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Interlayer Excitons in Transition-Metal Dichalcogenide Heterobilayers

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In heterobilayers consisting of different transition-metal dichalcogenide (TMDC) monolayers, optically excited electron–hole pairs can be spatially separated into the adjacent layers due to a type-II band alignment. However, they remain Coulomb correlated and form interlayer excitons (ILEs), which recombine radiatively. While these ILEs are observed in several TMDC material combinations, their characters and properties depend on the specific system. Herein, some of these peculiarities are demonstrated by comparing studies performed on two different heterobilayer combinations: MoS$_2$–WSe$_2$ and MoSe$_2$–WSe$_2$.

1. Introduction

In recent years, 2D crystal structures have emerged as a fascinating new field of solid-state physics. Many layered materials can easily be thinned down to 2D sheets by means of mechanical exfoliation,[1] and the electronic structure of these atomically thin layers may differ from that of their corresponding bulk crystals. For example, while MoS$_2$ and related transition-metal dichalcogenides (TMDCs) such as WSe$_2$ and MoSe$_2$ are indirect-gap semiconductors in the bulk, a transition to a direct band gap occurs as they are thinned down to a single layer.[2–4]

In TMDC monolayers, spin and valley degrees of freedom are coupled, and the different valleys are directly addressable using circularly polarized light,[5,6] making these materials highly interesting for potential valleytronic applications. Using external magnetic fields, the energy degeneracy of the different valleys can be lifted, and a valley polarization can be induced in some cases.[7–13] The optical response of TMDC monolayers is heavily modified due to excitonic effects. Given the strict 2D confinement of carriers within the monolayer and the weak, anisotropic dielectric screening due to the low-dielectric-constant environment surrounding the monolayer, excitons have binding energies on the order of 500 meV,[14–18] making them stable well beyond room temperature.

While various TMDCs share the same crystal structure and have qualitatively similar band structures, they are characterized by different lattice constants as well as different absolute values of band gap and band offsets with respect to the vacuum level.[19] Therefore, several combinations of TMDCs lead to a type-II (staggered) band alignment,[19,20] when they are stacked into a heterobilayer. Thus, electron–hole pairs that are optically generated in either of the constituent layers become spatially separated within a few tens of femtoseconds.[21,22] These spatially separated charge carriers can form interlayer excitons (ILEs) on ultrafast timescales,[23] which have been observed directly in photoluminescence (PL) for several

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TMDC heterobilayer combinations. Such ILEs have already been studied in conventional semiconductor heterostructures, e.g., coupled quantum wells. There, different aspects such as exciton–exciton interaction and bosonic condensation of excitons were investigated, but only at low temperatures, limited by the weak ILE binding energies in these systems. ILEs in TMDC heterostructures are characterized by binding energies which significantly exceed those in GaAs heterostructures, making them stable beyond room temperature and robust against dissociation in applied electric fields. In addition, they have long radiative lifetimes, in stark contrast to intralayer excitons in TMDC monolayers, where subpicosecond radiative recombination can be observed. These long lifetimes potentially allow for cooling ILEs to very low lattice temperatures, a prerequisite for exciton condensation. With this unique combination of properties, TMDC ILEs are a highly promising novel platform for studying exciton–exciton interactions. Furthermore, the combination of 2D crystals into a van der Waals heterostructure yields a novel degree of freedom, the relative crystallographic orientation of adjacent layers (twist angle). In the case of TMDC heterobilayers, this is of special importance due to the band structure of the TMDCs: the real-space alignment also influences the alignment of the band extrema, which are localized at the corners of the Brillouin zone. Depending on the twist angle, the band extrema for conduction and valence bands of different layers may become misaligned in reciprocal space or yield an alignment which is not found in naturally stacked homobilayers. For very small twist angles, Moiré effects lead to a periodic lateral potential modulation, which can strongly influence inter- and intralayer exciton properties in TMDC heterobilayers.

