Selective Exciton-Phonon-Phonon Coupling and Anharmonicity with Cavity Vibrational Phonons and MoS$_2$ Lattice Phonons in Hybrid Nanobeam Cavities

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We report selective coupling between neutral excitons $X^0$, vibrational phonon modes of a free-standing nanobeam cavity and lattice phonons of a MoS$_2$ monolayer fully encapsulated by hBN. Our experimental findings demonstrate that the cavity vibrational phonons selectively couple to neutral excitons ($X^0$), and the coupling to negatively charged trion ($X^−$) being significantly weaker. We establish this result by studying the lattice temperature induced broadening of exciton linewidths, where the contribution from the $X^0$-cavity phonon coupling is clearly observed while the $X^−$-cavity phonon coupling is not. Furthermore, when the Raman modes of MoS$_2$ lattice phonons $A_{1g}$ and 2LA are tuned into an outgoing resonance with exciton emissions, we observe the $X^0$-cavity phonon-lattice phonon coupling which inherits the characteristics rule the of $X^0$-cavity phonon coupling. As a result, $X^0$-induced Raman scatterings are enhanced, while $X^−$-induced scatterings are suppressed, revealed by the detuning-dependent Raman intensities and the ratio of $X^−/X^0$ emission intensities.

The phonon anharmonicity from the coupling between cavity vibrational phonons and MoS$_2$ lattice phonons is further demonstrated by the observed Raman linewidth. Such hybrid couplings between materials and nanostructures enable the control of phonon-induced processes in nanophotonic and nanomechanical systems incorporating 2D semiconductors.

Opto-mechanical systems are of very strong interest for cavity-QED experiments since they intentionally couple optical, electronic and vibrational degrees of freedom having very different eigenfrequencies. Hereby, the interband optical response of the system becomes sensitive to the local photon field and vibrational state-of-motion dynamics [1–5]. Monolayers of transition metal dichalcogenides (TMDs) are of particular interest in this context since they: (i) can be attached via van der Waals bonding to a wide range of different substrates, (ii) combine strong-light matter interactions mediated by excitonic processes at room temperature, (iii) have large photelastic coupling strengths and (iv) feature atomically-thin layers with electronic and optical properties that are intrinsically sensitive to both hydrostatic, biaxial, uniaxial and flexural strain [6–8]. Mechanical cavities formed from freely suspended TMD monolayers have been previously prepared to study their vibrational and torsional phonon modes [7–10]. However, in such studies the TMD monolayer is not usually fully encapsulated within hexagonal boron nitride (hBN), an approach known to enhance the optical linewidth and stability of the excitonic transitions. Without encapsulation, adsorption of molecules onto the active TMD monolayer and environmental disorder degrade excitonic properties [11, 12] and limits further investigations on the exciton-phonon couplings in cavities.

In contrast, recent work reports that fully encapsulated TMDs can be embedded into nanobeam cavities [13] which allow them to support both optical and vibrational modes confined in the cavity center [14]. The high optical and mechanical finesse combined with the small confinement volumes enhance the couplings between excitons, photons and cavity vibrational phonons [14, 15]. Moreover, the impact of environmental disorder is suppressed by the encapsulation [16]. As a result, pristine TMD excitons with linewidths approaching the homogeneous limit are achieved and disorder-induced noise is suppressed. Therefore, even without external mechanical driving the impact of the nanobeam vibrations and their coupling to TMDs can be investigated via the measured optical properties.

In this letter we report investigations of the coupling between the vibrational modes of the nanobeam cavity and excitonic transitions of a hBN encapsulated MoS$_2$ monolayer. We observe evidence for the coupling of cavity vibrational phonons to neutral excitons $X^0$, but find that coupling to trions $X^−$ is much weaker. Due to the low energy of cavity vibrational phonons ($\sim$ GHz), the exciton-cavity-phonon coupling contributes to the linear temperature broadening of exciton linewidths [16–19]. Compared to control experiments performed on samples without cavities, this additional temperature broadening is clearly observed for $X^0$, giving rise to a $\sim$ 2× larger temperature induced broadening. However, this additional temperature broadening is not observed for $X^−$. We explain this observation as being due to the fact that the exciton-phonon coupling strength is proportional to the difference of the deformation potentials for electrons ($D_{e,cav}$) and holes ($D_{h,cav}$): Since $X^−$-cavity phonon coupling strength is proportional to $2D_{e,cav} − D_{h,cav}$ and $D_{h,cav} \sim D_{e,cav}/2$ [20–23], this leads to a strongly reduced $X^−$-cavity phonon coupling for the ribbon-shaped MoS$_2$ monolayer [23]. Furthermore, when the Raman modes of MoS$_2$ lattice phonons, e.g. $A_{1g}$ and 2LA, are tuned to be near the exciton emission energies, the exci-
ton couples to both the cavity vibrational phonons and the MoS$_2$ lattice phonons simultaneously. In this case, anharmonic effects arise from the coupling between the cavity and lattice phonons, as evidenced by the temperature broadening of Raman linewidth [10, 24]. The selective $X^{0}$-cavity phonon-lattice phonon coupling results in the enhancement of $X^{0}$-induced Raman scatterings while $X^{-}$-induced scatterings are found to be suppressed, a conclusion based on their detuning-dependent Raman intensities [25, 26] and the ratio of $X^{-}/X^{0}$ emission intensities [26]. Our work reveals the interplay between excitons, cavity vibrational modes and MoS$_2$ lattice phonons, enabling the control of phonon-induced processes in the hybrid 2D-material-cQED systems.

