

Gate-induced layer sliding drives non-volatile switching of magnetism in tetralayer graphene

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Contents

1	Summary of devices	2
2	Fabrication procedure and structural relaxation	3
3	Transport measurement and domain configuration of device B	4
4	Discussion on domain proportions	7
5	Additional transport measurement of device D	9
6	Microscopic discussion on the sign of anomalous Hall effect in pure rhombohedral graphene	11
7	Calculation of the doping and displacement field dependence of the energy difference	12
8	Determination of $\Delta U(0,0)$	15
9	Effect of free-parameter variation on the relative energy spectrum	15
10	Comparison of band structures for different tight-binding parameters	18
11	Structural interpretation of the hysteresis	18
12	Comparison of the switching energy barrier with sliding ferroelectric	19
13	Determination of $\Delta\chi$	19
14	Orbital Magnetic susceptibility	20
15	Comparison of KPFM methods	22
16	Switching across all devices	23

1 Summary of devices

Table 1 summarizes all the transport devices which we include data for in this work.

Device	Stage Rotation	Top hBN Thickness	Bottom hBN Thickness	Figures [†]
A	180°*	5.8 nm	8 nm	M1, M2, M3, M4, E1, E2, E3, E4, E5, E6, S11, S12
B	0.03°	3.3 nm	4.2 nm	S2, S3
C	−0.02°	8 nm	13 nm	E7, E8
D	−0.02°	17 nm	13 nm	E9, E10, S4, S5, S6
E	0.02°	6 nm	1.5 nm	S15
F	180°*	7 nm	2 nm	S15
G	180°*	8.6 nm	7.5 nm	S15

*For details see Fig. S1 [†]M = Main, E = Extended Data, S = Supplementary

Table 1: Summary of devices. This table summarizes all the transport devices discussed in this work. Note that the actual twist angle in the sample could be different from the stage rotation angle due to strain and structural relaxation.

2 Fabrication procedure and structural relaxation

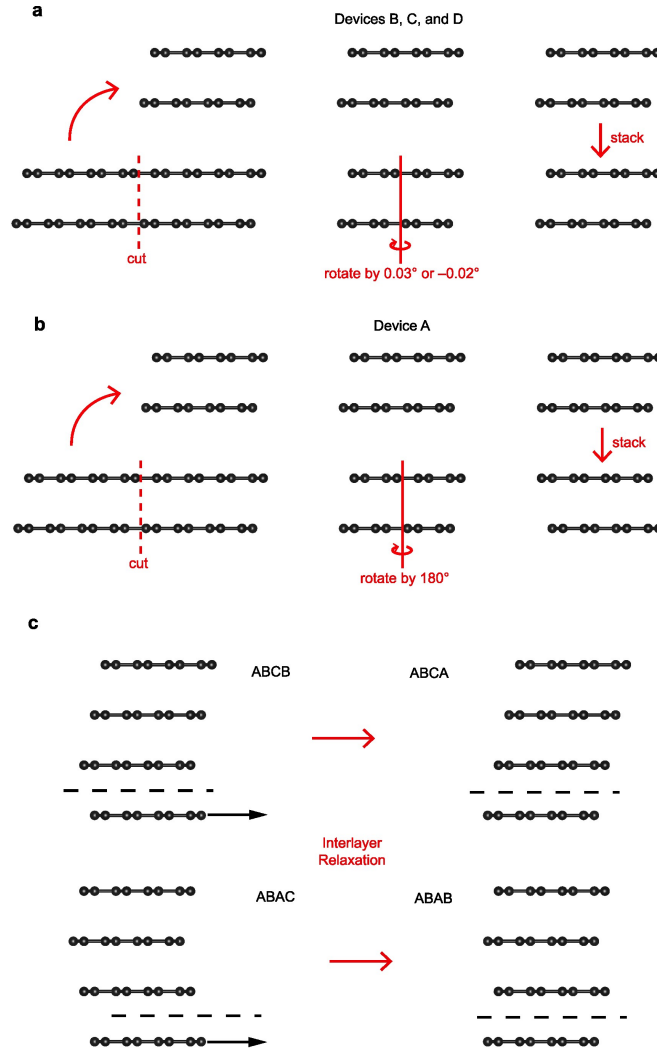


Fig. S1: Heterostructure fabrication procedure and structural relaxation. **a**, Stacking procedure for devices B, C, and D. A bilayer graphene sheet is cut in half and stacked with a twist angle close to 0° relative to the other half. For device B, two bilayer graphene sheets were stacked with a twist angle of 0.03° , and for devices C and D, an angle of -0.02° was used. **b**, Stacking procedure for device A. A bilayer graphene sheet is cut in half and stacked with a twist angle close to 180° relative to the other half. In practice, the twist angle slightly deviates from 180° , giving rise to a long-period moiré pattern as shown in Fig. 1b. **c**, Structural relaxation in device A. Since ABCB and ABAC stacking orders are energetically unstable as compared to both ABCA and ABAB structures [1, 2], they relax into rhombohedral (ABCA) and Bernal (ABAB) stacking orders through the sliding of one layer with respect to the other layers during stacking, as reported in prior studies [3]. Devices B, C, and D yield rhombohedral and Bernal stacking orders without sliding.

3 Transport measurement and domain configuration of device B

The gate-induced sliding transition depends sensitively on the initial domain configuration and the pinning of the domain walls. The sliding transition proceeds through the motion of preexisting domain walls, so a preexisting domain wall is a prerequisite for switching, as established by previous real-space imaging [4, 5]. For the sliding transition to occur, this domain wall must then be depinned from its pinning sites by the applied gate voltage. The pinning sites include the sample edge, the AA-stacking regions arising from the marginal twist angle between the layers, and bubbles or interfacial contamination [5]. Because the distribution of domain walls, the locations of the pinning sites, and the pinning energies all vary from device to device, the gate-induced switching behavior is device- and location-dependent. These factors determine not only whether gate-induced switching occurs, but also whether the AHE can be fully switched on and off.

Device B is a case where the AHE cannot be fully switched on and off, and this is consistent with the weak gate-induced switching we observed in this device. The original domain configuration is already largely mixed. This is evidenced by the weak quantum oscillations across the entire region of the gate-voltage phase diagram, including the electron-doped region in Fig. S2b. The weak quantum oscillations originate from the addition of the individual quantum oscillations of the Bernal and rhombohedral structures, meaning that both the Bernal and rhombohedral domains are of substantial size. We attribute this coexistence of Bernal and rhombohedral structures across the entire phase diagram to a slightly larger twist angle, which produces a domain network of many small triangular domains. Here, the large number of pinning sites impedes domain-wall motion, and the domains can no longer switch strongly [5]. This makes it impossible to fully switch on and off the AHE in device B. We emphasize that this originates from a device-specific domain configuration: in devices A, C and D, which likely have larger domains, we are able to fully switch the AHE on and off.

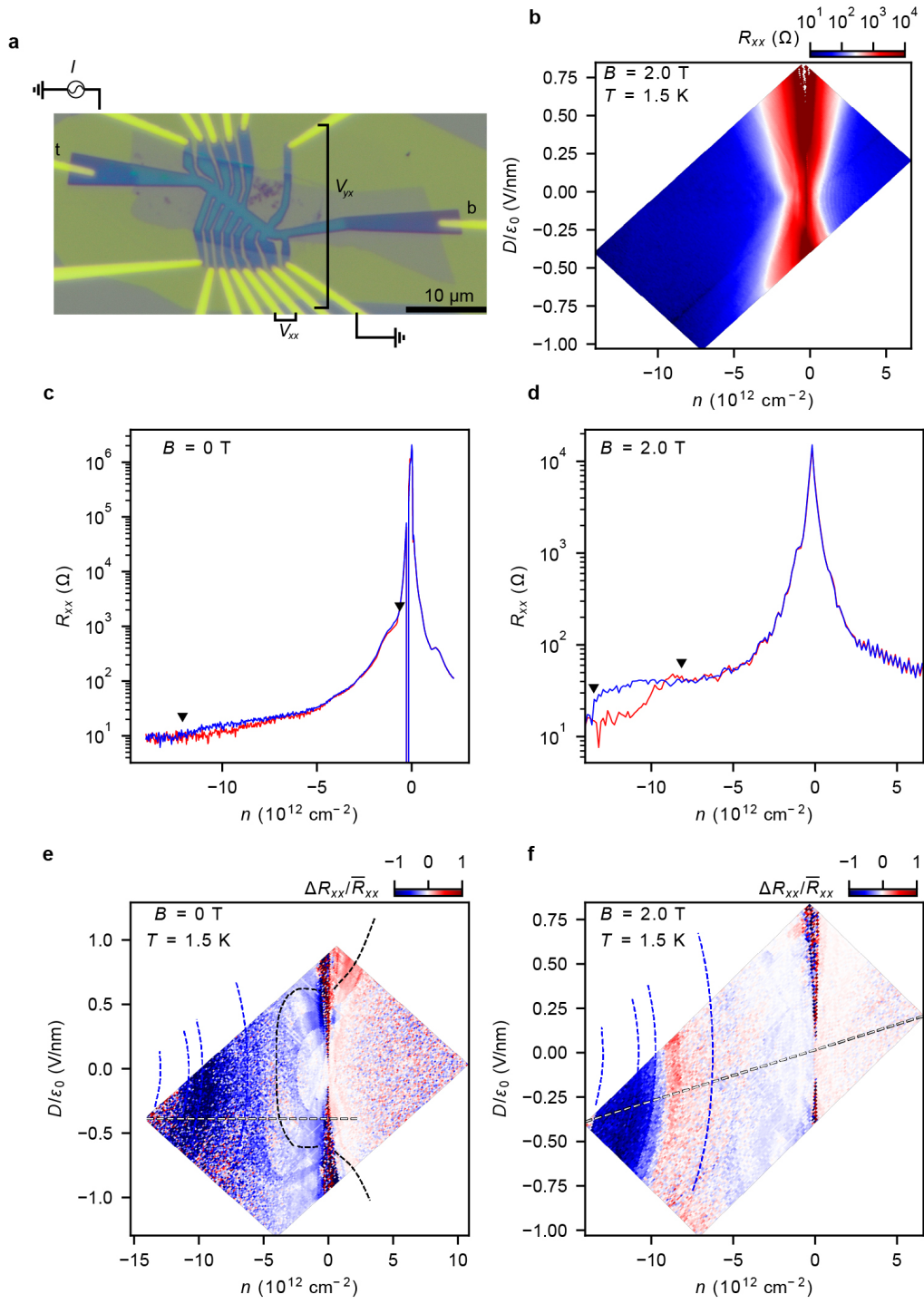


Fig. S2: Hysteresis in device B. **a**, Optical microscope image of device B with the measured contact configuration labeled. **b**, R_{xx} measured at $B = 2$ T. The absence of clear quantum oscillations indicates that Bernal and rhombohedral structures coexist across the entire gate voltage range. **c**, R_{xx} measured as a function of n in the forward (red) and backward (blue) sweeps along the line cut outlined in **e**. The triangles highlight the points at which switching happen. **d**, R_{xx} at $B = 2.0$ T as a function of n in the forward (red) and backward (blue) sweeps along the line cut outlined in **f**. **e**, $\Delta R_{xx}/\bar{R}_{xx}$ measured following the same procedure as in Figs. 2 and 3a. The shape of the switching boundaries is consistent with those observed in device A (black and blue dashed lines taken from Fig. 3a) and is consistent with the theoretical calculation in Fig. 3b. The much smaller $\Delta R_{xx}/\bar{R}_{xx}$ in device B compared to devices A, C, and D indicates that only a small fraction of the device area switches between Bernal and rhombohedral stacking. This is consistent with **b**, which shows that the device remains a mixture of Bernal and rhombohedral structures across the entire gate range. Therefore, device B switches between the Bernal-favored and rhombohedral-favored states according to the history of the gate voltages. **f**, $\Delta R_{xx}/\bar{R}_{xx}$ measured at $B = 2$ T. At this field, the switching boundaries at lower carrier density are more pronounced than at $B = 0$ T, while those near charge neutrality are less distinct. This field-induced change in contrast likely arises from differences in magnetoresistance between Bernal and rhombohedral structures. The switching boundaries at lower carrier density are also consistent with what was observed in device A (blue dashed lines).

