{r}), \quad (\text{S18})$$

where \hat{r} is the in-plane unit radial vector. The two derivatives in eq. S18 are considered constant.

We define $T_R = \partial R / \partial \rho$. We also have

$$\frac{\partial \rho}{\partial z_0} = \frac{\partial}{\partial z_0} (-C_A V_g) = -V_g \frac{\partial C_A}{\partial z_0} = -V_g \frac{\partial}{\partial z_0} \frac{\epsilon_0}{z_0} = -V_g \frac{\epsilon_0}{z_0^2},$$

where C_A is the capacitance per unit area. This yields

$$\nabla R = T_R \frac{V_g \epsilon_0}{d^2} \frac{z_0}{r_0^2} (2r\hat{r}),$$

where d is the distance between the gate and the undisplaced membrane, assuming that positive z_0 indicates downwards deflection towards the gate. The unperturbed potential gradient is given by

$$\nabla V_{hom} = -\frac{V_{sd}}{L} \hat{x}$$

so the right hand side of eq. S15 is given by

$$\begin{aligned}\frac{\nabla R \cdot \nabla V_{hom}}{R_0} &= \frac{T_R V_g \epsilon_0 z_0}{R_0 d^2 r_0^2} [2r\hat{r} \cdot (-\frac{V_{sd}}{L} \hat{x})] \\ &= \frac{-2T_R \epsilon_0 V_g V_{sd} z_0}{R_0 d^2 L} \frac{z_0}{r_0^2} x\end{aligned}$$

Taking the solution to the 2D Poisson equation

$$\nabla^2 V = \delta(\vec{x}),$$

where $\delta(\vec{x})$ is the Dirac delta function as

$$V = \frac{1}{4\pi} \ln(x^2 + y^2).$$

V_{in} is therefore given by

$$\begin{aligned}V_{in}(\vec{x}') &= - \int dx dy \frac{2T_R \epsilon_0 V_g V_{sd} z_0}{R_0 d^2 L} \frac{z_0}{r_0^2} x \frac{1}{4\pi} \ln |\vec{x}' - \vec{x}|^2. \\ &= A_0 \int dx dy x \ln |\vec{x}' - \vec{x}|^2,\end{aligned}$$

with

$$A_0 = -\frac{T_R \epsilon_0 V_g V_{sd} z_0}{2\pi R_0 d^2 L} \frac{z_0}{r_0^2}.$$

The integral is taken over the resonator area, a circle of radius r_0 . Converting this into a dimensionless integral over the unit circle by a change of coordinates and evaluating the integral numerically gives

$$\Delta V \cong \pi A_0 r_0^3.$$

Since the voltage and current are related by resistance R by $V = IR$,

$$\frac{\Delta V}{V} = \frac{\Delta R}{R} = -\frac{\Delta G}{G},$$

where $G = 1/R$ is the conductance. Then we have

$$\Delta G = -\frac{T_R V_g \epsilon_0 z_0}{4d^2 R_0} G = -\frac{T_R V_g \epsilon_0 z_0}{4d^2 R_0^2}.$$

Considering the vibration amplitude $|\delta z|$ this yields an expression for the ac current δI :

$$\delta I = -\frac{T_R V_g V_{sd} \epsilon_0 |\delta z|}{4d^2 R_0^2}.$$

We can rewrite this in terms of the transconductance $T = \partial G / \partial \rho$. We have

$$T = \frac{\partial G}{\partial \rho} = \frac{\partial(1/R)}{\partial \rho} = \frac{-1}{R^2} \frac{\partial R}{\partial \rho}.$$

Thus,

$$\delta I = \frac{TV_g V_{sd} \epsilon_0 |\delta z|}{4d^2}, \quad (\text{S19})$$

We can compare this to the parallel plate result

$$\begin{aligned} \delta I &= V_{sd} \Delta G \\ &= V_{sd} T V_g C' |\delta z| \\ &= \frac{V_{sd} T V_g \epsilon_0 |\delta z|}{d^2} \end{aligned}$$

Thus we have another 1/4 factor signal reduction because of the geometry. The total signal will be 1/16 of that expected from a parallel plate model for the resonator. With this factor of 1/4, the FM

signal is expected to be, using the parallel plate result from ref. 7

$$\delta I = \frac{1}{8} \frac{\partial G}{\partial V_g} V_{sd} V_g \frac{C'}{C} f_{\Delta} \frac{\partial}{\partial f} \Re[\delta z],$$

We have also the relation for a harmonic oscillator on resonance

$$\frac{d\Re[\delta z]}{df} = \frac{-2|\delta z|}{\delta f},$$

where δf is the frequency width of the resonance. Therefore the amplitude in terms of known experimental parameters is given by:

$$|\delta z| = \frac{4d\delta I\delta f}{(\partial G/\partial V_g)V_{sd}V_g f_{\Delta}} \quad (\text{S20})$$

S4: Dissipation and line broadening from intermodal interactions

Recent theoretical work has suggested that the linewidth in carbon nanotube⁸ or graphene⁹ resonators could result from fluctuations in the tension caused by the thermally excited modes. However, the work on carbon nanotubes considers tension fluctuations, which is not immediately applicable to two-dimensional membranes. It also does not consider the temporal behavior of the fluctuations, which is important for determining the contribution of motional narrowing to the linewidth. The work on graphene does not explicitly consider a tensioned membrane, which is relevant to our experimental situation. As a result, we first consider a model in which the modes of the sheet are divided conceptually into “fast” and “slow” modes by their frequency. As the fast modes oscillate due to thermal excitation, they perturb the slow modes’ resonance frequency through nonlinear coupling. Later in this section we consider dissipative line broadening due to intermodal energy transfer. This contribution to line broadening is found to be small compared to the frequency fluctuation broadening for our device geometry and typical parameters.

We now focus specifically in the fluctuations in the effective spring constant of the fundamental

mode. We take its equation of motion as that of a harmonic oscillator:

$$\frac{d^2z}{dt^2} + \omega_0^2[1 + x(t)]z = F_{ext}(t)/m, \quad (\text{S21})$$

where z is the displacement, t is the time, ω_0 is the characteristic frequency, $F_{ext}(t)$ is any external force applied to the oscillator, m the oscillator mass, and $x(t)$ is a random function with a mean of zero and autocorrelation function $R(\tau) = \langle x(t)x(t+\tau) \rangle$ which represents the frequency fluctuations $\delta\omega$ due to the fast modes. Since the frequency in the oscillator equation is a stochastic variable, the physical situation is reminiscent of that in nuclear magnetic resonance (NMR) in which a precessing spin undergoes Larmor frequency fluctuations due to fluctuating environmental magnetic fields.^{10,11} For this situation, this stochastic equation was successfully treated using the method of model coefficients in which the random variable is assumed to have a specific known behavior that produces an exact result for the mean solution of equation S21.^{10,11} (see also ref. 12 for review.)

Two limits exist with different behavior, determined by comparing the magnitude of the frequency fluctuations $\delta\omega$ with the inverse correlation time of the fluctuations ν . If $\nu \ll \delta\omega$ then the frequency fluctuations are sufficiently slow that the full intrinsic linewidth $\Gamma_i = \delta\omega$ results. On the other hand, if $\nu \gg \delta\omega$ the oscillator averages the fluctuations, resulting in a narrower linewidth Γ_{mn} than $\delta\omega$, which is the regime of motional narrowing in NMR.

