Deviation from the Normal Mode Expansion in a Coupled Graphene-Nanomechanical System

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A significant deviation from the normal mode expansion is observed in the optomechanically measured thermal noise of a graphene membrane suspended on a silicon nitride nanoresonator. This deviation is due to the heterogeneous character of mechanical dissipation over the spatial extent of coupled eigenmodes, which is tuned through an avoided anticrossing. We demonstrate that the fluctuation-dissipation theorem permits a proper evaluation of the thermal noise of the coupled nanomechanical system. Since a good spatial homogeneity is delicate to ensure at the nanoscale, this approach is fundamental to correctly describing the thermal noise of nanomechanical systems which ultimately impact their sensing capacity.

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I. INTRODUCTION

Nanomechanical oscillators are routinely used in fundamental and applied physics [1,2] as ultrasensitive force or mass sensors due to their increased sensitivity to their environment. The understanding of dissipation at the nanoscale is the key ingredient towards extreme sensitivity operation. Among others, carbon-based nanoresonators and alternative 2D materials [3] have revolutionized the field of nanomechanics [4–14] by pushing the oscillator dimensions down to a single atomic layer. The extreme sensitivities achieved are ultimately limited by the thermal noise of the nanoresonators, which underlines the importance of correctly understanding and describing their Brownian motion. The thermal noise of a vibrating nanomechanical system is commonly described using the normal mode expansion, which assumes that each eigenmode is driven by an independent fluctuating Langevin force, presenting no correlation with other eigenmodes. However, this intuitive description only holds when the mechanical dissipation is homogeneously distributed in the system [15–17]. Otherwise, inhomogeneous damping can create dissipative coupling between eigenmodes, leading to a violation of their assumed independence. Such deviations, which have been reported on macroscopic devices [18,19], are expected to be extremely important in nanomechanical systems since it becomes increasingly difficult to ensure and even measure a good spatial homogeneity over the entire nanosystem as its size is decreased. However, no deviations from the normal mode expansion have been observed at the nanoscale to date, despite the large variety of nanoresonators investigated.

In this article, we report on the deviation from the normal mode expansion in the optomechanically measured thermal noise of a nanomechanical arrangement made of a suspended graphene monolayer coupled to a silicon nitride nanomembrane which supports the graphene resonator. To fully explore the deviation from the normal mode expansion, we exploit the inertial coupling between both nanoresonators: upon temperature-controlled tunable hybridization, the coupled eigenmodes become spatially delocalized on the two subsystems whose intrinsic mechanical damping rates differ by 2 orders of magnitude. In this situation with strong coupling between the two nanoresonators, the damping homogeneity is, therefore, no longer maintained, which results in a pronounced deviation from the normal mode expansion that we report on and analyze. Then we measure the local mechanical susceptibility of the coupled nanomechanical system and prove that the fluctuation-dissipation theorem still holds across the entire observed anticrossing—that is, for both homogeneously and heterogeneously distributed mechanical damping.

These considerations are essential for correctly describing nanomechanical systems affected by inhomogeneous damping and point out the importance of having access to the local mechanical susceptibility to correctly estimate the thermal noise of complex nanomechanical systems.

II. SAMPLE PREPARATION

Our nanomechanical system is a fully suspended single-layer graphene sheet deposited on a square window opened in a Si_3N_4 nanoresonator, itself supported on an opened silicon wafer (see Fig. 1), which allows a dual optical access from both sides. It is obtained [see the Supplemental Material (SM) [20]] by transfer in the liquid phase of a monolayer, polycrystalline graphene grown by CVD on Cu [21,22] and suspended over up to $25 \times 25\ \mu m^2$ on a prepatterned stoichiometric Si_3N_4 membrane which is 500 nm thick and 100 μm wide.

