Berry curvature, spin Hall effect and nonlinear optical response in moiré transition metal dichalcogenides heterostructure

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Recently, the spin Hall effect was experimentally observed in the AB stacked MoTe$_2$/WSe$_2$ heterobilayer. In this work, we perform a systematic study of the Berry curvature effects in moiré transition metal dichalcogenides heterostructure. We point out that the moiré potential on the remote conduction band would induce a sizable periodic pseudo-magnetic field (PMF) on the valence band. This periodic PMF enables net Berry curvature flux in each valley. Due to the spin-valley locking, the PMF can thus cause the spin Hall effect observed in the experiment. Moreover, we propose that the valley-contrasting Berry curvature distribution induced by the PMF can be probed through nonlinear optical responses. Our work deepens the understanding of the Berry curvature effects and their relation with periodic PMFs in the moiré transition metal dichalcogenides heterostructure.

Introduction—Two-dimensional moiré materials have paved a new avenue toward engineering topology, correlation, superconductivity and magnetism in a flat-band system in recent years. These moiré materials provide rich platforms for studying novel transport and optical responses induced by Berry curvature effects. For example, magneto-electric and nonlinear Hall effects have been demonstrated in twisted graphene superlattice and twisted transition metal dichalcogenides (TMD) homobilayer.

Notably, moiré TMD heterobilayers, in which moiré pattern mainly originates from the lattice mismatching between two distinct TMD layers, have been observed to exhibit nontrivial topological and correlation properties. It was studied that a quantum anomalous Hall state at filling $\nu = 1$ (one hole per moiré unit cell) was observed in AB stacked moiré MoTe$_2$/WSe$_2$ heterobilayers. Very recently, the spin Hall torque has been demonstrated near $\nu = 1$ stemming from the large Berry curvature in this bilayer heterostructure. However, unlike the graphene moiré superlattice or twisted TMD homobilayer, the novel responses induced by the Berry curvature in TMD heterobilayer remain unknown theoretically. Moreover, in previous works, the model for TMD heterobilayer is simply described by

$$H = -\frac{p^2}{2m} + V(\mathbf{r}),$$

where $p$ is the crystal momentum operator, $m$ is an electron effective mass and $V(\mathbf{r})$ is the moiré potential. As $H$ simply represents a valence band free Fermion moving in a periodic potential, the finding of Berry curvature induced spin Hall effect in the experiment is quite surprising.

In this work, we describe the moiré TMD heterobilayer as a massive Dirac Fermion moving in periodic moiré potential, in which the moiré potential of both conduction band and valence band are taken into account. Given that the low energy states are near the valence band edge, we project out the freedom of conduction band by using the quantum commutation relation of crystal momentum $\hat{p}$ and position $\hat{r}$. Remarkably, we find the moiré potential on the conduction band, which although is $1 \sim 2$ eV away, contributes a periodic pseudo-magnetic field to the valence band in the low energy state. We next show that the periodic PMF results in a moiré valley-contrasting Berry curvature distribution, which exhibits net Berry curvature in each valley. Being consistent with the experiment, we find a large spin Hall effect in this case. It arises from a combination of the giant Ising SOC and the net Berry curvature induced by PMF. Finally, we show that the predicted moiré valley-contrasting Berry curvature distribution induced by the periodic PMF would exhibit a salient feature in the shift current response, which is a second-order DC response by applying a linear polarized light. The shift current response is tied to the quantum geometric properties of the system and microscopically varies due to changes in properties of the Bloch wavefunction upon excitation between bands.

Model Hamiltonian—Due to a large band offset (hundreds of meV) between the two layers in moiré TMD heterostructure, we assume the low energy states arise from one layer, while the other layer contributes to a periodic moiré potential. It is known that the 2H-TMD monolayer is described by massive Dirac Fermions. We thus model the TMD heterostructure with a massive Dirac Hamiltonian including slow-varying moiré potential on both conduction and valence band:

$$H = v_f \left( \begin{array}{cc} 0 & \pi^{\dagger} \\ \pi & 0 \end{array} \right) + \left( U_v(\hat{r}) 0 \\ 0 U_v(\hat{r}) \right) + \frac{\Delta}{2} \sigma_z,$$  

