

Defect Healing and Charge Transfer-Mediated Valley Polarization in MoS₂/MoSe₂/MoS₂ Trilayer van der Waals Heterostructures

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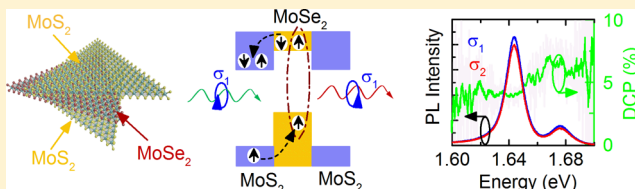
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Supporting Information

ABSTRACT: Monolayer transition metal dichalcogenides (TMDCs) grown by chemical vapor deposition (CVD) are plagued by a significantly lower optical quality compared to exfoliated TMDCs. In this work, we show that the optical quality of CVD-grown MoSe₂ is completely recovered if the material is sandwiched in MoS₂/MoSe₂/MoS₂ trilayer van der Waals heterostructures. We show by means of density functional theory that this remarkable and unexpected result is due to defect healing: S atoms of the more reactive MoS₂ layers are donated to heal Se vacancy defects in the middle MoSe₂ layer. In addition, the trilayer structure exhibits a considerable charge-transfer mediated valley polarization of MoSe₂ without the need for resonant excitation. Our fabrication approach, relying solely on simple flake transfer technique, paves the way for the scalable production of large-area TMDC materials with excellent optical quality.

KEYWORDS: Chemical vapor deposition, transition metal dichalcogenides, van der Waals heterostructures, defect healing, charge transfer mediated valley polarization



Monolayer transition metal dichalcogenides (TMDC) have a direct bandgap situated in the visible range, which makes them ideal building blocks for novel electronic and optoelectronic devices.^{1–10} The bandgap of monolayer TMDCs occurs at the inequivalent (but degenerate) K and K' points of the hexagonal Brillouin zone. The broken inversion symmetry of a TMDC monolayer combined with the time reversal symmetry imposes opposite magnetic moments at the K and K' valleys. This in turn determines the characteristic circular dichroism exhibited by these materials, wherein each valley can be addressed separately with circularly polarized light of a given helicity.^{11–13} Additionally, optical spectra are influenced by the strong spin–orbit coupling, which lifts the degeneracy of band states at the valence band edges, resulting in well-resolved A and B resonances, as observed in reflectivity or absorption spectra.^{2,14–16} The interplay of spin–orbit coupling with broken inversion symmetry and time reversal symmetry locks the valley and spin degrees of freedom, making TMDCs attractive candidates for valleytronics.¹⁷ The spin–valley index locking along with the large distance in the momentum space between K and K' valleys preserves the valley polarization observed in the degree of circular polarization (DCP) in helicity resolved photoluminescence emission.^{18–22}

Applications require a scalable fabrication platform providing high-quality large-area monolayer TMDC. Unfortunately, the

most promising approach today, namely chemical vapor deposition (CVD) growth^{23–27} struggles to compete with exfoliated TMDCs in terms of sample quality. Low-temperature photoluminescence (PL) spectroscopy of CVD-grown MoS₂ and MoSe₂ reveals broad emission from defect bound excitons, which is significantly more intense than the free exciton peak^{28–30} and is related to chalcogen vacancies induced during the CVD growth.^{29,30}

Here, we demonstrate a novel approach to neutralize the intrinsic defects of CVD-grown TMDCs, using flake transfer tools routinely employed in the fabrication of van der Waals heterostructures.^{31–33} We investigate the optical properties of trilayer stacks composed of external CVD-grown MoS₂ flakes²⁴ as capping layers and an internal CVD-grown MoSe₂ flake that has a smaller bandgap.^{34,35} Remarkably, this fabrication approach strongly suppresses the localized exciton emission in MoSe₂, yielding a low-temperature PL comparable to that observed in mechanically exfoliated samples. This striking result can be understood from density functional theory (DFT), which suggests that the more reactive MoS₂ donates chalcogen atoms to heal vacancy defects in MoSe₂. Incorporating MoS₂