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The used substrate is a silicon wafer coated on both sides with plasma-sputtered low-stress Si_3N_4 thin films of 500 nm thickness. Large windows of about 100×150 μm are defined using laser lithography on the wafer back side, while smaller square windows (4–25 μm sized) are defined on the top surface, centered above the bottom windows. The silicon nitride is etched away by reactive-ion etching in a SF_6 plasma. In order to permit optical transmission through the sample, and thus optical access from both sides, the silicon is subsequently etched away through the windows opened in the silicon nitride layers in a KOH solution. This etching away results in suspended silicon nitride membranes with predefined holes, as shown in Fig. 1 and the SM [20]. The graphene is grown on a copper foil by chemical vapor deposition from a methane precursor to form a continuous layer [21]. The monolayer graphene is covered by a spin-coated resist support layer (PMMA) and the copper is etched away in an ammonium persulfate solution. The graphene is then transferred onto the pre-patterned substrate. After removal of the PMMA layer in acetone and rinsing in isopropanol, the sample is dried in a supercritical carbon dioxide dryer to protect it from strong surface-tension forces that can be exerted by drying liquid droplets. The quality of the suspended graphene and the low levels of doping and residual strain are verified and estimated [23] using confocal micro-Raman mapping; see the SM [20].

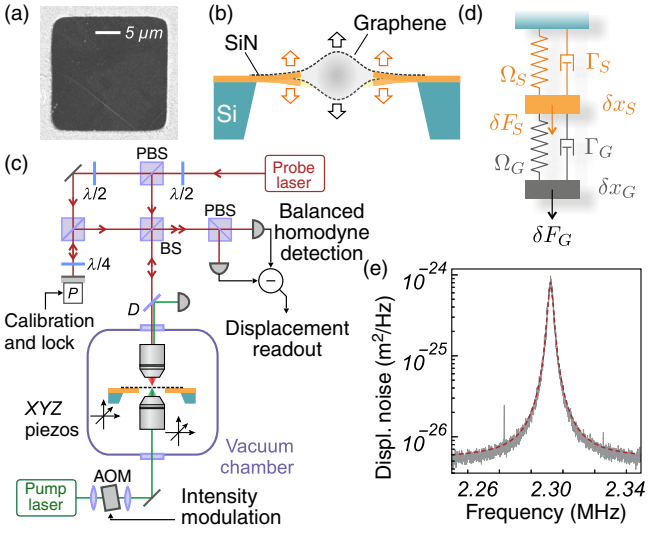

FIG. 1. The experimental setup. (a) SEM of a 20×20 μm^2 suspended CVD-grown graphene monolayer supported on a 300-nm-thick SiN nanomembrane, as sketched in (b). (c) Experimental setup. A balanced homodyne detection measures the phase fluctuations of the probe-laser field reflected by the sample and monitors its position fluctuations. A second counterpropagating pump-laser beam can be intensity modulated to optomechanically drive the coupled nanoresonators. The experiment is performed at pressures below 10^{-3} mbar. (d) Model describing the inertially coupled nanoresonators. (e) Thermal noise of a graphene membrane. The sharp peaks on each side are weakly coupled SiN eigenmodes.

III. THE EXPERIMENTAL SETUP

A 633-nm probe laser is focused on the graphene resonator with a high numerical-aperture objective (whose optical waist approximately equals 400 nm). The weak reflected beam constitutes the signal arm of a balanced homodyne detection [24] [see Fig. 1(c) and the SM [20]]. The sample is mounted vertically on an *XYZ* piezoelectric stage ($100 \times 100 \times 100$ μm scan range) permitting a precise positioning with respect to the probe-laser beam, which is fixed in space in order to ensure a stable operation of the interferometer. The long working distances (4 mm) and large numerical apertures (0.75) of the microscope objectives employed permit us to focus the laser beams down to optical waists of approximately 330 nm with half focusing angles approximately equal to 45°. This angle is smaller than the KOH chemical-etching angle of silicon (54°) so that a full laser-beam transmission is preserved all over the graphene membrane. The experiment is conducted in a vacuum chamber to suppress air damping, which limits the oscillators' quality factors. Static pressures below 0.01 mbar can be maintained over several days.

