Intrinsic valley Hall transport in atomically thin MoS$_2$

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Electrons hopping in two-dimensional honeycomb lattices possess a valley degree of freedom in addition to charge and spin. In the absence of inversion symmetry, these systems were predicted to exhibit opposite Hall effects for electrons from different valleys. Such valley Hall effects have been achieved only by extrinsic means, such as substrate coupling, dual gating, and light illuminating. Here we report the first observation of intrinsic valley Hall transport without any extrinsic symmetry breaking in the non-centrosymmetric monolayer and trilayer MoS$_2$, evidenced by considerable nonlocal resistance that scales cubically with local resistance. Such a hallmark survives even at room temperature with a valley diffusion length at micron scale. By contrast, no valley Hall signal is observed in the centrosymmetric bilayer MoS$_2$. Our work elucidates the topological origin of valley Hall effects and marks a significant step towards the purely electrical control of valley degree of freedom in topological valleytronics.
Electron valley degree of freedom emerges as local extrema in the electronic band structures. Inequivalent valleys, well separated in the Brillouin zone, can be energetically degenerate due to symmetry and serve as novel information carriers controllable via external fields1−6. A feasible means to manipulate such a valley degree of freedom is through a valley Hall effect (VHE)7−9. Analogous to an ordinary Hall effect, in which a transverse charge current is driven by a uniform magnetic field in real space, a transverse valley current in the VHE is produced by valley-contrasting Berry curvatures in momentum space. Upon the application of an external electric field, the curvatures drive carriers from different valleys to traverse in opposite directions. Therefore, the VHE has been a major theme in the study of valleytronics, particularly in those 2D materials featuring K and K’ valleys in their hexagonal Brillouin zones10−19.

As Berry curvature is even under spatial inversion (P) and odd under time reversal (T), the VHE cannot survive when both P and T symmetries are present. To achieve VHEs in monolayer and bilayer graphene, an elaborately aligned h-BN substrate10 and a strong dual gating field11,12 were respectively utilized to break the P symmetry. To excite VHEs in specific valleys17,18, circularly polarized light20−22 were used for breaking the T symmetry in atomically thin transition-metal dichalcogenides (TMDC). Monolayer TMDCs have direct band gaps of optical frequencies at two inequivalent K-valleys23,24, due to the intrinsic P asymmetry in their unit cells depicted in Fig. 1a. Thus, Berry curvatures with opposite signs naturally emerge at the two K-valleys. Moreover, the T and mirror symmetries lock the spin and valley indices of the sub-bands split by the spin-orbit couplings, both of which are flipped under T; the spin conservation suppresses the inter-valley scattering. Therefore, monolayer TMDCs have been deemed an ideal platform for realizing intrinsic VHE without extrinsic symmetry breaking15,16.

However, the quantum transport in atomically thin TMDCs has been a long-standing challenge due to the low carrier mobility and the large contact resistance in their field-effect devices prepared by an exfoliation method. Recent breakthroughs in the fabrication of low-temperature ohmic contacts for high-mobility 2D TMDC devices25−28 have already facilitated the observation of transport hallmarks of Q-valley electrons28,29, K-valley electrons30,31, K-valley holes32−34, and Γ-valley holes35. These discoveries have revealed the rich and unique valley physics in the platform of atomically thin TMDCs.

In this work, we design nonlocal, layer-dependent, transport measurements to systematically examine the intrinsic VHEs in n-type 2H-MoS2. For the first time, we observe nonlocal resistances that exhibit cubic power-law scaling with the local resistances in the monolayers and trilayers, evidencing intrinsic VHEs. Because of the large intrinsic bandgaps and spin-valley locking of TMDCs, such VHEs can even be observed at room temperature in our monolayer devices. Beyond critical carrier densities (~4.0 × 10^{11} cm^{−2} for monolayers and trilayers), the cubic scaling turns into linear scaling. Notably, only linear scaling is observed in bilayer MoS2, where the P symmetry is restored. Intriguingly, although the monolayer and trilayer feature respectively K- and Q-valleys near their conduction-band edges, they display comparable valence-band Berry curvatures, valley Hall signatures, and micron-sized valley diffusion lengths. Our results not only offer the first experimental evidence for the intrinsic VHE but also help elucidate its topological origin6 in odd-layer TMDCs and pave the way for realizing room-temperature low-dissipation valleytronics by purely electronic means.

