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Exciton Hall effect in monolayer MoS₂

Masaru Onga¹, Yijin Zhang^{2,3}, Toshiya Ideue¹, Yoshihiro Iwasa^{1,4*}

¹ Quantum-Phase Electronics Center (QPEC) and Department of Applied Physics, The University of Tokyo, Tokyo 113-8656, Japan

² The Institute of Scientific and Industrial Research, Osaka University, 8-1, Mihogaoka, Ibaraki, Osaka 067-0047, Japan

³ Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany

⁴ RIKEN Center for Emergent Matter Science (CEMS), Wako 351-0198, Japan

*Correspondence to: iwasa@ap.t.u-tokyo.ac.jp

1. Sample preparation

We note a remarkable effect of the sample quality on the observation of the exciton Hall effect (EHE). The exciton diffusions and EHE signals in our measurement is so sensitive to the surface state that the clean-up process (mentioned in Methods) seems to be important. Additional procedures such as electron beam lithography with resists can also affect or sometimes degrade the PL profile. In our measurements we intentionally used the flakes just after the clean-up process for the best condition.

2. Reproducibility in other samples

The EHE is also observed in other two flakes. Figure S1 shows the real-space observation of the EHE in another flake (sample B). The same phenomena explained in the main text (Fig. 2 & 3) were observed. Here long and narrow flakes were also chosen for

the observation of EHE. As shown in table S1, the α_{EHE} in each flake are summarized. The values are ~ 0.2 in all monolayer MoS₂ samples.

3. Control experiment: the case of a large sample

The narrow and long shape of the flakes is crucial for observing the EHE by causing the unidirectional transport of excitons. As a control experiment, we carried out the same experiment on a large flake as shown in Fig. S2a. Even though a clear exciton diffusion was caused by the laser illumination (Fig. S2b), no appreciable EHE signals were observed (Fig. S2c) because of the omnidirectional diffusion of excitons.

4. Definition and evaluation of the Hall angle

Geometrical definition of the Hall angle (α_{EHE}) introduced in the main text is simple and straightforward in the case that the Hall motion is traced directly, which is also discussed in a direct observation of the skyrmion Hall effect with spatially-resolved magneto-optical Kerr effect¹ and X-ray microscopy². Figure S3 shows the α_{EHE} as a function of x within the region (ii). The L_{xy} (ΔI_{norm} and I_{sum}) was too small to evaluate the α_{EHE} properly in the regions (i) (region (iii)).

On the other hand, the Hall angle is conventionally defined by the ratio of the spin Hall current to the total dissipative current along the driving force. For direct comparison with the common Hall angle, we define another exciton Hall angle β_{EHE} as following;

$$\beta_{\text{EHE}} = \frac{L_{xy} \sum_y |I_{\sigma+}(x, y) + I_{\sigma-}(x, y)|}{L_{xx} \sum_y (I_{\sigma+}(x, y) + I_{\sigma-}(x, y))},$$

where each notation is identical to the main text. Here, we estimate the exciton Hall current as $L_{xy} \sum_y |I_{\sigma+}(x, y) + I_{\sigma-}(x, y)|$ and total exciton current as $L_{xx} \sum_y (I_{\sigma+}(x, y) + I_{\sigma-}(x, y))$, which we can directly compare β_{EHE} with other Hall angles. In this estimation β_{EHE} in sample A is 0.035, which is comparable to the spin Hall angles of normal metals.

In both definitions (geometrically-defined α_{VHE} and conventionally-defined β_{EHE}), the values are more than two orders of magnitude larger than the valley Hall angle estimated from Ref. 3. This suggests that the origin of the Berry curvatures of composite particles (excitons) is different from that of the single particles such as electrons and holes.

5. The effect of the sample shape on the Hall angle

The present definition of the Hall angle is validated only in the adequate shape of the samples, which is also the reason why we chose such particular flakes. In wide flakes, the omnidirectional diffusion makes it impossible to define α_{EHE} (Supplementary Information Section 3 and Fig. S2). On the other hand, in quite narrow (wire-like) flakes, it is also difficult to define the L_{xy} and α_{EHE} because of the short widths.

Although the edges of the samples would affect the exciton recombination and the estimation of α_{EHE} , the influence of the edges is not dominant. Table S2 shows the sample widths and α_{EHE} determined at the same x in samples A and B. Despite the significant difference in the widths, the almost identical values of α_{EHE} were obtained. This indicates the edges do not affect the present estimation of α_{EHE} .

