

# Electronic transport on the Shastry-Sutherland lattice in Ising-type rare-earth tetraborides

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In the presence of a magnetic field frustrated spin systems may exhibit plateaus at fractional values of saturation magnetization. Such plateau states are stabilized by classical and quantum mechanisms including order by disorder, triplon crystallization, and various competing order effects. In the case of electrically conducting systems, free electrons represent an incisive probe for the plateau states. Here we study the electrical transport of Ising-type rare-earth tetraborides  $RB_4$  ( $R = \text{Er, Tm}$ ), a metallic Shastry-Sutherland lattice showing magnetization plateaus. We find that the longitudinal and transverse resistivities reflect scattering with both the static and the dynamic plateau structure. We model these results consistently with the expected strong uniaxial anisotropy on a quantitative level, providing a framework for the study of plateau states in metallic frustrated systems.

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## I. INTRODUCTION

Geometrically frustrated lattices play host to a number of emergent quantum mechanical phases including quantum spin liquids [1], resonating valence bond states [2], and complex magnetic orders [3]. Such systems are typically electronic insulators constructed from low-connectivity lattices that enforce competing magnetic interactions and enhanced quantum mechanical fluctuations [4]. While in many cases the introduction of charge carriers destabilizes such lattice-borne frustration, recently a variety of frustration-related effects have been discussed in this context in a class of materials termed frustrated metallic systems [5]. Examples include kagome lattice model realizations of the fractional quantum Hall effect [6] and superconductors with exotic pairing symmetries [7,8]. To what extent such phenomena can be realized in experiments is an open question.

A known materials system that has both lattice frustration and itinerant electronic behavior is the rare-earth ( $R$ ) tetraboride  $RB_4$ . The system is tetragonal (space group  $P4/mbm$ ) with magnetic  $R$  ions embedded in a boron network and forming a lattice topologically equivalent to the Shastry-Sutherland lattice (SSL) in the  $ab$  plane shown in Fig. 1(a). In the  $ab$  plane of the unit cell there are four  $R$  ions, located at  $(0.318a, 0.818a)$ ,  $(0.182a, 0.318a)$ ,  $(0.818a, 0.682a)$  and  $(0.682a, 0.182a)$ , where  $a$  is the lattice parameter (the complete boron network is described elsewhere [9]). While the  $4f$  electrons of the  $R$  ions are localized in a frustrated configuration, the  $5d$  electrons from  $R$  and  $2p$  from B act as itinerant carriers [9]. As with other SSL systems, the key parameters determining the frustration are the antiferromagnetic exchange  $J_1$  and  $J_2$  ( $J_1, J_2 > 0$ ) on diagonal and square bonds on alternating tiles [10]. Unlike the celebrated case of quantum spin-1/2  $\text{Cu}^{2+}$  ions in the insulating compound  $\text{SrCu}_2(\text{BO}_3)_2$ , which realizes the collective dimer singlet ground state predicted for the SSL [10,11],  $RB_4$  has large classical  $f$  moments with magnetic interactions mediated by itinerant electrons. Despite this, just as  $\text{SrCu}_2(\text{BO}_3)_2$  exhibits a series of fractional magnetization plateaus as a function of the magnetic field  $H$  with  $M/M_S = 1/q$  ( $q$  here is an integer from 2 to 9,  $M$  is the

magnetization, and  $M_S$  is the saturation  $M$ ) [11–14],  $RB_4$  also shows magnetization plateaus of unusual structure [15–18]. A particularly interesting limit is the trivalent  $R = \text{Er}$  and  $\text{Tm}$ , where a strong Ising single-ion anisotropy exists such that the  $f$ -electron moments may be described within up/down twofold degrees of freedom locked perpendicular to the SSL plane and the plateau transitions arise from complex spin-flip processes [19–22].

Herein we investigate how static and dynamic aspects of the magnetism in Ising-like  $RB_4$  influence transport and the view it offers into the energetics of the classical SSL magnetic phase diagram. The SSL network for  $\text{ErB}_4$  and  $\text{TmB}_4$  along with their Ising-type antiferromagnetic (AFM) ground states are shown in Figs. 1(b) and 1(c), respectively [16,23]. The  $a$  and  $c$  lattice constants are 7.071 and 4.000 Å, respectively, for  $\text{ErB}_4$  and 7.057 and 3.987 Å, respectively, for  $\text{TmB}_4$ . In both materials the magnetic structure repeats uniformly in layers along the  $c$  axis [9,18]. One view of the difference between the two systems is the connectivity of the spins: in  $\text{ErB}_4$  the spins on the diagonal bonds are antiparallel, while in  $\text{TmB}_4$  they are parallel. This can be understood in terms of exchange interactions, as while both compounds have  $J_1 \approx J_2 > 0$ , they differ in further neighbor interactions [16,21]. With  $H \parallel c$ , in  $\text{ErB}_4$  the possible sites for field-dependent spin flips occur on one-dimensional (1D) ferromagnetic chains connected by  $J_2$  that are decoupled unless a fourth neighbor interaction  $J_4$  is included. For  $\text{TmB}_4$  a third neighbor interaction  $J_3$  complementary to  $J_1$  allows instead for a 2D network of possible spin flips. These differences can be connected to the corresponding plateau structures, which are shown in Fig. 1(d). Common to both systems are plateaus at  $M_S/2$ , while  $\text{TmB}_4$  shows an additional plateau with a higher denominator depending on the history and  $q$  may take noninteger values, as also shown in Fig. 1(d) [16]. As we discuss below, these differences in magnetism also have a significant impact on electronic transport.

