Hysteretic magnetoresistance and unconventional anomalous Hall effect in the frustrated magnet TmB₄

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We study TmB₄, a frustrated magnet on the Archimedean Shastry-Sutherland lattice, through magnetization and transport experiments. The lack of anisotropy in resistivity shows that TmB₄ is an electronically three-dimensional system. The magnetoresistance (MR) is hysteretic at low temperature even though a corresponding hysteresis in magnetization is absent. The Hall resistivity shows unconventional anomalous Hall effect (AHE) and is linear above saturation despite a large MR. We propose that complex structures at magnetic domain walls may be responsible for the hysteretic MR and may also lead to the AHE.

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Geometric frustration in magnetic systems arises from competing magnetic interactions that cannot be satisfied simultaneously and leads to a variety of exotic ground states [1]. While insulating frustrated materials are well studied, metallic systems have received less attention [2]. In metallic materials, the conduction electrons mediate interactions between the magnetic moments. Additionally, the transport properties in such systems can be strongly influenced by the magnetic structure [1]. This interplay between magnetism and charge can be exploited in two ways: to engineer a highly field-tunable response of the transport properties [3] or to use transport experiments as an indirect probe of the complex magnetic structures that arise in such systems [4,5].

The rare-earth tetraboride family (RB₄, R is a rare earth) is a series of metallic frustrated magnets. RB₄ crystallizes in a tetragonal structure (space group P4/mmm, 127) [6], consisting of alternating layers of R and B ions [Fig. 1(a)]. The R ions form a frustrated Shastry-Sutherland lattice (SSL) with competing interactions J₁ and J₂ [7]. Quite remarkably, high-resolution structural refinement of LaB₄ [8] and HoB₄ [9] show that the R-R bonds corresponding to J₁ and J₂ appear equal in length, making the R sublattice a rare physical realization of one of the eleven Archimedean lattices [10] [Fig. 1(b)]. While other frustrated Archimedean lattices such as the triangular and Kagomé lattices are well studied [10,11], the RB₄ family is the only known realization of the Archimedean Shastry-Sutherland lattice.

In this article, we use magnetization and transport experiments to study TmB₄, a member of the RB₄ family that has attracted attention for its rich phase diagram [13–16] [Fig. 1(c)]. Crystal field effects at the Tm³⁺ sites (site symmetry mm) lift the degeneracy of the J = 6 multiplet and the ground state is the doublet MJ = ±6 [14]. A strong Ising anisotropy is present [17] and the interactions between the Tm³⁺ spins consist of both direct exchange and Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions. Below TN₁ = 9.7 K, an antiferromagnetic Néel phase is stable and the magnetization shows a striking field dependence: a wide half plateau is present at M/Msat = 1/2 (Msat is the saturation magnetization of 7 μB/Tm) and a narrow hysteretic fractional plateau at M/Msat ∼ 1/8 [14,15,18]. Between TN₁ = 11.7 K and TN₂, neutron scattering experiments find two long-range modulated phases, MP1 and MP2 [16]. While MP1 can be indexed by a single modulation vector of periodicity ∼8 unit cells (u.c.), MP2 requires an additional modulation of ∼80 u.c. [16]. Frustration in TmB₄ is reflected in the moderately large frustration parameter [15,19] and in the appearance of a diffuse peak in neutron scattering above TN₂ [16], indicative of short-range order. In the temperature range TN₁ > T > TN₂, the diffuse peak coexists with the sharp peaks from MP1 and MP2 [16].

Theoretical models for TmB₄, focused on explaining the unusual plateau structure, have assumed a two-dimensional (2D) nature (in analogy to another SSL compound SrCu(BO₃)₂ [20]). While a 2D SSL in the Ising limit cannot have a half plateau [21], several groups have demonstrated the existence of a half plateau by considering longer-range interactions [22–25]. Even so, the modulated phases and the fractional plateau remain unexplained, despite the relatively simple structure of TmB₄ and intense theoretical effort [21–25].

Here we present a combined transport and magnetization study of TmB₄. By measuring the resistivity anisotropy, we find that TmB₄ is an electronically three dimensional (3D) system. We find unusual hysteretic magnetoresistance (MR), which may arise from complex structures at magnetic domain walls. We further find the presence of an unconventional anomalous Hall effect (AHE).

Methods. TmB₄ single crystals were synthesized by solution growth method using an Al flux and oriented using x-ray diffraction in the Laue geometry to within ±5° [12]. Quantum Design (QD) MPMS XL SQUID magnetometer was used for magnetization measurements and QD PPMS for transport experiments [12]. Since the magnetization in the fractional plateau phase is known to vary with field history [14,18], a protocol was developed that reproduces the same magnetization curve at 2 K when the measurement is repeated [12].

