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Received: December 1, 2022; Accepted: February 14, 2023; Published Online: February 20, 2023; https://doi.org/10.1016/j.xinn.2023.100399
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REPORT

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GRAPHICAL ABSTRACT

PUBLIC SUMMARY
- Modulation of Weyl electronic state by external field is highly desired.
- Weyl electronic state and transverse transport are synchronously modulated.
- Such modulation is realized by magnetic field-induced spin canting.
- Non-collinear magnetism and topological electronic state can be connected.
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Received: December 1, 2022; Accepted: February 14, 2023; Published Online: February 20, 2023; https://doi.org/10.1016/j.xinn.2023.100399

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Citation: Shen J., Gao J., Yi C., et al., (2023). Magnetic-field modulation of topological electronic state and emergent magneto-transport in a magnetic Weyl semimetal. The Innovation 4(2), 100399.

The modulation of topological electronic state by an external magnetic field is highly desired for condensed-matter physics. Schemes to achieve this have been proposed theoretically, but few can be realized experimentally. Here, combining transverse transport, theoretical calculations, and scanning tunneling microscopy/spectroscopy (STM/S) investigations, we provide an observation that the topological electronic state, accompanied by an emergent magneto-transport phenomenon, was modulated by applying magnetic field through induced non-collinear magnetism in the magnetic Weyl semimetal EuB₆. A giant unconventional Hall effect (UAHE) is found during the magnetization re-orientation from easy axes to hard ones in magnetic field, with a UAHE peak around the low field of 5 kOe. Under the reasonable spin-canting effect, the folding of the topological anti-crossing bands occurs, generating a strong Berry curvature that accounts for the observed UAHE. Field-dependent STM/S reveals a highly synchronous evolution of electronic density of states, with a dI/dV peak around the same field of 5 kOe, which provides evidence to the folded bands and excited UAHE by external magnetic fields. This finding elucidates the connection between the real-space non-collinear magnetism and the k-space topological electronic state and establishes a novel manner to engineer the magneto-transport behaviors of correlated electrons for future topological spintronics.

INTRODUCTION

From the perspective of applications of spintronics and future quantum devices, it is highly desirable to modulate the electronic structures using external fields and further to control the electronic transport behavior. Recently, materials with topologically non-trivial electronic states have attracted much attention. In particular, magnetic topological materials, where the transport behavior, such as the intrinsic anomalous Hall effect (AHE), anomalous Nernst effect (ANE), and quantum AHE (QAHE), are dominated by sublattice topological electronic bands near the Fermi level. Therefore, magnetic topological materials have emerged as an ideal platform for relating the transport behavior and electronic structures in the external fields.

It is known that the electronic structures can be significantly influenced by the spin configuration by breaking or preserving the mirror symmetry. Therefore, modulating the spin structure using a magnetic field is a potential strategy for realizing the modulation of topological bands. According to the existing theories and experiments, two main effects of the magnetic field modulating the topological state are the movement of Weyl nodes and the generation of a topological state. Theoretical calculations have predicted that the position of the Weyl nodes could be manipulated through magnetization rotation and Zeeman splitting during the magnetization process, resulting in varying AHEs. In addition, the mixing of electronic bands with equal or opposite spin owing to spin-orbit coupling (SOC) was proposed based on the spin-canting effect, resulting in a non-linear geometrical Hall effect. However, there is no solid experimental evidence for the studies mentioned above, and the related physical mechanism is still being developed. Therefore, further studies are essential.

EuB₆, which has a simple band structure (see supplemental information and Figures S1 and S2), was recently predicted to be a magnetic topological semimetal. According to the magnetic symmetry of the system, EuB₆ could be a nodal-line semimetal or a Weyl semimetal. Moreover, a topological phase transition across the critical temperature was confirmed using angle-resolved photoemission spectroscopy and magneto-kerr effect microscopy. The magnetization measurements showed two magnetic transitions at 15.3 and 12.5 K, corresponding to the magnetic moments along the [100] and [111] directions, respectively. The theoretically calculated magneto-crystalline anisotropy energy of EuB₆ was only 8 meV, which indicated that the orientation of the magnetic moments could be easily driven by a magnetic field. In this case, the magnetic field controlling the spin orientation in real space changed the mirror symmetry of the system and affected the topological band structures in momentum space. Therefore, the magnetic Weyl semimetal EuB₆ is a potentially ideal system for modulating the topological bands using a magnetic field.

