Magnetization process in Er$_2$Ti$_2$O$_7$ at very low temperature

P Bonville$^1$, S Petit$^2$, I Mirebeau$^2$, J Robert$^2$, E Lhotel$^3$ and C Paulsen$^3$

$^1$CEA, Centre de Saclay, IRAMIS/Service de Physique de l’État Condensé, F-91191 Gif-sur-Yvette, France
$^2$CEA, Centre de Saclay, Laboratoire Léon Brillouin, F-91191 Gif-sur-Yvette, France
$^3$Institut Néel, CNRS & Université Joseph Fourier, F-38042 Grenoble, France

E-mail: pierre.bonville@cea.fr

Received 26 February 2013, in final form 26 April 2013
Published 14 June 2013
Online at stacks.iop.org/JPhysCM/25/275601

Abstract

We present a model which accounts for the high-field magnetization at very low temperature in the frustrated pyrochlore compound Er$_2$Ti$_2$O$_7$. In Er$_2$Ti$_2$O$_7$, the Er$^{3+}$ ion has a planar crystal field anisotropy and the material undergoes a transition to antiferromagnetism at $T_N = 1.2$ K. Our model is a mean-field self-consistent calculation involving the four rare earth sites of a tetrahedron, the building unit of the pyrochlore lattice. It includes the full crystal field Hamiltonian, the infinite range dipolar interaction and anisotropic nearest neighbour exchange described by a 4-component tensor. We discuss the equivalence of our treatment of the exchange tensor, taken to be diagonal in a frame linked to a rare earth–rare earth bond, with the pseudo-spin Hamiltonian recently developed for Kramers doublets in a pyrochlore lattice.

(Some figures may appear in colour only in the online journal)

1. Introduction

The pyrochlore titanates, with formula R$_2$Ti$_2$O$_7$, where R is a rare earth, have been the subject of intense studies for a decade [1]. The pyrochlore lattice, made of tetrahedra joined by their vertices, leads indeed to a frustration of the exchange interaction in some specific situations. Owing to different crystal field properties, to the Kramers or non-Kramers character, to the relative importance of the dipolar interaction with respect to exchange, these compounds display a great variety of low-temperature behaviours, the best known being the ‘spin-ice’ ground state occurring in Ho$_2$Ti$_2$O$_7$ and Dy$_2$Ti$_2$O$_7$ [2, 3]. In recent years, there has been a break-through towards a full understanding of the low-temperature properties of the Tb, Er and Yb members of the series, in particular with the recognition that the anisotropy of the nearest neighbour exchange interaction plays an essential role [4–6]. We are interested here in the magnetization at very low temperature in Er$_2$Ti$_2$O$_7$, in the antiferromagnetic (AF) phase, and we will show that the magnetization curves for different field orientations, together with the neutron diffuse scattering in the paramagnetic phase, can be accounted for by using a unique anisotropic exchange tensor.

Er$_2$Ti$_2$O$_7$ is an antiferromagnet with $T_N = 1.2$ K [7–9] and presents an easy magnetic plane, perpendicular to the local ⟨111⟩ ternary axes, arising from the crystal field ground doublet of the Er$^{3+}$ Kramers ion [4], which has a full angular momentum $J = 15/2$. The ground state was determined to be of the so-called $\psi_2$-type, where the Er$^{3+}$ magnetic moments are not coplanar [9, 10]. The ‘order by disorder’ mechanism [11] was suggested to drive this particular ground state selection [9], but other mechanisms have also been investigated [12]; recently, the ‘order by disorder’ scenario has been put on a more robust ground [13, 14]. The phase diagram and the evolution of the magnetic structure upon application of a magnetic field were also determined [15, 16]. A quantum critical point was suggested to occur at $H_q \approx 1.5$ T [15] and the evolution of the spin configuration was quantitatively interpreted in [16] in terms of anisotropic exchange, using the crude approximation of a two-component exchange tensor. A symmetry constrained 4-component anisotropic exchange tensor, in the frame of a $J–J$ coupling, was first introduced for the pyrochlore problem in [5]. Using another definition for
the exchange tensor [17], its four components were derived in Er$_2$Ti$_2$O$_7$ from fitting of the in-field spin wave dispersion laws [14]. In this latter work, the Hamiltonian is written in terms of effective $S = 1/2$ pseudo-spins which represent the ground doublet alone. Such an approach is adequate to describe the physics of Er$_2$Ti$_2$O$_7$ at low temperature (the two first excited doublets have an energy of 73 and 85 K above the ground state [9]) and in zero or moderate magnetic fields (a few T, of the order of the critical field $H_c \approx 1.7$ T, see below), where mixing with the excited doublet is negligible. The pseudo-spin Hamiltonian cannot however capture the high-field magnetization since herein mixing plays an important role. In this work, we start by showing the equivalence of the 4-component pseudo-spin exchange tensor used in [14] and of the exchange tensor used in the present work, whose symmetric part is diagonal in a R–R bond frame. Then we show that the single crystal magnetization curves, for various field directions, in the AF phase of Er$_2$Ti$_2$O$_7$ and the zero field AF ground state are described rather well by mean-field theory, considering the full crystal field interaction and anisotropic exchange in the $J$–$J$ coupling. We compare the results of the model with the field evolution of the individual moments in the ordered state, deduced from neutron diffraction [16]. We finally show that the same exchange tensor also allows the reproduction of the neutron diffuse scattering in the paramagnetic phase.

