

## **DISCLAIMER**

**This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof. Reference herein to any social initiative (including but not limited to Diversity, Equity, and Inclusion (DEI); Community Benefits Plans (CBP); Justice 40; etc.) is made by the Author independent of any current requirement by the United States Government and does not constitute or imply endorsement, recommendation, or support by the United States Government or any agency thereof.**

DOE award number: DE-SC0019259

Title: Topological magnons and topological electronic properties in metallic Kagome magnets

Name of receipt: Michigan State University

Principal Investigator: Xianglin Ke

## **Executive Summary:**

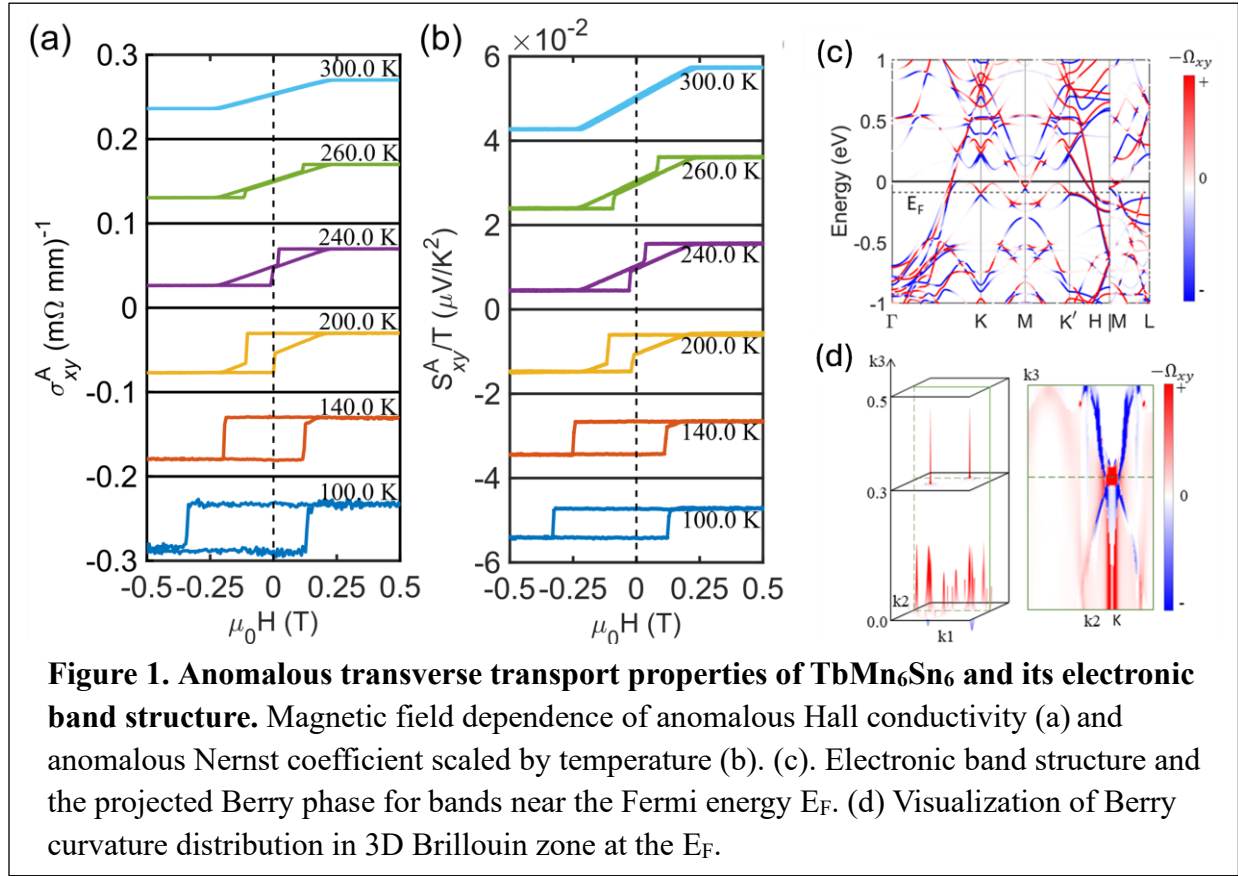
The primary goal of this project is to study metallic Kagome magnets that potentially host topological electronic properties and topological magnetic excitations, aiming to understand the effect of lattice geometry on topological character of magnons and electronic bands and to study the interplay between magnetism and topological electronic properties. Through this DOE-funded research program, we have unraveled novel electronic and thermoelectric properties of  $\text{RMn}_6\text{Sn}_6$  ( $\text{R} = \text{Y}, \text{Tb}, \text{and Er}$ ) and  $\text{Fe}_3\text{Ge}$  Kagome magnets, including anomalous Hall effect and anomalous Nernst effect, which are associated with the Berry curvature contribution from the massive Dirac gaps in the 3D momentum space. In addition, we have also revealed topological Hall effect and topological Nernst effect, and we ascribed these phenomena to the Berry curvature associated with the magnetic field-induced non-zero scalar spin chirality. These features highlight the synergic effects of the Berry phases in both momentum space and real space of Kagome magnets, which render them excellent candidates for room-temperature electronic and thermoelectric applications based on transverse transport. In addition, through this program we have also study novel thermal transport properties of quantum materials by probing the thermal Hall effect, which can provide crucial insights into nontrivial topological character and chirality of quasiparticles like magnons and magnon-polarons. This report provides an overview of some project activities related to these accomplishments.

