Giant anomalous Nernst effect in the magnetic Weyl semimetal Co$_3$Sn$_2$S$_2$

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In ferromagnetic solids, even in absence of magnetic field, a transverse voltage can be generated by a longitudinal temperature gradient. This thermoelectric counterpart of the Anomalous Hall effect (AHE) is dubbed the Anomalous Nernst effect (ANE). Expected to scale with spontaneous magnetization, both these effects arise because of the Berry curvature at the Fermi energy. Here, we report the observation of a giant ANE in a newly-discovered magnetic Weyl semimetal Co$_3$Sn$_2$S$_2$ crystal. Hall resistivity and Nernst signal both show sharp jumps at a threshold field and exhibit a clear hysteresis loop below the ferromagnetic transition temperature. The ANE signal peaks a maximum value of $\sim 5 \, \mu V/K$ which is comparable to the largest seen in any magnetic material. Moreover, the anomalous transverse thermoelectric conductivity $\alpha_{yx}$ becomes as large as $\sim 10 \, A/K.m$ at 70 K, the largest in known semimetals. The observed ANE signal is much larger than what is expected according to the magnetization.

I. INTRODUCTION

The Nernst effect, the transverse electric field generated by a longitudinal thermal gradient in the presence of a magnetic field, has triggered renewed attention in condensed matter physics since the discovery of the pseudogap phase in cuprates$^{[1, 2]}$. Conductors with a large Nernst coefficient are important for device applications as in cryogenic refrigerations$^{[3]}$, but such devices have not been realized in practice because of their low conversion efficiency. Recent years, the large Nernst effect has been observed in correlated electron systems$^{[4–6]}$, conventional semimetals$^{[7]}$, as well as in metallic ferromagnets$^{[8–10]}$. Consequently, a number of novel ground states$^{[11]}$ and exotic electronic orders$^{[12]}$ can be identified by measuring the Nernst effect. For some ferromagnetic metals, in particular, the Nernst signal was observed below $T_c$ even in the absence of external magnetic field$^{[8–10]}$. This phenomenon, known as the anomalous Nernst effect (ANE), is observed to be proportional to the magnetization. The underlying physics is that the spontaneous magnetization in these materials plays a role of the intrinsic magnetic field, geometrically connected to the Berry curvature of the Bloch bands at the Fermi energy$^{[13]}$.

In recent years topological Dirac/Weyl semimetal materials have been theoretically predicted and experimentally discovered. The electronic structures of these materials have the topologically robust$^{[14–18]}$ and symmetry-protected bulk energy bands which linearly intersect at some special points (the Dirac points) or symmetry axis near the Fermi level$^{[19, 20]}$. The breaking of inversion symmetry or time-reversal symmetry (TRS) can split a Dirac point into a pair of the Weyl points with opposite chiralities. The chiral Weyl points are then the source or sink of the Berry curvature $\Omega(k)$$^{[21]}$, meaning that the Berry curvature is singular at these points. So far, dozens of Dirac/Weyl semimetals have been investigated$^{[22–29]}$ and several unique physical phenomena such as the large magneto-resistivity (MR)$^{[29]}$ and ultrahigh mobility$^{[22]}$ have been observed. However, the defining properties of such topological semimetals, including the Fermi arc in surface states and chiral anomaly in charge transports, are not easy to be identified experimentally. So far, a direct evidence of the Fermi arc has been clarified by ARPES measurements in the TaAs-family$^{[30, 31]}$. While, a possible indirect signature of the chiral anomaly is associated with the negative longitudinal MR as investigated in several relevant materials$^{[20, 32, 33]}$. But some extrinsic factors such as the current jetting and crystal inhomogeneity$^{[34]}$ are hardly ruled out.