The interferometer permits a shot-noise-limited readout of the membrane's thermal noise, with injected optical powers ranging from 1 to 100 μW. A fast piezoelectric element driving the local oscillator mirror permits a robust calibration of the interferometer, particularly insensitive to spatial drifts or reflectivity variations due to nonhomogeneous graphene properties (wrinkles or grain boundaries). Reflectivities in the 1%–10% range are measured on monolayers depending on the level of contaminants. A typical calibrated-displacement noise spectrum is shown in Fig. 1. Its reproduction at varying optical powers permits us to verify the absence of optical backaction (see the SM [20]). The uncoupled graphene resonators present fundamental eigenmodes in the 1–10 MHz range, with quality factors from 10 to 500 in vacuum and effective masses ranging from 10^{-16} to 10^{-14} kg. Operating with fully transmitting systems permits the suppression of additional cavity effects [25] which could complicate the noise thermometry. The spatial profile of graphene eigenmodes can be mapped by probing thermal-noise spectra at varying positions on the graphene membrane; see the SM [20]. The slight elliptical structure and the frequency splitting observed on higher-order modes reflects the presence of a residual 20-MPa stress along the diagonal direction [26,27], attributed to the graphene transfer process. Also visible on the thermal-noise spectrum are sharp peaks corresponding to higher-order eigenmodes of the Si_3N_4 nanomembrane, whose fundamental mode oscillates at around 100 kHz. They present larger quality factors (above 1000) but higher masses, on the order of 10^{-12} kg. In the

following, we investigate the thermal noise of the coupled system.

IV. HYBRIDIZATION OF GRAPHENE EIGENMODES

In order to tune the eigenfrequencies, we exploit the partial absorption of a second laser beam at 532 nm focused down to an optical waist of ≃300 nm, spatially superimposed on the probe beam and injected from the opposite side of the sample. It generates a slight temperature increase which is almost nondetectable in the Brownian temperature [see Fig. 2(e)] but is sufficient to significantly thermally tune the graphene eigenfrequency. A clear hybridization between both the graphene and Si_3N_4 eigenmodes is shown in Fig. 2(b), where a pronounced frequency anticrossing can be seen, as well as a modification of the mechanical damping rates. Such signatures are fingerprints of strong

