

Large Anomalous Nernst Effect and Topological Nernst Effect in The Noncollinear Antiferromagnet NdMn₂Ge₂

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Abstract

The ThCr₂Si₂-type NdMn₂Ge₂ was recently proposed to be a magnetic Skyrmion candidate exhibiting topological Hall effect. In this paper, we report magnetic, electronic, and thermoelectric measurements on NdMn₂Ge₂ single crystals. We show that, in the canted antiferromagnetic phase and the conical magnetic phase with a large out-of-plane ferromagnetic component, this system exhibits both anomalous Nernst effect and topological Nernst effect. We argue that NdMn₂Ge₂ is a unique magnetic material with its topological characters associated with the Berry phases in both real space and momentum space.

Introduction

In nonmagnetic metals, in the presence of a longitudinal electric field and a perpendicular magnetic field, a transverse voltage is induced due to the Lorentz force, which is known as normal Hall effect. In ferromagnetic metallic systems, an additional transverse voltage emerges, termed as anomalous Hall effect (AHE), which is fundamentally driven by the spin-orbit interaction. The anomalous Hall resistivity is often proportional to spontaneous magnetization instead of external magnetic field [1,2]. Recently, a third contribution to the Hall effect, coined as topological Hall effect (THE), was proposed in some magnetic systems with noncoplanar spin configurations [3-7]. Owing to the noncoplanar spin structure, the resultant non-zero spin chirality $\chi = \mathbf{S}_i \cdot (\mathbf{S}_j \times \mathbf{S}_k)$, where $\mathbf{S}_i$, $\mathbf{S}_j$, and $\mathbf{S}_k$ are three nearest-neighboring spins, induces a nontrivial Berry phase and an associated fictitious magnetic field that gives rise to the THE [8-12]. The THE has been detected in magnetic Skyrmion systems which host swirling vortex-like spin textures.

As the counterpart of electronic transport measurements, thermoelectric transports measurements are effective probes to study thermoelectric properties of magnetic materials [12-17]. For instance, the anomalous Nernst effect (ANE), a transverse voltage generated in the presence of a longitudinal temperature gradient, is found to be associated with the Berry curvature of electronic bands near the Fermi level and thus has been recently applied in studying topological electronic materials [18,19]. From the technological viewpoint, materials showing appreciable Nernst effect are promising candidates for both thermoelectric and spintronics applications

[20,21]. In comparison to the electric Hall effect, both ANE and topological Nernst effect (TNE) have been much less studied due to the lack of suitable candidate materials.

Very recently, it was reported that NdMn_2Ge_2 , a centrosymmetric compound with ThCr_2Si_2 -type structure, exhibits a large THE with a maximum value of $\sim 2.05 \mu\Omega \text{ cm}$ over a wide range of magnetic field and temperature [22,23]. And the Lorentz transmission electron microscopy (LTEM) study revealed the existence of magnetic Skyrmions in this compound [22]. Therefore, NdMn_2Ge_2 is a great candidate material to study its thermoelectric properties. In this paper, we report comprehensive magnetic, electronic, and thermoelectric transport measurements on NdMn_2Ge_2 single crystals. We show that NdMn_2Ge_2 exhibits both ANE with $|S_{xy}^A| \sim 1.4 \mu\text{V K}^{-1}$ and TNE with $|S_{xy}^T| \sim 0.3 \mu\text{V K}^{-1}$. The anomalous thermoelectric linear response tensor $|\alpha_{xy}^A|$ reaches $\sim 0.6 \text{ A m}^{-1} \text{ K}^{-1}$ and the $|S_{xy}^T|$ is found to be comparable to the values reported previously in other topological systems.

