Diodic transport response and the loop current state in twisted trilayer graphene

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The discovery of magic-angle graphene moiré systems has unlocked a wide variety of intriguing emergent phenomena, such as superconductivity and orbital ferromagnetism. In this work, we report the discovery of a new type of electronic order in twisted trilayer graphene that breaks both parity and time-reversal symmetry. This is identified based on highly nonreciprocal, diode-like current-voltage characteristics throughout the moiré flatband, which exhibits a one-fold or three-fold symmetric angular dependence as a function of the azimuth direction of current flow. By analyzing the relationship between the nonreciprocal response and the orbital ferromagnetism, we show that this parity-breaking order can be described as a valley-polarized loop current state, which is highly tunable with magnetic field, current flow, and field-effect doping. Our findings point towards the universal presence of valley-polarized isospin order and rotational symmetry breaking across the moiré flatband, with important implications for understanding intertwined and competing orders, such as ferromagnetism, nematicity, and superconductivity, in graphene-based moiré systems.

Owing to the nearly dispersionless band structures, graphene-based moiré systems establish an exciting material platform to study strongly correlated 2D electrons. A series of previous observations have shown that the dominating influence of electron interaction drives a cascade of isospin transitions and Fermi surface reconstructions [1–7]. The combination of isospin symmetry breaking and strong correlation gives rise to an intricate landscape of emergent quantum phases, such as superconductivity and orbital ferromagnetism [8–10]. The subtle interplay between the flat moiré band and strong Coulomb interaction provides an ideal venue for exploring novel nematic phases [11–19]. Indeed, experimental evidence of even-parity electron nematic has been reported in the superconducting and normal phases of magic-angle graphene bilayer and trilayer, inferred from the twofold symmetry of the critical current and resistivity measured under an in-plane magnetic field [20–23]. A recent experiment reported a prominent zero-field superconducting diode effect in twisted trilayer graphene, revealing a new type of symmetry breaking order [24]. Since the presence of either time-reversal $T$, and two-fold rotational symmetry $C_2$, implies identical critical current for forward and reverse DC current bias, $I^+ = I^-$, the observation of direction-dependence in the critical current points towards spontaneous breaking of both $C_2$ and $T$ in the superconducting state [25–28]. The symmetry breaking observed in the superconducting state also motivates our search for a parity-breaking electronic order in metallic states of graphene-based moiré systems.

In a two-dimensional electron systems, electron nematic order may occur through the Pomeranchuk instability of the Fermi surface. In the simplest form, a Pomeranchuk instability induces a quadrupolar distortion in a circular Fermi surface (panel i in Fig. 1a), giving rise to a nematic Fermi liquid with resistivity anisotropy (panel ii in Fig. 1a) [29]. This nematic Fermi liquid, which preserves two-fold rotational symmetry $C_2$, has been studied extensively in strongly correlated materials, such as ruthenate and cuprate materials [29–31]. Beyond the two-fold symmetric nematic order, more exotic Pomeranchuk phases have been discussed in previous theoretical proposals [19, 32–34]. For example, charge Pomeranchuk instabilities in odd angular momentum ($\ell$) channels break both parity and time-reversal symmetry and are expected to exhibit orbital loop current orders [32, 33]. As shown in panel iii in Fig. 1a, the $\ell = 3$ state breaks two-fold rotational symmetry $C_2$ but remains three-fold symmetric. Further breaking the three-fold symmetry of the $\ell = 3$ order gives rise to a one-fold symmetric state (panel iv in Fig. 1a). Despite great interest, it has been unclear how loop current orders can be unambiguously detected in transport experiments.

In this work, we report direct identification of a new electronic order in twisted trilayer graphene, which simultaneously breaks parity and time-reversal symmetry, using angle-resolved transport measurement (ARTM). This measurement scheme is made possible by the “sunflower” sample geometry, as shown in Fig. 1b. The “sunflower” pattern has eight independent electrical contacts, allowing us to vary the azimuth direction of current flow with an increment of $\phi = 22.5^\circ$. By measuring the voltage drop across two contacts that are either parallel or per-
FIG. 1. Diodic transport response in magic-angle twisted trilayer graphene. (a) Schematic diagram of Fermi surface contours corresponding to different underlying symmetries: (i) an isotropic Fermi liquid with a circular Fermi surface, which preserves both in-plane rotational and time-reversal symmetries; (ii) a nematic Fermi liquid with quadrupole distortion, which breaks three-fold rotational symmetry $C_3$ but is invariant under two-fold rotation; (iii-iv) nonreciprocal states resulting from more exotic Pomeranchuk instabilities which breaks two-fold rotational symmetry $C_2$. The state in (iii) preserves three-fold rotational symmetry $C_3$, whereas both two-fold and three-fold symmetry are broken in (iv). (b) Schematic of transport measurement in the “sunflower” geometry. This configuration measures the transport response parallel and perpendicular to the direction of current flow. The diameter of the circular portion is 2$\mu$m, more than 200 times the moiré length. (c-d) Angular-resolved measurement of (c) longitudinal resistance and (d) transverse resistance. The right panels show the same result in polar-coordinate plots. Azimuth angle $\phi$ defines the direction of DC current flow. (c) and (d) are measured with DC current flowing along azimuth angle of $\phi = 35^\circ$. The state in (iii) preserves three-fold rotational symmetry $C_3$, whereas both two-fold and three-fold symmetry are broken in (iv). (f) The nonlinear component of the IV curve, which is defined as the difference between the measured voltage and the linear response $V_{DC} - I_{DC}R_0$. $R_0$ is sample resistance measured at small current bias. The solid line is a quadratic dependence of $I_{DC}$. The nonlinear component shows the same sign independent of current flow direction. (g) Angular dependence of nonreciprocity $\eta$, defined based on Eq. 2. All measurements are performed at $\nu = 0.2$, $B = 0$ and $T = 20$ mK.

