

# **Bipolarity of large anomalous Nernst effect in Weyl magnet-based alloy films**

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## Abstract

A thermopile device converts thermal energy to electrical energy. Controlling the polarity of the thermoelectric voltage that such a device generates is an important part of enhancing its thermoelectric output. Constructing thermopile devices where the mechanism is based on the anomalous Nernst effect in topological magnets and where one can control the bipolarity is still hard to do. Here we demonstrate the bipolarity of a large anomalous Nernst effect in a series of Weyl ferromagnet  $\text{Co}_3\text{Sn}_2\text{S}_2$ -based alloy films by tuning the Fermi energy. We illustrate the bipolarity of the anomalous Nernst signal originating from the intrinsic Berry curvature contribution by systematically regulating the Fermi energy by Nickel or Indium substitution while maintaining a topological band feature of the Weyl ferromagnet. The bipolarity enables the construction of the Weyl magnet-based anomalous Nernst thermopile that generates large thermoelectric output at zero magnetic field. These demonstrations of bipolar large anomalous Nernst effect in  $\text{Co}_3\text{Sn}_2\text{S}_2$ -based films will stimulate the device development of efficient thermoelectric energy conversion exploiting topological magnets.

Two types of thermoelectric energy conversions in a homogeneous material have been investigated as the voltage generation along longitudinal direction, Seebeck effect (**Fig. 1a**), and transverse direction driven by magnetization, anomalous Nernst effect (ANE, **Fig. 1b**), to

the direction of thermal flow<sup>1-5</sup>. In conventional semiconductor, the polarity of Seebeck effect is governed by the dominant carrier-type whether electron or hole, which can be controlled via Fermi energy  $E_F$  tuning by chemical doping (**Fig. 1c**) (ref. <sup>1</sup>). The polarity is defined as positive or negative slope of voltage generation against temperature variation as shown in **Fig. 1d**, corresponding to the positive Seebeck coefficient  $S_{xx}$  for  $p$ -type with hole or negative one for  $n$ -type with electron in the single band model. This response to the thermal flow can be widely applied to judge the carrier-type of the dominant conducting charge carrier in a single-band material as well as Hall effect measurement, because  $S_{xx}$  directly reflects the energy derivative of density of state at the  $E_F$ . By combining these semiconducting materials possessing different polarities, the thermoelectric output voltage is accumulated by the implementation as a thermopile device<sup>1</sup>.

Recently, ANE in topological magnets has been intensively studied because of a giant response originating from Berry curvature in the topological electronic bands<sup>2-4</sup>. For one example, the simple electronic structure around the Weyl point is schematically depicted in **Fig. 1e**. A gapped nodal line with a singularity point close to the  $E_F$  provides a crucial contribution of intrinsic mechanism via Berry curvature exhibiting giant anomalous Hall effect (AHE) and ANE<sup>4,6-8</sup>. According to the theoretical band calculation, the polarities of AHE and ANE in the topological magnets are dominated by the sign of exchange interaction between the linearly-dispersive band and magnetic moments. Experimentally, as depicted in **Fig. 1f**, the polarity of

ANE can be defined as positive (yellow) and negative (blue) of the hysteresis loop, yielding the positive/negative Nernst coefficient  $S_{xy}$  under the same magnetization  $M$  direction at zero-magnetic-field. These positive and negative  $S_{xy}$  of two magnets under parallel magnetization  $M$  are compatible to the combination of  $p$ -type and  $n$ -type in semiconductors, enabling us to fabricate the thermopile based on ANE<sup>9,10</sup>. Although the polarities of ANE for the topological magnets are expectedly controllable by shifting the  $E_F$  across the exchange gap (**Fig. 1e**), it is still difficult to control the bipolarity of ANE in a same base material due to the dominant contribution from large Fermi surface of magnetic metals and parasitic contribution of multiple gapped nodal lines and Fermi surfaces in semimetals.

Topological materials have been successively predicted by first-principles theoretical band calculation incorporating spin-orbit interaction, providing a challenging playground for materialization of a giant response of AHE and ANE<sup>4,6-8</sup>. In AHE in two-dimensional (2D) surface state of a magnetic topological insulator (TI), for example Cr-doped  $(\text{Bi,Sb})_2\text{Te}_3$ , the chemical substitution of Bi/Sb works well to adjust  $E_F$  to the exchange gap at singularity point, making the system to quantum anomalous Hall insulator state<sup>11-13</sup>. As for the polarity of AHE, the hysteresis direction of Cr- (ref.<sup>11,12</sup>) and V-doped  $(\text{Bi,Sb})_2\text{Te}_3$  (ref.<sup>14</sup>) is positive as opposed to negative for that of Mn-doped  $\text{Mn}_x\text{Bi}_{2-x}\text{Te}_{3-y}\text{Se}_y$  (ref.<sup>15</sup>) owing to the different sign of magnetic interaction. Regarding ANE in the 2D surface state of TI, the polarity reversal has been reported by electrostatic gating across the exchange gap<sup>16</sup>. In contrast to the controllable

