Layer-Contrasted Hall Effect in Twisted Bilayers with Time Reversal Symmetry

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We discover a layer-contrasted charge Hall effect in chiral bilayers with time reversal symmetry. The restriction by Onsager relation on having linear Hall response in individual layers is lifted by layer hybridization of electronic states in the chiral structures, allowing opposite Hall voltages to develop in opposite layers. This effect also amounts to a layer Hall effect, namely the Hall transport of layer pseudospin that carries charge dipole, and the underlying band geometric quantity is the momentum-space vorticity of the layer current. We demonstrate the effect in twisted bilayer graphene and twisted homobilayer transition metal dichalcogenides with a wide range of twist angles, which exhibits giant Hall ratios under experimentally practical conditions, with gate voltage controlled on-off switch. This work reveals novel Hall physics in chiral structures, and opens up a new direction of layertronics that exploits the quantum nature of layer degree of freedom to uncover exciting effects.

Within linear response, Hall effect is forbidden in systems respecting time reversal (TR) symmetry, by the Onsager relation of electrical conductivity. Charge Hall transport, and its quantized form, have been observed only with TR symmetry breaking, either by magnetic field or magnetic order [1–3]. Such a restriction has also inspired important research directions to explore in TR symmetric systems Hall-type transport of non-electrical quantum degrees of freedom like spin and valley pseudospin [4, 5]. By such effects only spin or valley polarizations, rather than charge, accumulate at the edges, leading to null Hall voltage in compliance with the Onsager relation.

In this work we reveal an opportunity offered by chiral structures for exploring charge Hall effects under TR symmetry. In a double layer geometry, chiral crystal symmetry permits layer-contrasted Hall transport and charge accumulations at edges, whereas interlayer quantum tunneling lifts the restriction of Onsager relation on having opposite Hall voltages from the two layers as subsystems. Such a phenomenon can be understood from the perspective of a dipole Hall effect, in which the out-of-plane layer-pseudospin (LPS) as a charge dipole moment is transported transverse to the in-plane electric field and accumulated near the edges [see Fig. 1(a)]. Our symmetry analysis reveals that this is a characteristic of general chiral bilayers with Fermi surface. Van der Waals bilayers with a twist [6–12] therefore offer natural experimental platforms to explore the effect, with layer-resolved access of Hall voltage facilitated by the vdW double layer geometry [13–15].

This effect is fundamentally distinct from the layer-polarized Hall effect in layered antiferromagnetic insulators [19–21] and the layer-dependent quantum Hall effect [13–18], both of which rely on the TR symmetry breaking, and from the “nonlinear Hall” effects [22, 23] where the Onsager relation is obviously irrelevant. We show that the layer-resolved linear Hall response here is rooted in a distinct band geometric quantity—the momentum space vorticity of the LPS current—that emerges from the interlayer hybridization of electronic states under TR and chiral symmetry. We demonstrate the effect using exemplary systems of twisted bilayer graphene (tBG) and twisted homobilayer transition metal dichalcogenides (tTMDs) [6–12] with a broad range of twist angles. Within experimentally feasible range of carrier doping [6–18, 24, 25], we find pronounced Hall responses accompanied by giant Hall ratios (up to $O(10)$ in tBG), with sign and magnitude controlled by the twist angle. The effects in tTMDs also feature remarkable on-off switchability by gate voltage that promises device applications.

Symmetry characters. We first understand the emergence of the layer (pseudospin) Hall effect from the perspective of symmetry. The LPS current generated at the linear order of a driving electric field $E$ is given by

$$j_a^l = \sigma_{ab}^l E_b,$$

(1)

where the Einstein summation convention is adopted for in-plane Cartesian Coordinates $a$ and $b$, and $j_a^l$ is the current along the $a$ direction of the out-of-plane interlayer dipole moment. Since the LPS current is odd under TR while the electric field is even, breaking TR is necessary to allow this effect, either by magnetic order in equilibrium or by nonequilibrium kinetics of electrons around the Fermi surface. In nonmagnetic metallic states the effect can only stem from the latter, and the resulting $\sigma_{ab}^l$ is a TR even tensor.

