Magnetic Properties of the Ising-like Rare Earth Pyrosilicate: D-Er$_2$Si$_2$O$_7$

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Ising-like spin-1/2 magnetic materials are of interest for their ready connection to theory, particularly in the context of quantum critical behavior. In this work we report detailed studies of the magnetic properties of a member of the rare earth pyrosilicate family, D-Er$_2$Si$_2$O$_7$, which is known to display a highly anisotropic Ising-like g-tensor and effective spin-1/2 magnetic moments. We used powder neutron diffraction, powder inelastic neutron spectroscopy (INS), and single crystal AC susceptibility to characterize its magnetic properties. Neutron diffraction enabled us to determine the magnetic structure below the known transition temperature ($T_N = 1.9$ K) in zero field, confirming that the magnetic state is a four-sublattice antiferromagnetic structure with two non-collinear Ising axes, as was previously hypothesized. Our powder INS data revealed a gapped excitation at zero field, consistent with anisotropic (possibly Ising) exchange. An applied field of 1 T produces a mode softening, which is consistent with a field-induced second order phase transition. To assess the relevance of D-Er$_2$Si$_2$O$_7$ to the transverse field Ising model, we performed AC susceptibility measurements on a single crystal with the magnetic field oriented in the direction transverse to the Ising axes. This revealed a transition at 2.65 T at 0.1 K, a field significantly higher than the mode softening field observed by powder INS, showing that the field-induced phase transitions are highly field-direction dependent as expected. These measurements suggest that D-Er$_2$Si$_2$O$_7$ may be a candidate for further exploration related to the transverse field Ising model.

I. INTRODUCTION

The identification of Ising-like magnetic materials has been historically important for verifying the many intriguing features of the classical Ising model.¹,² In the context of quantum magnetism, examples of Ising materials which can be tuned to a quantum phase transition (QPT) via a transverse magnetic field are in even higher demand. The Transverse Field Ising Model (TFIM) is one of the most tractable models with a QPT, and thus has been studied extensively theoretically, including in the burgeoning field of non-equilibrium quantum dynamics.³ However, despite the seemingly straightforward ingredients, there are only a few currently known magnetic materials which approximate the TFIM; CoNb$_2$O$_6$ (quasi-1D),⁴ (Ba/Sr)$_2$Co$_2$V$_2$O$_6$ (quasi-1D),⁵,⁶ and LiHoF$_4$ (dipolar coupled 3D).⁷,⁸ With each of these materials, many detailed comparisons to theoretical expectations have been pursued, and even their non-equilibrium behavior are now being explored.⁹ Yet, each material has its own deviations from the ideal models, and the identification of additional TFIM materials, particularly those representing the higher dimensional 2D or 3D (non-dipolar) models, which cannot be solved exactly, are of great interest. The first step in finding new TFIM materials is to find materials with predominantly Ising-like exchange. This type of anisotropic exchange is expected to be more prevalent in 4f rare earth based magnetic systems, as the high spin-orbit coupling provides inherent anisotropy to the system.

Indeed, rare earth based materials have become the subject of increased study in the realm of quantum magnetism in general. Due to the chemical similarity of the rare earths, the same structure can often be stabilized with a variety of rare earth ions. However, the magnetic interactions and anisotropies can be dramatically different between each instance; such is the case for the rare earth pyrochlores¹⁰,¹¹ and the rare earth delafossites.¹²⁻¹⁵ In this work we have investigated a member of the rare earth pyrosilicate (RE$_2$Si$_2$O$_7$) family of compounds which have become the subject of renewed interest due to the discovery of a dimer magnet with evidence for a field-induced Bose-Einstein condensate in Yb$_2$Si$_2$O$_7$.¹⁶,¹⁷ The present work focuses on the magnetic properties of one polymorph of Er$_2$Si$_2$O$_7$. It is worth noting that all of the lanthanide series can be synthesized in this stoichiometry (albeit with many possible structures) making this series an interesting playground for understanding the interplay of magnetic species and crystal structure on the ground state of quantum magnets.

