Intrinsic Spin and Orbital Hall Effects from Orbital-Dependent Level Splitting

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We show theoretically that orbital-dependent level splitting can generate both intrinsic spin Hall effect (SHE) and intrinsic orbital Hall effect (OHE) in centrosymmetric systems. This mechanism is stable against orbital quenching and applicable to a wide class of multi-orbital systems. OHE occurs even without the spin-orbit coupling (SOC). Its main role is to convert OHE into SHE. The conversion efficiency varies with the SOC strength in a nonmonotonic way. The resulting spin and orbital Hall conductivities can be large. This work suggests a path for an ongoing search for stronger SHE. It also calls for experimental efforts to probe orbital degrees of freedom in OHE and SHE. Possible ways for experimental detection are briefly discussed.

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Spin Hall effect (SHE) [1–5] is a phenomenon in which an external electric field \( \mathbf{E} \) generates a spin current in a transverse direction. Recent experiments [6, 7] demonstrated that when a spin current generated by SHE is injected to a neighboring ferromagnetic layer, it can even switch its magnetization direction. SHE is now regarded as an indispensable tool in spintronics to generate and detect a spin current [4, 5]. Of particular interest is intrinsic mechanisms [8–12], which do not rely on impurity scattering. Large SHE in 5d transition metals such as Pt is believed to be intrinsic [11–17].

The spin-orbit coupling (SOC) is a crucial element for intrinsic SHE and has sizable value only near atomic nuclei [18], where it can be approximated as

\[
H_{so} = \frac{2\alpha_{so}}{\hbar^2} \mathbf{S} \cdot \mathbf{L}.
\]

Here, \( \mathbf{L} \) denotes the orbital angular momentum near nuclei. Since the spin \( \mathbf{S} \) couples to other degrees of freedom only through Eq. (1) (at least in non-interacting systems without magnetism), \( \mathbf{L} \) is expected to play important roles for SHE. Although an orbital degree of freedom is taken into account in equilibrium band structure calculations, dynamical roles of \( \mathbf{L} \) are commonly ignored in literature. Only for a limited class of systems, it was argued [11, 12, 19, 20] that an Aharonov-Bohm phase generated by orbitals is responsible for SHE and that SHE is closely related to orbital Hall effect (OHE). Here OHE refers to an \( \mathbf{E} \)-induced transverse flow of \( \mathbf{L} \). Even for these materials, however, there is no experiment that probes roles of \( \mathbf{L} \) as far as we are aware of. It is partly due to the expectation that \( \mathbf{L} \) cannot play any important roles due to orbital quenching [21] in solids.

In this Letter, we demonstrate that in centrosymmetric systems with orbital-dependent level splitting, nonzero \( \mathbf{L} \) is dynamically generated by \( \mathbf{E} \) (even when \( \mathbf{L} \) is quenched in equilibrium) and this \( \mathbf{L} \) gives rise to intrinsic SHE and OHE. This demonstration (sketched in Fig. 1) provides not only an alternative theoretical picture to understand the SHE and OHE in Refs. [11, 12, 19] but also implies that many other systems may exhibit SHE and OHE since orbital-dependent level spacing is ubiquitous in multi-orbital systems. Specifically we demonstrate two points; (i) even when the SOC is absent and \( \mathbf{L} \) is completely quenched in equilibrium by crystal field splitting, orbital-dependent level splitting generates OHE universally. (ii) When the SOC is sizable, the OHE is efficiently converted into SHE. Thus OHE is more fundamental than SHE in this mechanism. Interestingly we find that the SOC being stronger does not necessarily imply stronger SHE. This result is relevant for ongoing search for materials with strong SHE.