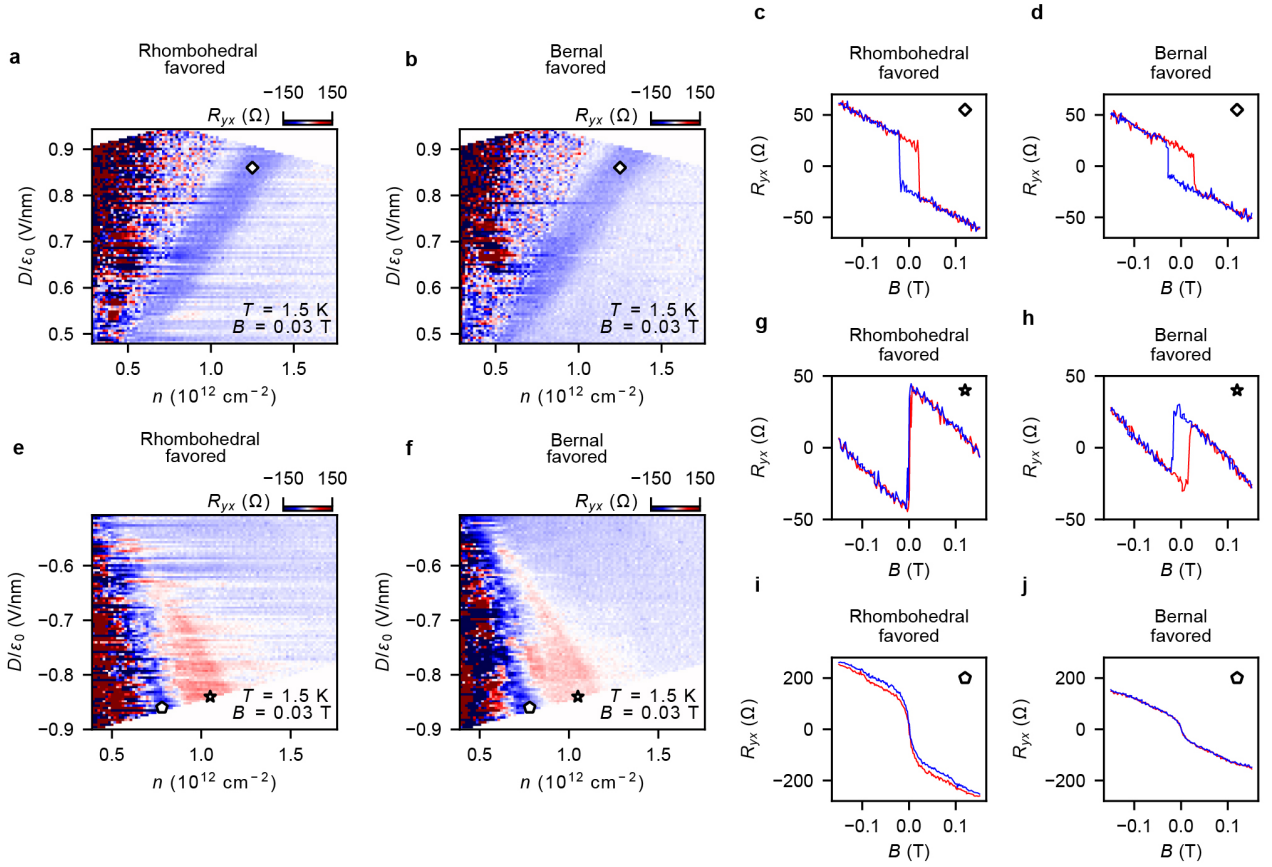


Fig. S3: Anomalous Hall effect in device B. We performed the anomalous Hall measurements of device B in the same manner as in Fig. 4. As discussed in Fig. S2, device B always consists of the mix of Bernal and rhombohedral structures. Depending on the history of the gate voltage sweep, we can switch between Bernal-favored and rhombohedral-favored states. **a,b**, R_{yx} measured at $B = 0.03$ T in the positive D and small positive n region, for the rhombohedral-favored (**a**) and Bernal-favored (**b**) states, respectively. As the device remains in a mixed state in both cases, the AHE is present in both states. The horizontal features in the rhombohedral-favored state likely originate from the stochastic nature of the structural domain configuration. As the switching boundary lies very close to the largest hole density corner, the structural domain configuration can vary each time we reach there. **c,d**, R_{yx} at $T = 1.5$ K as a function of B in the forward (red) and backward (blue) sweeps for the rhombohedral-favored (**c**) and Bernal-favored (**d**) states, respectively. Although complete switching between magnetic and non-magnetic states is absent, a larger AHE is observed in the rhombohedral-favored state. **e,f**, Same as **a** and **b**, but measured in the negative D region. Since the device remains mixed in both cases, a large positive AHE (and a negative AHE in the small- n region) appears in both cases. The sign and regions of AHE are consistent with those in device A. **g,h**, R_{yx} measured at $T = 1.5$ K at the star-marked positions in the rhombohedral-favored (**g**) and Bernal-favored (**h**) states. **i,j**, R_{yx} measured at $T = 1.5$ K at the star-marked positions in the rhombohedral-favored (**i**) and Bernal-favored (**j**) states. Nonlinearity in R_{yx} reveals a weak but finite AHE.

4 Discussion on domain proportions

Despite the absence of direct imaging, our quantum oscillation measurements provide strong evidence that a substantial change in the domain proportion occurs in our device. Extended Data Fig. 1 (device A) and Figure S4 (device D) show the quantum oscillations in both the Bernal-dominant and the rhombohedral-Bernal-mixed states. In the Bernal-dominant state, the device exhibits clear oscillations consistent with Bernal tetralayer graphene [6], indicating that the device is predominantly in the Bernal structure. In the rhombohedral-Bernal-mixed state, by contrast, the oscillations become

noticeably blurred. This arises from the addition of individual quantum oscillations of the Bernal and rhombohedral structures. This indicates an enhancement of rhombohedral domain proportion relative to the Bernal-dominant state.

We note, however, that this quantum-oscillation analysis is only a qualitative measure of the domain ratio. A quantitative determination of the proportion and arrangement of Bernal and rhombohedral domains would require simultaneous real-space imaging and transport measurement as a function of carrier density and displacement field.

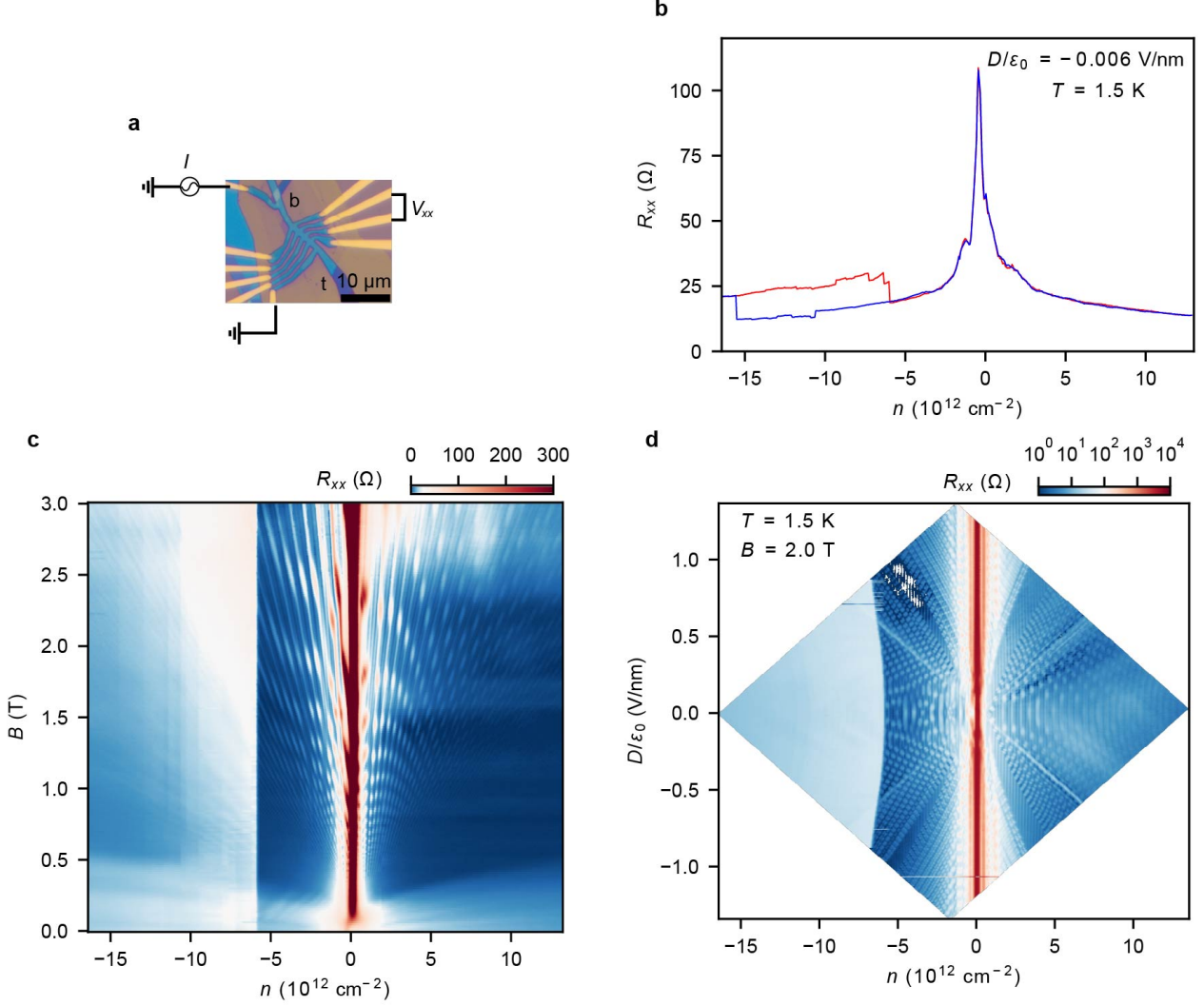


Fig. S4: Hysteresis and quantum oscillations in device D. **a**, Optical microscope image of the device with the measurement configuration labeled. **b**, R_{xx} measured in the forward (red) and backward (blue) scan directions of carrier density n at a constant displacement field of $D/\epsilon_0 = -0.006 \text{ V/nm}$, where ϵ_0 is the vacuum permittivity. **c**, Landau fan diagram of R_{xx} measured in the increasing direction of n at $D/\epsilon_0 = -0.006 \text{ V/nm}$. Clear oscillations consistent with Bernal tetralayer graphene appear after the transition to the Bernal-dominant state at $n = -6 \times 10^{12} \text{ cm}^{-2}$. **d**, R_{xx} measured by first going to the largest-hole-density corner (to realize the rhombohedral-Bernal-mixed structure), then sweeping n in the increasing direction at constant D at $B = 2 \text{ T}$. A clear transition is again observed, across which the quantum oscillations consistent with Bernal tetralayer graphene appear.

