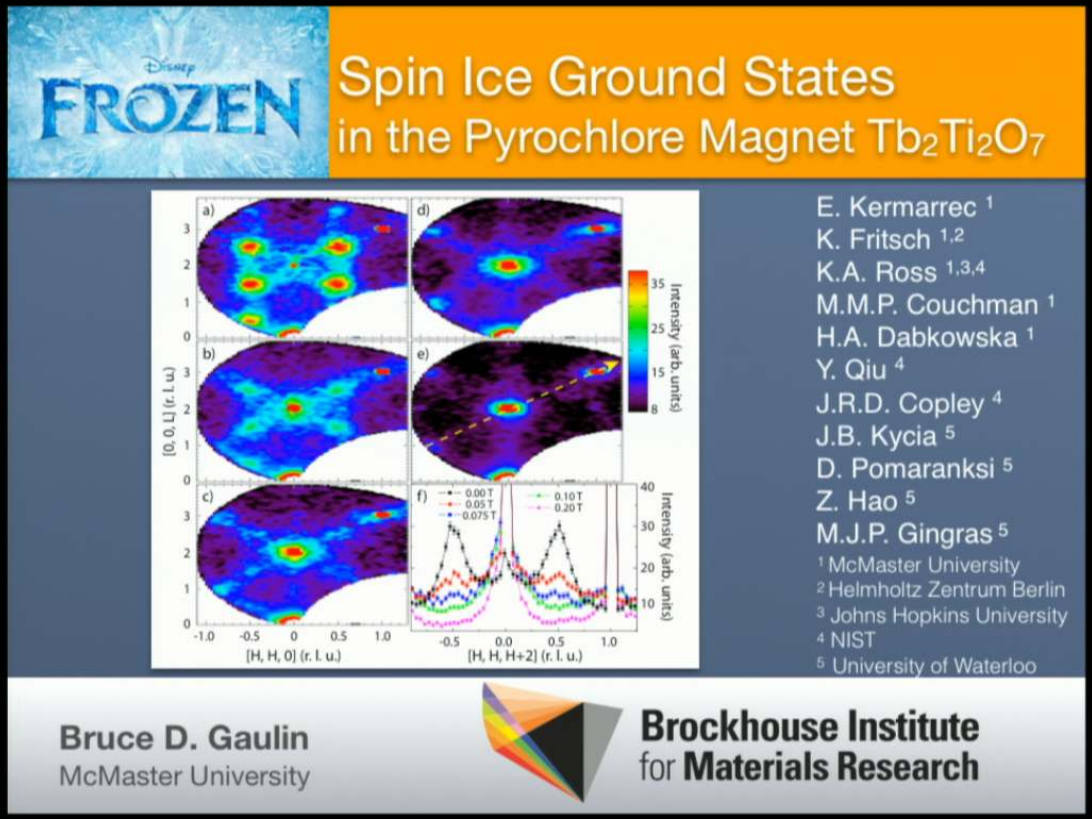
Title: Frozen Spin Ice Ground States in the Pyrochlore Magnet Tb2 Ti2 O7

Date: May 01, 2014 05:20 PM

URL: http://pirsa.org/14050019

Abstract: Tb2Ti2O7 was one of the first pyrochlore magnets to be studied as a candidate for a spin liquid or cooperative paramagnet, and its ground state has remained enigmatic for fifteen years. Recent time-of-flight neutron scattering studies have shown that it enters a glassy Spin Ice ground state, characterized by frozen short range order over about 8 conventional unit cells, and the formation of a ~ 0.08 meV gap in its spin excitation spectrum at the appropriate quasi-Bragg wave vectors. I will introduce the relevant Spin Ice physics background, and describe how the experiments are performed. The new H-T phase diagram for Tb2Ti2O7 in a [110] magnetic field will be presented. This shows that its frozen (i.e. glassy) Spin Ice ground state (at low temperature and zero field) and its conventional field-induced ordered phase (at low temperature and high fields) bracket the cooperative paramagnetic phase which generated the original interest in this fascinating magnet.


FROZEN
Spin Ice Ground States
in the Pyrochlore Magnet $Tb_2Ti_2O_7$
a)
b)
c)
d)
e)
f)
[0, 0, L] (r. l. u.)
[H, H, 0] (r. l. u.)
[H, H, H+2] (r. l. u.)
Intensity (arb. units)
0.00 T
0.05 T
0.075 T
0.10 T
0.20 T
E. Kermarrec [1]
K. Fritsch [1,2]
K.A. Ross [1,3,4]
M.M.P. Couchman [1]
H.A. Dabkowska [1]
Y. Qiu [4]
J.R.D. Copley [4]
J.B. Kycia [5]
D. Pomaranksi [5]
Z. Hao [5]
M.J.P. Gingras [5]
[1] McMaster University
[2] Helmholtz Zentrum Berlin
[3] Johns Hopkins University
[4] NIST
[5] University of Waterloo
Bruce D. Gaulin
McMaster University
Brockhouse Institute
for Materials Research

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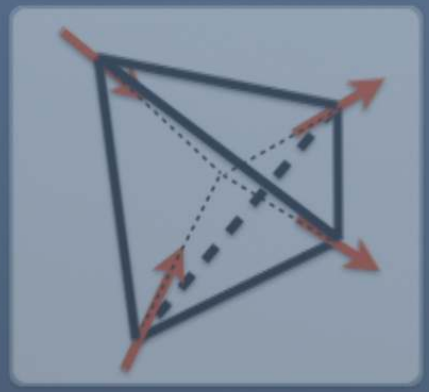
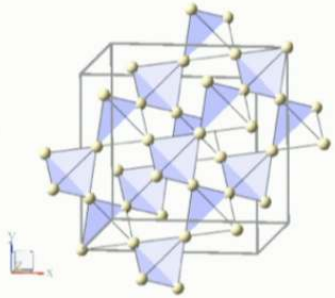
Geometric Frustration from Tetrahedra

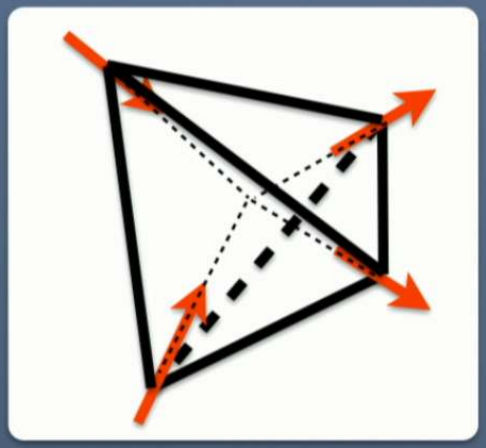


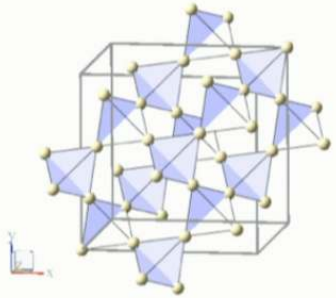


Pyrochlore

freedom of choice for each tetrahedron leads to a macroscopic degeneracy: **NO Long Range Order**

