



Supplementary Material for

The valley Hall effect in MoS₂ transistors

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The Valley Hall Effect in MoS₂ Transistors

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1. Materials and Methods:

1.1 Device fabrication.

MoS₂ monolayers were mechanically exfoliated from bulk MoS₂ crystals onto Si substrates coated by 300 nm of SiO₂. Monolayer samples were identified using a combination of optical contrast and photoluminescence spectroscopy (8). Standard electron beam lithography techniques were used to define metal contact areas on our exfoliated samples. Electron beam evaporation was used to deposit 0.5 nm Ti/50 nm Au contacts, followed by a standard methylene chloride/acetone lift-off procedure. Using electron beam lithography to create an etch mask, we defined the Hall bar geometry using a ten-second low-pressure SF₆ plasma etch. Finally, the

device was laser annealed in high vacuum (28, 34) ($\sim 10^{-6}$ torr) at 120 °C for ~ 10 hours before measurement. We note that the reasons for creating a Hall bar device with a long Hall probe and a short photoconduction channel (Fig. 1B in main text) are two-fold: 1) we want the photocurrent (which is generated most efficiently at the contacts) to be produced near the center of the device so that any Hall voltage can be efficiently picked up by the Hall probe; 2) we want to reduce the background photovoltage generated at the metal-semiconductor contacts of the Hall probe (see below). Under this Hall bar geometry, the Hall probe typically picks up a finite background voltage signal (due to remnant longitudinal-transverse coupling), at about one part per 100 of the source-drain bias V_x . This finite longitudinal-transverse coupling does not affect our anomalous Hall effect measurement, which is performed under modulations of the handedness of incident light (see below). Multiple devices have been investigated in this study (6 devices for monolayers and 2 for bilayers).

1.2 Photoconduction and Hall voltage measurements.

Measurements were performed in a Janis cryostat cooled by liquid nitrogen and placed on an inverted microscope. A standard Hall voltage measurement was performed with a source-drain voltage V_x applied across the short channel as shown in Fig. 1B in the main text. The voltage difference between the A and B contacts of the Hall probe was measured by a voltage amplifier, whose output was further sent to a lock-in amplifier. For our photocurrent measurement, a Fianium supercontinuum laser source with a monochromator (selecting a line width of ~ 5 nm for each color) was used for acquiring the photoconductivity and Hall conductivity spectra. A diode laser (centered at 1.9 eV) was used for all other optical excitations. Photocurrent and Hall voltage maps were obtained by scanning the laser spot across the samples

with a pair of scanning mirrors, and reflection images were obtained by collecting the reflected light in a silicon photodiode.

To modulate the polarization of the incident light, the laser was linearly polarized and passed through a photoelastic modulator before being focused onto the sample through a 40x long working distance objective (spot diameter between 1–3 μm depending on the specific measurement). The angle of incidence θ of the linearly polarized light with respect to the fast axis of the modulator was varied by a half waveplate so that the photon ellipticity could be continuously tuned while the phase retardation between the fast and slow components was modulated at 50 kHz. The main experimental results involving quarter-wave ($\Delta\lambda = 1/4$) modulation between circularly polarized photons of different handedness were obtained with a lock-in detection frequency of 50 kHz. On the other hand, the phase shift of the modulator was switched from quarter-wave ($\Delta\lambda = 1/4$) to half-wave ($\Delta\lambda = 1/2$) modulation for the control experiments involving modulation with linearly polarized light and the lock-in detection frequency was changed to twice the fundamental modulation frequency, i.e. 100 kHz, so that no modulation involving circularly polarized light could be picked up. Note that effects from possible modulation of the beam position were minimized by control experiments using expanded illumination (beam diameter $\sim 5 \mu\text{m}$). Moreover, undesirable power modulation of the optical excitation was reduced to a level less than 10^{-4} at the sample position, contributing negligible perturbations to our Hall voltage measurement.

2. Supplementary Text:

In the main text, we present data for one monolayer and one bilayer device. We name these devices M1 and B1, respectively, and continue this naming scheme with monolayer devices M2-M5 and bilayer device B2 in the following supplementary information.

2.1 Derivation of the anomalous Hall conductivity

According to Refs. 4 and 6, the Hall conductivity for the electrons in the K' valley of an MoS₂ monolayer originating from the intrinsic Berry curvature effect can be written as

$$\sigma_{H,K'} = \frac{\pi e^2}{h} \int_{E_g/2}^{\infty} d\epsilon g(\epsilon) \Omega_{e,K'}(\epsilon) f_e(\epsilon). \quad (S1)$$

Ignoring spin-orbit coupling, $g(\epsilon) = \frac{2m_e\epsilon}{\pi\hbar^2 E_g}$ is the electron density of states at the K' valley,

$\Omega_{e,K'}(\epsilon) = \frac{\hbar^2 E_g^2 / m_e}{8\epsilon^3}$ is the Berry curvature and $f_e(\epsilon)$ is the Fermi-Dirac distribution. In the degenerate limit, $\sigma_{H,K'}$ becomes

$$\sigma_{H,K'} \approx \frac{e^2}{h} \frac{\mu_{K'}}{E_g} = \frac{e^2}{h} \frac{\hbar^2 \pi n_{K'}}{2m_e E_g}, \quad (S2)$$

where $\mu_{K'}$ is the chemical potential and $n_{K'}$ is the total electron density at the K' point. In this limit, the anomalous Hall conductivity σ_H becomes (including only the electron contribution)

$$\sigma_H \approx \frac{e^2}{h} \frac{\hbar^2 \pi \Delta n_v}{2m_e E_g}. \quad (S3)$$

Here, Δn_v is the carrier density imbalance between the two valleys generated by photoexcitation and m_e is the electron band mass.

