

METHODS

Device fabrication Devices were assembled using a dry transfer method based on van der Waals adhesion[1], with top and bottom gates as well as the electrical contacts to the hBN-encapsulated Bernal stacked BLG device made from ~ 10 nm-thick graphite flakes. As shown in Figs. S1-S4, this leads to a significant improvement in sample quality as compared to samples with evaporated metal gates. Care was taken in order to match the top and bottom hBN thicknesses, which were approximately 40-50 nm for all three devices. The resulting mismatch in geometric capacitance was determined to be $\delta \equiv (c_t - c_b)/(c_t + c_b) = 0.018$ for Device A, and was $< .05$ for all devices. Despite efforts to rotationally misalign the hBN crystals proximate to the graphene bilayer, the effects of an intermediate wavelength moire pattern[2–4] are visible on one of the layers at high density in device A (Fig. S12). The resulting staggered sublattice potential is responsible for the asymmetry upon inversion of p_0 in some of the transitions visible in Fig. 1 of the main text. While no secondary fans were observed in devices B and C, similar offsets (albeit of differing magnitudes and signs) were observed in these devices as well, indicating that coupling to one or more of the hBN layers remains relevant (See Figs. S13, S14, and S15). Devices A and C were patterned by using a dry ICP etch in a mixture of $\text{CHF}_3 + \text{O}_2$, and the graphite contacts were themselves contacted along the edge with a Cr/Pd/Au metal stack. Device B was first exposed to a XeF_2 atmosphere to remove a sacrificial top hBN layer used for stack assembly, annealed at 400°C in forming gas, and the graphite contacts and gates were area contacted with a Ti/Au metal stack.

Capacitance circuit To measure the small capacitances of our device with high sensitivity, we utilize a cryogenic impedance transformer based on an FHX35X high electron mobility transistor[5] in a bridge configuration. This measurement works by effectively disconnecting the sample capacitance from the large capacitance of the cryostat cabling: the BLG device is connected only to the gate of the HEMT (with input capacitance of a few hundred fF) and a large resistor ($100\text{ M}\Omega$), with additional $\sim 1\text{pF}$ of stray capacitance. While the HEMT is operated at or below unity voltage gain to minimize power dissipation, it effectively transforms the picofarad sample impedance into a $1\text{k}\Omega$ output impedance. At 50 kHz , and assuming cryostat cable capacitances of $\approx 1\text{nF}$,

this translates to a power gain of approximated 1000.

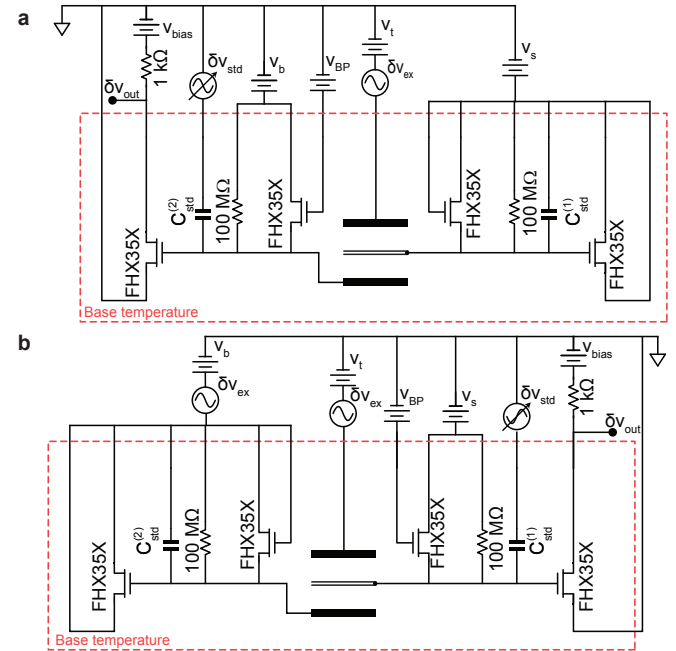


FIG. S1. Simplified measurement schematics. (a) Measurement of penetration field capacitance, C_P . **(b)** Measurement of symmetric capacitance, C_S . Here DC voltages are denoted v_i while AC voltages are denoted δv_i . $v_{BP} \approx -0.5\text{V}$ pinches off the multiplexing transistor (on bottom gate or sample, respectively for the two measurement schemes), tuning the input impedance of the measurement transistor on that terminal. δv_{ex} is the AC excitation applied across the relevant capacitor to be measured, and δv_{std} is chosen to balance the bridge such that $\delta v_{\text{out}} = 0$. v_t , v_s , and v_b in combination control p_0 and n_0 , with one of the three fixed to control the working point of the measurement transistor. n_0 and p_0 are swept and δv_{out} is monitored to extract the capacitance. In all measurements, the measurement transistor bias $v_{\text{bias}} \approx 20\text{ mV}$ was limited to prevent excessive heating of the sample.

As described in detail in references 6 and 7, measurements of three distinct capacitances—the top gate, bottom gate, and penetration field capacitance—provide a complete reconstruction of both the charge and layer polarization in the bilayer as a function of applied voltages. For devices in which top and bottom gate geometric capacitances are symmetric ($\delta \equiv \frac{c_t - c_b}{c_t + c_b} \rightarrow 0$, see supplementary information of 7

for a detailed derivation), one accesses the derivatives of the total density (n) and layer polarization (p) with respect to the applied voltages n_0 and p_0 through the relations

$$\frac{\partial n}{\partial n_0} \approx \frac{C_S}{2c} \quad (\text{S1})$$

$$\frac{\partial n}{\partial p_0} \approx -\frac{C_A}{2c} \quad (\text{S2})$$

$$\frac{\partial p}{\partial p_0} \approx \frac{c_0}{c} \frac{4C_P + C_S - 2c}{c} \quad (\text{S3})$$

$$\frac{\partial p}{\partial n_0} \approx -\frac{c_0}{c} \frac{C_A}{c}, \quad (\text{S4})$$

where $c = (c_t + c_b)/2$ is the average of the top and bottom gate geometric capacitances, $c_0 \gg c$ is the interlayer capacitance of the bilayer itself, and $C_{S(A)} = C_T \pm C_B$ are the symmetric and antisymmetric measured gate capacitances per unit area. The approximation is well justified as $\delta = .018$ for Device A and is similarly small for the other devices.

In order to measure all three quantities without warming up the sample, low-impedance access to all three terminals of the device must be possible in situ, in apparent conflict with the desire to maintain the amplifier input at high impedance. This problem is solved using a cryogenic multiplexer constructed out of two additional HEMTs, which allows either the bottom gate or the bilayer graphene flake to be brought to a high impedance. Figure S1a shows a simplified electrical schematic for measuring $C_P \equiv \partial n_T / \partial v_b$, where n_T is the charge on the top gate while v_b is the bottom gate voltage. $v_b \approx -3\text{V}$ is fixed to set the transistor operating point, $v_{bias} \approx 25\text{mV}$ is chosen so that no heating of the probe is observed (see the next subsection on likely electron temperature). Additional DC voltages on the top gate (v_t) and applied to the ohmic contacts of the graphene (v_s) are varied to control n_0 and p_0 , defined as

$$n_0 = c_t(v_t - v_s) + c_b(v_b - v_s) \approx c(v_t + v_b - 2v_s) \quad (\text{S5})$$

$$p_0 = c_t(v_t - v_s) - c_b(v_b - v_s) \approx c(v_t - v_b). \quad (\text{S6})$$

To measure the differential capacitance, a fixed AC excitation is applied to top gate (δv_{ex}). A second AC excitation at the same frequency is applied to a standard capacitor δv_{std} , with phase and amplitude chosen to balance the bridge ($\delta v_{out} = 0$). Crucially, for the C_P measurement the ‘bypass’ HEMT attached to the bottom gate is driven deep into depletion ($v_{BP} \approx v_b - .6\text{V}$), maintaining the high impedance of the measurement transistor input. A final DC voltage is applied to all pins of the second amplifier (v_s). Because the FHX35X is depletion mode, in this configuration it shorts out the 100 M Ω resistor, providing a low impedance connection to the sample. To measure C_S , the situation is reversed (Figure S1b), with the transistor amplifier on the sample now active and the bypass transistor on the bottom gate short circuited, providing a low impedance path for the bottom gate voltage excitation. Although not focus of the current work, C_A can also be measured by applying opposite phase excitation signals to the top

and bottom gate[7]. The frequency and excitation voltages for data shown in all figures are shown in Table S3.

All measurements below 14T were performed in a dry dilution refrigerator with a base temperature of $\approx 10\text{ mK}$. However, the sample temperature was likely higher due to heating from the cryogenic amplifiers, which were directly connected to the sample and only a few millimeters away. While we do not have a thermometer for the electron temperature in our devices, recent tunneling experiments, which use an identical amplification scheme in a dilution fridge with a similar base temperature, do allow for in situ thermometry[8]. In these experiments, electron temperatures below 100 mK are possible only with careful thermal isolation and heat sinking of the HEMT, which is a heat source directly tied to the sample[9]. Because we have not taken these precautions in our setup, our electron temperature is likely no less than 100mK for the capacitance measurements. Indeed, data taken at the NHMFL in Tallahassee in a 3He system with a base temperature of 300mK do not look qualitatively different.

