Frustrated, Satisfied and Fluctuating Ground States in Pyrochlore Magnets

B. D. Gaulin

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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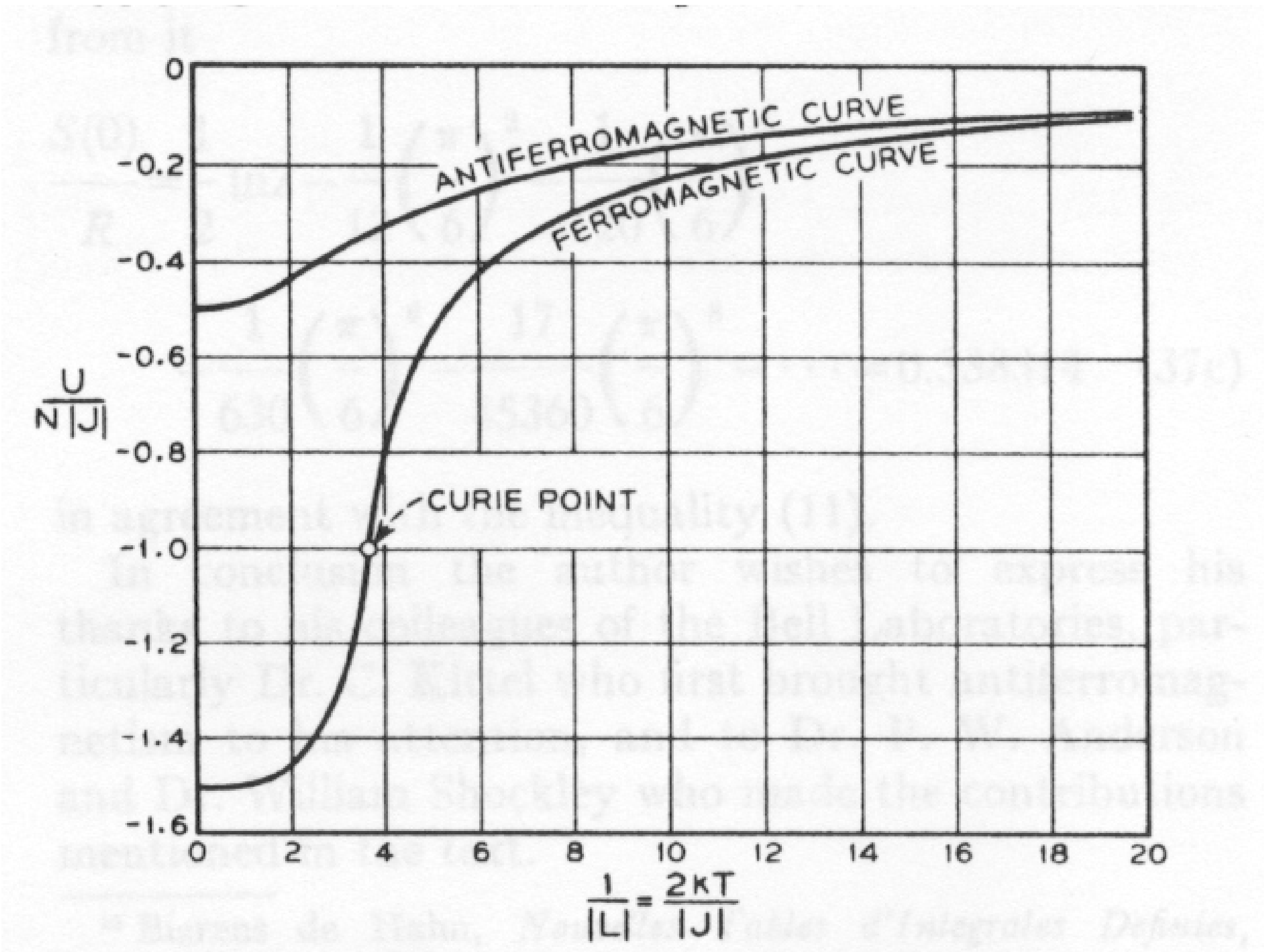
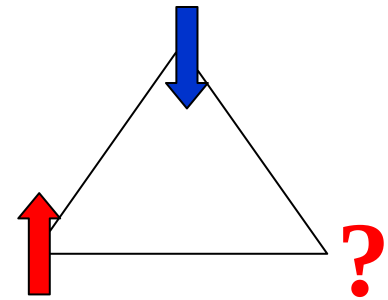
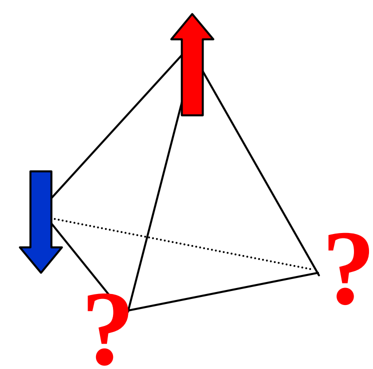
- **J.R.D. Copley**

now at : HMI Berlin[1]

Geometrical Frustration:
Antiferromagnetism + Triangles and Tetrahedra

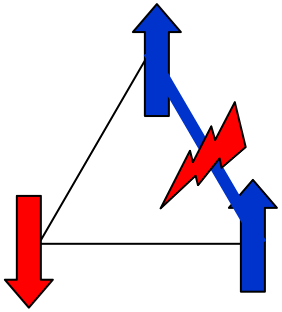


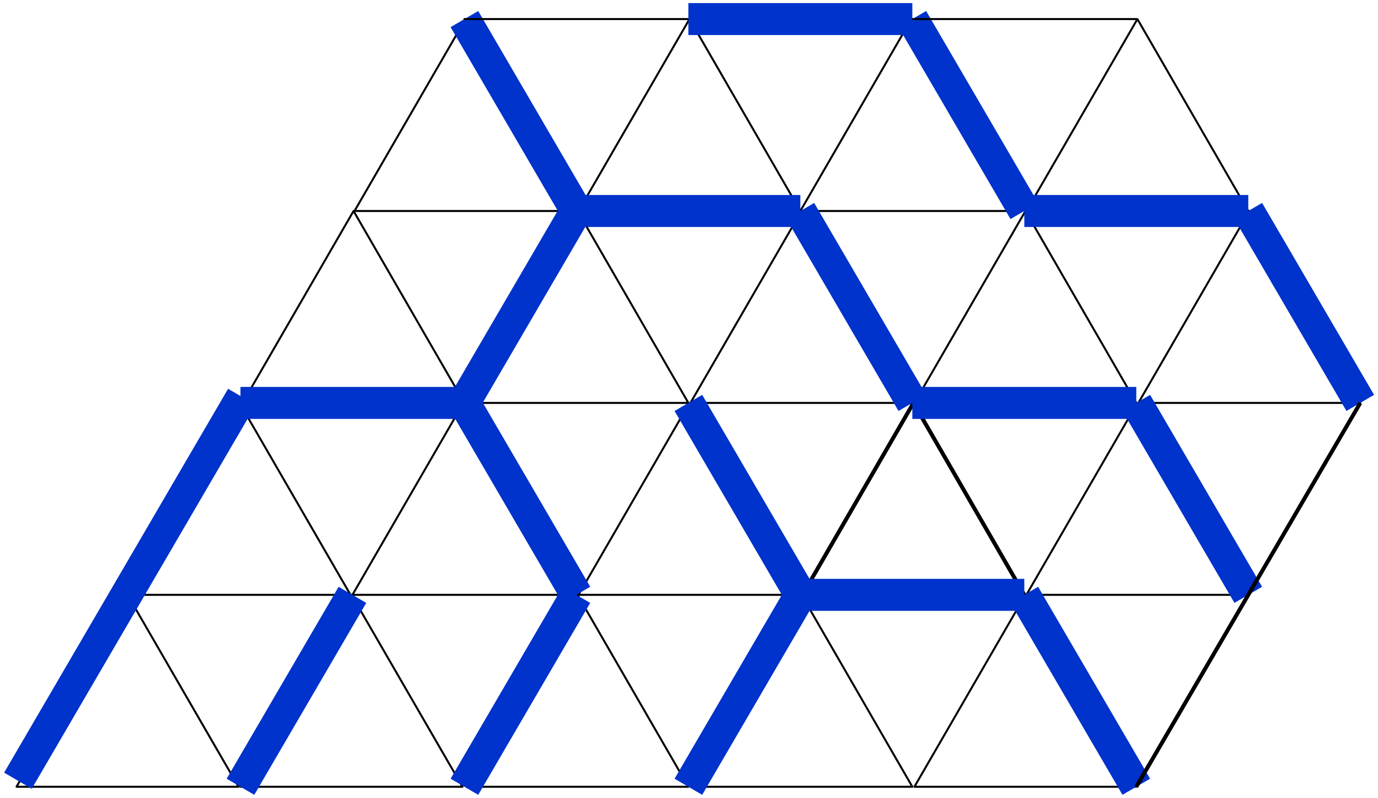




2D Triangular Lattice: G.H. Wannier Phys. Rev. 79, 357, 1950.

Ground State of the Ising AF on a Triangular Lattice

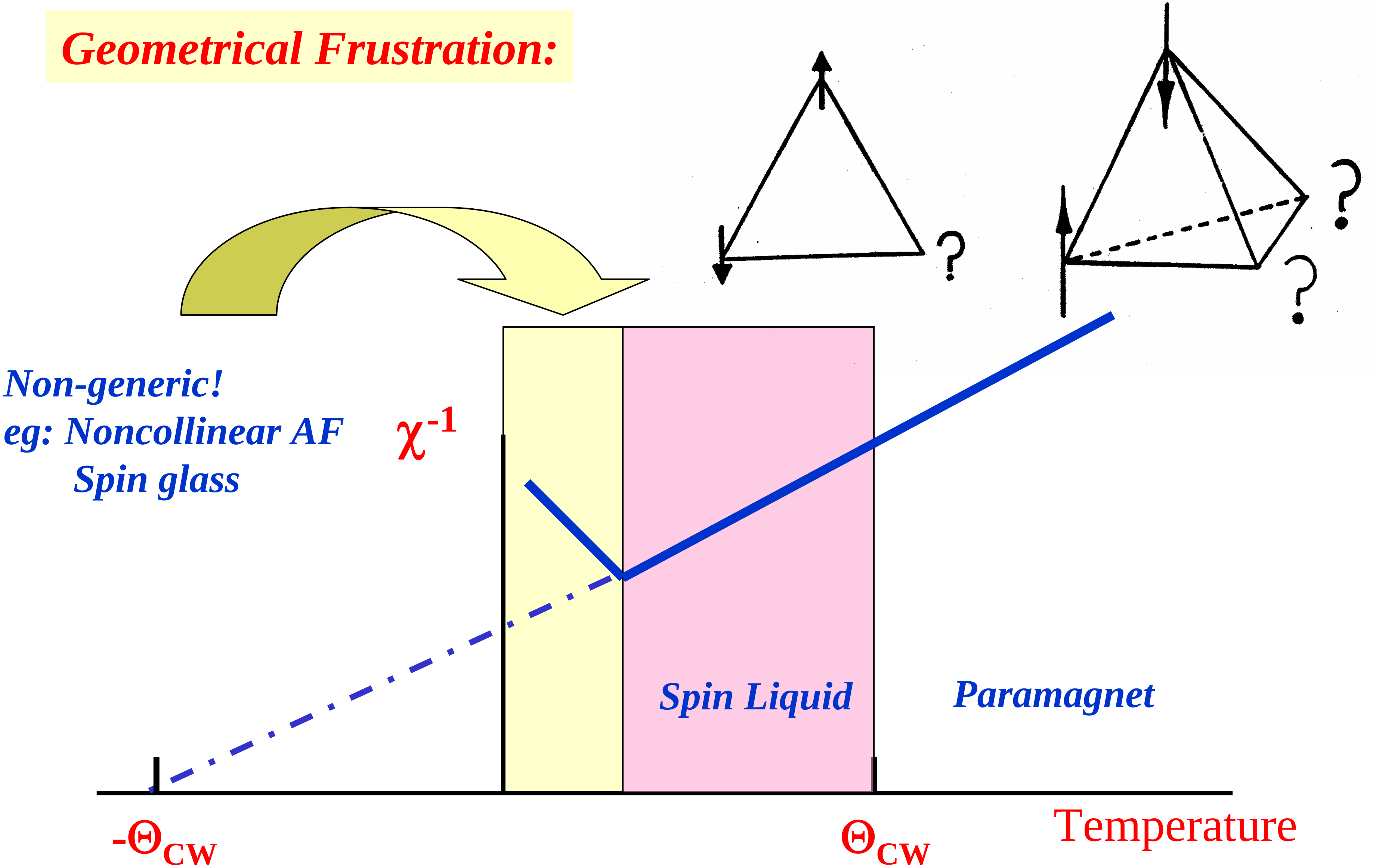




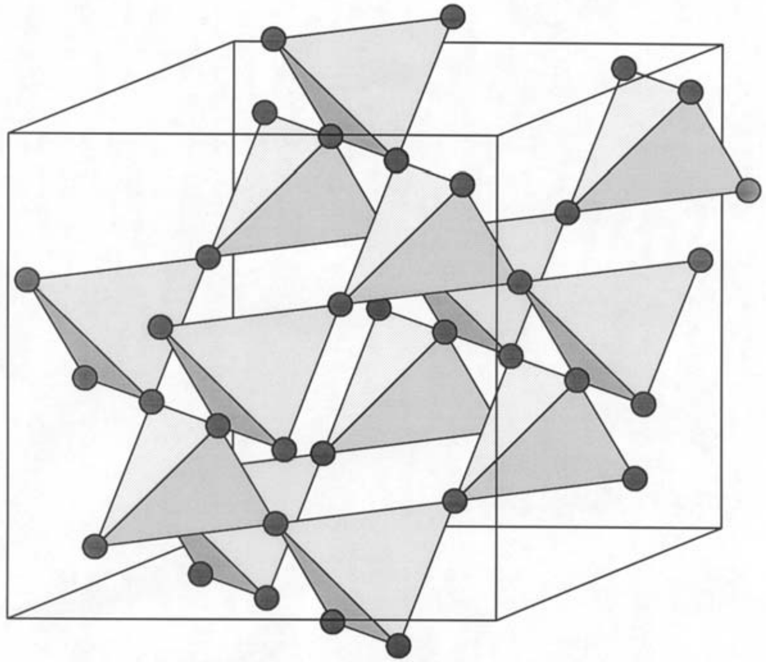
Entropy at T=0 is finite ~ 0.34 R

No LRO at any temperature

Geometrical Frustration:



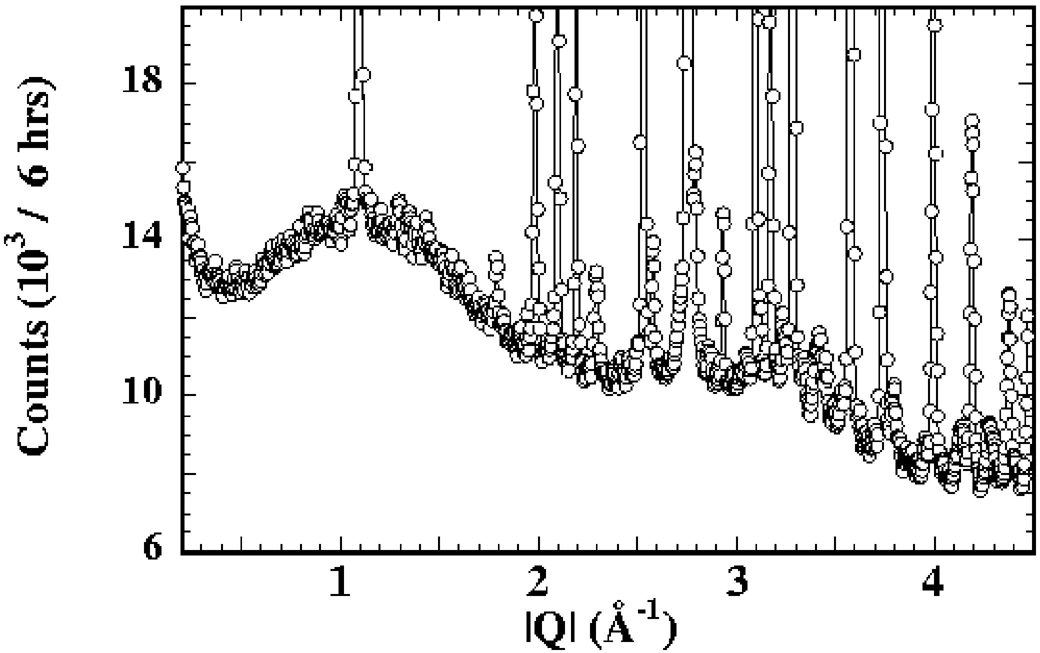
Mean Field Theory predicts a phase transition near T=| Θ_{CW} | ,
but materials remains disordered to much lower temperatures – <u>Spin Liquid</u>



Frustration in three dimensions:

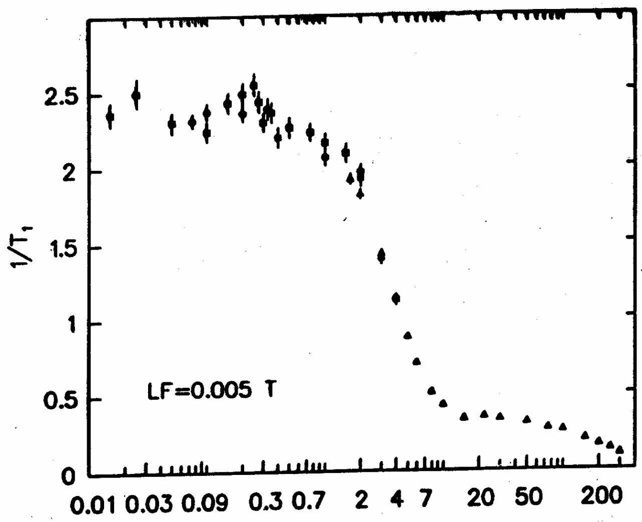
The cubic pyrochlore structure;
A network of corner-sharing tetrahedra

