Critical thickness for the emergence of Weyl features in Co$_3$Sn$_2$S$_2$ thin films

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Magnetic Weyl semimetals are quantum phases of matter arising from the interplay of linearly dispersive bands, spin-orbit coupling, and time reversal symmetry breaking. This can be realised, for example, in Co$_3$Sn$_2$S$_2$, based on a cobalt kagome lattice and characterised by intriguing phenomena such as large anomalous Hall effect, Nernst effect, and water oxidation. Here, we attempt to determine the robustness of the twofold necessary conditions for the emergence of the magnetic Weyl semimetal phase in Co$_3$Sn$_2$S$_2$ ultrathin films. Except for two-dimensional layered materials, a reduction of thickness generally makes it difficult to develop topological character and ferromagnetic long-range order. In Co$_3$Sn$_2$S$_2$ films, while ferromagnetic ordering appears robustly even in average thicknesses of one or two unit cells with island-like polycrystalline domains, the anomalous Hall conductivity appears only above a critical thickness of approximately 10 nm. The emergence of surface conduction and large anomalous Hall effect implies the distinct contribution of Weyl nodes and their Berry curvature. These findings reveal an exotic feature of Weyl physics in thin-film based superstructures as well as a potential for future applications in electronic devices.
The inset shows the reduction is independently proposed in the monolayer of the kagome lattice in Co3Sn2S2. The quantum anomalous Hall effect has been theoretically tilted from the magnetisation direction along the large anomalous phenomena in Co3Sn2S2. The electronic structure of Co3Sn2S2 is schematically drawn in Fig.1b. Finite contribution of spin-orbit coupling (SOC) produces a gap at the band crossing points except for the protected spatial symmetry positions (from left to centre of Fig. 1b). This is defined as Dirac semimetal (DSM) with possessing helical Fermi arcs (FAs). With broken time-reversal symmetry, the mWSM phase emerges with chiral FAs owing to ferromagnetic exchange splitting (right of Fig. 1b). On the projected surface normal to the plane of Co3Sn2S2 films (Fig. 1c), the surface FAs contribute to provide electrical conduction because the pairs of Weyl nodes are tilted from the magnetisation direction along z axis. The Weyl nodes in the electronic bands enlarge the summation of Berry curvature under the well-regulated Fermi energy (E_F), inducing the large anomalous phenomena. Ultimately, the emergence of the quantum anomalous Hall effect has been theoretically proposed in the monolayer of the kagome lattice in Co3Sn2S2. While the benefit of the SOC is maintained in the ultrathin films, the robustness of the mWSM phase against the thickness reduction is independently influenced by the twofold stability of linear dispersive band and ferromagnetism. On the one hand, for topological materials such as three-dimensional topological insulators and DSMs, the topological phase disappears in ultrathin films owing to topological phase transition driven by hybridisation of surface states and crossover by quantum size effect, respectively. On the other hand, the stability of ferromagnetism in the thin-limit is an unresolved fundamental problem in Co3Sn2S2. The itinerant ferromagnetism in the Co-kagome lattice of Co3Sn2S2 is generally considered with the Stoner mechanism. Except for two-dimensional layered compounds such as CrI3 and Cr2Ge2Te6, however, it has been well known that the ferromagnetic order becomes drastically weakened in the thin-limit of conventional ferromagnetic metals.

Because of the difficulty in the exfoliation from bulk Co3Sn2S2 crystals, we investigate the magnetisation and electrical transport properties in the films approaching the thin-limit by careful synthesis of Co3Sn2S2 thin films with the varied film thickness t using vacuum deposition technique. We here disclose the robustness of mWSM phase in Co3Sn2S2 via characterisation of ferromagnetism and Weyl transport features in Co3Sn2S2 thin films. While the ferromagnetism with perpendicular magnetic anisotropy persists down to a few unit cell thicknesses with

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**Fig. 1 Thin films of kagome-lattice ferromagnet Co3Sn2S2.**

- **a** Crystal structure of Co3Sn2S2 and the ferromagnetic spin ordering below T_C drawn with VESTA.
- **b** Schematic band structures of nodal line semimetal, DSM, mWSM (FM: ferromagnetic coupling). Band crossing points, that is, Dirac and Weyl points, are represented by black and grey ellipsoids.
- **c** Distribution of Weyl points in energy-wave number (E-k) space and electrical conduction via chiral FA in Co3Sn2S2.
- **d** Typical out-of-plane XRD patterns of Co3Sn2S2 with varied t. Three different targets (A, B and C) were used to check the reproducibility of the film composition and quality. For t ≥ 40 nm, the film compositions determined by EDX are shown.
- **e** M versus T curves measured for t = 41, 10, 2.7 and 1.3 nm (fabricated with the target A) at μ0H = 10 mT. The inset shows the t dependence of T_C.
island-like polycrystalline domains, the mWSM phase with large anomalous Hall effect (AHE) emerges above a critical thickness of ~10 nm.

