Rational design principles for giant spin Hall effect in 5d-transition metal oxides

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Spin Hall effect (SHE), a mechanism by which materials convert a charge current into a spin current, invokes interesting physics and promises to empower transformative, energy-efficient memory technology. However, fundamental questions remain about the essential factors that determine SHE. Here, we solve this open problem, presenting a comprehensive theory of five rational design principles for achieving giant intrinsic SHE in transition metal oxides. Arising from our key insight regarding the inherently geometric nature of SHE, we demonstrate that two of these design principles are weak crystal fields and the presence of structural distortions. Moreover, we discover that antiperovskites are a highly promising class of materials for achieving giant SHE, reaching SHE values an order of magnitude larger than that reported for any oxide. Additionally, we derive three other design principles for enhancing SHE. Our findings bring deeper insight into the physics driving SHE and could help enhance and externally control SHE values.

significance

The conversion of charge current into spin current, via spin Hall effect (SHE), attracts extensive interest due to its rich physics and significant technological impact. As an attractive method to control magnetism using electric currents, SHE could help achieve energy-efficient data storage technologies. Here, we answer the fundamental question, what are the key factors that determine the value of SHE? We present the foundational principles for the design of materials with giant values of SHE and demonstrate the essential physics at play. Additionally, we report the discovery of existing materials that promise giant SHE. These findings are a significant advance in the cutting-edge science and technology of SHE.

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Data deposition: The spin Hall code used to calculate these values has been deposited in GitHub (https://github.com/codellama/Spin-Hall-W90).

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enhance these SHE values. Moreover, the estimated longitudinal conductivities for our best materials are \( \geq 5 \times 10^{2} \) S/cm, making them suitable for applications in emergent memory technology (17). Altogether, our work answers fundamental questions about the physics of SHE, it discovers exciting spin Hall materials, it opens further avenues of materials research for giant SHE, and it leads to possibilities for the development of energy-efficient, next-generation memory devices.

**Results**

**General Origin of the SHE.** We now derive the general factors that control the value of SHE. Specifically, the type of SHE that arises from the intrinsic band structure of a material, called the intrinsic SHE (18, 19). We start with the expression for spin Hall conductivity (SHC) obtained from linear response theory. Let \( \hat{H} \) represent the total Hamiltonian of the system and \( \hat{h}_{0} \) represent the Hamiltonian without spin–orbit coupling (SOC) (19). Let \( \sigma^{n}_{\alpha\beta} \) and \( \Omega^{n}_{\alpha\beta} \) represent the \( \alpha\beta \) component of the band \( n \) resolved SHC and spin Berry curvature, respectively, of this system. The spin momentum is along the direction \( s \), and \( \alpha, \beta, s \) are along the coordinate axes \((x, y, z)\).

\[
\hat{H} = \hat{H}_{0} + \lambda \hat{s} \cdot \hat{s}.
\]

In the above, suppressing the index \( \lambda \):

\[
\sigma^{n}_{\alpha\beta} = -\frac{e}{\hbar} \oint_{\mathcal{BZ}} \frac{d\mathcal{k}}{(2\pi)^{3}} \sum_{n} f_{n}(\mathcal{k}) \Omega^{n}_{\alpha\beta}.
\]

These equations demonstrate that the spin Berry curvature originates from the product of the spin operator \( \hat{s} \) and the anomalous velocity operators \( \hat{v}_{\alpha} \). We assume that the spin-conserving part of the SOC operator \( \lambda \hat{s} \cdot \hat{s} \) makes the dominant contribution to the SHC, as compared with the spin-mixing part. We include our justification for this assumption in SI Appendix and show that this assumption is often, although not always, valid. Substituting 4 into 3, we obtain:

\[
\Omega^{n}_{\alpha\beta} = -\text{Im} \sum_{m\neq n,l} \left[ \langle n|\hat{s}_{l}|m\rangle \langle l|\hat{v}_{m}\rangle + \langle n|\hat{v}_{l}|m\rangle \langle l|\hat{s}_{m}\rangle \right] \left( \epsilon_{m} - \epsilon_{n} \right)^{2}.
\]

The total SHE of the material would involve adding the spin Berry curvature contributions from both the time-reversal partners. Comparing 10 and 11 and using \( \text{Im}(AB^{*}C^{*}) = -\text{Im}(ABC) \), we are left with only two nonzero terms that can contribute to the total SHE \( \sigma^{n}_{\alpha\beta} \). The first term is \( \left( \sigma_{n\alpha\beta}^{n} \right)^{\text{spin}} \equiv \sum_{m\neq n,l} \left[ \langle n|\hat{s}_{l}|m\rangle \langle l|\hat{v}_{m}\rangle + \langle n|\hat{v}_{l}|m\rangle \langle l|\hat{s}_{m}\rangle \right] \left( \epsilon_{m} - \epsilon_{n} \right)^{2} \) and \( \left( \sigma_{n\alpha\beta}^{n} \right)^{\text{spin-mix}} \equiv \left( \sigma_{n\alpha\beta}^{n} \right)^{\text{spin}} \). This term arises from the anomalous velocity originating in the rotation of eigenstates, or in other words, in the mixing of orbitals, caused by the spin-conserving part of SOC. The second term \( \left( \sigma_{n\alpha\beta}^{\text{spin}} \right)^{\text{spin}} \) arises from the anomalous velocity originating in the rotation of eigenstates caused by the spin independent part of the Hamiltonian \( \hat{H}_{0} \). For materials with inversion and time reversal symmetry, which includes all materials in this study, the second term, which is related to the Berry curvature, is zero (20).