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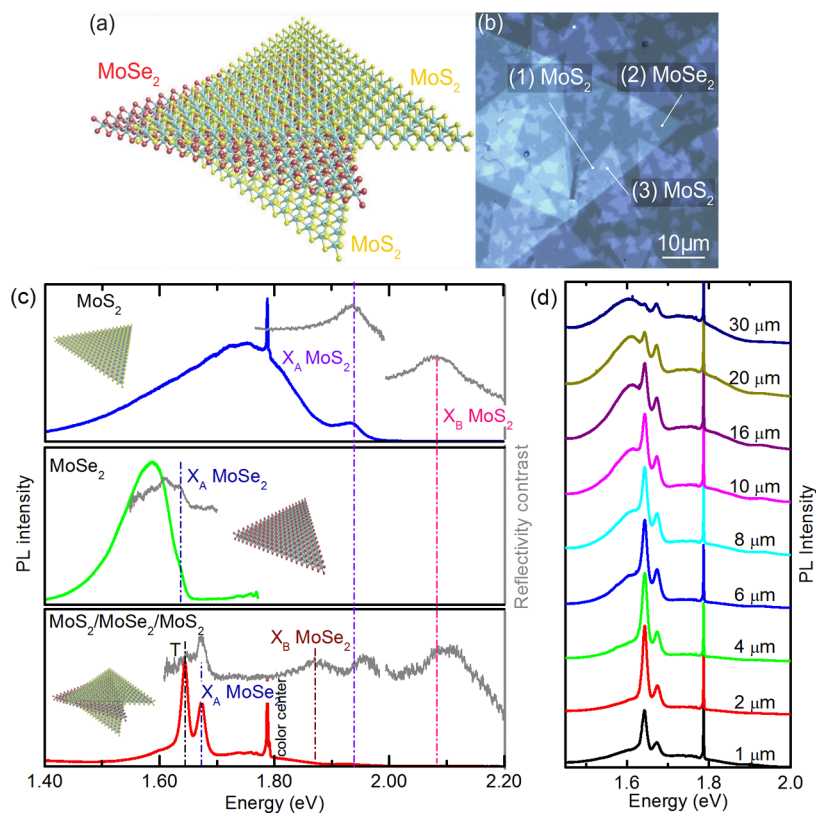


Figure 1. (a) Ball and stick model of MoSe₂ layer sandwiched between two MoS₂ flakes. (b) Optical micrograph of a representative transfer area. (c) Low-temperature μ PL spectra (colored curves) and reflectivity contrast (gray curves) of as-grown MoS₂, MoSe₂ monolayers and MoS₂/MoSe₂/MoS₂ trilayer stack. (d) PL spectra of trilayer stack measured using different spot sizes. The spectra are vertically offset for clarity. X_A indicates A exciton and T denotes the trion (charged A exciton).

into the trilayer heterostructure furthermore allows us to demonstrate a new way to introduce valley polarization in MoSe₂. Because of the type II band alignment in TMDC heterojunctions,³⁵ a significant charge transfer is observed in these systems.^{32,36,37} Our results show that spin of the hole is conserved upon charge transfer from MoS₂ to MoSe₂ after excitation in resonance with MoS₂ A exciton. This leads to nonzero steady-state valley polarization in MoSe₂, which has never been observed before under nonresonant excitation.^{38–40}

Defect Healing. The sample with MoS₂/MoSe₂/MoS₂ trilayer stacks and micrograph of a representative transfer area are schematically shown in Figure 1a,b. Low-temperature microPL (μ PL) spectroscopy has been used to characterize as-grown CVD samples on the sapphire substrate (prior to any transfer process) and the trilayer MoS₂/MoSe₂/MoS₂ stack. All spectra shown in Figure 1c,d were measured under nominally identical conditions with an excitation power of 100 μ W (see Figure S1 in Supporting Information (SI) for power dependent μ PL spectra). The as-grown MoS₂ and MoSe₂ monolayers both show a broad PL feature (full width at half-maximum, fwhm, of 258 and 106 meV, respectively) related to emission from excitons bound to defect or charge impurity states.^{28–30,41} A-exciton emission (labeled X_A in Figure 1c) is seen only as a weak peak or shoulder at higher energies. These assignments are confirmed by reflectivity contrast measured on the same spot. Reflectivity contrast is defined as $\Delta R/R_s = (R - R_s)/R_s$, where R is the reflectivity spectrum measured on the sample and R_s denotes the reflectivity spectrum measured on the substrate. In the case of transparent substrates such as sapphire, reflectivity contrast is proportional to the absorption of the

