Observation of a phase transition within the domain walls of ferromagnetic Co$_3$Sn$_2$S$_2$

Changmin Lee$^{1}$, Praveen Vir$^{2}$, Kaustuv Manna$^{2,3}$, Chandra Shekhar$^{2}$, J. E. Moore$^{1,4}$, M. A. Kastner$^{5,6,7}$, Claudia Felser$^{2}$ & Joseph Orenstein$^{1,4}$

The ferromagnetic phase of Co$_3$Sn$_2$S$_2$ is widely considered to be a topological Weyl semimetal, with evidence for momentum-space monopoles of Berry curvature from transport and spectroscopic probes. As the bandstructure is highly sensitive to the magnetic order, attention has focused on anomalies in magnetization, susceptibility and transport measurements that are seen well below the Curie temperature, leading to speculation that a “hidden” phase coexists with ferromagnetism. Here we report spatially-resolved measurements by Kerr effect microscopy that identify this phase. We find that the anomalies coincide with a deep minimum in domain wall (DW) mobility, indicating a crossover between two regimes of DW propagation. We demonstrate that this crossover is a manifestation of a 2D phase transition that occurs within the DW, in which the magnetization texture changes from continuous rotation to unidirectional variation. We propose that the existence of this 2D transition deep within the ferromagnetic state of the bulk is a consequence of a giant quality factor for magnetocrystalline anisotropy unique to this compound. This work broadens the horizon of the conventional binary classification of DWs into Bloch and Néel walls, and suggests new strategies for manipulation of domain walls and their role in electron and spin transport.
The crystal structure of Co₃Sn₂S₂ comprises quasi-2D Co₃Sn layers separated by sulfur atoms, with the magnetic Co atoms arranged on a kagomé lattice. The ferromagnetic state is characterized by strong easy-axis anisotropy favoring magnetization in the c direction, that is, perpendicular to the layers. Recent research has focused on evidence for Weyl semimetal topology in Co₃Sn₂S₂ based on a giant anomalous Hall effect in Co₃Sn₂S₂, and observation of Fermi arcs by photoemission and scanning tunneling spectroscopies. However, as the bandstructure is expected to be highly sensitive to the magnetization texture, attention has been drawn to unexpected structures in the temperature dependence near 130 K (well below the Curie temperature of 175 K) that is seen by static and ac susceptibility, muons, spin rotation, Hall effect, magneto-optic spectroscopy, and magnetic force microscopy measurements. Although these structures are relatively small in some cases, their ubiquity across laboratories and probes has led to proposals that a phase transition to a coexisting antiferromagnet or spin glass takes place deep within the dominant ferromagnetic phase. Here the scanning Kerr microscopy measurements described below show that such a phase transition does indeed occur, but within the 2D manifold of the magnetic domain walls (DWs) rather than the bulk of the 3D crystal.

**Results**

A schematic of the experimental setup is shown in Fig. 1a, which illustrates the local probing of magnetization dynamics using the polar Kerr effect (see Methods for details). Scanning the sample under the laser focus yields a map (Fig. 1b) of the Kerr ellipticity, Φ(r), revealing domains of magnetization directed parallel and anti-parallel to the surface normal, or c-axis direction. The change in Φ(r) across a DW is proportional to the zero-field magnetization (shown as a function of temperature in Fig. S1 of Supplementary Information). For local measurements of the DW dynamics, a coil surrounding the sample generates an oscillating magnetic field parallel to the easy axis, H(t) = cH_ac sin(ωt). This ac magnetic field tilts the energy landscape, inducing DW displacement in the direction that increases the volume of magnetization aligned parallel to H(t). The oscillating displacement, in turn, generates a synchronous modulation in ellipticity, δΦ_ac(r), that peaks at the domain boundaries. (Fig. S2(b) of Supplementary Information is a map of δΦ_ac(r) that shows peak contrast at the domain walls imaged in Fig. 1b).

