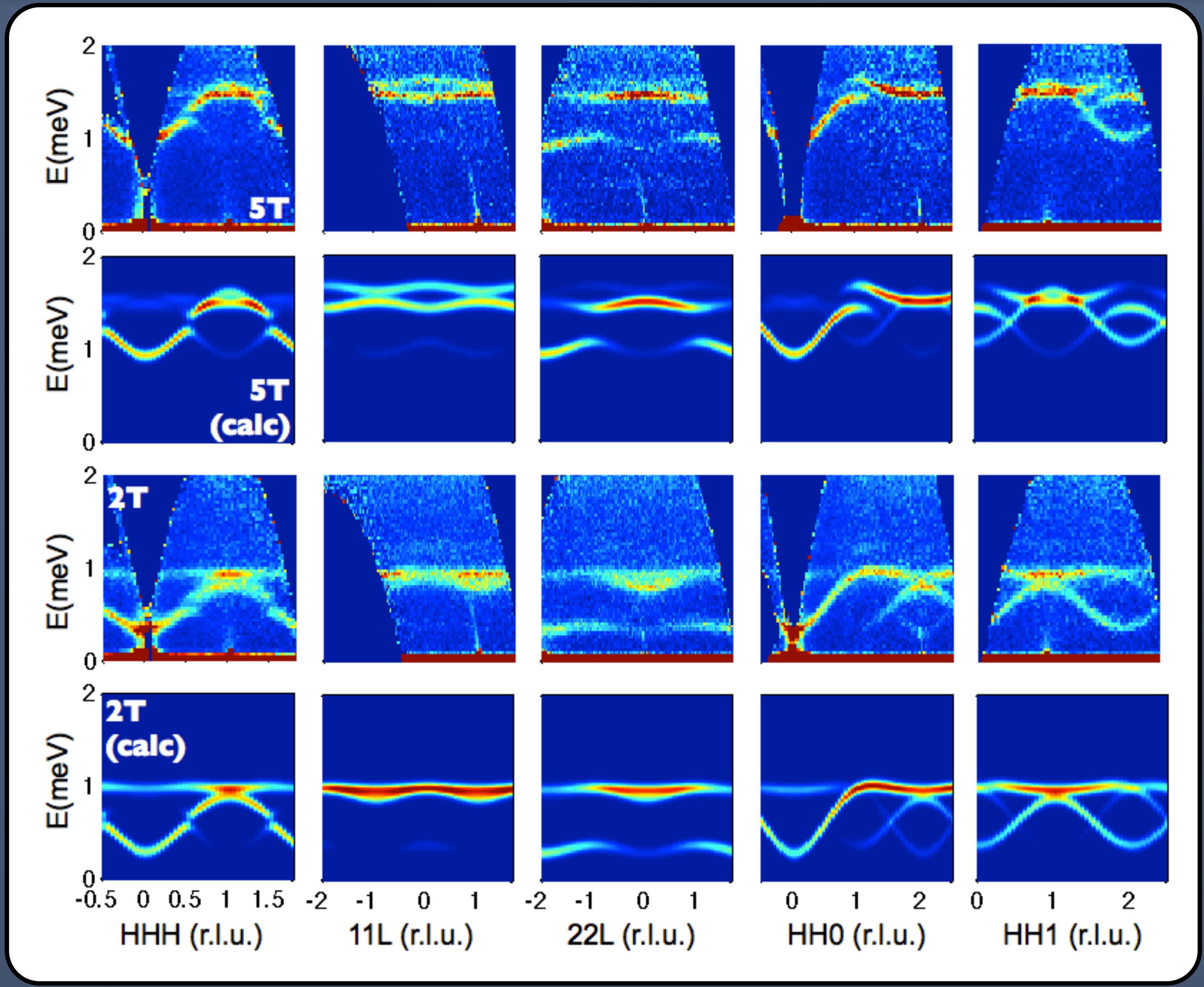
Quantum Ordered & Disordered Phases in XY Pyrochlores $Er_2Ti_2O_7$ and $Yb_2Ti_2O_7$



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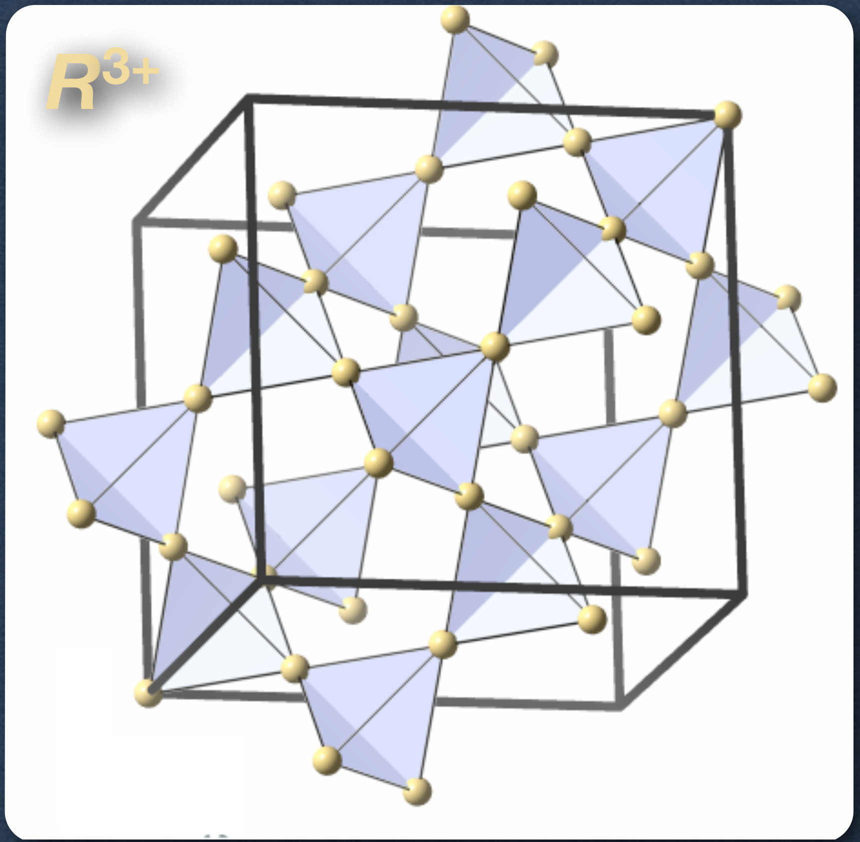
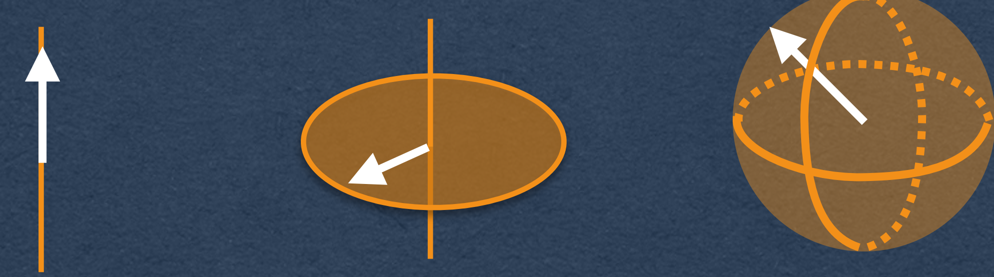
Brockhouse Institute for Materials Research

Real Pyrochlores: playgrounds for frustration

$R_2Ti_2O_7$ “Rare earth titanates”



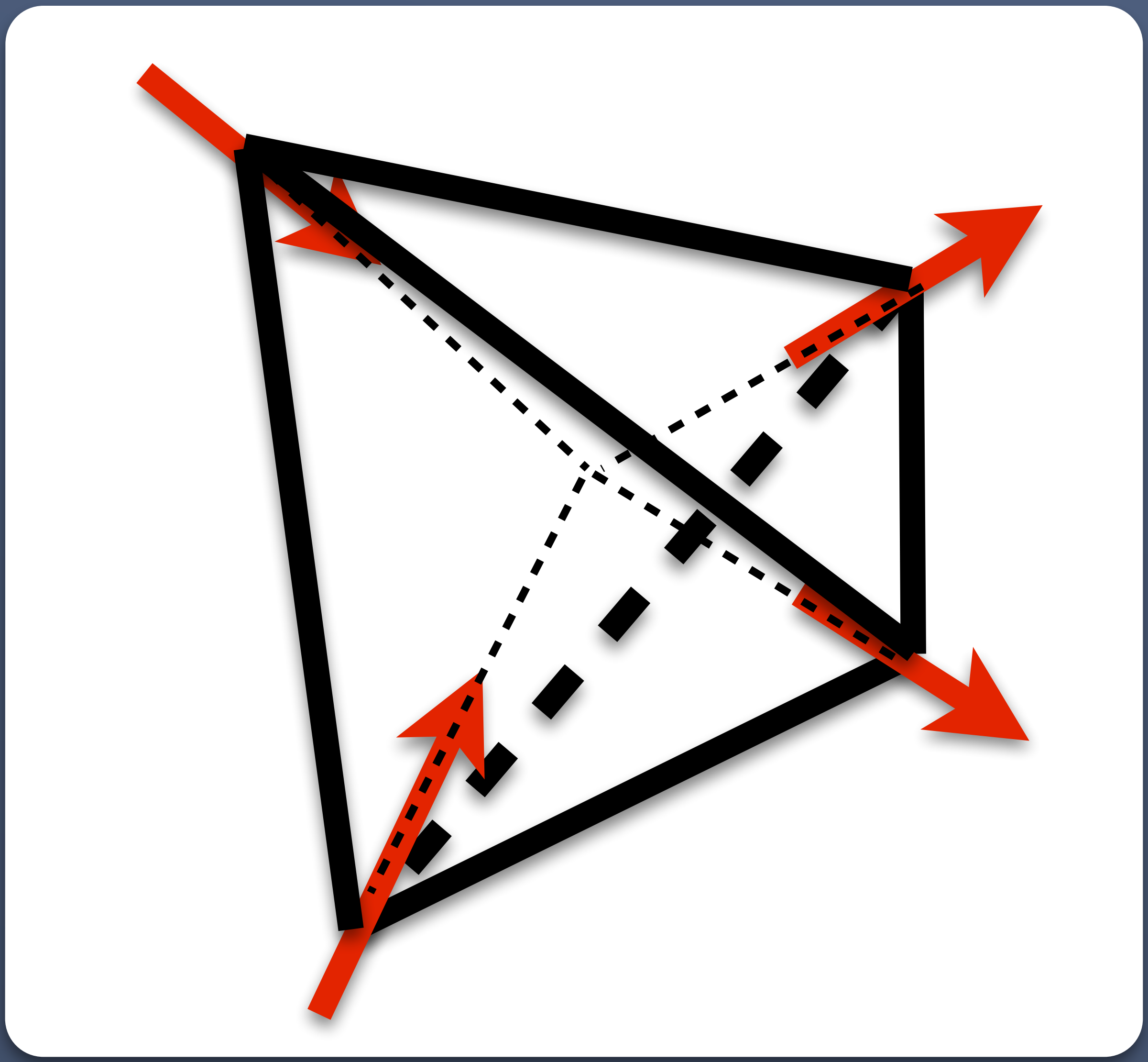
Ising XY Heisenberg

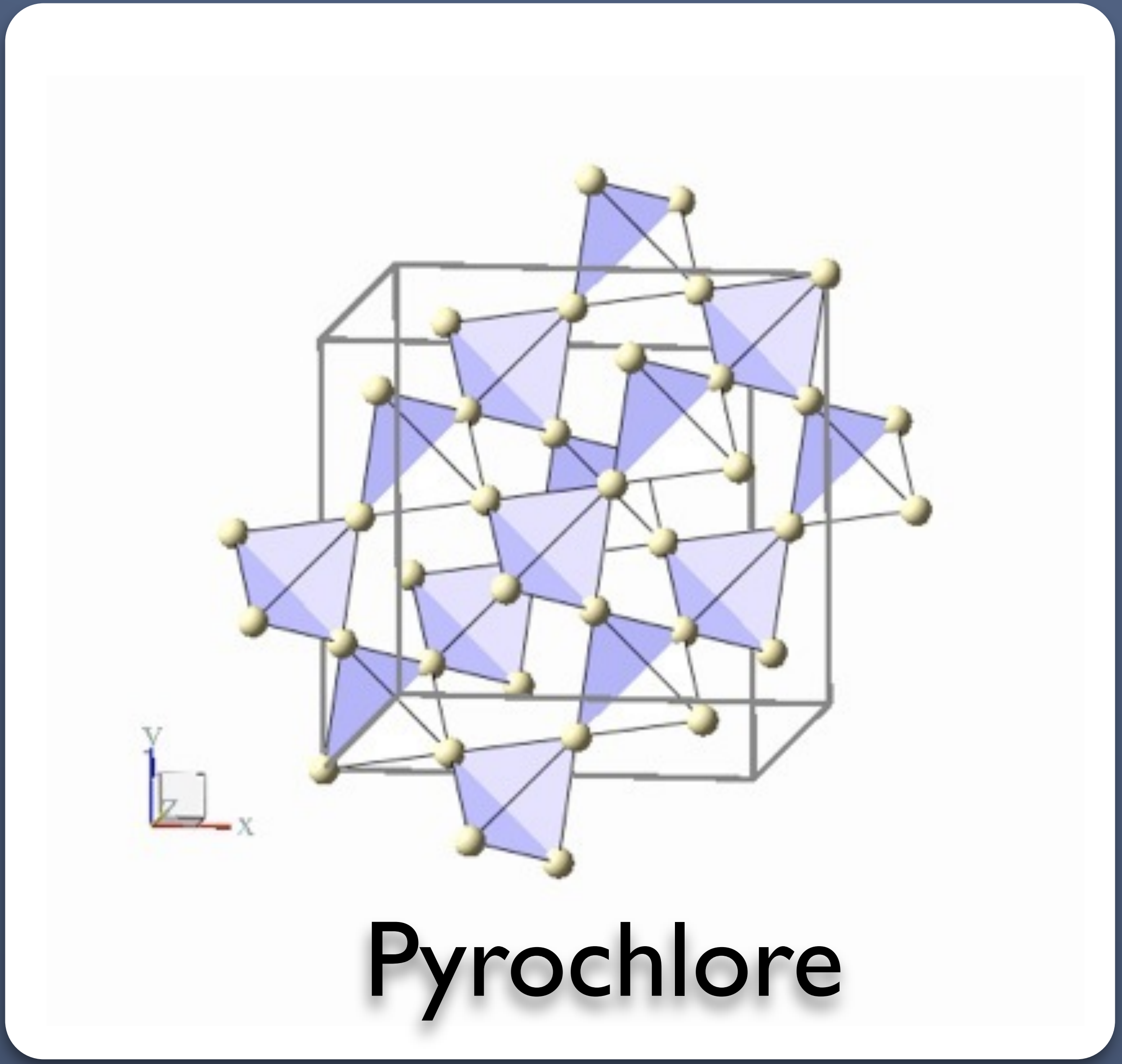


Differences in **Anisotropy** is very important

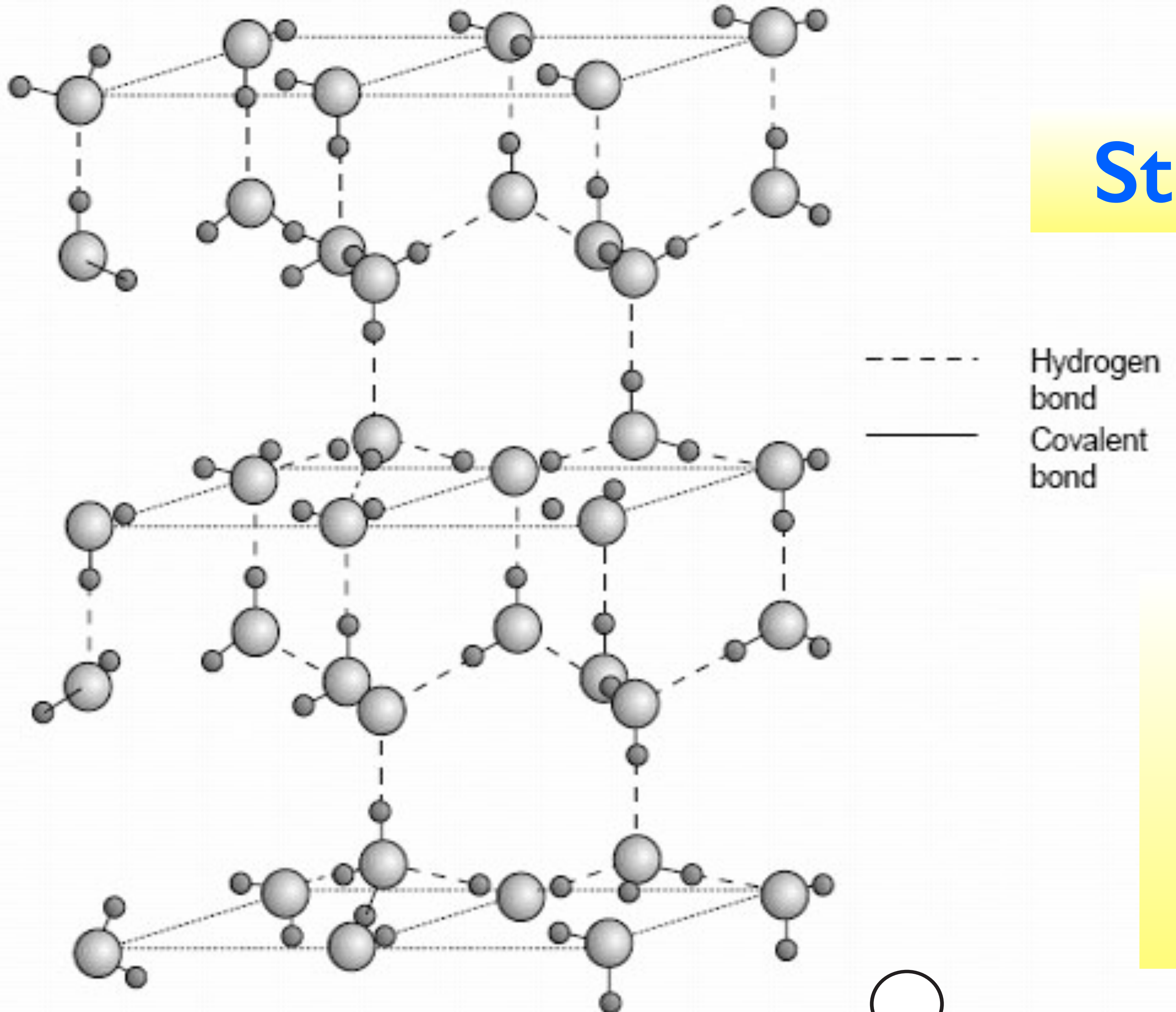
	Single Ion Anisotropy	Interactions	Ground state
Ho, Dy	Ising	FM	spin ice
Tb	Ising	AFM	spin liquid
Gd	Heisenberg	AFM	partial order
Er	XY	AFM	“order by disorder”
Yb	XY	FM	“quantum spin ice”

Geometric Frustration from Tetrahedra





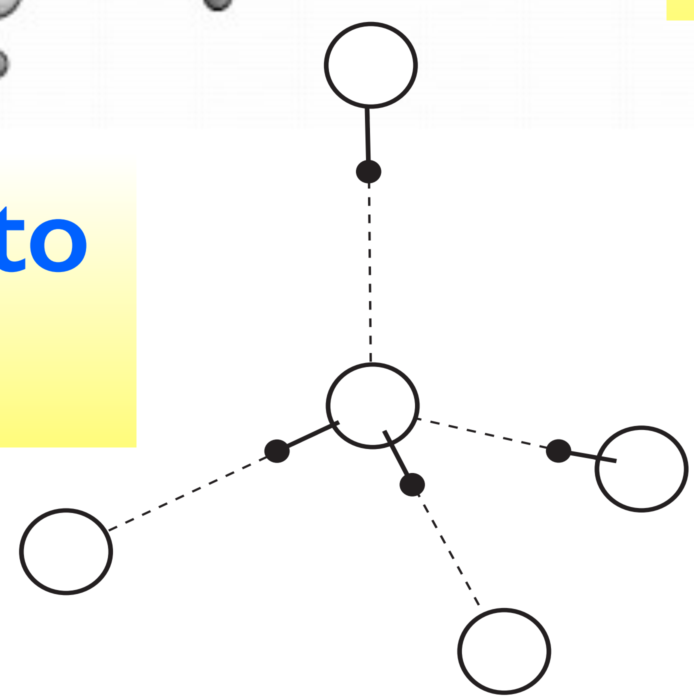
freedom of choice for each tetrahedron leads to a macroscopic degeneracy: **NO Long Range Order**

