Synthesis of ultra pure Co$_3$Sn$_2$S$_2$ crystals

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Abstract

As a ferromagnetic topological semimetal with a strong anomalous Hall effect (AHE), Co$_3$Sn$_2$S$_2$ has been extensively explored for manipulating the topological state via the changed magnetic order as well as device applications connecting dissipationless spin and charge transport$^{1-3}$. However, the main challenge is the improvement of the crystal quality, which is crucial for enhancing physical properties including AHE, carrier mobility, and magnetoresistance$^{4,5}$. Here, we report the synthesis of ultra pure Co$_3$Sn$_2$S$_2$ single crystals with an ultra low impurity density by an innovative crystal-sourced chemical vapor transport (CS-CVT) approach, achieving ultrahigh anomalous Hall angle (AHA) of 40%, carrier mobility and magnetoresistance ($MR$) of 10490 cm$^2$V$^{-1}$s$^{-1}$ and 2500%, respectively. These values represent a huge improvement on previously reported highest values of either pristine or doped Co$_3$Sn$_2$S$_2$ (of 33%, 2600 cm$^2$V$^{-1}$s$^{-1}$, and 180% respectively)$^{4,6}$. Equally importantly, the anomalous Hall conductivity (AHC) of our crystals reached 1600 Ω$^{-1}$cm$^{-1}$, which is larger than the theoretically predicted value of 1310 Ω$^{-1}$cm$^{-1}$ from the integral of Berry curvature$^7$. Observation of an ultranarrow flat band and topological surface states near the Fermi level provides good microscopic understanding of this gigantic anomalous Hall effect. Our high-quality magnetic topological materials are expected to facilitate a full understanding of the strong electronic correlation and may perhaps lead to the discovery of a quantum AHE in the 2D limit.

Full Text

Ferromagnetic topological Weyl semimetals of Co$_3$Sn$_2$S$_2$ with a kagome lattice, have attracted great attention because they host frustrated spin configurations, correlated electron interactions, and topological quantum states of matter$^{8,9}$. Such a material with its fully polarized spin and topological properties can give rise to rich physical phenomena, such as a Berry curvature-induced anomalous Hall effect (AHE), negative orbital magnetism and Chern gapped topological fermions$^{10,11}$. Experimental evidence for the bulk Weyl nodes near the Fermi energy and the magnetic topological state was provided by angle-resolved photoemission spectroscopy (ARPES) and magnetotransport measurements$^{12,13}$. Chiral-anomaly-induced $MR$, a much high anomalous Nernst signal, and a quantum AHE predicted in the two-dimensional (2D) limit have also been reported$^{8,14-16}$. However, direct visualization for the flat band related to the exotic correlated electron phenomena has not been detected in Co$_3$Sn$_2$S$_2$ crystals and there has still been a controversial issue of physical properties on the S-terminated and Sn-terminated surfaces$^{15,17}$. Moreover, the very high mobilities and magnetoresistance usually exhibited in topological Weyl semimetals, attributed to the high Fermi velocity of massless Dirac states and uncompensated electron and hole carriers, still need to be achieved in the Co$_3$Sn$_2$S$_2$ crystals. Therefore, a significant improvement of the crystal quality is strongly desired.

High-quality kagome lattice materials are indeed difficult to prepare due to the requirement of the well-controlled 2D ideal kagome lattice formed by transition metal atoms as well as the elimination of vacancies and impurities$^{18,19}$. The typical Co$_3$Sn$_2$S$_2$ single-crystals prepared by the self-flux method
reported by Liu et al. have an anomalous Hall conductivity (AHC), anomalous Hall angle (AHA), electron mobility, and $MR$ of 1130 $\Omega^{-1}$cm$^{-1}$ at 2 K, 20%, 730 cm$^2$V$^{-1}$s$^{-1}$, and $\sim$ 55%, respectively$^{8,20}$. Thin films of Co$_3$Sn$_2$S$_2$ have been prepared by molecular beam epitaxy, but showing a much lower AHC of 649 $\Omega^{-1}$cm$^{-1}$ at 2 K and an AHA of 11.4% due to the poor compositional uniformity$^{21}$. Recently, the chemical vapor transfer (CVT) method$^{6,22}$ has been applied to grow crystalline Co$_3$Sn$_2$S$_2$ nanoflakes with enhanced AHC of $\sim$1400 $\Omega^{-1}$cm$^{-1}$, AHA of $\sim$32%, $MR$ of $\sim$180, and an electron mobility of $\sim$2600 cm$^2$V$^{-1}$s$^{-1}$. Numerical calculations have suggested that residual impurities are the limiting factor for mobility in ultra-high-quality materials$^{23}$. These results suggest that the transport properties of Co$_3$Sn$_2$S$_2$ crystals could be improved by a CVT method which results in a lower residual impurity and a corresponding lower concentration of carrier scattering centers.

