

# Supplementary Information for

## Moiré Periodic and Quasiperiodic Crystals in Heterostructures of Twisted Bilayer Graphene and Hexagonal Boron Nitride

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### 1. Additional STS Mappings

We present another MIC with crystallographic directions of GG, GBN and the consequent MM aligned in Fig S1. The condition for such alignments is  $|\theta_{GBN}| = 0.55^\circ$ . The moiré-of-moiré-of-moiré (MMM) pattern as a second order interference of GBN and MM is observed for a wide range of energies, reflecting the incommensurability and scaling symmetry of such quasiperiodic structures. Additional STS mapping data at different energies from the MC, MIC and MQC regions in main text Fig. 4 and Fig. S1 is presented in Fig. S2-S4.

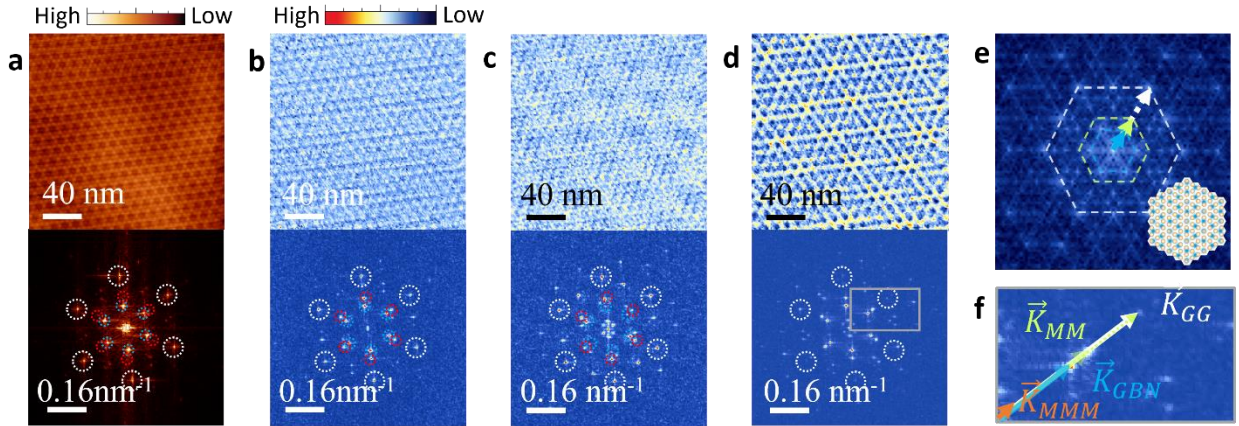


Fig. S1. **STS mapping of a MIC with the directions of  $\vec{K}_{GG}$  and  $\vec{K}_{GBN}$  aligned.** **a**, Topography of an example of incommensurate modulated crystal:  $L_{GG} = 5.43 \text{ nm}$ ;  $L_{GBN} = 12.47 \text{ nm}$  ( $\theta_{GG} = 2.58^\circ$ ;  $\theta_{GBN} = 0.56^\circ$ ). STS mapping of this region at energies  $V_B = -120 \text{ mV}$ ,  $-80 \text{ mV}$ ,  $-40 \text{ mV}$  are shown in **b-d**, respectively. **e**, A simulated aligned MIC (lower right inset) and its FFT. **f**, Zoom-in of FFT in **d**, within the grey rectangular region. The GG, GBN, MM and 2<sup>nd</sup> order interference induced moiré-of-moiré-of-moiré (MMM) wavevectors are illustrated with white, blue, green and yellow arrows, respectively. Tunneling parameters:  $V_B = -200 \text{ mV}$ ;  $V_G = 0 \text{ V}$ ;  $I = 100 \text{ pA}$

