Large Room-Temperature Pure Topological Hall Effect (THE) in Kagome Antiferromagnet Mn$_3$Sn, and Induced Giant Low-Temperature THE with Fe Doping

Achintya Low, Susanta Ghosh, and Setti Thirupathaiah

Department of Condensed Matter Physics and Material Sciences, S. N. Bose National Centre for Basic Sciences, Kolkata-700106

Mn$_3$Sn is a fascinating magnetic topological system, showing topological characteristics within the Kagome lattice network due to the non-vanishing Berry phase in the momentum space. In this study, for the first time, we show a large pure room-temperature topological Hall effect (THE) in the $xy$-plane (0001), while the anomalous Hall effect (AHE) has been noticed in the $zx$-plane (0110) of Mn$_3$Sn. With Fe doping, we can induce a giant $xy$-plane THE in addition to AHE at low temperatures, while still preserving the pure room-temperature THE in Mn$_{2.8}$Fe$_{0.2}$Sn. Moreover, the AHE in the $zx$-plane has been increased with Fe doping. Our studies indicate that the topological properties are highly anisotropic in these systems. Most importantly, the large room-temperature pure THE observed in Mn$_3$Sn is quite promising for the realization of room-temperature topotronic-based applications.

I. INTRODUCTION

The Hall effect has been one of the pioneering discoveries in condensed matter physics [1], gained a lot of attentiveness both from the fundamental and technological applications [2, 3]. Specifically, in the fundamental science, the Hall effect branched into various exotic phenomena such as the spin Hall effect (SHE) observed without external magnetic fields [4], the quantum Hall effect (QHE) observed in two-dimensional electron gas systems where the Hall resistance is quantized [5, 6], the anomalous Hall effect (AHE) that is mainly observed in the ferromagnetic systems which scales with the magnetization property [7], and the recently emerging topological Hall effect that is mostly observed in magnetic topological metals having non-zero Berry phase in real-space [8]. Magnetic topological metals are a special class of topological systems that originated from the interplay between electronic and magnetic phases of the matter leading to a skyrmionic lattice [12, 13]. In general, the bulk topological systems are mainly classified into two types based on the attributes of band crossing points proximity to the Fermi level. They are, i) the Dirac semimetals and ii) the Weyl semimetals [14]. In Dirac semimetals, the band crossing point or the Dirac point is protected by both the time-reversal and inversion symmetries [15, 16]. Whereas in Weyl semimetals, the Weyl point is protected either by the time-reversal or inversion symmetry. Thus, a Weyl point can be tuned from the Dirac point by breaking one of the two symmetries. Several Weyl systems with inversion symmetry breaking have been studied [17–19], while a very few Weyl systems are known till date with the broken time-reversal symmetry (TRS) [20]. Co$_3$Sn$_2$S$_2$ [21, 22], Fe$_3$Sn$_2$ [23], EuCd$_3$As$_2$ [24] and Mn$_3$X (X=Sn, Ge, and Ga) are some of the known magnetic Weyl systems belonging to this family of broken TRS [25].

Out of these, specially, Mn$_3$X systems are very interesting topological metals as they show large anomalous Hall effect, despite having antiferromagnetic ordering, originated from the momentum-space Berry curvature [26, 27]. They further show frustrated magnetism due to distorted kagome lattice, leading to spin-glass phase at low temperatures [28]. In addition, flat-band electronic structure near the Fermi level has been observed due to strong electronic correlations [27]. Though there exist several inter-metallic kagome lattice systems [29, 30], the antiferromagnetism with noncollinear magnetic structure together with topological quantum electronic states make Mn$_3$X promising materials for potential technological applications in spintronics and quantum computations [33, 34].

Mn$_3$X has the hexagonal crystal structure with P6$_3$/mmc space group and Mn atoms arranged in a slightly distorted kagome lattice on the $xy$-plane [36]. At room temperature, Mn$_3$X shows noncollinear antiferromagnetic ordering with 120$^0$ inverse triangular spin structure due to Dzyaloshinskii–Moriya interaction (DMI) driven by the strong spin-orbit coupling [37, 40]. Importantly, Mn$_3$X systems show non-vanishing $k$-space Berry phase at room temperature originated from the triangular coplanar spin structure, leading to a large anomalous Hall effect [26, 27, 41, 42]. Interestingly, in the case of Mn$_3$Sn, the triangular coplanar magnetic order reshapes into a spiral noncoplanar magnetic ordering with a finite net magnetization along the $c$-axis at a critical spin-reorientation transition temperature ($T_{SR}$) [37, 40] which is not found in Mn$_3$Ge [43]. As a result, the AHE suppresses below $T_{SR}$ in Mn$_3$Sn but not in Mn$_3$Ge [44].

