Valley-magnetophonon resonance for interlayer excitons

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Abstract

Heterobilayers consisting of MoSe$_2$ and WSe$_2$ monolayers can host optically bright interlayer excitons with intriguing properties such as ultralong lifetimes and pronounced circular polarization of their photoluminescence due to valley polarization, which can be induced by circularly polarized excitation or applied magnetic fields. Here, we report on the observation of an intrinsic valley-magnetophonon resonance for localized interlayer excitons promoted by intervalley hole scattering. It leads to a resonant increase of the photoluminescence polarization degree at the same field of 24.2 Tesla for H-type and R-type stacking configurations despite their vastly different excitonic energy splittings. As a microscopic mechanism of the hole intervalley scattering we identify the scattering with chiral TA phonons of MoSe$_2$ between excitonic states mixed by the long-range electron hole exchange interaction.

1. Introduction

Two-dimensional (2D) crystals and their van der Waals heterostructures (HSs) are promising candidates for novel optoelectronic devices. Among the 2D crystals, the semiconducting transition metal dichalcogenides (TMDCs) like MoS$_2$ have garnered a lot of attention due to their intriguing properties: in the monolayer (ML) limit, they are direct-gap semiconductors [1, 2] with large exciton binding energies [3] and peculiarities such as spin–valley locking [4]. The latter phenomenon, coupled with helicity-dependent interband selection rules, allows for optical initialization and readout of a coupled spin–valley polarization [5]. Many combinations of different TMDC MLs yield a type-II band alignment, which leads to interlayer charge separation. Spatially separated electron hole pairs can form so-called interlayer excitons (ILEs) in these heterobilayers [6]. Depending on the specific material combination, these ILEs may be optically bright only for specific crystallographic alignments [7] (interlayer twist) and their energy may be tunable via control of interlayer twist [8, 9]. These ILEs inherit some properties, such as spin–valley polarization [10], from the constituent TMDC MLs. However, in contrast to ML excitons, they are characterized by ultralong lifetimes [11–13] and diffusion over mesoscopic distances [10, 14], which makes them attractive for exciton-based optoelectronic devices, see [15] for a recent review.

Magneto-optical studies in high magnetic fields have been used very successfully to elucidate properties of TMDC MLs, such as exciton g factors [16], magnetic-field-induced valley polarization [17], exciton Bohr radii and masses [18, 19], dark exciton [20] and Rydberg exciton states [21, 22], as well as
the substructure and dynamics of more complex quasiparticles like biexcitons [23–26], see also [27] for a recent review. More recently, ILEs in TMDC heterobilayers have also been subjected to high magnetic fields, revealing a unique ability to engineer their effective g factor by changing the twist angle [28], which can be much larger and of the opposite sign as compared with excitons in TMDC MLs [29, 30].

For the specific material combination of WSe$_2$ and MoSe$_2$, optically bright ILEs are only observable for interlayer twist angles close to 0 or 60 degrees [7]. These configurations are also referred to as R-type (0 degree) or H-type (60 degrees) in accordance to the prevalent stacking polymorphs of TMDC multilayers. The optical selection rules for ILEs in these HSs depend on the local interlayer atomic registry [31], and therefore, the helicity of the emitted PL is not directly linked to ILE valley polarization, in contrast to TMDC MLs. In heterobilayers, the interlayer atomic registry can vary spatially due to two different effects:

- the formation of a moiré lattice [30, 32], which arises from an angular misalignment of the individual layers;
- atomic reconstruction [33, 34], in which the individual layers are slightly distorted to yield domains with perfect interlayer atomic registry separated by domain walls.

Both effects lead to exciton localization, which can be used to study highly tunable manybody phases of excitons and individual charge carriers [31, 35–37].

