Enhanced Trion Emission in Monolayer MoSe₂ by Constructing a Type-I Van Der Waals Heterostructure

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Trions, quasi-particles consisting of two electrons combined with one hole or of two holes with one electron, have recently been observed in transition metal dichalcogenides (TMDCs) and drawn increasing attention due to potential applications of these materials in light-emitting diodes, valleytronic devices as well as for being a testbed for understanding many-body phenomena. Therefore, it is important to enhance the trion emission and its stability. In this study, a MoSe₂/FePS₃ van der Waals heterostructure (vdWH) with type-I band alignment is constructed, which allows for carriers injection from FePS₃ to MoSe₂. At low temperatures, the neutral exciton (X⁰) emission in this vdWH is almost completely suppressed. The intensity ratio increases from 0.44 in a single MoSe₂ monolayer to 20 in this heterostructure with the trion charging state changing from negative in the monolayer to positive in the heterostructure. The optical pumping with circularly polarized light shows a 14% polarization for the trion emission in MoSe₂/FePS₃. Moreover, forming such type-I vdWH also gives rise to a 20-fold enhancement of the room temperature photoluminescence from monolayer MoSe₂. These results demonstrate a novel approach to convert excitons to trions in monolayer 2D TMDCs via interlayer doping effect using type-I band alignment in vdWH.

1. Introduction

2D transition metal dichalcogenides (TMDCs), MX₂ (M = Mo, W; X = S, Se, Te) with a hexagonal crystal structure are an exciting class of materials.[1–3] Monolayer TMDs feature a direct bandgap, a large binding energy of neutral exciton (X⁰) of up to several hundred meV because of the reduced dielectric screening, conventional semiconductor heterostructures like GaAs-AlGaAs, maximum (VBM) are located in the wider-bandgap (WBG) and narrower-bandgap (NBG) materials, respectively. Electrons and holes can be spatially separated, which results in the formation of the interlayer excitons and consequently in quenching of the photoluminescence (PL) in their component layers.[13]

However, the type-I band alignment, which is common in valley-selective optical coupling due to the lack of inversion symmetry, and a strong spin–orbit coupling.[4] Excitons in TMDCs can often trap an electron (X⁻) or a hole (X⁺) to form so-called trions.[4–7] With these exotic features and tunable bandgaps from visible to near-infrared regions, TMDCs are considered as a highly desired material class for the next-generation optoelectronic and valley-based electronic applications.[8]

Stacking 2D layered materials with pre-selected properties will give rise to the formation of van der Waals heterostructures (vdWHs) with atomically sharp and near-defect-free interfaces, which may exhibit novel physics and possess versatile properties.[9–11] So far, the most extensively studied TMDC vdWHs include MoS₂–MoSe₂,[12] MoS₂–WS₂,[13,14] MoS₂–WSe₂,[15,16] MoSe₂–WSe₂,[17] MoSe₂–WS₂,[18] and WS₂–WSe₂.[19] Their band alignment is found to be staggered type-II, where the conduction band minimum (CBM) and valence band maximum (VBM) are located in the wider-bandgap (WBG) and narrow-bandgap (NBG) materials, respectively. Electrons and holes can be spatially separated, which results in the formation of the interlayer excitons and consequently in quenching of the photoluminescence (PL) in their component layers.[13]

The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/adfm.202104960.
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is very rarely reported in vDWs. The CBM and VBM of type-I band alignment are both located in the NBG layer. Therefore, in the heterojunction with type-I band alignment, carriers will flow only from the WBG to the NBG material. Consequently, in the NBG layer, the carrier concentration increases and its PL intensity is enhanced. Nevertheless, besides the use of hexagonal boron nitride (h-BN) for passivation, up to now only a few implementations of the type-I band alignment in 2D materials, like PbI₂/WS₂ and MoS₂/ReS₂, have been reported. Those papers focus on the charge transfer process at the interface, which leads to a PL enhancement of the NBG layer. However, trion and exciton emission behavior in type-I vDW with TMCDs has not yet been investigated. The conversion from excitons to trions controlled by charge density can influence the exciton lifetime and leads to a large valley polarization via valley-selective optical pumping. Since in straddling bands carriers flow only in one direction, by proper selection of WBG materials (n-type or p-type) the charging state of trion can be controlled. Moreover, positively charged trions, X⁺, have a much longer dephasing time than negatively charged trions, X⁻ and thus a smaller linewidth, which makes them attractive for quantum technology.