In the basis of the \( d \)-orbitals of TMOs, \( \mathcal{L} = \hat{d}_{\alpha} \) either mixes \( \hat{d}_{xz} \) with \( \hat{d}_{y} \) or \( \hat{d}_{xy} \) with \( \hat{d}_{y} \) or \( \hat{d}_{xy} \) or \( \hat{d}_{x} \). Consequently, for \( \hat{v}_{x} = \partial/\partial k_{x} (\lambda \hat{s} \cdot \hat{s}) \), the only nonzero components of \( \hat{v}_{x} \) arise either from the mixing of \( \hat{d}_{x} \) with \( \hat{d}_{y} \) or \( \hat{d}_{xy} \) or \( \hat{d}_{y} \) or \( \hat{d}_{xy} \). From the derivation above, we deduce three general rules for obtaining significant SHE in materials with inversion and time reversal symmetry, as enumerated below.

**R1.** Nonzero SHE requires the bands of character \( d_{i} \) and \( d_{j} \) to be close enough in energy at a given \( \mathbf{k} \), such that the SOC can mix these states, where \( i = xz, j = yz, \) or \( i = xy, j = x^{2} - y^{2} \) for \( d \)-orbitals.

**R2.** For nonzero SHE, the Fermi level should pass through the region where the bands \( d_{i} \) and \( d_{j} \) are crossing or being mixed by SOC. If the Fermi level does not pass through the band mixing region, all interacting states will have the same occupation. As a result, \( f_{n}(\mathcal{k}) \hat{v}_{\alpha\beta}^{\text{spin}} \) will exactly cancel with \( f_{n}(\mathcal{k}) \hat{v}_{\alpha\beta}^{\text{spin}} \hat{v}_{\alpha\beta}^{\text{spin}} \), as \( l_{\alpha\beta} = l_{\alpha\beta}^{\text{spin}} \). Thus, the Fermi-level positioning is critical to the total value of SHE. This rule has been attained previously.
Rational Design Principles for Large SHE in TMOs. In *General Origin of the SHE*, we have derived three basic rules that govern the value of SHE in *d*-orbital materials. From these, we will now deduce the rational design principles for large SHE in TMOs. The band structure of a TMO is generally explained with the help of crystal field theory (CFT) (21, 22). CFT considers a transition metal atom with its detailed electronic structure in an environment of ligands that are considered structureless. The primary effect of the ligands is to alter the electronic structure of the transition metal due to repulsion between the transition metal electrons and the electronegative ligand ions. The repulsive ligand field, and consequently the electronic structure, is determined by the geometry of the ligands surrounding the transition metal. For instance, in most TMOs considered here, the transition metal atom with its detailed electronic structure in an octahedral crystal field (Fig. 1). For an octahedral crystal field, among the six ligands, resulting in what is called an octahedral crystal field (Fig. 1). In contrast, a cubic crystal field splits the otherwise degenerate \(5d\) orbitals into an \(e_g\) (DOF = 2) and a \(t_{2g}\) (DOF = 3) manifold. The splitting of the larger manifold into these smaller spaces reduces the possibilities for rotation of DOFs, thereby reducing the possible spin Berry curvature and consequently reducing SHE. Therefore, we deduce that large values of SHE are encouraged by weak crystal fields. This is Condition 1 (C1). Specifically for TMOs, we have shown above that there are two possible contributions to SHE, one that arises from the mixing of \(d_{xz}\) and \(d_{yz}\) orbitals and another that arises from the mixing of \(d_{xy}\) and \(d_{x^2-y^2}\) orbitals. It follows that SHE is maximized when both of these contributions are nonzero and add constructively to one another. From rule R1, for these two SHE contributions to be nonzero, we would require the \(d_{xz}\) and \(d_{yz}\) bands, as well as the \(d_{xy}\) and \(d_{x^2-y^2}\) bands, to cross one another or be close enough in energy to enable SOC to mix them. The splitting of two \(d\) bands by crystal field into separate manifolds will hamper the mixing of those two bands. As such, the first principle to obtain large values of SHE (C1) is that the crystal field should be weak. For instance, in a weak octahedral or cubic crystal field, the \(e_g\) and \(t_{2g}\) manifolds overlap, allowing band crossings between the \(d_{xy}\) and \(d_{x^2-y^2}\) on \(e_g\) (DOF = 3)