In this article, we report the synthesis of high-quality Co$_3$Sn$_2$S$_2$ crystals with the highest carrier mobility and AHA reported so far by an innovative CVT approach using single-crystal granules as source materials (named crystal-sourced CVT method, CS-CVT). Low-temperature magnetotransport measurements show that the carrier mobility, AHA, and $MR$ of our high-quality Co$_3$Sn$_2$S$_2$ crystals are remarkably enhanced to over $10^4$ cm$^2$V$^{-1}$s$^{-1}$, 40% and 2500%, respectively. Moreover, an ultranarrow flat band and topological surface states near the Fermi level are observed, which provide good microscopic understanding of the gigantic anomalous Hall effect. The significant enhancement in transport properties and the observation of the topological bands are attributed to our ultra-high-quality sample, as demonstrated by a combination of x-ray diffraction (XRD), scanning transmission electron microscopy (STEM) and STM measurements. Our study provides a feasible way to improve the quality of magnetic topological semimetal materials, showing promise for pursuing new topological properties such as a high-temperature quantum AHE in the 2D limit, as well as possible quantum devices based on topological physics.

**Synthesis and structural characterizations of the ultra pure Co$_3$Sn$_2$S$_2$ single crystals**

The high-quality ultra pure Co$_3$Sn$_2$S$_2$ single crystals were synthesized by the CS-CVT method, where single-crystal granules of Co$_3$Sn$_2$S$_2$ were used as the source material and NH$_4$Cl as the transport agent (Fig. 1a). The Co$_3$Sn$_2$S$_2$ crystal has a rhombohedral structure with the space group, R-3m (No. 166), where Co atoms are arranged into a kagome lattice in the $ab$ plane with a Sn atom at the center of each hexagon (Fig. 1b). All as-grown single crystals show a hexagonal shape with a lateral size ranging from 1–8 mm and a thickness of about 0.01mm (Supplementary Fig. 1). A typical optical photograph in Fig. 1c presents a Co$_3$Sn$_2$S$_2$ hexagonal single crystal of 5.5 mm in lateral size, indicating the prior growth of $ab$ plane with the CoSn kagome lattice. Energy dispersive X-ray spectroscopy (EDS) measurement on the as-grown crystals reveals an atomic ratio of Co:Sn:S very close to 3:2:2 (Supplementary Fig. 2). The x-ray diffraction (XRD) pattern of the Co$_3$Sn$_2$S$_2$ single crystal only shows the $(00l)$ diffraction peaks, confirming the pure phase of the as-grown single crystal with a preferred [001] orientation (Fig. 1d). The lattice parameters $a$, $b$, and $c$ are measured by single crystal diffraction to be 5.3657 Å, 5.3657 Å and
13.1710 Å, respectively. The double-crystal x-ray rocking curve for the (003) Bragg reflection (Fig. 1e) demonstrates a much smaller crystal mosaic of 0.058°, as compared to 0.105° in typical crystals obtained by the self-flux method, indicating a high crystallinity with a low density of lattice defects. The h0l plane diffraction pattern of the Co$_3$Sn$_2$S$_2$ (Supplementary Fig. 3) shows sharp diffraction spots practically without any split diffraction spots, which confirms the excellent single crystal nature of the material.

To further confirm the high quality of our Co$_3$Sn$_2$S$_2$ crystals, we carried out atomic-scale structural and chemical analysis on cross-sectional samples using aberration corrected scanning transmission electron microscopy (STEM). Figures 1f,g show the simultaneously acquired STEM high-angle annular dark-field (HAADF) Z-contrast image and annular bright-field (ABF) image of the Co$_3$Sn$_2$S$_2$ sample along the [100] zone axis, with the structural models overlaid and main crystallographic planes indexed. The distinct spatial distributions of Co, Sn and S atoms were further confirmed by the atomic-resolution chemical mapping shown in Supplementary Fig. 4e. All the images clearly reveal the perfect crystalline structure of the Co$_3$Sn$_2$S$_2$ without noticeable structural defects. STM images were further used to measure the point-defect density. We identified two kinds of cleaved surfaces, Sn surface and S surface. From the typical 60 nm × 60 nm STM images on the S surface of Co$_3$Sn$_2$S$_2$ obtained by the self-flux and CS-CVT method in Supplementary Fig. 5a,b, the average density of impurities in the Co$_3$Sn$_2$S$_2$ crystal obtained by the self-flux method is $1.2 \times 10^{-2}$ atoms/nm$^2$, which is 6.7 times higher than that of the Co$_3$Sn$_2$S$_2$ crystal obtained by the CS-CVT method ($1.8 \times 10^{-3}$ atoms/nm$^2$) (Supplementary Fig. 5e). Similarly, for Sn surface, the average density of impurities in the Co$_3$Sn$_2$S$_2$ crystal obtained by the self-flux method ($2.8 \times 10^{-2}$ atoms/nm$^2$) is 25.5 times higher than that of the Co$_3$Sn$_2$S$_2$ crystal obtained by the CS-CVT method ($1.1 \times 10^{-3}$ atoms/nm$^2$) (Supplementary Fig. 5c-d,f). It implies only 1 residual impurity for every 1500 S atoms or every 2400 Sn atoms in the Co$_3$Sn$_2$S$_2$ crystal obtained by the CS-CVT method. These results demonstrate that the use of high purity source materials with exact stoichiometry and clean vapor transportation process in the CS-CVT growth, can indeed significantly improve the crystal quality of Co$_3$Sn$_2$S$_2$, without introducing residual atoms of flux agents.

