

Bulk magnetic properties of distorted square lattice compounds M' - Ln TaO₄ ($Ln = Tb, Dy, Ho, Er$)Nicola D. Kelly^{1,*}, Ivan da Silva², and Siân E. Dutton^{1,†}¹*Cavendish Laboratory, University of Cambridge, JJ Thomson Avenue, Cambridge CB3 0HE, United Kingdom*²*ISIS Neutron and Muon Source, Rutherford Appleton Laboratory, Didcot OX11 0QX, United Kingdom*

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We report bulk magnetic properties of the monoclinic lanthanide tantalates, M' - Ln TaO₄ ($Ln = Tb, Dy, Ho, Er$), where the magnetic Ln^{3+} ions are arranged on a distorted 2D square lattice. The heavier analog M' -YbTaO₄ has been investigated as a spin-orbit-coupled, quasi-two-dimensional frustrated magnet, and the properties of the other M' - Ln TaO₄ are expected to vary depending on the electronic configuration of the Ln ion, namely, Kramers vs non-Kramers behavior and different crystal electric field parameters. In this work, powder neutron diffraction is used to confirm the crystal structure for $Ln = Tb, Ho, Er$, and to determine the magnetic structure of M' -TbTaO₄, which displays long-range antiferromagnetic (AFM) order below $T_N = 2.1$ K. The Tb^{3+} moments are aligned primarily along the c axis with AFM nearest-neighbor interactions. Susceptibility data suggest that M' -DyTaO₄ may display short-range ordering around 2.7 K, while M' -HoTaO₄ and M' -ErTaO₄ show AFM correlations but do not order above 1.8 K. Measurements of the magnetic specific heat provide evidence for a Kramers doublet ground state in M' -ErTaO₄, similar to its heavier analog M' -YbTaO₄.

DOI: [10.1103/q8ld-d9vp](https://doi.org/10.1103/q8ld-d9vp)**I. INTRODUCTION**

The spin- $\frac{1}{2}$ Heisenberg square lattice is an important model for frustrated magnetism studies. The two relevant interactions are the nearest-neighbor coupling J_1 , along the sides of the squares, and next-nearest-neighbor J_2 , along the diagonals of the squares. Depending on the relative signs and magnitudes of J_1 and J_2 , three types of classical long-range order (LRO) can be observed [ferromagnetic (FM), columnar antiferromagnetic (AFM), and Néel AFM], and there are also two boundary regions where spin-liquid behavior is predicted [1–3]. These arise from the competition between J_1 and J_2 when J_2/J_1 is close to ± 0.5 . Several experimental realizations of the Heisenberg square lattice have been studied recently, including YbBi₂IO₄ and YbBi₂ClO₄ [4], SrLaCuSbO₆ and SrLaCuNbO₆ [5], and α -KTi(C₂O₄)₂ · x H₂O [6], which all order antiferromagnetically at low temperatures. However, a small number of materials have been found to resist order and are therefore considered candidates for the spin-liquid behavior predicted at $J_2/J_1 \approx \pm \frac{1}{2}$, including transition-metal oxides Sr₂Cu(Te_{1-x}W_x)O₆ [7,8] and a rare-earth oxide NdKNaNbO₅ [9]. Therefore, there is interest in studying other layered materials with magnetic ions on a square lattice.

This work focuses on the lanthanide orthotantalates, Ln TaO₄, with the monoclinic M' crystal structure. The tantalates crystallize in several different structures depending on

the lanthanide ion, temperature, and/or pressure. Compounds of the largest lanthanides, La–Pr, have the monoclinic space group $P2_1/c$, while compounds of the later (smaller) lanthanides can form the M ($I2/a$) or M' ($P2/c$) monoclinic phases or the tetragonal T ($I4_1/a$) phase (which is only observed *in situ* when heated above 1400 °C) [10]. The magnetic Ln^{3+} ions in M - Ln TaO₄ occupy a three-dimensional distorted diamond lattice, which leads to geometric magnetic frustration from the competing nearest-neighbor interactions [11,12]. In contrast, the layered M' phase contains magnetic Ln^{3+} ions on a two-dimensional (2D) distorted square lattice, which also displays frustration.

The crystal structure of M' - Ln TaO₄ is shown in Fig. 1. It is built from alternating layers of edge-sharing TaO₆ octahedra and Ln O₈ square antiprisms, Fig. 1(a). Within each layer of lanthanide-oxygen polyhedra, the Ln^{3+} ions form a distorted square network, Fig. 1(b), with two distinct nearest-neighbor distances, denoted J_{1a} and J_{1b} . The interlayer $Ln - Ln$ separation (e.g., 5.15 Å for M' -TbTaO₄ at room temperature) is more than 30% longer than either of the intralayer $Ln - Ln$ distances (3.71 and 3.96 Å for M' -TbTaO₄). Therefore, we can consider this a quasi-2D system. The combination of highly localized $4f$ orbitals, a quasi-2D magnetic network, and geometric frustration makes it likely that long-range order might be partially or wholly suppressed in these materials.

The M' - Ln TaO₄ phase can be observed for a wide range of Ln ions, although it competes thermodynamically with the M phase: in general, M is favored for the larger lanthanides whereas M' is favored for the smallest lanthanides such as Yb [10]. Recently, two independent studies investigated frustrated magnetism in the spin- $\frac{1}{2}$ system, M' -YbTaO₄. They both determined that YbTaO₄ has a spin-orbit-driven $J_{eff} = \frac{1}{2}$ Kramers doublet ground state with a large gap to the first excited state. The Curie-Weiss temperature θ_{CW} was negative, indicating weak antiferromagnetic correlations between Yb³⁺

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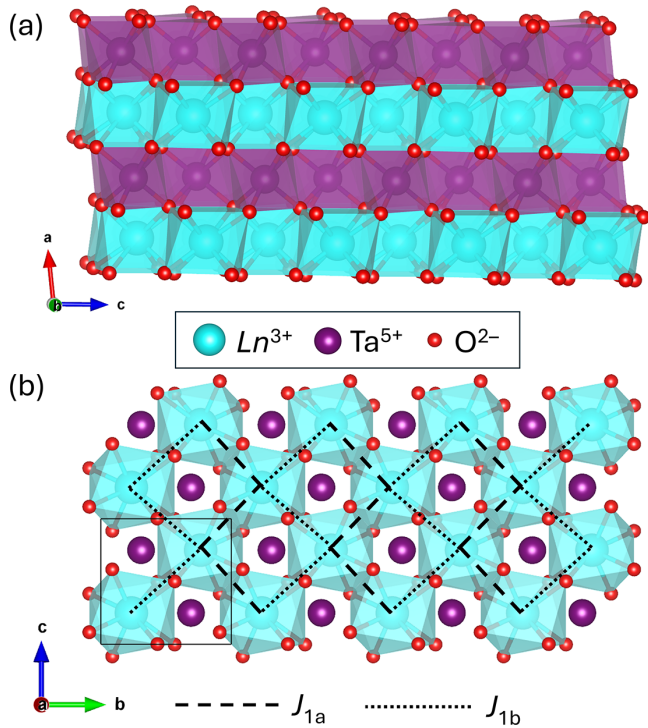


