Abstract: The pyrochlore magnet Yb2Ti2O7 has the remarkable property that it orders magnetically, but has no propagating magnons over wide regions of reciprocal space. Using inelastic neutron scattering we observe that at high magnetic fields, in addition to dispersive magnons there is also a two-magnon continuum, which grows in intensity upon reducing field, overlaps with one-magnon states at intermediate fields leading to strong dispersion renormalizations and magnon decays. We re-evaluate the Hamiltonian finding dominant quantum exchange terms, which we propose are responsible for the anomalously strong quantum fluctuation effects observed at low fields.
Quasiparticle breakdown in the quantum pyrochlore $\text{Yb}_2\text{Ti}_2\text{O}_7$ in magnetic field

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Inelastic Neutron Scattering on LET spectrometer @ ISIS Facility
Spin ice physics on the pyrochlore lattice

- corner-shared tetrahedra, Ising spins (local 111 axis) coupled FM many degenerate states ⇒ Spin Ice, 2 in 2 out, Ho$_2$Ti$_2$O$_7$

  \[ J_{zz} \]

  spin-flip (two monopole)

  \[ 0 \]

  spin-ice states

- additional “transverse exchange” terms ⇒ “quantum spin ice” (photon + propagating monopoles)

  \[ J_{zz} \]

  Magnetic Monopoles
  Electric Charges
  Spinons

  \[ J_{zz}^{1}/J_{zz}^{2} \]

  Visons
  Magnetic Monopoles

  \[ \text{Photon Excitations} \]
Yb$_2$Ti$_2$O$_7$ spin dynamics: open questions

- **how magnons disappear upon lowering field?**
- if broad scattering at B=0 is due to quantum fluctuations, are fluctuations still present at high field, what is their manifestation, **how do fluctuations evolve upon lowering field** (gradually or with a sharp onset below a critical field)?
Yb$_2$Ti$_2$O$_7$ single crystal specific heat

- our image furnace “slow”-grown & annealed single crystal shows a single, sharp specific heat peak at 0.214 K $\rightarrow$ similar in behaviour to high purity limit (stoichiometric powders have 1$^{\text{st}}$ order transition 0.24-0.26 K)

Spin dynamics at high magnetic field // [001] 0.15 K

- observe sharp modes with gap increasing in field, as seen by Ross et al. in B//[−1,1,0]
- coherently-propagating spin-flips on 4 sublattices
Parameterization by spin waves of a nn Hamiltonian

\[ H_{\text{Exchange}} = \sum_{\langle ij \rangle} \left\{ J_{zz} S^z_i S^z_j - J_{\pm} (S^+_i S^-_j + S^-_i S^+_j) \right\} \]

\[ J_{\pm \pm} (\gamma_{ij} S^+_i S^+_j + \gamma^*_{ij} S^-_i S^-_j) \quad g = \begin{pmatrix} g_\perp & 0 & 0 \\ 0 & g_\perp & 0 \\ 0 & 0 & -g_{||} \end{pmatrix} \]

\[ + J_{z \pm} [S^z_i (\zeta_{ij} S^+_j + \zeta^*_{ij} S^-_j) + (i \leftrightarrow j)] \}

- pick dispersion points throughout the full volume of data (17 directions total) get (h,k,l,E) and mode index 1-4
- **fit 6 parameters** (4 symmetry-allowed nn exchange terms) + g tensor (2 terms) to data from both neutron experiments B//[001] and published B//[−1,1,0]
to fit all Hamiltonian parameters without any constraints use neutron data for the **two field orientations** (over 550 dispersion points) and **magnetization near saturation** (7 T)

\[
\begin{align*}
J_{zz} &= 0.026(3) \text{ meV} \\
J_\pm &= 0.074(2) \text{ meV} \\
J_{\pm\pm} &= 0.048(2) \text{ meV} \\
J_{z\pm} &= -0.159(2) \text{ meV} \\
g_\parallel &= 2.14(3) \\
g_\perp &= 4.17(2).
\end{align*}
\]
Parameterization of dispersions at $B=5T // [-1,1,0]$