As depicted schematically in Fig. 1(a), the hBN/MoS$_2$/hBN heterostructures investigated were prepared using mechanical exfoliation and viscoelastic dry transfer methods [27], and features upper (lower) hBN layers that are around 15 (55) nm thick, transferred onto a 200 nm thick Si$_3$N$_4$ layer on a Si substrate. The sample was patterned into a series of photonic crystal nanobeams as described in Qian et al. [13]. We begin by discussing measurements of the lattice temperature dependent $X^{0}$ and $X^{-}$ linewidths recorded from four different positions of the hBN-encapsulated MoS$_2$ monolayer. The first case, denoted **bare-flakes** in Fig. 1(a), corresponds to the region of the sample away from the nanobeam photonic crystals that consists only of the hBN-encapsulated MoS$_2$ monolayer on the planar Si$_3$N$_4$ layer. This case is represented by the gray data. The second and third cases, denoted by the red and blue datasets in Fig. 1, correspond to data recorded from **standing** and **suspended** regions on the Si$_3$N$_4$ nanobeam. Since the photonic crystal trenches here have the periodicity of 2 $\mu$m (no confinement) and the size of 1 $\mu$m, the laser spot could be precisely and readily re-positioned on the standing and suspended regions of this sample. These three cases (bare-flakes, standing and suspended) represent the control experiments presented in this work, in contrast to the fourth case corresponding to the laser focal volume being placed directly on top of the high-Q photonic crystal cavity [13], denoted by the green data in Fig. 1 labelled **cavity**. The optical and vibrational mode of the cavity are formed by locally chirping the photonic crystal periodicity to create the bandgap confinement.

We study optomechanical phenomena originating from the cavity modes by comparing data recorded from the cavity position to the three control groups: **bare-flakes**, **standing** and **suspended**. Besides the presence of the confined optical and vibrational modes, all other factors in the cavity which might affect optical or acoustic properties, also exist in the control groups. For example, in the cavity the local static strain is induced in the MoS$_2$ monolayer from the unetched [28] or etched Si$_3$N$_4$, and the same strain exists in the standing (red) or suspended (blue) cases. Typical photoluminescence (PL) spectra recorded using a 532 nm cw-laser with a spot size $\sim 1$ $\mu$m and $\sim 30$ $\mu$W of excitation power are presented in Fig. 1(a) around 10, 96 and 162 K. In all four cases, the peak energy of the $X^{0}$ and $X^{-}$ transitions follow the hyperbolic cotangent shift with parameters consistent to

![Figure 1](https://example.com/figure1.png)

**FIG. 1.** (a) Schematic of four different laser positions used for spectroscopic studies in this work: **bare-flake** off to the side of the nanobeam, **standing** on the nanobeam but a region with Si$_3$N$_4$ below, **suspended** - a free-standing region on the etched Si$_3$N$_4$ and **cavity** - the center point of the nanobeam cavity. **Left panel:** Typical PL data recorded from these four positions around 10, 96 and 162 K. **(b) left:** $T$-dependent linewidths of the neutral and negatively charged exciton for the four detection positions, right: calculated optical and vibrational modes of the cavity together with the associated characteristic frequencies. In these vibrational modes, the MoS$_2$ deforms primarily along the axis of the nanobeam direction.
values reported in previous works [16] (Supplemental). In contrast, the linewidth of the $X^0$ peak recorded from the cavity behaves quite differently from the three reference positions: bare-flake, standing and suspended.

