Topological magnetic hysteresis in single crystals of CeAgSb$_2$ ferromagnet

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Abstract

Closed-topology magnetic domains are usually observed in thin films and in an applied magnetic field. Here we report the observation of rectangular cross-section tubular ferromagnetic domains in thick single crystals of CeAgSb$_2$ in zero applied field. Relatively low exchange energy, small net magnetic moment, and anisotropic in-plane crystal electric fields lower the domain wall energy and allow for the formation of the closed-topology patterns. The tubular domain structure irreversibly transforms into a dendritic pattern upon cycling the magnetic field. This transition between closed and open topologies results in a ‘topological magnetic hysteresis’—the actual hysteresis in magnetization, not due to the imperfections and pinning, but due to the difference in the pattern morphology. Similar physics was suggested before in pure type-I superconductors and is believed to be a generic feature of other nonlinear single (present case), or two-phase (type-I superconductor) systems where the effects similar to demagnetization (shape-dependent macroscopic variation of properties) lead to pattern formation.

Keywords: hysteresis, crystals, ferromagnet, magnetic domain, topological hysteresis, magnetic anisotropy, crystal field

(Some figures may appear in colour only in the online journal)

1. Introduction

The interpretation of experimental data in ferromagnetic samples is often difficult because the response is determined by both the microscopic physics of the exchange interactions and the macroscopic non-uniformity of the dipolar magnetic fields surrounding always-finite samples. In fact, the latter determines the low-field behavior of magnetization in ferromagnets and superconductors [1–6]. It is well-known that in macroscopic ferromagnetic systems, the minimization of the dipolar magnetic field energy leads to the formation of ferromagnetic domains where local magnetization points in different directions, thus reducing the amplitude of the outside field beyond some distance from the surface [1–3]. The price to pay for this reduction is the domain wall energy, and the competition between these two contributions to the total free energy determines pattern morphology and size of the domains. When an external magnetic field changes, domain walls move, leading to the expansion of the domains where local magnetization points in the direction of the applied field. Similar phenomena occur in ferroelectrics [1] and superconductors [7, 8]. In all cases, the macroscopic behavior is described by the effect of demagnetization—a magnetic field distortion proportional to the sample magnetic moment [1–3, 9, 10], strictly (mathematically) applicable only to ellipsoids [1], but extendable to arbitrary shapes in a straightforward manner [9, 10].

Considering the symmetry with respect to magnetization reversal and the necessity to move the domain walls in response to the applied magnetic field, open topology patterns (laminar, labyrinth, or stripes) are usually observed in bulk ferromagnets where domain wall energy is large [1, 2]. However, in thin ferromagnetic films with perpendicular to the film plane easy axis, closed topologies (bubbles...
or tubes) are sometimes more favorable [2, 11–14], although these tubes are very different from what we observe here. For example, in cobalt films, 10 nm thick, the bubbles (tubes) are of the same size, so this is a nanoscale phenomenon in a system with extremely large demagnetizing correction [1, 2, 10, 11]. Such cylindrical magnetic domain (CMD) patterns [2], are observed in doped iron garnets, where tubular structure is induced by a static or pulsed magnetic field [12–14]. They attracted significant attention in the 1970s–1980s due to their use in mageto-optical memory devices, which was short-lived after alternative better processes were identified [15]. However, the idea of using similar tubular patterns in other systems still attracts a lot of attention, for example, in microfluidic bubble logic [16]. In our case, the novelty could be in an efficient electric currents control of magnetic tubular domain patterns in CeAgSb2, a good metal [17–19], instead of conventional CMD garnets, which are insulating [2].

In general, the difference between the energies of tubular and stripe patterns is small and depends on the details of the material parameters, such as magnetic and crystal field anisotropies, exchange energy, and geometry of the sample [1–3]. The stability of these bubbles or tubes usually requires thin-film geometry (i.e., large demagnetization) and, often, an applied bias magnetic field that lifts the above-mentioned orientational degeneracy [2]. In such a case, a dominant domain with the magnetization along the applied bias magnetic field surrounds the other type of domain (with magnetization oriented opposite to the applied field) that may exist in the form of separate flux tubes. At zero applied field, such topological imbalance was believed to be possible only as a metastable state, which could be quenched by the annealing above the Curie temperature with subsequent cooling in zero field.