Measurement of electronic compressibility and thermodynamic energy gaps Most of the experimental data presented in the main text are C_P , which we interpret as proportional to electronic compressibility. As is clear from Eqs. S1-S3, this is not generally true: solving those equations for C_P gives

$$C_P = \frac{c}{2} \left(1 - \frac{\partial n}{\partial n_0}\right) + \frac{c^2}{4c_0} \frac{\partial p}{\partial p_0}. \quad (\text{S7})$$

The third term in this equation is unique to multilayers, denoting the layer polarizability. Near layer polarization phase transitions, where the layer occupation changes rapidly over a small range of p_0 , this term can be large; however, in the single component regimes where, for example, we perform our measurements of the thermodynamic gaps, this term is tiny. In these regimes, charge cannot move easily between the layers, and the bilayer system behaves as a single layer system (measurements of $\partial p / \partial p_0$, discussed below, are featureless in this regime). In this case, the conventional quantum capacitance[10] formula applies, so that

$$\frac{\partial n}{\partial n_0} = \frac{1}{1 + 2c \frac{\partial \mu}{\partial n}} \quad (\text{S8})$$

$$C_P = \frac{c}{2} - \frac{1}{4} \left(\frac{1}{2c} + \frac{1}{\partial n / \partial \mu} \right)^{-1} \quad (\text{S9})$$

$$= \frac{c^2}{2c + \partial n / \partial \mu} \quad (\text{S10})$$

From the above formulas, the change in chemical potential follows as

$$\Delta \mu_{12} = \int_{n^{(1)}}^{n^{(2)}} \frac{\partial \mu}{\partial n} dn = \int_{n_0^{(1)}}^{n_0^{(2)}} \frac{\partial \mu}{\partial n} \frac{\partial n}{\partial n_0} dn_0 \quad (\text{S11})$$

$$= \int_{n_0^{(1)}}^{n_0^{(2)}} \frac{C_P}{c} d(n_0/c) \quad (\text{S12})$$

We take the gap as the difference between the maximum and minimum in $\Delta \mu$ near a FQH state, as shown in Fig. 2b of the

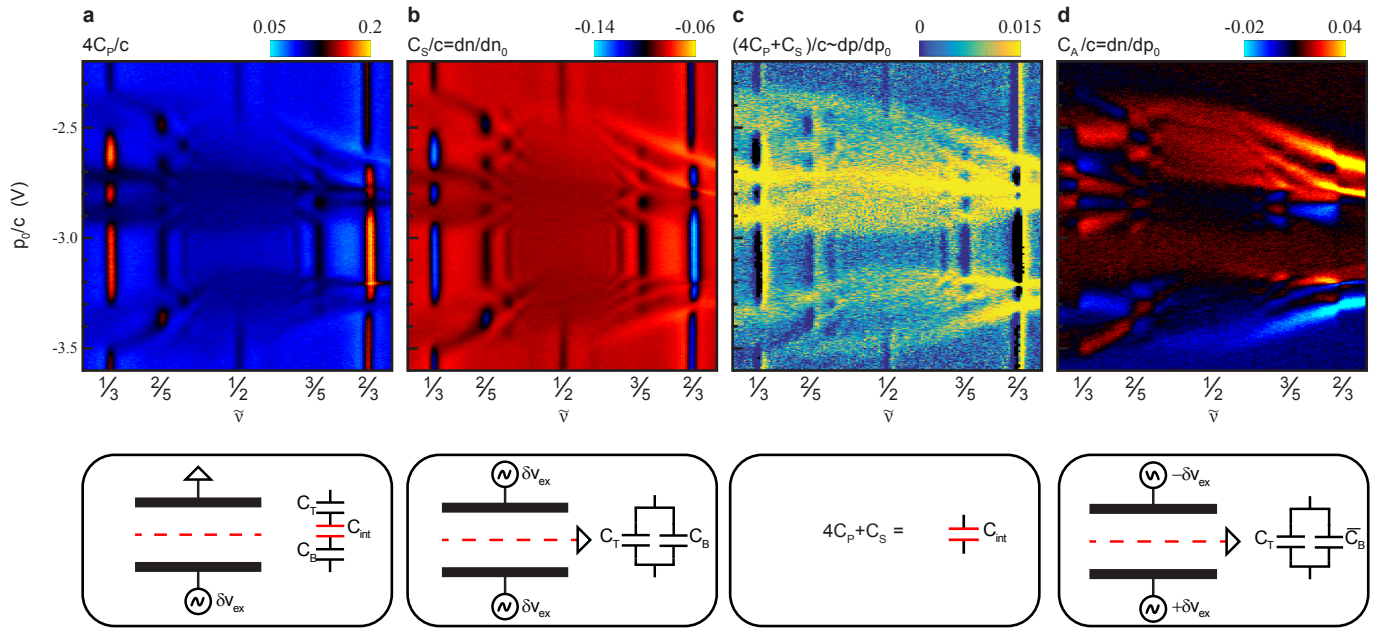


FIG. S2. Thermodynamic properties of interlayer correlated states. (a) Penetration field capacitance $4 \times C_P/c$. (b) Symmetric capacitance C_S/c . (c) $4C_P + C_S$, proportional to the polarizability. (d) anti-symmetric capacitance C_A . As presented, C_P/c and C_S/c were offset by constant parasitic capacitance levels $C_{\text{par}}/c = -0.55$ and $C_{\text{par}}/c = -2.84$ respectively. The lower panels illustrate the effective geometric capacitance measured in each case, as would be applicable for a classical four plate capacitor. C_P measures top, bottom, and interlayer capacitance in series, while C_S measures the sum of the top and bottom capacitance. $4C_P + C_S$ thus gives the interlayer capacitance, C_{int} , which is $dp/du \sim dp/dp_0$. C_A , discussed extensively elsewhere[7], measures the difference between top and bottom capacitances.

main text. As can be seen from Eq. S12, quantitative measurement of $\Delta\mu$ requires accurate knowledge of c . This is calibrated in situ from the capacitance in an integer quantum Hall gap (where the full $c/2$ penetration capacitance obtains). Unfortunately, this data was not be acquired in the limited time available at the National high magnetic field lab. For this reason, the gaps in Fig. 1e of the main text (right panel) are presented only in relative units, normalized to their value at $B=14\text{T}$.

Interpretation of any measured capacitance as a thermodynamic derivative requires that the sample is sufficiently conductive to fully charge over a time scale comparable to the inverse measurement frequency[11]. At low temperature and high magnetic fields, our sample becomes strongly insulating at all integer and many fractional filling factors, precluding this condition being satisfied at high frequency. This is unimportant for identifying the existence of fractional quantum Hall phases, which manifest as C_P peaks regardless of whether the contrast mechanism is due to low electronic compressibility or low bulk conductivity; however, it is critical to be in the low frequency limit for any quantitative analysis of energy gaps following Eq. S12. Failure to charge manifests as an increase in the out of phase, dissipative signal in the complex capacitance, $\tilde{C} = C + iD$, where we have plotted ‘C’ throughout the text. We can monitor charging across the parameter range by plotting ‘D’. In order to measure energy

gaps, we decrease the frequency until no features are visible in D. In this limit, it is justified to integrate C_P to extract energy gaps.

Measurement of layer polarizability, $\partial p/\partial p_0$ To measure $\partial p/\partial p_0$, as shown in Fig. 3e of the main text, we measure penetration field (C_P) and symmetric capacitance (C_S) over a range of n_0 and p_0 corresponding to the four-level crossing described in the main text Figs. 3a-b. As can be seen in Fig. S11d, there are diagonal features in C_S within the $n_0 - p_0$ plane that correspond to constant gate voltage. We ascribe these features either to incompressible LL gaps in the graphite gates themselves. To avoid them, we perform the measurement in the nearly identical level crossing at $p_0/c \approx -3V$ for $-1 < \nu < 0$, where diagonal features are not observed in the regime of the phase transition. C_P and C_S in this regime are shown in Fig S2, along with their weighted sum, $4C_P + C_S$ as per Eq. S3. The two measurements were calibrated against each other by measuring C_T , the top gate capacitance, accessible in either configuration.

FQH gaps show strong features in the $\partial p/\partial p_0$ data. This is spurious: digital subtraction of the two data sets measured with different amplifiers leads to systematic errors where C_P and C_S have large gradients, for example in FQH gaps. There, small offsets in n_0 and nonlinearities in the amplifier chain can lead to incomplete cancellation of the two signals, leading to the visibility of the gaps themselves in Fig. 3e of the

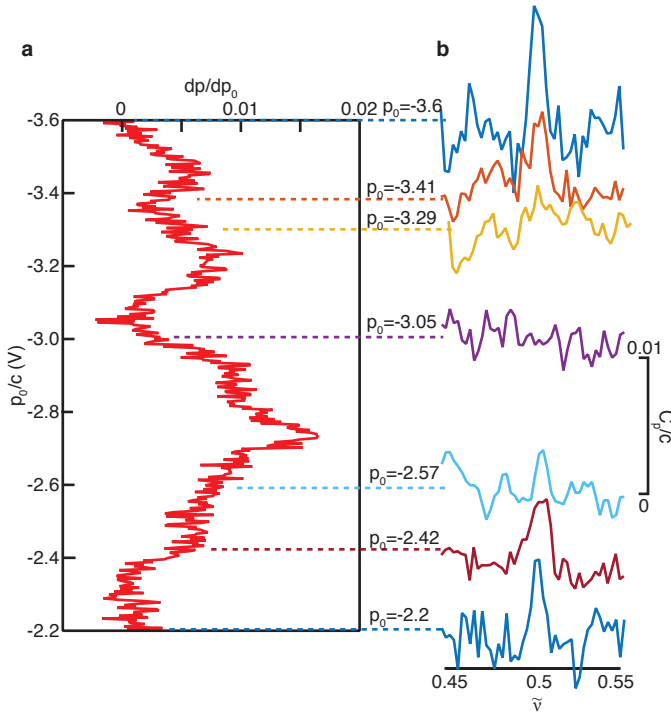


FIG. S3. **Qualitative comparison of incompressibility and depolarization at $\nu=1/2$** **a** $C_P + 4C_S \sim dp/dp_0$ near $\tilde{\nu} = 1/2$ as a function of p_0 and **(b)** C_P as a function of $\tilde{\nu}$ showing the incompressible peak at $\tilde{\nu} = 1/2$. The gap persists despite finite depolarization, fading with increasing p but vanishing only between $p_0/c \approx -3.3$ and $p_0/c \approx -2.6$.

