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# Control of coherently coupled exciton-polaritons in monolayer tungsten disulphide

Xiaoze Liu<sup>1†</sup>, Wei Bao<sup>1†</sup>, Quanwei Li<sup>1</sup>, Chad Ropp<sup>1</sup>, Yuan Wang<sup>1,2</sup>, Xiang Zhang<sup>1,2\*</sup>

<sup>1</sup>*NSF Nanoscale Science and Engineering Center, University of California, Berkeley, California  
94720, USA.*

<sup>2</sup>*Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California  
94720, USA.*

<sup>†</sup> These authors contributed equally to this work.

\* Email: [xiang@berkeley.edu](mailto:xiang@berkeley.edu)

**Abstract:** Monolayer transition metal dichalcogenides (TMD) with confined 2D Wannier-Mott excitons are intriguing for the fundamental study of strong light-matter interactions and the exploration of exciton-polaritons at high temperatures. However, the research of 2D exciton-polaritons has been hindered, because the polaritons in these atomically thin semiconductors discovered so far can hardly support strong nonlinear interactions and quantum coherence due to uncontrollable polariton dynamics and weakened coherent coupling. In this work, we demonstrate, for the first time, precisely controlled hybrid composition with angular dependence and dispersion-correlated polariton emission by tuning the polariton dispersion in TMD over a broad temperature range of 110-230 K in a single cavity. This tamed polariton emission is achieved by the realization of robust coherent exciton-photon coupling in a monolayer tungsten disulphide (WS<sub>2</sub>) with large splitting-to-linewidth ratios (SLR, >3.3). The unprecedented ability to manipulate the dispersion and correlated properties of TMD exciton-polariton at will offers new possibilities to explore important quantum phenomena such as inversionless lasing, Bose-Einstein condensation (BEC), and superfluidity.

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Two-dimensional (2D) semiconducting transition metal dichalcogenides (TMD) have been an attractive group of semiconductors since the discovery of the transition from indirect to direct band gap at the monolayer limit [1–3]. Due to their quantum confinement at reduced dimensionality and crystal symmetry, the TMD monolayers have very large exciton binding energies up to  $\sim 0.7$  eV [4–6], huge oscillator strengths [1,7], and the valley degree of freedom [8–11]. These exceptional properties bolster the formation of exciton-polaritons in optical cavities [12–19] with a great potential for applications in quantum information, communication, and computing. In semiconductor microcavities (MC), the part-light part-matter polaritons are formed in the strong coupling regime where the coupling rate is faster than the average dissipation rates of light and matter. When a distinct polariton energy dispersion is present, these bosonic quasiparticles can follow the Bose-Einstein statistics with small effective masses, enabling Bose-Einstein condensation (BEC) at elevated temperatures [20–22].

The realization of polariton condensation at higher temperatures requires excitons that are robust to thermal fluctuations, coherent coupling between the exciton and photon that are maintained under high polariton densities, and polariton dynamics that are optimized through a well-balanced hybrid composition within a distinct energy dispersion (such as the angular dependent energy diagram). In this context, TMD monolayers, that provide delocalized (Wannier-Mott) excitons with large binding energies and distinct valley degree of freedom [12,23], are a unique semiconductor system for room temperature polariton condensation with optical spin degree of freedom. However, the research of TMD exciton-polaritons has been hindered by (1) the weakened coherent coupling, indicated by small splitting-to-linewidth ratios (SLR), and (2) uncontrolled and extremely fast polariton dynamics. The coherent coupling is represented by SLR, i. e., the average number of energy oscillations

between excitons and photons during polariton lifetime [24](see [25] with more details). More importantly, to maintain unperturbed coherent coupling at high pump density, support strong nonlinear polariton interactions and slow down the polariton leakage rate with respect to the nonlinear interaction rate, a large SLR is essential for polariton condensation [24]. After the first demonstration of exciton-polaritons in a MoS<sub>2</sub> monolayer-based MC [12], various approaches [13–18] have been employed for further polariton study, but with limitations on reduced Rabi splitting, low temperature operation (< 20 K), and small SLR (typically < 2). These limitations prevent coherent strong coupling at high pump injection, which is the key for room temperature polariton condensation. On the other hand, polariton condensation strongly depends on the optimized dynamics via the well-balanced hybrid composition in the energy dispersion diagram, which holds only when the exciton component has a significantly slower decay rate than the photon leakage rate. Yet, the TMD exciton decay process, involving defect-mediated non-radiative recombination [26], trion [27,28] and dark exciton [29] quenching at low temperatures, is ultrafast (typically <10 ps) and comparable with the photon leakage lifetime. Hence, the optimized polariton dynamics in TMD has not been demonstrated. In this work, we show that the angular dependent hybrid composition with a full dispersion is, for the first time, controlled by directly tuning the TMD exciton energy over a broad temperature range (110-230 K), where coherently coupled TMD exciton-polaritons with a large SLR > 3.3 are sustained. Due to the enhanced long exciton lifetime (> 200 ps), the angular dependent intensity distribution of polariton PL reveals a clear correlation with the hybrid composition of excitons and photons as a function of temperature. This correlation provides a distinctive basis for control of polariton dynamics by tuning of the hybrid composition in a single cavity device, which is in contrast to the requirement that samples be varied in conventional semiconductor systems to span polariton

