Anomalous rotation of the linearly polarized emission of bright excitons in strained WSe₂ monolayers under high magnetic fields

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Linearly polarized microphotoluminescence (μ -PL) measurements of strained WSe₂ monolayers in out-of-plane high magnetic fields are presented. At low temperature, a splitting of the bright exciton emission into two exciton components is observed, which is attributed to an in-plane uniaxial strain based on the full polarization dependence of the photoluminescence spectrum. High magnetic field measurements directly reveal a distinct evolution of the linear polarization and allows us to extract the valley coherence time constants (T_{s2}^*) for both exciton components. For the high-energy transition of the exciton, the valley coherence time $\simeq 0.45$ ps, closely matching T_{s2}^* of an unstrained monolayer ($\simeq 0.34$ ps). For the low-energy exciton, however, T_{s2}^* is four times larger, $\simeq 1.97$ ps. This valley coherence time observed here may be explored for future valleytronic applications.

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I. INTRODUCTION

Two-dimensional transition-metal dichalcogenides (TMDCs), such as MoS₂, MoSe₂, WS₂, and WSe₂, have recently emerged as a promising class of semiconducting materials to investigate new physical effects related to strong Coulomb mediated electron-hole (e - h) interactions [1–3]. Due to a direct band gap located at two degenerate valleys at the corners of the hexagonal Brillouin zone (K^+ and K^-) and a strong spin-orbit interaction [4,5] in monolayers, bound e - h pairs (excitons) in these materials possess unique optical selection rules [6–8]. This makes it possible to individually address them by using circular polarization in both excitation and detection (valley polarization) [9,10]. Additionally, excitons can be prepared in a coherent superposition of the two K^+ and K^- valleys using linearly polarized optical excitation, thereby generating so-called *valley coherence* [11], which results in linearly polarized optical emission [11,12]. Recently, experimental evidence for control of the valley coherence where the linearly polarized optical emission is rotated with respect to the linear excitation polarization using external magnetic fields has been reported in monolayer WS₂ and WSe₂ [13–15]. These experimental results represent an important step towards full control of valley states in these materials and, in addition, serve as a playground for quantum optical experiments, in close analogy to the electron spin in coherent spintronics [16]. However, fundamental questions with respect to the nature of the valley decoherence mechanisms remain open. Possible reasons include intervalley scattering of carriers mediated by the Coulomb interaction

and/or interaction with phonons in between K^+ and K^- valleys or the K to Q valley of the Brillouin zone. In addition, the enhanced e - h exchange interaction that effectively couples the exciton center-of-mass motion with its valley pseudospin can also cause valley decoherence. Here, we are interested in the effect of strain on valley decoherence in monolayer TMDCs.

In low-dimensional semiconductors, such as quantum wells, wires, and dots, strain effects are known to influence profoundly the energy levels, the spatial extent of the wave functions, as well as the interaction between carriers, excitons, and phonons [17,18]. Recent experiments in molybdenum and tungsten-based monolayer TMDCs have revealed new insights into exciton-phonon coupling under uniaxial tensile strain using microphotoluminescence (μ -PL) measurements at room temperature [19]. In particular, the PL linewidth of the neutral exciton has been observed to decrease with an increasing amount of strain, indicating a weakening of the exciton-phonon coupling in WSe₂ monolayers. Moreover, the energy separation between the indirect and direct gaps increases due to suppression of the phonon-induced intervalley (K - Q) scattering [20]. At cryogenic temperatures, when the exciton linewidth is smaller due to a reduced phonon-assisted broadening, a splitting of the bright exciton emission into two exciton components, each with peculiar polarization properties, is observed [21]. In this respect, probing linear polarization of neutral exciton emission in strained monolayer TMDCs under high magnetic fields can provide important information regarding the mechanism of valley decoherence.