Generally, the $T$ dependence of $X^0$ and $X^-$ linewidth in TMD monolayers follows the phenomenological equation [16, 17, 29]

$$
\gamma = \gamma_0 + a_1 T + \frac{a_2}{\exp\left(\frac{(h\omega)_L}{k_B T}\right) - 1}
$$

(1)

where $\gamma_0$ is the intrinsic $X^0$ or $X^-$ linewidth, $a_1$ quantifies the strength of the linear temperature dependent broadening induced by the low energy acoustic phonons (given by the slope at low $T$), $a_2$ is the non-linear broadening arising from higher energy phonons with an average energy $\langle h\omega \rangle_L$, and $k_B$ is the Boltzmann constant. The temperature dependent linewidths of the $X^0$ ($\gamma_{X^0}$) and $X^-$ ($\gamma_{X^-}$) are presented in Fig. 1(b). For the bare-flake case (gray), the phonon coupling terms obtained by fitting the experimental data are $a_{1,X^0} = 18.5 \pm 1.7 \, \mu eV \cdot K^{-1}$ and $a_{2,X^0} = 29.8 \pm 5.8 \, meV$, in good quantitative agreement to previous works [16–19, 30–34]. Moreover, the average phonon energy $\langle h\omega \rangle_{L,X^0} = 23.1 \pm 5.8 \, meV$ agrees with that obtained from the hyperbolic cotangent fit of the $X^0$ energy shift (16.9±1.6 meV), within the experimental error. In the standing (red) and suspended (blue) cases, the $T$-dependence of $\gamma_{X^0}$ is also well described by Eq. 1. The best fit terms $a_{1,X^0} = 68.5 \pm 4.9 \, (77.5 \pm 3.2) \, \mu eV \cdot K^{-1}$ and $a_{2,X^0} = 91 \pm 48 \, (77 \pm 20) \, meV$ in the standing (suspended) case are around the upper limit of previous works with the strain induced [32–34]. In our samples the local static strain is induced by the Si$_3$N$_4$ substrate (standing) [28] or the suspension of the MoS$_2$ (suspended), consistent to these previous works [32–34].

In contrast to the three reference cases above, $\gamma_{X^0}$ for the detection position on the cavity exhibits different behaviour. We observe a linear temperature dependence of $\gamma_{X^0}$ up to $T = 150 \, K$, with a slope $a_{1,X^0} = 131 \pm 7 \, \mu eV \cdot K^{-1}$, almost double the value obtained for the three control measurements. This enhanced linear broadening indicates that the coupling between $X^0$ and low energy phonons is stronger for this case as compared to the three control experiments, revealing the contribution from cavity vibrational phonons. The cavity vibrational phonons have low energies, an expectation that is confirmed by finite element calculations, which revealed eigenfrequencies in the range $3.16 - 3.43 \, GHz$ for the first three modes. Typical results are presented in Fig. 1(b) (details see Supplemental). Such low-energy ($<< k_B T$) phonons contribute to the linear broadening $a_1 T$ of the exciton linewidth [16, 17].

In complete contrast to the $\gamma_{X^0}$ discussed above, the $T$ broadening of $X^-$ linewidth $\gamma_{X^-}$ reveals little difference between the four cases investigated, as presented in Fig. 1(b). In the control measurements (gray, red and blue) $\gamma_{X^-}$ is again very well described by Eq. 1 with the linear term $a_{1,X^-} = 106\pm31, \, 154\pm20, \, 114\pm8 \, \mu eV \cdot K^{-1}$. Although $\gamma_{X^-}$ in the cavity (green) cannot be appropriately described by Eq. 1, no additional temperature induced broadening beyond the control groups is observed. Based on the data presented in Fig. 1(b), we conclude that the cavity vibrational phonon couples more strongly to $X^0$ than to $X^-$. This non-trivial selective coupling can be qualitatively explained by the extended "ribbon shape" of the hBN-encapsulated MoS$_2$ monolayer in the nanobeam cavity. Hereby, the MoS$_2$ deforms primarily along the axis of the nanobeam direction. For such a situation Cai et al. [23] have calculated the nanobeam width (strain) dependent deformation potential for electrons and holes. Independent of strain, the deformation potential of holes $D_{h, cav}$ in such biaxially strained MoS$_2$ was found to be approximately twice that for electrons $D_{e, cav}$ [23]. Therefore, the $X^-$-cavity phonon coupling strength ($\times 2D_{e,cav} - D_{h,cav}$) is expected to be much smaller than the $X^0$-cavity phonon coupling strength ($\times D_{e,cav} - D_{h,cav}$) [20–22].

