# Supplementary Information: Local spectroscopy of a gate-switchable moiré quantum anomalous Hall insulator

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## Supplementary Note 1: Identifying the three stacking regions of tMBLG

STM topographic images of tMBLG (Fig. 1b, Fig. 3e,g) typically show three distinct regions within each moiré unit cell, which we call "bright", "intermediate" and "dark" based on their

apparent heights for  $|V_{\text{Bias}}| > 100 \text{ mV}$ . We determined their local stacking orders by analysing structural as well as electronic contributions to the apparent height. AAB stacking in tMBLG is energetically unfavourable (due to strong repulsion between the carbon atoms in the "AA" layers) and exhibits an out-of-plane structural displacement, so we identify the "bright" region as AAB stacking. ABC and BAB stackings, on the other hand, have similar binding energies to each other and similar structural heights, so atomic structure alone is inadequate to explain the difference between the observed "intermediate" and "dark" regions. We have calculated the local density of states (LDOS) at the ABC and BAB sites using the continuum model (see Supplementary Note 6) and integrated them from –200 to 0 meV (simulating a negative  $V_{\text{Bias}}$ ) or from 0 to 200 meV (simulating a positive  $V_{\text{Bias}}$ ), and find that ABC always displays a higher intensity than BAB. Since a larger integrated LDOS corresponds to a higher apparent height in constant-current STM measurements, we identify the "dark" region as corresponding to BAB stacking and the "intermediate" region ABC stacking.

### Supplementary Note 2: Comparison between transport and STM/STS measurements

In transport measurements on tMBLG the graphene layers are usually encapsulated between two pieces of hBN with a top gate and a bottom gate on two sides.<sup>1-4</sup> This allows independent tuning of carrier density *n* and out-of-plane electric field  $\mathbf{E} = (0, 0, E)$  in the graphene stack through combination of top and bottom gate voltages. The electric field creates a potential difference  $\Delta_U$  =  $eEd_a$  between adjacent graphene layers ( $d_a = 0.33$  nm is the inter-layer distance) which impacts the shape and alignment of flat bands and hence the correlated states of tMBLG. Supplementary Figure 2 shows a schematic of  $R_{xx}$  as a function of both n and E in a typical transport measurement of tMBLG (from Ref. <sup>1</sup>). Correlated insulating states emerge at v = 1, 2, 3 over a finite range of E, demonstrating electric-field-tuning of correlation effects in tMBLG. In our STM/STS measurement geometry, a dedicated top gate is not achievable due to the existence of the STM tip. Both n and E are controlled by the back-gate voltage  $V_{\rm G}$  and cannot be independently tuned (tip-induced gating is not observed in our experiment due to deliberate work function matching between graphene and the tip material). The different chemical environment of exposed carbon atoms in the top layer and those in contact with hBN in the bottom layer leads to an additional inter-layer potential difference  $\Delta_{U0}$ . The overall inter-layer potential difference is  $\Delta_U = \Delta_{U0} + eEd_a$  where E is directly related to n by  $E = \frac{ne}{2\varepsilon_{\text{off}}\varepsilon_0}$  ( $\varepsilon_{\text{eff}}$  is the effective out-of-plane dielectric constant of tMBLG). The parameter space in STM/STS measurements therefore corresponds to a "diagonal" line-cut in Supplementary Fig. 2,

which touches the v = 2 and the v = 3 correlated insulating states while missing the v = 1 one. This explains why correlation gaps appear in dI/dV at v = 2, 3 but not at v = 1 in our data.