Low temperature powder neutron diffraction from $Tb_2Ti_2O_7$

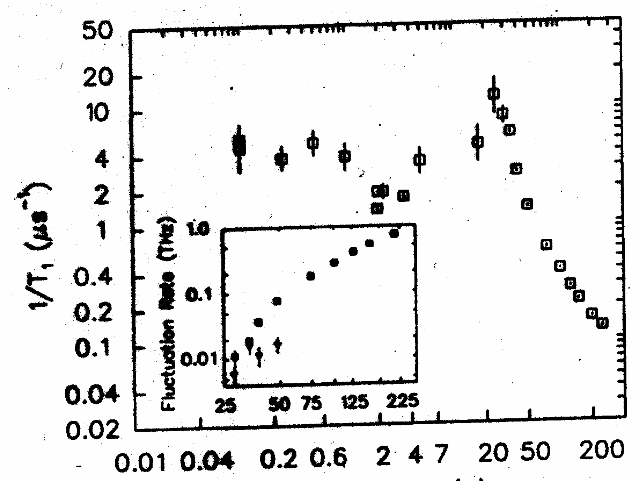


μSR Studies of Magnetic Ground States in:

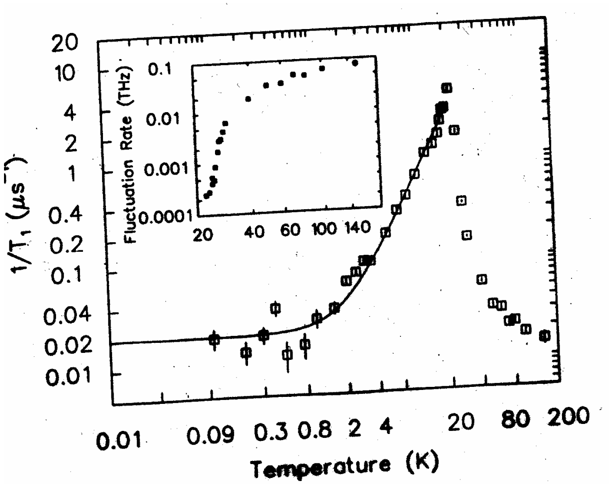
$Tb_2Ti_2O_7$
$Tb_2Mo_2O_7$
$Y_2Mo_2O_7$



$Tb_2Ti_2O_7$: *Spin Liquid*

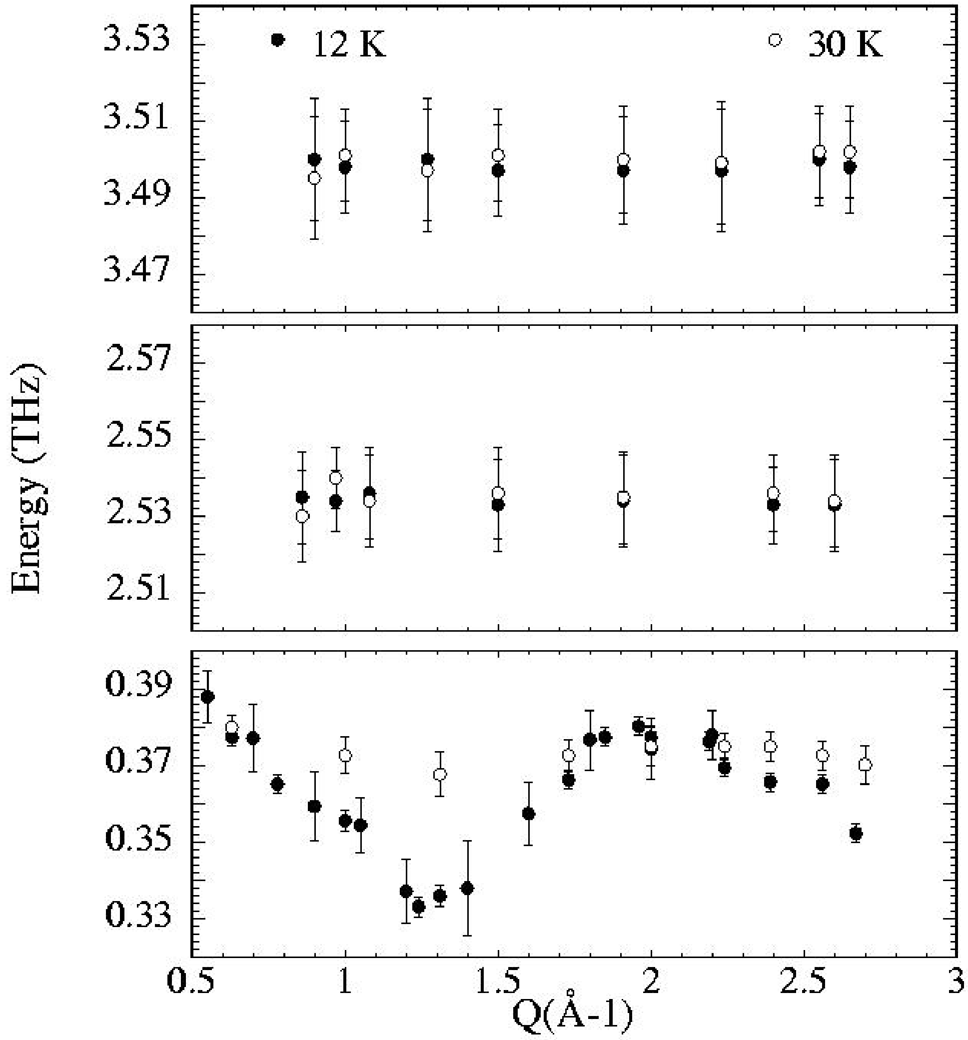


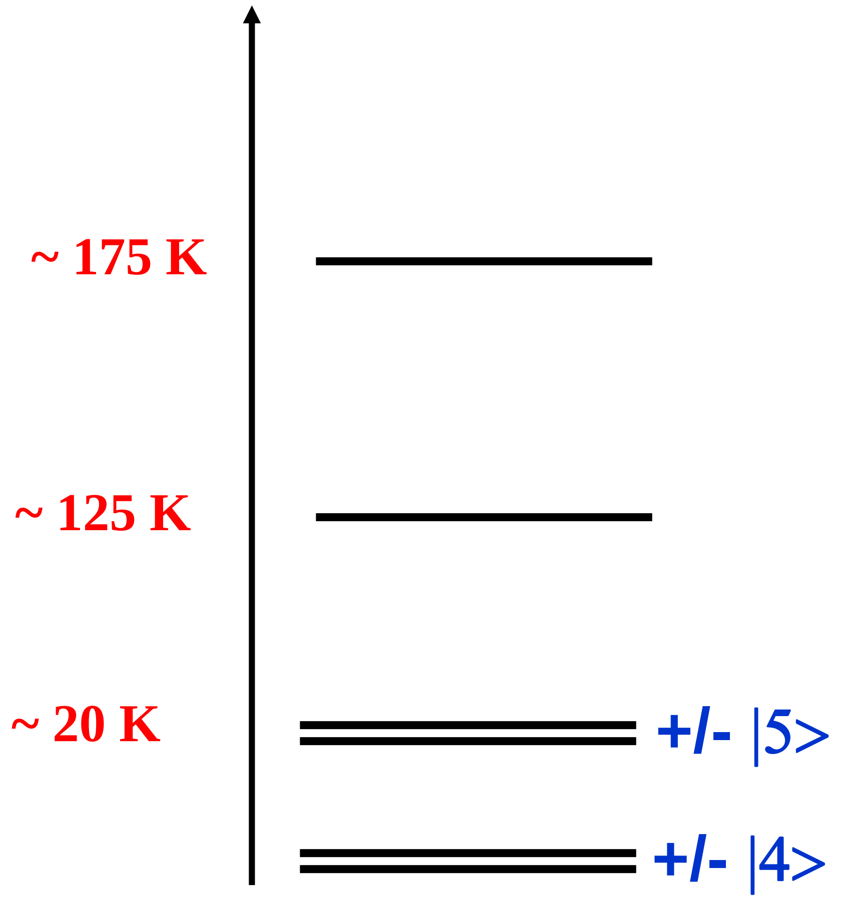
$Tb_2Mo_2O_7$: *Spin Liquid and Spin Glass*



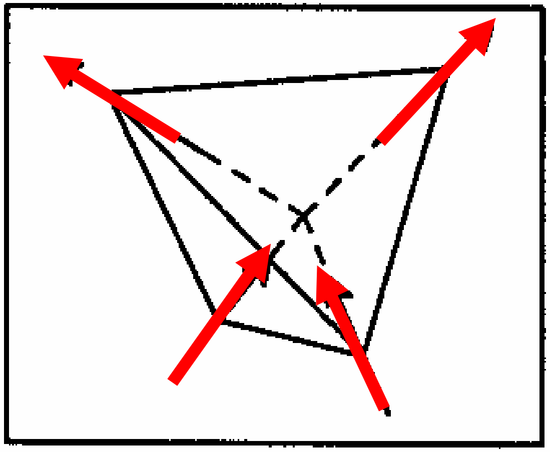
$Y_2Mo_2O_7$: *Spin Glass*

Inelastic neutron scattering on polycrystalline $Tb_2Ti_2O_7$



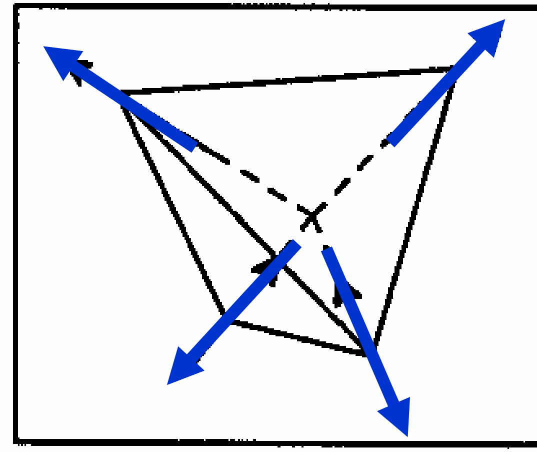


(Δ : $Ho_2Ti_2O_7$ ~ 240 K ; $Dy_2Ti_2O_7$ ~ 380 K)



Rare Earth moments:

Strong [111] anisotropy



Ferromagnetic exchange:

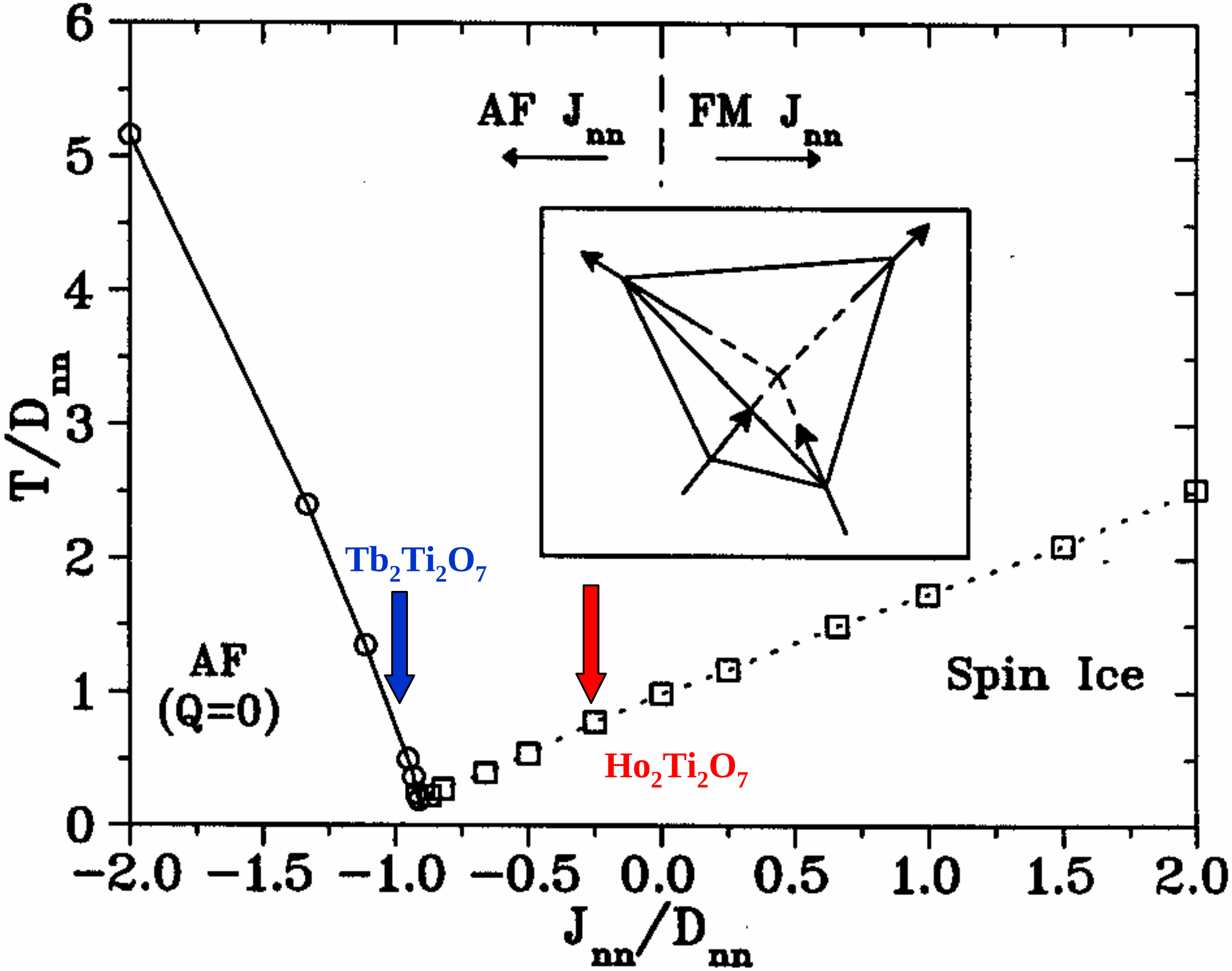
"Spin Ice" : 2 in 2 out

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

Antiferromagnetic exchange:

All in - All out

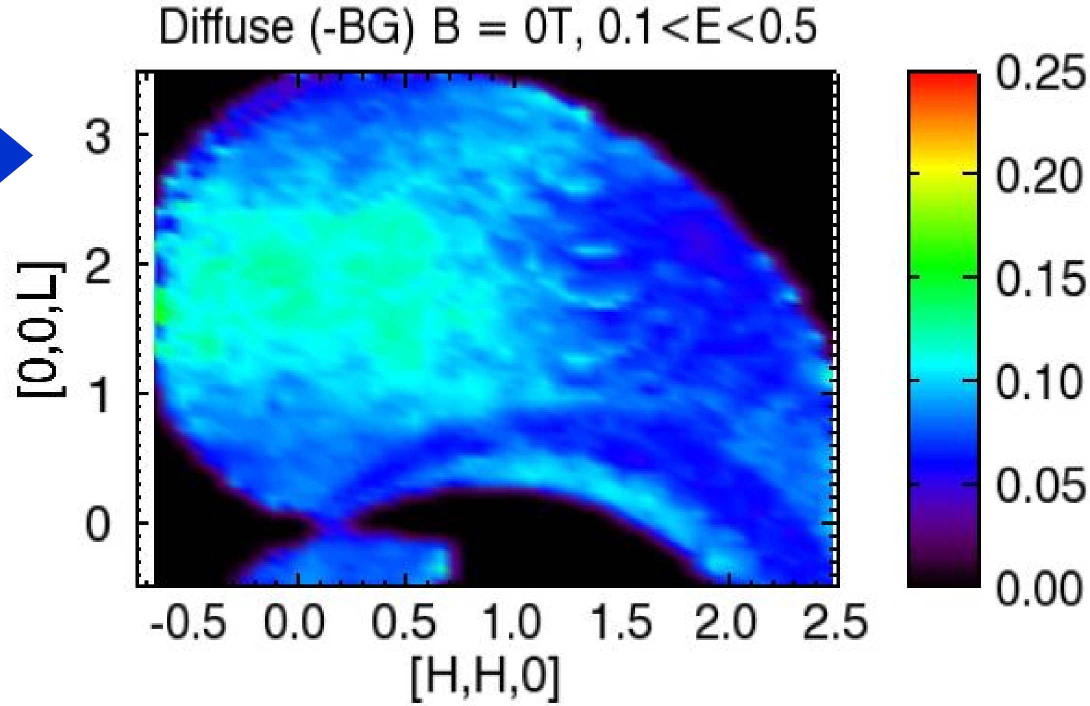
$$H = -J \sum_{\langle ij \rangle} \mathbf{S}_i^{z_i} \cdot \mathbf{S}_j^{z_j} + D r_{\mathrm{nn}}^3 \sum_{j>i} \frac{\mathbf{S}_i^{z_i} \cdot \mathbf{S}_j^{z_j}}{|\mathbf{r}_{ij}|^3} - \frac{3(\mathbf{S}_i^{z_i} \cdot \mathbf{r}_{ij})(\mathbf{S}_j^{z_j} \cdot \mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^5}$$

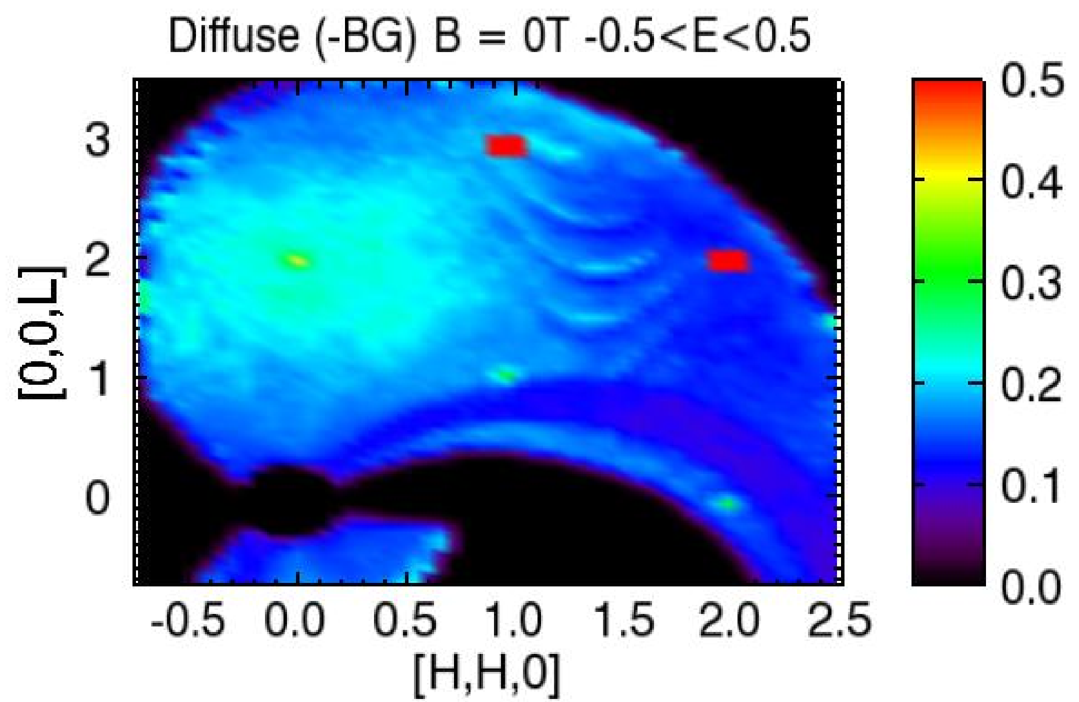


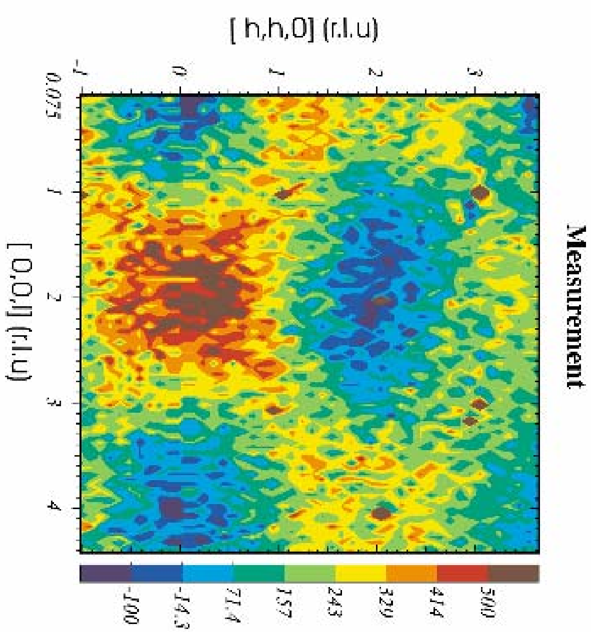
B.C. den Hertog and M.J.P. Gingras, PRL, 84, 3430, 2000.