## II. METHODS

Single crystals of  $\text{ErB}_4$  and  $\text{TmB}_4$  were grown using the floating-zone method. We reacted 99.99% pure  $\text{Er}_2\text{O}_3$  or 99.99% pure  $\text{Tm}_2\text{O}_3$  with 99% pure B in Ar flow to form polycrystalline tetraborides [23], from which single

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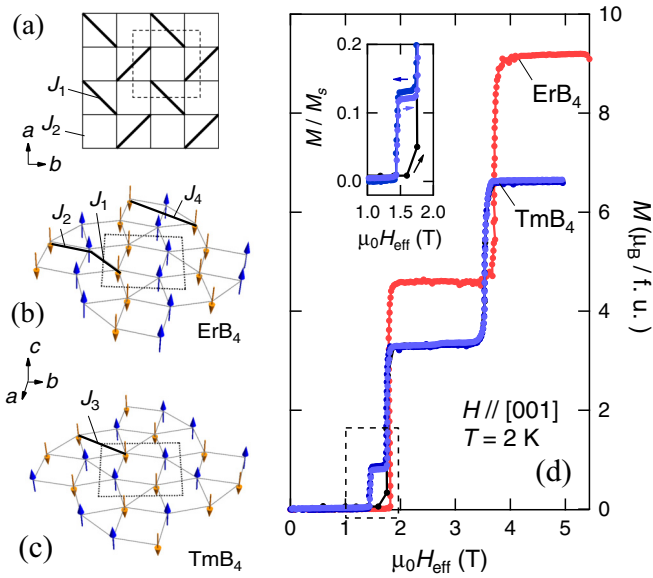


FIG. 1. Shastry-Sutherland lattice and magnetization plateaus in  $\text{ErB}_4$  and  $\text{TmB}_4$ . (a) SSL model with diagonal bond  $J_1$  and square bond  $J_2$ . (b, c) Spin configuration for antiferromagnetic ground states in  $\text{ErB}_4$  and  $\text{TmB}_4$ , respectively. Exchange couplings  $J_1$ ,  $J_2$ ,  $J_3$ , and  $J_4$  and the unit cell (dashed line) are shown. (d) Magnetization as a function of the field  $\mu_0 H_{\text{eff}}$  applied along the  $c$  axis for  $\text{ErB}_4$  and  $\text{TmB}_4$ . Inset: Magnified view near the  $(1/q)M_s$  phase in  $\text{TmB}_4$ . Light blue (dark blue) curves represent scans with an increasing (a decreasing) field between  $\pm 5$  T. The plateau values (defined at the midpoint of the plateau) for the two scan directions are  $0.121 M_s$  ( $q \simeq 8.3$ ) and  $0.132 M_s$  ( $q \simeq 7.6$ ), respectively.

crystals were obtained after further zone refining. Powder x-ray diffraction was done to confirm that the materials are of a single phase and single-crystal scattering was performed to orient crystals.

Measurements of  $M$  were performed using a commercial SQUID magnetometer with a field applied along the [001] tetragonal axis. The demagnetization factor  $N$  calculated from sample dimensions [24] and the measured  $M$  were used to obtain the effective field  $H_{\text{eff}} = H - NM$  and magnetic induction  $B = \mu_0(H_{\text{eff}} + M)$  for magnetization and transport measurements, respectively. Here  $\mu_0$  is the vacuum permeability. The contributions from  $R$  moments are significant, with  $\mu_0 M_s = 2.14$  and  $1.56$  T for  $\text{ErB}_4$  and  $\text{TmB}_4$ , respectively.

Electrical measurements were performed using a standard low-frequency (18.3-Hz) ac technique with a 2-mA excitation in a commercial cryostat. The magnetic field was applied along [001] as in magnetization measurements, with the current applied in the (001) Shastry-Sutherland plane. The dimensions of transport samples used here are  $0.71 \times 0.33(ab) \times 0.02(c)$  mm<sup>3</sup> ( $\text{ErB}_4$ ) and  $0.71 \times 0.28(ab) \times 0.03(c)$  mm<sup>3</sup> ( $\text{TmB}_4$ ).  $\rho_{xx}$  ( $\rho_{yy}$ ) is obtained from symmetrization (antisymmetrization) between time-reversed processes.