In the nondegenerate limit, we can show that $\sigma_{H,K'}$ becomes

$$\sigma_{H,K'} \approx \frac{e^2}{h} \frac{\hbar^2 \pi n_{K'}}{2m_e E_g} F\left(\frac{E_g}{2k_B T}\right), \quad (S4)$$

where $F \approx 1$ is dimensionless and is weakly dependent on temperature. Equation S4 thus reduces to Eq. S3, so the expression for the Hall conductivity σ_H when expressed in terms of the carrier density is approximately the same in both the degenerate and nondegenerate limits. Note that the above derivation is for the intrinsic Berry curvature effect. It is shown in Ref. 4 that the side-jump contribution is twice as big as the intrinsic effect and has the opposite sign. Thus, including the intrinsic and side-jump contributions, the anomalous Hall conductivity can be reduced to Eq. 1 in the main text.

2.2 Temperature-dependent electrical transport

To better understand the electrical transport properties of our devices, we examined the temperature and gate dependence of the resistivity. We show the temperature dependence of the resistivity ρ_{xx} versus gate voltage V_g for monolayer device M1 in Fig. S1A. We clearly see the presence of a metal-insulator transition across $V_g = 0$ V: the resistivity increases with decreasing temperature for $V_g < 0$ V (the insulating regime) and vice versa for $V_g > 0$ V (the metallic regime). This is further illustrated in Fig. S1B, which shows the temperature dependence of the resistivity at different V_g . Consistent with recent observations (28, 35) and with previous studies on 2D electron gases in various semiconductor systems (36), the transition occurs near a resistivity value of $\rho_{xx} \sim \frac{h}{e^2} = 2.6 \times 10^4 \Omega$ that obeys the Ioffe-Regel criterion (37) $k_F l \sim 1$. Here k_F and l are the Fermi wave-vector and the mean free path of the electrons, respectively.

2.3 Photocurrent and photoconduction

In Fig. S2, we compare the scanning photocurrent image of device M5 under 0 V and 0.5 V bias. The photocurrent under the zero-bias condition (< 5 nA) is negligible compared to that generated from photoconduction (> 300 nA).

2.4 Scanning Hall voltage microscopy

We characterized our devices under illumination by spatially mapping their photocurrent and Hall voltage responses. The maps for monolayer device M2 and bilayer device B1 are compared in Fig. S3; all of the maps were recorded at $V_g = 0$ V and $V_x = 0.5$ V, using a continuous wave laser (centered at 1.9 eV with spot diameter ~ 1 μ m) at an incident power of ~ 50 μ W. Figure S3A shows the scanning photocurrent image of monolayer device M2. The photocurrent is mainly generated at the center of the device where a source-drain bias voltage V_x is applied across the short channel. The corresponding scanning Hall voltage (V_H) images are shown in Figs. S3B and C for R-L and L-R modulations, respectively. We see that a finite Hall voltage is produced at the center of the device, coinciding with the location of photocurrent production. Furthermore, the sign of V_H reverses when the helicity of the modulation changes from R-L to L-R.

In Figs. S3D, E and F, we show the results from bilayer device B1 as a control experiment. Although a similar photocurrent is again produced at the center of the device, the Hall voltage is much smaller (by about a factor of 10) than that of the monolayer device. We note that significant photovoltages (particularly in the bilayer device) are also observed at the metal-semiconductor contacts of the Hall probe (both at zero and finite bias along the short channel). These photovoltages probably arise from the modification of the polarization state by the metal contacts, which leads to a corresponding power modulation. We used a long Hall probe

in our experiment so that laser illumination of our Hall probe contacts and the resulting effects of undesirable signals on our measurement would be minimal. Overall, the observation of a significant Hall voltage only at the centers of monolayer devices confirms the observation of the VHE.

2.5 Data from extra monolayer devices

Figure S4 shows Hall voltage measurements from monolayer devices M2-M5, recorded at gate voltage $V_g = 0$ V and under 1.9 eV excitation with intensity $100 \mu\text{W } \mu\text{m}^{-2}$. Again, we see a finite Hall voltage that scales linearly with the source-drain voltage V_x only for the L-R and R-L modulations of the incident laser beam, and the sign of the effect is the same for all devices. Furthermore, the Hall voltage vanishes in all devices under half-wave modulation.

Figure S5 shows scanning photocurrent and Hall voltage maps for monolayer devices M1, M4, and M5. The measurement schematic for all maps is indicated in Fig. S5A, and all maps were recorded with gate voltage $V_g = 0$ V and source-drain bias $V_x = 0.5$ V, under 1.9 eV excitation with intensity $50\text{-}150 \mu\text{W } \mu\text{m}^{-2}$. For all devices, the Hall voltage is strongest when the laser is focused on the center of the device. We observe a sign change when we switch from R-L to L-R modulation, and the signs are consistent with the data presented in Fig. S4.

