On the anomalous low-resistance state and exceptional Hall component in hard-magnetic Weyl nanoflakes

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Received March 23, 2021; accepted May 14, 2021; published online July 1, 2021

Magnetic topological materials, which combine magnetism and topology, are expected to host emerging topological states and exotic quantum phenomena. In this study, with the aid of greatly enhanced coercive fields in high-quality nanoflakes of the magnetic Weyl semimetal Co$_3$Sn$_2$S$_2$, we investigate anomalous electronic transport properties that are difficult to reveal in bulk Co$_3$Sn$_2$S$_2$ or other magnetic materials. When the magnetization is antiparallel to the applied magnetic field, the low longitudinal resistance state occurs, which is in sharp contrast to the high resistance state for the parallel case. Meanwhile, an exceptional Hall component that can be up to three times larger than conventional anomalous Hall resistivity is also observed for transverse transport. These anomalous transport behaviors can be further understood by considering nonlinear magnetic textures and the chiral magnetic field associated with Weyl fermions, extending the longitudinal and transverse transport physics and providing novel degrees of freedom in the spintronic applications of emerging topological magnets.

hard-magnetic material, magnetic Weyl semimetal, magnetoresistance, Hall effect

PACS number(s): 81.40.Rs, 85.70.Kh, 73.43.Qt

Citation: Q. Q. Zeng, G. X. Gu, G. Shi, J. L. Shen, B. Ding, S. Zhang, X. K. Xi, C. Felser, Y. Q. Li, and E. K. Liu, On the anomalous low-resistance state and exceptional Hall component in hard-magnetic Weyl nanoflakes, Sci. China-Phys. Mech. Astron. 64, 287512 (2021), https://doi.org/10.1007/s11433-021-1715-4

1 Introduction

Recently, the first magnetic Weyl semimetal Co$_3$Sn$_2$S$_2$ was discovered [1-6], which has significantly improved the understanding of the interplay of magnetic order and topological physics after the experimental realization of the quantum anomalous Hall effect (QAHE) [7]. Co$_3$Sn$_2$S$_2$ is a shandite compound with Weyl nodes at ~60 meV above the Fermi level and shows semi-metallicity with small Fermi surfaces and low carrier concentration. Transport properties are dominated by the electrons near Fermi energy. As the topological features in this system are sufficiently close to the Fermi energy and there are few trivial bands, it is promising to observe the transport behavior dominated by topological band structures. In Co$_3$Sn$_2$S$_2$ bulk single crystals, large intrinsic anomalous Hall conductivity and anomalous Hall angle have been detected [1,2]. In the two-dimensional (2D) limit, theoretical studies have predicted that the desired high-temperature QAHE could be realized [8-10], which has been indicated experimentally by the observation of chiral edge states in a recent STM measurement [11]. In addition,
regardless of the specific types of magnetic domain walls, there may exist a large magnetoresistance (MR) that can be retained against the finite strength of the disorder in Co₃Sn₂S₂ [10].

The application prospects of Co₃Sn₂S₂ make it a promising candidate for practical applications of topological materials in spintronic or advanced electronic devices. For topology-related spintronic applications and the realization of QAHE, the growth and physical study of Co₃Sn₂S₂ films are highly desired. Epitaxial polycrystalline Co₃Sn₂S₂ films can be grown via molecular beam epitaxy and co-sputtering deposition methods [12,13]. Moreover, reports on Co₃Sn₂S₂ nanoflakes grown via the chemical vapor transport (CVT) method exist [14,15]. Out-of-plane ferromagnetic order and large anomalous Hall effects were observed. These studies offer important indications for further studies of low-dimensional magnetic Weyl semimetals. In addition to studies on possible QAHE and its practical applications, the unexplored properties of this magnetic Weyl low-dimensional system are important. Anomalous transport properties in Co₃Sn₂S₂ nanoflakes have been observed in our previous work [14]. However, these intriguing behaviors still need to be studied and analyzed in detail.