We present polarized microphotoluminescence measurements on strained WSe₂ monolayers at 4.2 K in a linearly polarized basis (both in excitation and detection) in perpendicular magnetic fields up to 25 T. Control measurements are performed on unstrained monolayers. We investigate the

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degree of linear polarization of the neutral exciton emission and its field-induced rotation angle, since these are the key parameters towards the understanding of valley decoherence. At zero magnetic field, we show that the two exciton components, separated by a few meV, have optical properties that dependent strongly on linear excitation polarization. Interestingly, measurements in high magnetic fields show that the two exciton components exhibit a different rotation angle and a distinct decrease of linear polarization with field. We analyze our data using a simple four-level model system and extract a valley coherence time constant of $T_{s2}^* \simeq 0.45$ ps for the high-energy exciton, which is of the same order of magnitude as the value for an unstrained WSe_2 monolayer sample ($\simeq 0.34$ ps). Surprisingly, the valley coherence time constant for the low-energy exciton state is four times larger ($\simeq 1.97$ ps). Our results demonstrate that strain can be used to alter and optimize the optical properties in monolayer TMDCs, including complex effects such as valley coherence.

II. EXPERIMENT

WSe_2 monolayer flakes were prepared by mechanical exfoliation from bulk crystals and transferred onto 285-nm SiO_2/Si substrates. The monolayer region was confirmed by concomitant high-resolution polarized-resolved μ -Raman, μ -PL, and μ -PL excitation (μ -PLE) measurements at liquid-He temperature [21]. The strain in our monolayer samples is unintentional and is likely created during the exfoliation process. Mechanical stretching in one direction leads to uniaxial strain, which is enhanced during cooling the monolayer sample, because of the different thermal expansion coefficients of the lattices of the TMDC monolayer and the underlying SiO_2 substrate [21]. The distinction between strained and unstrained monolayer samples was made by using μ -Raman spectroscopy measurements (see Ref. [21]). Figure 1(a) illustrates the schematics of the optical setup in a high magnetic field. For all measurements, the monolayer samples were placed inside a liquid-He bath cryostat and cooled using helium exchange gas. The measurements were performed in the backscattering configuration in a Florida-Bitter magnet generating magnetic fields up to 30 T, oriented perpendicular to the plane of the monolayer (Faraday geometry). A continuous-wave laser, emitting at 640 nm (1.93 eV), was used for excitation via a microscope objective with a numerical aperture $\text{NA} = 0.55$ and $40\times$ magnification, resulting in a laser spot of approximately $1\ \mu\text{m}$ in diameter. The excitation energy was chosen to be energetically sufficiently close to the excitonic resonance, to observe a finite degree of circular and linear polarization at 0 T, but far enough to avoid resonant Raman scattering peaks.

μ -PL spectra in zero magnetic field were recorded using a triple-grating spectrometer in subtractive mode (0.5 m length, 1800 grooves/mm grating) equipped with a liquid-nitrogen-cooled CCD camera. Linearly polarized μ -PL measurements in high magnetic fields were performed using a single-grating spectrometer (0.3 m length) with a 1200 grooves/mm grating. The linearly polarized light for excitation and detection was implemented by using two pairs of optical elements, consisting of a half-wave plate and a linear polarizer, allowing for an independent control of polarization. The field-induced

FIG. 1. (a) Schematic illustration of the experiment. The laser polarization (labeled as “exc.”) is linear along the x axis, which allows for the generation of excitons in a superposition of K^+ - and K^- -valley states. The linear polarization of the PL is measured using a half-wave plate and linear polarizer. θ is the angle between the polarizations of the excitation and detection. We use $\theta = 0^\circ$ and 90° to denote the perpendicular and parallel polarizations, respectively. The plane of the monolayer was oriented perpendicularly to the direction of the magnetic field (Faraday geometry). (b) Top: Illustration of the uniaxial tensile strain in a WSe_2 monolayer. Bottom: Uniaxial strain causes an in-plane anisotropy splitting (Δ_0) between the exciton branches in the light cone [7,24]. (c) μ -PL spectrum (symbols) of a strained WSe_2 monolayer on a SiO_2/Si substrate measured at $T = 4.2$ K, including the simulated spectrum (solid line). The double structure of the bright A-exciton $1s$ ground state is shown by bi-Lorentzian fits (dashed lines). The neutral exciton peaks along the x and y directions are labeled as X_X and X_Y , respectively.

