Frustrated and Satisfied Ground States in Pyrochlore Magnets

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McMaster University

- **Neutron and X-ray Scattering from Exotic Magnets**
- **Spin liquid and Ordered Ground States in the Pyrochlore Antiferromagnet $Tb_2Ti_2O_7$**
- **Spin Ice Ground State in $Ho_2Ti_2O_7$**
- **Structural Fluctuations within the Spin Liquid State of $Tb_2Ti_2O_7$**

Collaborators

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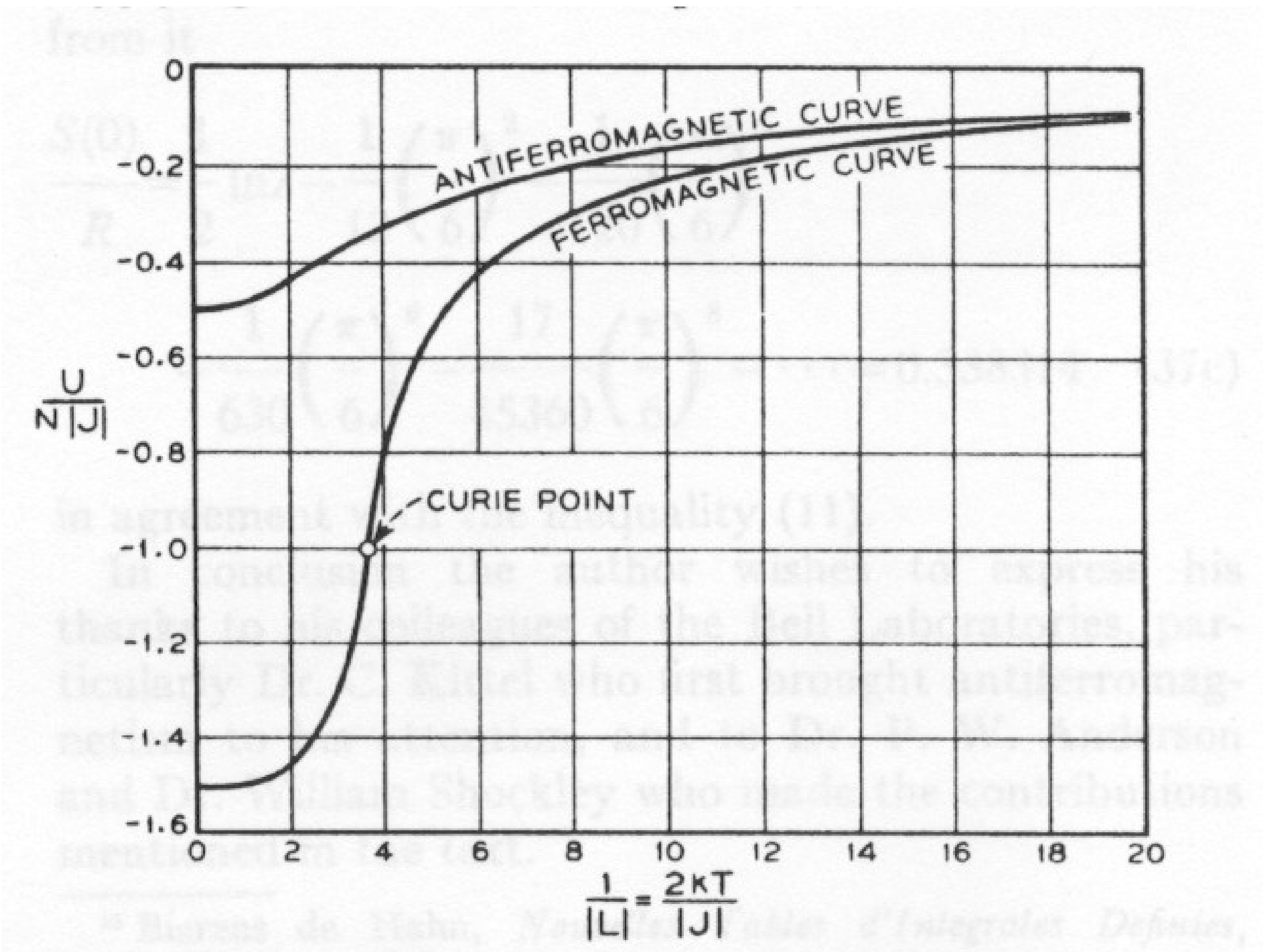
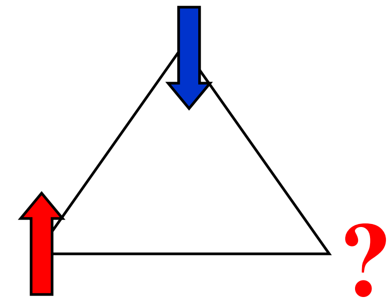
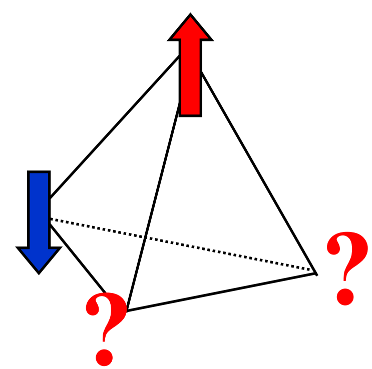
• J.S. Gardner • Y. Qiu • J.R.D. Copley	NIST

Geometrical Frustration:
Antiferromagnetism + Triangles and Tetrahedra

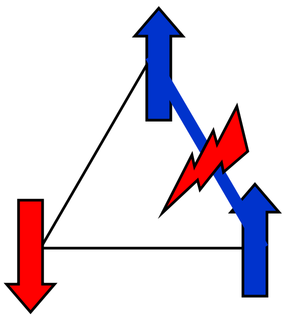


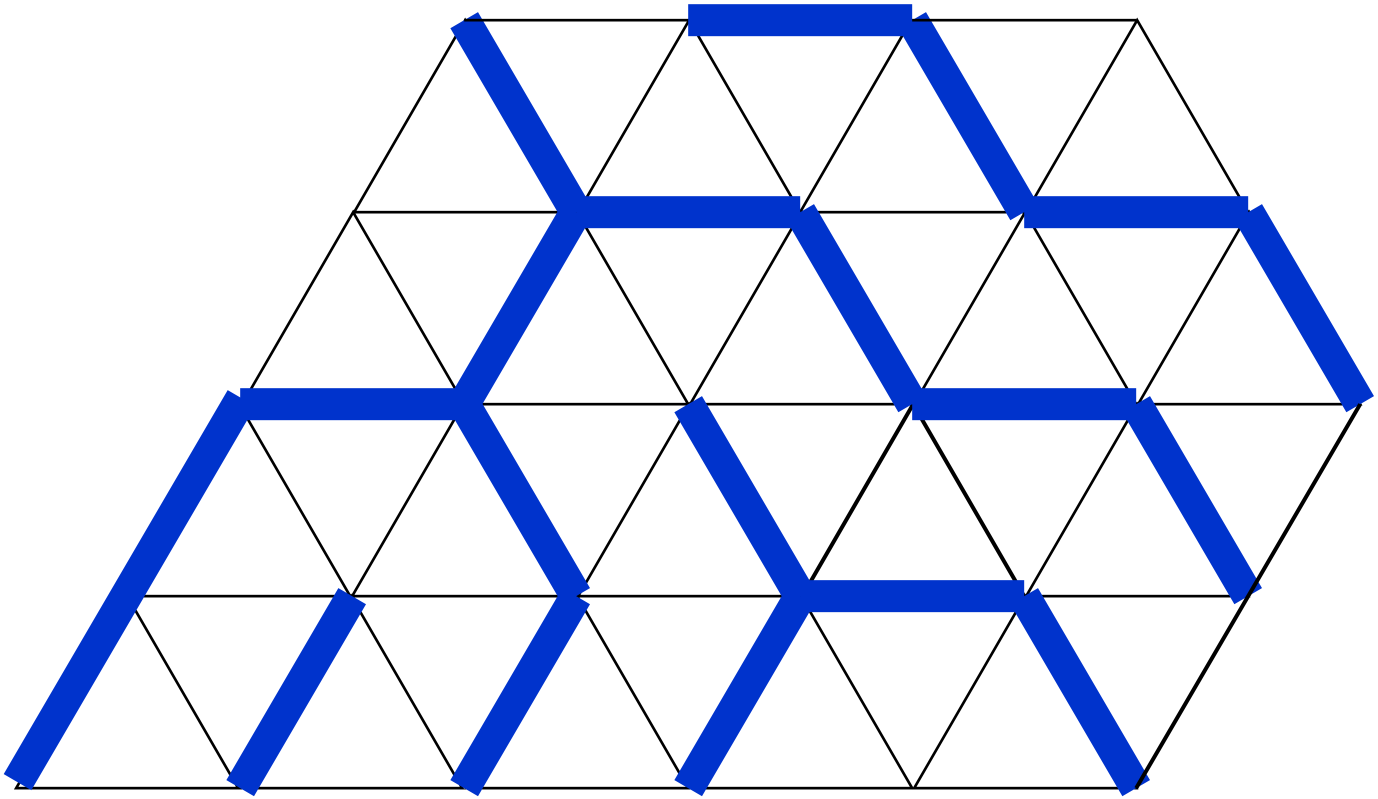




2D Triangular Lattice: G.H. Wannier Phys. Rev. 79, 357, 1950.

Ground State of the Ising AF on a Triangular Lattice

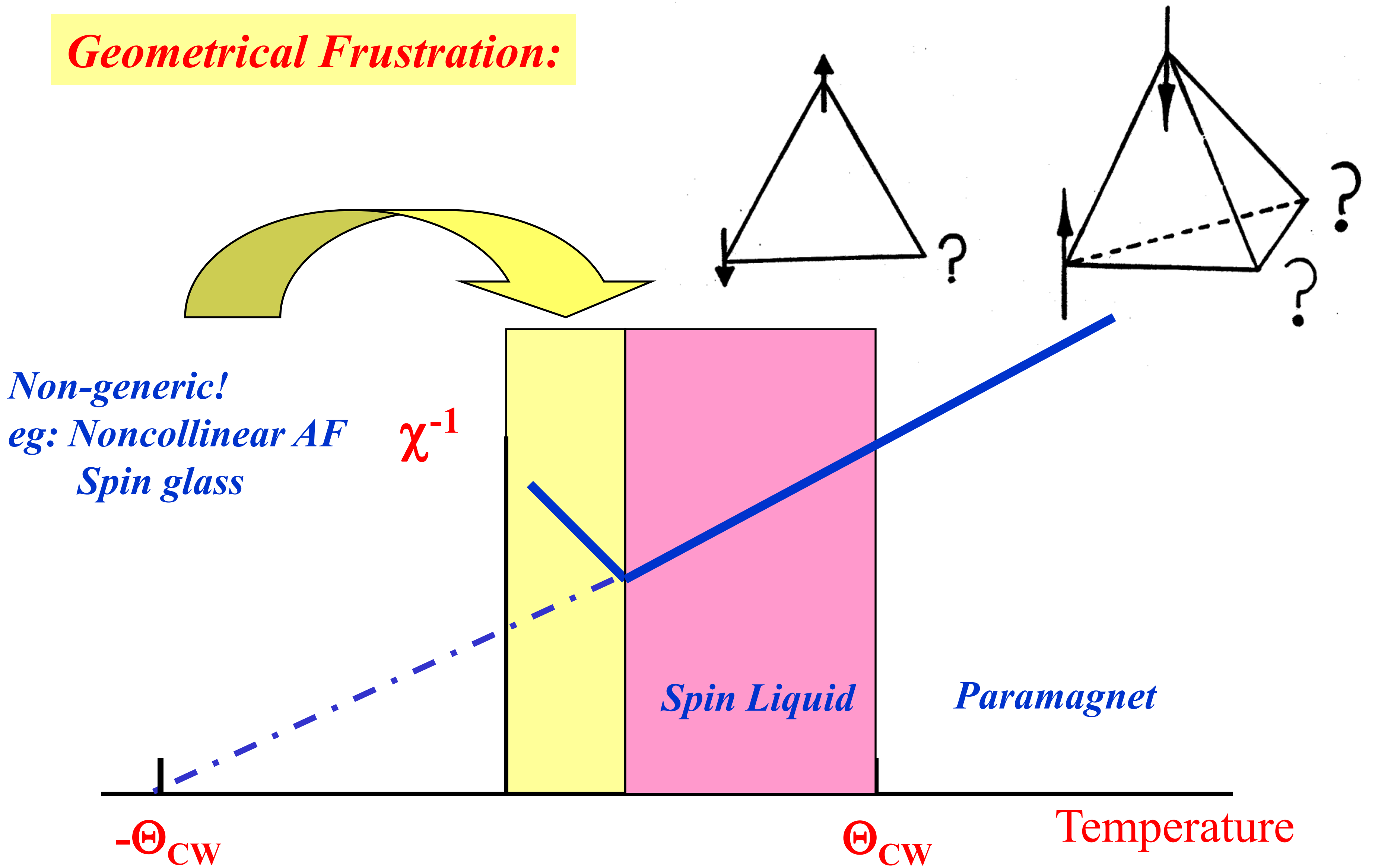




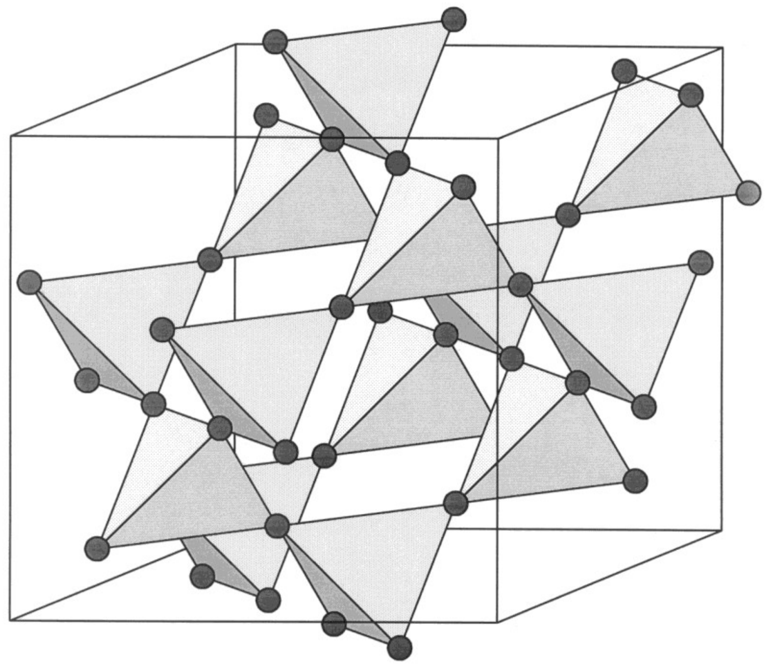
Entropy at T=0 is finite ~ 0.34 R

No LRO at any temperature

Geometrical Frustration:



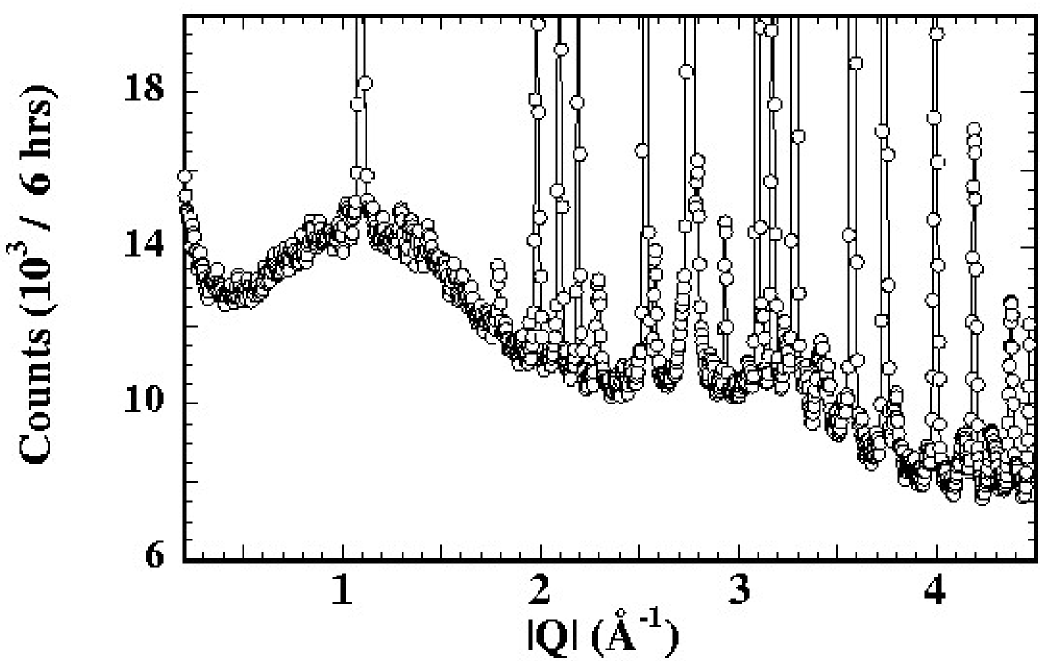
Mean Field Theory predicts a phase transition near T=| Θ_{CW} | ,
but materials remains disordered to much lower temperatures – <u>Spin Liquid</u>



Frustration in three dimensions:

The cubic pyrochlore structure;
A network of corner-sharing tetrahedra

Low temperature powder neutron diffraction from $Tb_2Ti_2O_7$

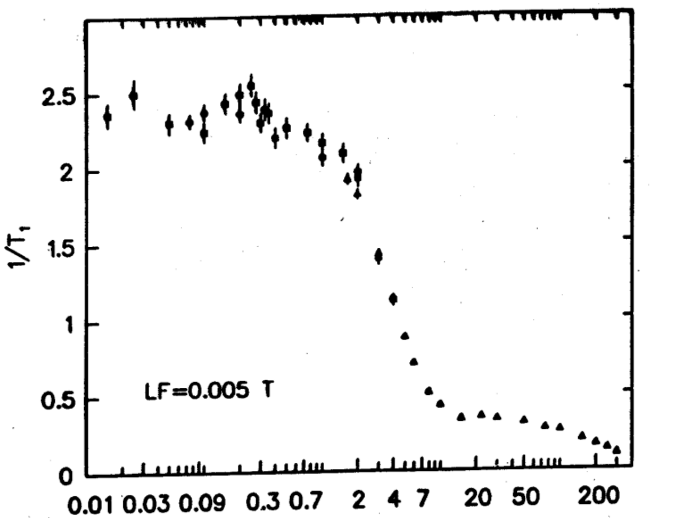


μSR Studies of Magnetic Ground States in:

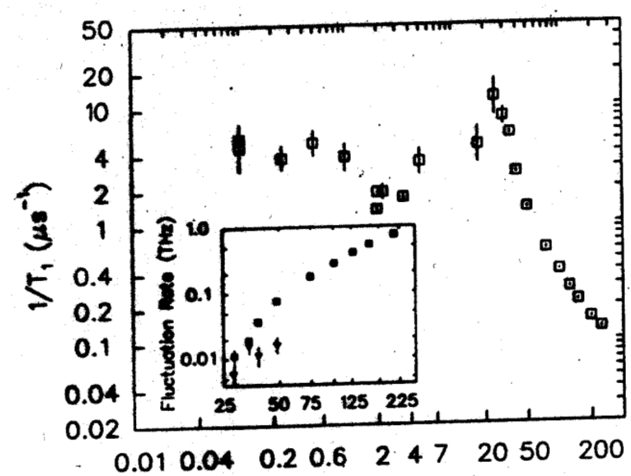
$Tb_2Ti_2O_7$

$Tb_2Mo_2O_7$

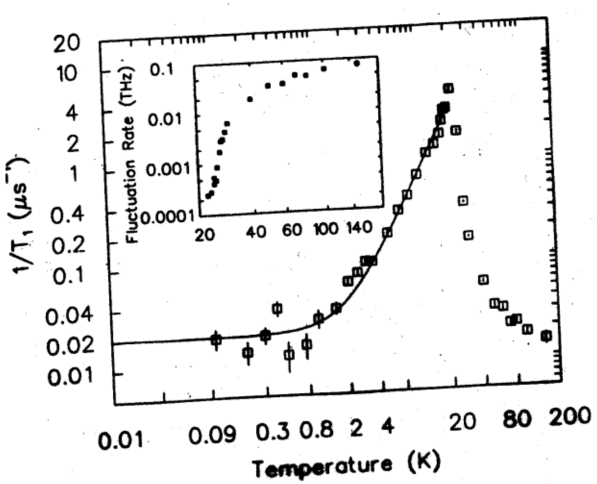
$Y_2Mo_2O_7$



$Tb_2Ti_2O_7$: Spin Liquid

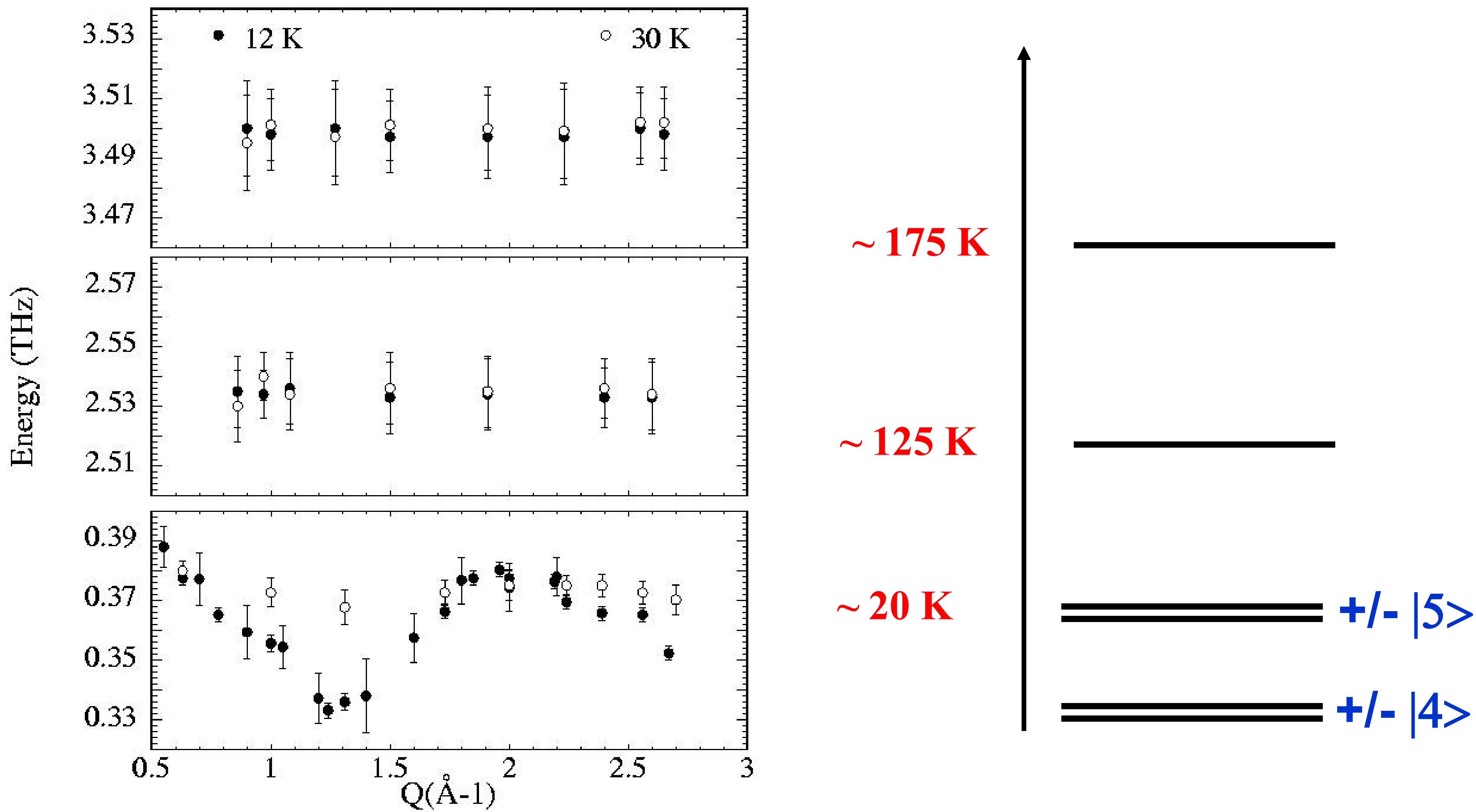


$Tb_2Mo_2O_7$: Spin Liquid and Spin Glass

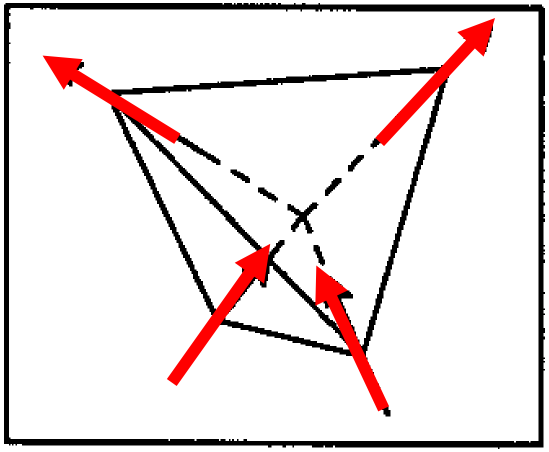


$Y_2Mo_2O_7$: Spin Glass

Inelastic neutron scattering on polycrystalline $Tb_2Ti_2O_7$

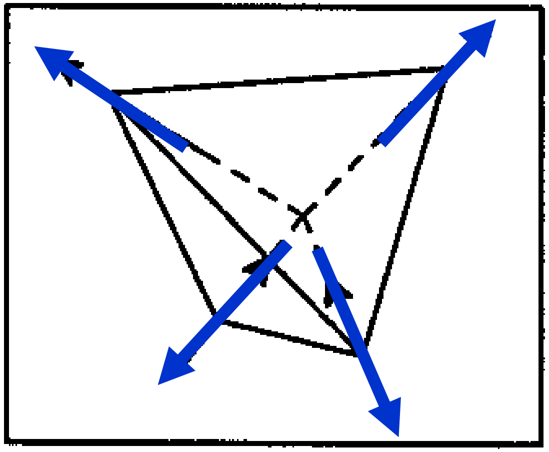


(Δ : $Ho_2Ti_2O_7$ ~ 240 K ; $Dy_2Ti_2O_7$ ~ 380 K)



Rare Earth moments:

Strong [111] anisotropy



Ferromagnetic exchange:

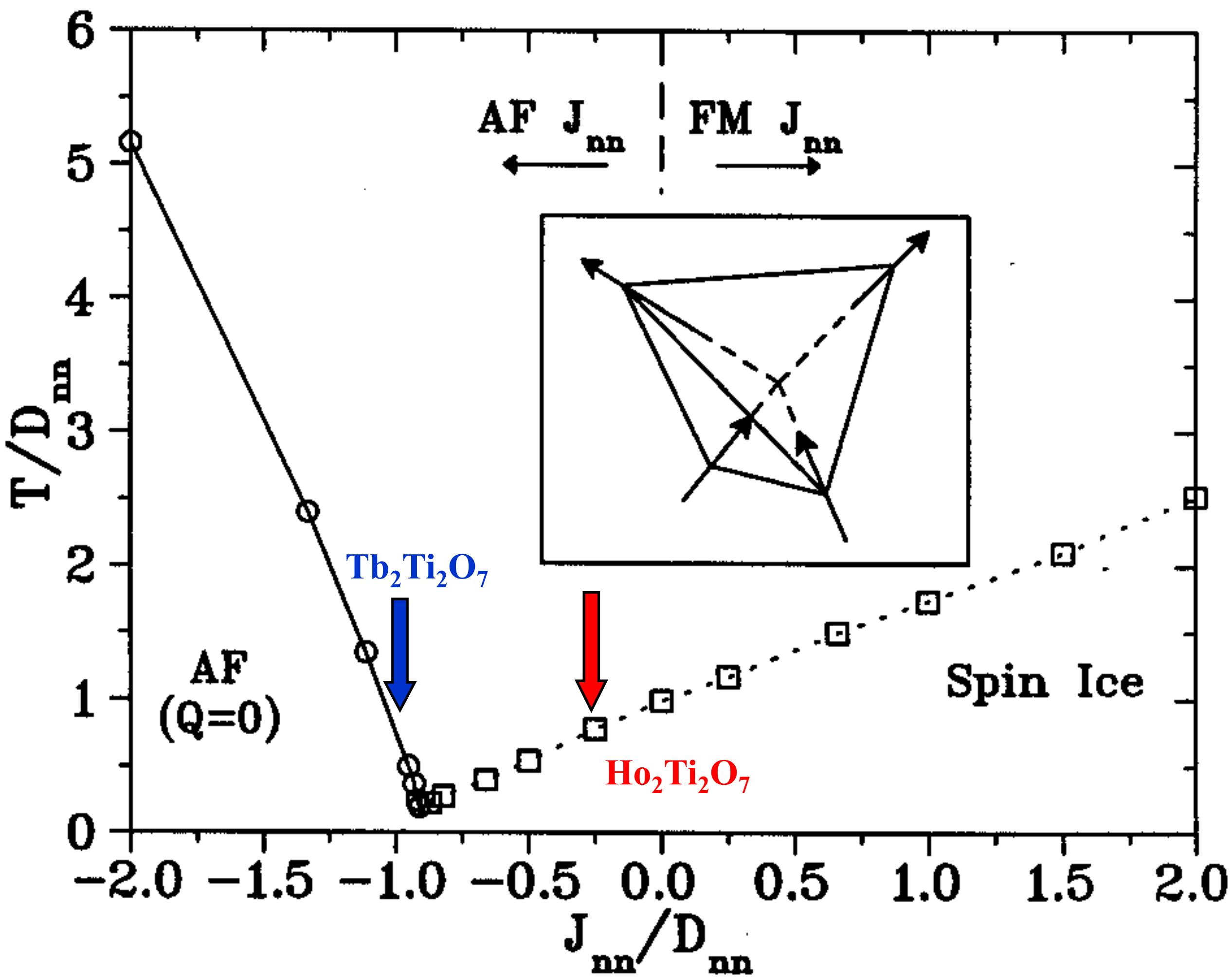
"Spin Ice" : 2 in 2 out

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

Antiferromagnetic exchange:

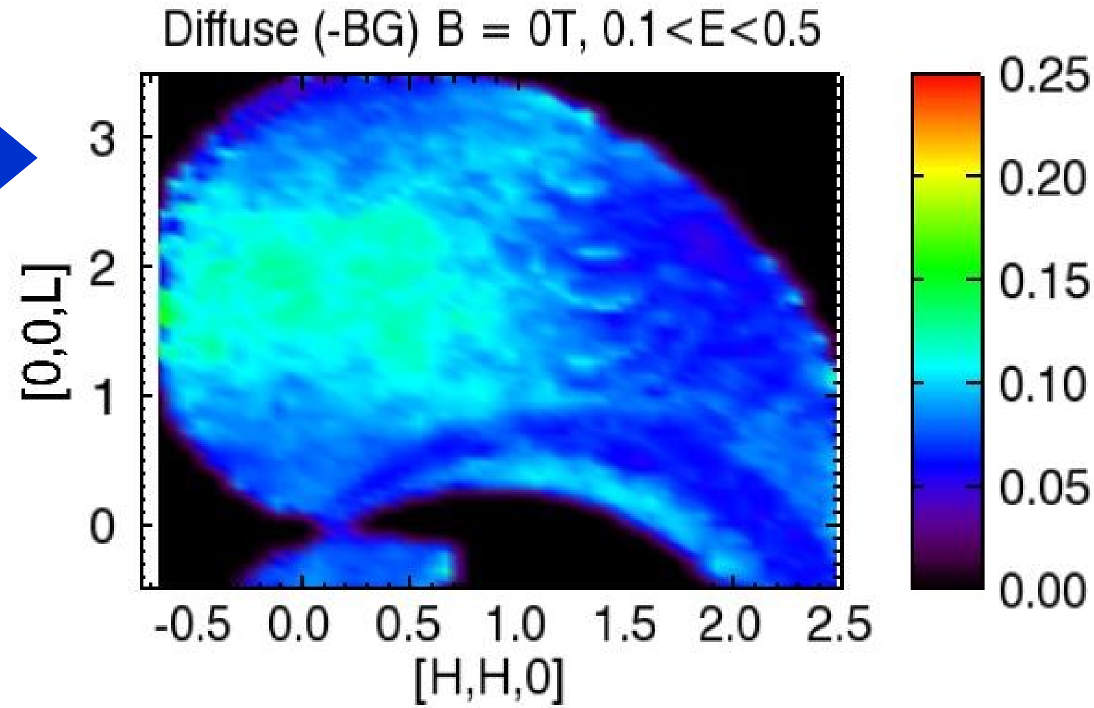
All in - All out

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

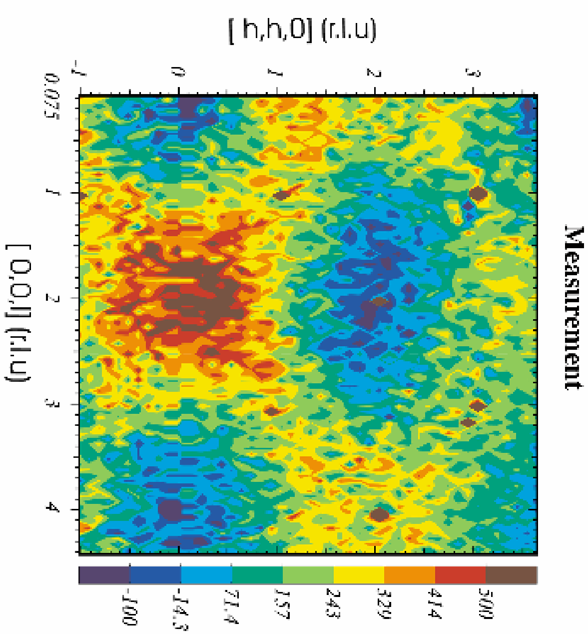


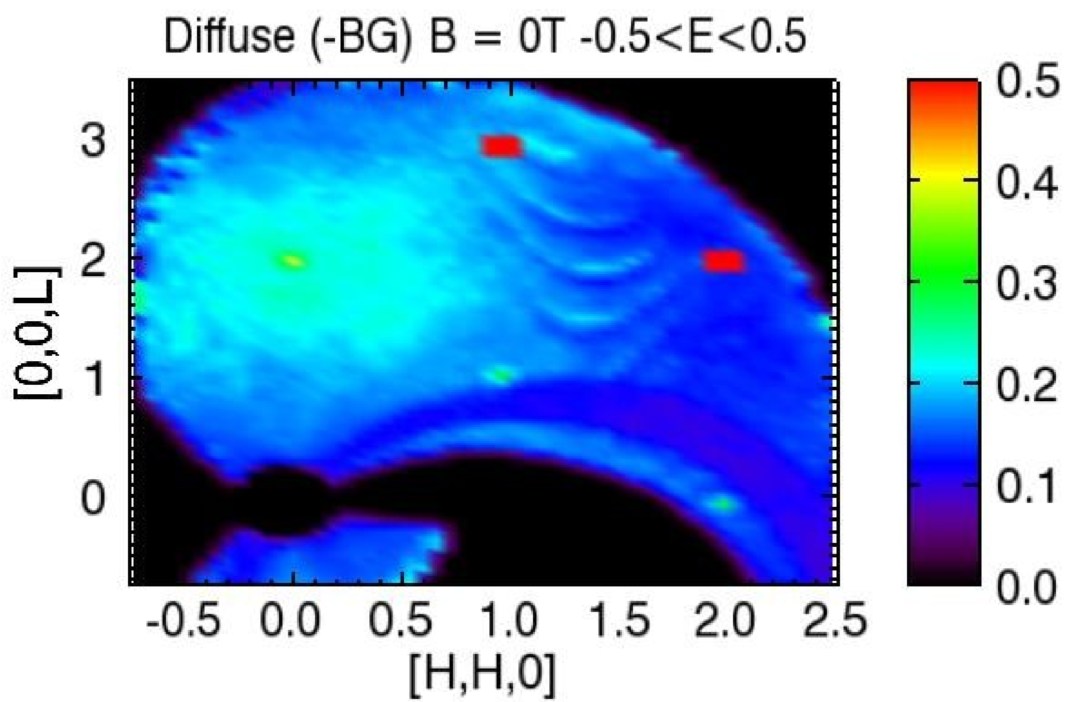
B.C. den Hertog and M.J.P. Gingras, PRL, 84, 3430, 2000.

TOF scattering
T=0.4 K



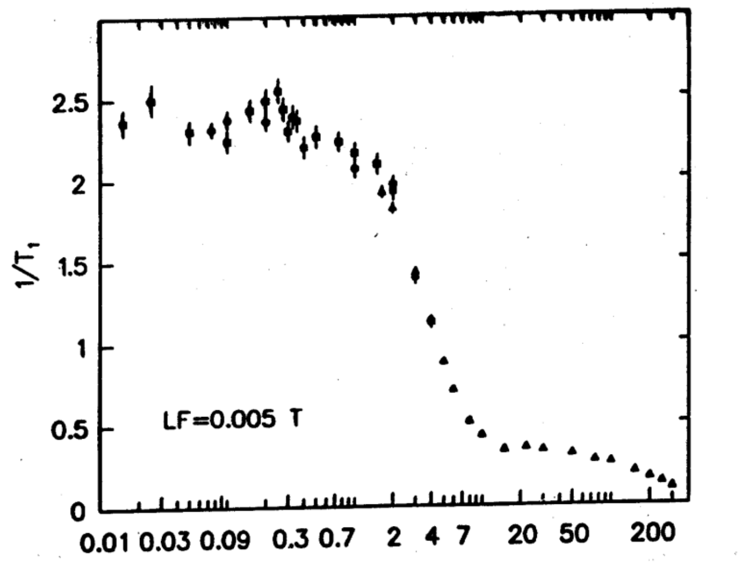
2-Axis Diffraction
T=9 K

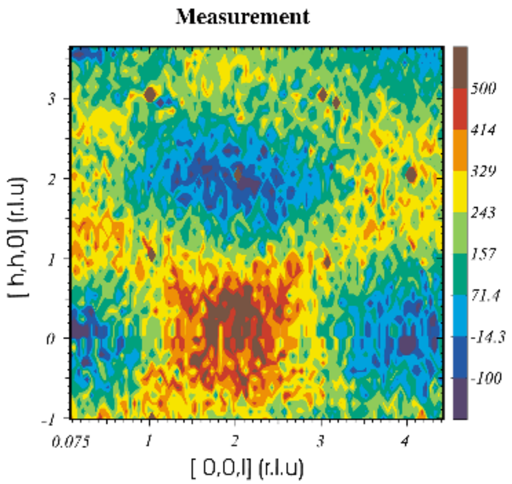




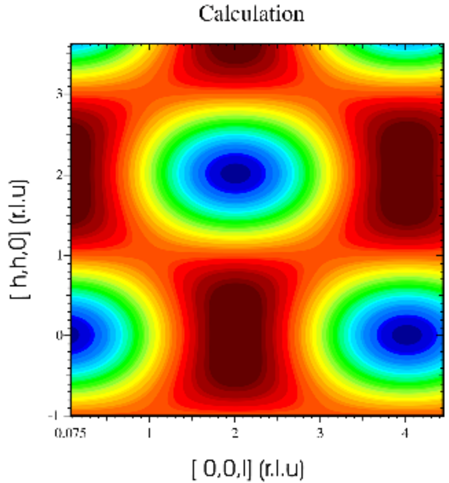
Outstanding Questions:

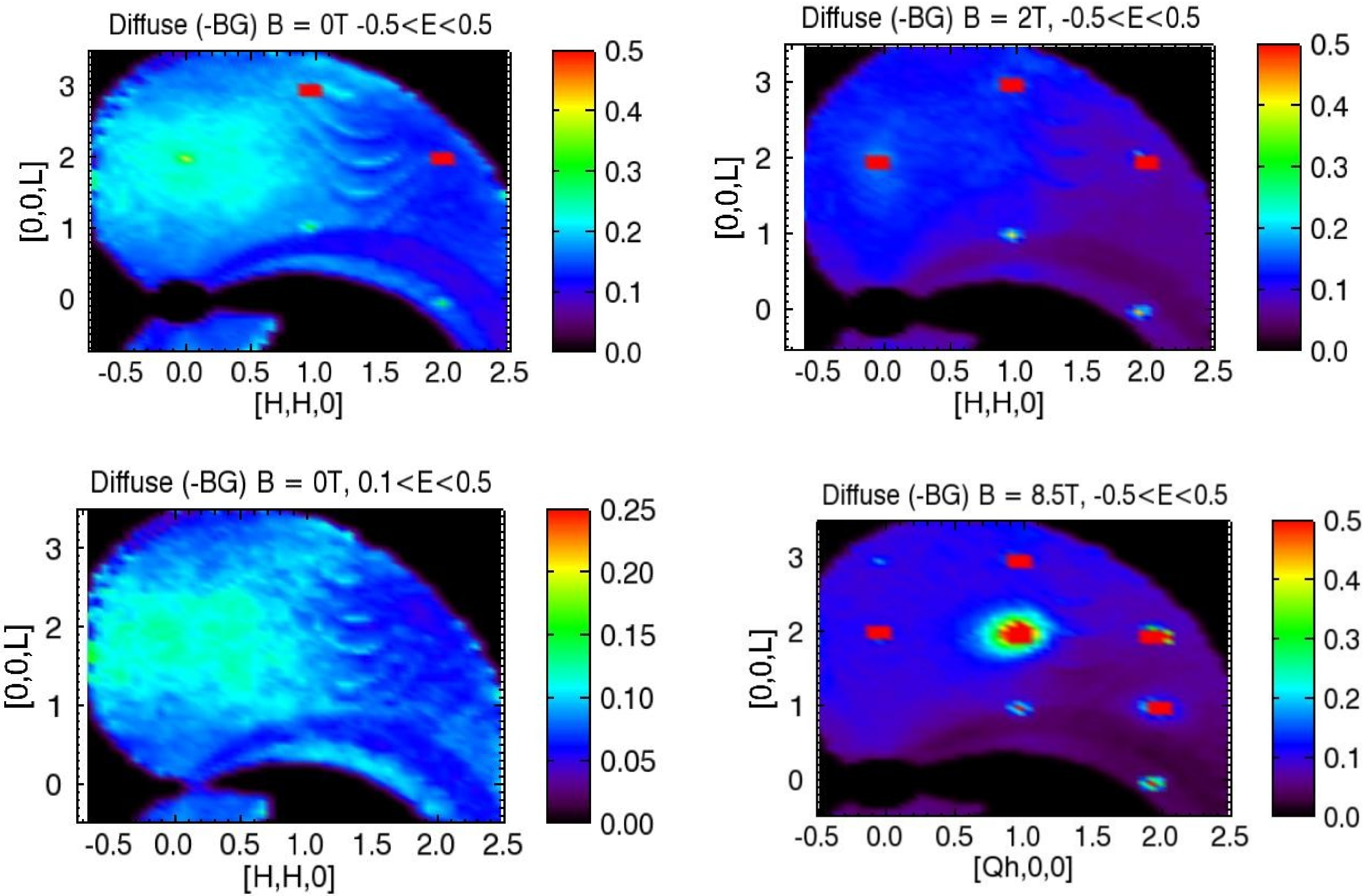
- **Why is $Tb_2Ti_2O_7$ disordered as low as 0.02 K (in H=0, P=0)?**