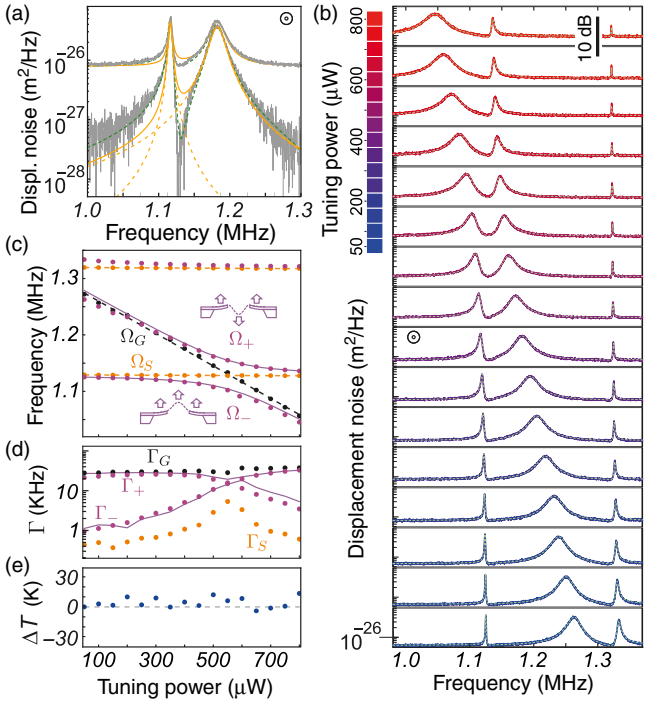

FIG. 2. Thermal noise of the hybridized eigenmodes. (a) Thermal noise of the coupled nanomechanical system measured in the middle of the graphene membrane ($S_{\delta x_G}[\Omega]$) when tuned to an anticrossing region by adjusting the pump intensity (400 μW). Lower traces are obtained after numerical background substraction. The solid lines are the best fits derived employing the normal mode expansion. The dashed green lines are fits using expression (4). (b) Spectra measured through the anticrossing for increasing tuning laser powers. The dashed lines are fits using Eq. (4) with the fitting parameters $\Omega_{S,G}$, $\Gamma_{S,G}$ reported in (c),(d) using $\mu = 0.002$. The purple disks represent the measured coupled eigenfrequencies $\Omega_\pm/2\pi$, and the solid lines are deduced from Eq. (3). (d) A similar analysis for damping rates $\Gamma_\pm/2\pi$. (e) Relative Brownian temperature variation deduced from the fits.

dual-mode coupling [28], which can also affect the force sensitivity [29–31].

The modelization of our inertially coupled nanomechanical system is based on cascaded mechanical oscillators [15,16], as sketched in Fig. 1(d). Their vibrations δx_G and δx_S around the rest positions are coupled through

$$\begin{aligned}\delta\ddot{x}_G &= -\Omega_G^2(\delta x_G - \delta x_S) - \Gamma_G(\delta\dot{x}_G - \delta\dot{x}_S) + \delta F_G/M_G \\ \delta\ddot{x}_S &= -\Omega_S^2\delta x_S - \Gamma_S\delta\dot{x}_S + \mu\Omega_G^2(\delta x_G - \delta x_S) \\ &\quad + \mu\Gamma_G(\delta\dot{x}_G - \delta\dot{x}_S) + \delta F_S/M_S, \end{aligned} \tag{1}$$

where $\Omega_{G,S}/2\pi$ ($\Gamma_{G,S}$) are the uncoupled frequencies (damping rates). δF_G is an external force applied on the graphene membrane and M_G the graphene effective mass at the measurement location [17], while $\mu \equiv M_G/M_S$ parametrizes the hybridization strength. Depending on the graphene and Si_3N_4 membrane geometries which govern the vibration-mode spectrum and their spatial profiles, anticrossings with varying strength can be observed (see the SM [20]). Intuitively, if graphene is positioned at a node of the membrane eigenmode, their hybridization will be reduced. In the Fourier domain, we have $\begin{pmatrix}\delta x_G \\ \delta x_S\end{pmatrix} = \boldsymbol{\chi}[\Omega]\cdot\begin{pmatrix}\delta F_G \\ \mu\delta F_S\end{pmatrix}$, using $\delta x_i[\Omega] \equiv \int_{\mathbb{R}} dt e^{i\Omega t}\delta x_i(t)$. The dynamical matrix $\boldsymbol{\chi}[\Omega]^{-1}$ is

$$\begin{pmatrix}\chi_G^{-1} & M_G\Omega^2 - \chi_G^{-1} \\ \mu(M_G\Omega^2 - \chi_G^{-1}) & \mu(\chi_S^{-1} + \chi_G^{-1} + M_G\Omega^2)\end{pmatrix}, \tag{2}$$

where we use the uncoupled mechanical susceptibilities $\chi_{G,S} \equiv M_{G,S}^{-1}(\Omega_{G,S}^2 - \Omega^2 - i\Omega\Gamma_{G,S})^{-1}$. Diagonalizing the restoring-force matrix $M_G^{-1}\boldsymbol{\chi}[0]^{-1}$ yields the new eigenfrequencies $\Omega_\pm/2\pi$ of the coupled system:

$$\Omega_\pm^2 \equiv \frac{(1+\mu)\Omega_G^2 + \Omega_S^2}{2} \pm \frac{\sqrt{(\Omega_S^2 - (1+\mu)\Omega_G^2)^2 + 4\mu\Omega_G^2\Omega_S^2}}{2}. \tag{3}$$

