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Tunable exchange bias in the magnetic Weyl semimetal math xmlns="http://www.w3.org/1998/Math/MathML">mrow>ms ub>mi>Co/mi>mn>3/mn>/msub>msub>mi>Sn/mi>mn> 2/mn>/msub>msub>mi mathvariant="normal">S /mi>mn>2/mn>/msub>mi mathvariant="normal">S /mi>mn>2/mn>/msub>/mrow>/math> Avia Noah, Filip Toric, Tomer D. Feld, Gilad Zissman, Alon Gutfreund, Dor Tsruya, T. R. Devidas, Hen Alpern, Atzmon Vakahi, Hadar Steinberg, Martin E. Huber, James G. Analytis, Snir Gazit, Ella Lachman, and Yonathan Anahory Phys. Rev. B **105**, 144423 — Published 20 April 2022 DOI: 10.1103/PhysRevB.105.144423

### Tunable exchange bias in magnetic Weyl semimetal Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub>

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**Abstract:** Exchange bias is a phenomenon critical to solid-state technologies that require spin valves or non-volatile magnetic memory. The phenomenon is usually studied in the context of magnetic interfaces between antiferromagnets and ferromagnets, where the exchange field of the former acts as a means to pin the polarization of the latter. In the present study, we report an unusual instance of this phenomenon in the topological Weyl semimetal Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub>, where the magnetic interfaces associated with domain walls suffice to bias the entire ferromagnetic bulk. Remarkably, our data suggests the presence of a hidden order parameter whose behavior can be independently tuned by applied magnetic fields. For micron-size samples, the domain walls are absent, and the exchange bias vanishes, suggesting the boundaries are a source of pinned uncompensated moment arising from the hidden order. The novelty of this mechanism suggests exciting opportunities lie ahead for the application of topological materials in spintronic technologies.

### 1. Introduction

The phenomenon of exchange bias (EB) was first realized in Co/CoO<sub>x</sub> heterostructure particles [1]. The magnetic hysteresis loop of this heterostructure was centered around a field  $H_{EB} = -(H_c^- + H_c^+)/2 \neq 0$ , where  $H_c^+$  and  $H_c^-$  represent the coercive fields for the positive and negative fields, respectively. The observed shift in the magnetic hysteresis loop was attributed to the pinning of the ferromagnetic (FM) Co moments by the exchange interactions with the anti-ferromagnetic (AFM) CoO<sub>x</sub>. Since then, EB was observed in other heterostructures of different FM and AFM materials [2] including spin glasses [3], superparamagnets [4], and ferrimagnets [5]. Moreover, some realizations of EB can be produced without heterostructures [6,7], and some do not require activation by cooling in a magnetic field and create spontaneous EB that is isothermally set at low temperatures [4,8]. This abundance of systems presenting EB have produced a plethora of suggested mechanisms to explain this phenomenon. The main component of these mechanisms lies in a source of uncompensated spins that induce a preferred direction.

Recently, EB was observed in a pure bulk Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub> single crystals [9], with no substrate effects [6] or material doping [7], invoking the need for a different mechanism for the EB they present. Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub> was recently discovered as a magnetic Weyl semimetal, where the breaking of time-reversal symmetry due to ferromagnetism satisfy the requirement of a broken symmetry to create the topological band structure [10–18]. Of particular interest is the non-trivial interplay between strong magnetic correlations and the emergent topological band structure of Weyl fermions in such materials [19–25], which is not present in their inversion-symmetry-breaking counterparts. Specifically, the magnetic moments in Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub> reside on the Co sites and are arranged in a layered kagome lattice. Experimentally, below 175 K, a dominant FM phase with an easy axis oriented out of the kagome plane is observed, although this phase may coexist with an in-plane (IP) antiferromagnetic phase [9,21]. Below this temperature, the electronic dispersion has been shown to host Weyl fermions [16,26] associated with a giant anomalous Hall effect [12,13,27].

