# 研　究　助　成　報　告　書（　終了　）　No.1

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| 研究課題名 | 金属間化合物におけるトポロジカルネルンスト効果 |
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＜研究内容・成果等の要約＞ **Summary of Content**


Ferromagnets and antiferromagnets are two basic examples of magnetic materials. The former, ferromagnets, are already widely used in consumer electronics for data storage and many other applications. On the other hand, antiferromagnetic materials are being actively developed for ‘antiferromagnetic spintronics’, which is an efficient and robust candidate for next-generation electronic devices. In addition, antiferromagnetic materials have many ‘emergent properties’, which are functional responses to external stimuli that are thought to be relevant to future science and technology.

For example, the thermoelectric Nernst effect appears in certain antiferromagnets where the magnetic moments are not all along the same direction (noncoplanar, or complex antiferromagnets). This Nernst effect is a thermoelectric effect, which converts ‘useless’ heat flow to a useful electric voltage and which is being actively explored recently for green technology applications. The principal investigator (PI) of this project has developed a new class of thermoelectric Nernst effect, the ‘topological Nernst effect’ due to the complex antiferromagnetic order, and further explored this direction in the research period supported by the Izumi Science and Technology Foundation.

The main results obtained during the grant period are: (a) Realization of a giant Nernst effect in the antiferromagnet $CoNb_3S_6$ [N.D. Khanh, M. Hirschberger, *et al.*, accepted to Nature Communications] and (b) a detailed investigation of the magnetic structure of $Nd_3Ru_4Al_{12}$, a material with complex magnetism and a large Nernst effect [P. Baral, Y. Ishihara, *et al.*, manuscript in preparation]. As for (a), we have done precision measurements of the thermoelectric effect in our custom-built measurement setup. We have further calculated the electronic structure of $CoNb_3S_6$ and obtained an estimate for the thermoelectric effect, which is in good quantitative agreement with the experiment. For the calculations, we have obtained a new software package and workstation with support of the Izumi foundation. As for (b), we have grown single crystals with raw materials supported by the Izumi foundation and performed high-quality neutron scattering experiments at JRR-3 facility in Tokaimura, Japan. Therefore, the magnetic structure of $Nd_3Ru_4Al_{12}$ was determined and it is now possible to compare to the observed thermoelectric effect and to electronic structure calculations. It is believed that a thorough understanding of the thermoelectric Nernst effect, which has been achieved in this grant period, will pave the way for applications not only in ‘energy harvesting’ or green technology, but also in the advanced sensing of small temperature gradients in zero magnetic field.

＜研究発表（口頭、ポスター、誌上別）＞

Research papers and theses

1. Y. Ishihara, "*Exploration of magnetic structure of Kagome lattice materials via neutron scattering*" (中性子散乱実験によるカゴメ格子物質の磁気構造の探索), 修士論文, 東京大学 大学院工学系研究科 物理工学専攻, submitted in January 2025
2. N. D. Khanh, S. Minami, M. M. Hirschmann, T. Nomoto, M.-C. Jiang, R. Yamada, D. Yamaguchi, Y. Hayashi, Y. Okamura, H. Watanabe, G.-Y. Guo, Y. Takahashi, S. Seki, Y. Taguchi, Y. Tokura, R. Arita, and M. Hirschberger, "*Gapped nodal planes drive a large topological Nernst effect in a chiral lattice antiferromagnet*", Nature Communications (in print, 2025)

Oral conference presentations

1. M. Hirschberger, "Novel superstructures of spin and charge, and effect on electronic structures", Ringberg – Symposium of the Max-Planck-Society (Germany) "Exotic States Of Quantum Condensed Matter", organized by Max-Planck-Institute Stuttgart, Germany, November 14, 2023
2. M. Hirschberger, "Probing chiral magnetic textures by quantum beam scattering", 8th International Symposium of Quantum Beam Science (ISQBS2023), Ibaraki University, Mito Campus, November 28, 2023
3. M. Hirschberger, N. D. Khanh, T. Nakajima, L. Spitz, *et al.,* "Energy harvesting using the thermoelectric Nernst effect of spin textures", The 1st U-Tokyo ISSP・RIKEN CEMS Collaboration Workshop (第1回東大ISSP・理研CEMS連携ワークショップ), Kashiwa, Japan, January 24, 2024
4. M. Hirschberger, "量子物質における電子構造由来の熱電効果の探索" (Thermoelectric studies of electronic structure in quantum materials), Spring Meeting of the Japanese Physical Society (online), March 19, 2024
5. M. Hirschberger, "Topology and anomalous Hall effect in reciprocal space from spin chirality in real space", Conference of Condensed Matter Physics, Yangtze River Delta Physics Research Center, Liyang, China, August 5, 2024