When $\mu \ll 1$, the minimum relative-frequency splitting amounts to $\sqrt{\mu}$, corresponding to a canonically defined coupling strength of $g = \Omega_G\sqrt{\mu}$ [28]. Depending upon the sample geometry, a large variety of coupling strengths can be observed, up to 200 kHz, largely entering the so-called strong-coupling regime ($g > \Gamma_S, \Gamma_G$). The experimentally measured coupled eigenfrequencies are shown in Fig. 2(c) for increasing pump-laser powers. They can be well fitted using Eq. (3) and a linear pump-power dependence for the uncoupled graphene and Si_3N_4 eigenfrequencies of -284 Hz/μW and -2 Hz/μW, respectively. The latter corresponds to a maximum static heating of the Si_3N_4 nanoresonator estimated at the level of ≃1 K [32]. Using the experimentally measured heat diffusion coefficient of

$5 \times 10^{-6} m^2/s$ (see the SM [20]), the thermal heat resistance of graphene is numerically estimated at 0.25 K per microwatt absorbed. The effective mechanical damping rates $\Gamma_\pm$ of the coupled modes can be roughly estimated using the FWHM of the thermal-noise spectra [see Fig. 2(d)] and then used to extrapolate the uncoupled damping rates (see the SM [20]).

V. VIOLATION OF THE NORMAL MODE EXPANSION

Meanwhile, a striking feature can be seen in the displacement noise spectra shown in Fig. 2: a characteristic peak asymmetry and a sharp noise minimum between both eigenmodes are clearly visible in the anticrossing region. These spectra cannot be fitted with two independent mechanical thermal-noise spectra [see Fig. 2(a)] with a deviation larger than 10 dB observed in the vicinity of Ω_S. Therefore, the measured thermal noise cannot be described by two eigenmodes driven with independent Langevin forces, which reveals the violation of the normal mode expansion. As illustrated in Fig. 3, this violation is a consequence of the spatial inhomogeneity of damping rates across the system: acoustic vibrations are more efficiently damped in graphene than in Si_3N_4. When the eigenmodes become hybridized, their spatial profiles are delocalized

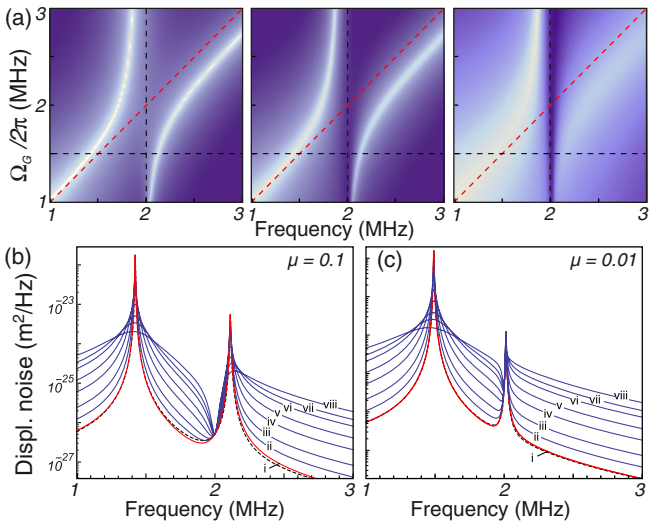

FIG. 3. Role of damping heterogeneity. (a) Numerical simulations of thermal-noise spectra deduced from the model (see the SM [20]) for varying uncoupled graphene frequency Ω_G and different graphene damping rates. From left to right, $\Gamma_G/2\pi = 5$, 50, and 500 kHz ($\Omega_S/2\pi = 2$ MHz, $\mu = 0.1$, $\Gamma_S/2\pi = 5$ kHz). (Bottom panels) Thermal-noise spectra calculated with $\Omega_G/2\pi = 1.5$ MHz for different graphene damping rates [5, 10, 20, 50, 100, 200, 300, and 500 kHz from (i) to (viii)] and respective coupling strength of $\mu = 0.1$ and 0.01 for (b) and (c), respectively. The peak asymmetry and the noise reduction are absent in the case of homogeneous damping (i), and the simulations are well described by a modal expansion (the dashed lines).

over both systems [see the Fig. 2(c) insets], so that mechanical damping becomes inhomogeneous over the eigenmode spatial extension. Thus, the spatial profile of the vibration pattern cannot be stationary anymore since it is nonhomogeneously damped and cannot be preserved over time. As such, dissipation is now able to couple eigenmodes, which breaks the fundamental hypothesis required to apply the normal mode expansion [15,17]. When $\Omega_G = \Omega_S$, the thermal-noise spectral density at the minimum noise frequency is measured at a level approximately $2\Gamma_G/\Gamma_S$ times lower than the prediction of the normal mode expansion; see the SM [20]. The understanding of this deviation is critical for patching the normal mode expansion and working out an analytical description of the system fluctuations.