Here, by combining transport measurements, theoretical calculations, and scanning tunneling microscopy/spectroscopy (STM/S), we present strong evidence that the magnetic field can significantly modulate the topological bands and further produce unconventional transport behavior through the field-induced spin-canting effect in magnetic Weyl semimetal EuB₆.

RESULTS AND DISCUSSION

EuB₆ crystallizes in a CaB₆ structure with a space group of 221 (Figure 1A). Two significant magnetic transition temperatures, T_C1 and T_C2, can be observed in inset of Figure 1C, which indicates that the spin orientation changes from the [100] to the [111] direction as the temperature decreases and that the easy axis is in the [111] direction (see also Figure 1A). Figure 1D shows the magnetic field dependence of isothermal M (top panel) and Hall resistivity ρ_xy (bottom panel) at 2 K. The magnetization curves for H along the [100], [110], and [111] directions exhibited weak magneto-crystalline anisotropy. The dashed lines shown in Figure 1D (bottom panels) represent the linear fit of ρ_xy above saturation field H_S. In the case of H//[100] and [111], the dashed lines did not pass through the origin point at 2 K, indicating the existence of the conventional AHE (CAHE). However, in the case of H//[110] the dashed line did pass through the origin point, meaning that the CAHE is unobvious. Interestingly, an additional component of AHE was unexpectedly observed before H_S for H//[100] and [110]. A significant Hall resistivity peak, referred to as an unconventional AHE (UAHE, ρ_UAHE), emerged before the magnetization saturation of the system. With increasing temperature, ρ_UAHE became weaker and vanished, and only the CAHE remained (see supplemental information and Figure S3).

In general, the total Hall resistivity is expressed as ρ_xy = ρ_0H + ρ_T,M + ρ_UAHE, where the three terms on the right of the equation are the normal Hall term, the conventional anomalous Hall term, and the unconventional anomalous Hall term. Based on ρ_xy measurement data, the high field part is the linear normal
Hall term $\rho_{yx}^{\text{H}} = R_0H$, and the total anomalous Hall term, which includes the CAHE and UAHE, can be extracted based on $\rho_{yx}^T = \rho_{yx} - \rho_{yx}^{\text{H}}$ as shown in Figure 1E. The CAHE and UAHE were obtained at 2 K for $H$ along the [100] and [111] directions, while only UAHE was observed for $H$ along the [110] direction (also see Figure 1D). For $H$ along [100] and [111], the conventional $\rho_{yx}^T$ was only 0.65 $\mu$cm at 2 K above $H_c$. To obtain $\rho_{yx}^U$ below $H_c$, $\rho_{yx}^T$ is defined as $\rho_{yx}^T = \rho_{yx} - \rho_{yx}^{\text{H}} - \rho_{yx}^M$, where $\rho_{yx}^M$ can be determined using the data above $H_c$ by considering $\rho_{yx}^M = 0$ and $M - M_z$ after magnetization saturation. Figure 1F shows the $\rho_{yx}^U$ after subtracting the normal and conventional anomalous Hall terms for $H$ along [100], [110], and [111]. The maximum $\rho_{yx}^U$ values were 4.4, 9.6, and 1.0 $\mu$cm at 2 K. As shown in Figure 1G, for $H$ along [100], [110], and [111], the maximum $\rho_{yx}^U$ values were 280, 976, and 120 $\Omega^{-1}$ cm$^{-1}$, respectively. In addition, the unconventional anomalous Hall angle (UAHA) was obtained using the relation $\theta_{\text{UA}}(H) = \rho_{yx}^U(H)/\rho_{yx}^T(H) + \rho_{yx}^M(H)$, as shown in Figure 1H. The maximum UAHA values were 4.4%, 9.6%, and 1% for [100], [110], and [111], respectively. The UAHE in EuB$_6$ is much larger than the CAHE and is also larger than that for many ferromagnetic (FM) systems.\textsuperscript{24–29}