pendicular to the current flow direction, as shown in Fig. 1b, we can extract a pair of quantities, $R_{\parallel}$ and $R_{\perp}$, which are directly comparable to the longitudinal and transverse resistance of the sample [35]. Here $R_{\parallel}$ and $R_{\perp}$ are operationally defined as $\Delta V_{\parallel}/I$ and $\Delta V_{\perp}/I$ (Fig. 1b). Near the charge neutrality point (CNP) at $\nu = 0.3$, both $R_{\parallel}$ and $R_{\perp}$ exhibit a two-fold oscillation with a period of 180$^\circ$ (Fig. 1c-d). The maximum values of $R_{\parallel}$ and $R_{\perp}$ are offset by $\phi = 45^\circ$. Combined, the angular dependence in Fig. 1c-d points towards an orthorhombic anisotropy that preserves two-fold rotational symmetry $C_2$ [23, 30, 31, 35].

In addition to the orthorhombic anisotropy, the transport response also exhibits nonreciprocity, which is evidenced by the nonlinear current-voltage (IV) characteristics, as shown in Fig. 1e. Fig. 1f shows the nonlinear component of the IV curve, which is obtained by subtracting the linear component of transport response. The resulting nonlinear response, $\Delta V_{\parallel} - R_0 I_{DC}$, exhibits a quadratic dependence on $I_{DC}$ (Fig. 1f), which provides a strong indication for a highly nonreciprocal transport behavior. We define the angular dependence of nonreciprocity based on the difference in the transport response between forward and reversed current flowing in the azimuth direction $\phi$.

$$\eta(I_{DC}, \phi) = R_{\parallel}(+I_{DC}, \phi) - R_{\parallel}(-I_{DC}, \phi).$$

(1)

Here $+I_{DC}$ and $-I_{DC}$ corresponds to forward and reverse current bias. Interestingly, the nonreciprocal response exhibits strong dependence on the azimuth direction of current flow. Unlike $R_{\parallel}$ and $R_{\perp}$, the angular dependence of $\eta$ is predominantly one-fold symmetric, as shown in Fig. 1g. The red solid line in the polar-coordinate plot denotes the polar-axis of the one-fold symmetric response, where $\eta$ is maximized.
FIG. 2. Second-harmonic nonlinear transport response at $B = 0$. (a-b) Second-harmonic nonlinear response ($\Delta V_i^{2\omega}$ and $\Delta V_\perp^{2\omega}$) as a function of $I_{AC}$, measured along azimuth angle $\phi = 112^\circ$ degree (blue) and $\phi = 292^\circ$ degree (red). Despite that the measurement setup respects chirality, the signal pairs measured are of opposite sign, each following a quadratic curve. (c-f) Angle dependence measurement of $\Delta V_i^{2\omega}$ and $\eta$ at $\nu = -1.85$, $\Delta V_{\perp}^{2\omega}$ and $\eta$ at $\nu = 1.45$. The remarkable match in the angle dependence between $\Delta V_{\parallel}^{2\omega}$ and $\eta$ at difference densities illustrates their correspondence relation. $\Delta V_{\parallel}^{2\omega}$ are measured at $I_{AC} = 100$ nA, $\eta$ are measured at $I_{DC} = 100$ nA. All measurements are performed at $B = 0$ and $T = 20$ mK.

The nonreciprocity is also manifested as an AC transport response at the second-harmonic frequency [36–40]. An AC current bias is applied symmetrically across the “sunflower” sample at a frequency of 13 Hz (see Fig. S6i), while $\Delta V_{\parallel}^{2\omega}$ and $\Delta V_{\perp}^{2\omega}$ are measured at the second-harmonic frequency of 26 Hz. Fig. 2a-b plots the second-harmonic response parallel and perpendicular to the current flow direction, $\Delta V_{\parallel}^{2\omega}$ and $\Delta V_{\perp}^{2\omega}$, which exhibit a quadratic behavior as a function of AC current bias. Both $\Delta V_{\parallel}^{2\omega}$ and $\Delta V_{\perp}^{2\omega}$ switch sign when the current direction and the voltage probes are reversed simultaneously, which is a hallmark of nonreciprocal transport response. This sign-reversal suggests that the potential influence of heating and contact resistance are of secondary importance at best. At the same time, the angular dependence of $\Delta V_{\parallel}^{2\omega}$ shows excellent agreement with that of $\eta$ (Fig. 2c-f). Since a parity-breaking electronic order can be captured by either one-fold and three-fold symmetric angular dependence, we will utilize a linear combination of these two to describe the angular-dependence of the nonreciprocal response,

$$\eta(\phi) = \eta_1 \cos(\phi - \beta_1) + \eta_3 \cos(3(\phi - \beta_3))$$

$$\Delta V_{\parallel}^{2\omega}(\phi) = V_1 \cos(\phi - \beta_1) + V_3 \cos(3(\phi - \beta_3)).$$

Here $\eta_1$ and $V_1$ ($\eta_3$ and $V_3$) define the oscillation amplitude for the one-fold (three-fold) symmetric component. At $\nu = -1.85$ and $+1.45$, the angular dependence is predominantly one-fold symmetry. For the one-fold symmetric response, we define a nonreciprocal axis that points along the azimuth direction of current flow, $\phi = \beta_1$, where $\eta$ and $\Delta V_{\parallel}^{2\omega}$ are maximized. While a three-fold symmetric response does not have a unique polar-axis, we define three nonreciprocal axes pointing along azimuth directions where the nonlinear response is maximized, $\phi = \beta_3$. Along these nonreciprocal axes, the behavior of $R_{\parallel}$ resembles a diode. Since the second-harmonic nonlinear response is observed in both parallel and perpendicular channels, we will investigate the nature of the parity-breaking electronic order by examining the non-linear response in the perpendicular channel.