FIG. 1. Crystal structure of M' - LnTaO_4 , space group $P2_1/c$ (No. 13), viewed along different crystallographic axes (b and a , respectively). Blue atoms are Ln^{3+} , purple are Ta^{5+} , and red are O^{2-} . In (b), dashed and dotted lines highlight the distorted square lattice of Ln^{3+} ions.

spins. Ramanathan *et al.* found no magnetic ordering down to 100 mK in specific heat measurements [13]. This means that the frustration index $f = |\theta_{\text{CW}}/T_N| \approx 10$, indicating strong frustration [14]. Kumar *et al.* similarly found a lack of magnetic order at $T \geq 1.8$ K; they also confirmed the presence of fluctuating Yb^{3+} spins using muon-spin relaxation (μSR) spectroscopy [15]. The combined experimental evidence supports a quantum disordered ground state in M' - YbTaO_4 .

In this study, we measure the bulk magnetic properties of the analogous lanthanide tantalate compounds with $\text{Ln} = \text{Tb}$, Dy , Ho , and Er , which each have unique electronic configurations and different quantum numbers L , S , and J . We find that M' - TbTaO_4 orders antiferromagnetically at 2.1 K, while the other three compounds have AFM correlations (Curie-Weiss constants ranging from -3 to -7.5 K) but do not display long-range order above 1.8 K. Neutron diffraction was used to confirm the nuclear crystal structures at room temperature for $\text{Ln} = \text{Ho}$ and Er . Neutron diffraction also allowed us to track the nuclear structure of M' - TbTaO_4 as a function of temperature on cooling to 1.5 K, and to determine its magnetic structure below T_N . We find that M' - TbTaO_4 displays long-range AFM order with an ordering vector of $\vec{k} = (\frac{1}{2}, \frac{1}{2}, 0)$ and the spins directed mainly along the c direction.

II. EXPERIMENTAL METHODS

A. Synthesis

Polycrystalline samples of LnTaO_4 in the monoclinic M' phase were synthesized using ceramic methods on a 0.5–4.0 g

scale. The lanthanide oxides, Tb_4O_7 (99.9%, Alfa Aesar), Dy_2O_3 (99.99%, Alfa Aesar), Ho_2O_3 (99.99%, Alfa Aesar), or Er_2O_3 (99.9%, Alfa Aesar), respectively, were ground in stoichiometric ratio with Ta_2O_5 (99.993%, Thermo Scientific) using a pestle and mortar. The powders were placed in alumina crucibles and heated in air at 3°C min^{-1} to 1200–1450 $^\circ\text{C}$ for 24 h (a more detailed discussion of the optimum temperature for each sample can be found in Sec. III A). The samples were cooled to room temperature at the natural rate of the furnace, ground, and reheated with the same heating profile until the weight fractions of impurity phases no longer decreased (typically 2–4 more times).

B. Powder x-ray diffraction

Phase purity was first checked using powder x-ray diffraction (PXRD) on a Bruker D8 diffractometer with $\text{Cu K}\alpha$ radiation, $\lambda = 1.541 \text{ \AA}$. High-resolution synchrotron PXRD patterns for structural refinement of phase-pure samples were collected at the I11 beamline, Diamond Light Source [16], with wavelength (refined from a silicon standard) $\lambda = 0.82869 \text{ \AA}$. Approximately 50 mg of each sample was mixed 1:1 by volume with ground glass to reduce absorption and packed into $\phi = 0.5$ mm borosilicate capillaries. Rietveld refinement [17] was carried out using the program TOPAS v5 [18]. The background was fitted with a Chebyshev polynomial with 12 coefficients and the peak shape was a Voigt function with axial divergence asymmetry. During refinement, the x and z coordinates of the Ln and Ta atoms were fixed at these values representing special positions in the cell: $x_{\text{Ln}} = 0$, $z_{\text{Ln}} = 0.25$; $x_{\text{Ta}} = 0.5$, $z_{\text{Ta}} = 0.75$. The atomic coordinates of the oxygen atoms were also fixed at values taken from the literature [19]. The refined parameters included the background coefficients, zero offset, peak shape parameters, y coordinates of the Ln and Ta atoms, and atomic displacement parameters B_{iso} for each element.

C. Powder neutron diffraction

Large samples (approximately 4 g each) of M' - TbTaO_4 , M' - HoTaO_4 , and M' - ErTaO_4 were investigated using time-of-flight (TOF) powder neutron diffraction (PND) at the GEM beamline, ISIS Neutron and Muon Source [20]. On GEM, we used six arrays (banks) of detectors to cover a wide range of scattering angles as follows: bank 1, $5 - 12^\circ$; bank 2, $13 - 21^\circ$; bank 3, $24 - 45^\circ$; bank 4, $50 - 74^\circ$; bank 5, $79 - 106^\circ$; bank 6, $142 - 149^\circ$ [21]. Note that M' - DyTaO_4 was excluded from the neutron diffraction study because of the very strong neutron absorption of some Dy isotopes [22] which would require isotopically enriched samples for PND experiments. The samples were packed into $\phi = 6$ mm vanadium cans and measured at 300 K (all samples) and at 100, 30, and 1.5 K (Tb only) using an ILL Orange cryostat [23]. Rietveld refinement [17] of the nuclear structure was carried out using the program TOPAS v5 [18] against banks 3–6 simultaneously, each with equal weighting. For the refinement of TbTaO_4 at 1.5 K, the lower-resolution banks 1 and 2 were also included in order to encompass the (110) magnetic peak at $d = 7.47 \text{ \AA}$.

D. Magnetometry

Magnetic measurements were made using a Quantum Design Magnetic Property Measurement System (MPMS-3). Approximately 10 mg of each sample (weighed accurately in each case) was contained in clingfilm and the standard plastic sample holders then inserted into a brass sample holder. The dc magnetic moment was measured as a function of temperature upon warming in the range 1.8–300 K at a field of 500 Oe, after cooling in zero field (ZFC) or in the applied field (FC). The dc moment was also measured as a function of magnetic field in the range 0–7 T at temperatures of 2, 4, 6, 8, 10 and 100 K.

