Films of rhombohedral graphite (RG; also described as chirally stacked ABC graphene multilayers) are predicted to possess nearly flat bands localized at the surfaces; these bands are particularly suitable for exploring electron–electron interactions. In RG, interlayer hopping (parameterized by $\gamma$; see Fig. 1a) dimerizes the electronic states of the opposite sublattices of all contiguous graphene layers in the bulk, shifting their energies away from the Fermi level. Electrons of only two sublattices, one in the top layer, and the other in the bottom layer, remain at low energies. These surface states are considered to be equivalent to topological edge states in the ‘polycyclic’ Su–Schrieffer–Heeger model. The surface states residing on the inequivalent sublattices of the opposite surfaces of $N$-layer-thick samples are expected to form nearly flat bands with approximate energy dispersion relations of the form $E \sim \pm p^4$ (refs. 3,12), where $p$ is the in-plane momentum. These surface states are further theorized to have strong electron–electron interactions, hypothetically leading to spontaneous quantum Hall states, ferromagnetism and superconductivity. When considering other hopping parameters ($\gamma_0$, $\gamma_1$, see Fig. 1a; Methods), the surface flat bands acquire a finite bandwidth of the order of $2\gamma_0/\gamma_1$ as well as electron–hole asymmetry, and trigonal warping due to $\gamma_1$ (Fig. 1b), which could affect electron–electron interactions, spontaneous symmetry breaking and the opening of a bandgap (Fig. 1c).

Owing to the metastable nature of RG, the reported transport experiments have been limited to ABC trilayer and ABCA tetralayer graphene, either in suspended two-terminal devices, or with the spectrum modified by moiré superlattices; longer ABC sequences have been studied using magneto-Raman and ARPES. Here we report electronic transport in high-quality RG of up to $N = 50$ graphene layers thick, where we observe strong electronic correlations and phase separation, phenomena typically reserved for unconventional superconductors and heavy fermion materials. Eight RG devices were studied in detail in this work, with thicknesses ranging from about 2.3 nm ($N = 7$) to about 16.5 nm ($N = 50$).

We identified the stacking order of graphite films using Raman spectroscopy and, to retain their high electronic quality, we encapsulated the exfoliated films with hexagonal boron nitride (hBN) crystals using the directional dry transfer technique (Methods). Figure 1d compares low-temperature ($T = 20$ mK) resistivity $\rho_{xx}$ of RG and hexagonal graphite (HG) devices of the same thickness (device 1, about 3 nm thick, 9 graphene layers thick) as a function of total carrier density $n = \frac{\rho_{xx}}{\rho_{xx}} < \frac{\rho_{xx}}{C_e}$. (Here $V_1$ and $V_2$ are the gate voltages, and $C_1$ and $C_2$ are the unit-area capacitances, of the top and bottom gates, respectively.)
elementary charge.) The RG device shows much stronger modulation of \( \rho_{xx}(n) \) as compared to the reference HG device, such that \( \rho_{xx} \) of the former spans 10 \( \Omega \) to 23 \( k\Omega \) and exhibits a profound high-resistivity region near \( n = 0 \). At the same time, the HG device displays metallic behaviour over the entire range of carrier densities (\( \rho_{xx} \) varies only from 8 \( \Omega \) to 63 \( \Omega \), similarly to the reported dual-gated devices of tetralayer RG.

Figure 1e shows three distinct high-resistance regions for RG device 1 (which are absent in the HG device) as a function of displacement field \( D = \frac{U}{n \varepsilon_{0}} \), measured at \( n = 0 \) (\( \varepsilon_{0} \) is the vacuum permittivity). The appearance of the high-resistivity region at the neutrality point (\( n = 0 \), \( D = 0 \)) suggests that an energy gap opens in the surface states spectrum, whereas the \( \rho_{xx} \) value close to the resistance quantum \( h/e^{2} \) hints at edge-state transport caused possibly by topological character of the insulating state. The resistivity peak at the neutrality point was observed only in sufficiently thin (<4 nm) RG devices and cannot be explained within a free-particle model (Methods). Before discussing this high-resistivity feature, we consider states induced by a large displacement field. These regions of high resistivity, observed in all RG devices, emerging above a critical displacement field, \( |D| > D_{c} \) (see Fig. 2a–d), are in agreement with the theoretical prediction that a high enough \( D \) opens an energy gap in the surface state spectrum of RG. The effect is similar to that observed in bilayer graphene under displacement but has much stronger nonlinear screening of charges at the two RG surfaces (Methods). Note that the observation of these displacement-induced gaps requires a gapped bulk of multilayer RG.
Effect of quantum spin Hall order parameter on the Landau spectrum. A pronounced gap at \( v = -18 \) followed by a series of crossings below 8 T could be traced back to the crossings between the 2N-fold degenerate zeroth LL and the valence-band LLs. The robust \( v = -18 \) state emerges when these crossings stop (Extended Data Fig. 4a). When only a single gate is used (Extended Data Fig. 5), the valley degeneracy becomes lifted, and a robust gap appears at \( v = -7 \) (for 7-layer-thick device 7), \( v = -9 \) (for 9-layer device 1) and at \( v = -11 \) (for 11-layer device 6). This provides additional support for our assignment of the observed spectrum to the \( N \)-degenerate zeroth LL, which is in turn a manifestation of an \( N \) Berry phase in \( N \)-layer RG.