manner for the 2D surface state, the control of ANE in the three-dimensional (3D) Weyl magnets is rather difficult. In fact, although many candidate compounds exhibiting large ANE,  $\text{Co}_2\text{MnGa}$  (ref. <sup>17</sup>),  $\text{UCo}_{0.8}\text{Ru}_{0.2}\text{Al}$  (ref. <sup>18</sup>),  $\text{Fe}_3X$  ( $X = \text{Al, Ga, Sn}$ ) (refs. <sup>19-22</sup>),  $\text{YbMnBi}_2$  (ref. <sup>23</sup>), and  $\text{Co}_3\text{Sn}_2\text{S}_2$  (refs. <sup>24,25</sup>), are categorized as topological magnets, it is still difficult to make a theoretical strategy to maximize the  $S_{xy}$  and/or to control the polarity of ANE because the singularity point often appears in multiple at the specific energy position far from  $E_F$ . In the metallic systems with alloying, the  $E_F$  tuning with rigidly keeping topological band feature is not trivial due to large Fermi surface. Since another requirement on magnetic properties is the out-of-plane  $M$  to yield ANE in case of the in-plane thermal gradient, the large coercivity enables the stable zero-field operation of magneto-thermoelectric energy conversion. Here, one of candidates of magnetic Weyl semimetal,  $\text{Co}_3\text{Sn}_2\text{S}_2$ , is a good platform for examining the polarity control of ANE via  $E_F$  tuning owing to small Fermi surface with relatively simple band structure as well as large ANE with the large coercivity<sup>26-29</sup>. In addition, the systematic control of the bipolarity of large ANE in  $\text{Co}_3\text{Sn}_2\text{S}_2$ -based alloy films enables us to examine the zero-field operation of the anomalous Nernst thermopile composed of the same base Weyl magnet.

### Considerations from band calculation

The Weyl points in  $\text{Co}_3\text{Sn}_2\text{S}_2$  are located around L points schematically shown in **Fig. 2a** (see also Supplementary Fig. 1, refs. <sup>28-30</sup>). Considering the Stoner ferromagnetism of  $\text{Co}_3\text{Sn}_2\text{S}_2$

(refs. <sup>31,32</sup>), the density of states at  $E_F$  is rather small at the ferromagnetic condition, resulting in the dominant contribution of Berry curvature to the large AHE and ANE thanks to the existence of gapped nodal lines with Weyl points. Based on this band structure, the anomalous Hall conductivity  $\sigma_{\text{AHE}}$  and anomalous Nernst conductivity  $\alpha_{\text{xy}}^{\text{A}}$  can be theoretically calculated as shown in **Fig. 2b** and **2c**, respectively<sup>4,6-8,30</sup>. In **Fig. 2b**, the positive maximum value of  $\sigma_{\text{AHE}}$  as large as  $1200 \text{ Scm}^{-1}$  appears at close to the  $E_F$  similar to AHE in the 2D surface state of magnetic TI (ref. <sup>11,12</sup>), which is an ideal situation to examine the control of ANE<sup>4</sup>. According to the Mott relation at low temperature  $\alpha_{\text{xy}}^{\text{A}} = \frac{\pi^2 k_{\text{B}}^2 T}{3e} \sigma'_{\text{AHE}}(E_F)$ , where  $k_{\text{B}}$  is Boltzmann constant,  $T$  temperature,  $e$  elementary charge, and  $\sigma'_{\text{AHE}}(E_F)$  energy derivative of  $\sigma_{\text{AHE}}$  at  $E_F$  (refs. <sup>4,6-8</sup>), the sign of  $\alpha_{\text{xy}}^{\text{A}}$  is theoretically predicted to be reversed across the  $E_F$  as shown in **Fig. 2c**. The positive and negative  $\alpha_{\text{xy}}^{\text{A}}$  values appear at the energy upper and lower than  $E_F$ , respectively. In contrast to the clear guiding principle, the experimental materialization is challenging because of difficulty of  $E_F$  tuning with rigidly keeping band feature and complexity of the  $S_{\text{xy}}$ . The anomalous component of  $S_{\text{xy}}$  at  $B = 0 \text{ T}$  ( $S_{\text{ANE}}$ ) is composed of two terms as a following equation,  $S_{\text{ANE}} = \rho_{\text{xx}} \alpha_{\text{xy}}^{\text{A}} - \rho_{\text{AHE}} \alpha_{\text{xx}}$ , which can be converted to  $\alpha_{\text{xy}}^{\text{A}} = \sigma_{\text{xx}} S_{\text{ANE}} + \sigma_{\text{AHE}} S_{\text{xx}}$ , where  $\rho_{\text{xx}}$  is resistivity,  $\rho_{\text{AHE}}$  anomalous Hall resistivity,  $\alpha_{\text{xx}}$  Peltier conductivity, and  $\sigma_{\text{xx}}$  longitudinal electrical conductivity<sup>3,4,7,8</sup>. In this study, we report on the demonstration of bipolar large ANE via systematic control of  $E_F$  in  $\text{Co}_3\text{Sn}_2\text{S}_2$ -based alloy thin films, where  $E_F$  could be tuned by substitution of Co with Ni or Sn with In. By applying these  $\text{Co}_3\text{Sn}_2\text{S}_2$ -based Weyl magnets with

positive and negative  $S_{xy}$  to fabricate the thermopile, the magneto-thermoelectric conversion at zero-magnetic-field was demonstrated by accumulating the ANE voltage.