Poster presentations

1. N. D. Khanh, S. Minami, M. Hirschmann, T. Nomoto, M. C. Jiang, R. Yamada, D. Yamaguchi, Y. Hayashi, Y. Okamura, G. Y. Guo, Y. Takahashi, S. Seki, Y. Taguchi, Y. Tokura, R. Arita, M. Hirschberger, "Topological Nernst effect in a chiral van der Waals magnet", JST CREST「トポロジー」領域 2023年度領域会, January 19, 2024 (presented by Max Hirschberger)
2. N. D. Khanh, S. Minami, M. Hirschmann, T. Nomoto, M. C. Jiang, R. Yamada, D. Yamaguchi, Y. Hayashi, Y. Okamura, G. Y. Guo, Y. Takahashi, S. Seki, Y. Taguchi, Y. Tokura, R. Arita, M. Hirschberger, "Anomalous Hall & Nernst effects in reciprocal space from spin chirality in real space", RIKEN Center for Emergent Matter Science Topical Meeting, Wako, Japan, May 21, 2024 (presented by Max Hirschberger)

＜研究の目的、経過、結果、考察（5000 字程度、中間報告は 2000 字程度）＞

研究の目的 **Purpose of Research**

The ultimate purpose of our research is the development of new thermoelectric devices based on the Nernst effect of antiferromagnets with topological properties. In particular, the winding number or topological charge of a spin texture is defined as [N. Nagaosa *et al.*, Nat. Nanotechnol. **8**, 899 (2013)]

$$n_{sk} = \frac{1}{4\pi}\int_{\mathrm{muc}} d^3r\, \boldsymbol{n}\cdot\left(\frac{\partial \boldsymbol{n}}{\partial x}\times\frac{\partial \boldsymbol{n}}{\partial y}\right) \tag{1}$$

where 'muc' indicates integration over a magnetic unit cell, $\boldsymbol{r} = (x, y, z)$ and $\boldsymbol{n}$ is a normalized magnetization field, $\boldsymbol{n} = \boldsymbol{m}/|\boldsymbol{m}|$. This topological charge becomes non-zero in certain noncollinear antiferromagnets, e.g. in skyrmion lattice magnets. The principal investigator's previous research has demonstrated that $n_{\mathrm{sk}}$ can generate a large thermoelectric effect, the topological Nernst effect, which may have technological applications if it can exist in a broad class of materials: M. Hirschberger *et al.*, Phys. Rev. Lett. **125**, 076602 (2020).

In this research, we pursued a more quantitative understanding of this topological Nernst effect by studying the interconnection of the magnetic structure of solids and $n_{\mathrm{sk}}$ (via neutron scattering), and the connection between $n_{\mathrm{sk}}$ and the thermoelectric effect (via thermoelectric measurements and electronic structure calculations). It is essential to demonstrate the connection of the magnetic structure and thermoelectric transport property, to develop the understanding of the topological Nernst effect and to pave the way for future thermoelectric devices based on antiferromagnetic materials.

研究の経過 **Progress**

Two materials are targeted in this research project: The first is $Nd_3Ru_4Al_{12}$, a ferromagnet with a noncollinear magnetic structure. We use neutron scattering experiments to demonstrate the finite $n_{\mathrm{sk}}$ in this material in the magnetic ground state. Combined with previously published thermoelectric measurements [K. Kolincio *et al.*, Proc. Natl. Acad. Sci. USA **118**, e2023588118 (2021)], this result demonstrates the existence of a topological Nernst effect in this material.

The second is $CoNb_3S_6$, whose magnetic structure was recently revealed through neutron scattering by our collaborators in Japan [Nat. Phys. **19**, 961–968 (2023)]. Here, we measured the thermoelectric properties in detail, using our custom-made experimental measurement setup, and compared quantitatively the resulting Nernst effect data to electronic structure calculations. Our state-of-the-art electronic structure calculation includes, in detail, the interplay of magnetic structure and conduction electrons. We found good agreement between experiment and theory. In addition, the observed Nernst effect of $CoNb_3S_6$ is extremely large for an antiferromagnet and on the same order of magnitude as the Nernst effect in ferromagnets. Note that, in the usual case, ferromagnets have much larger Nernst effects than antiferromagnets.

In combination, our experimental progress on magnetic properties and associated thermoelectric responses reflects the content of our research proposal to the Izumi foundation. The research content of the proposal document was faithfully executed.

研究の結果 **Results**

**① Careful study of magnetic order in new thermoelectric materials.**

Figure 1 shows our magnetic scattering result for the noncollinear magnet $Nd_3Ru_4Al_{12}$. In Fig. 1(a), we show the 'all-in-all-out' magnetic structure determined for $Nd_3Ru_4Al_{12}$ using various neutron diffraction experiments in Japan.