Most remarkably, a nonreciprocal response is present throughout the moiré flatband. As shown in Fig. 3a, the nonreciprocal response exhibits a large three-fold symmetric component near integer moiré band filling (top row of Fig. 3a), where Hall density measurement indicates a small Fermi surface (Fig. 3b). The density range of these predominantly three-fold response is marked by the blue shaded stripes in Fig. 3b-c.
FIG. 3. The density dependence of the diodic response. (a) Polar-coordinate plots of angular dependence of linear transport response $R_\parallel$ (top row) and second-harmonic nonlinear response $\Delta V_{2\omega}^T$ ($R_\parallel$ and ) (bottom row) measured at different moiré band fillings. The angular dependence of $\Delta V_{2\omega}^T$ and $R_\parallel$ are predominantly six-fold and three-fold symmetric. (b) Hall density $n_{\text{Hall}}$ and (c) oscillation amplitude of the angular dependence of $\Delta V_{2\omega}^T$, defined as $V_1 + V_3$ according to Eq. 2, as a function of moiré filling $\nu$. Vertical blue stripes mark the density regime where transport response exhibits a large $C_3$ breaking component. (d) angular dependence of $R_\parallel$ (top row) and $\Delta V_{2\omega}^T$ (bottom row), which display prominent $C_3$ breaking component. transport responses in these regimes exhibits a six-fold symmetric angular dependence, which are best fit using an angular dependence with a period of $\phi = 60^\circ$. Tuning moiré band filling away from integer values increases the size of the Fermi surface, stabilizing a diodic response with one-fold symmetric angular dependence (top row of Fig. 3d), which coincides with a prominent orthorhombic anisotropy (bottom row of Fig. 3d). Combined, linear and nonreciprocal transport responses suggest that $C_3$ breaking Pomeranchuk instability is favorable by a large Fermi surface. This points towards a close connection between the cascade of isospin transitions and the nonreciprocal response. Such connection is further confirmed by examining the strength of the nonreciprocal response, which is defined by the amplitude of the angle-dependent oscillation $V_1 + V_3$. A small amplitude is observed in the density range of $-2 < \nu < +1$, where the underlying Fermi surface is expected to be isospin-unequally polarized (see Fig. S1). The emergence of isospin order at $\nu = +1$ and $\nu = \pm 2$ coincides with a substantial increase in the diodic response. An exception is observed in a small density range near the charge neutrality point (CNP), where large nonreciprocal response is observed. This may be the result of additional flavor polarization in the spin, valley and sublattice degrees of freedom.

Notably, the nonreciprocal axes of the three-fold symmetric nonlinear response are predominantly aligned along characteristic angles of the six-fold angular dependence of the linear response, marked with green solid lines in Fig. 3a. Both the three-fold nonlinear and six-fold linear response display a lack of dependence on moiré band filling. This suggests that $C_3$ preserving transport responses, both linear and nonlinear, are linked with the angular dependence of the underlying Fermi surface.
This provides a strong indication that the three-fold symmetric nonreciprocal response originates from a valley-polarized isospin order. On the other hand, the non-reciprocal polar axis of the one-fold symmetric nonlinear response, $\beta_1$, along with the director axis of the orthorhombic anisotropy, are highly dependent on moiré band filling. The behavior of both $\beta_1$ and $\beta_3$ can be distinguished with previous observations of nonlinear Hall effect in few-layer WTe$_2$ [37, 38], where maximum nonlinear response is realized when the current flow is aligned perpendicular to the mirror axis of the crystal [37, 38].

Before proceeding further, it is instructive to consider and compare the symmetry requirements for the observed diodic response and for valley isospin polarization. Valley polarization spontaneously breaks both $C_2$ and $T$, but preserves their product $C_2 T$. A diodic response requires $C_2$ symmetry breaking and could arise from three possible symmetry breaking orders (see detailed discussion in SI): (i) $C_2$ is broken and $T$ is preserved. This scenario applies to the recently observed second-order Hall response in various systems where the crystal lattice breaks $C_2$. (ii) $C_2$ and $T$ are broken but their product $C_2 T$ is preserved, as realized by the presence of valley polarization; (iii) $C_2$, $T$ and $C_2 T$ are all broken. This scenario can arise from valley polarization in graphene moiré superlattices with sublattice symmetry breaking, which could be induced by the proximity with hBN or WSe$_2$ substrates that break $C_2$ symmetry [9, 10, 41].

