Palmer-Chalker correlations in the XY pyrochlore antiferromagnet Er$_2$Sn$_2$O$_7$

Solene Guitteny$^1$, Sylvain Petit$^1$, Elsa Lhotel$^2$, Julien Robert$^1$, Pierre Bonville$^3$, Anne Forget$^3$, Isabelle Mirebeau$^1$

$^1$ CEA, Centre de Saclay, DSM/IRAMIS/ Laboratoire Léon Brillouin, F-91191 Gif-sur-Yvette, France
$^2$ Institut Néel, CNRS, 25 Av des martyrs, BP 25, 38042 Grenoble Cedex, France
$^3$ CEA, Centre de Saclay, DSM/IRAMIS/ Service de Physique de l’Etat Condensé, F-91191 Gif-Sur-Yvette, France

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Er$_2$Sn$_2$O$_7$ is considered, together with Er$_2$Ti$_2$O$_7$, as a realization of the XY antiferromagnet on the pyrochlore lattice. We present magnetization measurements confirming that Er$_2$Sn$_2$O$_7$ does not order down to 100 mK but exhibits a freezing below 200 mK. Our neutron scattering experiments evidence the strong XY character of the Er$^{3+}$ moment and point out the existence of short range correlations in which the magnetic moments are in peculiar configurations, the Palmer-Chalker states, predicted theoretically for an XY pyrochlore antiferromagnet with dipolar interactions. Our estimation of the Er$_2$Sn$_2$O$_7$ parameters confirm the role of the latter interactions on top of relatively weak and isotropic exchange couplings.

I. INTRODUCTION

Geometrical frustration has become a central challenge in contemporary condensed matter physics. It is the source of many exotic ground states whose description remains challenging for both theoreticians and experimentalist.$^{1,2}$ These unconventional magnetic states often originate from the strong degeneracy of the ground state manifold, which prevents the stabilization of standard magnetic phases. Whatever their type, perturbations are often driving the low temperature behaviors by lifting partially or totally this extensive degeneracy. Quantum or thermal fluctuations may also enter into play to select and stabilize a particular configuration (or a subset of configurations), a phenomenon called “order by disorder” mechanism.$^{2}$ The family of pyrochlore compounds R$_2$T$_2$O$_7$ (R is a rare earth and T=Ti, Sn, Zr, ...), with the rare-earth magnetic moments localized at the vertices of corner-sharing tetrahedra are model systems to study these subtle order by disorder effects.$^{3,4}$

The case of R=Er compounds is of specific interest: they present a strong XY-like anisotropy, combined with antiferromagnetic interactions leading to a model with an extensive classical degeneracy.$^{5,6}$ The easy magnetic planes are perpendicular to the local $<111>$ ternary axes (XY character), arising from the crystal field properties of the Kramers Er$^{3+}$ ion. While no signature of long-range order could be detected down to 100 mK in Er$_2$Sn$_2$O$_7$, Er$_2$Ti$_2$O$_7$ undergoes a transition towards an antiferromagnetic Néel phase below $T_N = 1.2$ K.$^{7,8}$ This ordered phase has a non-collinear structure, in which the magnetic moments are perpendicular to the local $<111>$ axes in a peculiar configuration denoted $\psi_2$. In Er$_2$Ti$_2$O$_7$, this structure is surprising since dipolar interactions, which are an important perturbation to the isotropic exchange Hamiltonian, are expected to select other magnetic states, called Palmer-Chalker states.$^{9,10}$ However, by considering general anisotropic exchange parameters, it has been recently argued that a quantum order by disorder mechanism$^{11,12}$ explains this $\psi_2$ ordering and accounts for many experimental features. In this context, the reasons for the absence of ordering in Er$_2$Sn$_2$O$_7$ remain an open question.

In this paper, we address this issue by determining experimentally the key parameters of the Hamiltonian of Er$_2$Sn$_2$O$_7$: the crystal electric field (CEF) parameters obtained from inelastic neutron scattering experiments and the anisotropic exchange parameters deduced from the magnetization curves. The main difference between the titanate and stannate parameters is a weaker and less anisotropic exchange tensor. By analyzing neutron scattering data, we demonstrate the existence of short-range correlated domains frozen in the Palmer-Chalker configuration,$^{11,12}$ hence quite different from the $\psi_2$ configuration selected in Er$_2$Ti$_2$O$_7$.$^{13,14}$ We finally show that these configurations are indeed stabilized in a mean field calculation for this set of parameters.

