

Supplementary information for:
Moiré-engineered light-matter interactions in
MoS₂/WSe₂ heterobilayers at room
temperature

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Supplementary Notes

Note S1. Discussion about the energy tails of PL peaks

The low-energy tail of the PL peak of MoS₂/WSe₂ heterobilayer with a large twist angle (e.g., $\sim 2.8^\circ$, see Fig. 1b) is similar to that of monolayer WSe₂ [1, 2] and MoS₂ [3]. We expect the possible origin of defect-related phonon sidebands for the following reasons:

1) TMD monolayers are actually quite defective. Despite high-quality flux-grown crystals are used for fabricating our samples, a defect density of about 10^3 point defects per $1\mu\text{m}^2$ is expected [4]. The physical picture of the single photon emitters in TMD monolayers is commonly described as the localization of excitons in defects [5]. The presence of high-density defects can be reflected by the observation of quantum emitters in TMD monolayer/pillar structures [6, 7] arising from the activation of defects due to exciton funneling. Therefore, the optical properties of the stacked heterobilayers will inevitably be affected by defects.

2) According to previous work [3], the low-energy tail of PL peak of MoS₂ is suppressed after the passivation of non-radiative defects. Additionally, the exciton lifetime is significantly prolonged. In our case, the presence of non-radiative defects contributes to a short exciton lifetime of samples with large twist angles, and the suppression of exciton occupation in defects with moiré potential enhances the exciton lifetime, as shown in Fig. 3 and Fig. S6.

3) The phonon sidebands typically result in a low-energy tail of the PL peak of TMD monolayers [1, 2].

4) We carefully compared the PL spectrum of the samples with large twist angles, observing that the low-energy tail becomes more prominent when increasing the twist angle (see Fig. S15). We expect that the non-radiative centers or defects exist in samples with different twist angles, and the key point is the interplay between non-radiative centers and moiré effect. According to the observation in Fig. 3, the moiré potential passivates the non-radiative centers, featuring brighter excitons with longer lifetimes. Identifying how the different types of defects or disorders affect PL emission and how the moiré pattern can affect phonon sidebands is challenging, inspiring future work.

In stark contrast, the samples with a small twist angle ($\sim 0.5^\circ$) show high-energy tails of PL peaks and their presence is due to the quantized energy levels within a moiré potential trap. The invisible low-energy tail can be ascribed to the passivation of non-radiative defects by moiré potential.

Note S2. Calculation of exciton density

The dipolar nature of interlayer excitons enables the repulsive interaction among excitons, and the resulting energy shift can be used to derive exciton density, as described by the formula [8]: $n_{\text{IX}} = \delta E \epsilon_0 \epsilon_{\text{HS}} / qd$, where δE is the blue-shifted energy of IX, ϵ_0 is the vacuum permittivity, ϵ_{HS} is the effective relative permittivity of MoS₂/WSe₂ heterobilayers, q is an elementary charge of electron, and d is the interlayer spacing between the constituent monolayers.

Given an energy shift of 15 meV for IX1 at 335 uW (see Fig. 1c), we can derive an exciton density of $0.95 \times 10^{12} \text{ cm}^{-2}$ that reaching moiré density ($1.38 \times 10^{12} \text{ cm}^{-2}$). This indicates that the exciton number per moiré cell is approaching 1. The lateral distribution of the Gaussian beam of the pump laser could lead to an exciton number in a moiré cell at the center of the laser spot larger than unity, evidenced by the blue shift of IX1 upon increasing pump power (see Fig. 1c) owing to repulsive dipolar interaction [9].

Note S3. Calculation of the moiré period

The moiré period a_M is calculated via [10]:

$$a_M = \frac{a_{\text{MoS}_2} a_{\text{WSe}_2}}{\sqrt{a_{\text{MoS}_2}^2 + a_{\text{WSe}_2}^2 - 2a_{\text{MoS}_2} a_{\text{WSe}_2} \cos \theta}},$$

where a_{MoS_2} (a_{WSe_2}) is the lattice constant of monolayer MoS₂ (WSe₂). $a_{\text{MoS}_2} = 0.316 \text{ nm}$ and $a_{\text{WSe}_2} = 0.328 \text{ nm}$ [11] are used to calculate the moiré period. The largest moiré period is about 8.6 nm, which remains slightly larger than the measured size of exciton of 5.2 nm [12] and is thus beneficial for exciton confinement. It is worth noting that the large lattice mismatch of MoS₂/WSe₂ heterobilayer facilitates the retention of the small moiré cells against local twist angle disorder and prevents the formation of significantly enlarged moiré cells shown in previous works [13, 14].