sample.⁴² The reflectivity contrast spectrum of as-grown MoS₂ (see upper panel of Figure 1c) consists of a main peak at 1.937 eV and a higher energy, weaker feature at 2.085 eV, related to the spin-orbit split B exciton. The energy difference of 148 meV corresponds very well to theoretically predicted spin-orbit splitting of the valence band⁴³ and is very similar to the splitting determined with transmission measurements on similar samples.³⁴ The reflectivity contrast spectrum of as-grown MoSe₂, shown in the central panel of Figure 1c has a peak at 1.637 eV, which corresponds well to the high energy shoulder of the μ PL spectrum and hence is assigned to A exciton. As in our previous study,³⁴ we are unable to resolve the B exciton in reflectivity contrast measurements on as grown MoSe₂.

The optical properties of the trilayer stack are dramatically improved (bottom panel in Figure 1c). The most striking difference is the nearly total suppression of (i) emission from defect bound excitons, and (ii) significant quenching of both MoS₂ PL, barely seen as a weak peak at 1.929 eV, and of MoSe₂ PL (overall integrated intensity decreases by two and one orders of magnitude, respectively, see Figure S2 in SI). The measured PL spectrum is dominated by narrow free neutral (X_A) and charged exciton (T) emission in MoSe₂ (low power fwhm of 11 and 10 meV, respectively, see Figure S1 in SI), approaching the quality of exfoliated WSe₂ embedded in boron nitride (fwhm \sim 10 meV).⁴⁴ The free exciton emission overlaps with a weak broad background emission from the sapphire substrate (the narrow peak just below 1.8 eV corresponds to the emission from a color center in sapphire). The highest energy peak in the reflectivity contrast spectrum of the trilayer

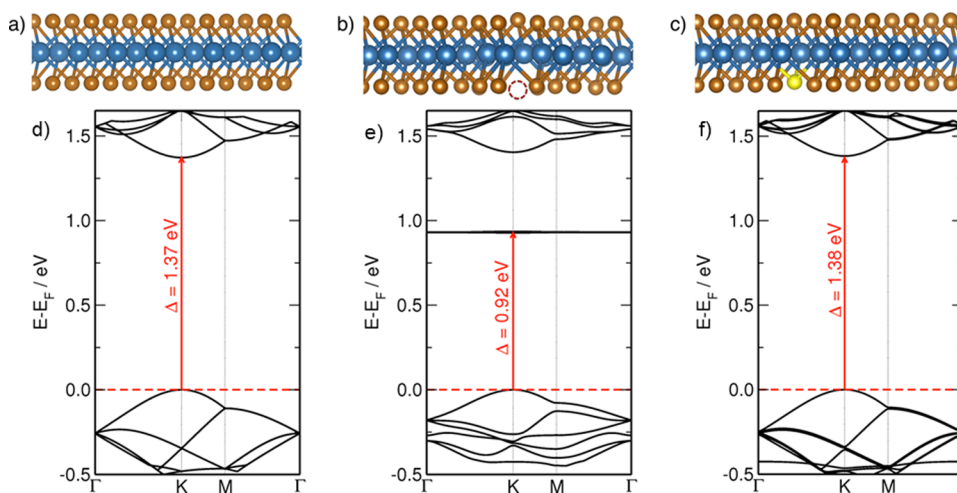


Figure 2. (a–c) Atomic representation of a perfect MoSe₂ monolayer, the MoSe₂ monolayer with 1 Se vacancy, and the MoSe₂ monolayer with 1 S substitution, respectively. (d–f) Corresponding calculated band structures. Fermi level (horizontal dashed lines) is shifted to the top of the valence band. Fundamental bandgaps are indicated.

