

Switchable moiré potentials in ferroelectric $\text{WTe}_2/\text{WSe}_2$ superlattices

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Moiré materials with superlattice periodicity many times the atomic length scale have shown strong electronic correlations and band topology with unprecedented tunability. Non-volatile control of the moiré potentials could allow on-demand switching of superlattice effects but has remained challenging to achieve. Here we demonstrate the switching of the correlated and moiré band insulating states, and the associated nonlinear anomalous Hall effect, by the ferroelectric effect. This is achieved in a ferroelectric WTe_2 bilayer of the T_d structure with a centred-rectangular moiré superlattice induced by interfacing with a WSe_2 monolayer of the H structure. The results can be understood in terms of polarization-dependent charge transfer between two WTe_2 monolayers, in which the interfacial layer has a much stronger moiré potential depth; ferroelectric switching thus turns on and off the moiré insulating states. Our study demonstrates the potential for creating new functional moiré materials by incorporating intrinsic symmetry-breaking orders.

Two-dimensional ferroelectric materials have attracted much recent interest due to their potential in non-volatile memory device applications¹. In addition to atomically thin ferroelectrics^{2–7}, such as SnTe , CuInP_2S_6 and $T_d\text{-WTe}_2$, ferroelectricity has also been realized in moiré heterostructures^{8–13}, enabling non-volatile control of correlated electronic states and superconductivity. However, non-volatile electrical switching of the moiré trapping potential for electrons and the associated superlattice effects remains a challenge. Here we achieve this goal by utilizing the intrinsic ferroelectricity in angle-aligned bilayer $T_d\text{-WTe}_2$ /monolayer H- WSe_2 moiré heterostructures.

Monolayer WTe_2 of the T_d crystal structure consists of a layer of tungsten atoms between two layers of tellurium atoms in a distorted octahedral coordination¹⁴. The structure is centrosymmetric with a mirror line along the crystal b axis. At charge neutrality, the material is a quantum spin Hall insulator^{15–17}. Bilayer $T_d\text{-WTe}_2$ comprises two anti-parallel monolayers; the structure becomes non-centrosymmetric with a polar axis¹⁸ (inclined from the out-of-plane direction). The presence of the polar axis stabilizes ferroelectricity⁴ and generates Berry curvature dipoles in the momentum space¹⁹ responsible for the nonlinear

anomalous Hall effect (NAHE)^{20–22}. Bilayer $T_d\text{-WTe}_2$ is a topologically trivial insulator. Figure 1c illustrates its schematic band structure with a flat valence band centred at the Γ point of the Brillouin zone and dispersive conduction bands away from the Γ point^{23,24}. The bands are layer-polarized by spontaneous out-of-plane electric polarization.

Ferroelectric moiré heterostructure

We introduce moiré potentials to bilayer WTe_2 by adding a WSe_2 monolayer on it^{25,26} (Fig. 1a). The WSe_2 monolayer is of the H crystal structure, in which the tungsten atoms are between the selenium atoms in the trigonal prismatic coordination. The zigzag crystal axis of WSe_2 is aligned to the crystal a axis of WTe_2 . A nearly centred-rectangular moiré superlattice is expected with periods 6.9 nm and 6.4 nm along the a and b axes of WTe_2 , respectively. These values are estimated from the lattice mismatch between the two materials (5% and 9%) along the two axes. The corresponding moiré density, n_M , is about $4.5 \times 10^{12} \text{ cm}^{-2}$. These estimates agree with the experimental moiré pattern of a typical aligned $\text{WTe}_2/\text{WSe}_2$ heterostructure obtained by piezoresponse force microscopy (PFM) (Fig. 1b; see Methods and Extended Data Fig. 1 for

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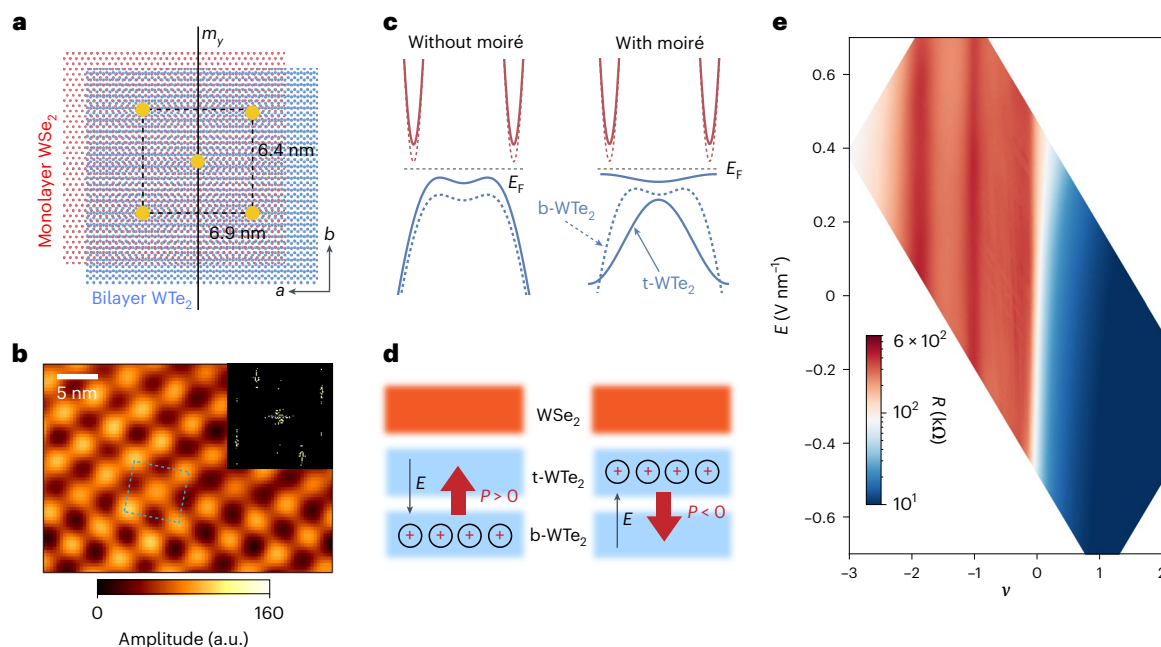


Fig. 1 | Bilayer T_d -WTe₂/monolayer H-WSe₂ moiré heterostructure.

a, Moiré superlattice for angle-aligned bilayer T_d -WTe₂/monolayer H-WSe₂ heterostructure. Only the tungsten atoms are shown. Yellow dots denote the high-symmetry MM (tungsten on tungsten) sites, which form a centred-rectangular lattice with lattice constants of 6.9 nm along the crystal a axis of WTe₂ and 6.4 nm along the crystal b axis of WTe₂ (m_y is the mirror line). The dashed rectangle marks the moiré unit cell. The black arrows mark the crystal a and b axes of bilayer WTe₂. **b**, PFM image of a typical moiré heterostructure. The dashed rectangle marks the moiré unit cell. Inset: Fourier transform of the PFM image; there are four diffraction peaks corresponding to the centred-rectangular moiré lattice. **c**, Schematics for the bilayer WTe₂ band structure with and without

moiré effects. Red and blue curves correspond to conduction and valence bands, respectively. As shown in the right-hand panels, moiré bands are formed only on the top WTe₂ (t-WTe₂) layer (solid blue curves). The bottom WTe₂ (b-WTe₂) layer experiences substantially weaker moiré potential; the bands are dispersive (dashed curves). The horizontal grey dashed line denotes the Fermi level. **d**, Polarization-dependent hole (red cross) distributions in bilayer WTe₂. P and E are, respectively, the spontaneous polarization and its corresponding internal electric field. **e**, Electric-field and filling-factor dependence measured at 30 K for the longitudinal resistance of angle-aligned monolayer T_d -WTe₂/monolayer H-WSe₂ heterostructure.

details). The two lattice vectors form an angle of about 84°. The moiré lattice is robust and insensitive to twist-angle variations in the sample as a result of the large lattice mismatch. The nearly centred-rectangular lattice also contrasts with the triangular and honeycomb moiré lattices examined by most existing studies to date^{27–31}.

The heterostructure has a type I band alignment with both the conduction band minimum and valence band maximum from bilayer WTe₂ (ref. 32). The WSe₂ layer remains charge-neutral at all times; it only provides a moiré potential responsible for the flat bands in the WTe₂ layer. Figure 1c shows the schematic band structure, including the zone-folding effect on the bilayer WTe₂ bands²⁰. The top WTe₂ layer directly interfacing with WSe₂ is expected to experience a substantially stronger moiré potential than the bottom WTe₂ layer; the moiré bands for the top layer are expected to be much flatter. Such layer-sensitive moiré effects in bilayer WTe₂, combined with its intrinsic ferroelectric order, form the basis for ferroelectric control of the moiré potential in this study.