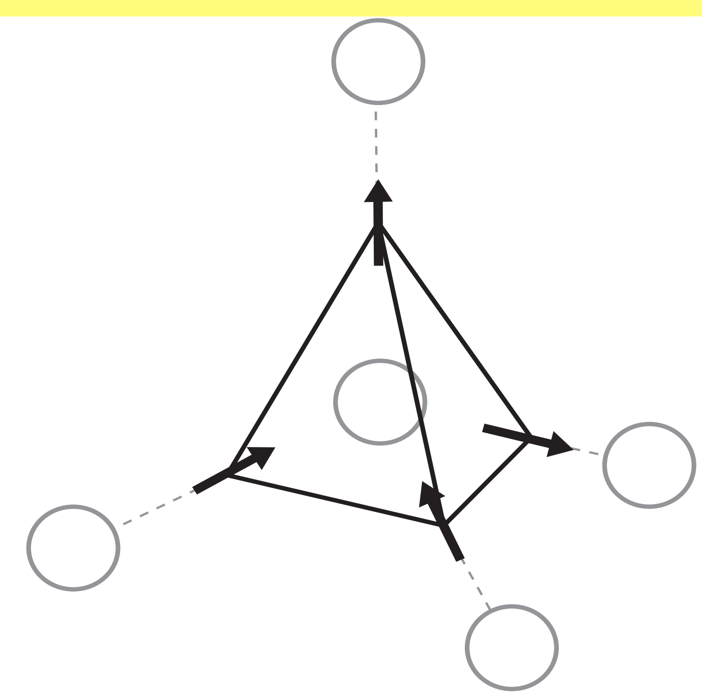


Structure of Ice

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

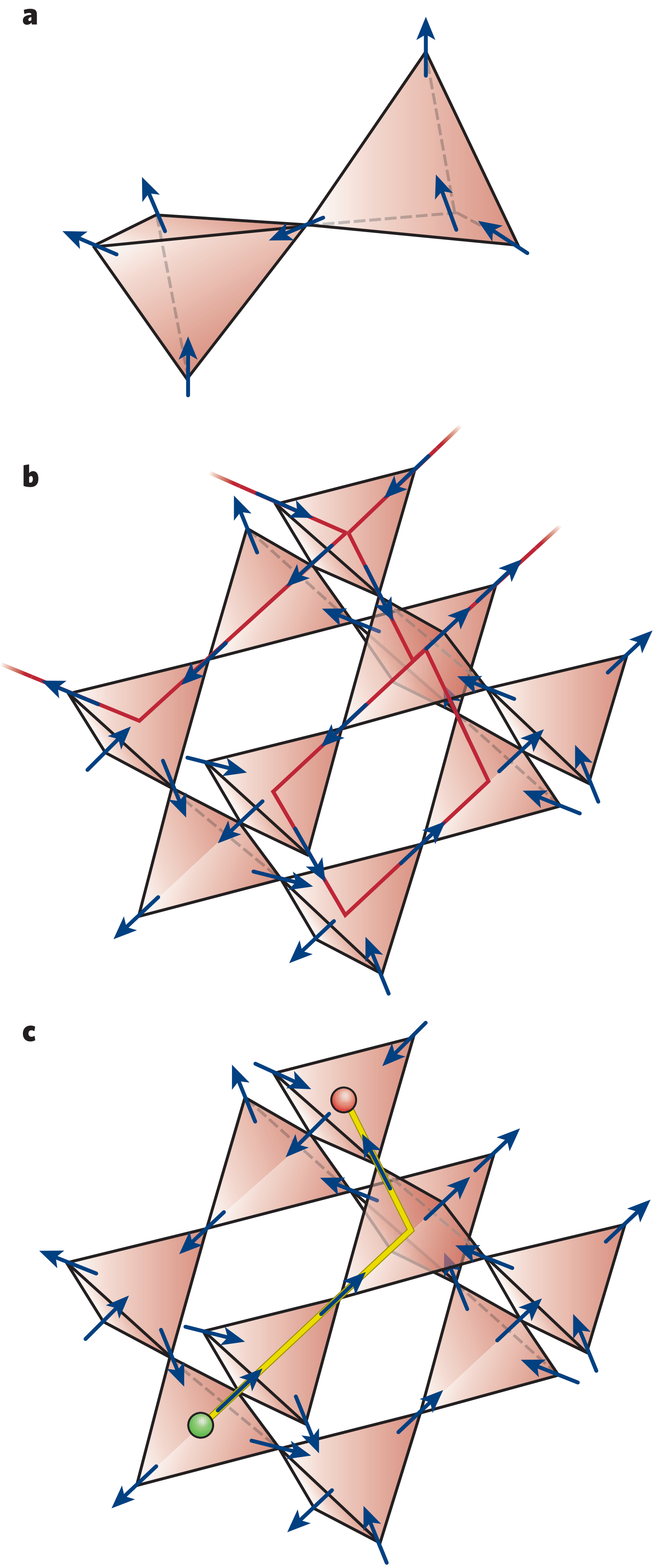
Correspondance to Spin Ice

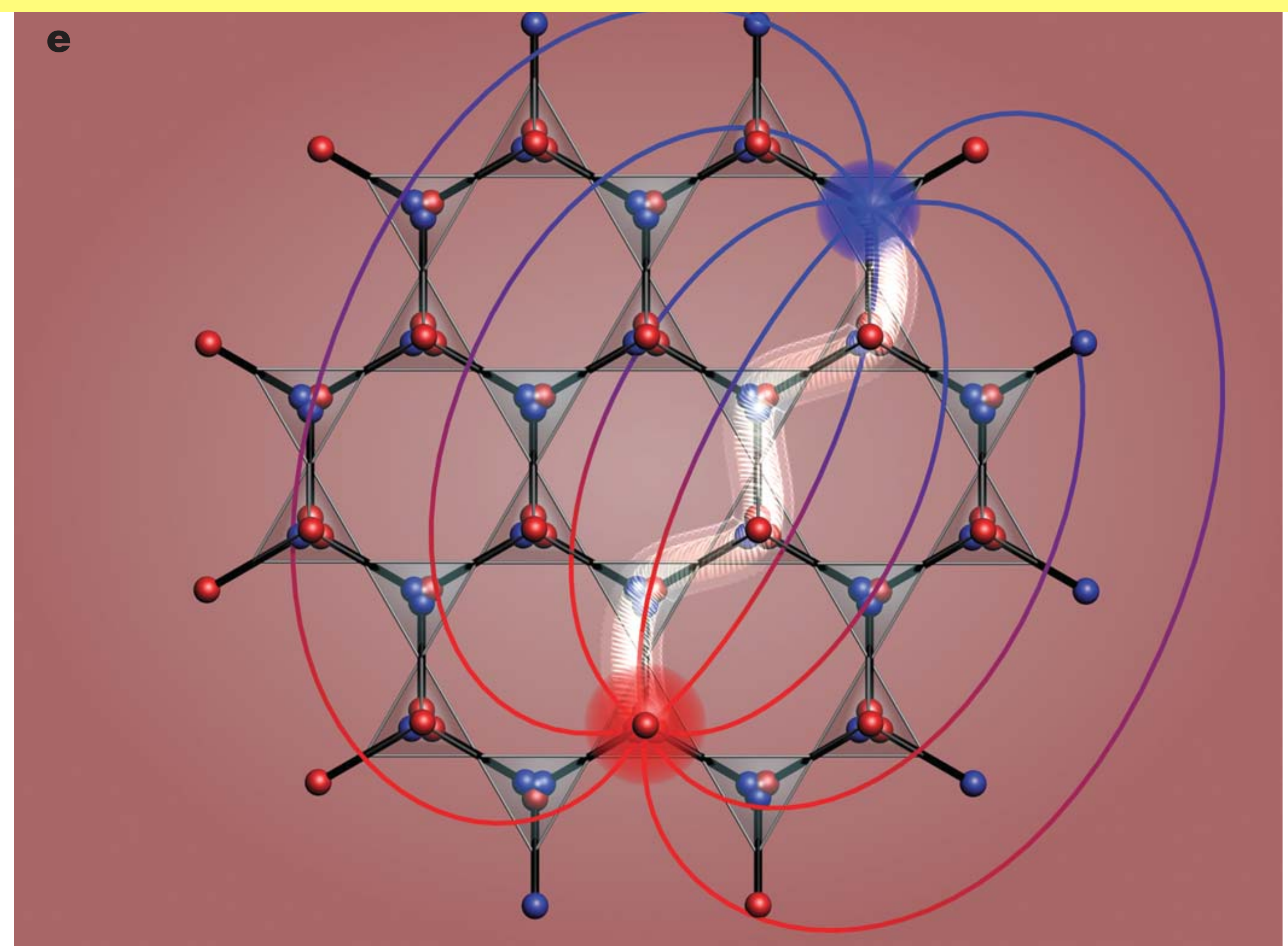




Spin Ice

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

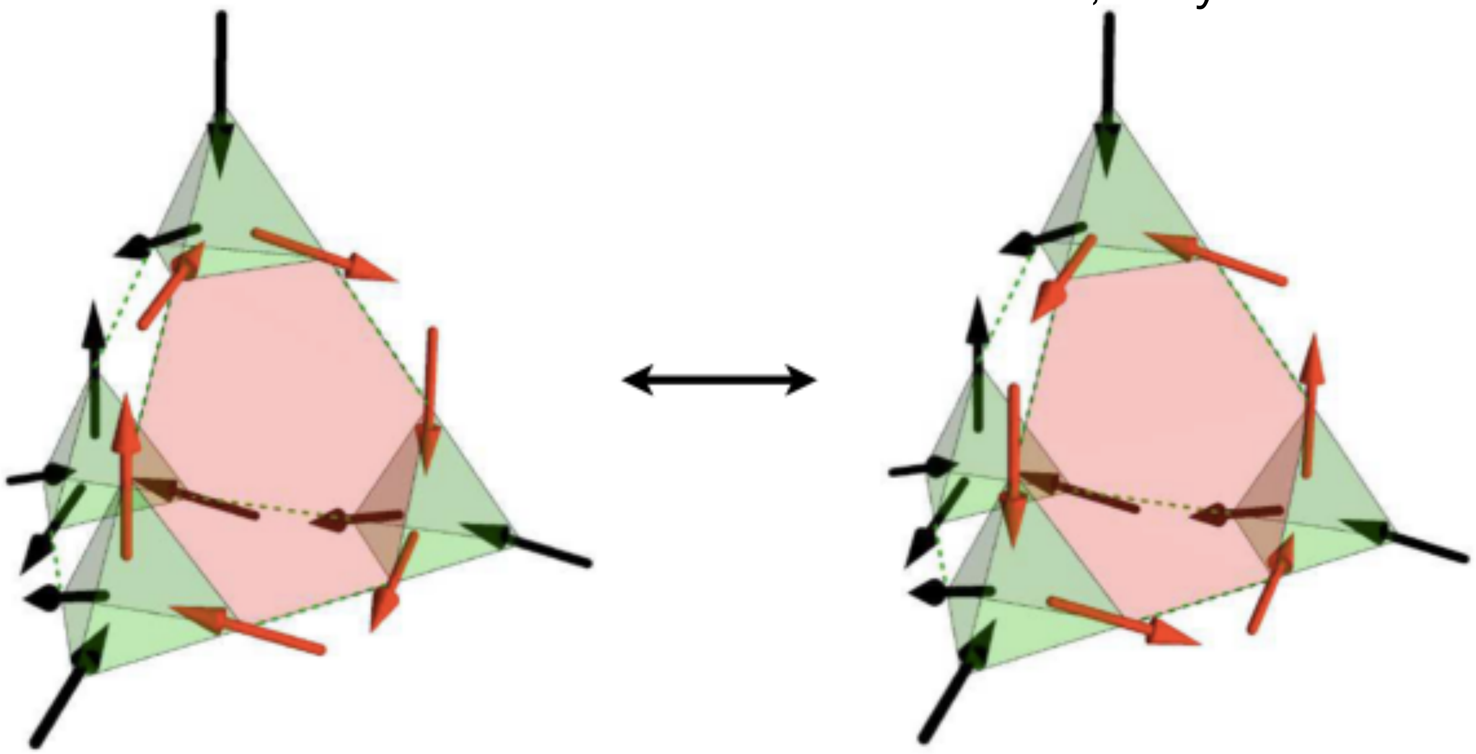




C. Castelnovo, R. Moessner, and S.L. Sondi, Nature, 451, 43 (2007)
L. Balents, Nature, 464, 199 (2010)

"Quantum" Spin Ice

O. Benton et al, Phys. Rev. B **86**, 2012



$$\vec{\nabla} \cdot \vec{B} = \rho_m$$

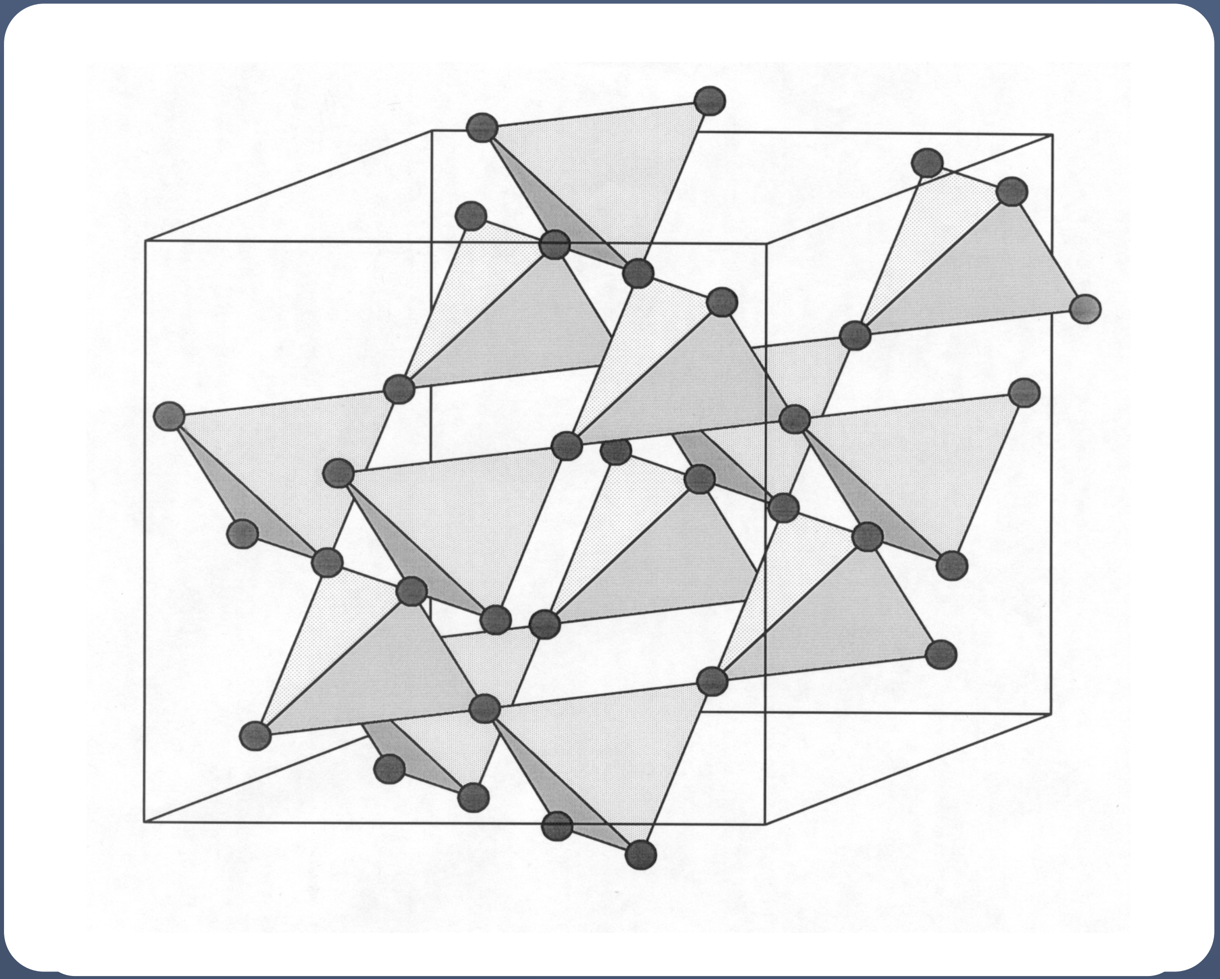
$$\vec{B} = \vec{\nabla} \times \vec{A}$$

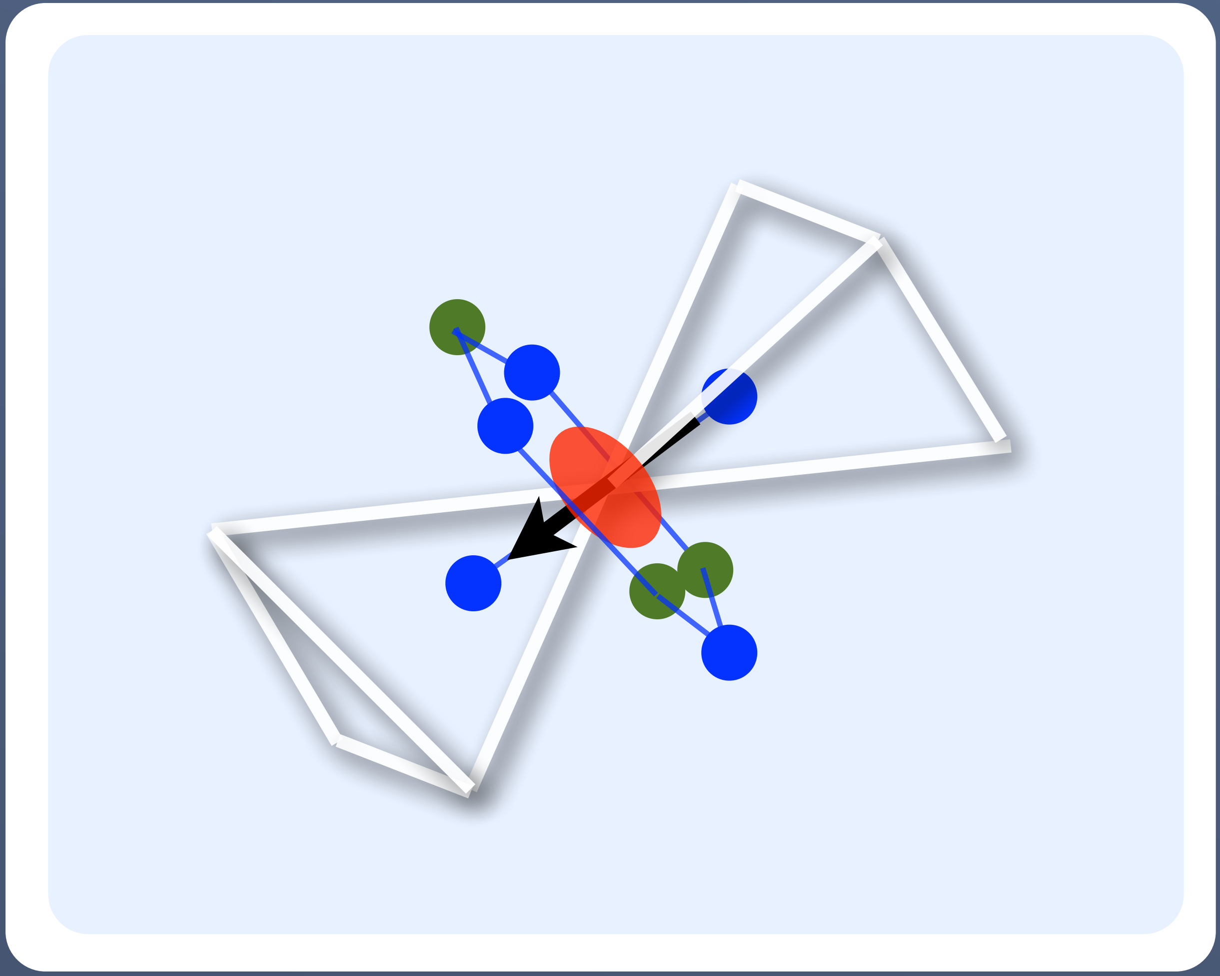
$$\vec{E} = -\frac{\partial \vec{A}}{\partial t}$$

$$\mathcal{H}_{\mathrm{U(1)}} = \frac{\mathcal{U}}{2} \sum_{\langle \mathbf{rr'} \rangle} [(\nabla_{\bigcirc} \times \mathcal{A})_{\mathbf{rr'}}]^2 + \frac{\mathcal{K}}{2} \sum_{\langle \mathbf{ss'} \rangle} \mathcal{E}^2_{\mathbf{ss'}}$$

- Can tunnel between ice rules states
- Introduces *fluctuations* in the gauge field
  - **Electric monopoles —** coherent, propagating wavepacket of ice configurations
  - **Magnetic monopoles —** violate ice rules, i.e. 3-in 1-out
  - **Gauge photons —** transverse fluctuations of gauge field

Crystal Field Environment at the RE Site





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

Crystal Field Effects

$Yb_2Ti_2O_7$

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

680K

$g_{||} = 1.78$

$g_{\perp} = 4.28$

$4f^{13}$ $J = 7/2$

$Er_2Ti_2O_7$

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

76K

$g_{||} = 2.32$

$g_{\perp} = 6.80$

$4f^{11}$ $J = 15/2$

$Dy_2Ti_2O_7$

Bertin et al., J. Phys: CM, 24, 256003, 2012

550K

500K

450K

300K

$g_{||} \sim 10$

$g_{\perp} \sim 0$

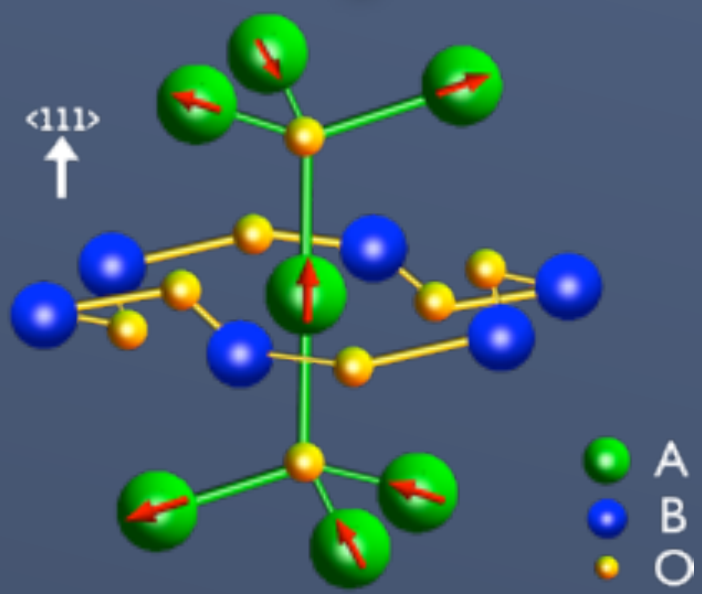
$4f^{9}$ $J = 15/2$

Crystal Field Effects:
How do you get $S_{effective}$=1/2 from J=big, ie J=15/2?

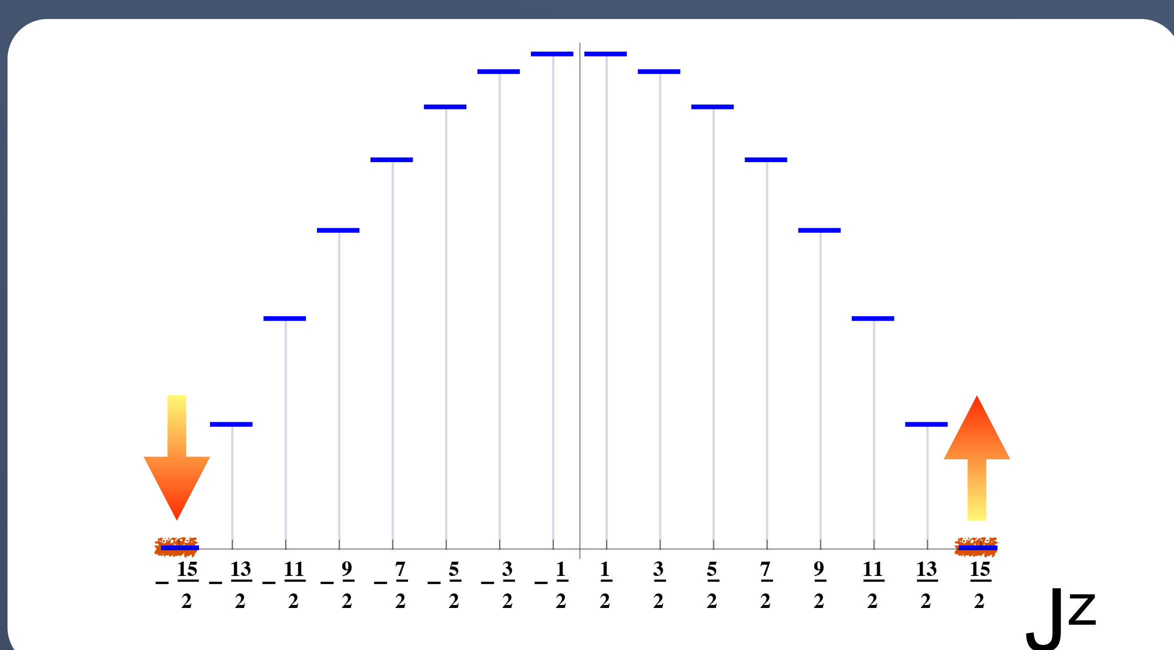
- Hund's rules: L+S=J states, split by crystal fields

following L. Balents

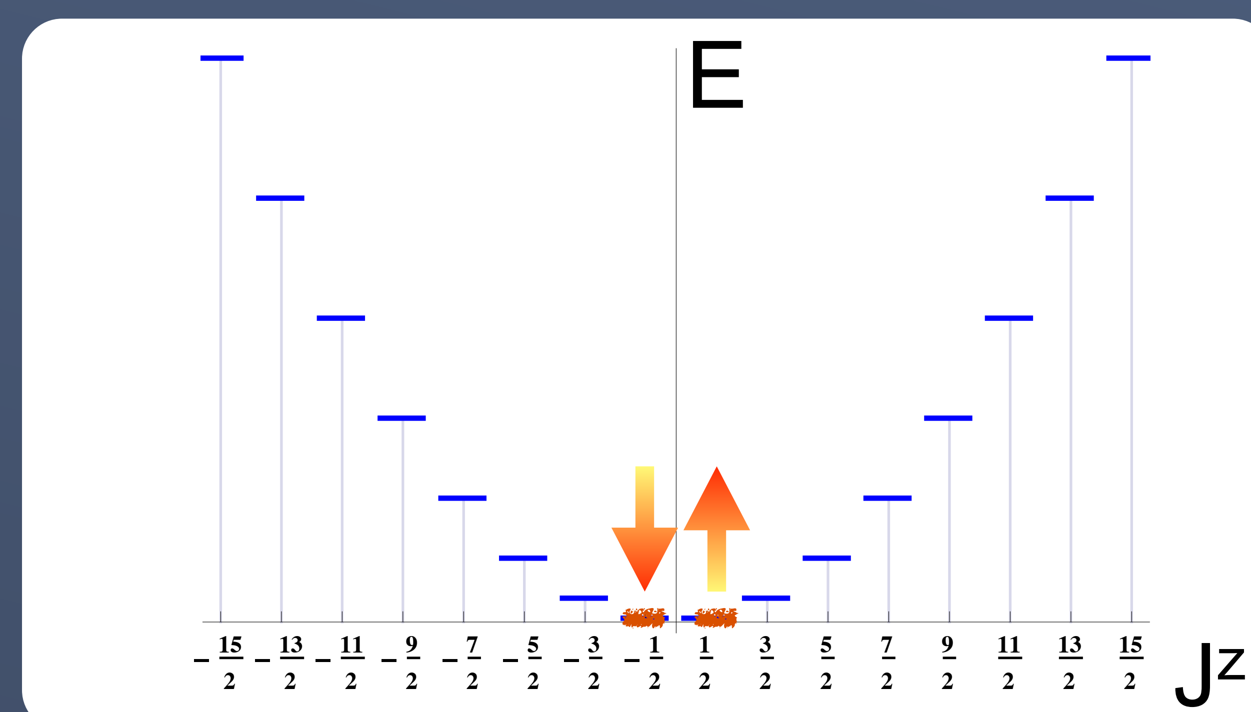
$$H_{\text{ion}} = -D\left(\vec{J}_i \cdot \hat{n}_i\right)^2$$



