The depinning of magnetic domain walls and the magnetization anomaly within the ferromagnetic phase of the Weyl semimetal Co$_3$Sn$_2$S$_2$

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We report bulk magnetization measurements and spatially resolved measurements of magnetic domains in Co$_3$Sn$_2$S$_2$ single crystals. The results indicate that a previously reported magnetic anomaly around 130 K is due to domain wall pinning and depinning. Our measurements also reveal a hysteresis between field-cooled-cooling (FCC) and field-cooled-warming (FCW) magnetization curves acquired under a constant magnetic field below 300 Oe. This observation rules out the possibility that the anomaly stems from a second-order AFM-FM phase transition. Our results further suggest that changes in the shape of hysteresis loops from 5 K to 170 K are caused by an unusual temperature-dependent domain nucleation field that changes sign around 130 K. The Kerr rotation images of the magnetic domains obtained between 120 K and 140 K support the notion that a domain wall pinning effect exists near 130 K.

INTRODUCTION

Weyl Semimetals (WSM) are material systems with non-degenerate bands that cross each other near the Fermi level when inversion symmetry or time-reversal symmetry is broken [1, 2]. Co$_3$Sn$_2$S$_2$ is a ferromagnetic material with a Curie temperature of $T_C = 174$ K [3], and was proposed to be a potential WSM [5]. Co$_3$Sn$_2$S$_2$ crystallizes in a hexagonal lattice with triangular S and Sn layers interspersed between the kagome lattice planes of Co. A recent ARPES study reveals large band shifts with the onset of magnetism due to the cluster effects of Co [6]. The topological features of this material lead to a large anomalous Hall conductivity [7], an anomalous Nernst Hall effect [8], a negative magnetoresistance due to a chiral anomaly [5], and an unusual breaking-down of Ohm’s law [9]. In addition, an unusually large magneto-optic effect associated with the anomalous Hall effect has also been observed [10].

Recently reported studies suggest that there may exist a hidden magnetic phase in Co$_3$Sn$_2$S$_2$ around 130 K [11, 12], such as revealed in AC susceptibility measurements of whole single crystal samples with low applied field [13]. A muon diffraction study revealed both a primary precession at 5 K and a secondary precession below $T_C$, which was interpreted as two different magnetic structures. The primary structure is the ferromagnetic phase (FM) while the secondary structure is suggested as a possible in-plane antiferromagnetic phase (AFM) [14]. The neutron diffraction study by Soh et al., however, rules out the existence of an antiferromagnetic structure [15]. A recent half-polarized neutron diffraction investigation reveals that there is a local symmetry broken below $T_C$, where the kagome lattice starts to distort [16]. Based on an AC magneto-optic Kerr effect (MOKE) study, Lee et al. suggest that the anomaly around 130 K in the FCC magnetization is likely due to a transition between linear domain walls and elliptical domain walls [17].

In this context, we report bulk magnetization measurements on Co$_3$Sn$_2$S$_2$ single crystals in external magnetic fields applied along the $c$ axis. We also present magneto-optic Kerr effect (MOKE) images of magnetic domains in Co$_3$Sn$_2$S$_2$ from 120 to 140 K obtained under an external magnetic field of 94 Oe applied along the $c$ axis. We extract fractions of single domains and domain walls near the magnetic anomaly around 130 K. We argue that the magnetic anomaly can be explained by a domain wall pinning-and-depinning effect. The origin of the domain wall pinning and depinning effect remains to be further investigated.

METHODS

Single Crystal Growth

Single crystals of Co$_3$Sn$_2$S$_2$ were synthesized by solution growth [9, 15, 16]. A ternary mixture with initial composition Co$_{12}$Sn$_8$S$_{40}$ was first heated to 400°C over two hours and held there for another two hours. It was then heated to 1050°C over six hours and held there for 10 hours, followed by a slow, 90 hour cool down to 740°C. The remaining molten flux was removed by centrifugation. Shiny hexagonal crystals were obtained. The powder x-ray diffraction data refines to a hexagonal unit cell with $a = 5.3641(8)$ Å and $c = 13.1724$ Å, which are consistent with previously reported values [20].