Note S4. IX energy-twist angle dependency

In our investigated MoS₂/WSe₂ heterobilayers, we find that the IX energy (E) strongly depends on twist angle and the experimental data can be fitted well by an exponential function: $E(\theta) = a - bc^\theta$. Here, the derived values of a , b , and c are 1.14767, 0.2242, and 0.60813, respectively. Regarding the energy of interlayer excitons, the energy shift can be ascribed to the compensation of momentum mismatch and the increased depth of moiré potential upon decreasing twist angle [15]. Additionally, we find that the twist-angle dependent energy shift can be up to $\sim 224 \text{ meV}$ according to the equation, in our case, is significantly larger than that ($\sim 60 \text{ meV}$) observed in MoSe₂/WS₂ moiré superlattices [16]. This finding reflects that the moiré potential in our system is more significant. Furthermore, moiré potential strongly depends on interlayer coupling, evidenced by the sufficient suppression with an additional monolayer hBN in the TMD heterobilayer [17]. Therefore, we expect that the significant energy

described by the equation implies the change of interlayer spacing between monolayers.

On the other hand, the equation is used as an empirical formula to derive the twist angle according to the measured IX energy in the PL spectrum. For example, regarding a measured IX emission at 1217 nm (i.e., 1.019 eV), a twist angle of $\sim 1.1^\circ$ can be obtained. This twist angle corresponds to a moiré period of ~ 7.7 nm, well consistent with the moiré period measured by AFM (~ 7.3 nm, see the inset of Fig. 2c). Moreover, determining the twist angle by straight edges and polarized SHG suffers from local twist-angle disorder induced by bubbles and wrinkles. Therefore, it is reasonable to use IX energy to derive twist angles of the twisted heterobilayers shown in Fig. 2 and Fig. 3.

Note S5. Exciton lifetime in MoS₂/WSe₂ heterobilayers with different twist angles

The radiative recombination rate of an exciton determines the brightness of the light emission. A short radiative lifetime (τ_r) is typically required for bright emitters. Experimentally, the measured exciton lifetime (τ) can be defined as $1/\tau = 1/\tau_r + 1/\tau_{nr}$, where τ_{nr} is the non-radiative lifetime. Therefore, a shorter exciton lifetime should give rise to a brighter PL emission if we assume a fixed non-radiative lifetime. However, we find that the IXs with longer exciton lifetime show brighter PL emission (see Fig. 3 and Fig. S6). This counter-intuitive behavior indicates that the non-radiative lifetime increases in regions with smaller twist angles and deeper moiré potentials. Non-radiative centers such as defects in TMD monolayers can capture excitons and result in two orders of magnitude lower PL quantum yield [18]. The brighter PL emission from regions with smaller twist angles reflects the enhanced PL quantum yield due to exciton confinement. Considering the increasing exciton density with pump power, exciton-exciton annihilation can play an important role in exciton lifetime. More specifically, the exciton-exciton annihilation results in a significant power-dependent exciton lifetime reduction [19]. In moiré superlattices, we find that the exciton lifetime of well-localized IX such as IX1 does not decrease with increasing pump power, indicating that the moiré potential in MoS₂/WSe₂ heterobilayers can effectively suppress non-radiative effects.

Note S6. Discussion on output behavior of the moiré superlattice-cavity devices

The output of the devices depends on whether exciton density exceeds the transparent density (n_{tr}) or threshold density (n_{th}). If $n < n_{tr}$, i.e., the exciton acts as a light absorber, will prevent one from observing the cavity mode, which is shown in a previous report with MoTe₂ [20]. Through PL measurements, we observe a pronounced sharp emission line of D1 with an enhancement factor of 41.5 (Fig. 4d), which is significantly larger than the reported enhancement factor from other TMD-cavity structures [21]. Interestingly, we find that the output of the cavity mode of D1 increased linearly with pump intensity

(Fig. S12), and the cavity mode keeps dominating the emission even at low pumping levels. Considering the high-quality factor ($>10^4$) of the cavity, we propose that the absence of a kink in the L-L curve is due to a low threshold beyond the measurement capability of our instruments.