The pyrosilicate compound Er$_2$Si$_2$O$_7$ can crystallize in three different structures based on the synthesis temperature: the low-temperature phase P1 (Type B), the intermediate-temperature phase C2/m (Type C) and the high-temperature phase P2$_1$/a (Type D).¹⁹,²⁰ The focus of this work is the high temperature phase (shown in Fig. 1), hereby referred to as D-Er$_2$Si$_2$O$_7$. Since Er$^{3+}$ is a Kramer’s ion the ground state of the CEF is protected by time-reversal symmetry and thus must be at least doubly degenerate. In the 1970’s the structure of the rare earth pyrosilicates was determined due to interest in the magnetic, electrical, and optical properties of rare earth materials. In particular, the rare earth pyrosilicates were of interest due to the 180° Si-O-Si bond in the [Si$_2$O$_7$]$^{6-}$ groups.²⁰ After some time the magnetic properties of D-Er$_2$Si$_2$O$_7$ were explored via Zeeman spectroscopy and magnetometry.²¹,²² The Zeeman spectroscopy measurements revealed crystal field levels at 27 cm$^{-1}$ (39 K, 3.3 meV) and 52 cm$^{-1}$ (74 K, 6.4 meV), the former of which we have confirmed via specific heat in Appendix VIA. Further, Leask et al. determined an Ising-like g-tensor anisotropy, with $g_x = 2.6$, $g_y = 3.4$, and $g_z = 13.4$.²³ The crystal symmetry and the trivial Er$^{3+}$ site symmetry ($C_1$) results in two orientations of these local axes, with the $x$ axis shared by both. The $z$ axis was found to be 28° (clockwise) from the $a$ axis and ±15° from the $c$-b plane, while $x$ is in the $a$-b plane (see Fig. 2c). However, there is a discrepancy in the $g$-tensor values identified by Leask, et al.²³ and those identified earlier by Maqsood.¹ⁱ This discrepancy could be due to Maqsood using Curie-Weiss fits to determine the values of the $g$-tensor. Curie-Weiss fits can prove unreliable for rare
FIG. 1: a) The crystal and magnetic structure (obtained from the refinement in Fig. 3a) of D-Er$_2$Si$_2$O$_7$ viewed along the a-axis. Bonds in orange dictate the equivalent "intrachain" interactions that form chains and bonds in blue dictate the interchain interactions that form a tessellated, distorted honeycomb lattice. Here Er atoms are green, Si are blue, and O are red. b) View of the crystal and magnetic structure along the b-axis showing the layered nature of the magnetic ions. All panels of this figure were created using the Vesta software.\textsuperscript{18}

