Thermal fluctuations of the optical properties of nanomechanical photonic metamaterials

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The combination of optical and mechanical resonances offers strong hybrid nonlinearities, bistability and the ability to efficiently control the optical response of nanomechanical photonic metamaterials with electric and magnetic field. While optical resonances can be characterized in routine transmission and reflection experiments, mapping the high frequency mechanical resonances of complex metamaterial structures is challenging. Here we report that high-frequency time-domain fluctuations of the optical transmission and reflection spectra of nanomechanical photonic metamaterials are directly linked to thermal motion of their components and can give information on the fundamental mechanical frequencies and damping of the mechanical modes. We demonstrate this by analyzing time-resolved fluctuations of the transmission and reflection of dielectric and plasmonic nanomembrane metamaterials at room temperature and low ambient gas pressure. These measurements reveal complex mechanical responses, understanding of which is essential for optimization of these functional photonic materials. At room temperature the magnitude of metamaterial transmission and reflection fluctuations is broadly on the scale of 0.1% but may reach 1% at optical resonances.

1. Introduction

The thermal vibration frequencies of components of nanomechanical devices increase as objects decrease in size and the amplitude of such oscillations becomes increasingly important. In nanomechanical devices, they are of picometric scale in the mega- to gigahertz range and add noise to induced controlled movements that underpin the functionality. For example, thermal vibrations in nanomechanical photonic metamaterials[1] result in small fluctuations of their optical properties and may perturb their switching characteristics. These fluctuations provide an opportunity for the characterization of mechanical properties. In particular, this applies to highly sensitive photonic metamaterials formed as periodic arrays of optical resonators supported by flexible nanomembrane structures that show giant electro-optical[2], magneto-electro-optical,[3] acousto-optical,[4] phase change[5] and nonlinear optical[6-8] responses. In this paper, we experimentally investigate thermal fluctuations in dielectric and plasmonic nanomechanical photonic metamaterials in the megahertz frequency range and demonstrate how measuring the spectra of thermal fluctuations of transmittance and reflectance can be used to determine the main frequencies and damping of the nanostructures' natural oscillations, at which they can be efficiently controlled by external stimuli. We will examine thermal fluctuations in widely used optical metamaterials, arrays of metamolecules supported on beams (strings) cut from membranes of nanoscale thickness, as illustrated by **Figure 1**.

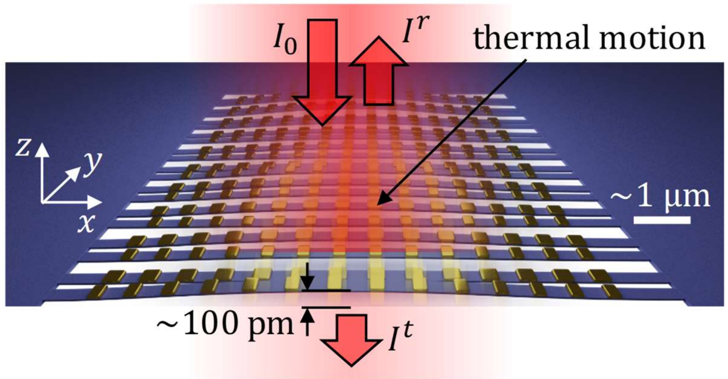

Figure 1. Thermal fluctuation of optical properties of nanomechanical metamaterials. The photonic metamaterial is an array of beams of nanoscale width and thickness located in the *xy* plane. The beams, typically a few tens of microns long, support a periodic array of optical resonators of subwavelength (nanoscale) dimensions. Optical properties of such arrays can be controlled by actuation driven by external stimuli such as electromagnetic forces, optical and acoustic waves.[2-4, 7, 8] Picometric thermal displacements of the individual beams also modulate the optical properties of the metamaterials, causing small fluctuations of their transmittance and reflectance.

2. Brownian motion and optical properties of nanostructured materials: Theoretical background

Classical Brownian motion experiments reveal how a microscopic particle is driven in chaotic motion *externally* by collisions with molecules of ambient gas or liquid. The majority of nanomechanical photonic metamaterials are constructed from cantilevers- or doubly-clamped beam-like components of microscopic length and nanoscopic cross-section. The thermal motion of microscopic/nanoscopic cantilevers or beams arises from interference of thermally populated ensembles of incoherent flexural phonons in the main mechanical modes. Here, the thermal motion of a cantilever or beam in vacuum is driven *internally*, by momentum transfer resulting from the annihilation and creation of phonons in the mode. (The emission and absorption of thermal photons are not important due to the low momentum of such photons.)

