Local susceptibility of the Yb$_2$Ti$_2$O$_7$ rare earth pyrochlore computed from a Hamiltonian with anisotropic exchange

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Abstract

The rare earth pyrochlore magnet Yb$_2$Ti$_2$O$_7$ is among a handful of materials that apparently exhibit no long range order down to the lowest explored temperatures and well below the Curie–Weiss temperature. Paramagnetic neutron scattering on a single crystal sample has revealed the presence of anisotropic correlations and recent work has led to the proposal of a detailed microscopic Hamiltonian for this material involving significantly anisotropic exchange. In this paper, we compute the local sublattice susceptibility of Yb$_2$Ti$_2$O$_7$ from the proposed model and compare with the measurements of Cao et al (2009 Phys. Rev. Lett. 103 056402), finding quite good agreement. In contrast, a model with only isotropic exchange and long range magnetostatic dipolar interactions gives rise to a local susceptibility that is inconsistent with the data.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The pyrochlore lattice of corner-sharing tetrahedra is a classic example of a lattice prone to geometric frustration, the inability of interacting magnetic moments on such a lattice to simultaneously minimize all of the pairwise interactions in the system. In the rare earth oxides, $A_2B_2O_7$, where $A$ is a trivalent rare earth ion (Ho, Dy, Tb, Gd, Yb) or yttrium (Y) and $B$ is a tetravalent transition metal ion (Ti, Sn, Mo, Mn), both $A$ and $B$ reside on distinct interpenetrating pyrochlore lattices. Such systems have been the subject of intense research for the past ten years as they are known to manifest a wide variety of exciting collective phenomena [1] including spin ices (Dy$_2$Ti$_2$O$_7$, Ho$_2$Ti$_2$O$_7$) [2], spin liquids (Tb$_2$Ti$_2$O$_7$) [3], spin glasses (Y$_2$Mo$_2$O$_7$) [4], and LRO with persistent spin dynamics (Gd$_2$Sn$_2$O$_7$, Er$_2$Ti$_2$O$_7$, and Yb$_2$Ti$_2$O$_7$) [5–7].

To better understand the many complex phenomena observed in magnetic pyrochlore oxides, knowledge of the microscopic interactions present in these materials is necessary. The pyrochlore lattice can be described as an fcc Bravais lattice with a tetrahedral basis at each fcc site. The magnetic rare earth ($A^{3+}$) ions reside on the pyrochlore lattice and are subject to a crystal field (CF), which introduces a magnetic anisotropy with respect to the local $\langle 111 \rangle$ axes that is reflected in the magnetic susceptibility on each of the four sublattices. Such systems have been the subject of intense research for the past ten years as they are known to manifest a wide variety of exciting collective phenomena [1] including spin ices (Dy$_2$Ti$_2$O$_7$, Ho$_2$Ti$_2$O$_7$) [2], spin liquids (Tb$_2$Ti$_2$O$_7$) [3], spin glasses (Y$_2$Mo$_2$O$_7$) [4], and LRO with persistent spin dynamics (Gd$_2$Sn$_2$O$_7$, Er$_2$Ti$_2$O$_7$, and Yb$_2$Ti$_2$O$_7$) [5–7].

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vehicle for the richness of phenomena observed in rare earth frustrated magnets [1]. Indeed, recent theoretical work has exposed large anisotropic superexchange in pyrochlore oxide materials [10, 11].