Results

Devices for nonlocal measurements. The structure of a monolayer MoS2 field-effect transistor is sketched in Fig. 1b. Its bright-field cross-sectional scanning transmission electron microscopy (STEM) image in Fig. 1c clearly shows the layered BN-MoS2-BN structure without any impurities in the interfaces down to the atomic scale. The device fabrication process includes a dry transfer step followed by a reactive ion etching step27,28,35 (see Methods and Supplementary Fig. 1 for details). A low contact barrier formed on the n-type MoS2 is evidenced by the 1-V curves, contact resistances (Supplementary Fig. 2), and the field-effect mobilities μ varied from 500−4000 cm^{2}V^{−1}s^{−1} for monolayers, 4000−23000 cm^{2}V^{−1}s^{−1} for bilayers, and 10000−25000 cm^{2}V^{−1}s^{−1} for trilayers at T = 2 K (Supplementary Figs. 3 and 4). The impurity-free STEM images and the high mobilities coincide well with the low residue carrier densities (n* ≈ 4 × 10^{10} cm^{−2}, see Supplementary Fig. 5).

As for the electronic measurement, an inverse VHE is exploited to detect a valley current, as sketched in Fig. 1d. An applied current I_{B1} through probes 1 and 2 induces charge imbalance in a remote region, as measured by the voltage drop V_{AA} between probes 3 and 4 (Supplementary Fig. 6). The nonlocal resistance R_{NL} = V_{AA}/I_{B2} mediated by the valley Hall current was predicted36 to present cubic power-law dependence on the local resistance R_{L} = V_{AA}/I_{B1}.

Nonlocal transport in monolayer MoS_{2}. Nonlocal resistance R_{NL} in a n-type monolayer MoS_{2} (sample B of length L = 6 μm and width W = 1.5 μm inscribed in the inset of Fig. 1e), measured as a function of gate voltage V_{g} at varied temperatures, is shown in Fig. 1e. A giant R_{NL} is observed in the range of V_{g} − −15 to −25 V that amounts to the electron density n ≈ 10^{10} to 10^{11} cm^{−2}. In particular, the observed R_{NL} ≈ 10^{6} Ω exceeds the classical ohmic contribution R_{CL} = R_{L} = μ_{g} e^{−V_{g}/W} ≈ 10^{4} Ω by two orders of magnitude in the range of V_{g} = −15 to −18 V at 2 K and V_{g} = −22 to −25 V at 300 K. Another unexpected feature of R_{NL} is its V_{g} dependence. In sharp contrast to the classical contribution R_{CL}, which decreases gradually with increasing V_{g}, the observed R_{NL} drops by at least one order of magnitude within an increase of several volts in V_{g}. Both the pronounced nonlocal signal and its unusual sensitivity to V_{g} suggest that the observed R_{NL} has a physical origin different from the classical ohmic contribution R_{CL}.

The temperature dependence of R_{L} and R_{NL} uncovers the mesoscopic mechanism of both the local and nonlocal transport. The conduction can be separated into three regimes: the thermal activation (TA) at 250 K > T > 130 K, the nearest-neighbor hopping (NNH) at 130 K > T > 60 K, and the variable-range hopping (VRH) below 60 K (sample A of L = 3.6 μm and W = 1.5 μm, see Fig. 2f and Supplementary Fig. 7a and 7b). These transport regimes are consistent with previous studies37,38. Since pronounced nonlocal signals are observed in all three transport regimes, there appears no clear connection between the transport regimes and the onset of strong nonlocal signals. Interestingly, the characteristic temperatures of both NNH and VRH for R_{NL} are much larger than those for R_{L} in the range of V_{g} ≈ −60 to −58 V (Supplementary Fig. 7d and 7e). This indicates a higher energy barrier in the nonlocal transport and an anomalous origin of the nonlocal signal.

To determine the origin of the observed R_{NL}, we investigate the scaling relation between R_{NL} and R_{L} as functions of V_{g} at different temperatures for both sample A (Fig. 2) and sample B (Supplementary Fig. 8). For a fixed V_{g}, both R_{L} and R_{NL} increase when the temperature is lowered. In sample A, two regimes with distinct scaling behaviors become clearly visible in Fig. 2c, d, the logarithmic plot of R_{L} and R_{NL} at different V_{g}. Above 160 K, the slopes of the InR_{NL} versus InR_{L} curves are 1, indicating that R_{NL} ∝ R_{L}. Below 160 K, the slopes turn to 3 in the low electron density regime (R_{L} ≈ 10^{8} to 10^{9} Ω), which amounts to R_{NL} ∝ R_{L}^{3}.

