Magnetic domain walls depinning 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 at 130 K is due to domain wall pinning. 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. The 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 is caused by an unusual temperature-dependent domain nucleation field that changes the sign at 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 crossing near the Fermi level when inversion symmetry or time-reversal symmetry is broken [1,3]. Co$_3$Sn$_2$S$_2$ is a ferromagnetic WSM and has a Curie temperature of $T_C = 174$ K [4]. 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 [5]. The topological features of this material lead to a large anomalous Hall conductivity [6], an anomalous Nernst effect [7], an unusual breaking-down of Ohm’s law [9]. In addition, an unusually large magneto-optic effect associated with the anomalous Hall effect have 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 indicates 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 Solt et al., however, rules out the existence of an antiferromagnetic structure [13]. A recent half-polarized neutron diffraction investigation reveals 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 kink around 130 K in the FCC magnetization is likely due to a transition between linear domain walls and circular 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 140 K to 120 K obtained under an external magnetic field of 97 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 resolved.

METHODS

Single Crystal Growth

Single crystals of Co$_3$Sn$_2$S$_2$ were synthesized by solution growth [8,18,19]. A ternary mixture with initial composition Co$_{12}$S$_8$Sn$_8$ 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) methods. 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 remnant 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 Co₃Sn₂S₂(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 to produce a variable field up to 100 Oe (0.01 T) along the c-axis (the surface normal) of the sample.

RESULTS AND DISCUSSION

Magnetization measurements

The FCC curve with magnetic field of 1000 Oe parallel to c axis is shown in Figure 1 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 μB per Co and a Curie-Weiss temperature around θ_{CW} = 175 K, which are consistent with a previously published work [13]. 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_A. Second, a downward kink arises at T_A = 130 K in the FCC data. This kink does not shift when the externally applied field changes and is also reported in other studies [14] [22]. A similar kink also emerges in the FCW magnetization, albeit at the temperature where the FCC and FCW curves start to depart. Previously, T_A was proposed to be an AFM-FM transition by a muon diffraction experiment [14]. A second order AFM-FM phase transition, however, should not depend on the magnetic history. Therefore, the hysteresis between FCC and FCW magnetizations indicates an alternative mechanism is responsible for the kink at T_A.

Hysteresis loops at various temperatures are shown in Fig. 2. The spontaneous magnetization, M_s, is 0.3 μB/Co at 5 K [6] [20]. However, the hysteresis loops are not centered about zero field. There is an offset between the negative coercive field H^-_c = 0.195 ± 0.003 T and the positive coercive field H^+_c = 0.13 ± 0.003 T. This offset disappears above 100 K. Similar behavior was also observed in Hall resistivity [23] [24]. 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, which is typically an AFM state [25]. Besides EB, Fig. 2 also shows that increasing temperature causes the shape of the hysteresis loop to change from a perfect square, to a triangular-tailed square, to a bi-triangular shape, until the hysteresis disappears completely. The shape of hysteresis loops can be characterized by magnetic domain nucleation fields H^\pm_{nuc} where the hysteresis loop shows a sudden jump, and saturation field H^\pm_{sat} where magnetization becomes saturated, as shown in Figure 3.

The proportional part of the hysteresis loops above ∼ 100 K can be understood with a domain-wall motion model [26]. A two-domain system is characterized by the difference between the fraction of field-aligned up do-
mains and the fraction of field-antialigned down domains

\[ \delta = n_{\text{up}} - n_{\text{down}}. \tag{1} \]

If \( \delta = 1 \), the system only has up domains. The measured magnetization is \( M = M_s \delta \). 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 \( \frac{\partial E}{\partial \delta} |_{\delta_0} = 0 \),

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

where \( |H_{d,\text{max}}| = N_c M_s \) is the maximum demagnetization field, which tends to push domain walls outside the system to become single-domain. When \( H_{\text{nuc}} < H < |H_{d,\text{max}}| \), as \( H \) increases, domain walls are pushed further inside the system, and therefore 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 \( N_c = 0.608 \). \( |H_{d,\text{max}}| \) is then obtained from \( M_s \).