E. Specific heat

Approximately 40 mg of each sample (weighed accurately in each case) was ground with an equal mass of Ag powder (Alfa Aesar, 99.99%, –635 mesh) using a pestle and mortar, then pressed into a $\phi = 5$ mm pellet. Portions of this pellet weighing 5–20 mg were attached to the sample holder using Apiezon N grease. The heat capacity was measured at $T = 1.8$ –30 K using a Quantum Design Physical Properties Measurement System (PPMS); an initial addenda (background) measurement was made before adding the pellet, in order to subtract the heat capacity contribution arising from the grease. The Ag lattice contribution to the total heat capacity was then subtracted using values from the literature [24]. Furthermore, for each compound M' - Ln TaO₄, the lattice contribution C_{latt} was modelled using the Debye law:

$$C_{\text{latt}} = \frac{9nRT^3}{\theta_D^3} \int_0^{\frac{\theta_D}{T}} \frac{x^4 e^x}{(e^x - 1)^2} dx \quad (1)$$

where n is the number of atoms per formula unit, R is the molar gas constant, T the temperature in K, and θ_D the Debye temperature in K [25]. A scalar parameter p was substituted for $9nR$ in the fitting equation and refined to account for the sample mass error and imperfect coupling to the sample stage. The lattice contribution was then subtracted from all datasets to leave the magnetic heat capacity, $C_{\text{mag}}(T)$, for each compound. The obtained Debye temperatures were 256(2), 197(2), 127(3), and 148(2) K for $Ln = \text{Tb, Dy, Ho, and Er}$ respectively.

TABLE I. Unit cell parameters for M' - Ln TaO₄ ($Ln = \text{Tb, Dy, Ho, Er}$) in space group $P2_1/c$. Values obtained by Rietveld refinement against synchrotron PXRD data, $T = 300$ K, $\lambda = 0.82869$ Å.

	M' -TbTaO ₄	M' -DyTaO ₄	M' -HoTaO ₄	M' -ErTaO ₄
a (Å)	5.14126(4)	5.12917(2)	5.11433(2)	5.09892(4)
b (Å)	5.48935(4)	5.47022(3)	5.45273(2)	5.43875(5)
c (Å)	5.33646(4)	5.31716(2)	5.30149(2)	5.28448(5)
β (deg)	96.6668(5)	96.5857(3)	96.5011(2)	96.4289(4)
V (Å ³)	149.588(2)	148.203(1)	146.893(1)	145.626(2)
y_{Ln}	0.73406(16)	0.73436(15)	0.73385(13)	0.73369(18)
y_{Ta}	0.80846(15)	0.80704(14)	0.80600(12)	0.80578(17)
χ^2	3.68	3.98	4.72	4.21
R_{wp} (%)	2.65	2.93	3.26	2.88

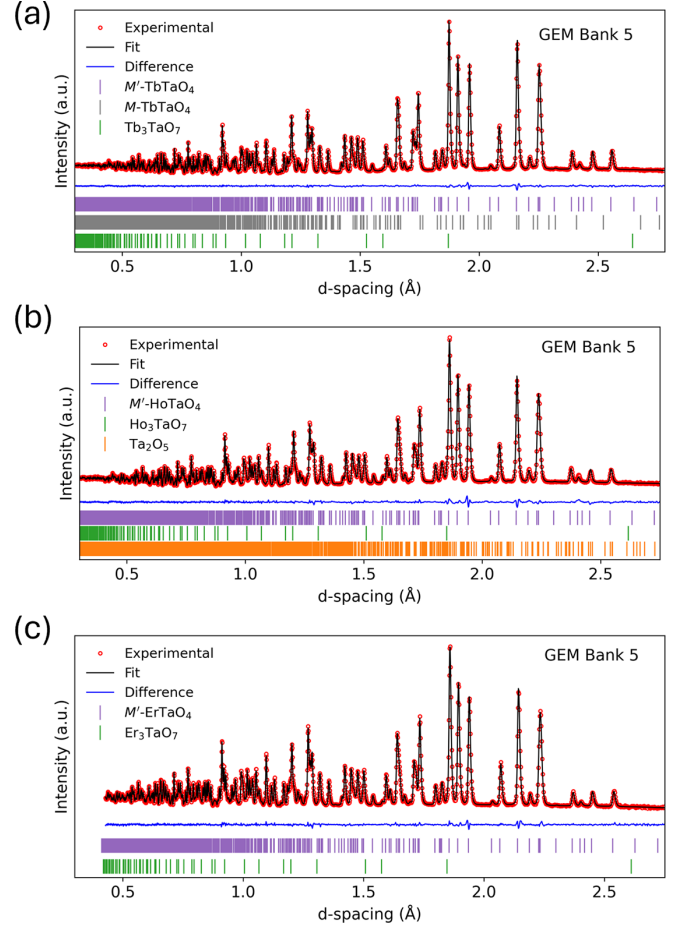


FIG. 2. Rietveld refinement against room-temperature TOF PND data (GEM bank 5) for (a) M' -TbTaO₄, (b) M' -HoTaO₄, (c) M' -ErTaO₄.

III. RESULTS AND DISCUSSION

A. Structural characterization

The phase space of the lanthanide tantalates Ln TaO₄ contains several distinct polymorphs. For $Ln = \text{Tb–Er}$, the equilibrium phase at room temperature is M' , which on heating to around 1400 °C transforms into the tetragonal T (scheelite) phase. However, the T phase cannot be isolated at room temperature and upon cooling transforms into the M (fergusonite) phase [10,26]. Therefore, targeted synthesis of the M' phase requires the temperature to be maintained as high as possible, to facilitate atomic diffusion, whilst not exceeding the $M' - T$ transition temperature [27,28]. The optimum furnace temperature was found to be 1350 °C for $Ln = \text{Tb, Dy and Ho}$, and 1450 °C for $Ln = \text{Er}$. It proved impossible to stabilise phase-pure M' -GdTaO₄ (or any pure M' samples with larger lanthanide ions) because when the synthesis temperature was kept low enough to avoid forming the M phase, there was always competition from Gd₃TaO₇ [29].

The crystal structures of M' - Ln TaO₄ were refined against high-resolution synchrotron powder X-ray diffraction (PXRD) data using the Rietveld method. The refined parameters for all four compounds are given in Table I and the refinement plots are in the Supplemental Material,

Figs. S1–S4 [30]. The PXRD data showed no presence of the competing M -phase in these samples and the refined structural parameters of the four samples agree well with reports from the literature [10,19]. Note: with respect to those literature reports, the origin has been shifted by half a unit cell in the b direction in order to achieve consistency between the nuclear and magnetic unit cell models (see later Sec. III D and Table S2 in Supplemental Material [30]).

Further structural characterization was carried out using powder neutron diffraction (PND) on three samples: M' -TbTaO₄, HoTaO₄, and ErTaO₄ [20]. Figure 2 shows the Rietveld refinement for room-temperature data on all three compounds (bank 5 only; refinements for the other banks are presented in the Supplemental Material [30]). Both PXRD and PND on the neutron samples showed small amounts (≤ 1 wt %) of impurities: Ln_3TaO_7 [28,29]; the competing M phase of TbTaO₄; and Ta₂O₅ in the Ho sample, which unfortunately could not be eliminated after repeated firing of the larger neutron samples. However, all magnetometry and heat capacity measurements were done on smaller samples which were found to be phase pure by high-resolution synchrotron PXRD. Unlike PXRD, the relative neutron scattering lengths [22] permitted refinement of the fractional coordinates of O ions as well as those of the metal ions in our samples, and the refined values (Table II) matched those reported in the literature [19]. The unit cell volumes are linearly related to the lanthanide ionic radius [31] and are consistent with literature reports, as shown by Fig. 3. The differences in refined lattice parameters between synchrotron PXRD and PND data were very small ($<0.05\%$ for representative TbTaO₄), which can be accounted for by small fluctuations in temperature between the different experimental environments.

