5-1-2016

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

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Abstract

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.

Disciplines

Condensed Matter Physics

Comments

This article is published as Sunku, Sai Swaroop, Tai Kong, Toshimitsu Ito, Paul C. Canfield, B. Sriram Shastry, Pinaki Sengupta, and Christos Panagopoulos. "Hysteretic magnetoresistance and unconventional anomalous Hall effect in the frustrated magnet TmB 4." *Physical Review B* 93, no. 17 (2016): 174408. DOI: 10.1103/PhysRevB.93.174408. Posted with permission.

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Hysteretic magnetoresistance and unconventional anomalous Hall effect in the frustrated magnet TmB_4

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(Received 21 May 2015; revised manuscript received 7 February 2016; published 11 May 2016)

We study TmB_4, a frustrated magnet on the Archimedean Shastry-Sutherland lattice, through magnetization and transport experiments. The lack of anisotropy in resistivity shows that TmB_4 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.

DOI: 10.1103/PhysRevB.93.174408

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 (RB_4, R is a rare earth) is a series of metallic frustrated magnets. RB_4 crystallizes in a tetragonal structure (space group *P4/mbm*, 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 $\mathcal{J}_1$ and $\mathcal{J}_2$ [7]. Quite remarkably, high-resolution structural refinement of LaB_4 [8] and HoB_4 [9] show that the R-R bonds corresponding to $\mathcal{J}_1$ and $\mathcal{J}_2$ 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 RB_4 family is the only known realization of the Archimedean Shastry-Sutherland lattice.

In this article, we use magnetization and transport experiments to study TmB_4, a member of the RB_4 family that has attracted attention for its rich phase diagram [13–16] [Fig. 1(c)]. Crystal field effects at the Tm^{3+} sites (site symmetry mm) lift the degeneracy of the $J = 6$ multiplet and the ground state is the doublet $M_J = \pm 6$ [14]. A strong Ising anisotropy is present [17] and the interactions between the Tm^{3+} spins consist of both direct exchange and Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions. Below $T_{N2} = 9.7$ K, an antiferromagnetic Néel phase is stable and the magnetization shows a striking field dependence: a wide half plateau is present at $M/M_{sat} = 1/2$ (M_{sat} is the saturation magnetization of 7 μ_B/Tm) and a narrow hysteretic fractional plateau at $M/M_{sat} \sim 1/8$ [14,15,18]. Between $T_{N1} = 11.7$ K and T_{N2}, 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 $\sim$8 unit cells (u.c.), MP2 requires an additional modulation of $\sim$80 u.c. [16]. Frustration in TmB_4 is reflected in the moderately large frustration parameter [15,19] and in the appearance of a diffuse peak in neutron scattering above T_{N2} [16], indicative of short-range order. In the temperature range $T_{N1} > T > T_{N2}$, the diffuse peak coexists with the sharp peaks from MP1 and MP2 [16].

Theoretical models for TmB_4, focused on explaining the unusual plateau structure, have assumed a two-dimensional (2D) nature (in analogy to another SSL compound $SrCu(BO_3)_2$ [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 TmB_4 and intense theoretical effort [21–25].

Here we present a combined transport and magnetization study of TmB_4. By measuring the resistivity anisotropy, we find that TmB_4 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. TmB_4 single crystals were synthesized by solution growth method using an Al flux and oriented using x-ray diffraction in the Laue geometry to within $\pm 5^\circ$ [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 [ρ_{xx} and ρ_{zz}, Fig. 2(a)] reveals two

