Nonlinear anomalous Hall effects probe topological phase-transitions in twisted double bilayer graphene

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Nonlinear anomalous Hall effect is the Berry curvature dipole induced second-order Hall voltage or temperature difference in response to a longitudinal electric field or temperature gradient. These are the prominent Hall responses in time reversal symmetric systems. Here, we investigate the family of second-order nonlinear anomalous Hall effects, the electrical, thermoelectric and thermal nonlinear Hall effects in the moiré system of twisted double bilayer graphene. We demonstrate that the nonlinear anomalous Hall signals can be used to probe the topological phase-transitions in moiré systems, induced by a perpendicular electric field. Specifically, we show that the whole family of nonlinear anomalous Hall responses undergo a sign reversal across a topological phase-transition.

I. INTRODUCTION

The experimental realization of novel quantum phases in small angle twisted bilayer graphene (TDBG)1–3 has propelled moiré systems to the forefront of quantum material research. For very small twist angles, the enhanced impact of electron-electron interactions due to narrow bandwidth enables a rich phase diagram of correlated phases in TDBG2–5. In addition to TDBG, other stacked moiré systems such as twisted double bilayer graphene (TDBG)6–15, twisted trilayer graphene16–19, and marginally twisted transition metal dichalcogenides20–22 also show fascinating strong correlation effects. Moiré systems also offer a promising platform for exploring the valley and topological physics. In addition to large Berry curvature hotspots, valley-Chern number based topological phase transitions23 have also been demonstrated in moiré systems such as TBG24, TDBG10,15,25–29, multi-layered twisted systems30,31, and twisted transition metal dichalcogenides32,33.

The non-interacting topological aspects of moiré systems are relatively less explored. However, the presence of time reversal symmetry (TRS) suppresses the Berry curvature related linear response phenomena, such as the anomalous Hall effect34,35, in these materials. However, the nonlinear anomalous (NLA) Hall effect induced by the Berry curvature dipole (BCD)36–41 can probe the Berry curvature in systems with TRS [Fig. 1(a)]. The NLA Hall effect requires the breaking of space inversion symmetry (SIS), and it appears in crystals with reduced symmetry32,33,42–51. In addition to the NLA Hall charge response to the electric field, there are also thermoelectric and thermal NLA responses that have a similar origin. The former is known as the NLA Nernst effect52,53, and the latter is termed as the NLA thermal Hall (or Righi-Leduc) effect54. The family of the NLA charge and heat responses originate from the finite Berry curvature of the electronic bands, and the asymmetry of the electronic dispersion.

In this paper, we demonstrate that the electrical, thermoelectric and thermal NLA Hall responses in strained TDBG can probe the topological phase transition, as they undergo a sign reversal across the transition. We present a systematic study of the family of NLA Hall effects: the electrical NLA Hall, the thermoelectric NLA Nernst [Fig. 1(b)] and the NLA thermal Hall [Fig. 1(c)] effect in strained TDBG. In particular, we highlight the impact of the band structure tunability of TDBG through the perpendicular electric field on the whole family of NLA Hall effects. While the electrical NLA Hall effect has been studied earlier in other moiré sys-
tems\textsuperscript{32,33,48–50}, the thermoelectric and thermal NLA Hall effects have not been explored earlier in moiré systems. We connect the family of NLA Hall responses to the topological phase-transitions of the valley-Chern number\textsuperscript{32,46,55} in TDBG. In addition to tuning the electronic structure, the perpendicular electric field in TDBG also induces topological phase-transition of the valley-Chern type. We find that such transitions, accompanied by the band gap closing at a specific $k$ point, change the distribution of the Berry curvature hotspot in the vicinity of the band crossing point [Fig. 1(d)]. The change in the Berry curvature distribution results in sign change of all three NLA Hall responses across the phase transition. This establishes the NLA Hall responses as a unique probe of the band topology in moiré and other topological systems.

II. ELECTRONIC BAND STRUCTURE OF TDBG

TDBG is fabricated by placing two sheets of Bernal stacked (AB-stacked) bilayer graphene on top of each other and rotating them by an angle $\theta$ with respect to each other around an axis perpendicular to the plane. Accordingly, its Hamiltonian is constructed by combining the Hamiltonian of the individual bilayers along with a coupling between the two, known as moiré coupling. To obtain the electronic structure of TDBG, here we extend the low energy continuum approach of Bistritzer and MacDonald\textsuperscript{1}. We particularly focus on the perpendicular electric field tunability and strain induced symmetry reductions of the TDBG which play pivotal roles in generating NLA Hall effects in TDBG. We emphasize that a perpendicular electric field can be experimentally applied in a specially designed field-effect-transistor like device structure with dual gates\textsuperscript{13–15}. Strain, on the other hand, generally appears in 2D systems during fabrication on a substrate and is ubiquitous in almost every moiré platform\textsuperscript{56}.

