Optical spin pumping induced pseudomagnetic field in two-dimensional heterostructuresChongyun Jiang,¹ Abdullah Rasmita,¹ Weigao Xu,¹ Atac Imamoğlu,² Qihua Xiong,^{1,3,4,*} and Wei-bo Gao^{1,4,5,*}¹*Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore*²*Institute of Quantum Electronics, ETH Zürich, CH-8093 Zürich, Switzerland*³*NOVITAS, Nanoelectronics Center of Excellence, School of Electrical and Electronic Engineering, Nanyang Technological University, Singapore 637371, Singapore*⁴*MajuLab, CNRS-Université de Nice-NUS-NTU International Joint Research Unit UMI 3654, Singapore*⁵*The Photonics Institute and Centre for Disruptive Photonic Technologies, Nanyang Technological University, Singapore 637371, Singapore*

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Two-dimensional heterostructures are likely to provide new avenues for the manipulation of magnetization that is crucial for spintronics or magnetoelectronics. Here, we demonstrate that optical spin pumping can generate a large effective magnetic field in two-dimensional MoSe₂/WSe₂ heterostructures. We determine the strength of the generated field by polarization-resolved measurement of the interlayer exciton photoluminescence spectrum: the measured splitting exceeding 10 millielectron volts (meV) between the emission originating from the two valleys corresponds to an effective magnetic field of ~ 30 T. The strength of this optically induced field can be controlled by the excitation light polarization. Our finding opens up possibilities for optically controlled spintronic devices based on van der Waals heterostructures.

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Magnetic fields or pseudomagnetic fields interacting with a low-dimensional system not only show rich phenomena in condensed-matter physics [1,2], but are also likely to play a key role in realizing the technological promise of spin-based devices [3–5]. In direct analogy with spintronics, the coupling of valley degrees of freedom with a magnetic field in an atomically thin layer of graphene [6] or transition-metal dichalcogenides (TMDs) provides a different perspective towards the emerging field of valleytronics [7], as shown in cases of valley polarization [8,9], valley-polarized Landau levels [10,11], magnetic proximity effect [12,13], valley Zeeman splitting [14,15], and giant spin/valley susceptibility of TMD monolayers [16].

The introduction of a magnetic field in two-dimensional (2D) materials can be done in different ways, either internally or externally. Internally, intrinsic magnetism has been demonstrated in two-dimensional crystals CrI₃ [17] or Cr₂Ge₂Te₆ atomic layers [18]. Externally, the pseudomagnetic field can be generated by other stimuli such as strain [19], molecular doping [20], and light [21–26]. Regarding previous pioneer works using light, all of the reported approaches require ultrafast lasers with high peak power for realizing inverse Faraday effect [21–23], ac Stark shift [24,25], or, recently, Bloch-Siegert shift [26].

Here, we demonstrate a way to generate an optically induced pseudomagnetic field by an optical spin pumping mechanism in 2D TMD heterostructures. The introduction of a pseudomagnetic field relies on the ultrafast charge transfer [27,28] and optical spin pumping. The coupling between this field and the interlayer exciton results in optically detectable

valley degeneracy breaking. We observe a spectrum splitting between different valley transitions of more than 10 meV, which corresponds to a ~ 30 T pseudomagnetic field. We show that the degeneracy between the valleys can be lifted by using continuous-wave optical excitation and that the valley-splitting magnitude can be controlled by changing the excitation laser degree of circular polarization.

Our sample consists of a MoSe₂/WSe₂ heterostructure on a SiO₂/Si substrate with the MoSe₂ monolayer stacked on top of the WSe₂ monolayer [29,30]. Due to the band alignment of the MoSe₂/WSe₂ heterostructure, electrons and holes excited with light will relax into the conduction band of MoSe₂ and the valence band of WSe₂, respectively, after a short time. Electrons in MoSe₂ and holes in WSe₂ bound by Coulomb attraction form the interlayer exciton. Similar to the case of intralayer exciton in monolayer TMD [31–33], the spin and valley degrees of freedom of the interlayer exciton in a TMD heterostructure can be addressed using circularly polarized optical excitation [8,9]. For simplicity, without considering Moiré pattern polarization correction [34,35] (see the Supplemental Material, Sec. I [36]), here we denote right circularly polarized light (σ_-) coupled to spin up in the K valley and left circularly polarized light (σ_+) coupled to spin down in the K' valley.