Details of the D-Er$_2$Si$_2$O$_7$ sample synthesis have been outlined elsewhere,\textsuperscript{23} but broadly the synthesis was performed by mixing stoichiometric amounts of Er$_2$O$_3$ and SiO$_2$, pressing the powder into dense rods, and heating the rods between 1400°C and 1500°C four times with intermediate re-grinding. Two types of samples were used in the present study. For susceptibility measurements, a small single crystal of pure D-Er$_2$Si$_2$O$_7$ - grown via the optical floating zone technique - was used. The second sample was a powder, which was used for neutron scattering and heat capacity. Rietveld refinement of room-temperature powder x-ray diffraction data indicated that Er$_2$SiO$_5$, a common (and stubborn) impurity in the synthesis of D-Er$_2$Si$_2$O$_7$, made up approximately 9% of the sample. The powder x-ray diffraction data for D-Er$_2$Si$_2$O$_7$ yielded the lattice parameters: $a = 4.68878(8)$ Å, $b = 5.56029(7)$ Å, $c = 10.79659(10)$ Å, $\alpha = 90^\circ$, $\beta = 90^\circ$, $\gamma = 96.043(1)^\circ$. These parameters are consistent with previously published values.\textsuperscript{20} Heat capacity measurements were performed using a Quantum Design PPMS with the heat capacity option on a sintered powder sample of D-Er$_2$Si$_2$O$_7$. AC susceptibility measurements were performed on a single crystal of D-Er$_2$Si$_2$O$_7$ (as confirmed by Laue x-ray diffraction) using a Quantum Design PPMS with the dilution refrigerator and AC susceptor. These measurements were performed at numerous temperatures with $f = 1000$ Hz, $H_{AC} = 0.2$ mT, and with the DC magnetic field applied transverse to the "average Ising direction" ($x$ in Fig. 2). Inelastic neutron scattering (INS) measurements were performed on approximately 5 grams of a sintered powder rod loaded in a Cu canister at the NIST Center for Neutron Research in Gaithersburg, MD, USA using the Multi-Axis Crystal Spectrometer (MACS)\textsuperscript{23} with a fixed final energy ($E_f$) of 2.5 meV, the double focusing monochromator, and Be filters on the incident and scattered beams. Neutron diffraction measurements were performed on the same sample at the High Flux Isotope Reactor at Oak Ridge National Laboratory using the HB-2A (POWDER) diffractometer.\textsuperscript{25} The HB-2A data was collected at 10 K, 2 K, and 0.28 K with the Ge(113) monochromator ($\lambda = 2.41$ Å) and a collimation of open-21°-12°. All errorbars shown in this work indicate $\pm$ one standard deviation. AC susceptibility measurement results (at T = 0.1 K) with a field in the transverse direction (i.e. with AC and DC fields applied along $x$) are shown in Fig. 2b-c). The directions of the Ising axes ($z$) and transverse direction ($x$) in relation to the crystallographic axes is shown in Fig. 2a. We note that the magnetization along this transverse field direction of D-Er$_2$Si$_2$O$_7$ has not been previously studied. For the AC susceptibility measurements, we aligned the sample so that the field was applied along $x$, defined by the angles $\theta = 21.3^\circ$ (the angle from the $a$-axis) and $\phi = 12.8^\circ$ (the angle from the $a$-$b$ plane) as determined by our neutron diffraction measurements discussed in section III B. The decision to use the moment direction found via neutron diffraction for the Ising direction - as opposed to the direction found by Leask et al.\textsuperscript{22} - was based on the expectation that the ordered moment direction is determined in part by the exchange tensor, rather than solely the single-ion

III. RESULTS AND DISCUSSION

A. AC Susceptibility

AC susceptibility measurement results (at T = 0.1 K) with a field in the transverse direction (i.e. with AC and DC fields applied along $x$) are shown in Fig. 2b-c). The directions of the Ising axes ($z$) and transverse direction ($x$) in relation to the crystallographic axes is shown in Fig. 2a. We note that the magnetization along this transverse field direction of D-Er$_2$Si$_2$O$_7$ has not been previously studied. For the AC susceptibility measurements, we aligned the sample so that the field was applied along $x$, defined by the angles $\theta = 21.3^\circ$ (the angle from the $a$-axis) and $\phi = 12.8^\circ$ (the angle from the $a$-$b$ plane) as determined by our neutron diffraction measurements discussed in section III B. The decision to use the moment direction found via neutron diffraction for the Ising direction - as opposed to the direction found by Leask et al.\textsuperscript{22} - was based on the expectation that the ordered moment direction is determined in part by the exchange tensor, rather than solely the single-ion
FIG. 2: a) Schematic diagram showing the average Ising direction “m” and the two local Ising axes. The x-axis is transverse to both Ising axes. Only one of the two y-axes is shown for clarity. Here θ is the angle from the crystallographic a-axis and φ is the angle from the a-b plane. Leask et al.\textsuperscript{25} previously found the single-ion g-tensor to be oriented towards \(θ = 28^\circ\) and \(φ = 15^\circ\). In contrast, we found based on neutron diffraction data that the \textit{ordered} moments lie at \(θ = 21.3^\circ\) and \(φ = 12.8^\circ\). For our transverse-field measurements, we chose to align the crystal perpendicular to the \textit{ordered} moment. b-c) The real (\(χ'\)) and imaginary (\(χ''\)) components of the AC susceptibility at \(T = 0.1\) K, \(f = 1000\) Hz, and \(b_{ac} = 2mT\) for a field ramp from 2.25 T to 3.25 T (blue circle) and a field ramp from 3.25 T to 2.25 T (orange square). A transition is observed at \(\sim 2.65\) T, which shows "hysteresis" in both the real and imaginary components of the susceptibility. It is unclear if this hysteresis is an experimental artifact and this is discussed further in the main text.