TOF scattering
T=0.4 K

2-Axis Diffraction
T=9 K

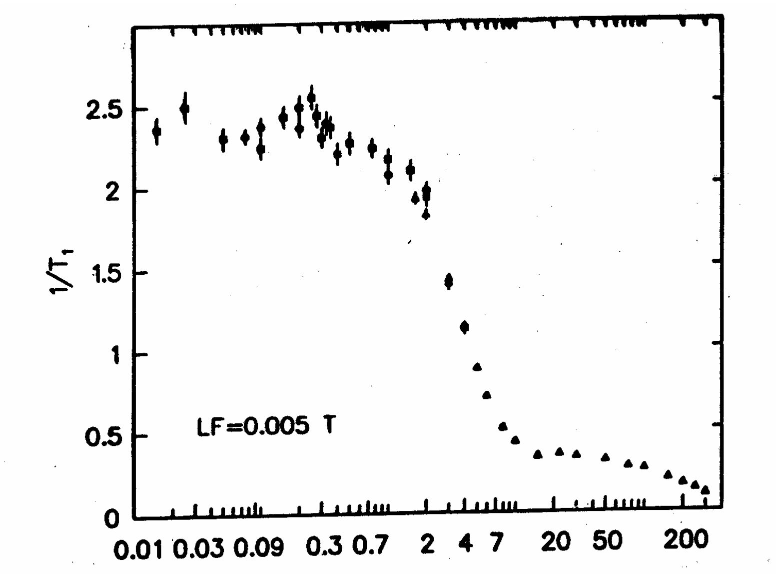


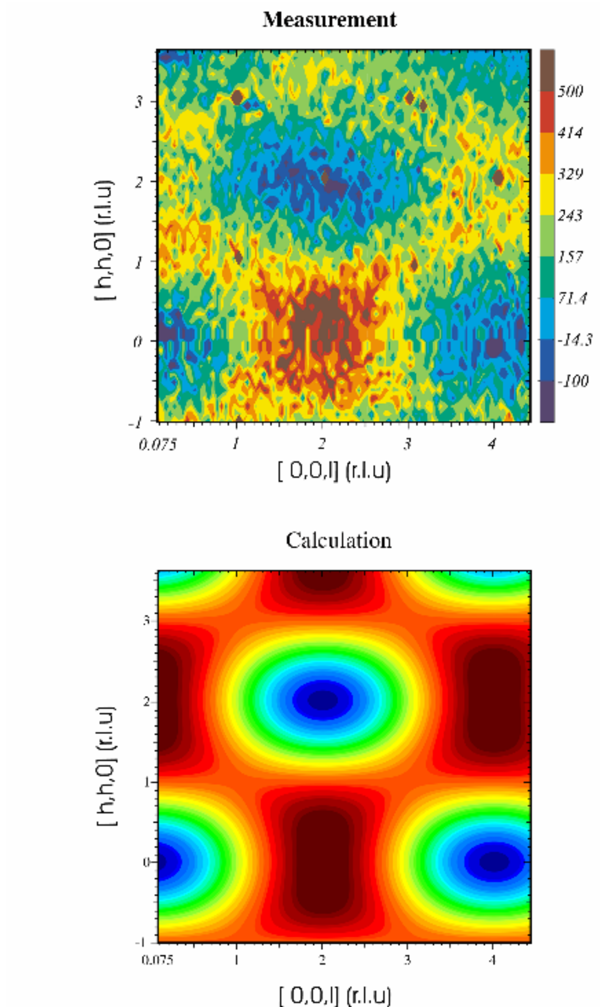




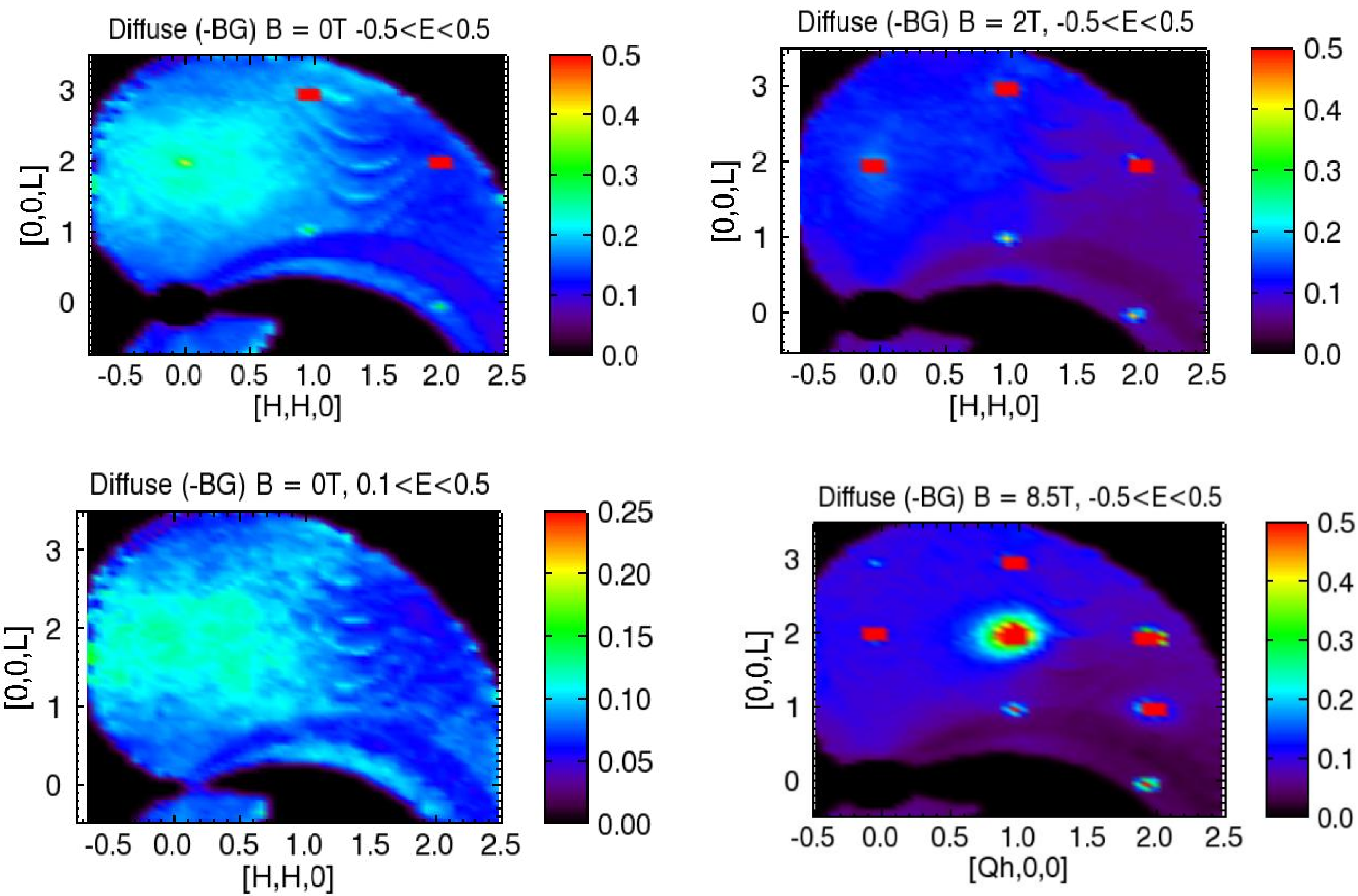
Outstanding Questions:

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

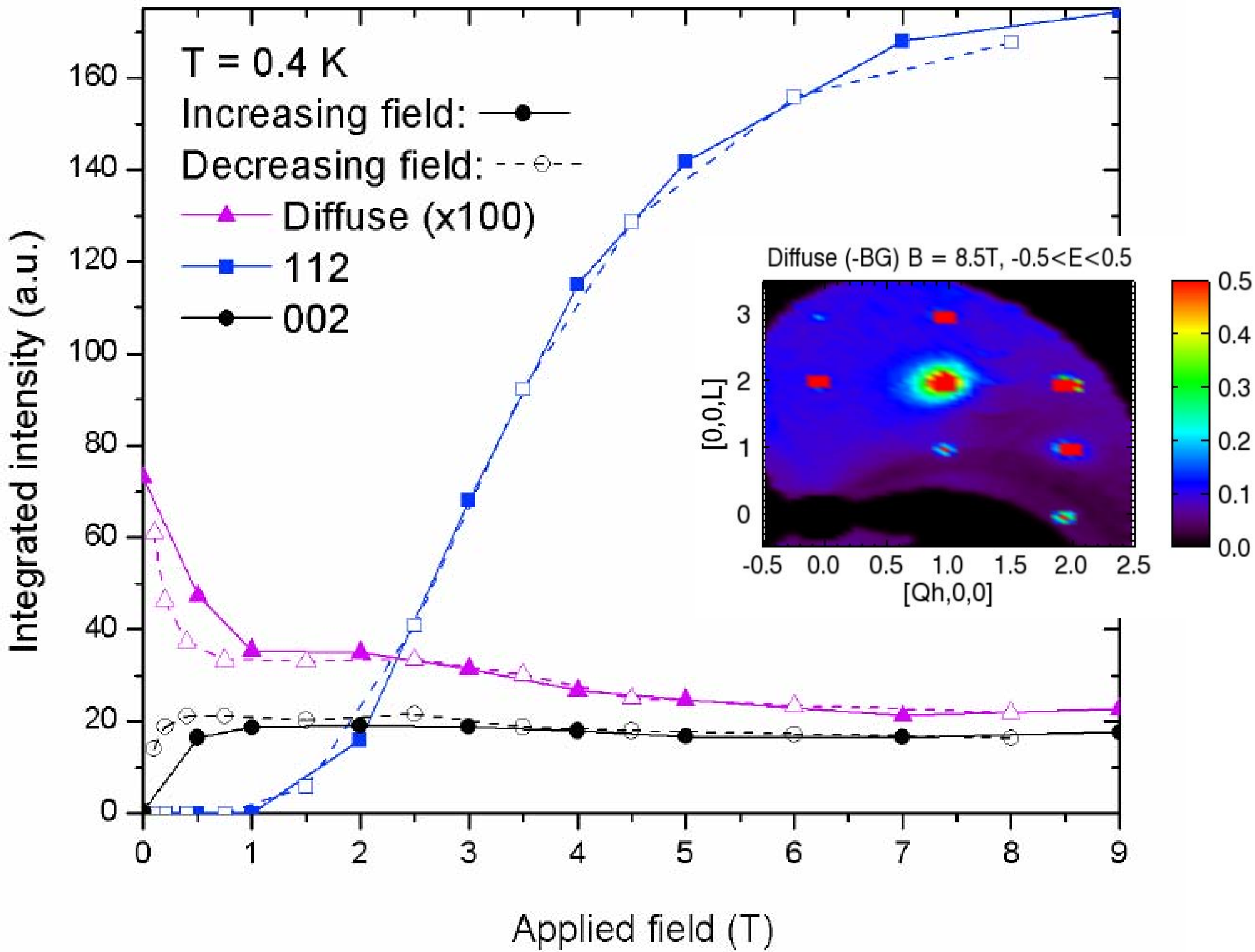




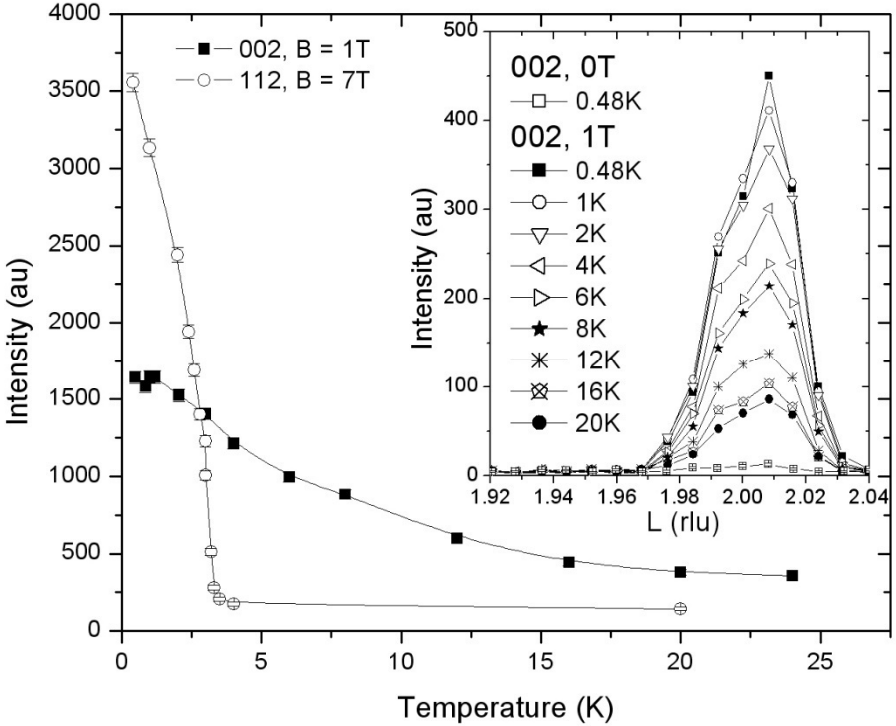
- **S(Q) at low T is (perhaps naively) incompatable with Ising (111) anisotropy. What is going on?**

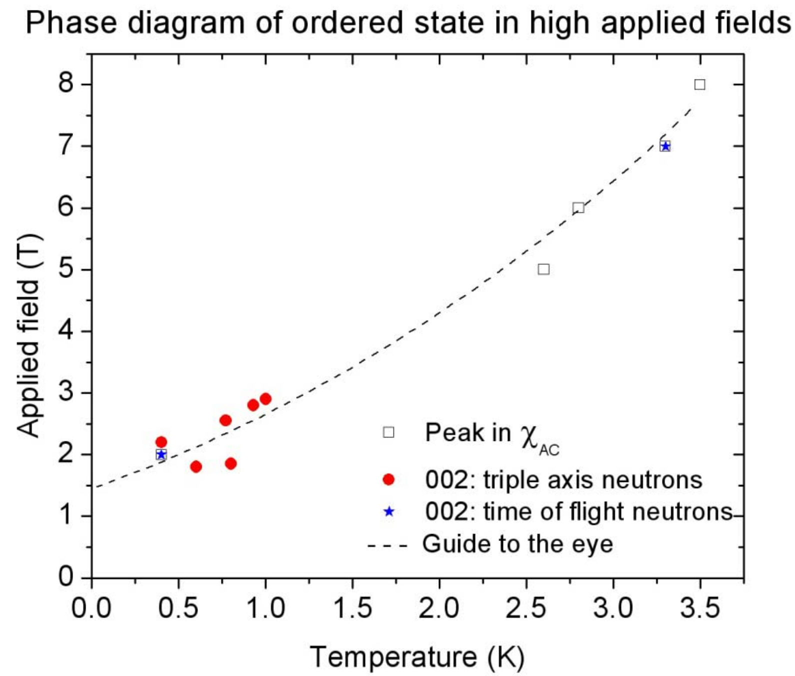


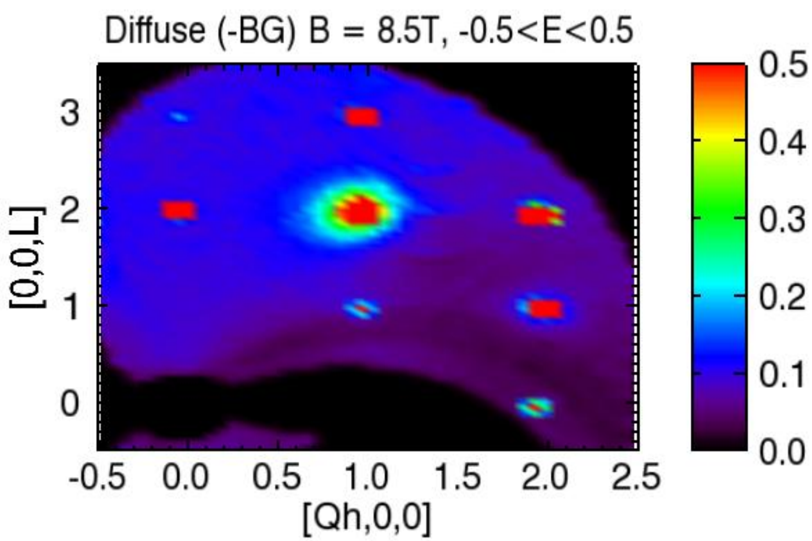
Ordered phases appear on application of H // 110



T = 0.4 K
Increasing field:
Decreasing field:
Diffuse (x100)
112
002
Integrated intensity (a.u.)
Applied field (T)
Diffuse (-BG) B = 8.5T, -0.5<E<0.5
[0,0,L]
[Qh,0,0]