With the Bernal stacked bilayer graphene Hamiltonian, TDBG can be constructed to be either of AB-AB or AB-BA type\textsuperscript{10,25–27}. Here, we consider the AB-AB configuration. However, we emphasize that the physics we highlight in this paper is independent of the stacking order. We assume that the bilayer at the top (layer-1) is rotated by an angle $-\theta/2$ and the bilayer at the bottom (layer-2) is rotated by an angle $\theta/2$. Such rotation of individual layers creates a moiré pattern with an effective twist angle $\theta$. A schematic of the moiré structure in the reciprocal space is presented in Fig. 2(a). The big hexagons (orange and cyan) represent the rotated BZ of the (top and bottom) bilayer graphene and the small hexagons represent the moiré BZ. The neighbouring Dirac points (vertices of the large hexagons) of the rotated bilayers are connected by $q_{tr}$ and $q_{tl}$. The details of the low energy model based band structure calculation of TDBG are summarized in Appendix A.

The band structure of TDBG for twist angle $\theta = 1.1^\circ$ is shown in Fig. 2(b). In the absence of a perpendicular electric field, the band structure consists of a pair of low energy flat bands separated from the higher energy bands. The higher energy bands are referred to as the moiré bands and the corresponding gap is known as the moiré gap. The broad features of the electronic spectrum is similar to that found in TBG\textsuperscript{1} and to other moiré systems\textsuperscript{32}. However, TDBG offers better tunability of its electronic properties, which are accessed by the external perpendicular electric field [onsite parameter $\Delta$ in Eq. (A3)], compared to TBG. We have included that in our calculations through the gap parameters as $\Delta'_p = -\Delta_p = 1/2 \Delta$ and $\Delta'_f = -\Delta_f = 1/2 \Delta$ which basically represents a constant gradient in the potential. With this parameter $\Delta$, we can induce metal-insulator transition\textsuperscript{57} with out changing doping, along with topological phase-transitions of the valley-Chern type\textsuperscript{23}. The topological phase transitions are associated with closure of the band gaps at a specific $k$-point and it plays a very fundamental role in NLA Hall effects.

We now focus on the effect of strain on the band struc-
ture of TDBG. Strain usually appears in 2D moiré systems when mounting on a substrate. In case of twisted moiré systems where two layers are put on top of each other and sandwiched between the substrates, strain may appear on both of the layers\cite{50,58}. However, for simplicity, in our calculation we assume strain (\(\mathcal{E}\)) acts only on a single (bottom) layer\cite{59}. In presence of strain, the primitive lattice vectors and hence the reciprocal lattice vectors get distorted. For a given strain matrix \(\mathcal{E}\), (which satisfies \(\mathcal{E}^T = \mathcal{E}\) with \(T\) denoting the transpose), the real space vectors distort as \(r \rightarrow (1 + \mathcal{E})r\) and the reciprocal vectors as \(k \rightarrow (1 - \mathcal{E}^T)k\). We obtain the strained moiré lattice vectors as \(G_{\text{nn}} = \beta_3^{(1)} - \beta_3^{(2)}\) where \(\beta\) represents the modified reciprocal lattice vectors due to combined effect of rotation and strain. More details of the impact of strain on the electronic properties of TDBG are summarized in Appendix B.

To calculate the strained band structure we consider the uni-axial strain of strength \(\mathcal{E}\) at an angle \(\phi\) relative to zigzag direction as\cite{40,50,58}

\[
\mathcal{E} = \varepsilon \left( -\cos^2 \phi + \nu \sin^2 \phi \frac{1}{1+\nu} \sin \phi \cos \phi \right).
\]  

Equation (1) represents the system when it is more stretched in one direction and less stretched in the perpendicular direction. With this strain matrix, the calculated electronic structure of TDBG for \(\varepsilon = 0.2\%\) strain (with \(\phi = 0\)) is shown in Fig. 2(b). We find that the strain can lift the degeneracy between the flat bands observed in the absence of a perpendicular electric field (\(\Delta = 0\)).

More importantly, even a small but finite strain significantly reduces the symmetry of the system. In Fig. 2(c) and (d), we highlight the breaking of the \(C_3\) rotational symmetry, one of the key symmetry of the hexagonal lattice structure, due to strain. We convey this by plotting the energy dispersion along three different \(C_3\)-symmetric paths. We show that in absence of strain [see Fig. 2(c)] the energy dispersion in the three marked paths lie on top of each other. In contrast, the presence of strain in the Fig. 2(d) lifts this degeneracy, clearly indicating the breakdown of the \(C_3\) symmetry.