Here, we directly compare experimental results for two different TMDC heterobilayer combinations. While ILE can be observed in both systems at room temperature, there are pronounced differences: ILE are observable in the MoS$_2$–WSe$_2$ combination for any twist angle. By contrast, for MoSe$_2$–WSe$_2$, only careful crystallographic alignment of the layers yields an observable ILE PL emission. However, for such structures, the ILE dominates the PL spectrum at low temperatures, allowing us to study subtle effects, such as exciton–exciton interaction and the influence of external magnetic fields.

2. Results and Discussion

2.1. Comparison of Different Material Combinations

We directly compare room-temperature PL spectra for MoS$_2$–WSe$_2$ and MoSe$_2$–WSe$_2$ heterobilayers in Figure 1. The twist angle for these samples was determined by second-harmonic-generation (SHG) microscopy (not shown), exploiting the peculiar polarization dependence of SHG from TMDC monolayers. For both structures, we observe pronounced PL emission corresponding to the A exciton emission of the constituent monolayers. In the case of MoS$_2$–WSe$_2$, we find an additional emission feature, which we attribute to the ILE, close to, but slightly below, the WSe$_2$ emission, in agreement with the initial study on this material combination. For the MoSe$_2$–WSe$_2$ heterobilayer, the ILE emission is spectrally well separated by more than 200 meV from the residual monolayer emission, making it potentially more suitable for spectroscopic studies.

This is confirmed in low-temperature PL measurements of the two samples, as shown in Figure 2. For MoS$_2$–WSe$_2$, the ILE is still observable at 90 and 60 K (the red dotted line serves as guide to the eye). However, as the temperature is lowered further, pronounced emission from the WSe$_2$ trion (T) and emission associated with defects (L) in WSe$_2$ monolayers (marked with green dotted lines) overlap the ILE spectral region. For comparison, a scaled ILE spectrum recorded at 90 K is superimposed onto the spectrum measured at 4 K. Thus, it is not easily possible to
identify the ILE emission at liquid-helium temperature. In later sections, we will discuss how the peculiar character of ILEs in this material combination, which is revealed by analyzing the effect of twist angle on ILE energy, contributes to the low ILE PL yield at low temperatures.

By contrast, in the MoSe$_2$–WSe$_2$ heterobilayer, decreasing the sample temperature drastically increases, both, the absolute PL yield of the ILE and its emission intensity relative to the residual PL from the monolayer exciton features, as Figure 2b shows. The increased PL yield is accompanied by a pronounced reduction of the ILE PL linewidth, which is reduced to about 25 meV at 4 K. This makes this material combination ideal for studying subtle effects on ILE by low-temperature PL spectroscopy. We will discuss two examples in the following sections.

2.2. Material-Specific Properties: MoS$_2$–WSe$_2$

2.2.1. Twist-Angle Dependence of ILE Emission

ILEs in TMDCs were first reported in a seminal work\cite{24} in 2014 for MoS$_2$–WSe$_2$ heterobilayers. To elucidate the properties of the ILEs in this material combination, we fabricated a large number of heterobilayers with different twist angles, which we determined by SHG microscopy. Each of these samples was studied by PL at room temperature. Figure 3a representatively shows two typical spectra. The samples depicted here present two limiting cases of twist angle, with the one shown in the top panel having a twist angle close to 30° and the one in the bottom panel having a twist angle close to 60°, corresponding to antialignment of the adjacent layers. Clearly, for both samples, we see a pronounced ILE emission peak at about 1.6 eV, as well as the emission associated with the MoS$_2$ and WSe$_2$ monolayers. We note that the emission associated with the monolayers is substantially quenched in the heterobilayer region compared with isolated monolayers. The spectra are well described using three Gaussian fit functions (solid lines) for ILE, MoS$_2$, and WSe$_2$, respectively. Remarkably, there is a strong energetic shift of the ILE peak position between the samples (the red dotted lines serve as guide to the eye), whereas the monolayer emission does not shift significantly. For each of the heterobilayers in our study, we performed micro-PL mapping and analyzed the spectra for the region where ILE emission was observed. By averaging the ILE PL peak energies extracted from these maps, we were able to compensate for the spatial inhomogeneity of ILE emission. The results of this process are summarized in Figure 3b (red solid dots, with the error bars indicating the standard deviation). We clearly observe a systematic dependence of the ILE emission energy on twist angle, with samples twisted close to 30° having emission energies about 50 meV higher than aligned (0°) or antialigned (60°) samples.