Below we demonstrate the effects of ferroelectric switching on the correlated insulating states and the Berry curvature dipole distribution in the correlated flat bands. The heterostructure is encapsulated between top and bottom boron nitride and graphite gates for independent controls of the charge carrier density ν (in units of n_M) and the electric field E perpendicular to the sample plane (positive field defined as pointing from WTe₂ to WSe₂). See Methods for details on device fabrications, calibration of moiré density and measurements.

Switchable moiré potential by ferroelectricity

To contrast the behaviour of bilayer WTe₂ devices, we first examine the density- and electric-field-dependent transport properties in

an angle-aligned monolayer WTe₂/monolayer WSe₂ heterostructure (Fig. 1e). Because monolayer WTe₂ is a quantum spin Hall insulator^{15–17}, we fabricate an edge-avoiding device to access the bulk resistivity of the material (Methods and Extended Data Fig. 2). We only perform measurements down to 30 K because the sample resistance increases substantially below this temperature. In Fig. 1e, we observe resistance peaks at filling factors of $\nu = 0, -1, -2$. They correspond to the band insulating state when WTe₂ is at charge neutrality ($\nu = 0$), the moiré band insulating state when the first moiré valence band is fully filled with holes ($\nu = -2$), and the correlated insulating state (probably a Mott insulator) for a half-filled moiré band ($\nu = -1$). The insulating states at $\nu = -1, -2$ confirm the formation of flat moiré valence bands in monolayer WTe₂ (see Extended Data Fig. 3 for results on other devices). In contrast, no insulating state is observed at $\nu = 1, 2$ on the electron doping side (see also Figs. 2 and 3). The result demonstrates the substantially weaker moiré effects on the conduction bands, possibly due to the small conduction band mass ($\sim 1/3$ of the valence band mass) in WTe₂ (refs. 20,24,33) and the small moiré periods in the WTe₂/WSe₂ heterostructure. Moreover, all observed insulating states show weak and non-hysteretic electric field dependence, consistent with the absence of ferroelectricity in monolayer WTe₂ (ref. 4).

Next, we examine the density- and electric-field-dependent resistance in a bilayer WTe₂ device under forward (from negative to positive) and backward (from positive to negative) scans of the electric field in Fig. 2a,b, respectively. The edge-avoiding geometry is no longer needed because bilayer WTe₂ is a topologically trivial insulator^{23,24}. Lower-temperature measurements are also possible. Similar to the monolayer device, we observe insulating states at $\nu = -1, -2$ for hole doping, a charge-neutral insulating state at $\nu = 0$, and no

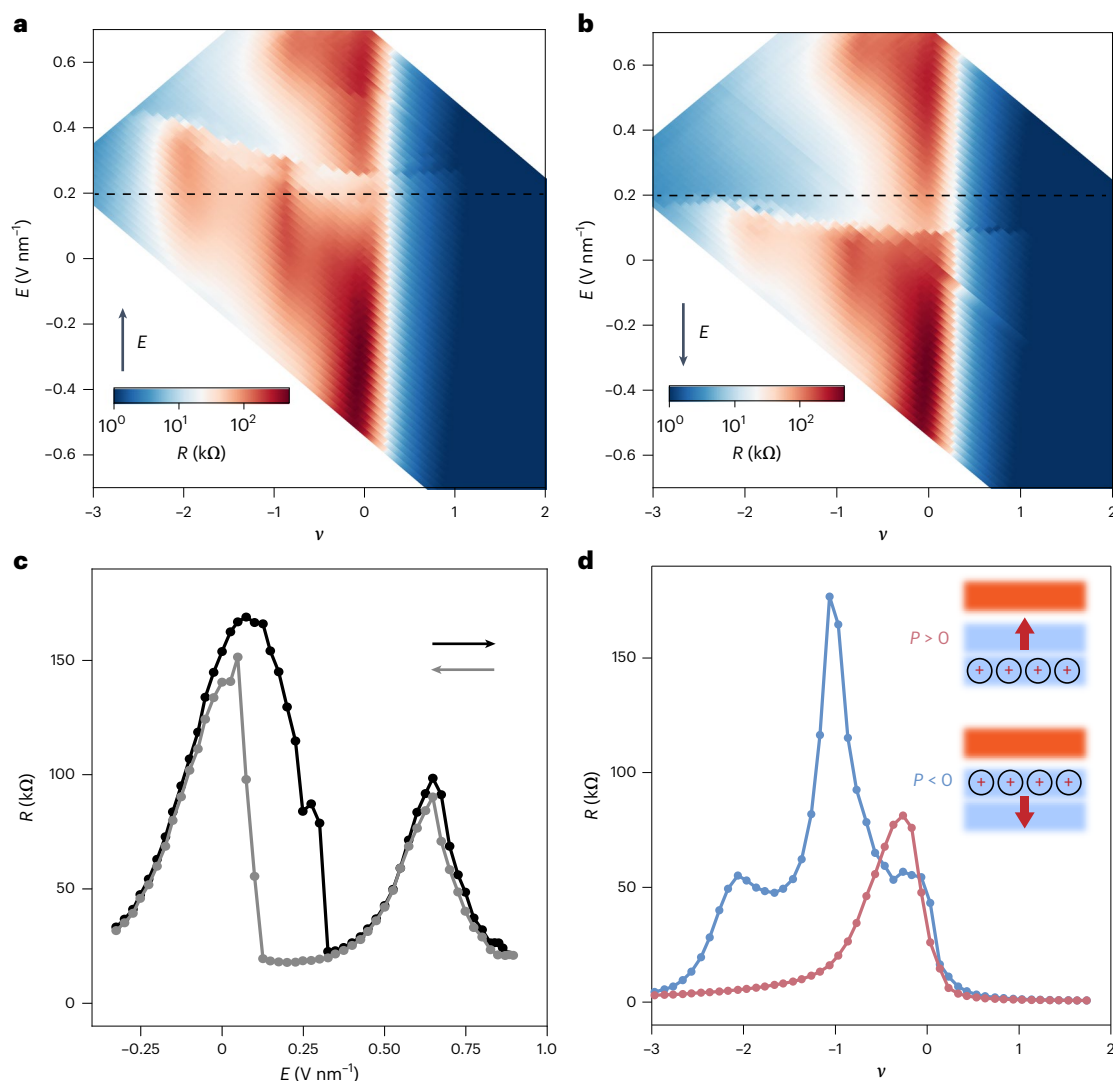


Fig. 2 | Switching moiré potential by ferroelectricity. **a, b**, Electric-field and filling-factor dependence for the longitudinal resistance of an angle-aligned bilayer $T_d\text{-WTe}_2$ /monolayer H-WSe_2 heterostructure at 10 K. The black arrows label the forward (**a**) and backward (**b**) electric field scan directions. Hysteretic electric-field dependence corresponding to ferroelectric switching is observed.

c, Electric-field dependence of the longitudinal resistance at $\nu = -1$. The arrows mark the field scan directions. **d**, Filling-factor-dependent longitudinal resistance extracted from **a** (blue) and **b** (red) at $E = 0.2$ $V\text{ nm}^{-1}$ (the black dashed lines in **a** and **b**). They denote the two spontaneous polarization states $P > 0$ and $P < 0$ (inset). Moiré insulating states are observed only for $P < 0$.

moiré insulating states for electron doping. However, unlike the monolayer device, strong electric-field dependence is observed. In particular, robust insulating states at $\nu = -1, -2$ are observed mainly for $E \lesssim 0.3$ $V\text{ nm}^{-1}$ in the forward scan (Fig. 2a) and for $E \lesssim 0.1$ $V\text{ nm}^{-1}$ in the backward scan (Fig. 2b). A sharp resistance jump and an electric-field hysteresis are also observed near these critical fields, as exemplified by the electric-field-dependent resistance at $\nu = -1$ in Fig. 2c (similar data at other filling factors are shown in Extended Data Fig. 4). (The extra resistance step near $E = 0.25$ $V\text{ nm}^{-1}$ is probably caused by the formation and propagation of ferroelectric domains in the bilayer WTe_2 (refs. 6,34,35).) In addition to the hysteresis, a non-monotonic electric field dependence is also observed (the origin is discussed in Methods). The results demonstrate the effects of ferroelectric switching (in bilayer WTe_2) on the moiré physics. The centre of the ferroelectric switching is shifted from 0 $V\text{ nm}^{-1}$ to -0.2 $V\text{ nm}^{-1}$ and disperses with hole doping because of the built-in electric field in the asymmetric heterostructure (WSe_2 is on top of WTe_2).

We further illustrate the effects of ferroelectric switching by plotting the density-dependent resistance near the centre of the hysteresis

(at fixed $E = 0.2$ $V\text{ nm}^{-1}$) for both forward and backward field scans (Fig. 2d). The two cases represent the two remnant states of spontaneous polarization P in the ferroelectric WTe_2 : positive polarization ($P > 0$, pointing from WTe_2 to WSe_2) for backward scan and negative polarization ($P < 0$, pointing from WSe_2 to WTe_2) for the forward scan. Insulating states at $\nu = -1, -2$ associated with the moiré valence band are observed only for $P < 0$; only the $\nu = 0$ insulating state remains for $P > 0$. The charge neutrality point is also nearly independent of the sign of P , showing the negligible change in the total doping density by ferroelectric switching.