D>0: spin ice



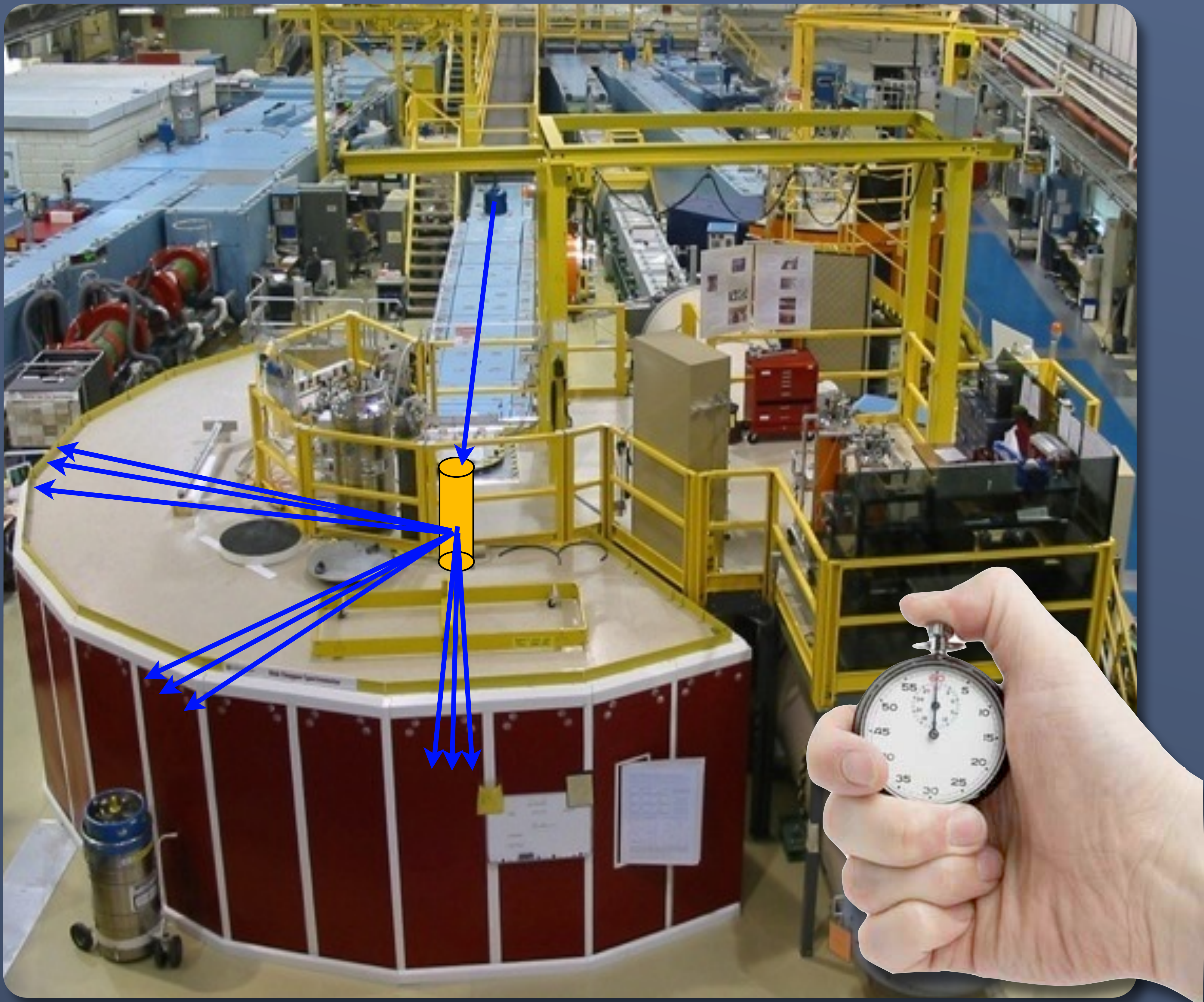
D<0: $Yb_2Ti_2O_7$, $Er_2Ti_2O_7$



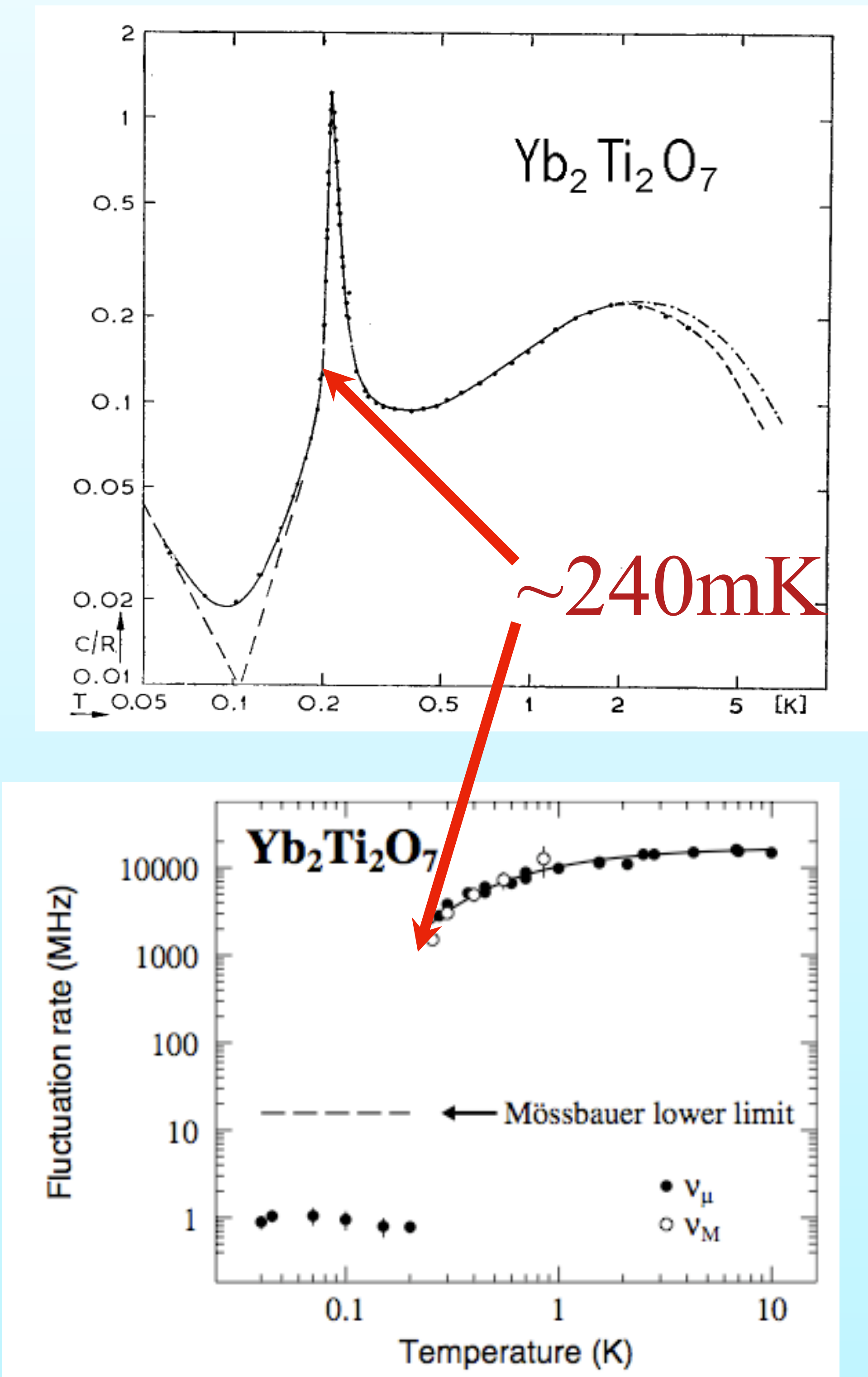
Time of Flight Neutron Scattering

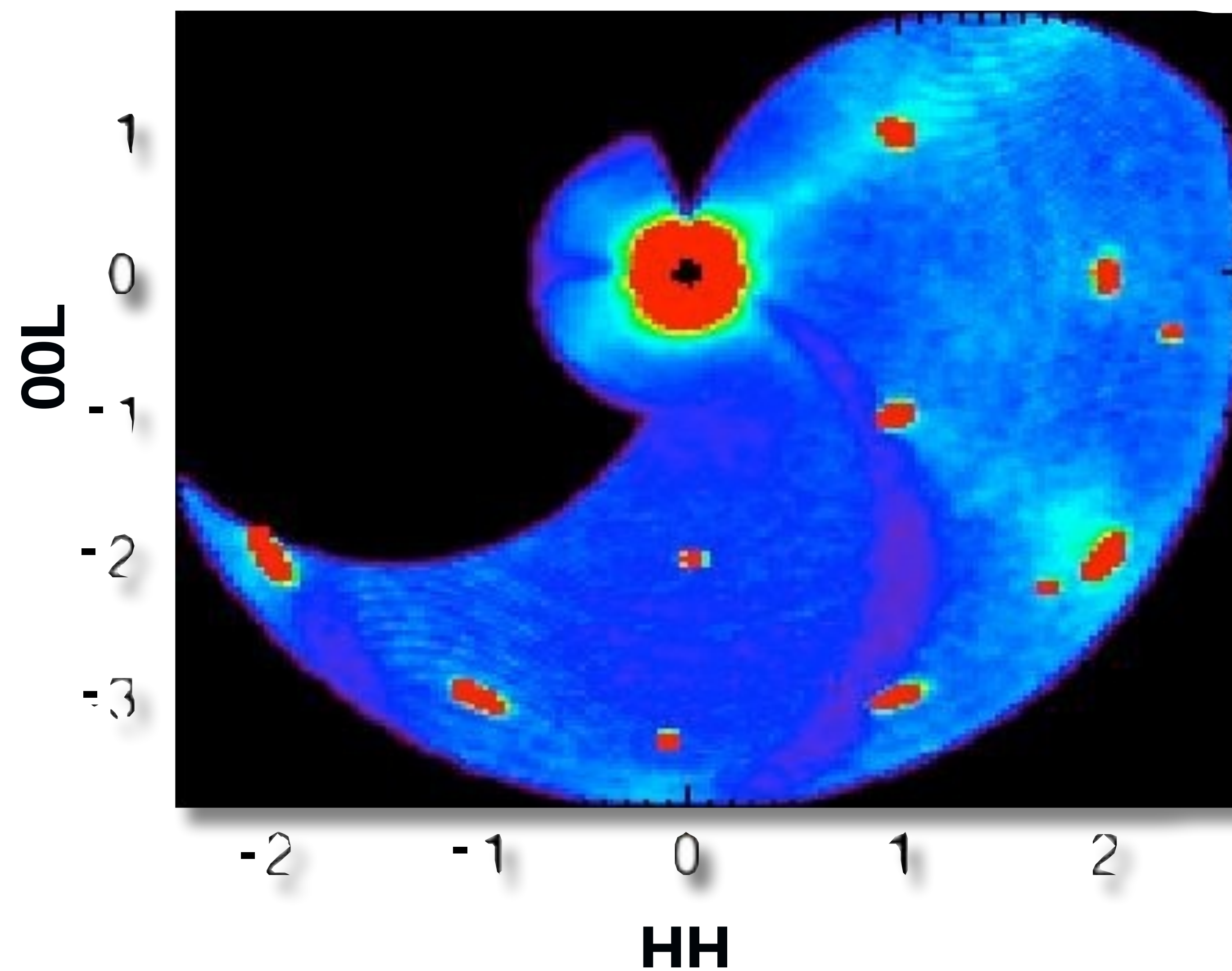
“Disk Chopper Spectrometer” (DCS)

@ NIST Center for Neutron Research



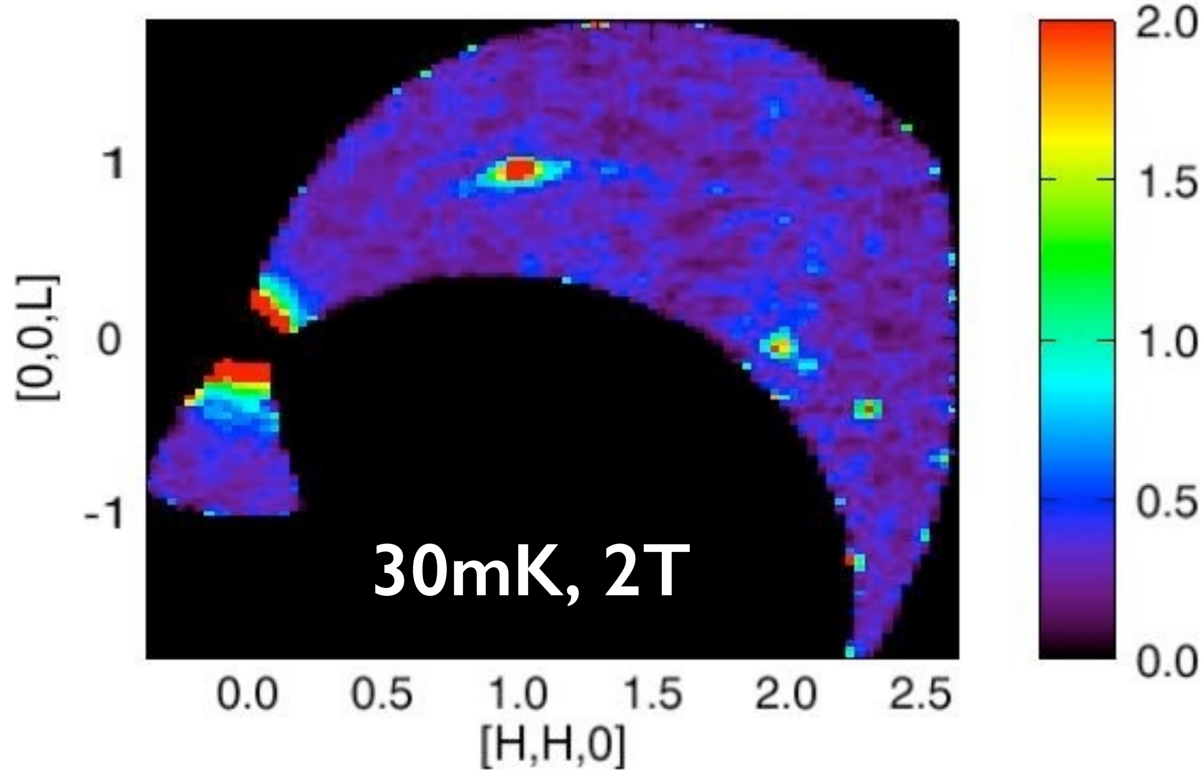
$Yb_2Ti_2O_7$ by the numbers:



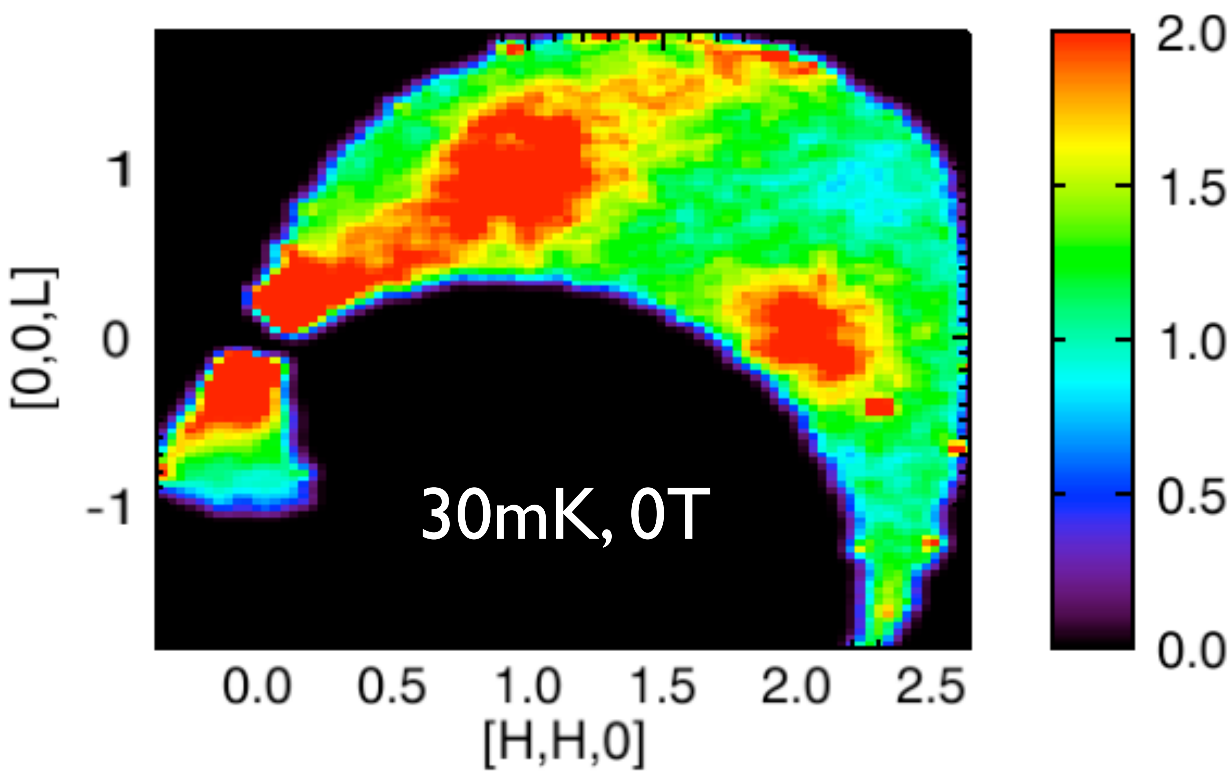


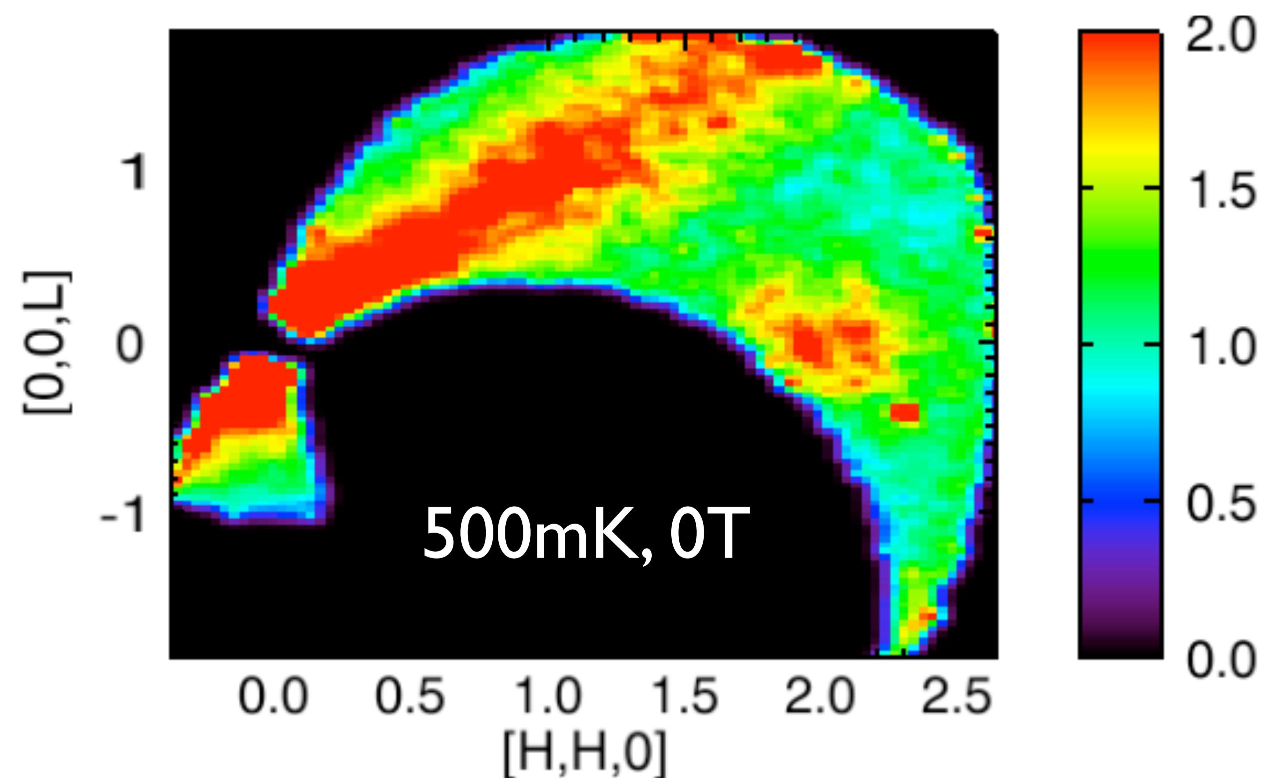
- Ferromagnetic "XY" pyrochlore
- "T_C" ~ 240 mK
- CW_T ~ +0.6 K $g_\perp/g_{||}$ ~ 2.4
- Rods of scattering observed previously by Bonville et al.

