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Spin depolarization dynamics of WSe_2 bilayer*

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In this work, the spin dynamics of a centrosymmetric WSe_2 bilayer has been investigated by the two-color time-resolved Kerr rotation together with helicity-resolved transient reflectance techniques. Two depolarization processes associated with the direct transition are discovered at a low temperature of 10 K, with the characteristic decaying time of ~3.8 ps and ~20 ps, respectively. The short decay time of ~3.8 ps is suggested to be the exciton spin lifetime of the WSe_2 bilayer, which is limited by the short exciton lifetime of the WSe_2 bilayer and the rapid intervalley electron–hole exchange interaction between K^+ and K^- valley in the same layer as that of monolayer. The long decay time of ~20 ps is suggested to be the spin lifetime of photo-excited electrons, whose spin relaxation is governed by the rapid intervalley scattering from the K valley to the global minimum Σ valley and the subsequent interlayer charge transfer in WSe_2 bilayer. Our experimental results prove the existence of the spin-polarized excitons and carriers even in centrosymmetric transition metal dichalcogenides (TMDCs) bilayers, suggesting their potential valleytronic and spintronic device applications.

Keywords: spin depolarization dynamics, WSe_2 bilayer, time-resolved Kerr rotation

PACS: 72.25.Rb, 31.70.Hq, 78.55.–m **DOI:** 10.1088/1674-1056/27/5/057202

1. Introduction

Transition metal dichalcogenides (TMDCs) thin films, such as MoS_2, $MoSe_2$, WS_2, and WSe_2, have drawn considerable attention for their promising applications in novel optoelectronics and valleytronics as well as quantum manipulation.[1,2] WSe_2 is a representative material in the TMDCs family characterized by its high photoluminescence (PL) quantum yield and strong spin–orbit coupling (SOC) induced spin splitting.[3–5] Up to now, WSe_2 monolayer has been widely investigated.[3,6–12] Thanks to the broken inversion symmetry and strong SOC, a valley-dependent optical selection rule is allowed,[13,14] so that the circular polarization of the absorbed or emitted photons can be easily related to the selective carrier excitation in the corresponding non-equivalent K^+ and K^- valleys. Different valley polarization degree by varying temperature and excitation wavelength for different TMDCs thus has been investigated extensively by using the steady-state polarization-resolved PL technique.[6,8,9,15–20] Electron–hole Coulomb exchange interaction between K^+ and K^- valley has been regarded as the main scattering mechanism for the valley depolarization in monolayer TMDCs.[21–24]

Consisting of two monolayers of WSe_2 with a layer degree of freedom added, bilayer WSe_2 exhibits rich optoelectronic properties different from its monolayer counterpart. As the upper layer is 180° rotated in-plane with respect to the lower layer, crystal inversion symmetry is recovered.[25–28] Thus it is expected that the circular polarization of excitons arising from the valley-dependent optical selection rule is absent in centrosymmetric bilayer WSe_2. However, in contrast to this expectation, polarized PL emission associated with direct transition has shown a rather high polarization degree as reported for bilayer WSe_2,[26,29] WS_2[30] and electric-field induced symmetry breaking MoS_2.[31] These unusual observations have stimulated further theoretical and experimental studies for the intrinsic origination of the observed circular polarization in centrosymmetric bilayer TMDCs materials.[25,32–34] Though the crystal inversion symmetry is recovered in bilayer WSe_2, strong SOC strength in each monolayer can efficiently suppress the interlayer hopping of carriers, known as the spin–layer locking effect, leading to distinct high circular polarization of PL for WSe_2 bilayer.[29] The intrinsic circular polarization for bilayer TMDCs has also been theoretically investigated with strong SOC as a local effect in monolayer, which can induce spin polarization even for bilayers with global inversion symmetry.[33]