In this paper, we describe a fascinating example of a closed topology (tubular) pattern in a zero magnetic field that still preserves the topological symmetry on the scale of the entire sample. Remarkably, this is observed not in a thin film but a thick single crystal of CeAgSb2. This intermetallic compound is a rare example of a ferromagnetically ordered Kondo lattice [20–22]. It exhibits many unusual properties [20–26], such as magnetic field [21, 25, 26] and pressure [21] tuned quantum critical point, strong effects of spin–orbit coupling on the magnetic ground state [23], and a possible orbital crossover in ultrafast quasiparticle dynamics [20]. Our work adds to this remarkable set of properties the observation of closed topology (tubular) patterns in a zero applied magnetic field that still preserves the topological symmetry on the scale of the entire thick crystal.

2. Experimental

Large single crystals of CeAgSb2 have been grown out of excess Sb. High-purity elements were placed in alumina crucibles with an atomic ratio of Ce: 0.045, Ag: 0.091, Sb: 0.864, and sealed in evacuated amorphous silica tubes. The ampules were heated to 1180 °C and then cooled over 100 hours to 670 °C, after which the excess Sb was decanted with the help of a centrifuge [27]. Single crystals with masses as large as ~2 grams were grown (see reference [18] for a representative picture). Previous studies reported detailed structural, magnetic, transport, thermodynamic, and quantum oscillations measurements [17, 19–26, 28].

Direct magneto-optical Kerr imaging was used to reveal the magnetic domain structure. Specifically, the sample is mounted on a copper cold-stage with its natural (as grown) [001] facet perpendicular to the propagation direction of the linearly polarized light. The copper cold-stage is located inside the optical flow-type liquid helium cryostat in vacuum. The c-axis (and, in this case, also an easy magnetic axis) is parallel to the light propagation direction and is, therefore, optimal for the magneto-optical polar Kerr effect [2]. Upon reflection, linear polarization direction (parallel to the surface) rotates by the angle proportional to the amplitude of the surface magnetization component perpendicular to the surface. Due to the chirality of the problem, opposite magnetic moments lead to the opposite directions of the polarization rotation. When viewed through an analyzer rotated almost perpendicular (but about 0.5° or so away) to the polarizer, a two-dimensional (2D) image of the magnetic pattern emerges. We emphasize this is not a scanning technique, and the entire 2D picture is acquired by the CCD camera at once. Bright regions correspond to UP domains in all images, whereas dark areas correspond to DOWN domains. A more detailed discussion of magneto-optical techniques can be found elsewhere [2, 7, 29].

Magnetic measurements were conducted using a commercial Quantum Design Magnetic Property Measurement System (MPMS).

3. Results and discussion

In general, understanding the domain patterns and their evolution with changing external parameters is essential for many reasons. Not only does it involve the microscopic physics of the material, but it is also directly linked to the coercivity and magnetic losses in time-dependent magnetic field. The measured magnetization is a sum of the projections of all local magnetic moments onto the measurement axis. Therefore, different \( M(H) \) magnetization loops will be recorded for different patterns. The magnetic hysteresis is more often than not observed in ferromagnetic samplers, and it is related to the magnetic domains and their motion [2]. Usually, it is attributed to the imperfections of the crystal lattice and various impurities (collectively known as the ‘pinning centers’) that lead to the position-dependent energy landscape that leads to the pinning of the domain walls and prevents them from moving [2, 3]. However, a different kind of so-called ‘topological hysteresis’ has been demonstrated in clean type-I superconductors, where it results from the differences in the topology of the intermediate state patterns for increasing and decreasing magnetic fields [6]. Is it possible to have a similar phenomenon in a ferromagnetic system? As we show here, the answer is a resounding yes.

Overall, this subject belongs to a more general discussion of patterns formation in highly nonlinear systems. There are many examples in chemistry, biology, astrophysics, mathematical complexity, and virtually all fields of science [30–33].
Ferromagnetic and superconducting materials are particularly attractive, because electronic and magnetic patterns can be easily manipulated and tuned by varying temperature, magnetic field, and specimen geometry. They have no inertia and are easy to reset, compared, for example, to real-life diffusive mass transport, irreversible chemical reactions, and froth coarsening. Magnetic systems can be tuned as close to the minimum energy state as experimentally possible, and in magnetic and superconducting systems, a precisely-tuned magnetic field plays the role of time in the process of coarsening (ripening) but, unlike real froth, is reversible [5, 31].