2.6 Dependence of the Hall voltage map on excitation photon energy

In Fig. S6 we compare the response of monolayer device M1 under on- (1.9 eV) and off- (2.3 eV) resonance excitation by displaying scanning photocurrent and Hall voltage maps recorded at source-drain bias $V_x = 0.5$ V and gate voltage $V_g = 0$ V. The incident intensity was $100 \mu\text{W } \mu\text{m}^{-2}$. While the device displays a similar photocurrent response under both the 1.9 eV

(Fig. S6A) and 2.3 eV (Fig. S6C) excitation energies, we only observe a significant Hall voltage under R-L modulation when the device is excited at its band gap, 1.9 eV, as in Fig. S6B.

2.7 Electrical characterization of bilayer devices

Figure S7A shows the 4-point conductivity as a function of back gate voltage for bilayer device B2. Similar to the monolayer devices, only n-type behavior is seen. A 4-point mobility of about $500 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ is deduced for this particular device. In general, bilayer devices have slightly higher electron mobilities (typically between 100 and $500 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) compared to their monolayer counterparts (typically between 50 and $300 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$).

In Fig. S7B we compare the photoconduction spectra for monolayer device M2 and bilayer device B2. The similar photoconduction spectra reflect their similar absorption spectra originating from direct optical transitions. The result is consistent with recent optical studies of the absorption spectra of MoS_2 samples with varying thickness (8). Given all these similar characteristics, bilayer MoS_2 devices provide an ideal platform for control experiments demonstrating the absence of the VHE in an inversion-symmetric system.

2.8 Comparison of the second mono- and bilayer devices

In Fig. S8, we show more detailed comparisons of the monolayer device M2 and bilayer device B2. Figure S8A shows the bias (V_x) dependence of the anomalous Hall voltage. Again, finite Hall voltages that depend weakly on the gate voltage (V_g) are seen for the monolayer device, and no Hall voltage up to a bias of $\pm 1 \text{ V}$ is seen in the bilayer device for all gate voltages. Figure S8B shows the corresponding gate dependence of the anomalous Hall resistance. A Hall

resistance of a few Ω is seen for the monolayer device while that for the bilayer is less than 0.1 Ω .

2.9 Photodoping density

We extract the photoexcited carrier density Δn_{ph} by measuring the gate dependence of $\Delta\sigma_{xx}$ at different incident laser powers (inset of Fig. S9A). As mentioned in the main text, Δn_{ph} can be obtained from the relation $\Delta\sigma_{xx} = \Delta n_{ph} e \mu$ with $\mu = \frac{1}{C_g} \frac{d\sigma_{xx}}{dV_g}$. The photoexcited carrier density Δn_{ph} as a function of back gate voltage V_g at different excitation intensities for monolayer device M1 is shown in Fig. S9A. At high excitation intensities, a charge density Δn_{ph} on the order of 10^{11} cm^{-2} is seen.

Figure S9B shows the dependence of the carrier density Δn_{ph} on the incident laser intensity P at different gate voltages. The observed saturation behavior might be explained by the presence of trapped-charge contributions to the photoconduction, as it is similar to the observed intensity saturation in disorder-induced photoluminescence that originates from the change in occupancy of the trapped states (37). More systematic studies of the dependence of the photoconduction on the amount of disorder in the system are required for a better understanding of the laser power dependence of the photoresponse.

2.10 Gate voltage dependence of σ_H

The gate voltage (V_g) dependence of the anomalous Hall conductivity σ_H from monolayer devices M1-M3 and M5 under R-L modulation (centered at 1.9 eV with an excitation intensity of $150 \mu\text{W } \mu\text{m}^{-2}$) is shown in Fig. S10; note that it increases with electron doping. As mentioned in the main text, no dependence on the gate voltage is expected according to the simplest theoretical

model. Possible explanations for this discrepancy have been discussed in the main text by considering the portion of Δn_{ph} that contributes to the Hall effect and the presence of extrinsic contributions.

Supplementary Figures:

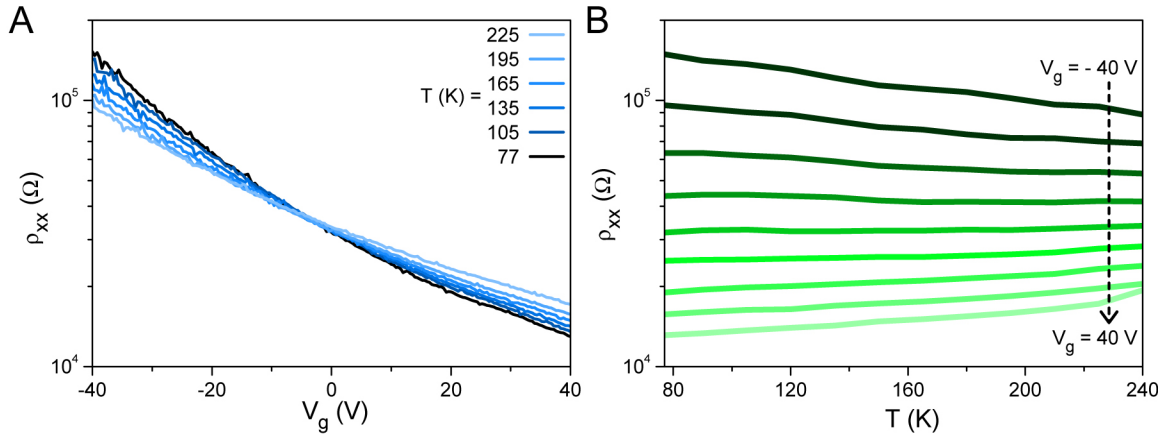


Fig. S1. Temperature-dependent electrical transport in monolayer device M1. (A) Resistivity ρ_{xx} as a function of back gate voltage V_g at different temperatures. **(B)** Temperature (T) dependence of ρ_{xx} at different back gate voltages. A metal-insulator transition is observed near $\rho_{xx} \sim \frac{h}{e^2} = 2.6 \times 10^4 \Omega$.