We begin with an illustration of the point (i) for a \( p \)-orbital system [Fig. 1(a)], the simplest multi-orbital system. We assume \( \alpha_{so} = 0 \) since the SOC is not essential for (i). We
also assume that all orbital degeneracy is lifted due to crystalfield splitting and the expectation value of \( L \) is suppressed to zero for all eigenstates. Nevertheless orbital character may still vary from bands to bands and also with the crystal moment \( k \). Such orbital-dependent level splitting is the only requirement for our mechanism. For concreteness, we assume that for \( k = |k| (\cos \phi, \sin \phi, 0) \) in the \( k_x, k_y \) plane, the wavefunction in the lower(upper) band has tangential(radial) \( p \)-orbital character, that is, \( |u_{1,k} \rangle \sim |p_{\phi+\pi/2} \rangle \) \( |u_{2,k} \rangle \sim |p_\phi \rangle \), where \( |p_\phi \rangle \equiv \cos \phi |p_x \rangle + \sin \phi |p_y \rangle \). Here, \( |u_{1(2),k} \rangle \) is the periodic part of the Bloch wavefunction of the lower(upper) band. Note that the expectation value \( \langle L \rangle \) vanishes for these states. Figure 1(b) shows schematically the wavefunction character of eigenstates in the lower band at the Fermi surface. Suppose \( E \) is applied to the \(-x\) direction. Then \( k \) is shifted in the \(+x\) direction to \( k + \delta k = |k + \delta k| (\cos(\phi + \delta \phi), \sin(\phi + \delta \phi), 0) \), where \( \delta \phi \) is negative(positive) for positive(negative) \( k_y \). Under this \( k \) shift, \( |u_{1,k} \rangle \sim |p_{\phi+\pi/2} \rangle \), which can be written as \( |p_{\phi+\delta \phi+\pi/2} \rangle + \delta \phi |p_{\phi+\delta \phi} \rangle \), evolves to \( \exp(-iE_{1,k+\delta k} \delta \phi/h) |u_{1,k+\delta k} \rangle \). Thus, interband hybridization is induced. Note that the ratio between the two coefficients of the states \( |u_{1(2),k+\delta k} \rangle \) is complex, which is important since it makes the expectation value \( \langle L \rangle \) of the hybridized state nonzero. Thus, even when \( L \) is completely quenched in equilibrium, dynamically induced hybridized states have nonzero \( \langle L \rangle \). An explicit calculation results in \( \langle L \rangle \times \delta \phi \delta \phi \). Recalling that \( \delta \phi \) has opposite signs for positive and negative \( k_y \)'s, the sum of \( \langle L \rangle \) over all occupied hybridized states may vanish. However the orbital Hall current \( \sum \langle L \rangle \) can be finite. This illustrates an intrinsic mechanism of OHE based on the orbital-dependent level splitting. Similar demonstrations are possible for \( d \)- and \( f \)-orbital systems.

Next we restore the SOC. Then due to the correlation between \( L \) and \( S \), nonzero \( \langle L \rangle \) for a hybridized state in non-equilibrium implies \( \langle S \rangle \) to be nonzero. Thus the SOC generates SHE as a concomitant effect of OHE. The sign of the spin Hall conductivity (SHC) is the same or opposite to that of the orbital Hall conductivity (OHC) depending on whether the correlation is positive or negative (that is, \( S \) is parallel or antiparallel to \( L \)) [Figs. 1(c), (d)].

To examine the intuitive picture in Fig. 1 quantitatively, we adopt a tight-binding model description of a very simple system, a simple cubic lattice with four orbitals \( s \), \( p_x \), \( p_y \), and \( p_z \) at each lattice point [Fig. 2(a)]. Possible nearest-neighbor hoppings are shown in Fig. 2(a) with their hopping amplitudes (see Ref. [22] for details of the system). Figure 2(b) shows the band structure of this system. The three doubly-degenerate lower bands have \( p \)-character whereas the topmost doubly-degenerate band (with \( \Gamma \) point band edge at 0.3 eV) has \( s \)-character. The color in Fig. 2(b) represents the expectation value of \( L \cdot S \) for each eigenstate. The \( p \)-character bands have negative(positive) correlation between \( S \) and \( L \) in the lower(higher) energy branches due to the SOC. Figure 2(c) shows the orbital character of the states at \( E = -0.8 \) eV with \( k_z = 0 \). The inner(outer) band has radial(tangential) character, confirming the presence of orbital-dependent level splitting. In this simple system, it arises from the \( sp \) hybridization and vanishes if the hopping amplitude \( t_{sp} \) between \( s \) and \( p \) orbitals vanishes. Numerical values of the Hamiltonian parameters are (unless mentioned otherwise) as follows; \( E_s = 3.2 \), \( E_{px} = E_{py} = E_{pz} = -0.5 \) for on-site energies, \( t_s = 0.5 \), \( t_{px} = 0.5 \), \( t_{py} = 0.2 \), \( \gamma_{sp} = 0.5 \) for nearest-neighbor hopping amplitudes, and \( \alpha_{SO} = 0.1 \) for SOC, all in unit of eV.