Results

Film growth and magnetisation measurements. We fabricated c axis oriented Co$_3$Sn$_2$S$_2$ films on Al$_2$O$_3$(0001) substrates by co-sputtering technique (Method). The $t$ was controlled from nominal 1.3 nm (roughly one unit cell) to 61 nm. Figure 1d shows typical X-ray diffraction (XRD) patterns of nearly stoichiometric Co$_3$Sn$_2$S$_2$ films capped with SiO$_x$ (a thickness of ~50 nm), where the systematic variation of thickness fringes indicates the uniform film quality and tunable manner of our t control at least down to t~10 nm (Supplementary Figs. 1 and 2). Although the diffraction intensity for t < 4 nm was very weak, the deposition rate for these films was reliably reproducible by the sputtering method. The surface morphology of thin films for t = 4 and 11 nm was fairly flat (Supplementary Fig. 3). Based on these observations, we apply the nominal value of t estimated from the deposition rate in the following discussions.

Figure 1e shows magnetisation M versus temperature T curves measured for films with nominal t values of 41, 10, 2.7 and 1.3 nm. A sharp rise in M occurs, respectively, at T = 181, 177, 143 and 130 K, which correspond to Curie temperature $T_C$. For t = 41 nm, the saturated value of M ($M_s$) of 0.29 $\mu_B$ per Co (where $\mu_B$ is Bohr magneton) at T = 10 K is comparable to the reported bulk value of 0.29–0.33 $\mu_B$ per Co$^{5-7,16,17}$. With decreasing t, both the $M_s$ and the $T_C$ (inset) decrease monotonically. Considering the Stoner mechanism$^{16,17}$, these decreases are induced by the reduction of the density of states (DOS) at $E_F$ in the ultrathin films. In fact, the Hall coefficient $R_H$ in the paramagnetic state is systematically decreased$^{5-3}$ (see Supplementary Fig. 4). Given that Co$_3$Sn$_2$S$_2$ thin films have semimetallic bands as in the bulk, the decrease in $R_H$ should correspond to the decrease of carriers that contribute to the ordinary Hall effect, that is, the decrease in the DOS at $E_F$. However, the monotonous systematic trend of $M_s$ and $T_C$ and $R_H$ at t > 4 nm in Supplementary Fig. 4 allows us to extrapolate the finite value of $R_H$ in the ultrathin region, indicating that the ferromagnetic order is maintained with finite DOS even in the ultrathin films.

Structure-property relationship in Co$_3$Sn$_2$S$_2$ films. In contrast to these monotonous changes in magnetism, electrical resistivity $\rho_{xx} (=R_{xx} \times t$, where $R_{xx}$ is the measured sheet resistance) dramatically increases with the t reduction. In Fig. 2a, b, the t dependences of $\rho_{xx}$ at (a) $T=200 \ K > T_C$ and (b) $2 \ K < T_C$ are shown, respectively. Here, $\rho_{xx}$ is estimated based on the assumption that electrical conduction is uniform over the whole region of a film without surface conduction. Above approximately t = 15 nm, both $\rho_{xx}$ values at T = 200 K and 2 K are virtually constant; the bulk component of electrical conduction is comparable in these thick films. In the intermediate range of ~3 nm < t < ~15 nm, $\rho_{xx}$ at T = 200 K increases as t decreases, whereas $\rho_{xx}$ at T = 2 K increases more significantly with a rough relation of $\rho_{xx} \propto t^{-1}$. Below t ~3 nm, the electrical resistance exceeds the measurement limit. To comprehensively understand the impact of t on $\rho_{xx}$ and also magnetism, we performed cross-sectional transmission electron microscopy (TEM) experiments for typical samples in these three t regions (nominal t = 45, 10 and 2.7 nm), as displayed in Fig. 2c-e. The 45-nm-thick sample is flat and the nominal t value estimated from the deposition rate is in excellent agreement with the t (t$_{obs}$) value observed by TEM. In the 10-nm-thick sample, crystalline domains form a continuous network albeit with t$_{obs}$ fluctuating between 8 and 12 nm. Island-like polycrystalline domains eventually appear in the nominal 2.7-nm-thick sample (t$_{obs}$ = 0–6 nm). Overall, the averaged t$_{obs}$ matches well with the nominal t value, guaranteeing that our calculation of M using the film volume is reasonable (Fig. 1e).