The following question is why the crystal quality using CS-CVT is better than other growth methods previously reported. Compared to the self-flux method, the CS-CVT method can avoid contamination from flux agents, and provide stable thermal dynamics under a suitable constant growth temperature, preferring a growth in (00l) plane of Co$_3$Sn$_2$S$_2$, resulting in the formation of perfect kagome plane. Furthermore, using Co$_3$Sn$_2$S$_2$ single-crystal granules as the precursor ensures the purity of source materials and the initial stoichiometric ratio of Co, Sn, S elements. Compared to CVT method previously reported 6,22, what we used is the NH$_4$Cl as transport agent, which is decomposed to NH$_3$ and HCl gas at about 300 °C, providing a clean crystallization process. The Cl ions have a strong transport ability for Co and Sn ions, leading to a growth of Co$_3$Sn$_2$S$_2$ with the stoichiometric ratio at a cleaner condition and single-atom process.
Magnetization, resistivity, and large magnetoresistance

In the temperature dependence of magnetization of the Co$_3$Sn$_2$S$_2$ crystal obtained by the CS-CVT method, a sharp magnetic transition was observed with a Curie Temperature ($T_c$) of 177 K (Fig. 2a and Supplementary Fig. 6). A non-collinear magnetic transition around 150 K is unnoticeable due to the high quality of the Co$_3$Sn kagome lattice in our sample. The field dependence of out-of-plane magnetization reveals a large coercivity ($H_c\sim5000$ Oe), while the in-plane magnetization displays no sign of saturation in magnetic fields up to 2 T, suggesting that the Co$_3$Sn$_2$S$_2$ crystal has strong magnetic crystalline anisotropy (Supplementary Fig. 6e,f). From the temperature-dependent resistivity curves (Fig. 2b), a kink at $T_c = 181$ K is clearly observed, which is slightly higher than the 177 K observed in magnetization measurements. The residual resistivity ratio (RRR) $\rho_{300K}/\rho_{2K}$ is over 128, which is about one order of magnitude higher than the values of 5–20 reported in previous works$^{7,8,22}$, indicating the high quality of the present Co$_3$Sn$_2$S$_2$ crystals. Importantly, we firstly observed a magnetic-field-induced metal-insulator-like transition, suggesting a high carrier mobility in the present Co$_3$Sn$_2$S$_2$ crystal, which is similar to other topological semimetals such as LaSb, TaP and NbP$^{24,25}$. The inset in Fig. 2b shows the conductivity reaches $2.3 \times 10^5$ $\Omega^{-1}$cm$^{-1}$ at 2 K, which is one order magnitude higher than the reported values$^8,22$, indicating that our Co$_3$Sn$_2$S$_2$ single crystal is close to clean-limit regime$^{26}$.

Figure 2c manifests the magnetic field dependence of transverse magnetoresistance under out-of-plane and in-plane magnetic fields at different temperatures. Notably, the non-saturated positive $MR$ reached 2500% under a magnetic field of 15 T at 2 K at $B \perp I$, which is one order of magnitude larger than the previously reported results ranging from 55 to 250% in Co$_3$Sn$_2$S$_2$ crystals$^{6,8,27-31}$. The large $MR$ in the present Co$_3$Sn$_2$S$_2$ crystals is ascribed to a high carrier mobility and a low scattering rate of the carriers. Besides, a negative $MR$ due to the chiral anomaly was observed at high temperature regime. The chiral-anomaly-induced $MR$ is attributed to the Dirac band of the kagome lattice$^{32}$. Longitudinal magnetoresistances measured with $B$ parallel to the current at various temperatures in Fig. 2d show a net weakly positive $MR$ below 40 K and a negative $MR$ above 40 K due to the chiral anomaly, which represents a crucial peculiarity of Weyl fermions for Co$_3$Sn$_2$S$_2$ crystals$^{22,33}$. The different behavior of the longitudinal and the transverse magnetoresistance is on account of the strong out-of-plane anisotropy.