Nano-components such as beams and cantilevers can be modelled as damped mechanical oscillators.[9] Considering nanomechanical structures located in the *xy*-plane, and engaged in thermal motion in the *z* direction, the Langevin equation for the thermal motion of such a component can be written as[10]

$$\ddot{z} + 2\pi\gamma\dot{z} + \omega_0^2 z = F_{\text{th}}(t)/m_{\text{eff}} = \sqrt{4\pi k_B T\gamma/m_{\text{eff}}}\eta(t) \quad (1)$$

where $F_{\text{th}}(t)$ is the time dependent thermal force experienced by the oscillator related to the dissipation factor γ through the fluctuation-dissipation theorem, $\eta(t)$ is a normalized white noise term, m_{eff} is the effective mass of the object, k_B is the Boltzmann constant, T is the temperature, $\omega_0 = 2\pi f_0 = \sqrt{(k/m_{\text{eff}})}$ is the natural angular frequency of oscillation, f_0 is the natural frequency and k is the spring constant. The resonance quality factor $Q = {}^{f_0}/_{\gamma}$ in the limit of small damping, $\gamma \ll f_0$, which we assume here.

The origins of mechanical dissipation have been intensively studied over the last decades.[10, 11] The most relevant loss mechanisms include clamping losses due to propagation of elastic waves into the substrate through the supports of the oscillator, fundamental anharmonic effects such as thermoelastic damping and phonon-phonon interactions, materials-induced losses caused by the relaxation of intrinsic or extrinsic defect states in the bulk or surface of the resonator, and, where applicable, viscous damping caused by interactions with surrounding gas atoms or by compression of thin fluidic layers.

Thermomechanical fluctuations of a component's position $z(t)$ are transduced to fluctuations of intensity of light scattered on the component, $\delta I(t) = \frac{\partial\mu(z,\Omega)}{\partial z} \cdot I_0 \cdot \delta z(t)$, where I_0 and $I = \mu(z,\mathrm{F})I_0$ are the intensities of the incident and scattered light, and $\mu(z,F)$ is, generally, a nonlinear function of the component's displacement z and optical frequency F. As, in a stochastic process, the power spectral density is equal to the Fourier transform of its autocorrelation function,[12] the scattered light amplitude spectral density $S(f)$ resulting from small thermomechanical fluctuations in position $\delta z(t)$ is:

$$S(f) = \frac{\delta I}{\sqrt{\delta f}} = \left(\frac{\partial\mu(z,\mathrm{F})}{\partial z}\bigg|_{z=0} \cdot I_0\right) \times \sqrt{\frac{k_B T f_0}{2\pi^3 m_{\text{eff}} Q\left[(f_0^2 - f^2)^2 + (f f_0/Q)^2\right]}} \quad (2)$$

In a nanomechanical photonic metamaterial, a non-diffracting array of identical oscillating components, the same formula will describe the spectra of fluctuations of the intensity of light reflected I^r and transmitted I^t through the metamaterial, see Figure 1. Fluctuations $S^{r,t}$ of the metamaterial's specular reflectance/transmittance correspond to the ratio between the amplitudes of fluctuating and non-fluctuating components of the transmitted/reflected light, that is Equation (2) divided by the incident intensity I_0 and optical reflectance/transmittance of the metamaterial without displacement, $\mu_0^{r,t}(\mathrm{F}) = \mu^{r,t}(0, \mathrm{F})$.

$$S^{r,t}(f) = \frac{\delta I^{r,t}/I^{r,t}}{\sqrt{\delta f}} = \frac{1}{\mu_0^{r,t}(\mathrm{F})} \times \left.\frac{\partial \mu^{r,t}(z,\mathrm{F})}{\partial z}\right|_{z=0} \times \sqrt{\frac{k_B T f_0}{2\pi^3 m_{\mathrm{eff}} Q\left[(f_0^2-f^2)^2+(f f_0/Q)^2\right]}} \quad (3)$$

At the mechanical resonance, $f = f_0$, this modulation depth spectral density reaches a peak value of

$$S^{r,t}(f_0) = \frac{1}{\mu_0^{r,t}(\mathrm{F})} \times \left.\frac{\partial \mu^{r,t}(z,\mathrm{F})}{\partial z}\right|_{z=0} \times \sqrt{\frac{k_B T\, Q}{2\pi^3 m_{\mathrm{eff}} f_0^3}} \quad (4)$$

