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The spin liquid state of the $Tb_2Ti_2O_7$ pyrochlore antiferromagnet: a puzzling state of affairs

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Abstract

The pyrochlore antiferromagnet $Tb_2Ti_2O_7$ has proven to be an enigma to experimentalists and theorists working on frustrated magnetic systems. The experimentally determined energy level structure suggests a local $\langle 111 \rangle$ Ising antiferromagnet at low temperatures, $T \lesssim 10$ K. An appropriate model then predicts a long-range ordered $\mathbf{Q} = \mathbf{0}$ state below approximately 2 K. However, muon spin resonance (μSR) experiments reveal a paramagnetic structure down to tens of millikelvins. The importance of fluctuations out of the ground state effective Ising doublet has been recently understood, for the measured paramagnetic correlations cannot be described without including the higher crystal field states. However, these fluctuations treated within the random phase approximation (RPA) fail to account for the lack of ordering in this system below 2 K. In this work, we briefly review the experimental evidence for the collective paramagnetic state of $Tb_2Ti_2O_7$. The basic theoretical picture for this system is discussed, where results from classical spin models are used to motivate the investigation of quantum effects to lowest order via the RPA. Avenues for future experimental and theoretical work on $Tb_2Ti_2O_7$ are presented.

1. Introduction

Frustrated magnetism has been actively studied for decades [1–3], and interest continues to grow. In models such as the antiferromagnetic (AFM) J_1–J_2 model on a bipartite lattice, frustration results from a competition between two different ground state spin structures, one

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favoured by J_1 and the other by J_2 [4]. The J_1–J_2 model has motivated many interesting theoretical studies, but the tuning of energy scales to induce frustration is not easily achieved experimentally. Another class of materials and models exhibit frustration at the level of nearest neighbour interactions via a competition imposed by the lattice geometry, creating geometric frustration[6]. Crystal geometries formed from unit triangles and AFM interactions are frustrated because the spins about a triangle cannot arrange themselves so that all pairwise interactions are satisfied (i.e., $-J_1\mathbf{S}_i \cdot \mathbf{S}_j$ for $J_1 < 0$ cannot be minimized for each pair (i, j)). Hence, the triangular and Kagomé lattices are common frustrated geometries in 2D. In three dimensions, lattices of corner sharing triangles (garnet structures) or tetrahedra (pyrochlore and spinel structures) are frustrated with nearest neighbour AFM interactions.

The pyrochlore lattice has received increased attention recently because the $S = 1/2$ AFM model is predicted to be a collective paramagnet, or spin liquid, in the ground state [5, 6]. A 3D spin liquid is unusual [7], but the role of lattice dimensionality appears to be reduced in the face of large quantum fluctuations driven by frustration [5]. The stabilization of a non-collinear long-range ordered state is expected when long-range interactions are included [8], or when constraints arising from single-ion anisotropy are imposed [9]. Magnetic insulating pyrochlores, general formula $A_2B_2O_7$, where A is a rare earth ion (Ho^{3+}, Dy^{3+}, Tb^{3+}, Gd^{3+}), are a subset of frustrated magnets that are at the focus of much experimental and theoretical work [1, 2]. This family of materials displays long-range order [10], a novel 'ice-like' phase with residual entropy [2, 11, 12], and collective paramagnetic behaviour at millikelvin temperatures [13]. The focus of our brief review is the candidate 3D spin liquid $Tb_2Ti_2O_7$ [13]. The many conflicting experimental results and theoretical predictions for this material are discussed, and the recent progress toward a general picture for $Tb_2Ti_2O_7$ is presented.

2. Experimental picture of $Tb_2Ti_2O_7$

The pyrochlore $Tb_2Ti_2O_7$ has dominant AFM interactions as determined from dc magnetization measurements on a polycrystalline sample; $\theta_{CW} \approx -20$ K [13]. Results from muon spin resonance (μSR) clearly indicate dynamic moments down to 70 mK despite the development of short-range correlations at approximately 50 K, as seen in neutron scattering measurements [13, 14]. Suppression of an ordering temperature in this material by two orders of magnitude (i.e., $|\theta_{CW}|/70$ mK $= O(10^2)$) can be understood in the context of a nearest neighbour AFM. However, $Tb_2Ti_2O_7$ has a measured spin anisotropy gap of $\Delta \approx 18$ K [13, 15], attributable to crystal field effects, as well as reasonably strong dipolar interactions [15, 16], so such a large reduction in T_c is quite puzzling.