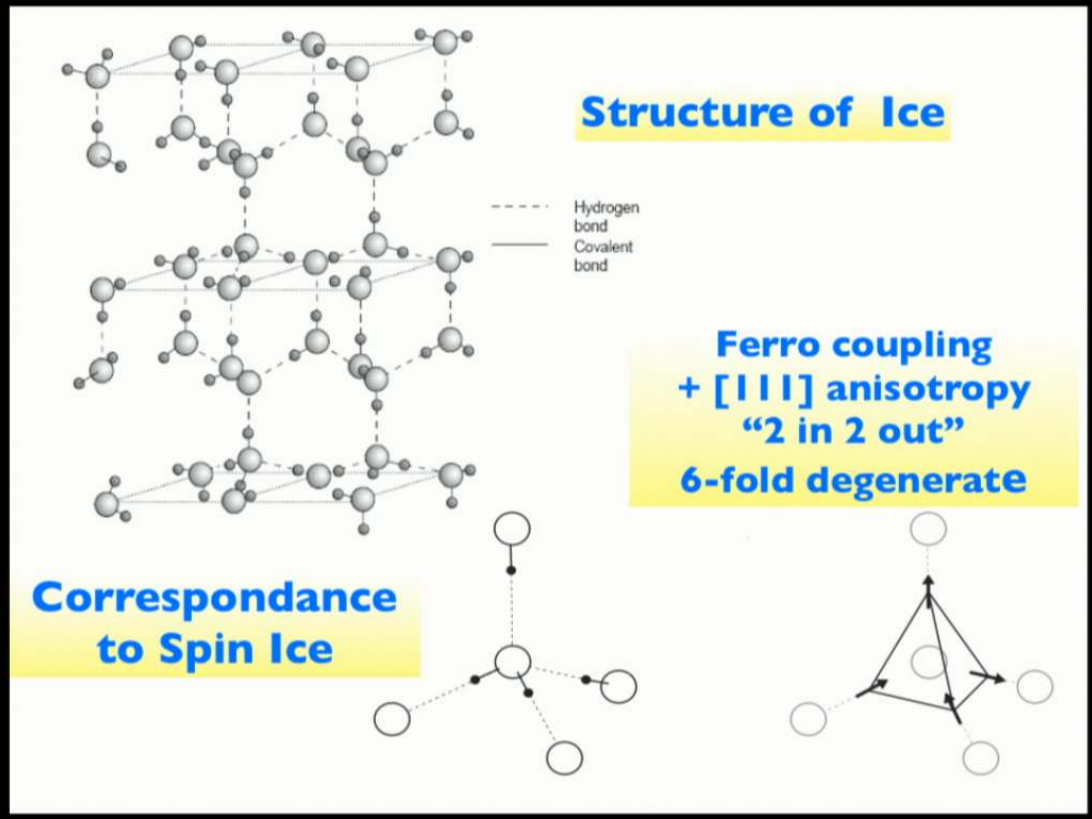
Geometric Frustration from Tetrahedra

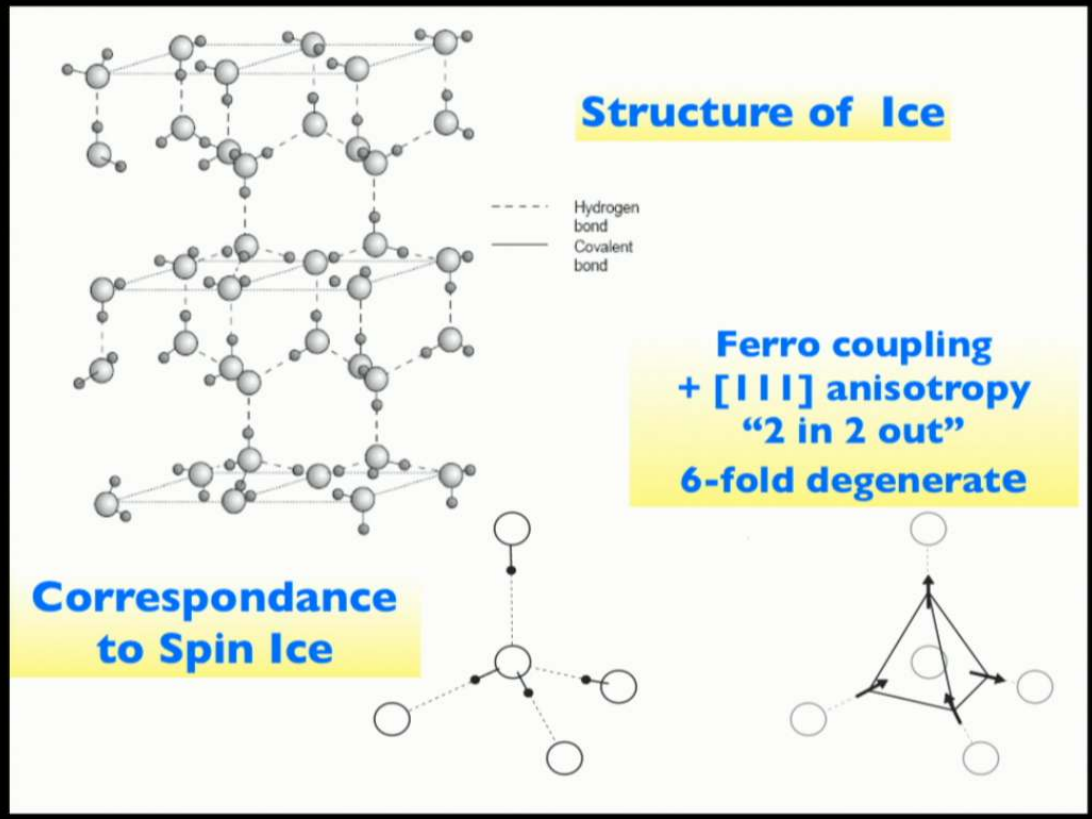




Pyrochlore

freedom of choice for each tetrahedron leads to a macroscopic degeneracy: **NO Long Range Order**


