Site-controlled interlayer coupling in WSe₂/2D perovskite heterostructure

Qinglin Wei¹, Xinglin Wen^{1,2*}, Junchao Hu¹, Yingying Chen¹, Zeyi Liu¹, Tinghao Lin¹ and Dehui Li^{1,2*}

ABSTRACT Interlayer excitons (IXs) formed in transition metal dichalcogenides (TMDs)/two-dimensional (2D) perovskite heterostructures are emerging as new platforms in the research of excitons. Compared with IXs in TMD van der Waals heterostructures, IXs can be robustly formed in TMDs/2D perovskite heterostructures regardless of the twist angle and thermal annealing process. Efficient control of interlayer coupling is essential for realizing their functionalities and enhancing their performances. Nevertheless, the study on the control of interlayer coupling strength between TMD and 2D perovskites is elusive. Therefore, we realize the control of interlayer coupling between monolayer WSe₂ and (iso-BA)₂PbI₄ with SiO₂ pillars *in situ*. An abnormal 10-nm blue shift and 2.5 times photoluminescence intensity enhancement were observed for heterostructures on the pillar, which was contrary to the red shift observed in TMD heterobilayers. We attributed the abnormal blue shift to the enhanced interlayer coupling arising from the reduced gap between constituent layers. In addition, IXs became more dominant over intralayer excitons with enhanced coupling. The interlayer coupling could be further engineered by tuning the height (*h*) and diameter (*d*) of pillars. In particular, an additional triplet IX showed up for the pillar with an *h/d* ratio of 0.6 due to the symmetry breaking of monolayer WSe₂. The symmetry breaking also induced an anisotropic response of IXs. Our study is beneficial for tuning and enhancing the performance of IX-based devices, exciton localization and quantum emitters.

Keywords: transition metal dichalcogenides, 2D perovskites, interlayer exciton, interlayer coupling strength, pillar

INTRODUCTION

Van der Waals (vdW) stacking of two distinct transition metal dichalcogenide (TMD) monolayers usually features a type-II band alignment. Charge transfer between two layers results in a population of electrons in the lower energy conduction band (CB) and holes in the higher energy valence band (VB). Although electrons and holes reside in separate layers, the atomically thin nature of TMDs makes them strongly bond together *via* Coulomb interaction, thus facilitating the formation of interlayer excitons (IXs) [1–5]. The reduced overlap of electrons' and holes' wavefunctions caused by spatial separation leads to a lifetime as long as tens of nanoseconds, which is several orders of magnitude longer than intralayer excitons (tens of picoseconds) [1,6]. In addition, the presence of out-of-plane

dipole moment makes it feasible to control IXs with an electric field, which is demanded in excitonic circuits [7–9]. IXs also carry the valley information and have applications in valleytronics [10,11]. The emergence of IXs promotes fundamental research of many-body interactions [12] and exciton condensation [13]. It also triggers applications in excitonic circuits and valleytronics.

However, properties of IXs in TMD heterostructures highly depend on the twist angle between adjacent layers due to the momentum mismatch, making it difficult to fabricate samples with consistent performance. The formation of moiré potential in vdW heterostructures will change the dynamics and transport properties of IXs significantly [14,15]. Alternative to TMD vdW heterostructures, TMDs can also be combined with other layered materials to achieve interlayer coupling [16–19]. Recently, it has been demonstrated that robust IXs could be observed in TMD/two-dimensional (2D) perovskite heterojunctions with various material combinations [20]. IXs in TMD/perovskites have the following advantages: (i) thermal annealing is not necessary during fabrication; (ii) optical responses are independent of the twist angle; (iii) various perovskites with a tunable bandgap can produce IXs with a broad wavelength range [20]. In other words, 2D perovskites possess advantages of easy synthesis, diverse material library, tunable bandgap, and large exciton binding energy; thus, they provide a new platform for research of excitons [21–26].