Results. An examination of the in plane and out of plane longitudinal resistivities [ρzz, and ρzz, Fig. 2(a)] reveals two
FIG. 1. (a) Crystal structure of RB₄. The R and B layers are labeled. (b) The R sublattice viewed along the c axis, showing the Archimedean Shastry-Sutherland lattice. (c) Phase diagram of TmB₄ as determined from our data [12].

key features. First, ρₓₓ and ρzz show a significant drop at Tₜ₁ and Tₜ₂ due to decrease in scattering from disordered spins. Second, both ρₓₓ and ρzz are very similar in magnitude and T dependence. The second result is in sharp contrast with the assumption of TmB₄ being a quasi-2D system [22–25]. To rule out a possible misalignment, we confirmed the orientation of the crystal used for c-axis transport after the experiments [12]. We conclude that TmB₄ is an electronically 3D system. This result is expected from the 3D crystal structure: the smallest distance between the Tm ions along the c axis is 3.987 Å while the corresponding in plane distance is 3.64 Å [6]. Further support comes from band structure calculations [27] and quantum oscillation measurements on the related compound YB₄ [28], which show that the Fermi surface is 3D.

The isotropic nature of the resistivity implies that the out of plane magnetic interactions between Tm spins are non-negligible in comparison to the in plane interactions J₁ and J₂. Future theoretical models must take this result into consideration. We suggest that an anisotropic Kondo lattice model, similar to that used for β-YbAlB₄ [29], may be more appropriate for TmB₄, although further experiments are needed to establish such a picture.

The in-plane Hall resistivity [ρₓᵧ, Fig. 2(b)] decreases at high temperature but shows a sharp upturn at Tₜ₁ and a change of slope at Tₜ₂. To investigate this unusual behavior in ρₓᵧ, we measured the magnetic field dependence of M, ρₓₓ, and ρₓᵧ at three temperature regimes: T < Tₜ₂ (2K), Tₜ₂ < T < Tₜ₁ (10.5K), and T > Tₜ₁ (15K), shown in Fig. 3.

The magnetization at 2 K, shown in Fig. 3(a) as a function of magnetic flux density B = μ₀H + M [12], displays the previously reported plateau structure [13–15]. ρₓₓ at 2 K [Fig. 3(b)], shows features at the magnetic transitions indicating a strong influence of the magnetic structure on ρₓₓ. Similar features have been observed in other metallic magnets such as SiCo₆O₁₁ [30] and RNi₂Ge₂ [31]. Surprisingly, ρₓₓ shows a strong hysteresis at all magnetic fields below saturation, including zero field, even though the magnetization shows a noticeable hysteresis only at the fractional plateau.

The vanishing of hysteresis in MR above saturation allows us to exclude nonmagnetic explanations such as structural defects and extrinsic impurities. Hysteretic MR has previously
been observed in phase-separated perovskite manganites [32] and ferromagnets such as Fe\textsubscript{1/4}TaS\textsubscript{2} [33], where it is the result of a change in the bulk magnetic structure. The presence of a hysteresis in MR with no corresponding hysteresis in magnetization is counterintuitive (because the lack of hysteresis in the magnetization suggests that the magnetic structure remains the same). We return to this result later.

We now examine the Hall resistivity in TmB\textsubscript{4}. Conventionally, the Hall resistivity of a magnetic material can be decomposed into its ordinary contribution, which depends on $B$ [12], and an anomalous contribution, which depends on $M$ and the scattering rate (through $\rho_{xx}$) [34]:

$$\rho_{xy} = R_0 B + (a\rho_{xx} + b\rho_{xx}^2)M,$$

where $R_0$ is the ordinary Hall coefficient and $a$ and $b$ are constants. The second term ($\rho_{xy} \sim \rho_{xx} M$) is due to the skew scattering mechanism [35,36], while the third term ($\rho_{xy} \sim \rho_{xx}^2 M$) is a combination of intrinsic AHE and side jump mechanisms [37–39]. By comparing our data to Eq. (1), we can test if the AHE in TmB\textsubscript{4} can be explained by conventional theories. While some of the magnetic phases, especially the fractional plateau phase, extend over a narrow $H$ range to allow a definite comparison, our conclusions remain unaffected.

$\rho_{xy}$ at 2 K [Fig. 3(c)] consists of regions of linear behavior separated by sharp jumps and shows hysteresis between 1.4 T and 2.5 T. We notice that $\rho_{xy}$ does not scale with magnetization. As we go from the Néel phase (brown) to the fractional plateau phase (green), the magnetization increases and $\rho_{xy}$ shows a corresponding increase. However, as we reach the half plateau (orange), $\rho_{xy}$ drops. Saturation (white) leads to an even larger drop in $\rho_{xy}$. Moreover, $\rho_{xy}$ is linear above saturation despite the presence of a large, nonsaturating MR. This result shows that ordinary contributions to $\rho_{xy}$ dominate above saturation and conventional contributions to AHE are negligibly small [$a \simeq 0, b \simeq 0$ in Eq. (1)]. A best fit of the down sweep to Eq. (1), while showing good agreement between 2T and 4T, deviates significantly below 2 T and is strongly nonlinear above saturation (Fig. S7 in Ref. [12]).

