Zero-Field Nernst Effect in a Ferromagnetic Kagome-Lattice Weyl-Semimetal Co₃Sn₂S₂

Satya N. Guin,* Praveen Vir, Yang Zhang, Nitesh Kumar, Sarah J. Watzman, Chenguang Fu, Enke Liu, Kaustuv Manna, Walter Schnelle, Johannes Gooth, Chandra Shekhar, Yan Sun, and Claudia Felser*

The discovery of magnetic topological semimetals has recently attracted significant attention in the field of topology and thermoelectrics. In a thermoelectric device based on the Nernst geometry, an external magnet is required as an integral part. Reported is a zero-field Nernst effect in a newly discovered hard-ferromagnetic kagome-lattice Weyl-semimetal Co₃Sn₂S₂. A maximum Nernst thermopower of \( \pm 3 \mu \text{V K}^{-1} \) at 80 K in zero field is achieved in this magnetic Weyl-semimetal. The results demonstrate the possibility of application of topological hard magnetic semimetals for low-power thermoelectric devices based on the Nernst effect and are thus valuable for the comprehensive understanding of transport properties in this class of materials.

The advantages such as the absence of moving parts and noise make the thermoelectric technology promising for energy conversion and solid-state cooling.[1–6] Its progress largely relies on the Seebeck effect, i.e., the generation of an electrical voltage longitudinal to a temperature gradient. In contrast, configurations based on the Nernst effect, which is the generation of a transverse electrical signal in a magnetic field, have been significantly less studied.[7–9] partly as an external magnetic field is usually required to observe a Nernst signal. In conventional thermoelectric devices based on the Seebeck effect, the heat reservoir is required to be a part of the electrical circuit (Figure 1a). In contrast, the multi-terminal thermoelectric devices based on the Nernst effect enable spatial separation of the heat reservoir from the electric circuitry.[10] Moreover, in Nernst devices, there is no need for both p- and n-type materials as the polarity of the voltage can be reversed by reversing the magnetic field direction.[8,11] Therefore, Nernst devices overcome certain problems of Seebeck devices, where the different thermal expansion coefficients of p- and n-type materials lead to compatibility issues.

In a ferromagnetic material, the generation of an additional electric voltage orthogonal to the applied temperature gradient due to the internal magnetization is referred to as the anomalous Nernst effect (ANE).[11,12] In general, the Nernst thermopower (\( S_{xy} \)) of a soft magnetic material is zero once the magnetic field is removed. Therefore, a part generating an external magnetic field needs to be integrated in devices based on the Nernst effect, which is a big obstacle for the thermomagnetic devices. A possible solution could be the use of a hard magnetic material, where an anomalous Nernst signal (\( S_{xy}^\text{h} \)) can be obtained in zero field. Such a permanent magnetic material remains in a magnetized state even when the external magnetic field is removed.[13,14] Magnetic ferrites and rare-earth-based hard magnets have been extensively studied owing to their wide range of functional applications including magnetic motors, magnetic recording media, magnetic fluids, and electromagnetic wave filters.[13–16] A novel potential application with hard magnets is energy conversion based on the Nernst effect, which has not attracted significant attention. Few examples of such materials include thin films of the alloys FePt, FePd, L₁₁₋ₓMnₓGa, D0₂₂₋ₓMnₓGa, CoₓNi, dilute magnetic semiconductor (DMS) Ga₁₋ₓMnₓAs, and recently Mn₃Sn.[17–20] Therefore, novel hard-magnetic materials are required. Studies on their thermomagnetic properties are of significance in terms of both technological application and fundamental understanding. A promising approach is to search for a new candidate from the library of topological materials as they share many mutual features with the thermoelectric materials.

