PRESSURE INDUCED CRYSTALLIZATION OF A SPIN LIQUID

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$Tb_2Ti_2O_7$ is an insulating pyrochlore oxide in which localized Tb^{3+} spins occupy a lattice of corner-linked tetrahedra. Antiferromagnetic Heisenberg interactions on this lattice are highly frustrated, giving rise to macroscopic degeneracy in the ground state. In general this degeneracy is lifted by perturbations arising from chemical disorder or additional magnetic interactions, yielding either spin glass-like or long range ordering transitions. $Tb_2Ti_2O_7$ is the only compound which remains in a fluctuating paramagnetic state down to 70 mK [1]. Short range antiferromagnetic correlations between the Tb spins develop below 100 K, corresponding to a "spin liquid" state. The single ion ground state in $Tb_2Ti_2O_7$ is a crystal field doublet, which interacts with its neighbors via superexchange and dipolar coupling. Given these simple interactions, the absence of magnetic order is surprising. Applied pressure allows one to study this stability, by perturbing the balance of the various interactions, which have different dependencies on interatomic distance.

We used LLb-Kurchatov pressure cells and the specialized high pressure diffractometer G6-1, the only apparatus that allows neutron scattering at both very low temperatures (1.4 K) and very high pressures, (10-50 GPa) (2). Figure 1 shows neutron diffraction spectra at 1.4 K for three pressures, focusing on the diffuse intensity in the region of the 111 nuclear peak. At P = 0, the diffuse intensity arising from liquid-like magnetic correlations shows no indication of magnetic long range order. At 1.5 GPa, small magnetic Bragg peaks start to emerge from the diffuse background. At 8.6 GPa, the average intensity becomes much lower, but the magnetic peaks are now very clearly seen. Concomitantly, the diffuse intensity shows a stronger modulation. The onset of narrow Bragg peaks shows the development of magnetic long range order, or crystallization of the spin liquid state, induced by pressure.

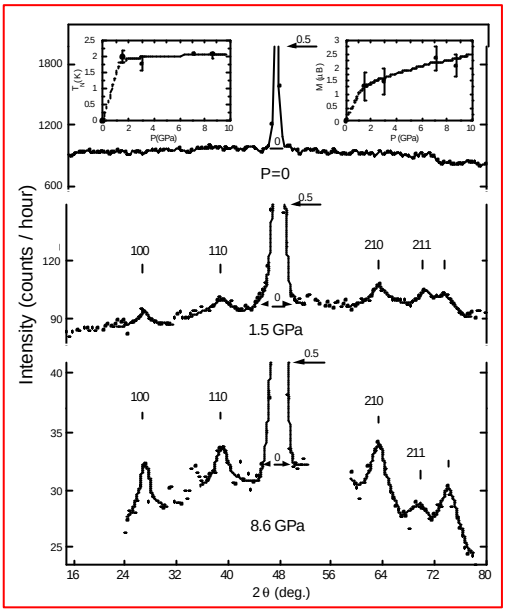

Figure 1. $Tb_2Ti_2O_7$: raw neutron diffraction spectra (neutron counts/hour) for three pressures at 1.4 K. The incident neutron wavelength is 4.741 Å. Intensity scales are chosen to show the magnetic peaks as compared with the 111 structural peak. Half intensity of the 111 peak is shown at the center of the spectra. Insets: the Néel temperature (left) and ordered magnetic moment at 1.4 K (right) vs. pressure.

The ordered magnetic structure has a simple cubic unit cell, derived from the chemical one of Fd-3m symmetry by a propagation vector **k** = 100 or 110. There is no magnetic contribution to the structural peaks. The antiferromagnetic structure is not predicted by theoretical models and differs from the few ordered structures reported in pyrochlore compounds. The Néel temperature T_N = 2.1 K is pressure independent. Below T_N, the ordered and spin liquid phases coexist. The ordered moment increases with pressure, but remains always below the calculated value of 5 μ_B, as expected due to the coexistence of liquid and ordered states.

The modulation amplitude of the diffuse scattering A(P,T) shows clear evidence of this coexistence. A(P,T) defined experimentally as I_{max}-I_{min}, where I_{max} and I_{min} are the extrema of the diffuse intensity, is proportional to the thermal average of the first neighbor spin correlations. It increases with decreasing temperature, and this effect becomes more pronounced as pressure increases (Fig. 2). The onset of long range order at T_N coincides with a sharp kink of A. Below T_N, the decrease of A mirrors the increase of the Bragg intensity, showing that spin liquid and ordered phases coexist. The magnetic state below T_N reminds one of a mixed solid-liquid phase, with both static and dynamical character, and a gradual transfer of intensity from liquid to ordered state as temperature decreases.

The coexistence of spin liquid and ordered states and the fact that T_N is pressure independent suggest that the transition may be of first order. The pressure induced order provides further compelling evidence that the spin liquid state in $Tb_2Ti_2O_7$ is most unusual. In liquid helium, pressure induces crystallization by simply strenghtening the interatomic interactions, which reduces the quantum fluctuations. Here the role of pressure is more intricate. One possibility is that the frustration is relieved by a pressure induced structural distortion, but this has not been observed. A more intriguing possibility is that in the spin liquid state, exchange dipolar and crystal field interactions are naturally balanced in such a way as to make quantum fluctuations significant. This delicate balance is then destroyed by pressure, resulting in magnetic order.

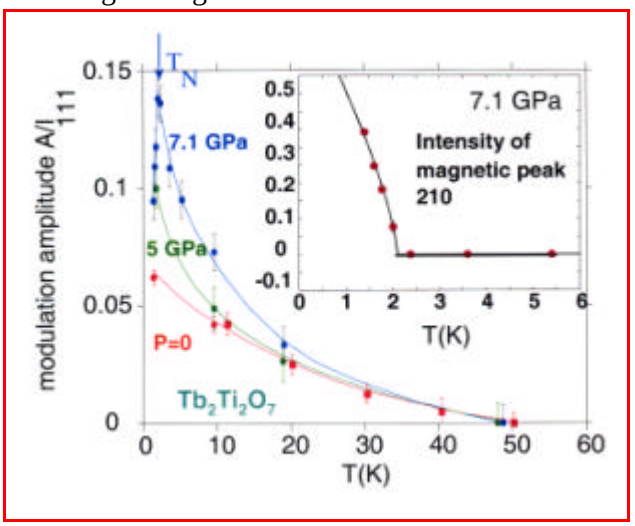

Figure 2. Temperature dependence of the modulation amplitude A(P,T) for the pressures P = 0, 5 and 7.1 GPa, where A(P,T) = I_{max}-I_{min}, the difference in extrema of intensity in our experimental range. To compare data at different pressures, A(P,T) is scaled to the integrated intensity of the 111 Bragg peak. At 5 and 7.1 GPa, solid lines above T_N are fits with the law -ln(T/T_{up}), T_{up} = 46 K. At 5 GPa, only the paramagnetic regime was investigated. Inset: integrated intensity of the 210 magnetic peak (scaled to the 111 peak intensity) vs. temperature at 7.1 GPa. The solid line is a guide to the eye.

References

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[2] I. Goncharenko and I. Mirebeau, *Rev. High Pressure Sci. and Technol.* **7** (1998) 475.

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