configurations [22,30–34]. Our findings demonstrate an ideal platform for manipulating the TMD polaritons at will and open the door towards realization of 2D polariton condensation at room temperature.

To achieve a large SLR for strongly coupled exciton-polaritons we introduce an efficient strategy to incorporate monolayer tungsten disulphide ( $\text{WS}_2$ ) into a compact all-dielectric MC. Coherent coupling with a larger SLR requires small cavity length to increase Rabi splitting, and a larger quality factor to reduce polariton linewidth. The cavity structure is shown in the schematic of Fig. 1a, where the bottom mirror is 12.5 pairs of a  $\text{SiO}_2/\text{Si}_3\text{N}_4$  distributed Bragg reflector (DBR) while top mirror is 7.5 pairs of a  $\text{SiO}_2/\text{Si}_3\text{N}_4$  DBR. Since the monolayer  $\text{WS}_2$  exciton is fragile to the fabrication process [3,26,35], we utilize hydrogen silsesquioxane (HSQ) to form a sandwich structure around the exfoliated monolayer  $\text{WS}_2$  (HQ Graphene, Inc) that preserves the electronic and optical quality of TMD monolayers [36,37]. An  $\text{Al}_2\text{O}_3$  layer is grown on top of capping HSQ to further protect the  $\text{WS}_2$  layer from the high temperature plasma during growth of the top DBR. The monolayer area is shown in the microscopic image of Fig. 1b inset. The HSQ thickness is controlled to achieve a MC cavity resonance at 2.052 eV for normal incidence (the in-plane wavenumber  $k_{\parallel}=0$ ), determined by the reflectivity (see [25] Fig. S1). The cavity resonance needs to overlap with the exciton energy over a broad temperature range with only negligible influences from the trions and dark excitons, in order to demonstrate the clear feature of coherently coupled exciton-polariton and the control of polariton dispersion as a function of detuning (determined by temperature). Based on the normal cavity resonance  $E_{ph}$  at 2.052 eV, the cavity shows angular dependence of  $E_{cav}(\theta) = E_{ph} / \sqrt{1 - (\sin(\theta) / n_{eff})^2}$  as in Fig. S1(a). Moreover, at the normal incidence the total quality factor of the MC is measured to be  $\sim 400$ , corresponding to a cavity a full width half maximum (FWHM) of  $\sim 5\text{meV}$ . Our strategy thus ensures the

excitonic performance of WS<sub>2</sub> is boosted without sacrificing the cavity quality factor or expanding the cavity length.

The exciton linewidth and decay process can not only affect the coherent coupling, but also modify the polariton dynamics. The excitonic performance can be characterized by the exciton PL measurements. PL spectra of WS<sub>2</sub> monolayer measured with different substrates and capping conditions (Fig. 1b), along with time-resolved PL of WS<sub>2</sub> on HSQ (Fig. S2), confirm that HSQ enhances the excitonic performance. Compared with the PL spectra of WS<sub>2</sub> on substrates of SiO<sub>2</sub>/DBR and on HSQ/DBR, the PL of WS<sub>2</sub> sandwiched by two HSQ layers shows a peak intensity that is increased by a factor of >10 and a lifetime ~220 ps at low pump fluence, corresponding to a PL quantum efficiency of ~10%. Moreover, the PL spectra all show a consistent dominant excitonic peak at ~2.006 eV with FWHM of ~30 meV. Note here that the observed PL quantum efficiency is still within the optical linear regime and stays unchanged up to a pump fluence of 0.1 μJ/cm<sup>2</sup> (Fig. S2), different from the reported chemically treated monolayer MoS<sub>2</sub> [26] whose high quantum efficiency becomes suppressed at pump fluences > 0.01 μJ/cm<sup>2</sup> due to the Auger recombination. With the HSQ capping layers, the WS<sub>2</sub> excitons preserve their apparent temperature-dependent shift (from 2.006 eV around 295 K to 2.085 eV around 10 K) as shown in Fig. S3, distinct from other temperature-insensitive cases [38,39]. Due to the emergence of trions [27] and dark excitons [29] at low temperature, our MC sample is only studied at temperatures > 110 K, where the exciton peak dominates.