Although the free-particle theory adequately describes the experimental results at high doping (Fig. 3a, d above \( n > 2.5 \times 10^{12} \text{ cm}^{-2} \)), it fails to explain the pronounced gaps observed at \( v = 0 \) and \( v = -6 \) (Fig. 3a). Apart from the crisscross pattern on the hole side (Fig. 3b), which agrees with the free-particle theory, there is a series of crossings on the electron side at the symmetry broken states \( v = 10, 14, 18, 22 \ldots \). The crossings are better seen if we replott the Landau fan as a function of \( v \) (Fig. 3c; the crossing points are marked with red arrows). These crossings also cannot be explained using the free-particle picture. To account for the latter features, we suggest a spontaneous appearance of an interaction-induced spin and/or valley symmetry-breaking order parameter. It leads to a bandgap when the resulting band splitting exceeds the single-particle overlap between the conduction and valence bands, thus allowing the observation of quantum Hall states at \( v = 0 \) and \( -6 \) (Fig. 3a, d). With increasing \( B \), the gap switches from \( v = 0 \) to \( v = -6 \). This can be explained by large orbital magnetic moments that have opposite signs for opposite spins (or valleys) and lead to closure of the \( v = 0 \) gap, as shown in Fig. 3d. The bandgap disappears with increasing doping, but the observed crossings on the electron side (Fig. 3c, and also Extended Data Fig. 4c, for 3.3-nm-thick device 5) suggest that a small interaction-induced order parameter remains present up to \( n = 2.5 \times 10^{12} \text{ cm}^{-2} \).

The interplay between the orbital, valley and spin degrees of freedom leads to several possible states with spontaneously broken symmetry in multilayer RG. They include quantum valley Hall, quantum anomalous Hall, layer antiferromagnetic insulator and quantum spin Hall states (see Methods). The crossing points on the electron side (Fig. 3c, and Extended Data Fig. 4d for similar results in device 5) can be explained by the phase transition from spin-polarized states due to Zeeman splitting at higher electron concentration to quantum spin Hall states at electron concentration \( n = 2.5 \times 10^{12} \text{ cm}^{-2} \) (Fig. 3e). The quantum spin Hall phase is accompanied by a giant Berry curvature and a related giant orbital magnetic moment (the same in both valleys but opposite for the two spin directions). The observed crossings cannot be explained by any other of the conjectured phases listed above. For instance, the
The hysteresis rapidly decreases with increasing $T$ and disappears if the RG enters a metallic state. Note that the hysteresis strongly favours electron doping, reflecting the electron–hole asymmetry of the RG band structure. In addition to the hysteresis as a function of $n$, we also observed pronounced hysteresis in response to both perpendicular ($B_\perp$) and parallel ($B_\parallel$) magnetic fields. $B_\perp$ suppresses the insulating state (Fig. 4b) but the accompanying Landau quantization unfortunately complicates analysis of the observed $T$ dependence. In contrast, measurements in $B_\parallel$ (Fig. 4c), where the orbital degree of freedom remains unaffected, provide a clearer picture. With increasing $T$, both the hysteresis and the transition field rapidly decrease, and the $B_\parallel$- $T$ phase diagram follows the power-law $T = (B_\perp - B_\parallel)^\alpha$ where $\alpha = 0.4$ is the critical exponent (Fig. 4d). Our fit is comparable to those reported for the ‘hidden order’ phase in strongly correlated heavy fermion systems like $^{27}$URu$_2$Si$_2$. The pronounced hysteretic behaviour with abrupt switching hints at spontaneous formation of insulating electronic phases that are separated into domains, similar to those observed in other correlated systems such as manganites, cuprates, and transition metal oxides. The hysteresis as a function of both electric and magnetic fields is characteristic of multiferroic materials, and advanced scanning probe techniques could be helpful in identifying the nature of those electronic domains in future.