The optical properties of IXs can be dramatically manipulated by engineering interlayer coupling. For instance, strong coupling yields pure IXs and intralayer excitons vanish, while IXs and intralayer excitons occur simultaneously in a moderate coupling regime, and only intralayer excitons are present for weak coupling [27–29]. For TMD heterostructures, various strategies have been used to engineer the coupling strength. For example, strong coupling of WSe₂/MoSe₂ heterobilayers could be achieved by applying high pressure to reduce the gap between constituent layers [29]. It has also been demonstrated that the resonance wavelength, exciton localization, and anisotropy of IXs could be largely tuned *via* strain [30–33]. In addition, moiré potential could modify IX dipole-dipole interactions [15,34]. Though several approaches have been used to control the coupling between TMD layers, the interlayer coupling manipulation between TMD and 2D perovskites is still elusive. Optical responses of IXs, such as spectra line shape, resonance wavelength, photoluminescence (PL) intensity, and the ratio between IXs and intralayer excitons, highly depend on the interlayer coupling. It appeared that the optical responses of IXs of TMDs/

¹ School of Optical and Electronic Information, Huazhong University of Science and Technology, Wuhan 430074, China

² Wuhan National Laboratory for Optoelectronics, Optical Valley Laboratory, Huazhong University of Science and Technology, Wuhan 430074, China

* Corresponding authors (emails: wenxl@hust.edu.cn (Wen X); dehuili@hust.edu.cn (Li D))

2D perovskites were different for individual samples, which may be attributed to the deviation of the interlayer coupling [20,35]. Efficient control of interlayer coupling between TMDs and 2D perovskites is critical for manipulating the optical properties of IXs, realizing their functionalities, and enhancing their performance. Moreover, 2D perovskites are thicker than TMD monolayers. The coupling strength manipulation is different from the TMD/TMD scenario. Therefore, it is desirable to investigate how to manipulate interlayer coupling between TMDs and 2D perovskites.

We present the site-control of IXs in $\text{WSe}_2/(\text{iso-BA})_2\text{PbI}_4$ (iso-BA stands for $\text{C}_4\text{H}_9\text{NH}_3$) heterostructure with SiO_2 pillars. Our results show that the coupling strength is enhanced with a pillar to yield higher IX intensity and IX/intralayer exciton ratio. Contrary to a strain-induced red shift of IX wavelength in TMD vdW heterostructures, the blue shift was observed for $\text{WSe}_2/(\text{iso-BA})_2\text{PbI}_4$ placed on the pillar. This is attributed to the enhanced interlayer coupling strength instead of the bandgap narrowing of WSe_2 . Following the fact that pillars have negligible influence on $(\text{iso-BA})_2\text{PbI}_4$, we interpret that the interlayer coupling manipulation arises from the modified gap between WSe_2 and $(\text{iso-BA})_2\text{PbI}_4$ in the presence of mechanical force on the pillar. In addition, the coupling strength could be tuned by varying the diameter (d) and height (h) of the pillar. It appeared that interlayer coupling strength increased when the h/d ratio changed from 0.2 to 0.4. Specifically, an additional triplet IX was observed when the h/d ratio was further increased to 0.6 due to the out-of-plane symmetry breaking of WSe_2 . The symmetry breaking also induced the anisotropic response of IXs with the linear dichroism of 1.7. The site-control of interlayer coupling is beneficial for tunable IX-based optoelectronic devices and promotes the realization of IX localization and quantum emitter.

EXPERIMENTAL SECTION

Sample preparation

A polymethyl methacrylate (PMMA) layer was spin-coated on a SiO_2/Si substrate with an oxide layer of 100 or 300 nm. Then, electron beam lithography (EBL, TESCAN VEGA3) was used to define the pattern. After developing exposed PMMA, 5-nm Cr was deposited and then a lift-off process was conducted to obtain a Cr array sample. Cr layer served as the mask, and the sample was etched with inductively coupled plasma (ICP) to remove the SiO_2 layer. Subsequently, a SiO_2 pillar array was achieved after removing the residual Cr with HCl. Monolayer WSe_2 was exfoliated from commercial crystal and dry-transferred to an as-fabricated pillar with alignment. Another $(\text{iso-BA})_2\text{PbI}_4$ layer was exfoliated from the synthesized crystal and transferred to the top of WSe_2 with careful alignment.

Optical measurement

An iHR 550 Raman spectrometer equipped with liquid nitrogen charge coupled device was used for optical measurement. The equipment was set to the backscattering mode. All PL measurement was conducted in a cryostat cooled with liquid nitrogen at the excitation of 473 or 633 nm, and the signal was collected with a 50 $\times$ long working distance objective.

