I. Introduction

Magnetic topological materials have attracted tremendous recent attention\([1–4]\). \(\text{Co}_3\text{S}_2\text{Sn}_2\) is one of them, which has a kagome lattice and becomes ferromagnetic below 177 K\([5]\). It is a half-metal, as the spin degeneracy of its carriers is almost entirely lifted in the ferromagnetic phase\([6, 7]\). It is a semimetal with an equal number of electrons and holes. The density of these carriers is in the range of \(10^{19} \text{cm}^{-3}\)\([9]\), so that one electron and one hole is shared by more than a thousand atoms.

The observation of a sizeable anomalous Hall effect in this context makes this solid an appealing platform. Because of the low carrier density, the anomalous Hall angle becomes remarkably large\([10, 11]\), as large as 33\% in iron-doped samples at low temperature\([12]\). Angle-resolved photoemission spectroscopy\([13]\) and scanning tunneling microscopy\([14]\) and magneto-optical studies\([15]\) confirm that it is a magnetic Weyl semimetal with a topologically non-trivial electronic structure. The high mobility of carriers in this low-density semi-metal generates an ordinary Nernst coefficient\([8]\) on top of the anomalous Nernst response. A study of its thermoelectric response\([9]\) has documented the contrasting evolution of the ordinary and anomalous components of the Nernst response. As expected in a Berry-curvature picture, increasing disorder enhances the anomalous Nernst effect and reduces the ordinary Nernst effect.\([9]\).

In this paper, we present two distinct experimental studies of \(\text{Co}_3\text{S}_2\text{Sn}_2\). First of all, we report on the fermiology of this system by studying quantum oscillations on high-mobility crystals in presence of strong magnetic fields. Our sample has a residual resistivity ratio (RRR) of 30 and its mobility is as large as \(2.7 \times 10^3 \text{cm}^2\text{V}^{-1}\text{s}^{-1}\). Using pulsed magnetic fields up to 61 T and low temperatures, we tracked quantum oscillations with magnetic field perpendicular and parallel to the \(z\)-axis([0001]). The angle dependence of the observed frequencies was compared with what is expected according to the density functional theory (DFT) calculations. There were consistent. In addition, we found an unexpected frequency emerging at high field and attributed it to a magnetic breakdown orbit between a hole-like and an electron-like orbit. In a type II Weyl semimetal, magnetic breakdown between Weyl nodes in momentum space is reminiscent of Klein tunneling\([16]\). However, the observed breakdown frequency is close to the sum of the two fundamental frequencies (and not their difference, which was predicted\([15]\)). In this respect, magnetic breakdown in \(\text{Co}_3\text{S}_2\text{Sn}_2\) is reminiscent of what has been observed in \(\text{WTe}_2\)\([17]\) and in elemental metals, Mg\([18]\) and Be\([18]\) and contrasts with what was reported in the case of \(\text{ZrS}_2\text{S}_2\)[19]. The second component of the present report is a quantitative study of the thermal Hall ef-
fect. This complements previous studies of the anomalous Hall [9,11] and anomalous Nernst [9] effects in this system. Putting the anomalous Lorenz ratio (i.e. the ratio of anomalous thermal and electrical conductivities divided by temperature) under scrutiny, we found that the anomalous transverse Wiedemann-Franz law ((WF law)) holds at low temperature, but a downward deviation starts above 100K. In this regard, Co$_3$Sn$_2$S$_2$ is reminiscent of Mn$_3$Ge [20] and contrasts with Mn$_3$Sn [21], in which the anomalous WF law holds even at room temperature. As argued previously [20], the temperature at which the departure from the anomalous WF law occurs depends on the details of the Berry curvature in the vicinity of the Fermi level.