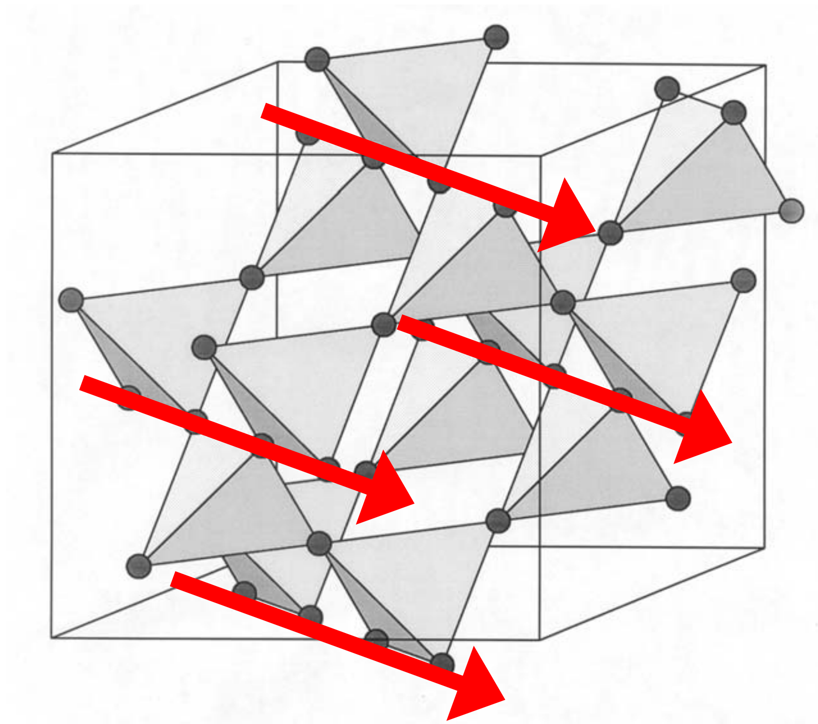






Low field (002) phase persists to very high T > 25 K

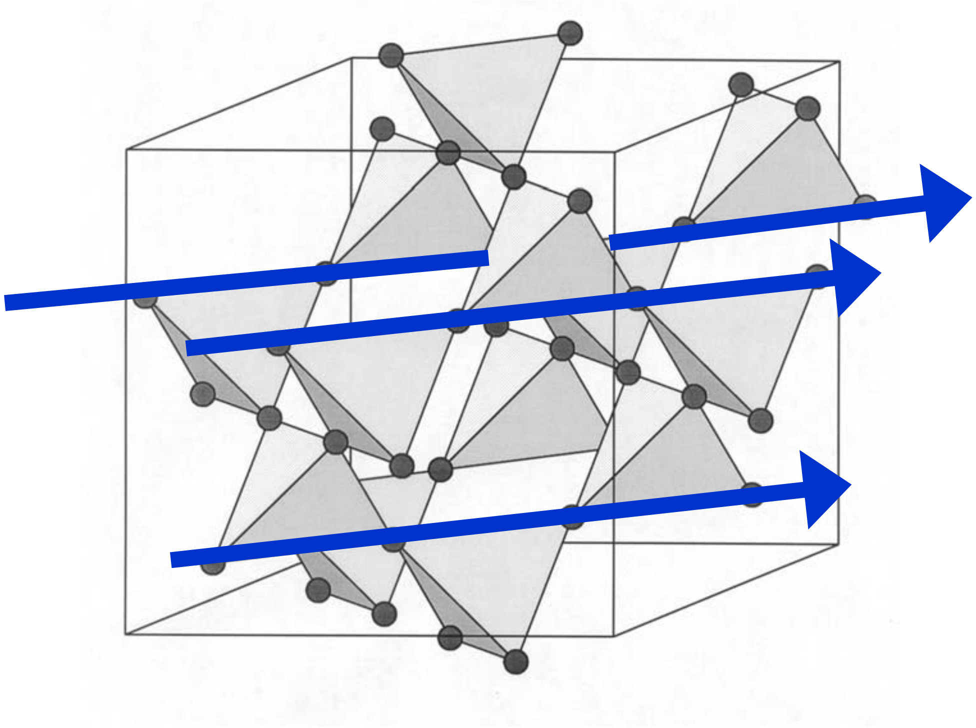
High field (112) phase exists on expected T_N ~ 2 K



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

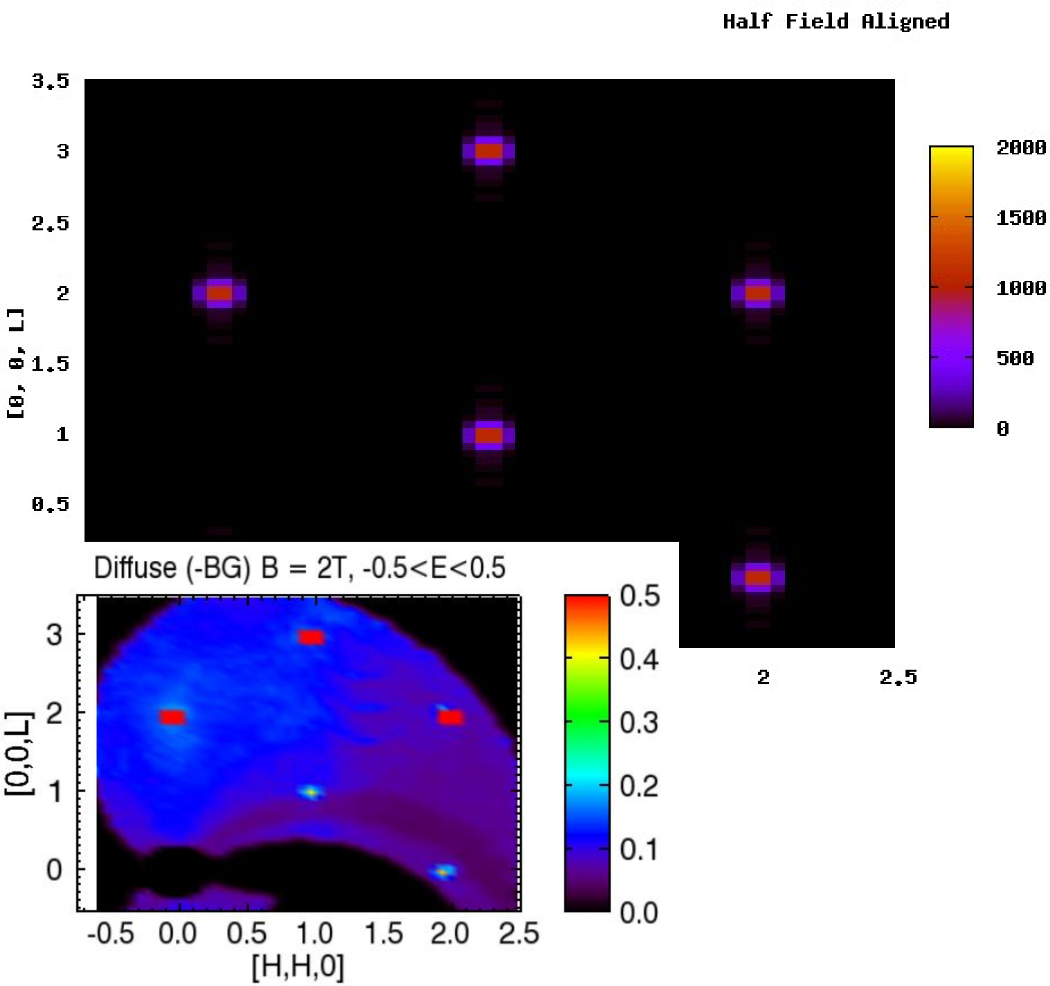


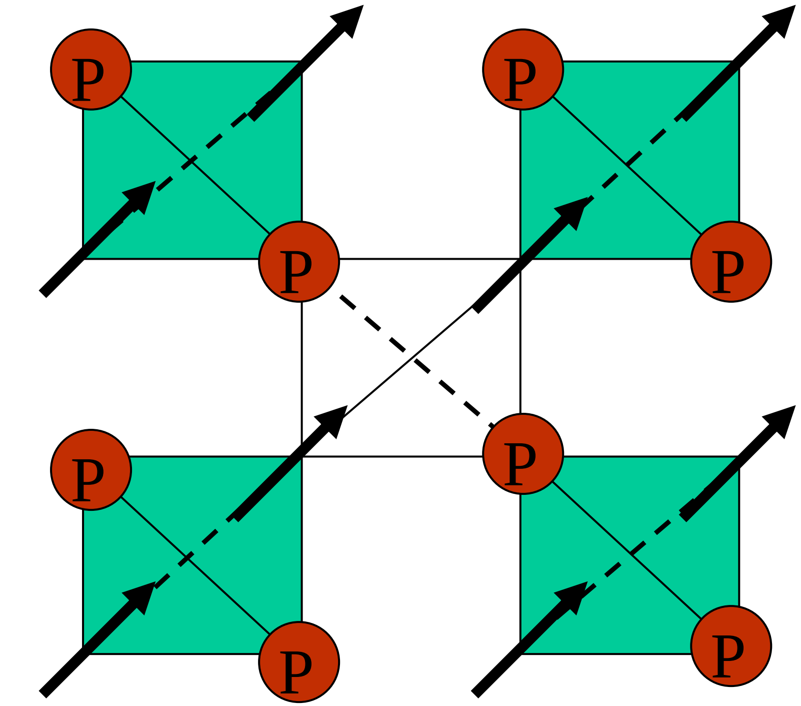
Application of weak [1-10] magnetic field breaks system up into α and β chains.

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

Perpendicular β-[110] chains

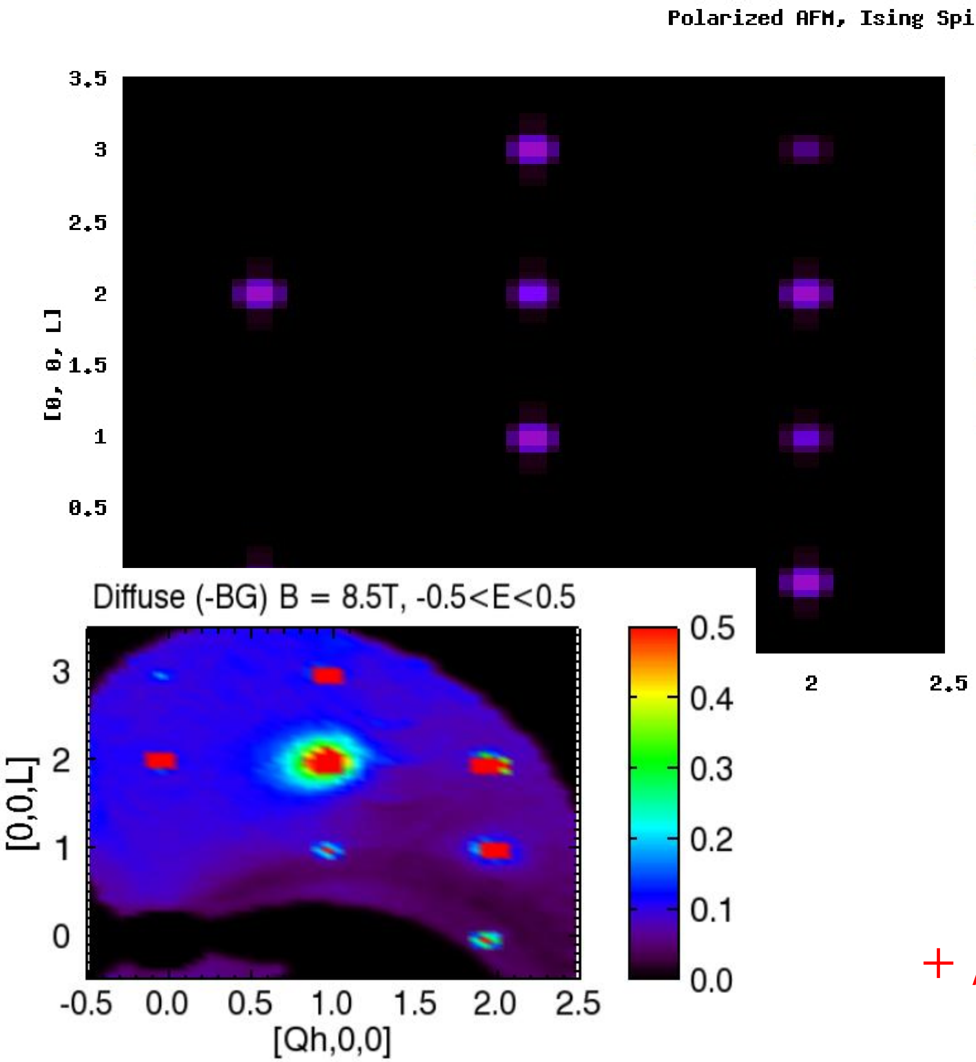
α-chains polarized along the [1,1,0] Direction