### III. NONLINEAR ANOMALOUS HALL EFFECTS IN TDBG

Having explored the electronic band structure of TDBG, we now focus on the electric, thermoelectric and thermal NLA Hall effects\cite{38,39}. It has been recently discovered\cite{39} that SIS broken systems possess a second-order nonlinear Hall charge current, which is finite even in the presence of TRS. This has been termed as the NLA Hall effect. The corresponding conductivity \(\sigma_{\text{abc}}\) relating the NLA current to the in-plane electric field, \(\mathbf{E}\) (\(j^{(2)} = \sigma_{\text{abc}}F_b E_c\), with \(a\), \(b\) and \(c\) as spatial indices) is given by

\[
\sigma_{\text{abc}} = \epsilon_{\text{abcd}} \frac{e^3 \tau}{h^2} \Lambda^a_{\text{dc}}. 
\]  

Table I. The family of the nonlinear Hall conductivity (electric, thermoelectric and thermal) and the corresponding generalizations of the Berry curvature dipoles. The NLA Hall effect \(\sigma_{\text{abc}}\) is quantified by the BCD \(\Lambda^a_{\text{dc}}\); the NLA Nernst effect is quantified by \(A^a_{\text{dc}}\), and the NLA thermal Hall effect is quantified by \(A^a_{\text{dc}}\). Clearly, all three of these are Fermi surface effects.

Here, \(-e\) is the electronic charge, \(\epsilon_{\text{abcd}}\) is the antisymmetric Levi-civita tensor, \(\tau\) is the scattering time and \(\Lambda^a_{\text{dc}}\) is the BCD which is given by,

\[
\Lambda^a_{\text{dc}} = -\sum_n \int |dk| \frac{\partial f_0}{\partial k_c} \Omega^a_{\text{dc}}. 
\]  

In Eq. (3), \(f_0^a\) is the Fermi distribution function of the \(n\)-th band and \(|dk|/(2\pi)^D\) stands for \(dk/(2\pi)^D\), where \(D\) represents the spatial dimension of the system and \(\Omega^a_{\text{dc}}\) is the \(d\)-th component of Berry curvature vector. It is defined as \(\Omega^a_{\text{dc}} = \frac{1}{2} e_{\text{dc}} \Omega^n_{\text{ac}}\), where

\[
\Omega^n_{\text{ac}} = -2\text{Im} \sum_{m \neq n} \frac{\langle u_n | \partial_k \mathcal{H} | u_m \rangle \langle u_m | \partial_k \mathcal{H} | u_n \rangle}{(\epsilon_n - \epsilon_m)^2}. 
\]  

Here, \(u_m\) is the periodic part of the Bloch wave-function with \(\mathcal{H} | u_n \rangle = \epsilon_n | u_n \rangle\). The discovery of electrical NLA Hall effect was followed up by predictions where a longitudinal temperature gradient is shown to generate second-order NLA Hall charge current as well as NLA Hall heat current. The phenomena of temperature gradient induced NLA Hall charge current, \(j_{\text{a}}^{(2)} = \sigma_{\text{abc}}(\nabla_b T \nabla_c T)\), has been named as NLA Nernst effect\cite{52,60}. The NLA Nernst coefficient can be expressed as \(\sigma_{\text{abc}} = \epsilon_{\text{abcd}} \frac{e^3 \tau}{h^2} \Lambda^a_{\text{dc}}\), with \(\Lambda^a_{\text{dc}}\) being the thermoelectric counterpart of the BCD. Similarly, the NLA heat current generation due to temperature gradient \(j_{\text{Q,a}}^{(2)} = \bar{\sigma}_{\text{abc}}(\nabla_b T \nabla_c T)\) is called the NLA Right-Leduc effect\cite{54}. The corresponding NL conductivity can be expressed in terms of the thermal counterpart of the BCD \(\Lambda^a_{\text{dc}}\) as \(\bar{\sigma}_{\text{abc}} = \epsilon_{\text{abcd}} \frac{e^3 \tau}{h^2} \Lambda^a_{\text{dc}}\). The expressions of all the NLA Hall conductivities and the related generalization of the Berry curvature dipoles are summarized in Table I.

Similar to other second-order NL transport phenomena, the NLA Hall effects vanish in systems with the SIS. This can be seen from the expressions of Table I using the fact that in presence of SIS, both the energy and Berry curvature are even functions of the crystal momentum. Interestingly, the breaking of SIS is not sufficient
to generate the NL Hall responses and further reduction in the crystal symmetries is required\[^{36}\]. For a detailed crystal symmetry analysis for materials which can host a finite BCD, we refer the readers to Ref. \[^{36}\]. Furthermore, from the expressions of the NLA Hall effects given in Table I, the different NLA conductivity coefficients are expected to be related on the basis of the Sommerfeld expansion. In the low temperature limit, \(\mu \gg k_B T\), the thermoelectric counterpart (Nernst) satisfies the relation \(\sigma_{abc} (\mu) \propto \kappa_{abc} (\mu)\) while the NL thermal Hall coefficient satisfies \(\kappa_{abc} (\mu) = \frac{\partial \mu}{\partial \sigma_{abc}} (\mu)\).