The dynamics of direct and indirect excitons in bilayer WSe_2 has been previously studied,[26] and the depolarization of a direct exciton is reported to be as short as a few picoseconds by time-resolved PL (TRPL) measurement, similar to the measured lifetime of a direct exciton.[26] However, the experimental study of spin dynamics with the time-resolved Kerr rotation (TRKR) technique, which is capable to directly probe the intrinsic valley/spin polarization in TMDCs and overcome the constrain of low time-resolution and short electron–hole

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recombination lifetime in TRPL measurement,[3,7,11,24,35–38] has not yet been performed for bilayer WSe_2, which is essential for understanding the spin- and valley-related physics in interlayer-coupled TMDCs. In this letter, the two-color TRKR technique together with helicity-resolved transient reflectance spectrum has been employed to characterize the spin dynamics in WSe_2 bilayer. Strong direct and indirect transitions, associated with the direct transition in K valley and the indirect transition between holes at K valley and electrons at Σ valley, are observed in the PL spectrum. The spin dynamics associated with the direct transition in WSe_2 bilayer has been characterized by two decay times of ~3.8 ps and ~20 ps at the low temperature of 10 K. Similar results are obtained by the helicity-resolved transient reflectance study. The short decay time of ~3.8 ps and its temperature dependence agree well with the valley lifetime of neutral exciton in the monolayer WSe_2 reported previously, while the long spin lifetime of ~20 ps is discussed to be associated with the electron spins in WSe_2 bilayer. Our results are helpful for understanding the spin properties for interlayer-coupled multilayer TMDCs films.

2. Experiment

The investigated bilayer WSe_2 flakes are fabricated by mechanical exfoliation with adhesive tape from a bulk crystal (2D semiconductors, Inc.) onto 285-nm-SiO_2/Si substrates. The typical optical images of few-layer and bilayer WSe_2 are firstly identified by optical contrast under a microscope, as depicted in Fig. 1(a). Due to AB stacking in WSe_2 bilayer, there exists 180° rotation between layers, combining with a strong SOC effect. The spin, valley, and layer configuration in the first Brillouin zone for WSe_2 bilayer is sketched in Fig. 1(b).[29] The steady-state microscope PL under the excitations of a 532 nm CW laser is collected by a Si CCD combined with Horiba Jobin Yvon iHR550 spectrometer. For the circular-polarization resolved experiment, the PL emission passes through a quarter wave plate and a beam-displacer enabling the left- and right-circularly polarized component of PL response detection at the same time.

The TRKR and helicity-resolved transient reflectance spectrum is taken with a Ti: sapphire laser system (Chameleon Ultra II, Coherent Inc.) which provides a 150 fs pulse with a repetition rate of 80 MHz. The experiments are performed with the optical setup shown in Fig. 1(c). In our two-color pump–probe experiment, the output of the Ti: sapphire laser is directly used as a pump beam, while the probe beam is chosen from a supercontinuum white light source which is generated by a nonlinear photonic crystal fiber under excitation of a femtosecond laser pulse of Ti: sapphire laser. The tunable probe beam is chosen by combining two band-pass filters (TBP01-790 and FF01-747, Semrock, Inc.). The pump and probe beams are focused onto the sample collinearly using an Olympus 50× objective lens. The spatial resolution is about 1.5 μm. The time delay of the probe is controlled by a remote-controlled mechanical stage. The probe beam is linearly polarized, while the pump beam is circularly polarized and cut off by a long-wavelength pass filter (FF01-715/LP, Semrock, Inc.) before being directed into the photodetector. The Kerr rotation signal is collected by a sensitive optical bridge balanced Si photodetector combined with a Stanford lock-in amplifier.

