

Stability Diagram of Layer-polarized Quantum Hall States in Twisted Trilayer Graphene

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1. Calculation of total charge carrier density and displacement field from gate capacitive coupling.

To calculate the total charge carrier density n (that is subsequently used to find the overall Landau level filling and total Chern number) and displacement field D in tTLG between the top graphite and silicon back gate, we follow a ubiquitously used (for example, in^{1,2}) parallel-plate capacitor model:

$$n = \frac{C_{TG}V_{TG} + C_{BG}V_{BG}}{e} + n_0, \quad (S1)$$

$$D = \frac{-C_{TG}V_{TG} + C_{BG}V_{BG}}{2} + D_0. \quad (S2)$$

In these formulas, $V_{TG}(V_{BG})$ is the voltage applied to the top (back) gate; $C_{TG}(C_{BG})$ are the top(back) gate capacitances per unit area; n_0 , D_0 are offset possibly due to Schottky barriers at the layer interfaces or slight intrinsic doping of graphene; e is the elementary charge. The top gate geometric capacitance per unit area is calculated from $C_{TG} = \epsilon_{hBN}/d_t$, where the hBN permittivity is $\epsilon_{hBN} = 3.76\epsilon_0$, ϵ_0 is the permittivity of vacuum, $d_t = 57$ nm is the thickness of hBN between the top gate and tTLG. The back gate has a capacitance of two capacitors in series with the respective separations equal the SiO₂ and bottom hBN (Fig. 1a) thicknesses. Thus, the geometric capacitance of the back gate is estimated according to $1/C_{BG} = 1/C_{SiO_2} + 1/C_b$, where $C_{SiO_2} = \epsilon_{SiO_2}/d_{SiO_2}$ and $C_b = \epsilon_{hBN}/d_b$ with the SiO₂ permittivity $\epsilon_{SiO_2} = 3.9\epsilon_0$; and SiO₂ and bottom hBN dielectric thicknesses $d_{SiO_2} = 285$ nm and $d_b = 13$ nm respectively. The total distance between the tTLG and silicon back gate introduced in the main manuscript is $d_{bg} = d_b + d_{SiO_2} = 298$ nm. The value of $n_0 \sim 1 \times 10^{11}$ cm⁻² is found from the gate voltage needed to compensate for the offset of the charge neutrality resistance peak from zero charge carrier density. Same voltages give an estimate for D_0/ϵ_0 to be ~ 0.01 V/nm.

2. Data from additional region of measured device.

The longitudinal, R_{xx} , (transverse, R_{xy}) resistances presented in the main manuscript are calculated from the voltage measured between contact 9 and contact 8 (contact 9 and contact 3) under a 10 nA current driven between contact 1 and contacts 6 shown in Figure 1b. The key results discussed in the main text have been reproduced in a different region of the same device where R_{xx} (R_{xy}) was measured between contact 8 and contact 7 (contact 8 and contact 4) under the same 10 nA current excitation between contact 1 and contacts 6.