We reach the strong coupling regime by first cooling down the MC to 110K where the cavity detuning is  $\Delta = E_{ph} - E_{ex} = -26$  meV. The k-space (angle-resolved) reflectivity is then characterized using white light as shown in Fig. 2a. More details of k-space characterization is discussed in Supplementary Information (SI) and Fig. S4 in SI, and the data shown here is based

on TM polarization unless otherwise noted. Two reflectivity modes appear with the higher-energy mode's dispersion flattened at small incidence angles and the lower-energy mode's dispersion flattened at larger angles. This is a typical signature of a two-branch polariton dispersion, indicating that the system is in the strong coupling regime. Note here the polariton states are the superposition of original cavity mode and exciton resonance, so they show a very distinct angular dependence with apparently anti-crossed branches from the original cavity dispersion (Fig. S1) and flat exciton dispersion (Fig. S5). The cavity photon dispersion is shown as the blue dashed curve while the exciton energy, at 2.078 eV, is shown as the red dashed line, which is based on the temperature-dependent PL (Fig. S3). The FWHM of the polariton branches (identified by the magenta curves) are both less than 12 meV, with the Rabi splitting read directly as  $\sim 40$  meV at  $\sin(\theta)$  of 0.25, yielding a high SLR  $> 3.3$ . This large SLR is clear from the splitting feature without peak fitting, demonstrating sufficient round-trip energy oscillations during the exciton-photon coupling and thus obvious coherent coupling.

This Rabi splitting is consistent with a coupled oscillator model as

$$\begin{pmatrix} E_{ex} + i\hbar\Gamma_{ex} & V_A \\ V_A & E_{cav}(\theta) + i\hbar\Gamma_{cav} \end{pmatrix} \begin{pmatrix} \alpha \\ \beta \end{pmatrix} = E \begin{pmatrix} \alpha \\ \beta \end{pmatrix}. \quad (1)$$

$E_{ex}$  and  $E_{cav}$  are the exciton and cavity photon energies respectively. And the  $\Gamma_{cav}$  and  $\Gamma_{ex}$  are the half width half maximum (HWHM) of cavity photon and exciton, respectively.  $E$  are the eigenvalues corresponding to the energies of polariton modes.  $\alpha$  and  $\beta$  construct the eigenvectors where  $|\alpha|^2 + |\beta|^2 = 1$ .  $V_A$  is the coupling strength. This leads to the

eigenvalues  $E = \left(\frac{E_{ex} + E_{cav}}{2}\right) + i\left(\frac{\hbar\Gamma_{ex} + \hbar\Gamma_{cav}}{2}\right) \pm \sqrt{V_A^2 + \frac{1}{4}(E_{ex} - E_{cav} + i\hbar\Gamma_{ex} - i\hbar\Gamma_{cav})^2}$  where the

Rabi splitting  $\hbar\Omega_{\text{Rabi}} = 2\sqrt{V_A^2 - \frac{1}{4}(\hbar\Gamma_{\text{ex}} - \hbar\Gamma_{\text{cav}})^2}$  at detuning  $\Delta = E_{\text{cav}} - E_{\text{ex}} = 0$ . More details of this model are also elaborated in the SI.

To fit this model with the data, the reflectivity minima are extracted from the k-space reflectivity and the calculated polariton dispersion is obtained, which overlays exactly with the data. Specifically, at the zero detuning where polariton linewidth is around 12 meV, the difference of HWHM between excitons and cavity photons are estimated to  $\sim 7$  meV, the Rabi splitting of 40 meV yields a coupling strength  $V_A$  of 20.3 meV.