Faraday rotation of both the excitation and emitted light in the microscope objective was compensated by appropriate rotation of the linear polarization.

III. RESULTS

A typical μ -PL spectrum of a strained WSe_2 monolayer sample at 4.2 K and at zero magnetic field is presented in Fig. 1(c). The highest-energy peak around 1750 meV labeled exciton corresponds to the bright neutral A-exciton transition. The feature around 1718 meV labeled trion corresponds to the negatively charged exciton. These peaks assignments are in good agreement with previously published data [11,13,22,23]. However, due to the presence of strain in our samples, both neutral exciton and trion peaks are split. To further clarify the exciton fine structure, we include in Fig. 1(c) fitted spectra obtained using bi-Lorentzian functions. In the case of the trion peak, the observed fine structure corresponds to the optically bright, negatively charged excitons—inter- and

intravalley trions. This arises from the strong e - h exchange interaction, which recently has been predicted [7,24] and observed [23,25]. In the neutral exciton case, the observed split-exciton peak arises from a combined effect of strain and electron-hole exchange [see Fig. 1(b)], where the peaks correspond to anisotropic excitons. We label these features X_X and X_Y , to refer to neutral excitons along the x and y directions, respectively [21]. The corresponding energy splitting Δ_0 of about 4 meV is consistent with the extracted mechanical strain values and is in good agreement with theoretical predictions [24]. We note that the neutral exciton PL linewidth in our strained monolayer samples amounts to ≈ 9 meV, smaller than in unstrained samples, whose linewidth is about 12 meV. This is consistent with the effect of the weakening of the exciton-phonon coupling with increasing the uniaxial strain [19], as well as the increase of the energy separation between the indirect (Q -valley) and direct (K -valley) gaps [20], and thus provides an additional confirmation of the presence of uniaxial strain in our WSe₂ monolayers.

A. Linear polarization at $B = 0$ T

To gain more information about the anisotropy of the neutral exciton emission, we first characterize our sample at zero magnetic field. Figure 2(a) shows the linearly polarized PL spectra for horizontally (top panel) and vertically (bottom panel) polarized excitation of a strained WSe₂ monolayer corresponding to excitation along the y and x directions. The PL emission is analyzed into its two linearly polarized components (vertical and horizontal). Three main observations are immediately apparent from the data. First, the overall neutral exciton emission partially preserves the linear polarization (valley coherence), in contrast to the trion emission, which is unpolarized (no valley coherence) in both configurations (horizontally and vertically polarized excitation) [11]. Second, the PL intensity of the neutral exciton strongly depends on the polarization orientation in excitation: In horizontally polarized excitation (labeled as Y), the horizontally polarized emission is almost two times stronger than the vertically polarized one; in vertically polarized excitation (labeled as X), the vertically polarized emission is only slightly stronger than the horizontally polarized one. Third, the overall PL emission intensity is distributed differently over the X_X and X_Y excitons, for excitation along the x and y directions. The relevant quantity in this experiment is the degree of linear polarization of the emitted light defined as $DLP = (I_{\parallel} - I_{\perp}) / (I_{\parallel} + I_{\perp})$, where $I_{\parallel}$ ($I_{\perp}$) denotes the integrated intensity of the PL component that is parallel (perpendicular) to the excitation. Having established that the neutral exciton PL peak consists of two Lorentzian line shapes [see Fig. 2(a)], we now can extract the DLP for each anisotropic exciton in horizontally (vertically) polarized excitation. In horizontally polarized excitation, the DLP amounts to about 13% for the X_X and 39% for the X_Y exciton. In vertically polarized excitation, the DLP $\simeq 24\%$ for the X_X and 21% for the X_Y exciton. Therefore, depending on the linear polarization in excitation, the degree of linear polarization is stronger for either the X_X or X_Y exciton. In contrast to unstrained WSe₂ monolayers, where the degree of linear polarization is independent of the crystal orientation and exactly follows the polarization of the optical excitation