Methods

NdMn_2Ge_2 single crystals were grown using In-flux method. High-purity elements Nd, Mn, Ge, and In as starting materials were mixed with the molar ratio of 1:2:2:30 and sealed in a vacuumed quartz tube. The quartz tube was loaded into a box-type furnace which was heated to 1100°C and kept at this temperature for 10 hours for the raw materials to be thoroughly melted. Then the furnace was slowly cooled down to 700°C at a rate of 3°C per hour before taking out the quartz tube for centrifuging. Afterwards, the as-grown shiny, plate-like single crystals with a typical

size of $3\text{mm} \times 2\text{mm} \times 0.2\text{mm}$ were obtained. The magnetic susceptibility was measured using a Quantum Design SQUID magnetometer (MPMS3), and the electronic transport measurements were carried out using a Quantum Design cryostat (PPMS). Single crystal neutron diffraction measurements were performed using the HB1A triple-axis spectrometer at High Flux Isotope Reactor in Oak Ridge National Laboratory. The energy of incident neutrons was fixed as 14.6 meV. A single crystal sample was oriented in the $(H\ 0\ L)$ and $(H\ K\ 0)$ planes, where H , K , and L are in reciprocal lattice units (r.l.u.). The sample was mounted inside a closed-cycle helium refrigerator with a measurement temperature range of 5 - 300 K. Thermoelectric measurements were carried out using a modified sample puck designed to be compatible with the PPMS cryostat. The temperature was probed using type-E (Constantan-Chromel) thermocouples, and the thermoelectric voltage was measured using a Keithley Instruments K2182A nanovoltmeter. For all measurements presented in this work, the magnetic field was applied along the c -axis.

Results

NdMn_2Ge_2 compound crystallizes in tetragonal space group $I4/mmm$ (No. 139) with lattice parameters $a = b = 4.110\ \text{\AA}$ and $c = 10.845\ \text{\AA}$. Figure 1(a) illustrates the schematic crystal structure of NdMn_2Ge_2 . The neighboring Mn (and Nd) atoms form square lattices in the ab plane with the neighboring Mn (Nd) layers separated from each other by $c/2$. Previous neutron powder diffraction measurements revealed that this system undergoes multiple magnetic phase transitions upon cooling: a paramagnetic-to-antiferromagnetic transition occurs at $\sim 480\ \text{K}$ below which Mn

spins form a collinear antiferromagnetic spin structure; below ~ 330 K, the magnetic structure changes to a canted antiferromagnetic structure with a net ferromagnetic component of Mn spins aligned along the c -axis, which is subsequently followed by another magnetic transition to a conical spin structure below ~ 250 K; finally a ferromagnetic ordering of the Nd sublattice at ~ 100 K was reported [22,24].

In Fig. 1(b), we plot the temperature dependence of magnetic susceptibility $\chi(T)$ measured with a magnetic field of 0.1 T applied along the c -axis under zero-field cooling (ZFC) and field-cooling (FC) conditions. As clearly seen from the $d\chi/dT$ (blue) curve, within the measurement temperature range NdMn_2Ge_2 exhibits three magnetic phase transitions at $T_c \sim 337$ K, $T_{\text{SR}} \sim 216$ K, and $T_{\text{Nd}} \sim 21.5$ K, corresponding to the collinear-to-canted antiferromagnetic transition of Mn spins, the spin-reorientation of Mn spins, and the ferromagnetic ordering of Nd spins, respectively. These features are consistent with a recent report [23]. Additionally, another anomaly at $T_{\text{icm}} \sim 246$ K is also visible, as seen in the expanded view of $\chi(T)$ shown in the inset, which corresponds to the transition to incommensurate conical spin structure (as will be discussed later).

Figure 1(c) shows the temperature dependence of in-plane resistivity ρ_{xx} and its derivative $\frac{d\rho_{xx}}{dT}$ measured at zero field. The residual-resistivity-ratio (RRR) reaches a value of ~ 160 , indicating high-quality of the as-grown single crystals. Notably, while ρ_{xx} monotonically increases with increasing temperature, an anomaly is clearly seen in $\frac{d\rho_{xx}}{dT}$ at T_{SR} . In addition, a kink in $\frac{d\rho_{xx}}{dT}$ near the canted antiferromagnetic phase transition T_c is also observed (see the dashed line). In Fig.1(d), we present the

temperature dependent Seebeck coefficient S_{xx} with a thermal gradient applied in the ab plane. The positive S_{xx} value over the whole measurement temperature range indicates that the p-type (hole) charge carriers dominate in NdMn₂Ge₂. S_{xx} shows a broad peak with a value of $13.6 \mu\text{V K}^{-1}$ around T_{icm} and a kink near T_c . These features suggest that both electronic and thermoelectric transport properties of NdMn₂Ge₂ are closely correlated with the complex magnetic configurations of this system.