Thickness dependence of electrical and AHE properties. The anomalous electrical transport properties are pronounced on the t dependent systematic variation in Fig. 3. As shown in Fig. 3a, thick films with t ~20 nm exhibit metallic behaviour down to the lowest T of 2 K. The $\rho_{xx}$ shows an inflection near T = 180 K, which agrees well with $T_C$ detected by magnetisation measurements (Fig. 1e). In addition, two samples for t = 41 and 59 nm (broken lines) represent superior conduction behaviour in low T region with a comparable residual resistivity ratio to that for bulk single crystals$^{5-7,10}$. A decrease in t to <10 nm substantially increases the $\rho_{xx}$ without an inflection around $T_C$ (t ~5 and 4 nm) although M develops ferromagnetically. The $\rho_{xx}$ for the films with nominal t = 2.7 and 1.3 nm was undetectable above the measurement limit (Fig. 2a, b) because of the disconnection of film domains (Fig. 2e). In Hall conductivity, $\sigma_{xy} = \rho_{yx}/\rho_{xx}$ (Supplementary Fig. 6a for the T dependence of $\rho_{yx}$ in Fig. 3b, the $\sigma_{xy}$ dramatically increases at T comparable with $T_C$ for M (Fig. 1e) and saturates at low T. Although many of the thick films (t > 10 nm) show $\sigma_{xy}$ exceeding 1000 $\Omega^{-1}$ cm$^{-1}$ at T = 2 K, which is comparable or even higher than the bulk values$^{5,6}$ and theoretically calculated values$^{12}$, the $\sigma_{xy}$ for t = 5 and 4 nm is much suppressed. These bulk-comparable T-dependent $\rho_{xx}$ and $\sigma_{xy}$ strongly support that the AHE character in thick films t > 10 nm reflects the Weyl features of electronic bands. In addition to the high $\sigma_{xy}$, we observed a negative magnetoresistance in an in-plane H configuration, called the chiral anomaly$^{4-25}$, which has been discussed as a hallmark of the mWSM state$^5$ (Supplementary Fig. 7).

To find the surface-specific conductance contributions by surface FAs (Fig. 1c), we plot sheet conductance $\kappa_{xx}$ as a function of t in Fig. 3c, where $\rho_{xx}^{bulk}$ is the bulk resistivity of Co$_3$Sn$_2$S$_2$ film and $\sigma_t$ is the t-independent surface conductance. We here summarise all data acquired for films fabricated with the three sputtering targets A (circles), B (triangles) and C (squares).
Fig. 2 Structure-property relationship. a, b t dependence of \( \rho_{xx} \) at \( T = 200 \) K and 2 K, respectively. The symbols in a and b correspond to the used sputtering targets: A (circles), B (triangles) and C (squares). The red solid line in a is a guide to the eye, corresponding to the value of \( \rho_{xx}^{\text{bulk}} = 2.9 \times 10^{-4} \) \( \Omega \) cm. The blue solid line in b represents a rough relation of \( \rho_{xx} \propto t^{-1} \). These data suggest three \( t \) regions with distinct conduction mechanisms: mWSM for \( t > 15 \) nm, bad metal without surface conduction for \( 3 \) nm \( < t < 15 \) nm, and structurally disordered islands without electrical connection for \( t < 3 \) nm. c–e Cross-sectional TEM images for \( t = 45, 10 \) and 2.7 (nominal) nm, respectively. The \( t_{\text{obs}} \) values represent rough thicknesses of \( \text{Co}_3\text{Sn}_2\text{S}_2 \) estimated from the TEM images. f–h \( M \) versus \( \mu_0H \) curves for \( t = 41, 10 \) and 2.7 (nominal) nm, respectively. The used sputtering targets were A for f–h, B for d, and C for c.