### **Sample fabrication and measurement setup**

$\text{Co}_3\text{Sn}_2\text{S}_2$  and  $\text{Co}_3\text{Sn}_2\text{S}_2$ -based alloy thin films of  $\text{Co}_3\text{In}_y\text{Sn}_{2-y}\text{S}_2$  ( $y = 0.08, 0.24, 0.41,$  and  $0.60$ ) and  $\text{Co}_{3-x}\text{Ni}_x\text{Sn}_2\text{S}_2$  ( $x = 0.11, 0.21,$  and  $0.44$ ) were fabricated by radio-frequency magnetron sputtering on  $\text{SrTiO}_3$  (111) substrates<sup>33</sup>. Detail fabrication process and composition analysis are described in the Methods section. Crystal structure was characterized by x-ray diffraction with  $\text{Cu } K\alpha$ , whose results are summarized in supplementary information Supplementary Fig. 2a. The systematic variation of lattice parameters indicates the substitution of Ni and In into Co and Sn, respectively. Additionally, the substitution has been examined by variation of magnetic properties, magnetization  $M$  and ferromagnetic transition temperature  $T_C$  in the alloy films<sup>34</sup>. The variation of these parameters is consistent to those of bulk studies on  $\text{Co}_3\text{In}_y\text{Sn}_{2-y}\text{S}_2$  (refs. <sup>35,36</sup>) and  $\text{Co}_{3-x}\text{Ni}_x\text{Sn}_2\text{S}_2$  (refs. <sup>37,38</sup>). The thickness of films is almost 40 nm, which is thick enough to evaluate AHE and ANE (ref. <sup>39</sup>). We applied  $\text{SrTiO}_3$  substrate in this study because thermal conductance of  $\text{SrTiO}_3$  is sufficiently low to apply reliably-measurable temperature gradient in the film down to 50 K (ref. <sup>40</sup>). The electrical and thermoelectric transport measurements were carried out with a home-made sample stage to apply temperature gradient in physical properties measurement system (PPMS, Quantum Design, Inc.). A picture

of the measurement setup for thermoelectric effect is displayed in **Fig. 2d** (See also Supplementary Fig. 3). The local temperatures were measured by Pt thermometers on the substrate. Temperature and magnetic field dependences of resistivity and Hall effect were simultaneously measured at the same measurement sweeps. In addition, with applying temperature gradient by heating one side (Supplementary Fig. 3d), the thermoelectric effects were characterized with applying magnetic field at various temperatures.

### **Bipolarity of $S_{xy}$ in $\text{Co}_3\text{Sn}_2\text{S}_2$ -based alloy films**

A typical hysteresis curve of magnetization and AHE for  $\text{Co}_3\text{Sn}_2\text{S}_2$  film at  $T = 150$  K in **Fig. 2e** shows perpendicular magnetic anisotropy with a large coercive field  $B_c$  of about 1 T. Saturation value of  $M$  at low temperature (Supplementary Fig. 4) is consistent with that in bulk crystals and previous film study on  $\text{Al}_2\text{O}_3$  substrates<sup>26,27,41</sup>. Magnetic field dependence of Hall resistivity  $\rho_{yx}$  presents a clear hysteresis with a comparably large  $B_c$  to that of  $M$ - $B$  curve, implying large AHE. In **Fig. 2f**,  $S_{xy}$  of  $\text{Co}_3\text{Sn}_2\text{S}_2$  film as a function of  $B$  is summarized for elevated temperatures. Across the  $T_C$  at around  $T = 180$  K (Supplementary Fig. 4), the clockwise hysteresis (negative) appears, being opposite hysteresis direction to that of  $M$  and AHE in **Fig. 2e**. At lower temperature than 125 K, the direction of hysteresis is reversed to counterclockwise (positive). The polarity reversal against temperature variation likely reflects the switching of dominant Berry curvature contribution in the intricate band structure around  $E_F$ . The  $B_c$

increases with decreasing the temperature regardless of the hysteresis direction, indicating the  $M$  directing out-of-plane is stabilized. The  $S_{xy}$  value of  $3 \mu\text{V}/\text{K}$  comparable to the bulk value<sup>24,25</sup> is evident to the high crystalline quality of the film.

By substituting Ni and In, the bipolarity of large ANE was demonstrated while the polarity of AHE remains positive in all the compositions in this study. In **Fig. 3a**,  $\rho_{yx}$  with AHE is summarized, displaying the counterclockwise direction of all hysteresis loops for six compositions. Although the two films with rich In ( $y = 0.60$ ) and Ni ( $x = 0.44$ ) present small hysteresis at  $T = 100 \text{ K}$  due to low  $T_C$  (ref. <sup>34</sup>), apparent hysteresis appeared at  $T = 50 \text{ K}$  (Supplementary Fig. 5). Large AHE with positive hysteresis direction for all the films indicates that the Weyl band feature is kept in the films with these compositions (Supplementary Fig. 6). On the contrary, in **Fig. 3b**, the  $S_{xy}$  for four films obviously presents polarity reversal from counterclockwise hysteresis for  $\text{Co}_3\text{Sn}_2\text{S}_2$  and  $x = 0.11$  alloy films to clockwise one for alloy films (See also Supplementary Fig. 7 for magnetic field dependence of transverse thermoelectric conductivity  $\alpha_{xy}$  at  $T = 100$  and  $50 \text{ K}$ ). The counterclockwise hysteresis loop is consistent with the previous  $\text{Co}_3\text{Sn}_2\text{S}_2$  bulk studies<sup>24,25</sup>. At  $T = 50 \text{ K}$ , the negative hysteresis was also observed for  $x = 0.44$  and  $y = 0.61$  (Supplementary Fig. 5). Although the band structure is slightly modified by In substitution, the Weyl feature expectedly remains at less than  $y \sim 0.8$  in theoretical calculation<sup>30</sup>. In addition to the consistent result of the experiments and theoretical calculation for the In substitution, further polarity reversal was experimentally obtained for the

