

Magnetic phase diagram of ErB_4 as explored by neutron scattering

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The tetragonal $4f$ -electron intermetallic ErB_4 is characterized by strong Ising anisotropy along the tetragonal c axis. The magnetic moments on the erbium sites can be mapped onto a Shastry-Sutherland lattice, resulting in geometrical frustration. At zero magnetic field ErB_4 exhibits collinear columnar antiferromagnetic (CAFM) order below $T_N = 15.4$ K. In the presence of a magnetic field parallel to the c axis, ErB_4 exhibits a plateau at $1/2$ of the saturation magnetization M_S , which arises at a spin-flip transition at $H_1 = 1.9$ T. Fractional magnetization plateaus and other exotic spin phases are a well-established characteristic feature of frustrated spin systems. Monte Carlo simulations propose that ErB_4 is an ideal candidate to feature a spin supersolid phase in close vicinity of H_1 between the CAFM and $M/M_S = 1/2$ plateau (HP) phase. Here, we combine single-crystal neutron diffraction and inelastic neutron scattering to study the magnetic phase diagram and the crystal electric-field (CEF) ground state of ErB_4 . Our measurements as a function of magnetic field find no signature of the spin supersolid phase but allow us to determine the magnetic structure of the HP phase to be of the up-up-up-down (uud) type consistent with an Ising material. The magnetic moment $\mu_{\text{CEF}} = 8.96 \mu_B$ expected from the CEF configuration determined by our inelastic neutron-scattering measurements is also consistent with the ordered moment observed in neutron diffraction, showing that the moments are fully ordered and close to the Er^{3+} free ion moment ($9.6 \mu_B$).

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I. INTRODUCTION

Magnetic frustration generally arises when the sum of all constraints determined by the underlying microscopic magnetic short-range interactions cannot be simultaneously satisfied. This typically results in the suppression of long-range magnetic order and a large degeneracy of the ground state [1–3]. In insulating quantum magnets that exhibit well-localized magnetic moments, magnetic frustration is an extensively studied phenomenon resulting in rich and exotic physics ranging from spin ice [4,5] to spin liquids [6], and experimental realizations of Kitaev magnets, which have the potential to host Majorana fermions [7]. In turn, insulating frustrated magnets are relevant both for our fundamental understanding of solids as well as for applications in quantum computing and quantum information systems [8].

In stark contrast, much less attention has been paid to magnetic frustration in metallic quantum materials. This is because the effects of magnetic frustration in metallic

systems are substantially weaker due to the long-range nature of itinerant magnetic exchange interactions [3,9]. Nevertheless, it has been recently demonstrated that frustration is also a relevant ingredient for understanding the magnetism in strongly correlated electron materials. Examples are heavy fermion materials [10–13], where metallic quantum spin liquid states have been proposed [14,15], skyrmion lattice materials [16–18], cubic geometrically frustrated rare-earth intermetallics [19,20], as well as van der Waals metals [21]. A common theme in all of these metals is the interplay between frustration and magnetic anisotropy.

Here we study the material ErB_4 , which belongs to the family of metallic rare-earth tetraborides RB_4 (R = rare earth). The tetraborides crystallize in the tetragonal space-group $P4/mbm$, as illustrated in Fig. 1(a). The magnetic moments on the rare-earth sites can be mapped onto a Shastry-Sutherland lattice (SSL) [22] [see Fig. 1(b)]. The SSL is a square lattice with antiferromagnetic nearest-neighbor interactions in every square [J_1 ; cf. Fig. 2(a)] and antiferromagnetic next-nearest neighbor interactions in every second square [J_2 ; cf. Fig. 2(a)], resulting in geometrical frustration [23]. An iconic realization of the SSL in an insulator is $\text{SrCu}_2(\text{BO}_3)_2$ [24], which at low temperature exhibits a sequence of magnetization plateaus at fractional values of the saturation magnetization M_S , and a plethora of exotic properties and excitations [25,26]. Interestingly, despite being metals, various members of the rare-earth tetraborides similarly exhibit magnetization plateaus at fractional values [27–34], demonstrating that frustration is an

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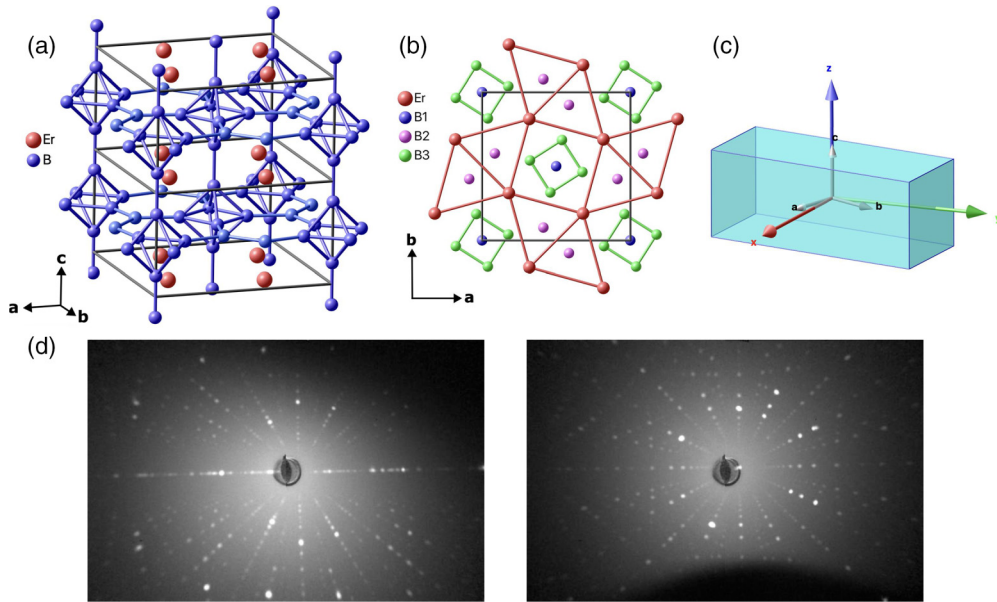


FIG. 1. (a) The tetragonal crystal structure of ErB_4 (space group $P4/mbm$) is shown. (b) Projection of the crystal structure onto the tetragonal basal plane. The red tubes denote the bonds between the Er atoms, which sit on a Shastry-Sutherland lattice (SSL). The black lines denote the boundaries of the conventional unit cell. (c) The model of the sample shape, which was used to calculate the scattering path lengths for absorption and extinction correction, is illustrated in the experimental scattering geometry. Here, $\mathbf{a}$, $\mathbf{b}$, and $\mathbf{c}$ define the crystallographic axes, whereas $\mathbf{x}$, $\mathbf{y}$, and $\mathbf{z}$ represent the laboratory coordinate system. (d) X-ray Laue backscattering photographs taken along the (left) ab plane and (right) c direction of an aligned sample.

important tuning parameter. Here, the magnetic structure and the number of successive magnetic transitions as a function of magnetic field resulting in the plateau phases changes depending on the rare-earth ion. HoB_4 [30] and TmB_4 [33,34] feature complex magnetic order with plateaus at different fractions of M_S . Even rare-earth tetraborides with seemingly simple magnetic structures can show plateau phases. TbB_4 [31,32] orders in an antiferromagnetic structure, with magnetic moments within the ab plane, and shows multiple plateaus at $M/M_S = 2/9, 1/3, 4/9, 1/2, \dots$, under applied magnetic field along the c axis.