The temperature dependent PL results discussed above reveal that the cavity vibrational phonons influence the phonon-induced exciton linewidth broadening, a property that is entirely determined by MoS$_2$ lattice phonons in control measurements. To further investigate the interplay between excitons, cavity vibrational modes and MoS$_2$ lattice phonons, we continue by presenting Raman spectroscopy measurements. Hereby, we tune the Raman modes of MoS$_2$ lattice phonons through the $X^0$ and $X^-$ peaks (outgoing Raman resonance) by varying the lattice temperature $T$. This was done since all Raman spectroscopy measurements were performed using a 632 nm cw-laser with a spot size $\sim 1 \, \mu m$ and $\sim 100 \, \mu W$ of excitation power. These excitation conditions produce both PL from the $X^0$ and $X^-$ as well as Raman scatterings. Typical raw data measured from the bare-flake is presented in the leftmost panel of Fig. 2(a), and the extracted Raman intensity with the background PL subtracted is presented in the rightmost panel for comparison. Typical background subtracted Raman spectra measured from the cavity are presented in Fig. 2(b). We observe three dominant Raman modes: the Si$_3$N$_4$ phonon (525 cm$^{-1}$) and two MoS$_2$ lattice phonons: the acoustic phonon 2LA (450 – 480 cm$^{-1}$) and optical phonon $A_{1g}$ (409 cm$^{-1}$) [35]. The Raman intensities are normalized by dividing the integrated Raman peak intensity by the Bose factor and the Si$_3$N$_4$ peak, i.e.

$$
I'_{A_{1g}} = \frac{I_{A_{1g}}}{I_{SiN}/(n_{SiN} + 1)}, \quad I'_{2LA} = \frac{I_{2LA}}{I_{SiN}/(n_{SiN} + 1)}
$$

where $I_{A_{1g}}, I_{SiN}$ and $I_{2LA}$ are peak intensities extracted from Raman spectra, and $n_{A_{1g}}, n_{SiN}, n_{LA}$ are the $T$-dependent Bose distribution factors. Theoretically, the normalized Raman intensity is determined by the integral...
of all possible intermediate states described by [36]

$$\sum_{m} \left| \frac{M_{fm} M_{mp} M_{mi}}{(\omega_{m} - (i/2)\gamma_{m} - \omega_{laser}) (\omega_{m} - (i/2)\gamma_{m} - \omega_{p})} \right|^2$$

where $i, m, f$ denote the initial, intermediate and final states, $\omega_{laser}$ ($\omega_{p}$) is the energy of the laser (Raman mode), $\omega_{m}$ and $\gamma_{m}$ are the energy and lifetime of the intermediate states $m$, $M_{fm}$ ($M_{mi}$) is the matrix element of the optical $f \rightarrow m$ ($m \rightarrow i$) transition, and $M_{ep}$ is the matrix element characterising exciton-phonon coupling. In our measurement, the outgoing Raman lines are near resonant to $X^0$ and $X^-$, thus their detuning plays the major role in the variation of Raman intensities. Therefore, we approximately fit the normalized Raman intensities using

$$I'_p = \sum_{X} g_{X,p} R_{X,p}, X = \{X^0, X^-\}, p = \{A_{1g}, 2LA\}$$

$$R_{X,p} = 1/|\omega_X - (i/2)\gamma_X - \omega_p|^2$$

where $g_{X,p}$ reflects the exciton-phonon coupling strength and $R_{X,p}$ is the exciton-Raman detuning modelled by the Fermi’s golden rule [25, 37]. The exciton energy $\omega_X$ and linewidth $\gamma_X$ are all extracted from the PL spectra, and the Raman mode energy $\omega_p$ is extracted from the Raman spectra. The normalized Raman intensities of the $A_{1g}$ and 2LA modes are plotted in Fig. 2(c) and (d), respectively, as a function of the detuning to $X^0$. The Raman intensities recorded from the bare-flake (gray), standing (red) and suspended (blue) control experiments are compared with results obtained from two nanobeam cavities having widths 520 nm (C2) and 420 nm (C12). Raman data was obtained on several different cavities (see Supplemental), all of which were qualitatively similar to the measurements presented in Fig. 2(c) and (d). A clear resonance is observed for $A_{1g}$ at the energy of $X^0$ peak for the two-cavity measurements, as shown by the dashed lines on the figure. In comparison, the dotted line on the figure represents the expected resonance arising from coupling to $X^-$, $\sim 35$ meV lower in energy than $X^0$ [25, 26]. In the three control groups (gray, red, and blue) the coupling strength $g_{X,p}$ is nearly constant [25, 37, 38], and the fitting reveals that for $A_{1g}$ only $g_{X^-, A_{1g}}$ has a significant amplitude, while $g_{X^0, A_{1g}}$ vanishes. In contrast, for 2LA both $g_{X^0, 2LA}$ and $g_{X^-, 2LA}$ have finite amplitudes. Another distinctive feature of the $X^-$-phonon coupling is the clear enhancement of the relative PL intensity of $X^-$ compared to $X^0$ around the $X^-$-Raman resonance [26]. This can be clearly observed in all three control measurements presented in Fig. 2(d) in which spectra recorded
at \( \sim 65\text{K} \) using a 632nm-laser (close to \( X^-\)–Raman resonance) are compared with reference data recorded using a 532nm-laser (far away from \( X^-\)–Raman resonance). This enhancement of \( X^- \) arises since \( X^-\)-phonon coupling assists the \( X^0 / X^- \) conversion via a doubly resonant Raman scattering process, as depicted schematically in the inset of Fig. 2(d) [39–41].

