

Peer Review File

Moiré-engineered light-matter interactions in MoS₂/WSe₂
heterobilayers at room temperature



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REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The authors reported the observation of multiple interlayer excitons (IX) peaks in the photoluminescence (PL) spectrum of MoS₂/WSe₂ heterobilayer at room temperature, which they claimed to be moiré IX peaks for the small twist angle sample. They support this assignment by observing that the low-energy peaks show low power saturation, indicating localized states. The existence of deep moiré superlattice is further probed using atomic force microscopy (AFM), showing moiré modulated mechanical deformation. Further experiments, including reflection contrast of the intralayer exciton, are used to claim that the atomic reconstruction is the cause of the deep moiré superlattice trap. They also compare the lifetime and intensity of two moiré superlattices, showing that the moiré superlattice suppresses IX nonradiative decay. Finally, by coupling different IX species to the photonic crystal cavity, they show that the laser device utilizing moiré trapped IX could have one order lower lasing threshold compared to that using unlocalized IX.

Overall, the results, especially the low threshold demonstration, are very promising for the IX application. However, there are some points that are confusing to me. I will support publication in Nature Communication provided the comments below are addressed satisfactorily.

1. The optical pump power applied is quite high (reaching mW regime). Is this already the regime where IX density reaches or exceeds the moiré density? Could the authors estimate the IX density vs power in their experiment, e.g., based on IX blueshift or other means?
2. What are the values of saturation powers for IX1 and IX2 in Fig. 1d? They seem quite different, with the IX2 power saturation behavior being very weak. Could the authors give a more detailed analysis of what mechanisms can contribute to the saturation behavior for each IX species?
3. While the localization can be due to moiré superlattice, it can also be due to disorder. How can the author rule out the disorder contribution in the IX PL emission?
4. Is the energy vs. twist angle shown in Fig. 1e related to IX1 emission? Was the peak energy obtained from the multi-peak fitting? The uncertainty in the energy value has to be presented in the graph or the figure caption. Can the authors also present the energy vs twist angle for IX2 and IX3?
5. What is the origin of IX2? Is it because IX-IX repulsion caused a change in the moiré potential experienced by the IX (like in <https://doi.org/10.1021/acs.nanolett.3c01160>), or is it because there are multiple IX energy levels due to a deep trap? Will these two mechanisms result in different power dependence for IX2?
6. The authors observed a low-energy tail for the large twist angle ($> 1^\circ$) samples. In the discussion, they said it is similar to the phonon sideband in monolayer WSe₂. However, in the following sentence, they said it comes from defects. Could the authors clarify which of these two is the mechanism they believe to be the main reason for the low-energy tail? These two mechanisms differ in temperature dependence behavior, i.e., increasing the temperature will induce detrapping while increasing the effect of the phonon sideband. Could the authors provide such a temperature-dependence study to justify the origin of the low-energy tail?

7. In Fig. 2a, one of the samples is uncoupled bilayer (2L) MoS₂/WSe₂. What is the meaning of uncoupled here, and how could it be ensured that it is uncoupled?
8. In Fig. 2c, $X_{\text{MoS}_2}^A$ in MoS₂/WSe₂ heterobilayer has energy below that in 2L-MoS₂. The authors said that this must be due to expanded lattice constant. However, it is also possible that this is caused by the influence of the moiré superlattice since the superlattice potential can create moiré minibands with energy below the MoS₂ monolayer conduction band or higher than MoS₂ monolayer valence band. How could this effect be ruled out?
9. The authors said:
“..., monolayer WSe₂ with a smaller lattice constant (0.328 nm) expands while monolayer MoS₂ with a larger lattice constant (0.316 nm) shrinks.”
 This statement is strange. First, $0.328 > 0.316$, shows the statement must be wrong. Instead, I expect WSe₂ with a larger lattice constant to shrink while MoS₂ expands.
10. The authors observed that, for a twist angle $< 0.9^\circ$, an additional higher energy state is detected in the reflection contrast. First, I want to confirm that this is related to the WSe₂ exciton peak (which seems to be the case based on Fig. 2a). If this is the case, why is it absent in the MoS₂ exciton peak? Given the reason is that this is intralayer exciton localized at a different site in a moiré cell, shouldn't a similar feature be observed for the MoS₂ exciton peak also? Is it possible that this is not due to such a reason, but other reasons, including phonon-replica or intervalley exciton?
11. The authors said that the equation, $E(\theta) = a - b\cos\theta$, in Note S2, implies that the significant energy change of IX is related to the interlayer spacing between monolayers. Why is that so?

Reviewer #2 (Remarks to the Author):

The manuscript by Q. Lin & H. Fang et al. reports optical properties of twisted MoS₂/WSe₂ heterostructures at room temperature. The optical spectrum is investigated using luminescence and reflectance spectroscopy, and the morphology is studied by atomic force microscopy. Moreover, the authors place hBN encapsulated heterostructures on top of a nanobeam cavity to study the coupling of differently confined interlayer excitons with cavity photons.

The discussion of the data presented in Figs. 1-3 is sound and the conclusions seem solid. Since the authors mention light-matter interactions in their title, it would be nice if they presented a consistent analysis of the oscillator strength as a function of twist angle. The analysis and discussion of the data shown in Fig. 4, however, is quite unsatisfactory (see comments below). In the current presentation, I do not see any interesting insights from this part of the manuscript. As outlined below, it is not clear why the results are discussed in the context of lasing and threshold behavior, as the effects involved might simply result from the coupling of the PL emission to cavity modes and Purcell physics, which is all well explored.

If the authors were able to show compelling evidence that different localization of moire excitons can significantly improve optical properties at room temperature, I would be willing to consider recommending the manuscript for publication. Currently, however, I am not convinced.

Comments:

1) The authors mention lasing and talk about the threshold behavior of the power-dependent PL intensity they measure from three different samples on top of the nanobeam cavity. I noticed a few issues:

a) It is repeatedly mentioned that the cavity is “topological”, yet besides a reference, neither the impact of such properties on the optical mode spectrum nor the emission of the heterostructures are discussed in the manuscript. What are significant differences of the “topological nanobeam cavity” compared to any conventional nanobeam cavity? If the authors want to make claims about topological features, these claims should be supported by their data, otherwise this language is unnecessary and simply distracting.

b) All relevant data is shown in Supplementary Figs. 9 & 10, while Fig. 4 of the main manuscript merely shows the result of an analysis (cf. Fig 4e) that is not very well explained in the manuscript. What is the spectral range the authors use to integrate the PL peak intensity? The background emission clearly varies significantly, which – depending on the exact analysis – can easily lead to artifacts and the features the authors use to make claims about the different “threshold” of differently localized moire excitons. It is further not clear to me what “threshold” even refers to in the absence of clear evidence for lasing.

2) I am also not very fond of the definition of the enhancement factor “x41.5”. It reflects the difference between the outcoupled emission and that of the background emission. This does not automatically mean enhancement. The factor 41.5 is, for example, impacted by the suppression of background emission by the nanobeam cavity. It might be better to compare the PL from a sample on SiO₂/Si under identical excitation conditions.

3) The authors fail to mention a recent work on moire excitons coupled to nanocavities that clearly shows lasing at low temperatures: Qian et al., Sci. Adv. 10, eadk6359 (2024). In particular, to justify the term “threshold”, I would expect to see behavior similar to the power-dependent emission in Fig. 2A.

Reviewer #3 (Remarks to the Author):

The authors measure MoS₂/WSe₂ heterobilayers at room temperature and identify the interlayer exciton (IX) emission at ~1 eV. For heterobilayers with small twist angles, the authors find saturation behaviour on the PL emission with increasing optical pump power. They also report that when decreasing the twist angle, the IX lifetime becomes longer and light emission is enhanced. Finally, heterobilayers with small twist angles are then integrated with silicon topological cavities.

Below I write some questions and comments about the manuscript.