Ni substitution. This is an unexpected finding from the naive expectation based on the rigid band consideration in **Fig. 2c** presenting positive  $\alpha_{xy}^A$  at the upper energy region. The sign reversal of  $S_{ANE}$  is dominated by the first term of  $\rho_{xx}\alpha_{xy}^A$  in the definition due to no sign change in  $\rho_{AHE}\alpha_{xx}$  (Supplementary Fig. 8). In fact, all the  $\alpha_{xy}^A$  for Ni-substituted films is negative value (Supplementary Fig. 9d), which is inconsistent to the naive expectation. While the  $E_F$  shift by Ni substitution seems rigid with keeping Weyl feature in  $\sigma_{xy}$  (ref. <sup>37</sup>), the appearance of polarity reversal in  $S_{xy}$  implies that the realistic band structure of the  $\text{Co}_{3-x}\text{Ni}_x\text{Sn}_2\text{S}_2$  film would be rather different from that of  $\text{Co}_3\text{Sn}_2\text{S}_2$ . The experimental demonstration of bipolarity of  $S_{xy}$  in  $\text{Co}_3\text{Sn}_2\text{S}_2$ -based alloy films is a great step forward to the implementation of controllable ANE in the topological magnets to the thermoelectric energy conversion.

Composition dependences of  $\sigma_{AHE}$  and  $S_{ANE}$ , Hall conductivity  $\sigma_{xy}$  and  $S_{xy}$  at  $B = 0$  T, at  $T = 100$  K are summarized in **Fig. 3c** and **3d**, respectively. The  $\sigma_{AHE}$  value decreases with increasing the substitution content in both cases of In and Ni. This trend comes from the Stoner mechanism for the ferromagnet with a Weyl band structure in  $\text{Co}_3\text{Sn}_2\text{S}_2$  (refs. <sup>31,32</sup>). In contrast, the sign reversal occurs in  $S_{ANE}$ . The largest values for positive and negative  $S_{ANE}$  at  $T = 100$  K are +2.7 and -2.8  $\mu\text{V}/\text{K}$ , respectively. The comparable value is a distinct feature of ANE in topological magnets, in fact ANE in magnetic TI also exhibits comparable positive and negative  $S_{xy}$  by electrostatic tuning<sup>16</sup>. This feature may be widely applied to the topological magnets in consideration of the Fermi energy dependence of  $\alpha_{xy}^A$ , while  $S_{ANE}$  is determined by the balance

of two components in  $S_{ANE} = \rho_{xx}\alpha_{xy}^A - \rho_{AHE}\alpha_{xx}$ . In semiconductors, the large deviation between positive and negative  $S_{xx}$  is well known<sup>1</sup>, which is attributed to the asymmetry of band dispersion at conduction band and valence band. Because the large ANE originates from the Berry curvature around the contribution of band singularity point, it will be possible to extract both large positive and negative ANE by systematic regulation of  $E_F$  across the gap. This distinct feature of topological magnets enables the construction of thermopile with a same base material. In the relation of  $S_{ANE}$  and  $\alpha_{xy}^A$ , we summarized the related physical parameters in supplementary information Supplementary Fig. 8-13. The sign change of  $\alpha_{xy}^A$  originates from the large  $\sigma_{AHE}$  in  $\text{Co}_3\text{Sn}_2\text{S}_2$ -based films (Supplementary Fig. 9), relating on the bipolarity of  $S_{ANE}$ . It is noteworthy that the experimental values of  $\sigma_{AHE}$  at  $T = 2$  K and  $\alpha_{xy}^A$  at  $T = 50$  K are consistent (Supplementary Fig. 12) to the calculated values<sup>30,37</sup>, which evidences the reliable systematic regulation of  $E_F$ . According to the Mott relation, the ratio of  $\alpha_{xy}^A/\sigma_{AHE}$  remains consistently larger values than  $0.1 k_B/e$  for alloy films (Supplementary Fig. 13), which is comparable to the previous values in the bulk alloys<sup>42</sup>. Although  $T_C$  and characteristic temperature exhibiting highest  $S_{ANE}$  value for those films do not match well (Supplementary Fig. 8a and 8b), the peak values of  $S_{ANE}$  are expectedly implemented in the thermopile, demonstrated below.

