Picoscale Magnetoelasticity Governs Heterogeneous Magnetic Domains in a Noncentrosymmetric Ferromagnetic Weyl Semimetal

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Magnetic Weyl semimetals are predicted to host emergent electromagnetic fields at heterogeneous strained phases or at the magnetic domain walls. Tunability and control of the topological and magnetic properties are crucial for revealing these phenomena, which are not well understood or fully realized yet. Here, a scanning superconducting quantum interference device microscope is used to image spontaneous magnetization and magnetic susceptibility of CeAlSi, a noncentrosymmetric ferromagnetic Weyl semimetal candidate. Large metastable domains are observed alongside stable ferromagnetic domains. The metastable domains most likely embody a type of frustrated or glassy magnetic phase, with excitations that may be of an emergent and exotic nature. Evidence is found that the heterogeneity of the two types of domains arises from magnetoelastic or magnetostriction effects. It is shown how these domains form, how they interact, and how they can be manipulated or stabilized with lattice strains estimated to be on picometer levels. This knowledge can be used in designing and fabricating devices made from CeAlSi and related materials for magnetic field sensing and magnetic memory applications.

1. Introduction

Topological materials, namely the materials whose electronic structures are protected by nontrivial symmetries, have drawn great interest in the past decade. Topological insulators, with robust conducting surface states induced by the topology of the bulk band structure, led to the realization of various novel phenomena such as the quantum anomalous Hall effect. With the more recent discovery of Dirac Weyl fermions in semimetals, research focus has shifted to topological semimetals. Topological Weyl semimetals (WSM) are characterized by the linear dispersion around the band crossing points, termed the Weyl nodes. Their non-closed surface states, the Fermi arcs, and the band crossing have been experimentally observed through angle-resolved photoemission spectroscopy. Weyl nodes are protected by breaking either spatial-inversion symmetry or time reversal symmetry. Magnetic Weyl semimetals, such as antiferromagnetic semimetal Mn3Z and kagome ferromagnetic (FM) semimetal Co3Sn2S2, are particularly interesting since their topological states are easily tunable using an external magnetic field. Experimental studies show that magnetic Weyl semimetals with broken time reversal symmetry may generate strong anomalous Hall effects due to their large Berry curvature. The Nernst effect was also shown to be connected to the Berry curvature. Moreover, the magnetic textures, such as magnetic domain walls, in such materials can induce localized charges and be effectively treated as axial gauge fields. Magnetic domain walls, as a result of topological defects in the real space, may largely affect the electronic behaviors in momentum space. A closer look at domain walls at mesoscopic level is desirable to understand their dynamics, and it may lead to a practical pathway to unveil the interplay between Weyl electrons and magnetic textures.

Recently, the RAlX (R = rare earth elements, X = Ge or Si) family of compounds emerged in studies of magnetism in Weyl semimetals as they were proposed to be potential candidates of magnetic Weyl semimetals with remarkable tunability. Depending on the choice of rare earth ions, RAlX can be ferromagnetic, antiferromagnetic or nonmagnetic and hosts either type I or type II Weyl fermions. PrAlGe was expected to be a type I magnetic WSM and had indirect evidence of hosting nanoscale ferromagnetic domain walls which are tunable by changing the temperature and external magnetic field. In addition, CeAlGe was experimentally proven to be a type II antiferromagnetic...
WSM with a phase transition at 5K.\cite{27,28} A singular angular magnetoresistance study performed on CeAlGe also shows the existence of controllable high-resistance domain walls, which result from broken magnetic symmetry and Fermi surface mismatch in a nearly nodal electronic structure.\cite{29}

CeAlSi, which has the same $I4_{1}m d$ space group, was recently shown by us to be a noncentrosymmetric ferromagnetic WSM and exhibit an anisotropic anomalous Hall effect with different behaviors when measured along the easy- and hard-axes.\cite{30} The large anisotropy of bulk magnetization and the ferromagnetic domains observed in CeAlSi below $T_c = 8.3$ K in ref. \cite{30} suggest that the interplay between magnetic order and topological band structure may be responsible for a loop-shape Hall effect observed with a magnetic field along the hard-axis.

In this paper, we address a question of whether we understand the magnetic behavior of the ferromagnetic Weyl systems. Here, we study magnetic domains in CeAlSi crystals by investigating the origin of their heterogeneity and showing ways to control them. The results below will guide future efforts in building classical and quantum devices from these materials harvesting the magnetic and the electronic topological properties.