The results above can be understood based on the interlayer transfer of mobile holes in bilayer WTe_2 driven by its spontaneous ferroelectric polarization, which originates from the electric-field-switchable ionic displacements in the material^{6,36}. When positive polarization ($P > 0$) is prepared, the internal electric field from the displaced ions (or bound charges) within the bilayer WTe_2 is negative; the field splits the electronic bands of the top and bottom WTe_2 layer via the Stark effect and pushes the mobile holes to the bottom layer (Fig. 1d). The reverse happens when negative polarization ($P < 0$) is prepared (Fig. 1d).

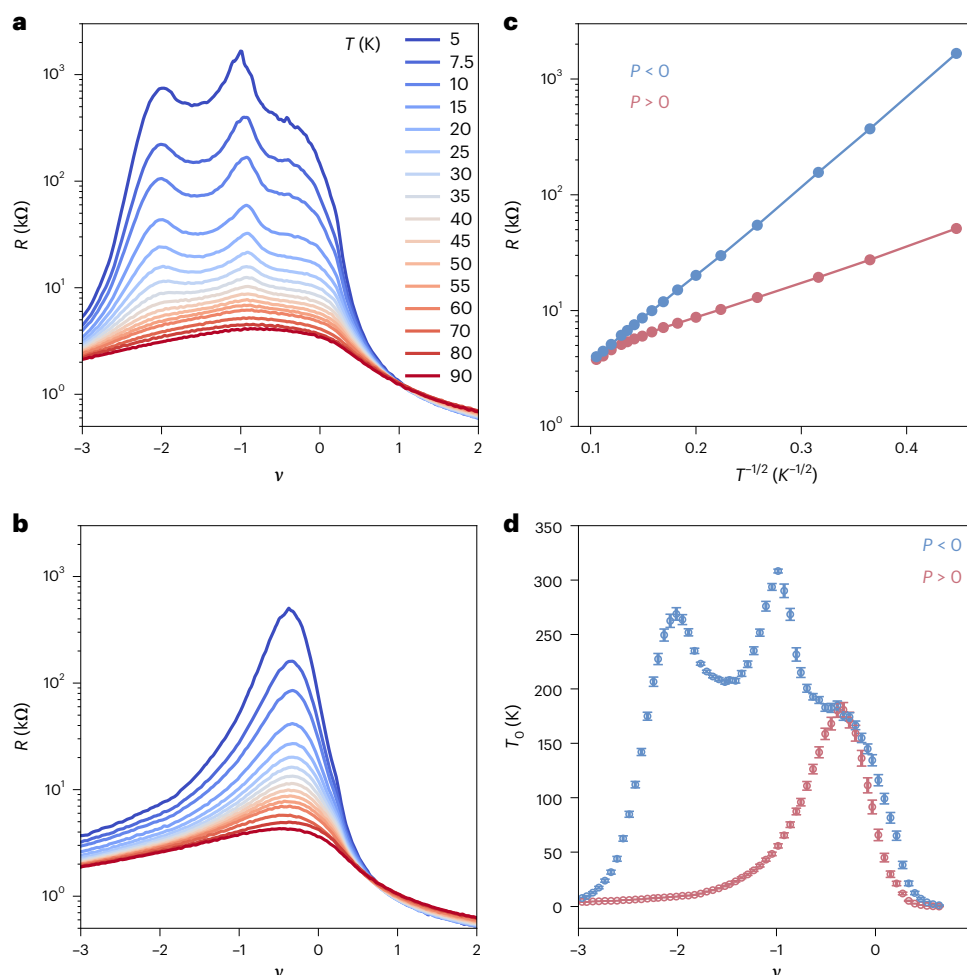


Fig. 3 | Temperature-dependent electrical transport. a, b, Filling-factor-dependent longitudinal resistance at varying temperatures from 5 K to 90 K for $P < 0$ (a) and $P > 0$ (b). Insulating-like behaviour is observed for the entire hole-doping region in both cases. **c,** Resistance (log scale) versus $T^{-1/2}$ at $\nu = -1$ for both $P < 0$ and $P > 0$. The linear dependence demonstrates Efros–Shklovskii variable-

range hopping. **d,** Filling-factor dependence of the extracted temperature T_0 for both $P < 0$ and $P > 0$. Enhancements at the moiré insulating states are observed, showing the shortened localization lengths for $P < 0$. Error bars denote the standard deviations from fitting the data in **c** ($N = 16$ data points have been used).

Because holes in the top layer interfacing directly with the WSe₂ layer experience a strong moiré potential, robust insulating states at $\nu = -1, -2$ are observed for $P < 0$. Conversely, the absence of these insulating states for $P > 0$ shows the substantially weaker moiré potential in the bottom WTe₂ layer.

Note that the total electric polarization is a sum of both the bound charge and mobile hole contributions; the two contributions point in opposite directions. The bound charge contribution dominates the entire hole doping range; this guarantees nearly complete (that is, 100%) interlayer hole transfer upon ferroelectric switching and the emergence of the moiré insulating states at $\nu = -1, -2$ for $P < 0$. Otherwise, the more mobile holes in the bottom layer would electrically shunt the bilayer conduction irrespective of the sign of P ; moiré insulating states would not have been observed.

Variable-range hopping transport

To gain further insights into how ferroelectric switching influences the charge transport in bilayer WTe₂, we perform temperature-dependence studies for both $P < 0$ and $P > 0$, which correspond to with and without moiré effects, respectively. Figure 3a,b shows the density-dependent resistance at varying temperatures (5–90 K) for the two states. Metallic transport (decreasing resistance with decreasing temperature)

is observed only on the electron doping side, where moiré physics is not observable; a metal–insulator transition is also observed near $\nu = 0.7$. The material exhibits insulator-like behaviour for the entire hole-doping range (increasing resistance with decreasing temperature), even when there is no moiré physics for $P > 0$. The $\nu = -1, -2$ insulating states for $P < 0$ survive up to about 70 K while the ferroelectricity persists to near room temperature.

The temperature-dependent resistance at $\nu = -1$ for both $P < 0$ and $P > 0$ is shown in Fig. 3c (similar dependence at other densities is shown in Extended Data Fig. 5). For temperatures below about 100 K, the resistance R scales with the temperature T according to the Efros–Shklovskii variable-range hopping model^{37,38}, $R \propto \exp(T_0/T)^{1/2}$, for over two orders of magnitude in R . The characteristic temperature $T_0 = \frac{2.8e^2}{4\pi\epsilon\epsilon_0k_B\xi}$ is inversely proportional to the localization length ξ of holes in the bilayer WTe₂ (here e , ϵ_0 , ϵ and k_B denote the electron charge, the vacuum permittivity, the dielectric constant of the substrate and the Boltzmann constant, respectively). The density dependence of T_0 is extracted in Fig. 3d. The temperature T_0 is substantially enhanced over the entire hole-doping range for $P < 0$; additional enhancement at the $\nu = -1, -2$ insulating states is also observed. The results demonstrate the additional localization of holes

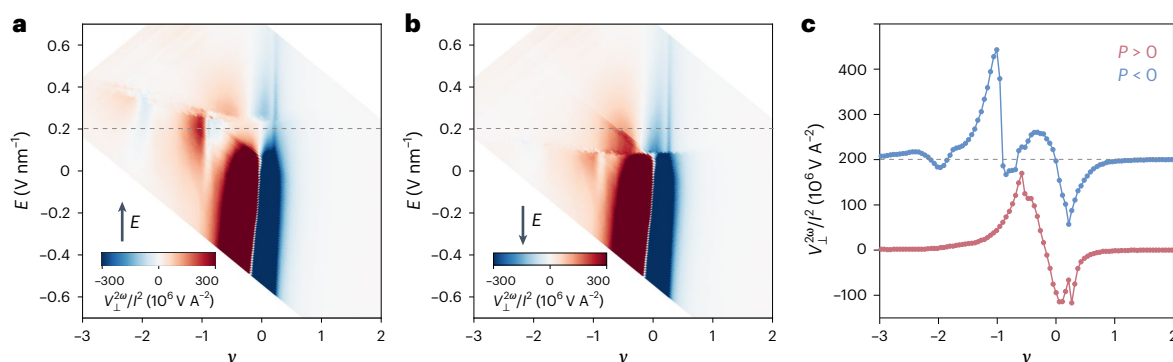


Fig. 4 | Switching the nonlinear anomalous Hall transport by ferroelectricity. **a,b**, Electric-field- and filling-factor-dependent nonlinear anomalous Hall response $V_{\perp}^{2\omega}/I^2$ at 30 K for the forward (**a**) and backward (**b**) electric field scan directions (black arrows). Hysteretic electric-field dependence corresponding to ferroelectric switching is observed. **c**, Filling-factor-dependent $V_{\perp}^{2\omega}/I^2$ for $P < 0$

and $P > 0$ extracted from **a** (blue) and **b** (red), respectively, at $E = 0.2 \text{ V nm}^{-1}$ (the black dashed lines in **a** and **b**). The two curves are vertically displaced for clarity. The horizontal dashed line marks $V_{\perp}^{2\omega}/I^2 = 0$ for $P < 0$. Nonlinear anomalous Hall resets are observed at the moiré insulating states at $\nu = -1, -2$ only for $P < 0$.

that shortens ξ ($\approx 35 \text{ nm}$ for $T_0 = 300 \text{ K}$) when the moiré potential becomes stronger for $P < 0$.