In TDBG, the TRS is present and the SIS is broken. Broken SIS combined with the absence of even-fold rotational symmetries, guarantees the presence of a finite Berry curvature\[^{61}\]. Since TDBG is a 2D-system, the Berry curvature has only one component perpendicular to the 2D plane (chosen to be the \(z\)-component). The distribution of the Berry curvature in the \(K\)-valley, in absence and in presence of strain are shown in Fig. 3(a)-(d) for two values of the perpendicular electric field. We emphasize that the presence of TRS in TDBG guarantees that \(\Omega_{z,K}(k) = -\Omega_{z,K^{'}}(-k)\). The non-zero Berry curvature combines with the velocity distribution over the BZ to dictate the NL responses at the Fermi surface. Interestingly, we find that although each valley possesses large Berry curvature, the BCD and its thermoelectric and thermal counterparts are identically zero in each valley of pristine TDBG due to the presence of \(C_3\) symmetry. However, in any realistic system, strain breaks the \(C_3\) symmetry to generate a finite BCD and its thermoelectric and thermal counterparts. The \(K\)-valley distribution of the Berry curvature multiplied with velocity at the first conduction band has been highlighted in Fig. 3(a)-(d) both in absence and presence of \(C_3\) symmetry. The asymmetry of Fig. 3(b) and (d) promotes a net dipole within the moiré unit cell. Note that the presence of TRS implies \(v_{n,K}(k) = v_{n,K^{'}}(-k)\). Consequently, the Berry curvature dipoles of the two valleys are identical.

In 2D systems like the TDBG, where the Berry curvature has only one component, the Berry curvature dipoles \(\Lambda_{dc}^{\alpha/\beta}\) are pseudo-tensors. Consequently it can be represented by two components of a vector decided by the direction of the velocity (subscript \(c\)). In Fig. 3(e) we have plotted the Berry curvature dipoles \(\Lambda_{x/y}^{\alpha}\) and \(\Lambda_{x/y}^{\alpha}\) of TDBG as a function of energy in presence of 0.2% strain for perpendicular electric field \(\Delta = 11\) meV. This is supplemented by the energy dispersion weighted by the Berry curvature along some high-symmetry points in Fig. 3(f). In the BCD distributions along the energy axis, broadly four regions of non-zero BCD and its counterparts can be seen. Each of these regions are separated by the band gaps. Among them two regions (in the middle) can be attributed to the flat conduction and valence bands, while the other two (on top and bottom) originate from the higher moiré bands.

From the plots we find that the BCD in the flat bands is of the order of magnitude \(\sim 10\) Å. The thermoelectric and thermal counterparts of BCD have larger magnitudes \(\sim 50\) Å and \(\sim 75\) Å, respectively. As expected, the thermoelectric counterpart of the BCD follows the same trend as the BCD. However, the thermal counterpart provides more subtle feature that are dictated by the derivative with respect to energy. We note that, although the BCD and its counterparts as a whole concentrate near certain regions, the \(x\)-component and the \(y\)-component of the Berry curvature dipoles have different features.

To highlight impact of the perpendicular electric field \(\Delta\) on the nature of the BCD and its thermal counterparts, we have plotted them for two different values of \(\Delta\) in Fig. 4. Specifically, we have plotted \(x\)- and \(y\)-components of \(\Lambda_{x}^{\alpha}\) and \(\Lambda_{x}^{\alpha}\) on the conduction band side.
the valleys. Specifically, we define the valley-Chern number for calculated by integrating the Berry curvature near the val-
the whole Brillouin zone, the valley-Chern number is cal-
ber is calculated by integrating the Berry curvature over
insulators (TRS broken systems). While the Chern num-
ber analog of the Chern number usually defined in Chern
TDBG possesses valley-Chern number, which is the val-
ley analog of the Chern number in Chern insulators (TRS broken systems). We add
here that in presence of TRS in TDBG, the total Chern number obtained by adding the contribution of both the valleys is always zero.

Interestingly, the valley-Chern number in TDBG can be changed by applying a perpendicular electric field inducing a topological phase-transition of valley-Chern type. This is different from the familiar topological phase-transitions in Chern insulators and defined in systems with TRS where the Chern number vanishes. In Fig. 5(a)-(b), we present the variation of the valley-Chern number of the first conduction and first valence bands in the K-valley on changing the perpendicular electric field (Δ) and strain (ε). We find multiple phase transitions in TDBG on varying strain and the perpendicular electric field. This is consistent with earlier reports of the valley-Chern phase transitions in TDBG with perpendicular electric field in Refs. [26 and 27], in absence of strain. Here, we find that the strain impacts the phase-transition boundaries significantly and also induces new phases for higher values of the electric field strength [see green region in Fig. 5(b)]. We note that the valley-Chern phase (2,−2), representing the conduction and valence band valley-Chern numbers respectively, is the dominant phase in TDBG. In addition, there are many other phases with valley-Chern number ranging from −3 to 3 depending on the specific choice of the parameters.