Figure 1c–j shows a series of maps that illustrate the temperature dependence of δΦ_ac(r)/Φ_dc in the range from 120 K to the Curie temperature. Two features are immediately clear from the sequence of images. First, as the temperature is increased to the Curie temperature, each domain boundary becomes increasingly convoluted, suggesting a progressive decrease in surface tension. A second, and less expected feature is that the DWs are nearly invisible in the 140 K map, indicating a deep minimum in δΦ_ac(r)/Φ_dc and therefore the oscillating displacement Δx as a function of T.

The key advantage of the scanned local probe over global ac susceptibility measurements is the ability to determine the displacement, Δx, of individual segments of DWs. The displacement is obtained with ≈1 nm sensitivity from the ratio of δΦ_ac at the domain wall center, δΦ_ac(0), to the step in ellipticity across the DW, δΦ_ac, through the relation δΦ_ac(0)/Φ_dc = erf(Δx/√2σ), where r is the probe focal radius (see Supplementary Information Section III). Panels a–d of Fig. 2 illustrate the local dynamics of a representative segment of a DW (see Supplementary Information Section IV for details on the spatial variation of DW dynamics). Figure 2a presents an overview of δΦ_ac(0)/Φ_dc in the −T plane using a color scale; Fig. 2b shows several line cuts through the plane at constant ω. A deep minimum in δΦ_ac(0)/Φ_dc is evident in both plots. Figure 2c shows the corresponding DW displacement as a function of H_ac for several temperatures. The displacement displays a threshold, indicating collective pinning behavior; Δx(H_ac) is nearly zero below a frequency-dependent field, H_th, and increases linearly with H − H_th for H > H_th. Finally, the wall displacement measured with H_ac = 28 Oe is plotted in Fig. 2d on an expanded temperature scale that highlights the step-like change centered on T = 135 K.

The step-change in displacement near 135 K cannot be understood within the standard theory of DW dynamics, which assumes a Bloch wall texture in which M rotates with constant magnitude in the plane of the DW. In this theory Bloch DW mobility is proportional to the product of the Gilbert parameter, which describes the damping rate of transverse magnetization fluctuations, and the DW width. Both of these parameters are expected to vary smoothly and monotonically at temperatures well below Tc. While the disorder that gives rise to collective pinning could account for a monotonic decrease in mobility as T is lowered, it cannot explain the subsequent increase of DW displacement below ≈ 135 K.
Studies of the ac susceptibility, $\chi_a(T)$, of insulating hexaferrite magnets performed in the early 1990’s provide a clue as to the origin of the anomalous DW displacement in Co₃Sn₂S₂. These measurements revealed nonmonotonicity in $\chi_a(T)$, resembling the minimum in DW displacement in Co₃Sn₂S₂, but with the qualitative difference that it was found in the critical regime, $T = 0.995T_c$, rather than deep within the ferromagnetic phase. This phenomenon was traced to the prediction by Bulaevskii and Ginzburg (BG) that a competition can be quantified by free energy of the form,

$$F \propto \rho_s(VM)^2 + KM_T^2 + \frac{1}{8\chi_c}(M^2 - M_i^2)^2/M_i^2,$$

where $\rho_s$ is the spin stiffness, $K \equiv 1/2K_{an}$ is the uniaxial (easy axis) anisotropy factor, and $\chi_c$ is a susceptibility that parameterizes the energy cost of changing the amplitude of the magnetization from its equilibrium value $M_T$ and $M_i$ are the in-plane (or transverse) and saturation magnetizations, respectively. As $T \to T_c$, $\chi_c(T)$ diverges, whereas $K(T)$ does not, and therefore variations in the amplitude of $M$ cost less energy than rotations. Consequently, in a range of temperatures below $T_c$, that depends on parameters, the energy to create a linear wall is less than the energy of a Bloch wall. Minimizing the free energy in Eq. 1 yields a continuous phase transition from linear to elliptical to circular (Bloch) walls. The order parameter of the BG transition is $M_f(0)$, the amplitude of the magnetization transverse to the propagation direction evaluated at the center of the DW. This order parameter manifests in DW dynamics because propagation of a linear wall in which $M_f(0) = 0$ involves purely longitudinal changes in magnetization, whereas the motion of a Bloch wall, where $M_f(0) = M_z$, requires rotation of $M$.