In this paper, we focus on local susceptibility measurements on one material, Yb$_2$Ti$_2$O$_7$ [8], which is remarkable for exhibiting a phase transition at $T_c \approx 240$ mK, albeit with no signs of long range magnetic order at lower temperatures [12]. The nature of the low temperature phase in Yb$_2$Ti$_2$O$_7$ is currently somewhat controversial. Yasui et al [13] report the onset of hysteresis in the vicinity of 240 mK and the presence of magnetic Bragg peaks at lower temperatures. Other studies, however, have found no evidence of magnetic reflections [12, 14, 15] in zero applied field. Hodges et al [12] present muon spin relaxation data revealing that the spin fluctuation rate drops sharply at about 240 mK, but remains nonzero and temperature independent well below $T_c$, perhaps indicating a spin gas to spin liquid type phase transition. Particularly, the collective paramagnetic state exhibits strongly anisotropic rod-like correlations in the diffuse neutron scattering which have been conjectured to arise from a weak decoupling of the kagome layers in the pyrochlore lattice [15]. We have investigated the origin of the correlations seen in neutron scattering by using a random phase approximation calculation of the paramagnetic neutron scattering intensity pattern and found a set of interactions that produce a theoretical scattering pattern that is in very good agreement with the experimental data [16]. The foremost characteristic of these interactions is that they are strongly anisotropic. The purposes of the present work are (i) to provide a comparison of the predictions of our microscopic Hamiltonian [16] with the local susceptibility ($\chi_a$) data of [8] and (ii) to provide a discussion of the sensitivity of $\chi_a$ to the nature of the interactions and compare with the results of [8, 9]. We show that our model can successfully account for the observed temperature dependence of $\chi_a$ without further adjustment to the exchange couplings determined in [16].

Figure 1. (a) Tetrahedral basis of the pyrochlore structure showing the four anisotropy axes as red arrows and the applied magnetic field $\mathbf{B}$ orientation (lower orange arrow). Sublattices 1 and 2 make up the $\alpha$ chains and sublattices 3 and 4, the $\beta$ chains. (b) Local susceptibility (open symbols) as reported in [8] in a 1 T field along [110], along with single ion local susceptibilities computed using the CFs of [8] (dashed and solid lines) and [17] (dot–dash line—the perpendicular part having been omitted as it almost coincides with the [8] CF result) in the same field, but without including the $J_e$ exchange interactions.