In the current decade, topological materials attract significant research interest. These newly discovered materials exhibit various physical properties such as chiral anomaly, high carrier mobility, giant magnetoresistance, and mixed axial-gravitational anomaly due to the topological band structure.[21–26] They are
also potential candidates for functional applications in quantum computing, infrared sensors, and heterogeneous catalysis.\cite{27–29} In this context, the discovery of magnetic topological materials created a new frontier in the field.\cite{30–37} These exotic magnetic topological materials provide a new scope in the thermoelectric research as the topological band structure effect can enhance the thermoelectric response.

Here, we show that the topological ferromagnets with large coercive fields ($H_c$) could be potential candidates for observation of the Nernst effect in zero field. As a case study material, we choose Co$_3$Sn$_2$S$_2$, a newly discovered hard-magnetic kagome-lattice Weyl-semimetal. Recently, a large intrinsic anomalous Hall conductivity (AHC) and giant anomalous Hall angle (AHA) have been observed in Co$_3$Sn$_2$S$_2$, originating from the topological band structure.\cite{38,39} We show that a single-crystalline Co$_3$Sn$_2$S$_2$ exhibits a maximum $S_{xy}$ value of $\approx 3 \mu V K^{-1}$ at 80 K in zero field, which is significantly high considering its magnetic moment of $\approx 0.89 \mu_B$ f.u.\cite{40,41} We employed a combined approach including electrical and thermoelectric measurements and first-principles calculations to elucidate this observation. Our complementary electrical transport measurement and density functional theory (DFT)-based calculations indicate that the high Nernst signal in Co$_3$Sn$_2$S$_2$ originates from the topological band structure.

The ternary chalcogenide Co$_3$Sn$_2$S$_2$ is a member of the shandite family $A_3M_2X_2$ (A = Ni, Co, Rh, Pd; M = Pb, In, Sn, Tl; X = S, Se) and crystallizes in a rhombohedral structure (space group: $R3m$).\cite{46,47} In this structure, Sn atoms are distributed over interlayers (Sn1) and in the kagome layers (marked as Sn(2)). The structure consists of the flat hexagonal kagome unit composed of Co and Sn(2) (see the $c$-axis view). The Sn(1) atoms are surrounded by a couple of Co$_3$ triangles in a trigonal-antiprismatic configuration. S atoms capped the Co$_3$ triangles above or below the kagome sheets. The inset shows a magnified plot.

The single crystals of Co$_3$Sn$_2$S$_2$ were synthesized from the elements using a melting reaction followed by slow cooling. The as-grown crystal was characterized by Luue X-ray diffraction (XRD) and powder XRD (see the Experimental Section, and Figure S1, Supporting Information). For the electrical and thermoelectric measurements, the crystal was oriented and cut into a bar shape. As the material exhibits a highly anisotropic out-of-plane magnetization, we used the following configuration: magnetic field ($\mu_0 H$) $||$ $c$-axis and electrical current ($I$) or thermal gradient ($\Delta T$) $||$ $ab$-plane.

Figure 1d shows the magnetization ($M$) as a function of the magnetic field ($\mu_0 H$) for Co$_3$Sn$_2$S$_2$ at different temperatures. Distinct rectangular-shaped hysteresis loops with large coercive fields $H_c$ of $\approx 3650$ and $\approx 570$ Oe are observed at 2 and 100 K, respectively. The origin of the large coercivity is attributed to the strong magnetic anisotropy of the compound (see Discussion, Supporting Information).\cite{42} This distinct hysteresis loop is observed up to $\approx 100$ K; it becomes less pronounced with the further increase in
temperature and vanishes as the temperature approaches $T_C$. The temperature-dependent magnetization measurement indicates a magnetic transition at $\approx 177$ K (Figure S2, Supporting Information). Overall, the magnetic measurement data indicate that Co$_3$Sn$_2$S$_2$ exhibits a sizable $H_C$ up to $\approx 120$ K.