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Stability of mWSM phase in \( \text{Co}_3\text{Sn}_2\text{S}_2 \) films. Using \( \sigma_{xy} \) and electrical conductivity \( \sigma_{xx} = \frac{\rho_{xx}^{\text{bulk}}}{\rho_{xx}^{\text{bulk}} + \rho_{xx}} \), we calculated a tangent of the Hall angle for AHE, \( \sigma_{xy}/\sigma_{xx} \), which is directly linked to the Berry curvature of electronic bands in the intrinsic mechanism. As shown in Fig. 4a, the 40-nm-thick film exhibits \( \sigma_{xy}/\sigma_{xx} \) as large as 0.24 at \( T = 130 \) K. With a decrease in \( t \), the \( T \) at which \( \sigma_{xy}/\sigma_{xx} \) becomes the largest shifts to low T. The T-dependent \( \sigma_{xy}/\sigma_{xx} \) can be apparently classified to two trends with/without peak at finite T (Supplementary Fig. 6b), which indicates that the AHE in all films is governed by the intrinsic mechanism identical to that for the bulk with different T-dependent \( \sigma_{xx} \).
Fig. 3 Critical $t$ for the emergence of Weyl features in electrical transport properties. a, b $T$ dependence of $\rho_{xx}$ and $\sigma_{xy}$ measured at zero field after field-cooled at $\mu_0 H = 1$ T, respectively. The used sputtering targets were A for $t = 41$ nm, B for $t = 40, 20, 10, 5$ and 4 nm, and C for $t = 59$ nm. c $t$ dependence of $R_s^{-1}$ at $T = 2$ K (blue symbols) and 200 K (red symbols). The red solid line is a linear fit to the data at $T = 200$ K. The blue solid and dashed lines for the data at $T = 2$ K represent two sample groups with different $\sigma_s$ values. The blue solid curve is a guide to the eye. d $t$ dependence of $\sigma_{xy}$ at $T = 2$ K. The blue solid curve is a guide to the eye. Symbols in c and d correspond to the used sputtering targets: A (circles), B (triangles) and C (squares).

Fig. 4 Phase diagram of Co$_3$Sn$_2$S$_2$ films for magnetic WSM. a $T$ dependence of $\sigma_{xy}/\sigma_{xx}$ measured for films with $t = 40, 20, 10, 5$ and 4 nm (fabricated with the target B) at zero field after field-cooled at $\mu_0 H = 1$ T. b Contour plot of $\sigma_{xy}/\sigma_{xx}$ as a function of $T$ and the nominal number of Co-kagome layers in the Co$_3$Sn$_2$S$_2$ films (PM: paramagnetic). The nominal number of Co-kagome layers was calculated using the c axis length determined by XRD (Supplementary Fig. 1). $T_C$ (Fig. 1e) and $T_{\text{peak}}$ are included for comparison. c Sheet Hall conductance $\sigma_{xy}$ at $T = 2$ K as a function of the number of Co-kagome layers. The solid line indicates the fitting result using a linear relation of $\log \sigma_{xy} = a + \log N_{\text{Co}}$. 
(Supplementary Fig. 8). To emphasise this trend, we made a contour plot of the $\sigma_{xy}/\sigma_{xx}$ as functions of $T$ and the sample thickness in Fig. 4b, where the nominal number of Co-kagome layers (three Co-kagome layers ~1.3 nm) is used as the sample thickness instead of $t$. The $T_c$ values (black circles) determined by magnetisation measurements (Fig. 1e) define the ferromagnetic/paramagnetic regions in the diagram. As discussed in Figs. 1e and 2, the average one- and two-unit-cells-thick samples with island-like polycrystalline domains maintain the ferromagnetic ordering, securing that the time reversal symmetry is broken in overall $t$ regions. It is now more obvious that a large $\sigma_{xy}/\sigma_{xx}$ region vanishes below a few tens Co-kagome layers, unveiling a mWSM phase emerges in the thicker region. The linear dispersive electronic bands would be intrinsically diminished in the thinner $t$ region similarly to band renormalisation in DSM of Cd$_3$As$_2$ thin film.\textsuperscript{15,27} Though it is difficult to fully understand the additional role of disorder by lattice distortion and roughness in the disappearance of AHE in the ultrathin films, the intrinsic band modification in ultrathin films may be examined in future study by spectroscopy after overcoming the difficulty of surface treatment or exfoliation.