In two independent magneto-optical studies on ILEs in H-type HSs [28, 38], a peculiar enhancement of ILE valley polarization was found in magnetic fields of about 24 Tesla. In the latter study, this enhancement was associated with a coupling between ILEs and chiral optical phonons [39–41]. The strong electron–phonon and exciton–phonon interactions were also shown to limit the mobility [42–45], lead to formation of polarons [46–49], and produce phonon cascades [50–53].

Here, we present a joint experimental and theoretical study of ILEs in both H-type and R-type WSe$_2$/MoSe$_2$ HSs. In magneto-photoluminescence measurements, we observe a pronounced enhancement of the ILE valley polarization at about 24.2 Tesla for both types of HSs, even though their g factors, and the corresponding valley Zeeman splitting of the ILEs differ by a factor of about 3. This observation is explained as a valley-magnetophonon resonance of a hole in the localized exciton. Our theoretical analysis demonstrates that the dominant mechanism of the valley-magnetophonon resonance is the electron-spin-conserving scattering with a chiral TA phonon originating from the MoSe$_2$ ML between the excitonic states mixed by the long-range exchange interaction.

The magnetophonon resonance was predicted more than half a century ago by Gurevich and Firsov [54] as an intrinsic resonance between a pair of Landau levels and the optical phonon energy. Soon after, the spin–magnetophonon resonance between opposite electron spin states was predicted [55] and observed [56]. Spin-conserving intervalley magnetophonon resonances were also studied in conventional semiconductors [57], graphene [58] and TMDC MLs [49]. Eventually the magnetophonon resonance evolved into a powerful tool to study both the phonon and electron properties of metals, semiconductors and semiconductor nanostructures [57, 59–61]. However, despite numerous investigations and applications of the magnetophonon resonance, an intervalley spin-flip resonance was never observed before to the best of our knowledge. Thus the hole valley–magnetophonon resonance represents a novel aspect of this tool highly relevant for TMDC HSs.

2. Experimental results

Our results are summarized in figure 1. ILEs in H-type and R-type HSs are directly distinguishable by their emission energy and spectral linewidth, even in zero-field photoluminescence (PL) spectra [62]. As figure 1(a) shows, at 0 Tesla the ILE in the H-type HS has its emission peak at around 1399 meV, with a linewidth of about 13 meV. By contrast, the ILE in R-type HS emits at the significantly lower energy of about 1368 meV and has a larger linewidth of about 37 meV. In a high magnetic field (29 T spectra are depicted in figure 1(a)), helicity-resolved PL spectra reveal a pronounced energetic splitting of $\sigma^+$ and $\sigma^-$ polarized emission, combined with a pronounced change in relative emission intensities. For both H-type and R-type HSs, the lower-energy emission becomes more intense than the high-energy emission. However, for the two HSs, $\sigma^+$ and $\sigma^-$ components shift in opposite ways: for H-type, the $\sigma^-$ component shifts to higher energies, while for R-type, it is the $\sigma^+$ component. It is also directly evident that the magnitude of the field-induced shifts is far larger in the H-type HS. In order to quantify these observations, we performed continuous sweeps of the magnetic field from 0 T to 30 T (H-type) or 29 T (R-type), with helicity-resolved PL spectra taken at fixed time intervals corresponding to about 27 mT spacing between spectra. For each spectrum, the ILE signal was analyzed using an automatized Gaussian fit routine to extract its peak position and integrated intensity. From these datasets, we were able to determine the dependence of the valley splitting $\Delta E$ (defined as $\Delta E = E_{\sigma^+} - E_{\sigma^-}$) on magnetic field, as depicted in figure 1(b) (the curves consist of ~1000 connected discrete points). We clearly see a linear dependence for both types of HSs, with opposite sign
Figure 1. (a) PL spectra of R-type and H-type HSs at 0 T (black lines) and helicity-resolved PL spectra at 29 T. The vertical dashed lines indicate the splitting between $\sigma^+$ and $\sigma^-$ emission at 29 T. (b) Valley splitting in R-type (orange line) and H-type (black line) HSs as a function of magnetic field. The red lines show linear fits to the data. The vertical dashed line indicates the resonance field of 24.2 T. The red arrows indicate the valley splitting values for the different HSs at this field. (c), (d) Helicity-resolved PL intensity for H-type (c) and R-type (d) HSs as a function of magnetic field. The blue arrows indicate the resonantly increased (c)/decreased (d) PL intensity at the resonance field. The light blue arrows indicate the weaker resonant features. (e), (f) ILE PL circular DOP for H-type (e) and R-type (f) HSs calculated from data in (c) and (d). The red solid lines in (e) and (f) show fits to the data using the theoretical model. Blue arrows indicate the resonantly enhanced DOP.