Structure of Ice
Hydrogen bond
Covalent bond
Ferro coupling
+ [111] anisotropy
"2 in 2 out"
6-fold degenerate
Correspondance to Spin Ice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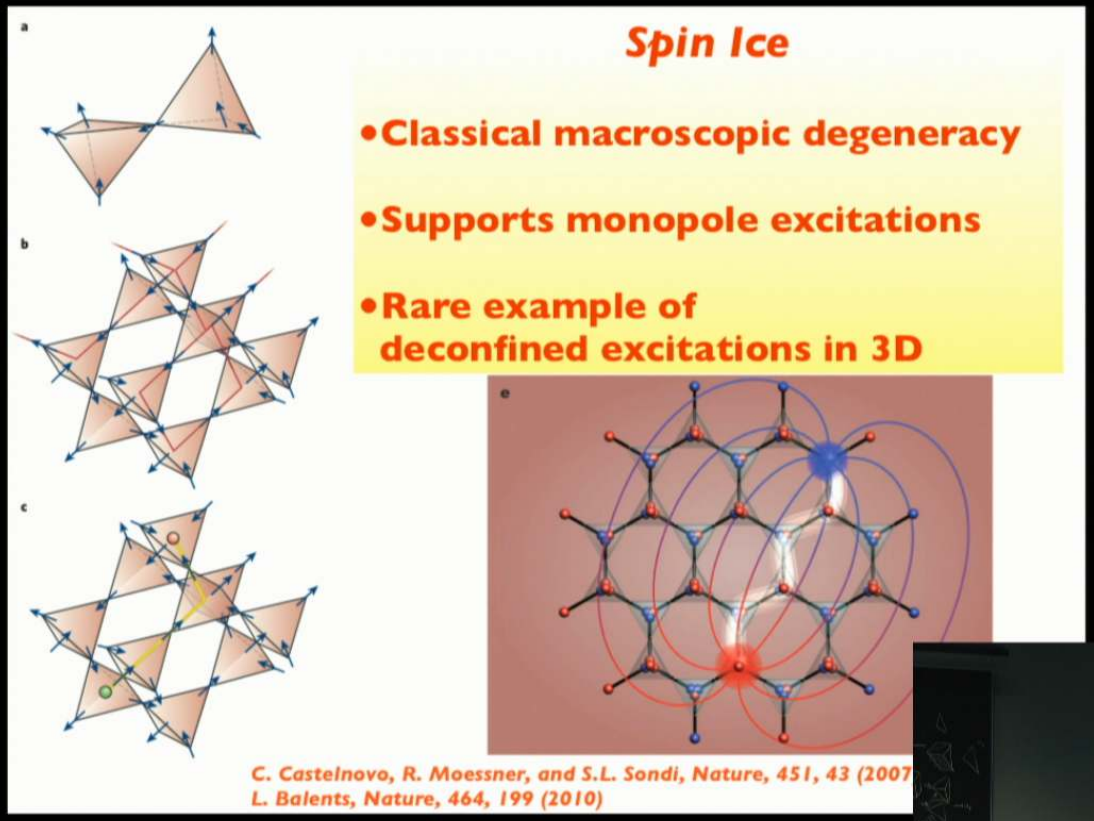
Spin Ice
•Classical macroscopic degeneracy
•Supports monopole excitations
•Rare example of deconfined excitations in 3D
C. Castelnovo, R. Moessner, and S.L. Sondi, Nature, 451, 43 (2007
L. Balents, Nature, 464, 199 (2010)

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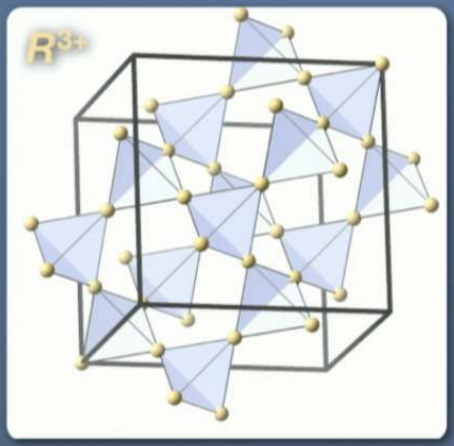
Real Pyrochlores

$R_2Ti_2O_7$

"Rare earth titanates"

Single-ion Anisotropy:
Crystal Field Effects

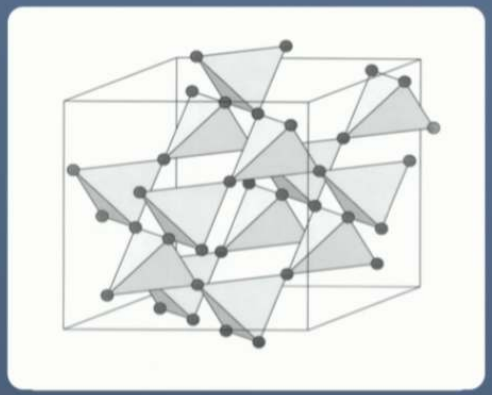
Exchange Anisotropy:
Spin orbit coupling

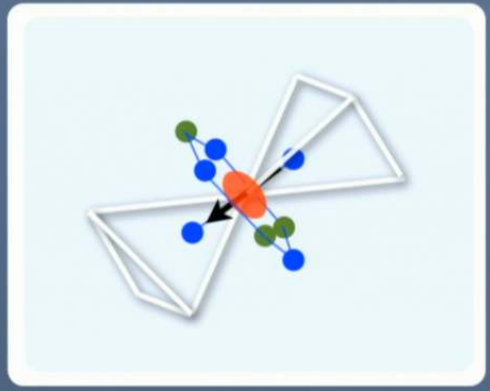


$R^{3+} =$

- Ho, Dy → Spin Ice, "Magnetic Monopoles", Coulomb phase
- Er → Long range order, Quantum critical point, Order by Disorder
- **Tb** → **Spin Liquid (?) ; Cooperative paramagnet (?)**
- Gd → Partial Ordering
- Yb → Quantum Spin Ice, Coulomb phase?

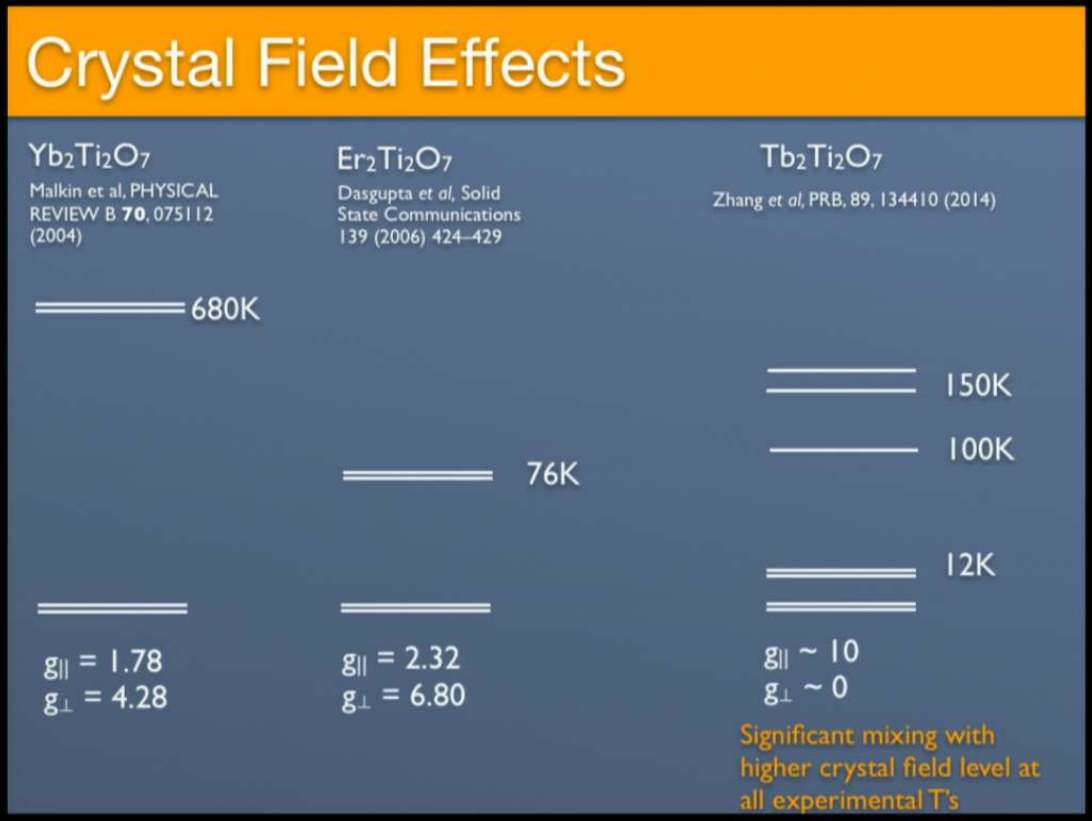
Crystal Field Environment at the RE Site





