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研究課題名（英文） Electrical manipulation of Antiferromagnetic State in Non-centrosymmetric Systems

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研究成果の概要（和文）：本研究では、インターカレートvdW物質CoNb3S6とCoTa3S6における時間反転対称性の破れた反強磁性構造の研究を通じて、反強磁性状態を「読み」、「書き」制御する新しいパラダイムを実証したい。このプロジェクトの主な成果は以下の通りである：(i)CoNb3S6のオール・イン・オールアウト型非平面反強磁性構造の解明、(ii)非自明なベリー曲率から発生する架空磁場に関連したトポロジカル・ホール効果やネルンスト効果を含むトポロジカル輸送現象の研究、(iii)非相反的電気輸送の観測-「ダイオード」効果、(iv)直流電流による反強磁性状態のスイッチングの実証。

研究成果の学術的意義や社会的意義
The insights gained from this research could drive innovations in information technology, particularly in the fields of data storage and processing. Spintronic devices based on AFM materials promise higher speeds and lower energy consumption compared to traditional electronic devices.

研究成果の概要（英文）：In this work, via investigation of antiferromagnetic structure with broken time reversal symmetry in intercalated vdW material CoNb3S6 and CoTa3S6, we would like to demonstrate a new paradigm to identify-“ read ” and control-“ write ” the antiferromagnetic states. The main achievements of this project can be summarized as: (i) Elucidation of the all-in all-out type non-coplanar antiferromagnetic structure of CoNb3S6; (ii) Investigation of topological transport phenomena, including topological Hall and Nernst effects, associated with fictitious field generated from non-trivial Berry curvature; (iii) observation of non-reciprocal electrical transport - “ diode ” effect and (iv) demonstration of switching the antiferromagnetic states by DC electric current. These results contribute to establish a foundation for further exploration new property of this intriguing class of antiferromagnetic materials with broken time reversal symmetry.

研究分野：Magnetism

キーワード：Antiferromagnet　Symmetry Breaking　Nonreciprocal transport　Data Storage

１．研究開始当初の背景

The manipulation of magnetism by electric current or field without heating has attracted significant attention for the development of next - generation spintronics devices. Several advances have been made utilizing spin-transfer torque (STT) and spin-orbit torque (SOT) switching mechanisms. However, most of these technologies are based on ferromagnetic materials with net magnetization. In the past decade, antiferromagnets have started to gain substantial attention. Compared to ferromagnetic (FM) materials, antiferromagnetic (AFM) states are more challenging to detect and control, despite their significant advantages. Recently, research has focused on the topological AFM Mn_3Sn, which exhibits a large Anomalous Hall Effect (AHE) despite its AFM nature. By passing a current through a metal layer (such as Pt or W) adjacent to Mn_3Sn, AFM states can be switched via the SOT mechanism. The ability to switch AFM states using protocols similar to those used for FM metals can drive scientific and technological advancements in the fields of spintronics and topological materials. Motivated by these works, further exploration of materials and mechanisms to switch AFM states by electrical means is highly desired. Very recently, a large Anomalous Hall Effect (AHE) has been discovered in the chiral lattice antiferromagnet $CoNb_3S_6$, characterized by two antiferromagnetic (AFM) states with broken time-reversal (TR) symmetry. However, unlike Mn_3Sn, $CoNb_3S_6$ lacks an inversion center, meaning its spatial inversion (SI) symmetry is broken. Such features suggest that $CoNb_3S_6$ and its sibling compounds can be promising platforms for investigating the interplay between antiferromagnetism and symmetry breaking.

２．研究の目的

This study aims to investigate the antiferromagnetic structures with broken time-reversal symmetry in intercalated van der Waals materials, specifically $CoNb_3S_6$ and $CoTa_3S_6$. Our objective is to introduce a new approach for identifying ("reading") and controlling ("writing") antiferromagnetic states, based on below targers: (i) Exploring Topological Transport Phenomena associated with recently discovered non-coplanar spin texture in $CoNb_3S_6$; (ii) Detecting Non-Reciprocal Electrical Transport phenomena; (iii) Demonstrating the Switching of Antiferromagnetic States by DC Electric Current.