where the ladder operator $\hat{\pi} = \tau \hat{p}_x + i \hat{p}_y$, $v_f$ is the Fermi velocity, $\Delta$ is the energy gap between conduc-
The effective mass $U$ is described by a massive Dirac model with a modified moiré and green atoms. The low energy physics of the top layer arises from lattice relaxation can also induce uniform strain distribution imposed on moiré TMD heterobilayers arising from lattice relaxation [30, 46]. The low energy Hamiltonian adopts a topological phase transition [30]. The physical origin of the topology is shown to be understood in terms of the Haldane model with zero magnetic flux in a single unit cell [30] [46]. The low energy Hamiltonian adopted in [30] is almost the same as Eq. (3), but the origin of the PMF term arises from lattice relaxation. In this work, we define the PMF $B_{ps}(r)$ as:

$$B_{ps}(r) = \partial_x A_y - \partial_y A_x = \tau B_0 \sum_{i=1}^3 \cos(G_i \cdot r + \phi_c)$$ (4)

with the PMF strength $B_0 = \frac{hU^2 c^2}{e\Delta}$. The PMF strength is mainly determined by the energy gap $\Delta$ and the conduction band moiré potential $U_c$. It is worth noting that the moiré potential on valence band has no influence on the PMF though it plays an important role to the band structure.

To see how the PMF affects the moiré band structure of a TMD heterobilayer, we can then diagonalize the effective Hamiltonian with plane wave basis. The resulting conduction band moiré potential $U_c = 40$ meV and energy gap $\Delta = 1$ eV, we find the PMF strength $B_0$ is as sizable as 55 T. Naively, it seems one can completely neglect the conduction band and its moiré potential as $\Delta$ is very large in this case. However, our finding points out that the conduction band moiré potential would enable the states at valence band to feel an effective PMF.
The calculated spin valley Hall conductivity $\sigma_{xy}$ as a function of chemical potential $\mu$ ($\mu \approx -40$ meV near the gap of top two moiré bands in Fig.1(c)) with different strength of $B_0$. We set $U_v = 15$ meV, $\phi_v = 0.3\pi$, $\phi_c = 0.5\pi$. We point out an intrinsic origin to generate the PMF with conduction band moiré potential in common TMD heterobilayers.

Spin Hall effect induced by moiré valley contrasting Berry curvature—Besides the nontrivial topology, the question is whether the valley-contrasting Berry curvature would induce some novel responses, which would help to identify the PMF effects in moiré TMD heterobilayer. In this section, we propose that a large spin Hall effect could be induced by the PMF, which may provide a plausible explanation to the spin Hall torque seen in MoTe$_2$/WSe$_2$ heterobilayer recently.

Using the effective Hamiltonian in Eq.(3), we can calculate the spin-valley Hall conductivity $\sigma_{xy}^{sv}$:

$$\sigma_{xy}^{sv} = \frac{2e^2}{h} \int \frac{d^2k}{(2\pi)^2} \left[ f_1(k)\Omega_1(k) + f_2(k)\Omega_2(k) \right]$$

where $1(2)$ is the band index of the first(second) moiré band in Fig.1(c), $\Omega_n(k)$ is the Berry curvature of $n$-th band, the integral is calculated over moiré Brillouin zone, and $f_{1,2}(k) = \{1 + \exp[(E_{1,2}(k) - \mu)/k_BT]\}^{-1}$ are the Fermi-Dirac functions. Note that $\Omega$ is valley-contrasting due to the time-reversal symmetry ($\Omega_{1,2}^{\pm} = -\Omega_{2,1}^{\mp}$). As a result, under an in-plane electric field, $\Omega$ can drive charge carriers at opposite valleys to flow in opposite transverse directions, which leads to transverse spin-valley currents (Fig.2(a)).

In Fig.2(b) we show the spin-valley Hall conductivity $\sigma_{xy}^{sv}$ for different $B_0$. For $B_0 = 0$, $\sigma_{xy}^{sv} = 0$ because the spinless time reversal symmetry enforces $\Omega_k = -\Omega_{-k}$ [30]. In contrast, the spin-Hall conductance becomes finite in the presence of the PMF. It is clear that $\sigma_{xy}^{sv}$ increases as the PMF strength $B_0$ increases, and the maximum value of $\sigma_{xy}^{sv}$ shows a linear increase at different value of $B_0$ which is shown in the inside panel. Therefore, we have demonstrated that in spite of the large gap between conduction and valence band in a massive Dirac model, the PMF on the valence band is generated by a moiré modulation. Such PMF would enable the presence of a large spin-Hall effects induced by the valley contrasting Berry curvature between top two moiré bands in moiré heterobilayer TMD.

