Frustrated, Satisfied and Fluctuating Ground States in Pyrochlore Magnets

B. D. Gaulin

- Neutron and X-ray Scattering from Exotic Magnets
- Spin liquid and Ordered Ground States in the Pyrochlore Antiferromagnet $\text{Tb}_2\text{Ti}_2\text{O}_7$
- Spin Ice Ground State in $\text{Ho}_2\text{Ti}_2\text{O}_7$
- Structural Fluctuations within the Spin Liquid State of $\text{Tb}_2\text{Ti}_2\text{O}_7$
Collaborators

• J.P.C. Ruff
• S.R. Dunsiger
• K.C. Rule
• J.P. Clancy
• M.J. Lewis

McMaster University

• A.J. Berlinsky
• H.A. Dabkowska
• C. Kallin
• J.E. Greedan

• M.J.P. Gingras

University of Waterloo

• R.F. Kiefl

University of British Columbia

• J.S. Gardner
• Y. Qiu
• J.R.D. Copley

NIST

now at: HMI Berlin
Geometrical Frustration:
Antiferromagnetism + Triangles and Tetrahedra

Ground State of the Ising AF on a Triangular Lattice

Entropy at T=0 is finite ~ 0.34 R

No LRO at any temperature
Geometrical Frustration:

Mean Field Theory predicts a phase transition near \( T = |\Theta_{CW}| \), but materials remains disordered to much lower temperatures – **Spin Liquid**

Non-generic!

*eg: Noncollinear AF Spin glass*

\[ \chi^{-1} \]

\[ \Theta_{CW} \]

**Spin Liquid**

**Paramagnet**

\[ -\Theta_{CW} \]

\[ \Theta_{CW} \]

Temperature

Mean Field Theory predicts a phase transition near \( T = |\Theta_{CW}| \), but materials remains disordered to much lower temperatures – **Spin Liquid**
Frustration in three dimensions:
The cubic pyrochlore structure;
A network of corner-sharing tetrahedra

Low temperature powder neutron diffraction from 
$\text{Tb}_2\text{Ti}_2\text{O}_7$
μSR Studies of Magnetic Ground States in:

\[
\begin{align*}
\text{Tb}_2\text{Ti}_2\text{O}_7 & : \text{Spin Liquid} \\
\text{Tb}_2\text{Mo}_2\text{O}_7 & : \text{Spin Liquid and Spin Glass} \\
\text{Y}_2\text{Mo}_2\text{O}_7 & : \text{Spin Glass}
\end{align*}
\]
Inelastic neutron scattering on polycrystalline \( \text{Tb}_2\text{Ti}_2\text{O}_7 \)

\(~ \Delta : \text{Ho}_2\text{Ti}_2\text{O}_7 \sim 240 \text{ K} ; \text{Dy}_2\text{Ti}_2\text{O}_7 \sim 380 \text{ K} \)
Rare Earth moments:

Strong [111] anisotropy

Ferromagnetic exchange:

“Spin Ice”: 2 in 2 out

Harris, Bramwell et al, PRL, 79, 2554, 1997

\[
H = - J \sum_{\langle ij \rangle} S_i^{z_i} \cdot S_j^{z_j} + D r_{nn}^3 \sum_{j>i} \frac{S_i^{z_i} \cdot S_j^{z_j}}{|r_{ij}|^3} - \frac{3(S_i^{z_i} \cdot r_{ij}) (S_j^{z_j} \cdot r_{ij})}{|r_{ij}|^5}
\]
2-Axis Diffraction
T=9 K

TOF scattering
T=0.4 K
Outstanding Questions:

• Why is $\text{Tb}_2\text{Ti}_2\text{O}_7$ disordered as low as 0.02 K (in $H=0$, $P=0$)?

• $S(Q)$ at low $T$ is (perhaps naively) incompatible with Ising (111) anisotropy. What is going on?
Ordered phases appear on application of $H // 110$
Low field (002) phase persists to very high $T > 25$ K

High field (112) phase exists on expected $T_N \sim 2$ K
Application of weak [1-10] magnetic field breaks system up into $\alpha$ and $\beta$ chains.

Polarizable $\alpha$-[1-10] chains (parallel to field)

Perpendicular $\beta$-[110] chains
Application of weak [1-10] magnetic field breaks system up into $\alpha$ and $\beta$ chains.

Polarizable $\alpha$-[1-10] chains (parallel to field)

Perpendicular $\beta$-[110] chains
$\alpha$-chains polarized along the [1,1,0] Direction

Diffuse (BG) $B = 2T, -0.5 < E < 0.5$

Half Field Aligned

$P$ = Paramagnetic Site
Field Aligned, Local $<111>$ AFM

Denotes Z-Component

“3 in – 1 out” local structure
Canted AFM, Moments not respecting Ising 111 axes. (Magnetization is ~11% of saturation along [110])

+ / - Denotes Z-Component
One Transition in Zero Field

Five Transitions in Non-Zero Field
Spin Ice Ground State in Ho$_2$Ti$_2$O$_7$
Ho$_2$Ti$_2$O$_7$ vs Tb$_2$Ti$_2$O$_7$

Static Spin Ice vs Dynamic Spin Liquid (H=0)

Magnetic Structure Factors appear complementary
Ferro-ordering of $\alpha$-chains

1D-correlations along $\beta$-chains
Spin Ice Ground State in $\text{Ho}_2\text{Ti}_2\text{O}_7$ is static

Spin Liquid Ground State in $\text{Tb}_2\text{Ti}_2\text{O}_7$ is dynamic
Very High Resolution
Inelastic Neutron Scattering (Backscattering)
Sees “Spin Ice” Freeze

Energy Scale of Freezing is 100 neV!
High Resolution X-Ray Scattering from Tb$_2$Ti$_2$O$_7$

Ruff et al., cond-mat/0707.1682
TbVO$_4$

Cooperative Jahn-Teller
Tetragonal - Orthorhombic
$T_C \sim 33.3$ K

(12, 0, 0) splits in the longitudinal direction

(8, 8, 0) splits in the transverse direction
TbVO$_4$

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**Conclusions:**

- New neutron scattering infrastructure leads to new sensitivity
  New time-of-flight neutron infrastructure at SNS, JSNS, 2TS@ISIS
  will give FOM improved by factors of ~ 50 – New Science!

- **Antiferromagnetic Pyrochlore** \( \text{Tb}_2\text{Ti}_2\text{O}_7 \):  
  
  Spin Liquid State in \( H=0 \) comes to order in small(ish) fields  
  Dispersive collective spin excitations observed in ordered states  
  – evidence for continuous spin symmetry

- Structural fluctuations characteristic of cooperative Jahn-Teller  
  phase transition accompany appearance of spin liquid state in \( \text{Tb}_2\text{Ti}_2\text{O}_7 \)  
  – grow continuously with decreasing \( T \) to \( T \sim 0.3 \text{ K} \).