RESULTS AND DISCUSSIONS

Fig. 1a shows a typical type-II band alignment of the $\text{WSe}_2/(\text{iso-BA})_2\text{PbI}_4$ heterostructure. A 633-nm (1.96 eV) laser was used to excite the $\text{WSe}_2/(\text{iso-BA})_2\text{PbI}_4$ heterostructure in our measurement to avoid the absorption of perovskites, obtaining a larger penetration depth to excite underneath WSe_2 . The holes would transfer from the VB of WSe_2 to $(\text{iso-BA})_2\text{PbI}_4$ and the

Figure 1 Fabrication of $(\text{iso-BA})_2\text{PbI}_4/\text{WSe}_2$ heterostructure and interlayer exciton (IX) measurement. (a) Illustration of IXs formed in the $\text{WSe}_2/(\text{iso-BA})_2\text{PbI}_4$ heterostructure. (b) Schematic of the heterostructure on SiO_2 pillar. (c) Optical image of the as-fabricated sample, where red and yellow dashed lines indicate the monolayer WSe_2 and $(\text{iso-BA})_2\text{PbI}_4$, respectively. The black square is the PL mapping area in the main text. (d) SEM (top) and AFM (bottom) images of the pillars; the inset is the AFM height profile across a pillar. (e) PL of $(\text{iso-BA})_2\text{PbI}_4$ under 473-nm excitation (black); PL spectra of WSe_2 (blue) and heterostructure (magenta) under 633-nm excitation. X^0 , X^- and IX represent neutral, charged, and interlayer excitons, respectively. The spectra are shifted vertically for clarity. (f) PL of the heterostructure with (w/t) and without (w/o) pillars. Blue shift and enhanced intensity are observed for IX on the pillar.

electrons still resided in the CB of WSe₂. Consequently, IXs between WSe₂ and (iso-BA)₂PbI₄ were formed. To control coupling *in situ*, a SiO₂ pillar array was fabricated with EBL and ICP etching from Si wafer with 100- or 300-nm oxide layer (See details in the EXPERIMENTAL SECTION). Fig. 1b shows a schematic illustration of the as-designed device. Monolayer WSe₂ was exfoliated from a single crystal (HQ graphene) and transferred to the pillar array as highlighted by the red dashed line in the optical image in Fig. 1c. Then, a (iso-BA)₂PbI₄ flake with a thickness of about 20 nm was exfoliated from the crystal and transferred to the top of WSe₂ with precise alignment as highlighted by the yellow dashed line in Fig. 1c. Fig. 1d shows the typical scanning electron microscopy (SEM, top panel) and atomic force microscopy (AFM, bottom panel) images of SiO₂ pillars. They show a diameter of 400 nm and a height of 80 nm (shown as inset). The periodicity of the array was set to 5 μ m to eliminate the influence from adjacent pillars.

For comparison, we plot the PL spectra of (iso-BA)₂PbI₄ (under 473-nm excitation), pristine WSe₂, and WSe₂/(iso-BA)₂PbI₄ heterostructure (under 633-nm excitation) at 78 K in Fig. 1e. Exciton emission of (iso-BA)₂PbI₄ is observed at 525 nm (black curve). Simultaneously, neutral (X⁰) and charged exciton (X⁻) of monolayer WSe₂ are resolved at 710 and 727 nm (blue curve), respectively. IX of WSe₂/(iso-BA)₂PbI₄ is observed at 750 nm along with a shoulder at 720 nm (magenta curve), which is consistent with the previous report [20]. Power-dependent measurement of WSe₂/(iso-BA)₂PbI₄ heterostructure shows that lower energy peak (750 nm) blue shifts with increased power, whereas the higher energy peak (720 nm) is independent of power (Fig. S1, Supplementary Information), verifying that 720

and 750 nm peaks are assigned to X⁰ and IX, respectively. The saturation of the blue shift of the 750 nm peak is due to the saturated exciton concentration with increased power. Furthermore, circularly polarized PL reveals that a higher energy peak has a valley degree, whereas a lower energy peak shows valley independence (Fig. S2), serving as another evidence that the lower energy peak belongs to IX and the higher energy peak belongs to X⁰ of 1L WSe₂. Fig. 1f shows typical PL spectra measured on and outside a pillar ($d = 600$ nm, $h = 240$ nm). It is worth noting that the PL intensity of IX for the sample on the pillar is stronger and the wavelength is blue shifted about 4 nm.