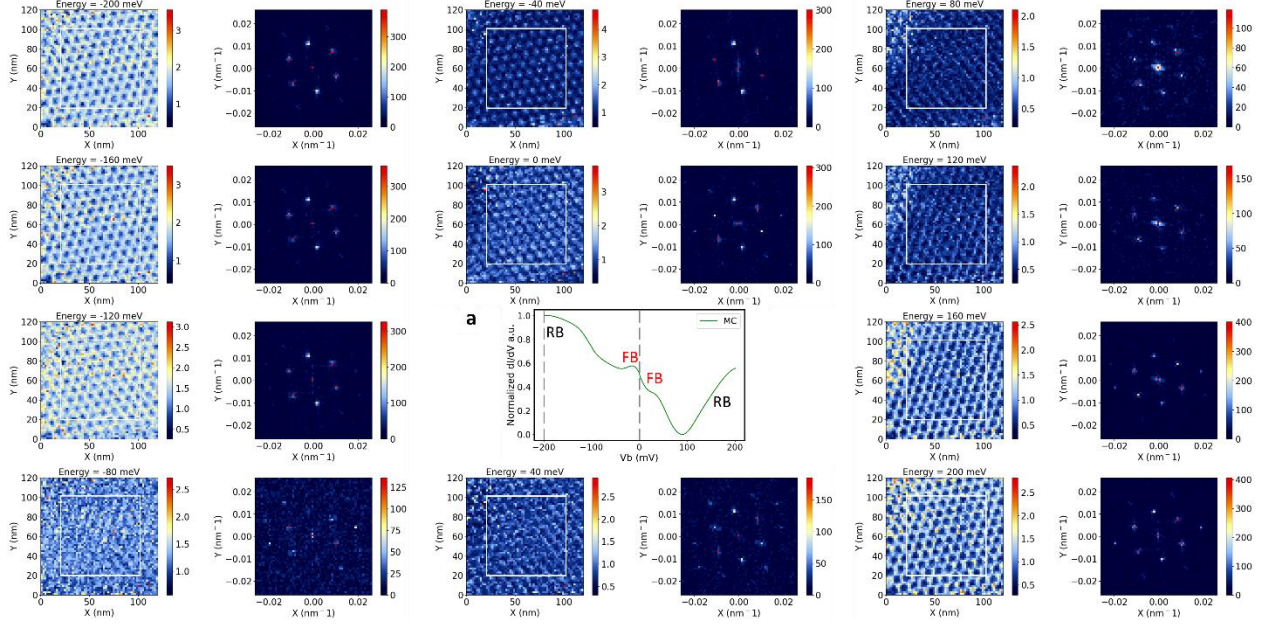


Fig. S2. **Additional STS Mapping of MC.** The center panel (a) shows averaged STS from the STS mapping of a MC region with  $L_{GG} = L_{GBN} = 10.8 \text{ nm}$  in Fig 4b. The moiré flat band are marked as FB and the remote bands are marked as RB. The rest of surrounding figures are STS mappings (left panels) and the corresponding FFTs (right panels) of the MC region presented in Fig. 4 at different  $V_B$  (from -200 mV to 200 mV). The energies of the mappings are included in their titles. The FFT only shows peaks at the 1<sup>st</sup> and 2<sup>nd</sup> BZ, indicating the C3 symmetry of the real space pattern. We note that there is a domain boundary at the upper left corner of the mapping region. Fig 4b is from the zoom-in to the regions in the white square. Tunneling parameters:  $V_B = -300 \text{ mV}$ ;  $V_G = 10 \text{ V}$ ;  $I = 100 \text{ pA}$