Switchable NAHE

Finally, we examine the effects of ferroelectric switching on the topological transport properties. The presence of an in-plane polar axis (along the mirror line of WTe_2) in the non-centrosymmetric centred-rectangular moiré lattice allows the generation of an out-of-plane magnetization under a bias current perpendicular to the mirror line^{19,21}. The current-induced magnetization can in turn induce an anomalous Hall effect probed by the same bias current. The anomalous Hall voltage thus scales with the bias current squared, giving rise to an NAHE^{19–22}. The effect provides a powerful probe of the Berry curvature dipoles (an imbalance in the Berry curvature occupations) in moiré materials^{39,40}.

To measure the NAHE in the $\text{WTe}_2/\text{WSe}_2$ moiré heterostructure, we bias an a.c. current (I) perpendicular to the mirror line of the moiré lattice and simultaneously measure the first-harmonic longitudinal voltage ($V_{\parallel}$) and the second-harmonic transverse voltage ($V_{\perp}^{2\omega}$) (Methods). Figure 4a,b shows the density and electric-field dependence of $V_{\perp}^{2\omega}/I^2$ under forward and backward scans in the electric field, respectively. The quantity provides a measure of the nonlinear anomalous Hall response (Extended Data Fig. 6). The measurement temperature is kept above 25 K to avoid large sample resistance. The results correlate with the behaviour for the resistance in Fig. 2a,b. In particular, a strong NAHE with sign reversal in $V_{\perp}^{2\omega}/I^2$ is observed at $\nu = -1, -2$ for $E \lesssim 0.3 \text{ V nm}^{-1}$ in the forward scan and for $E \lesssim 0.1 \text{ V nm}^{-1}$ in the backward scan; otherwise only the $\nu = 0$ region shows a strong NAHE. There is also a sharp jump in $V_{\perp}^{2\omega}/I^2$ and an electric-field hysteresis near these critical electric fields. We extract the density-dependent $V_{\perp}^{2\omega}/I^2$ for the two remnant states of spontaneous polarization $P < 0$ and $P > 0$ in Fig. 4c. (Temperature dependence is shown in Extended Data Fig. 7.) A strong NAHE with sign reversal in $V_{\perp}^{2\omega}/I^2$ is observed at $\nu = 0, -1, -2$ for $P < 0$; only the characteristic response at $\nu = 0$ remains for $P > 0$.

The results demonstrate the ferroelectric switching of the NAHE. In the absence of the moiré potential ($P > 0$), the Berry curvature and its dipole are concentrated near the WTe_2 conduction and valence band edges. The NAHE is strong near charge neutrality ($\nu = 0$); the sign reversal in $V_{\perp}^{2\omega}/I^2$ reflects the opposite Berry curvature dipoles for the conduction and valence bands^{20,23}. When flat bands are formed due to the moiré potential ($P < 0$), the Berry curvature and its dipole are redistributed within the moiré Brillouin zone⁴¹; the small moiré band gap is expected to enhance the Berry curvature effects substantially^{41,42}. In the strong correlation limit (at which Coulomb repulsion exceeds the

electronic bandwidth), Hubbard bands are formed; Berry curvatures of opposite signs are expected to concentrate near the Hubbard band edges, resulting in the characteristic NAHE response observed at $\nu = -1, -2$. Interestingly, the sign reversal in $V_{\perp}^{2\omega}/I^2$ at $\nu = -1, -2$ resembles the resets of the Hall density at integer fillings in graphene moiré materials⁴³. The resets reflect the changing carrier type upon doping an insulating state.

In conclusion, we have demonstrated electrical switching of the moiré potential by ferroelectricity in angle-aligned bilayer $\text{WTe}_2/\text{monolayer WSe}_2$; the heterostructure forms a centred-rectangular moiré lattice. The emergence of moiré insulating states at integer filling factors is fully controlled by the spontaneous polarization direction. Ferroelectric switching can also redistribute the Berry curvature dipole in the momentum space, giving rise to a strong NAHE at integer fillings of the moiré lattice. Although the experimental results can be qualitatively understood based on the formation of Hubbard bands, the distribution of Berry curvature dipole within the Hubbard bands in the strong correlation limit (versus that within the single-particle moiré bands in the non-interacting limit) is an open question for future studies; understanding the NAHE in the hopping transport regime⁴⁴ (Fig. 3 and Extended Data Fig. 7) also requires further investigations.

Online content

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at <https://doi.org/10.1038/s41565-023-01376-5>.

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Methods

Device fabrication

T_d-WTe₂/H-WSe₂ moiré heterostructures were fabricated using the standard layer-by-layer stacking method⁴⁵. In short, air-sensitive mono-layer and bilayer WTe₂ flakes were exfoliated from bulk crystals (HQ Graphene) and identified based on their optical contrast on silicon substrates inside a nitrogen-filled glovebox with <1 ppm of oxygen and H₂O. The *a* and *b* axes of the WTe₂ flakes were determined according to the sharp sample edges. Monolayer WSe₂ flakes were exfoliated under ambient conditions. The WSe₂ crystal axes were determined by polarization- and angle-resolved second-harmonic generation spectroscopy⁴⁶. The zigzag axis of WSe₂ was aligned to the crystal *a* axis of WTe₂ during the stacking process. To complete the devices, we first defined platinum electrodes in the Hall-bar geometry on a hexagonal boron nitride (hBN, 10–20 nm thick)/graphite (2–5 layers, serving as a bottom gate) heterostructure using electron-beam lithography. We then used a polymer stamp consisting of a polycarbonate film on polydimethylsiloxane to sequentially pick up flakes of graphite, hBN (~3–4 nm), a WSe₂ monolayer and a WTe₂ monolayer or bilayer. The finished heterostructure on the stamp was released onto the prepatterned platinum electrodes (directly touching WTe₂) at 180 °C. The polymer stamp was then dissolved in chloroform. To fabricate the edge-avoiding devices, we inserted a thin hBN spacer (~2 nm) between the moiré heterostructure and the platinum electrodes so that the WTe₂ sample edges do not touch the metal electrodes^{47,48}. See Extended Data Fig. 2 for the device geometry.

Linear and nonlinear electrical measurements

Electrical transport measurements were carried out in an Oxford Teslatron cryostat down to 1.8 K. Both linear and nonlinear electrical measurements were performed using the standard lock-in technique. Specifically, current with a constant amplitude of 0.5 μA and modulation frequency of 17 Hz was biased along the crystal *a* axis of WTe₂. The longitudinal voltage drop at the same frequency (*V*_{||}) and the Hall voltage drop at the second-harmonic frequency (*V*_⊥^{2ω}) were simultaneously recorded by lock-in amplifiers (Stanford Research SR830). In general, the sample resistance increases substantially at low temperatures; therefore, we limited the nonlinear anomalous Hall measurements to temperatures above 25 K. The nonlinear anomalous Hall response was verified to be independent of the excitation frequency (Extended Data Fig. 6). In addition, we also measured the longitudinal resistance with current biased along the crystal *b* axis of WTe₂ (Extended Data Fig. 8). The linear transport behaviour is largely independent of the bias current direction.

Due to the large sample resistance at low temperatures and possible excitonic interactions⁴⁹, calibration of the carrier density using the ordinary Hall effect is unreliable. Instead, we calibrate the carrier density (*n*) and moiré density (*n*_M) of the device shown in the main text using the parallel-plate capacitor model: $n = \frac{\varepsilon \varepsilon_0 V_{\text{tg}}}{d_{\text{tg}}} + \frac{\varepsilon \varepsilon_0 V_{\text{bg}}}{d_{\text{bg}}}$. Here *d*_{tg} ≈ 4 nm and *d*_{bg} ≈ 22 nm are the top and bottom hBN gate dielectric thicknesses measured by atomic force microscopy; *V*_{tg} and *V*_{bg} are the top and bottom gate voltages; and ε ≈ 3 is the hBN dielectric constant. A hole density of (5.0–5.5) × 10¹² cm⁻² at *v* = -1 was obtained using the applied gate voltages relative to those at *v* = 0; the value is about 10–20% larger than the moiré density (4.5 × 10¹² cm⁻²) estimated using the known lattice parameters (see main text). The discrepancy may arise from the misalignment of the WTe₂ and WSe₂ crystal axes (up to about 2°) and from the identification of the gate voltage corresponding to zero doping density.