In this work, we focus on the Hall resistivity of Mn$_3$Sn and Fe doped Mn$_{2.8}$Fe$_{0.2}$Sn single crystals to understand the topological properties changes with Fe doping. Our studies reveal anisotropic anomalous Hall resistivity between $xy$-plane ($\rho_{xy}$) and $zx$-plane ($\rho_{zx}$) in Mn$_3$Sn. We also observe a large pure room-temperature topological...
Hall effect with $\rho_{xy} \approx 2 \mu \Omega \cdot \text{cm}$, in Mn$_3$Sn that is not reported so far on any magnetic topological system to date. This value increases further to 2.3 $\mu \Omega \cdot \text{cm}$ as the temperature is decreased to 260 K, but abruptly disappears below 260 K. Anomalous Hall effect has not been detected on the $xy$-plane of Mn$_3$Sn down to the lowest possible measured temperature of 2 K. On the other hand, in agreement with the previous reports [20, 27, 11, 22], we observe a large anomalous Hall effect in the $xz$-plane at room temperature, which is nearly constant down to 260 K and then disappears below 260 K. However, we did not observe THE in the $xx$-plane at any measured temperature. With Fe doping, we uncovered that Fe induces a large $xy$-plane AHE and THE at low temperatures without affecting much the high temperature THE. We further notice a large $zz$-plane AHE at low temperatures induced by Fe doping that increases gradually from 320 K to 125 K and then decreases with decreasing temperature before it saturates below 40 K. We find that Mn$_2$Sn is metallic and show isotropic electrical resistivity between the $c$-axis and the $a$-axis, while Mn$_2.8$Fe$_{0.2}$Sn is metallic in the $c$-axis but is a bad-metal along the $a$-axis. Thus, Mn$_2.8$Fe$_{0.2}$Sn shows anisotropic electrical transport between $a$-axis and $c$-axis. Nevertheless, we find both systems show spin-glass-like magnetic structure below 40 K. But from the magnetization dynamics data, we find that Mn$_2.8$Fe$_{0.2}$Sn shows a relatively low relaxation rate compared to Mn$_3$Sn, suggesting a reduced glassy spin nature with Fe doping.

II. EXPERIMENTAL DETAILS

Single crystals of Mn$_3$Sn were prepared by the self-flux method as reported in [11]. In this method, Manganese (Alfa Aesar 99.995%) and Tin powders (Alfa Aesar 99.998%) were taken in the 7 : 3 ratio and then were mixed thoroughly before sealing in a preheated quartz ampoule under partial Argon pressure. The powder mixture was then heated up to 1273 K for 24 hours and slowly cooled to 1173 K. Then the ampoule was taken out of the furnace and let cool to room temperature. In this way, we obtained several shiny hexagonal-rod-shaped single crystals with typical dimensions of $2 \times 1 \times 1$ mm$^3$. For the growth of Fe doped single crystals, we followed the same method by adding the desired amount of iron powder (Alfa Aesar, 99.99%) to the Mn-Sn mixture.

Powder X-ray diffraction (XRD) was performed using Rigaku SmartLab 9kW Cu K$_\alpha$ X-ray source. Stoichiometry of the crystals were found to be Mn$_4$Fe$_{0.13}$Sn$_{1.05}$ and Mn$_2.76$Fe$_{0.21}$Sn$_{1.03}$ using Energy Dispersive X-ray Spectroscopy (EDXS). From the EDXS data, we can observe that both compounds have marginally higher Sn concentrations. For simplicity, we denote the compositions as Mn$_3$Sn and Mn$_2.8$Fe$_{0.2}$Sn. Electrical transport and Hall effect measurements were performed using the four-probe technique. Copper leads were attached to the sample using EPO-TEK H21D silver epoxy.