First of all, I agree with the authors that the presented data supports the conclusions drawn in

the first section “Localized interlayer excitons at room temperature”. Indeed, the authors supply sufficient experimental evidence to demonstrate the observation of confined IX states at RT. Their data shows strong dipole-dipole repulsion.

With respect to the next section, titled “Features of intralayer excitons in moire superlattices” I have a few questions. The authors provide a set of experimental data showing clear twist dependent phenomena on the RC spectra acquired in a set of samples. While I support the claim that the experimental data is indicative of the presence of moire patterns in their samples, I do not quite agree with the explanation provided by the authors. In the manuscript, the authors explain the observed phenomena by two effects, biaxial strain and atomic reconstructions, and provide an AFM showing the reconstruction effects. Unfortunately, I am not able to see the reconstructed domains in the AFM scan. Moreover, the measured energy dependence on the WSe₂ and MoS₂ A-excitons can be explained by considering the arise of resonant hybridized states and moire minibands on the electronic structure of the heterobilayers, as reported in Ref. <https://doi.org/10.1038/s41586-019-0986-9>. In fact, the measured RC spectra follows the behaviour predicted on that article in Fig. 3e.

Now I will focus on the next section “Engineering exciton lifetimes with moire potentials”. I would really appreciate if the authors could comment on the stacking angle of regions 1 and 2, since without that information the discussion it is hard to follow. I would have expected that the difference in the exciton lifetimes are due to the tuning of momentum mismatch for different stacking angles. I think it is remarkable that the authors observe the opposite behaviour. To make this point clearer, I would suggest to modify the lines 357-376 to “This feature cannot be explained by the reduced momentum mismatch that typically leads to a shorter exciton lifetime, which is the opposite of the observed trend in Fig. 3”. Moreover, how does the fact that the IX exciton lifetime is independent of pump power relate to the reported phenomena on the first section on the manuscript, where the authors reported a strong dipole-dipole interaction as a function of pump power? What is the reason behind the absence of any impact from this dipole interaction on the measured lifetime?

In the last section of the manuscript the authors present optical spectra of heterobilayers embedded in silicon cavities. L-L curves are used to obtain threshold values for different IX-cavity detunings. They show that the IX photon collection efficiency improves by tuning the cavity mode to the IX emission. I agree with the careful treatment of the data and the conclusions drawn, stating that the measurements hint at rich physics but are not conclusive to claim lasing behaviour. In lines 732-734, the authors explain that the D1 cavity might have a low threshold power that could not be measured. I was wondering why the authors could not apply less power to the sample, since the low power point in Fig. S10 is at $\sim 0.5 \text{ kW/cm}^2$ and the authors claim a $1.6 \mu\text{m}$ spot size, giving rise to $0.5 \text{ kW/cm}^2 \times \pi \times (1.7 \mu\text{m}/2)^2 = 12 \mu\text{W}$ applied on the sample, and typical optical experiments are able to apply $\sim \text{nW}$ laser powers.

Overall, I think the work presented is novel and can be published, provided that the authors answer the questions presented above.

POINT BY POINT RESPONSE TO REVIEWERS' COMMENTS

We answer the referees' criticism point-by-point. To this end, we keep the order of the referees' comments and mark it > **in blue and bold font** <. Our answers are in black and normal text. For each point, we indicate which parts of the manuscript have been changed $\gg$ in black in response to the referee's comments.

When we refer to Figures, Tables, Citations etc., we use the numbering of the revised version of the manuscript and supplemental material.

We add a marked-up version of our revised manuscript, where **added text is in blue, normal text**, **and removed text is in red, strikethrough text**.

***** Addressing Report of Reviewer 1 *****

> 1. The authors reported the observation of multiple interlayer excitons (IX) peaks in the photoluminescence (PL) spectrum of MoS₂/WSe₂ heterobilayer at room temperature, which they claimed to be moiré IX peaks for the small twist angle sample. They support this assignment by observing that the low-energy peaks show low power saturation, indicating localized states. The existence of deep moiré superlattice is further probed using atomic force microscopy (AFM), showing moiré modulated mechanical deformation. Further experiments, including reflection contrast of the intralayer exciton, are used to claim that the atomic reconstruction is the cause of the deep moiré superlattice trap. They also compare the lifetime and intensity of two moiré superlattices, showing that the moiré superlattice suppresses IX nonradiative decay. Finally, by coupling different IX species to the photonic crystal cavity, they show that the laser device utilizing moiré trapped IX could have one order lower lasing threshold compared to that using unlocalized IX. Overall, the results, especially the low threshold demonstration, are very promising for the IX application. However, there are some points that are confusing to me. I will support publication in Nature Communication provided the comments below are addressed satisfactorily (see attachment). <

Thank you for the clear summary of our work and acknowledging its impact on the community, and for the clear recommendation to publish our work in Nature Communications after addressing the confusing points. We address all comments in the following point-by-point.

> 2. The optical pump power applied is quite high (reaching mW regime). Is this already the regime where IX density reaches or exceeds the moiré density? Could the authors estimate the IX density vs power in their experiment, e.g., based on IX blueshift or other means? <

Thank you for the suggestion about estimating exciton density, which helped us to clarify the occupation of interlayer excitons in moiré cells. The moiré density for the sample with a twist angle of 0.5° is about $1/(8.5\text{ nm})^2 = 1.38 \times 10^{12} \text{ cm}^{-2}$. According to the blueshift of

IX1 (see Fig. 1c), we can derive its exciton density of $0.95 \times 10^{12} \text{ cm}^{-2}$ under the maximum pump power of $335 \mu\text{W}$, which is reaching the moiré density. This means that the average exciton number in single moiré cells is approaching unity. The detailed calculation is shown in the newly added Note S2. 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 1, as evidenced by the blueshift of IX1 upon increasing pump power (see Fig. 1c) owing to repulsive dipolar interaction (see ref. Nano Letters 23, 4627 (2023)).

» We added Note S2 that shows the estimation of exciton density and an additional description for the exciton density in [lines 225-227](#).

> **3. What are the values of saturation powers for IX1 and IX2 in Fig. 1d?** <

The saturation powers for IX1 and IX2 are $144 \mu\text{W}$ and $714 \mu\text{W}$, respectively.

» We added the saturation powers for IX1 and IX2 into the caption of Fig. 1.

> **4. They seem quite different, with the IX2 power saturation behavior being very weak. Could the authors give a more detailed analysis of what mechanisms can contribute to the saturation behavior for each IX species?** <

Interlayer excitons can be treated as bosons when the exciton density is dilute, that is, excitons can occupy the same energy level. When the exciton density increases with pump power, the repulsive interaction among excitons becomes prominent, and the ground state is occupied by several excitons that result in the blue shift and decreased moiré peak splitting (see ref. Nano Lett. 23, 4627 (2023)). Such a repulsion poses a limit on the exciton number in the same ground state.

On the other hand, moiré superlattices have been reported to create periodically deep potential traps, leading to the emergence of quantized energy levels (see ref. Nature 567, 71–75 (2019) and Nature Communications 14, 6910 (2023)). Excitons have the largest occupation possibility in the ground state at low pump power, and then the occupation possibility of higher energy states increases upon pump power. Such an exciton-filling effect agrees well with our observation. Provided that IX1 and IX2 originate from different potential traps, we would expect to observe the PL emission from IX1 and IX2 since the pump power is lowest, like the observation of defect-related states in this work: Nat. Commun. 14, 6910 (2023). Therefore, we expect that IX1 and IX2 originate from the same potential trap arising from moiré effect, as shown Fig. R1. The combined effect of moiré potential and repulsive interaction lead to the different saturation behavior of interlayer exciton states.

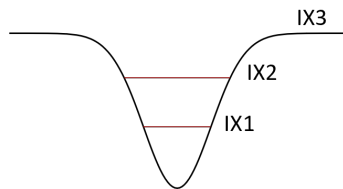


Figure R1: Energy levels within a potential trap.