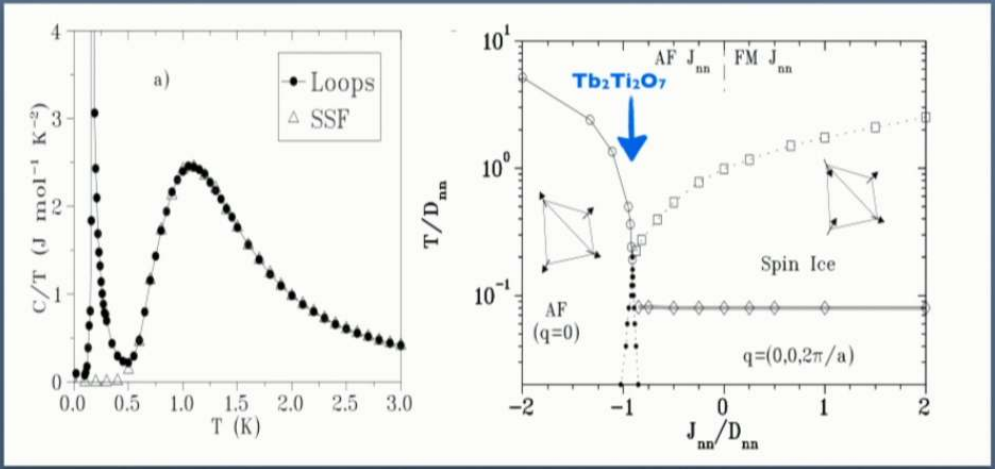
(2J+1) degenerate multiplet splits in presence of strong crystalline electric field from O^{2-} neighbours

Crystal Field Effects



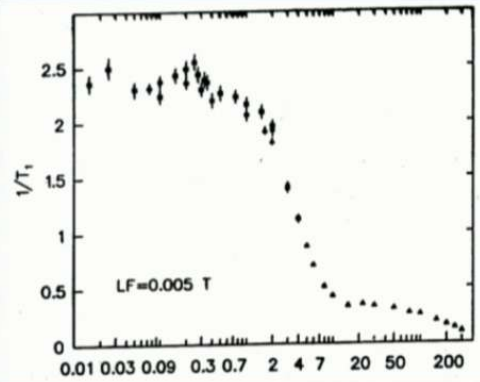
Minimal dipolar spin ice Hamiltonian

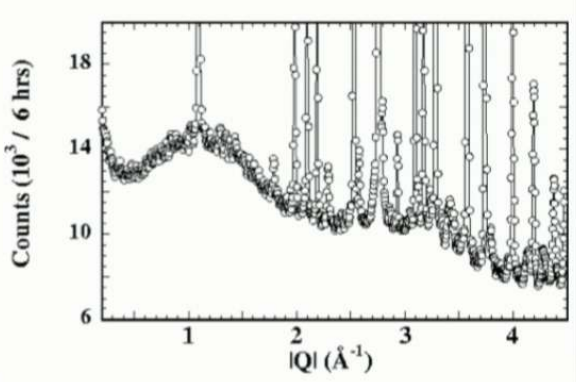
$$H = -J \sum_{\langle ij \rangle} \mathbf{S}_i^{z_i} \cdot \mathbf{S}_j^{z_j} + D r_{nn}^3 \sum_{i>j} \frac{\mathbf{S}_i^{z_i} \cdot \mathbf{S}_j^{z_j}}{|\mathbf{r}_{ij}|^3} - \frac{3(\mathbf{S}_i^{z_i} \cdot \mathbf{r}_{ij})(\mathbf{S}_j^{z_j} \cdot \mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^5}$$



R. G. Melko et al, Phys. Rev. Lett, 87, 067203, 2001

but, $Tb_2Ti_2O_7$ *doesn't* order at low T (~ 20 mK)



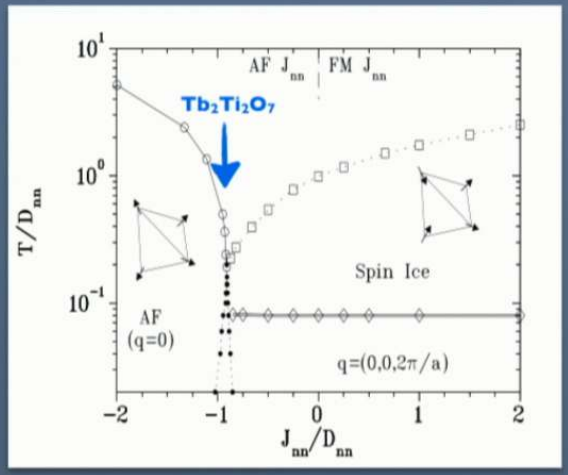


$1/T_1$ relaxation rate muSR

Low T powder neutron diffraction

J.S. Gardner et al, Phys. Rev. Lett, 82, 1012, 1999

Two Scenarios:
I: Virtual CEF transitions induce Quantum Spin Ice



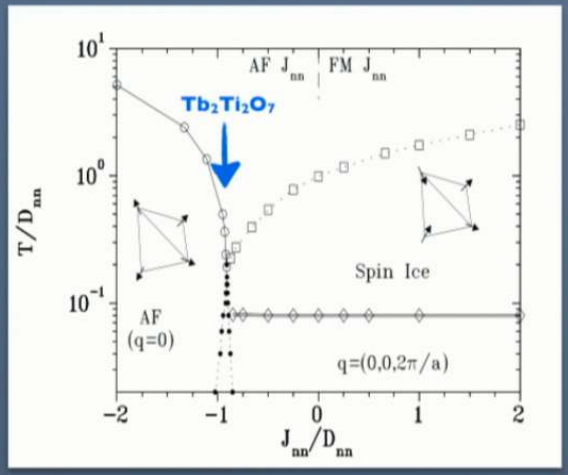
150K

100K

Two Scenarios:
I: Virtual CEF transitions induce Quantum Spin Ice

$Tb_2Ti_2O_7$

Zhang *et al*, PRB, 89, 134410 (2014)



$g_{||} \sim 10$

$g_{\perp} \sim 0$

Significant mixing with higher crystal field level at all experimental T's

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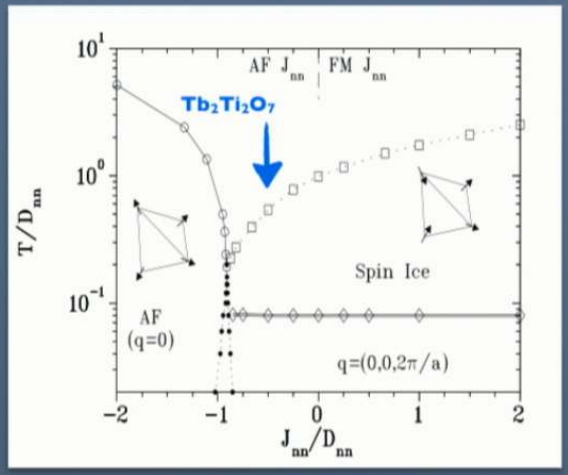

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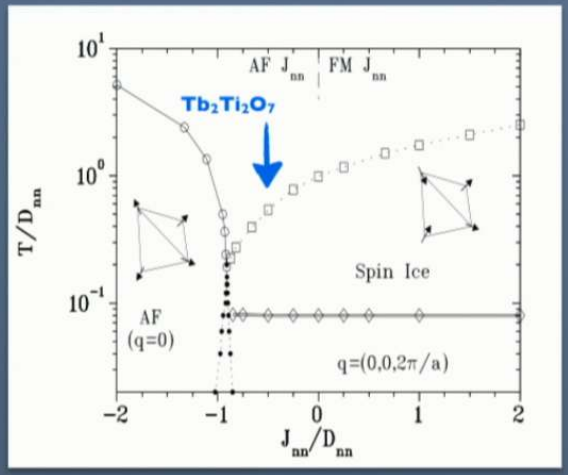

