

Title: Frozen Spin Ice Ground States in the Pyrochlore Magnet Tb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>

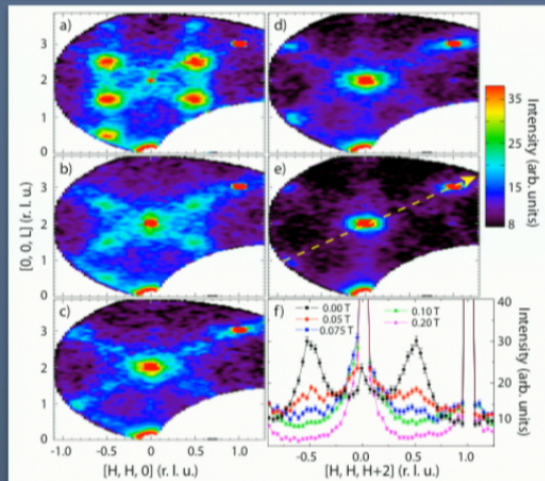
Date: May 01, 2014 05:20 PM

URL: <http://pirsa.org/14050019>

Abstract: Tb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> 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 Tb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> 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.



# Spin Ice Ground States in the Pyrochlore Magnet $Tb_2Ti_2O_7$



E. Kermarrec <sup>1</sup>  
K. Fritsch <sup>1,2</sup>  
K.A. Ross <sup>1,3,4</sup>  
M.M.P. Couchman <sup>1</sup>  
H.A. Dabkowska <sup>1</sup>  
Y. Qiu <sup>4</sup>  
J.R.D. Copley <sup>4</sup>  
J.B. Kycia <sup>5</sup>  
D. Pomaranski <sup>5</sup>  
Z. Hao <sup>5</sup>  
M.J.P. Gingras <sup>5</sup>  
<sup>1</sup> McMaster University  
<sup>2</sup> Helmholtz Zentrum Berlin  
<sup>3</sup> Johns Hopkins University  
<sup>4</sup> NIST  
<sup>5</sup> University of Waterloo

**Bruce D. Gaulin**  
McMaster University

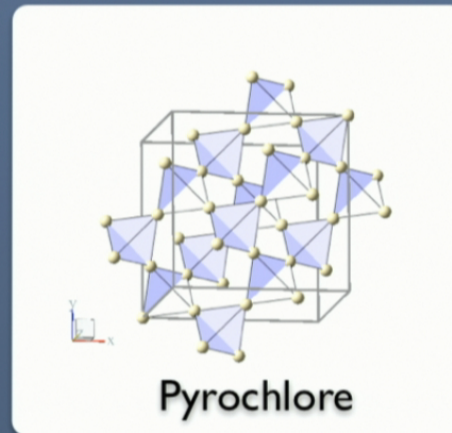
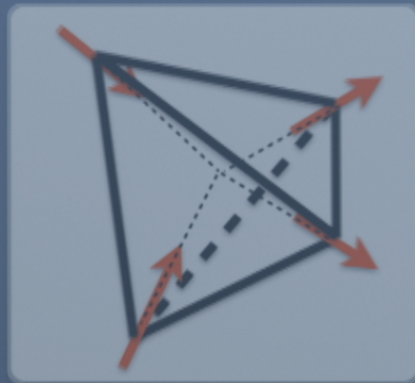


**Brockhouse Institute**  
for **Materials Research**





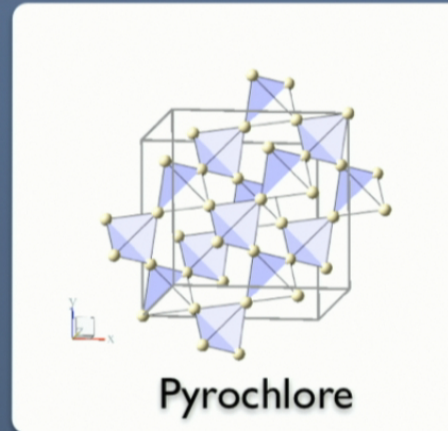
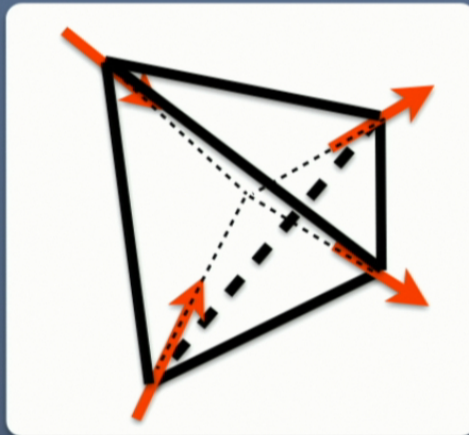
# Geometric Frustration from Tetrahedra



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

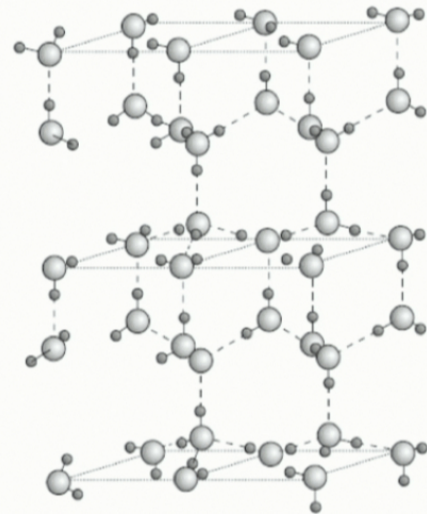


## Geometric Frustration from Tetrahedra



Pyrochlore

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

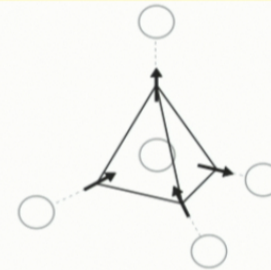
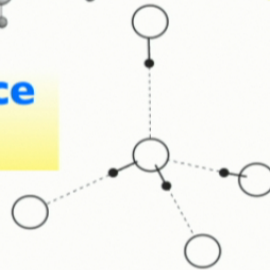


