Supplementary information

Coupled ferroelectricity and correlated states in a twisted quadrilayer MoS₂

moiré superlattice

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Figure S1. Scanning rate dependence of resistance hysteresis. Scan displacement field back and forth at 20.2 K. Interval of every two points measured is 0.001 s (a) and 0.01 s (b). The measurement time required in (b) is tenfold longer than the time in (a). But the hysteresis window is near same considering a natural decline of build-in electrical field.



Figure S2. Scanning direction dependence of resistance hysteresis. (a) Two lines are almost overlapped without hysteresis when scanning from -0.15 V/nm to 0.15 V/nm twice at 22.2 K. (b) Two lines show an obvious hysteresis window when scanning firstly from about 0.175 V/nm to -0.125 V/nm and then from about -0.125 V/nm to 0.175 V/nm at 22.2 K.



Figure S3. The resistance hysteresis against the scanning directions for v = 3 at 14 K. For each direction, we repeated the measurements four times. The resistance lines are almost overlapped without hysteresis when scanned in the same direction. By contrast, the resistance lines show an obvious hysteresis window when scanned along different directions.



Figure S4. Doping dependent hysteresis at different filling factors. There is no obvious hysteresis when scanning carrier density back and forth at a constant displacement field.



Figure S5. Ferroelectric hysteresis at various filling factors. Universal ferroelectric hysteresis lines are seen at various filling factors in (a)-(f).



Figure S6. Temperature dependence of ferroelectric polarization at v = 1.2. The red solid line represents the exponential decay fit $\rho \propto e^{-\alpha/T}$.



Figure S7. The absence of ferroelectric polarization in 57. 5° twisted bilayer MoS₂. The measured temperature is 12.5 K. Although correlated states at moiré band filling factors v = 1, 2, 3 can be clearly observed in 57. 5° twisted bilayer MoS₂ moiré superlattice, the resistance lines are almost overlapped without hysteresis when scanned along different directions. This strongly indicates the absence of ferroelectric polarization in 57. 5° twisted bilayer MoS₂.