Indeed, a diffusive model has predicted such power-law
pristine and free of impurities down to the atomic scale (scale bar 3 nm).

Nonlocal transport. The applied charge current in the left circuit generates a pure valley current in the transverse direction via a VHE. This valley current is inversion asymmetric (symmetric).

sandwiched between the top and bottom h-BN layers.

Dirac band structure of monolayer MoS2 produces large valley splitting (estimated as \( E_s/k_B \sim 169 \text{ K} \), see Supplementary Fig. 9) at high temperatures. Below 160 K, Eq. (1) can be employed to estimate \( l_V \). For the case of intermediate inter-valley scattering and edge roughness, \( l_V \sim 0.36 \mu m \) if we assume \( \sigma_{xy}^V \sim 1 e^2/h \). In the limit of strong inter-valley scattering and edge roughness, \( l_V \sim 0.43 \mu m \) if we assume \( \sigma_{xy}^V \sim 0.1 e^2/h \). These values of \( l_V \) are comparable to those obtained in graphene systems.

We further investigated the length dependence of the nonlocal valley transport. Apart from sample A \((L = 3.6 \mu m)\) and sample B \((L = 6 \mu m)\), two more samples \((L = 11 \mu m\) and \(16 \mu m)\) are investigated (Supplementary Fig. 8). The semilog plot of \( R_{NL} \) at \( n = 4 \times 10^{11} \text{ cm}^{-2} \) (extracted from the Hall measurement, see Supplementary Fig. 10) versus the sample length yields an estimate of \( l_V \sim 1 \mu m \) (Fig. 2g). This value is very close to \( W \) and much larger than the electron mean-free path \( l_m \sim 20 \text{ nm} \) (estimated from the sample mobility \( \mu \) for the range of \( n \) where the cubic scaling appears) and the localization length \( \xi \sim 50 \text{ nm} \).

relations (see below) but much weaker spin Hall effect (see Discussion).

The \( R_{NL} \) and \( R_{e} \) data measured at different temperatures for the case of \( V_g = −60 \text{ V} \) are plotted in Fig. 2d. The cubic law is not applicable above 160 K, due to the enhancement of inter-valley scattering by the smear of the lowest conduction sub-band spin splitting (estimated as \( E_s/k_B \sim 169 \text{ K} \), see Supplementary Fig. 9) at high temperatures. Below 160 K, Eq. (1) can be employed to estimate \( l_V \). For the case of intermediate inter-valley scattering and edge roughness, \( l_V \sim 0.36 \mu m \) if we assume \( \sigma_{xy}^V \sim 1 e^2/h \). In the limit of strong inter-valley scattering and edge roughness, \( l_V \sim 0.43 \mu m \) if we assume \( \sigma_{xy}^V \sim 0.1 e^2/h \). These values of \( l_V \) are comparable to those obtained in graphene systems.

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Nonlocal transport in bilayer and trilayer MoS2. For bilayer MoS2, the measured $R_L$ and $R_{NL}$ as functions of $V_g$ at different temperatures are plotted in Fig. 3a, b. As the carrier density increases, $R_L$ and $R_{NL}$ decrease in a similar fashion in the temperature range of 5–50 K. This yields a linear scaling behavior between $R_L$ and $R_{NL}$, as analyzed in Fig. 3c, d, and no cubic scaling is detected. We note that extrinsic $P$ symmetry breaking can be introduced into atomically thin bilayers via external gating, as achieved in bilayer graphene[11,12], and that detecting a nonlocal signal in gated bilayer graphene requires a threshold gating strength[11,12]. In our devices, however, $V_g$ is too low to reach the threshold estimated by an recent optical experiment[18], the estimated potential difference between the top and bottom layers is $\sim 9.2$ meV at $V_g = -60$ V. This weak symmetry breaking produces little change in the total Berry curvature as compared with the pristine case (Supplementary Fig. 12), given the facts that the induced potential is much smaller than the bandgap and that the valence-band contribution to $\sigma_{xy}^b$ is dominant. In light of this analysis, the gating-induced $P$ symmetry breaking is negligible in our bilayer MoS2. Therefore, we conclude that the absence of cubic scaling in bilayer MoS2 indicates the crucial role of strong $P$ symmetry breaking in generating VHE. This is consistent with the theoretical understanding of VHE[5–9], as aforementioned in Introduction.