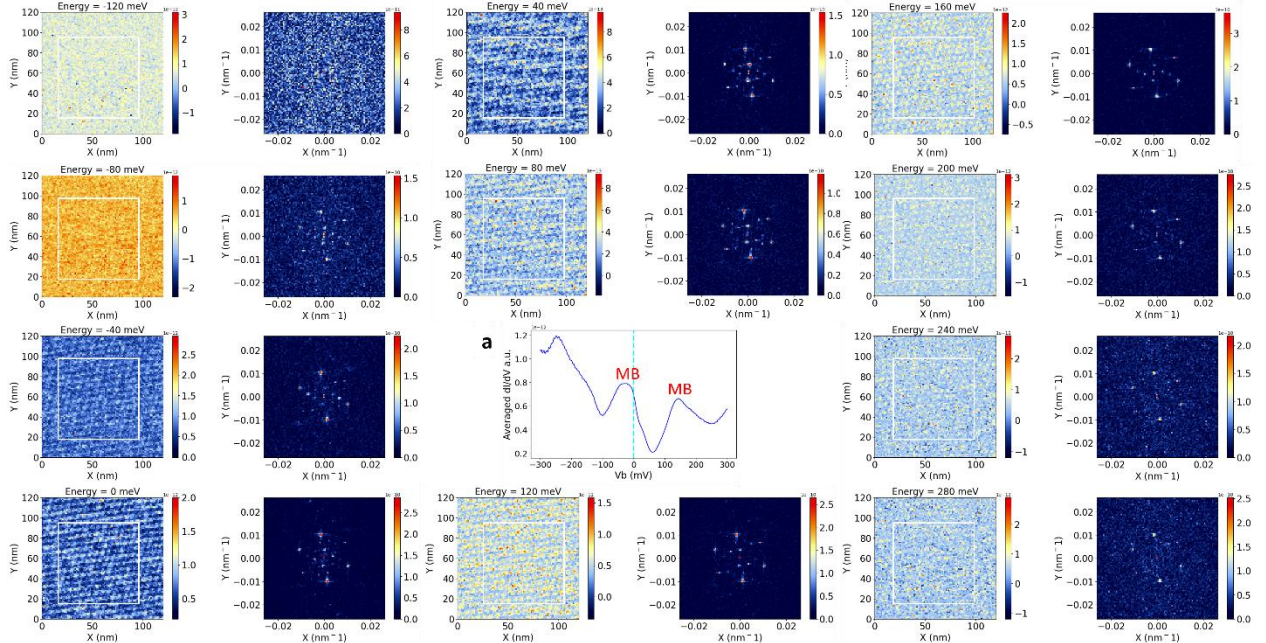
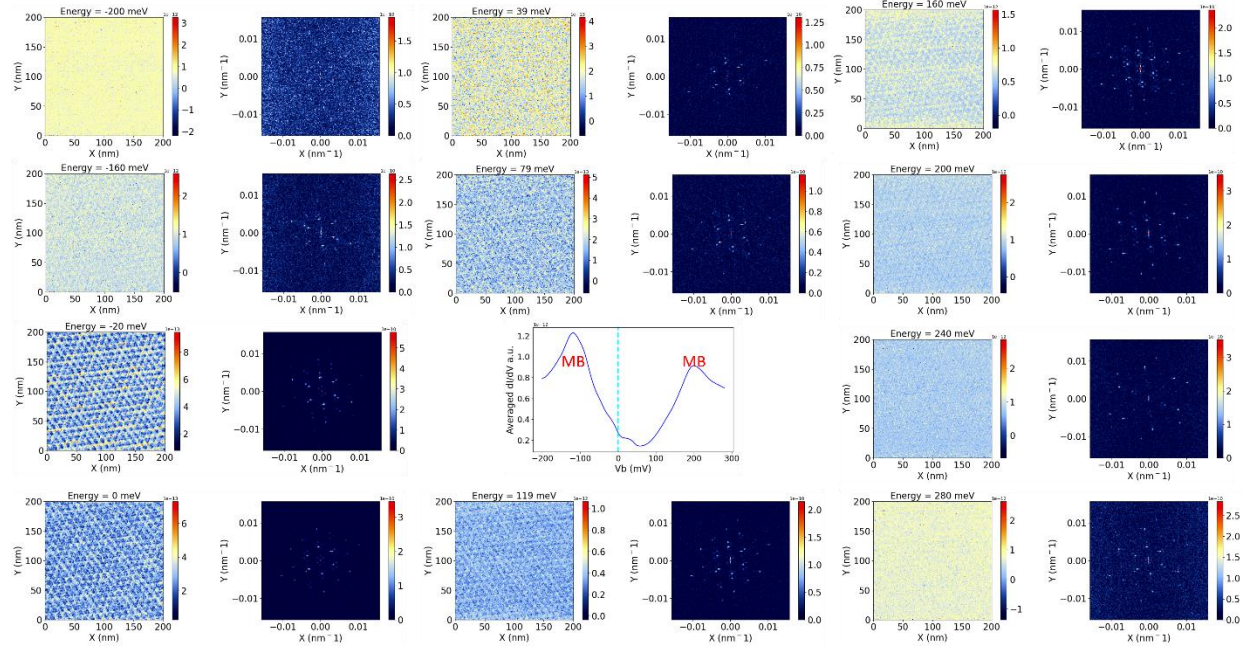


