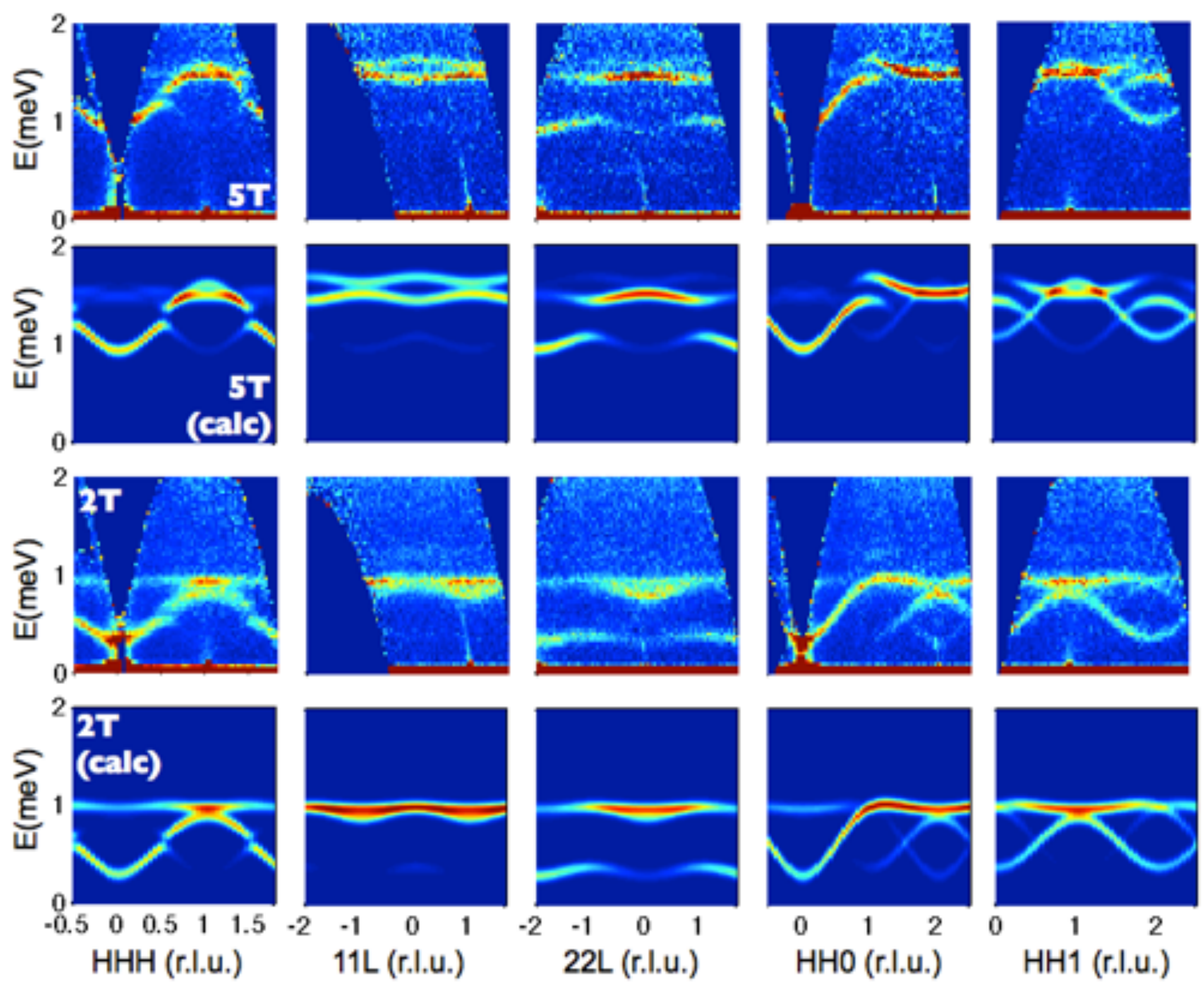
Ground State Selection in Quantum Pyrochlore Magnets



J. Gaudet [1]
E. Kermarrec [1, 2]
A.M. Hallas [1]
K.A. Ross [1, 3]
G. Sala [1]
K. Fritsch [1, 4]
G.M. Luke [1]
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J.P.C. Ruff [1, 5]
M.M.P. Couchman [1]
H.A. Dabkowska [1]
D. Pomaranski [6]
J.B. Kycia [6]
M.A. White [7]
C.R. Wiebe [1, 8]
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[7] Dalhousie University
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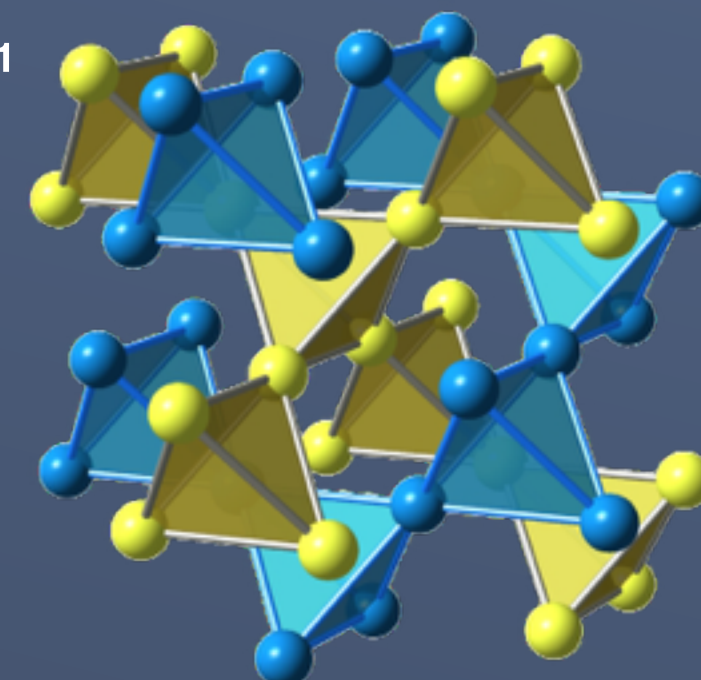
[10] MIT



Bruce D. Gaulin
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Collaborators at International Neutron and Muon Sources

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G.E. Granroth
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NIST Center for Neutron Research

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J.R.D. Copley

ILL Neutrons for Science

C. Ritter

Science & Technology Facilities Council ISIS

R.I. Bewley
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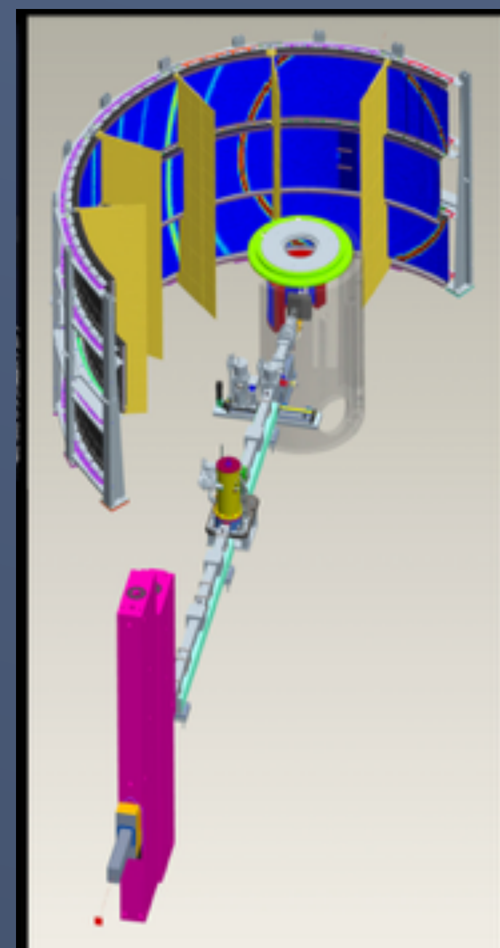

Paul Scherrer Institut PSI

R. Khasanov
Z. Guguchia

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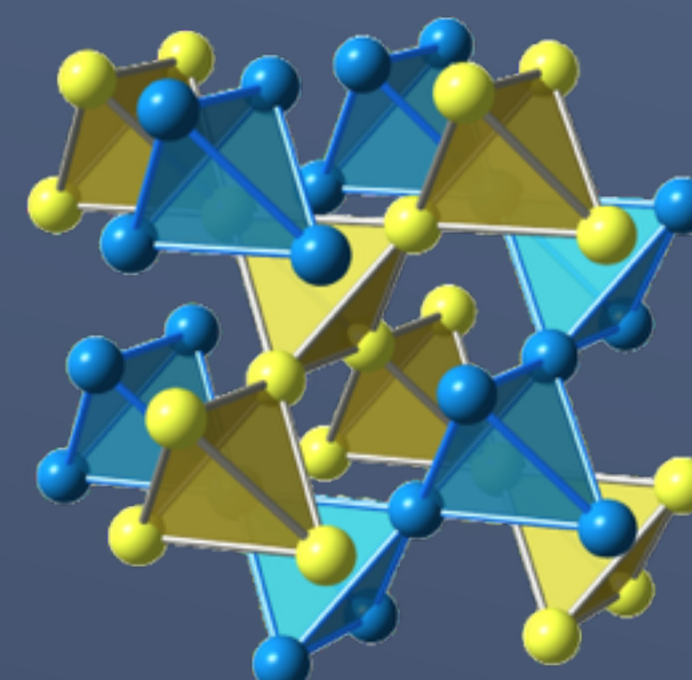
B. Hitti



