Anomalous transport properties of the antiferromagnetic Weyl semimetals \( \text{Mn}_3X \) (\( X = \text{Sn, Ge} \))

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Abstract. Noncollinear antiferromagnets \( \text{Mn}_3X \) (\( X = \text{Sn, Ge} \)) are characterized by a large anomalous Hall effect originating from a large Berry curvature despite a vanishingly small magnetization. From recent first-principle theories, the large Berry curvature is predicted to be induced by a existence of Weyl nodes broken time-reversal symmetry. The large anomalous Nernst effect is also contributed by the magnetic Weyl state around the Fermi level \( E_F \), and likely shares its origin with the anomalous Hall effect. The thermoelectric transport \( S(T) \) and thermomagnetic transport \( S_{ji}(T) \) are thus investigated in single crystals of \( \text{Mn}_3X \). Here, \( \text{Mn}_3X \) exhibits a large anomalous Nernst effect; in particular, the signal magnitude of \( \text{Mn}_3\text{Ge} \) exceeds \( 1 \mu \text{V/K} \), which is 1.5 times that of \( \text{Mn}_3\text{Sn} \). The Weyl properties are discussed by analyzing the thermal conductivity, specific heat, and Seebeck and Nernst effects. We also evaluate the zero-field Nernst-driven thermoelectric figure of merit for device applications in the antiferromagnets \( \text{Mn}_3X \).

1. Introduction

The large anomalous Hall effect (AHE) observed in the noncollinear antiferromagnets (AFMs) \( \text{Mn}_3X \) (\( X = \text{Sn and Ge} \)) is known to be given by the intrinsic contribution due to large Berry curvature generated by time-reversal-breaking Weyl metals \cite{1, 2}. The existence of the magnetic Weyl nodes at Fermi level \( E_F \) are suggested by recent first-principles calculations of the electronic band structure \cite{3, 4, 5, 6}. Additionally, the planar Hall effect accompanied by positive longitudinal magnetoconductance has been observed recently in magnetotransport measurements. Such a phenomenon is considered to be induced by the chiral anomaly corroborated with the magnetic Weyl semimetal owing to the clear evidence of Weyl nodes, which is provided by angle-resolved photoemission spectroscopy and the corresponding theoretical band structure.

The sum of the Berry curvature over the occupied states below the \( E_F \) theoretically constitutes the intrinsic contribution of AHE. When the \( E_F \) are tuned just at the Weyl
nodes, an ideally maximized AHE appears. The theoretically calculated value of AHE agrees reasonably well with the experimental value [7]. The Berry curvature is also detected using the thermoelectric counterpart of the AHE, the so-called anomalous Nernst effect (ANE). In fact, the observed anomalous Nernst effect is stronger than the effect reported for ferromagnets in the antiferromagnets Mn$_3$X [7, 8]. When the finite Berry curvature at Weyl nodes appears only in the $k_BT$ energy region around $E_F$, the ANE is provided by the contribution of the Weyl nodes in the $k$-space because the ANE intrinsically determines the spectrum of the Berry curvature at $E_F$ [7]. The ANE thus provides us important properties on the Berry curvature and its relation to the Weyl nodes near $E_F$ [6, 7].

Here, a comprehensive study of the large spontaneous ANE are performed in the chiral antiferromagnets Mn$_3$X. In antiferromagnet Mn$_3$Sn, the inverse-triangular spin structure is only magnetically stable from the spin-glass transition temperature $T_s$ ($=\sim 50$ K) to the antiferromagnetic Néel temperature $T_N$ ($=\sim 430$ K). Although Mn$_3$Ge has the same type of spin structure, it is stable above 300 mK and has no additional transition below $=\sim 380$ K. The low-temperature Nernst effect $S_{ji}$ is affected by the spectrum of the Berry curvature at the effective $E_F$, and reveals a unique property of Weyl metals. It is thus essential to investigate the low-temperature ANE of Mn$_3$Ge in elucidating the magnetic Weyl phenomenon for future application to evaluate the thermodynamic properties of the Nernst-type modules as well as our previous studies of Mn$_3$Sn [9, 10].