Terahertz optical responses—As we have shown in previous section, the PMF would influence the Berry curvature effects of TMD heterobilayers significantly. Next, we show the PMF strength can be explicitly seen in the experiment by studying the terahertz optical responses of TMD heterobilayers. We set the chemical potential near $\nu = 2$(two holes per moiré unit cell) so that the relevant states contributing to the terahertz response would contain the information of the PMF (see Fig.3(a)).

Before presenting the results of nonlinear terahertz optical responses, we actually first looked at linear optical conductivity $\sigma_{\alpha\beta}(\omega)$, where $\alpha, \beta$ label the polarized direction of the light. As shown in Supplementary Note 4, we find the longitudinal optical conductivity $\sigma_{xx}$ is almost insensitive to the PMF. It is because the value of $\sigma_{xx}$ mainly reflects interband linear resonant optical response strength while the Berry curvature is not that essential in this case. Interestingly, we find that the spin-valley optical conductivity defined as $\sigma_{xy}^{sv} = \frac{1}{2}(\sigma_{xy}^{v+} - \sigma_{xy}^{v-})$ can be enhanced by the PMF. See more detailed results about the calculated $\sigma_{\alpha\alpha}$ in Supplementary Note 4. But we still find that in general, it is hard to intuitively see the strength of PMF from the linear optical response only.
According to the previous works\cite{44,48,49}, nonlinear terahertz optical responses can reflect the topological nature of wavefunctions. On the other hand, we have shown the PMF can induce a valley-contrasting Berry curvature. To manifest the PMF strength through optical responses, we thus now look at the second order nonlinear terahertz optical response. As we will show the shift current response can fit our purpose, which measures a DC photocurrent driven in second order optical response in noncentrosymmetric quantum materials by shining a linear polarized light.

The shift current arises from the interband and intraband Berry connection by shielding a linear polarized light. It characterizes the nontrivial band topology of the moiré bands during the optical transition process. With a electric field $E_{\beta}(\omega)$ at frequency $\omega$ and linearly polarized in the $\beta$ direction, the shift current $J_{\alpha}$ in the $\alpha$ direction has the form \cite{44}

$$J_{\alpha} = \sigma_{\beta\beta}^{\alpha} E_{\beta}(\omega) E_{\beta}(-\omega)$$

where the second-order conductivity tensor $\sigma_{\beta\beta}^{\alpha}$ has the form\cite{44}

$$\sigma_{\beta\beta}^{\alpha}(\omega) = \frac{2g\pi e^3}{\hbar^2 S} \sum_{nm,k} f_{nm} \text{Im}(r_{mn}^{\beta} r_{nm;\alpha}^{\beta}) \delta(\hbar\omega_{nm} - \hbar\omega)$$

where $S$ is the sample area, $g = 2$ is the spin(valley) degeneracy, $n$ and $m$ are band indexes and $\hbar\omega$ is the photon energy. The occupation difference $f_{nm} = f_n - f_m$ with $f_n$ is the Fermi-Dirac distribution of band $n$. $r_{mn}^{\beta}$ are the inter-band Berry connections defined as $r_{mn}^{\beta} = i\langle m|\partial_{k_\beta}|n\rangle$. And the generalized derivative $r_{nm;\alpha}^{\beta} = \partial_{k_\alpha} r_{mn}^{\beta} - i(A_{nm}^{\alpha} - A_{mn}^{\alpha}) r_{nm}^{\beta}$, with $A_{nm}^{\alpha} = i\langle n|\partial_{k_\alpha}|m\rangle$ are intraband Berry connections for band $n$. The nonvanishing tensor $\sigma_{\beta\beta}^{\alpha}(\omega)$ can be deduced from $D_{3}$ point group symmetry generated by $C_{3z}$ and $C_{2y}$. According to the symmetry constraint of the $D_{3}$ point group, the non-zero elements in shift current optical conductivity tensor are $\sigma_{xx}^{\alpha\alpha} = \sigma_{yy}^{\alpha\alpha} = \sigma_{zz}^{\alpha\alpha} = -\sigma_{yx}^{\alpha\alpha}$. Without loss of generality, we display the results with $\sigma_{xx}^{\alpha\alpha}$ in the following.