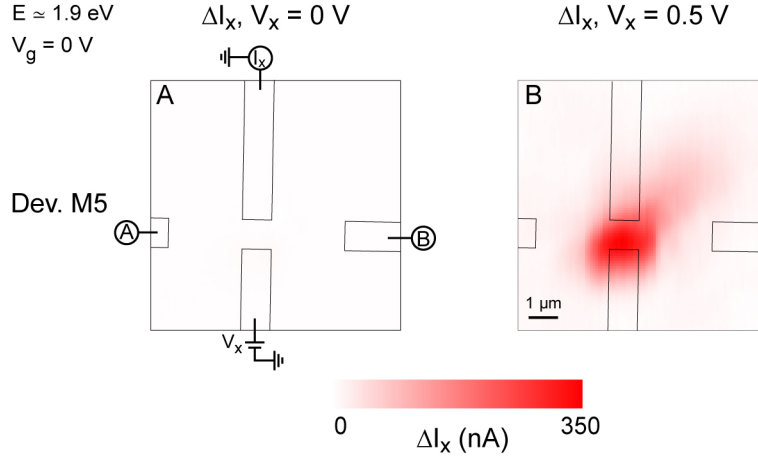


Fig. S2. Photocurrent and photoconduction in monolayer device M5. The measurement schematic for both maps is indicated in (A); both maps were recorded with gate voltage $V_g = 0$ V under 1.9 eV excitation at a power of ~ 50 μW and a spot diameter of ~ 1 μm . (A) Scanning photocurrent image of monolayer device M5 under zero bias ($V_x = 0$ V). (B) Scanning photocurrent image of monolayer device M5 under bias $V_x = 0.5$ V.

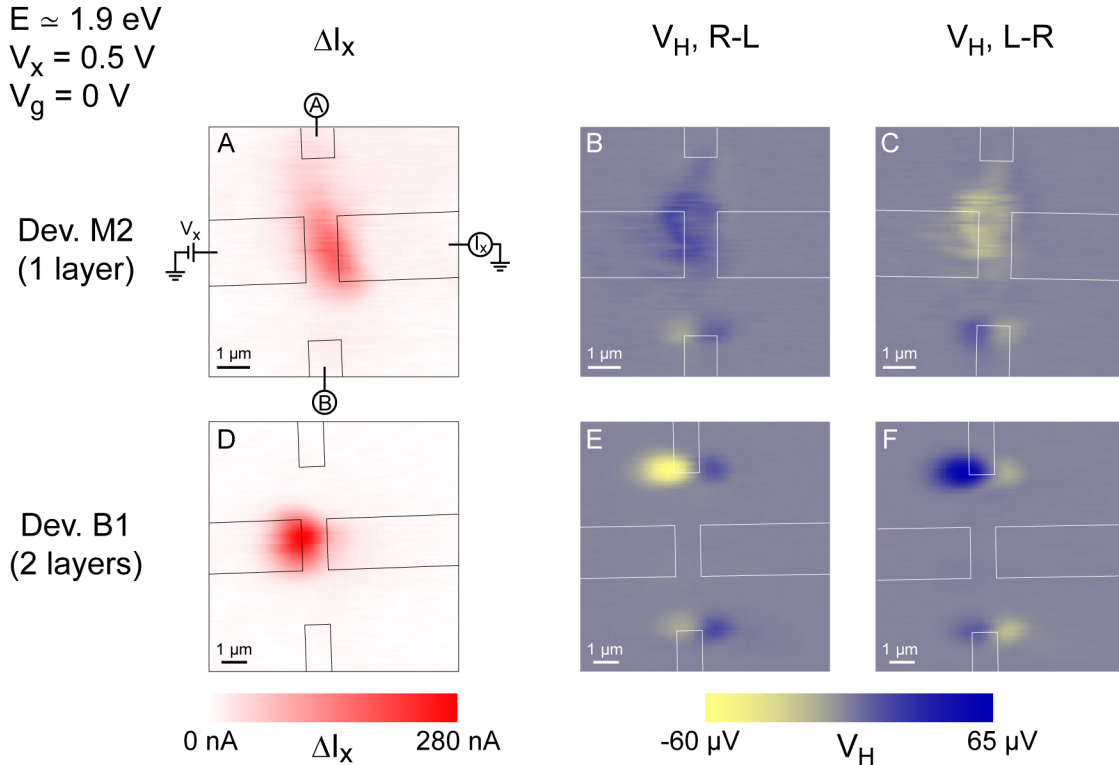


Fig. S3. Comparison of scanning photocurrent and Hall voltage images for monolayer device M2 and bilayer device B1. The measurement schematic for all maps is indicated in (A); all maps were recorded with gate voltage $V_g = 0$ V and source-drain bias $V_x = 0.5$ V, under 1.9 eV excitation at a power of ~ 50 μW and a spot diameter of ~ 1 μm . (A) Scanning photocurrent image of monolayer device M2. The corresponding scanning Hall voltage image under (B) R-L and (C) L-R modulations, respectively. (D-F) are the corresponding images of bilayer device B1.

