Picosecond optomechanical oscillations in metal-polymer microcavities

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We experimentally study mechanical vibrations in planar Fabry–Perot microcavities made of metallic mirrors and a polymer spacer, using broadband pump-probe spectroscopy. These acoustic waves oscillate at a picosecond time-scale and result in spectral oscillations of the cavity transmission spectrum. We find that the oscillations are initiated at the metal mirrors and that their temporal dynamics match the elastic modes of the polymer layer, indicating that mechanical momentum is transferred within the structure. Such structures combine the strong optical absorption of metals with the elasticity and the processability of polymers, which open the road to a new class of optomechanical devices.

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The sensitivity of optical resonators to small morphological distortions provides an efficient coupling mechanism between light and acoustic oscillations, which lies at the heart of cavity optomechanics. Microcavities of different geometries were shown to exhibit spontaneous mechanical oscillations when CW light was injected into a cavity [1,2], as a result of the interplay between optical forces acting on the structure and the optical back-action on light. Similarly, excitation of acoustic modes in semiconductor DBR microcavities was revealed by Raman spectroscopy [3–5], and later observed in the time domain using pump-probe measurements [6–8]. In these experiments, the mechanical vibrations of the cavity structure shifted the optical resonance, resulting in a periodic modulation of cavity optical response. Similar microcavity structures, designed to have high quality-factors for both the optical and mechanical modes [5,9], were implemented in the so-called phonon laser, where the optomechanical coupling provides the gain mechanism for coherent mechanical oscillations. At the nano scale, optomechanical coupling has been extensively studied in metallic nanoparticles [10–15], where the plasmonic resonance couples to the breathing modes of particles of various shapes and compositions. Such optomechanical coupling was used to study the mechanical properties of nano objects using pump-probe spectroscopy [13,15].

Here we study optomechanical coupling in metallic Fabry–Perot cavities filled with a thin polymer film as a transparent spacer, by using time-dependent pump-probe spectroscopy. We show that a short laser pulse impulsively excites one or several vibrational modes in the structure, which corresponds to longitudinal elastic waves in the polymer. The oscillation period scales linearly with the cavity thickness in accordance with the mechanical properties of the polymer spacer, and by analyzing the time-dependent spectral response of the cavity, we reveal that the magnitude of the vibrations is on the order of 0.1 Angstrom. Furthermore, we show that the vibrational modes are excited through absorption inside the metal mirrors, followed by momentum transfer into the polymer layer.

The structure used in our experiments is a metallic Fabry–Perot microcavity made of two silver mirrors separated by a polyvinyl alcohol (PVA, Mw 205,000, Sigma Aldrich) polymer spacer, as illustrated in the inset of Fig. 1. The sample was prepared by first depositing a 31 nm layer of silver on a precleaned microscope slide using a sputter-coater, followed by spin-coating a PVA − H2O solution (2.5 wt%) at 1000 rpm. Finally, a second Ag mirror was sputtered on top of the PVA layer, forming a cavity with a fundamental-mode transmission

Fig. 1. Cavity transmission spectrum measured at normal incidence (solid black line) and the simulated spectrum (dashed red line). The inset shows a sketch of a cavity structure with the thicknesses extracted from the numerical fit of the transmission.
resonance at 606 nm (solid black curve). The cavity Q factor of \(~20\) is dominated by losses in the metal. The second peak seen at \(\sim 330\) nm is associated with the transmission of silver as its dielectric function crosses zero at the plasma frequency [16]. Figure 1 also shows simulation results for the transmission spectrum, calculated by the transfer-matrix method, where the PVA layer thickness was found to be 149 nm by fitting the calculated cavity resonance to the measured spectrum. The dielectric function of silver used in the simulations was acquired by ellipsometry, and the PVA was taken to have a refractive index of 1.52. Other cavity samples were fabricated similarly, using different PVA solution concentrations or spin-coating speeds to vary the cavity thickness, which was extracted by numerical fitting.