The UAHE is very similar to the topological Hall effect (THE) in materials with topological spin textures in real space,\textsuperscript{30–33} in which the Dzyaloshinskii-Moriya interaction (DMI) plays an important role. However, it is considered that DMI does not exist in centrosymmetric EuB$_6$. Therefore, the UAHE in EuB$_6$ most likely results from the evolution of the Berry curvature in momentum space. A similar electronic structure analysis was also performed in the magnetic Weyl semi-metal EuCd$_2$As$_2$.\textsuperscript{34} For EuB$_6$, a very low magneto-crystalline anisotropy energy of 8 meV was reported in a previous theoretical study.\textsuperscript{29} This indicates that the competition between the magneto-crystalline anisotropy energy and the Zeeman energy gained in the magnetic field balance each other. In this case, the applied magnetic field potentially induced the non-collinear spin canting, which may further alter the topological bands and electronic transport properties of EuB$_6$. It is also different from the CAHE, which depends on the unchanged magnetism and the electronic structures (Berry curvature). During the magnetizing process, the reverse magnetic domain decreases, and the AHE increases. After the magnetization saturation, both the electronic structures and the CAHE remain unchanged. For the UAHE in EuB$_6$, in contrast, the spin canting was generated just during the magnetization re-orientation process, which results in an AHE peak with the evolution of topological bands and Berry curvature. collinear magnetism on the electronic structures. Starting from the 2-directed FM order, we added in-plane spin components ($\mu_y$) to different Eu atoms (marked in Figure 2C), thereby doubling the size of unit cells along all three lattice vectors. The energy difference was 8 meV when the canting angle $\theta$ was 45° ($\tan \theta = \mu_y/\mu_x$), which indicates that the spin orientations can be easily tuned using external fields. Because of the band-folding effect, the band inversions (consisting of $\pi$ and Eu-d orbits; see supplemental information and Figure S2) appearing at the high-symmetry k-points X, Y, and Z folded to the $\Gamma$ point. When the spin vector components became zero and the system was FM, 2-fold degenerate bands existed around the $\Gamma$ point (marked in Figure 2A) because of the band folding owing to the equivalence of the x and y directions. Upon increasing the canted spin components, the degenerated bands split, and all crossings along the $\Gamma$-Z path become gapped under SOC, as shown in Figures 2D and 2E. Although the energy scale of the spin-canting-induced band splitting was small (less than 20 meV at the $\Gamma$ point), the bands changed considerably upon coupling with the previous single degenerate band (green regions in Figures 2D and 2E), leading to an enhancement in the Berry curvature in the coupled area. The Berry curvature can be calculated directly using the linear response Kubo formula.\textsuperscript{35} We calculated the Berry curvature $\Omega_{xy}$ along the high-symmetry $\Gamma$-Z path, on which $\Omega_{xy}$ and $\Omega_{yz}$ always have the same value because of the $\text{C}_4v$ symmetry. Each peak in the Berry curvature spectrum corresponds to the band-crossing regime of the band structure. When the canting angle was zero, the bands arising from the high-symmetry k-points X and Y (doubly degenerate bands) did not couple with the bands from the Z point (single degenerate band) (Figure 2F). Owing to the spin-canting effect, these bands couple and open gaps (Figure 2H), which induce two additional peaks in the Berry curvature spectrum (left in Figure 2G). When $H$ was smaller than $H_c$, the electronic band structure could be modulated by this behavior of the non-collinear magnetism. The folding of the electronic band structure thus resulted in the UAHE during the magnetization process. However, when $H$ was larger than $H_c$, all moments followed the direction of the external magnetic field, and both the spin-canting effect and the UAHE disappeared, and only the CAHE remained owing to collinear FM. Furthermore, when $H$ exceeded $H_c$ at 2 K, the conventional AHCs measured in our study were only 24 and 89 $\mu$cm$^{-1}$ for [100] and [111], respectively (see supplemental information and Figure S5), which are consistent with the theoretical calculations for the collinear FM state.\textsuperscript{20}
We further extracted the field-dependent intensity of the spectra in Figure 3C and plotted the intensity values in a series of energies as a function of the magnetic field in Figure 3D. For all energy cuts, the intensity of the di/dv spectrum showed an initial increase, peaked at approximately 5 kOe, and then gradually decreased as the field reached 10 kOe. This observation is reminiscent of the transport measurements (Figures 1D–1H), where the UAHE demonstrates a highly concurrent field dependence, with an exact same maximum at 5 kOe. The spin-canting effect causes the band folding and changes the DOS near the Fermi level (Figures 2D, 2E, 3C, and 3D), which further leads to the UAHE. We plotted the intensity map (Figure 3E) of the negative second derivative of the spectra in Figure 3C, and the field-dependent STS features could be observed more clearly. The substantial enhancement of negative second derivative during the magnetization indicates a change in LDOS of EuB₆ with the application of external magnetic field.

