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2009 J. Phys.: Condens. Matter 21 492202

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Anisotropic exchange in frustrated pyrochlore $\text{Yb}_2\text{Ti}_2\text{O}_7$

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Received 29 September 2009, in final form 5 November 2009

Published 19 November 2009

Online at stacks.iop.org/JPhysCM/21/492202**Abstract**

The local Yb^{3+} magnetic susceptibility tensor was recently measured in the frustrated pyrochlore compound $\text{Yb}_2\text{Ti}_2\text{O}_7$ by means of in-field polarized neutron scattering in a single crystal. A very anisotropic effective exchange tensor was derived for the Yb^{3+} ion. Using this result, we reinterpret here the data for the powder susceptibility in $\text{Yb}_2\text{Ti}_2\text{O}_7$. We show that, in the case of a well-isolated Kramers doublet with anisotropic g and exchange tensors, the inverse susceptibility for a powder sample does not strictly obey a Curie–Weiss law at low temperature. We discuss the consequences regarding the paramagnetic Curie temperature, usually taken as a measure of the exchange/dipolar interaction, and the exotic ‘slow fluctuation’ ground state of $\text{Yb}_2\text{Ti}_2\text{O}_7$.

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

1. Introduction

The pyrochlore lattice is a cubic lattice (space group $Fd\bar{3}m$) made of regular tetrahedra linked by their vertices, a unit cell containing four tetrahedra whose threefold axes are along the $\langle 111 \rangle$ cubic directions. In the materials with formula $\text{R}_2\text{M}_2\text{O}_7$, where R is a rare earth and M a 4d transition metal (or Sn), both R^{3+} and M^{4+} ions occupy a pyrochlore lattice, displaced from each other by a translation along the cubic diagonal $[111]$. The pyrochlore materials have been at the centre of renewed interest for a decade because their lattice is prone to geometrical frustration of the exchange interactions between magnetic moments [1]. For instance, in the presence of isotropic (Heisenberg) antiferromagnetic (AF) exchange, it is clear that the four magnetic moments sitting at the corners of a single tetrahedron cannot adopt a collinear Néel-like AF arrangement such that all AF pairwise interactions be minimized. As a general rule, geometric frustration prevents long range magnetic order (LRO) settling at low temperature, giving rise, in the titanate ($\text{M} = \text{Ti}$) pyrochlore family, to the

so-called spin ice ground state in $\text{Ho}_2\text{Ti}_2\text{O}_7$ and $\text{Dy}_2\text{Ti}_2\text{O}_7$ [2] and to a spin-liquid ground state in $\text{Tb}_2\text{Ti}_2\text{O}_7$ [3]. In $\text{Yb}_2\text{Ti}_2\text{O}_7$, a transition takes place at 0.25 K, but the ground state is not a magnetic LRO phase. It shows slowly fluctuating Yb^{3+} magnetic moments with short range correlations [4, 6]. A polarized neutron study [7] confirmed this fluctuating picture, contradicting a neutron diffraction study [8] where small Bragg peaks had been observed. It seems now well established that the ground state in $\text{Yb}_2\text{Ti}_2\text{O}_7$ is dynamic, with short range two-dimensional spin correlations present both below and above the transition [5, 6, 9].

The pyrochlore lattice possesses the property that an external field, for instance an applied magnetic field, cannot be parallel simultaneously to the four local symmetry axes of a tetrahedron in a single crystal. This results in the impossibility of measuring the local magnetic properties, i.e. the susceptibility tensor, using classical bulk magnetometry methods. Indeed, the obtained data are always an average over the four local directions. In a recent work, we presented a method that circumvents this drawback: in-field polarized neutron diffraction in a single-crystal sample [10]. The applied field makes a different angle with the four ternary axes of

