Exotic Quantum and Classical Ground States in Geometrically Frustrated Magnets

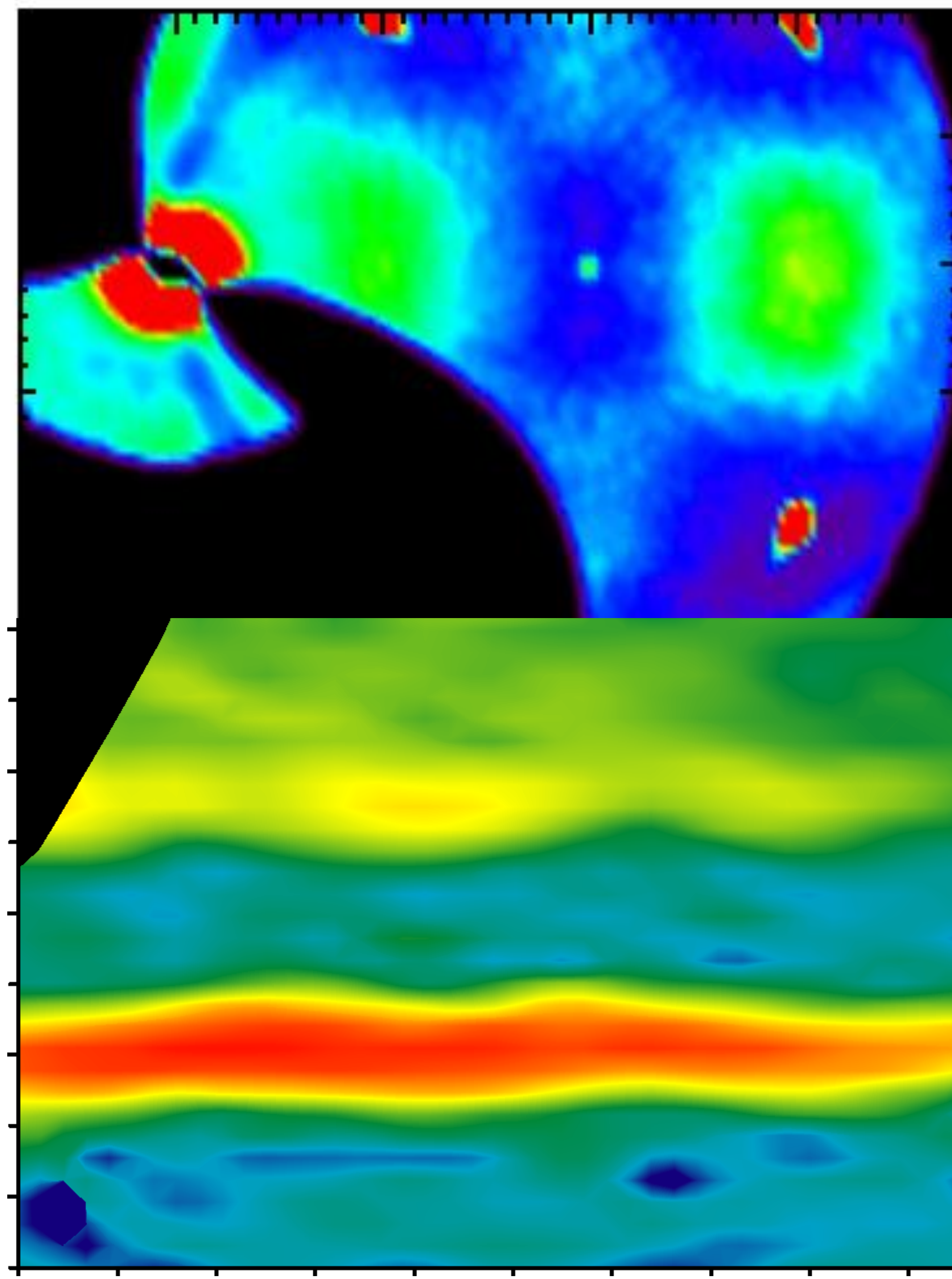
Bruce D Gaulin

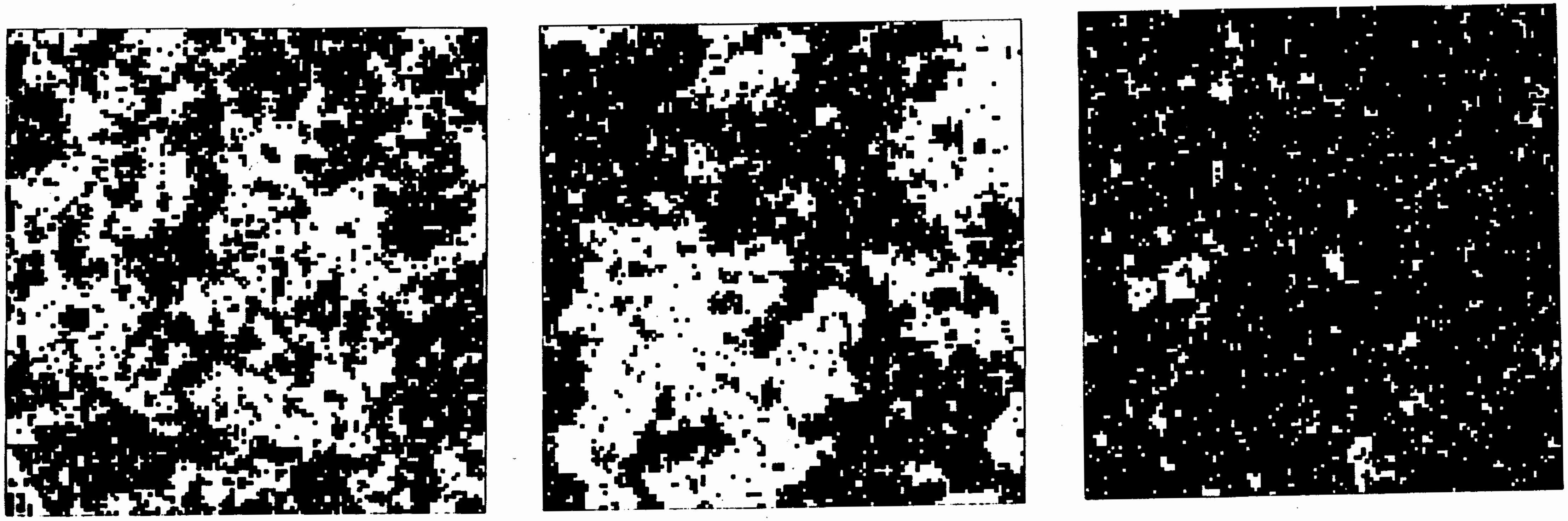
McMaster University

Brockhouse Institute for Materials Research

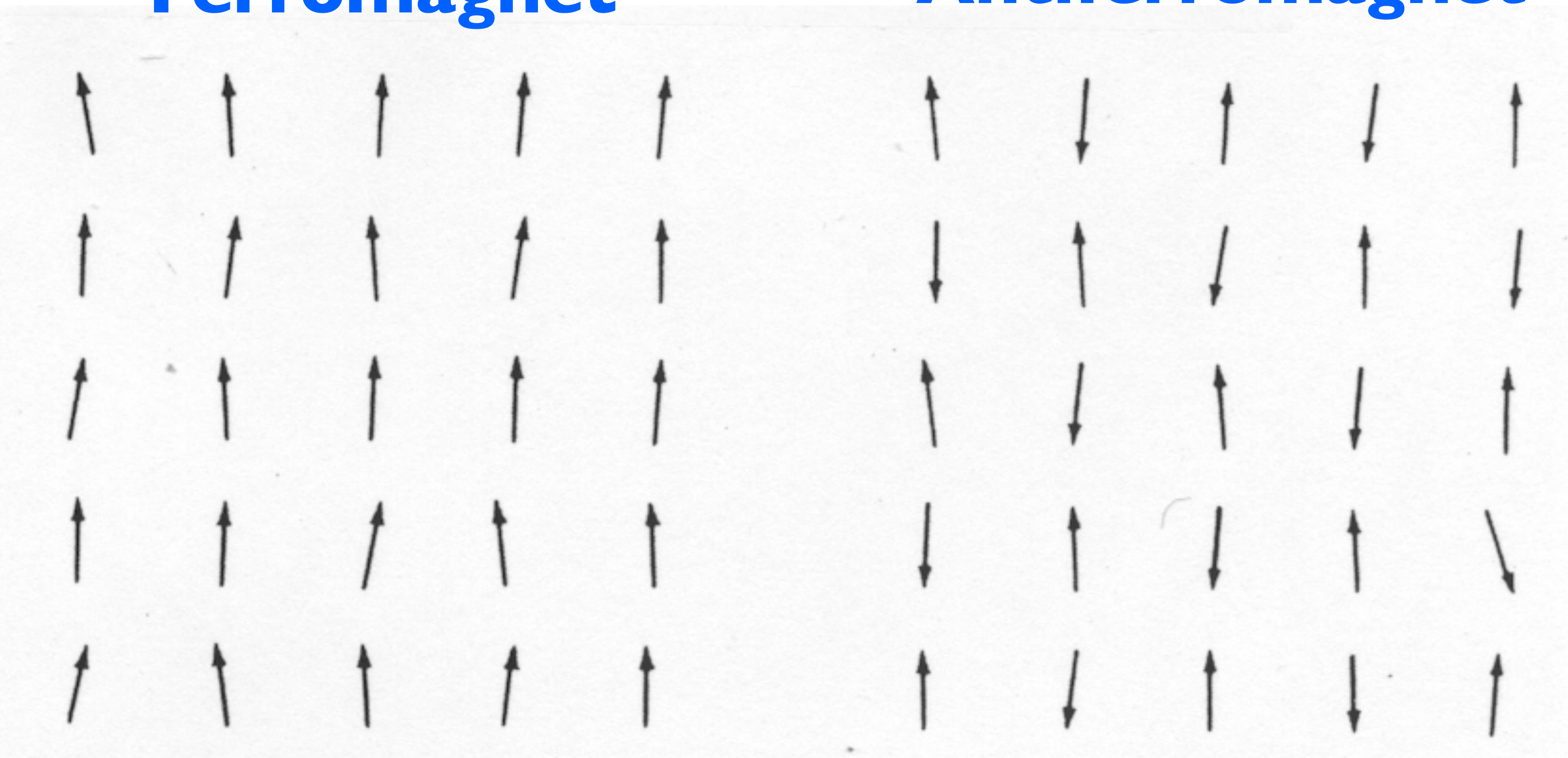
J.P. Clancy [1]
J.P.C. Ruff [1]
S. Haravifard [1]
K.A. Ross [1]
K. Fritsch [1]
C.P. Adams [1,2]
S.R. Dunsiger [1,3]
M. Ramazanoglu [1]
K.C. Rule [1,4]
J.S. Gardner [5]
R. Higashinaka [6]
H. Takagi [7]
H.A. Dabkowska [1]
A. Dabkowski [1]
Y. Qiu [5]
J.R.D. Copley [5]

[1] McMaster U
[2] St. Francis Xavier U
[3] TU Munich
[4] HZ Berlin
[5] NIST
[6] Tokyo Metro U
[7] U of Tokyo



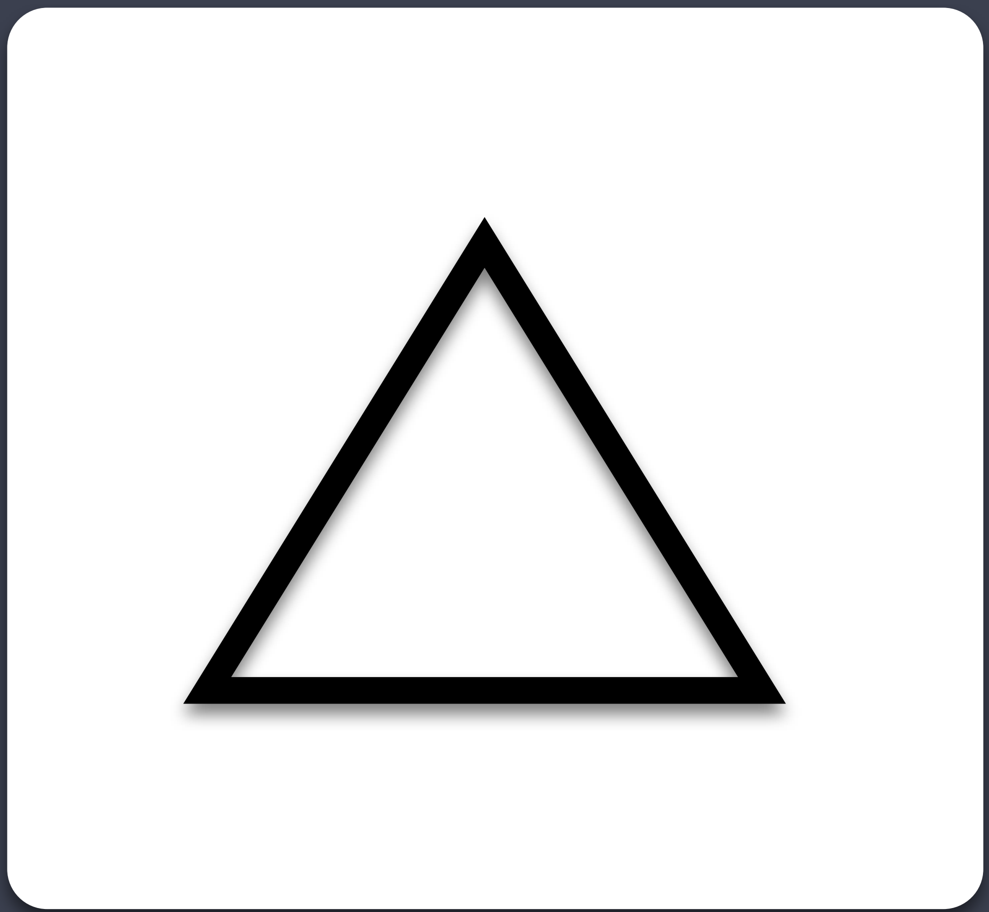


1.1 Tc | Tc | 0.9 Tc



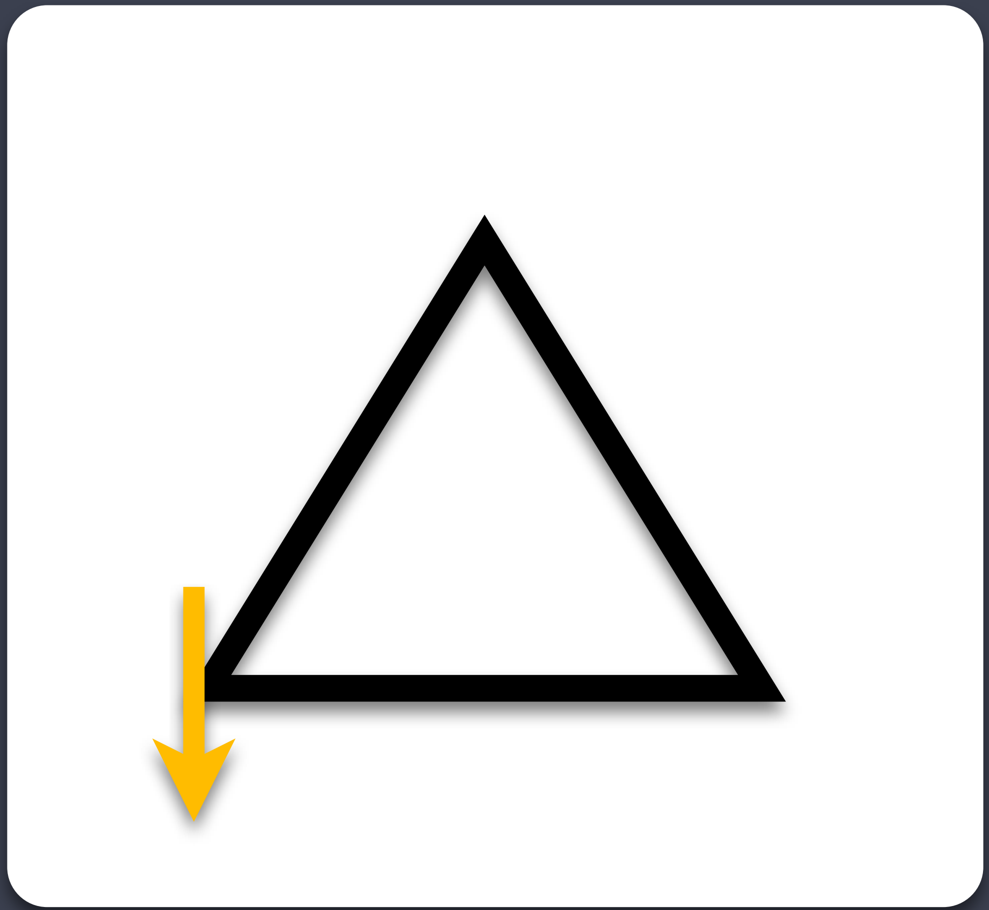
$$H = 2J\ \Sigma_{ij}\ \mathbf{S_i} \cdot \mathbf{S_j}$$

Geometric Frustration in 2D



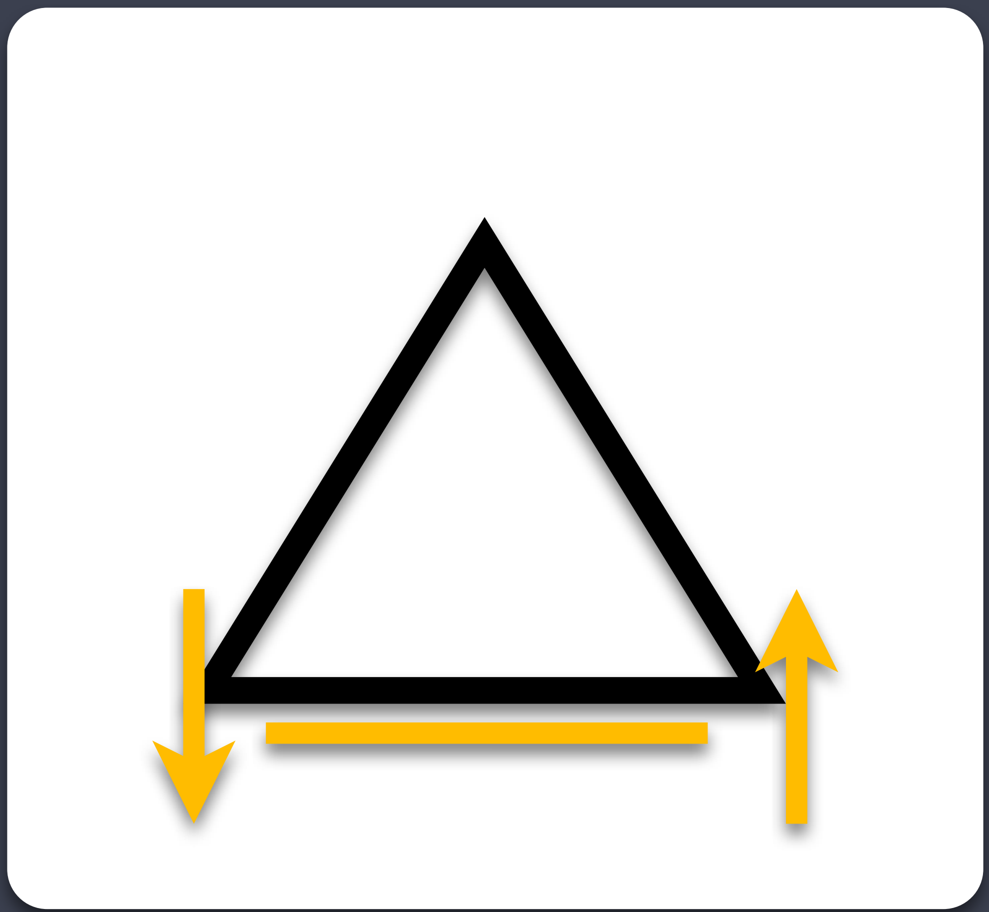
prefer ↑↓ alignment,
but choice of 3rd spin direction is unclear

Geometric Frustration in 2D



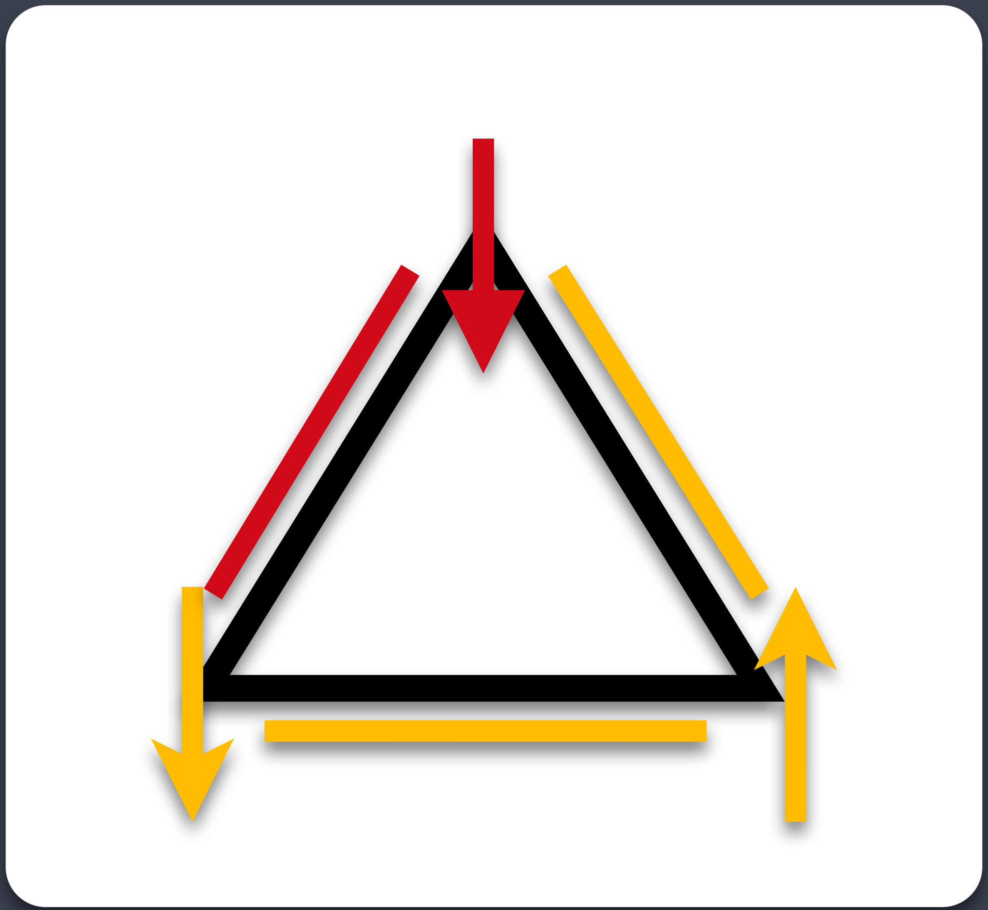
prefer ↑↓ alignment,
but choice of 3rd spin direction is unclear

Geometric Frustration in 2D



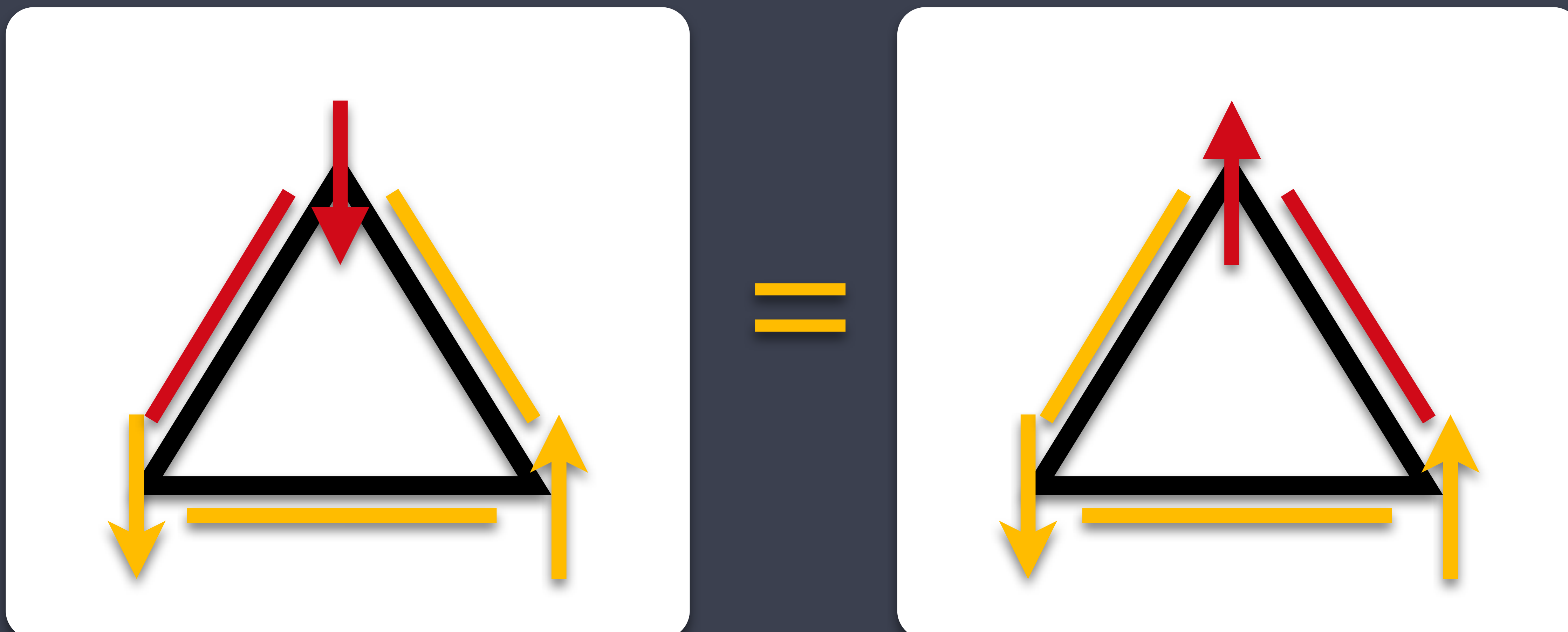
prefer ↑↓ alignment,
but choice of 3rd spin direction is unclear

Geometric Frustration in 2D



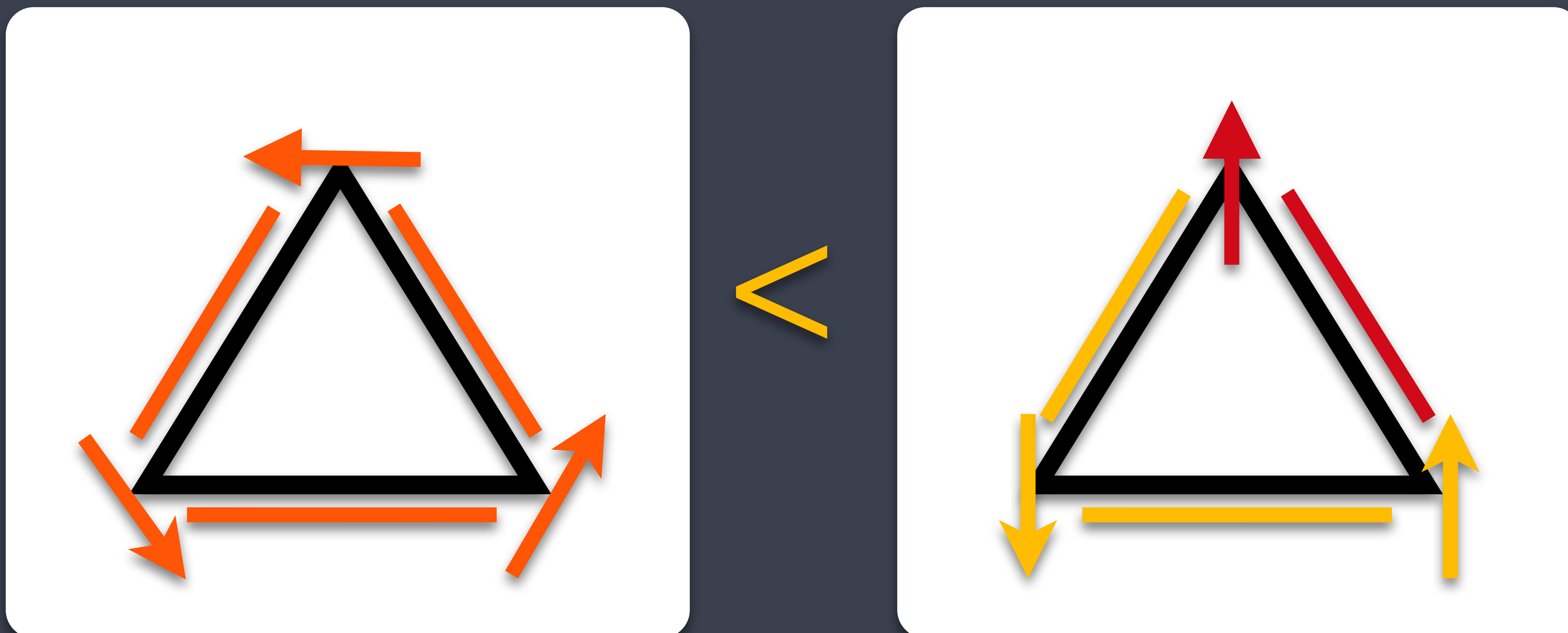
prefer ↑↓ alignment,
but choice of 3rd spin direction is unclear

Geometric Frustration in 2D



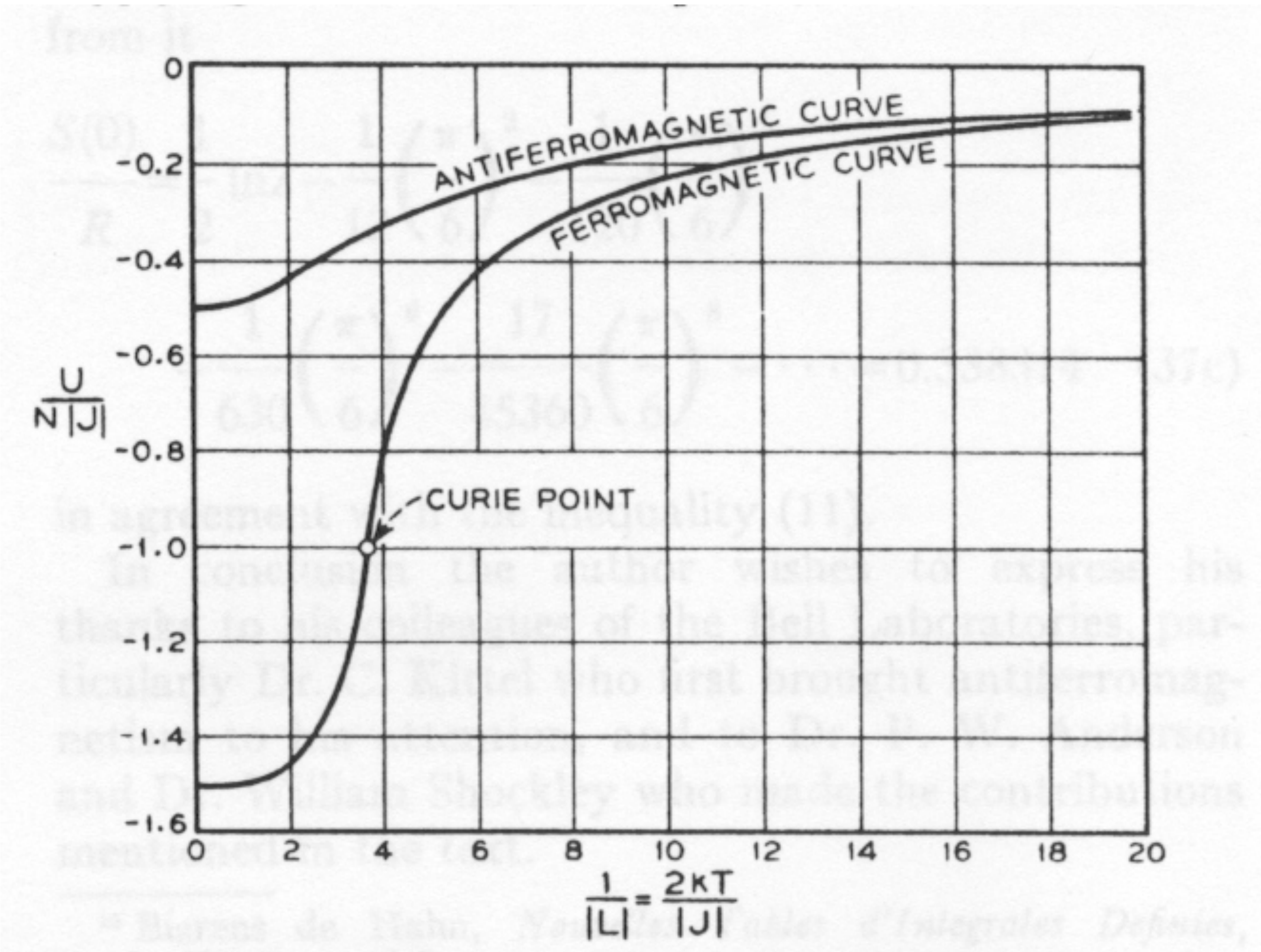
prefer ↑↓ alignment,
but choice of 3rd spin direction is unclear

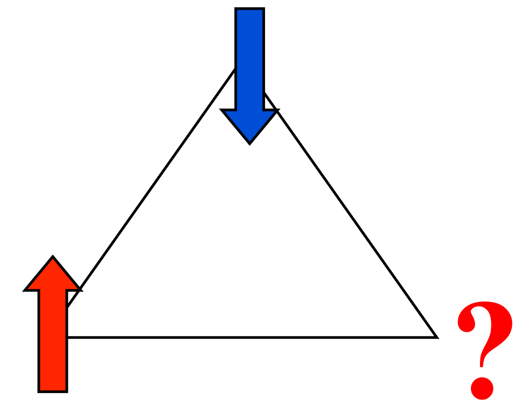
Geometric Frustration in 2D

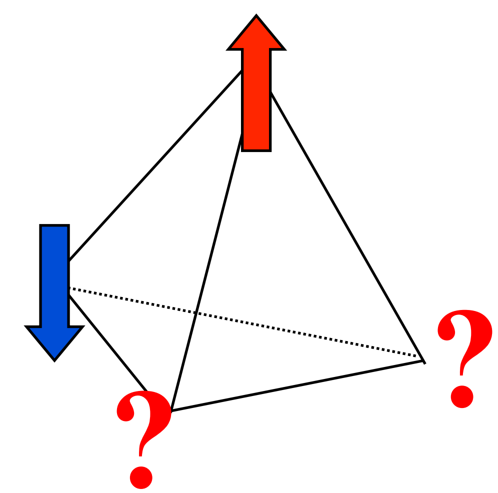


Can SOMETIMES lower energy by making a compromise

Geometrical Frustration:
Antiferromagnetism + Triangles and Tetrahedra

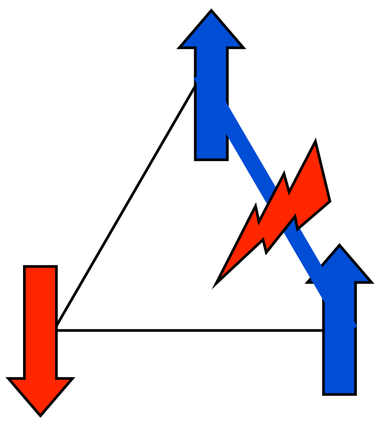


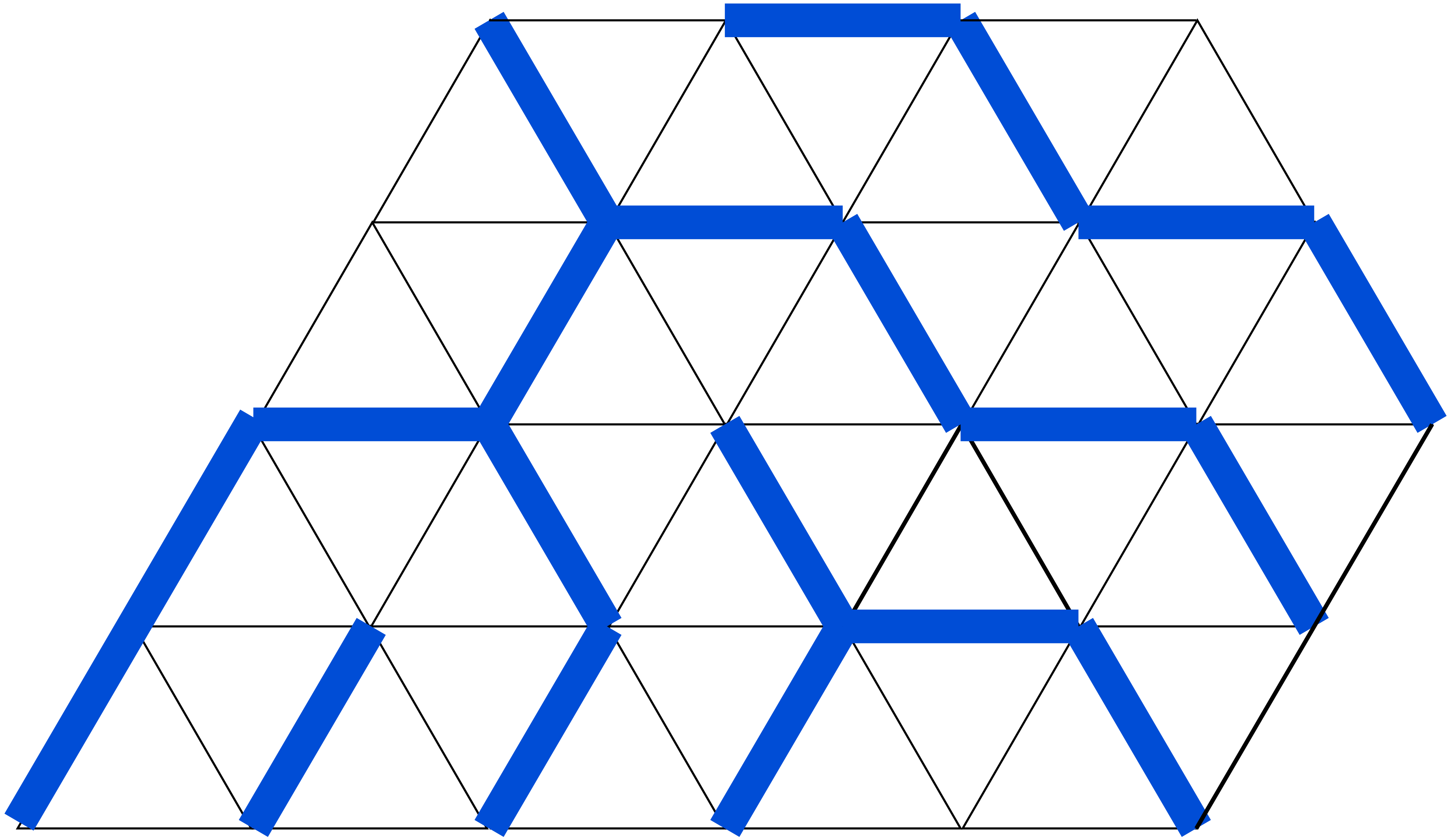




2D Triangular Lattice: G.H. Wannier Phys. Rev. 79, 357, 1950.

Ground State of the Ising AF on a Triangular Lattice

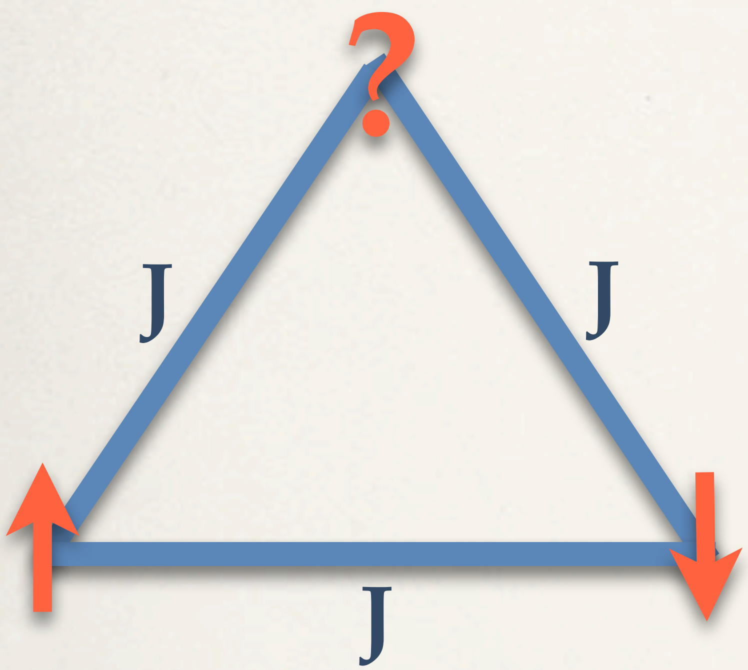




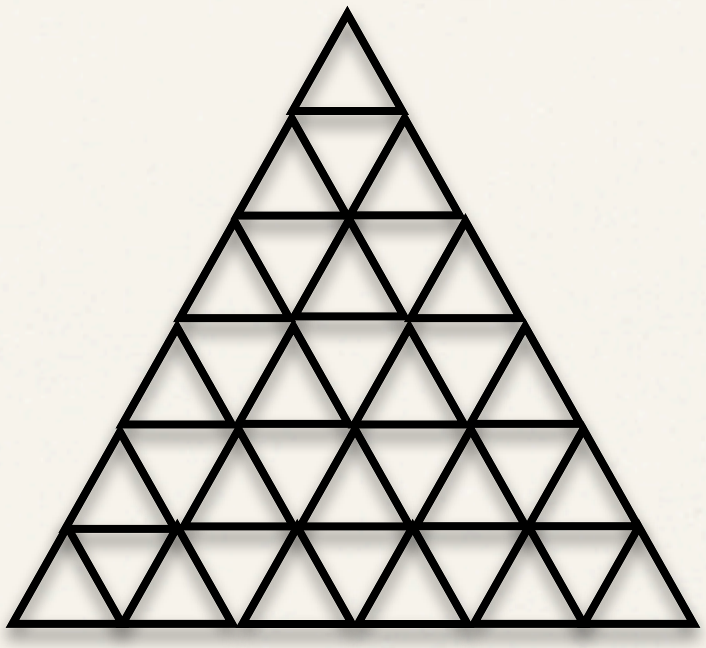
Entropy at T=0 is finite ~ 0.34 R

No LRO at any temperature

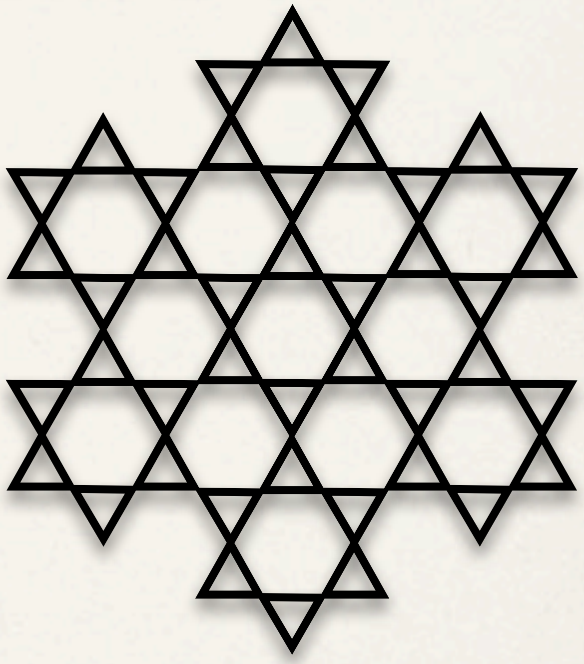
Geometrical Frustration



Frustrated Lattices in 2D



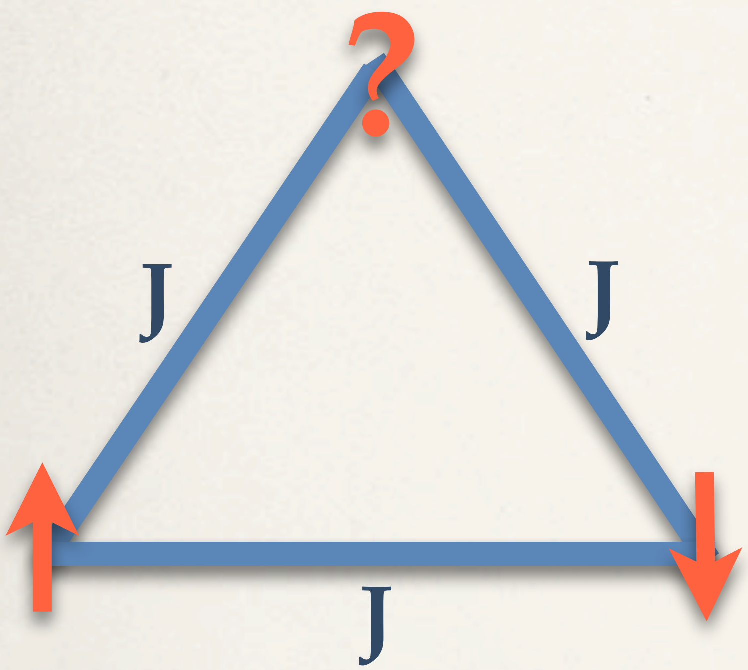
Triangular



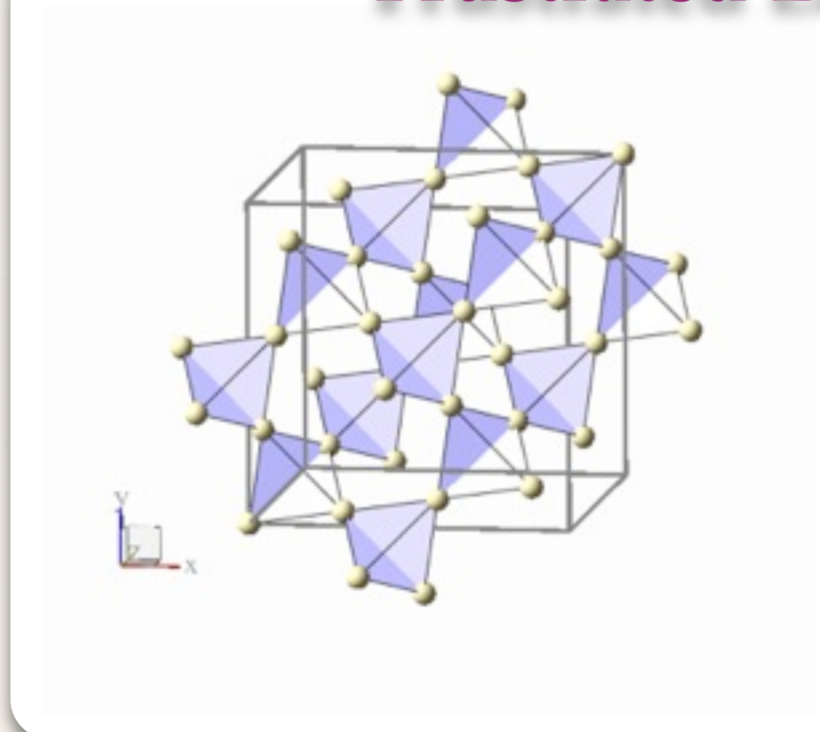
Kagome

- **Non-frustrated** quantum magnets **order** in 2D and 3D

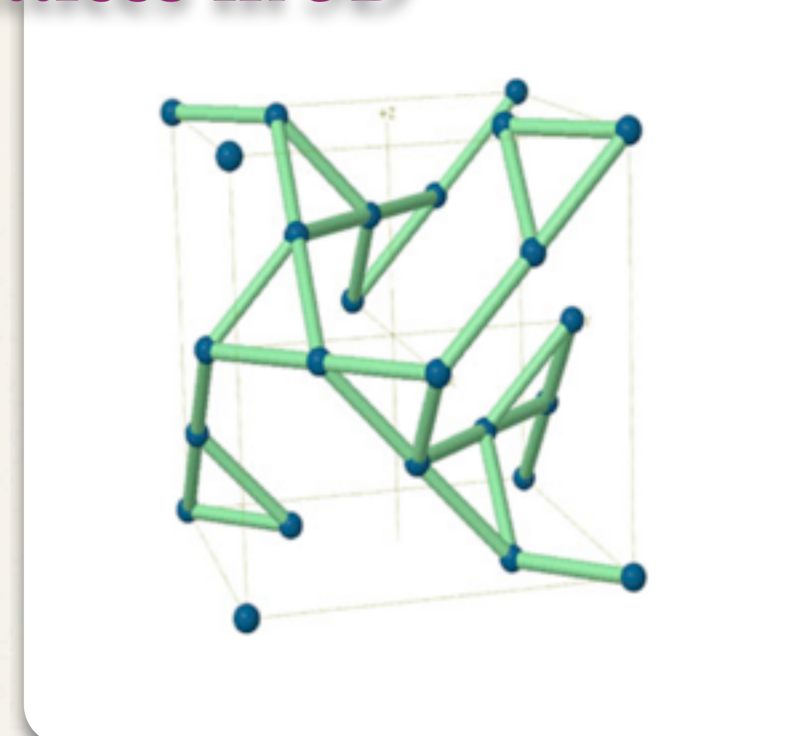
Geometrical Frustration



Frustrated Lattices in 3D



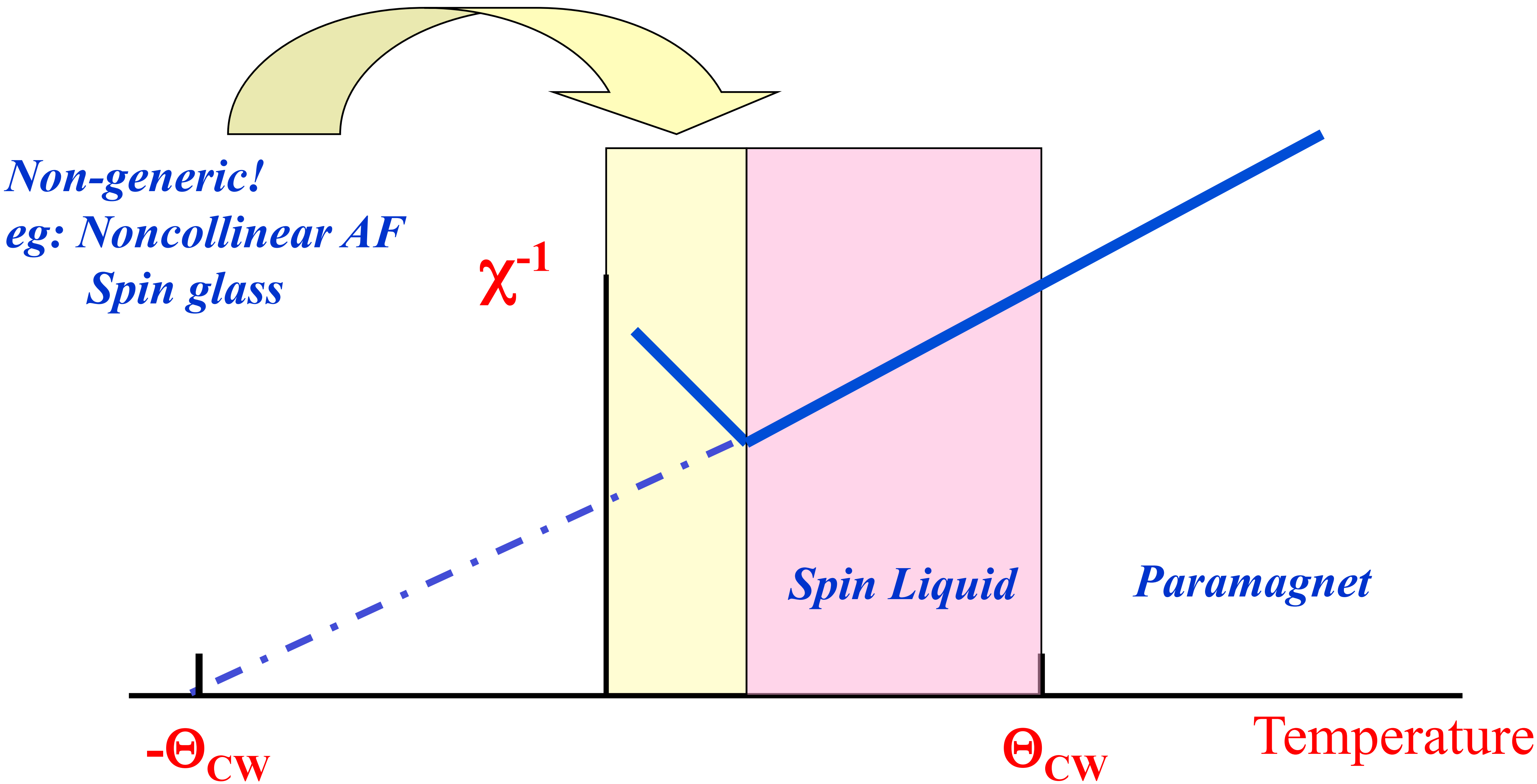
Pyrochlore



Hyper-kagome

- Non-frustrated quantum magnets order in 2D and 3D

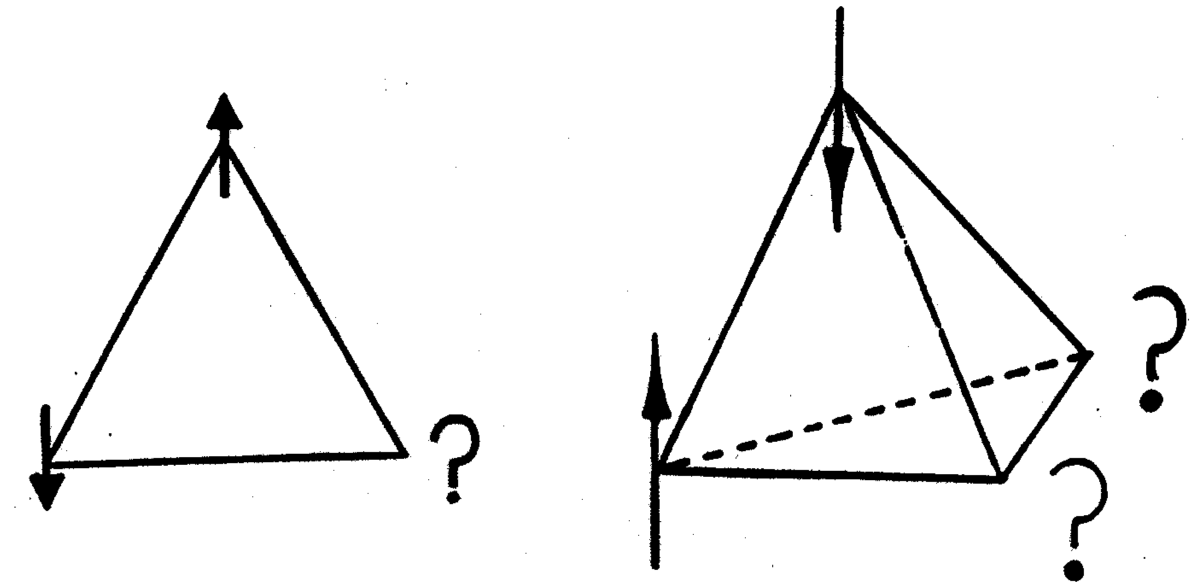
Geometrical Frustration:



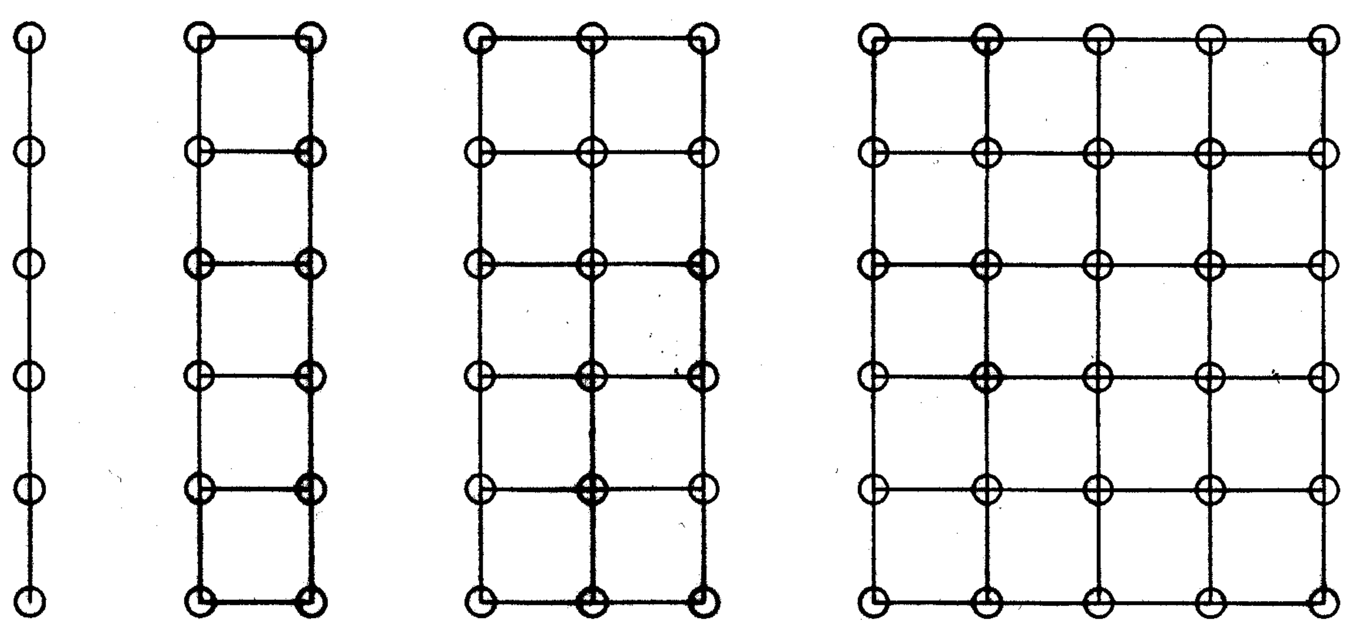
Mean Field Theory predicts a phase transition near T=| Θ_{CW} | ,
but materials remains disordered to much lower temperatures – Spin Liquid