As shown in Fig. 1d, the lowest-energy state has the highest possibility of being occupied by photon-generated IXs at low pump levels, and then the occupation possibility of higher energy states becomes higher with increased pump power. In this case, we expect that a cavity mode that couples to a higher-energy IX state will give rise to a higher threshold. Therefore, D2 and D3 are fabricated with higher-energy cavity modes that are mainly coupled to the adjacent higher- and highest-energy IX states in similarly twisted moiré superlattices, respectively. We note that the emission peaks of the cavity modes can be well-fitted by a Lorentzian function, as shown in the example with D3 (see Fig. S16). Moreover, the integrated PL intensities extracted from the fitting show that both D2 and D3 show a clear kink in the L-L curve, which is known as threshold behavior. To compare all three devices, the threshold is extracted by finding the cross-point of the fitting line of the L-L curve and the pump-power axis (see Fig. S12). In this case, the thresholds of D1, D2, and D3 are $\sim 0.11 \text{ kW/cm}^2$, $\sim 0.18 \text{ kW/cm}^2$, and $\sim 4.53 \text{ kW/cm}^2$, respectively. The comparison of the thresholds of these devices indicates that the lowest-energy IX state supplies the largest optical gain at low pump levels. Therefore, we conclude that the absence of a threshold in D1 is due to the low threshold that is beyond the capability of our measurement setup.

While the observed threshold indicates the presence of optical gain, it is still unclear whether the exciton density can reach the threshold density (i.e., $n > n_{th}$) for lasing. This is because cavity-enhanced spontaneous emission can also show similar threshold behavior [22]. The second-order auto-correlation function at zero time delay (i.e., $g^2(0)$) can be measured by the HBT experiment to distinguish the phase transition from chaotic light ($g^2(0)=2$) to coherent light ($g^2(0)=1$). The $g^2(0)$ of cavity-enhanced spontaneous emission cannot go to unity after the threshold density is reached. Correlating the photon statistics with the L-L curve is an interesting and inspiring direction for future work.

Supplementary Figures

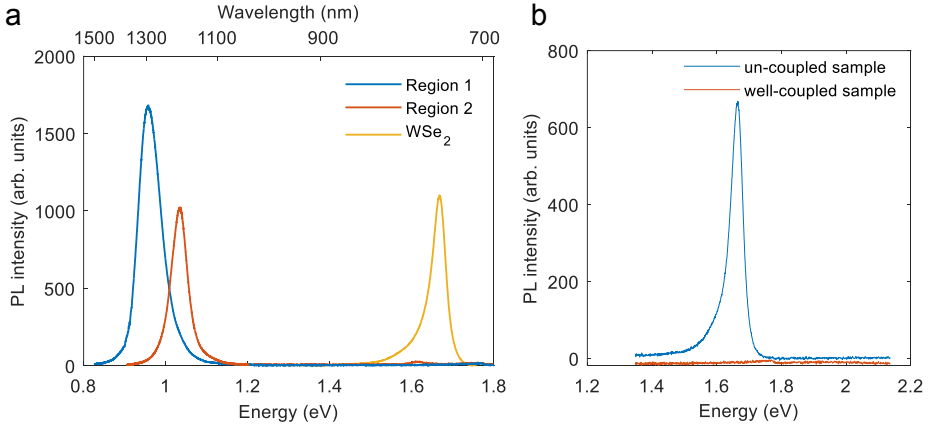


Fig. S1 PL quenching of WSe₂ A-excitons. (a) PL spectrum of the WSe₂ A-exciton in monolayer WSe₂ and MoS₂/WSe₂ heterobilayer shown in Fig. 3. (b) Comparison of WSe₂ A-exciton emission from uncoupled 2L-MoS₂/WSe₂ and well-coupled MoS₂/WSe₂. The strong WSe₂ exciton emission, together with the unchanged exciton absorption energy (see Fig. 2a) compared to that of monolayer WSe₂, confirm the negligible interlayer coupling between 2L-MoS₂ and WSe₂. The PL spectrum of the MoS₂ A-exciton is not measured because its energy is too close to that of the pump laser.