and different slope. A linear fit yields the effective ILE $g$ factors of $g_{\text{eff}} = -14.8$ for the H-type and $g_{\text{eff}} = +4.7$ for the R-type HS. Close to the resonance field of 24.2 T, we note a slight deviation of the measured valley splitting from the linear behavior for both HSs. Noteworthy, the valley splitting at this resonance field differs by a factor of more than 3 between the HSs, as indicated by the red arrows.

In addition to the valley-selective shifts of ILE energies, the magnetic field also modifies the relative intensities of $\sigma^+$ and $\sigma^-$ emission. We plot the helicity-resolved integrated PL intensities as functions of magnetic field for both HSs in figures 1(c) (H-type) and 1(d) (R-type). For the H-type HS, the $\sigma^+$ emission increases almost monotonously with magnetic field, while $\sigma^-$ decreases almost monotonously. However, we note a pronounced, resonant increase of the $\sigma^+$ emission at the resonance field of 24.2 T (marked by blue arrow). In the H-type HS, this is accompanied by a resonant reduction of the $\sigma^-$ emission at the same field. By contrast, in the R-type HS, the $\sigma^-$ emission initially increases up to about 10 T, then decreases. The $\sigma^+$ emission decreases almost monotonously, but we notice a pronounced, resonant decrease at the resonance field (marked by blue arrow). In the R-type HS, this is not accompanied by an increased emission in the opposite helicity. Looking more closely, we also see two weaker resonant features for both HSs at fields slightly above and below the resonance field (marked by light blue arrows).

From these datasets, we calculate the circular degree of polarization (DOP) of the ILE emission, defined as

$$DOP = \frac{p^+ - p^-}{p^+ + p^-}$$

with the helicity-resolved PL intensities $p^+$ and $p^-$. The DOP as a function of magnetic field is depicted in figures 1(e) and (f). We note that, based on our definition (1), it is positive for the H-type HS and negative for the R-type HS. For both HSs, the absolute value of the DOP increases with increase of magnetic field. In both cases, we clearly see a resonantly increased absolute value of the DOP at the resonance field, accompanied by two additional, weaker resonant features below and above the main resonance. While the DOP for the H-type HS reaches values above 90 percent at the largest applied magnetic field, the maximum absolute value for the R-type HS is smaller than 37 percent and actually achieved at the resonance field. This difference in the maximum DOP closely corresponds to the difference of the valley splittings.

Our most surprising observation is the resonant enhancement of the DOP at the same field of 24.2 T despite the large difference of the valley splittings.
Below, we demonstrate that this is a consequence of the hole valley-magnetophonon resonance.

3. Theory

The effective exciton $g$ factors for H-type and R-type HSs and the oscillator strengths inferred from the bright PL indicate the dominant contribution of $H^h_{\text{h}}$ (A′A′) [28, 35, 63] and $R^h_{\text{h}}$ (A′B′) [29, 64] interlayer atomic registries to the optical properties in agreement with the previous studies of MoSe$_2$/WSe$_2$ HSs. These $g$ factors stem from the individual electron and hole $g$ factors as $-g_e \mp g_h$, respectively, where ‘hole’ refers to the vacant state in the valence band. From the measured values of $-14.8$ and $+4.7$ we estimate the electron and hole $g$ factors to be $g_e = 5.05$ and $g_h = 9.75$ in agreement with first principle calculations [65–68].