5 Additional transport measurement of device D

After cooling device D down again after the temperature dependence measurement in Extended Data Figs. 7 and 8, we found that the switching boundary was changed, as shown in Fig. S5, implying a change in the rhombohedral and Bernal domain configuration after the thermal cycling. This new boundary enabled us to measure the Hall resistance in the rhombohedral-Bernal mixed and Bernal-dominant states in the large- $|D|$, low- n regions. Upon measuring the Hall resistance in both states, we find an anomalous Hall effect only in the rhombohedral-Bernal mixed state, as shown in Fig. S6.

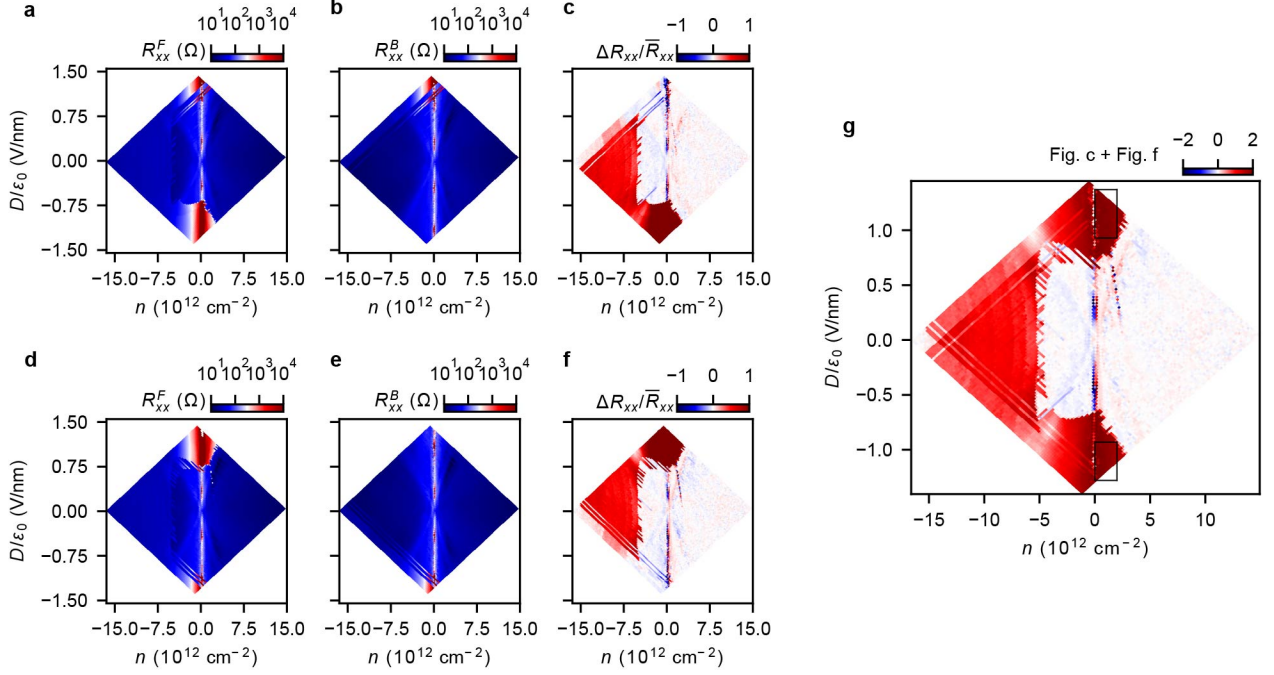


Fig. S5: Switching boundary after thermal cycle in device D. a-f, Measurement of R_{xx} and $\Delta R_{xx}/\bar{R}_{xx}$ following the same measurement scheme of Fig. 2. g, The sum of c and f. The black boxes indicate the areas in which we measured the Hall resistance.

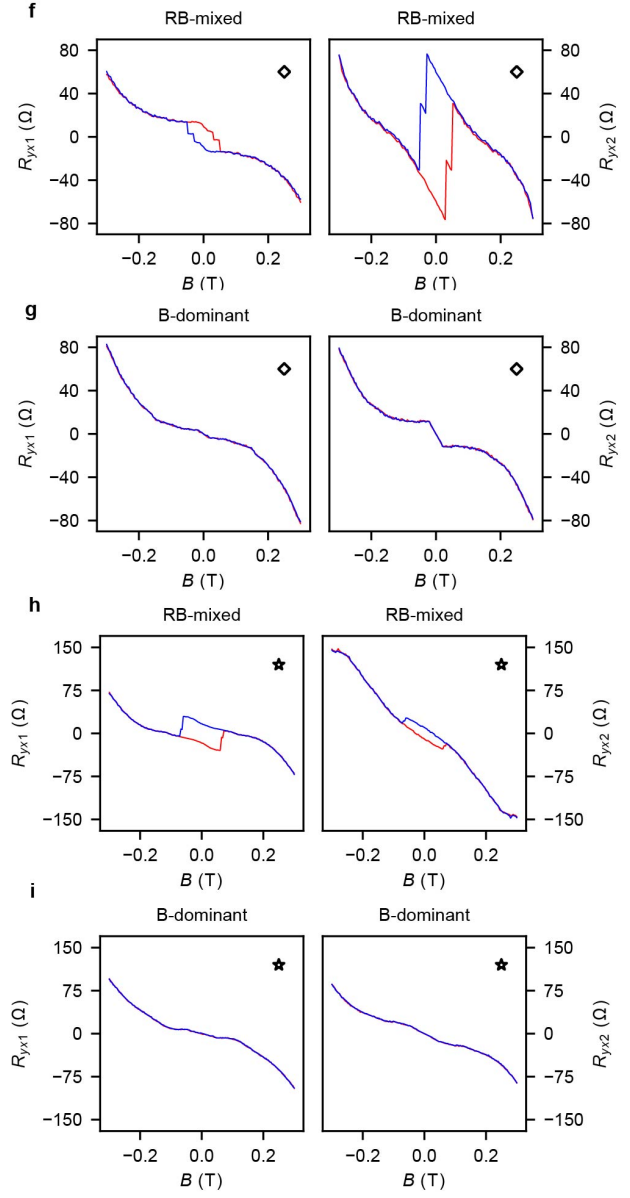
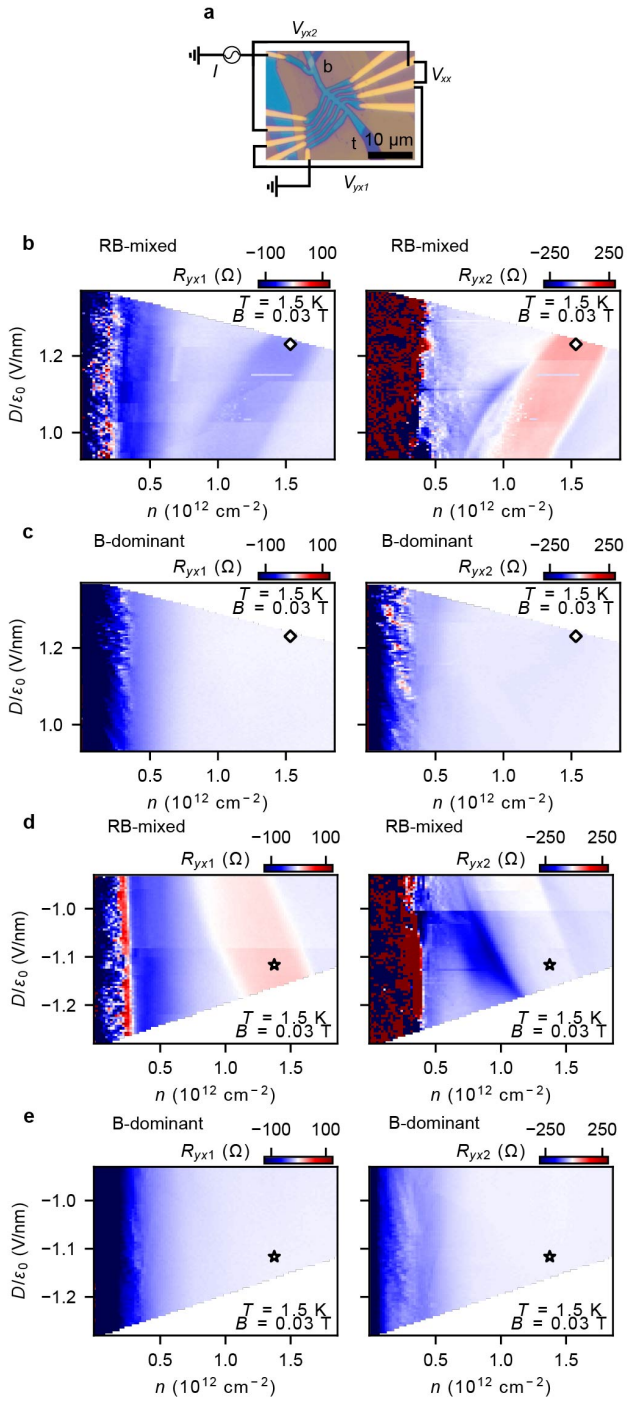


Fig. S6: Hall resistance measurements in device D. **a**, Optical microscope image of the device with the measurement configuration denoted. **b,c**, R_{yx1} and R_{yx2} measured at $B = 0.03$ T in the positive D and small positive n region, in the RB-mixed (**b**) and B-dominant state (**c**), respectively. R_{yx1} and R_{yx2} are measured at the locations denoted in **a** by V_{yx1} and V_{yx2} . **d,e**, R_{yx1} and R_{yx2} measured at $B = 0.03$ T in the negative D and small positive n region, in the RB-mixed (**d**) and B-dominant state (**e**), respectively. The horizontal line features at $D/\varepsilon_0 = -1.08$ V/nm and $D/\varepsilon_0 = -1.01$ V/nm in **d** originate from a slight change in the structural domain configuration upon reaching one of the domain-wall depinning voltages. **f,g**, R_{yx1} and R_{yx2} measured at $T = 1.5$ K at the diamond position as a function of B in both the forward (red) and backward (blue) directions for the RB-mixed (**f**) and B-dominant state (**g**), respectively. A nonzero R_{yx} at $B = 0$ T and the hysteresis appear only in the RB-mixed state. The nonlinear magnetic field dependence in the B-dominant state may suggest the multicarrier nature of the device. **h,i**, R_{yx1} and R_{yx2} measured at $T = 1.5$ K at the star position in the RB-mixed (**h**) and B-dominant state (**i**), respectively. Again, a nonzero R_{yx} at $B = 0$ T and the hysteresis appear only in the RB-mixed state. We note that the sign of the AHE for R_{yx1} in device D differs from those observed in devices A, B, and C. In those devices, the AHE is negative at positive D and positive at negative D . The same behavior is observed for R_{yx2} in device D, whereas for R_{yx1} in device D, the AHE is positive on both sides of the displacement field. We believe this difference in AHE within the same device is related to differences in domain configuration and to how the rhombohedral region and the domain boundary contribute to the AHE. Simultaneous transport measurement and real-space imaging would be required to better understand this behavior, which remains future work. It is important to note, however, that device D supports our main conclusion that the AHE can be switched on and off in a nonvolatile manner by controlling the crystal structure through gating.