PFM

To characterize the moiré pattern using PFM, we fabricated an angle-aligned monolayer T_d-WTe₂/H-WSe₂ heterostructure on a polymer stamp consisting of polycarbonate on polydimethylsiloxane. Flakes of hBN, monolayer WTe₂ and monolayer WSe₂ were sequentially

picked up by the polymer stamp. We then flipped the stamp for the PFM characterization with WSe₂ on the top surface to protect WTe₂ from oxidation. PFM was performed using a Bruker Veeco Icon atomic force microscope with Oxford Instrument Asylum Research ASYLEC-01 Ti/Ir-coated silicon probes. An a.c. bias voltage with a magnitude of 4 V and frequency of 680 kHz was applied to drive the probe during the scan. To improve the signal-to-noise ratio of the obtained moiré pattern, the raw PFM images were Fourier filtered with a threshold of 72% of the maximum intensity (Extended Data Fig. 1). We further fit the fast Fourier transform satellite peak intensity to a Gaussian function, which gives a standard deviation of ~8.8%, corresponding to 1–2° variation in the twist angle. Such twist angle variation is probably exaggerated by the PFM noise, the limited fast Fourier transform resolution and the random buckling of the sample on top of a polymer film.

Estimate of the first moiré valence band bandwidth

The bandwidth of the first moiré valence band $\sim \frac{h^2}{8m^*a_M^2} \approx 17$ meV can be estimated using the WTe₂ effective mass *m*^{*} ≈ 0.9*m*₀ (ref. 33). Here *h*, *m*₀ and *a*_M ≈ 6.5 nm denote the Planck's constant, the free electron mass and the moiré period, respectively. The bandwidth is comparable to or even smaller than the existing moiré materials^{27–31}.

Non-monotonic electric-field-dependent resistance

Extended Data Fig. 4 shows the electric-field-dependent longitudinal resistance for both field scan directions at selected filling factors. We focus our discussions on *v* = 0 and -1, at which high out-of-plane electric fields can be applied. Non-monotonic electric field dependence of the resistance is observed. In particular, resistance peaks are observed at electric fields *E* ≈ -0.3 V nm⁻¹ and 0.55 V nm⁻¹ for *v* = 0 and at *E* ≈ 0.1 V nm⁻¹ and 0.65 V nm⁻¹ for *v* = -1.

To understand the observation, we consider the band structure and interlayer charge distribution in bilayer WTe₂ under an applied electric field *E*. In the presence of a spontaneous polarization *P* (dominated by the bound charge contribution), the total electric field is given by *E*_{tot} = *E* - *P*/χε₀ (where χ is the electric susceptibility). At *v* = 0 (charge neutrality), the resistance reflects the charge gap size of bilayer WTe₂. The gap size is the largest for *E*_{tot} = 0. It decreases linearly with *E*_{tot} due to the interlayer Stark effect²⁰ (Fig. 1c). The maximum gap size is therefore expected at *E* = ±|*P*|/χε₀, which is consistent with the observed resistance peaks at *E* ≈ -0.32 V nm⁻¹ and 0.58 V nm⁻¹ in Extended Data Fig. 4. The asymmetry in the observed electric fields is caused by the heterostructure asymmetry, which shifts the centre of the electric-field dependence by about 0.2 V nm⁻¹. Away from these two fields, the gap size decreases with *E*_{tot}, and the resistance decreases.

On the other hand, the resistance at *v* = -1 is primarily determined by the location of the doped holes, specifically, high (low) resistance for holes residing in the top (bottom) WTe₂ layer. For *P* < 0, a negative electric field (*E* < 0) pushes the doped holes away from the WSe₂/WTe₂ interface, thus reducing the moiré effects (Fig. 1d); the resistance decreases monotonically (Extended Data Fig. 4). For *P* > 0, an electric field *E* > 0.4 V nm⁻¹ pushes the doped holes towards the WSe₂/WTe₂ interface, thereby enhancing the moiré effects (Fig. 1d); the resistance increases and peaks at *E* ≈ 0.65 V nm⁻¹, beyond which the charge gap decreases due to the interlayer Stark effect (as discussed above). Finally, hysteretic switching behaviour is observed for 0 < *E* < 0.4 V nm⁻¹; the resistance depends on the direction of *P*. The centre of the ferroelectric switching is shifted by about 0.2 V nm⁻¹ because of the heterostructure asymmetry.

Data availability

Source data are provided with this paper. Additional data related to this paper are available from the corresponding authors upon reasonable request. Source data are provided with this paper.

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Author contributions

K.K. fabricated the devices. K.K. and W.Z. performed the electrical measurements. Y.Z. and K.K. performed the PFM measurements. K.K. analysed the data. K.W. and T.T. grew the hBN crystals. K.K., J.S. and K.F.M. designed the scientific objectives, oversaw the project and co-wrote the paper. All authors discussed the results and commented on the paper.

Competing interests

The authors declare no competing interests.

Additional information

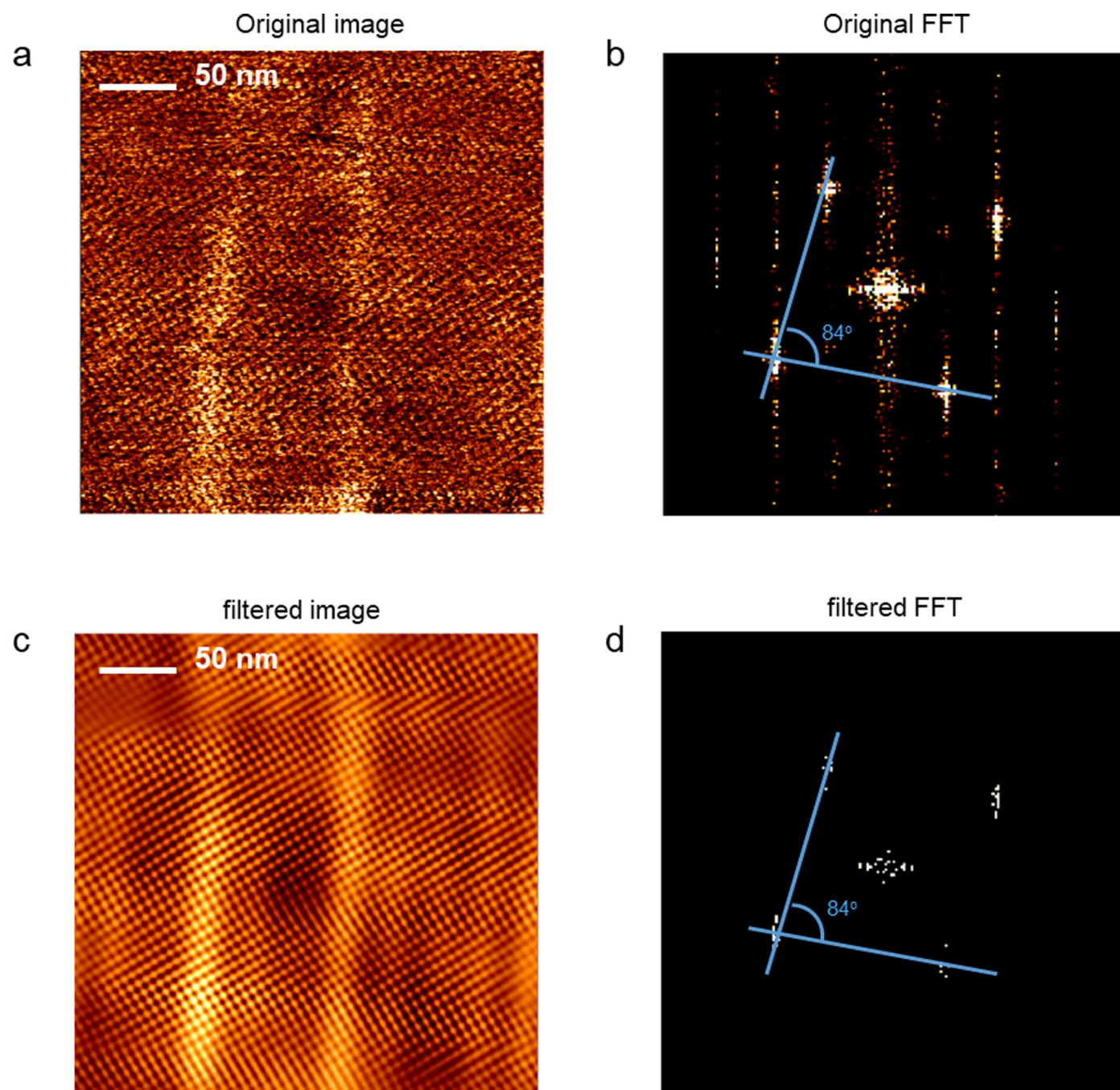
Extended data is available for this paper at <https://doi.org/10.1038/s41565-023-01376-5>.