main text. This is confirmed by the measurement of $\frac{dp}{dn_0}$: if the changes in polarization were real, they would manifest in this quantity as well. $\frac{\partial p}{\partial n_0}$ is shown in Fig. S2d. Because $\frac{\partial p}{\partial n_0}$ can be measured directly with a single amplifier (see, for example, [7]), subtraction-induced systematic errors are automatically canceled. We find no change in polarization of the $\tilde{\nu} = \frac{1}{2}$ state relative to its immediate background, indicating that the features in $\partial p/\partial p_0$ at fractional filling are indeed spurious. We thus integrate $\frac{\partial p}{\partial p_0}$ at $\tilde{\nu} = 0.495$ and $\tilde{\nu} = 0.505$ and average the results to determine Δp . Figure S2c shows raw $\frac{\partial p}{\partial p_0}$ data,

To determine the layer polarization change for arbitrary ν and p_0 , we integrate the measured $C_S + 4C_P$ signal, as

$$\Delta p_{12} = 2\pi\ell_B^2 \int_{p_0^{(1)}}^{p_0^{(2)}} \frac{\partial p}{\partial p_0} dp_0 \quad (\text{S13})$$

$$= 2\pi\ell_B^2 \frac{2c_0}{c} \int_{p_0^{(1)}}^{p_0^{(2)}} \left(\frac{C_S + 4C_P - 2c}{c} \right) dp_0 \quad (\text{S14})$$

$$= 2\pi\ell_B^2 2c_0 \int_{p_0^{(1)}}^{p_0^{(2)}} \left(\frac{C_S + 4C_P - 2c}{c} \right) d(p_0/c) \quad (\text{S15})$$

where Δp is expressed in filling factor units and all capac-

itances are understood to be in units of particle number per area per volt.

The measured data may be subject to an arbitrary offset due to differing parasitic capacitances in the C_S and C_P measurements which need not cancel. In addition, an overall magnitude error can lead to a systematic under- or over estimate of the change in polarization. To compensate these errors, we take advantage of the fact that we know, from band structure, the total layer polarization change that must occur between the two extremes in Fig. 3e (and Fig. S2a-d). At the bottom of these plots, an $N=0$ orbital on the bottom layer is fully occupied, while an $N=1$ orbital on the bottom layer has occupation $\tilde{\nu}$. At the top, these orbitals are empty while an $N=0$ orbital on the top layer is fully occupied while the $N=1$ orbital on the top layer has occupation $\tilde{\nu}$. Thus, for a given $\tilde{\nu}$ in this regime, we expect a total occupation transfer of $\Delta\nu = 2(1 + \alpha\nu)$, where $\alpha = .84$ is the layer polarization of the $N=1$ orbitals at 12T determined from tight binding calculations. We use this to fit two constants to the integrated data, a and b , so that

$$a \int_{-3.6}^{-2.2} \left(\frac{C_S + 4C_P}{c} - b \right) d\left(\frac{p_0}{c}\right) = 2(1 + \alpha\nu) \quad (\text{S16})$$

where the contour of integration is at constant $\tilde{\nu}$ from the bottom to the top of Fig. S2c. The constant b can be determined directly by measuring the background level in a single component region, where we expect $\partial p/\partial p_0 = 0$, and this background is subtracted from the $4C_P + C_S$ data set shown in Fig. S2c. The error with which this background can be determined is $\sigma_b = 2.3 \times 10^{-4}$. We then calculate a from Eq. S16 by performing this integration for 35 combinations of 7 different regions of $\tilde{\nu}$, from which we find $a = (422 \pm 22) \text{ V}^{-1}$. Following Eq. S15 allows us to extract the interlayer dielectric constant of the graphene bilayer, ϵ_{BLG} since,

$$a = 4\pi c_0 \ell_B^2 \quad (\text{S17})$$

$$= 4\pi \frac{\epsilon_{BLG}}{d_{BLG}} \frac{\epsilon_0}{e} \ell_B^2 \quad (\text{S18})$$

$$\therefore \epsilon_{BLG} = \frac{a}{113.6 \text{ V}^{-1}} = 3.71 \pm .19 \quad (\text{S19})$$

for $d_{BLG} = .335 \text{ nm}$, $B = 12 \text{ T}$. This is roughly consistent (to within 25%) with $\epsilon_{BLG} = 2.8$ determined in recent experiments at higher magnetic fields[7]. We note that ϵ_{BLG} receives contributions from filled Landau levels, and may well be field dependent. To generate Fig. 3f of the main text, we integrate along p_0/c , for example

$$\Delta p(p_0) = a \int_{-3.6}^{p_0/c} \left(\frac{C_S + 4C_P}{c} - b \right) d\left(\frac{p_0}{c}\right) \quad (\text{S20})$$

$$\sigma_{\Delta p}(p_0) = \frac{\sigma_a}{a} \Delta p(p_0) + \sigma_b(p_0/c + 3.6) \quad (\text{S21})$$

Because the layer polarization is known also for $p_0/c = -2.2 \text{ V}$, either the increasing or decreasing integral are equivalent, and error bars are defined by the lesser of the two.

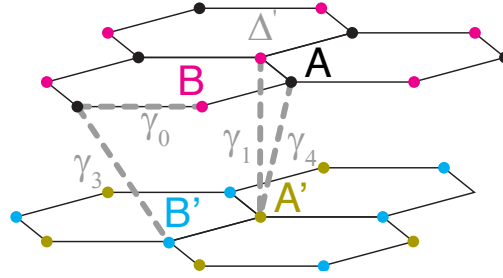


FIG. S4. **Bilayer graphene hopping parameters used in the single particle model for the wavefunctions.** In the calculations, $\gamma_0 = -2.61$ eV, $\gamma_1 = 0.361$ eV, $\gamma_3 = 0$, $\gamma_4 = 0.138$ eV, and $\Delta' = 0.015$ eV.

The largest uncertainty thus obtains approximately midway between the extremes near $p_0/c = -2.9$ V.

This analysis provides a quantitative view of the correlation of depolarization with gap size, the existence of a gapped phase despite depolarization is visible in the raw data. This is evident in Fig. S3, where a finite incompressibility peak is visible after considerable polarization has been transferred.

Single particle model We use the same single-particle model for bilayer graphene as described in the supplementary

information of 7, which includes tight binding parameters γ_0 , γ_1 , γ_4 , and Δ' (see Fig. S4). The single-particle spectrum at $B = 14$ T is shown in Fig. S6, with a level ordering which is typical for all the B -fields used in the experiment. Within this model, the ZLL wavefunctions in the different valleys (written in the lattice basis $\psi_{N\xi} = (\phi_A, \phi_{B'}, \phi_{A'}, \phi_B)$) are

$$\psi_{0+} = \begin{pmatrix} |0\rangle \\ 0 \\ 0 \\ 0 \end{pmatrix} \quad \psi_{0-} = \begin{pmatrix} 0 \\ |0\rangle \\ 0 \\ 0 \end{pmatrix} \quad \psi_{1+} = \begin{pmatrix} \cos \Theta |1\rangle \\ 0 \\ \cos \Phi \sin \Theta |0\rangle \\ \sin \Phi \sin \Theta |0\rangle \end{pmatrix} \quad \psi_{1-} = \begin{pmatrix} 0 \\ \cos \Theta |1\rangle \\ \sin \Phi \sin \Theta |0\rangle \\ \cos \Phi \sin \Theta |0\rangle \end{pmatrix} \quad (\text{S22})$$

$|0\rangle$ and $|1\rangle$ denote the $n = 0, 1$ magnetic oscillator states of a conventional parabolically dispersing system. The layer polarization of the $|1\rangle$ orbitals is then $\alpha = \cos^2 \Theta - \sin^2 \Theta (\cos^2 \Phi - \sin^2 \Phi)$. However, Φ is very small ($< .033$ for fields below 35 T). Thus Φ does not shift the balance between $|0\rangle$ and $|1\rangle$ oscillator states, and so does not enter calculations of long range Coulomb effects such as fractional quantum Hall. Θ controls the degree to which $N=1$ orbitals are strictly analogous to a parabolic electron system, i.e., purely composed of $|1\rangle$ oscillator states. A plot of $\Theta(B)$ is shown in Fig. S5.

Numerical simulations The phase diagram of Fig. 2d of the main text was calculated using the density matrix renormalization group (DMRG) for multicomponent quantum Hall

systems[13, 14]. For a detailed exposition and justification of the approach used here, we refer to Ref.7, which benchmarked similar numerical computations against the experimentally determined layer polarization data.