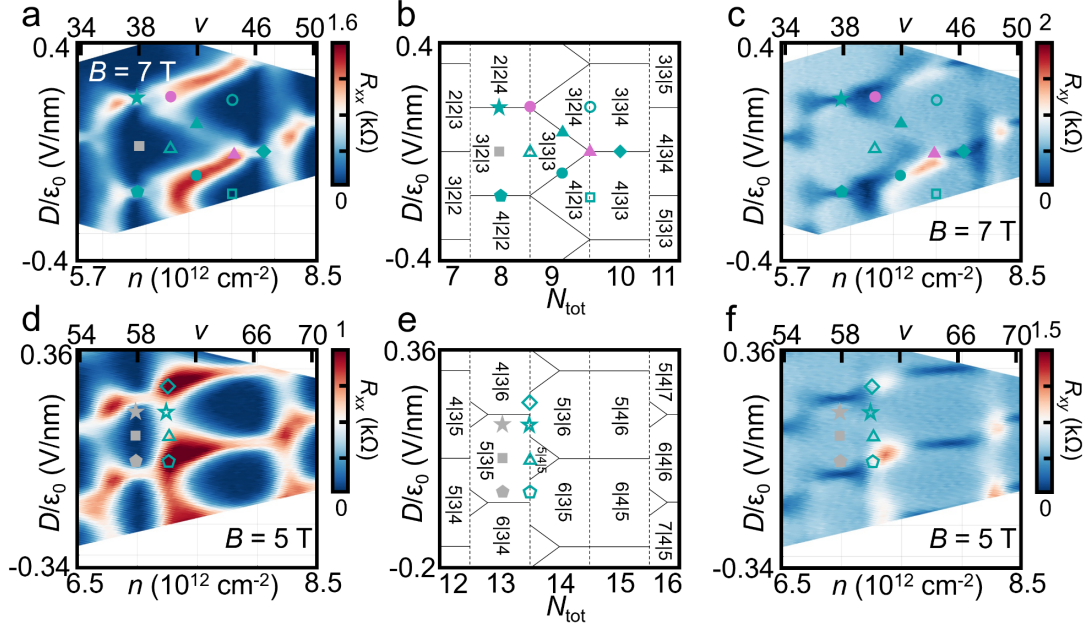


Fig. S1. Results from additional region of the device. (a) Longitudinal resistance (R_{xx}) measured in an additional region of the device as a function of charge carrier density n and displacement field D at 7 T. (b) The stability diagram [outlining transitions from (a)] showing layer-polarized quantum Hall states with an integer number (matching the layer Chern number) of filled fourfold degenerate LLs in each graphene layer (top|middle|bottom = $N_T|N_M|N_B$) with the total number of filled LL N_{tot} . Transitions between layer-polarized QH states are traced by solid (constant N_{tot}) and dashed (changing N_{tot}) lines. Selected transitions are labeled by grey, blue and red markers of various shapes consistent with the main manuscript. (c) Transverse Hall resistance (R_{xy}) measured in the additional region of the device at varying n and D under $B = 7 \text{ T}$. (d), (e), (f) same as (a), (b), (c) but measured at $B = 5 \text{ T}$.

Figure S1 summarizes magneto-transport data in the second region of the device reproducing the main features of the stability diagrams of the layer-polarized quantum Hall (QH) states described in the main manuscript. At 7 T, the measured longitudinal resistance R_{xx} (Fig. S1a) as a function of n and D reproduces Fig. 2b with transitions between layer-polarized quantum Hall states summarized in the same stability diagram (Fig. S1b) as in the main manuscript (Fig. 2c). Each layer-polarized QH state is characterized by layer Chern numbers $N_T|N_M|N_B$ equal to the number of filled fourfold-degenerate Landau levels (LLs) in the top|middle|bottom graphene. As in the main manuscript, two types of transitions between layer-polarized QH states are observed. The first type is interlayer transitions (labeled with solid blue and red markers in Figs. S1a, b) when the Chern numbers of two selected layers are being redistributed under changing D field

while the total Chern number N_{tot} remains the same. At these transitions, LLs of two layers are partially filled resulting in dissipative bulk conduction of two layers manifested as resistance peaks in Fig. S1a. The second type of transitions are intralayer ones (hollow markers in Figs. S1a, b) when only the middle layer becomes dissipative while its layer Chern number is changing. Figure S1a reproduces the triangular low-resistance domains corresponding to the $3|3|3$, $3|2|4$, $4|2|3$ layer-polarized QH states being a signature feature of the tTLG system.