### 2. Results

Figure 1A shows typical domains in the magnetization; also visible are in-phase and out-of-phase ac magnetic susceptibility components. The DC flux images display the stray field piercing the sample surface parallel to the a-b crystallographic plane. The in-phase and out-of-phase susceptibilities show two types of domains with different strengths of response. The in-phase susceptibility shows that these domains differ in the strength of paramagnetic-like response to local field of about 0.3 G at a typical frequency of 1800 Hz. The increased out-of-phase susceptibility in some domains correspond to those domains exhibiting energy losses (hysteresis) on the timescales of the local excitation ac magnetic field (note, the negative sign of the out-of-phase component is a matter of convention\cite{31,32}).

We hypothesized that the different domains arise due to magnetoelastic and magnetostriction effects that modify some of the regions of the sample. These regions experience larger strains due to the internal magnetization interacting with the lattice; see Figure 1B. To confirm that there is such a lattice-field connection in CeAlSi crystals (see sample A shown in Figure 1C), we...
Figure 2. The growth of domains with magnetic field in a CeAlSi crystal that was as-cooled through the transition at ≈8 K. (Top panel) Upper row: dc magnetizations in flux units. Middle and lower rows: The in-phase component and the out-of-phase component of the ac susceptibility. Dark green and dark marine areas in the in-phase and out-of-phase components, respectively, correspond to stable in-plane domains. Light green and light marine areas in the in-phase and out-of-phase components, respectively, are metastable domains. The domains are lined up with the crystallographic 〈100〉 directions. (Bottom panel) The histograms quantify how the stable domains grow with field, while the metastable domains shrink (see also Figure S5, Supporting Information).

attached one of the crystals to a miniature 1 mm long strain gauge positioned inside a small solenoid along with three other gauges comprising a Wheatstone bridge. This resulted in the high accuracy resistance measurements required to detect the tiny magnetostriction effects. We observed a magnetoelastic response from the sample at low temperatures below the ferromagnetic transition (see Figure 1D). The magnetostriction response is small but clearly detectable for the limited fields we applied: on the order of $10^{-5}$ at a few tens of Gauss in-plane fields. This strain corresponds to single nanometers of the total sample length changes.

As we show in Figure 2, such small external fields suffice to substantially change the landscape of the domains. In this figure, field cooled data are shown for representative field values ranging from a single Gauss to a couple of tens of Gauss. At lower fields, a substantial fraction of metastable domains is apparent alongside small stable domains. Stable FM domains were enhanced with presence of larger external fields. The average area of stable domains grows to more than twice the zero-field domain areas, as we apply a field of just over 10 G.

Applying external field is not the only method that can be used to manipulate the domains. By cycling the sample through the transition several times in zero field, but every time lowering the
temperature to which the sample was warmed up until it reached 6 K, we were able to train the domains even without the external field. Figure 3 shows the magnetic landscapes of both the zero-field-cooled sample and the sample treated by the training process. The ferromagnetic phase was modified due to the considerable growth of the stable FM domains after the sample was trained. Figure 3 shows the histogram of the stable domains. This annealing confirms that the domains with smaller susceptibility are the thermodynamically more stable ones; hence the other domains were defined as “metastable.”

We illustrated the FM phase transition itself in CeAlSi by showing the temperature evolution of FM domains in Figure 4. The presented sample A (also see sample B in Figure S6, Supporting Information) was first zero field cooled down to 6 K and then warmed to over 9 K with images taken at several temperatures between. Metastable domains, identified by light marine (higher signal) and light green (higher negative signal) areas in the in-phase susceptibility and the out-of-phase susceptibility, grew as the temperature increased. Susceptibility signals reached a maximum value at temperatures near $T_c$, where both stable and metastable domains were no longer distinguishable, and then eventually turned into narrow features along the lower temperature domain boundaries. No strong magnetic signals or FM domains were observed at 8.26 K, consistent with the bulk transition temperature around 8.3 K.\cite{30} In addition, the observed domain boundaries are within 3 µm sharpness, which is supportive of vertical walls.