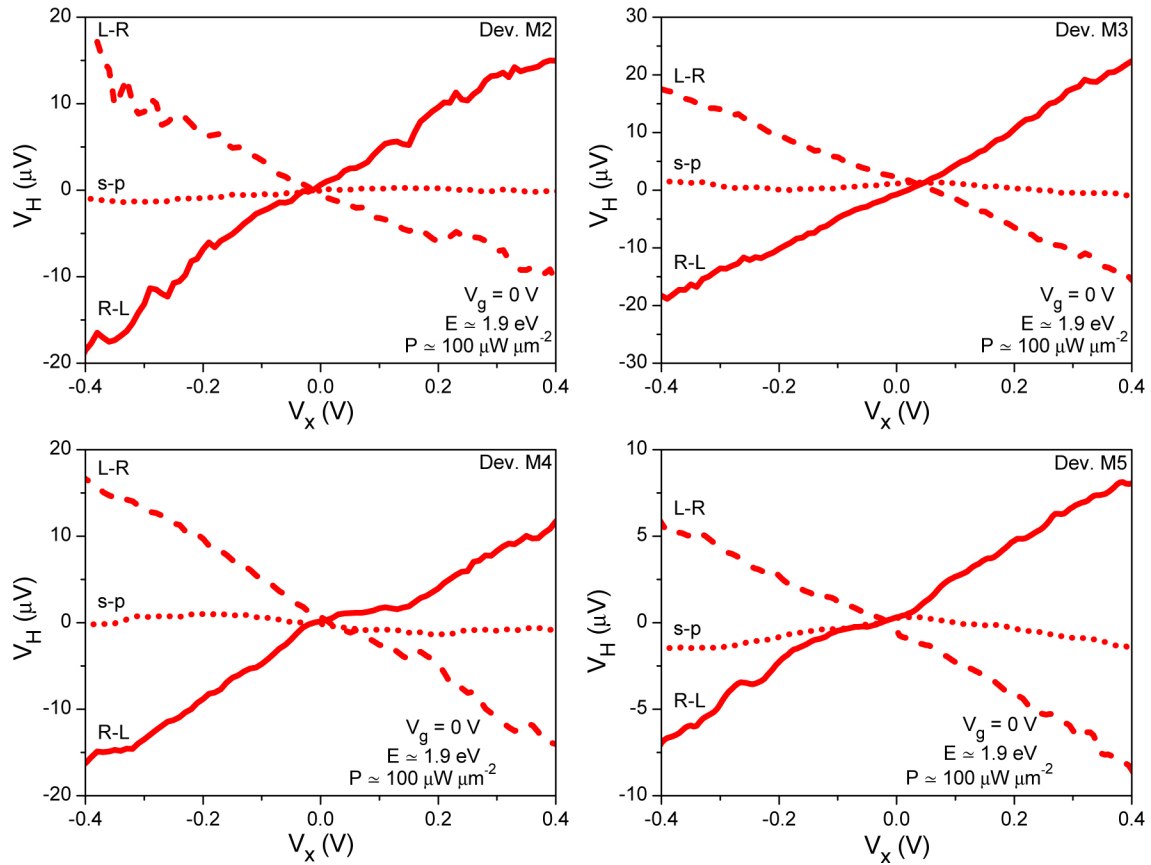


Fig. S4. Data showing the VHE in monolayer devices M2-M5. All devices were measured at gate voltage $V_g = 0$ V and under 1.9 eV excitation with intensity 100 $\mu\text{W} \mu\text{m}^{-2}$. For all devices, the source-drain bias dependence of the Hall voltage for R-L (red, solid) and L-R (red, dashed) modulations is shown, as are the results for half-wave (s-p) modulation (red, dotted).

