Low temperature phase diagram for the pyrochlore compound Tb$_2$Ti$_2$O$_7$

P Bonville$^1$, I Mirebeau$^2$, A Gukasov$^2$, S Petit$^2$, J Robert$^2$

$^1$ CEA, Centre de Saclay, DSM/IRAMIS/Service de Physique de l’Etat Condensé, 91191 Gif-sur-Yvette, France
$^2$ CEA, Centre de Saclay, DSM/IRAMIS/Laboratoire Léon Brillouin, 91191 Gif-sur-Yvette, France
E-mail: pierre.bonville@cea.fr

Abstract. The pyrochlore material Tb$_2$Ti$_2$O$_7$, although possessing a negative paramagnetic Curie temperature witnessing the presence of antiferromagnetic exchange interactions, shows no long range magnetic order of Tb$^{3+}$ moments down to 0.05 K. The reason for this behaviour is not thoroughly understood up to now. We present here a model based on the presence of a distortion from trigonal symmetry at low temperature, making this material a two-singlet system. Application of a 4-site mean field theory allows us to construct the phase diagram for this system and to understand how, in the presence of a distortion, Tb$_2$Ti$_2$O$_7$ can remain in a paramagnetic spin liquid phase down to T=0. Although the existence of a static distortion at low temperature in these materials is not yet convincingly assessed, we believe that our model can give useful hints towards an understanding of this interesting material.

1. Introduction
Frustration in materials crystallizing in the pyrochlore lattice has been a subject of interest since the pioneering work of J. Villain [1], and an intense burst of activity in the field has taken place in the last ten years (for a review, see Ref.[2]). Among the various compounds, the titanates R$_2$Ti$_2$O$_7$ (R=rare earth) occupy a special position since many different ground states are observed for the different R$^{3+}$ ions and an understanding of their basic properties is now achieved [2]. The Tb titanate Tb$_2$Ti$_2$O$_7$ stands as an exception, since its most salient property, i.e. the absence of long range magnetic ordering of the Tb$^{3+}$ moments down to at least 0.05 K [3], remains unexplained. Numerous tentative were carried out to explain why this material remains in a spin-liquid state [4]. Its peculiar crystal electric field (CEF) level scheme, consisting of two close non-Kramers doublets separated by about 18 K and well isolated from the rest of the levels [5, 6], has been invoked to play an important role: by taking it properly into account, the spin correlations in the paramagnetic phase could be understood [7] and the formation of a “quantum spin-ice” phase was also put forward [8]. Indeed, the ground doublet is rather anisotropic, which is one of the requirements for spin-ice behaviour [9], but the effective interionic interaction is antiferromagnetic [3], which precludes a description in terms of canonical spin-ice. All these models predict a $\mathbf{k}=0$ Néel order at low temperature, indicating that some essential ingredient is lacking to obtain a correct description of the physics of Tb$_2$Ti$_2$O$_7$.  

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2. The singlet ground state systems

The Tb$^{3+}$ ion is a non-Kramers (or Van Vleck) ion, with an even number (8) of electrons in its 4f shell. Its ground spin-orbit multiplet has a total momentum $J=6$ and a Landé factor $g_J=3/2$. A specific property of non-Kramers ions is that the $(2J+1)$-fold degeneracy of this multiplet can be entirely lifted by a crystal electric field (CEF) of sufficiently low symmetry. By contrast, for Kramers ions, with an odd number of 4f electrons, an electric field cannot lift completely the spin-orbit degeneracy, even in low symmetry environments, and Kramers doublets are the eigenstates of the CEF interaction. Therefore, the low temperature properties of non-Kramers ions like Tb$^{3+}$, with a doublet ground state, are very sensitive to small perturbations of the CEF interaction (impurities, dislocations, vacancies) which lift the degeneracy of the doublet into two “non-magnetic” singlets, usually in a random way. The term “non-magnetic” refers to the fact that the mean value of the total momentum $J$ in these singlets is zero. This picture actually holds for dilute rare earth ions, where no other interaction is at play at low temperature. In concentrated rare earth systems, the interionic exchange (and/or dipole-dipole) interaction tends to establish long range magnetic order, with a “magnetic” ground state, below a critical temperature. In the case of a singlet ground state system, there occurs thus a competition between crystal field and exchange, which has long been recognised and studied [10, 11, 12]. The main result of these early works is that magnetic long range ordering (LRO) in such systems can only settle in if the exchange is larger than a critical value:

$$J_c = \frac{\Delta}{4\alpha^2},$$

where $\Delta$ is the energy separation between the singlets and $\alpha$ the matrix element of $J_z$ linking the two singlets. A non-Kramers rare earth material where the CEF interaction yields a singlet ground state can thus remain in a paramagnetic state down to very low temperature (no magnetic LRO) if the exchange integral is not large enough. Nevertheless, the exchange interaction induces short range dynamic spin correlations in this phase, hence the name Spin Liquid (SL) which is often given to this state.

3. The low temperature distortion in Tb$_2$Ti$_2$O$_7$ and the inelastic neutron spectrum

Recently, high resolution x-ray scattering measurements in Tb$_2$Ti$_2$O$_7$ [13] have evidenced anisotropic broadenings of some Bragg peaks below 15 K which were interpreted as due to cubic to tetragonal structural fluctuations, precursors to a $T=0$ cooperative Jahn-Teller (JT) transition. A cooperative Jahn-Teller transition can occur for non-Kramers ions [14] with degenerate ground state and consists of a symmetry lowering which lifts the degeneracy and lowers the energy of the system. In Tb$_2$Ti$_2$O$_7$, although a true JT transition is not observed down to 0.3 K, the distortion from trigonal symmetry has a sizeable coherence length of about 20 lattice cells at the lowest temperature. Another intriguing experimental result is the presence of a low energy excitation ($\approx 0.2$ meV) in the low temperature inelastic neutron spectra in zero field [6] and in finite fields [15, 16]. This line cannot arise from the sole trigonal symmetry CEF interaction appropriate for the rare earth site in the pyrochlore lattice ($D_{bd}$), since the first excitation has an energy of 1.45 meV in Tb$_2$Ti$_2$O$_7$. The presence of a distortion from trigonal symmetry at the rare earth site, that could be coherent over several lattice cells and during a finite time, can then be envisaged as a possible cause for the two above mentioned experimental anomalies at low temperature. In particular, the two lowest energy states would be two singlets separated by a small energy gap. The presence of a structural distortion in Tb$_2$Ti$_2$O$_7$ has already been invoked for explaining the “apparent isolation” of Tb tetrahedra from the point of view of magnetic correlations [17], but another work [18] claims for the absence of a static distortion in Tb$_2$Ti$_2$O$_7$ from $\mu$SR measurements. We will discuss the relevance of the $\mu$SR findings to our work in a later section.
In this work, we shall rely on the x-ray and neutron scattering works mentioned above to explore the consequences of the presence of a small distortion from trigonal symmetry at the Tb site in Tb$_2$Ti$_2$O$_7$. We shall assume a distortion with tetragonal symmetry along a cubic axis, which can be considered as static during a finite time, following the suggestion in Ref.[13], stating that “cubic-to-tetragonal structural fluctuations” are present below 15 K in Tb$_2$Ti$_2$O$_7$. This may not correspond to the actual symmetry breaking, but we argue that the true symmetry lowering does not play an important role in the problem. Thus, at low temperature, the degeneracy of the ground doublet is lifted and we are now dealing with a two singlet system. In earlier studies [5, 6], the Tb$^{3+}$ ground doublet in Tb$_2$Ti$_2$O$_7$ was shown to be close to the state $|J = 6; J_z = \pm 4\rangle$. Assuming here for simplicity that these are the true trigonal CEF wavefunctions, then the eigenstates in the presence of the distortion are the symmetric and antisymmetric wavefunctions:

$$
\psi_a = \frac{1}{\sqrt{2}} [|+4\rangle - |-4\rangle] \quad \text{and} \quad \psi_s = \frac{1}{\sqrt{2}} [|+4\rangle + |-4\rangle].
$$

(2)

The energy separation $\delta$ between the singlets depends on the particular form of the symmetry breaking distortion. We write the tetragonal CEF perturbation as:

$$
H_{\text{dist}} = D_Q J_Z^2 = \frac{D_Q}{3} [2J_x^2 + J_z^2 + \sqrt{2}(J_x J_z + J_z J_x)],
$$

(3)

where $OZ$ is the axis of the tetragonal distortion (chosen as the cubic $<001>$ axis). For the local frame, the local anisotropy axis $Oz$ is the cubic $<111>$ axis associated with each Tb site, and $Ox$ is the cubic $<\bar{1}\bar{1}2>$ axis.