Time-resolved spectroscopic measurements were performed using a fs pump-probe setup (Helios, Ultrafast Systems), pumped by an 80 fs, tunable-wavelength OPA (Topas, Light Conversion) and using a white-light probe (400–800 nm) generated by a sapphire plate. The system was operated in transmission mode, with the pump focused on the sample to a \(\sim 100\) \(\mu\)m spot. We found that transient reflection measurements gave similar behavior, however, with a reduced signal-to-noise ratio. Figure 2(a) shows the relative transient transmission spectrum \(\Delta T/T\) as a function of the time delay between the pump and the probe, using a 530 nm pump with a pulse energy-density of 0.2 J/cm\(^2\). The transient signal clearly shows spectral oscillations with a period of \(~50\) ps with a \(\pi\) phase-shift when going from negative to positive detuning of the probe wavelength with respect to the cavity resonance (606 nm). This behavior is also seen in Figs. 2(b) and 2(c), showing the transient spectra at several time delays, and in the temporal kinetic measured at 590 nm. We found that the signal is linear with the pump intensity, with hardly any effect of the intensity or the pump wavelength on the temporal and spectral characteristics of the measured kinetics, within the range of intensities used. In these measurements, we may identify several processes occurring on different time-scales: immediately after the pump, a strong signal appears and decays within \(2–3\) ps. On a long time-scale of several hundreds of ps, we observed irregular dynamics, accompanied by a slow spectral shift. We associate the first, fast process with the excitation of hot electrons in the metal mirrors, which thermally relax within several ps [11,17,18], while the slower (ns-scale) dynamics can be attributed to heat diffusion from the metal to its surroundings. These two processes modify the dielectric function of the mirrors and of the polymer layer, which results in a shift of the cavity resonance in a complex manner. Superimposed on the latter, slow dynamics, we find an oscillatory behavior of the transmission spectrum, which is still above the measurement noise level even 1 ns after the excitation [see inset of Fig. 2(c)]. This periodic signal, which is the focus of the current study, is reminiscent of the spectral oscillations previously seen in pump-probe spectroscopic measurements conducted on dielectric microcavities and metallic nanoparticles [6,7,10–15], and similarly we attribute this oscillating spectral component to mechanical vibrations which are impulsively excited in the microcavity structure.

To characterize the spectral oscillations and to isolate them from the rest of the dynamic response of the structure, we Fourier-transformed the transient signal, over a full time-window of 1 ns. Figure 3(a) shows the power spectral density (PSD) of the kinetics, summed over all wavelengths in the range of 560–650 nm. The contribution of the periodic signal is clearly observed as a sharp peak at 22.8 \(\pm 0.9\) GHz, corresponding to an oscillation period of 44 ps. Moreover, the characteristic decay time of the oscillations is approximately 0.5 ns [Fig. 2(c)], corresponding to an acoustic Q-factor of \(~20\). Next, we tested how the acoustic mode frequency depends on the cavity dimensions. For this purpose, we fabricated cavities with different thicknesses and then repeated the pump-probe measurements and extracted the oscillation period using Fourier decomposition. Figure 3(b) shows that the oscillation period scales linearly with the thickness of the PVA spacer, indicating that the periodic signal originates from an acoustic compression mode of the polymer film. This, in turn, periodically modulates the distance between the metallic mirrors, resulting in a periodic shift of the cavity mode around its steady-state resonance wavelength. Taking the wavelength of the acoustic wave as the thickness of the polymer spacer, the inverse of the sound velocity of the mechanical oscillations is given by the slope of the linear curve in Fig. 3(b). Our measurements yield a value of 3650 m/s, which is of the same order of magnitude as typical sound velocity measured for polymers [19], confirming once again that the acoustic mode is primarily concentrated on the polymer spacer. Furthermore, by calculating the amplitude of the Fourier transform at the oscillation frequency for each wavelength separately (without wavelength-averaging), we can isolate the spectral component of the oscillations from the rest of the spectral dynamics. Performing this procedure...
for the transient spectrum of Figure 2 (at a frequency of 22.8 GHz), we find that the oscillation’s spectral signature is asymmetric at approximately 603 nm, as shown by the solid line in Fig. 3(c). In addition, we used our simulation to calculate the transient spectrum expected for a slight decrease in the polymer thickness, which is shown by the dashed line in Fig. 3(c). As shown, the simulated spectrum accurately fits the oscillation spectrum as extracted from our measurement, confirming that the observed oscillations are indeed the result of period compression and expansion of the polymer layer, which gives rise to a periodic back-and-forth shift in the cavity resonance (note that for the half-cycle corresponding to expansion the transient spectrum will be inverted). Moreover, by matching the amplitudes of the two spectra in Fig. 3(c), we find that the amplitude of the mechanical oscillations is approximately 0.1 Å, corresponding to a spectral shift of 0.3 nm for the cavity resonance and a mechanical strain of $6 \times 10^{-5}$.