Figure 1. (a) Powder XRD data taken from the crushed single crystals of Mn$_3$Sn, single crystal of Mn$_3$Sn, and Mn$_2.8$Fe$_{0.2}$Sn. Inset in (a) shows photographic image of Mn$_3$Sn and Mn$_2.8$Fe$_{0.2}$Sn single crystals. Top panel in (b) shows primitive unit cell of the hexagonal crystal structure of Mn$_3$Sn and bottom panel in (b) defines the crystal planes, $xx$-plane (0110) and $xy$-plane (001) on the hexagonal unit cell. (c) Resistivity measured along the $a$-axis, $\rho_{xx}$, as a function of temperature is plotted for both Mn$_3$Sn and Mn$_2.8$Fe$_{0.2}$Sn. (d) Resistivity measured along the $c$-axis, $\rho_{zz}$, as a function of temperature is plotted for both Mn$_3$Sn and Mn$_2.8$Fe$_{0.2}$Sn. Right-side inset in (d) shows schematic representation of the spin-reorientation above and below the transition temperature of 260 K for Mn$_3$Sn and 125 K for Mn$_2.8$Fe$_{0.2}$Sn. Left-side inset in (d) shows thermal hysteresis of the resistivity, $\rho_{zz}$ ($T$), between heating and cooling cycles of data collection on Mn$_3$Sn. (e) First derivative of $\rho_{zz}$ with respect to the temperature.
Temperature-dependent resistivity data were recorded from physical properties measurement system (PPMS, Dynacool, Quantum Design) within the temperature range of 2-350 K. Hall measurements were done by sweeping magnetic fields between -4 T and 4 T. Magnetic properties studies were done using the vibrating sample magnetometer (VSM) option of PPMS (Dynacool, Quantum Design) within the temperature range of 2-350 K and magnetic field was varied between -4 T and 4 T.

III. RESULTS AND DISCUSSIONS

A. Structural and Electrical Transport Properties

The powder XRD pattern of the crushed single crystals shown in the bottom panel of figure 1(a) confirms that Mn₃Sn crystalizes into hexagonal phase with a space group of P6₃/mmc (194). No impurity peaks have been detected from the XRD pattern, which suggests pure-phase Mn₃Sn single crystals. From the XRD pattern refinement of Mn₃Sn we obtain lattice parameters a=b=5.679(2) Å and c=4.533(4) Å which are comparable to the reported values of Mn₃Sn [41, 45]. The XRD patterns have taken on the single crystals of Mn₃Sn and Mn₂.₈Fe₀.₂Sn show multiple reflections corresponding to (0002) and (0004) planes.

Electrical resistivity measured along the a-axis ($\rho_{xx}$) plotted as a function of temperature in Fig. 1(c) for both Mn₃Sn and Mn₂.₈Fe₀.₂Sn. From Fig. 1(c) we can observe that the $\rho_{xx}$ resistivity of the parent compound shows a metallic nature, and interestingly for the first time, we find a hump-like structure at around 260 K [shown in the inset of Fig. 1(c)], whereas, in the case of Fe doped Mn₂.₈Fe₀.₂Sn, the resistivity decreases with increasing temperature which is a kind of bad-metallic behaviour [45]. Similarly, electrical resistivity measured along the c-axis ($\rho_{zz}$) plotted as a function of temperature in Fig. 1(d) for both Mn₃Sn and Mn₂.₈Fe₀.₂Sn. As can be seen from the $\rho_{zz}$ resistivity data, both parent and Fe doped systems show metallic behavior at low temperatures except for a significant increase in the impurity resistivity from 0.3 mΩ-cm to 0.63 mΩ-cm with Fe doping. Further, we observe a kink at around 260 K in Mn₃Sn which has been ascribed earlier to the spin structure reorientation of Mn atoms from a high-temperature noncollinear inverse triangular structure to a low-temperature noncoplanar spiral structure [31, 32]. But with Fe doping, we find that the spin reorientation transition temperature decreased to 125 K at which the kink has been observed from the $\rho_{zz}$ resistivity of Mn₂.₈Fe₀.₂Sn as shown in Fig. 1(d). Inset in Fig. 1(d) shows thermal hysteresis in the $\rho_{zz}$ resistivity of Mn₃Sn taken around 260 K between heating and cooling cycles. Thermal hysteresis in the $\rho_{zz}$ resistivity is consistent with the previous report on Mn₃Sn except that it was found at 270 K [42] and 275 K [41]. Such a thermal hysteresis is attributed to the first-order type magnetic transition from triangular to spiral structure in this system [41].