6 Microscopic discussion on the sign of anomalous Hall effect in pure rhombohedral graphene

We discuss the sign of anomalous Hall effect (AHE) in a spin- and valley-polarized quarter metal phases of a pure tetralayer graphene. Pure rhombohedral tetralayer graphene becomes layer-polarized under a displacement field, to either the top or bottom layer depending on the direction of the field. Flipping the sign of D reverses two quantities simultaneously: within a given valley, both the sign of the orbital magnetism and the sign of the Berry curvature change [7]. This is summarized in Table 2.

The applied magnetic field couples to the orbital magnetism and selects which valley is occupied, while the Berry curvature of that valley sets the sign of the AHE. As we apply a positive magnetic field under $D > 0$, the valley with positive orbital magnetism is selected (K valley), whose Berry curvature gives a negative AHE. Under $D < 0$, the same positive magnetic field again selects the valley with positive orbital magnetism, but this is now the K' valley, and its Berry curvature again gives a negative AHE. Because flipping D reverses both the orbital magnetism and the Berry curvature, their relative sign is fixed. Therefore, the sign of AHE for a given magnetic field remains the same upon reversing D , as long as we consider pure rhombohedral graphene. This microscopic explanation is consistent with our macroscopic symmetry argument in the main text, which states that the sign of the AHE should remain the same upon reversing D in an inversion-symmetric system.

This is the reason we hypothesized that the domain boundary between rhombohedral and Bernal tetralayer graphene is a possible source of AHE. This boundary naturally breaks inversion symmetry and may host an AHE that reverses sign as a function of D . One possibility is that, at the domain boundary, the relation between the orbital magnetism and the Berry curvature changes with the sign of the displacement field due to a mechanism we do not fully understand yet. Here, the displacement field is related to the layer polarization. Although this is an intriguing possibility, it remains speculation without solid experimental and theoretical evidence, and further research is required to test this hypothesis.

D-field	Valley	Orbital magnetism	Berry curvature
$D > 0$ (Bottom layer polarized)	K	+	-
	K'	-	+
$D < 0$ (Top layer polarized)	K	-	+
	K'	+	-

Table 2: Orbital magnetism and Berry curvature for different displacement fields in pure rhombohedral graphene. Sign of orbital magnetism and Berry curvature for different displacement fields. The external magnetic field couples to the orbital magnetism, which in turn sets the sign of the anomalous Hall effect (AHE) through the Berry curvature. Within this picture, the sign of the AHE should not change upon reversing the displacement field.

7 Calculation of the doping and displacement field dependence of the energy difference

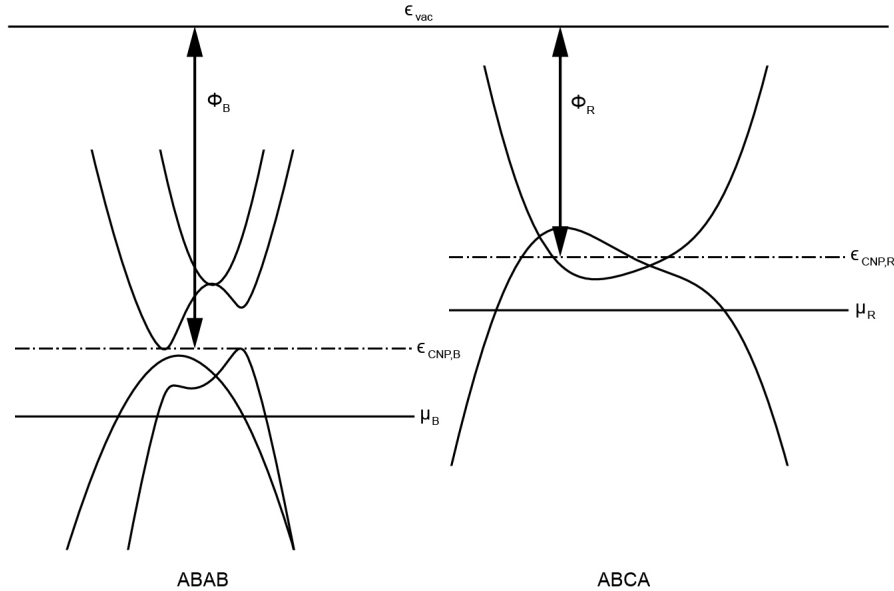


Fig. S7: Schematic illustration for calculating the energy difference. The tight-binding band structure of Bernal (ABAB, left) and rhombohedral (ABCA, right) at $D = 0$. Due to the difference in work function, $\Delta\Phi = \Phi_R - \Phi_B$, the band structures are shifted at the charge neutrality point relative to the vacuum energy, denoted $\epsilon_{\text{CNP},i}$ and ϵ_{vac} , respectively. Because of this shift the relative energy between the two structures changes as the system is doped to some chemical potential, μ_i .

In this section, we explain how we calculate the internal energy difference $\Delta U(n, D)$ between rhombohedral (ABCA) and Bernal (ABAB)-stacked tetralayer graphene. We follow the derivation of Ref. [4]. In the basis of $\{\sigma_l\} = \{A_1, B_1, \dots, A_4, B_4\}$ ¹, the tight-binding Hamiltonians are:

¹We use a slightly different naming convention for different orbitals compared with Ref. [4] and reference therein [8–10]. In our convention, A_{n+1} is right on top of B_n for ABCA stacking. For ABAB stacking A_2 is on top of B_1 , B_3 is on top of A_2 , A_4 is on top of B_3 .

$$\begin{aligned}
H_{ABCA} &= \begin{pmatrix} D_1 & H_{12} & H_{13} & H_{14} \\ H_{12}^\dagger & D_2 & H_{23} & H_{24} \\ H_{13}^\dagger & H_{23}^\dagger & D_3 & H_{34} \\ H_{14}^\dagger & H_{24}^\dagger & H_{34}^\dagger & D_4 \end{pmatrix} + H^{u_D} \\
H_{\sigma_1 l_1, \sigma_2 l_2}^{u_D} &= \delta_{l_1 l_2} \delta_{\sigma_1 \sigma_2} (l - \frac{5}{2}) \\
D_1 = D_2 = D_3 = D_4 &= \begin{pmatrix} 0 & \gamma_0 f \\ \gamma_0 f^* & 0 \end{pmatrix} \\
H_{12} = H_{23} = H_{34} &= \begin{pmatrix} -\gamma_4 f & \gamma_3 f^* \\ \gamma_1 & -\gamma_4 f \end{pmatrix} \\
H_{13} = H_{24} &= \begin{pmatrix} 0 & \frac{\gamma_2}{2} \\ 0 & 0 \end{pmatrix} \quad H_{14} = \begin{pmatrix} 0 & 0 \\ 0 & 0 \end{pmatrix} \\
f &= \exp(i\mathbf{k} \cdot \frac{a}{\sqrt{3}}(1, 0)) + \exp(i\mathbf{k} \cdot \frac{a}{\sqrt{3}}(-\frac{1}{2}, \frac{\sqrt{3}}{2})) + \exp(i\mathbf{k} \cdot \frac{a}{\sqrt{3}}(-\frac{1}{2}, -\frac{\sqrt{3}}{2}))
\end{aligned} \tag{1}$$

$$\begin{aligned}
H_{ABAB} &= \begin{pmatrix} D_1 & H_{12} & H_{13} & H_{14} \\ H_{12}^\dagger & D_2 & H_{23} & H_{24} \\ H_{13}^\dagger & H_{23}^\dagger & D_3 & H_{34} \\ H_{14}^\dagger & H_{24}^\dagger & H_{34}^\dagger & D_4 \end{pmatrix} + H^{u_D} \\
H_{\sigma_1 l_1, \sigma_2 l_2}^{u_D} &= \delta_{l_1 l_2} \delta_{\sigma_1 \sigma_2} (l - \frac{5}{2}) \\
D_1 = D_3 &= \begin{pmatrix} \gamma_2 & \gamma_0 f \\ \gamma_0 f^* & \gamma_5 + \delta \end{pmatrix} \quad D_2 = D_4 = \begin{pmatrix} \gamma_5 + \delta & \gamma_0 f \\ \gamma_0 f^* & \gamma_2 \end{pmatrix} \\
H_{12} = H_{34} &= \begin{pmatrix} -\gamma_4 f & \gamma_3 f^* \\ \gamma_1 & -\gamma_4 f \end{pmatrix} \quad H_{23} = H_{12}^\dagger \\
H_{13} &= \begin{pmatrix} \frac{\gamma_2}{2} & 0 \\ 0 & \frac{\gamma_5}{2} \end{pmatrix} \quad H_{24} = \begin{pmatrix} \frac{\gamma_5}{2} & 0 \\ 0 & \frac{\gamma_2}{2} \end{pmatrix} \quad H_{14} = \begin{pmatrix} 0 & 0 \\ 0 & 0 \end{pmatrix}
\end{aligned} \tag{2}$$

Here, $a = 0.246$ nm is the graphene's lattice constant. The hopping parameters are $(\gamma_0, \gamma_1, \gamma_2, \gamma_3, \gamma_4, \gamma_5, \delta) = (3120, 377, -20.6, 290, 120, 25, -9)$ meV [8–10]. u_D is the layer potential energy difference. We relate it to the displacement field D by $u_D = Dd/\epsilon_r$, where $d = 0.335$ nm, and ϵ_r is the effective dielectric constant of tetralayer graphene, where we used the value of 4.1 [4].