CeAgSb₂ is a ferromagnetic moderately heavy-fermion compound with Curie temperature, \( T_c = 9.8 \text{ K} \), and a small net magnetic moment of about \( 0.4 \mu_B \) on Ce\(^{3+} \) sites [17, 18, 28, 34]. Spontaneous magnetization points in the c-axis direction, which was understood within the ferromagnetic ground state in anisotropic exchange scheme [23, 28, 34]. Remarkably, there are competing, ab-plane easy axes dictated by the crystal electric field (CEF) at least in the paramagnetic state [28]. This may have profound implications on the domain wall energetics, hence patterns. Specifically, relatively small exchange energy (small \( T_c \)), small net magnetic moment, and perpendicular to magnetization direction CEF easy plane point toward relatively small domain wall energy, which favors the tubular domain structure in a system with uniaxial magnetic anisotropy.

The extremely soft nature of the magnetic domains in CeAgSb₂ became apparent when we realized that even the Earth’s magnetic field was sufficient to alter the domain pattern! In the first set of experiments, we observed the structure shown in figure 1(a). However, there is an problem with this picture—such domain pattern breaks the topological symmetry in the sense that one type of domain is not topologically equivalent to the opposite-sign domains, and this is the case on the scale of the entire sample, as shown in figure 1(c). Clearly, there is a reason to break the expected topological invariance, and, as is often the case with a very soft magnetic response, the problem is the Earth’s magnetic field. Yet, this is quite extraordinary that a minuscule 0.5 Oe magnetic field can alter the entire domain structure! With \( T_c = 9.8 \text{ K} \), the characteristic magnetic field at which the magnetostatic energy equates to thermal energy is \( B = k_B T / 0.4 \mu_B \approx 36.5 \text{ T} \), almost a million times greater than the magnetic field required to move the domains. Indeed, by applying a precisely controlled magnetic field, we could recover the topological invariance with respect to the direction of magnetization in the neighboring domains. However, as shown in figure 1(b), this symmetry is multiscale—two types of domains are observed. Each large domain contains closed-topology domains of the opposite orientation, and, together, the overall magnetic structure is symmetric with respect to the field reversal. Cooling in a slightly larger magnetic field, + 1 Oe, turns the whole picture, figure 1(c), into a negative image of figure 1(a). This highly unusual compensation at the scale of the entire sample signifies the contribution of the far regions around the sample to the overall magnetostatic energy.

Clearly, the observed domain structure is more complex than simple circular domains induced in garnets and other films. For example, in figure 1(a), the dark continuous domain contains the opposite sign, bright domain, rectangular cross-section regions with rectangular and round dots in the center. Unfortunately, we do not have a way to look into the bulk and determine whether these patterns are continuous throughout the thickness or only appear at the surface due to so-called domain branching [2]. A study similar to Nd₂Fe₁₄B with bulk x-ray spectroscopy needs to be performed to answer this question [29]. In our opinion, it is most likely that the rectangular regions are continuous flux tubes with very low domain wall energy so that they follow the in-plane crystal field anisotropy in a tetragonal crystal. Their evolution is reflected in the \( M(H) \) loops described below, causing substantial changes in the total magnetization. If they were just surface features, the total magnetization would not be affected nearly that much. Yet, the central dots in the rectangular tubes may well be the surface of inverted pyramid-shaped pockets that provide additional near-field magnetostatic compensation, similar to Landau’s idea of surface branching [1].

To the best of our knowledge, this is the first observation of such a domain pattern when a bulk ferromagnet splits into super-domains, each containing the rectangular tubes of one spin orientation embedded in a matrix of the opposite spin sign. When the external magnetic field is strictly zero, the total magnetization is zero, and topological symmetry is preserved on the scale of the entire sample, but not within each super-domain where tubular structure is favored due to low

\[
B = k_B T / 0.4 \mu_B \approx 36.5 \text{ T}.
\]
domain-wall energy compared to typical ferromagnets. This is similar to the behavior of the suprafroth in superconducting lead, which is a weak type-I superconductor with very low energy of the superconductor/normal phase interface [31].