As a first step to unraveling the mystery of $Tb_2Ti_2O_7$, one has to determine the single-ion properties of Tb^{3+} in the dense material. From experiments and calculations, Gingras *et al* find that the Tb^{3+} moment is approximately 5 μ_B, a substantial reduction from the free ion result 9.6 μ_B [15]. In addition, the crystal field effects on Tb^{3+} ($S = 3$, $L = 3$ so $J = 6$) yield an effective Ising ground state level structure with a quantization axis oriented along the local $\langle 111 \rangle$ direction [15, 17], with a gap to the next energy levels, an excited doublet, of the order of $\Delta \approx 18$ K. This $\langle 111 \rangle$ anisotropy is significant since for AFM effective nearest neighbour interactions it greatly reduces the frustration from that of a Heisenberg-like model [9]. With a better grip on the dipolar strength and the crystal field levels, a revised Curie–Weiss temperature of $\theta_{CW} \approx -14$ K for the dipoles and exchange is calculated and an ordering temperature of 1 K deduced [15, 16]. Also, from theoretical work on a dipolar spin ice model ($\langle 111 \rangle$ Ising)

[6] Frustration also results if there is distribution of interaction strengths between moments, such systems (spin glasses) are not considered here.

one is able to show that for the parameters of $Tb_2Ti_2O_7$, a long-range ordered state with zero net moment about each tetrahedron (an all-in all-out state) is expected at $T_c \approx 1.1$ K [16]. That experiments prove these predictions wrong is possibly due to the effects of long-range exchange beyond nearest neighbour or quantum fluctuations.

With the single-ion properties suggesting a $\langle 111 \rangle$ Ising dipolar model for $Tb_2Ti_2O_7$ at $T < 10$ K, but the absence of any agreement between this model and experiments on an ordering temperature, a re-evaluation of the model is necessary. Hence, the focus of the discussion shifts to the paramagnetic (PM) regime and the short-range correlations. Early inelastic neutron data on a single crystal found evidence for the partial softening of the lowest energy modes about [002] and [220] [14, 18]. Elastic scattering measurements on a single crystal of $Tb_2Ti_2O_7$ clearly display diffusive scattering about these same regions in the (hhl) plane with the strongest correlations centred about [002] [18, 19]. The qualitative features of the structure factor ($S(\mathbf{q})$) in the PM phase can be roughly fitted with a near neighbour AFM Heisenberg model [18], suggesting that the strict Ising anisotropy might not hold for temperatures defining the PM regime of this material, i.e., $T \gtrsim 0.5$ K. Note that the PM $S(\mathbf{q})$ is measured at 9 K in Gardner *et al* [18] but at 0.4 K in Yasui *et al* [19]. A relaxation of the $\langle 111 \rangle$ spin anisotropy in a long-range dipolar AFM pyrochlore model does yield good agreement with the PM $S(\mathbf{q})$ in this material for temperatures $T \gtrsim |\theta_{CW}| \gtrsim 10$ K [20, 21].

Recent data for $Tb_2Ti_2O_7$ in the low temperature regime suggests the existence of a spin glass-like state. Hysteresis is observed in the field cooled and zero-field cooled neutron scattering data below 1.5 K [19]. Thermodynamic measurements of the static susceptibility also indicate spin glass physics, albeit at a much lower temperature, 70 mK [22].

Hence, at present the general consensus is that $Tb_2Ti_2O_7$ remains a collective paramagnet down to $T = 50$–70 mK at ambient pressures. Under high pressure, 2–8 GPa, the material orders at $T_c \approx 2$ K [23], but the spin structure is not the $\mathbf{Q} = \mathbf{0}$ all-in all-out structure predicted from a $\langle 111 \rangle$ Ising model [16, 21], or the 'nearby' long-range ordered spin ice state [24]. An applied field can also induce an ordered state in this material [25]. Field driven ordered states is a current topic for the spin ice material $Dy_2Ti_2O_7$ [26], but unlike the spin ice materials case, moderate field strengths along high symmetry directions indicate weaker spin anisotropy for $Tb_2Ti_2O_7$ [19]. We note that the gap between the ground state doublet and the first excited states in $Tb_2Ti_2O_7$ is an order of magnitude smaller than the gap measured in the spin ice materials [17].