Figure 3 (b) shows the photon energy dependence of the shift current photoconductivity $\sigma_{xx}^{\alpha\alpha}(\omega)$ at different PMF strength $B_0$, where the Fermi energy is in the gap between first and second moiré bands. We note that (i) the order of photoconductivity is $\sim 10^4 \mu\text{A-nm}/\text{V}^2$, which is giant and in the same order as the one in twist bilayer graphene\cite{48}; (ii) the photoconductivity curve develops two peaks and their separation increases with the PMF strength.

The two peaks stems from the concentration of Berry curvature near $K_m$ and $-K_m$ pockets, and the photon energy difference between the two peaks stems the shifting the Dirac mass by PMF in opposite way at $K_m$ and $-K_m$ pockets. In other words, the separation of two peaks can be estimated by the gap difference at $K$ and $-K$ points, which we denote as $E_d$. As shown in Supplementary Note 3, $E_d$ can be solved as

$$E_d = \frac{\hbar e B_0}{m^*} \sin(\phi_c + \frac{\pi}{6})$$

In the inside panel of Fig 3(b), we compare the peak to peak frequency difference $\Delta\hbar\omega$ between numerical result (from continuum model) and theoretical calculation (in Eq.(9)), which shows a good agreement. The $\Delta\hbar\omega$ is monochromatically linear with $B_0$ and is about $9 \text{ meV}$ when $B_0 = 50 \text{T}$, which is resolvable in a terahertz optical measurement.

Conclusions.— In a conclusion, we have studied the Berry curvature effects in heterobilayer TMD superlattice in this work. In particular, we have found that the periodic PMF plays a crucial role in affecting the Berry
curvature distribution of moiré bands. Importantly, we found that the conduction band moiré potential within a massive Dirac Hamiltonian naturally induces a periodic PMF upon the valence band. We also have pointed out how the large spin Hall effect observed in the experiment could be explained by the moiré valley-contrasting Berry curvature distribution induced by the PMF.

Furthermore, we have demonstrated the observation of a two-peak splitting in shift current photoconductivity would provide a direct evidence of periodic PMF in TMD heterobilayers. Our theoretical findings in this work is general, which can be verified via transport and optical measurements in various recent fabricated TMD heterobilayers, such as MoTe$_2$/WSe$_2$, MoSe$_2$/WSe$_2$ and MoS$_2$/WSe$_2$.

Note added.—During the preparation of this manuscript, we noticed that the PMF arising from the massive Dirac Hamiltonian was also realized in [35]. But the special role of conduction band moiré potential giving rise to this PMF and the responses related to the PMF are not discussed in [35].

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Supplementary Material for ‘Berry curvature, Spin Hall effect and nonlinear optical response in moiré transition metal dichalcogenides heterostructure’

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I. DERIVATION OF THE CONTINUUM MODEL

In this section we give the derivation of the model Hamiltonian in detail. We start from a massive Dirac model including moiré potential as the same as Eq.(1) in main text

\[ H = v_f(\tau \hat{p}_x \sigma_x + \hat{p}_y \sigma_y) + \frac{\Delta}{2} \sigma_z + \left( \begin{array}{cc} U_c(\hat{r}) & 0 \\ 0 & U_v(\hat{r}) \end{array} \right) \]  

(S1)

\( U_c, U_v \) represent the moiré potential of conduction and valence band with \( U_c(r) = 2U_c \sum_{i=1}^{3} \cos(G_i \cdot r + \phi_c), U_v(r) = 2U_v \sum_{i=1}^{3} \cos(G_i \cdot r + \phi_v) \).

\( G_1 = (0, 1)G_0, G_2 = (-\sqrt{3}/2, -1/2)G_0, G_3 = (\sqrt{3}/2, -1/2)G_0, G_0 = \frac{4\pi}{\sqrt{3}LM} \).