## **Project Activities:**

**A: Topological electronic and thermoelectric properties of Kagome magnets.** The unique Kagome lattice geometry gives rise to peculiar electronic properties, such as flat band, topological Dirac band, and van Hove singularity. By integrating magnetism with topological electronic band structure, Kagome magnets provide a promising platform to explore new quantum phenomena and understand the novel physics. Along this theme, we have investigated electronic and thermoelectric properties of  $\text{RMn}_6\text{Sn}_6$  ( $\text{R} = \text{Y}, \text{Tb}, \text{and Er}$ ) and  $\text{Fe}_3\text{Ge}$  Kagome magnets.

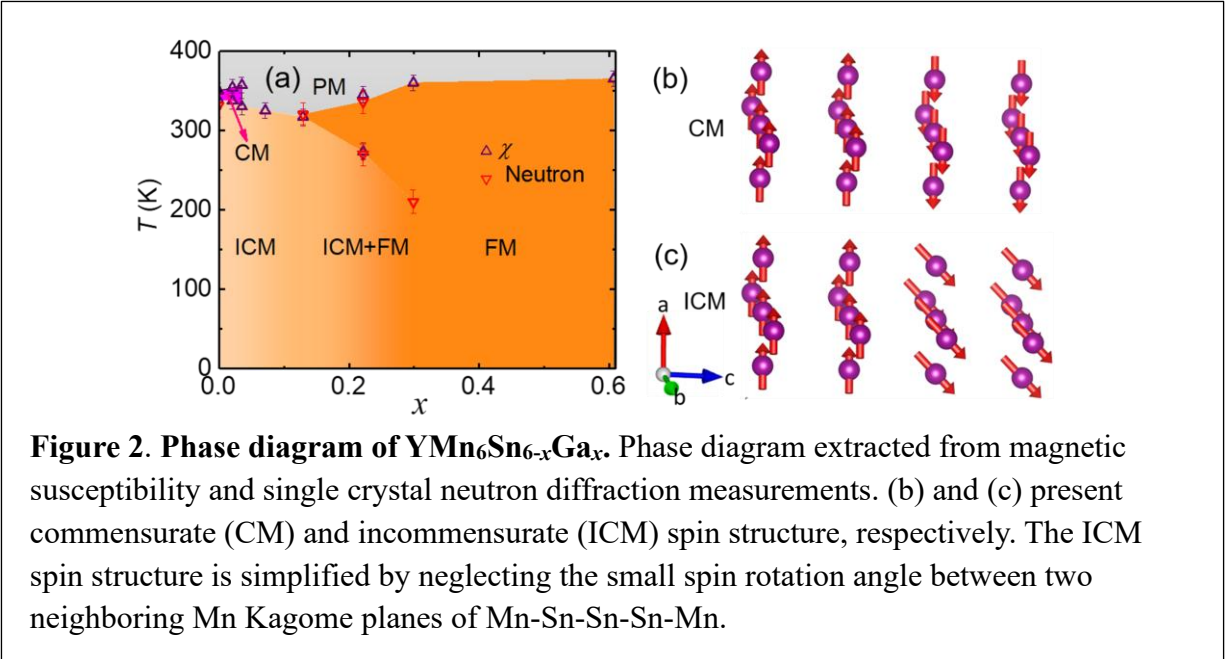
A.1) Exchange-biased topological transverse thermoelectric effect in a Kagome ferrimagnet  $\text{TbMn}_6\text{Sn}_6$ . Kagome metal  $\text{TbMn}_6\text{Sn}_6$  was recently discovered to be a ferrimagnetic topological Dirac material by scanning tunneling microscopy/spectroscopy measurements. In this project, we found that  $\text{TbMn}_6\text{Sn}_6$  exhibits large anomalous Hall effect and anomalous Nernst effect (Figure 1). We showed that these anomalous transverse transport features stem from the Berry curvature contribution from the massive Dirac gaps in the 3D momentum space. Furthermore, the transverse thermoelectric transport exhibits asymmetry with respect to the applied magnetic field,