In order to determine the value of $D_Q$, we proceed to the computation of the powder inelastic neutron scattering spectrum at 1.6 K. This spectrum reveals a sizeable low energy line near 0.05 THz (0.2 meV), on the side of the elastic line, for large $Q$ values (left panel of Fig.1). We attribute this line to the transition between the two singlets arising from the symmetry breaking. The spectrum, calculated using the CEF parameters as in Ref.[6] for the trigonal term, shows that a value $D_Q=0.25$ K reproduces correctly the position of this low energy transition (right panel of Fig.1). The eigenstates of type (2) yield a sizeable matrix element of $J_z$ between the two singlets, resulting in a non-extinct line since the transition line intensity is proportional to the square of this matrix element. Our single ion mean field calculation cannot integrate the Q-dependence due to exchange, and thus it cannot reproduce the dispersion of the CEF transition energies. The trigonal CEF parameters were chosen so as to yield an agreement with...
the mean energy value, which explains why the calculation slightly overestimates the energy of the line near 0.35 THz (1.45 meV). Since the characteristic time for these inelastic neutron data is about $10^{-11}$ s, the lifetime of the distortion must be longer than $10^{-11}$ s.

4. The phase diagrams in case of isotropic exchange

We proceed by computing the phase diagram for antiferromagnetic exchange, with the CEF scheme of Tb$_2$Ti$_2$O$_7$, obtained when varying the exchange integral, or rather the molecular field constant $\lambda$ (negative), which is related to the usual exchange constant $J$ between true spins by $J = (\frac{g J g - 1}{g J g})^2 \lambda \mu_B^2$. For this purpose, we obtain the magnetic configuration on one tetrahedron of the pyrochlore lattice by performing a self-consistent mean field calculation involving the 4 Tb ions of the tetrahedron. We take into account the short range exchange interaction with the 6 nearest neighbours of a given Tb ion. This exchange coupling can be chosen isotropic (one $\lambda$ value) or anisotropic (a three-component $\lambda$ tensor). We have also included the infinite range dipolar interaction through Ewald summations [19]. Since we consider only one tetrahedron, our calculation can simulate only long range ordered magnetic structures with a propagation vector $k = 0$. In the search for the ground configuration, we also impose the constraint that all 4 sites must be magnetically equivalent. We will compare the phase diagram obtained first in the absence of tetragonal distortion, then including the symmetry breaking. In the presence of the tetragonal distortion, one should observe a two singlet-like behaviour akin to that described in Refs.[10, 11, 12], i.e. there should be a range of $\lambda$ values where the system remains a spin liquid state down to very low temperature.

In the absence of distortion, the obtained phase diagram is shown in Fig.2. As $|\lambda|$ decreases, the ground state is first an antiferromagnetic (AF) phase (“all spins in or all spins out”), then, above a critical value $\lambda_c = -0.10 \text{T/} \mu_B$, an Ordered Spin Ice (OSI) phase (“two spins in and two spins out”), which differs from the standard Spin Ice by its long range order character. In the region with an AF ground state, there is a limited range: $\lambda_c < \lambda < \lambda_c = -0.10 \text{T/} \mu_B$, two transitions occur as temperature decreases. In the OSI phase, the Tb$^{3+}$ moment lies in the (1\bar{1}0) plane, i.e. between the cubic [001] and [110] axes. Thus, for all $\lambda$ values, the system shows magnetic Long Range Ordering, as expected since the ground Tb$^{3+}$ CEF state is a doublet. This phase diagram can be compared with that of Ref.[20], where the moments are described by

![Figure 2. (T,\lambda) phase diagram for the trigonal CEF level scheme (no distortion) of Tb$_2$Ti$_2$O$_7$ as a function of the isotropic molecular field constant $\lambda$. The green (resp. red) line and points are the transition line to an AF (resp. OSI) phase. In the range $-0.175 \text{T/} \mu_B < \lambda < \lambda_c = -0.10 \text{T/} \mu_B$, two transitions occur as temperature decreases.](image)
is −2.5. We believe the differences between the phase diagrams obtained with the two models stem from the neglect of the finite CEF anisotropy in Ref.[20].