Gigantic AHE and underlying mechanism

The field-dependent $\rho_{yx}$ measured at different temperatures from 2 K to 180 K, in an out-of-plane configuration with $B \perp I$, shows the coexistence of AHE with hysteretic behavior and the normal Hall effect (NHE) with nonlinear profile (Supplementary Fig. 8). The $\rho_{yx}$ versus temperature shows a remarkable nonlinear change from 2 K to 20 K, which is attributed to the dual-carrier conduction mechanism. The field-dependence of the Hall conductivity ($\sigma_{yx}$) from 2 K and 160 K calculated according to the formula

$$\sigma_{yx} = \frac{-\rho_{yx}}{\rho_{yx}^2 + \rho_{xx}^2} \quad (\text{Fig. 3a and Supplementary Fig. 9}),$$

exhibits significant nonlinear behavior as the temperature decreases, indicating the enhancement of the dual-carrier conduction mechanism. The
shaded areas in Fig. 3a represent the contribution of AHE. After subtracting the AHE signals, we thus extract the carrier densities and carrier mobilities by using the semiclassical Drude model (Supplementary Fig. 10a,b). The extracted carrier densities of electrons and holes (2 K) are about $7.9 \times 10^{19}$ cm$^{-3}$ and $2.2 \times 10^{20}$ cm$^{-3}$, respectively, and the corresponding mobilities of electrons and holes at 2 K are $10490$ cm$^{-2}$V$^{-1}$s$^{-1}$ and $2480$ cm$^{-2}$V$^{-1}$s$^{-1}$, respectively (Supplementary Fig. 10c). The electron mobility observed in our Co$_3$Sn$_2$S$_2$ crystals is the largest among all reported magnetic topological semimetals, (e.g., Co$_3$Sn$_2$S$_2$ $\sim 2600$ cm$^{-2}$V$^{-1}$s$^{-1}$, MnBi$_2$Te$_4$ $\sim 3110$ cm$^{-2}$V$^{-1}$s$^{-1}$, GaPtBi $\sim 1500$ cm$^{-2}$V$^{-1}$s$^{-1}$, Co$_2$MnGa $\sim 35$ cm$^{-2}$V$^{-1}$s$^{-1}$). The carrier mobility and $MR$ are useful metrics to quantify the quality of a semimetal crystal. The electron mobility is straightforward to measure and is inversely proportional to the average electron scattering rate in semimetal crystals. High mobility values in crystals imply that electrons are less likely to scatter over prolonged trajectories. The electrical transport in a semimetal usually consists of two types of carriers (electrons and holes) and an extremely high MR is usually attributed to a perfectly balanced hole-electron resonance condition that is much stronger than that in normal metals and semiconductors. Thus, it is crucial to obtain high-purity Co$_3$Sn$_2$S$_2$ samples to realize a balance between electrons and holes and a high carrier mobility ($\mu$), both of which will enhance the $MR$ effect.