Quite interestingly, there are two kinds of transverse transport properties, the anomalous Hall effect (AHE) and the anomalous Nernst effect (ANE), can help to probe the topological nature of charge carriers in the ferromagnetic Weyl semimetals. This is because both the transverse transport properties are contributed from the intrinsic magnetic field in the occupied bands and thus deemed as strong proofs of the finite Berry curvature originating from the separation of Weyl nodes. Recently, a magnetic Weyl semimetal Co$_3$Sn$_2$S$_2$ with a ferromagnetic kagome-lattice has been reported to show an intrinsic anomalous Hall effect$^{[35–37]}$, but no experiment on the Nernst effect in this material has been reported yet. It should be noticed that unlike the Hall effect where the normal contribution in a metal is always finite, the Nernst effect generally vanishes in ordinary metals due to the, and thus the anomalous contribution may become very prominent. On the other hand, a large AHE is not necessary to cause a large ANE. This is because the AHE...
The sample shows a magnetic moment of stacked along the c-axis with a corner-sharing octahedra. CoSn-layer, and the kagome lattices in different layers are lattice with corner-sharing triangles of Co atoms in the with the space group of R-3m. Co atoms form a kagome correlation between AHE and ANE.

Correlation between AHE and ANE is highly useful to confirm the contribution of the Berry curvature and in turn verify the ANE signal reaches a maximum value of ∼ 10 A/K.m, much larger than those of known ferromagnetic metals. This result shows that CoSn2 crystallizes in a rhombohedral structure at high temperature but starts to decrease below 200 K. The in-plane magnetic field dependence as the magnetization curve broadens and becomes very smooth under 9 T, as seen in the literatures[35, 36]. Temperature dependence of longitudinal resistivity ρxx and thermopower Sxx of CoSn2 single crystal are shown in figure 1a. Overall the resistivity shows a metallic behavior in the whole temperature regime but exhibits an unexpected kink associated with a ferromagnetic transition(FM) at about Te = 175 K, similar to the previous report[38]. As a magnetic field up to 9 T is applied, the resistivity does not change too much at high temperature but starts to decrease below 200 K. The kink in ρxy(T) near Te broadens and becomes very smooth under 9 T, as seen in the literatures[35, 36].

Figure 2a and 2c display the Hall resistivity and conductivity vs. the applied magnetic fields B || c \perp I at a representative temperature of 70 K. The ρxy(B) significantly exhibits a steep rectangular hysteretic jump at low fields, and then becomes almost flat at a higher field. Its magnitude is comparable to what was reported previously [35, 36] and is reversible with a field of 0.15 T at 70 K. The largest coercive field is about 0.7 T at 5 K (See figure S3 in SI). The Nernst signal Syx (figure 2b) shows a similar jump of about 10 µ V/K, which is over one order of magnitude larger than those of typical materials for the ANE such as Mn3Sn[39], pure metal Fe[40] CuCr2Se4−xBrx[9] and single crystal Fe3O4[10]. The large Sxx is also verified by the large Nernst angle θN = |Sxy/Sxx| of 16% (Figure 3b). Note that both Hall resistivity and Nernst signal show nearly the same field dependence as the magnetization curve (See figure S2 in SI), implying that the contribution of

**FIG. 1.** a Temperature dependence of the longitudinal electric resistivity(ρxx) of CoSn2 sample under zero field and 9 T. b Temperature dependence of magnetization with the FC modes at B = 0.1 T. The inset shows the saturated magnetization vs. fields at 5 K.

**FIG. 2.** Field dependence of transport coefficients at 70 K as B \parallel c-axis \perp I. (a) Hall resistivity ρxy vs. magnetic fields. (b) The Nernst signal Syx, extracted from ρyx and ρxy. (c) The Hall conductivity σyx, extracted from ρxx, ρyx, Sxx and Syx. The hysteric behavior is clearly observed at 70 K.

II. RESULTS AND DISCUSSION

CoSn2 crystallizes in a rhombohedral structure with the space group of R-3m. Co atoms form a kagome lattice with corner-sharing triangles of Co atoms in the CoSn-layer, and the kagome lattices in different layers are stacked along the c-axis with a corner-sharing octahedra. The sample shows a magnetic moment of ∼ 0.33µB/Co along the c-axis with a Neel temperature of about 175 K in figure 1b. The FC and ZFC curves overlap each other very well at 0.1 T, implying that the magnetic domains have been arranged along c-axis by the small external fields. The clear hysteresis loop with the saturated magnetization M of 0.33 µB/Co at 5 K is observed in the inset of figure 1b, consistent with the theoretical prediction[33, 36].
the anomalous Hall and Nernst effects dominates and the normal contribution (proportional to B) is negligibly small at low fields.