Magnetization measurements and magnetic domain imaging

Magnetization measurements of Co$_3$Sn$_2$S$_2$ single crystals were performed in a Quantum Design MPMS SQUID up to 7 T with the magnetic field parallel to the $c$ axis. Magnetization as a function of temperature was measured under various applied fields in both field-cooled-
cooling (FCC) and field-cooled-warming (FCW) conditions. Hysteresis loops were measured by first cooling down to the desired temperature in zero applied field and then sweeping the field. Before each measurement, we followed a demagnetization procedure to minimize the remanent field in the magnet and in the sample chamber by systematically oscillating the magnet to zero field at room temperature.

Polar MOKE images are acquired from as-grown Co$_3$Sn$_2$S$_2$(0001) single crystal samples, using a normal-incidence Sagnac interferometric scanning microscope [21]. The microscope measures Kerr rotation as opposed to Kerr ellipticity [10]. It has a spatial resolution of 0.85 µm and a sensitivity of 4.4 µrad. The single crystal sample is mounted on a cold finger inside a cryostat. The sample is optically accessible from outside through an optical window. The temperature of the sample is variable from 300 K to 5 K. We use a permanent magnet placed outside of the cryostat to produce a variable magnetic field of up to 100 Oe (0.01 T) along the c axis (the surface normal) of the sample. For this experiment, the field is held constant at 94 Oe, measured by a Hall effect sensor.

RESULTS AND DISCUSSION

Magnetization measurements

The FCC curve with a magnetic field of 1000 Oe parallel to the c axis is shown in Fig. 1a and illustrates the ferromagnetic-paramagnetic transition. The inverse susceptibility is fit to the Curie-Weiss law and we obtain an effective moment of 1.14 µ$_B$ per Co and a Curie-Weiss temperature around $\theta_{CW} = 175$ K, which are consistent with a previously published work [15]. There is no difference in the FCC and FCW magnetization at fields above 300 Oe. If the applied field is below 300 Oe, however, two anomalies emerge. First, the FCC and FCW curves start to deviate near $T_C$. Second, a local maximum arises at $T_A$ near 130 K in the FCC data, which is also reported in other studies [14][22]. It disappears above 300 Oe and below 50 Oe, while a small hysteresis between FCC and FCW still remains at 50 Oe. The field dependence of $T_A$ is relatively small, as shown in the inset of Fig. 1b. A local minimum also emerges in the FCW magnetization albeit at higher temperature. As we will show later, this local minimum originates from the magnetic domain wall depinning upon warming, which is relatively common since depinning is enabled by increasing thermal energy. On the other hand, the depinning upon cooling observed in the FCC at $T_A$ is anomalous, and the focus of this study. The hysteresis between FCC and FCW magnetizations indicates it is unlikely that the local maximum and minimum in the FCC and FCW magnetization are due to a second order phase transition, which should not depend on magnetic history.

Hysteresis loops at various temperatures are shown in Fig. 2. The spontaneous magnetization, $M_s$, is about 0.3 µ$_B$/Co at 5 K [7][20]. We note that the hysteresis loop is slightly off centered about zero field at 5 K with the negative coercive field $H_c^- = -0.415(5)$ T and the positive coercive field $H_c^+ = 0.405(5)$ T. The asymmetry of the hysteresis loop, depending on the magnetic history, can be attributed to Exchange Bias (EB), which is a result of interaction between a FM and another ordered magnetic phase, typically an AFM state [24]. We observe that the bias field with field up to 7 T is smaller than the one previously observed with smaller applied field [23][25]. Besides EB, Fig. 2 also shows that increasing temperature causes the shape of the hysteresis loop to change from a rectangular shape, to a triangular-tailed rectangle, to a bi-triangular shape, until the hysteresis

FIG. 1. a) Magnetization as a function of temperature measured under FCC with 1000 Oe applied field parallel to the c axis. The blue curve shows a Curie-Weiss fitting with an effective moment 1.14 µ$_B$ per Co. The inset shows a Co$_3$Sn$_2$S$_2$ single crystal grown from solution growth. b) FCC and FCW curves show a discrepancy between 100 K and near $T_A$. $T_A$ is the temperature of the magnetic anomaly where the FCC magnetization reaches a local maximum followed by a downward kink. The inset shows the field dependence of $T_A$. 