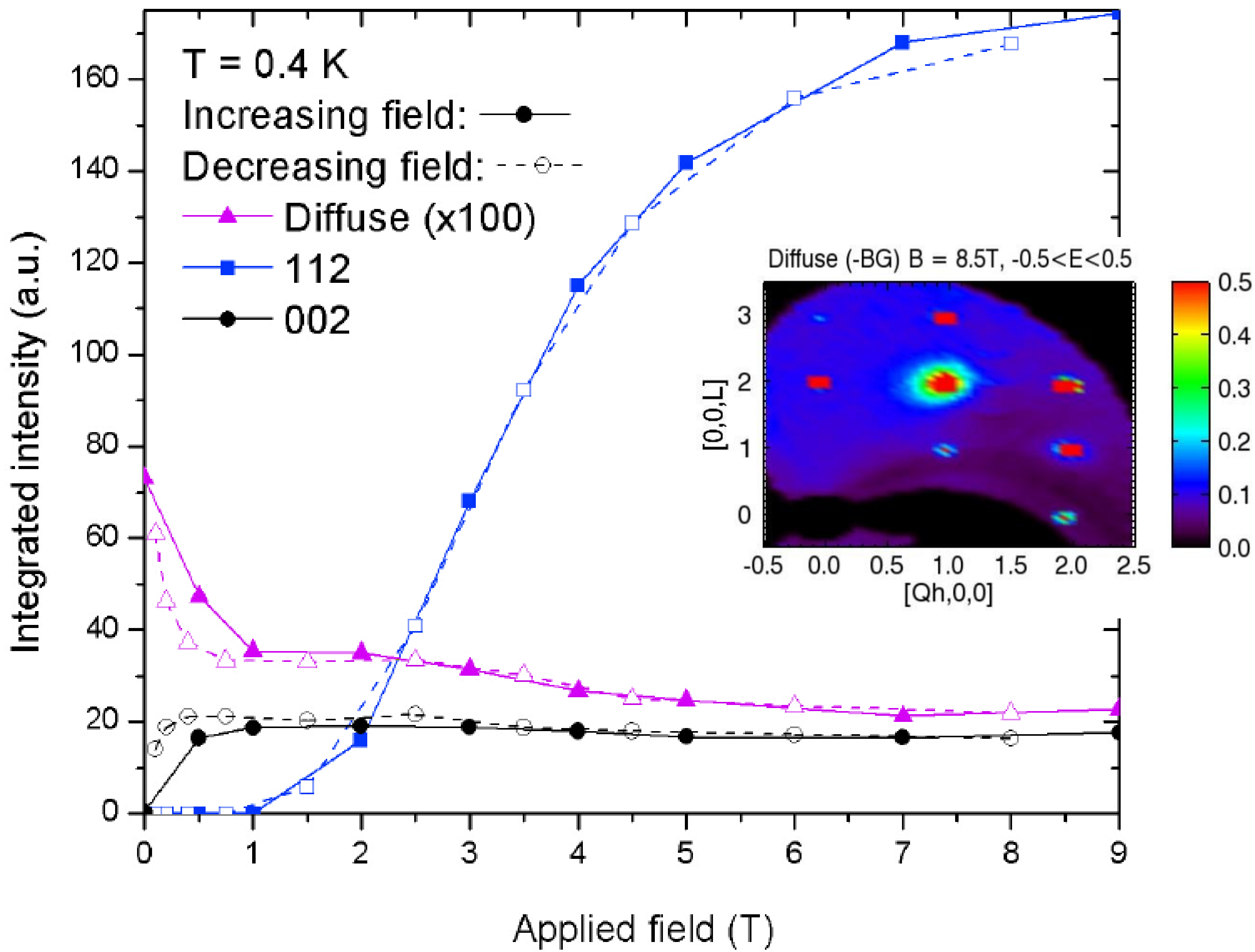


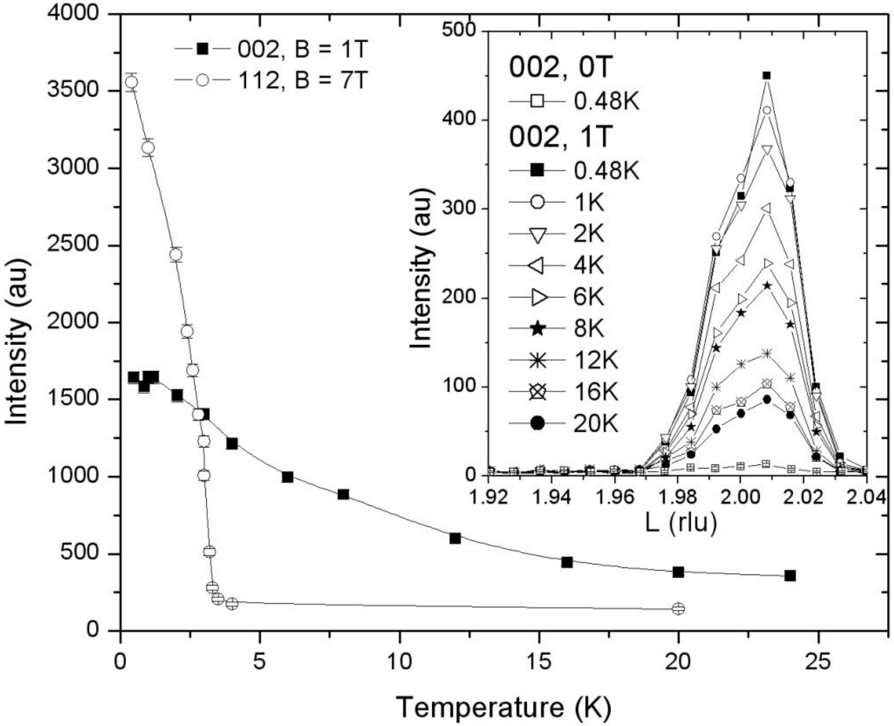
- **S(Q) at low T is (perhaps naively) incompatable with Ising (111) anisotropy. What is going on?**

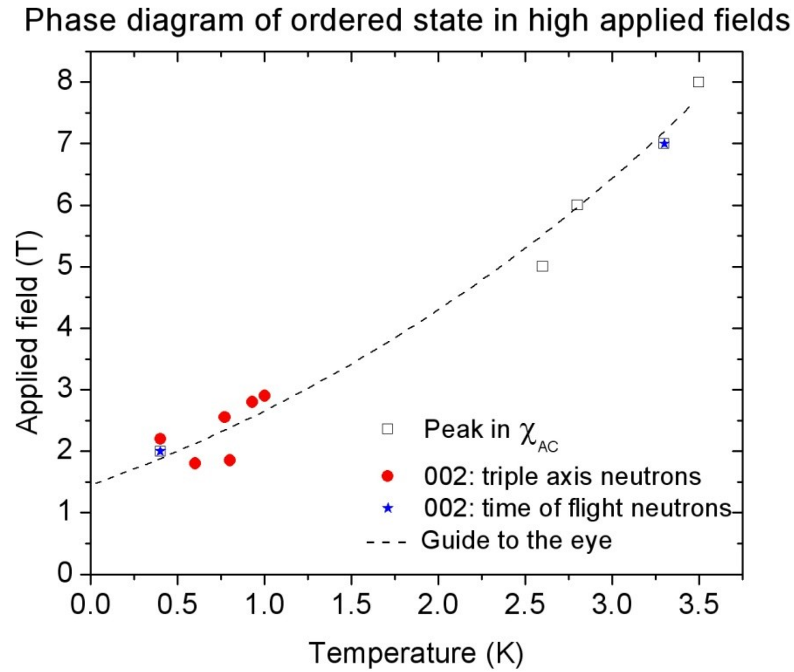


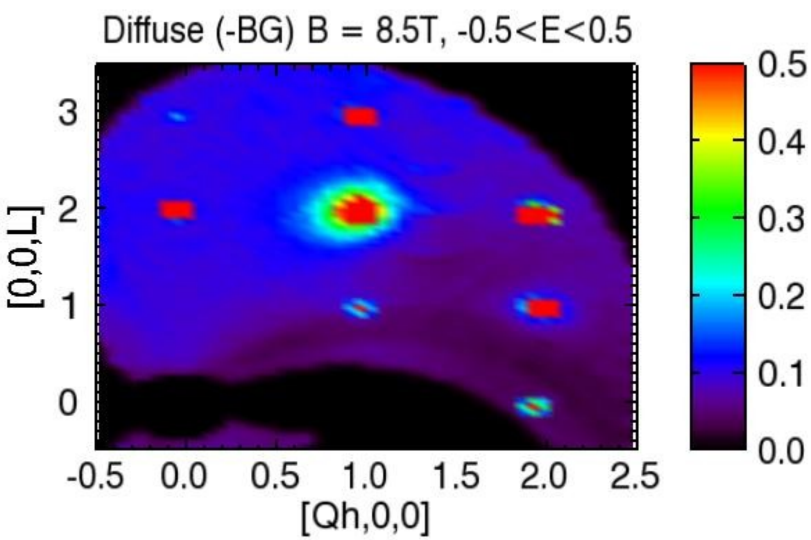


Ordered phases appear on application of H // 110


T = 0.4 K
Increasing field:
Decreasing field:
Diffuse (x100)
112
002
Integrated intensity (a.u.)
Applied field (T)
0
20
40
60
80
100
120
140
160
0
1
2
3
4
5
6
7
8
9
Diffuse (-BG) B = 8.5T, -0.5<E<0.5
[0,0,L]
[Qh,0,0]
-0.5
0.0
0.5
1.0
1.5
2.0
2.5
0.1
0.2
0.3
0.4
0.5

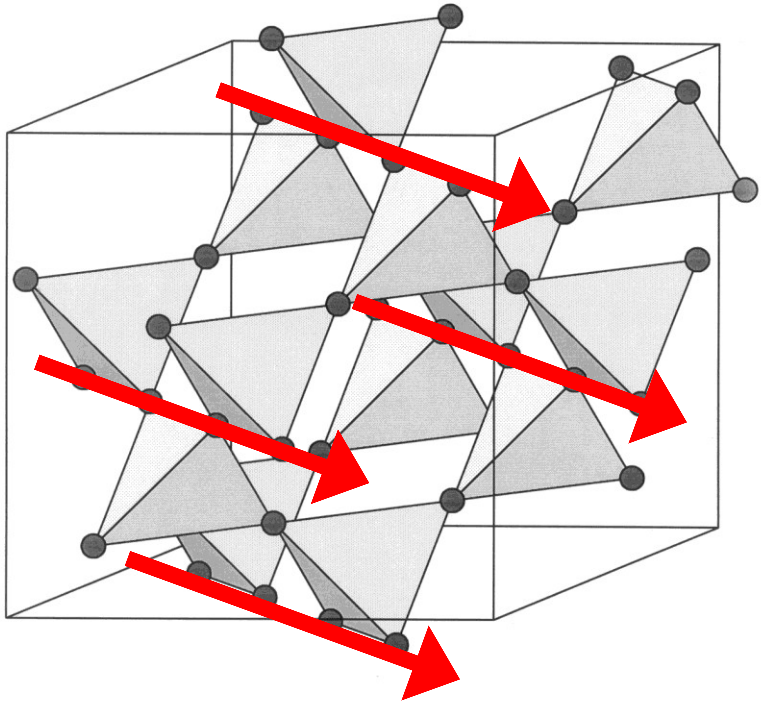






Low field (002) phase persists to very high T > 25 K

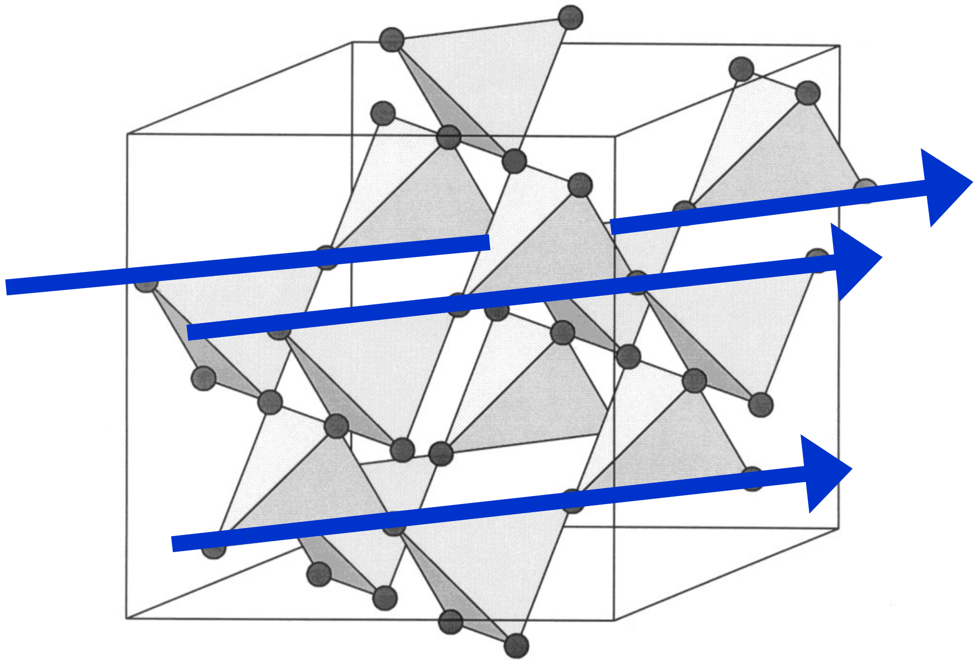
High field (112) phase exists on expected T_N ~ 2 K



Application of weak [1-10] magnetic field breaks system up into α and β chains.

