Intercorrelated anomalous Hall and spin Hall effect in kagome-lattice Co$_3$Sn$_2$S$_2$-based shandite films

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Magnetic Weyl semimetals (mWSMs) is characterized by linearly dispersive bands with chiral Weyl node pairs associated with broken time reversal symmetry. One of the hallmarks of mWSMs is the emergence of large intrinsic anomalous Hall effect. On heating the mWSM above its Curie temperature, the magnetism vanishes while exchange-split Weyl point pairs collapse into doubly-degenerated gapped Dirac states. Here, we reveal the attractive potential of these Dirac nodes in paramagnetic state for efficient spin current generation at room temperature via the spin Hall effect. Ni and In are introduced to separately substitute Co and Sn in a prototypical mWSM Co$_3$Sn$_2$S$_2$ shandite film and tune the Fermi level. Composition dependence of spin Hall conductivity for paramagnetic shandite at room temperature resembles that of anomalous Hall conductivity for ferromagnetic shandite at low temperature: exhibiting peak-like dependence centering around the Ni-substituted Co$_2$Ni$_2$Sn$_3$S$_2$ and undoped Co$_3$Sn$_2$S$_2$ composition, respectively. The peak shift is consistent with the redistribution of electrons’ filling upon crossing the ferromagnetic-paramagnetic transition, suggesting intercorrelation between the two Hall effects. Our findings highlight a novel strategy for the quest of spin Hall materials, guided by the abundant experimental anomalous Hall effect data of ferromagnets in the literature.

Non-trivial topology in the band structure of a solid can give rise to large Berry curvature $^{1,2}$ acting as an effective magnetic field in real space. This field can deflect the electrons in motion, leading to an intrinsic off-diagonal transport contribution that does not depend on the extrinsic electrons’ scattering rate. Typical examples are the anomalous Hall effect (AHE) $^3$ in ferromagnets and its spin counterpart the spin Hall effect (SHE) $^4$. The latter often involves non-magnetic materials with strong spin-orbit coupling and allows generation of a transverse spin current capable of manipulating the magnetization of an adjacent nanomagnet. The resulting spin-orbit torques (SOT) $^5$ is promising for applications including non-volatile memory, magnetic logic, field sensing and neuromorphic computing. Finding material systems that exhibit high charge-to-spin conversion efficiency is a key to realize competitive spin-orbitronic devices with low power consumption.

SHE has thus far only been investigated for a small subset of all the known materials. One primary challenge for probing the SHE is the non-conservative nature of the spin current, thus necessitates material integration into devices of comparable length scale (e.g., the thickness) with the commonly nanometric spin diffusion length $\lambda$ $^6$. In contrast, owing to its ease of evaluation and prolonged history, the AHE for many conducting magnetic materials are known and available in the literature. In view of the very similar origin and scaling relationship for AHE $^7$ and SHE $^8$, it is tempting to study the intercorrelation between them. Using the abundant AHE data as a facile indicator, one may unveil new design principles for materials with large SHE. Different from the trivial electronic bands in conventional 3d ferromagnetic metals $^9$, we focus on one of the topological bands in a magnetic Weyl semimetal (mWSM) $^{10,11}$ prototype cobalt shandite Co$_3$Sn$_2$S$_2$ (CSS) to reveal the intriguing correlation between the AHE and SHE.