### **Zero-field operation of anomalous Nernst thermopile**

To materialize the thermoelectric energy conversion via ANE under in-plane thermal gradient, the out-of-plane  $M$  is a fundamental requisite. Large coercive field coming from the large magnetic anisotropy is of great advantage for zero-field operation of ANE devices. Here we compare the positive and negative  $S_{xy}$  values and coercive field  $B_c$  of various materials for availability to the thermopile operation at zero-magnetic-field in **Fig. 4**; large positive and negative  $S_{xy}$  values with the large  $B_c$  are preferable to develop a stable operation of thermoelectric energy conversion. To date, there are many reports on the positive  $S_{xy}$  and rare examples of negative one for MnGa (ref. <sup>9</sup>) and NdFeB (ref. <sup>43</sup>). Under this situation, the reversible feature of  $\text{Co}_3\text{Sn}_2\text{S}_2$ -based alloy films is a significant advantage providing negative  $S_{xy}$  in thermopiles by appropriate selection of In or Ni content. In addition,  $\text{Co}_3\text{Sn}_2\text{S}_2$ -alloy films have large  $B_c$  originating from strong domain-wall pinning (ref. <sup>44</sup>), which is distinct from the bulk  $\text{Co}_3\text{Sn}_2\text{S}_2$ . To apply the film device of thermopile, this feature is critically important because the large  $B_c$  with strong magnetic anisotropy is a few examples in nature.

Finally, we demonstrate the thermoelectric conversion by anomalous Nernst thermopile composed of Weyl magnets of  $\text{Co}_3\text{Sn}_2\text{S}_2$  and  $\text{Co}_3(\text{In},\text{Sn})_2\text{S}_2$ , which provides positive and negative  $S_{xy}$ , respectively. As shown in the schematic (**Fig. 5a**) and a device photograph (**Fig. 5b**),  $\text{Co}_3\text{Sn}_2\text{S}_2$  (yellow) and  $\text{Co}_3(\text{In},\text{Sn})_2\text{S}_2$  (blue) are connected in series. The device was fabricated by repeated sputtering with contact shadow mask. By applying thermal gradient to the device shown in **Fig. 5b**, the voltage ( $V_1$ ,  $V_2$ ,  $V_3$ ,  $V_4$ ,  $V_5$ , and  $V_6$ ) was measured along

orthogonal direction under out-of-plane  $M$ . The temperature gradient in the device is roughly 1.2 K/mm by applying 75 mA. Typical magnetic field dependences of voltage at  $T = 70$  K are presented in **Fig. 5c**. Note that the data points are anti-symmetrized, leading to the extraction of ANE contribution (See also Supplementary Fig. 14). Compared to single line of  $\text{Co}_3\text{Sn}_2\text{S}_2$  (positive) and  $\text{Co}_3(\text{In},\text{Sn})_2\text{S}_2$  (negative), one pair  $n = 1$  is apparently added up from the generated voltages in the each lines. By increasing to  $n = 3$ , the voltage is clearly accumulated by thermopile structure. This is the thermopile operation. As summarized in **Fig. 5d**, the voltage at  $B = 0$  T increases in proportion to the connection number. The linearity of the accumulation comes from the comparable values of positive and negative  $S_{xy}$ . The voltage generation will be increased more by fabrication of large number of pairs with narrow channels. Zero-field operation was characterized at elevated temperature after field-cooling as shown in **Fig. 5e**. Since the Seebeck contribution at the connecting point is superimposed in the raw data (black solid and broken lines), we analyzed it by anti-symmetrized process; the symmetric (gray) and anti-symmetric (red) terms correspond to Seebeck and anomalous Nernst output, respectively. The anomalous Nernst output follows the consistent temperature dependence to the temperature dependence of  $S_{\text{ANE}}$  at  $B = 0$  T in a single film of both  $\text{Co}_3\text{Sn}_2\text{S}_2$  and  $\text{Co}_3(\text{In},\text{Sn})_2\text{S}_2$  (Supplementary Fig. 8a). This result is a good demonstration of thermopile with a same base material of a Weyl magnet at zero-field.

In summary, the polarity reversal of  $S_{xy}$  is clearly demonstrated in a magnetic Weyl semimetal of  $\text{Co}_3\text{Sn}_2\text{S}_2$ -based alloy thin films. The bipolarity of ANE is based on the intrinsic mechanism driven by Berry curvature contribution in a Weyl band feature. In addition to the experimental results consistent to the theoretical prediction for In substitution, the sign reversal by Ni substitution was observed contrary to the theoretical expectation of simple rigid band model, implying the Nernst measurement is sensitive to the variation of band character around the  $E_F$ . By improvement of the availability of theoretical band calculation for alloy materials, the superior topological features will be explored more. A base material of Weyl magnet  $\text{Co}_3\text{Sn}_2\text{S}_2$  with large coercive field is a preferable platform for the anomalous thermoelectric thermopile. The output voltage at zero-magnetic-field was effectively accumulated by fabrication of thermopile with positive and negative  $S_{\text{ANE}}$  in two  $\text{Co}_3\text{Sn}_2\text{S}_2$ -based alloy films. These demonstrations will stimulate the development of methodology and device fabrication for further extracting the topological band feature by systematic  $E_F$  tuning with substitution.

## **Acknowledgments**

We thank S. Nishimura and J. Shiogai for fruitful discussion about the measurement setup for magneto-thermoelectric properties and the NEOARK Corporation for the use of a maskless lithography system PALET. This work was supported by JSPS KAKENHI (Grant Nos. 22H00288, A.T., 21H01789, M.-T.S., 21H04437, M.-T.S., 19H01842, M.-T.S., 20K05299, Y.Y., 21H01031, Y.Y.), JST ERATO (Grant No. JPMJER2201, K.U.), and JST CREST (Grant No. JPMJCR18T2, A.T.).