B. Neutron Powder Diffraction

Neutron powder diffraction (NPD) data obtained at HB-2A was refined using the FullProf software\textsuperscript{27} and the SARA\textit{h} suite (using the Kovalev tables).\textsuperscript{28,29} Peaks corresponding to D-Er\textsubscript{2}Si\textsubscript{2}O\textsubscript{7} and Cu (from the sample can) were observed at 10 K and 2 K (both are above \(T_N\), but no sign of the impurity (Er\textsubscript{2}SiO\textsubscript{5}) was observed at these temperatures. This is likely due to the strongest nuclear peaks of Er\textsubscript{2}SiO\textsubscript{5} occurring at positions obscured by nuclear peaks of D-Er\textsubscript{2}Si\textsubscript{2}O\textsubscript{7}. A Rietveld analysis (Appendix VI B) of powder neutron diffraction data obtained at 10 K was also performed. The powder diffraction data at 2 K (Appendix VI B) shows diffuse scattering, as expected for a system approaching a continuous phase transition. Data at 0.280 K (below \(T_N\)) show an increase in intensity on peaks corresponding to the D-Er\textsubscript{2}Si\textsubscript{2}O\textsubscript{7} nuclear structure, aside from two peaks at 1.074 Å\textsuperscript{-1} and 1.819 Å\textsuperscript{-1} which can be indexed to the (110) and (310) positions of the impurity phase, Er\textsubscript{2}SiO\textsubscript{5} (space group C2/c, \(a = 14.366(2)\) Å, \(b = 6.6976(6)\) Å, \(c = 10.363(16)\) Å, \(α = 90^\circ\), \(β = 122.219(10)^\circ\), \(γ = 90^\circ\)).\textsuperscript{30} The magnetism of Er\textsubscript{2}SiO\textsubscript{5} has not previously been reported. Thus, we note in passing that since no magnetic impurity peaks were observed at 10 K or 2 K, the magnetic transition in Er\textsubscript{2}SiO\textsubscript{5} is likely between 0.280 K and 2 K, to a |\(\vec{m}\)|= 0 magnetic structure.

In a previous preprint version of this manuscript,\textsuperscript{26} a different set of AC susceptibility measurements was presented, which we later determined were obtained with the field not correctly oriented to the transverse (\(x\)) axis. This was shown via a subsequent set of DC magnetization measurements on the same crystal which showed a higher saturation magnetization than expected for that direction. This misorientation was confirmed by Laue x-ray diffraction to be 37° away from the transverse axis, approximately lying along the (491) direction. In the event that these measurements may still be useful to others, we have provided these DC magnetometry and AC susceptibility measurements in the Appendix (Section VIC 1).
that the ordered moments deduced from our refinement lie 6.9(5)° away from the single-ion Ising direction determined by Leask et. al. However, the overall ordered moment is similar. We measured an ordered moment of 6.56(3) µB at 250 mK and the moment found by Leask was 6.7 µB at 4.2 K. The discrepancy in the Ising axis direction is potentially due to the presence of the (unaccounted for, but small) Er₂Si₂O₇ impurity in our magnetic NPD data. Alternatively, it could be due to a difference in the exchange anisotropy directions compared to the single-ion anisotropy directions. The direction of the ordered moment direction would be influenced by both types of anisotropies. The details of the interactions in D-Er₂Si₂O₇ require further study, ideally by INS on single crystal samples.