Routes to Exotica

Geometrical Frustration



Quantum Fluctuations:
$S=1/2$ $H_{Transverse}$



Low Dimensional Structures

^{235}U + n

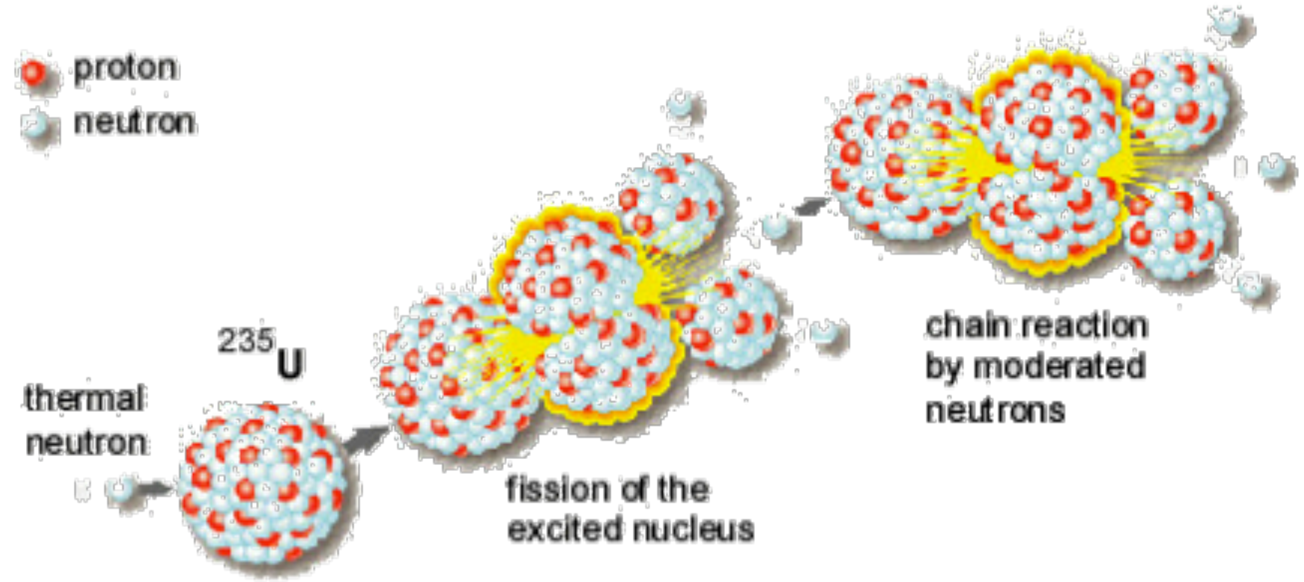
→

daughter nuclei +
2-3 n + gammas

neutrons:

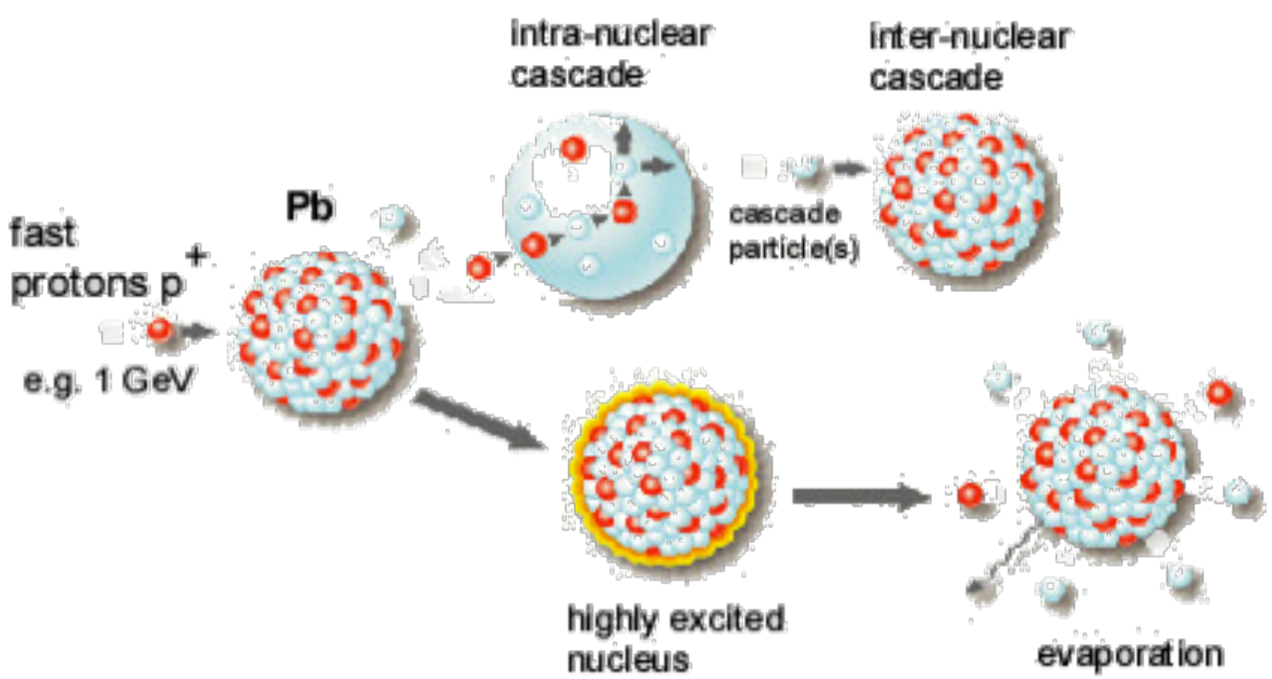
no charge
s=1/2
massive: mc^2~1 GeV

How do we produce neutrons



Fission

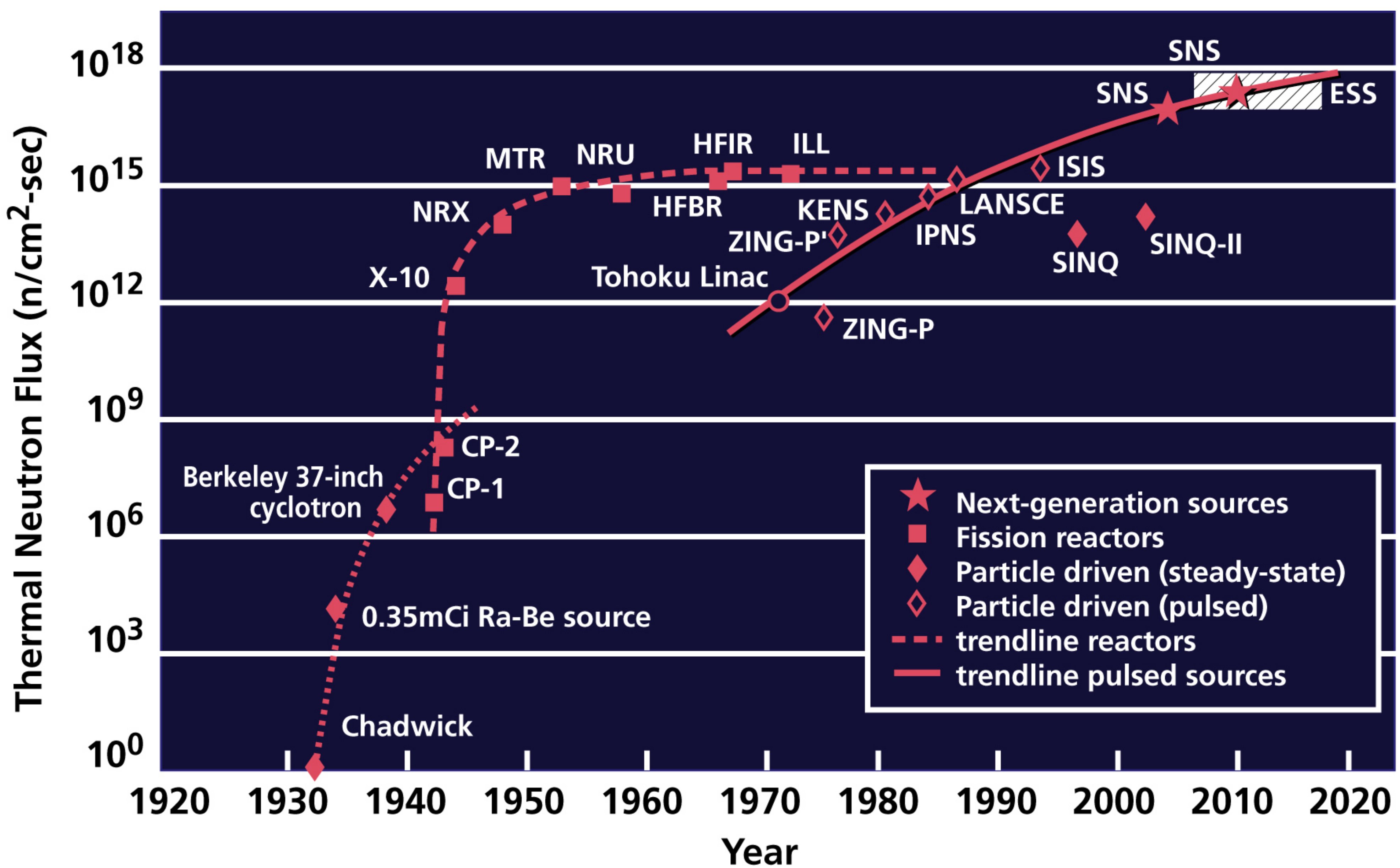
- chain reaction
- continuous flow
- 1 neutron/fission



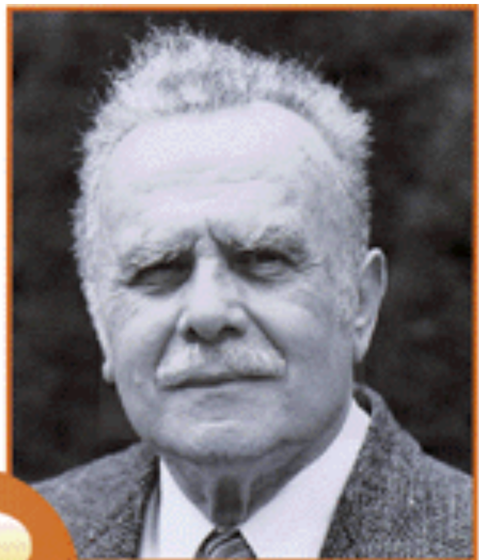
Spallation

- no chain reaction
- pulsed operation
- 30 neutrons/proton

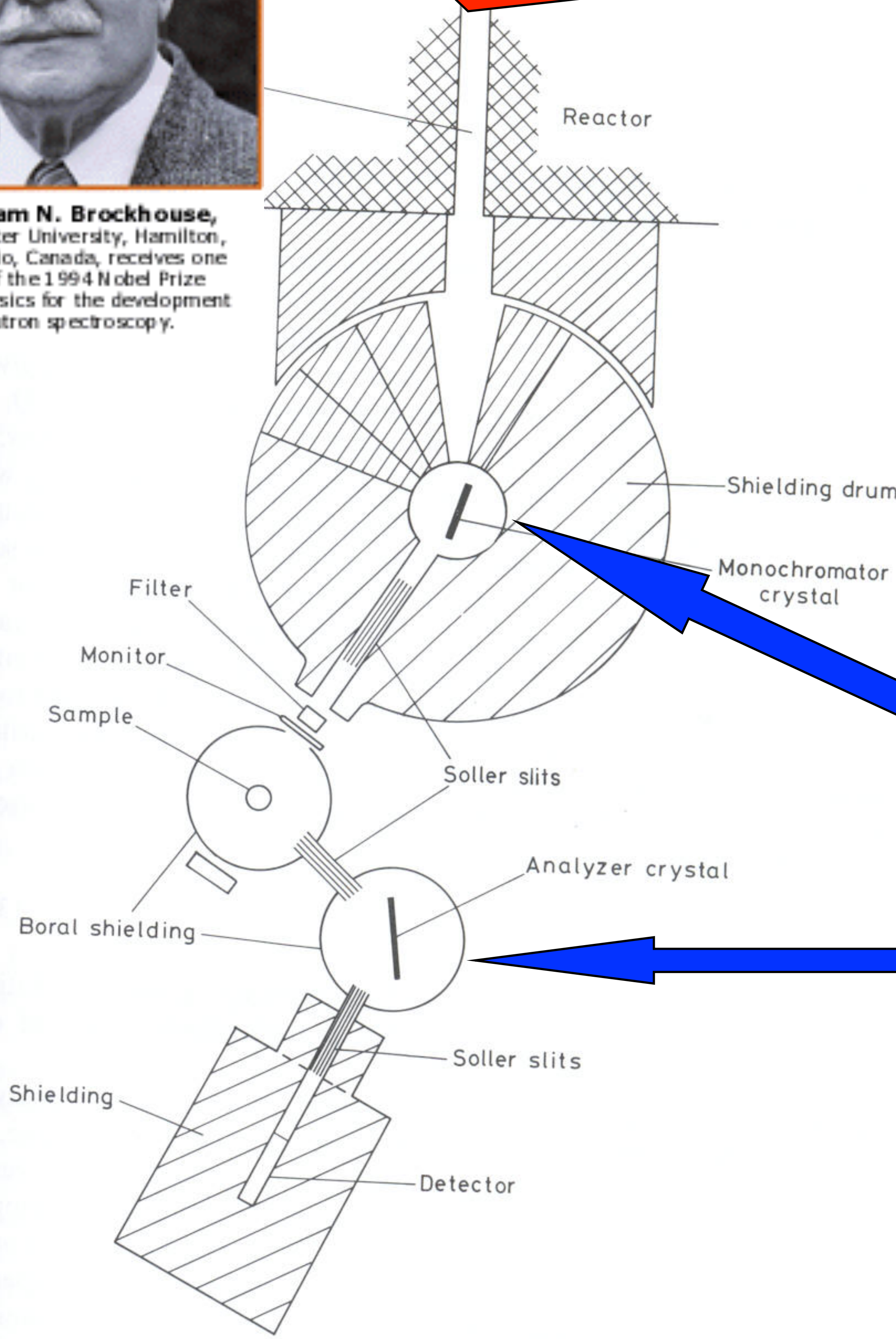
Development of Neutron Science Facilities

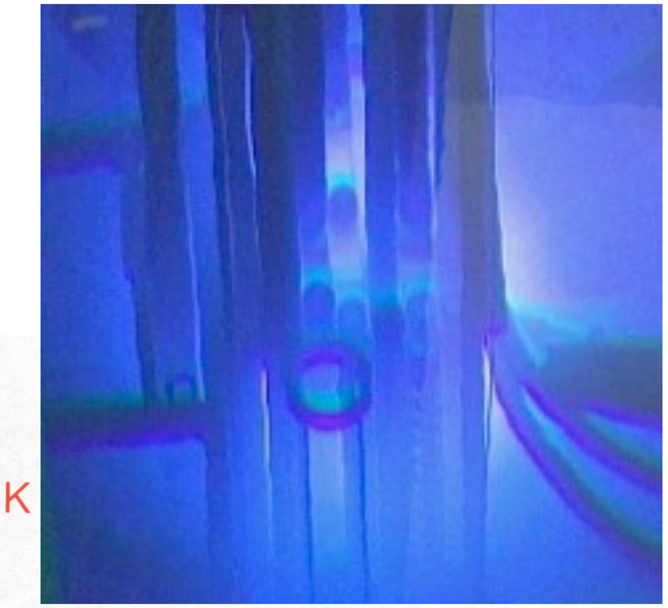


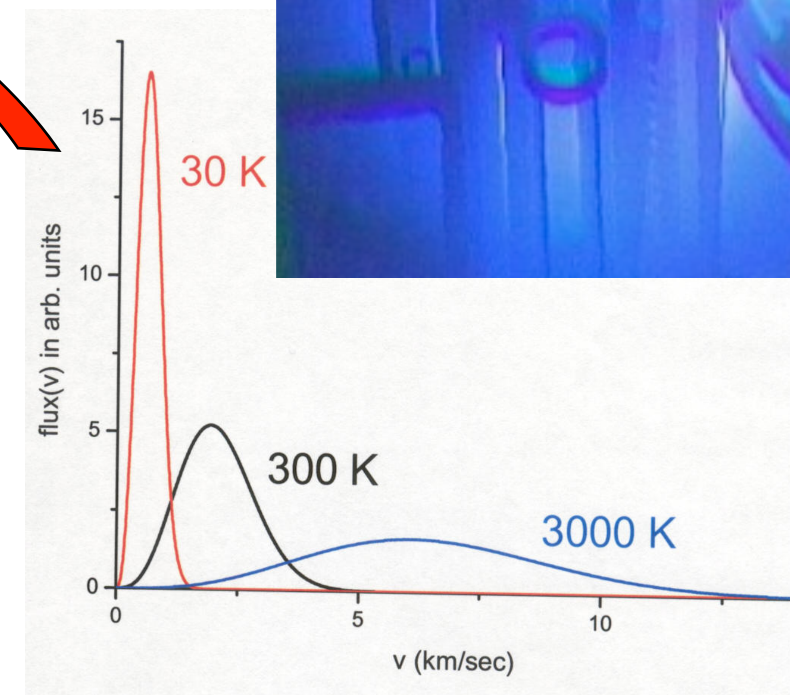
(Updated from *Neutron Scattering*, K. Skold and D. L. Price: eds., Academic Press, 1986)

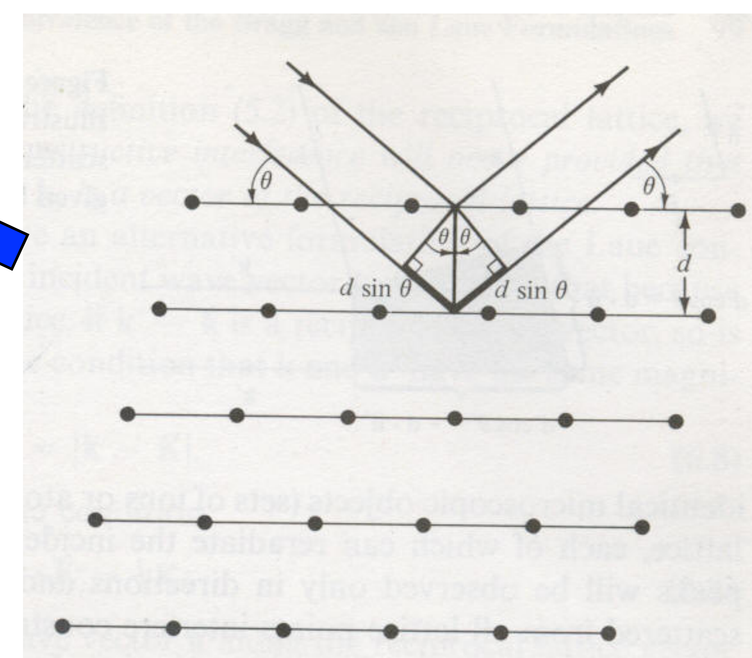


Betram N. Brockhouse,
McMater University, Hamilton, Ontario, Canada, receives one half of the 1994 Nobel Prize in Physics for the development of neutron spectroscopy.

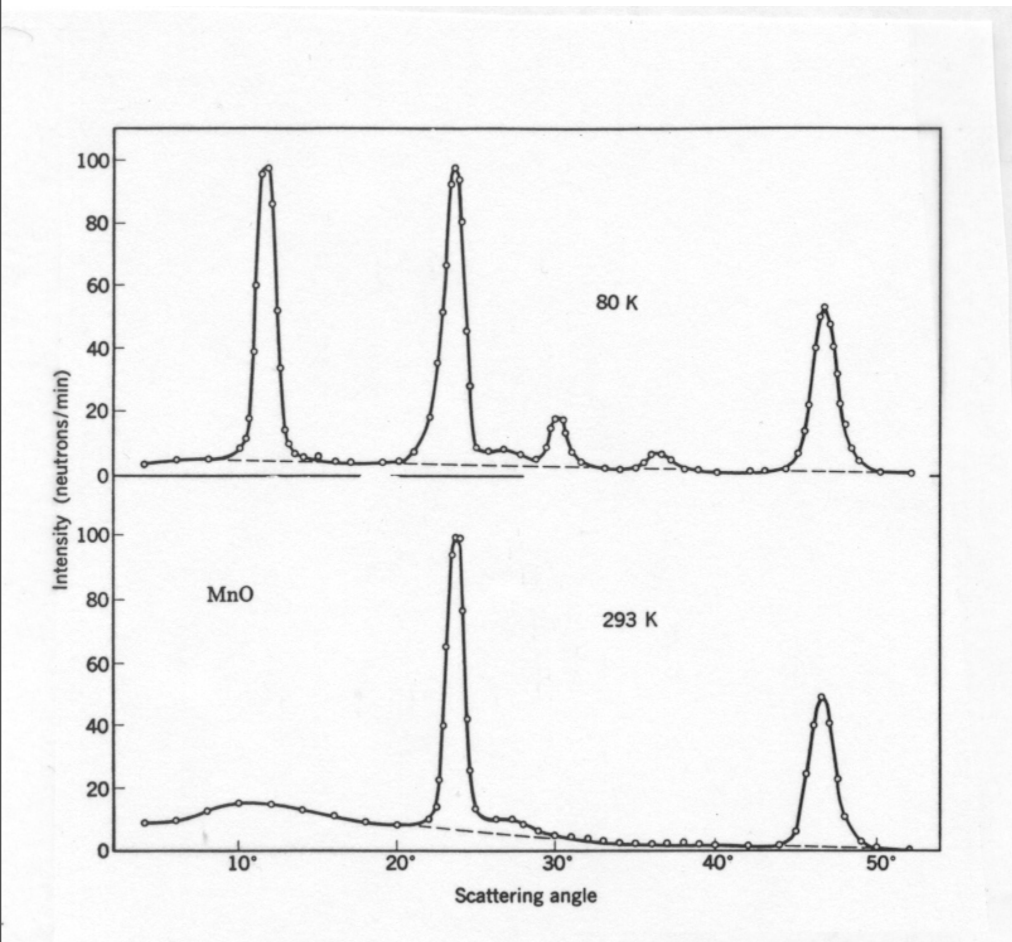


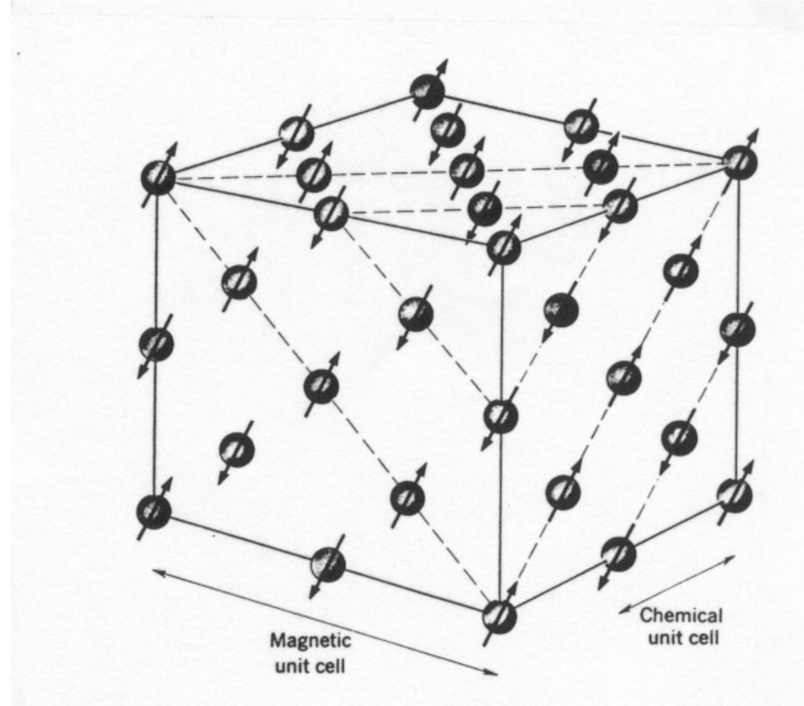


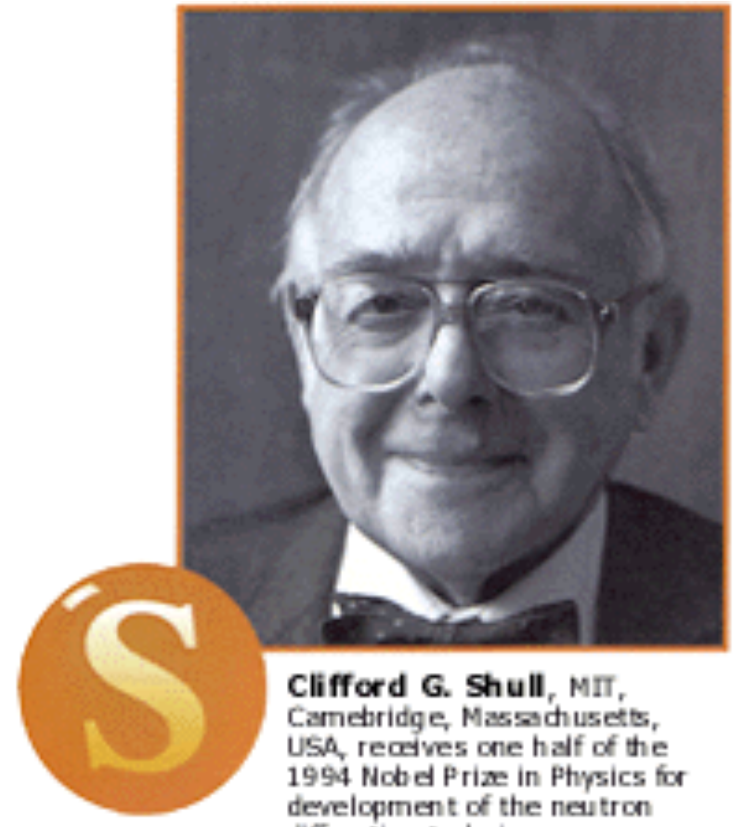




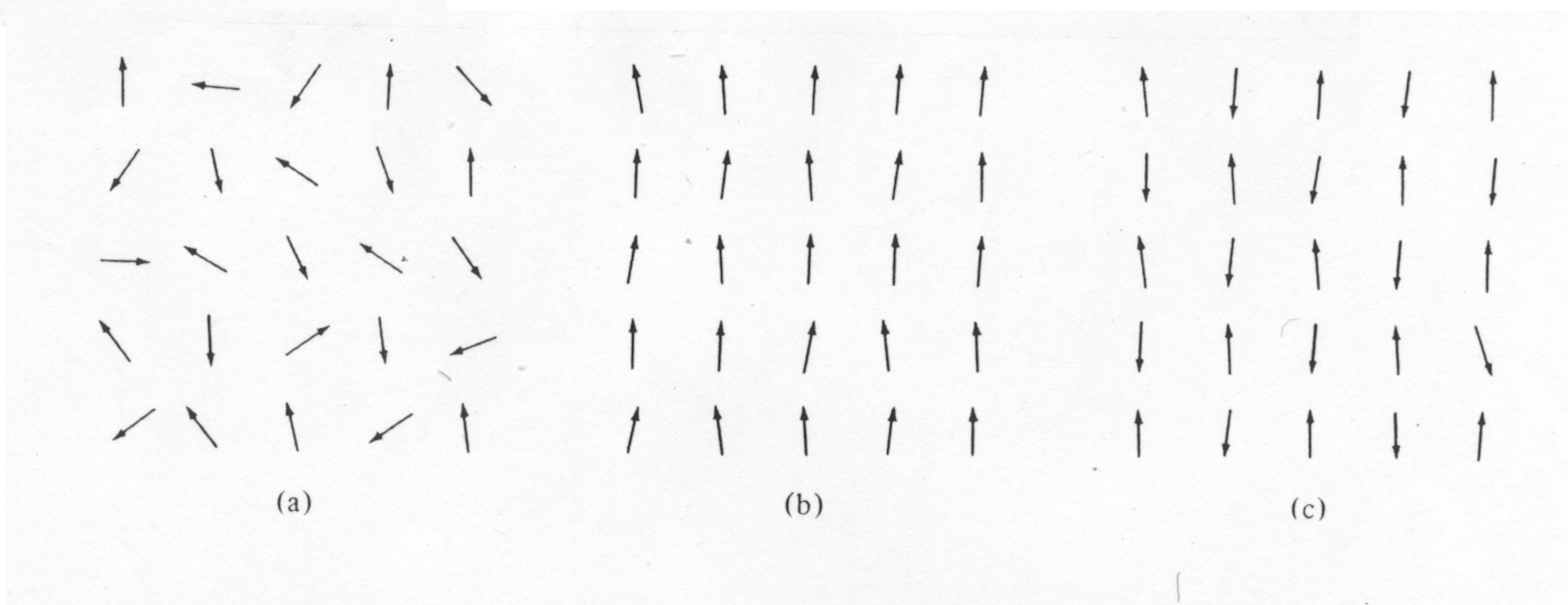
Bragg's Law: nλ = 2d sin(θ)







Clifford G. Shull, MIT, Camebridge, Massachusetts, USA, receives one half of the 1994 Nobel Prize in Physics for development of the neutron diffraction technique.

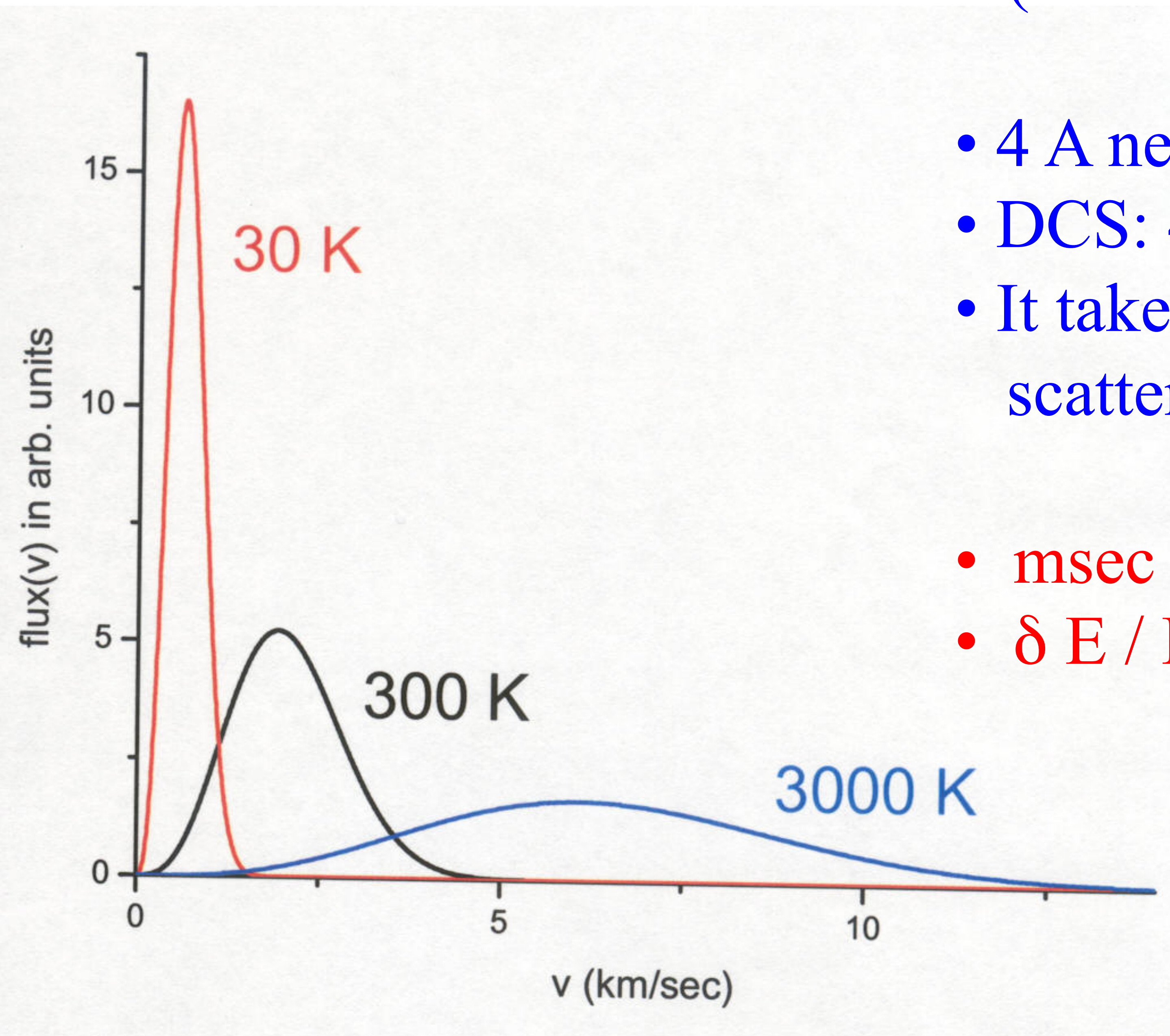


T > Tc

Ferro Antiferro

T < Tc

Neutrons have *mass*
so higher energy means faster – lower energy means slower

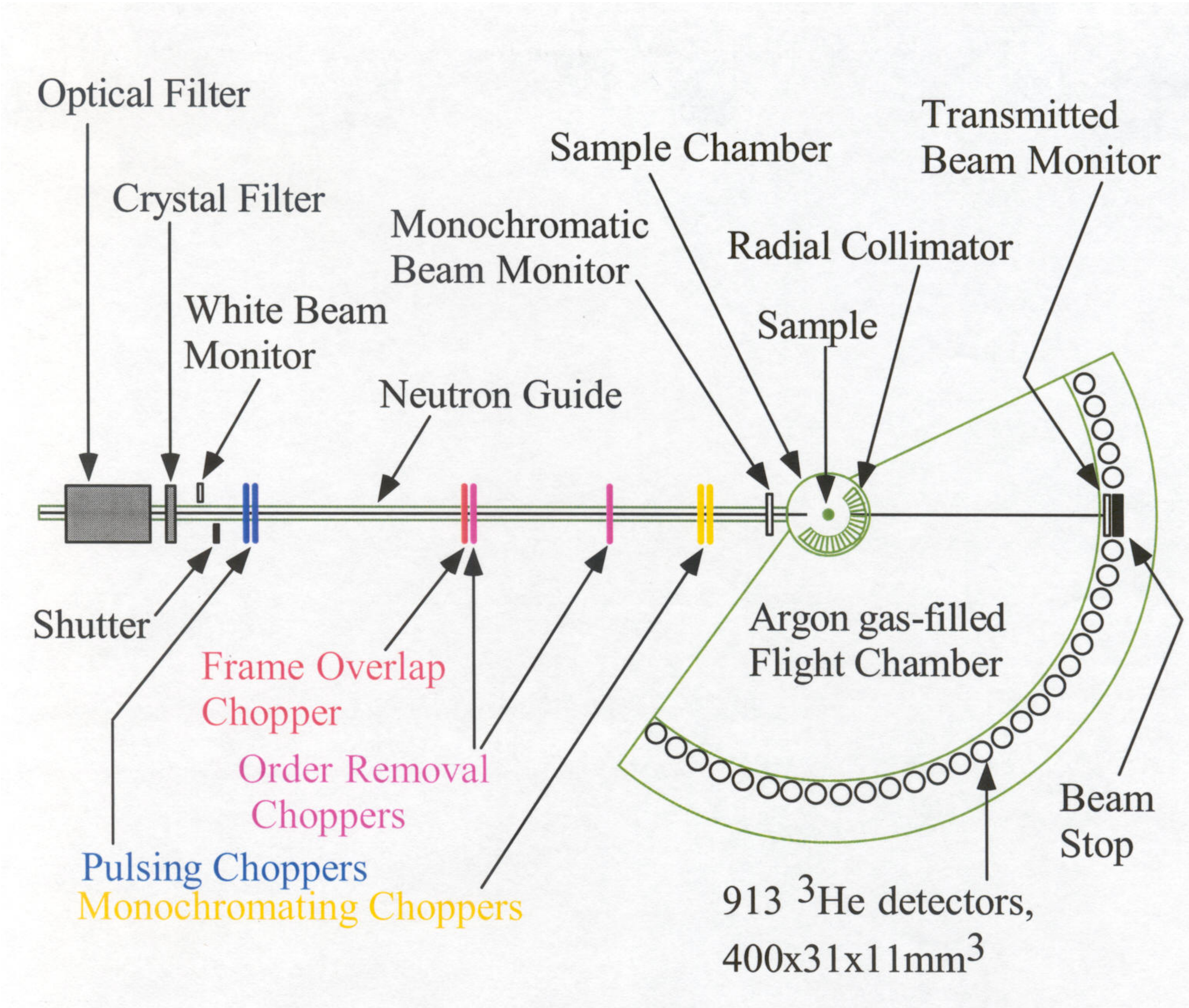


v (km/sec) = 3.96 / λ (A)

- 4 A neutrons move at ~ 1 km/sec
- DCS: 4 m from sample to detector
- It takes 4 msec for elastically scattered 4 A neutrons to travel 4 m

- msec timing of neutrons is easy
- δ E / E ~ 1-3 % - very good !

We can measure a neutron's energy, wavelength by measuring its *speed*


Optical Filter
Crystal Filter
White Beam Monitor
Neutron Guide
Monochromatic Beam Monitor
Sample Chamber
Radial Collimator
Sample
Transmitted Beam Monitor
Shutter
Frame Overlap Chopper
Order Removal Choppers
Pulsing Choppers
Monochromating Choppers
Argon gas-filled Flight Chamber
913 ^{3}He detectors, 400x31x11mm^3
Beam Stop

Motivation for Singlet Basis

$$H = J \sum_{<i,j>} \vec{S}_i \cdot \vec{S}_j$$

$$[\vec{S}_i \cdot \vec{S}_j = S_{\mathrm{z}i} S_{\mathrm{z}j} + S_{\mathrm{x}i} S_{\mathrm{x}j} + S_{\mathrm{y}i} S_{\mathrm{y}j}]$$

Neel type state for 2 spins:

$$\langle \uparrow_i \downarrow_j | \, S_{\mathrm{z}i} S_{\mathrm{z}j} \, | \uparrow_i \downarrow_j \rangle = -\frac{1}{4}$$

$$\langle \uparrow_i \downarrow_j | \, S_{\mathrm{x}i} S_{\mathrm{x}j} \, | \uparrow_i \downarrow_j \rangle = 0$$

$$\langle \uparrow_i \downarrow_j | \, S_{\mathrm{y}i} S_{\mathrm{y}j} \, | \uparrow_i \downarrow_j \rangle = 0$$

$$\langle \uparrow_i \downarrow_j | \, H \, | \uparrow_i \downarrow_j \rangle = -\frac{1}{4} J$$

Entangled (singlet) state:

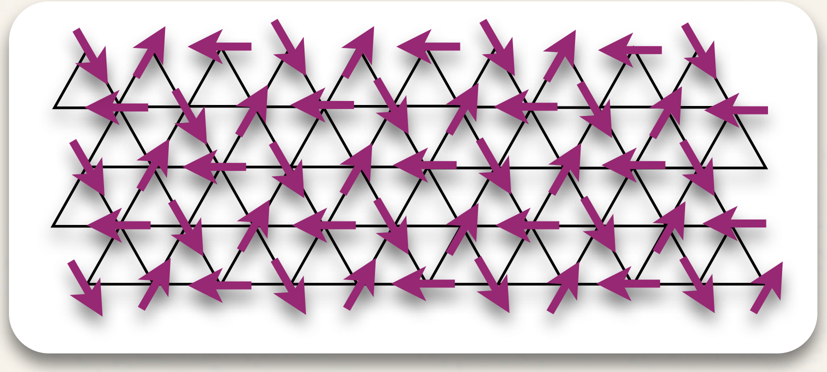
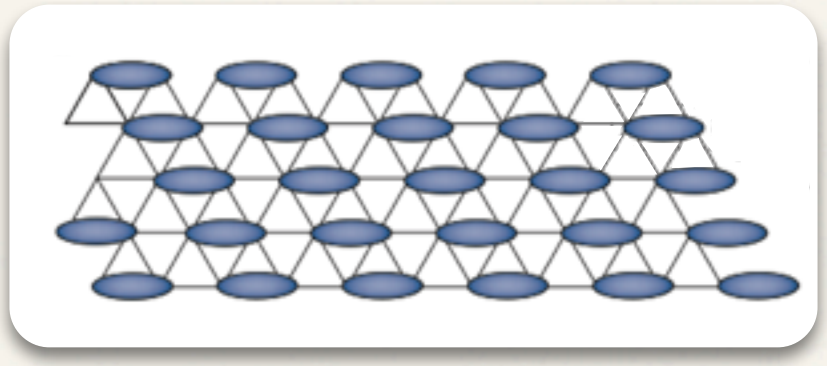
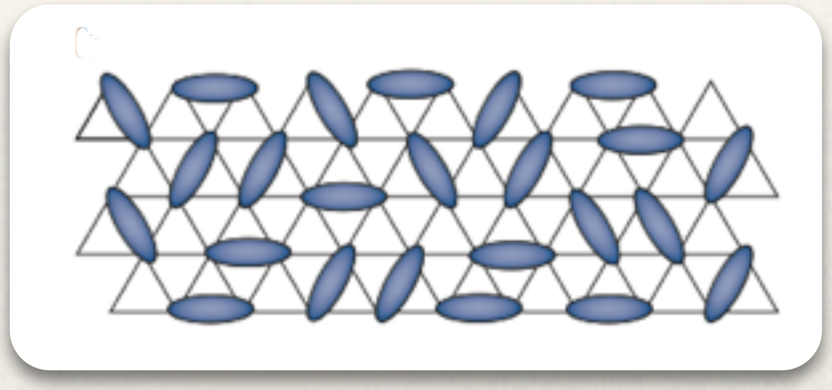
$$(\langle \uparrow_i \downarrow_j | - \langle \downarrow_i \uparrow_j |) S_{\mathrm{z}i} S_{\mathrm{z}j} (| \uparrow_i \downarrow_j \rangle - | \downarrow_i \uparrow_j \rangle) = -\frac{1}{4}$$

$$(\langle \uparrow_i \downarrow_j | - \langle \downarrow_i \uparrow_j |) S_{\mathrm{x}i} S_{\mathrm{x}j} (| \uparrow_i \downarrow_j \rangle - | \downarrow_i \uparrow_j \rangle) = -\frac{1}{4}$$

$$(\langle \uparrow_i \downarrow_j | - \langle \downarrow_i \uparrow_j |) S_{\mathrm{y}i} S_{\mathrm{y}j} (| \uparrow_i \downarrow_j \rangle - | \downarrow_i \uparrow_j \rangle) = -\frac{1}{4}$$

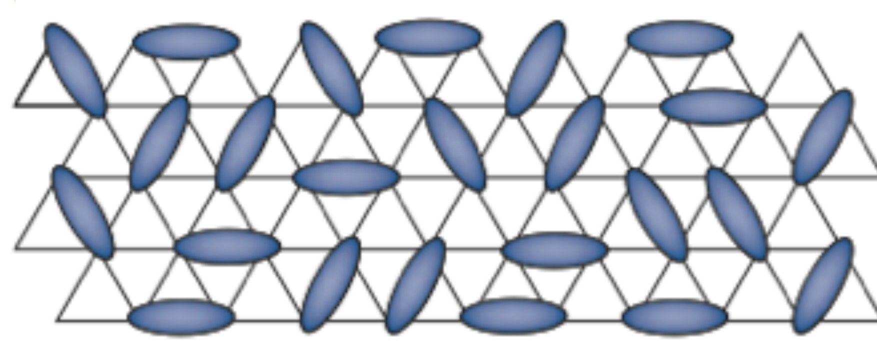
$$(\langle \uparrow_i \downarrow_j | - \langle \downarrow_i \uparrow_j |) H (| \uparrow_i \downarrow_j \rangle - | \downarrow_i \uparrow_j \rangle) = -\frac{3}{4} J$$

Symmetry in the Ground State

Neel State		Broken Spin Rotational Symmetry
Valence Bond Crystal		Broken Translational (Lattice) Symmetry
Spin Liquid (RVB)		Spin and Lattice Symmetry Preserved

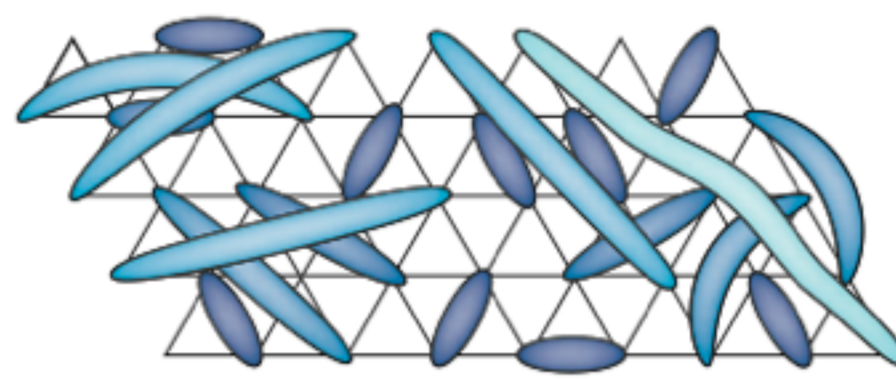
Possible Ground State: Only Nearest Neighbour Bonds

$$| \Psi_{\mathrm{sRVB}} \rangle = \Big| \quad \Big\rangle + \cdots$$



“Short Range RVB”

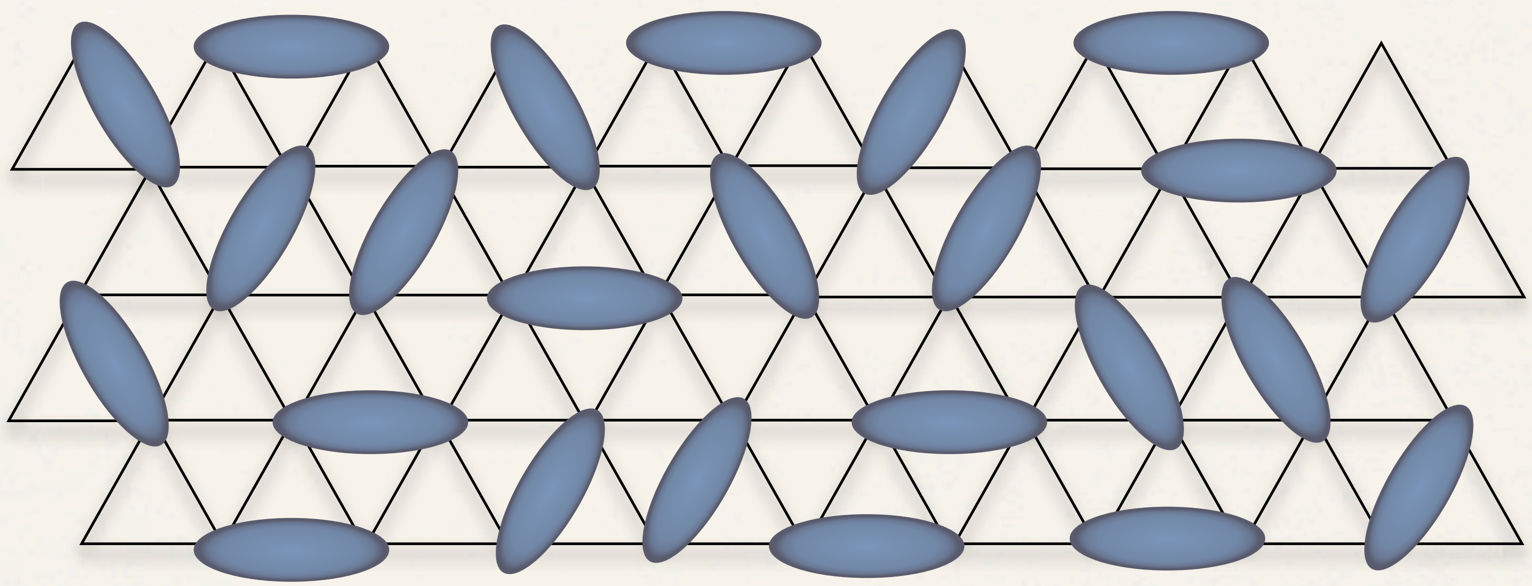
Possible Ground State: All Bond Lengths

$$| \, \Psi_{\mathrm{lRVB}} \, \rangle = \Big|$$  $$\Big\rangle + \cdots$$

"Long Range RVB"

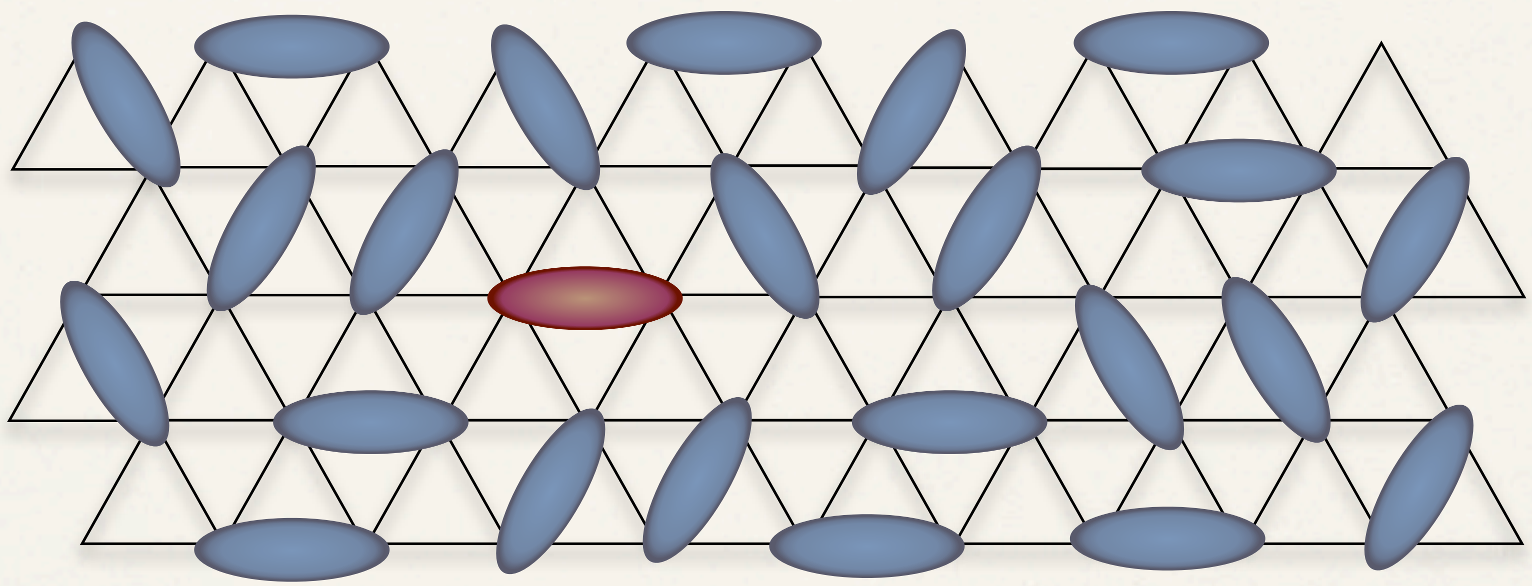
Spin Excitations: Spinons in sRVB

in sRVB state, creating 2 spinons costs $\Delta E = +J$, but then the spinons are free to move around by reorganizing singlets



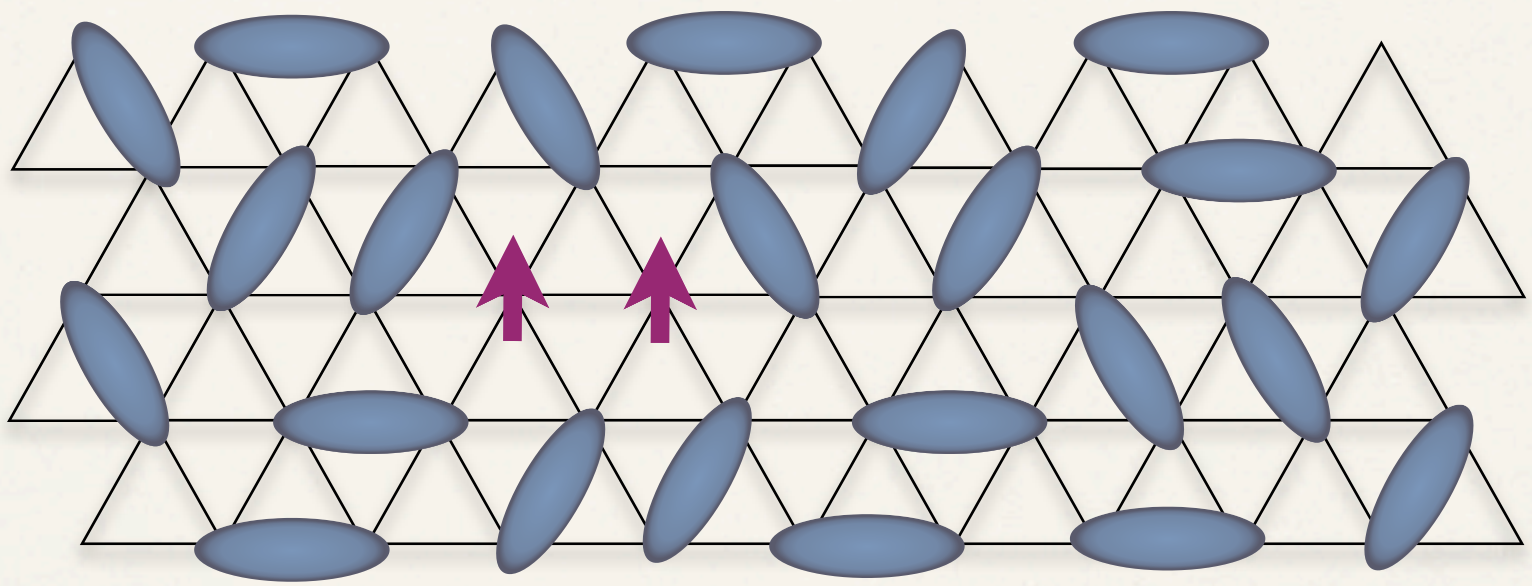
Spin Excitations: Spinons in sRVB

in sRVB state, creating 2 spinons costs $\Delta E = +J$, but then the spinons are free to move around by reorganizing singlets



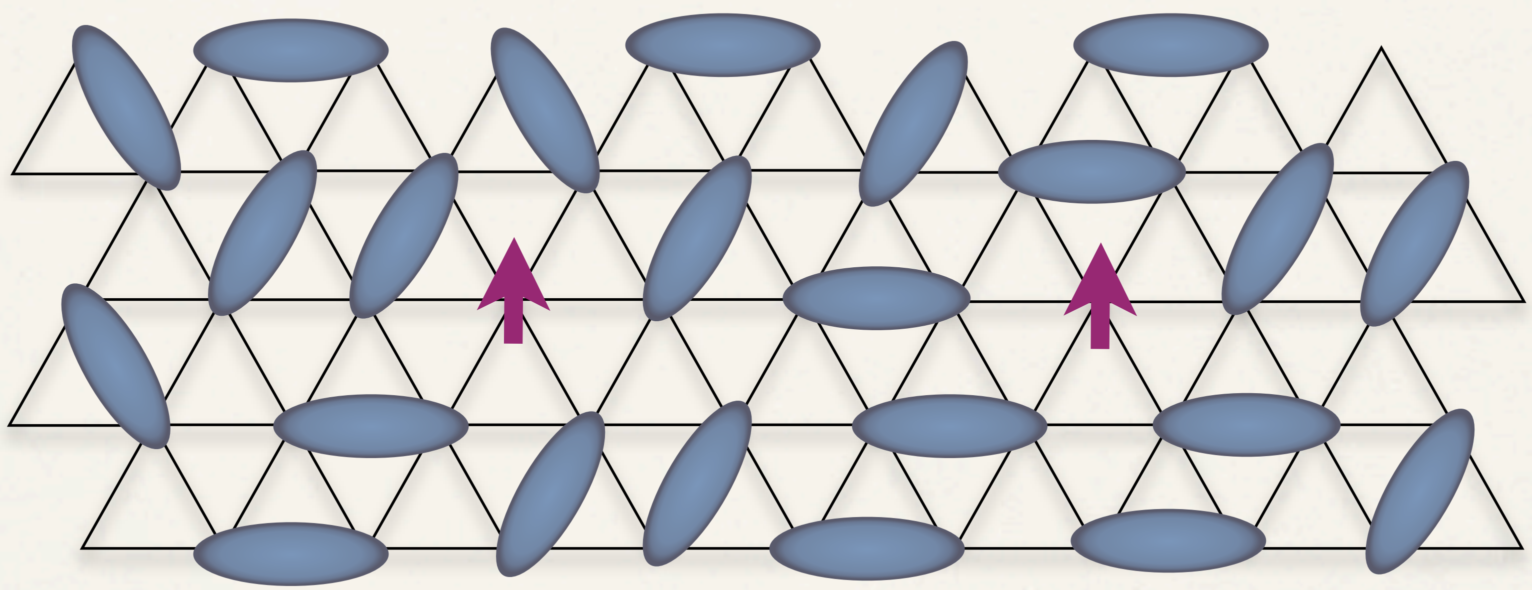
Spin Excitations: Spinons in sRVB

in sRVB state, creating 2 spinons costs $\Delta E = +J$, but then the spinons are free to move around by reorganizing singlets