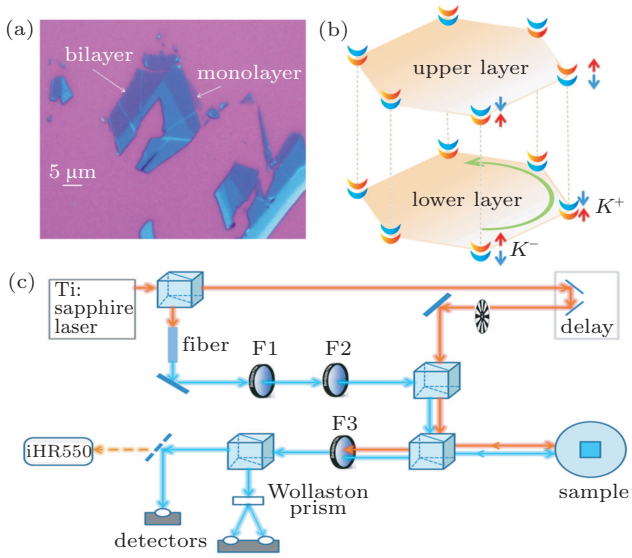


Fig. 1. (color online) (a) Optical microscopic image of WSe_2 flakes. (b) Cartoon of WSe_2 bilayer with its spin, valley, and layer configuration depicted. The upper layer is 180° rotated in-plane with respect to the lower layer. (c) Sketch of the time-resolved Kerr rotation (TRKR) setup. F1, F2, and F3 represent three kinds of optical filters.

3. Results and discussion

The temperature dependent PL response of WSe_2 bilayer is first measured with the spectra shown in Fig. 2(a). The two individual peaks can be easily distinguished when temperature is below 230 K. These two PL peaks will then merge together at higher temperatures. According to the previous works,[5,26,27,29,39–41] the PL peaks at 1.701 eV and 1.566 eV at 10 K are assigned to be associated with the direct (A exciton) and indirect transition, respectively. One can clearly see that the direct PL emission peak is red-shifted with increasing temperature. The temperature variation of the direct transition energy can be perfectly fitted by the Varshni function which describes the band gap decrease with increasing temperature for bulk semiconductors, as shown by the solid line in Fig. 2(b):[42,43] $E(T) = E(0) - \alpha T^2/(T+\beta)$, where $E(0)$ is the PL emission energy at 0 K, α and β are Varshni coefficients. For WSe_2 bilayer, $E(0) = 1.698$ eV, $\alpha = 3.9 \times 10^{-4}$ eV/K, and $\beta = 190$ K are extracted. The peak energy of the indirect PL emission shows rather weak temperature dependence, which is a character of the indirect transition that requires additional absorption or emission of phonons.

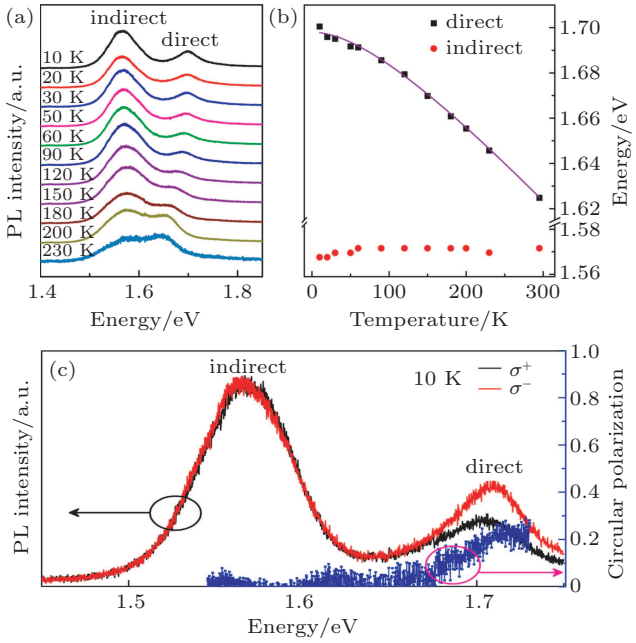


Fig. 2. (color online) (a) The PL spectrum of WSe_2 bilayer measured at different temperature. (b) Temperature dependence of the direct and indirect PL emission peak energy in WSe_2 bilayer. (c) Right (σ^+) and left (σ^-) circularly-polarized components of circular-polarization resolved PL spectra under σ^+-polarized laser excitation (left axis), and the polarization degree of circularly polarized PL of WSe_2 bilayer (right axis).