Even at the level of basic magnetic properties, experimental observations display puzzling inconsistencies. Explicitly, the reported values of magnetic saturation field,  $H_s$ , range from 0.06 T to 0.5 T in the bulk [9,12,13,21,26,28–30], and may reach values of a few Tesla in micron-size samples [31–35]. Such large variations are surprising since  $H_s$  is a material property determined by the crystalline structure and electronic orbitals, and the large variability is unlikely to be due to a simple geometrical factor or sample quality. Presently, these unusual magnetic properties including the presence of EB and their underlying physics are far from understood.

In this work, we address these questions by using a combination of transport measurements and magnetic imaging to visualize the magnetic domains and elucidate the mechanism underlying the EB in Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub>. We reveal a new kind of EB, that can be set and changed isothermally at low temperatures.

We attribute this to a hidden IP magnetic order that is likely related to the domain walls. These findings suggest a promising future for the application of magnetic Weyl semimetals in memory and spintronic technologies, although they may require further miniaturization for practical applications.

### 2. Results

Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub> crystals were grown by the self-flux method commonly used for these crystals [9,36,37]. For electronics transport measurements, four aluminum wires were glued to a 80 µm-thick sample using silver epoxy, as shown in **Figure 1a** (see Methods and Supplementary Note 6 [38]). The sample was zero-field cooled (ZFC) down to 4.2 K, where the sample displays FM characteristics [21]. A 2 mA current was applied along the *x* axis while the voltage  $V_y$  was measured along the *y* axis (see Figure 1a). We report the Hall resistance  $R_{xy} = V_y/I_x$  as a function of the out-of-plane (OOP) magnetic field  $\mu_0 H_z$  under distinct magnetic field protocols up to 6 Tesla.

As an example, Figure 1b (black curve) presents  $R_{xy}$  measurements starting from ZFC and ramping  $H_z$  at a rate of 4 mT/s to +1 T. Subsequently, we carried out a full -1 T to +1 T magnetic field loop. During the initial field ramp,  $R_{xy}$  grows smoothly, starting from zero and reaching saturation at 95 mT (see **Figure 3b** blue curve). The following  $R_{xy}$  curve exhibits sharp jumps associated with two distinct coercive fields,  $H_c^+ = 140$  mT and  $H_c^- = -240$  mT, for positive and negative  $H_z$ , respectively (Figure 1b, black curve). The asymmetry between the coercive fields yields  $H_{EB} = 50$  mT. We note that these  $H_c^{\pm}$  values are reproducible within our experimental resolution (better than 5 mT), as long as the field is swept symmetrically from +1 T to -1 T ( $\mu_0 | H_{max}^{\pm} | = 1$  T). As previously [9] reported for ZFC conditions, the lower coercive field is determined by the direction of the first field excursion. Here the positive direction was chosen initially, and we obtain  $|H_c^+| < |H_c^-|$ , and thus  $H_{EB} > 0$ .

Our results differ from those in previous reports, which did not observe a finite  $H_{EB}$  when performing a magnetization loop with a much larger  $\mu_0 H_{max}^{\pm} \sim 9$  T. It is therefore interesting to explore the evolution of the hysteresis loop at intermediate fields, and for asymmetric field-sweep protocols. With that goal in mind, we apply such a protocol with  $\mu_0 H_{max}^- = -2$ , -3, -6 T while fixing  $\mu_0 H_{max}^+ = 1$  T (Figure 1b blue curves). Interestingly, we find that  $H_c^+$  values increase by more than a factor of 5, ranging from 140 mT ( $\mu_0 H_{max}^- = -1$  T) to 736 mT ( $\mu_0 H_{max}^- = -6$  T). In contrast,  $H_c^-$  varies only slightly from the -240 mT observed in the  $\pm 1$  T symmetric sweep to -140 mT for all the other sweeps. Consequently, over these protocols,  $H_{EB}$  changes both sign and magnitude from  $H_{EB} = 50$  mT after ZFC, to -300 mT after  $\mu_0 H_{max}^- = -6$  T. Different values of  $|H_{max}^\pm|$  yield different values of  $H_c^\pm$  (Figure 1c). Curiously,  $H_c^+$  is constant for distinct values of  $H_{max}^-$  resulting in a plateau structure as shown in Figure 1c. The reverse protocol i.e., keeping  $H_{max}^- = -1$  T and varying  $H_{max}^+$  shows similar results that are detailed in supplementary note 2 and supplementary Figure 2 [38]. The observed sign reversibility in  $H_{EB}$  implies that it can be set to be vanishingly small, as demonstrated in Supplementary Note 3 [38].