This key conclusion can be immediately tested in thicker MoS2 samples. Given that $P$ symmetry is broken (respected) in pristine odd-layer (even-layer) MoS2, one might wonder whether the intrinsic VHE and its cubic scaling could be detected in trilayer MoS2. Figure 3e, f display our $R_L$ and $R_{NL}$ data measured in trilayer MoS2 as functions of $V_g$ at different temperatures. Evidently, the measured $R_{NL}$ rapidly decreases as $V_g$ increases in the narrow range of $-20 \text{ V} < V_g < -18.4 \text{ V}$, which is reminiscent of the behavior of $R_{NL}$ in our monolayer devices in the low density regime. Similar to the monolayer case, the logarithmic
plots of $R_L$ and $R_{NL}$ in Fig. 3g exhibit clear changes in slope from 1 to 3 near $V_g = -18.4 \text{ V}$, further confirming the observation of the nonlocal signal of VHE in trilayer MoS$_2$. To illustrate the temperature dependence, Fig. 3h plots the scaling relation between $R_L$ and $R_{NL}$ at different temperatures for the case of $V_g = -20 \text{ V}$. Again, there is a clear change in slope from 1 to 3 near 30 K. Moreover, the valley diffusion length can be extracted based on Fig. 3h and Eq. (1). We obtain $\nu_L \sim 0.5 \text{ pm}$ and $\sim 1 \text{ pm}$, respectively, for the aforementioned two limits $\sigma_{xx}^L \sim 1 e^2/h$ and $\sim 0.1 e^2/h$. Both the observed amplitude of nonlocal signal and the estimated valley diffusion length in the trilayer MoS$_2$ devices are comparable to those in the monolayer case. In addition to the crucial role of $P$ symmetry breaking, significantly, these observations are suggestive of a universal physical origin of VHEs in odd-layer TMDCs, as discussed below.

**Layer-dependent Berry curvatures.** To better understand the thickness dependent observations, we calculate the electronic band structures and Berry curvatures$^{15,16}$ for monolayer, bilayer, and trilayer MoS$_2$. The band structures in Fig. 4a, c are indeed thickness dependent. In particular, the conduction-band minima lie at the K-valleys for the monolayer, whereas they shift to the Q-valleys for the bilayer and trilayer. Given the low electron densities in our samples ($\sim 4 \times 10^{11} \text{ cm}^{-2}$ in monolayers and trilayers, $\sim 1 \times 10^{12} \text{ cm}^{-2}$ in bilayers), the Fermi levels only cross the lowest conduction sub-bands, as indicated by the green lines in Fig. 4a, c. As bilayer MoS$_2$ has a restored $P$ symmetry that is intrinsically broken in odd-layer MoS$_2$, the sub-bands are spin degenerate in the bilayer yet spin split in the monolayer and trilayer. With these band structures, we further compute the Berry curvatures that drive the VHEs. Berry curvature vanishes if both $P$ and $T$ are present. As plotted in Fig. 4d, f, our calculations reveal that the curvatures are indeed trivial in the bilayer yet substantial in the monolayer and trilayer. This explains the reason why no cubic scaling is observed in bilayer MoS$_2$ and highlights the role of $P$ symmetry breaking in producing VHEs.

It is puzzling to understand and compare the nonlocal signals of VHEs in monolayer and trilayer MoS$_2$. Similar cubic scaling behaviors and their transitions to linear ones above the critical densities or temperatures are observed in both cases. However, the conduction-band Berry curvatures (the difference between the blue and orange curves in Fig. 4d, f) because of the extremely small valence-band contributions are different but very minor (the difference between the blue and orange curves in Fig. 4d, f), due to the extremely weak interlayer couplings. By contrast, the conduction-band contributions are different but very minor (the difference between the blue and orange curves in Fig. 4d, f) because of the low electron densities. Therefore, the valence-band contributions dominate the valley Hall conductivities, leading to similar nonlocal signals of VHEs in monolayer and trilayer. Recently, nearly quantized edge transports have been observed along the designed or selected domain walls in graphene systems$^{13,14}$ and
Fig. 4 Band structures and Berry curvatures of atomically thin MoS$_2$. **a-c** Band structure of (**a**) monolayer, (**b**) bilayer, and (**c**) trilayer MoS$_2$. The conduction band edges lie at the K valleys in the monolayer but at the Q valleys in the bilayer and trilayer. Insets of **a-c**: The Fermi levels only cross the lowest sub-bands, which are spin degenerate in total curvatures of all valence-band states. The red arrow in curves are the total curvatures of all occupied states below the Fermi levels (~2 meV from the conduction band bottom), whereas the orange curves are the total curvatures of all valence-band states. The red arrow in **f** points out a tiny bump at a Q valley. Insets of **d-f** 2D mapping of Berry curvatures in the 2D Brillouin zone (white dashed lines)