C. Field-dependent Elastic Neutron Scattering

Elastic neutron scattering data measured using MACS, using the same powder sample as used at HB-2A, is shown in Fig. 3b and Fig. 3c. An “empty can” background was subtracted. Fig. 3b shows the field evolution of the elastic scattering at an average temperature (Tavg) = 0.16 K. Magnetic Bragg peaks from the impurity (Er₂Si₂O₇) are not resolved, due to the coarser Q-resolution of MACS compared to HB-2A. Intensity versus field cuts for the (002), (011), and (012) reflections are shown in Fig. 3c. The intensity of the peaks does not change significantly between 0 T and 0.25 T, even though the 0.25 T data was measured after going to high field (see caption to Fig. 3 for more detail). Dramatic changes in magnetic peak intensities are observed between 0.25 T and 2 T. Since the experiment was performed on a powder sample, all field directions are averaged here, including the field perpendicular to the Ising direction, which we have shown induces a transition at 2.65 T (Fig. 2). Other field directions are already known to induce transitions in the field range of about 0.5 T. Overall, the dramatic changes in Bragg intensity all occur below about 1 T. Above 1 T, the intensity of the peaks gradually changes, likely due the Er³⁺ moments approaching saturation along the field direction, as much as would be allowed by the Ising axes. An interesting effect is observed away from the Bragg peaks, shown in Fig. 3c as a cut at Q = 0.5 Å⁻¹ (integrated from 0.45 to 0.55 Å⁻¹). The incoherent background appears to increase in intensity at 0.5 T and beyond. The exact nature of this signal is not currently known, but it is not due to an irreversible change in sample environment such as water condensation on the cryostat (we have confirmed this by comparing to scans at similar field ranges taken before and after the dataset shown in Fig. 3c). It is possible that this background is indicative of short-range spin correlations. Near a second order transition, one does expect the critical behavior to manifest as a sharp increase of diffuse scattering. However, this diffuse scattering should diminish rapidly away from the transition, which we do not observe on the high-field side (the intensity seems to plateau after 2 T). We do not see any signatures of the transverse field transition at ~2.65 T in our elastic neutron scattering data. It is likely that the transition field strongly depends on the field direction, thus very few grains in the powder would be in the correct orientation to probe this transition, leading to a small signal in the neutron scattering measurements.

D. Inelastic Neutron Scattering

Finally we turn to the INS data from MACS, shown in Fig. 4a-f. In zero field, two branches of excitations are observed at 0.2 meV and 0.6 meV, which we refer to as Branch 1 and Branch 2, respectively. The branches are gapped and appear to be dispersionless within the resolution of the measurement (the energy step size was chosen to be 0.1 meV, which is similar to the instrument resolution for our settings, ~0.075 meV). 24 The gapped nature of the excitations is consistent with anisotropic interactions, such as Ising exchange, and the presence of two branches implies at least two non-equivalent sites in the magnetic unit cell. This can be easily understood based on the two local Ising axes. As discussed above, we have found a  |⃗k| = 0 antiferromagnetic structure that consists of AFM moments which are collinear when considering sites with the same Ising axis orientation. Thus, each branch is
expected to be two-fold degenerate. Something that is more difficult to understand is that as the field increases, we observe only one branch clearly, despite there still being two distinct Ising axes. At 0.25 T (Fig. 4b), only Branch 2 remains, and is significantly broadened. The disappearance of Branch 1 might indicate a very low field transition for some field directions, but Leask et al. did not report any magnetic transitions below 0.5 T at 500 mK. However, Leask et al. did predict that the transition near 0.5 T was first order in nature for some field directions (significant hysteresis was observed in the simulated magnetization). The absence of Branch 1 in our 0.25 T data may be related to this predicted first order transition; the 0.25 T data was collected after the sample was subjected to a very high field (8 T), thus the data at 0.25 T represents the decreasing field part of the hysteresis curve, which is likely to be in a different state than the 0 T data. However, to understand all the details of the field evolution of excitations would require further study on single crystal samples.

We also obtained INS data at fields up to 8 T (see Appendix V1D), where the excitations could in principle be modelled by linear spin wave theory. Additionally, Leask et al. obtained exchange interactions based on a mean field approach to describing features in the magnetization curves (these parameters, when used in a Monte Carlo simulation, did reproduce many of the observed features of the magnetization curves in several directions). Unfortunately, the exchange interactions as reported in that work are not uniquely assignable to specific pairs of ions in the unit cell, so a useful comparison to our data is greatly complicated. We would like to note that at least the interlayer interaction (along $a$) seems to be clearly defined, and this interaction is small compared to the other exchange interactions. This suggests that the magnetic interactions in D-Er$_2$Si$_2$O$_7$ could be quasi-2D. Indeed, the layered structure of Er$^{3+}$ in D-Er$_2$Si$_2$O$_7$ also suggests that the magnetic interactions may be quasi-2D.