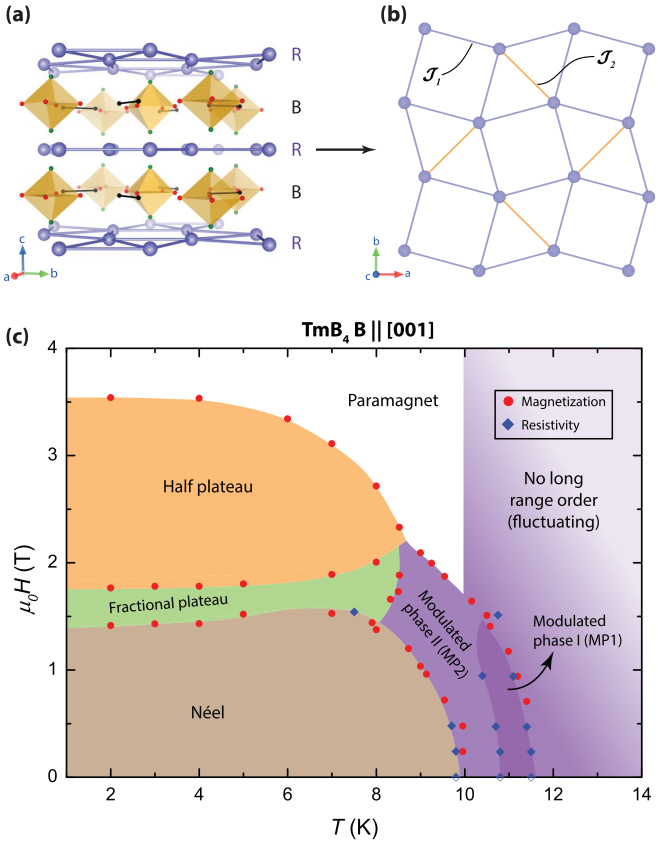

FIG. 1. (a) Crystal structure of RB$_4$. The R and B layers are labeled. (b) The R sublattice viewed along the c axis, showing the Archimedean Shastry-Sutherland lattice. (c) Phase diagram of TmB$_4$ as determined from our data [12].

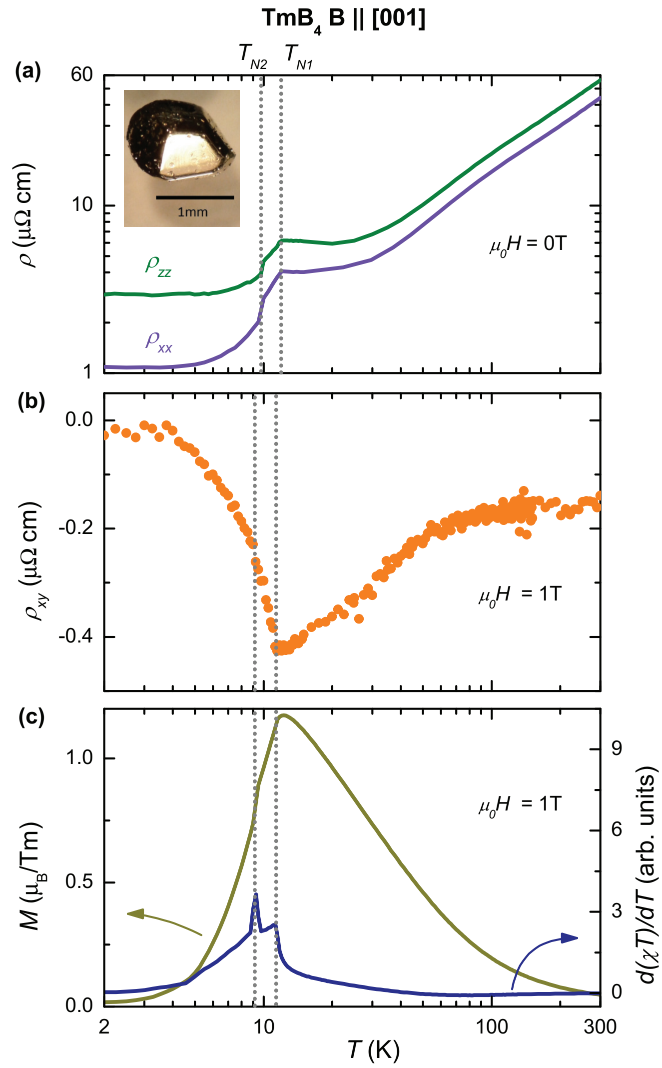

FIG. 2. (a) In-plane (ρ_{xx}) and out-of-plane (ρ_{zz}) longitudinal resistivities at zero field. Inset: photograph of a TmB$_4$ single crystal used in our experiments. The c axis is perpendicular to the shiny facet. (b) In-plane Hall resistivity ρ_{xy} at $\mu_0 H = 1$T. (c) Magnetization M and $d(\chi T)/dT$ (χ is the dc susceptibility) at $\mu_0 H = 1$T. The magnetic field is not corrected for demagnetization. Vertical dotted lines represent T_{N1} and T_{N2}. We estimate an error of 20% on the absolute values of ρ_{xx}, ρ_{xy} and ρ_{zz} [26].