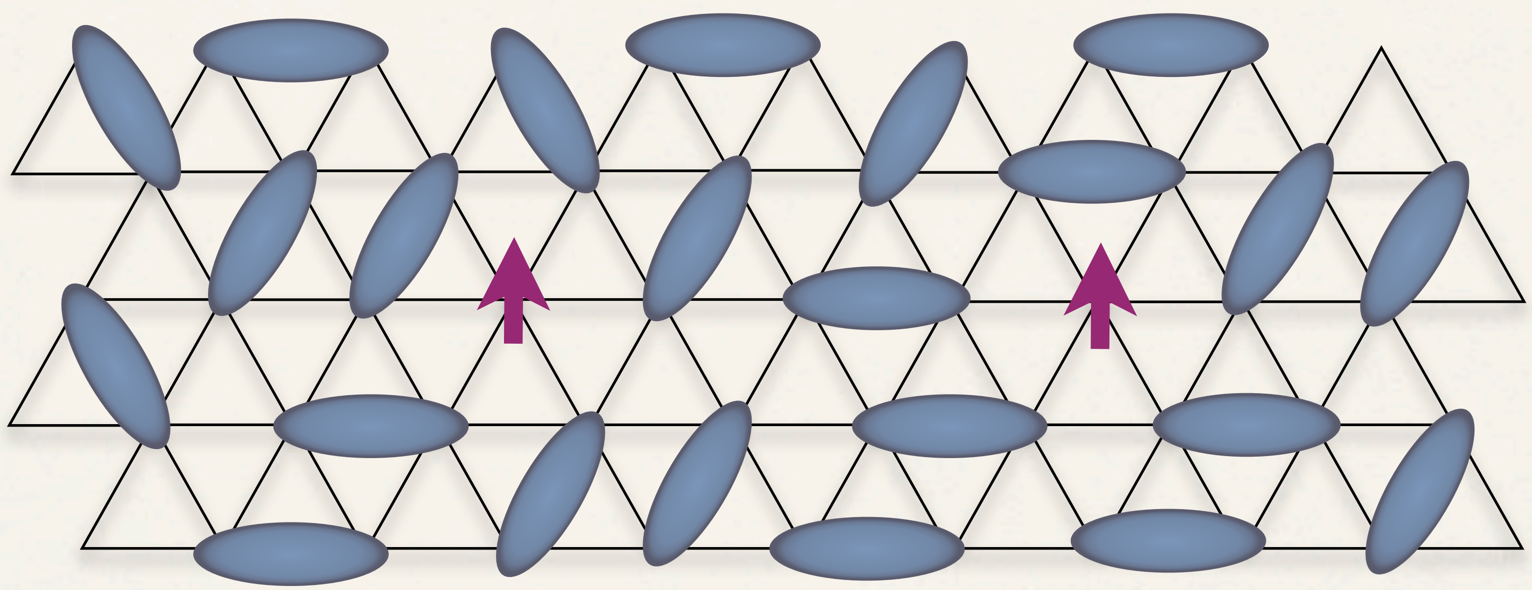
Spin Excitations: Spinons in sRVB

in sRVB state, creating 2 spinons costs $\Delta E = +J$, but then the spinons are free to move around by reorganizing singlets



Spin Excitations: Spinons in sRVB

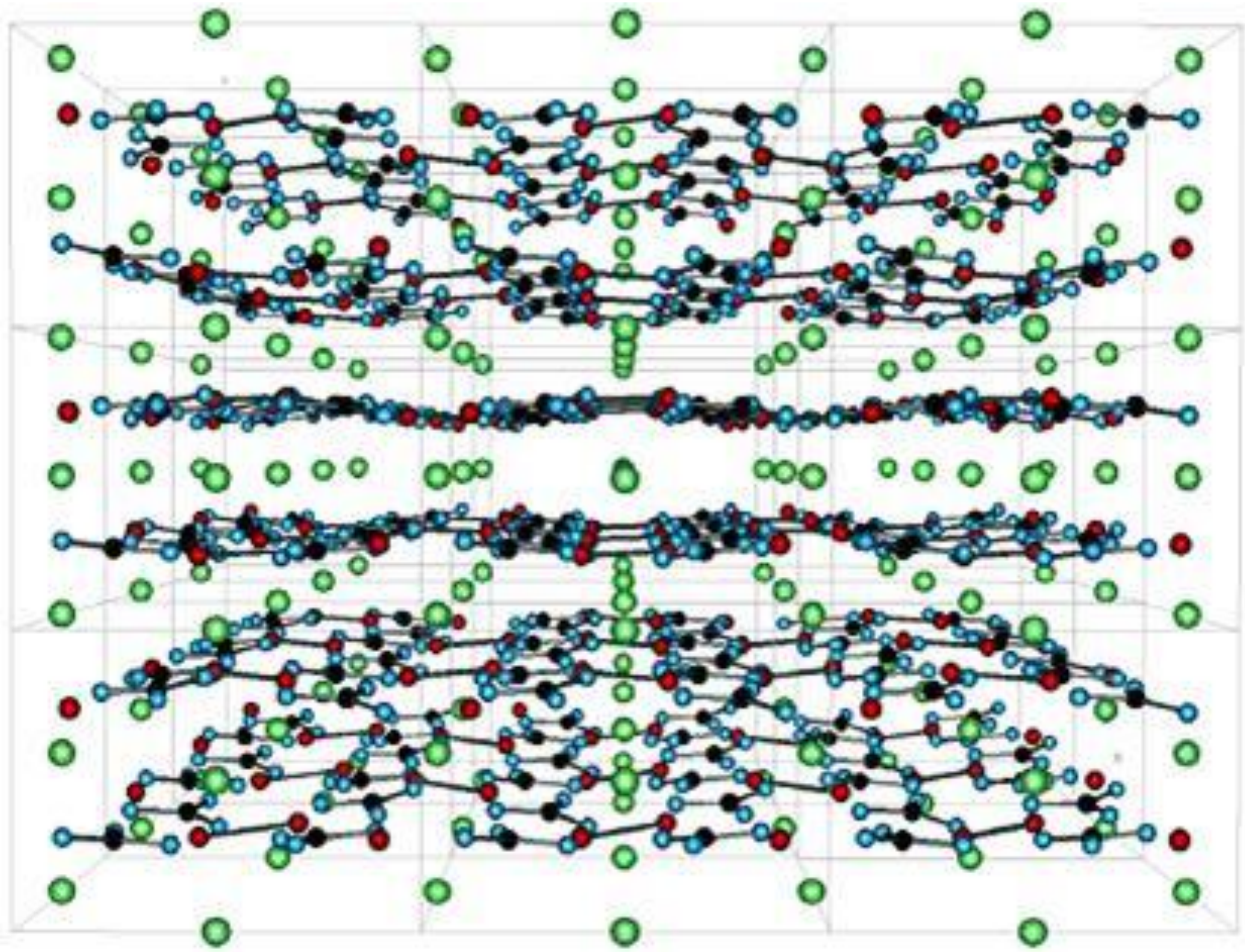
in sRVB state, creating 2 spinons costs $\Delta E = +J$, but then the spinons are free to move around by reorganizing singlets



NOTES:

1) Spinons are *deconfined*, but there is a *"spin gap"* ($\Delta E = +J$)

2) Charges are not mobile, only spins - it is like half an electron

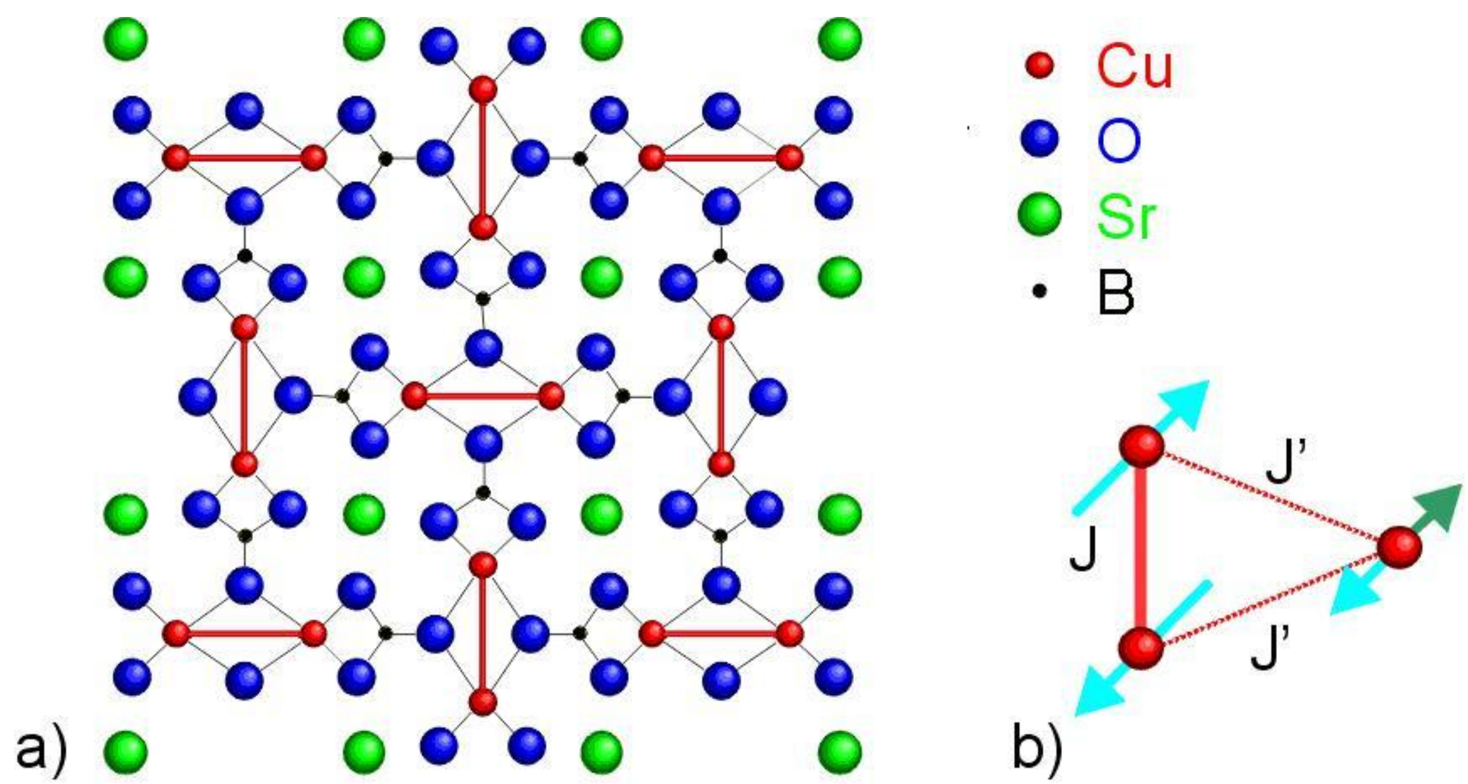


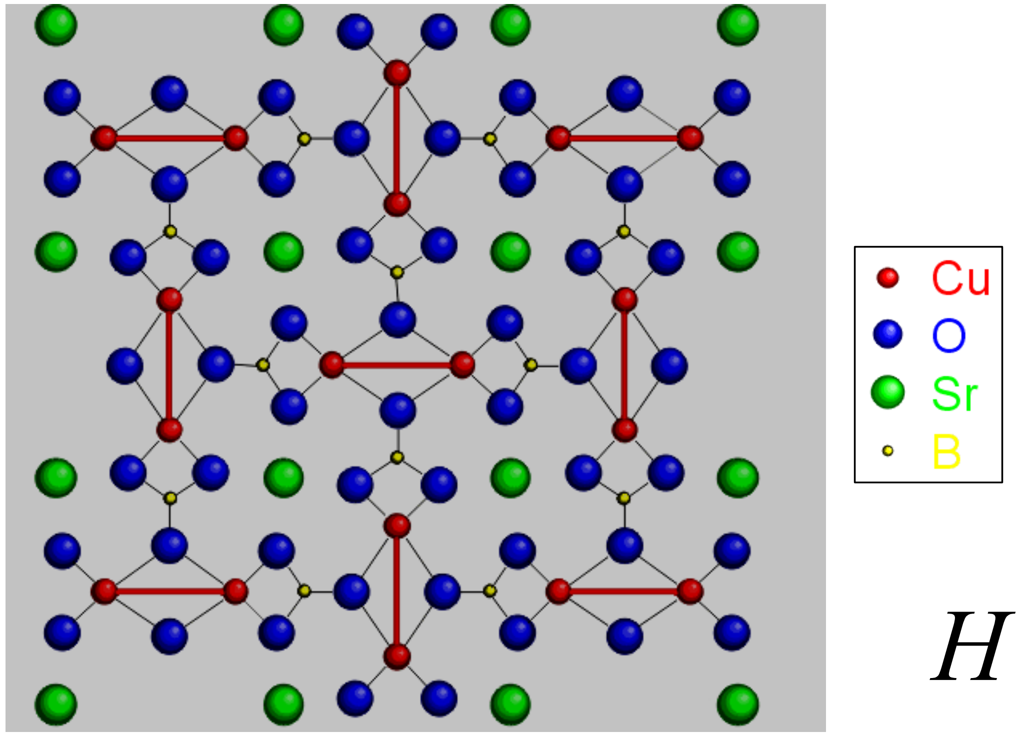
$SrCu_2(BO_3)_2$

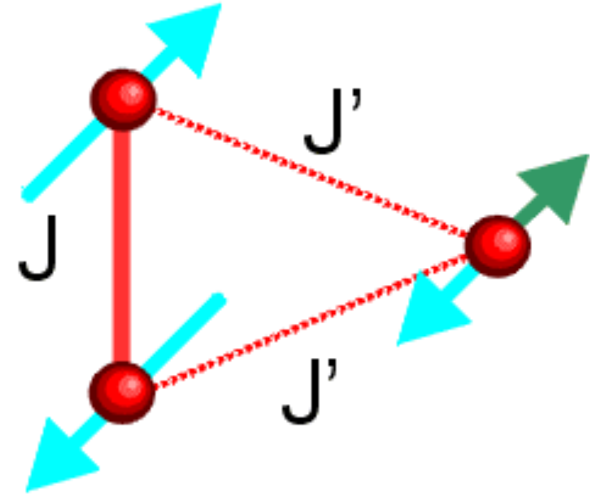
- Quasi-2D Cu-BO_3 planes
- Sr ions between planes
- Mott insulator

S=1/2 moments at Cu^{2+} sites arranged in orthogonal dimers on square lattice

- Shastry-Sutherland model







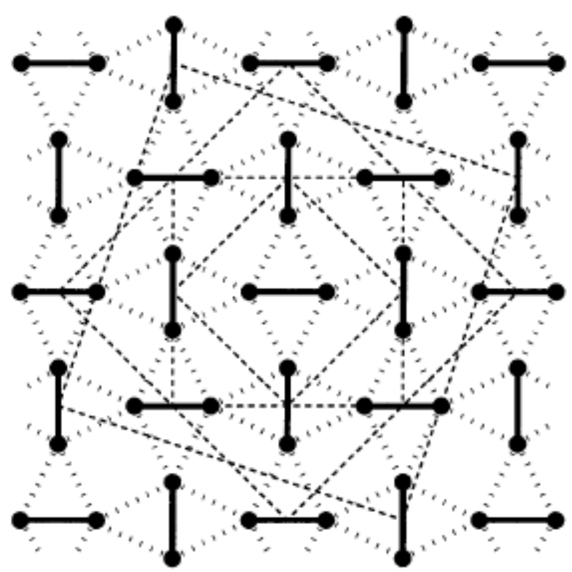
$$H = J\sum_{nn} S_i \cdot S_j + J'\sum_{nnn} S_i \cdot S_j$$

$|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$

J'/J = x_{crit} ~ 0.69

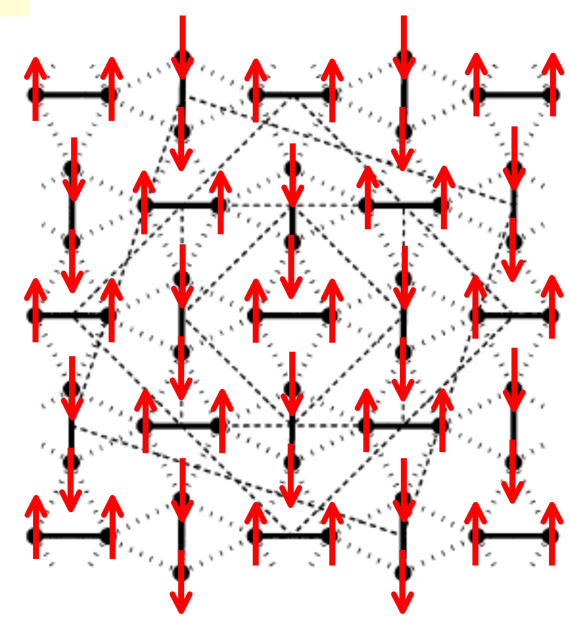
Case I

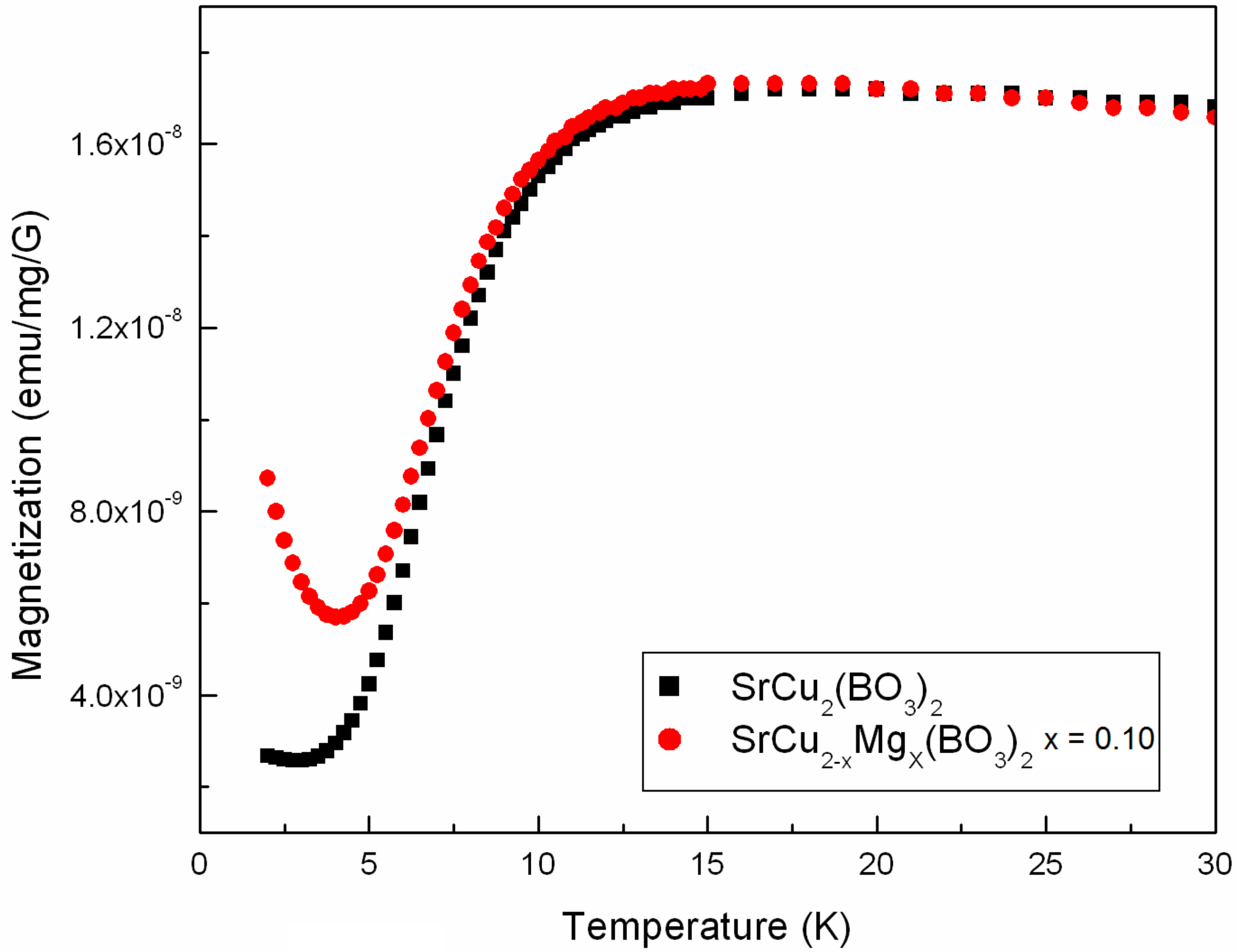
J>>J' : Ground State – Isolated singlet dimer on *nn* (diagonal) bonds



Case II

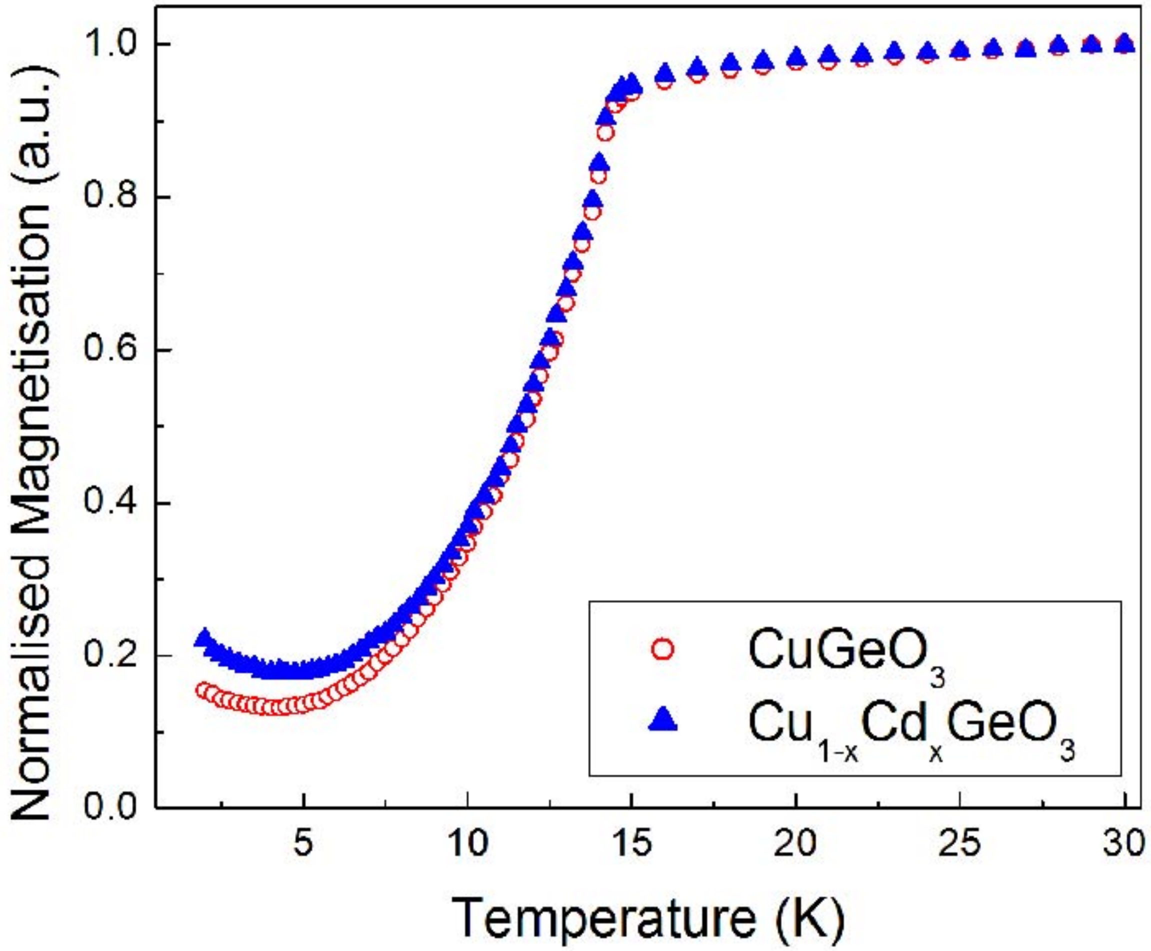
J<<J' : Ground State Ordered Neel antiferromagnetic

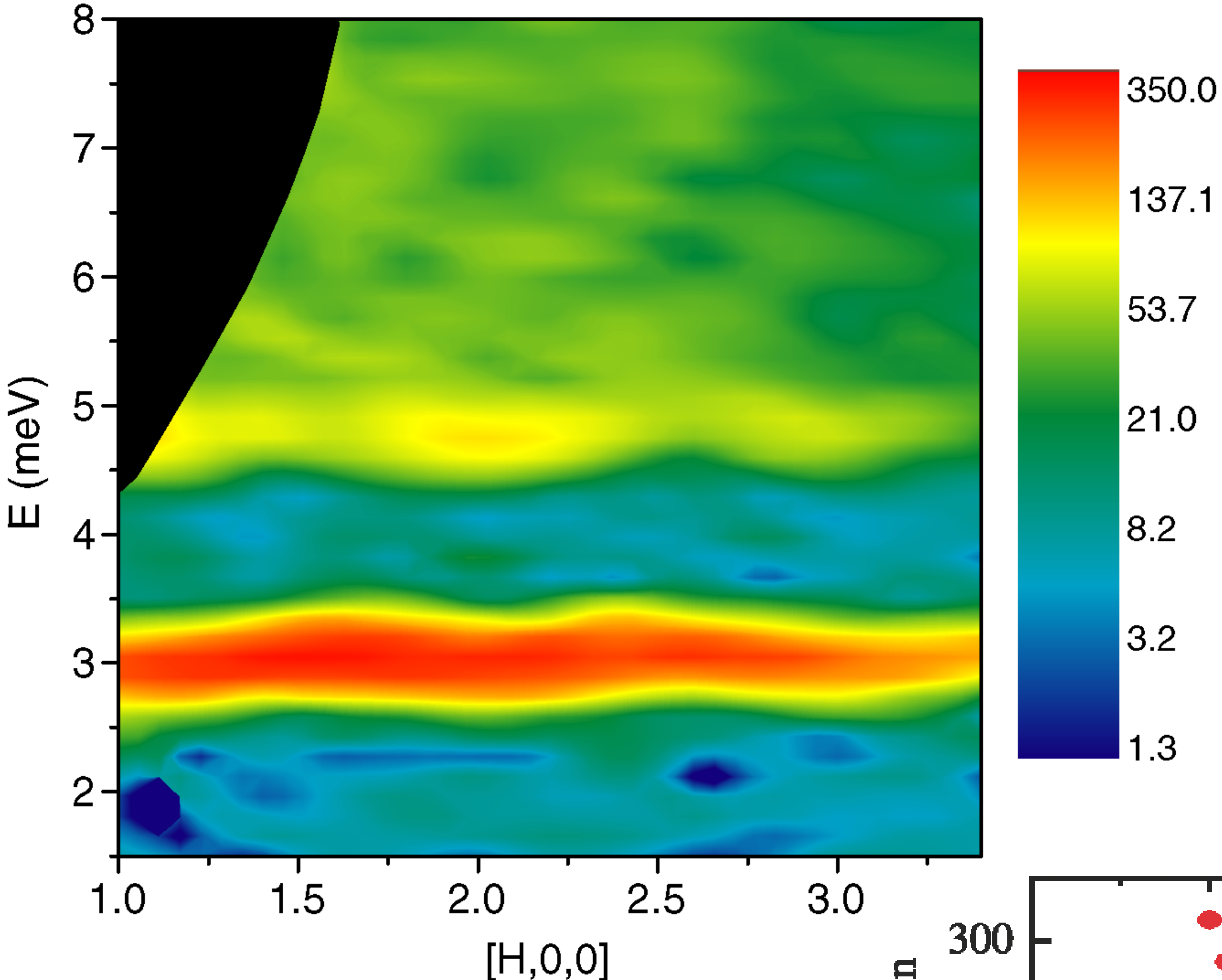




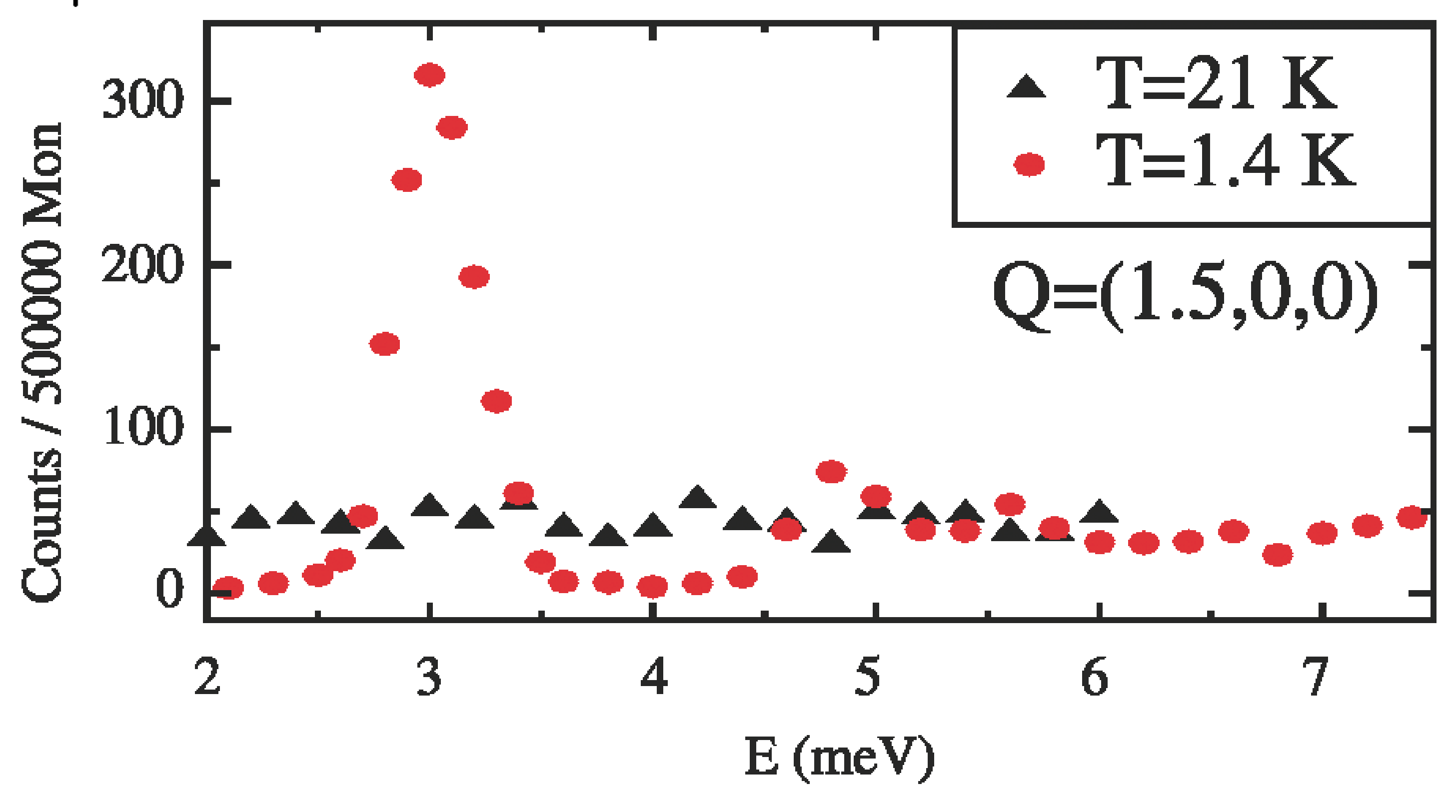
DC susceptibility shows singlet ground state – no phase transition in $SrCu_2(BO_3)_2$

DC susceptibility shows singlet ground state – Spin-Peierls phase transition in $CuGeO_3$





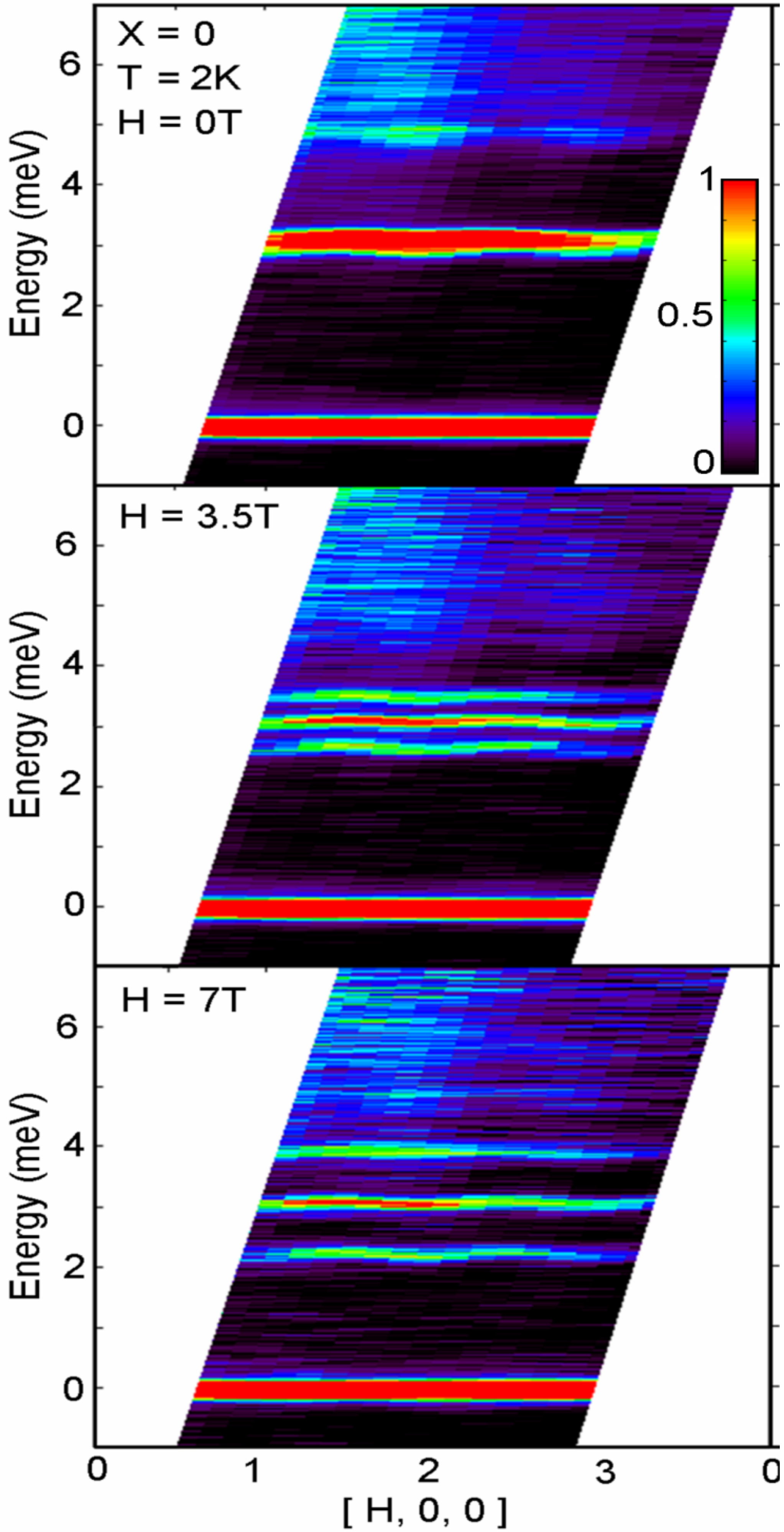
3 Bands of Singlet-Triplet excitations directly observable with neutron scattering



Δ~ 3 meV

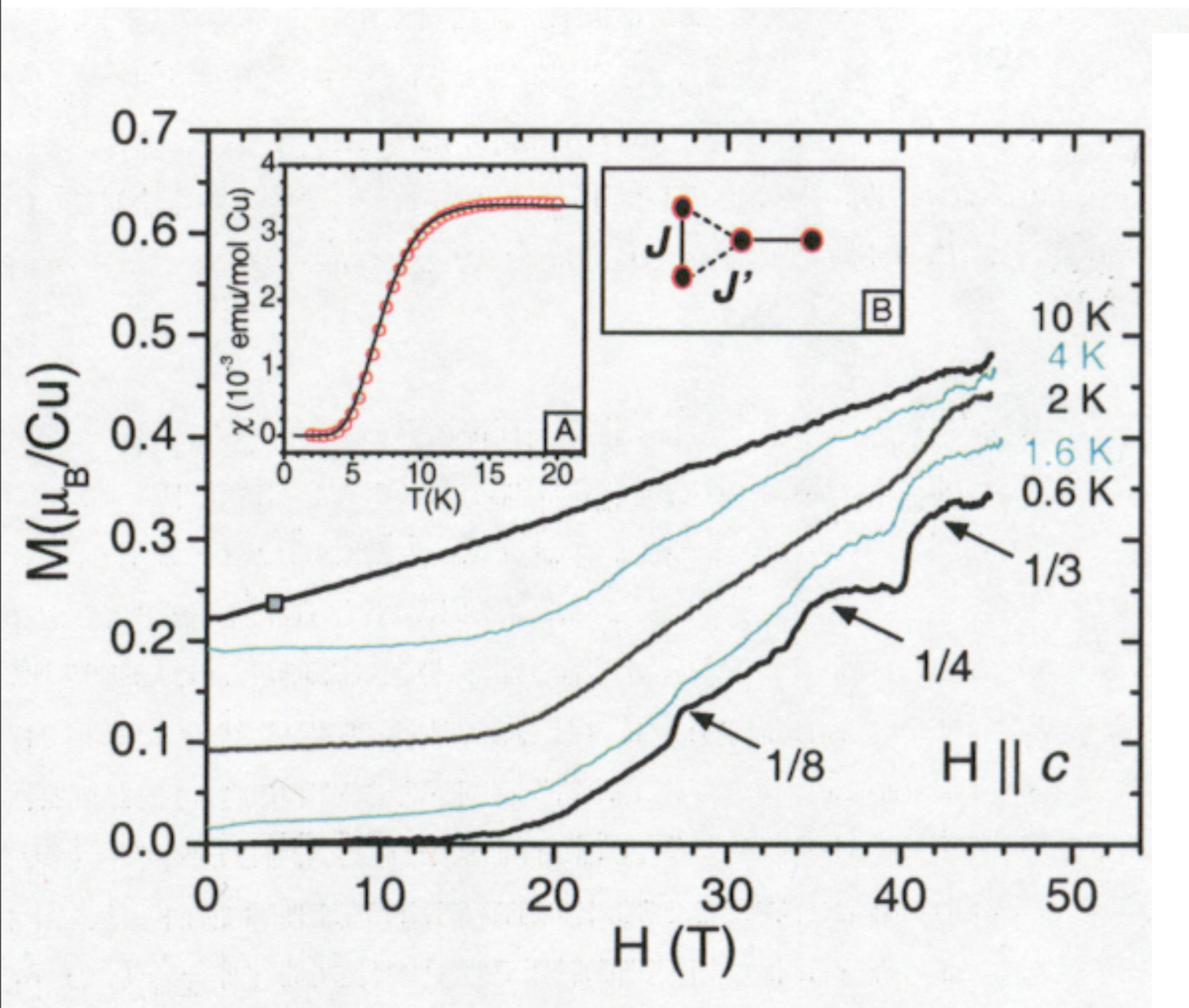
N=2 ~ 4.85 meV

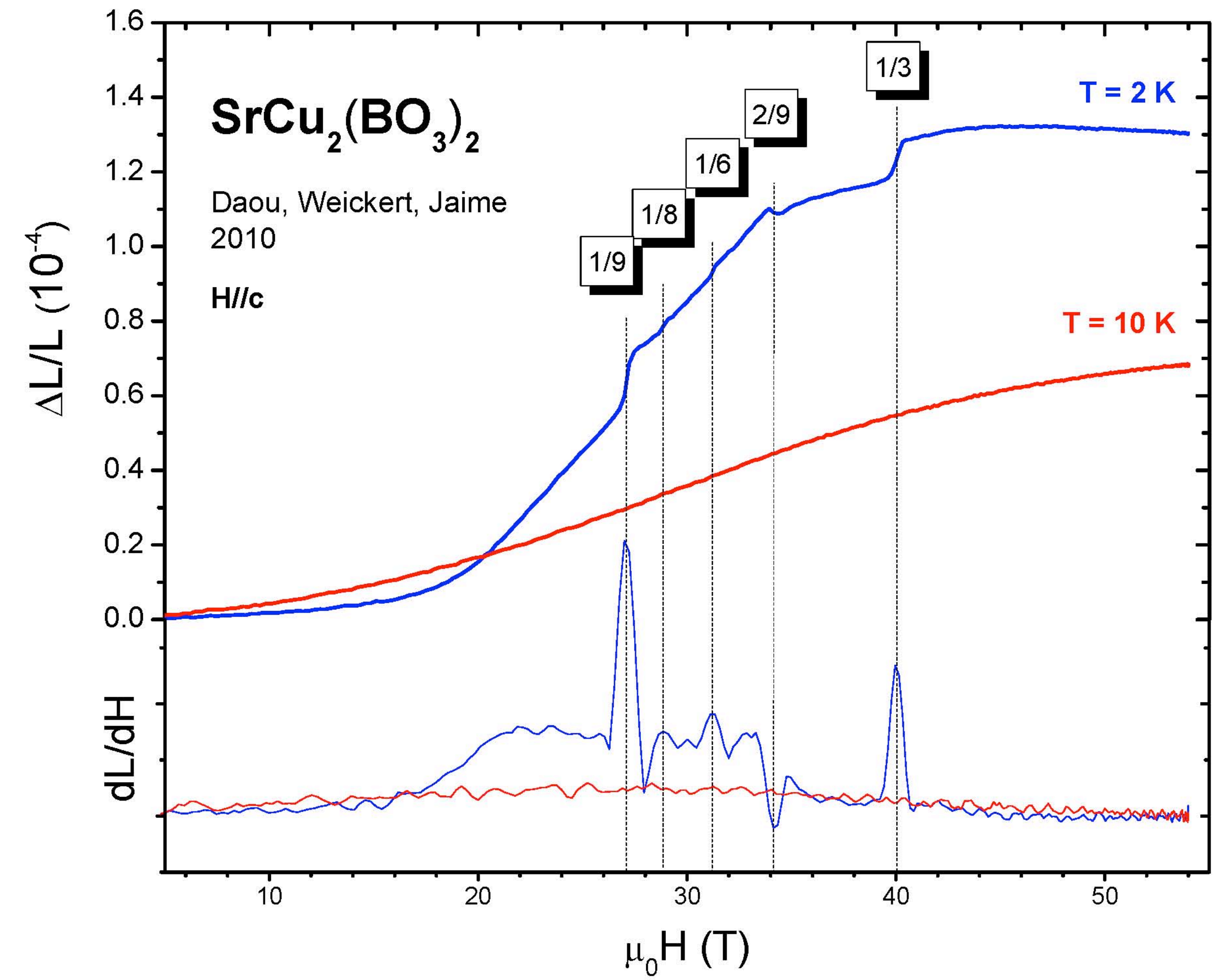
N=3 ~ 9.5 meV



Triplets split in a magnetic field

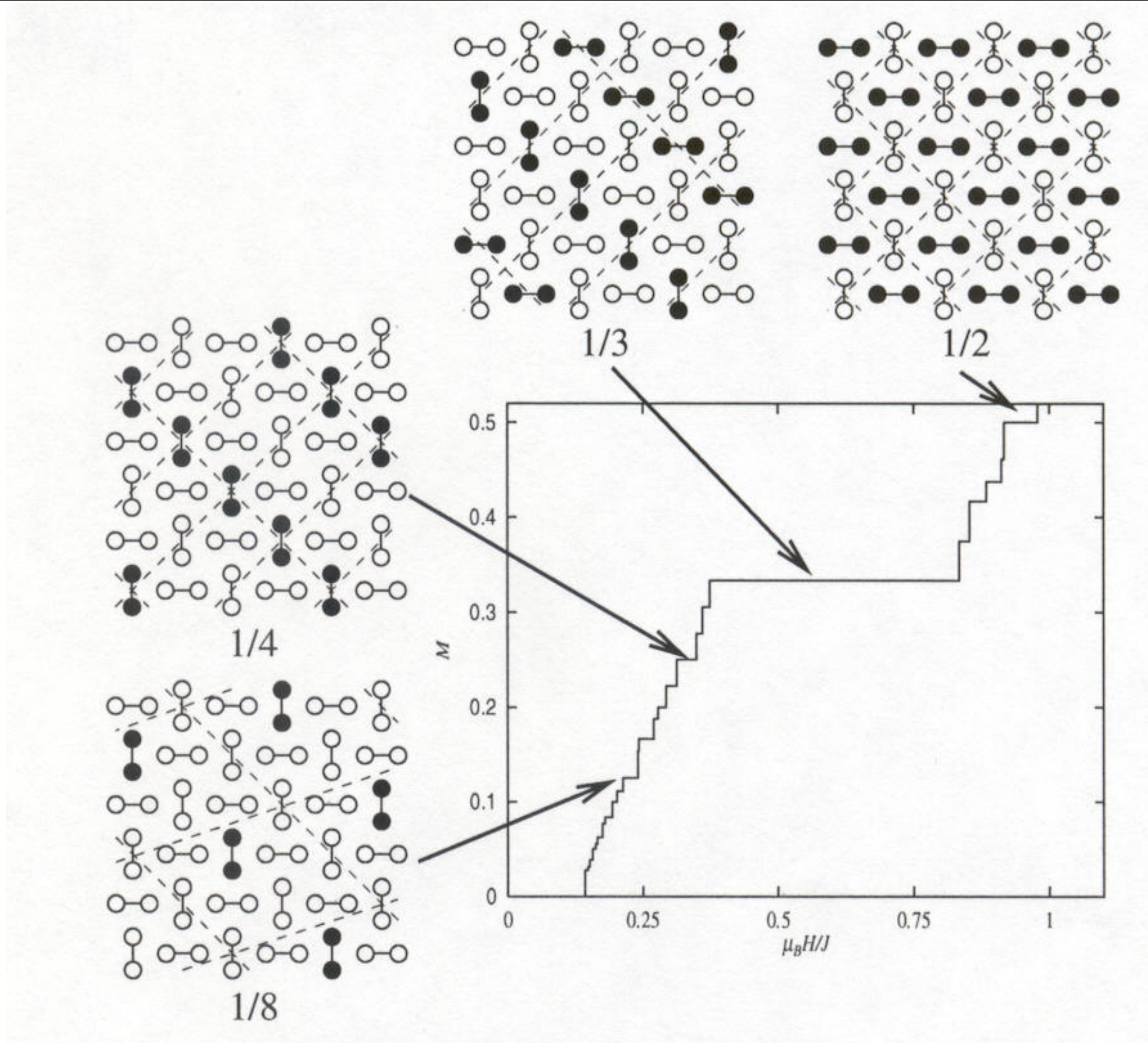
$$\Delta\mathrm{E} = \mathrm{g}\mu_B\mathrm{H}$$





SrCu$_2$(BO$_3$)$_2$ enters collective singlet state at low temperature:

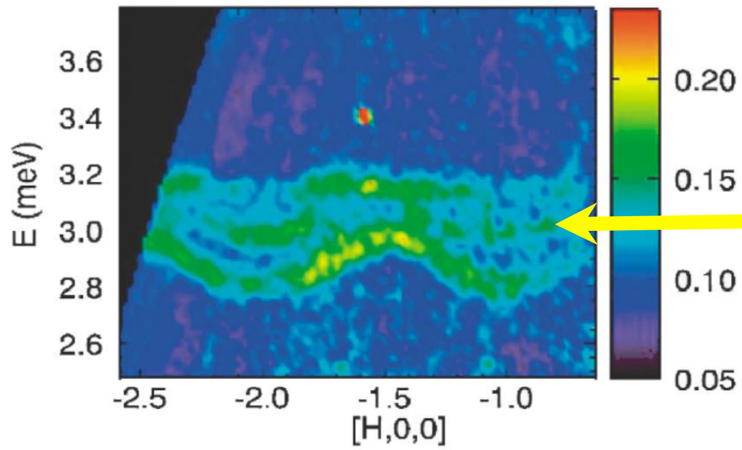
Large magnetic fields drive triplets to zero energy; producing steps in the magnetization.