The splitting Δ_{10} between the $N = 0, 1$ orbitals of the ZLL is small, while the splitting between the ZLL and the higher LLs is large, of order $\hbar\omega_c$. Our approach is based on the hierarchy $\Delta_{10} < E_C < \hbar\omega_c$, where E_C is the Coulomb scale, which holds throughout the experimental range of B and p_0 (see Fig S6). Because of the large splitting between the ZLL and the $N \geq 2$ LLs, we project the problem into the ZLL, though will account for screening from the filled LLs through an effective interaction V_{eff} we will discuss shortly. The ZLL projected Hamiltonian takes the form

$$H = \int d^2q n_{\text{ZLL}}(q) V_{\text{eff}}(q) n_{\text{ZLL}}(-q) + \Delta_{10} \hat{N}_1 + [\text{isospin splittings}] + [\text{SU(4)-interaction anisotropies}] \quad (\text{S23})$$

Here $n_{\text{ZLL}}(q)$ is the Fourier transform of the 2D electron den-

sity projected into the ZLL, V_{eff} is the effective interaction, $\hat{N}_1$

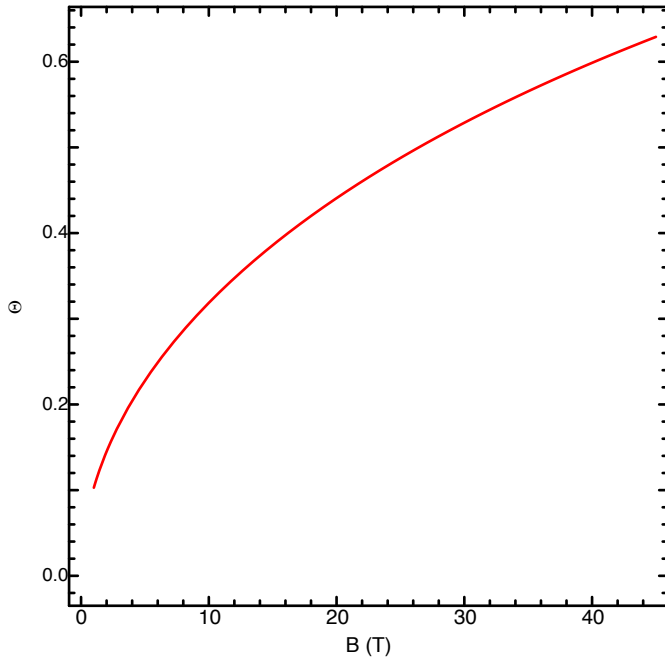


FIG. S5. **Band structure parameter** Θ , as calculated from the tight binding model as a function of magnetic field.

is the electron number in the $N = 1$ orbital, and Δ_{10} is the splitting between the $N = 0, 1$ orbitals. There are also single-particle isospin (spin-valley) splittings and small SU(4) interaction anisotropies, but since our interest is in regions where the valence electrons partially occupy a single isospin, while all other isospins are either full or empty, these can be dropped from the problem.

The bare Coulomb interaction is screened by the encapsulating hBN, the graphite gates at a distance $d/2$ from the sample, and the filled LLs below the ZLL. Screening from the hBN is accounted for in the Coulomb scale $E_C = \frac{e^2}{4\pi\epsilon_{\text{hBN}}\ell_B}$, where $\epsilon_{\text{hBN}} \sim 6.6$ is the dielectric constant of the hBN[12]. We assume the graphite behaves as a metal, so in units of ℓ_B , E_C , the gate screened interaction is $V(q) = \frac{2\pi}{q} \tanh(qd)$. The phase diagram here is presented for device A, $d = 40\text{nm}$, $d/\ell_B = 1.56\sqrt{B/\text{T}}$, but the results are largely insensitive to d at these fields since $d/\ell_B \gg 1$.

The residual response of the filled LLs below the ZLL is controlled by the ratio $\frac{E_C}{\hbar\omega_c} \approx 0.5 - .8$. We account for it within a phenomenological RPA-type treatment discussed in Refs.[7, 15], taking

$$V_{\text{eff}}(q) = \frac{V(q)}{1 + a_{\text{scr}} V(q) \tanh(0.62q^2\ell_B^2) 4\log(4)/2\pi}. \quad (\text{S24})$$

Within the two-band model of BLG at $\nu = 0$, RPA calculations give $a_{\text{scr}} = \frac{E_C}{\hbar\omega_c} \equiv a_*$. However, this value will not be quantitatively correct due to 4-band corrections and the filling of the isospins within the ZLL. For this reason, we treat a_{scr} as a phenomenological parameter. In Ref.7, quantitative

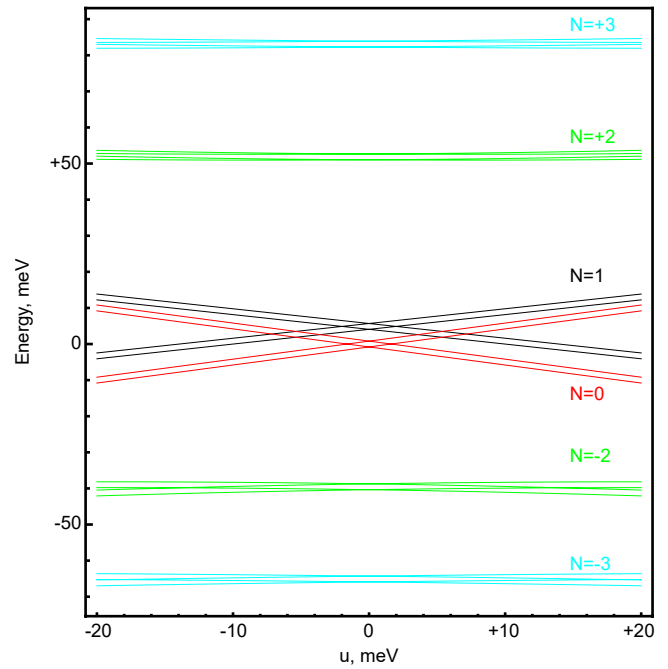


FIG. S6. **Single particle energy spectrum of bilayer graphene at $B = 14\text{T}$** , calculated from a tight binding model and including the Zeeman splitting and interlayer potential. The interlayer potential difference $u \approx \frac{p_0}{c} \frac{c}{2\epsilon_0} \approx 3.3 \times 10^{-3} \frac{p_0}{c}$, does not exceed 15 meV in the data presented. For reference, the bare Coulomb energy at 14T is $e^2/\epsilon_{\text{hBN}}\ell_B \approx 31\text{ meV}$, where we have taken $\epsilon_{\text{hBN}} = 6.6$ [12]. Interactions thus strongly mix the $N=0$ and $N=1$ levels (and in fact change the order of level filling within the ZLL, as shown in Ref. 7), while mixing of the 0-1 manifold with the $|N| \geq 2$ levels is comparatively weak.

agreement between numerics and experiment at 31T was obtained for $a_{\text{scr}} \sim 0.38a_*$, which is the value used in the phase diagram of the main text. To check that our conclusions are qualitatively insensitive to screening, the calculations below will be repeated for $a_{\text{scr}} = 0, 0.25a_*, 0.5a_*$ and $0.75a_*$.

We note that the RPA treatment only renormalizes the two-body Hamiltonian, while in principle higher-body terms are also generated. However, the two-body screening diagrams are larger by a factor of $N_f = 4$ relative to three-body corrections, and three-body corrections actually vanish within the “standard” model of graphene, which only accounts for the nearest neighbor hoppings, due to particle-hole symmetry. When accounting for the further-neighbor hoppings, there is a small amount of particle-hole symmetry breaking, but taken together this suggests the effective three-body interactions from outside the ZLL are much smaller than those which will be generated (and fully accounted for) from LL-mixing within the ZLL itself.

Under the assumption of isospin polarization, the $n_{\text{ZLL}}(q)$ can be restricted to the contribution from a single isospin, which contains the two LLs $N = 0, 1$. In this study we keep the full Hilbert space of both, since the splitting Δ_{10}

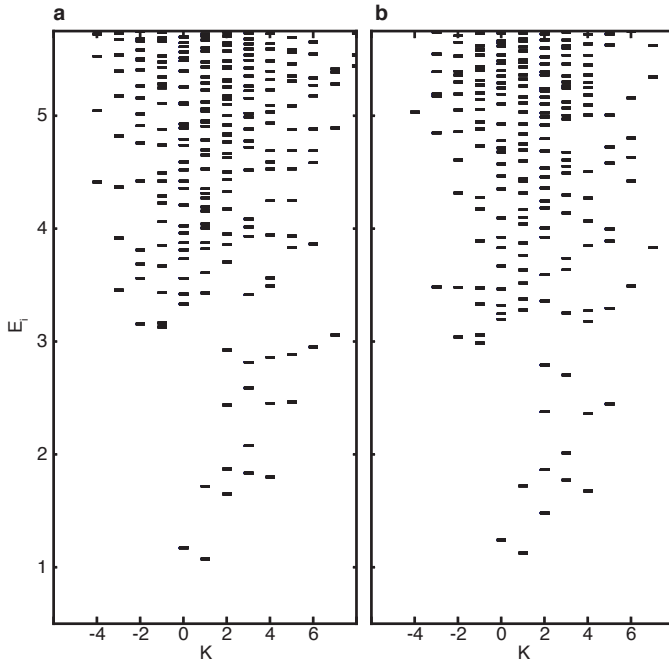


FIG. S7. **Entanglement spectrum from cylinder DMRG simulations of the incompressible half-filled Landau level.** E_i is the spectrum of the reduced density matrix for one half of the cylinder, $E_i = -\log(\rho_L)$, plotted against the momentum k around the cylinder. The counting of low lying levels, $1, 2, 4, \dots$, mimics the energy spectrum of the chiral edge theory of the Pfaffian phase, which we use to identify the state as the Pfaffian. The spectrum is shown for BLG model parameters $\Delta_{10} = 0.1E_C$, $B = 12\text{T}$ on a circumference $L = 19\ell_B$ cylinder. We repeat the calculations for two different values of the screening length (a) $a_{scr} = 0$ and (b) $a_{scr} = 0.75a_*$, where $a_* = \frac{E_C}{\hbar\omega_c}$. We find that the Pfaffian is stable over a wide range.