» We improved our descriptions about the observation of the different interlayer excitons in [lines 210-218](#), and used Fig. R1 as the inset of Fig. 1d.

> **5. While the localization can be due to moiré superlattice, it can also be**

due to disorder. How can the author rule out the disorder contribution in the IX PL emission? <

Let us assume that the exciton localization results from the disorder. In this case, we should be able to observe the exciton localization in samples with different twist angles. However, we find that only samples with a small twist angle ($\sim 0.5^\circ$) show the noticeable multi-peaks with increasing pump power, while not for the case with a relatively large twist angle (2.8°) (see Fig. S2). This twist-angle-dependent behavior helps us rule out the disorder's contribution.

>> We added this description in **lines 220-221**: "This twist-angle-dependence helps us to rule out the contribution of disorder in the materials".

> 6. Is the energy vs. twist angle shown in Fig. 1e related to IX1 emission? Was the peak energy obtained from the multi-peak fitting? The uncertainty in the energy value has to be presented in the graph or the figure caption. <

The appearance of multiple peaks only occurs for the samples with small twist angles ($\sim 0.5^\circ$). Therefore, it is challenging to define IX1, IX2, and IX3 for the samples with different twist angles. To obtain the twist-angle-dependence, the exciton energy in Fig. 1e is extracted from the PL peak energy in the PL spectrum under the same low pump power ($33.5 \mu\text{W}$) where single emission peaks dominate the PL spectrum, exemplified in Fig. 1b and that is why the uncertainty is not presented in Fig. 1e.

>> We note the absence of twist-angle-dependence for various excitonic states in **lines 236-238**.

> 7. Can the authors also present the energy vs twist angle for IX2 and IX3? <

We wish we could observe all three interlayer exciton states in the sample with different twist angles, and then monitor their responses. However, the fact is that the multi-peak feature is only visible for the samples with a small twist angle ($\sim 0.5^\circ$). For the sample with a relatively larger twist angle ($\sim 2.8^\circ$), we do not observe multiple peaks, as shown in Fig. S2, which can be explained by the decreasing effective depth of the moiré potential traps upon increasing twist angle, see ref. Science 376, 406-410 (2022). This impedes us from presenting the energy vs twist angle for IX2 and IX3.

> 8. What is the origin of IX2? Is it because IX-IX repulsion caused a change in the moiré potential experienced by the IX (like in <https://doi.org/10.1021/acs.nanolett.3c01160>), or is it because there are multiple IX energy levels due to a deep trap? <

As addressed in point 4, we expect that IX2 originates from the quantized energy levels within a deep moiré potential. The combined effect of exciton confinement arising from moiré potential and repulsive dipolar interaction leads to the appearance of IX2 with increasing pump power.

>> To make the exciton physics about the different interlayer excitonic states clearer, we improved the discussion about pump-power dependent behaviors in Fig. 1c and 1d, see **lines 210-218 and 220-221**.

> 9. Will these two mechanisms result in different power dependence for IX2?

<

Yes, the combined effect of the two mechanisms leads to a higher saturation power of IX2 than that of IX1, as shown in Fig. 1d.

> 10. The authors observed a low-energy tail for the large twist angle ($>1^\circ$) samples. In the discussion, they said it is similar to the phonon sideband in monolayer WSe₂. However, in the following sentence, they said it comes from defects. Could the authors clarify which of these two is the mechanism they believe to be the main reason for the low-energy tail? <

Thank you for pointing out this inconsistency and helping us realize that our writing logic here is jumpy. We relate the phonon sideband in monolayers to defects due to the following reasons:

1) TMD monolayers are actually quite defective. While 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 (see ref. Nano letters 19, 4371 (2019)). The physical picture of the single photon emitters in TMD monolayers is commonly described as the localization of excitons in defects (Phys. Rev. Lett. 123, 146401 (2019)). The presence of high-density defects can also be reflected by the observation of quantum emitters in TMD monolayer/pillar structures (Nature Communications 8, 15053 (2017) and Nature Communications 8, 15093 (2017)) 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 ACS Nano 11, 5179–5185 (2017), the low-energy tail of PL peak of MoS₂ is suppressed after the passivation of non-radiative defects (see Fig. R2). 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.

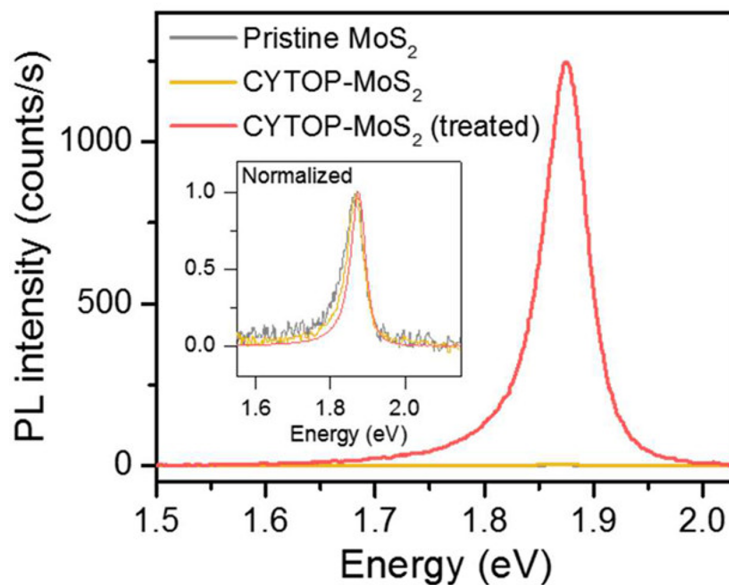


Figure R2: Defect passivation of monolayer MoS₂, taken from ref. ACS Nano 11,

5179–5185 (2017).

3) The phonon sidebands typically result in a low-energy tail of the PL peak of TMD monolayers, see ref. Phys. Rev. Research 3, L042019 (2021) and ACS Photonics 7, 2756 (2020).

In stark contrast, our samples with a small twist angle ($\sim 0.5^\circ$) show high-energy tails of PL peaks and its presence is due to the quantized energy levels within a moiré potential trap, which is discussed in the main text. On the other hand, we believe that non-radiative defects are also present in the moiré superlattices with small twist angles. The invisible low energy tail can be ascribed to the passivation of non-radiative defects by moiré potential.

Your comments help us realize that the focus of the corresponding section is to show a clear physical picture of the appearance of the multiple exciton states.

» We added a detailed discussion about the different exciton states and moved the part about the possible origins of the energy tails to the new Note S1.

> **11. These two mechanisms differ in temperature dependence behavior, i.e., increasing the temperature will induce detrapping while increasing the effect of the phonon sideband. Could the authors provide such a temperature-dependence study to justify the origin of the low-energy tail?** <

As you suggested, we conducted additional measurements by increasing the temperature and measuring the PL spectrum, as shown in Fig. R3. We observe that the peak intensity decreases with increasing temperature, and the energy redshifts while the shape remains unchanged.