### III. RESULTS AND DISCUSSION

$\text{RB}_4$  are metals and the metallicity of  $\text{ErB}_4$  and  $\text{TmB}_4$  is similar. Starting with  $\text{ErB}_4$ , as shown in Fig. 2(a) the resistivity  $\rho$  as a function of  $T$  is metallic over the range  $T = 2$  to

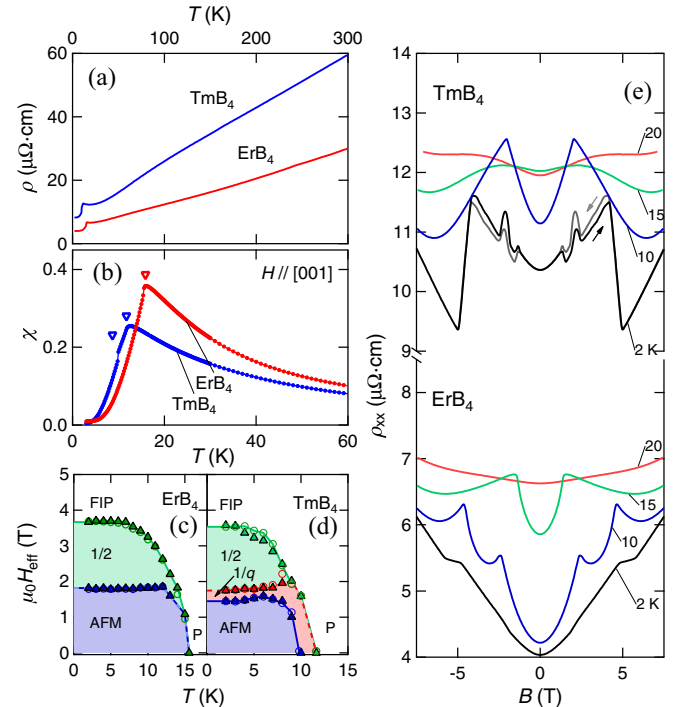


FIG. 2. Magnetic phase diagram of  $\text{ErB}_4$  and  $\text{TmB}_4$ . (a) Resistivity  $\rho$  as a function of temperature  $T$  for the SSL plane of  $\text{ErB}_4$  and  $\text{TmB}_4$  single crystals. (b) Volume magnetic susceptibility  $\chi$  measured along the  $c$  axis for  $\text{ErB}_4$  and  $\text{TmB}_4$ . Triangles denote transition temperatures. (c, d) Phase diagram in the  $H$ - $T$  plane for  $\text{ErB}_4$  and (trained)  $\text{TmB}_4$ , respectively. The boundaries determined from transport are represented by triangles; those determined from magnetization, by circles. (e) Magnetic-field dependence of the longitudinal resistivity  $\rho_{xx}(B)$  at selected  $T$  for  $\text{TmB}_4$  and  $\text{ErB}_4$ .

300 K. There is a kink in  $\rho(T)$  observed at low  $T$  which corresponds to the AFM ordering temperature  $T_N$  as observed in the temperature dependence of the magnetic susceptibility  $\chi(T)$  shown in Fig. 2(b). The response is distinct from the shoulderlike features observed for typical antiferromagnetic metals such as Cr and Dy [25], where the AFM ordering opens superzone gaps on the Fermi surface. Here this indicates an absence of Brillouin zone folding consistent with that the AFM magnetic unit cell is identical to the crystallographic unit cell. The field-temperature phase diagram is shown in Fig. 2(c); with increasing  $\mu_0 H_{\text{eff}}$   $\text{ErB}_4$  realizes a plateau state with  $M_S/2$  and eventually enters a field-induced paramagnetic (FIP) phase (see also Fig. 1(d)). As shown in Fig. 2(e), below  $T_N$  a series of magnetoresistance features appears at the phase boundaries in Fig. 2(c). In particular, prominent peaks are observed at the magnetic transitions at moderate  $T$  but are suppressed at the lowest  $T$ , 2 K.

The overall behavior of  $\text{TmB}_4$  is similar to that of  $\text{ErB}_4$ , but with an additional magnetic transition observed in  $\rho(T)$  and  $\chi(T)$  [Figs. 2(a) and 2(b), respectively], resulting in the phase diagram shown in Fig. 2(d). We denote the additional intermediate phase  $1/q$ , as the value of  $M$  in this region has been reported to be history dependent ( $q$  may take a value of 7, 9, or 11 [16]) and may not be precisely quantized [26]. Interestingly, this higher degree of complexity