To revisit the magnetic structure of NdMn₂Ge₂, we performed single crystal neutron diffraction measurements. Figures 2(a-c) show false-color contour maps of T - $[1\ 0\ L]$, T - $[0\ 0\ L]$, and T - $[H\ H\ 0]$ measured at 0 T. Two features are clearly seen. First, a commensurate-to-incommensurate phase transition occurs at T_{icm} with incommensurate magnetic Bragg peaks of $(1\ 0\ 1\pm\delta)$, where δ increases with decreasing temperature and then nearly saturates with a value of 0.217(1) at low temperature. Second, the neutron diffraction intensity of nuclear Bragg reflections also varies with temperature, indicating the contribution from magnetic moment that superimposes with the nuclear Bragg peak intensity. Figure 2(d) shows the temperature dependence of integrated neutron scattering intensity of $(0\ 0\ 2)$, $(1\ 1\ 0)$, $(2\ 0\ 0)$, and $(1\ 0\ 1)$, and $(1\ 0\ 1\pm\delta)$ Bragg reflections. Note that a ferromagnetic component of Mn spins gives rise to enhanced diffraction intensity of Bragg reflections with $H + K = \text{even}$, while an antiferromagnetic ordering of Mn spins yields an enhancement in diffraction intensity of Bragg reflections with $H + K = \text{odd}$ [24]. Since neutrons couple to the magnetic moment perpendicular to the moment transfer

$\mathbf{q}$, the observed monotonic increase in the diffraction intensities of (0 0 2), (1 1 0) and (2 0 0) over the whole measurement temperature range suggests that the net ferromagnetic moment of Mn spins in both the canted antiferromagnetic state ($T_{\text{icm}} < T < T_c$) and the conical spin state for $T_{\text{SR}} < T < T_{\text{icm}}$ and $T < T_{\text{SR}}$ is not strictly aligned either along the c -axis or in the ab plane. This is distinct from the previous neutron power diffraction studies [24] which concluded that the net ferromagnetic moment of Mn is along the c -axis for $T_{\text{SR}} < T < T_c$ and in the ab plane for $T < T_{\text{SR}}$, while indeed the net ferromagnetic component of Mn tends to tilt from the c -axis toward the ab plane when cooling across T_{SR} as seen from the magnetic susceptibility data shown in Fig. 1(b) (also see Ref. [23]). The sharp increase in the (0 0 2) Bragg intensity below 25 K, which is absent in both (2 0 0) and (1 1 0) Bragg reflections, arises from the long-range ordering of Nd spins aligned in the ab -plane. In addition, the commensurate-to-incommensurate phase transition below T_{icm} is clearly indicated by the decrease/increase of the diffraction intensity of (1 0 1)/(1 0 1 $\pm\delta$) Bragg peaks. The schematic sketches of magnetic structures are presented in Fig. S1 of Supplemental Material (SM) [25].

We now turn to the discussion on thermoelectric measurement results. Figure 3(a) shows the magnetic field dependence of Nernst coefficient $-S_{xy}$ measured at various temperatures. A thermal gradient is applied along the longitudinal direction (x -axis) and the magnetic field is applied along the perpendicular direction ($c(z)$ -axis), and the voltage is measured along the transverse direction (y -axis). One can see that above 220 K $-S_{xy}$ increases rapidly at low fields and then saturates while the saturation