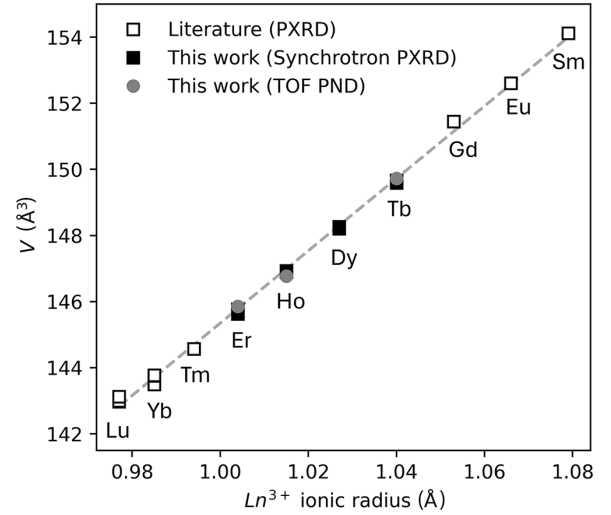


FIG. 3. Unit cell volume of M' - $LnTaO_4$ as a function of eight-coordinate Ln^{3+} ionic radius [31] from Rietveld refinements against both high-resolution x-ray and neutron diffraction data. Error bars are smaller than the data points. Literature data taken from Refs. [10,15,27,32,33].

PND was also measured on the sample of M' -TbTaO₄ at low temperatures using a cryostat. Upon cooling, the a , b , and c lattice parameters decreased while the angle β increased. Overall, the unit cell volume decreased by less than 0.5% between 300 and 1.5 K. The contraction of the unit cell is as expected for normal thermal expansion behavior and no anomalous movements of the atomic positions were detected. The Supplemental Material [30] includes a table of the refined

TABLE II. Refined structural data for M' - $LnTaO_4$ ($Ln = Tb, Ho, Er$) at room temperature from time-of-flight PND data (GEM, banks 3–6). Space group $P2_1/c$. $Ln1$ atom at (0, y_{Ln} , 0.25); Ta1 atom at (0.5, y_{Ta} , 0.75).

		Tb	Ho	Er
a (Å)		5.14258(2)	5.11201(3)	5.1007(15)
b (Å)		5.49176(2)	5.45320(3)	5.4432(15)
c (Å)		5.33763(2)	5.29881(3)	5.2864(15)
β (deg)		96.6566(3)	96.4658(5)	96.4040(5)
V (Å ³)		149.728(1)	146.774(1)	145.86(7)
$Ln1$	y	0.73439(5)	0.73414(7)	0.73380(7)
Ta1	y	0.80767(6)	0.80476(7)	0.80471(8)
O1	x	0.74789(5)	0.74879(7)	0.74955(7)
	y	0.41318(5)	0.41620(7)	0.41672(7)
	z	0.39731(5)	0.39987(8)	0.40129(8)
O2	x	0.27091(6)	0.26904(8)	0.26824(8)
	y	0.06469(5)	0.06442(7)	0.06462(7)
	z	0.49543(6)	0.49575(8)	0.49621(8)
B_{iso} (Å ²)	Ln	0.229(4)	0.148(5)	0.328(7)
	Ta	0.218(4)	0.200(6)	0.281(7)
	O	0.430(3)	0.418(4)	0.498(5)
R_{wp} (%)		1.41	1.73	1.83
χ^2		3.19	3.34	2.29
Impurities		0.47 wt % Tb ₃ TaO ₇ , 1.06 wt % M' -TbTaO ₄	2.7 wt % Ho ₃ TaO ₇ , 1.1 wt % Ta ₂ O ₅	0.8 wt % Er ₃ TaO ₇

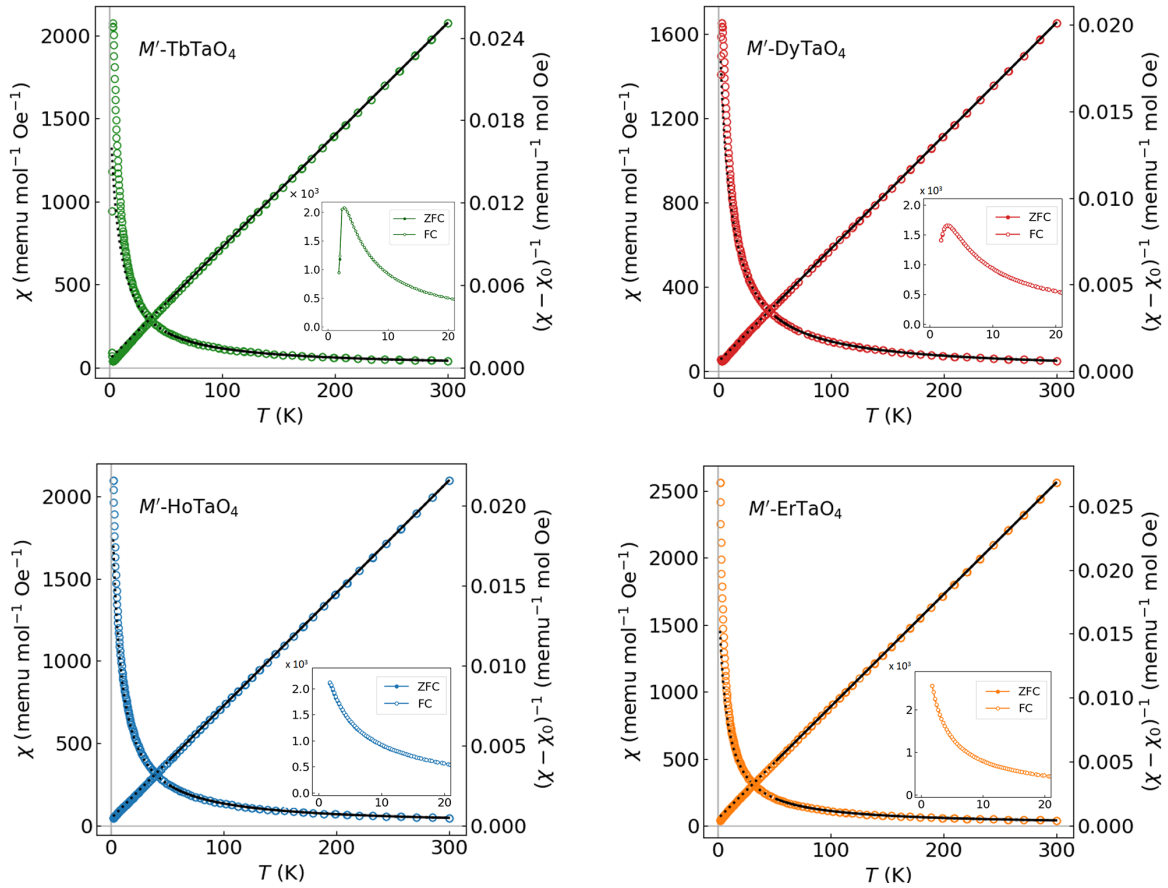


FIG. 4. Magnetic susceptibility χ and inverse susceptibility $(\chi - \chi_0)^{-1}$, fitted to the Curie-Weiss law in the range 50–300 K (black lines), for the four M' - Ln TaO₄ compounds in this study. The black dashed lines show the extrapolation of the fit below 50 K. Insets show the low-temperature region.

lattice parameters, Table S1, and plots of lattice parameters (Fig. S10) and metal-oxygen bond lengths (Fig. S11) as a function of T . At 1.5 K, large new peaks appeared which indicated long-range magnetic order in M' -TbTaO₄; the magnetic structure is discussed in Sec. III D.