Reflectance/transmittance fluctuations over a range of mechanical frequencies can be calculated by integration over the power spectral density of the fluctuations.

$$\delta I^{r,t}/I^{r,t} = \sqrt{\int \left(S^{r,t}(f)\right)^2 df} = \frac{1}{\mu_0^{r,t}(\mathrm{F})} \times \left.\frac{\partial \mu^{r,t}(z,\mathrm{F})}{\partial z}\right|_{z=0} \times \sqrt{\frac{k_B T}{2\pi^3 m_{\mathrm{eff}}}} \sqrt{\int \frac{f_0/Q}{(f_0^2-f^2)^2+(f f_0/Q)^2} df} \quad (5)$$

Integration from 0 to ∞, or at least over the whole resonance, gives the root-mean-square (RMS) fluctuations.

$$\delta I_{\mathrm{RMS}}^{r,t}/I^{r,t} = \frac{1}{\mu_0^{r,t}(\mathrm{F})} \times \left.\frac{\partial \mu^{r,t}(z,\mathrm{F})}{\partial z}\right|_{z=0} \times \sqrt{\frac{k_B T}{4\pi^2 m_{\mathrm{eff}} f_0^2}} \quad (6)$$

where the final term corresponds to the RMS beam displacement of

$$\delta z_{\mathrm{RMS}} = \sqrt{\frac{k_B T}{4\pi^2 m_{\mathrm{eff}} f_0^2}} \quad (7)$$

From here is apparent that fluctuations of transmission and reflection will be largest at high temperatures, in metamaterials constructed from very light (low m_{eff}) building blocks, and at optical frequencies where transmission and reflection depend strongly on displacements of said building blocks. The largest fluctuations over a narrow spectral range δf will be seen at high quality mechanical resonances [Equation (4)]. As such, we should expect thermal fluctuations of optical properties to be strongest in highly optically dispersive nanomechanical metamaterials.

Assuming metamaterial beams, such as those shown by Figure 1, with an effective mass m_{eff} = 1 pg and mechanical quality factor of Q = 1000 moving at a damped frequency f_0 = 2 MHz, and a typical change in optical properties (i.e. specular reflectance and transmittance) with beam displacement of $\partial\mu/(\mu_0\partial z) \sim$ 1 %/nm, one may expect to observe an RMS thermomechanical displacement amplitude of $\delta z_{\text{RMS}} = 160$ pm at the centre of the beams at room temperature, resulting in a 0.16% RMS fluctuation of optical properties. At the mechanical resonance, the corresponding spectral densities of displacement and optical property modulation reach peak values of 3 pm/$\sqrt{\text{Hz}}$ and $3 \times 10^{-5}/\sqrt{\text{Hz}}$, respectively.

Thus, in nanomechanical photonic metamaterials, thermal fluctuations are transduced to fluctuations of optical properties that determine the functional noise floor and dynamic range of nanomechanical photonic metadevices. At the same time, observation of the spectra of thermal oscillations gives direct access to the resonant frequencies of natural mechanical modes at which the photonic metamaterial will be most responsive to external stimuli. This information on fluctuation and responsivity of the mechanical sub-system can help in the design of highly efficient metadevices.[13] Moreover, nanomechanical photonic metamaterials typically comprise of large arrays of nominally identical elements (e.g. metamolecules, beams, etc.). However, in practice, with physical characteristics affected by fabrication tolerances, individual nanomechanical oscillators are endowed with slightly different natural frequencies, resulting in the degradation of the optical functionality. In the case where the natural frequencies of individual oscillators are closely spaced compared to the widths of their characteristically Lorentzian lines, the result will be an inhomogeneously broadened metamaterial mechanical resonance. Where the spacing is larger, resonances of individual oscillators may be resolved. Measurements of the spectra of thermal oscillations can provide insight into the nature of such metamaterial resonance broadening and splitting. Previously, transduction of natural oscillations to modulation of light has been observed in cantilevers,[14] antennas[15], microresonators[16] and optomechanical systems[17], and exploited in atomic force microscopy.[18] Here we focus on the role of thermal oscillation in forming the optical properties of nanomechanical photonic metamaterials.

3. Brownian motion and the optical properties of metamaterials: Experimental observation

Below we examine thermal fluctuations in the optical properties of two common types of photonic metamaterial; plasmonic and all-dielectric nanomechanical metamaterials fabricated on membranes of nanoscale thickness.[2, 3, 5-8, 19-21] Both the plasmonic and dielectric metamaterials consist of a planar array of nanoscale optical resonators supported by a 1d array of mechanical resonators, which are flexible beams of microscale length cut from a silicon-nitride membrane by focused ion beam (FIB) milling. Due to their surface-like overall geometry, such metamaterial structures are also known as metasurfaces.