To show how the polariton states are populated by scattering and thermal relaxation from the non-resonantly pumped exciton reservoir, the MC is non-resonantly pumped by a pulsed laser with a wavelength of 510 nm, which lies just outside the reflectivity stopband of the MC (see [25] for PL measurement details). In Fig. 2b, the k-space PL map shows a dominant lower polariton (LP) PL intensity with a maximum peak at the lowest polariton state. The upper polariton (UP) intensity is much weaker than the LP, so the PL intensity at higher photon energy panel is magnified by 10 times to be visible in the figure. The dispersion plot overlaid with this PL map is directly translated from the reflectivity dispersion without any fitting. As clearly shown, the dispersion curves are very consistent with the PL distribution, confirming that the polariton states are stable after the scattering and thermal relaxation in this non-resonantly pumped process. Moreover, the large contrast of PL intensity between UP and LP states indicates a more photon-like LP, with faster decay rate, as well as the most photon-like LP state (lowest LP state) with fastest rate, uncovering a hybrid composition-determined polariton dynamics.



If the polariton decay rate is slower than the polariton relaxation process, it can be determined by the exciton and photon rates with weighing fraction (i. e., Hopfield coefficient, see [25]), varying along the dispersion as:

$$\begin{aligned}\gamma_{LP} &= |\alpha|^2 \gamma_{ex} + |\beta|^2 \gamma_{cav} \\ \gamma_{UP} &= |\beta|^2 \gamma_{ex} + |\alpha|^2 \gamma_{cav}\end{aligned}\tag{2}$$

where  $|\alpha|^2, |\beta|^2$  represent weighing fractions of exciton and cavity photon respectively;  $\gamma_{LP}, \gamma_{UP}$  refer to the decay rates of LP and UP states respectively;  $\gamma_{ex}, \gamma_{cav}$  represent the decay rates of exciton (lifetime  $\sim 200$  ps, Fig. S2) and cavity photon (lifetime  $\sim 1$  ps), respectively. Due to the much faster cavity decay rate, polaritons with larger photon fraction will decay faster, resulting in a varying emission intensity distribution that depends on the hybrid composition.

The exciton-polariton composition, as a vital physical quantity for controlling the nonlinear interactions in polariton condensates [32,40], can be directly managed through cavity-excitation detuning ( $\Delta = E_{ph} - E_{ex}$ , see [25]). This detuning is most often achieved through cumbersome modification of the photon part [22,30–34] due to the difficulty in controlling the exciton part. However, TMD monolayers have Wannier-Mott excitons with large binding energies [4] that enable the tuning of the exciton part over a large temperature range. This strong temperature dependence is extremely difficult to achieve with other material systems because of the weak exciton binding energies in the conventional quantum well systems, the multi-exciton convolutions in wide-bandgap semiconductors, as well as the temperature-insensitivity of Frenkel excitons in organic materials. Here we demonstrate, for the first time, the tuning of the exciton part of the polariton as a function of temperature to control the cavity detuning in a single MC sample. The sample temperature is set to be 130K, 210K and 230K, corresponding to the detuning of -20meV, 2meV and 10meV, respectively. In Fig. 3a, the exciton dashed line red-

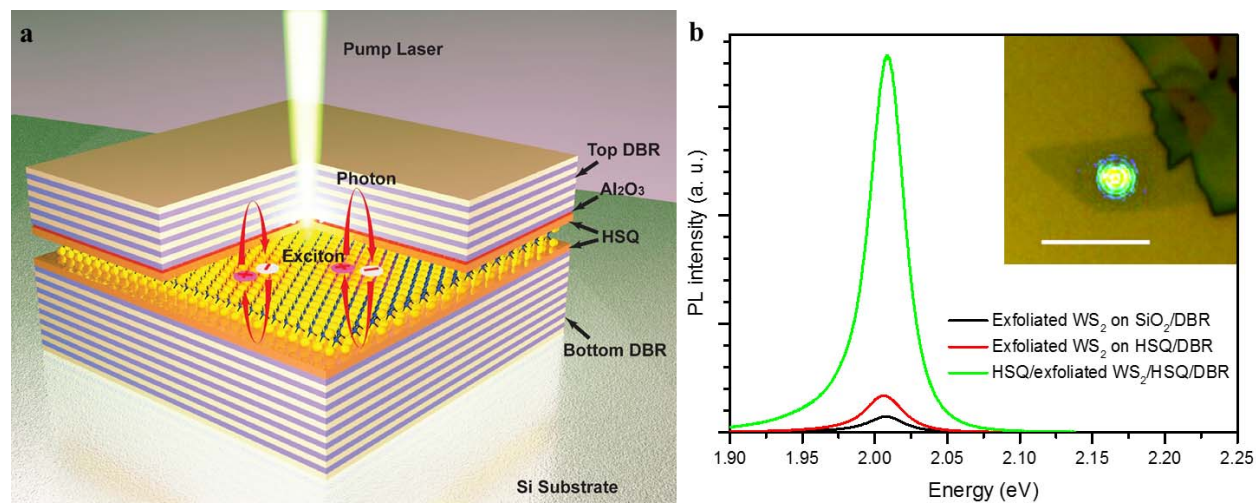
shifts as the temperature increases while the cavity resonance stays almost the same, creating the crossing and non-crossing features in the exciton and original cavity dispersion. The polariton branches, though modified significantly due to the various cavity detunings, are both unambiguously identified based on the anti-crossing reflectivity modes. Using the same model with corresponding exciton energies, the Rabi splittings, are fitted to be 39 meV ( $V_A = 19.8$  meV) at 130K, 37 meV ( $V_A = 18.8$  meV) at 210K and 36 meV ( $V_A = 18.3$  meV) at 230K, which are consistent across all these temperatures. The k-space PL at these temperatures is also shown in Fig. 3b, where the overlaid reflectivity dispersions agree well with the PL intensity distribution. The UP intensity is still magnified to be comparable with the LP intensity, indicating that the LP states are similarly stable over a large temperature range. Note that the magnifications of the upper panels become smaller as temperature increases, due to the significant change in the polariton composition.