Polarizable α-[1-10] chains (parallel to field)

Perpendicular β-[110] chains

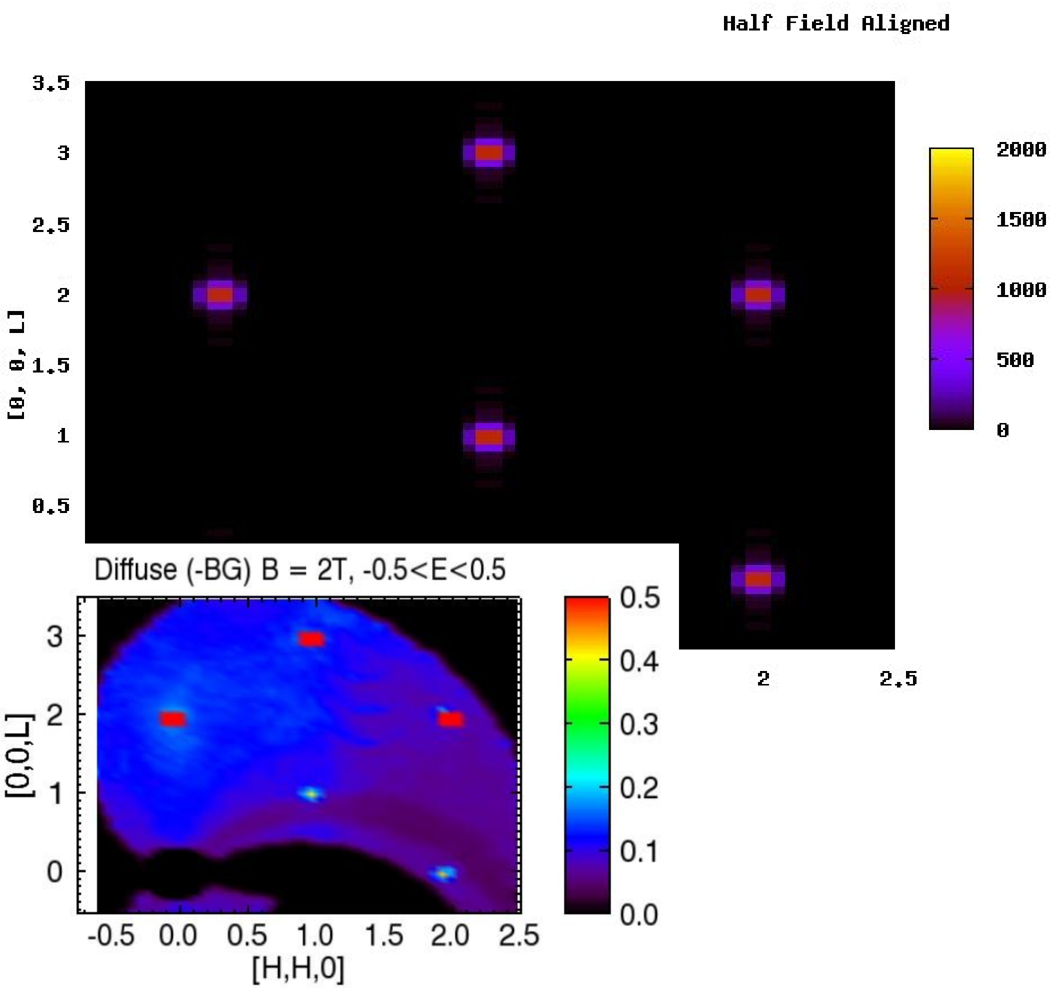


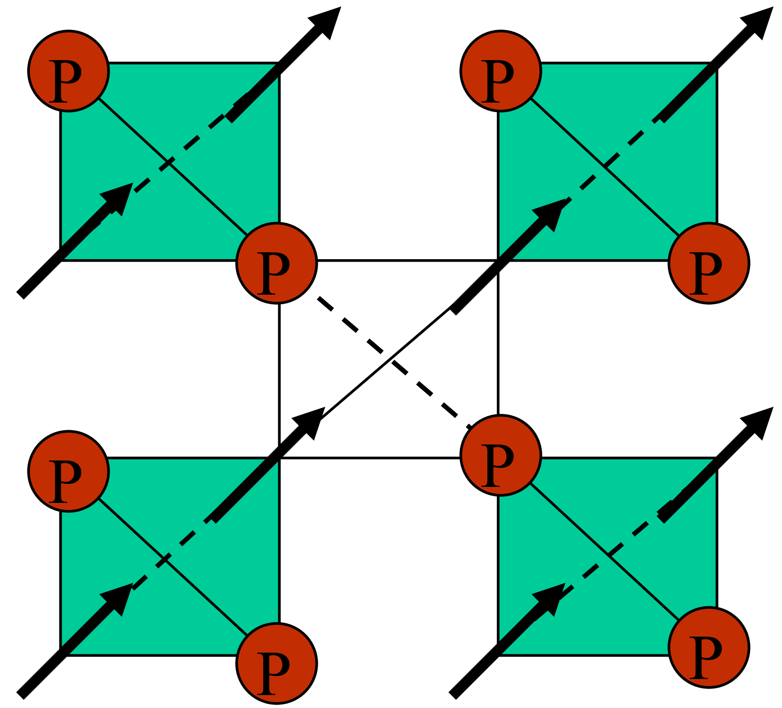
Application of weak [1-10] magnetic field breaks system up into α and β chains.

Polarizable α-[1-10] chains (parallel to field)

Perpendicular β-[110] chains

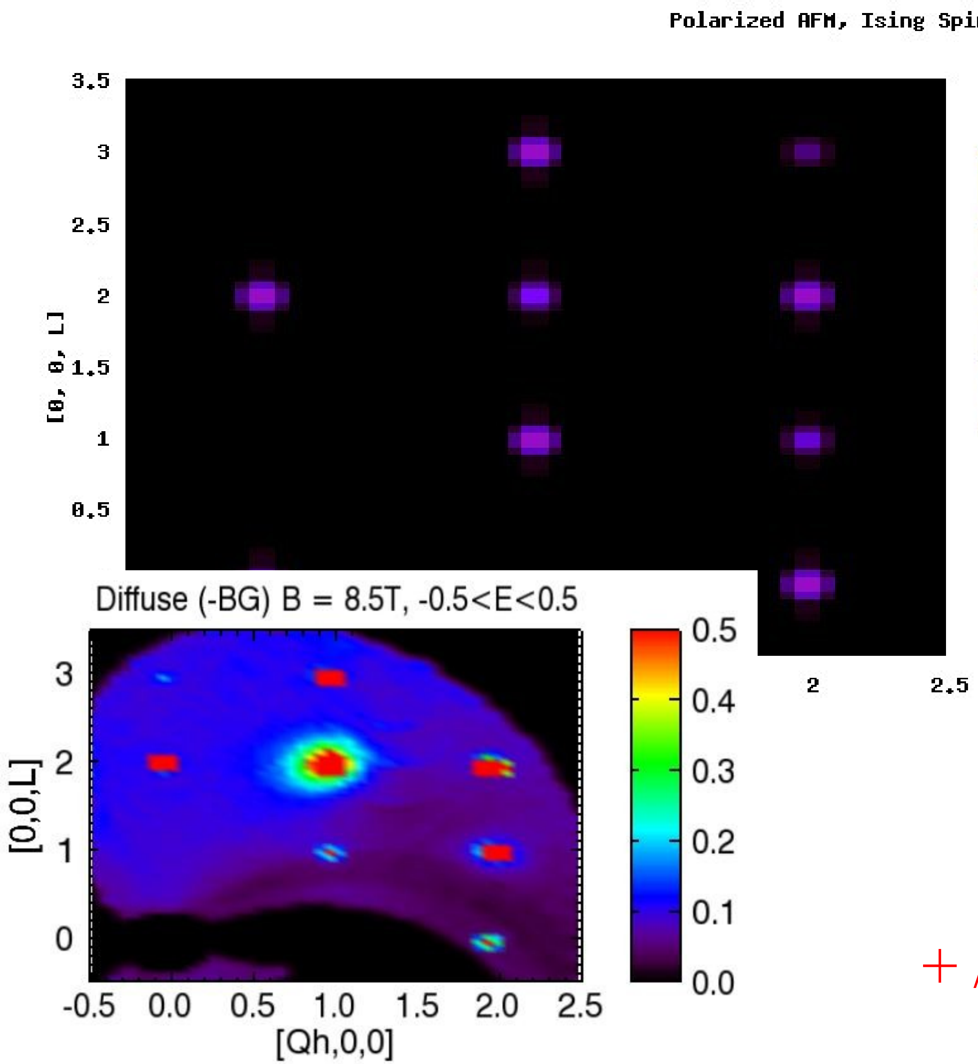
α-chains polarized along the [1,1,0] Direction