2. The anisotropic exchange tensor and the self-consistent calculations

Various conventions have been used to define the anisotropic exchange tensor in the pyrochlores [5, 6, 17, 18]. In [5], a transformation of $J$–$J$ coupling exchange integrals into pseudo-spin parameters was performed, but with conventions different from those of the present work. We choose here to consider an exchange tensor whose principal axis is the vector linking two rare earth ions (the R–R bond) [18], which is natural since exchange is a two-ion interaction. Therefore, we take a 3-component exchange tensor $\tilde{J}$ which is diagonal in a ‘bond frame’ $(a, b, c)$, where $c$ is the R–R bond axis (see the appendix for a definition of the bond frames). One can also add an antisymmetric Dzyaloshinskii–Moriya exchange term, with parameter $J_4$. In this frame, the exchange interaction between ions $i$ and $j$ reads as:

$$H_{\text{ex}}^{ij} = \frac{1}{2}[J_a J_x \cdot J_x + J_b J_y \cdot J_y + J_c J_z \cdot J_z + \sqrt{2} J_4 (J' \times J')],$$

where the $J'_{\mu}$ are the components of the full angular momentum. Another choice is made in [14, 17], where an $S = 1/2$ pseudo-spin is considered to describe the ground doublet and the exchange Hamiltonian is written in terms of the pseudo-spin components. In the case of a Kramers doublet in the local frame with trigonal symmetry, the 3-component axial $g$-tensor reads as $\tilde{g} = \{g_{\perp}, g_{\parallel}, g_z\}$, and the projection onto the ground doublet yields the relationship: $g_i J = \tilde{g} S$, where $g_i$ is the ionic Landé factor.

In the appendix, we demonstrate that these two formulations of the exchange Hamiltonian are equivalent, and we show how to derive the relationships between the $\{J_a, J_b, J_c, J_4\}$ tensor used here and the exchange parameters $\{J_{zz}, J_{\pm}, J_{\pm\pm}, J_{\pm\pm}\}$ entering the exchange Hamiltonian in terms of pseudo-spins. They read as follows (for a Kramers doublet):

$$J_{zz} = \left(\frac{g_z}{g_1}\right)^2 - \frac{J_a + 2J_c + 4J_4}{3}$$
$$J_\pm = \left(\frac{g_\perp}{g_1}\right)^2 \frac{2J_a - 3J_b - J_c + 4J_4}{12}$$
$$J_{\pm\pm} = -\frac{3\sqrt{2}}{g_1^2} \frac{J_a + J_c - J_4}{12}$$
$$J_{\pm\pm\pm} = \left(\frac{g_\perp}{g_1}\right)^2 - \frac{2J_a - 3J_b + J_c - 4J_4}{12}.$$  

The reciprocal relations read as:

$$J_a = \left[\frac{1}{3} - \left(-\frac{g_1}{g_\perp}\right)^2 J_{zz} + 4 \left(\frac{g_1}{g_\perp}\right)^2 (J_\pm - J_{\pm\pm}) \right]$$
$$J_b = -2 \left(\frac{g_1}{g_\perp}\right)^2 (J_\pm + J_{\pm\pm})$$
$$J_c = \left[\frac{1}{3} + \left(-\frac{g_1}{g_\perp}\right)^2 J_{zz} + 4 \left(\frac{g_1}{g_\perp}\right)^2 (J_\pm - J_{\pm\pm}) \right]$$
$$J_4 = \left[\frac{1}{3} - \left(-\frac{g_1}{g_\perp}\right)^2 J_{zz} + 2 \left(\frac{g_1}{g_\perp}\right)^2 (J_\pm - J_{\pm\pm}) \right].$$

We emphasize that the physical parameters here are $J_a$, $J_b$, $J_c$, $J_4$ and the two components $g_z$ and $g_\perp$ of the $g$-tensor, the pseudo-spin constants being effective parameters.