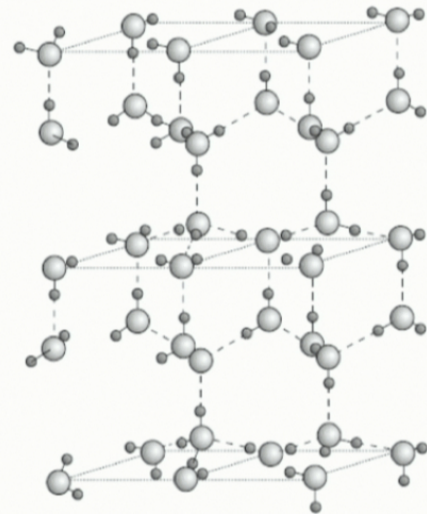
## Structure of Ice

--- Hydrogen bond  
 — Covalent bond

**Ferro coupling  
 + [111] anisotropy  
 "2 in 2 out"  
 6-fold degenerate**

## Correspondance to Spin Ice



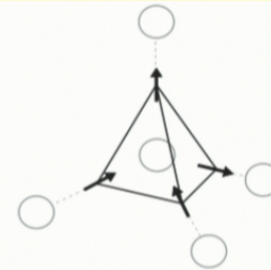


## Structure of Ice

--- Hydrogen bond  
 — Covalent bond

**Ferro coupling  
 + [111] anisotropy  
 "2 in 2 out"  
 6-fold degenerate**

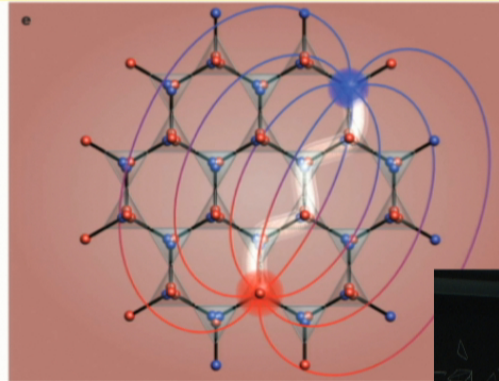
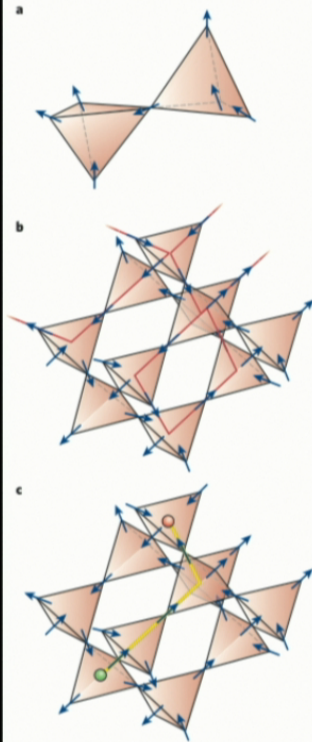
## Correspondance to Spin Ice





## Spin Ice

- Classical macroscopic degeneracy
- Supports monopole excitations
- Rare example of deconfined excitations in 3D



C. Castelnovo, R. Moessner, and S.L. Sondhi, *Nature*, 451, 43 (2007)  
L. Balents, *Nature*, 464, 199 (2010)







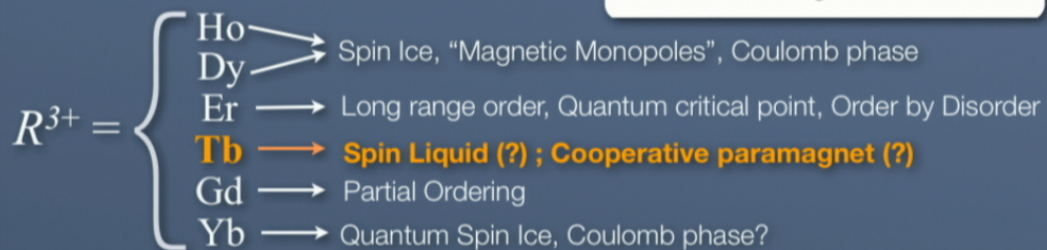
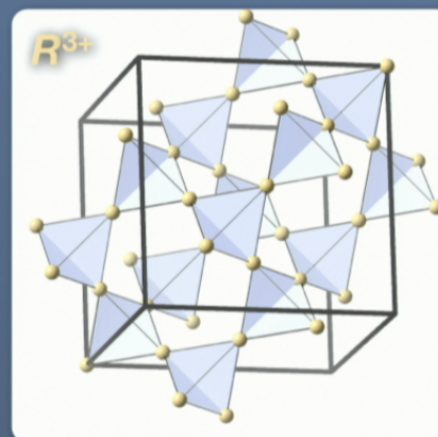
# Real Pyrochlores



“Rare earth titanates”

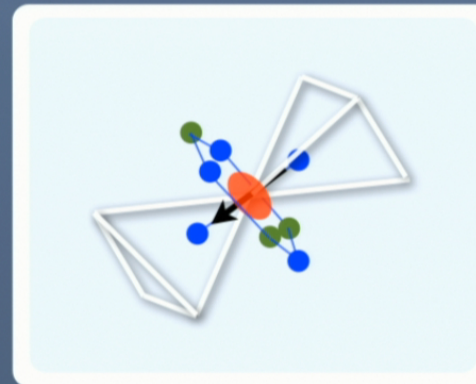
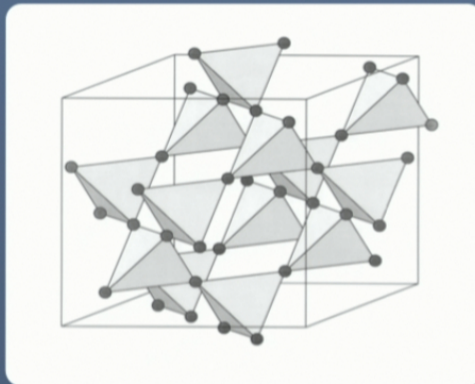
Single-ion Anisotropy:  
Crystal Field Effects

Exchange Anisotropy:  
Spin orbit coupling



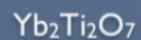


## Crystal Field Environment at the RE Site



( $2J+1$ ) degenerate multiplet splits in presence of strong crystalline electric field from O<sup>2-</sup> neighbours