The polarization-resolved PL measurement is then performed by excitation of circularly-polarized (σ^+ or σ^-) laser with photon energy of 2.33 eV, as shown in Fig. 2(c). Since the intensity difference between σ^+- and σ^--components reflects the population imbalance between spin-up and spin-down excitons, the obvious circular polarization is observed for direct excitons, and the degree of circular polarization is evaluated through $P_c = [I(\sigma^+) - I(\sigma^-)]/[I(\sigma^+) + I(\sigma^-)]$, where $I(\sigma^+)$ and $I(\sigma^-)$ correspond to the PL intensities of σ^+ and σ^- components, respectively.[6,11,25,44] A sizable PL circular polarization degree ($P_c \sim 27\%$) for direct A exciton is obtained, as shown in Fig. 2(c), while there is no PL polarization for the indirect exciton, which is in line with the previous works.[26,29,30] Though the circular polarization of excitons in WSe_2 bilayer should be absent from the point of view of the existence of inversion symmetry, the experimentally observed circular polarization of WSe_2 bilayer has been explained to result from the strong SOC effect, which competes over the interlayer hopping of both electrons and holes, leading to the so-called spin–layer locking effect for which a carrier is localized in either the upper or lower layer depending on its valley and spin state.[29]

The steady PL polarization degree is $P_c = P_0/(1 + 2\tau_r/\tau_s)$, where P_c is the steady PL polarization degree, P_0 is the initial PL polarization degree, τ_r is the exciton lifetime, and τ_s is the spin lifetime of excitons. A reasonable larger P_c is thus expected when τ_s is longer than τ_r. The lifetime of direct exciton for WSe_2 bilayer has been reported to be only $\sim$3 ps at a low temperature of 4 K.[26] The longer spin lifetime of WSe_2 bilayer is thus expected and further studied by the TRKR and helicity-resolved transient reflectance measurements as below.

Figure 3(a) shows the TRKR response measured at 10 K under excitation of right (σ^+) and left (σ^-) circularly-polarized pump laser with photon energy of 1.746 eV. The