In this work, anomalous transport properties are studied in high-quality Co₃Sn₂S₂ single-crystalline nanoflakes with 30-100-nm thickness. The system clearly enters a lower electrical resistance state once the external magnetic field is antiparallel to the magnetization. Furthermore, an exceptional Hall component besides the normal and anomalous Hall effects is observed concurrently. Possible magnetic textures induced by an antiparallel magnetic field are possibly responsible for the anomalous transport behaviors, as the interaction between Weyl electrons and magnetic textures can result in local states and conductive modes [16-20]. This study will attract interest in the interplay of magnetic order and topological transport behavior and offer a platform for magnetic topological material-based applications.

2 Material and methods

Single-crystalline Co₃Sn₂S₂ nanoflakes were grown via CVT using Co₃Sn₂S₂ polycrystalline powder as the precursor. The morphology and thickness were investigated using atomic force microscopy. Hall bar pattern with a long edge along the a axis of Co₃Sn₂S₂ for out-of-plane transport measurement was fabricated using microfabrication technology (Supporting Information Note 1). Transport data (temperature dependent resistivity in Supporting Information Note 2, for example) were measured using a Janis system and collected by employing lock-in amplifier technology. Symmetric and antisymmetric processes were performed on the longitudinal and transverse resistivity, respectively (details in Supporting Information Note 3).

3 Results and discussion

The morphology and thickness of a single-crystalline nanoflake are shown in Figure 1(a) (more details are given in Supporting Information Note 1). Samples of thicknesses 30, 41, 71, and 94 nm were studied in this work. Figure 1(b) shows one of the Hall bar patterns for transport measurements. The Curie temperatures of the two selected samples were determined as 181 K through the kink temperature in the temperature-dependent resistivity curve (Supporting Information Note 2). The single-crystalline Co₃Sn₂S₂ nanoflakes being studied show consistent basic physical properties with bulk single crystals [1,21] (Supporting Information Note 4). Furthermore, the nanoflakes show high quality with a residual resistivity ratio of ~20, an anomalous Hall conductivity of ~1000 Ω⁻¹ m⁻¹, and a large coercive field as high as 5.5 T.

Figure 1(c)-(j) show the magnetic field dependence of the longitudinal and transverse resistivity measured at various temperatures for the 30-nm-thick sample (data of other samples are shown in Supporting Information Note 4). The longitudinal data (Figure 1(c)-(f)) show positive parabolic-like MR at all measured temperatures. Apart from the ordinary MR, hysteresis behavior exists where the data with decreasing and increasing fields are not coincident between zero and coercive fields (μ0Hc). Thus, the value of the longitudinal resistivity depends on the magnetization history, being lower (50 K) or higher (1.6 K) for the demagnetization compared with the magnetization process.

As shown in Figure 1(g)-(j), the Hall data exhibit hysteresis loop behavior like the shape of the magnetic field dependence of magnetization in a hard-ferromagnetic material. The anomalous Hall resistivity, in addition to the normal Hall effect, is detected. The loop behavior and the anomalous Hall effect like that of the bulk sample [1] suggest that the easy axis of Co₃Sn₂S₂ single-crystalline nanoflake is still along the c axis. Clear nonlinear normal Hall curves are observed at low temperatures, reflecting that both electron and hole carriers contribute to the transport properties in Co₃Sn₂S₂ single-crystalline nanoflakes. We performed two-band [1,22,23] analyses on all measured samples. The carrier concentration of holes and electrons are both ~1×10²⁰ cm⁻³ in all studied samples (details in Supporting Information Note 4), indicating a near compensation of carriers in Co₃Sn₂S₂ single-crystalline nanoflakes with the above-mentioned different thicknesses. The abovementioned results reveal the consistent carrier properties of Co₃Sn₂S₂ single-crystalline nanoflake and bulk samples [1,14,15].