It is essential to contrast the above result with those from minor loop protocols used in conventional ferromagnets, where distinct  $H_c$  values can be reached by applying  $|H_{max}| < H_s$ . Physically, this phenomenon can be attributed to residual magnetic domains that are anti-parallel with the external field [39–42]. The present case is different, since  $|H_{max}| > H_s$  in all the protocols described above. Our measured  $H_s$  is in good agreement with previous reports where  $H_s \leq 500$  mT [9,12,13,21,28–30].

The non-trivial evolution of  $H_c^{\pm}$  as a function of  $|H_{max}|$  beyond  $H_s$  indicates that  $H_c^{\pm}$  does **not** depend solely on OOP magnetization. Notably, previous reports could not rule out a slight canting of magnetic moments below 125K, even though the magnetization was predominantly oriented OOP [12,21]. Moreover, theoretical calculations predict that even slight canting might dramatically affect the Weyl nodes [19] and associated magnetic textures [20]. Motivated by these theoretical predictions, it is also interesting to study the effect of an IP field,  $H_{IP}$ , on  $H_c^{\pm}$ .

To reveal the influence of  $H_{IP}$  on  $H_c^{\pm}$ , we conduct IP field excursions  $H_{IP}^{ex} = \pm 2$  T, followed by the  $\mu_0 |H_{max}^{\pm}| = 1$  T loop protocol at  $H_{IP} = 0$ . Considering that the IP saturation field is 23 T and given that the IP magnetization varies linearly [29] from 0° for  $H_{IP} = 0$  to 90° for  $H_{IP} = 23$  T we deduce that our field excursion cause a tilt of ~ 8°. We start by measuring  $R_{xy}$  after ZFC, before applying any IP field. Under these conditions,  $R_{xy}(H_z)$  shows the expected  $H_c^+ = 128$  mT and  $H_c^- = -241$  mT with  $H_{EB} = 56$  mT (black line, Figure 1d). An IP field is applied reaching  $\mu_0 H_{IP}^{ex} = 2$  T and then ramped down to zero before measuring again  $R_{xy}(H_z)$ . The  $R_{xy}(H_z)$  curve reveals a small increase in both  $H_c^+ = 139$  mT,  $H_c^- = -275$  mT and  $H_{EB} = 68$  mT (blue line) with respect to the ZFC measurements. On the other hand, applying an IP field excursion in the other direction up to  $\mu_0 H_{IP}^{ex} = -2$  T and back to zero, results in a change of sign in  $H_{EB}$ , with  $H_c^+ = 244$  mT,  $H_c^- = -128$  mT, and  $H_{EB} = -58$  mT (red line). This experimental observation indicates that even a small degree of canting has a dramatic effect on  $H_{EB}$ . We emphasize that the material retains a memory of the  $H_{EB}$  magnitude and sign, even after the IP field is set to zero. We also note that no hysteresis behavior is observed for the IP magnetization as a function of the IP field [12,29].