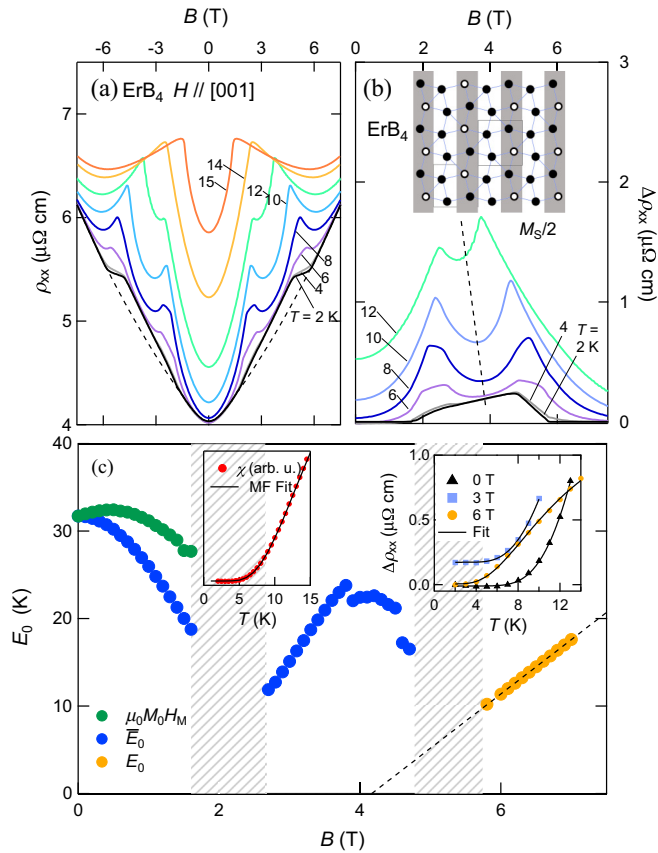


FIG. 3. Magnetic scattering in  $\text{ErB}_4$ . (a) Detailed magnetic-field dependence of the longitudinal resistivity  $\rho_{xx}(B)$  of  $\text{ErB}_4$  at  $T < T_N$ . (b) Magnetic contribution to resistivity  $\Delta\rho_{xx}$  (see text). Inset: Possible configuration of the half-plateau state, with AFM stripes marked in gray. Here the black (white) circles represent spins parallel (antiparallel) to  $H$ . Dashed square frames enclose two types of unit cells for the  $M_S/2$  phase. (c) Magnetic energy  $E_0$  as a function of  $B$ . The hatched area represents regions with phase coexistence and the dashed line is a linear fit to the FIP phase. Left inset: Mean-field fitting to the magnetic susceptibility of  $\text{ErB}_4$ . Right inset: Fitting of  $\Delta\rho_{xx}(T)$  at selected  $B$ .

is also reflected qualitatively in  $\rho_{xx}(B)$ . As shown in Fig. 2(e), a low-temperature hysteresis is observed in addition to sharp features corresponding to the magnetic transition.

### A. Magnetoresistance in $\text{ErB}_4$

Detailed study of  $\rho_{xx}(B)$  below  $T_N$  reveals connections to the magnetic phases and transitions in the Ising SSL system. We first focus on  $\text{ErB}_4$  with  $\rho_{xx}(B)$  shown in Fig. 3(a). The response can be understood as the sum of a conventional orbital magnetoresistance with additional scattering due to magnetic disorder and spin excitations as the plateau state evolves in field. To isolate the magnetic contribution, we calculate  $\Delta\rho_{xx}(B, T) \equiv \rho_{xx}(B, T) - \rho_{xx}^N(B, 2 \text{ K})$ , where we approximate the nonmagnetic contribution  $\rho_{xx}^N(B, 2 \text{ K})$  [dashed line in Fig. 3(a)] as a second-order polynomial fit to the AFM and FIP phases, where  $M$  is constant at low  $T$ . As shown in Fig. 3(b),  $\Delta\rho_{xx}$  exhibits a series of peaks at elevated  $T$  and a residual enhancement at intermediate  $B$ .

Unlike  $B$ -induced changes in resistivity for  $\rho_{xx}^N$  due to the Lorentz force, those in  $\Delta\rho_{xx}$  arise from interaction of carriers with the magnetic state and therefore reflect a change in carrier relaxation time  $\tau$ . The coexisting  $f$  moments and conduction electrons interact via a contact exchange interaction  $\mathcal{H}_{cf} = J_{c-f} \mathbf{s} \cdot \mathbf{S}$ , where  $\mathbf{s}$  is the conduction electron spin and  $\mathbf{S}$  is the total spin of localized magnetic moments [25,27]. It has been proposed that the  $M_S/2$  state is comprised of alternating AFM and ferromagnetic stripes [see the inset in Fig. 3(b)] where a large degeneracy of ordering of the AFM stripes exists [21]. Such an additional degree of freedom can be expected to increase irregularities in the spin structure and therefore also in the periodic potential seen by the charge carriers causing increased scattering. This is consistent with the steplike rise seen in both the raw  $\rho_{xx}(B)$  trace and the  $\Delta\rho_{xx}(B)$  peak in the  $M_S/2$  phase.

The pattern at elevated  $T$  in Fig. 3(b) suggests thermally enhanced magnetic scattering. For antiferromagnets in the strong Ising limit (where the exchange energy is less than the anisotropy energy), the lowest magnetic excitations are spin flips, as classical spin waves cost considerable anisotropy energies. In this context, the  $T$  excitation of the spin flips causes an increase in the spin-disorder resistivity (see Appendix) in the form [28,29]

$$\rho_m(T) \sim \text{sech}^2(E_0/k_B T), \quad (1)$$

where  $E_0$  represents the magnetic energy at each site and  $k_B$  is the Boltzmann constant. At  $B = 0$ ,  $E_0$  equals  $\mu_0 M_0 H_M$ , with  $M_0$  the rare-earth magnetic moment and  $H_M$  the effective molecular field at each site, and we get  $E_0 = 32 \text{ K}$  from fitting  $\Delta\rho_{xx}(T)$  with Eq. (1). This is comparable to the  $E_0 = 23 \text{ K}$  obtained from the mean-field fitting to the magnetic susceptibility of the Ising moments in  $\text{ErB}_4$  [30] [fit shown in Fig. 3(c), left inset],

$$\chi(T) = \frac{1 - m^2(T)}{T + E_0(1 - m^2(T))} + \chi_0, \quad (2)$$

where  $m(T)$  stands for the solution of sublattice magnetization at each  $T$  to  $m(T) = \tanh[E_0 m(T)/T]$ .  $\chi_0$  represents the residual susceptibility, which is rarely  $T$  dependent.