The additional MR peak or dip emerging in the background of the parabolic-like MR (the curves without magnetization reversal, i.e., from ±9 T to zero fields) was extracted and denoted as Δρₓₓ. At 50 K, when the field decreased from ±9 T to zero and then increased in the opposite
direction, a linear field-dependent positive $\Delta \rho_{xx}$ appeared immediately and became zero instantaneously after the magnetization reversal (Figure 2(a)). However, a negative linear $\Delta \rho_{xx}$ was unexpectedly observed at 1.6 K when the applied field approached the coercive field (Figure 2(b)). We further analyzed the emerging additional MR when magnetization was antiparallel to the applied magnetic field. In a widely accepted case, a system will be more magnetically ordered when the magnetic field is much higher than $\mu_0 H_c$. Between zero and coercive fields, the magnetic field gradually turns the antiparallel moments into parallel moments. Thus, the antiparallel moments become more disordered as the field increases before the magnetization reversal. Hence, the spin-dependent scattering [7,24,25] would be strong, increasing the longitudinal resistivity. Therefore, positive $\Delta \rho_{xx}$, like the behavior in Figure 2(a), is normally observed in magnetic systems [7,26]. The difference from the common case is that the positive $\Delta \rho_{xx}$ in the studied sample appears immediately when the antiparallel magnetic field starts increasing. More widely observed positive MR peaks appear only near the coercive field [27-29]. The linear magnetic field dependence is also a unique and unexplained phenomenon.

Interestingly, a contrary situation emerges when the system goes to low temperatures. The longitudinal resistivity in the case when the applied field is antiparallel to the magnetization (antiparallel configuration) is even lower than that of the parallel case (Figure 2(b)), i.e., $\Delta \rho_{xx}$ is negative. This behavior has been previously observed in our 180-nm-thick Co$_3$Sn$_2$S$_2$ single-crystalline nanoflakes [14] and can also be recognized in polycrystalline films (although the spin-related positive MR is mixed in) [13]. Evidently, the trend is a universal behavior in this low-dimensional Weyl system. Nevertheless, negative $\Delta \rho_{xx}$ has scarcely been observed among common magnetic materials or magnetic functional films in the past. A distinct mechanism resulting in a low-resistance state against the antiparallel external field may emerge in this magnetic topological system.

In magnetic information storage technology, the giant MR effect can be produced by switching the parallel and antiparallel directions of ferromagnetic and antiferromagnetic layers in magnetic tunnel junctions, which use different electrical resistance states between parallel and antiparallel cases [30-32]. The anomalous negative MR effect observed
Similarly, the effect exists with increasing applied field within the coercive field (the lower panel of Figure 3). Evidently, the 30-nm-thick sample. The arrows indicate the direction of field variation. For simplicity, two parts, we assume that one part, called the low-temperature part (low-T), occurs only below the kink in Δρxx to ρxx on the parabolic-like MR background at μ0Hc (dark blue dots) and separate contributions (green and orange symbols).

Figure 2 (Color online) (a), (b) Parabolic-like MR-subtracted magnetic field dependence of the longitudinal resistivity (Δρxx) at 50 and 1.6 K for the 30-nm-thick sample. The arrows indicate the direction of field variation. (c) ρxx difference before and after the jump at μ0Hc (Δρxx(μ0Hc)). (d) Ratio of Δρxx to ρxx on the parabolic-like MR background at μ0Hc (dark blue dots) and separate contributions (green and orange symbols).

in this magnetic nanoflake may provide a novel degree of freedom for spintronic applications.

Figure 2(c) and (d) show Δρxx at μ0Hc (Δρxx(μ0Hc)) and the Δρxx(μ0Hc)-to-ρxx(μ0Hc) ratio on the background of the parabolic-like MR (dark blue dots). Above and below 30 K, Δρxx(μ0Hc) is positive and negative, respectively. As a kink of Δρxx(μ0Hc) exists around 100 K, we considered the existence of competing components leading to different field dependences of longitudinal resistivity and their dominant position variation with temperature. For simplicity, two factors are currently assumed in this system. To separate these two parts, we assume that one part, called the low-temperature part (low-T part), occurs only below the kink temperature in Δρxx(μ0Hc). The other part is the high-temperature part (high-T part), which is fitted using the data just above 100 K. By adopting the form of y = Ae−x+b+c, the experimental data (dark blue dots) can be well fitted (green square symbols) above 80 K (see Supporting Information Note 5 for details). The low-T part is then extracted by subtracting the high-T part from the experimental results, denoted by orange triangle symbols in Figure 2(d). The current straightforward but effective fitting indicates that the high-T part contributes to longitudinal resistivity continuously below the Curie temperature. By contrast, the low-T part appears only below 70 K. Here, we stress that the low-resistance state can be observed directly from the original MR data without separation (Figure 1(e) and (f)). Thus, the current separation will not affect the main results.