We note that ingoing resonant Raman effects have little impact on the observed Raman intensities in our experiments, since the laser is far detuned from both excitonic peaks (\( X^0\)-Laser resonance corresponds to 60 meV in \( X^0\)-Raman mode detuning). Moreover, here \( A_{1g} \) and 2LA both contain doubly resonant Raman scatterings [37–39], and thereby the process leading to the Raman signal is not limited to phonons at the Brillouin zone center, but also involves a larger region of the vibrational phase space. Therefore, we approximately fit the experimental data in Fig. 2(c) and (d) via the outgoing resonance \( R_{X,p} \) which plays the major role in the detuning dependent Raman intensity. Nevertheless, we emphasize that the major conclusions reached here can still be obtained from the raw data directly, without making any approximations or quantitative fitting of data. Detailed discussions on the role of the excitation (ingoing) and additional qualitative analysis of raw data can be found in the Supplemental.

The Raman spectra of our control measurements generally agree well with previous works [25, 26]. In contrast, the Raman spectra recorded from the nanobeam cavities exhibit entirely different behavior that can be traced to the presence of the cavity vibrational mode. As shown by the position-dependent spectra in Fig. 2(b), the Raman intensities are largest at the cavity center in accord with our expectation that confined optical and vibrational modes of the cavity (both maximum at the midpoint of the nanobeam cavity) are not excluded by control groups. The normalized intensities of \( A_{1g} \) and 2LA from the cavity C2 (green) and C12 (purple) are presented in Fig. 2(c) and (d), respectively. As shown in Fig. 2(c), \( g_{X^0,A_{1g}} \) is clearly non-zero as evidenced by the resonance in the Raman intensity (dashed peak), a resonance that is completely absent in the control groups. In comparison, the data for 2LA shown in Fig. 2(d) shows that \( g_{X^0,2LA} \) (dashed peak) is finite for the cavities whilst the amplitude \( g_{X^-,2LA} \) (dotted peak) vanishes. Thus, we conclude that the \( X^0 \)-phonon coupling strengths \( g_{X^0,p} \) \((p = \{A_{1g},2LA\})\) in the nanobeam cavities are enhanced whilst the \( X^-\)-phonon coupling strengths \( g_{X^-,p} \) are suppressed. The suppression of \( X^-\)-phonon coupling is further supported by the \( X^-\) emission intensity presented in Fig. 2(d). The \( X^-\) emission enhancement [26] arising from coupling to phonons (Fig. 2(d inset), is not observed in the cavity.