6. Theoretical differences between the neutral and charged excitons

We note that there are crucial differences between Hall effect of charged/spin-polarized particles and neutral/spin-unpolarized excitations. If the contributions of trions are dominant, the observed excitation Hall effect can be also spin Hall effect of charged particles. Quantum statistics might be also different between trions and neutral excitons, leading to distinct theoretical consequences of the particles' transport. In the main text, we raise the possible perspective to distinguish these two.

We also note the importance of spin-orbit coupling in this system. The effects of spin-orbit interaction (SOI) are included only as the Zeeman-type spin splitting in the present discussion. That is the same assumption as previous works about valley transport in monolayer TMDs³⁻⁵. However, the direct contribution of the SOI to the quantum transport should be considered for further understandings of spin-valley transport in this system. Especially, it could be important to directly calculate the spin-dependent Berry curvature by fully considered tight-binding model with LS term^{6,7}, possibly revealing different origins between neutral excitons and spin-polarized trions in future.

7. Excitation-position dependence

In Fig. S4, we show the position and polarization dependence of the PL spectra in sample A. We did not observe significant differences in linewidths and peak positions among all spectra, indicating that the pumped excitonic states do not depend on the position and polarization. Although the intensity ratios of A-exciton and localized exciton are slightly modified at different positions possibly due to the non-uniformity of the sample, it should not affect the EHE.

8. Comparison with the measurements at room-temperature

We also attempted to measure the EHE at room temperature to evaluate the contribution to the EHE from $\nabla\mu$ and ∇T . Figures S5a and b display the distributions of excitation laser and PL of Sample C at room temperature (300 K) and 30 K, respectively. At 300K, the diffusion of excitons was smaller than that at 30 K, and therefore, we couldn't observe any discernible signals at 300 K in contrast to the clear EHE signals at 30 K (Figs. S5c, d). There are two scenarios to explain this result. One is that the diffusion coefficient at 30 K is much larger than that at 300 K, and the other is that the temperature gradient is the main driving force of excitons and the effective gradient $\nabla T/T$ increases as T decreases. Because the heating effect in shining the laser is inevitable in principle, further sensitive experiments are needed to distinguish these two contribution.

9. Excitation-power dependence

Here we show the power dependence of the spectra and exciton diffusion at 30 K in Fig. S6. Figure S6a demonstrates the power-dependent spectra showing almost no difference within the range of our setup. In Fig. S6b, we plot the integrated PL intensities for A-exciton and localized exciton separated by using Gaussian-fitting. The solid line corresponds to (Integrated intensity) $\propto$ (Excitation power) $^\gamma$, where $\gamma = 0.95 \pm 0.03$ (0.79 ± 0.05) for A-exciton (localized exciton). The deduced γ of A-exciton, which we now focus on, reaches almost 1, implying that exciton generation and recombination would be proportional to the excitation intensity. This also supports that we would neglect any non-linear and non-radiative effects such as exciton-exciton annihilation⁸.

Such linearity is also observed in the diffusion profiles (Fig. R4c). The PL intensities are normalized at $x = 0 \text{ }\mu\text{m}$, thus the almost same PL profiles among the all power indicate the exciton diffusion is independent of the excitation power within the present range. This does not identify whether the driving force of excitons is the gradients of temperature or that of chemical potential, because both gradients are supposed to depend linearly on the laser fluence.

Further experiments with the external temperature gradient by a micro-heater⁹ will be helpful to separate the gradient of temperature from the gradient of exciton densities.

10. Profile of the temperature gradient

We also estimated the temperature gradient induced by excitation laser, using a micro-thermometer made of Cr/Au (5 nm / 30 nm) (Fig. S7a). We measured the base temperature dependence of the four-terminal resistance of the thermometer for calibration. And used the resistance as a local thermometer (the detailed methods are mentioned in Ref. 9). Now $\Delta T = T_{\text{light}} - T_{\text{dark}}$, where T_{light} (T_{dark}) is a temperature measured by the micro-thermometer with (without) illumination of the laser (200 μW). We moved the laser spot to measure the x -dependency of ΔT instead of changing the position of the thermometer, showing a spatial profile of temperature under excitation laser (Fig. S7b). The maximum temperature gradient reaches $\sim 0.5 \text{ K}/\mu\text{m}$ ($\sim 5000 \text{ K}/\text{cm}$). This large gradient is supposed to be the driving force for the exciton transport and the EHE.