II. Experimental and computational details

High quality single crystals of Co$_3$Sn$_2$S$_2$ were grown by the self-flux method as in a previous work [22]. A stoichiometric ratio of Co, Sn and S powders was sealed in a quartz tube. The quartz tube was loaded into a box furnace and heated to 673 K and held for 24 hours. Then the tube was heated to 1273 K and held for 12 hours, then slowly cooled to 1073 K and the furnace was turned off. Shining crystals were obtained by mechanical separation carefully. The low-field transport measured by a standard four-probe method was performed on a physical property measurement system (Quantum Design PPMS-9). High-field magnetotransport measurements were carried out under pulsed magnetic field in Wuhan National High Magnetic Field Center, each contact resistance was less than 3 Ω in all the measurements.

Our calculations were performed using the density-functional theory (DFT) in the framework of the generalized gradient approximation [23] with the Vienna ab initio package [24]. To interpolate the 3D Fermi surfaces, we obtained by a tight-binding Hamiltonian, based on localized Wannier functions [25] projected from the DFT Bloch wave functions. We note that the orientations of $x$ is defined as the current direction, and the angle between $x$ and the [2110] orientation is about 10° (see more details in supplementary material [26] and Figs.1(b), z is defined as [0001] orientation.

III. Carrier density, mobility and the anomalous Hall conductivity

Fig. 1(a) shows the temperature dependence of resistivity measured as the current was applied along $x$-axis at zero field (black) and 9 T (red) along $z$-axis. A kink at Curie temperature ~175 K is visible in $\rho(T)$ at zero field. The temperature dependence of resistivity shows a metallic behavior with a residual resistivity rate of RRR $= \rho(300\text{K})/\rho(2\text{K}) \sim 30$. This is larger than what has been reported previously [9,11,27,28], indicating the high quality of the sample. Accordingly, the residual resistivity $\rho_0 = 12 \mu \Omega \text{cm}$ is also smaller than was found before [10,11]. The higher mobility leads to a larger magnetoresistance. This has been explored in other semi-metals, such as WTe$_2$ [17,29], WP$_2$ [30], PtBi$_2$ [31,32] and in Sb [33]. Fig. 1(b) displays the non-saturated positive magnetoresistance ($MR = |\rho_{xx}(\mu_0 H) - \rho_{xx}(0)|/\rho_{xx}(0) \times 100\%$) under the magnetic field of 9 T. As shown in the inset, the MR curve shows $H^{1.46}$ dependence up to 61 T at 1.6 K. Fig. 1(c) shows the nonlinear field dependence of the Hall resistivity at 2 K. The inset shows the field dependence of the ordinary Hall conductivity ($\sigma_{xy} = \rho_{yx}/(\rho_{yx}^2 + \rho_{xx}^2)$) after subtracting anomalous part [9]. The red line represents a two-band fit to the data, using the expression:

$$\sigma_{yz}^O = \frac{[\mu^2_0 \mu^2_H]}{[1 + \mu^2_0 (\mu_0 H)]} - \frac{[\mu^2_0 \mu^2_H]}{[1 + \mu^2_0 (\mu_0 H)^2]} e(\mu_0 H)$$

This allowed us to deduce the carrier density of holes (electrons): $\rho = 8.9 \times 10^{19} \text{ cm}^{-3}$ ($n = 8.7 \times 10^{19} \text{ cm}^{-3}$) and hole (electron) mobilities of $\mu_0 = 2713 \text{ cm}^2 \text{V}^{-1}\text{s}^{-1}$ ($\mu_e = 2673 \text{ cm}^2 \text{V}^{-1}\text{s}^{-1}$). The average transport mobility can also be extracted from residual resistivity at zero magnetic field $\rho_0$, using $\mu_0 = \frac{1}{\rho_0(\mu_0 + p)} = 2969 \text{ cm}^2 \text{V}^{-1}\text{s}^{-1}$.