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Significant mixing with higher crystal field level at all experimental T's

Two Scenarios:
II: Tb^{3+} ground state is a non-magnetic singlet

$Tb_2Ti_2O_7$

Zhang *et al*, PRB, 89, 134410 (2014)

Doublet nature of Tb^{3+} CEF eigenvalues *not* protected by Kramer's theorem

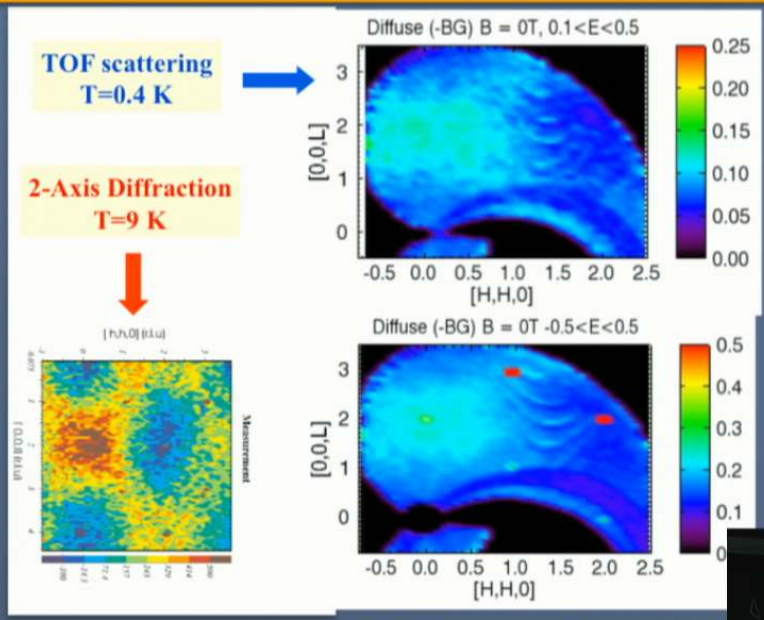
Tb^{3+} ground state could be non-magnetic singlet

Bonville et al, Phys. Rev. B, 89, 085115, 2014

In due course, single crystals were grown and measured

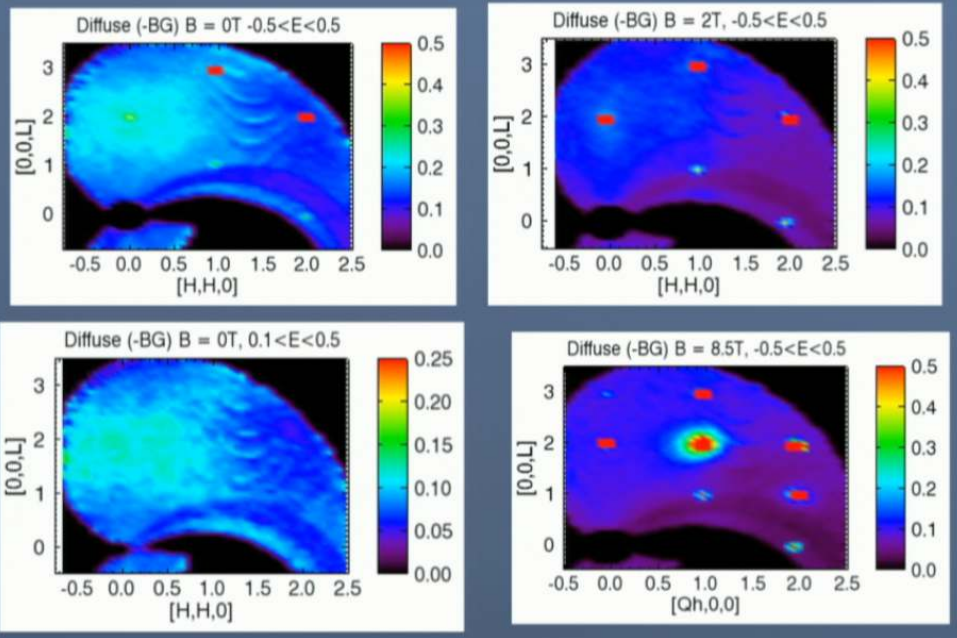
TOF scattering
T=0.4 K

2-Axis Diffraction
T=9 K



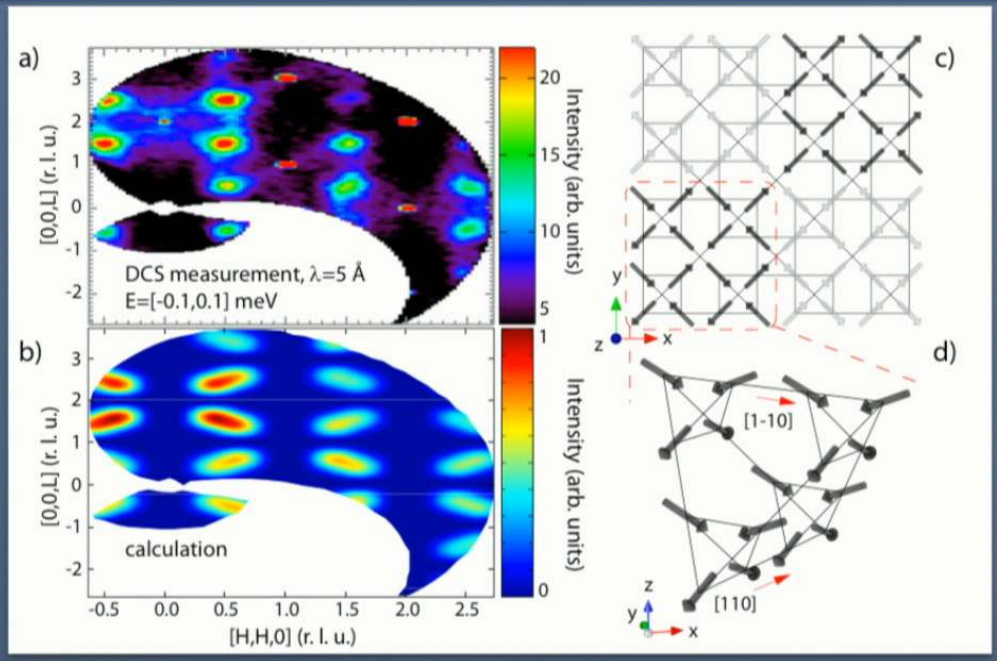
J.S. Gardner et al, Phys. Rev. B, 64, 224416, 2001
K.C. Rule et al, Phys. Rev. Lett, 96, 177201, 2006

Ordered Phase Appears on Application of a 110 Magnetic Field



K.C. Rule et al, Phys. Rev. Lett, 96, 177201, 2006

Unexpected observation of Bragg-like 1/2 1/2 1/2 reflections for T ~0.1 K! Modelled on AF frozen spin ice