Like the usual phase-transition in Chern insulators, the valley-Chern number changes in TDBG are also associated with band gap closing at a specific k point. To show this explicitly we have explored the evolution of the band movement in Fig. 5(c)-(e). Specifically, we have plotted the minimum of direct band-gaps as a function of Δ for the four lowest energy bands near the charge neutrality point (two flat bands along with the two moiré bands enveloping them), for strain ε = 0.2%. It is evident from Fig. 5(d) that the valley-phase transition for small perpendicular electric field (Δ ≈ 8 meV) in both the bands are associated with band gap closing between the pair of flat bands. Similarly, the phase-transitions of the conduction band for relatively higher value of perpendicular electric field (Δ ≈ 28 meV) can be attributed to the band gap closing between the first and second conduction band as highlighted in Fig. 5(e). The change in the valley Chern number of the valence flat band, for larger field strength, are associated with successive closing of band gaps between first and second valence bands, see Fig. 5(c).

A peculiarity of these topological phase transitions is that they are difficult to detect experimentally. This is because there is no net circulating edge state as the total Chern number, summed over both the two valleys, is zero. We show below that the NLA Hall measurements offer a unique probe to detect these valley-Chern phase transitions. At the heart of this idea is the fact that band geometric quantities dictate the band topology, and in this case of time reversal symmetric system they also manifest in the NLA Hall transport phenomena. This can be easily seen via the simple model calculation for gapped graphene presented in Appendix C and summarized in Fig. 6.

To highlight the connection between the valley-Chern phase transition and the NLA Hall responses in TDBG, we show the variation of the BCD (λx) with energy and

\begin{equation}
\sigma_{xy} = \frac{1}{2\pi} \int_{\text{valley}} dk \Omega_{\alpha}^n.
\end{equation}

Strictly speaking, the concept of valley-Chern number is an approximate idea that depends only on the integration near a specific valley and does not map on to the full BZ. However, since the continuum model of the moiré systems provides valley specific BZ and electronic structure, we can calculate the valley-Chern number in moiré systems just like the usual Chern number. We add here that in presence of TRS in TDBG, the total Chern number obtained by adding the contribution of both the valleys is always zero.
perpendicular electric field ($\Delta$) space for $\varepsilon = 0.2\%$ in Figs. 5(f) and 5(h). The corresponding thermal counterpart is shown in Figs. 5(g) and 5(i). The NL thermoelectric counterpart of the BCD is simply proportional to the BCD, and not presented separately. The key features in the $\Lambda^x_i$ plot are the appearance of butterfly-like patterns with four lobes. In the $\Lambda^x_i$ plot the corresponding patterns are replaced by butterfly-like patterns with eight lobes. The doubling of the number of lobes in $\Lambda^x_i$ is a consequence of the fact that $\Lambda^x_i$ is the energy derivative of the $\Lambda^y_i$ [see Fig. 1 (c)].

A careful analysis reveals that the butterfly-like structures accompany the topological phase transition, and these are a unique signature of the transition. To highlight this, we have encircled two regions in the topological phase diagram of the conduction band [see Fig. 5(a)], one near $\Delta = 8$ meV and the other near $\Delta = 34$ meV. Corresponding to these two points we have encircled two regions in the BCD phase-space plot [see Fig. 5(f)], which show butterfly-like structures. Similarly, we have encircled three phase-transition points in the valence band valley-Chern phase diagram in vicinity of $\Delta = 8$ meV, $\Delta = 28$ meV and $\Delta = 34$ meV. The corresponding regions having the butterfly-like structures are also marked in the BCD phase-space diagram of Fig. 5(f). Experimentally, we can fix the filling factor in vicinity of a Berry curvature hotspot (near a band edge), and measure the variation of the BCD with change in the perpendicular electric field. The butterfly structure described above, will feature in such experiments via the sign reversal in the NLA Hall conductivities across the phase transition, as shown in Fig. 4. This establishes that the sign reversal in the NLA Hall conductivities can be used as an experimental probe for the topological phase transition. The physical origin of the butterfly-like structure near the phase transition is a bit subtle, and it can be understood as follows. If we imagine a horizontal line through the center of the butterfly, then each of the lower two wings (lobes) of the butterfly corresponds to the BCD contribution of the two bands which touch each other at the phase-transition point. The corresponding colors represent opposite BCD before the transition. On the other hand, the upper two lobes of the butterfly-like structure are the BCD contribution of the same bands after the phase transition and the change in color at the same energy (imagine a vertical line) represents the phase transition. For instance, consider the butterfly around $\Delta = 34$ meV and $\mu \approx 35$ meV. The lower left (right) lobe is due to the first (second) conduction bands before phase-transition. The upper left (right) lobe represents the same band BCD contributions, but after the phase-transition.