To provide a clear physical picture, a schematic of the modulation of the electronic band structure and transverse electronic transport characteristics of the magnetic Weyl semimetal EuB₆ during the entire magnetization process is shown in Figure 4. According to the magnetic measurement results, the ground-state magnetic moment of EuB₆ aligns with the [111] easy axis (Figures 4A and 4B). For H//[111], there are no spin-canting and band-folding effects during the entire magnetization process. Therefore, it only shows the CAHE upon the unchanged topological bands, as shown in the top part of Figures 4B–4E. As long as the moments deviate from the easy axis to the hard axis, the competition between the magneto-crystalline anisotropy and the external magnetic field leads to the spin canting, which results in a band-folding effect in momentum space, leading to splitting of the pristine bands and production of additional bands, as shown by the red color in the middle and bottom regions of Figure 4C. With the aid of split bands and SOC-induced anti-crossings, an enhancement of the Berry curvature can be obtained. For H along the [100] and [110] main directions, the enhanced Berry curvature leads to the large UAHE during the magnetization process. Once the system enters the saturation state of magnetization, both spin canting and band folding disappear again, as shown in the middle and bottom regions of Figure 4D. Therefore, the UAHE vanishes, and the CAHE produced by the linear FM state remains, as shown in Figure 4E. This implies that as long as the magnetic field deviates from the [111] easy axis in EuB₆, a dynamic picture of spin canting, band folding, and the UAHE during the process of magnetization and demagnetization is very likely to be observed.

In current studies, the peak of the Hall sign during the magnetization process was widely considered to be THE, attributed to the topological spin structures in real space. In our study, we observed band splitting and LDOS evolution, resulting in a prominent UAHE. An external field can change the magnetic structure and further modulate the topological electronic structures, revealing an interplay between the magnetism and topology. The physical origin of the observed Hall peaks during the magnetization process can also be ascribed to the alteration of topological electronic structure in momentum space, not only the topological spin structure in real space.

**CONCLUSION**

In summary, the topological electronic structure was modulated owing to the spin-canting effect in magnetic Weyl semimetal EuB₆ during the magnetization re-orientation with the application of a magnetic field. An emergent UAHE was observed with a maximum UAHC of approximately 1,000 Ω⁻¹ cm⁻¹ and a UAH of approximately 10% at a low field of 5 kOe. Combining the experiments and theory, we present strong evidence that the magnetic field significantly modulates the topological electronic state and the unconventional transport behavior. Our findings provide not only an insight into the relationship between the magnetic and electronic structures but also guidance for manipulating the electronic transport properties through the external magnetic field, particularly in emergent magnetic correlated electronic materials for topological spintronics.