i.e., an exchange-bias behavior. Together, these features place  $\text{TbMn}_6\text{Sn}_6$  as a promising system for the outstanding thermoelectric performance based on anomalous Nernst effect.



A.2) Complex magnetic phase diagram and anomalous Hall effect in Ga-substituted  $\text{YMn}_6\text{Sn}_6$ . In contrast to a ferrimagnetic ground state with out-of-plane magnetic moment in  $\text{TbMn}_6\text{Sn}_6$ , the pristine  $\text{YMn}_6\text{Sn}_6$  is an incommensurate antiferromagnet with competing antiferromagnetic couplings between adjacent planes of ferromagnetically aligned Mn spins along the  $c$ -axis. We found that the magnetic ground state of  $\text{YMn}_6\text{Sn}_{6-x}\text{Ga}_x$  ( $0 < x < 0.61$ ) evolves from incommensurate antiferromagnet to ferromagnet with increasing Ga substitution  $x$  (Figure 2), a feature which is accompanied by the decrease in magnetoresistance. Furthermore, the topological Hall effect observed in the pristine compound is absent in the Ga-substituted ones; instead, the anomalous Hall effect persists which may be associated with the Berry curvature of gapped Dirac bands near the Fermi energy. Our results highlight strong interplay between magnetism and topological electronic properties in this system that can be readily tuned via Ga substitution.

A.3) Anomalous and topological Nernst effect in Kagome magnets  $\text{ErMn}_6\text{Sn}_6$  and  $\text{Fe}_3\text{Ge}$ . Distinct from both ferrimagnetic  $\text{TbMn}_6\text{Sn}_6$  and antiferromagnetic  $\text{YMn}_6\text{Sn}_6$ ,  $\text{ErMn}_6\text{Sn}_6$  is an intermediate compound exhibiting an incommensurate antiferromagnetic phase followed by a ferrimagnetic phase transition upon cooling. We have investigated the electronic and thermoelectric properties of this compound, and we found that in the antiferromagnetic phase