## Supplementary Note 3: Response of the v = 3 correlation gap size to an applied out-of-plane magnetic field

The size of the v = 3 correlation gap should be modified by application of an out-of-plane magnetic field **B** = (0, 0, *B*) due to the Zeeman energy associated with non-zero orbital magnetic moments, analogous to the case in Ref. <sup>5</sup>. We can estimate the magnitude of this effect by considering single-particle effects and assuming that each C = +2 sub-band in the CFB manifold has a magnetization of  $m_{C = \pm 2}$  as defined in Supplementary Note 6. For the K+ valley-polarized  $C_{\text{tot}} = +2$  state (Fig. 4a), the doubly-occupied C = +2 sub-bands then shift downward by  $B | m_{C = \pm 2} |$  (assuming B > 0), whereas both the occupied and the unoccupied C = -2 sub-bands shift upward by the same amount. The correlation gap should remain the same since it lies between the two C = -2 sub-bands. For the K- valley-polarized  $C_{\text{tot}} = -2$  state (Fig. 4b), on the other hand, the correlation gap (which lies between the doubly occupied C = -2 sub-bands and the unoccupied C = +2 sub-band) should decrease by  $2B | m_{C = \pm 2} |$ . We estimate this shift to be less than 2 meV at B = 2 T based on calculated  $m_{C = \pm 2}$  values (Supplementary Fig. 9b-d), and so this effect is obscured by thermal and instrumental broadening and is not significant in our data.

### Supplementary Note 4: Comparison between QAH phases in tMBLG and MA-tBLG

In addition to tMBLG, the QAH effect has also been observed in magic-angle twisted bilayer graphene (MA-tBLG) aligned with an hBN substrate.<sup>6</sup> The underlying physics of the QAH phases in tMBLG and hBN-aligned MA-tBLG are similar, but two major differences have been reported in transport experiments. First, the Hall conductance is quantized to  $\pm e^2/h$  for MA-tBLG and  $\pm 2e^2/h$  for tMBLG, since the Chern number for QAH states of MA-tBLG is  $\pm 1$  while for tMBLG it is  $\pm 2$ . Second, gate-induced Chern number switching has only been observed in tMBLG. This likely depends on the details of band structure, as discussed in Ref.<sup>7</sup>. In terms of local spectroscopy, signatures of Chern insulating behaviour have been detected via STM/STS in hBN-aligned MA-tBLG under low magnetic fields,<sup>8</sup> but no systematic study on the QAH phase has been performed yet.

### **Supplementary Note 5: Determining the correlation gap position**

We determined the  $V_{\rm G}(v)$  values at which the correlation gap appears (Fig. 3f,h) using the following procedure. Supplementary Figure 7a shows normalized dI/dV at  $V_{\text{Bias}} = 0 \text{ mV} (E_{\text{F}})$  as a function of  $V_{\rm G}$  for different *B* values (this is for a small-strain region; the data is the same as in Fig. 2j). The single dip at v = 3 for B = 0.0 T and two separate dips above and below v = 3 for finite B signify the formation of correlation gaps at these filling factors. The dip features, however, are often accompanied by prominent peaks indicated by red arrows in Supplementary Fig. 7a. To understand their origin, we carefully examine Fig. 2d-i (Fig. 2d is reproduced with slightly different color scale in Supplementary Fig. 7b as an example) and focus on the features that give rise to the dip-adjacent peaks in Supplementary Fig. 7a. As V<sub>G</sub> increases, they rapidly shift to higher energies (highlighted by the dashed box in Supplementary Fig. 7b), which is the opposite direction compared to the movement of CFB and VFB peaks. This suggests that they likely arise from tip-induced charging<sup>9,10</sup> and do not reflect the actual LDOS in tMBLG. For every curve in Supplementary Fig. 7a, we thus exclude the dip-adjacent peaks and perform linear fitting on the two sides of each dip (fitted lines are shown for B = 0.0 T as an example). The data points in Fig. 3f indicate intersection of fitted lines and the error bars come from the fitting errors. The same procedure is conducted for data from the largestrain region (Supplementary Fig. 7c; the data is the same as in Supplementary Fig. 6f) and the results are plotted in Fig. 3h.