between them is small and mixing between them plays a crucial role in stabilizing the Pfaffian. As discussed, within the four-band model of graphene, the $N = 0$ orbital has the character of a conventional $n=0$ LL of GaAs, while the $N = 1$ orbital is an admixture of the conventional $n=0$ and $n=1$ LLs. Letting $\bar{\rho}_{NN'}(q) \equiv \sum_k e^{-ikq_y} \ell_B^2 c_{N,k+q_x/2}^\dagger c_{N',k-q_x/2}$ denote the “guiding center” density operator projected into orbitals N, N' , [16] the density operator is

$$n_{ZLL}(q) = \sum_{N,N'} \mathcal{F}_{N,N'}(q) \bar{\rho}_{NN'}(q) \quad (\text{S25})$$

where $\mathcal{F}$ are the BLG “form factors” for LL projection, which depend on the lattice structure of the wavefunctions, Eq.S22. Up to small corrections at the lattice scale of order $\frac{a_0}{\ell_B}$, the BLG form factors $\mathcal{F}_{MN}$ can be expressed as linear combinations of the conventional form factors F_{mn} as

$$\mathcal{F}_{00} = F_{00} \quad (\text{S26})$$

$$\mathcal{F}_{11} = \cos^2(\Theta) F_{11} + \sin^2(\Theta) F_{00} \quad (\text{S27})$$

$$\mathcal{F}_{01} = \cos(\Theta) F_{01}. \quad (\text{S28})$$

where Θ is determined from the B -dependent band structure, Eq.S22 and

$$F_{mn}(q_x, q_y) = e^{-q^2/4} \sqrt{\frac{m!}{n!}} \left(\frac{q_x + iq_y}{\sqrt{2}} \right)^{n-m} L_m^{(n-m)}(q^2/2). \quad (\text{S29})$$

The two-band model is recovered in the limit $\Theta \rightarrow 0$, while in the four-band model of bilayer graphene, Θ grows with perpendicular magnetic field B , Fig.S5 [7, 17].

Having defined the Hamiltonian, which depends on $B, \Delta_{10}/E_C$, and a , infinite-DMRG was used to obtain the ground state of an infinitely long, circumference $L_y = 19\ell_B$ cylinder. In the main text, the ground state was found on a 6×16 grid of points in the $\Delta_{10}/E_C, B$ plane at screening strength $a_{scr} = 0.38a_*$, keeping $m = 12000$ DMRG states, with truncation errors less than 10^{-6} . The computations required around 32000 cpu-hours. The Pfaffian ground state is identified by a finite correlation length ($\xi \sim 3\ell_B$ in the most robust regions) and distinctive entanglement spectrum characteristic of the associated edge chiral CFT [18, 19] (see Fig. S7). The anti-Pfaffian, in contrast, would have an entanglement spectrum with the opposite chirality. Unlike GaAs, scattering between the $N = 0$ and $N = 1$ orbitals strongly breaks particle-hole symmetry, unambiguously stabilizing the Pfaffian order over the particle-hole conjugated anti-Pfaffian.

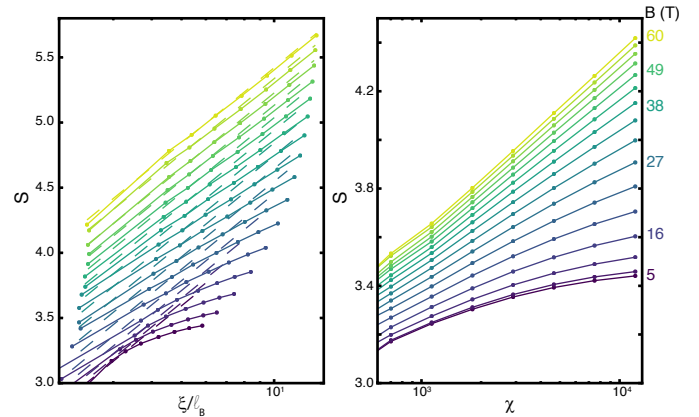


FIG. S8. **Finite entanglement scaling for the Pfaffian to CFL transition.** Data is shown for $L = 19$, $a_{scr} = 0.5a_*$, $\Delta_{10}/E_C = 0.5$ over a range of B . For each B , the DMRG simulations are repeated with increasing bond dimension (e.g., accuracy) $\chi = 600, \dots, 12000$. The bipartite entanglement S and correlation length ξ are measured at each point. (a) Numerically calculated $S(\xi)$ (solid) compared with the theoretical prediction for a CFL, $S = \frac{c}{6} \log(\xi) + s_0$, $c = 5$ (dashed). The log-linear scaling at high B is consistent with a CFL, while the behavior at low B is not, indicating a Pfaffian. (Right) $S(\chi)$. For low B , the entanglement is converging with bond dimension, indicating the state is the gapped Pfaffian. For high B , the scaling is log-linear up to the largest χ , consistent with the log-divergent entanglement of the CFL. Arguably the entanglement shows signs of saturating up $B \sim 34 - 38\text{T}$, beyond which we can't tell, suggesting the Pfaffian is stable up to at least this range.

For the reader more interested in how we have distinguished between the Pfaffian and anti-Pfaffian, we refer to the detailed DMRG study of this question described in Ref. 14, which studied a cut equivalent to $a_{\text{scr}} = 0$, $\Theta = 0$, Δ_{10} .

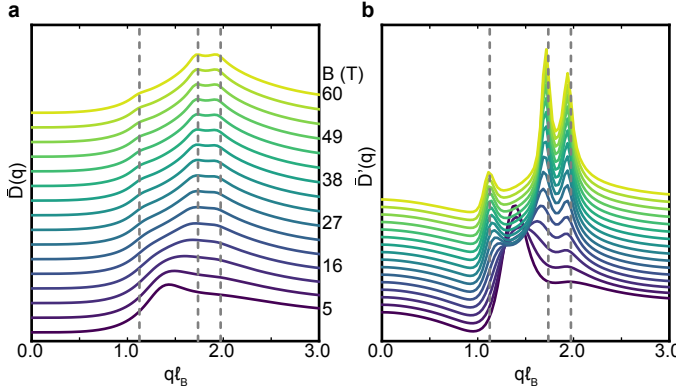


FIG. S9. Structure factor across the Pfaffian-composite fermi liquid transition. The guiding center density-density structure factor $\bar{D}(q) = \langle : n(q_x = q, q_y = 0) n(q_x = -q, q_y = 0) : \rangle$ (a) and its derivative, $\bar{D}'(q)$ (b). The gray vertical lines indicate the location of predicted kinks for a perfectly circular composite Fermi surface. At low B , where the state is a Pfaffian, there is only a single broad peak unattached to these locations. At high B , where the state is transitioning to a CFL, three sharp peaks develop. They are slightly displaced from the naive prediction because the circular Fermi deforms slightly when it is placed on a cylinder.[20]

Larger Θ (possibly achievable at the very highest B fields) increases the correlation length of the Pfaffian and drives a transition into the compressible CFL phase, which is to be expected since sharper interactions favor the CFL phase.[21–23] Since the competing CFL phase is gapless, computing the precise location of the Pfaffian-CFL phase boundary is extremely difficult; finite size or finite entanglement effects turn the continuous transition into a crossover. In particular, because DMRG simulations at bond dimension χ can only capture at most $S = \log(\chi)$ entanglement, while the CFL has a log-divergent entanglement, it is impossible to exactly capture the CFL in DMRG. As discussed in detail in Ref.[20], the DMRG thus induces a “finite-entanglement” correlation length ξ . In the CFL phase, $\xi \sim \chi^\kappa$ diverges with increasing bond dimension, while in the Pfaffian phase, ξ should converge to its physical value. Furthermore, in the CFL, the entanglement should scale with the finite-entanglement correlation length as $S(\chi) = \frac{c}{6} \log(\xi(\chi)) + s_0$, where $c = 5$ at circumference $L = 19$.

In Fig. S8, we plot both $S(\chi)$ and $S(\xi)$ at $L = 19$, $\Delta_{10}/E_C = 0.5$, $a_{\text{scr}} = 0.5a_*$ as the field increases from $B = 5 \cdots 60$ T. There is a regime where the simulations are clearly converging with χ , indicating that the state is the gapped Pfaffian, and regions where the scaling is consistent with the CFL up to the largest χ . Precisely pinpointing the transition is clearly difficult, since we can’t really distinguish between a Pfaffian phase of correlation length $\xi > 13$ and the

CFL.

Further insight is provided by the guiding-center density-density structure factor $\bar{D}(q)$ shown in Fig. S9. As discussed in Ref.20, in the CFL phase $\bar{D}(q)$ is predicted to have non-analyticities at wave vectors associated with scattering a composite Fermion across the Fermi surface. At low B , there is only a broad bump, consistent with a Pfaffian, while at high B three kinks develop close to the positions predicted for a circular Fermi surface. These kinks are brought into sharper relief by plotting “ $\partial_{|q|} \bar{D}(q)$,”[20] which is defined by multiplying the real-space correlations by $|x|$ before Fourier transforming to q_x . Like the scaling of the entropy, the structure factor is consistent with a Pfaffian to CFL transition.