Application of a Field



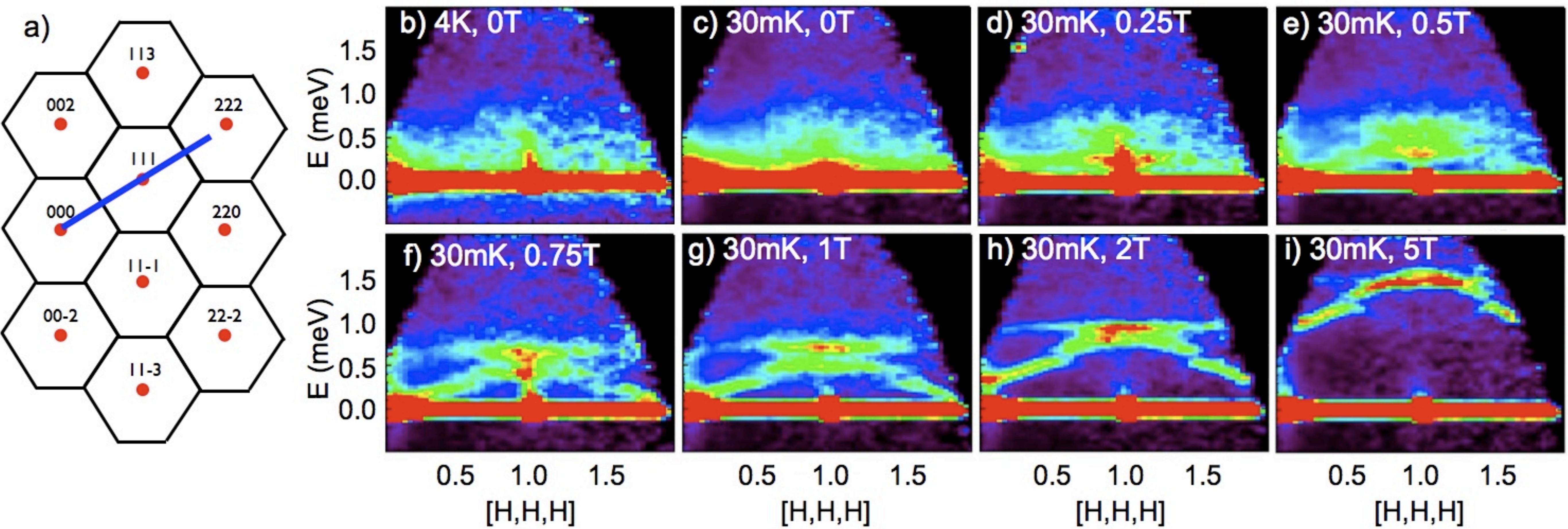
Field removes diffuse scattering

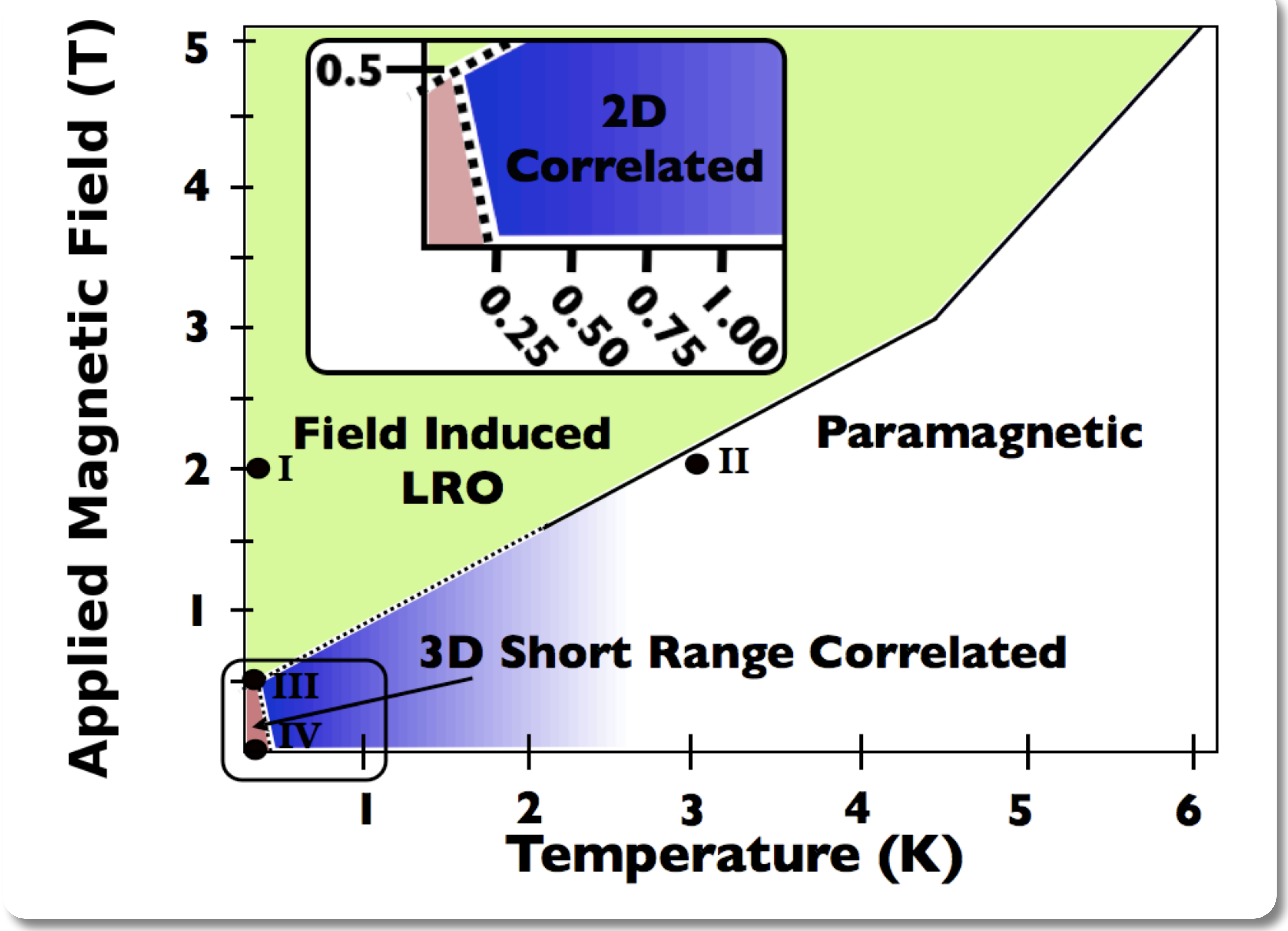




Weak magnetic field // [110] induces LRO:

appearance of long-lived spin waves at low T and moderate H

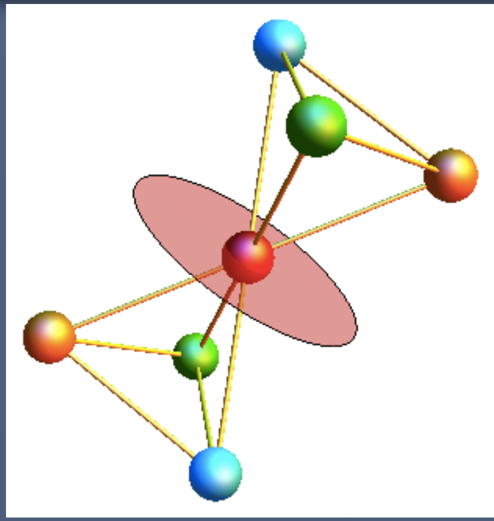


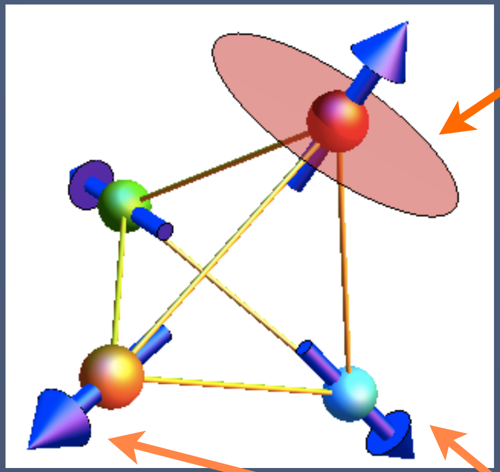


K. A. Ross, J. P. C. Ruff, C. P. Adams, J. S. Gardner, H. A. Dabkowska, Y. Qiu, J. R. D. Copley, and B. D. Gaulin, Phys. Rev. Lett. 103, 227202 (2009)

Anisotropic Exchange

RE ions are heavy - spin orbit coupling is strong
→ anisotropic exchange possible





local XY-plane

local z-axes

4 symmetry-allowed terms for exchange tensor

S. Curnoe. Phys. Rev. B 78, 094418 (2008).

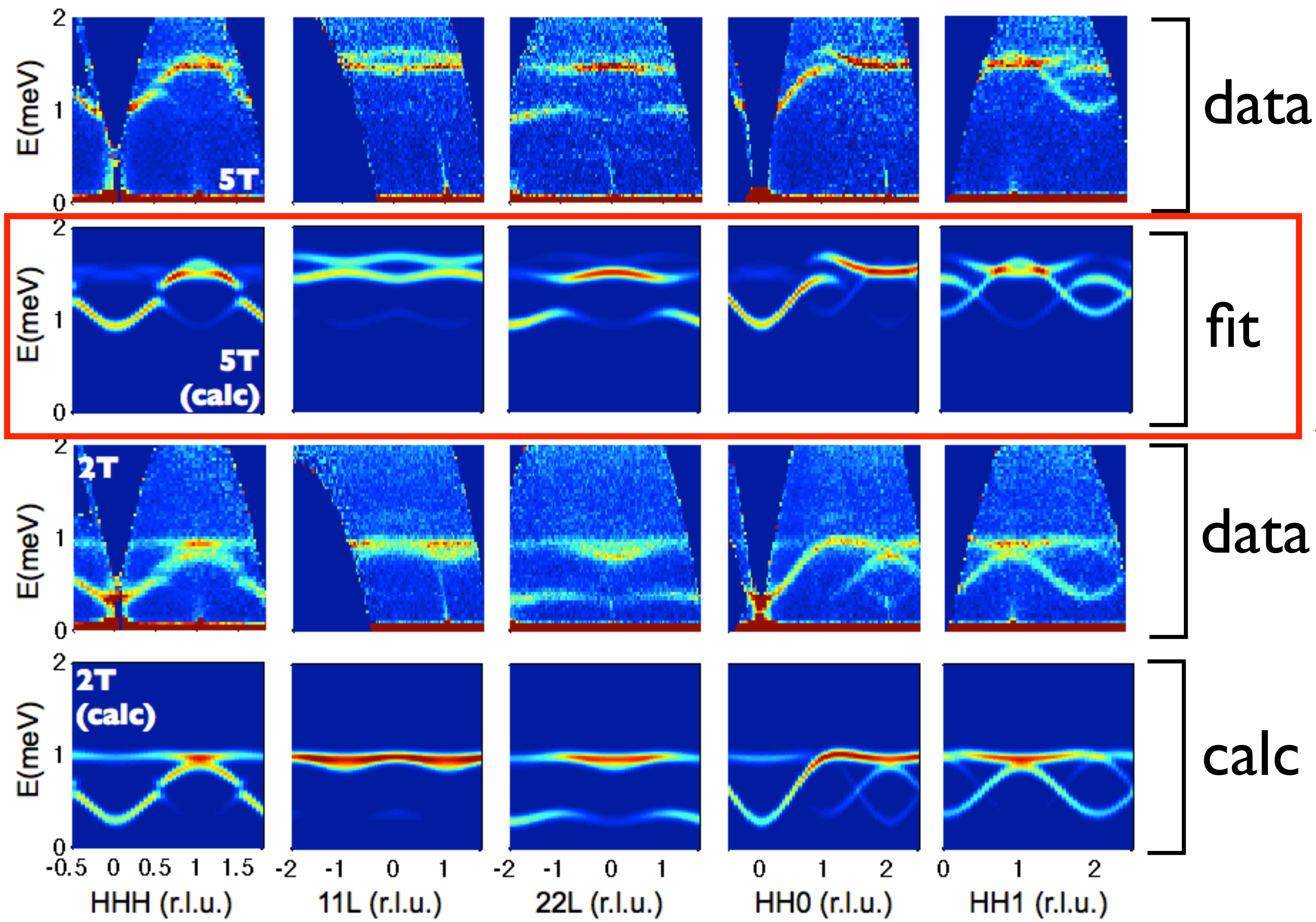
$$H = \sum_{\langle ij \rangle} \left\{ J_{zz} \mathsf{S}_i^z \mathsf{S}_j^z - J_{\pm} (\mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+) + J_{++} \left[\gamma_{ij} \mathsf{S}_i^+ \mathsf{S}_j^+ + \gamma_{ij}^* \mathsf{S}_i^- \mathsf{S}_j^- \right] + J_{z\pm} \left[\mathsf{S}_i^z (\zeta_{ij} \mathsf{S}_j^+ + \zeta_{ij}^* \mathsf{S}_j^-) + i \leftrightarrow j \right] \right\},$$

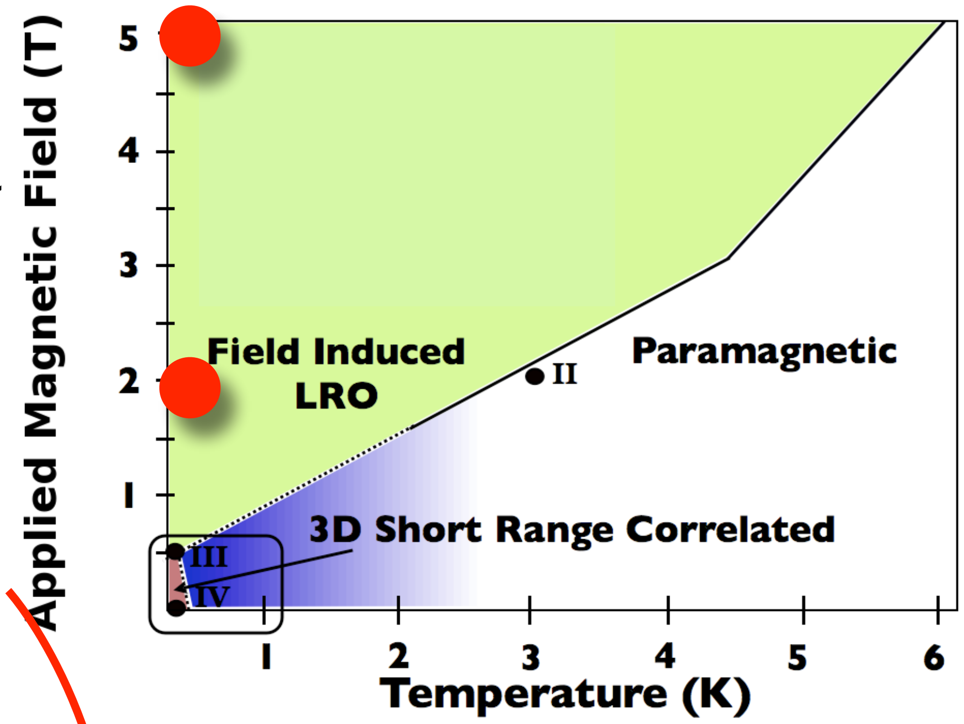
Hermele, M., Fisher, M. & Balents, L. Phys. Rev. B 69, 064404 (2004)

L. Savary, L. Balents, Phys. Rev. Lett. 108, 037202 (2012)

$Yb_2Ti_2O_7$ field polarized state

H along [1-10]

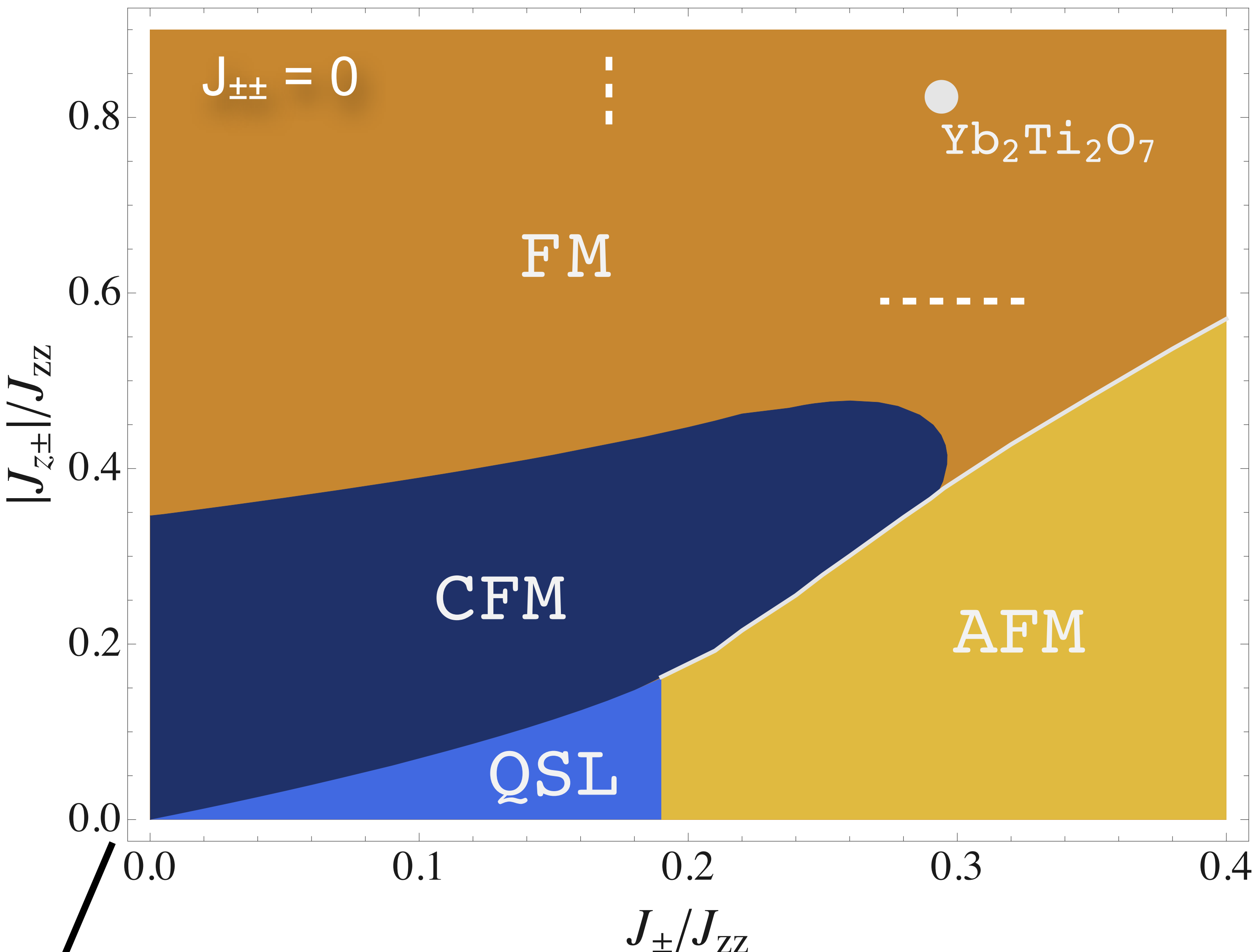


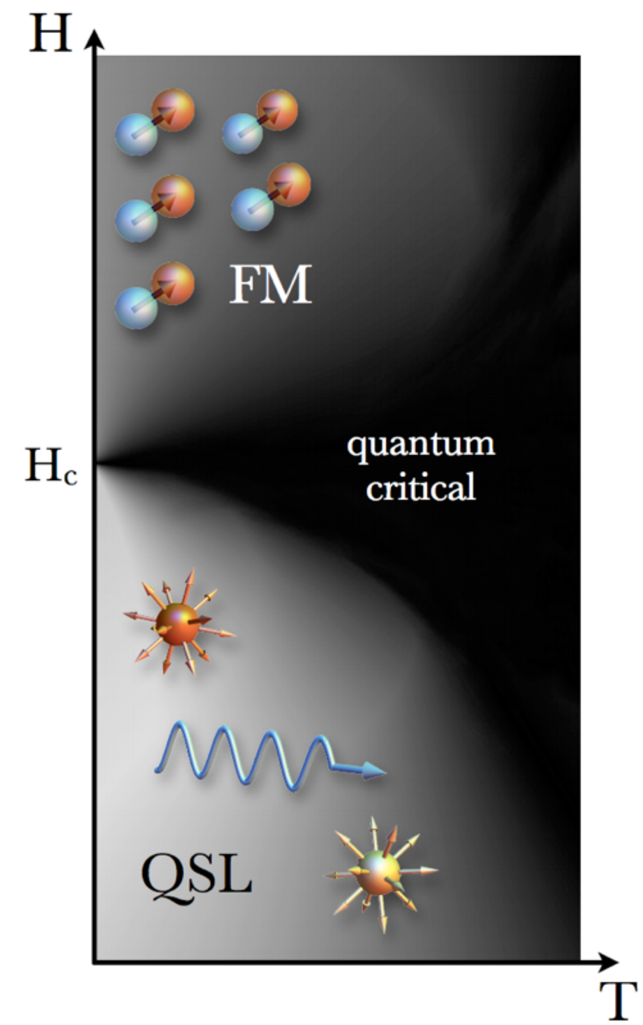


"Quantum Spin Ice"

$$J_{zz} = 0.17 \pm 0.04, \; J_{\pm} = 0.05 \pm 0.01, \; J_{\pm\pm} = 0.05 \pm 0.01, \; J_{z\pm} = -0.14 \pm 0.01 \quad \text{(meV)}$$

Gauge Mean Field Phase Diagram





L. Savary, L. Balents, Phys. Rev. Lett. 108, 037202 (2012)

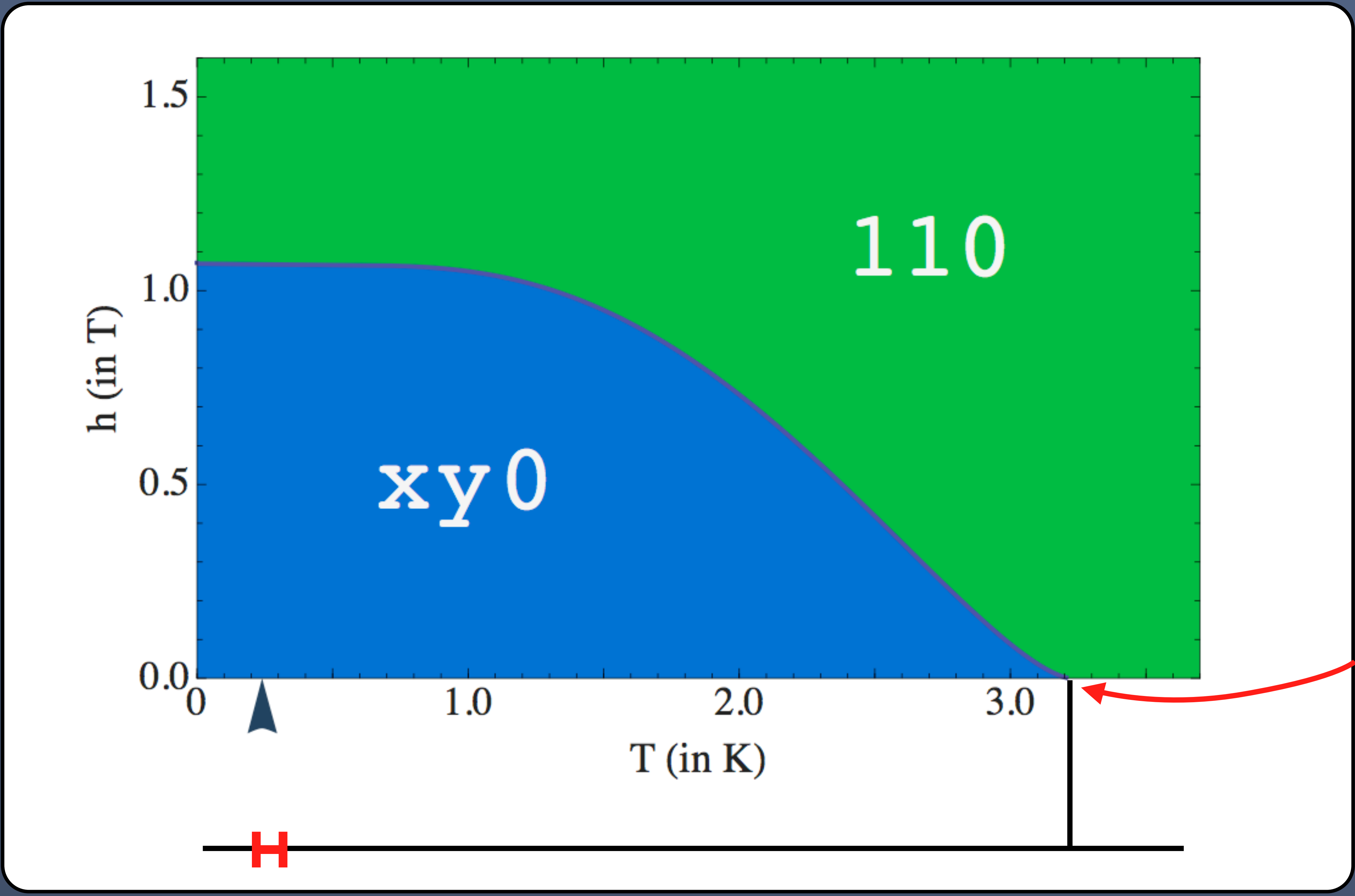
see also: H. Yan, O. Benton, L. Jaubert, N. Shannon, arXiv 1311.3501v1 (2013)

$J_{\pm\pm}/J_{zz}$

How close are we to the Coulomb QSL phase or Coulomb FM phase?