The resonant changes of the PL at the same magnetic field $B_{\text{res}} = 24.2$ T in both H-type and R-type HSs suggest a common resonance. Despite the large difference in the exciton valley splittings the individual electron and hole Zeeman energies are the same at a given magnetic field for both stacking configurations. Therefore, we attribute the observed resonances to the individual charge carriers.

Electron or hole intervalley scattering requires a spin flip and absorption or emission of a chiral phonon at the corner of the Brillouin zone ($K$ points). The large density of chiral phonon states strongly increases the scattering rate. Intervallel spin-magnetophonon resonances were never observed before to the best of our knowledge. Here we coin it valley-magnetophonon resonance for brevity.

The electron and hole Zeeman splittings in the field $B_{\text{res}}$ are $g_e\mu_B B_{\text{res}} = 7.1$ meV and $g_h\mu_B B_{\text{res}} = 13.7$ meV. Calculations of the phonon energies in MoSe$_2$ and WSe$_2$ [43, 69–73] demonstrate the absence of $K$ phonon modes at the Zeeman splitting of the electron. However in the vicinity of the hole Zeeman splitting there are the chiral ZA phonon mode of WSe$_2$ at 15 meV and the TA phonon mode of MoSe$_2$ at 14.7 meV. The exact energies of chiral phonons can be measured using inelastic x-ray scattering [74]. Taking into account the possible phonon energy renormalization [75, 76] we conclude that we observe a valley-magnetophonon resonance of a hole.

3.1. Valley-magnetophonon resonance in PL polarization

In order to demonstrate that the hole valley magnetophonon resonance leads to the resonant enhancement of the PL polarization we consider the exciton spin dynamics using rate equations for the four lowest intravalley and intervalley excitonic states shown in figures 2(a) and (b) (see supplementary material). We take into account radiative and nonradiative exciton recombination times, $\tau_R$ and $\tau_{\text{NR}}$, respectively, as well as the electron, $\tau_e(B)$, and hole, $\tau_h(B)$, intervalley scattering times to the lower energy states. Note that the actual hole scattering mechanism may be quite complex and involve both charge carriers in a single scattering event, as shown in the next subsection. The scattering times to the higher energy states are larger by the corresponding factors $\exp(g_e \mu_B B/k_BT)$ with $T$ being the temperature and $k_b$ being the Boltzmann constant.

For the hole valley relaxation time we assume the following form:

$$\frac{1}{\tau_h(B)} = \frac{1}{\tau_{\text{res}}} \exp\left[\frac{(B - B_{\text{res}})^2}{\Delta B^2}\right] + \frac{1}{\tau_e^{(0)}} \left(\frac{B}{B_{\text{res}}}\right)^2.$$

Here the first term stands for the resonant intervalley scattering at field $B_{\text{res}}$ with the minimum time $\tau_{\text{res}}$ and the width of the resonance $\Delta B$. The second term describes the phenomenological scattering time $\tau_e(B) \propto 1/B^2$, which corresponds to the direct spin–phonon coupling in strained HSs in small magnetic fields [77]. Similarly to this, for the electron intervalley scattering we assume $\tau_e(B) = \tau_e^{(0)}(B_{\text{res}}/B)^2$.