In this work, we demonstrate the manipulation of the trion charging state and population in monolayer MoSe₂ by constructing a vdWH MoSe₂/FePS₃ with type-I band alignment. FePS₃ with an indirect bandgap is chosen as the WBG semiconductor. The type-I band alignment is supported by density-functional theory (DFT) calculation, such that MoSe₂ with the narrower bandgap acts as the carrier extraction layer and p-type FePS₃ is a source of holes. We have achieved an enhancement of the trion emission with the Iₜ/Iₓ₀ ratio increasing from 0.44 to 20, and a 20-fold enhancement of PL emission from the MoSe₂ monolayer due to carrier injection from FePS₃. Moreover, both the exciton emission in a single MoSe₂ monolayer and the trion emission in MoSe₂/FePS₃ depend on the chirality of the excitation light, indicating the valley polarization selectivity. Our study provides essential insight into the underlying carrier transport mechanism and points to potential device application based on type-I band alignment vDW.

2. Results and Discussion

2.1. Raman Results

Figure 1a shows the schematic configuration of MoSe₂/FePS₃ vdWH covered with a few layers of h-BN. Except protecting samples from air, the h-BN encapsulation also reduces the linewidth of the trion and exciton emission from the MoSe₂ monolayer. The MoSe₂ monolayer was fully laying on the top of FePS₃ and the heterostructure area is around 100 µm² which is much larger than the laser spot size (about 7 µm²). Therefore, all the optical signals (including micro-Raman and micro-PL) are coming from the heterojunction. For the reference, individual MoSe₂ monolayers and FePS₃ flakes covered with h-BN were prepared as well. Figure 1b shows the optical microscope image of MoSe₂/FePS₃ vdWH, where regions with magenta and orange outlines represent FePS₃ and MoSe₂, respectively.

Figure 1c,d shows the Raman spectra of monolayer MoSe₂ combined with 22 nm (corresponding to 31 layers) FePS₃.
defined as 1ML MoSe$_2$/3ML FePS$_3$, multilayer FePS$_3$, and individual MoSe$_2$ flakes with different thicknesses (1ML, 2ML, and multilayer). For MoSe$_2$, the A$_{1g}$ mode is an out-of-plane vibration modes in which Se atoms in all layers oscillate in phase with reference to the corresponding central Mo atom.[27] Figure 1d shows the micro-Raman spectra taken from the MoSe$_2$ layer with different thicknesses. The peak positions of the A$_{1g}$ mode at 240.6 cm$^{-1}$ for 1ML MoSe$_2$, 241.7 cm$^{-1}$ for 2ML, and 242.3 cm$^{-1}$ for multilayer MoSe$_2$ are in good agreement with previous reports,[27] confirming the thickness of MoSe$_2$ in our study. Besides the A$_{1g}$ phonon mode, the MoSe$_2$ flake has other Raman active phonon modes like E$_{2g}^1$ and B$_{2g}^1$ visible at about 286.9 and 355.7 cm$^{-1}$, as well as E$_{1g}$ at 169.7 cm$^{-1}$ for 1ML MoSe$_2$ (see Figure 1c). Their peak positions shift to lower wave-numbers as the number of layers increases and the highest peak intensities are observed for the MoSe$_2$ bilayer. The other peaks labeled in Figure 1c with a, b, and c showing much broader features especially pronounced in monolayer and bilayer MoSe$_2$ are assigned to second-order Raman processes.[28] Interestingly, the A$_{1g}$ phonon mode from 1ML MoSe$_2$ in vdWH has the same peak position as that from individual 1ML MoSe$_2$, but the peak intensity is significantly reduced, probably due to the softening of the phonons by phonon–carrier interaction.