# Crystal Field Effects

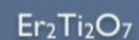


Malkin et al, PHYSICAL  
REVIEW B **70**, 075112  
(2004)

==== 680K

====

$$g_{\parallel} = 1.78$$
$$g_{\perp} = 4.28$$



Dasgupta et al, Solid  
State Communications  
139 (2006) 424–429

==== 76K

====

$$g_{\parallel} = 2.32$$
$$g_{\perp} = 6.80$$



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

==== 150K

==== 100K

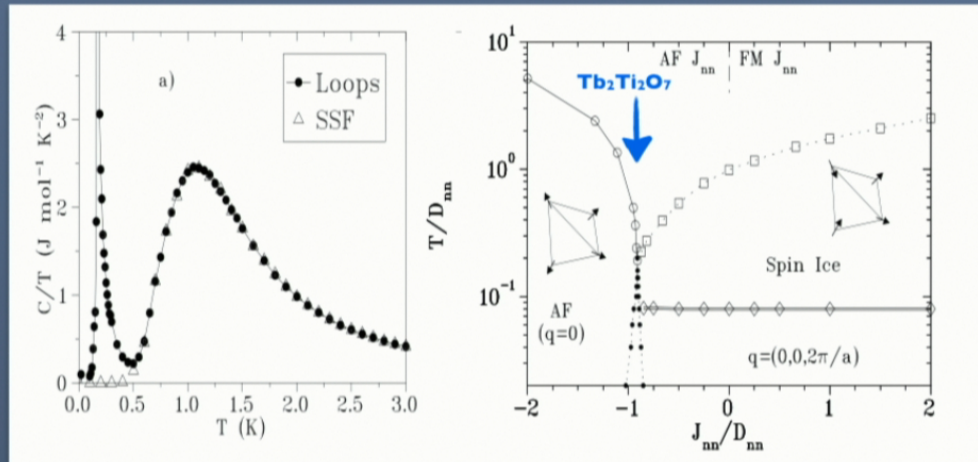
==== 12K

$$g_{\parallel} \sim 10$$
$$g_{\perp} \sim 0$$

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

## Minimal dipolar spin ice Hamiltonian

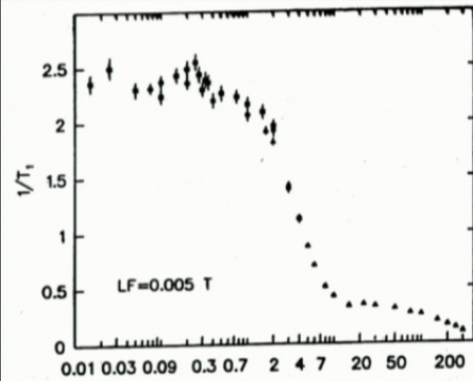
$$H = -J \sum_{\langle ij \rangle} \mathbf{S}_i^{z_i} \cdot \mathbf{S}_j^{z_j} + D r_{nn}^3 \sum_{i>j} \frac{\mathbf{S}_i^{z_i} \cdot \mathbf{S}_j^{z_j}}{|\mathbf{r}_{ij}|^3} - \frac{3(\mathbf{S}_i^{z_i} \cdot \mathbf{r}_{ij})(\mathbf{S}_j^{z_j} \cdot \mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^5}$$



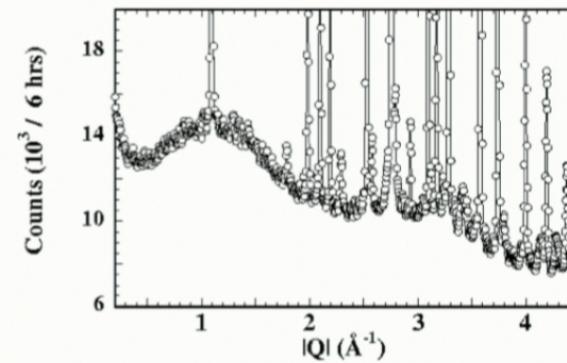
R. G. Melko et al, Phys. Rev. Lett, 87, 067203, 2001