Field dependence of the Hall conductivity \( -\sigma_{yx} \) and the transverse thermoelectric conductivity \( \alpha_{yx} \) at 70 K is shown in figure 2c and 2d. The \( \sigma_{yx} \) and \( \alpha_{yx} \) can be written as the formulas \( \sigma_{yx} = -\rho_{yx}/(\rho_{yx} + \rho^2) \) and \( \alpha_{yx} = (\rho_{xx}^2 - \rho_{yx}^2)/(\rho_{xx}^2 + \rho_{yx}^2) \). The obtained \( |\sigma_{yx}| \) at the saturated field 0.5 T reaches \( \sim 1320 \, \Omega cm^{-1} \), which is very close to the calculated result (1310 (\Omega cm)^{-1}) from the integral of Berry curvature along \( k_z \) in the system [35][36]. Similar to the large \( \sigma_{yx} \), the \( \alpha_{yx} \) is found to be very large, reaching approximately 11 A/K.m at 70 K. This value is one or two orders magnitude of larger than the other typical ferromagnets or Mn$_3$Sn with \( \alpha_{yx} \sim 0.01 - 1 \, A/K.m \), as shown in figure 4b.

Temperature dependence of the AHR, \( \rho_{yx}^{A} \), (extrapolating the high-field part of \( \rho_{yx} \) back to zero field) and AHC, \( \sigma_{yx}^{A} \), are illustrated in figure 3a. The \( \rho_{yx}^{A} \) shows a strong temperature-dependence below \( T_c \), and peaks a maximum value of 21 \( \mu \Omega cm \) around 140 K. While the \( \sigma_{yx}^{A} \) is almost temperature-independence below 100 K. Such feature is also observed in pure materials such as Fe, Co, Ni and Gd, where the mechanism of the AHE has been suggested to be the intrinsic Berry-phase mechanism [8]. We plot the \( \sigma_{xy}^{A} \) as a function of \( \sigma_{xx} \) in the main plane of figure 3b. It describes that \( \sigma_{xy}^{A} \) is weakly dependent of \( \sigma_{xx} \) at low temperature regime( < 100 K), similar to the case in Fe, Ni pure metals [8] (See SI). A large anomalous Hall angle \( |\sigma_{yx}/\sigma| \) reaches a maximum value about 15% at 150 K and gradually decreases to 6.3% at 5 K, as shown in the inset of figure 3b. The large Hall angle in Co$_3$Sn$_2$S$_2$ is close to the value of 16% in GdPtBi, a typical field-induced Weyl-semimetal [11], but is much larger than that of the nonlinear antiferromagnetic Mn$_3$Sn (3.2%) [42] and Mn$_3$Ge (5%) [59].

The ANE signal \( S_{yx}^{A} \) as a function of temperature is displayed in figure 3c. The \( S_{yx}^{A} \) strongly depends on temperature, rapidly increases below \( T_c \) and then peaks at around 70 K, a half of the peak temperature of \( \rho_{yx}^{A} \) in figure 3a. Similar to the large Hall angle, the large \( S_{yx}^{A} \) can be verified by the Nernst angle \( \theta_N = |S_{xy}/S_{xx}| \) in the inset of figure 3c. The value reaches 17% at 70 K, which is comparable with the previous findings in the Co$_2$MnGa system [43], a FM semiconductor. Correspondingly, temperature dependence of the \( \alpha_{yx}^{A} \) is mapped in the inset of figure 3d, and the \( \alpha_{yx}^{A} \) rapidly increases below 200 K, showing a maximum value of \( \sim 11 \, A/K.m \) at 50 K, followed by a monotonous decrease to low temperatures. Such large \( \alpha_{yx}^{A} \) is one or two orders magnitude larger than the other ferromagnets with a small \( \alpha_{yx} \sim 0.01-1 \, A/K.m \) (See figure S4 in SI). Note that the \( \alpha_{yx}^{A} \) is roughly obey the linear T-dependence at low temperatures, but closely follow the relation \( \propto T \log(T) \) at a temperature regime from 70 K to 175 K, as shown in figure 3d. Similar behavior has been reported in Co$_2$MnGa system [43].