The behavior of R_{xy} as a function of n and D at $B = 7$ T is also reproduced in the second region of the device (Fig. S1c). At layer-polarized QH states, R_{xy} reaches a constant value $1/G_{xy}$, where $G_{xy} = 4(3/2 + N_{\text{tot}}e^2/h)$ is consistent with the layer Chern number assignment in Fig. S1b. As in the main manuscript, R_{xy} changes non-monotonically at interlayer transitions reaching a local minimum at transitions involving the top and bottom layers and a maximum at transitions with two adjacent layers. As discussed in the main text, such unusual behavior signifies a non-trivial Coulomb coupling between partially filled LLs in two layers with a potential of realization of exciton condensates and Coulomb drag in the tTLG platform.

The magneto-transport data at 5 T from the main text (Fig. 3) is also mainly reproduced in the second region of the device (Figs. S1d to S1f) exhibiting the same transitions between layer-polarized QH states. However, in the second region, the triangular domains of R_{xy} approaching zero are not well-developed implying a sensitive dependence of partially filled LL coupling on the exact angle combination tuning the interlayer interactions.

3. Characterization of two moiré periodicities.

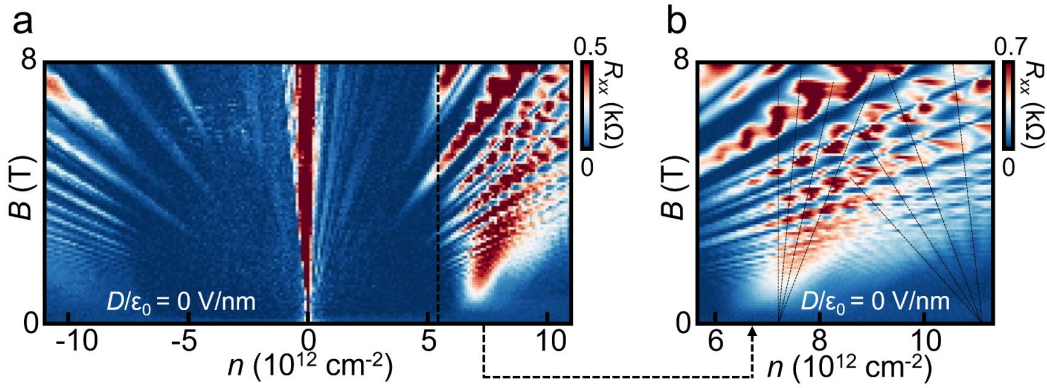


Fig. S2. Effect of moiré periodicities on magneto-transport. (a) Longitudinal resistance R_{xx} as a function of charge carrier density n and magnetic field B in the second region of the device at $D/\epsilon_0 = 0 \text{ V/nm}$. (b) Zoomed-in scan of (a) with two satellite Landau fans (highlighted by the dashed lines) due to the two moiré periodicities. The fans emanate from the charge carrier densities corresponding to four electrons per respective moiré superlattice supercell.

To confirm the three graphene layers being tunnel-coupled instead of strongly hybridized, we perform magneto-transport measurements to see the effect of the two moiré periodicities between adjacent layers. Figure S2 presents R_{xx} (as an example, measured in the same region of the tTLG device as in Supporting Information-2) as a function of magnetic field B and varying charge carrier density n at constant displacement field $D/\epsilon_0 = 0 \text{ V/nm}$. At $B = 0$ T, there is no signature of resistance peaks corresponding to band insulator states expected from either of the