The present work offers a novel experimental system in which to explore strongly correlated phenomena that does not rely on spin–orbit or exchange interactions in transition metal oxides or on precise structuring of graphene superlattices. Electronic coupling between top and bottom surfaces in thin RG films is similar to that observed in thin films of topological insulators and could be responsible for the spontaneous gap opening in our case. However, this coupling cannot explain the observed hysteretic behaviour. We believe that multilayer RG provides an interesting system in which to explore strong correlations, quantum criticality and other many-body phenomena, usually reserved for $f$-element-based heavy fermions or $d$-element-based transition metal oxides.

**Online content**

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shifted away from zero energy, leading to a bulk gap of about 3πγ/N for N layers, making only the surface bands relevant for electronic transport at low T. The surface states have shallow dispersion for quasi-momenta p < p_s = γ/ν, where ν is the Dirac velocity determined by intralayer nearest-neighbour hopping γ_o. There is also a quadratic term in the dispersion of surface states 2m_v^2/2 m^2 (with m^2 = 2 γ^2/2 γ_0^2 + δ/γ_0 = (0.4 m_e)^2), which breaks the electron–hole symmetry, and trigonal warping terms controlled by γ_e and γ_r. Here, m_e is the free electron mass. Since the SWMC parameters are poorly known for RG, we use the conventional HG values 23,36 for the larger hopping amplitudes (γ_e = 3.16 eV, γ_r = 0.39 eV, ν = 0.25 eV) while tuning the unknown γ_o to -0.01 eV and m^2 to 0.4 m_e to better describe the experiment.

We also set the energy difference of dimerized (that carry bulk states) and non-dimerized (that carry surface states) sublattices to zero, δ = 0, and tuned γ_e to about 0.13 eV. These parameter values have been chosen by comparing Fig. 3a with Extended Data Fig. 4a and matching the electron density of the first crossing of valence and conduction bands (n = -6.3 × 10^{12} cm^{-2}), which corresponds to the valence-band population where the chemical potential is at the bottom of the conduction band. Other crossings lead to extra constraints that can be satisfied reasonably well at positive filling factors (4 < ν < 40), where crossings are in the range 5 T < B < 2 T), while the matching becomes worse near zero filling and at larger magnetic fields, suggesting an increased role of interaction effects.

### Bandgap induced by displacement field

Introducing a vertical electric displacement field D via asymmetric gating leads to a potential difference Δ between the top and the bottom layers, resulting in a density redistribution that strongly screens the external electric field. The dispersion of surface states with non-zero Δ is shown in Extended Data Fig. 9. Note that the difference between the energies of valence- and conduction-band states at the K point equals Δ (only γ_e, vertical hopping and the Δ potential contribute to the Hamiltonian at K points), leading to states localized at A and B sublattices with energies ±Δ/2, while all the other states belong to dimers, connected by γ_r vertical hopping and having energies ±γ_r). After calculating the energy dispersion, the screening electron density accumulated at layer i is calculated as

\[ n_i = 2 \int_{BZ} \frac{d^2k}{(2\pi)^2} \sum_{l=1}^{2N} \left( |\Psi^l_A(k)|^2 + |\Psi^l_B(k)|^2 \right) f(e_i - E_i) - 1 \]

where is the layer number in N-layer RG, the integration is over the Brillouin zone (BZ), l stands for band index, Ψ^l_A(k) and Ψ^l_B(k) are normalized wavefunctions of two sublattices in layer i, E_i is the Fermi function, E_i is the Fermi level, e_i is the energy of band i, and –1 subtracts a homogeneous charge jelly that neutralizes the Fermi sea electron density. Denoting the total screening charge accumulated near the top and bottom surfaces by n^{scr}_B(Δ) and n^{scr}_A(Δ) respectively, we relate the displacement field to the charge density and the potential asymmetry as

\[ \frac{e(n_A - n_B)}{2 \varepsilon_0} = D = \frac{e(n^{scr}_B(Δ) - n^{scr}_A(Δ))}{2 \varepsilon_0} + \frac{e D}{d} \]

where e is the electron charge, d is the thickness of the graphite film, and \( \varepsilon_0 \) is the vacuum permittivity. We employed the Hartree approximation and used the fact that the dominant screening charge is localized at the surface layers. By varying the potential difference of the surface layers, tuning the chemical potential to maintain overall charge neutrality, and finding the electron density distribution, we determined the relation between the bandgap and applied external displacement field, as shown in Fig. 2f inset. The bandgap Δ, defined as the difference between the conduction-band minimum and the valence-band maximum, is shown in Extended Data Fig. 9.

### Surface and bulk contributions

Extended Data Figure 8 shows the temperature dependence of resistivity, \( \rho_{xx}(T) \), at n = 0 for 16.5- and 3.3-nm-thick RG (devices 4 and 5, respectively). In contrast to the metallic behaviour of Bernal-stacked graphite 23, we observed a semiconductor-to-metal-transition with decreasing temperature. We attribute this transition to the interplay between rhombohedral stacking and Bernal stacking occurs during the shift of graphene layers along armchair directions, whereas it does not happen along zig-zag directions.