The all-dielectric metamaterial shown in **Figure 2**a,c was fabricated on a 200 nm thick silicon nitride membrane coated with a 115 nm layer of amorphous silicon by plasma-enhanced chemical vapor deposition. This bilayer was then structured by FIB milling to define an array of asymmetric nanorod pairs in the amorphous silicon layer, on 20.3 μm long silicon nitride beams. The structure supports a closed mode optical resonance[22] at a wavelength of 1530 nm, underpinned by the excitation of antiparallel displacement currents in the pair of amorphous silicon nanorods,[23] see Figure 2a and **Figure 3**.

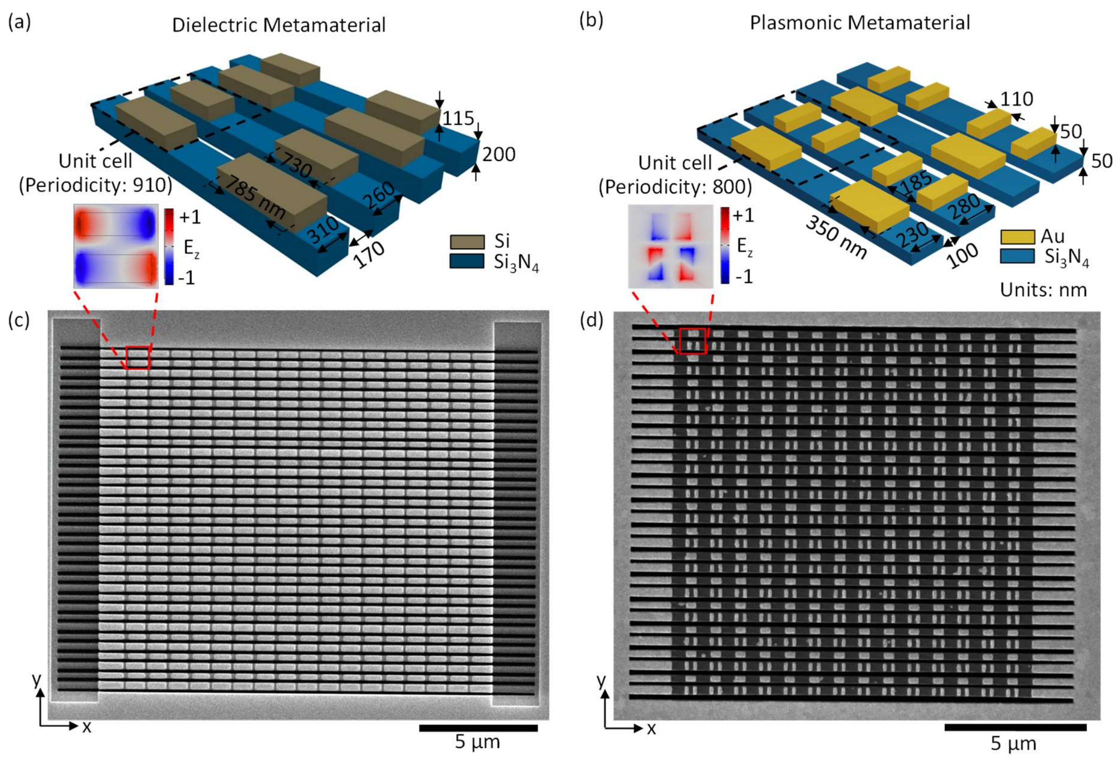

Figure 2. Nanomechanical photonic metamaterials. Structural schematic and SEM images of planar dielectric (a, c) and plasmonic (b, d) metamaterials fabricated on silicon nitride membranes. Insets show the resonant field distributions excited by *x*-polarized incident light at wavelengths of 1550 nm (a) and 1310 nm (b).

The plasmonic metamaterial shown in Figure 2b,d was manufactured on a 50 nm thick silicon nitride membrane coated with a 50 nm layer of gold by thermal evaporation. This bilayer was then structured by FIB milling to define gold nanorods, in groups of three per unit cell in a Π-shaped arrangement split across two adjacent 14.4 μm long silicon nitride beams. The Π-shaped nanorod arrangement supports a Fano-type optical resonance[7, 24] at a wavelength of 1350 nm, resulting from the interference between a "bright" dipole mode excited in the larger gold bar and an antisymmetric and "dark" magnetic dipole mode induced in the pair of smaller bars, see Figure 2b and Figure 3.