The internal energy of graphene per area is denoted by U_i , where $i = B, R$ refers to Bernal and rhombohedral, respectively.

$$U_i(n, D) = \int D_i(\epsilon) f(\epsilon - \mu_i(n, D)) \epsilon d\epsilon + U_{p,i}. \tag{3}$$

Here, ϵ and μ are energy of the states and chemical potential, both measured from the vacuum level. D_i is the density of states. f is the Fermi-Dirac distribution function at temperature T . $U_{p,i}$ is the energy of ions for stacking configuration i . We assume $U_{p,i}$ to be independent of the carrier density n and the displacement field D . We define the charge neutrality point (CNP) measured from the vacuum level to be $-\Phi_i$ as shown in Fig. S7. Φ_i is the work function at CNP for stacking configuration i . Then the energy of the states and chemical potential, measured from the CNP, are $\epsilon' = \epsilon + \Phi_i$,

$\mu'_i = \mu_i + \Phi_i$. We also redefine the density of states such that $D'_i(\epsilon') \equiv D_i(\epsilon)$.

$$\begin{aligned} U_i(n, D) &= \int D'_i(\epsilon') f(\epsilon' - \mu'_i(n, D)) (\epsilon' - \Phi_i) d\epsilon' + U_{p,i} \\ &= \int D'_i(\epsilon') f(\epsilon' - \mu'_i(n, D)) \epsilon' d\epsilon' + U_{p,i} - (N + n) \Phi_i \end{aligned} \quad (4)$$

Here, N is the number of background electrons at CNP, and n is the change in the carrier density from CNP due to electrostatic doping. The energy difference is defined as

$$\Delta U(n, D) \equiv U_R(n, D) - U_B(n, D). \quad (5)$$

For regularization, we subtract $\Delta U(0, 0)$ from $\Delta U(n, D)$

$$\begin{aligned} \Delta U(n, D) &= \int D'_2(\epsilon') f(\epsilon' - \mu'_2(n, D)) \epsilon' d\epsilon' - \int D'_1(\epsilon') f(\epsilon' - \mu'_1(n, D)) \epsilon' d\epsilon' \\ &\quad - \int D'_2(\epsilon') f(\epsilon' - \mu'_2(0, 0)) \epsilon' d\epsilon' + \int D'_1(\epsilon') f(\epsilon' - \mu'_1(0, 0)) \epsilon' d\epsilon' - n \Delta \Phi + \Delta U(0, 0). \end{aligned} \quad (6)$$

Here, the contribution of $-n \Delta \Phi$ stabilizes the rhombohedral structure through hole doping. The chemical potential μ'_i is determined by solving the equation:

$$\int D_i(\epsilon') f(\epsilon' - \mu'_i) d\epsilon' = n + 8/A_{uc}. \quad (7)$$

Here, A_{uc} is the unit cell area of monolayer graphene, and this equation takes spin degeneracy into account. In practice, we take a very fine grid in the atomic Brillouin zone, and do the corresponding sum on the grid to calculate these integrals. We further define $\text{CNP}_i \equiv \mu'_i(0, 0)$. The reason we do not define it as $\mu'_i(0, D)$ is that due to the particle-hole symmetry breaking terms in the tight-binding Hamiltonian, $\mu'_i(0, D)$ will move around for different D even if we impose the layer potential energy in a symmetric manner, which is not physical.

In summary, the procedures for calculating $\Delta U(n, D)$ are:

1. Calculate the eigenvalues of the tight-binding Hamiltonian, Eq.1, for each point on the momentum grid in the atomic Brillouin zone at $D = 0$.
2. Use the eigenvalues obtained in step (1) as ϵ'_i , calculate $\mu'_i(0, 0)$ using Eq.7.
3. Calculate the eigenvalues of the tight-binding Hamiltonian, Eq.1, for each point on the momentum grid in the Brillouin zone for physical D .
4. Subtract $\mu'_i(0, 0)$ from the eigenvalues obtained in step (3). This gives ϵ'_i .
5. Calculate $\mu'_i(n, D)$ using Eq.7. Finally, calculate the energy difference using Eq.6.
6. In our calculation, for convergence, we take a 3600 by 3600 grid in the parallelogram Brillouin zone. The temperature is $T = 1.5$ K. We took the value of $\Delta \Phi = \Phi_R - \Phi_B = -19$ mV from [11]. $\Delta U(0, 0) = 2.404$ meV/nm² is taken from the theoretical calculation [12]. The reason for the choice is discussed in detail in section 8.

Note that although we calculated ΔU , the Helmholtz free energy F is identical to U under our calculation precision, since the entropy contribution ST is four orders of magnitude smaller as compared to U at $T = 1.5$ K.

8 Determination of $\Delta U(0, 0)$

$\Delta U(0, 0)$ determines the overall energy stability between rhombohedral and Bernal tetralayer graphene at CNP and $D = 0$. Various DFT approaches have been used to estimate this energy difference—differing, for example, in their treatment of dispersion interactions—and there is no universal consensus on the optimal scheme. Because the energy gap between the two stacking structures is small, the predicted values vary appreciably across different approaches [12]. Accordingly, we adopt the value that most faithfully reproduces our experimental results.

Comparing calculated parameters with our measured data, we find that 2.404 meV/nm^2 , obtained using the Local Density Approximation (LDA) [12], captures the features of our switching behavior most appropriately. The switching from B-dominant to RB-mixed occurs at $n_1 = -15.5 \times 10^{12} \text{ cm}^{-2}$, and the major switching from RB-mixed to B-dominant occurs at $n_2 = -4.1 \times 10^{12} \text{ cm}^{-2}$ under $D/\epsilon_0 = -0.067 \text{ V/nm}$ (See Fig. 1c in the main text). Since the switching takes place when the energy difference overcomes the energy barrier associated with domain depinning, we expect that the switching barrier in two cases are similar, namely, $\Delta U(n_1, D) \simeq -\Delta U(n_2, D)$. As shown in Fig. S8, $\Delta U(0, 0) = 2.404 \text{ meV/nm}^2$ satisfies this condition.

We note that our choice of $\Delta U(0, 0)$ carries some uncertainty. However, because it only sets the overall energy difference as an offset, the comparison between constant-energy-difference contours and the switching boundary in Fig. 3b remains valid regardless of the precise value of $\Delta U(0, 0)$.

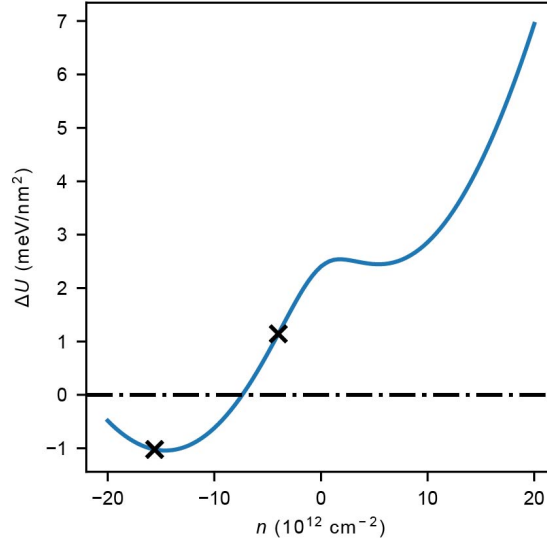


Fig. S8: Determination of $\Delta U(0, 0)$. The calculated energy difference, ΔU , at $D/\epsilon_0 = -0.05 \text{ V/nm}$ with $\Delta U(0, 0) = 2.404 \text{ meV/nm}^2$. The black dashed line indicates $\Delta U = 0$. The black crosses indicate n_1 and n_2 where we observe the sliding transitions in device A.

9 Effect of free-parameter variation on the relative energy spectrum

The discrepancy in fitting between the extracted contours and the calculated energy spectrum likely originates from our approximation of treating the effective dielectric constant of tetralayer graphene ϵ_r as a constant value, which is used to convert the displacement field D to the interlayer energy difference u_D in tetralayer graphene. This quantity ϵ_r is not a constant in reality but depends on both the displacement field D and the carrier density n , since the out-of-plane polarizability of the multilayer is itself a function of band filling and band structure. A fully rigorous treatment would require solving the layer-resolved charge distribution and the internal electric field in tetralayer graphene self-consistently

at each (n, D) point via a Hartree calculation [13]. Such a treatment is computationally demanding across the full phase diagram. We therefore adopted the standard approach in the multilayer graphene literature of using a single effective ε_r to convert D to u_D . The value used in the main calculation, $\varepsilon_r = 4.1$, was taken from a related calculation [4].

This approximation is expected to be most reliable in the moderate-carrier-density regime and less accurate at high carrier density, where screening effects become stronger. We believe this density-dependent screening is one likely reason why the quantitative agreement degrades in the negative high-carrier-density regime. Importantly, this limitation cannot be fully resolved simply by choosing a different constant ε_r ; a quantitatively accurate treatment would require a density- and field-dependent, self-consistent calculation, which lies beyond the scope of the present work.

To draw out a meaningful conclusion within this approximation, we focus on the robustness of the qualitative conclusion rather than on optimizing a quantitative fit. To this end, we recomputed the energy spectrum under several reasonable variations of the model parameters, as shown in Fig. S9. Specifically, we varied $\Delta\Phi$ around our measured average value of -22 mV (taken from our measured surface potential average value from KPFM, see Supplementary Information section 15); tested a larger $\varepsilon_r = 5.5$ from [14]; and used alternative tight-binding parameters from [13, 15].

Across these variations, the same qualitative behavior persists: rhombohedral stacking becomes energetically favorable in the large-hole-doping region, and in the large- $|D|$ close to the charge-neutral region, consistent with the experimental observations. Some parameter choices, for example, those shown in Fig. S9f and S9j, give particularly good apparent agreement with the experimental contours. However, we do not claim that these are the uniquely correct physical parameters. Given the size of the parameter space, it would likely be possible to find parameter combinations that improve the quantitative agreement, but such agreement alone would not establish the physical correctness of those parameters. The purpose of the parameter study is instead to demonstrate that the qualitative result is robust against reasonable parameter variations.