An interesting feature of the OSI phase is that the Tb moment direction is “tilted” with respect to the easy \langle 111 \rangle axis. On Figure 3 is represented the variation of the angle \Theta=m, [111] as a function of the molecular field constant, in the OSI phase. Our convention is that \Theta is taken negative if the Tb\textsuperscript{3+} moment lies between [001] and [111], and positive for \textbf{m} lying between [111] and [110]. For the standard (non ordered) Spin Ice, \Theta = 0. One observes a continuous decrease of \Theta from a position between the [110] and [111] axes for AF exchange, towards the [001] axis for positive \lambda values. The configuration with \textbf{m} \parallel [111] (\Theta = 0) is obtained for a small non-zero value \lambda_{cr} = −0.015 T/\mu_B. These findings, for \lambda > 0, are in agreement with those of Ref.[21], where the competition between exchange and CEF anisotropy was investigated in a pyrochlore lattice, with ferromagnetic interionic exchange. For finite anisotropy, an OSI phase was obtained with \Theta < 0 ranging from −54.7° (\textbf{m} \parallel [001]) for infinitely large \lambda, to 0 for infinitely large anisotropy (Spin Ice). In the present work, the anisotropy has a fixed value determined by the CEF interaction and the \Theta = 0 limit cannot be reached for ferromagnetic exchange. It is reached for small AF exchange due to the continuous reorientation of \textbf{m} towards [110] as \lambda decreases.

We proceed now to the phase diagram including the tetragonal distortion (3), with \( D_Q = 0.25 \) K, shown in Fig.4. The main difference with the zero distortion phase diagram is the appearance of a new critical value \( \lambda_{SL} = −0.028 \) T/\mu_B above which the spontaneous Tb\textsuperscript{3+} moment is zero, at least down to 0.05 K which is the lowest temperature of our calculation. This
phase has therefore a Spin Liquid (SL), or “no LRO”, ground state and corresponds to the case studied in Ref.[10] where exchange is not strong enough so as to induce a long range ordered magnetic state. Due to the complexity of the pyrochlore lattice, with 4 magnetic sublattices, and to that inherent to the dipolar interaction, an approximate analytical expression for the critical magnetic state. Due to the complexity of the pyrochlore lattice, with 4 magnetic sublattices, and to that inherent to the dipolar interaction, an approximate analytical expression for the critical exchange value could not be obtained, similar to expression (1) derived in Ref.[10], but work is in progress in that direction. The presence of a symmetry lowering at the Tb site in Tb$_2$Ti$_2$O$_7$, making this material a two singlet system at low temperature, could therefore stand at the origin for the lack of magnetic LRO down to very low temperature. This holds provided the experimental value of the molecular field constant corresponds to the SL phase; an estimation for λ in Tb$_2$Ti$_2$O$_7$, from the Curie-Weiss behaviour of the powder magnetic susceptibility, has been given in Ref.[6]: λ ≃ −0.05 T/µ$_B$[23]. This value lies not far from, but outside the range giving rise to the SL phase, as seen in Fig.4. This phase diagram could be relevant for Tb$_2$Sn$_2$O$_7$ where an OSI phase is observed below 0.9 K and where Θ ≃13° [22]. Work is currently in progress in this direction.