To explain this systematic dependency, we turn to density functional theory (DFT) calculations. Here, the band structure of commensurate, periodic supercells constructed for several twist angles is calculated (see supplementary information in the study by Kunstmann et al.\cite{33} for details). From these calculations, we are able to extract the systematic dependency of, e.g., the K–K and Γ–K interlayer transition energies on twist angle. To allow for direct comparison with experimental data, we rigidly shift the values obtained by DFT by 445 meV. The results are depicted by red and yellow crosses in Figure 3b. Remarkably, the twist angle-dependent shift of the Γ–K transition quantitatively matches the twist angle dependency observed in the experiments, even including the slight asymmetry between aligned and antialigned structures. By contrast, the K–K transition shows the opposite functional behavior, with an increase in transition energy toward 0° and 60° twist angles. Thus, we can assign the ILE transition we observe in the MoS$_2$–WSe$_2$ heterobilayer to be of a momentum space-indirect Γ–K type. This association also serves to explain the lack of ILE PL intensity dependency on the twist angle, as the Γ–K transition always corresponds to the same momentum space mismatch, regardless of twist. By contrast, the K–K transition is only direct in momentum space for 0° and 60° twist angles, so that a change in the transition character from direct for aligned to indirect for twisted structures is expected. In fact, this is observed in experiments on the MoSe$_2$–WSe$_2$ heterobilayer system and manifests itself as a drop in the PL intensity for twisted structures.\cite{29}

The momentum space-indirect character of the ILE in our structures also serves to explain the lack of PL yield increase with decreasing temperatures (see Figure 2a), as the Γ–K ILE recombination is phonon assisted. Our DFT calculations allow us to further elucidate the properties of the Γ–K ILE by considering the partial charge densities of electron and hole states. We find that, as expected from a basic band alignment model, electrons are fully localized in the MoS$_2$ layer, whereas holes are fully localized in the WSe$_2$ layer at the respective K points, leading to a vanishing real-space overlap of K-point electron and hole wave functions. In stark contrast, holes at Γ are strongly delocalized and form a hybrid state, which has about 24% partial charge density within the MoS$_2$ layer. Thus, there is a significant real-space wave function overlap for the Γ–K ILE, facilitating radiative recombination and resulting in a large binding energy. Calculating the binding energies of the different ILEs using the quantum-electrostatic heterostructure model\cite{39} and a variational wave function ansatz, we find significantly different values. For the Γ–K ILE, we obtain 550 meV, similar to values for TMDC intralayer excitons,\cite{16} due to the hole delocalization enhancing the electron–hole Coulomb interaction. By contrast, for the
K–K ILE, we find a much smaller value of 290 meV, in line with previous calculations.^[40,51]^2

2.3. Material-Specific Properties: MoSe$_2$–WSe$_2$

Due to its spectrally well-separated ILE emission peak and its narrow emission linewidth at low temperatures, the MoSe$_2$–WSe$_2$ heterobilayer system is ideally suited for the study of subtle effects. Here, we present two examples.

2.3.1. Exciton–Exciton Interaction

First, we study the power dependence of the ILE peak in a wide range of excitation densities. Figure 4a shows a series of normalized PL spectra of ILE emissions in an antialigned MoSe$_2$–WSe$_2$ heterobilayer measured at 4.2 K. The power of the excitation laser (pulse length full width at half maximum 180 fs, pulse repetition rate 80 MHz, and central wavelength 560 nm) was tuned over more than two orders of magnitude for this measurement series. Remarkably, we observe that the ILE emission peak systematically blueshifts by more than 10 meV as the pump power is increased, initially without any change in the spectral shape or linewidth. Such pronounced shifts are absent for the residual emission from the constituent monolayers in this sample (not shown). A saturation of the blueshift, together with an increasing linewidth of the PL emission, is only observed for the largest excitation powers. The ILE peak position data extracted from this series are shown in Figure 4b.