FIG. 2. (a) High-resolution linearly polarized PL spectra for horizontally (top panel) and vertically (bottom panel) polarized excitation. Green and brown symbols represent vertically and horizontally polarized detection. Dashed and solid spectra show bi-Lorentzian fits to the doublet structure of the neutral exciton peak. The solid spectra indicate the fits in cross polarization configuration. Solid lines show the total fitted curves. (b) PL spectra as a function of the linear polarization detection angle. The orientation of the excitation (brown arrows) and detection (red arrows) linear polarization are indicated for the co- and cross-polarization configuration. (c) Polar plots of the extracted PL intensity for X_X and X_Y excitons at $B = 0$ T (symbols: measurement data; solid lines: sinusoidal function fit). $I_{\parallel}$ ($I_{\perp}$) denotes the integrated PL intensity that is parallel (perpendicular) to the excitation.

(see Jones *et al.* ref.[11]), in strained monolayers the DLP is dependent on linear polarization in excitation.

Collectively, these experimental results at zero field provide evidence that the polarization of the bright exciton PL emission depends on the in-plane (xy) anisotropy. At the same time, however, for the moderate strain strength ($\approx 0.2\%$) in our samples, both PL peaks show a finite degree of circular polarization [21]. This suggests that these two anisotropic excitons, X_X and X_Y , can nevertheless preserve clear chiral optical selection rules at the $K^{\pm}$ points of the Brillouin zone. A similar observation has been also reported for the monolayer (ML) MoS₂ free-exciton emission under a controllable applied uniaxial tensile strain [26]. Therefore, the observed linear polarization in detection is a combination of both anisotropic and valley coherence effects. Due to the small mechanical strain in our samples, we presume that the linear polarization is mainly dominated by valley coherence which, in this case, is a result of the coherent superposition of valley states in each of the individual X_X and X_Y exciton levels.

FIG. 3. (a) 2D false-color plots presenting bright A-exciton PL spectra for several magnetic fields using vertical linear polarization (x polarized) in excitation and different angles θ of linear polarization in detection. The copolarized configurations at $B = 0$ T are indicated by horizontal white dashed lines on each plot. The green and brown dashed lines are a guide to the eye and allow one to follow the evolution of the PL linear polarization angle of the two excitons (X_X and X_Y) in the magnetic field. (b) and (c) Polar plots of the extracted PL intensity (symbols) for each exciton at selected magnetic fields ($B = 0, 10$, and 20 T). Solid lines are sinusoidal function fits to the data.

To further characterize the linear polarization properties of X_X and X_Y , we measured μ -PL spectra with a fixed linear polarization direction in the excitation (vertical) and detection polarization angles rotating through 180° . The raw data are presented in Fig. 2(b) as a nonpolar two-dimensional (2D) false-color plot. The exciton PL intensity changes significantly as a function of the linear polarization detection angle. Because of the small energy separation between the two exciton PL peaks, it is hard to distinguish them in this figure. However, in Fig. 2(c) we present the polar plot of the extracted emission intensity of both excitons, which clearly depends on the detection polarization angle. In agreement with previous reports [11,15], the variation of the PL intensity with the detection angle shows a characteristic figure of a dipole shape dependence. The data can be well described using $I(\theta) = I_0 + I_x \cos^2(\theta + \theta_0)$, from which we extract the angle between the maximum PL intensity for each exciton. Therefore, we can observe that the principal polarization axes of the excitons are rotated by about few degrees with respect to each other.

B. Linear polarization in high magnetic fields

We now turn to the linearly polarized μ -PL in high magnetic fields. These measurements have been performed under vertically polarized optical excitation. Figure 3(a) displays typical 2D false-color plots for different values of B , which show the PL spectra as a function of the linear polarization detection angle. The two main effects of B on the linear polarization of the two excitons X_X and X_Y are visible in the experimental data. First, the linearly polarized optical emission angle of the low- and high-energy exciton shifts with increasing B towards a higher angle [see the maxima of the brown and green dashed lines with respect to white horizontal lines in Fig. 3(a)]. This effect is more pronounced for the X_X than the X_Y exciton. Second, the degree of linear polarization is strong at low and zero B , but gradually reduces

with increasing magnetic field, again with a behavior that is different for the X_X and X_Y excitons.