We continue to demonstrate that, in contrast to the constant exciton-phonon coupling strengths \((g)\) observed in control measurements, the cavity enhanced value of \( g_{X^0,A_{1g}} \) or \( g_{X^0,2LA} \) follows a temperature dependent power law \( (T^N) \). Fig. 3(a) shows the temperature dependence of \( R_{X^0,2LA} = I_{2LA} / I_{X^0,2LA} \) recorded from the midpoint of the cavities C2 (green curve) and C12 (purple curve). The selective enhancement of \( g_{X^0,A_{1g}} \) or \( g_{X^0,2LA} \), and the \( T^N \) dependence are two key observations in the cavities, indicating that the excitonic transitions couple to both lattice phonons and the cavity vibrational phonons. This type of exciton-phonon-phonon coupling we expect are illustrated in the inset of Fig. 3(a), where the cavity vibrational phonons provide additional intermediate states (gray replicas) that provide resonance conditions for the scattering of MoS\(_2\) lattice phonons (arrows). Similar vibronic sub-level mediated processes have been reported in photonic [42, 43] and plasmonic systems [44, 45]. Before further discussion, we first exclude other possibilities. As discussed in relation to Fig. 1(a) and 2(b), all other factors besides the optical and vibrational modes of the cavity are excluded by the three control measurements performed. We note that the resonant cavity optical mode could also enhance Raman intensity by increasing the local field amplitude within the MoS\(_2\) monolayer [46, 47] or exciton spontaneous emission rate [44, 47, 48]. However, these effects are expected to be strongly dependent on the detuning of the laser and exciton transitions from the cavity mode. In our experiments, both the excitation laser and observed Raman modes are far detuned from the cavity optical mode (> 50 meV), and the detuning is nearly constant with \( T \). At the same time the coupling between \( X^0 \) and the cavity optical mode is expected to be small [13]. Therefore, we do not believe that the cavity optical mode plays a major role here in determining the temperature dependence of \( g_{X^0,A_{1g}} \) or \( g_{X^0,2LA} \) and we conclude that the observed results are fully consistent with coupling to the cavity vibrational mode.
Indeed, the exciton-phonon-phonon coupling between excitons, cavity vibrational phonons and MoS$_2$ lattice phonons (Fig. 3(a) inset) well explains both the selective enhancement of X$^0$-induced Raman scattering and the power temperature dependence of the Raman cross section. Hereby, the intermediate vibronic states (gray replicas) depicted in the inset of Fig. 3(a) would only be expected to occur for X$^0$ since our findings (Fig. 1) show that only X$^0$ couples significantly to cavity vibrational phonons. Meanwhile, the $T^N$ dependence is consistent with the Bose occupation of cavity vibrational phonons $1 + n_{vP} \approx n_{vP} \propto T$, and $N$ would be determined by the number of cavity vibrational phonons mediating the resonant Raman scattering process. Finally, additional supporting evidence for the exciton-phonon-phonon coupling is observed from the $T$-dependent Raman linewidth, where the phonon-phonon coupling (anharmonicity) between cavity and lattice phonons introduces a $T$-broadening [10, 24]. Figure 3(b) presents the Raman linewidth of one 2LA peak extracted from multi-Lorentz fitting (details in Supplement). Compared to the constant Raman linewidth observed for the bare-flake (gray), the $T$-broadening in the cavity (purple) is observed with the slope $4.8 \pm 1.8 \mu$eV $\cdot$ K$^{-1}$, demonstrating the phonon-phonon coupling between the cavity vibrational phonons and MoS$_2$ lattice phonons. In addition, we also observe the non-monotonic Raman linewidth behaviour from the exciton-phonon coupling [24] for A$_{1g}$, but present the results in Supplemental for brevity.

In summary, we demonstrated coupling between neutral exciton X$^0$, cavity phonon and lattice phonon modes in the cavity-MoS$_2$ system. Compared to control groups without cavities, an additional temperature broadening of X$^0$ linewidth was observed from the PL spectra, and an enhancement of X$^0$-induced scatterings was observed from the Raman spectra. In contrast, the X$^-$-cavity phonon coupling was shown to be much weaker, resulting in the suppression of X$^-$-induced Raman scattering in cavities. Usually, these phonon-induced processes are determined by lattice phonons in the 2D semiconductors. Our results illustrate how the interplay between excitons, cavity vibrational phonons and MoS$_2$ lattice phonons enables control of phonon-induced processes in nanophotonic and nanomechanical systems. In addition, since our results are observed with the natural nanobeam vibrations without external mechanical driving, similar optomechanical phenomena would intrinsically exist in most 2D-material-based systems with potential for future applications. One recent example [13] includes the control of free exciton transport via phonon couplings, illustrating how fundamental properties of light-matter interactions in 2D materials can be affected by the phonon-induced processes.

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Supplementary Information for Selective Exciton-Phonon-Phonon Coupling and Anharmonicity with Cavity Vibrational Phonons and MoS$_2$ Lattice Phonons in Hybrid Nanobeam Cavities

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SFig. 1. Design and FEM simulation of the cavity. (a) Schematic of cavity structures [1]. The whole nanobeam has the length $l_x = 20 \ \mu m$, width $d_y = 500 \ \text{nm}$ and depth $d_z = 250 \ \text{nm}$. Nanotrenches have length $h_x = 120 \ \text{nm}$ and depth $h_z = 150 \ \text{nm}$, following a Gaussian spatial distribution with the separation between trenches $a_i/a = 1 - A \cdot \exp(-i^2/(2\sigma^2))$. $a = 250 \ \text{nm}$ is the lattice constant and $A = 0.1, \sigma = 4$ define a smoothly varying photon and phonon confinement. Since mechanical properties of hBN are not well known yet, here we set all material as Si$_3$N$_4$ in the calculation. (b) The cavity phonon modes. Energy unit is GHz. First three lowest energy modes are already plotted in Fig. 1 in the main paper. In addition, there are also phonon modes from the whole nanobeam as shown in SFig. 10, which have huge mode volume thus their coupling to excitons is much weaker than the confined phonon mode here.