The Hall conductivity, which is antisymmetric with respect to the directions of the in-plane driving field and response current, is dual to $\sigma_z^l \equiv (\sigma_{xy}^l - \sigma_{yx}^l)/2$, which transforms as a pseudoscalar (the $zz$ component of a TR even pseudotensor). It remains unchanged under rotation, but changes sign under space inversion, mirror reflection and roto-reflection. Such a layer Hall effect is therefore allowed provided that the bilayer crystal structure is chiral.

This chiral symmetry requirement matches perfectly
with the twisted bilayer van der Waals structures, such as the tBG and tTMDs. These most studied systems, which are also our foci, are all based on the honeycomb-lattice monolayer and preserve the threefold rotation symmetry in the $z$ direction. This symmetry forbids the off-diagonal components of the symmetric part of $\sigma_{ab}^l$ with respect to $a$ and $b$, thus enforces a pure Hall current quantified by $\sigma_{xy}^l = -\sigma_{yz}^l = \sigma_{zx}^l$.

By symmetry analysis, we also find that chiral bilayers isolate the layer Hall effect, which features the interlayer charge dipole accumulation near the layer boundary, from the LPS counterpart of the Edelstein effect [29], in which the interlayer dipole is generated in the 2D bulk of the sample at the linear order of in-plane $E$ field. The latter effect is forbidden in all the 9 chiral crystal classes relevant to 2D materials but the $C_1$ class without any symmetry, including the cyclic point groups $C_m$ and dihedral point groups $D_m$ with $m = 2, 3, 4, 6$ (summarized in Table I). Moreover, the in-plane two-fold rotation axis in dihedral groups prohibits any interlayer polarization in equilibrium, while the cyclic groups with an out-of-plane polar axis may allow it. In the latter case, the edge interlayer dipole accumulation by the layer Hall effect can be measured by taking the difference between the signals in the presence and absence of external field. In practice, the interlayer polarization can be measured locally [27, 28]. Nevertheless, we will not go into pertinent details, instead we spotlight the concurrent layer-contrasted charge Hall effect, which is also a direct experimental manifestation of the layer Hall effect.

By symmetry arguments one can see that the layer Hall effect amounts to a layer-contrasted charge Hall effect in coupled bilayer systems. The total charge current is the sum of the charge flows on the two layers $\mathbf{j} = \mathbf{j}^i + \mathbf{j}^b$. As the driving current is applied on both layers, the Onsager relation for the electrical conductivity of a TR symmetric bulk material [1] forbids any total charge Hall current flowing out of the bilayer system within linear response. This restriction, however, allows the double-layer driving current to induce layer-contrasted Hall voltages that appear only in the subsystem—each layer—of the coupled bilayer system. Here the interlayer coupling is indispensable, otherwise the linear charge Hall effect in each independent layer is again prohibited by the Onsager relation. Additionally, the LPS current, which measures the difference of the charge flows on two layers, must be finite, i.e., $\mathbf{j}^l = \mathbf{j}^i - \mathbf{j}^b \neq 0$. The layer Hall effect—available only in chiral bilayers with TR symmetry—thus offers a unique scenario to realize the layer-contrasted charge Hall effect without magnetic order: $\sigma_{xy}^{l(b)} = -\sigma_{xy}^{b(a)} = \sigma_{xy}^{l}/2$, where the conductivity $\sigma_{xy}^{l(b)}$ quantifies the charge Hall effect in the top (bottom) layer. In practice, the layer-contrasted Hall voltage can be measured by Hall bar setup on a specific layer, as is illustrated in Fig. 1(a).

**Theory: Layer current $k$-space vorticity.** The LPS current density is given by the integral of the current carried by each electron with velocity $\mathbf{v}_n^l(\mathbf{k})$ weighted by the distribution function $f_n(\mathbf{k})$:

$$j^l = e \sum_n \int \frac{d^2k}{(2\pi)^2} f_n(\mathbf{k}) \mathbf{v}_n^l(\mathbf{k}),$$