key features. First, ρ_{xx} and ρ_{zz} show a significant drop at T_{N1} and T_{N2} due to decrease in scattering from disordered spins. Second, both ρ_{xx} and ρ_{zz} are very similar in magnitude and T dependence. The second result is in sharp contrast with the assumption of TmB$_4$ 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 TmB$_4$ 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 3.987Å 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 YB$_4$ [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 $\mathcal{J}_1$ and $\mathcal{J}_2$. Future theoretical models must take this result into consideration. We suggest that an anisotropic Kondo lattice model, similar to that used for β-YbAlB$_4$ [29], may be more appropriate for TmB$_4$, although further experiments are needed to establish such a picture.

The in-plane Hall resistivity [ρ_{xy}, Fig. 2(b)] decreases at high temperature but shows a sharp upturn at T_{N1} and a change of slope at T_{N2}. To investigate this unusual behavior in ρ_{xy}, we measured the magnetic field dependence of M, ρ_{xx}, and ρ_{xy} at three temperature regimes: $T < T_{N2}$ (2K), $T_{N2} < T < T_{N1}$ (10.5K), and $T > T_{N1}$ (15K), shown in Fig. 3.

The magnetization at 2 K, shown in Fig. 3(a) as a function of magnetic flux density $B = \mu_0 H + M$ [12], displays the previously reported plateau structure [13–15]. ρ_{xx} at 2 K [Fig. 3(b)], shows features at the magnetic transitions indicating a strong influence of the magnetic structure on ρ_{xx}. Similar features have been observed in other metallic magnets such as SrCo$_6$O$_{11}$ [30] and RNi$_2$Ge$_2$ [31]. Surprisingly, ρ_{xx} 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