VI. THERMAL NOISE OF THE HYBRIDIZED NANOMECHANICAL SYSTEM

To properly describe the nanosystem thermal noise, it is necessary to return to the original formulation of the fluctuation-dissipation theorem [15,33]:

$$S_{\delta x_G}[\Omega] = \frac{2k_B T}{|\Omega|} |\mathrm{Im}\chi_{GG}[\Omega]|, \tag{4}$$

which relates the measured displacement noise spectral density to the local mechanical susceptibility χ_{GG}. The latter connects the optomechanically measured deformations of the graphene membrane $\delta x_G[\Omega]$ to the external force δF_G applied on the graphene membrane at the measurement point: $\delta x_G[\Omega] = \chi_{GG}[\Omega]\delta F_G$. First, we pursue the analysis based on the model employed above. Inverting Eq. (2), we obtain

$$\chi_{GG}[\Omega]^{-1} = \chi_G^{-1} - \frac{(\chi_G^{-1} + M_G\Omega^2)^2}{\chi_G^{-1} + \chi_S^{-1} + M_G\Omega^2}, \tag{5}$$

which permits a derivation of the expected thermal noise (see the SM [20]) using Eq. (4). Our experimental results can be well fitted with this model [see Figs. 2(a) and 2(b)] using the fitting parameters which are reported in Figs. 2(c), 2(d), and 2(e). The magnitude of the coupling parameter $\mu = 0.002$ is also in agreement with the ratio of bare effective masses of both nanoresonators. No significant variation in the fitted noise temperature could be detected [see Fig. 2(e)], which places an upper bound of $\simeq$10 K on the maximum temperature increase induced by the tuning laser. This observation is also consistent with the estimated thermal resistance given above and allows us to neglect the role of temperature inhomogeneities in our modelization.

VII. VALIDITY OF THE FLUCTUATION-DISSIPATION THEOREM IN THE COUPLED NANOMECHANICAL SYSTEM

Verifying the validity of the fluctuation-dissipation theorem is essential in order to assess whether the measured

spectra correspond to the thermal noise of the system. Following the principles of linear response theory [33], this verification requires measuring the local mechanical susceptibility χ_{GG} of the coupled nanomechanical system. To do so, we modulate the pump beam intensity by means of an acousto-optic modulator (AOM) and realize response measurements by sweeping the modulation frequency while recording the driven displacement. Both laser spots are carefully superimposed on the graphene membrane to access the local susceptibility; it is worth mentioning that this measurement cannot be realized with electrostatic gate or with piezoelectric actuations since their spatial excitation profile is not localized on the measurement spot. We first verify the linearity of the actuation [see Fig. 4(a)] by varying the optical modulation depth δP over 2 orders of magnitude without modifying the mean pump power (60 μW) to ensure a stable graphene frequency, away from the anticrossings. No deviation from linearity are observed in the driven oscillations up to a maximum amplitude of 1 nm, a few times the monolayer thickness (0.3 nm), so that we perfectly sit in the linear actuation and measurement regime. A typical actuation efficiency of 17 pm/μW is measured, corresponding to an optical force of 540 fN/μW. This value is significantly larger than the radiation-pressure-force contribution of 0.3 fN/μW for a 10% absorption coefficient, which confirms the dominant role of photothermal forces [34,35] in the optical actuation of graphene [25]. The backaction noise resulting from the intensity fluctuations of the shot-noise-limited laser beams can thus be evaluated at the level of $\simeq$0.1 fm/$\sqrt{\text{Hz}}$ for $P_0 = 100$ μW. This value is largely negligible compared to the measured thermal noise, so backaction cancellation [36] and classical noise-squashing mechanisms [37] can be safely excluded while interpreting our results.