(see Figure 1c, bottom panel), blue-shifted with respect to the A exciton peak of MoS₂ by 145 meV is attributed to the B exciton of MoSe₂. The peak related to MoS₂ A exciton in the reflectivity contrast spectrum is blue-shifted by 27 meV with respect to the corresponding PL peak. This Stokes shift has been attributed to the presence of a high doping level in MoS₂,¹⁶ also present in our layers.⁴⁵ On the low energy side of the μ PL spectrum, the two distinct peaks at 1.674 and 1.645 eV are assigned to the A exciton and to the trion of MoSe₂, respectively. The trion binding energy of 29 meV is very similar to that reported in other studies on MoSe₂.^{29,46}

The high optical quality of the MoSe₂ embedded in the heterostructure enabled us to resolve an additional peak at 1.87 eV in the reflectivity contrast spectrum, assigned to the B exciton of MoSe₂.^{43,47} The vastly improved optical properties suggest a defect healing process in which the contact with MoS₂ is enough to drastically reduce the number of defects in MoSe₂. The quenching of the intralayer emission^{32,37} in the trilayer is manifestation of a fast charge transfer mechanism^{36,48} related to the type II band alignment in MoS₂/MoSe₂ heterostructures.³⁵ The weak luminescence of MoS₂ is consistent with a background n-doping⁴⁵ of the as grown layers and with an additional charge transfer after the formation of the heterostructure. We assign the brighter emission from MoSe₂ (in the trilayer) to the hole transfer to an intrinsically n-doped material and to the defect healing effect, combined with luminescence resulting from higher energy states, similar to hot luminescence of direct exciton in multilayer MoSe₂.^{49,50} The long-range optical uniformity of the trilayer stacks has been monitored by acquiring PL with a gradually defocused excitation beam. The acquired PL spectra are displayed in Figure 1d. For an excitation spot size of 10 μ m, the defect emission remains strongly suppressed. For larger spot sizes, a broad low-energy peak starts to emerge, probably due to defect related emission in MoSe₂ in areas which are not fully capped. These measurements are a proof of concept, demonstrating that this approach, when optimized, should enable the fabrication of large area CVD-grown heterostructures with excellent optical quality.

Previous attempts at improving the optical properties of CVD-grown MoSe₂ using HBr treatment²⁹ or the isoelectronic impurity substitution³⁰ have met with only partial success;

impurity-bound excitons still remained the most prominent component of the emission spectrum. Low-temperature PL spectra consisted in broad features, wherein free exciton emission could be identified only after fitting. The optical properties of exfoliated MoS₂ have been improved by superacid treatment.⁵¹ In our case, the simple act of bringing MoSe₂ in intimate physical contact with MoS₂, a procedure that can be performed after growth and does not require any chemical functionalization, results in a virtually complete suppression of emission from the impurity-bound states and a spectrum in which the trion and exciton resonances can be clearly resolved.

Our defect healing hypothesis is further supported by the results obtained from DFT simulations. We have calculated the gain of energy in a heterobilayer MoSe₂/MoS₂ using two models: A single Se vacancy in the MoSe₂ layer (Model 1) and a single S vacancy in the MoS₂ layer together with a single S substitution in the MoSe₂ layer (Model 2). This corresponds to the transfer energy of a S atom from pristine MoS₂ to heal a defect in MoSe₂. We observe a significant energy gain of 18 kJ mol⁻¹ (180 meV) per S transfer from MoS₂ to the MoSe₂ defect, which shows that defect healing in MoSe₂ by MoS₂ is thermodynamically favored. The formation energy of a S vacancy in a MoS₂ monolayer has been theoretically estimated in the 1.3–1.5 eV range.^{52,53} We consider these as upper bounds for the energy barrier of the transfer of a S atom to fill a Se vacancy in MoSe₂, because this is not a static process but a transfer between two neighboring layers.