Figure 1. Controlling the exchange bias of  $Co_3Sn_2S_2$  single crystal sample by applying out-of-plane and in-plane field protocols at 4.2 K. (a) Optical image of the sample and a schematic depicting paths for current  $I_x$ , voltage probe  $V_y$ , and the crystal axes. (b) Hall resistance  $R_{xy} = V_y/I_x$  as a function of the out-of-plane field  $\mu_0H_z$ . After ZFC the field is swept between  $\pm 1$  T resulting in a rectangular  $R_{xy}$  with  $H_{EB} = 50$  mT (black). Controlling the positive coercive field  $H_c^+$  by performing a larger field excursion in the negative direction  $\mu_0H_{max}^- = -2$ , -3, -6 T while keeping  $\mu_0H_{max}^+ = 1$  T resulting in  $H_{EB} = -170$ , -200, -300 mT, respectively (dark blue to light blue). (c)  $H_c^+$  as a function of  $\mu_0H_{max}^-$ , with colored dots corresponding to the measurements shown in **b**. (d) Each measurement is performed at  $H_{IP} = 0$  after ZFC (black), after an excursion to  $H_{IP}^{ex} = 2$  T resulting in  $H_c^+ = 139$  mT and  $H_c^- = -275$  mT,  $H_{EB} = 68$  mT (blue) and to  $\mu_0H_{IP}^{ex} = -2$  T yielding  $H_c^+ = 244$  mT,  $H_c^- = -128$  mT and  $H_{EB} = -58$  mT (red). Three loops performed for each measurement gave similar results, and one representative result is shown for clarity. In both **b** and **d** the curves for different protocols are shifted vertically for clarity.

In order to further explore the possibility that the  $H_{EB}$  information is not encoded in the OOP magnetization, we now attempt to demagnetize the sample, using a minor loop protocol. Explicitly, we carry out one minor loop sweep with  $\mu_0 |H_{max}^{\pm}| \sim H_c^{\pm}$  (Figure 2a Red curve number 1 and 2). The results reveal a vanishingly small  $R_{xy}$  (number 3), which naively indicates that the sample is demagnetized. A priori, one would expect a demagnetized sample to lose all  $H_{EB}$  information and return to the initial random ZFC conditions. To test this expectation, we carry out an OOP field excursion oppositely aligned with the initial ZFC excursion (namely, we sweep toward negative fields). Remarkably, we find precisely the same hysteresis loop as after the ZFC loop (Figure 2b blue curve). This finding suggests that even though the sample appears demagnetized in transport, it retains the information regarding the direction of the first field excursion after ZFC. More broadly, this provides experimental evidence that  $H_{EB}$  information is stored in degrees of freedom other than the global OOP magnetization.



**Figure 2. Demagnetization and magnetization (a)** Demagnetization protocol, the field swept from -1T to 1T to initialize the sample and from +1 until  $H_c^-$  is reached (blue). Sweeping the field between the distinct  $H_c^{\pm}$ , after three repetitions produces a vanishingly small  $R_{xy}$  (red). (b) Magnetization of the sample. The field is first swept to -1T (red) and then a +/- 1T loop is performed (blue).

To gain a better insight into the magnetic structure, we conduct local magnetic field imaging  $B_z(x, y)$  using a scanning SQUID that resides at the end of a sharp tip (Figure 3a). The SQUID-on-tip (SOT) provides high spatial resolution for magnetic imaging [43,44] reaching single-spin sensitivity [45,46]. Images are taken at 4.2 K with SQUIDs having a diameter ranging from 110 to 130 nm (see Methods and Supplementary Note 1 [38]).

Figure 3c presents the results of imaging the ZFC sample with magnetic features of a few microns yielding a magnetic contrast of ~ 50 mT. With increasing field, domains parallel to the field grow at the expense of the anti-parallel domains (Figure 3 c-f and Movie 1). Above the saturation field, 95 mT, starting from ZFC conditions, the magnetic contrast drops below 1.5 mT. The local magnetic structure does not appear to further evolve on this scale, which is in agreement with transport and global magnetization measurements [9,12,13,21,28–30]. These small micron-sized magnetic features were not observed in previous magnetic imaging work done on this material [47,48] most likely because the authors did not image the sample in ZFC conditions.