Figures 1(e) and (f) show that this model nicely fits the PL DOP almost in the whole range of magnetic fields from 0 to 29 T including the resonant enhancement at 24.2 T. The fit parameters are given...
3.2. Mechanism of hole valley-magnetophonon resonance

Direct intervalley scattering between Kramers degenerate states of the hole is forbidden by time reversal symmetry [43, 78, 79]. However, it becomes possible in the presence of an external magnetic field [80] or hole electron exchange interaction [81]. The excitons in our MoSe$_2$/WSe$_2$ HSSs are localized either due to the moiré potential or the domains formed by atomic reconstruction. We find that the dominant mechanism of the hole valley-magnetophonon resonance in this case is a two-step process (see supplementary material), as illustrated in figure 2(c). To be specific, let us consider the scattering from an intervalley excitonic state with longer lifetime to an intravalley state with shorter lifetime, when a hole in the exciton scatters from $K_-$ to $K_+$ valley decreasing the exciton energy, as shown in figure 2(c). At the first step, an electron from the $K_+$ valley virtually scatters to the upper (spin split) subband in the $K_-$ valley emitting a chiral TA phonon of the MoSe$_2$ ML. The phonon emission ensures energy conservation for the entire two-step scattering, and the intermediate (auxiliary) exciton state has a very short lifetime limited by the time-energy uncertainty relation. At the second step, the exciton scatters as a whole from the $K_-$ valley to the ground state in the $K_+$ valley due to the long-range electron hole exchange interaction. This step can be described as emission and reabsorption of a virtual longitudinal photon [78, 82, 83]. The efficiency of this scattering is ensured by the brightened spin-triplet exciton states and specific optical selection rules for $H^e_0$ and $R^e_0$ interlayer atomic registrys [84]. In total, the electron remains in the same state, but the hole flips its valley. The hole scattering from an intravalley exciton state in $K_-$ valley to the $K_+$ valley has the same rate, and the scattering with increase of the energy is suppressed by a Boltzmann factor.

For the scattering shown in figure 2(c), Fermi's golden rule for the scattering rate reads

$$\frac{1}{\tau_{\text{res}}} = \frac{2\pi}{\hbar} \sum_{q} \left| \langle f_q | \mathcal{H}_{\text{exch}} | a_q \rangle | \langle a_q | \mathcal{H}_{\text{e-ph}} | f_q \rangle \right|^2 \delta(E_i - E_f - \hbar \Omega_q),$$

(3)

where $i, a_q, f_q$ represent the initial, auxiliary and final states of exciton and emitted phonon with the wave vector $K_-$, $q$ and energy $\hbar \Omega_q$, $E_{i,f,a}$ are the exciton energies in the corresponding states, and $\mathcal{H}_{\text{exch}}$ and $\mathcal{H}_{\text{e-ph}}$ stand for the exchange and electron–phonon interaction Hamiltonians. In the vicinity of the magnetophonon resonance one has $E_i - E_f = \Delta_c + (g_h - g_e^c)\mu_B B_{\text{res}}$, where $\Delta_c$ is the spin orbit splitting of the MoSe$_2$ conduction band and $g_e^c$ is the electron valley $g$ factor [85, 86].

From the symmetry analysis we find that the interaction of electrons with chiral TA phonons in MoSe$_2$ ML is described by (see supplementary material)

$$\mathcal{H}_{\text{e-ph}} = \sum_{q, \pm} \frac{\hbar}{2\rho \Omega_q a^e} \Xi q \pm | b_{K_+}^e + q e^{-iq r_e} + \text{H.c.},$$

(4)

where $b_{K_+}^e$ are the phonon creation operators, $\rho$ is 2D mass density of the ML, $A$ is the normalization area, $q = (q_x \pm i q_y)/\sqrt{2}$, $\Xi$ is the intervalley deformation potential, $r_e$ is the electron coordinate, and $r_{K_+}$ are the electron valley rising and lowering operators, which conserve the electron spin. This Hamiltonian can be derived taking into account the fact that electrons and phonons at $K_\pm$ valleys have orbital angular momenta $\pm 1$ and $\mp 1$, respectively, and that the angular momentum modulo 3 should be conserved during the scattering. The phonon dispersion at the corners of the Brillouin zone has the form $\hbar \Omega_q = \hbar \Omega_0 + (\hbar q)^2/(2M)$ [71], where $M$ is the effective phonon mass at the $K$ point.