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the rare earth ions in a given tetrahedron. The resulting induced moments can be described, in the linear approximation holding for low fields, using a single two-component local susceptibility tensor $\{\chi_z, \chi_\perp\}$ appropriate for the threefold symmetry of the Yb^{3+} site: $\mathbf{m}_i = \tilde{\chi} \mathbf{H}_i$, where the index i labels the four sites. Refinement of the polarized neutron diffraction patterns allows the $\tilde{\chi}$ tensor to be determined [10]. In this latter work, the method was applied to the pyrochlore titanates with $R = \text{Ho, Tb, Er}$ and Yb , whose crystal field (CF) level scheme is relatively well known. In order to interpret the $\tilde{\chi}(T)$ data, we were led to introduce interionic interactions through a molecular field tensor $\tilde{\lambda} = \{\lambda_z, \lambda_\perp\}$ (such that the molecular field on ion i is $\mathbf{H}_{\text{mol}} = \tilde{\lambda} \mathbf{m}_i$), which turned out to be quite anisotropic in most cases.

In this work, we focus on $\text{Yb}_2\text{Ti}_2\text{O}_7$ at low temperature, when the ground CF doublet alone is populated. We show that, in the presence of anisotropy due to both crystal field and interionic couplings, the thermal behaviour of the inverse susceptibility of a powder sample cannot be described by a linear Curie–Weiss law. We also show that the paramagnetic Curie temperature measured by the usual methods either in a powder or single-crystal sample may yield unreliable results when interpreted classically in terms of isotropic interionic interactions.

2. The polarized neutron data in $\text{Yb}_2\text{Ti}_2\text{O}_7$

The susceptibility data derived from the polarized neutron analysis is shown in figure 1. The main thermal variation is due to the progressive population of the Yb^{3+} crystal field Kramers doublets as temperature increases. The parameters of the crystal field interaction in $\text{Yb}_2\text{Ti}_2\text{O}_7$ are given in [10] and the calculated curves for $\chi_z(T)$ (resp. $\chi_\perp(T)$) obtained by including a Zeeman interaction with a field of 1 T parallel (resp. perpendicular) to a local [111] axis are shown in figure 1 as dashed lines. For χ_\perp , the CF/Zeeaman curve reproduces well the experimental data, which is not the case for $\chi_z(T)$ below 20 K. We thus introduced a molecular field tensor $\{\lambda_z, \lambda_\perp\}$ and performed a self-consistent mean-field calculation of the susceptibility in order to improve agreement with the data. For the transverse direction, we obtain $\lambda_\perp \sim 0$, but the axial component is found to be large and positive (ferromagnetic): $\lambda_z = 2.5(5) T/\mu_B$ (solid line in figure 1). When only the ground Kramers doublet $\{|\psi\rangle, |\psi'\rangle\}$ is populated, one can assign it a fictitious spin $S' = 1/2$ and describe its magnetic properties using a g tensor such that: $g_i \mathbf{J} = \tilde{g} \mathbf{S}'$, where $g_J = 6/5$ for Yb^{3+} . Then $g_z = 2g_J \langle \psi | J_z | \psi \rangle$ and $g_\perp = 2g_J \langle \psi | J_x | \psi' \rangle$. In $\text{Yb}_2\text{Ti}_2\text{O}_7$, the excited Kramers doublets lie at approx. 700 K above the ground state [10, 11], which has $g_z = 2.25$ and $g_\perp = 4.1$; so the g and λ tensors have opposite anisotropies in $\text{Yb}_2\text{Ti}_2\text{O}_7$.

3. Calculation of the powder susceptibility in the presence of anisotropy

We consider a Kramers doublet in a site with axial symmetry, and with anisotropic $\{g_i\}$ and $\{\lambda_i\}$ tensors. We assume that the temperature is much lower than the energy gap between

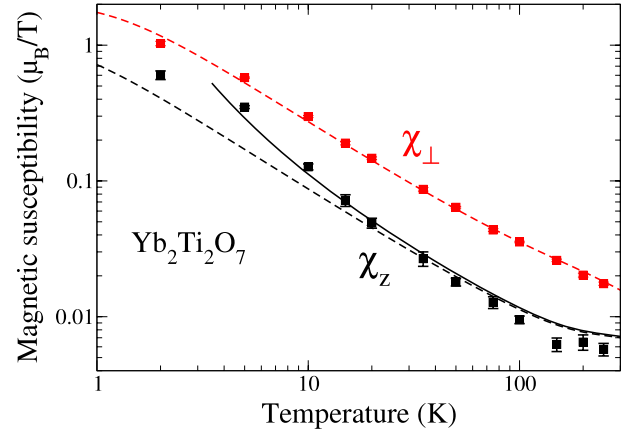


Figure 1. Thermal variation of χ_z and χ_\perp in $\text{Yb}_2\text{Ti}_2\text{O}_7$ (from [10]), together with calculations involving crystal field only (dashed line), and crystal field and anisotropic exchange, treated self-consistently in the mean-field approximation (solid lines). For $\chi_\perp(T)$, the dashed curve alone is shown because the crystal-field-only calculation matches well with the data.