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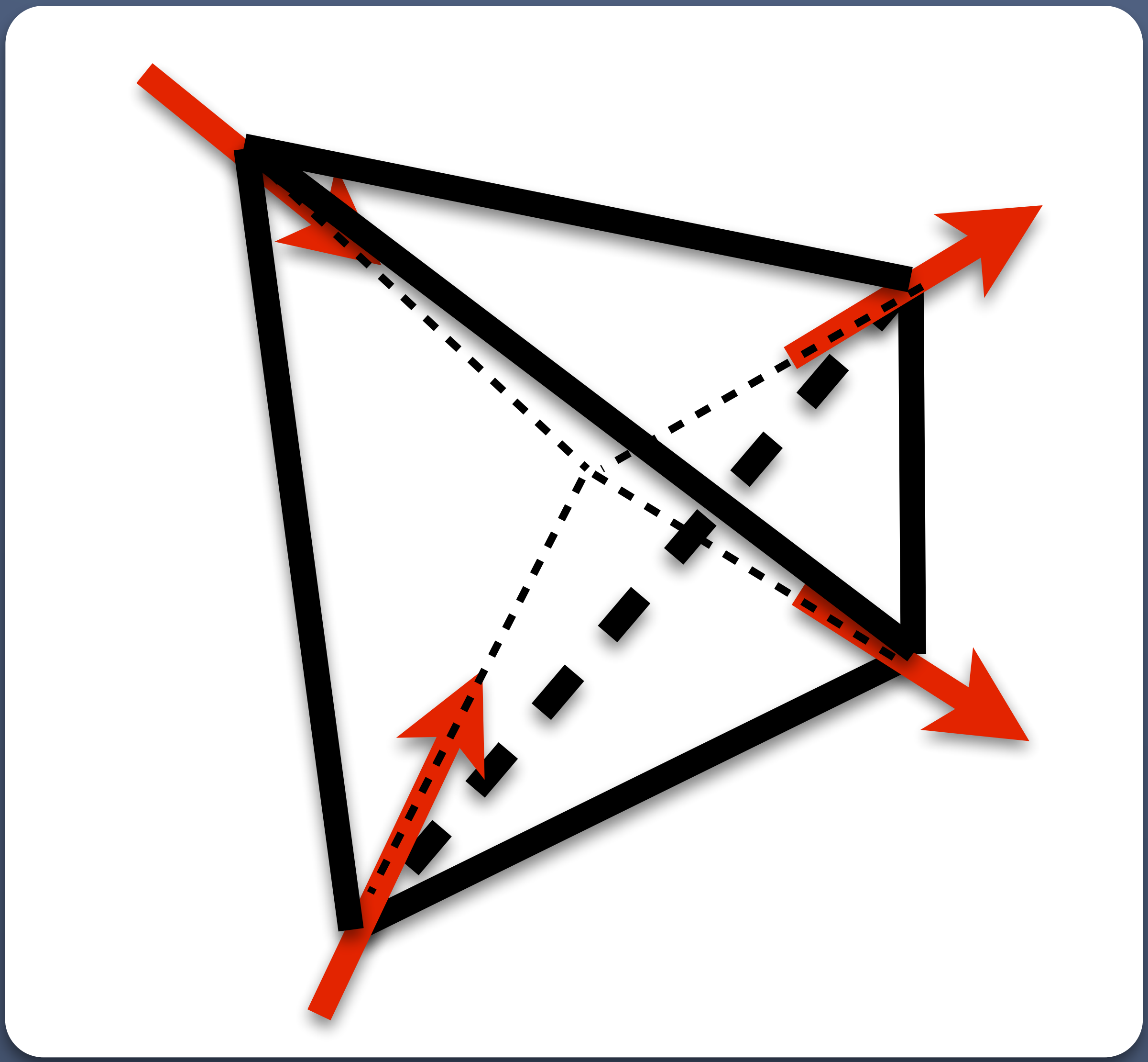
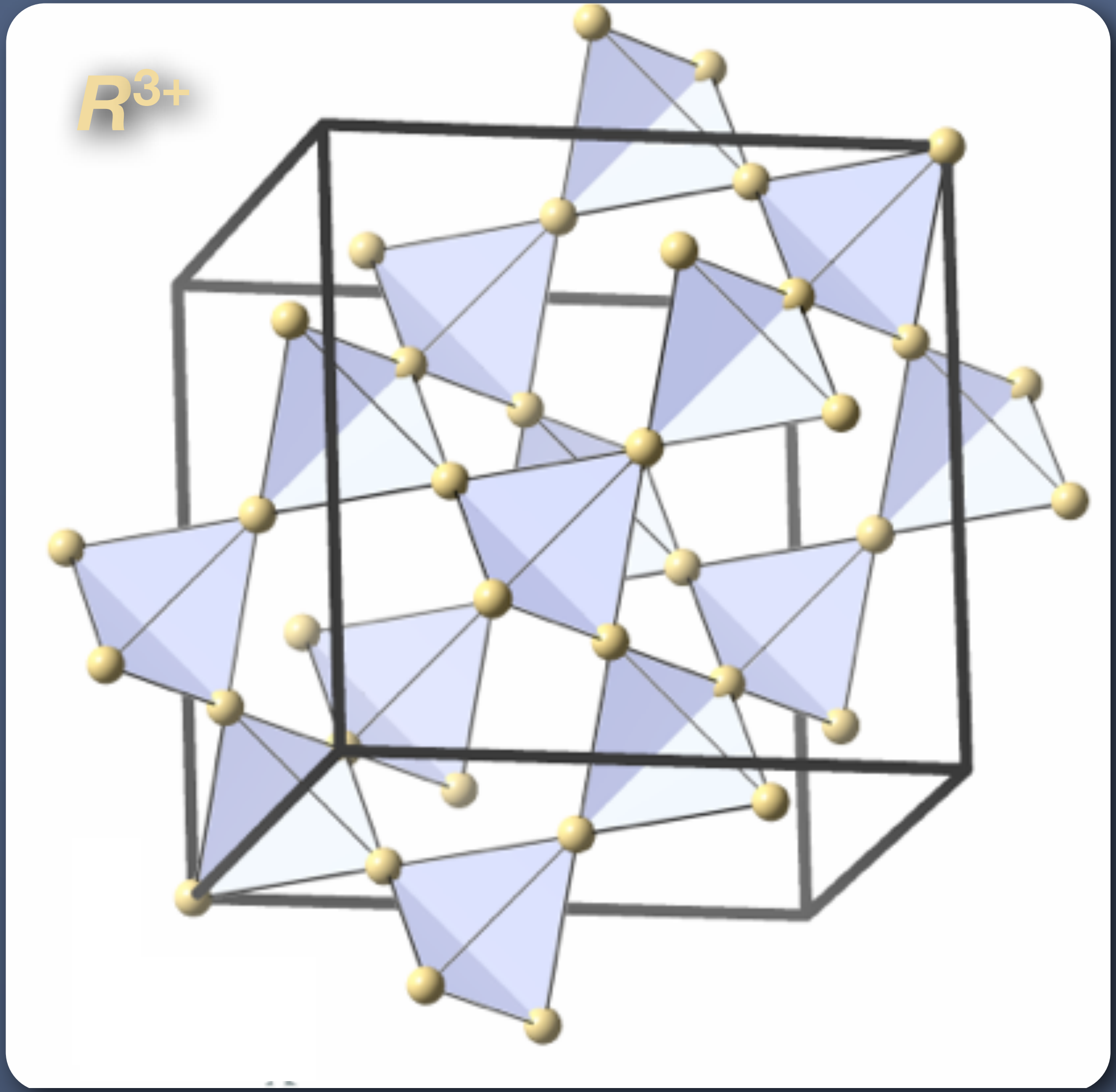
Physics of Frustration in 3D on Tetrahedra

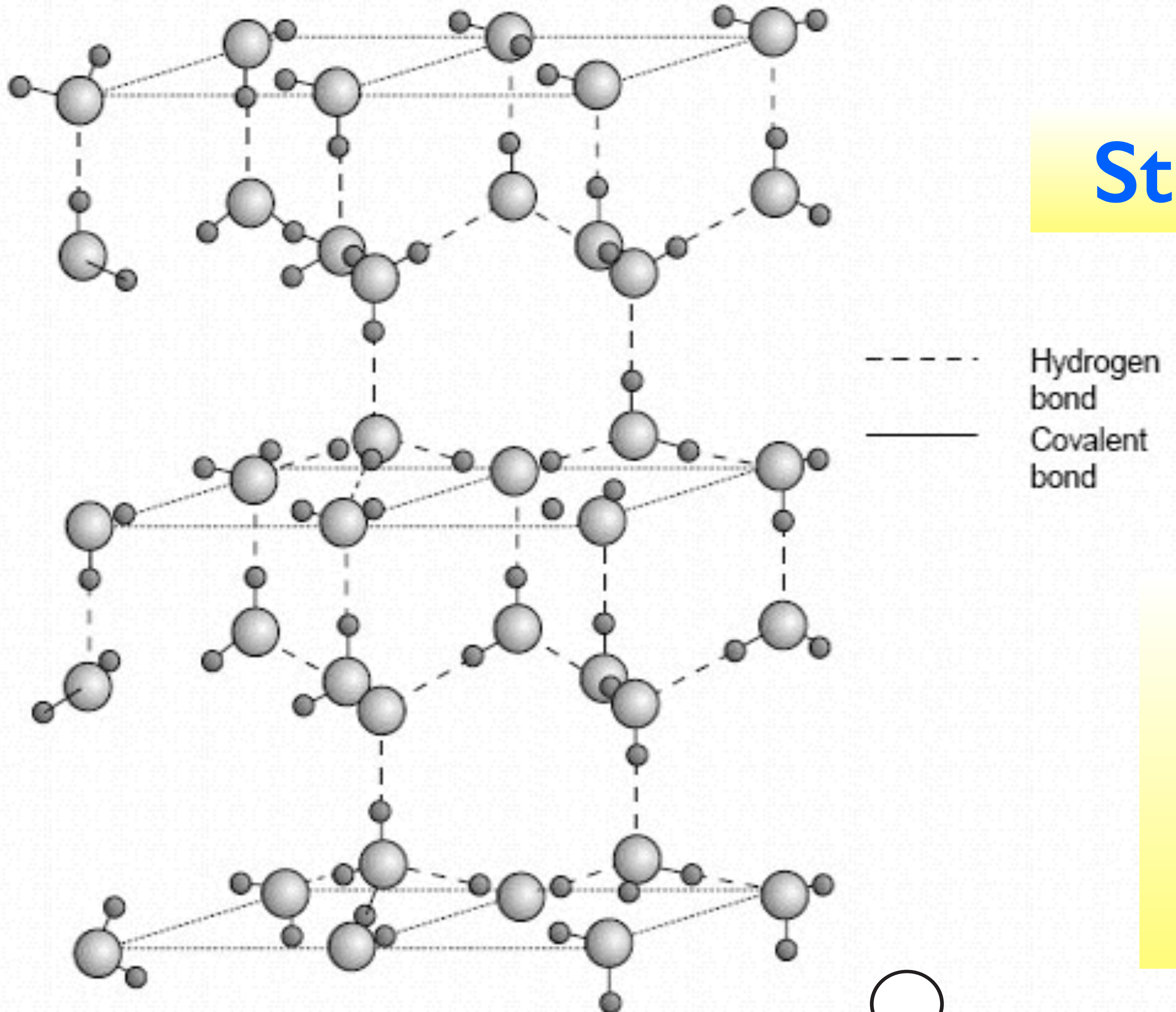




Cubic Pyrochlore Lattice

***Spin Ice*: **Ferromagnetic interactions combined with local Ising anisotropy leads to 6-fold degeneracy on a single tetrahedron - macroscopic degeneracy on 3D crystal

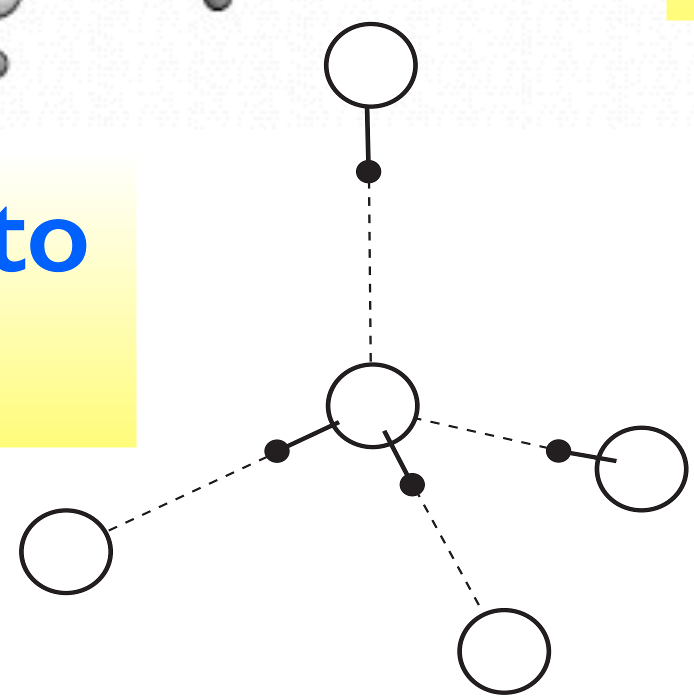
M.J. Harris, S.T. Bramwell, D.F. McMorrow, T. Zeiske, and K.W. Godfrey, PRL 79 2554 (1997)