moirés to be near $n = 9 \times 10^{12} \text{ cm}^{-2}$ for the $\sim 2^\circ$ twist angles thus confirming the weak hybridization between the layers. Under finite magnetic field, in addition to the Landau fan at the charge neutrality point ($n = 0$), R_{xx} exhibits a pair of satellite fans (highlighted with dashed lines in Fig. S2b, a zoomed-in scan of Fig. S2a) emanating from $n_1 = 7 \times 10^{12} \text{ cm}^{-2}$ and $n_2 = 11 \times 10^{12} \text{ cm}^{-2}$ corresponding to four electrons per supercell of the two moiré interfaces. Interestingly, at hole doping, no satellite Landau fans are present. This can potentially be due to the electron-hole asymmetry of the bands of each constituent MLG layer resulting in reduced interlayer coupling and thus ill-defined moiré band gap. From the satellite fan positions, the twist angles $\theta_1 = 1.7^\circ$ ($\theta_2 = 2.1^\circ$) between two adjacent graphene layers can be extracted according $n_1 = 4/A_1$ ($n_2 = 4/A_2$), where $A_j = (\sqrt{3}/2)\lambda_j^2$ is the moiré supercell area, $\lambda_j = a/[2\sin(\theta_j/2)]$ is the moiré supercell lattice constant, and $a = 0.246 \text{ nm}$ is the graphene lattice constant, $j = 1, 2$ is the moiré interface index. The verified values of the twist angles being sufficiently away from the magic angle (where a strong hybridization between the layers is expected) gives additional evidence of weak tunnel coupling between the individual graphene monolayers in tTLG.

4. Electrostatic simulation of layer Landau level fillings.

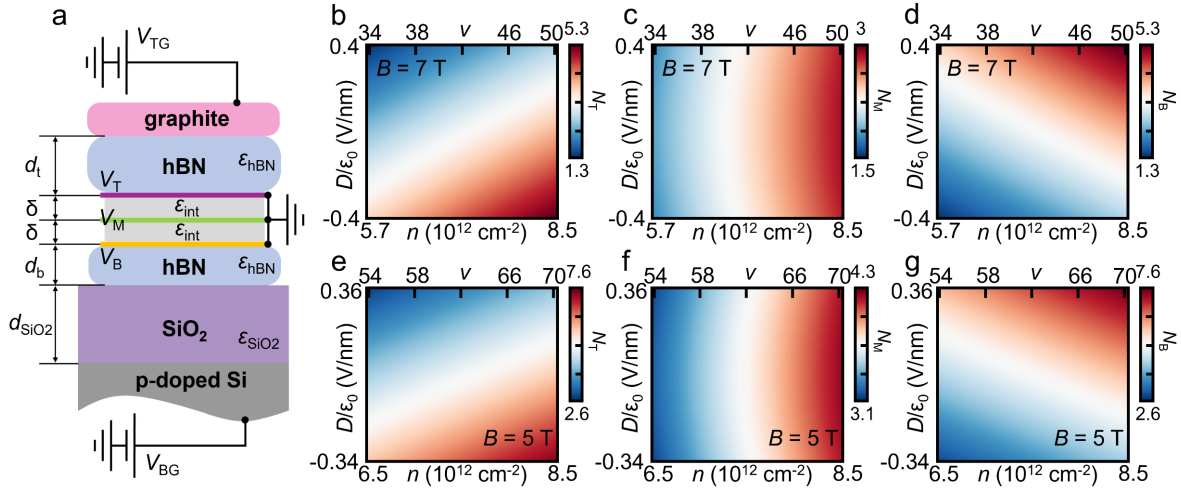


Fig. S3. Electrostatic simulation of Landau level filling in individual layers of graphene. (a) Geometry of the five-plate capacitor model with the graphite top (silicon back) gate having a voltage of V_{TG} (V_{BG}) while the three graphene layers are grounded. (b)-(d) Simulated charge carrier density in the top (b), middle (c), bottom (d) graphene converted into the corresponding layer-specific Chern numbers N_i as a function of total charge carrier density n and displacement field D/ϵ_0 at $B = 7 \text{ T}$, where $i = T, M, B$ are the indices corresponding to the top, middle and bottom layer. (e), (f), (g) same as (b), (c), (d) but the layer Chern numbers are calculated at $B = 5 \text{ T}$.