The primary finding is that the energy of the emission from the K or K' valley depends on the excitation light polarization. Moreover, unlike in the case of the excitation-induced exciton energy splitting in a semiconductor quantum well [37–40] where the cross-polarized component (i.e., the emission polarization is orthogonal to the excitation polarization) has lower energy than the copolarized component (i.e., the emission polarization is the same as the excitation polarization), we observed that the cross-polarized component has higher

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FIG. 1. (a) Interlayer exciton spectrum at 2 K and magnetic field $B = 2$ T for σ_+ and σ_- excitation. The red (blue) solid and dashed lines indicate the spectrum and main peak position (obtained using multiple peak fitting) of σ_+ (σ_-) collection, respectively. The difference of peak position for the same collection polarization indicates optically induced valley-degeneracy breaking. (b) Magnetic field dependence of the valley splitting. The valley splitting is defined as the difference between σ_- and σ_+ collection peak position. Three different cases are shown: σ_- excitation, σ_+ excitation, and copolarized (collection state = excitation state) case. The excitation-state dependence of the valley splitting is attributed to the optically pseudomagnetic field. The copolarized case shows Zeeman-like linear behavior, while the other two show a nonlinear trend (a dip) near $B = 0$ T, which is attributed to the suppression of intervalley scattering by the out-of-plane magnetic field.

energy than the copolarized one. Figure 1(a) shows the interlayer exciton spectrum and peak position under different excitation polarizations at $B = 2$ T and temperature 2 K. As shown in Fig. 1(a), with σ_+ light excitation, light emission with polarization σ_+ has a lower energy than the light emission with polarization σ_- (fitting method shown in Supplemental Material, Sec. II [36]). More interestingly, the σ_+ emission energy becomes higher than the σ_- emission energy if we use σ_- light excitation, as shown in the lower spectrums in Fig. 1(a). We emphasize that we monitor the energy splitting of the σ_+ and σ_- emission, but not their intensity [9].

In order to know more about the spectrum splitting, we further measured the cases under a series of different out-of-plane magnetic fields. From Fig. 1(b) (red and blue dots), one can see that such spectrum splitting exists in all magnetic field strength (for data from another sample, see the Supplemental Material, Sec. III [36]). Even in the absence of magnetic field, such energy splitting exists, even though with a smaller value. The splitting here is much larger than the normal valley Zeeman splitting in an external magnetic field as measured in the copolarized cases [14,15], as denoted with purple triangles in Fig. 1(b).

Next, we analyze the mechanism of this optically controlled spectrum splitting and compare with previous pseudomagnetic-field-generation schemes. Unlike phenomena observed in the TMD monolayer [24,25], the observed optically induced valley-degeneracy breaking cannot be attributed to the valley-selective optical Stark effect. The optical excitation energy (1.707 eV) is too far blue-detuned from the interlayer exciton energy level range (1.3–1.4 eV) and its intensity is too small (<100 kW/cm²). For comparison, the detuning and the intensity of the optical pump used to generate

FIG. 2. (a) Schematic of the generation of the nonzero net spin. (i) Initial heterostructures. (ii) The heterostructure is excited with a circularly polarized laser creating the electron and hole in the WSe₂ layer. (iii) Due to the ultrafast charge (electron) transfer from WSe₂ to MoSe₂, most of the created conduction-band electrons migrate to the MoSe₂ layer. (iv) Both carriers (i.e., hole and electron) undergo depolarization mainly due to the intervalley scattering. Since the electron scattering rate in MoSe₂ (k_e) is much faster than the hole scattering rate in WSe₂ (k_h), the system net spin is not zero. This net spin creates a pseudomagnetic field that affects the exciton emission energy. (b) Rate-equation model for the process described in (a). In this model, $N_K^e(N_{K'}^e)$ and $N_K^h(N_{K'}^h)$ are the electron and hole population in the K (K') valley, respectively, N_R is the nonexcited population density, $\Gamma(\Gamma')$ is the electron-hole injection rate in the K (K') valley, γ is the electron-hole recombination rate, and $\sigma_K(\sigma_{K'})$ is the optical state that is coupled to the K (K') valley.

the valley-degeneracy breaking of more than 2 meV is around 0.2 eV red-detuned and 1 GW/cm², respectively [24]. Hence, the contribution of the valley-selective optical Stark effect can be neglected.

The emergence of the effective magnetic field using the strong spin-orbit coupling of valence band states in WSe₂ is sketched in Fig. 2(a). When we resonantly excite the exciton transition of WSe₂ with σ_- light, electrons and holes with spin up are generated in WSe₂. Due to the energy band alignment, electrons will tunnel to the conduction band of MoSe₂ in a short time, while spin-up holes remain in WSe₂, which provides the lowest-energy states available for holes. In this way, the electrons and holes are separated, ensuring that electron-hole exchange is ineffective in effecting a joint electron-hole valley flip. Strong spin-orbit coupling ensures that holes have a long spin/valley lifetime exceeding a microsecond [41]. Due to the Coulomb exchange interaction, the presence

of optically injected spin-polarized holes implies that in the presence of a large hole population in the K valley, the net Coulomb repulsion experienced by a K -valley (spin-up) hole is reduced as compared to that of a K' -valley hole. As a consequence, the energy of a K -valley interlayer exciton is reduced as compared to that of a K' valley, leading to the observed splitting in the emission spectrum. Note also that the spin polarization of the electron will also contribute additively to the splitting. However, the spin/valley lifetime of the electron is much shorter compared to that of the hole, which resulted in much smaller net polarization of the electron.