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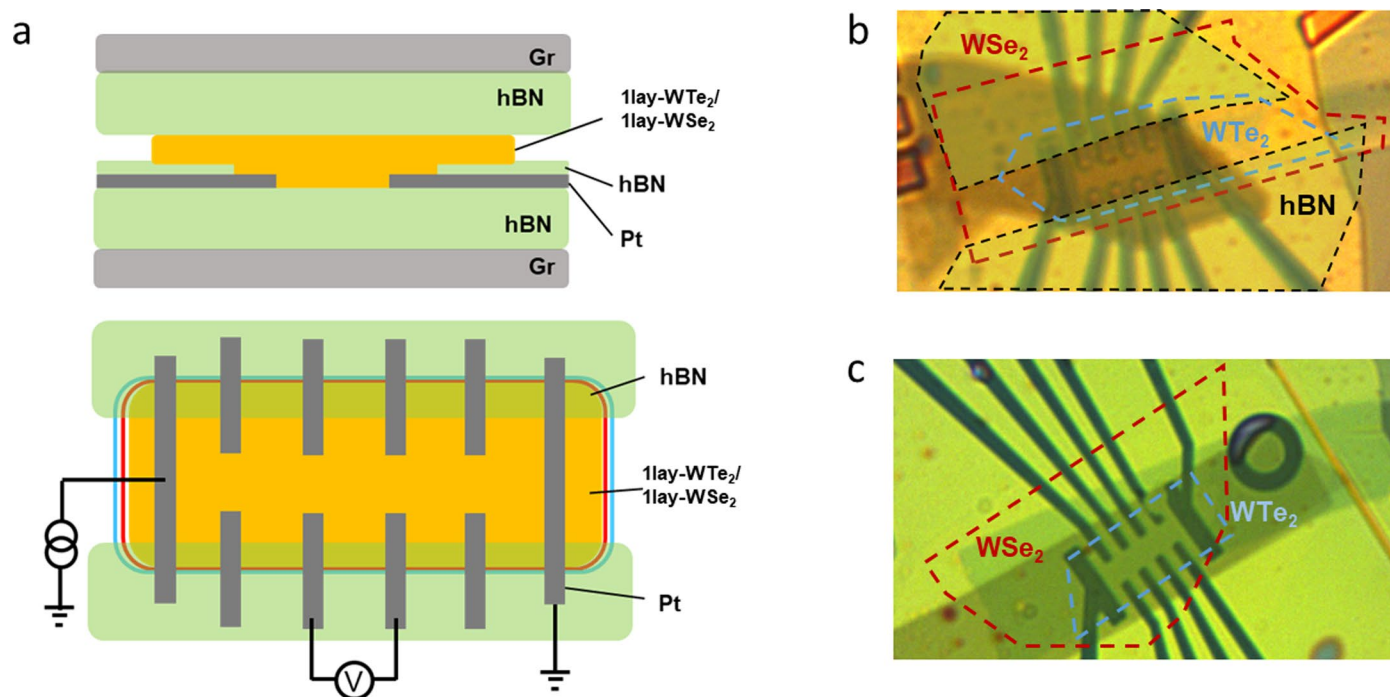
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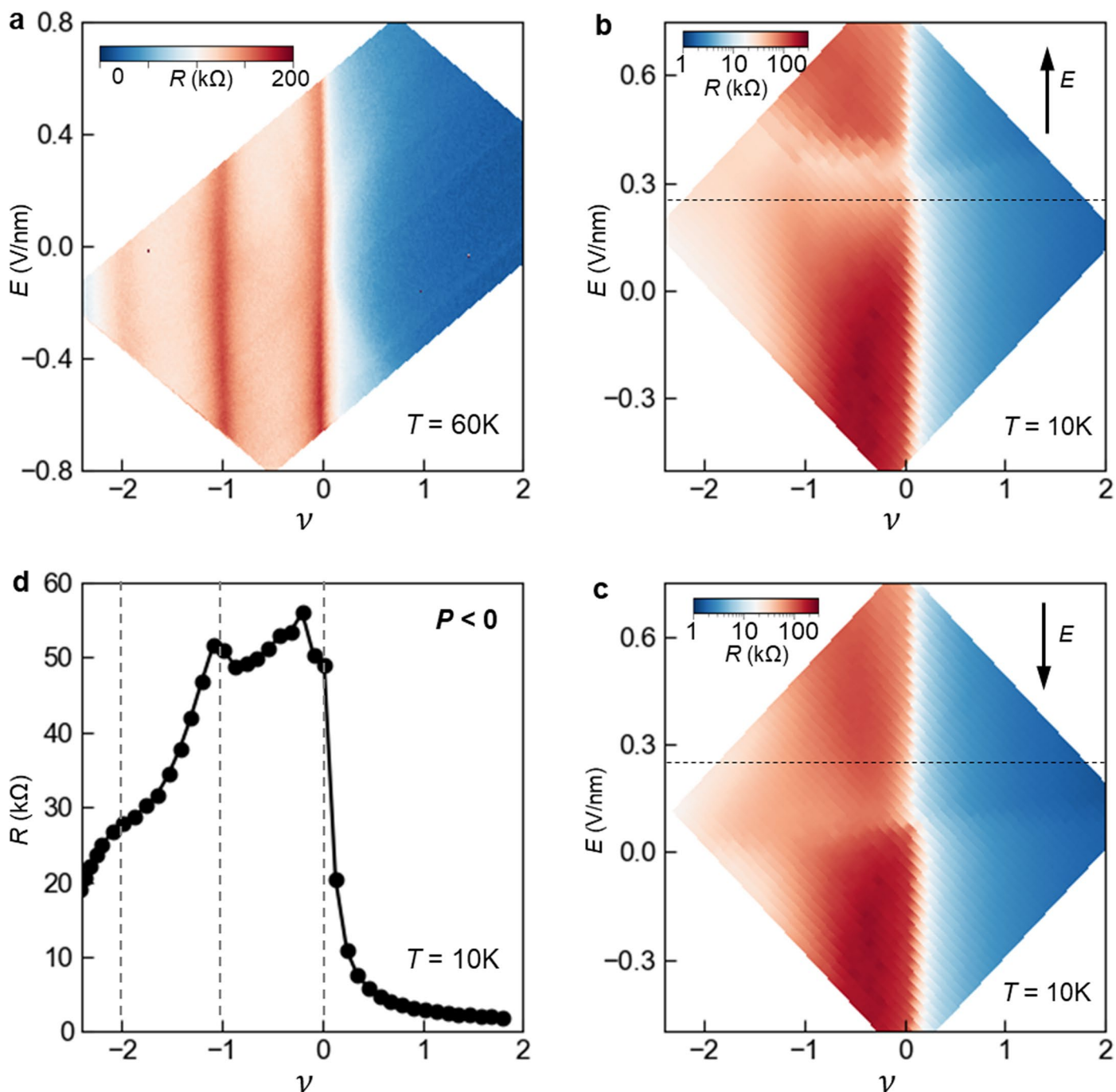
Extended Data Fig. 1 | PFM images and analysis. **a**, Raw PFM image of an angle-aligned monolayer $T_d\text{-WTe}_2$ /monolayer H-WSe_2 heterostructure. **b**, Fourier transform of the raw image. Fourier peaks corresponding to a

centred-rectangular lattice can be clearly observed. **c**, **d**, The Fourier-filtered PFM image (**c**) and the corresponding Fourier transform (**d**). A high-pass filter is applied with a threshold of 72 % of the maximum intensity.