To confirm the fillings of layer-specific fourfold degenerate Landau levels discussed in the main manuscript (Figs. 2c, 3c, 3g), we perform a simulation of layer charge carrier densities by following an established modelling² of dual-gated twisted trilayer graphene as a five-plate

capacitor (Fig. S3a). Within the model, adjacent layers of the tTLG have a vertical interlayer distance of $\delta = 0.33$ nm, the value estimated in the previous work² on twisted graphene. In the simulation, we neglect the second-order effect of the different top and bottom moiré twist angles on the interlayer distances in tTLG and thus assume the graphene layers being equally separated. The top graphite gate (with an electric potential V_{TG}) is separated from the three grounded graphene layers by an hBN layer with a thickness of $d_t = 57$ nm. The silicon back gate (at an electric potential V_{BG}) is isolated from the tTLG by another hBN flake with a thickness of $d_b = 13$ nm, and a layer of SiO₂ having a thickness of $d_{SiO_2} = 285$ nm. Doping graphene electrostatically costs charging energy, and, therefore, each layer has a non-zero electric potential V_i related to the layer charge carrier density n_i according $n_i = \text{sign}(V_i) \left(\frac{eV_i}{\hbar v_F} \right)^2 / \pi$, where e is the elementary charge, $\hbar$ is the reduced Planck's constant, $v_F = 1 \times 10^6$ m/s is the Fermi velocity in graphene, the index $i = T, M, B$ refers to the top, middle, bottom layer.

Layer-specific charge carrier density can be calculated from the difference of displacement fields across each graphene layer (Gauss' law) given by the following equations

$$\begin{aligned} en_T &= C_{TG}(V_{TG} - V_T) + C_0(V_M - V_T), \\ en_M &= C_0(V_T + V_B - 2V_M), \\ en_B &= C_{BG}(V_{BG} - V_B) + C_0(V_M - V_B), \end{aligned} \quad (S3)$$

where $C_{TG} = \epsilon_{hBN}/d_t$ is the geometric top gate capacitance per unit area; C_{BG} is the geometric back gate capacitance found from the capacitance of the bottom hBN (C_b) and SiO₂ (C_{SiO_2}) according $1/C_{BG} = 1/C_{SiO_2} + 1/C_b$ (see Supporting Information-1 for additional details); $C_0 = \epsilon_{int}/\delta$ is the interlayer capacitance per unit area with an interlayer dielectric constant $\epsilon_{int} = 2.5\epsilon_0$ as reported in the previous studies.^{2,3} The equations are consistent with the introduced formula (S1) in Supporting Information-1 for the total charge carrier density $n = n_T + n_M + n_B = (C_{TG}V_{TG} + C_{BG}V_{BG})/e$, where the offset is $n_0 = 0$ in the simulation.

We solve the equations (S3) self-consistently for a set of top and bottom gate voltages corresponding to the same range of the total charge carrier density n and displacement field D as in Figs. 2b, 3b to model the layer charge carrier density. To verify the assignment of LL fillings at finite magnetic field B for each layer-polarized quantum Hall (QH) state in Figs. 2c, 3c, and 3g, the layer charge carrier densities are converted into layer-specific Chern numbers N_i . The latter are defined as $4(N_i + 1/2) = n_i \hbar / (eB)$ characterizing the number of filled (in this section, a non-integer Chern number indicates partial LL filling) fourfold degenerate LLs in each graphene layer at $B = 7$ T (Figs. S3b–S3d corresponding to Fig. 2 of the main manuscript) and $B = 5$ T (Figs. S3e–S3g corresponding to Fig. 3) where i is the layer index. In layer-polarized QH states at the absence of screening^{4,5} of electric field by partially filled LLs, the layer Chern numbers found from the simulation approximately match those in Fig. 2c and Figs. 3c, 3g. Nonetheless, the simulation does not cover the effects on the charge distribution in the system from screening and coupling between partially filled Landau levels, thus the modelled data does not represent the complete structure of transitions between layer-polarized QH states.