Figure 1(a) illustrates a rhombohedral structure of CSS (Space group No. 166: R3m) which consists of alternate Co$_3$Sn/ SnS$_2$ planes, stacking along the c-axis in the hexagonal representation. Co atoms form a kagome lattice within the ab plane and exhibit strong perpendicular magnetic anisotropy with Curie temperature ($T_C$) of $\sim 177$ K. Recent spectroscopic studies $^{14,15}$ have established ferromagnetic CSS (FM-CSS) as an exotic mWSM with pairs of Weyl points (WPs) near the Fermi level ($E_F$) connected by the chiral surface Fermi arcs. This leads to large summation of the Berry curvature in FM-CSS, exhibiting a record-high anomalous Hall angle exceeding 0.2 $^{16,17}$. The relatively low $T_C$ of CSS, however, hinders the prospect of exploiting its topological properties for many practical applications. So far, very limited studies $^{18}$ were devoted to explore the usefulness of paramagnetic CSS (PM-CSS) at room temperature. Here, we highlight the intercorrelation between the intrinsic AHE and SHE for CSS-based compounds, across the ferromagnetic-paramagnetic transition. We
represent the position of the Weyl/Dirac points. The arrows in (σ) the Fermi level points in DOS and Hall conductivities. d of states (DOS) (σ), that flows along xy with polarization along z(y). (c-e) Calculated electronic band structure (c), spin-resolved density of states (DOS) (d), and anomalous Hall conductivity σxy (e) as a function of Fermi energy for ferromagnetic (FM) Co3Sn2S2 in the magnetic Weyl semimetal state. (f-h) Calculated electronic band structure (f), spin-resolved DOS (g), and two selected components σxy and σxz of the spin Hall conductivity tensor (h) for paramagnetic (PM) Co3Sn2S2. E/kT = 0 represents the Fermi level E/F for undoped Co3Sn2S2 in the FM and PM states, respectively. Red and blue dashed boxes in (c) and (f) represent the position of the Weyl/Dirac points. The arrows in (d), (e), (g), and (h) indicate contributions of these Weyl/Dirac points in DOS and Hall conductivities.

further demonstrate that Fermi-level tuning via isostructural substitutional alloying is an effective strategy allowing full exploitation of the topological features in the band structure of PM-CSS, for efficient spin current generation via SHE at room temperature.

To elucidate our concept, we consider a two-orbital effective tight-binding model of CSS [19, 20]. A longitudinal charge current jx is applied along x in the ab kagome plane, as illustrated in Fig. (a). The magnetization is along z coinciding with the c-axis. We first focus on FM-CSS in the mWSM state, corresponding to Figs. (b), (c), and (e). Highlighted by the dashed red box in the simplified electronic band structure of half-metallic FM-CSS in Fig. (c), the two WPs in the vicinity of EF (represented by E/t1 = 0), manifest themselves in the spin-resolved density of states (DOS) [Fig. (d)] as a minima of the majority spin-up DOS. The anomalous Hall conductivity σxy (AHC) [Fig. (e)] exhibits a peak centering around the WPs at EF. We highlight that at higher energy, a similar σxy peak of opposite sign appears (blue arrow), which is attributed to the two conjugated WPs from the spin-down bands [Fig. (c); dashed blue box]. Using the lattice parameter of CSS a = 0.538 nm and recognizing e2/h is half of the conductance quantum, at EF, we obtain σxy ≈ 1.0 × 103 Ω−1 cm−1, which agrees with the experiment and first-principles calculations [14].

The calculated properties for PM-CSS where the exchange splitting is absent are shown in Figs. (f), (g), and (h). The conjugated WPs annihilate into doubly-degenerated Dirac points followed by gap opening due to spin-orbit coupling. [Fig. (f); dashed boxes] 21, 22 The spin-resolved DOS of the two spins channels are equal, as shown in Fig. (g). σxy vanishes because contributions from the two spins exactly cancel each other, as expected for a paramagnet. In contrast, the spin current is time reversal invariant, e.g., a spin-up spin current flowing along y is equivalent to a spin-down spin current flowing along −y. The spin Berry curvature contributions from the two spin channels are additive and linked to the position of the gapped Dirac point. As depicted in Fig. (b), for jx along x, we define the spin Hall conductivity (SHC) σxy(i, j = x, y, z) where the spin current is flowing along i with polarization along j. Lateral SHC σxy [Fig. (h); skyblue] of approximately twice-as-large to AHC is expected near the gapped Dirac point. Using the same σxy and σxy ≈ 1.9 × 1011 (h/2e)Ω−1 cm−1, which approaches that of a typical spin Hall metal Pt 23, 24. In practice, for (0001)-textured CSS film in this work, it is more convenient to detect the spin current flowing along z with polarization along y, i.e., σxz. Calculations suggest a peak near the gapped Dirac point for σxz [Fig. (h); pink]. This maximum is however smaller than that of σxy where charge and spin current are both flowing in the kagome plane, a feature resembles another kagome semimetal Fe3Sn2 with highly anisotropic AHC tensor 25. The strong anisotropy reflects that the interplay between the conduction electron, kagome lattice, and spin-orbit coupling determines the Hall effects. We emphasize that SHC of undoped PM-CSS is unoptimized.
as $E_F$ is well below the gapped Dirac point. Within rigid band approximations, an electron doping of $\sim 1$ electron per formula unit is required to reposition $E_F$ close to the gapped Dirac point, for which a dramatic enhancement of SHE is expected.