2. Anisotropic exchange model

In this section, we provide a description of a candidate theory for the magnetism of Yb$_2$Ti$_2$O$_7$ introduced in [16]. The magnetic Yb$^{3+}$ ion has electronic configuration $^4$I$_g$, such that $J = 7/2$ and Landé factor $g_J = 8/7$. The nearest neighbour distance between Yb$^{3+}$ ions is $r_{nn} = (a/4)\sqrt{2}$, where $a = 10.026$ Å is the size of the conventional cubic unit cell [14]. The single ion CF interaction $H_{ci}$ is the largest magnetic energy scale. The form of $H_{ci}$ is fixed by the symmetry of the Yb$^{3+}$ environment. To specify $H_{ci}$ completely, we require the values of the six CF parameters. We use two independent sets of CF parameters determined in [8, 17]. The magnetic Hamiltonian is $H = H_{ci} + H_{int}$, which includes both the CF and spin–spin interactions of the form $H_{int} = H_{ex} + H_{pd}$, which is composed of exchange, $H_{ex}$, and long range magnetostatic dipolar interactions $H_{pd} = \frac{1}{2} \sum_{i,a,j,b} \frac{D_{int}(a,b)}{|\mathbf{r}_{ij}|^6} (\mathbf{J}_i \cdot \mathbf{J}_j - 3(\mathbf{J}_i \cdot \mathbf{n}_{ij})\mathbf{n}_{ij} (\mathbf{J}_j \cdot \mathbf{n}_{ij}))$ with coupling $D = \frac{4\mu_0 \mu_n n_{ij}^2}{6\pi a^6}$, $H_{ex}$ contains all nearest neighbour exchange interactions, $J_{ex}$, that respect lattice symmetries. There are four such nearest neighbour interactions [18]: $H_{iso} = -J_{iso} \sum_{i,a,j,b} (\mathbf{J}_i \cdot \mathbf{n}_{ij})(\mathbf{J}_j \cdot \mathbf{n}_{ij})$, the standard isotropic exchange, $H_{pd} = -J_{pd} \sum_{i,j} (\mathbf{J}_i \cdot \mathbf{J}_j - 3(\mathbf{J}_i \cdot \mathbf{n}_{ij})\mathbf{n}_{ij} (\mathbf{J}_j \cdot \mathbf{n}_{ij}))$, a pseudo-dipolar interaction of exchange origin and not part of $H_{pd}$ and, finally, $H_{DM} = -\sum_{i,a,j,b} \Omega_{DM}^{a,b} (\mathbf{J}_i \times \mathbf{J}_j)$, the Dzyaloshinskii–Moriya (DM) interaction [19]. In all of these terms, $J_{ex}$ denotes the angular momentum of the Yb$^{3+}$ located at lattice $\mathbf{R}_i$ (fcc lattice site $i$, and tetrahedral sublattice site $a$) and $\mathbf{n}_{ij}$ is a unit vector directed along $\mathbf{R}_{ij} - \mathbf{R}_i$. In our recent work [16], we were able to determine the values of $J_e = \{J_{iso}, J_{iso}, J_{pd}, J_{DM}\}$. We accomplished this by fitting the diffuse paramagnetic neutron scattering measurements to neutron scattering patterns generated using the random phase approximation [16]. The fitting was...
shown that large DM interactions are allowed in Yb$_2$Ti$_2$O$_7$ due to neutron polarization vector $\vec{R}$. Yb$_2$Ti$_2$O$_7$, the applied magnetic field was 1 T in the [110] direction with all diagonal elements equal and all off-diagonal elements (in fcc sites) was taken to have two independent components $\chi_{\alpha\beta}$, sublattice labelling as well as the local Ising directions. Transforming $\chi_a$ to the local frame $\chi_a^{\nu} = \hat{u}_a \cdot \vec{u}_\nu \chi_a^{\nu}$, we extract $\chi_a$ with $\chi_a^{\nu} = \chi_a^{zz}$. Our choice of local frame is such that the [110] field has vanishing local y component for sublattices 1 and 2 (along the $\alpha$ and $\beta$ chains) and vanishing local x and z components for sublattices 3 and 4 (along the $\beta$ chains). Hence, for sublattices 1 and 2, with an applied [110] field, we take $\chi_{a,\perp} = \chi^{1x}$ and for sublattices 3 and 4, $\chi_{a,\perp} = \chi^{yy}$.

### 4. Results

Figure 1(b) shows the single ion $\chi_a = \{\chi_{||}, \chi_{\perp}\}$, computed only from the Zeeman term and CF Hamiltonian using the CF parameters of [8, 17], and the experimentally determined values of $\chi_a$ [8]. Both sets of CF parameters give rise to a $\chi_{\perp}$ that fits the experimental data well over the whole temperature range even in the absence of interactions except for the lowest temperature point, $T = 2$ K, where the experimental $\chi_{\perp}$ falls below the CF prediction. However, the $\chi_{||}$ calculations for the two sets of CF parameters are significantly different from one another and from the experiment. Whereas, the

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4 The measurements that have been made of the Curie–Weiss temperature $T_C$ all lie in the range $0.65 \pm 0.15$ K.
parameters of [8] give a $\chi_1$ that fits reasonably well between 20 and 70 K, deviating from experiment at the highest and lowest measured temperatures, those of [17] do not fit well over most of the temperature range. The addition of anisotropic exchange of the form given in equation (1), does not improve the fit to experiment of $\chi_1$ computed using the CF parameters of [17]. Because of the poor fit to $\chi_1$, even when anisotropic exchange is included, we do not use the CF parameters of [17] any further in this work despite their fair success in reproducing paramagnetic neutron scattering measurements in [16]. Another point to be drawn from figure 1(b) is that there is a difference between the local susceptibilities for the $\alpha$ and $\beta$ chains. In particular, $\chi_1$ is not probed for the 3 and 4 sublattices because the [110] field does not induce a moment in the local [111] Ising direction on these sublattices. Also, $\chi_\perp$, which is probed on all four sublattices, is different for the $\alpha$ and $\beta$ chains. As the temperature is raised, the cubic symmetry between different sublattices is partially restored.