K. Fritsch et al, Phys. Rev. B, 87, 490410, 2013

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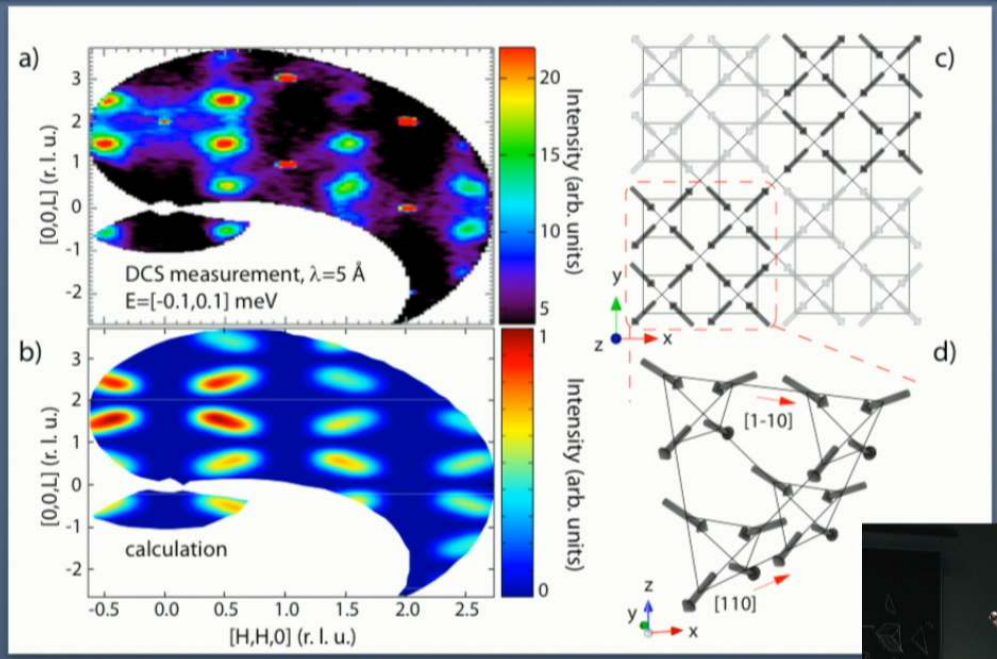

K. Fritsch et al, Phys. Rev. B, 87, 490410, 2013

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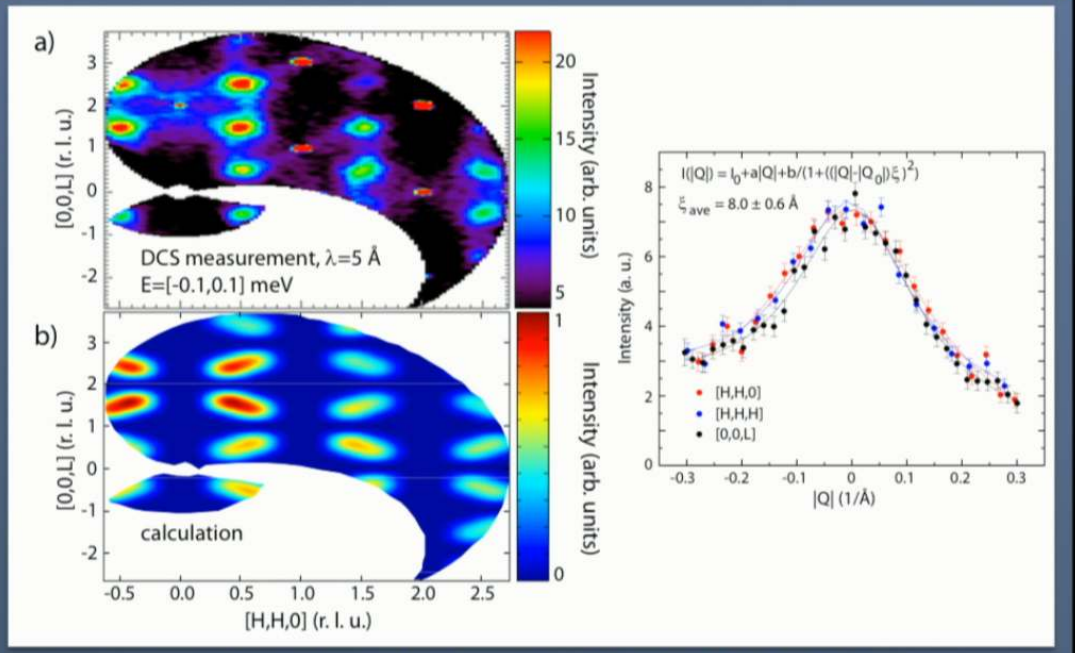
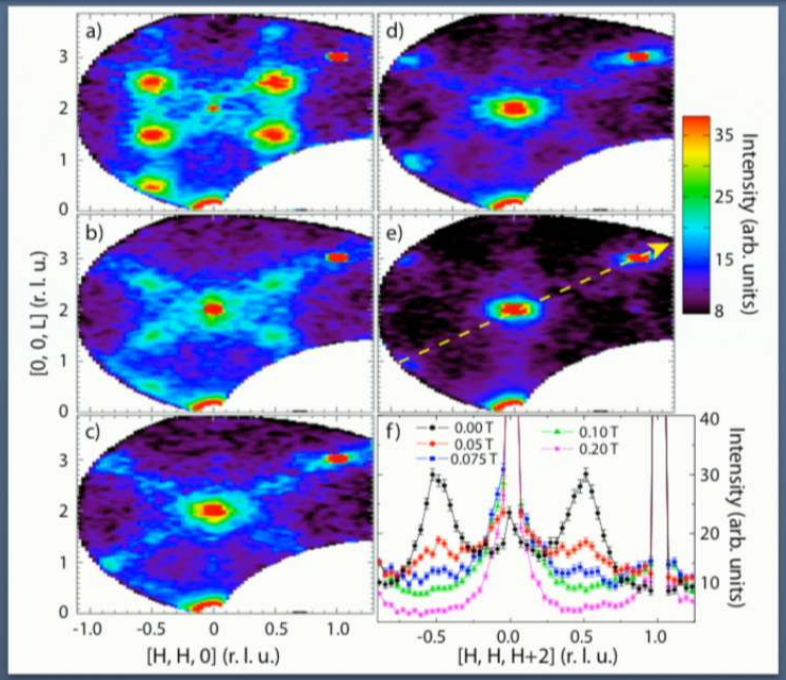
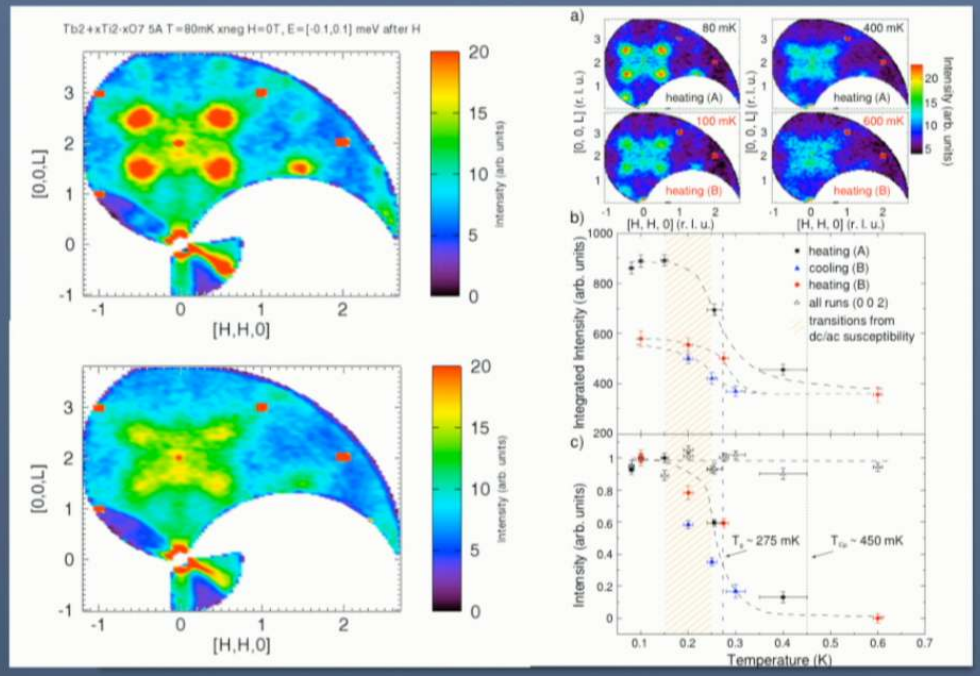