To retain the high electronic quality of RG, we used a directional van der Waals assembly technique as follows. First, we identified the crystal orientation of graphite from the edge chirality of graphite films using Raman spectroscopy. Next, we used a polymer (a polymethylmethacrylate (PMMA)/polydimethylsiloxane (PDMS) stack mounted on a glass slide)-mediated dry transfer method to encapsulate RG films in hexagonal boron nitride (hBN). During the encapsulation process, the pressing-down direction of the PDMS/PDMS layers was controlled along the zig-zag direction of the graphite films, so that the stacking transformation could be avoided. Then, we used the above-mentioned Raman-ratio map again to confirm that rhombohedral stacking was retained in the hBN-encapsulated graphite. Finally, multiterminal Hall bars were fabricated on the hBN/RG films/hBN heterostructures using standard electron-beam lithography. Examples of our Raman analysis during device fabrication are presented in Extended Data Fig. 7 for device 6 (3.6-nm-thick RG).

### Tight-binding model

To describe multilayer RG, we employ the Slonczewski–Weiss–McClure (SWMC) parameterization of graphite 20,24,26,36, with interlayer hopping parameters that take into account the lattice arrangement of rhombohedral graphite, as shown in Fig. 1a. Multilayer RG has bulk subbands respectively, we relate γ_δ and δ respectively, we relate γ_δ and δ respectively. In contrast to the metallic behaviour of Bernal-stacked graphite we observed a semiconductor-to-metal-transition with decreasing temperature. We attribute this transition to the interplay between rhombohedral stacking and Bernal stacking occurring during the shift of graphene layers along armchair directions, whereas it does not happen along zig-zag directions.
Possibility of stacking faults

It is important to have a reliable method to check for possible stacking faults in the sample. Apart from Raman (Extended Data Fig. 1a–d) and electronic Raman scattering signatures\textsuperscript{35}, measurements of an in-plane transport gap opened by the vertical displacement field (asymmetric gating) provides another diagnostic tool (Extended Data Fig. 1e–g). Our calculations reveal that it is impossible to open a transport gap if stacking faults or thicker inclinations of ABA stacking are present. We considered the most elementary ABCBA stacking fault that joins ABC stacking with its mirror-image CBA stacking (Extended Data Fig. 2a); a Bernal stacking fault that joins two ABC multilayers, as shown in Extended Data Fig. 2d for the ABCBCA multilayer; and surface stacking fault ABCAC (Extended Data Fig. 2g). The bold characters indicate the type and position of stacking faults.

At the K points of the Brillouin zone of RG, the in-plane hoppings \( y_0 \) destructively interfere, leaving only \( y_1 \) as a high-energy scale. For the twin-boundary stacking, the low-energy states are localized on sublattices A\(_1\), A\(_5\) and B\(_4\), and on a zero-energy eigenstate \( \psi_{1,y_0} \) of a B\(_2\)A\(_3\)B\(_4\) trimer, which we denote as B\(_2\)–B\(_4\). Extended Data Fig. 2a. Away from the K point, chiral decomposition\textsuperscript{31} leads to the appearance of effective 2- and 3-layer ABC chiral subsystems with obvious generalization to films thicker than 5 graphene layers. So, there are four low-energy sub-bands: two with energies growing away from the K point (valence-band-type) and two with energies decreasing away from the K point (valence-band-type). Similarly to RG discussed before, the hopping \( y_1 \) leads to an overlap of valence and conduction sub-bands of the order of 20 meV. Application of the vertical electric field (creating layer potentials \( U_1 < U_2 < \ldots < U_N \) splits the four almost degenerate sub-bands into two outer valence and conduction bands, localized on A\(_1\) and A\(_5\) (with energies \( U_1 \) and \( U_5 \) at the K point), and two inner valence and conduction bands, localized on B\(_1\) and B\(_5\) (with energies \( U_3 \) and \( U_7 \) at the K point). The energy splitting of the two inner bands is \( U_3^2 - U_7^2 = 2\left( d^2 \alpha \right) \), where \( d = 0.335 \) nm is the distance between layers, and it only depends on the charge density of the third layer, \( n_{0,3} \).