We have also calculated the band structures of a perfect MoSe₂ monolayer, MoSe₂ monolayer with one Se vacancy, and MoSe₂ monolayer with one Se→S substitution. We observe that Se vacancies introduce strongly localized states in the bandgap of MoSe₂, 0.92 eV above the top of the valence band (see Figure 2). These are dispersionless and act as trap centers. Healing the Se vacancy with S substitution restores the band structure of a nearly perfect MoSe₂ monolayer. In the studied 5 \times 5 supercell model (2% Se→S substitutions), the bandgap increases by only 1 meV.

Charge Transfer-Mediated Valley Polarization. First signatures of charge transfer between the layers are seen in the comparison between PL intensities of as grown layers and trilayers, shown in Figure S2 of SI. Additional insights is provided by the spatial correlation of the PL intensity of the

MoS₂ and MoSe₂. The integrated spatial map of the MoSe₂ A exciton is shown in Figure 3a. The signal is particularly intense

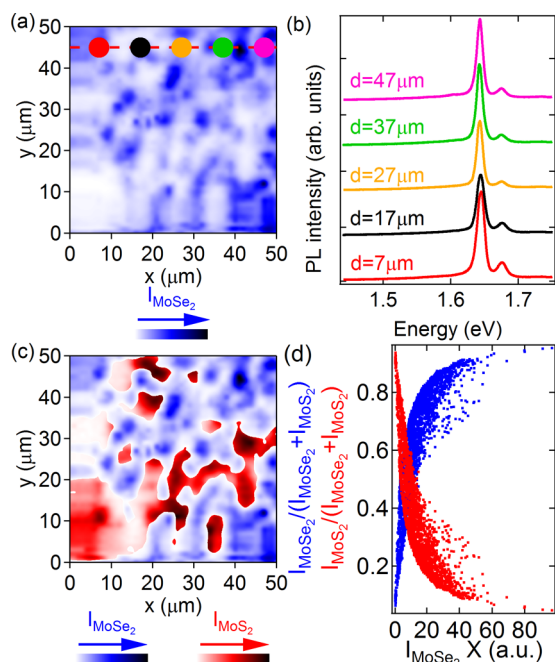


Figure 3. (a) Spatial map of the integrated intensity of A exciton of encapsulated MoSe₂. The dashed line schematically illustrates the direction along which the spectra of panel b have been extracted. The colored circles mark the position within the map of the spectra having the corresponding color in panel b. (b) μ PL spectra extracted at positions marked by circles of the corresponding color in panel a. The spectra are vertically offset for clarity. (c) Mapping of the integrated intensity of MoS₂ X_A overlaid on the same spatial map of MoSe₂ X_A. (d) Integrated intensity of MoSe₂ X_A and MoS₂ X_A as a function of the intensity of MoSe₂ X_A.

at positions where the overlap between the three layers is good and the material does not have a large number of defects. This implies that the bright spots do not necessarily have a triangular shape. This map provides an additional opportunity to

demonstrate the high degree of uniformity of the emission of MoSe₂ incorporated in a heterostructure, by extracting μ PL spectra. We show in Figure 3b five μ PL spectra measured at 10 μ m distance from one another. The spectra have been normalized by the integration time. We note that emission from defect states is consistently absent in the five spectra and the similar line shape points to a good uniformity of the emission over the full mapped area. In Figure 3c, we overlay the intensity map of MoS₂ with that of MoSe₂, forcing the areas having the lowest signal from the latter to be transparent. These areas correspond to zones where the signal from MoS₂ is highest. We quantify the observed intensity (anti)correlation by plotting in Figure 3d the ratio between the intensity of A exciton in one material normalized by the total emission of both materials [$I_{\text{MoX}_2}/(I_{\text{MoS}_2} + I_{\text{MoSe}_2})$, where X = S or Se]. It can be noted that when the emission of MoSe₂ becomes more pronounced, the emission of MoS₂ decreases correspondingly. This is fully consistent with charge transfer. In positions where the three layers overlap efficiently, charge transfer induces a quenching of the MoS₂ PL. At the same time, the PL from MoSe₂ is particularly intense at these positions owing to efficient defect healing, but still weaker than in as-grown MoSe₂ samples (see Figure S2 in SI).