We have grown textured undoped CSS, Ni-substituted Co$_{y}$−Ni$_x$Sn$_2$S$_2$ (CNSS), and In-substituted Co$_3$Sn$_2$−yIn$_y$S$_2$ (CSIS) films with thicknesses ranging from 11 to 40 nm on Al$_2$O$_3$ (0001) substrates by co-sputtering as described previously and in Section S1 of the Supplemental Material. $x$ and $y$ denote the composition of Ni and In, respectively. Upon replacing Co with Ni (Sn with In), the electron (hole) doping is expected to shift $E_F$ to higher (lower) energies. The crystal structure of CSS is maintained throughout because CSS, Ni$_3$Sn$_2$S$_2$ and Co$_3$In$_2$S$_2$ are isostructural compounds. Figure 2(a) shows typical x-ray diffraction (XRD) spectrum for an undoped CSS film exhibiting clear Laue fringes around the CSS (0006) reflection, indicative of strong (0001) texture with sharp interfaces. In-plane XRD $\Phi$ scan of the CSS (1120) reflection, however, shows peaks that are $30^\circ$ apart, reflecting the presence of in-plane twinned domains. The out-of-plane lattice parameter $c$ in Fig. 2(b) was extracted from the (0006) peak position and suggests systematic tuning of $x$ and $y$.

The magneto-transport as a function of temperature $T$ in Figs. 2(c) and 2(d) provides another evidence of systematic tuning of film properties by alloying. Solid lines denote data for undoped CSS (black), CNSS (red) and CSIS (blue) films covered by a thick $\sim 75$ nm SiO$_x$ protective layer, whereas dashed lines represent the data for samples after removing the SiO$_x$ layer by Ar ion milling followed by the deposition of 3 nm AlO$_x$ capping. The high quality of our undoped CSS film gives rise to a residual resistivity ratio, $\rho_{xx}/\rho_{xx}(T = 300\text{ K})/\rho_{xx}(T = 10\text{ K}) \sim 3$. For doped films, the kink in $\rho_{xx}$ reflecting $T_C$ and the average resistance are systematically controlled by In and Ni contents. Meanwhile, $\sigma_{xy}$ [Fig. 2(d)] is nearly a plateau for $T$ sufficiently far below $T_C$, suggesting the intrinsic nature of the AHE. $T_C$, $\rho_{xx}$ at $T = 300\text{ K}$, the maximum anomalous Hall angle $\theta_{\text{AHE, max}} \equiv \sigma_{xy}/\rho_{xx}$ extracted at $T = T_{\text{AHE, max}}$ (g), and $\sigma_{xy}$ at 10 K are summarized in Figs. 2(e)–2(h). Referring to Stoner criterion, both electron and hole doping reduce $T_C$ because $E_F$ of undoped PM-CSS falls on a local maxima of the DOS [Fig. 2(g)]. Similarly, $\sigma_{xy}$ at 10 K and $\theta_{\text{AHE, max}}$ are maximized for undoped FM-CSS and fall rapidly with increasing $x$ and $y$. These observations confirm the correlation between the large intrinsic $\sigma_{xy}$ and the $E_F$ positioning relative to the magnetic WPs.

We next fabricate undoped CSS(1.8)/CoFeB(2)
(thicknesses in nanometer) trilayers [See Supplemental Material 20] for investigating the charge-to-spin conversion at $T = 300$ K, i.e., when CSS is paramagnetic. $t$ denotes the thickness of the CSS layer. Figure 3(a) shows a typical high-resolution cross-sectional transmission electron microscopy (HR-TEM) image of CSS(18.5)/Cu(1.8)/CoFeB(2) trilayer where the layered structure of high-quality CSS is clearly visible. The average grain size of CSS ($> 50$ nm) is significantly larger than that of Cu/CoFeB ($\sim 10$ nm). Energy dispersive x-ray spectroscopy (EDX) mapping confirms that all layers of the heterostructure are continuous with limited interdiffusion [See Fig. S1 in Supplemental Material 20]. Typical atomic force microscopy (AFM) micrograph [Fig. 3(b)] of the trilayer for $t = 7.9$ nm reveals flat surface morphology with a low root mean square (r.m.s.) surface roughness of $\sim 0.3$ nm, which is a prerequisite for spintronic device integration in the future.