By using the two spinor wavefunction \( (\Psi_c, \Psi_v)^T \), the Schrödinger equation can be written in the form of two coupled equations:

\[ \begin{align*}
(\frac{\Delta}{2} + U_c(\hat{r}))\Psi_c + v_f(\tau \hat{p}_x - i\hat{p}_y)\Psi_v &= E\Psi_c \\
v_f(\tau \hat{p}_x + i\hat{p}_y)\Psi_c - (\frac{\Delta}{2} - U_v(\hat{r}))\Psi_v &= E\Psi_v
\end{align*} \]

(S2) (S3)

Since the energy gap \( \Delta \) is relatively large, we can do the approximation that \( E \approx -\Delta/2 \) after considering the states near valence band edge. Thus from Eq.(S2) we obtain:

\[ \Psi_c = -\frac{v_f}{\Delta + U_c(\hat{r})}(\tau \hat{p}_x - i\hat{p}_y)\Psi_v \]  

(S4)

Insert Eq.(S4) into Eq.(S3), we obtain:

\[ \begin{align*}
[-v_f^2(\tau \hat{p}_x + i\hat{p}_y) + 1 \overline{\Delta + U_c(\hat{r})}](\tau \hat{p}_x - i\hat{p}_y) - \frac{\Delta}{2} + U_v(\hat{r})]\Psi_v &= E\Psi_v
\end{align*} \]

(S5)

By expanding \( \frac{1}{\Delta + U_c(\hat{r})} \) to the first order, we get the effective Hamiltonian:

\[ H_{eff} = -\frac{v_f^2}{\Delta} \hat{\pi}(1 - \frac{U_c(\hat{r})}{\Delta})\hat{\pi}^\dagger - \frac{\Delta}{2} + U_v(\hat{r}) \]

(S6)

with the ladder operator \( \hat{\pi} = \tau \hat{p}_x + i\hat{p}_y \). To deal with the term \( \hat{\pi}U_c(\hat{r})\hat{\pi}^\dagger \), we first divide it into a self-hermitian operator:

\[ \hat{\pi}U_c(\hat{r})\hat{\pi}^\dagger = 1/2 * ([\hat{\pi}, U_c(\hat{r})]\hat{\pi}^\dagger + \hat{\pi}[U_c(\hat{r}), \hat{\pi}^\dagger] + U_c(\hat{r})\hat{p}_x^2 + \hat{\pi}^2 U_c(\hat{r})) \]  

(S7)

with \( \hat{\pi}^2 = \hat{\pi}_x^2 + \hat{\pi}_y^2 \). The commutation relation \( [\hat{\pi}, U_c(\hat{r})] = h(-i\tau \partial_\tau + \partial_y)U_c(\hat{r}) \). Using the plane waves \( |k\rangle = e^{ikr}, \hat{\pi}|k\rangle = (\tau \hat{p}_x + i\hat{p}_y)|k\rangle \) and \( \hat{\pi}^\dagger |k\rangle = (\tau \hat{p}_x - i\hat{p}_y)|k\rangle \).

Thus we can write the effective continuum model:

\[ H_{eff} = -\frac{v_f^2}{\Delta} \pi(1 - \frac{U_c(\hat{r})}{\Delta})\pi^\dagger - \frac{\Delta}{2} + U_v(\hat{r}) \]

\[ = -\frac{v_f^2}{\Delta}[(1 - \frac{U_c(\hat{r})}{\Delta})(\hat{p}_x^2 + \hat{p}_y^2) + 2e\mathbf{p} \cdot \mathbf{A}] + U_v(\hat{r}) - \frac{\Delta}{2} \]

(S8)
The vector potential $\mathbf{A}$ satisfies

\begin{align}
A_x &= \tau A_0 \sin(\mathbf{G}_1 \cdot \mathbf{r} + \phi_c) - \frac{1}{2} \sin(\mathbf{G}_2 \cdot \mathbf{r} + \phi_c) \\
&\quad - \frac{1}{2} \sin(\mathbf{G}_3 \cdot \mathbf{r} + \phi_c) \tag{S9}
\end{align}

and

\begin{align}
A_y &= \tau A_0 \left( \frac{\sqrt{3}}{2} \sin(\mathbf{G}_2 \cdot \mathbf{r} + \phi_c) - \frac{\sqrt{3}}{2} \sin(\mathbf{G}_3 \cdot \mathbf{r} + \phi_c) \right) \tag{S10}
\end{align}

where $A_0 = \frac{\hbar U_c G_2}{e \Delta}$.