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



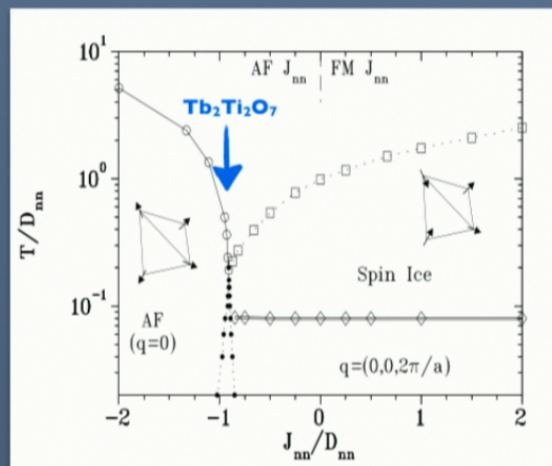
**$1/T_1$  relaxation rate  
muSR**



**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



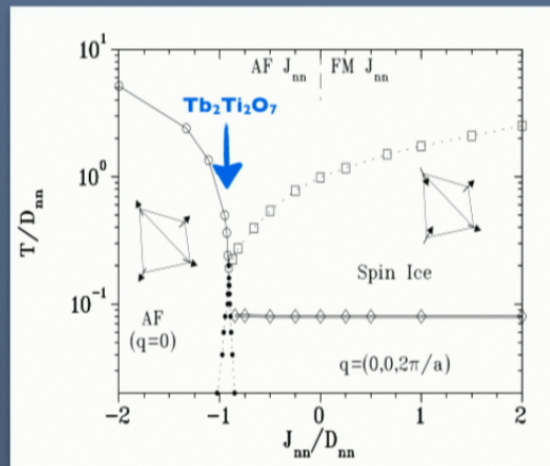
==== 150K  
==== 100K



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

$\text{Tb}_2\text{Ti}_2\text{O}_7$

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



==== 150K

==== 100K

==== 12K

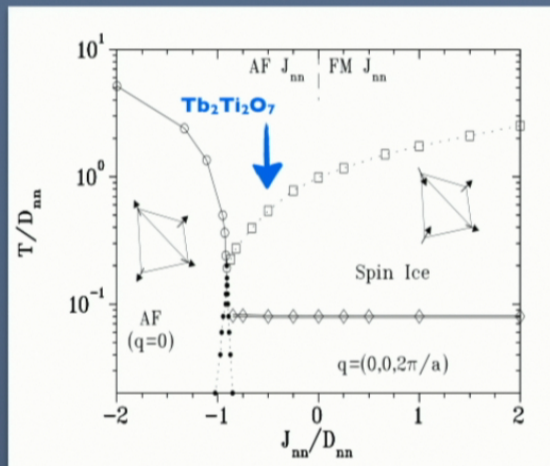
$g_{||} \sim 10$

$g_{\perp} \sim 0$

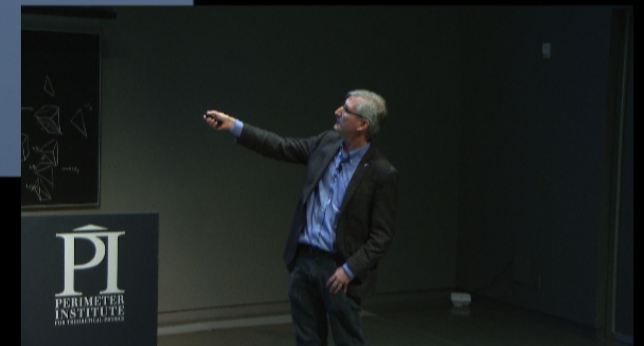
Significant mixing with  
higher crystal field level at  
all experimental T's



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



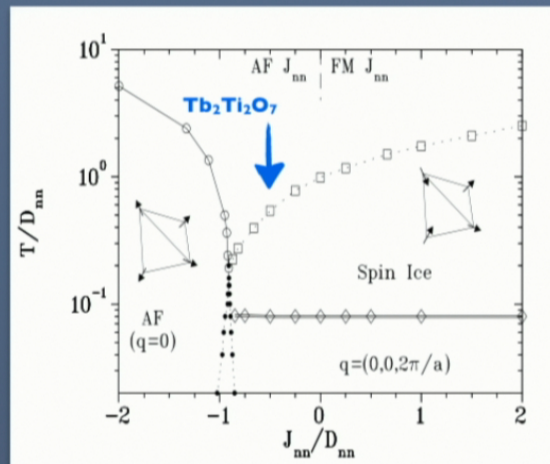
==== 150K  
==== 100K



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

$\text{Tb}_2\text{Ti}_2\text{O}_7$

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



==== 150K

==== 100K

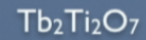
==== 12K

$g_{||} \sim 10$

$g_{\perp} \sim 0$

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

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



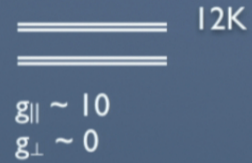
Zhang et al, PRB, 89, 134410 (2014)

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

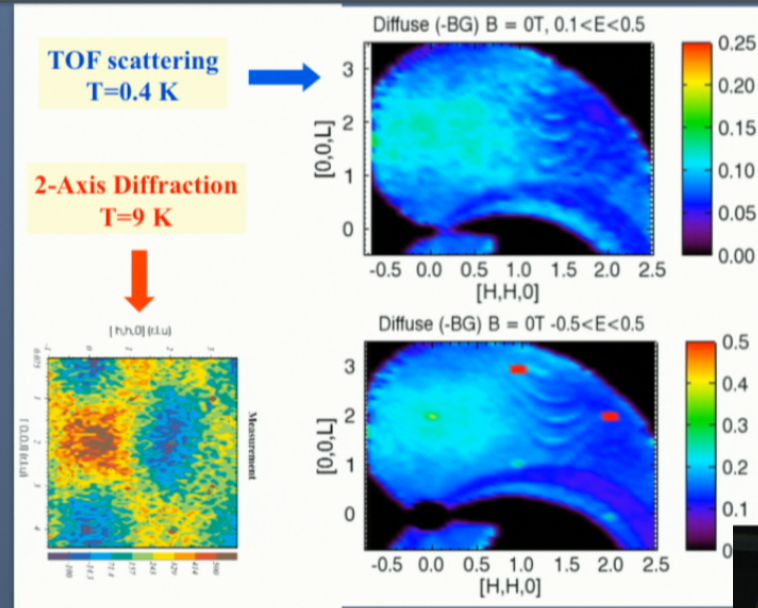
< 0.5 K



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



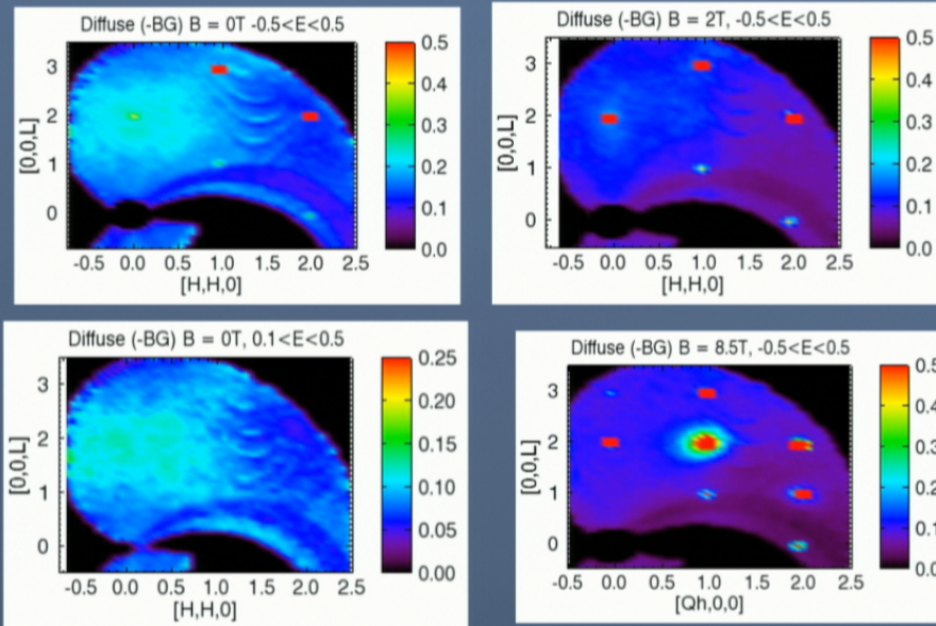
## In due course, single crystals were grown and measured