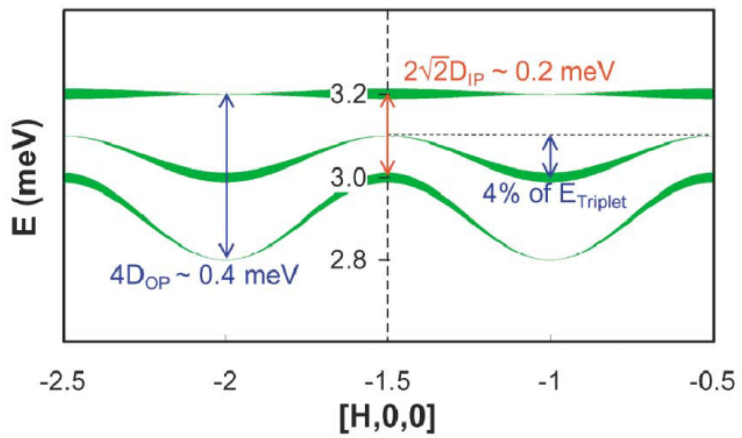


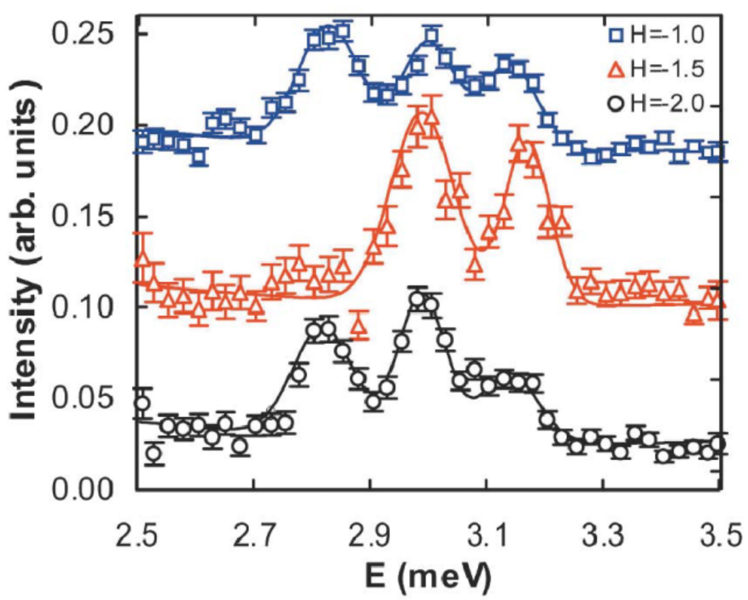
Hard core Boson models for interacting triplets (Miyahara and Ueda, J. Phys. CM 15, R327, 2003)

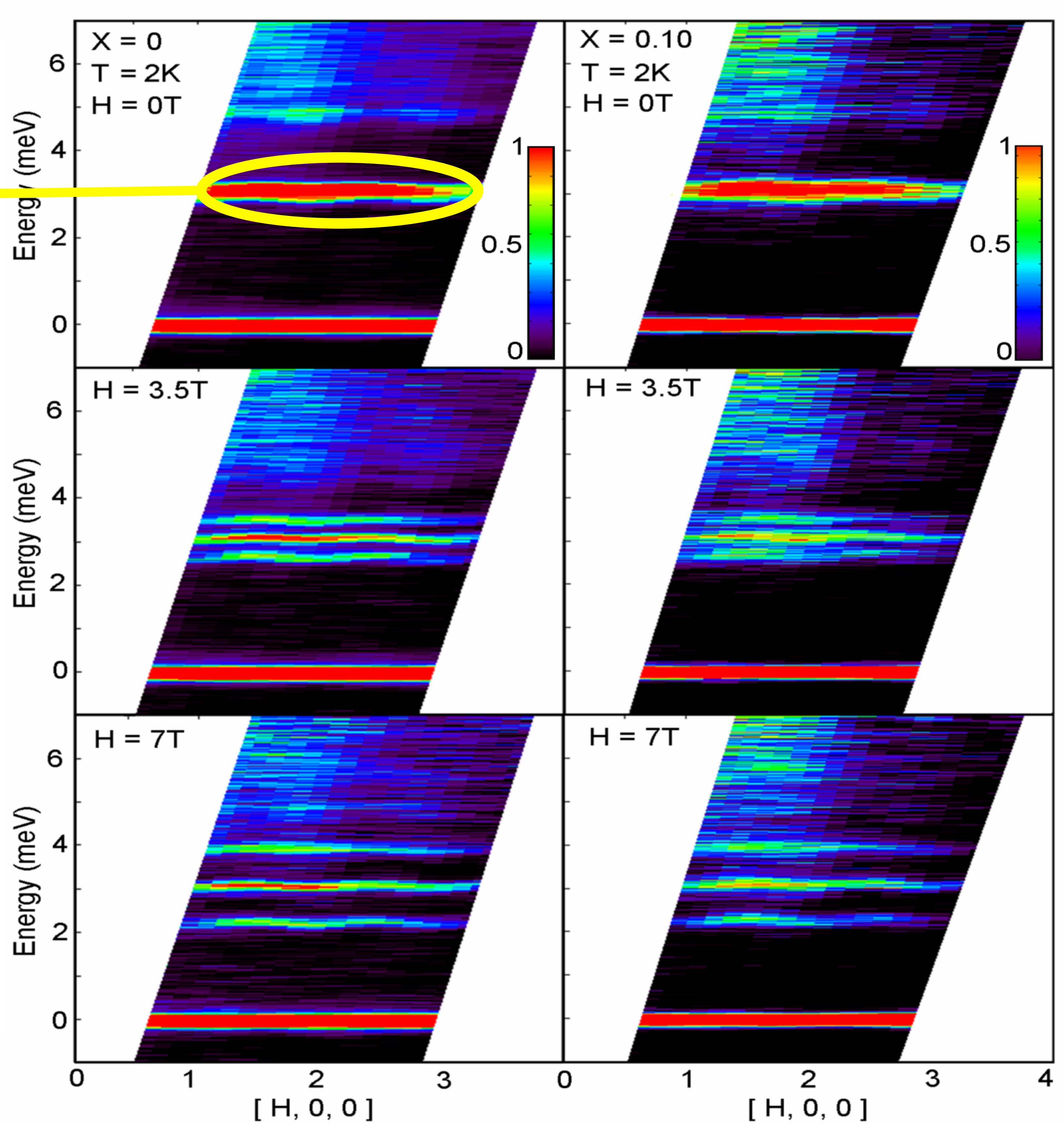
Plateaus appear at commensurate filling of lattice with triplets

Time of Flight Neutron Scattering Data

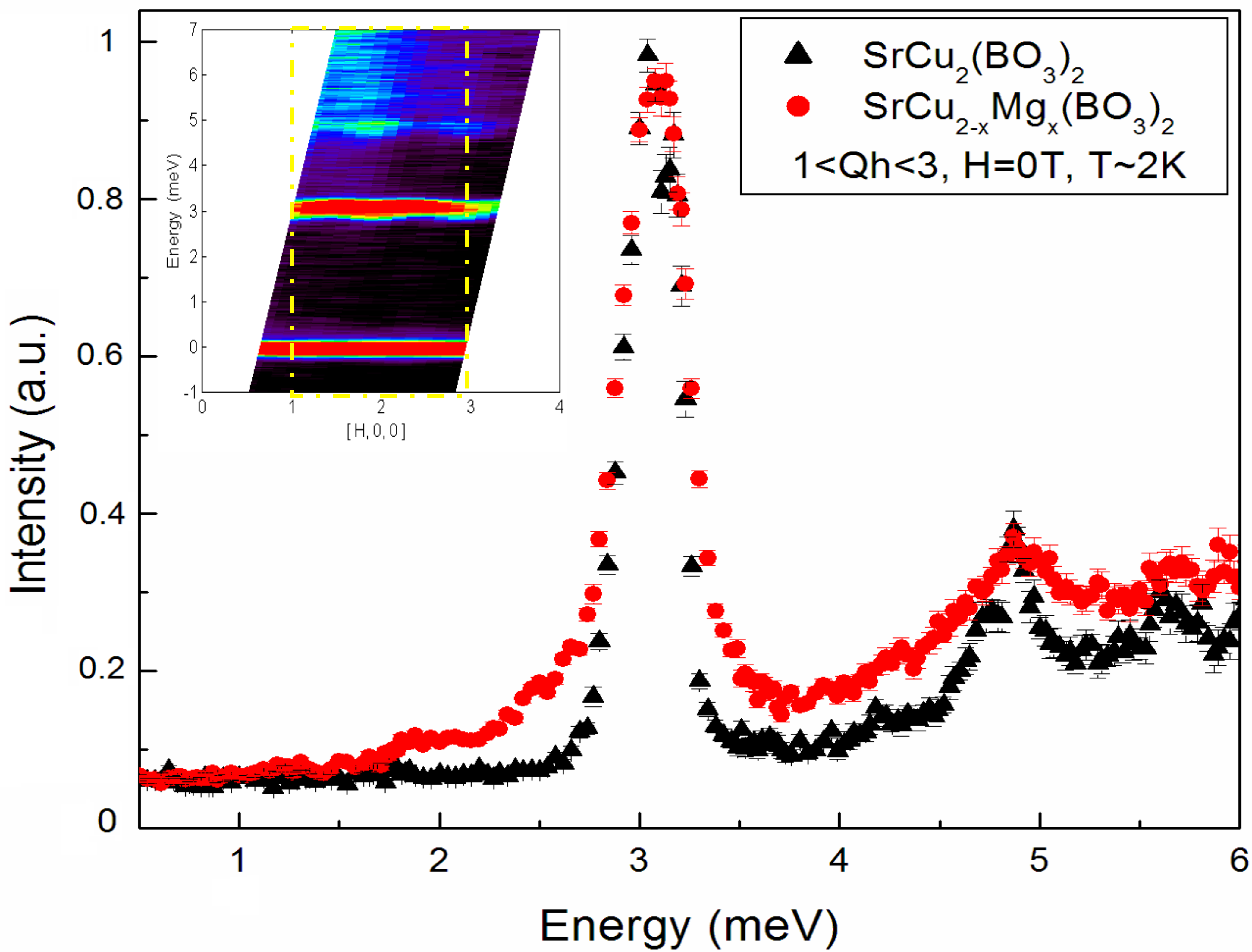








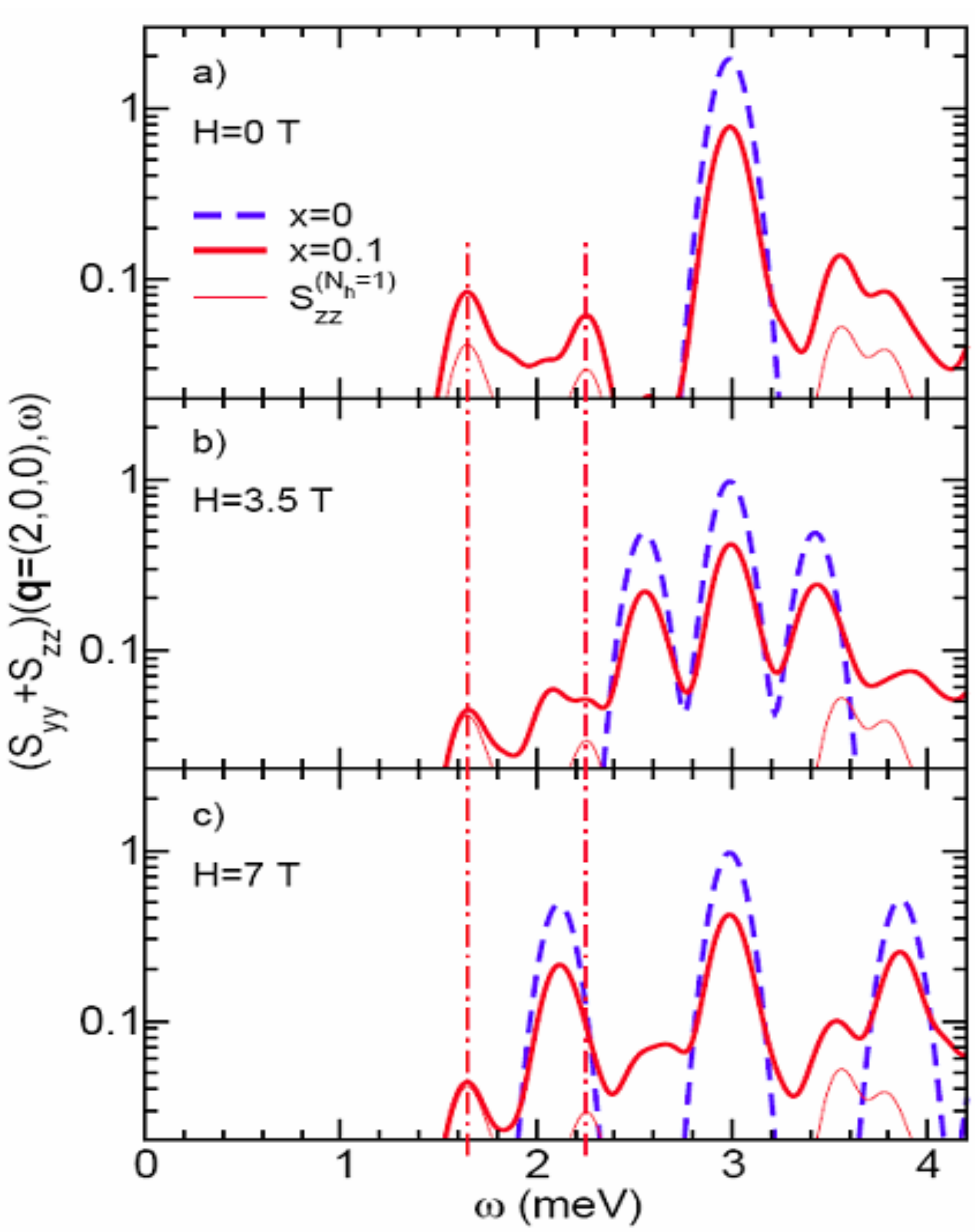
B.D. Gaulin et al,
PRL 93, 267202 (2004)

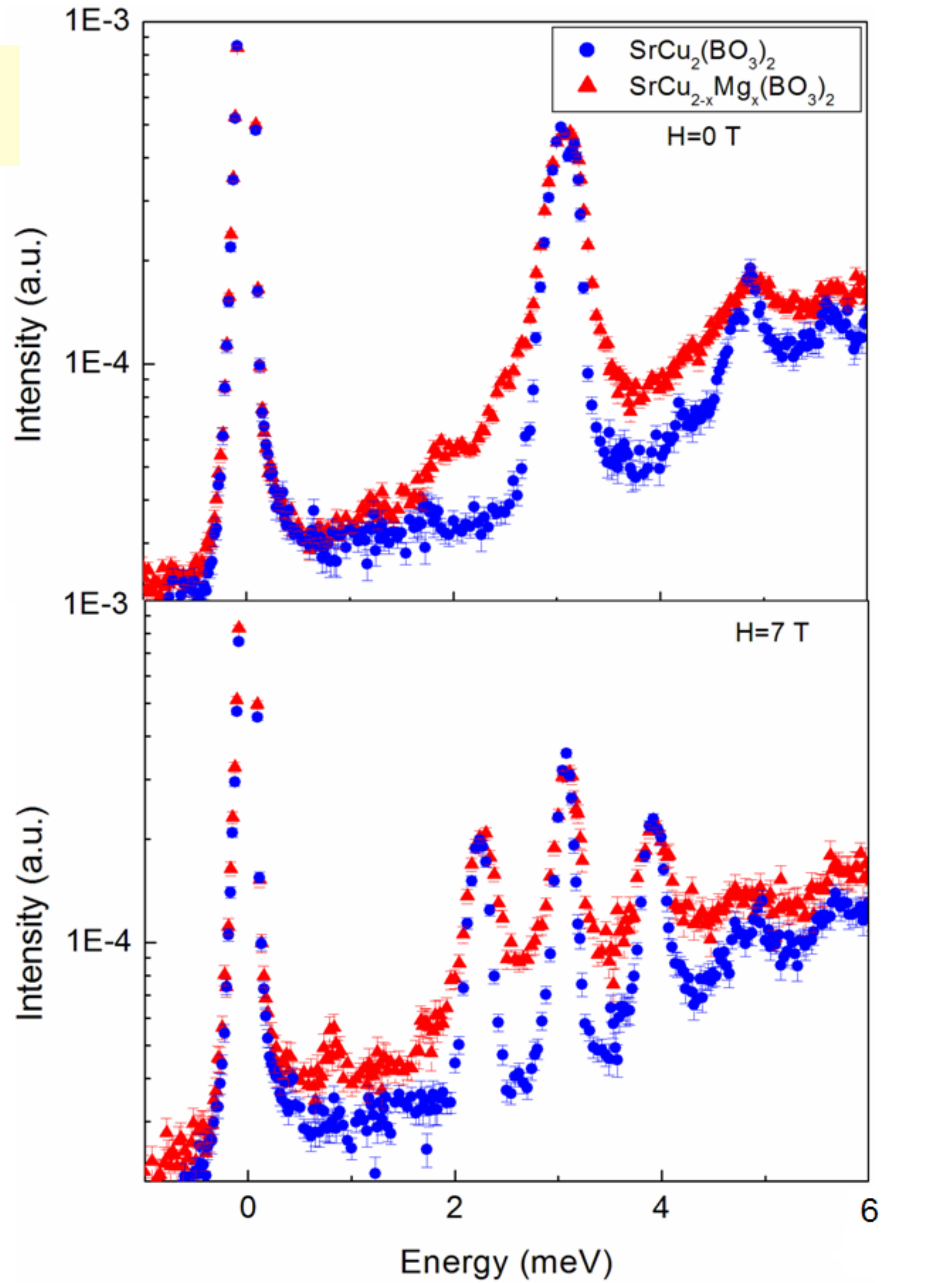


Wednesday, May 5, 2010

Comparison to Theory

Calculated dynamical spin structure factor using the zero temperature Lanczos method.

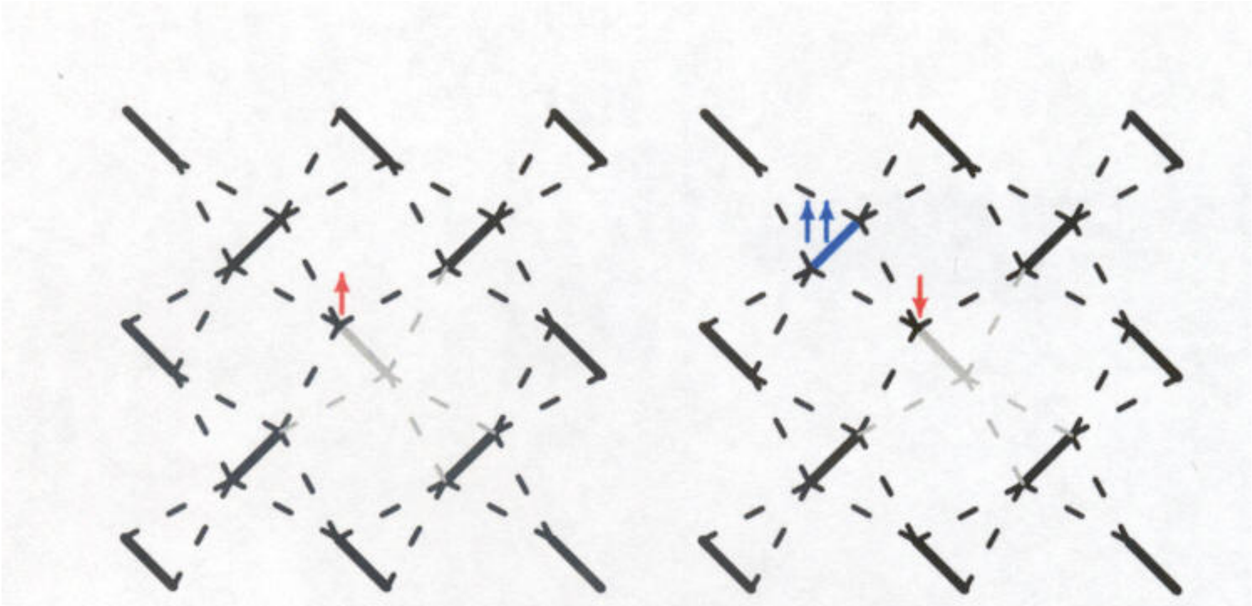




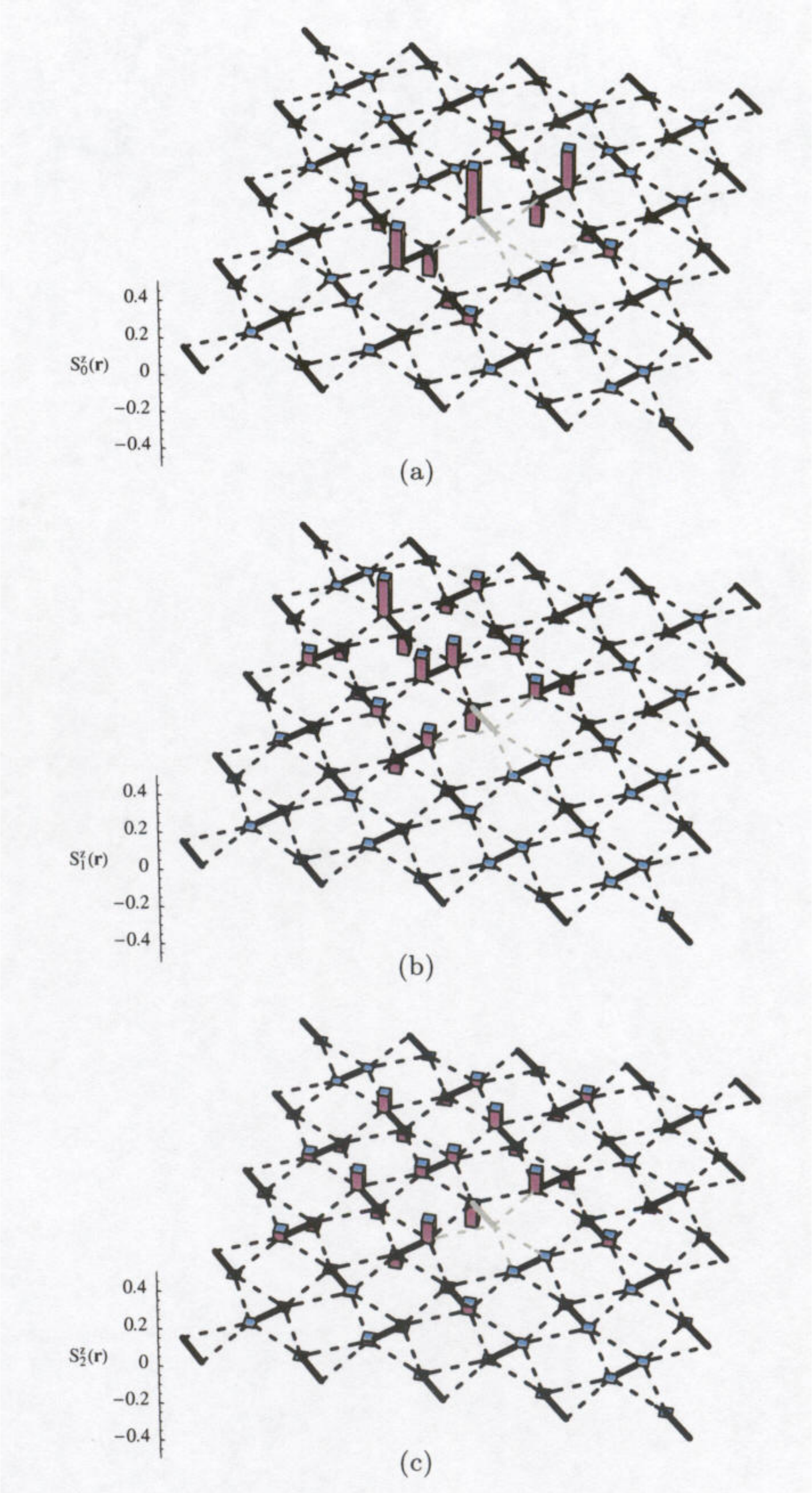
S. Haravifard, S. R. Dunsiger, S. El Shawish, B. D. Gaulin, H. A. Dabkowska, M. T. F. Telling, T. G. Perring, and J. Bonča, Phys. Rev. Lett. 97, 247206 (2006)

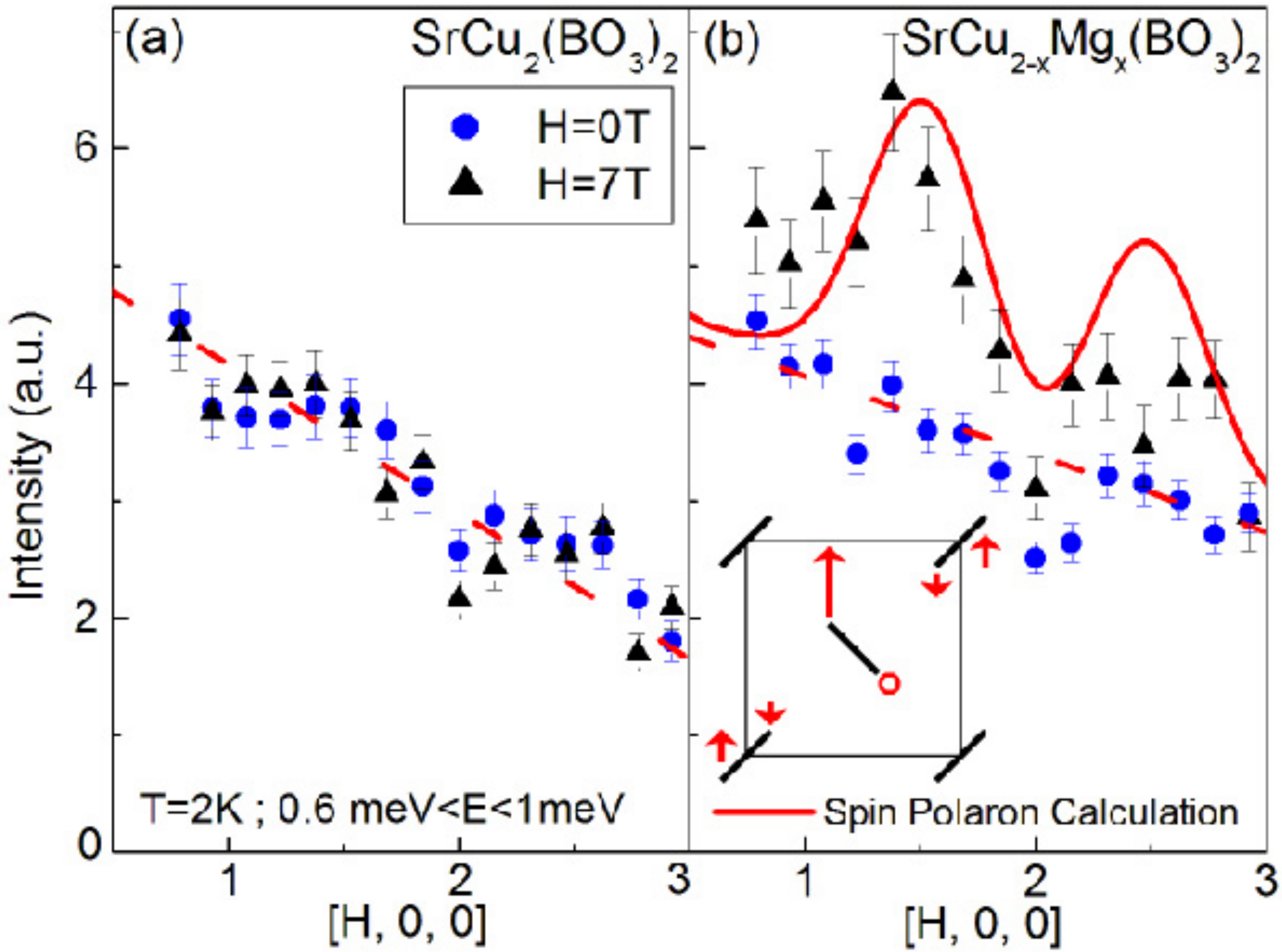
Variational Calculation
(El Shawish and Bonca, PRB 74, 174420, 2006)

Starting point for spin defect around impurity


(a) (b)

Starting point *not* an eigenfunction of *H*





(a)
$SrCu_2(BO_3)_2$
H=0T
H=7T
Intensity (a.u.)
0
2
4
6
1
2
3
[H, 0, 0]
T=2K ; 0.6 meV<E<1meV
(b)
$SrCu_{2-x}Mg_x(BO_3)_2$
1
2
3
[H, 0, 0]
Spin Polaron Calculation

Sara Haravivard

Sarah Dunsiger

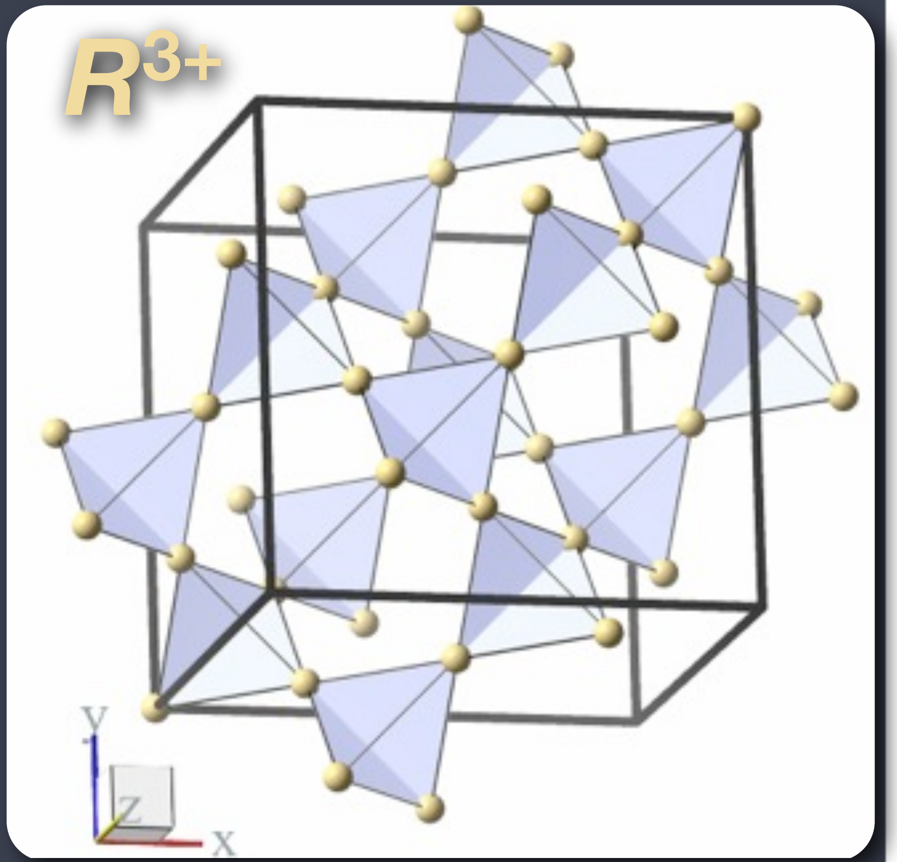
- *Renaissance in neutron science with figures of merit increases ~ 100*
  TOF techniques well suited to exotic magnetism as it measures very effectively across wide dynamic range in Q and E

- *Shastry-Sutherland singlet ground state in $SrCu_2(BO_3)_2$:*
  Frustration drives localized triplets + plateaus in triplet density with field

- *Quenched impurities give spin polarons:*
  extended S=1/2 states which can be measured with neutron scattering

Real Pyrochlores: Rare Earth Titanates

$$R_2\mathrm{Ti}_2\mathrm{O}_7$$



$$R = \begin{cases} \mathrm{Ho} \\ \mathrm{Dy} \\ \mathrm{Er} \\ \mathrm{Tb} \\ \mathrm{Gd} \\ \mathrm{Yb} \end{cases}$$

Ho, Dy → Spin Ice, Magnetic Monopole Excitations

Er → LRO, Quantum Critical Point

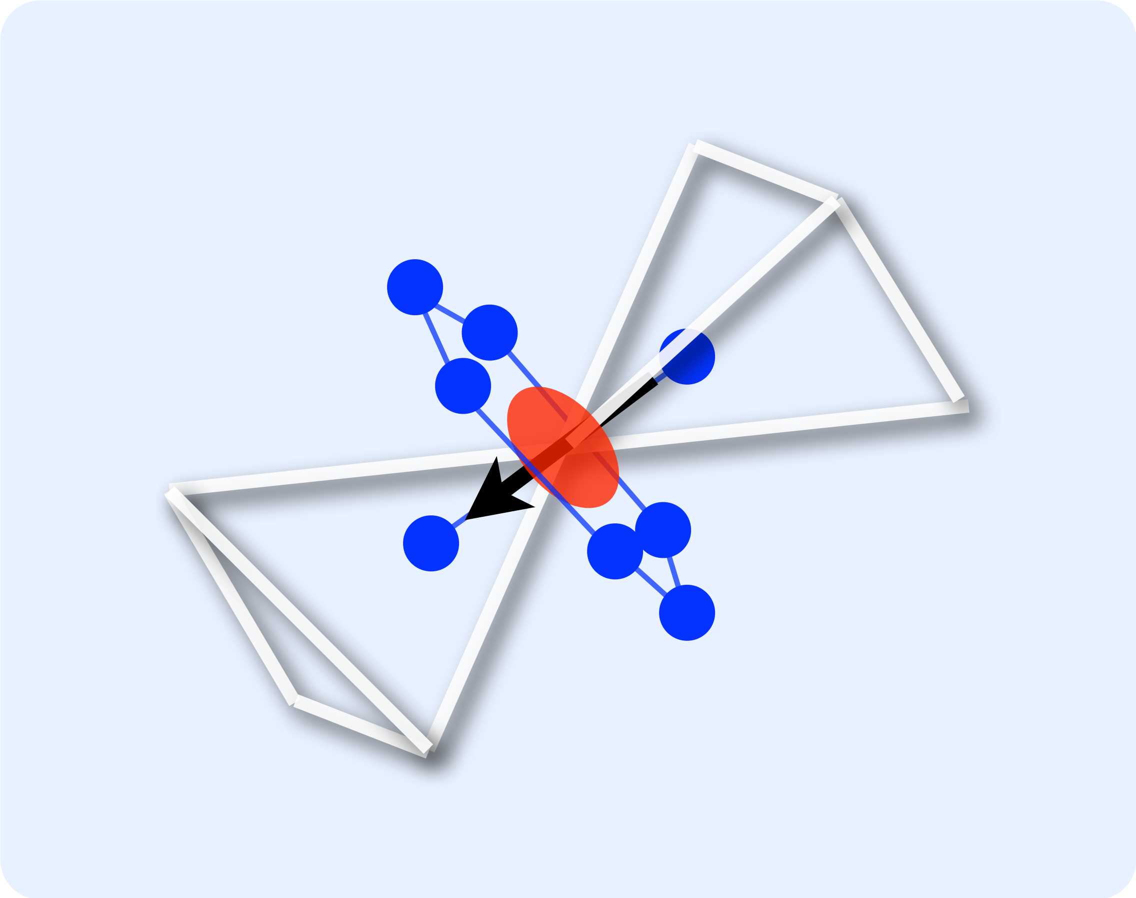
Tb → Spin Liquid

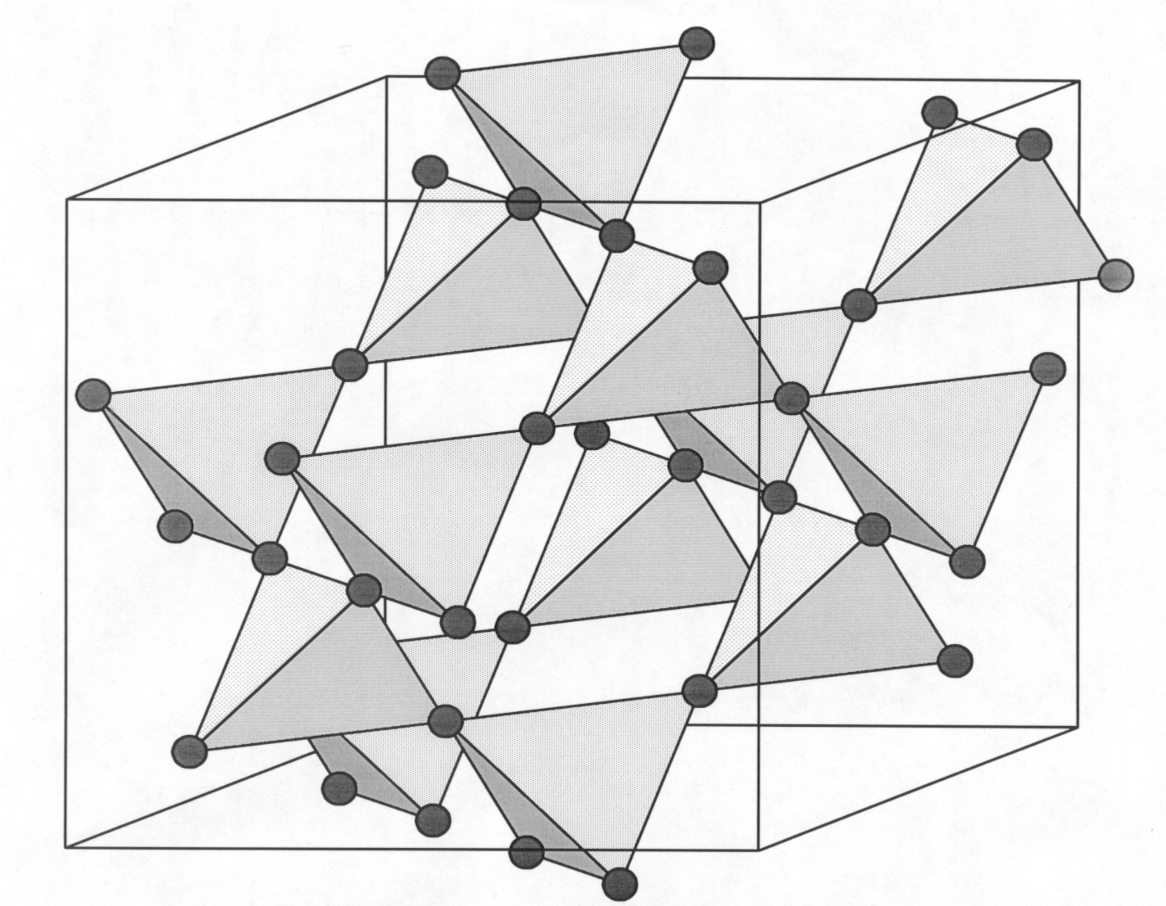
Gd → Partial Ordering

Yb → Low-Dimensional Correlations

Cubic Pyrochlores:

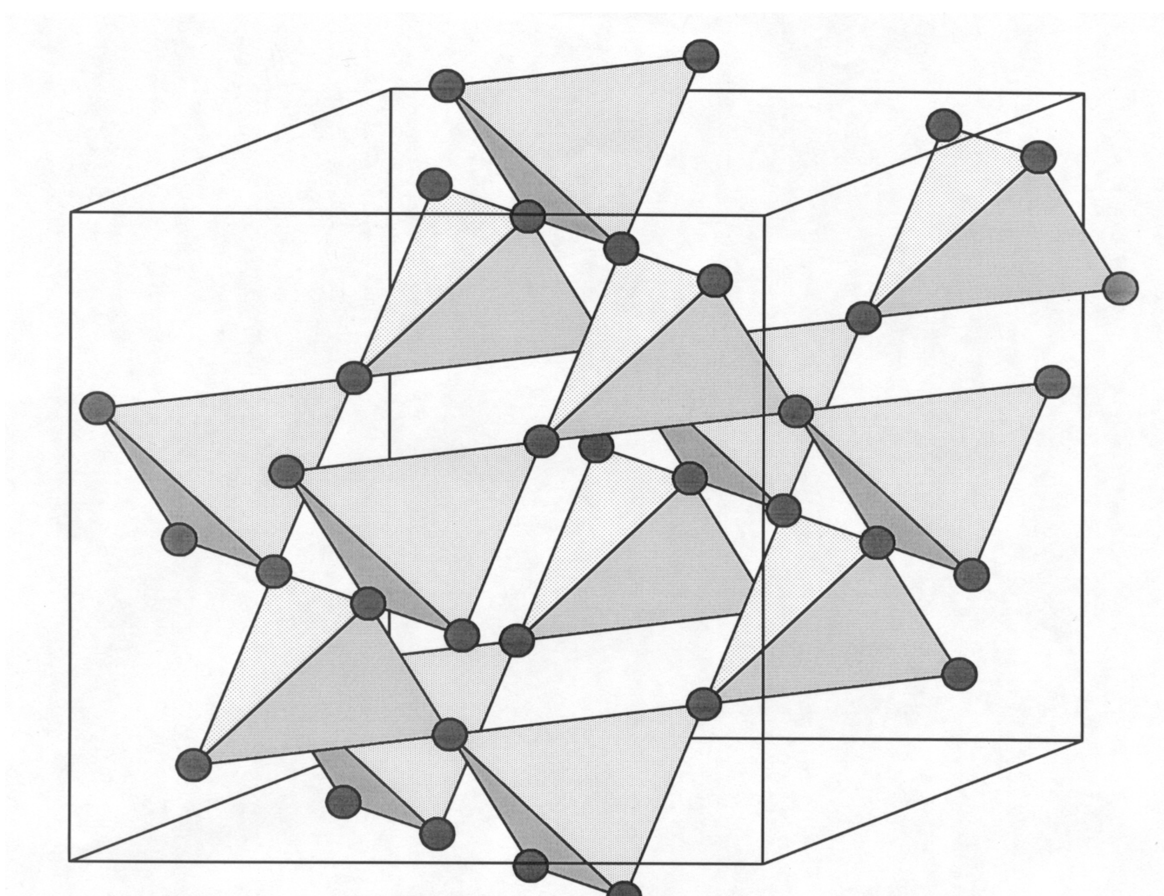
- Spins on a network of corner-sharing tetrahedra
- $A_2Ti_2O_7$
- A site is RE $^{3+}$ (many magnetic possibilities)

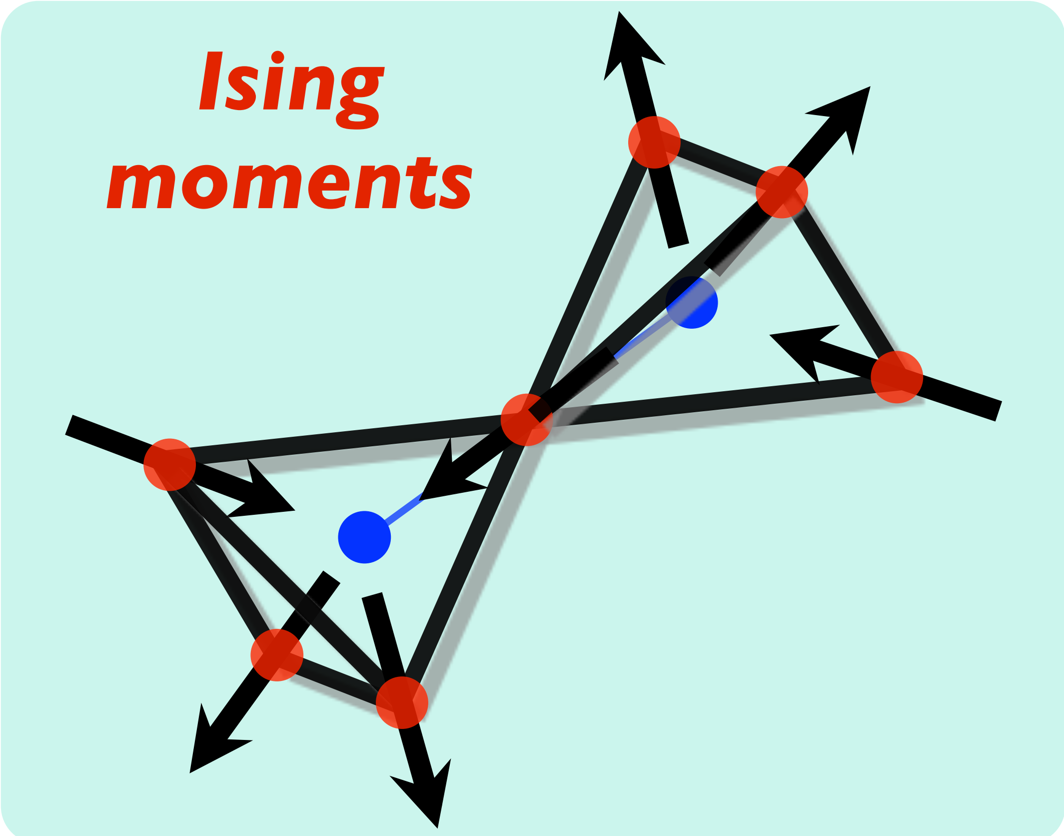


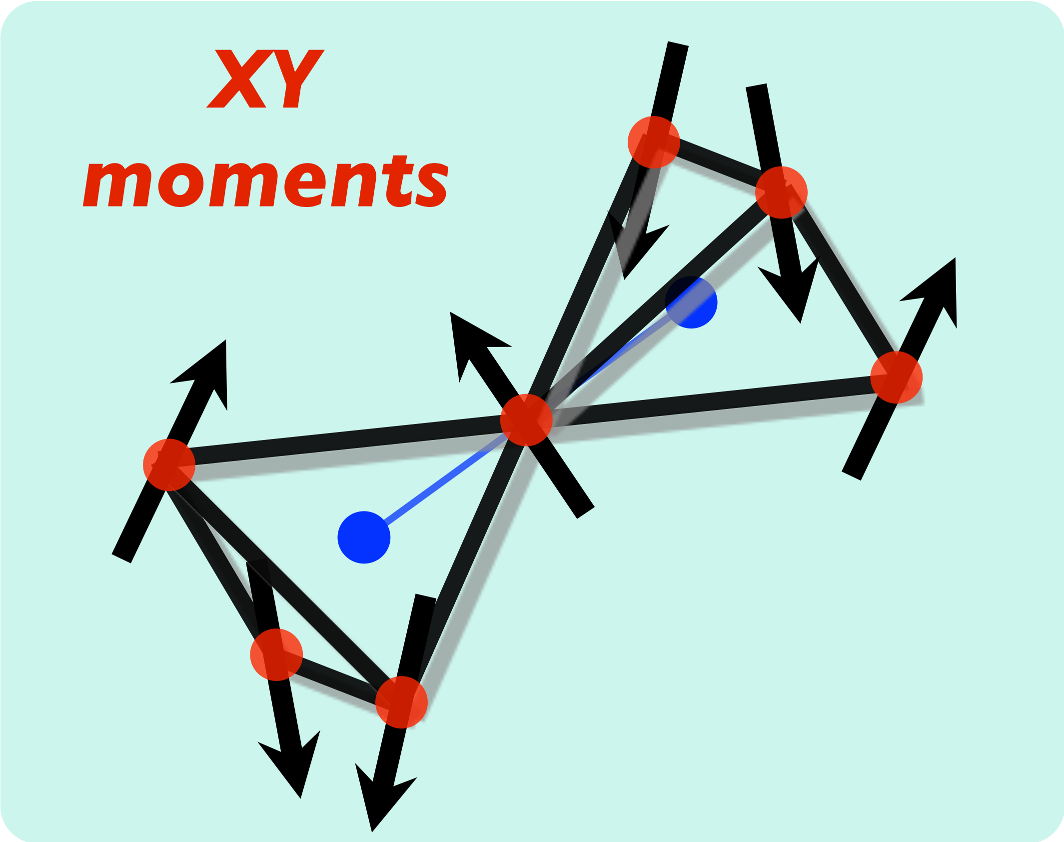


Cubic Pyrochlores:

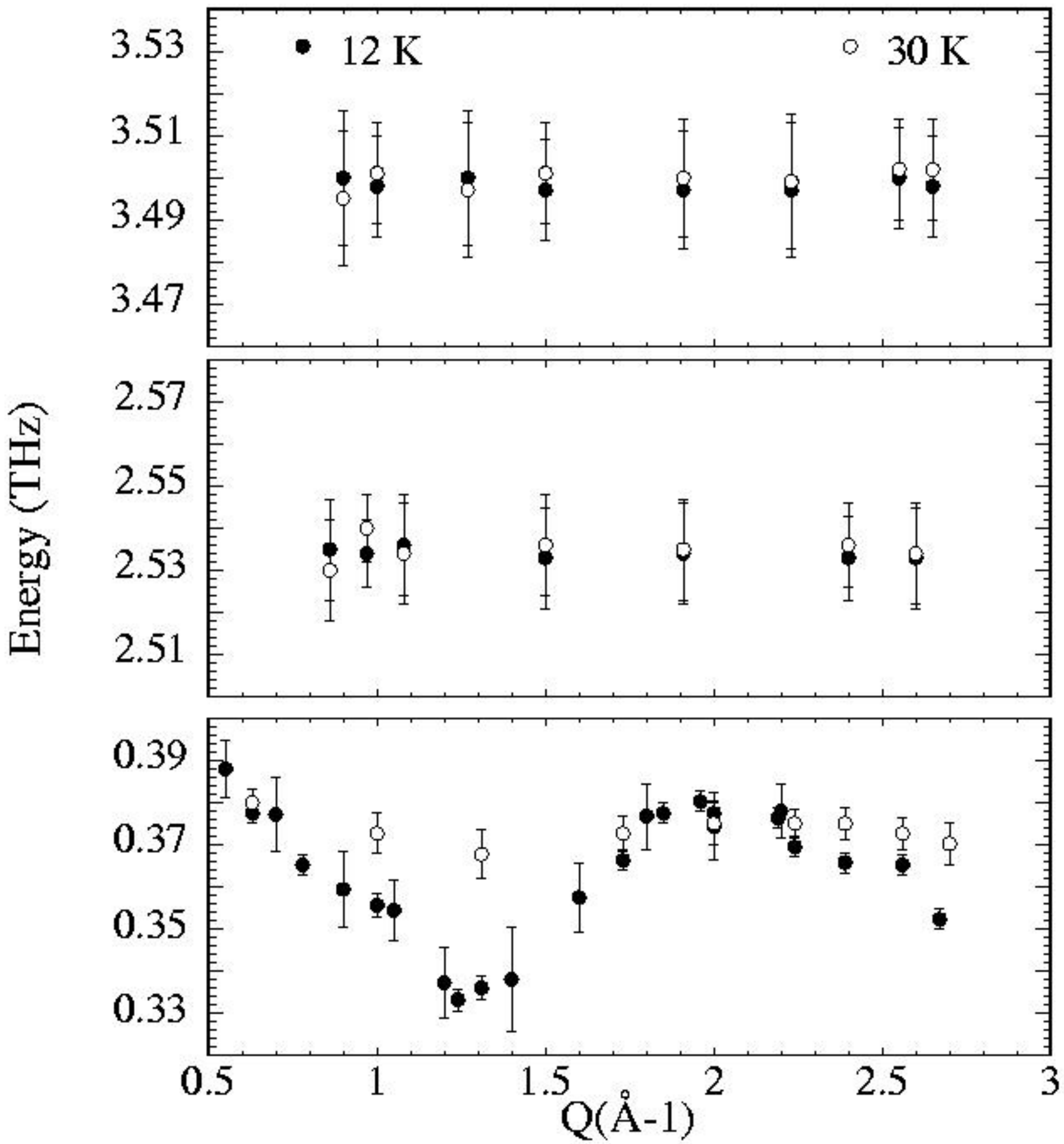
- *Spins on a network of corner-sharing tetrahedra*
- $A_2Ti_2O_7$
- *A site is RE $^{3+}$ (many magnetic possibilities)*

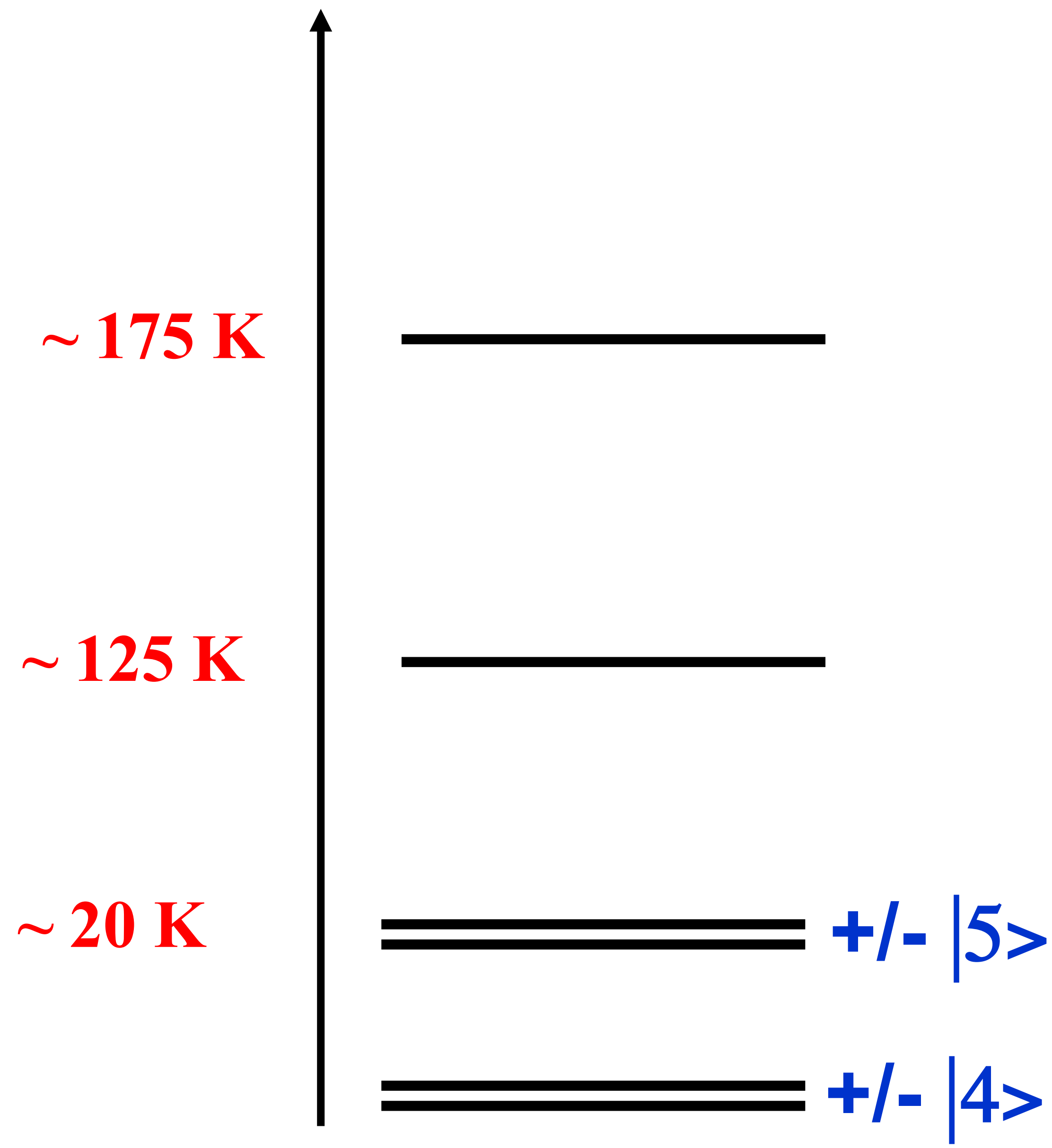




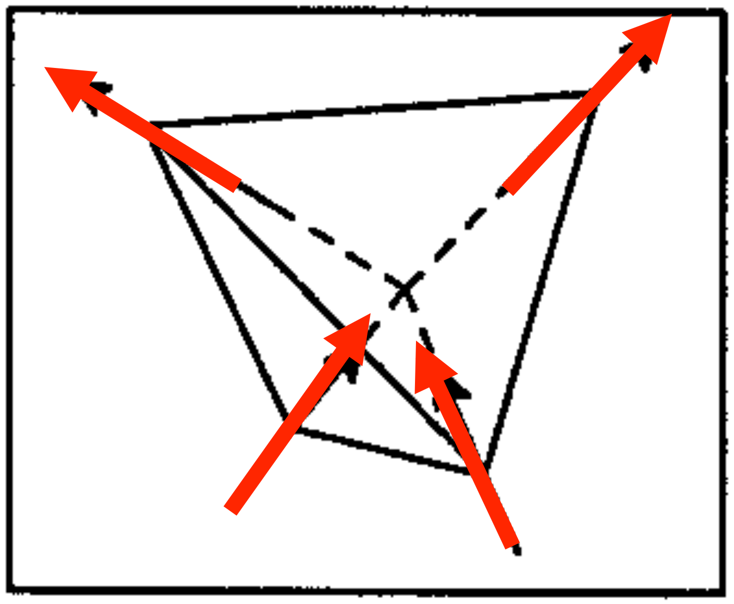


Inelastic neutron scattering on polycrystalline $Tb_2Ti_2O_7$



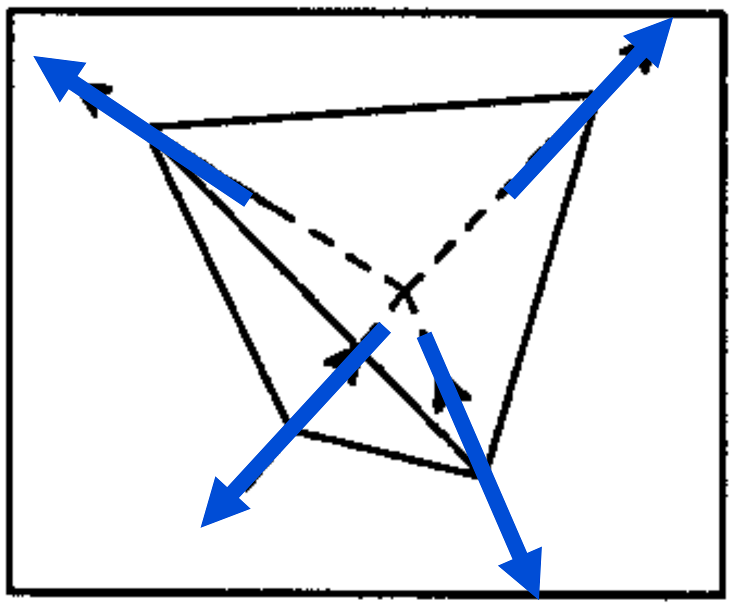


(Δ : $Ho_2Ti_2O_7$ ~ 240 K ; $Dy_2Ti_2O_7$ ~ 380 K)



Rare Earth moments:

Strong [111] anisotropy



Ferromagnetic exchange:

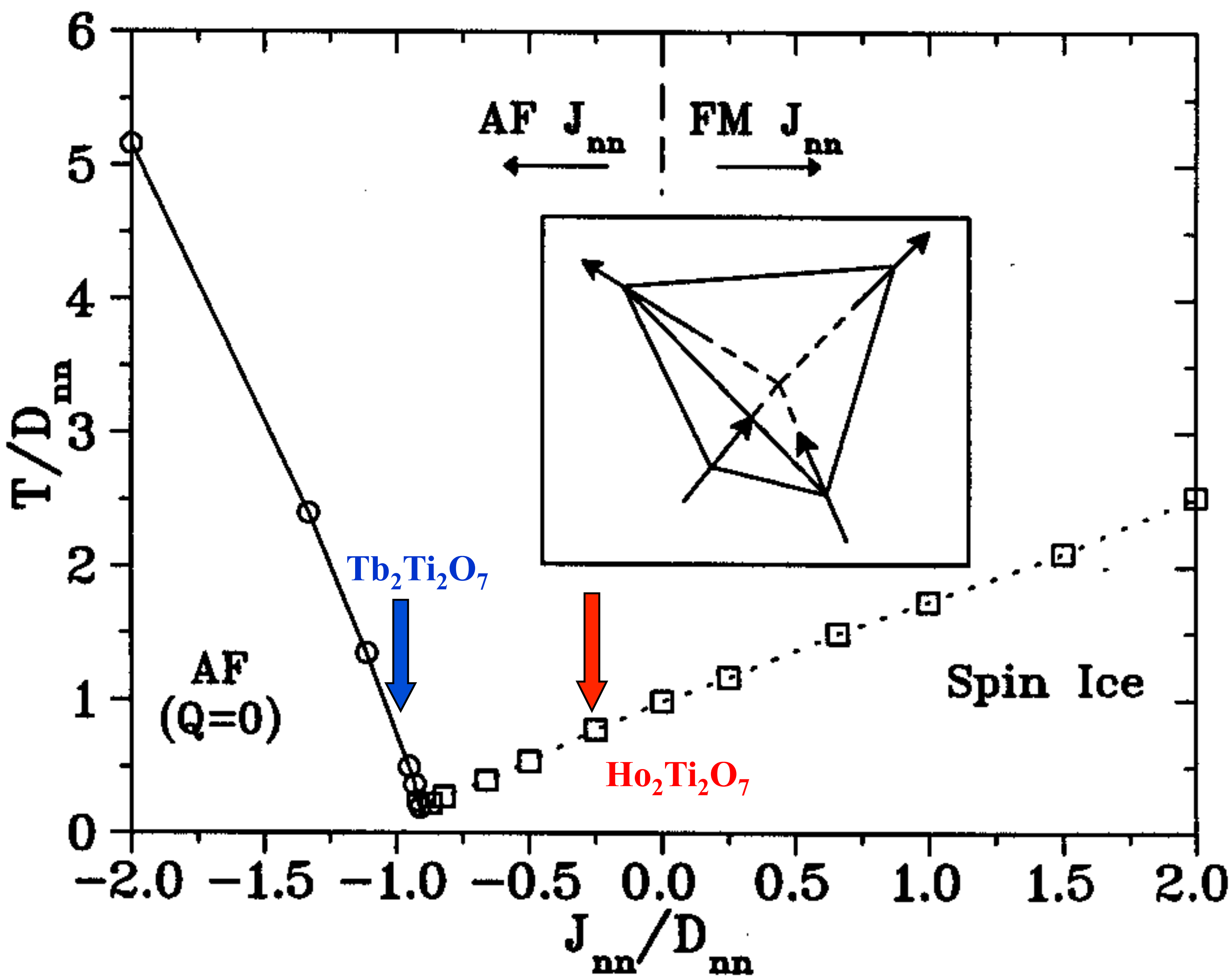
“Spin Ice” : 2 in 2 out

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

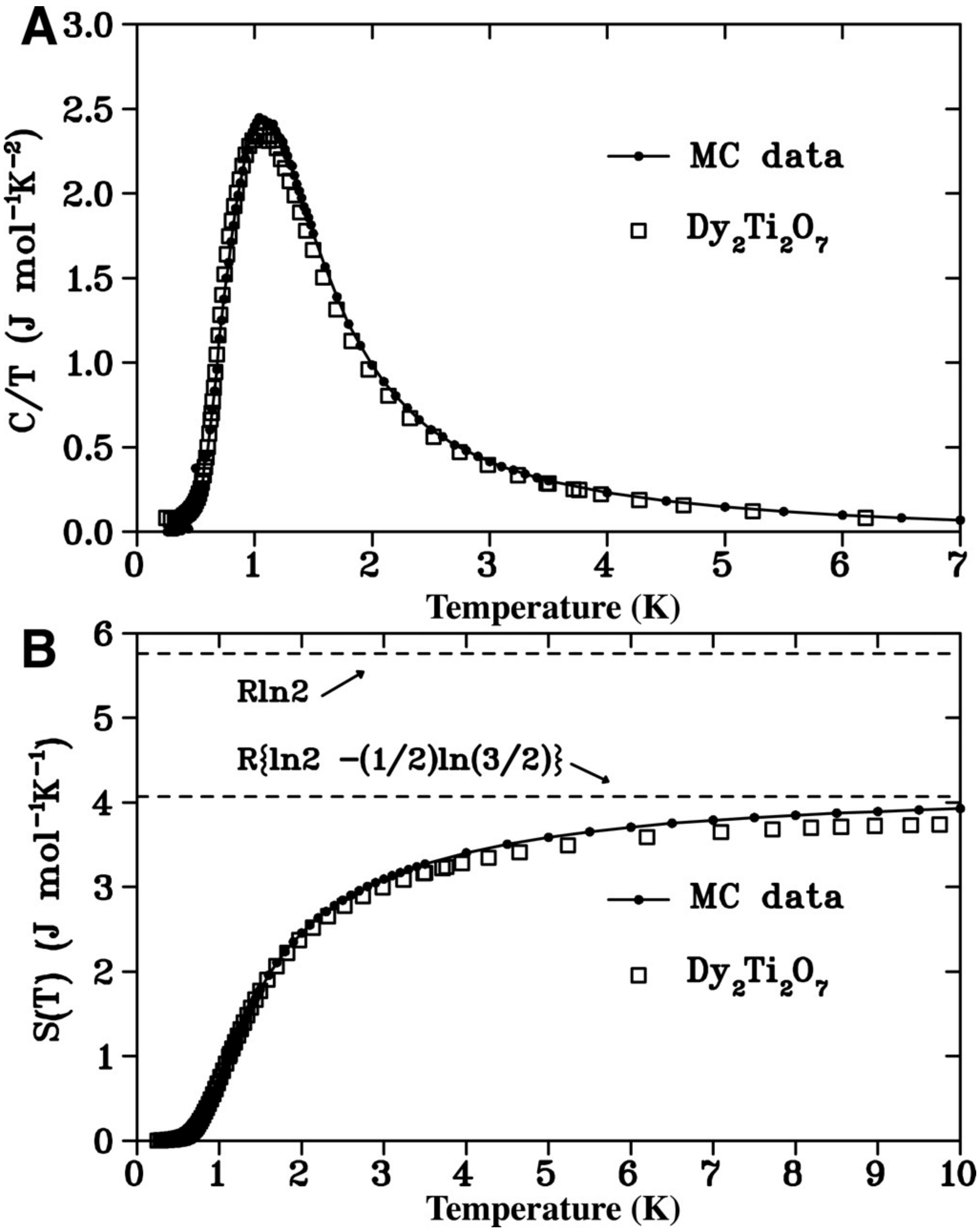
Antiferromagnetic exchange:

All in - All out

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



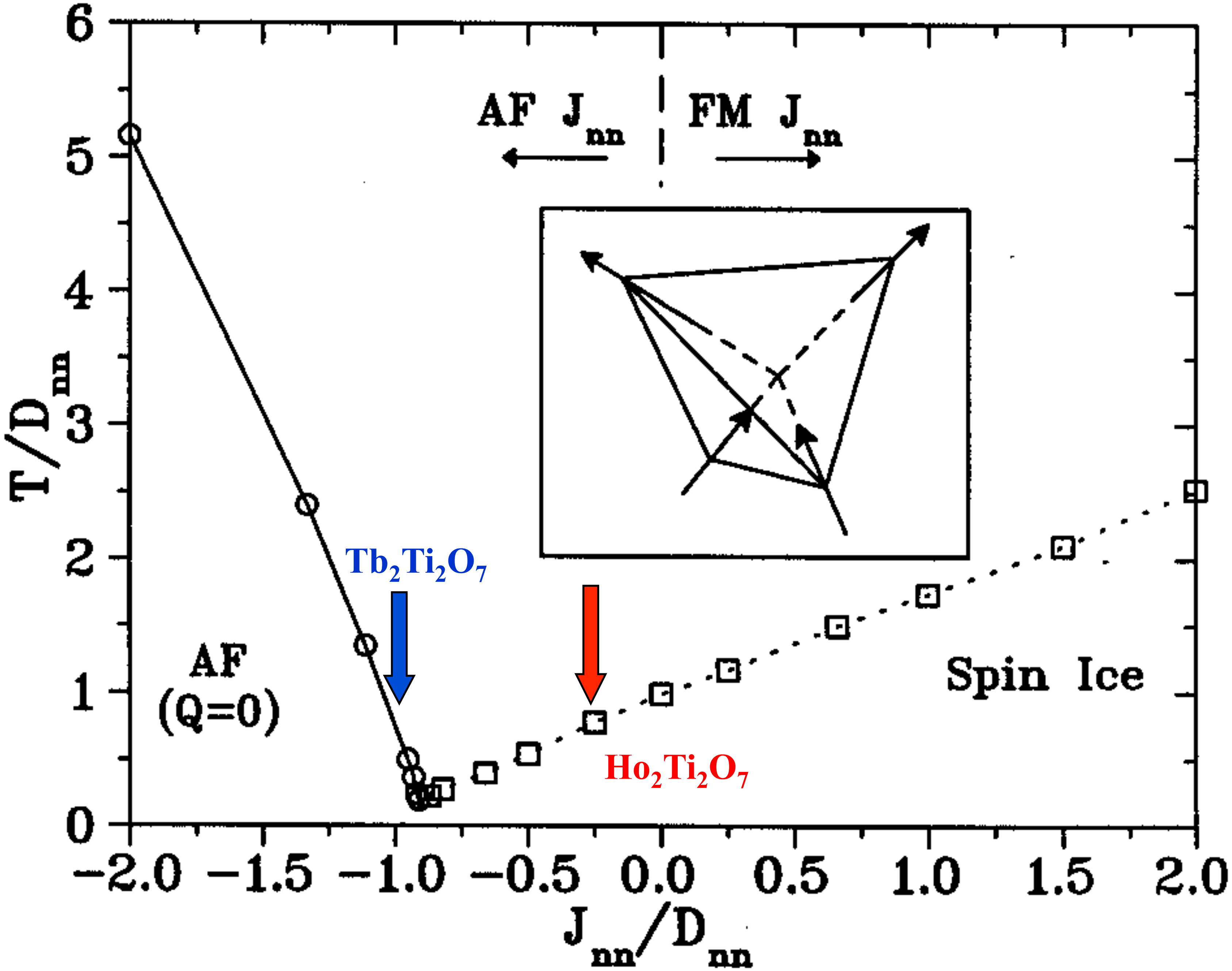
B.C. den Hertog and M.J.P. Gingras, PRL, 84, 3430, 2000.