This blueshift is a direct consequence of the nature of the ILE: due to the spatial separation of electron and hole into adjacent layers, they have a permanent, well-oriented electric dipole moment. With increasing ILE density, dipole–dipole interaction between ILE (as shown in the inset of Figure 4b) blueshifts the ILE energy. This effect has previously been studied in coupled quantum well systems^[34,35]^ and was also observed by several groups in TMDC heterobilayers.^[^36,37,38,39]^ In the limit of low ILE density, it is expected to scale linearly with the density. However, as shown in Figure 4b, we observe a near-logarithmic dependency of the blueshift on excitation power. This seemingly contradictory observation can be reconciled with the expected linear behavior by studying ILE dynamics using time-resolved techniques. In our study, we found a very strong dependence of the effective ILE PL lifetime on excitation power, with a significant decrease for increasing excitation power. Thus, the time-averaged ILE density has a sublinear dependence on excitation power, details are provided in the study by Nagler et al.^[50]^ By comparing the energetic positions of the ILE for strong, pulsed excitation with those for weak, continuous-wave excitation (not shown), we find a maximum blueshift of about 20 meV. Using a basic electrostatic approach,^[52]^ we can determine a lower boundary for the ILE density $n_{\text{ILE}}$ corresponding to this blueshift. We find the value of $n_{\text{ILE}} = 4 \times 10^{10}$ cm$^{-2}$. Given that this value is extracted using the time-integrated PL spectra, the initial ILE density after pulsed excitation will be considerably higher, easily exceeding the values that can be generated in coupled quantum well systems.

2.4. Giant Magnetic Splitting due to Intervalley Transitions

Their relatively low PL linewidth and high PL yield at low temperatures make ILE in MoSe$_2$–WSe$_2$ heterobilayers ideally suited for studying the effects of an external magnetic field. Figure 5a shows helicity-resolved ILE PL spectra measured using linearly polarized excitation, so that both valleys of the constituent monolayers are excited equally. Without magnetic field (top panel), the $\sigma^+$ and $\sigma^-$ emission peaks have the same energy and intensity. By contrast, for a magnetic field of 30 T applied perpendicular to

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**Figure 4.** a) Excitation power-dependent PL spectra of antialigned MoSe$_2$–WSe$_2$ heterobilayer measured at 4.2 K. b) ILE peak position as a function of excitation power (note the logarithmic scale). The inset shows the dipolar interaction of ILE.

**Figure 5.** a) Helicity-resolved PL spectra of an antialigned (twist angle 54°) MoSe$_2$–WSe$_2$ heterobilayer measured at 4.2 K for applied magnetic fields of 0 and 30 T in Faraday configuration. b) Magnetic field dependence of valley splitting. The solid line indicates a linear fit to the data (blue dots). c) Schematic of valley alignment for antialigned heterobilayer in reciprocal space. MoSe$_2$ conduction band valleys and WSe$_2$ valence band valleys with different indices are aligned, so that intervalley ILE transitions are direct in reciprocal space. d) Schematic of magnetic field-induced shifts of conduction and valence band valleys corresponding to the momentum space-direct ILE transition in a magnetic field.
the layer plane, we find a pronounced energetic splitting between $\sigma^+$ and $\sigma^-$ of about 26 meV, exceeding the linewidth of the ILE peaks. In addition, the higher-energy ILE emission is strongly quenched, indicating a pronounced, magnetic field-induced valley polarization. In a series of helicity-resolved PL measurements at different applied magnetic fields, we are able to track the evolution of these effects. We find a splitting of the $\sigma^+$ and $\sigma^-$ ILE emissions, which scale linearly with the magnitude of the applied magnetic field, as shown in Figure 5b. From a fit to this data, we are able to determine an effective g factor of $-15.1 \pm 0.1$ for the ILE, which was recently confirmed in another study.[46] This value is about four times larger than the values observed for excitons in TMDC monolayers, where $g$ factors close to $-4$ were reported for many materials.[7–13]