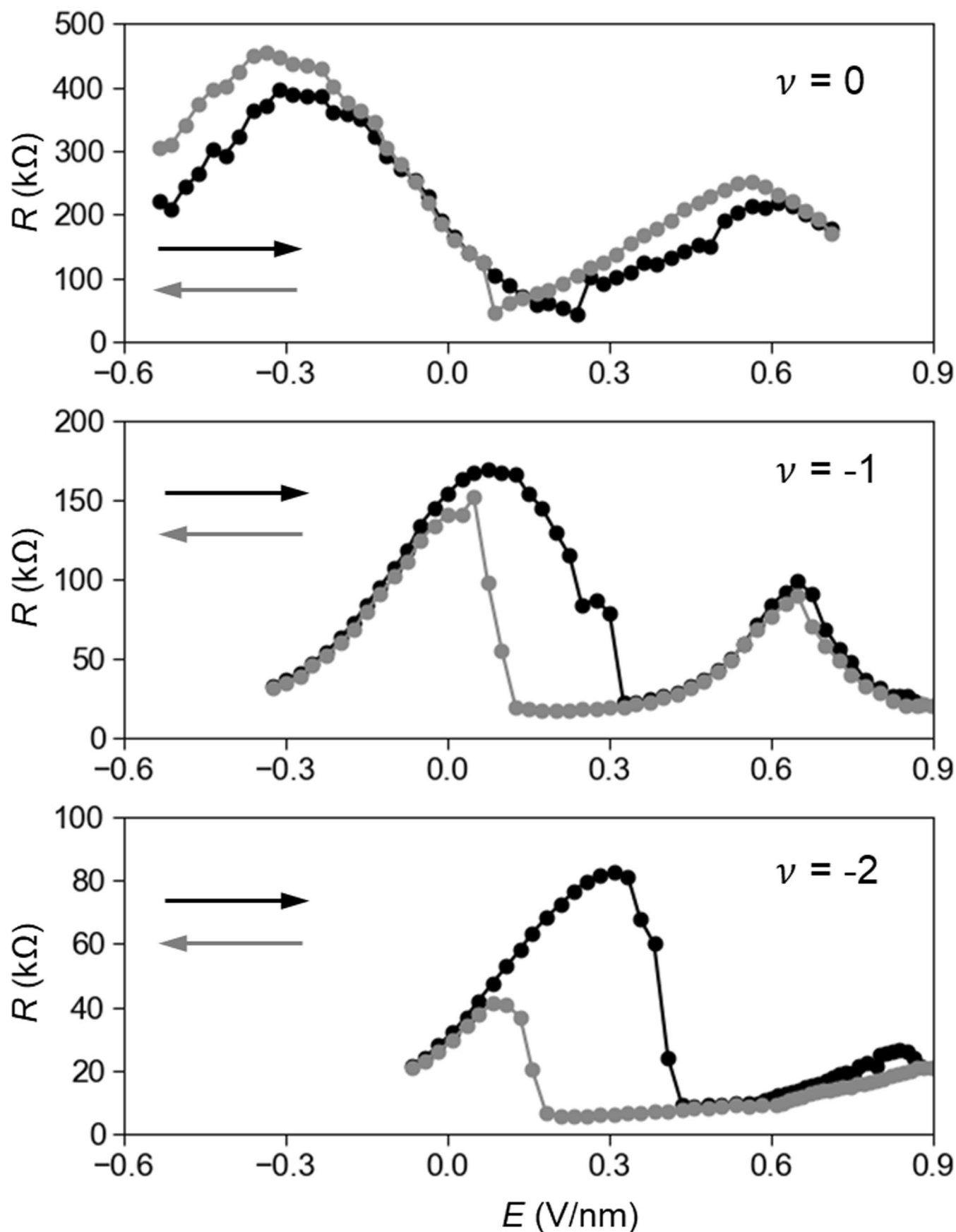
Extended Data Fig. 2 | Device structures for aligned T_d -WTe₂/monolayer H-WSe₂ devices. **a**, schematics of an angle-aligned monolayer WTe₂/monolayer WSe₂ device. Top: cross-section of the device. A thin hBN spacer is inserted between the WTe₂ and the platinum (Pt) electrodes to avoid direct edge contacts. Gr stands for few-layer graphite gate electrode. Bottom: top view of the device. The blue and red lines denote the helical edge states of a quantum spin Hall

insulator. The measurement configuration is also shown. **b**, optical image of a typical angle-aligned monolayer WTe₂/monolayer WSe₂ device. Red, blue and black dashed lines mark the WSe₂, the WTe₂ and the hBN spacer flakes, separately. **c**, optical image of the a typical angle-aligned bilayer WTe₂/monolayer WSe₂ device. Red and blue dashed lines mark the WSe₂ and the bilayer WTe₂ flakes, separately.



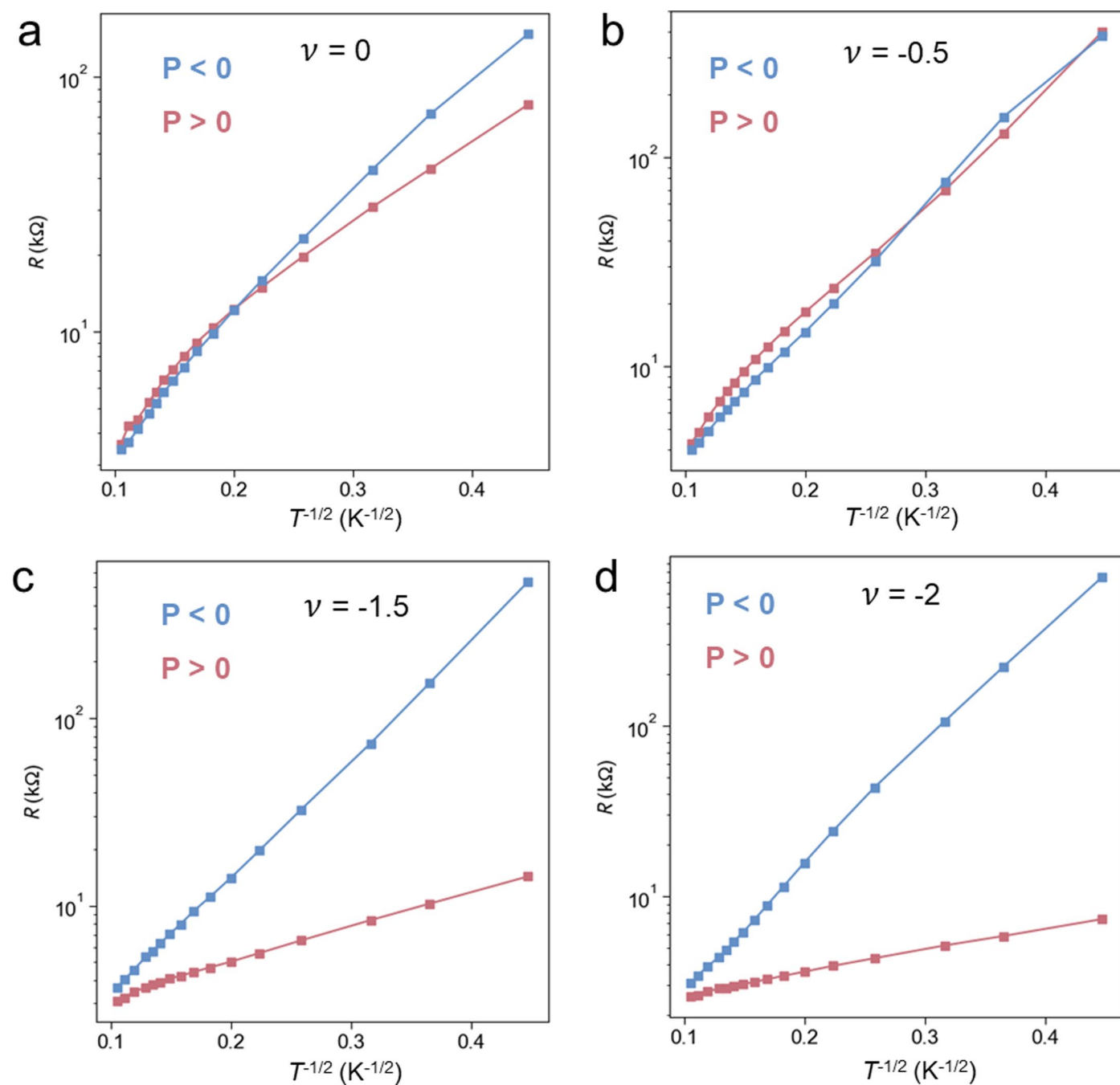
Extended Data Fig. 3 | Repeatability of the data. **a**, Two-terminal sample resistance as a function of filling factor and electric field in another angle-aligned 1lay-WTe₂/1lay-WSe₂ moiré device at $T = 60$ K. **b,c**, Four-terminal sample resistance as a function of filling factor and electric field in another angle-aligned 2lay-WTe₂/1lay-WSe₂ moiré device at $T = 10$ K. Both forward and backward

field scans are shown. **d**, Filling factor dependence of the sample resistance at $E = -0.25$ V/nm for $P < 0$. The moiré insulating states are weaker in this device likely caused by the larger unintentional twist angle introduced in the sample fabrication process.