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

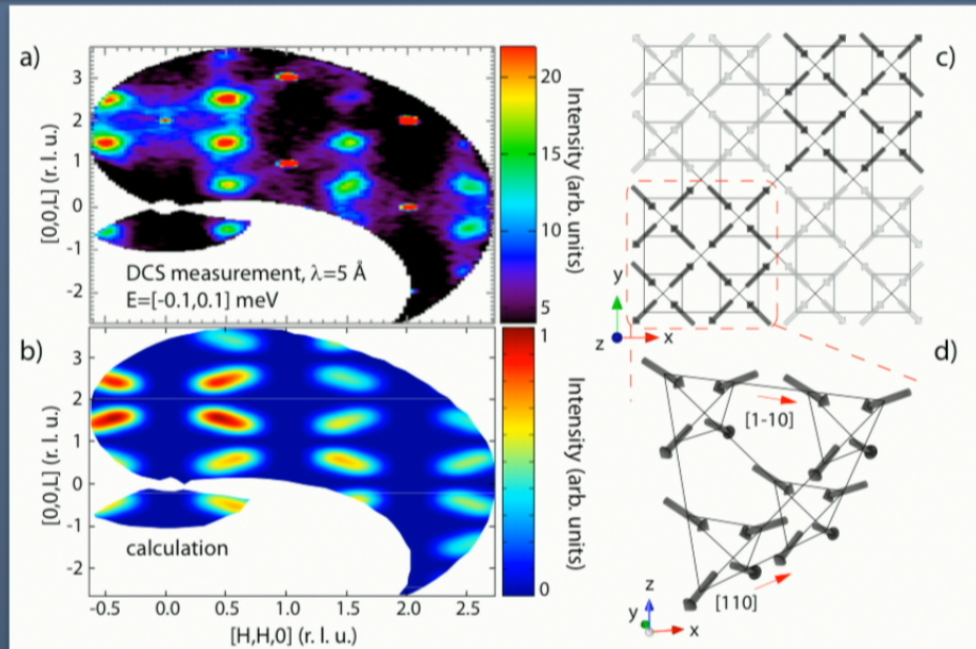


## Ordered Phase Appears on Application of a $\parallel 10$ Magnetic Field



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

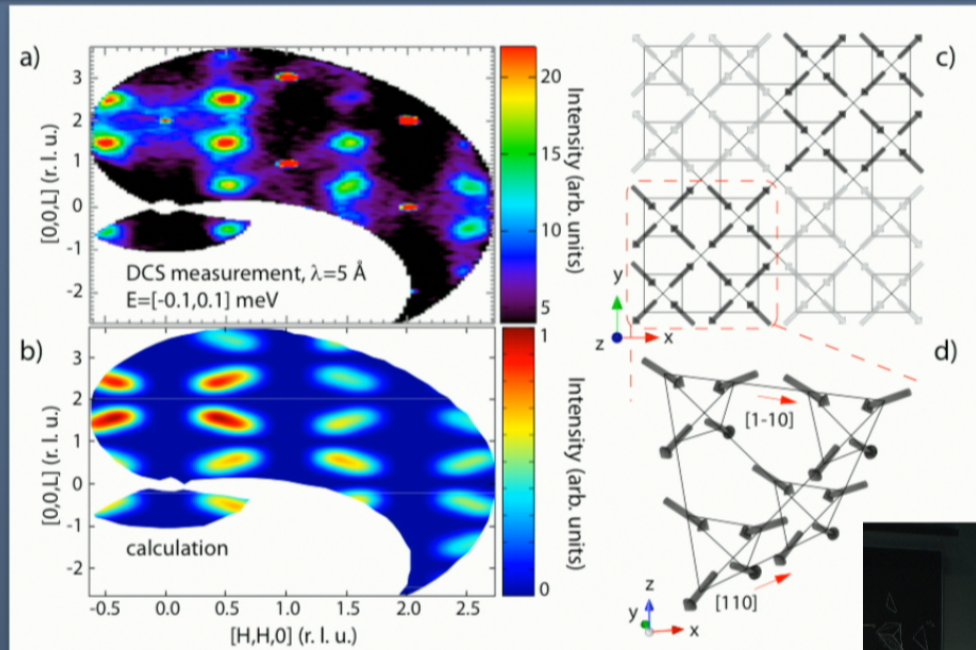
**Unexpected observation of Bragg-like  $1/2 \ 1/2 \ 1/2$  reflections for  $T \sim 0.1$  K! Modelled on AF frozen spin ice**



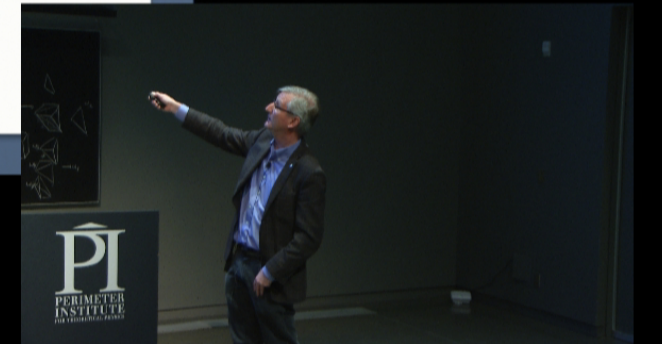
**K. Fritsch et al, Phys. Rev. B, 87, 490410, 2013**