Fig. 1(d) shows the temperature dependence of the anomalous Hall conductivity (AHC) attains 1290
\[\Omega^{-1}\text{cm}^{-1}\] at 40 K and \(1093\ \Omega^{-1}\text{cm}^{-1}\) at 2 K, the anomalous Hall angle \(AHA=\sigma_{xy}/\sigma_{xx}\times100\%\) reaches 23\% at 140 K, similar to previous reports\[10,11\]. In contrast to dirtier samples, the anomalous Hall conductivity slightly decreases below 50 K. Such a behavior was previously noticed in cleaner samples\[9\].

IV. The Fermi surface

Fig. 2 (a) and Fig. 2 (c) display the temperature dependence of magnetoresistance under high magnetic field along \(z\)-axis and \(y\)-axis. Quantum oscillations of resistivity, known as the Shubnikov-de Haas (SdH) effect, are observed when the magnetic field exceeds 20 T. A plateau in magnetoresistance appears around 20 T when the magnetic field is parallel to the \(y\)-axis. When the magnetic field is parallel to the \(xy\) plane, the saturation field for magnetization is about 23 T\[34\]. Therefore, the resistivity plateau is concomitant with the field-induced rotation of the magnetic moments from \(z\)-axis to the \(xy\) plane.

As shown in Fig. 2 (b) and (f), the SdH oscillations become clearly visible by subtracting a polynomial background from the data. Fig. 2 (c) and (g) present the fast Fourier transformation (FFT) spectrum of the oscillating component, \(\Delta \rho\) at different temperatures. Some of the broad asymmetric peaks in the FFT spectrum, such as the peak around 430 T for \(H \parallel z\) are caused by superposition of multiple peaks. We fit them, using the Gauss multi-peak method\[35\]. The evolution of these peaks at different temperatures is shown in the inset of the Fig.2g.

We can identify four peaks in the case of \(H \parallel z\) and five peaks in \(H \parallel y\), respectively. These frequencies observed in FFT spectra are closely related with the orthogonal extremal cross-sectional area \(A_F\) of the Fermi pockets by the Onsager relation \(F=(\hbar/2\pi e)A_F\). We extracted the effective mass by fitting the temperature dependence of the oscillation amplitude with the thermal damping term \(R_T=X/\sinh(x)\) of the Lifshitz-Kosevich (L-K) formula\[36\], where \(X=14.69m^*T/B\) and \(m^*\) is the effective cyclotron mass. We obtain the effective masses of different frequencies as shown in the Fig. 2(d) and (h). The parameters for pockets in two orientations are summarized in Table I. We note that the Dingle mobility is about 30 times lower than the transport mobility. A much larger difference between the two mobilities has been reported in other semi-metals, such as Sb, WP\(_2\) and Cd\(_3\)As\(_2\). In all cases, transport mobility is significantly larger than quantum mobility. In other words, the mean-free-path for momentum relaxation is longer than the one associated with the distance between collisions which broaden the Landau levels\[37\].

We studied the evolution of the SdH oscillations at 1.6 K with rotation. Fig. 3(a) presents the angle-dependence of MR which decreases gradually when the field rotates away from the \(z\)-axis to the \(y\)-axis. \(\theta\) is defined by the angle between the field and the \(z\)-axis, the MR reaches a minimum value at \(\theta=90^\circ\) when the field is parallel to the \(y\) axis. Fig. 3(b) shows SdH oscillations for each angle after subtracting a fitted polynomial. The angle dependencies of the frequencies extracted from the SdH oscillations are summarized in Fig. 3 (d). Symbols show the data and lines represent the DFT calculations.

FIG. 2. (a) and (e) the magnetoresistance as a function of the magnetic field \(\mu_0H\) at different temperatures for \(H \parallel z\)-axis and \(H \parallel y\)-axis respectively. (b) and (f) The oscillatory part of resistivity \(\Delta \rho\) for \(H \parallel z\)-axis and \(H \parallel y\)-axis respectively. (c) and (g) show the fast Fourier transformation spectrum at various temperature. The inset shows the multi-peak fitting used to distinguish between superposed frequencies. (d) and (h) the L-K fitting with the effective masses.
According to the DFT calculations, Co$_3$Sn$_2$S$_2$ has two types of electron pockets and two types of hole pockets. We labeled each pocket as shown in Fig. 4. In the case of electrons, there are twelve banana-shape pockets (E1) across the Brillouin zone, and six triangle-like pockets (E2) near the Γ point shown in the Fig. 4(a). In the case of holes, there are six cylinder-like pockets (H1) across the Brillouin zone and six triangular-like pockets (H2) close to the k$_z$ boundary shown in the Fig. 4(c).