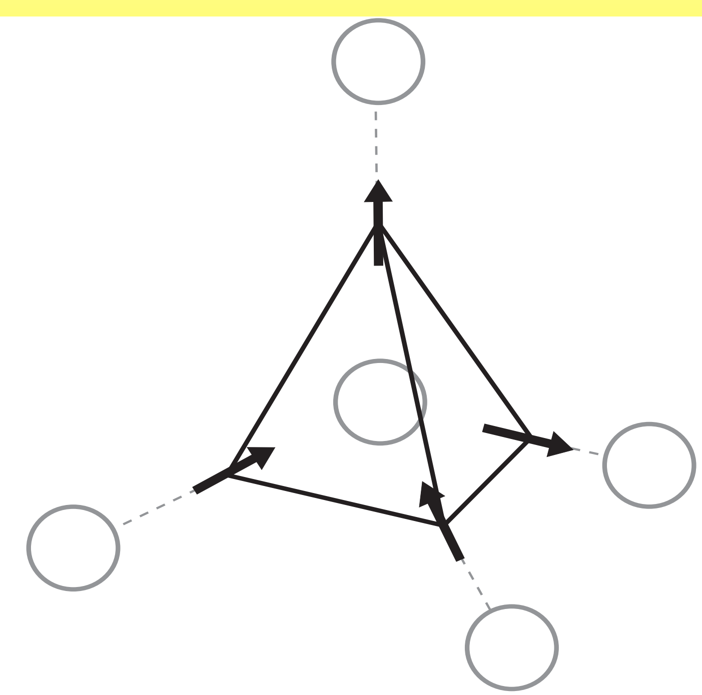


Structure of Ice

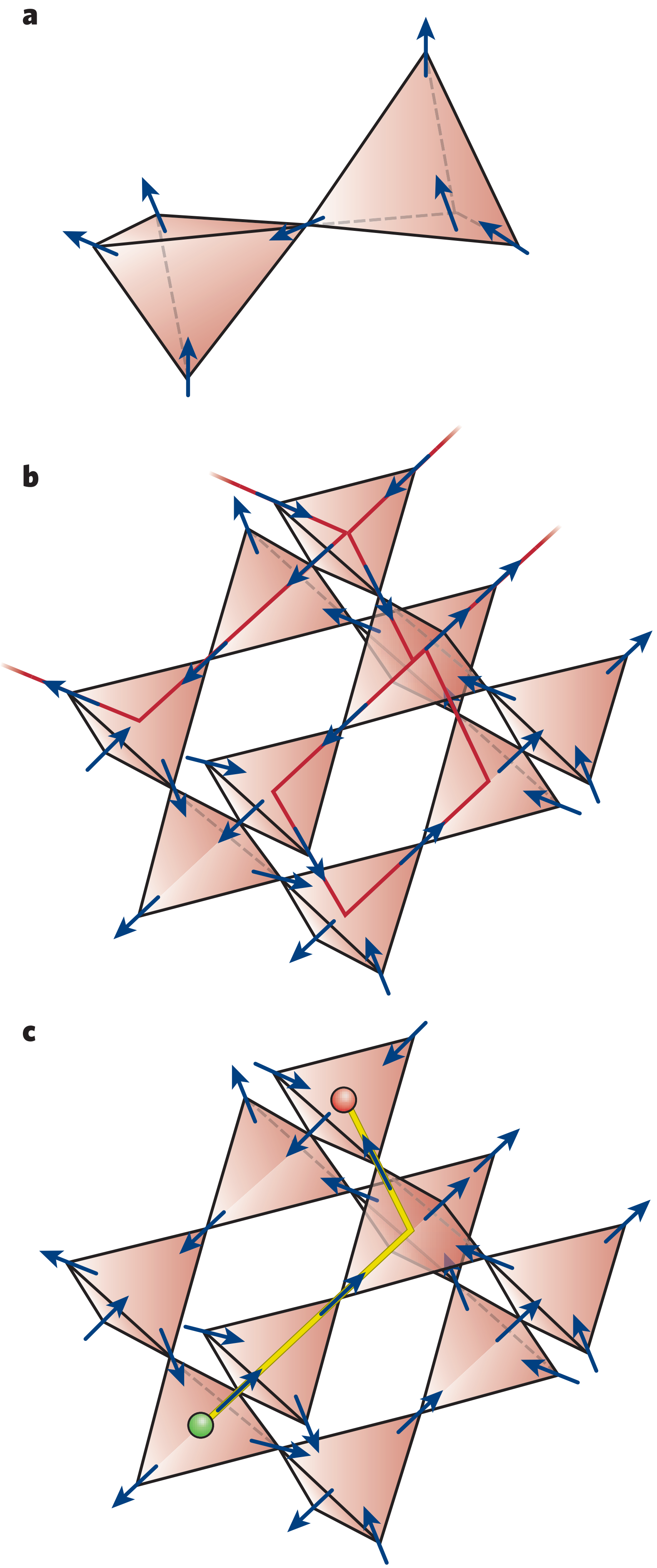
Ferro coupling + [111] anisotropy “2 in 2 out” 6-fold degenerate