For the ABCBCA stacking fault in Extended Data Fig. 2d, the low-energy states at the K point are localized on sublattices A\(_1\), B\(_3\), A\(_4\) and B\(_4\), with the splitting of the inner band determined by the electric field between layers 3 and 4. In both ABCBA and ABCBCA stacking fault cases, to open a gap one requires a displacement field sufficient to overcome the doubled screening of four low-energy bands and, additionally, to create a potential difference of about 20 meV between the two consecutive layers forming a stacking fault. Such displacement fields would strongly exceed the values plotted in Fig. 2e, and are beyond our experimental reach. Large but experimentally attainable displacement fields would split the inner B\(_1\) and A\(_4\) bands at the K point, as shown in Extended Data Fig. 2e, f, but such high fields would also substantially increase the dispersion of these bands, resulting in band overlap near \( p = p_0 \), thus closing the gap. As for the ABA surface stacking fault in Extended Data Fig. 2g–i, the ABA surface bands overlap with a Dirac cone having velocity \( v_1/\sqrt{2} \), which shunts the gapped state.

Thus we conclude that any stacking faults make opening of a transport gap impossible. We also note that stacking faults would, generally, lead to asymmetric behaviour with respect to reversals of the direction of \( D \). Calculated dispersions with non-zero displacement field are shown for the twin-boundary defect in Extended Data Fig. 2b, c for the stacking fault buried inside the ABC bulk in Extended Data Fig. 2e, f, and for the surface stacking fault in Extended Data Fig. 2h, i. Examples of transport data for multilayer graphite devices with stacking faults are shown in Extended Data Fig. 1. The presence of faults is established by measuring Raman spectra.

Landau levels in multilayer RG

We have calculated the LL spectrum by employing the same method as in ref. \textsuperscript{36}. Numerical diagonalization is performed in a regularized finite basis consisting of oscillator states for each sublattice component, namely \( \left( \psi_{1,y_0}, \psi_{1,y_1}, \ldots, \psi_{1,y_N} \right) \) for sublattice A\(_1\), and \( \left( \psi_{2,y_0}, \psi_{2,y_1}, \ldots, \psi_{2,y_N} \right) \) for B\(_2\), where \( N_{AB} = 250 \) is a cut-off LL index, sufficient for convergence at \( B > 0.5 \) T.

To qualitatively understand the LL spectrum, let us start with the minimal model\textsuperscript{34} with only the nearest-neighbour in-plane \( (y_{ij}) \) and interlayer \( (y_{ij}) \) hoppings. In the absence of a displacement field \( (D = 0) \), the low-energy LL spectrum consists of \( 4N \)-degenerate levels at zero energy, which we refer to as zeroth LLs (where 4 is due to double-spin and double-valley degeneracy, and \( N \) comes from \( N \)-fold orbital degeneracy), plus four-fold degenerate levels arising from the conduction and valence bands that disperse as \( E = \pm B N/2 \). Within the two-band model, the vector wavefunctions of zeroth LLs of one valley reside on one (for example, the top) surface, \( \psi_{N_{AB}} \), and for the other valley the wavefunctions sit on the opposite (for example, the bottom) surface, \( \psi_{N_{AB}} \). Therefore, for zeroth LLs the valley degeneracy is equivalent to the top/bottom surface degeneracy.

The minimal model is insufficient to describe the experimental results, making it necessary to consider further electron hopping amplitudes. The presence of inter-layer same-sublattice hopping \( y_1 \) leads to strong electron–hole asymmetry and breaks the orbital degeneracy of zeroth LLs, leading to dispersion \( E_n = n \hbar \omega_c \), \( n = 0, \ldots, N - 1 \), with \( \omega_c = e B/m^* \), \( m^* = 0.4 m_e \). The conduction-band LLs continue the sequence of four-fold degenerate LLs as \( E_n = n \hbar \omega_c \), \( n = N, N + 1, \ldots \). The valence-band dispersion is non-monotonic and produces a series of four-fold degenerate LLs \( E_n = n \hbar \omega_c \), \( n = 1, \ldots, \) near zero energy, as well as a sequence of 12-fold degenerate levels at higher energies, originating from the three equivalent valence-band maxima caused by trigonal warping (hoppings \( y_3, y_4 \)).

At moderate \( B \), valence-band LLs bend down, forming numerous crossings with zeroth LLs and conduction-band LLs\textsuperscript{35}. This explains the multiple crossings of the LLs on the hole side in Fig. 3a, b. The two-fold degeneracy of the filling factors (for example, \( n = 6, 8, 10, \ldots \)) at \( D = 0 \) can be attributed to the preserved valley degeneracy, while the gaps are given by combination of the orbital splitting (\( n = 8, 10, \ldots \)) for odd \( N \) and spin splitting (\( n = 4, 8, 12, 16, \ldots \)) for odd \( N \) and \( n = 6, 10, 14, \ldots \) for even \( N \). Although the theory qualitatively agrees with the experiment, it predicts a twice higher value of \( B \) at which the last crossings occur. This discrepancy can be attributed to exchange interactions.