As shown previously, while the ZFC and the demagnetized state both exhibit a vanishing  $R_{xy}$  (Figure 2 and Figure 3b, green curve), there is a crucial difference between these states in that the latter retains the  $H_{EB}$  memory, while the former does not. It is therefore interesting to observe whether this difference is also apparent in the local magnetic structure. To examine this, we compare the respective magnetic structures of the two states (Figure 3 g-j and movie 2). The results indicate that, unlike the ZFC state, the demagnetized state exhibits a stripe pattern with a typical width of 10 µm, and a length that extends beyond our field of view (45  $\mu$ m). The memory of a finite exchange bias  $H_{EB}$  is retained only by the larger domain.

Similar magnetic domain structures are observed when the magnetization reverses (Figure 3 k-n and movie 3). We also note that similar domains of typical size of a few tens of microns were recently observed by MFM and by scanning Kerr microscopy in similar conditions [47,48]. Notably, the appearance of these features always coincides precisely with a sign change in  $R_{xy}$  (Figure 3b red curve), indicating that our microscopic images ( $45 \times 45 \ \mu m^2$ ) are representative of the much larger sample (mm size). We note that this transitory state is observed only for loops where  $|H_{max}^{\pm}|$  is relatively low  $|H_{max}^{\pm}| < 300 \ mT$ , and is only visible for less than 30 mT beyond the field at which  $R_{xy}$  changes sign. For protocols with a larger  $|H_{max}^{\pm}|$ , no transitory state was observed and instead an abrupt change in local magnetic field was recorded by the SOT between two field steps (smaller than 5 mT) coinciding with the change of sign of  $R_{xy}$ . This indicates that the magnetization reversal in such case occurs abruptly throughout the sample with no evolution of the magnetic landscape observed on either side of the magnetic transition.

![](_page_7_Figure_2.jpeg)

**Figure 3. Scanning SOT microscopy images of Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub> bulk at 4.2 K. (a)** Optical image of the SOT pointing down, the sample, and the reflection of the SOT on the sample. (b)  $R_{xy}$  as a function of applied magnetic field for different phases of the sample. ZFC to saturated field (blue), demagnetized state to saturated field (green) and magnetization reversal with transitory state (red).  $R_{xy}$  corresponding to a loop with  $\mu_0 |H_{max}^{\pm}| = 1$  T is shown for comparison (black dashed). The fields at which the images were taken are marked with dots. The curves for different protocols are shifted vertically for clarity. (c-n) Sequence of magnetic images of different states of the sample at distinct values of applied out-of-plane field  $\mu_0 H_z$ . (c-f). Evolution from ZFC to the saturation field.  $\mu_0 H_z = 45$  mT **c**,  $\mu_0 H_z = 65$  mT **d**,  $\mu_0 H_z = 85$  mT **e**,  $\mu_0 H_z = 95$  mT **f**. (g-j) Evolution of demagnetization to the saturation field.  $\mu_0 H_z = 30$  mT **g**,  $\mu_0 H_z = 50$  mT **h**,  $\mu_0 H_z = 75$  mT **i**,  $\mu_0 H_z = 95$  mT **j**. (k-n) Magnetization reversal with transitory state.  $\mu_0 H_z = 35$  mT **k**,  $\mu_0 H_z = 45$  mT **l**,  $\mu_0 H_z = 75$  mT **m**. All images are 45x45  $\mu$ m<sup>2</sup>, pixel size 480 nm **c-f** and 980 nm **g-n**, acquisition time 8 min/image **c-f** and 5.8 min/image **g-n**. The bright to

dark color scale represents 50 mT and is the same for all images. See Supplementary Movies 1-3 corresponding to images c-f, g-j, k-n, respectively