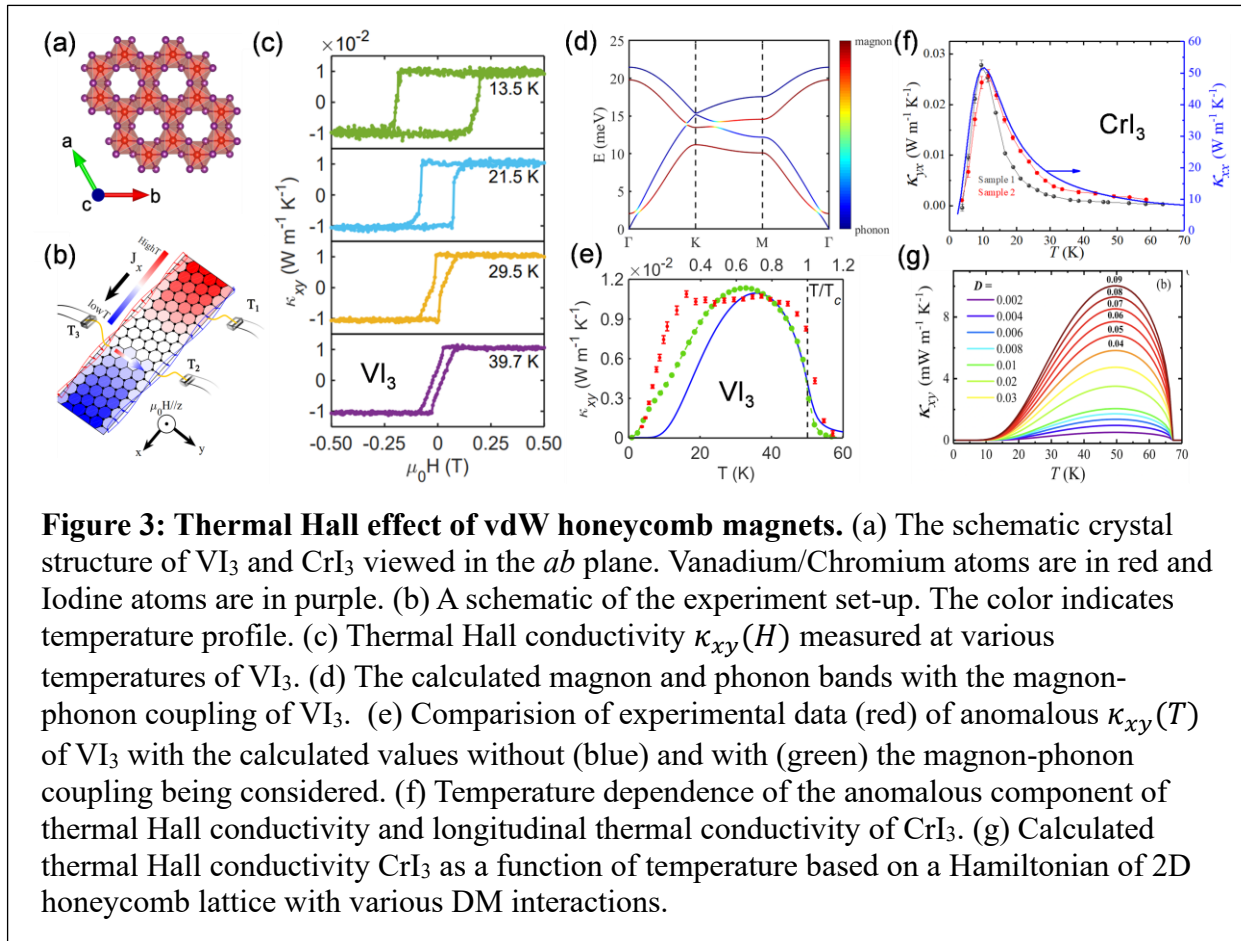


$ErMn_6Sn_6$  exhibits both topological Nernst effect and anomalous Nernst effect with a large Nernst coefficient reaching  $1.71 \mu V K^{-1}$  at 300 K and 3 T. This value surpasses that of most of the previously reported state-of-the-art canted antiferromagnetic materials. Similarly, we also found that  $Fe_3Ge$ , which has a slightly distorted magnetic Kagome lattice, exhibits large anomalous and topological Hall effect and Nernst effect. The topological Hall effect and topological Nernst effect are ascribed to the Berry curvature associated with the field-induced non-zero spin chirality. These studies highlight the synergic effects of the Berry phases in both momentum space and real space in metallic Kagome magnets.