Furthermore, small areas were repeatedly scanned without an external field and showed an unusual slowly fluctuating behavior in the out-of-phase susceptibility at 6 K, see Figure 5. The wave-like features across the metastable domain were found to change their shape over the timescale of minutes and the slow-dynamics persisted over hours. The persistent dynamics of the out-of-phase component is surprising given that all the susceptibility measurements are done in the linear response regime (no dependence on the ac field amplitude, see Figure S2, Supporting Information). Typically, what fluctuates in magnetic systems is magnetization, but not susceptibility.\cite{33,34} Furthermore, we find that the so-called Cole–Cole plots (see Figure S3, Supporting Information) of the in-phase versus out-of-phase components of the magnetic susceptibility show short, incomplete arcs that are a signature of a wide distribution of activation times in a frustrated system.\cite{31,32,35}

3. Discussion

The DC magnetic flux images are consistent with the results from bulk measurements\cite{30} in terms of the direction of magnetization being in-plane (normal to the $c$-axis). The DC magnetic flux images show that the stray field penetrates out of the $a$-$b$ surface at the in-plane domain boundaries. We found that zero-field-fasted-cooled domains are rather small (a few micrometers in size) and a weak in-plane field of a few Gauss is enough to develop very large in-plane FM domains (a few hundreds of micrometers in size). The typical level of the observed DC flux variations is a few $\Phi_0$, which is consistent with the remnant magnetization determined from the $c$-axis bulk magnetization measurements.\cite{30} The remnant $a$-axis magnetization predicts DC signal on the order of...
Figure 4. The temperature evolution of the magnetic domains in the CeAlSi crystal. Upper row: dc magnetization in flux units. Middle and lower rows are the in-phase component and the out-of-phase component of the ac susceptibility. Dark green and dark marine areas in the in-phase and out-of-phase components, respectively, are stable in-plane domains. Light green and light marine areas in the in-phase and out-of-phase components, respectively, are metastable domains. The stable domains shrink, while the metastable grow toward the transition, and eventually collapse to narrow features reminiscent of the domain boundaries roughly lined up with the crystallographic \(\langle 100\rangle\) direction. The dc magnetization contrast reduces toward the transition, while the amplitude of the in-phase and out-of-phase signals grow. The metastable regions show enhanced signals. The \(\approx 8.3\) K transition temperature is in agreement with the bulk results.\(^{[30]}\) Above the bulk transition temperature of \(8.3\) K, the bulk ferromagnetic domains disappear. Some small paramagnetic (not ferromagnetic) signal persists for another \(\approx 0.5–1\) K, see Figure S1 in the Supporting Information. These might be fluctuations above the critical temperature.

hundreds of \(\Phi_0\) if the domains were to be polarized out-of-plane (note that we could not invert the DC flux images reliably to determine the exact in-plane configuration due to the intricacy of the patterns). Thus, the magnitude of the signal is consistent with the in-plane magnetization in the stable domains.

In our experiments, the in-phase c-axis AC susceptibility channel probes small canting of the in-plane atomic magnetic moments in the stable FM domains, which exhibit very small c-axis signal at 6 K. Domains identified as metastable showed larger responses, likely due to a magnetically disordered frustrated state, or due to under-resolved finer structure to be studied in future works (such as magnetic spirals or skyrmions,\(^{[36]}\) see the ripples in Figure 5). These domains’ types could be distinguished in the susceptibility channel but not in the DC magnetization, highlighting the power of our susceptibility imaging technique. The location of the domains is random with temperature cycling through the transition which rules out chemical phase separation. Interestingly, domain walls exhibited signal in the out-of-phase component, indicating that the metastable domains and domain walls have some intimate connection of yet undetermined origin.

The metastability is likely associated with the interlayer antiferromagnetic-like (partial) ordering. Our neutron scattering and bulk magnetization data\(^{[30]}\) show that the magnetization lies in the \(a-b\) crystallographic plane. In the proper ferromagnetic phase, in-plane Ce magnetic moments within the unit cell are stacked in \([\cos(45° + \theta/2) \sin(45° + \theta/2) 0]\) and \([\cos(45° - \theta/2) \sin(45° - \theta/2) 0]\) directions with the angle between the Ce moments estimated to be \(\theta = 68°.\)\(^{[30]}\) This stacking, when coarsely-grained, produces the net in-plane magnetization in the \([1 1 0]\) direction in this example (or three other degenerate directions in different proper domains). Under a built-up magnetoelastic strain, the c-axis length changes, and the interlayer interactions become modified. Hence, differently stacked configurations in terms of the Ce moment alternation may emerge, which we believe is the likely origin of the metastable domains that are identified in the susceptibility channels. Individually magnetized in-plane Ce layers can be organized in an antiferromagnetic-like sequence in the c-axis direction, or can be organized in disordered stacking, or even in a chiral (rotating) stacking order. Chiral magnetization can possibly even lead to phenomena like long-range magnetic spirals, skyrmions, and other incommensurate magnetic arrangements. This interpretation is fully consistent with our previous bulk results. These stacking orders would be impossible to distinguish in neutron scattering because even a small external magnetic field, as our current scanning superconducting quantum interference device (SQUID) work shows, can wipe out the regions with metastable stacking orders. Further, such nonferromagnetic stackings might result in a fine in-plane structure, hence the suggestion about skyrmions.