We rotated the magnetic field in the yz plane. When the magnetic field is aligned along the z-axis ($\theta = 0$) the extreme cross-section is identical for each type of pocket and one expects to see four distinct frequencies. When the magnetic field has a finite angle with the axis, the extreme cross-section degeneracy of pockets of the same type is lifted and more than four frequencies is expected. The calculated H1 pocket can have two maximum extreme cross-section and one local minimum cross-section, since it has a quasi-cylindrical shape. Note that the calculated SdH frequencies are not symmetric between +z to y direction and –z to y direction. (see the SM for more discussion).

Fig. 4 shows the calculated Fermi surface pockets and their projections along high-symmetry z-axis. The electron pockets are in blue and the hole pockets are in red. The Fermi pockets distribution follow the $M_y$ symmetry and inversion symmetry. The total volume of the Fermi pockets yields the theoretical carrier densities. The calculated hole density is $1.82 \times 10^{20}$ cm$^{-3}$ and for the electron carrier density is $1.59 \times 10^{20}$ cm$^{-3}$ at $\mu = -2$ meV with respect to the charge neutral point.
FIG. 4. The calculated Fermi surface of Co₃Sn₂S₂. (a) The electron pocket with two types, banana-shaped pockets called E1 with blue color and triangular shape pockets called E2 with cyan color. (c) The hole pocket with two types, cylinder-shaped pockets called H1 with a red color and triangular shape pockets called H2 with a purple color. The possible cross-section with magnetic field align z direction for the electron pocket with a red line (b) and for the hole pocket with blue line (d).

As discussed above, our experimental Hall conductivity together with two-band fitting yields a carrier density, which is approximately twice lower. A similar discrepancy between experimental and theoretical carrier density was observed in the case of WTe₂ [17].

V. Magnetic breakdown

As shown in the Fig. 3(d), there is a satisfactory consistency between experimental and theoretical results. DFT calculations do not expect any frequency larger than 600 T. However, a FFT peak is present at 760 T, when the magnetic field is parallel to the y-axis. We attribute this frequency to a magnetic breakdown orbit. As shown in Fig. 3(d), this frequency is observed in the angular range of θ = 30° to 90° and it corresponds roughly to sum of E₂ and H₂ frequencies. Fig. 5(a) shows the cross sections of the E₂ and H₂ pockets when the field is parallel to y-axis.

For θ = 90°, the 760 T frequency suddenly appears above a threshold field of 41.7 T. This can be seen in Fig. 5(b), which presents the FFT spectrum for different field windows. We suggest that above 41.7 T magnetic breakdown occurs between the electron pocket E₂ and the hole pocket H₂, which according to the DFT calculations are close to each other in the momentum space. When the field is in the y axis (θ = 90°), each of these pockets has two distinct extreme orbits and therefore two distinct frequencies. The fundamental frequencies of 343 T (for E₂) and 429 (for H₂) are the larger ones and their sum is close to the observed frequency of 760 T. The angle dependence of the magnetic breakdown frequency is roughly consistent with the sum of fundamental frequencies of the E₂ and H₂ in the Fig. 3(d). The breakdown field which is defined as \( H_B \) varies with rotation between 35 T and 48 T (See the SM for more details).