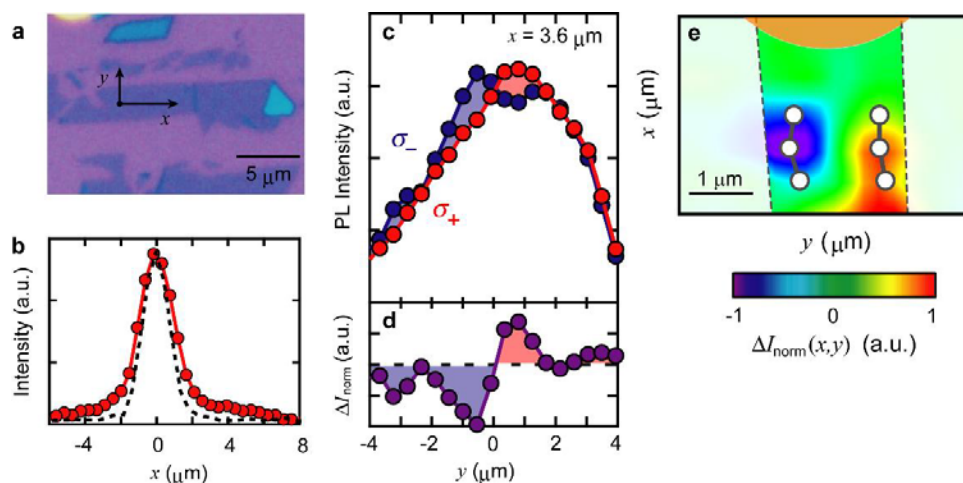


Fig. S1. Observation of the EHE in sample B. **a**, Optical image of sample B. **b**, Longitudinal profile along with x -axis in **a**. **c**, **d**, Polarization-resolved transverse profile of the PL intensity and ΔI at $x = 3.6 \mu\text{m}$, representing the occurrence of the EHE. **e**, Color plot of ΔI .

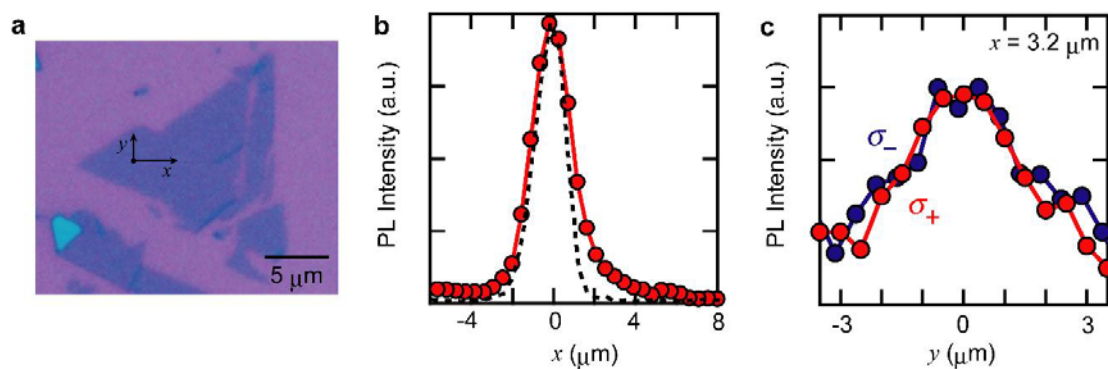


Fig. S2 Measurements in a large flake. **a**, Optical image of sample D. **b**, Longitudinal profile along with x -axis in **a**. **c**, Typical transverse profile of the PL intensity at $x = 3.2$ μm .

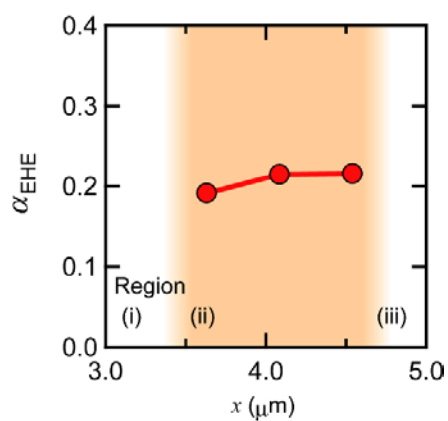


Fig. S3 The exciton Hall angle vs. x in region (ii). There is almost no x -dependency in α_{EHE} .