To study the large AHE of the high-quality Co$_3$Sn$_2$S$_2$ crystals, we subtract the NHE contributions according to the dual-carrier Drude model (Supplementary Fig. 11, 12). The AHC $\sigma_{yx}^A$ shows a strong dependence on temperature and reaches a maximum of $1600$ $\Omega^{-1}$cm$^{-1}$ around 50 K (Fig. 3b), which is the largest value among all the reported pure Co$_3$Sn$_2$S$_2$ crystals so far. In addition to a large AHC, the Co$_3$Sn$_2$S$_2$ crystals show a gigantic AHA of $\sigma_{yx}^A / \sigma_{xx}$. The temperature dependence of $\sigma_{yx}^A / \sigma_{xx}$ (Fig. 3c) shows that AHA firstly increases and then decreases with increasing temperature. The $\sigma_{yx}^A / \sigma_{xx}$ exceeds 30% within a broad temperature range of 100 K to 160 K, and has a maximum of approximately 40% around 140 K. It is well established that a large intrinsic AHE is present in the magnetic Weyl semimetal Co$_3$Sn$_2$S$_2$ due to the topologically enhanced Berry curvature, resulting in a giant intrinsic AHC. The theoretical AHC obtained from the integral of Berry curvature is about $1310$ $\Omega^{-1}$cm$^{-1}$ at 0 K$^7$. However, the maximum AHC measured in our Co$_3$Sn$_2$S$_2$ crystals reaches $1600$ $\Omega^{-1}$cm$^{-1}$ (Fig. 3d), suggesting that other mechanisms exist in the high-quality Co$_3$Sn$_2$S$_2$ crystal. Smit and Berger proposed the skew scattering and side-jump effects that are affected by the spin-orbit interaction$^{37,38}$. However, such mechanisms are usually observed in materials with a high conductivity, larger than $10^4$ $\Omega^{-1}$cm$^{-1}$, since the skew scattering can be suppressed by the increasing impurity scattering$^{26}$. Indeed, the conductivity of our Co$_3$Sn$_2$S$_2$ crystals reaches $10^4$ $\Omega^{-1}$cm$^{-1}$ around 100 K and rapidly increases to $2.3 \times 10^5$ $\Omega^{-1}$cm$^{-1}$ at 2 K (Supplementary Fig. 13a). We also note that the measured AHC reaches the theoretical maximum for the intrinsic mechanism ($1310$ $\Omega^{-1}$cm$^{-1}$) at 100 K. With further decreasing temperature, the AHC first increases to its maximum ($1600$ $\Omega^{-1}$cm$^{-1}$) around 50 K then decreases to about $970$ $\Omega^{-1}$cm$^{-1}$ at 2 K with a linear dependence on the longitudinal conductivity $\sigma_{xx}$, as shown in Fig. 3d. Our results are very different from the previous results$^8$, where AHC is almost independent to longitudinal conductivity $\sigma_{xx}$ in the relatively low temperature range.
To understand the origin of the large AHC, we study the relationship between the anomalous Hall resistivity $\rho_{yx}$ and the longitudinal resistivity $\rho_{xx}$ by using the formula $\rho_{xy}^A = (a \rho_{xx} + b \rho_{xx}^2) \cdot M$. The first term represents the skew scattering contribution, while the second term corresponds to the intrinsic or side-jump contributions. Both parts are linearly proportional to magnetization $M$. As shown in Fig. 3e, we plot the $\rho_{yx}/(\rho_{xx} \cdot M)$ against $\rho_{xx}$, which exhibits a good linear relationship below 80 K. Therefore, we extract the contribution from the skew scattering, and the sum of side-jump and intrinsic contributions. The results are shown in Fig. 3f. We find that the fitted AHC of $\sigma_{\text{total}}$ is in good agreement with the experimentally observed values. Remarkably, the skew scattering gives a negative contribution to AHC, which is consistent with previous results$^{6,7}$. Moreover, it becomes more significant with decreasing temperature due to the enhancement of the longitudinal conductivity. Therefore, we conclude that the reduction of AHC below 50 K is ascribed to the large negative contribution from skew scattering.

To distinguish the intrinsic and side-jump contributions to the AHC, we adopt a new scaling relation for the Hall conductivity $\sigma_{yx}$ and longitudinal conductivity $\sigma_{xx}$ in the limit of $\rho_{yx} \ll \rho_{xx}$ range. As shown in Supplementary Fig. 13b, we find that the intrinsic AHC is about 1400 $\Omega^{-1}\text{cm}^{-1}$, close to the theoretical value. On the other hand, we also note that the skew scattering is almost negligible when the temperature is between 50 K and 80 K. In this range, therefore, the AHC increases with decreasing temperatures and exceeds the maximum intrinsic value, which is mainly due to the significant contribution from the side-jump effects. The observation of remarkable contributions of skew scattering and side-jump to AHE in our Co$_3$Sn$_2$S$_2$ crystals further indicates that the sample is close to the clean-limit, i.e., ultra-low densities of defects and impurities$^{39}$. We compare the previously reported key transport parameters of Co$_3$Sn$_2$S$_2$ crystals and other magnetic topological materials in Supplementary Figs. 14 and 15, and the values of the carrier mobility, AHA, and $MR$ observed in our Co$_3$Sn$_2$S$_2$ samples are all the largest$^{40-46}$.

**Visualization of ultranarrow kagome flat bands using ARPES.**

To understand the possible mechanism for the great improvement in the electronic transport properties of Co$_3$Sn$_2$S$_2$ crystals, its Fermi surface and band structures were measured by high-resolution vacuum ultraviolet (VUV) laser ARPES at 20 K$^{47}$. Figure 4a shows the Fermi surface mapping of Co$_3$Sn$_2$S$_2$ crystals by integrating the spectra weight within ±10 meV energy window with respect to the Fermi level. The Fermi surface is obtained by symmetrizing the original mapping considering the three-fold and mirror symmetry. Six small pockets are clearly observed firstly surrounding the Brillouin zone center $\Gamma$. The experimental Fermi surface in Fig. 4a is in conformity with the projections of the calculated Fermi surface (Fig. 4b). For the sake of understanding the origin of the strong spectral weights around the Brillouin zone corner $k(k')$, two tangent lines (marked Cut1 and Cut2 in Fig. 4a) are taken along two different momentum directions to display the band structure near point K. The detailed band structures near $E_F$(Cut1) along $\Gamma$-K directions are shown in Fig. 4c. The topological surface state marked by the red circle (labeled TSS) is observed with $k_F$ about 0.4 Å$^{-1}$ away from the K point, which can be more clearly identified in the corresponding second derivative spectrum. The positions of the topological surface
states observed in the experiment are consistent with the results reported previously\textsuperscript{48}. The topological surface states across the Fermi level are usually considered to enhance the carrier mobility due to the low effective mass\textsuperscript{49}.