The spectral density of the frequency fluctuations in x , $S(\omega)$, defined as

$$S(\omega) = \int_{-\infty}^{\infty} R(\tau)e^{i\omega\tau}d\tau,$$

at zero frequency can be written as $S(0) = \Delta t \langle x^2 \rangle = 4\Delta t \langle \delta\omega^2 \rangle / \omega_0^2$, where Δt is a characteristic correlation time for the fluctuations. Setting $\Delta t = 1/\nu$, the frequency width Γ_{mn} is given by¹⁰⁻¹²

$$\Gamma_{mn} = \frac{\omega_0^2}{2} S(0), \quad (\text{S22})$$

The linewidth is therefore

$$\Gamma = \min[\Gamma_{mn}, \Gamma_i] \quad (\text{S23})$$

To compute Γ_i and Γ_{mn} , we begin by Fourier transforming the energy given in eq. S1, by using the Fourier transform relations

$$\begin{aligned} h(\vec{q}) &= \int h(\vec{x}) e^{-i\vec{q}\cdot\vec{x}} d^2\vec{x} \\ h(\vec{x}) &= \int h(\vec{q}) e^{i\vec{q}\cdot\vec{x}} \frac{d^2\vec{q}}{(2\pi)^2} \end{aligned}$$

which yields

$$\begin{aligned} \mathcal{U} &= \int \frac{d^2\vec{q}}{(2\pi)^2} h^*(\vec{q}) h(\vec{q}) \left[c(\lambda + \mu)q^2 + \frac{1}{2}\kappa q^4 \right] \\ &+ \int \frac{d^2\vec{q}_1 d^2\vec{q}_2 d^2\vec{q}_3 d^2\vec{q}_4}{(2\pi)^6} h^*(\vec{q}_1) h^*(\vec{q}_2) h(\vec{q}_3) h(\vec{q}_4) u(\vec{q}_1, \vec{q}_2, \vec{q}_3, \vec{q}_4) \\ &\times \delta(\vec{q}_4 + \vec{q}_3 - \vec{q}_2 - \vec{q}_1) \end{aligned} \quad (\text{S24})$$

where $u(\vec{q}_1, \vec{q}_2, \vec{q}_3, \vec{q}_4)$ is given by

$$u(\vec{q}_1, \vec{q}_2, \vec{q}_3, \vec{q}_4) = \frac{1}{16} \lambda [(\vec{q}_1 \cdot \vec{q}_3)(\vec{q}_2 \cdot \vec{q}_4) + (\vec{q}_1 \cdot \vec{q}_4)(\vec{q}_2 \cdot \vec{q}_3)] + \frac{1}{4} \mu (\vec{q}_1 \cdot \vec{q}_2)(\vec{q}_3 \cdot \vec{q}_4)$$

If u were zero then \mathcal{U} would be that of noninteracting quadratic normal modes. The u function thus describes the nonlinear modal interactions to 4th order in h . For a given configuration of $h(q)$ values the shift in the effective spring constant of the mode with wavevector \vec{q} is determined by adding a cosine wave to the sheet and collecting quadratic terms in its amplitude A_m . The height Fourier components then become

$$h_m(\vec{q}) = h(\vec{q}) + \frac{(2\pi)^2 A_m}{2} [\delta(\vec{q} - \vec{q}) + \delta(\vec{q} + \vec{q})]$$

Then

$$\begin{aligned}
\delta\mathcal{U} &= \int \frac{d\vec{q}_1 d\vec{q}_2 d\vec{q}_3 d\vec{q}_4}{(2\pi)^6} \left(h^*(\vec{q}_1) + \frac{(2\pi)^2 A_m}{2} [\delta(\vec{q}_1 - \vec{q}) + \delta(\vec{q}_1 + \vec{q})] \right) \\
&\times \left(h^*(\vec{q}_2) + \frac{(2\pi)^2 A_m}{2} [\delta(\vec{q}_2 - \vec{q}) + \delta(\vec{q}_2 + \vec{q})] \right) \\
&\times \left(h(\vec{q}_3) + \frac{(2\pi)^2 A_m}{2} [\delta(\vec{q}_3 - \vec{q}) + \delta(\vec{q}_3 + \vec{q})] \right) \\
&\times \left(h(\vec{q}_4) + \frac{(2\pi)^2 A_m}{2} [\delta(\vec{q}_4 - \vec{q}) + \delta(\vec{q}_4 + \vec{q})] \right) \\
&\times u(\vec{q}_1, \vec{q}_2, \vec{q}_3, \vec{q}_4) \delta(\vec{q}_1 + \vec{q}_2 - \vec{q}_3 - \vec{q}_4)
\end{aligned} \tag{S25}$$

Collecting the terms quadratic in A_m , the shift is related to integrals over the mean squares of the modal displacements. In thermal equilibrium, the mean square modal displacements undergo thermal fluctuations, broadening the frequency response. The contribution of a given fast mode with frequency ω' to $S(0)$, labeled $S_{\omega'}(0)$ is

$$S_{\omega'}(0) = \frac{4}{\omega_0^2} \langle \delta\omega^2 \rangle_{\omega'} \Delta t_{\omega'},$$

where $\langle \delta\omega^2 \rangle_{\omega'}$ is the mean square fluctuation in the slow mode with frequency ω_0 caused by the fast mode with frequency ω' , and $\Delta t_{\omega'}$ is the characteristic time scale of those fluctuations. Since the bandwidth of a harmonic oscillator with characteristic frequency ω' and quality factor $Q_{\omega'}$ is $\omega'/Q_{\omega'}$ this gives $\Delta t_{\omega'} \approx Q_{\omega'}/\omega'$. We then have

$$S(0) = \int S_{\omega'}(0) \rho(\omega') d\omega', \tag{S26}$$

where $\rho(\omega')$ is the density of modes at frequency ω' .

The quantity $\langle \delta\omega^2 \rangle_{\omega'}$ is computed by first computing the total $\langle \delta\omega^2 \rangle$ by squaring equation S25 and thermally averaging it with respect to the quadratic part of \mathcal{U} , using the relation that $\frac{1}{2} \delta k_{eff}/k_{eff} = \delta\omega/\omega$, where k_{eff} is the slow mode effective spring constant. To perform the

thermal average, we use the relation that

$$\begin{aligned} \langle h^*(\vec{q}_4)h^*(\vec{q}_3)h(\vec{q}_2)h(\vec{q}_1) \rangle_{0>} &= \frac{1}{4} \frac{(2\pi)^2 \delta(\vec{q}_4 - \vec{q}_2)}{\beta(A\vec{q}_4^2 + B\vec{q}_4^4)} \frac{(2\pi)^2 \delta(\vec{q}_3 - \vec{q}_1)}{\beta(A\vec{q}_3^2 + B\vec{q}_3^4)} + \frac{1}{4} \frac{(2\pi)^2 \delta(\vec{q}_3 - \vec{q}_2)}{\beta(A\vec{q}_3^2 + B\vec{q}_3^4)} \frac{(2\pi)^2 \delta(\vec{q}_4 - \vec{q}_1)}{\beta(A\vec{q}_4^2 + B\vec{q}_4^4)}, \\ &+ \frac{1}{4} \frac{(2\pi)^2 \delta(\vec{q}_4 + \vec{q}_3)}{\beta(A\vec{q}_4^2 + B\vec{q}_4^4)} \frac{(2\pi)^2 \delta(\vec{q}_2 + \vec{q}_1)}{\beta(A\vec{q}_2^2 + B\vec{q}_2^4)} \end{aligned} \quad (\text{S27})$$

where $A = c(\lambda + \mu)$ and $B = \frac{1}{2}\kappa$ are elastic constants that define the quadratic part of \mathcal{U} , and $\beta = 1/k_B T$, with k_B the Boltzmann constant and T the temperature. (See for example, ref. 13.) The factor of 1/4 and the extra term compared to the work in ref. 13 arise because the height is real, so $h(\vec{q}) = h^*(-\vec{q})$. The result is that the total mean square frequency fluctuations are given by

$$\langle \delta\omega^2 \rangle = \frac{27\bar{A}\bar{q}^4 \omega_0^2 E}{512\beta^2 k_{eff}^2} \int \frac{dq}{2\pi} \frac{q^5}{(Aq^2 + Bq^4)^2} = \frac{27\bar{A}\bar{q}^4 \omega_0^2 E}{2048\pi\beta^2 k_{eff}^2 AB}, \quad (\text{S28})$$

where $E = N^2(\lambda^2 + 4\lambda\mu + 4\mu^2)$ is an elastic constant and N is the number of layers. Converting this to an integral over frequency using the density of modes obtained from the frequency relation $\omega = \sqrt{2(Aq^2 + Bq^4)}/\sigma$ gives,

$$\langle \delta\omega^2 \rangle = \frac{27\bar{q}^4 \omega_0^2 E}{512\beta^2 k_{eff}^2} \int d\omega' \left(\frac{-A + \sqrt{A^2 + 2\sigma\omega'^2 B}}{B} \right)^2 \frac{1}{\sigma^2 \omega'^4} \rho(\omega')$$