### **Supplementary Note 6: Theoretical model and calculations**

**Continuum model of tMBLG.** Our calculations were based on the Bistritzer–MacDonald continuum approach to moiré structures.<sup>11,12</sup> Here the monolayer graphene is modelled using a twoband tight-binding model with  $t_0 = 2.8$  eV while the Bernal-stacked bilayer is modelled using a fourband model with  $t_0 = 2.61$  eV,  $t_1 = 0.361$  eV,  $t_3 = 0.283$  eV,  $t_4 = 0.138$  eV,  $\Delta = 0.015$  eV.<sup>13</sup> The bilayer is then rotated by angle  $\theta$  and hybridized with the monolayer with intra-sublattice strength  $w_{AA} = 87.75$  meV and inter-sublattice strength  $w_{AB} = 117$  meV. The hetero-strain is taken into consideration as an artificial vector field, following the treatment in Ref.<sup>14</sup>. We further add a potential difference  $\Delta_U = (12.6 + 4.69 v)$  meV between adjacent graphene layers to account for the gate-induced out-of-plane electric field as well as the built-in asymmetry between top and bottom layers due to the presence of the hBN substrate (Supplementary Note 2). The resulting continuum model is truncated by keeping all states within a radius of 6 mini-Brillouin zones (mBZs) of the  $\gamma$ point. **Decomposing orbital magnetization into bulk and edge components.** When the chemical potential  $\mu$  resides inside the v = 3 correlation gap, the total orbital magnetization can be decomposed into the bulk part which is independent of  $\mu$  and the edge part which depends linearly on  $\mu$ . There is no unique way of defining these two pieces. We find it convenient to use the following convention:

$$M_{\text{bulk}} = \frac{e}{2\hbar} \sum_{n} \int d^2 \mathbf{k} \,\epsilon^{ab} \,\operatorname{Im} \langle \partial_a u_{n,\mathbf{k}} | H_{\mathbf{k}} + \mathcal{E}_{n,\mathbf{k}} - 2\mathcal{E}_{\text{max}}^{\text{OC}} | \partial_b u_{n,\mathbf{k}} \rangle \tag{1}$$

$$M_{\text{edge}}(\mu) = -\frac{e}{\hbar} \sum_{n} \int d^2 \mathbf{k} \, \epsilon^{ab} \, \text{Im} \langle \partial_a u_{n,\mathbf{k}} | \mu - \mathcal{E}_{\text{max}}^{\text{OC}} | \partial_b u_{n,\mathbf{k}} \rangle = \frac{e}{\hbar} \mathcal{C}_{\text{tot}}(\mu - \mathcal{E}_{\text{max}}^{\text{OC}}) \tag{2}$$

where  $\epsilon^{ab}$  is the antisymmetric tensor,  $\mathcal{E}_{n,k}$  is the single particle energy, and  $\mathcal{E}_{max}^{OC}$  is the energy of the top of the occupied bands (i.e., the bottom of the correlation gap). This convention has the advantage that  $M_{edge}(\mathcal{E}_{max}^{OC}) = 0$ , as befitting the interpretation that  $M_{edge}$  is the contribution from chiral edge modes. Appropriate care needs to be taken to compute these quantities on a discrete grid, as described earlier in Refs. <sup>15,16</sup>.

**Calculating**  $M_{edge}$ . For a given band structure, the sign of  $M_{edge}$  always follows that of  $C_{tot}$ , while its magnitude reaches the maximum when  $\mu = \mathcal{E}_{min}^{UO}$  ( $\mathcal{E}_{min}^{UO}$  is the energy of the bottom of the unoccupied bands, i.e., the top of the correlation gap):

$$M_{\rm edge}(\mu = \mathcal{E}_{\rm min}^{\rm UO}) = \frac{e}{h} C_{\rm tot}(\mathcal{E}_{\rm min}^{\rm UO} - \mathcal{E}_{\rm max}^{\rm OC}) = \frac{e}{h} C_{\rm tot} \mathcal{E}_{\rm g}$$
(3)