The polariton composition from the exciton and photon can be characterized by the weighing fractions, calculated as the Hopfield coefficients as a function of cavity detunings [25]. The Hopfield coefficients for the LP branch are plotted in Fig. 4 for various temperatures. With the large binding energy and strong temperature dependence, the excitons are robust and tunable over a much broad temperature range. As the cavity detuning changes from negative (130K), to near-zero (210K), to positive (230K), the Hopfield coefficients show that the LPs can be flexibly tuned from a more photon-like, to a photon-exciton-mixed, and to a more exciton-like states at small incidence angles. This tuneability provides the freedom to control the polariton composition at will, and thus to optimize the polariton dynamics.

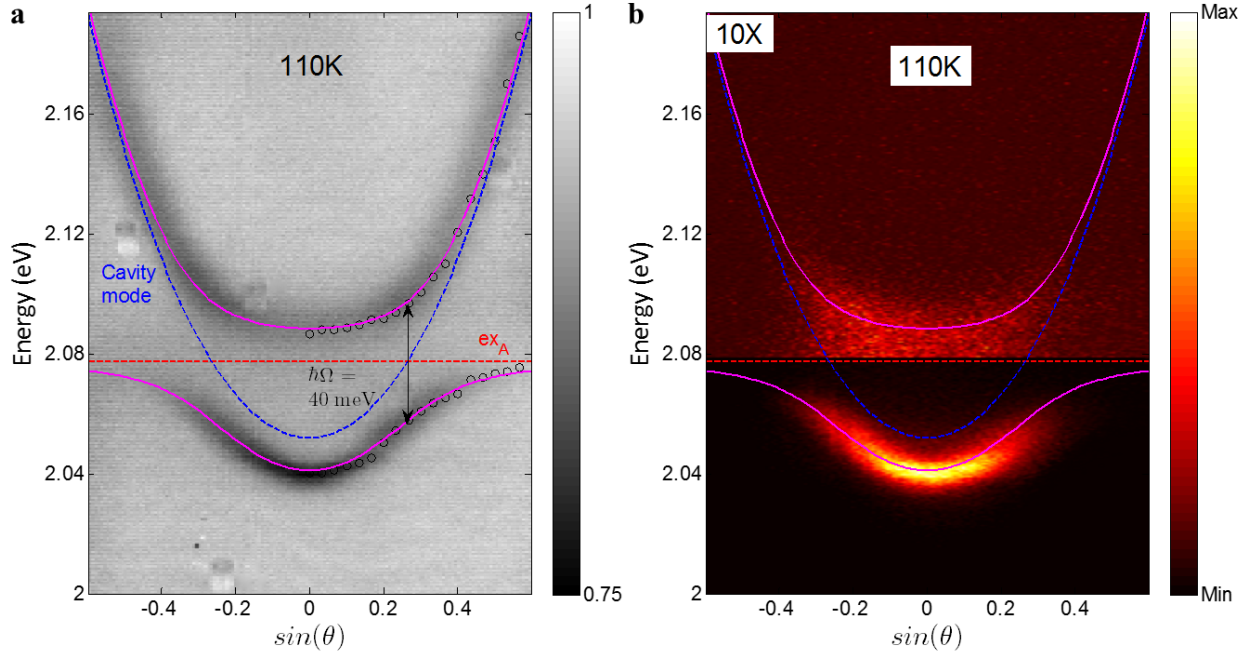
The polariton composition reveals clear correlation with the PL intensity distribution at different temperatures as shown in Fig. 3b. At negative detuning, the LP is more photon-like at