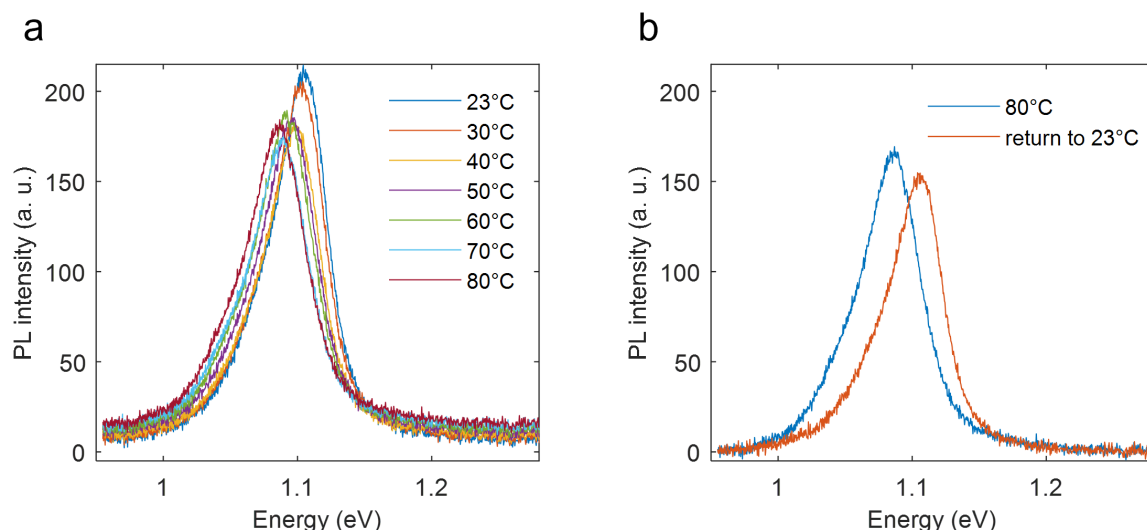


Figure R3: Temperature dependence of the $\sim 2.8^\circ$ MoS₂/WSe₂ heterobilayer. (a) PL spectrum at temperatures from 23 °C to 80°C. The redshift is due to the reduced bandgap upon increasing temperature. The decrease in PL intensity is mainly ascribed to the degradation of sample quality after heating, which is evidenced by the comparison of the PL spectrum shown in (b).

Furthermore, 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. R4). Combined with the reasons mentioned in point 10, we expect that the non-radiative centers or defects are always there, no matter the twist angle. The key point should be the interplay between non-radiative centers and moiré effect. According to the observation in Fig. 3, we expect that the moiré effect 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 to solve the issue.

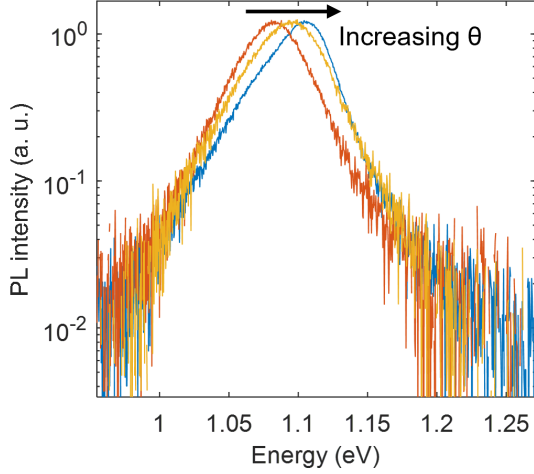


Figure R4: Comparison of PL spectrum of samples with large twist angles. The twist angles are derived by the empirical equation shown in Note S4.

» We added Note S1 to discuss the energy tail of the PL peaks.

> **12. In Fig. 2a, one of the samples is uncoupled bilayer (2L) MoS₂/WSe₂. What is the meaning of uncoupled here, and how could it be ensured that it is uncoupled?** <

The 'uncoupled' means that the interlayer coupling between bilayer MoS₂ and WSe₂ is poor, which is evidenced by the bright WSe₂ A-exciton emission from the formed heterostructure (orders of magnitude larger than the well-coupled heterobilayer, see Fig. R5) and the unshifted WSe₂ absorption peak compared to that of WSe₂ monolayer (see Fig. 2a). We choose this sample due to two main reasons: 1) it clearly shows that the exciton resonance does not shift if the interlayer coupling is poor; 2) the exciton energy of 2L-MoS₂ is a good indicator for interlayer coupling. The exciton energy of monolayer MoS₂ shifts to a lower value when stacked with WSe₂. Naturally, we would expect that the exfoliated bilayer should have a better interlayer coupling than the stacked heterobilayer. In this case, we can gain more insight into the moiré superlattices by comparing their MoS₂ exciton resonances to that for the bilayer MoS₂.

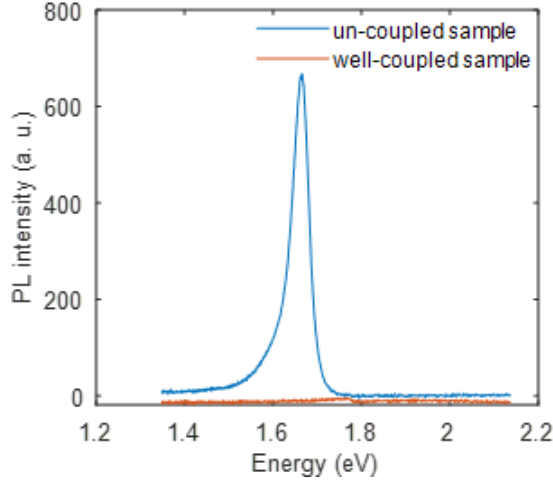


Figure R5: Comparison of WSe₂ A-exciton emission from uncoupled 2L-MoS₂/WSe₂ and well-coupled MoS₂/WSe₂. The strong WSe₂ exciton emission, together with the unchanged exciton absorption energy (see Fig. 2a) compared to that of monolayer WSe₂, confirm the negligible interlayer coupling between 2L-MoS₂ and WSe₂.

≫ We added an explanation how we identify the uncoupled and well-coupled heterostructures in [lines 250-252](#) and use Fig. R5 as a subplot of Fig. S1 to show their different PL quenching of WSe₂ excitons.

> 13. In Fig. 2c, $X_{MoS_2}^A$ in MoS₂/WSe₂ heterobilayer has energy below that in 2L-MoS₂. The authors said that this must be due to expanded lattice constant. However, it is also possible that this is caused by the influence of the moiré superlattice since the superlattice potential can create moiré minibands with energy below the MoS₂ monolayer conduction band or higher than MoS₂ monolayer valence band. How could this effect be ruled out? <

Interlayer coupling and lattice constant are the two factors that have the greatest impact on exciton energy. We expect that the exfoliated bilayer MoS₂ has a better interlayer coupling than the stacked MoS₂/WSe₂ heterobilayers. Therefore, we believe that the energy shift of the exciton resonance with twist angle is not just because of the interlayer coupling but also due to changes in the lattice constant.

Furthermore, the observation of moiré pattern in both the height channel and stiffness channel manifests the spatial modulation of the lattice constant, indicating the presence of atomic reconstruction. It is worth noting that the energy of $X_{MoS_2}^A$ is related to atomic reconstruction (feature as periodically modulated lattice constant), which does not mean that the moiré potential can be excluded; instead, the reconstruction can lead to a deeper moiré potential or more flat bands that promote exciton localization, see ref. Science 378, 1235-1239 (2022). Therefore, we believe that the moiré effect plays a crucial role in tailoring the exciton energy and cannot be ruled out.

≫ We added an additional description to relate the atomic reconstruction and moiré effect, see [lines 337-339](#).

> 14. The authors said: "... , monolayer WSe₂ with a smaller lattice constant (0.328 nm) expands while monolayer MoS₂ with a larger lattice constant

(0.316 nm) shrinks.” This statement is strange. First, $0.328 > 0.316$, shows the statement must be wrong. Instead, I expect WSe₂ with a larger lattice constant to shrink while MoS₂ expands. <

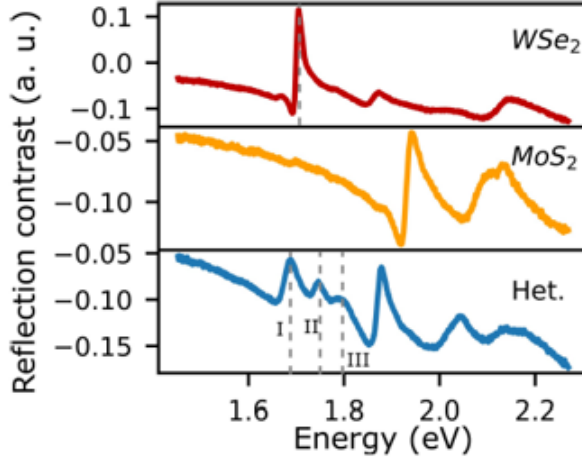
We greatly appreciate you for pointing out our mistake. The fact is that WSe₂ with a larger lattice constant shrinks while MoS₂ expands.