The signature of such a continuous phase transition is enhanced susceptibility of the order parameter to its conjugate field, in this case a static magnetic field $H_T$ applied perpendicular to the easy axis. Figure 3a shows the DW displacement vs. temperature for $H_T = 0$, 500 Oe and 1000 Oe; Fig. 3b is a plot of $\Delta_x$ vs. $H_{ac}$ at $H_T = 28$ Oe for temperatures near the transition. Near the transition temperature a transverse field of 1000 Oe is sufficient to switch the DW from its low to high mobility state, that is, changing $M_f(0)$ from zero to $M_z$. This indicates a remarkable enhancement of transverse susceptibility, as the field required to overcome the bulk anisotropy and rotate the spins to the basal plane is ~20 T.

The free energy in Eq. 1 predicts a mean-field transition occurring when the control parameter $r = 4\chi_c/K_{ab}$ reaches unity and an order parameter critical exponent of one-half, such that $\rho \equiv M_f(0)/M_z = [1 - r(T)]^{1/2}$. However, given that the transition occurs in a two-dimensional (2D) manifold, one might anticipate strong fluctuation effects. Indeed, Lawrie and Lowe have shown theoretically that the BG transition is in the universality class of 2D Ising models in which fluctuations reduce the transition temperature and lead to an order parameter critical exponent of $1/8$. The Ising nature of the transition reflects the two discrete choices for the helicity of the Bloch wall.
To test the prediction of a 2D Ising transition within the DW, we determined $\tau(T)$ through measurements of $M$ vs. $H$ for both in- and out-of-plane directions at intervals of 1 K in the temperature range from 110 to 160 K. An example of the isothermal magnetization data at $T = 120$ K, characteristic of a ferromagnet with strong uniaxial anisotropy, is shown in Fig. 4a. The in-plane susceptibility $\chi_{ab}$ is extracted from the slope of $M_a$ vs. $H_a$ and is related to the anisotropy parameter through the relation $K = 1/2 \chi_{ab}$. The longitudinal susceptibility $\chi_{c}$ is obtained from the slope of $M_c$ vs. $H_c$ just above the saturation point (between 0.2 and 1 T). The temperature dependence of $\chi_c$ and $\chi_{ab}$ (divided by 8 for clarity) is plotted in Fig. 4b.

Figure 4c shows the local DW dynamics over the same expanded temperature range as in Fig. 2d, but now plotted as a function of $\tau(T)$ rather than $T$. To compare with a theory for the dynamics to be discussed below, we have converted the vertical axis scale from displacement, $\Delta x$, to effective mobility, $\mu_{\text{eff}}$, through the relation, $\mu_{\text{eff}} = \omega \Delta x/(H_{ac} - H_{ab})$. The observation that the transition takes place when $\tau(T)$ is of order unity gives additional strong support for the identifying the step in mobility as the BG transition. Moreover, the fact that the transition occurs at $\tau(T) \approx 0.45$ rather than precise unity suggests a reduction in critical temperature typical of 2D Ising models, where for example the $T_c$ for the Ising transition on a square lattice is reduced from the mean-field value by a factor 0.5724.

To further pursue the mean-field vs. 2D Ising comparison, we consider the rate at which the order parameter grows below the transition, using the mobility as a proxy. To do so requires a phenomenological model for the mobility as a function of the BG order parameter. We introduce the coefficients $\Gamma_L$ and $\Gamma_T$, the relaxation rate of longitudinal and transverse fluctuations of $M$, respectively, which quantify the rate at which energy is dissipated as the DW propagates. The DW mobility is obtained from the steady-state condition that the dissipation rate equals the rate at which energy is gained as the magnetization aligns with the applied field (see Supplementary Section VI for details). The resulting expression for $\mu_{\text{eff}}(\tau)$ interpolates between the mobilities of perfectly circular walls where $\mu_{\text{eff}} = w \Gamma_L/M_s$ and linear walls for which $\mu_{\text{eff}} = 3 w \Gamma_L/2 M_s$. The dashed blue line in Fig. 4c, which is a plot of $\mu_{\text{eff}}(\tau)$ assuming $\rho = (1 - \tau)^{1/8}$, shows that a mean-field description fails not only for the critical value of $\tau$, but the order parameter exponent as well. The mean-field or 3D Ising fits with lower critical values of $\tau$ near 0.5 also fail to describe the data accurately near the phase transition (see Supplementary Section VII for details).