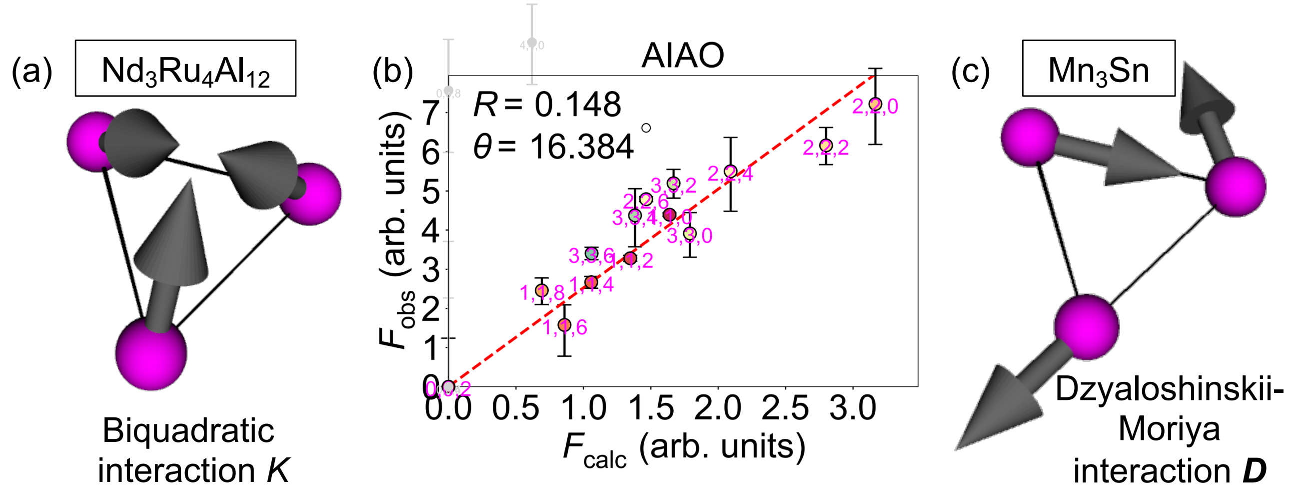


**Figure 1**: Magnetic order in $Nd_3Ru_4Al_{12}$. See text for discussion.

We used beamline PONTA-5G of JRR-3 in Tokaimura, Ibaraki prefecture to conduct polarized and non-polarized neutron scattering experiments. In addition, we obtained supporting data from beamline TAIKAN at J-PARC facility. The experiments were carefully analyzed by a student, Ms. Ishihara, and the magnetic structure was determined. As an example, Fig. 1(b) shows the comparison of calculated ($x$ -axis) and observed ($y$ -axis) magnetic structure factors, where the neutron scattering intensity is proportional to the square of the structure factor; the line indicates perfect agreement of model and experiment. The quality of the agreement is good and thus the magnetic structure was confirmed.

At this point, we draw an analogy between our target material $Nd_3Ru_4Al_{12}$ and the well-known compound $Mn_3Sn$, which is known to realize a giant thermoelectric Nernst effect at high temperatures due to its noncollinear magnetic structure [Fig. 1(c); Nat. Phys. **13**, 1085 (2017)]. The two materials have the same crystallographic space group $P6_3/mmc$, i.e., their crystal structures are closely related. Therefore, our study on magnetism $Nd_3Ru_4Al_{12}$ can provide important insights into the magnetism of a broad class of next-generation thermoelectric materials, including $Mn_3Sn$.

Specifically, the total energy of two neighboring spins $\boldsymbol{S}_i$, $\boldsymbol{S}_j$ is defined by the spin Hamiltonian,

$$\mathcal{H} = J\boldsymbol{S}_i \cdot \boldsymbol{S}_j + \boldsymbol{D} \cdot (\boldsymbol{S}_i \times \boldsymbol{S}_j) + K(\boldsymbol{S}_i \cdot \boldsymbol{S}_j)^2 \quad (2)$$

where the first, second, and third term represent the conventional Heisenberg exchange interaction $J$, the Dzyaloshinskii-Moriya (antisymmetric) exchange interaction $D$, and the biquadratic interaction $K$, respectively. The $D$ has been studied extensively, and it is known that the magnetic structure of $Mn_3Sn$ can be explained by two terms, $J$ and $D < 0$, in principle. Further, the role of $K$ is more controversial: recent theoretical calculations predict that metallic materials should have a large $K$ [Phys. Rev. B **95**, 224424 (2017)]. In collaboration with researchers in Germany, who probed various magnetic interactions by detailed numerical 'Monte Carlo' calculations, we confirmed that the $K$ term is very likely to stabilize the magnetic order observed in $Nd_3Ru_4Al_{12}$. Now, we are still studying the effect of long-range interactions, such as the coupling between two different triangles, on the magnetic order in this material.