small angles, while becoming more exciton-like at large angles. Since the WS<sub>2</sub> exciton lifetime of ~200 ps is much longer than the cavity photon leakage lifetime of ~1 ps, the more photon-like polariton (LP branch) lifetime is generally smaller than more exciton-like polariton (UP branch) lifetime. The LP states have a faster lifetime, and are thus the preferred states for polariton leakage emission, which is consistent with PL intensity of the LP state being maximal throughout all temperatures 110-230 K. Once the detuning becomes positive, at small angle the LP starts to become more exciton-like while the UP becomes more photon-like. The photon-like trend of UP branch at positive detuning decreases the UP leakage lifetime, enhancing the UP PL intensity. This transition of the hybrid polariton composition leads to less emission contrast between the UP and LP states at higher temperatures.

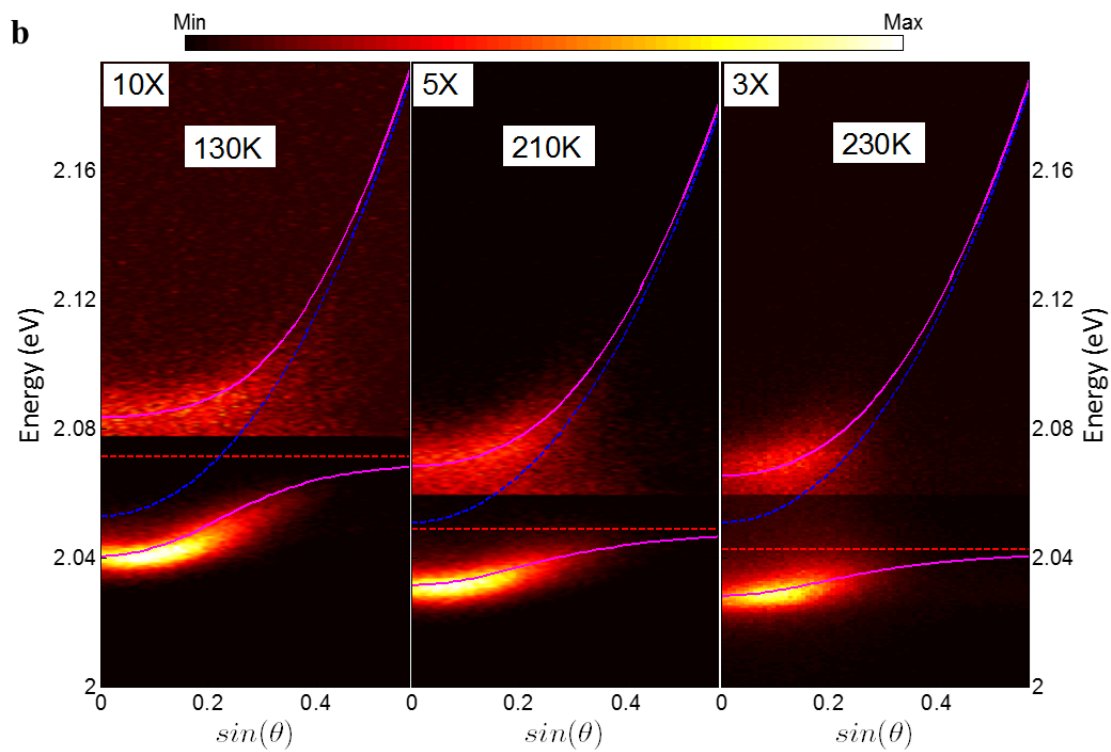
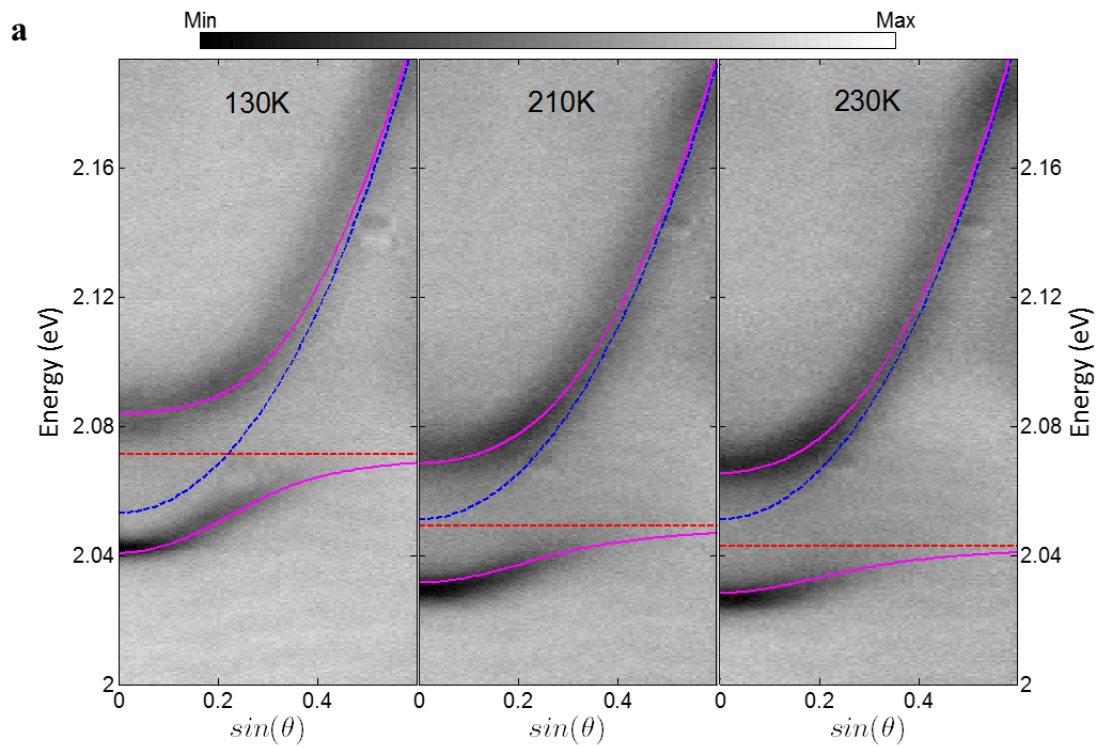
In summary, the unambiguous control of polariton dispersion is demonstrated in a WS<sub>2</sub> MC, due to the large exciton binding energy, strong temperature-dependence of Wannier excitons, unperturbed exciton quantum yield and lifetime over a large temperature range. By tuning the polariton dispersion, the angular dependent polariton composition can be flexibly controlled, enabling the control of polariton emission dynamics. This controllability illustrates a powerful way to optimize polariton dynamics and paves the way to realize polariton condensation in TMD monolayers. Furthermore, the coherent strong coupling with a large SLR of 3.3 not only ensures the fundamental basis for polariton condensation, but also holds great promise to preserve the coherence of valley excitons in TMD monolayers.