where  $\mathcal{E}_{g}$  is the size of the correlation gap. To obtain  $\mathcal{E}_{g}$  values as a function of hetero-strain, we performed Hartree-Fock calculation in a momentum-space approach analogous to earlier Hartree-Fock studies of twisted graphene systems. Our code is an extension of the tBLG code used in Ref. <sup>17</sup>, the twisted double bilayer graphene (tDBLG) code used in Ref. <sup>18</sup>, and the tMBLG code used in Ref. <sup>19</sup>. The Coulomb interaction (screened by the graphene, the hBN/SiO<sub>2</sub> substrate and the metallic STM tip) is assumed to take the single-plane-screened form  $V(\mathbf{q}) = \frac{e^2}{2\epsilon_{\text{eff}}\epsilon_0 q} [1 - \exp(-2qd_S)] (\varepsilon_{\text{eff}}$ = 12 and  $d_{\text{S}} = 20$  nm were chosen on phenomenological grounds). The Coulomb matrix elements are evaluated in the basis of the continuum band structure and projected into the six bands nearest to the charge neutrality per valley and spin, for a total of 24 bands. We then consider a Slater-determinant ansatz  $|u\rangle$  that is diagonal in the mBZ momentum **k**. Discretizing the model on a  $16 \times 16 \text{ k-grid}$ , the *u*'s are iteratively adjusted to minimize the energy  $\langle u|H|u\rangle$ , using the optimal damping algorithm in Ref. <sup>20</sup> to achieve Hartree-Fock self-consistency. Supplementary Figure 8a-c shows the resulting Hartree-Fock tMBLG band structures for selected hetero-strain values. Here positive (negative) strain is defined as stretching (compressing) the bilayer and compressing (stretching) the monolayer (bilayer). Increasing the hetero-strain leads to a reduction in  $\mathcal{E}_{g}$  (Supplementary Fig. 8d) and also in  $M_{\text{edge}}$  as plotted in Fig. 4d (positive strain is assumed in the main text, but the results remain qualitatively the same for negative strain).

**Calculating**  $M_{\text{bulk}}$ . As  $M_{\text{bulk}}$  is heavily dependent on the detailed band structure, a full calculation based on the Hartree-Fock approach is extremely costly and beyond our current capabilities. Instead, we adopt the phenomenological model proposed in Refs. <sup>1,7</sup> to take electron-electron interaction into consideration. For our purpose it is convenient to take an alternative decomposition of orbital magnetization

$$M_{\text{bulk}} = \frac{e}{2\hbar} \sum_{n} \int d^{2}\mathbf{k} \,\epsilon^{ab} \,\operatorname{Im}\langle\partial_{a}u_{n,\mathbf{k}}|H_{\mathbf{k}} + \mathcal{E}_{n,\mathbf{k}} - 2\mathcal{E}_{\max,n}|\partial_{b}u_{n,\mathbf{k}}\rangle$$

$$- \frac{e}{\hbar} \sum_{n} \int d^{2}\mathbf{k} \,\epsilon^{ab} \,\operatorname{Im}\langle\partial_{a}u_{n,\mathbf{k}}|\mu - \mathcal{E}_{\max,n}|\partial_{b}u_{n,\mathbf{k}}\rangle$$

$$= \sum_{n} m_{n} + \frac{e}{\hbar} \sum_{n} C_{n}(\mu - \mathcal{E}_{\max,n})$$
(4)

where  $\mathcal{E}_{\max,n}$  is the energy of the top of the *n*-th band and  $m_n$  can be interpreted as the total orbital magnetization of that band when the chemical potential is at  $\mathcal{E}_{\max,n}$ . We start from the single-particle band structure (for which the  $m_n$ 's can be straightforwardly calculated using the dual state method of Ref. <sup>16</sup>) and analyse how electron-electron interaction impacts  $M_{\text{bulk}}$ . The primary effect of interaction is to introduce exchange energy difference between different sub-bands which causes spontaneous polarization in the spin-valley space. We expect the nature of each sub-band to remain unchanged to a good approximation. Therefore, we make several simplifying assumptions similar to those in Ref. <sup>1</sup> with slight modifications:

- 1. Interaction opens up a gap between the three occupied CFB- sub-bands and the one unoccupied CFB+ sub-band;
- 2. Interaction introduces an exchange-driven energy difference  $\delta_{\text{VFB}}$  among the VFB subbands (which have valley-dependent  $C = \mp 1$ ) (Supplementary Fig. 9a);
- 3. The energies of other bands remain unchanged;
- 4. Each  $m_n$ , evaluated at the top of each band after taking  $\delta_{\text{VFB}}$  into account, remains unchanged due to interaction.