Magnetization and ac susceptibility measurements were performed on a powder sample down to 100 mK using a superconducting quantum interference device (SQUID) magnetometer equipped with a dilution refrigerator developed at the Institut Néel-CNRS Grenoble.$^{15}$ The neutron measurements were performed on the same sample at the cold triple-axis spectrometer 4F2 of LLB-Orphée reactor.$^{16}$

II. MAGNETIZATION AND SUSCEPTIBILITY

Our magnetization measurements first confirm the absence of transition towards a long range ordered state down to 100 mK. The dc susceptibility keeps increasing with decreasing temperature. It presents an upturn below about 2 K, hence deviating from a Curie-Weiss behavior (see the top inset of Figure 1), in agreement with Ref. 7. Below 200 mK, a freezing is observed, as shown by an irreversibility in the zero field cooled - field cooled (ZFC-FC) magnetization and by a frequency dependence in the ac susceptibility (see top Figure 1). The imaginary part of the susceptibility $\chi''$ exhibits a peak whose frequency dependence can be accounted for by an Arrhenius law $\tau = \tau_0 \exp(E/k_BT)$ where $\tau=1/2\pi f$, $\tau_0 = 4 \times 10^{-5}$ s and $E/k_B = 0.9$ K (see bottom Figure 1) in the measured frequency range (0.57 - 211 Hz).
Qualitatively similar features have been observed in this temperature range in other spin liquid compounds such as Gd₃Ga₅O₁₂ [23] or Tb₂Ti₂O₇ [22, 24]. At the moment, no clear picture emerges to explain this freezing, but it could be associated with slow dynamics of correlated spins.

The magnetization curves as a function of field present an inflection point around 1 T for temperatures below 750 mK (see Figure 2). This behavior is reminiscent of the field induced transition observed in Er₂Sn₄O₇ [25, 26], thus suggesting that a field induced order might be stabilized above this field in Er₂Ti₂O₇. Unfortunately, the powder nature of the sample prevents from a detailed analysis of this metamagnetic-like behavior. However, it is worth mentioning that preliminary calculations (using the mean-field model developed in Section V) indicate that reorientations of the magnetic moments occur in the 1 - 1.5 T field range for the three main symmetry directions [110], [100] and [111].

Below 200 mK, an additional curvature develops in the magnetization curve around 0.2 T (see inset of Figure 2) which might be associated with the freezing observed in low field at these temperatures.

FIG. 1: (color online) ac and dc susceptibility vs temperature. Ac susceptibility is plotted for 5.7 Hz < f < 211 Hz with Hac = 1.4 Oe. Top: M/H and real part of the ac susceptibility χ′ vs T. M was measured using the ZFC-FC procedure with H = 50 Oe. The dotted line is a fit to the equation H/M = 0.35 + 0.07T. Bottom: χ″ vs T. Inset: τ vs 1/Tmax showing the Arrhenius behavior with τ₀ = 4 x 10⁻⁵ s and E/kB = 0.9 K.

FIG. 2: (color online) Magnetization M vs internal field Hi (points), along with simulation (lines) (see text). Corrections for demagnetizing effects were made assuming a demagnetizing factor N = 4π/3 (cgs units) [23]. Results for different sets of parameters (assuming Jₐ = 0) are presented to illustrate the sensitivity of the calculation. Inset: M vs Hi for μ₀H < 1 T at 100 (green squares), 200 (blue triangles) and 500 mK (red points).

III. XY ANISOTROPY AND CRYSTAL FIELD ANALYSIS

Aiming at a precise determination of the Er³⁺ anisotropy, the CEF excitations were measured by means of inelastic neutron scattering experiments (see Figure 3), carried out at temperatures of 1.5, 10, 50 and 100 K. Between 0 and 20 meV, three CEF levels are observed at E₁ = 5.1, E₂ = 7.6 and E₃ = 17.2 meV, in agreement with Ref [8]. With increasing temperature, excited states are populated to the detriment of the ground CEF state, giving rise to new modes at hω = E₁ - E₂. The analysis of these spectra is based on the simulation of the scattering function S(Q, ω):

\[
S(Q, \omega) = \sum_{m,n} \frac{e^{-E_m/k_BT}}{Z} (m|\vec{J}|n) \langle n|\vec{J}|m \rangle \times \delta(\omega + E_n - E_m)
\]

where the |m⟩ and E_m are respectively the eigen wavefunctions and eigenvalues of the CEF Hamiltonian HCEF (J = 15/2, g_J = 6/5 for Er³⁺):

\[
H_{CEF} = \sum_{m,n} B_{nm} O_{nm}
\]

The O_nm are the Stevens operators and the B_nm the associated coefficients that remain to be determined (Ref. 29 and references therein). Z is the partition function defined by Z = \sum_m e^{-E_m/k_BT}. Fitting the data through this model yields the coefficients listed in table I (see also Appendix A). The wavefunctions of the ground doublet lead to g_⊥ = 7.52 ± 0.1 and g_/\parallel = 0.054 ± 0.02. For
comparison, the $\text{Er}_2\text{Ti}_2\text{O}_7$ values from Ref~[29] are also given, showing that both compounds have rather similar CEF schemes, but that the $\text{Er}^{3+}$ magnetic moment has a stronger planar character in $\text{Er}_2\text{Sn}_2\text{O}_7$.