field gradually increases at lower temperatures. The latter feature is associated with the spin reorientation transition at $\sim T_{\text{SR}}$ with the net ferromagnetic moment tilting from nearly c -axis toward the ab plane, consistent with $M(H)$ data shown in Fig. S2 [25]. There is no visible hysteresis loop in both $-S_{xy}$ and $M(H)$ curves, implying that domain walls propagate smoothly without flux pinning in NdMn_2Ge_2 during the field sweeping process. Alike the field-dependent ρ_{yx} (Fig. S3(a)) [25], the saturated $-S_{xy}$ at high fields suggests the nature of anomalous Nernst effect (ANE). As discussed previously, recent electronic transport measurements revealed that, in addition to the normal Hall effect and AHE contribution, the third contribution-THE, which stems from the finite scalar spin chirality associated with the noncoplanar spin configuration that gives rise to non-zero real-space Berry curvature, emerges [23]. As a result, one anticipates observing the topological Nernst effect (TNE) contribution to the total Nernst effect. That is, $S_{xy} = S_0 + S_{xy}^A + S_{xy}^T$, where S_0 , S_{xy}^A , and S_{xy}^T represent the normal ($\sim H$), anomalous ($\sim M$), and topological contributions to the Nernst effect, respectively.

To extract the topological contribution to the observed Nernst signal, S_{xy}^T , we overplot the $-\Delta S_{xy}(H)$ and the magnetization data $M(H)$ for each temperature measured. $\Delta S_{xy}(H)$ is obtained after subtracting the S_0 term from the measured S_{xy} by performing linear fitting using the high field data. For instance, we present such data measured at $T = 260$ K in Fig. 3(b). It is clearly seen that the magnetic field dependence of ΔS_{xy} and M do not overlap exactly in the low field region, suggesting a topological contribution to the observed Nernst effect. By assuming that S_{xy}^A is

proportional to M , we can further subtract the S_{xy}^A term from ΔS_{xy} to obtain the TNE contribution S_{xy}^T . The thus-extracted $-S_{xy}^T$ at 260 K is represented by the blue curve in Fig. 3(b), which shows a broad peak with a maximum value of $\sim -0.26 \mu V K^{-1}$ at the low field. $-S_{xy}^T$ approaches zero when H is close to the saturation field. These features are similar to those found in Fe_3Sn_2 which hosts magnetic Skyrmions [26].

In Fig. 3(c) we present the field dependence of $-S_{xy}^T$ at various temperatures, and a T - H contour map of $-S_{xy}^T$ is shown in Fig. 3(d). It is obviously seen that $-S_{xy}^T$ reaches high values within a narrow T - H region, a feature similar to what is found in the THE (ρ_{yx}^T) data shown in Fig. S3(b-d) [25]. The temperature dependence of the maximum $|-S_{xy}^T|$ measured at each temperature is shown in the inset of Fig. 3(d). For comparison, we also plot the temperature dependence of the maximum $|\rho_{yx}^T|$. Overall, the shapes of $|-S_{xy}^T|$ and $|\rho_{yx}^T|$ are similar, showing nonmonotonic temperature dependence. Both curves exhibit a plateau-like feature with large TNE and THE between 220 K to 320 K, implying that the TNE and THE behaviors are of the common physical origin. Recent LTEM studies revealed the existence of Skyrmion bubbles, however, the Skyrmion bubbles persist in a wide temperature range from 310 K down to 150 K (at which both TNE and THE signal are quite small) and do not show noticeable magnetic field dependence. This suggests that the TNE and THE behaviors are not mainly driven by the non-zero spin chirality associated with Skyrmion bubbles. Instead, a plausible mechanism can be ascribed to the non-coplanar spin structure with a large out-of-plane ferromagnetic component in $NdMn_2Ge_2$. As discussed previously, between T_{SR} and T_{icm} $NdMn_2Ge_2$ exhibits a

conical spin structure while between T_{icm} and T_c it exhibits a canted antiferromagnetic spin structure in the absence of magnetic field with a net ferromagnetic moment having components both in the ab plane and along the c -axis. This suggests a delicate energetic competition between the canted antiferromagnetic phase and the incommensurate conical phase. As a result, an applied magnetic field along the c -axis may tip the energetic balance and give rise to the non-coplanar spin structure between T_{icm} and T_c , which accounts for the non-vanishing THE and TNE observed. It is desirable to probe the field-induced spin structure using neutron diffraction measurements.