» We corrected this in [lines 317-319](#).

> 15. The authors observed that, for a twist angle $< 0.9^\circ$, an additional higher energy state is detected in the reflection contrast. First, I want to confirm that this is related to the WSe₂ exciton peak (which seems to be the case based on Fig. 2a). If this is the case, why is it absent in the MoS₂ exciton peak? Given the reason is that this is intralayer exciton localized at a different site in a moiré cell, shouldn’t a similar feature be observed for the MoS₂ exciton peak also? Is it possible that this is not due to such a reason, but other reasons, including phonon-replica or intervalley exciton? <

Excitons can be captured in the different atomic registries of moiré superlattices, and the absorption varies for different localized excitons, see ref. Nature 567, 76–80 (2019). In our case, although a different combination of TMD monolayers was used, different atomic registries were created after stacking. The observation of the emergence of a higher-energy state of WSe₂ exciton at a small twist angle was used as evidence for the emergence of a moiré pattern, see ref. Nature 603, 247–252 (2022). Additionally, the presence of moiré pattern is manifested by the AFM measurements, see Fig. 2c and Fig. S4. Therefore, we expect that the varying absorption and/or potential depth for the localized excitons results in the different responses of WSe₂ and MoS₂ excitons. To further examine the localization of interlayer excitons, the electron-beam-based hyperspectral imaging reported in ref. Science 378, 1235–1239 (2022) can possibly provide the answer. However, it is out of reach of our current measurement setups, and we are seeking new collaborations to answer this question.

Although the formation of a moiré pattern is commonly used to explain the observation of multiple WSe₂ exciton resonances at small twist angles, the absence of the multipeak feature for MoS₂ exciton has not been discussed, see the previous report shown in Fig. R6. Therefore, we appreciate that you put forward new possible origins for this phenomenon. Phonons in 2D materials can cause phonon-replica and intervalley exciton, which are potentially affected by the moiré pattern (see ref. Nature Materials 20, 1100–1105 (2021)). In the MoS₂/WSe₂ heterobilayers that we studied, the modulation of phonons by the moiré pattern has remained unexplored. At present, it is not clear how the moiré pattern can affect phonons and consequently cause different responses of WSe₂ exciton and MoS₂ exciton. Studying the contribution of phonons on exciton absorption is interesting and challenging. While it is out of the scope of this work, we are inspired to explore this direction in the future. Moreover, in Fig. 2, the exciton resonances are determined in the RC measurement, which is expected to characterize only direct valley transition states. Therefore, we do not expect the contribution of intervalley excitons in the RC spectra.



Extended Data Fig. 1 | Reflection contrast spectra of the sample. Top, reflection of the 1L WSe_2 region, showing its lowest excitonic resonance at 1.7 eV, marked with a grey dashed line. Middle, the same, for 1L MoS_2 , with its lowest resonance at 1.95 eV. Bottom, the same, for the heterobilayer region. It shows several resonances, marked I, II and III, instead of a single line in the vicinity of the WSe_2 resonance. This signature has been categorized as evidence for the emergence of the moiré pattern¹⁶. All measurements were done at 80 K.

Figure R6: Refection contrast spectrum of a $\text{MoS}_2/\text{WSe}_2$ heterobilayer, taken from ref. Nature 603, 247–252 (2022).

» We added a discussion on the absence of splitting of MoS_2 exciton in lines 343-345.

> 16. The authors said that the equation, $E(\theta) = a - bc^\theta$, in Note S2, implies that the significant energy change of IX is related to the interlayer spacing between monolayers. Why is that so? <

Thank you for finding this unclear description. Regarding the energy of interlayer excitons, the energy shift can be ascribed to the compensation of momentum mismatch and the increased depth of the moiré potential upon decreasing twist angle, similar to ref. Science 376, 406-410 (2022). Additionally, we find that the twist-angle dependent energy shift can be up to ~ 224 meV according to the equation, which in our case is significantly larger than that (~ 60 meV) observed in $\text{MoSe}_2/\text{WS}_2$ moiré superlattices (see ref. Nature 567, 81–86 (2019)). This finding reflects that the moiré potential in our system is more significant. Furthermore, the moiré potential strongly depends on interlayer coupling, as evidenced by the sufficient suppression with an additional monolayer hBN in the TMD heterobilayer, see ref. Nature 580, 472–477 (2020). Therefore, we expect that the significant energy described by the equation implies the change of interlayer spacing between monolayers.

» We improved our description by adding the above explanation, as shown in lines 724-735.

***** Addressing Report of Reviewer 2 *****

> 1. The manuscript by Q. Lin & H. Fang et al. reports optical properties of twisted $\text{MoS}_2/\text{WSe}_2$ heterostructures at room temperature. The optical

spectrum is investigated using luminescence and reflectance spectroscopy, and the morphology is studied by atomic force microscopy. Moreover, the authors place hBN encapsulated heterostructures on top of a nanobeam cavity to study the coupling of differently confined interlayer excitons with cavity photons. The discussion of the data presented in Figs. 1-3 is sound and the conclusions seem solid. Since the authors mention light-matter interactions in their title, it would be nice if they presented a consistent analysis of the oscillator strength as a function of twist angle. The analysis and discussion of the data shown in Fig. 4, however, is quite unsatisfactory (see comments below). In the current presentation, I do not see any interesting insights from this part of the manuscript. As outlined below, it is not clear why the results are discussed in the context of lasing and threshold behavior, as the effects involved might simply result from the coupling of the PL emission to cavity modes and Purcell physics, which is all well explored. If the authors were able to show compelling evidence that different localization of moiré excitons can significantly improve optical properties at room temperature, I would be willing to consider recommending the manuscript for publication. Currently, however, I am not convinced. <

Thank you for your insightful comments and for raising important points. We would like to analyze the oscillator strength of the moiré superlattice as a function of twist angle. However, the weak optical absorption of interlayer exciton hinders us from performing the measurements. According to Fig. 3 and Fig. S6, the regions with small twist angles show significantly stronger PL emissions than those from the regions with larger twist angles, indicating the improved optical properties of the heterobilayers with moiré effect.

Furthermore, we note that the cavity-enhanced emission cannot only be explained by the coupling of the PL emission to cavity modes and Purcell physics, but it is also related to the optical properties of the gain material. To evaluate the significance of the origin of optical gain, we prepared reference samples where nanobeam cavities are covered by bilayer MoTe₂ that intralayer excitons dominate the optical properties of the host material, as shown in Fig. R7. We observe a prominent PL emission from bilayer MoTe₂, but cavity mode is absent when the cavity is fully covered by MoTe₂, that is, the absorption of intralayer exciton dominates. Additionally, in the case that cavity resonance is well below the intralayer exciton resonance (i.e., low optical absorption), the cavity mode becomes prominent and lasing-like behaviors were observed, see the previous work *Advanced Optical Materials* 7, 1900538 (2019).

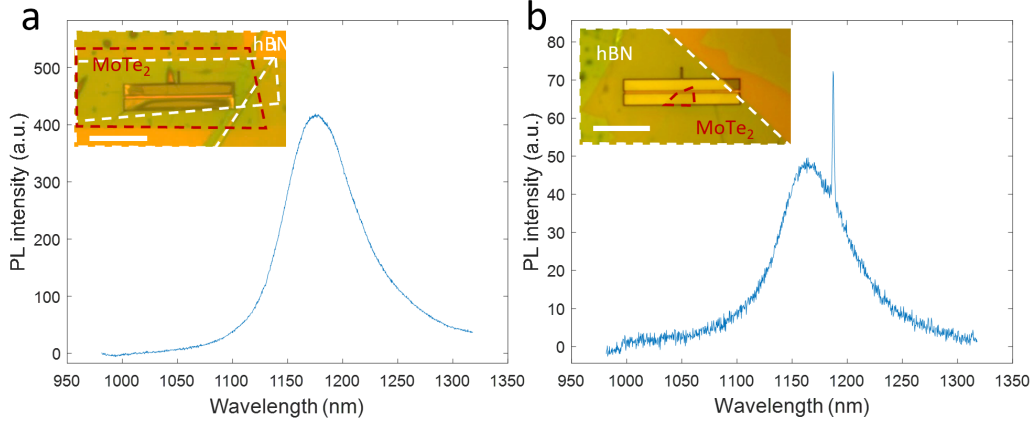


Figure R7: Bilayer MoTe₂-cavity devices. The hBN-caped MoTe₂ flakes fully cover and partially cover the cavities with the same parameters, as shown in Fig. R7a and Fig. R7b, respectively. The samples are characterized by the same PL measurement setup. The partially-covered sample with only a small fragment 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 un-pumped MoTe₂ area can impede the formation of the cavity mode.