The Raman spectrum from FePS$_3$ is also presented in Figure 1c. FePS$_3$ shows six distinguishable peaks labeled with P$_1$–P$_6$. The observed phonon modes are in agreement with previously reported ones.[29] The P$_1$–P$_6$ phonon modes are assigned to the ($P_2S_6$)$_{4^{-}}$ bipyramid structures and P$_1$–P$_2$ are related to the Fe atoms and are sensitive to the magnetic coupling between Fe atoms.[30] According to the temperature-dependent micro-Raman spectra, the evolution of the P$_1$ and P$_2$ phonon modes in FePS$_3$ is very similar both in the individual flake and in the vdWH. We found that the Néel temperature of FePS$_3$ is at around 120 K (see Figure S1, Supporting Information), which is in good agreement with the data reported in the literature.[29]

2.2. Band Alignment from DFT Calculations

We used DFT calculations to investigate the band alignment for monolayer MoSe$_2$/FePS$_3$. For the calculations, we have considered a monolayer heterojunction 1ML MoSe$_2$/1ML FePS$_3$ and a heterojunction prepared of 3ML MoSe$_2$ on monolayer FePS$_3$, as shown in Figure 2a. After structural optimization, an average interlayer distance of $d = 3.63$ Å was found between FePS$_3$ and MoSe$_2$. The binding energy between FePS$_3$ and MoSe$_2$ was calculated to be 16 meV Å$^{-2}$ which is close to the values in other 2D vdWH interfaces.[31] Our electronic structure calculations indicate that the MoSe$_2$ monolayer has a direct band gap of 1.33 eV at the K point (Figure S2, Supporting Information). The FePS$_3$ monolayer shows a quasi-direct bandgap of 2.27 eV where VBM and CBM are both located at the Σ point (midpoint of Γ and K points). Although DFT band gap cannot directly be compared with the optical gap measured in the experiment, our
results are in line with the experimental band gap (2.2 eV)\cite{32} and the theoretical band gap (2.5 eV)\cite{33} for FePS$_3$ monolayer. The slight difference of band gap values for FePS$_3$ monolayer between our calculation and the previous theoretical report\cite{33} is due to different calculation methods. Based on the electronic structure calculations, the CBM of the MoSe$_2$ is lower than that of the FePS$_3$ while the VBM is higher than that of the FePS$_3$, forming type-I band alignment, as schematically illustrated in Figure 2b,c. Both heterostructures with 1ML-MoSe$_2$ and 3ML-MoSe$_2$ show direct band gaps of 1.51 and 1.07 eV, situated at the $\Gamma$ point in Figure 2b,c. The projected densities of states suggest that the VBM mainly consists of Mo d and Se p orbitals, whereas the CBM is mainly composed of the Mo d orbitals and slight contributions from Fe d states. To further analyze the effect of coupling between FePS$_3$ and MoSe$_2$, the average potential profile across the heterostructure and the charge difference between the combined system and isolated parts are shown in Figure S3 in the Supporting Information. Owing to the higher potential energy of MoSe$_2$, the charge is redistributed in the system with predominant accumulation at the side of MoSe$_2$ layer facing FePS$_3$.

2.3. Photoluminescence Results

Figure 3a shows the room temperature PL (RTPL) spectra of 1ML MoSe$_2$/31ML FePS$_3$ and individual MoSe$_2$ monolayer. The RTPL spectra obtained from 2ML individual MoSe$_2$ and its heterostructure are presented in Figure 3b. Note that the RTPL measurement was performed in air under ambient conditions. Insets show optical images of investigated samples. 1ML and 2ML MoSe$_2$ are specially chosen here since MoSe$_2$ possesses thickness-dependent bandgap features\cite{2,34}, i.e., a direct bandgap for monolayer MoSe$_2$ and an indirect bandgap for thicker MoSe$_2$. Similar to Raman spectra, the PL measurements can be used to determine the thickness of investigated MoSe$_2$ flakes. All the samples for PL measurement were covered with h-BN in order to prevent oxygen/moisture contaminations and obtain a high quality of optical emission spectra\cite{35}. Since FePS$_3$ is an indirect band gap semiconductor\cite{33} and its photogenerated carriers can quickly relax to MoSe$_2$ due to their type-I band alignment, all the PL peaks presented here originate only from MoSe$_2$ flakes. The single MoSe$_2$ monolayer flake shows the RT excitonic PL emission at 1.573 eV while the PL