Correspondance to Spin Ice

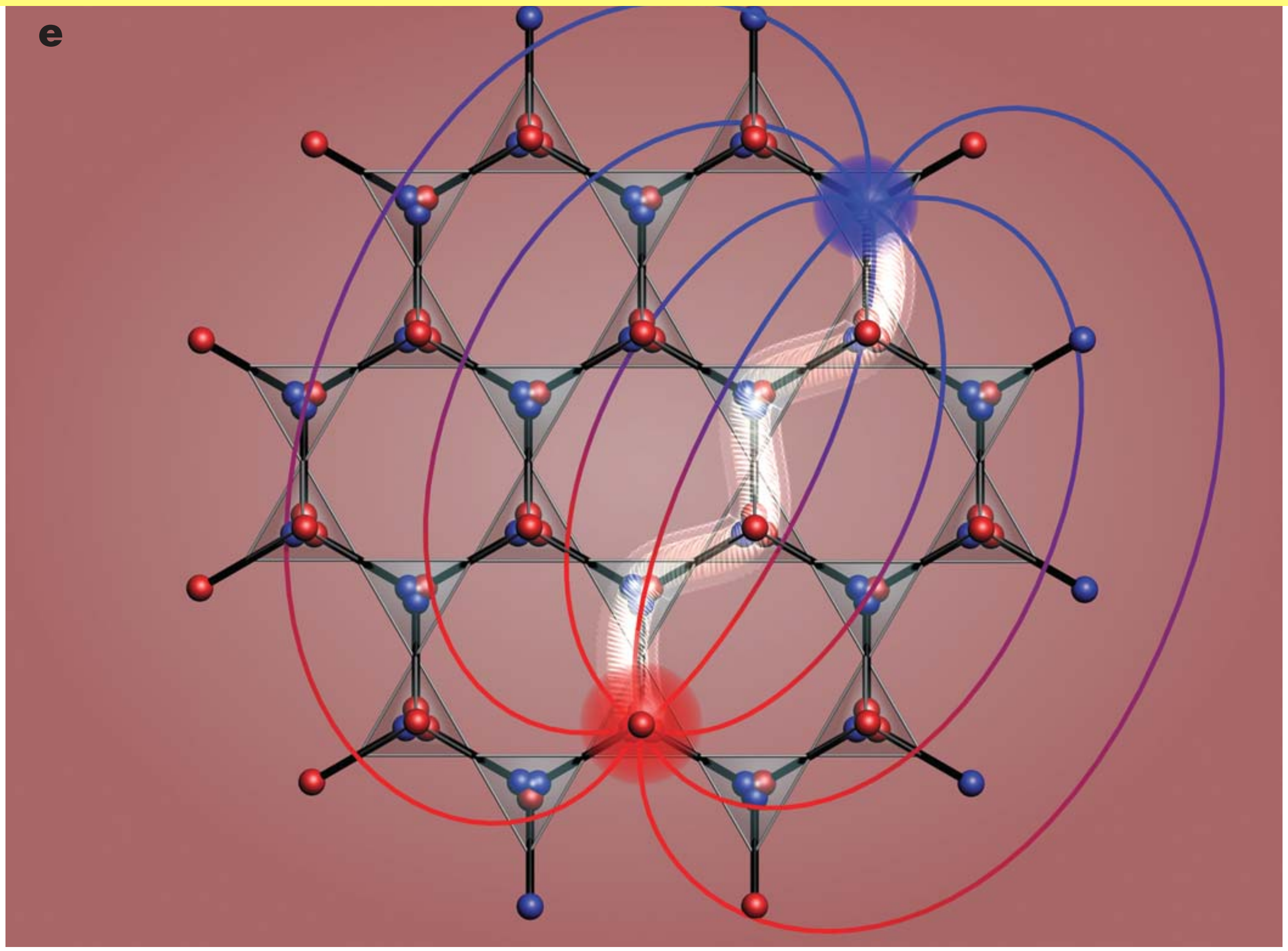




Spin Ice



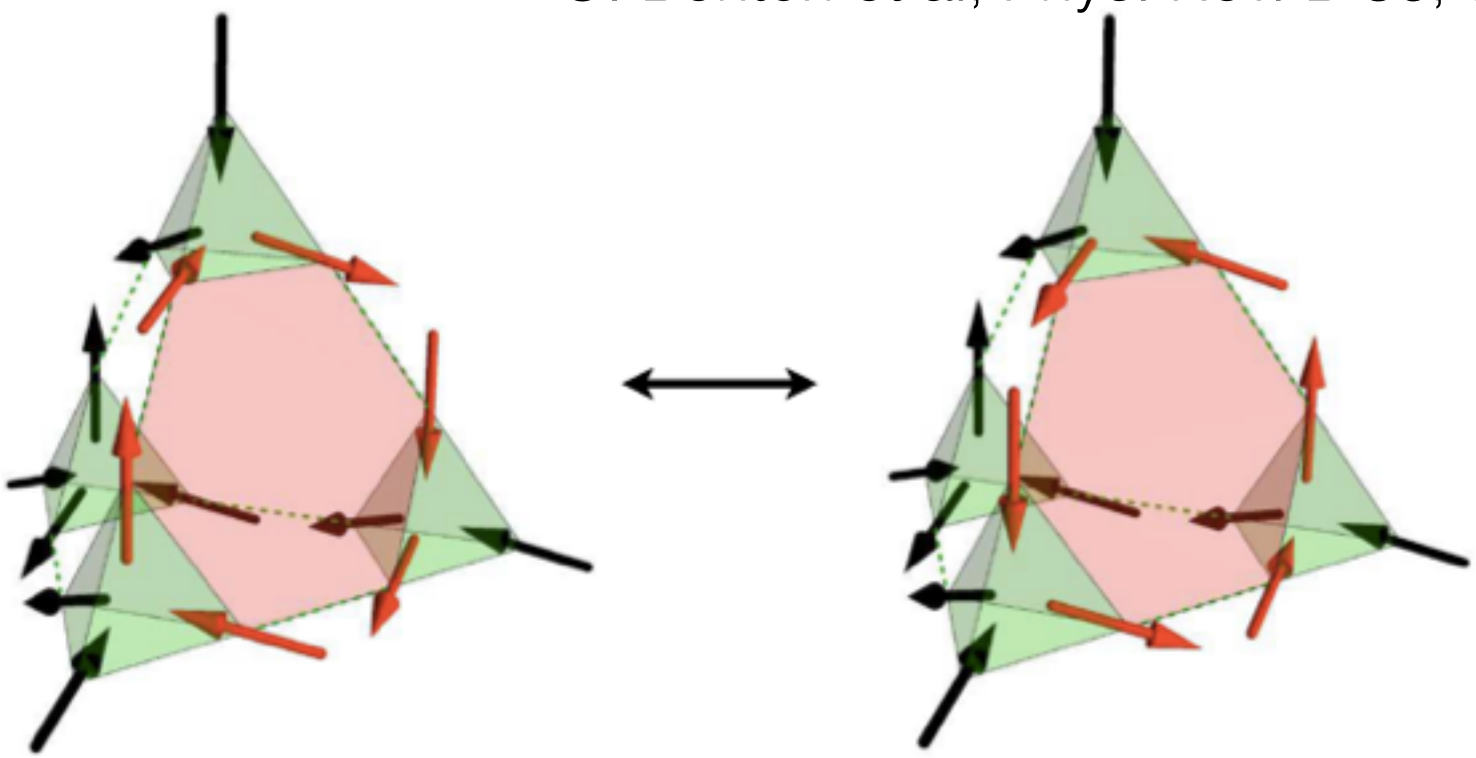
- Classical macroscopic degeneracy
- Supports monopole excitations
- Rare example of deconfined excitations in 3D



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**, 075154, 2012



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

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

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

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

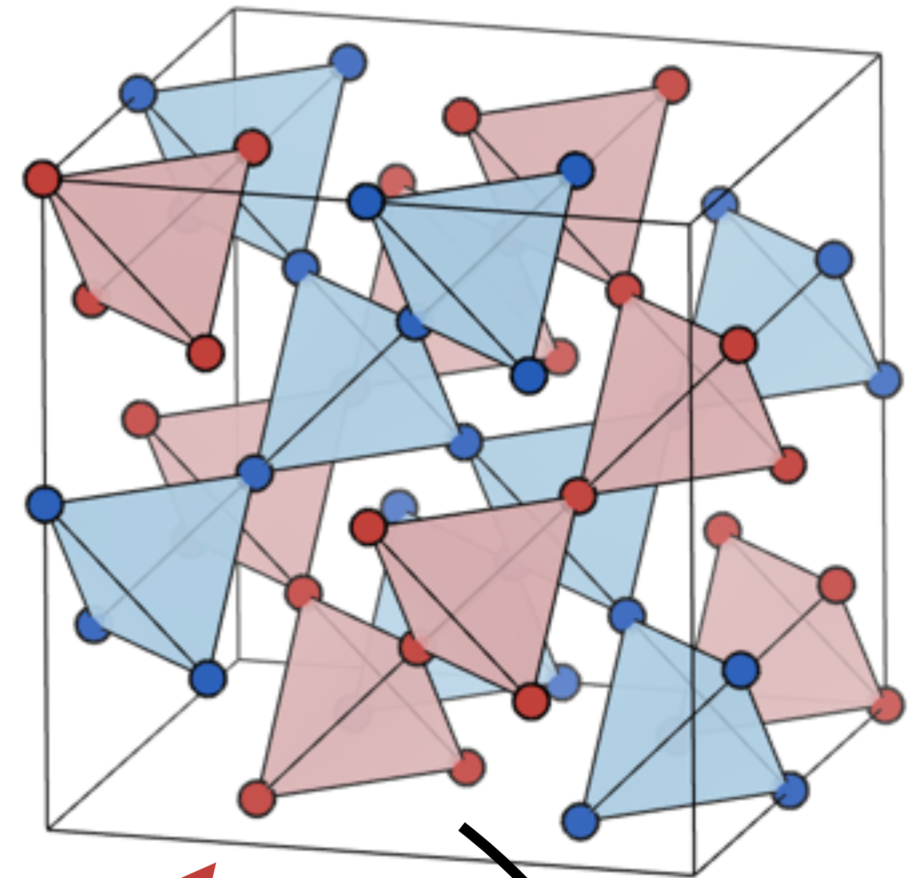
- Can tunnel between ice rules states
- Introduces *fluctuations* in the gauge field
    - **Electric monopoles —** coherent, propagating wavepacket of ice configurations
    - **Magnetic monopoles —** violate ice rules, i.e. 3-in 1-out
    - **Gauge photons —** transverse fluctuations of gauge field

Pyrochlores have the quintessential lattice for the phenomena of magnetic frustration in 3D.

$(Er^{3+})_2(Ti^{4+})_2O_7$
4f^{11} J=15/2

$(Yb^{3+})_2(Ti^{4+})_2O_7$
4f^{13} J=7/2

H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
				Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
				Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

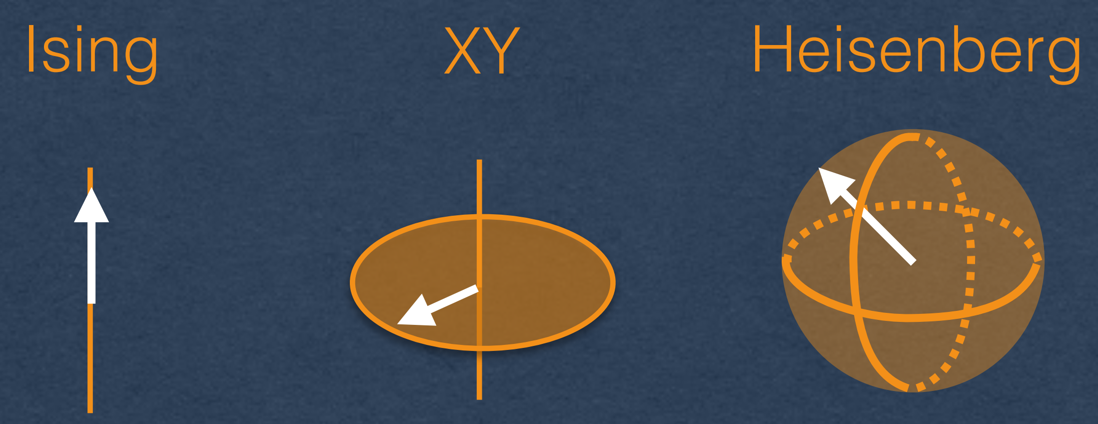


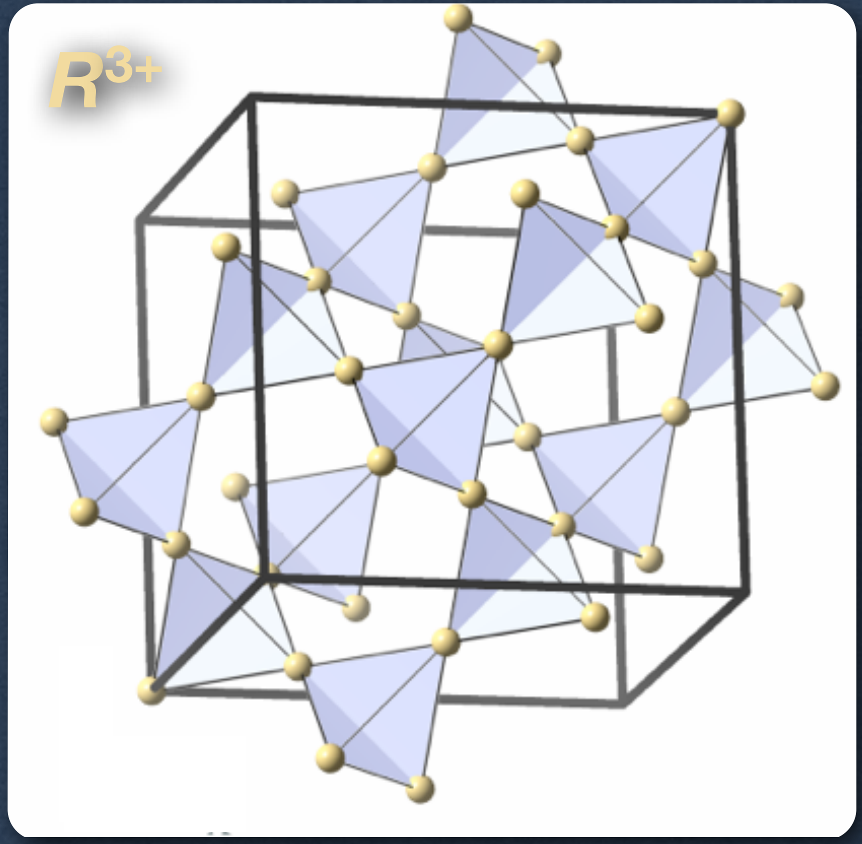
...
....
"disorder-free" spin glass
spin ice
metal-insulator transitions
spin liquid
order-by-disorder
fragmentation

Gardner, Gingras, and Greedan, Rev. Mod. Phys **82**, 53 (2010)
Wiebe and Hallas, APL Materials **3**, 041519 (2015)

Real Pyrochlores: playgrounds for frustration

$R_2Ti_2O_7$ "Rare earth titanates"