3. Theoretical interpretation of $Tb_2Ti_2O_7$

The Heisenberg AFM on the pyrochlore lattice is defined by the simple Hamiltonian $H = -J_1 \sum_{\langle (i,a),(j,b) \rangle} \mathbf{S}_i^a \cdot \mathbf{S}_j^b$, where $J_1 < 0$, (i, a) describes the location of a spin by its fcc lattice position $\mathbf{R}_i$ and tetrahedral basis point $\mathbf{r}^a$, and interactions are among nearest neighbours only. The Hamiltonian H represents a highly frustrated model that produces a spin liquid ground state for quantum spins ($\mathbf{S}_i^a = 1/2$) [5, 6] or classical spins ($\mathbf{S}_i^a = \hat{x} S_i^{a,x} + \hat{y} S_i^{a,y} + \hat{z} S_i^{a,z}$) [27–30]. In the classical model, an ordered state can be induced by perturbations arising from exchange interactions beyond nearest neighbours [29] or by constraints imposed by spin isotropy [9]. If the single-ion anisotropy is along the local $\langle 111 \rangle$ direction (i.e., $\mathbf{S}_i^a = \hat{z}^a \sigma_i^a$ where $\hat{z}^a$ is the local $\langle 111 \rangle$ direction and $\sigma_i^a = \pm 1$), then it is the nearest neighbour ferromagnetic pyrochlore (nearest neighbour spin ice) that is frustrated [2, 9, 11]. For rare earth magnets such as the spin ice materials and $Tb_2Ti_2O_7$, long-range dipolar interactions are a significant contribution to the Hamiltonian. Therefore, the basic model for rare earth pyrochlore insulators can be

written out:

$$H = -J_1 \sum_{\langle (i,a),(j,b)\rangle} \mathbf{S}_i^a \cdot \mathbf{S}_j^b + D_{\rm dd} \sum_{(i,a)>(j,b)} \left(\frac{\mathbf{S}_i^a \cdot \mathbf{S}_j^b}{|\mathbf{R}_{ij}^{ab}|^3} - \frac{3(\mathbf{S}_i^a \cdot \mathbf{R}_{ij}^{ab})(\mathbf{S}_j^b \cdot \mathbf{R}_{ij}^{ab})}{|\mathbf{R}_{ij}^{ab}|^5} \right), \tag{1}$$

where $D_{\rm dd} = DR_{\rm nn}^3$ is the nearest neighbour dipole strength, $D = \mu_0\mu^2/4\pi R_{\rm nn}^3$, μ is the moment on the rare earth ion, $R_{\rm nn}$ is the nearest neighbour distance, and $\mathbf{R}_{ij}^{ab}$ is the vector separation between spins $\mathbf{S}_i^a$ and $\mathbf{S}_j^b$. If the spins in equation (1) are constrained to their local $\langle 111\rangle$ Ising axes, then equation (1) represents the dipolar $\langle 111\rangle$ Ising model (DIM) that successfully describes the specific heat [16] and paramagnetic correlations [31] of spin ice. When the DIM is simulated by a Monte Carlo algorithm employing nonlocal loop moves, a first-order transition to a spin ice ground state is found [24], with an ordering wavevector and spin structure in agreement with mean-field calculations [20].

The phase diagram of the DIM on the pyrochlore lattice is generated by defining the sign and magnitude of J_1 and the strength of the dipole–dipole interaction D [16]. The DIM exhibits two phases depending on the ratio J_1/D: for $J_1/D \gtrsim -4.525$ a spin ice manifold results (macroscopic degeneracy), where two spins point in and two spins point out along their local $\langle 111\rangle$ axes for each unit tetrahedron. For $J_1/D < -4.525$, a non-collinear long-range ordered state results, where all spins about a unit tetrahedron point either in or out along their local $\langle 111\rangle$ axes; this $\mathbf{Q} = \mathbf{0}$ structure is replicated over the lattice. We note that going from the PM to the $\mathbf{Q} = \mathbf{0}$ state occurs via a second-order phase transition, but the path from the PM to spin ice manifold represents a crossover to a dynamically frozen state without long-range order. In the DIM, the temperature of the phase transition or crossover is signalled by the peak in the calculated specific heat, whose position depends on the value of J_1/D [16]. Hence, the important energy scale in this model is the effective nearest neighbour exchange, $J_{\rm eff} = J_1/3 + 5D/3$, where $1/3$ and $5/3$ are geometric factors arising from the inner product of the local $\langle 111\rangle$ spin quantization axes. With $J_1/D \approx -5.5$ for $Tb_2Ti_2O_7$, the prediction is a $\mathbf{Q} = \mathbf{0}$ state at approximately 1 K [15, 16]. The subtle point about $Tb_2Ti_2O_7$ in the DIM is that it sits near the boundary between the two phases, making it susceptible to fine tuning of the model parameters (exchange or dipolar energy scales and single-ion crystal field state wavefunctions). It is conceivable that $Tb_2Ti_2O_7$ might sit close to a line of disorder in the phase diagram where fluctuations dominate and a tendency to order is suppressed. The simulation of the DIM near the phase boundary presents some numerical challenges and is not well understood [24].