2. Experimental and Results

Figures 1a and 1b present the specific heats $C_{ab}(T)$ and $C_{ab}(T)/T$ of Mn$_3$Ge (Mn$_3$Sn). The peak is observed at 65 K (70 K) for Mn$_3$Ge (Mn$_3$Sn). According to the Einstein–Debye equation of $C_{ab}/T = \gamma + \beta T^2$, where $\gamma$ and $\beta$ are respectively parameters of the electronic and lattice contributions to the specific heat, the best parameter fittings to the Mn$_3$Ge (Mn$_3$Sn) data are obtained in the low-temperature regime of 2–15 K as $\gamma = 22.5$ mJ/mol K$^2$ ($=\sim 32.5$ mJ/mol K$^2$) and $\beta = 156$ μJ/mol ($=\sim 300$ μJ/mol). Here, the Debye temperature is evaluated as $T_\theta = 291$ K ($T_\theta = 234$ K) using $C_{ab}/T = (12\pi^4N_k)/(5T_\theta^3)T^2$ and Boltzmann’s constant $N_k$ [5]. In Figure 1c, the positive Seebeck coefficient $S_{ji}$ is shown at 300 K and indicates that hole carriers are dominated by electron carriers [5, 10]. $S_{ji}(T)$ changes sign at $=\sim 100$ K and peaks at $=\sim 50$ K in both the Q||[2110] and Q||[0110] of Mn$_3$Ge. Additionally, we obtain from the estimated Debye temperatures a crossover temperature of $T_\theta/5 = 58.6$ and 46.8 K for the minimum Seebeck coefficient of Mn$_3$Ge and Mn$_3$Sn respectively. Figure 1d displays the zero-field thermal conductivity $\kappa(T)$ in the warming process after annealing at a temperature above $T_N$, which aligns the domain along $B$. The value $\kappa(300$ K) $=8–12$ W/Km of Mn$_3$Sn is slightly more conductive than that of Mn$_3$Ge. The spontaneous Nernst voltage can be applied to thermoelectric modules that convert only a temperature gradient into electrical energy and ordinary Seebeck-type at room temperature [11]. As shown in Fig. 2a, the temperature dependence of $S_{ji}$ is maximized at 100 K (200 K) for Mn$_3$Ge (Mn$_3$Sn). We find that the Nernst coefficient is largest in Mn$_3$Ge as the antiferromagnet. The Seebeck-driven thermoelectric figure of merit $ZT = (S^2/\rho\kappa)T$ is a quantity used to characterize the performance of a device and defined for a maximally efficient thermoelectric generator. Similarly, the Nernst-driven $Z_N T = (S_{ji}^2/\rho\kappa)T$ is defined in Mn$_3$Ge and is only $=\sim 9 \times 10^{-6}$ at 300 K, which indicates higher value compared with the value for Mn$_3$Sn ($=\sim 2 \times 10^{-6}$ at 300 K) reported in our previous study [10] and is still quite low for fabricating a Nernst-driven thermoelectric device.

3. Conclusion

A sizable Nernst coefficient $S_{ji}$ is obtained for antiferromagnetic Mn$_3$Ge, which exceeds 1 μV/K at around 100 K. This value is comparable to the Nernst coefficients of ordinal ferromagnets and twice that of Mn$_3$Sn. The ANE in Mn$_3$Ge is thus assumed to be induced by large
Figure 1. (a) Zero-field specific heat divided by temperature $C_{ab}(T)/T$ of Mn$_3$Ge (red) and Mn$_3$Sn (blue) in (a) the temperature range of 2–200 K and (b) the low-temperature range below 20 K. The calculated entropy $S(T)$ is plotted in the inset in (a). The dashed lines show the linear fitting of $C_{ab}/T = \gamma + \beta T^2$ to the low-temperature data in $C_{ab}/T$ vs $T^2$ of panel (b). (c) Zero-field Seebeck coefficients $S(T)$ of Mn$_3$Ge in three different configurations: $Q||[2\bar{1}\bar{1}0]$ (green), $Q||[01\bar{1}0]$ (blue), and $Q||[0001]$ (red). (d) Zero-field thermal conductivity $\kappa(T)$ for the three different configurations of Mn$_3$X ($X = \text{Sn, Ge}$): $Q||[2\bar{1}\bar{1}0]$ (green), $Q||[01\bar{1}0]$ (blue), and $Q||[0001]$ (red).