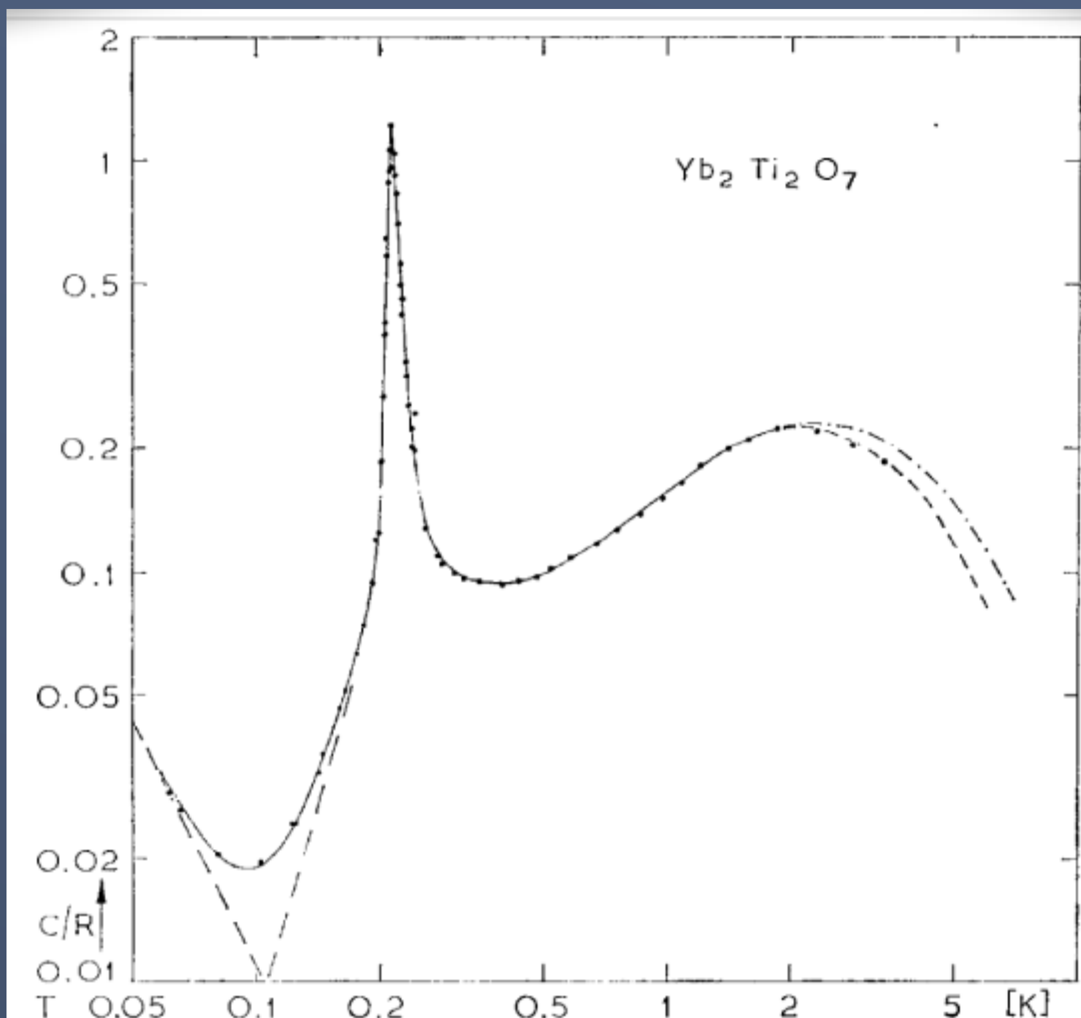


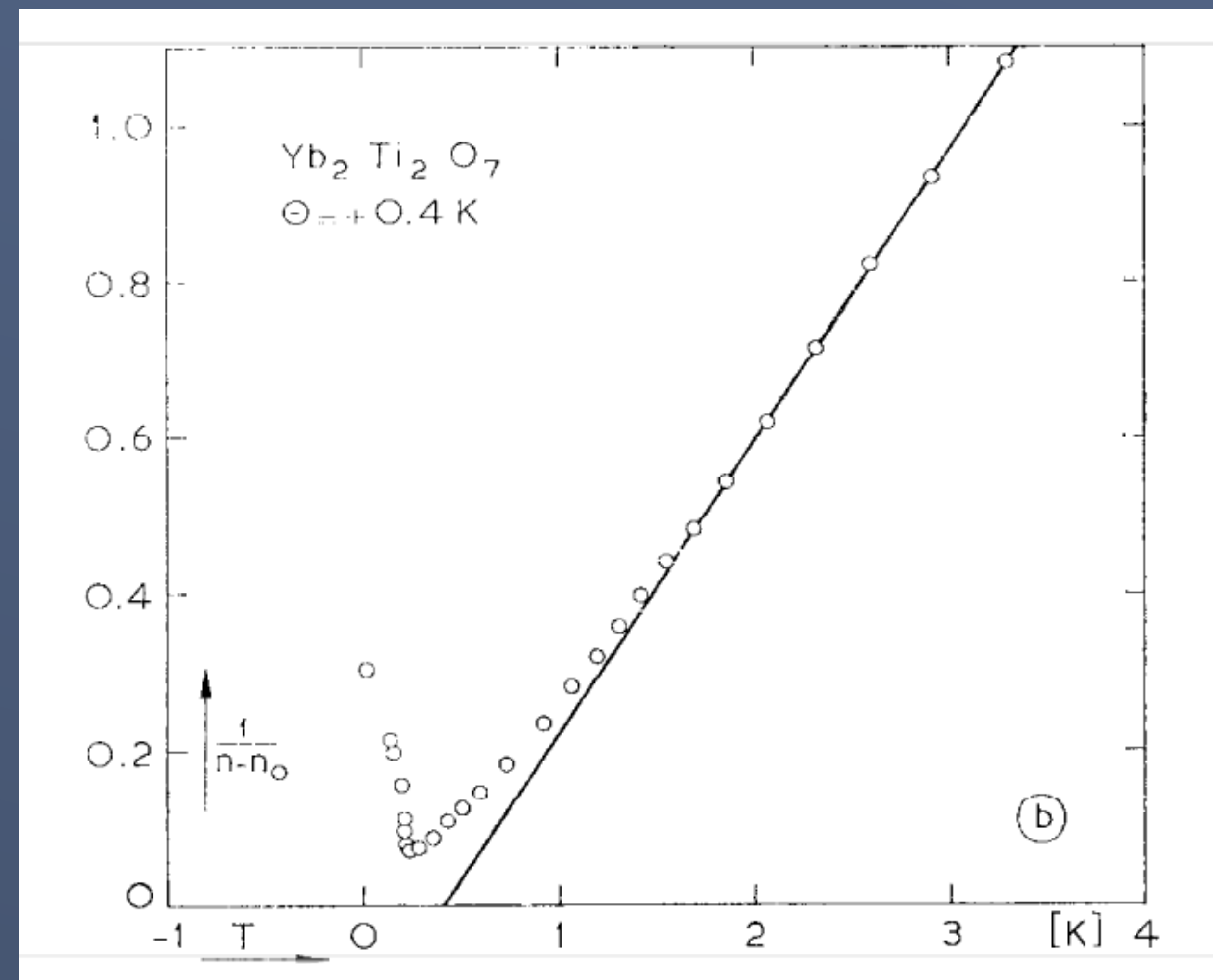
Anisotropy and ***Interactions*** combine for diverse exotic ground states

	Single Ion Anisotropy	Interactions	Ground state
Ho, Dy	Ising	FM	spin ice
Tb	Ising	AF	"quantum spin ice"
Gd	Heisenberg	AF	partial order
Er	**XY**	**AF**	**Neel state via ObD**
Yb	**XY**	**FM**	**"quantum spin ice"**

$Yb_2Ti_2O_7$: An XY Ferromagnetic Pyrochlore

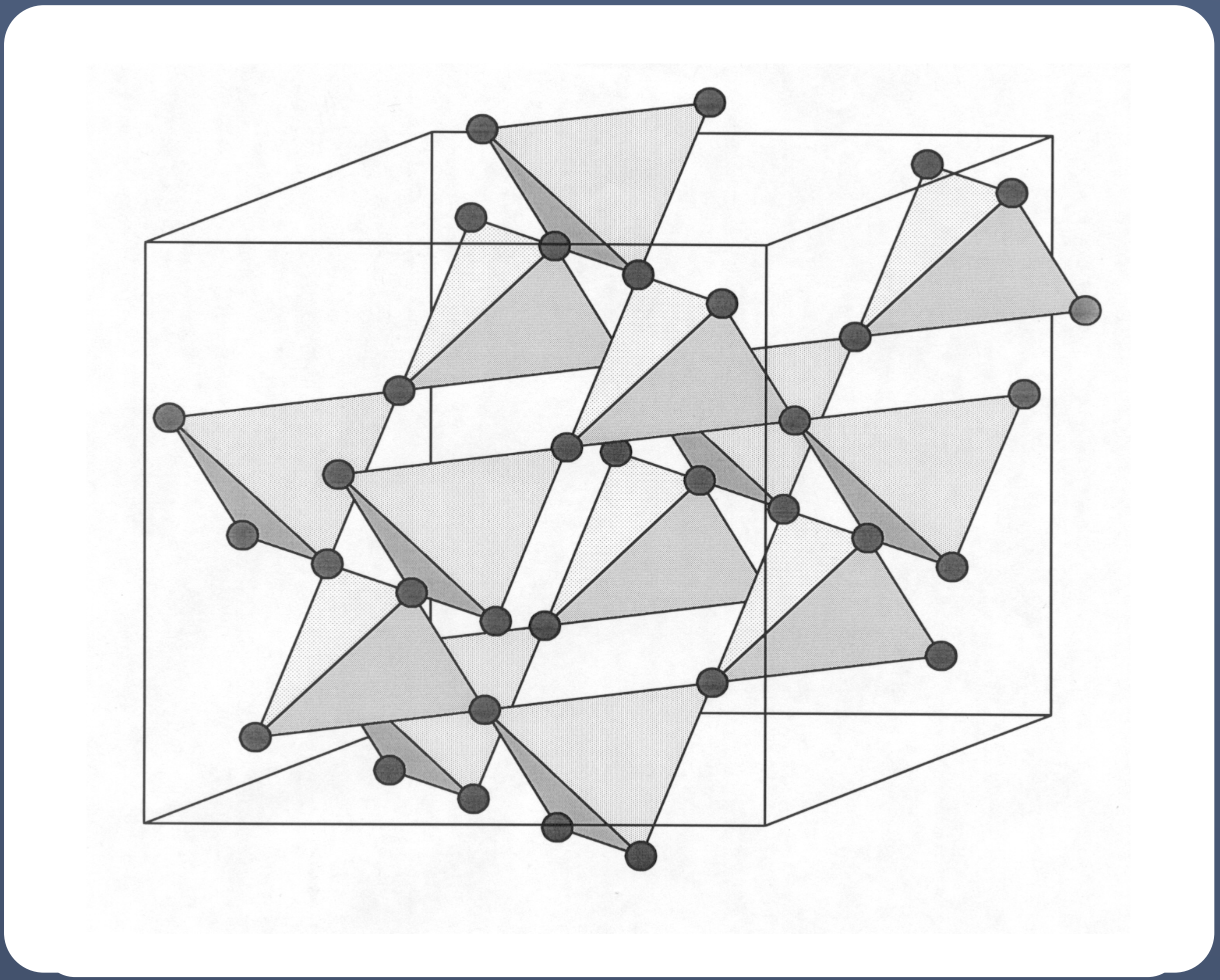
H. W. J. Blöte, R. F. Wielinga and W. J. Huiskamp, Physica 43, 549 (1969).