The fast change of valley splitting between $B = 0$ T to $B = \pm 2$ T [Fig. 1(b)] is observed for the case of σ_- and σ_+ excitation, but not observed for the copolarized case. To explain the rapid increase of the strength of the optically generated field with the applied external B field, we note that the electron-hole exchange-interaction-induced valley flip of intralayer excitons can be suppressed with B fields. Consequently, the increase in hole spin polarization with increasing B field stems from the suppression of hole valley flips taking place before the electron is transferred to MoSe₂ [42]. For a magnetic field magnitude bigger than 2 T, the expected linear behavior of valley splitting [1,2] is observed since the interlayer scattering has been largely suppressed. This nonlinear behavior is not observed in the copolarized case because the pseudomagnetic field due to two orthogonal optical excitations will have an opposite sign. Hence, the valley splitting is only affected by the external magnetic field for the copolarized case. More evidence that the valley splitting follows valley polarization is shown in the Supplemental Material, Sec. IV [36], for excitation laser power and wavelength-dependent measurement.

We use the rate-equation model shown in Fig. 2(b) to obtain an expression for the pseudomagnetic field. The pseudomagnetic field is proportional to the difference between the population of the spin-up and spin-down holes plus the difference between the populations of the spin-up and spin-down electrons. It can be written as

$$B \propto -\frac{2(\gamma + k_e + k_h)}{(\gamma + 2k_e)(\gamma + 2k_h)}(\Gamma_K - \Gamma_{K'}), \quad (1)$$

where B represents the pseudomagnetic field, $k_e(k_h)$ is the depolarization rate of the electron (hole), $\Gamma_K(\Gamma_{K'})$ is the electron-hole injection rate in the K (K') valley, and γ is the electron-hole recombination rate. In the Supplemental Material, Sec. V [36], we also give a microscopic description for the observed transition energy shift [43–45].

Based on Eq. (1), it is possible to tune the valley splitting by changing the proportion of the σ_+ and σ_- part of the optical excitation. In order to prove this, we measure the valley splitting as a function of the polarization state, $|\Psi_1\rangle = \frac{1}{\sqrt{2}}(|H\rangle + e^{i\alpha}|V\rangle)$ and $|\Psi_2\rangle = \cos(\frac{\alpha}{2})|H\rangle + ie^{i\phi}\sin(\frac{\alpha}{2})|V\rangle$, with $\phi = -136 \pm 5^\circ$ and various values of α . The data is collected for two different values of magnetic field ($B = -2$ and 2 T). The result is shown in Fig. 3. As shown in this figure, we have successfully controlled the valley degeneracy by excitation polarization manipulation. The data fit well with the pseudomagnetic field model (for the fitting formula, see Supplemental Material, Sec. I [36]). Based on this data, by comparing the amplitude of the oscillation with the constant

FIG. 3. (a),(b) Illustration of the trajectory of the optical state on the surface of the Poincaré sphere. The polarization state of the optical excitation is swept through two different trajectories. The state for those trajectories can be expressed as $|\Psi_1\rangle = \frac{1}{\sqrt{2}}(|H\rangle + e^{i\alpha}|V\rangle)$ and $|\Psi_2\rangle = \cos(\frac{\alpha}{2})|H\rangle + ie^{i\phi}\sin(\frac{\alpha}{2})|V\rangle$ with $\phi = -136 \pm 5^\circ$ for the trajectory in (a) and (b), respectively. (c),(d) Valley splitting as a function of the excitation polarization state. Here the dots represent the experimental data, while the lines are the theoretical fitting result. The result in (c) corresponds to the trajectory in (a), while (d) corresponds to (b).

difference between the valley splitting at $B = -2$ and 2 T, as shown by the difference of two oscillation curves in Figs. 3(c) and 3(d), we estimated that the pseudomagnetic field induced by circularly polarized light is $\sim \pm 31$ T.

In summary, we show that the degeneracy between the valleys can be lifted by using optical excitation even for vanishing external magnetic fields, which is induced by an optically induced spin pumping mechanism. Furthermore, we show that the valley splitting can be well controlled by manipulating the optical excitation state. The realization of an optically induced pseudomagnetic field will pave the way for optical control of valleytronics devices, which is considerably faster than controlling the external magnetic field.

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APPENDIX: MATERIALS AND METHODS

The MoSe₂/WSe₂ heterostructure is fabricated via mechanical exfoliation and aligned transfer method. The stacking sequence of the sample is checked using second harmonic generation (SHG)-based measurement and it is found to be AA stacking. A homemade fiber-based confocal microscope is used for performing the polarization-resolved photolumi-

nescence (PL) spectroscopy. The polarization state of the excitation and collection is controlled using a combination of polarizer, quarter-wave plate, and half-wave plate. The PL emission is directed by a multimode optical fiber into a spectrometer (Andor Shamrock) with a CCD detector for spectroscopic recording. The sample is loaded into a magnetocryostat to control the magnetic field and sample temperature.

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