The out-of-phase AC susceptibility measurements probe dissipation effects in the “movement” of the in-plane spins. They do not only probe mere canting. The temporally changing out-of-phase component must be accompanied by some hysteresis and it is not enough to have simple canting of the spins. We notice that at low temperature, the stable FM domains show no
out-of-phase component as one expects for simple, fully reversible canting. However, the metastable regions are now more clearly identified as those that exhibit nonzero out-of-phase signal. The out-of-phase component grows in magnitude with temperature. Thus, it appears to be intimately related to the mechanism of the phase transition (see also temperature dependence of the permeability in the Supporting Information).

The magnetic phases of RAIX (R = rare-earth and X = Si, Ge) materials are known to be very sensitive to small lattice variations between the family members, even though the microscopic mechanism for such sensitivity is not known.\textsuperscript{[37–42]} Our work shows evidence that such sensitivity to strain can occur within a specific material. We probed how the size of the samples change under small in-plane field and found that in terms of the lattice deformations such magnetostriction strains could correspond to picometer level displacements for the fields used here.

Note, the off-stoichiometry of Al and Si in our material is less than 2%, a variation that produces no major changes in the magnetic behavior of the material as pointed out in ref. [30]. Bulk magnetization curves in that reference are identical between several samples grown under different conditions, but the Hall effect shows mild variations because a slight off-stoichiometry can shift the Fermi-level. However, note that magnetism comes from the \( f \) electrons of Ce that are 6 eV below the Fermi-level. Thus, a change of Fermi level by a few meV has no impact on the localized moments responsible for magnetism in CeAlSi.

The internal coarse-grained magnetization corresponds to a higher internal magnetic field: larger than our test fields in Figure 1D. We estimate the internal field (magnetic flux density, \( B \)) to be on the order of 1000 G (from the bulk results\textsuperscript{[30]}), which means that the built-in strain in or around the domains is likely an order of magnitude or two larger than the strain measured here in the \( \approx 30 \) G external field. Estimated \( 10^{-3} \) strains at internal fields of \( \approx 1000 \) G would correspond, then, to an order of a picometer distortion per unit cell.

Upon formation of internal magnetization in the stable ferromagnetic domains, the domains could build up internal strain due to the magnetostriction or magnetoelastic effects\textsuperscript{[43,44]} and effectively apply a somewhat larger strain at the surrounding boundaries, producing a different magnetic phase around them due to the excessive strain (surface-origin of the signal is ruled out based on the approach curves, see Figure S4, Supporting Information). We believe that this is the reason for the observed magnetic phase separation represented by domains with small and large magnetic AC susceptibility: one has large uniform domains and another is either frustrated or a fine-structure matrix modified by strain.

The out-of-phase component of the susceptibility is related to magnetization noise via the fluctuation dissipation theorem. In our case the out-of-phase component is itself “noisy” in the metastable domains, which is highly unusual and is probably a reflection of some magnetic frustration in the magnetic state. To the best of our knowledge the only case where susceptibility was found to be fluctuating is a system exhibiting superconductor to insulator transition, with slow quantum fluctuations in the diamagnetism.\textsuperscript{[45]} We speculate that the two material systems might be related through the role of frustration: frustration in the lattice of spins in the magnetic system and frustration in the superconducting vortices in the superconducting system.

What this combination of stable and metastable phases offers are the opportunities for combining different topologically nontrivial interfaces in different magnetic domains of the noncentrosymmetric Weyl semimetal. The metastable domains exhibit magnetic fluctuations that may be of magnetically nontrivial nature by themselves, as expected in other materials.\textsuperscript{[46,47]} Further, the coexistence of these phases may be related to the puzzling Hall effect signals in these materials.\textsuperscript{[30]} We envision the facilitation of emergent electromagnetics\textsuperscript{[48]} by controlling these phases and the interfaces between them with magnetic fields and strain knobs. While we do not claim to have demonstrated the ultimate electronic topological effects in this work, our result is a critical milestone toward this goal. Simply put, we need to know how to control and manipulate magnetic domains and boundaries in devices made from this material. The current work provides a keystone playbook for these future experiments in CeAlSi and other related materials.