These effects are clearly visible in Figs. 3(b) and 3(c) in which the same results are plotted in polar coordinates: At $B = 0$ T, the two excitons have almost the same linear polarization angle, which also coincides with the polarization direction of the excitation laser [brown arrows in Figs. 3(b) and 3(c)]. However, at $B = 20$ T the rotation of the principal polarization axis for the X_X exciton is almost two times larger than for the X_Y exciton [see pink and blue symbols in Figs. 3(b) and 3(c)]. Moreover, the decrease in the DLP with increasing B (decoherence effect [13,15]), which is different for the X_X and X_Y exciton, is clearly observed in polar coordinates by analyzing the evolution of the polarization pattern shape. At $B = 20$ T, where the two excitons still have a finite degree of linear polarization, the X_X exciton pattern is close to a dipole, whereas the X_Y pattern clearly deviates from a dipole shape.

To obtain more quantitative information about the dependence of the rotation angle on B for both exciton components, we fitted their integrated PL intensities with a sinusoidal function at each magnetic field, as shown by the solid lines in Figs. 3(b) and 3(c). The results of the analysis are presented in Fig. 4(a). For the X_X exciton component, we extracted a rotation angle value of about 40° , whereas for the X_Y about 23° at the maximum field of $B = 25$ T used in our experiment. The almost twice difference in rotation angle of the two exciton components is surprising, reflecting a more complex valley decoherence mechanism. We notice that the rotation angle of the X_X exciton obtained here is comparable to the value of the neutral exciton reported in WS_2 monolayers [14]. In order to verify the rotation angle in WSe_2 monolayers without strain, we also performed control measurements under similar conditions. The complete data set is given in the Appendix, whereas the extracted rotation angle values are shown in Fig. 4(a) (purple symbols) for comparison. Indeed, in our unstrained WSe_2 monolayer sample we observe a very similar rotation angle of

FIG. 4. (a) Rotation angle and (b) degree of linear polarization (DLP) for strained (X_X , X_Y) and unstrained (X) neutral excitons in WSe₂ monolayers at different magnetic fields (symbols). The solid lines are fits to the experimental results by $\theta_{X(Y)} = \arctan(\Omega_X T_{s2}^*)/2$. (c) Low-temperature μ -PL spectra (open symbols) of strained WSe₂ at $B = 0$ and 21 T. σ^- polarized light was used for excitation, with detection in σ^+ (red) and σ^- (blue) polarization. Solid lines show the Lorentzian fits to the spectra. (d) Transition energies vs magnetic field. The upper states (X_Y^{K-} and X_X^{K-}) are σ^- polarized (blue) and the lower states (X_Y^{K+} and X_X^{K+}) σ^+ polarized (red). (e) Schematic diagram of magnetic field valley decoherence process in strained WSe₂ monolayers.

39° at $B = 25$ T. Therefore, the smaller rotation angle value for X_Y suggests a more complex picture of valley decoherence in strained monolayer TMDCs. We also notice that in this sample, we can observe a saturation of rotation angle above 22 T, previously not seen in this material, which might be due to the low magnetic fields employed in those experiments ($B \leq 9$ T) [13,15]. Interestingly, in the strained monolayer sample, the linear polarization rotation angle of the X_X exciton saturates at a much lower field (≈ 15 T), whereas for the X_Y component we see no saturation effect. This demonstrates that

the uniaxial strain in monolayer TMDCs can be used to vary the magnetic field value at which the saturation of rotation angle emerges in the linearly polarized exciton emission.

In Fig. 4(b) we show the extracted values for the degree of linear polarization for both exciton components, along with the data for the neutral exciton (X) of the unstrained monolayer. We observe a decrease of the DLP in both cases with increasing magnetic field: from about 22% at $B = 0$ T to 13% for the X_X and about 5% for the X_Y at $B = 25$ T. We notice, in particular, a faster decrease of the DLP of the X_Y component, which can be no longer resolved at fields larger than $B = 22.5$ T. Concerning the neutral exciton of the unstrained monolayer, the DLP decreases more rapidly than that of the X_X exciton but slower than that of the X_Y . Such an overall distinct magnetic-field-induced behavior of the degree of linear polarization is particularly remarkable given the fact that both states have initially equal values at zero field.