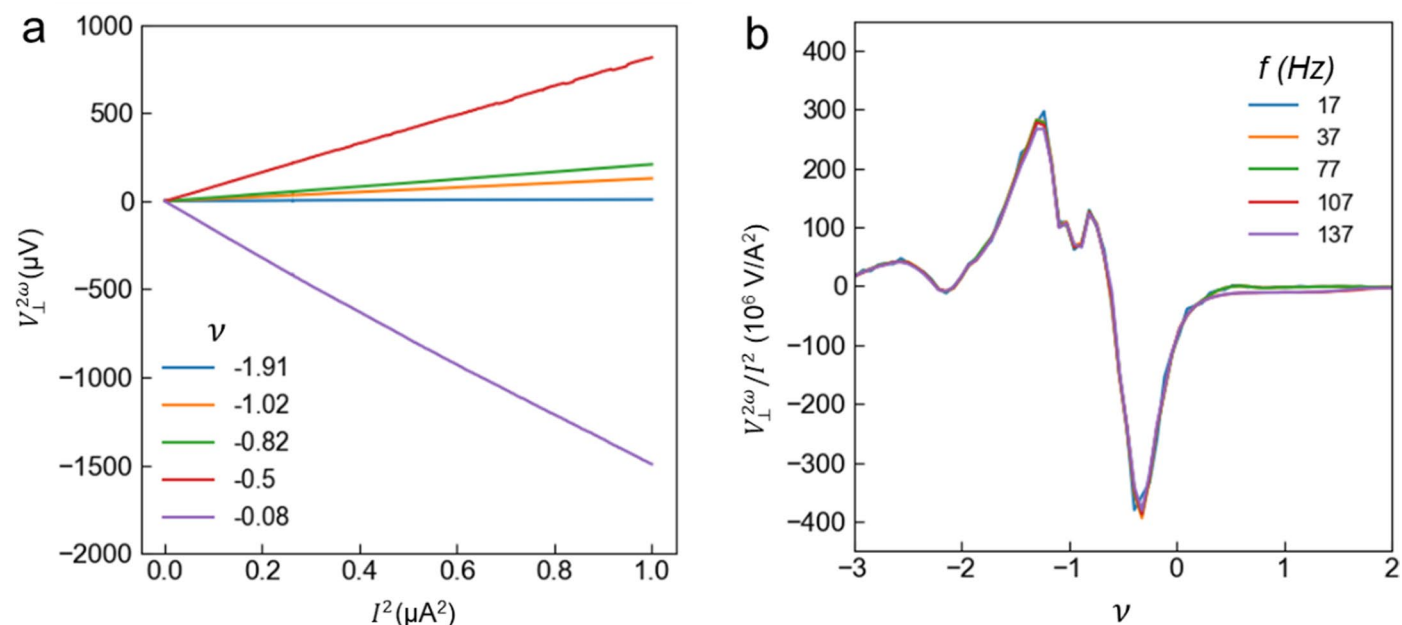


Extended Data Fig. 4 | Electric field dependent resistance at selected filling factors of $\nu=0, -1, -2$. Arrows denote the electric field scan directions. The measurement temperature is 10 K. A clear hysteresis corresponding to

ferroelectric switching is observed. In addition to the ferroelectric switching, non-monotonic electric field dependence is also observed (see discussions in Methods).

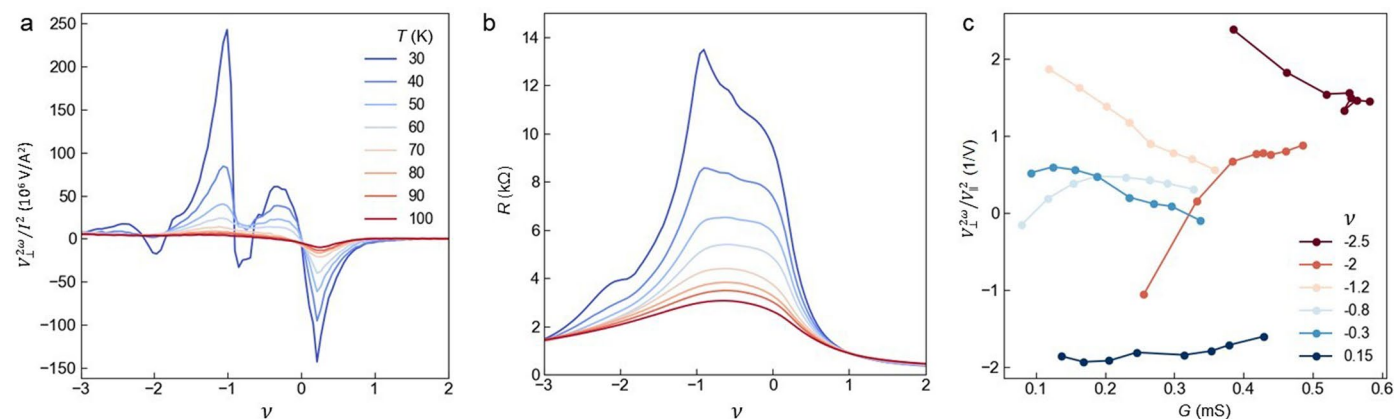


Extended Data Fig. 5 | Variable-range hopping transport at different filling factors. a-d, Resistance (in log scale) versus $T^{-1/2}$ for both $P < 0$ and $P > 0$ at $\nu = 0$ (a), -0.5 (b), -1.5 (c) and -2 (d). The linear dependence demonstrates the Efros-Shklovskii variable-range hopping.



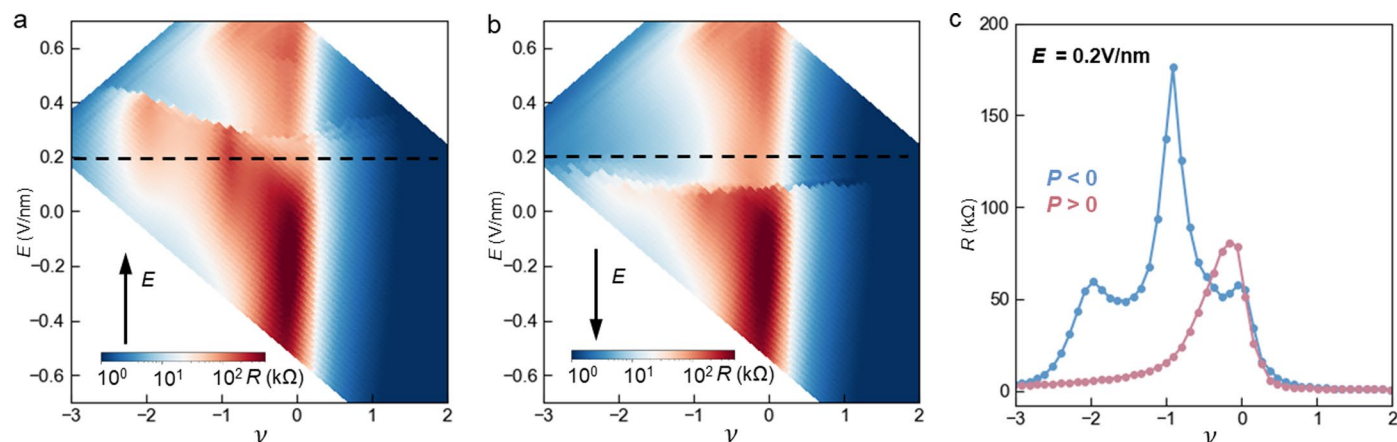
Extended Data Fig. 6 | Nonlinear anomalous Hall response. a, Linear dependence of the second-harmonic Hall voltage on the bias current squared at varying filling factors. The current modulation frequency is 17 Hz. The

measurement temperature is 25 K. **b,** Filling factor dependence of the nonlinear anomalous Hall response $V_L^{2\omega}/I^2$ at varying excitation frequencies of 17, 37, 77, 107, and 137 Hz. The response is independent of the excitation frequency.



Extended Data Fig. 7 | Temperature dependent nonlinear anomalous Hall effect at $E=0.2 \text{ V/nm}$ and $P<0$. **a, b**, Filling factor dependence of $\frac{V_{\perp}^{2\omega}}{I^2}$ (**a**) and the longitudinal resistance R (**b**) at varying temperatures from 30 K to 100 K. **c**, Extracted $V_{\perp}^{2\omega}/V_{\parallel}^2$ as a function of the sample conductance G at selected moiré filling factors from -2.5, to 0.15. Unlike the extrinsic NAHE in the coherent

metallic transport regime, in which $V_{\perp}^{2\omega}/V_{\parallel}^2 \propto G^2$ is expected and has been observed²⁶, complicated dependence of $V_{\perp}^{2\omega}/V_{\parallel}^2$ on G is observed for the variable-range hopping transport regime here. Future studies are required to better understand the NAHE in the hopping transport regime.



Extended Data Fig. 8 | Transport studies along the WTe_2 crystal b -axis.

a, b, Electric field and filling factor dependence for the longitudinal resistance of angle-aligned bilayer $\text{T}_d\text{-WTe}_2$ /monolayer H-WSe_2 heterostructure at 10 K. The current is biased along the crystal b -axis of WTe_2 . The black arrows label the forward (**a**) and backward (**b**) electric field scan directions. Hysteretic electric

field dependence corresponding to ferroelectric switching is observed. **c,** Filling factor dependent longitudinal resistance extracted from **a** (blue) and **b** (red) at $E = 0.2 \text{ V/nm}$ (the black dashed lines in **a, b**). They denote the two spontaneous polarization states $P > 0$ and $P < 0$. Moiré insulating states are observed only for $P < 0$.