From the above discussion, we expect a butterfly-like structures in the BCD diagram for each of the phase-transition points. However, there are few more subtleties worth mentioning here. The BCD and its thermal counterparts are Fermi surface phenomena and hence the total contribution at a given doping (or chemical poten-
V. EXPERIMENTAL IMPLICATIONS

In the last section, we have shown that the topological phase-transitions can be probed through the sign change in different BCD induced NLA Hall coefficients. However, in an experimental set up, the total NLA Hall response can have additional contributions from skew-scattering and side jump processes along with the BCD contribution\textsuperscript{65}. To separate out the BCD induced contribution of the NLA Hall signal from the rest and observe the sign change due to topological phase-transition is challenging and require some experimental sophistication. This can be done as follows. The different contributions of the NLA Hall signal scale differently with the scattering timescale or with the linear DC conductivity. By studying the scaling of total NLA Hall response with the Drude conductivity, the BCD dependent contributions can be extracted from the total NLA Hall response. In particular, the perpendicular electric field provides a useful knob to vary the conductivity at a fixed temperature to study the scaling in TDBG and detect topological transition from the sign-reversal of BCD\textsuperscript{15}. A fixed temperature ensures that the scaling parameters are not tuned with temperature\textsuperscript{66} and helps in accurate estimation of BCD. We note that measuring the other two counterparts of BCD (thermoelectric and thermal) requires control over heat current generation and detection and this is relatively harder to measure experimentally. However, recent advancements in thermal measurements\textsuperscript{67,68} may provide a way forward in the near future.

VI. CONCLUSION

To summarize, we have investigated the NLA Hall effect, the NLA Nernst effect, and the NLA thermal Hall effect in TDBG. We highlight that the breaking of the $C_3$ rotational symmetry in TDBG, by strain, is crucial for having finite NL Hall responses in moiré systems with trigonal symmetry. We find that the NLA Hall conductivities for charge and heat transport are maximum near the band edges, which are also the hotspots of the Berry curvature. Additionally, we show that the tunability of the band structure of TDBG through the perpendicular electric field can be exploited to tune the NLA Hall responses. Further, we demonstrate that the measurement of NLA Hall effects can be used as an experimental tool to probe topological phase-transitions of the valley-Chern type in time reversal symmetric systems. Specifically, we find that each topological transition is accompanied with a sign change in the NLA Hall coefficients, as reflected by the butterfly-like patterns in our calculations. Our study establishes the NLA Hall responses as an effective probe for detecting topological phase-transitions in moiré superlattices and in other systems.

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Appendix A: Band structure of TDBG in absence of strain

In this appendix, we construct the Hamiltonian of TDBG. We start with an initial brief review of the Bernal stacked bilayer graphene Hamiltonian, which is the building block of the TDBG. The Brillouin zone (BZ) of the AB-stacked bilayer graphene is identical to that of a monolayer graphene. The primitive lattice vectors $\mathbf{a}_1 = a(1,0)$ and $\mathbf{a}_2 = a(1/2, \sqrt{3}/2)$ yield the reciprocal lattice vectors to be $b_1^{(0)} = \frac{4\pi}{\sqrt{3}a}(\sqrt{3}/2, -1/2)$ and $b_2^{(0)} = \frac{4\pi}{\sqrt{3}a}(0,1)$. Here, $a$ is the lattice constant which is $\sqrt{3}$ times of the carbon-carbon bond length $d = 1.42$ Å. The coordinates of the vertices of the hexagonal first BZ are $K_\xi^{(0)} = \xi(2b_1^{(0)} + b_2^{(0)})/3$ with $\xi$ being the valley index. Near these BZ corners the electronic band dispersion\textsuperscript{69,70} and it can be described by an effective four band low energy model. Including the effects of hexagonal warping, the Hamiltonian near the $K$-valley can be expressed in terms of the fermion operators of the $A$ and the $B$ sublattice of the top and the bottom layers.
Here, the block diagonal matrices $h^{t/b}_k$ represents the massive Dirac Hamiltonian of the top and bottom monolayers and $t_k$ represents the effect of inter-layer hopping. The corresponding matrices are

$$ h^{t/b}_k = h_{v0} \sigma \cdot k + \frac{\delta}{2} (\mathbb{I} \mp \sigma_z), \quad t_k = \begin{pmatrix} -h_{v4}^{\pi \downarrow} & -h_{v3}^{\pi \downarrow} \\ h_{v4}^{\pi \uparrow} & -h_{v3}^{\pi \uparrow} \end{pmatrix}, $$