In Eq. (S8), we find besides the kinetic energy part, the effective Hamiltonian includes a $\mathbf{p} \cdot \mathbf{A}$ like term induced by a pseudo-magnetic field. The pseudo-magnetic field is given by

\begin{align}
B(r) = \tau \frac{\hbar U_c G_2^2}{e \Delta} \sum_{i=1}^{3} \cos(\mathbf{G}_i \cdot \mathbf{r} + \phi_c) \tag{S11}
\end{align}

The result reveals that the moiré potential of conduction band triggers a gauge potential on valence band, with opposite sign in two valleys.

### II. TOPOLOGICAL PHASE TRANSITION

By further tuning the conduction band moiré potential $U_c$ to change PMF, the top two moiré bands can further exchange Berry curvature by gap closing and reopening, and undergo a topological phase transition. In Fig. S1 (a) and (b), we plot the Berry curvature distribution for $B_0 = 55T(U_c = 40meV)$ and $B_0 = 200T(U_c = 144meV)$ for the first moiré band. The chern number goes from $C = 0$ to $C = 1$.

From the effective Hamiltonian Eq. (S18), the topological phase transition occurs at $B_0 \approx 103T(U_c = 74meV)$.

### III. DERIVATION OF THREE-BAND EFFECTIVE HAMILTONIAN

We derive the three-band effective model from continuum model Eq. (S8) at the Brillouin corners (in Fig S2 (a)). First we consider the case $U_e(r) = 0$ and $U_v(r) = 0$. Because the three corners of moiré Brillouin are connected by the superlattice reciprocal vectors, using the plane waves $|k\rangle = e^{ik \cdot \mathbf{r}}$, the effective Hamiltonian near $\pm \mathbf{K}$ is written as:

\begin{align}
H_{\pm}^U(k) &= \epsilon_0 \mathbb{I} + \begin{pmatrix}
0 & 0 & 0 \\
0 & \frac{1}{2} k_x + \frac{\sqrt{3}}{2} k_y & 0 \\
0 & 0 & \frac{1}{2} k_x - \frac{\sqrt{3}}{2} k_y
\end{pmatrix} \tag{S12}
\end{align}

where $\epsilon_0 = -\hbar^2 |K|^2/(2m^*)$, $v = \hbar^2 |K|/m^*$. For the moiré potential $U_v(r)$,

\begin{align}
H_{\pm}^U(k) &= \begin{pmatrix}
0 & U_v e^{\mp i \phi_v} & U_v e^{\pm i \phi_v} \\
U_v e^{\pm i \phi_v} & 0 & U_v e^{\mp i \phi_v} \\
U_v e^{\pm i \phi_v} & U_v e^{\mp i \phi_v} & 0
\end{pmatrix} \tag{S13}
\end{align}

For the gauge field term,

\begin{align}
H_{\pm}^A(k) &= \begin{pmatrix}
0 & -\frac{g}{2} e^{\pm i \phi_c} & \frac{g}{2} e^{\mp i \phi_c} \\
\frac{g}{2} e^{\pm i \phi_c} & 0 & -\frac{g}{2} e^{\mp i \phi_c} \\
-\frac{g}{2} e^{\mp i \phi_c} & \frac{g}{2} e^{\pm i \phi_c} & 0
\end{pmatrix} \tag{S14}
\end{align}

with $g = \hbar e B_0/(2\sqrt{3}m^*)$. At the Brillouin zone corners, the eigenenergies and eigenfunctions of $H^U + H^A$ are:

\begin{align}
E_1 &= 2(U_v \cos \phi_v \mp \frac{g}{2} \sin \phi_c), \\
|\psi_1\rangle &= \frac{1}{\sqrt{3}} (|\pm K_1\rangle + |\pm K_2\rangle + |\pm K_1\rangle) \tag{S15}
\end{align}
FIG. S1: (a),(b) The band structure for $B_0 = 55T(U_c = 40meV)$ and $B_0 = 200T(U_c = 144meV)$. We set $U_v = 15meV, \phi_v = 0.3\pi, \phi_c = 0$. (c),(d) The corresponding Berry curvature.

$$E_2 = 2U_v \cos(\phi_v + \frac{2\pi}{3}) \pm g \sin(\phi_c - \frac{\pi}{3}),$$  
$$|\psi_2\rangle = \frac{i}{\sqrt{3}}(|\pm K_1\rangle + e^{\pm i \frac{2\pi}{3}}|\pm K_2\rangle + e^{\mp i \frac{2\pi}{3}}|\pm K_3\rangle)$$  