ErB_4 is well suited to investigate the interplay of magnetic frustration and anisotropy due to the Ising-like character of its magnetism [28,29,35]. In fact, several theoretical studies have treated the SSL in the strong Ising limit and have identified ErB_4 as an ideal candidate to experimentally study this problem [36–40]. ErB_4 orders in a collinear columnar antiferromagnetic (CAFM) ground state below $T_N = 15.4$ K, where the Er moments point along the crystallographic c axis [cf. Fig. 2(a)] [29,35]. Neutron-diffraction studies [28,35] and magnetization [29] measurements for magnetic fields applied along the tetragonal c axis further revealed a metamagnetic transition to a ferrimagnetic phase with $M/M_S = 1/2$ at $H_1 = 1.9$ T [see Figs. 2(b) and 2(c)]. However, the full magnetic structure of the $1/2M_S$ -plateau phase (HP) above H_1 remains unknown.

Based on extended Monte Carlo simulations, it has further been proposed that ErB_4 may host exotic spin phases in magnetic field [40]. Notably, a supersolid phase is expected to exist in a narrow region between the CAFM and HP phase. Similarly, one expects a spin superfluid in the vicinity of the phase transition between the HP phase and

the field-polarized (FP) state. However, stabilizing the spin superliquid and -solid phases requires the existence of additional higher-order exchange interactions J_3 and J_4 , which are illustrated in Fig. 2(a). Here, J_3 is ferromagnetic and J_4 is much weaker and antiferromagnetic. Because for ErB_4 the local magnetic moments arising from $4f$ electrons on the erbium sites are embedded into a metallic host, long-range Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions mediated by the surrounding conduction electrons may indeed lead to finite exchange integrals J_3 and J_4 . The authors of Ref. [40] further argued based on their Monte Carlo simulations that the sequence of phases observed in ErB_4 may only be stabilized in case J_3 and J_4 are finite.

In this paper, we present single-crystal neutron-diffraction studies of the magnetic phase diagram of ErB_4 in order to investigate whether ErB_4 shows signatures in the neutron-scattering data that agree with the calculated magnetic structure factor for the supersolid phase [40]. In addition, we present an inelastic neutron-scattering study of the crystal electrical field ground state.

II. EXPERIMENTAL METHODS

The growth conditions described in Ref. [41] were employed for the growth of the single-crystal ErB_4 sample via the traveling solvent floating zone (TSFZ) method, in an optical image furnace with a two-mirror vertical configuration and a 5-kW xenon arc lamp (model HKZ, SciDre). The initial polycrystalline compound was prepared by means of the arc-melting method utilizing Edmund Bühler apparatus (model MAM-1). The starting materials used in the synthesis process were 3N erbium rods (Goodfellow) and 99.5%-enriched

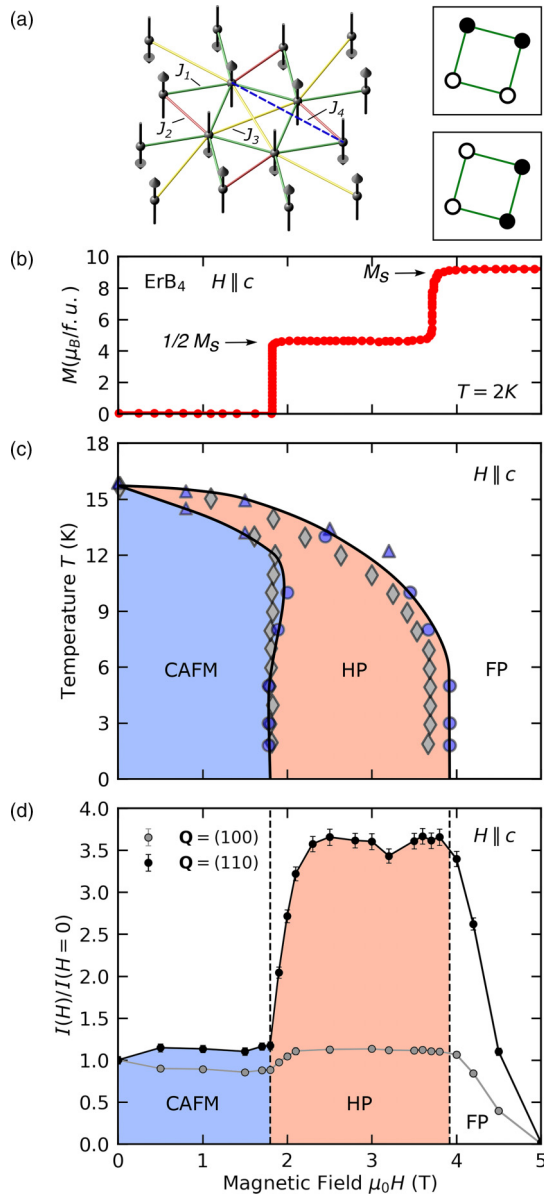


FIG. 2. (a) The magnetic structure of ErB_4 at zero magnetic field is illustrated. The diagonal bond J_1 (green) and the square bond J_2 (red) according to the Shastry-Sutherland model are also indicated. For ErB_4 the additional exchange interactions to higher-order neighbors J_3 and J_4 are also relevant and are shown as yellow solid and blue dashed lines, respectively (see text for details). The top and bottom insets on the right show the two domains $Pb'am$ and $Pba'm$ of the magnetic symmetry group $Pbam$, where up moments are depicted by black-filled circles and down moments depicted by white-filled circles. The magnetization as a function of magnetic field applied along the c axis, measured at $T = 2$ K (data adapted from Ref. [29]) and the corresponding temperature T vs magnetic field H phase diagram derived from transport measurements (gray diamond symbols, from Ref. [29]) are shown in (b) and (c), respectively. The blue triangles and circles denote phase transitions as determined from our neutron diffraction data using the magnetic field and temperature sweeps shown in Figs. 5 and 6, respectively. (d) Magnetic field dependence of the relative intensities for different Bragg peak positions measured at $T = 1.8$ K. The plotted intensity of each reflection was normalized with regard to the intensity observed at zero magnetic field.

isotopic ^{11}B pieces (Eurisotop). The $^{11}\text{ErB}_4$ crystal growth process was conducted at a pressure of 5 bar of argon with a flow rate of 1 l/min (5N, Messer) with a growth rate of 15 mm/h and a shutter opening of 95–100%. The feed and seed rods ($\phi \sim 8$ mm) were rotated clockwise at a rate of 15 rpm during the growth. The phase and crystallographic parameters of the TSFZ single-crystal samples were confirmed by single-crystal x-ray diffraction (SC-XRD), the details of which can be found in the Supplemental Material [42]. The orientation of single-crystal samples was performed using a Siemens Kristalloflex 760 Laue x-ray diffractometer equipped with a Photonic-Science Laue camera. Example x-ray Laue backscattering photographs of aligned samples are shown in Fig. 1(d), confirming the high crystalline quality.