To investigate how the pillars affect the optical responses of IXs, PL mapping of the WSe₂/(iso-BA)₂PbI₄ heterostructure in the vicinity of an isolated pillar (area of the black square in Fig. 1c) was conducted and the intensity of IX peak was integrated, as shown in Fig. 2a. It is clearly seen that IXs are the strongest when the heterostructure is right on top of the pillar, and they decline by about four times when the heterostructure is 4 μ m away from the top. Fig. 2b shows the IX wavelength mapping, and the wavelength is blue shifted about 4 nm on the pillar. The stronger IXs are accompanied by the blue shifted peak on the pillar. It is worth noting that the exciton funneling effect can also increase the PL intensity, in which the strain of the pillar creates a potential well (energy minimum) and the excitons can be drifted from a flat unstrained region to the potential well. Nevertheless, the trap energy level is lower than CB for the funneling effect; thus, IX emission should be red shifted as reported previously [31,32,36], which contradicts the blue shift in our measurement. Thus, exciton funneling is excluded.

Figure 2 PL mapping of IXs. (a) Integrated IX PL mapping around a pillar. The isolated pillar is located at the center of the map. The black dashed arrow indicates the direction where PL spectra are extracted. (b) IX peak wavelength mapping corresponding to (a). (c) Extracted PL spectra at different distances away from the pillar as indicated by the dashed arrow in (a). (d) Extracted IX peak wavelengths and IX/X⁰ ratios at different distances away from the pillar as indicated by the dashed arrow in (a).

In addition, isolated pristine (iso-BA)₂PbI₄ flake and monolayer WSe₂ were exfoliated onto the same pillar array to conduct the measurement separately. By measuring the PL of (iso-BA)₂PbI₄ with and without the pillar, it could be concluded that the pillar almost had a negligible influence on the perovskite because the PL wavelength and intensity were almost unchanged (Fig. S3). For pristine WSe₂, the pillar could induce a 3-nm red shift of neutral exciton and the intensity increased slightly, which is attributed to the pillar-induced strain effects (Fig. S4). As mentioned above, the bandgap narrowing of WSe₂ arising from the strain is incompatible with the blue shift of IX emission on the pillar. We attribute the blue shift to the enhanced interlayer coupling on top of the pillar. A smaller gap between constituent layers leads to enhanced interlayer coupling, leading to a stronger IX dipole moment. Consequently, stronger dipole-dipole repulsion will produce the blue shift of the PL peak. The blue shift of PL of IXs in WSe₂/MoSe₂ heterobilayers has also been reported by reducing the gap between the two layers to enhance interlayer coupling, similar to our scenario [29]. It is assumed that monolayer WSe₂ exhibits a tent-like shape around the pillar, as reported in several studies [17,32,37]. Consequently, the gap between WSe₂ and (iso-BA)₂PbI₄ is shortened on top of the pillar, yielding enhanced interlayer coupling. In addition, the temperature-dependent PL in Fig. S5 shows that heterostructures can sustain IXs at 160 and 140 K for cases w/t and w/o the pillar, respectively, serving another evidence of enhanced interlayer coupling. Since the modulation of IXs arises

from the strain underneath monolayer WS₂, the thickness of perovskite has a negligible effect on the IX modulation. Thus, the stronger PL intensity of IXs and blue shift of PL peaks are evidence of enhanced interlayer coupling.

PL at different distances away from the pillar was extracted and displayed in Fig. 2c. The emission intensity of IXs decreases with the distance away from the pillar. To see the trend of IX emission against distance, IX wavelength and intensity ratio of IX to X⁰ were extracted and plotted against distance in Fig. 2d. As the distance increases, the wavelength of IX emission red shifts from 745.5 to 749.0 nm and saturates when distance reaches 3.5 μm (black dots), suggesting that the pillar can control the IX emission within a small length scale (about 3.5 μm), and the PL wavelength of IXs is maintained outside 3.5 μm. Apart from the blue shifted peak, the intensity ratio of IX to X⁰ decreases from 9.5 to 6.0 with increased distance (magenta dots), serving as another evidence that interlayer coupling is enhanced with the pillar.