From the above observations, we can conjecture that there is a minimal length scale of the domains required for the appearance of EB. This hypothesis can be tested by repeating the same field sweep protocols on samples with lateral dimensions of a few tens of microns, which is comparable to the size of the domains following the initial field sweep ( $\geq 45 \,\mu$ m, Figure 3 g-n). For such samples, we expect to see a single, or at most a few, magnetic domains. We use a Focused Ion Beam (FIB), to cut a slab of area of 60 x 30  $\mu$ m<sup>2</sup> from the bulk crystal with thicknesses ranging from 1  $\mu$ m to 42  $\mu$ m and the *c* axis pointing out of the plane (**Figure 4a**). Platinum is then used as a contact to lithographically defined 50 nm-thick Nb tracks to the sample. Chemical analysis reveals that the sample maintained stoichiometric ratio of its constituent elements except near the surface where C and Ga were detected (Supplementary Note 6 [38]). The FIB sample is ZFC only down to 10 K in order to avoid the complications arising from the superconductivity in Nb ( $T_c < 9.2 \,$  K). Currents ranging from 0.4 mA to 0.8 mA are applied while the transverse voltage is measured as shown in figure 4b, maintaining similar current densities as in the bulk measurements.

The results of these experiments for a 6  $\mu$ m thick sample are shown in Figure 4c-d. First, we note that the  $H_{EB}$  resulting from a complete  $\mu_0|H_{max}| = H_c$  loop protocol is vanishingly small ( $H_{EB} < 5$  mT). Secondly, starting from ZFC conditions (blue curve), we find that  $H_c > 1$  T. This value should be compared with the  $H_c \sim 0.2$  T that was previously obtained with an identical protocol on mm-sized samples. We find that the  $H_c$  values are remarkably robust to radically different protocols. In particular, we did not observe demagnetization (Figure 4c) or variation in  $H_c$  under the application of  $\mu_0 H_z = 6$  T (Figure 4d). We further corroborate the absence of magnetic domain walls via SOT microscopy. The entire portion of the sample that was probed (more than 50 % of the area) changed magnetization abruptly in coincidence with the jump in  $R_{xy}$  (Figure 4c insets, Supplementary movie 8 [38]). This observation provides strong experimental evidence for the presence of a single sample-sized magnetic domain. These large values of  $H_c$  are similar to those previously reported in recent works [31–34].

Results from applying the same analysis to samples with different thicknesses (SM 4), revealed only a small, or no clear dependence of  $H_c$  on the sample thickness. In addition, the demagnetization protocol has no effect on  $H_c$  for any of the thicknesses tested. Finally, in all but the thickest (42 µm) sample we did not detect any measurable EB (Figure 4e-f). The fact that the thickest microcrystal showed some EB suggests that the lateral dimensions are near the critical dimensions to obtain EB (few tens of microns). A summary of  $H_{EB}$ ,  $H_c^{max}$ , and  $R_{xy}$  measured for all the samples presented in this work is shown in Figure S7.

![](_page_10_Figure_0.jpeg)

Figure 4.  $R_{xy}$  measurements of Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub> FIB samples with distinct thicknesses ranging from 6 to 42 µm at 10 K. (a) SEM image of four different Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub> single crystals cut by FIB with an area of 60 x 30 µm<sup>2</sup> and thicknesses ranging from 6 to 42 µm. (b) SEM image at higher magnification of the 6 µm thickness sample including a schematic of the measurement and the crystal axes. c-f R<sub>xy</sub> measurements under different out-of-plane protocols. c-d. Sample thickness 6 µm. e-f Sample thickness 42 µm. (c,e) Blue: After ZFC, the field is swept three times between ±1T. Red: Finding the positive and negative coercive fields. Sweeping the field six times between the positive and negative coercive fields, does not change the coercive field, therefore no demagnetization process is possible in the FIB sample. Only two curves are shown for clarity. (d,f) Applying a positive 6 T field and then sweeping the field until the negative coercive field does not change the coercive field of the 6 µm sample. The negative coercive field of the 42 µm sample grows by 200 mT resulting in  $H_{EB} = 100$  mT. The R<sub>xy</sub> measurements of samples with other thickness are presented in SM (4). C inset: SOT imaging of the 6 µm sample before and after  $H_c^+$ presenting a single domain structure which flips at  $H_c^+$ . All images are 45x45 µm<sup>2</sup>, pixel size 480 nm, acquisition time 9 min/image. The bright to dark color scale represents 10 mT and is the same for all images. See Supplementary Movie 8 [38].