FIG. 2. The spontaneous magnetization, $M_s$, is about 0.3 µ$_B$/Co at 5 K [7][20]. We note that the hysteresis loop is slightly off centered about zero field at 5 K with the negative coercive field $H_c^- = -0.415(5)$ T and the positive coercive field $H_c^+ = 0.405(5)$ T. The asymmetry of the hysteresis loop, depending on the magnetic history, can be attributed to Exchange Bias (EB), which is a result of interaction between a FM and another ordered magnetic phase, typically an AFM state [24]. We observe that the bias field with field up to 7 T is smaller than the one previously observed with smaller applied field [23][25]. Besides EB, Fig. 2 also shows that increasing temperature causes the shape of the hysteresis loop to change from a rectangular shape, to a triangular-tailed rectangle, to a bi-triangular shape, until the hysteresis...
disappears completely. The shape of hysteresis loops can be characterized by magnetic domain nucleation fields $H_{nuc}^\pm$, and saturation field $H_{sat}^\pm$. We note that these fields depend on defects and the shape of a sample [26, 27]. For this reason, we use the same sample for all the bulk magnetization measurements presented in this article.

\[
\delta = n_\uparrow - n_\downarrow. \tag{1}
\]

If $\delta = 1$, the system only has up domains. The measured magnetization is $M = M_0 \delta$. Therefore, the average energy per site is

\[
E = \frac{N_c}{2} M_s^2 \delta^2 - M_s H \delta, \tag{2}
\]

where the first term is the demagnetization energy with demagnetization factor $N_c$, and second term is the Zeeman energy. The equilibrium $\delta_0$ is given by $\frac{\partial E}{\partial \delta}\big|_{\delta_0} = 0$,

\[
\delta_0 = \frac{H}{|H_{d,max}|}, \tag{3}
\]

where $|H_{d,max}| = N_c M_s$ is the maximum demagnetization field. When $H_{nuc}^- < H < |H_{d,max}|$, as $H$ increases, the fraction of up-domains grows. Similarly, the fraction of down-domains grows when $-|H_{d,max}| < H < H_{nuc}^+$ while $H$ decreases. Therefore, the magnetization follows as

\[
M = M_s \delta_0 = \frac{H}{N_c}, \tag{4}
\]

which characterizes the hypotenuse of the triangle or triangular tail in the hysteresis loop. The demagnetization factor of the sample is obtained from the inverse of the slope of the hypotenuse as $N_c = 0.608$. $|H_{d,max}|$ is then obtained from $M_c$.

The proportional part of the hysteresis loops above $\sim 100$ K can be understood with a domain-wall motion model [28]. A two-domain system is characterized by the difference between the fraction of up domains (field-aligned) and the fraction of down domains (field-antialigned):

\[
\delta = n_\uparrow - n_\downarrow. \tag{1}
\]

If $\delta = 1$, the system only has up domains. The measured magnetization is $M = M_0 \delta$. Therefore, the average energy per site is

\[
E = \frac{N_c}{2} M_s^2 \delta^2 - M_s H \delta, \tag{2}
\]

where the first term is the demagnetization energy with demagnetization factor $N_c$, and second term is the Zeeman energy. The equilibrium $\delta_0$ is given by $\frac{\partial E}{\partial \delta}\big|_{\delta_0} = 0$,

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\delta_0 = \frac{H}{|H_{d,max}|}, \tag{3}
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where $|H_{d,max}| = N_c M_s$ is the maximum demagnetization field. When $H_{nuc}^- < H < |H_{d,max}|$, as $H$ increases, the fraction of up-domains grows. Similarly, the fraction of down-domains grows when $-|H_{d,max}| < H < H_{nuc}^+$ while $H$ decreases. Therefore, the magnetization follows as

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![FIG. 2. Hysteresis loops with the applied field up to 7 T parallel to the $c$ axis at various temperatures. Only a selection of temperatures are shown. The hysteresis gradually changes from rectangle to bi-triangular shape, which is related to domain-wall pinning.](image)