As mentioned in section 2, $Tb_2Ti_2O_7$ does not order at $T \approx 1$ K, and suggestions from the PM $S(\mathbf{q})$ results [18, 19] that the $\langle 111\rangle$ Ising constraint should be relaxed in this case provide a hint of where to venture next. In considering the symmetry of the DIM, one can show that no strict $\langle 111\rangle$ Ising model on the pyrochlore lattice can reproduce the short-range correlations observed in experiments, but a model in which the rotational symmetry is partially restored is found to admit a solution compatible with experiments [20]. Mean-field and Monte Carlo calculations make these claims more quantitative [20]. Hence, in the PM regime the physics of $Tb_2Ti_2O_7$ cannot be ascribed to the same $\langle 111\rangle$ Ising interpretation of the moments that works so well for the spin ice materials.

A pitfall with a classical Heisenberg description for $Tb_2Ti_2O_7$ is that complete restoration of spin isotropy does not agree with experiments and calculations of the single-ion properties of the Tb^{3+} [15, 17]. The g-tensor of Tb^{3+} in $Tb_2Ti_2O_7$ exhibits $\langle 111\rangle$-like anisotropy below 10 K and a ground state doublet structure that is separated from a first excited doublet by about 18 K, which is of the order of the Curie–Weiss temperature for exchange and dipolar interactions ($\theta_{\rm CW} = -14$ K) [15]. For the spin–orbit coupled Tb^{3+} moments ($J = 6$), the angular momentum for the two lowest lying doublets is predominantly $|M_J = \pm 4\rangle$ for the

ground state and $|M_J = \pm 5\rangle$ for the first excited state [15]. Hence, transverse fluctuations between these two lowest lying doublets are allowed and non-negligible at the temperatures of the $S(\mathbf{q})$ experiments, 5–10 K [18, 21].

Applying a random phase approximation (RPA) to a model with exchange and long-range dipoles, the fluctuations between the lowest lying crystal field states can be studied, i.e., the $\langle 111 \rangle$ Ising limit is recovered when transitions between states $|\psi_0\rangle$ (ground state) and $|\psi_1\rangle$ (first excited state) have a vanishing matrix element, $\langle \psi_1 | S^{\pm} | \psi_0 \rangle = 0$. Using $|\psi_0\rangle = |M_J = \pm 4\rangle$ and $|\psi_1\rangle = |M_J = \pm 5\rangle$, the RPA [21] gives excellent semi-quantitative agreement with the PM $S(\mathbf{q})$ and the energy dispersion between the low lying magnetic states of the neutron experiments [18]. However, the RPA still predicts an ordered state at $T_c \approx 2$ K. Hence, the RPA demonstrates the sensitivity of the PM correlations observed in neutron scattering to quantum fluctuations in this unusual magnetic system ($|\psi_0\rangle = |M_J = \pm 4\rangle$ and $|\psi_1\rangle = |M_J = \pm 5\rangle$), but it still gives a T_c of approximately 2 K for the above all-in all-out $\mathbf{Q} = \mathbf{0}$ state.

4. Avenues for future studies and conclusions

In order to understand the physics of $Tb_2Ti_2O_7$ below 2 K, we need to go beyond the first-order approximation of the RPA and consider quantum effects more rigorously. A reasonable first step would be to employ a more detailed description of the crystal field states of the Tb^{3+} ion, and then form an effective low energy Hamiltonian from the more complicated model following a procedure similar to that used in the derivation of the t–J or Heisenberg models from the Hubbard model. Experimentally, low temperature neutron scattering (elastic and inelastic) measurements on single crystals are needed to probe the fluctuations between crystal field states [13, 14, 18]. Exploring these same properties but at hydrostatic pressures below 2 GPa could provide a valuable link to the high pressure ordered state of $Tb_2Ti_2O_7$ [23].

In conclusion, there is strong evidence that $Tb_2Ti_2O_7$ cannot be described by a classical $\langle 111 \rangle$ Ising Hamiltonian similar to the successful model of the spin ice materials [2]. Spin fluctuations must be incorporated into any model of $Tb_2Ti_2O_7$ in order to capture the PM correlations, and a simple description of quantum fluctuations via the RPA is sufficient to achieve good agreement with experiments. However, to study the lack of an ordering transition in $Tb_2Ti_2O_7$, a more careful treatment of the quantum fluctuations is essential. Finally, we come back to the $\langle 111 \rangle$ Ising dipolar model phase diagram and note that $Tb_2Ti_2O_7$ is very close to the AFM/spin ice phase boundary [16]. There may be issues with the quantum fluctuations, long-range exchange, microscopic disorder at very low temperatures, or tuning of material or model parameters that places $Tb_2Ti_2O_7$ on a line or region of disordered semi-classical ground states.

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