We perform the magnetization calculations starting from the trigonal crystal field Hamiltonian, to which is added a Zeeman term, with the same notations and conventions as in [4]. A molecular field is built in the mean-field approximation for each ion on its site from exchange Hamiltonian (1) and considering the six nearest neighbours. The infinite range dipole field on each ion is computed using the Ewald summation method [19, 20] and assuming that all ions in each of the four fcc sublattices forming the pyrochlore lattice have the same moment orientation. The molecular and dipole field are added to the applied field and a self-consistent calculation is performed, which involves the four sites of a tetrahedron. This type of calculation is thus limited to magnetic structures with a $\mathbf{k} = 0$ propagation vector, either spontaneous or field induced. It gives access to the moment arrangement within a tetrahedron and to the magnetization.
3. Magnetic properties in the antiferromagnetic phase

3.1. Isothermal magnetization curves

The magnetization versus field has been measured in the AF phase of an Er$_2$Ti$_2$O$_7$ single crystal, at 0.15 K, using a SQUID magnetometer equipped with a dilution refrigerator developed at the Institut Néel-CNRS. The magnetic field was applied along the three symmetry directions [111], [110] and [100], and the data are shown as black dots in figure 1. The demagnetization factor was negligible for the three directions. The data for H || [111] are in good agreement with previous measurements at 0.5 K [21]. For H || [111] and [110] (figures 1(a) and (b)), one observes a steep increase of the magnetization with a slight upwards curvature up to a critical field $H_C \simeq 1.6$ T; for H || [100] (figure 1(c)), the initial increase is linear and the critical field slightly higher: $H_C \simeq 1.8$ T. Above $H_C$, a much slower linear increase is observed up to the maximum field of 7 T. Using the method described in section 2, we have computed the field variation of the magnetization for each field direction. The magnetic structure has $K = 0$ when the field is applied along the [110] direction [15, 16] and it is probably so for H || [111] and [100], as can be concluded a posteriori from our study. The Er$^{3+}$ ion ($J = 15/2, g_J = 6/5$) is described by its full angular momentum J, and the g-tensor of the ground doublet has components: $g_z = 2.6$ and $g_\perp = 6.8$ [4].

Starting from the pseudo-spin exchange parameters derived for Er$_2$Ti$_2$O$_7$ in [14], relations (3) allow us to obtain the exchange integrals $J_a, J_b, J_c$ and $J_d$. Actually, the set of pseudo-spin parameters $\{J_{2z}, J_\perp, J_{z\perp}, J_{\perp\perp}\}$ includes the dipolar interaction limited to first neighbours, whereas our calculation makes use of the exchange only integrals and of the infinite range dipolar interaction. Then one must replace $J_a, J_b$ and $J_c$ in relations (3) respectively by $J_a - D, J_b - D$ and $J_c + 2D$ (see appendix), where D is the characteristic first neighbour dipolar energy of 0.020 K in Er$_2$Ti$_2$O$_7$. One obtains, taking into account the error bars given in [14] for the pseudo-spin parameters: $J_a = 0.067 \pm 0.048$ K, $J_b = -0.057 \pm 0.01$ K, $J_c = -0.071 \pm 0.060$ K and $J_d = -0.15 \pm 0.015$ K. Except for $J_b$, the range of acceptable values is quite large. The isothermal magnetization being quite sensitive to the particular values of the exchange parameters, the calculation of its field variation at 0.15 K for H || [111], [110] and [100] allows us to reduce significantly the error bars. We obtain the best match to the data (see figure 1) with the following parameter values: $J_a = 0.030 \pm 0.005$ K, $J_b = -0.050 \pm 0.005$ K, $J_c = -0.105 \pm 0.01$ K and $J_d = \pm 0.005$ K. The slight upward curvature at low field for H || [111] and [110] and its absence for H || [100], the critical field values and the linear increase of the magnetization above $H_C$ are well reproduced. The derived exchange parameter values, while lying within the range determined in [14] from the fit of the spin wave dispersion laws, represent a much more precise set for the exchange tensor. In this latter work, the fitted g-values ($g_z \simeq 2.5$ and $g_\perp \simeq 6$) are close to those obtained in [4]. We note that the g-tensor derived from EPR measurements [22], i.e. $g_z \simeq 0.24$ and $g_\perp \simeq 7.6$, is rather far from our values and much more anisotropic.