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Fig. 2. PL spectra and fittings. (a) T-dependent PL spectra and (b) multi-Lorentz fittings in the suspended case as examples. Peaks from neutral exciton X⁰, trion X⁻, dark trion X⁻,D [2] and localized excitons LX (2peaks) are observed. (c) X⁰ energy shift is well fitted with standard hyperbolic cotangent relation [3]. For the four cases, S is 1.52 ± 0.06, 1.87 ± 0.06, 1.89 ± 0.04, 1.88 ± 0.06, and ⟨ℏω⟩K is 16 ± 2, 23 ± 1, 22 ± 1, 24 ± 1 meV. (d) X⁻ energy is also well fitted. For the four cases, S is 1.37 ± 0.10, 1.69 ± 0.08, 1.88 ± 0.08, 1.68 ± 0.07, and ⟨ℏω⟩K is 14 ± 3, 21 ± 2, 23 ± 2, 22 ± 2 meV. Little difference is observed in the energy shift between different cases, and all fitting parameters agree well to previous works. (e) X⁰ linewidth and (f) X⁻ linewidth, of which the low T data are already plotted in Fig. 1 in the main paper. The suppression of γX⁰ broadening at T > 200 K may originate from the saturation of phonon couplings or other complex dynamics which need further investigations. The fitting accuracy of γX⁻ at high T is limited by the merge of X⁻ and X⁻,D peaks as shown in (b) inset.
Sfig. 3. Subtraction and fitting of Raman spectra. (a) One example of the raw spectrum (black) and the corresponding Raman spectrum (red) after subtracted with the emission baseline (blue). (b) Multi-Lorentz fitting. The intensity of 2LA discussed in the main paper is the sum of two peaks (blue and green). Besides the A_{1g} and 2LA, some other known Raman modes reported previously are also labelled here. However, these modes are too weak when they are far detuned from X^0 and X^-, thus are not discussed in this work.

Sfig. 4. T-dependent phonon energy shifts and linear fittings, agreeing well with freedoms in volume thermal effects. (a) Schematic of sample structures. (b) The A_{1g} mode in the MoS_2 monolayer. (c) The Si_3N_4 Raman mode. T-dependent phonon energy shift is related to volume thermal coefficient, where more freedom of volume thermal expansion results in slower energy shift [4]. In the four cases from left to right in (a), this freedom of MoS_2 or Si_3N_4 increases, agreeing well to the experimental energy shift rate in (b)(c). Since the phonon energy shifts are consistent to previous theories and not related to the focus of this work, we did not discuss them in the main paper.
SFig. 5. Additional Raman results on the cavity sample. The 2D flakes for the control group sample and cavity sample are from the same bulk materials, by the same exfoliation and transfer method. Nevertheless, there might be wonderings that the differences between cavities and control groups are from uncertainties during the fabrication. Here we show the Raman results measured from the bare flake on the cavity sample but outside the cavity (gray), and another cavity C7 (yellow). The key results (peaks) of the bare flake on the cavity sample are same to the bare flake on the control group sample shown in Fig. 2 in the main paper, and the results of cavity C7 are also same to that of C2,12 in the main paper. These agreements exclude the uncertainties during the fabrication.