Berry curvature, indicating the existence of paired Weyl points. In both Mn$_3$Sn and Mn$_3$Ge, the AHE and ANE are considered to share the same origin. The large anomalous Nernst coefficient $S_{ji}$ for Mn$_3$Ge beyond the magnetization scaling relation are shown in Fig. 2b [8]. From a viewpoint of the ANE, however, the zero-field Nernst-driven thermopower of antiferromagnetic Weyl metals still does not have sufficient $ZT$ for practical application, in contrast with Seebeck-driven thermopower in semiconductors. In future practical applications, antiferromagnetic Weyl semimetals are expected to be available for topological memory besides Nernst-driven thermoelectric modules as discussed here. These Weyl semimetals are suitable for microfabricated devices owing to the weak magnetic leakage due to integration. We need to investigate further their possible use in spintronic applications [12, 13] and continue searching for the antiferromagnetic type of such novel materials.

acknowledgments
This work is partially supported by CREST (JPMJCR18T3), Japan Science and Technology Agency, by Grants-in-Aids for Scientific Research on Innovative Areas (15H05882 and 15H05883) from the Ministry of Education, Culture, Sports, Science, and Technology of Japan, and by Grants-in-Aid for Scientific Research (19H00650) from the Japanese Society for the Promotion
Figure 2. (a) Zero-field Nernst signals $S_{ji}(T)$ of Mn$_3$X (with open and closed symbols indicating data for Mn$_3$Sn and Mn$_3$Ge respectively). (b) Nernst coefficient $S_{ji}$ as a function of magnetization $M$ in Mn$_3$X and various ferromagnetic materials [7, 8]. The red-shaded area indicates the regions in which $S_{ij}(M)$ follows the linear scaling relation for conventional ferromagnets [7, 8].

of Science (JSPS). The work at IQM was supported by the US Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under grant DE-FG02-08ER46544. The work on the first-principles calculation was supported in part by JSPS Grant-in-Aid for Scientific Research on Innovative Areas (18H04481 and 19H05825) and by MEXT as a social and scientific priority issue (Creation of new functional devices and high-performance materials to support next-generation industries) to be tackled using the Post-K computer (hp180206 and hp190169). The use of the facilities of the Materials Design and Characterization Laboratory at the Institute for Solid State Physics, The University of Tokyo, is gratefully acknowledged.

References
[1] Nakatsuji S, Kiyohara N and Higo T 2015 Nature 527 212–215
[2] Kiyohara N, Tomita T and Nakatsuji S 2016 Phys. Rev. Applied 5(6) 064009
[3] Yang H, Sun Y, Zhang Y, Shi W J, Parkin S S P and Yan B 2017 New J. Phys. 19 015008
[4] Suzuki M T, Koretsune T, Ochi M and Arita R 2017 Phys. Rev. B 95(9) 094406
[5] Ziman J M 1960 Electrons and phonons: the theory of transport phenomena in solids (Oxford University Press)
[6] Kuroda K, Tomita T, Suzuki M T, Bareille C, Nugroho A A, Goswami P, Ochi M, Ikhlas M, Nakayama M, Akebi S, Noguchi R, Ishii R, Inami N, Ono K, Kumigashira H, Varykhalov A, Muro T, Koretsune T, Arita R, Shin S, Kondo T and Nakatsuji S 2017 Nature Materials 16 1090–1095
[7] Ikhlas M, Tomita T, Koretsune T, Suzuki M T, Nishio-Hamane D, Arita R, Otani Y and Nakatsuji S 2017 Nature Phys 13 1085–1090
[8] Chen T, Tomita T, Minami S, Fu M, Koretsune T, Kitatani M, Muhammad I, Nishio-Hamane D, Ishii R, Ishii F, Arita R and Nakatsuji S 2020 Nature Communications 12 572
[9] Sugii K, Imai Y, Shimozawa M, Ikhlas M, Kiyohara N, Tomita T, Suzuki M T, Koretsune T, Arita R, Nakatsuji S and Yamashita M arXiv preprint arXiv:1902.06601
[10] Tomita T, Ikhlas M and Nakatsuji S 2020 JPS Conference Proceedings 30 011009
[11] Snyder G J and Toberer E S 2008 Nature Materials 7 105–114
[12] Sugimoto S, Nakatani Y, Yamane Y, Ikhlas M, Kondou K, Kimata M, Tomita T, Nakatsuji S and Otani Y 2020 Communications Physics 3 111
[13] Miwa S, Iihama S, Nomoto T, Tomita T, Higo T, Ikhlas M, Sakamoto S, Otani Y, Mizukami S, Arita R and Nakatsuji S 2021 Small Science 1 2000062