### IV. DIFFUSE ELASTIC SCATTERING AND PALMER-CHALKER CORRELATIONS

To further describe the spin liquid state of $\text{Er}_2\text{Sn}_2\text{O}_7$, we have measured the spin-spin correlation function $S(Q,\omega)$ at 1.5 K. An elastic response is observed, forming a broad peak centered at $Q=1.1$ Å$^{-1}$, consistent with the results of Ref~[8]. This response is typical of an elastic diffuse scattering where spin correlations extend over a few inter-atomic distances and are frozen at the time scale of the neutron probe. It is accompanied by a quasi-elastic contribution corresponding to fluctuations of this short range ordered pattern with typical rate $\gamma = 0.5$ meV, namely a typical time of $\tau \sim 10$ ps. The intensities of both contributions increase continuously with decreasing temperature.

Such a diffuse peak does not preclude of any type of magnetic correlations in general. However, given the similarities between $\text{Er}_2\text{Sn}_2\text{O}_7$ and $\text{Er}_2\text{Ti}_2\text{O}_7$, both being antiferromagnets and sharing an XY anisotropy, we propose to model the magnetic ground state in $\text{Er}_2\text{Sn}_2\text{O}_7$ by considering finite size magnetic domains (to account for the peak broadening), chosen among the symmetry allowed patterns for a $k=0$ propagation vector. This modeling is based on a refinement which is constrained by symmetry and physical arguments, as explained below.

The symmetry analysis, performed in the space group $Fd-3m$ using the Basreps software[30] shows that the basis states of the $k=0$ manifold transform as linear combinations of the basis vectors of four Irreducible Representations (IR), labeled $\Gamma_1, \Gamma_5$ in group theory[13,14]. The XY anisotropy is minimized only for: i) linear combinations of the two basis vectors $\psi_1$ and $\psi_2$ which transform according to $\Gamma_5$; ii) a discrete set of basis vectors $\psi_{3,4,5}$ which transform according to $\Gamma_7$. The ground state of $\text{Er}_2\text{Ti}_2\text{O}_7$ and the Palmer-Chalker states (PC)[15] correspond to $\psi_2$ and $\psi_{3,4,5}$, respectively, namely to different IR’s. Table II and the right side of Figure 4 provide the coordinates of these basis vectors and a sketch of the $\psi_2$ and $\psi_3$ magnetic structures (see also Appendix B).

We proceed by fitting the crystalline structure at 50K to determine the overall scaling factor and the lattice parameters. Using these values and assuming a given $\psi$ set, the two remaining parameters of the proposed model are the amplitude of the $\text{Er}^{3+}$ moment and the coherence length of the magnetic domains, which determines the width of the diffuse peaks. As shown in Figure 4, subtracting the high temperature data (50 K) to focus on the magnetic signal only, a very good refinement is obtained with the vectors $\psi_{3,4,5}$ of $\Gamma_7$, yielding an $\text{Er}^{3+}$ moment of 2.8 $\mu_B$ at 1.5 K and a coherence length of about 10 Å[32].

A much worse agreement is obtained with the vectors $\psi_1$ or $\psi_2$ of $\Gamma_5$. In the data ($Q < 1.7$ Å$^{-1}$) of Ref. [8], we have measured the spin-spin correlation function at 1.5 K. At 100 K, the slight discrepancy is attributed to a small evolution of the parameters with temperature.

### TABLE I: Stevens coefficients (in K) for $\text{Er}_2\text{Sn}_2\text{O}_7$ (present work) and $\text{Er}_2\text{Ti}_2\text{O}_7$ (from[29]).

|       | B20 | B22 | B40 | B42 | B43 | B60 | B63 | B66 |
|-------|-----|-----|-----|-----|-----|-----|-----|-----|
| $\text{Er}_2\text{Ti}_2\text{O}_7$ | 616 | 0   | 2850| 0   | 795 | 858 | -493| 980 |
| $\text{Er}_2\text{Sn}_2\text{O}_7$ | 656 | 0   | 3010| 0   | 755 | 738 | -653| 990 |

Inelastic neutron scattering spectra showing the CEF excitations. The lines correspond to the calculation (see text) with the parameters obtained from the fit at 1.5 K. At 100 K, the slight discrepancy is attributed to a small evolution of the parameters with temperature.
denotes an (anisotropic) exchange tensor and \( \tilde{\mathbf{m}}_i \) in this expression, assume an exchange tensor which is diagonal in the (nearest neighbours. Various conventions have been used the dipolar interaction limited to the contribution of the frame linked with a R-R bond 