**This work**

- $J_{zz} = 0.026(3)\text{ meV}$
- $J_{zz} = 0.17 \pm 0.04$,
- $J_{\pm} = 0.074(2)\text{ meV}$
- $J_{\pm} = 0.05 \pm 0.01$,
- $J_{++} = 0.048(2)\text{ meV}$
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- $J_{z\pm} = -0.159(2)\text{ meV}$
- $J_{z\pm} = -0.14 \pm 0.01$

**Ross et al (2011)**

- $g_{\parallel} = 2.14(3)$
- $g_{z} = 1.80$
- $g_{\perp} = 4.17(2)$. $g_{xy} = 4.32$

*(see also Robert et al PRB 2015)*

$g_{xy}/g_{z} = 2.4$ fixed
Parameterization of dispersions at B = 5T // [001]

- earlier parameterization does not fit data in [001] field
Convergence of refinement of Hamiltonian parameters

**This work**  
**Ross et al (2011)**

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| $g_\parallel / g_\parallel$ | 1.95(3)     | $g_{xy} / g_z = 2.4$ | fixed

- unique solution explains **all existing dispersions data (2 field orientations) + saturation magnetization**
- refined g-factor ratio agrees with recent crystal field parameterization 1.96 +/- 0.13  
  *J. Gaudet et al (2015)*
- agrees with parameterization of diffuse scattering at 0.4 K Roberts et al PRB (2015)  
  *DiLong et al (2014)*
- agrees with THz data (energies &polarization)
Parameterization by spin waves of a nn Hamiltonian

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### Convergence of refinement of Hamiltonian parameters

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Semi-classical Phase Diagram

- revised parameters put system almost on phase boundary Splayed FM – AFM $\Psi_{2,3}$

=> strongly frustrated

- mean-field $T_C \sim 3$ K >> actual $T_c = 0.21-26$ K

Classical Spin Ice point

classical degeneracy of ice states preserved
Two-magnon scattering continuum

- at higher energies see additional weak continuum scattering (1-2% of one-magnon weight)

2 magnon continuum

1 magnon dispersions

Energy (meV)

Intensity (arb. units)

Energy (meV)

Energy (meV)

Magnetic Field $\mu_0 H_{int}$ (T)

Magnetization $M/T$ (T)

MPWS Data

Model

0.21 T

2 Magnon Continuum

1 Magnon Bandwidth
Gap vs magnetic field // [001]

- gap increases in field, cross-over canted FM -> field-polarized
Conclusions

- In [001] field, Canted FM and Field-Polarized are smoothly connected, no phase transition, gap grows in field.

- At high field, see sharp magnons + 2-magnon continuum that grows rapidly upon lowering field, when overlap occurs, top magnon decays and lower magnon dispersions are strongly renormalized; continuum dominates at zero field.

- At high fields, sharp magnons captured well by spin waves of nn Hamiltonian with revised parameters, negligible $J_{zz}$ & dominant $J_{zz}$ almost on (mean-field) phase boundary between Canted FM and AFM $\Psi_{2,3}$, strongly frustrated.

arxiv: 1703:04506
Exchange Hamiltonian – global cubic axes

\[
J_{01} = \begin{pmatrix}
J_2 & J_4 & J_4 \\
-J_4 & J_1 & J_3 \\
-J_4 & J_3 & J_1
\end{pmatrix}
\]

\[
[J_1 \ J_2 \ J_3 \ J_4] = \begin{pmatrix}
-0.028 & -0.326 & -0.272 & 0.049
\end{pmatrix} \text{ meV}
\]

FM Ising coupling $J_2 S_x S_x$ (“Kitaev”-type)
+ “pseudo-dipolar” symmetric exchange $+ J_3 (S_y S_z + S_z S_y)$ $\Gamma$-term

- for spins $\parallel z$ 4/6 Kitaev bonds + 6 $\Gamma$ bonds create quantum dynamics