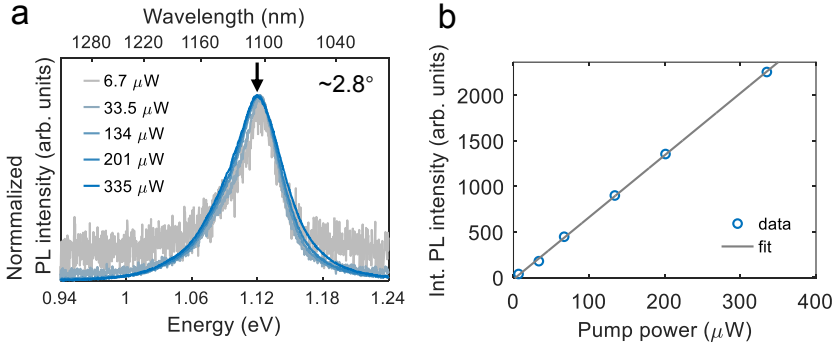


Fig. S2 PL emission from a $\sim 2.8^\circ$ MoS₂/WSe₂ heterobilayer at different pump powers. (a) Power-dependent PL spectra, with the arrow indicating the peak energy. No clear change in the PL peak and the peak energy shift is observed, indicating that the moiré effect is negligible. (b) The integrated PL intensity of the IX emission peak as a function of pump power.

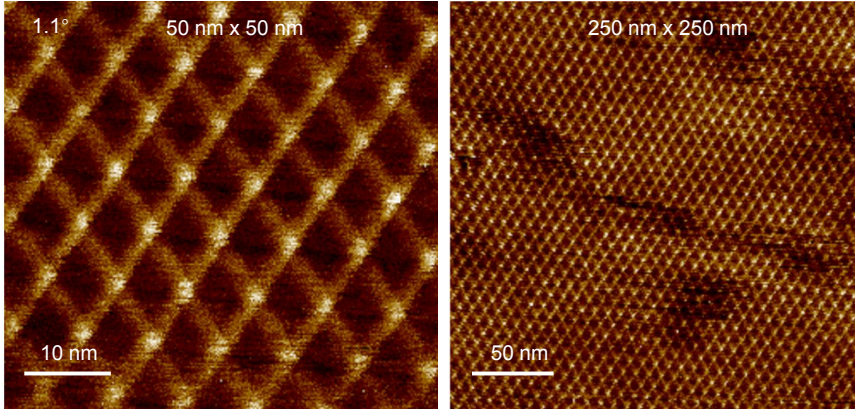


Fig. S3 Moiré pattern of the sample shown in the inset of Fig. 2c in the stiffness channel with different scanning ranges. The amplitude of the stiffness modulation is about 19 MPa. The moiré pattern in this channel reflects the periodically modulated Young's modulus.

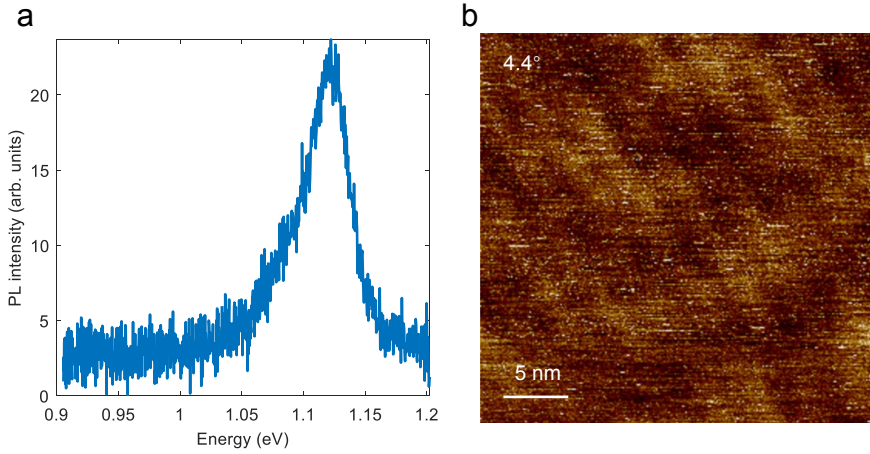


Fig. S4 Characterizations of a $\sim 4.4^\circ$ $\text{MoS}_2/\text{WSe}_2$ heterobilayer. (a) PL spectrum of the sample. (b) The measured height profile with AFM. The moiré pattern is invisible because of the absence of atomic reconstruction, or the amplitude of the height modulation is below the resolution limit of ~ 30 pm.