Additional ^{11}B -enriched high-quality single crystals were grown using the flux method with Al flux [45]. Erbium pieces (Ames Laboratory; 99.99%), boron powder (99.99%), and aluminum shots (99.999%) in a 1:4:250 ratio were placed in a 50-mL alumina crucible and loaded in a horizontal tube furnace with argon flow (99.999%). The reagents were heated to 1550 °C, held at 1550 °C for 12 h, and slow cooled at 2.5 °C/h to 1050 °C. The furnace was then turned off, and chemical etching with a NaOH solution was used to remove the Al flux at room temperature. Two of these crystals with a total mass of $m = 0.105$ g were stacked in a mosaic for inelastic neutron-scattering experiments with a combined mosaic spread of less than 0.5°.

The neutron-diffraction experiment was carried out on the thermal single-crystal diffractometer ZEBRA at Paul Scherrer Institute (PSI). The neutron wavelength $\lambda = 1.178$ Å as provided by the (311) reflection of a Ge monochromator was used. The crystal was aligned with the crystallographic c axis vertically. A magnetic field up to 6 T along the c axis was provided by a vertical field cryomagnet that allows to perform diffraction experiments down to 1.8 K. The nuclear- and magnetic structure refinements were carried out using the program MAG2POL [46]. Note that even though our sample was enriched with ^{11}B (absorption cross section 0.0055 barn), the absorption of ^{10}B is so large [3835.9 barn] that even a small fraction results in strong absorption effects and poses a challenge with regard to high-quality structural refinements. In order to overcome this challenge, we determined the transmission factor and applied an absorption correction by using a convex-hull sample shape model in MAG2POL, which enables the calculation of beam path lengths inside the sample, before and after the scattering process. The model used for the sample shape is illustrated in Fig. 1(c). By applying the *on-the-fly* absorption option [46] of MAG2POL, the linear absorption coefficient, which highly depends on the ^{11}B concentration, is recalculated in every iteration of the nuclear refinement using the ^{11}B concentration as a refinable parameter.

We further carried out inelastic neutron-scattering measurements on the multiplexing spectrometer CAMEA [47,48] at PSI to determine the crystal electrical field (CEF) excitation of ErB_4 . Various incident energies between $E_i = 5$ and 13 meV were used to investigate the excitation spectrum over an energy transfer of 0–10 meV and a Q transfer $\sim 0.5 - 2$ Å $^{-1}$ at 1.6 K. Preliminary inelastic neutron-scattering measurements were additionally performed on the time-of-flight

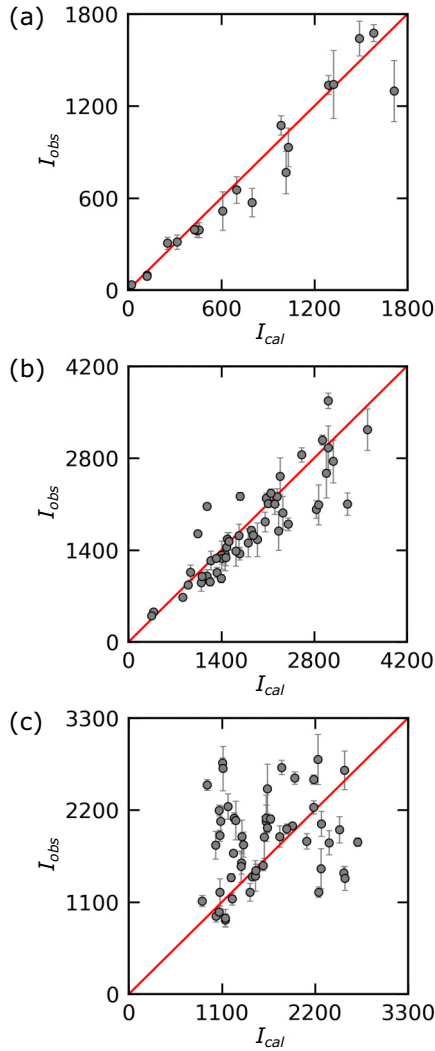


FIG. 3. Calculated integrated intensities (I_{cal}) vs measured integrated intensities (I_{obs}) for the best fits obtained in our refinements of all measured Bragg peaks. The red line indicates the ideal condition of $I_{\text{obs}} = I_{\text{cal}}$. Panels (a), (b), and (c) show the refinements for the nuclear structure measured at $T = 20$ K and $H = 0$, the zero-field CAFM magnetic structure at $T = 1.8$ K and $H = 0$, and the HP phase at $T = 1.8$ K and $H = 3$ T, respectively.

spectrometer FOCUS [49] located at PSI. The neutron data were analyzed using the PYTHON package PYCRYSTALFIELD [50].

III. EXPERIMENTAL RESULTS

A. Refinement of crystal structure

To confirm the crystal structure of our samples, the nuclear structure factor was measured on 112 structural Bragg reflections indexed in the space group $P4/mbm$ [No. 127; see Figure 1(a)] in the paramagnetic state at 20 K. The structural refinement results in an agreement factor of $R_F = 6.88$ for the previously reported tetragonal crystal structure of ErB_4 . The calculated versus the observed integrated intensities for the best fit are shown in Fig. 3(a). The refined parameters were the atomic positions, two of the diagonal elements (x_{11}, x_{22})

TABLE I. Atomic parameters for the tetragonal crystal structure (space group $P4/mbm$) of ErB_4 as obtained via refinement of our neutron-diffraction data taken at $T = 20$ K. The table lists all atoms, their Wyckoff positions, and their coordinates in a conventional tetragonal unit cell.