![Figure 3](https://i.imgur.com/xy.png)

Figure 3. a) RTPL spectra of 1ML MoSe$_2$/31ML FePS$_3$ and 1ML MoSe$_2$-single layer. b) RTPL spectra of 2ML MoSe$_2$/24ML FePS$_3$ and 2ML MoSe$_2$-single layer. Insets are optical images of samples. c) Band alignment of MoSe$_2$/FePS$_3$ before contact of constituent layers, after contact of constituent layers, and under laser excitation. The shadowed areas visualize the band alignment for 1ML MoSe$_2$/multilayer FePS$_3$. d) PL spectra of 1ML MoSe$_2$/31ML FePS$_3$ and single 1ML MoSe$_2$ at 15 K. e) Normalized TDPL spectra for 1ML MoSe$_2$/31ML FePS$_3$ (blue-shaded areas) and 1ML MoSe$_2$ (black-shaded areas).
emission from 1ML MoSe2/FePS3 is slightly blue-shifted to 1.579 eV. More importantly, the heterostructure shows 20-fold enhancement of PL emission. Since the exciton lifetime in indirect semiconductors is generally much longer than in direct ones, the photogenerated carriers in FePS3 can quickly diffuse into MoSe2 and radiatively recombine there, which accounts for this giant PL enhancement in 1ML MoSe2/31ML FePS3. The PL emission from bilayer MoSe2 on FePS3 observed at 1.543 eV is enhanced by a factor of 1.3 in comparison to the individual flake. Zhang et al. have shown strong enhancement of the exciton emission from the WS2 monolayer covered with WO3 flake. Zhang et al. have shown strong enhancement of the exciton emission from the WS2 monolayer covered with WO3 due to the reduction of nonradiative recombination and the charge transfer within the heterostructure.[36]

As concluded from DFT calculation, 1ML MoSe2/FePS3 shows a straddling type-I band alignment between MoSe2 (NBG) and FePS3 (WBG). In addition, the conductivity type of single MoSe2 and FePS3 flakes is confirmed by electrical measurements presented in Figure S4 in the Supporting Information. MoSe2 and FePS3 possess n-type and p-type conductivity, respectively, which is consistent with other reports.[37,38] After forming a heterojunction with type-I band alignment, holes will flow from FePS3 to MoSe2 to achieve an equilibrium state (see the middle panel in Figure 3c). In our case, the thickness of the MoSe2 is limited to one monolayer (about 0.7 nm), which can be completely converted to p-type when combining with 31 ML FePS3. The shallowed areas in Figure 3c indicate the band alignment for 1ML MoSe2/multilayer FePS3. Under excitation, both electrons and holes generated in FePS3 can diffuse to MoSe2, with a result of its PL enhancement due to type-I band alignment (see the right panel in Figure 3c).

The smaller enhancement in 2ML MoSe2/24ML FePS3 is due to the fact that 2ML MoSe2 is an indirect bandgap semiconductor with much lower radiative recombination rate.[34] Since the MoSe2 layer is placed fully on the FePS3 layer, we can exclude the possibility that surface charges in the SiO2 substrate can donate carriers into MoSe2, as it was previously reported.[35,39] Therefore, in our case only FePS3 can be the extra source of photogenerated carriers in MoSe2. A PL emission enhancement for the narrower bandgap material in type-I van der Waals heterojunctions has been reported before, e.g., for MoTe2–WS2,[21] Pbl2–WS2,[40] WSe2–black phosphorus,[41] MoS2–ReS2,[20] GaSe–GaTe,[22] etc. This distinct feature is substantially different from that of the widespread type-II band alignment for TMDcs heterostructures, like MoS2–WS2,[15] MoS2–MoSe2,[12] MoS2–WS1, and WSe2–MoS2,[13,42] where the PL emissions of two constituent layers are both quenched along with the appearance of an additional interlayer exciton peak with low energy.[17,43]