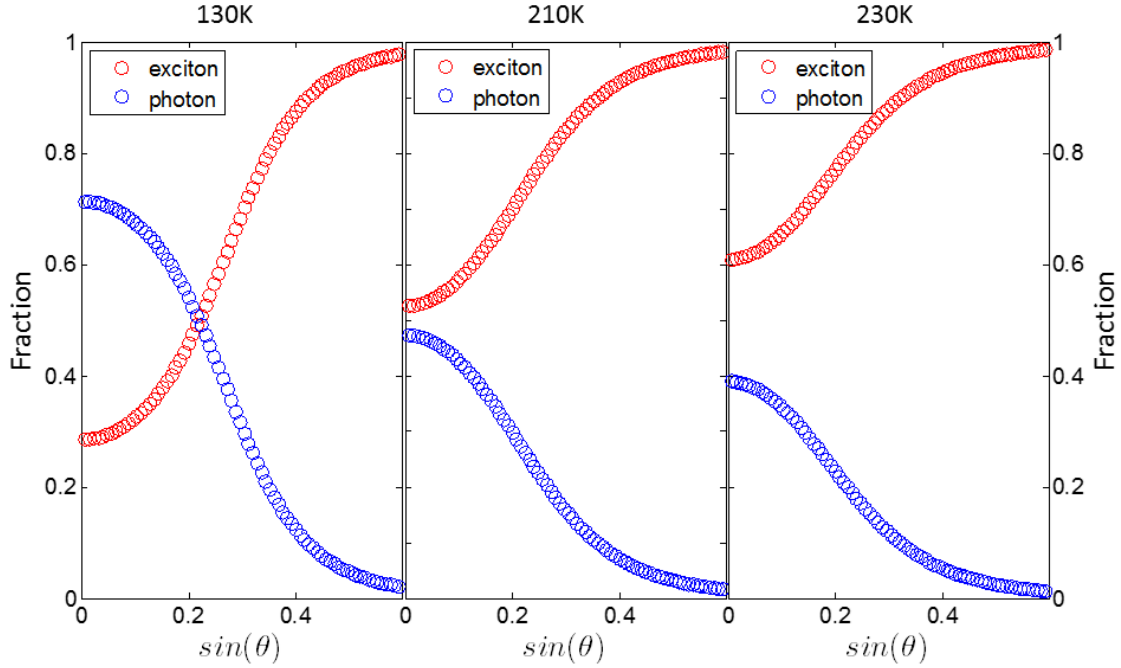
**Figure 1. Schematic of TMD microcavity and improved WS<sub>2</sub> exciton emission.** **a**, Schematic of the microcavity (MC) structure. It consists of 12.5 periods of a SiO<sub>2</sub>/Si<sub>3</sub>N<sub>4</sub> bottom distributed Bragg reflector (DBR) and 7.5 periods of a top DBR. The cavity layer structure includes an exfoliated monolayer WS<sub>2</sub> flake sandwiched by two hydrogen silsesquioxane (HSQ) layers. There is a thin Al<sub>2</sub>O<sub>3</sub> film atop of the top HSQ layer to protect the WS<sub>2</sub> during the top DBR growth. **b**, The WS<sub>2</sub> photoluminescence (PL) spectra of exfoliated WS<sub>2</sub> on SiO<sub>2</sub>/DBR and HSQ/DBR substrates, and WS<sub>2</sub> on HSQ/DBR capped with HSQ and Al<sub>2</sub>O<sub>3</sub> layer under the same pump and collection conditions. Besides the intensity enhancement, the PL peak position and FWHM are all consistent. The inset shows the optical image of the monolayer WS<sub>2</sub> flake (the light-green trapezoid shape) with a laser beam at the center (the white circular shape) before depositing the top DBR, the scale bar corresponds to 10 μm.