Putting together CFT with the general rules (R) controlling the value of SHE that we had derived before, we will now deduce our five rational design principles to obtain large SHE. Our first principle originates in the inherently geometric nature of the Berry curvature. As a geometric property, Berry curvature results from the rotation of a DOF of the electronic wave function with a variation of some periodic parameter (20). For SHE, which derives from the spin Berry curvature, the relevant DOF is the orbital angular momentum \(\hat{l}\) and the relevant parameter is the wave vector \(\hat{q}\). For \(d\)-orbitals, there are five possible orbital DOFs. However, in TMOs, the crystal field splits this orbital space into smaller subspaces spanned by fewer DOFs. For instance, in octahedral and cubic fields the \(d\)-bands are split into an \(e_g\) (DOF = 2) and a \(t_{2g}\) (DOF = 3) manifold. The splitting of the larger manifold into these smaller spaces reduces the possibilities for rotation of DOFs, thereby reducing the possible spin Berry curvature and consequently reducing SHE. Therefore, we deduce that large values of SHE are encouraged by weak crystal fields. This is Condition 1 (C1). Specifically for TMOs, we have shown above that there are two possible contributions to SHE, one that arises from the mixing of \(d_{xz}\) and \(d_{yz}\) orbitals and another that arises from the mixing of \(d_{xy}\) and \(d_{x^2-y^2}\) orbitals. It follows that SHE is maximized when both of these contributions are nonzero and add constructively to one another. From rule R1, for these two SHE contributions to be nonzero, we would require the \(d_{xz}\) and \(d_{yz}\) bands, as well as the \(d_{xy}\) and \(d_{x^2-y^2}\) bands, to cross one another or be close enough in energy to enable SOC to mix them. The splitting of two \(d\) bands by crystal field into separate manifolds will hamper the mixing of those two bands. As such, the first principle to obtain large values of SHE (C1) is that the crystal field should be weak. For instance, in a weak octahedral or cubic crystal field, the \(e_g\) and \(t_{2g}\) manifolds overlap, allowing band crossings between the \(d_{xy}\) and \(d_{x^2-y^2}\) on \(e_g\) (DOF = 3). 

In materials with strong crystal fields, mixing between otherwise separate manifolds can be enhanced if structural distortions are present. Structural distortions of octahedral crystal fields, such as octahedral rotation and tilting, have been shown to break up the \(e_g\) and \(t_{2g}\) manifolds into further subbands for SrIrO$_3$ (23). This splitting up of a manifold increases the total bandwidth of that manifold, which encourages overlaps or transitions between separate manifolds. In other words, structural distortions increase the space of rotations of the orbital DOFs. We would thus expect the presence of structural distortions to mitigate the dampening effect of a strong crystal field on SHE and generate large SHE values. We call this condition of structural distortions Condition 2 (C2). The enhancement of SHE via distortions has been experimentally reported for SrIrO$_3$ (24). Although we have derived Conditions 1 and 2 (C1 and C2) specifically for octahedral crystal fields, they are general conditions extending beyond octahedral or even cubic crystal fields. This is because, in general, the weaker the crystal field, and the greater the structural distortions, the greater the possibility of mixing between various \(d\)-orbitals. Condition 3 (C3) is analogous as the general rule 2 (R2), i.e., the optimal positioning of the Fermi level. As mentioned before, this rule has been attained previously (e.g., ref. 20). Using an alternative approach, we have reobtained this rule and include it in our design principles for completeness.

To deduce Condition 4 (C4), we consider materials with octahedral or cubic crystal fields and with symmetry preserved between two cubic axes (say, \(X\) and \(Y\)). In other words, any structural distortions that may be present in the material do not break the symmetry between \(X\) and \(Y\) axes. We focus only on the \(t_{2g}\) manifold of the octahedral and cubic crystal fields and the transitions therein between the \(d_{xz}\) and \(d_{yz}\) orbitals. The impact of SOC is known to couple the spin and orbital states and to split

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**Fig. 1.** Illustration of crystal field splitting in TMOs. A demonstrates structural diagrams of octahedral, cubic, and linear crystal fields (CFs), with the transition metal atom marked in red and the ligand atom marked in cyan. B plots the energy splittings of \(d\)-orbitals generated under these crystal fields.
the $t_{2g}$ manifold into a lower-energy $J_{eff} = 3/2$ and a higher-energy $J_{eff} = 1/2$ submanifold (25). The electron wave functions for the states in these $J_{eff} = 3/2$ and $1/2$ submanifolds can be written as:

$$|j_{eff} = 3/2, m_j = 3/2\rangle = -\frac{1}{\sqrt{2}} d_{yz} \otimes |\uparrow\rangle - \frac{i}{\sqrt{2}} d_{zx} \otimes |\uparrow\rangle$$
$$|j_{eff} = 3/2, m_j = 1/2\rangle = -\frac{1}{\sqrt{6}} d_{yz} \otimes |\downarrow\rangle - \frac{i}{\sqrt{6}} d_{zx} \otimes |\downarrow\rangle + \sqrt{\frac{2}{3}} d_{xy} \otimes |\uparrow\rangle$$

$$|j_{eff} = 3/2, m_j = -1/2\rangle = \frac{1}{\sqrt{6}} d_{yz} \otimes |\uparrow\rangle - \frac{i}{\sqrt{6}} d_{zx} \otimes |\uparrow\rangle$$
$$|j_{eff} = 3/2, m_j = -3/2\rangle = -\frac{1}{\sqrt{2}} d_{yz} \otimes |\downarrow\rangle - \frac{i}{\sqrt{2}} d_{zx} \otimes |\downarrow\rangle$$

$$|j_{eff} = 1/2, m_j = 1/2\rangle = \frac{1}{\sqrt{3}} d_{yz} \otimes |\downarrow\rangle + \frac{i}{\sqrt{3}} d_{zx} \otimes |\downarrow\rangle$$
$$|j_{eff} = 1/2, m_j = -1/2\rangle = \frac{1}{\sqrt{3}} d_{yz} \otimes |\uparrow\rangle - \frac{i}{\sqrt{3}} d_{zx} \otimes |\uparrow\rangle$$
$$-\sqrt{\frac{3}{2}} d_{xy} \otimes |\downarrow\rangle,$$

where $|\uparrow\rangle$ and $|\downarrow\rangle$ are spin-up and spin-down states, respectively. In materials with octahedral or cubic crystal fields and $X-Y$ symmetry, absent SOC, the electron wave functions will have equal contributions from the $d_{xz}$ and $d_{yz}$ orbitals. Once SOC is turned on, if the $t_{2g}$ manifold is split into well-separated $J_{eff} = 3/2$ and $1/2$ submanifolds (25), these coupled $J_{eff} = 3/2$ and $1/2$ states will be given by Eqs. 12 and 13. However, these coupled states also will have equal contributions from the $d_{xz}$ and $d_{yz}$ orbitals. Since the electron states before and after turning on SOC would have equal contributions from the $d_{xz}$ and $d_{yz}$ orbitals, we can deduce that turning on SOC would not result in any net $d_{xz} - d_{yz}$ transitions and would not contribute to SHE (refer to rule R3). In contrast, the same structural conditions, octahedral or cubic crystal fields and $X-Y$ symmetry but with mixing or overlap between $J_{eff} = 3/2$ and $1/2$ submanifolds, could result in enhanced SHE values due to net transitions between the $d_{xz}$ and $d_{yz}$ orbitals. We call this Condition 4 (C4). As we will see later, this condition can be met by 1) a moderate SOC regime where the $J_{eff} = 3/2$ and $1/2$ submanifolds are not well separated or 2) orthorhombic structural distortions. There is some overlap between conditions C2 and C4 since structural distortions could also enhance the mixing of $J_{eff} = 3/2$ and $1/2$ states. However, since distortions are not the only method to result in this mixing of $J_{eff} = 3/2$ and $1/2$ states, we count this mixing as a separate condition. Finally, Condition 5 (C5) is that the presence of moderate electron correlation may enhance SHE by localizing electrons, as has been proposed in literature (26). However, later we show that this effect is nuanced and depends on band structure details.

To summarize, the five general conditions for achieving for giant SHE are weak crystal fields (C1), structural distortions (C2), optimal positioning of the Fermi level (C3), mixing of $J_{eff} = 1/2$ and $3/2$ bands for materials under octahedral or cubic crystal fields with $X-Y$ symmetry (C4), and, in some cases, moderate electron correlations (C5). These conditions also serve as rational design principles. In the remaining sections, we report our findings on SHE in various $5d$-TMOS and explain them on the basis of these five rational design principles.