**B: Topological excitations probed via thermal Hall effect studies.** Probing the thermal Hall effect (THE) is a powerful approach for unraveling emergent thermal transport properties of quantum materials, where a transverse heat current emerges in the presence of vertical magnetic field in response to a longitudinal temperature gradient, an analogy to the electric Hall effect in metallic systems. The THE phenomenon bridges thermodynamics and quantum mechanics, probing exotic quasiparticles such as magnons, phonons, and their hybridization (i.e, magnon-polarons), serving as an important complementary tool to inelastic neutron scattering. Importantly, the THE can provide crucial insights into nontrivial topological character and chirality of these quasiparticles. In this program, we have homebuilt a multifunctional setup which allows to investigate the THE of various quantum materials.

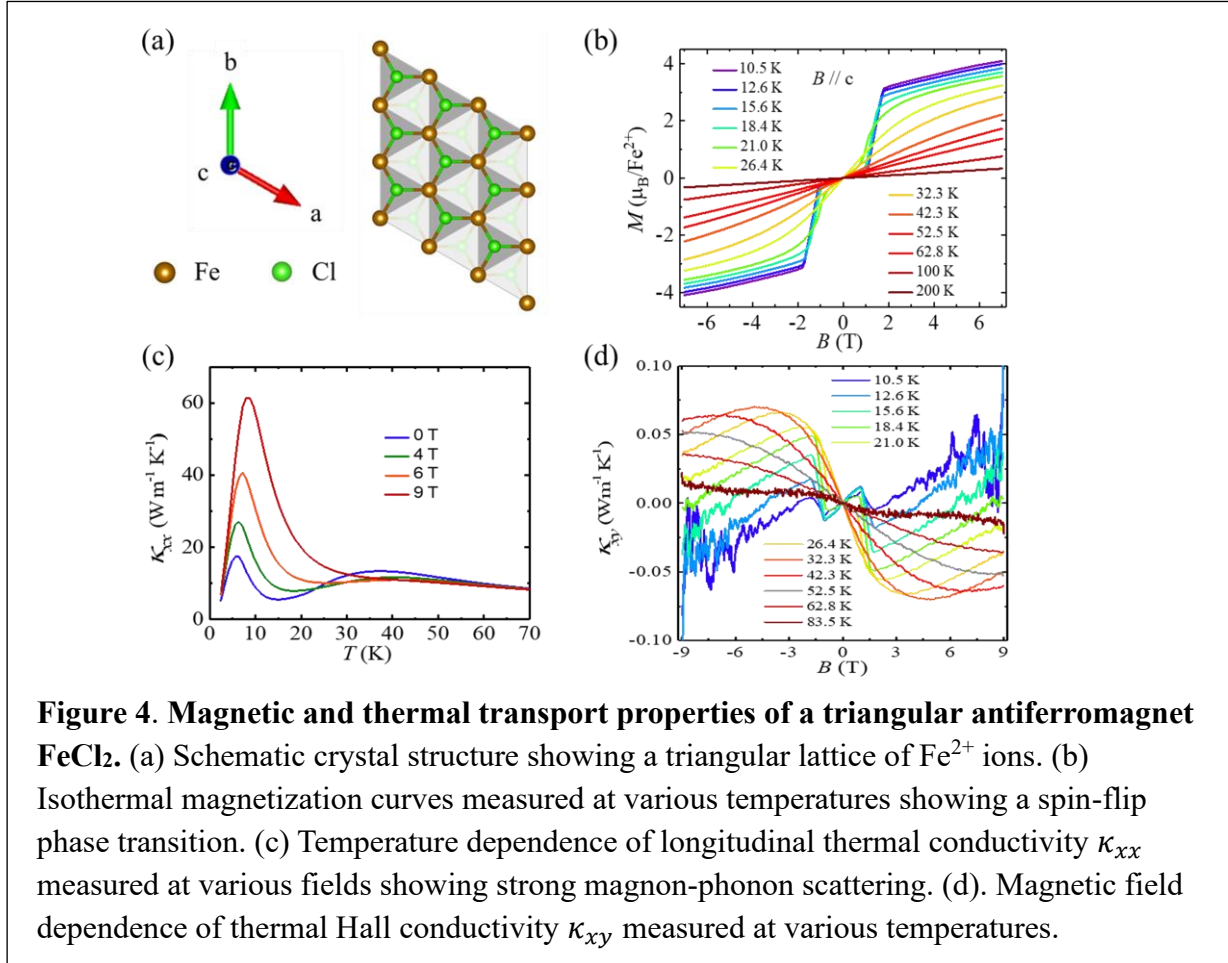
B.1) Thermal Hall effect driven by topological magnons. We had initiated studies of thermal transport properties in a honeycomb ferromagnet  $VI_3$ , in which the symmetry-allowed Dzyaloshinskii–Moriya interaction (DMI) is anticipated to give rise to topological magnetic excitations (magnons). We have observed a large THE over a wide range of temperature in  $VI_3$  with the thermal Hall conductivity  $\kappa_{xy}$  reaching a value of  $\sim 0.01 W K^{-1}m^{-1}$  which is an order