the ground doublet and the excited CF states (≈ 700 K in $\text{Yb}_2\text{Ti}_2\text{O}_7$). Then the magnetic properties of the system are well described in terms of those of the ground doublet alone. In computing the susceptibility, we will neglect the Van Vleck contribution due to the quantum mixing of excited CF states by the Zeeman interaction. For the field in the i direction (i being z or \perp), the Curie constant is $C_i = \frac{g_i^2 \mu_B^2}{4k_B}$ and the susceptibility in the presence of a molecular field is

$$\chi_i = \frac{C_i/T}{1 - \lambda_i C_i/T}. \quad (1)$$

The powder susceptibility $\chi_{\text{pdr}} = \frac{1}{3}(\chi_z + 2\chi_\perp)$ can then be easily obtained as

$$\chi_{\text{pdr}} = \frac{\bar{g}^2 \mu_B^2}{4k_B} \frac{T - \Lambda}{(T - G_z)(T - G_\perp)}, \quad (2)$$

where $\bar{g}^2 = \frac{1}{3}(g_z^2 + 2g_\perp^2)$, $G_i = \lambda_i \frac{g_i^2 \mu_B^2}{4k_B}$ is the (opposite of) the interionic coupling energy in the i direction and $\Lambda = \frac{1}{3} \frac{g_\perp^2 G_\perp + 2g_z^2 G_z}{\bar{g}^2}$. So, due to the crystal field and interionic coupling anisotropies, the inverse susceptibility $1/\chi_{\text{pdr}}$ for an isolated doublet does not follow a linear Curie–Weiss law, but instead has a hyperbolic shape with an oblique asymptote. Expression (2) should hold for $T > T_i = \max(|G_z|, |G_\perp|)$, which is the mean-field ‘instability temperature’ for the uniform susceptibility. At higher temperature, a Curie–Weiss law is recovered with an effective moment $\mu_{\text{eff}} = \frac{\sqrt{3}}{2} \bar{g} \mu_B$ and

$$\theta_p = G_\perp + G_z - \Lambda = \frac{\mu_B^2}{4k_B} \frac{\lambda_z g_z^4 + 2\lambda_\perp g_\perp^4}{3\bar{g}^2}. \quad (3)$$

Hence, care must be exercised in the presence of anisotropies since (i) the determination of θ_p depends strongly on the temperature range because of the curvature of $1/\chi_{\text{pdr}}(T)$ and (ii) no direct information about the sign and magnitude of the interionic interactions can be derived since each λ_i value is weighed by g_i^4 .