MFT phase diagram: $Yb_2Ti_2O_7$

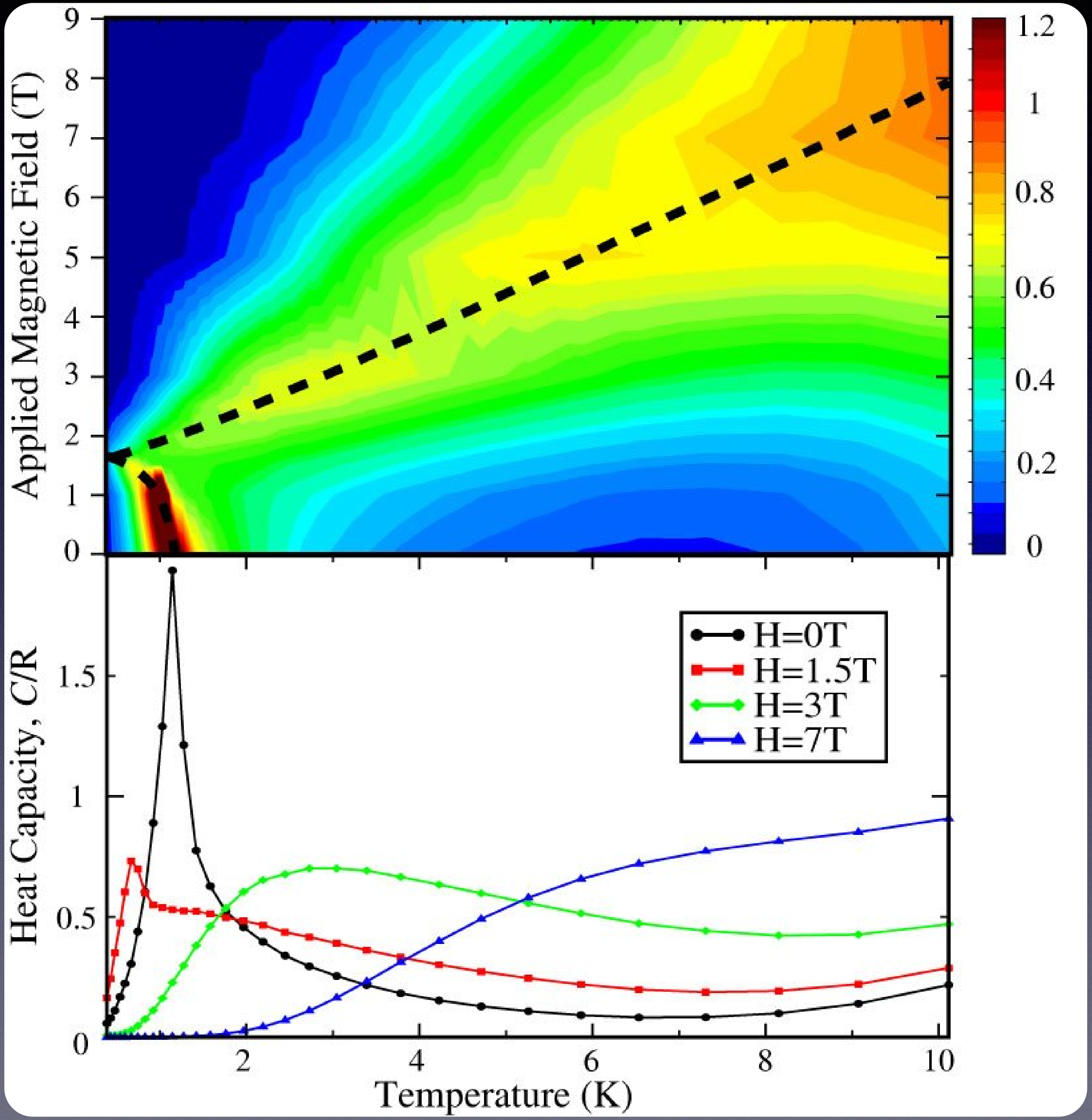
Huge suppression of T_c because of quantum fluctuations



MFT Transition from spinwave fits

Observed range of sensitivity of T_c in specific heat

AF planar pyrochlore: $Er_2Ti_2O_7$: $\Theta_{CW} \sim$ -22 K



2003-2012: The nine-year $Er_2Ti_2O_7$ ground state puzzle

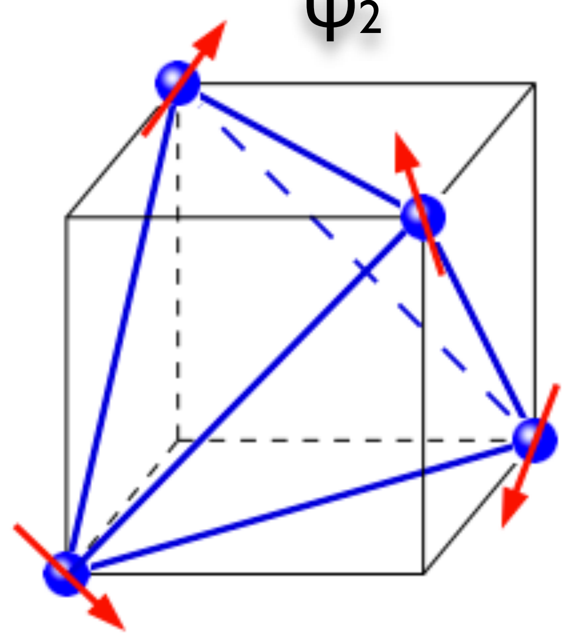
"What is the mechanism leading to ordered state selection?"

P. Stasiak, P. A. McClarty, M. J. P. Gingras, Phys. Rev. B 89, 024425 (2014)

- Not dipolar interactions → leads to "ψ_3" state (roughly Palmer-Chalker)

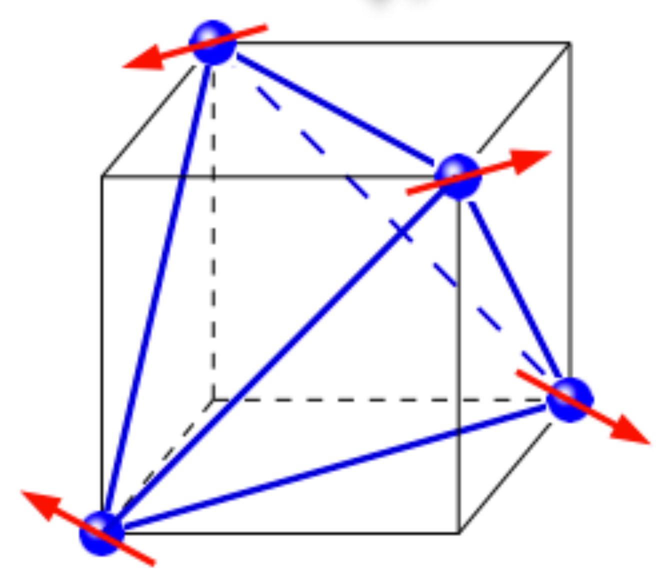
Observed state

ψ_2

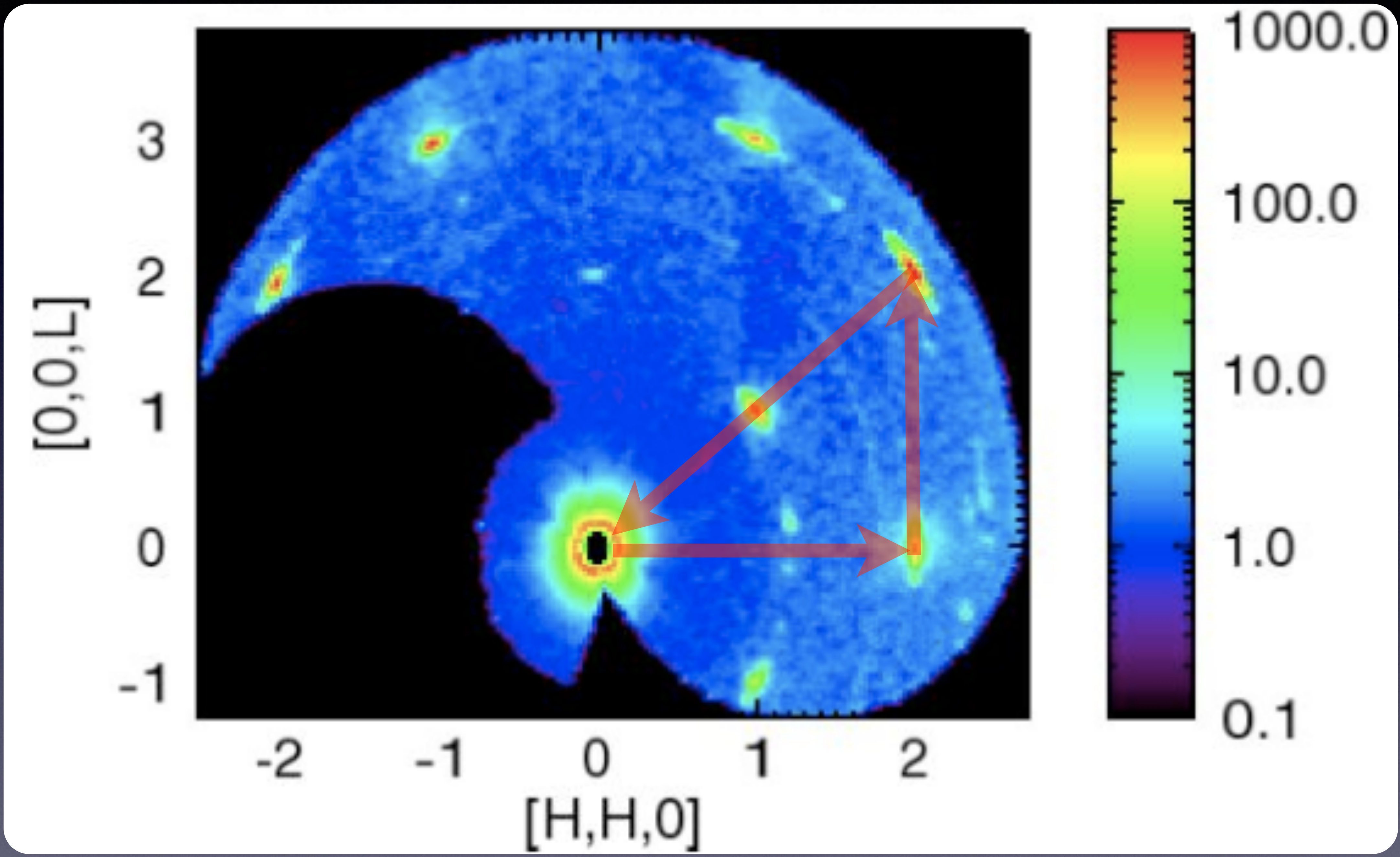


State selected by isotropic J plus long range dipolar

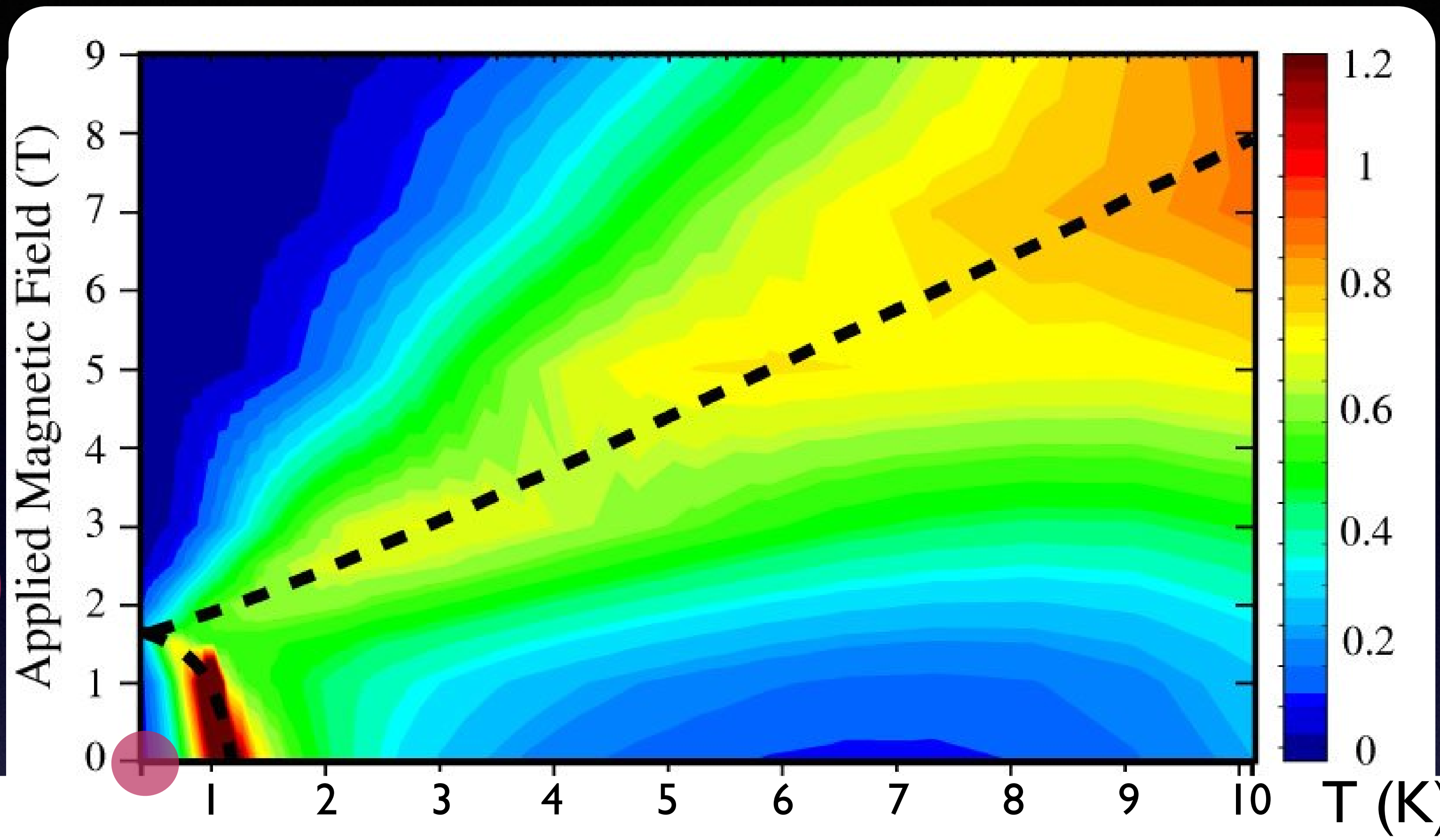
ψ_3



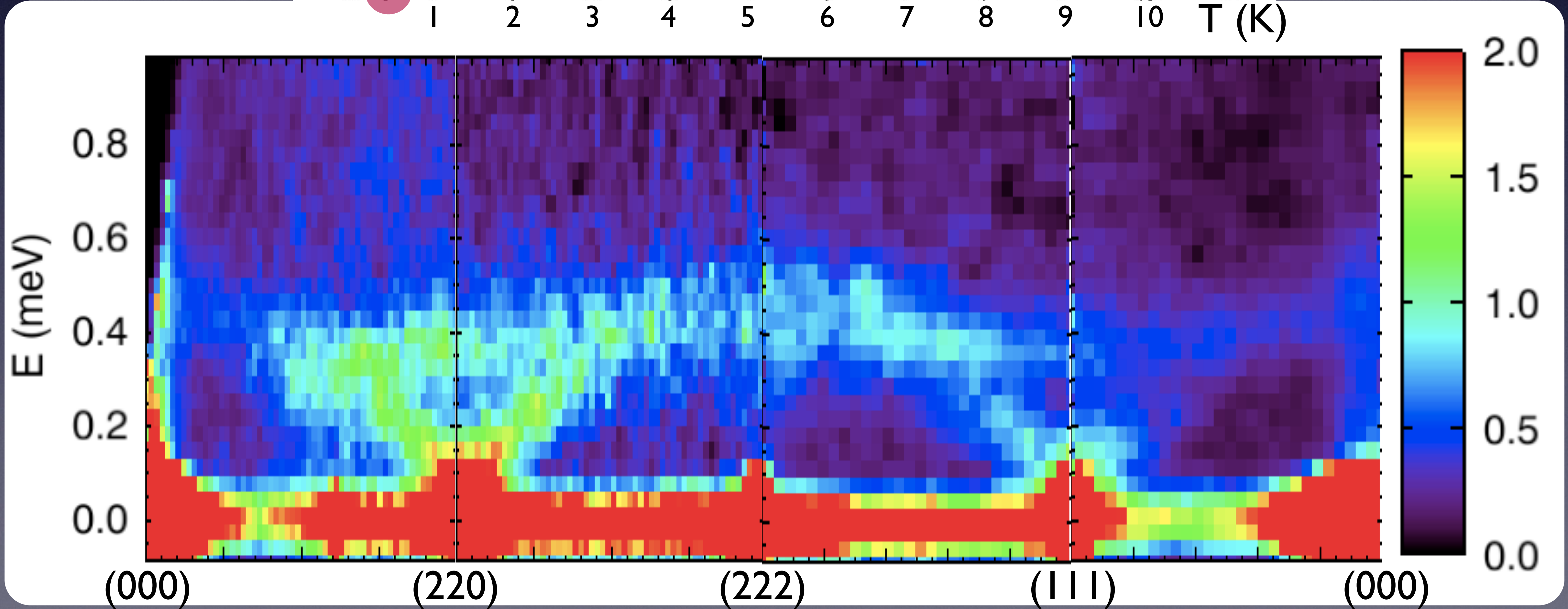
$Er_2Ti_2O_7$ @ 50 mK



50 mK, 0 T

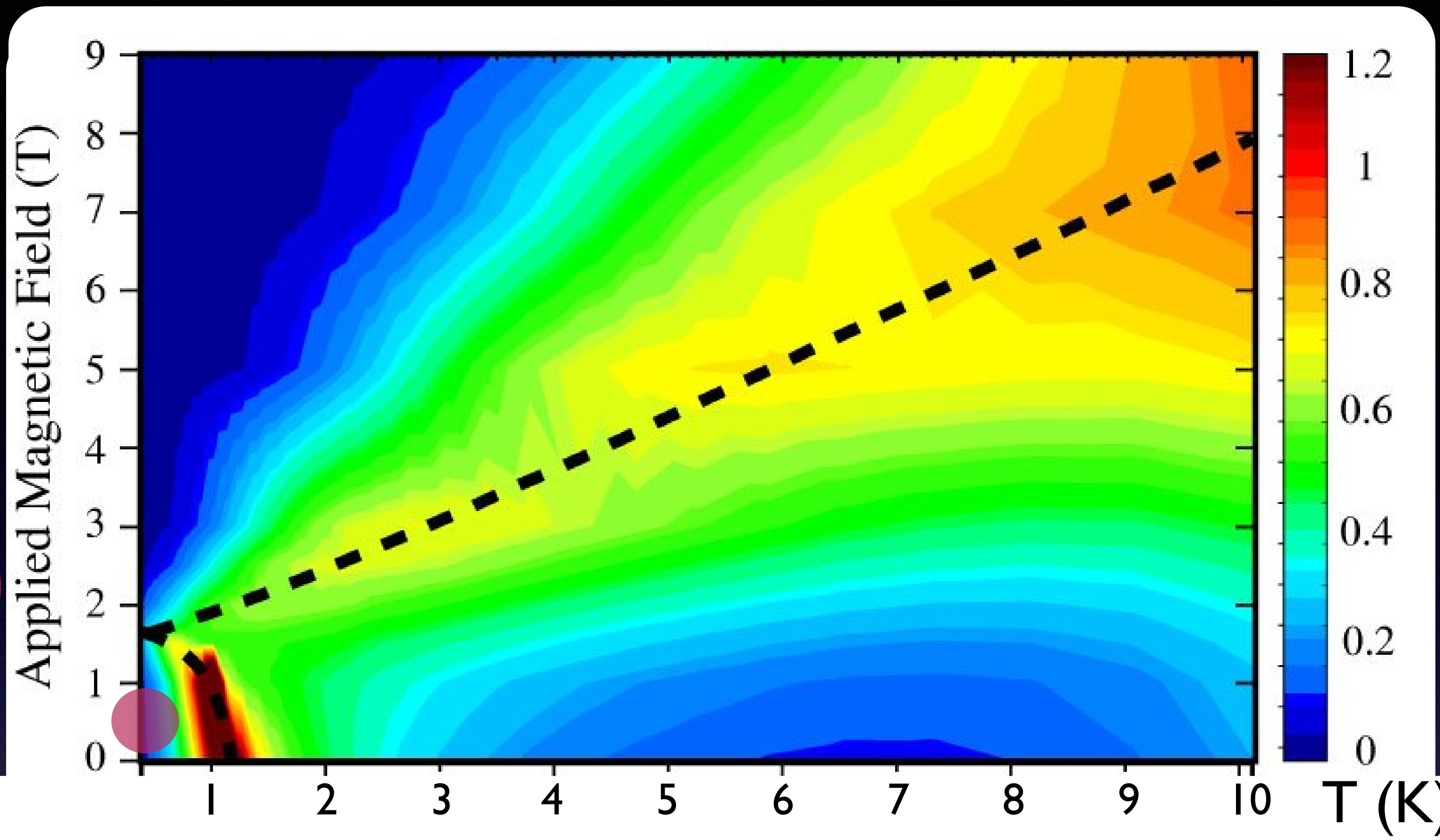


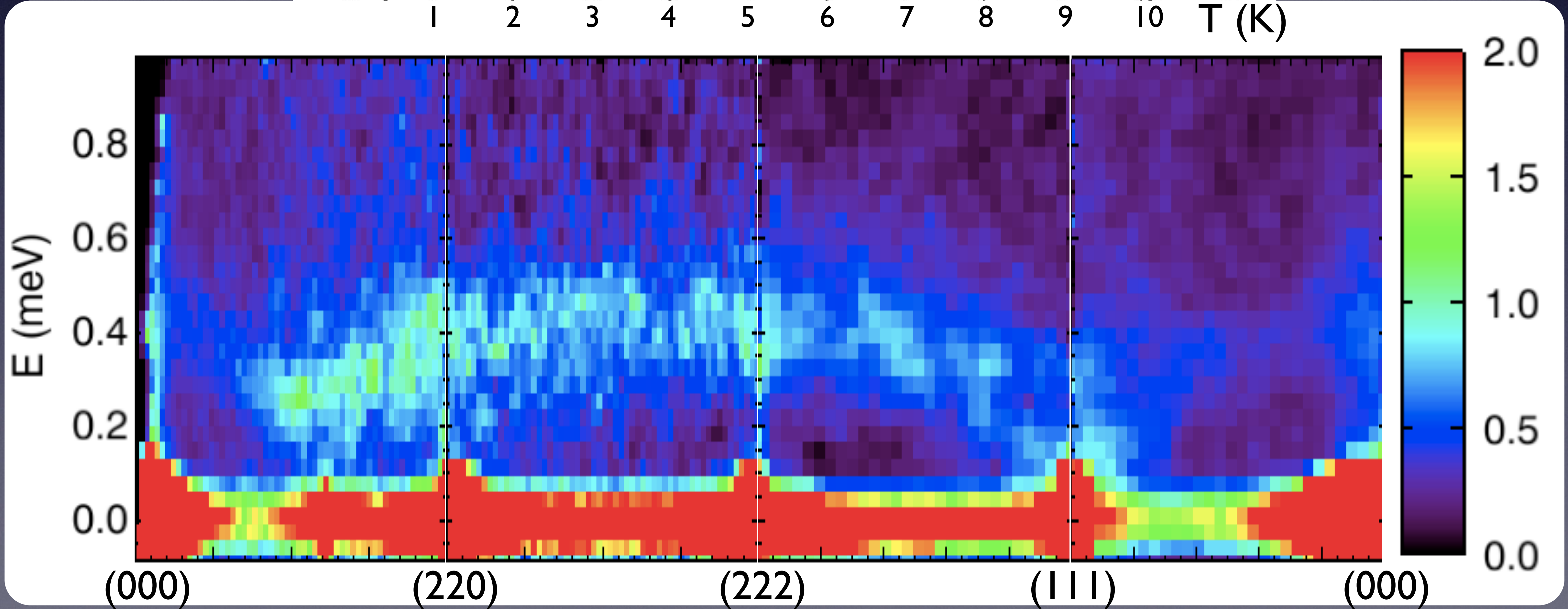
Applied Magnetic Field (T)
9
8
7
6
5
4
3
2
1
0
1
2
3
4
5
6
7
8
9
10
T (K)
1.2
1
0.8
0.6
0.4
0.2
0