(A2)

with $\pi \equiv k_x + ik_y$. In the different intra-layer and inter-layer couplings have been introduced through the hopping parameter $\gamma_i$ or equivalently by $v_i = \sqrt{\frac{3}{2} |a_j|/(2\hbar)}$. The nearest neighbor intra-layer coupling between the A and the B sublattice is represented by the parameter $v_0$. The inter-layer intra-dimer coupling is represented through $\gamma_1$. Parameters $\gamma_3$ and $\gamma_4$ are the couplings between the inter-layer non-dimer sites coupling and the inter-layer coupling between dimer and non-dimer sites respectively. For our calculations we consider $\delta = 15$ meV, $\gamma_0 = -3.1$ eV, $\gamma_3 = 283$ meV and $\gamma_4 = 138$ meV.

A schematic of a moiré pattern is shown in Fig. 2(a) where the right-side arrow orientations (red, blue and green) represent the K-valley, while the left-side represents the K’-valley. The reciprocal lattice vectors of the moiré lattice are obtained as $G^i_m = b^{(1)}_i - b^{(2)}_i$, with the rotated reciprocal lattice vectors of each bilayer being specified by $b^{(l)}_i = R(\mp \theta/2) b^{(0)}_i$ with $\mp$ for bilayer $l = 1, 2$, respectively. Using this we obtain the pair of primitive moiré lattice vectors to be $G^x_m = \frac{8\pi}{3a}\sin \frac{\theta}{2}(-1/2, \sqrt{3}/2)$ and $G^y_m = \frac{8\pi}{3a}\sin \frac{\theta}{2}(1/2, \sqrt{3}/2)$. Using the low energy Hamiltonian [Eq. (A1)] for each lattice points, vertices of the small hexagons in Fig. 2(a), and the moiré hopping matrix, we construct the continuum Hamiltonian. A certain cut-off in the reciprocal space is used to truncate the lattice. The smallest TDBG Hamiltonian for the K-valley can be written as

$$ H = \begin{pmatrix} h^{+\downarrow}_{k,t} + \Delta_t^+ & 0 & 0 \\ 0 & h^{+\uparrow}_{k,b} + \Delta_b^+ & 0 \\ T^t & 0 & T^b \end{pmatrix}. $$

(A3)

Here, the superscripts on $h^{+\downarrow}_{k,t}/b$ represents rotated Dirac Hamiltonian as $h^\pm = R(\mp \theta/2) k \cdot \sigma$ and $\Delta_{t/b}^\pm$ represents the effect of a perpendicular electric field. In Eq. (A3), $T(r)$ represents the moiré coupling matrix, which connects the bottom layer of bilayer-1 to the top layer of bilayer-2. In this smallest TDBG Hamiltonian, only the nearest neighbor coupling will be considered which are connected by the vectors $q_{b} = \frac{8\pi}{3a}\sin \frac{\theta}{2}(0, -1)$ and $q_{t} = \frac{8\pi}{3a}\sin \frac{\theta}{2}(-\sqrt{3}/2, 1/2)$ and $q_{tr} = \frac{8\pi}{3a}\sin \frac{\theta}{2}(\sqrt{3}/2, 1/2)$. The moiré hopping matrices are given by

$$ T(r) = \sum_{j=b, tr, tl} T_{ij} e^{-i q_j \cdot r}, \quad \text{where} \quad (A4) $$

$$ T_b = \begin{pmatrix} \omega & \omega' \\ \omega' & \omega \end{pmatrix}, \quad T_{tr/tl} = \begin{pmatrix} \omega & \omega e^{i 2 \pi /3} \\ \omega e^{i 2 \pi /3} & \omega \end{pmatrix}. $$

(A5)

Here, $\omega$ and $\omega'$ denote the diagonal and the off-diagonal hopping strengths, respectively. We emphasize here that an unequal $\omega$ and $\omega'$, specifically $\omega' > \omega$, is known to be crucial to match the calculated low energy band structure with the experimentally observed spectral gap [10,25]. In this paper, we consider $\omega' = 106$ meV and $\omega = 79$ meV [10,26].

### Appendix B: Electronic band structure of TDBG in presence of strain

In this section of Appendix we highlight the details of the strain implementation in TDBG. In presence of strain, the Dirac Hamiltonian of Eq. (A3) modifies to

$$ h_{k,l} = h_{v0} R(\mp \theta/2) [(k+nE_T)](k-D_\xi)\cdot(\xi \sigma_x, \sigma_y) + \frac{\delta}{2} (\mathbb{I} \mp \sigma_z). $$

(B1)

Here, the strain matrix operate over the position of the twisted Dirac points given by

$$ D_\xi = (\mathbb{I} - E_T) K^T_\xi - \xi A, $$

(B2)

with $A$ representing the gauge field that has the dimension of reciprocal lattice vector. The appearance of the gauge field can be attributed to the fact that the strain causes the inter-atomic distance in each layer to become different in different directions. This results in the difference of hopping parameters which displaces the Dirac point from its original position. The gauge potential $A$ in terms of the elements of the strain matrix is given by

$$ A = \frac{\sqrt{3}}{2a} \beta (E_{xx} - E_{yy}, -2E_{xy}) . $$

(B3)

Here, $\beta = 1.57$ and $E_{ij}$ are the elements of the strain matrix.