where $n$ and $\hbar$ are the band index and crystal momentum, and $\mathbf{v}_n^l(\mathbf{k}) = (u_n(\mathbf{k}))^\dagger \mathbf{v}^l |u_n(\mathbf{k})\rangle$ is the LPS velocity, represented by the operator $\mathbf{v}^l = \frac{1}{2} \{ \mathbf{\hat{n}}, \mathbf{\hat{\sigma}} \}$ in a bilayer system, carried by the state $|u_n(\mathbf{k})\rangle$. Here the Pauli matrix $\mathbf{\hat{\sigma}}$ is the out-of-plane LPS operator operating in the layer index subspace [29]. Because of the TR symmetry, $\mathbf{v}_n^l(\mathbf{k}) = -\mathbf{v}_n^b(-\mathbf{k})$, hence a nonzero LPS current requires a distribution function in $k$-space that breaks the occupation symmetry at $\mathbf{k}$ and $-\mathbf{k}$. Such an off-equilibrium distribution can be driven by an electric field and described by the Boltzmann transport equation. Within the simplest constant relaxation time approximation, the deviation from the equilibrium Fermi distribution $f_0 \equiv f_0(\varepsilon_n)$ is of a dipole structure in $k$-space: $f_n - f_0 = -\frac{\tau}{\hbar} \mathbf{E} \cdot \partial_{\varepsilon_n} f_0$, with $\varepsilon_n$ being the band energy and $\tau$ the transport relaxation time. This approximation is usually taken so that the specific content of disorder usually unknown does not pose a difficulty and that the band origin of the effect can be manifested [22, 23, 30].

The layer Hall conductivity thus reads

$$\sigma_{xy}^l = \frac{e^2}{\hbar} \mathbf{v} \cdot \mathbf{\mathcal{V}},$$

where

$$\mathcal{V} = -\frac{\hbar}{2} \sum_n \int \frac{d^2k}{(2\pi)^2} f_0 \left[ \mathbf{v}_n(\mathbf{k}) \times \mathbf{v}_n(\mathbf{k}) \right]_z$$

is intrinsic to the band structure, has the dimension of frequency, and is indeed a TR even pseudoscalar conforming to the symmetry analysis. Here $\mathbf{v}_n$ is the group velocity, and $f_0 = \partial f_0/\partial \varepsilon_n$ implies that the layer Hall effect is a Fermi surface property. If the interlayer coupling is absent, one has $\mathbf{v}_n^l(\mathbf{k}) = \mathbf{v}_n(\mathbf{k}) \sigma_{xy}^{l(b)}$ hence no layer Hall effect. If the LPS current in Eq. (4) is replaced

| Chiral point groups | equilibrium interlayer polarization | Layer Hall effect | Layer Edelstein effect |
|--------------------|------------------------------------|------------------|------------------------|
| $C_2$ (out-of-plane axis); $C_m$, $m = 3, 4, 6$ | ✔ | ✔ | ✗ |
| $C_2$ (in-plane axis); $D_m$, $m = 2, 3, 4, 6$ | ✗ | ✔ | ✗ |
by the total charge current of a bilayer system, one gets a vanishing total charge Hall effect. The formal theory thus confirms the conclusions of the foregoing symmetry arguments.

Now we show that the layer Hall effect has a band origin in the layer current $k$-space vorticity. Via an integration by parts, Eq. (4) is recast into

$$\mathcal{V} = \sum_n \int \frac{d^2 k}{(2\pi)^2} f_n \omega_n(k),$$

which measures the $k$-space vorticity of the LPS current $\omega_n(k) = \frac{1}{2i} \partial_\theta \times \mathbf{v}^l_n(k)_z$ integrated over the occupied states. As the integral of this vorticity over any full band vanishes, only $\omega_n(k)$ of partially occupied bands contribute to a net layer Hall effect. The layer current vorticity can be expressed in an enlightening form

$$\omega_n(k) = \hbar \text{Re} \sum_{n_1 \neq n} \frac{[\mathbf{v}_{nn_1}(k) \times \mathbf{v}^l_{n_1}(k)]_z}{\varepsilon_n(k) - \varepsilon_{n_1}(k)},$$

where the numerator involves interband matrix elements of velocity and LPS current operators. This expression of $\omega_n(k)$ shares a striking similarity with the band geometric quantity called $k$-space Berry curvature [31]: The former becomes the latter if $\mathbf{v}^l_{n_1}(k)$ is replaced with the $k$-space interband Berry connection $A_{nn_1} = \langle u_n | i \partial_\theta | u_{n_1} \rangle$. As such, the layer Hall effect, despite being described by the Boltzman transport theory, encodes the information of interband coherence, which has up to now mostly connected to intrinsic transport effects induced by various Berry-phase effects [11] [4] [5] [31].