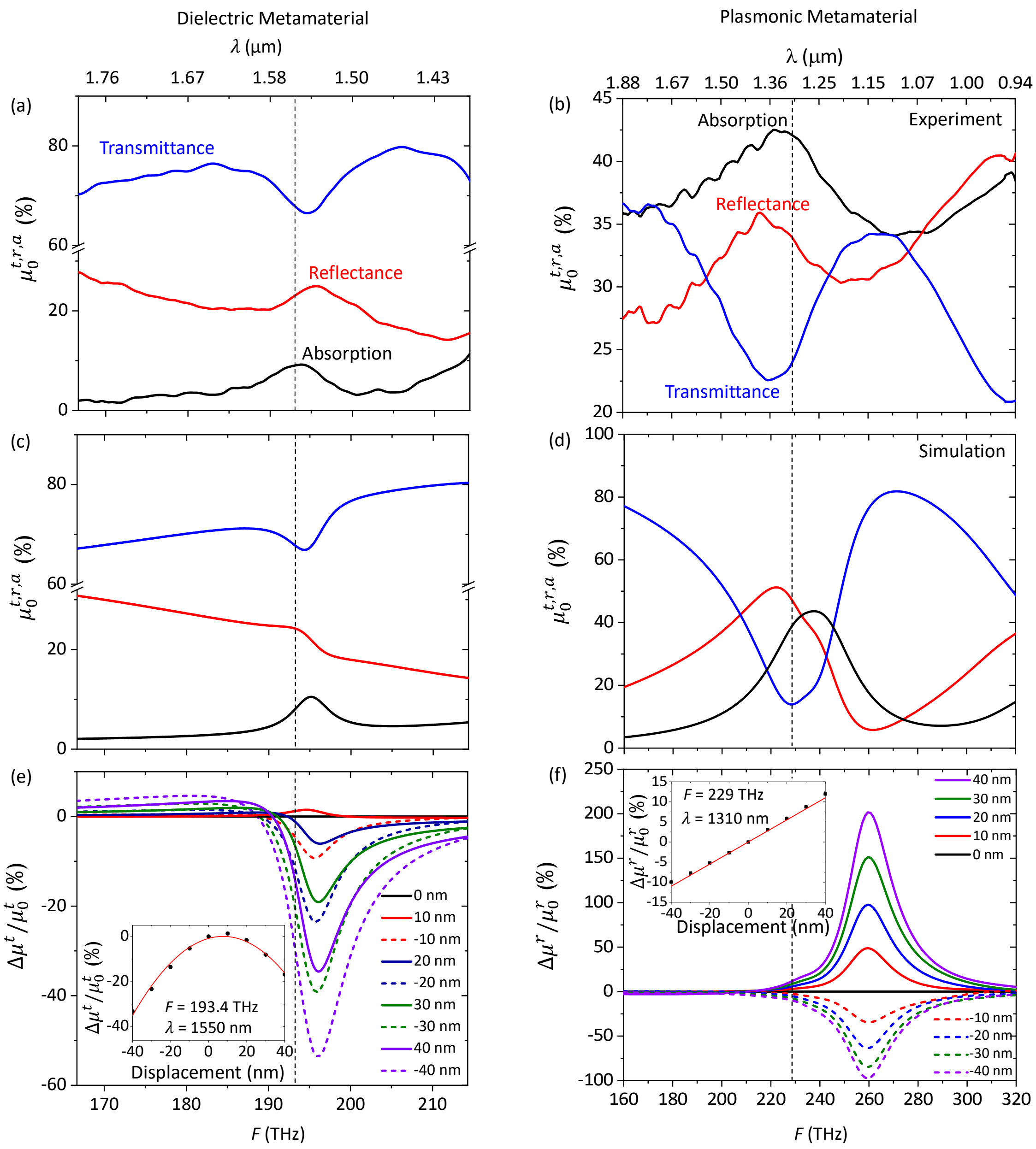

Figure 3. Optical properties of the dielectric and plasmonic nanomechanical metamaterials. Experimentally measured (a, b) and computed (c, d) optical reflection (μ_0^r), transmission (μ_0^t) and absorption (μ_0^a) spectra of the dielectric and plasmonic metamaterials. Computed values (e, f) of $(\Delta\mu^{t,r}/\mu_0^{t,r})$, a figure of merit of responsivity of the metamaterial's optical properties to the relative displacement of neighbouring beams along z at different levels of displacement. The insets show the dependence of $(\Delta\mu^{t,r}/\mu_0^{t,r})$ on displacement at wavelengths of 1550 nm and 1310 nm, respectively. Positive displacement corresponds to the movement of narrower beams along $+z$ relative to wider beams; all results are for x-polarized light.