Atom	Wyckoff	x	y	z
Er	4g	0.3193(7)	0.8193(7)	0.0000
B(1)	4e	0.0000	0.0000	0.2045(3)
B(2)	4g	0.0849(1)	0.5849(1)	0.5000
B(3)	8j	0.1767(9)	0.0400(9)	0.5000

of the extinction correction tensor and an overall scale factor. The third diagonal element x_{33} could not be refined due to the small number of accessible out-of-plane reflections (hkl), $l \neq 0$. Due to the relatively small number of recorded reflections combined with a neutron absorbing sample, refinement of the isotropic temperature factor was not possible and therefore fixed to zero. Finally, we also refined the site-occupation boron sites with ^{11}B and obtained 0.987(6), which is in good agreement with the purity of ^{11}B -enriched boron used for the growth. The original dataset was corrected for absorption and averaged in $P4/mbm$ symmetry using the refined ^{11}B occupancy. The refined atomic positions are tabulated in Table I together with the extinction parameters $x_{11} = 1.09(4)$ and $x_{22} = 0.86(3)$. Our results agree well with previous structural refinements of ErB_4 [51,52].

B. Crystal electric-field excitations

The magnetic moment on the erbium sites in ErB_4 is a result of their partially filled $4f$ shell. The electronic configuration for the Er^{3+} ions is $[\text{Xe}]4f^{11}$ and, following Hund's rules, the total angular momentum of the spin-orbit ground state is $J = 15/2$ with $L = 6$ and $S = 3/2$. In absence of the crystal electric field, the corresponding spin-orbit ground state is $2J + 1 = 16$ -fold degenerate. The neighboring B ions generate the CEF, which typically corresponds to an energy scale of about 100 meV. Note that for Er^{3+} the spin-orbit gap between the first-excited and the spin-orbit ground state is $\lambda J \approx 2.5$ eV [53]. In turn, for ErB_4 , we can limit ourselves to lower- J multiplets for our CEF analysis.

The form of the CEF Hamiltonian is defined by the position of the boron ligands surrounding the magnetic Er^{3+} ion. Using PYCRYSTALFIELD [50], we built a point-charge model containing 18 ligands and a mirror plane along the $[1\bar{1}0]$ direction. The mirror symmetry suppresses $-m$ terms such that the CEF Hamiltonian is given by

$$\mathcal{H}_{CF}^{\text{ErB}_4} = B_2^0 O_2^0 + B_2^2 O_2^2 + B_4^0 O_4^0 + B_4^2 O_4^2 + B_4^4 O_4^4 + B_6^0 O_6^0 + B_6^2 O_6^2 + B_6^4 O_6^4 + B_6^6 O_6^6, \quad (1)$$

where the O_n^m are Stevenson operators, which are written in terms of the J^+ , J^- , and J_z operators [54]. The calculated CEF parameters of the underlying point-charge model reveal that Eq. (1) is predominantly determined by the first two terms, allowing us to approximate the CEF Hamiltonian for ErB_4 by

$$\mathcal{H}_{CF}^{\text{ErB}_4} \approx B_2^0 O_2^0 + B_2^2 O_2^2. \quad (2)$$

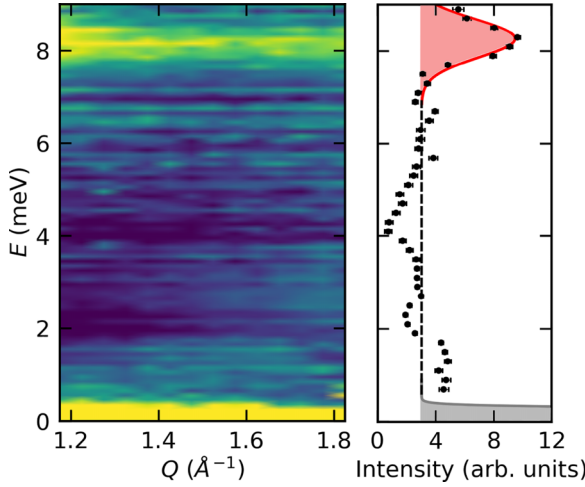


FIG. 4. Inelastic neutron-scattering measurement up to an energy loss of 9.2 meV at 1.6 K. The energy vs intensity plot on the right is the Q integrated, where the red line shows the fit of the CEF level using the CEF Hamiltonian in Eq. (2). The black dashed line illustrates the background level and the gray line shows the peak shape of the elastic line.

Figure 4 shows the results of our inelastic neutron measurements on CAMEA. In the left panel of Fig. 4, we show the neutron energy-loss channel up to 9.2 meV as a function of momentum transfer. Only a single dispersionless excitation is observed at 8.25 meV in this energy window. As CEF excitations are single-ion properties, they typically lack dispersion and exhibit an intensity distribution, which decreases as a function of momentum transfer following the magnetic form factor of Er^{3+} .

Because these measurements were carried out at a temperature $T = 1.6$ K, the first-excited state cannot be significantly thermally populated. Therefore, this excitation stems from the CEF ground state. Notably, of the total of seven transitions between the eight Kramers doublets belonging to the lowest-energy manifold for Er^{3+} with $J = \frac{15}{2}$, this is the first transition between the ground state and the next-highest doublet.

To determine the CEF Hamiltonian, we have fitted the momentum-transfer integrated neutron spectrum shown in the

right panel of Fig. 4 to Eq. (2). The resulting parameters are $B_2^0 O_2^0 = 0.140\,42$ meV and $B_2^2 O_2^2 = -0.262\,40$ meV, respectively. Table II shows the calculated eigenvalues and eigenvectors based on the fitted CEF Hamiltonian up to the first-excited doublet. The expected magnetic moment calculated from the fitted CEF Hamiltonian is $\mu_{\text{CEF}} = 8.96(3) \mu_B$. This compares well with the magnetic saturation moment identified previously in magnetization measurements [29].

C. Magnetic phase diagram probed by neutron diffraction

Before performing symmetry analysis to determine the magnetic structures from our neutron-diffraction data, we first establish the magnetic phase diagram based on neutron-scattering data. In Fig. 2(d), we show the integrated neutron intensity for the (100) and (110) Bragg reflections. Here, the plotted intensity of each reflection was normalized with regard to the intensity observed at zero magnetic field. Due to symmetry, the (100) reflection represents purely antiferromagnetic contributions, whereas the (110) reflection corresponds to an additional uniform ferromagnetic contribution. The relative intensity of the (110) Bragg peak shows a clear plateau as a function of magnetic field, whereas the antiferromagnetic contribution on (100) remains almost unchanged. This shows that an additional uniform ferromagnetic contribution to the magnetization emerges when entering the HP phase, suggesting that the metamagnetic transition above H_1 can be characterized as a spin-flip transition.