It is interesting to note that $\omega_n(k) = -\text{Im} \mathcal{C}$, where $\mathcal{C} = \sum_{n_1 \neq n} (A_{nn_1} \times \mathbf{v}^l_{n_1})_z$. Being the imaginary part of $\mathcal{C}$, the $k$-space vorticity of the LPS current is connected to the real-space circulation of this current around the electron wave-packet center $r_\mathcal{P}$. $\mathcal{C}$: $(\mathbf{r} - r_\mathcal{P}) \times \mathbf{v}^l = \text{Re} \mathcal{C}$. While $\text{Re} \mathcal{C}$ stems from the self-rotational motion of the electron wave packet, $\omega_n(k) = -\text{Im} \mathcal{C}$ results from the center-of-mass motion. Their relation is analogous to the $k$-space Berry curvature and quantum metric, which are the imaginary and real parts of the quantum geometric tensor [32].

The nonzero layer current vorticity requires the quantum interlayer hybridization of electronic states, which is a characteristic property not shared by the Berry curvature. If the states $|n_1\rangle$ and $|n_1\rangle$ involved in Eq. (6) are polarized in the same layer around some momentum $k$, one gets $\mathbf{v}^l_{n_1}(k) = \mathbf{v}^l_{n_1}(k)\sigma^l_{n_1}(k)$ and thus vanishing layer current vorticity $\omega_n(k) = 0$. On the other hand, if the two states are polarized in different layers, then $\mathbf{v}^l_{n_1}(k)$ is suppressed, so does $\omega_n(k)$. Therefore, the layer current vorticity favors strongly layer-hybridized states around band near-degeneracy regions.

**Application to tBG.** We now apply our theory to tBG. For small twist angles $\theta$, we employ the continuum model with parameters taken from Ref. [33]. The results are corroborated by tight-binding calculations, which are also applicable at large twist angles. All model details are presented in the Supplemental Material [34].

The calculation results for $\theta = 1.05^\circ$ and $1.47^\circ$ are shown in Figs. (1b) and (c). The central bands around zero-energy are separated from their neighbors with a global gap at such small angles. When the Fermi level intersects the central bands, $\sigma^l_{xy}$ shows two narrow peaks.
with opposite signs for electron and hole doping. When the Fermi level is located in the global gap, \( \sigma_{xy} \) vanishes as a Fermi surface property. Its magnitude starts to increase again when the Fermi level is shifted outside the gap. Assuming a relaxation time of 1 ps \[35–37\], the layer Hall conductivity can reach dozens of \( e^2/h \) upon Fermi level shifts within 20 meV. Such slight shifts can be readily achieved by dual gate. The experimental measurement shall also be facilitated by a large Hall ratio \( \sigma_{xy}/\sigma_{xx} \) which is an intrinsic quantity if the longitudinal charge conductivity \( \sigma_{xx} \) is also evaluated using the constant \( \tau \) approximation. In the current case, \( \sigma_{xx} \) is strongly suppressed by the quite flat dispersion, thus the Hall ratio can be \( \gg 1 \), as shown in the inset of Figs. 1(b) and (c).

The layer Hall effect is not restricted to long-range moiré lattices at small \( \theta \). When \( \theta \) increases, the profiles of \( \sigma_{xy} \) remain similar, but the width and magnitude of its peaks increase. This is illustrated in Fig. 4(d), where the central peaks of \( |\sigma_{xy}| \) are presented for a series of \( \theta \). While the two peaks become more separated as \( \theta \) increases, sizable \( \sigma_{xy} \) \( \sim \mathcal{O}(1) \) \( e^2/h \) within dozens of meV around zero-energy is still achievable for a wide range of \( \theta \).