To provide further evidence attributing diodic response to valley polarization, we measure the Hall resistance $\bar{R}^{1\omega}_y$ (top panels) and the second-harmonic nonlinear response $\bar{R}^{2\omega}_y$ (bottom panels) as a function of an out-of-plane magnetic field that is swept back and forth between $B = \pm 30$ mT, shown in Fig. 4a-f plots. $\bar{R}^{1\omega}_y$ is obtained by subtracting the $B$-dependent background of the Hall resistance $R_{xy}$ measured at the first-harmonic frequency (see Fig. S10). On the other hand, $\bar{R}^{2\omega}_y$ is defined as $\Delta V^{2\omega} / I_{AC}$, which is measured at the second-harmonic frequency $2\omega$. The $B$-dependence of $\bar{R}^{1\omega}_y$ reveals a series of hysteretic transitions near integer filling of the electron moiré band at $\nu = +1$, $+2$ and $+3$ (Fig. 4a-c). The observation of hysteresis loops reveals orbital...
ferromagnetism, which arises from the combination of a valley-polarized isospin order and sublattice symmetry breaking [9, 10, 41]. Interestingly, the second-harmonic response $\Delta V_{2\omega}$ near $\nu = +1$ also exhibits discontinuous jumps that coincide with the reversal in the underlying valley polarization. Fig. 4g plots the magnetic hysteresis loop in Hall resistance measured at different temperature. With decreasing temperature, the size of the hysteresis loop resulting from spontaneous valley polarization, $\Delta R_{\text{xy}}$, exhibits a sharp onset near $T = 2.5$ K (Fig. 4h). The second-harmonic nonlinear response $\Delta V_{2\omega}$ exhibits a similar onset that coincides in temperature (Fig. 4i). These observations strongly suggest that the dionic response arises from the valley-polarized isospin order, which breaks $C_2$ and $T$ symmetries.

Importantly, valley polarization preserves $C_2T$ and therefore cannot by itself generate orbital ferromagnetism or anomalous Hall effect, which requires breaking both $T$ and $C_2T$ symmetries. On the other hand, our symmetry analysis above shows that valley polarization alone should give rise to dionic response, without the need of additional $C_2T$ symmetry breaking. Indeed, on the hole-doping side of the CNP, anomalous Hall effect is absent, but strong dionic response is observed (Fig. 2f). The absence of magnetic hysteresis $R_{\text{xy}}$ or $\Delta V_{2\omega}$ near $\nu = -1$, $-2$ and $-3$ (Fig. 4d-f), and its presence on the electron doping side, can be understood by considering the influence of moiré bandwidth and Coulomb interaction strength on $C_2T$ symmetry breaking. It has been recognized that correlation strength is weaker in the hole-doping band, due to its larger moiré bandwidth. In the scenario where $C_2T$ breaking arises from Coulomb-driven sublattice polarization, it is plausible that $C_2T$ breaking is absent for the hole-doping band. Alternatively, $C_2T$ breaking could result from the proximity-induced spin-orbit coupling (SOC) [41, 42], which is inversely proportional to the band width. As such, a more dispersive band effectively suppresses the influence of SOC [42], which accounts for the weakened $C_2T$ breaking on the hole-doping side. That a prominent dionic response is observed in the hole-doping band without magnetic hysteresis reveals an electronic order that breaks $C_2$ and $T$ but preserves their product, $C_2T$. This is in excellent agreement with valley-polarized isospin order without additional $C_2T$ symmetry breaking.

Going beyond symmetry arguments, we now discuss the microscopic origin of dionic response in the valley polarized state. Notably, the moiré band structure of each valley is asymmetric with respect to the center of the mini-Brillouin zone ($k = 0$): $\epsilon_v(k) \neq \epsilon_v(-k)$, where $v = \pm$ denotes the valley isospin. Recent theory [43] predicts that noncentrosymmetric energy dispersion can give rise to second-order (dionic) current-voltage relation: $j_{2\omega} = \sigma_{abc} E_v^a E_\omega^c$, where $\sigma_{abc}$ is obtained from Boltzman transport equation:

$$\sigma_{abc} = -e^3 \tau^2 \sum_k \int \partial_{k_a} \partial_{k_b} \partial_{k_c} \epsilon_v(k) f_v(k) \quad (3)$$

![FIG. 5. Current-driven transition in the loop current state. Schematic of (a) the three-fold symmetric and (b) predominantly one-fold symmetric loop current state in a graphene moiré superlattice. The black circles mark the positions of charge islands on AA sites. Red arrows denote current flow direction between adjacent AA sites. $C_3$ is preserved in (a) as current flows in all three directions exhibit equal magnitude, whereas $C_3$ is broken by a dominating current flow direction in (b). The inset of (a) and (b) are polar-coordinate plot of nonreciprocal response measured at $\nu = 0.3$ and different current bias (see Fig. S11). (c) Second-harmonic nonlinear response measured with current bias is swept back and forth. Current flow is applied along $\phi = 112^\circ$. A hysteresis loop is observed as the amplitude of the AC current is swept between 0 and 2$\mu$A.](image)
where \( f_v(k) = 1/(e^{\beta(\epsilon_v(k)-\mu)} + 1) \) is the equilibrium distribution function for a given valley. In the absence of interaction, moiré bands of the two valleys are related by either \( C_2 \) or \( T \) symmetry, which guarantees that \( \epsilon_+(k) = \epsilon_-(-k) \). As a result, the contributions to \( \sigma_{abc} \) from opposite valleys completely cancel. In the interaction-driven valley polarized state, however, \( C_2 \) and \( T \) symmetries are spontaneously broken and the degeneracy between the energy bands and distribution functions of the two valleys is lifted, thus leading to nonzero \( \sigma_{abc} \) with three-fold angular dependence.