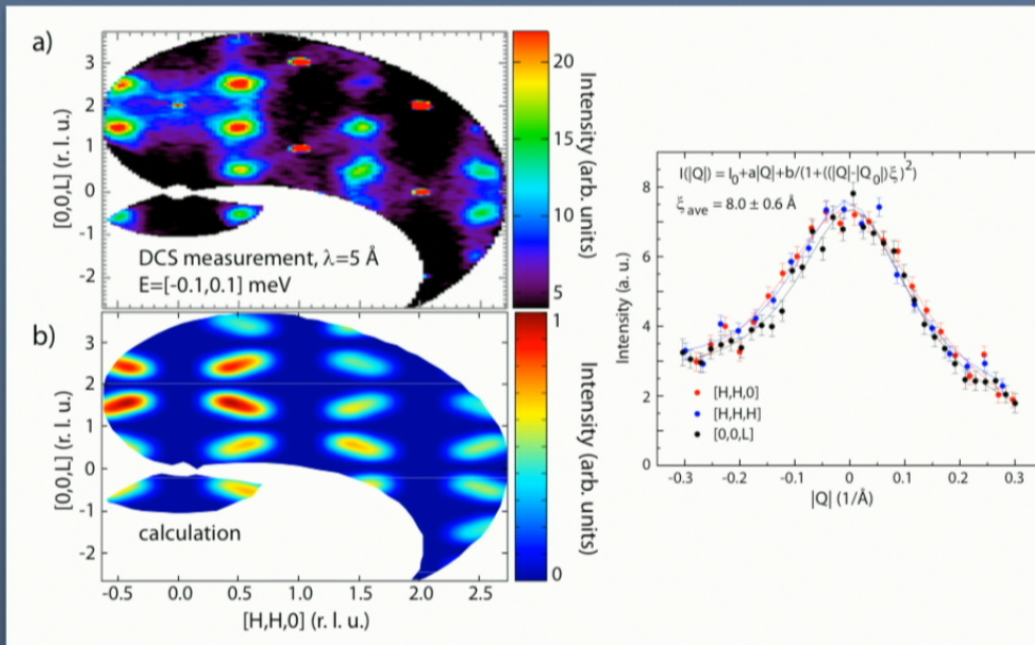
## Unexpected observation of Bragg-like $1/2 \ 1/2 \ 1/2$ reflections for $T \sim 0.1 \text{ K}$ ! Modelled on AF frozen spin ice



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

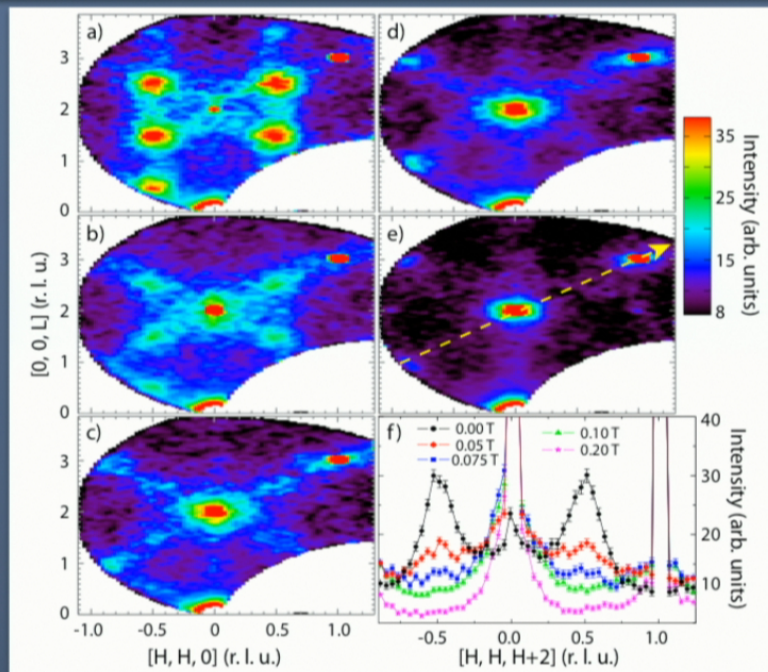


**Unexpected observation of Bragg-like  $1/2 \ 1/2 \ 1/2$  reflections for  $T \sim 0.1$  K! Modelled on AF frozen spin ice**



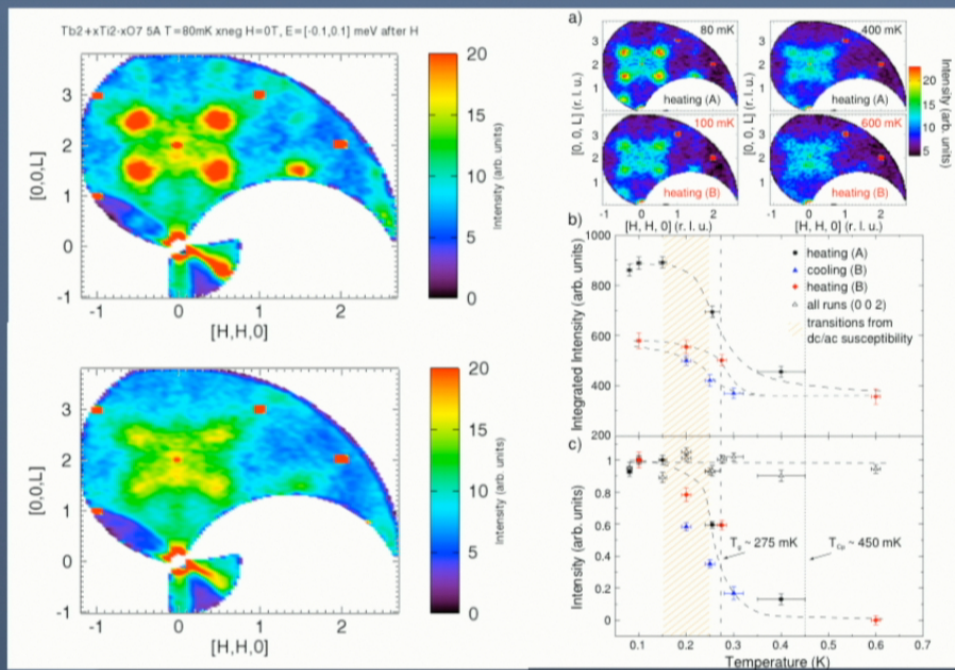
**K. Fritsch et al, Phys. Rev. B, 87, 490410, 2013**