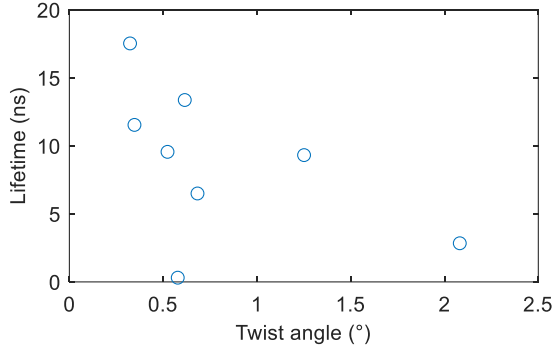


Fig. S5 IX lifetime of $\text{MoS}_2/\text{WSe}_2$ heterobilayers as a function of twist angle. No clear trend in twist angle-dependent IX lifetime is visible, which we ascribe to the different optical qualities of the fabricated samples.

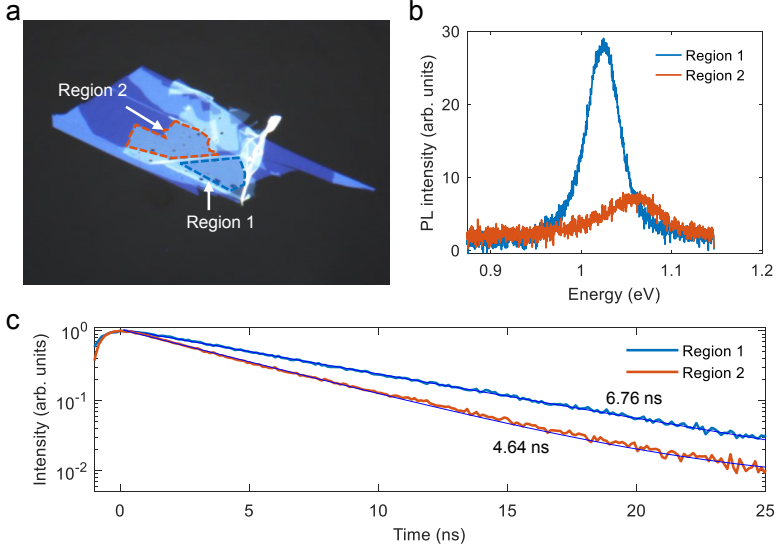


Fig. S6 Reproducibility of twist-angle-dependent IX lifetime. (a) Optical microscope image of a $\text{MoS}_2/\text{WSe}_2$ heterobilayer that is divided into two different twisted regions by a wrinkle. (b) PL spectrum of the different twisted regions (region 1 and region 2 with twist angles of $\sim 1.2^\circ$ and $\sim 1.9^\circ$ respectively) under the same pump power. (c) Time-resolved PL dynamics from two regions with different twist angles. The black curves are exponential fits to the data, giving an IX lifetime of 6.76 ns (4.64 ns) for Region 1 (Region 2).

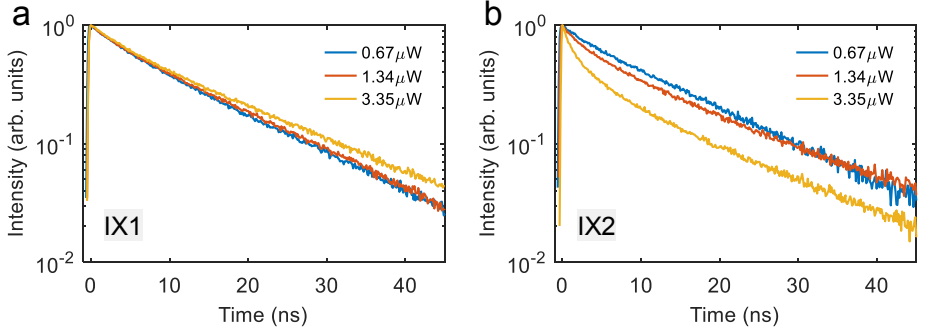


Fig. S7 IX lifetime of Region 1 in Fig. 3 under different pump powers. A bandpass filter with a center wavelength of 1300 nm and a bandwidth of 12 nm is used to measure the exciton lifetime of the lowest energy state (IX1), as shown in (a). A bandpass filter with a center wavelength of 1200 nm and a bandwidth of 10 nm is used to measure the exciton lifetime of a higher energy state (IX2), as shown in (a). A faster additional decay in (b) at 3.35 μW can be ascribed to the contribution of delocalization of IX2, which is also observed in WS_2/WSe_2 moiré superlattices [23].