Strain also modifies the lattice vectors and consequently the hopping matrices and the hopping vectors. We calculate the strained moiré vectors starting from un-rotated and un-strained lattice vectors. Following Refs. [9,19], we obtain the lattice vectors using $G^{x\downarrow}_m = R_{-2}(1-E_T)b_1 - R_{-2} b_1$ and $G^{y\downarrow}_m = R_{-2}(1-E_T) b_2 - R_{-2} b_2$ which yields...
Here, the band. Equation (C2) depends on the valley index number for the massive Dirac low energy model (without respective bands is evident. Using the expression of Berry A sign change in the Berry curvature distribution in the s

Here, \( \phi \) changes. We calculate the change in the phase factor as

\[
\frac{\phi_{tr}}{2} = \frac{2\pi}{3} \left( 1 + \sqrt{3} \varepsilon_{xx} \varepsilon_{zy} + \sqrt{3} \varepsilon_{xy} \varepsilon_{yy} - \varepsilon_{xy}^2 - \varepsilon_{yy}^2 \right) \tag{B7a}
\]

\[
\frac{\phi_{tr}}{2} = \frac{2\pi}{3} \left( 1 - \sqrt{3} \varepsilon_{xx} \varepsilon_{zy} - \sqrt{3} \varepsilon_{xy} \varepsilon_{yy} - \varepsilon_{xy}^2 - \varepsilon_{yy}^2 \right) \tag{B7b}
\]

Appendix C: Topological phase transition in tilted massive Dirac Hamiltonian

In this Appendix, we convey the central idea of topological phase-transition of valley-Chern type employing a simplistic model for tilted massive Dirac Hamiltonian. For that we consider a model Hamiltonian as

\[
H_s = \hbar v_F (k_x \sigma_y - s k_y \sigma_x) + \hbar \nu_t k_x + \Delta \sigma_z \ . \tag{C1}
\]

Here, \( v_F \) is the Fermi velocity and \( s = \pm \) is the valley index and \( \Delta \) is the gap parameter that captures the potential induced by the perpendicular electric field. Here, \( \nu_t \) is the top and bottom gate and its sign can be reversed. The Berry curvature for this model is given by

\[
\Omega_z = \mp s \frac{\hbar^2 v_F^2 \Delta}{2(\hbar^2 v_F^2 k^2 + \Delta^2)^{3/2}} \ . \tag{C2}
\]

Here, the \( \mp \) sign stands for the conduction and valence band. Equation (C2) depends on the valley index \( s \). For \( s = + \) we have shown the Berry curvature weighted dispersion for \( \Delta < 0 \) in Fig. 6(a) and for \( \Delta > 0 \) in Fig. 6(b). A sign change in the Berry curvature distribution in the respective bands is evident. Using the expression of Berry curvature, we can calculate the valley-Chern number. For that we need to first construct a definition of valley-Chern number for the massive Dirac low energy model (without properly defined BZ). The valley-Chern number may be defined in the infinite Dirac Fermi sea as

\[
C_v = \frac{1}{2\pi} \int \Omega_z dk \ . \tag{C3}
\]

This expression can be computed numerically choosing the integration regime appropriately. However, for analytical calculation of the valley-Chern number we first calculate \( \int \Omega_z f_0 dk \) and then consider \( \Delta \ll \mu \) the limit. Using this recipe for the isotropic Hamiltonian \((\nu_t = 0)\)
we calculate the valley-Chern number for the conduction band with $\Delta > 0$, to be

$$C_v = -\frac{s}{2} \left(1 - \frac{\Delta}{\mu}\right) \tag{C4}$$

Now in the infinite Fermi surface limit ($\Delta \ll \mu$) we get $C_v = -s/2$. We note that the two valley have opposite valley-Chern number. Since the valley-Chern numbers are opposite and sums up to zero, so a topological index can be defined as $Z_2 = (C_+ - C_-)/2$. Now, if we reverse the sign of the perpendicular electric field then the valley-wise Chern number gets altered. This electric field induced topological phase-transition has been highlighted in Fig. 6(c). We emphasize that other than the perpendicular electric field, a topological transition can also occur by changing the stacking from AB to BA registry\textsuperscript{23}. The consequence of the phase-transition on the NLA Hall effect has been highlighted in Fig. 6(d).

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