When the field ramps down from 7 T, negative domains start to nucleate below a certain field and cause a sudden jump in the hysteresis loop. The onset of the jump is taken to be \( H_{\text{nuc}}^- \), indicating the nucleation of negative domains. \( H_{\text{nuc}}^+ \) is taken in a similar manner when the field ramps up. As the field further ramps down, the system becomes single-domained and its magnetization saturates, as shown in Figure 3(a). Figure 3(b) shows \( H_{\text{nuc}}^\pm \), \( H_{\text{sat}}^\pm \), and \( |H_{d,\text{max}}| \) as functions of temperature. Below 85 K, \( H_{\text{nuc}} > |H_{d,\text{max}}| \), once positive domains nucleate, the demagnetization field pushes domain walls outside the system, and the magnetization saturates immediately. Therefore, the hysteresis loop is square-shaped, and \( H_{\text{sat}} \) is identical to \( H_{\text{nuc}}^- \). Above 85 K, \( H_{\text{nuc}} < |H_{d,\text{max}}| \), when positive domains nucleate, the demagnetization field pushes domain walls to the equilibrium position and then gradually outside the system. Hence, the hypotenuse of the hysteresis loop is formed, and \( H_{\text{sat}} \) is identical to \( |H_{d,\text{max}}| \). Below 130 K, \( H_{\text{nuc}}^+ \) is positive, therefore hysteresis loop is a triangular-tailed square. Above 130 K, \( H_{\text{nuc}}^+ \) becomes negative, and hysteresis loop becomes double-triangular shaped. We further note that above 300 Oe, negative domains stop nucleating, which is consistent with the fact that the
anomaly at 130 K disappears above 300 Oe, indicating that the magnetic anomaly is driven by domain depinning which occurs between 132 K and 120 K, as will be discussed later.

To investigate the connection between domain-wall pinning and the magnetic anomaly, we compared $H_{\text{nuc}}^{\pm}$ with the FCC and FCW magnetization curves at 100 Oe, as shown in Figure 4. Below 120 K, the applied 100 Oe field is smaller than $H_{\text{nuc}}^{+}$ but larger than $H_{\text{nuc}}^{-}$, and consequently, both positive and negative domains stop nucleating. Therefore, the overall magnetization must grow proportionally to $M_s$, where $M = \delta M_s$. In this case, choosing $\delta = 0.31$ matches the FCC data perfectly below 120 K (black solid line). The nucleation temperature for negative domains that we get from our model (indicated by the black vertical line in Fig. 3), however, does not match the temperature at which we see negative domains actually form during FCC at $T_A$. This discrepancy can be understood because the nucleation field $H_{\text{nuc}}$ was obtained from measuring hysteresis loops up to 7 T, where a small fraction of negative domains must nucleate out of a saturated positive-domain environment. In contrast, while measuring FCC and FCW curves at low fields, such a saturation is never reached and therefore, the actual nucleation field for these measurements should be smaller than the nucleation field presented in Figure 3.