We next analyze the transverse transport behavior in Co3Sn2S2 single-crystalline nanoflakes. Conventional magnetic materials usually show both normal and anomalous Hall effects. For a hard-magnetic material with coercivity and sharp magnetization reversal, the typical Hall curve is shown in the upper panel of Figure 3(a). We denote the Hall resistivity curve from +9 T to −9 T as ρyx+. Similarly, the curve from −9 T to +9 T is denoted as ρyx−. Evidently, ρyx−−ρyx+ maintains a stable plateau (two times as anomalous Hall resistivity (ρAH)) within the coercive field (the lower panel of Figure 3(a)). For Co3Sn2S2 nanoflakes, the difference between the Hall data with different directions of the magnetic field variation is shown in Figure 3(b). In sharp contrast, an increasing ρyx−−ρyx+ effect exists with increasing applied fields up to the coercive field, which is quite different from the conventional case.

The corresponding Hall loop, subtracting the normal Hall component, is depicted in Figure 3(c). In a conventional hard-magnetic material, the normal Hall-subtracted Hall curve should be a square loop [7], indicated by the red-dashed lines in Figure 3(c). However, unexpected Hall contribution (highlighted by light-blue-colored filling, denoted as exceptional Hall resistivity Δρyx in this text) exists except normal and anomalous Hall components in the current system.

Figure 3(d) shows the Δρyx at different temperatures in the negative field region. The Δρyx appears from the zero-field and increases noticeably with increasing antiparallel field up to the coercivity. Once the magnetization and applied field are parallel to each other after the magnetization reversal at the coercive field, the exceptional Hall resistivity suddenly drops to zero. Compared with the bulk case having a small coercive field (~0.5 T) [1], the enhanced coercivity in Co3Sn2S2 nanoflakes makes it possible to clearly observe Δρyx in a wide antiparallel field range. By further adopting the function form of Δρyx≈aB3−Δρyx is found to evolve from a ~B3 law at 50 K to a stable ~B3 law below 30 K (Figure 3(e)).

Evidently, Δρyx emerges around 70 K and becomes more notable at lower temperatures. A large value of 1.5 μΩ cm is observed at 1.6 K. Because the low-T negative MR (Figure 2(d)) also appears around 70 K, we conclude that there should be connections between anomalous longitudinal and transverse transport behaviors.

Figure 3(f) shows Δρyx at μ0Hc and its ratio to ρAH. Evidently, the exceptional Hall component can be more than 2.5 times larger than ρAH, indicating a notable effect driven by a distinct mechanism in Co3Sn2S2 nanoflakes at low temperatures.

Figure 4(a) and (b) summarize the anomalous transport behaviors of four different thick samples. The Hall conductivity analysis can be found in Supporting Information.
All samples show similar temperature-dependent transport behaviors, exhibiting anomalous negative MR and exceptional Hall resistivity below 70 K. According to the data, no obvious thickness dependence exists for these anomalous transport behaviors in the current thickness range. In addition, the 41-nm-thick sample shows smaller negative MR and exceptional Hall values compared with other samples. Further, it shows lower mobility (Supporting Information Note 4), possibly attributed to the deviation of sample quality.

Next, we analyze the underlying origin of anomalous transport behaviors. The longitudinal resistance states for a certain magnetic field value are different during field-decreasing and -increasing processes. Thus, the anomalous transport behaviors could not result from the samples’ basic properties (e.g., carrier mobility), which may vary with the magnetic field. The two-band analysis (Figure S5(c)) also indicates additional components apart from the normal and anomalous Hall conductivities, emerging only under an antiparallel magnetic field. Considering the coexistence of negative MR and exceptional Hall effect, these two effects may simultaneously originate from a specific physical state that emerges under an antiparallel magnetic field at low temperatures. It has been reported that nontrivial topological magnetic textures may emerge in a magnetic Weyl system [19,33-35]. Once the magnetic textures (even without topological property) are formed, they will interact with the Weyl conduction electrons and affect the transport behavior in a system through an emergent chiral magnetic field [16-20]. The fictitious magnetic field may be reminiscent of the real-space Berry curvature in conventional materials with trivial band structures. However, the chiral magnetic field shows a richer degree of freedom as the related chiral gauge potential is closely linked to the Weyl nature and depends on the direction of the background magnetization itself [19,20].
Theoretical calculations show that there could be bound states and potential current, which are highly localized around a magnetic domain wall or texture [16,17,20]. In experiments, Co₃Sn₂S₂ bulk samples indeed showed other possible magnetic interactions besides the out-of-plane ferromagnetic interaction, and thus, nonuniform magnetization behavior below the Curie temperature [1,21,36,37]. A round-shaped domain structure has also been observed in the Co₃Sn₂S₂ system [38]. Based on the above theoretical and experimental reports, a promising possibility responsible for the anomalous negative MR and exceptional Hall effect in Co₃Sn₂S₂ nanoﬂakes is the noncollinear magnetic structure. Based on our experiment results, we consider it the magnetic texture induced by the antiparallel magnetic field.