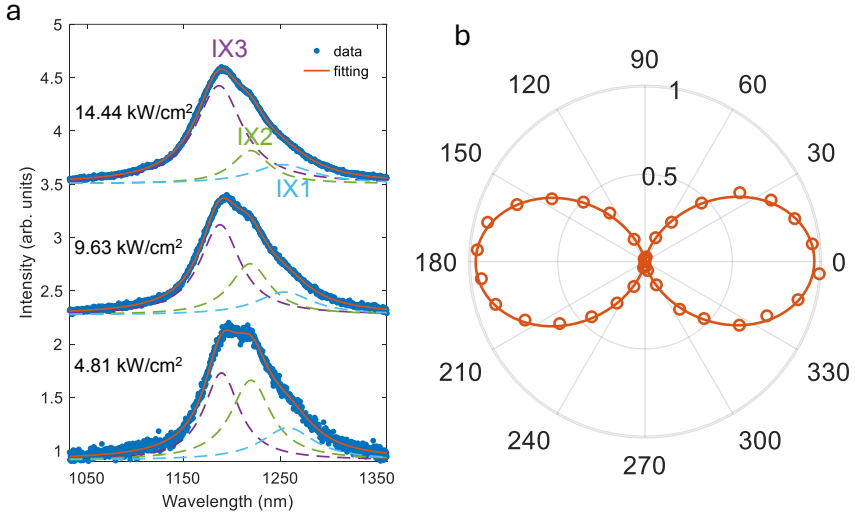


Fig. S8 D3. (a) PL spectra of hBN-encapsulated $\text{MoS}_2/\text{WSe}_2$ on the cavity, which are normalized by their peak intensities. The relatively increased PL intensity of high-energy peaks is due to the increased occupation probability of high-energy IX states, which is in good agreement with that observed in Fig. 1d. The cavity mode is filtered out by a linear polarizer due to its excellent linear polarization, as shown in (b).

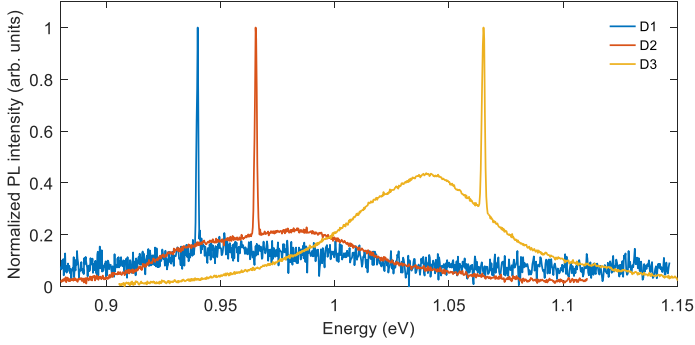


Fig. S9 PL spectra of the different moiré superlattice-cavity devices. The PL spectra are acquired when the cavity modes become prominent under different pump power densities with a grating of 150 grooves/mm, which are normalized by their peak intensities. D1, D2, and D3 are pumped by a power density of $\sim 1.39 \text{ kW/cm}^2$, $\sim 1.73 \text{ kW/cm}^2$, and $\sim 33.97 \text{ kW/cm}^2$, respectively. This difference mainly originates from the transition of the dominating IX state upon increasing pump power (see Fig. S8 and Fig. 1c). We note that the low-resolution grating is used to clearly see the PL emission background.