The Effect of Weak Crystal Field (C1). We first report the finding of giant SHE in BCC-Pt$_3$O$_5$. Pt$_3$O$_5$ has two possible proposed crystal structures (27), body centered cubic (BCC) (28) and simple cubic (SC) (29). BCC-Pt$_3$O$_5$ consists of Pt atoms surrounded by a cubic oxygen crystal field, while SC-Pt$_3$O$_5$ has Pt atoms inside a square planar crystal field. As we show below, the crystal field in BCC-Pt$_3$O$_5$ is weak which satisfies C1, leading to a giant SHE. The calculated SHE for BCC-Pt$_3$O$_5$ is $3.7 \times 10^5 \ h/2e^2 s/cm$ with an estimated giant spin Hall angle $\Theta_{SH}$ of $\sim 7.4$. In sharp contrast to BCC-Pt$_3$O$_5$, SC-Pt$_3$O$_5$ displays a small SHE as a result of its strong crystal field which violates C1. Another exciting result is the finding of a hypothetically giant SHE in the rare earth antiperovskite Yb$_2$PbO. Yb$_2$PbO has Yb in a weak crystal field, satisfying C1. However, unfortunately, the nominal Fermi level in Yb$_2$PbO is poorly placed which violates C3. Engineering the Fermi level to place it at 0.6 eV above the nominal Fermi level of Yb$_2$PbO satisfies C3, leading to an SHE of $-2.4 \times 10^3 \ h/2e$ S/cm with an estimated giant $\Theta_{SH}$ of $-2.5$ to $-4.9$. Although hypothetical, this latter result, alerts us to the possibility of finding giant SHE in 5d-transition metal antiperovskites via careful engineering of the Fermi level. Our preliminary results for SHE in TM$_2$PbO, where Yb atoms have been substituted by 5d-transition-metal atoms Hf and Ta, show promise toward achieving giant SHE in antiperovskites. We will report these results in detail in future work. The giant values of $\Theta_{SH}$ described in this section and in Table 1 are at least an order of magnitude larger than those reported for any TMOs so far. Additionally, these values are greater than those observed for all other measured materials except for Bi$_2$Se$_3$ and Bi$_2$Te$_3$. Our spin Hall results give further support to the rational design principles deduced above.

BCC-Pt$_3$O$_5$ has a cubic crystal field that splits the $5d$-bands into a lower-energy $e_g$ and a higher-energy $t_{2g}$ manifold. A large Pt-O bond length of 2.7 Å leads to a weak crystal field (C1), which is evident from a significant overlap between the $d_{xy}$ and $d_{yz}$ orbitals. We call this Condition 5 (C5). As we will see later, this condition can be met by 1) a moderate SOC regime where the $J_{eff} = 3/2$ and $1/2$ submanifolds are not well separated or 2) orthorhombic structural distortions. There is some overlap between conditions C2 and C4 since structural distortions could also enhance the mixing of $J_{eff} = 3/2$ and $1/2$ states. However, since distortions are not the only method to result in this mixing of $J_{eff} = 3/2$ and $1/2$ states, we count this mixing as a separate condition. Finally, Condition 5 (C5) is that the presence of moderate electron correlation may enhance SHE by localizing electrons, as has been proposed in literature (26). However, later we show that this effect is nuanced and depends on band structure details.

To summarize, the five general conditions for achieving for giant SHE are weak crystal fields (C1), structural distortions (C2), optimal positioning of the Fermi level (C3), mixing of $J_{eff} = 1/2$ and $3/2$ bands for materials under octahedral or cubic crystal fields with $X-Y$ symmetry (C4), and, in some cases, moderate electron correlations (C5). These conditions also serve as rational design principles. In the remaining sections, we report our findings on SHE in various $5d$-TMOS and explain them on the basis of these five rational design principles.
Comprising only two ligand atoms makes the linear crystal field weak, satisfying C1 and allowing for a large SHE. This crystal field splits the 5d-bands into smaller manifolds, comprising degenerate $d_{xy}$ and $d_{yz}$ bands and separately degenerate $d_{xz}$ and $d_{2z^2-r^2}$ bands. The weakness of this linear crystal field is evident from the overlap between these manifolds as shown in Fig. 2 C, b and e. The degeneracy between $d_{xy}$ and $d_{2z^2-r^2}$ orbitals is further broken by a weak repulsion of the Yb-$d_{xy}$ orbitals from the B site Pb ions (Fig. 2 C, a) that pushes the $d_{xy}$ bands higher in energy (see Fig. 2 C, b). The concerted effort of the weak ligand crystal field (C1), and a weak repulsive field from the cation Pb (modified C1), along with a hypothetical increase in the Fermi level by 0.6 eV (C3), leads to a giant SHE. At 0.6 eV above $E_F$, close to the $M$ point, there is a band crossing between the lower Yb-$d_{2z^2-r^2}$ bands and the upper Yb-$d_{xy}$ bands, along with the presence of $d_{xz}$ and $d_{yz}$ orbitals (Fig. 2 C, b and e). SOC is expected to mix these bands and create a large hot spot for spin Berry curvature around $M$, which is shown in Fig. 2 C, d. As a result, we see a giant estimated SH angle in Yb$_3$PbO for a Fermi-level positioning of 0.6 eV above the nominal value. Yb$_3$PbO is identified as a possible topological crystalline insulator (30) with a Dirac node at the $E_F$ (31); however, we do not see any significant SHE contribution from this node.