IV. CONCLUSIONS

In this work we have used AC susceptibility, neutron diffraction, and inelastic neutron scattering to study the Ising-like antiferromagnetic order and field-induced behavior of the rare earth pyrosilicate, D-Er$_2$Si$_2$O$_7$. AC susceptibility measurements with a field transverse to the Ising direction show a transition at a magnetic field strength of 2.65 T. Using neutron diffraction we have determined the magnetic structure of D-Er$_2$Si$_2$O$_7$, which consists of moments pointing along a local direction which is close to the Ising direction determined by Leask et al. Our powder INS measurements reveal gapped excitations, one of which softens under an applied field and becomes gapless near 1 T as expected for a field-induced second order phase transition. Beyond ~ 1 T, the branch energy increases again, consistent with entering the field-polarized paramagnetic regime. The absence of Gap 1 in the 0.25 T data may be related to this predicted first order transition; the 0.25 T data was collected after the sample was subjected to a very high field (8 T), thus the data at 0.25 T represents the decreasing field part of the hysteresis curve, which is likely to be in a different state than the 0 T data. However, to understand all the details of the field evolution of excitations would require further study on single crystal samples.

We concern ourselves now with the behavior of Branch 2 at higher fields, which exhibits clearer signatures. As the field is increased to 1 T, Branch 2 broadens (likely due to the anisotropic $g$-tensor) and softens dramatically. The excitation becomes gapless near 1 T as expected for a field-induced second order phase transition. Beyond ~ 1 T, the branch energy increases again, consistent with entering the field-polarized paramagnetic regime. No signature of the transverse field transition is observed in the inelastic spectrum. It is worth noting that we have not attributed any signatures in the INS spectrum to the impurity (Er$_2$SiO$_3$), even though it is also magnetic. Due to the relatively low concentration in the sample (9%), we expect the impurity will not contribute appreciably to the INS signal.

We also obtained INS data at fields up to 8 T (see Appendix V1D), where the excitations could in principle be modelled by linear spin wave theory. Additionally, Leask et al. obtained exchange interactions based on a mean field approach to describing features in the magnetization curves (these parameters, when used in a Monte Carlo simulation, did reproduce many of the observed features of the magnetization curves in several directions). Unfortunately, the exchange interactions as reported in that work are not uniquely assignable to specific pairs of ions in the unit cell, so a useful comparison to our data is greatly complicated. We would like to note that at least the interlayer interaction (along $a$) seems to be clearly defined, and this interaction is small compared to the other exchange interactions. This suggests that the magnetic interactions in D-Er$_2$Si$_2$O$_7$ could be quasi-2D. Indeed, the layered structure of Er$^{3+}$ in D-Er$_2$Si$_2$O$_7$ also suggests that the magnetic interactions may be quasi-2D.
become gapless near 1 T. Due to both neutron scattering experiments being performed on powder samples, it is difficult to directly connect the transition observed in AC susceptibility on a single crystal to any signature in the neutron scattering data. However, we can state that there is a transition for a field applied perpendicular to the Ising axes, the zero-field INS spectra is consistent with Ising-like exchange, and the magnetic structure has been determined via neutron diffraction. This work sets the stage for future studies confirming the Ising-like nature of interactions in D-Er$_2$Si$_2$O$_7$ and for further study of the transverse field transition. Future work should include INS measurements on single crystals in order to further elucidate the nature of the transverse-field-induced transitions in this material.

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VI. APPENDIX

A. Heat Capacity Measurements

The heat capacity for 1.8 K to 100 K is shown in Fig. 5. A broad feature on top of the phonon contribution is observed. The peak of this feature occurs at \( \sim 16 \) K and can be attributed to a crystal field Schottky anomaly around 39 K, consistent with the lowest crystal electric field level measured by Leask et. al. at 27 cm\(^{-1}\) (39 K).\(^{22}\)

![Schottky Anomaly](image)