The significant difference between the giant ANE in Co$_3$Sn$_2$S$_2$, Mn$_3$Sn and in ferromagnets is demonstrated in figure 4a. We plot the ANE as functions of the magnetization with a logarithmic scale for various ferromagnetic metals, Mn$_3$Sn and Co$_3$Sn$_2$S$_2$ (See Methods). The ANE in ferromagnets is known to be roughly proportional to the magnetization \( M \), \( |S_{yx}| = |Q_s|\rho_0 M \), where \( |Q_s| \) is the anomalous Nernst coefficient. It can be seen that almost all of ferromagnetic metals follow this re-
ulation and their AHE signal becomes more larger with increasing magnetization. The shaded region covers all of the data points for ferromagnets. An exception for the antiferromagnetic Mn$_3$Sn observed strongly violates this scaling, which is ascribed to the enhanced Berry curvature at the Fermi energy [42]. Similarly, the Co$_3$Sn$_2$S$_2$ does not follow this relation. The $S_{yx}$ is concretely far away from this trend and more than one and two order magnitude larger than what would be expected based on the scaling relation. In addition, the intrinsic transverse thermoelectric conductivity $|\alpha_{yx}|$ is close to 11 A/K.m for Co$_3$Sn$_2$S$_2$, the largest value compared to the other ferromagnets and semimetals as shown in figure 4b. The observed large $|\alpha_{yx}|$ in the Weyl magnets are potentially useful to realize the thermoelectric devices for cryogenic refrigeration.

The mechanism of the significantly large intrinsic ANE in Co$_3$Sn$_2$S$_2$ should be distinct from the conventional one for ferromagnets. In the forwork of the Boltzmann approach, the anomalous Hall conductivity $\sigma_{yx}^A$ and the anomalous transverse thermoelectric conductivity $\alpha_{yx}^A$ can be written as the formula [27]:

$$\sigma_{yx}^A = -\frac{e^2}{h} \int \frac{dk}{2\pi^2} \Omega_{n,z}(k) f_{nk},$$

and

$$\alpha_{yx}^A = -\frac{eT}{\Omega_{n,k}} \int \frac{dk}{2\pi^2} \Omega_{n,z}(k)(\varepsilon_{n,k} - \mu) f_{nk} + k_B T \ln [1 + e^{-\beta (\varepsilon_{n,k} - \mu)}].$$

Here, $\Omega_{n,k}$ is the Berry curvature along the z direction, and $f_{nk} = f(E_{nk})$ is the Fermi distribution function with the band index $n$ and the wave vector $k$. $\varepsilon_{n,k}$ and $\beta$ donate the band energy and 1/kBT. From the equations, the $\alpha_{yx}^A$ is governed by the Berry curvature around the Fermi energy, while the $\sigma_{yx}^A$ is given by the summation of the Berry curvature for all the occupied bands. According to these equations, the $\alpha_{yx}^A$ at low temperatures can be reduced to the Mott relations $\alpha_{yx}^A = \frac{\pi^2 k_B^2 T}{3e} \frac{\partial \sigma_{yx}^A}{\partial \mu}$. Our result in $\alpha_{yx}^A(T)$ showing a roughly linear temperature-dependence at low temperatures is consistent with the Mott rule.

### III. CONCLUSION

In summary, we report an observation of the giant ANE and the large intrinsic transverse thermoelectric conductivity in the magnetic Weyl semimetal Co$_3$Sn$_2$S$_2$. The Nernst signal $S_{yx}$ reaches $\sim$ 5 $\mu$ V/K at 70 K, much larger than those of known ferromagnetic metals. Furthermore, the $\alpha_{yx}^A \sim 10^4$ A/K.m in the sample is over one or two order magnitude larger than those of other ferromagnets where $\alpha_{yx}^A$ = 0.01 – 1 A/K.m. Our experimental results indicate that the giant ANE in Co$_3$Sn$_2$S$_2$ can be ascribed to the enhanced contribution from the Berry curvature very close to Fermi levels. The enhanced $\alpha_{yx}^A$ in Weyl magnets paves a path to realize potentially thermopile devices for the thermoelectric power generation. Our experimental results also highlight the complementary roles of ANE and AHE in revealing the intrinsic Weyl state in the magnetic Weyl semimetals.

### IV. METHOD

Large single crystals of Co$_3$Sn$_2$S$_2$ with the mininiter size were grown through the Bridgman technique, as reported in the previous reports [38].