The OHC \( \sigma_{OHE} \) and the SHC \( \sigma_{SH} \) are calculated as a function of the Fermi energy \( E_F \) by using the Kubo formula:

\[
\sigma_{OHE} = \frac{e}{h} \sum_{n \neq m} \int \frac{d^3k}{(2\pi)^3} (f_{mk} - f_{nk}) \Omega_{nmk}^X, \quad (2a)
\]

\[
\Omega_{nmk}^X = \hbar^2 \text{Im} \left[ \frac{\langle u_{nk} | j_x^X_S | u_{mk} \rangle \langle u_{mk} | v_x | u_{nk} \rangle}{(E_{nk} - E_{mk} + i\eta)^2} \right], \quad (2b)
\]

where \( j_x^X_S = (v_y X_z + X_z v_y) / 2 \) is the conventional orbital(spin) current operator with \( X_z = L_z (S_z) \), \( v_x(y) \) is the velocity operator along the \( x(y) \) direction, and \( f_{nk} \) is the Fermi-Dirac distribution function. In view of the picture in Fig. 1, \( \Omega_{nmk}^X \) may be regarded as the contribution to \( \sigma_{OHE} \) from the hybridization between the bands \( n \) and \( m \). Figure 3 shows the calculated \( \sigma_{OHE} \) as a function of \( E_F \). Note that both \( \sigma_{OHE} \) [Fig. 3(a)] and \( \sigma_{SH} \) [Fig. 3(b)] vanish for \( \gamma_{sp} = 0 \) and grow with increasing \( \gamma_{sp} \), as expected from the intuitive picture in Fig. 1(b). Interestingly \( \sigma_{OHE} \) does not vary much with \( \alpha_{SO} \) and stays finite even when \( \alpha_{SO} = 0 \) [Fig. 3(c)]. On the other hand, \( \sigma_{SH} \) vanishes for \( \alpha_{SO} = 0 \) and grows with in-
This implies efficient dynamical generation of $L$ and $S$ saturates once a system enters the strong SOC regime and the enhanced SOC may reduce $\sigma_{\text{OH}}$ [Fig. 3(c)]. Considering that $\alpha_{\text{so}}$ grows rapidly with the atomic number $Z$, this result may be relevant for large $Z$ materials (see Discussion), implying that materials with overly strong SOC may not be good choices for ongoing efforts to achieve large SHC.

We emphasize that large OHC and SHC remain stable against crystal field splitting. To demonstrate this, we shift the on-site energies of $p_x$ and $p_y$ orbitals by $\pm \Delta_d$ and calculate $\sigma_{\text{OH/SH}}$ as a function of $\Delta_d$. Figures 3(e) and (f) show that $\Delta_d$ does not crucially affect $\sigma_{\text{OH/SH}}$ even for $\Delta_d$ as large as 0.3 eV. Considering that the picture in Fig. 1(a) assumes the strongly quenched orbital regime, this stability against $\Delta_d$ is understandable.

An important technical issue is the stability of OHE and SHE against impurity scattering, which may significantly modify $\sigma_{\text{OH/SH}}$ through the so called vertex correction [23]. For systems with the inversion symmetry, on the other hand, the vertex corrections for $\sigma_{\text{SH}}$ and $\sigma_{\text{OH}}$ vanish in the weak scattering limit due to the symmetry [11, 24, 25]. We thus expect that our results for centrosymmetric systems remain stable in the weak scattering regime.

Next we compare our work with other theoretical works. Reference [9] on two-dimensional Rashba systems reports that spin-dependent level splitting generates an interband hybridization upon the application of $E = E_x \hat{x}$ and $\langle S_z \rangle$ for the hybridized state has opposite signs for opposite signs of $k_y$’s. This mechanism (Fig. 1 of Ref. [9]) is thus very similar to ours (Fig. 1 of this Letter) and our work may be regarded as an orbital counterpart of Ref. [9]. There are clear differences, however. Reference [9] completely ignores the orbital degree of freedom and is applicable only to noncentrosymmetric systems, whereas ours is applicable to centrosymmetric systems.

References [11, 12] on 4$d$ and 5$d$ transition metals report that the inter-atomic hopping between $s$ and $d$ orbitals is crucial for OHE and SHE, of which dependences on the hopping strength and $\alpha_{\text{so}}$ closely resemble the results in Figs. 3(a)-(d). This indicates that results in Refs. [11, 12] can be explained by the orbital-dependent level splitting picture (Fig. 1). We argue that this picture provides a more detailed microscopic understanding (such as $k$-resolved contributions [22] to OHC and SHE) than the Aharonov-Bohm phase picture proposed by Refs. [11, 12] since the former/latter picture provides a momentum(real)-space description.