With these exchange integral values, our mean-field calculation yields the zero field ground configuration to be the $\psi_2$ state, as determined from neutron diffraction [9, 10]. The selection of $\psi_2$ in this case results from the crystal field anisotropy alone (although rather weak [12]) induced by mixing of the excited crystal field states into the ground state by the exchange/dipole interaction. The alternative ‘order by disorder’ scenario for selecting the ground state [13, 14] is probably more robust than the crystal field anisotropy for establishing the correct ground configuration; we note that it implies the existence of a small gap of 0.02 meV in the spin wave spectrum which has not yet been observed.
In addition, we find that the Er moments in the ground configuration do not lie exactly in their easy plane, but have a small \((\approx 0.01 \, \mu_B)\) out-of-plane \(z\)-component as a consequence of the above mentioned anisotropy. The mean-field Néel temperature is \(T_N = 2.62\, K\), higher than the experimental value 1.2 K. This enhancement of the mean-field \(T_N\) value with respect to the actual value is probably due to the effect of spin fluctuations, which are not taken into account in the mean-field description [14].

3.2. The in-field magnetic structures

Our model also gives vectorial information about the individual Er moments. The calculated field variations of the moduli of the four moments in a tetrahedron are represented in the insets of figure 1. For \(H \parallel [111]\), the Er moment at the site with its ternary axis parallel to the field has a rather simple behaviour. Its modulus (black curve in the inset of figure 1(a)) is seen to strongly decrease as \(H\) increases, signalling a departure from the easy plane; it reaches a minimum value near the critical field \(H_c \approx 1.6\, T\), and increases linearly with \(H\) above \(H_c\). For \(H \parallel [110]\), a similar behaviour is obtained for the \(\alpha\)-sites (black and red curve in the inset of figure 1(b)). For both these field directions, there are three different behaviours for the moments as a function of field, which implies that the field direction is not a symmetry axis for the magnetic structure. By contrast, for \(H \parallel [100]\) (figure 1(c)), a single behaviour is obtained for the four sites, suggesting that [100] is a symmetry axis for the field induced structure, which could be checked by in-field neutron diffraction. Neutron diffraction data were obtained at 0.3 K for \(H \parallel [110]\) in Er\(_2\)Ti\(_2\)O\(_7\) [15, 16] and were interpreted in [16] using, as a crude approximation, a two-component exchange tensor diagonal in the local frame. We replotted here the data of [16] (points in figure 2) and compare the evolution of the magnetic structure with calculations using the presently derived 3-component exchange tensor diagonal in the bond frames, to which is added Dzyaloshinskii–Moriya exchange (lines in figure 2). This gives a physical insight about the field evolution of the magnetic structures, which present similar features whatever the field direction. The field variation of the angles of the moments with their local axis (figure 2(b)) and with the field (figure 2(c)) is rather well reproduced, showing that the moments rotate towards the field direction up to the critical field value. Above this field, they align along (or close to) the field, and thus the critical field can be considered as the ‘spin flip’ field of the AF structure, the linear increase of the moment moduli (and of the magnetization) above \(H_c\) being caused by quantum mixing with the excited crystal field states. By contrast, the experimental values of the moment moduli (figure 2(a)) are below the calculated curves, especially around the critical field at 1.6 T. We note that the magnetization along the same field direction (figure 1(b)) is rather close to the calculated curve, but also lies below it. The experimental moment values can be subject to some uncertainty since they rely on the refinement of the neutron diffraction lines for a \(k = 0\) structure, which have a rather small intensity (the moment value is 2–3 \(\mu_B\)) growing on the nuclear diffraction peaks. Another cause for the discrepancy could be the presence of the quantum critical point at 1.6 T for this field direction [15], causing enhanced spin fluctuations which the mean-field model cannot take into account.