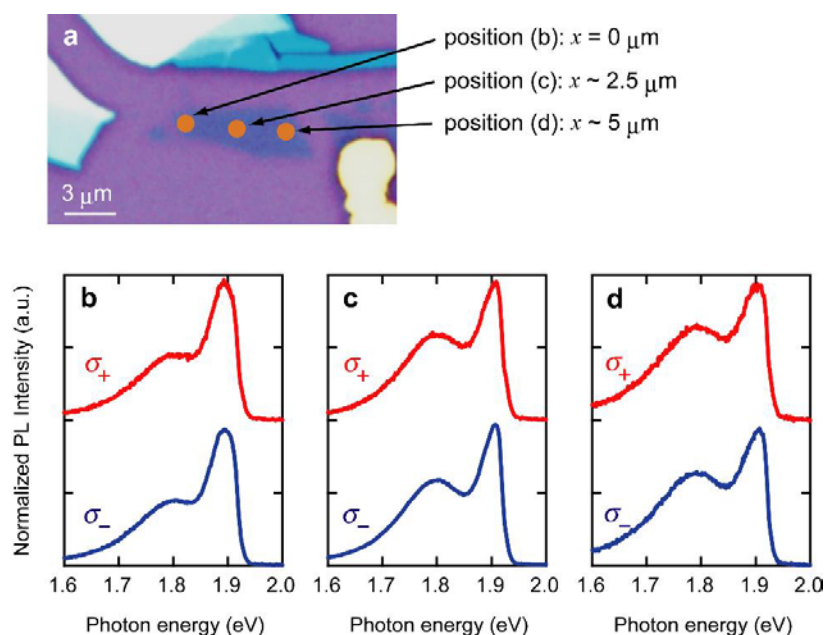


Fig. S4. Position and polarization dependence of PL spectra. **a**, Optical image of the sample A and excitation positions in **b**, **c**, and **d**. **b-d**, Polarization-resolved PL spectra at different excitation points. We simultaneously measured each couple of spectra by shining linearly polarized light.

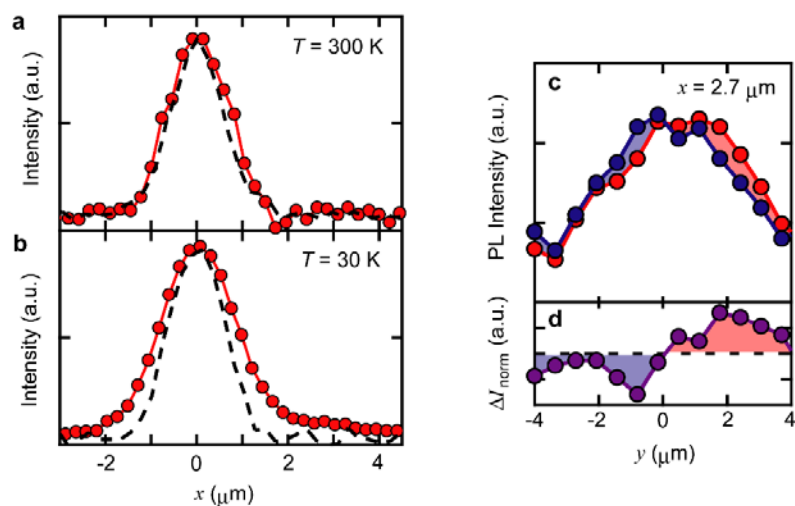


Fig. S5. Temperature dependence of the diffusion. **a, b,** Profiles of diffusion of excitons at 300 K and 30 K in sample C. **c, d,** Signals of the EHE at 30 K in this sample at $x = 2.7$ μm .