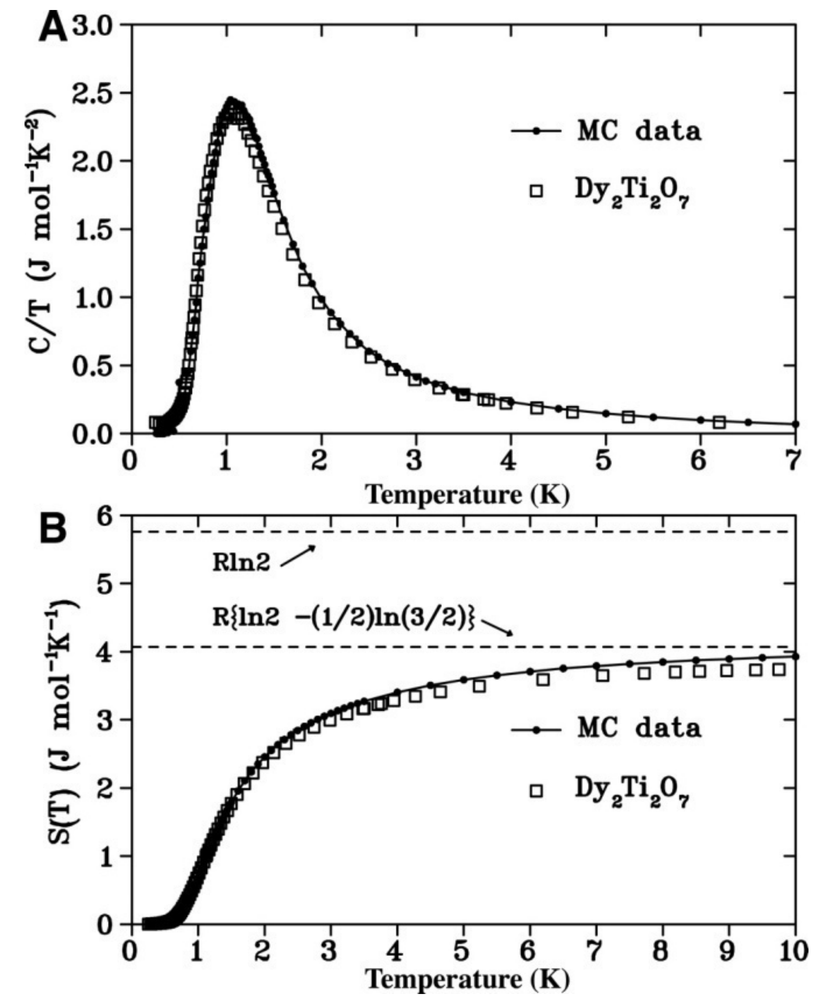
Spin Ice Ground State indicated by Shottky-like anomaly at $T_{SI} \sim 2$ K in both $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$

Missing entropy is the same as for proton disorder in solid H_2O, hence “Spin Ice”

A.P. Ramirez et al, Nature, 399, 333, 1999

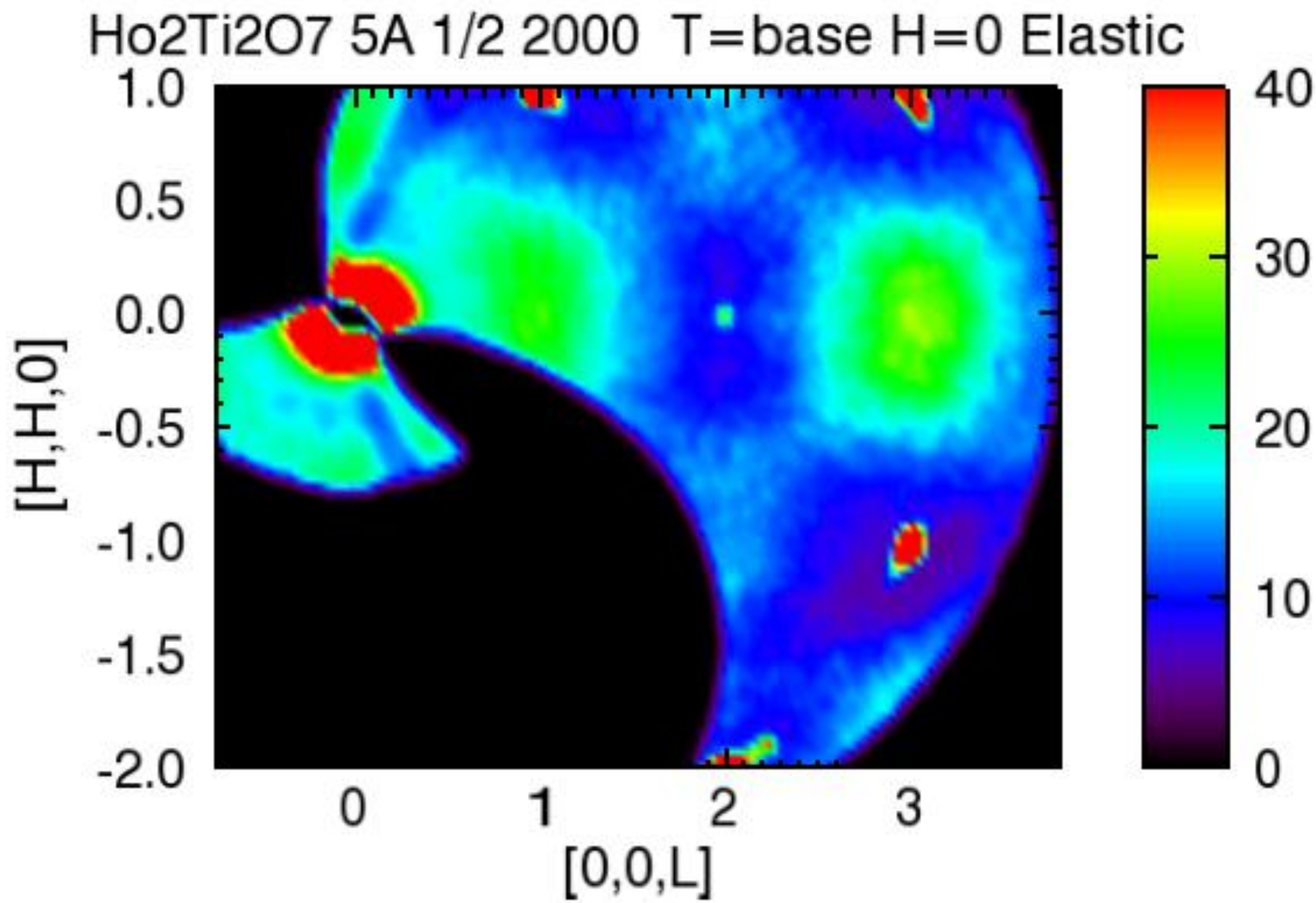


T/D_{nn}
J_{nn}/D_{nn}
AF J_{nn}
FM J_{nn}
$Tb_2Ti_2O_7$
$Ho_2Ti_2O_7$
AF
(Q=0)
Spin Ice



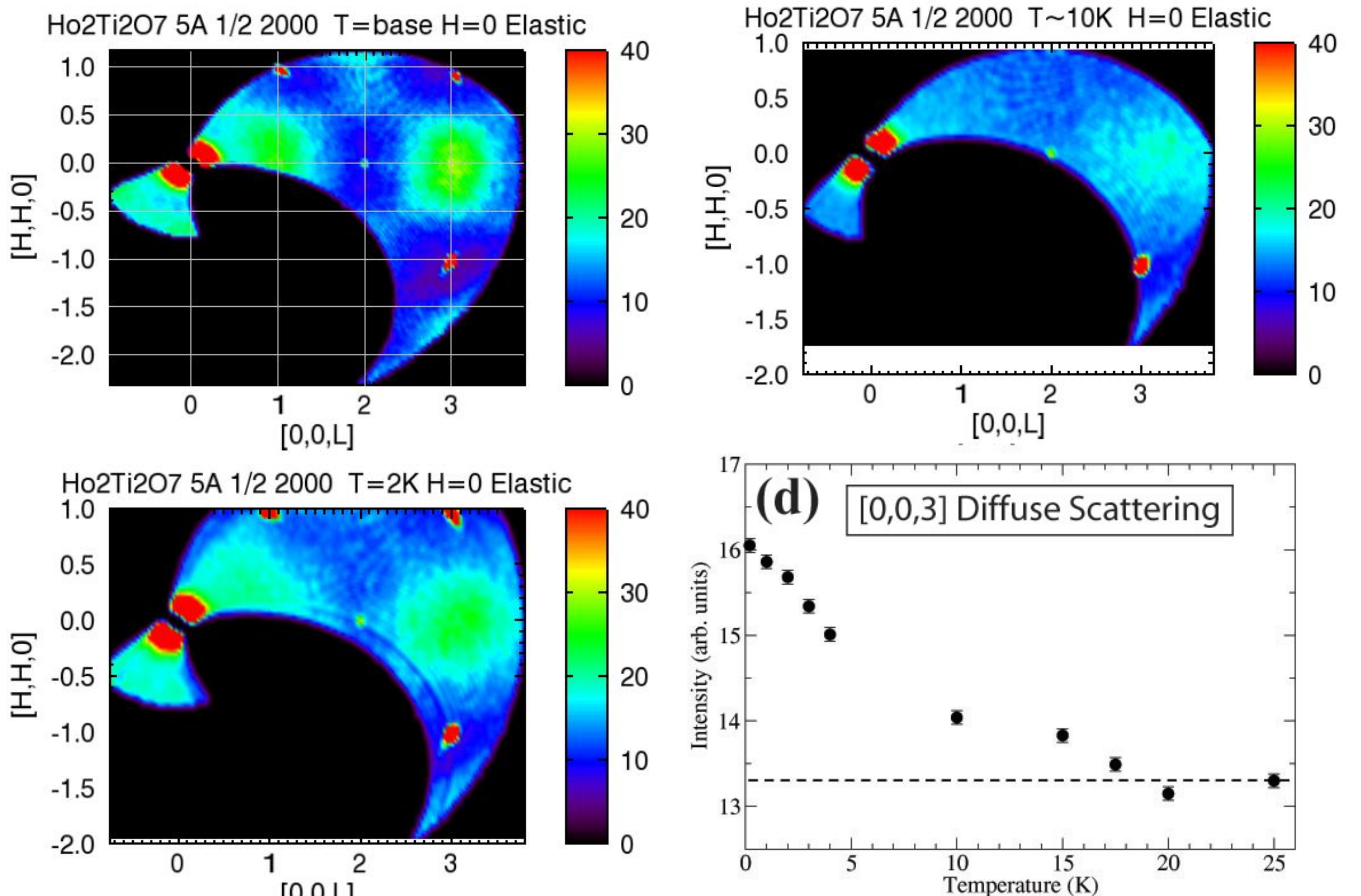
A
C/T (J mol^{-1}K^{-2})
Temperature (K)
MC data
$Dy_2Ti_2O_7$
B
S(T) (J mol^{-1}K^{-1})
Rln2
R{ln2 −(1/2)ln(3/2)}
Temperature (K)
MC data
$Dy_2Ti_2O_7$

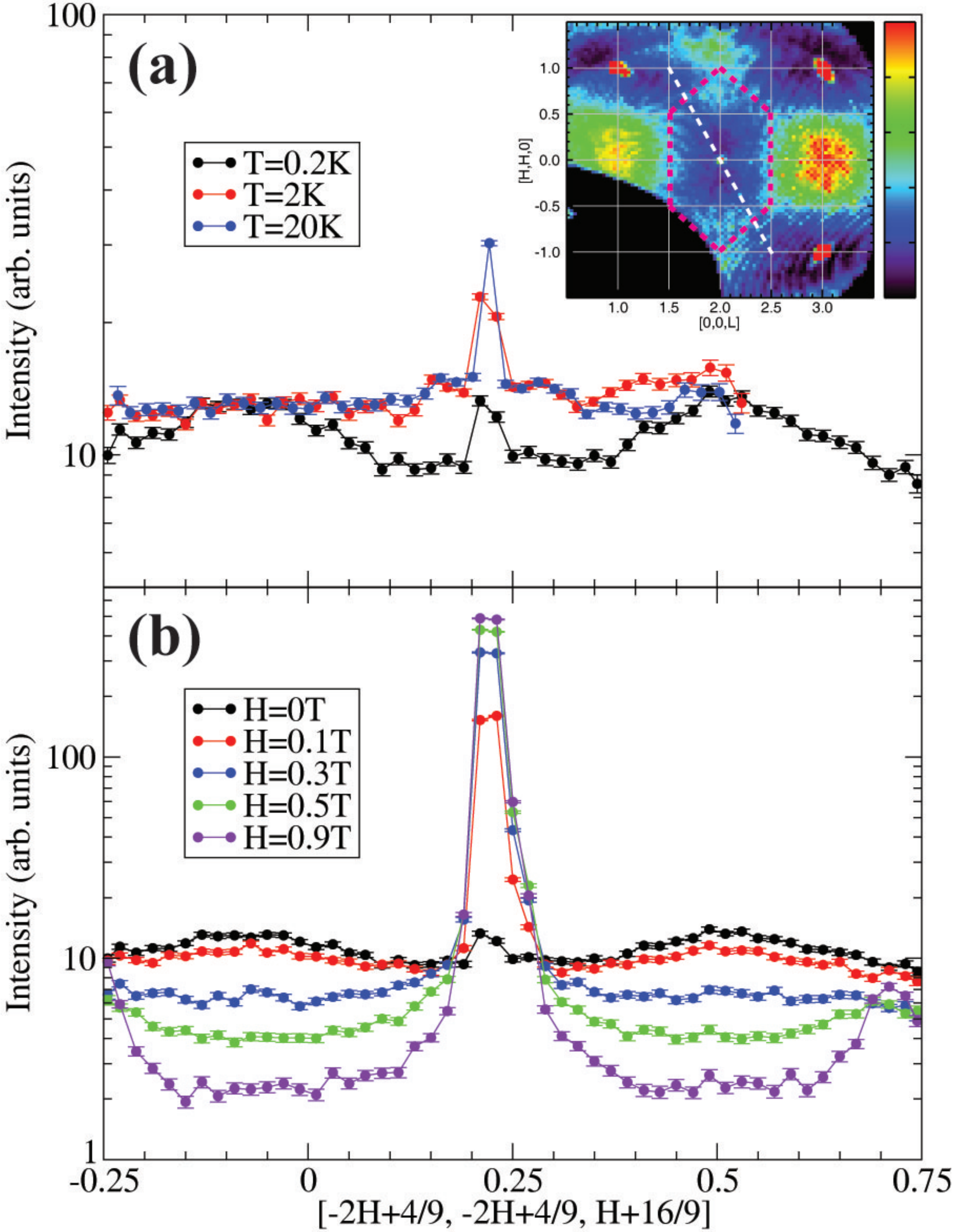
Spin Ice Ground State in $Ho_2Ti_2O_7$



J. P. Clancy, J. P. C. Ruff, S. R. Dunsiger, Y. Zhao, H. A. Dabkowska, J. S. Gardner, Y. Qiu, J. R. D. Copley, T. Jenkins, and B. D. Gaulin, Phys. Rev. B 79, 014408 (2009)

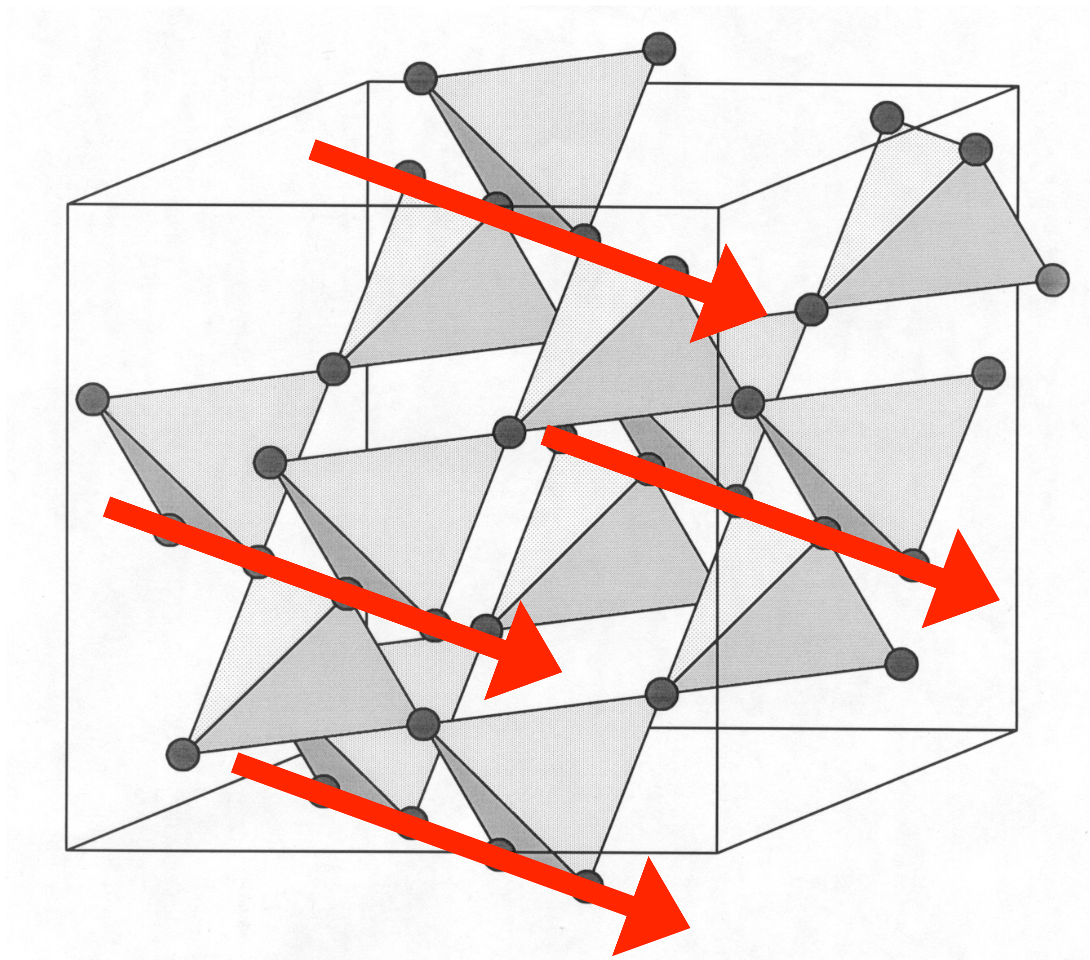
Diffuse Scattering Evolves with T on scale of ~ 17 K





Spin Ice correlations due to enforced "ice rules" (2 spins in – 2 spins out / tetrahedron) gives rise to "Zone Boundary Scattering"

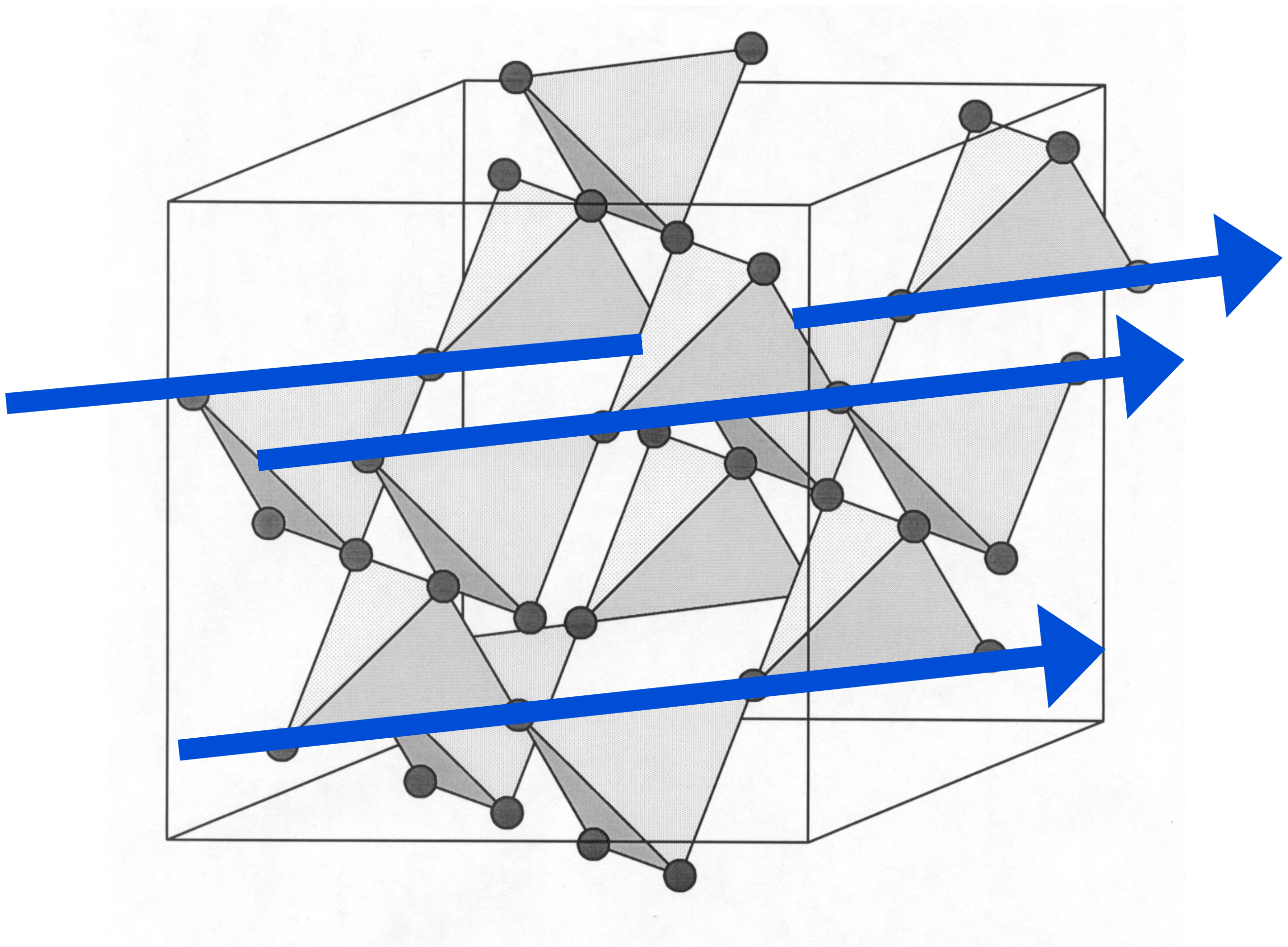
Evolve on scale of $T_{SI} \sim 2$ K



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

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

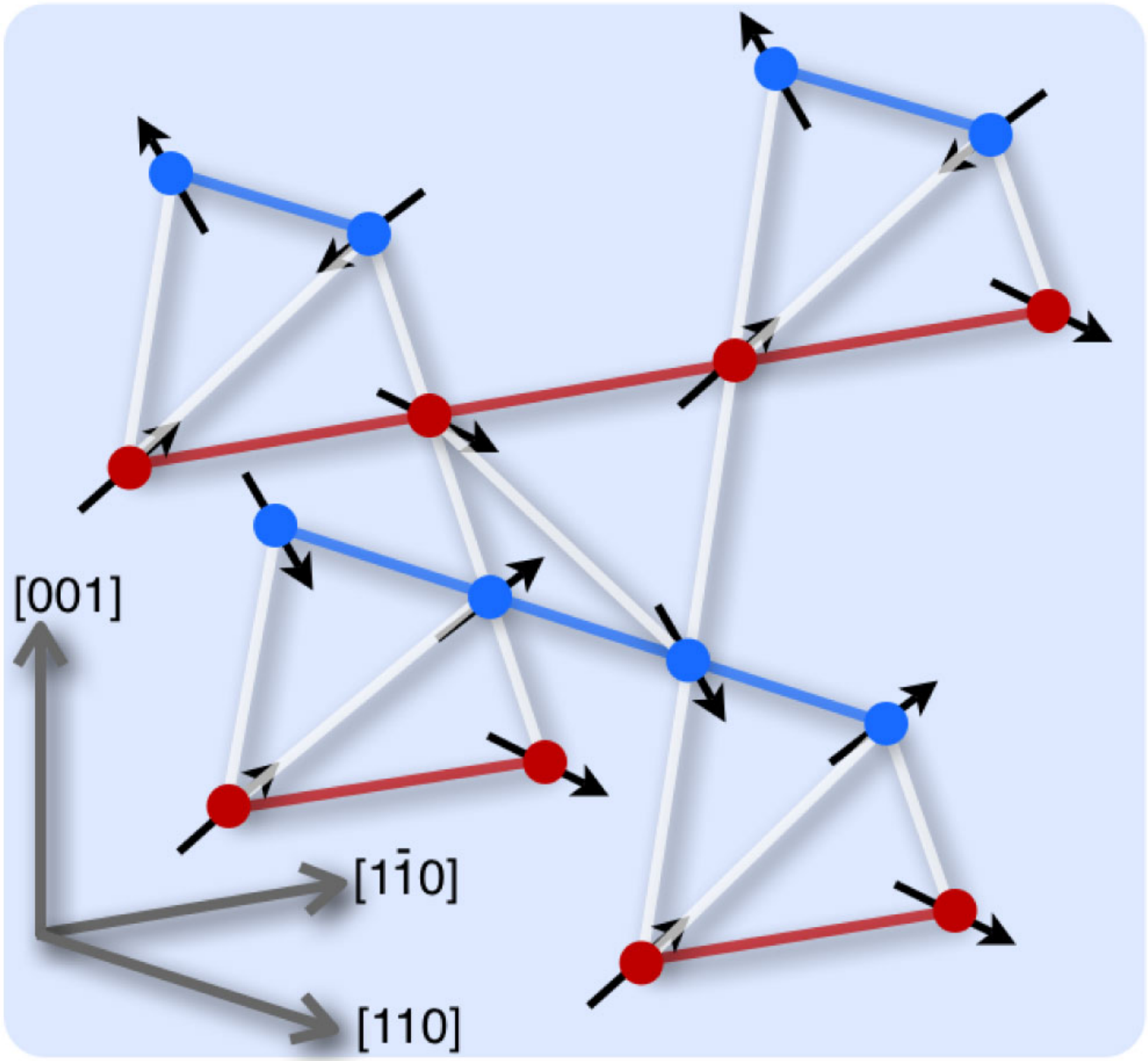
Perpendicular β-[110] chains



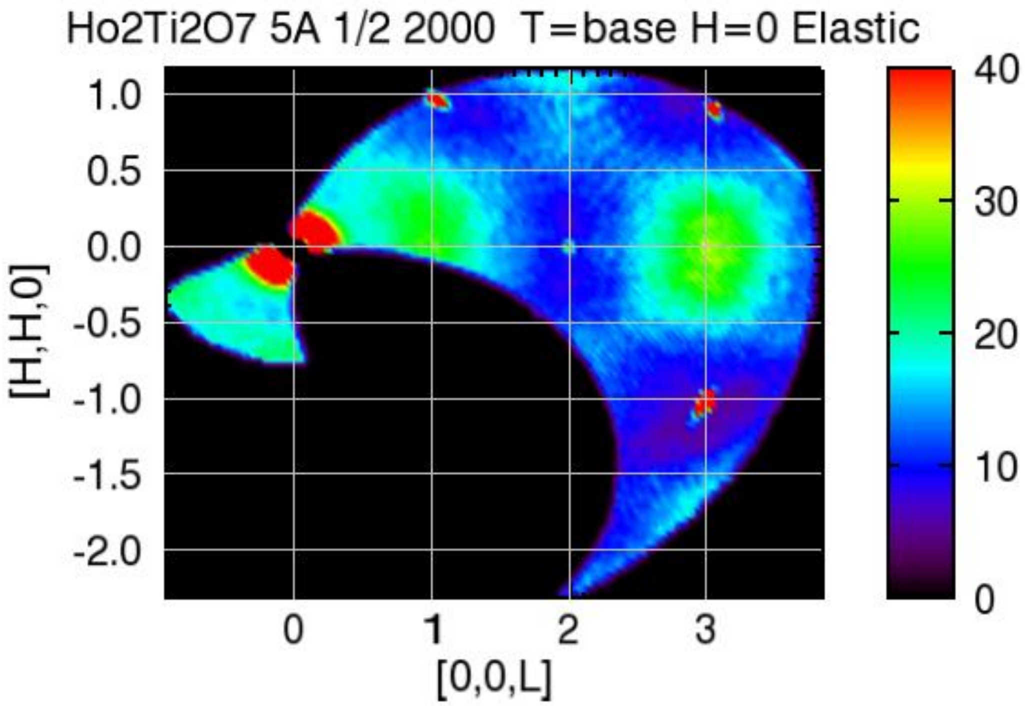
Application of weak [1-10] magnetic field breaks system up into α and β chains.

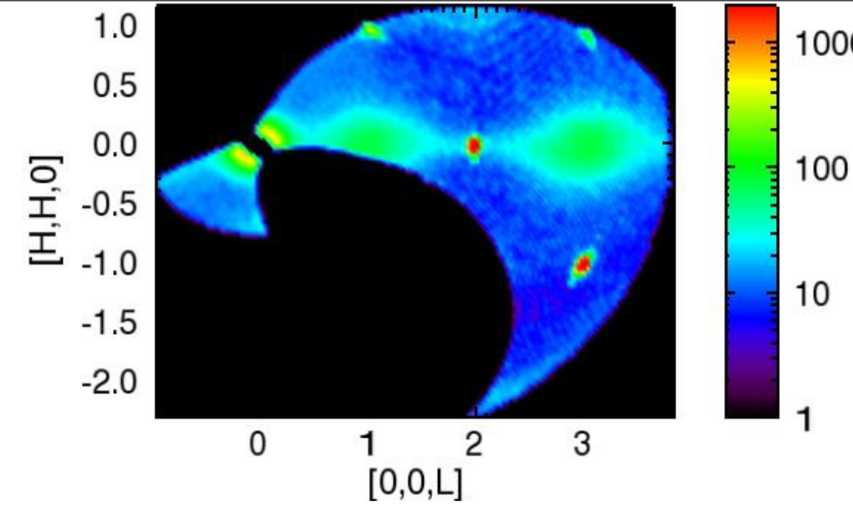
Polarizable α-[1-10] chains (parallel to field)

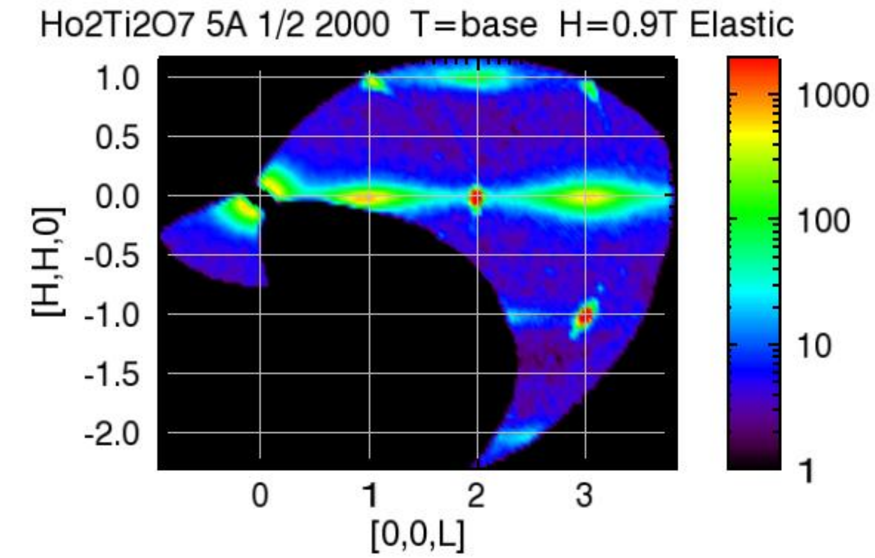
Perpendicular β-[110] chains



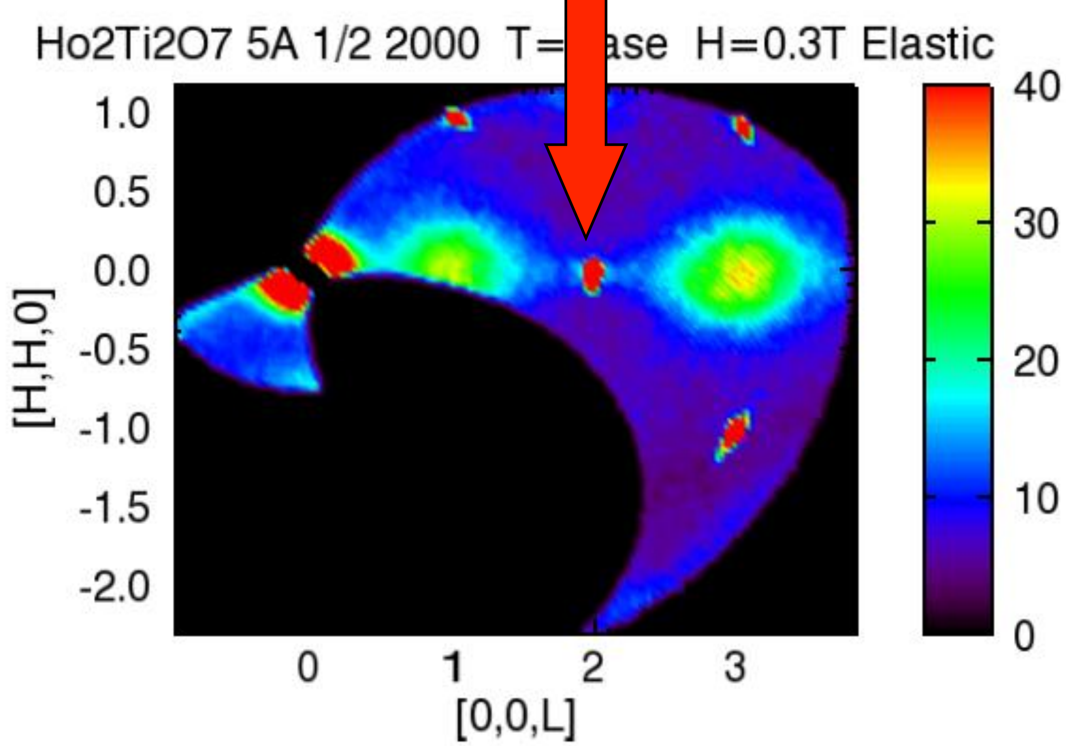
[1-10] Magnetic field should decompose pyrochlore lattice into polarized α chains (red chains) and decoupled qausi-1D β chains (blue chains).



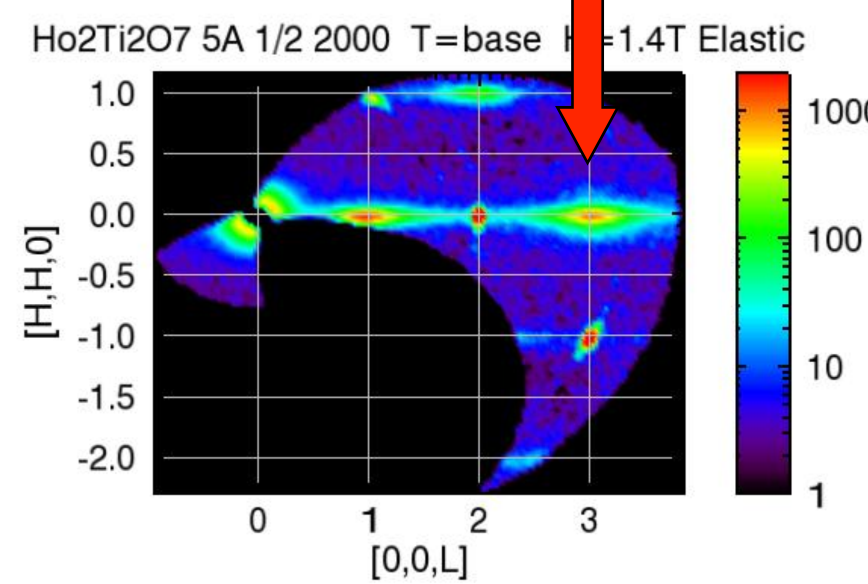




Ferro-ordering of α-chains

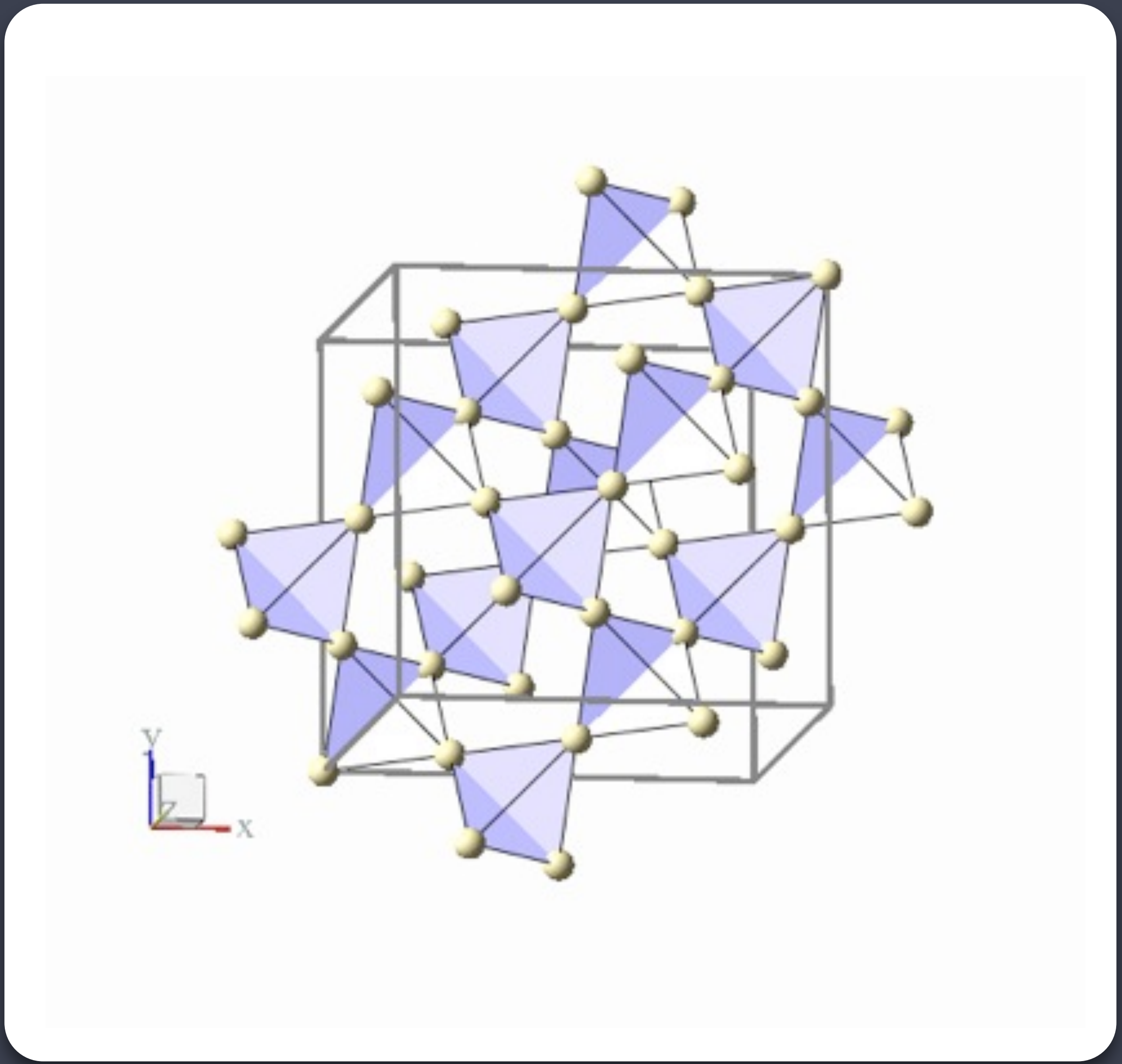


1D-correlations along β-chains



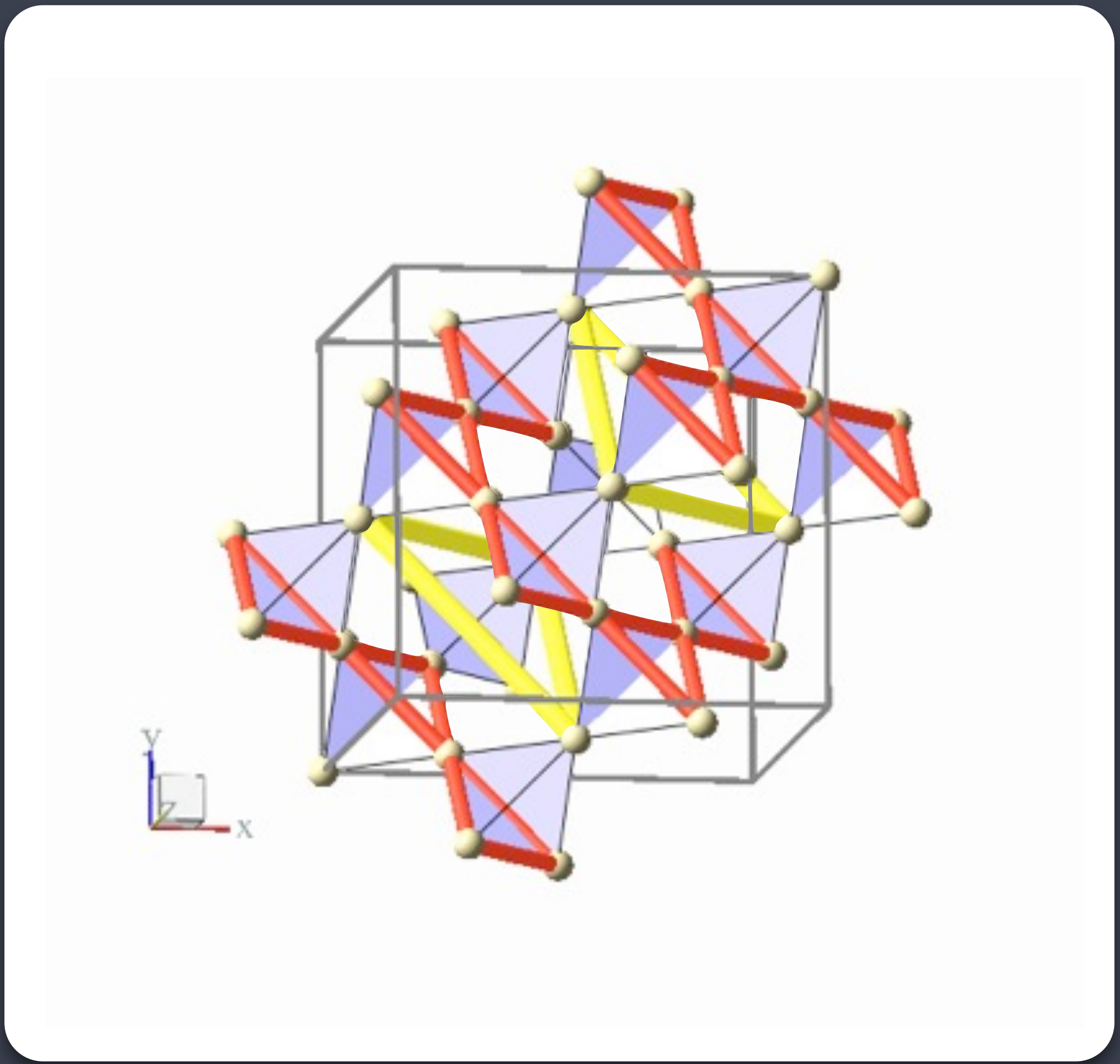
J. P. Clancy, J. P. C. Ruff, S. R. Dunsiger, Y. Zhao, H. A. Dabkowska, J. S. Gardner, Y. Qiu, J. R. D. Copley, T. Jenkins, and B. D. Gaulin, Phys. Rev. B 79, 014408 (2009)

Geometric Frustration in 3D: The Pyrochlore Lattice

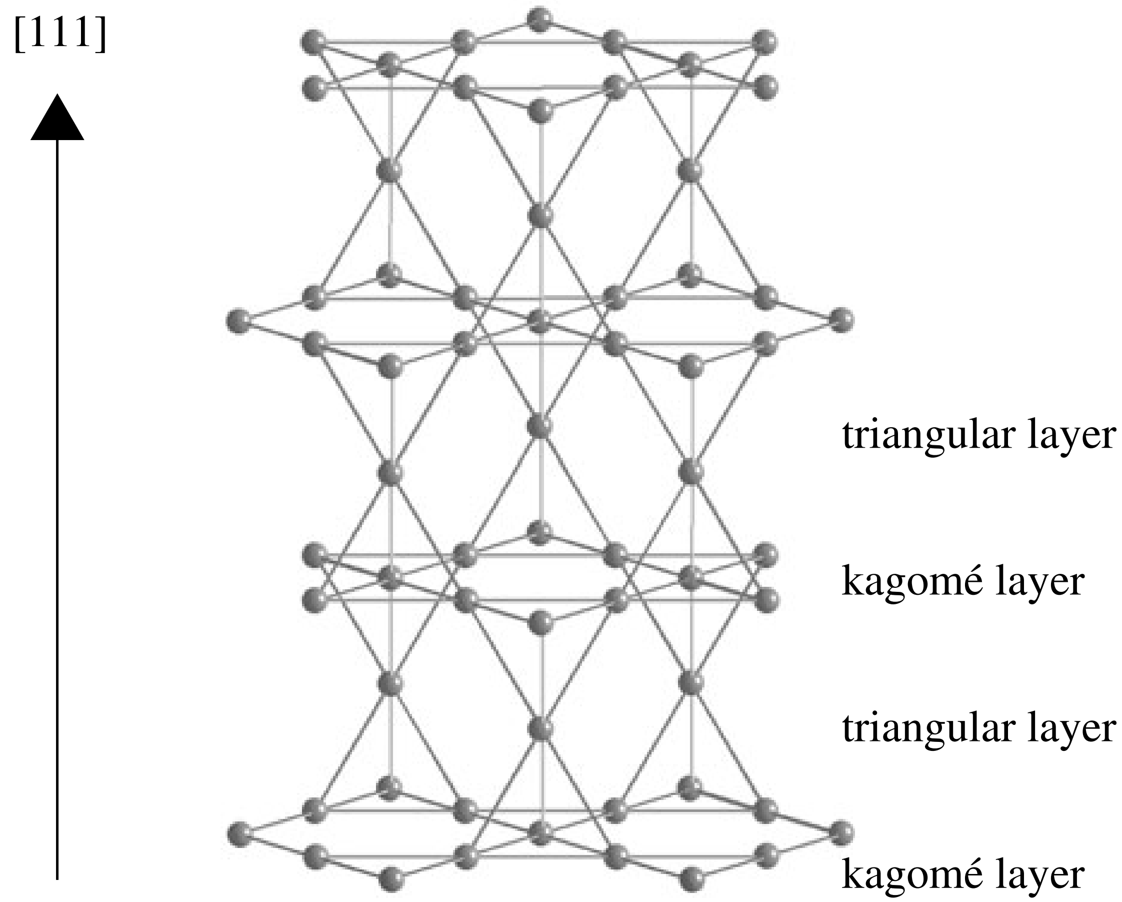


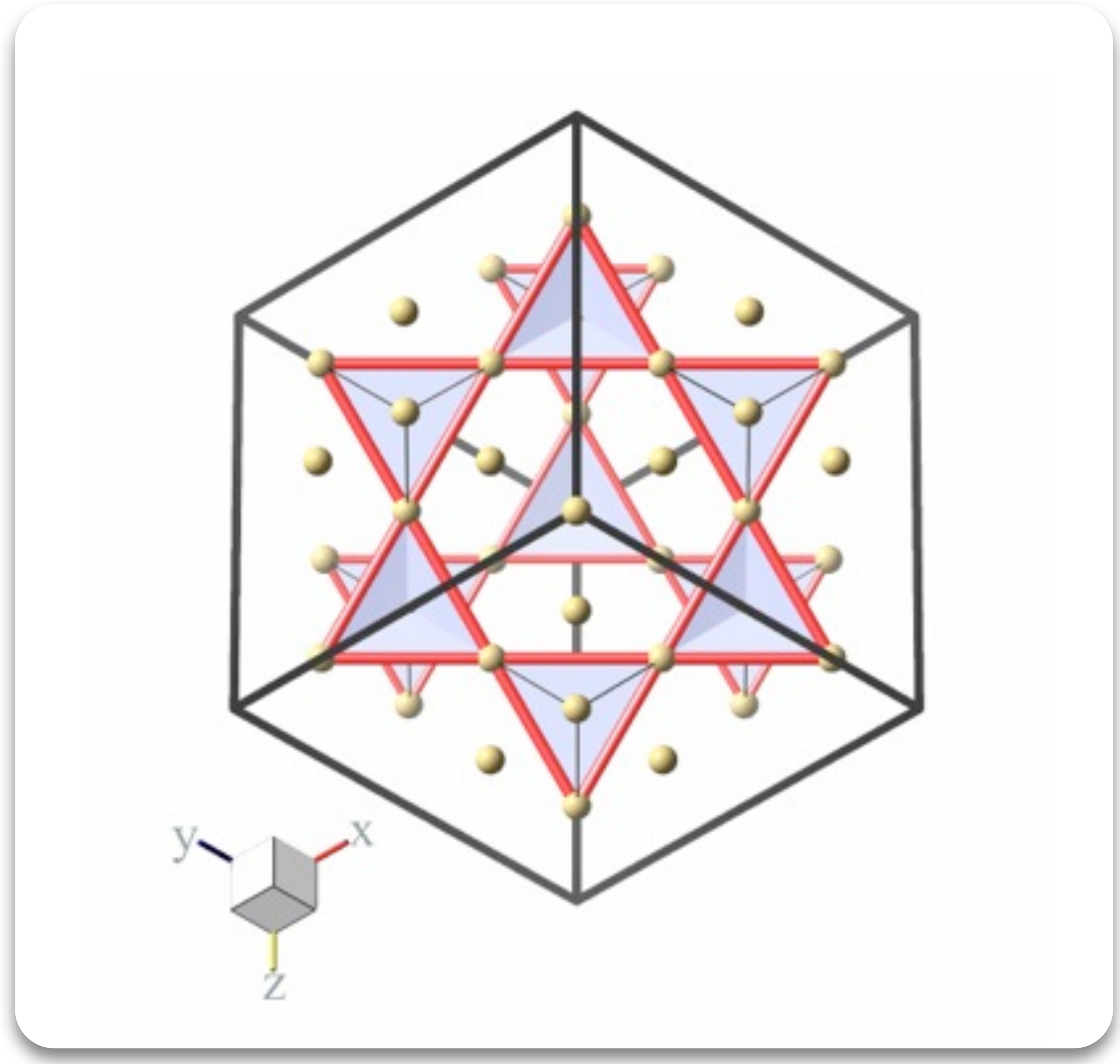
Corner-Sharing Tetrahedra

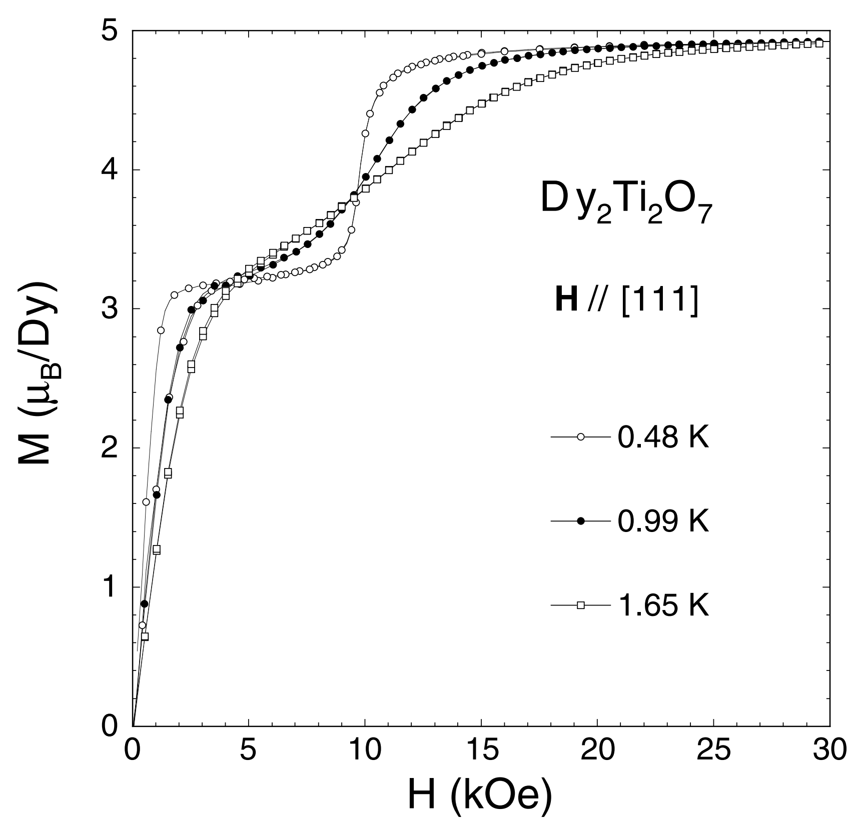
Geometric Frustration in 3D: The Pyrochlore Lattice