Magnetic breakdown in momentum space between an electron and hole pocket resembles the Klein tunneling at a \( p-n \) junction in real space [16]. We note, however, that theory expects that this Klein tunneling leads to a breakdown frequency equal to the difference between the two original fundamental frequencies and not the sum as we observe here. Previous experiments have reported the observation of magnetic breakdown in two other topological materials, WTe₂[17] and ZrSiS[19]. In the case of WTe₂, the breakdown frequency is the sum of two fundamental frequencies. But, this breakdown may occur between the adjacent two hole pockets. In ZrSiS[19], magnetic break-
The temperature dependence of the anomalous Hall conductivity (a), the anomalous thermal Hall conductivity (b) and the anomalous Lorenz number (c). The anomalous WF law is obeyed at low temperature, but a deviation starts above 100 K.

Non-vanishing summation of the electronic Berry curvature is known to generate the intrinsic anomalous Hall effect (AHE). Measuring the thermoelectric (Nernst) and the thermal (Righi-Leduc) counterparts of the anomalous Hall effect provides additional information on the topological origin of the AHE. The amplitude of the anomalous transverse thermoelectric conductivity has been found to correlate with the amplitude of the anomalous Hall conductivity [39]. In this section we focus on the correlation between anomalous electrical and thermal Hall conductivities.

Fig. 6(a,b) show the temperature dependence of the anomalous Hall conductivity ($-\sigma_{yx}^A$) and the anomalous thermal Hall conductivity ($-\kappa_{yx}^A/T$) in another sample #29 (See the supplement for the raw data[26]), grown by chemical vapor transport method [9]. Below the Curie temperature, both increase rapidly with decreasing temperature and saturate to an almost temperature-independent magnitude below 100 K. The magnitude of $\sigma_{yx}^A$ is consistent with the previous reports [9][11]. Our $\kappa_{yx}^A/T$ data allows to check the validity of the anomalous Wiedemann-Franz Law for transverse response [20][21] by comparing the anomalous Lorenz number $L_A^A = \frac{\kappa_{yx}^A}{\sigma_{yx}^A}$ with the Sommerfeld value $L_0 = \frac{e^2}{\pi^2} (\frac{h}{k_B})^2$ where $e$ and $k_B$ are the elementary charge of electron and the Boltzmann constant, respectively.

As seen in Fig. 6(c), the Wiedemann-Franz Law holds below 100 K. This confirms once again that the anomalous Hall response is a Fermi surface (and not a Fermi sea) property as originally argued by Haldane [40]. However, above 100 K, a downward deviation in the the Wiedemann-Franz Law emerges. As argued previously [20], this deviation can be understood as the manifestation of the way the Berry curvature is averaged by thermal and electric probes. In a manner similar to Mn$_3$Ge [20], the downward deviation from the WF law in Co$_3$Sn$_2$S$_2$ arises when $\sigma_{yx}^A$ is evolving with temperature. Interestingly, according to a recent study of optical conductivity, in the ferromagnetic phase of Co$_3$Sn$_2$S$_2$, bands shift with increasing magnetization on Co sites. Such an evolution with temperature approaching the Curie temperature can generate a difference between the way Berry curvature is integrated in the electrical and the thermal channels [38].

VII. Summary

In summary, we performed a study of quantum oscillations of magnetoresistance in high-quality single crystals of Co$_3$Sn$_2$S$_2$ up to 61 T. We found a good agreement between the frequency of these oscillations and the Fermi surface of Co$_3$Sn$_2$S$_2$ according to DFT calculations. The Fermi surface consists of two types of electron pockets and two types of hole pockets. We detected an additional frequency and attributed it to magnetic breakdown between an electron and a hole pocket. We also found that the anomalous transverse Wiedemann-Franz law holds at low temperature, but a downward deviation emerges at finite temperature.

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FIG. S1. The in-plane orientation of the sample was determined by a single crystal XRD.

FIG. S2. (a) The temperature dependence of in-plane resistance and (b) thermal conductivity of the sample #29. The field dependence of (c) the Hall conductivity and (d) thermal Hall conductivity at various temperatures.