② **Observation of large thermoelectric effects in magnetic materials.**

Next, we investigate the thermoelectric effect in a noncollinear antiferromagnet where the magnetic structure has already been reported. In $CoNb_3S_6$, the structural blocks of magnetic order are cobalt tetrahedra, on which cobalt magnetic moments point either 'into' or 'out of' a tetrahedron.

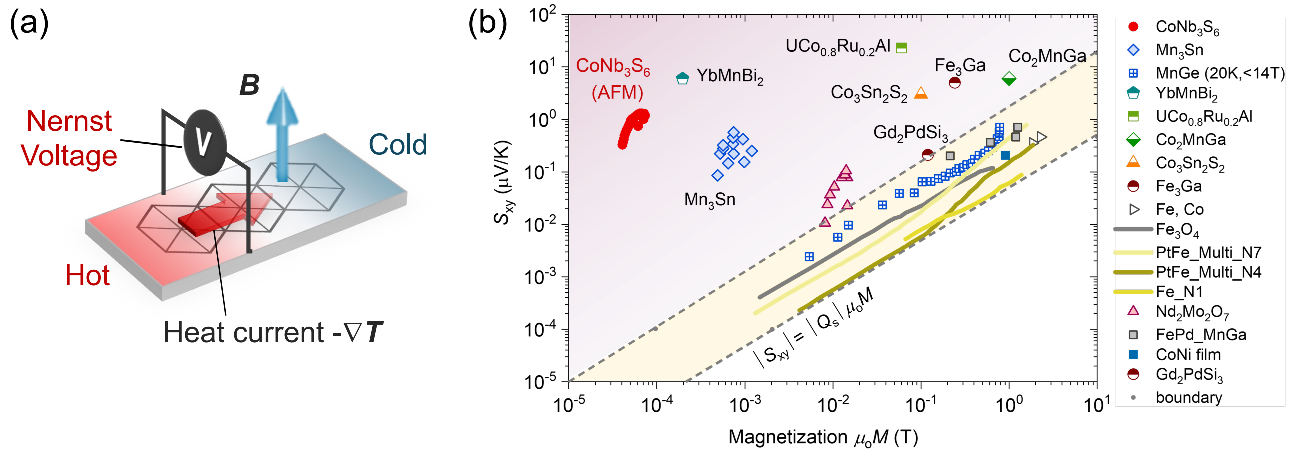


**Figure 2**: Thermoelectric Nernst effect in $CoNb_3S_6$. In panel (b), the signal of $CoNb_3S_6$ appears at low magnetization $M$ but is as large as the Nernst effect of ferromagnets.

Following the neutron scattering research recently reported by Takagi *et al.*, Nat. Phys. **19**, 961–968 (2023), we decided that a large topological Nernst effect is likely for this antiferromagnetic structure with zero net magnetization, $M = 0$. Therefore, we prepared high-quality single crystals and cut them into the shape of a tetragonal plate. We attached gold wires by silver paint and thus connected electrical connections and thermometers to the sample. As shown in Fig. 2(a), we applied a heat current along the $x$-direction of a coordinate system using a chip heater, detected the resulting thermoelectric voltage $V_y$ perpendicular to the heat current, and thus calculated $S_{xy} = V_y/|-\nabla_x T|$, the thermoelectric Nernst effect. It was found that even in zero magnetic field, a very large Nernst effect appears below the magnetic transition temperature $T_N$. This is attributed to the spin winding of this magnetic structure according to Eq. (1).

Quantitatively, we compare $CoNb_3S_6$'s Nernst effect to other antiferromagnets, and to ferromagnets, in Fig. 2(c). Data from $CoNb_3S_6$ is located on the upper left side of the plot, indicating a very large Nernst effect in combination with a nearly zero net magnetization $M$. By comparison to theoretical electronic structure calculations for $CoNb_3S_6$ with including the details of the magnetic structure, we further confirm that the large thermoelectric effect originates from $n_{sk}$ and should therefore be called a 'topological Nernst effect'.

研究の考察 **Discussion**

The thermoelectric Nernst effect of antiferromagnets provides a vast arena for material exploration and development of advanced functionality. This project, sponsored by the Izumi foundation, has added new candidate materials $CoNb_3S_6$ and $Nd_3Ru_4Al_{12}$ which are relevant to understand the fundamental principles of the thermoelectric Nernst effect. More recently, the design of novel devices based on the Nernst effect, such as 'thermopile' devices, represents progress towards technologically relevant device architectures for the Nernst effect [Nat. Phys. 20, 254–260 (2024)]. It is believed that further progress in fundamental understanding and device design will make energy-saving, Nernst-based thermoelectrics economically viable.