(S16)

$$E_3 = 2U_v \cos(\phi_v - \frac{2\pi}{3}) \pm g \sin(\phi_c + \frac{\pi}{3}),$$  
$$|\psi_3\rangle = \frac{-i}{\sqrt{3}}(|\pm K_1\rangle + e^{\mp i \frac{2\pi}{3}}|\pm K_2\rangle + e^{\pm i \frac{2\pi}{3}}|\pm K_3\rangle)$$  

(S17)
Thus in the basis spanned by \(|\psi_1\rangle, |\psi_2\rangle, |\psi_3\rangle\), we can write the effective model which describes the states near \(\pm K\) of the first three moiré bands:

\[
H_{\text{eff}}^\text{f}(k) = \begin{pmatrix}
2(U_v \cos \phi_c + \frac{g}{2} \sin \phi_c) & \frac{1}{2}v(k_y \pm ik_x) & \frac{1}{2}v(k_y \pm ik_x) \\
\frac{1}{2}v(k_y \pm ik_x) & 2U_v \cos(\phi_c + \frac{\pi}{4} \pm g \sin(\phi_c + \frac{\pi}{4})) & 2U_v \cos(\phi_c + \frac{\pi}{4} \pm g \sin(\phi_c + \frac{\pi}{4})) \\
\frac{1}{2}v(k_y \pm ik_x) & 2U_v \cos(\phi_c + \frac{\pi}{4} \pm g \sin(\phi_c + \frac{\pi}{4})) & \frac{1}{2}v(k_y \pm ik_x)
\end{pmatrix}
\]  

(S18)

The energy gap difference between \(K\) and \(-K\) points can be evaluated as \(E_d = 2\sqrt{3}g \sin(\phi_c + \pi/6)\). To calculate the spin Hall effect, we can only consider the first two bands and it becomes a massive Dirac model with the Fermi velocity \(v_F = v/2\) and the mass \(m_0^\pm = \sqrt{3}U_v \cos(\phi_c + \pi/6) + \frac{\sqrt{3}}{2}g \sin(\phi_c + \frac{\pi}{6})\). We can calculate the Berry curvature near \(\pm K_m\) points:

\[
\Omega_{k}^{\pm} = -\frac{s}{2} \frac{v_F^2 m_0^\pm}{2(m_0^+ + v_F^2 k^2)^{3/2}} 
\]  

(S19)

with \(s = \pm 1\) for \(\pm K\) points, and \(l = \pm 1\) for band index (+1 for upper band and -1 for lower band). By using Eq.(5) and integrating over \(K\) and \(-K\) points, we can obtain the spin valley Hall conductivity \(\sigma_{xy}^v\):

\[
\sigma_{xy}^v = \begin{cases} 
0 & 0 < |\mu| < m_0^+ \\
sgn(\mu) \frac{e^2}{2\pi \hbar} [1 - \frac{m_0^+}{|\mu|}] & m_0^+ < |\mu| < m_0^- \\
-sgn(\mu) \frac{e^2}{2\pi \hbar} \frac{m_0^- - m_0^+}{|\mu|} & m_0^- < |\mu| < m_0^-
\end{cases}
\]  

(S20)

In Fig S2(b) we plot the \(\sigma_{xy}^v\) as the function of \(\mu\) at zero temperature. And we obtain:

\[
\max(|\sigma_{xy}^v|) = \frac{e^2}{2\pi \hbar} \sqrt{3}g \sin(\phi_c + \pi/6) \\
\approx \frac{e^2}{2\pi \hbar} \sqrt{3}g \sin(\phi_c + \pi/6)
\]  

(S21)

which means in the low \(B\) field region, \(\max(|\sigma_{xy}^v|) \sim B_0 \sin(\phi_c + \pi/6)\). It is worth noting that the theoretical calculations in this part do not correspond well to numerical calculations in Fig S2(b), and it just provide a responsible explanation of the \(\sigma_{xy}^v - \mu\) curve.