For TMDC monolayers, the observed $g$ factor of about $-4$ can be explained by considering the contributions to the valley-selective energy shifts of conduction and valence band states[7–11] in conjunction with the selection rules for interband recombination of monolayer excitons. The three relevant contributions arise from magnetic moments related to the spin, the atomic orbitals, and the valleys in conduction and valence bands. The excitonic interband transitions in the monolayers are spin conserving; thus, assuming identical spin-related magnetic moments for conduction and valence bands, both bands shift in the same direction and by the same amount in a magnetic field. Hence, there is no valley-selective energetic shift due to the spin magnetic moments. The valence bands carry a nonzero, valley-contrasting orbital magnetic moment $\mu_B = \pm 2$ leading to an overall splitting between the valley-selective transitions of $-4\mu_B B$. The contribution from the valley magnetic moments is given by $\pm \mu^v_k = m_0/m_e$ for the conduction band and $\pm \mu^c_k = m_0/m_e$ for the valence band in the K+/K− valley, respectively.

As the effective masses in valence and conduction bands are very close to each other for most TMDCs,[53] the valley magnetic moments are almost identical in conduction and valence bands. The excitonic interband transitions in TMDC monolayers are valley conserving, and therefore, just as discussed earlier for the spin, conduction and valence bands move by similar amounts in the same direction for each valley, yielding almost no valley-selective energetic shift, so that the total effective g factor expected for TMDC monolayers is about $-4$, arising almost exclusively due to the valley-contrasting valley band atomic orbital magnetic moments, with slight deviations due to effective mass differences modifying valley magnetic moments in conduction and valence bands. However, our antialigned heterobilayer has a peculiar alignment in reciprocal space that does not occur in TMDC monolayers: the conduction-band K+ valleys of MoSe$_2$ are situated directly above the valence-band K− valleys of WSe$_2$ and vice versa, so that intervalley ILE transitions are direct in momentum space, whereas intravalley transitions have a large momentum mismatch. Thus, the ILE emission observed in our PL experiments corresponds to intervalley transitions between the MoSe$_2$ conduction and WSe$_2$ valence bands, as shown in Figure 5c.

The total valley-selective energy shift for these intervalley ILE transitions can be determined based on the three contributions discussed earlier for the monolayers and is shown in Figure 5d. Assuming that the ILE transitions are spin conserving, there is no valley-selective shift due to the spin. The valley-contrasting orbital magnetic moments in the valence band yield a splitting of $-4\mu_B B$, just as for monolayers. However, as the valley index changes in the ILE transitions, the contributions of the valley magnetic moments to the valley-selective splitting do not cancel, but add up, leading to the observed large effective $g$ factor for the ILE.[32] Using theory values[33] for the conduction- and valence-band effective masses of the constituent layers, we find an effective $g$ factor of $-13.1$ for the interlayer transition, in qualitative agreement with the experimentally observed value.

3. Conclusion

In summary, we have presented experimental and theoretical studies of ILEs in two different TMDC heterobilayer systems, MoS$_2$–WSe$_2$ and MoSe$_2$–WSe$_2$. While both material combinations host optically bright ILEs which are readily observable by PL at room temperature, there are system-specific peculiarities. The ILEs that we observe in MoS$_2$–WSe$_2$ are indirect in $k$ space, corresponding to a $\Gamma$–K interlayer transition, and thus observable irrespective of the interlayer twist angle. Due to a pronounced hybridization of the hole wave function at $\Gamma$, they have only partial interlayer character. By contrast, in MoSe$_2$–WSe$_2$, ILEs only recombine radiatively for heterobilayers close to crystallographic (anti)alignment. For this material system, the ILE transition dominates the PL spectrum at low temperatures and shows a very low linewidth, so that subtle effects such as exciton–exciton interaction become discernible in PL spectroscopy. The twist angle can be exploited in this material system to make intervalley interlayer transitions direct in reciprocal space, leading to a giant magnetic splitting of the ILE in an external magnetic field.