After we demonstrated the hard-magnetic nature of Co$_3$Sn$_2$S$_2$, we investigated the anomalous Nernst thermopower ($S_{xy}^A$) in zero magnetic field. We magnetized the sample by applying a field of $\pm 1$ T ($H_H$) to orient the magnetic moments along the $c$-axis and then switched off the field. Figure 2a presents $S_{xy}^A$ of Co$_3$Sn$_2$S$_2$ in zero field as a function of the temperature. $S_{xy}^A$ increases with the temperature and reaches a peak value of $\approx 3 \mu V K^{-1}$ at $\approx 80$ K. The further increase in the temperature leads to a decrease in $S_{xy}^A$, above $\approx 120$ K, the signal is almost zero as the compound loses its hard-magnetic structure. The observed maximum value of $S_{xy}^A$ for Co$_3$Sn$_2$S$_2$ is remarkable considering the magnetic moment of $\approx 0.89$ $\mu_B$ f.u.$^{-1}$ (see Discussion in the Supporting Information). Further, we investigated the magnetic-field dependence of the Nernst thermopower $S_{xy}(\mu_0H)$ at different temperatures (Figure 2b and Figure S4, Supporting Information). As discussed in the introduction that ferromagnets exhibit ANE below their $T_C$, this system starts to exhibit an anomalous behavior in $S_{xy}$. The effect of the large magnetocrystalline anisotropy of this compound is evident in the $S_{xy}(\mu_0H)$ data. The anomalous behavior in $S_{xy}(\mu_0H)$ (Figure 2b and Figure S4, Supporting Information) can be observed up to the Curie temperature of the compound ($T_C = 177$ K). Above $T_C$ the Co$_3$Sn$_2$S$_2$ does not show any ANE as it is no longer magnetic in nature. Below 100 K, a rectangular-shaped hysteresis loop was observed in the $S_{xy}(\mu_0H)$ data; $S_{xy}$ maintains a plateau value after a certain field. The change of the sign of $S_{xy}$ is due to the flipping of the magnetic moment of the Co atoms with the direction of $\mu_0H$ (positive to negative). This observation is consistent with the zero-field measurement of a magnetized sample. The finite $H_C$ in the data below $\approx 120$ K enables to estimate the zero-field $S_{xy}$, i.e., $S_{xy}$. The temperature-dependent estimated values of $S_{xy}$ from the field sweep data are in good agreement with the temperature-dependent zero-field measurement results (inset in Figure 2b). In the $S_{xy}(\mu_0H)$ data, we observed $S_{xy}^A$ up to $\approx 170$ K by extrapolating the slope of the high-field data; however, estimated $S_{xy}^A$ above $\approx 120$ K is not a true zero-field anomalous value, as the hard-magnetic nature disappears above $\approx 120$ K.

In order to understand the origin of the large $S_{xy}^A$ of Co$_3$Sn$_2$S$_2$, we measured the electrical resistivity ($\rho_{xx}$), longitudinal thermoelectric response, i.e., the Seebeck coefficient ($\alpha_{xx}$), and Hall resistivity ($\rho_{yx}$). The temperature dependence of the electrical resistivity ($\rho_{xx}$) is presented in Figure 3a. $\rho_{xx}$ decreases with the decrease in the temperature reaching a value of $\approx 55 \mu\Omega$ cm at 2 K, indicating the metallic nature of the compound. The anomaly at $T_C = 177$ K in $\rho_{xx}$ reflects the onset of a magnetic transition. The negative sign of $S_{xx}$ is attributed to the dominant n-type charge carriers in Co$_3$Sn$_2$S$_2$ (Figure 3b). $S_{xx}$ linearly increases with the temperature and becomes approximately constant in the range of 50–120 K. Above 120 K, $S_{xx}$ increases with the temperature and exhibits an anomaly near $T_C$.