4. Summary and Outlook

Future studies should focus on measuring the expression of these magnetic states in the electronic properties and on fine-tuning these phases with external strain. Building devices that will utilize the domains and domain walls to uncover useful electromagnetics behavior becomes a more realistic target with our findings: We show how magnetic domains form in CeAlSi, how they interact, and how they can be manipulated or stabilized with lattice strains estimated to be on picometer levels, which means it should be possible to induce such small strains externally using a strain cell and perhaps even enhance the ferromagnetic transition temperature.\textsuperscript{[49–52]}

Figure 5. Slow fluctuations in the out-of-phase susceptibility across one of the metastable domains. Time between images is about 10 min, each image has 75 lines, and line-time is 3.1 s. If one follows the whiter spots, they can be noticed to change from one image to another, while the apparent size of the features of the wavy pattern is similar. Such fluctuations are extremely unusual and indicate a possible switching mechanism in a magnetically frustrated system.
To make it more concrete, Figure 2 shows an extreme sensitivity of the domain wall structure to an external field. Only 2 G is enough to change the domain wall arrangement in CeAlSi. The electrical resistance of a magnetic material is expected to respond to domain wall arrangement.\(^{[33]}\) Thus, CeAlSi can be used as a platform for new type of a sensor of magnetic field or magnetic memory, specifically, for low-temperature and low-noise applications.

We have performed a comprehensive scanning SQUID study of single crystals of the ferromagnetic noncentrosymmetric Weyl semimetal candidate CeAlSi. We discovered a heterogeneous state that is formed by the proper ferromagnetic in-plane domains and a type of metastable domain characterized by energy dissipation. The latter forms most likely due to the magnetostrictive or magnetoelastic effects and enhanced strain around the ferromagnetic domains, surrounded by the matrix of a possibly magnetically frustrated configuration. Coexistence of such spatially separated strained phases will allow for the engineering of topologically nontrivial domain boundaries with possible emergent electrodynamics and new functionalities in the future. Picometer atomic displacement can lead to dramatic changes in correlated systems.\(^{[14,55]}\) The fact that such a small deformation causes a dramatic change in magnetic behavior hints that there is a proximity to some type of structural instability. Whether that instability could be related to Weyl fermions interacting with magnetic moments remains to be seen, but this is one of the more exciting directions that our work motivates.

5. Experimental Section

Imaging of magnetic domains in CeAlSi was reported using scanning SQUID microscopy (SSM). SSM is one of the most sensitive tools to measure tiny magnetic features near a sample surface. In the sensors, the gradiometric sensing pick-up loops are integrated with the well shielded body of the SQUID and are concentric with the microcoils, which allows to apply a local magnetic field to the sample.\(^{[16-58]}\) Thus, the sensors can measure and image magnetic susceptibility in addition to spontaneous magnetization or dc magnetic flux. The ability to image magnetic ac susceptibility allows for the discrimination between domains with different magnetic characters, which otherwise look practically indistinguishable in the magnetic dc signal.

In this experiment, a SQUID\(^{[46]}\) was used, which has a niobium pick-up loop with an inner diameter of 3 µm and an outer diameter of 3.5 µm and a one-turn field coil with an inner diameter of 6 µm and an outer diameter of 7 µm. Spatial resolution of this sensor is about 3–6 µm depending on the sample features (sharp changes can be resolved with better resolution). The SQUID was attached to a custom-built piezoelectric scanner. The SQUID, scanner, and samples were cooled inside a Montana Instruments Fusion closed-cycle cryostat similar to the one described by Shaper et al.\(^{[39]}\)

CeAlSi crystals were grown using a self-flux method.\(^{[30]}\) The starting materials Ce, Al, and Si were mixed in a mole ratio of 1:10:1 and placed in an alumina crucible inside an evacuated silica tube. The mixture was heated to 1000 °C for 12 h, cooled to 700 °C at 0.1 °C min\(^{-1}\), annealed at 700 °C for 12 h and then centrifuged to decant the residual Al-flux. The crystals are several milliliters wide with metallic luster and they are stable in air. The CeAlSi samples have a ferromagnetic transition temperature of \(T_{\text{N}} = 8.3\) K. The [001] surface of the samples was polished using a standard polishing jig and a diamond lapping film with a 0.5 µm grade to obtain a flat surface suitable for the scanning SQUID imaging. The structure and the ferromagnetic phase transition were previously verified by optical second harmonic generation and neutron scattering.\(^{[30]}\)

In this work, magnetostriction was measured by applying a small field in a \((1\overline{1}0)\) direction to a sample glued to a strain gauge.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Author Contributions

B.X., J.F., and I.S. performed scanning SQUID imaging experiments, H.-Y.Y. and F.T. synthesized the crystals and performed their characterization. All authors contributed to the paper preparation.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Research data are not shared.

Keywords

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