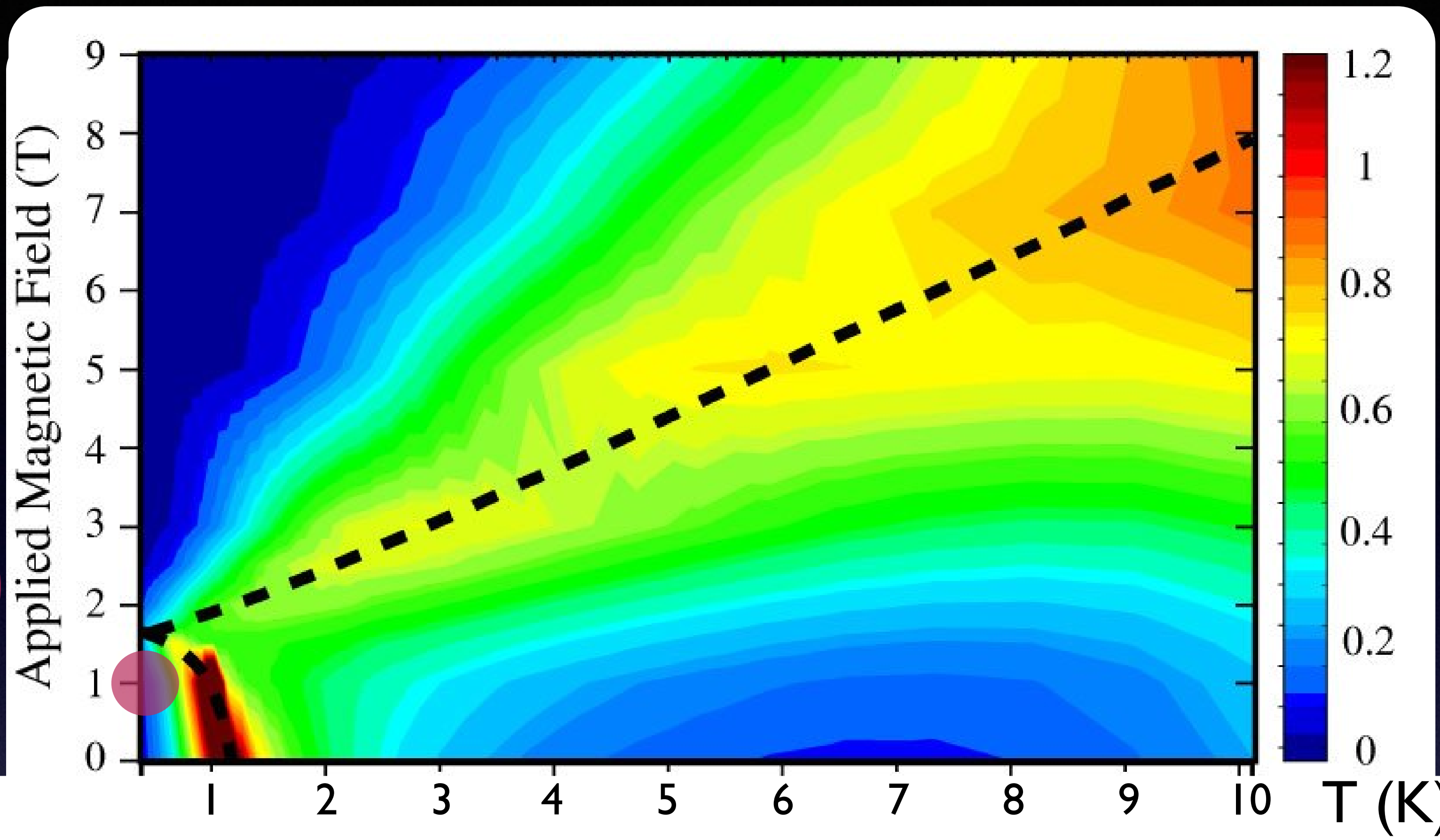
E (meV)
0.8
0.6
0.4
0.2
0.0
(000)
(220)
(222)
(111)
(000)
2.0
1.5
1.0
0.5
0.0

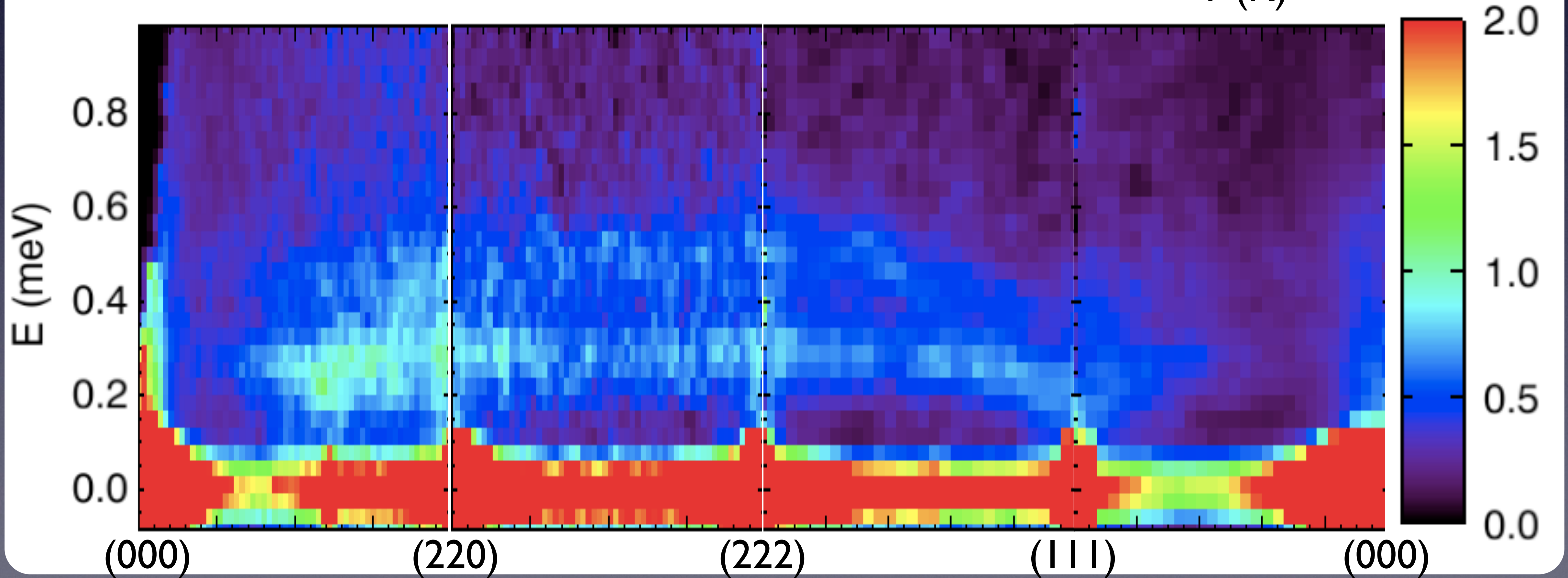
50 mK, 0.5 T


Applied Magnetic Field (T)
9
8
7
6
5
4
3
2
1
0
1
2
3
4
5
6
7
8
9
10
T (K)
1.2
1
0.8
0.6
0.4
0.2
0

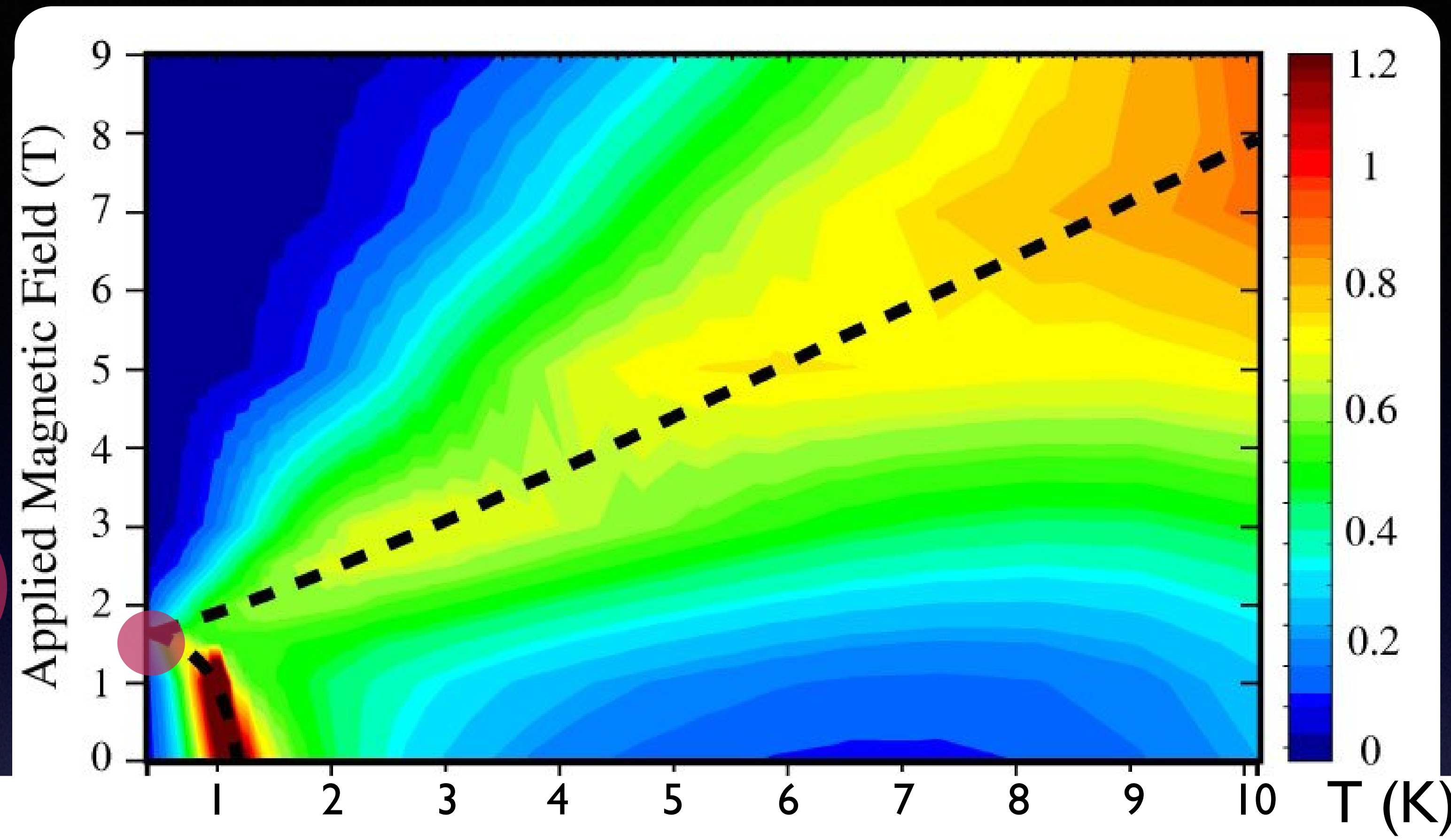

E (meV)
0.8
0.6
0.4
0.2
0.0
(000)
(220)
(222)
(111)
(000)
2.0
1.5
1.0
0.5
0.0

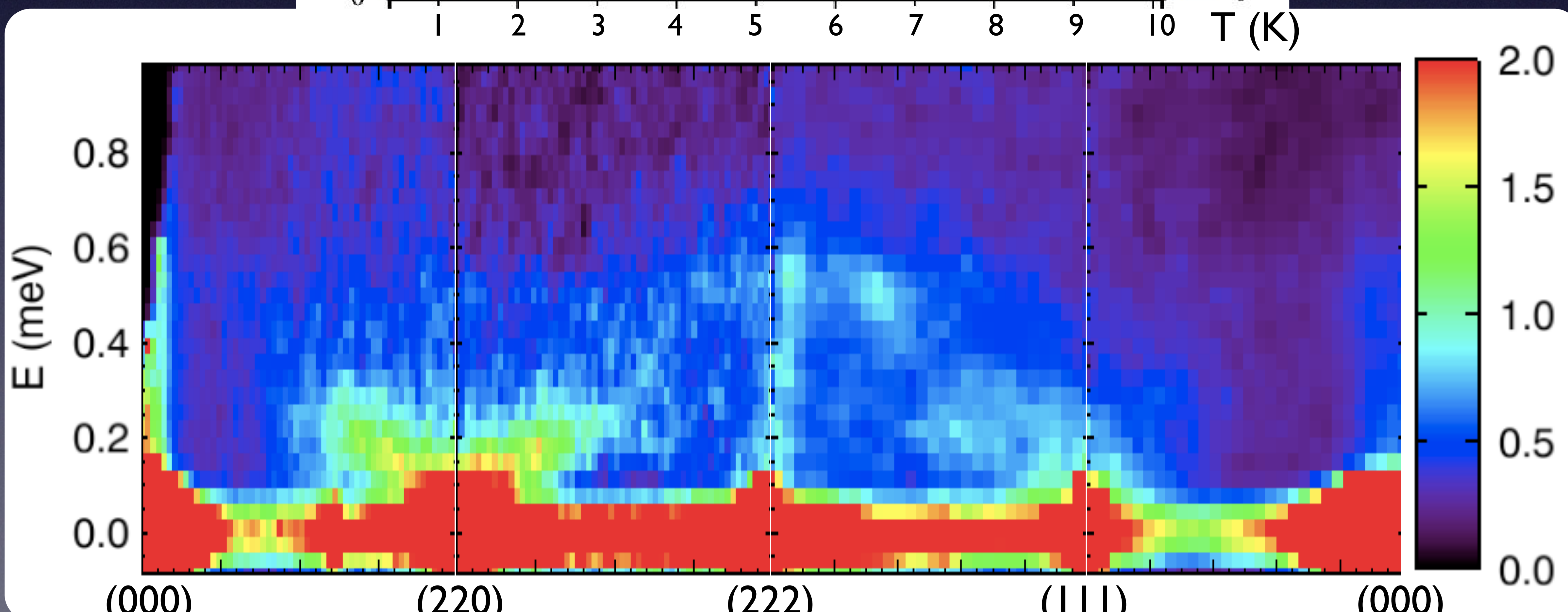
50 mK, 1.0 T



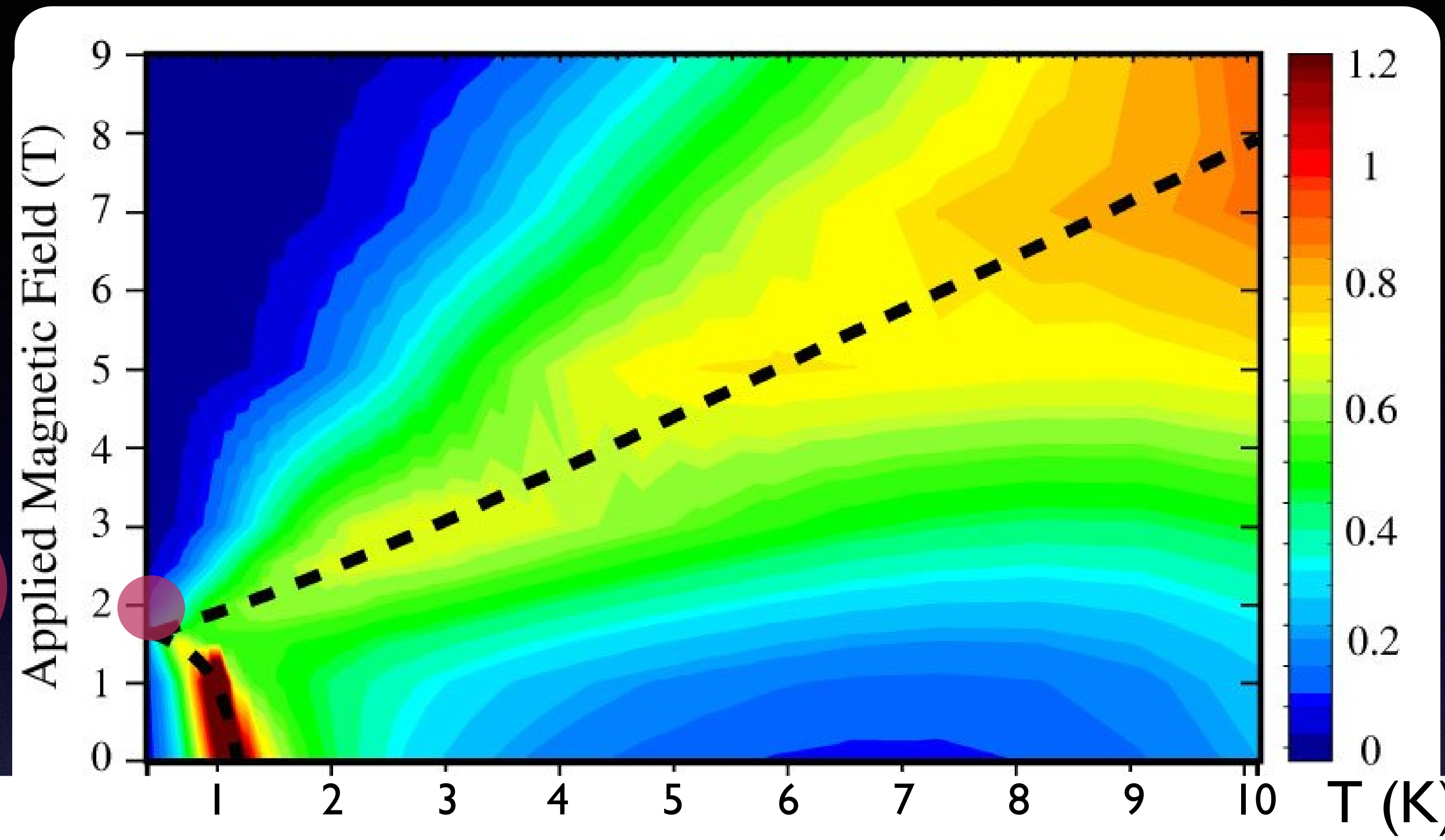


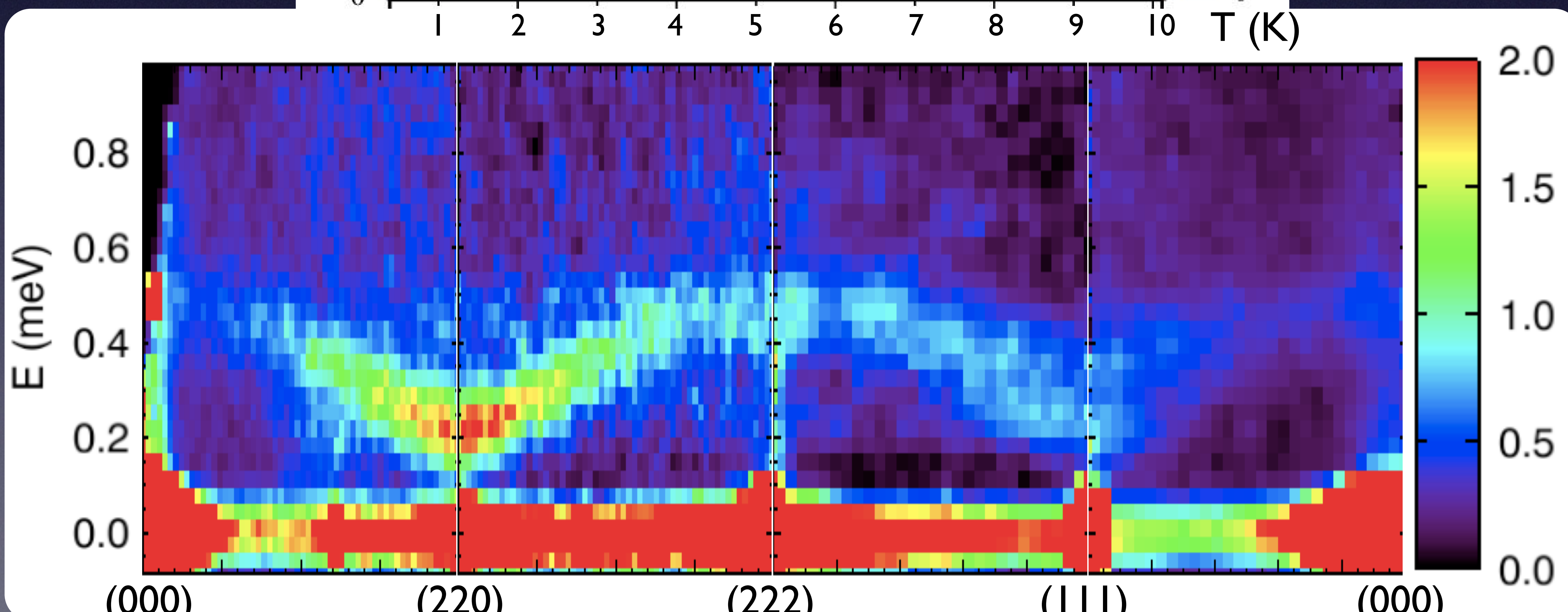
50 mK, 1.5 T


Applied Magnetic Field (T)
0
1
2
3
4
5
6
7
8
9
1
2
3
4
5
6
7
8
9
10
T (K)
0
0.2
0.4
0.6
0.8
1
1.2