References [8, 17, 26, 27] studied SHE in hole-doped semiconductors. Whereas these studies did not recognize the importance of $L$, their results can be easily understood by the orbital-dependent level splitting picture (the Aharonov-Bohm phase picture, on the other hand, appears to be difficult to apply to this system). A crucial point is that for the semiconductors in the diamond or the zinc blende structure, the inter-atomic hopping between $p$ orbitals themselves induces rapid $k$-dependent variation of $|u_{nk}|$ near $k = 0$, where $p$-character bands touch each other. Then together with the energy separation between the $p$-character bands away from the band

![FIG. 3. (Color online) The Fermi energy ($E_F$) dependence of the OHC $\sigma_{\text{OH}}$ (a,c,e) and the SHC $\sigma_{\text{SH}}$ (b,d,e) for different values of (a,b) the inter-orbital hopping amplitude $\gamma_{sp}$ between $s$ and $p$ orbitals, (c,d) the SOC strength $\alpha_{\text{so}}$, and (e,f) the crystal field splitting strength $\Delta_d$, respectively.](image-url)
touching point, all requirements of the orbital-dependent level splitting picture are satisfied. Thus when \( E_\gamma \) is near the band touching point, both OHC and SHC should grow as the separation between the \( p \)-character bands grows. This explains the OHC in the spinless limit [24] and the SHC in the opposite limit [26], where \( L \) is completely locked to \( S \).

The Berry curvature [28] is a commonly invoked concept to analyze intrinsic Hall effects. Reference [8] attributed SHE in hole-doped semiconductors to the non-Abelian \( k \)-space Berry curvature that diverges at the band touching point. Here we calculate the \( k \)-space Berry curvature for the \( sp \) system in a simple limiting case, \( \alpha_{sp} = 0 \) [29] and \( \Delta_{sp} = 0 \), for which three \( p \)-character bands touch each other at \( k = 0 \). For the \( p \) character bands, we obtain [22] the Berry curvature

\[
\Omega^{(p)}(k) = -2\lambda_{sp}L^{(p)} + O(k)^2,
\]

where \( \lambda_{sp} \equiv a^2\gamma_{sp}/2\hbar E_g^2 \); \( a \) is the lattice spacing of the cubic lattice in Fig. 2(a), \( E_g \) is the band gap between the \( s \)-character band and the \( p \)-character sub-Hilbert space. Note that \( \Omega^{(p)}(k) \) does not diverge at the 3-fold band touching point \( (k = 0) \). Thus the band touching within \( p \)-character bands is not important. It is instead the hybridization between \( s \)- and \( p \)-character orbitals, of which strength is governed by \( \gamma_{sp} \) and \( E_g \). Note also that \( \Omega^{(p)}(k) \) is collinear to \( L^{(p)} \), which implies intrinsic tendency towards OHE.

Here we emphasize that \( \Omega^{(p)}(k) \) is non-Abelian since it is defined within the sub-Hilbert space spanned by the three \( p \)-character bands. Its non-Abelian nature manifests itself in its matrix representation. When the three real orbitals \( |x⟩ \), \( |y⟩ \), \( |z⟩ \) are used as bases of \( |u_{mk}⟩ \)'s in the \( p \)-character sub-Hilbert space, the matrix representations of the \( L^{(p)} = (L_x^{(p)}, L_y^{(p)}, L_z^{(p)}) \) in Eq. (3) become

\[
\frac{L_x^{(p)}}{\hbar} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ i & 0 & 0 \end{pmatrix}, \quad \frac{L_y^{(p)}}{\hbar} = \begin{pmatrix} 0 & i & 0 \\ -i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \frac{L_z^{(p)}}{\hbar} = \begin{pmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},
\]

which satisfy the usual angular momentum commutation relations. The non-Abelian nature of the Berry curvature is a consequence of symmetries since Abelian Berry curvatures are forced to vanish [30] by the combination of the space inversion and the time-reversal symmetries. Only non-Abelian Berry curvatures can survive the symmetry constraints. When real wavefunctions are used as bases of representations as in Eq. (4), the symmetries force only diagonal components of the \( k \)-space Berry curvature to vanish and off-diagonal components are free from such constraints. It is exactly these off-diagonal components of \( \Omega^{(p)}(k) \) that generate intrinsic OHE. This can be made manifest in \( \Omega_{mnk}^{(O)} \) [Eq. (2b)]. When \( m \) denotes the \( s \)-character band and \( n \) one of the \( p \)-character bands, one obtains [22] near \( k = 0 \),

\[
\Omega_{mnk}^{(O)} \approx \frac{\hbar}{4} \sum_{\mu,\nu=x,y,z} \text{Re} \left[ \langle u_{nk}|p_{\mu}⟩ \left( L_z^{(p)}\Omega^{(p)} \right)_{\mu\nu} |p_{\nu}|u_{nk}⟩ \right].
\]

**Discussion.**— Since orbital and spin currents are not directly measurable, predictions in this Letter should be probed through accumulations of the spin and orbital moments at edges of systems [31]. The magneto-optical Kerr effect is used in Refs. [32, 33] to probe accumulated magnetic moments at edges. To differentiate the orbital and spin accumulations, one may utilize the x-ray magnetic circular dichroism [34, 35].