The observation of the magnetic domains is usually performed well below the Curie temperature without monitoring their transformation across the transition, which is often too high - well above room temperature. Here we show that it is imperative to zero-field cool the sample, because otherwise the equilibrium domain structure may never be attained. In CeAgSb₂, $T_c = 9.8$ K, and we could observe these transformations. Figure 2 shows the change in the magnetic domain structure upon warming up after cooling in Earth magnetic field. Comparison of figures 1(a) and 2(a) shows that little change occurs between 5 K and 8.5 K. However, the domains become more rounded and branch out closer to $T_c$. Both effects can be linked to a further decrease of the magnetic anisotropy along the $c$-axis and, perhaps, the desire of the ordered moment to follow the in-plane CEF. Notably, the domain structure changes and never recovers when the magnetic field is cycled at temperatures below $T_c$. It makes us wonder, how many possible domain patterns were missed in ferromagnetic materials with $T_c$ too high for such experiments?

Let us now examine the correlation of the global magnetic response and mesoscopic domain patterns. Figure 3 shows the evolution of the total magnetic moment presented in the units of $\mu_B$ per Ce measured at 5 K starting in a fully demagnetized state after zero-field cooling in a true zero field, then ramping the magnetic field to $+500$ Oe, beyond the saturation around 300 Oe, then reducing it to $-500$ Oe, and finally returning to $+500$ Oe. This is a classical magnetization $M(H)$ loop with a pristine branch [3]. The characteristic images of the magnetic domains are shown at the indicated magnetic field values. Magnetic domains growth after zero-field cooling appears to proceed in a more ordered fashion than domains shrinkage when the magnetic field is reduced after reaching saturation field. The difference is that in the former case of entering the magnetic field, the up-domains (collinear with the applied field) form a stable closed topology of tubes embedded in the anti-domain (anti-collinear) matrix. These tubular domains simply grow, absorbing the neighbors. Their size increases until a fully polarized state is reached. There is no reason for such tubes to break apart, so the topology remains tubular. In stark contrast, reducing the applied magnetic field requires anti-domains to appear, which has a higher probability of happening at the edges due to non-uniformity of the magnetic field due to demagnetizing effects. Therefore, these domains channel inside starting from the edges in a labyrinth way, preventing uniform tubular positive domains from forming and shrinking.

This hysteresis is further investigated in figure 4, which examines the magnetic field dependence of global magnetization obtained differently. Here we arrived at each point of fixed $T$ and $H$ on the $M(H)$ loop following three different paths in the $T-H$ phase diagram. The first two are from the conventional $M(H)$ hysteresis loop measured at a constant temperature, shown in figure 4 by triangles and squares.
The yellow squares show the ‘initial’ magnetization measured in an increasing magnetic field after cooling in true zero magnetic field (ZFC) to $T = 5$ K. This path is accompanied by the gradual transformation of the tubular domain structure shown in figure 3. The green triangles in figure 4 show the same branch of a full $M(H)$ loop after magnetization in large positive and large negative magnetic fields. The third protocol, shown by the red stars, is $M(H)$ measured after cooling in the indicated magnetic field from above $T_c$ to 5 K. Obviously, after saturated uniform state, an equilibrium tubular structure is triggered below 200 Oe, and the magnetization there matches exactly that obtained after ZFC pristine state! This provides crucial evidence that the tubular structure is the equilibrium state of this ferromagnetic system. The difference between the green triangles in the descending branch and red circles is the topological hysteresis in the sense that the macroscopic magnetization hysteresis depends on the topology of the domain pattern, which in turn is determined by the path to a given point in the $T$–$H$ phase diagram.

In conclusion, we discovered the rectangular tubular pattern of the magnetic domains in thick ferromagnetic crystals of CeAgSb$_2$, which represent the equilibrium (minimum energy) topology after cooling in an actual magnetic field. This result proves that such closed-topology patterns are not the property of thin-film geometry, applied magnetic field, or the result of a particular metastable state but may be formed if domain wall energy is unusually small and there are competing anisotropic contributions to total magnetostatic energy.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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