## Frozen Spin Ice Structure Polarized by $\sim 0.075$ T (110) Magnetic Field

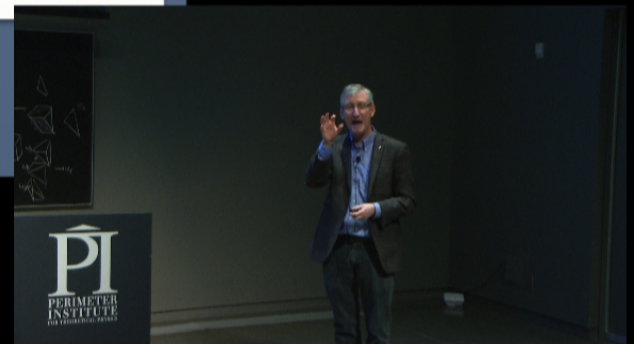
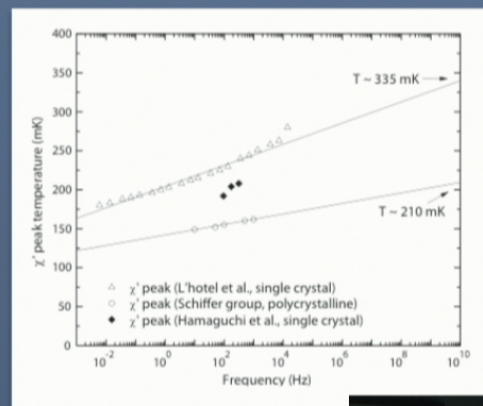
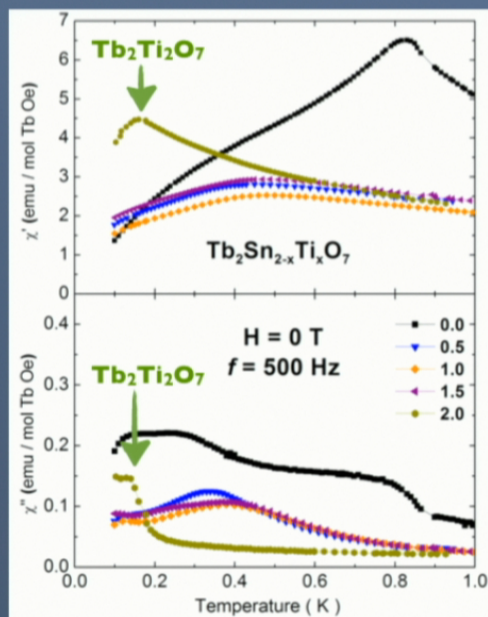