Here, we would like to note that while technically Yb$_3$PbO is a rare-earth (RE) oxide and not a TMO, a Fermi level that is 0.6 eV above the nominal Fermi level of Yb$_3$PbO passes through the empty 5d-bands of Yb, which brings our design principles into effect. As mentioned before, the optimal positioning of the Fermi level in antiperovskites for giant SHE can be achieved by substituting Yb atoms with the 5d-transition metal atoms Hf and Ta, such that the Fermi level passes through the 5d-bands. We also note that there exist many known 5d-transition metal antiperovskite compounds (beyond oxides) such as Pt$_2$REB (RE: rare earth) (32) and Pt$_2$AP (A = Sr, Ca, La) (33), while many more are theoretically predicted to be stable (34) and remain to be synthesized. Our preliminary calculations on these transition metal antiperovskites show that giant spin Hall values can be obtained in antiperovskites by careful engineering of the materials’ chemical composition. We will report these results in detail in future work.

**The Role of Structural Distortions (C2).** We now examine cubic perovskites where the transition metal atom is under a typically strong, octahedral crystal field. BaOsO$_3$ (35, 36) and SrOsO$_3$ (36) are both perovskite oxides with different sizes of the A site cation. As a result, BaOsO$_3$ adopts a perfect perovskite structure free from distortions and demonstrates a low SHC, on account of its strong crystal field violating C1. In contrast, SrOsO$_3$ demonstrates large octahedral distortions, satisfying C2, which leads to a larger SHE for SrOsO$_3$. To account for correlation effects in BaOsO$_3$ and SrOsO$_3$, we use an local-density approximation (LDA)+U scheme with U = 2 eV, which is taken from ref. 36. Like SrOsO$_3$, SrIrO$_3$ (23) also demonstrates significant distortions of the O6 octahedra, which satisfies C2, imparting a larger SHC to SrIrO$_3$ than BaOsO$_3$. The enhancement of SHC by structural distortions has been experimentally shown for SrIrO$_3$ (24). The values of SHC and $\Theta_{SH}$ for these materials are calculated along the pseudo-cubic axis and are enumerated in Table 2. An analysis of SHE in the rutile binary oxides OsO$_2$ and IrO$_2$ is included in SI Appendix.

BaOsO$_3$ is a distortion-free cubic perovskite, with a metallic conductivity (35, 36), and an octahedral crystal field that splits the 5d-bands into a lower $t_{2g}$ and an upper $e_g$ manifold, with the Fermi level lying inside the former. The strong crystal field precludes any $e_g - t_{2g}$ overlap, violating C1, as is evident from Fig. 3 A, b, and resulting in a small SHC (Table 2). As BaOsO$_3$ shows an octahedral crystal field, $X$–$Y$ symmetry, and no clear

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**Fig. 2.** SHE in BCC-Pt$_3$O$_4$ (A), SC-Pt$_3$O$_4$ (B), and Yb$_3$PbO (C). A–C, a display the structure of these oxides with Pt in silver, Yb in cyan, Pb in dark gray, and O in red. A–C, b and c show the band structure projected onto $d_{xy}$ orbitals (green) and $d_{2z^2-r^2}$ orbitals (yellow), without and with SOC, respectively. A–C, e and f plot the band structure projected onto $d_{xy}$ orbitals (green) and $d_{yz}$ orbitals (yellow), without and with SOC, respectively. A–C, d portray the spin Berry curvature.

$\Theta_{SH}$ of 0.5. A square planar crystal field splits the 5d-bands into multiple manifolds, starting from a higher-energy $d_{2z^2-r^2}$ level, followed by $d_{xy}$ and even lower $d_{2z}$ levels, and ending with the lowest energy level comprised of degenerate $d_{xz}$ and $d_{yz}$ bands. The strength of the square planar crystal field in SC-Pt$_3$O$_4$ is evident from the insignificant overlap between the $d_{xy}$ and $d_{2z^2-r^2}$ orbitals, as shown in Fig. 2 B, b. Additionally, in the absence of SOC, we observe a minimal presence of $d_{xy}$ and $d_{yz}$ bands at the Fermi level, all of which leads to a much smaller SHC than that seen for BCC-Pt$_3$O$_4$.

Yb$_3$PbO is an antiperovskite, where the A and B sites of a regular perovskite have been swapped such that the central atom (Yb) is on the A site and Pb is on the B site. Every Yb atom is bonded to only two oxygen atoms with an O-Yb-O bond angle of 180°, constituting a linear ligand crystal field.
splitting between the $J_{g}$ = 3/2 and 1/2 submanifolds (Fig. 3 A, e and f), we expect C4, i.e., mixing of the $J_{\text{eff}}$ = 3/2 and 1/2 states, to contribute to the SHE in this case.

SrOsO$_3$ (36) and SrIrO$_3$ (23) are orthorhombic perovskites with a strong octahedral crystal field (C1), as well as the presence of structural distortions (C2). If acting alone, a strong crystal field violates C1, leading to a low SHE, as seen in BaOsO$_3$. However, structural distortions satisfy C2 and mitigate the effect of crystal field, resulting in a larger SHE for SrOsO$_3$ and SrIrO$_3$ (Table 2). The mechanism by which structural distortions enhance SHE is evident from a comparison of the projected band structures for BaOsO$_3$ (Fig. 3 A, b and c), with that for SrOsO$_3$ (Fig. 3 B, b and c) or SrIrO$_3$ (Fig. 3 C, b and c). Distortions split the $e_g$ and $t_{2g}$ manifolds into multiple subbands, increasing the energy span of both manifolds. This splitting, in turn, increases the likelihood of crossings between different subbands within a manifold, as well as crossings between different manifolds. This mechanism underpins C2.