Equation (1) may be further modified to describe the effects of finite fields taking  $E_0 = \mu_0 M_0 |H_M \pm H_{\text{eff}}|$  and the sign depends on whether the magnetic moments align or antialign with the applied magnetic field. The green circles in Fig. 3(c) show the fit results of  $\mu_0 M_0 H_M$  taking half of all spins to be parallel and half antiparallel to  $H_{\text{eff}}$ , where  $\mu_0 M_0 H_M$  depends weakly on  $B$  within  $30 \pm 5 \text{ K}$ . Alternatively, we show the average  $\bar{E}_0$  obtained by assuming a single uniform  $E_0$  using blue circles, and the evolution of  $\bar{E}_0$  with  $B$  is shown in Fig. 3(c), with representative fits to Eq. (1) shown in the inset. As  $B$  is increased and the magnetic state is destabilized we see a drop in  $\bar{E}_0$  from the zero-field value, 32 K. At the magnetic transitions [regions corresponding to transitions in  $M(H_{\text{eff}})$  shown as hatched areas in Fig. 3(c)] a mixed magnetic phase is likely to exist and not be captured by the present model [31]. Upon entering the  $M_S/2$  phase we see a rise in  $\bar{E}_0$  to approximately 25 K, where the state is most stable, before it decreases again as the system approaches the transition to the FIP.

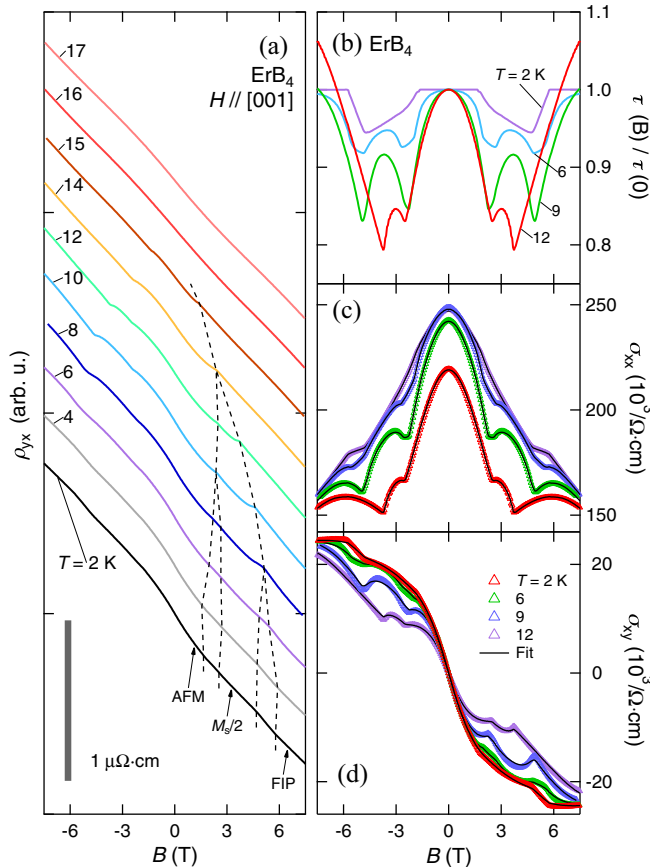


FIG. 4. Hall effect in ErB<sub>4</sub>. (a) Field dependence of the transverse resistivity  $\rho_{yx}(B)$  for ErB<sub>4</sub>. Dashed lines represent the singularities observed in  $\rho_{xx}$ . (b) Relative relaxation time  $\tau(B)/\tau_0$  as a function of  $B$ . (c) Longitudinal conductivity  $\sigma_{xx}$  and (d) transverse conductivity  $\sigma_{xy}$  fit with the modified two-band model using  $\tau(B)/\tau_0$  [Eqs. (3) and (4)]. The legend is the same for both panels.

In the FIP phase, all the magnetic moments are uniformly aligned with  $B$  and  $E_0 = \mu_0 M_0 (H_{\text{eff}} - H_M)$ , with the Zeeman energy gain associated with the applied field overwhelming the AFM interactions. Here we expect a linear  $B$  dependence of  $E_0$  as observed for fit results in the FIP phase [orange circles in Fig. 3(c)]. The slope yields  $M_0 = 9.24\mu_B$ , quantitatively consistent with the magnetic moment of  $\text{Er}^{3+}$  ( $M_s = 9.6\mu_B/\text{Er}$ ). The positive intercept on  $B$  implies that the underlying interaction of the system is antiferromagnetic, and the FIP phase is destabilized at magnetic fields below 4 T.