In Figs. 5 and 6, we additionally show integrated intensities measured on the (010) and (020) reflections as a function of magnetic field $\mu_0 H$ and temperature T , which we used to establish the H - T phase diagram of ErB_4 . In Fig. 5, we illustrate the magnetic field dependence of the integrated intensity of the (010) pure magnetic Bragg peak for various temperatures T . When entering the HP phase the intensity displays a clear jump and stays nearly constant as a function of field analogous to the magnetization plateau. For increasing temperature, the integrated intensity decreases as a function of field and the jump in intensity reduces to a small kink in the data. The field values for the phase transitions between the CAFM and HP phase (H_1) and between the HP and the FP state (H_2), as determined from our neutron data, are denoted with black triangles in Fig. 5.

TABLE II. Eigenvalues and eigenvectors for crystal electrical field Hamiltonian in Eq. (2) for the Er^{3+} ions in ErB_4 fitted to the inelastic neutron data shown in Fig. 4. The values are shown up to the first-excited doublet of the Er^{3+} $J = \frac{15}{2}$ ground-state manifold that was accessible in our measurements. The top numbers in a row correspond to $|+J_z\rangle$ and the bottom numbers to $|-J_z\rangle$.

E (meV)	$ \frac{+15}{-2}\rangle$	$ \frac{+13}{-2}\rangle$	$ \frac{+11}{-2}\rangle$	$ \frac{+9}{-2}\rangle$	$ \frac{+7}{-2}\rangle$	$ \frac{+5}{-2}\rangle$	$ \frac{+3}{-2}\rangle$	$ \frac{+1}{-2}\rangle$
0.000	-0.0048 0.0	0.0 -0.021	-0.062 0.0	0.0 -0.1418	-0.2651 0.0	0.0 -0.4167	-0.5595 0.0	0.0 -0.647
0.000	-0.0048 0.0	0.0 -0.021	-0.062 0.0	0.0 -0.1418	-0.2651 0.0	0.0 -0.4167	-0.5595 0.0	0.0 -0.647
8.254	0.0 0.0182	-0.0709 0.0	0.0 0.1808	-0.3442 0.0	0.0 0.508	-0.5771 0.0	0.0 0.4686	-0.1814 0.0
8.254	-0.0182 0.0	0.0 0.0709	-0.1808 0.0	0.0 0.3442	-0.508 0.0	0.0 0.5771	-0.4686 0.0	0.0 0.1814

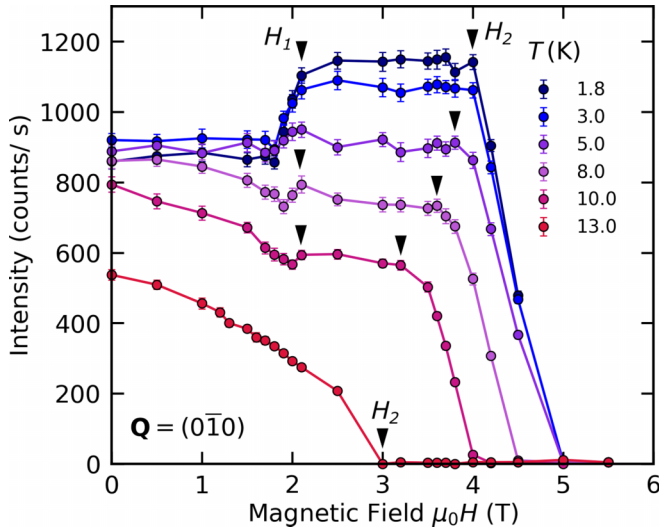


FIG. 5. Integrated intensity as a function of magnetic field for the purely magnetic Bragg signal at $(0\bar{1}0)$, recorded at different temperatures. The magnetic field was applied along the crystallographic c axis.

Figure 6(a) shows the temperature dependence of the magnetic contribution to the $(0\bar{2}0)$ Bragg peak for different magnetic fields. Here, the magnetic part of the $(0\bar{2}0)$ Bragg peak was obtained by subtracting the Bragg-peak signal well above the ordering temperature. To determine the critical temperature T_{N1} , the integrated intensity I was fitted to

$$I(T) = M^2(T) = \left(\frac{T_{N1} - T}{T_{N1}} \right)^{2\beta}, \quad (3)$$

where M is the staggered magnetization (order parameter) and β is the critical exponent of the order parameter. The best fits are represented by the solid lines in Fig. 6(a). The corresponding T_{N1} determined by these fits is denoted with a black triangle in Fig. 6(a). At zero field we obtain $\beta = 0.23(1)$, whereas at the highest fields we find $\beta = 0.20(2)$. However, for intermediate field values $\mu_0 H = 0.8$ and 1.5 T, the temperature dependence [see Figs. 6(b) and 6(c)] shows a weak shoulderlike feature and Eq. (3) is not suitable to describe the temperature dependence for the entire temperature range below T_{N1} . Comparing with the phase diagram previously established from bulk measurements (cf. Ref. [29]) shown in Fig. 2(c), it becomes apparent that for those fields, the system undergoes two phase transitions as a function of temperature. Notably, the shoulder is associated with the phase transition between the CAFM and the HP, which we label with T_{N2} .

In Figs. 6(b) and 6(c), we demonstrate that the temperature dependence of the integrated intensity I of the $(0\bar{2}0)$ reflection for 0.8 and 1.5 T can be well accounted for by fitting the region above and below T_{N2} with distinct critical exponents. Notably, we find $\beta = 0.14(1)$ describes the temperature dependence of the order parameter below T_{N2} best.

All phase boundaries that were determined from our neutron-diffraction data are also denoted in Fig. 2(c). The phase transitions determined from magnetic field (see Fig. 5) and temperature scans (see Fig. 6) are marked with blue triangles and circles, respectively. This shows that the phase