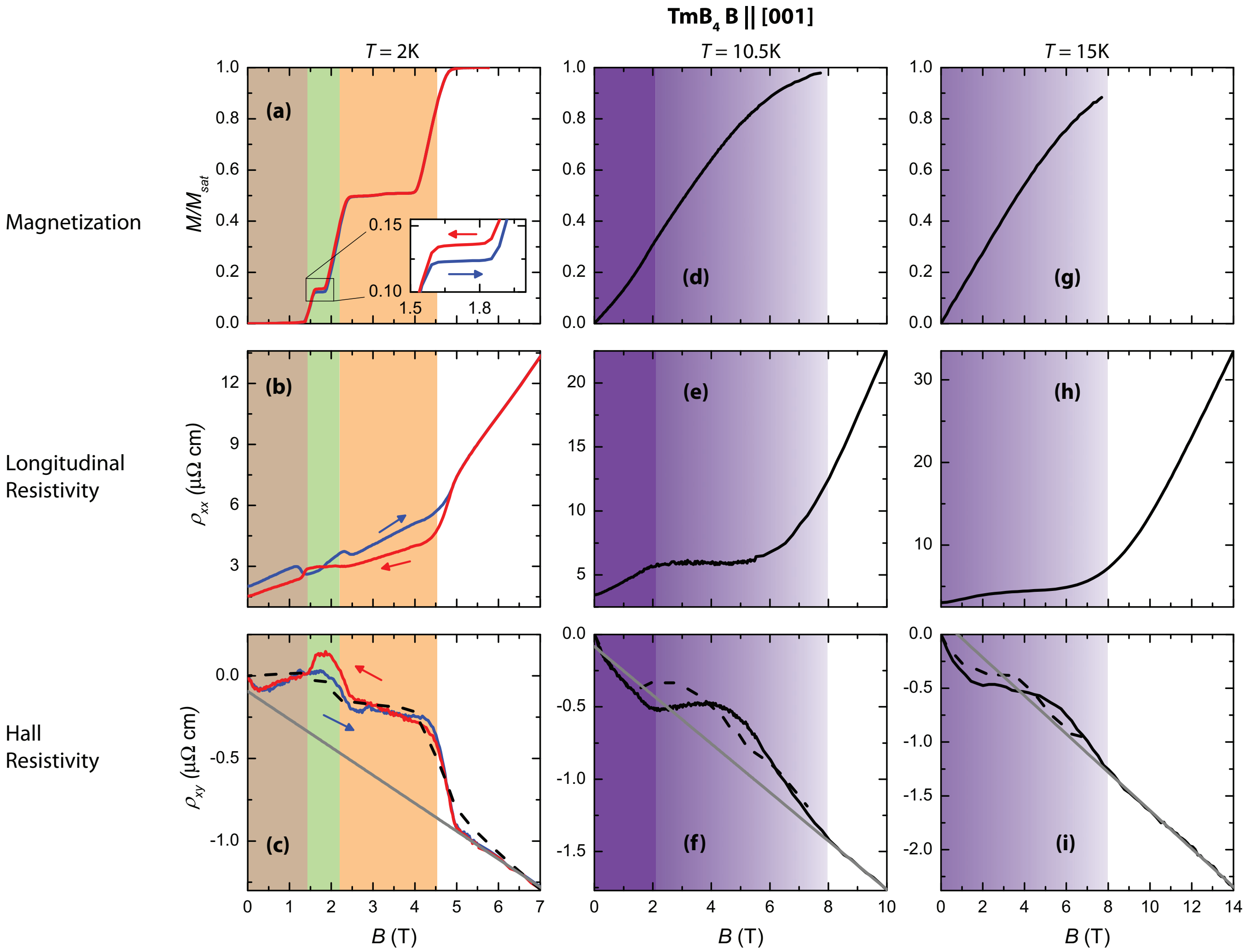

FIG. 3. (a)–(c) M, ρ_{xx}, and ρ_{xy} at 2K. (d)–(f) M, ρ_{xx}, and ρ_{xy} at 10.5K. (g)–(i) M, ρ_{xx}, and ρ_{xy} at 15K. The dashed lines are best fits to conventional AHE theories [Eq. (1)] and the solid gray lines are linear fits to ρ_{xy} above saturation. At 2K, the best fit is to the down sweep. The colored backgrounds correspond to different magnetic phases [Fig. 1(c)].

been observed in phase-separated perovskite manganites [32] and ferromagnets such as $Fe_{1/4}TaS_2$ [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_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 ρ_{xx}) [34]:

$$\rho_{xy} = R_0 B + \left(a\rho_{xx} + b\rho_{xx}^2\right)M, \qquad (1)$$

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_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.

ρ_{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 ρ_{xy} does not scale with magnetization. As we go from the Néel phase (brown) to the fractional plateau phase (green), the magnetization increases and ρ_{xy} shows a corresponding increase. However, as we reach the half plateau (orange), ρ_{xy} drops. Saturation (white) leads to an even larger drop in ρ_{xy}. Moreover, ρ_{xy} is linear above saturation despite the presence of a large, nonsaturating MR. This result shows that ordinary contributions to ρ_{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)]. ρ_{xy} 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 ρ_{xx} [Fig. 3(e)], both of which do not show a hump, conventional contributions to AHE cannot lead to the observed ρ_{xy}. Despite the presence of a strong MR above saturation, ρ_{xy} is linear, indicating that conventional contributions to AHE can be neglected. A best fit of ρ_{xy} 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 ρ_{xx} and ρ_{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. ρ_{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 ρ_{xy} to Eq. (1) deviates strongly from the measured data [Fig. 3(i)].

An unusual feature common to the ρ_{xy} data at all three temperatures is the nonzero y intercept of the linear fit above saturation. However, the slope of linear fit to the ρ_{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 YB_4 (Sec. IX in Ref. [12]). This correspondence suggests that the high-field behavior of ρ_{xy} is the sum of a linear contribution from ordinary Hall effect and a constant term.

Discussion. The MR of TmB_4 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 TmB_4 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 TmB_4. 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 TmB_4. 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 TmB_4 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 ρ_{xy} in TmB_4.

In conclusion, we discovered that TmB_4, and likely other RB_4, 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 TmB_4.

Acknowledgments. We thank Y. Ozaki for technical assistance in crystal alignment. S.S.S. thanks Tanmoy Das and Anjan Soumyanarayanan for helpful discussions. Work in Singapore was supported by Grant No. MOE2011-T2-1-108 from the Ministry of Education, Singapore and the National Research Foundation (NRF), NRF-Investigatorship (NRF-NRFI2015-04). Work at Ames Laboratory was supported by the U.S. Department of Energy, Office of Basic Energy Science, Division of Materials Sciences and Engineering. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. DE-AC02-07CH11358. Work at UCSC was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences under Award No. FG02-06ER46319.

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