In graphene systems, the valley polarized state with \( C_2T \) symmetry is an ordered state characterized by spontaneous loop currents [44]. Schematic illustrations of possible loop current pattern in twisted trilayer graphene are shown in Fig. 5a-b. Generally speaking, loop-current order cannot be detected by conventional transport probes. Its broken time reversal symmetry does not lead to anomalous Hall effect. Its broken parity cannot be revealed from resistivity in the linear transport channel, which necessarily has two-fold symmetry. Our findings show that angle-resolved second-order nonlinear transport responses, including diodic and nonlinear Hall effects, provide a powerful method capable of directly probe loop current order. Notably, the loop current state accounts for both the three-fold and one-fold symmetric patterns. The three-fold symmetric nonreciprocal response is naturally explained by a valley-polarized loop current state, where the magnitude of current flow is equal in three directions (Fig. 5a). On the other hand, the one-fold symmetric response corresponds to a loop current state with asymmetric current flows (Fig. 5b).

The one-fold symmetric loop current state, which breaks parity, time-reversal and three-fold rotational symmetry simultaneously, is highly tunable using a variety of experimental parameters. As shown in Fig. 5c, a one-fold symmetric angular dependence is observed at \( \nu = -2.2 \) with small current bias. At this density, sweeping the current bias back and forth induces a hysteresis loop in the nonlinear response \( \Delta V_{\perp}^L \) (Fig. 5c). The angular-dependence of \( \Delta V_{\perp}^L \) measured before and after the hysteretic transition exhibit a prominent rotation in the polar axis of the one-fold symmetric loop current state. Before the hysteretic transition, the polar axis is aligned along the azimuth angle of \( \beta = 100^\circ \), as shown in the upper inset of Fig. 5c. A large current induces a rotation in the polar-axis of around \( \Delta \beta = 120^\circ \) in the counter-clock-wise direction. This is evidenced by the angular-dependent measured with small current bias after the hysteretic transition loop (lower inset of Fig. 5c). Current and doping-induced hysteretic behaviors have been associated with the orbital ferromagnetic order in previous observations, which is primarily identified based on \( B \)-induced hysteresis in the anomalous Hall effect [9, 10, 41, 45, 46]. Unlike these previous observations, current and doping-induced hysteresis behavior is observed in the hole-doped moiré flatband in the absence of magnetic hysteresis (Fig. S8). This is consistent with a loop current state that preserves the \( C_2T \) symmetry.

As shown in Fig. 4b-i and Fig. S4, the loop current state exhibits a sharp onset near \( T = 3 \) K with decreasing temperature. This provides a strong indication that the valley-polarized isospin order is only stable at low temperature. It is worth noting that the valley-polarized loop current state exhibits a sharp onset temperature, whereas the temperature dependence of the even-parity orthorhombic anisotropy is more gradual (see Fig. S4 and Fig. S5). This discrepancy can be understood by considering the role of uniaxial strain in the underlying moiré superlattice. The gradual temperature-dependence in the orthorhombic anisotropy can be naturally explained by the strong coupling with lattice distortions. On the other hand, a valley-polarized loop current state does not couple to lattice distortion, owing to the presence of time-reversal symmetry breaking. As a result, Coulomb interaction is the likely origin of the loop current state. A loop current order in the metallic state also offers a natural explanation for the previously reported zero-field superconducting diode effect [24]. The universal presence of the valley-polarized loop current state also provides important symmetry constraints for theories aiming to model various emergent phenomena, such as the cascade of Dirac revival, correlation-driven insulators and superconductivity.

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SUPPLEMENTARY MATERIALS

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SUPPLEMENTARY TEXT

Isospin degeneracy

Fig. S1 shows a cascade of Fermi surface reconstructions at integer fillings of the moiré band. These reconstructions are evidenced by quantum oscillations emanating from integer fillings of the moiré bands (Fig. S1a–c), as well as resets in $n_{Hall}$ (Fig. S1d). The reconstruction gives rise to a series of energy gaps in the moiré band. The location of these energy gaps exhibits a slope of $±2$ in the $ν–B$ map, which are marked with solid lines in Fig. S1c.

The isospin degeneracy underlying the reconstructed Fermi surface can be determined based on the slope of the quantum oscillation in the fundamental sequence emanating from each integer moiré filling. The lowest order state of each fundamental sequence is marked by the solid red lines in Fig. S1c. The trajectory of each inexpressible state is described by a pair of quantum numbers $(t, s)$ from the Diophantine equation $ν = tφ/φ_0 + s$, where $t$ corresponds to the slope and $s$ the intercept in the $ν–B$ map. The isospin degeneracy of the Fermi surface equals the difference between slope of the Coulomb-driven energy gap in the moiré band, which is $±2$, and that of the lowest order quantum oscillation. For instance, the moiré energy gap at $ν = 2$ traces a slope of $t = 2$, whereas the fundamental sequence of quantum oscillation has a slope of $t = 2 + 2$. This points to a degeneracy of 2 for the reconstructed Fermi surface at half moiré filling. Similarly, the reconstructed Fermi surface at quarter and three-quarter fillings have 3 and 1 fold degeneracy. This hierarchy of isospin degeneracy is in excellent agreement with previous report in magic-angle twisted bilayer graphene [49, 50], suggesting that the underlying moiré band of twisted trilayer graphene that is directly comparable to the flatband physics studied in MATBG.