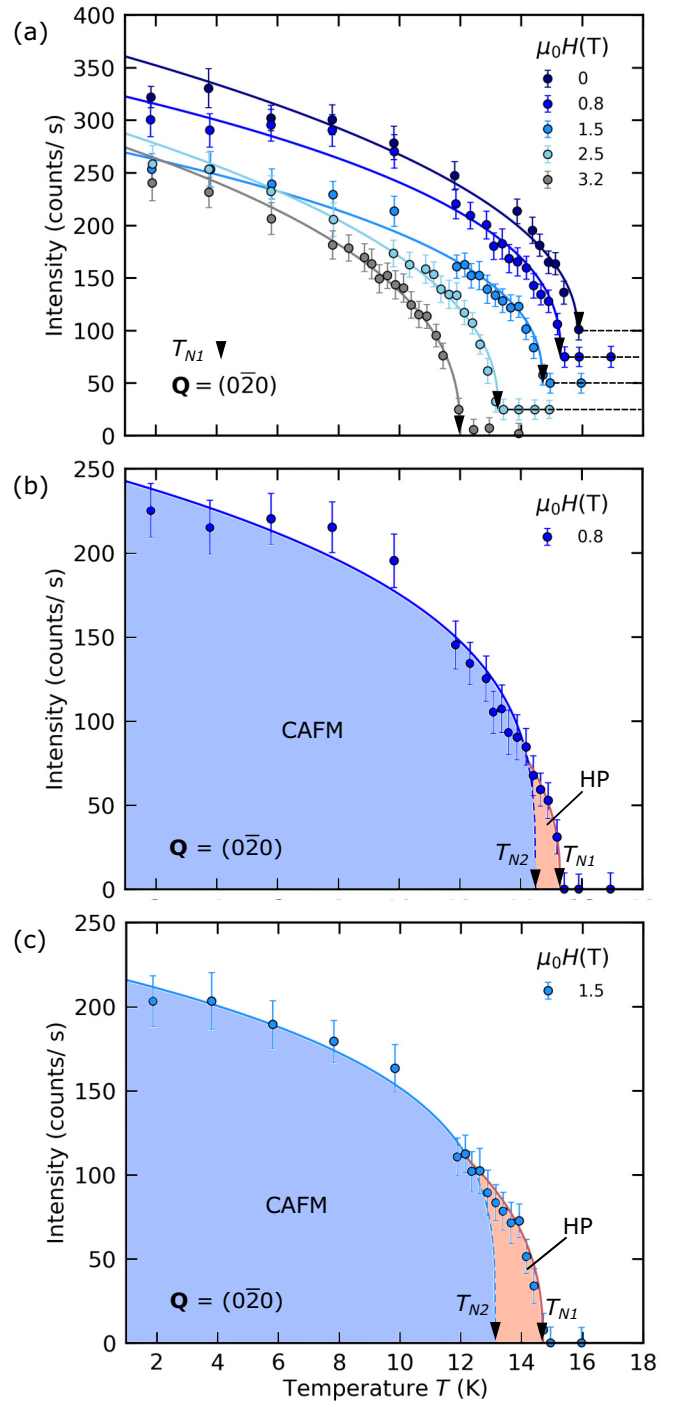


FIG. 6. (a) Integrated intensity for the $(0\bar{2}0)$ Bragg peak as a function of temperature measured at different magnetic fields. Here, we only show the magnetic contribution to the reflection, which was obtained by subtracting the intensity obtained at a temperature well above the critical temperature. For better readability the datasets were shifted by a constant offset of 25 with respect to each other, such that the dashed lines indicate zero intensity for each dataset. The solid lines represent a fit to Eq. (3). For intermediate field values $\mu_0 H = 0.8$ and 1.5 T, Eq. (3) is not suitable to describe the temperature dependence for the entire temperature range below T_{N1} (see text for details). As shown in panels (b) and (c), the situation is more complex for $\mu_0 H = 0.8$ and 1.5 T, respectively (see text for details).

TABLE III. Irreducible representation and basis vector composition for space group $P4/mbm$ with $\mathbf{k} = 0$ found using the representation analysis option of MAG2POL. The erbium atomic positions are defined as Er1 = (0.31875, 0.81875, 0), Er2 = (0.68125, 0.18125, 0), Er3 = (0.18125, 0.31875, 0), Er4 = (0.81875, 0.68125, 0).

IR	BV	Atom	BV components			IR	BV	Atom	BV components		
Γ_2 (AFM)	Ψ_1	Er1	0.707	0.707	0	Γ_9 (fm)	Ψ_1	Er1	1	0	0
		Er2	-0.707	-0.707	0			Er2	1	0	0
		Er3	-0.707	0.707	0			Er3	0	-i	0
		Er4	0.707	-0.707	0			Er4	0	-i	0
Γ_3 (FM)	Ψ_1	Er1	0	0	1	Γ_9	Ψ_2	Er1	0	1	0
		Er2	0	0	1			Er2	0	1	0
		Er3	0	0	1			Er3	i	0	0
		Er4	0	0	1			Er4	i	0	0
Γ_4 (AFM)	Ψ_1	Er1	0.707	-0.707	0	Γ_9	Ψ_3	Er1	0	-i	0
		Er2	-0.707	0.707	0			Er2	0	-i	0
		Er3	0.707	0.707	0			Er3	-1	0	0
		Er4	-0.707	-0.707	0			Er4	-1	0	0
Γ_5 (AFM)	Ψ_1	Er1	0	0	1	Γ_9	Ψ_4	Er1	-i	0	0
		Er2	0	0	1			Er2	-i	0	0
		Er3	0	0	-1			Er3	0	1	0
		Er4	0	0	-1			Er4	0	1	0
Γ_6 (AFM)	Ψ_1	Er1	0.707	-0.707	0	Γ_{10} (AFM)	Ψ_1	Er1	0	0	1
		Er2	-0.707	0.707	0			Er2	0	0	-1
		Er3	-0.707	-0.707	0			Er3	0	0	-i
		Er4	0.707	0.707	0			Er4	0	0	i
Γ_8 (AFM)	Ψ_1	Er1	0.707	0.707	0	Γ_{10}	Ψ_2	Er1	0	0	i
		Er2	-0.707	-0.707	0			Er2	0	0	-i
		Er3	0.707	-0.707	0			Er3	0	0	-1
		Er4	-0.707	0.707	0			Er4	0	0	1

diagram derived from neutron measurements agrees well with the phase diagram previously constructed from bulk transport measurements in Ref. [29].

We note that according to Ref. [40] the magnetic structure factor for the proposed spin supersolid phase should lead to additional magnetic intensities at peaks corresponding to magnetic propagation vectors of the type (100), (110), and (000). The phase is expected in a narrow magnetic field regime around H_1 . As shown in Figs. 2(d) and 5, we do not observe any extra intensity in this field range and observed intensities can be attributed either to the CAFM phase or the HP phase as we show below.

D. Magnetic structure at zero magnetic field

We now proceed to confirm the magnetic structure at zero magnetic field. The structure is characterized by the propagation vector $\mathbf{k} = (1, 0, 0)$ [28,35] that is equivalent to $\mathbf{k} = 0$. By applying group-theoretical symmetry analysis using MAG2POL and SARAh, the decomposition of the representation of the magnetic Er site occupying the 4g Wyckoff position of the $P4/mbm$ space group based on $\mathbf{k} = 0$ into irreducible representations (IRs) is given by

$$\Gamma = \Gamma_2^1 + \Gamma_3^1 + \Gamma_4^1 + \Gamma_5^1 + \Gamma_6^1 + \Gamma_8^1 + 2\Gamma_9^2 + \Gamma_{10}^1, \quad (4)$$

where the superscripts define the order of the IR. The calculated irreducible representations containing the basis vector components for the different erbium atom positions are presented in Table III. The IRs Γ_2 , Γ_4 , Γ_5 , Γ_6 , Γ_8 , and Γ_{10}

are associated with antiferromagnetic configurations and Γ_3 , Γ_9 with ferromagnetic configurations. Due to the vanishing uniform magnetization in the zero-magnetic field phase [cf. Fig. 2(a)], only IRs representing antiferromagnetic configurations are allowed. To perform the magnetic structure determination, 152 Bragg reflections were recorded at a temperature of 1.8 K. The reflections were integrated and averaged in $Pmmm$ symmetry to allow for lower-symmetry models. The magnetic form factor was described by the analytic approximation to the $\langle j_0 \rangle$ integrals for the f electrons of the Er^{3+} ions [55].