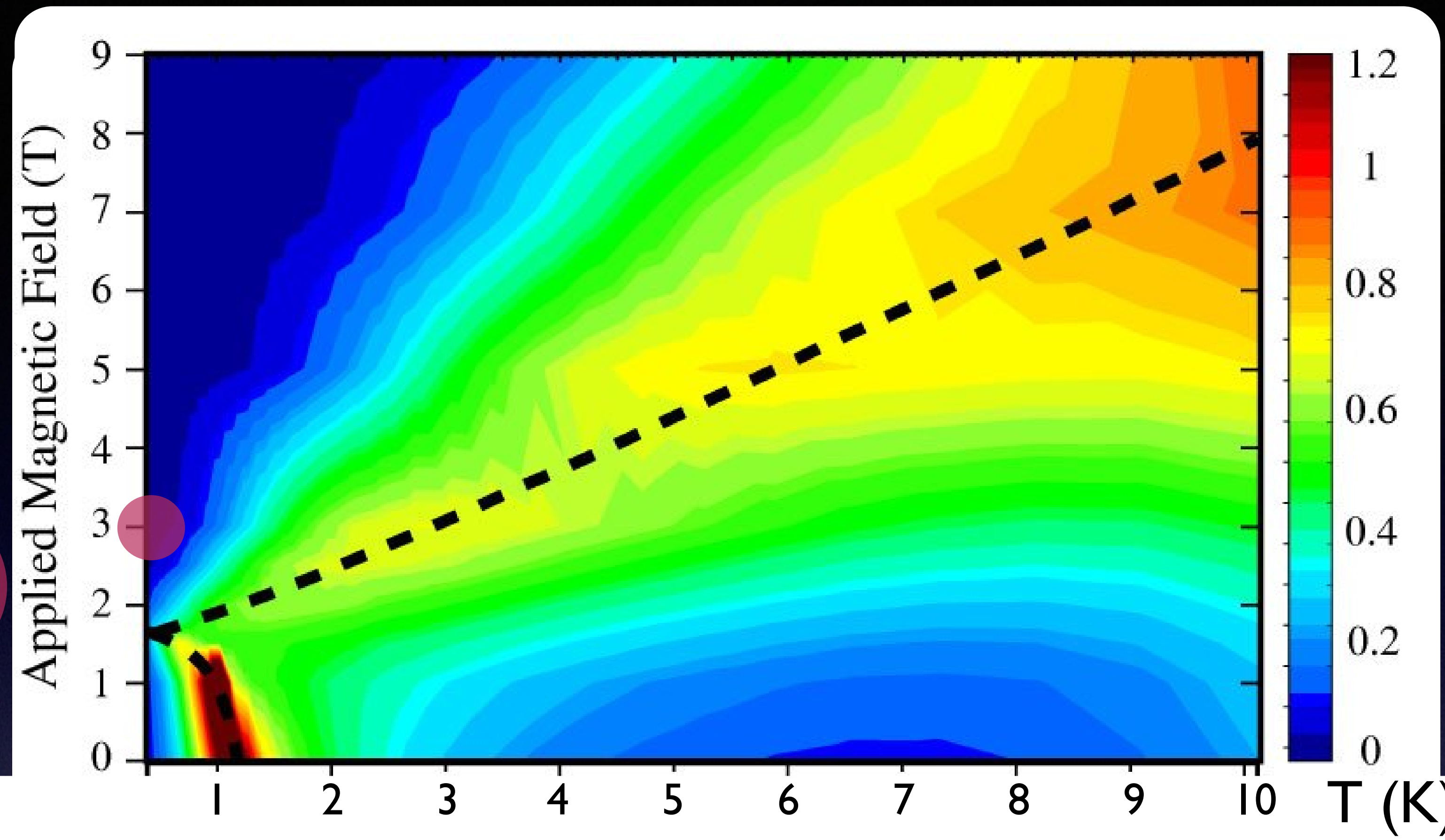

E (meV)
0.0
0.2
0.4
0.6
0.8
(000)
(220)
(222)
(111)
(000)
0.0
0.5
1.0
1.5
2.0

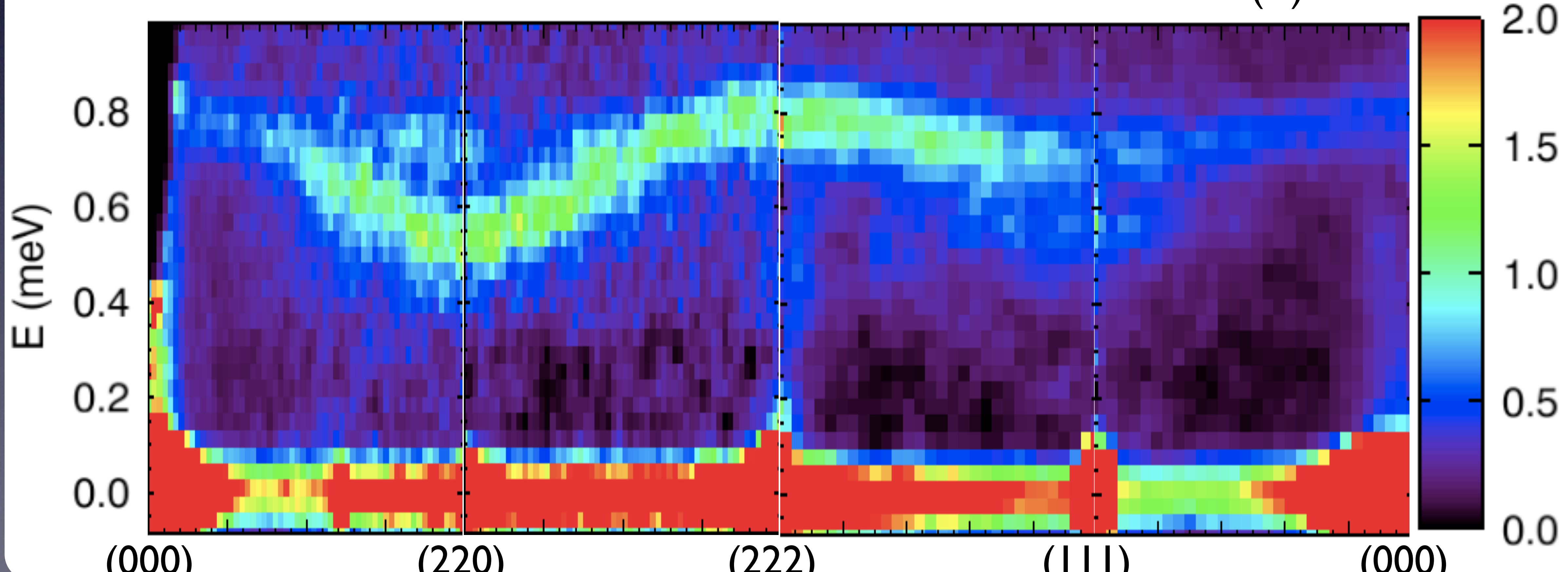
50 mK, 2.0 T

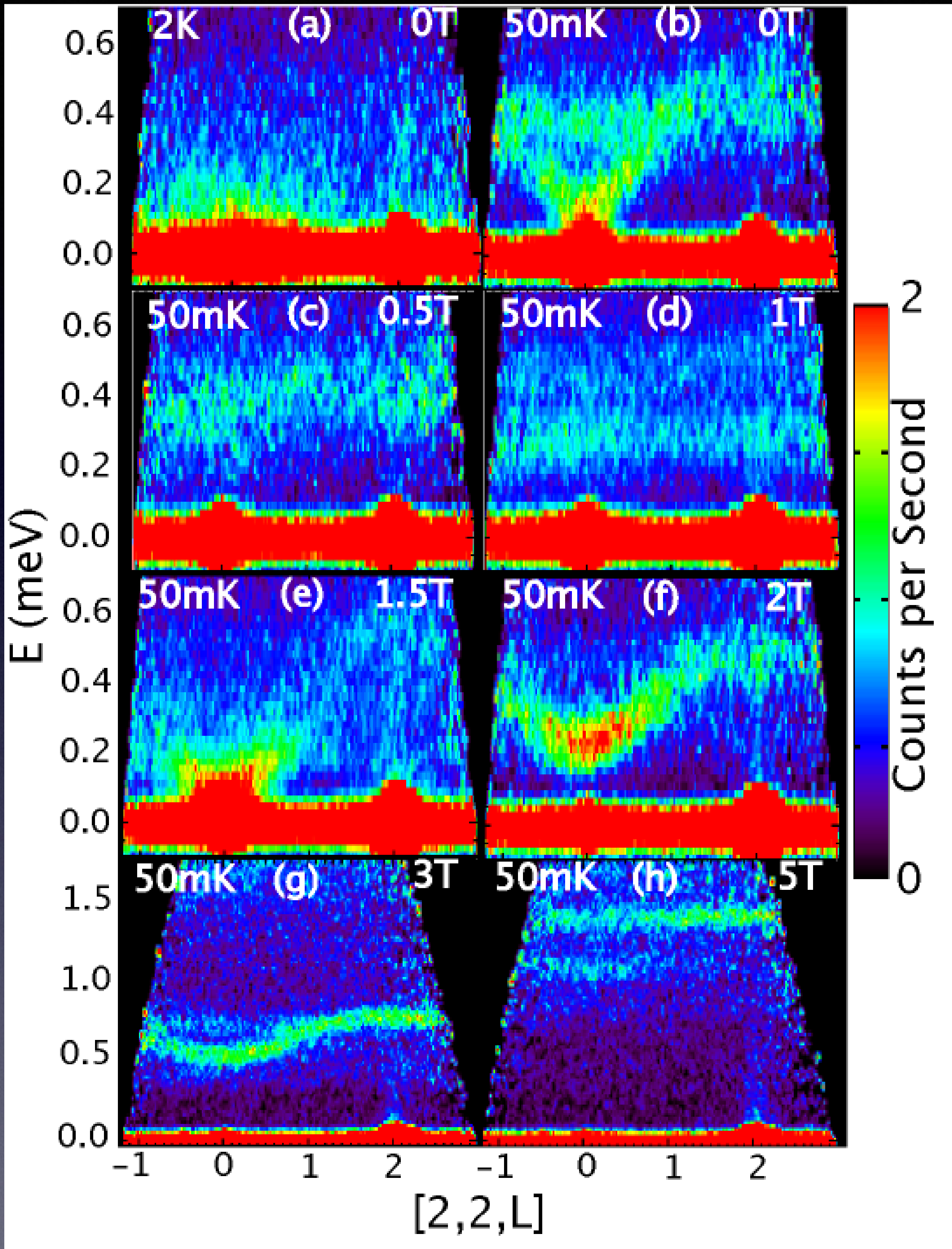

Applied Magnetic Field (T)
0
1
2
3
4
5
6
7
8
9
10
T (K)
1.2
1
0.8
0.6
0.4
0.2
0


E (meV)
0.8
0.6
0.4
0.2
0.0
2.0
1.5
1.0
0.5
0.0
(000)
(220)
(222)
(111)
(000)

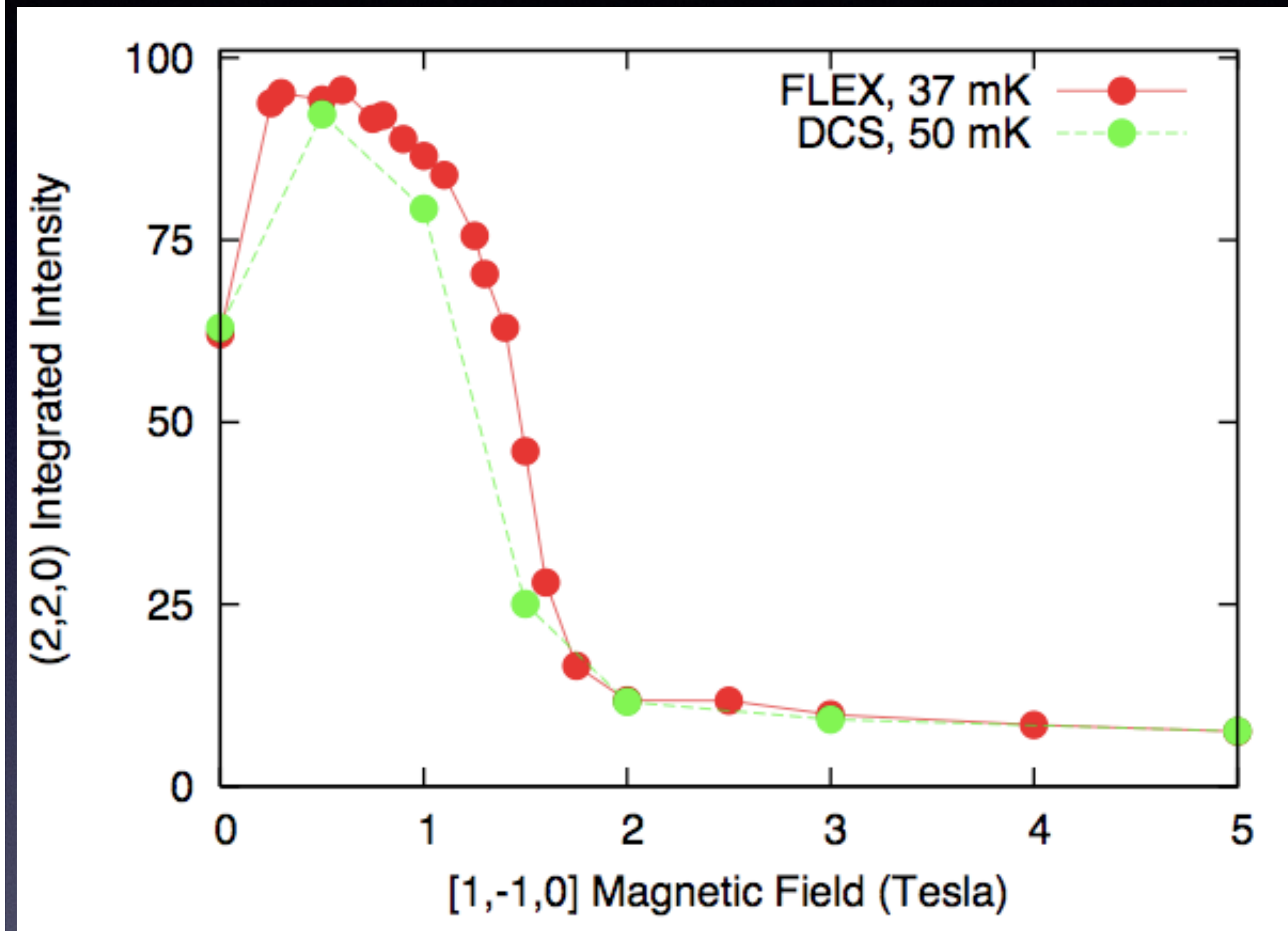
50 mK, 3.0 T


Applied Magnetic Field (T)
0
1
2
3
4
5
6
7
8
9
1
2
3
4
5
6
7
8
9
10
T (K)
0
0.2
0.4
0.6
0.8
1
1.2


E (meV)
0.0
0.2
0.4
0.6
0.8
(000)
(220)
(222)
(111)
(000)
0.0
0.5
1.0
1.5
2.0



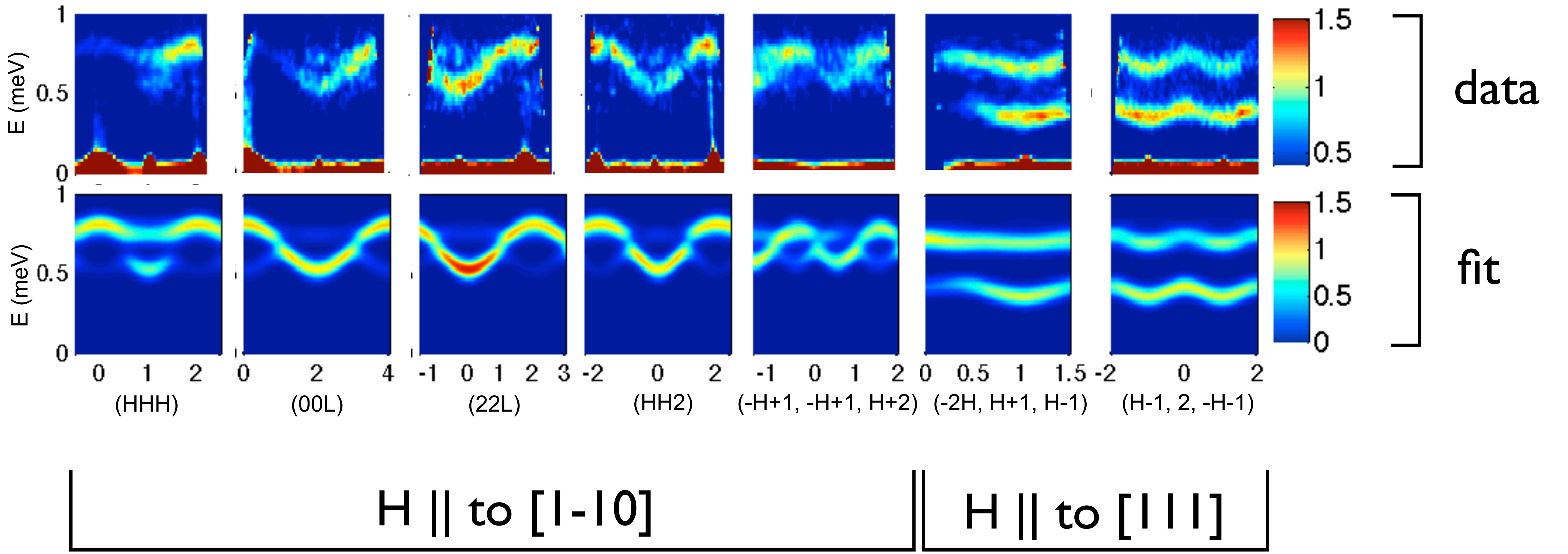
$Er_2Ti_2O_7$



J. P. C. Ruff, J.P. Clancy, A. Bourque, M.A. White, M. Ramazanoglu, J.S. Gardner, Y. Qiu, J. R. D. Copley, M.B. Johnson, H.A. Dabkowska, and B. D. Gaulin, Phys. Rev. Lett. 101, 147205 (2008)

$Er_2Ti_2O_7$: two experiments and fits

H = 3T



$$J_{zz} = -2.5\times10^{-2} \pm 1.8\times10^{-2}, J_{\pm} = 6.5\times10^{-2} \pm 7.5\times10^{-3}$$
$$J_{\pm\pm} = 4.2\times10^{-2} \pm 5.0\times10^{-3}, J_{z\pm} = -8.8\times10^{-3} \pm 1.5\times10^{-2}$$

(meV)

Degeneracy of Ground State

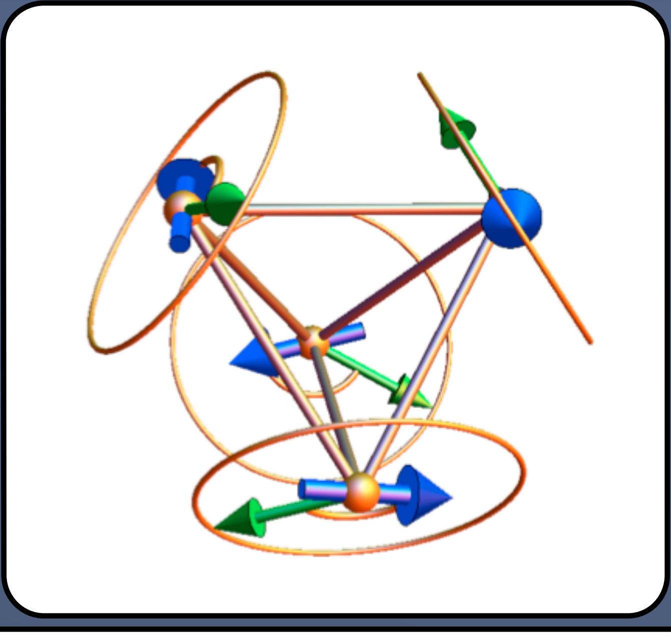
→ continuous degeneracy at Mean Field level

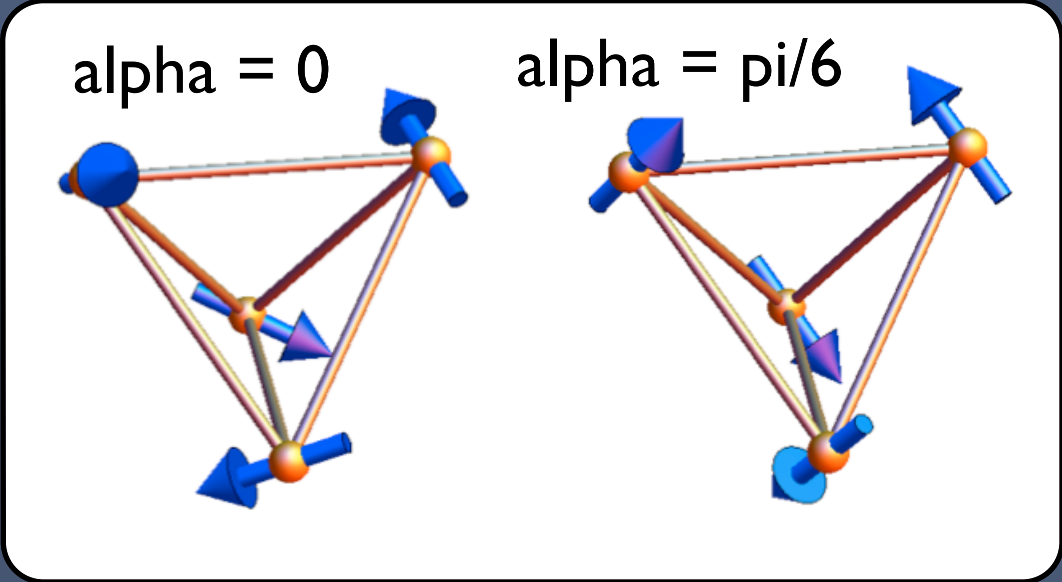
→ **Cannot be broken by dipolar or further range interactions**

→ parameterized by single angle parameter: alpha

→ degeneracy broken by OBD gives states with alpha = 0, pi/3, etc.

→ does the data show the 6 OBD states?





$$H = \sum_{\langle ij \rangle} \{ J_{zz} \mathsf{S}_i^z \mathsf{S}_j^z - J_{\pm} (\mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+) + J_{++} \left[\gamma_{ij} \mathsf{S}_i^+ \mathsf{S}_j^+ + \gamma_{ij}^* \mathsf{S}_i^- \mathsf{S}_j^- \right]$$

$$+ J_{z\pm} \left[\mathsf{S}_i^z (\zeta_{ij} \mathsf{S}_j^+ + \zeta_{ij}^* \mathsf{S}_j^-) + i \leftrightarrow j \right] \},$$

Order by Disorder (quantum and thermal)

'accidental degeneracy':
at the mean field level, the ground state shows a continuous symmetry that is not present in the Hamiltonian.

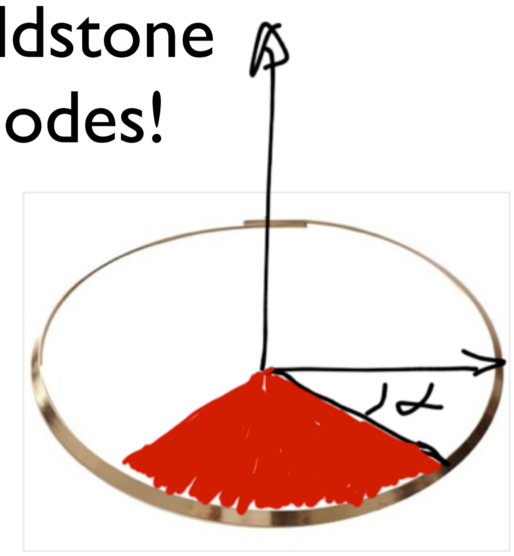
When dynamics are *softer* along specific directions, higher density of low E modes = more microstates available at specific "alphas"

→ the entropic term in F = E-TS selects the ordered state at non-zero T (thermal ObD)

OR

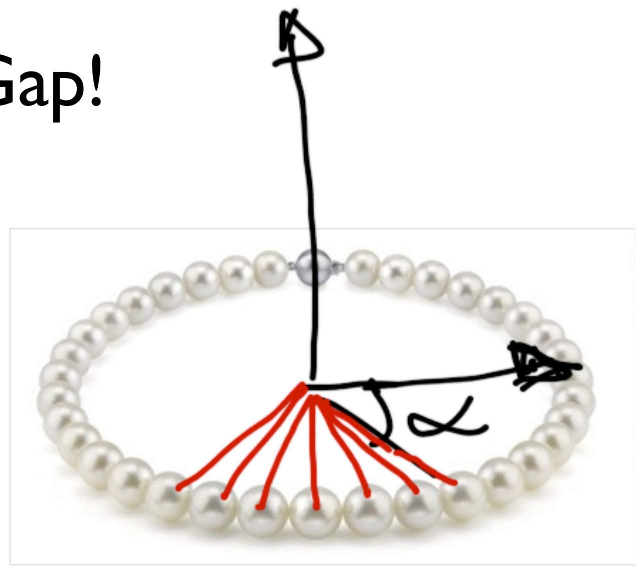
→ Quantum fluctuations select the ordered state even at zero T (Quantum ObD) i.e. fluctuations introduce an effective term to the Hamiltonian that breaks the accidental degeneracy

Goldstone modes!