On the other hand, we do not observe such a thermal hysteresis in the $\rho_{zz}$ resistivity of Mn₂.₈Fe₀.₂Sn despite having the magnetic transition at around 125 K.

To precisely identify the spin-reorientation transition temperature ($T_{SR}$), we plotted $d\rho_{zz}/dT$ as a function of temperature as shown in Fig. 1(e). From the first derivative, we can reaffirm $T_{SR}$=260 K for Mn₃Sn and 125 K for Mn₂.₈Fe₀.₂Sn. In addition, we also observe a decrease in $d\rho_{zz}/dT$ with increasing $T$ for both systems below 10 K possibly due to weak local potentials at low temperatures [47] and above 143 K due to an electronic phase transition. Eventually, $d\rho_{zz}/dT$ becomes zero at 265 K and beyond this temperature, it is negative for Mn₂.₈Fe₀.₂Sn. That means, Mn₂.₈Fe₀.₂Sn shows a metal-insulator (MI) transition at around 265 K. On the other hand, in Mn₃Sn, the MI transition seems to be happening at much-elevated temperatures as $d\rho_{zz}/dT$ approaches
Figure 3. Magnetization isotherms, M(H), of Mn$_3$Sn for field applied parallel (a) and perpendicular (b) to the c-axis measured at various temperatures. Similarly, magnetization isotherms of Mn$_2$Fe$_{0.2}$Sn for field applied parallel (c) and perpendicular (d) to the c-axis measured at various temperatures. (e) and (f) show magnetization relaxation data plotted as a function of time, measured at 5 K with an external field of 500 Oe applied parallel and perpendicular to the c-axis from Mn$_3$Sn and Mn$_2$Fe$_{0.2}$Sn, respectively.

zero at around 345 K [48]. To emphasize here, Mn$_3$Sn shows nearly isotropic resistivity between a and c axes, while Mn$_2$Fe$_{0.2}$Sn shows a large resistivity anisotropy.

**B. Magnetic Properties**

Magnetization as a function of temperature M(T), plotted for zero-field cooling (ZFC) and field cooling (FC) modes with an external magnetic field of 500 Oe applied parallel and perpendicular to c-axis is shown in Fig. 2(a) for Mn$_3$Sn and in Fig. 2(b) for Mn$_2$Fe$_{0.2}$Sn. When the field is applied perpendicular to c-axis (H ⊥ c), in Mn$_3$Sn, a huge drop in magnetization is noticed at 260 K for both FC and ZFC modes, while the magnetization drop still present for the field applied parallel to c-axis (H ∥ c) but relatively small compared to H ⊥ c. The magnetization drop at 260 K for H ⊥ c can be understood as a result of spin-reorientation from inverse-triangular to spin-spiral structure [37–40]. Further reduction in temperature leads to spin-glass transition at 125 K due to the spin-reorientation that is found at 260 K in the parent system. In addition, below 40 K, we find a rapid decrease in magnetization from the ZFC and almost saturated magnetization from the FC data. This is due to the spin-glass-like transition as also observed in Mn$_3$Sn. Next, for the magnetic field applied parallel to c-axis, we do not see any significant spin-reorientation transition-like feature but we do find a fer-
romagnetic transition almost at the same temperature of 240 K similar to $H \perp c$. Also, the spin-glass-like transition occurs almost at the same temperature of 40 K similar to $H \perp c$. Overall, on comparing the magnetization $M(T)$ data between Mn$_3$Sn and Mn$_{2.8}$Fe$_{0.2}$Sn, we find that Fe doping enhances the magnetization in Mn$_3$Sn, while it does not affect the low-temperature spin-glass transition temperature. Perhaps, higher Fe doping concentration may increase the spin-glass transition temperature [52].