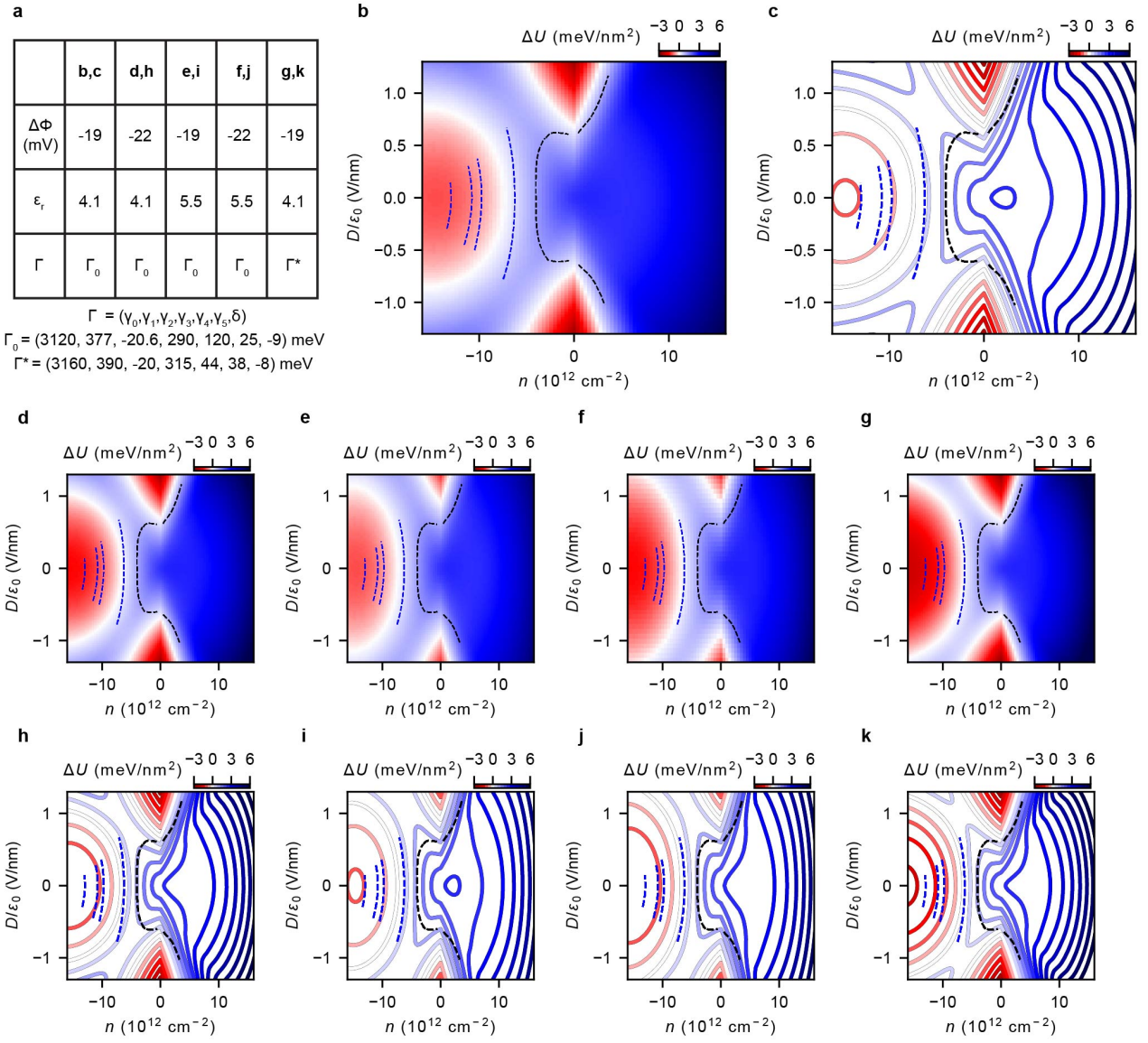


Fig. S9: Comparison of different parameters on ΔU . **a**, Table of the different parameters used to calculate ΔU . We test different values of the work function difference $\Delta\Phi$, dielectric constant ϵ_r , and tight-binding parameters. **b**, Calculated ΔU using the parameters outlined in **a**. This is the same as Fig. 3b. **c**, Contour map of **b**. **d-k**, Calculated ΔU and contour maps using the parameters outlined in **a**.

10 Comparison of band structures for different tight-binding parameters

We calculate the band structures of Bernal and rhombohedral tetralayer graphene under different tight-binding parameters for comparison. We find that most features of the band structure are consistent between the calculations using the two different sets of tight-binding parameters.

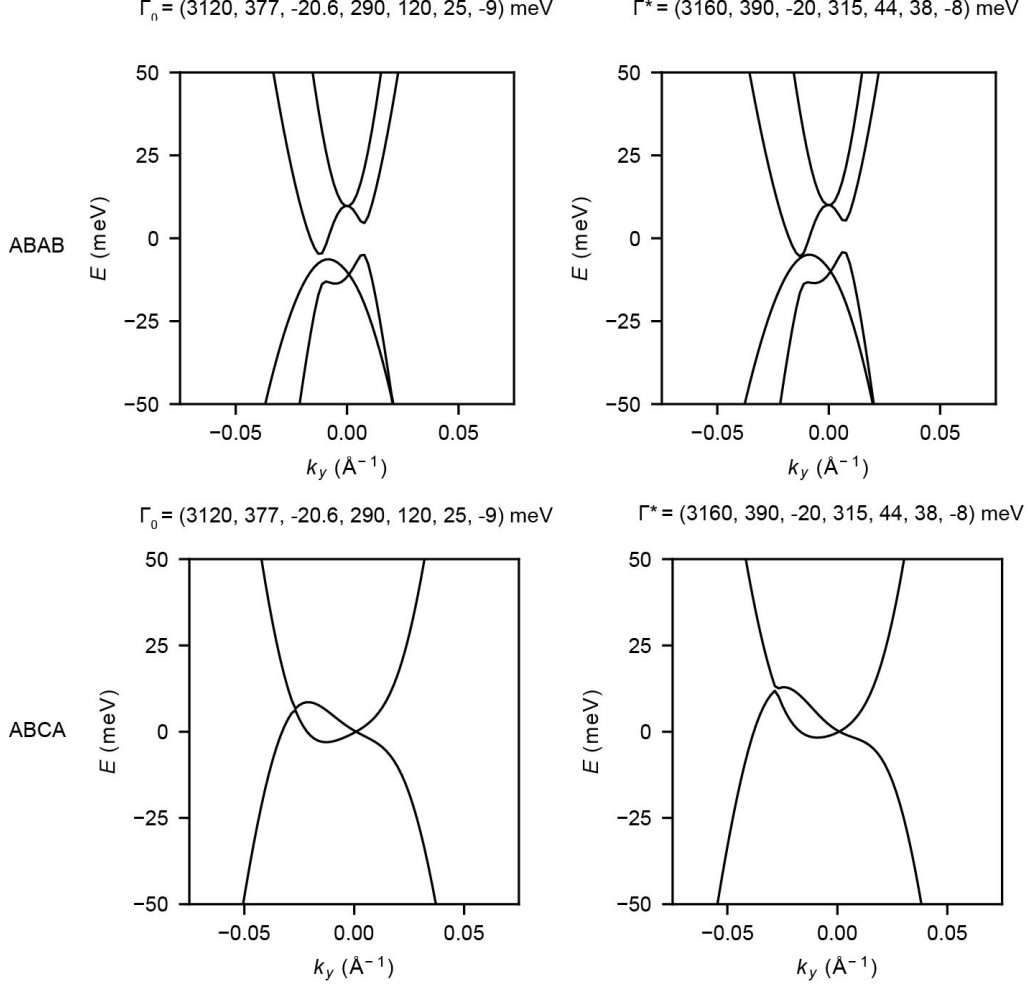


Fig. S10: Band structures calculated for different tight-binding parameters. The tight-binding band structure of Bernal (ABAB, top) and rhombohedral (ABCA, bottom) at $D = 0$, for tight-binding parameters taken from [8] and from [13, 15].

11 Structural interpretation of the hysteresis

First-order phase transitions of correlated electronic phases can produce hysteresis, which has been reported before in multilayer rhombohedral graphene [16, 17]. While such electronic phase transitions can produce hysteresis, we do not believe that this effect is causing the hysteresis in our device for the following reasons.

First, our hysteresis appears in the high-carrier-density regions where correlated phases are not observed. The hysteresis due to electron correlation appears along the phase boundaries of the interaction-driven correlated phases in rhombohedral graphene. These phases exist only at low densities in rhombohedral graphene, typically around or less than 10^{12} cm^{-2} , where the Fermi energy is

close to the flat-band regions of the rhombohedral band structure [16, 17]. Meanwhile, the hysteresis in our device is observable up to a much higher carrier density of $1.55 \times 10^{13} \text{ cm}^{-2}$, where the band dispersion is much larger, and the effect of electron correlations is nearly negligible. This discrepancy indicates that the gate-induced hysteresis we observe is not of electron-correlation origin.

Second, our hysteresis occurs at room temperature, well above the temperature at which correlated phases are observed. We measured the temperature dependence of the gate-induced switching in device D, which we present in Extended Data Figs. 9 and 10. The hysteresis in R_{xx} as a function of n persists all the way to room temperature, as shown in Extended Data Fig. 9. We further investigated the switching boundary within the space of carrier density and displacement field across multiple temperatures, as shown in Extended Data Fig. 10, which again shows hysteretic switching up to room temperature. The persistence of hysteretic switching well above the transition temperature of correlated phases provides further evidence that the gate-induced hysteresis we observe does not originate from electron correlations.

Finally, the structural switching between Bernal and rhombohedral graphene through electrostatic gating has been imaged in real space previously, with infrared scattering-type scanning near-field optical microscopy (IR-SNOM) [4] and with scanning tunneling microscopy (STM) [18]. We prepared the coexisting Bernal and rhombohedral graphene structure in a manner similar to that of the previous report and obtained a similar free-energy phase diagram, but with a different probe: real-space imaging in their case [4] and transport measurements in ours. Therefore, it is reasonable to attribute our hysteresis observation to the structural phase transition between Bernal and rhombohedral graphene.

12 Comparison of the switching energy barrier with sliding ferroelectric

Here, we compare the energy barrier for the sliding phase transition with that of sliding ferroelectric bilayer hBN [19]. The switching barrier for a sliding ferroelectric can be estimated from the polarization magnitude, $P = 2.25 \times 10^{-12} \text{ C/m}$, and the coercive field, $E = 0.1 \text{ V/nm}$. The corresponding switching energy is $2PE = 2.8 \text{ meV/nm}^2$. This value is of the same order of magnitude as the switching energy of ΔU , which is on the order of 1 meV/nm^2 .

It is notable that the switching energies for the sliding transition are comparable despite the different underlying physical mechanisms. In the case of bilayer hBN, the electric field drives the sliding phase transition through the sliding ferroelectric mechanism. In contrast, in our system, switching is induced by doping-induced change in the stability of different stacking configurations. The consistency of the energy scales, despite these distinct driving forces, can be attributed to the fact that both systems undergo transitions through interlayer sliding. The similar energy scale is especially reasonable considering the similar mechanical properties of hBN and graphene.