The filling factor at a specified chemical potential is determined as the number of filled LLs in the regularized basis, with half the total number of levels subtracted to account for the background charge neutralizing the Fermi sea. Normally, this would place the zero electron density between LLs originating from valence and conduction bands that form the pairs related by approximate electron–hole symmetry \( (\psi_{1,y_0}, \psi_{1,y_1}) + (\psi_{1,y_1}, \psi_{1,y_0}) \). However, there are \( N \) additional (spin and valley degenerate) zero LLs (numbered as \( 0, \ldots, N - 1 \)) with wavefunctions localized only on one of the sublattices. At low \( B \), zero LLs overlap with valence-band LLs, leading to complicated counting of the filling factors but, for \( B \geq 10 \) T, conventional valence-band LLs are below the zero energy, leading to zero electron density \( (n = 0) \) located in the middle point of zeroth LLs. This middle point is located either in between the
Zeeman-split zeroth LLs with index \((N-1)/2\) for odd \(N\), or between the orbital-split zeroth LLs with indices \(N/2-1\) and \(N/2\) for even \(N\).

Spin- and valley-polarized states

Coulomb and phonon-mediated interactions between electrons can lead to a multitude of patterns for spontaneous symmetry breaking. These include breaking a ‘top–bottom’ symmetry of the two surfaces, an spin/valley symmetry, a six-fold rotation symmetry (nematic phase), or a \(U(1)\) gauge symmetry (superconductivity)\(^{36-40}\). The presented experiments provide evidence that symmetry breaking occurs for a limited range of sample thicknesses (<10 layers), hinting at the importance of interaction between the surfaces, which suggests a ‘top–bottom’ type of symmetry breaking order. The candidates are thus the quantum valley Hall (VH; ferroelectric) phase with order parameter \(\Delta^{\text{VH}} = 0\) \(\lambda\) \(\sigma_z^{\text{layer}}\), quantum layer antiferromagnetism (LAF) \(\Delta^{\text{LAF}} = \lambda\) \(\sigma_z^{\text{layer}} \otimes \sigma_z^{\text{spin}}\), the quantum spin Hall (QSH) phase (also called the ‘spin flux’ phase) \(\Delta^{\text{QSH}} = \lambda\) \(\sigma_z^{\text{layer}} \otimes \sigma_z^{\text{spin}} \otimes \sigma_z^{\text{valley}}\), or the quantum anomalous Hall (AH) phase \(\Delta^{\text{AH}} = \lambda\) \(\sigma_z^{\text{layer}} \otimes \sigma_z^{\text{spin}} \otimes \sigma_z^{\text{valley}}\). Here, \(\sigma_z\) is a Pauli matrix. In the absence of a \(B\) field, only the relative orientation of spins matters, while the presence of a \(B\) field lifts this degeneracy to fix an optimal ‘canted’ orientation of spins\(^{41}\). The AH phase breaks the time-reversal symmetry and implies a Hall signal at \(B = 0\), which has not been observed. The VH phase is the one induced by displacement field and discussed above, but it is not continuously connected to the observed spontaneous insulating phase, and, instead, competes with it. So we are left with two candidates: LAF and the QSH effect. Both phases lead to giant Berry curvatures and, consequently, to large orbital magnetic moments, which peak near the three degenerate maxima of the valence band\(^{17}\). The orbital magnetic moment has opposite signs in the two valleys for the \(\Delta^{\text{QSH}}\), quantum layer antiferromagnetism (LAF) order parameters, leading to pronounced lifting of the 12-fold degeneracy of LLs originating from valence-band maxima, which results in two groups of six-fold LLs. This explains the observed robust gap at \(\nu = -6\) (Fig. 3a, b, d). The six unoccupied valence-band LLs lead to the layer-antiferromagnet (three \(K\) and three \(K'\) levels) in the case of the quantum LAF order parameter and to a ferromagnet (three \(K\) and three \(K'\)) in the case of QSH ordering. Canting effects can be neglected since the magnitude of the order parameter is much larger than the Zeeman splitting.

The effect of order parameters on the spectrum of zeroth LLs follows from localization of zeroth LLs on the top/bottom surfaces in the K/K’ valleys. Hence, \(\Delta^{\text{LAF}}\) breaks the valley degeneracy, leading to the appearance of gaps at odd filling factors (Fig. 3f) whereas the valley degeneracy remains intact for \(\Delta^{\text{QSH}}\) (Fig. 3e). The experimental data in Fig. 3c are consistent with the QSH order for electron doping below \(2.5 \times 10^{12} \text{ cm}^{-2}\), where the orbital and spin magnetic moments align. Note that a QSH order parameter fits finite magnetic field data at substantial electron doping, and does not necessarily connect to the observed spontaneous gap near charge neutrality.