Since orbital(spin) is not conserved, the connection between edge accumulation and \( \sigma_{OH(SH)} \) is not straightforward and there is ongoing discussion [36] on this connection. To assess this connection, we calculate the non-equilibrium orbital(spin) density generated by \( E \) as a function of position for a finite size system. We verify [22] that two opposite edges have opposite signs of orbital(spin) accumulations and for a given edge, the orbital(spin) accumulations at different \( E \) values \((-1.3 \text{ and } +0.0 \text{ eV})\) respectively have the opposite(same) signs. This result agrees qualitatively with the behaviors of \( \sigma_{OH(SH)} \) in Fig. 3. However further study is needed to clarify the bulk-boundary connection, which goes beyond the scope of this Letter.

In addition to the \( sp \) hybridized system in a simple cubic lattice, we perform calculations for other orbital hybridizations in other centrosymmetric crystal structures and verify similar results [37]: \( \sigma_{OH} \) of order \( 10^3 \left( \hbar/2|e| \right) \left( \Omega \cdot \text{cm} \right)^{-1} \) is rather ubiquitous and \( \sigma_{SH} \) does not monotonically increase with \( \alpha_{so} \). We thus argue this mechanism to be applicable to various centrosymmetric systems.

To conclude, we demonstrated that orbital-dependent level splitting in multi-orbital centrosymmetric systems can generate OHE, which is then converted to SHE by the SOC. The conversion efficiency depends on the SOC strength in a non-monotonic way, which provides one possible explanation why experiments [see Table III in Ref. [38] and Ref.[39]] on \( f \) orbital rare-earth systems with very strong SOC find \( \sigma_{SH} \) to be only \( 100 \sim 200 \left( \hbar/2|e| \right) \left( \Omega \cdot \text{cm} \right)^{-1} \), which is about one order of magnitude smaller than that for Pt. According to our preliminary calculation [37], systems with much weaker SOC such as vanadium can have \( \sigma_{SH} \sim 200 \left( \hbar/2|e| \right) \left( \Omega \cdot \text{cm} \right)^{-1} \), which is converted from exceptionally large \( \sigma_{OH} \sim 10^4 \left( \hbar/2|e| \right) \left( \Omega \cdot \text{cm} \right)^{-1} \) by weak SOC. Considering the importance of orbital hybridization, we argue that stronger OHE and SHE may be realized in binary compounds, in which orbitals of different character from different atomic elements share similar energies and generate strong orbital hybridization. Our result calls for experimental efforts to probe dynamically generated \( L \) in materials with strong OHE or SHE.

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Supplementary Information for
“Intrinsic Spin and Orbital Hall Effects from Orbital-Dependent Level Splitting”

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Tight-binding model

In this section, detailed information is presented on the tight-binding model considered in the Letter. We assume that there are s, px, py, pz orbitals at each Bravais lattice in the simple cubic structure. By using Bloch-like states

$$|\varphi_{n\sigma k}\rangle = \sum_R e^{i k \cdot (R - r)} |\varphi_{n\sigma R}\rangle,$$

(S1)
as basis, where n = s, px, py, pz is the orbital character and \(\sigma = \uparrow, \downarrow\) is the spin, the tight-binding Hamiltonian in k-space is written as

$$H_{\text{tot}}(k) = H_{\text{kin}}(k) + H_{\text{so}},$$

(S2)

where \(H_{\text{kin}}(k)\) is the kinetic part of the Hamiltonian arising from the hoppings and \(H_{\text{so}}\) describes the spin-orbit coupling (SOC) near the atomic nucleus. First, \(H_{\text{kin}}(k)\) is independent of the spin and its nonzero matrix elements are

$$\langle \varphi_{\sigma k'} | H_{\text{kin}} | \varphi_{\sigma k}\rangle = E_s - 2t_s \left[ \cos(k_x a) + \cos(k_y a) + \cos(k_z a) \right],$$

(S3)

$$\langle \varphi_{p x\sigma k'} | H_{\text{kin}} | \varphi_{p y\sigma k}\rangle = E_{px} + 2t_{px} \cos(k_x a) - 2t_{py} \cos(k_y a) + \cos(k_z a),$$

(S4)

$$\langle \varphi_{p y\sigma k'} | H_{\text{kin}} | \varphi_{p x\sigma k}\rangle = E_{py} + 2t_{py} \cos(k_y a) - 2t_{px} \cos(k_z a) + \cos(k_x a),$$

(S5)

$$\langle \varphi_{p z\sigma k'} | H_{\text{kin}} | \varphi_{p \sigma k}\rangle = E_{pz} + 2t_{pz} \cos(k_z a) - 2t_{pr} \cos(k_x a) + \cos(k_y a),$$