in magnitude larger than the thermal Hall signal observed in other ordered magnets. The thermal Hall signal persists in the absence of an external magnetic field with  $\kappa_{xy}$  being nearly the same as the saturated value, suggesting the nature of anomalous THE. In conjunction with theoretical calculations, we showed that  $\text{VI}_3$  exhibits a dual nature of the THE, which is dominated by topological magnons hosted by the ferromagnetic honeycomb lattice at higher temperatures and by magnon-polarons via magnon-phonon hybridization at lower temperatures (Figure 3(a-e)). Our results not only position  $\text{VI}_3$  as the first material to investigate THE arising from topological magnons and magnon-polarons, but also render it as a potential platform for spintronics/magnonics applications. In contrast, in another honeycomb ferromagnet  $\text{CrI}_3$ , in which previous inelastic neutron scattering studies have suggested topological nature of its magnetic excitations, we found that that the long-sought thermal Hall signal anticipated for topological magnons is fairly small (Figure 3(f-g)). These findings are anticipated to stimulate further neutron scattering studies on  $\text{CrI}_3$ , which can shed light on the intrinsic topological nature of magnetic excitations.



B.2) Thermal Hall effect driven by topological magnon-polarons. We also studied thermal transport properties of other van der Waals magnets, such as  $\text{FeCl}_2$  which is a triangular antiferromagnet. We revealed large magneto-thermal conductivity and thermal Hall effect in this material. The magneto-thermal conductivity reaches over  $\sim 700\%$ , indicating strong magnon-phonon coupling. Furthermore, we found an appreciable thermal Hall signal which changes sign

concurrently with the spin-flip transition from the antiferromagnetic state to the polarized ferromagnetic state (Figure 4). And our theoretical calculations suggested that the large Berry curvature induced at the anticrossing points of the hybridized magnon and acoustic phonon modes of FeCl<sub>2</sub> can lead to the observed thermal Hall effect.



**Figure 4. Magnetic and thermal transport properties of a triangular antiferromagnet FeCl<sub>2</sub>.** (a) Schematic crystal structure showing a triangular lattice of Fe<sup>2+</sup> ions. (b) Isothermal magnetization curves measured at various temperatures showing a spin-flip phase transition. (c) Temperature dependence of longitudinal thermal conductivity  $\kappa_{xx}$  measured at various fields showing strong magnon-phonon scattering. (d). Magnetic field dependence of thermal Hall conductivity  $\kappa_{xy}$  measured at various temperatures.

This award has partially supported the training of one postdoc and two graduate students. And this award has led to 19 publications and another 3 manuscripts under review, which are listed below.

1. C.Q. Xu, T.W. Heitmann, H. Zhang, X. Xu, and X. Ke, “Magnetic phase transition, magnetoresistance, and anomalous Hall effect in Ga substituted YMn<sub>6</sub>Sn<sub>6</sub> with a ferromagnetic kagome lattice”, Phys. Rev. B **104**, 024413 (2021).
2. M. Sretenovic, S. Okamoto, G. Peiker, T.X. Tang, H. Zhang, C.Q. Xu, T.W. Heitmann, Q. Zhang, C.R. dela Cruz, and X Ke, “Competing energetic states in Fe<sub>2</sub>WO<sub>6</sub> with strong spin-charge-lattice coupling”, Phys. Rev. B **104**, 134413 (2021).
3. H. Zhang, C.Q. Xu, C. Carnahan, M. Sretenovic, N. Suri, D. Xiao, and X Ke, “Anomalous thermal Hall effect in an insulating van der Waals magnet”, Phys. Rev. Lett. **127**, 247202 (2021).