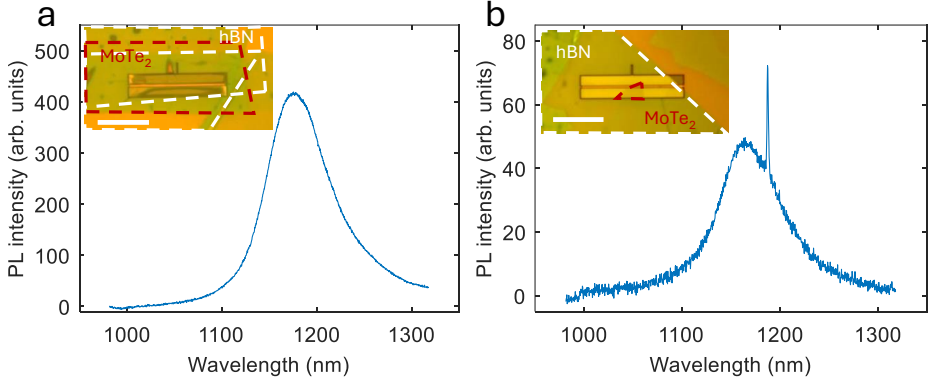


Fig. S10 Bilayer MoTe_2 -cavity devices. The hBN-capped MoTe_2 flakes fully cover and partially cover the cavities with the same parameters, as shown in (a) and (b), respectively. The samples are characterized by the same PL measurement setup. The partially-covered sample with only a small fragment in the center features a clearly cavity-enhanced PL, while the cavity mode is absent in the fully-covered sample, indicating large intralayer excitonic absorption introduced by the un-pumped MoTe_2 area can impede the formation of the cavity mode.

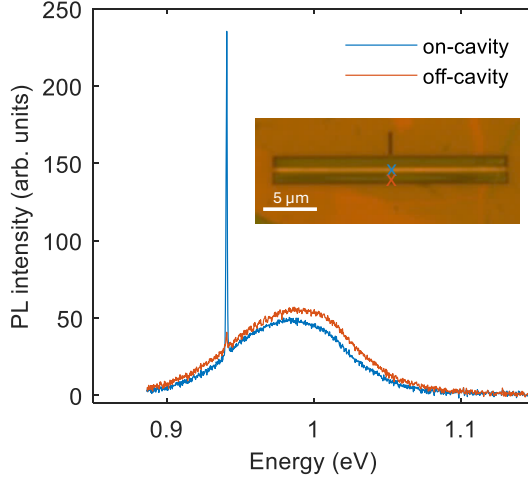


Fig. S11 PL emission from the heterostructure on- and off-cavity of D1. It is worth noting that the PL spectra are acquired by a grating of 150 grooves/mm to capture the PL background, resulting in an averaged cavity emission peak (i.e., the linewidth broadens and peak intensity decreases) compared to the data in Fig. 4e acquired by a grating of 1200 grooves/mm.

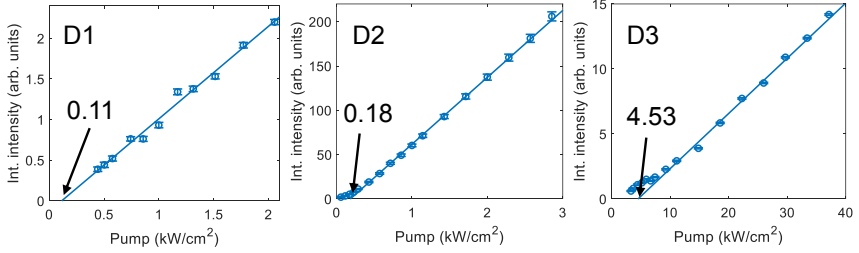


Fig. S12 The L-L curve of different devices acquired with a grating of 1200 grooves/mm. The integrated PL intensities with error bars are extracted by fitting the data with a Lorentzian function. The threshold is determined by identifying the intersection point (pointed out by the black arrow) of the fitting line of the L-L curve with the pump axis.