(S6)

$$\langle \varphi_{\sigma k'} | H_{\text{kin}} | \varphi_{p \sigma k}\rangle = 2\gamma_{sp} \sin(k_x a),$$

(S7)

$$\langle \varphi_{\sigma k'} | H_{\text{kin}} | \varphi_{p \sigma k}\rangle = 2\gamma_{sp} \sin(k_y a),$$

(S8)

$$\langle \varphi_{\sigma k'} | H_{\text{kin}} | \varphi_{p \sigma k}\rangle = 2\gamma_{sp} \sin(k_z a).$$

(S9)

Here, \(E_s\) and \(E_{pi}\) (\(i = x, y, z\)) are on-site energies for s and pi orbitals, respectively, and \(t_s, t_{ps(\pi)}, \gamma_{sp}\) are the nearest hopping amplitudes between s orbitals, between p orbitals via \(\sigma(\pi)\) bonding, and between s and p orbitals, respectively. Second, the SOC is

$$H_{\text{so}} = \frac{2\alpha_{so}}{\hbar^2} L \cdot S,$$

(S10)

where \(L(S)\) is the orbital/spin angular momentum operator. The orbital angular momentum operators are defined within p orbitals, whose matrix representations are

$$L_x = \hbar \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{pmatrix}, \quad L_y = \hbar \begin{pmatrix} 0 & 0 & i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{pmatrix}, \quad L_z = \hbar \begin{pmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

(S11)

by using the basis states \(\varphi_{p x\sigma k}, \varphi_{p y\sigma k}, \text{ and } \varphi_{p z\sigma k}\). The spin angular momentum operators are represented by the Pauli matrices within each orbital:

$$\langle \varphi_{n\sigma k} | S | \varphi_{n'\sigma' k}\rangle = \frac{\hbar}{2} [\sigma],$$

(S12)

Values of the parameters used in the Letter are

\[ E_s = 3.2, \ E_p = E_{px} = E_{py} = E_{pz} = -0.5, \ t_s = 0.5, \ t_{ps} = 0.5, \ t_{pr} = 0.2, \ \gamma_{sp} = 0.5, \ \alpha_{so} = 0.1, \]

(all in the unit of eV). All parameters are set as above unless specified otherwise, such as in Fig. 3. For Figs. 3(e) and 3(f) showing the crystal field splitting dependence, on-site energies are set as \(E_{px} = E_p + \Delta_c f, E_{py} = E_p - \Delta_c f, E_{pz} = E_p\).

k-resolved orbital and spin Hall conductivities in the band structure

In the Letter, we argued that spin Hall effect (SHE) is a secondary effect of orbital Hall effect (OHE) by the SOC. The sign of the spin Hall conductivity (SHC) is the same(opposite) to that of the orbital Hall conductivity (OHC) if the correlation
By comparing with Eq. (2) in the Letter, we find that

\[ \langle \mathbf{L} \cdot \mathbf{S} \rangle \] (shown in Fig. 2(b)) is positive(negative). In order to compare each contribution in the band structure, we define k- and band-resolved contribution \( \sigma_{nk}^{\text{OH(SH)}} \) as

\[ \sigma_{nk}^{\text{OH(SH)}} = \sum_n \int \frac{d^3k}{(2\pi)^3} f_{nk} \sigma_{nk}^{\text{OH(SH)}}. \]  

(S14)

By comparing with Eq. (2) in the Letter, we find that

\[ \sigma_{nk}^{\text{OH(SH)}} = -2e\hbar \sum_{m \neq n} \text{Im} \left[ \frac{\langle u_{nk} | j_{y}^{X_{z}} | u_{mk} \rangle \langle u_{mk} | v_{z} | u_{nk} \rangle}{(E_{nk} - E_{mk} + i\eta)^2} \right], \]  

(S15)

where \( j_{y}^{X_{z}} = (v_{y}X_{z} + X_{z}v_{y})/2 \) is the conventional orbital(spin) current with \( X_{z} = L_{z}(S_{z}). \) In Fig. S1(a) and S1(b), the OHC and SHC are shown in color on top of the band structures, respectively. It can be seen that this interpretation holds for \( \Gamma \) and \( \text{H}. \) This qualitatively explains the correlation between OHC and SHC, although there are quantitative deviations in sign-changing position of \( \sigma_{nk}^{\text{OH(SH)}}. \)