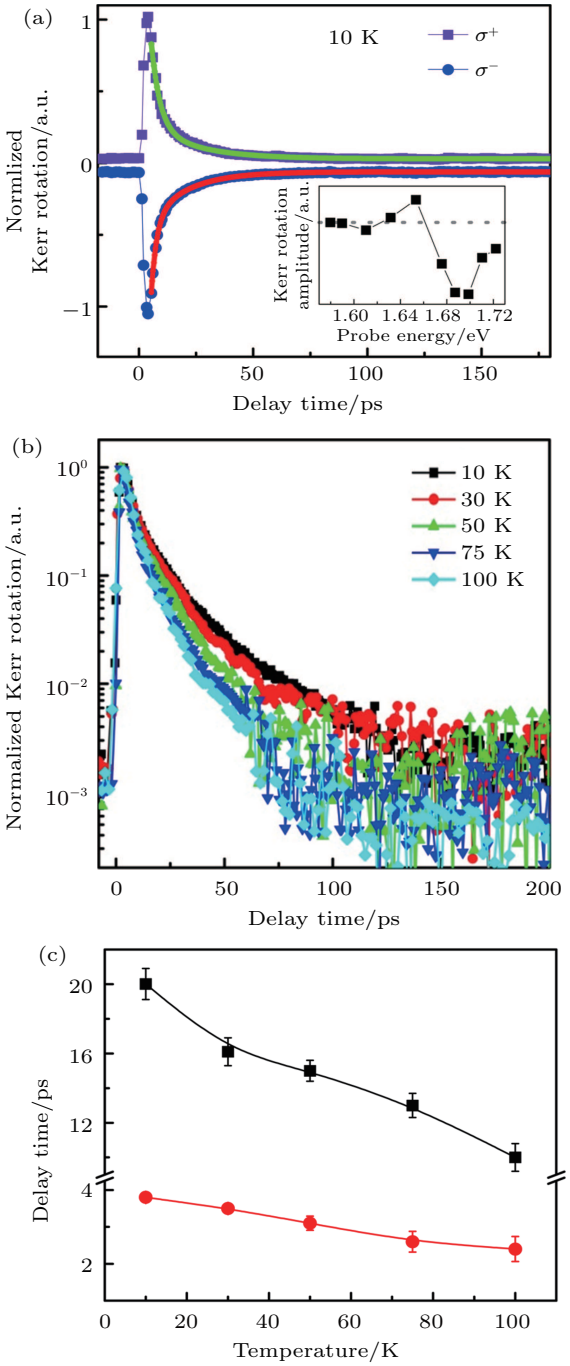


Fig. 3. (color online) (a) Time-resolved Kerr rotation response measured at 10 K under excitation of right (σ^+, violet trace) and left (σ^-, blue trace) circularly-polarized pump laser with photon energy of 1.746 eV and probe energy of 1.701 eV. These two traces are fitted by a bi-exponential decay function shown in green and red lines, respectively. The inset shows the amplitude of transient Kerr rotation signals at near zero delay time as a function of the probe laser energy with pump energy of 1.746 eV. (b) Kerr rotation dynamics excited by a σ^+-polarized pump pulse at 1.746 eV under different temperatures. The probe beam is set at the direct exciton resonance energy of 1.701 eV. (c) Temperature dependence of the depolarization time measured by TRKR.

probe laser energy is set at 1.701 eV, which is in resonance with the direct transition peak. By reversing the helicity of the pump pulse, a reversal sign of the Kerr signal is probed. The clear Kerr rotation response probed at direct transition energy indicates a large population imbalance between spin-up and spin-down excitons or carriers, which also conforms to the observed circular polarization for the direct exciton at the K valley. The probe-energy-dependent transient Kerr rotation amplitude which is recorded at near zero delay time with pumping energy of 1.746 eV, as illustrated in the inset of Fig. 3(a), clearly shows that the large spin polarization is present for the direct-transition exciton. However, no Kerr rotation signal can be detected when tuning the probe energy in resonance with the indirect exciton, which is coincident with the results of Fig. 2(c). Consequently, the temporal decay of Kerr signal indicates the dynamic depolarization process, which can be fitted well by a bi-exponential decay function. A fast decay time of ∼3.8 ps and slow decay time of ∼20 ps at temperature of 10 K is thus extracted.

Figure 3(b) shows the TRKR response measured at different temperatures, with pump excitation energy set at 1.746 eV and probe beam set at resonance energy of 1.701 eV for direct exciton. The decay process of TRKR at all temperatures can be fitted well by a bi-exponential decay function, with a short and long decay time extracted as shown in Fig. 3(c). It is seen that the fast decay time slightly reduces from 3.8 ps to about 2.5 ps as the temperature increases from 10 K to 100 K. The timescale of this short lifetime and its temperature dependent behavior agree well with the valley depolarization lifetime of a neutral exciton in monolayer WSe_2, as we previously studied.[22,24] This implies that the rapid polarization decay process with a short time scale of ∼3.8 ps is mainly dominated by the electron–hole exchange interaction between the K^+ and K^- valley in the same layer, as illustrated by the dashed green curve in Fig. 4(b), which has been proved to be the main valley depolarization mechanism in monolayer TMDCs.[21–24] In addition, the reported radiative lifetime of direct exciton in bilayer WSe_2 is as short as < 3 ps at 4 K.[26] The measured fast polarization decay of ∼3.8 ps in bilayer WSe_2 by TRKR is thus the combined consequence of short exciton lifetime and strong intervalley electron–hole exchange interaction, which is also the inherent feature of monolayer.