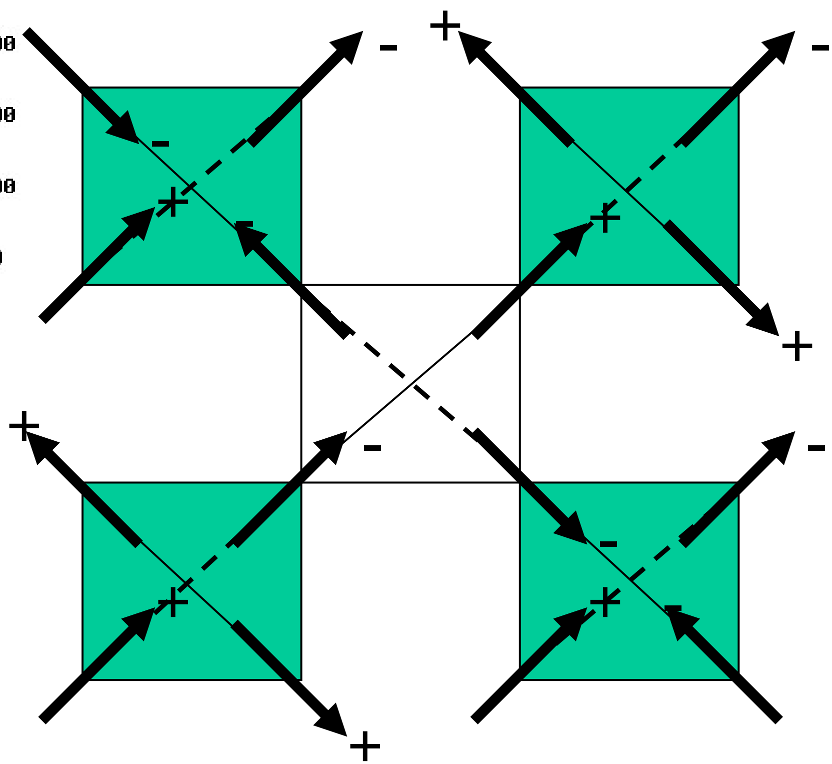




P = Paramagnetic Site

Field Aligned, Local <111> AFM

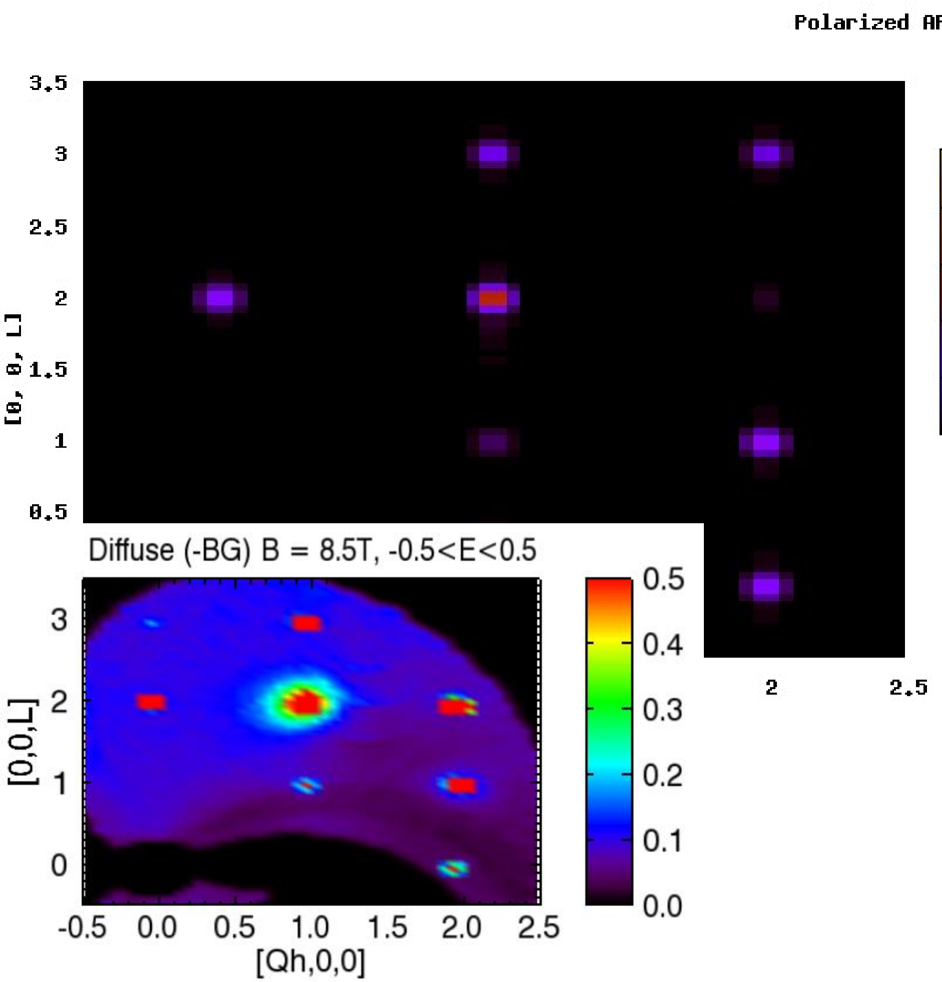


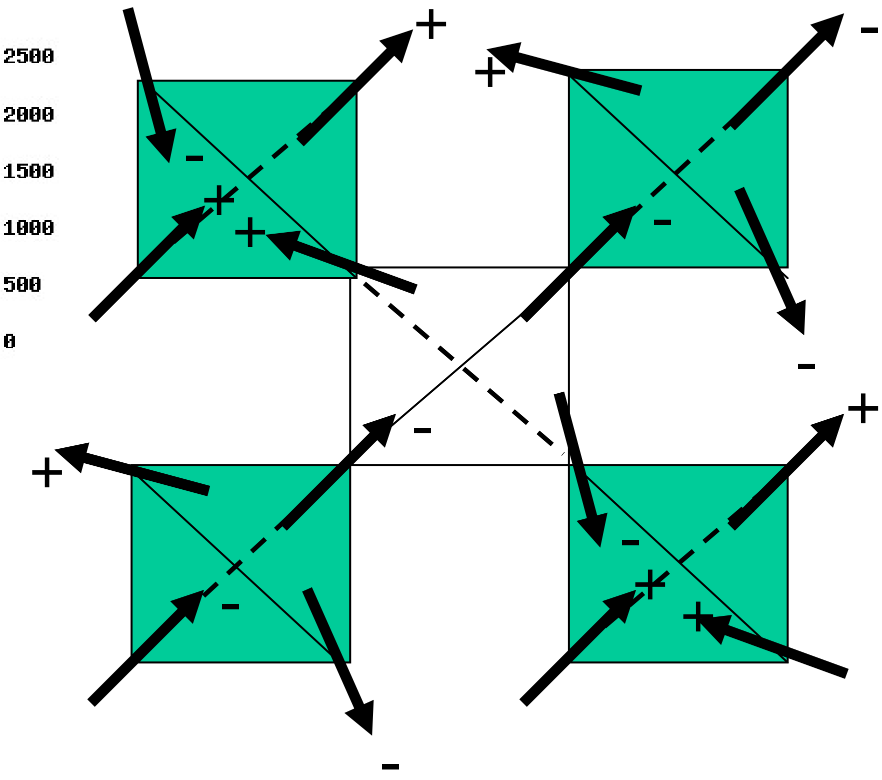


+ / - Denotes Z-Component

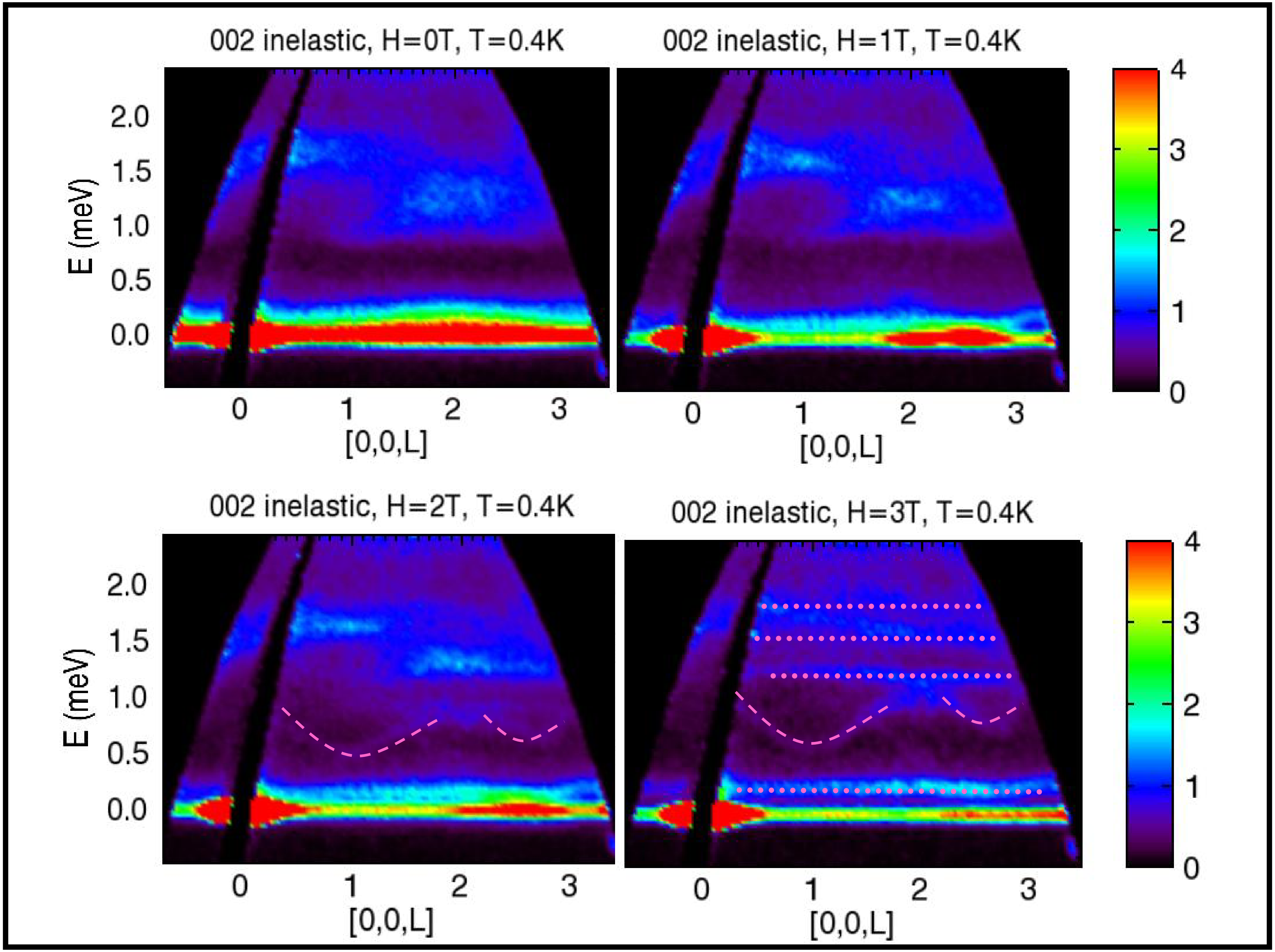
"3 in – 1 out" local structure

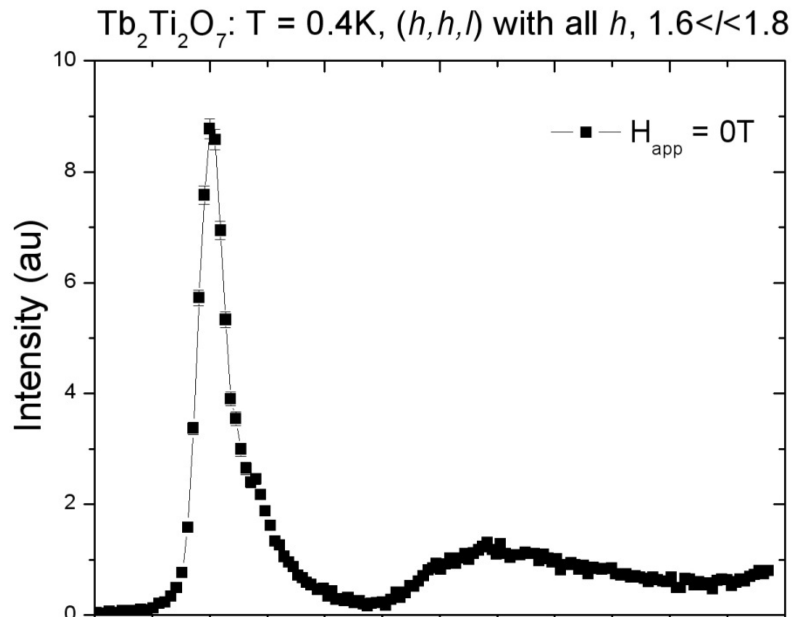
Canted AFM, Moments not respecting Ising 111 axes.
(Magnetization is ~11% of saturation along [110])



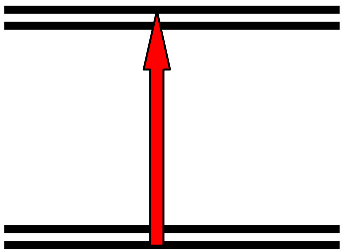


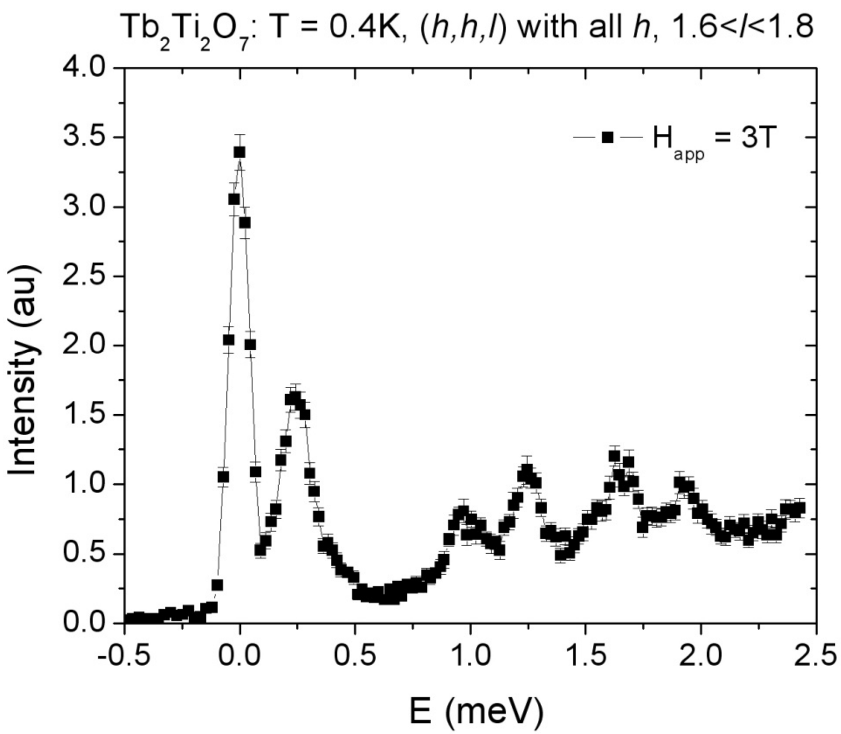
+ / - Denotes Z-Component


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)
2.0
1.5
1.0
0.5
0.0
0
1
2
3
[0,0,L]
4
3
2
1
0

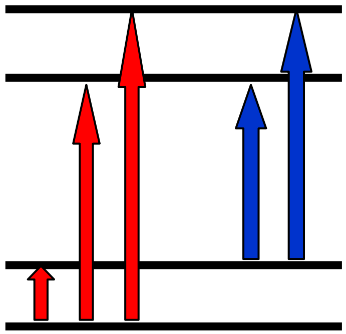


One Transition in Zero Field

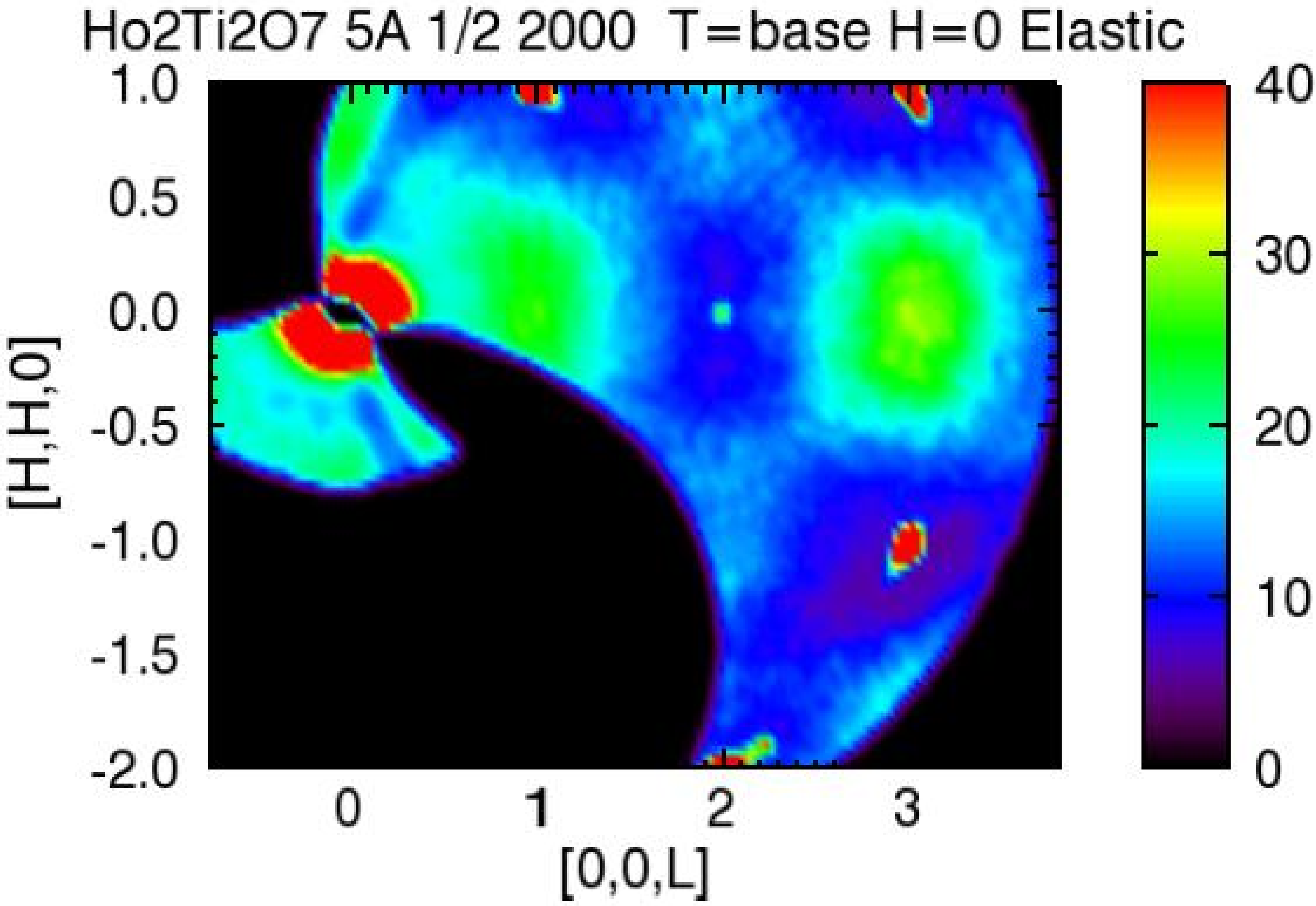




Five Transitions in Non-Zero Field

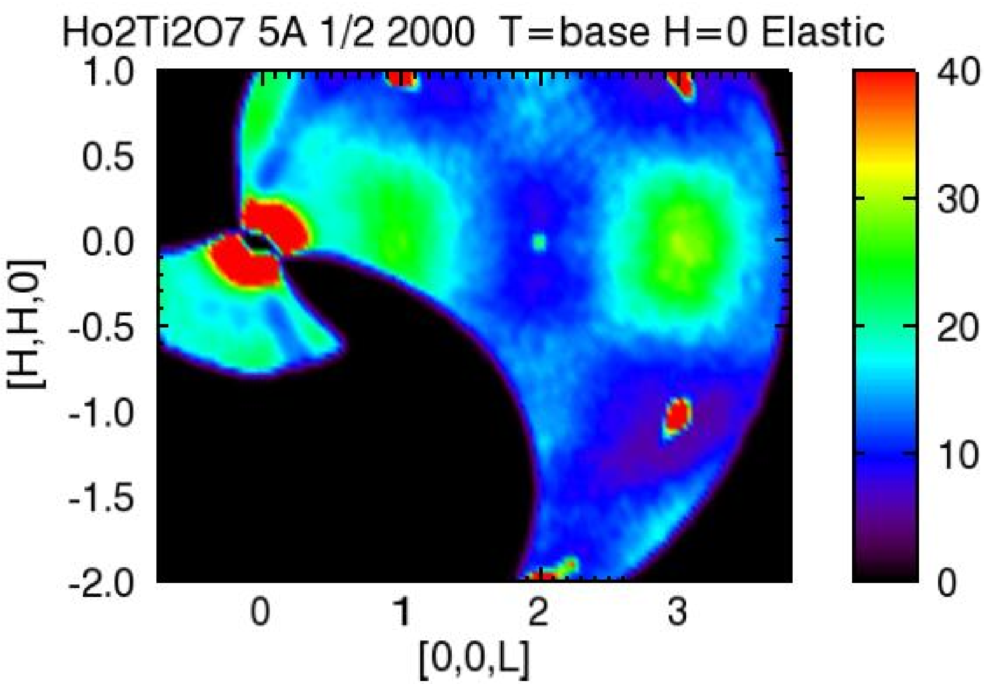


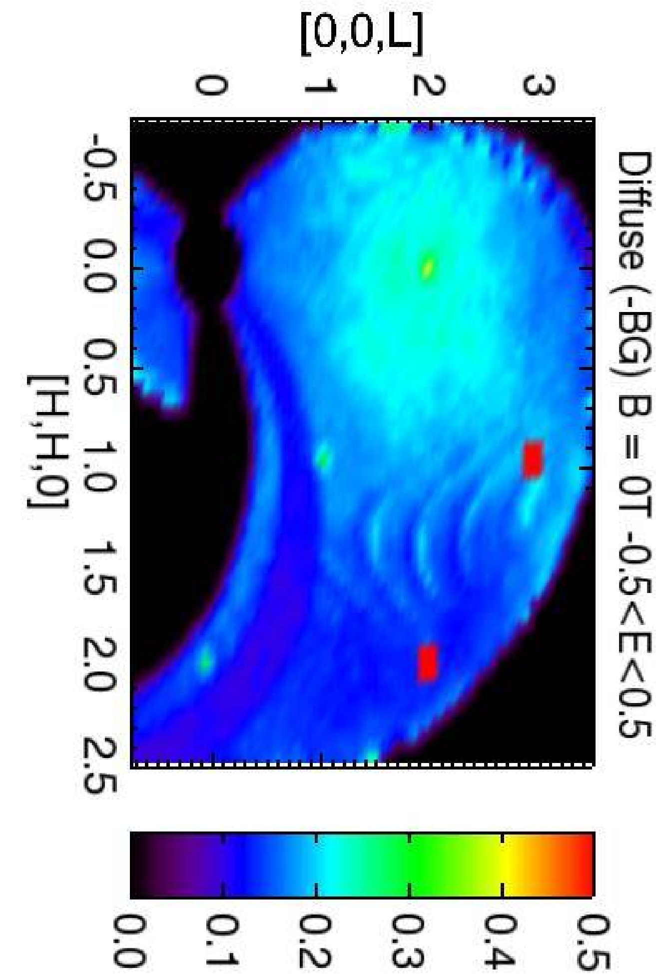
Spin Ice Ground State in $Ho_2Ti_2O_7$



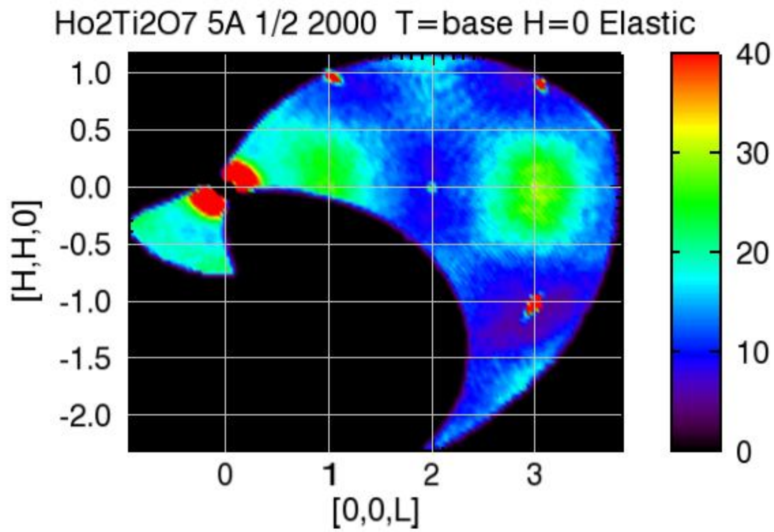
$Ho_2Ti_2O_7$ vs $Tb_2Ti_2O_7$

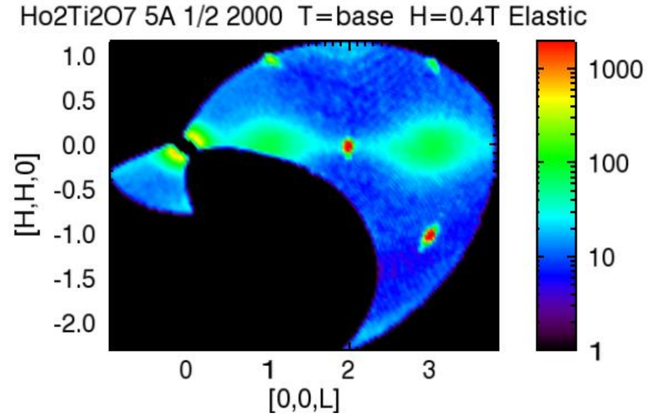
Static Spin Ice vs Dynamic Spin Liquid (H=0)