On passing a charge current along $x$, CSS generates a spin current flowing along $z$ that traverses the Cu spacer and exerts SOTs on the CoFeB with in-plane magnetization. Limited by the thin film geometry, only $\sigma_{zz}^{DL}$ is accessible. The thin Cu spacer with long $\lambda$ physically separates CSS and CoFeB, thus avoiding local enrichment of Co at the interface which may alter the properties of CSS. The observation of $\sim 1\%$ current-in-plane giant magnetoresistance for the trilayer at $T = 50$ K (i.e., when CSS is ferromagnetic) confirms the finite spin transparency across the Cu spacer [See Fig. S2 in Supplemental Material 20]. We first employ the harmonic Hall technique 29, 30 to quantify the damping-like and field-like spin-orbit effective fields ($H_{DL}$ and $H_{FL}$, respectively) acting on the CoFeB magnetization [Fig. 3(c)]. The dependence of second harmonic Hall resistance $R_{2\omega}$ on the external field $H_{ext}$ and its azimuthal angle $\varphi$ allows separation of the SOT contribution $\propto \pm \theta$ from the parasitic thermoelectric effects 31, 32. Figure 3(d) plots the $\varphi$-dependence of $R_{2\omega}$ measured at various $H_{ext}$ for trilayer with $t = 7.9$ nm and a current density flowing in the CSS $j_{CSS} \sim$
1.6 × 10^6 A cm^{-2}. \( R_{DL}(\varphi) \) is dominated by the cos \( \varphi \) term, defined with a prefactor \( A \). [See Section S2 of the Supplemental Material for the detailed analysis of the harmonic Hall measurement.] \[ H_{DL} \] is extracted by linear fitting \( A \) against the inverse of the effective in-plane field \( 1/(H_x + H_{sat}) \) [insets of Fig. 3(b)]. We found \( H_{DL}/j_{CSS} = 1.4 \times 10^{-6} \) Oe A^{-1} cm^2, corresponding to a DL spin Hall efficiency \( \xi_{DL} = 2e H_{DL} M_{CoFeB} j_{CSS} \approx +0.10 \) where \( e \) is the elementary charge, \( h \) the Planck constant, \( M_s = 1200 \) emu cm^{-3} the saturation magnetization, and \( j_{CoFeB} = 2 \) nm the CoFeB thickness. \( \xi_{DL} \) of undoped PM-CSS is of the same sign as that of Pt. Its absolute magnitude is larger than another mWSM prototype Co_{0.8}Mn_{0.2}Ga (\( \xi_{DL} \sim -0.07 \) in the ferromagnetic state) \[24\]. With \( \rho_{xx} \approx 340 \) \( \mu \)Ω cm, we obtained \( \sigma_{xx}^S = \xi_{DL}/\rho_{xx} \approx 300(\hbar/2e)\Omega^{-1} \) cm^{-1}.

For an independent verification, we performed spin-torque ferromagnetic resonance (ST-FMR) [set-up depicted in Fig. 3(e)] measurement \[25\] on microstripes fabricated on the same substrate. A representative FMR spectrum measured from the mixing voltage \( V_{mix} \) while applying \( H_{sat} \) along \( \varphi = 45^\circ \) is shown in Fig. 3(f). Data is fitted by the sum of a symmetric and an antisymmetric Lorentzian. More details of the analysis can be found in the Section S3 of the Supplemental Material \[20\]. The emergence of an appreciable symmetric component (blue) confirms the generation of DL-SOT from the undoped PM-CSS at room temperature. Line-shape analysis taking into account current shuttling in the Cu spacer yields \( \xi_{DL} = +0.11 \), in excellent agreement with the harmonic Hall results. The full \( \varphi \) dependence of ST-FMR confirms the dominant role of spin current with polarization along \( y \) [See Fig. S4(a) in Supplemental Material \[20\]. Figures 3(g) and 3(h) summarize the CSS thickness \( t \)-dependence of \( \xi_{DL} \) and \( \sigma_{xx}^S \), obtained from the two techniques. The almost constant trend for \( \xi_{DL} \) and \( \sigma_{xx}^S \) against \( t \) is consistent with bulk-like SHE and \( \lambda \) much shorter than \( t = 7.9 \) nm.