To further tune the interlayer coupling strength, we measured PL of WSe₂/(iso-BA)₂PbI₄ heterostructures on various pillars with different *h/d* ratios. PL spectra of WSe₂/(iso-BA)₂PbI₄ on pillars with different *h/d* ratios are shown in Fig. 3a. For each pillar, we measured IX emission on top of the pillar (red curve) and outside pillar (black curve) to make the comparison more reliable. As shown in Fig. 3a, we can see that a pillar with a ratio of 0.2 does not influence the IX spectrum. In contrast, an obviously enhanced PL intensity and blue shifted wavelength are

Figure 3 Modulation of IXs with various pillars. (a) PL of WSe₂/(iso-BA)₂PbI₄ heterostructures on pillars with different height (*h*) to diameter (*d*) ratios (red). The PL of heterostructures without pillar (black) is also shown for comparison. Neutral exciton (X⁰) and interlayer exciton (IX) are labeled; the blue dashed line is located at the IX peak of the sample without pillar for guidelines. (b) Extracted Δλ and IX PL enhancement (*I*_{w/o}/*I*_{w/t}) on different pillars, where the negative value means blue shift. (c, d) Power- and temperature-dependent Δλ of the heterostructure on a pillar with an *h/d* ratio of 0.4, respectively. Notes: Δλ is the IX wavelength difference between heterostructures w/t and w/o pillar.

observed for pillars with ratios of 0.3 and 0.4. The PL of IX red shifts slightly, and an additional peak near 770 nm (donated as IX^2) is observed for the heterostructure on top of the pillar with a ratio of 0.6. To see the trend, we extracted IX peak shift and intensity enhancement ($I_{on}/I_{outside}$) (Fig. 3b). The blue shift and IX intensity enhancement are the largest at the h/d ratio of 0.4, with a value of 4.0 nm and 2.5 times, respectively. For pillars with ratios increased from 0.2 to 0.4, the increased strain perturbation produces a shorter gap, thus enhancing the interlayer coupling strength. We will discuss the abnormal response and origin of the 770 nm peak (bottom panel, Fig. 3a) shown for the pillar with a ratio of 0.6 later. It is worth noting that IX wavelength has some deviations for different samples w/o the pillar in Fig. 3a, originating from the deviation of individual sample fabrication. However, we compare the PL w/t and w/o pillar within the same sample, which is reasonable.

Furthermore, power-dependent measurements were conducted for the pillar with the h/d ratio of 0.4, and the IX wavelength shift is summarized in Fig. 3c. The IX peak shift $\Delta\lambda$ ($\Delta\lambda$ is the difference between IX wavelengths w/t and w/o the pillar) decreased from 10 to 4 nm with increased power and finally saturated. Because the laser power has negligible effects on the bandgap of WSe_2 and $(iso-BA)_2PbI_4$, $\Delta\lambda$ should be a constant. We ascribe the power-dependent $\Delta\lambda$ to the increased interlayer coupling with the pillar. The blue shift is primarily contributed by the stronger dipole moment discussed in Fig. 2 at low powers. However, IX dipole density increases dramatically with increasing power and dominates the repulsion process. Consequently, $\Delta\lambda$ exhibits saturation. Temperature-dependent performance was also measured in Fig. 3d. It can be observed that $\Delta\lambda$ is almost unchanged, which may be because the temperature can only change the bandgap of WSe_2 and $(iso-BA)_2PbI_4$.

PbI_4 instead of IX dipole-dipole interaction conditions.

For the abnormal red shift of IX wavelength and appearance of an additional 770 nm peak, we suspect that IX^2 is the spin-triplet IX, and it is brightened in the presence of a pillar with a high h/d ratio. The process can be explained with the pictures in Fig. 4a, b. It is well known that two split CBs exist with opposite spins (CB1 and CB2) because of the strong spin-orbit coupling in monolayer (1L) WSe_2 in Fig. 4a [38]. Spin-up electrons on higher energy CB1 can form spin-singlet IX, whereas spin-down electrons on CB2 produce spin-triplet IX^2 [39,40]. Normally, singlet and triplet IXs can couple with photons with in-plane and out-of-plane polarization, respectively. Nevertheless, the experimental and calculation results showed that singlet and triplet IXs could occur simultaneously by engineering heterostructure alignment or out-of-plane symmetry (Fig. 4b) [1,39–41]. In our case, the excitation polarization is in-plane with no pillar, and only the higher energy singlet IX (about 735 nm) can be observed. However, the out-of-plane symmetry of WSe_2 is broken in the presence of pillar, leading to the appearance of the lower energy triplet IX^2 . The allowed triplet dipole transition from CB2 will expedite the decay of electrons in CB1, thus reducing the intensity of the singlet. This is consistent with our observation (Fig. 3a) that the intensity of singlet IX (about 735 nm) is reduced, and the peak is red shifted. To further validate our hypothesis, we conducted power-dependent measurements (Fig. 4c). The total PL is represented by a gray curve; it can be fitted with singlet IX (blue) and triplet IX^2 (magenta) peaks. The integrated PL intensities of IX and IX^2 are plotted against power in logarithmic scale, and the slope is 0.64 and 0.57, respectively (Fig. 4d). The sublinear dependence is strong evidence that IX^2 is a kind of interlayer exciton instead of free exciton of 1L WSe_2 due to a longer IX lifetime compared with