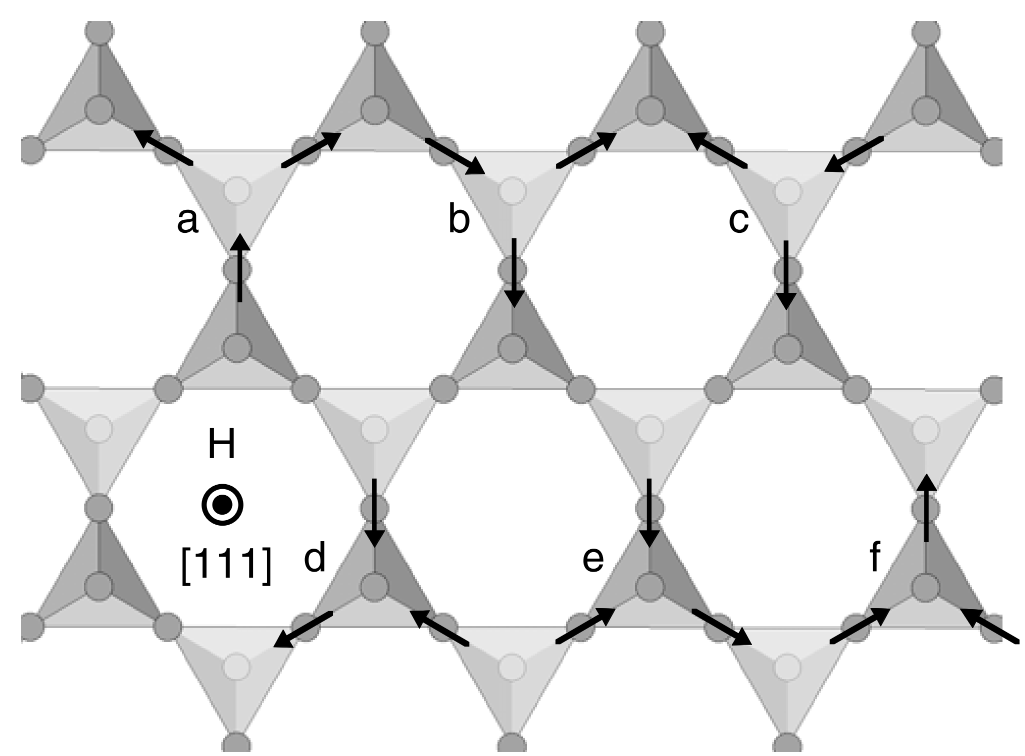


Corner-Sharing Tetrahedra

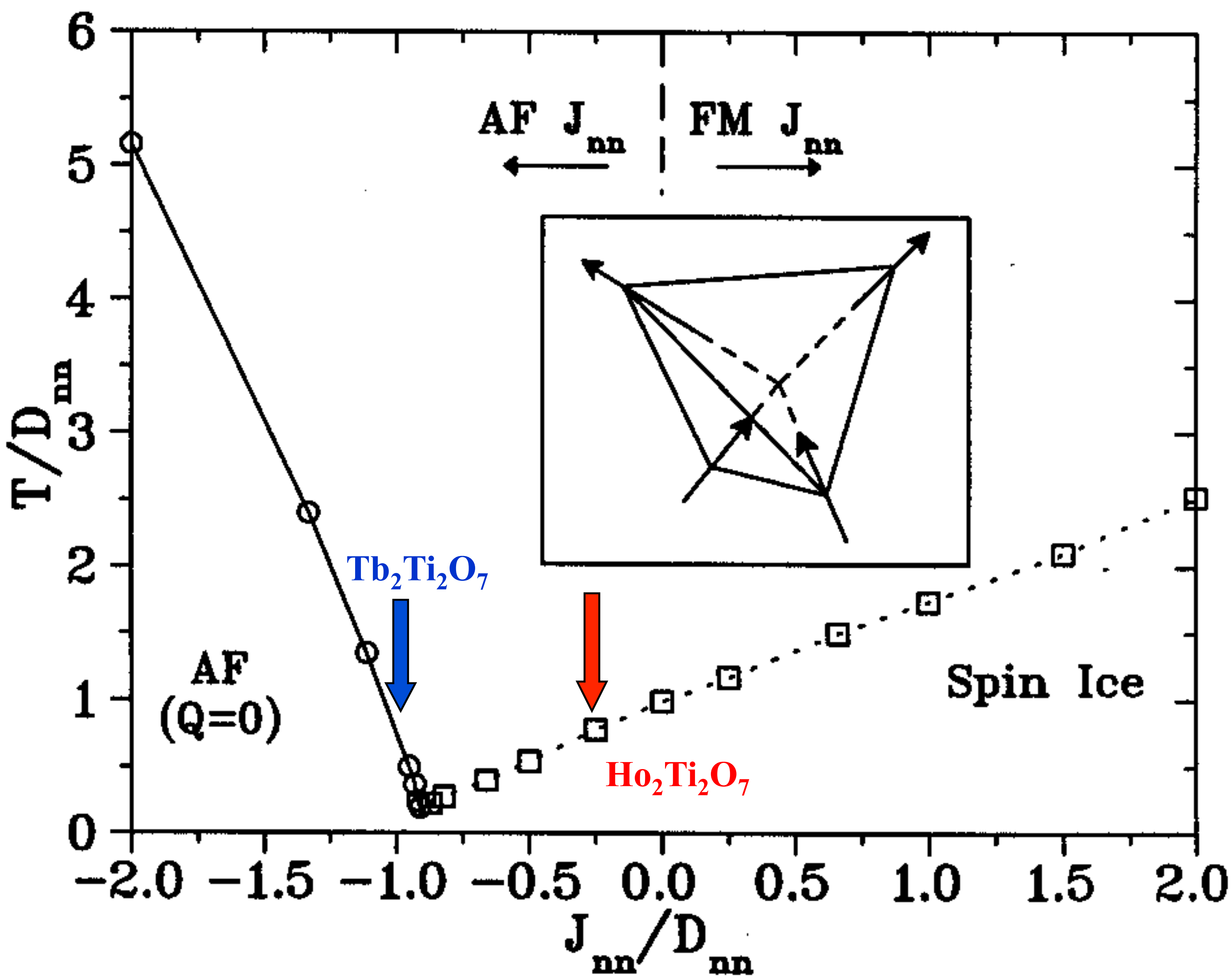






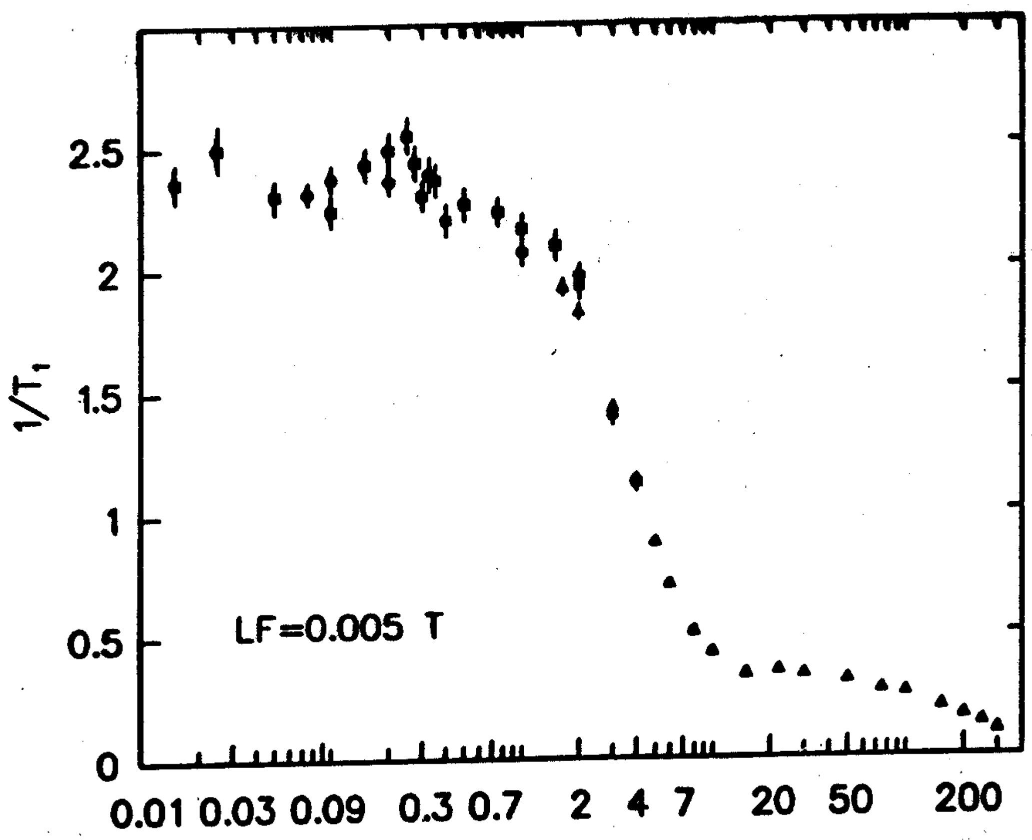


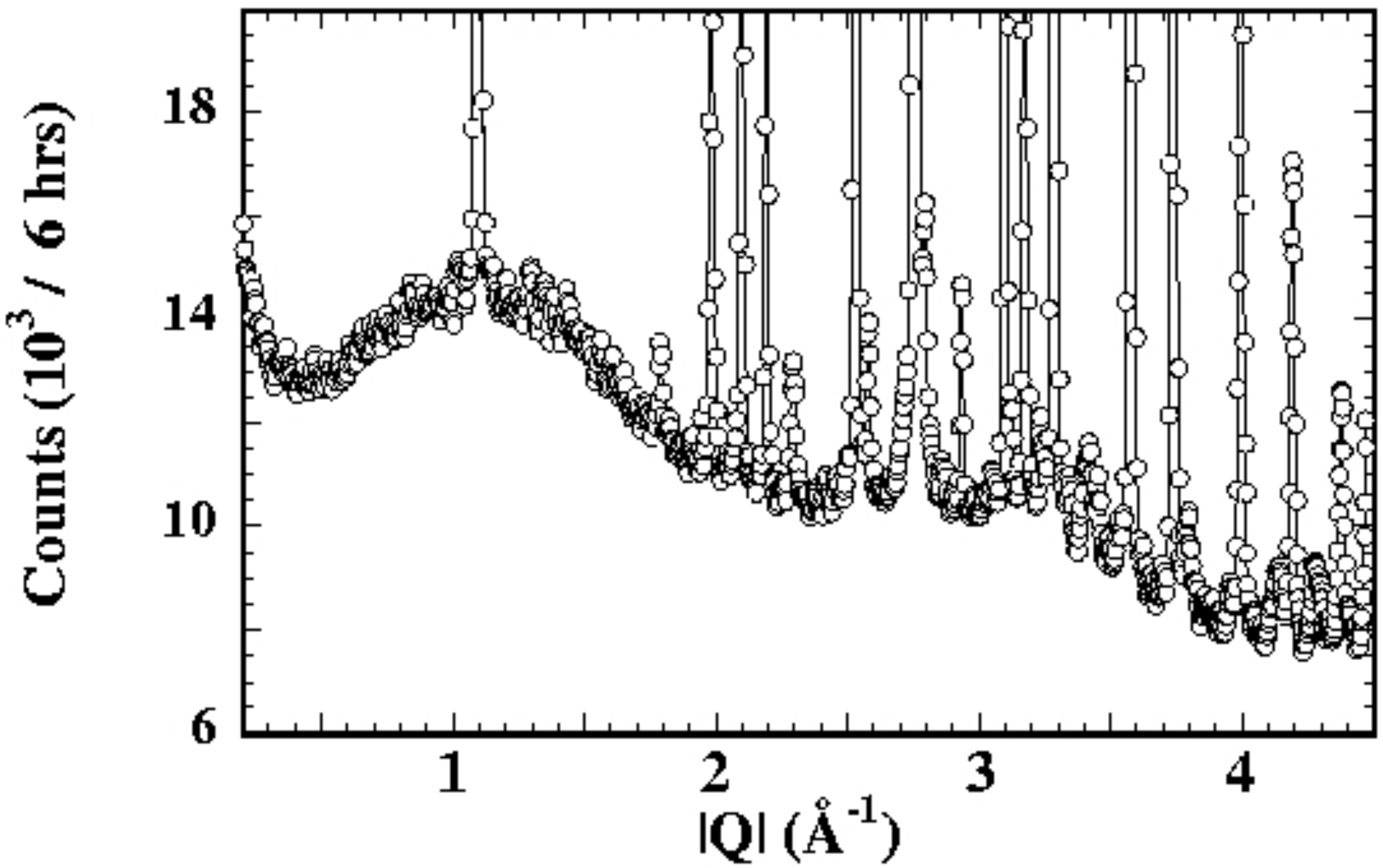
K. Matsuhira et al,
J. Phys. Cond. Matter, 14, L559, 2002



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

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

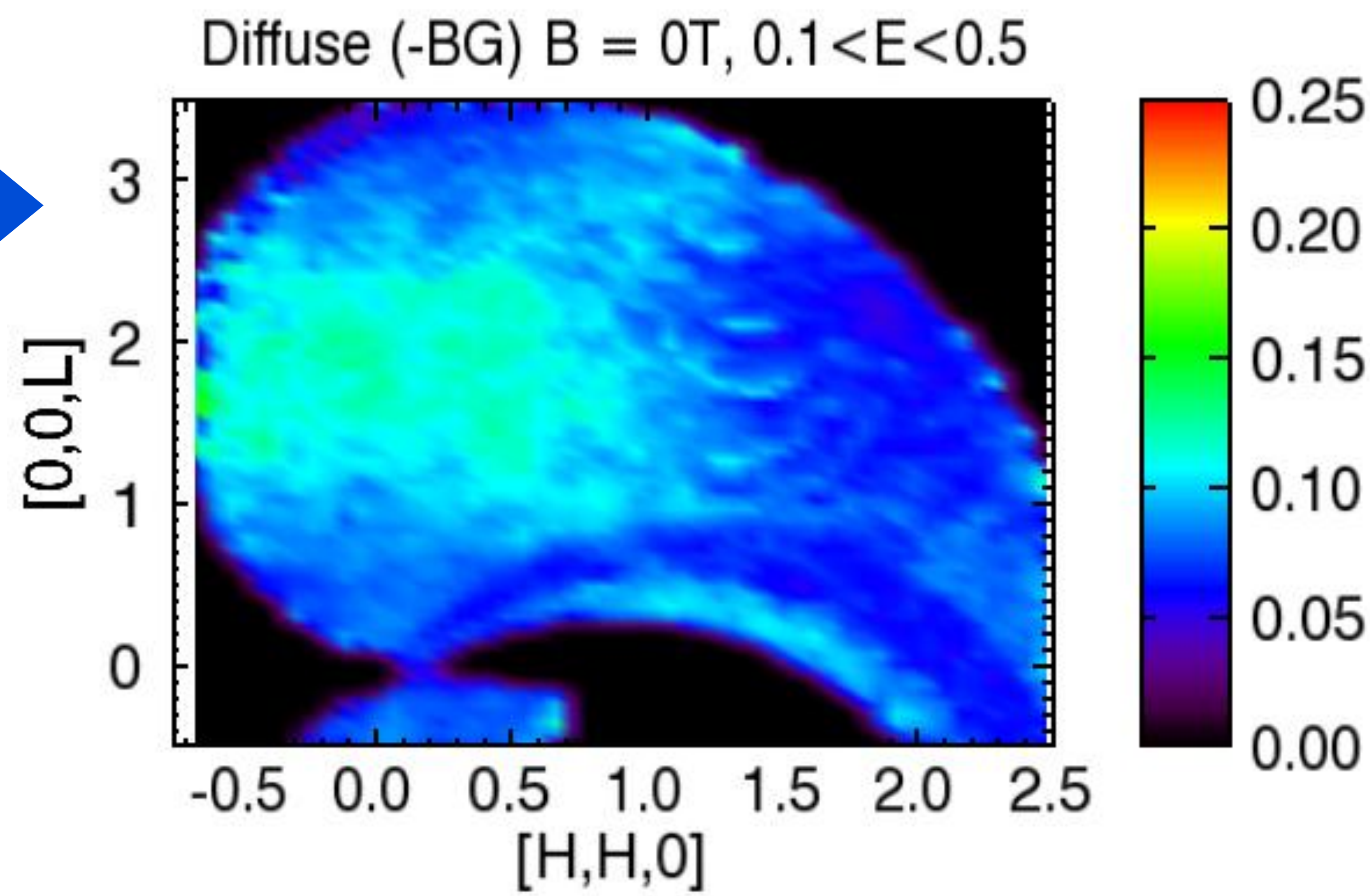




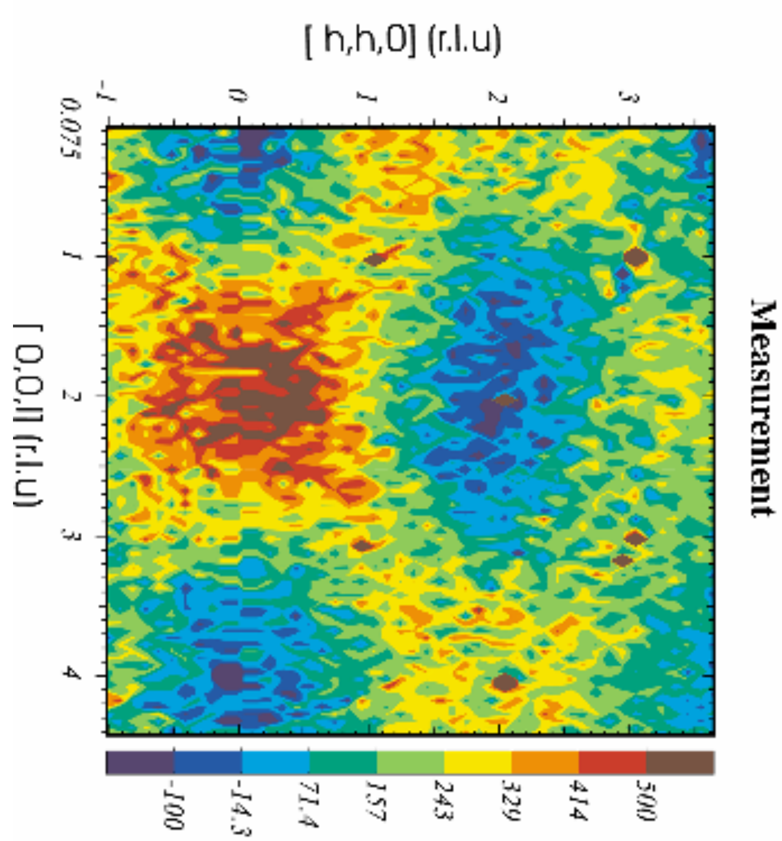
1/T_1 relaxation rate muSR

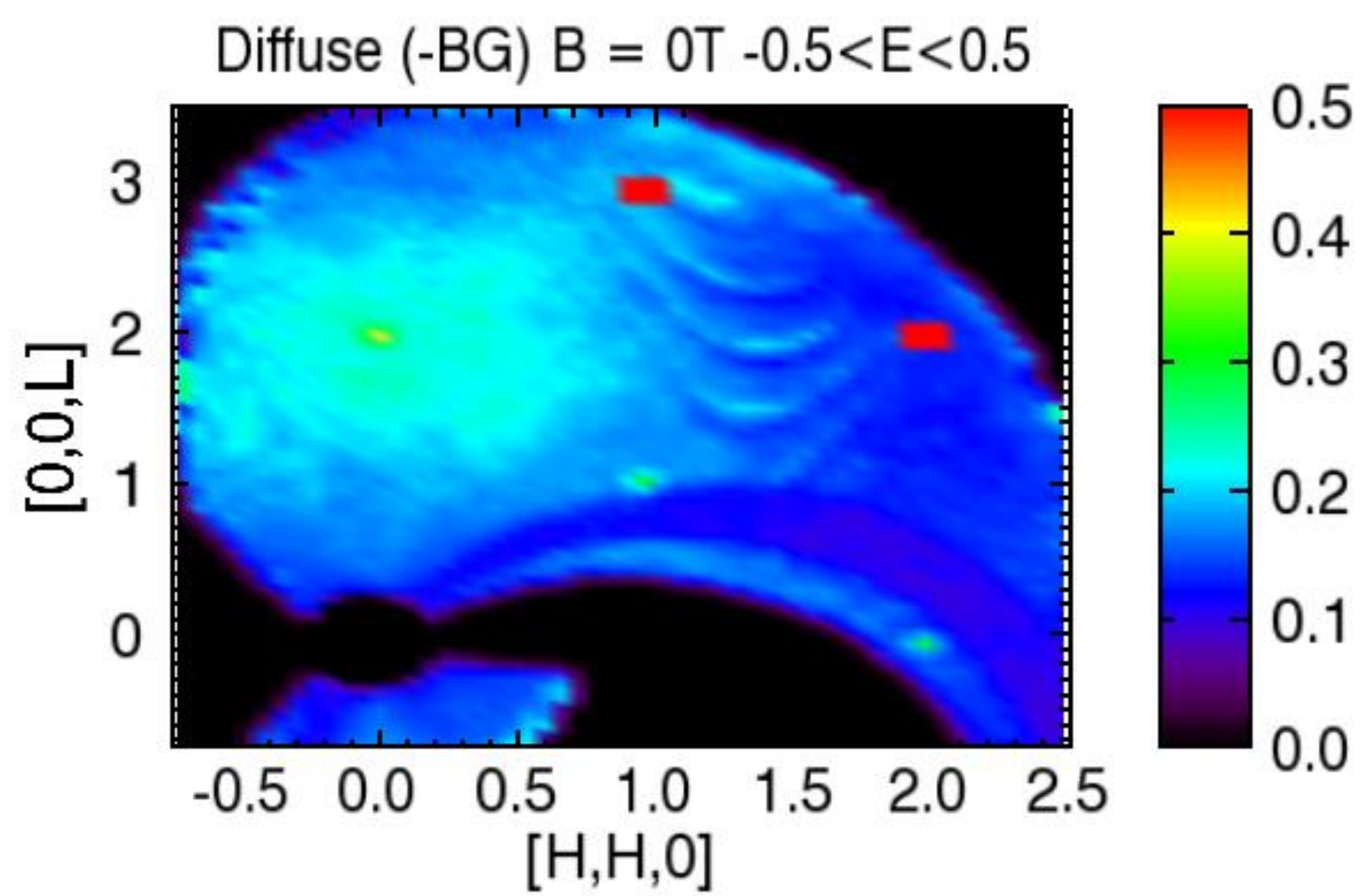
Low T powder neutron diffraction

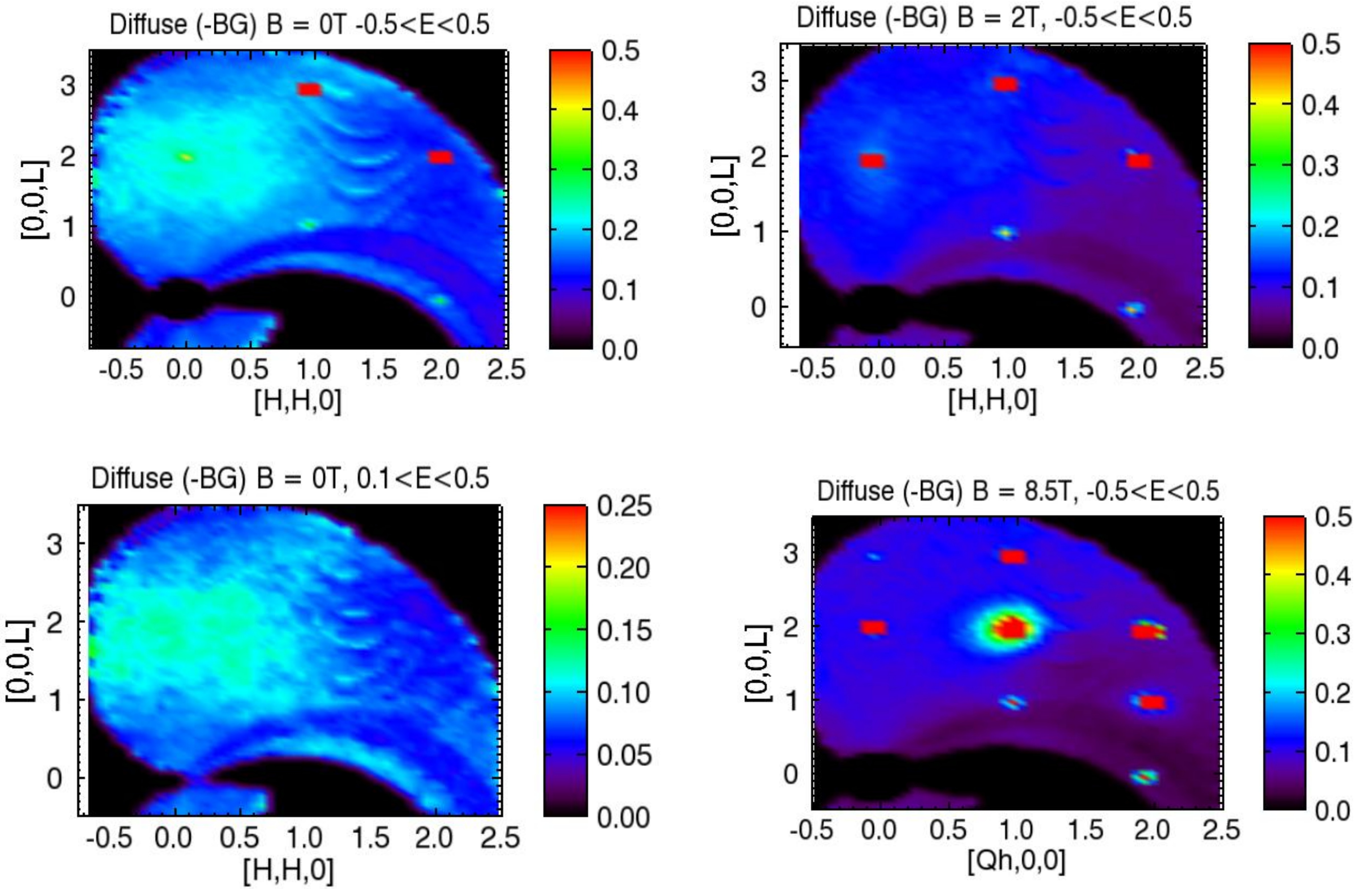
TOF scattering
T=0.4 K



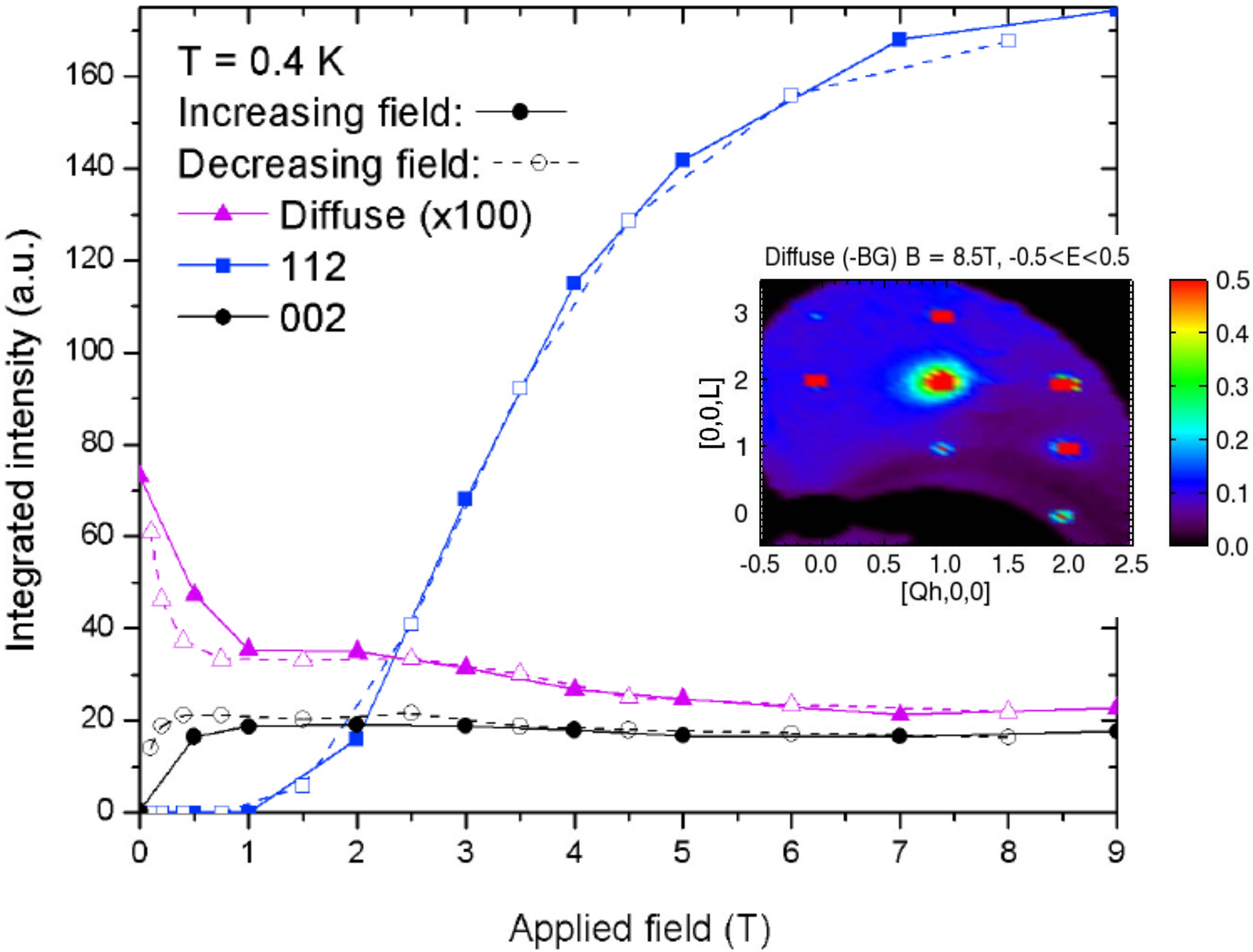
2-Axis Diffraction
T=9 K

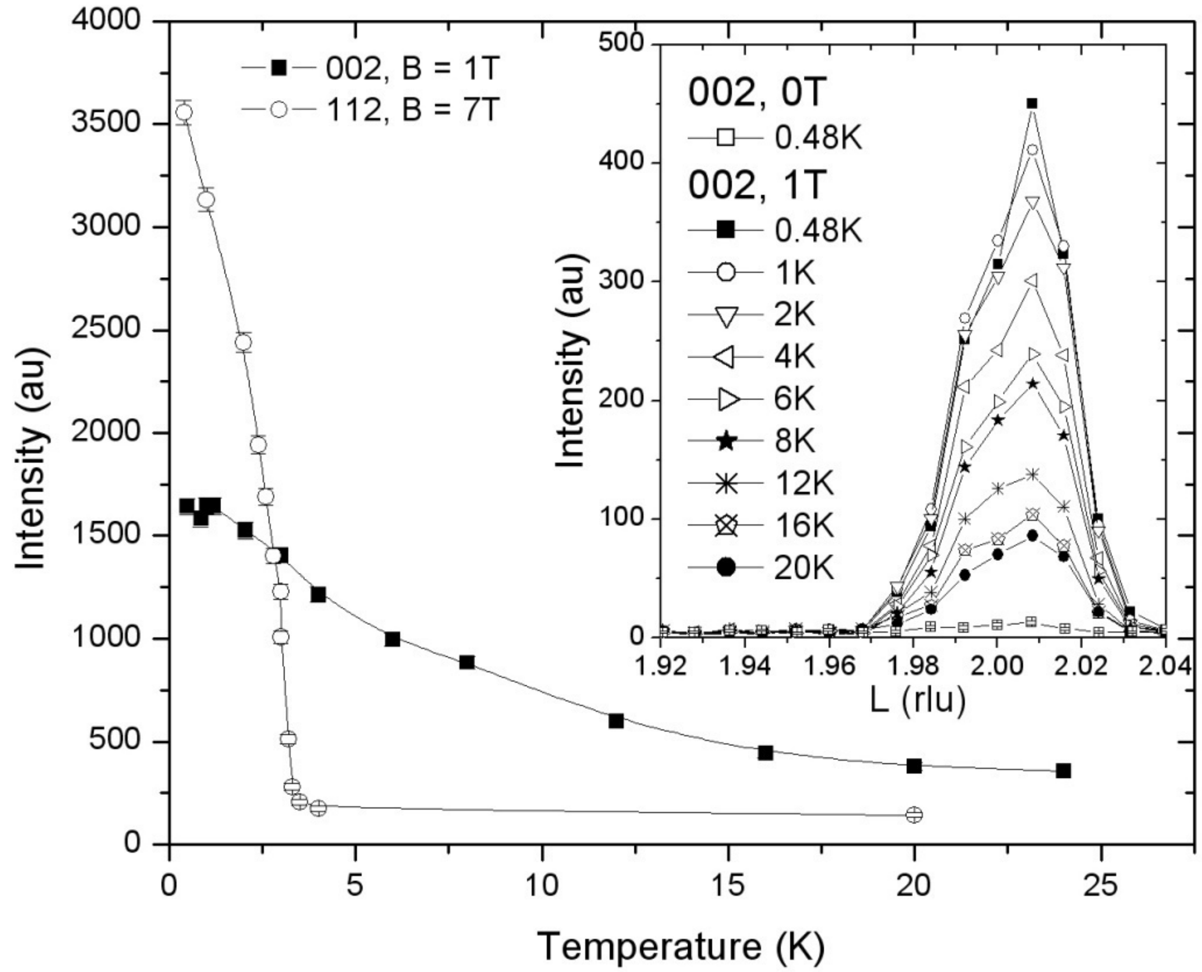


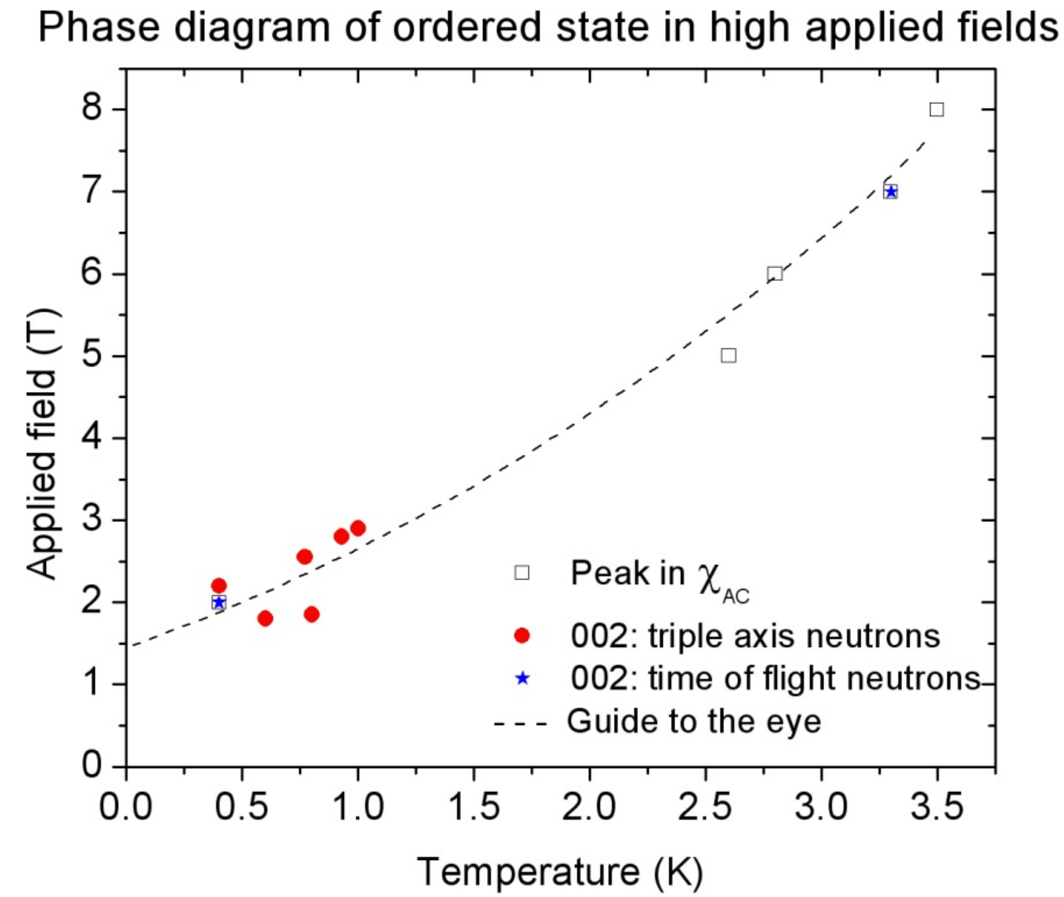


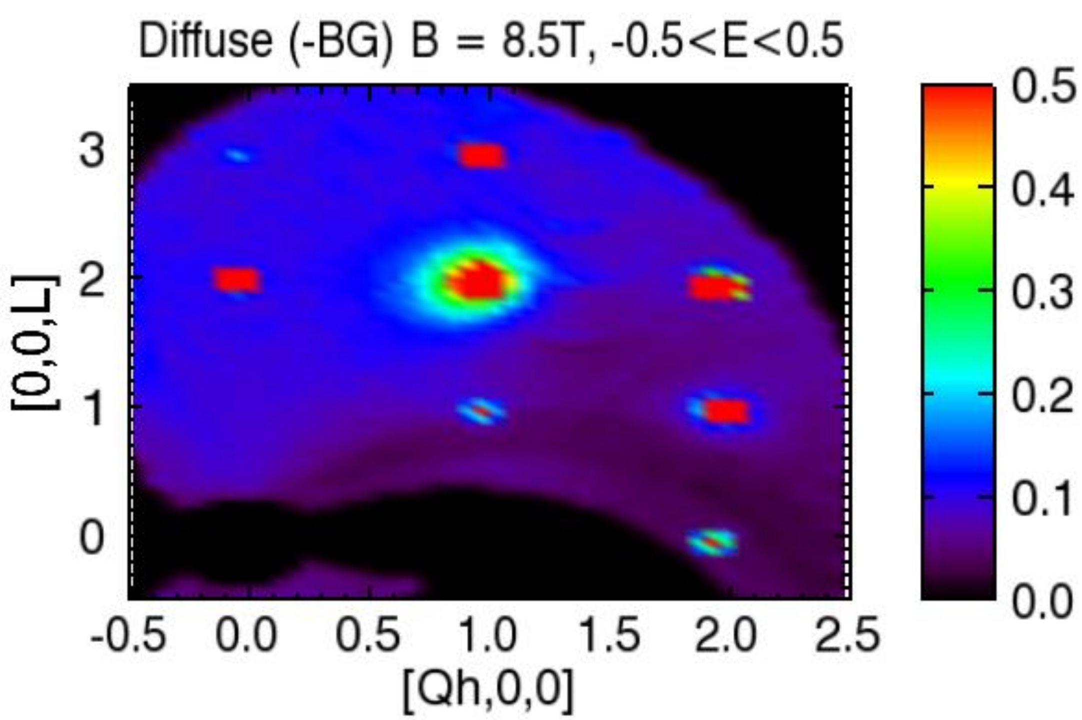


Ordered phase(s) appear(s) on application of H // 110


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
0.0
0.1
0.2
0.3
0.4
0.5

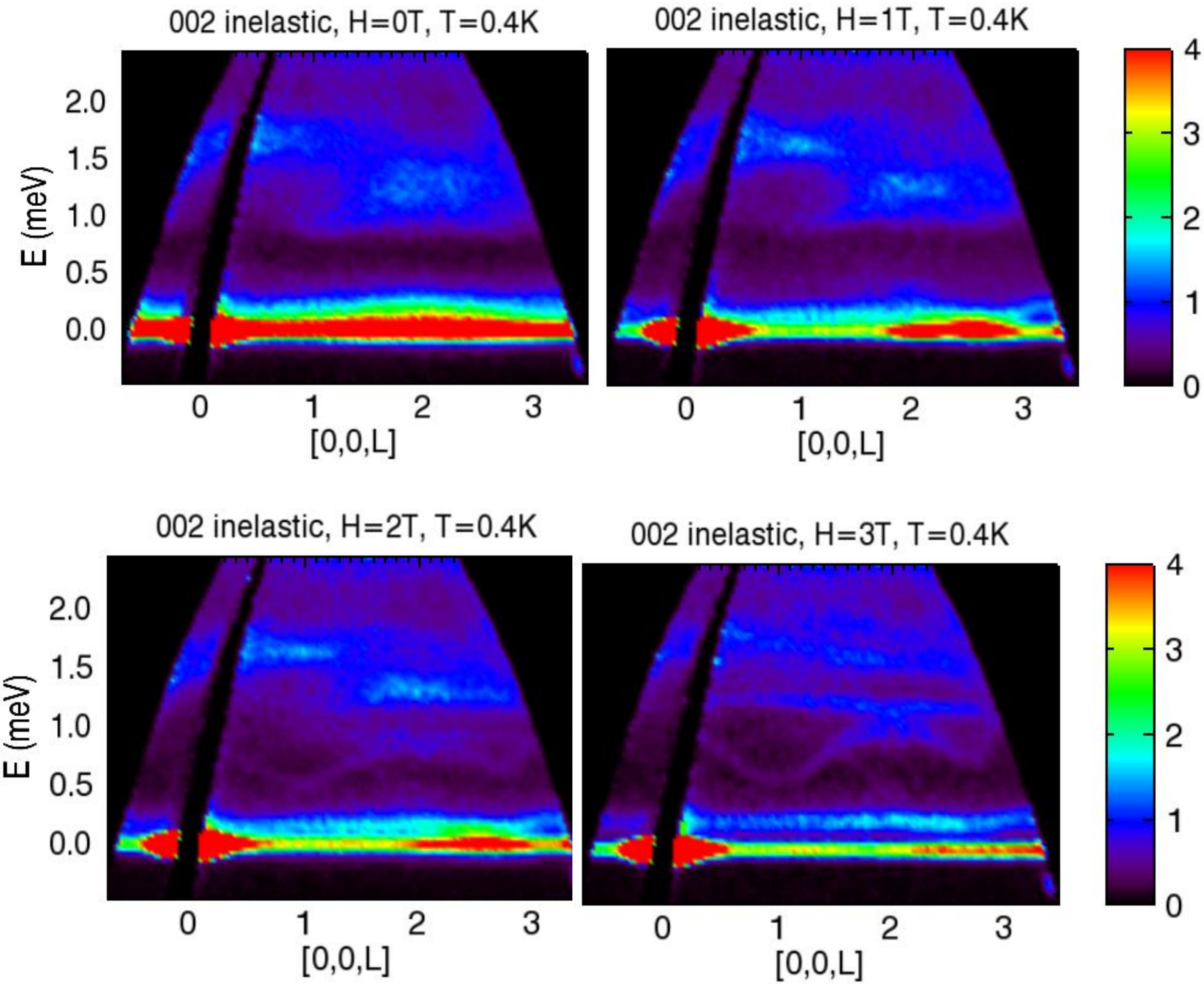




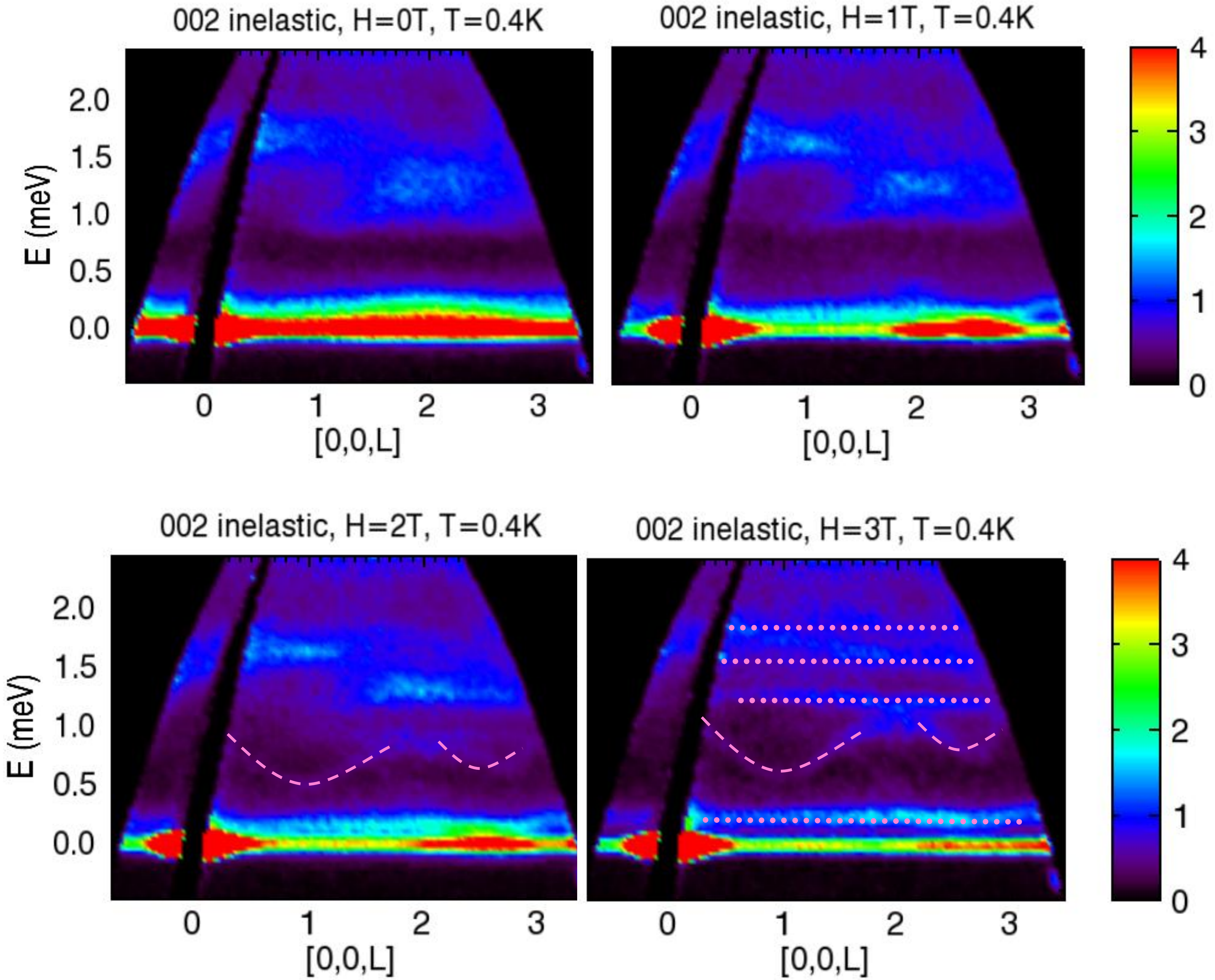


Low field (002) phase persists to very high T > 25 K

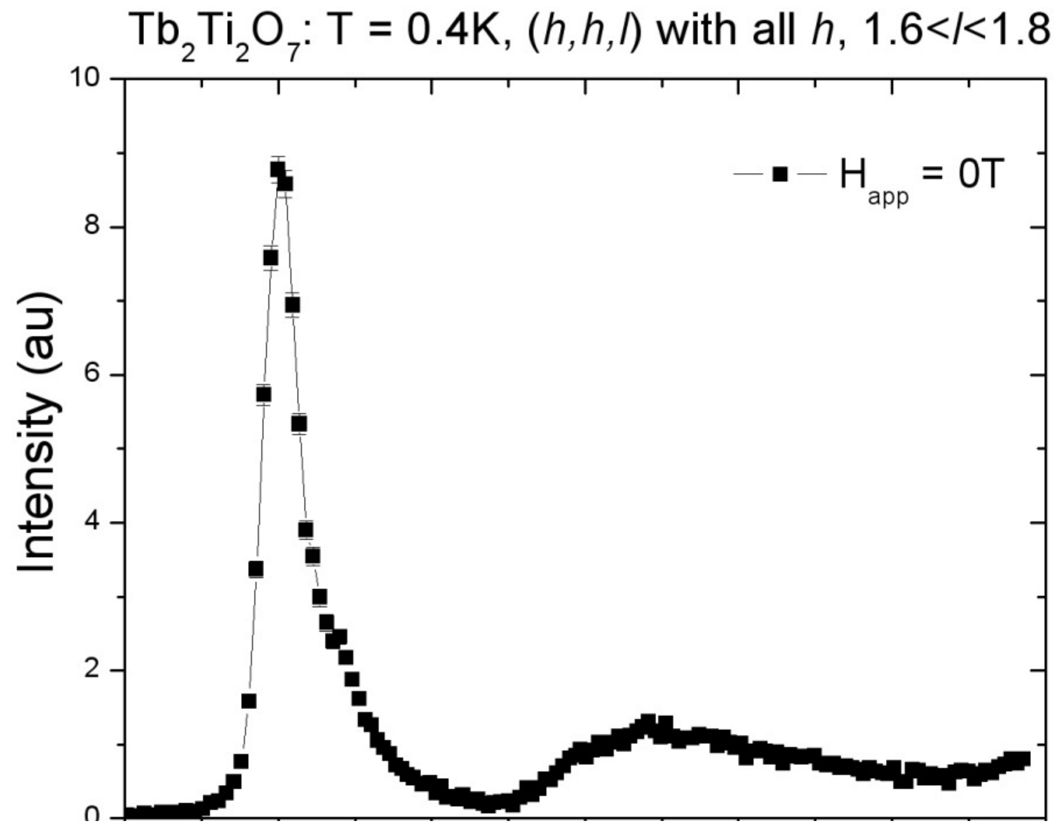
High field (112) phase exists on expected $T_N \sim 2$ K



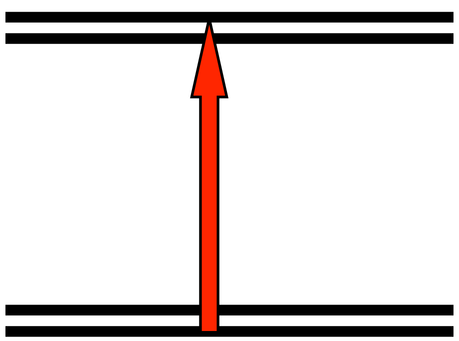
002 inelastic, H=0T, T=0.4K
002 inelastic, H=1T, T=0.4K
002 inelastic, H=2T, T=0.4K
002 inelastic, H=3T, T=0.4K
E (meV)
[0,0,L]

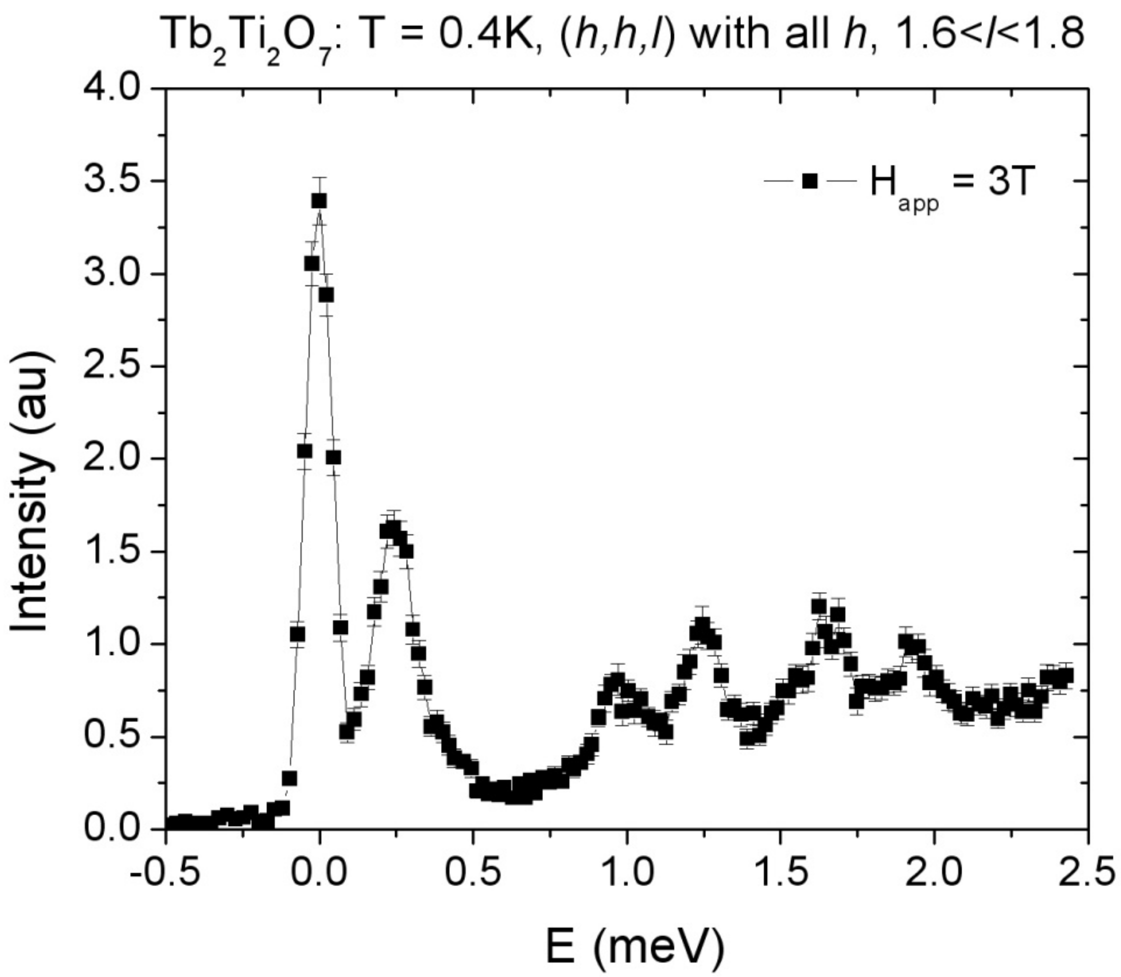


K. C. Rule, J. P. C. Ruff, B. D. Gaulin, S. R. Dunsiger, J. S. Gardner, J. P. Clancy, M. J. Lewis, H. A. Dabkowska, I. Mirebeau, P. Manuel, Y. Qiu, and J. R. D. Copley, Phys. Rev. Lett. 96, 177201 (2006)