For evidence of domain-wall pinning effects at low fields, we measured hysteresis loops with the applied field only up to 100 Oe as shown in Figure 5. The virgin curve at 130 K at first remains flat, then increases with a constant slope, which is the hallmark of a domain-wall pinning-type virgin curve. The magnetization of the field-increasing part of the hysteresis loop is smaller than that of the virgin curve, indicating that the positive domains encounter some pinning effect. Asymmetry also emerges up to 160 K when the maximum applied field is only 100 Oe. At 130 K, the negative coercive field is $H_{\text{c}}^{-} = 34 \pm 0.5$ Oe, while the positive coercive field is $H_{\text{c}}^{+} = 45 \pm 0.3$ Oe, and the actual coercive field, $H_{\text{c}}$, is taken to be the average of the two. Figure 5(b) reveals a local maximum of $H_{\text{c}}$ at 132 K, which is consistent with the anomaly in FCC curve at 100 Oe. Figure 6 shows the evolution of positive domains fraction as a function of temperature. As shown by the blue curve, while cooling down from $T_C$, the system encounters some domain-wall pinning effect, which reaches the crest around 132 K, and releases afterwards. Consequently, negative domains start to nucleate more below 132 K, which causes the decreasing magnetization in the FCC. Further below 120 K, both positive and negative domains stop nucleating, resulting in the magnetization proportional to spontaneous magnetization, where $M = \delta M_s$. While warming up, as shown as the red curve, since excessive positive domains already exist, they remain unchanged until around 155 K, where the demagnetisation requires more positive domains to form. Due to the presence of domain-wall pinning, the energy when cooled down and warmed up
follows two different pathways, resulting in the hysteresis in the FCC and FCW. The domain-wall depinning below 132 K 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.

![Graph showing evolution of up domains of FCC (blue), FCW (red), and theoretical modeling with $N_c = 0.6$ (black), as a function of temperature under 100 Oe applied field.](image1)

**FIG. 6.** Evolution of up domains of FCC (blue), FCW (red), and theoretical modeling with $N_c = 0.6$ (black), as a function of temperature under 100 Oe applied field. The intersect of black dotted lines with red and black curves shows the onset of depinning as warming up.

**Magnetic domain structures from 140 K to 120 K**

Except for a recent MOKE study reported by Lee and coworkers [17], domain structures of Co$_3$Sn$_2$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 measurements done on whole samples.

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 the magnetic field of $H_{||} = +94$ Oe. They are shown in Figure 7. 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 75% of the average value as up or down domains, and points below that value as domain walls. Figure 8 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. For a ferromagnetic sample of finite size, the demagnetization theory can be used to describe the effect of ferromagnetic domains that form when $H_{\text{applied}} < |H_{d,\text{max}}|$. By equating the expected magnetization from the demagnetization theory, to the magnetization from a fraction of up domains $n_{\text{up}}$ [27], we get

$$\frac{2(n_{\text{up}} - 1)M_s}{N_c} = H_{\text{applied}}$$

In eq. 5, the applied field, $H_{\text{applied}}$ and the demagneti-
zation factor $N_c$, which is determined from the shape of the sample, are temperature independent. Eq. 5 can be rewritten solving for the domain fraction

$$n_{up} = \frac{1}{2M_s} \frac{H_{\text{applied}}}{N_c} + \frac{1}{2}$$

(6)

Eq. 6 indeed gives us the result that the fraction of up domains should decrease as temperature decreases while $M_s$ increases. However, the domain walls seem to experience some pinning effect between 128 K and 132 K. An ac MOKE study of Co$_3$Sn$_2$S$_2$ reveals a domain wall mobility minimum near 140 K [17, 28], corroborating the domain wall pinning effect. The abnormal behavior of domain walls near 130 K might suggest a domain wall phase transition where linear wall evolves to elliptical wall [17].

![FIG. 8. Evolution of up and down domains and domain walls as cooling down from MOKE image. The up domains shrink while down domains grow. Domain wall pinning is observed between 128 K and 132 K.](image)

**CONCLUSION**

We attribute the magnetic anomaly at $T_A = 130$ 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 loop to change from a triangular-tailed square into a double-triangle. While our findings corroborate the minimum mobility of domain walls near 140 K [17], the origin 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 unpinning and local symmetry breaking, as well as its possible relation to the observed exchange bias effect at low temperatures.

**ACKNOWLEDGMENTS**

V.T., Z.S. and R.R.U. acknowledge support from the UC Lab Fees Research Program (LFR-20-653926) and UC Davis Startup funds. X.D.Z. acknowledges a Visiting Lecture Professorship from Fudan University in support of this work. We acknowledge support from the Physics Liquid Helium Laboratory fund.

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