**FIG. 5:** Specific heat measured on a powder sample of D-Er\(_2\)Si\(_2\)O\(_7\).

B. Magnetic Structure Refinement

Additional neutron diffraction data obtained on a powder sample of D-Er\(_2\)Si\(_2\)O\(_7\) is shown in Fig. 6. The data at 10 K (Fig. 6a) was used to perform a Rietveld refinement on the nuclear structure of D-Er\(_2\)Si\(_2\)O\(_7\) with the Cu peaks masked. This yielded the following lattice parameters at 10K for D-Er\(_2\)Si\(_2\)O\(_7\): \(a = 4.6808(8)\) Å, \(b = 5.5566(2)\) Å, \(c = 10.7864(4)\) Å, \(\alpha = 90^\circ\), \(\beta = 90^\circ\), \(\gamma = 96.064(2)^\circ\). These values found for the nuclear structure of D-Er\(_2\)Si\(_2\)O\(_7\) were used in the refinement of the magnetic structure in Fig. 3a. The neutron diffraction data shown in Fig. 6b is a subtraction of the 10 K data from the 2 K data. This shows the development of diffuse scattering expected in the vicinity of a second order phase transition. This is consistent with the magnetic ordering transition observed in D-Er\(_2\)Si\(_2\)O\(_7\) at 1.9 K.

The basis vector composition of the three basis vectors (\(\psi_{10}\), \(\psi_{11}\), \(\psi_{12}\)) in the \(\Gamma_4\) irreducible representation are shown in Table I. These basis vectors are part of the \(\Gamma_4\) irreducible representation which relates to the “m” point group symmetry and magnetic space group P2\(_1\) /c.

![Neutron diffraction data obtained using HB-2A at 10 K. The fit was performed after removing peaks from the Cu canister.](image)

**FIG. 6:** a) Neutron diffraction data obtained using HB-2A at 10 K. The fit was performed after removing peaks from the Cu canister. b) Neutron diffraction data obtained using HB-2A with the 10 K data subtracted from the 2 K data showing the diffuse scattering expected in the vicinity of a second order phase transition, with a peak centered at 1.2 Å.

C. AC Susceptibility and Magnetometry

1. Measurements along the transverse direction

Additional AC susceptibility measurements for a field applied perpendicular to the Ising direction are shown in Fig. 7. All data in this shown in this section is measured with \(f = 1000\) Hz and \(H_{AC} = 2\) mT. The data shown in Fig. 7a demonstrates the susceptibility up to a DC field of 10 T. The transition observed in Fig. 2 at 2.65 T is also observed here. At high fields the signal becomes diamagnetic, partly from the sample, partly from the mount and glue, and is likely due to the small moment along the transverse field direction which becomes saturated, leading to a small paramagnetic response. The data shown in Fig. 7b-e consists of four constant-temperature field sweeps (increasing field) and demonstrates how the 2.65 T transverse field transition evolves with temperature. There is little change in the imaginary component...
TABLE I: A table showing the positions (x, y, z) of the four Er$^{3+}$ sublattices in the unit cell, the representation for each basis vector in terms of the (non-orthogonal) crystallographic $a, b, c$ axes, the refined contributions ($C_{10}, C_{11}, C_{12}$) of each basis vector, and the resulting refined moment vectors for each Er$^{3+}$ site.

| Atom Info | $\psi_{10}$ | $\psi_{11}$ | $\psi_{12}$ | Moment Direction |
|-----------|------------|------------|------------|-----------------|
| Atom | $x$ | $y$ | $z$ | $m_a$ | $m_b$ | $m_c$ | $m_a$ | $m_b$ | $m_c$ | $(a, b, c)$ basis |
| 1 | 0.88829 | 0.09318 | 0.34934 | 1 | 0 | 0 | 1 | 0 | 0 | 1 | (5.72(3), -2.34(6), -1.45(6)) |
| 2 | 0.11171 | 0.40682 | 0.84934 | 1 | 0 | 0 | 1 | 0 | 0 | -1 | (5.72(3), 2.34(6), 1.45(6)) |
| 3 | 0.11171 | 0.90682 | 0.65066 | -1 | 0 | 0 | -1 | 0 | 0 | -1 | (-5.72(3), 2.34(6), -1.45(6)) |
| 4 | 0.88829 | 0.59318 | 0.15066 | -1 | 0 | 0 | -1 | 0 | 0 | 1 | (-5.72(3), -2.34(6), -1.45(6)) |