When interactions are included in the computation of $\chi_a$ as described in section 3, we obtain the results shown in figure 2. Figure 2(a) shows the local susceptibility, computed for two different models over the temperature range 2–300 K, together with the experimental data of [8]. The inclusion of interactions does not significantly affect $\chi_\perp$ so, for these results, we merely observe that the splitting between $\alpha$ and $\beta$ chains is present once again because of the symmetry breaking field. The first model to be compared with the experimental $\chi_\parallel$ data is one with long ranged dipoles and purely isotropic exchange ($J_{sing} = J_{pol} = J_{DM} = 0$), with a coupling $J_{iso} = -0.06 K$ constrained by the Curie–Weiss temperature, which is taken to be $\theta_{CW} = 0.75 K$, within the experimental error margins given above [16] (blue dot–dash line). The second model is the candidate anisotropic exchange model described in section 2 and [16] (blue dashed line). The $\chi_\perp$ results (red lines) do not strongly distinguish between the two models. At low temperatures, $T \lesssim 30 K$, where the effect of interactions produces significant deviations of $\chi_1$ from the noninteracting case, the isotropic exchange model does not fit the experimental data as well as the anisotropic exchange model, except at the lowest temperature point (2 K). However, neither model captures the 2 K point for $\chi_\perp$.

Figure 2(b) shows once again the experimental data and the anisotropic model predictions for $\chi_a$ as well as $\chi_\alpha$ computed using the molecular field method of [8]. The fit in [8] of $\chi_\parallel$ is only provided down to approximately 3.5 K in that work (and no results for $\chi_\perp$ for the single ion MF case are reported in that reference). For the temperature range over which the models can be compared, our anisotropic exchange model describes the experimental data as well as the model of [8]. However, our model achieves this while correctly preserving the lattice symmetries, unlike the model of [8], and therefore corresponds to a physically acceptable microscopic model. We discuss further the comparison of our results with those of [8, 9] in the appendix.

At temperatures $T \gg \theta_{CW}$, interactions are expected to have little effect on $\chi_a$ which is borne out by our calculations. Therefore the differences between the $\chi_a$ calculations and experimental results at high temperatures ($T > 100 K$) can be attributed to an incorrect spectral decomposition of the excited CF states [8, 17].

5. Discussion

We have investigated the local susceptibility computed from the anisotropic exchange model described in section 2 for Yb$_2$Ti$_2$O$_7$. The model was obtained from an independent previous fit to the paramagnetic neutron scattering [16]. The calculations presented here fit the $\chi_a$ experimental data well with no adjustable parameters over much of the experimental temperature range 2–250 K. There are, however, discrepancies in the fit above approximately 100 K, presumably due to the fact that the available CF parameters do not capture the correct wavefunctions for the excited CF energy levels. The fit is also
imperfect at the lowest observed temperature of 2 K where the experimental $\chi_a$ susceptibility flattens out while it does not in the MF treatment of the Hamiltonian $H = H_{\text{zz}} + H_{\text{int}} + H_{\text{Z}}$.