4. Diffuse scattering in the paramagnetic phase

The neutron diffuse scattering in the paramagnetic phase at 2 K reported in [23] is displayed in the left part of figure 3. It shows intense spots near the Bragg positions (111), (133), (311), (333) . . . , and less intense maxima near (000), (022) . . . . Our calculations (figure 3 right part) of this diffuse scattering are performed using the RPA approximation [24] with the anisotropic exchange tensor derived above from the magnetization curve, and considering the Fourier transform of the infinite range dipolar interaction [25, 26]. The temperature was chosen at 3.2 K since the mean-field Néel temperature with these parameters is 2.62 K. One can see that our
Figure 3. Diffuse neutron scattering at 2 K in Er$_2$Ti$_2$O$_7$ in the \((hk0)\) plane (upper panel) and in the \((hkk)\) plane (lower panel) of the reciprocal space: left: experimental data from [23]; right: simulated maps (at 3.2 K) using the exchange tensor $J_a = 0.030$ K, $J_b = -0.050$ K, $J_c = -0.105$ K and $J_4 = 0$ in the 4-site RPA approximation.

simulations reproduce satisfactorily the experimental data of [23]. We note that they capture the high-intensity diffuse spots at Bragg positions, precursor to the long-range magnetic ordering with $\mathbf{k} = 0$ occurring at lower temperature, whereas the simulations performed in [23] do not. We believe this is due to the fact that the approach used in this latter work, i.e. the diagonalization of the 4-site exchange/dipolar Hamiltonian on a tetrahedron, cannot describe the critical correlations above $T_N$ responsible for these diffuse Bragg spots.

5. Conclusion

We have presented a mean-field approach for frustrated pyrochlore systems which takes into account the full crystal field level scheme of the rare earth ion, the anisotropic nearest neighbour exchange and the infinite range dipolar interaction. The model is applied to investigate the high-field low-temperature magnetization curves in the antiferromagnet Er$_2$Ti$_2$O$_7$, together with other physical properties. We show the equivalence of the exchange tensor in $J-J$ coupling diagonal in the bond frame we use here and of the pseudo-spin $S = 1/2$ exchange tensor recently proposed. Our fit of the magnetization curves in the AF phase for different field directions, where the quantum mixing with excited crystal field states is well accounted for, allows us to derive more precise values for the exchange parameters. These parameters also describe well the diffuse neutron scattering in the paramagnetic phase. The advantage of the present approach with respect to models dealing with $S = 1/2$ pseudo-spins is that it can take into account the effects linked with quantum mixing of crystal field states by the magnetic field, which become important above the critical, of 'spin flip', field of the magnetic structure.

Acknowledgments

We are grateful to P Dalmas de Réotier and C Marin (CEA Grenoble, SPMS, France) for providing the Er$_2$Ti$_2$O$_7$ single crystal used for the magnetization measurements, and to C Decorse (ICMMO, Université Paris-Sud, Orsay, France) for the single crystal used for the neutron diffraction measurements.

Appendix. Relations between the exchange tensor appropriate to the $S = 1/2$ pseudo-spin Hamiltonian and the exchange tensor in the ‘bond frame’

We start by choosing a set of vectors forming an orthonormal frame attached to the bond linking rare earth neighbours $i$ and $j$. Calling $\mathbf{e}_i$ the unit vector along the \((111)\) trigonal axis at site $i$, we define the following unit vectors forming the ‘bond frame’, where $\mathbf{e}_{ij}$ lies along the link between the two
neighbouring sites:
\[
\begin{align*}
  c_{ij} &= \frac{\sqrt{3}}{2\sqrt{2}} (e_i - e_j) \\
  a_{ij} &= \frac{\sqrt{3}}{2} (e_i + e_j) \\
  b_{ij} &= c_{ij} \times a_{ij}.
\end{align*}
\]