### 3. Discussion

We now turn to discuss the physical consequences of our experimental findings. It is clear that  $H_{EB}$  is not a static material property and can be tuned by applying IP and OOP field protocols. That  $H_c^{\pm}$  evolves with  $H_{max}^{\mp}$  well beyond the saturation field implies an undetectable evolution in the spin texture, which dramatically affects  $H_c$  of the entire sample. The fact that the sign of  $H_{EB}$  is determined by the direction of the IP field, even after that field is no longer applied, provides experimental evidence that IP spin canting determines the exchange bias of the OOP magnetic moment in both sign and size.

In contrast, for microcrystals thinner than 40  $\mu$ m, we measure a vanishingly small  $H_{EB}$  and the value of  $H_c^{\pm}$  were found to be independent of the applied magnetic field protocol. The measured values of  $H_c^{\pm}$  were also more than five times larger than the  $H_c^{\pm}$  measured for mm-sized sample using a  $H_{max}^{\pm} = 1$  T protocol (See figure S7). Looking at the thickest measured microcrystal, we note the appearance of a finite  $H_{EB} = 10$  mT and a measurable drop in  $H_c^{\pm}$ . Our magnetic imaging results show that domain walls are not formed in microcrystals (Figure 4c and movie 8). The correlation between the magnitude of  $H_c^{\pm}$  and the presence of finite  $H_{EB}$  is yet to be formally explained, however, the fact that they both depend on the sample size on length scales that fit the domain size (few tens of microns) suggest that the presence of domain wall is one of the plausible explanations. Broadly, a finite EB entails the presence of uncompensated moments. Indeed, for larger  $H_c$ , these moments are more susceptible to polarization and hence their contribution to the EB will be diminished.

Although the precise mechanism for the EB in this material remains to be identified, previous reports combined with our present findings, may point to a possible physical mechanism. A good candidate should be an uncompensated spin texture such as an AFM phase that would coexist with the dominating FM order. At the domain walls, ferromagnetism is weak and permits the appearance of competing magnetic orders. In that regard, Ref. [21] reported that the coexistence of AFM and FM survives all the way down to 25 K, in proximity to the saturation field where the presence of domain walls is expected. Our results show that these domain walls are only stable enough to be measured for protocols where  $H_{max}^{\pm} < 0.3$  T (transitory state Figure 3). In the work of Ref. [21] the authors of that work used a protocol with  $H_{max}^{\pm} = \pm 6$  T. We speculate that this is the reason why they did not observe the AFM phase down to 4.2 K. The AFM state presents a chiral magnetic structure of the IP spin component that could be caused by the inherent magnetic frustration of kagome lattice or Dzyaloshinsky-Moriya (DM) interlayer interaction. The two possible chiralities of the IP AFM state could be toggled by an IP field and affect  $H_c$ . Moreover, it is interesting to relate these results to other ferromagnets exhibiting exchange bias due to interlayer DM interaction [49] which are allowed by symmetry in Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub>. These results could also be related to the memory effect emerging from the chiral domains in an in-plane AFM kagome [50]. It is worth noting that DM interaction is allowed by symmetry argument since the inversion centers present in Co<sub>3</sub>Sn<sub>2</sub>S<sub>2</sub> are not located at the bisecting point of the straight line between two out-of-plane Co atoms [51]. The experimental evidence in favor of local AFM order is only circumstantial and invites future explorations, which will allow for direct confirmation of this scenario or equally interesting alternative.