Photoluminescence excitation (PLE) spectroscopy is a useful tool to investigate more thoroughly the charge transfer processes. We focus our measurements on MoSe₂ incorporated in a trilayer stack. In Figure 4a, we show the PLE measured without making use of polarization optics. Both PLEs of the A exciton and trion have been normalized by the weakest intensity measured at an excitation energy of 1.999 eV in order to be able to compare the enhancement effects for the exciton and trion. The integrated intensities of both peaks show a pronounced maximum when the excitation energy is close to the resonance with the B exciton of MoSe₂.⁵⁴ The integrated intensity is consistently lower than this maximum at other excitation energies, including those corresponding to resonances in MoS₂ (see, for example, the weak peak corresponding to MoS₂ X_A, appearing as a shoulder of the main PLE peak in Figure 4a). This is the exact opposite of what is observed in a heterostructure system where energy transfer has been

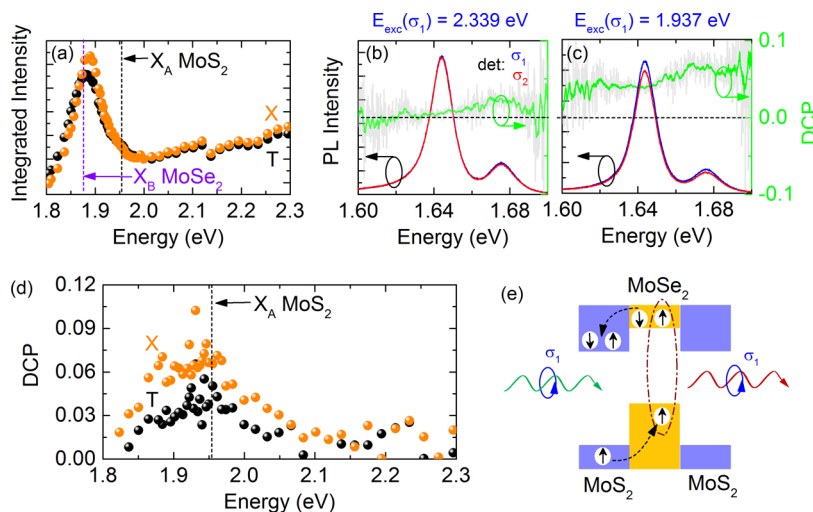


Figure 4. (a) Normalized integrated intensity of MoSe₂ X_A and T as a function of the excitation energy. Polarization-resolved μ PL spectra and DCP of MoSe₂ excited (b) far off MoS₂ X_A resonance and (c) on resonance with MoS₂ X_A. (d) Integrated DCP as a function of the excitation energy. (e) Schematic illustration of spin transfer under excitation resonant with MoS₂ X_A.

demonstrated. In such system, the emission intensity of one material is significantly enhanced when the excitation energy is resonant with excitonic transitions of the other material.⁵⁵ This consideration allows us to safely rule out energy transfer between the different layers.

To investigate whether the trilayer stack shows charge transfer, we initially consider the normalized intensity of exciton and trion when the excitation energy is resonant with excitonic transitions of MoS₂. For resonances with both A and B exciton of MoS₂, the emission intensity is enhanced more for MoSe₂ A exciton than for the trion. This is consistent with the presence of a large n-type background doping (free electrons) in CVD-grown MoSe₂, which gives rise to strong charged exciton emission even in the absence of gating (see Figure 1a). For a MoS₂/MoSe₂ heterojunction, the band alignment promotes the transfer of holes from MoS₂ to MoSe₂.³⁵ When we optically excite the trilayer stack, we induce a net transfer of holes from MoS₂ to MoSe₂, resulting in a relatively stronger emission of the neutral exciton as compared to the trion.⁵⁵