In the bond frame, we consider the general anisotropic exchange Hamiltonian, written in terms of the full angular momenta \( \mathbf{J} \) and where the last term is the antisymmetric Dzyaloshinskii–Moriya exchange (the convention used here is that an antiferromagnetic exchange integral is negative):
\[
\mathcal{H} = -\sum_{\langle ij \rangle} \mathcal{J}_a (\mathbf{J}_i \cdot \mathbf{a}_{ij} \cdot \mathbf{J}_j) + \mathcal{J}_b (\mathbf{J}_i \cdot \mathbf{b}_{ij} \cdot \mathbf{J}_j) + \mathcal{J}_c (\mathbf{J}_i \cdot \mathbf{c}_{ij} \cdot \mathbf{J}_j) + \sqrt{2} J_4 \mathbf{b}_{ij} \cdot \mathbf{J}_i \times \mathbf{J}_j \right) \right].
\]

where \( \sum_{\langle ij \rangle} \) means a summation over the first neighbour pairs. This can be written in terms of a global exchange matrix \( \mathbf{J} \):
\[
\mathcal{H} = -\sum_{\langle ij \rangle, uv} J_{ij}^{uv} \left[ \mathcal{J}_a a_{ij}^u a_{ij}^v + \mathcal{J}_b b_{ij}^u b_{ij}^v + \mathcal{J}_c c_{ij}^u c_{ij}^v \right]
\]

where \( \eta \) is an antisymmetric 3 \( \times \) 3 matrix with only two non-zero elements: \( \eta_{13} = 1 \) and \( \eta_{31} = -1 \). In the ‘bond frame’, the matrix \( \mathbf{J} \) reads:
\[
\mathbf{J} = \begin{pmatrix}
  J_a & 0 & \sqrt{2} J_4 \\
  0 & J_b & 0 \\
  -\sqrt{2} J_4 & 0 & J_c
\end{pmatrix}
\]

In [14], the general anisotropic exchange Hamiltonian is written in terms of the spin components within the local frame of an \( S = 1/2 \) pseudo-spin:
\[
\mathcal{H} = \sum_{ij} J_\pm \left( S_i^+ S_j^- + S_j^+ S_i^- \right) + J_\pm \left( \gamma \eta_n^i S_i^+ S_j^- + \gamma \eta_n^j S_j^+ S_i^- \right) + J_\pm \left( \xi \eta_\pi^i S_i^+ S_j^- + \xi \eta_\pi^j S_j^+ S_i^- \right) + i \leftrightarrow j,
\]

where the ‘sanserif’ notations refer to spin components in the local bases. The two states of this \( S = 1/2 \) pseudo-spin span the states of the ground CEF doublet and, for the case of a Kramers ion (like \( \text{Er}^{3+} \)), one can define a local g-tensor \( \tilde{g} \) such that: \( g_{ij} \mathbf{J} = \tilde{g} \mathbf{S} \). In the context of pyroclores with local trigonal symmetry, the \( \tilde{g} \) matrix is diagonal and takes the form:
\[
\tilde{g} = \begin{pmatrix}
g_+ & 0 & 0 \\
0 & g_+ & 0 \\
0 & 0 & g_z
\end{pmatrix}
\]

Our goal is to determine the relation between the ‘bond frame’ exchange parameters \( \mathcal{J}_a, \mathcal{J}_b, \mathcal{J}_c \) and \( J_\pm \), on the one hand, and the pseudo-spin parameters \( J_{z\pm}, J_{\pm\pm}, J_{\pm} \) and \( J_{\pm\pm} \) on the other hand. To this end, we call \( B_{ij} \) (resp. \( M_{ij} \)) the matrix transforming the coordinates in the ‘bond frame’ \( \{ij\} \) (resp. the local frame \( i \)) to the cubic (Cartesian) coordinates, \( R \) the rotation matrix of \(-\pi/3\) around a local axis, and \( A \) the matrix transforming \((S_z, S_y, S_x)\) into \((S_+, S_-, S_0)\) in the local basis (we omitted the site indices for sake of clarity): \( g_{ij} \mathbf{J} = \tilde{g} \mathbf{S} \), with \( \tilde{S} = (S_+, S_-, S_0) \), and:
\[
A = \begin{pmatrix}
  1/2 & 1/2 & 0 \\
-1/2 & i/2 & 0 \\
  0   &  0    &  1
\end{pmatrix}.
\]

A straightforward transformation of Hamiltonian (A.3) shows that:
\[
\mathcal{H} = \sum_{\langle ij \rangle, uv} \epsilon_{ij}^{uv} \left[ \frac{1}{\tilde{g}_{ij}} A^T \tilde{g} R^T M_i^u B_{ij} \tilde{J} B_{ij}^0 M_j^v R \tilde{g} A \right] S_i^v,
\]

This allows the relations between the two sets of parameters appearing in (A.4) and (A.5) to be determined; they are given in section 2 of the main text. The conventions as to the sign of the exchange integrals are different for the two sets: for an AF interaction, they are positive (except \( J_{\pm\pi} \)) in the pseudo-spin representation and negative in the ‘bond frame’ description.