For a better understanding of the magnetism of these systems, we performed isothermal magnetization $M(H)$ measurements as shown in figure 3. Figures 3(a) and 3(b) depict $M(H)$ isotherms from Mn$_3$Sn for $H \parallel c$ and $H \perp c$, respectively. From Fig. 3(a), we see significant magnetic hysteresis in Mn$_3$Sn for $H \parallel c$ at 2 K without saturation even at an applied field of 4 T. But at higher temperatures, the magnetic hysteresis disappears. The observed magnetic hysteresis at 2 K is possibly due to short-range magnetic ordering due to spin fluctuations in the spin-glass state. As shown in Fig. 3(b), for $H \perp c$ also the isotherms $M(H)$ are found to be similar to $H \parallel c$ with significant magnetic hysteresis at 2 K again due to glassy spin nature. In contrast, we find a weak ferromagnetic-like hysteresis with large coercivity and remanence at 300 K. Moreover, the slope $dM/dH$ obtained at higher fields ($>3$ T) decreases with increasing temperature, hinting at reduced antiferromagnetism in Mn$_3$Sn at higher temperatures. Therefore, we conclude that the observed magnetic hysteresis at low temperatures for both $H \perp c$ and $H \parallel c$ in Mn$_3$Sn is due to the short-range magnetic interactions below $<40$ K. The anisotropic magnetization between $H \perp c$ and $H \parallel c$ in Mn$_3$Sn is in good agreement with previous magnetic studies on Mn$_3$Sn [45].

Figures 3(c) and 3(d) depict $M(H)$ isotherms measured on Mn$_{2.8}$Fe$_{0.2}$Sn at various temperatures for both $H \parallel c$ and $H \perp c$, respectively. From Fig. 3(c), for $H \parallel c$, we can see that the Fe doping induces a long-range ferromagnetic ordering as the magnetic hysteresis is found with increased coercivity at 2 K. But the magnetic hysteresis disappears at temperatures of 100 and 150 K, while still having a sigmoid-like $M(H)$ curve. On the other hand, at the temperatures of 260 and 300 K, we observe that the system completely transforms into antiferromagnet as we find linear $M(H)$ isotherms. Next from Fig. 3(d), for $H \perp c$, we observe $M(H)$ isotherm with a coercivity of 850 Oe at 2 K. Interestingly, we also notice a field-induced asymmetric $M(H)$ curve at 2 K that is not visible for $H \parallel c$. We further observe reduced coercivity of 220 Oe at 260 K, which is nearly constant up to 300 K without any field-induced asymmetry in the $M(H)$ loop. Note here that the coercivity of Mn$_{2.8}$Fe$_{0.2}$Sn is almost a factor of 2 less compared to the parent system for $H \perp c$ at 300 K. Whereas, the coercivity of Mn$_{2.8}$Fe$_{0.2}$Sn is nearly 2 times higher compared to Mn$_3$Sn at 2 K for $H \parallel c$.

The glassy-spin nature of the systems is further examined by performing magnetization relaxation measurements for both Mn$_3$Sn and Mn$_{2.8}$Fe$_{0.2}$Sn as shown Figs. 3(e) and 3(f), respectively, measured with an applied magnetic field of 500 Oe in the FC mode at 5 K. Magnetization relaxation for a spin-glass system can be explained using the Stretched function, $M(t) = A \exp[-(\frac{t}{\tau})^\alpha]$ [54]. Here, $A$ is an exponential factor, $t$ is the time, $\tau$ is a characteristic relaxation time constant, and $\alpha$ is stretching exponent which is temperature dependent and can take the values $0 < \alpha < 1$ [55]. From the Stretched function fitting to Mn$_3$Sn [see Fig. 3(c)], we obtain $\alpha=0.96 \pm 0.04$ and $\tau=(1.03 \pm 0.17) \times 10^5$ s for $H \parallel c$. Similarly, $\alpha=0.85 \pm 0.1$ and $\tau=(1.83 \pm 0.77) \times 10^5$ s are obtained for H $\perp c$. The $\alpha$ values close to 1 and higher relaxation time constants suggest Mn$_3$Sn to be nearly an isotropic spin-glass system. On the other hand, the best fit for the magnetization relaxation curves from Mn$_{2.8}$Fe$_{0.2}$Sn is obtained using the modified Stretched function $M(t) = M_0 + A \exp[-(\frac{t}{\tau})^\alpha]$ [56, 58]. Here, the additional term $M_0$ is the magnetization due to long-range ferromagnetic ordering at $t=\infty$ [59]. From the fitting [see Fig. 3(f)] we obtained $M_0=0.21 \pm 0.1$ emu/g, $\tau=5.67 \pm 512$ s and $\alpha=0.47 \pm 0.01$ for $H \parallel c$. Similarly, we obtained $M_0=0.14 \pm 0.01$ emu/g, $\tau=7.16 \pm 31.9$ s and $\alpha=0.76 \pm 0.02$ for H $\perp c$. Here, different $\alpha$ values (<1) for different crystal orientations found in Mn$_{2.8}$Fe$_{0.2}$Sn suggest for anisotropic ferromagnetism induced with Fe doping, which is consistent with the anisotropic magnetization isotherms, M ($H$), shown in Figs. 3(c) and 3(d).
C. Anomalous and Topological Hall Effects