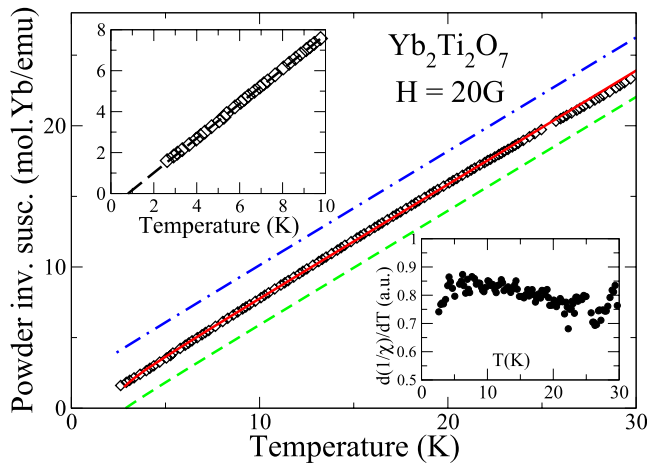


Figure 2. Thermal variation of the inverse powder susceptibility in $\text{Yb}_2\text{Ti}_2\text{O}_7$, measured with a field $H = 20$ G. The curves represent calculations using expression (2) for a doublet with $g_z = 2.3$ and $g_\perp = 4.15$, with various hypotheses about the interionic coupling: isotropic with $\lambda = 1 T/\mu_B$ (dashed green curve) or $-1T/\mu_B$ (dash-dotted blue curve), and anisotropic with $\lambda_z = 2.5 T/\mu_B$, $\lambda_\perp = 0$ (solid red curve). Upper inset: low temperature part of $1/\chi_{\text{pdr}}(T)$ and the Curie–Weiss law with $\theta_p = 0.8$ K (dashed line). Lower inset: derivative of $1/\chi_{\text{pdr}}$ with temperature.

4. Low temperature powder susceptibility in $\text{Yb}_2\text{Ti}_2\text{O}_7$

Early measurements of the powder susceptibility in $\text{Yb}_2\text{Ti}_2\text{O}_7$ below 10 K [11] yielded a small positive value: $\theta_p \simeq 0.75$ K (ferromagnetic). Prompted by our results [10] about the anisotropy of the interactions, we measured $\chi_{\text{pdr}}(T)$ anew in the temperature range 2.5–30 K in a SQUID magnetometer (figure 2, where $1/\chi_{\text{pdr}}$ is represented). The data are very close to a straight line going through zero. However, a slight curvature is observed, illustrated by the thermal variation of $d(1/\chi_{\text{pdr}})/dT$ shown in the lower inset of figure 2. This curvature corresponds to the conclusion made above that $1/\chi_{\text{pdr}}(T)$ has a hyperbolic shape, and it is responsible for the small value $\theta_p \simeq 0.75$ K obtained in [11] when considering data below 10 K (see the upper inset of figure 2). In this figure are represented curves obtained using expression (2), with $g_z = 2.3$ and $g_\perp = 4.15$ appropriate for the ground doublet, and with different hypotheses about the molecular field tensor. The curves with isotropic coupling (dash-dotted blue and dashed green), with $\lambda = \pm 1 T/\mu_B$, do not match the data. The solid red curve is obtained, with no adjustable parameters, using the anisotropic λ tensor $\{\lambda_z = 2.5 T/\mu_B, \lambda_\perp = 0\}$ obtained from an entirely different technique, the polarized neutron data [10]. An almost perfect agreement with experiment is obtained, which gives confidence in the derived λ values. The (opposite of the) interionic coupling energies are then $G_z = 2.15$ K and $G_\perp \simeq 0$, which are the actual interaction strengths. This value of G_z matches that of the peak temperature of the specific heat bump (near 2 K) observed in $\text{Yb}_2\text{Ti}_2\text{O}_7$ in [12]. Indeed, the specific heat is due to the degeneracy lifting by the (short range) exchange/dipolar interaction and one expects $T_{\text{peak}} \sim G_z$.

One can note that, even in the case of isotropic coupling, a linear Curie–Weiss law is not obeyed by $1/\chi(T)$. The deviation is the larger, the more the ratio g_\perp/g_z differs from unity. Strictly speaking, for a ground CF Kramers doublet a linear Curie–Weiss law holds only for isotropic g and molecular field tensors.

5. Discussion

5.1. $\text{Yb}_2\text{Ti}_2\text{O}_7$ an ‘exchange spin ice’?