From this we find that $S_{\omega'}(0)$ is given by

$$S_{\omega'}(0) = \frac{27\bar{q}^4 E}{128\beta^2 k_{eff}^2} \left(\frac{-A + \sqrt{A^2 + 2\sigma\omega'^2 B}}{B} \right)^2 \frac{Q_{\omega'}}{\sigma^2 \omega'^5} \quad (\text{S29})$$

Fast mode behavior

Completing the calculation requires finding the $Q_{\omega'}$ in eq. S29. Similar to the fundamental mode, the linewidth and $Q_{\omega'}$ of the fast modes also have two potential sources, frequency fluctuations and energy damping. However, frequency fluctuations are ineffective at causing the fast mode amplitude fluctuations that produce the frequency shifts in the fundamental mode. These amplitudes fluctuate on a timescale determined by the modal energy damping rate. Therefore a de-

termination of the energy damping rate for the fast modes is necessary to find the appropriate $Q_{\omega'}$. A variety of energy relaxation mechanisms are possible including coupling to electrons, clamping losses, or to in-plane phonons which are expected to produce relatively high quality factors $\sim 10^5$ or higher for geometries similar to our experiment.^{14–16} Here, we also consider a mechanism of energy transfer from one vibrational mode to another. The tension fluctuations within the sheet give rise to spatially inhomogeneous wave velocity fluctuations. Such fluctuations also scatter the standing waves of the sheet in a manner similar to Rayleigh scattering. However, because the fluctuations are time dependent, scattering among modes with different frequencies is expected to occur. We compute the energy damping rate of the fast modes due to such scattering using perturbation theory for the wave equation. The Lagrange density for the membrane is

$$\mathcal{L} = \frac{1}{2\sigma} \left(\frac{\partial h}{\partial t} \right)^2 - A(\nabla h)^2 - \frac{1}{8}(\lambda + 2\mu)(\nabla h)^4 - B(\nabla^2 h)^2.$$

Neglecting bending for modes that are longer wavelength than the cutoff $\sqrt{A/B}$ the equation of motion is

$$\sigma \frac{\partial^2 h}{\partial t^2} - A\nabla^2 h - \left\{ \frac{1}{4}(\lambda + 2\mu) [\nabla(\nabla h)^2 \cdot \nabla h + (\nabla h)^2 \nabla^2 h] \right\} = 0$$

We consider the term in braces as a perturbation. The unperturbed problem has time-periodic solutions satisfying the eigenvalue equation

$$\nabla^2 h + k_l^2 h = 0,$$

where k_l^2 is the l th eigenvalue for each solution. The perturbation scatters a given mode into other modes, giving it a finite lifetime. We denote the instantaneous state of the membrane as a superposition of eigenmodes $\Psi_l = 1/\sqrt{A} e^{i\vec{k}_l \cdot \vec{x}}$, where l is the mode index.

$$h(t) = \sum_l \Psi_l e^{-i\omega_l t} c_l(t),$$

where $c_l(t)$ are coefficients for which we want to obtain a differential equation. Substituting the

expression for $h(t)$ into the equation of motion, we get

$$\sum_l \Psi_l e^{-i\omega_l t} \left\{ -2i\omega_l \dot{c}_l(t) + \ddot{c}_l(t) + \frac{1}{4\sigma} (\lambda + 2\mu) \left[2(\vec{k}_l \cdot \nabla h)^2 + (\nabla h)^2 k_l^2 \right] \right\} = 0$$

Taking the inner product denoted by

$$\langle f, g \rangle = \int d^2\vec{x} f^*(\vec{x}) g(\vec{x})$$

of this equation with $\Psi_m e^{i\omega_m t}$ yields

$$-2i\omega_m \dot{c}_m(t) + \ddot{c}_m(t) = \sum_l \frac{1}{4\sigma} (\lambda + 2\mu) \langle \Psi_m, [2(\vec{k}_l \cdot \nabla h)^2 + (\nabla h)^2 k_l^2] \Psi_l \rangle e^{i\omega_m t} c_l(t)$$

where $\omega_{lm} = \omega_m - \omega_l$. Assuming only one of the modes has non-negligible amplitude so that $c_l(t) \approx 0 \ll c_n(t)$ except for one particular mode n , the equation becomes

$$-2i\omega_m \dot{c}_m(t) + \ddot{c}_m(t) = \frac{1}{4\sigma} (\lambda + 2\mu) \langle \Psi_m, [2(\vec{k}_n \cdot \nabla h)^2 + (\nabla h)^2 k_n^2] \Psi_n \rangle e^{i\omega_{nm} t} c_n(t)$$

For large times, growing amplitude solutions for c_m occur when the right hand side of the above includes zero frequency components. In this case the \ddot{c}_m term yields a fast oscillating homogeneous solution and no time-averaged energy transfer so long as that the perturbation is sufficiently weak that $c_n(t)$ varies slowly on the time scale of $1/\omega_m$. We will see below that this condition is satisfied to first order. Neglecting this term, which makes a negligible contribution to the energy transfer, the equation then becomes essentially the same as time-dependent perturbation theory in quantum mechanics. The lowest order solution is

$$c_m = \frac{i}{2\omega_m} \int_0^t \frac{c_n(0)}{4\sigma} (\lambda + 2\mu) \left[\langle \Psi_m, (\nabla h)^2 k_n^2 \Psi_n \rangle + \langle \Psi_m, 2(\vec{k}_n \cdot \nabla h)^2 \Psi_n \rangle \right] e^{i\omega_{nm} t'} dt' \quad (\text{S30})$$

The first matrix element term in the brackets results from velocity fluctuations. The thermal expectation value of the second term is the same as the first. Therefore we replace the entire integrand

with twice the first term. Factoring out the k_n the matrix element is therefore

$$V_{mn} = \langle \Psi_m, (\nabla h)^2 \Psi_n \rangle = \frac{1}{A} \int d^2\vec{x} (\nabla h)^2 e^{i(\vec{k}_n - \vec{k}_m) \cdot \vec{x}} \quad (\text{S31})$$

Thus the matrix element is the Fourier transform of $(\nabla h)^2$ at the scattering wave vector $\Delta\vec{k} = \vec{k}_n - \vec{k}_m$. This should depend on the squared height modulations with wavevector $\Delta\vec{k}$, which have a characteristic frequency $\omega(\Delta\vec{k})$.