The Hamiltonian of the long-range exchange interaction between auxiliary and final states can be obtained similarly to the ML case [83]. It reads

$$\mathcal{H}_{\text{exch}} = \frac{2\pi e^2 \delta(\rho) (Kp_{\text{ex}}^e) (Kp_{\text{ex}}^f)}{\rho_0 m_0 \omega_0 \Gamma_0} K,$$

(5)

where $\rho$ is the distance between electron and hole, $\omega_0$ is the background dielectric constant, $m_0$ is the free electron mass, $\omega_0$ is the exciton resonance frequency, $K$ is the exciton center of mass momentum, and $p_{\text{ex}}^e,f$ are the interband momentum matrix elements for the auxiliary and final states.

To calculate the scattering rate we consider the wave function of the localized exciton $\propto e^{-\rho/a_0 - R^2/(2\ell^2)}$, where $a_0$ and $\ell$ are the exciton Bohr radius and the localization length, and $R$ is the exciton center of mass coordinate. Then, under the assumption of low temperatures, $k_B T \ll g_h \mu_B B_{\text{res}}$ we obtain the hole intervalley scattering rate (see supplementary material)

$$\frac{1}{\tau_{\text{res}}} = \frac{\pi m e^2}{4\hbar^3} \Gamma_0 \eta_0 \xi E_{\text{loc}} \Xi^2 (B - B_{\text{res}}) \theta(B - B_{\text{res}}) \exp \left( -\frac{M g_h \mu_B (B - B_{\text{res}})}{m E_{\text{loc}}} \right),$$

(6)

where $E_{\text{loc}} = h^2/(m\ell^2)$ is the exciton localization energy with $m$ being the exciton mass, $k = \sqrt{2\hbar \omega_0}/c$ is the light wave vector, and $\Gamma_0 = 2\pi k^2 e^2 [\xi^2 \varphi(0)^2] / (h^2 \omega_0^2 m_0^3)$ are the free exciton radiative decay rates in the auxiliary and final states with $\varphi(0) = \sqrt{2/\pi a_0}$ being the wave function of the relative electron hole motion at $\rho = 0$. We note...
that a spread of exciton localization energies in the sample does not affect the position of the resonance.

One can see that the hole intervalley scattering rate vanishes at magnetic fields below $B_{\text{res}}$, as there are no phonons of the required energy, which is described by the Heaviside step function $\theta(B - B_{\text{res}})$. Just above $B_{\text{res}}$ the scattering rate grows linearly with increase of $B - B_{\text{res}}$ because of the increase of the electron-phonon interaction matrix element. However at high magnetic fields the phonons with large wave vectors are needed, so the exciton-phonon matrix element decreases exponentially because of the exciton localization. As a result, the hole intervalley scattering rate has a narrow maximum at $B \approx B_{\text{res}}$ with the width of the order of $mE_{\text{loc}}/(\hbar g_\text{\mu B} M)$. In the maximum it reaches

$$\frac{1}{\tau_{\text{res}}} = \frac{\pi m^2 M^2 \Upsilon^2 \Gamma_0^f E_{\text{loc}}^2 \Xi^2}{4\hbar^2 \rho k^2 (E_a - E_g)^2 g_\text{\mu B} B_{\text{res}}}.$$  \hspace{1cm} (7)

Substitution of the material parameters yields (see supplementary material) $\tau_{\text{res}} = 12$ ns and 20 ns for H-type and R-type HSs, which agrees with the timescales observed for ILE PL polarization saturation [62].

4. Discussion and conclusion

The resonant increase of the PL polarization in the same magnetic field for H-type and R-type HSs unambiguously reveals a valley-magnetophonon resonance.