Fig. 4(a) depicts Hall resistivity in the $xy$-plane ($\rho_{xy}$) and in the $zx$-plane ($\rho_{zx}$) plotted as a function of temperature from Mn$_3$Sn measured with a magnetic field of 1 T applied parallel and perpendicular to the $c$-axis. From Fig. 4(a), we notice an increase in $\rho_{zx}$ from 320 K down to 260 K, which then instantaneously become nearly zero below 260 K. This sudden decrease in $\rho_{zx}$ at 260 K coincides with the spin-reorientation transition temperature of the studied sample. The observation of drastic changes in the $\rho_{zx}$ Hall resistivity at 260 K is in good agreement with previous studies on a similar system except for a slightly higher transition temperature of 270 K $\pm$ 2 and 275 K $\pm$ 1. On the other hand, we observe no significant change in $\rho_{xy}$ Hall resistivity from 320 K down to 2 K, which is nearly zero all the time. Next, Fig. 4(b) depicts $\rho_{xy}$ and $\rho_{zx}$ Hall resistivity plotted as a function of temperature for Mn$_2$S$_{8-x}$Fe$_x$S$_{32}$ measured with the magnetic field of 1 T applied parallel and perpendicular to the $c$-axis. From Fig. 4(b), we notice an increase in $\rho_{xy}$ from 320 K down to 125 K, which gradually decreases with temperature from 125 K down to 40 K, and then saturates below 40 K. The decrease in $\rho_{xy}$ coincides with the spin-reorientation transition temperature of 125 K as observed from the magnetization measurements on the doped sample [see Fig. 2(b) of H || c]. On the other hand, the $\rho_{xy}$ Hall resistivity slowly increases between 320 to 125 K and then exponentially increases from 125 K down to 40 K, and then tends to saturate below 40 K. Interestingly, we notice an extremely large $\rho_{xy}$ value of 31 $\mu$Ω-cm under 1 T magnetic field at 2 K which is found to be negligible in the parent system.

Fig. 5(a) depicts $\rho_{xy}$ Hall resistivity plotted as a function of the applied magnetic field from Mn$_3$Sn for H || c. From Fig. 5(a), we can realize that at low temperatures (T < 260 K) the Hall resistivity does not change much with the field and is always close to zero between -4 T and 4 T. However, at the sample temperature of 260 K, we observe a non-zero Hall resistivity as high as 2.3 $\mu$Ω-cm at a critical field of 0.3 T which then slightly reduced to 2 $\mu$Ω-cm at 300 K. Similarly, Fig. 5(b) depicts $\rho_{zx}$ Hall resistivity plotted as a function of magnetic field from Mn$_3$Sn for H || c. From Fig. 5(b), we can realize that for temperatures < 260 K the $\rho_{zx}$ Hall resistivity does not change much with the magnetic field and is always close to zero between -4 T and 4 T. However, for the sample temperatures of ≥ 260 K, we observe an anomalous Hall resistivity as high as 6.5 $\mu$Ω-cm whose sign depends on the direction of the applied magnetic field [42, 60].

Fig. 5(c) depicts $\rho_{xy}$ Hall resistivity measured from Mn$_2$S$_{8-x}$Fe$_x$S$_{32}$ for H || c. Interestingly, unlike in Mn$_3$Sn in which no significant Hall resistivity was found at low temperatures, we observe a gigantic $\rho_{xy}$ anomalous Hall resistivity in Mn$_2$S$_{8-x}$Fe$_x$S$_{32}$ as high as 36 $\mu$Ω-cm at 2 K with an applied field of 4 T. We further observe hysteresis in $\rho_{xy}(H)$ data at 2 and 5 K, which is consistent with magnetization data of M(H) as we find a large magnetic hysteresis at 2 K [see Fig. 3(c)]. This suggests that the anomalous Hall resistivity observed in Mn$_2$S$_{8-x}$Fe$_x$S$_{32}$ is mainly originated by the ferromagnetism induced by the Fe doping. The anomalous $\rho_{xy}$ Hall resistivity gradually decreases with increasing temperature and the hysteresis disappears for the temperatures of ≥ 100 K. To our surprise, similar to the parent system, we observe non-zero $\rho_{xy}$ Hall resistivity as high as 2 $\mu$Ω-cm at temperatures ≥ 260 K in Mn$_2$S$_{8-x}$Fe$_x$S$_{32}$ at a critical field of 0.2 T which then reduces to 1.6 $\mu$Ω-cm at 300 K.