The results of the magnetic refinements are presented in Table IV, showing that the best refinement was obtained for the IR Γ_{10} , with an agreement factor of $R_F = 7.88$. Refining the magnetic components C_1 and C_2 associated with the normalized basis vectors Ψ_1^{10} , Ψ_2^{10} of Γ_{10} , with the constraint of the magnetic moments on each site having the same modulus, results in $C_1 = C_2 = 8.8(9) \mu_B$, with an equal population of the two magnetic domains, 0.50(2) ($Pb'am$) and 0.49(2)

TABLE IV. Agreement factors obtained by fitting the irreducible representations to the recorded structure factors at 1.8 K and zero magnetic field. The fits were performed using the software MAG2POL and the normalized basis vectors of the IRs in Table III.

IR	Γ_2	Γ_4	Γ_5	Γ_6	Γ_8	Γ_{10}
R_F	21.51	20.08	30.15	21.99	20.17	7.88

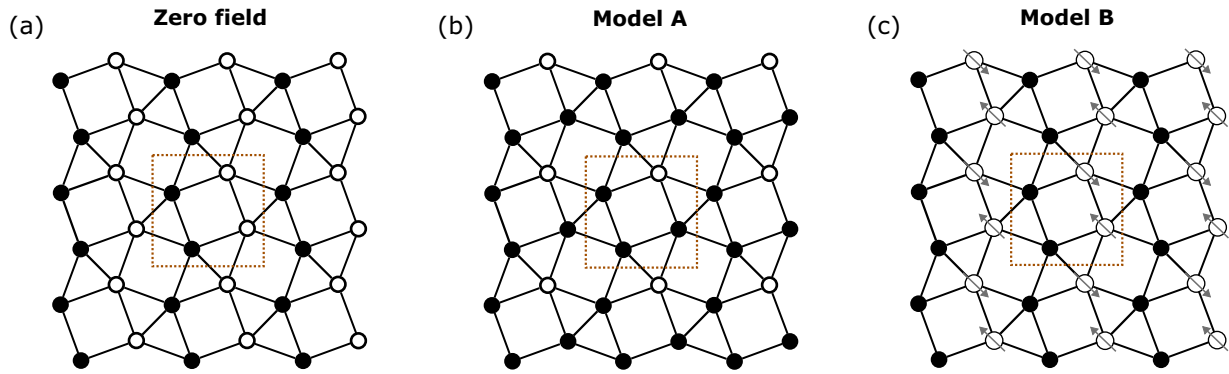


FIG. 7. (a) Collinear columnar antiferromagnetic (CAFM) structure at zero field. (b), (c) Spin-flip models used for the refinement of the magnetic structure in the half-plateau phase. Model A denotes the situation, where starting from the collinear antiferromagnetic ground state at zero field, one of the moments is flipped 180° . Model B describes the situation where both down moments are flipped by 90° , such that they are oriented within the basal plane. These moments are illustrated by gray circles with arrows denoting the moment direction within the plane.

($Pba'm$) [cf. Fig. 2(a)]. The ordered magnetic moment agrees well with the magnetic moment $\mu_{\text{CEF}} = 8.96(3) \mu_B$ expected from the determined crystal electrical field Hamiltonian. The calculated versus the observed integrated intensities for the best fit to IR Γ_{10} are shown in Fig. 3(b). The magnetic structure identified in our refinements is in agreement with the zero-field structure proposed in Ref. [35] and is illustrated in Figs. 2(a) and 7(a).

E. Magnetic structure of the HP phase

As we have demonstrated above, the metamagnetic transition at H_1 is characterized by the emergence of an additional uniform ferromagnetic contribution to the magnetization suggesting a spin-flip transition. Thus, to determine the magnetic structure in the HP phase we study two different spin-flip models that are allowed by the symmetry group $Pbam$, and are illustrated in Fig. 7.

Both models have in common that half of the spins in the magnetic unit cell are arranged in pairs, in an antiparallel fashion, such that they do not contribute to the uniform magnetization. In turn, each model results in a uniform magnetization that corresponds to $1/2$ of the saturation magnetization M_S so that they comply with the bulk magnetization measurements. Model A represents the Ising-type uuud model, where one of moments pointing downwards in the zero-field ground state flips by 180° (cf. Fig. 7). Model B represents the situation where both of the spins pointing downwards at $H = 0$ T become flipped by 90° at H_1 , such that they are coplanar within the basal plane.

We note that the refinement of the magnetic structure in the plateau phase is affected by additional challenges. Notably, a first-order phase transition driven by the application of magnetic field, as observed at H_1 (cf. the jump in the magnetic order shown in Fig. 2), typically results in an abrupt rearrangement of magnetic domains. Due to magnetostriction, this also influences the nuclear intensities, which impacts the correction of extinction. In turn, this requires refining the extinction parameters for the plateau phase separately. Thus, the magnetic structure refinements of the HP phase were per-

formed by treating the extinction parameters x_{11} and x_{22} as free parameters.

The separate refinement of x_{11} and x_{22} , however, poses the problem that a reliable refinement of the size of magnetic moment is difficult. In the case of the ErB_4 , this can be mitigated, because the ordered moment in the zero-field phase is known and corresponds to the magnetic moment $\mu_{\text{CEF}} = 8.96(3) \mu_B$ expected from the CEF configuration. Moreover, both previous bulk magnetization measurements as well as our neutron-diffraction results show that the transition in the HP phase is characterized by a spin-flip transition, which does not affect the magnitude of the magnetic moments. In turn, the refinement was carried out with a fixed magnetic moment μ_{CEF} and the scale parameter determined for the zero-field magnetic structure. We note that we also attempted an alternative strategy to refine the integrated intensities, where $x_{11} = x_{22}$ was enforced. However, this strategy resulted in unphysically large extinction parameters without improving the agreement factors, and was therefore disregarded. In addition, model B was refined by choosing the rotation angle φ in the basal plane as a refinement parameter under the constraint of a vanishing net magnetic moment within the basal plane. Canting of the moments out of plane did not lead to better refinements. Because our limited dataset is not appropriate to differentiate between models where canting angles differ just by a few degrees, we constrained the moments within the basal plane.