The slope of the Coulomb-driven energy gap, $t = ±2$ could result from two possible scenarios. First, the energy gap of the moiré flatband has Chern number zero, and the slope of $t = ±2$ derives from the $N = 0$ Landau level of the monolayer band [7, 51, 52]. However, this scenario is not consistent with the observed orbital ferromagnetism at $ν = 1, 2$ and 3 (Fig. 4). Another possibility is that the slope of $t = ±2$ results from the Chern number of the underlying orbital ferromagnetic state, whereas the monolayer band is spontaneously gapped out near integer fillings. To definitively distinguish between these two scenarios requires a thorough mapping of the energy gap as a function of $B$. This is beyond the scope of this report.

The Landau fan diagram in Fig. S1 provides an accurate identification for the twist angle at $θ = 1.44°$. While the twist angle is detuned from the magic angle of $≈ 1.55°$, the emergence of Landau fan at each integer filling, along with Hall density reset, exhibit the same characteristic as magic-angle twisted trilayer graphene in previous observations [7, 51, 52]. It has been proposed that the proximity effect between tTLG and a few-layer WSe$_2$ could potentially expand the flatband condition to a twist angle that is much smaller compared to the magic angle of tTLG [53]. A more definitive identification on the influence of the proximity effect requires an thorough analysis of many samples, which is beyond the scope of this work. Nevertheless, the transport response in Fig. S1 is consistent with the proposed scenario, where the flatband condition is modified by the proximity effect.

Cascade of isospin transitions

The $ν–T$ map of the moiré flatband is divided into different areas based on the underlying isospin polarization. The boundaries of different isospin polarizations are defined by peaks in longitudinal resistance, concomitant with reset in the Hall density [1, 5–7]. Fig. S3 shows the $ν–T$ map of $R_∥$ and Hall density $n_{Hall}$. Isospin transitions are marked by white circles in the top panel. The cascade of isospin transition is clearly detectable at $T > 30$ K. This onset of isospin polarization transitions provides a characteristic for the Coulomb energy scale, which is believed to be the main driver behind the observed cascade phenomenon.

The onset temperature of the valley-polarized loop current state

Fig. 4h–i shows that the onset temperature of the nonreciprocal response, indicative of the emergence of the valley-polarized isospin order, coincides with the formation of orbital ferromagnetic state at $T = 3$ K. By plotting the temperature dependence, Fig. S4 shows that sharp onsets of the nonreciprocal response $ΔV_{2ω}^∥$ at $T < 5$ K across the moiré flatband. This implies that valley polarization only occurs below 5 K, which is much lower compared to the onset temperature for the cascade of isospin transitions. This discrepancy has interesting implications on the nature of isospin transitions at high temperature.
FIG. S1. Magneto-transport measurement across the moiré flatband. (a) $\Delta V_\parallel /I$ and (b) $\Delta V_\perp /I$ across the moiré filling-magnet field ($\nu - B$) map. Incompressible states are manifested as minima in $\Delta V_\parallel /I$, concomitant with quantized plateau in $\Delta V_\perp /I$. (c) The most prominent incompressible states are marked with black dashed line in the schematic $\nu - B$ map, where each trajectory is described by a pair of quantum numbers $(t, s)$ from the Diophantine equation $\nu = t\phi /\phi_0 + s$. Here $\nu$ is the moiré filling factor at the incompressible state, whereas $t$ and $s$ describe the slope and intercept of each trajectory [47, 48]. (d) Hall density $n_{\text{Hall}}$ as a function of moiré filling measured at $B = 0.5$ T.

Comparing the nonlinear and linear transport response

As shown in Fig. S5, the orthorhombic anisotropy onsets at $T \sim 30$ K, which is in excellent agreement with the energy scale of Coulomb interaction in tTLG [6, 7]. In comparison, the onset temperature of the diodic response $\Delta V_\perp ^{2\omega}$ indicates a much lower energy scale for the loop current phase. The sharpness of nematic transition is consistent
FIG. S2. The amplitude of the one-fold and three-fold component. $V_{p+3}^{2\omega}$ (top panel), $V_1^{2\omega}$ (red trace in the bottom panel), and $V_3^{2\omega}$ (red trace in the bottom panel) as a function of moiré band filling.

FIG. S3. Cascade of isospin transitions. $\Delta V_1/I$ (top panel) and Hall density $n_{Hall}$ (bottom panel) measured with linear transport (small current bias of 5 nA) as a function of moiré filling and temperature. The peak position of $\Delta V_1/I$ marks the boundary between different isospin orders [6, 7], which are marked with white circles. The transition between different isospin orders coincide with resets in the Hall density. The cascade of isospin transitions are detectable at $T = 34$ K, much higher compared to the onset temperature of valley-polarized loop current state, which is $T < 5$ K.

with the fact that the influence of distortion in the moiré superlattice is of secondary importance. This is consistent with the density induced transition between $C_3$ preserving and $C_3$ breaking phases in Fig. 3. At the same time, the loop current state is expected to be insensitive to uniaxial strain, owing to the presence of time-reversal symmetry breaking. Taken together, our findings implies that the odd- and even-parity nematic state arise from different types of Pomeranchuk instabilities.
FIG. S4. The temperature-onset of the loop current state. The temperature dependence of $\Delta V_{2\omega}^2$ as a function of temperature measured at (a) $\nu = 1.2$, (a) $\nu = 1.8$, (c) $\nu = 2.6$, (d) $\nu = -2.1$, (e) $\nu = -3.4$, and (f) $\nu = -1.0$. In (a-b), the loop current phase exhibits one-fold symmetric $\Delta V_{2\omega}^2$. In (c-f), the loop current phase exhibits predominantly three-fold symmetric $\Delta V_{2\omega}^2$. The onset of $\Delta V_{2\omega}^2$ occurs at temperature below 5 K, regardless of the one-fold or three-fold angular dependence. $\Delta V_{2\omega}^2$ is the nonlinear response measured at the second-harmonic frequency with an AC current of $I_{AC} = 100$ nA.