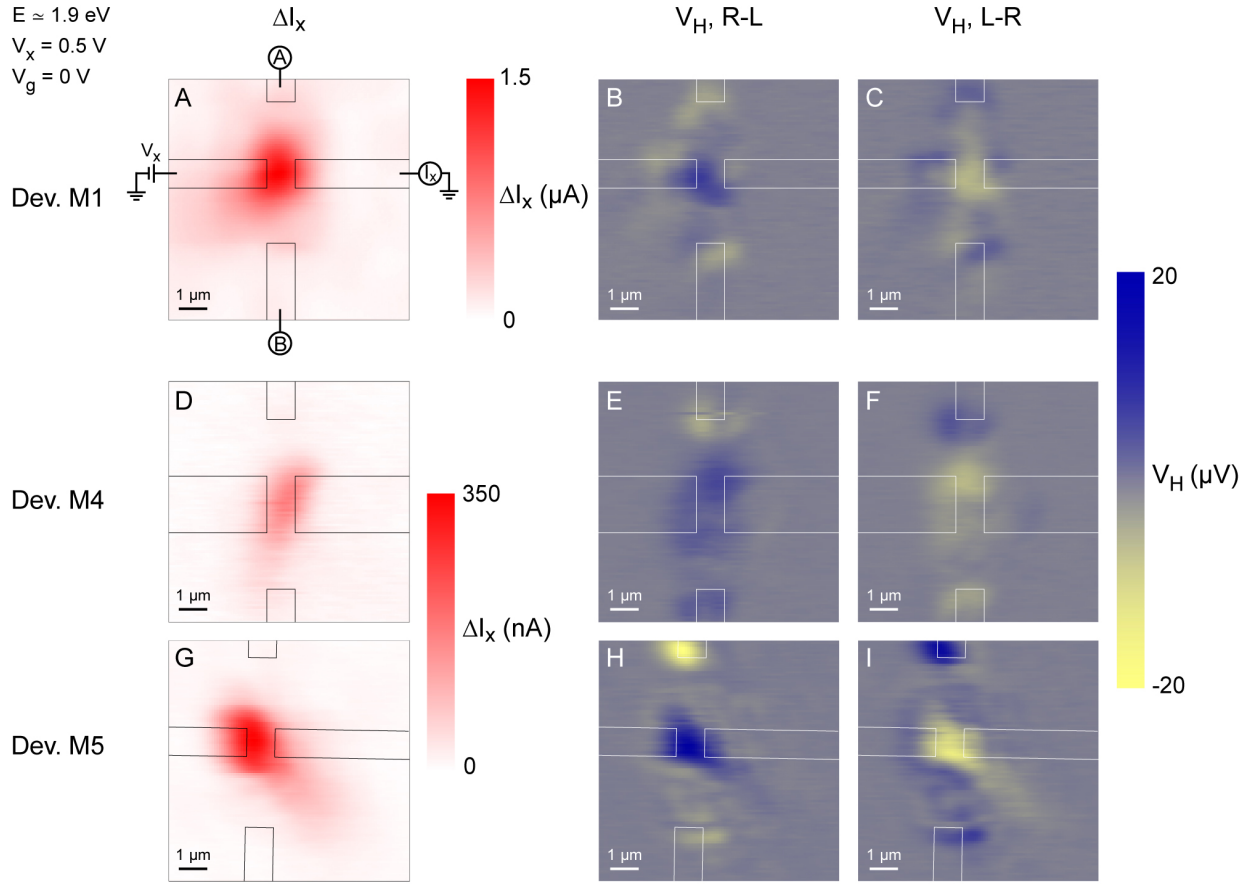


Fig. S5. Scanning photocurrent and Hall voltage maps for monolayer devices M1, M4, and M5. The measurement schematic for all maps is indicated in (A); all maps were recorded with gate voltage $V_g = 0 \text{ V}$ and source-drain bias $V_x = 0.5 \text{ V}$, under 1.9 eV excitation and intensity $50\text{--}150 \mu\text{W } \mu\text{m}^{-2}$. (A) Scanning photocurrent image of monolayer device M1. The corresponding scanning Hall voltage image under (B) R-L and (C) L-R modulations, respectively. (D-F) are the corresponding images of monolayer device M4, and (G-I) of monolayer device M5.

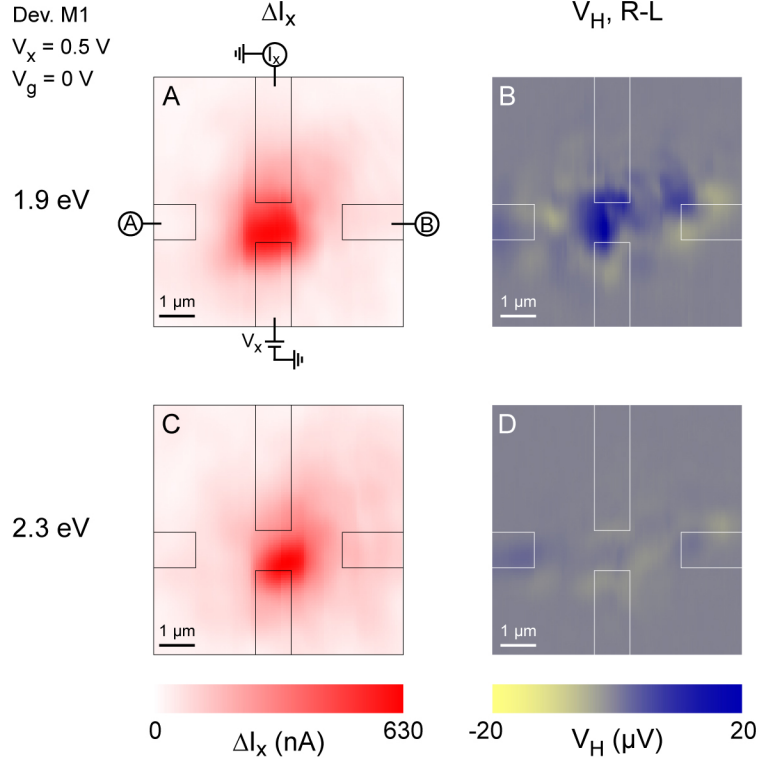


Fig. S6. Dependence of Hall voltage maps on excitation photon energy in monolayer device M1. The measurement schematic for all maps is indicated in (A); all maps were recorded with gate voltage $V_g = 0$ V and source-drain bias $V_x = 0.5$ V, under an excitation intensity of $100 \mu\text{W} \mu\text{m}^{-2}$. (A) Scanning photocurrent image of monolayer device M1 under 1.9 eV excitation. (B) The corresponding scanning Hall voltage image under R-L modulation. (C-D) are the corresponding images for 2.3 eV excitation.