We now extend the harmonic Hall SOT quantification at \( T = 300 \) K to Ni-substituted and In-substituted CSS/Cu/CoFeB trilayers. Contrary to \( \sigma_{xy} \) that is maximized for undoped FM-CSS, both \( \xi_{DL} \) and \( \sigma_{xx}^S \) [Figs. 4(a) and 4(b)] exhibit a pronounced peak for electron-doped PM shandites. The highest \( \xi_{DL} = +0.15 \) is achieved for PM-Co_{0.25}Ni_{0.75}Sn_{0.2}S_2, for \( x \sim 0.75 \), showing a large controllability of one order of magnitude by the composition variation. Taking into account the conductivity enhancement on increasing \( x \) [Fig. 2(f)], the peak of \( \sigma_{xx}^S \approx 500(\hbar/2e)\Omega^{-1} \) cm^{-1} is shifted to Co_{0.98}Ni_{0.02}Sn_{0.2}S_2, \( x \sim 1.0 \). Both \( \xi_{DL} \) and \( \sigma_{xx}^S \) decrease dramatically upon increasing Ni composition to \( x \sim 1.5 \) or introducing In for hole doping. A direct comparison of the harmonic Hall results for CSS(\( x \sim 1.0 \))/Cu/CoFeB trilayer and CSIS(\( y = 0.6 \))/Cu/CoFeB trilayer is shown in Fig. S3 in the Supplemental Material \[20\]. Compared to the \( \theta_{AHE,max} \) and AHC \( (\sigma_{xy}) \) peaks near \( x \sim 0 \) for FM-CSS [Right axes and dark yellow lines in Figs. 4(a) and 4(b)], reproduced from Figs. 2(g) and 2(h)], the observed peak shifts in the composition dependence of \( \xi_{DL} \) and \( \sigma_{xx}^S \) for PM-CSS agree with our calculations in Fig. 1. Since the conductivity of CSSNs is in the "moderately dirty" regime, one may consider the intrinsic contribution dominates the SHC. The observed evolution of SHC against \( x \) and \( y \) thus mainly reflects the relative position between \( E_F \) and a source of spin Berry curvature in momentum space, e.g., a gapped Dirac point. The intercorrelation between the AHE and SHE shown here may constitute a transport signature of Weyl-Dirac topological transition in mWSM and the associated redistribution of the electron’s filling \[21, 22\].

FIG. 4. (a,b) Nickel (Ni) composition \( x \) and Indium (In) composition \( y \) dependence of the damping-like spin Hall efficiency \( \xi_{DL} \) (a) and spin Hall conductivity \( \sigma_{xx}^S \) (b), for paramagnetic shandites based on the harmonic Hall technique, measured at 300 K. Right axes and dark yellow lines show the maximum anomalous Hall angle \( \theta_{AHE,max} \) and anomalous Hall conductivity \( \sigma_{xy} \) measured at low temperature. (c) \( \sigma_{xx}^S \) against the longitudinal conductivity \( \sigma_{xx} \) for all the samples. The calculated spin Hall conductivity maxima for \( \sigma_{xx}^S \) and \( \sigma_{xy}^S \) are indicated by the dashed lines.
potentially enhanced intrinsic SHE. This rule of thumb is best applied to ferromagnets where contribution from one spin channel dominates its intrinsic AHE and Berry curvature, as exemplified by the half-metallic mWSM Co$_3$Sn$_2$S$_2$. Another prototypical mWSM Co$_2$MnGa belonging to the highly tunable full Heusler family may also work, provided a proper dopant that simultaneously introduces additional electrons and reduces $T_C$ can be identified. Reciprocally, introducing a ferromagnetic hole dopant into well-established paramagnetic spin Hall materials may lead to the discovery of new ferromagnets with large AHC. This may explain the recent demonstration of large AHC in Li$_2$-ordered CrPt$_3$ compound. One should however be cautious in view of the metallic nature of these materials and the strong spin-orbit coupling of Pt. As a final remark, this strategy is readily extendable to the material screening for thermoelectric generation via the anomalous Nernst effect and spin Nernst effect, provided in addition the Mott relation is satisfied.

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