4. Heda Zhang, Jahyun Koo, Chunqiang Xu, Milos Sretenovic, Binghai Yan, and **Xianglin Ke**, “*Exchange-biased topological transverse thermoelectric effects in a Kagome ferrimagnet*”, Nature Communications **13**, 1091(2022).
5. Lujin Min, Milos Sretenovic, Thomas Heitmann, Tyler W. Valerntine, Rui Zu, Venkatraman Gopanlan, Christina M. Rost, **Xianglin Ke**, and Zhiqiang Mao, “*A topological Kagome magnet in high entropy form*”, Communications Physics 5, 63 (2022).
6. H. Zhang, C.Q. Xu, S.H. Lee, Z.Q. Mao, and **X. Ke**, “*Thermal and thermoelectric properties of an antiferromagnetic topological insulator  $MnBi_2Te_4$* ”, Phys. Rev. B **105**, 184411 (2022).
7. Y.D. Guan, C.H. Yan, S.H. Lee, X. Gui, W. Ning, J.L. Ning, Y.L. Zhu, M. Kothakonda, C.Q. Xu, **X.L. Ke**, J.W. Sun, W.W. Xie, S.L. Yang, and Z.Q. Mao, “*Ferromagnetic  $MnBi_4Te_7$  obtained with low concentration Sb doping: a promising platform for exploring topological quantum states*”, Phys. Rev. Mater. **6**, 054203 (2022).
8. V. Petkov , T Durga Rao, A. Zafar, AM Milinda Abeykoon, E. Fletcher, J. Peng, Z.Q. Mao, and **X. Ke**, “*Lattice distortions and the metal-insulator transition in pure and Ti-substituted  $Ca_3Ru_2O_7$* ”, J. Phys. Condens. Matter. **51**, 015402 (2023).
9. Chunqiang Xu, Caitlin Carnahan, Heda Zhang, Milos Sretenovic, Pengpeng Zhang, Di Xiao, and **Xianglin Ke**, “*Thermal Hall effect in a van der Waals triangular magnet  $FeCl_2$* ”, Phys. Rev. B **107**, L060404 (2023).
10. Chunqiang Xu, Wei Tian, Pengpeng Zhang, **Xianglin Ke**, “*Large anomalous Nernst effect and topological Nernst effect in the noncollinear antiferromagnet  $NdMn_2Ge_2$* ”, Phys. Rev. B **108**, 064430(2023).
11. V Petkov, R Amin, M Jakhar, V Barone, AM Milinda Abeykoon, M Sretenovic, **X. Ke**, “*Charge density wave order, local lattice distortions, and topological electronic states in  $NbTe_4$* ”, Phys. Rev. B **108**, 174112(2023).
12. Chunqiang Xu, Heda Zhang, Caitlin Carnahan, Pengpeng Zhang, Di Xiao, **Xianglin Ke**, “*Thermal Hall effect in the van der Waals ferromagnet  $CrI_3$* ”, Phys. Rev. B **109**, 094415(2024).
13. M. Boswell, Q. Zhang, **X. Ke**, M. Li, ad W. Xie, “*Triangular Lattice of  $Ho^{3+}$  with  $S_{eff} = \frac{1}{2}$  in antiferromagnetic  $KHoSe_2$* ”, ACS Applied Electronic Materials **6**, 3827 (2024).
14. Mingyu Xu, Shuyuan Huyan, Haozhe Wang, Sergey L Bud'ko, Xinglong Chen, **Xianglin Ke**, John F Mitchell, Paul C Canfield, Jie Li, Weiwei Xie, “*Pressure-dependent “insulator-metal-insulator” behavior in Sr-doped  $La_3Ni_2O_7$* ”, Advanced Electronic Materials 10, 2400078 (2024).
15. Mingyu Xu, Yongbin Lee, **Xianglin Ke**, Min-Chul Kang, Matt Boswell, Sergey L Bud'ko, Lin Zhou, Liqin Ke, Mingda Li, Paul C Canfield, Weiwei Xie, “*Giant uniaxial magnetocrystalline anisotropy in  $SmCrGe_3$* ”, J. Am. Chem. Soc. **146**, 30294 (2024).
16. Shuvankar Gupta, Olajumoke Oluwatobiloba Emmanuel, Yasemin Ozbek, Mingyu Xu, Weiwei Xie, Pengpeng Zhang, **Xianglin Ke**, “*Giant Nernst angle in self-intercalated van der Waals magnet  $Cr_{1.25}Te_2$* ”, Materials Today Physics **50**, 101627 (2025).
17. Chandan De, Yu Liu, Sai Venkata Gayathri Ayyagari, Boyang Zheng, Kyle P. Kelley, Sankalpa Hazra, Jingyang He, Sylwia Pawledzio, Subin Mali, Samaresh Guchaaait, Suguru Yoshida, Yingdong Guan, Seng Huat Lee, Milos Sretenovic, **Xianglin Ke**, Le Wang, Mark H. Engelhard, Yingge Du, Weiwei Xie, Xiaoping Wang, Vincent H. Crespi, Nasim Alem, Venkatraman Gopalan, Qiang Zhang, Zhiqiang Mao, “*Discovery of a layered multiferroic compound  $Cu_{1-x}Mn_{1+y}SiTe_3$  with strong magnetoelectric coupling*”, Science Advances **11**, eadp9379 (2025).