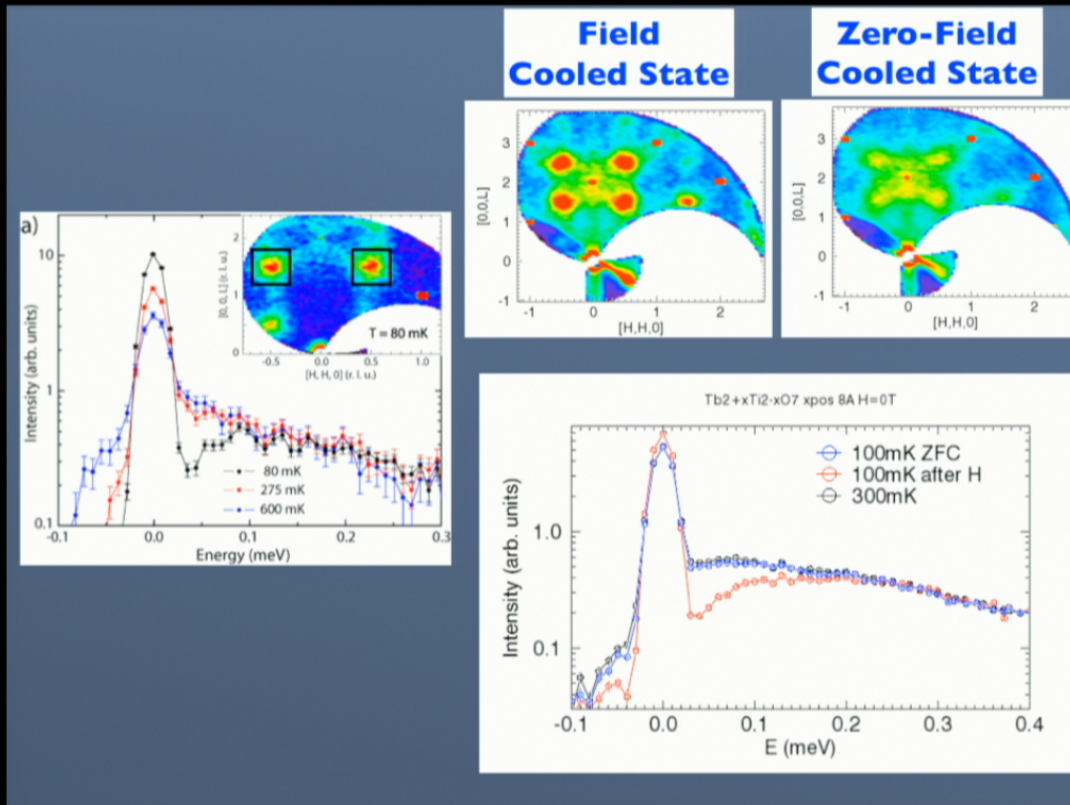


## Low T Spin Ice Phase is History Dependent

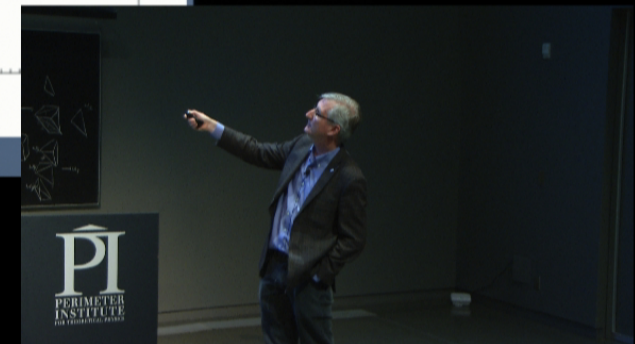
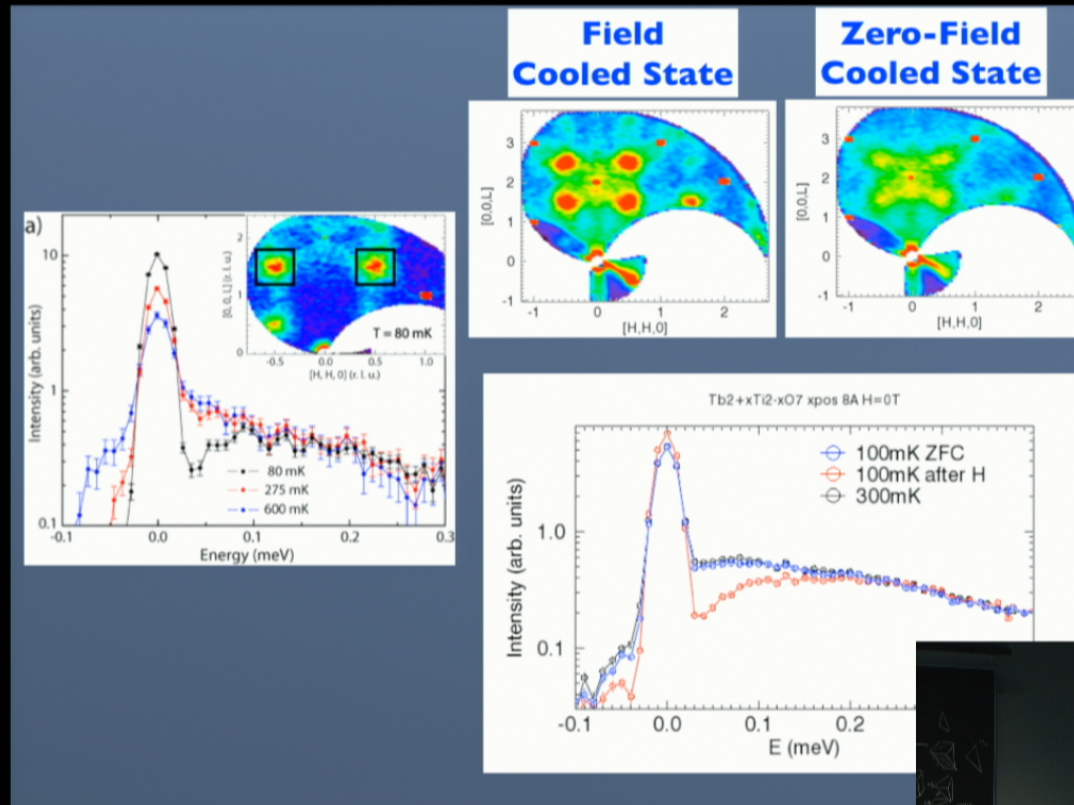


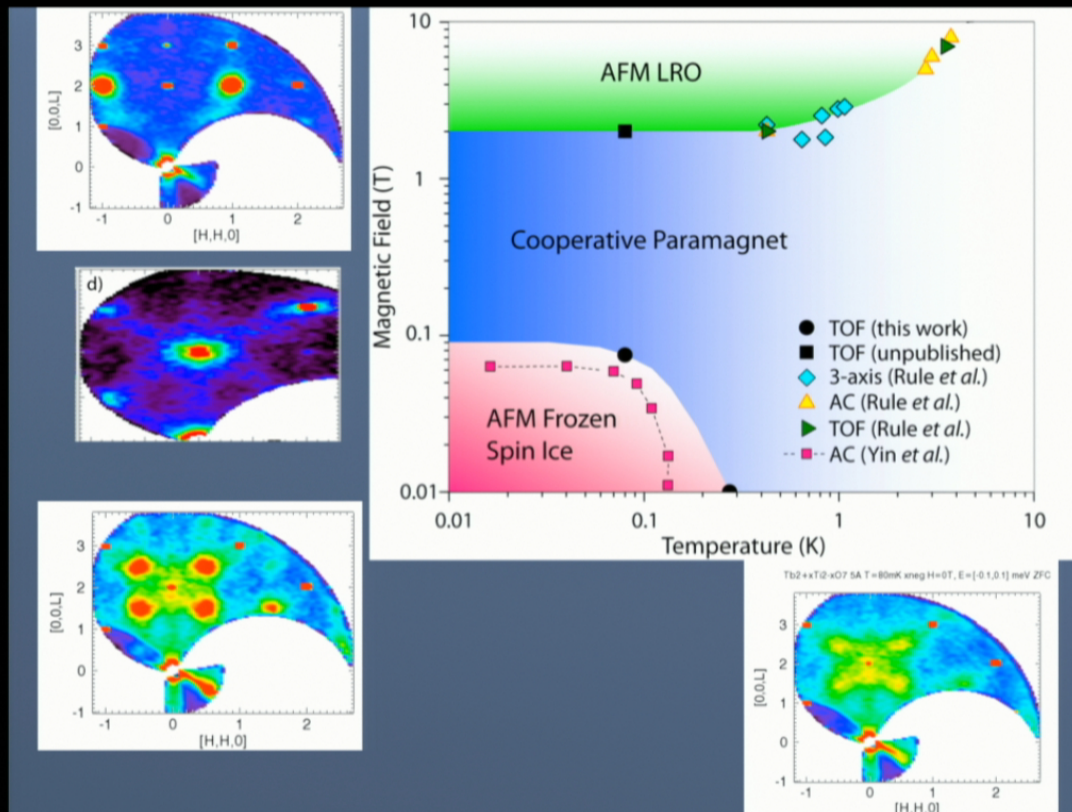
## Glassy-behaviour seen below $\sim 0.25$ K in $\text{Tb}_2\text{Ti}_2\text{O}_7$













### 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 ( $\sim 8$  unit cell) correlations and an  $\sim 0.1$  meV  $\sim 1$  K gap*
- *ZFC state is gapless and shows “checkerboard” correlations*