IV. DISCUSSION

In monolayer TMDCs, applying an out-of-plane magnetic field lifts the valley degeneracy, resulting in a change of oscillation frequency of the coherent superposition of valley states by $\Delta E = \hbar\Omega_B = g\mu_B B$, where g is the exciton g -factor and $\mu_B = 0.058$ meV/T is the Bohr magneton. According to Jones and Xu *et al.* [11], the exciton pseudospin can be illustrated on the equator of a Bloch sphere (in close analogy with the formalism used for spin). Thus, the evolution in time of the pseudospin after an initial laser light polarization along the x direction is given by $|X\rangle = 1/\sqrt{2}(|K^+\rangle e^{-i\Omega_B t/2} + |K^-\rangle e^{i\Omega_B t/2})$. As a result, after a certain time and at a finite magnetic field, the exciton pseudospin will have evolved to another position on the Bloch sphere [15]. This corresponds to a rotation of the exciton PL linear polarization considering that ΔE has a nonzero value. The extracted rotation angle as a function of magnetic field can be analyzed according to $\theta = \arctan(\Omega_B T_{s2}^*)/2$. Here, T_{s2}^* is the fitted coherence time and can be expressed as $1/T_{s2}^* = 1/\tau + 1/T_{s2}$, where τ is the exciton lifetime and T_{s2} is the pure dephasing time, respectively.

In strained monolayer TMDCs, however, the picture becomes more complex since each of the two exciton components, being separated by Δ_0 at zero magnetic field, split up upon the application of an external magnetic field. Therefore, in this case we have four distinct PL lines at high fields (X_X^{K+} , X_Y^{K+} , X_X^{K-} , and X_Y^{K-}) as can be seen in the circularly polarized resolved μ -PL experiment shown in Figs. 4(c) and 4(d) and schematically illustrated in Fig. 4(e). The magnetic-field-induced splitting of the two exciton components ΔE_X and ΔE_Y is different and, in addition, is twice smaller than ΔE in unstrained monolayers. Therefore, we can expand the model derived by Wang *et al.* [15] to our strained ML considering a four-level model system (see Ref. [27] for more details). Since the rotation angle is directly proportional to the exciton g -factor (i.e., valley Zeeman splitting) and T_{s2}^* parameter, we can define $\theta_X = \arctan(\Omega_X T_{s2}^*)/2$ and $\theta_Y = \arctan(\Omega_Y T_{s2}^*)/2$ for each of two exciton components, where $\Omega_{X(Y)} = \Delta E_{X(Y)}/\hbar = g_{X(Y)}\mu_B B/\hbar$. The fits yield T_{s2}^* values of ≈ 0.45 and 1.97 ps using the exciton g -factors of $g_Y \approx -1.8 \pm 0.05$ and $g_X \approx -2.6 \pm 0.05$, for X_Y and X_X , extracted

FIG. 5. (a) μ -PL spectrum of an unstrained WSe₂ monolayer on a SiO₂/Si substrate measured at $T = 4.2$ K and $B = 0$ T. (b) Polar plots of extracted PL intensity for a neutral exciton in an unstrained WSe₂ monolayer flake at selected magnetic fields of $B = 0, 5, 10, 15, 20$, and 25 T. Solid lines are sinusoidal function fits to the data.

from circularly polarized PL measurements in high magnetic fields [21]. The data for the unstrained WSe₂ monolayer are presented in Fig. 4(a) for direct comparison. Similarly, we extract a value of $T_{s2}^* \simeq 0.34$ ps using an exciton g -factor of $\simeq -4.02 \pm 0.05$. It is interesting to note that while the T_{s2}^* parameter is smaller in the unstrained sample, the higher values found in the strained sample are related to the smaller exciton g -factor values [21].