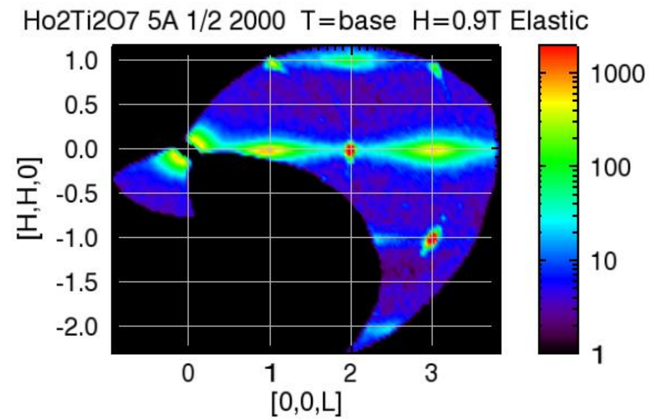




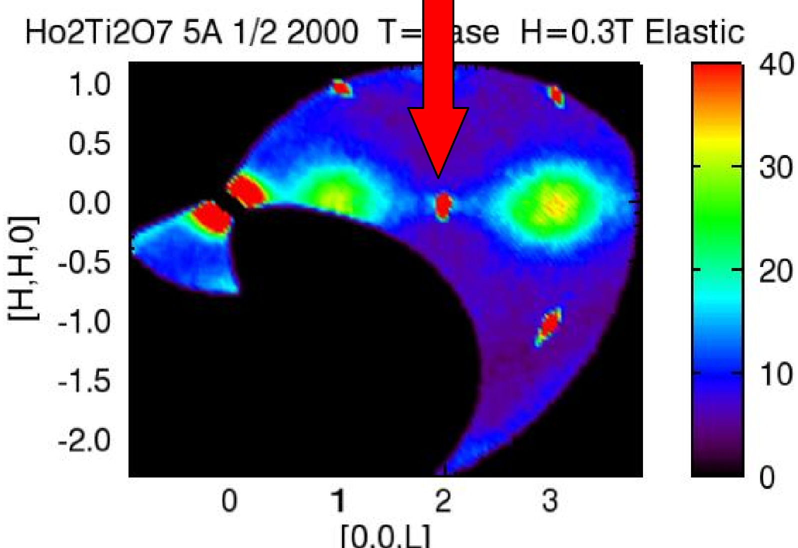
Magnetic Structure Factors appear complementary



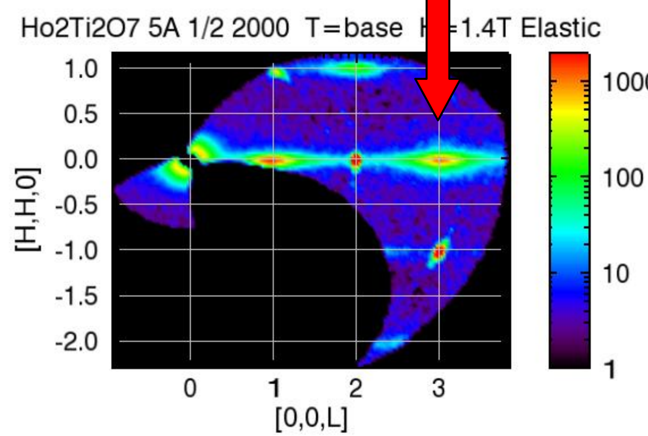


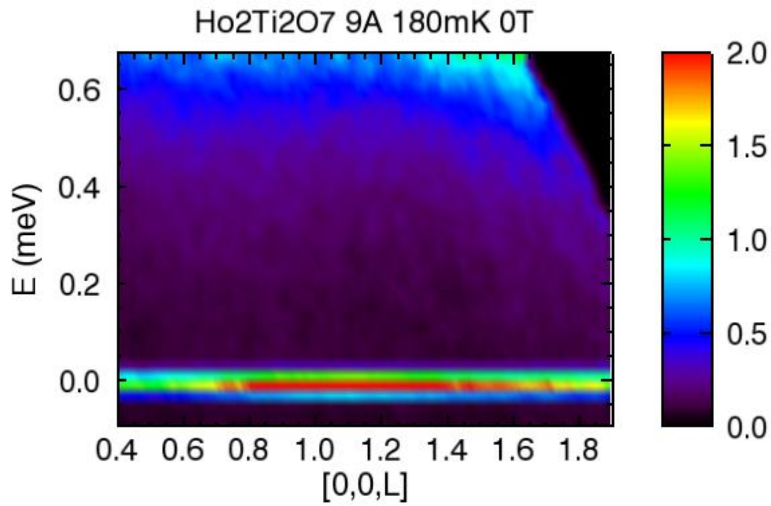


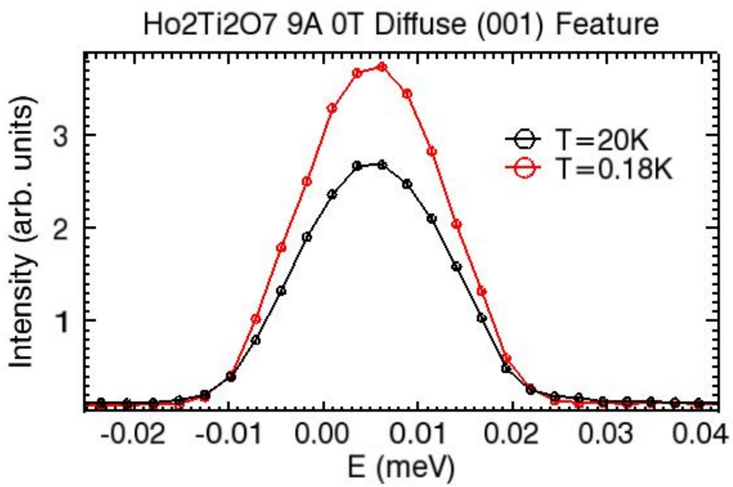
Ferro-ordering of α-chains



1D-correlations along β-chains

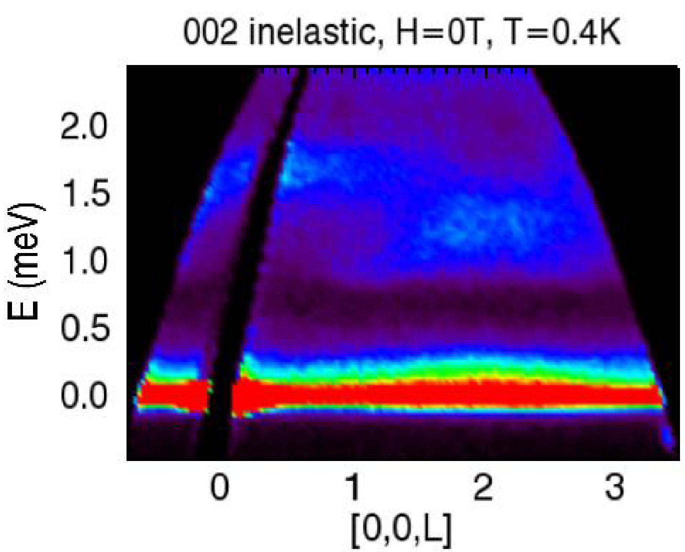


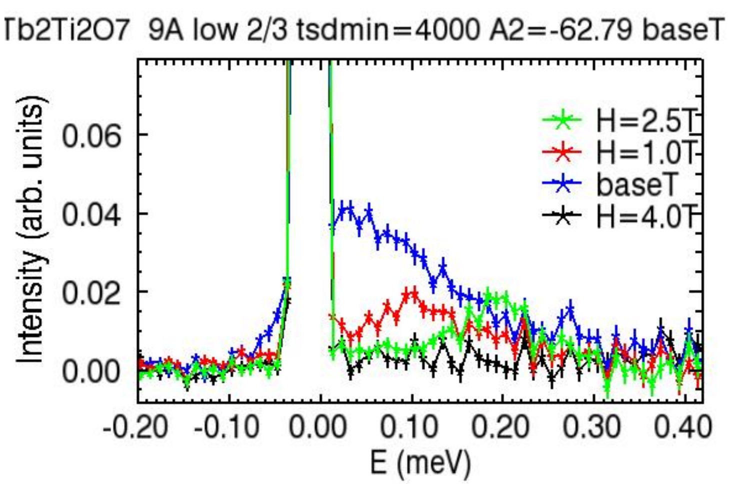




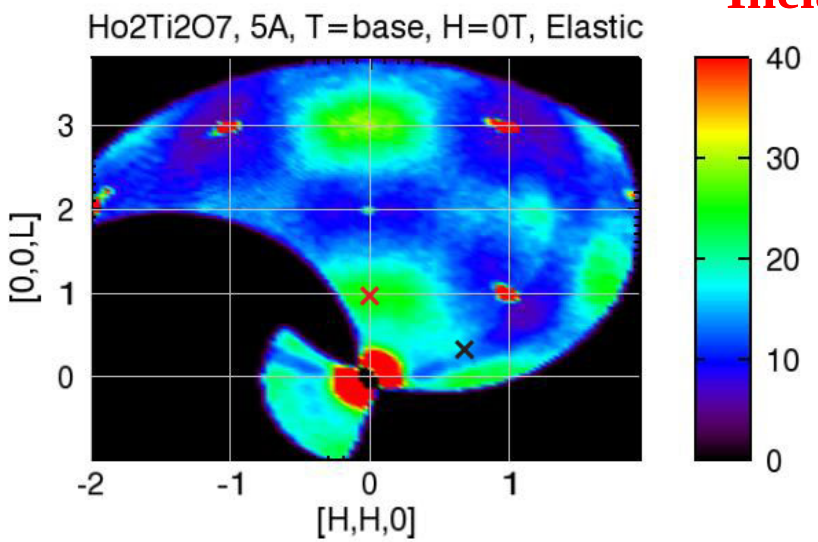
Spin Ice Ground State in $Ho_2Ti_2O_7$ is static

Spin Liquid Ground State in $Tb_2Ti_2O_7$ is dynamic

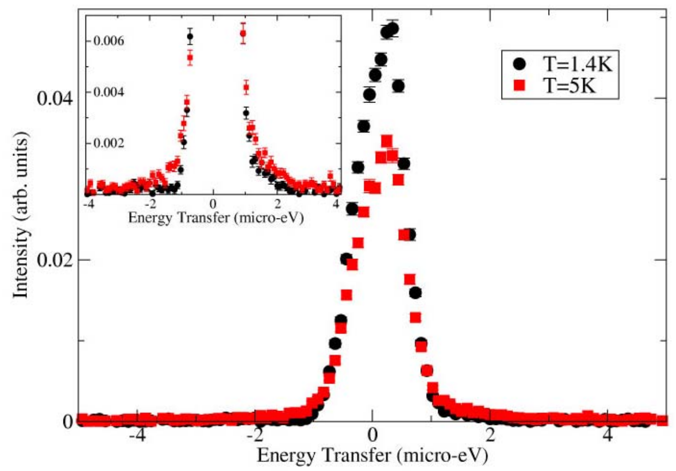


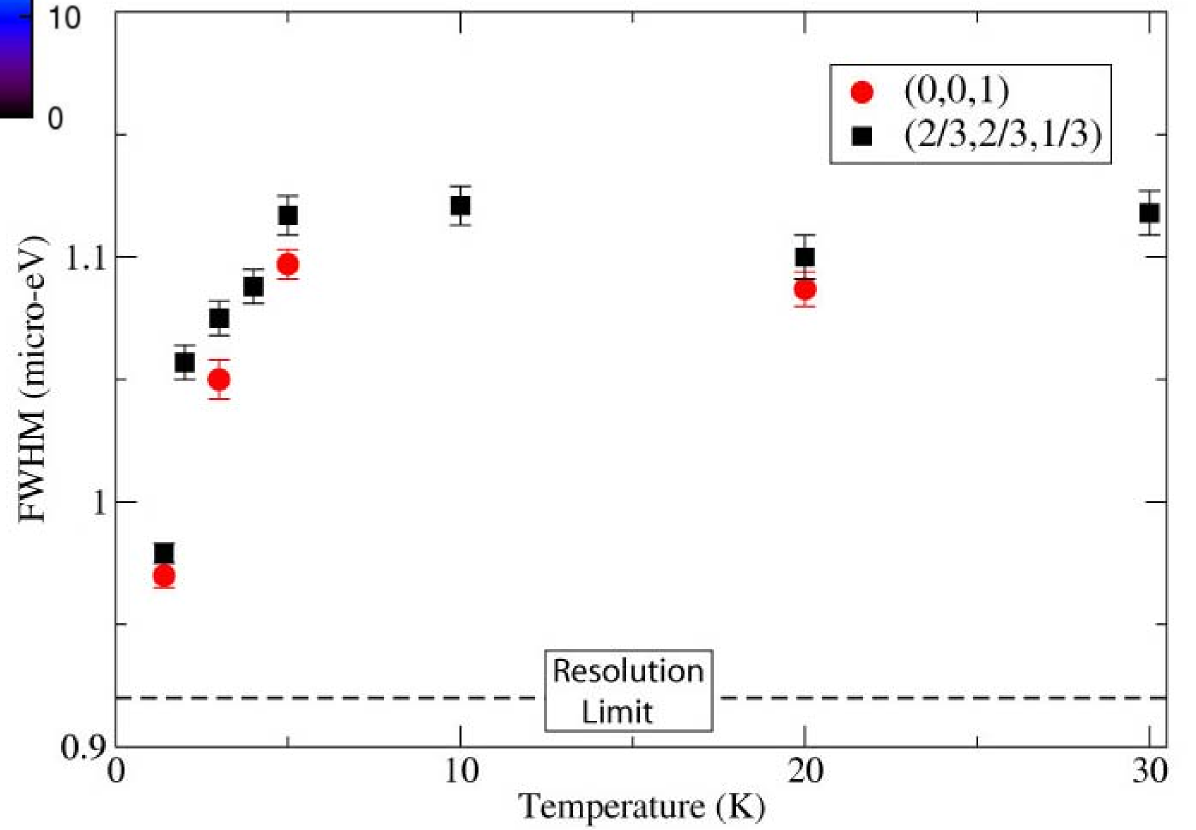


Very High Resolution
Inelastic Neutron Scattering (Backscattering)
Sees "Spin Ice" Freeze



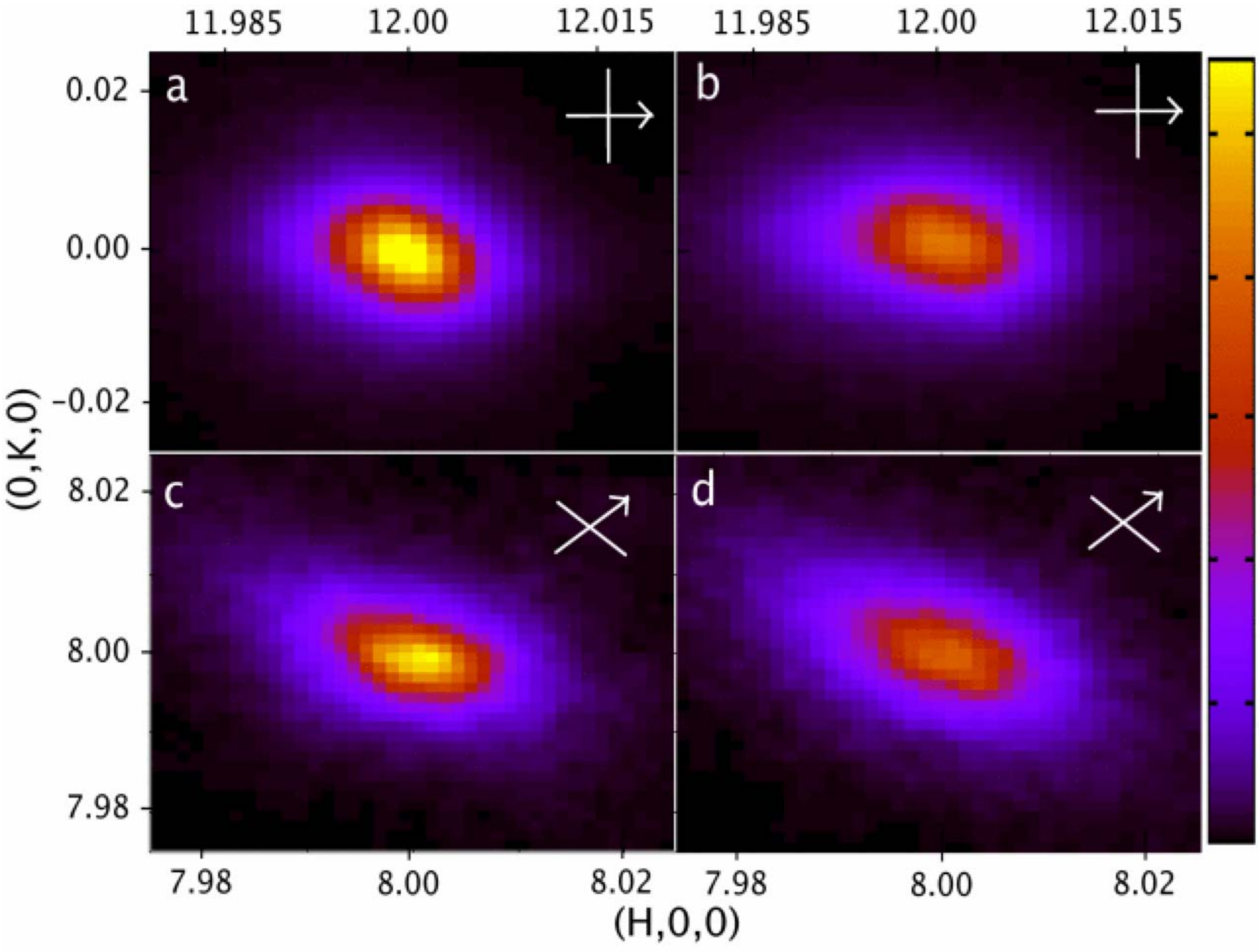
Energy Scale of Freezing is 100 neV!