The crystal field scheme used in [10] for $\text{Yb}_2\text{Ti}_2\text{O}_7$ is rescaled from that for other rare earths (Ho and Tb) and it is somehow different from that used in an earlier work [11]: the values for the CF parameters B_4^0 and B_6^0 are interchanged. With the previous parameter set of [11], the ground Kramers doublet has $g_z = 1.8$, to be compared with $g_z = 2.3$ with the present set. This has some consequence regarding the low temperature slowly fluctuating state. In this phase ($T < 0.25$ K), the magnitude of the Yb^{3+} short range correlated moment is $1.15 \mu_B$ [4]. For an anisotropic Kramers doublet, the saturated moment value is $\frac{1}{2}g_i\mu_B$ when the applied/molecular field is along a principal direction (or plane) i . Using the previous parameters of [11], $g_z = 1.8$ and $g_\perp = 4.2$, the Yb moment could vary between $0.9 \mu_B$ for a field along z and $2.1 \mu_B$ for a field perpendicular to z . The experimental value $1.15 \mu_B$ was then taken as an indication that the moment was tilted (by 44°) from the z axis. With the present parameter set, the moment value is compatible with the Yb moment lying along the local [111] axis. This also matches with the interionic coupling energy balance $G_z \gg G_\perp$, which shows that the two anisotropies combine such that the local z axis is a strongly energetically favoured axis. The origin of this very anisotropic interionic coupling is probably to be searched for in the exchange itself, the pairwise dipole–dipole coupling being rather weak: $|E_{\text{dip}}| \simeq 0.04$ K for a low temperature configuration where the two Yb moments are along their local axes. This is much smaller than the overall interaction $G_z \simeq 2.15$ K. The picture one can now get of the low temperature phase of $\text{Yb}_2\text{Ti}_2\text{O}_7$ is that of short range correlated Yb^{3+} moments slowly fluctuating along their local [111] axis. Rather unexpectedly, the low temperature coupling in $\text{Yb}_2\text{Ti}_2\text{O}_7$ is Ising-like ferromagnetic; the Ising and ferromagnetic characters do not come resp. from the CF and the dipole–dipole interaction, as in the canonical spin ice $\text{Dy}_2\text{Ti}_2\text{O}_7$ [13], but from the exchange alone. Thus, $\text{Yb}_2\text{Ti}_2\text{O}_7$ could be labelled an ‘exchange spin ice’ and it would be of interest to check whether the two-dimensional short range spin correlations observed in $\text{Yb}_2\text{Ti}_2\text{O}_7$ [5, 6, 9] can be explained with such a model.

5.2. Low temperature spin dynamics and distortions in pyrochlores

Another important difference with the standard spin ice lies in the fact that the g tensor has a sizeable transverse value in $\text{Yb}_2\text{Ti}_2\text{O}_7$, allowing fast fluctuations ($\nu \simeq 10$ GHz around 1 K) to occur between the two states of the ground Kramers doublet. Indeed, the occurrence of fast spin fluctuations at

low temperature, due to exchange/dipole-induced two-ion spin flips, is linked to the presence of non-zero matrix elements of J_{\pm} (i.e. g_{\perp}) and/or J_z ($g_{zp} = 2g_J|\langle\psi|J_z|\psi'\rangle|$) between the states of the ground CF multiplet. This multiplet must be a doublet for Kramers ions (Yb^{3+} , Dy^{3+}), and it seems also to be a doublet in all pyrochlores with non-Kramers ions (Pr^{3+} , Tb^{3+} , Ho^{3+}). In the ‘canonical’ spin ices $\text{Ho}_2\text{Ti}_2\text{O}_7$ and $\text{Dy}_2\text{Ti}_2\text{O}_7$, both g_{\perp} and g_{zp} are zero in the ground doublet (Ising ions). This is an intrinsic property for non-Kramers ions in zero field [14], while for Kramers ions only g_{zp} is zero. For Dy^{3+} in $\text{Dy}_2\text{Ti}_2\text{O}_7$, $g_{\perp} = 0$ because the ground wavefunction is $|J = 15/2; J_z = \pm 15/2\rangle$ [15] with possibly a small admixture of the $|J_z = \pm 9/2\rangle$ state [16]. The exchange/dipole coupling can make g_{\perp} depart from zero by mixing with excited CF states and this effect can be large if there is an excited state close to the ground state. In $\text{Ho}_2\text{Ti}_2\text{O}_7$ and $\text{Dy}_2\text{Ti}_2\text{O}_7$, the gap to the excited states is around 200–300 K [16], which is two orders of magnitude larger than the exchange/dipole coupling (a few K). Therefore, g_{\perp} remains vanishingly small and the spin fluctuations are very slow below 10 K [17, 18]. In contrast, there is an excited CF state near 15–20 K in $\text{Tb}_2\text{Ti}_2\text{O}_7$ and in $\text{Tb}_2\text{Sn}_2\text{O}_7$ [19, 20], and the Ising character is lost at low temperature due to the exchange/dipole coupling [21] which can induce a sizeable g_{\perp} value. This is likely to be the cause of the fast fluctuations observed in $\text{Tb}_2\text{Ti}_2\text{O}_7$ [3] and $\text{Tb}_2\text{Sn}_2\text{O}_7$ [22–24] down to very low temperatures.