### B. Hall resistivity of ErB<sub>4</sub>

We next examine the transverse resistivity  $\rho_{yx}$ . As shown in Fig. 4(a), there is an overall electronlike response, with weak kinks appearing as a function of  $B$ . The magnetic phase boundaries from the phase diagram in Fig. 2(c) are shown as dashed lines and closely track the features in  $\rho_{yx}$ . These features can be understood by the magnetic modifications to  $\tau$  introduced above for  $\rho_{xx}$ . We employ a modified two-band model incorporating a field-dependent relaxation time  $\tau(B)$

TABLE I. Fitting parameters for the two-band model for ErB<sub>4</sub>. Sample A was measured between  $\pm 9$  T; sample B, between  $\pm 18$  T.

	$n_1$ (/cm <sup>3</sup> )	$\mu_1$ (cm <sup>2</sup> /V·s)	$n_2$ (/cm <sup>3</sup> )	$\mu_2$ (cm <sup>2</sup> /V·s)
<b>Sample A</b>				
$\sigma_{xx}$	$1.74 \times 10^{21}$	716.9	$8.15 \times 10^{19}$	3680.5
$\sigma_{xy}$	$3.17 \times 10^{20}$	1218	$1.816 \times 10^{19}$	4036
<b>Sample B</b>				
$\sigma_{xx}$	$1.26 \times 10^{21}$	484.3	$2.04 \times 10^{20}$	2064
$\sigma_{xy}$	$2.2 \times 10^{20}$	1186	$1.83 \times 10^{19}$	4441.7

for the longitudinal conductivity  $\sigma_{xx}$ ,

$$\sigma_{xx} = \sum_i \sigma_{xx}^i = \sum_i \frac{n_i e \mu_i (\tau(B)/\tau_0)}{1 + (\mu_i B)^2 (\tau(B)/\tau_0)^2}, \quad (3)$$

where  $\sigma_{xx}^i$ ,  $n_i$ , and  $\mu_i$  are the conductivity, carrier density, and mobility of each band, and  $\tau_0$  is the zero-field relaxation time at a given  $T$ . The total transverse conductivity  $\sigma_{xy}$  is written as

$$\sigma_{xy} = \sum_i \sigma_{xx}^i \cdot (\mu_i B) \cdot (\tau(B)/\tau_0). \quad (4)$$

The ratio  $\tau(B)/\tau_0$  shown in Fig. 4(b) is obtained from  $\Delta\rho_{xx}$ , viz,  $\tau(B)/\tau_0 = \rho_{xx}(0, T) / [\Delta\rho_{xx}(B, T) + \rho_{xx}(B, 2 \text{ K})]$ .

As shown in Figs. 4(c) and 4(d), Eqs. (3) and (4) provide satisfactory fits for  $\sigma_{xx}$  and  $\sigma_{xy}$ , respectively. The best fits for  $\sigma_{xx}$  and  $\sigma_{xy}$  at  $T = 2$  K are listed in Table I (also for a second sample, B). The set of parameters is similar for both fits, though there is a factor of 4–5 difference in carrier densities that optimize the longitudinal and transverse fits. We hypothesize that the lack of convergence is related to the Fermi surface's being composed of more than two bands [9]. However, higher order fitting is not a satisfactory proof of this, given the large number of parameters it introduces.

More generally, we suggest that this demonstrates that the features in  $\rho_{yx}$  may be captured by a field-induced scattering rate without showing clear signatures of the anomalous Hall effect conventionally observed in ferromagnets as a Hall effect proportional to  $M$  [32]. We point out that the magnitude of anomalous Hall conductivity  $\sigma_{xy}^A$  expected for the current system from the scaling relation between  $\sigma_{xy}^A$  and  $\sigma_{xx}$  is of the order  $10^3 / \Omega \cdot \text{cm}$  [33], which is difficult to decompose unambiguously from the background Hall conductivities with prominent features upon magnetic phase transitions [see black fit curves in Fig. 4(d)]. We suggest that systems with reduced background  $\sigma_{xy}$  from the normal Hall conductivity  $\sigma_{xy}^N$  may provide a clearer view of the extrinsic/intrinsic anomalous Hall contributions in magnetization plateau systems. As  $\sigma_{xy}^N \sim \tau$ , this may be achieved by doping the boron sites in  $RB_4$  with nonmagnetic elements to suppress  $\tau$  while minimizing the influence on the magnetic subsystem. Low carrier compounds are also favorable as they possess a smaller  $\sigma_{xy}^N$  background, though care must be taken as small carrier systems may exist under a different physical regime in the universal scaling [33].