３．研究の方法

* Crystal growth. $CoNb_3S_6$ and $CoTa_3S_6$ polycrystalline samples were synthesized by combining Co, Ta, Nb, and S in stoichiometric proportions inside a silica ampoule under vacuum. The mixture was then heated first at 300 °C for 2 days and then at 850 °C for 1 day. To grow single crystals, chemical vapor transport employing iodine as the transport medium was utilized. Crystal orientations were determined using back-reflection X-ray Laue photography, while the sample's purity was verified through powder X-ray diffraction.

* Magnetic and electrical transport properties. Magnetic susceptibility and magnetization measurements were conducted utilizing a superconducting quantum interference device magnetometer (MPMS, Quantum Design). Meanwhile, measurements of resistivity (longitudinal and transverse) were carried out using the A.C transport option within a PPMS (Quantum Design).

* Measurement of thermoelectric Nernst effect. Thermoelectric experiments are carried out using

a customized sample stage mounted in a commercial PPMS cryostat (Quantum Design, Inc.). Thermopower (Seebeck) and Nernst effect are recorded using a one-heater two-thermometer technique in steady-state mode. A temperature gradient $-\nabla T$ is applied along the a-direction within the basal plane, while the magnetic field is parallel to the *c*-axis.

* Measurement of electric current switching magnetic state. We first fabricated the $CoNb_3S_6$ FIB devices (size 22μm x 12μm x 1.5μm). A conventional four-probe method is utilized to measure the Hall voltage with current of 0.2 mA. Before that, a writing electrical pulse current is applied with a duration of 100 ms and then stop. Following, the reading current is applied, and Hall voltage is measured after a wait time of 600 ms.

４．研究成果

4.1. Non-coplanar antiferromagnetic structure and topological Hall effect

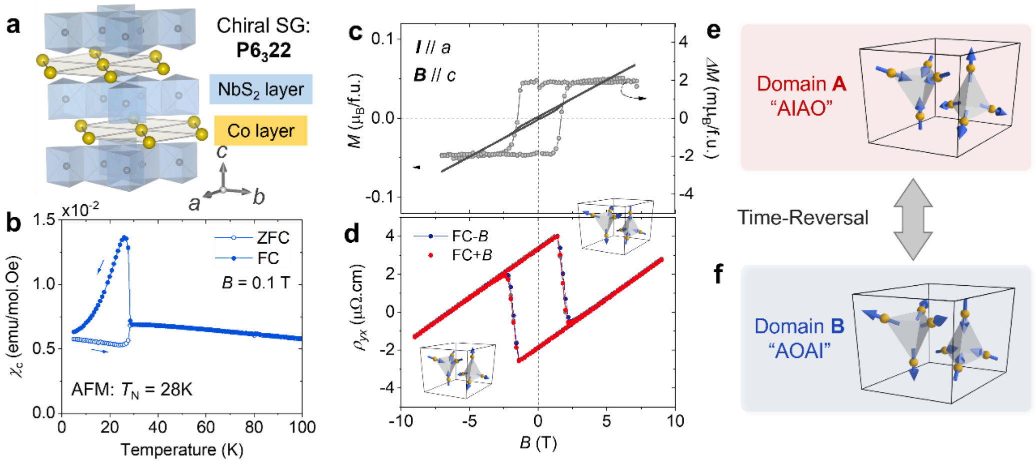

Figure 1. (**a**) Crystal structure of $CoNb_3S_6$. (**b**) Temperature dependence of magnetic susceptibility in *B // c*. (**c, d**) *B* dependence of *M* and Hall resistivity ρ_{yx} at 26K. (**e, f**) Non-coplanar antiferromagnetic states in $CoNb_3S_6$: All-in All-out and All-out All-in.