Figure 4(c) and (d) show schematic illustrations of the magnetic structure when an external magnetic field is antiparallel to the magnetization. As the magnetization reversal process is sharp, we consider the antiparallel magnetic domains formed close to the coercivity sweeping out the entire sample immediately. Hence, only a slight tilt of moments occurs when the applied field is less than the coercivity. Moments may tilt randomly owing to the antiparallel magnetic field perturbation at temperatures higher than 70 K (Figure 4(c)), while the ordered magnetic textures (sketched as red patterns in Figure 4(d)) may emerge at low temperatures due to the competition of ferromagnetic and other magnetic interactions.

Once the abovementioned potential magnetic textures are induced by the antiparallel magnetic field, a Weyl-fermion-correlated chiral gauge field \( b_\text{ex}(\mathbf{r}) = \nabla \times \frac{J_{\text{ex}}}{V_F} M(\mathbf{r}) \) [16-20] (where \( J_{\text{ex}} \) is the exchange interaction strength between localized magnetic moments and Weyl electrons, and \( V_F \) is the Fermi velocity) emerges around the magnetic textures. Furthermore, this localized chiral gauge field may lead to bound states near the textures [16,17,20]. When an electric field is applied to the sample, potential bound states can also offer emergent conductive modes in this system. As anomalous transport behaviors exist in both longitudinal and transverse channels in Co₃Sn₂S₂ nanoﬂakes, a varying \( M(\mathbf{r}) \) with a complicated order (not varied within parallel planes, schematic sketched in Figure 4(d)) may occur, leading to \( j_\text{Hall} \) with both longitudinal (corresponding to low-resistance state) and transverse (corresponding to exceptional Hall effect) components. The speciﬁc structure of the possible magnetic textures in Co₃Sn₂S₂ single-crystalline nanoﬂakes and the conﬁrmation of its inﬂuence on the transport behaviors require further study.

4 Conclusions

Herein, an anomalous low electrical resistance state and exceptional Hall component are reported in high-quality single-crystalline nanoﬂakes of magnetic Weyl semimetal Co₃Sn₂S₂ once the magnetization is antiparallel to external magnetic fields. Possible antiparallel ﬁeld-induced magnetic textures and Weyl-fermion-associated chiral magnetic fields are expected to account for the decrease and increase in the longitudinal and transverse resistivities, respectively, through the possible additional conductive modes. Our report will draw more interest in the interplay of magnetism and topological transport phenomena, offering new elementary states for the applications of magnetic topological materials.

This work was supported by the National Natural Science Foundation of China (Grant Nos. 520888101, and 11974394), the National Key R&D Program of China (Grant No. 2019YFE0704900), the Beijing Natural Science Foundation (Grant No. Z190009), the Strategic Priority Research Program (B) of the Chinese Academy of Sciences (CAS) (Grant No. XDB33000000), the Scientiﬁc Instrument Developing Project of CAS (Grant No. ZDKYYQ20216003), Users with Excellence Program of Hefei Science Center CAS (Grant No. 2019HSC-UE009), and the Youth Innovation Promotion Association of CAS (Grant No. 2013002). Shu Zhang is supported by the National Science Foundation, United States (Grant No. DMR-1742928).

Supporting Information

The supporting information is available online at phys.scichina.com and link.springer.com. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientiﬁc accuracy and content remains entirely with the authors.

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