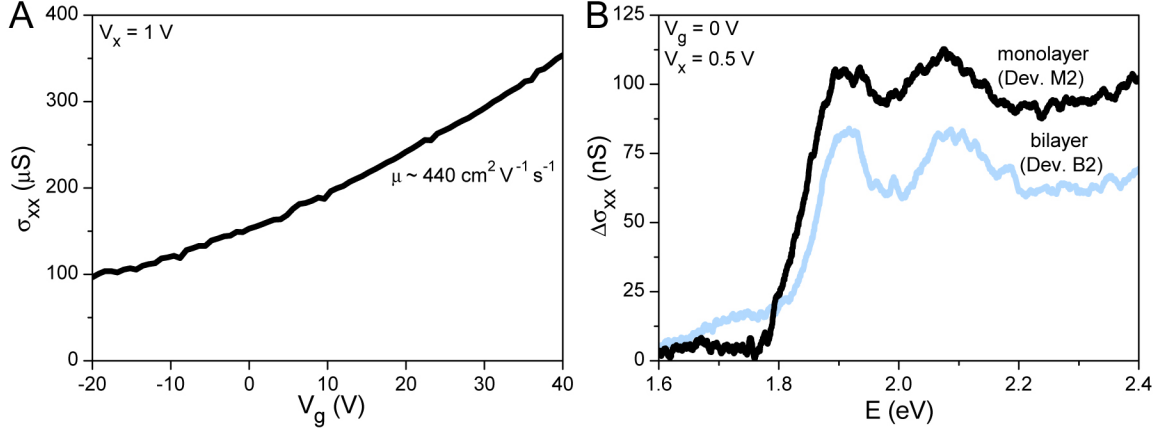


Fig. S7. Electrical characterization of bilayer device B2. (A) 4-point conductivity as a function of back gate voltage. (B) The photoconduction spectra of monolayer device M2 and bilayer device B2 as a function of incident photon energy obtained at bias voltage $V_x = 0.5$ V and gate voltage $V_g = 0$ V.

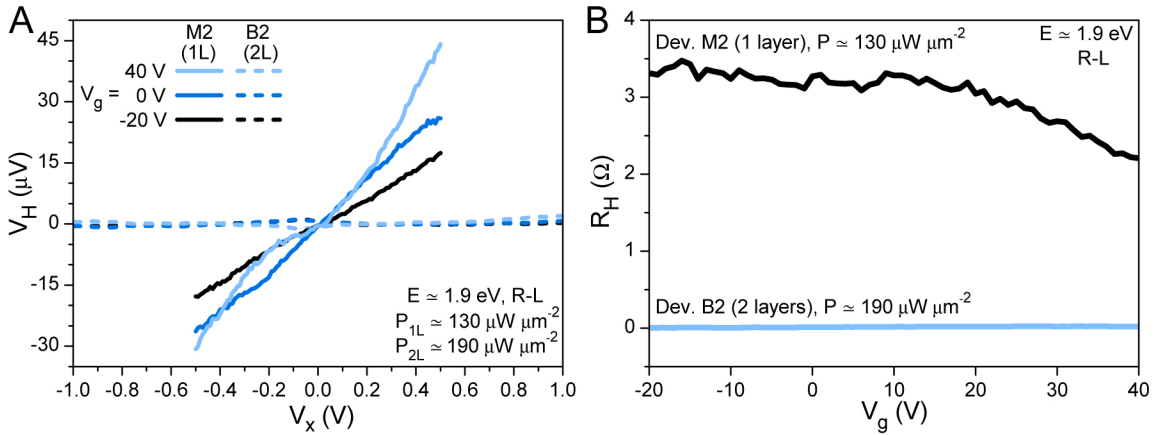


Fig. S8. Detailed comparison of monolayer device M2 and bilayer device B2. (A) The anomalous Hall voltage V_H for typical mono- and bilayer devices as a function of bias voltage V_x at different back gate voltages V_g . (B) The corresponding anomalous Hall resistances R_H as a function of back gate voltage V_g .