13 Determination of $\Delta\chi$

The carrier density at which switching happens evolves as a function of magnetic field B as shown in Figs. S11 and S12. This evolution allows us to extract the difference in the magnetic susceptibility $\Delta\chi = \chi_R - \chi_B$, where χ_R and χ_B are the magnetic susceptibility in the rhombohedral and Bernal structures, respectively. As the switching occurs at constant free energy, ΔF , the switching point in n versus B maps out a curve along

$$\Delta F(B, n_0 + \Delta n) - \Delta F(B, n_0) = \Delta F(0, n_0) - \Delta F(B, n_0), \quad (8)$$

where n_0 and $n_0 + \Delta n$ are the carrier densities at which switching happens at 0 T and finite magnetic field, respectively. By first order approximation, we get

$$\Delta n \frac{\partial \Delta F}{\partial n} = \frac{1}{2} \Delta\chi B^2. \quad (9)$$

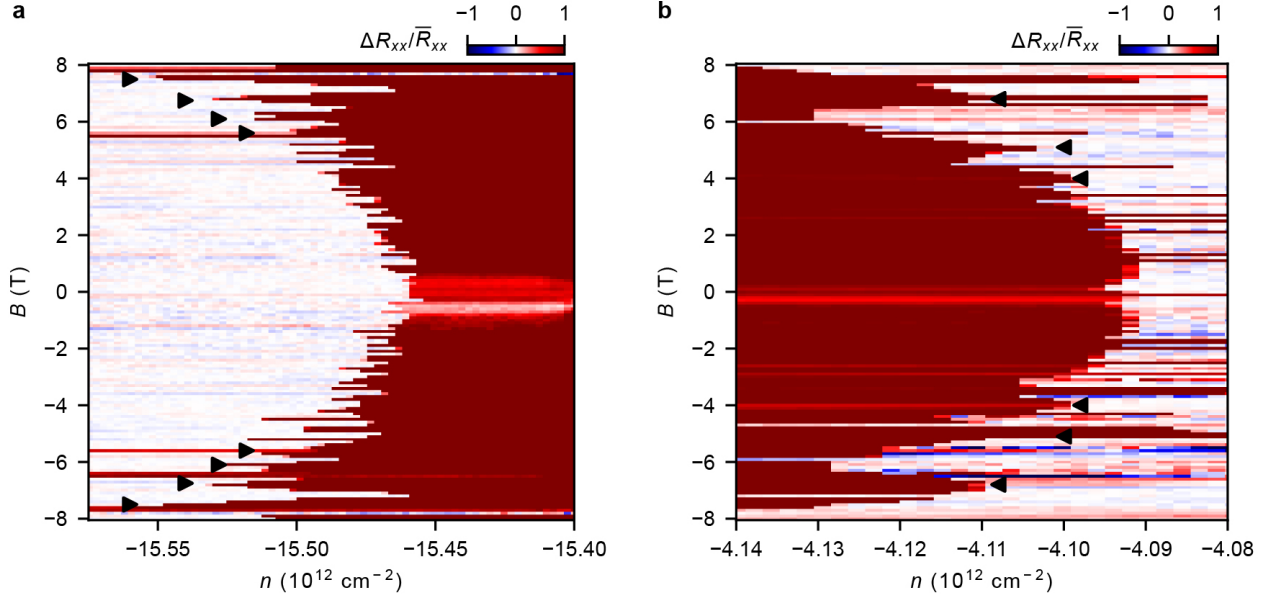


Fig. S11: Magnetic field dependence of the switching boundary. **a,b,** $\Delta R_{xx}/\bar{R}_{xx}$ as a function of B and n near the switching boundaries highlighted by the triangles in Fig. 1c. In both cases, rhombohedral stacking becomes destabilized with increasing B . This measurement allows extraction of the difference in magnetic susceptibility between rhombohedral and Bernal stacking, $\Delta\chi$, which agrees well with theoretical calculations (see Supplementary Information for the details). The switching values of n oscillate with B under high magnetic fields, as indicated by the black triangles. These oscillations originate from de Haas–van Alphen (dHvA) oscillations of the Helmholtz free energy difference ΔF and the work function difference $\Delta\Phi$, reflecting an interference pattern between the quantum oscillations from the rhombohedral and Bernal tetralayer graphene.

We can obtain $\partial\Delta F/\partial n \simeq \partial\Delta U/\partial n$ from the calculation in section 7 and Fig. S8. By fitting the evolution of the carrier density at which switching happens with $\Delta n = aB^2$ as shown in Fig. S12, we can solve for $\Delta\chi$.

$$\Delta\chi = 2a \frac{\partial\Delta U}{\partial n}. \quad (10)$$

The calculated values are

$$\Delta\chi = -4.5 \times 10^{-7} \text{ Jm}^{-2}\text{T}^{-2} \text{ for } n = -4.1 \times 10^{12} \text{ cm}^{-2}, \text{ and}$$

$$\Delta\chi = 0.1 \times 10^{-7} \text{ Jm}^{-2}\text{T}^{-2} \text{ for } n = -15.5 \times 10^{12} \text{ cm}^{-2}.$$

14 Orbital Magnetic susceptibility

In this section, we explain how we calculate the orbital magnetic susceptibility of different stacking configurations. We closely follow Ref. [20].

Throughout this section, we use the continuum expansion of the tight-binding Hamiltonian near the valley $\mathbf{K} = (0, \frac{4\pi}{3a})$, $f(\mathbf{K} + \mathbf{k}) \approx \frac{\sqrt{3}a}{2}(ik_x - k_y)$. We denote the Hamiltonian obtained from the continuum expansion by $\mathcal{H}$.

In the linear response theory, magnetic susceptibility χ is calculated by

$$\chi(T, \mu) = \text{Im} \int_{-\infty}^{\infty} d\epsilon f(\epsilon - \mu) F(\epsilon + i0^+) \quad (11)$$

$$f(\epsilon - \mu) = (\exp(\frac{\epsilon - \mu}{T}) + 1)^{-1} \quad (12)$$

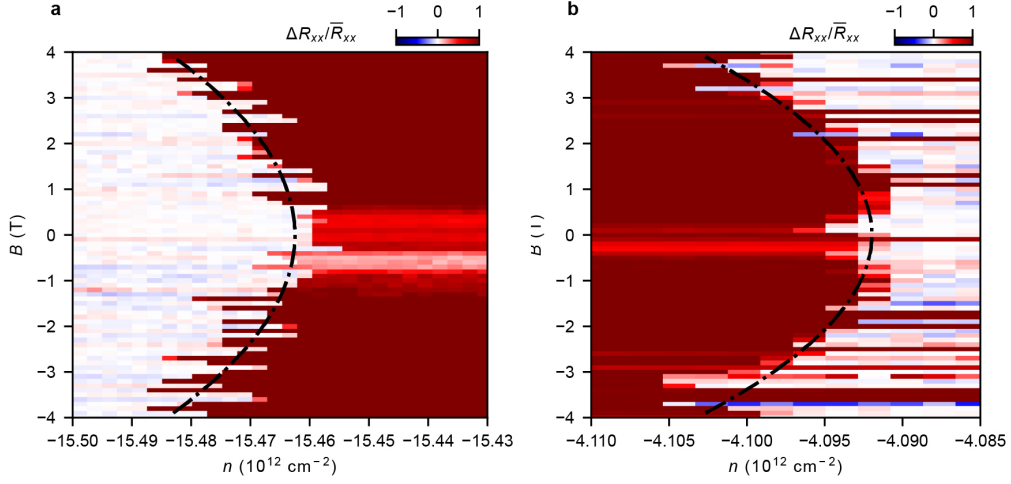


Fig. S12: The evolution of the carrier density at which switching happens as a function of B . a, b, Zoomed in view of Fig. S5. The black dashed line is the fit with a quadratic of the form, $\Delta n = aB^2$. Only values of B from -4 T to 4 T are considered so as to avoid the additional interference pattern.

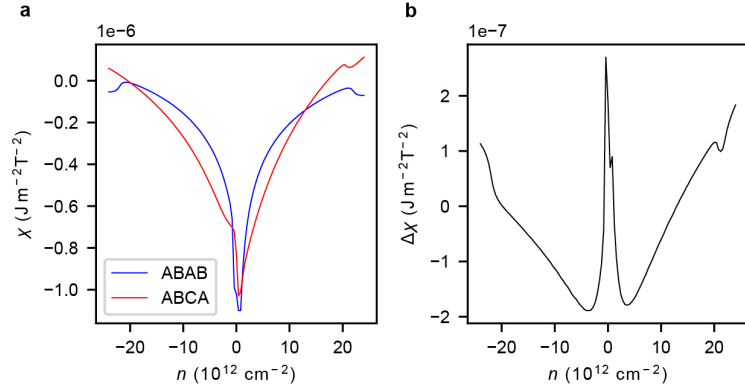


Fig. S13: Zero temperature magnetic susceptibility of rhombohedral and Bernal tetralayer graphene. a, Calculated magnetic susceptibility χ for Bernal (ABAB) and rhombohedral (ABCA). b, The difference in magnetic susceptibility, $\Delta\chi = \chi_R - \chi_B$. We assume spin-valley degeneracy in our calculation.

$$F(z) = -\frac{g_v g_s e^2}{2\pi A \hbar^2} \sum_{\mathbf{k}} \text{tr}(G\mathcal{H}_x G\mathcal{H}_y G\mathcal{H}_x G\mathcal{H}_y) \quad (13)$$

For graphene-based system, $g_v = g_s = 2$ are valley/spin degeneracy. $G(z) \equiv \frac{1}{z-H}$ is the Green's function. We define $H_x \equiv \frac{\partial \mathcal{H}}{\partial k_x}$, $H_y \equiv \frac{\partial \mathcal{H}}{\partial k_y}$. To model the impurity scattering rate, $i0^+$ in the Green's function is replaced by $i\Gamma$. In encapsulated samples like ours, we expect a realistic Γ of around 0.5 meV. For this calculation, a value of 4 meV was used to ensure convergence. The result shows little sensitivity to Γ , as confirmed by our calculations varying Γ from 4 to 20 meV.

We take a momentum space square with side length 8 nm^{-1} centered around the origin, with roughly 4000 by 4000 grid points inside it.

Figure S13 shows the result of the calculated magnetic susceptibility for both rhombohedral and Bernal structures, along with their difference. The calculated values are

$$\begin{aligned} \Delta\chi &= -1.9 \times 10^{-7} \text{ Jm}^{-2}\text{T}^{-2} \text{ for } n = -4.1 \times 10^{12} \text{ cm}^{-2}, \text{ and} \\ \Delta\chi &= -0.5 \times 10^{-7} \text{ Jm}^{-2}\text{T}^{-2} \text{ for } n = -15.5 \times 10^{12} \text{ cm}^{-2}. \end{aligned}$$

The order of magnitude of the theoretically calculated $\Delta\chi$ compares well with the calculated from experiment values in section 13, which confirms that our method is effective in estimating thermodynamic quantities in 2D devices.

15 Comparison of KPFM methods

We performed KPFM measurements on our two twisted double bilayer graphene samples and obtained values of -30.05 and -13.60 mV, which averages to -22 mV (Fig. S14). Meanwhile, Atri *et al.* obtains the value of -19 mV and claims that this value is constant and independent of the region where the measurement is taken [11]. These values are comparable in magnitude, but we have larger sample-to-sample dependence in the measurement. We believe that the value of Atri *et al.* (-19 mV) is likely closer to the intrinsic value of the work function difference. Their result is established using two-pass sideband KPFM in an N₂ atmosphere at 100 mbar. Our measurements, by contrast, were performed in ambient conditions using single-pass frequency-modulation KPFM. These differences in the measurement scheme and the measurement environment affect the apparent surface potential contrast between rhombohedral and Bernal regions.

First, in single-pass frequency-modulation KPFM, the topographic and electrostatic responses are acquired simultaneously; hence, the measured potential is convolved with topographic crosstalk. Two-pass sideband KPFM, by contrast, measures topography and surface potential in separate passes, suppressing the topographic crosstalk. The two-pass scan yields a more faithful measure of the surface potential contrast, but we could not implement this method due to the limited capability of our instrument.