To carry out this calculation we assume a \( k=0 \) magnetic structure in the Fd-3m space group with face centered cubic (fcc) symmetry. In other words, the 4 \( \text{Er}^{3+} \) moments of a given tetrahedron may be different, but the spin configurations on tetrahedra connected by fcc lattice translations are the same. Following a self-consistent treatment, \( \mathcal{H} \) is diagonalized numerically for each site to determine the energies \( E_{i,\mu} \) and the wave functions \( |\psi_{i,\mu}\rangle \). This yields the magnetic moment (see also Appendix C):

\[
\vec{m}_i = -g_J \mu_B \langle \vec{J}_i \rangle = -g_J \mu_B \sum_{\mu} \frac{e^{-E_{i,\mu}/k_BT}}{Z} \langle \psi_{i,\mu} | \vec{J}_i | \psi_{i,\mu} \rangle
\]

where \( Z = \sum_{\mu} e^{-E_{i,\mu}/k_BT} \). For a given field amplitude, this procedure is repeated for different directions to account for the powder average.

Since \( \mathcal{J}_4 \) is an anti-symmetric exchange constant (Dzyaloshinskii-Moriya like), it is expected to be smaller than the symmetric ones \( \mathcal{J}_{a,b,c} \). Assuming \( \mathcal{J}_4 = \pm 0.005 \) \( K \), the magnetization curve is then well reproduced by the blue and black sets of parameters in Figure 2.

Incorporating the nearest neighbors contribution of the dipolar interaction (\( D_{nn}=0.022 \) \( K \)), see Appendix C) in these anisotropic exchange constants leads to the effective parameters \( \mathcal{J}_a' \sim 0.05 \pm 0.017 \) \( K \), \( \mathcal{J}_b' \sim 0.05 \pm 0.005 \) \( K \) and \( \mathcal{J}_c' \sim 0.005 \) \( K \). Next, it is of great interest to compare these results with the \( \text{Er}_2\text{Ti}_2\text{O}_7 \) exchange parameters listed in Table III and obtained from spin-wave or magnetization curve analysis. We first note that the \( \mathcal{J}_a' \) value in \( \text{Er}_2\text{Ti}_2\text{O}_7 \) is also almost zero when considering the error bars. Interestingly, the symmetric exchange couplings in \( \text{Er}_2\text{Sn}_2\text{O}_7 \) are smaller and more isotropic than in \( \text{Er}_2\text{Ti}_2\text{O}_7 \), thus making the dipolar interaction the main anisotropic interaction.

In this context, according to a number of theoretical work, the ground states in \( \text{Er}_2\text{Sn}_2\text{O}_7 \) should belong to the \( \Gamma_7 \) representation, that is to say to the Palmer-Chalker states. The mean-field phase diagram (see Figure 5) computed in zero field as a function of \( \mathcal{J}_a/\mathcal{J}_c \) and \( \mathcal{J}_b/\mathcal{J}_c \) confirms this assumption. As quoted in Ref. 16, the energetic selection at play in this approach is quite weak and neglects the influence of quantum and thermal fluctuations. Nonetheless, it is useful to explore the type of correlations that might develop depending on the exchange parameters. First, the calculation predicts a canted ferromagnetic "CF" state in the negative \( \mathcal{J}_b/\mathcal{J}_c \) region, which might be relevant in the case of other XY

| Site | 1       | 2       | 3       | 4       |
|------|---------|---------|---------|---------|
|      | CEF axis |         |         |         |
| \( \Gamma_5 \) | \psi_1   | \(-1,1,0\) | \(-1,1,0\) | \(-1,1,0\) |
|      | \psi_2   | \(0,1,1\)  | \(-1,0,1\) | \(-1,0,1\) |
| \( \Gamma_7 \) | \psi_3   | \(-1,1,0\) | \(-1,1,0\) | \(-1,1,0\) |
|      | \psi_4   | \(-1,0,-1\) | \(-1,0,-1\) | \(-1,0,-1\) |

TABLE II: Coordinates of the moments at the 4 sites of a tetrahedron in the different \( \psi \) sets (see text). Note that \( \psi_{3,4,5} \) are obtained by reversing a pair of anti-parallel spins in the \( \psi_1 \) series.

down to 100 mK. By comparison with the present results, the \( \text{Er}^{3+} \) moment likely reaches 3.8 \( \mu_B \) at 100 mK. Note that powder measurements cannot distinguish between the basis vectors of either \( \Gamma_5 \) or \( \Gamma_7 \). However, for a given representation, the relative intensities of the (111) and (220) peaks are fixed. The choice between \( \Gamma_5 \) and \( \Gamma_7 \) IR is thus unambiguous.