In Fig. 3(c), the slow decay process is characterized by a longer lifetime which decreases from ∼20 ps to ∼10 ps with increasing temperature from 10 K to 100 K. In order to understand this long spin decay time, we further performed transient helicity-resolved reflectance measurement at a temperature of 10 K, as displayed in Fig. 4(a). Under excitation via a σ^+-polarized pump pulse at 1.746 eV, the spin dynamics is investigated using a $\sigma^+(\sigma^-)$-polarized probe pulse at direct transition of 1.701 eV. The transient helicity-resolved reflectance response $\Delta R/R$ can be fitted by an exponential decay function with a characteristic decay time of ∼70 ps for the same circularly-polarized σ^+ probe (SCP, black trace) and ∼100 ps for opposite circularly-polarized probe σ^- probe (OCP, red trace). The 10-times longer lifetime obtained by $\Delta R/R$ measurement compared to that of exciton lifetime measured by time-resolved PL in Ref. [26] suggests that the lifetime obtained by $\Delta R/R$ reflects the optically-excited carrier lifetime which is in the timescale of tens of picoseconds. The signal difference between SCP and OCP, as presented in Fig. 4(a) with blue trace, indicates a clear circular dichroic effect which can be fitted well by a single-exponential decay function (magenta line). The spin lifetime is thus deduced to be around ∼25 ps, and this time scale is in accordance with the long spin lifetime measured by TRKR as presented in Fig. 3(a).

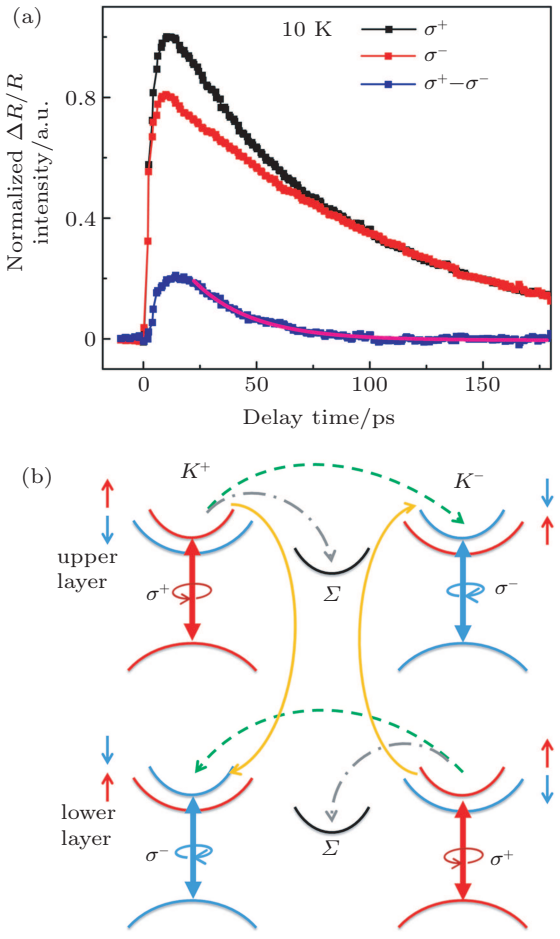


Fig. 4. (color online) (a) Helicity-resolved transient reflectance response ($\Delta R/R$) of the direct exciton at temperature of 10 K. The same (SCP, black trace) and opposite (OCP, red trace) circularly-polarized $\Delta R/R$ is recorded by σ^+- and σ^--polarized probes under σ^+-polarized pumping, respectively. The blue trace is the difference between the SCP and OCP signal, which is fitted by a single-exponential decay function shown in the magenta line. (b) Schematic of the spin flip and intervalley scattering process that induces spin relaxation in WSe_2 bilayer, under excitation of σ^+-polarized light (red thick arrow). The red or blue thin arrow represents the spin up or down configuration of electrons in the conduction band.