The red dashed line, which assumes the 2D Ising critical exponent of 1/8 and a reduced critical value of $\tau$, provides a better description of the mobility data.

**Discussion**

Finally, a natural question is why a DW phase transition deep in the FM state is uniquely observed in Co$_3$Sn$_2$S$_2$ and not in other metallic uniaxial ferromagnets. We suggest that what sets this compound apart is the giant value of its dimensionless anisotropy factor, $K$. Table 1 summarizes the magnetocrystalline anisotropy of several uniaxial ferromagnets that are receiving attention for potential applications. Notice that while the dimensionful anisotropy energy, $\mu_B K M_s^2$, is similar for each compound, the
saturation magnetization of Co₃Sn₂S₂ is much smaller; consequently, the ratio of anisotropy to magnetostatic energy is anomalously large. This leads to the absence of the domain branching that complicates the structure of domain walls at the surface and disrupts the BG transition in other FM systems. The large value of $K$ likely originates from the extreme sensitivity of the low-energy electronic states to the magnetization. The giant anisotropy in Co₃Sn₂S₂, and potentially other systems in which the electron dispersion is highly sensitive to magnetization texture, may lead to new strategies for manipulation of domain walls and their role in electron and spin transport.

### Methods

**Scanning MOKE microscopy.** A linearly polarized 633 nm He/Ne laser beam was focused at normal incidence onto a 1 μm spot on the sample surface with an objective lens (Olympus LMPFLN 50×, NA = 0.5). The reflected beam was then collected by a 50:50 non-polarizing beam splitter, which directs the beam to a quarter wave plate and a Wollaston prism for balanced photodetection of Kerr ellipticity. The sample was raster scanned with a piezoelectric scanner (Attocube ANPx101) and a Wollaston prism for balanced photodetection of Kerr ellipticity. The sample stage was adjusted with piezoelectric scanners (Attocube ANPz102). For local measurements of the DW dynamics, an out-of-plane AC magnetic field was applied through a coil (Woodruff Scientific, 156 turns, inner diameter: 5 mm, height: 1.5 mm) that surrounds the sample. The modulation in Kerr ellipticity $\delta_{\text{AC}}(r)$ was measured using a standard lock-in detection at the frequency of the coil. The sensitivity of DC Kerr measurements is typically ~1 mrad, due to various sources of fluctuations. By shifting the observation frequency using modulation of $H_m$, we are able to measure Kerr effect amplitudes of less than 1 μrad.

### Data availability

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

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**Table 1 Summary of various magnetic constants in uniaxial ferromagnets.**

| Compound     | Magnetization ($M_s$) $\times 10^5$ A/m | Magnetostatic energy ($\mu_0 M_s^2$) $\times 10^5$ J/m³ | Anisotropy energy ($\mu_0 K_{eff}$) $\times 10^5$ J/m³ | Anisotropy factor (K) |
|--------------|----------------------------------------|----------------------------------------------------|----------------------------------------------------|----------------------|
| Co₃Sn₂S₂     | 0.56                                   | 0.042                                              | 3.8                                               | 90                   |
| Fe₃Ge₂Te₂     | 4.0                                    | 2.0                                                | 12.1                                              | 6.0                  |
| CrI₃         | 2.1                                    | 0.58                                               | 3.1                                               | 5.3                  |

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Author contributions
C.L. performed the optical measurements. C.L. and J.O. analyzed the data and wrote the manuscript. P.V., K.M., C.S., and C.F. grew and characterized the crystals. C.S. performed the magnetic susceptibility measurements. J.O. and J.E.M. developed the theoretical models. All authors commented on the manuscript.

Competing interests
The authors declare no competing interests.

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Correspondence and requests for materials should be addressed to Joseph Orenstein.

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