Figure 4 Emission of triplet IXs. (a, b) Schematics of singlet IX and triplet IX^2 emission, respectively. The red arrows represent the spin direction. (c) Power-dependent PL of the heterostructure on the pillar with an h/d ratio of 0.6. Grey curve is the measured PL, blue and magenta curves are IX and IX^2 fittings. (d) Power-dependent integrated PL intensity of IX and IX^2 under logarithmic scale. The solid line corresponds to the linear fitting, and they show sublinear slopes. (e) Extracted IX and IX^2 peaks, both of which exhibit a blue shift with power. (f) PL intensity ratio of IX to IX^2 .

direct exciton transition and charge transfer time [42].

The peak wavelength was also extracted and plotted in Fig. 4e. Both IX and IX² show a blue shift with increased power, which is an additional signature of interlayer excitons. The energy difference between IX and IX² is fixed around 70 meV, determined by the CB splitting energy. Our splitting energy is about 20 meV higher than the highest value reported in the literature [43,44]. However, the reported splitting energy varies among literature, no matter in the experiments or theoretical calculations, which is affected by sample quality, measurement temperature and substrates [45]. Fig. 4f shows the integrated PL intensity ratio of IX to IX². It can be seen that the fraction of higher energy singlet IX increases with power, serving as another evidence that it is the band splitting, since the lower band will be populated first, and the higher energy band starts to be filled at high excitation power [1].

We conducted the polarization-dependent measurement to verify that the symmetry is broken for the heterostructure on the pillar, and the results are shown in Fig. 5. Then, we fixed the laser polarization and rotated the collection polarizer during the measurement. An isotropic response was resolved for the sample without pillar (grey dots), while two-folds anisotropy was observed for the pillar-addressed samples. IXs of the heterostructure on a pillar with an h/d ratio of 0.2 have linear dichroism of 1.2 (magenta dots). If the h/d ratio is further increased to 0.6, the linear dichroism of IXs increases to 1.7 correspondingly (orange dots). In addition, the anisotropy of IX² is presented (blue dots), and it shares the same anisotropic trend with IX. The different linear dichroisms of IX and IX² may result from the deviated symmetry breaking in parallel and vertical directions since IX and IX² have in-plane and out-of-plane dipole orientations, respectively. The anisotropy originates probably from the strain effects where the local strain breaks the symmetry of WSe₂. The existence of strain on WSe₂ has already been confirmed by spectra in Fig. S4. Pillars with a higher h/d ratio can provide higher strain and induce larger anisotropy. This is the reason we can observe triplet IXs only at a large h/d ratio (0.6). In addition, inhomogeneous local strain renders the

line shape of IXs different from each sample [36]. It is consistent with our observation in Fig. 2a that the IX line shape varies from each other (red curve). A similar phenomenon has been observed in MoSe₂/WSe₂ heterobilayers whose IX shape and anisotropy were largely affected by the local strain [36]. The out-of-plane symmetry is broken with the pillar, making it feasible to brighten the triplet interlayer exciton, as we discussed previously.