Several response measurements are subsequently performed through the anticrossing in the same measurement conditions as in Fig. 2(a) by progressively increasing the pump intensity while maintaining a fixed modulation depth ($\delta P/P_0 = 30\%$). The response curves shown in Fig. 4(c) permit us, once combined with the optical to force conversion factor measured in Fig. 4(a) in the absence of hybridization, to determine the complex local mechanical susceptibility, $\chi_{GG}[\Omega]$, as shown in Fig. 4(d). Its proper determination requires us to take into account the weak residual contribution of the interferometer feedback loop in the measurement span, the transfer function of all photodetectors employed, and the spectral response of the AOM. With this determination, the expected thermal noise can be properly estimated using Eq. (4) and compared to the measured thermal-noise spectrum, as shown in Fig. 4(e). The excellent quantitative agreement found between both measurements all across the hybridization

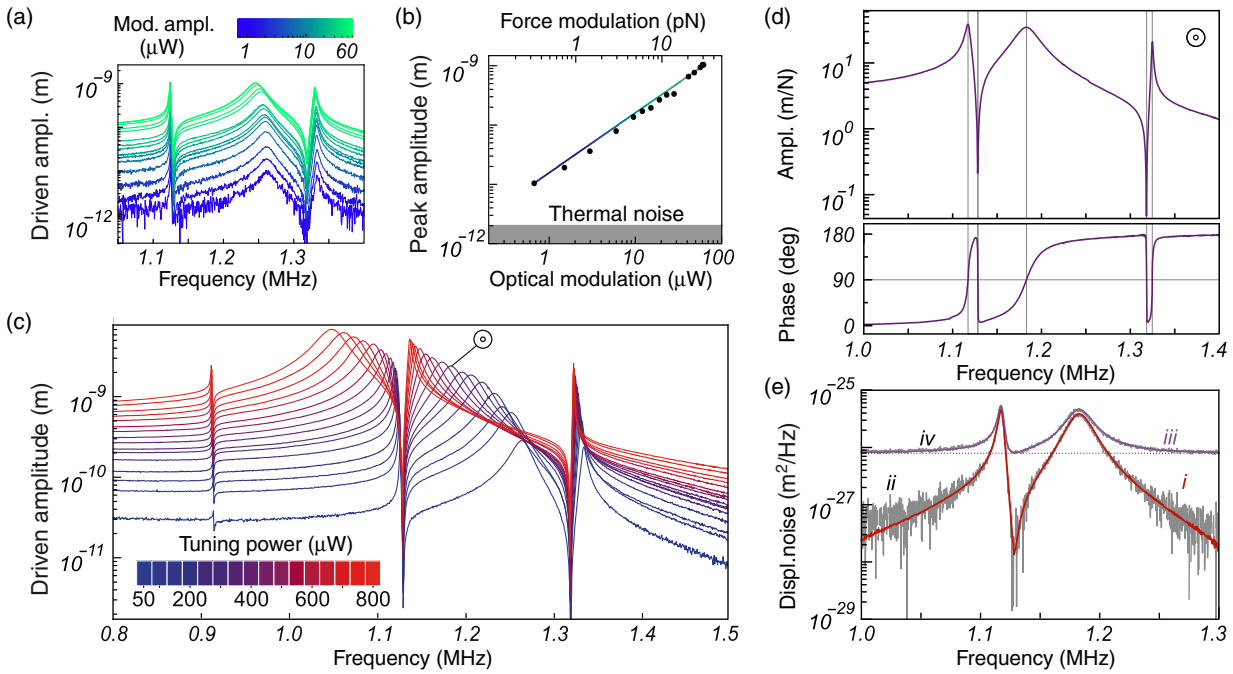

FIG. 4. Optomechanical response of the hybridized nanomechanical system. (a) Optomechanical response obtained by modulating the pump intensity for increasing modulation depths δP with a fixed average tuning power ($P_0 = 60$ μW). (b) Maximum driven displacement reported as a function of δP. The solid line has a slope of 17 μm/W. (c) Optomechanical responses obtained for an increasing optical pump power P_0 (30% modulation strength). (d) Amplitude and phase of the mechanical susceptibility χ_{GG} derived for 400 μW of tuning power. The corresponding thermal-noise spectrum expected using Eq. (4) is reported in (e)(i) and presents a very good agreement with the measured spectrum (ii). The detection noise is included in traces (iii) and (iv). A 30% correction is used here on the optical to the force conversion factor determined in Fig. 4(a) to account for a slight modification of the actuation efficiency between both measurements.

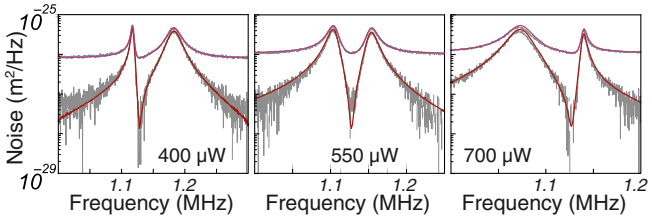

FIG. 5. Probing fluctuation-dissipation theorem validity at different tuning powers while following the same conventions as in Fig. 4(e). Response measurements of Fig. 4 are used to compute the local effective mechanical susceptibility subsequently injected in Eq. (4) and compared to the experimental spectra. For the entire data set, the only free parameter is an overall correction factor (from 0.8 to 1.2) to account for slight drifts in the readout and actuation efficiencies.