We also performed circular polarization resolved PLE focusing on MoSe₂ incorporated in a trilayer stack. When the excitation energy is far from resonance (A or B excitons of both materials), MoSe₂ shows an extremely small valley polarization. This is illustrated by the polarization-resolved μ PL spectra of Figure 4b, where a negligibly small DCP, defined as $DCP = [I_{\sigma_1} - I_{\sigma_2}] / [I_{\sigma_1} + I_{\sigma_2}]$, is observed across the entire energy range of interest.³⁸ When the excitation energy of the laser is tuned to the proximity of the resonance with the A exciton of MoS₂, a significant valley polarization accumulates. An example of polarization-resolved spectra at an excitation energy resonant with A exciton of MoS₂ is shown in Figure 4c. The valley polarization is quantified by a slightly positive DCP at energies around A exciton and trion of MoSe₂ (see Figure 4c). In Figure 4d, we illustrate the excitation energy dependence of the integrated DCP (estimated by extracting the relevant integrated intensity of exciton and trion with Gaussian fits). We notice a significant increase of the integrated DCP at energies corresponding to the A exciton resonance of MoS₂ (see dashed line in Figure 4d) with a low energy shoulder possibly related to an enhanced DCP at excitation energies corresponding to MoSe₂ B exciton.³⁸ Polarization-resolved electroluminescence of single and multilayer MoSe₂⁵⁶ as well as polarization-resolved PL of indirect excitons emitted by a WSe₂/MoSe₂ heterostructure⁵⁷ have demonstrated higher degree of circular polarization. However, the mechanisms leading to polarized emission in these systems are fundamentally different from those yielding polarized PL of MoSe₂,⁵⁶ which is virtually impossible to achieve unless the PL is excited using quasi-resonant excitation in X_A of MoSe₂.^{38–40}

We ascribe the observed MoSe₂ valley polarization to the hole transfer from MoS₂ with a mechanism schematically illustrated in Figure 4e. We excite the trilayer stack with circularly polarized light with a given helicity and in resonance with the A exciton of MoS₂. The valley polarization directly created in MoSe₂ is quickly lost, which results in a negligible DCP, similarly to nonresonant excitation (see Figure 4d). Resonant excitation in MoS₂ creates valley polarization for a duration estimated in the hundreds of femtoseconds range.⁵⁸ Charge transfer in van der Waals heterostructures is an ultrafast process with upper bounds in the tens of femtoseconds range (hole transfer from MoS₂ has been reported to be faster than 50 fs³⁶). During this very rapid transfer, we assume that the hole spin (and thus valley due to the large spin orbit splitting in the

valence band) is conserved, and because of the excess electron population in the MoSe₂ layer, the injected hole forms an exciton populating the valley corresponding to the helicity of the incoming light. These excitons have presumably a low kinetic energy (no excess energy of the photocreated hole), which slows down significantly the intervalley scattering rate due to electron–hole exchange interaction.⁵⁹ As a result, this hole transfer is responsible for the observed valley polarization.

Conclusions. A detailed investigation of the optical properties of MoS₂/MoSe₂/MoS₂ trilayers reveals that stacking dramatically improves the optical quality of CVD-grown MoSe₂, essentially eliminating all defect bound exciton and MoS₂-related emission. These results open the way to using CVD-grown TMDCs for applications and studies that require materials with excellent optical quality. Photoluminescence spectra from MoSe₂ in a trilayer stack are dominated by narrow neutral and charged A exciton emission, resembling the spectrum of a high quality mechanically exfoliated flake. Density functional calculations confirm a defect healing scenario in which S atoms replace Se vacancies. Circular polarization resolved PL measurements demonstrate that MoSe₂ exhibits a significant valley polarization even when the excitation energy is far from the A exciton resonance. This behavior is the signature of an efficient spin-conserving hole transfer from MoS₂ to MoSe₂. Our approach provides a robust and straightforward method of healing defects in CVD-grown samples, which might also be beneficial for the transport properties of these materials.