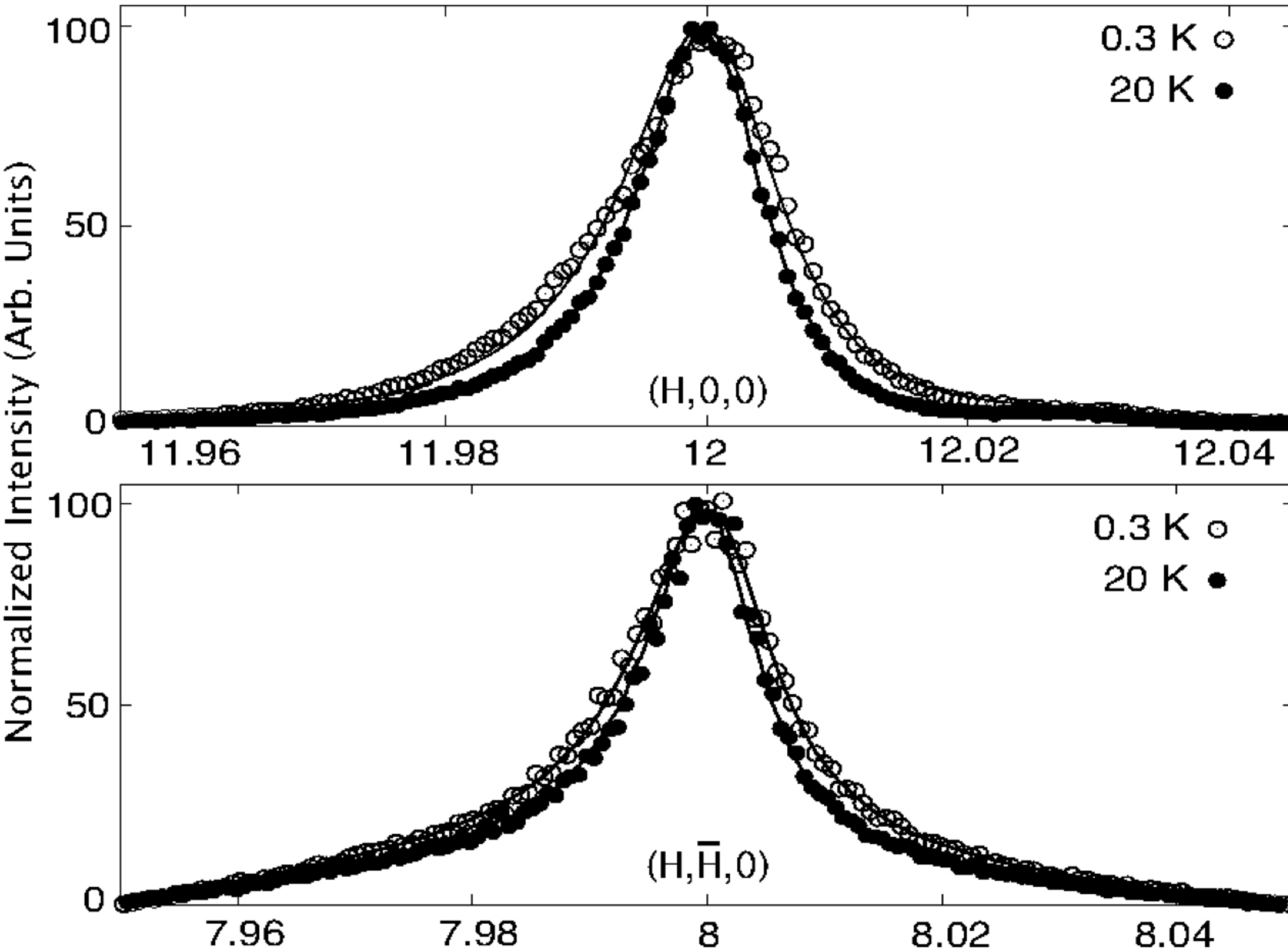




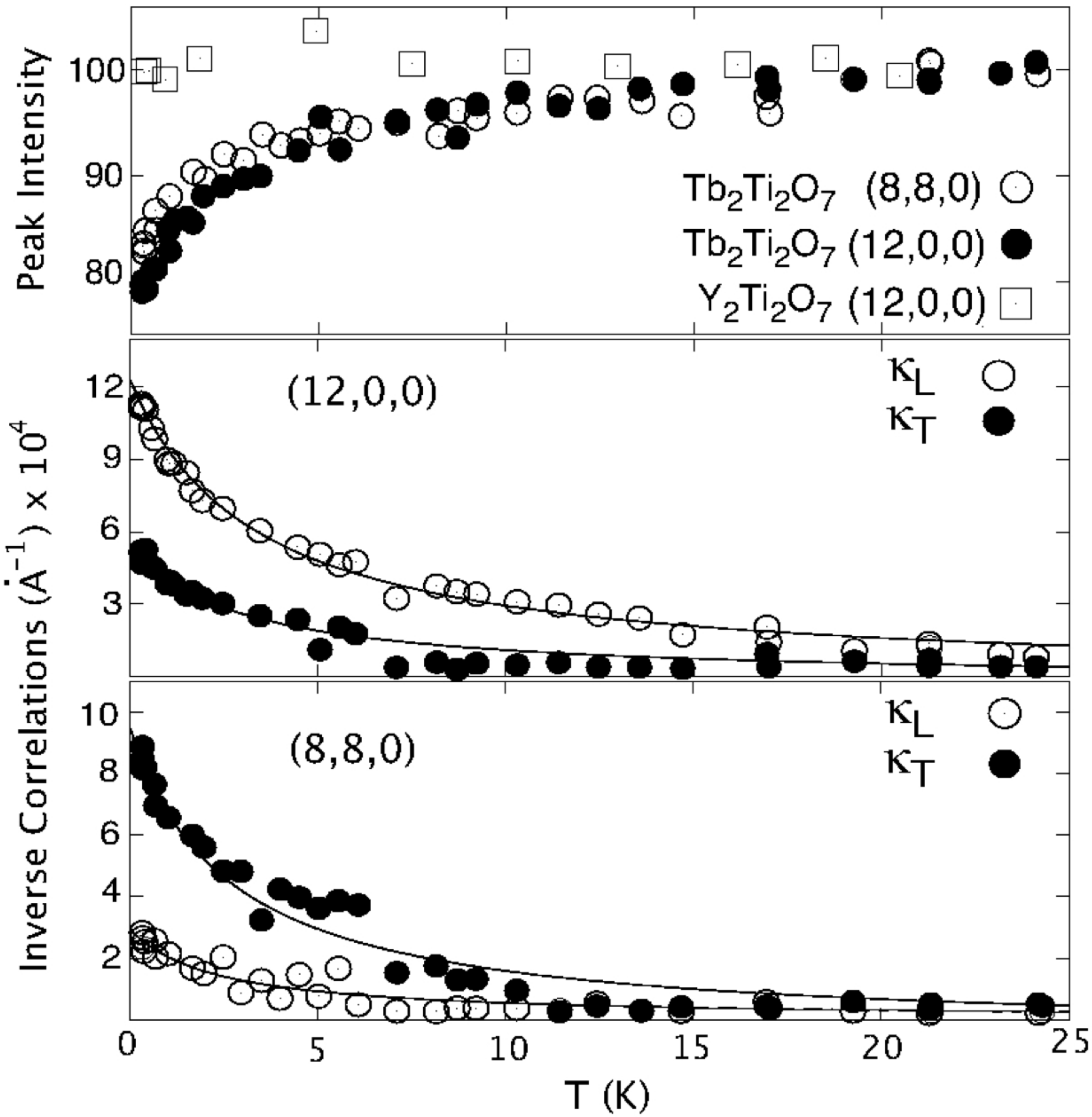
High Resolution *X-Ray* Scattering from $Tb_2Ti_2O_7$

Ruff et al., cond-mat/0707.1682

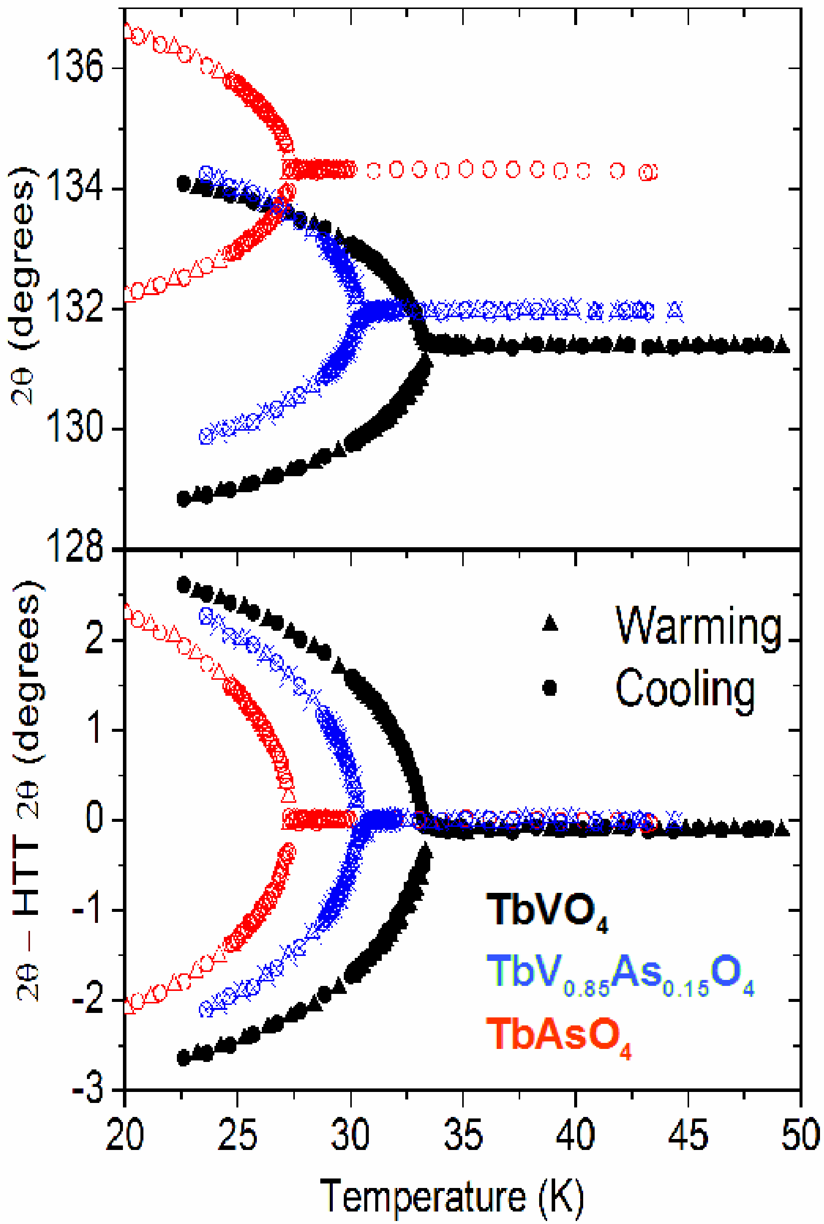


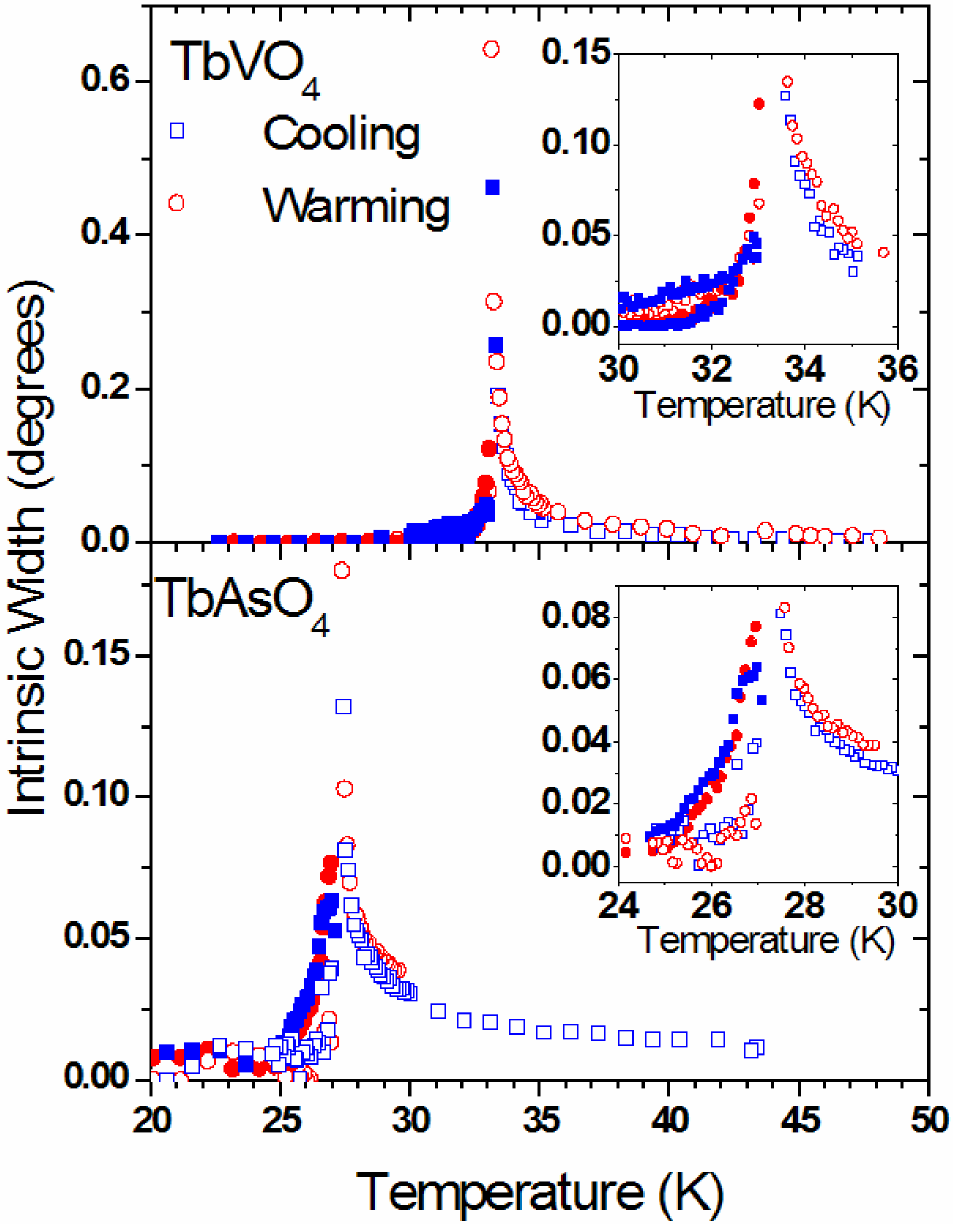


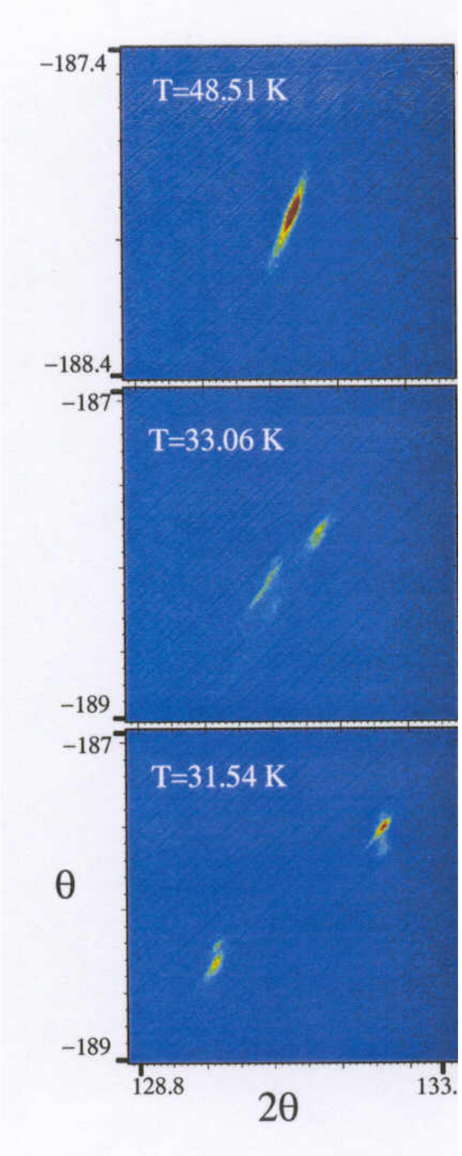
0.3 K
20 K
(H,0,0)
11.96
11.98
12
12.02
12.04
Normalized Intensity (Arb. Units)
100
50
0
0.3 K
20 K
(H,H̄,0)
7.96
7.98
8
8.02
8.04



Peak Intensity
100
90
80
Tb2Ti2O7 (8,8,0)
Tb2Ti2O7 (12,0,0)
Y2Ti2O7 (12,0,0)
(12,0,0)
κL
κT
12
9
6
3
Inverse Correlations (Å-1) x 10^4
(8,8,0)
κL
κT
10
8
6
4
2
0
5
10
15
20
25
T (K)