It is known that the intrinsic band structure effect from the Berry curvature can lead to a large anomalous Hall effect (AHE) in a topological material (see the Experimental Section). Therefore, we estimated the Hall conductivity ($\sigma_{yx}$) of Co$_3$Sn$_2$S$_2$ from the measured electrical resistivity ($\rho_{xx}$) and Hall resistivity ($\rho_{yx}$) using the measured components of resistivity ($\rho_{xx}$, $\rho_{yx}$) and Seebeck and Nernst thermopowers ($S_{xx}$ and $S_{yx}$).

$$\sigma_{yx} = \frac{\rho_{yx}}{\rho_{xx}^2 + \rho_{yx}^2}$$  \hspace{1cm} (1)

Figure 3c presents $\sigma_{yx}$ of Co$_3$Sn$_2$S$_2$ as a function of the magnetic field at different temperatures. The trend and values of the AHC ($\sigma_{yx}$) are fully consistent with previous observations.[18] Below 100 K, a large AHE is observed with a sharp rectangular-shaped loop, which arises from the topological-band-structure-enhanced Berry curvature effect.[38] This observation indicates the topological-band-structure-related origin of the large $S_{yx}$ of Co$_3$Sn$_2$S$_2$ as the transverse thermoelectric response is associated with the Berry curvature, as discussed below.

The notion of the topological band structure effect in transverse thermoelectric responses can be understood by analyzing the temperature dependence of the anomalous transverse thermoelectric conductivity ($\alpha_{yx}$). Therefore, we estimated the transverse thermoelectric conductivity ($\alpha_{yx}$) using the measured components of resistivity ($\rho_{xx}$, $\rho_{yx}$) and Seebeck and Nernst thermopowers ($S_{xx}$ and $S_{yx}$).

$$\alpha_{yx} = \frac{S_{yx}^A \rho_{yx} - S_{xx} \rho_{yx}}{\rho_{xx}^2 + \rho_{yx}^2}$$  \hspace{1cm} (2)
Figure 3. Electrical transport, Seebeck coefficient, and thermoelectric conductivity. a) $T$-dependent electrical resistivity ($\rho_{xy}$). The change in slope in $\rho_{xy}$ around $\approx 177$ K corresponds to a magnetic transition (marked with an arrow). b) $T$ dependence of the Seebeck coefficient ($S_{xy}$) of Co$_3$Sn$_2$S$_2$. The $T$ dependence of the AHC ($\sigma_{xy}^\text{ANC}$) is shown in the inset. d) $T$ dependence of the experimentally determined anomalous transverse thermoelectric conductivity ($\sigma_{xy}^\text{ANC}$). The dotted line indicates magnetic transition temperature.

Figure 3d shows the anomalous part of $\alpha_{xy}^N$ of Co$_3$Sn$_2$S$_2$. The $S_{xy}$ and $\rho_{xy}$ exhibit anomalous behavior below $T_C$. Therefore, a nonzero $\alpha_{xy}^N$ value can be observed only below $T_C$. In Co$_3$Sn$_2$S$_2$, the $\alpha_{xy}^N$ shows a peak value around 150 K and decreases upon cooling, although the saturation magnetization increases. A similar observation was reported in an anomalous Hall resistance measurement.[38] The observation is an evidence that the anomalous Nernst signal originates from a different source, rather than from the magnetization.

For a deeper understanding, we carried out DFT- and ab-initio-based electronic structure calculations and numerical simulations of $\sigma_{xy}^N$ and $\alpha_{xy}^N$ using the Kubo formula. Figure 4a shows the calculated electronic structures of Co$_3$Sn$_2$S$_2$ with and without spin-orbit coupling (SOC). The spin-down channel of the bands has a gap of $\approx 0.35$ eV, while the spin-up channel is semimetallic (see also Figure S6, Supporting Information). Moreover, for the spin-up channel along the $\Gamma$–L and L–U paths, linear band crossings can be observed near the charge-neutral Fermi point ($E_F$). In the absence of SOC, the full Hamiltonian could be split into the direct product of a spinless Hamiltonian and spin-Hamiltonian, in which the band degeneracy of each spin channel is determined by the spinless Hamiltonian. Owing to the mirror symmetry of the lattice, the band inversion of the spin-up channel forms a nodal line in the mirror plane ($M_L$). Considering the combination of inversion and $C_1$ rotational symmetries, there are six nodal lines in total (Figure 4b). The application of SOC breaks the mirror symmetry and lifts the degeneracy of the nodal lines, with one pair of Weyl points remaining on it.