In contrast to the strong absorption of intralayer excitons in MoTe₂, the absorption of interlayer excitons in heterobilayers is invisible, leading to a prominent cavity mode from all moiré superlattice-cavity devices where the cavity is fully covered by heterobilayers. This big difference in the emission behavior between these two types of devices manifests the significance of the material properties including optical absorption and gain.

As mentioned in the main text, we designed and fabricated three representative devices (D1, D2, and D3) with optical cavity resonances that are mainly coupled to three different IX states ($|IX1\rangle$, $|IX2\rangle$, and $|IX3\rangle$), respectively. These three moiré superlattice-cavity devices show a dominant cavity mode (i.e., a significant PL enhancement over emission background) at different pump power densities, see Fig. S10. The enhancement factor of D3 reaches approximately 3 at a high pump power density of 33.97 kW/cm², while D1 shows an enhancement factor of about 6 at a low pump power density of 1.39 kW/cm². It is worth noting that the 150 grooves/mm grating facilitates the observation of PL emission background but results in a decreased enhancement factor due to the broadened linewidth. Furthermore, the contrasting thresholds derived from L-L curves (see Fig. S12) for these three devices reflect different optical gains provided by the various excitonic states. We emphasize that D1 and D2 devices show a one-order of magnitude lower threshold when compared with the D3 (see Fig. S12), implying the significance of moiré effects. Understanding and quantifying the gain contribution from the different excitonic states is challenging but meaningful, and it will be one of our focuses in the future.

» We added Fig. R7 into the SI and labeled it as Fig. S9. Moreover, we added corresponding discussions in [lines 465-471](#).

> 2. The authors mention lasing and talk about the threshold behavior of the power-dependent PL intensity they measure from three different samples on top of the nanobeam cavity. I noticed a few issues: a) It is repeatedly mentioned that the cavity is “topological”, yet besides a reference, neither

the impact of such properties on the optical mode spectrum nor the emission of the heterostructures are discussed in the manuscript. What are significant differences of the "topological nanobeam cavity" compared to any conventional nanobeam cavity? <

We agree that it is not necessary to repeatedly mention the 'topological', which is actually not the focus of this work. The design of the cavity has been realized in previous works (Communications Physics 1, 86 (2018) and Advanced Optical Materials 7, 1900538 (2019)) and mentioned in Methods. We use the design mainly because of its single-mode operation that can simplify our study of the coupling between different interlayer exciton states and cavity modes. These topological cavities typically feature single-mode operation and enable a lower lasing threshold compared to conventional cavities, see ref: Bandres et al., Science 359, eaar4005, (2018).

>> We have replaced some 'topological' with 'single-mode' in the revised manuscript.

> 3. b) All relevant data is shown in Supplementary Figs. 9 & 10, while Fig. 4 of the main manuscript merely shows the result of an analysis (cf. Fig 4e) that is not very well explained in the manuscript. What is the spectral range the authors use to integrate the PL peak intensity? The background emission clearly varies significantly, which – depending on the exact analysis – can easily lead to artifacts and the features the authors use to make claims about the different "threshold" of differently localized moiré excitons. It is further not clear to me what "threshold" even refers to in the absence of clear evidence for lasing. <

Thank you for guiding us to improve the description of the analysis of Fig. 4e. We agree that the background emission varies for the different devices and the D3 shows the most significant PL background. Let us take D3 as an example. The integrated PL intensity is extracted by fitting the cavity mode with a Lorentzian function, and thus the PL background is not included (see Fig. R8). Moreover, according to the power dependence of the different interlayer excitons shown in Fig. 1 and Fig. S2, we do not expect that the PL background can give rise to the observed threshold behavior.

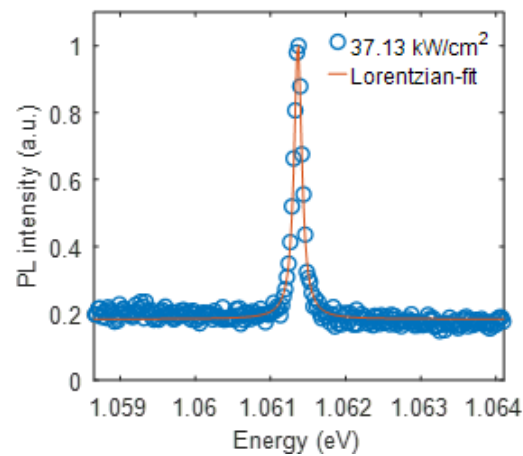


Figure R8: Fitting of the emission peak of D3 with a Lorentzian function. The spectrum is acquired by a grating of 1200 grooves/mm, showing the PL background is less significant than that acquired by a grating of 150 grooves/mm.

Regarding the meaning of "threshold" in our case, we expect that it reflects the occurrence of population inversion in moiré superlattices, i.e., optical gain. According to ref. Light: Science & Applications 6, e17030 (2017), both cavity-enhanced spontaneous emission and stimulated emission show threshold behavior. The difference is that the optical gain in the former case is not large enough to compensate for the system loss (see a detailed discussion in Note S6).

» We improved the description in the caption of Fig. S10 and Fig. S12, added Fig. S16 to illustrate the fitting, and included explanations about our data analysis in [lines 792-795](#).

> **4. I am also not very fond of the definition of the enhancement factor "x41.5". It reflects the difference between the outcoupled emission and that of the background emission. This does not automatically mean enhancement. The factor 41.5 is, for example, impacted by the suppression of background emission by the nanobeam cavity. It might be better to compare the PL from a sample on SiO₂/Si under identical excitation conditions.** <

It is challenging to prepare another sample with a heterostructure with the same twist angle and optical quality on a SiO₂/Si substrate for comparing the PL intensity. Alternatively, we measured the PL spectrum from the heterostructure on the center of the cavity and on the SiO₂ beside the cavity for comparison, as shown in Fig. R9. We clearly see that the PL background emission from on- and off-cavity is similar, and thus the way we define the enhancement factor is reasonable. According to ref. Nature Nanotechnology 12, 987 (2017), the PL background mainly originates from the gain material outside the cavity.

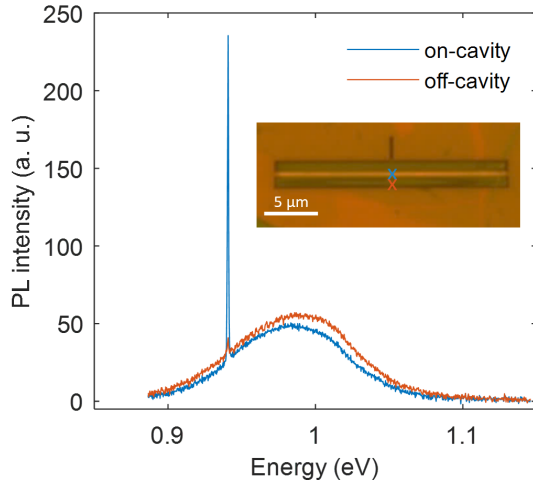


Figure R9: PL emission from the heterostructure on- and off-cavity of D1. It is worth noting that the PL spectra are acquired by a grating of 150 grooves/mm to capture the PL background, resulting in an averaged cavity emission peak (i.e., the linewidth broadens and peak intensity decreases) compared to the data in Fig. 4e acquired by a grating of 1200 grooves/mm.