**MATERIALS AND METHODS**

**Sample preparation and physical property measurements**

EuB₆ single crystals were prepared using the flux method. The ingredients with a ratio of Eu:B:Al = 1:8:100 were placed in an alumina crucible and heated to 1,773 K in a furnace under an atmosphere of high-purity argon. Subsequently, the furnace was slowly cooled to 1,573 K.
at a rate of 2 K/h and then rapidly cooled to room temperature at a rate of 200 K/h. Single-crystalline samples were obtained and then washed with sodium hydroxide solution and deionized water and present with a black cubic shape (see inset in Figure 1B). The crystal structure of single-crystal samples was characterized by X-ray diffraction (XRD). Room temperature XRD patterns of EuB$_6$ show only the (h00) and (hk0) Bragg peaks, which indicate that the two exposed surfaces are the (100) and (110) planes, as shown in Figure 1B. Magnetic properties were measured by a superconducting quantum interference device (SQUID) magnetometer. Electric transport properties were measured by the physical property measurement system (PPMS).

**STM/S measurements**

EuB$_6$ single crystals were mounted on the metal sample holders by conductive epoxy. The crystals were mechanically cleaved under 12 K and then immediately transferred to the STM scanner (Unisoku). STM/S measurements were performed under 0.45 K. The differential conductance (dI/dV) spectra were obtained with a standard lock-in amplifier at a frequency of 973.0 Hz, with a modulation voltage of 0.2 mV. Magnetic fields were applied perpendicular to the samples.

**First-principles calculations**

First-principles calculations were performed with the Vienna ab-initio simulation package (VASP) based on the density functional theory (DFT) with the projector augmented wave (PAW) method. The exchange-correlation potential is treated using the generalized gradient approximation (GGA) parametrized by Perdew, Burke, and Ernzerhof (PBE). The rotationally invariant on-site coulomb interactions on 4f electrons are taken to be 8 eV. For the non-collinear magnetic calculations, the 400 eV energy cutoff and the 4 x 4 x 4 $\Gamma$-centered k-mesh are adopted in the self-consistent calculations. The momentum-dependent Berry curvature is calculated according to the following Kubo formula:

$$\Omega_{nab} = -2 im \sum_{\mathbf{k}} \frac{\text{Im} \langle \mathbf{u}_m(k) | \mathbf{u}_n(k') \rangle}{\epsilon_n(k') - \epsilon_m(k)}$$

The velocity operators are calculated by using the PAW wave functions as proposed in Song et al. The Berry curvature calculations, we take 280 valence bands and 220 conduction bands to ensure the convergence.
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ACKNOWLEDGMENTS
This work was supported by the National Key R&D Program of China (nos. 2022YFA1403800, 2022YFA1403400, and 2019YFA0704900); the Fundamental Science Center of the National Natural Science Foundation of China (no. 52088101); the Synergistic Extreme Condition User Facility (SECUF); the Beijing National Science Foundation (no. Z190009); the National Natural Science Foundation of China (nos. 11974394, 12174426, 12104280, and 12004416); the Strategic Priority Research Program (B) of the Chinese Academy of Sciences (CAS) (XDB33000000); the Key Research Program of CAS (no. ZDRW-CN-2021-3); the CAS Project for Young Scientists in Basic Research (YSSB-003); the Scientific Instrument Developing Project of CAS (no. ZDKY-YQ2021003); and the Basic Research Plan of Shanxi Province (no. 20210302124160).

AUTHOR CONTRIBUTIONS
E.L. designed research; J.S., S.Z., and J.Y. performed transport measurements. J.G. and Z.W. carried out DFT calculations. C.Y. and Y.S. grew crystals. M.L. and G.L. performed STM/AFM measurements. Z.W. carried out DFT calculations. C.Y. and Y.S. grew crystals. M.L. and G.L. performed STM/AFM measurements.

DECLARATION OF INTERESTS
The authors declare no competing interests.

SUPPLEMENTAL INFORMATION
It can be found online at https://doi.org/10.1016/j.xinn.2023.100399.

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