At high temperatures $T \gg 6\text{K}$, $\chi_a$ is insensitive to the interactions and the quality of fit is determined by the CF Hamiltonian and the Zeeman term. Figure 1 illustrates that the CF provides an adequate fit to $\chi_a$ from 100 K down to about 30 K and to $\chi_\perp$ over the entire range of temperatures. Figure 2 shows that the quality of fit for $T > 30$ K does not change with the inclusion of interactions. Therefore, the only nontrivial test of our model comes from the five lowest temperature points ($T = 2, 5, 10, 15, 20$ K) in $\chi_\parallel$ where the noninteracting Hamiltonian, $H = H_{\text{zz}} + H_{\text{Z}}$, gives a $\chi_\parallel$ that falls significantly below the experimental result (see figure 1(b)). The anisotropic exchange model determined in [16] provides a good description of $\chi_\parallel$, whereas isotropic exchange interactions do not—this is the main conclusion of this work. Although the relevant test of our model comes from only a few (five) data points, it achieves the goals of providing a consistency check of our model determined previously on the basis of diffuse paramagnetic neutron scattering [16] while also establishing that anisotropy in the exchange is indeed an intrinsic ingredient of the spin Hamiltonian of Yb$_2$Ti$_2$O$_7$.

The authors of [8, 9] both extract exchange parameters from the local susceptibility data, and present fits to the local susceptibility that result from these exchange parameters. As stated above, we find that this method for extracting exchange parameters is underconstrained. In the appendix, we describe in some detail the approaches taken in these two papers and compare with our own results.

6. Conclusion

In this work, we have shown that a model with four non-adjustable bilinear nearest neighbor exchange couplings, extracted from neutron scattering measurements in a previous work [16], produces a good fit to local susceptibility measurements on the rare earth pyrochlore material Yb$_2$Ti$_2$O$_7$ for temperatures in the range $T \lesssim 100$ K. The fit of our model to the experimental measurements of $\chi_a$ provides further independent evidence for the correctness of our anisotropic exchange model [16].

We also found that the origin of the discrepancy between the experiment and our model at high temperatures, and indirectly, a similar failure to fit the local susceptibility $\chi_a$ also at $T \gtrsim 10^2$ K in [8], lies in the excited CF states obtained from the CF parameters of [8, 17]. Further experimental work would be required to probe more accurately the spectral decomposition of the excited states of the CF in order to better determine their structure. That said, we do not expect that the corrections to the spectral decomposition of those excited states would, as a byproduct, necessitate revisiting the values of the anisotropic exchange parameters $\mathcal{J}_a$. We are thus confident, combining the results presented in this paper and those of [16], that we have in hand the correct microscopic Hamiltonian of Yb$_2$Ti$_2$O$_7$ that includes symmetry-allowed anisotropic bilinear nearest neighbour couplings. One may therefore aim to proceed to unravel the nature of the low temperature ($T \lesssim 2$ K) state of this exotic material. In this context, it is worth commenting on the validity of the mean field theory treatment of our model at low temperatures ($T \lesssim 2$ K) and/or in weak field. Mean field theory predicts a transition to a $q = 0$ structure at approximately 1.1 K [16]. This contrasts with the experimental $T_c = 240$ mK [12]. As usual, mean field calculations neglect the effects of both thermal and quantum fluctuations. These could be fairly large in this system and greatly decrease the critical temperature. The role of thermal and quantum fluctuations in the proposed Hamiltonian $H = H_{\text{int}} + H_{\text{Z}}$ for Yb$_2$Ti$_2$O$_7$ remains to be investigated. To the best of our knowledge, and notwithstanding the difficulties with mean field theory, the sharp specific heat feature signalling a phase transition in Yb$_2$Ti$_2$O$_7$ has only been reported in powder samples. It would be useful to confirm the existence of this feature in single crystals.

Two interesting observations that arise from this work are that (i) the isotropic exchange $J_{\text{iso}}$ is ferromagnetic (positive) in Yb$_2$Ti$_2$O$_7$ while it is found to be antiferromagnetic (negative) for the other A$_2$Ti$_2$O$_7$ compounds [1] and (ii) there appears to exist a large antisymmetric anisotropic exchange described above in terms of the form of a DM interaction. Very recent theoretical work [11] has found that a large anisotropic exchange, including large DM interactions, can in principle arise in Yb$_2$Ti$_2$O$_7$. It would be interesting to investigate this issue further.