We can evaluate the total orbital magnetization when  $\mu = \mathcal{E}_{\text{max}}^{\text{OC}}$  with these assumptions. Focusing on the K+ valley-polarized  $C_{\text{tot}} = +2$  state, we get

$$M_{\text{bulk}} = \sum_{n} m_{n} + \frac{e}{h} \sum_{n} C_{n} (\mathcal{E}_{\text{max}}^{\text{OC}} - \mathcal{E}_{\text{max},n})$$

$$= m_{C = +2} + m_{C = +2} + m_{C = -2}$$

$$+ \frac{e}{h} [(-1) (\mathcal{E}_{\text{max}}^{\text{OC}} - \mathcal{E}_{\text{max},\text{VFB}}) + (-1) (\mathcal{E}_{\text{max}}^{\text{OC}} - \mathcal{E}_{\text{max},\text{VFB}})$$

$$+ (1) (\mathcal{E}_{\text{max}}^{\text{OC}} - \mathcal{E}_{\text{max},\text{VFB}}) + (1) (\mathcal{E}_{\text{max}}^{\text{OC}} - \mathcal{E}_{\text{max},\text{VFB}} - \delta_{\text{VFB}})]$$

$$= m_{C = +2} - \frac{e}{h} \delta_{\text{VFB}}$$
(5)

 $M_{\text{bulk}}$  for the  $C_{\text{tot}} = -2$  state is the exact opposite. Here the first term (arising from single-particle effects) is parallel to  $M_{\text{edge}}$  while the second term characterizing electron-electron interaction strength is antiparallel to  $M_{\text{edge}}$ .

We find that  $m_{C = +2}$  becomes larger as the hetero-strain is increased due to a redistribution of Berry curvature and band dispersion throughout the mBZ.  $\delta_{VFB}$  is treated as a fitting parameter since it is difficult to accurately estimate the exchange coupling. Supplementary Figure 9b-d shows  $M_{\text{bulk}}(C_{\text{tot}} = +2)$  as a function of the hetero-strain for different choices of  $\delta_{VFB}$  when the twist angle  $\theta$ is fixed at 1.26°, 1.28°, and 1.32°, respectively. In the main text we use  $\delta_{VFB} = 13 \text{ meV}$  for  $\theta = 1.26^{\circ}$ (Fig. 4d), which best matches our observation that increasing hetero-strain causes  $M_{\text{bulk}}(C_{\text{tot}} = +2)$  to flip from negative to positive and the system to change from switchable to non-switchable. For 1.30°  $< \theta < 1.34^{\circ}$ , only non-switchable QAH states have been observed experimentally (Fig. 3a), but this does not preclude a similar strain-induced transition since  $M_{\text{bulk}}$  could flip its sign for strains below 0.12%, the lowest amount observed within this angle range.

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**Supplementary Figure 1: Optical microscope image of the tMBLG device.** MLG = monolayer graphene; BLG = Bernal-stacked bilayer graphene.



Supplementary Figure 2: Parameter space in transport and STM/STS measurements. The dark blue regions indicate insulating phases in transport measurement (adapted from Ref. <sup>1</sup>). The dashed line is the STM/STS parameter space.  $\varepsilon_{eff}$  is the effective dielectric constant of tMBLG, *E* is the outof-plane electric field, and  $\Delta_U$  is the corresponding inter-layer potential difference.



Supplementary Figure 3: Additional data for correlated insulating states at v = 2, 3. a, Gatedependent d*I*/d*V* density plot for the ABC stacking region over the gate range  $-70 \text{ V} \le V_G \le 70 \text{ V}$ . The vertical dashed line denotes the Fermi energy  $E_F$ . Spectroscopy parameters: modulation voltage  $V_{\text{RMS}} = 1 \text{ mV}$ ; setpoint  $V_{\text{Bias}} = 100 \text{ mV}$ ,  $I_0 = 1.55 \text{ nA}$  for  $-70 \text{ V} \le V_G \le -2 \text{ V}$ ; setpoint  $V_{\text{Bias}} = -100$ mV,  $I_0 = 0.8 \text{ nA}$  for  $0 \text{ V} \le V_G \le 70 \text{ V}$ . b, Same as a, but for the AAB stacking region. Spectroscopy parameters: modulation voltage  $V_{\text{RMS}} = 1 \text{ mV}$ ; setpoint  $V_{\text{Bias}} = 100 \text{ mV}$ ,  $I_0 = 1.4 \text{ nA}$  for  $-70 \text{ V} \le V_G \le$ -2 V; setpoint  $V_{\text{Bias}} = -100 \text{ mV}$ ,  $I_0 = 0.8 \text{ nA}$  for  $0 \text{ V} \le V_G \le 70 \text{ V}$ . Splitting of the CFB peak around v= 2, 3 is observed in spectra measured in all three regions (see also Fig. 1c). c-e, Normalized d*I*/d*V* at  $V_{\text{Bias}} = 0 \text{ mV}$  ( $E_F$ ) as a function of  $V_G(v)$  for all three stacking regions. This type of plot is analogous to a transport conductance measurement since in both cases only electronic states near  $E_F$  are being probed. Dips going down to nearly vanishing d*I*/d*V* that are centred at v = 2, 3, and 4 indicate emergence of insulating phases at these filling factors.