Discussion

The final remark in this study is the verification of quantum Hall conductance $e^2/h$ ($e$: elementary charge, $h$: Planck constant) of one Co-kagome layer in the mWSM by a systematic extrapolation of sheet Hall conductance $\tau_{xy}$ shown in Fig. 4c, which has been theoretically expected in kagome layer with a flat band feature.\textsuperscript{3,11,12} The good agreement of a linear relationship in the certain region may exemplify the two-dimensional contribution of each Co-kagome layer to the Hall conductivity with close $E_F$ to the gap. In comparison with the 1.28 $e^2/h$ in each kagome layer calculated from the $\sigma_{xy}$ value of 1130 $\Omega^{-1}$ cm$^{-1}$ for bulk crystal,\textsuperscript{9} the extrapolated value of 1.3 $e^2/h$ by fitting in Fig. 4c using a linear relation of $\log(\tau_{xy}) = a + \log(N_{Co})$, where $a$ is a fitting parameter and $N_{Co}$ is the number of Co-kagome layer, is further reasonable (Supplementary Fig 9). The robust ferromagnetism in the ultrathin films, large AHE, and a verification of quantum conductance experimentally prove the significant feature of mWSM Co$_3$Sn$_2$S$_2$ thin films. Stabilisation of ultimate thin-limit of Co-kagome monolayer is a future interesting challenge to perform direct measurement of magnetic interaction in the layer and quantum conductance. In view of a wide variety of mWSMs and magnets with kagome lattice, heterostructure engineering and $E_F$ tuning will pave a way to find emergent phenomena that relate with Weyl nodes.

Methods

Thin-film growth. The Co$_3$Sn$_2$S$_2$ films were grown on Al$_2$O$_3$(0001) substrates by radio-frequency magnetron sputtering\textsuperscript{22} with Co–SnS$_2$$_{13}$ mosaic targets. The mosaic target was prepared using an SnS$_{1.35}$ disc (mixed-phase of SnS and SnS$_2$) and Co metal chips. The film composition was controlled by the number and location of the Co metal chips. Three Co–SnS$_2$$_{13}$ mosaic targets were used, which are referred to as A, B and C in the text. To suppress possible influences by impurities, e.g., unreacted/segregated ferromagnetic Co, particular attention was paid to the reproducibility of the film composition using the three different targets. Prior to the film growth, the substrates were annealed at 1000 °C in air to obtain atomically smooth surfaces. The films were deposited at 400 °C and then capped with SiO$_x$, followed by in situ annealing at 800 °C in a vacuum. The crystal structure and composition of the films were analysed by XRD using Cu $K\alpha$ radiation and energy-dispersive X-ray spectroscopy, respectively.

Electrical and magnetic measurements. Electrical measurements were performed with a Physical Property Measurement System (Quantum Design, Inc.). A Hall-bar shaped channel was patterned by mechanically scratching the film. Electrical contacts were made with indium solder. The measured $\mu_{H}$ versus $\mu_{H}$ curves were anti-symmetrized. The $T$ dependence of $\sigma_{xy}$ was obtained by anti-symmetrizing data taken at zero field after field-cooled at $\mu_{H} = 1$ T. Magnetisation measurements were performed with a Magnetic Property Measurement System (Quantum Design, Inc.). Before the measurements, the bottom surface of Al$_2$O$_3$ substrate was polished to remove possible magnetic contaminations from the substrate holder used. For the $\mu_{H}$ versus $T$ measurements in Fig. 1c, the samples were cooled from $T = 300$ K to 10 K in an out-of-plane magnetic field of $\mu_{H} = 1$ T. Subsequently, $\mu_{H} = 10$ mT was measured in a heating process. The measured $\mu_{H}$ versus $\mu_{H}$ curves were anti-symmetrized for a comparison with $\sigma_{xy}$ versus $\mu_{H}$ characteristics.

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Author contributions
J.I. and K.F. grew the films and measured the electrical transport properties. J.S. performed the magnetoresistance measurements. K.F. performed the magnetisation measurements under the support by J.S., T.S. and K.T. J.I. and K.F. analysed the measured data. K.N. contributed to theoretical interpretations of the experimental results. J.I., K.F. and A.T. wrote the manuscript with input from other authors. All authors discussed the results. A.T. supervised the project.

Competing interests
The authors declare no competing interests.

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