(see Fig. 5) demonstrates the validity of the fluctuation-dissipation theorem in our strongly coupled nanomechanical arrangement.

The hybridization dramatically modifies the graphene mechanical response and has an impact on the signal-to-noise ratio (SNR) observed in a force measurement. For a monochromatic force of amplitude δF_G applied in the center of the graphene membrane, the SNR can be expressed as $\mathrm{SNR}[\Omega]/(\mathrm{SNR}_G) = |\mathrm{Im}\,\chi_G^{-1}/\mathrm{Im}\,\chi_{GG}^{-1}|$, where $\mathrm{SNR}_G \equiv \delta F_G^2/2M_G\Gamma_G k_B T$ represents the SNR of the uncoupled graphene alone. As verified experimentally and confirmed with the model (see the SM [20]), the SNR can be improved with respect to the uncoupled graphene resonator in narrow frequency bands in the vicinity of the Si_3N_4 resonance. As already employed with macroscopic devices [19], this approach constitutes a strategy for achieving larger sensitivities in hybrid nanosensors.

VIII. CONCLUSIONS

In this paper, we demonstrate the violation of the normal mode expansion in a multimode nanomechanical system and verify that the fluctuation-dissipation theorem well describes its thermal noise despite the large mass and damping asymmetries. This work underlines the importance of measuring the local mechanical susceptibility of a nanosystem to correctly understand its thermal noise. Since a good sample homogeneity is more delicate to ensure in extremely downsized nanomechanical devices, we anticipate that these deviations will play an important role in the future of nanomechanical sensors. Our observations, realized on inertially coupled nanomechanical oscillators, have a more general reach and are also valid when mechanical modes are externally coupled, such as by optical or electrostatic force-field gradients [8,14,38–44]. Such a fundamental approach could be used for developing alternative force-detection protocols based on multimodal nanosystems. Furthermore, optomechanical cooling is intrinsically responsible for both mode cross-coupling and nonhomogeneous modification of the damping rates when the feedback actuation profile does not perfectly match the vibration profile of the mode of interest. Therefore, our conclusions and our approach should be directly transposed to correctly understand the noise of actively cooled multimode optomechanical systems [38,45–47]. Finally, we emphasize that the coupling mechanism involved here is of a conservative nature in the sense that it does derive from a potential energy.

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