![FIG. 3. a) Hysteresis loop at 131 K with the applied field up to 7 T and parallel to the $c$ axis. $H_{nuc}^<$ and $H_{sat}^<$ are shown with arrows. b) $H_{nuc}^<$ and $H_{sat}^<$ as functions of temperature with maximum applied field up to 7 T. The green curve is the theoretical value of $-|H_{d,max}|$. The black dashed line shows the 300 Oe applied field.](image)
greater than the maximum demagnetization field, and
the magnetization saturates immediately. Therefore, the
hysteresis loop is rectangular-shaped, and $H_{\text{sat}}$ is identi-
cal to $H_{\text{nuc}}^\downarrow$. Above 85 K, $|H_{\text{nuc}}^\downarrow| < |H_{\text{d,max}}|$, when down
domains nucleate, their relative fraction depends on the
applied field, and gradually increases as the applied field
ramps down. Hence, the hypotenuse of the hysteresis
loop is formed, and $H_{\text{nuc}}^\downarrow$ is identical to $-|H_{\text{d,max}}|$. We
can see that $H_{\text{nuc}}^\downarrow$ changes sign upon cooling. For our
sample, this happens at around 130 K, below which $H_{\text{nuc}}^\downarrow$
is negative, and the hysteresis loops are triangular-tailed
rectangles. Above 130 K, $H_{\text{nuc}}^\downarrow$ becomes positive, and the
hysteresis loops become double-triangular shaped.

We further note that above $H_{\text{nuc}}^\downarrow$ down domains can-
not grow. As will be shown later, the local maximum
at $T_A$ is due to the depinning of domain walls allowing
the previously deficient down domains to grow. The
local maximum disappears at applied fields above 300 Oe
because this field is greater than $H_{\text{nuc}}^\downarrow$ at any temper-
\n
\begin{align*}
\text{Asymmetry also emerges up to 160 K when the maxi-
mum applied field is only 100 Oe as shown in Fig. 4. The virgin curve at
130 K remains flat at first, then increases with a constant
slope, which is the hallmark of a domain-wall pinning-
type virgin curve [29]. The magnetization of the field-
increasing part of the hysteresis loop is smaller than that
of the virgin curve, indicating that the domains encounter
some pinning effect.}
\end{align*}

\begin{align*}
\text{FIG. 4. a) Hysteresis loop with applied field parallel to the
c axis up to 100 Oe. The arrow shows the coercive field $H_c$.}
\text{Nucleation field $H_{\text{nuc}}^\downarrow$ is obtained by the intersection of red}
\text{lines. b) $H_c$ and $H_{\text{nuc}}^\downarrow$, with maximum field up to 100 Oe, as}
\text{a function of temperature.}
\end{align*}

\begin{align*}
\text{Although the particular values of $H_c$ and $H_{\text{nuc}}^\downarrow$ are sample}
\text{dependent, we expect that their temperature dependence}
\text{and the creation of a domain depinning region are general}
\text{observations in the Co$_3$Sn$_2$S$_2$ system since the local max-
imum in $MvT$ has been observed in many studies [14, 22].}
\end{align*}

\begin{align*}
\text{Figure 5(a) shows the temperature dependence of the up-domain fraction extracted from the FCC (blue curve)
and FCW (red curve) magnetization data at 100 Oe and}
\text{from demagnetization theory (black curve). By equating the expected magnetization from demagnetization the-
ory, to the magnetization from a fraction of up domains $n_\uparrow$, we get}
\end{align*}

\begin{align*}
(2n_\uparrow - 1)M_s = \frac{1}{N_c}H_{\text{applied}}
\end{align*}

\begin{align*}
\text{In Eq. 5, the applied field, $H_{\text{applied}}$ and the demagnetiza-
tion factor $N_c$ are temperature independent. Eq. 6 can be}
\text{rewritten solving for the domain fraction}
\end{align*}
\[ n_\uparrow = \frac{1}{2M_s} \frac{H_{\text{applied}}}{N_c} + \frac{1}{2} \]  \hspace{1cm} (6)