In this part, we investigate the magnetic structure and transport property of $CoNb_3S_6$, belonging to a huge family of transition metal intercalated in dichalcogenide MT_3X_6 (*M*, *T* = 3d and 5d transition metals, respectively; *X* = S, Se), which crystallizes in chiral structure (space group $P6_322$) (**Figure 1a**). Below T_N ~ 28 K, $CoNb_3S_6$ undergoes a magnetic phase transition (**Figure 1b**). In contrast with previous study proposed a collinear AFM structure with Co^{2+} moments ⊥ *c*-axis, we demonstrate that $CoNb_3S_6$ hosts a non-coplanar all-in all-out (AIAO) type AFM structure characterized by the co-existence of multiple magnetic modulation vector $\boldsymbol{Q}$ = (1/2, 1/2, 0) (a triple-$\boldsymbol{Q}$ state). The 2 degenerated magnetic domains all-in all-out (AIAO) and all-out all-in (AOAI) (**Figure 1e, f**) break the TRS, giving rise to the emergence of large spontaneous Hall effect originated from Berry curvature proportional to a scalar spin chirality χ_{ijk} defined by the solid angle between neighboring spins $\boldsymbol{S}_{i,j,k}$ as $\chi_{ijk} \equiv \boldsymbol{S}_i \cdot (\boldsymbol{S}_j \times \boldsymbol{S}_k)$, despite its tiny magnetization *M* (**Figure 1c**). Spontaneous Hall response in $CoNb_3S_6$ can be termed as topological Hall effect (THE), which is driven by fictitious magnetic field associated with non-coplanar antiferromagnetic structure. Part of this result was published in H. Takagi *et al.*, Nat. Phys. **19**, 961 (2023).

4.2. Topological Nernst effect

We demonstrate in **Figure 2** the TNE in $CoNb_3S_6$, which corresponds to antiferromagnetic domains in absence of a sizable net magnetization. Here, the thermal gradient $-\nabla T$, magnetization *M*, and detected Nernst voltage V_{xy} lie orthogonal (**Figure 2a**). **Figure 2b** shows the Nernst thermopower $S_{xy} = V_{xy} / (-\nabla T)$ measured at T = 26 K, just below T_N. A clear hysteresis in S_{xy} can be observed upon sweeping the magnetic field B between ±9 T with hysteresis width of ~4 T, a hallmark of the transformation between two antiferromagnetic domain states (i.e., AIAO and AOAI). The enormous Nernst thermopower S_{xy} ~ 1 μV/K observed in zero field, which is comparable to that of conventional ferromagnets. Meanwhile, the tiny spontaneous magnetization $M \sim 2 \times 10^{-3}$ μ_B/f.u. of $CoNb_3S_6$ is much smaller than the saturated magnetization value expected for Co^{2+} moments (S = 3/2); In view of the nearly compensated AFM state with vanishingly small net magnetization, the Nernst effect in $CoNb_3S_6$ is evidently not simply proportional to M as in ferromagnets (**Figure 2c**). Instead, the large TNE can be attributed to Berry curvature originating from non-coplanar AIAO type antiferromagnetic order. Preprint of this work was at N. D. Khanh *et al*, arXiv:2403.01113 (2024) (The manuscript is under reviewed for publication).