The dielectric and plasmonic metamaterial structures presented in Figure 2 and studied in this work are planar nanomechanical photonic metamaterial designs, which can be reconfigured: a) thermally, by changing the external temperature due to differential thermal expansion of neighbouring beams,[21] b) optically, when the displacement of neighbouring beams is driven by optical heating and the interaction between oscillating electric dipoles induced by the incident light,[7, 8] c) acoustically, by ultrasound vibrations,[4] d) electrostatically, by the Coulomb force between charged neighbouring beams[2] or selected beams and another electrode,[20] and e) magnetically, by the Lorentz force acting in a static magnetic field on a current passing through the beams.[3] (In the latter two cases an electrically conductive layer is added to the structure.)

In both types of metamaterial, differential movements between neighbouring beams change the unit cell geometry and thereby the optical properties of the array. In such structures, the frequencies of the fundamental mechanical oscillatory modes f_0 of the beams lie in the megahertz range. At the same time, the optical properties of the metamaterials are most sensitive to the beam movements when the optical frequency F is near either a plasmonic or dielectric resonance, where the value of $\left.\frac{\partial \mu^{r,t}(z,F)}{\partial z}\right|_{z=0}$ [see Equation (3)-(6)] can be much higher than off-resonance. Here, the variation of optical properties is generally a nonlinear function of beam displacement, but may be approximately linear for sufficiently small displacements. This is illustrated by 3D finite element Maxwell solver simulations of the resonant optical properties of the arrays for different levels of mutual displacement between neighbouring beams, Figure 3e-f. For instance, for displacements of up to 40 nm, the reflectivity of the plasmonic metamaterial changes linearly with displacement at 1310 nm (Figure 3f), while transmission of the dielectric metamaterial changes approximately parabolically with displacement at 1550 nm (Figure 3e).

The thermal fluctuation of the optical properties of the dielectric and plasmonic nanomechanical metamaterials were studied at wavelengths of 1550 nm (transmission mode, 19.6 μW incident power) and 1310 nm (reflection mode, 48.2 μW power and) respectively with *x*-polarized CW laser light and ~5 μm spot size. The metamaterial samples were mounted in a vacuum chamber at a pressure of 4-5 μbar to reduce air damping of the mechanical modes. The intensity of light transmitted and reflected from the samples was monitored with a photodetector and a radio frequency spectrum analyser.