4. Experimental Section

Sample Preparation: Our heterostructures were fabricated by means of a deterministic transfer process.[54] For this, we initially exfoliated TMDC flakes from bulk crystals (HQ graphene and 2D semiconductors) onto intermediate polymethylsiloxane (PDMS) substrates. Monolayer regions of these flakes were identified via optical microscopy. Then, we subsequently transferred the constituent flakes of a heterostructure onto the target substrate, a silicon wafer piece covered with a SiO$_2$ layer and predefined metal markers. For the MoSe$_2$–WSe$_2$ material combination, we carefully aligned well-cleaved edges of the monolayer parts of the two flakes to yield crystallographic alignment of the layers. For the MoS$_2$–WSe$_2$ material combination, a large number of samples with different alignments were fabricated. Subsequent to the transfer, the samples were annealed in vacuum at a temperature of 150 °C for several hours to improve interlayer coupling.[55]

Optical Spectroscopy: Temperature-dependent PL measurements were performed in a self-built confocal microscope setup. The excitation laser was coupled into a 100× microscope objective and focused to a spot diameter of less than 1 μm on the sample surface. The PL from the sample was collected using the same objective and coupled into a grating spectrometer, where it was detected using a charge-coupled device (CCD) sensor. The sample was mounted on the cold finger of a small He-flow cryostat and scanned beneath the microscope objective.

Low-temperature PL measurements in large magnetic fields were conducted at the HFML facility in Nijmegen. The sample was placed on an $x$–$y$–$z$ piezoelectric stage and cooled down to 4.2 K in a cryostat filled with liquid helium. Magnetic fields up to 30 T were applied by means of a resistive magnet in Faraday configuration. The excitation laser was focused...
onto the sample with a microscope objective resulting in a spot size of about 4 μm. The polarization of the PL was analyzed with a quarter-wave plate and a linear polarizer.

Estimation of ILE Density: We considered a mean-field approximation, which was valid in a regime where the ILE density \( n_{\text{ILE}} \) was so small that the average exciton–exciton distance was substantially larger than the interlayer distance \( d \). In addition, the exciton thermal wavelength also needs to be smaller than the average exciton–exciton distance. For a temperature of 4.5 K, we found \( \lambda_\text{th} \approx 10 \text{ nm} \). Within this approximation, \( n_{\text{ILE}} \) was related to the blueshift \( \Delta E_b \) by

\[
  n_{\text{ILE}} = \frac{\Delta E_b s_{\text{FSO}}}{4 \varepsilon c \epsilon_0 d^2} \quad (1)
\]

Here, we utilized \( s_{\text{FSO}} = 4.5 \) and an interlayer distance \( d = 1 \text{ nm} \).

DFT Calculations: DFT calculations were conducted using the PBE (Perdew, Burke and Ernzerhof) functional[56] and the DFT-TS (Tkatchenko-Scheffler) dispersion-interaction-correction scheme[57] using the projector augmented wave method[58] and a plane wave basis set with a cutoff energy of 259 eV, as implemented in the Vienna Ab initio Simulation Package (VASP). For the k-point sampling, an in-plane sampling density of 0.1 Å\(^{-1}\) was used. It was carefully checked that this density leads to converged total energies (energy differences are smaller than 1 meV atom\(^{-1}\)). The momentum space integration was conducted with a Gaussian smearing method using an energy width of 0.05 eV for all calculations. All unit cells were built with at least 10 Å separation between replicas in the perpendicular direction to achieve negligible interaction. All systems were fully structurally optimized until all interatomic forces and stresses on the unit cell were below 0.01 eV Å\(^{-1}\) and 10 kbar, respectively.

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Conflict of Interest

The authors declare no conflict of interest.

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