B. Magnetic measurements

Peaks in the magnetic susceptibility were visible for M' -TbTaO₄ and M' -DyTaO₄ at approximately 2.7 K for both samples (Fig. 4). There was no divergence between the ZFC and FC susceptibility data which might have indicated spin-glass behavior or other hysteretic effects such as metamagnetism. This, combined with the shape of the peak for M' -TbTaO₄ suggests a transition from paramagnetism to a long-range-ordered AFM state upon cooling. The peak for M' -DyTaO₄ is significantly broader, which may indicate that below 2.7 K the magnetic moments have a short correlation length. Furthermore, the shape of the derivative $d(\chi T)/dT$ [34] shows a clear feature for M' -TbTaO₄ but not for M' -DyTaO₄ (Supplemental Material, Fig. S13 [30]), despite the peaks in $\chi(T)$ occurring at similar temperatures for the two compounds. Meanwhile, M' -HoTaO₄ and M' -ErTaO₄ show only Curie-Weiss behavior with no susceptibility peaks visible in the temperature range measured.

The magnetic susceptibility $\chi = M/H$ was fitted to the modified Curie-Weiss law:

$$\chi = \chi_0 + \frac{C}{T - \theta_{CW}}, \quad (2)$$

where χ_0 is a small temperature-independent susceptibility, C is the Curie constant ($C = \mu_{\text{eff}}^2/8$), and θ_{CW} is the Curie-Weiss temperature. All chemical elements and compounds have a negative, temperature-independent diamagnetic contribution to the susceptibility, χ_{Di} , which can be estimated using Pascal's constants [35] as -8×10^{-5} emu mol⁻¹ for these four M' - Ln TaO₄ compounds. However, diamagnetism is a weak effect and is typically overridden by stronger magnetic effects such as paramagnetism [36], so lanthanide-containing solids are expected to have positive values of χ_0 . We used a Nelder-Mead optimization method (Supplemental Material, Fig. S14 [30]) to find the best linear fit to the inverse susceptibility as a function of temperature. This approach minimizes the scaled least-squares difference between $(T - \theta)/C$ and $(\chi - \chi_0)^{-1}$, which is more effective than regular Curie-Weiss fitting with free parameters on both sides of the equation [Eq. (2)]. This yielded small positive values of χ_0 , which are given in Table III along with the fitted values of θ_{CW} and μ_{eff} . Figure 4 shows the Curie-Weiss fits as black solid lines with extrapolation below 50 K as dashed lines. The negative Curie-Weiss temperatures indicate antiferromagnetic interactions and the

TABLE III. Magnetic parameters for M' - $LnTaO_4$ ($Ln = Tb, Dy, Ho, Er$) from Curie-Weiss fitting in the range 50–300 K.

	M' -TbTaO ₄	M' -DyTaO ₄	M' -HoTaO ₄	M' -ErTaO ₄
Sample mass (mg)	6.2	7.6	6.8	10.8
θ_{CW} (K)	−3.50(12)	−6.75(7)	−5.12(2)	−3.93(5)
μ_{eff} (μ_B)	9.65(6)	10.93(3)	10.58(11)	9.41(3)
Free-ion moment (μ_B)	9.72	10.65	10.61	9.58
χ_0 (emu mol ^{−1} Oe ^{−1})	2.0×10^{-3}	6.9×10^{-4}	1.3×10^{-3}	2.5×10^{-3}

effective magnetic moments μ_{eff} agree quite well with the expected moments for free ions, $\mu = g_J \sqrt{J(J+1)}$.

Magnetic isotherms were measured for all samples at $T = 2, 4, 6, 8, 10$, and 100 K and are shown in Fig. 5. At $T \leq 10$ K and at high fields, the magnetization saturates to a straight line representing the underlying paramagnetism or crystal electric field (CEF) effects. The data at $T = 2$ K were therefore fitted to a linear function between 5 and 7 T and the line was extrapolated back to zero field (grey dashed line) to obtain an estimate for the saturation magnetization, M_{sat} . The magnitude of M_{sat} can be compared with the predicted value ($g_J J$ for Heisenberg, $g_J J/2$ for Ising, or $2g_J J/3$ for XY anisotropy, in powder samples [37]) to give an indication of the single-ion anisotropy in these systems. The data indicate that all compounds exhibit significant single-ion anisotropy and, in particular, that M' -HoTaO₄ may be an Ising system

($M_{sat} = 5$ and $g_J J = 10 \mu_B/\text{f.u.}$) and M' -ErTaO₄ an XY system ($M_{sat} = 6$ and $g_J J = 9 \mu_B/\text{f.u.}$). However, errors arising from the extrapolation and/or the sample mass mean that caution should be exercised in drawing these conclusions from the $M(H)$ data alone. For example, the magnitude of M_{sat} for M' -TbTaO₄ lies somewhat between the expected values for Ising and XY anisotropy, yet the neutron diffraction data (Sec. III D) indicate an almost easy-axis (Ising) behavior. Additional neutron diffraction down to lower temperatures on the Ho and Er samples, and/or an isotopically enriched Dy sample, would be required to confirm the type of anisotropy. Spectroscopic measurements to determine the CEF levels would also provide information on anisotropy.