The single crystal X-ray diffraction were obtained using a D/Mx-max3a diffractometer with CuK$_\alpha$ radiation and a graphite monochromator at the room temperature, which determines the crystal grown orientation. The composition of the crystals were obtained by energy dispersive X-ray (EDX) spectroscopy. The sample is polished and then cut into a bar-shape with a size of 3.5$\times$2.0 mm$^3$. The magnetization measurements were done using a commercial SQUID magnetometer. The (magnetoresistivity and Hall coefficient measurements were performed using the standard four-terminal method in a commercial Quantum Design PPMS-9 system. The thermal power measurement were performed with a one-heater-two-thermometers technique in PPMS with a high-vacuum environment. Two Chromel-constantan (type E) thermocouples were employed to measure the temperature difference generated by a small heater chip.

In figure 4a, the Nernst signal $S_{yx}$ for various ferromagnets below the Curie temperatures is abstracted the ANE results, as reported in those literatures. For example Fe(300 K) [44]; Co(300K) [44], Fe$_3$O$_4$(300K) [10], MnGe(140K) [43], MnGe(100K) [43] and MnGe(20K) [43], Nd$_2$MoO$_7$[46]; Co/Ni film(300K) [47]; Pt/Fe multilayer(300K) [48]; MnGa(300K) [11], $\alpha_{xy}^A \sim 0.2$ and 0.12 A/Km for La$_{1-x}$Sr$_{x}$CoO$_3$(x=0.3 and 0.25) [8], $\alpha_{xy}^A \sim 0.3$ A/Km for Nd$_2$MoO$_7$[46], $\alpha_{xy}^A \sim 0.25$, and $\alpha_{xy}^A \sim 0.0045$ A/Km for Fe$_3$O$_4$[10], and $\alpha_{xy}^A \sim 0.9$, 0.52 and 0.21 A/Km for FePt, Co and Fe$_3$O$_4$. $\alpha_{xy}^A \sim 0.9$, 0.52 and 0.21 A/Km for FePt, Co and Fe$_3$O$_4$.

In figure 4b, the maximum $|\alpha_{yx}^A|$ values are taken from each magnetic material, Mn$_3$Sn and Co$_2$MnGa.

$\alpha_{xy}^A \sim 1$ A/Km for Mn$_3$Sn [49], $\alpha_{xy}^A \sim 0.6$ A/Km for SrRuO$_3$[8], $\alpha_{xy}^A \sim 0.3$ A/Km for Mn$_3$Sn [42], $\alpha_{xy}^A \sim 0.2$ and 0.12 A/Km for La$_{1-x}$Sr$_x$CoO$_3$(x=0.3 and 0.25) [8], $\alpha_{xy}^A \sim 0.03$ A/Km for Nd$_2$MoO$_7$[46], $\alpha_{xy}^A \sim 0.25$, and $\alpha_{xy}^A \sim 0.0045$ A/Km for Fe$_3$O$_4$[10], and $\alpha_{xy}^A \sim 0.9$, 0.52 and 0.21 A/Km for FePt, Co and Fe$_3$O$_4$. $\alpha_{xy}^A \sim 0.9$, 0.52 and 0.21 A/Km for FePt, Co and Fe$_3$O$_4$.

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ACKNOWLEDGEMENTS

We thank K. Beluia for inspiring discussions and suggestions. We also thank H. Wang, and J. Yang for some technical support. This research was supported in part by the NSF of China (under Grants No. 11474082) and the National Key Projects for Research and Development of China (Contract No. 2016YFA0000402). Yu-Ke Li was supported by an open program from Wuhan National High Magnetic Field Center (2016KF03).

AUTHOR CONTRIBUTIONS

Y. Li designed the research. H. Yang synthesized the samples and performed the electronic and thermal transport measurements. W. You performed the XRD measurements and analyzed the structure parameters. C. Cao performed the first-principle calculation. C. Xi, and J. Wang assisted the measurements. C. Cao, M. L. Tian, Z. A. Xu, J. Dai, and Y. Li, discussed the data, interpreted the results. J. Dai and Y. Li wrote the paper.

ADDITIONAL INFORMATION

Competing financial interests: The authors declare no competing financial interests. Correspondence and requests for materials should be addressed to Yuke Li (email: yklee@hznu.edu.cn).