IV. LINEAR OPTICAL CONDUCTIVITY

For a circular polarized light, the optical conductivity is written in terms of the longitudinal part \(\sigma_{xx}\) and transverse part \(\sigma_{xy}\):

\[
\sigma_{\pm}(\omega) = \sigma_{xx}(\omega) \pm i \sigma_{xy}(\omega) 
\]  

(S22)

where \(\pm\) for left (+1) or right (-1) circular polarization. Thus the dissipative components of the conductivity tensor is \(\text{Re}(\sigma_{xx})\) and \(\text{Im}(\sigma_{xy})\). The optical conductivity from interband transition can be calculated using standard linear response theory:

\[
\sigma_{\alpha \beta}(\omega) = -\frac{i e^2}{\hbar} \sum_{m \neq n} \int \frac{d^2 k}{(2\pi)^2} \frac{f_{k,m} - f_{k,n}}{E_{k,m} - E_{k,n}} \langle u_{k,m} | \hat{v}_\alpha(k) | u_{k,n} \rangle \langle u_{k,n} | \hat{v}_\beta(k) | u_{k,m} \rangle \\
\frac{1}{\hbar \omega + i\eta + E_{k,m} - E_{k,n}}
\]  

(S23)

Because of the time-reversal symmetry, the conductivity in the transverse direction \(\sigma_{xy} = -\sigma_{yx}\). As a result, \(\sigma_{xy}\) is spin-resolved due to the opposite spin from two valleys. We have the optical spin-valley conductivity \(\sigma_{xy}^v = \frac{1}{2}(\sigma_{xy}^{+1} - \sigma_{xy}^{-1})\).

In Fig S3(a) and (b) we plot the \(\text{Re}(\sigma_{xx})\) for different \(B_0\) and \(\phi_c\), and we find it does not depend on \(B_0\), for the longitudinal part of optical conductivity has a peak at the energy which corresponds to the mean gap of the two bands near Brillouin zone boundary. In Fig S3(c),(d) we show our results for the \(\sigma_{xy}^v\) vs. \(\hbar \omega\) in units of \(e/\hbar\) for the four values of \(B_0\) and find it zero when \(B_0 = 0\). The spinless time-reversal symmetry enforces the integral in Eq.(S23) to be zero and the \(k \cdot A\) term in the Hamiltonian will break the spinless time-reversal symmetry and results in finite optical conductivity. It is clear that \(\text{Im}(\sigma_{xy}^v)\) get enhanced as \(B_0\) increases.
V. SYMMETRY ANALYSIS OF THE NONLINEAR OPTICAL RESPONSE

In this section we discuss the symmetry properties of the shift current conductivity tensor. In the matrix form, the shift current conductivity tensor is expressed as:

$$\hat{\sigma}_c^{ab} = \begin{pmatrix} \sigma_{xx} & \sigma_{xy} & \sigma_{yx} & \sigma_{yy} \\ \sigma_{yx} & \sigma_{yy} & \sigma_{yx} & \sigma_{yy} \\ \sigma_{yx} & \sigma_{yx} & \sigma_{xx} & \sigma_{yy} \\ \sigma_{yy} & \sigma_{yx} & \sigma_{yx} & \sigma_{xx} \end{pmatrix}$$  \hspace{1cm} (S24)

In a symmetry operator $g$, $\hat{\sigma}_c^{ab}$ is transformed as:

$$\hat{\sigma}_c^{ab} \rightarrow \hat{U}^\dagger(g) \hat{\sigma}_c^{ab} \hat{U}(g) \otimes \hat{U}(g)$$  \hspace{1cm} (S25)

In $C_3$ rotation, $\hat{U}(C_3) = e^{i\frac{2\pi}{3}} \sigma_y$, it enforces:

$$\sigma_{xx} = \sigma_{yy} = \sigma_{yx} = \sigma_{xy} = -\sigma_{xx}$$  \hspace{1cm} (S26)

$$\sigma_{yy} = \sigma_{yx} = \sigma_{xy} = -\sigma_{yy}$$  \hspace{1cm} (S27)

Similarly, in $C_{2y}$ rotation symmetry, it enforces:

$$\sigma_{xx} = \sigma_{yy} = \sigma_{yx} = \sigma_{xy} = -\sigma_{xx} = 0;$$  \hspace{1cm} (S28)

Thus the only nonzero and nonequivalent term in $\hat{\sigma}_c^{ab}$ is $\sigma_{yx}^c$, which is calculated in the main text.