Moreover, around the K point, a nearly dispersionless flat band is captured (obviously presented in its relevant second derivative spectra). The flat band around the K point can expand a larger momentum space along the momentum Cut2 (indicated by red arrow in Fig. 4d). The corresponding EDCs are shown in Fig. 4e. Around the Fermi level, all the EDCs show very sharp peaks and the linewidths are all below 10 meV. Subtracting the temperature broadening of 7 meV at 20 K, the intrinsic line widths are about 6–7 meV. Such an ultranarrow flat band directly visualized by ARPES was first reported in the kagome materials. In order to understand the origin of the flat bands, the band structure of Co\textsubscript{3}Sn\textsubscript{2}S\textsubscript{2} is calculated based on relativistic density functional theory (DFT) projected on the Sn plane (Fig. 4f). The red line highlights the manifestation of the kagome flat band, showing quite flat and dispersionless behavior around both the Γ and K points. The calculated results are quite consistent with our ARPES results. In detail, the calculated band bottom around Γ is slightly above the Fermi level and the band bottom around K is just below the Fermi level. This also agrees with our ARPES results that a strong flat spectral weight is measured around the K point and quite weak spectral weight is measured around the Γ point.

Combining the ARPES results and the band calculation, we conclude there is a kagome flat band around the Fermi level in Co\textsubscript{3}Sn\textsubscript{2}S\textsubscript{2} which means the electron kinetic energy is strongly quenched by quantum interference effects, preventing delocalization of the wave function across the lattice. The appearance of flat bands is expected to increase the distance between a pair of Weyl nodes with opposite chirality which is nearly proportional to intrinsic AHE in magnetic Weyl semimetal magnetic\textsuperscript{50,51}.

In summary, using a “crystal-sourced chemical vapor transport” approach, we have successfully synthesized large-size, high-quality, hexagonal Co\textsubscript{3}Sn\textsubscript{2}S\textsubscript{2} single crystals with an ultra-low impurity density. The improvement of crystal quality enables the first observation of an ultranarrow topological flat band and makes a huge enhancement for nearly all the physical properties of Co\textsubscript{3}Sn\textsubscript{2}S\textsubscript{2}. Our high-quality samples display an ultrahigh AHA of 40%, intrinsic carrier mobility of 10490 cm\textsuperscript{2}V\textsuperscript{-1}s\textsuperscript{-1}, AHC of 1600 Ω\textsuperscript{-1} cm\textsuperscript{-1}, and MR of 2500%, which are the highest among all reported results for Co\textsubscript{3}Sn\textsubscript{2}S\textsubscript{2}. Besides the contribution to AHE from Berry curvature, the observation of the skew scattering and side-jump effects on AHE suggests that our sample is close to the clean-limit regime. This CS-CVT method can be extended to fabricate other kagome materials with improved crystal quality and physical properties compared with other methods reported in the past. For example, we have successfully synthesized Co\textsubscript{3}In\textsubscript{2}S\textsubscript{2} and Ni\textsubscript{3}In\textsubscript{2}S\textsubscript{2} crystals of the best quality and highest carrier mobility (Supplementary Figs. 16 and 17). The present results provide a useful way towards the growth of high-quality magnetic topological materials and suggest a bright future for the investigation of exotic physics in topological materials and kagome lattice materials.

Methods
Growth of the Co$_3$Sn$_2$S$_2$ single crystals.

The hexagonal Co$_3$Sn$_2$S$_2$ single crystals were successfully synthesized by a new chemical vapor transport approach using single-crystal granules as source materials (the CS-CVT method). First, single crystals of Co$_3$Sn$_2$S$_2$ with no regular shape were prepared by the self-flux method. Raw materials of Co, Sn, and S were mixed in a molar ratio of Co:Sn:S = 3:2:2 and placed in a crucible, then sealed in a quartz tube using a high temperature flame gun under high vacuum. The quartz tube was slowly heated for 55 hours to 1000 °C, then kept constant at that temperature for 72 hours and slowly cooled to 600 °C at a rate of 2 °C/h. Subsequently, the single-crystal granules were sealed in an evacuated quartz tube with NH$_4$Cl as a transport agent. The hexagonal Co$_3$Sn$_2$S$_2$ single crystals were produced by CS-CVT in a temperature gradient from 1000 °C to 900 °C for about a week.

Sample characterization.