C. Specific heat

The zero-field magnetic specific heat for M' -TbTaO₄ is given in Fig. 6. A sharp λ -type transition is observed at $T = 2.1$ K, indicating long-range AFM ordering. The temperature of the peak in heat capacity is lower than that observed in the magnetic susceptibility (2.7 K), which is similar behavior to the other polymorph of this compound, M -TbTaO₄ [11]. In addition, a peak in $d(\chi T)/dT$ occurs at the same temperature as the peak in the specific heat (Supplemental Material, Fig. S13 [30]), which is typical for antiferromagnets [34]. Integration of C_{mag}/T yields the magnetic entropy, ΔS_{mag} , which approaches $4 \text{ J K}^{-1} \text{ mol}_{Tb}^{-1}$ at 30 K. This value is less than 20% of the expected $\Delta S_{mag} = R \ln(2J+1) = 21.32 \text{ J K}^{-1} \text{ mol}^{-1}$ ($J = 6$); clearly, since the lower-temperature side of the λ -like peak is incomplete, there is still significant fur-

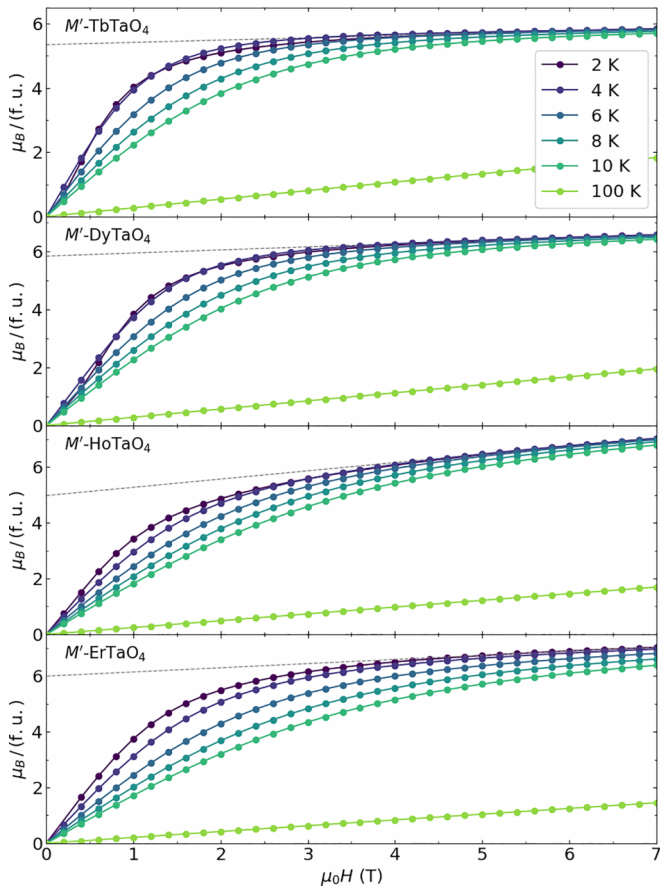


FIG. 5. Magnetic isotherms for M' - $LnTaO_4$. The dashed lines show the linear fits in the region 5–7 T, extrapolated back to 0 T.

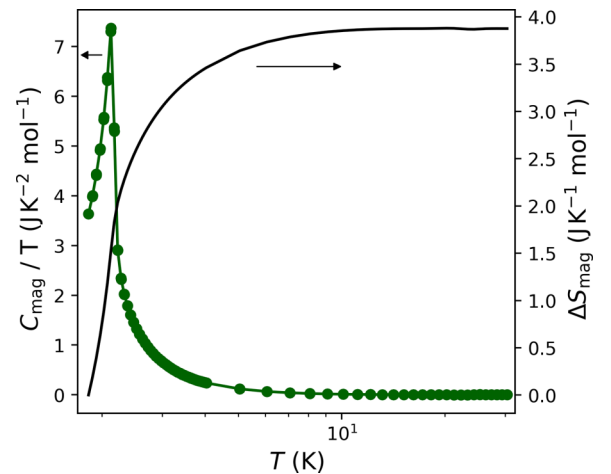


FIG. 6. M' -TbTaO₄ magnetic specific heat data (left axis) and magnetic entropy change (right axis).

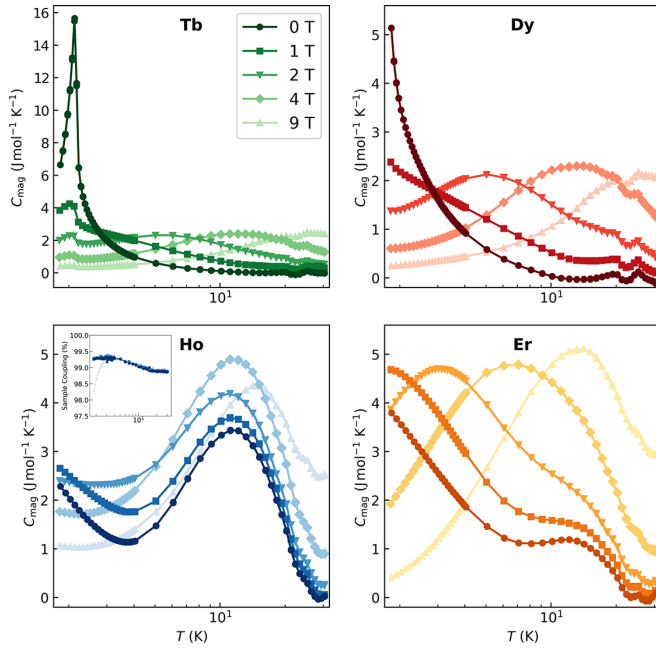


FIG. 7. Magnetic specific heat data at various applied fields for the four M' - Ln TaO₄ samples. Inset for $Ln = \text{Ho}$ shows the sample coupling.

ther entropy to be lost on cooling the material below 1.8 K. The specific heat in zero field and applied fields (1, 2, 4 and 9 T) is also shown in Fig. 7, top left. Upon application of a magnetic field, the intensity of the sharp peak is suppressed and it first shifts to slightly lower temperatures before becoming a broad Schottky-like feature which moves to higher temperatures with increasing field.

In contrast, for M' -DyTaO₄ no such sharp peak is observed (Fig. 7, top right), which agrees with the absence of a peak in $d(\chi T)/dT$ [34] (Supplemental Material, Fig. S13 [30]). There is no evidence for either long-range order or spin freezing above 1.8 K in either the zero-field or 1 T specific heat data. At 2 T, we observe a broad peak, centered around 5 K. This peak then shifts to higher temperatures as the field increases, which is typical of a Schottky anomaly. Such behavior arises from Zeeman splitting of a Kramers doublet state with $J_{\text{eff}} = \frac{1}{2}$, as observed in the isostructural M' -YbTaO₄ at all fields [15]. However, the absence of a peak in the lower-field datasets suggests that the ground state at zero field is not a Kramers doublet, and that there is a field-induced transition between 1 and 2 T, perhaps reflecting suppression of the short-range order suggested by the magnetic susceptibility. In Fig. 5, the magnetization curves for M' -DyTaO₄ at 2 K and 4 K cross over twice at low fields, which also hints at magnetic ordering. There are also two small anomalies in the M' -DyTaO₄ data around $T = 20$ and 25 K whose origin is unknown. They may be an artifact of measurements or related to the sample environment because less pronounced features appear at the same temperatures in the HoTaO₄ and ErTaO₄ data and they are independent of the applied field.

Like Dy³⁺ ions, Er³⁺ ions have an odd number of electrons and are therefore also Kramers ions. M' -ErTaO₄ displays a Schottky-like peak similar to that of M' -DyTaO₄ (Fig. 7,

bottom right) but, unlike DyTaO₄, this behavior appears to extend to the lowest fields. There is a second broad feature between around 7 and 22 K in the zero-field data, which seems to be suppressed in applied magnetic fields and may represent crystal electric field effects or short-range magnetic correlations; the feature might be investigated in the future using neutron diffraction and/or inelastic scattering in this temperature range. The plot of χ^{-1} vs T for this sample showed a significant downturn away from linearity below 30 K, which is consistent with the hypothesis of short-range magnetic correlations in this temperature range. This contrasts with the χ^{-1} vs T plot for HoTaO₄ which remains linear well below 10 K.