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Author contributions

A.M. conceived the presented idea and directed the project. Y.S., S.V.M., S.O., C.M., J.B., J.Y., A.I.B. and B.A.P. performed transport measurements. S.X., Y.Y. and S.-K.S. fabricated devices. Y.S., S.O., C.M. and J.Y. performed data analysis. S.S. and V.I.F. developed the theory and performed theoretical calculations. Y.S., S.O., A.K.G., V.I.F. and A.M. contributed to the interpretation of data. K.W. and T.T. grew hBN single crystals. A.M., Y.S., A.K.G., S.S., V.I.F., S.O., C.M., J.B., J.Y., A.I.B. and B.A.P. performed transport measurements.

Additional information

Correspondence and requests for materials should be addressed to A.M. Peer review information Nature thanks Dmitri Efetov and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

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Extended Data Fig. 1 | Effect of stacking sequence on the displacement-field-induced bandgap. a, b, Raman maps of device 3, a 6.5-nm graphite flake with domains of differing local stacking sequence, shown before (a) and after (b) encapsulation. The colour coding indicates the ratio of the integrated area of the low-frequency component of the graphite Raman 2D band (ranging around 2,670–2,700 cm\(^{-1}\)) to that of the high-frequency component (ranging around 2,700–2,730 cm\(^{-1}\)). Coloured dots in a and b mark the positions where the Raman spectra shown in c and d respectively were taken. Scale bars, 10 μm.

c, d, Typical 2D Raman peaks of HG (black curves A and a), RG (red curves C and c) and graphite of mixed ABA and ABC stacking (blue curves B and b), before (c) and after (d) encapsulation. Laser wavelength, 532 nm. e, \(\rho_{xx}\) as a function of \(D\) for Hall bar devices made using domains at point b (light blue) and point c (red), respectively, \(T = 1.6\) K. Inset, optical micrograph of the devices before shaping them into the Hall bar geometry. f, g, Resistivity maps \(\rho_{xx}(n_b, n_t)\) of graphite with ABC stacking (f) and mixed stacking (g); \(T = 1.6\) K, \(B = 0\) T.
Extended Data Fig. 2 | Band structures of multilayer RG with stacking faults. 

a, Schematic of twin-boundary defect. b, c, Calculated spectra for the ABCABCABCABC sequence at positive (b) and negative (c) \( D \).

d, Schematic of a buried Bernal stacking fault ABCBA. e, f, Calculated band structure for such a defect: ABCABCABCABC sequence at positive (e) and negative (f) \( D \).

g, Schematic of surface stacking fault ABCAC. h, i, Spectra for the ABCABCABCABCABCABC sequence at positive (h) and negative (i) \( D \). In all the calculations we used \( D = \pm 1 \text{ V nm}^{-1} \). See Methods section ‘Possibility of stacking faults’ for details of nomenclature used in a, d, g. \( E \) is the band energy, \( \rho \) is the in-plane momentum, and \( \rho_c = \gamma_1/v \), where \( v \) is the Dirac velocity.
Extended Data Fig. 3 | The quantum Hall effect in thin RG. a, Hall resistivity $\rho_{xy}$ (red curve) and longitudinal resistivity $\rho_{xx}$ (black curve) as a function of magnetic field $B$ measured at 20 mK for the same device as in main text Fig. 3 (device 1); $n = 2.3 \times 10^{12}$ cm$^{-2}$, $D = 0$ V nm$^{-1}$. b, $\rho_{xy}$ and $\rho_{xx}$ as a function of $n$ for the same device as in a; $D = 0$, $B = 10$ T, $T = 20$ mK.
Extended Data Fig. 4 | Landau levels from surface states of multilayer RG.

a, Calculated free-particle spectrum. b, Conductivity map $\sigma_{xx}(n_t, n_b)$ for 3-nm-thick RG (device 1) measured at $B = 9\, \text{T}$ and $T = 20\, \text{mK}$. Note that, for two independent surfaces, LLs should form sets of horizontal and vertical lines. The observed behaviour suggests that top and bottom surfaces of our RG devices are nearly independent 2D systems. c, Landau fan $\sigma_{xx}(n, B)$ for device 5 (3.3-nm-thick RG); $D = 0, T = 0.25\, \text{K}$. d, Differential $d\rho_{xx}/dv(n, B)$ map on the electron side (same device as in e). The red arrows indicate LL crossings. e, $\sigma_{xx}(n_b, B)$ for device 8 (7.2-nm-thick RG) measured at $T = 1.7\, \text{K}$. f, $\sigma_{xx}(n, B)$ for device 4 (16.5-nm-thick RG) at $D = 0$ and $T = 0.25\, \text{K}$. 
Extended Data Fig. 5 | Single-gate ($D \neq 0$) Landau fan diagrams highlighting the robust $\nu = N$ quantum Hall state in $N$-layer-thick RG.