Considering the results in Fig. 3, one may argue that the oscillations are initiated directly at the PVA layer. One mechanism which might be considered is a fast thermal expansion due to heat deposited through absorption, as was observed in time-domain thermoreflectance measurements of thin polymeric films [19]. Because the polymer itself is nearly transparent for the pump wavelength (with optical losses of less than 1%), it is more reasonable to assume that heat is mainly generated in the metal mirrors, and only then diffuses into the PVA spacer. However, when taking into account the thermal conductivity of PVA ($\Lambda = 0.3 \text{ Wm}^{-1} \text{K}^{-1}$) and its heat capacity $C = 1.6 \times 10^{6} \text{ Jm}^{-3} \text{ K}^{-1}$ [19] together with a typical polymer thickness of $L = 150 \text{ nm}$, one can obtain an estimate for the time scale for thermal conduction into the polymer of $\tau = \frac{CL}{\Lambda^2} = 30 \text{ nsec}$. This time scale is much longer than both the oscillation period and the measurement time window, and we therefore conclude that the initial “kick” acting on the structure cannot originate from thermal effects in the PVA spacer itself. Moreover, with PVA being an insulating, disordered material, processes such as photogeneration of excitons or the inverse piezoelectric effect which are relevant to phonon excitations in semiconductor microcavities [20] cannot occur in our system. Instead, similar to the mechanism taking place in metallic films [20–22] and metallic nanoparticles [10–13], the ultrafast heating and subsequent rise in pressure of the free electron cloud in the metal mirrors results in the generation of a coherent strain pulse. This mechanical energy is then transferred from the metal layer into the polymeric film [23–25], initiating the acoustic vibrations observed in our experiments.

The oscillations described above represent the lowest breathing mode of the polymer layer. However, as the dimensions of the structure are increased, higher-order modes are expected to appear [26]. Indeed, for a 255-nm-thick PVA spacer, the Fourier transform gives two distinct peaks, as seen in Fig. 4(a)—the fundamental mode at 14 GHz and a second mode at a double frequency (28 GHz). This half-period oscillation is also clearly visible in the kinetics of the signal at 460 nm, shown in the inset of Fig. 4(a). A similar behavior is also observed for a cavity with a 580 nm-thick spacer: The Fourier transform, presented in Fig. 4(b), reveals higher oscillation modes at 12, 18, and 24 GHz, representing the second, third, and fourth harmonic of the fundamental frequency (which is obscured by low-frequency background originating from slow thermal effects). Note that the fundamental period (166 ps) aligns well with the linear dispersion of Fig. 3(b). Although high-harmonic modes can in principle always exist, we did not observe modes with a frequency higher than 30 GHz in any of our measurements, indicating that a strong loss mechanism suppresses these high-frequency vibrations and prohibits their observation. Notice that this upper frequency is well below the cutoff-frequency of our measurement (1 THz) set by the 0.5 ps time-steps used for the delay stage.