K. Fritsch et al, Phys. Rev. B, 87, 490410, 2013

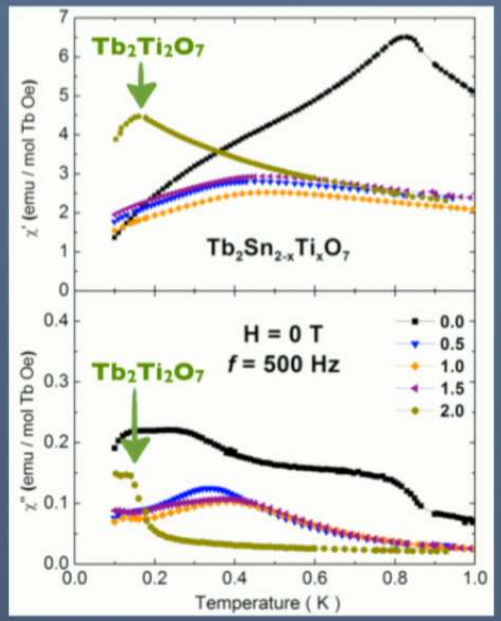
Frozen Spin Ice Structure Polarized by ~ 0.075 T (110) Magnetic Field

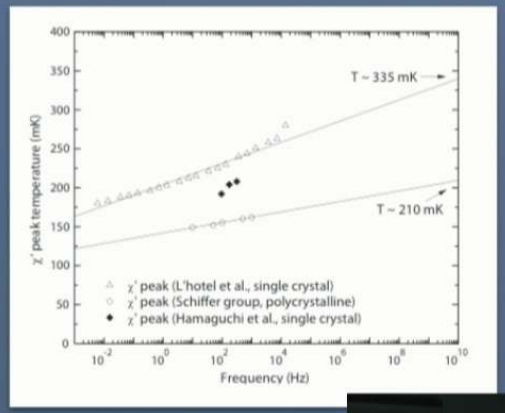


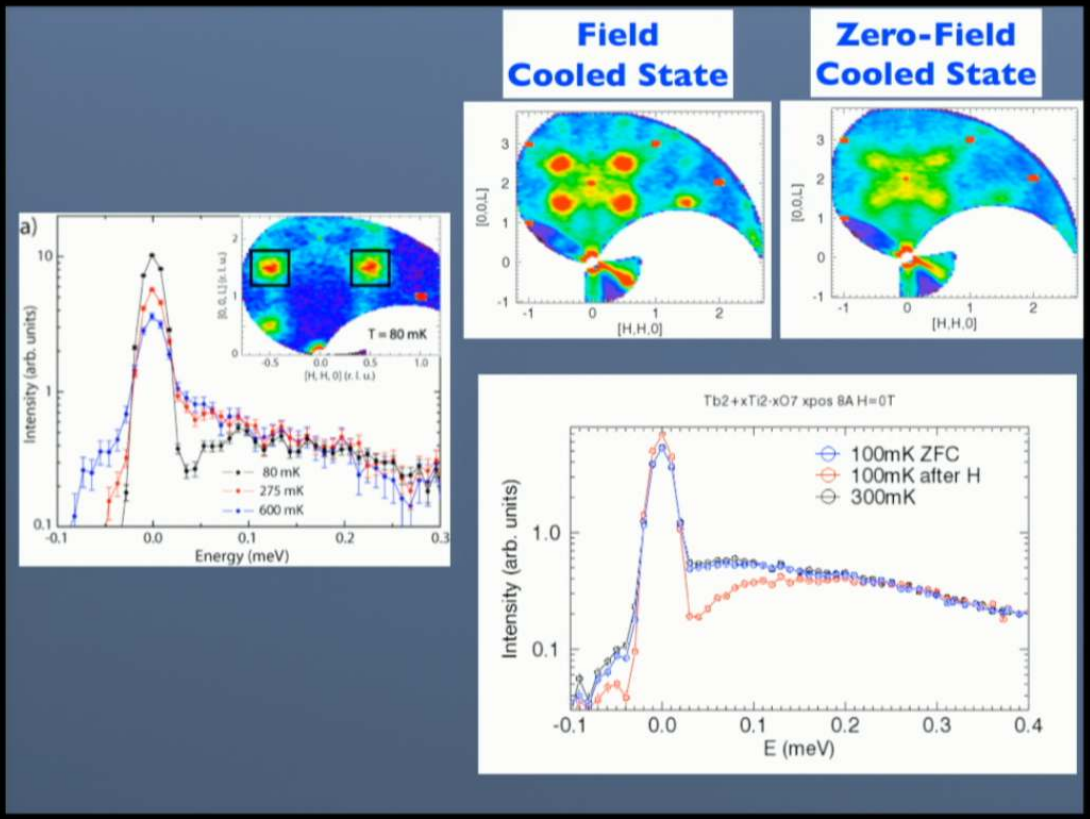
Low T Spin Ice Phase is History Dependent