5. Characterization of electrostatic screening by constituent graphene layers.

To characterize the degree of electric field screening^{4,5} by each graphene layer, we examine the behavior of Shubnikov-de Haas (SdH) oscillations in tTLG. Figure S4 presents a zoomed-in scan of Fig. 1d from the main manuscript at high electron doping where longitudinal resistance R_{xx} at $B = 7$ T is plotted as a function of electric field due to the back ($E_{BG} = V_{BG}/d_{bg}$) and top ($E_{TG} =$

V_{TG}/d_t) gate. $V_{BG}(V_{TG})$ is the voltage applied to the back(top) gate, $d_{bg}(d_t)$ is the separation between the tTLG and the respective gate. Figure S4 shows three distinct sets (each having a characteristic slope as a function of E_{BG} and E_{TG}) of resistance peaks (SdH oscillations maxima). The first set (traced by the green dashed lines in Fig. S4) is SdH maxima changing with a slope $|\Delta E_{TG}/\Delta E_{BG}| = 1$ indicates an equal coupling to the top and back gate consistent with quantum Hall (QH) states residing in the middle graphene layer equally screened by the remaining two graphene layers. Peaks from the other two sets are indicated by purple (yellow) markers having slopes $|\Delta E_{TG}/\Delta E_{BG}| < (>)1$ corresponding to QH states in the top (bottom) graphene layer with reduced capacitive coupling to the back (top) gate due to electrostatic screening by the middle and bottom (top) graphene layer. The purple markers are assigned to data points with $R_{xx} > 2 \text{ k}\Omega$ forming two clusters that are individually fitted with a linear function (purple dashed lines in Fig. S4) of E_{BG} and E_{TG} . The average slope of the fitted lines is found to be $\Delta E_{TG}/\Delta E_{BG} = -0.33 \pm 0.03$. Similarly, the linear fit (yellow dashed line in Fig. S4) of the yellow markers ($R_{xx} > 1.2 \text{ k}\Omega$) has a slope of $\Delta E_{TG}/\Delta E_{BG} = -2.35 \pm 0.15$. From this, we can quantitatively estimate the effect of electric field screening by the middle and bottom (top) graphene layer reducing the electric field at the top (bottom) graphene to $|\Delta E_{TG}/\Delta E_{BG}| = 33\% \pm 3\%$ ($|\Delta E_{BG}/\Delta E_{TG}| = 43\% \pm 3\%$). The disparity in screening at the top (bottom) graphene can potentially be explained by structural and electronic microscopic differences of the bottom (top) moiré interfaces having distinct stacking order landscapes due to unequal twist angles between the layers.

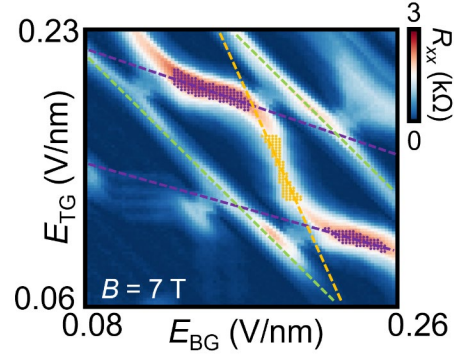


Fig. S4. Characterization of electrostatic screening by graphene layers. Longitudinal resistance R_{xx} as a function of electric field from the top (E_{TG}) and back (E_{BG}) gate at $B = 7 \text{ T}$. The resistance peaks are Shubnikov-de Haas (SdH) oscillations maxima corresponding to quantum Hall (QH) states in different graphene layers. The peaks with a slope $|\Delta E_{TG}/\Delta E_{BG}| = 1$ (traced by the green dashed lines) correspond to QH states in the middle graphene. Peaks with slopes $|\Delta E_{TG}/\Delta E_{BG}| < (>)1$ are from QH states in the top (bottom) graphene layer with selected values of $R_{xx} > 2 \text{ k}\Omega$ ($R_{xx} > 1.2 \text{ k}\Omega$) highlighted by purple (yellow) markers whose dependence on E_{BG} and E_{TG} is fitted with purple (yellow) lines.