Non-Abelian Berry curvature from the \( sp \) orbital hybridization

In this section, we explain how the \( sp \) orbital hybridization gives rise to a non-Abelian Berry curvature in k-space. We use the quasi-degenerate perturbation theory explained in Refs. [S1, S2]. In the \( sp \) model, the Hamiltonian in k-space is formally written as

\[ H_{\text{eff}}(\mathbf{k}) = \begin{pmatrix} H_{p}(\mathbf{k}) & h(\mathbf{k}) \\ \bar{h}^{\dagger}(\mathbf{k}) & H_{s}(\mathbf{k}) \end{pmatrix}, \]  

(S16)

where \( H_{p(s)}(\mathbf{k}) \) is the Hamiltonian within a subspace spanned by \( p(s) \) orbitals, and \( h(\mathbf{k}) \) describes the \( sp \) hybridization. The Berry phase effect becomes manifest by projecting the dynamics in the ground state within the adiabatic approximation. This can be achieved by applying a unitary operator \( U(\mathbf{k}) \) which eliminates the \( sp \) hybridization:

\[ H_{\text{eff}}(\mathbf{k}) = U(\mathbf{k})H(\mathbf{k})U^{\dagger}(\mathbf{k}) = \begin{pmatrix} H_{p,\text{eff}}(\mathbf{k}) & 0 \\ 0 & H_{s,\text{eff}}(\mathbf{k}) \end{pmatrix}. \]  

(S17)

Now, the \( sp \) hybridization is effectively included in \( H_{p(s),\text{eff}}(\mathbf{k}) \). Moreover, it is important to note that the canonical position operator transforms as

\[ \mathbf{r} \mapsto \mathbf{R} = U(\mathbf{k})\mathbf{r}U^{\dagger}(\mathbf{k}) = \mathbf{r} + \mathcal{A}(\mathbf{k}), \]  

(S18)

where \( \mathcal{A}(\mathbf{k}) = iU(\mathbf{k})\partial_{\mathbf{k}}U^{\dagger}(\mathbf{k}) \) is the Berry connection arising from the \( sp \) hybridization. In a perturbative regime where \( |h(\mathbf{k})| \ll E_{g} \), where \( E_{g} = E_{p}(\mathbf{k}) - E_{s}(\mathbf{k}) \) is the energy gap between \( s \) and \( p \) bands, the Berry connection in the \( p \) orbital subspace can be calculated as

\[ \mathcal{A}^{(p)}(\mathbf{k}) = \frac{i}{2} \left[ h(\mathbf{k}) \frac{1}{E_{g}} \{ \partial_{\mathbf{k}}h(\mathbf{k}) \} \right] = \frac{1}{E_{g}} h^{\dagger}(\mathbf{k}). \]  

(S19)
The assumption above always holds near the high-symmetry points since
\[ h(k) = -2i\gamma_{sp} [\sin(k_x a), \sin(k_y a), \sin(k_z a)]^T. \tag{S20} \]

Similarly, the Berry connection in the \(s\) orbital subspace can be found by interchanging \(h(k)\) and \(h^\dagger(k)\).

In the absence of the SOC, the Berry connection near the \(\Gamma\)-point in the \(p\) orbital subspace is found as
\[ \mathbf{A}^{(p)}(k) = \lambda_{sp} k \times \mathbf{L}^{(p)}, \tag{S21} \]
where \(\lambda_{sp} = a^2 \gamma_{sp}^2 / 2\hbar E_g^2\). On the other hand, the Berry connection in the \(s\) orbital subspace is zero because there is no internal degree of freedom in the absence of the SOC. The corresponding Berry curvature is
\[ \Omega_{\gamma}(k) = \frac{1}{2} \varepsilon_{\alpha\beta\gamma} F_{\alpha\beta}(k), \tag{S22} \]
where
\[ F_{\alpha\beta}(k) = \partial_{k_\alpha} A_\beta(k) - \partial_{k_\beta} A_\alpha(k) - i [A_\alpha(k), A_\beta(k)]. \tag{S23} \]

This leads to Eq. (3) in the Letter. This procedure is schematically shown in Fig. S3. The effect of the \(sp\) hybridization can be effectively described as an interaction with an emergent gauge field or the Berry connection after the downfolding the \(sp\) hybridization.

In the presence of the SOC, \(p\) bands splits into \(j = 3/2\) and \(j = 1/2\) multiplets. By downfolding the Hamiltonian in each multiplet, we find that the Berry connection becomes
\[ \mathbf{A}^{(j)}(k) = \lambda_{sp}^{(j)} k \times \mathbf{J}^{(j)}, \tag{S24} \]
where \(\mathbf{J} = \mathbf{L} + \mathbf{S}\) is the total angular momentum, and the superscript \(j\) represents its projection to \(j = 3/2\) or \(j = 1/2\) multiplet. The proportionality constant slightly changes from the \(\lambda_{sp}\) obtained in the absence of the SOC:
\[ \lambda_{sp}^{(j)} = \lambda_{sp} \frac{E_g^2}{E_g + \alpha_{so} \{j(j + 1) - 11/4\}^2}. \tag{S25} \]