To shine more light on the interaction and charge transfer between FePS3 and MoSe2, temperature-dependent PL (TDPL) measurements were performed. In this measurement, we focus on the sample 1ML MoSe2/31ML FePS3. As the reference sample, an individual MoSe2 monolayer was also investigated. Figure 3d shows the PL spectra of 1ML MoSe2/31ML FePS3 and individual MoSe2 monolayer at 15 K. The peaks at 1.66 and 1.63 eV in both spectra correspond to neutral exciton (X) and charged trion (T) emission from the MoSe2 monolayer, respectively. The appearance of the trion emission in the individual MoSe2 monolayer is due to the unintentional n-type doping as a consequence of impurities attached to reactive chalcogenide vacancies or defects in the SiO2 substrate.[44–46] In Figure 3d, we observe a giant enhancement of the trion emission and a dramatic suppression of the exciton emission (the PL intensity ratio $\frac{I_{\text{Trion}}}{I_{\text{X}}}$ = 20) for MoSe2 in the heterojunction in comparison with $\frac{I_{\text{Trion}}}{I_{\text{X}}}$ = 0.44 for individual 1ML MoSe2. The strong enhancement of the trion emission implies a strong doping of MoSe2 in vdWH. Again, this agrees well with the definition of type-I band alignment where carriers can flow only in one direction from WBG to NBG. In heavily doped semiconductors, excitons can trap free electrons or holes to form trions. This phenomenon also can be explained by a model based on the law of mass action:[47,48]

$$\frac{N_{X}N_{e}}{N_{T}} = \left(\frac{4m_{X}m_{e}}{\pi\hbar^{2}m_{T}}\right) k_{B}T \exp\left(-\frac{E_{B}}{k_{B}T}\right) \tag{1}$$

where $N_{X}$, $N_{T}$, and $N_{e}$ are trion, exciton, and charge carrier concentrations, $\hbar$ is the reduced Planck’s constant, $k_{B}$ is the Boltzmann constant, $T$ is the temperature, $E_{B}$ is the trion binding energy, $m_{X}$ is effective mass of electrons, $m_{X}$ and $m_{T}$ are exciton and trion effective masses, respectively. According to this model, there is a positive correlation between $\frac{N_{T}}{N_{X}}$ and $n_{e}$, i.e., $\frac{N_{T}}{N_{X}}$ increases with increasing carrier concentration $n_{e}$. Since the concentrations of trions and excitons are proportional to their PL intensities $\frac{N_{T}}{N_{X}} \propto \frac{I_{T}}{I_{X}}$, it means that doping (higher $n_{e}$ or $n_{h}$) can lead to a higher PL intensity ratio $\frac{I_{T}}{I_{X}}$. Besides, the increased trion binding energy, which can be modified by the dielectric environment, could also lead to a higher $\frac{I_{T}}{I_{X}}$. Specifically, a lower dielectric constant of the environment can reduce the total screening effect on 1ML TMDCs, which can result in a higher trion binding energy and further can contribute to a higher $\frac{I_{T}}{I_{X}}$.[35] Due to $\epsilon_{\text{FePS3}} < \epsilon_{\text{SiO2}}$, it means that FePS3 in vdWHs may provide an environment that is more energetically favorable for trion formation in MoSe2/FePS3 than in individual MoSe2 being directly contacted with a SiO2 substrate.