To further quantify the ANE in NdMn_2Ge_2 , we calculate the anomalous transverse thermoelectric conductivity α_{xy}^A . Using the measured components of σ_{xx} , σ_{xy}^A , S_{xx} and S_{xy}^A , we determine α_{xy}^A in terms of $\alpha_{xy}^A = \sigma_{xx}S_{xy}^A + \sigma_{xy}^AS_{xx}$ [27,28], where σ_{xx} and σ_{xy}^A are calculated as $\sigma_{xx} = \frac{\rho_{xx}}{\rho_{xx}^2 + \rho_{yx}^2}$ and $\sigma_{xy}^A = \frac{\rho_{yx}^A}{\rho_{xx}^2 + \rho_{yx}^2}$, respectively. Note that these transverse thermoelectric and electronic conductivity tensors are directly associated with the Berry curvature Ω^Z , with σ_{xy}^A being proportional to the integration of Ω^Z of the whole Fermi Sea while α_{xy}^A being proportional to the integrated Ω^Z in the momentum space near the Fermi surface [18,29]. Figure 4 presents the temperature dependence of $\sigma_{xy}^AS_{xx}$, $\sigma_{xx}S_{xy}^A$, and α_{xy}^A extracted at 1.8 T, the magnitude of which all increase with decreasing temperature within the measurement temperature range. Note that the first two terms have an opposite sign, and such a feature is similar to that observed in other topological semimetals, such as Mn_3Ge [18] and Fe_3Sn_2 [26]. The maximum value (at $T = 160$ K) of $|\alpha_{xy}^A|$ reaches $\sim$

$0.6 \text{ Am}^{-1}\text{K}^{-1}$ which is comparable with results reported in Mn_3Ge and Fe_3Sn_2 with $\alpha_{xy}^A \sim 1.0 \text{ Am}^{-1}\text{K}^{-1}$ [18,26]. By simply fitting ρ_{xy} to ρ_{xx} (i.e., $\rho_{xy} \sim \rho_{xx}$), it was previously argued that the AHE between 150 K and 250 K is driven by skew scattering of the charge carriers [23]. Note that skew scattering tends to be the dominant factor in highly conductive metals whose resistivity is smaller than $1 \mu\Omega \text{ cm}$ [29]. However, as shown in Fig. 1 (c) (also see Ref. [23]), within this temperature range the resistivity of NdMn_2Ge_2 is $100 \sim 200 \mu\Omega \text{ cm}$, suggesting that NdMn_2Ge_2 is not a highly conductive metal, which implies that skew scattering is not dominant. Furthermore, instead of linearly fitting ρ_{yx} to ρ_{xx} between 150 K and 250 K, as shown in Fig. S4 [25], we find that our own ρ_{yx}/M (instead of just ρ_{yx}) data can be nicely fitted with both linear and quadratic terms of ρ_{xx} between 160 K and 320 K, i.e., $\rho_{xy} \sim a(M)\rho_{xx} + b(M)\rho_{xx}^2$, assuming that both $a(M)$ and $b(M)$ terms are proportional to M [29]. This suggests the contributions from both skew scattering and intrinsic contribution associated with the electronic structure to the AHE. The *ab initio* calculations of electronic band structure are desirable to further understand the origin of the anomalous Hall and anomalous thermoelectric conductivity in this system.

Conclusion

In summary, we have synthesized NdMn_2Ge_2 single crystals and performed neutron diffraction, electronic and thermoelectric transport studies. We show that NdMn_2Ge_2 exhibits large topological Nernst effect and anomalous Nernst effect, with the former feature driven by the non-coplanar spin structure and the latter feature

related to the combined effects of skew scattering and Berry curvature of electronic bands. These results suggest NdMn_2Ge_2 be a rare candidate which possesses topological properties associated with Berry phases in both real space and momentum space.