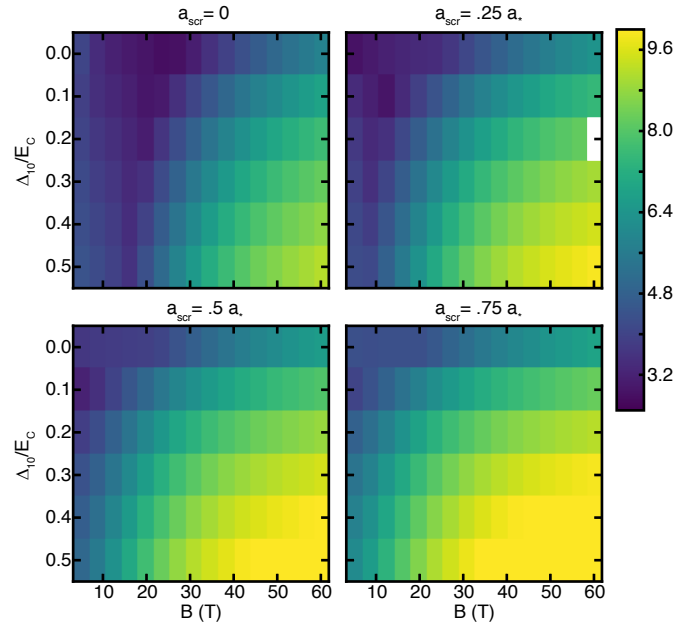


FIG. S10. Correlation length in the $B - \Delta_{10}/E_C$ plane. The computed DMRG correlation length ξ/ℓ_B is computed for four values of the screening strength a_{scr} . The plots are analogous to that in Fig. 2d of the main text. Computations are performed on an $L = 19\ell_B$ cylinder, with $\chi = 6000$ DMRG states. Larger screening increases the correlation length somewhat, but the general features are changed (e.g. the entanglement spectrum is consistent with the Pfaffian in the low- ξ regime, Fig.S7)

We note that a third competing phase is a striped phase, which breaks translation invariance. We have checked for this possibility by initializing the DMRG simulations with a charge density wave state close to the known wavelength of the stripe.[22] Throughout the phase diagram studied here, the stripes melt and form a liquid at sufficiently high DMRG accuracy. However, we do find that there is a critical $\Delta_{10} < 0$ where the Pfaffian phase is destroyed in favor of a stripe. Potentially, experiments could reach this regime by tuning Δ_{10} with very large electric fields (since the $N = 0, 1$ orbitals have slightly different layer polarization), but none of our experiments are in this regime.

To address the uncertainty in the screening strength, we present calculations ($m=6000$, truncation error less than 10^{-5}) for a variety of screening strengths a_{scr} . Again, the Pfaffian is preferred throughout most of the phase diagram. The screening has some quantitative effect on the growth of the correlation length, and hence presumably the precise location of the Pfaffian to CFL transition.

While we computed a phase diagram in the $\Delta_{10}/E_C, B$ plane, we do have a microscopic estimate of the splitting Δ_{10} . As discussed in detail in Ref.[7], it has three contributions: a B -dependent single particle splitting due to four-band corrections, a single particle splitting proportional the applied electric field p_0 , and a “Lamb-shift” contribution arising from the exchange interaction with the levels below the ZLL. The data from Sample C shown in Fig. 2e of the main text were taken at $p_0=6.0\text{V}$. The trajectory shown in Fig. 2d of the main text is calculated including the effects of B - and p_0 induced orbital splitting for $p_0 = 6\text{V}$ within the tight binding model described above, and also include the Lamb shift. The p_0 dependence of Δ_{10} is an interesting feature, since it can be used to tune the strength of the LL-mixing in-situ.

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ADDITIONAL DATA

TABLE S1. Thermodynamic energy gaps in the single component N=0 orbital regime (Sample A). Gaps are measured at B=14T and expressed in Kelvin.

	Temperature, mK								
$\tilde{\nu}$	20	160	310	450	577	711	810	1000	1150
2/7	0.63	0.77	0.59	0.38	0.3	0.2	-	-	-
1/3	-	-	-	-	-	-	-	3.77	3.1
2/5	-	-	-	3.85	3.25	2.63	2.25	1.4	1.15
3/7	-	-	1.89	1.46	1.03	0.625	0.53	0.31	-
4/9	0.83	0.8	0.62	0.40	0.29	0.15	-	-	-
5/11	0.345	0.22	0.13	0.11	-	-	-	-	-
6/13	0.12	0.07	-	-	-	-	-	-	-
7/13	0.07	0.06	0.04	-	-	-	-	-	-
6/11	0.35	0.22	0.18	0.13	-	-	-	-	-
5/9	0.96	0.98	0.79	0.57	0.38	0.2	0.18	-	-
4/7	2.9	2.67	2.34	1.83	1.36	0.87	0.89	0.48	0.34
3/5	-	-	-	6.0	5.23	4.36	3.8	2.7	2.1
2/3	-	-	-	-	-	-	-	10.5	9.56

TABLE S2. Thermodynamic energy gaps in the single component N=1 orbital regime. Gaps are measured at B=14T and expressed in Kelvin.

	Temperature, mK								
$\tilde{\nu}$	20	160	310	450	577	711	810	1000	1150
1/5	1.3	0.9	0.94	0.51	0.3	-	-	-	-
2/7	0.9	0.39	0.33	0.15	0.1	-	-	-	-
1/3	-	-	-	3.15	2.94	2.4	1.76	0.78	0.63
1/2	3.99	3.17	2.68	1.7	1.01	0.57	0.46	-	-
7/13	0.9	0.44	0.4	0.28	-	-	-	-	-
2/3	-	-	-	4.35	3.79	3.12	2.6	1.7	1.1
5/7	1.73	1.02	0.8	0.53	0.43	0.2	0.18	-	-
4/5	1.14	0.98	0.81	0.57	0.43	0.21	0.16	-	-

TABLE S3. Frequency and excitation voltages used in capacitance data presented in main text, methods, and supplementary figures.

Figure	δV_{ex} (mV)	f (kHz)
1b	1.4	71.77
1d	0.22	85.77
1e	0.9	2.48
2a,b	0.9	2.48
2e (L)	0.9	2.48
2e (R)	1.6	17.7
3c	0.9	71.77
3e	2.96	8.17
S2a-c	2.96	8.17
S2d	0.9	8.17
S11	2.96	71.77
S12	2.1	81.7
S13	1	71.77
S14	.2	87.77
S15	4.7	84.77
S16	1.6	17.7
S17	0.9	2.48

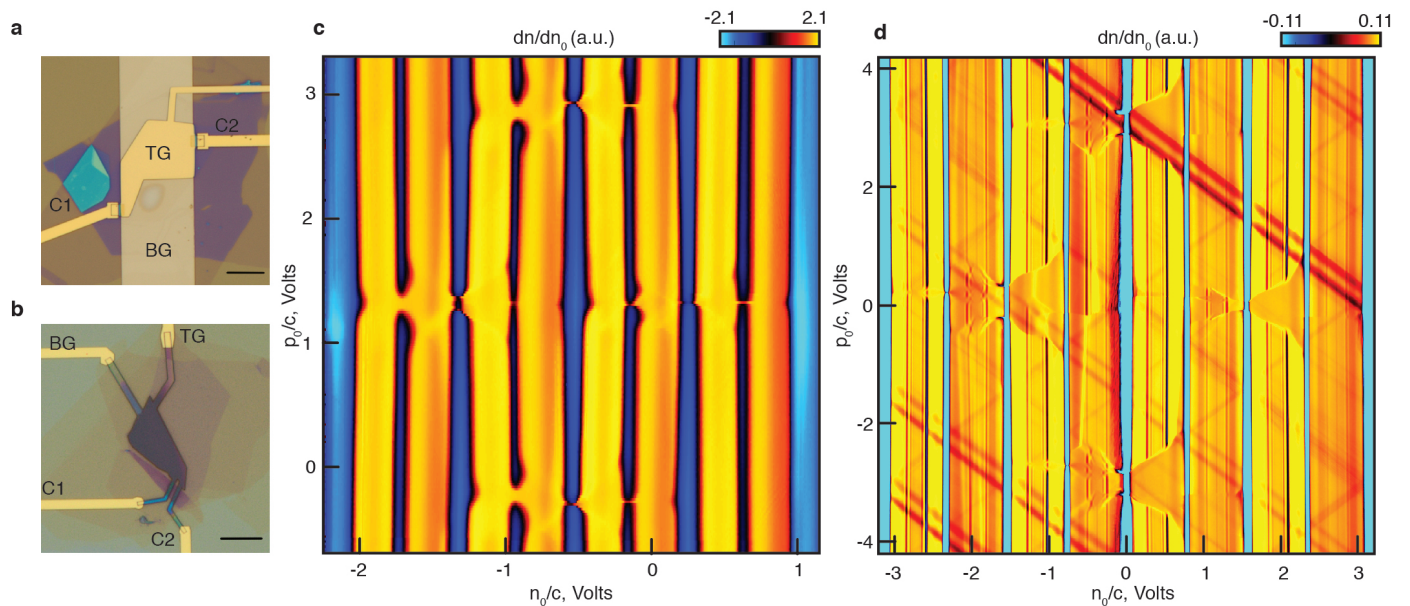


FIG. S11. **Comparison of metal- and graphite-gated devices.** (a) Optical micrograph of a metal gated device. Scale bar corresponds to 10 μm . (b) Optical micrograph of a graphite gated device. Scale bar corresponds to 10 μm . (c) Symmetric capacitance, $C_S = C_T + C_B = \partial n / \partial n_0$, for a metal gated device at $B=10$ T and $T \approx 50$ mK. (d) C_S for a graphite gated device at $B=12$ T. Graphite gated devices show much narrower integer QH states, along with many fractional states. Diagonal features in (d) are features that depend on only one of the gates (top or bottom) potentials V_B or V_T , indicating either single gated regions or electronic structure within the graphite gates.