Finally, the specific heat data for M' -HoTaO₄ are shown in Fig. 7 (bottom left). A broad hump centered around 8 K is clearly visible and, with the exception of the 9 T data, the temperature of the maximum remains fairly independent of applied field, unlike in the Dy and Er analogues. Broad features like this have been observed in other compounds containing Ho³⁺ (a non-Kramers ion), especially when the Ho³⁺ ions are in low-symmetry crystallographic environments; in π -HoBO₃, the behavior was ascribed to a nonmagnetic singlet ground state, with the broad feature arising from van Vleck paramagnetism [38]. The low-temperature upturn in the heat capacity probably represents the nuclear hyperfine interaction (nuclear Schottky anomaly) as observed in other Ho-containing compounds [38]. In M' -HoTaO₄, the Ho³⁺ ion is on the 2e Wyckoff position of space group 13 ($P2_1/c$) with site symmetry C_2 . The low site symmetry and non-Kramers nature of Ho³⁺ mean that its $2J + 1 = 15$ -fold degeneracy may be lifted by interaction with the surrounding CEF, i.e., there is no symmetry-enforced ground state doublet, unlike in many Er³⁺ or Yb³⁺ compounds. Hence, we suggest that the specific heat data on M' -HoTaO₄ represent a singlet ground state. The higher-temperature peak in the 9 T dataset is probably an anomaly, related to some slippage of the sample platform or thermocouples between the 4 T and 9 T measurements, since there is no evidence in the $M(H)$ data for a field-induced transition in this range. The inset to Fig. 7 shows how the sample coupling for M' -HoTaO₄ is different for the 9 T data at low temperatures, supporting this hypothesis; for the other three compounds, the sample coupling had a slight T dependence but was independent of applied field.

D. Magnetic structure of M' -TbTaO₄

The magnetic structure of M' -TbTaO₄ was investigated using PND [20] below the ordering temperature of $T_N = 2.1$ K. Additional peaks appeared between the 30 K and 1.5 K datasets in positions which indicated commensurate magnetic ordering. Using the FullProf Suite [39], a suitable magnetic ordering vector was found to be $\vec{k} = (\frac{1}{2}, \frac{1}{2}, 0)$. The magnetic peaks were then modeled in TOPAS v5 [18] in magnetic space group 14.80 (P_a2_1/c). Each Tb³⁺ ion carries a magnetic moment with refined components $-0.702(8)$, 0, and $8.053(6)$ μ_B along the a , b , and c axes, respectively. This corresponds to $-1.652(12)$, 0, and $7.996(6)$ μ_B in Cartesian axes and an ordered moment magnitude of $8.17(9)$ μ_B . This is consistent with the expectation of $g_J J = 9$ μ_B for Tb³⁺ ions. Note that this magnetic space group does permit a nonzero

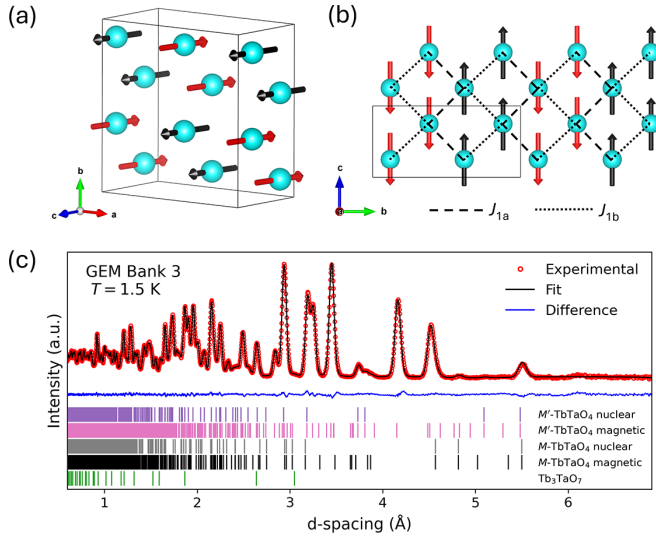


FIG. 8. (a) Magnetic structure model for one magnetic unit cell of M' -TbTaO₄ at 1.5 K. (b) Magnetic structure model for a single layer (distorted square lattice). Dashed lines are the nearest-neighbor distances at 3.7054(4) Å, dotted lines are the next-nearest-neighbor distances at 3.9493(4) Å. (c) Rietveld refinement against TOF PND data at $T = 1.5$ K: GEM bank 3.

y component to the magnetic moment. However, its value always refined to zero (from different trial starting points) with a large uncertainty reported by the TOPAS software, so it was subsequently fixed at zero. Furthermore, a nonzero y component would produce magnetic intensity at the (100) reflection with $d \approx 10.19$ Å, which is completely absent in our data. Thus, in this model, the magnetic moments are primarily directed along the c direction and are fractionally canted toward a . The nearest-neighbor magnetic moments are coupled antiferromagnetically within the bc plane, and the next-nearest-neighbor interactions are ferromagnetic, creating a zigzag-like ordering pattern. Adjacent layers along the a axis are coupled antiferromagnetically.

The magnetic structure model for M' -TbTaO₄ is shown in Figs. 8(a) and 8(b) and the Rietveld refinement at 1.5 K (bank 3) in Fig. 8(c). Rietveld refinements for all banks are available in the Supplemental Material, Fig. S12 [30], along with a table giving the structural parameters for a one-phase combined nuclear and magnetic structure, Table S2. Note that

we were also able to model the magnetic structure successfully by using ISODISTORT [40] and selecting $\vec{k} = (\frac{1}{2}, \frac{1}{2}, 0)$ and irrep mC2+, mk9t3.

In addition to the M' -TbTaO₄, two other minor phases contribute intensity to the 1.5 K neutron diffraction pattern: 1.06 wt % M -TbTaO₄ (both nuclear and magnetic reflections [11]), and 0.47 wt % Tb₃TaO₇. Tb₃TaO₇ is known to undergo two magnetic ordering transitions at 2.9 K and 3.6 K [29], which may be the origin of the very small regions of unindexed intensity in the 1.5 K data. However, the magnetic structure of Tb₃TaO₇ has not been reported and could not be deduced from our data because of the very low weight fraction of Tb₃TaO₇ in this sample.