The position of the SdH peaks in Fig. S4 from QH states of either the top or bottom layer is determined by a constant charge carrier density (constant Landau level filling) in the corresponding layer given by

$$C_{\text{BG}}^* V_{\text{BG}} + C_{\text{TG}}^* V_{\text{TG}} = \text{const}, \quad (\text{S4})$$

where $C_{\text{BG}}^*(C_{\text{TG}}^*)$ is the effective capacitance of the back (top) gate per unit area accounting for the screening effect. For the QH states in the bottom (top) layer $C_{\text{BG}}^* = C_{\text{BG}}$ ($C_{\text{TG}}^* = C_{\text{TG}}$), where C_{BG} and C_{TG} are the geometric gate capacitances defined in Supporting Information-1. Equation (S4) for the peak positions can be rewritten in terms of electric field from the back (top) gate $E_{\text{BG}}(E_{\text{TG}})$ and distances to the respective gates:

$$C_{\text{BG}}^* d_{\text{bg}} E_{\text{BG}} + C_{\text{TG}}^* d_{\text{t}} E_{\text{TG}} = \text{const}. \quad (\text{S5})$$

Using this equation, the ratio between the effective capacitances of the top and back gate can be estimated from the slope of the SdH maximum lines: $C_{\text{BG}}^*/C_{\text{TG}}^* = |(d_{\text{t}} \Delta E_{\text{TG}})/(d_{\text{bg}} \Delta E_{\text{BG}})| = (d_{\text{t}}/d_{\text{bg}}) |\Delta E_{\text{TG}}/\Delta E_{\text{BG}}|$. By substituting the extracted slope for the QH states in the top (bottom) graphene layer, the effective capacitance ratio $C_{\text{BG}}^*/C_{\text{TG}}^*$ is found to be 0.063 ± 0.005 (0.45 ± 0.03).

6. Effect of disorder on Landau level broadening.

The sample having disorders such as charged impurities or lattice distortions due to strain results in Landau level (LL) broadening and subsequent widening of the SdH oscillations peaks which can potentially obscure the studied QH transitions. As a consequence, the observed transport signatures of filling the N_i -th LL in an i -th graphene layer become smeared out when the energy difference (Landau gap) between the nearest LLs, $\Delta_i = \sqrt{2} \hbar v_F (\sqrt{N_i + 1} - \sqrt{N_i})/l_B$, is comparable to the LL disorder broadening, Γ , where $l_B = \sqrt{\hbar/eB}$ is the magnetic length. A typical Landau gap in the QH stability diagram (Figs. 2, 3) with $N_i = 5$ has a characteristic size of ~ 20 meV at $B = 5$ T. The LL energy broadening, Γ , is determined⁶ by the quantum scattering time, τ_q , according to the uncertainty relation $\Gamma \sim \hbar/\tau_q$. Estimating the quantum scattering time as the charge carriers' collision time, τ , extracted from their mean free path^{7,8} $l \sim v_F \tau \sim 6 \mu\text{m}$ at $7 \times 10^{12} \text{ cm}^{-2}$ gives a LL energy broadening of $\Gamma \sim 0.1$ meV, the value which is more than two orders of magnitude smaller than the typical size of the Landau gap in tTLG. However, as reported in a previous study⁶, τ_q in graphene is up to one order of magnitude smaller than the collision time, τ , giving the realistic LL broadening of ~ 1 meV⁹ which is still considerably less than the Landau gaps in the studied system allowing us to resolve the Chern transitions between most of the layer-polarized QH states. The LL energy broadening becomes relevant when the LLs from different graphene layers are closely aligned. As an example, in Fig. 3h, the LLs are misaligned by $\Delta_M/8 \sim 3$ meV comparable to $\Gamma \sim 1$ meV resulting in an R_{xx} peak spanning over the entire 5|4|5 domain.