XRD patterns were collected using a Rigaku SmartLab SE X-ray diffractometer with Cu K$_\alpha$ radiation ($\lambda = 0.15418$ nm) at room temperature. Single crystal diffraction patterns and rocking curves were taken by a Bruker D8 Venture. Scanning electron microscopy (SEM) and X-ray energy-dispersive spectroscopy (EDS) were performed using a HITACHI S5000 with an energy dispersive analysis system Bruker XFlash 6|60. Magnetization measurements were carried out on oriented crystals with the magnetic field applied along both the $a$ and $c$ axes using a vibrating sample magnetometer (MPMS 3, Quantum Design). Both in-plane electrical resistivity and Hall resistivity data were collected on a Quantum Design Physical Properties Measurement System (PPMS).

Scanning transmission electron microscopy and electron energy-loss spectroscopy.

Electron-transparent lamellas for STEM investigation were prepared by conventional lift-off focused-ion-beam (FIB) methodology using a Thermo Scientific Helios G4 CX DualBeam system, operated at accelerating voltages of 30 kV down to 2 kV to ensure minimum sample damage. Aberration-corrected STEM characterizations were performed using a Nion HERMES-100, operated at 100 kV and a probe forming semi-angle of 30 mrad. HAADF and ABF images were acquired simultaneously using two annular detectors with collection semi-angle of 92–210 and 15-30 mrad, respectively. EELS measurements were performed using a collection semi-angle of 91 mrad, an energy dispersion of 0.5 eV per channel, and a probe current of $\sim 20$ pA. The Co-L (779 eV), Sn-M (485 eV), and S-L (165 eV) absorption edges were integrated for elemental mapping after background subtraction. The parent spectrum image was processed with the principal component analysis (PCA) tool to reduce random noise.

Scanning tunneling microscopy/spectroscopy.

The experiments were carried out in a Createc low-temperature STM system in ultra-high vacuum (UHV) with a base pressure better than $3.0\times10^{-10}$ mbar. STM characterization was performed with a Pt-Ir tip in constant current mode and bias refers to the voltage on the sample with respect to the tip. The STM
scanning was performed on surfaces of Co$_3$Sn$_2$S$_2$ crystals that were freshly cleaved at 10 K in the UHV chamber. The measurements were conducted at 4.5 K. The images were processed using the WSxM software package.

**High resolution ARPES measurements.**

High-resolution angle-resolved photoemission measurements were carried out at 20 K by using ultraviolet laser as the light source that can provide a photon energy of $h\nu = 6.994$ eV with a bandwidth of 0.26 meV. The energy resolution was set at $\sim 2.5$ meV for the measurements. The angular resolution was $\sim 0.3^\circ$. The Fermi level was referenced by measuring on a clean polycrystalline gold that was electrically connected to the sample. The sample was cleaved in situ and measured in vacuum with a base pressure better than $5 \times 10^{-11}$ Torr.

**DFT calculations**

DFT calculations were performed using the generalized gradient approximation for the exchange-correlation potential, the projector augmented wave method\textsuperscript{52} and a plane-wave basis set as implemented in the Vienna ab-initio simulation package\textsuperscript{53,54}. The energy cutoff for the plane-wave basis was set to 400 eV for structural relaxations and 600 eV for the energy and electronic structure calculations. Two k-meshes of $7 \times 7 \times 1$ and $11 \times 11 \times 1$ were adopted for the structural relaxations and total energy (electronic structure) calculations, respectively. The mesh density of the k points was fixed when performing the related calculations with primitive cells. In geometric structure relaxation, van der Waals (vdW) interactions were considered at the vdW-DF level with the optB86b functional as the exchange functional (optB86b-vdW)\textsuperscript{55,56}. Symmetrical slab models were employed, and the surface atoms were fully relaxed until the residual force per atom was less than 0.005 eV/Å. To avoid image interactions between adjacent unit cells, a vacuum layer of more than 20 Å thick was added to the slab cell perpendicular to the surface. The optimized lattice constants of bulk Co$_3$Sn$_2$S$_2$ are 5.37 and 13.15 Å along the a and c directions, respectively.

**Declarations**

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**Author Contributions:** H.-J.G. designed the project. H.T.Y., S.H.L. and K.Z. prepared the samples. S.H.L., J.H., and H.T.Y. performed the magnetization measurements. S.H.L., H.G., G.X., Q.Z.C., Q.Q. and H.T.Y. performed the transport experiments. Y.L., H.C., Q.Z. and L.H. performed the STM and nc-AFM
experiments with the guidance of H.-J.G. R.G. and W.Z. performed the STEM experiments. J.G.Y, L.Z. and X.J.Z. perform the ARPES experiments. All authors participated in the data analysis and manuscript writing.

**Competing Interests:** The authors declare that they have no competing interests.

**Data availability**

Data measured or analyzed during this study are available from the corresponding author on reasonable request.