In the literature, the intrinsic exciton radiative lifetime (τ) for monolayer WSe₂ at low temperature (4.2 K) is reported to be from 0.2 to 4 ps [28–30]. Assuming an exciton lifetime τ of 2 ps [30] in our work, we can estimate the valley decoherence time T_{s2} for each of the two exciton resonances. The error here, given by both the data analysis uncertainty and the sample-to-sample variation, is less than 2 ps and leads to an uncertainty of the estimated valley decoherence time values of about ± 0.08 ps. For X_Y we obtain a value

of $T_{s2} \simeq 0.58 \pm 0.08$ ps, which is of the same order of magnitude as our derived value for the unstrained sample (0.41 ± 0.08 ps) and is in very good agreement with recent theoretical predictions and experimental data [15,31]. Interestingly, the valley decoherence time estimated for the X_X exciton is 131 ± 0.08 ps, which is two orders of magnitude larger than for the high-energy exciton. This is surprising since the rotation angle for the low-energy exciton is only twice larger than for the high-energy exciton and the extracted exciton g -factors cannot explain this large difference in their decoherence times. However, this effect might be related to the longer exciton lifetime for the higher-energy state than the low-energy state, although the exact reason requires additional investigation. Recent experiments in uniaxially strained WSe₂ monolayers have revealed that the exciton linewidth decreases with increasing mechanical strain [20]. This has been explained on the basis of an increased energy separation between the indirect (K - Q) and direct (K - K) gaps, suppressing the phonon-induced intervalley scattering. As a result, it is expected that the exciton spin, valley, and coherence lifetimes will be longer in strained WSe₂ MLs. However, at this moment it is not clear why the exciton lifetime for one state should be longer than for the other one. Nevertheless, this result provides information about the valley decoherence mechanism, and furthermore it suggests interesting approaches to manipulate the valley coherence at longer times in atomically thin TMDCs.

Finally, we will discuss the degree of linear polarization in magnetic fields. Figure 4(b) shows the simulated linear polarization degree of the experimental data using the same model as in Fig. 4(a). As expected, for both unstrained (purple) and strained (green and brown) MLs, the DLP strongly decreases with increasing magnetic field. This drop in DLP originates from the dephase induced during the exciton lifetime [13,32]. The decrease in DLP of the low-energy exciton is lower compared to the high-energy exciton, which might be attributed to the longer exciton lifetime for the higher-energy state, consistent with the observation of the larger decoherence time discussed above. However, future time-resolved PL measurements which resolve the dynamics of each exciton should provide more definitive conclusions on this matter.

V. CONCLUSION

In conclusion, we have measured linearly polarized PL of bright excitons in strained WSe₂ monolayers in an out-of-plane magnetic field up to 25 T. We find that the PL linear polarization rotation angle increases and its degree of polarization is reduced with increasing magnetic field strength for both exciton components. Interestingly, the rotation angle increase for the low-energy exciton is almost twice larger than for the high-energy exciton. In contrast, the degree of polarization has a stronger decrease for the high-energy exciton. We determine the valley coherence time constant of $T_{s2}^* \simeq 0.45$ and $\simeq 1.97$ ps for the high- and low-energy exciton, respectively. Our results contribute to a better understanding of how strain changes the excitonic properties, provide crucial information about exciton valley coherence, and open different ways to control this effect in strained monolayer TMDCs.

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APPENDIX: ADDITIONAL DATA FROM UNSTRAINED WSe₂ MONOLAYER FLAKE

As a control experiment, we measured a WSe₂ monolayer flake without the presence of uniaxial strain (as deter-

mined from polarized microphotoluminescence and Raman measurements—see Ref. [21] including the Supplemental Material therein). The μ -PL spectrum of an unstrained WSe₂ monolayer on a SiO₂/Si substrate measured at $T = 4.2$ K and $B = 0$ T is shown in Fig. 5(a), where we also highlight the PL peak associated with the neutral exciton (X), charged exciton (T), and localized exciton (L). Following the approach we used in the main text, we systematically map the linearly polarized PL of this sample as a function of magnetic field. In Fig. 5(b) we show the extracted PL intensity for the neutral exciton in polar coordinates. The analysis of these data is discussed in the main text. These results confirm that an out-of-plane magnetic field leads to a rotation of polarization angle together with a decrease of the degree of the linear polarization, previously observed in WS₂ monolayers [14].

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