The energy transfer rate is determined from eq. S30.

$$\frac{1}{E_n} \frac{dE_m}{dt} = \frac{d}{dt} \frac{k_{eff,m} |c_m(t)|^2}{k_{eff,n} |c_n(0)|^2} = \frac{k_{eff,m} k_n^4 (\lambda + 2\mu)^2}{k_{eff,n} 64\omega_m^2} \int_0^t V_{mn}(t) V_{mn}^*(t) e^{i\omega_{mn}\tau} d\tau + c.c.$$

Taking the thermal average gives

$$\left\langle \frac{1}{E_n} \frac{dE_m}{dt} \right\rangle = \frac{k_{eff,m} k_n^4 (\lambda + 2\mu)^2}{k_{eff,n} 64\omega_m^2} \int_0^t \langle V_{mn}(t) V_{mn}^*(t) \rangle e^{i\omega_{mn}\tau} d\tau + c.c.$$

for large t , this becomes

$$\left\langle \frac{1}{E_n} \frac{dE_m}{dt} \right\rangle = \frac{k_{eff,m} k_n^4 (\lambda + 2\mu)^2}{k_{eff,n} 32\omega_m^2} \Re[S_{V_{mn}}(\omega_{mn})], \quad (\text{S32})$$

where $S_{V_{mn}}$ is the power spectral density of V_{mn} at frequency ω_{mn} . This should be peaked at frequency $\omega(\Delta\vec{k})$ with width $\sim \omega(\Delta\vec{k})$, while the area under the peak vs. frequency is the thermal average of $|V_{mn}|^2$. We therefore expect an analog of Fermi's golden rule to apply, i.e.

$$\begin{aligned} \left\langle \frac{1}{E_n} \frac{dE_m}{dt} \right\rangle &= \frac{k_{eff,m} k_n^4 (\lambda + 2\mu)^2}{k_{eff,n} 32\omega_m^2} \langle |V_{mn}|^2 \rangle \delta[\omega(q_n) - \omega(q_m) \pm \omega(q_n - q_m)] \\ &= \frac{k_n^2 (\lambda + 2\mu)^2}{32\sigma^2 u^2} \langle |V_{mn}|^2 \rangle \delta[\omega(q_n) - \omega(q_m) \pm \omega(q_n - q_m)], \end{aligned} \quad (\text{S33})$$

where $k_{eff,n} \approx \bar{A}Aq_n^2$, u is the wave velocity and

$$\omega(q) = \sqrt{\frac{2(Aq^2 + Bq^4)}{\sigma}},$$

where the final result for the total normalized energy loss rate is obtained by an integration over the final states. Evaluating eq. S33 numerically for typical parameters yields $Q_f \sim 10^4$ at room temperature and $\sim 10^6$ at low temperature, scaling as $1/T^2$. Note that using a broader frequency function than a δ -function to account for the finite frequency width of $S_{V_{mn}}$ leads to a larger predicted Q , thus the above estimates constitute a lower bound on Q from this mechanism. Having found Q_f , we must now compare Γ_{mn} to Γ_i to determine whether motional narrowing is expected to be important. To first evaluate Γ_i we use eq. S28 using $k_{eff} \approx \bar{A}Aq^2$, yielding

$$Q_i = \frac{\xi T_0^{3/2} r_0}{k_B T} \sqrt{\frac{\kappa}{E}} \quad (\text{S34})$$

where $\xi \approx 6.84$ is a numerical factor, $E = N^2(\lambda^2 + 4\lambda\mu + 4\mu^2) \approx N^2 \times 1.1 \times 10^5 \text{ (N/m)}^2$ with N the number of layers, $\kappa \approx 10^{-16} \text{ Nm}$ is the bending modulus, T_0 the tension, k_B is the Boltzmann constant, and r_0 is the resonator radius. Evaluation of eq. S26 using $Q_{\omega'} = Q_i \sim 100 - 1000$, obtained from eq. S34 using typical parameters gives $\Gamma_i \sim \Gamma_{mn}$. Since we have instead $Q_{\omega'} = Q_f \gg Q_i$, the fluctuations are sufficiently slow so that we find that $\Gamma_i \ll \Gamma_{mn}$ and thus the static limit is the relevant one, since other energy damping mechanisms¹⁴⁻¹⁶ such as clamping loss mentioned above are also expected to produce significantly larger Q values than Q_i . Therefore we expect the effects of motional narrowing to be minimal and the measured quality factor should follow the relation given for Q_i , eq. S34, as observed.

Energy Transfer to fast modes from fundamental mode

In the steady state, vibrations of the fundamental mode produce a periodic modulation of the membrane tension with frequency ω_0 and amplitude $\delta\omega$. Each fast mode, approximated as an independent oscillator with frequency ω_f , then has its frequency modulated at frequency ω_0 . The equation of motion for each fast mode can then be written as¹⁷

$$\ddot{z} + \Gamma\dot{z} + \omega_f^2[1 + \varepsilon \cos(\omega_0 t)]z = \sqrt{\Gamma D}\xi(t)$$

where $D = k_B T / m_{eff}$ is a diffusion constant and $\sqrt{\Gamma D}\xi(t)$ is a delta-correlated stochastic function that models the Langevin force. In the steady state the root mean square amplitude of the oscillator is given by (assuming $Q \gg 1$ and neglecting terms of order higher than ε^2 ($\varepsilon = 2\delta\omega_f/\omega_f$ or terms with frequency $2\omega_0$ or higher))

$$\sigma_{zz} = \frac{D}{\omega_f^2} + \frac{3D}{2\omega_f^4}\delta\omega^2 - 2\delta\omega\frac{D}{\omega_f} \left[\frac{1}{2\omega_f^2}\cos(\omega_0 t) + \frac{\Gamma}{4\omega_0\omega_f^2}\sin(\omega_0 t) \right]. \quad (\text{S35})$$

Here we neglect frequency broadening, since we are in the static limit where $v \ll \delta\omega$, and the moments of the solutions $\sigma_{zz}(\omega_f)$ corresponding to the equations with different ω_f in the ensemble would be expected to be very similar since $\omega_0 \ll \omega_f$. When the parametric drive is zero, the modal amplitude thermal fluctuations are given by $\sigma_{zz} = D/\omega_f^2$. For finite parametric drive caused by the vibrations in the fundamental mode, the relative frequency modulations $\delta\omega_f/\omega_f = \frac{1}{2}\delta T_0/T_0$ and are therefore the same for each fast mode. The sine and cosine terms thus represent a coherent response of all the fast modes to the vibration of the fundamental mode. Thus the fundamental mode vibration modulates the sheet tension at the same frequency ω_0 . Since the membrane is under electrostatic pressure, the changing tension displaces the equilibrium point of the fundamental mode, which corresponds to a force $z_0\delta k_{eff}$. The term proportional to the sine therefore yields a frictional damping force on the fundamental mode that is proportional to its velocity.

In addition, the second term of eq. S35 shows that in steady state each fast mode has excess energy above the equipartition energy, $E_{excess} = \frac{3D}{4\omega_f^4}\delta\omega_f^2 k_{fast}$, where k_{fast} is the stiffness of the fast mode. Each fast mode oscillator dissipates this excess energy at an average rate $\frac{3D}{4\omega_f^4}\delta\omega_f^2\Gamma k_{fast}$, which balances the input power due to the parametric drive. The total energy loss per unit time from all the fast modes is given by

$$\left\langle \frac{dE}{dt} \right\rangle = \frac{\delta\omega_0^2}{\omega_0^2} \frac{3\bar{A}^2 A}{4m\beta} \int_0^{q_{max}} \frac{dq}{2\pi} \frac{q^3}{Q\omega(q)} \quad (\text{S36})$$

where $q_{max} = \sqrt{A/B}$. The damping force F_d due to the coherent tension modulations is given by (setting $\langle h^*(\vec{q}_1)h(\vec{q}_2) \rangle = \frac{1}{2}\bar{A}\sigma_{zz}(2\pi)^2\delta(\vec{q}_1 - \vec{q}_2)$ and taking the expectation value of the eq. S25 to find the mean stiffness modulation)

$$F_d = \dot{z} \frac{\tilde{E}\bar{q}^2\bar{A}^2\delta\omega_0}{2\omega_0^3 m\beta} \int_0^{q_{max}} \frac{dq}{2\pi} \frac{q^3}{Q\omega(q)}, \quad (\text{S37})$$

where \tilde{E} is an elastic constant on the order of the Young's modulus. It can be verified that the damping force given by eq. S37 leads to essentially the same energy loss from the excited slow resonator mode as the steady state loss from the fast modes given by eq. S36 when the tension is dominated by the stretching from the displacement. Thus, within this picture, energy flows from the slow modes to the fast modes via coherent tension oscillations in the sheet. Energy relaxation can also occur to the wider environment through loss mechanisms such as clamping loss or radiation of in-plane phonons.

For typical device parameters and at room temperature and below this friction force produces a frequency broadening less than Γ_i . We thus expect the frequency broadening to dominate the linewidth. This is consistent with the observed $\propto T$ linewidth dependence.