7. Electric field screening by partially filled Landau levels.

A direct consequence of disorder is the presence of localized and extended states in Landau levels. In contrast to localized QH states, for a given graphene layer, charge carriers in a partially filled Landau level at an extended state can significantly screen¹⁰ external electric field while reducing its effect on the other layers. This phenomenon is particularly prominent near the $5|3|5 \Leftrightarrow 5|4|5$ intra-middle-layer transition at $B = 5$ T where at zero displacement field (Fig. S5a, blue hollow triangle), the Landau levels of all three layers are closely aligned (Fig. S5b) while the Fermi level E_F (Fig. S5b) is at the maximum of the middle layer 4th Landau level's density of states (Fig. S5b, right). At this LL alignment, even a slight displacement field can move the LLs of the top/bottom layer to the Fermi energy E_F making the corresponding layer compressible (in an extended QH state) thus significantly reducing the effect of electric field on the middle graphene. Specifically, while keeping the charge carrier density constant, applying a small positive displacement field (by adding negative/positive voltage to the top/back gate) leads to partial filling of the bottom layer LL (left of Fig. S5c represented by the hollow star in Fig. S5a) corresponding to the bottom graphene being in a compressible QH state. The latter screens the additional positive voltage from the back gate thus lowering the LL filling of the middle layer. As a result, the middle graphene transitions from the extended QH state at the half-filled LL (right of Fig. S5b) to a localized one (right of Fig. S5c) passing the bottom mobility edge. Further applying positive D field at constant n , aligns the top layer LL with E_F (Fig. S5d corresponding to the hollow diamond in Fig. S5a) making the top graphene compressible and screening the added negative voltage to the top gate. Thus, the absence of the additional negative top gate voltage completely fills the middle layer LL pushing the Fermi level through the LL top mobility edge (left of Fig. S5d). Similarly, a decreasing D (at added positive/negative voltage to the top/back gate) makes the top layer LL half filled (Fig. S5e, hollow pentagon in Fig. S5a), screening the increased voltage from the top gate. Consequently, the reduced positive top gate voltage moves the E_F across the same bottom mobility edge as in Fig. S5c.

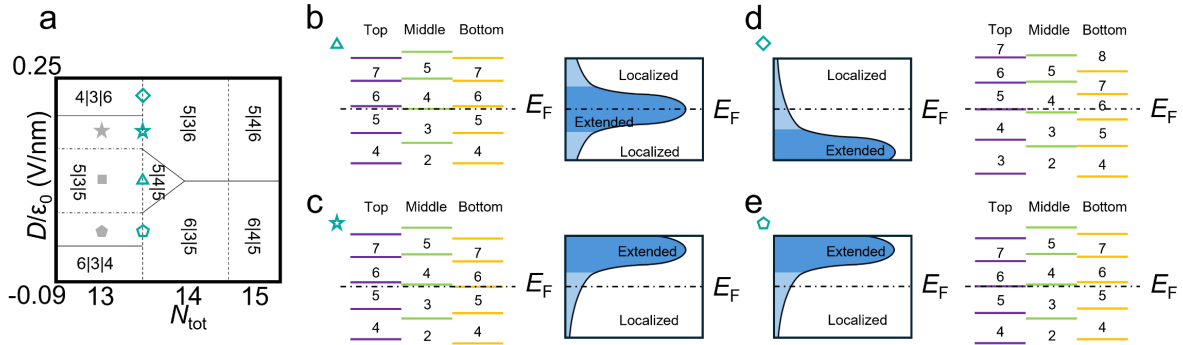


Fig. S5. Effect of screening by partially filled Landau levels. (a) Stability diagram of layer-polarized QH states centered near the $5|4|5$ QH state at $B = 5$ T. (b)-(e) Layer-specific Landau level alignment with respect to the Fermi energy E_F at selected QH state transitions labeled with the hollow blue markers in (a). The cartoons on the right in (b), (c) and on the left in (d), (e) schematically show the density of states of the $N_M = 4$ middle layer Landau level with respect to the Fermi energy E_F . The dash-dotted lines are the mobility edges separating the localized and extended QH regimes.

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