## **Author contributions**

K.F. grew the thin films. S.N. and K.F. prepared the sample for thermoelectric measurements. S.N. carried out the electrical and thermoelectric measurements with support from K.F.. Magnetization measurement was carried out by K.F. and T.S.. Calibration of the measurement setup for Seebeck effect was carried out by T.H., K.U., and K.F.. First principle band calculation was carried out by Y.Y. and M.-T.S.. S.N., K.F., and A.T. wrote the draft. All the authors discussed the results and commented on the manuscript. K.F. and A.T. conceived the project.

## **Competing Interests**

The authors declare no competing interests.

## Figure legends

**Figure 1. Polarity of Seebeck effect and anomalous Nernst effect.** **a**, Seebeck effect of  $p$ -type and  $n$ -type semiconductors under temperature gradient  $\nabla T$ . **b**, Simple band structure of conventional semiconductor with Fermi energy (broken lines, blue and orange), which corresponds to  $n$ -type to  $p$ -type, respectively. **c**, Longitudinal voltage  $V_{xx}$  induced by temperature difference in  $n$ -type and  $p$ -type semiconductors. Seebeck coefficient  $S_{xx}$  is defined by the slope of this plot; positive and negative corresponds to holes in  $p$ -type and electrons in  $n$ -type semiconductors, respectively. **d**, Anomalous Nernst effect induced orthogonal direction by  $\nabla T$  and magnetization  $M$ . **e**, Simple linearly-dispersive band with Weyl point in Weyl ferromagnets. **f**, Transverse voltage  $V_{xy}$  via Nernst effect exhibiting positive (counterclockwise, orange) and negative (clockwise, blue) hysteretic loops.

**Figure 2. Concept and experimental verification of Weyl feature in  $\text{Co}_3\text{Sn}_2\text{S}_2$  film.** **a**, Band structure of  $\text{Co}_3\text{Sn}_2\text{S}_2$  around Weyl points. **b**, Energy distribution of anomalous Hall conductivity  $\sigma_{\text{AHE}}$ . **c**, Energy distribution of anomalous Nernst conductivity  $\alpha_{xy}^{\text{A}}$ . Data in **a-c** is replotted from ref. <sup>30</sup> with  $M$  directing upward (Supplementary Fig. 1). **d**, Measurement setup for electrical and thermoelectric properties. Top and bottom are a schematic of side view and a photo of top view. In the sample photograph, the upper brownish region represents the film region. The dumbbell-shaped five lines at lower position are Pt/Ti thermometers to evaluate the

temperature at each position. **e**, Magnetic field dependence of magnetization  $M$  and Hall resistivity  $\rho_{yx}$  for  $\text{Co}_3\text{Sn}_2\text{S}_2$  film at  $T = 150$  K. **f**, Nernst coefficient  $S_{xy}$  of  $\text{Co}_3\text{Sn}_2\text{S}_2$  film at various temperatures as a function of magnetic field under applying  $I_{\text{heater}} = 75$  mA. See Methods for the details about the estimation of the sample temperature  $T_{\text{center}} = (T_{\text{high}} + T_{\text{low}})/2$  under thermoelectric measurements.  $T(T_{\text{center}}) = 50$  K (54 K), 75 K (84 K), 100 K (107 K), 125 K (133 K), 150 K (159 K), 160 K (169 K), 165 K (174 K), 175 K (183 K), 200 K (208 K), and 250 K (256 K).

**Figure 3. Bipolar large anomalous Nernst effect.** **a** and **b**, Hall resistivity  $\rho_{yx}$  at  $T = 100$  K and Nernst coefficient  $S_{xy}$  at  $T(T_{\text{center}}) = 100$  K ( $\sim 108$  K) as a function of magnetic field for  $\text{Co}_3\text{Sn}_2\text{S}_2$ -based alloy films, respectively. The  $S_{xy}$  was measured with applying  $I_{\text{heater}} = 75$  mA. The compositions of the alloy films are In ( $y = 0.08, 0.24, 0.41, \text{ and } 0.60$ ) and Ni ( $x = 0.11, 0.21, \text{ and } 0.44$ ). **c** and **d**, Composition dependence of anomalous Hall conductivity  $\sigma_{\text{AHE}}$  and anomalous Nernst coefficient  $S_{\text{ANE}}$  at  $B = 0$  T and  $T(T_{\text{center}}) = 100$  K ( $\sim 108$  K), respectively. The error bars in **d** are due to measurement errors of the sample dimensions.