CONCLUSIONS

We investigated the site-control of interlayer coupling in the WSe₂/(iso-BA)₂PbI₄ heterostructure. The heterostructure was placed on pillar arrays with different h/d ratios. PL measurement shows a blue shift wavelength and enhanced PL intensity of IXs with the pillar, which we attribute to the enhanced interlayer coupling between constituent layers. We also interpreted that the reduced gap between two layers is responsible for the enhanced coupling strength. Importantly, IXs become more dominant over intralayer exciton with enhanced coupling. Furthermore, we compared the pillars with different h/d ratios. The results show that the interlayer coupling strength can be engineered by tuning the h/d ratio. The interlayer coupling is enhanced when the h/d ratio increases from 0.2 to 0.6. An additional triplet IX² shows up for the pillar with an h/d ratio of 0.6 due to out-of-plane symmetry breaking. It is also observed that $\Delta\lambda$ declines with power, while a constant $\Delta\lambda$ is observed for different temperatures. In contrast to isotropic PL from the heterostructure on a flat surface, the strain-induced anisotropic response is observed for samples on the pillar because of the symmetry breaking of 1L WSe₂. Our results show the ability to manipulate interlayer coupling strength as desired. All IX PL intensity, resonance wavelength, and IX species can be tuned to meet the requirement for excitonic device applications. The strain-induced anisotropy is also useful in applications of polarization-dependent optoelectronic devices. It is worth noting that IXs can also be formed in perovskite/perovskite heterostructures [46]. The perovskites-based IXs are emerging as another system for practical applications.

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Figure 5 Polarization-dependent IX emission. IX emission without pillar is isotropic (gray). IX emission of the heterostructure on a pillar with an h/d ratio of 0.2 is slightly anisotropic with linear dichroism of 1.2 (magenta). IX² (blue) and IX (orange) of the heterostructure on a pillar with the h/d ratio of 0.6 exhibit linear dichroism of 1.3 and 1.7, respectively.

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Author contributions Wen X and Li D conceived the idea and supervised the project; Wei Q prepared the heterostructures and conducted the optical measurement; Hu J synthesized 2D perovskites; Chen Y, Liu Z, and Lin T helped in TMD transfer and EBL fabrication. All authors analyzed the data and commented on the manuscript.

Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Supporting data are available in the online version of the paper.

Qinglin Wei received her BS degree from Huazhong University of Science and Technology (HUST) in 2019. She is currently pursuing her Master degree at the School of Optical and Electronic Information, HUST. Her research focuses on spectroscopy of transition metal dichalcogenides, two-dimensional perovskites and their heterostructures.

Xinglin Wen received his BS degree in physics from Wuhan University in 2011 and PhD degree from Nanyang Technological University in 2016. He was a post-doctor at Nanyang Technological University from 2016 to 2018. He is currently an associate professor at the School of Optical and Electronic Information, HUST. His research interests include two-dimensional materials, plasmonics and plasmon-exciton coupled system.

Dehui Li received his BS degree in physics from Xi'an Jiaotong University in 2006 and his MS degree from the Institute of Modern Physics, Chinese Academy of Sciences in 2009. He obtained his PhD degree from Nanyang Technological University in 2013. He was a post-doctor at the University of California, Los Angeles from 2013 to 2016. Currently, he is a professor at the School of Optical and Electronic Information, HUST. His research focuses on the synthesis and optoelectronic properties of functional semiconducting nanomaterials and heterostructures.

单层硒化钨/二维钙钛矿异质结层间耦合强度的定点调控

魏青林¹, 温兴林^{1,2*}, 胡俊超¹, 陈瑛瑛¹, 刘泽一¹, 林庭浩¹, 李德慧^{1,2*}

摘要 与过渡金属硫化物中的层间激子相比, 过渡金属硫化物与二维钙钛矿间的层间激子有更高的鲁棒性, 且其光学性质与层间旋转角和是否退火无关. 因此, 过渡金属硫化物与二维钙钛矿间的层间激子正成为一个新的研究激子的体系. 层间激子的有效控制是实现其功能和提高其性能的关键, 然而, 目前缺少对过渡金属硫化物与二维钙钛矿间层间激子有效控制的研究. 本文利用二氧化硅纳米柱实现了单层WSe₂和(iso-BA)₂PbI₄二维钙钛矿间层间激子的定点调控. 与过渡金属硫化物层间激子在应力作用下红移的现象相反, 我们在过渡金属硫化物和二维钙钛矿间的层间激子中观测到10 nm的蓝移以及2.5倍的信号增强. 我们推断该蓝移是由层间间距减小导致的层间耦合强度增大引起的. 该推断也由观测到的层间激子相对自由激子的比例随着耦合强度的增大而增大的现象验证. 同时, 层间耦合还可以通过改变纳米柱的直径和高度进行调控. 特别地, 由于对称性被破坏, 我们在直径/高度比为0.6的纳米柱上观测到三线态层间激子, 且层间激子的各项异性证明其对称性确实已被破坏. 总之, 本研究对调控基于层间激子的光电器件、激子局域态和量子光源具有一定促进作用.