a, Conductivity map $\sigma_{xy}(\nu, B)$ for 7-layer-thick RG (device 7).

b, Conductivity map $\sigma_{xx}(\nu, B)$ for 9-layer-thick RG (device 1).

c, Conductivity map $\sigma_{xx}(\nu, B)$ for 11-layer-thick RG (device 6).
Extended Data Fig. 6 | See next page for caption.
Extended Data Fig. 6 | Insulating states and hysteretic behaviour in multilayer RG. a, b, Additional data for 3.3-nm-thick RG (device 5, same as in main text Fig. 4a, b). a, Main panel, temperature dependence of $\rho_{xx}(n)$ around the insulating state, $B = 0$ T. Inset, Arrhenius plot for the peak resistivity, indicating the presence of a bandgap of about 2–3 meV. The y axis shows peak resistivity, and red line indicates the fit. b, Main panel, histogram of the conductivity values found on the hysteretic curves $\sigma_{xx}(n)$, such as those shown in the inset. Eighty-three such curves were used to make the histogram, where data from forward and backward sweeps are plotted in blue and red, respectively. $D = 0, T = 250$ mK, $B = 0$. c–e, Hysteresis (shaded red) in $\rho_{xx}(B)$ observed in device 7 (2.3-nm-thick RG) at the charge neutrality point; d, hysteresis (shaded red) in $\rho_{xx}(n)$ at $B = 5$ T. For c and d, $T = 250$ mK. e–g, Hysteretic behaviour of device 1 for different cooling cycles. No noticeable hysteresis was found for the first cooling event (e). Hysteresis in $\rho_{xx}(n)$ (f) and $\rho_{xx}(D)$ (g) is clearly seen after another cooling. Solid (dashed) lines indicate positive (negative) sweep directions and the coloured areas highlight the difference between the sweep directions. $T = 20$ mK; $B = 0$ T.
Extended Data Fig. 7 | Stacking order of device 6 at different fabrication stages. a, Optical image of the graphite flake (left) and the corresponding Raman map with a step size of 0.8 μm × 0.8 μm (right). Scale bar, 10 μm. b, Optical image of the graphite flake encapsulated by hBN (left) and the corresponding Raman map with a step size of 0.7 μm × 0.7 μm (right). Scale bar, 10 μm. c, Optical image of the finished Hall bar (left) and the corresponding Raman map with a step size of 0.5 μm × 0.5 μm (right). Scale bar, 4 μm. The colour coding of the Raman maps in a–c indicates the ratio of the integrated area of the low-frequency component (ranging around 2,635–2,665 cm⁻¹) to that of the high-frequency component (ranging around 2,665–2,695 cm⁻¹) of the graphite Raman 2D band. The coloured dots in a–c mark the positions where the Raman spectra shown in e were taken. d, Optical image showing that the Hall bar device is made from the ABC stacked region. Scale bar, 10 μm. e, Typical 2D Raman peaks of RG (dots a, c and d) and of graphite of mixed ABA and ABC stacking (dot b). Laser excitation wavelength, 633 nm.
Extended Data Fig. 8 | Temperature dependence of resistivity. a, $\rho_{xx}$ as a function of $T$ at zero gate doping for RG device 4 with a thickness of 16.5 nm (blue solid curve), for RG device 5 with a thickness of 3.3 nm (red solid curve), and for 6-nm HG (grey dashed line). b, Same data as a but plotted on a log scale. While cooling down, $\rho_{xx}$ of RG first increases for $T > T_c$ and then decreases, in sharp contrast to the monotonic decrease of $\rho_{xx}$ for HG. The critical temperature $T_c$ decreases with increasing thickness of RG. Besides the presence of $T_c$, $\rho_{xx}$ of the 3.3-nm RG device 5 shows a sharp increase for $T < 6$ K owing to the phase transition to the insulating state.
**Extended Data Fig. 9 | Bandgap opening by displacement field.**

**a**, Band dispersion of RG under an applied displacement field; bandgap \( \tilde{\Delta} \) is masked by a bandwidth of \( 2\gamma \gamma \gamma /\gamma \gamma \) such that only the effective gap \( \Delta = \tilde{\Delta} - 2\gamma \gamma \gamma /\gamma \gamma \) is visible in transport measurements. **b**, Calculated dependence of \( \tilde{\Delta} \) and \( \Delta \) on displacement field for \( N = 9 \) layers.