Another important consequence of the SOC is that it gives rise to the nonzero Berry connection in \(s\) band as well. By applying the same procedure as above, it is found as
\[ \mathbf{A}^{(s)} = \lambda_{sp}^{(s)} \mathbf{S} \times \mathbf{k}, \tag{S26} \]
where
\[ \lambda_{sp}^{(s)} = \frac{2}{3} (\lambda_{sp}^{3/2} - \lambda_{sp}^{1/2}), \tag{S27} \]
thus it vanishes in the absence of the SOC. For this limit of strong SOC, similar result was also found from the Kane model for semiconductors [S1–S4].

FIG. S2. Emergence of the Berry connection in an effective theory. After downfolding, the \(sp\) hybridization can be effectively described as interactions with Berry connections in each subspace.
Relation between the non-Abelian Berry curvature and orbital Hall conductivity

In this section, we demonstrate a relation between the non-Abelian Berry curvature described in the previous section and the OHC near the high-symmetry point such as $\Gamma$, which is shown in Eq. (5) in the Letter. Near the $\Gamma$-point, Bloch states have almost pure $s$- and $p$-characters, and the velocity operator is written as

$$v_{x(y)} \approx \frac{2i\gamma_{sp}a}{\hbar} \left( |\varphi_{p_{x(y)}k}| \langle \varphi_{sk} | - |\varphi_{p_{x(y)}k}| \langle \varphi_{sk} | \right). \quad (S28)$$

Then we expand

$$\Omega_{nmk}^{L_z} = \hbar^2 \text{Im} \left[ \frac{\langle u_{nk} | L_z | u_{mk} \rangle \langle u_{mk} | v_y | u_{nk} \rangle}{(E_{nk} - E_{mk} + i\eta)^2} \right] \quad (S29)$$

in the lowest order in $k$ for band indices $n$ and $m$ denoting one of $p$-character bands and $s$-character band, respectively. Note that the $v_{x(y)}$ couples $s$- and $p$-character bands, while $L_z$ couples two $p$-character bands. Thus, it follows that

$$\Omega_{nmk}^{L_z} \approx \frac{\hbar^2}{2} \sum_{l \in p_x, p_y, p_z} \text{Im} \left[ \frac{\langle u_{nk} | L_z | u_{lk} \rangle \langle u_{lk} | v_y | u_{mk} \rangle \langle u_{mk} | v_x | u_{nk} \rangle}{(E_{nk} - E_{mk} + i\eta)^2} \right]. \quad (S30)$$

Also, the energy eigenvalues for all $p$-character bands are the same at the $\Gamma$-point, then

$$\Omega_{nmk}^{L_z} \approx \frac{\hbar^2}{2} \sum_{l \in p_x, p_y, p_z} \text{Im} \left( \frac{\langle u_{nk} | L_z | u_{lk} \rangle \langle u_{lk} | v_y | u_{mk} \rangle \langle u_{mk} | v_x | u_{nk} \rangle}{(E_{lk} - E_{mk} + i\eta)(E_{nk} - E_{mk} + i\eta)} \right) = \frac{1}{2} \sum_{l \in p_x, p_y, p_z} \text{Im} \left( \langle u_{nk} | L_z | u_{lk} \rangle \langle \partial_{k_y} u_{lk} | u_{mk} \rangle \text{Im} \langle u_{mk} | \partial_{k_y} u_{nk} \rangle \right) = \frac{1}{4} \sum_{l \in p_x, p_y, p_z} \text{Re} \left[ \langle u_{nk} | L_z | u_{lk} \rangle \Omega_{z,ln}^{(p)} (k) \right]. \quad (S33)$$

This proves Eq. (5) in the Letter.

Boundary accumulation of the orbital and spin moments

Since the orbital and spin currents are not directly measurable in the experiment, we calculate the edge accumulations for the orbital and spin moments in a finite system from the Kubo formula. We considered a film structure with its thickness of 40 atomic layers. In Fig. S4, current-induced orbital and spin moments are shown for Fermi energies $E_F = -1.30$ eV and $E_F = 0.00$ eV. While the spin and orbital accumulations have the opposite signs for $E_F = -1.30$ eV [Fig. S4(a)], the signs are the same for $E_F = 0.00$ eV [Fig. S4(b)]. This qualitatively agrees with the OHC and SHC calculated in the bulk.

![FIG. S3. Current-induced orbital and spin moments from OHE and SHE in a finite system for (a) $E_F = -1.30$ eV and (b) $E_F = 0.00$ eV.](image)

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