V. ESTIMATION OF THE EXCHANGE CONSTANTS IN \( \text{Er}_2\text{Sn}_2\text{O}_7 \), ANALYSIS AND DISCUSSION

We proceed with the estimation of the exchange constants in \( \text{Er}_2\text{Sn}_2\text{O}_7 \) by combining neutron data and magnetization curve analyses. As emphasized above, applying a magnetic field drives the system towards an ordered state, hence making a mean field treatment an acceptable starting point. We thus follow the mean field approach proposed in Ref. 16 and consider the Heisenberg Hamiltonian for R moments \( \tilde{\mathbf{J}}_i \) at sites \( i \) of the pyrochlore lattice:

\[
\mathcal{H} = \mathcal{H}_{\text{CEF}} + \sum_{\langle i,j \rangle} \tilde{\mathbf{J}}_i (\mathbf{\tilde{J}} + \tilde{\mathbf{J}}_{\text{dip}})_{ij} \langle \mathbf{\tilde{J}} \rangle + g_J \mu_B \mathbf{H} \cdot \tilde{\mathbf{J}}_i
\]

In this expression, \( \mathbf{H} \) is an applied magnetic field, \( \tilde{\mathbf{J}} \) denotes an (anisotropic) exchange tensor and \( \tilde{\mathbf{J}}_{\text{dip}} \) the dipolar interaction limited to the contribution of the nearest neighbours. Various conventions have been used to define this anisotropic exchange \( [16][11][14][30] \). Here, we assume an exchange tensor which is diagonal in the \( \{\tilde{a}, \tilde{b}, \tilde{c}\} \) frame linked with a R-R bond \( [25] \):

\[
\tilde{J}_i \tilde{J}_j = \sum_{\mu, \nu = x, y, z} J^\mu_{i,j} (J_0 \delta^\nu_{i,j} + J_0 b_{i,j} \delta^\nu_{i,j} + J_0 c_{i,j} \delta^\nu_{i,j}) J^\nu_j + J_4 \sqrt{2} \tilde{b}_{i,j} \langle \tilde{J}_i \times \tilde{J}_j \rangle
\]

Considering for instance the pair of \( \text{Er}^{3+} \) ions at \( \tilde{r}_1 = (1/4, 3/4, 0) \) \( a \) and \( \tilde{r}_2 = (0, 1, 2, 0) \) \( a \), where \( a \) is the cubic lattice constant, we define the local bond frame as:

\[
\tilde{a}_{12} = (0, 0, -1), \quad \tilde{b}_{12} = 1/\sqrt{2} (1, -1, 0) \quad \text{and} \quad \tilde{c}_{12} = 1/\sqrt{2} (-1, 1, 0).
\]

This Hamiltonian, written in terms of bond-exchange constants, has the great advantage to provide a direct physical interpretation of the different parameters.

The magnetization is given by \( M(\mathbf{H}) = \sum_i \tilde{m}_i \mathbf{H} / \mathbf{H} \), where the \( \tilde{m}_i \) denote the individual magnetic moments.
The shaded area corresponds to the region that accounts for the 4 states of $\Gamma_7$ (with the above determined CEF coefficients). $J_c$ and $J_4$ are fixed to 0.04 K and 0 respectively, while dipolar interaction is included. The AF1 phase resembles very much the $\psi_2$ state of $\Gamma_7$. This ground state is obtained for a strongly anisotropic exchange tensor and especially for ferromagnetic and weakly antiferromagnetic $J_a/J_c$. Finally, the $Er_{2}Sn_{2}O_{7}$ parameters lead to a different ground state labeled AF2 which exactly corresponds to the Palmer-Chalker states ($\Gamma_7$), with a magnetic moment of 3.9 $\mu_B$ and a mean-field ordering temperature $T_N \sim 1.3$ K.

The obtained energy difference between the three states of $\Gamma_7$ is very small so that the ultimate selection is expected to be very fragile with respect to any fluctuations. This mean-field phase diagram thus confirms that the anisotropy and exchange parameters in $Er_{2}Sn_{2}O_{7}$ stabilize Palmer-Chalker correlations as measured experimentally, suggesting that $Er_{2}Sn_{2}O_{7}$ is akin to an XY pyrochlore antiferromagnet with dipolar interactions.

The role of the latter interactions in stabilizing Palmer-Chalker states was pointed out in the case of the Heisenberg pyrochlore $Gd_{2}Sn_{2}O_{7}$. It undergoes a first order transition towards a long range order at about 1 K this ordering being robust with respect to quantum fluctuations. The lack of ordering in $Er_{2}Sn_{2}O_{7}$ thus remains puzzling, but the XY anisotropy as well as the proximity of the AF1 phase might be key ingredients to explain it.