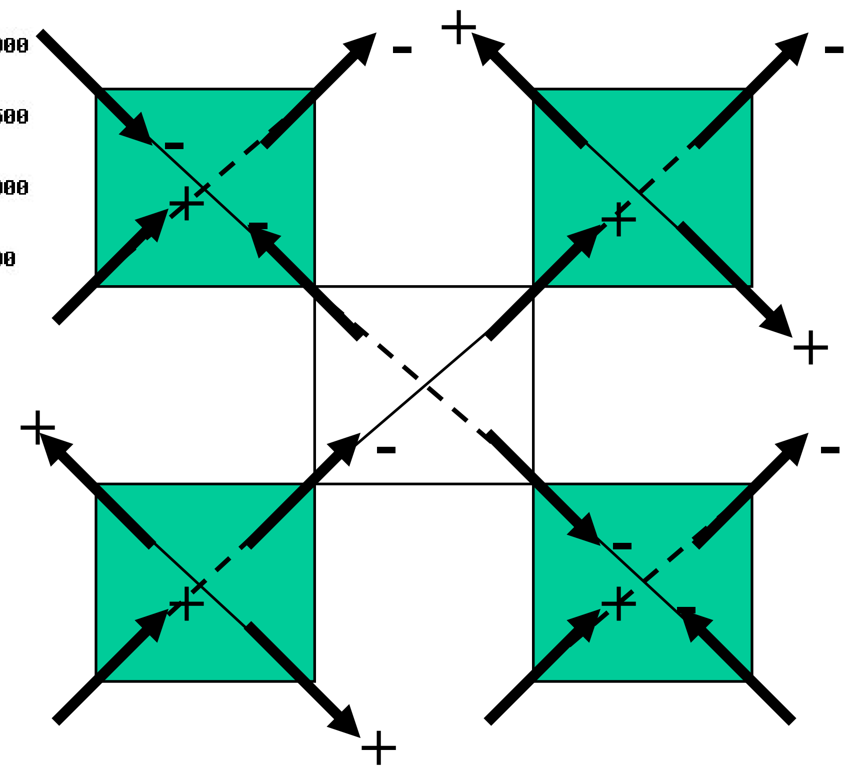




P = Paramagnetic Site

Field Aligned, Local <111> AFM

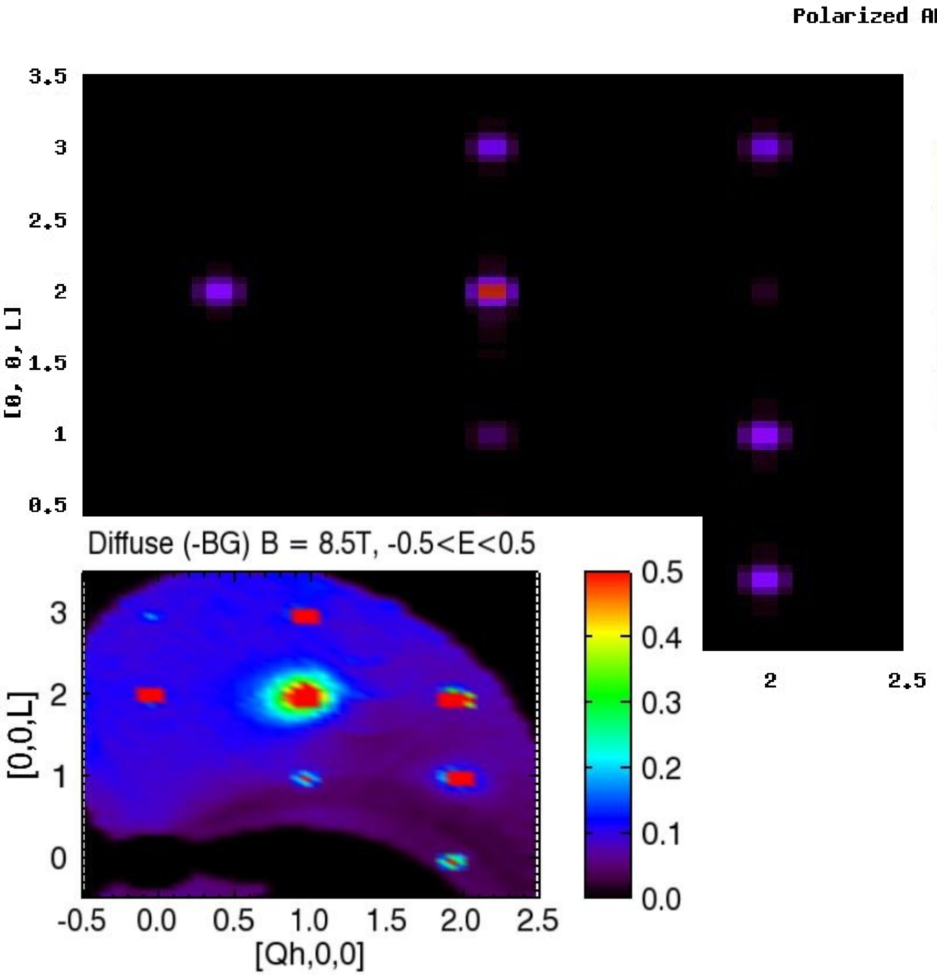


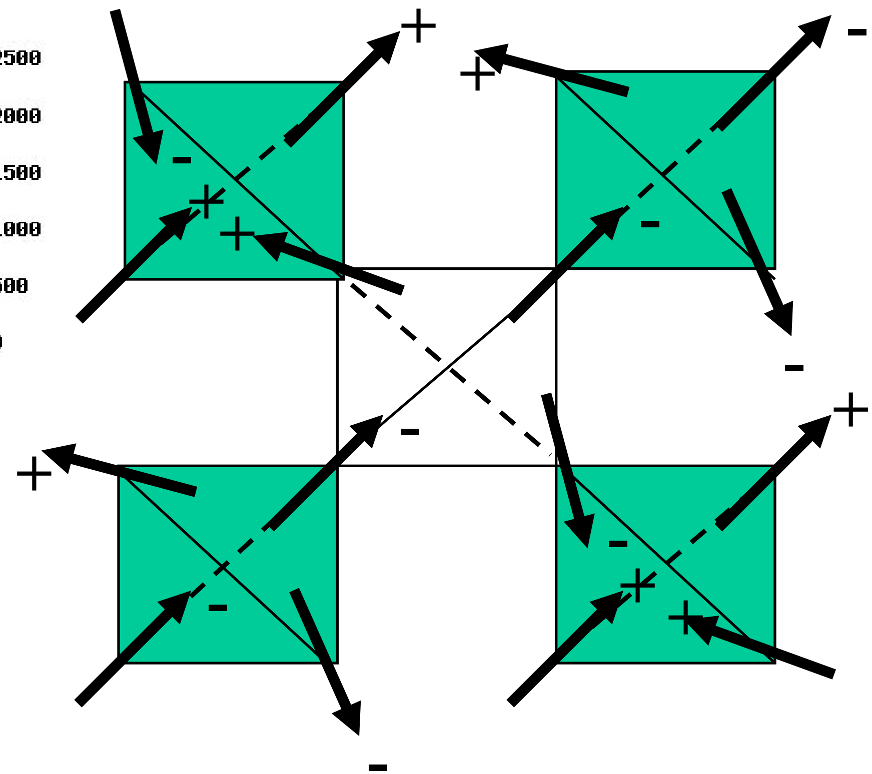


+ / - Denotes Z-Component

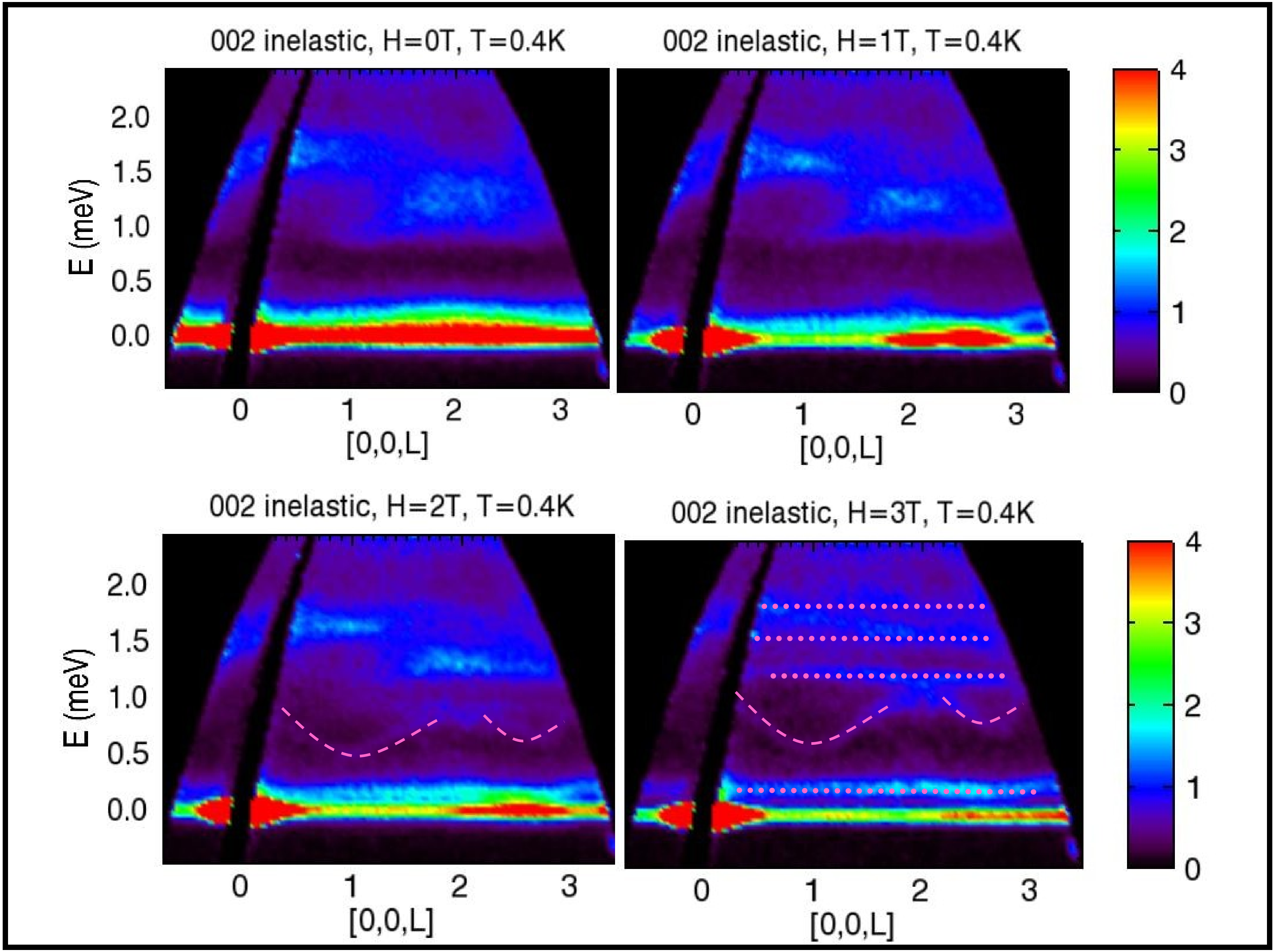
“3 in – 1 out” local structure

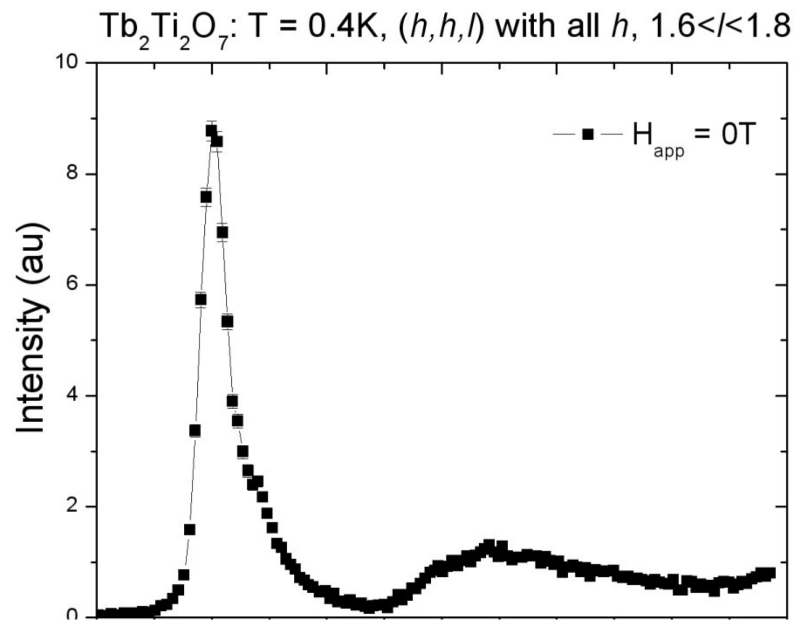
Canted AFM, Moments not respecting Ising 111 axes.
(Magnetization is ~11% of saturation along [110])



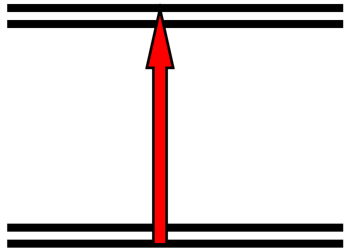


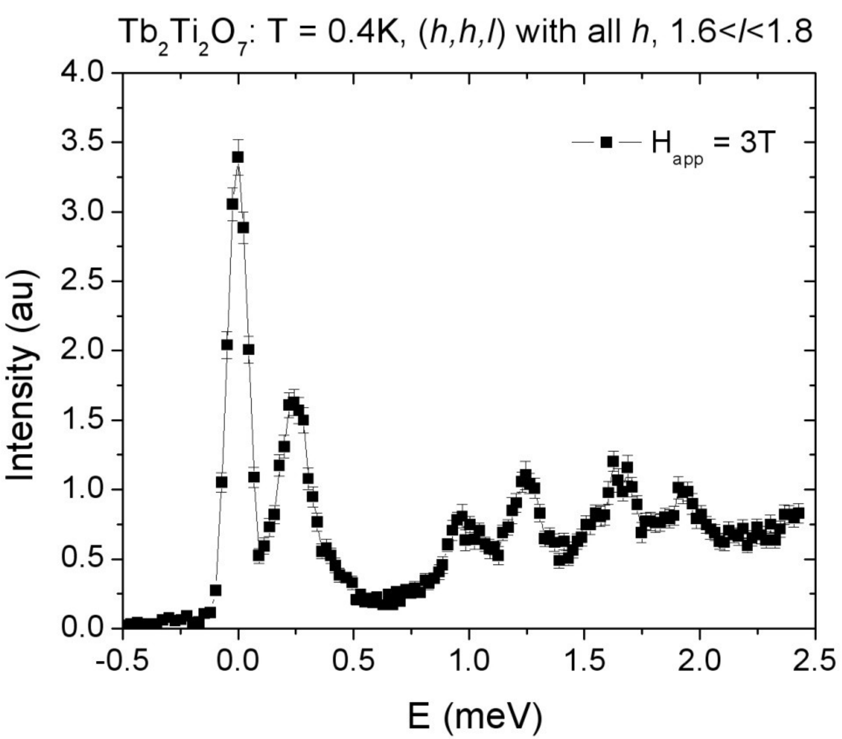
+ / - Denotes Z-Component


002 inelastic, H=0T, T=0.4K
002 inelastic, H=1T, T=0.4K
002 inelastic, H=2T, T=0.4K
002 inelastic, H=3T, T=0.4K
E (meV)
[0,0,L]
2.0
1.5
1.0
0.5
0.0
0
1
2
3
4