Fig. 5(d) depicts $\rho_{zx}$ Hall resistivity from Mn$_2$S$_{8-x}$Fe$_x$S$_{32}$ for H || c. From Fig. 5(d), we can see non-zero anomalous $\rho_{zx}$ Hall resistivity in Mn$_2$S$_{8-x}$Fe$_x$S$_{32}$ even at 2 K which increases with temperature. We find $\rho_{zx}$ $\approx$ 9.5 $\mu$Ω-cm at 150 K which then decreases with increasing temperature. We further notice a field-induced asymmetric hysteresis in the $\rho_{zx}$ (H) Hall resistivity at 2 and 5 K. This observation of asymmetric hysteresis in $\rho_{zx}$ (H) is consistent with the field-induced asymmetric M(H) isotherm measured at 2 K [see Fig. 3(d)].

Figs. 6(a) and 6(b) depicts $\rho_{xy}^T$ topological Hall resistivity from Mn$_3$Sn and Mn$_2$S$_{8-x}$Fe$_x$S$_{32}$, respectively extracted from the total Hall resistivity data shown in Figs. 5(a) and 5(c). Note here that the total Hall resistivity shown in Fig. 5 has contributions from normal Hall
resistivity ($\rho_N^H$) which varies linearly with the applied magnetic field ($\rho_N^H = R_0 \mu_0 H$), anomalous Hall resistivity ($\rho_A^H$) which depends on the sample magnetization ($\rho_A^H = S_A \rho^2 M$), and topological Hall resistivity ($\rho_T^H$). All these contributions lead to a total Hall resistivity as per the relation $\rho_H = \rho_N^H + \rho_A^H + \rho_T^H = R_0 \mu_0 H + S_A \rho^2 M + \rho_T^H$. Here $R_0$ is a normal Hall coefficient and $S_A$ is an anomalous Hall coefficient. Now, to extract the topological Hall resistivity one has to subtract the normal and anomalous Hall contributions from the total Hall resistivity, following a method that has been explained thoroughly in several earlier reports [27, 39, 61] [62]. In this way, we extracted $\rho_{xy}^T$ topological Hall resistivity from Mn$_3$Sn and Mn$_{2.8}$Fe$_{0.2}$Sn to be 2 $\mu$Ω-cm and 1.6 $\mu$Ω-cm, respectively at 300 K. While Mn$_3$Sn shows no topological Hall resistivity in the $xy$-plane below 260 K, we find Mn$_{2.8}$Fe$_{0.2}$Sn shows topological Hall resistivity as high as 4.5 $\mu$Ω-cm at 2 K, which gradually decreases with increasing temperature. But at 100 K, we notice that the sign of $\rho_{xy}^T$ switches from negative to positive for the magnetic fields and from positive to negative for the negative magnetic fields as shown in Fig. 6(b).

Fig. 6(c) shows the normal Hall coefficient ($R_0$) and the anomalous Hall coefficient ($S_A$) plotted as a function of temperature, extracted from the fittings of the data shown in Fig. 6(b). From Fig. 6(c), we can notice that the normal Hall coefficient is negative at low temperatures, suggesting for dominant electron carriers and at high temperatures $R_0$ becomes positive, suggesting for dominant hole carriers. The normal Hall coefficient sign switching is in-line with the sign change in the topological Hall resistivity shown in Fig. 6(b). Further, we observe that the anomalous Hall coefficient is 0.18 V$^{-1}$ at 2 K which gradually decreases with increasing temperature to 0.01 V$^{-1}$ at 150 K and becomes temperature independent after that. Note here that we did not observe any noticeable $\rho_{xx}^T$ topological Hall resistivity from neither Mn$_3$Sn nor Mn$_{2.8}$Fe$_{0.2}$Sn despite observing a large anomalous $\rho_{xx}$ Hall resistivity [see Figs. 5(b) and 5(d)].