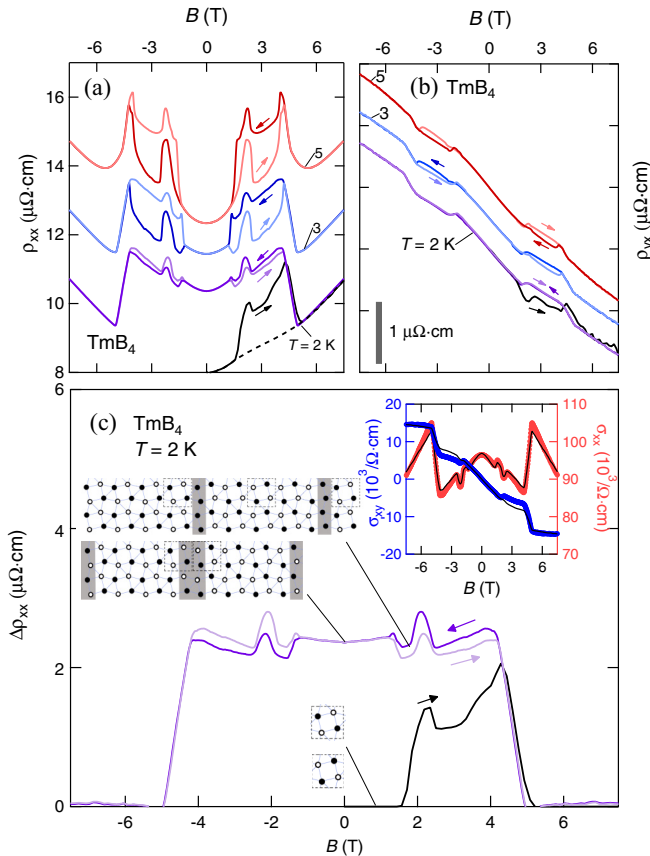


FIG. 5. Magnetotransport in  $\text{TmB}_4$ . (a) Longitudinal resistivity  $\rho_{xx}$  for  $\text{TmB}_4$  at low  $T$ . The  $T = 3$  K and  $T = 5$  K curves are offset for clarity. (b) Transverse resistivity of  $\text{TmB}_4$  at low  $T$ . (c) Magnetic contribution to resistivity  $\Delta\rho_{xx}$  and possible magnetic configurations [16,26]. Inset:  $\sigma_{xx}$  and  $\sigma_{xy}$  fit with Eqs. (3) and (4). For clarity, only scans from negative to positive  $B$  are shown.

### C. Transport in $\text{TmB}_4$

Turning to the detailed magnetotransport of  $\text{TmB}_4$ , the low- $T$  behavior of  $\rho_{xx}$  and  $\rho_{yx}$  is shown in Figs. 5(a) and 5(b), respectively. Unlike the case of  $\text{ErB}_4$ , we observe hysteresis in both transport channels (also recently reported in another study [34]). Here hysteresis refers to the difference between time-reversed full-field sweeps. As shown in Fig. 1(d) hysteresis is observed in  $M(H)$  in the vicinity of the  $(1/q)M_s$  phase; in transport hysteresis appears across a  $B$  range corresponding approximately to both the  $(1/q)M_s$  and the  $M_s/2$  phases. Additionally, for  $\rho_{xx}$  we observe a difference between the zero-field cooled (virgin) state and the trained state (that seen after once reaching the FIP phase).

To probe the origin of these effects, we construct  $\Delta\rho_{xx}$  in a manner analogous to that for  $\text{ErB}_4$ . In this case the normal component  $\rho_{xx}^N$ , which connects the AFM and FIP states, appears to belong to the virgin state, as shown by the dashed line in Fig. 5(a). Subtraction of this component yields  $\Delta\rho_{xx}$  as shown in Fig. 5(c). The presence of additional scattering is evident in the trained phase. We note that this is contrary to the case of conventional domain wall scattering in ferromagnets, in which the virgin state typically has a higher resistivity [24].

TABLE II. Fitting parameters for the two-band model for  $\text{TmB}_4$  (scan with increasing  $B$ ).

	$n_1$ (/cm <sup>3</sup> )	$\mu_1$ (cm <sup>2</sup> /V·s)	$n_2$ (/cm <sup>3</sup> )	$\mu_2$ (cm <sup>2</sup> /V·s)
$\sigma_{xx}$	$1.12 \times 10^{21}$	624	$3.95 \times 10^{18}$	10467
$\sigma_{xy}$	$6.8 \times 10^{19}$	1024	$8.12 \times 10^{19}$	1063

Considerations of the detailed real-space magnetic textures resulting from the 2D spin flip network in this system offer insight into this unusual behavior and, more broadly, the appearance of the  $(1/q)M_s$  phase [26]. The spin configuration for the zero-field cooled AFM state is known to have a magnetic unit cell identical to that of the crystallographic unit cell, as shown in Fig. 5(c) [16]. Starting from this simple AFM phase, with increasing  $B$  the  $M_s/2$  phase and then the FIP phase are stabilized. The subsequent decrease in  $B$  to 0 realizes a cascade of phases with  $M = M_s/2$ ,  $(1/q)M_s$ , and 0. However, the latter states are known to have larger real-space magnetic structures, which are evidently nearly degenerate in energy and accessible along this thermodynamic path [16,26]. One example of the expected long-period structure at  $M = 0$  is shown in Fig. 5(c), with AFM domains in an antiphase periodic structure. It has been suggested that the alignment/shift of those AFM domains every  $4/5$  unit cells leads to the  $(1/q)M_s$  phase in  $\text{TmB}_4$  [26]. This characteristic of training and complexity is a hallmark of strong magnetic frustration in  $\text{TmB}_4$ ; the resulting increase in  $\rho_{xx}$  can then be viewed as being due to domain wall scattering or the opening of superzone gaps in the Fermi surface if such structures are macroscopically ordered. In contrast, time-reversal antisymmetric quantities  $M$  and  $\rho_{yx}$  do not show training.