» Fig. R9 is added into supplementary information and named Fig. S11.

> **5. The authors fail to mention a recent work on moiré excitons coupled to nanocavities that clearly shows lasing at low temperatures: Qian et al., Sci. Adv.10, eadk6359 (2024). In particular, to justify the term "threshold", I**

would expect to see behavior similar to the power-dependent emission in Fig. 2A. <

Thank you for reminding the newly published work about moiré exciton lasers, and now we cite the paper in the Introduction of the revised manuscript, see [line 95](#). To see if our moiré exciton-cavity devices show similar power-dependent emission to Fig. 2A of the ref. Sci. Adv.10, eadk6359 (2024), we plot the L-L curve of D2 on a log-log scale, which shows a visible threshold (see Fig. R10).

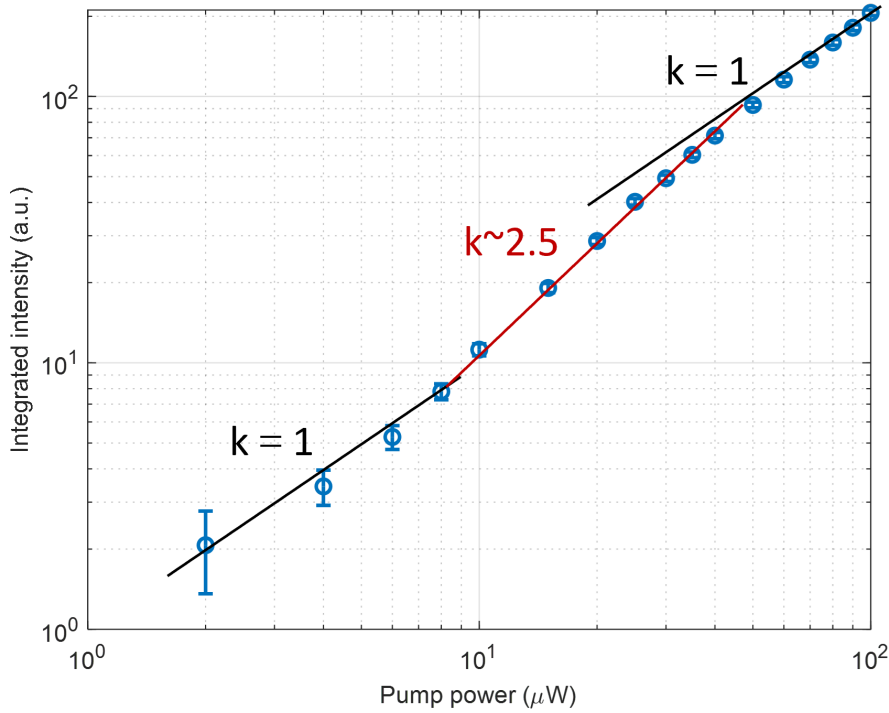


Figure R10: L-L curve of D2 on a log-log scale. The integrated intensity is obtained by fitting the cavity mode with a Lorentzian function.

We observe a clear S-shape like the one in Fig. 2A of the suggested paper and hope it is good enough for examining the lasing action. However, we notify that both cavity-enhanced spontaneous emission and stimulated emission show an S-shape L-L curve, see ref: Light: Science & Applications 6, e17030 (2017). Therefore, we expect that the conclusive demonstration of lasing-dominated emission requires the measurement of photon statistics by the Hanbury-Brown and Twiss experiment, as discussed in Note S6. Correlating the photon statistics with the L-L curve is an interesting and inspiring direction for future work.

» We added an additional description to inspire future work on correlating photon statistics with the L-L curve, see [lines 811-813](#).

***** Addressing Report of Reviewer 3 *****

> 1. The authors measure MoS₂/WSe₂ heterobilayers at room temperature and identify the interlayer exciton (IX) emission at ~ 1 eV. For heterobilayers with small twist angles, the authors find saturation behaviour on the PL emission with increasing optical pump power. They also report that when decreasing the twist angle, the IX lifetime becomes longer and light emission is enhanced. Finally, heterobilayers with small twist angles are then integrated with silicon topological cavities. First of all, I agree with the authors that the presented data supports the conclusions drawn in the first section “Localized interlayer excitons at room temperature”. Indeed, the authors supply sufficient experimental evidence to demonstrate the observation of confined IX states at RT. <

Thank you for summarizing the experiments we performed and for supporting our statements regarding exciton localization at room temperature.

> 2. Their data shows strong dipole-dipole repulsion. With respect to the next section, titled “Features of intralayer excitons in moiré superlattices” I have a few questions. The authors provide a set of experimental data showing clear twist dependent phenomena on the RC spectra acquired in a set of samples. While I support the claim that the experimental data is indicative of the presence of moiré patterns in their samples, I do not quite agree with the explanation provided by the authors. In the manuscript, the authors explain the observed phenomena by two effects, biaxial strain and atomic reconstructions, and provide an AFM showing the reconstruction effects. Unfortunately, I am not able to see the reconstructed domains in the AFM scan. <

We totally agree that the moiré pattern in the height channel shown in the inset of Fig. 2c is a little bit noisy. The signal-to-noise is not good due to the small modulation height of the moiré pattern and the resolution limit of our AFM. A much clearer moiré pattern in the stiffness channel can be found in Fig. S4. Moreover, such a pattern becomes invisible for the sample with a larger twist angle of 4.4° (see Fig. S3), which is consistent with ref. Nature Physics 10, 451–456 (2014) and rules out the measurement artifacts. To make the moiré pattern in the inset of Fig. 2c more distinguishable, we place it together with the one in Fig. S4, as shown in Fig. R11.

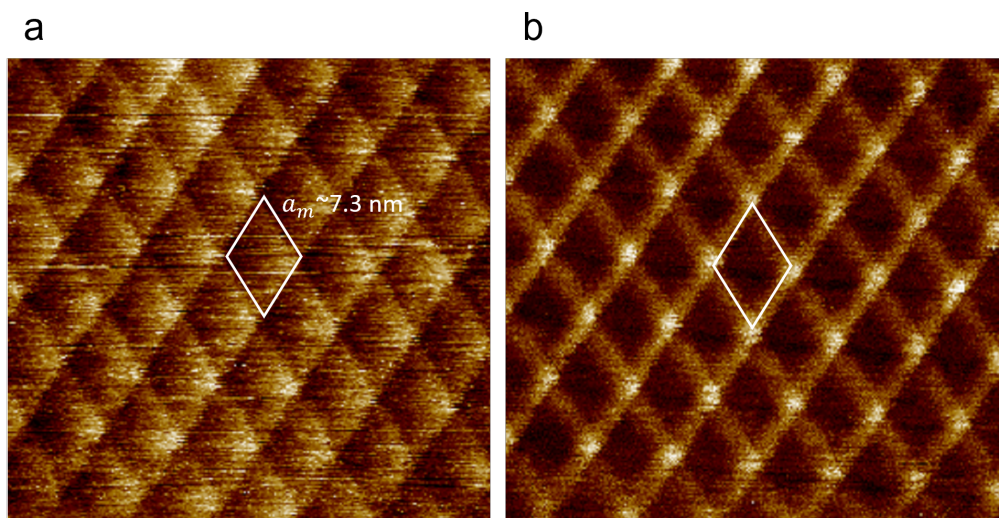


Figure R11: Moiré pattern in height channel (a) and stiffness of AFM measurement (b).

In our case, we chose the noisy moiré pattern in the height channel as the inset of Fig. 2c because it directly manifests that the constituent monolayers cannot be treated as rigid lattices, i.e., atomic reconstruction happens. Additionally, to the best of our knowledge, this is the first demonstration of the moiré pattern in the height channel of AFM measurements. The occurrence of atomic reconstruction in the heterobilayers with large lattice mismatch leads to significant strain distribution and thus alters the exciton energy in the monolayers (see Fig. 2a).