**Figure 4. Correlations of key parameters, anomalous Nernst coefficient  $S_{\text{ANE}}$  and coercive field  $B_c$ , guiding for stable device operation based on ANE under in-plane thermal gradient and zero-magnetic-field.** The  $S_{\text{ANE}}$  and  $B_c$  for magnetic materials and topological

magnets are summarized to find the specific features of those materials. The values are from  $\text{Co}_2\text{MnGa}$  (ref. <sup>17</sup>),  $\text{UCo}_{0.8}\text{Ru}_{0.2}\text{Al}$  (ref. <sup>18</sup>),  $\text{Co}_3\text{Sn}_2\text{S}_2$  (refs. <sup>24,25</sup>), CBST (Cr-doped  $(\text{Bi,Sb})_2\text{Te}_3$ ) (ref. <sup>16</sup>), FePt and MnGa (ref. <sup>9</sup>), NdFeB,  $\text{Sm}_2\text{Co}_{17}$ , and  $\text{SmCo}_5$  (ref. <sup>43</sup>), and  $\text{YbMnBi}_2$  (ref. <sup>23</sup>).  $\text{Co}_3\text{Sn}_2\text{S}_2$ ,  $\text{Co}_3\text{In}_y\text{Sn}_{2-y}\text{S}_2$ , and  $\text{Co}_{3-x}\text{Ni}_x\text{Sn}_2\text{S}_2$  are displayed as CSS, CISS, and CNSS, respectively. The data for CSS, CISS, and CNSS are the values at  $T = 100$  K shown in Fig. 3b. The plotted values are listed in Supplementary Table 1 in supplementary information. The indexes b (open symbols) and f (solid symbols) mean the bulk and film, respectively.

**Figure 5. Demonstration of zero-field thermopile operation based on Weyl magnet**

**$\text{Co}_3\text{Sn}_2\text{S}_2$ -based alloy films.** **a**, Schematic illustration of thermopile with  $\text{Co}_3\text{Sn}_2\text{S}_2$  (orange region, CSS) and  $\text{Co}_3\text{In}_{0.24}\text{Sn}_{1.83}\text{S}_2$  (blue region, CISS) in series. **b**, A sample picture on the measurement stage. **c**, Thermoelectric voltage  $V$  under applying heater current 75 mA, which typically induces temperature gradient of about 1.2 K/mm in the sample. The measured voltage was anti-symmetrized against  $B$  to extract ANE-derived thermoelectric voltage. **d**, Thermoelectric voltage as a function of connection number. **e**, Temperature dependence of raw data (black solid line for  $+M$  and broken line for  $-M$  condition), and the analyzed data, symmetric (gray) and anti-symmetric (red) terms, are shown for  $n = 3$ . During cooling,  $B = |1|$  T was applied to align the  $M$ . Symmetric and anti-symmetric terms correspond to Seebeck and Nernst term, respectively.



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## Methods

**Sample preparation.** In- or Ni-substituted  $\text{Co}_3\text{Sn}_2\text{S}_2$  films were fabricated by radio-frequency magnetron sputtering method on  $\text{SrTiO}_3$  (111) substrates.  $\text{SrTiO}_3$  substrates were cut and cleaned with ~30 mL acetone and ~30 mL ethanol for 1 min, respectively, in the ultrasonic cleaning bath at room temperature followed by drying with a high-purity (> 99.99995%) nitrogen gas. The substrates were annealed at 1000 °C for two hours in air before installation to the vacuum chamber. After the deposition at 400 °C, the films were annealed at 800 °C for one hour. The films were capped by  $\text{SiO}_x$  (approximately 70 nm) to suppress the re-evaporation during annealing in vacuum and oxidation in the air. We select the elements of In for reducing electrons and Ni for increasing electrons to substitute into Sn and Co site, respectively. We expectedly substitute large composition of In and Ni because there are stable phases of  $\text{Co}_3\text{In}_2\text{S}_2$  and  $\text{Ni}_3\text{Sn}_2\text{S}_2$  in shandite crystal structure<sup>45,46</sup>. The Weyl band structure is expectedly kept up to In  $y = 0.80$  (ref. <sup>30</sup>) and Ni  $x = 0.60$  (ref. <sup>37</sup>) by consideration with theoretical band calculation. There are two Sn sites in the shandite structure, it between Co kagome layers and at Co kagome layer. Although we could not experimentally determine the mainly substituted site in the films, it is important to discuss the In site. Considering our fabrication process with high temperature annealing, the In site seems prefer to the thermodynamically stable Sn site between Co kagome layers as similar as bulk study<sup>47</sup>. The compositions of the alloy films with In ( $y = 0.08, 0.24, 0.41, \text{ and } 0.60$ ) and Ni ( $x = 0.11, 0.21, \text{ and } 0.44$ ) were tuned by target compositions with Ni,

In<sub>2</sub>S<sub>3</sub>, and Co chips on SnS<sub>1.5</sub> plate. A photo of the typical sputtering target is shown in Supplementary Fig. 2d. The composition of films was characterized by energy-dispersive x-ray spectroscopy (EDX). The lattice parameters were evaluated by x-ray diffraction with Cu *K*α. Film thickness is almost 40 nm, which was estimated by x-ray Laue fringes associated with the Co<sub>3</sub>Sn<sub>2</sub>Sn<sub>2</sub>(0006) diffraction (Supplementary Fig. 2a). To measure the electrical and thermoelectric properties, the films were patterned to half area of the substrate. On the other half, Pt/Ti bilayer metal was evaporated in the form of five dumbbells for the thermometer at each position of the substrate surface (see Fig. 2d and S3c). The thermopile was fabricated with contact metal mask. By applying the mask, each film, Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub> and Co<sub>3</sub>In<sub>0.24</sub>Sn<sub>1.83</sub>S<sub>2</sub>, can be selectively fabricated on one substrate. The anomalous Nernst thermopile composed of bipolar materials generates the larger density of output power than that composed of an unipolar material and a nonmagnetic material.