5. Anisotropic exchange

Up to now, we have considered that exchange is isotropic in Tb$_2$Ti$_2$O$_7$, but recent investigations [24, 25] have put forward the presence of anisotropic exchange in the pyrochlore titanates. Anisotropic exchange has been known to be present in rare earth insulating compounds since the first developments of rare earth studies [26, 27, 28]. We will use here a simple model where anisotropy is introduced through the principal values λ$_a$, λ$_b$ and λ$_c$ of the molecular field tensor $\hat{\lambda}$. We assume that the $\hat{\lambda}$ tensor is diagonal in the {a,b,c} frame where the c-axis is along the bond joining two rare earth ions. The local susceptibility data in single crystals of the rare earth titanates, obtained from neutron diffraction experiments, were analysed in Ref.[24] in terms of a single site self-consistent calculation, and the dipolar interaction was not included. We give here a new analysis in terms of the present model: the experimental data and the calculated curves are shown in Fig.5. The dashed curves in this figure were calculated with an isotropic molecular field constant $\lambda = -0.025$ T/µ$_B$ corresponding to the largest |$\lambda$| value in the SL phase of Fig.4. The agreement with experiment is not very good at low temperature, especially for $\chi_\perp(T)$. By introducing exchange anisotropy, the agreement with experiment is improved; the susceptibility data however is not very sensitive to the $\lambda_i$ values. A good match to the data is obtained with $\lambda_a$ in the range $-0.06$ - $-0.04$ T/µ$_B$, $\lambda_b$ in the range $-0.12$ - $-0.08$ T/µ$_B$ and $\lambda_c$ ≃ $-0.07$ T/µ$_B$. We note that the small distortion from trigonal symmetry is not relevant for these susceptibility curves since its effect is sizeable only below 1-2 K, as will be shown in the next section.

Next, the phase diagram at T=0 as a function of the components of the $\hat{\lambda}$ tensor has been...
computed. We find that, for not too small values of the $\lambda_i$, it does not depend strongly on $\lambda_b$. We therefore show in Fig.6 a cut of the 3-dimensional phase diagram at the value $\lambda_b = -0.09 T/\mu_B$. For large $|\lambda_a|$ values, an OSI phase (in red) is obtained, and for smaller $|\lambda_a|$ values, a SL phase (in blue) is obtained for small $\lambda_c$ and an AF phase (in green) for large $\lambda_c$ values. The SL region of the phase diagram is relatively large, and one can mark a point in this region (pink star in Fig.6) whose coordinates are $\lambda_a = -0.04$, $\lambda_b = -0.09$ and $\lambda_c = -0.07 T/\mu_B$ and which could represent the molecular field tensor in Tb$_2$Ti$_2$O$_7$. Indeed, calculations of the low temperature properties using this particular tensor yield good agreement: i) with the local susceptibility data as discussed above, ii) with the field evolution of the magnetic structure as discussed in Ref.[29], especially with the observation of a spin-melting of some Tb moments at a critical field of about 1.5 T, and they yield a qualitative agreement with the very low temperature susceptibility data as discussed below.

6. The low temperature susceptibility

For magnetic systems with a singlet ground state, separated from the excited states by an energy $\Delta$, the magnetic susceptibility is expected to show a so-called Van-Vleck plateau for temperatures $T < \Delta/k_B$ (in case of a Spin Liquid ground state). The static magnetic susceptibility $\chi(T)$ has been measured in a powder Tb$_2$Ti$_2$O$_7$ sample, down to very low temperature (0.01 K) in Ref.[30]. These data are reported (as $1/\chi$) in Fig.7, together with the calculations using the

Figure 6. ($\lambda_c, \lambda_a$) phase diagram at $T=0$ for $\lambda_b = -0.09 T/\mu_B$, for the trigonal CEF level scheme of Tb$_2$Ti$_2$O$_7$ with a tetragonal distortion of type (3) and anisotropic exchange. The pink star marks the point in the $\lambda$-space ($\lambda_a = -0.04$, $\lambda_b = -0.09$ and $\lambda_c = -0.07 T/\mu_B$) which could represent the position of Tb$_2$Ti$_2$O$_7$.