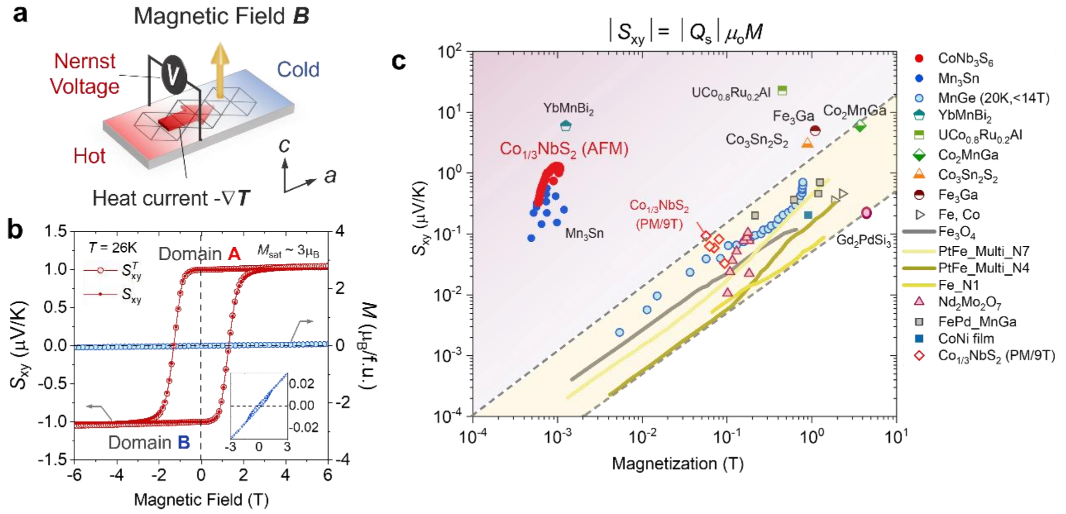

Figure 2. (**a**) Schematic illustration of experimental setup for Nernst measurement. (**b**) Large spontaneous Nernst effect at 26K, contrasting to tiny magnetization *M.* (**c**) Scaling of Nernst coefficient of $CoNb_3S_6$ and other materials versus magnetization M, excluding the magnetization origin of Nernst signal.

4.3. Nonreciprocal transport of electric current

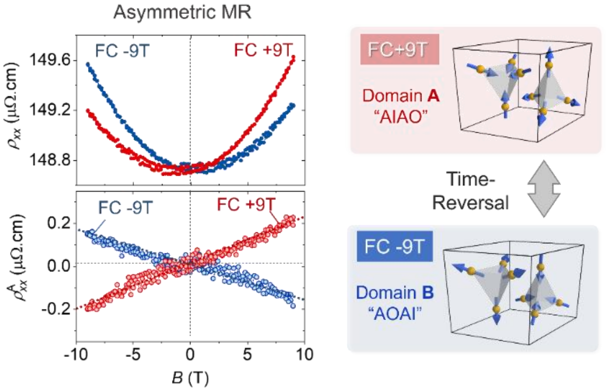

Figure 3. Asymmetric magnetoresistance (MR) of $CoNb_3S_6$ measured at 5K in I // a, B // c. The opposite slope of asymmetric MR reflects the 2 AFM states with broken TRS.

$CoNb_3S_6$ is also a promising candidate to explore non-reciprocal electrical transport phenomena associated with broken time-reversal (TR) and spatial inversion (SI) symmetries, enabling many intriguing phenomena, such as non-reciprocal propagation of quasiparticles (electron, spin, magnon…) or magnetochiral anisotropy or inverse Edelstein effect. To this aspect, we investigate the nonreciprocal electrical transport phenomena in $CoNb_3S_6$, and observed odd-parity linear magnetoresistance (MR) $\boldsymbol{R(+B) \neq R(-B)}$ in AFM phase. In principle, MR can be expressed as $R(B)$

= R_o + a.B + b.B^2. While B^2 term exhibits the conventional MR, the B-linear term reflects the TRS breaking. The asymmetric MR and magnitude of spontaneous Hall resistivity acquired at zero field exhibits similar behavior upon the variation of temperature and cooling field, which evidences the correlation between AFM order and AHE (**Figure 3**).