We note that, in the ‘bond frame’, the dipolar interaction limited to a nearest neighbour pair \( \{ij\} : \mathcal{H}_{\text{dip}} = D [\mathbf{J}_i \mathbf{J}_j - 3 (\mathbf{J}_i \cdot \mathbf{c}_{ij} \cdot \mathbf{J}_j)] = \mathbf{J}_i \cdot \mathbf{D} \cdot \mathbf{J}_j \) is diagonal with:
\[
\mathbf{D} = \begin{pmatrix}
  D & 0 & 0 \\
  0 & D & 0 \\
  0 & 0 & -2D
\end{pmatrix}
\]

and \( D = \frac{a}{4 \pi} \left( \frac{\mu_0 a}{4 \pi} \right)^2 \frac{13 \pi^2}{16} \), where \( a \) is the cubic cell lattice parameter.

References

[1] Gardner J S, Gingras M J P and Greedan J E 2010 Rev. Mod. Phys. 82 53
[2] Ramirez A P, Hayashi A, Cava R J, Siddhartan R and Shastry B S 1999 Nature 399 333
[3] Bramwell S T et al. 2001 Phys. Rev. Lett. 87 047205
[4] Cao H B, Gukasov A, Mirebeau I, Bonville P, Decorse C and Sorge A and Gingras M J P 2011 Phys. Rev. Lett. 106 187202
[5] Thompson J D, McClarty P A, Rønnow H M, Regnault L P, Sorge A and Gingras M J P 2011 Phys. Rev. Lett. 106 187202
[6] Onoda S and Tanaka Y 2011 Phys. Rev. B 83 094411
[7] Blöte H W J, Wielenia R F and Huiskamp W J 1969 Physica 43 549
[8] Siddhartan R, Shastry B S, Ramirez A P, Hayashi A, Cava R J and Rosenkranz S 1999 Phys. Rev. Lett. 83 1854
[9] Champion J D M et al. 2003 Phys. Rev. B 68 020401(R)
[10] Poole A, Wills A S and Lelièvre-Berna E 2007 J. Phys.: Condens. Matter 19 452201
[11] Villain J 1980 J. Physique 41 1263
[12] McClarty P A, Curnoe S H and Gingras M J P 2009 J. Phys.: Conf. Ser. 145 012032
[13] Zhitomirsky M E, Gvozdikova M V, Holdsworth P C W and Moessner R 2012 Phys. Rev. Lett. 109 077204
[14] Savary L, Ross K A, Gaulin B D, Ruff J P C and Balents L 2012 Phys. Rev. Lett. 109 167201
[15] Ruff J P C et al 2008 Phys. Rev. Lett. 101 147205
[16] Cao H B, Mirebeau I, Gukasov A, Bonville P and Decorse C 2010 Phys. Rev. B 82 104431
[17] Ross K A, Savary L, Gaulin B D and Balents L 2011 Phys. Rev. X 1 021002
[18] Malkin B Z, Lummen T T A, van Loosdrecht P H M, Dhalenne G and Zakirov A R 2010 J. Phys.: Condens. Matter 22 276003
[19] Born M and Huang K 1954 Dynamical Theory of Crystal Lattices (London: Oxford University Press)
[20] Toukmaji A, Sagui C, Board J and Darden T 2000 J. Chem. Phys. 113 10913
[21] Petrenko O A, Lees M R and Balakrishnan G 2011 J. Phys.: Condens. Matter 23 164218
[22] Sosin S S, Prozorova L A, Lees M R, Balakrishnan G and Petrenko O A 2010 Phys. Rev. B 82 094428
[23] Dalmas de Réotier P, Yaouanc A, Chapuis Y, Curnoe S H, Grenier B, Ressouche E, Marin C, Lago J, Baines C and Giblin S R 2012 Phys. Rev. B 86 104424
[24] Kao Y J, Enjalran M, Del Maestro A, Molavian H R and Gingras M J P 2003 Phys. Rev. B 68 172407
[25] Del Maestro A G and Gingras M J P 2004 J. Phys.: Condens. Matter 16 3339
[26] Enjalran M and Gingras M J P 2004 Phys. Rev. B 70 174426