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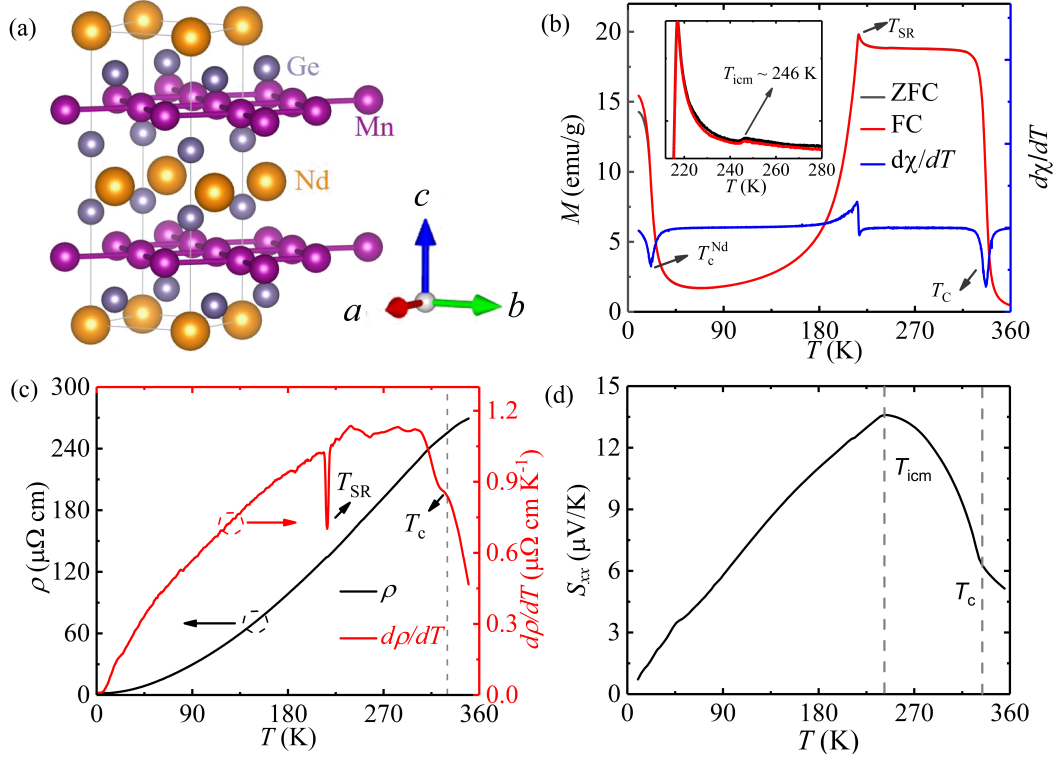


Figure 1: (a) The schematic crystal structure of NdMn₂Ge₂; (b) Temperature dependence of magnetic moment measured under field-cooling (FC) and zero-field cooling (ZFC) conditions; Inset shows an expanded view near T_{icm} . (c) Temperature dependence of longitudinal resistivity ρ_{xx} (left axis) and its derivative $\frac{d\rho_{xx}}{dT}$ (right axis); (d) Temperature dependence of Seebeck coefficient S_{xx} .