**Figure 2. k-space reflectivity and photoluminescence (PL) at 110 K.** **a**, The reflectivity map at 110 K obtained by k-space spectroscopy, where the horizontal axis represents the sine function of the incidence angles ( $\theta$ ) and the vertical axis is the photon energy. The grey colour scale represents the reflectivity with darker areas corresponding to lower reflectivity. Two reflectivity modes are identified as the two anti-crossing polariton branches with dispersion fitted by a coupled oscillator model [25] with a 40 meV Rabi splitting. The dispersion plot is overlaid with the reflectivity map, with the dashed red line representing the exciton energy (2.078 eV), the dashed blue curve as the cavity photon dispersion, and the two solid magenta curves are the fitted polariton dispersion. This coupled oscillator model shows excellent agreement with the experimental reflectivity. **b**, The non-resonantly pumped PL map obtained by k-space spectroscopy at 110 K. The orange colour scale represents the PL intensity. The intensity of the upper polariton PL is magnified by 10 times due to its weak emission. The reflectivity dispersion is directly translated here and shows excellent agreement with the PL dispersion.



**Figure 3. Temperature-dependent reflectivity and PL.** **a**, The k-space reflectivity map at temperatures of 130 K, 210 K and 230 K. As the temperature increases, the exciton (the red dashed line) red shifts from 2.072 eV to 2.042 eV, while the cavity photon dispersion (the blue dashed curve) does not change, creating various cavity detunings over this temperature range. The coupled oscillator model is calculated to fit the dispersions (magenta solid curves) showing that the strong coupling regime holds at all these temperatures with large splitting-to-linewidth ratio. **b**, The k-space PL map at temperatures of 130 K, 210 K and 230 K. The reflectivity dispersions are directly translated here showing the agreement between the reflectivity and PL dispersions. Since the UP PL is much weaker, the intensity is still magnified by 10X, 5X and 3X at the upper panels at all these temperatures, respectively. Note here that the UP PL intensity becomes comparable with lower polariton (LP) intensity as the UP becomes more photon-like at higher temperatures.



**Figure 4. Hopfield coefficients of lower polariton (LP) states at three temperatures.** Based on the coupled oscillator model, the contributions from exciton and photon components on the polariton states can be characterized by Hopfield coefficients [25]. The Hopfield coefficients given as a function of the incidence angle indicate how the polaritons are hybridized by the excitons and photons. As temperature increases, the cavity detuning changes from negative to positive, leading to a transition from a more photon-like to a more exciton-like state. This transition also alters the emission dynamics of the polaritons and thus the PL intensity distribution as seen in Fig. 3b.



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