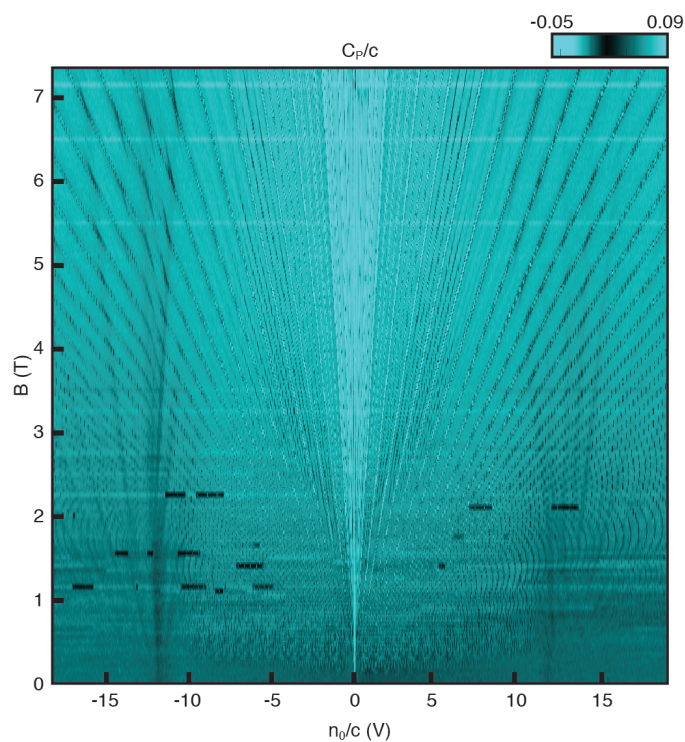


FIG. S12. **Moire features in Sample A.** Landau fan plot of C_p/c in Sample A. The main fan centered at charge neutrality is visible, along with satellite fans originating from superlattice zone boundary at $n_0/c = \pm 11.8$ V. The location of the satellite peaks imply a superlattice constant $\lambda \approx 13.3$ nm, corresponding to near-perfect (within experimental error) angular alignment of the Bernal bilayer to the underlying boron nitride.

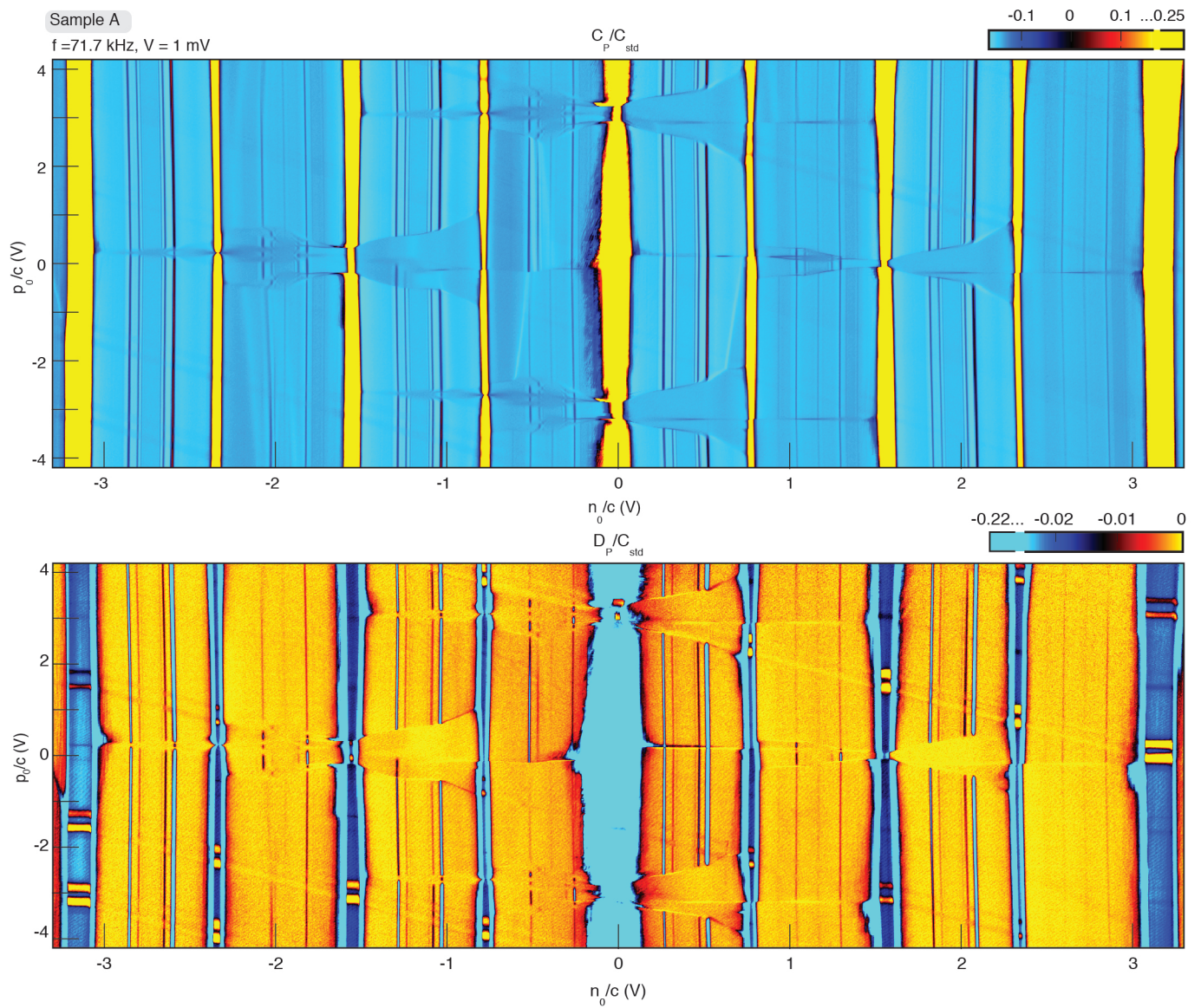


FIG. S13. **Additional data from Device A.** Penetration field capacitance and dissipation for the data set shown in Figure 1 of the main text from Device A (#SZ13), taken at base temperature and $B=12T$. Data was taken with a 1 mV AC excitation at 71.77 kHz applied to the top gate, and is shown in units of an arbitrary fixed reference capacitor.

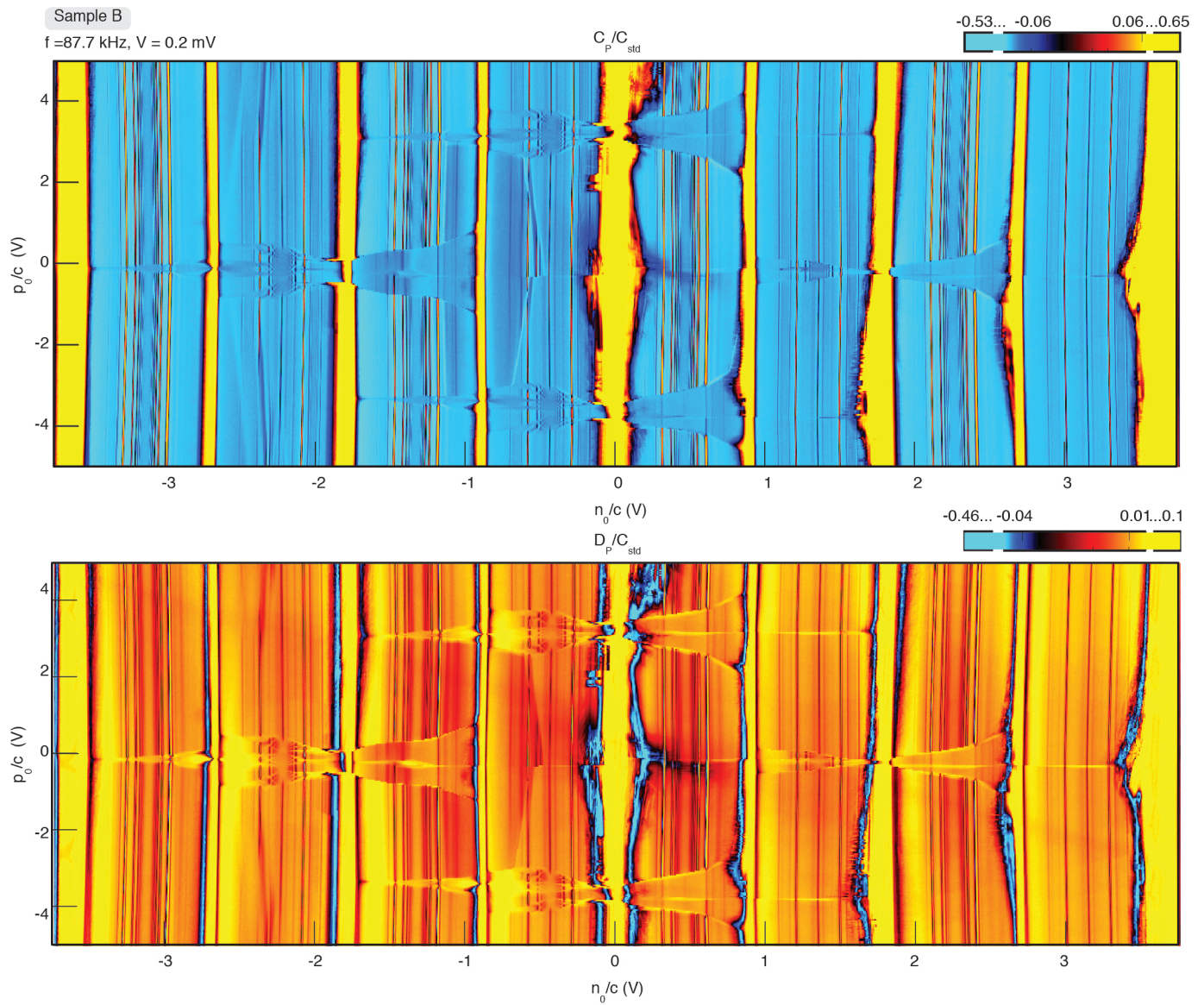


FIG. S14. **Additional data from Device B.** Penetration field capacitance and dissipation for Device B (#EMS13), taken at base temperature and $B=14\text{T}$. Data was taken with a 0.2 mV AC excitation at 87.77 kHz applied to the top gate, and is shown in units of an arbitrary fixed reference capacitor.