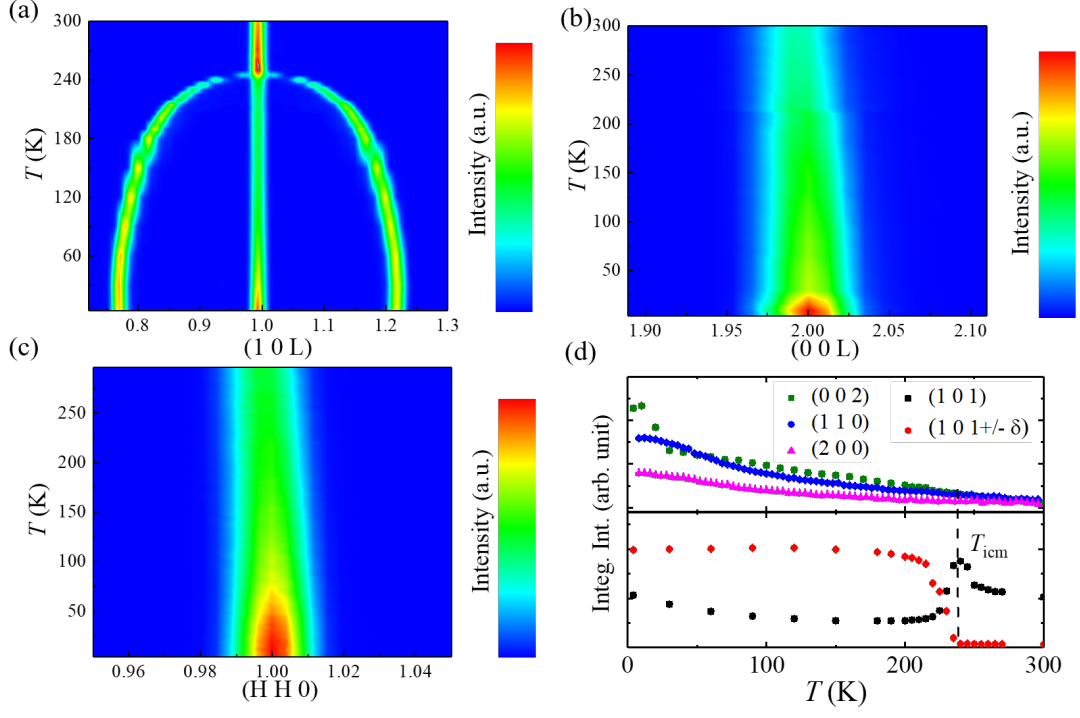


Figure 2: (a-c) False-color contour maps of T - $[1\ 0\ L]$, T - $[0\ 0\ L]$, and T - $[H\ H\ 0]$ measured at 0 T; (d) Temperature dependence of integrated neutron scattering intensity of $(0\ 0\ 2)$, $(1\ 1\ 0)$, $(2\ 0\ 0)$, and $(1\ 0\ 1)$, and $(1\ 0\ 1 \pm \delta)$ Bragg reflections.