Overall, the large pure room-temperature topological Hall effect observed in Mn$_3$Sn is a novel experimental result of this study as to date a pure THE has not been observed on any system with $\rho_{xy}$ as high as 2 $\mu$Ω-cm at 300 K without dominant coexisting AHE. This means, so far existing reports on the Mn$_3$Sn single crystals demonstrated the room temperature THE within the range of $\rho_{xy} \approx 0.3 - 2.1$ $\mu$Ω-cm but always in the presence of AHE [12, 63, 64]. There also exist few systems other than Mn$_3$Sn showing large room-temperature THE but again with coexisting AHE. For instance, noncollinear ferromagnet LaMn$_2$Ge$_2$ shows topological Hall resistivity of 1 $\mu$Ω-cm along with a comparable anomalous Hall resistivity of 0.5 $\mu$Ω-cm at 300 K [65]. Frustrated kagome ferromagnet Fe$_2$S$_2$ shows topological Hall resistivity of 2 $\mu$Ω-cm but with a large anomalous Hall resistivity of 4.5 $\mu$Ω-cm at 300 K [66]. Similarly, there exists many chiral and skyrmion systems such as CrTe$_2$ [67], NiMnGa [68], and Mn$_2$PtSn [69] showing room-temperature topological Hall resistivity but always accompanies with a significant anomalous Hall resistivity. On the other hand, the skyrmion system Gd$_2$PdSi$_3$ shows a large topological Hall resistivity of 2.5 $\mu$Ω-cm without AHE signal but only at very low-temperatures (< 25 K) [70].

Interestingly, we notice a pure room-temperature topological Hall effect in Mn$_{2.8}$Fe$_{0.2}$Sn with $\rho_{xy} \approx 1.6$ $\mu$Ω-cm at 300 K for a critical field of 0.2 T, which then increases with decreasing temperature up to $\approx 4.5$ $\mu$Ω-cm at 2 K for a critical field of 1.7 T. Most importantly, we find THE with a large coexisting AHE at 2 and 5 K. On comparing our $\rho_{xy}$ Hall resistivity of Mn$_{2.8}$Fe$_{0.2}$Sn at 2 K [see Fig. 5(c)], we suggest that the AHE signal found in Ref. 62 could be originated from the excess Mn that is commonly present in these systems as we find that the Fe doping at the Mn site induces a large $xy$-plane anomalous Hall effect. Nevertheless, the absence of $xy$-plane AHE in our Mn$_3$Sn system further confirms that the studied sample has no excess Mn. Thus, to find pure THE in these systems, one should get the crystals without excess Mn. Finally, we attribute the room-temperature pure $xy$-plane THE signal found in Mn$_3$Sn and Mn$_{2.8}$Fe$_{0.2}$Sn to the real-space Berry phase due to the skyrmion lattice generated from the field-induced-do-
IV. SUMMARY

In summary, we studied the kagome antiferromagnet Mn$_3$Sn and Fe doped Mn$_2$Fe$_2$Sn single crystals to understand the influence of Fe doping on the electronic and magnetic properties. Our results reveal a nearly isotropic metallic nature of electrical resistivity in Mn$_3$Sn between $a$-axis and $c$-axis, while the Fe doping leads to a total anisotropic electrical resistivity in Mn$_3$Sn. From the magnetic property studies of M(T) on Mn$_3$Sn, we observe a sudden decrease in magnetization at the spin-reorientation transition temperature of 260 K. Although Fe doping decreases the spin-reorientation transition temperature to 125 K, it has no effect on the spin-glass transition temperature which was always found at around 40 K. We observe a large xy-plane topological Hall effect in Mn$_3$Sn at room temperature which disappears below 260 K. Though Fe doping shows no significant effect on the room temperature pure topological Hall resistivity, induces a giant xy-plane THE signal at low temperatures. In addition, we notice a giant anomalous Hall effect induced in the $xy$-plane with Fe doping, while a little change has been found on the $zx$-plane AHE with Fe doping. Our results suggest that the room-temperature THE observed in both Mn$_3$Sn and Mn$_2$Fe$_2$Sn is mainly originated from the domain wall-based skyrmion lattice, while the low-temperature THE observed from Mn$_2$Fe$_2$Sn is mostly due to the field-induced skyrmion lattice generated by the spin-canting with Fe doping.

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