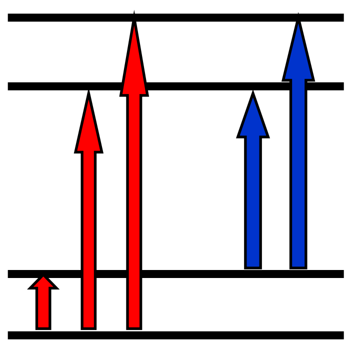


One Transition in Zero Field

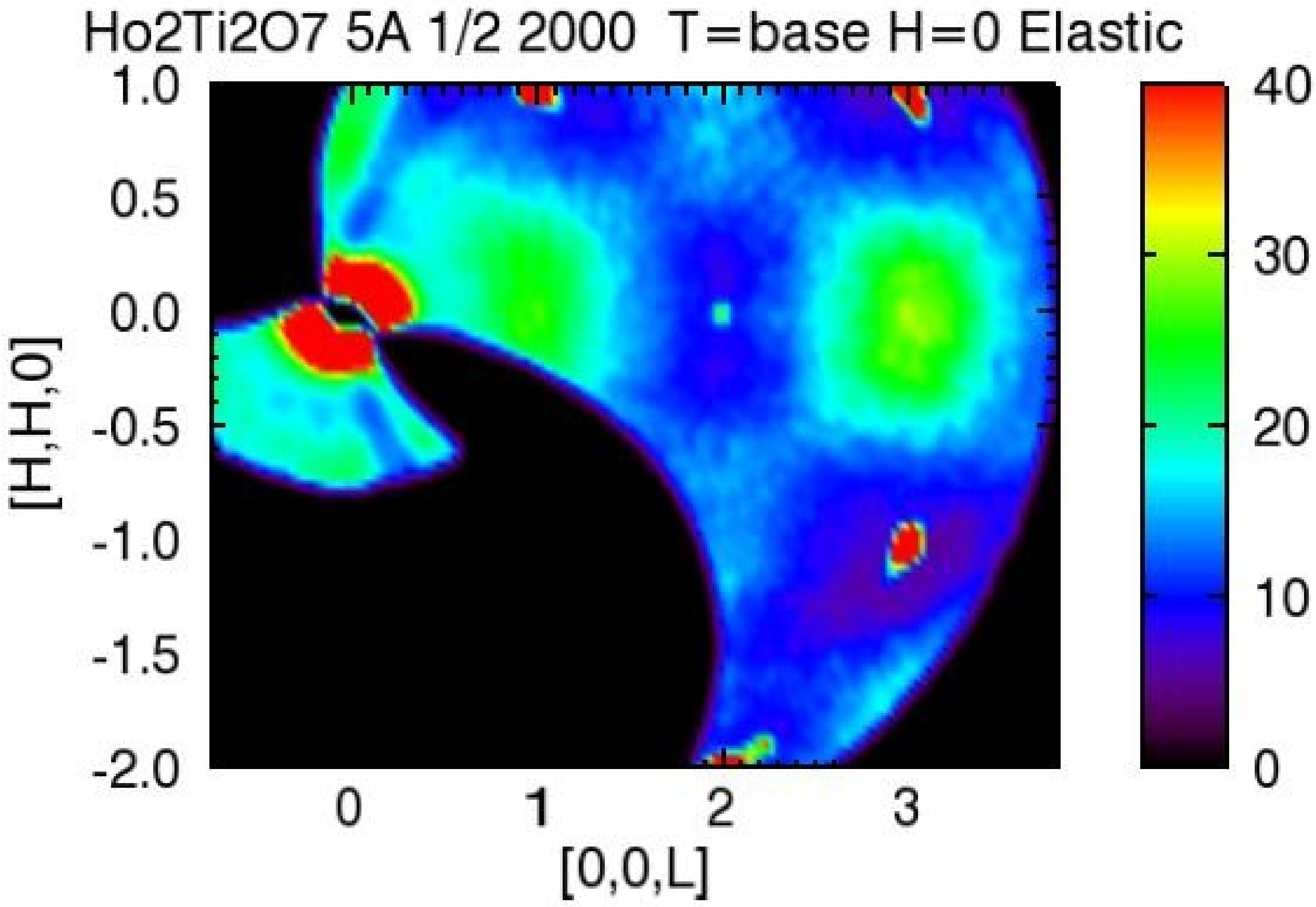




Five Transitions in Non-Zero Field

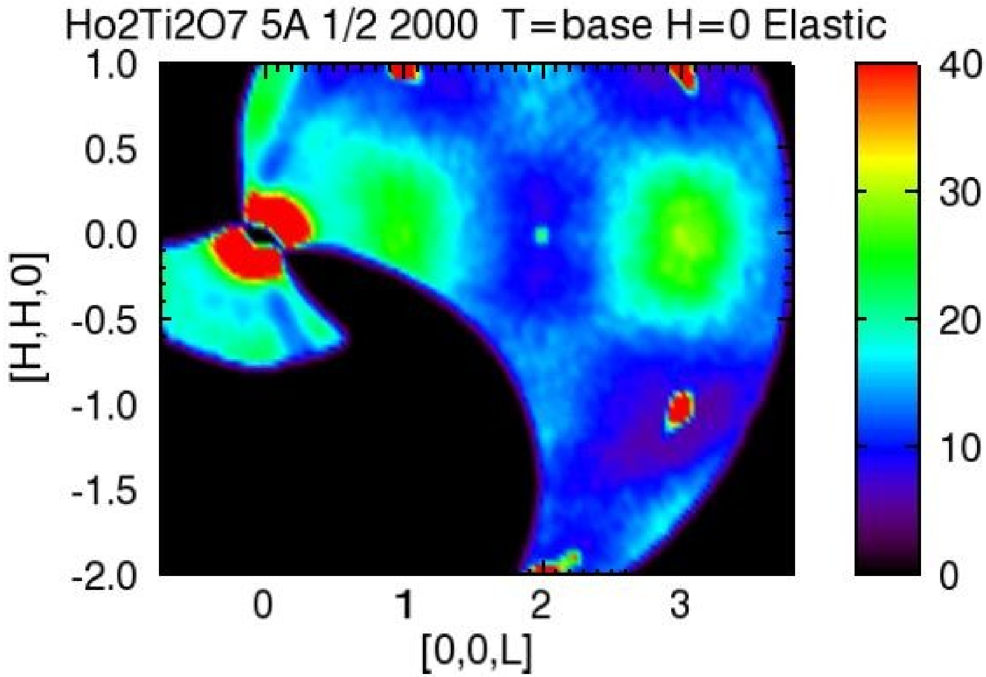


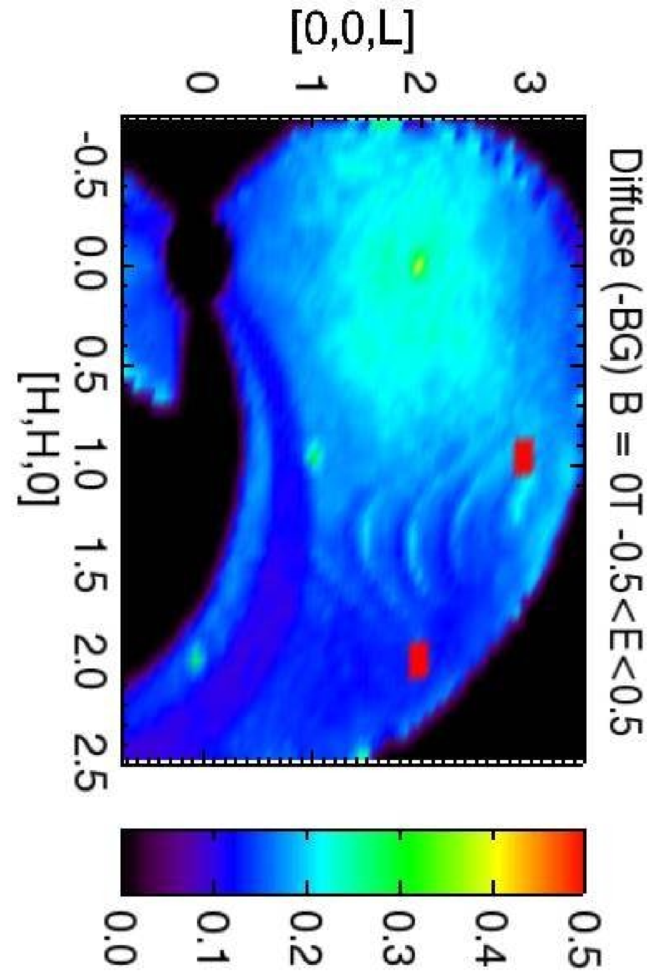
Spin Ice Ground State in $Ho_2Ti_2O_7$



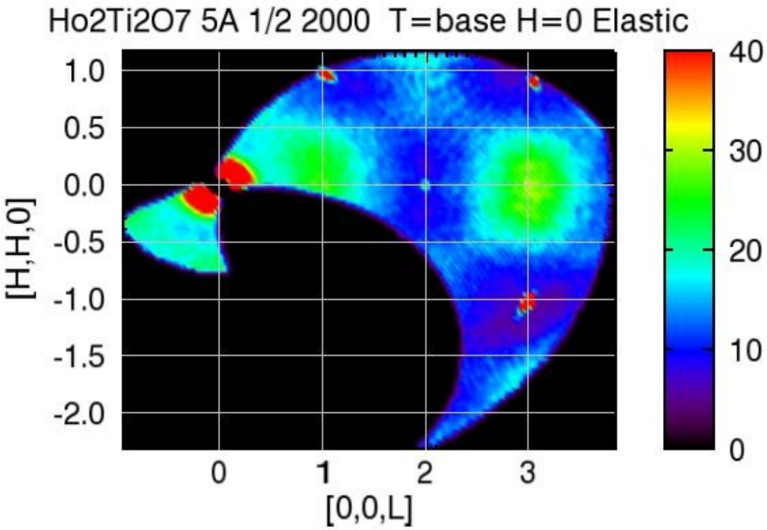
$Ho_2Ti_2O_7$ vs $Tb_2Ti_2O_7$

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

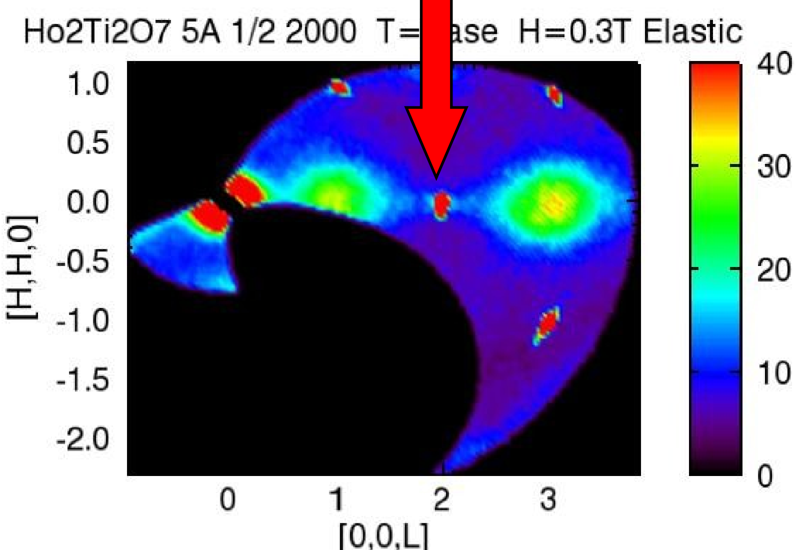


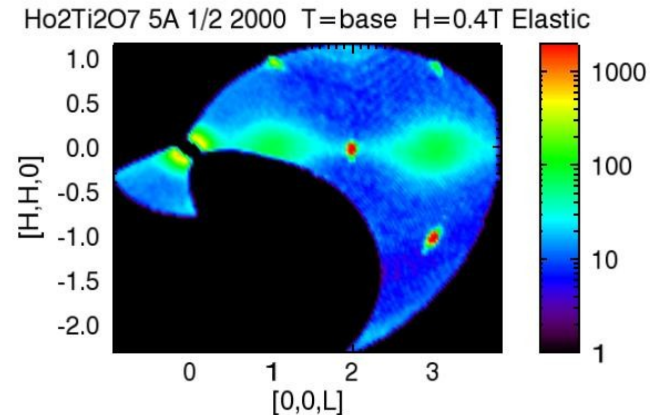


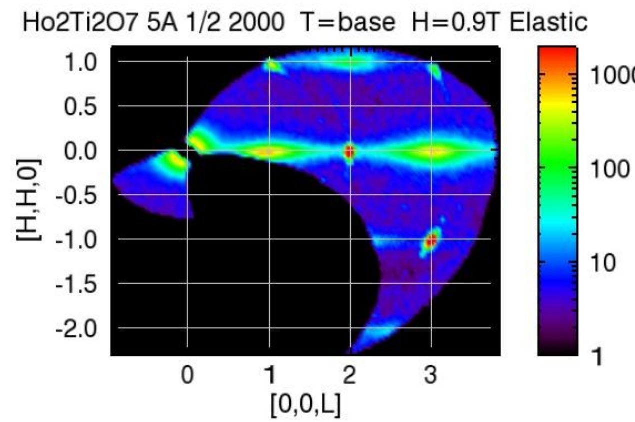
Magnetic Structure Factors appear complementary



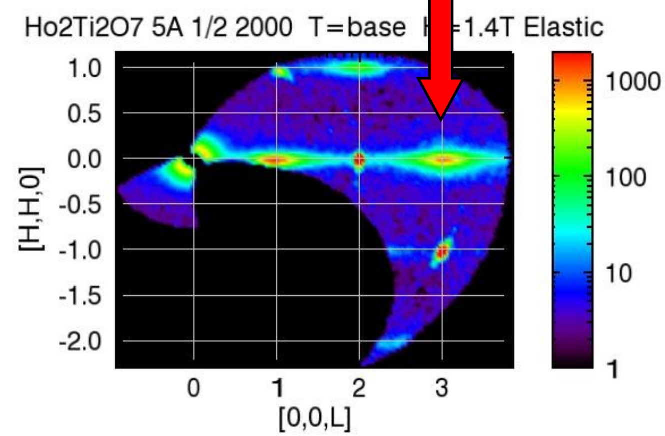
Ferro-ordering of α-chains

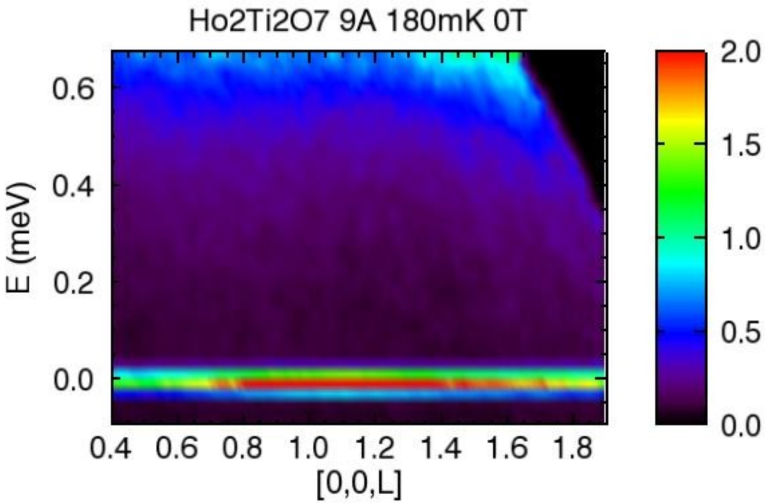


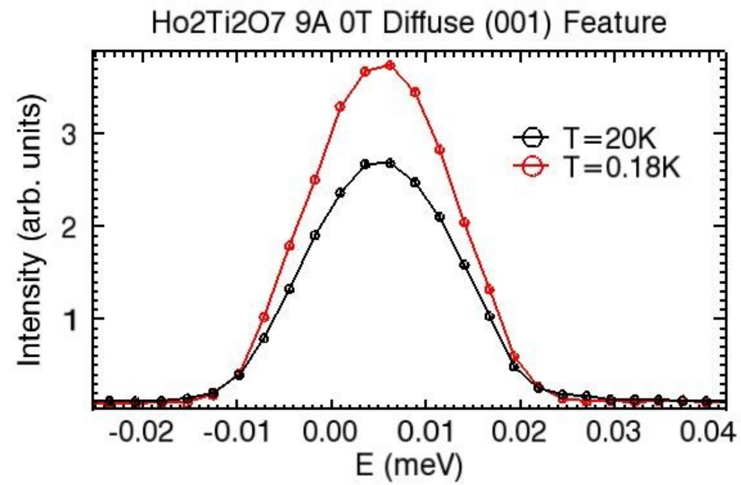




1D-correlations along β-chains

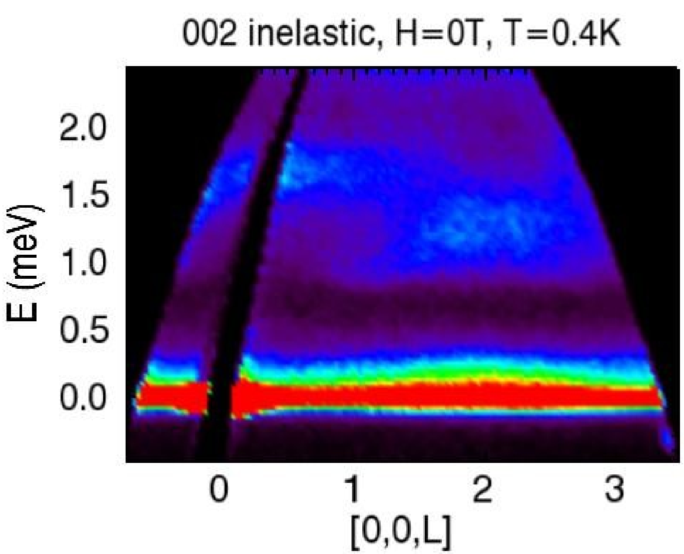


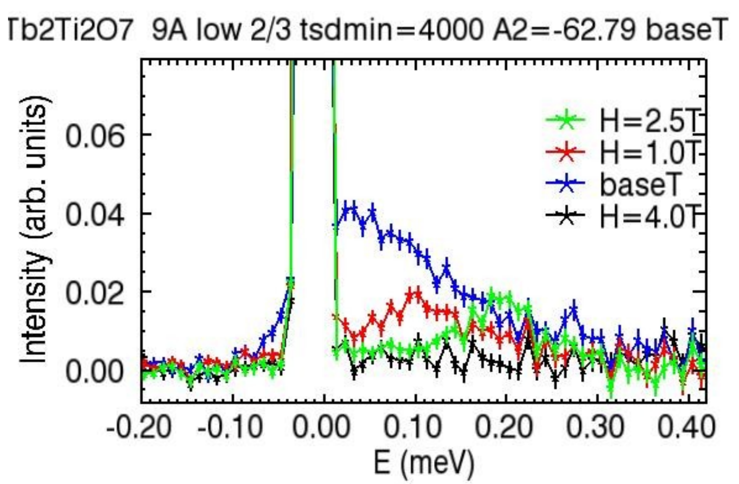




Spin Ice Ground State in $Ho_2Ti_2O_7$ is static

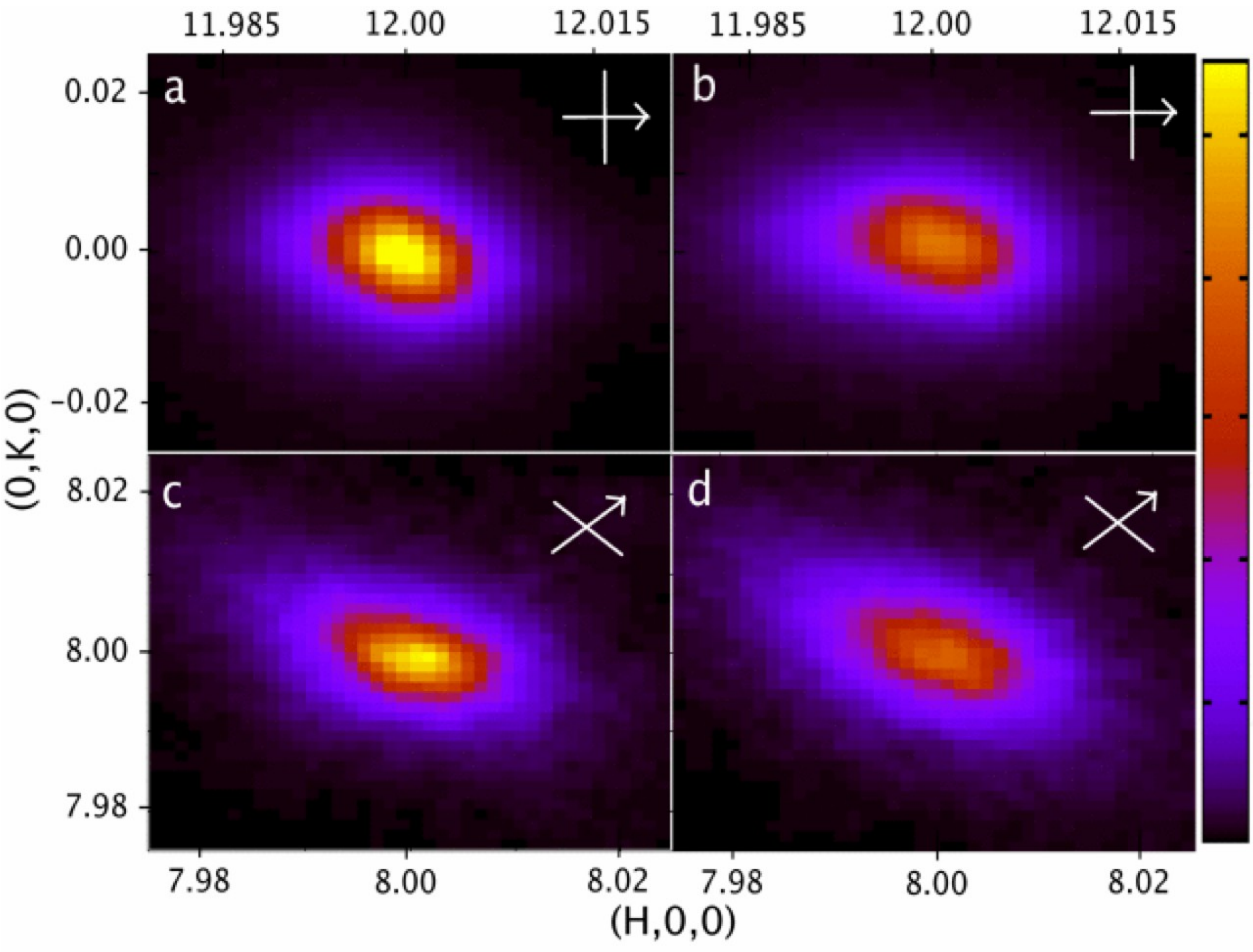
Spin Liquid Ground State in $Tb_2Ti_2O_7$ is dynamic