SFig. 6. Qualitative analysis of experimental data. As discussed later in SFig. 7, the Raman scatterings are complex and we use approximation to quantitatively fit experimental data as presented in Fig. 2 in the main paper. Here we emphasize that, our conclusions do not rely on the quantitative fitting, and can be directly obtained from the raw data. Generally, we obtain two curves in the detuning-dependent Raman intensities, shown by the dashed line and dotted line. The dashed line is surely from the $X^0$-phonon detuning. By comparing the cavity and the control group, the enhancement of dashed line is clearly observed. Therefore, the first conclusion that the $X^0$-phonon coupling is enhanced in the cavity, is achieved. There might be wonderings about the dotted line. E.g., the dotted line in (b), surely contains the contribution from the $X^0$-2LA detuning, but might also contains contributions from the $X^0$-LA or $X^0$-Laser detuning. Nonetheless, we do not need to care how much percentage is from which detuning, because the whole dotted line vanishes in the cavity which means all contributions become zero. Therefore, the secondly conclusion that the $X^-$-phonon coupling is suppressed, is also achieved.
SFig. 7. Schematic of Raman scatterings and discussions about ingoing resonance. (a) $I'_{A1g}$ on the bare flake which is also plotted in Fig. 2(c) in the main paper. By comparison $R_{X^-, A1g}$ (gray line) is the best approximation. (b) The ideal first-order Raman scattering. Due to the scattering limited at zone center, the Raman intensity follows $R_{X^0, A1g}$ and $R_{X^0, Laser}$ (dark yellow line in (a)). Obviously, this is not the case in our work. Firstly, $R_{X^0, A1g}$ and $R_{X^0, Laser}$ involves two sections (ingoing and outgoing) in one process. Thus, it is contradictory that $R_{X^0, Laser}$ exists while $R_{X^0, A1g}$ vanishes. Even if only considering $R_{X^0, Laser}$ (cyan line in (a)), the variation is relatively small due to the large $X^0$-Laser detuning thus cannot fit the experimental data. (c) The phonon-assisted $X^0/X^-$ conversion [5]. The outgoing section (downward solid arrow) is around the zone center thus follows $R_{X^-, A1g}$. In contrast in the ingoing section (upward solid arrow), the $X^0$ created by the laser is not at the valley center, thus does not follow $R_{X^0, Laser}$. Indeed, the ingoing section creates one $X^0$ with the laser energy $\omega_{Laser}$ at the real energy level. Therefore, we think this probability is dominated by $1 + n_{Laser}$ since excitons in the valley follows the Bose distribution $n_{Laser}$ [6]. In our experiment, $\hbar \omega_{Laser} = \hbar \omega_X > \kappa T$, thereby $1 + n_{Laser} \approx 1$ is approximately constant. As shown in (a), $R_{X^-, A1g}$ fits the experimental data better than $R_{X^0, A1g}$ and $R_{X^0, Laser}$ (orange line). (d) The second-order 2LA scattering [7]. Strictly, the Raman intensity in (c) and (d) is very complex with the integral over all possible intermediate states. But as discussed in the main paper, in our measurement range the outgoing section is around resonance, thus $R_{X^0}$ plays the major role in the Raman intensity variation. Indeed, this approximation achieves good agreements between Raman and PL spectra. The fitting $I^{2LA}_X = g_{X^0, 2LA} R_{X^0, 2LA} + g_{X^-, 2LA} R_{X^-, 2LA}$ in Fig. 2(d) in the main paper gives the exciton-phonon coupling strength $g_{X^0, 2LA}$ and $g_{X^-, 2LA}$. The PL spectra in Fig. 1 in the main paper also gives the coupling strength between excitons and acoustic phonons $a_{1, X^0}$ and $a_{1, X^-}$. The $g_{X^0, 2LA}/g_{X^-, 2LA}$ 5.1, 2.0 and 1.5 (bare flake, standing and suspended) quantitatively agree with $a_{1, X^0}/a_{1, X^-}$ which are 5.7, 2.2, and 1.5 correspondingly. This agreement further supports our fitting a valid approximation of the complex Raman scatterings.
Fig. 8. Raman intensities $I_{\text{A}_1g} / (n_{\text{A}_1g} + 1)$ in (a) $I_{\text{2LA}} / (n_{\text{2LA}} + 1)^2$ in (b) without the normalization to the Si$_3$N$_4$ peak. Compared to normalized intensities in Fig. 2(c)(d) in the main paper, the two key results in the cavity including the selective enhancement of $X^0$-phonon coupling and the $T_N^N$ dependence of the enhancement are the same. We note that, the theoretical laser spot size is $1 \mu m$. In contrast, the nanobeam cavity width is $< 0.5 \mu m$. Thus, it makes no sense to compare the absolute intensity between difference cases. This is why we normalized to the Si$_3$N$_4$ peak in the main paper, which provides the comparison between different cases and further helps removing potential noises from experimental variations [8]. Nonetheless, as shown here, the key conclusions do not rely on the normalization.
SFig. 9. Linewidth of Raman modes. (a)(b) $A_{1g}$ linewidth in control groups (a) and cavities (b). Solid lines are the the quadratic fitting. Data at high $T > 170$ K has huge error bar (hollow points) from the low signal to noise (SNR) of $A_{1g}$ peak in raw data. Due to the exciton-cavity-phonon-lattice-phonon coupling discussed in the main paper, the non-monotonic linewidth [9] is significant in the cavities (b) compared to the control groups (a). (c)(d) Linewidths of two 2LA peaks (SFig. 3(b)) and the linear fittings (solid lines). The SNR of the 2LA peak is low at low $T$ (hollow points). The flake (gray) and C12 (purple) case in (d) are already plotted in Fig. 4(b) in the main paper. Through the linear fittings (solid lines), the broadening of linewidths which reflects the phonon-phonon coupling is clearly observed in cavities. (f) Raw Raman spectra normalized by the 2LA peak, where the $T$ broadening of the linewidth in the cavity is clearly observed compared to the control group.
SFig. 10. Some series vibration modes of the whole nanobeam. Frequency unit is GHz. These modes have huge mode volumes.
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