Figure 4c shows the computed $\alpha_{xy}^N$ as a function of the position of the Fermi level ($E_F$) at 80 K. We would like to note that unlike intrinsic AHE, the intrinsic part of $\alpha_{xy}^N$ depends on the strength of the Berry curvature near $E_F$, rather than on the Berry curvature of all of the occupied bands (see the Experimental Section).[33,44] The anomalous Nernst conductivity (ANC) of Co$_3$Sn$_2$S$_2$ exhibits a strong $E_F$ dependence, which is not observed for $\sigma_{xy}^N$ (Figure S7, Supporting Information). $\sigma_{xy}^N$ maintains a plateau value ($\approx 1000$ S cm$^{-1}$) over an energy window of $\approx 100$ meV around $E_F$. In contrast, $\alpha_{xy}^N$ has a peak value slightly away from $E_F$ and then rapidly decreases away from $E_F$ due to the modulation of the Berry curvature strength. This finding was further supported by our temperature-dependent $\sigma_{xy}^N$ and $\alpha_{xy}^N$ data. The experimental $\sigma_{xy}^N$ exhibits a small change below $\approx 100$ K. In contrast, $\alpha_{xy}^N$ exhibits a monotonous decreasing trend with the decrease in temperature.

In a previous study, angle-resolved photoelectron spectroscopy and low-temperature Shubnikov–de Haas quantum oscillation analysis indicated that $E_F$ was slightly shifted toward the valence bands in Co$_3$Sn$_2$S$_2$.[38] In this study, we use a sample with a similar quality to that for the AHE, indicating a similar position of $E_F$. We calculated the temperature dependence of $\alpha_{xy}^N$ below ordering temperature using different positions of $E_F$ of Co$_3$Sn$_2$S$_2$ (Figure S8, Supporting Information). $\alpha_{xy}^N$ exhibits a monotonous increase with $T$ for $E_F = E_0 = 0.07$ eV to $E_F = 0.10$ eV and well agrees with the experimental value when $E_F = E_0 = 0.08$ eV (Figure 4d). Moreover, the temperature-modified Berry curvature distributions ($\Omega_{\sigma}$) at 80 and 150 K for $E_F = E_0 = 0.08$ eV...
also have large nonzero values (Figure 4b and Figure S9, Supporting Information), suggesting a topological band structure contribution in the transverse thermoelectric transport.