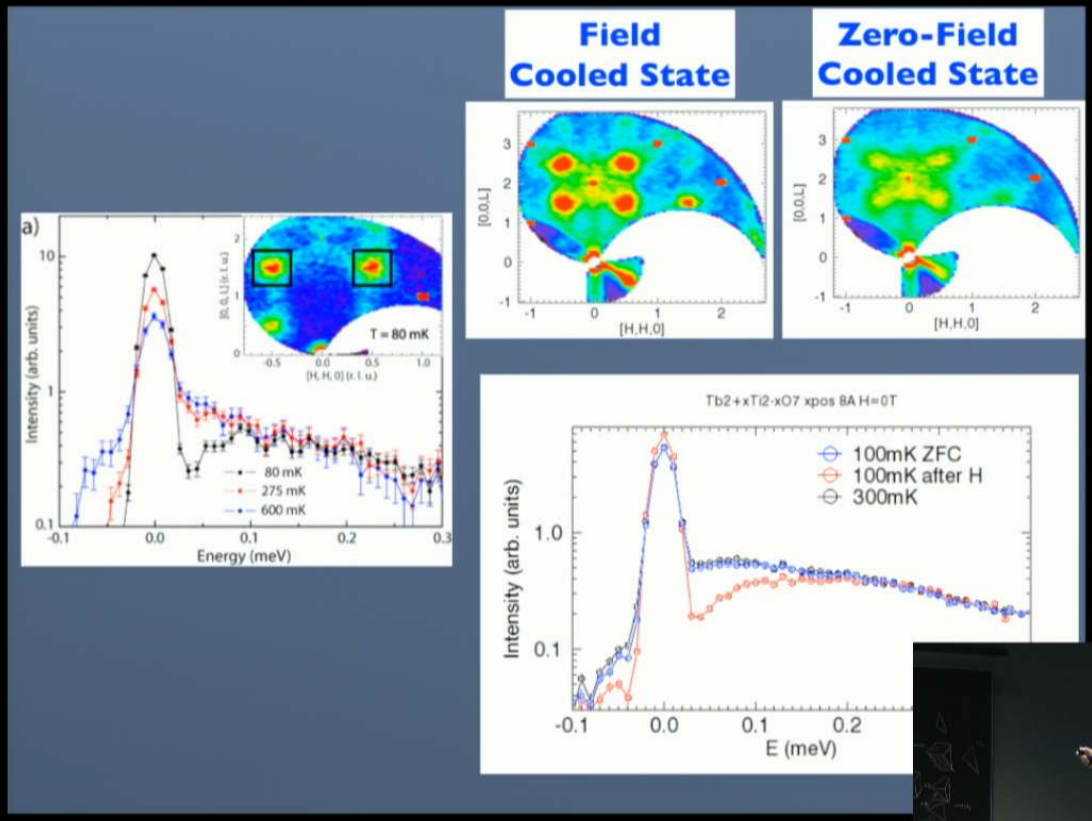


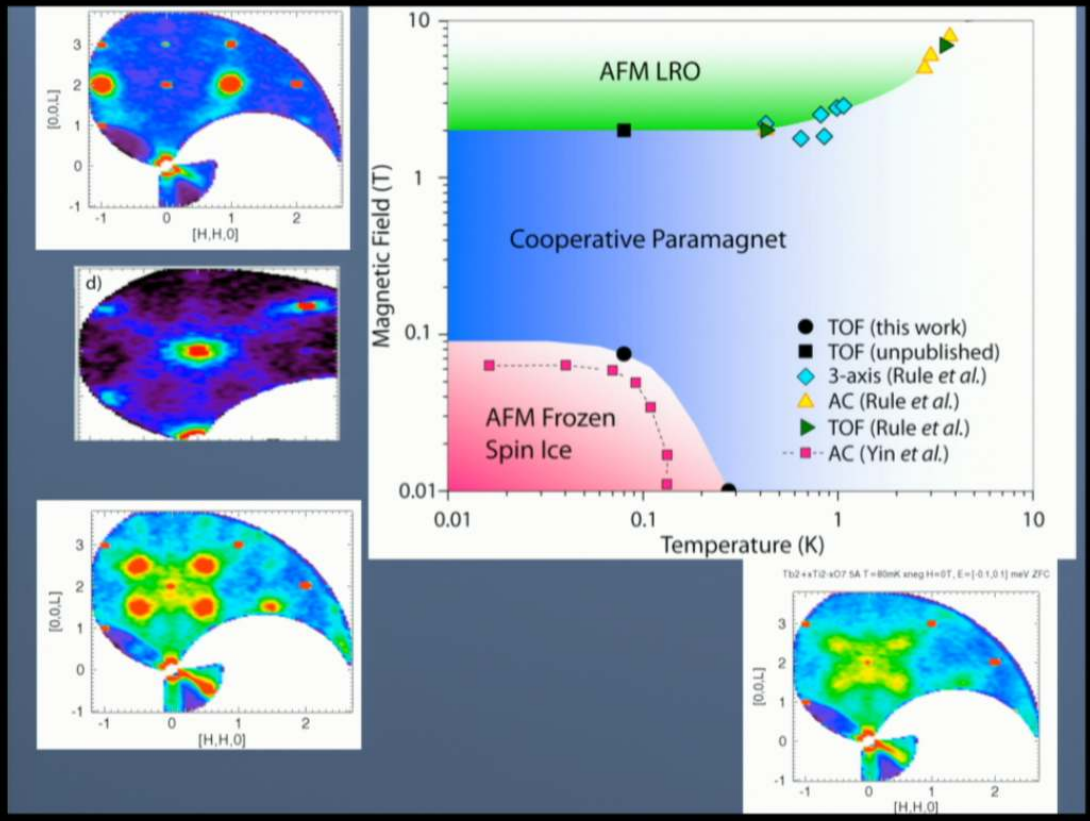
Glassy-behaviour seen below ~ 0.25 K in $Tb_2Ti_2O_7$

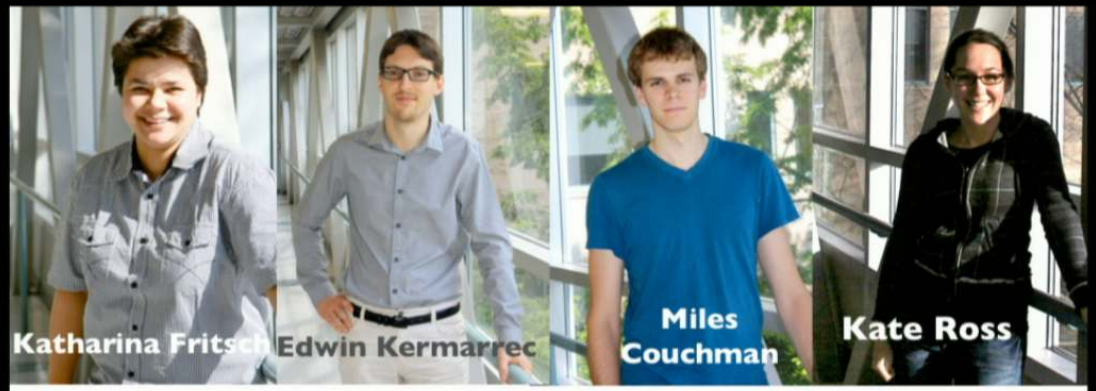





Field Cooled State
Zero-Field Cooled State
[0,0,L]
[H,H,0]
a)
Intensity (arb. units)
Energy (meV)
T = 80 mK
80 mK
275 mK
600 mK
Tb2+xTi2-xO7 xpos 8A H=0T
100mK ZFC
100mK after H
300mK
Intensity (arb. units)
E (meV)


Field Cooled State
Zero-Field Cooled State
a)
T = 80 mK
Intensity (arb. units)
Energy (meV)
80 mK
275 mK
600 mK
[0,0,L]
[H,H,0]
Tb2+xTi2-xO7 xpos 8A H=0T
100mK ZFC
100mK after H
300mK
Intensity (arb. units)
E (meV)


[0,0,L]
[H,H,0]
d)
AFM LRO
Cooperative Paramagnet
AFM Frozen
Spin Ice
Magnetic Field (T)
Temperature (K)
TOF (this work)
TOF (unpublished)
3-axis (Rule et al.)
AC (Rule et al.)
TOF (Rule et al.)
AC (Yin et al.)



Conclusions

- *TOF techniques well suited to exotic magnetism as it measures very effectively across wide dynamic range in Q and energy*
- *$Tb_2Ti_2O_7$ displays frozen AF spin ice state at low temperatures (< 0.25 K) and fields (< 0.075 T)*
- *FC state shows 1/2 1/2 1/2 short range (~ 8 unit cell) correlations and an ~ 0.1 meV ~ 1 K gap*
- *ZFC state is gapless and shows "checkerboard" correlations*

EXIT
EXIT

EXIT