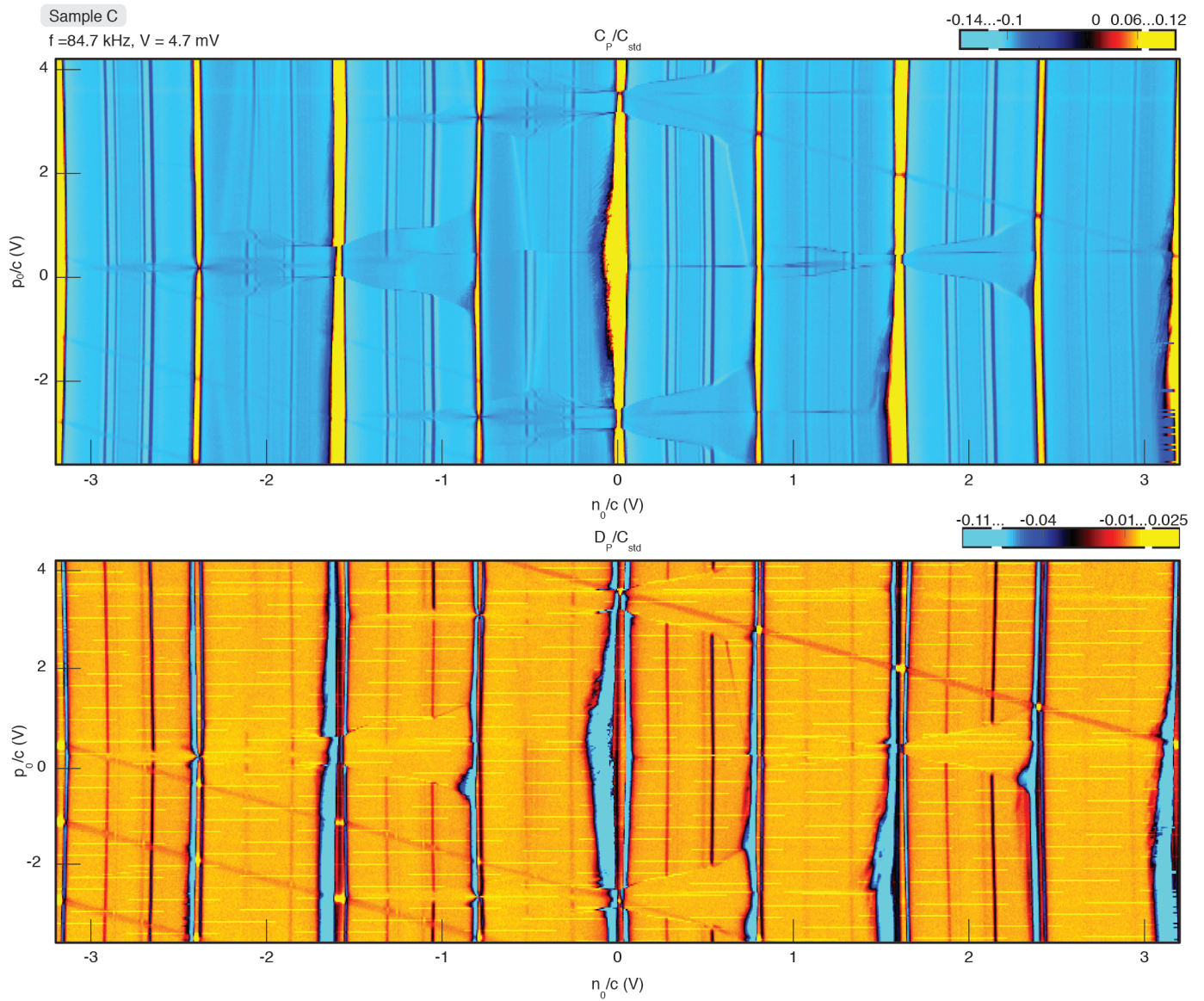


FIG. S15. **Additional data from Device C.** Penetration field capacitance and dissipation for Device C (#HZS63), taken at base temperature and $B=14$ T. Data was taken with a 4.7 mV AC excitation at 84.77 kHz applied to the top gate, and is shown in units of an arbitrary fixed reference capacitor.

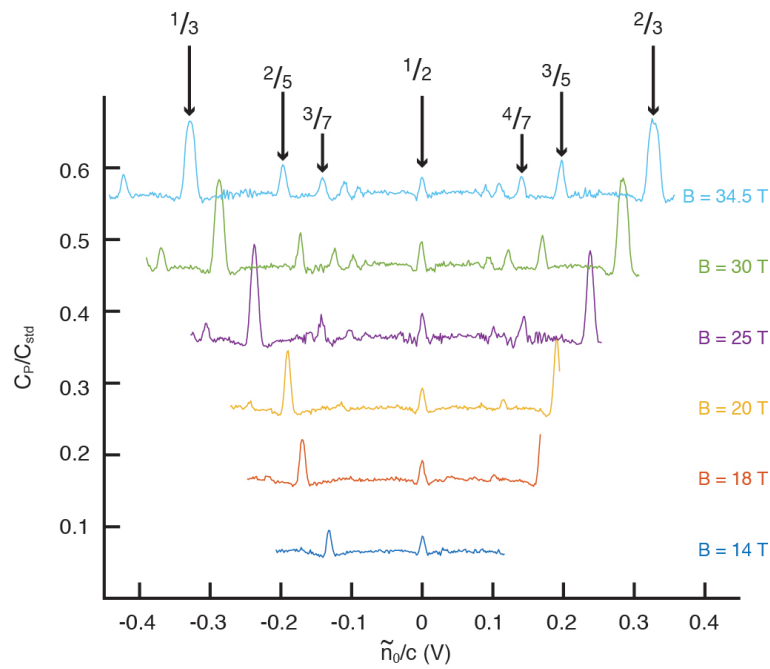


FIG. S16. **Pseudopotential tuning.** C_{pen} at different magnetic fields in Device C. Data are taken between $\nu = 1$ and $\nu = 2$ at $p_0/c = 6$ V, corresponding to partial occupation of an $N=1$ orbital level. The line-traces are offset for visibility. As the magnetic field is increased the structure of the wavefunction is tuned from $N=0$ -like to $N=1$ -like, and a hierarchy sequence of odd denominator states appears.

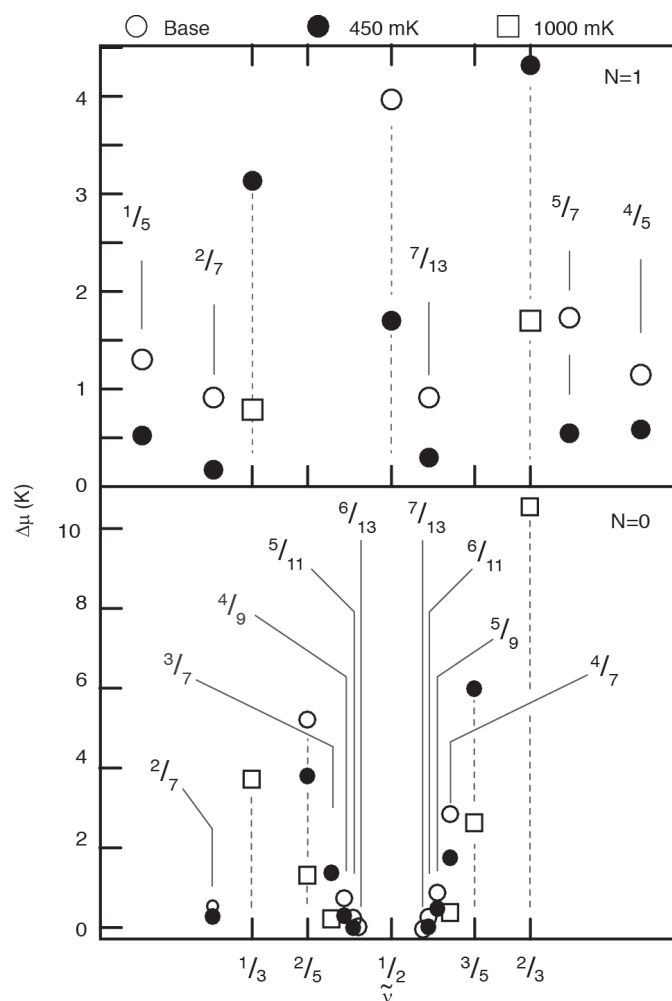


FIG. S17. **Measured gaps of fractional quantum Hall states** Data are taken from Device A at $B=14$ T, within one of the single component regimes between $\nu = 0$ and $\nu = 1$ (for the $N=0$ orbital) and $\nu = 1$ and $\nu = 2$ (for the $N=1$ orbital). Measurements are taken at temperatures from 20 mK to 1.1 K