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SUPPLEMENTARY INFORMATION

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Signatures of tunable superconductivity in a trilayer graphene moiré superlattice

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Supplementary Information

Signatures of Gate-Tunable Superconductivity in Trilayer Graphene/Boron Nitride Moiré Superlattice

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In our transport results, we found important electron-hole asymmetry and asymmetry in the displacement field direction in TLG/hBN. To understand the two asymmetries, we calculated the band structure (see Ref. 5 for calculation details) and show the results in Extended Data Fig. 7.

The single-particle bandstructure of the heterostructure is described by the Hamiltonian $H = H_{ABC} + V_M$, where H_{ABC} is the TLG Hamiltonian under a weak vertical electrical field, and V_M describes the effective potential acting on TLG from the moiré superlattice. The low-energy electronic structure of ABC-stacked TLG can be captured by an effective two-component Hamiltonian in the K valley that describes hopping between the A atom in the top graphene layer and the C atom in the bottom graphene layer[7,8],

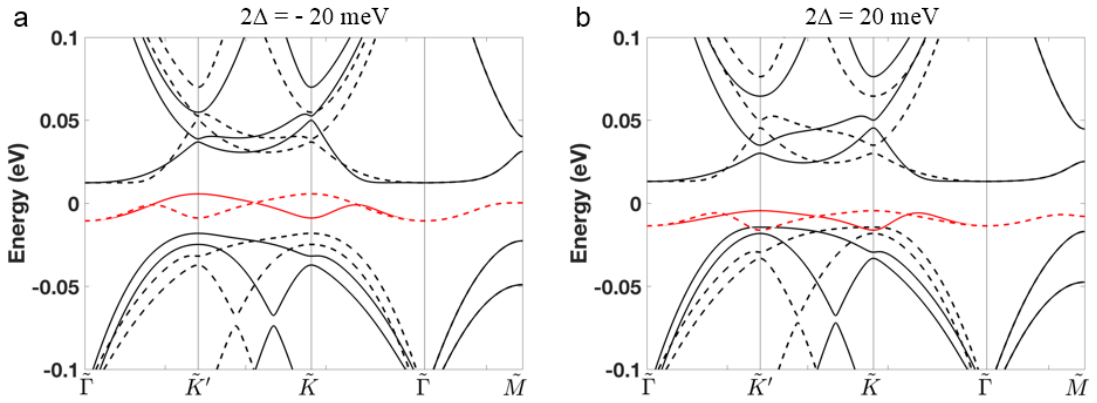
$$H_{ABC} = \frac{{v_0}^3}{t_1^2}\begin{pmatrix} 0 & (\pi^+)^3 \\ \pi^3 & 0 \end{pmatrix} + \left(\frac{2v_0v_3p^2}{t_1} + t_2\right)\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} + \left(\frac{2v_0v_4p^2}{t_1} - \Delta'\right)\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} + \left(\frac{3{v_0}^2p^2}{{t_1}^2} - 1\right)\Delta''\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} + \Delta\begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix};$$

where $\pi = p_x + ip_y, p$ is the electron momentum, 2Δ is the electron energy difference between the top and bottom layer due to the vertical electrical field $v_i \equiv \left(\frac{\sqrt{3}}{2}\right)at_i/\hbar$, a is the carbon-carbon lattice constant ~2.46 Å, $\Delta' \approx 0.0122$ eV, $\Delta'' \approx -0.0095$ eV, and $t_0, t_1, t_2, t_{3,} t_4$ are tight binding parameters in ABC TLG obtained from LDA *ab initio* calculations with values of 2.62 eV, 0.358 eV, -0.0083 eV, 0.293 eV and 0.144 eV, respectively. We consider that the encapsulated ABC TLG forms a

near-zero-twisting moiré superlattice with the top hBN film. The TLG/hBN interaction in the K valley can be approximated by a potential of the form $V_M^A(\boldsymbol{r}) = 2\,C_A\,Re\left[e^{i\varphi_A} f(\boldsymbol{r})\right]\begin{pmatrix}0 & 0\\ 0 & 1\end{pmatrix}$ acting at the low energy A site of the effective ABC trilayer in contact with the top hBN film, where $f(\boldsymbol{r}) = \sum_{j=1}^{6} e^{i\boldsymbol{q}_j \boldsymbol{r}}\,(1+(-1)^j)/2$ and $\boldsymbol{q}_j$ are the six reciprocal lattice vectors of the triangular moiré superlattice with $|\boldsymbol{q}_j| = \boldsymbol{q}_M \equiv \frac{4\pi}{\sqrt{3}L_M}$. The hBN layer periodically modulates the potential in the bottom layer carbon atom whose magnitude and phase parameters are $C_A = -14.88$ meV, $\varphi_A = 50.19^o$. We solve the Hamiltonian numerically by direct diagonalization with a momentum cutoff at 5 q_M.

The calculated electron and hole minibands are asymmetric for both positive and negative displacement field. We found that for interlayer potential difference 2Δ = -20 meV (D = -0.4V/nm), the bandwidth of the electron miniband is 27 meV, while the hole miniband bandwidth is ~15 meV. For 2Δ = 20 meV (D = 0.4V/nm), the bandwidth of the electron and hole miniband are similar, while the electron miniband has more overlap with other minibands. In both positive and negative displacement fields, the hole miniband has narrower bandwidth and less overlap with other minibands than the electron miniband. The configuration that gives rise to the narrowest hole minibands experimentally exhibits the strongest Mott insulator behavior at ¼ and ½ filling. It is also the configuration that shows superconductivity behavior at low temperature close to the ¼ filling.

The asymmetry of positive and negative displacement field can be clearly identified from Fig. S1. In the negative displacement field, as shown in Fig. S1a, the hole miniband shows narrower bandwidth (11.7 meV) and is totally isolated from other minibands compared to the case in Fig. S1b which corresponds to the positive displacement field (bandwidth is 16.2 meV with overlap to the remote band).



Extended Data Figure 7 Calculated band structure of TLG/hBN with negative (**a**) and positive (**b**) displacement fields. The solid and dashed lines correspond to band structure at K and K' valley, respectively. The energy difference between top and bottom layer graphene $2\Delta = 20$ meV corresponds to the displacement field $D = 0.4$ V/nm.