Figure 5 compares the absolute value of peak $S_{xy}$ of different class of materials from the literature with Co$_3$Sn$_2$S$_2$.\[17–19,34,45–49\] A maximum $S_{xy}$ of $\approx 3 \mu V K^{-1}$ at $T = 80$ K is measured in the case of Co$_3$Sn$_2$S$_2$. In addition, Co$_3$Sn$_2$S$_2$ exhibits a zero-field signal, which is not the case for nonmagnetic and soft-ferromagnetic metals. Although the maximum $S_{xy}$ of Co$_3$Sn$_2$S$_2$ ($\approx 3 \mu V K^{-1}$ at $T = 80$ K) is lower than that of topological ferromagnet Co$_2$MnGa ($\approx 6 \mu V K^{-1}$ at $T = 300$ K) and DMS Ga$_{0.93}$Mn$_{0.07}$As ($\approx 8.1 \mu V K^{-1}$ at $T = 10$ K), but the present results provide a possible guiding principle for the observation of large zero-field anomalous Nernst thermopower in a hard-ferromagnetic topological semimetal at elevated temperature. Moreover, considering the magnetic moments of $\approx 0.79 \mu B f.u.$ for Co$_3$Sn$_2$S$_2$ ($T = 80$ K) and $\approx 3.7 \mu B f.u.$ for Co$_2$MnGa ($T = 300$ K), the present result is more interesting and due to lower magnetic moment the inherent stray field for Co$_3$Sn$_2$S$_2$ will be low. Figure S10 in the Supporting Information compares the ratios of the strengths of the anomalous Nernst signals to their saturation magnetizations of different hard and soft ferromagnetic metals and antiferromagnet Mn$_3$Sn.\[17,19,34,45–49\] The Y-axis of the plot ($S_{xy}/\mu_B M$) represents the ratio of $S_{xy}$ to the magnetic moment ($\mu_B M$). The values of $S_{xy}/\mu_B M$ for the topological ferromagnets are higher than those of the trivial ferromagnetic materials. A similar result was observed for the topological chiral antiferromagnet Mn$_3$Sn; however, it exhibited a lower Nernst signal than that of Co$_3$Sn$_2$S$_2$ (Figure S10, Supporting Information).\[19\] $S_{xy}/\mu_B M$ indicates that the strength of the transverse signal is significantly higher in Co$_3$Sn$_2$S$_2$ and indeed highest among those of ferromagnetic metals. This implies that the anomalous transport properties originate from the topological band structure effect and are independent of the strength of the magnetic moments.

Figure 4. Electronic structure and calculated transverse thermoelectric conductivity. a) Electronic structures of Co$_3$Sn$_2$S$_2$ with and without SOC. The application of SOC breaks the mirror symmetry leading to gapping of the nodal lines due to the degeneracy lifting of the bands. b) Temperature-modified Berry curvature distribution ($\Omega_N$) at 80 K in the Brillouin zone with the magnetization along the c-axis for $E_F = E_0 - 0.08$ eV. The green lines indicate the nodal rings. c) Theoretically calculated ANC ($\alpha_{\alpha N}$) as a function of the chemical potential with respect to the charge neutrality point $E_0$ at $T = 80$ K. d) Theoretically estimated temperature-dependent $\alpha_{\alpha N}$ with $E_F = E_0 - 0.08$ eV below ordering temperature. The calculated value is in good agreement with the measured value.

Figure 5. Absolute value of anomalous Nernst thermopowers ($S_{xy}$) for ferromagnetic metals, antiferromagnet Mn$_3$Sn, DMS Ga$_{0.93}$Mn$_{0.07}$As from the literature and Co$_3$Sn$_2$S$_2$. The shaded region indicates hard and soft ferromagnetic metals.\[17–19,34,45–49\]
In conclusion, magnetic topological semimetals are potential candidates for the observation of exotic anomalous transport properties. We showed that the hard ferromagnetic kagome-lattice Weyl-semimetal \( \text{Co}_3\text{Sn}_2\text{S}_2 \) from the shandite family exhibited a large ANE in zero field. Our experimental and DFT calculation results showed that the large ANE originated from \( \text{Co}_3\text{Sn}_2\text{S}_2 \) was not sufficiently high for applications. This study shows that the topological material with a large \( H_L \) could be a potential candidate for a transverse thermoelectric and that the large signal can be achieved in zero field. Therefore, it is of interest to search for new hard magnets with high Curie temperatures from the library of magnetic topological materials.

**Experimental Section**

Single-Crystal Growth of \( \text{Co}_3\text{Sn}_2\text{S}_2 \) and Characterization: The single crystals of \( \text{Co}_3\text{Sn}_2\text{S}_2 \) were grown using an elemental melting reaction followed by a slow cooling. As-purchased high-quality elemental cobalt (99.999%), tin (99.999%), and sulfur (99.999%) were used for the synthesis. Stoichiometric amounts of the elements (Co:Sn:S \( =3:2:2 \)) were loaded in an alumina crucible and then sealed in a quartz tube. The tube was heated to \( 1323 \text{ K} \) over a period of \( 48 \text{ h} \), soaked for \( 24 \text{ h} \), and then slowly cooled down to \( 873 \text{ K} \) over 7 days. The power XRD measurement was performed at room temperature using a Huber Image Plate Guinier Camera G670 operated with Cu K\( _{\alpha1} \) radiation (\( \lambda = 1.54056 \text{ Å} \)).