Supplemental Materials for "Bulk Fermi surface of the ferromagnetic Kagome-lattice semimetal Co$_3$Sn$_2$S$_2""

S1. determining the in-plane orientation of the sample

High quality single crystals of Co$_3$Sn$_2$S$_2$ were grown by the self-flux method, the nature cleavage plane is the ab-plane of the single crystals. The c-axis can be identified easily, while the orientations of the single crystal in ab-plane were confirmed by single-crystal XRD technique. The angle between a-axis and the x is about 10°, as the figure S1 shows.

By measuring the field dependence of resistivity Fig. S2(a) and Hall resistivity Fig. S2(b), we can deduce the Hall conductivity through $\sigma_{yx} = -\frac{\rho_{yx}}{\rho_{xx}^2 + \rho_{yx}^2}$ as the field is along c or the z axis. After plotted the Hall conductivity in the Fig. S2(c), the anomalous Hall conductivity can be easily obtained by extrapolate the normal part of the Hall effect to zero field. The intersect at the the axis of the $\mu_0H=0$ in the Fig. S2(d) is the anomalous Hall conductivity. On the other hand, the thermal Hall conductivity can be obtained by measuring transverse thermal gradient with the measured longitudinal thermal conductivity.
FIG. S4. Angular dependence of the frequencies extracted from the SdH oscillations, the lines are from the theory calculation results for $z$ to $y$ direction (a) and $-z$ to $y$ direction(b).

\[ \kappa_{yx} = \frac{\partial_y T \kappa_{yy} \kappa_{xz}}{J_{qz}} \] \[ \text{[1]} \]

The breakdown fields for different angles were determined by carrying out FFT in different field ranges in the Fig. S3. The black lines are for a field range from 5 T to 61 T. The blue lines are for the field range where no breakdown peak presents, while the red lines for the field range where breakdown peak presents. The table S1 shows the summary of the results from experiments. The absence of the E2 in the table S1 is due to the small amplitude of the peak in the FFT. Below 30°, the breakdown frequency is not observable.

S4. The angle-dependence of FFT lines in theoretical calculation

As mentioned in the main text, the calculated SdH frequencies are not symmetric between $+z$ to $y$ direction (a) and $-z$ to $y$ direction(b), because of the low symmetry in the $yz$ plane. From our experimental data, we can’t distinguish the plane the sample was rotated from $+z$ to $y$ direction or $-z$ to $y$ direction. By the comprising with the experimental data in the Fig.S4(a), the Fig. 3d shows better overlap between theoretical and experimental results.

TABLE S1. The extracted frequency of the E2, H2 and the magnetic breakdowns at different angles, $H_B$ is where the magnetic breakdown field occurs.

| $\theta$ | E2 (T) | H2 (T) | MB (T) | $H_B$ (T) |
|----------|--------|--------|--------|-----------|
| 90°      | 343    | 429    | 760    | 41.7      |
| 85°      | 323    | 416    | 789    | 43.4      |
| 75°      | 403    | 751    | 47.6   |           |
| 65°      | 376    | 702    | 35.1   |           |
| 60°      | 305    | 473    | 854    | 37.7      |
| 55°      | 299    | 462    | 810    | 38.2      |
| 40°      | 293    | 468    | 718    | 38.4      |
| 30°      | 468    | 724    | 41.7   |           |

As mentioned in the main text, the calculated SdH frequencies are not symmetric between $+z$ to $y$ direction (a) and $-z$ to $y$ direction(b), because of the low symmetry in the $yz$ plane. From our experimental data, we can’t distinguish the plane the sample was rotated from $+z$ to $y$ direction or $-z$ to $y$ direction. By the comprising with the experimental data in the Fig.S4(a), the Fig. 3d shows better overlap between theoretical and experimental results.

[1] Li X, Xu L, Ding L, Wang J, Shen M, Lu X, Zhu Z and Behnia K 2017 Phys. Rev. Lett. 119 056601