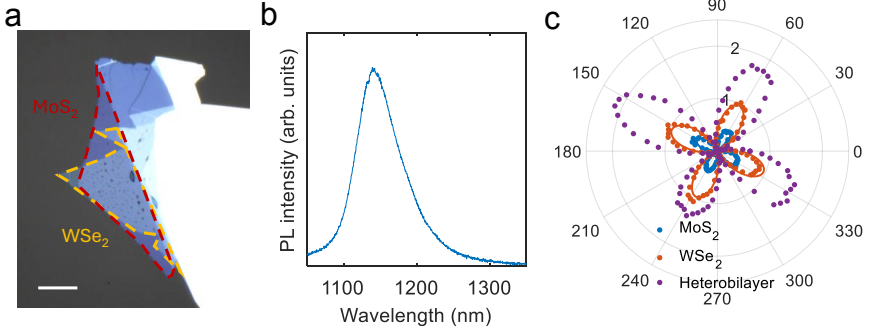


Fig. S13 Correlation between twist angle and IX energy. (a) The optical microscope image of the sample. Scale bar: $10\ \mu\text{m}$ (b) IX emission spectra. (c) SHG from monolayers and heterobilayer. The twist angle derived from SHG measurements is about 2.7° , corresponding to an IX energy of 1.089 eV. This correlation agrees well with the twist angle dependence of the IX energy in Fig. 1b.

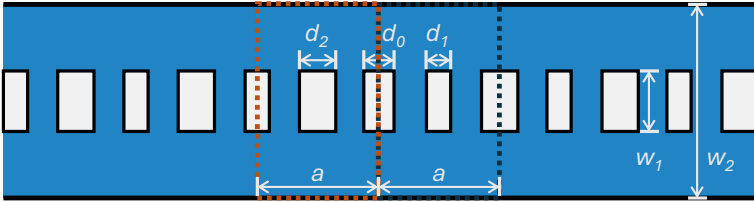


Fig. S14 Sketch of the top-view of the topological photonic crystal nanobeam cavity. The red and blue unit cells (dashed boxes) have the same periodicity (denoted as a) with different air-hole arrangements. The interface lies at the center where the red and blue unit cells meet. The parameters of the cavity are marked by d_0 ($=0.25a$), d_1 ($=0.2a$), d_2 ($=0.3a$), w_1 ($=0.5a$), and w_2 ($=1.6a$). The thickness and length of the nanobeam are fixed at 220 nm and $70a$, respectively. By varying a , we tune the cavity resonance to match the multiple peaks in the small twisted moiré superlattices. The devices (D1, D2, and D3) mentioned in the main text were fabricated with $a=300\ \text{nm}$, $280\ \text{nm}$, and $260\ \text{nm}$, respectively. In this design, the simulated cavities show a similar Q-factor of about 20000.

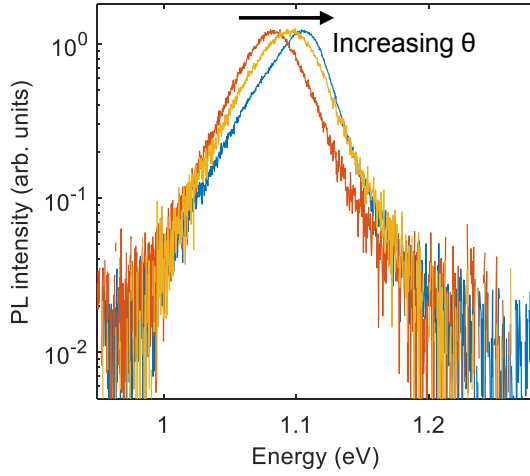


Fig. S15 Comparison of PL spectrum of samples with large twist angles. The twist angles are derived by the empirical equation shown in Note S4.

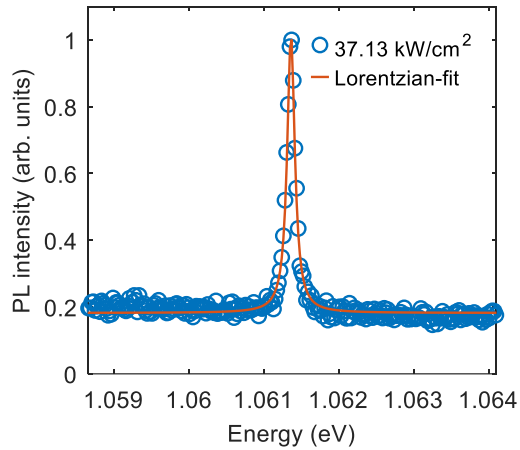


Fig. S16 Fitting of the emission peak of D3 with a Lorentzian function. The spectrum is acquired by a grating of 1200 grooves/mm, showing the PL background is less significant than that acquired by a grating of 150 grooves/mm.

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