» We add the reasons why we choose the moiré pattern in the height channel in [lines 330-332](#).

> **3. Moreover, the measured energy dependence on the WSe₂ and MoS₂ A-excitons can be explained by considering the arise of resonant hybridized states and moiré minibands on the electronic structure of the heterobilayers, as reported in Ref. <https://doi.org/10.1038/s41586-019-0986-9>. In fact, the measured RC spectra follows the behaviour predicted on that article in Fig. 3e.** <

It is great to see similar behaviors in different semiconducting moiré superlattices, which will be helpful for the discovery of the main mechanisms that dominate its excitonic effects. In ref. <https://doi.org/10.1038/s41586-019-0986-9>, WS₂/MoSe₂ heterobilayers have a band offset of about 20 meV, see ref. Appl. Phys. Lett. 102, 012111 (2013). This small band offset promotes the hybridization between the conduction bands of the constituent monolayers. In our case, the band offset of MoS₂ and WSe₂ is hundreds of meV, leading to interlayer exciton energies far below those of intralayer excitons, see Fig. 1 and Fig. 2. Therefore, we do not expect the contribution of hybridization in MoS₂/WSe₂ heterobilayers. Instead, both types of heterobilayers should have atomic reconstruction at small twist angles, leading to flat bands (see ref. Nature Materials 20, 945–950 (2021)), and thus we expect that reconstruction is the main reason why we see similar twist angle-dependent RC spectra.

> **4. Now I will focus on the next section “Engineering exciton lifetimes with moiré potentials”. I would really appreciate if the authors could comment on the stacking angle of regions 1 and 2, since without that information the discussion it is hard to follow.** <

We greatly appreciate your constructive suggestion about adding the twist angles of the different regions.

» We add the twist angles in the corresponding positions, see the changes in [lines 365-366 and 941-942](#).

> **5. I would have expected that the difference in the exciton lifetimes are due to the tuning of momentum mismatch for different stacking angles. I think it is remarkable that the authors observe the opposite behaviour. To make this point clearer, I would suggest to modify the lines 357-376 to “This feature cannot be explained by the reduced momentum mismatch that typically leads to a shorter exciton lifetime, which is the opposite of the observed trend in Fig. 3”.** <

Thank you for helping us improve our explanation.

» The change is shown in [lines 396-397](#).

> 6. Moreover, how does the fact that the IX exciton lifetime is independent of pump power relate to the reported phenomena on the first section on the manuscript, where the authors reported a strong dipole-dipole interaction as a function of pump power? What is the reason behind the absence of any impact from this dipole interaction on the measured lifetime? <

Without the potential traps, the interlayer excitons can be transported to other positions through repulsive interaction, as demonstrated in ref. Nature Photonics 16, 79–85 (2022) where the moiré potential is suppressed by a hBN spacer. The traveling excitons can easily be captured by other origins such as defects, see Nature Communications 13, 5354 (2022). In two-dimensional TMDs, the exciton lifetime typically decreases upon increasing the pump power owing to enhanced Auger-induced exciton-exciton annihilation, see ref. Nano Lett. 14, 5625–5629 (2014). The passivation of non-radiative centers can significantly prolong the exciton lifetime, see ref. ACS Nano 11, 5179–5185 (2017).

In our case, we discovered a significant moiré potential that suppresses the non-radiative centers, evidenced by observation of a brighter exciton with a longer lifetime, see Fig. 3b and 3c. On the other hand, the strong dipolar interaction reflects that excitons are well-localized in moiré potential traps. In stark contrast, the sample with a large twist angle (i.e., negligible moiré potential) does not show the dipolar interaction, see Fig. S2. Therefore, we expect that the weak dependence of the IX1 lifetime on pump power results from the protection of moiré trapping against exciton-exciton annihilation.

» We improved our explanation in [lines 405-407](#).

> 7. In the last section of the manuscript the authors present optical spectra of heterobilayers embedded in silicon cavities. L-L curves are used to obtain threshold values for different IX-cavity detunings. They show that the IX photon collection efficiency improves by tuning the cavity mode to the IX emission. I agree with the careful treatment of the data and the conclusions drawn, stating that the measurements hint at rich physics but are not conclusive to claim lasing behaviour. In lines 732-734, the authors explain that the D1 cavity might have a low threshold power that could not be measured. I was wondering why the authors could not apply less power to the sample, since the low power point in Fig. S10 is at $\sim 0.5 \text{ kW/cm}^2$ and the authors claim a $1.6 \mu\text{m}$ spot size, giving rise to $0.5 \text{ kW/cm}^2 \times \pi \times (1.7 \mu\text{m}/2)^2 = 12 \mu\text{W}$ applied on the sample, and typical optical experiments are able to apply $\sim \text{nW}$ laser powers. <

We totally agree with you that typical optical experiments can apply laser powers in the order of 1 nW, which is also accessible in our lab. In our case, the challenge is to detect the weak cavity mode under a small pump power. The detector is cooled electrically rather than with liquid nitrogen, and thus limits the sensitivity of our measurement setup for detecting weak signals. The extracted threshold of 0.11 kW/cm^2 corresponds to a power as low as $2.2 \mu\text{W}$, impeding us from observing the cavity mode and then deriving the threshold power of D1.

> 9. Overall, I think the work presented is novel and can be published, provided that the authors answer the questions presented above. <

We greatly appreciate all your helpful suggestions and comments that improved our manuscript and a clear recommendation to publish our work in Nature Communications. We hope we have answered your questions well.

REVIEWERS' COMMENTS

Reviewer #1 (Remarks to the Author):

The authors have satisfactorily addressed my concerns in the rebuttal letter and the revised draft. The origin of different IXs is now explained clearly. The arguments for the evidence of moiré IX and possible lattice reconstruction contribution are now quite convincing.

Considering this and their answers to other referees' questions, I support the publication in Nature Communications.

I have only one minor correction: The unit of exciton density and moiré density (line 695-696) should be cm^{-2} instead of cm^2 .

Reviewer #2 (Remarks to the Author):

All queries have been addressed. I recommend the manuscript for publication.

Reviewer #3 (Remarks to the Author):

I am satisfied with the corrections and explanations added by the authors. Therefore, I would be pleased to accept it for publication.

POINT BY POINT RESPONSE TO REVIEWERS' COMMENTS

We answer the referees' criticism point-by-point. To this end, we keep the order of the referees' comments and mark it > **in blue and bold font** <. Our answers are in black and normal text.

***** Addressing Report of Reviewer 1 *****

> **1. The authors have satisfactorily addressed my concerns in the rebuttal letter and the revised draft. The origin of different IXs is now explained clearly. The arguments for the evidence of moiré IX and possible lattice reconstruction contribution are now quite convincing. Considering this and their answers to other referees' questions, I support the publication in Nature Communications.**
<

We are delighted to have fully addressed your questions and greatly appreciate your recommendation for our work's publication in Nature Communications.

> **2. I have only one minor correction: The unit of exciton density and moiré density (line 695-696) should be cm^{-2} instead of cm^2 .** <

Thank you for finding this mistake. Now we have corrected it in the revised version. To further improve our manuscript, we have rearranged the figures in the Supplementary Information so that they are cited in sequential order in the main text, with the content remaining unchanged.

***** Addressing Report of Reviewer 2 *****

> **1. All queries have been addressed. I recommend the manuscript for publication.** <

We are delighted to have fully addressed your questions and greatly appreciate your recommendation for our work's publication in Nature Communications.

***** Addressing Report of Reviewer 3 *****

> **1. I am satisfied with the corrections and explanations added by the authors. Therefore, I would be pleased to accept it for publication.** <

We are delighted to have fully addressed your questions and greatly appreciate your recommendation for our work's publication in Nature Communications.