Relation between steady-state dissipation and the friction force

In steady state, we assume that the motion is effectively ergodic and the time and ensemble averages of quantities related to the motion are equal. The oscillator susceptibility determines the steady state response of the fundamental mode. The imaginary part determines the mean rate of energy dissipation in the fundamental mode due to energy transfer to the environment. If the frictional damping force is represented by $m\Gamma_d\dot{z}$, then the power dissipated is $P = \frac{1}{2}m\omega_0^2\Gamma_d\langle\delta z^2\rangle$. On the other hand the imaginary part of the susceptibility including frequency fluctuations on resonance is

$$\chi''(\omega) = \frac{1}{\omega_0\Gamma},$$

with Γ determined by the fluctuation broadening, eq. S34, assuming that $\Gamma_d \ll \Gamma$. The dissipated

power is then $P = F^2 Q / (m\omega_0)$, where F is the magnitude of the sinusoidal force drive. Equating these, we find the mean square amplitude of the motion is

$$\langle \delta z^2 \rangle = \frac{F^2}{\Gamma \Gamma_d m^2 \omega_0^2}. \quad (\text{S38})$$

Note that since $\Gamma_d < \Gamma$ this is in general larger than $\langle \delta z^2 \rangle$ would be if Γ originated from a frictional force. Nevertheless, the amplitude response measured at the drive frequency, e.g. by a lock-in amplifier will be identical to that determined by $\chi(\omega)$, just as if Γ represented a frictional force. This is because unlike the case of purely frictional force damping, the amplitude response power spectrum is spread out in frequency $\sim \delta\omega$ because of the fluctuating stiffness, while a lock-in measures the amplitude only in a narrow bandwidth around the excitation frequency.

S5: Electrostatic forces effect on quality factor

The electric field up to second order in the height is given by,¹⁸

$$\begin{aligned} E(\vec{r}) = & \left[-\frac{V}{d} + \frac{V}{d} \int \frac{d^2 \vec{q}}{(2\pi)^2} \frac{\cosh(qz)}{\sinh(qd)} qh(q) e^{i\vec{q}\cdot\vec{x}} \right. \\ & - \frac{V}{d} \int \frac{d^2 \vec{q}_1}{(2\pi)^2} \frac{d^2 \vec{q}_2}{(2\pi)^2} \frac{\cosh(q_2 d)}{\sinh(q_2 d)} \frac{\cosh(q_1 z)}{\sinh(q_1 d)} q_1 q_2 h(\vec{q}_2) h(\vec{q}_1 - \vec{q}_2) e^{i\vec{q}_1 \cdot \vec{x}} \Big] \hat{z} \\ & - \frac{V}{d} \int \frac{d^2 \vec{q}}{(2\pi)^2} \frac{\sinh(qz)}{\sinh(qd)} qh(\vec{q}) e^{i\vec{q}\cdot\vec{x}} (i\hat{q}) \\ & + \frac{V}{d} \int \frac{d^2 \vec{q}_1}{(2\pi)^2} \frac{d^2 \vec{q}_2}{(2\pi)^2} \frac{\cosh(q_2 d)}{\sinh(q_2 d)} \frac{\sinh(q_1 z)}{\sinh(q_1 d)} q_2 h(\vec{q}_2) h(\vec{q}_1 - \vec{q}_2) e^{i\vec{q}_1 \cdot \vec{x}} (i\vec{q}_1) \end{aligned} \quad (\text{S39})$$

where $\vec{r} = (\vec{x}, z)$ is the three dimensional position vector for in-plane position $\vec{x} = (x, y)$. Using the expression of the electric field the capacitance correction can be computed from the surface charge density $\sigma_s = \epsilon_0 \vec{E} \cdot \hat{n}$ with the unit normal to the graphene membrane $\hat{n} = (\nabla h - \hat{z}) / [1 + (\nabla h)^2]^{1/2}$. The capacitance C is given by $C = Q/V = \int \sigma_s d\vec{A} / V$. Including this electrostatic term modifies \mathcal{U} . Keeping terms up to 2nd order yields additional terms $\Delta \mathcal{U}$ (The 4th order electrostatic term is

negligible compared to the mechanical one under typical physical conditions in our experiment).

$$\Delta\mathcal{U} = \int \frac{d^2\vec{q}}{(2\pi)^2} h^*(\vec{q})h(\vec{q}) \left(-\frac{\varepsilon_0 V_g^2}{2d^2} \frac{\cosh qd}{\sinh qd} q - \frac{\varepsilon_0 V_g^2}{4d} q^2 \right)$$

The addition of these terms modifies eq. S28 to

$$\langle \delta\omega^2 \rangle = \frac{27}{512} \frac{\bar{A}\bar{q}^4 \omega_0^2 E}{\beta^2 k_{eff}^2} \int \frac{dq}{2\pi} \frac{q^5}{\left[-\frac{\varepsilon_0 V_g^2}{2d^2} \frac{\cosh qd}{\sinh qd} q + \left(A - \frac{\varepsilon_0 V_g^2}{4d} \right) q^2 + Bq^4 \right]^2}, \quad (\text{S40})$$

This can be approximated by

$$\langle \delta\omega^2 \rangle = \frac{27}{512} \frac{\bar{A}\bar{q}^4 \omega_0^2 E}{\beta^2 k_{eff}^2} \int \frac{dq}{2\pi} \frac{q^5}{(C + Dq + Aq^2 + Bq^4)^2}, \quad (\text{S41})$$

with $A = c(V_g)(\lambda + \mu) - \frac{\varepsilon_0 V_g^2}{4d}$, $B = \frac{1}{2}\kappa$, $C = -\frac{\varepsilon_0 V_g^2}{2d^3}$, $D = -\frac{\varepsilon_0 V_g^2}{2d^2}$. Equation S41 is used to compute Q by numerical integration and plotted for appropriate parameters in the main text Fig. 3b.

S6: Nonlinear resonator dynamics

The equation of motion for a nonlinear oscillator can be expressed as

$$m \frac{d^2 \bar{z}}{d\bar{t}^2} + \Gamma \frac{d\bar{z}}{d\bar{t}} + m\omega_0^2 \bar{z} + \bar{\chi} \bar{z}^2 + \bar{\mu}_0 \bar{z} \frac{d\bar{z}}{d\bar{t}} + \bar{\alpha} \bar{z}^3 + \bar{\eta} \bar{z}^2 \frac{d\bar{z}}{d\bar{t}} = \bar{G}_0 \cos(\bar{\omega} \bar{t} + \phi). \quad (\text{S42})$$

To switch to units such that $\alpha = m = \omega_0 = 1$ the dimensionless length variable is taken to be $z = \bar{z} \sqrt{\bar{\alpha}/m\omega_0^2}$, and the parameters of the oscillator equation of motion can be expressed in dimensionless form as follows¹⁹

$$\varepsilon = Q^{-1} = \frac{\Gamma}{m\omega_0}; \eta = \frac{\bar{\eta}\omega_0}{\bar{\alpha}}; G_0 = \frac{\bar{G}_0}{\omega_0^3} \sqrt{\frac{\bar{\alpha}}{m^3}}; \chi = \frac{\bar{\chi}}{\omega_0 \sqrt{m\bar{\alpha}}}; \mu_0 = \frac{\bar{\mu}_0}{\sqrt{m\bar{\alpha}}}; \omega = \frac{\bar{\omega}}{\omega_0}.$$

Here we assume that Q is determined from frictional forces alone, and ignore frequency broadening. We take this as a good approximation when the resonator is driven into the regime where

nonlinear damping is important. The equation of motion can then be scaled to yield

$$\ddot{z} + \varepsilon \dot{z} + z + \chi z^2 + \mu_0 z \dot{z} + z^3 + \eta z^2 \dot{z} + [h_0 \varepsilon \cos(2\omega t + 2\phi)]z = \varepsilon^{3/2} g_0 \cos(\omega t + \phi) \quad (\text{S43})$$