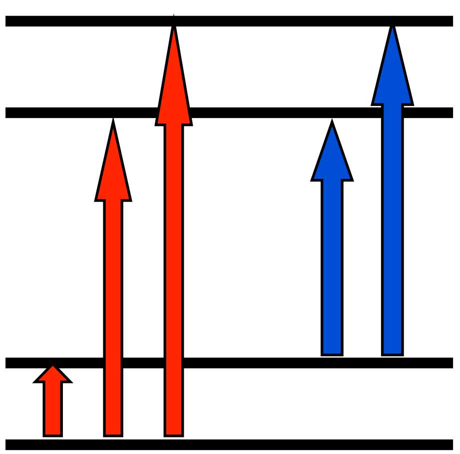


One Transition in Zero Field



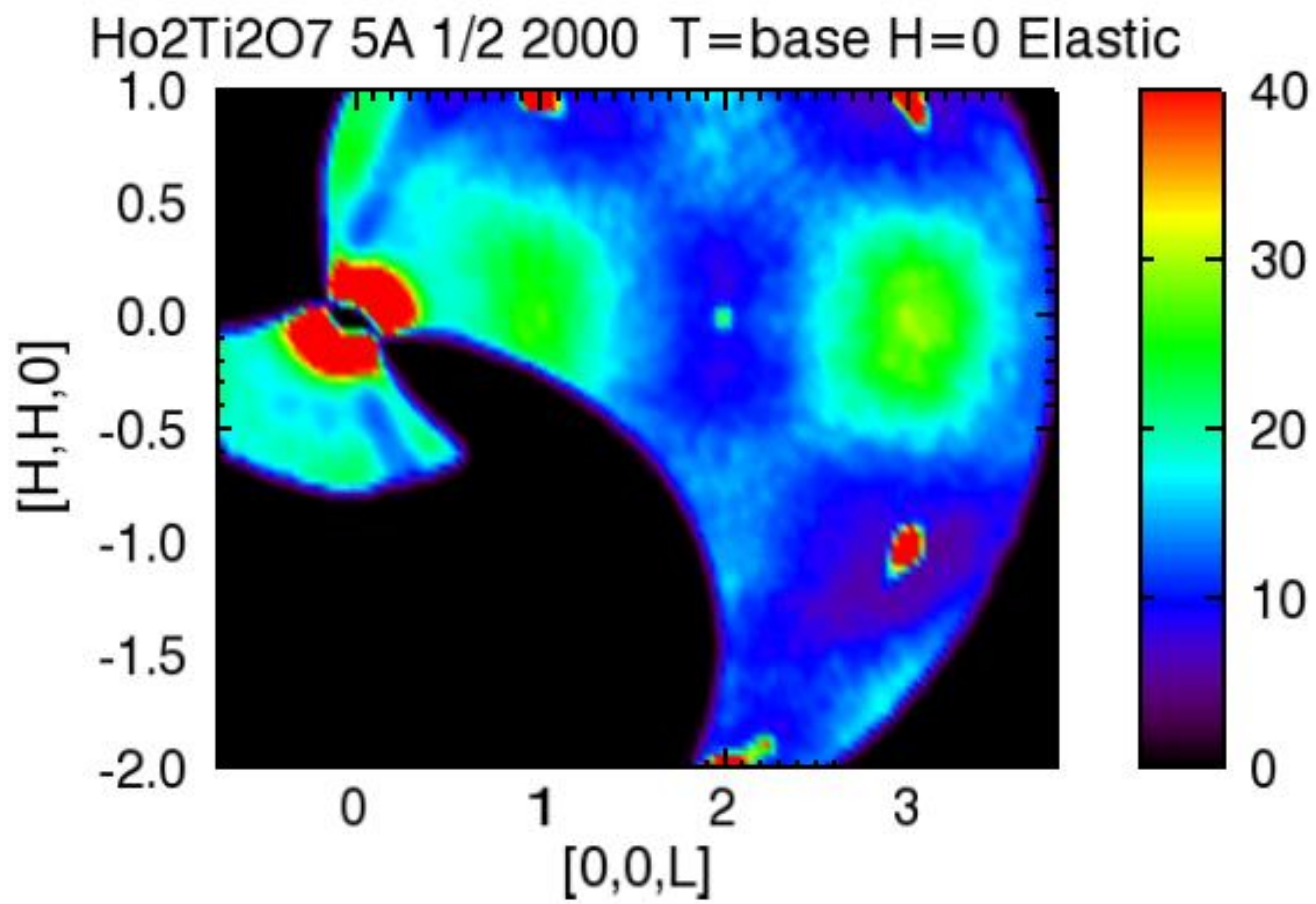


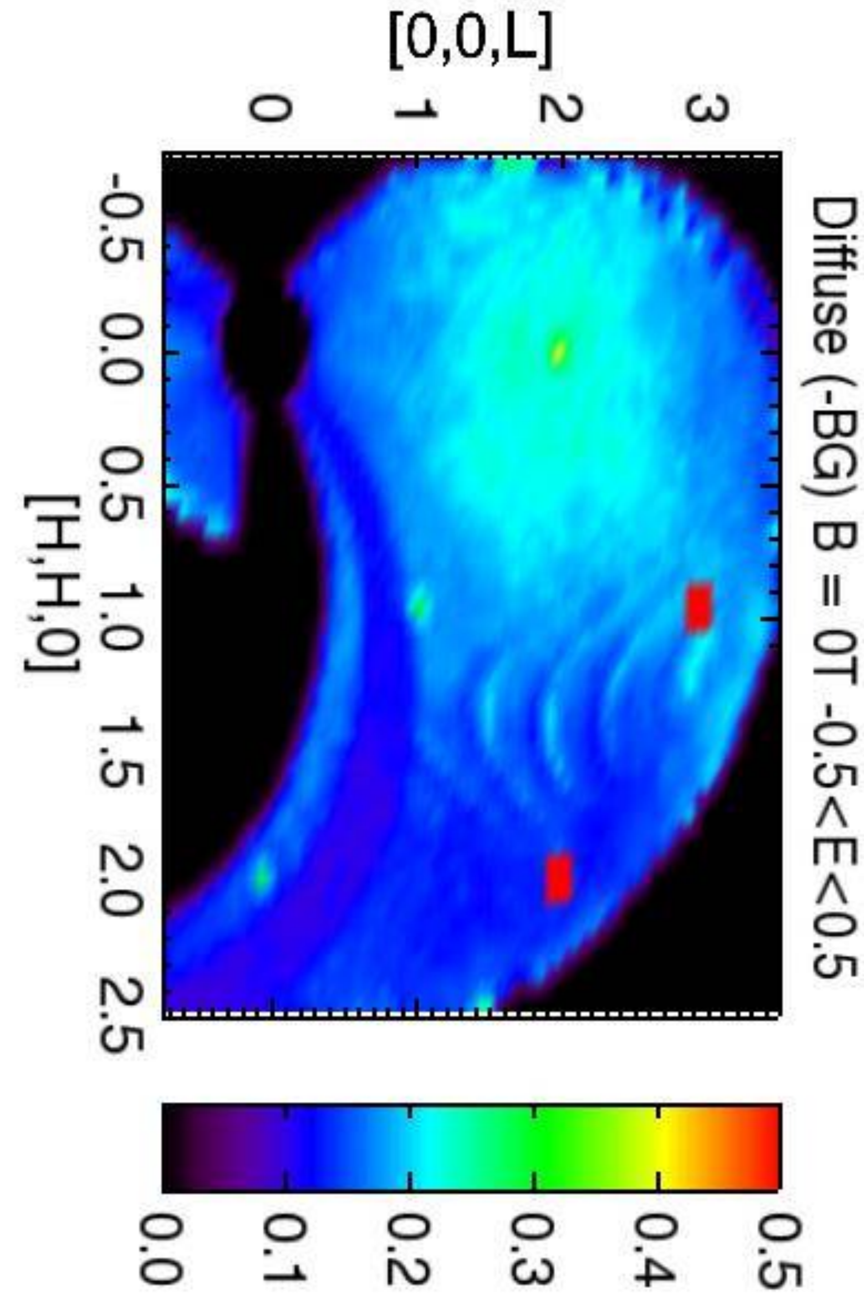
Five Transitions in Non-Zero Field



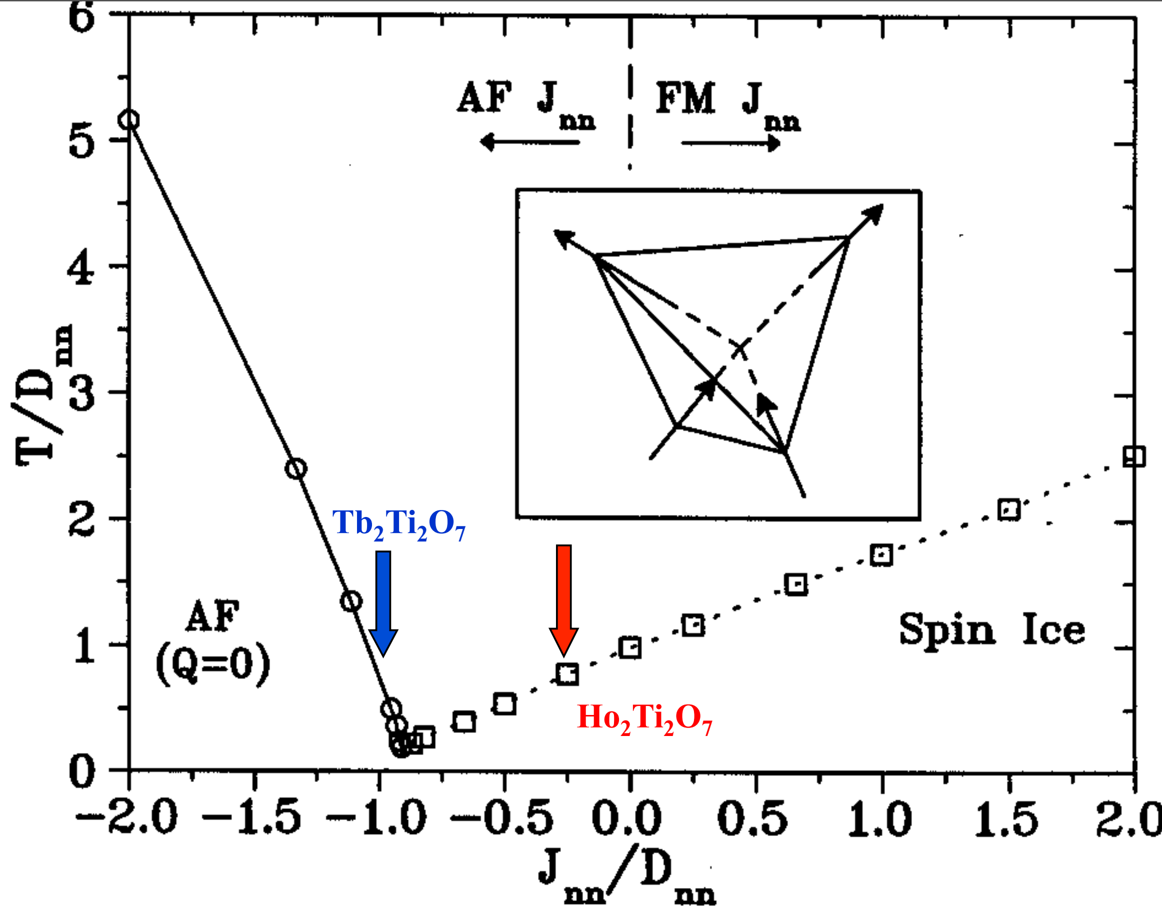
$Ho_2Ti_2O_7$ vs $Tb_2Ti_2O_7$

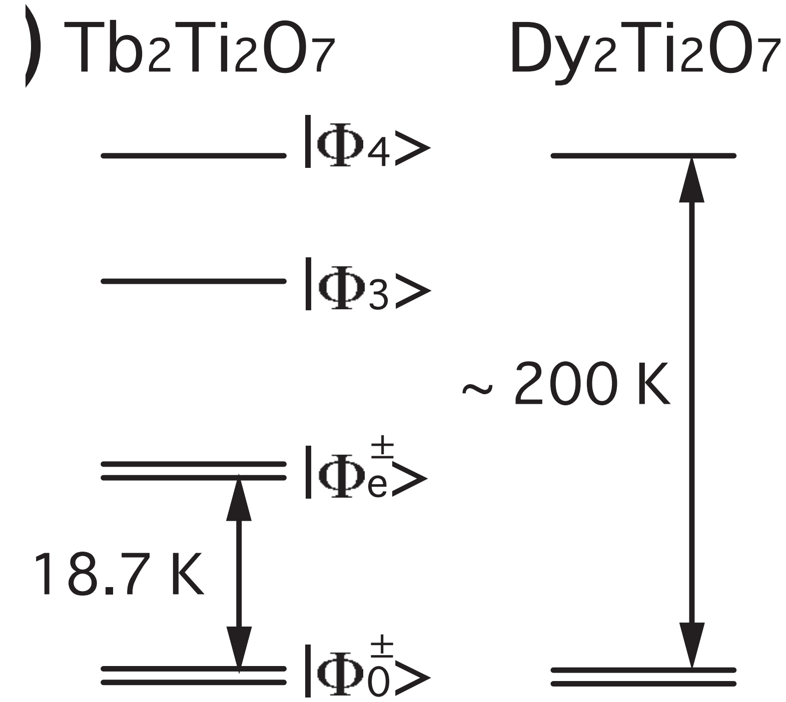
Static Spin Ice vs Dynamic Spin Liquid (H=0)

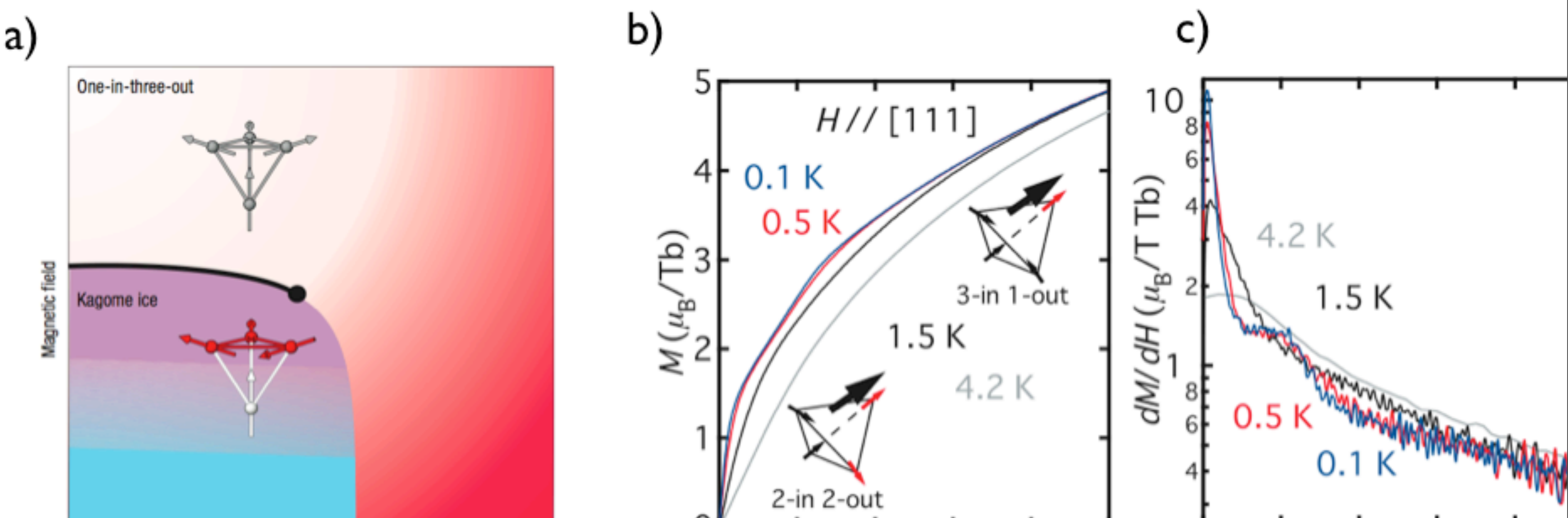




Magnetic Structure Factors appear complementary







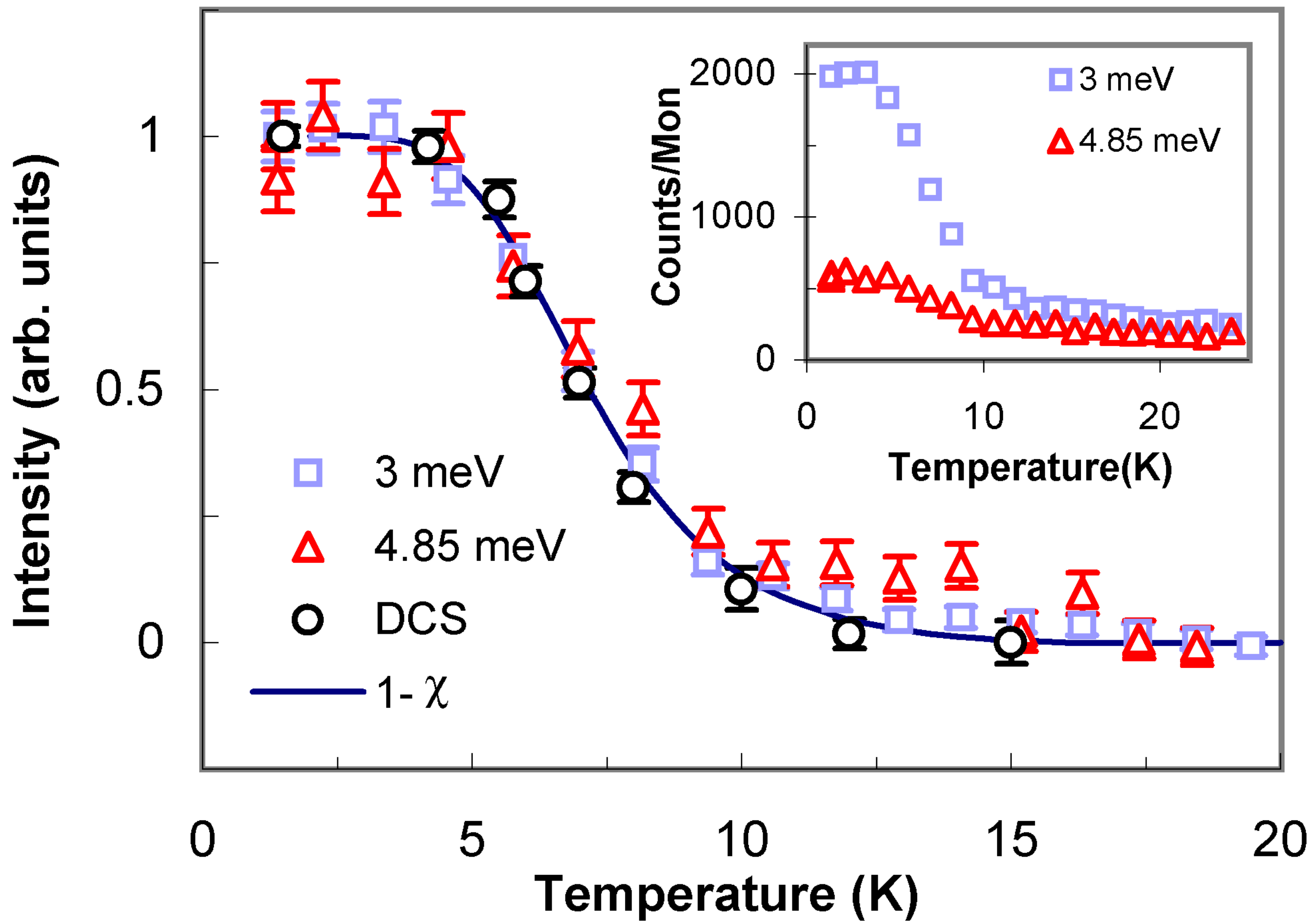
$Tb_2Ti_2O_7$ displays evidence for magnetization plateau in [111] magnetic field:
Signature of spin ice state - quantum spin ice?

Kate Ross

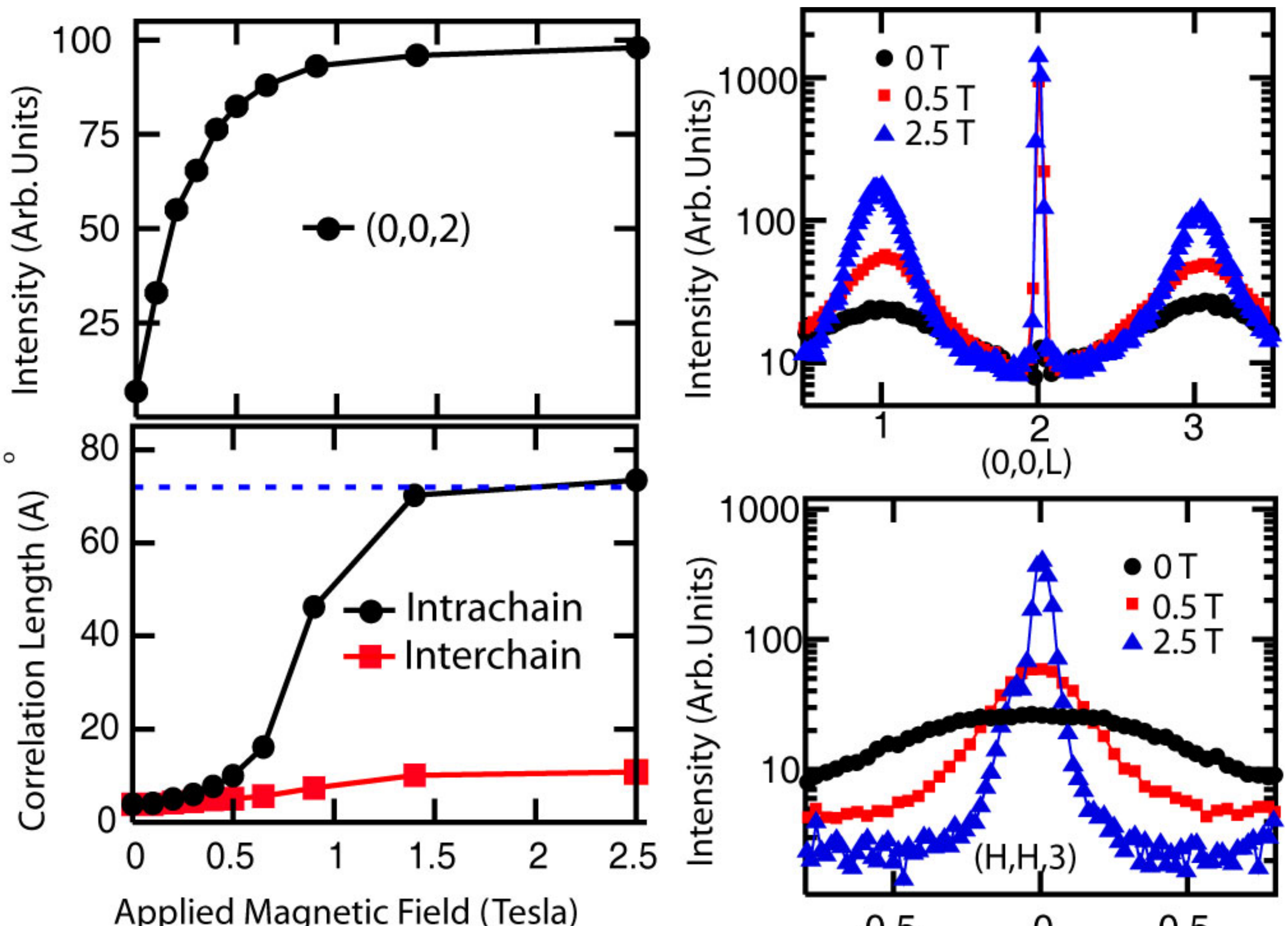
Pat Clancy

Jacob Ruff

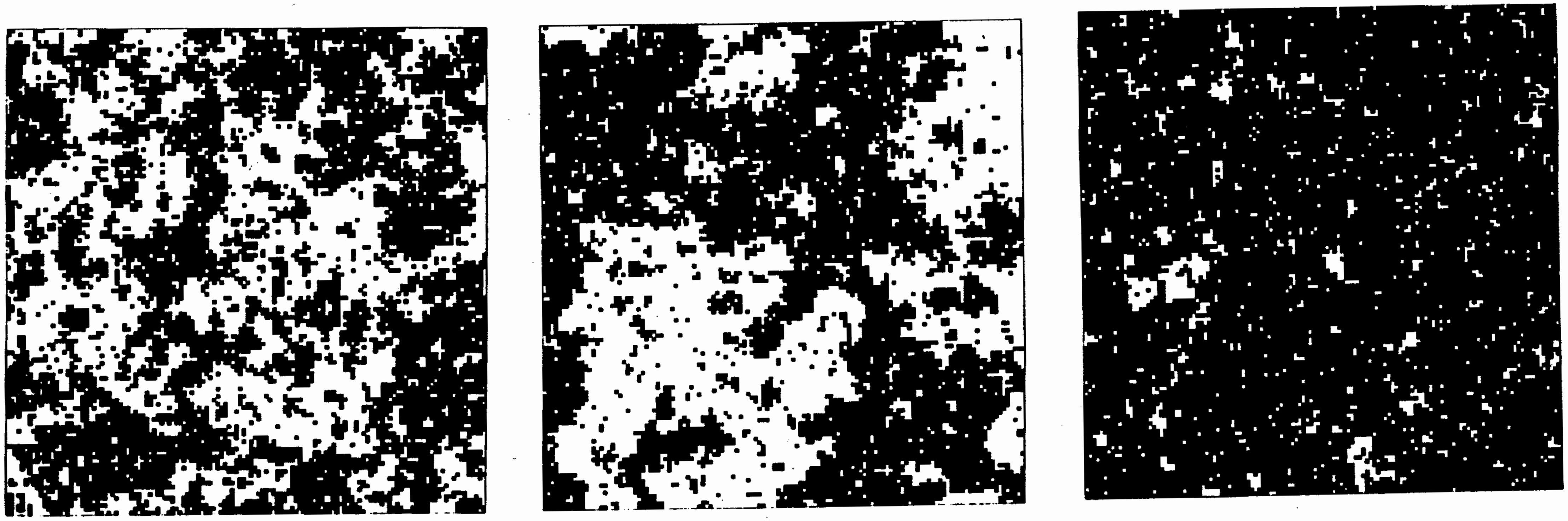
- *Rare earth titanates: playground for exotic frustrated ground states*
- *$Ho_2Ti_2O_7$: Canonical spin ice*
  1D decomposition in [110] field; kagome ice + plateau in [111] field
- *$Tb_2Ti_2O_7$: enigmatic spin liquid*
  [110] field induced LRO; evidence for plateau in [111] field - quantum spin ice?



Temperature dependence of 1-triplet excitation and 2-triplet excitation are identical and follow the complement of the dc-susceptibility (1-χ).

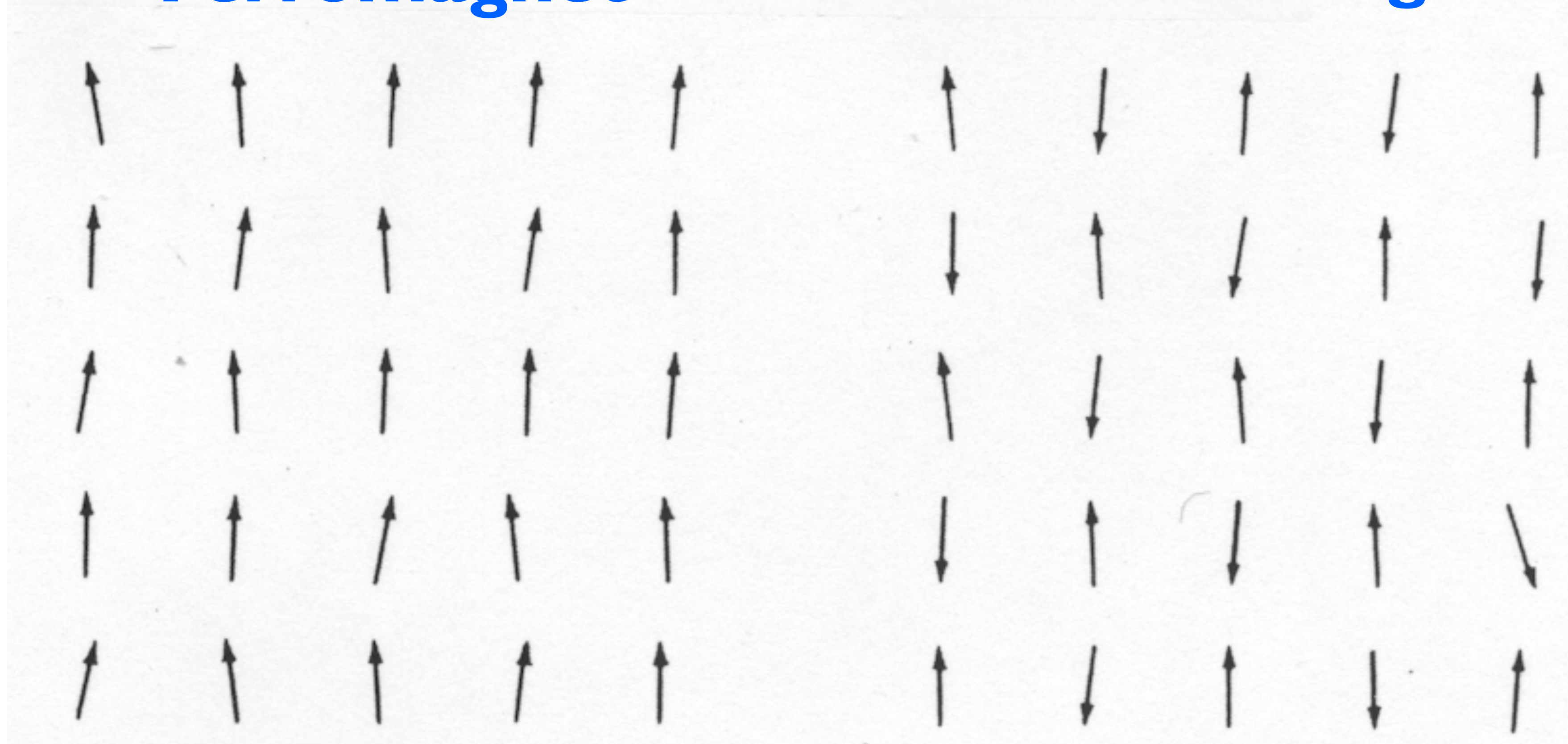


100
75
50
25
Intensity (Arb. Units)
(0,0,2)
80
60
40
20
0
Correlation Length (Å)
Intrachain
Interchain
0
0.5
1
1.5
2
2.5
Applied Magnetic Field (Tesla)
0 T
0.5 T
2.5 T
1000
100
10
Intensity (Arb. Units)
1
2
3
(0,0,L)
1000
100
10
Intensity (Arb. Units)
0 T
0.5 T
2.5 T
(H,H,3)
-0.5
0
0.5

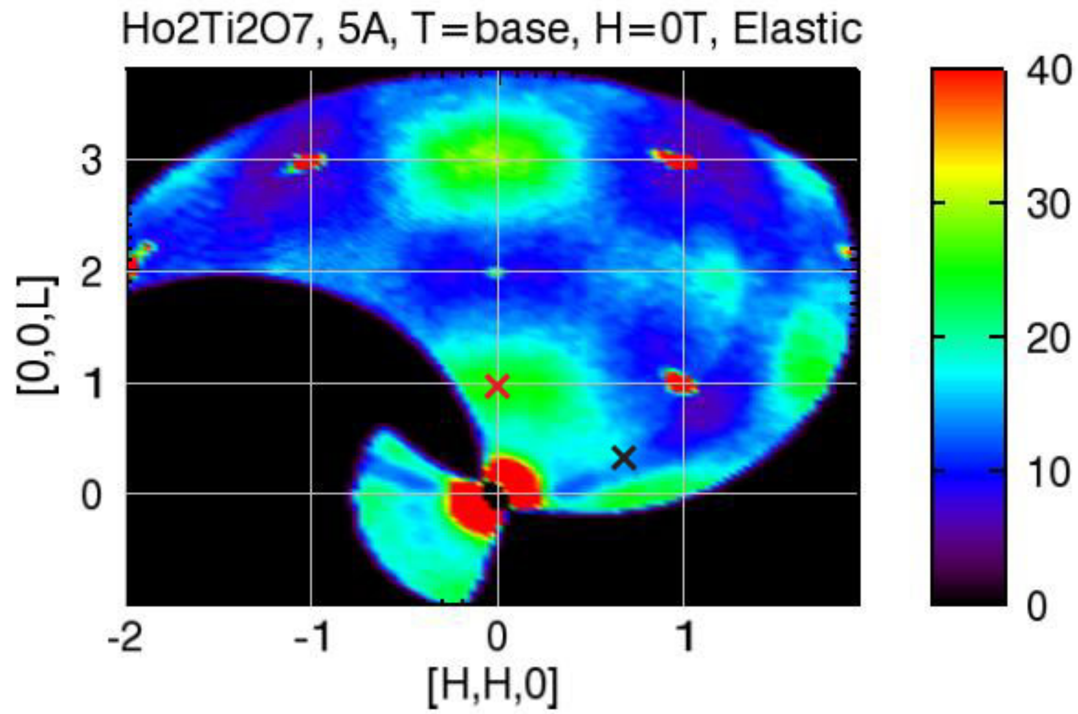


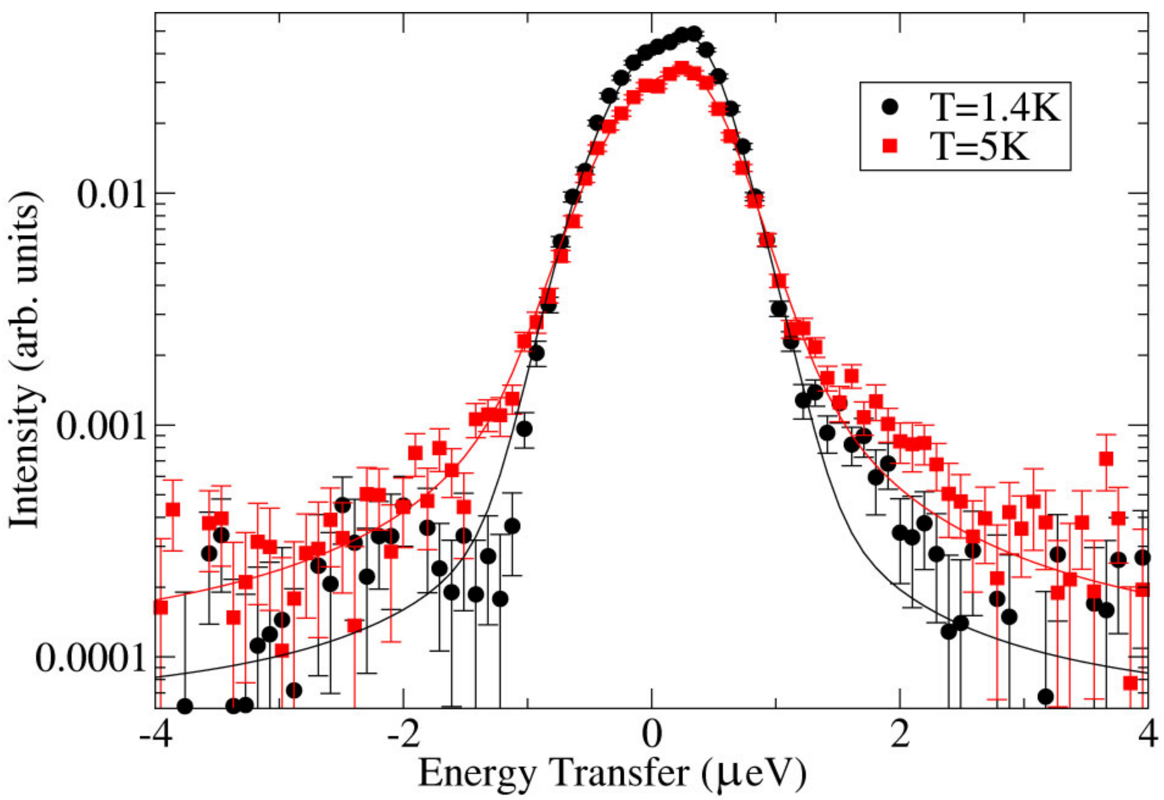
1.1 Tc   Tc   0.9 Tc

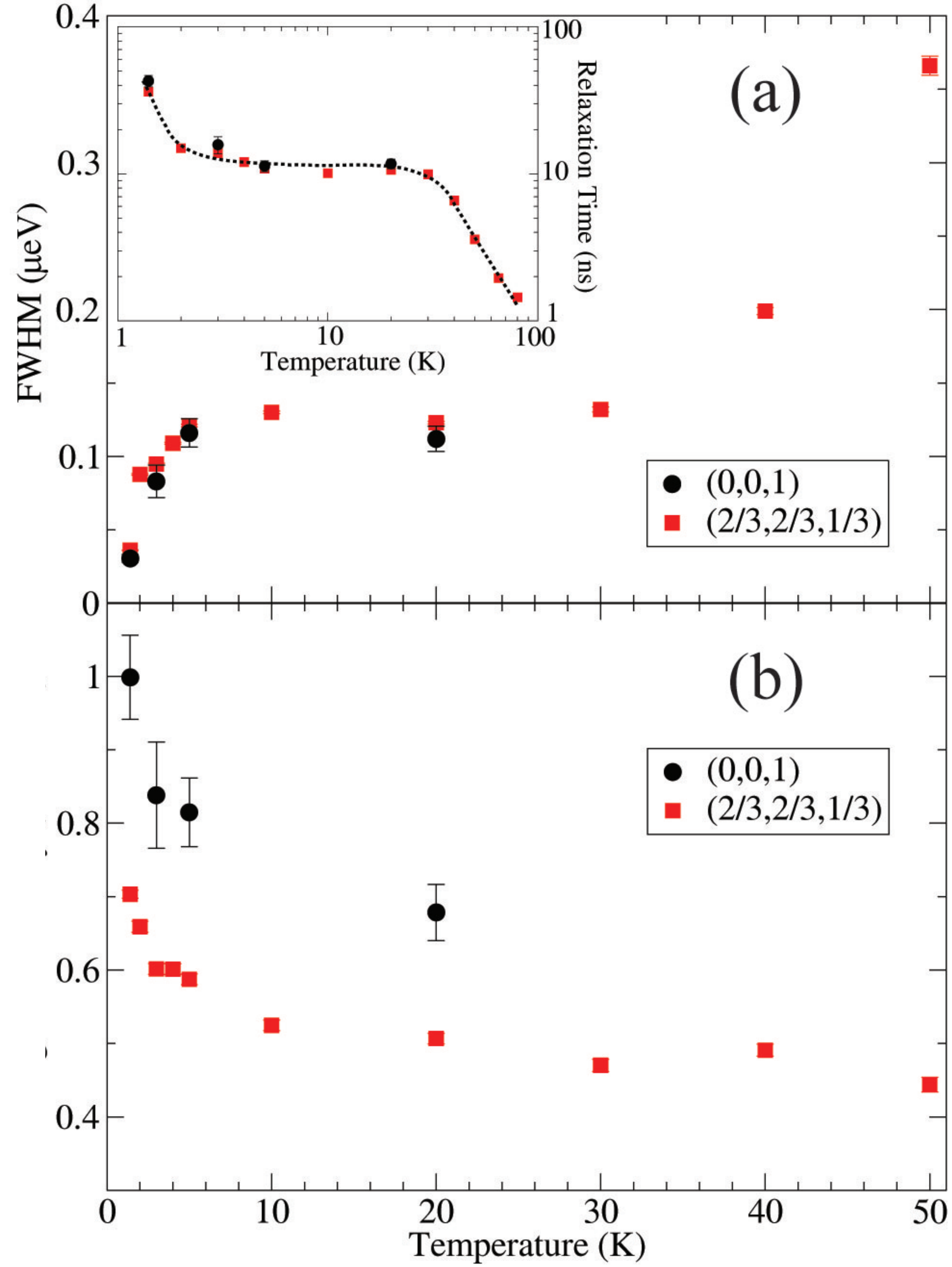
Ferromagnet   Antiferromagnet

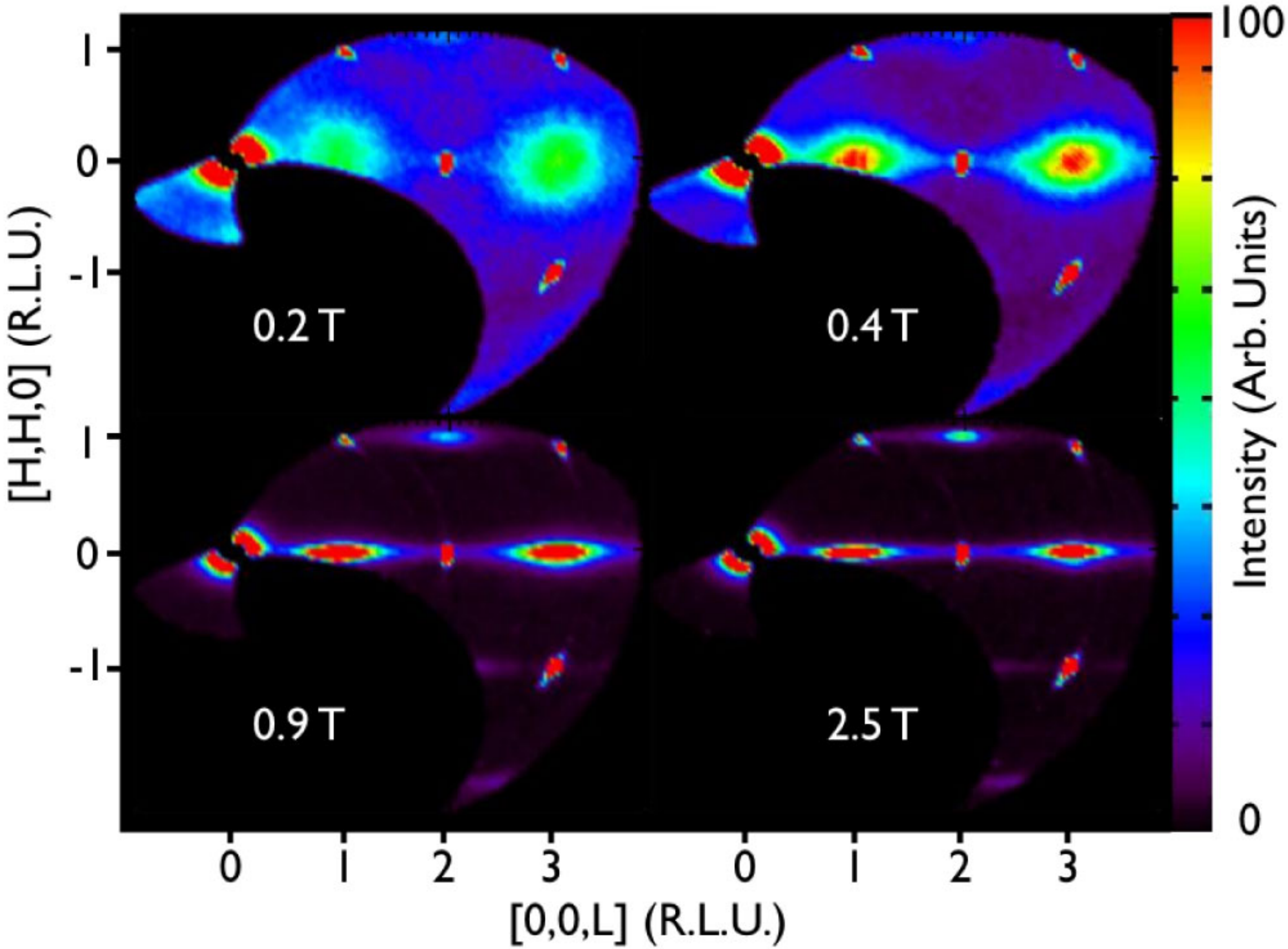


$$\mathrm{H} = 2\mathrm{J}\ \Sigma_{ij}\ \mathbf{S_i} \cdot \mathbf{S_j}$$


Ho2Ti2O7, 5A, T=base, H=0T, Elastic
[0,0,L]
[H,H,0]
-2
-1
0
1
2
3
40
30
20
10
0


T=1.4K
T=5K
Intensity (arb. units)
0.01
0.001
0.0001
-4
-2
0
2
4
Energy Transfer (μeV)

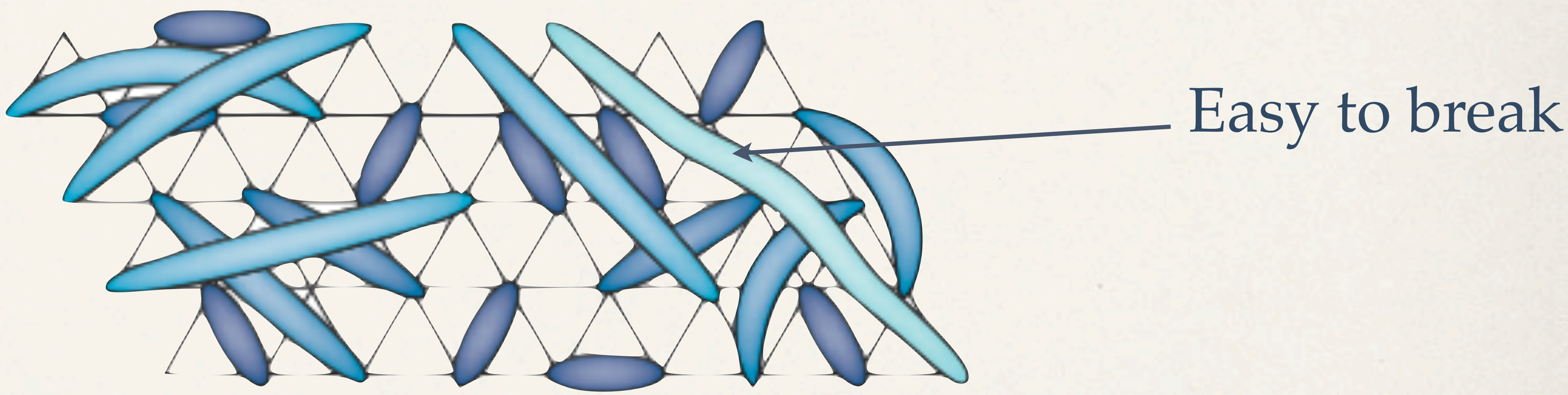

(a)
FWHM (μeV)
0.4
0.3
0.2
0.1
0
Relaxation Time (ns)
100
10
1
Temperature (K)
(0,0,1)
(2/3,2/3,1/3)
(b)
1
0.8
0.6
0.4
0
10
20
30
40
50
Temperature (K)



100
0
Intensity (Arb. Units)
[H,H,0] (R.L.U.)
[0,0,L] (R.L.U.)
0.2 T
0.4 T
0.9 T
2.5 T
1
0
-1
0
1
2
3

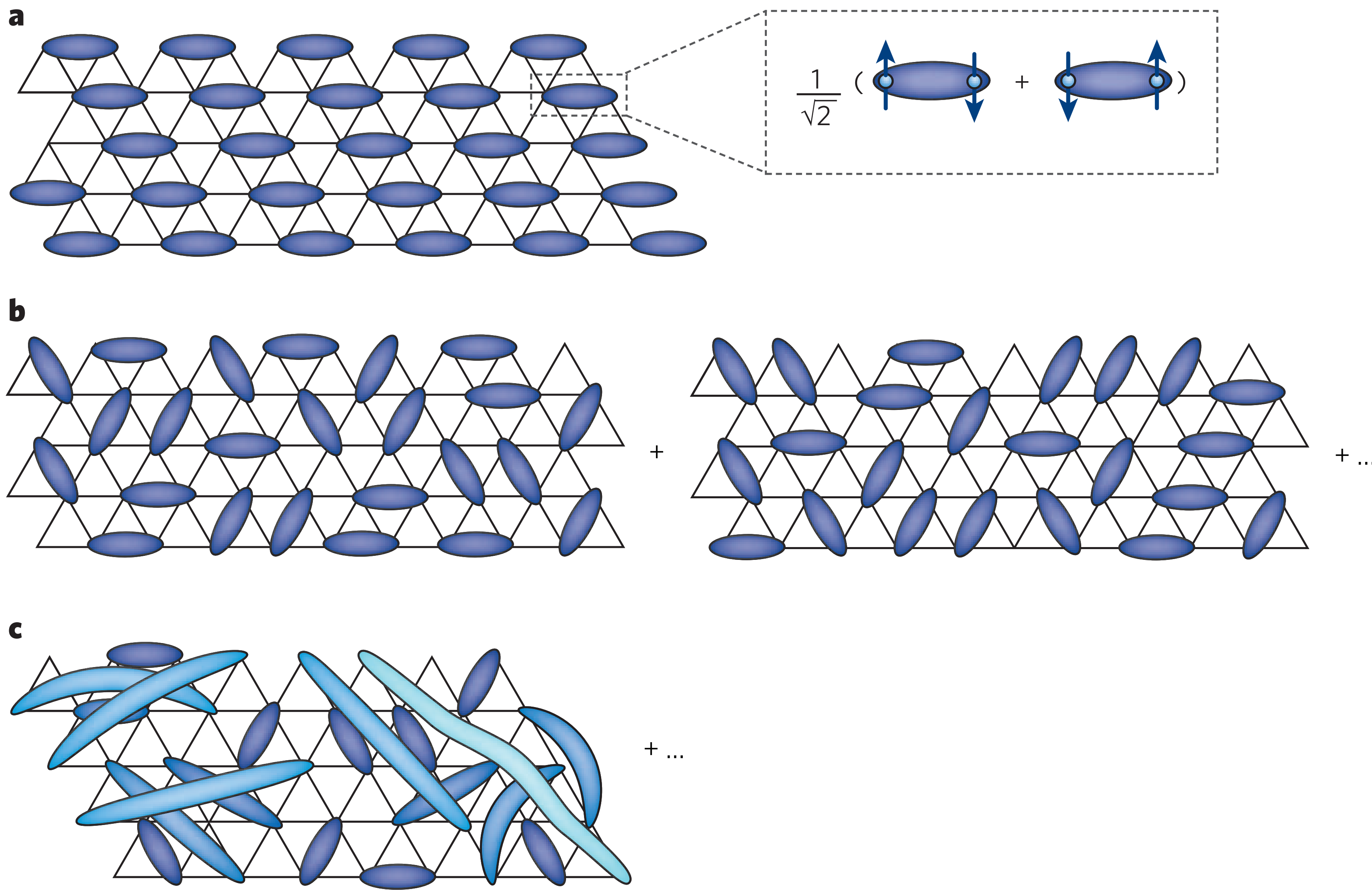
Spin Excitations: Spinons in lRVB

in lRVB state, creating 2 spinons can cost NO energy because long bonds are bound by distant neighbor exchange only



NOTE:

1) Spinons are *gapless* and *deconfined*


a
$\frac{1}{\sqrt{2}}$ (+)
b
+
+ ...
c
+ ...