The magnetic and transport properties of MP1 are qualitatively similar to those of MP2 [12] and we focus our analysis...
on the latter. At 10.5 K, the long-range modulation of MP2 disappears at 1.6 T and the magnetization saturates at ∼7 T [Fig. 3(d)]. ρxy shows a sharp kink at 1.6 T, then a broad hump at ∼4 T before finally becoming linear above saturation [Fig. 3(f)]. Considering the behavior of M and ρxy [Fig. 3(e)], both of which do not show a hump, conventional contributions to AHE cannot lead to the observed ρxy. Despite the presence of a strong MR above saturation, ρxy is linear, indicating that conventional contributions to AHE can be neglected. A best fit of ρxy to Eq. (1) deviates strongly from the measured data [Fig. 3(f)].

At $T > T_{N1}$, no long-range magnetic order is present and $M$ [Fig. 3(g)] increases smoothly until the maximum measured field. Both $\rho_{xx}$ and $\rho_{xy}$ at 15 K [Figs. 3(h)–3(i)] are very similar to the corresponding curves at 10.5 K, despite the absence of long-range order at 15 K. $\rho_{xy}$ shows a kink at 1 T and a broad hump at ∼5 T before becoming linear above saturation. Using the same arguments as those at 10.5 K, we conclude that conventional contributions to AHE are negligibly small at 15 K and a best fit of $\rho_{xy}$ to Eq. (1) deviates strongly from the measured data [Fig. 3(i)].

An unusual feature common to the $\rho_{xy}$ data at all three temperatures is the nonzero intercept of the linear fit above saturation. However, the slope of linear fit to the $\rho_{xy}$ data is comparable at all three temperatures (Sec. IX in Ref. [12]). The carrier concentration calculated at 2 K matches well with the value at 300 K (where no AHE is expected to be present) as well as the experimentally measured value on the nonmagnetic compound YB4 (Sec. IX in Ref. [12]). This correspondence suggests that the high-field behavior of $\rho_{xy}$ is the sum of a linear contribution from ordinary Hall effect and a constant term.

Discussion. The MR of TmB4 shows strong hysteresis at 2 K despite the absence of corresponding hysteresis in the magnetization. We suggest that subtle changes occur in the magnetic structure of TmB4 that strongly influence the MR but not the bulk magnetization. Neutron scattering experiments have shown that the magnetic structure in the modulated and the plateau phases consists of stripes or domains [14–16]. However, the microscopic structure at the domain walls is unknown. The domain walls could contain unusual magnetic structures or disordered spins or both, a possibility not considered in previous studies on TmB4. Changes in those structures can lead to a hysteretic MR while leaving the bulk magnetization unaffected.

By considering the behavior of Hall resistivity above saturation, we find that conventional contributions to AHE are negligibly small in TmB4. Therefore, all deviations from the ordinary, linear field dependence are due to unconventional mechanisms. One possibility is topological Hall effect (THE) where conduction electrons moving through a noncoplanar structure accumulate a Berry phase due to net spin chirality leading to a Hall contribution. However, neutron scattering experiments on TmB4 have not found any evidence for a global noncoplanar structure [14,16]. We suggest that noncoplanar structures could arise at domain walls, which in turn lead to both hysteretic MR and THE. Further experiments are necessary to confirm this hypothesis. Above saturation, the magnetic structure is coplanar and any potential THE contributions must be zero. In contrast, our data shows that a constant term is present. Therefore, additional contributions to AHE must be present. Other possibilities are AHE arising from phonons and spin waves [34,40]. Further work is necessary to determine if they can account for the measured $\rho_{xy}$ in TmB4.

In conclusion, we discovered that TmB4, and likely other RB4, are electronically 3D systems and future theoretical models must take this result into consideration. Our hysteretic MR results suggest that complex structures arise at magnetic domain walls that strongly affect the transport properties. Our Hall resistivity results show the presence of AHE. Further analysis reveals that conventional contributions to the AHE are negligible and hence unconventional contributions must be present. A combination of high-resolution neutron scattering, microscopic experiments, and theoretical modeling are required to determine the magnetic structure and the origin of unconventional AHE in TmB4.

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[26] Considerable uncertainties in measuring the distance between the electrical contacts, because of the small size of the samples, lead to an error bar of 20% on the absolute values of all transport quantities. Our conclusions are unaffected by this error.