The dominant microscopic mechanism of the hole valley-magnetophonon resonance is found to be the scattering with a chiral TA phonon of MoSe$_2$ ML between the excitonic states mixed by the long-range exchange interaction. Noteworthy it has a few solid advantages: (a) it does not require spin-dependent electron–phonon interaction, (b) it profits from the large density of chiral phonon states, (c) it is free of van Vleck cancellation, (d) the long-range exchange interaction is enhanced by exciton localization, and (e) optical selection rules for excitons in H$_{\text{HH}}$ and R$_{\text{HH}}$ atomic registries exactly match the requirements for the exchange interaction. The unique observation of the valley-magnetophonon resonance in TMDC HSs is made possible by the strong spin splittings of the bands.

The weaker resonances observed about 3.8 T below and above the main resonance in both HSs may be related to two-phonon processes involving an additional zone-center phonon with an energy of about 2 meV. This might be an interlayer phonon [75], such as the interlayer shear mode [76, 87], which is observable in the samples used in our study (see supplementary material). The tiny splitting of the main resonance in figure 1(c) of the order of 50 mT is probably related to the small spread of $g$ factors between the localization sites. We note that doping and transverse electric fields can be used to tune the exciton oscillator strength. This would change the electron hole long-range exchange interaction, see equation (5), and thus the hole intervalley scattering rate. This could potentially be used to identify this scattering mechanism in other experiments.

For delocalized ILEs, the same mechanism would give a step-like enhancement of the PL polarization and decrease of the ILE lifetime at fields above $B_{\text{res}}$, provided the spin relaxation time exceeds $\tau_{\text{res}}$. Thus, the valley-magnetophonon resonance could be observed also in measurements of ILE diffusion despite the fact that the contribution from the valley-magnetophonon resonance to the momentum relaxation is expected to be much smaller than that from scattering by static disorder.

In summary, we have observed a hole valley-magnetophonon resonance of localized ILEs in both H-type and R-type MoSe$_2$/WSe$_2$ HSs at a magnetic field of 24.2 T. It leads to a resonant enhancement of the PL DOP under nonresonant excitation at low temperatures. The hole spin-flip intervalley scattering involves a chiral TA phonon originating from the MoSe$_2$ ML and long-range exchange interaction. The valley-magnetophonon resonance is important for both the transport properties in moiré HSs and optical manipulation of the valley degree of freedom of charge carriers.

5. Methods

5.1. Sample preparation

Our HSs were fabricated by means of a deterministic transfer process [88] using bulk crystals supplied by HQ graphene. MLs of the constituent materials are prepared on an intermediate polydimethylsiloxane substrate and reliably identified based on their absorption contrast in microscopy. They are subsequently stacked on top of each other on a silicon substrate covered with a silicon oxide layer. In order to achieve crystallographic alignment, we pre-select ML flakes which have at least one very long (>30 µm) straight edge, which indicates that the edge is along a crystallographic high-symmetry direction. Then the well-cleaved edges of the constituent layers are optically aligned parallel to each other during the transfer to reach well-defined interlayer twist angles close to zero or 60 degrees. While this process does not allow us to specifically achieve H-type or R-type alignment, this was identified a posteriori based on the effective $g$ factors and emission energies of ILEs in the samples, see supplementary material. Further details are published elsewhere [62].

5.2. Optical spectroscopy

Low-temperature PL measurements in high magnetic fields were performed at the HFML facility in
Nijmegen. The sample was placed on a x−y−z piezo-electric 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. A diode laser (emission wavelength 640 nm) was used for excitation. The laser light was linearly polarized and 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. The PL was then coupled into a grating spectrometer, where it was detected using a CCD sensor. For field sweeps, the magnetic field was ramped continuously from 0 T up to 30 T (for technical reasons, the field sweep for the R-type HS was limited to 29 T), and spectra for a fixed detection helicity were recorded at fixed time intervals. At the maximum field, the detection helicity was flipped and the field was ramped down continuously to 0 T, so that spectra for the other helicity could be recorded.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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