Figure 7. Low temperature part of the thermal variation of the powder inverse susceptibility in Tb$_2$Ti$_2$O$_7$. The data are from Ref.[30], the solid line is the calculation using the present model with a tetragonal distortion and an anisotropic molecular field tensor, and the dashed red line is the calculation without tetragonal distortion. Insert: high temperature part of the inverse powder susceptibility. The solid line is a calculation with the anisotropic $\tilde{\lambda}$ tensor derived above (see text).
model developed in the present work: the black solid line represents the curve obtained with a tetragonal distortion \((D_Q = 0.25\, \text{K})\) from the trigonal local symmetry, the red dashed curve without distortion \((D_Q = 0)\). The expected Van-Vleck plateau for \(D_Q = 0.25\, \text{K}\) is clearly not observed in the data, which follow a kind of Curie-Weiss law down to 0.1 K, but the experimental value of \(1/\chi(0.1\, \text{K})\) matches well with the calculated plateau value. The curve obtained without distortion (red dashed line) is rather far from the data points. The high temperature Curie-Weiss law shown in the insert of Fig.7 is well reproduced with the anisotropic molecular field tensor determined above. We have so far no explanation for the discrepancy between the low temperature data and our calculation. It is worth noticing that, below 0.1 K, irreversibilities are observed in Ref.[30] with respect to FC and ZFC measurements procedures, showing that the ground state could be more intricate than described here. The spins I=3/2 of the \(^{159}\text{Tb}\) nucleus could also play a role, since the hyperfine interaction is evidenced in specific heat measurements below 0.4 K [31]. The problem of the very low temperature state of the \(^{3+}\text{Tb}\) ion should then be determined by considering a hamiltonian with two terms of comparable magnitude: the distortion from trigonal symmetry and the hyperfine interaction, which is present on each site since \(^{159}\text{Tb}\) is monoisotopic.

The \(\mu\)SR measurements presented in Ref.[18] conclude to the “lack of static lattice distortion in \(\text{Tb}_2\text{Ti}_2\text{O}_7\)”, which seems to contradict our statements, although we believe the distortion is probably dynamic. Actually, the \(\mu\)SR data show that the width of the dipolar field distribution experienced by the \(\mu^+\) is approximately independent of temperature. This means that no extra width is observable that would be caused by a distribution of distortions developing at low temperature. But this is fully compatible with our picture of a single distortion arising below 15 K, probably during a finite time and over a finite distance. Another conclusion of this \(\mu\)SR study is that the \(\mu^+\) Knight-shift, which is proportional to the \(\text{Tb}^{3+}\) magnetic susceptibility, saturates below about 1 K. This is in agreement with our prediction of a Van-Vleck like behaviour for \(\chi(T)\) below about 1 K (see Fig.7). The time window of the \(\mu\)SR technique being about 1 \(\mu\)s, this observation means that the lifetime of the distortion is longer than 1 \(\mu\)s.

7. Conclusion and perspectives
This work introduces a spontaneous crystal field distortion, due to an incipient Jahn-Teller transition, as a possible mechanism to account for the spin liquid ground state of \(\text{Tb}_2\text{Ti}_2\text{O}_7\), unexplained so far. Our current studies suggest that this mechanism is also at play in the parent \(\text{Tb}_2\text{Sn}_2\text{O}_7\) Ordered Spin Ice, where it could account for specific aspects of the magnetic structure and spin excitations. The spin lattice coupling, which plays a key role to relieve magnetic frustration in spinels and multiferroics with transition metals [32, 33], is here at play in terbium pyrochlores due to the peculiar \(\text{Tb}\) crystal field. Several points remain to be clarified, both from experimental and theoretical points of view. Experimentally, the precise nature of the distortion, its length and time scale remain to be determined, noticing that there is no direct observation of a symmetry lowering, and that only its consequences on microscopic magnetism are accessible to experiment. Theoretically, the influence of a distortion on the cooperative spin fluctuations (Q-dependence of the crystal field levels and/or spin waves) could be checked, among other consequences.

In contrast with spinels and multiferroics, in \(\text{Tb}_2\text{Ti}_2\text{O}_7\), the ground state selected by the spontaneous distortion is not magnetically ordered. It is stabilized in a region of the phase diagram which also involves anisotropic exchange and dipolar contributions to the energy. In \(\text{Tb}_2\text{Ti}_2\text{O}_7\), the spin liquid state induced by the distortion in zero magnetic field at ambient pressure has a counterpart in the long range antiferromagnetic orders induced by a magnetic field and/or an applied stress. Both phenomena are a manifestation of the spin-lattice coupling, whose effect is enhanced by the proximity of the first excited crystal field level of the \(\text{Tb}^{3+}\) ion. The magnetic field couples to the \(\text{Tb}^{3+}\) electric environment by mixing the CF levels (or not)
depending on its orientation with respect to the $<111>$ local axis. This induces a giant and anisotropic magnetostriction arising from a field induced distortion. In such a case, one expects the symmetry of the distortion to be intimately correlated with the symmetry of the magnetic structure. It could also provide some hints to elucidate the influence of the spin lattice coupling in this system.

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