**References**


**Figures**
Figure 1

**CS-CVT synthesis strategy for ultra pure Co$_3$Sn$_2$S$_2$ single crystals and structural characterizations.** a, Schematic of the CS-CVT synthesis process for ultra pure Co$_3$Sn$_2$S$_2$ single crystals. The Co$_3$Sn$_2$S$_2$ crystals prepared by self-flux method were placed with a small amount of NH$_4$Cl as the transport agent at the high-temperature part. After about a week, large-size, ultra pure, regular-hexagonal Co$_3$Sn$_2$S$_2$ single
crystals were collected at the low-temperature part. **b**, Unit cell in a hexagonal setting, consisting of two hexagonal planes of S and Sn as well as a Co$_3$Sn kagome layer sandwiched between the S atoms. Blue, green and pink balls represent Co, Sn and S atoms, respectively. **c**, An optical photograph of the hexagonal Co$_3$Sn$_2$S$_2$ crystal prepared by the CS-CVT method. **d**, The x-ray 2θ scan conducted on the $ab$ plane of the Co$_3$Sn$_2$S$_2$ crystal prepared by the CS-CVT method with the corresponding Miller indices (00$l$). **e**, The X-ray rocking curves of (003) reflection for samples prepared by self-flux (black) and CS-CVT (red) methods, showing a smaller FWHM of 0.058° for the Co$_3$Sn$_2$S$_2$ crystal prepared by the CS-CVT method. **f**, **g**, Simultaneously acquired high-resolution HAADF and ABF images of the Co$_3$Sn$_2$S$_2$ crystal structure seen along the [100] zone axis, up and down respectively. The inset in the HAADF is a close-up image with the atomic model of one unit cell overlayed, where blue balls correspond to Co, green to Sn and pink to S atoms. The inset in the ABF is an FFT pattern with the main planes indexed.
Fig. 2 | Magnetization, electrical resistivity, and magnetoresistance of Co₃Sn₂S₂ crystals. a, Temperature dependence of magnetization with ZFC and FC procedures of Co₃Sn₂S₂ crystals under 20 Oe. b, Temperature dependence of the resistivity under magnetic fields of 0 T, 5 T, and 9 T along z direction (c axis), respectively. The inset shows temperature dependence of \( \sigma_{xx} \) at zero magnetic field. c, Magnetoresistance measured under applied fields up to 15 T at different temperatures from 2 K to 200 K for \( B \perp I \), showing a non-saturated positive magnetoresistance below 100 K and a non-saturated negative magnetoresistance above 100 K. d, Magnetoresistance measured under applied fields up to 15 T at different temperatures from 2 K to 200 K for \( B \parallel I \), showing a complex magnetoresistance behavior below 40 K and a non-saturated negative magnetoresistance above 40 K.

Figure 2

See image above for figure legend.
**Fig. 3** | Transport measurements of the gigantic AHE. 

**a,** The field-dependent $\sigma_{y\chi}$ measured at 2 K and 50 K in an out-of-plane configuration with $B \perp I$, showing AHE with a hysteretic behavior. The contribution of AHE is marked by shaded areas. 

**b,** Temperature dependences of the AHC ($\sigma^A_{y\chi}$), showing a AHC maximum of $1600 \Omega^{-1}\text{cm}^{-1}$ at 50 K. 

**c,** Temperature dependence of the AHA ($\sigma^A_{y\chi}/\sigma$) at zero magnetic field, showing a maximum of 40% at 140 K. 

**d,** $\sigma^A_{y\chi}$ dependence of $\sigma_{xx}$ at zero magnetic field from 2 K to 100 K, displaying a decreasing behavior below 50 K. 

**e,** The $\rho_{yx}/\rho_{xx}/M$ dependence of $\rho_{xx}$ from 2 K to 80 K, indicating the contribution of the intrinsic and extrinsic mechanism on the large AHE. 

**f,** Temperature dependence of the AHC ($\sigma^A_{y\chi}$) at the zero magnetic field, indicating the intrinsic mechanism and the skew scattering mechanism.

**Figure 3**

See image above for figure legend.
Figure 4

Electronic structure and observation of a flat band around the Fermi level. **a**, Fermi surface mapping acquired by integrating the spectral weight within a ± 10 meV energy window. The red dotted line along $K_y$ is marked as Cut 1. The red dotted along $K_x$ is labelled as Cut 2. Cuts 1 and 2 signify the momentum locations of the experimental band structures in (c) and (d), respectively. **b**, Projections of calculated Fermi surfaces. Blue lines denote the Brillouin zones. **c**, The band structure at Cut 1 in (a) and the corresponding second derivative, indicating the existence of TSS marked by the red arrow. **d**, The band structure at Cut 2 in (a), showing a flat band near the Fermi level. **e**, EDC of Cut 2 at different $K_x$, showing a narrow coherent peak corresponding to the flat band with a large DOS near Fermi level. **f**, Calculated band dispersions along different high-symmetry directions on the Sn-terminated surface, showing a nearly flat band near the Fermi level.

Supplementary Files

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