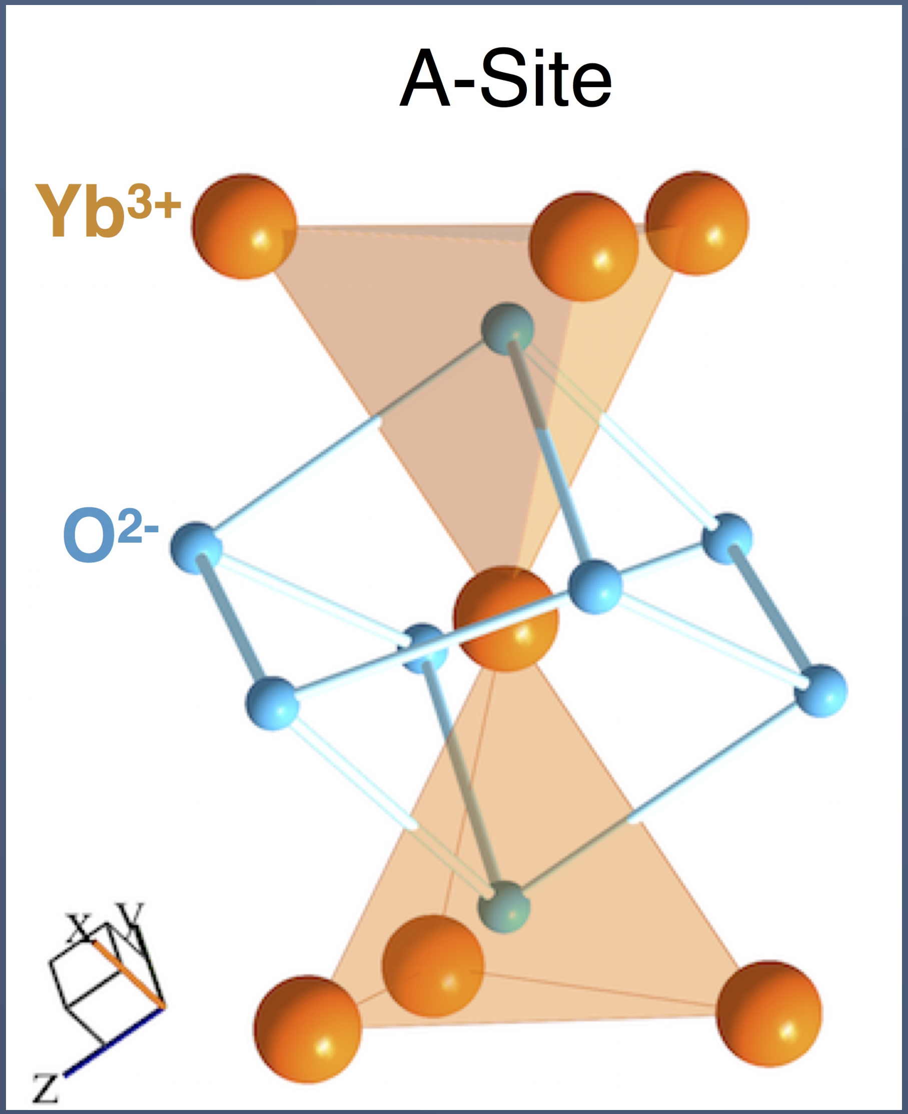




- Ferromagnetic "XY" pyrochlore
- " T_C " ~ 240 mK
- C_P (T) has 2 peaks: one at "T_C" ; one at ~ 3 K
- CW_T ~ +0.5 K $g_{\perp}/g_{||}$ ~ 2

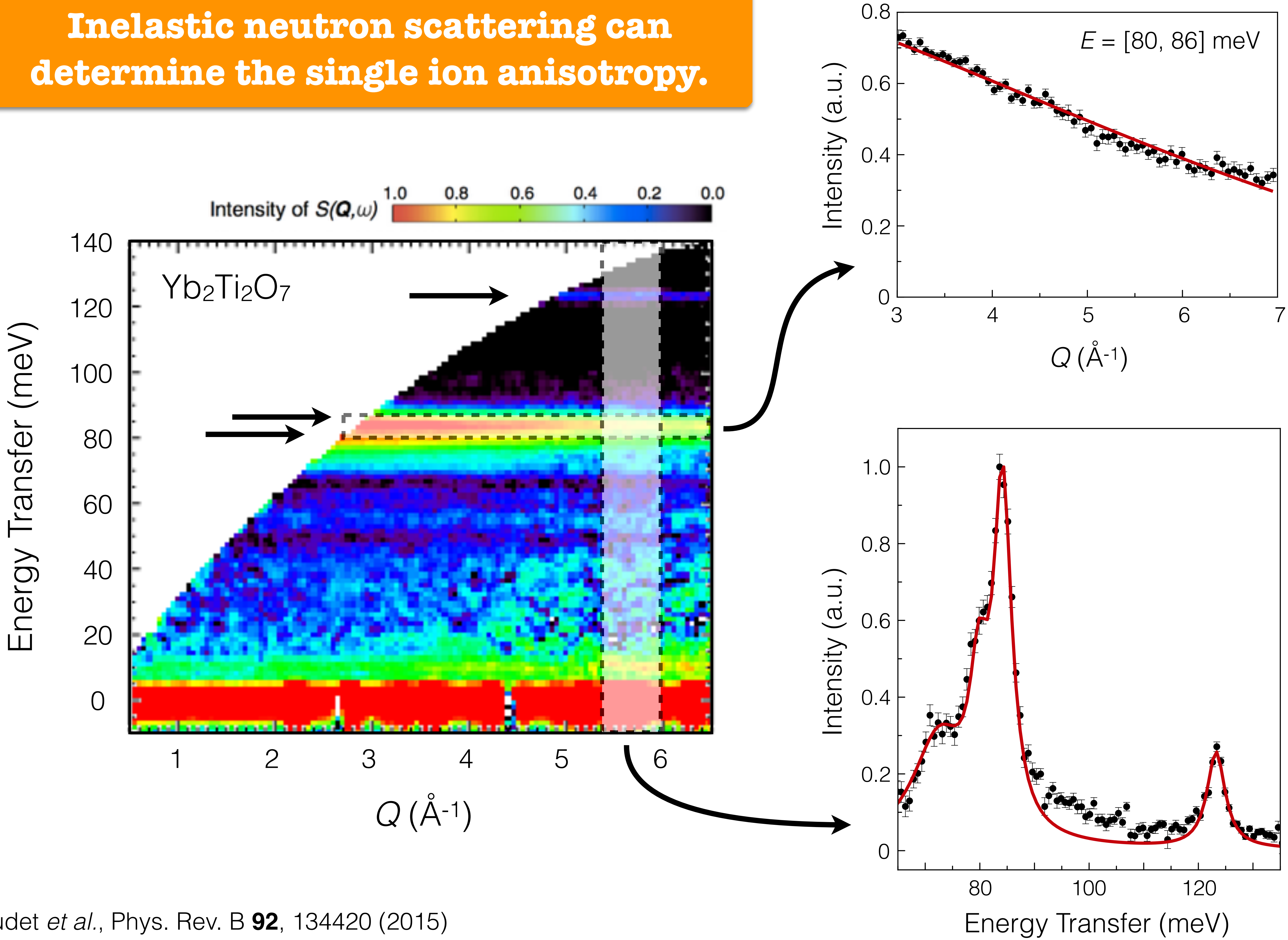
Crystal Field Environment at the RE Site





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

Inelastic neutron scattering can determine the single ion anisotropy.



Gaudet *et al.*, Phys. Rev. B **92**, 134420 (2015)
Hallas *et al.*, Phys. Rev. B **93**, 104405 (2016)