The single crystal of the as-grown crystal was evaluated by the white-beam backscattering Laue XRD method. Well-characterized and aligned crystals were cut into bar shapes for transport and magnetization measurements. The typical dimensions of the crystals used for the electrical and thermal transport measurements were \( 7 \times 1.3 \times 0.4 \text{ mm}^3 \).

Electrical Transport and Magnetization Measurements: The electrical transport properties were measured using a PPMS9 instrument (ACT option, Quantum Design). The standard four-probe method was used in all of the measurements. In order to correct for contact misalignment, the measured data were field-symmetrized and antisymmetrized. The magnetization measurement was performed using a Quantum Design PPMS1 instrument.

Thermoelectric Transport Measurements: All of the thermal transport experiments were performed in a physical property measurement system (PPMS) cryostat. The Seebeck and Nernst thermoelectric measurements were carried out in the one-heater two-thermometer configuration. In the field sweep experiments in a temperature range of \( 13\text{–}310 \text{ K} \), the PPMS as well as an external nanovoltmeter and current source (Keithley) were controlled using LabVIEW. The temperature gradient was generated using a resistive heater, connected to a gold-coated flat copper wire at one end of the sample. The thermal gradient \( \Delta T \) was applied along the ab-plane of the sample, while the magnetic field was applied along the c-axis. Another gold-plated flat copper wire was attached to the puck clamp for the heat sink. In order to measure the temperature gradient, two gold-plated copper leads were attached directly to the sample using a silver-filled epoxy along the thermal gradient direction. The distance between the thermometers was \( =3.5 \text{ mm} \). In the Seebeck and Nernst measurements, \( \Delta T \) was typically set to \( =1\text{–}3\% \) of the base temperature. Two copper wires were attached using the silver epoxy, orthogonal to the thermal gradient direction, to measure the transverse voltage. In order to correct the data for contact misalignment, the measured data were field-symmetrized and antisymmetrized. The thermal conductivity measurement was carried out in the PPMS using the thermal transport option.

Anomalous Nernst Thermopower of Different Class of Materials from the Literature: The highest absolute value of anomalous Nernst thermopowers (\( \Delta S_n \)) were used for the different soft and hard ferromagnets, antiferromagnet \( \text{MnS}_n \), and DMS \( \text{Ga}_{0.93}\text{Mn}_{0.07}\text{As} \) well below their Curie temperatures. Their magnetizations (\( \mu_0 \text{M} \) in Tesla) were used to calculate the ratios (\( \text{Co/NI film (300 K; ref. [17])} \), \( \text{Li}_2\text{FePt (300 K; ref. [17])} \), \( \text{DO}_{32}\text{MnS}_9\text{Ga (300 K; ref. [17])} \), \( \text{Li}_2\text{MnGa (300 K; ref. [17])} \), \( \text{Li}_2\text{FePd (300 K; ref. [17])} \), \( \text{Ga}_{0.93}\text{Mn}_{0.07}\text{As} (10 K, ref. [18]) \), \( \text{MnS}_n (200 K; ref. [19]) \), \( \text{Co/MnGa (300 K; refs. [34,36])} \), \( \text{NdM}_{0.2}\text{O}_{0.8} (T < \text{T}_c = 73 \text{ K}, B = 1 \text{ T} \) [111] \), \( \text{Fe (300 K; ref. [46])} \), \( \text{Co (300 K; ref. [46])} \), \( \text{FeO}_4 (300 \text{ K}, B < 0.8 \text{ T}; ref. [47]) \), \( \text{MnGe (100 K, B > 5 \text{ T}; ref. [48])} \), and \( \text{PtFe multilayer (N = 9, 300 K, B < 5 \text{ T}; ref. [49])} \).