Recently, a new CF-induced spin ice has seemingly been discovered, namely $\text{Pr}_2\text{Sn}_2\text{O}_7$ [25], which shows very fast spin fluctuations ($\nu \simeq 8$ GHz) below 1 K and has been labelled a ‘dynamic spin ice’. Since the first excited CF level in $\text{Pr}_2\text{Sn}_2\text{O}_7$ lies at 210 K above the ground state [25], the occurrence of fast fluctuations is rather unexpected. For non-Kramers ions, however, a small CF distortion from the nominal threefold symmetry (of magnitude 0.1–1 K) can greatly perturb the magnetic properties. Indeed, it lifts the degeneracy of the ground doublet and the new wavefunctions of the quasi-degenerate singlets $\{\psi_D, \psi'_D\}$ are then the symmetric and antisymmetric combinations of the zero distortion wavefunctions. The following relation holds:

$$g_{zp}^D = 2g_J|\langle\psi_D|J_z|\psi'_D\rangle| = g_z, \quad (4)$$

where g_z is the zero distortion longitudinal component of the g tensor defined above, which can be quite large (20 for Ho^{3+} in $\text{Ho}_2\text{Ti}_2\text{O}_7$, and about 5.3 for Pr^{3+} in $\text{Pr}_2\text{Sn}_2\text{O}_7$). One also has $g_z^D = 0$ and $g_{\perp}^D \simeq 0$, with self-explanatory notations. Although the g_{zp}^D value can be much depleted by an exchange/dipole field, the presence of a distortion opens a new relaxation channel within the lowest levels of a non-Kramers ion. Recently, some evidence was found of a small tetragonal distortion appearing at low temperature in $\text{Tb}_2\text{Ti}_2\text{O}_7$ and $\text{Tb}_2\text{Sn}_2\text{O}_7$ [26, 27]. Assuming that it is also present in other pyrochlores, then the g_{zp}^D channel for relaxation can be effective and can lead to exchange/dipolar-induced fluctuations within a non-Kramers doublet. According to our estimations and assuming that the spin fluctuation frequency ν is proportional to the square of the largest matrix element of J_z or J_{\pm} , the enhancement factor of ν when a distortion is ‘switched on’ is about 50 for $\text{Tb}_2\text{Ti}_2\text{O}_7$ and $\text{Ho}_2\text{Ti}_2\text{O}_7$, but it reaches the very

large value of 10^6 for $\text{Pr}_2\text{Sn}_2\text{O}_7$. In this latter material, the small activation energy $E_A \simeq 1.4$ K found in [25] for the thermal variation of the fluctuation time could represent the splitting of the ground doublet by the distortion.

For Kramers ions, a CF distortion cannot lift the degeneracy of the doublets and its effect on the wavefunctions is very weak.

6. Conclusion

In the frustrated pyrochlore $\text{Yb}_2\text{Ti}_2\text{O}_7$, we examined some consequences of the presence of a very anisotropic molecular field tensor at the Yb^{3+} site, determined in a previous polarized neutron study [10]. We show that the exchange interaction is Ising-like ferromagnetic at low temperature, so that $\text{Yb}_2\text{Ti}_2\text{O}_7$ could be an example of ‘exchange spin ice’. However, in contrast with ‘dipolar spin ices’ like $\text{Dy}_2\text{Ti}_2\text{O}_7$, the CF anisotropy in $\text{Yb}_2\text{Ti}_2\text{O}_7$ allows fast spin fluctuations to occur at low temperature. We also suggest that the spin fluctuations recently observed in the ‘dynamic spin ice’ $\text{Pr}_2\text{Sn}_2\text{O}_7$ can be due to the presence of a CF distortion with respect to trigonal symmetry.

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