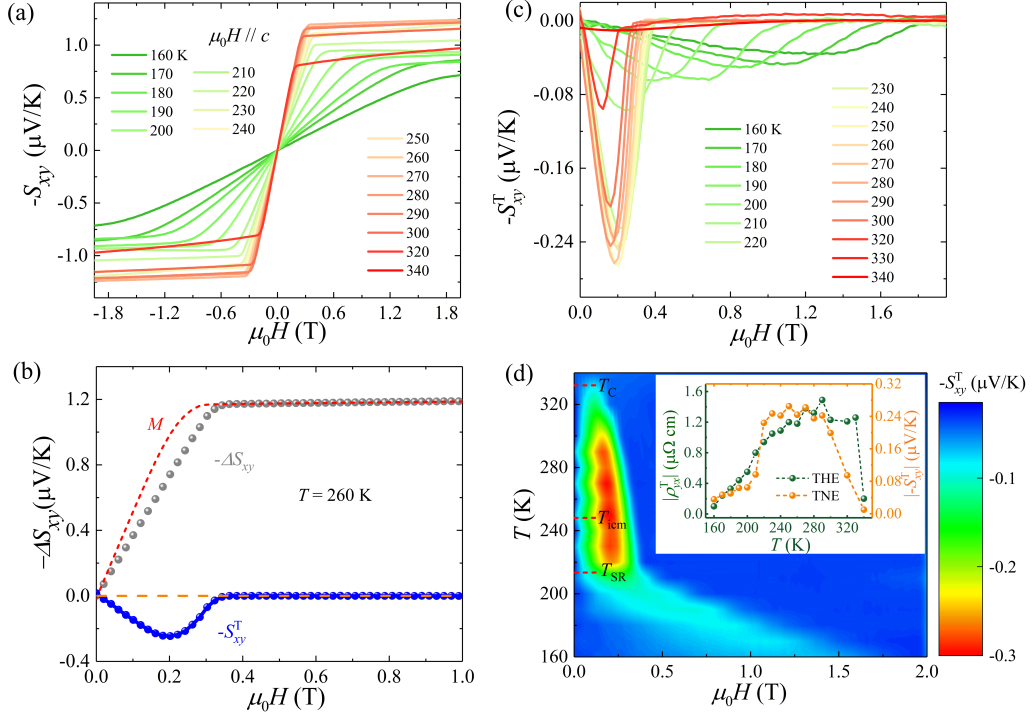


Figure 3: (a) Magnetic field dependence of Nernst thermopower $-S_{xy}$ measured at various temperatures; (b) Magnetic field dependence of magnetic moment M , $-\Delta S_{xy}$, and the extracted topological Nernst $-S_{xy}^T$ at 260. (c) Magnetic field dependence of $-S_{xy}^T$ measured at different temperatures. Each curve is obtained using the method as described in panel (b); (d) The T - H contour map of $-S_{xy}^T$. The transition temperatures were denoted using dashed red lines. The inset is the temperature dependence of maximum values of $|\rho_{yx}^T|$ (left axis) and $|-S_{xy}^T|$ (right axis).

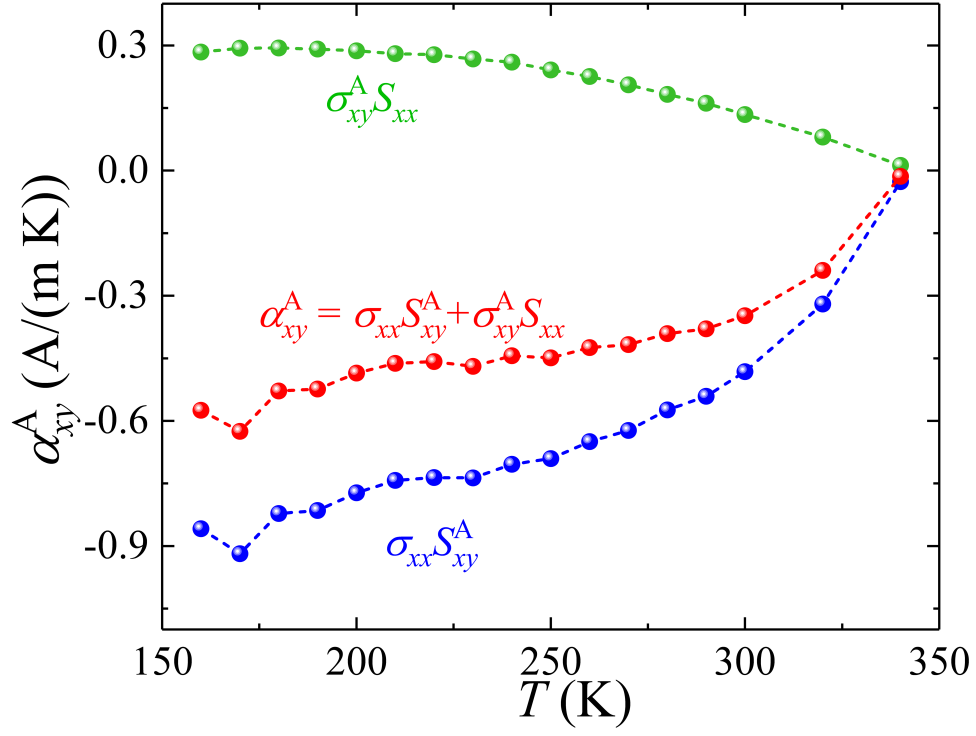


Figure 4: The temperature dependence of total anomalous transverse thermoelectric conductivity α_{xy}^A and its two components $\sigma_{xx} S_{xy}^A$ and $\sigma_{xy}^A S_{xx}$.

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