In comparison to the M' phase described above, the M polymorph of TbTaO₄, which has a three-dimensional stretched diamond network of Tb³⁺ ions, displays AFM order below $T_N = 2.25$ K with the moments directed along the a axis [11]. Both phases of TbTaO₄ display almost easy-axis anisotropy and antiferromagnetic coupling between nearest-neighbor Tb ions. However, differences in their crystal structures lead to important differences in the magnetic structures. In the M phase, there are four near-neighbor AFM interactions at similar distances ($2 \times J_{1a}$ at 3.75 Å, $2 \times J_{1b}$ at 3.84 Å) and the closest ferromagnetically coupled pair of spins are much further apart at ~ 5.05 Å. In contrast, the M' phase has AFM $2 \times J_{1a}$ at 3.71 Å and FM $2 \times J_{1b}$ at 3.95 Å, plus AFM interlayer coupling J_2 (5.13 Å). The tilting of the magnetic moments out of the plane in the M' phase might be a way to partially relieve the frustration that arises from these competing AFM and FM nearest-neighbor interactions. The experimental T_N is slightly lower for the M' phase than the M phase (2.1 vs 2.25 K). However, the Curie-Weiss temperature is smaller for M' than M , and the estimated frustration parameter $f = |\theta_{CW}/T_N|$ is therefore also smaller, although both values are modest (M : $f \approx 4.2$, M' : $f \approx 1.7$). We can use mean-field estimates—neglecting any single-ion anisotropy or longer-range interactions—of the nearest-neighbor exchange interaction (J_{nn}) and dipolar energy scale (D), Eqs. (3) and (4), to rationalize the difference in magnetic ordering temperatures [41–43]:

$$J_{nn} = \frac{3k_B|\theta_{CW}|}{2nJ(J+1)}, \quad (3)$$

$$D = \frac{\mu_0\mu_{\text{eff}}^2}{4\pi R^3}, \quad (4)$$

TABLE IV. Estimates of the nearest-neighbor exchange and dipolar energy scales for M' - and M -LnTaO₄ ($Ln = \text{Tb, Dy, Ho, Er}$). The Curie-Weiss temperatures and effective magnetic moments for the M phase are taken from Ref. [11].

	TbTaO ₄		DyTaO ₄		HoTaO ₄		ErTaO ₄	
	M	M'	M	M'	M	M'	M	M'
θ_{CW} (K)	−9.49	−3.50	−6.88	−3.0	−7.84	−6.38	−7.43	−6.05
μ_{eff} (μ_B)	10.18	9.65	10.71	10.64	10.57	10.66	9.44	9.55
n	4	4	4	4	4	4	4	4
J	6	6	7.5	7.5	8	8	7.5	7.5
R_{nn} (Å)	3.799	3.830	3.786	3.816	3.775	3.805	3.763	3.794
J_{nn} (K)	0.0847	0.0313	0.0405	0.0176	0.0408	0.0332	0.0437	0.0356
D (K)	1.18	1.03	1.32	1.27	1.29	1.28	1.04	1.04

where n is the number of nearest neighbors and J is the total angular momentum quantum number, $J = 6$ for Tb^{3+} . An exact calculation of the dipolar interactions would require knowledge of the magnetic structure, which is not available for all of these compounds, but we can estimate the dipolar energy scale (around 1 K in these materials). We took $n = 4$ for both M and M' structures; for R_{nn} , we used an average of the four closest Tb–Tb distances, which are within 7% of each other (compared with the next closest ion, which is at a distance more than 30% larger than R_{nn} in both phases). In M' -TbTaO₄, its smaller Curie-Weiss temperature and greater average Tb–Tb separation lead to smaller values for both the superexchange and dipolar interactions. These weaker interactions are reflected in the slightly lower magnetic ordering temperature. Table IV gives the relevant parameters and estimated interaction energies for all four samples of M' -LnTaO₄ with comparisons to the M polymorphs [11]. For each pair of compounds with the same Ln , the values of J_{nn} and D are smaller for the M' phase than for the M phase. We also note that the difference in D for M' versus M gets smaller across the lanthanide series (as the average Ln – Ln distance, R , gets smaller), until D is the same for M - and M' -ErTaO₄. This demonstrates an interesting correlation with the minimum Ln^{3+} radius, below which the M phase can no longer be synthesized under ambient pressure conditions [10,44]. The use of pressure to produce compounds (magnetic or nonmagnetic) which are inaccessible by conventional synthesis methods is well documented [45] but typically limited to small volumes of polycrystalline samples. However, the application of a magnetic field during synthesis has lately been revealed as another possible control parameter for synthesis. Under the so-called magnetosynthesis, large single crystals of BaIrO₃ were produced with different structures and magnetic behaviors depending on the field applied during crystal growth [46]. The lanthanide tantalates, with their range of different crystal structures and magnetic properties, may be good candidates for future investigations into the wider applicability of this technique.

Notably, both phases of TbTaO₄ are the only compounds in their respective M' -LnTaO₄ ($Ln = \text{Tb–Yb}$) or M -LnTaO₄ series ($Ln = \text{Nd–Yb}$) which have been found to display long-range order above 1.8 K. Ramanathan *et al.* reported that M' -YbTaO₄ has no long-range order down to 0.1 K, but to our knowledge no other compounds in this family have been studied below 1.8 K. Further low-temperature experiments on the other LnTaO₄ compounds should be carried out where possible, to compare and contrast between different lanthanide ions and to determine whether there is any correlation between the Néel temperatures or magnetic structures of the M and M' phases for lanthanide ions other than Tb^{3+} .

IV. CONCLUSIONS

We have investigated the crystal structure and bulk magnetic properties of the lanthanide tantalum oxides,

M' -LnTaO₄, where $Ln = \text{Tb, Dy, Ho, and Er}$. Powder x-ray and neutron diffraction were used to study the crystal structure. Using magnetometry and heat capacity measurements, we observe LRO at $T_N = 2.1$ K for TbTaO₄ and no LRO above 1.8 K for the compounds where $Ln = \text{Dy, Ho, or Er}$, although the susceptibility of M' -DyTaO₄ suggests possible short-range magnetic ordering below $T \approx 2.7$ K. M' -ErTaO₄ has a Kramers doublet ground state as evidenced by the Schottky-type anomaly in field-dependent specific heat data, similar to the previously reported M' -YbTaO₄. A similar feature was seen for M' -DyTaO₄ above 2 T, but there was no peak in the heat capacity at lower fields. This behavior could be related to short-range order and/or CEF effects.

Powder neutron diffraction indicates that the magnetic moments in M' -TbTaO₄ below T_N are directed primarily along the c axis in a long-range antiferromagnetic structure with $\vec{k} = (\frac{1}{2}, \frac{1}{2}, 0)$, with an ordered magnitude of $8.17(9) \mu_B$ at 1.5 K. We compared the magnetic ordering temperatures of the M and M' polymorphs of TbTaO₄ and related these to the estimated strengths of superexchange and dipolar coupling between Tb^{3+} ions. For all Ln , the interactions are weaker for the M' phase than for the M phase, which correlates with the smaller T_N for M' - vs M -TbTaO₄. This work provides insight into how the magnetic anisotropy and ordering behavior of rare-earth oxides may be significantly affected, either by substitution of different magnetic ions into a given crystal structure or by differences in the crystal structure for a given magnetic ion (specifically Tb^{3+}). Future investigations at subkelvin temperatures will enable us to extend this work to other lanthanide ions.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [47].

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