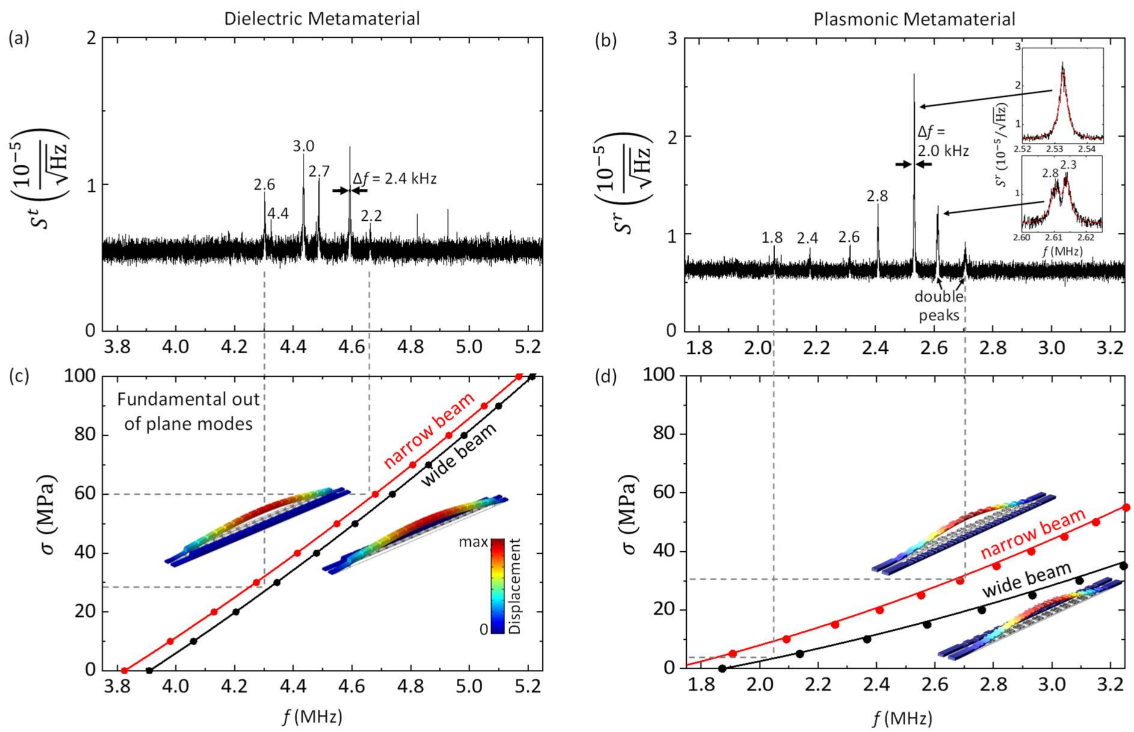

Figure 4. Thermal fluctuation of the optical properties of nanomechanical metamaterials. Measured spectral density of transmission modulation S^t (a) and reflectance modulation S^r (b) by dielectric and plasmonic metamaterials, respectively. Resonant peak widths Δf are labelled in kHz and insets show enlarged examples of single (double) resonant peaks with single (double) Lorentzian fits. Frequencies of the fundamental out-of-plane resonances (insets) of the dielectric (c) and plasmonic (d) metamaterial beams for different levels of tensile stress σ. Simulated resonance frequencies of the nanostructured beams (data points) are shown with a fit according to Equation (8) (lines).

As expected from Equation (3), in the spectra of transmitted and reflected light we observe a range of sharp peaks linked to the frequencies of thermal movements of the silicon nitride beams at their natural frequencies in the megahertz range, **Figure 4**a,b. Here, the peaks are observed against a flat background of the photodetector noise. The magnitude of the underlying thermal RMS beam displacements is ~50 pm (~160 pm) for the dielectric (plasmonic) metamaterial beams at room temperature according to Eq. (7). The average quality factor of the observed mechanical resonances for the dielectric metamaterial is 1.6×10^3, slightly higher than the quality factor of 1.0×10^3 for the plasmonic metamaterial.

In both cases, we see an isolated group of several peaks with Lorentzian profiles, that overlap in the case of closely-spaced resonances (insets of Figure 4b). The Lorentzian line shape indicates that different peaks are related to different beams within the structure. Thus, the collective mechanical response of the metamaterial is the sum of the individual responses of its mechanical elements. Due to a wide distribution of resonance frequencies of a small number of mechanical resonators, we resolve the individual resonances, resulting in the observation of a group of Lorentzian lines, rather than spectral broadening of a single peak. (The metamaterial beams are necessarily coupled through the supporting membrane, but we do not observe any evidence of coupling effects such as synchronization. The observation multiple resonance peaks indicates oscillation at different frequencies.) Here, the main cause of variations between the resonance frequencies of individual, nominally identical beams is most likely disparities in beam tension across the sample resulting from the non-uniformity of intrinsic stress in the membrane, rather than variations of their physical dimensions. These variations in tensile stress across the metamaterials, leading to the spread of individual peak frequencies, can be evaluated from Euler-Bernoulli beam theory,[12, 25] which gives the stress-dependent fundamental frequency of a doubly clamped beam of rectangular cross-section as:

$$f_0 = 1.03 \frac{t}{L^2} \sqrt{\frac{E}{\rho} \left(1 + \frac{\sigma L^2}{3.4 E t^2}\right)} \quad (8)$$

where t is the beam thickness, E the Young's modulus, ρ the density of the material and σ the tensile stress along the beam length.

Figure 4c,d, illustrating the relationship between stress and fundamental frequency, shows that for both the plasmonic and dielectric metamaterials, the observed variation of the fundamental mechanical resonance frequencies of beams are explainable by tensile stress variations from beam to beam of ~30 MPa. This is not a surprizing number as stresses of between several hundred megapascals and a gigapascal are common in unstructured silicon nitride membranes,[26] resulting in significant post FIB-fabrication stress variations across metamaterial arrays.