VI. SUMMARY

In summary, $Er_{2}Sn_{2}O_{7}$ does not exhibit long range order down to the base temperature probed of 100 mK, but shows a macroscopic freezing below 200 mK. The magnetic moments have a very strong planar character. From the analysis of the magnetization curve within a mean-field model, the exchange couplings are found to be relatively weak and isotropic. At 1.5 K, the diffuse magnetic scattering is well reproduced by considering short-range correlations corresponding to Palmer-Chalker configurations. These results can be accounted for by a mean-field model which confirms that, with the $Er_{2}Sn_{2}O_{7}$ parameters deduced from the experiments, Palmer-Chalker configurations, stabilized by the dipolar interactions, should be the ground state. In that context, the absence of ordering in $Er_{2}Sn_{2}O_{7}$ remains an open issue, but the present estimation of the CEF parameters and exchange couplings appears to be a starting point for further theoretical calculation.

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Here, we adopt the notations of \cite{19,20} and note that they are different from those used in \cite{20,21}.

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The sample being a powder, the demagnetizing factor \( N \) is hard to estimate. However, the main characteristics of the exchange tensor deduced from the refinement of the magnetization curve are only slightly affected by the chosen value of \( N \), so that the present analysis does not depend on its exact value.

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Fits were performed setting \( J_1 \) to a values of series between -0.02 and +0.02 (which is about 50\% of the symmetric constants. For \( J_1 \) larger than 0.01, the magnetization curve could not be reproduced. Note that, whatever \( J_1 \), all the sets of parameters that reproduce the magnetization curve fall in the Palmer-Chalker part of the mean-field phase diagram of Figure 5.

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**Appendix A: Crystal field**

The CEF parameters are determined using the standard Hamiltonian.

\[
H_{CEF} = \sum_{m,n} B_{nm} O_{nm}
\]

The transition between levels give rise to dispersionless modes in elastic neutron scattering data. The positions and intensities of these modes are fitted in the present study. To illustrate the sensitivity of our determination,
TABLE IV: CEF calculations for different $B_{nm}$ parameters: the two first transitions $E_{1,2}$ along with the g-Landé factors $g_{\perp}$ and $g_{//}$ are listed.

| Site | $B_{20}$ (K) | $B_{40}$ (K) | $B_{43}$ (K) | $B_{60}$ (K) | $B_{66}$ (K) | $E_1$ (meV) | $E_2$ (meV) | $g_{\perp}$ | $g_{//}$ |
|------|-------------|-------------|-------------|-------------|-------------|-----------|-----------|-----------|---------|
| 1    | 656         | 3010        | 755         | 738         | -653        | 990       | 5.08      | 7.64      | 0.054   |
| 2    | 700         | 5.03        | 7.70        | 0.125       | 7.57        |           |           |           |         |
| 3    | 600         | 5.15        | 7.56        | 0.034       | 7.47        |           |           |           |         |
| 4    | 2710        | 5.47        | 8.59        | 1.065       | 7.81        |           |           |           |         |
| 5    | 3310        | 4.77        | 6.92        | 1.405       | 7.09        |           |           |           |         |
| 6    | 800         | 4.90        | 7.41        | 0.013       | 7.52        |           |           |           |         |
| 7    | 700         | 5.30        | 7.91        | 0.10        | 7.53        |           |           |           |         |
| 8    | 800         | 5.74        | 8.14        | 0.84        | 7.68        |           |           |           |         |
| 9    | -600        | 4.93        | 7.18        | 0.98        | 7.28        |           |           |           |         |
| 10   | -700        | 5.24        | 8.14        | 0.84        | 7.68        |           |           |           |         |
| 11   | 1100        | 5.38        | 8.64        | 0.22        | 7.64        |           |           |           |         |
| 12   | 900         | 4.81        | 6.75        | 0.17        | 7.39        |           |           |           |         |

TABLE V: $(\vec{a}, \vec{b}, \vec{c})$ frame for the different sites of a tetrahedron.

| Site | $\vec{a}$ | $\vec{b}$ | $\vec{c}$ |
|------|----------|----------|----------|
| 1    | (1,1,1)  | (1,-1,1) | (1,1,1)  |
| 2    | (1,1,1)  | (1,1,1)  | (1,1,1)  |
| 3    | (1,1,1)  | (1,1,1)  | (1,1,1)  |
| 4    | (1,1,1)  | (1,1,1)  | (1,1,1)  |

Appendix B: Details about the $\psi_2$ basis vector

The description of the possible magnetic structures in Er$_2$Ti$_2$O$_7$ and Er$_2$Sn$_2$O$_7$ XY antiferromagnets is based on the symmetry analysis performed in the Fd-3m space group for a k = 0 propagation vector. As explained in the main text, Er$_2$Ti$_2$O$_7$ undergoes a transition towards an antiferromagnetic Neél phase below $T_N = 1.2$ K[10,12].