The busy plots of band structure and spin Berry curvature for SrOsO$_3$ (Fig. 3 B, b-f) and SrIrO$_3$ (Fig. 3 C, b-f) make it difficult to isolate the exact regions of spin Hall generation. However, a broad inspection reveals multiple degeneracy splitting by the action of SOC. Inspection of projected band structures without SOC at the Fermi level reveals the presence of $e_g$–$t_{2g}$ overlap that is greater than that seen in BaOsO$_3$. The $e_g$–$t_{2g}$ manifold overlap is greater for SrIrO$_3$ than SrOsO$_3$, owing to the increased band filling of the former. This overlap likely explains the order of SHC values seen that are largest for SrIrO$_3$, followed by SrOsO$_3$, and smallest for BaOsO$_3$. Our estimated values of $\Theta_{SH}$ for SrIrO$_3$ are $\Theta_{SH}^{S_{\text{y}}}$ ≈ 0.2 and $\Theta_{SH}^{S_{\text{z}}}$ ≈ 0.34 (Table 2), which are comparable to the experimentally observed values of $\Theta_{SH}^{S_{\text{y}}}$ ≈ 0.5 and $\Theta_{SH}^{S_{\text{z}}}$ ≈ 0.3 (24).

The Effect of Electron Correlations (C5). In this section, we focus on the relationship between electron correlations and SHE captured in C5. It has been proposed that moderate electron correlations enhance SOC by localizing electrons (26) and therefore should enhance SHE as well. However, here, we find that while moderate correlations may impact SHE, this effect of correlations is more nuanced and depends on band structure details (C5). This nuanced effect is due to the presence of competing spin Berry curvature hot spots, as well as local changes in band occupation caused by electron correlations.

Pyrochlore iridates, in particular RE pyrochlore iridates, have generated keen interest for their interesting properties resulting from the interaction of moderate correlation and SOC (37). These properties include the anomalous Hall effect (38), spin-liquid state (39), Weyl semimetal (40, 41), etc. Here, we study the SHE in a RE pyrochlore iridate, Tb$_2$Ir$_2$O$_7$ (42, 43). In contrast, the pyrochlore iridate Bi$_2$Ir$_2$O$_7$ (44–46) has low electron correlations due to strong hybridization between Ir 5d and Bi 6p electrons (44). We include electron correlations for Tb$_2$Ir$_2$O$_7$ under an LDA+U scheme with U ranging from 0 to 2.5 eV. Our results for SHE in Tb$_2$Ir$_2$O$_7$ and Bi$_2$Ir$_2$O$_7$ are listed in Table 2, with the pyrochlore lattice vectors $\vec{a}$, $\vec{b}$, and $\vec{c}$ defined along $\hat{x} + \hat{y}$, $\hat{x} - \hat{y}$, and $\hat{z}$ directions, respectively. With increasing correlation (U), the SHE for Tb$_2$Ir$_2$O$_7$ shows nonlinear, complex behavior. For Bi$_2$Ir$_2$O$_7$, we predict a SHC of −122 to −512 $\hbar$/2e S/cm. The complicated behavior of SHC with correlations (U) observed in Tb$_2$Ir$_2$O$_7$ arises due to competing spin Berry correlations.

Table 2. List of SHC $\sigma_{\text{SH}}^{S_{\alpha}}$ ($\hbar$/2e S/cm), longitudinal conductivity $\sigma$, and spin Hall angle $\Theta_{SH}^{S_{\alpha}}$ for cubic perovskites

| Structure | $\sigma_{xy}$ | $\sigma_{yz}$ | $\sigma_{zx}$ | $\sigma$ (S/cm) | $\Theta_{SH}^{S_{x}}$ | $\Theta_{SH}^{S_{y}}$ | $\Theta_{SH}^{S_{z}}$ |
|-----------|--------------|--------------|--------------|-----------------|---------------------|---------------------|---------------------|
| BaOsO$_3$ | −150         | −134         | −150         | 55 (51)         | −2.7                | −2.4                | −2.7                |
| SrOsO$_3$ | −294         | 30           | 276          | 128 (51)        | −2.3                | 0.23                | 2.15                |
| SrIrO$_3$ | 382          | 680          | 84           | $2 \times 10^3$ (23) | 0.2                  | 0.34                | 0.05                |