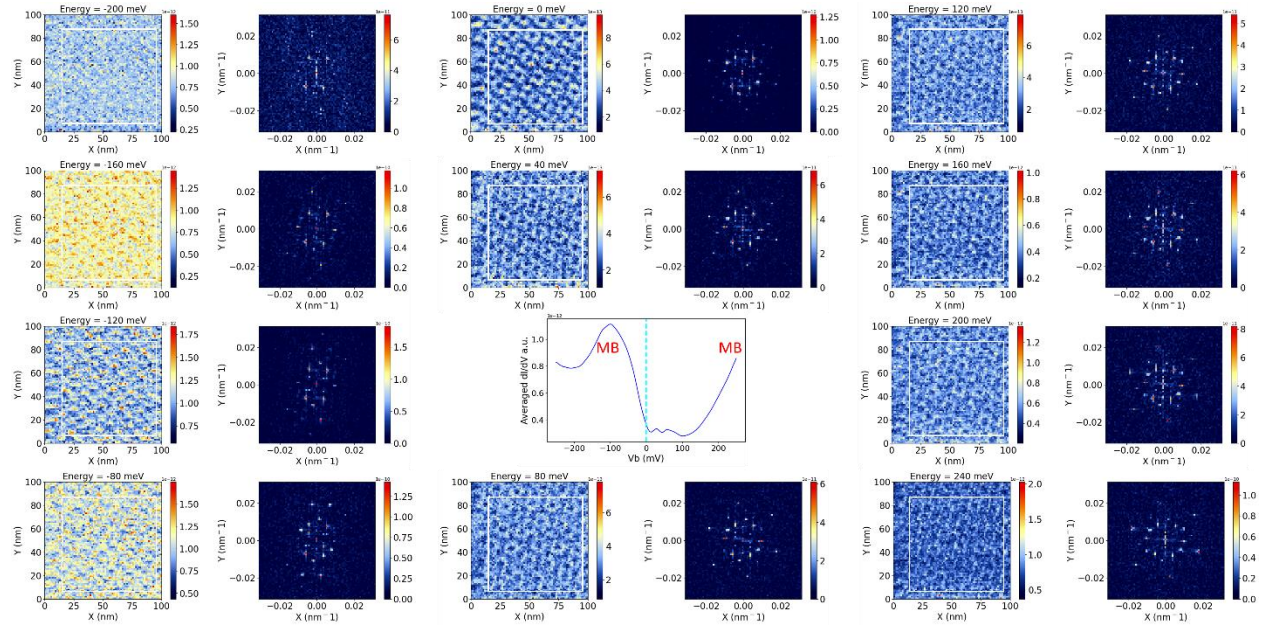
Fig. S3. **Additional STS Mapping of MIC.** The center panel (a) is averaged STS from the STS mapping of the MIC region in Fig 4f with  $L_{GG} = 7.1 \text{ nm}$ ;  $L_{GBN} = 10.9 \text{ nm}$ . The moiré bands are marked as MB in the center panel, the averaged  $dI/dV$  from all points in the map. The rest of surrounding figures are STS mappings (left panels) and the corresponding FFTs (right panels) of the MIC presented in Fig. 4f region at different  $V_B$  (from -200 mV to 200 mV)



mV). The energies of the mappings are included in their titles. Fig 4f is the zoom-in to the regions in the white square. Tunneling parameters:  $V_B = -300\text{mV}$ ;  $V_G = 0\text{V}$ ;  $I = 100\text{pA}$



**Fig. S4 Additional STS mapping of a MIC with the directions of  $\vec{K}_{GG}$  and  $\vec{K}_{GBN}$  aligned.** The center panel (a) is averaged STS from the STS mapping of the MIC region in Fig. S1 with  $L_{GG} = 5.43\text{ nm}$ ;  $L_{GBN} = 12.47\text{ nm}$ . The moiré bands are marked as MB in the center panel, the averaged  $dI/dV$  from all points in the map. The rest of surrounding figures are STS mappings (left panels) and the corresponding FFTs (right panels) of the MIC region at different  $V_b$  (from  $-200\text{ mV}$  to  $200\text{ mV}$ ). The energies of the mappings are included in their titles. Tunneling parameters:  $V_B = -200\text{mV}$ ;  $V_G = 0\text{V}$ ;  $I = 100\text{pA}$



**Fig. S5. Additional STS Mapping of MQC.** The center panel (a) is averaged STS from the STS mapping of the MQC region in Fig. S4j. The rest of surrounding figures are STS mappings (left panels) and the corresponding FFTs (right panels) of the MQC region (marked with white box in each panel here) at different  $V_b$  (from  $-200\text{ mV}$  to  $200\text{ mV}$ ).

240 mV) indicated by their titles. The intensity of GG, GBN and MM periods has different energy dependence, giving rise to the real space pattern with evolving symmetries. Fig 4j is the zoom-in to the regions in the white square. Tunneling parameters:  $V_B = -300\text{mV}$ ;  $V_G=0\text{V}$ ;  $I = 100\text{pA}$