FIG. S5. Comparing the onset of the loop current state with orthorhombic anisotropy. $\Delta V_{2\omega}^2$ (blue trace) and $\Delta R/R_0$ (red trace) as a function of temperature measured at (a) $\nu = -2.2$ and (b) $\nu = 0.3$. $\Delta R/R_0$ describes the strength of orthorhombic anisotropy, which is defined based on the angular dependence $R_\parallel(\phi) = R_0 + \Delta R \cos(2(\phi + \alpha))$. 
MATERIALS AND METHOD

Device Fabrication

The doubly encapsulated tTLG is assembled using the “cut-and-stack” technique. All components of the structure are assembled from top to bottom using the same poly(bisphenol A carbonate) (PC)/polydimethylsiloxane (PDMS) stamp mounted on a glass slide. The sequence of stacking is: graphite as top gate electrode, 24 nm thick hBN as top dielectric, bilayer WSe$_2$, tTLG, 24 nm thick hBN as bottom dielectric, bottom graphite as bottom gate electrode.

The entire structure is deposited onto a doped Si/SiO$_2$ substrate, as shown in Fig. S7a. Electrical contacts to tTLG are made by CHF$_3$/O$_2$ etching and deposition of the Cr/Au (2/100 nm) metal edge contacts. The sample is shaped into a sunflower geometry with an inner radius of 1.9 $\mu$m for the circular part of the sample. In this geometry, the electrical contacts are separated by an azimuth angle of 45°, allowing an increment in the azimuth angle that is 22.5° (Fig. S6).

Transport measurement

The carrier density in tTLG is tuned by applying a DC voltage bias to the bottom gate electrode. The electrical potential of the top gate electrode is held at zero. As a result, the tTLG sample experience a non-zero displacement field $D$ at large carrier density, which induces hybridization between the monolayer band and the moiré flatband. We note that the dependence of Hall density on moiré band filling is in excellent agreement with $D = 0$ behavior from previous observations. This indicates that the influence of $D$ on the moiré flatband is not substantial. This is further confirmed by the Landau fan diagram in Fig. S1, which is also consistent with the expected behavior at $D = 0$.

Transport measurement is performed in a BlueFors LD400 dilution refrigerator with a base temperature of 20 mK. Temperature is measured using a resistance thermometer located on the cold finger connecting the mixing chamber and the sample. An external multi-stage low-pass filter is installed on the mixing chamber of the dilution unit. The filter contains two filter banks, one with RC circuits and one with LC circuits. The radio frequency low-pass filter bank (RF) attenuates above 80 MHz, whereas the low frequency low-pass filter bank (RC) attenuates from 50 kHz. The filter is commercially available from QDevil.

The current-voltage characteristics is measured using two methods. In the DC measurements, we sweep the amplitude of the DC current with a small, fixed AC excitation of 5 nA at a frequency of 13 Hz. The differential voltage is measured using standard lock-in techniques with Stanford Research SR830 amplifier. In the AC measurements, we sweep the amplitude of the AC current at a frequency of 13 Hz. The nonlinear response is measured at the second harmonic frequency using Stanford Research SR830 amplifier. To avoid the potential influence of contact resistance, the AC current is applied symmetrically across the sample, as shown in Fig. S6i.

Transport response is measured across voltage leads that are parallel and perpendicular to the current flow direction. The setup for the parallel response, $\Delta V_{||}$, is shown in Fig. S6a-h. For current flowing in the azimuth angle $0 - 180^\circ_{\text{circ}}$, Fig. S6a-h display 8 measurement configurations with an increment of 22.5° in the azimuth direction of current flow. The voltage measurement in panel e-h is different by a geometric factor compared to that of panel a-d. This geometric factor is characterized in Ref. [35] and shown to be 1.09.
FIG. S6. **Measurement setup.** (a-h) Measurement setup with DC current bias. DC current bias is applied to different combination of electrical contacts so that DC current flows along different azimuth angles $\phi$. Transport response for each azimuth direction is extracted by measuring the voltage drop across two contacts that are parallel to the direction of the DC current flow, $\Delta V_\parallel$. Similarly, $\Delta V_\perp$ denotes the voltage drop across two contacts that are perpendicular to the current flow direction. $\Delta V_\parallel/I$ and $\Delta V_\perp/I$ are in excellent agreement with longitudinal and transverse resistance, as shown in Fig. S1. (i) In AC current measurement, AC current bias are applied to the same electrical contact as in panel (a-h). Here we use the configuration in panel (a) as an example. To do AC current measurement, two current bias with a phase difference of $\pi$ are applied simultaneously to the two current bias contacts. This ensures that the center of the sample has zero electric potential, thus eliminating potential influence of contact resistance and thermal-electric effects.