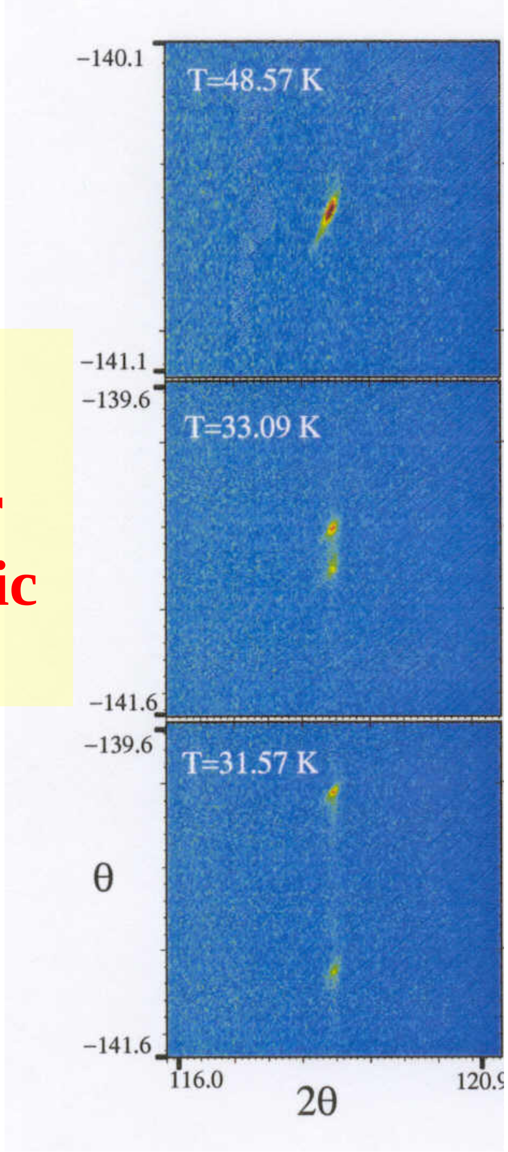

136
134
132
130
128
2θ (degrees)
2
1
0
-1
-2
-3
2θ – HTT 2θ (degrees)
Warming
Cooling
TbVO4
TbV0.85As0.15O4
TbAsO4
20
25
30
35
40
45
50
Temperature (K)


TbVO4
Cooling
Warming
0.6
0.4
0.2
0.0
0.15
0.10
0.05
0.00
30
32
34
36
Temperature (K)
TbAsO4
0.15
0.10
0.05
0.00
0.08
0.06
0.04
0.02
0.00
24
26
28
30
Temperature (K)
Intrinsic Width (degrees)
20
25
30
35
40
45
50
Temperature (K)



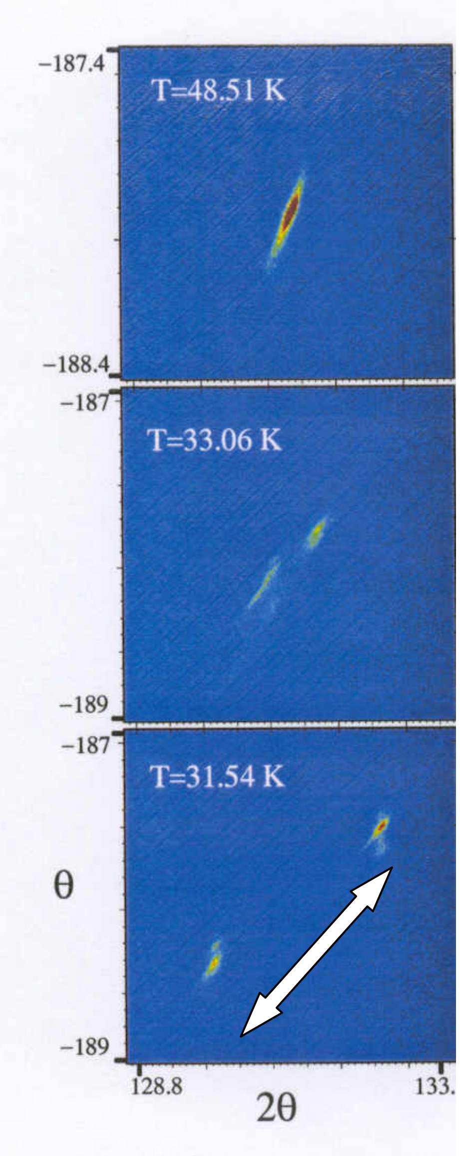
$TbVO_4$

Cooperative Jahn-Teller
Tetragonal - Orthorhombic
T_C ~ 33.3 K



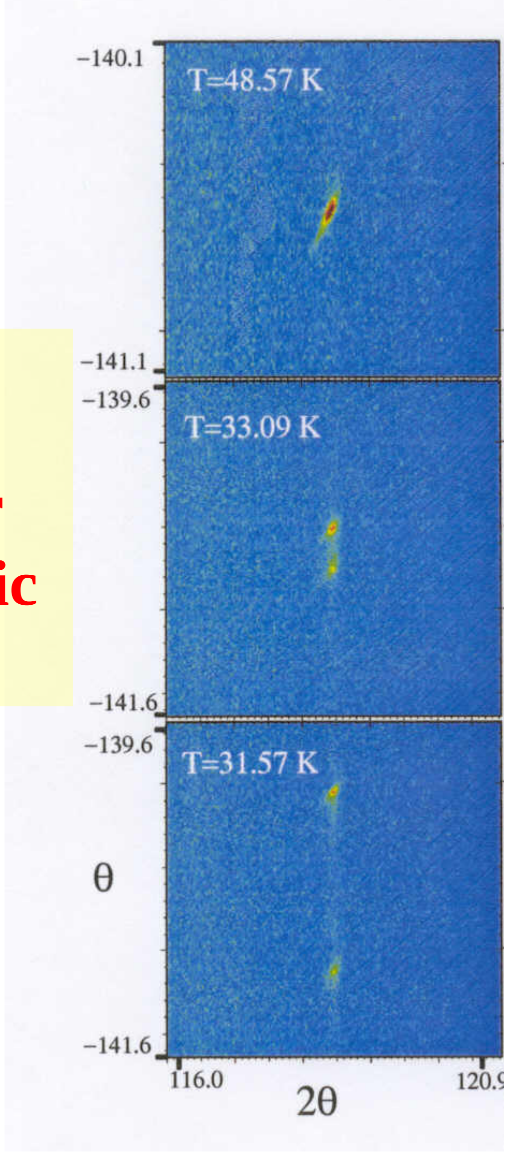
(12, 0, 0) splits in the *longitudinal direction*

(8, 8, 0) splits in the *transverse direction*



$TbVO_4$

Cooperative Jahn-Teller
Tetragonal - Orthorhombic
T_C ~ 33.3 K



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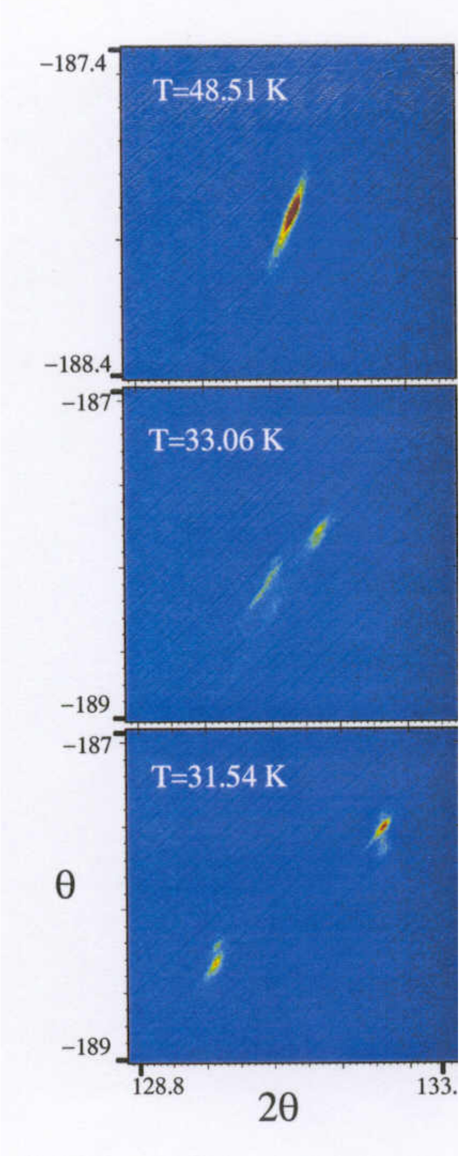
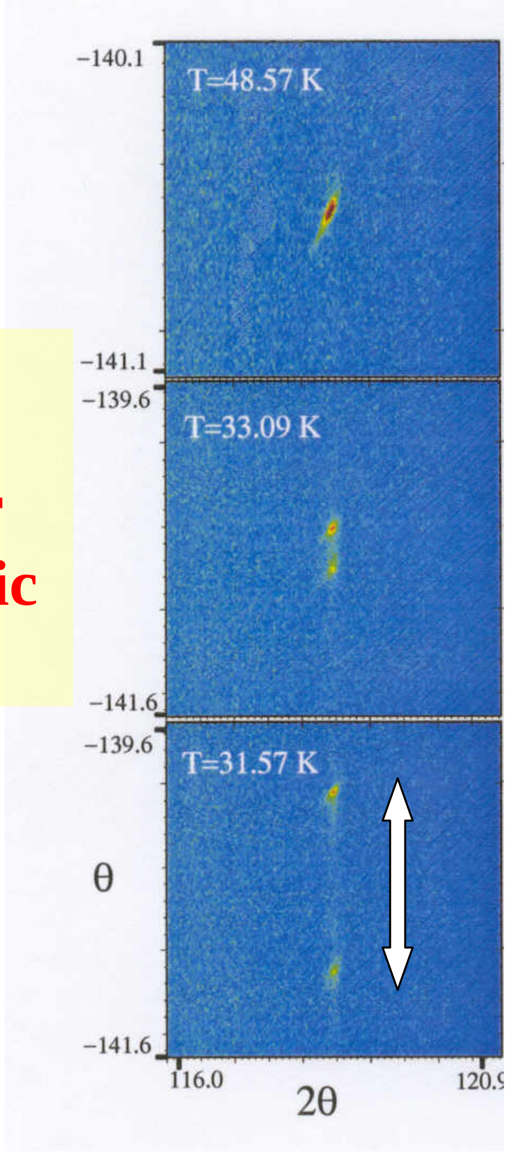


$TbVO_4$

Cooperative Jahn-Teller
Tetragonal - Orthorhombic
T_C ~ 33.3 K



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

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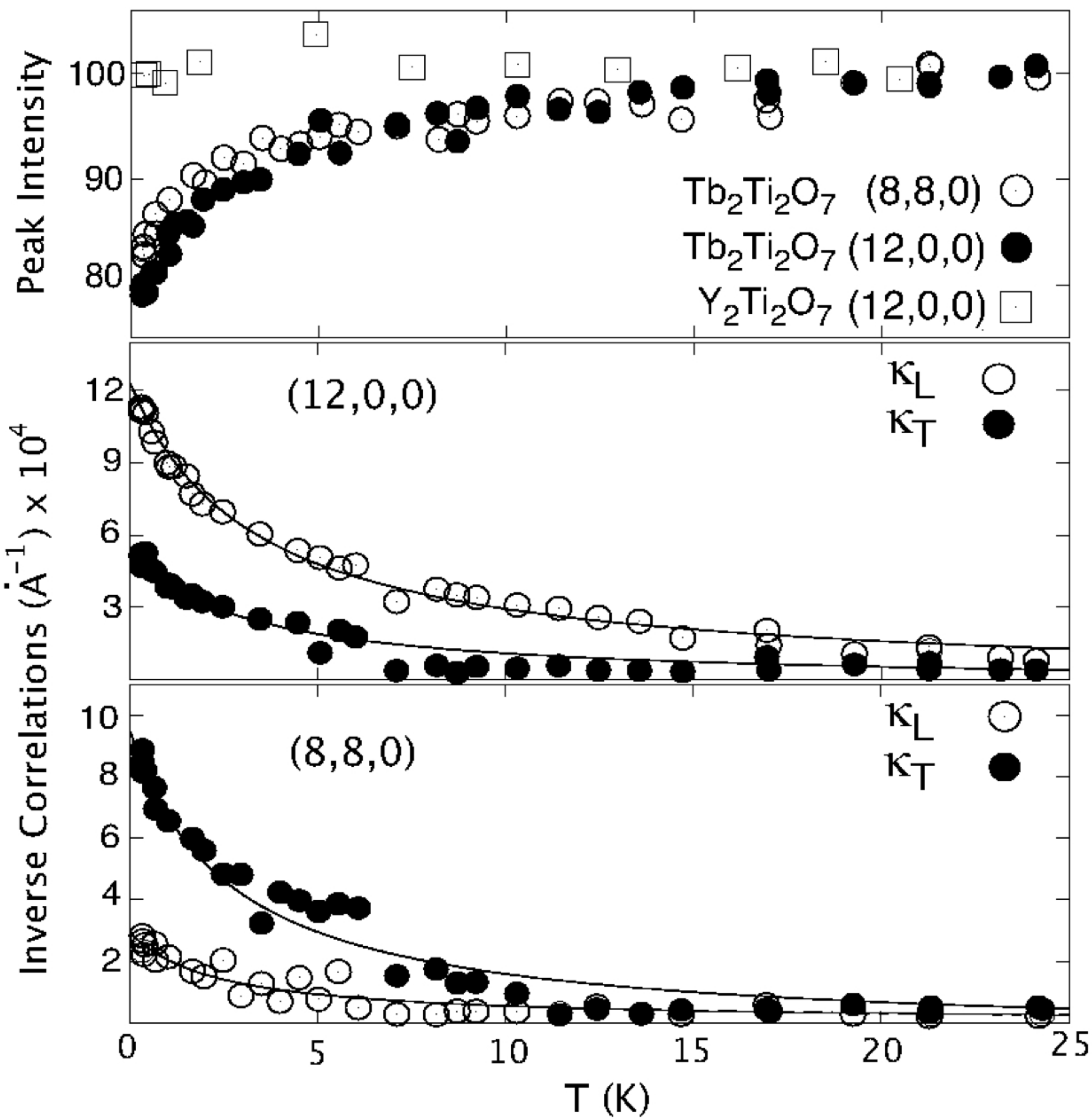
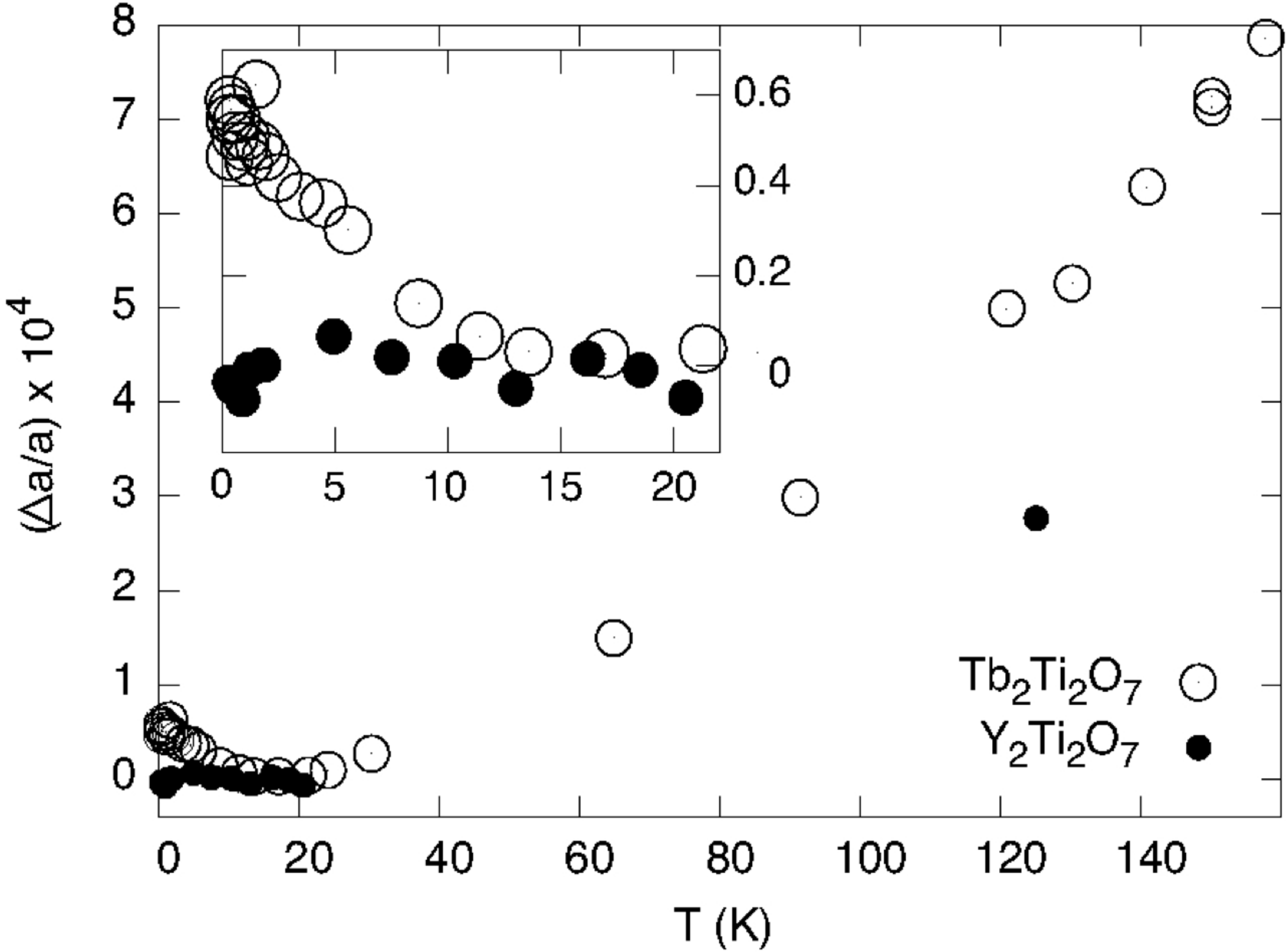

Peak Intensity
100
90
80
$Tb_2Ti_2O_7$ (8,8,0)
$Tb_2Ti_2O_7$ (12,0,0)
$Y_2Ti_2O_7$ (12,0,0)
(12,0,0)
κ_L
κ_T
12
9
6
3
Inverse Correlations ($\AA^{-1}$) x 10^4
(8,8,0)
κ_L
κ_T
10
8
6
4
2
0
5
10
15
20
25
T (K)


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

- **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 $Tb_2Ti_2O_7$ – grow continuously with decreasing T to T~ 0.3 K.**