18. Xu, Mingyu; Gonzalez Jimenez, Jose; Jose, Greeshma; Boonkird, Artittaya; Xing, Chengkun; Harrod, Chelsea; Li, Xinle; Zhou, Haidong; **Ke, Xianglin**; Bi, Wenli; Li, Mingda; Xie, Weiwei, “*Spin-flop and metamagnetic transition in monoclinic  $\text{Eu}_4\text{Bi}_6\text{Se}_{13}$* ”, *Chemistry of Materials* **37**, 1935 (2025).
19. Olajumoke Oluwatobiloba Emmanuel, Shuvankar Gupta, and **Xianglin Ke**, “ *$\text{ErMn}_6\text{Sn}_6$ : A Promising Kagome Antiferromagnetic Candidate for Room-Temperature Nernst Effect-based thermoelectrics*”, *Advanced Functional Materials* 2418715 (2025).
20. Wang, Haozhe; Xu, Mingyu; Koirala, Krishna Prasad; Melnick, Corey; Peng, Cheng; Gonzalez-Rivas, Mario U.; Lu, Jiaqi; Freese, Jessica; Wang, Le; Engelhard, Mark; Du, Yingge; **Ke, Xianglin**; Green, Robert; Hallas, Alannah; Li, Jie; Kotliar, Gabriel; Xie, Weiwei, “*Tetragonal  $\text{BaCoO}_3$ : A  $\text{Co}^{4+}$  ferromagnetic Mott insulator with inverted spin crossover*”, under review in *ACS Central Science* (2025).
21. Chunqiang Xu, Shuvankar Gupta, Hengxin Tan, Heyonhu Bae, Oluwatobiloba Olajumoke, Mingyu Xu, Yan Wu, Xiaofeng Xu, Pengpeng Zhang, Weiwei Xie, Binghai Yan, and **Xianglin Ke**, “*Large anomalous and topological Hall effect and Nernst effect in a Dirac Kagome magnet  $\text{Fe}_3\text{Ge}$* ”, under review in *Advanced Functional Materials* (2025).
22. Shuvankar Gupta, Yasemin Ozbek, Oluwatobiloba Olajumoke, Maya Bostock, Johnathan Kowalski, Pengpeng Zhang, and **Xianglin Ke**, “*Compelling evidence of second charge-density-wave in two dimensional  $\text{DyTe}_3$  and its impacts on magneto-transport*”, under review in *Phys. Rev. B* (2025).