Motivation for Singlet Basis

$$H = J \sum_{<i,j>} \vec{S}_i \cdot \vec{S}_j$$

Broken Rotational Symmetry

Rotational Symmetry Intact

Neel type state for 2 spins:

$$\langle \uparrow_i \downarrow_j | \, S_{\mathrm{z}i} S_{\mathrm{z}j} \, | \uparrow_i \downarrow_j \rangle = -\frac{1}{4}$$

$$\langle \uparrow_i \downarrow_j | \, S_{\mathrm{x}i} S_{\mathrm{x}j} \, | \uparrow_i \downarrow_j \rangle = 0$$

$$\langle \uparrow_i \downarrow_j | \, S_{\mathrm{y}i} S_{\mathrm{y}j} \, | \uparrow_i \downarrow_j \rangle = 0$$

Entangled (singlet) state:

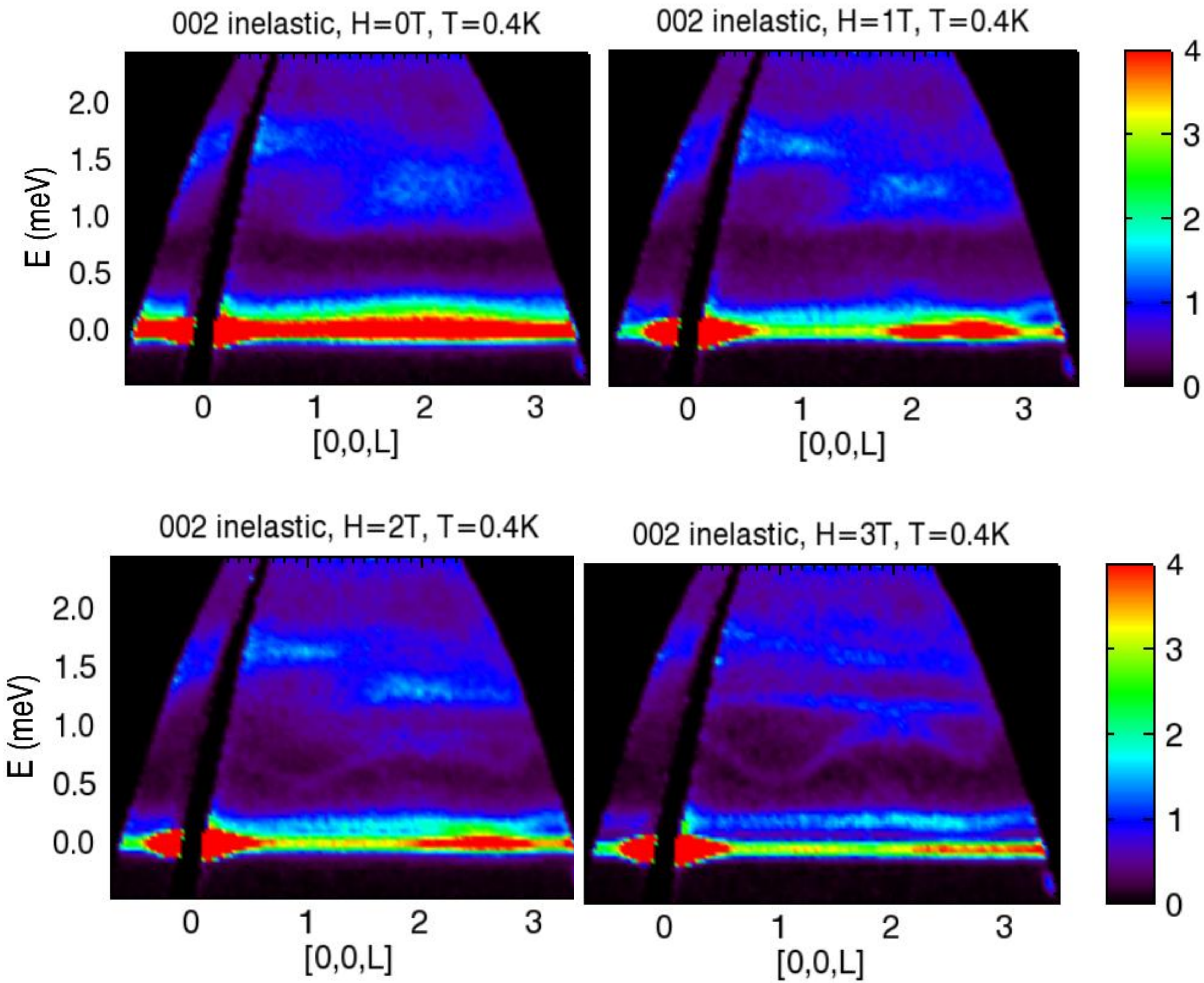
$$(\langle \uparrow_i \downarrow_j | - \langle \downarrow_i \uparrow_j |) S_{\mathrm{z}i} S_{\mathrm{z}j} (| \uparrow_i \downarrow_j \rangle - | \downarrow_i \uparrow_j \rangle) = -\frac{1}{4}$$

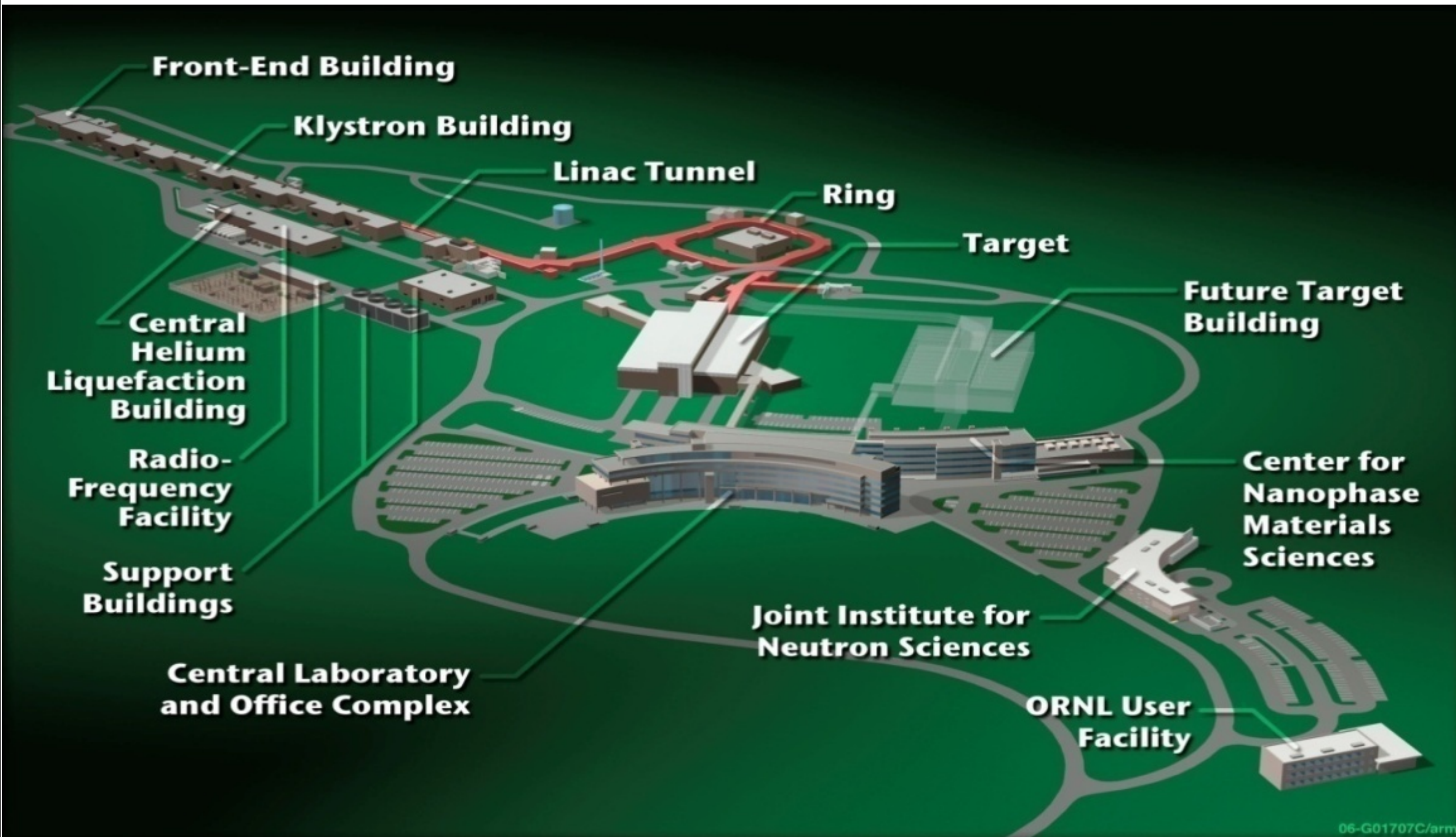
$$(\langle \uparrow_i \downarrow_j | - \langle \downarrow_i \uparrow_j |) S_{\mathrm{x}i} S_{\mathrm{x}j} (| \uparrow_i \downarrow_j \rangle - | \downarrow_i \uparrow_j \rangle) = -\frac{1}{4}$$

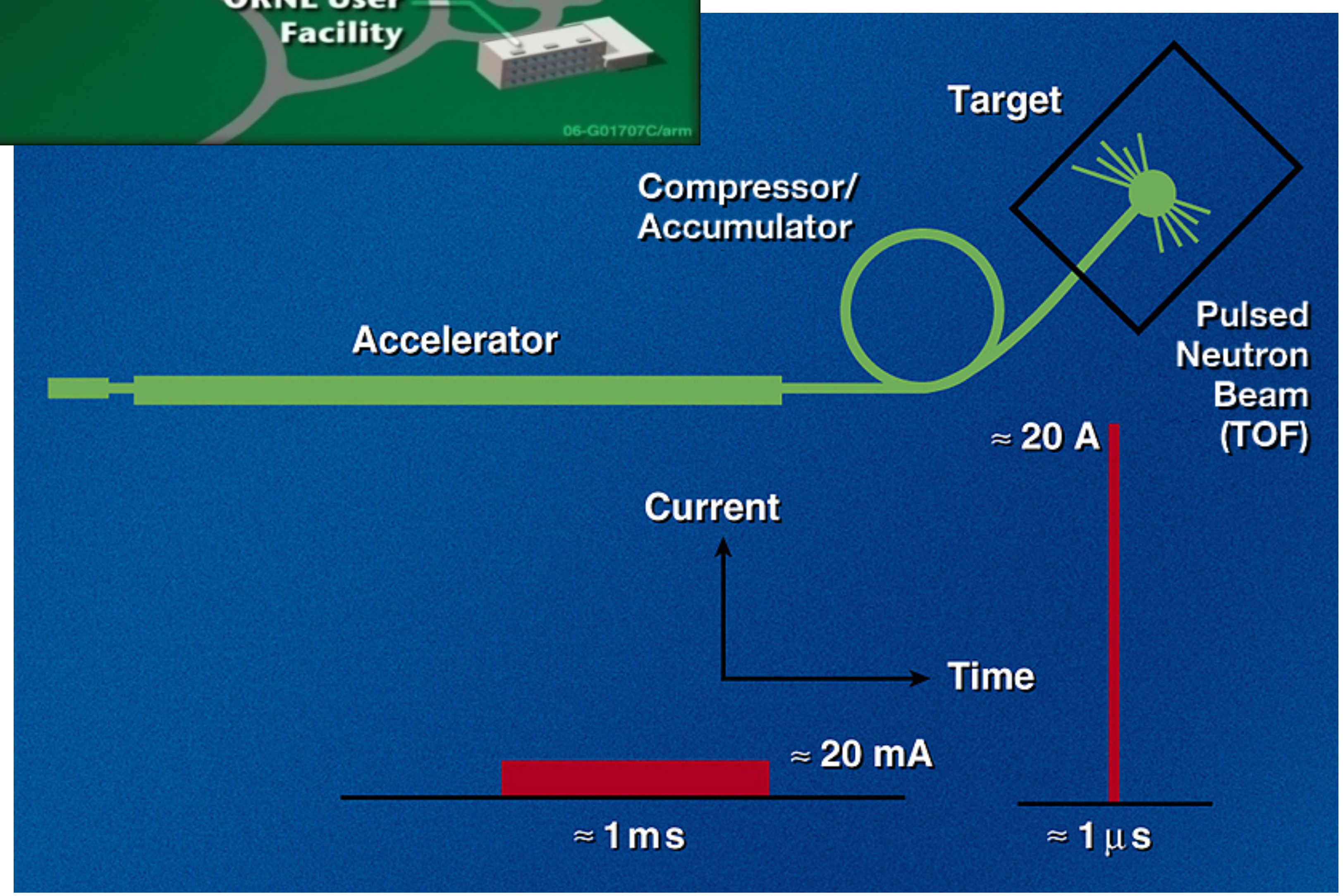
$$(\langle \uparrow_i \downarrow_j | - \langle \downarrow_i \uparrow_j |) S_{\mathrm{y}i} S_{\mathrm{y}j} (| \uparrow_i \downarrow_j \rangle - | \downarrow_i \uparrow_j \rangle) = -\frac{1}{4}$$

$$\langle \uparrow_i \downarrow_j | \, H \, | \uparrow_i \downarrow_j \rangle = -\frac{1}{4} J$$

$$(\langle \uparrow_i \downarrow_j | - \langle \downarrow_i \uparrow_j |) H (| \uparrow_i \downarrow_j \rangle - | \downarrow_i \uparrow_j \rangle) = -\frac{3}{4} J$$


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

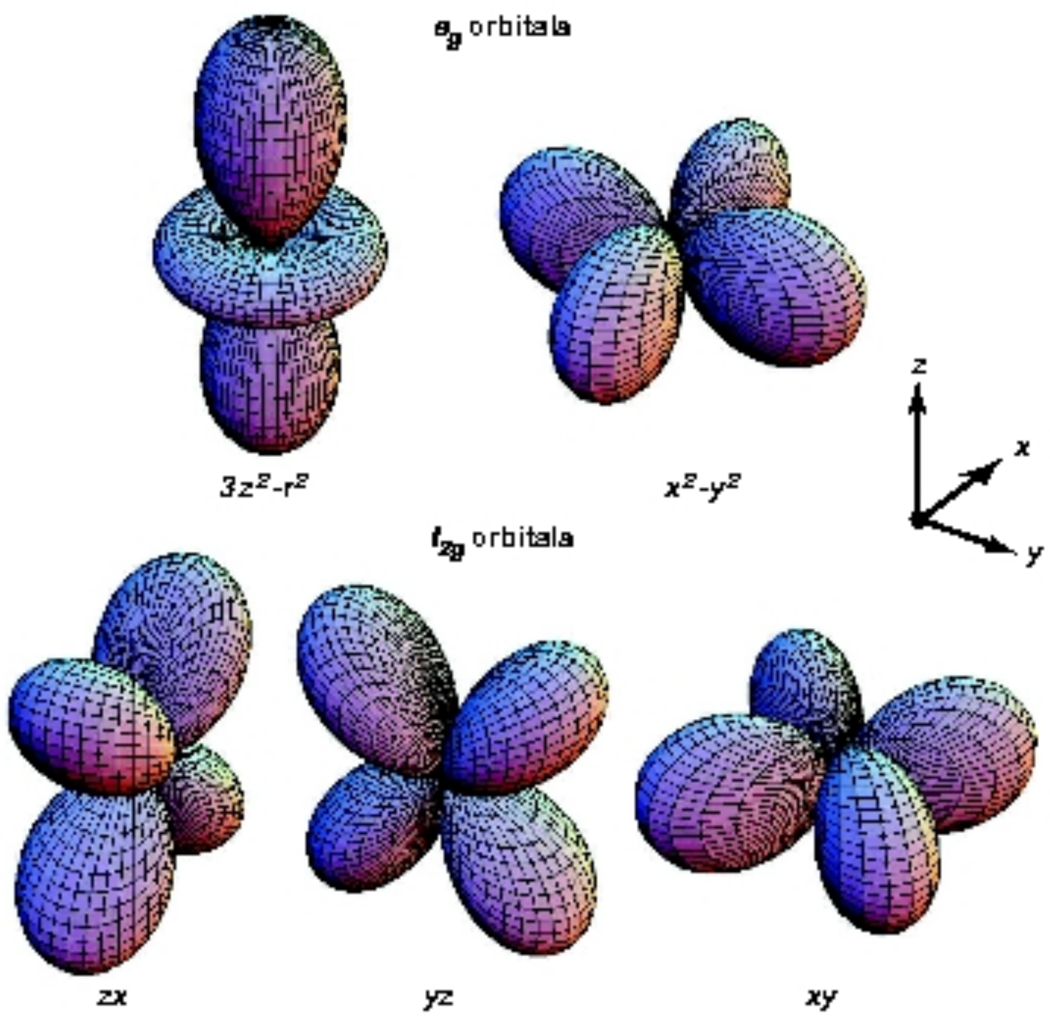

Front-End Building
Klystron Building
Linac Tunnel
Ring
Target
Future Target Building
Central Helium Liquefaction Building
Radio-Frequency Facility
Support Buildings
Central Laboratory and Office Complex
Joint Institute for Neutron Sciences
Center for Nanophase Materials Sciences
ORNL User Facility
06-G01707C/arm

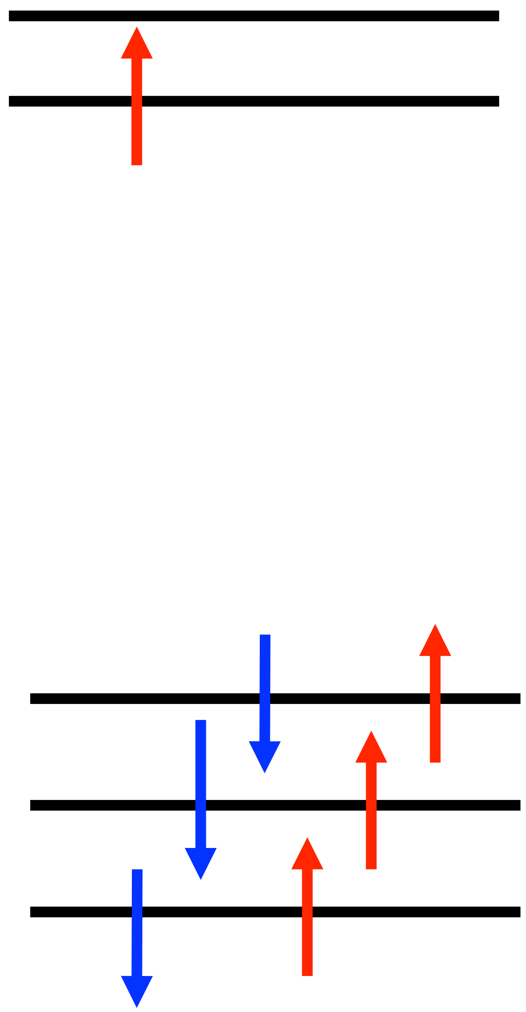

Target
Compressor/ Accumulator
Accelerator
Pulsed Neutron Beam (TOF)
≈ 20 A
Current
Time
≈ 20 mA
≈ 1 ms
≈ 1 μs

Where do quantum spins come from?

$3d^7$

big CEF effect: low spin state

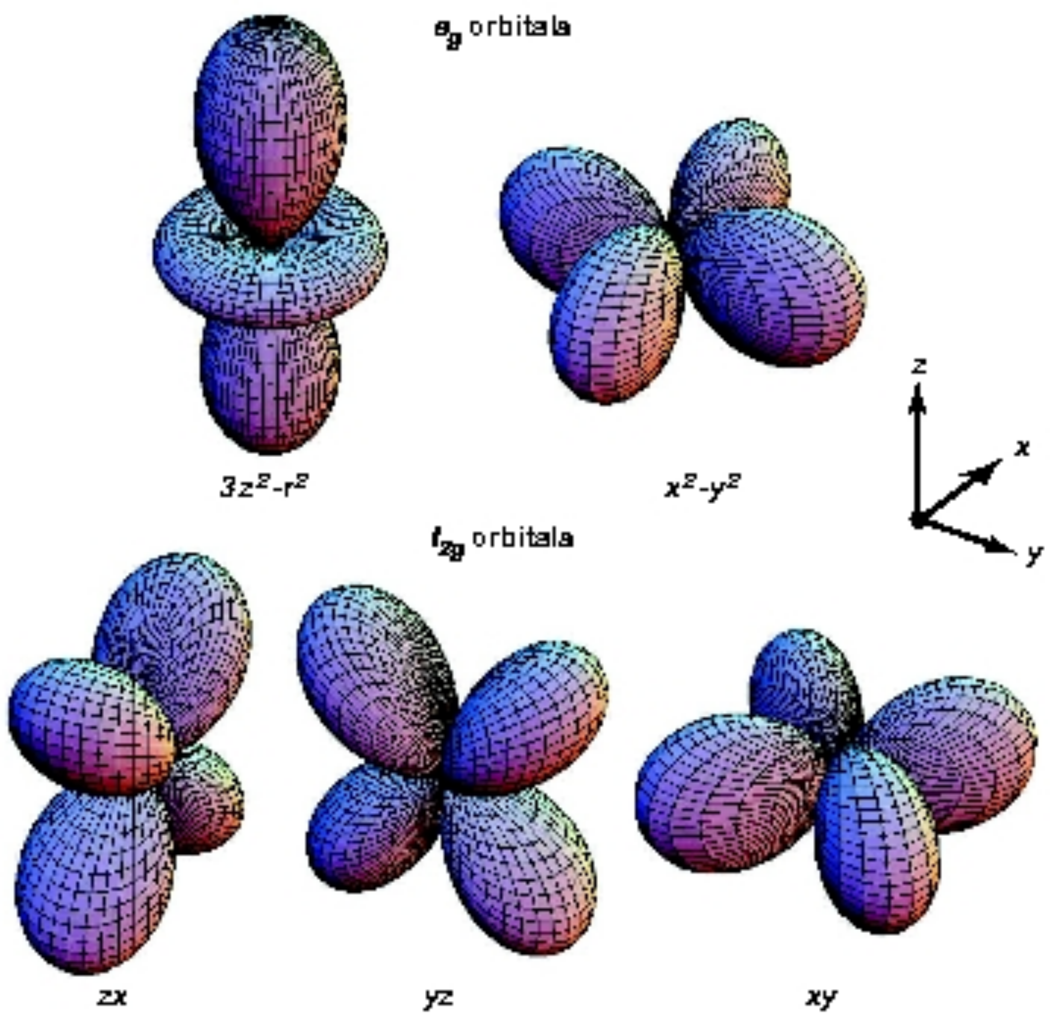


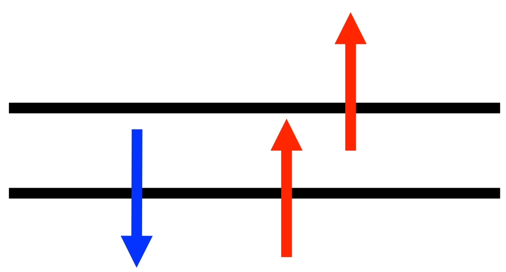


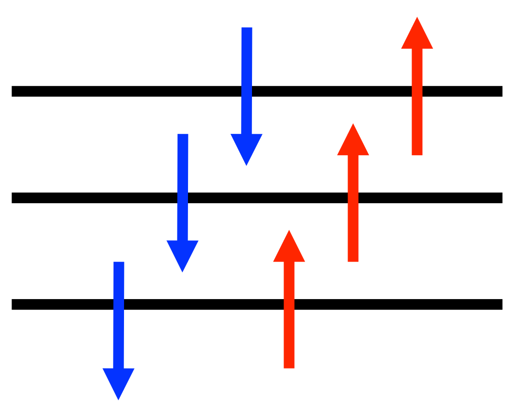
Where do quantum spins come from?

$3d^9$

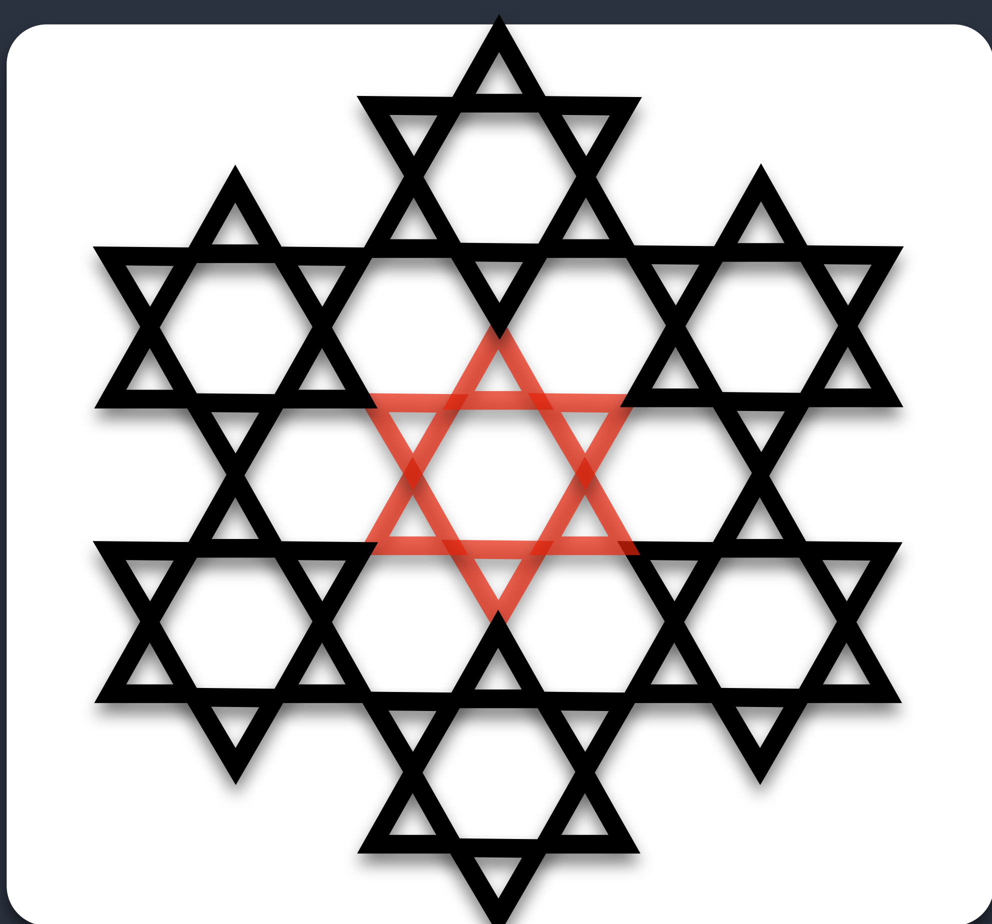
CEF → orbitally quenched orbitals + Hund's rules

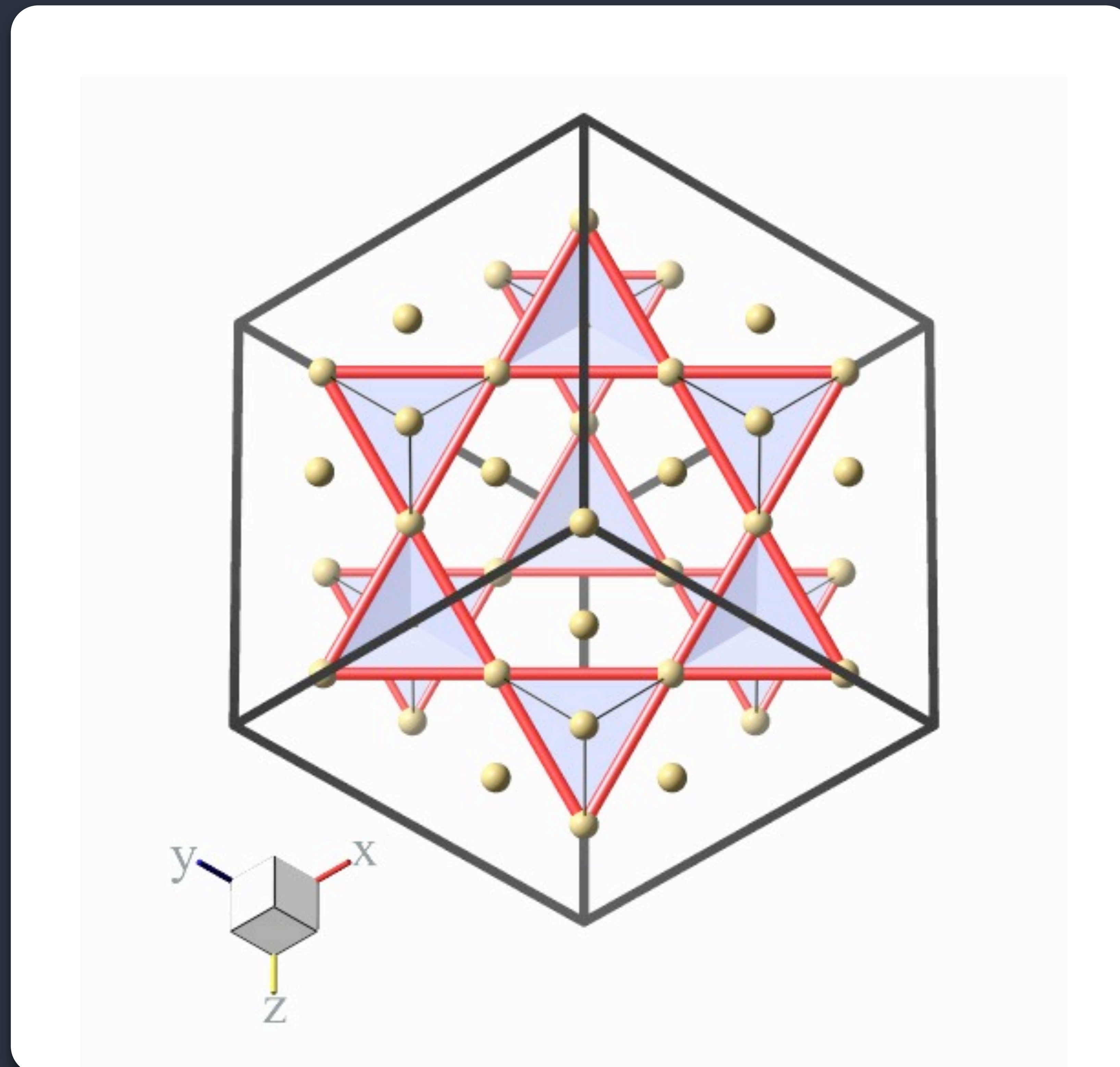




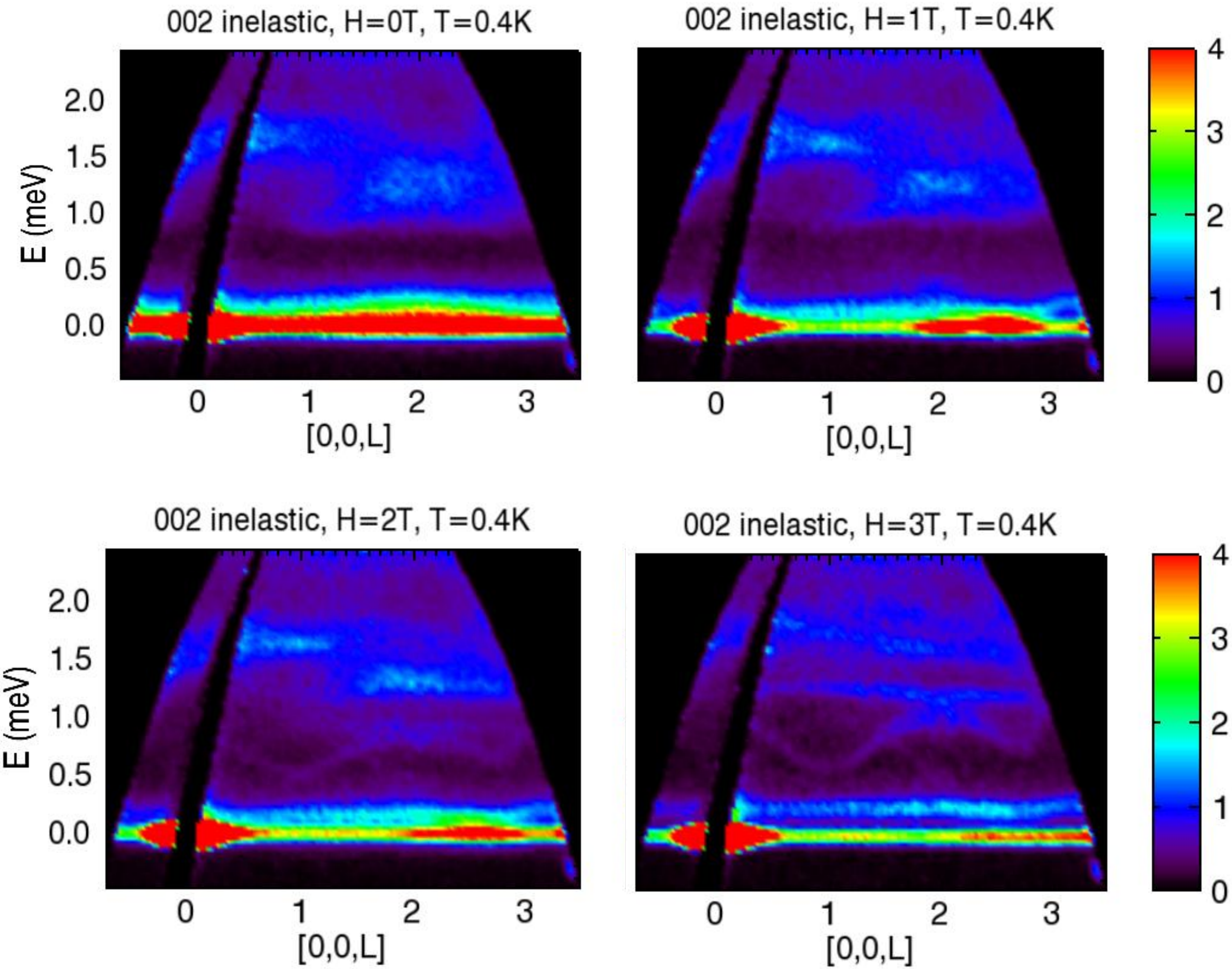


Pyrochlore Lattice



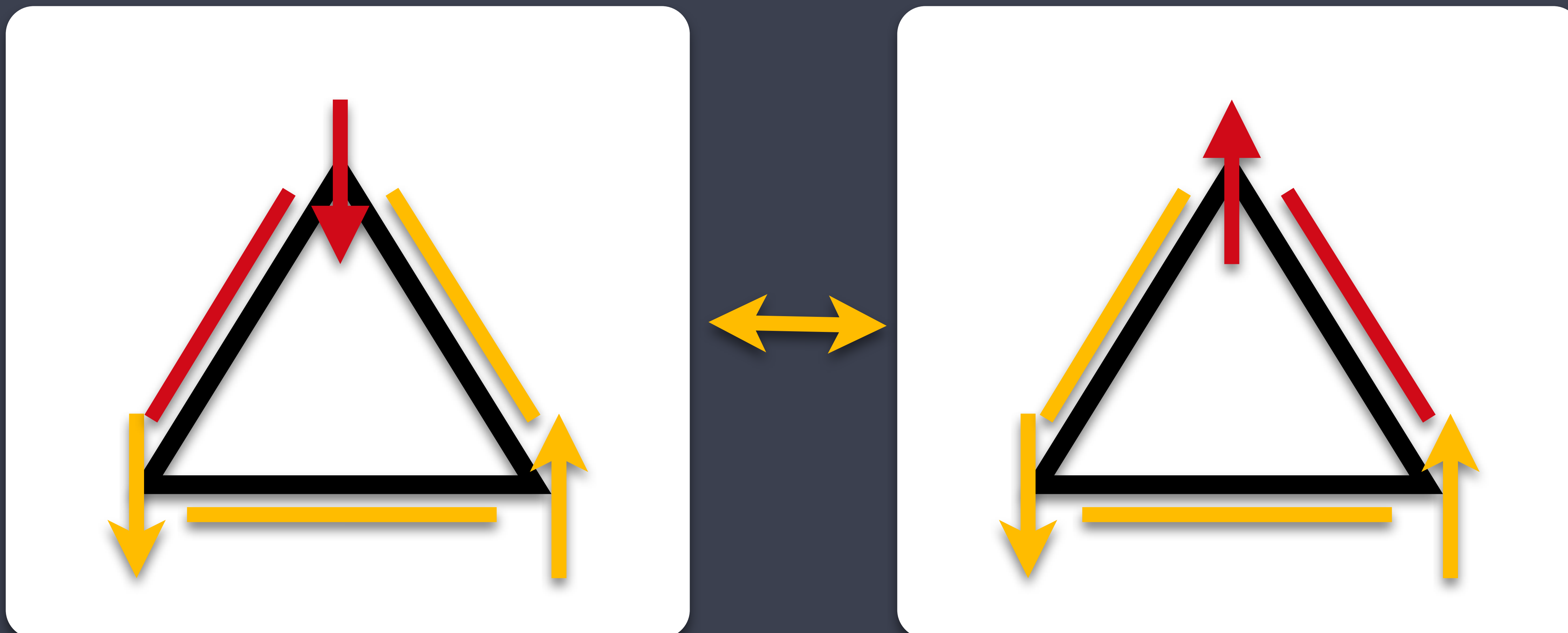


The (111) planes of the Pyrochlore structure are interleaved Kagome and triangular planes

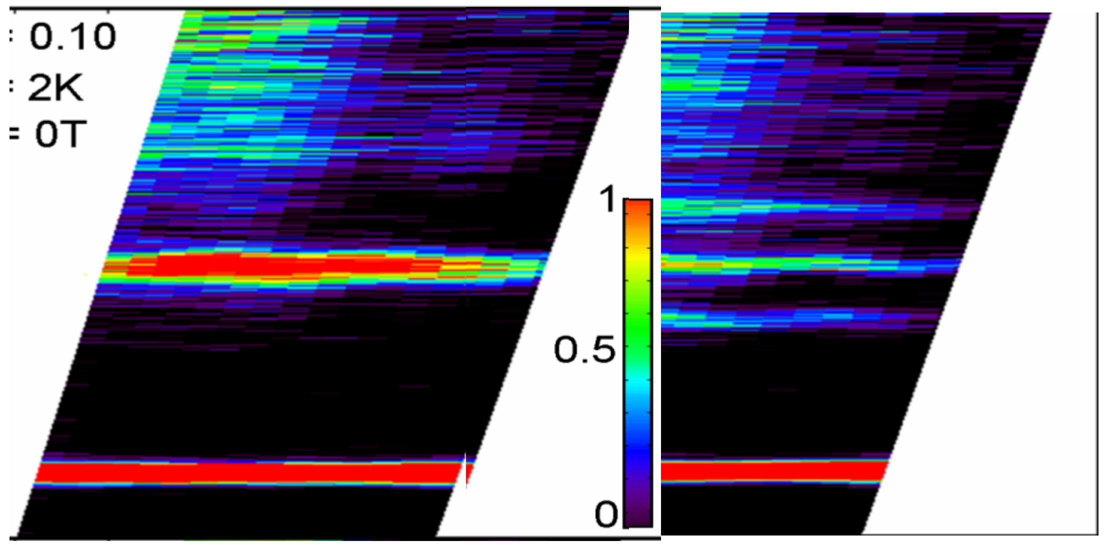


002 inelastic, H=0T, T=0.4K
002 inelastic, H=1T, T=0.4K
002 inelastic, H=2T, T=0.4K
002 inelastic, H=3T, T=0.4K
E (meV)
2.0
1.5
1.0
0.5
0.0
[0,0,L]
0
1
2
3
4

Geometric Frustration in 2D



Sometimes the system will fluctuate between these degenerate states, increasing entropy: NO Long Range Order



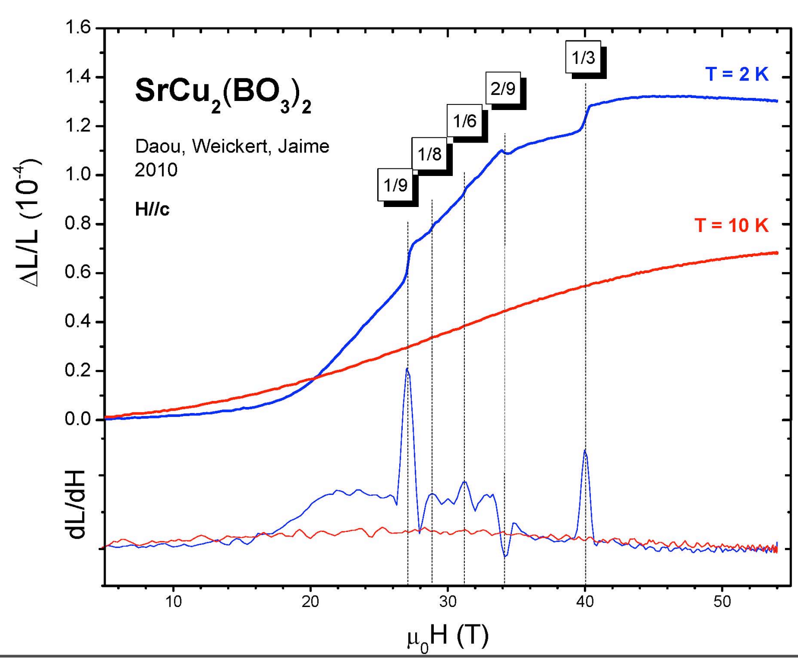
0.10
2K
0T
1
0.5
0

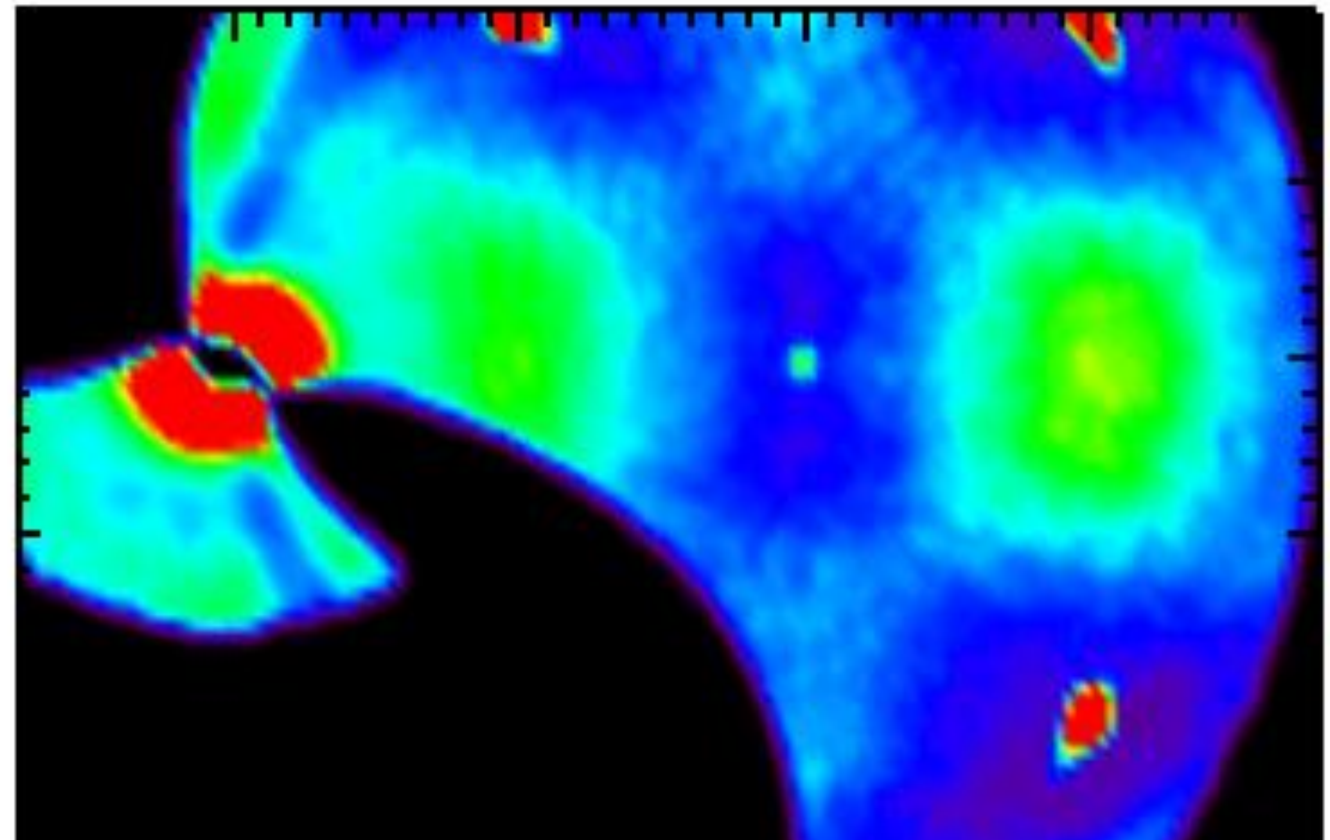
Time-of-flight neutron scattering and 2D spin correlations in 3D crystals

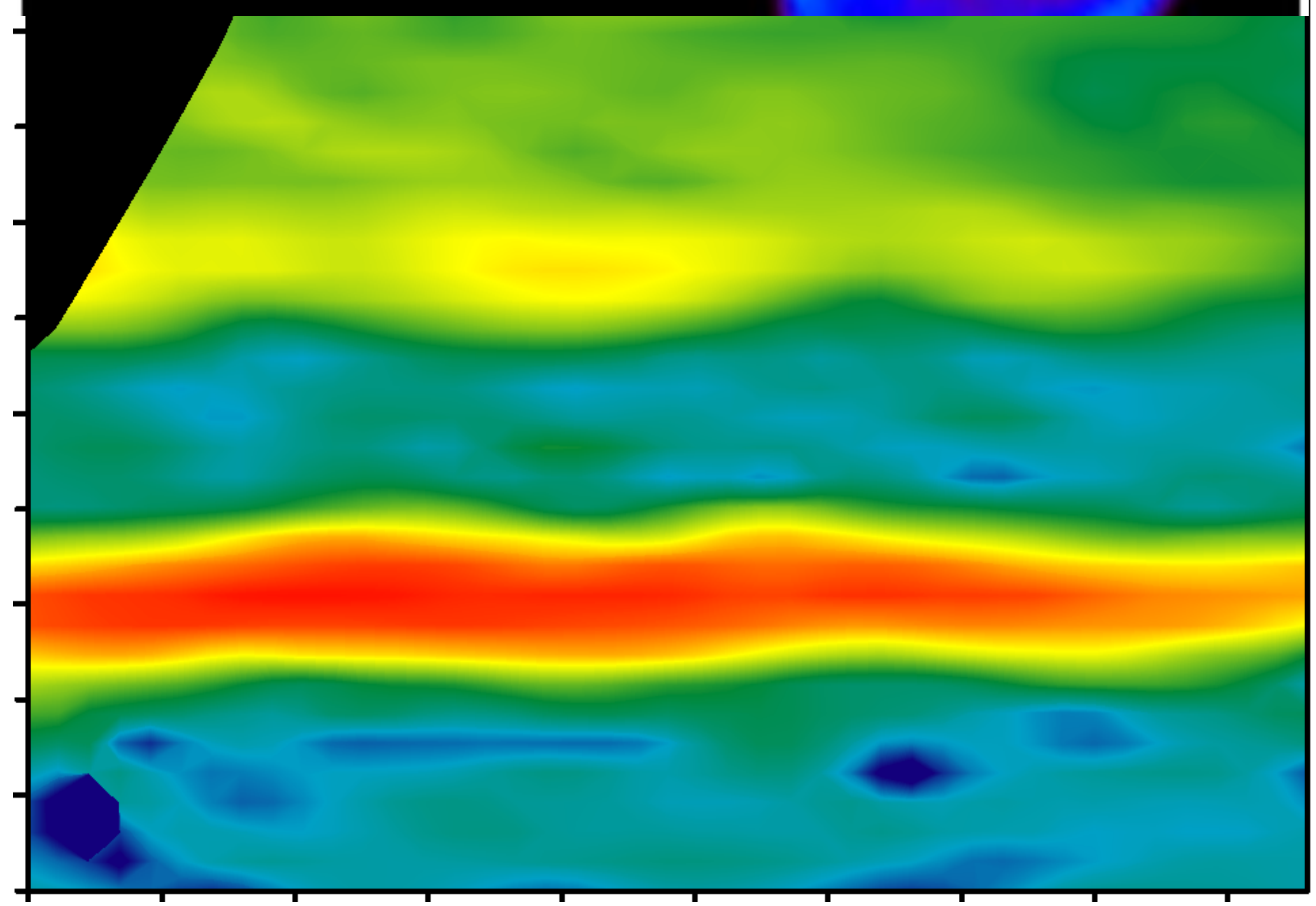
Bruce D Gaulin
BIMR
McMaster University

[1]McMaster University
[2]St. Francis Xavier U
[3]NIST
[4]*SNS, ORNL*

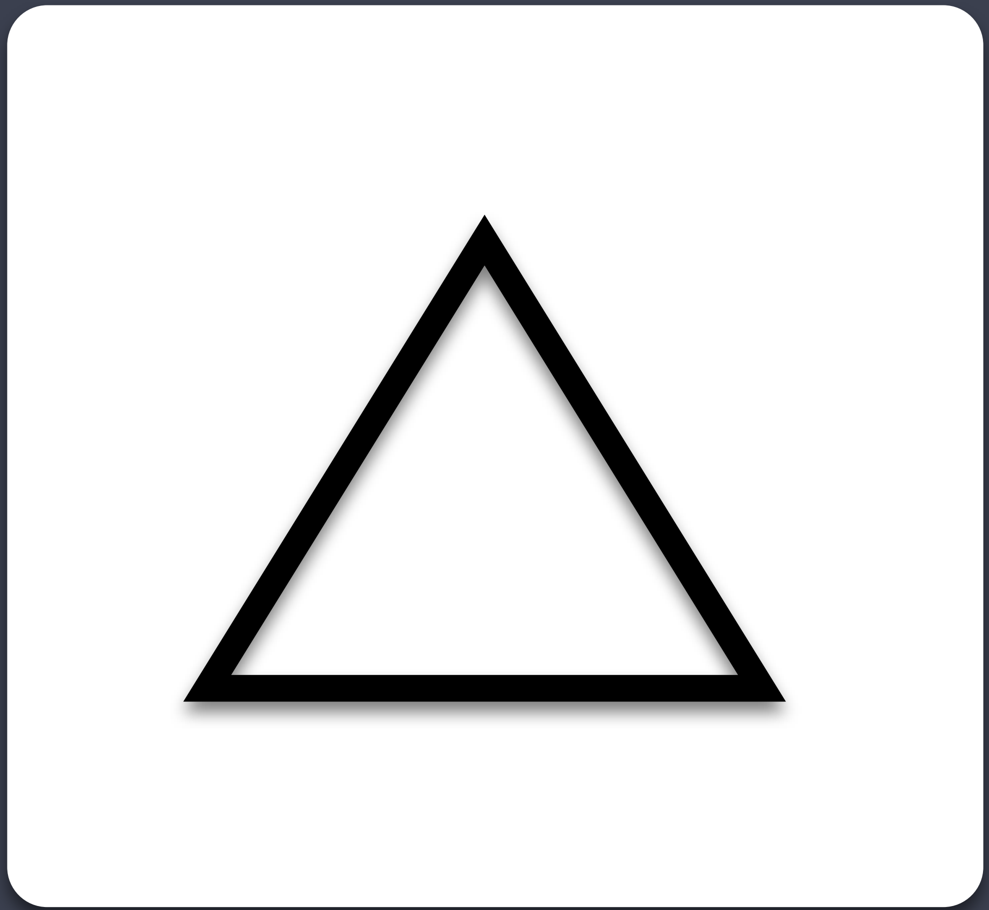
ka [1]



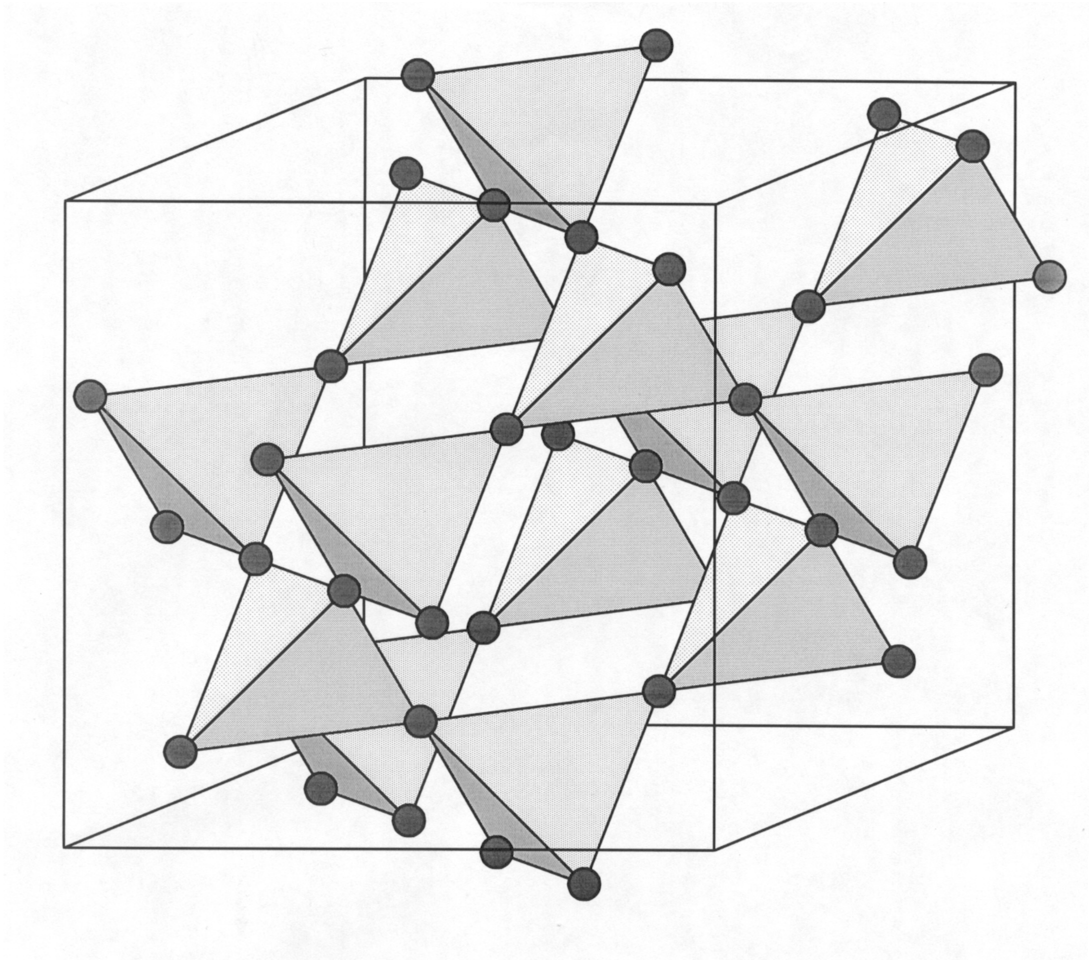




Geometric Frustration in 2D



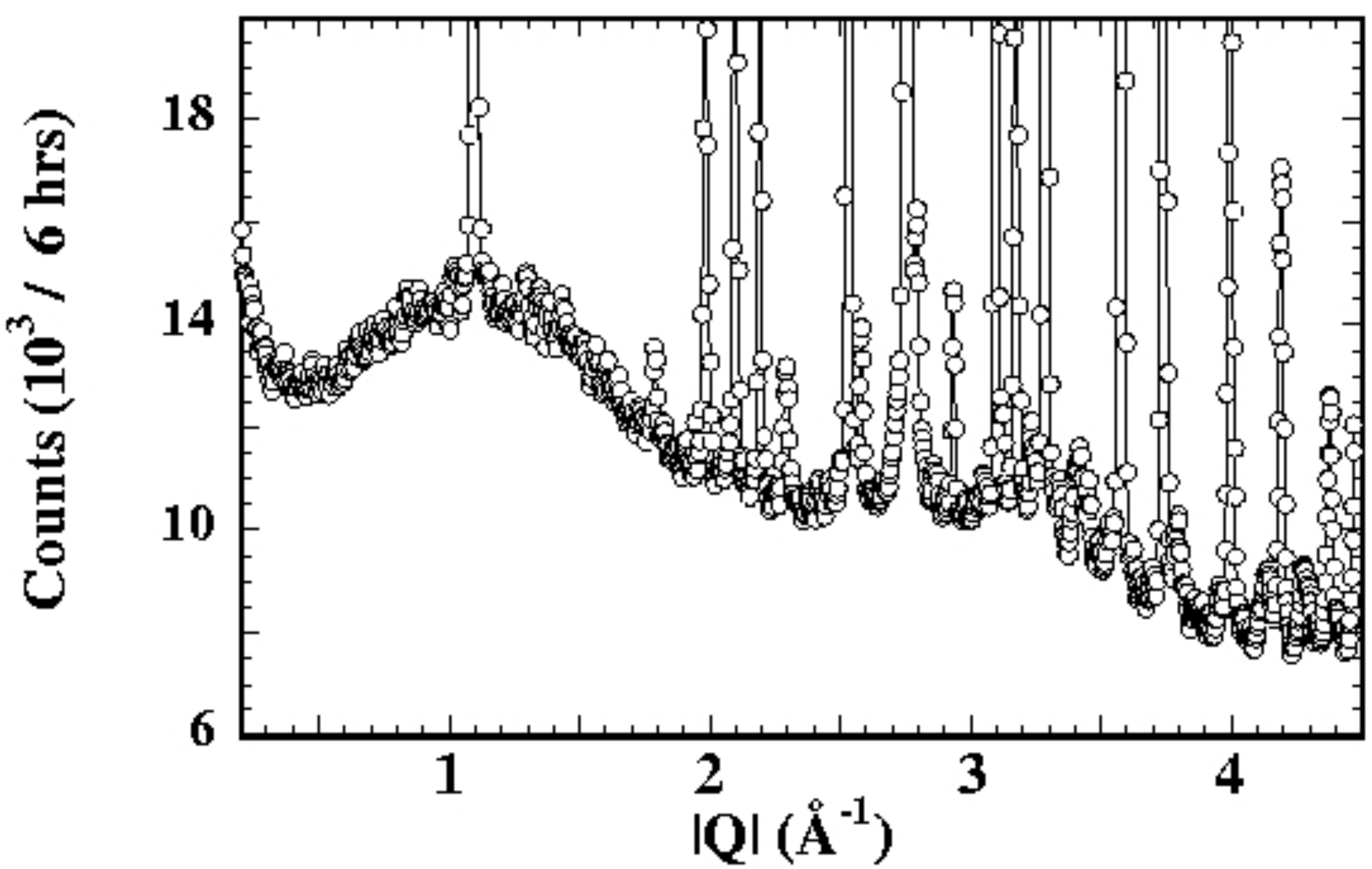
prefer anti-ferromagnetic alignment



Frustration in three dimensions:

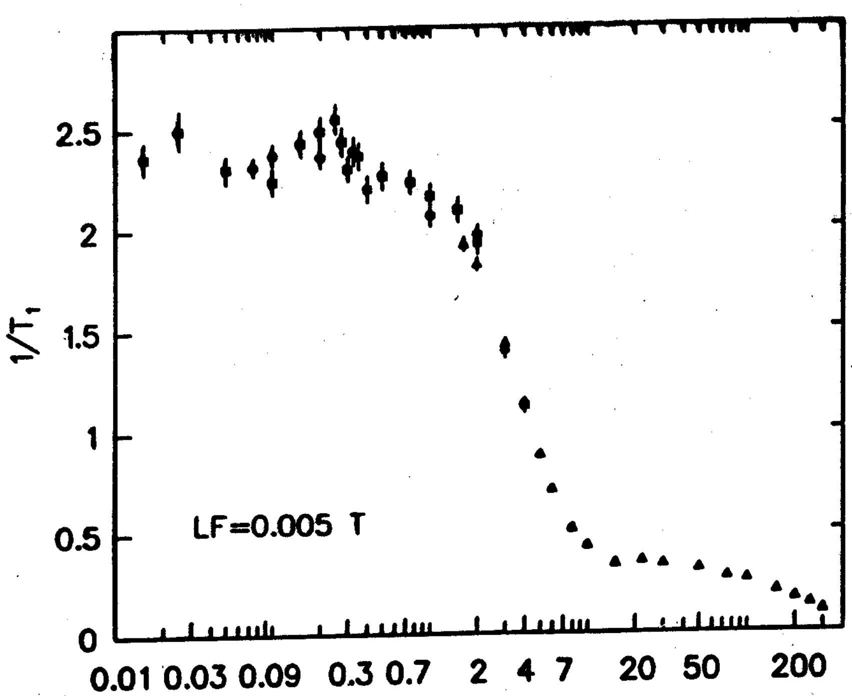
The cubic pyrochlore structure;
A network of corner-sharing tetrahedra

Low temperature powder neutron diffraction from $Tb_2Ti_2O_7$

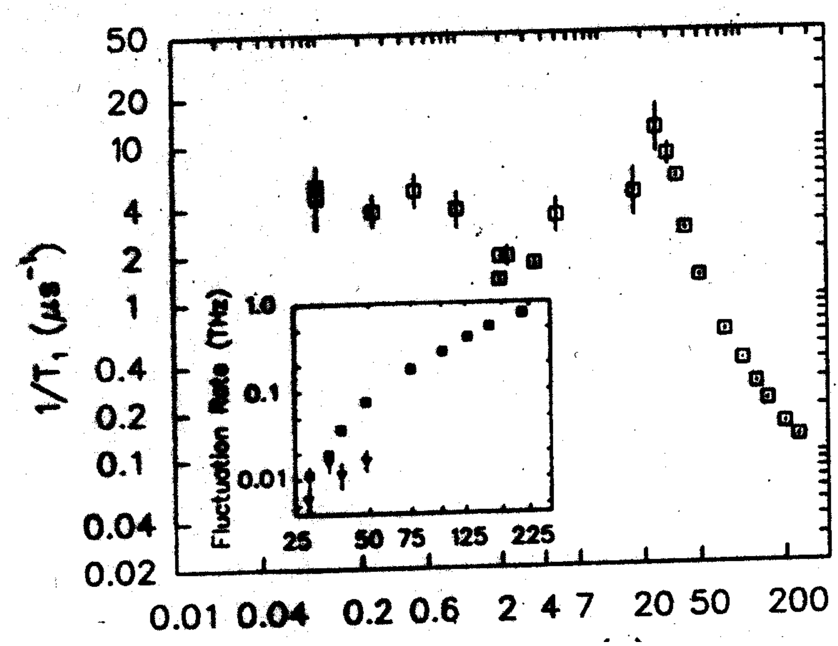


μSR Studies of Magnetic Ground States in:

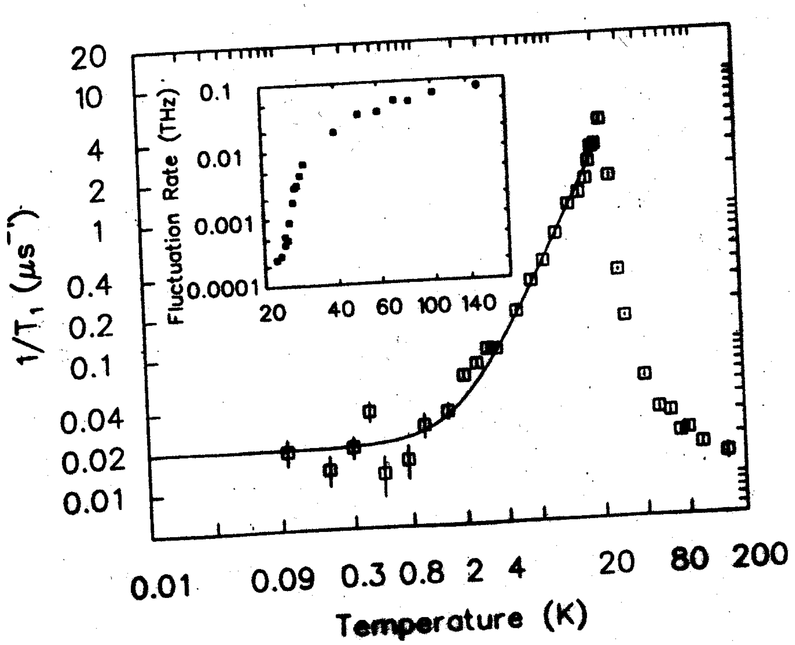
$Tb_2Ti_2O_7$
$Tb_2Mo_2O_7$
$Y_2Mo_2O_7$



$Tb_2Ti_2O_7$: *Spin Liquid*



$Tb_2Mo_2O_7$: *Spin Liquid and Spin Glass*



$Y_2Mo_2O_7$: *Spin Glass*