Equation (6) gives us the result that the fraction of up domains should decrease as temperature decreases while \( M_s \) increases (black curve). However, domain wall pinning keeps the FCC and FCW up-domain fractions above the black curve for all but the temperatures near \( T_C \). By following the blue curve, we can understand the local maximum in the FCC \( M_eT \) curve. While cooling down below \( T_C \), the system encounters a domain-wall pinning effect, as seen in Fig. 5(a) where the blue curve begins to deviate from the black curve. This domain-wall pinning causes the magnetization to increase nearly proportionally to \( M_s \). Below around 132 K, the system enters a depinning region where the domain-wall pinning strength diminishes and as a result, the up-domain fraction decreases towards its theoretical no pinning value (the black curve) and the magnetization consequently decreases. Below about 120 K, however, the domain wall pinning strength increases again, and the domain fraction is held relatively constant. As shown in Fig. 5(b), the FCC magnetization shows a local minimum around 120 K, below which the magnetization increases. The repinning is consistent with the fact that domain walls rarely move below 119 K reported by a Lorentz microscopy study 31. Eventually, below about 95 K, domain walls are completely pinned and the magnetization becomes proportional to \( M_s \), i.e. \( M = \delta M_s \) with \( \delta = 0.31 \). While warming up, as shown by the red curve in Fig. 5, since excessive up domains already exist, they remain unchanged until around 155 K, where the reduced demagnetization field requires more up domains to form. Due to the presence of domain-wall pinning, the total magnetization of the sample when cooled down and warmed up follows two different pathways, resulting in the hysteresis between the FCC and FCW magnetization curves. The domain-wall depinning below 132 K might originate from the local symmetry breaking below \( T_C \) 10, where the distortion of lattice reduces the anisotropy energy and thus allows domains to flip.

Magnetic domain structures from 140 K to 120 K

Except for a recent MOKE study reported by Lee and coworkers 17, domain structures of \( \text{C}_{6} \text{S}_{13} \text{Sn}_{2} \text{S}_{2} (0001) \) single crystal samples over a wide range of temperature and applied magnetic fields are largely unknown. By measuring spatially resolved magnetization through the Kerr effect as a function of temperature, we can examine whether the magnetic anomaly is intrinsic to the physics in a single domain or due to the presence of multiple domains and domain walls. Such detailed information is elusive from bulk measurements averaged over the entire sample.

To examine domain structures near \( T = 130 \) K, we acquired a set of MOKE images at 140 K, 136 K, 132 K, 128 K, 124 K, and 120 K after the sample is field cooled in a magnetic field of \( H_\parallel = +94 \text{ Oe} \). These images are shown in Fig. 6. Other than the disappearance of a small embedded red domain in the upper left corner (between 136 K and 132 K) and that of an embedded blue domain to its immediate right (between 128 K and 124 K), the domain structure remains essentially unchanged between 140 K and 120 K. Limited by the spatial resolution of the scanning microscope (0.85 µm), we cannot rule out transitions in this temperature range that involve domain walls from these MOKE images alone.

We can extract the fraction of up domains, down domains, and domain walls from these images as follows. First, we measure the average absolute Kerr rotation of up and down domains. Then, we count points above...
FIG. 6. Polar Kerr rotation images of Co$_3$Sn$_2$S$_2$(0001) after FCC in $H_l = +94$ Oe (pointing out of paper along the c-axis) from room temperature acquired at (a) 140 K; (b) 136 K; (c) 132 K; (d) 128 K; (e) 124 K; (f) 120 K. Red regions are domains with magnetization pointing out of paper along the direction of the applied magnetic field; blue regions are domains with magnetization pointing in the opposite direction. The unit of Kerr rotation is µrad. The scale bar is 50 µm. The image size is 240 µm x 240 µm.

75% of the average value as up or down domains, and points below that value as domain walls. Figure 7 shows the evolution of these fractions as a function of temperature. As the sample is cooled down, the fraction of up domains shrinks while that of down domains grows due to the demagnetization effect. The domain walls seem to experience some pinning effect between 128 K and 136 K.

CONCLUSION

We attribute the magnetic anomaly $T_A$ near 130 K in the FCC magnetization curve in Co$_3$Sn$_2$S$_2$ to a domain wall pinning effect. This pinning effect is evident in both spatially averaged and spatially-resolved (via a MOKE microscope) measurements in low fields (below $\sim$ 300 Oe) and reaches a maximum around 132 K before releasing at lower temperatures. In addition, this domain wall pinning is responsible for the hysteresis between the bulk FCC and FCW magnetizations. We also find that the nucleation field changes its sign around the anomaly temperature $T_A = 130$ K, causing the shape of the hysteresis loops to change from triangular-tailed rectangles into double-triangles. While our findings corroborate the low mobility of domain walls near 119 K [31], the origin of the domain wall pinning and ultimately, unpinning at $T_A = 130$ K, remains unknown. It might originate from the local symmetry breaking below $T_C$ [16], where the distortion of lattice reduces the anisotropy energy and thus allows domains to flip. Further investigations are needed to explore the connection between domain wall depinning and local symmetry breaking, as well as its possible relation to the observed exchange bias effect at low temperatures.

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