The conclusion of this work, that the magnetic exchange interactions in Yb$_2$Ti$_2$O$_7$ are anisotropic, hints at the possibility of significant multipolar interactions [10, 24] in this material and, perhaps, in other rare earth pyrochlores. That said, since the CF gap is much larger in Yb$_2$Ti$_2$O$_7$ than the scale of the interactions, anisotropic bilinear couplings between the J operators as well as multipolar couplings between the Yb$^{3+}$ moments manifest themselves, at low energies, in the form of generally anisotropic bilinear exchange couplings between effective pseudospins one-half [11].

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Appendix. Previous works on local susceptibility in Yb$_2$Ti$_2$O$_7$

In this appendix we discuss the anisotropic exchange fits of [8], for Yb$_2$Ti$_2$O$_7$, and [9] for various rare earth pyrochlore titanates.

The fit to $\chi_a$ presented in [8] and reproduced in figure 2 is based on a self-consistent mean field theory that considers a single sublattice. The moment ($\mathbf{J}$), at a temperature $T$, was computed from a mean field Hamiltonian consisting of the CF, a Zeeman term, and a mean field term arising from the interactions of the form $-g_{\mu B} \lambda_a^\alpha (J_a^\alpha)$, where $\alpha$ specifies the components $||$ and $\perp$, the directions parallel and perpendicular to the local [111] direction. Solving for the moment, the two couplings $\lambda_||$ and $\lambda_\perp$ were determined from a fit to $\chi_a$. Since
this model fails to take into account the sublattice structure of the pyrochlore lattice, the ensuing fit is unlikely to correctly reflect the underlying microscopic couplings in Yb$_2$Ti$_2$O$_7$.

The model of [9] includes the CF anisotropy as well as nearest neighbour exchange couplings and the long range dipole interaction (treated via an Ewald summation). The model correctly handles the sublattice structure of the pyrochlore lattice. The model includes, at the outset, three exchange couplings which respect the lattice symmetries and which are denoted: $\lambda_{\perp,1}^M$, $\lambda_{\perp,2}^M$ and $\lambda_{\parallel}^M$. These can be expressed in terms of the couplings $\mathcal{J}_c$ as follows:

$$\lambda_{\perp,1}^M = \frac{1}{2} \mathcal{J}_{\text{sing}} + \mathcal{J}_{\text{iso}} + \mathcal{J}_{\text{pd}} \quad (A.1)$$

$$\lambda_{\perp,2}^M = \mathcal{J}_{\text{iso}} + \frac{1}{2} \mathcal{J}_{\text{pd}} \quad (A.2)$$

$$\lambda_{\parallel}^M = -\frac{2}{3} \mathcal{J}_{\text{sing}} + \mathcal{J}_{\text{iso}} - 2 \mathcal{J}_{\text{pd}} \quad (A.3)$$

This set of couplings does not include the DM coupling, $\mathcal{J}_{\text{DM}}$. Furthermore, in the fits to $\chi_a$, [9] imposes the constraint $\lambda_{\perp,1}^M = \lambda_{\perp,2}^M$. While this choice most likely circumvents the problem of having an underconstrained set of couplings to fit the $\chi_a$ data, the reduction of the parameter space to a particular two-dimensional surface within a four-dimensional space of couplings appears to us to be somewhat arbitrary. Indeed, the couplings we have obtained from neutron scattering data and which we used in our fit to $\chi_a$ do not lie within the parameter space considered in [9]. For example, $\mathcal{J}_{\text{DM}} = -0.25$ K for our model with CF parameters taken from [8] (see equation (2)), which is of the same order of magnitude as the other three couplings. Malkin et al [9] make use of their model to study the exchange couplings of a number of rare earth pyrochlore titanates finding a significant degree of anisotropy in the exchange in a number of cases. For the case of Yb$_2$Ti$_2$O$_7$, which is relevant to this paper, they mention couplings that are much larger than those of the other materials considered but do not report these couplings.

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