$Yb_2Ti_2O_7$ Crystal Field Scheme

$Yb_2Ti_2O_7$

1278K

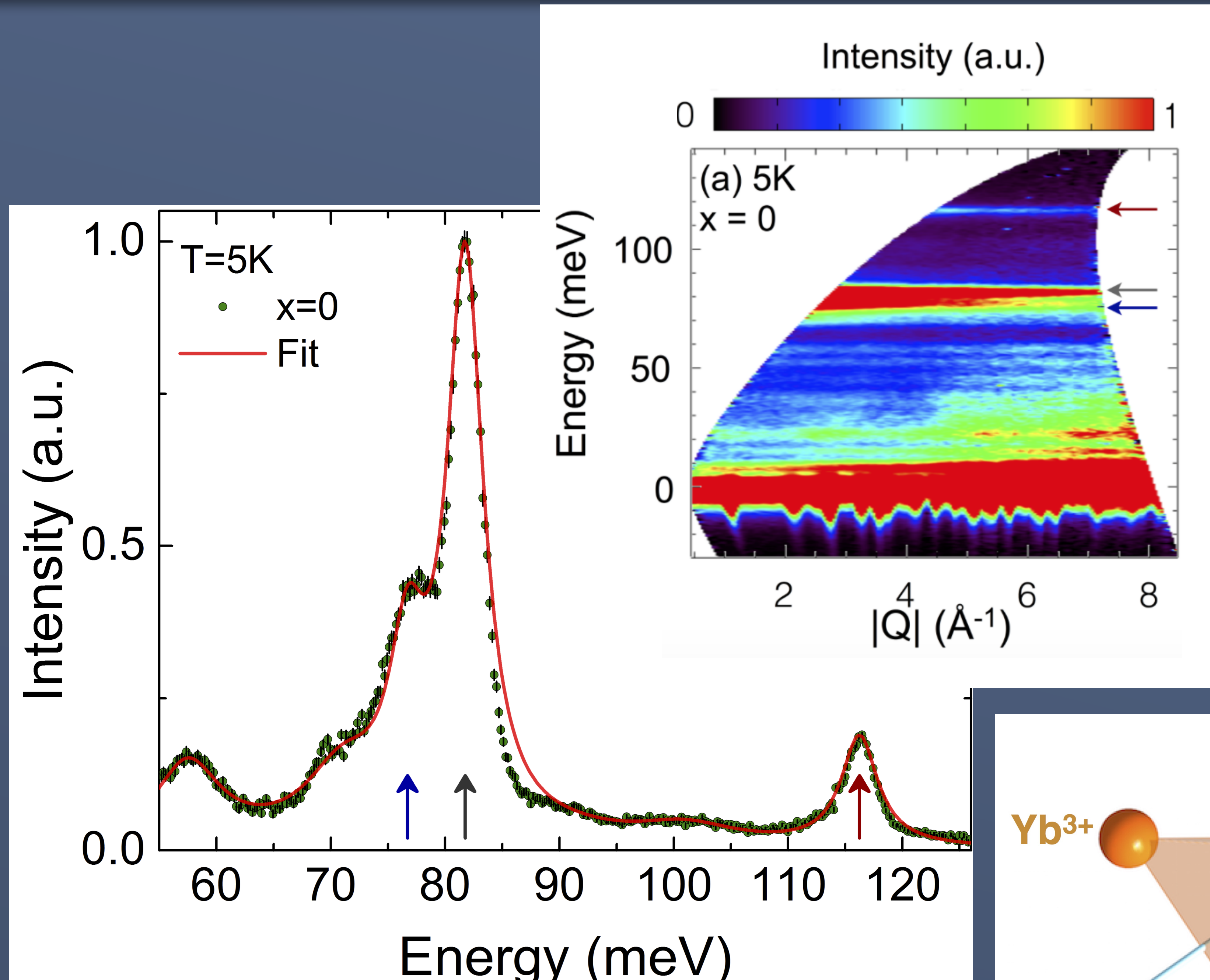
900K

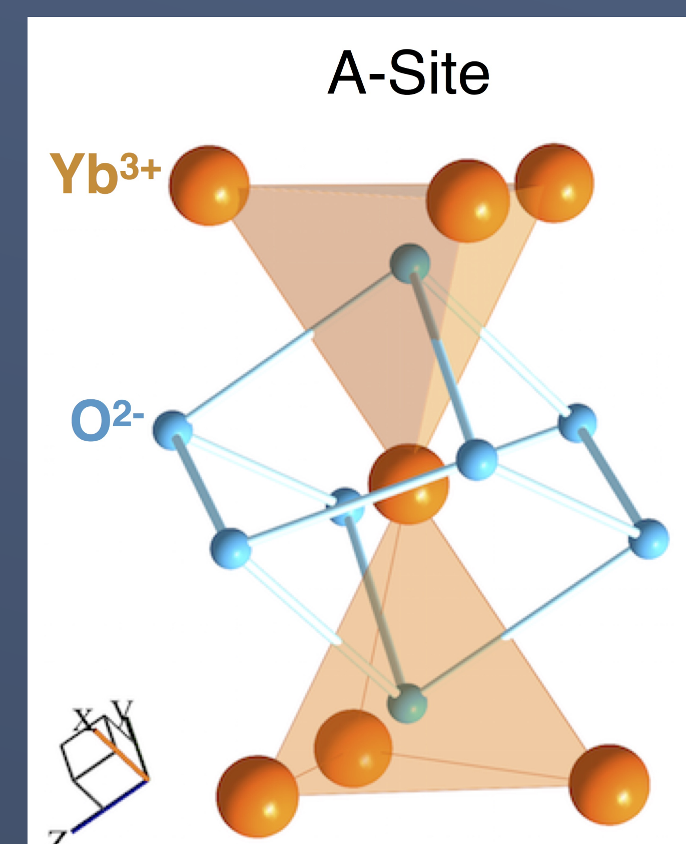
843K

$g_{||}$ = 1.92

$g_{\perp}$ = 3.69

4f [13] J = 7/2





Ground state wavefunctions:

$$|\phi^{\mp}\rangle = \boxed{0.91|\mp\tfrac{1}{2}\rangle} \pm 0.40|\mp\tfrac{7}{2}\rangle \pm 0.13|\pm\tfrac{5}{2}\rangle$$

J. Gaudet et al, PRB **92, 134420** (2015)

Crystal Field Effects

$Yb_2Ti_2O_7$

Gaudet et al, PRB **92**, **134420** (2015)

1278K

900K

843K

$g_{||} = 1.92$

$g_{\perp} = 3.69$

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

$Er_2Ti_2O_7$

Gaudet et al, unpublished.

900K

680K

250K

76K

$g_{||} = 3.91$

$g_{\perp} = 6.30$

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

$Dy_2Ti_2O_7$

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

550K

500K

450K

300K

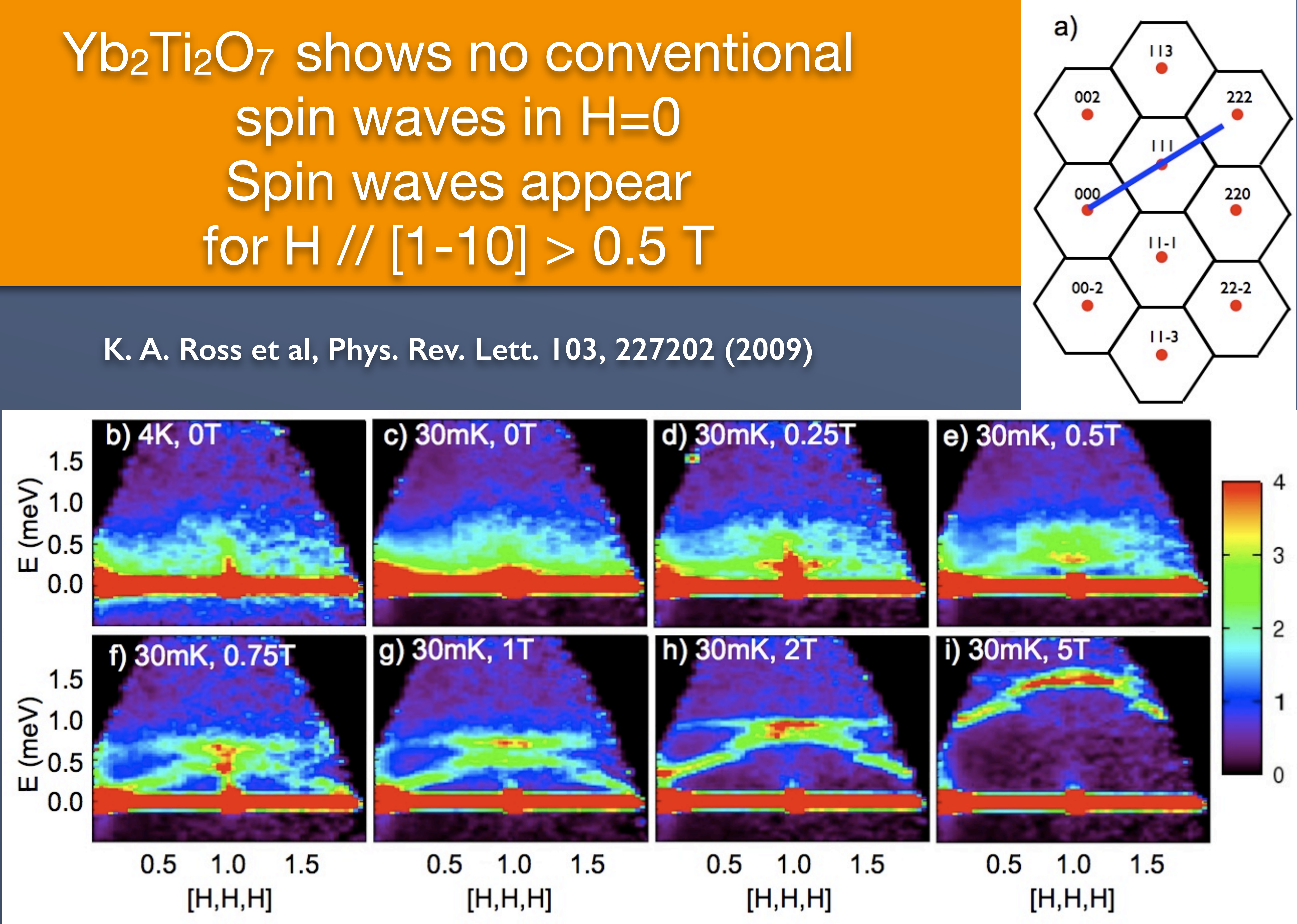
$g_{||} \sim 10$

$g_{\perp} \sim 0$

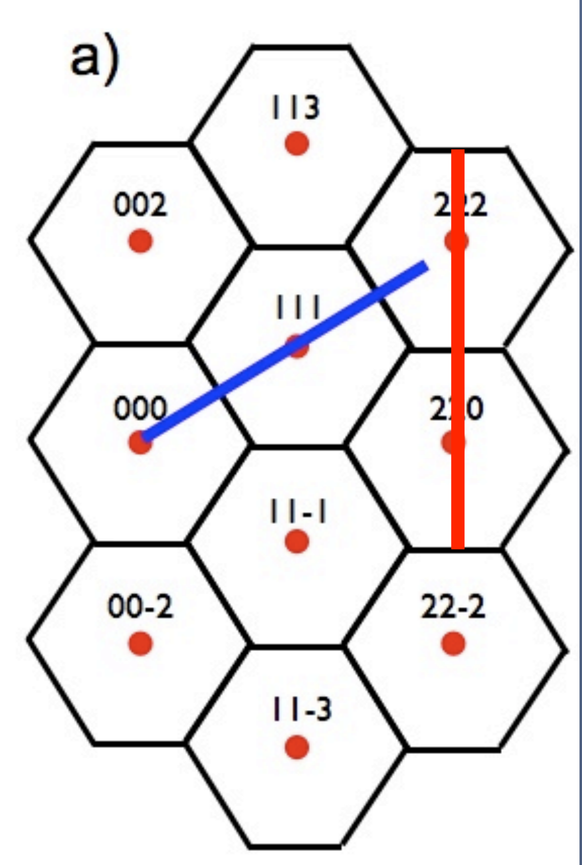
$4f^{9}$ J = 15/2

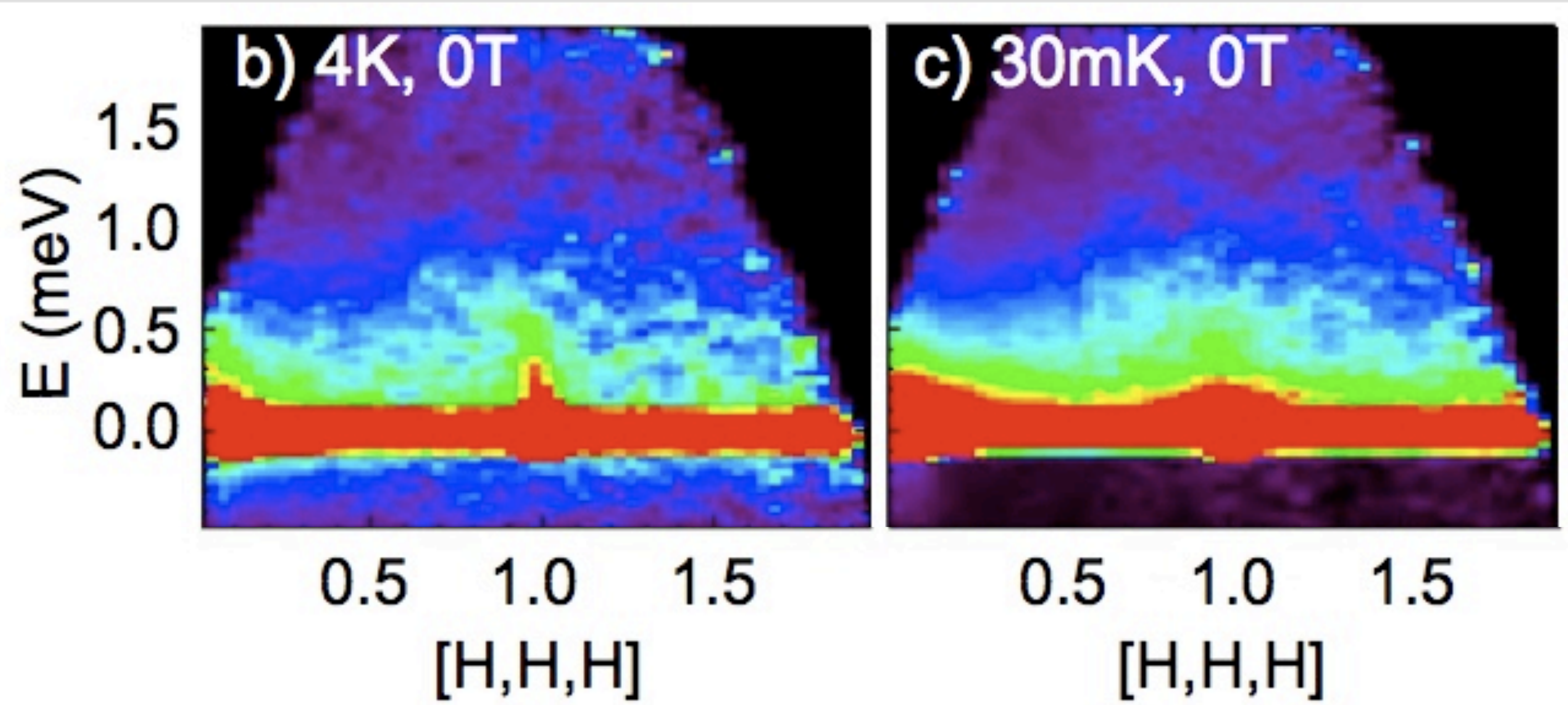
$Yb_2Ti_2O_7$ shows no conventional spin waves in H=0 Spin waves appear for H // [1-10] > 0.5 T