Similarly to the case of  $\text{ErB}_4$ , the patterns observed in both  $\rho_{xx}$  and  $\rho_{yx}$  for  $\text{TmB}_4$  can largely be explained by the magnetic structure-sensitive changes in  $\tau$  and spin disorder in the plateau phases. The fitting of  $\sigma_{xx}$  and  $\sigma_{xy}$  using Eqs. (3) and (4) is shown in the inset in Fig. 5(c) (parameters are listed in Table II). Fitting of the transport reproduces the experimental curves apart from in the  $M_s/2$  phase. As deviations in the Hall response in magnetic systems are often due to the anomalous Hall effect, we suggest that this may be due to a skew scattering contribution from the ferromagnetically aligned domain walls [16,26]. In terms of modeling as employed in  $\text{ErB}_4$ , analysis of  $\rho_{xx}(T)$  in the FIP phase yields a magnetic moment of  $6.84\mu_B$  ( $M_s = 6.66\mu_B/\text{Tm}$ ), molecular field of 1.74 T, and corresponding exchange energy of  $-0.69$  meV. Here again transport offers a quantitative measure of the underlying energy scales for the SSL.

## IV. CONCLUSION

The present study demonstrates that transport is a sensitive probe of magnetic disorder and excitations in model metallic frustrated systems. In particular, the magnetotransport processes are found to be sensitive to static and dynamic magnetic disorder across plateau transitions and allow for quantitative characterization of the underlying magnetic order and its excitations. These results are consistent with the strong Ising anisotropy expected for  $R = \text{Er}$  and  $\text{Tm}$ . Generally,

the enhancement in resistivity observed at the intermediate plateau phases favors a partially disordered state [21] or an additional number of domain walls [16,26] rather than uniform structures, examples of which have been proposed previously [20]. The results provide a framework in which to study the more complex  $RB_4$  magnetization plateau series such as  $TbB_4$  [17] and  $HoB_4$  [18] with non-Ising-type anisotropies. More broadly, our study offers a new approach to a central question in frustrated magnetic systems, i.e., the nature of their elementary excitations.  $Yb_2Pt_2Pb$  is a metal recently identified as an anomalous quasi-1D quantum magnet in which electronic transport may be a probe of spinon dynamics [35]. Further application to systems with novel excitations such as monopoles in spin ice [36], spinons in spin liquids [37], and quasi-1D quantum magnets [38] could offer new insights into these phenomena.

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### APPENDIX

#### 1. Resistivity analysis in $ErB_4$

We attribute the  $T$  evolution of  $\rho_{xx}$  to the inelastic scattering of conduction electrons by the magnetic subsystem. Due to the strong Ising anisotropy, the local moments can be adequately viewed as individual two-level systems split by molecular exchange fields. The level splitting is given by  $2E_0 = 2\mu_0 H_1 M_0$ .

The contribution to resistivity from inelastic scattering on localized quantum levels can be modeled as (following the

description of crystal-field scattering [28])

$$\rho \sim \frac{1}{\tau} \sim |J_{c-f}|^2 \sum_{i,i'} |\langle m'_s, i' | s \cdot S | m_s, i \rangle|^2 p_i f_{ii'}, \quad (A1)$$

where  $i$  and  $i'$  ( $m_s$  and  $m'_s$ ) denote the initial and final states of the magnetic moments (conduction electron spin), respectively. We define the occupation probability of the  $i$ th level as  $p_i$  and the Fermi factor as  $f_{ii'}$ , where

$$p_i = \frac{e^{-E_i/k_B T}}{\sum_j e^{-E_j/k_B T}}, \quad f_{ii'} = \frac{2}{1 + e^{(E_{i'} - E_i)/k_B T}}. \quad (A2)$$

Here  $E_i$  and  $E_{i'}$  are the energy of the localized moments before and after the scattering event, respectively.

Using  $\pm$  to denote the two local levels with energies  $\pm E_0$  we get

$$p_{\pm} = \frac{e^{\mp E_0/k_B T}}{e^{E_0/k_B T} + e^{-E_0/k_B T}}, \quad (A3)$$

and the Fermi factor raising (lowering) the energy of the magnetic system is

$$f_{\mp, \pm} = \frac{2}{1 + e^{\pm 2E_0/k_B T}} = \frac{2}{e^{\pm E_0/k_B T} (e^{E_0/k_B T} + e^{-E_0/k_B T})}. \quad (A4)$$

Substituting Eq. (A3) and Eq. (A4) into Eq. (A1) we obtain the  $T$  dependence of  $\rho$  as

$$\rho \sim \text{sech}^2(E_0/k_B T). \quad (A5)$$

#### 2. Parameters for two-band fitting

For  $ErB_4$  we fit Eqs. (3) and (4) to  $\sigma_{xx}$  and  $\sigma_{xy}$  of two samples, A and B, respectively. In Table I we show the fitting parameters for sample A at 2 K up to 9 T and for sample B at 1.6 K up to 18 T. In each case there exist two electron bands with a relatively high (low) density and low (high) mobility.

For  $TmB_4$ , to avoid complications of the observed hysteresis we fit the negative-to-positive field scan with the resulting parameters listed in Table II. Similarly, two electronlike bands contribute to the conductivity.

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