The parameter ε which is the inverse quality factor is considered a small parameter that enables approximate solutions that are expanded in ε . Motivated by our experimental results, in eq. S43 we have also included a parametric drive parametrized by h_0 that modulates the resonance frequency by $\delta\omega_0$ at a frequency 2ω as well as force drive parametrized by g_0 such that $G_0 = \varepsilon^{3/2} g_0$. The quantity $h_0 \varepsilon = 2\delta\omega_0/\omega_0$ gives the fractional amplitude of the frequency modulation relative to the undriven characteristic frequency ω_0 . To solve this equation, following the original method used by Duffing, (See for example ref. 20) we first assume to zero order the time dependence for the oscillator displacement with amplitude parameter Γ_0 and frequency ω given by

$$z_0 = \Gamma_0 \varepsilon^{1/2} \sin \omega t. \quad (\text{S44})$$

Here we reference the phases of the force and parametric drives to the response, which is taken to be a sine function. For a linear oscillator, the $\phi = 0$ on resonance (the drive being proportional to a cosine when the response is a sine). Note that in this definition of the phase ϕ occurs in the drive term rather than the response. This differs from the conventional treatment of the harmonic oscillator but simplifies the calculations below. The equation of motion S43 is rearranged to yield an expression for the acceleration:

$$\ddot{z} = -\varepsilon \dot{z} - z - \chi z^2 - \mu_0 z \dot{z} - z^3 - \eta z^2 \dot{z} - [h_0 \varepsilon \cos(2\omega t + 2\phi)]z + \varepsilon^{3/2} g_0 \cos(\omega t + \phi) \quad (\text{S45})$$

the expression for z_0 is substituted into the right hand side of the above equation, and integrated twice to yield a solution for z_1 . which produces the following result:

$$\begin{aligned}
z_1 = & -\frac{1}{4}\Gamma_0^2\chi\epsilon t^2 + \frac{\Gamma_0^3\epsilon^{(\frac{3}{2})}\eta\cos(\omega t)}{4\omega} \\
& -\frac{\Gamma_0^3\epsilon^{(\frac{3}{2})}\eta\cos(3\omega t)}{36\omega} + \frac{\Gamma_0^2\epsilon\mu_0\sin(2\omega t)}{8\omega} \\
& +\frac{3\Gamma_0^3\epsilon^{(\frac{3}{2})}\sin(\omega t)}{4\omega^2} - \frac{\Gamma_0^3\epsilon^{(\frac{3}{2})}\sin(3\omega t)}{36\omega^2} \\
& -\frac{\Gamma_0^2\chi\epsilon\cos(2\omega t)}{8\omega^2} - \frac{\Gamma_0\epsilon^{(\frac{3}{2})}h_0\sin(\omega t + 2\phi)}{2\omega^2} + \frac{\Gamma_0\epsilon^{(\frac{3}{2})}h_0\sin(3\omega t + 2\phi)}{18\omega^2} \\
& +\frac{\Gamma_0\epsilon^{(\frac{3}{2})}\cos(\omega t)}{\omega} - \frac{\epsilon^{(\frac{3}{2})}g_0\cos(\omega t + \phi)}{\omega^2} + \frac{\Gamma_0\sqrt{\epsilon}\sin(\omega t)}{\omega^2}
\end{aligned} \tag{S46}$$

Equating the coefficients of $\sin(\omega t)$ and $\cos(\omega t)$ between z_0 and z_1 produces a set of equations that can then be solved for Γ_0 and ϕ . The resulting solution is approximate, and when substituted into the original equation of motion produces non-zero error terms. However, these error terms will be at other harmonics besides those at frequency ω and of order ϵ or smaller. Thus the obtained values for Γ_0 and ϕ will be correct to lowest order in ϵ . In the present case, there are two additional issues to the expression given for z_0 in eq. S44. One is that it generates a term quadratic in t of order ϵ in z_1 . This can be eliminated by adding an ϵ -order constant to z_0 and setting it so the t^2 term vanishes. The other issue is that z_1 contains terms of order ϵ and frequency 2ω . If one were to proceed to higher order, the quadratic terms in the equation of motion would generate additional terms of order $\epsilon^{3/2}$ which are at the same order as the terms with frequency ω . To avoid this, z_0 must be corrected to add these terms so that z_1 is correct to order $\epsilon^{3/2}$ with only one iteration. The necessary expression is then:

$$z_0 = \Gamma_0\sqrt{\epsilon}\sin(\omega t) + \Lambda_1\epsilon\sin(2\omega t) + \Lambda_2\epsilon\cos(2\omega t) + \Lambda_3\epsilon,$$

with Λ_1 , Λ_2 and Λ_3 constants to be determined. After substituting z_0 into the right hand side of eq. S45 and matching the appropriate coefficients we get the following coupled cubic equations for Γ_0

and ϕ with effective cubic parameters η and α :

$$\begin{aligned} \left(2\Omega + \frac{1}{2}h_0 \cos 2\phi - \frac{3}{4}\alpha\Gamma_0^2\right) \Gamma_0 - g_0 \sin \phi &= 0 \\ \frac{1}{4}\Gamma_0^3\eta - \frac{1}{2}\Gamma_0 h_0 \sin 2\phi + \Gamma_0 - g_0 \cos \phi &= 0 \end{aligned} \quad (\text{S47})$$

The frequency width in the FM technique is determined by the frequency spacing between the stationary points of the real part of the response.⁷ The real part is given by the in-phase response to the drive, which we find based on eq. S43 to yield $\Re[z] = -\Gamma_0 \sin \phi$. Thus we want to find the stationary points of $-\Gamma_0 \sin \phi$ subject to the constraints given by the coupled cubic equations in eq. S47. Thus we use the method of Lagrange multipliers, which requires finding the stationary points of the function

$$\begin{aligned} f &= -\Gamma_0 \sin \phi + \lambda_1 \left[\left(2\Omega + \frac{1}{2}h_0 \cos 2\phi - \frac{3}{4}\alpha\Gamma_0^2\right) \Gamma_0 - g_0 \sin \phi \right] \\ &\quad + \lambda_2 \left(\frac{1}{4}\Gamma_0^3\eta - \frac{1}{2}\Gamma_0 h_0 \sin 2\phi + \Gamma_0 - g_0 \cos \phi \right), \end{aligned}$$

introducing auxiliary Lagrange multiplier variables λ_1 , and λ_2 that multiply the two constraint equations. Optimizing this gives 3 equations, which together with the original constraints eq. S47 yield 5 equations and 5 unknowns for the variables $\Omega, \phi, \Gamma_0, \lambda_1$, and λ_2 .

$$\begin{aligned} \frac{1}{4}(3\Gamma_0^2\eta - 2h_0 \sin 2\phi + 4)\lambda_2 - \frac{1}{4}[9\Gamma_0^2\alpha - 2h_0 \cos 2\phi - 8\Omega]\lambda_1 - \sin \phi &= 0 \\ -(\Gamma_0 h_0 \cos 2\phi - g_0 \sin \phi)\lambda_2 - (\Gamma_0 h_0 \sin 2\phi + g_0 \cos \phi)\lambda_1 - \Gamma_0 \cos \phi &= 0 \\ 2\Gamma_0\lambda_1 &= 0 \\ \left(2\Omega + \frac{1}{2}h_0 \cos 2\phi - \frac{3}{4}\alpha\Gamma_0^2\right) \Gamma_0 - g_0 \sin \phi &= 0 \\ \frac{1}{4}\Gamma_0^3\eta - \frac{1}{2}\Gamma_0 h_0 \sin 2\phi + \Gamma_0 - g_0 \cos \phi &= 0 \end{aligned} \quad (\text{S48})$$

From the third equation down in the above equation, we see that unless Γ_0 is zero then $\lambda_1 = 0$. The

system of equations can then be simplified to

$$\frac{1}{4}(3\Gamma_0^2\eta - 2h_0 \sin 2\phi + 4)\lambda_2 - \sin \phi = 0 \quad (\text{S49})$$

$$-(\Gamma_0 h_0 \cos 2\phi - g_0 \sin \phi)\lambda_2 - \Gamma_0 \cos \phi = 0 \quad (\text{S50})$$

$$\left(2\Omega + \frac{1}{2}h_0 \cos 2\phi - \frac{3}{4}\alpha\Gamma_0^2\right)\Gamma_0 - g_0 \sin \phi = 0 \quad (\text{S51})$$

$$\frac{1}{4}\Gamma_0^3\eta - \frac{1}{2}\Gamma_0 h_0 \sin 2\phi + \Gamma_0 - g_0 \cos \phi = 0 \quad (\text{S52})$$

We first seek an asymptotic solution to the system of equations S49-S52 valid for large g_0 and h_0 .