The second factor is the measurement environment. Because our measurements were performed under ambient conditions, adsorbed water and other airborne contaminants form a thin layer on the sample surface, producing surface dipoles that contribute directly to the measured surface potential. The N₂ atmosphere backfilled after high-vacuum pumping, as used by Atri *et al.*, eliminates this contribution and yields an intrinsic surface potential.

For these reasons, we believe the value of Atri *et al.* (-19 mV) is more reliable, and we used it in our comparison with theory, but have included our own measurement for reference.

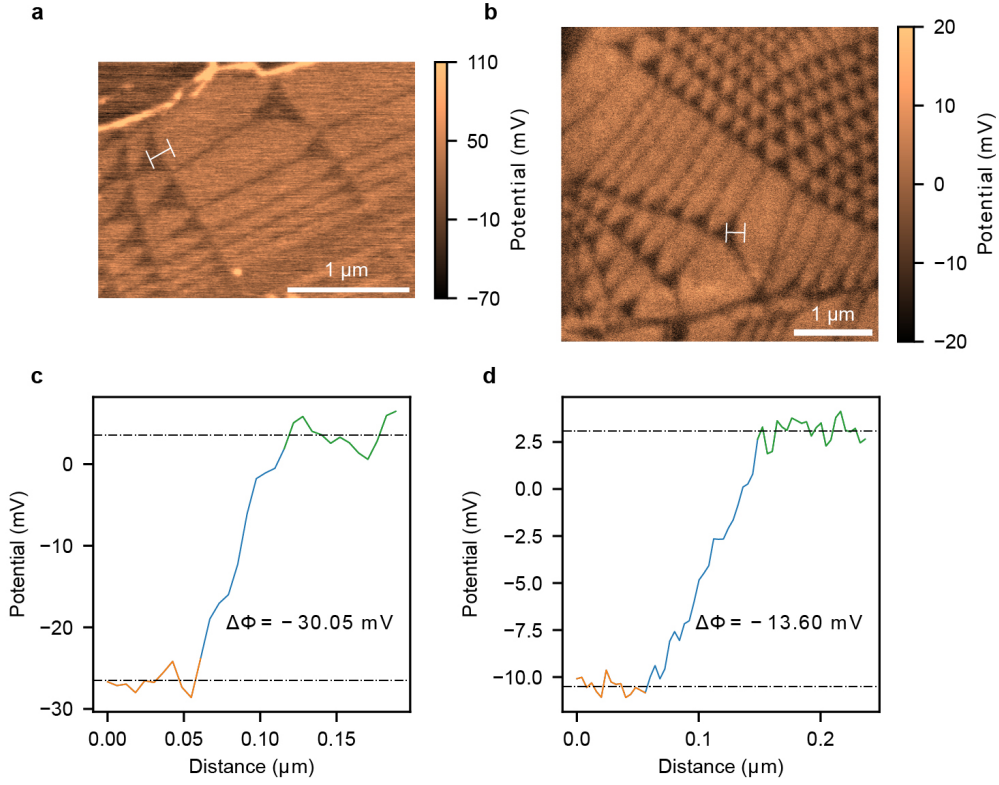


Fig. S14: Extraction of work function difference. **a,b**, Kelvin probe force microscopy (KPFM) images of mixed rhombohedral and Bernal structures taken at room temperature. The white lines indicate the positions of the line cuts shown in **c** and **d**. **c,d**, Surface potential profiles measured along the line cuts in **a** and **b**, respectively. The work function difference $\Delta\Phi = \Phi_R - \Phi_B$ between rhombohedral and Bernal stackings is extracted from the potential difference between rhombohedral (orange) and Bernal (green) regions.

16 Switching across all devices

We include a list of all the fabricated devices in Table 3. In total, we fabricated 15 dual-gated, twisted double bilayer graphene devices. Seven of these devices (devices X1 to X7) had major problems, including a small gate range, a gate that did not work properly, or a twist angle between the layers that was too large. The angle between the layers is sometimes off from the targeted twist angle, due to unintentional strain. For example, devices X3 and X4 had a final twist-angle misalignment between bilayers greater than 1 degree, creating a nanometer-scale moiré pattern instead of the micrometer-scale moiré pattern we require for the sliding transition.

Among the 8 devices without major problems, we observed gate-induced structural switching in 7 of them (devices A to G). We observed the anomalous Hall effect (AHE) in 4 of them (devices A to D), for which we show detailed measurements in the main text, Extended Data, and Supplementary Information. Devices E, F, and G show gate-induced structural switching, as shown in Fig. S15. In devices E and F, owing to the limited gate range, we could not measure the region of phase space where the AHE may appear (slightly electron-doped, large $|D|$ regions). In device G, while we observed gate-induced structural switching, we could only realize the Bernal-dominant state in the region of phase space where the AHE may appear. Here, we did not observe any AHE, consistent with the Bernal-dominant state. Device B is the only device in which we observed the AHE in both gate-history-dependent states. This is consistent with the weak structural switching in this device: the weak quantum oscillations across the entire gate-voltage range indicate that substantial rhombohedral and Bernal domains coexist even in the Bernal-favored state, and consequently the AHE is not completely

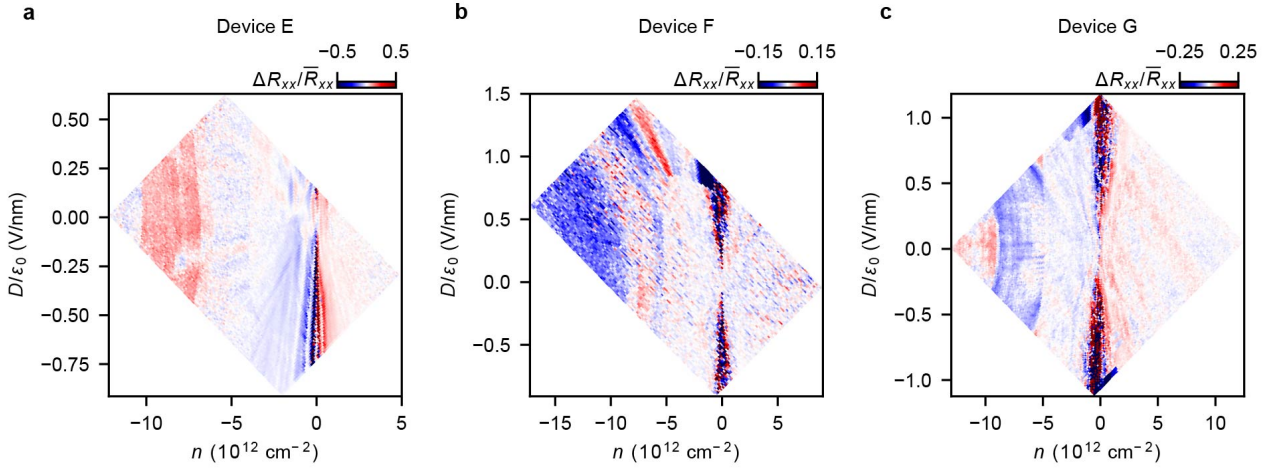


Fig. S15: Gate-induced structural switching in three other devices. a-c, Measurement of $\Delta R_{xx}/\bar{R}_{xx}$ on devices E-G following the same measurement scheme as in Fig. 2.

switched off, as discussed in “Transport measurement and domain configuration of device B” section of Supplementary Information.

Among the 8 devices without major problems, we did not observe gate-induced structural switching in 1 of them (device H). This device had a moderate gate range, around $-1.6 \times 10^{13} \text{ cm}^{-2}$ at $|D/\epsilon_0| > 0.5 \text{ V/nm}$, reaching similar levels to device F, but did not exhibit gate-induced structural switching. Although it is nontrivial to identify the reason why we do not observe structural switching, we speculate that it is related to the initial structural domain configuration of the device. Even in device A, where we observed substantial structural switching in two contact pairs, we did not observe any structural switching in the remaining two contact pairs, as illustrated in Extended Data Fig. 2. This observation suggests that the initial structural domain configuration of the device is critical for observing the structural switching. For example, we cannot achieve any switching if we had a pure Bernal tetralayer graphene device without a domain wall, which is possible through relaxation even if we made marginally-twisted double bilayer graphene. Therefore, it is not surprising that we do not observe the structural switching in one device out of eight.

To summarize:

Fabrication yield: 8/15 devices sufficiently functional for evaluating the gate-induced switching: They had no misalignment of twist angle and could reach a carrier density of at least $-1.3 \times 10^{13} \text{ cm}^{-2}$ (devices A to H)

Gate-induced structural-switching yield: 7/8 devices showed gate-induced structural (sliding) switching (devices A to G)

AHE switching: Devices A, C, and D show complete on/off switching; Device B shows incomplete switching. Devices E and F could not access the AHE-relevant phase space, and device G could access only the B-dominant state there.

Characterized devices	Lowest carrier density	Structural switching	AHE region accessible?	AHE observed?	Other comments
A	$-1.6 \times 10^{13} \text{ cm}^{-2}$	Yes	Yes	Yes	
B	$-1.4 \times 10^{13} \text{ cm}^{-2}$	Yes	Yes	Yes	Only device with AHE in both states, likely due to the mixed structure in both states.
C	$-1.7 \times 10^{13} \text{ cm}^{-2}$	Yes	Yes	Yes	
D	$-1.6 \times 10^{13} \text{ cm}^{-2}$	Yes	Yes	Yes	
E	$-1.3 \times 10^{13} \text{ cm}^{-2}$	Yes	Not accessible	N/A	Could not measure the region where the AHE exists.
F	$-1.6 \times 10^{13} \text{ cm}^{-2}$ only at $ D/\epsilon_0 > 0.5 \text{ V/nm}$	Yes	Not accessible	N/A	Could not measure the region where the AHE exists.
G	$-1.3 \times 10^{13} \text{ cm}^{-2}$	Yes	Yes, only in B-dominant state.	No	As we can only access the AHE region in the B-dominant state in this device, we observed no AHE.
H	$-1.6 \times 10^{13} \text{ cm}^{-2}$ only at $ D/\epsilon_0 > 0.5 \text{ V/nm}$	No	Yes, only in B-dominant state.	No	
Devices with major problems	Primary issue with device				
X1	Lowest carrier density is only $-0.6 \times 10^{13} \text{ cm}^{-2}$	N/A	N/A	N/A	
X2	Lowest carrier density is only $-0.5 \times 10^{13} \text{ cm}^{-2}$	N/A	N/A	N/A	
X3	Angle between bilayers > 1 degree	N/A	N/A	N/A	
X4	Angle between bilayers > 1 degree	N/A	N/A	N/A	
X5	Gates did not work properly, likely due to defective dielectric	N/A	N/A	N/A	
X6	Top gate does not function	N/A	N/A	N/A	
X7	Top gate shorted to device	N/A	N/A	N/A	

Table 3: Summary of all the devices we fabricated.

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