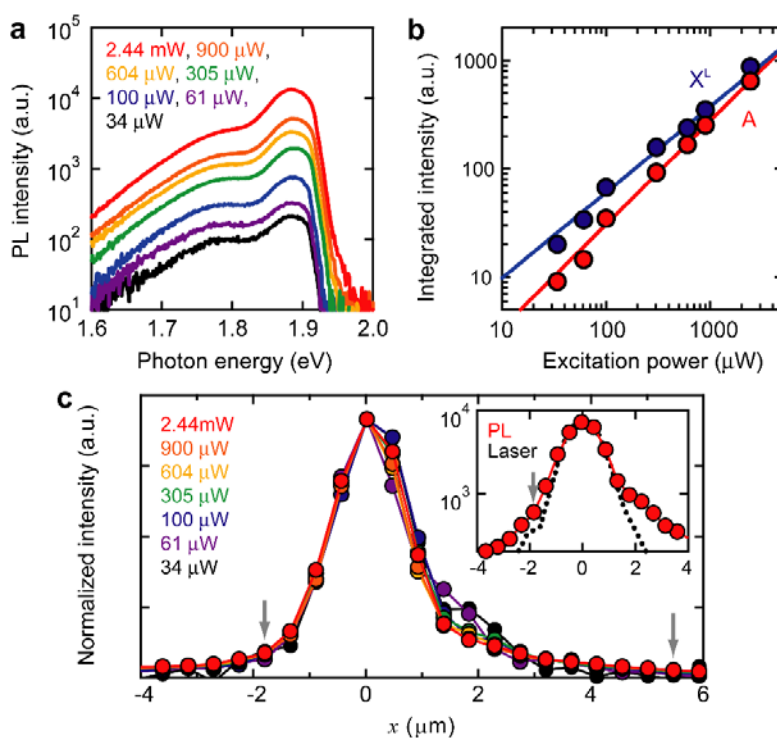


Fig. S6. Excitation-power dependence. **a**, Excitation-power dependence of PL spectra at 30 K. Different colors correspond to the spectra in the different excitation power. **b**, Integrated intensity vs. excitation power. Each point is calculated by Gaussian-fitting of the spectra. Red (Blue) data correspond to A-exciton (localized exciton), and the lines denote the fitting lines. **c**, Fluent-dependent cross-sectional profiles of the exciton diffusion (similar to Fig. 2d). The PL intensities are normalized at $x = 0$ μ m. Gray arrows denote the sample edges. Inset: the comparison between laser and a typical PL profile. Both axes are same as the main Fig. R6c. Note that the excitation power of the experiment in the main text is around 300 μ W.

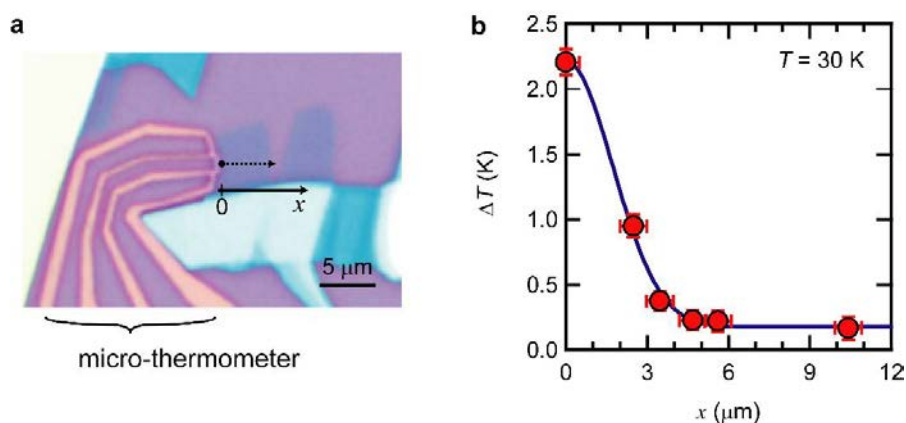


Fig. S7. Temperature gradient induced by laser. **a**, Optical image of a typical device for measuring the temperature profile. We used the Cr/Au (5 nm/30 nm) electrodes as a micro-thermometer, by measuring the change of the four-terminal resistance⁹. **b**, Temperature difference of the electrodes with and without illumination of the laser (200 μW) as a function of x -coordinate. The base temperature is 30 K, and the blue line is a fitting curve with a Gaussian function.

Sample	A	B	C
α_{EHE}	0.20	0.19	0.24

Table S1. Summary of the α_{EHE} . We summarized the average α_{EHE} in three flakes of monolayer MoS₂.

(at $x = 3.6 \mu\text{m}$)	Sample widths	α_{EHE}
Sample A	2.81 μm	0.191
Sample B	2.17 μm	0.188

Table S2. Sample widths vs. α_{EHE} . Summary of the sample widths and α_{EHE} of the samples A and B. All values are estimated at the same x ($= 3.6 \mu\text{m}$) in order to exclude other geometrical effects except the sample widths.

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