Supplementary Figure 4: Topological behaviour of the v = 3 state at B = 0.2 T. Gate-dependent dI/dV density plot near v = 3 at (a) B = 0.0 T and (b) B = 0.2 T ( $V_{RMS} = 1$  mV; setpoint  $V_{Bias} = -60$  mV,  $I_0 = 0.5$  nA). Arrows indicate correlation gaps.



Supplementary Figure 5: Gate-dependent dI/dV without the v = 3 correlation gap. These data were obtained in a region with  $\theta = 1.23^{\circ}$  and hetero-strain = 0.19%. The vertical dashed line denotes the Fermi energy. Spectroscopy parameters: modulation voltage  $V_{\text{RMS}} = 1$  mV; setpoint  $V_{\text{Bias}} = 100$ mV,  $I_0 = 2$  nA for -70 V  $\leq V_G \leq -2$  V; setpoint  $V_{\text{Bias}} = -100$  mV,  $I_0 = 1$  nA for 0 V  $\leq V_G \leq 70$  V. VFB = valence flat band, CFB- = lower branch of conduction flat band, CFB+ = upper branch of conduction flat band.



Supplementary Figure 6: Topological behaviour of the v = 3 state for the large-strain region. ae, Gate-dependent d*I*/d*V* density plot near v = 3 for a region with  $\theta = 1.26^{\circ}$  and hetero-strain = 0.24% at (a) B = 0.0 T, (b) B = 0.2 T, (c) B = 0.4 T, (d) B = 0.6 T, and (e) B = 0.8 T (modulation voltage  $V_{\text{RMS}} = 1$  mV; setpoint  $V_{\text{Bias}} = -60$  mV,  $I_0 = 0.3$  nA). Arrows indicate correlation gaps. **f**, Normalized d*I*/d*V* at  $V_{\text{Bias}} = 0$  mV ( $E_{\text{F}}$ ) as a function of  $V_{\text{G}}(v)$  and B. The dashed line is a guide to the eye following the Středa formula with  $C_{\text{tot}} = +2$ .



Supplementary Figure 7: Extracting correlation gap positions from gate-dependent dI/dV. a, Normalized dI/dV at  $V_{\text{Bias}} = 0 \text{ mV} (E_{\text{F}})$  as a function of  $V_{\text{G}}(v)$  for different magnetic fields (smallstrain region). Red arrows indicate tip-induced charging features. Squares with error bars are correlation gap positions. Fitted lines are plotted in grey for B = 0.0 T. b, Reproduction of gatedependent dI/dV in Fig. 2d with the charging feature highlighted (dashed white box). c, same as a, but with data from the large-strain region.



Supplementary Figure 8: Strain tuning Hartree-Fock band structure for tMBLG. a-c, Hartree-Fock band structures plotted along a high-symmetry line in the mBZ for hetero-strains of (a) -0.2%, (b) -0.1%, and (c) 0.0%. Red and blue curves represent sub-bands with different spins. d, Extracted correlation gap size as a function of hetero-strain.



Supplementary Figure 9: Calculating bulk magnetization in the presence of electron-electron interaction. **a**, Energy configuration of the K+ valley-polarized state at v = 3 showing sub-bands from both the CFB manifold and the VFB manifold.  $\delta_{\text{VFB}}$  is the interaction-induced energy offset among the VFB sub-bands. **b-d**,  $M_{\text{bulk}}$  for  $C_{\text{tot}} = +2$  state as a function of hetero-strain for different choices of  $\delta_{\text{VFB}}$  at (**b**)  $\theta = 1.26^{\circ}$ , (**c**)  $1.28^{\circ}$ , and (**d**)  $1.32^{\circ}$ .