Methods. Sample Preparation. The sample with MoS₂/MoSe₂/MoS₂ trilayer stacks, schematically shown in Figure 1a, was obtained by two separate transfer steps using a wet transfer KOH method.³² First of all, the upper-MoS₂ was transferred onto the as-grown MoSe₂ monolayer on sapphire.^{24,34} Subsequently, the MoS₂/MoSe₂ stack was transferred onto an as-grown bottom MoS₂ monolayer on sapphire. For both transfers, sapphire chips with material (upper-MoS₂ or MoS₂/MoSe₂ stack) were first spin coated with PMMA 950 at 1500 rpm for 60 s and baked at 180 °C for 5 min. The films were detached in KOH (30%) at moderate temperatures (70 °C), washed three times in deionized water, transferred onto sapphire with the stacking layer (MoSe₂ or bottom-MoS₂), and dried at 50 °C for 30 min. The PMMA was removed by dipping the sample in acetone for 12 h, followed by rinsing with isopropanol and drying in a N₂ flow. Such method provides a polymer clean interface of stacks and minimal damage of material caused by the transfer process. Using this method, a large area film with monolayer (MoS₂), bilayer (MoSe₂/MoS₂, MoS₂/MoSe₂, and MoS₂/MoSe₂ stacks) and trilayer (MoS₂/MoSe₂/MoS₂ stack) was obtained. A micrograph of a representative transfer area is shown in Figure 1b. To determine the position of areas with different number of stacked layers, Ni markers were deposited.

Optical Measurements. For the optical characterization, the sapphire substrate was mounted on the cold finger of a He-flow cryostat. The excitation was provided either by a CW frequency-doubled solid-state laser emitting at 532 nm or by the frequency-doubled output of an optical parametric oscillator (OPO), synchronously pumped by a mode-locked Ti:sapphire laser. The typical temporal pulse width was 300 fs, with a repetition rate of 80 MHz. The excitation beam was focused on the sample by a 50× microscope objective, giving a spot size of approximately 1 μ m and having a numerical aperture of 0.55. The emitted PL was collected through the

same objective and redirected to a spectrometer equipped with a liquid nitrogen cooled CCD camera or (for time-resolved measurements) to an imaging spectrometer and detected using a synchroscan streak camera with the temporal resolution set to 5 ps. All the spectra have been measured at 5K, unless otherwise specified.

For spatial mapping the emission has been monitored while the optical cryostat was displaced with respect to the microscope objective using high precision motorized x - y translation stages (1 μm step). The integrated intensity of given features (e.g., A exciton emission) has been obtained by performing Gaussian fitting of the measured μPL spectra.

Density Functional Theory of Band Structure in MoSe₂/MoS₂ Heterobilayers. We have calculated heterobilayers made of MoSe₂ and MoS₂ monolayers, using DFT as implemented in the Crystal09 software.⁶⁰ We employed all-electron Gaussian-type bases of triple-quality,^{61,62} while Mo atoms were treated with the HAYWSC-311(d31)G basis with effective core potential set,⁶³ together with the PBE gradient corrected density functional.⁶⁴ London-dispersion interactions were accounted for using the approach proposed by Grimme (DFT-D3).⁶⁵ Full optimization of atomic positions and lattice vectors was performed on both models: Model 1 with perfect MoS₂ monolayer and one Se vacancy in the MoSe₂ monolayer, and Model 2 with one S vacancy in the MoS₂ monolayer and one S substitution in the MoSe₂ monolayer. The models are built of 5 \times 5 supercells (see Figure 2).

Geometry optimization only slightly alters the lattice geometry (see Table 1).

Table 1. Structural Parameters of MoS₂/MoSe₂ Defective Heterostructures Giving the Lattice Constant (a) and Interlayer Metal-to-Metal Distances (d)

System	a (Å)	d (Å)
MoS ₂ 1L	3.171	
MoSe ₂ 1L	3.251	
MoS ₂ /MoSe ₂	3.214	6.21
Model 1	3.206	6.30
Model 2	3.203	6.26

For the large supercells, which still overestimate the defect density in experiment, the defects studied here do not introduce any drastic changes into the structural properties of the systems. However, we note that we are constrained with the commensurate models of a heterobilayer, in which the corresponding monolayers are slightly distorted compared with the relaxed monolayers 3.251 and 3.171 Å for MoSe₂ and MoS₂, respectively. For a perfect heterobilayer (3.214 Å), this gives 1.15% compression of MoSe₂ and 1.4% elongation of MoS₂.

■ ASSOCIATED CONTENT

● Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.7b00904.

Low-temperature μPL spectra, comparison of μPL spectra of as-grown versus trilayer samples, temperature-dependent μPL spectra, and time-resolved PL (PDF)

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Notes

The authors declare no competing financial interest.

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