FIG. 7: a)-e) AC susceptibility data measured at several temperatures with $f = 1000$ Hz, $H_{AC} = 2$ mT up to a DC field of 10 T (panel a) or 3.5 T (panels b through c) with the field applied perpendicular to the Ising direction (i.e. applied along the x direction). The field-induced transitions are observable as a kink in the real component of the susceptibility ($\chi'$). f) Phase diagram for D-Er$_2$Si$_2$O$_7$ based on the AC susceptibility measurements shown in panels a-e. The transition points represent the points where the real part of the susceptibility ($\chi'$) abruptly changes due to the transition. The shaded region indicates AFM order.

2. Measurements along (491)

As mentioned in the main text, a previous version of this manuscript showed AC susceptibility data that was presented as a field applied transverse to the Ising direction, but it was later discovered that the measurements were actually for a field applied 37° from the average Ising direction, approximately along the (491) direction. We have included these data here (Fig. 8). A comparison of data with $f = 100$ Hz at $T = 0.075$ K (originally shown in the main text) and 0.5 K is shown in Fig. 8a. The data at 0.075 K shows a sharp peak in both the real ($\chi'$) and imaginary ($\chi''$) components at 0.9 T, with a small shoulder near 0.65 T. This shoulder has no significant imaginary component. This changes at 0.5 K, where the shoulder increase significantly in magnitude in the real part of the susceptibility. We also see that the peak in the imaginary component shifts to coincide with the shoulder at 0.65 T. Similar changes are observed for a comparison of data with $f = 1000$ Hz and $T = 0.05$ K and 0.5 K, shown in Fig. 8b. Overall, there is not a large difference between data measured at $f = 100$ Hz and $f = 1000$ Hz. One notable signature is that the transition at 0.65° T changes in magnitude with higher frequencies, indicating there may be some frequency dependence to this transition. The cause for these changes is not currently known, but may indicate a more complex field-induced phase diagram.

The DC magnetometry measurements that enabled us to identify the issue with the alignment are shown in Fig. 9. These measurements were performed with a $^3$He insert for the Quantum Design MPMS3. Measurements were performed for a field applied 37° from the average Ising direction, approxi-
FIG. 8: a) AC susceptibility at 0.075 K and 0.5 K performed with a frequency of 100 Hz in an DC magnetic field applied 37° from the average Ising direction, approximately along the [431] direction. and a 2 Oe AC magnetic field. b) AC susceptibility at 0.05 K and 0.5 K performed with a frequency of 1000 Hz in an DC magnetic field applied 37° from the average Ising direction, approximately along the [431] direction. and a 2 mT AC magnetic field.

The fact that the measured moment is so large along this field direction (6 \( \mu_B \)) led to the discovery that the sample was misaligned, as the transverse field direction would be expected to have a maximum moment of 1.3 \( \mu_B \) (based off Leask et al’s g-tensor value of 2.6 for the transverse field direction).

FIG. 9: a) A grayscale view of numerous constant-temperature field sweeps for the field-derivative of the DC magnetic susceptibility, showing a transition at 0.65 T that decreases in field as the temperature is increased. b-e) Characteristic field sweeps from the data shown in panel a).
D. MACS Data

Additional inelastic slices from the MACS data at 5 T (panel a) and 7 T (panel b) are shown in Fig. 10. Both sets of data show Branch 2 increasing in energy relative to the 3 T data shown in Fig. 4e. Note, in Fig. 10 the energy window was increased from 1 meV to 2 meV to allow all of Branch 2 to be visible.

FIG. 10: a) Energy vs. |Q| slices ($T_{avg} = 0.16$ K) at $\mu_0H = 5$ T. Branch 2 has increased relative to 3 T (Fig. 4e) due to Zeeman splitting. b) Energy vs. |Q| slices ($T_{avg} = 0.16$ K) at $\mu_0H = 7$ T. Branch 2 has increased relative to 5 T, causing the energy window displayed to be increased from 1 meV to 2 meV.
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30 Referring to the notation in Ref., this is the interaction between the atom on “sublattice 1” with itself in the next unit cell.