This ordered phase has a non-collinear structure, in which the magnetic moments are perpendicular to the local $< 111 >$ axes. This configuration corresponds to the $\psi_2$ basis vector of the $\Gamma_5$ irreducible representation.

To better figure out this peculiar configuration, a different approach can be followed[17,18], considering a series of XY degenerate classical configurations where the magnetic moment at site $i$ is defined in a local frame $(\vec{a}_i, \vec{b}_i, \vec{c}_i)$ given in Table V. Each magnetic moment points along $\vec{a}_i = \cos \phi \hat{a} + \sin \phi \hat{b}_i$, where $\phi$ is a continuous parameter. $\vec{c}_i$ is the local CEF axis. With these notations, the 6 domains of the $\psi_2$ magnetic structure are obtained for $\phi = n\pi/3, n = 0, \ldots, 5$, while the $\psi_1$ (Ref [13,17] also called $\psi_2$ in Ref. [14,18]) are generated for $\phi = \pi/6 + n\pi/3, n = 0, \ldots, 5$.

These configurations are classically degenerate since for arbitrary $\phi$, the classical energy given by $E_c = (4J_{a} - 2(3J_{a} + J_{c}))m^2$, where $(J_{a}, J_{b}, J_{c})$ are the bond exchange parameters defined in the main text, does not depend on $\phi$. The studies published in[17,18] have shown further that the zero point energy $E_0(\phi)$, calculated in the spin wave approximation as a function of $\phi$, breaks this degeneracy, exhibiting weak minima for the 6 $\psi_2$ domains. A particular ordered ground state is thus selected by this quantum order by disorder mechanism.

Appendix C: Mean field model

The present mean field study follows the approach of Ref [16] it is based on the following Hamiltonian for R moments $\vec{J}_i$ at site $i$:

$$
\mathcal{H} = \mathcal{H}_{CEF} + \sum_{<i,j>} \vec{J}_i(\vec{\xi} + \vec{\xi}_{\text{dip}}(\vec{J}_i))(\vec{J}_j) + g_J \mu_B \vec{H}.\vec{J}_i
$$

In this expression, $\vec{H}$ is an applied magnetic field, $\vec{\xi}$ denotes the anisotropic exchange tensor and $\vec{\xi}_{\text{dip}}$ the dipolar interaction limited to the contribution of the nearest neighbours. Various conventions have been used to define this anisotropic exchange[16,15,14]. Here, we assume an exchange tensor which is diagonal in the $(\vec{a}, \vec{b}, \vec{c})$ frame linked with a R-R bond. Considering for instance the pair of Er$^{3+}$ ions at $r_1 = (1/4,3/4,0)a$ and $r_2 = (0,1/2,0)a$, where a is the cubic lattice constant, we define the local bond frame as: $\vec{a}_{12} = (0,0,-1)$, $\vec{b}_{12} = 1/\sqrt{2}(1, -1, 0)$ and $\vec{c}_{12} = 1/\sqrt{2}(-1, -1, 0)$.

$$
\vec{J}_i \vec{J}_j = \sum_{\mu, \nu = x, y, z} J^\mu_{i,\nu} \tau^\mu_{i,\nu} \tau^\nu_{i,\nu} + J^\mu_{i,\nu} \tau^\nu_{i,\nu} \tau^\mu_{i,\nu} + \vec{J}_i(\vec{J}_j \times \vec{J}_j)
$$

Owing to the form of the dipolar interaction, we have:

$$
\vec{\xi}_{\text{dip}}(\vec{J}_i, \vec{J}_j) = D_{nn} \left( \vec{a}_{ij} \vec{a}_{ij} + \vec{b}_{ij} \vec{b}_{ij} - 2 \vec{c}_{ij} \vec{c}_{ij} \right)
$$

with $D_{nn} = \frac{\mu_o}{4\pi} \left( \frac{g_J \mu_B}{r_{nn}^3} \right)^2$ and where $r_{nn}$ is the nearest neighbour distance in the pyrochlore lattice. If we incorporate it in the anisotropic exchange constants $(J_a, J_b, J_c)$, we obtain:

$$
J'_a = J_a + D_{nn}
$$

$$
J'_b = J_b + D_{nn}
$$

$$
J'_c = J_c - 2D_{nn}
$$
Table VI: Anisotropic exchange parameters for Er$_2$Ti$_2$O$_7$ and Er$_2$Sn$_2$O$_7$. $\langle J_{ij}^{\text{coupling Er}} \rangle$ is given in $10^{-2}$ meV while the other sets are in K. Positive values correspond to AF couplings.