The spin-flip model A, where one of the magnetic moments flips, leads to the best agreement factor of $R_F = 14.77$ (see Table V). The refinement of model A is presented in Fig. 3(c). The large difference between the calculated and recorded intensities is a direct result of the small number of recorded Bragg peaks, the strong absorption and extinction in

TABLE V. Agreement factor and anisotropic extinction parameters of the refined spin-flip models.

Model	R_F	x_{11}	x_{22}
A	14.77	0.10(2)	0.06(1)
B	17.64	0.05(2)	0.06(2)

the material, and the resulting errors propagating through all refinements, but does not change the conclusion that model A provides the best fit of the HP phase in ErB₄.

IV. DISCUSSION

Our neutron-scattering study offers insight into the magnetic phase diagram of ErB₄. Most notably, our neutron-diffraction results in magnetic field show no signatures that may be related to the emergence of a spin supersolid phase between the CAFM and HP phases. This suggests that the exact combination of exchange interactions J_1 to J_4 that lead to the supersolid phase in the Monte Carlo simulations [40] does not reflect the magnetic interactions in ErB₄. Although, it has been argued in Ref. [40] that substantial finite ferromagnetic J_3 and a small antiferromagnetic J_4 are able to stabilize the CAFM and HP phases, we note that various sets of interactions may lead to this sequence of phases. For example, anisotropic interactions J_2 in combination with a small J_3 may also stabilize these phases [56]. However, we also note that our measurements were limited to $T = 1.8$ K and the supersolid phase may arise at lower temperatures. Previous transport and magnetization measurements [29] similarly do not detect any signatures of the supersolid phase. Further, the magnetic phase diagram established via our neutron-diffraction experiments agrees well with the previously established phase diagram [29].

Our neutron-diffraction results also reveal the critical exponents β of the magnetic order parameter for the both the CAFM and the HP phase. For both, we find that for the phase transition from the paramagnetic state into the ordered state, β is approximately 0.22. In contrast, for the transition from the HP into the CAFM phase as a function of temperature we find that $\beta = 0.14(1)$. In principle, due to the strong uniaxial anisotropy of ErB₄, we expect that the critical exponents are within the Ising universality class. For two- (2D) and three-dimensional (3D) Ising systems the calculated exponent β is $1/8$ and $0.326\,419(3)$, respectively. This suggests that ErB₄ is close to a 2D Ising system. That the critical exponent deviates from $1/8$ may be because ErB₄ is not a true 2D system, and weak interlayer exchange between different SSL layers exists. This offers an additional explanation of why the supersolid phase is not stabilized in ErB₄.

Using symmetry analysis to refine the integrated intensities measured in our single-crystal neutron-diffraction experiments in applied magnetic field, we further reproduce previous results [35] and show that the zero-field magnetic ground state is indeed a columnar antiferromagnet. Further for the HP phase, we find that the Ising-like spin-flip model A, corresponding to a uuud structure, provides the best agreement with our data. Although it is possible that other spin-flip models, where all spins are slightly canted, may be stabilized, our measurements are not sensitive to these small deviations. In addition, it is not expected that such an arrangement of the magnetic moments would be stable over the entire field range of the HP phase, and other plateaus at fractional magnetization values would be expected as observed in TbB₄ [31,32]. In turn, ErB₄ behaves similar to TmB₄ [34], where due to the strong Ising anisotropy, the $1/2M_S$ plateau arises due to a spin-flip transition in agreement with the uuud model.

Finally, our inelastic neutron measurements resolve the lowest-lying CEF excitation of Er³⁺ with $J = \frac{15}{2}$. This allowed us to determine a minimal CEF Hamiltonian [cf. Eq. (2)]. The expected magnetic moment calculated from the fitted CEF Hamiltonian is $\mu_{\text{CEF}} = 8.96(3) \mu_B$, which agrees well with both the size of the magnetic moments determined from our neutron-diffraction results $\mu_{\text{Er}} = 8.8(9) \mu_B$, as well as the saturation magnetization $M_S \approx 9 \mu_B$ determined from magnetization measurements [29]. This demonstrates that the full Er³⁺ moment expected from the CEF ground state orders. Moreover, the ordered moment is close to the free ion moment of $9.6 \mu_B$. This suggests that the magnetic frustration in ErB₄ is relatively weak and that, consistent with the observed magnetic phase diagram, its low-temperature magnetic properties are mostly driven by strong Ising anisotropy. This is also consistent with the empirical frustration parameter defined as $f = |\theta_{\text{CW}}|/T_N$. Using the Curie-Weiss temperatures $\theta_{\text{CW}} = 11.24$ K ($\mathbf{H} \parallel c$) or $\theta_{\text{CW}} = -23.26$ K ($\mathbf{H} \perp c$) obtained in Ref. [57], no ($f \sim 0.73$) or very weak ($f \sim 1.5$) magnetic frustration would be expected.

A potential explanation for the relatively small frustration parameter, even within the tetragonal plane of ErB₄, is—as discussed in the Introduction—that the presence of the RKKY interaction leads to long-range exchange interactions, which are known to weaken the geometrical frustration resulting from the SSL. In turn, it remains an interesting open question why ErB₄ shows magnetization plateaus similar to insulating SSL materials in spite of the weak frustration. Our measurements motivate neutron spectroscopy studies to determine the underlying magnetic interactions in this system and to shed some light on this question. Apart from clarifying how the interplay of RKKY interactions and Ising anisotropy results in a weakly frustrated ground state in ErB₄ with interesting properties, this will also reveal the exact magnitude of higher-order exchange integrals, such as J_3 and J_4 , that go beyond the nearest- and next-nearest neighbor exchange terms included in the SSL Hamiltonian. Knowledge of J_3 and J_4 may also answer the question whether ErB₄ may be tuned towards the supersolid phase predicted in Ref. [40] using external tuning parameters.

V. SUMMARY

Employing single-crystal neutron diffraction and inelastic neutron scattering, we have investigated the magnetic phase diagram and crystal electric-field ground state of ErB₄. The phase diagram as a function of magnetic field as determined by our study agrees well with previous bulk measurements [29] and shows no sign of the previously proposed spin supersolid phase [40]. Our measurements confirm the magnetic order at zero magnetic field to be of the CAFM type. Further, we show that a uuud spin arrangement (cf. Fig. 7), which corresponds to a spin-flip transition into the HP phase, provides the best description of our neutron-diffraction data and the previously published magnetization data [29]. The critical exponent β of the magnetic order parameter for ErB₄ is close to a 2D Ising model, suggesting that ErB₄ is not purely characterized by a Shastry-Sutherland model but may exhibit finite interlayer exchange interactions. The crystal electric-field ground state of ErB₄ determined by our study

results in an expected magnetic moment $\mu_{\text{CEF}} = 8.96(3) \mu_B$, showing that the moments in the CAFM and HP phases are fully ordered.

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