Discussion

Finally, we note that the VHE and spin Hall effects are distinct in TMDCs, in spite of the spin-valley locking. The spin-valley locking is a property at Fermi level only when it lies in the lowest conduction or highest valence sub-band. Yet, all states below Fermi level contribute to the spin and valley Hall conductivities (see Supplementary Note 1). Although a similar line of analysis based on Eq. (1) can be done for a theoretical hypothesis of spin Hall effect as well, it appears that this is not the case for three reasons. First, the spin Hall conductivities are predicted to be very small for pristine odd-layer TMDCs when the valence bands are degenerate in spin and valley symmetries (T). Nonlocal resistances have little response to a magnetic field up to 9 T (Supplementary Fig. 13). Third, the spin diffusion length in TMDCs is at the scale of several tens of nanometers ($\approx$ 100 nm) via van der Waals interactions. The h-BN/MoS$_2$ flake is then transferred onto a fresh thick h-BN substrate, to form a BN-MoS$_2$ heterostructure (step 1 in Supplementary Fig. 1).

Layer numbers and stacking orders. To determine the number of layers for a MoS$_2$ sample, we carried out microRaman and photoluminescence measurements before making a device (Supplementary Fig. 14). We also took cross-sectional STEM (JEOL JEM-ARM200F Cs-corrected TEM, operating at 60 kV) images after the electronic measurement. The STEM images can clearly determine the number of MoS$_2$ layers (Supplementary Fig. 14) and distinguishes the 2H stacking order from other stacking orders such as 1T and 3R (Supplementary Fig. 15).

Selective etching process. A hard mask is patterned on the heterostructure by the standard e-beam lithography technique using PMMA (step 1 in Supplementary Fig. 1). The exposed top BN layer and MoS$_2$ are then etched via reactive ion etching (RIE), forming a Hall bar geometry (steps 3 & 4 in Supplementary Fig. 1). Then a second-round e-beam lithography and RIE is carried out to expose the MoS$_2$ layer (steps 5 & 6 in Supplementary Fig. 1). The electrodes are then patterned by a third-round e-beam lithography followed by a standard e-beam evaporation (steps 7 & 8)
Electronic measurement. The I–V curves are measured by Keithley 6430. Other transport measurements are carried out by using (i) low-frequency lock-in technique (SR 830 with SR550 as the preamplifier and DS 360 as the function generator, or (ii) Keithley 6430 source meter (>10^12 Ω input resistance on voltage measurements). The cryogenic system provides stable temperatures ranging from 1.4 to 300 K. A detailed discussion of the nonlocal measurement is presented in Supplementary Fig. 6.

Data availability
The authors declare that the major data supporting the findings of this study are available within the paper and its Supplementary Information. Extra data are available from the authors upon reasonable request.

Received: 24 April 2018 Accepted: 23 January 2019
Published online: 05 February 2019

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Author contributions
Z.W. and N.W. conceived and designed the experiments. F.Z. provided the theoretical support. N.W. and F.Z. supervised the work. Z.W. fabricated the devices, performed the measurements, and analyzed the data with the help from M.H., J.L., T.H., L.A., Y.W., S.X., G.L., C.C. and K.T.L. X.C. carried out the STEM characterizations. G.-B.L. comments on the spin Hall effect regime. Phys. Rev. B 79, 035304 (2009).

Acknowledgements
We acknowledge the financial support from the Research Grants Council of Hong Kong (Project Nos. 16300717, 16302215, 16324216, C7036-17W, C6026-16W, and HKUST3/ CRF13G), the Croucher Foundation, the Dr. Taichin Lo Foundation, the National Key R&D Program of China (with grant number 2017YFB0701600), US Army Research Office (under grant number W911NF-18-1-0416), and UT-Dallas Research Enhancement Funds. We acknowledge the technical support from Wing Ki Wong and the RAITH-HKUST Nanotechnology Laboratory at MCPF. Z.W. acknowledges useful conversations with Danru Qu. F.Z. is grateful to Allan MacDonald, Joe Qu, and Di Xiao for valuable discussions.

Additional information
Supplementary Information accompanies this paper at https://doi.org/10.1038/s41467-019-08629-9.

Competing interests: The authors declare no competing interests.

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Journal peer review information: Nature Communications thanks the anonymous reviewers for their contribution to the peer review of this work.

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