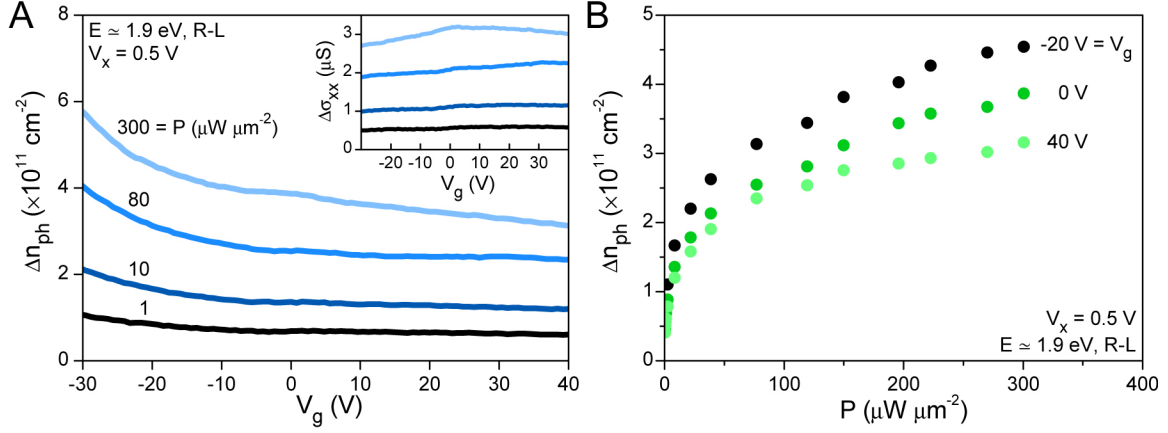


Fig. S9. Photoexcited carrier density in monolayer device M1. (A) The photoexcited carrier density Δn_{ph} as a function of gate voltage V_g at different laser excitation intensities P . The inset shows the corresponding V_g dependence of $\Delta\sigma_{xx}$ from which the carrier densities are extracted. (B) The carrier density Δn_{ph} as a function of laser intensity P at different gate voltages V_g .

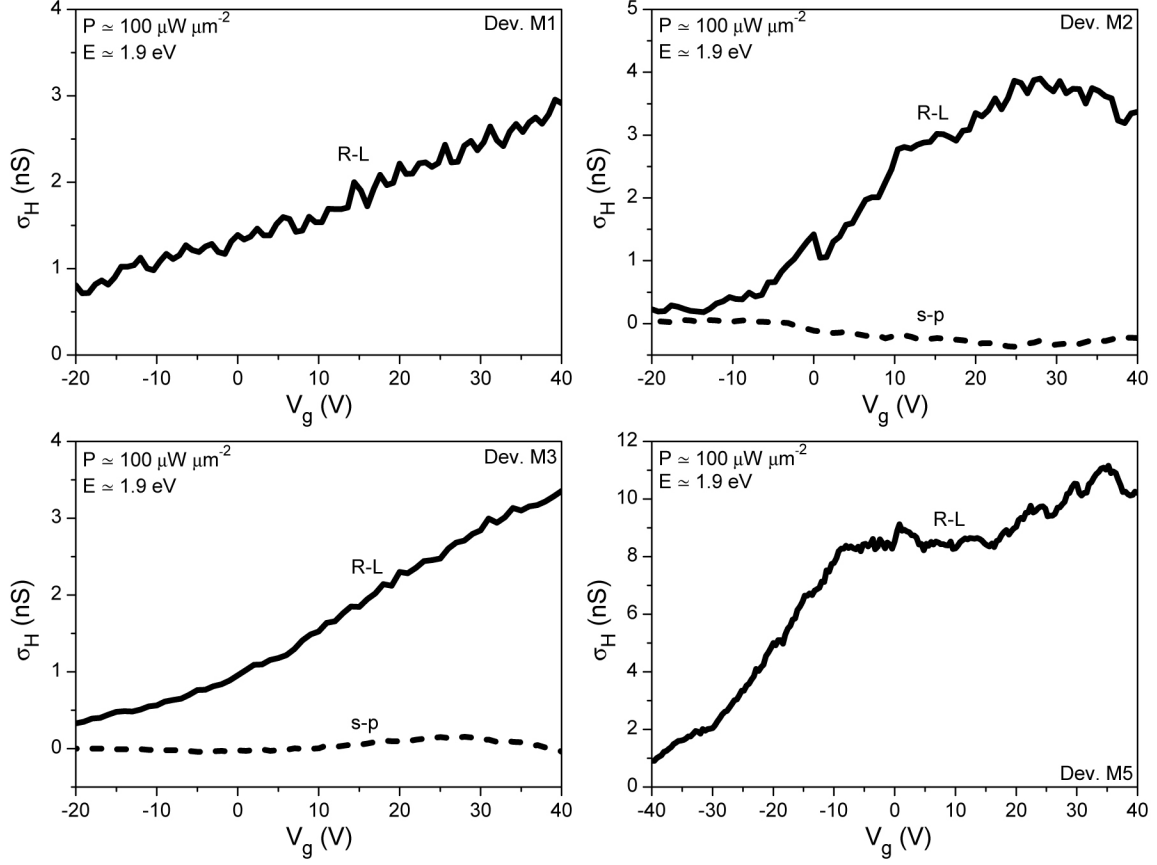


Fig. S10. Gate dependence of the anomalous Hall conductivity for monolayer devices M1-M3 and M5. All data shown was collected under 1.9 eV excitation with intensity $100 \mu\text{W } \mu\text{m}^{-2}$. We show the gate (V_g) dependence of the anomalous Hall conductivity σ_H under R-L modulation for all four devices, and we show the control results under s-p modulation for devices M2 and M4.

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23. Materials and methods are available as supplementary materials on *Science Online*.
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31. Strictly speaking, a bilayer device with slightly broken inversion symmetry by the substrate and/or by unintentional doping could also produce a finite VHE, but these effects are expected to be much smaller as compared with the VHE in monolayer devices (13).
32. Here, our assumption that only the photoexcited electrons contribute to the Hall response is reasonable because the holes are much more vulnerable to traps than are the electrons, given our highly n-doped device. Unlike the electron side, the VHE and the SHE become equivalent on the hole side owing to the spin-valley coupled valence band.
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