Another tantalizing mechanism involves electronic boundary states that were theoretically predicted to form at the interface between two magnetic domains in magnetic Weyl semimetals. While an electronic mechanism is less likely to drive the reported EB phenomena, our work provides a concrete protocol for nucleating magnetic domain walls that are essential for the observation of topological boundary effects in magnetic Weyl semimetals. The presence of magnetic boundaries encompass exciting transport and electrostatic phenomena related to the axial magnetic field [20,22] that inspired recent experimental efforts [22,24,25]. Though beyond the scope of the current work, our experimental system provides a unique opportunity to address these exotic phenomena present at magnetic domain walls.

# 4. Methods

- **4.1. Single crystal growth.** Single crystals were grown from a stochiometric ratio of elements using the self-flux method (Sn flux). The elements were placed in AlO<sub>x</sub> crucible and sealed in an evacuated quartz tube.
- **4.2. Transport measurements.** Transport measurements were performed at 4.2 K cooling inside a Liquid Helium Dewar. Unless otherwise mentioned, a current of 2 mA was applied along the ab plane. Distance between current contacts was 1.6 mm and between  $V_{\gamma}$  contacts 1.2 mm. In all measurements, 25 µm Al wires were glued using silver epoxy to a 80 µm-thick sample as measured using a 3D profilometer. In bulk samples, each transport data set was measured after ZFC to initialize the sample. A vector magnet was used to apply IP and OOP magnetic fields. The magnet consists of a standard coil for OOP field and a split coil of IP field.
- **4.3. Scanning Squid-On-Tip microscopy:** The SOT was fabricated using self-aligned three-step thermal deposition of Pb at cryogenic temperatures, as described previously [45]. Supplementary Figure S1 [38] shows the measured quantum interference pattern of one of the SOTs used for this work with an effective diameter of 130 nm and a maximum critical current of 98 μA. The asymmetric structure of the SOT gives rise to a shift of the interference pattern resulting in good sensitivity in a wide range of fields. All measurements were performed at 4.2 K in a low pressure He of ~1 mbar.
- 4.4. Statistic and uncertainties: Statistic and uncertainties: The resolution of the magnetic field is 4 mT and is determined by the magnetic sweep rate (4 mT/s) and the integration time of the signal (1 s). A systematic error on the magnetic field of a few G at most can be attributed to trapped flux in the magnet and the residual magnetization of surrounding parts. Typically, we did not observe drifts

of  $H_c$  larger than our field resolution (4 mT) for identical protocols on given sample. The uncertainty on  $V_{xy}$  is dominated by electrical noise. A usual systematic DC offset was observed and removed by symmetrizing the data around  $V_{xy} = 0$ . The uncertainty on  $V_{xy}$  is the main source of uncertainty of  $R_{xy}$ . The uncertainty on the distance is better than the last digit displayed. The SOT images are slightly distorted due to the piezoelectric hysteresis estimated to be 10-20 %.

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### 6. Author contributions

Y.A., E.L., A.N. and S.G. conceived the experiment and analyzed the data. A.N. and Y.A. performed the scanning SOT measurements and the transport measurements. A.N., T.D.F., F.T., G.Z. and Y.A. constructed the scanning SOT microscope. A.N., F.T., T.D.F., G.Z., A.G., D.T, and Y.A. Fabricated and characterized the SOT devices. M.E.H. developed the SOT readout system. E.L. and J.G.A. grew the crystals. A.V., Y.A., A.N., G.Z., A.G. and H.A. fabricated and measured the FIB samples. A.N., T.R.D. and H.S. in-plane transport measurements. Y.A., S.G., A.N., E.L., F.T. and J.G.A. wrote the paper with contributions from all authors.

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