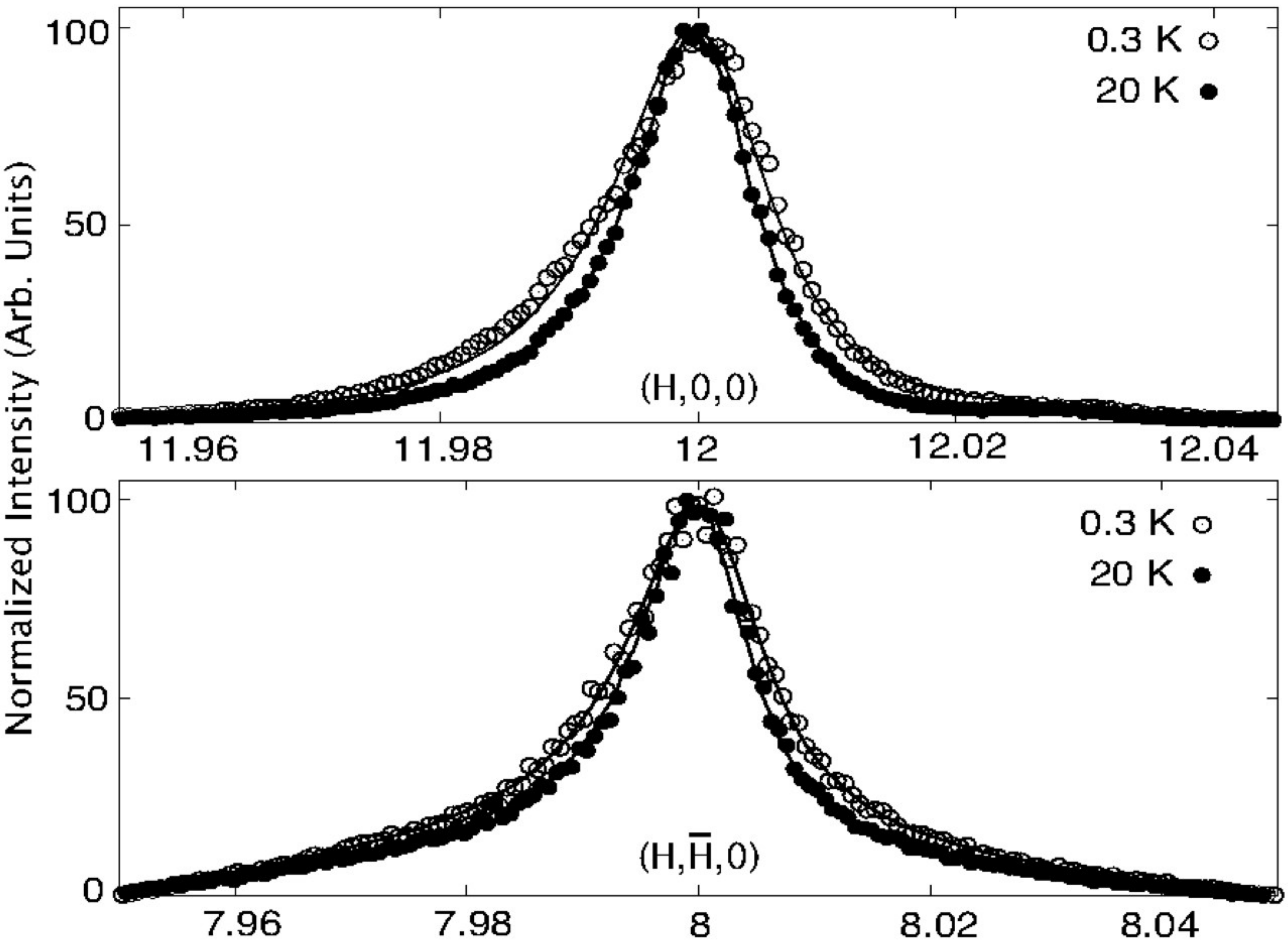




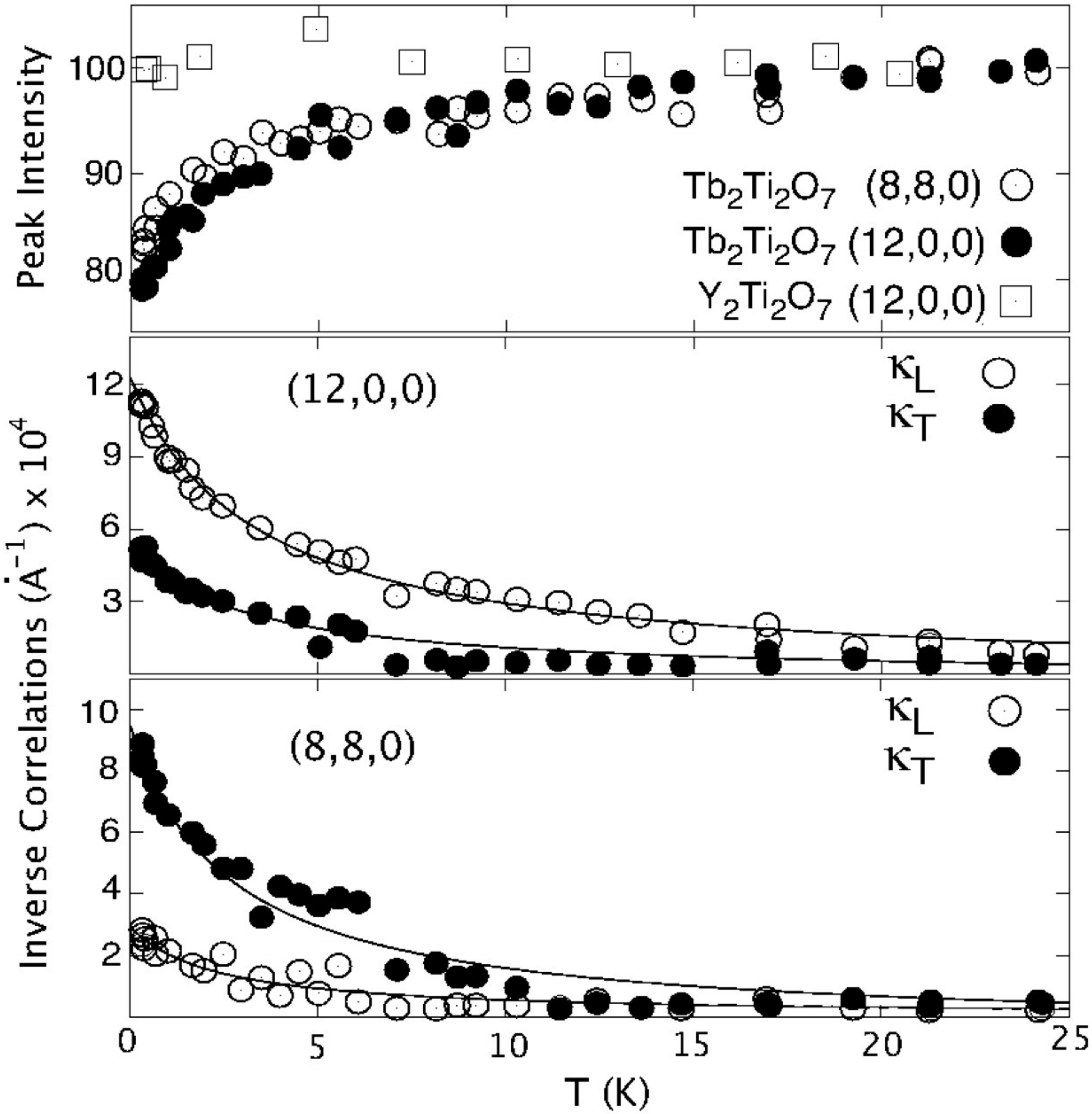
High Resolution *X-Ray* Scattering from $Tb_2Ti_2O_7$

Ruff et al., cond-mat/0707.1682, PRL to appear

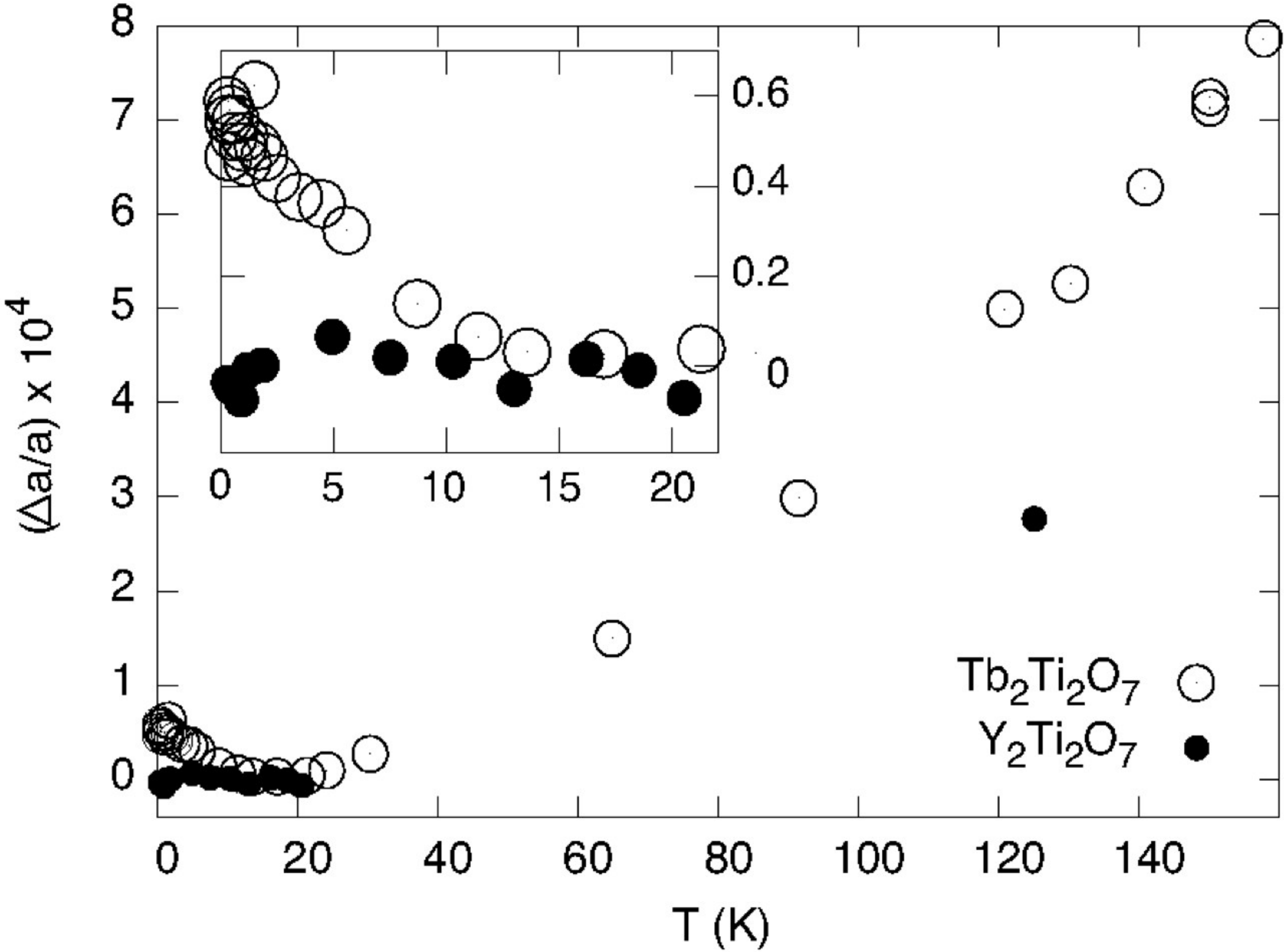




Normalized Intensity (Arb. Units)
100
50
0
0.3 K
20 K
(H,0,0)
11.96
11.98
12
12.02
12.04
100
50
0
0.3 K
20 K
(H,$\bar{H}$,0)
7.96
7.98
8
8.02
8.04



Peak Intensity
Tb2Ti2O7 (8,8,0)
Tb2Ti2O7 (12,0,0)
Y2Ti2O7 (12,0,0)
(12,0,0)
κL
κT
(8,8,0)
κL
κT
Inverse Correlations (Å^-1) x 10^4
T (K)



(Δa/a) x 10^4
T (K)
Tb2Ti2O7
Y2Ti2O7
0
1
2
3
4
5
6
7
8
0
20
40
60
80
100
120
140
0.6
0.4
0.2
0
0
5
10
15
20

- ***Conclusions:***

- **New neutron scattering infrastructure leads to New Science**
  DCS has been a *tremendous success* in elucidating exotic magnetism
  New time-of-flight neutron infrastructure at SNS, JSNS, 2TS@ISIS

- **Antiferromagnetic Pyrochlore $Tb_2Ti_2O_7$:**

  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 $Tb_2Ti_2O_7$

- **"Ferromagnetic" Pyrochlore $Ho_2Ti_2O_7$:**

  Static Spin Ice Ground State