**Electrical and thermoelectric measurements.** The electrical and thermoelectric properties were measured in the physical properties measurement system (PPMS, Quantum Design, Inc.) with home-made heater blocks for applying in-plane temperature gradient. The electrical and thermoelectric measurements were carried out with externally connected electronic equipment (Supplementary Fig. 3), nanovoltmeter (Keithley 2182A), digital multimeter (Keysight 34461A), source measure unit (Keithley 2612A), current source (Keithley 6221A), and semiconductor parameter analyzer (Agilent 4155C). The data acquisition from the equipment

was carried out via Labview program. The data were obtained in a DC mode. The measurement protocol for the temperature dependence of the thermoelectric measurement was the following. First, the sample was cooled to  $T = 200$  K ( $> T_C$ ) at  $B = 0$  T, following the application of a heater current to induce a temperature gradient in the film. Subsequently, the sample was field-cooled to  $T = 10$  K at  $B = +1$  T. After decreasing  $B$  to 0 T, the temperature was increased from  $T = 10$  K to 300 K with sufficient waiting durations for the stable temperature condition. Second, the same sequence with  $B = -1$  T was applied to measure the different magnetization direction. We applied anti-symmetrized calculation to separately evaluate longitudinal and orthogonal terms. The protocol for the magnetic field dependence of the thermoelectric measurement was the following. First, a heater current was applied to induce a temperature gradient at each  $T$ . After waiting for the stable temperature condition, the magnetic field dependence was measured. The temperature for all measurement was controlled by PPMS temperature. The suspended sample setup and measurement configuration are usually applied to the evaluation of the Seebeck and Nernst effect for not only film samples but also bulk samples. It is likely that strain effect is rather weak, which was confirmed by the consistent result of  $\sigma_{\text{AHE}}$  of identical samples in two measurement configurations with usual and suspended one. The heater current was applied to make temperature gradient. For the quantitative estimation of  $S_{xx}$  and  $S_{xy}$ , it is important to evaluate the temperature difference and temperature gradient, respectively. The temperature gradient was evaluated by two/three Pt thermometers on the substrate. The linearity

of temperature versus distance in the sample was confirmed by the measurement using three Pt thermometers under applying  $I_{\text{heater}} = 75$  mA (Supplementary Fig. 3f and 3g). As the heat bath side of the sample was thermally contacted with the base part of the sample holder, the temperature of which was controlled as the system temperature of PPMS, the low-temperature-side temperature was close to it during heating (Supplementary Fig. 3e). The plotted temperature  $T$  in the figure is the temperature of system in PPMS. The difference between PPMS temperature and the temperature at the center of the film obtained by Pt/Ti thermometer under heating was typically 3 ~ 8 K in the examined temperature range of 50 ~ 200 K (Supplementary Fig. 3e and 3g). The temperature at the center of the film  $T_{\text{center}}$  can also be estimated as  $T_{\text{center}} = (T_{\text{high}} + T_{\text{low}})/2$  owing to the linearity of temperature versus distance under applying  $I_{\text{heater}} = 75$  mA (Supplementary Fig. 3f and 3g). We applied the data ( $\sigma_{xx}$ ,  $\sigma_{xy}$ ,  $S_{xx}$  and  $S_{xy}$ ) obtained at the identical temperature of PPMS to calculate the  $\alpha_{xy}^A$  and  $\alpha_{xx}$ . The measured transverse voltage was anti-symmetrized against  $B$  to eliminate spurious contributions, e.g., from the misalignment of potential probes. The electrical contacts were made with an Au wire (diameter = 0.03 mm, purity 99.95%) and an indium solder (purity 99.99%). Contrary to the anti-symmetrized  $S_{xy}$ , the  $S_{xx}$  includes a small but finite thermoelectric contribution induced by the Seebeck effect in the Au wires connected to the sample. To eliminate such a parasitic contribution in our measurement set-up, the  $S_{xx}$  value of Au estimated from the literature<sup>48</sup> was subtracted from the measured  $S_{xx}$  data. The validity of this  $S_{xx}$  correction was confirmed by

evaluating  $S_{xx}$  in a reference 37-nm-thick  $\text{Fe}_{0.63}\text{Sn}_{0.37}$  film on the  $\text{SrTiO}_3$  (111) near room temperature using the above setup and Seebeck Coefficient/Electric Resistivity Measurement System (ZEM-3, ADVANCE RIKO, Inc.), where the thermoelectric contribution of voltage probes is calibrated and the absolute Seebeck coefficient of the sample can be extracted. The  $\text{Co}_3\text{Sn}_2\text{S}_2$  films cannot be used for this calibration because of the presence of the thick  $\text{SiO}_x$  capping layer, which disturbs the good electrical contact between the films and voltage probes. The wire contribution is not changed for different samples unless the wiring configuration is changed.

**Magnetization measurement.** Magnetization of the films was measured using a magnetic properties measurement system (MPMS3, Quantum Design, Inc.).

### **Data Availability**

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

### **Code Availability**

Computer codes used in this study are available from the corresponding author upon reasonable request.

### **Methods-only references**

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