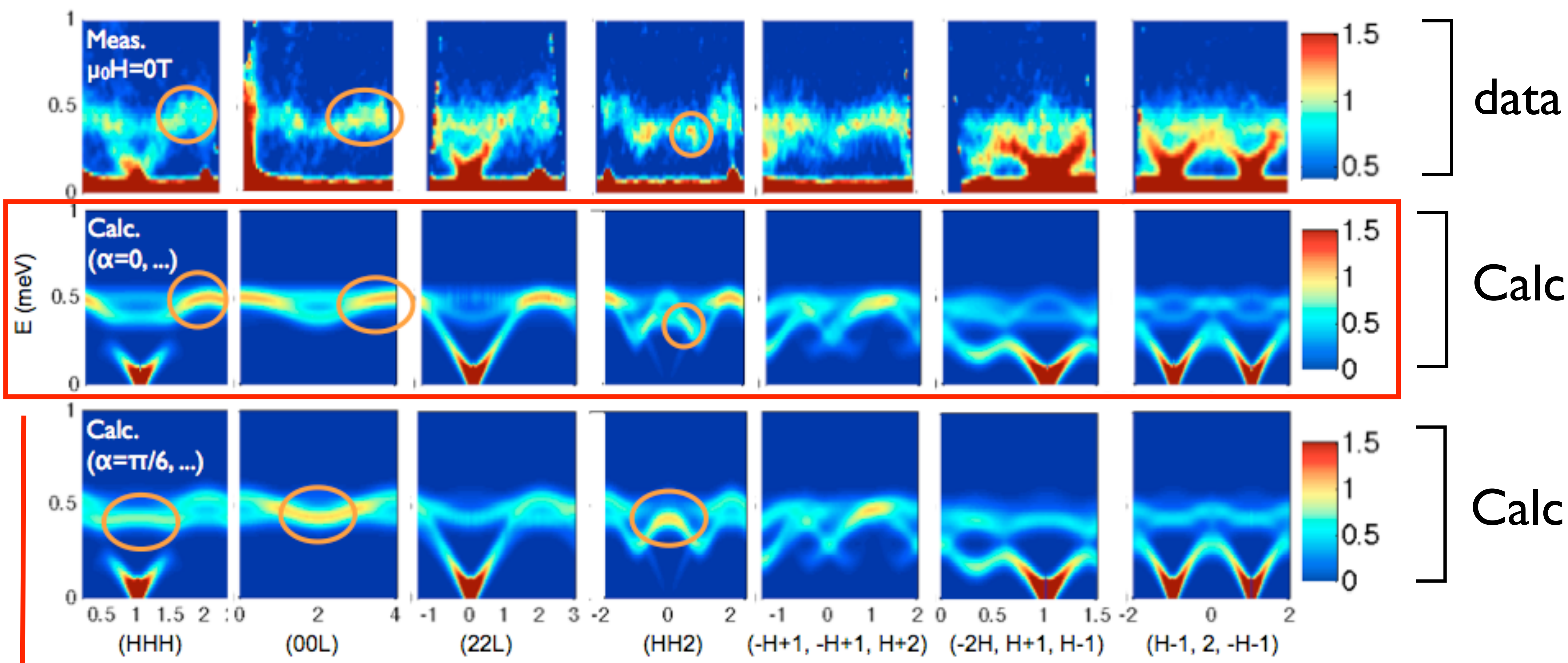
The necklaces represent surfaces of constant free energy in configuration space

Gap!



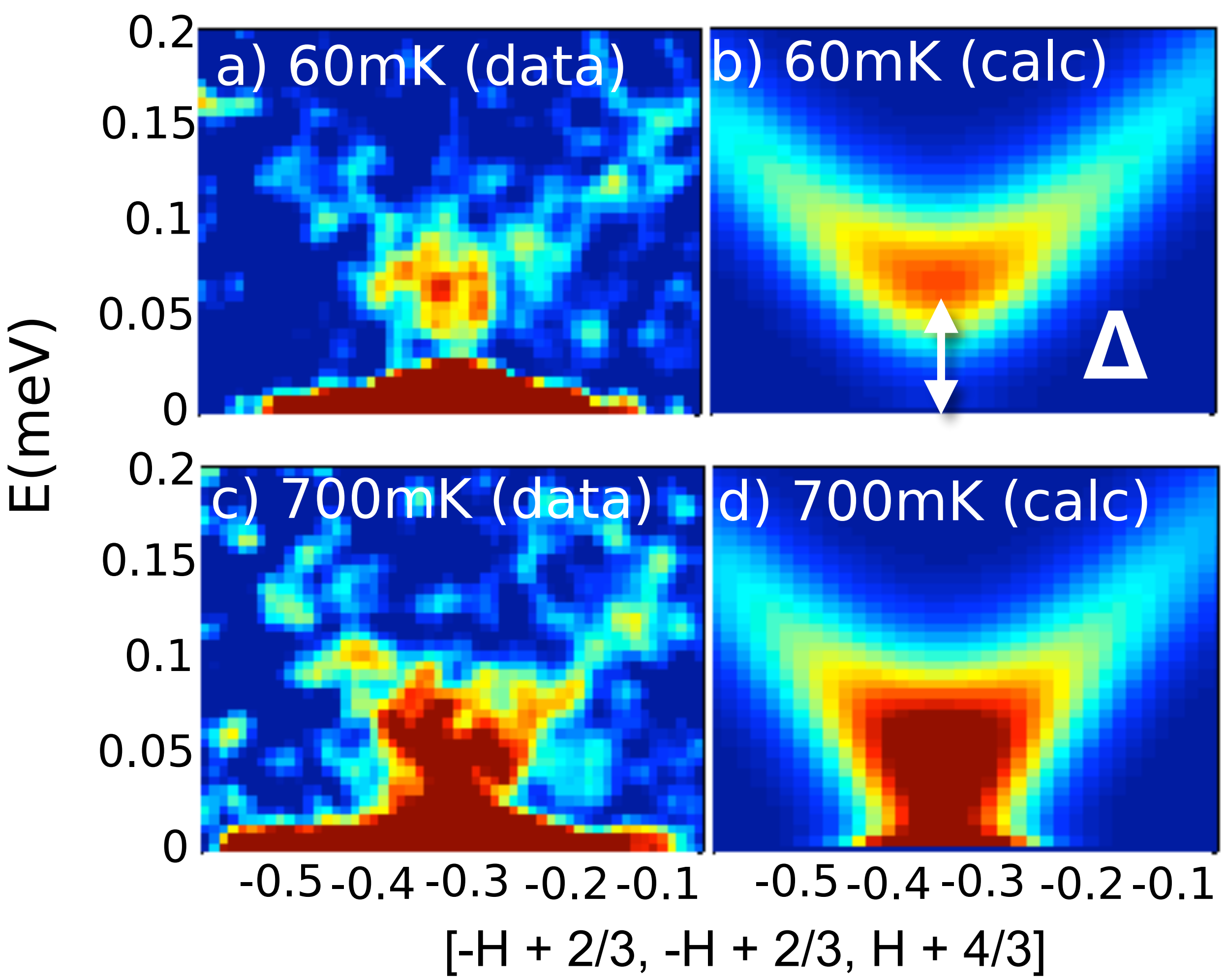
$Er_2Ti_2O_7$: zero field calculation

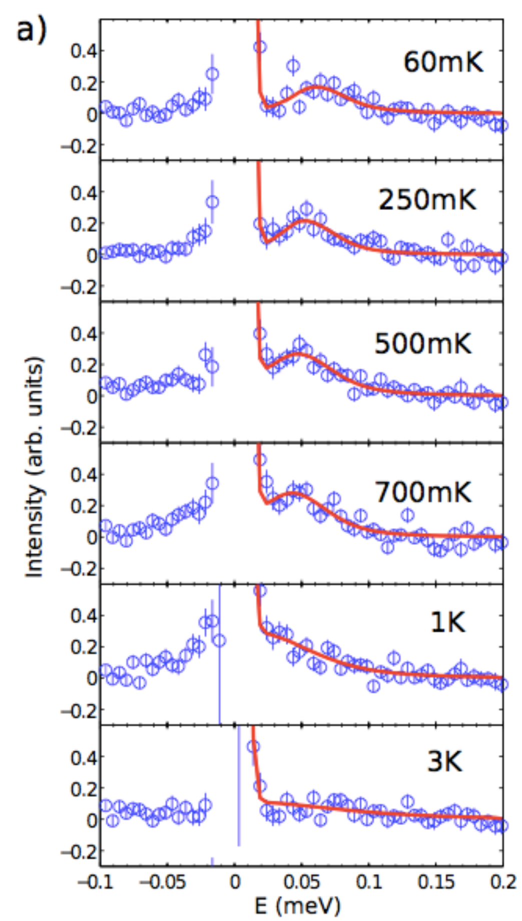
H = 0T

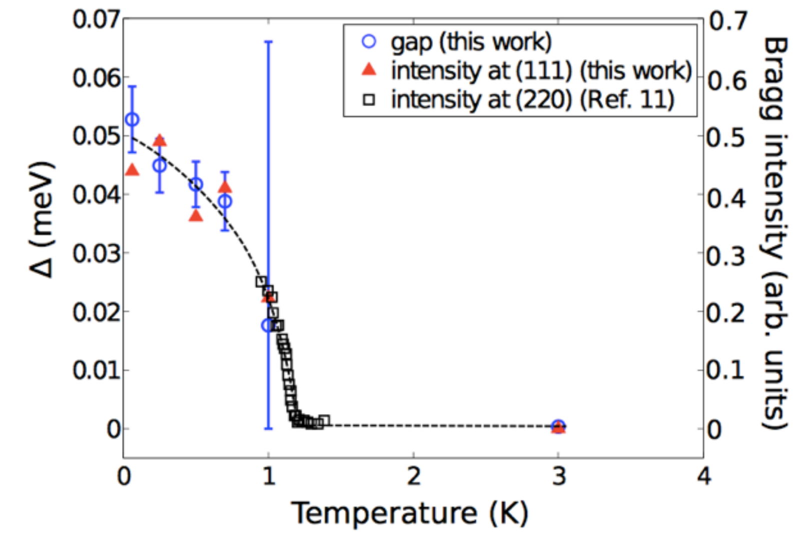


States selected by Order by Disorder show better agreement

A Very Small Gap Exists!

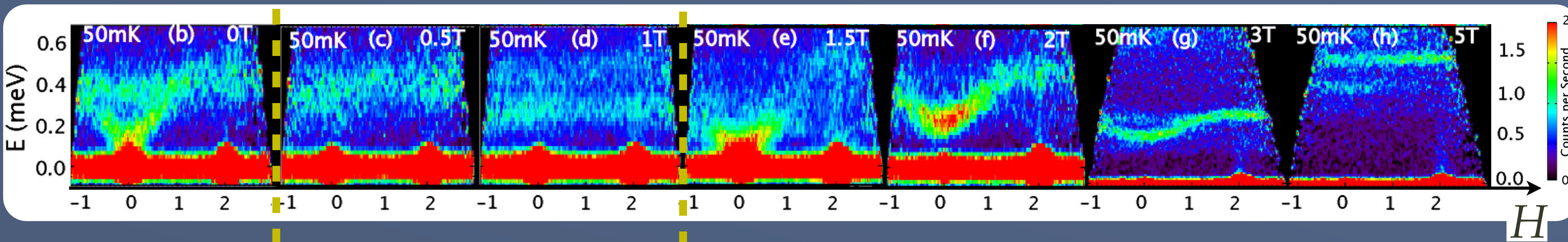


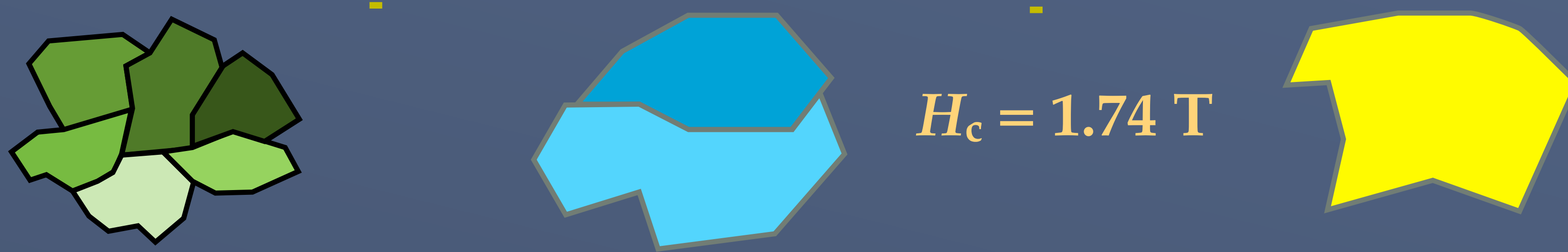




Extremely high energy resolution measurements
at the NCNR (NIST)

24 hours of counting on a 7 gram crystal



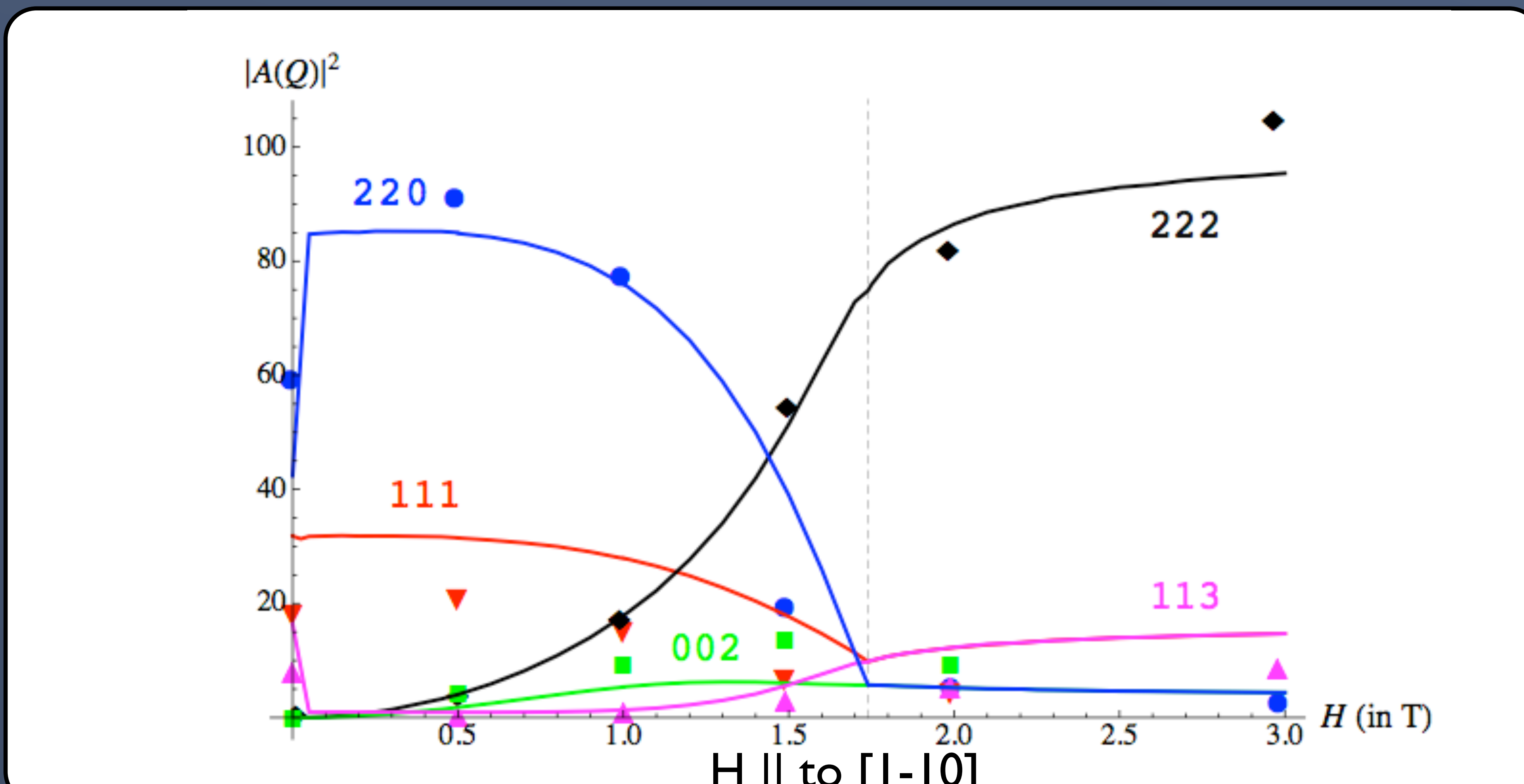


$H_c = 1.74$ T

6 domains

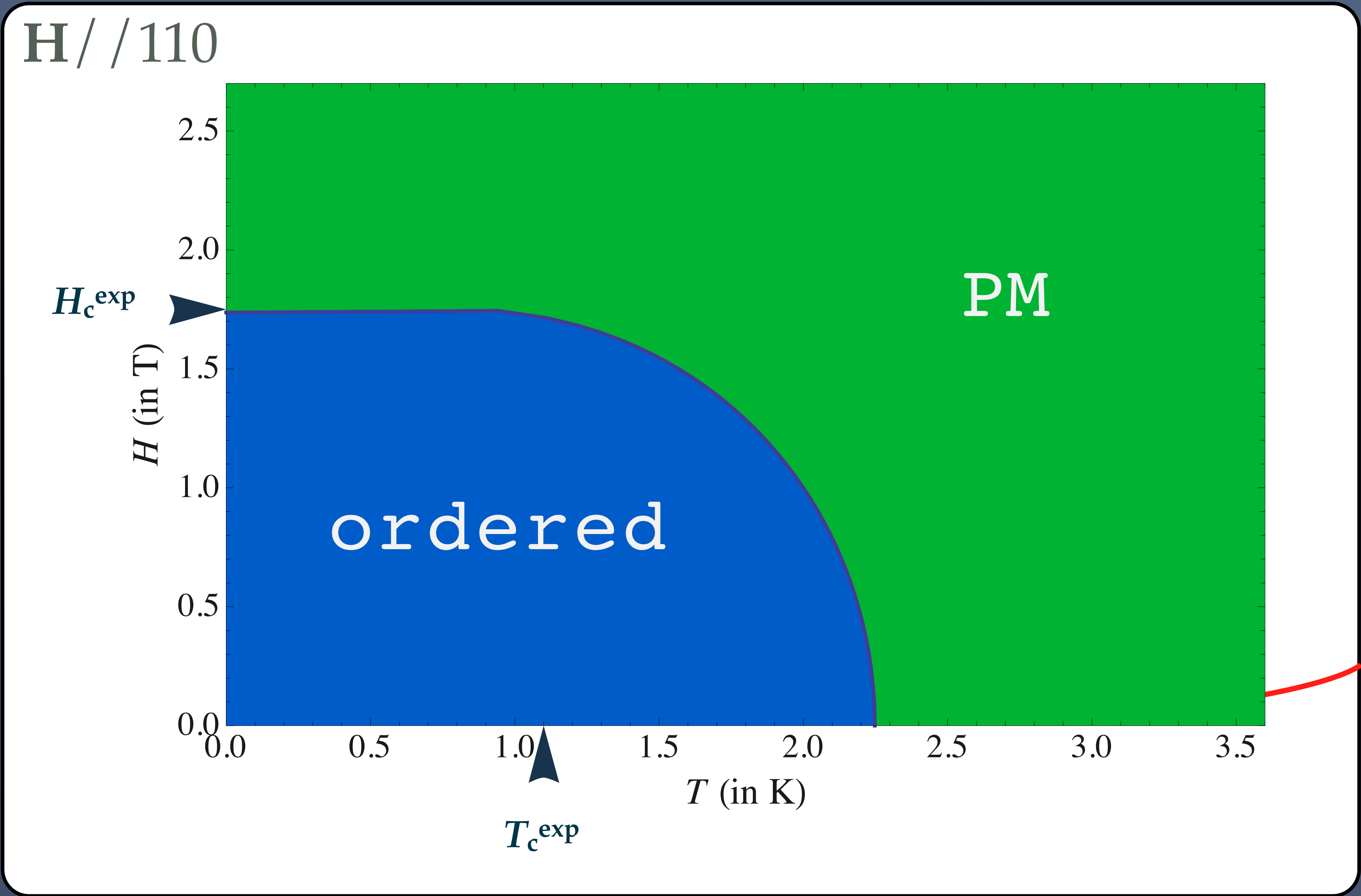
2 domains

1 domain



MFT phase diagram: $Er_2Ti_2O_7$

Little suppression of T_c due to frustration, fluctuations

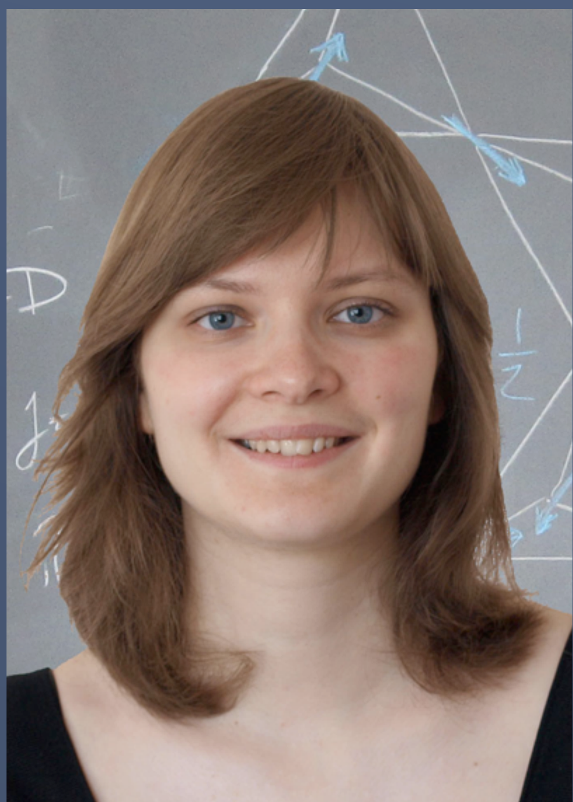


MFT Transition from spinwave fits

$T_C^{MF}/T_C^{exp} \sim 2.1$ $H_C^{MF} = H_C^{exp}$!

Collaboration

Lucile Savary



Leon Balents

Kate Ross

Edwin Kermarrec

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K.A. Ross, L. Savary, B. D. Gaulin, and L. Balents,
Quantum Excitations in Quantum Spin Ice, Phys. Rev. X **1**, 021002 (2011).

L. Savary, K. A. Ross, B.D. Gaulin, J.P.C. Ruff, and L. Balents,
Order by Quantum Disorder in $Er_2Ti_2O_7$, Phys. Rev. Lett. 109, 167201 (2012).

K. A. Ross, Y. Qiu, J.R.D. Copley, H.A. Dabkowska, and B.D. Gaulin,
Order by Quantum Disorder Spin Wave Gap in $Er_2Ti_2O_7$, Phys. Rev. Lett. 112, 057201 (2013).