4.4. Demonstration of switching AFM states by electric current

In $CoNb_3S_6$, the THE is associated with two AFM states, AIAO and AOAI with broken TRS, as seen by opposite signs of Hall voltage V_H upon sweeping magnetic field (**Figure 1d**). We aim to use electric current to switch these two AFM states. Here, the current is passed directly on AFM layer (**Figure 4b**). A magnetic field will be applied to create a single domain of AFM state (AIAO or AOAI) before passing a large DC current pulse (several 10 milliamperes) in a very short time (~millisecond) to avoid heating. After the pulse is turned off, V_H will be probed by small current (**Figure 4a, b**). Depending on the value of V_H, the AFM states can be identified.

Here, we observed a clear hysteresis of V_H respecting to the DC current. The amplitude of V_H corresponds to the Hall signal of non-coplanar AFM states (**Figure 4c**). However, one of the problems is that the switching occurs indeterministic, which might be due to the effect of heating at the contact. To overcome this difficulty, we would like to improve the device fabrication process to achieve smaller FIB samples. This may allow to enhance current density large enough to switch the AFM states, while the current magnitude is still unchanged to keep the effect of heating as minimal as possible. Such proposal is the issue for future study.

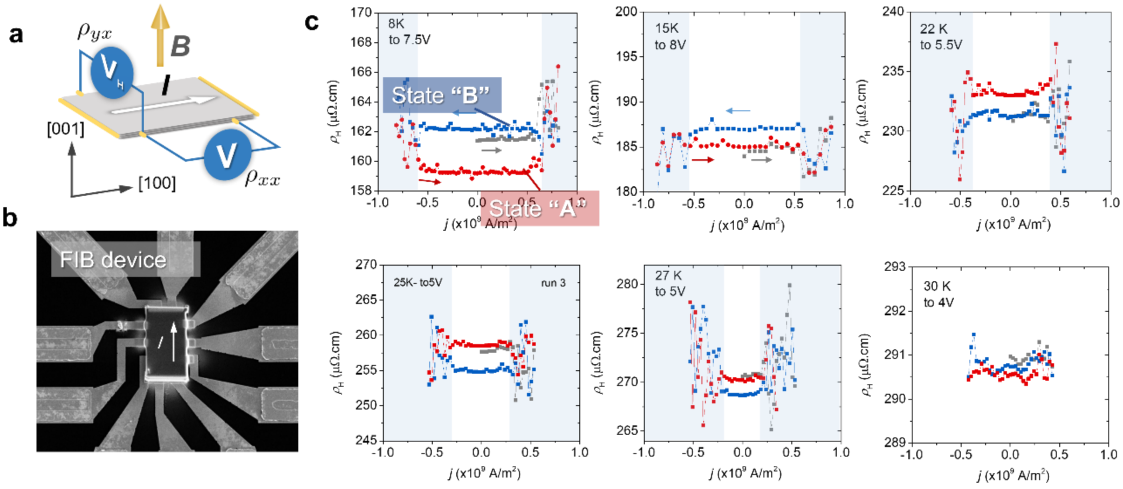

Figure 4. (**a**) Schematic illustration of experiment configuration. (**b**) SEM image of FIB device. (**c**) Demonstration of current switching AFM domains in $CoNb_3S_6$ micro-device at several temperatures via measurement of DC current dependence, suggesting the possibility of switching behavior. However, miniaturizing the heating effect is required to obtain deterministic switching.

５．主な発表論文等

〔雑誌論文〕　計4件（うち査読付論文　3件／うち国際共著　3件／うちオープンアクセス　1件）

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掲載論文のDOI（デジタルオブジェクト識別子） なし	査読の有無 無
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掲載論文のDOI（デジタルオブジェクト識別子） 10.1038/s41567-023-02017-3	査読の有無 有
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掲載論文のDOI（デジタルオブジェクト識別子） 10.1103/PhysRevB.107.104421	査読の有無 有
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〔図書〕　計0件

〔産業財産権〕

〔その他〕

https://www.t.u-tokyo.ac.jp/en/press/pr2023-04-21-002

６．研究組織

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７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関