Next we look at the \( k \)-space distributions of LPS and LPS current vorticity to have a better understanding of the features of the layer Hall effect. We illustrate these in Fig. 2 using a 2.65° tBG and focus on the first conduction band. The band projection of the LPS is denoted by color in Fig. 2(a). At low energies, the layer hybridization is weak, thus the states are dominantly upper (lower) layer polarized around \( \kappa \) (\( \kappa' \)). At higher energies, Dirac cones come from the two layers intersect and hybridize strongly along the \( \gamma \) point, rendering LPS \( \sim 0 \). Such layer polarizations or hybridizations in different band regions are also manifested in Fig. 2(b) for the distribution of the layer current vorticity. It is concentrated along the path from \( \gamma \) to \( m \), which is characterized by regions with strongly layer-hybridized and nearly-degenerate bands, and is suppressed in the layer polarized regions around \( \kappa \) and \( \kappa' \). White curves in Fig. 2(b) show two different Fermi surfaces. At low electron doping, \( \sigma_{xy} \) is contributed by the dark blue area in Fig. 2(b) with \( \omega_n < 0 \), thus it is negative and increases in magnitude as the Fermi level is lifted towards the middle of the band [see Eq. (6) and brown curve in Fig. 2(a)]. As the Fermi level is further increased, regions with highly concentrated \( \omega_n > 0 \) start to contribute, hence the magnitude of \( \sigma_{xy} \) drops. Evolution of \( \sigma_{xy} \) with the Fermi level can also be understood from the distribution of \( v_n \times v'_n \) shown in Fig. 2(c). Since it is dominantly negative in the blue regions, according to Eq. (4), \( \sigma_{xy} \) shall be negative and maximal when the Fermi level locates around the middle of the band.

**Application to tTMDs.** Now we briefly address the layer Hall effect in tTMDs and focus on its tuning. We consider the continuum model of tMoTe\(_2\) \[38, 39\] as an example (details in Ref. [34]). The calculation results for \( \theta = 1.2° \) and \( 3° \) are shown in Fig. 3. The obtained layer Hall conductivity can reach \( \sim e^2/h \) and the Hall ratio \( \sigma_{xy}/\sigma_{xx} \sim \mathcal{O}(0.1) \). It exhibits rich profiles, which can be attributed to the complexity of TMDs band structures that feature multiple isolated wide bands, and the efficient layer hybridization in this material. As is shown in Fig. 3(a) for the case of \( \theta = 2° \), most band regions are strongly layer hybridized.

Properties of tTMDs can be sensitively tuned via the interlayer bias \( V_z \). The band structures of 2° tMoTe\(_2\) with different \( V_z \) are shown in Figs. 3(a) – (c), and the corresponding layer Hall conductivities are presented in Fig. 3(d). The profiles of \( \sigma_{xy} \) vary dramatically for different \( V_z \), including the magnitude and sign. A prominent impact of interlayer bias is the suppression of \( \sigma_{xy} \) at low energies [grey area in Fig. 3(d)], which indicates that the layer Hall effect can be turned on/off with gate control. This is because the interlayer bias polarizes low-energy electrons into one of the layers [see the dominance of green color in Figs. 3(b) and (c)], thus reduces interlayer coupling.

This gate suppressed layer Hall effect totally differs from the gate induced layer-polarized Hall effect appearing in the even-layer antiferromagnet MnBi\(_2\)Te\(_4\) \[10, 20\]. This distinction arises from the fact that the former and latter effects favor strong layer hybridization and polarization, respectively. It is also noted that the linear Hall effect in a nonmagnetic bilayer can only appear in the
layer-contrasted manner in line with the Onsager relation, irrespective of the gate voltage; while the layer-resolved Hall effects in each ferromagnetic layer of antiferromagnetic bilayer MnBi$_2$Te$_4$ can be quite different when the combined symmetry of TR and space inversion is broken by the gate.

In summary, we have revealed that the chiral crystal symmetry and quantum interlayer hybridization of electronic states render a unique approach to the layer-contrasted Hall effect in bilayer systems with TR symmetry and quantum interlayer hybridization of electronic states. This effect not only uncovers novel Hall physics based on the quantum layer-hybridization, but also serves as an efficient probe of chiral bilayers. It opens up a new research direction of layertronics that exploits the quantum nature of the layer degree of freedom.

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