Dividing eq. S52 through by $\Gamma_0 h_0$ gives

$$\frac{\Gamma_0^2\eta}{4h_0} - \frac{1}{2}\sin 2\phi + \frac{1}{h_0} - \frac{g_0 \cos \phi}{\Gamma_0 h_0} = 0 \quad (\text{S53})$$

If we assume the parametric drive comes from heating as discussed in the main text, then we expect the parametric drive h_0 will be related to the force drive by $h_0 = bg_0^2$, where b is a constant.

Therefore in the limit of large g_0 , keeping the dominant term in Γ_0 eq. S53 reduces to

$$\frac{\Gamma_0^2\eta}{h_0} = 2 \sin 2\phi$$

Eq. S50 can then be solved for λ_2 :

$$\lambda_2 = -\frac{\Gamma_0 \cos(\phi)}{\Gamma_0 h_0 \cos(2\phi) - g_0 \sin(\phi)}$$

and substituted in to eq. S49, which gives

$$\frac{1}{4} \frac{(2\Gamma_0^2\eta + 4)\Gamma_0 \cos \phi}{\Gamma_0 h_0 \cos 2\phi - g_0 \sin \phi} + \sin \phi = 0$$

Neglecting the 4 compared to $2\Gamma_0^2\eta$, as appropriate for the large amplitude limit, and dividing the

top and bottom of the fraction by h_0 gives:

$$\frac{\frac{1}{4} \frac{(2\Gamma_0^2 \eta)}{h_0} \Gamma_0 \cos \phi}{\Gamma_0 \cos 2\phi - (g_0/h_0) \sin \phi} + \sin \phi = 0 \quad (\text{S54})$$

In the limit of large h_0 this reduces to

$$\frac{1}{4} (4 \sin 2\phi) \frac{\cos \phi}{\cos 2\phi} + \sin \phi = 0$$

which gives

$$\sin 2\phi \cos \phi + \cos 2\phi \sin \phi = 0$$

$$\sin 3\phi = 0$$

$$\phi = 0, \pm\pi/3, \pm2\pi/3 \dots$$

The values $\phi = 0, \pi$ are not considered since in conjunction with eq. S54 these values imply that $\Gamma_0 = 0$. Since the *low-amplitude* resonance has $\phi = 0$, we expect the flanking minima will have $\phi = \pm\pi/3$. These values for the phase can then be substituted into eq. S52. For $\phi = \pi/3$, we get the cubic equation

$$\Gamma_0^3 + \left(\frac{4 - \sqrt{3}h_0}{\eta} \right) \Gamma_0 - \frac{2g_0}{\eta} = 0$$

For large h_0 , it can be shown that there are three real roots, and the one that matches continuously to the single real root for small h_0 is given approximately by

$$\Gamma_0 \approx \sqrt{\frac{\sqrt{3}h_0 - 4}{\eta}}$$

This value for Γ_0 can then be substituted back into eq. S51 along with ϕ to determine Ω corre-

sponding to a stationary value of the phase. Solving eq. S51 for Ω yields

$$\Omega = \frac{3}{8}\Gamma_0^2\alpha - \frac{1}{4}h_0\cos(2\phi) + \frac{g_0\sin(\phi)}{2\Gamma_0}$$

This gives for large h_0 the frequency of the upper frequency minimum in the FM signal, Ω_+ :

$$\begin{aligned}\Omega_+ &= \frac{1}{8}h_0 + \frac{3\alpha}{8\eta}(\sqrt{3}h_0 - 4) + \frac{3^{1/4}}{4}\sqrt{\frac{\eta}{b}} \\ &= \left(\frac{1}{8} + \frac{3\sqrt{3}\alpha}{8\eta}\right)bg_0^2 + \text{const.}\end{aligned}$$

for the case $\phi = -\pi/3$ the equation for Γ_0 is given by

$$\Gamma_0^3 + \left(\frac{4 + \sqrt{3}h_0}{\eta}\right)\Gamma_0 - \frac{2g_0}{\eta} = 0$$

It can be shown that this equation has one real root,

$$\Gamma_0 = \frac{2\sqrt{3}g_0}{3h_0}$$

When substituted into eq. S51 along with the value for ϕ , we find for large h_0 the frequency of the lower frequency minimum Ω_- :

$$\Omega_- = -\frac{1}{4}h_0 = -\frac{1}{4}bg_0^2$$

This gives in the large drive asymptotic limit

$$\Delta\Omega = \Omega_+ - \Omega_- = \frac{3}{8}\left(1 + \frac{\sqrt{3}\alpha}{\eta}\right)bg_0^2 + \frac{3^{1/4}}{4}\sqrt{\frac{\eta}{b}} - \frac{3\alpha}{2\eta}. \quad (\text{S55})$$

This is quadratic in the drive as observed experimentally. Going back to physical units gives

$$\delta f = \frac{3}{8}\left(1 + \frac{\sqrt{3}\alpha}{2\pi\eta f_0}\right)bF_{AC}^2 f_0 + \text{const.}, \quad (\text{S56})$$

where F_{AC} is the ac electrostatic force on resonator as in the main text.

To obtain an approximate solution valid for any drive strength, numerical solutions of the systems of equations S49-S52 are used to show that the two oscillator phases ϕ_+ and ϕ_- corresponding to the two minima in the FM signal can be serviceably approximated by the mathematical functions

$$\begin{aligned}\phi_+ &= \pi/4 + \pi/12 \tanh(bg_0^2) \\ \phi_- &= -\pi/4 - \pi/12 \tanh(bg_0^2)\end{aligned}$$

These phases can be substituted into eq. S52 and the resulting cubic equation for Γ_0 solved for each phase. When these values for Γ_0 are substituted back into eq. S51 the resulting equation can be solved for Ω . The difference between the two Ω solutions corresponding to ϕ_+ and ϕ_- gives the frequency width function. The solution can be expressed in physical units and then fit to the data.

S7: Estimate for b and expression for α/η

Using the Fourier heat law, the temperature rise ΔT at the center of the graphene membrane assuming thermal equilibration to the ambient substrate temperature at the boundary is

$$\Delta T = \frac{P_0 r^2}{4\kappa_{2D}\bar{A}},$$

where \bar{A} is the membrane area, P_0 is the power dissipated, r is the radius, and κ_{2D} is the two-dimensional thermal sheet conductivity. Taking $P_0 = V_{sd}^2/R$, where R is the device resistance then

$$\Delta T = \frac{V_{sd}^2}{4\pi R \kappa_{2D}}.$$

If a_T is the (negative valued) thermal expansion coefficient, then the change in strain δc is given by

$$\delta c \approx \frac{V_{sd}^2 |a_T|}{4\pi R \kappa_{2D}},$$

which changes the membrane tension T_0 by an amount δT_0 given by

$$\delta T_0 = \frac{V_{sd}^2 |a_T| (\lambda + \mu)}{2\pi R \kappa_{2D}}$$

The parametric drive H is given by $2\delta f_{res}/f_{res}$, where δf_{res} is the modulation of the resonance frequency f_{res} . In terms of the tension, we have $\delta f_{res}/f_{res} = \frac{1}{2}\delta T_0/T_0$. Thus,

$$H = \frac{V_{sd}^2 |a_T| (\lambda + \mu)}{2\pi R \kappa_{2D} T_0}$$

Using the result for the force drive from eq. S13, we find

$$H = \left(\frac{4d^2}{\varepsilon_0 \bar{A} V_g} \right)^2 G_0^2 \frac{|a_T| (\lambda + \mu)}{2\pi R \kappa_{2D} T_0},$$

and therefore the constant b is given by

$$b = \left(\frac{4d^2}{\varepsilon_0 \bar{A} V_g} \right)^2 \frac{|a_T| (\lambda + \mu)}{2\pi R \kappa_{2D} T_0} \quad (\text{S57})$$