Ab Initio Calculations: For ab initio calculations, the DFT was employed using the Vienna ab initio simulation package.[50] including the exchange–correlation energy through the Perdew–Burke–Ernzerhof functional. For the integrations in the \( k \)-space, a grid of \( 19 \times 19 \times 19 \) points was used. The Wannier functions from the resulting band structure was extracted using WANNIER90[51] to set up a tight-binding Hamiltonian, which reproduced the DFT band structure within a few millielectronvolts. With this Hamiltonian, the Berry curvature \( \Omega_{ij} \) was calculated as

\[
\Omega_{ij} = \lim_{\Delta \omega \to 0} \sum_{\mathbf{k}} \frac{1}{2\pi i} \int \frac{d \mathbf{k}}{2\pi} \epsilon_{\mathbf{k}} \partial_{\mathbf{k}j} \left( \partial_{\mathbf{k}i} \epsilon_{\mathbf{k}} \right) \left( \delta_{\mathbf{H}} \right) \left( \mathbf{k} \right) \left( \delta_{\mathbf{H}} \right) \left( \mathbf{k} \right)
\]

where \( m \) and \( n \) are the eigenstates and \( \epsilon \) are the eigenenergies of the Hamiltonian \( H \).

Subsequently, the AHC \( \alpha_{ij} \) was calculated as

\[
\alpha_{ij} = -\frac{\epsilon_{ij}}{\hbar} \sum_{\mathbf{k}} \frac{d \mathbf{k}}{2\pi} \epsilon_{\mathbf{k}} \left( \delta_{\mathbf{H}} \right) \left( \mathbf{k} \right) f_{\mathbf{k}}
\]

where \( f_{\mathbf{k}} \) is the Fermi–Dirac distribution for a band \( n \) at a \( k \)-point. The ANC \( \alpha_{ij}^N \) below ordering temperature is

\[
\alpha_{ij}^N = \frac{\hbar}{2T} \int \frac{d \mathbf{k}}{2\pi} \epsilon_{\mathbf{k}} \left( \delta_{\mathbf{H}} \right) \left( \mathbf{k} \right) \left( \delta_{\mathbf{H}} \right) \left( \mathbf{k} \right)
\]

If \( \beta = k_B T \) was defined, where \( k_B \) is the Boltzmann constant, the temperature-modified Berry curvature distribution can be expressed as

\[
\Omega_N = \sum_{\mathbf{k}} \int_{\mathbf{B}_N} \left( \epsilon_{\mathbf{k}} - \mu \right) f_{\mathbf{k}} + k_B T \ln \left( 1 + e^{-\left( \epsilon_{\mathbf{k}} - \mu \right)/k_B T} \right)
\]

\( \Omega_N \) has a finite value only around the Fermi energy. Therefore, \( \alpha_{ij}^N \) is related to the summation of the Berry curvatures of all of the occupied bands below the Fermi energy. In contrast, \( \alpha_{ij}^N \) is related to the Berry curvature of the occupied bands near the Fermi energy. For the integrations over the whole Brillouin zone in the AHE and ANE calculations, a mesh of \( 251 \times 251 \times 251 \) \( k \)-points was employed, providing converged results. Discussion on finite-temperature DFT calculation has been given in the Supporting Information.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

**Acknowledgements**

This study was financially supported by the ERC Advanced grant no. (742068) "TOP-MAT", and Deutsche Forschungsgemeinschaft DFG under SFB 1143. S.N.G., Ch.F., and E.K.L. thank the Alexander von Humboldt Foundation for the fellowships. E.K.L. also thanks the National Natural Science Foundation of China (no. 51722106).

**Conflict of Interest**

The authors declare no conflict of interest.