Finally, we investigated the dependence of the oscillation amplitude on pump wavelength and the effect of the optical resonance of the cavity on the optomechanical coupling. We repeated the pump-probe measurement with a 318-nm-thick cavity while scanning the pump-wavelength across the cavity

\[ \Delta T / T [\%] \] 

\[ \text{Wavelength [nm]} \] 

\[ \text{Frequency [GHz]} \] 

\[ \text{Oscillation Period [\text{ps}]} \] 

\[ \text{Kinetics PSD [AU]} \] 

\[ \text{m = 1} \] 

\[ \text{m = 2} \] 

\[ \text{m = 3} \] 

\[ \text{m = 4} \] 

\[ \text{Delay [ps]} \] 

\[ \text{m = 2} \] 

\[ \text{m = 3} \]

Fig. 3. (a) Power spectrum of the transient spectrum of Fig. 2, averaged over all wavelengths. (b) Measured oscillation period for different cavity thicknesses (circles), with a linear fit (dashed line) corresponding to a sound velocity of 3650 m/s. (c) Optical spectrum of the 22.8 GHz oscillation (solid line), as extracted from the Fourier decomposition of (a), compared to a simulated differential transmission spectrum calculated for a 0.1 Å variation in the cavity thickness (dashed line).

Fig. 4. Power spectrum of the transient transmission measured for cavities with (a) a 255-nm-thick spacer and (b) a 580-nm-thick spacer, revealing the simultaneous excitation of acoustic resonances at multiple harmonics. The indices $m$ indicate the harmonic order of the vibrational modes and the inset in (a) shows the temporal kinetics of the signal, measured at a wavelength of 460 nm.
resonance, and calculated the oscillation amplitude as described above (Fig. 3). To avoid spectral overlap of the interrogated probe wavelength region (near the 560 nm cavity resonance at normal incidence) with scattering of the pump we took advantage of the angular dependence of the cavity resonance by launching the pump beam at 45° with respect to the cavity. Under these conditions, the cavity resonance, as seen by the pump beam (for TE polarization), is located at 503 nm. The results are presented in Fig. 5, showing the dependence of the oscillation intensity on the pump wavelength (full circles), normalized by the pump intensity at each wavelength. These measurements are superimposed on the cavity absorbance profile (taken as A = 1 − T − R, where T and R are the transmission and reflection spectra, respectively, measured at 45°). The excitation efficiency of the mechanical oscillations is maximal when the pump wavelength is tuned to the optical cavity resonance and decreases around it in accordance with the cavity absorption line shape. This proves that the mechanical oscillations are indeed caused by deposition of heat in the structure, which, as discussed above, predominantly takes place in the metallic mirrors.

In conclusion, we presented an extensive experimental study of optomechanical coupling in metal-polymer microcavities and studied the dynamics of the excited vibrations using time-dependent pump-probe spectroscopy. We found that these vibrations correspond to elastic waves which are excited in the polymer layer, and that their period scales linearly with the polymer thickness, matching a phase velocity of 3650 m/s. We observed high-order modes up to a fourth harmonic of the fundamental frequency, with a frequency cutoff at ∼30 GHz, above which no vibrational modes were observed. Furthermore, we found that the optomechanical coupling is maximal when the pump wavelength is tuned to the cavity resonance, where the absorption of light into the metallic mirror is most efficient. Our measurements indicate that the mechanical vibrations are initiated by absorption in the metal mirrors, followed by a fast expansion of free electrons and momentum transfer into the polymer layer. Such a mechanism can be used for a new class of optomechanical devices, taking advantage of the strong absorption in metals along with the wide variety of material-processing methods applicable to polymers. Moreover, we showed that broad-band pump-probe spectroscopy can detect minute (subangstrom) structural changes in the cavity. Therefore, such systems may be used for studying the mechanical properties of polymers at the GHz regime, relying on the enhanced optical sensitivity provided by the cavity.

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