Considering the fact that the measured spin lifetime of ∼20 ps is obviously a few times longer than the fast direct exciton recombination time of ≤ 3 ps in bilayer WSe_2 reported

previously[26] and is also within the same order of magnitude with the carrier lifetime deduced from the transient helicity-resolved reflectance response $\Delta R/R$, this long spin lifetime of ~20 ps is suggested to be associated with the optically-excited spin-polarized electrons. This argument is further supported by the recent theoretical study of the intrinsic circular polarization in centrosymmetric TMDCs stack layers.[33] Under σ^+ excitation, only the electrons with pure right-handed character in the K^+ valley of the upper layer and the K^- valley of the lower layer can be excited effectively, as shown in Fig. 4(b), so that the excited electrons would possess a net up-spin that equals the local spin polarization on each monolayer. However, the spin relaxation of holes in the stacked TMDCs layers is expected to be fast, since the complex interstate spin-conserving and spin-flip relaxation processes for holes are expected to happen rapidly in the bilayer owing to the mixed feature of right- and left-handed characteristics of holes based on a theoretical study.[33] Moreover, the interlayer hole hopping at the K valley is expected to occur more likely compared to that of electrons, owing to the much larger interlayer hopping strength of holes for bilayer WSe_2.[29] The recent time- and angle-resolved photoelectron spectroscopy study has also reported the generation of spin-polarized carriers in inversion-symmetric bulk WSe_2 upon excitation with circularly-polarized pump pulses.[34] In this work, the band structure of WSe_2 bilayers calculated by density functional theory (DFT) shows that the global minimum of the conduction band is at the Σ point, which is located between Γ and K point in k-space. The time-dependent DFT reveals the two-dimensional character of the spin-polarized electrons that are initially excited within individual layers and can scatter extremely fast (< 100 fs) towards the global minimum Σ valley of the conduction band as sketched in Fig. 4(b), leading to relaxation of spin-polarized electrons. Moreover, the three-dimensional character of the global minimum band could then facilitate the interlayer charge transfer that further accelerates electron spin relaxation. Since the intervalley scattering from K to Σ valley and interlayer charge transfer are expected to be enhanced at higher temperature, this can cause a reduced electron spin lifetime as presented in Fig. 3(c). In addition, the photon energy of the pump laser is 45 meV higher than the direct A exciton resonance energy in our TRKR experiment, and the photo-excited electrons in the K^+ valley of the upper layer (K^- valley of the lower layer) could possibly relax to the lower layer (upper layer) within the same valley (shown in Fig. 4(b) with yellow arrows) accompanied by spin flip and excess energy dissipation to the environment.[29] The spin physics of excitons and carriers in bilayer TMDCs thus requires further theoretical and experimental investigation, including material design and interface engineering.[34]

4. Conclusion

We have investigated the spin relaxation dynamics of exciton and carriers in WSe_2 bilayer by combining time-resolved Kerr rotation and helicity-resolved transient reflectance measurements. The spin dynamics associated with the direct transition in WSe_2 bilayer has been revealed to exhibit a short decay time of ~3.8 ps and a long decay time of ~20 ps at the low temperature of 10 K. The short decay time of ~3.8 ps is assigned to be the exciton spin lifetime of WSe_2 bilayer, which is limited by the strong intervalley electron–hole exchange interaction between the K^+ and K^- valley in the same layer and short exciton lifetime of WSe_2 bilayer. Whereas the long decay time of ~20 ps is assigned to be the electron spin lifetime in WSe_2 bilayer, which is governed by the rapid intervalley scattering from K valley to global minimum Σ valley and the subsequent interlayer charge transfer in WSe_2 bilayer. Our experimental results further prove the robust circular polarization in centrosymmetric bilayers and especially the spin-polarized photoexcited carriers in bilayers, suggesting the possible engineering of carrier spins in multilayer TMDCs for their potential application in valleytronic and spintronic devices.

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