Expression for α/η

The ratio α/η can be determined from the data at small drive, from the equations S49-S52 setting $h = 0$, and considering g to be small. The phase at the minima of the FM signal is taken to be $\approx \pm\pi/4$. Using these approximations, the system of equations can be solved to yield in dimensionless units

$$\Delta\Omega = 1 + \frac{3}{4}\eta\Gamma_0^2$$

Using also the expression for the frequency shift with drive in terms of α ^{21,22}

$$f_{res} = f_0 + \frac{3\alpha|\delta z|^2}{32\pi^2 f_0 m} \quad (\text{S58})$$

where f_0 is the linear response resonance frequency, and setting $\Delta f = f_{res} - f_0$ and $\delta f = \varepsilon f_0 \Delta\Omega$

the linewidth we find

$$\frac{\alpha}{\eta} = 4\pi \frac{d\Delta f}{d\delta f} f_0, \quad (\text{S59})$$

which gives a way of measuring the ratio α/η as discussed in the main text.

Supporting Information Figure S1: Schematic diagram of device geometry

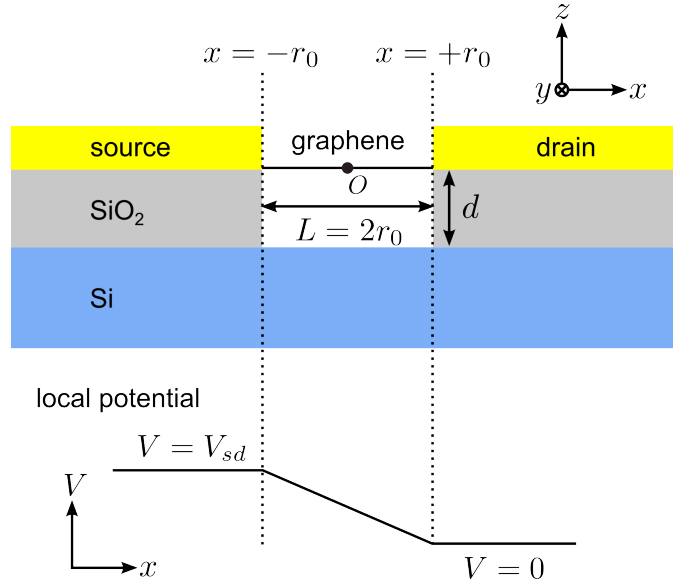


Figure S1: Schematic diagram of device geometry on an oxidized Si wafer, with a source, drain, and Si gate. The coordinate directions are indicated as well as the origin by O at the center of the resonator. The diameter of the drum membrane is L , and the radius is r_0 . The spacing from the substrate is d . The voltage drop profile along the sample is also shown below the device diagram.

Supporting Information S2: Room temperature variation of frequency and Q with gate voltage

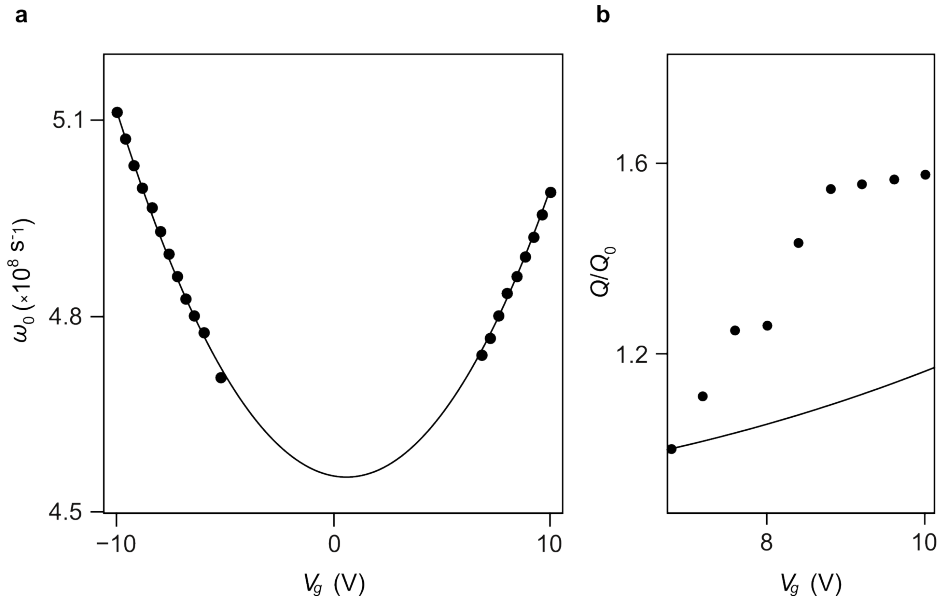


Figure S2: (a) Plot of the frequency vs. gate voltage measured at room temperature for the device discussed in the main text taken with $V_{sd} = 70$ mV. The solid line is a fit to a 2nd order polynomial. Since the dependence of the frequency on V_g is relatively strong, we assume that the frequency is dominated by the tension and neglect electrostatic effects. The tension is then related to the frequency by $\omega = \sqrt{2\pi T_0/m}$. (b) The measured Q vs. V_g is shown as the black circles. We then use the method discussed in section S5 to compute the expected Q , which is plotted as the solid line. Since Q was not easily extrapolated back to zero frequency, the data and theory were matched at the lowest measured V_g .

Supporting Information S3: Nonlinear parameter analysis for temperature $T = 57$ K.

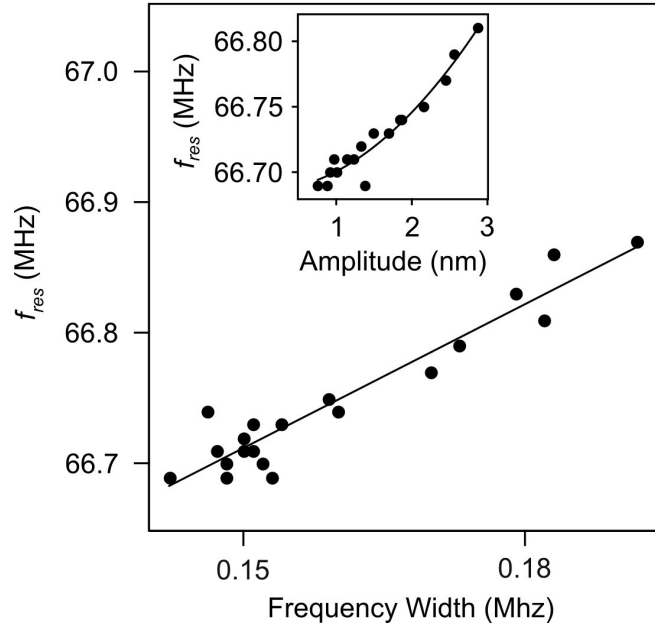


Figure S3: Nonlinear oscillator dynamics of the lowest-frequency mode. Main panel: resonance frequency vs. frequency width at $T = 57$ K and $V_g = 18$ V plotted as black circles. Solid curve is a straight line fit to the data. Inset: resonance frequency vs. amplitude at $T = 57$ K and $V_g = 18$ V plotted as black circles. The amplitude is obtained from the FM lineshape when it is still approximately Lorentzian (eq. S20). Solid curve is a fit to the theoretical expectation (eq. S58). The data analysis yields $\eta = 8 \times 10^4$ Ns/m³ and $\alpha = 2.4 \times 10^{14}$ N/m³. The lower value for α than at $T = 16$ K may indicate the presence of quadratic displacement terms as in eq. S43.

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