Inhomogeneous illumination of several beams within the optical spot profile can also contribute to inhomogeneous shifts of beam frequencies through thermal expansion of the beam length, which reduces stress: $\sigma = \sigma_0 - \alpha E \Delta T$, where σ_0 is the initial tensile stress, α the thermal expansion coefficient of the material and ΔT the temperature change. Considering conductive cooling, in our experiments, the laser-induced temperature increases reach ~10 K in the plasmonic metamaterial and ~1 K in the dielectric metamaterial. For a silicon nitride beam with an initial stress of 30 MPa, such a temperature increase translates to around 20% stress reduction in the plasmonic metamaterial (2% for the dielectric metamaterial). In the present cases, this translates to a 6% (2%) decrease in resonance frequency, see Figure 3c,d. We observe a significantly larger spread of resonance frequencies, with the lowest resonance frequency being 24% (7%) smaller than the largest, suggesting that the observed spread of resonance frequencies is mainly due to intrinsic stress variations of the beams.

The fluctuation of the optical properties of the metamaterials is presented in terms of $S^{r,t}$, the relative spectral density of transmission/reflectance modulation, see Figure 4a,b. To evaluate the root-mean-square of relative fluctuation of optical properties $\delta I^{r,t}_{\mathrm{RMS}}/I^{r,t}$ resulting from thermomechanical fluctuations we need to integrate, as shown by Equation (5), over the entire frequency range. If only fundamental mechanical modes of the metamaterial beams are taken into account, this integral can be approximated as $S_0\sqrt{N\Delta f}$, where N is the number of peaks/modes and S_0 and Δf are the average amplitude and width of the peaks.

From the data presented in Figure 4a,b, we can evaluate the level of transmission fluctuation as $\frac{\delta I^{t}_{\mathrm{RMS}}}{I^{t}} = 0.05$ % for the dielectric metamaterial at 1550 nm, and the level of reflection fluctuation as $\frac{\delta I^{r}_{\mathrm{RMS}}}{I^{r}} = 0.1\%$ for the plasmonic metamaterial at 1310 nm. These values can increase to about $\frac{\delta I^{t}_{\mathrm{RMS}}}{I^{t}} = 0.1$ % and $\frac{\delta I^{r}_{\mathrm{RMS}}}{I^{r}} = 1.5\%$ at the dielectric and plasmonic resonances, see Figure 3e,f.

Finally, we shall note that thermal fluctuations can give rise to nonlinear effects. The nonlinearity of coupling between metamaterial optical properties $\mu^{r,t}(z,F)$ and beam displacements z will result in fluctuations of the optical properties at harmonics of the fundamental mechanical oscillation frequency f_0. Moreover, the mechanical motion itself may be thermally excited into the nonlinear regime. In the case of beams anchored at both ends, the oscillation becomes nonlinear when the amplitude of oscillation becomes comparable to the beam thickness t divided by the square root of the mechanical quality factor Q:[27] a beam with length L, width w and thickness t enters the nonlinear regime of thermal motion for

$$L > 2.6\left(\frac{Ewt^5}{Qk_BT}\right)^{1/3} \qquad (9)$$

As an example, at room temperature, a silicon nitride beam of rectangular 100 nm × 100 nm cross-section and quality factor Q = 1000 will enter the nonlinear regime of Brownian motion at L >100 µm. Such effects though lie beyond the scope of the present study, as the sub-200-

pm RMS displacements of our metamaterial beams are too small to enter the nonlinear regime, which would require displacements of several nanometres in our structures.

4. Conclusion

In conclusion, we have observed that the optical properties of dielectric and plasmonic nanomechanical photonic metamaterials at near-infrared (telecoms) wavelengths exhibit thermal fluctuations of order 0.1%., rising potentially to ~1% at optical resonances. The spectra of fluctuations enable metamaterial characterization at the level of individual mechanical elements. They provide exact information on the frequencies at which a nanomechanical photonic metamaterial can be efficiently actuated by external stimuli, providing insight to mechanisms of broadening and splitting of mechanical resonances in metamaterials, and aiding in the optimization of their performance. Beyond metamaterials, our approach may be applied to the characterization of mechanical resonances of similar micro-/nanostructures, e.g. in nano-electromechanical and biological systems.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Data Availability Statement
The data that support the findings of this study are openly available from the University of Southampton ePrints research repository at https://doi.org/10.5258/SOTON/D1887 .[28]

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Brownian motion leads to small fluctuations of transmissivity and reflectivity of nanomechanical photonic metamaterials. Observation of such perturbations gives direct information on fundamental frequencies and quality factors of mechanical modes of metamaterials, and reveals the frequencies at which the metamaterial can be actuated efficiently.

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Thermal fluctuations of the optical properties of nanomechanical photonic metamaterials