Figure 3e shows the normalized TDPL spectra for vdWHs (blue-shaded areas) and individual 1ML MoSe2 (black-shaded areas). The red shift of exciton and trion peaks for both samples is observed with increasing temperature, which is due to temperature-induced bandgap shrinkage. Below 50 K, the trion emission gives a pronounced peak and dominates the spectra of vdWH, while in individual 1ML MoSe2 the exciton emission dominates the PL spectra because of the lower carrier concentration and thus lower trion concentration. At 50 K, the trion and exciton emissions from the heterojunction have comparable intensity, but with increasing temperature the trion emission vanishes. This is due to thermal fluctuations resulting in the conversion from trions to excitons. In individual 1ML MoSe2, the trion emission is already negligible as compared with the exciton peak at 50 K as a consequence of lower carrier concentration. In the temperature range of 90–300 K, only the exciton emission peak is observed for both
Figure 4. Low-temperature PL emission after excitation with left- or right-handed circularly polarized light obtained a) from the heterojunction and b) from the individual MoSe2 monolayer. The inset shows the chiral optical selection rules for interband optical transitions in nonequivalent valleys K+ and K− in monolayer MoSe2.

vdWHs and individual MoSe2. Note that we assume FePS3 has no effect on the exciton peak position of MoSe2 in vdWHs. In addition, the thermal stability of the trion also depends on its charge state. Positively charged trions, X+2, have localized holes that are less prone to scattering, leading to the reduced line width.30

In order to clarify the charge sign of trions in both systems and to explore their potential impacts on valleytronics, we have employed PL excitation with circularly polarized light (see Figure 4). Using left-(σ−) or right-handed (σ+) excitation in both systems, we have seen differences in the PL emission. Here, the degree of circular polarization is evaluated as $P_c = [I(\sigma +) − I(\sigma −)]/[I(\sigma +) + I(\sigma −)]$, where $I(\sigma +)$ and $I(\sigma −)$ correspond to the PL intensities under left-(σ−) or right-handed (σ+) excitation. In the individual monolayer MoSe2, the exciton emission is enhanced by 13% for σ+ excitation while the trion emission is almost not affected with a small broadening. The exciton emission polarization is resulted from the broken inversion symmetry and the strong SOC in monolayer TMDCs,49 which gives rise to two inequivalent K+ and K− valleys with different population. Due to the spin-valley locking effect, the right-handed (σ+) circularly polarized photon initializes a carrier in the K+ valley and the left-handed (σ−) initializes a carrier in the K− valley, i.e., valley-dependent optical selection rules.50,51 Zeng et al. reported that 30% valley polarization can be achieved in pristine monolayer MoS2 with circularly polarized light pump.34 For the PbI2/WS2 heterostructure with type-I band alignment, Zhang and co-workers have shown near unity spin polarization in PbI2 due to the modification of the carrier lifetime.52 In the heterostructure MoSe2/FePS3, the exciton emission is the same for both excitations, while the trion emission exhibits a polarization of about 14%. It suggests that no carrier spin polarization is introduced from Ising-type antiferromagnetic FePS3 to MoSe2. Moreover, the trion emission from the heterostructure is narrower than from the individual MoSe2 monolayer and the peak position is blue shifted for the heterostructure by about 2 meV. Since holes are more localized than electrons in MoSe2, X+ are immobile and less prone to scattering, leading to a narrow linewidth of X+ than X−.30 A similar energy difference for the X+ and X− emission and narrowing of the PL emission for X+ in MoSe2 were observed by Shepard et al.30 Therefore, we can conclude that in our case negatively charged trions X− form at low temperature in the individual MoSe2 monolayer, while in the heterostructures the trions are positively charged due to hole injection from p-type FePS3 into the MoSe2 monolayer.

3. Conclusion

In conclusion, we have performed a systematic study of optical properties for a novel semiconductor-antiferromagnetic MoSe2/FePS3 vdWH. DFT calculations show the type-I band alignment in ultrathin MoSe2/FePS3, which is verified by the PL results. The room temperature PL shows 20-fold enhancement of the emission from 1ML MoSe2 in MoSe2/FePS3 vdWH, which is due to carrier injection from WBG FePS3 to NBG MoSe2. At low temperatures, the intensity ratio between trion and exciton emission increases from 0.44 to about 20. Moreover, the heterojunction exhibits trion emission polarization, which makes it attractive for valleytronics. Our results present a novel approach to modify the population of trions and excitons as well as the trion charge state by forming a heterostructure with type-I band alignment. This approach is transferable to many other 2D materials and is opening a broad playground for fundamental physics and for valleytronic applications.