As usual in mean field approximations, a self-consistent treatment is carried out to solve the problem: starting from a random configuration for the $\langle J_j^z \rangle$, the contribution to $H$ at site $i$ is diagonalized in the Hilbert space of the Er$^{3+}$ magnetic moment defined by the $\{ J_j \}$, $J_z = -15/2, \ldots, 15/2$ basis vectors, and taking into account the external magnetic field $H$ as well as the molecular field $\sum_{<i,j>} J_i(\vec{J} + \vec{J}_{\text{dip}}_{i,j}) \langle J_j^z \rangle$. This yields the energies $E_{i,\mu}$ and the wave functions $|\psi_{i,\mu}\rangle$. The updated magnetic moments:

$$\langle \vec{J}_i \rangle = \frac{1}{Z} \sum_{\mu} e^{-E_{i,\mu}/k_BT} |\psi_{i,\mu}\rangle \langle \vec{J}_{\mu} | \psi_{i,\mu}\rangle$$

$$Z = \sum_{\mu} \exp -E_{i,\mu}/k_BT$$

is used to proceed at site $j$, and this is repeated until convergence. The magnetization is then given by:

$$M(H) = \sum_i \vec{m}_i \cdot \vec{H}/H$$

Appendix D: Relation with quantum pseudo-spin half models

The anisotropic exchange Hamiltonian can be rewritten in terms of couplings between the spin components of a pseudo spin half defined in the subspace of the ground CEF doublet:

$$\mathcal{H}' = \sum_{i,j} J_{zz} S_i^z S_j^z - J_\pm (S_i^+ S_j^- + S_i^- S_j^+) + J_{\pm \perp} (\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij} S_i^- S_j^-) + J_{\pm \perp} [\zeta_{ij} S_i^+ S_j^- + \zeta_{ij} S_i^- S_j^+] + \delta ij$$

$$g_j \vec{J} = g \vec{S} \quad \text{or} \quad \vec{J} = \lambda \vec{S}$$

In the context of pyrochlores, the $\lambda = \frac{g \mu_B}{g_j}$ matrix is diagonal and takes the form:

$$\lambda = \begin{pmatrix} \lambda_{\perp} & \lambda_{\perp} \\ \lambda_{\perp} & \lambda_{\perp} \end{pmatrix}$$

We call $M$ the matrix that connects the local and global bases and $A$ the matrix connecting $(S_x, S_y, S_z)$ and $(S_+, S_-)$ (we omit the indexes for sake of clarity), so that:

$$\vec{J} = M \lambda A \vec{S}$$

and finally get the following relations:

$$J_{zz} = \lambda_{\perp}^2 \frac{J_a - 2 J_c - 4 J_4}{3}$$

$$J_\pm = \lambda_{\perp}^2 \frac{2 J_a - 3 J_b - J_c + 4 J_4}{12}$$

$$J_{\pm \perp} = \lambda_{\perp} \lambda_{\perp} \frac{J_a + J_c - J_4}{3 \sqrt{2}}$$

and conversely:

$$J_a = 4 \frac{J_{\pm \pm} - J_\pm}{3 \lambda_{\perp}^2} + \frac{4 \sqrt{2}}{3} \frac{J_{\pm \perp}}{\lambda_{\perp} \lambda_{\perp}} - \frac{J_{zz}}{3 \lambda_{\perp}^2}$$

$$J_b = 2 \frac{J_{\pm \pm} + J_\pm}{\lambda_{\perp}^2}$$

$$J_c = \frac{2}{3} \frac{-J_{\pm \pm} + J_\pm}{\lambda_{\perp}^2} + \frac{4 \sqrt{2}}{3} \frac{J_{\pm \perp}}{\lambda_{\perp} \lambda_{\perp}} - \frac{2 J_{zz}}{3 \lambda_{\perp}^2}$$

$$J_4 = \frac{2}{3} \frac{J_{\pm \pm} - J_\pm}{\lambda_{\perp}^2} - \frac{\sqrt{2}}{3} \frac{J_{\pm \perp}}{\lambda_{\perp} \lambda_{\perp}} - \frac{1}{3} \frac{J_{zz}}{\lambda_{\perp}^2}$$

Table VI provides the different sets of anisotropic exchange parameters for Er$_2$Ti$_2$O$_7$ (Ref. [17,36]) and Er$_2$Sn$_2$O$_7$ (present work) deduced from this transformation. This procedure is similar to the ones detailed in references [17,36].