Spin Hall prediction for cubic perovskites

| Correlation | $U = 0.0$ eV | $U = 0.5$ eV | $U = 1.0$ eV | $U = 1.5$ eV | $U = 2.0$ eV |
|-------------|-------------|-------------|-------------|-------------|-------------|
| $\Theta_{SH}^{S_{x}}$ | −111        | 186         | 16          | 284         | −374        |
| $\Theta_{SH}^{S_{y}}$ | 115         | 160         | 15          | 134         | 528         |
| $\Theta_{SH}^{S_{z}}$ | −10         | −136        | −56         | −56         | −56         |

Spin Hall prediction for cubic perovskites

| Correlation | $U = 0.0$ eV | $U = 0.5$ eV | $U = 1.0$ eV | $U = 1.5$ eV | $U = 2.0$ eV |
|-------------|-------------|-------------|-------------|-------------|-------------|
| $\Theta_{SH}^{S_{x}}$ | −122        | −512        | −158        | −714 (45)   | −0.2        |
| $\Theta_{SH}^{S_{y}}$ | −0.7        | −0.2        | −0.2        | −0.2        | −0.2        |
Fig. 4. SHE in pyrochlore oxides \(\text{A}_{2}\text{Ir}_{2}\text{O}_{7}\), A = Tb, or Bi. A displays the structure with Tb/Bi in cyan, Ir in purple, and O in red. For B, a–e display the spin Berry curvature for \(\text{Tb}_{2}\text{Ir}_{2}\text{O}_{7}\) with correlation (U) values between 0 and 1.5 eV, and f portrays the corresponding band structure. For C, a shows the band structure of \(\text{Bi}_{2}\text{Ir}_{2}\text{O}_{7}\) without (blue) and with (orange) SOC, b displays the band structure projected onto \(d_{xy}\) orbitals (green) and \(d_{z^2−r^2}\) orbitals (yellow) without SOC, c plots the spin Berry curvature, and d shows the band structure projected onto \(d_{xy}\) orbitals (green) and \(d_{yz}\) orbitals (yellow) without SOC.

The behavior of the hot spot contributions with U is a result of local band fillings. For the first hot spot (\(L−\Gamma\)), the band fillings remain essentially unchanged as U increases (Fig. 4 B, f). Therefore, overall, increasing U increases the SHC contribution from this hot spot. However, for the second hot spot (\(\Gamma−X\)) the band filling changes substantially for one of the bands as U increases. When U is small, the Fermi level goes cleanly through the gap created by SOC, which should maximize the total Berry curvature contribution from this hot spot (Fig. 4 B, f). When U is increased, the lower band is pushed just above the Fermi level, thereby decreasing the SHC contribution. Therefore, the second hot spot contribution initially increases and then decreases with increasing U. Overall, we find that moderate electron correlations may impact SHE. However, the exact effect of correlations on SHE depends band-structure details, such as competing spin Berry curvature hot spots and changes in local band occupation. These inferences are encapsulated in C3 of our rational design principles.

Discussion

In this paper, we present a comprehensive theory of the foundational factors that determine the value of intrinsic SHE in TMOs. SHE is of fundamental interest owing to its rich physics. It is also highly attractive for enabling energy-efficient, next-generation memory technology. Therefore, our results not only bring important insight into interesting, fundamental physics, they also carry significant promise for helping emerging memory technology. Our findings partly originate from the key realization that intrinsic SHE is inherently a geometric property. Such geometric properties are maximized when the rotation of the DOFs of a wave function is maximized. This condition is fulfilled under weak crystal fields and structural distortions. Therefore, we report that weak crystal fields and structural distortions are factors that enhance the value of SHE. Moreover, we derive three other factors that control SHE values, optimal positioning of the Fermi level, mixing of \(d_{z^2}\) = 1/2 and 3/2 bands for materials under octahedral or cubic crystal fields with \(X−Y\) symmetry, and, in some cases, electron correlations. Our design principles are general enough to apply to transition metal compounds, as well as to other geometric properties like the anomalous Hall effect, orbital Hall effect, etc. Our design principles are also impactful because of the inherent tunability of transition metal compounds. In the future, our design principles could be used to develop the highly attractive, in situ, external field control of SHE.

We also report that previously unexplored materials (BCC-\(\text{Pt}_3\text{O}_4\) and \(\text{Sd}\)-antiperovskites) promise to demonstrate giant efficiencies of conversion of charge current to spin current that are an order of magnitude larger than that reported for any oxide so far. Additionally, we enumerate large efficiency values found in a significant number of other oxides. Our discoveries of exciting spin Hall materials along with the implementation of our rational design principles could significantly advance the spin Hall values observed to date. Put together, our work answers fundamental questions about the physics of SHE, it discovers attractive spin Hall materials, it opens further avenues of materials research for giant SHE, and leads to possibilities for the development of transformative memory devices with external field control of SHE.

Materials and Methods

We performed calculations using QUANTUM ESPRESSO (52), WANNIER90 (53), and our in-house code, which calculates SHC from the output of the
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