4. Experimental Section

Sample Preparation: The 2D material flakes were mechanically exfoliated through blue tape and then transferred onto SiO2/Si substrates (90 nm thick SiO2 on heavily doped Si) by using a polymer stamp transfer technique within a nitrogen atmosphere in a glove box environment. This technique provided good crystalline quality, intact atomic interfaces, and less contamination of the flake in comparison with other transfer techniques.53 Also, prepatterned electrodes with Ti/Au (5/45 nm) on SiO2/Si substrates were fabricated by standard photolithography and electron-beam evaporation processes. To fabricate the MoSe2/FePS3 vdWH, FePS3 was first transferred onto the SiO2/Si substrate, and then the monolayer MoSe2 was transferred onto the top of the FePS3 flake. For reference, the single MoSe2 monolayer flakes were prepared in the same way. Finally, both the vdWH and single MoSe2 flakes were covered with h-BN in order to reduce flake contamination and degradation resulting from contact with air.

Characterization: For the micro-PL and micro-Raman measurements, a cw frequency-doubled Nd:YAG laser was used for excitation at a wavelength of $\lambda = 532$ nm. The laser power was controlled by introducing neutral density filters. The maximum laser power used here was 3.2 mW to avoid heating of the flake. The spot diameter of the laser on the sample was ~3 µm, i.e., smaller than the size of the target flakes. Both PL and micro-Raman investigations were performed in the temperature range from 4 to 300 K using a liquid-He cooled chamber and temperature controller. A liquid nitrogen-cooled Si-CCD camera was used to detect the PL emission, which was dispersed in a spectrometer. To check the PL polarization of trion and exciton in MoSe2, the excitation with circular left-(right-) handed polarized light was performed. The measurement was done at 4 K. The thickness of FePS3 was measured with atomic force microscopy (AFM) with a Bruker Multimode 8 system.
FePS$_3$, a unit cell of tetragonal shape with 20 atoms, was considered as a baseline for the calculations. In order to adopt zigzag antiferromagnetic ordering in the basal plane, an isolated slab approach was used to implement this ordering. The Brillouin zone of the system was sampled using an 8$\times$8$\times$1 k-mesh for primitive cells and a 5$\times$5$\times$1 k-mesh for supercells. London dispersion interactions were included in the total energy as proposed by Grimme in the DFT-D2 method. The geometry optimization was carried out until the atomic forces were less than 0.01 eV Å$^{-1}$. A vacuum layer of 20 Å perpendicular to the basal plane was introduced to implement the isolated slab condition. In order to adopt zigzag antiferromagnetic ordering in FePS$_3$, a unit cell of tetragonal shape with 20 atoms was considered, as shown in Figure 2a. The heterostructures were constructed by using an interface consisting of 1$\times$1 unit cell of FePS$_3$ and 1$\times$3 unit cells of monolayer MoSe$_2$, corresponding to a lattice mismatch of only 0.6%. The supercells contained 38 atoms in two layers and 74 atoms in four layers (MoSe$_2$ triple layers + FePS$_3$ monolayer) configurations.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

J.M.D. thanks China Scholarship Council (File no. 2017068900037). L.H. thanks the National Natural Science Foundation of China (project number 61804098) and the Zhejiang Provincial Natural Science Foundation of China (project number LZ21E020002). Y.J.Z. thanks the Shenzhen Science and Technology Project under Grant number JCYJ20180507182246321. A.V.K. also thanks the DFG for support within the projects KR 4866/2-1 (project number 339 406129719). The computational support from the Technical University of Dresden computing cluster (TAURUS) and from High Performance Computing Center (HLRS) in Stuttgart, Germany is gratefully acknowledged. The authors thank Scheumann for the metal deposition of the substrates. The nanofabrication facilities (NanoFaRo) at the Ion Beam Center at the HZDR are also gratefully acknowledged. Open access funding enabled and organized by Projekt DEAL.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Research data are not shared.

Keywords

photoluminescence enhancement, polarization, trion/exciton intensity ratio, type I, van der Waals heterostructure

Received: May 25, 2021
Revised: June 25, 2021
Published online:

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