FIG. S7. **“sunflower” geometry.** (a) Schematic of the “sunflower” geometry sample. (b) Optical image of the tTLG sample patterned into the “sunflower” geometry. Carrier density is varied by tuning the DC voltage bias applied to the bottom graphite gate, which only covers the circular part of the “sunflower” sample. Electrical contacts between graphene and metal are outside of the graphite gate. Therefore, eight “patels” are fixed at a constant value of carrier density. As such, the dependence of linear and nonlinear transport responses on moiré band filling is decoupled from the potential influence of contact resistance. Detailed characterization of transport response across such sample geometry is discussed in Ref. [35]. (c) Schematic diagram of wiring used in the ARTM.
**FIG. S8.** **Doping-induced hysteresis in second-harmonic nonlinear response.** Second-harmonic nonlinear anomalous Hall resistance as a function of moiré filling at zero external magnetic field, while the gate is swept in different directions at 20mK (top panel) and 2K (bottom panel). The sweeping direction is labeled by the arrows in corresponding colors. Note that at 20mK the second-harmonic resistance exhibits prominent hysteresis near \( \nu = -2 \) and \(-3\), and the hysteresis quickly disappears as temperature is raised to 2K.

**FIG. S9.** **Angle dependence of \( \eta \) measured at different DC current bias.** Polar-coordinate plot of the angle dependence of \( \eta \), measured at \( B = 0 \), \( T = 20 \) mK and \( \nu = 0.2 \). With increasing DC current bias, a similar angular dependence that is predominantly one-fold symmetric is observed up to \( I_{DC} = 100 \) nA. The polar-axis of the one-fold symmetric response is aligned near \( \beta = 135^\circ \).
FIG. S10. **B-induced hysteresis loops.** $B$-dependence of Hall resistance $R_{xy}$ (top panel) and corrected Hall resistance $\bar{R}_{xy}$ (bottom panel) measured at different moiré band fillings. $\bar{R}_{xy}$ is obtained by subtracting the $B$-dependent background from $R_{xy}$. 
FIG. S11. Current-driven one-fold to three-fold transition in both AC and DC measurements. (a) $\Delta V_{2\omega}^\perp$ as a function of AC current bias $I_{AC}$ measured at $T = 20$ mK and $\nu = -0.3$. A sign reversal occurs at $I_{AC} = 150$ nA. The current dependence of $\Delta V_{2\omega}^\perp$ measured near the CNP, which exhibits a sign-reversal in $\Delta V_{2\omega}^\perp$ at $I_{AC} = 150$ nA. This sign-reversal corresponds to a current-dependence in the angular-dependence of $\Delta V_{2\omega}^\perp$; a three-fold symmetric angular dependence is stable at large current, whereas a one-fold symmetric response is stable at small current bias. This is evidenced by the angle dependence of (b) $\eta$ and (c) $\Delta V_{2\omega}^\perp$ measured at different DC and AC current bias, respectively. The polar-axis of the nonreciprocal response is marked with the red solid line, which is aligned along the azimuth direction of maximum nonreciprocal response. The solid black line is the best fit to the angular dependence using Eq. 2. The green dashed circle denotes zero in $\eta$ and $\Delta V_{2\omega}^\perp$. 
FIG. S12.  

**B-induced hysteresis loops.**  
(a-b) Size of the $B$-induced hysteresis loop as a function of (a) moiré band filling and (b) temperature $T$. Robust magnetic hysteresis is observed near $\nu = +1$ and $+2$. At $\nu = 0.9$, the hysteresis is shown to onset at $T = 2.5$ K. (c) Transverse resistance measured at the first-harmonic frequency $R_{\perp 1}^\nu$ and (d) second-harmonic nonlinear response $R_{\perp 2}^\nu$ measured with $B$-field sweeping back and forth. The measurement is performed at $\nu = 1.17$. While Hysteretic transitions in $R_{\perp 1}^\nu$ and $R_{\perp 2}^\nu$ are linked to the same valley-polarization transition and generally occur at similar density and temperature range, they do not always occur at the same time. This discrepancy can be understood by considering the angle dependence of the second harmonic nonlinear response, which is shown to be one-fold symmetric in the density range of the anomalous Hall effect. A hysteresis loop in the second harmonic nonlinear response could arise from a $B$-induced rotation in the polar axis. This is distinct compared to a sign reversal, which corresponds to a rotation of $180^\circ$. In this work, we showed that the polar axis of the loop current state is tunable with varying $B$ near integer filling in the electron-doped moiré band. While $B$-induced rotation is not observed in the hole-doping band, Fig. 5c and Fig. S8 show that the polar axis orientation is tunable with current and field-effect doping.

FIG. S13.  

**Six-fold symmetric angular dependence of the linear transport response $R_\parallel$, $R_\perp$** as a function of azimuth angle $\phi$ measured at $\nu = 2.2$ and (a) $T = 0.6$ K, (b) $T = 34$ K. The red dashed line is the best fit to the angular dependence. At low temperature, the angular dependence shows a period of $60^\circ$. At high temperature, the angular dependence is predominantly isotropic. The emergence of the six-fold pattern at low temperature is consistent with the onset temperature of the three-fold symmetric loop current state.
FIG. S14. **Scheme for measuring the angular dependence of nonreciprocity.** In order to make sure the polar axis of the one-fold symmetric nonreciprocal response does not rotate during an angular dependence measurement, we adopt the following measurement scheme. 8 directions of current flow between $0 < \phi < 180^\circ$ are marked with letter $a$ to $h$. The next 8 with reversed current flow are marked with $a'$ to $h'$. According to Fig. 2a-b, switching the current flow direction always gives rise to a sign reversal in the nonreciprocal response. This provides a nice way to verify that the polar axis remains the same. We perform the angular measurement in the following sequence: $a, b, c, a', d, e, f, b'$ ... if $a'$ shows sign reversal compared to $a$, it confirms that the polar-axis has not rotated and the measurement continues.