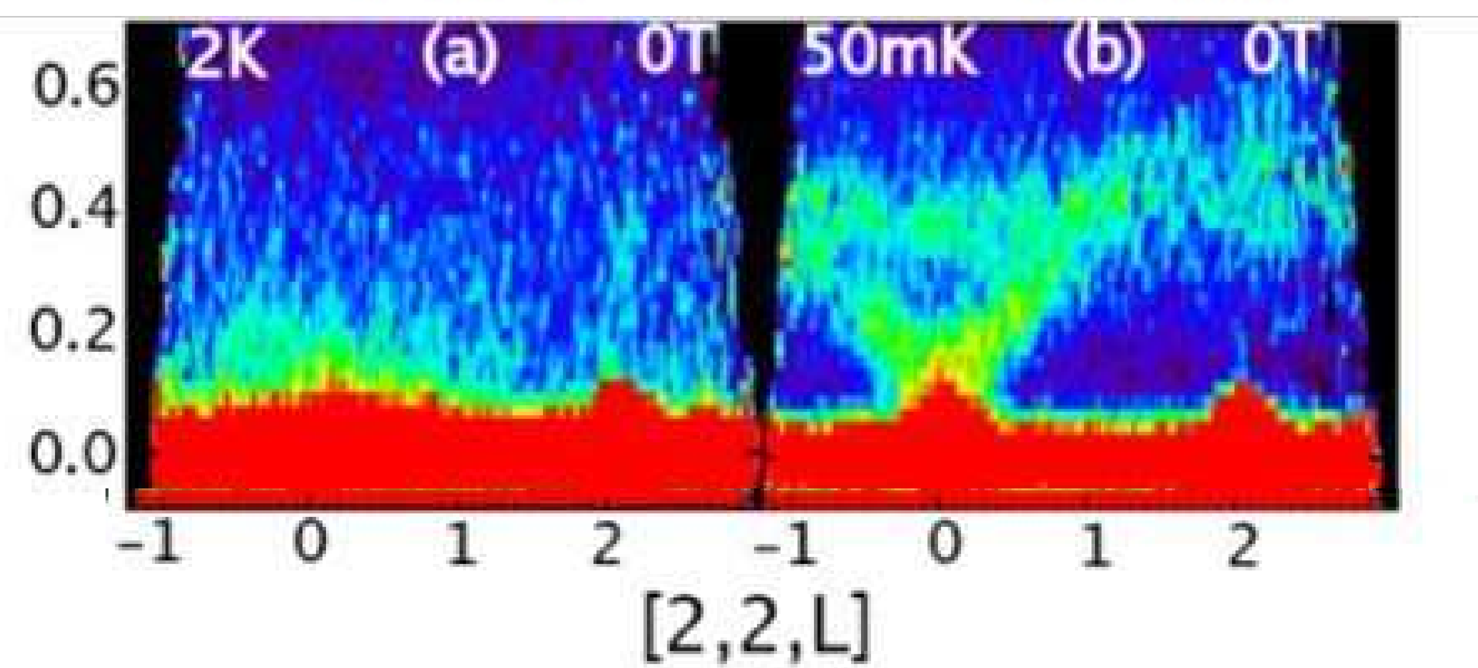
K. A. Ross et al, Phys. Rev. Lett. 103, 227202 (2009)



$Yb_2Ti_2O_7$ shows no conventional spin waves in H=0
Spin waves appear for H // [1-10] > 0.5 T

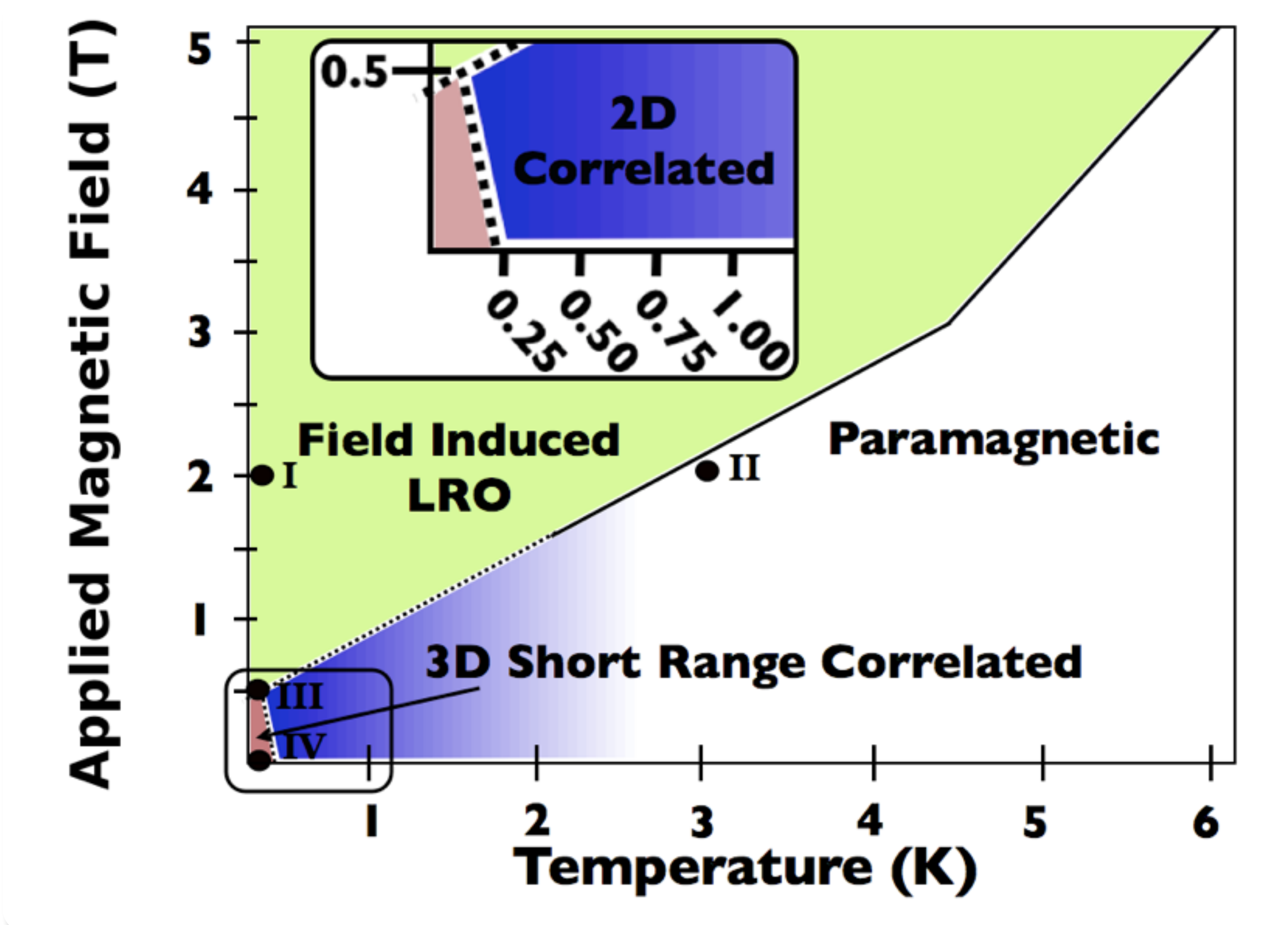






$Yb_2Ti_2O_7$: T_C = 0.26 K
Q_{ord} = (1 1 1)

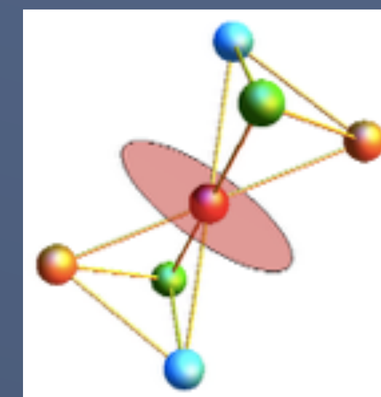
$Er_2Ti_2O_7$: T_N=1.2 K
Q_{ord}=(220)



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)

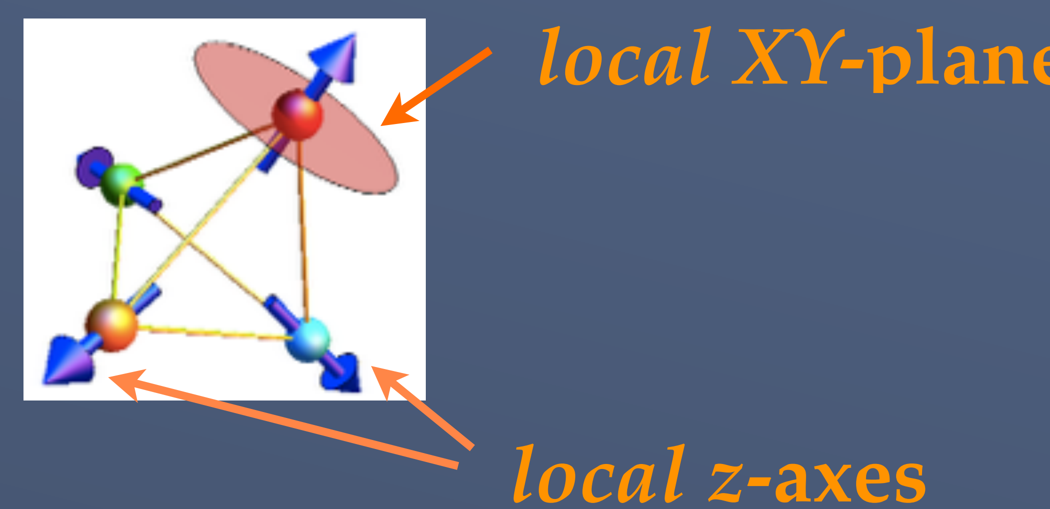
Anisotropic Exchange

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



4 symmetry-allowed terms for exchange tensor

S. Curnoe. Phys. Rev. B 78, 094418 (2008).
see also S. Onoda and Y. Tanaka, Phys. Rev. B 83, 094411 (2011).

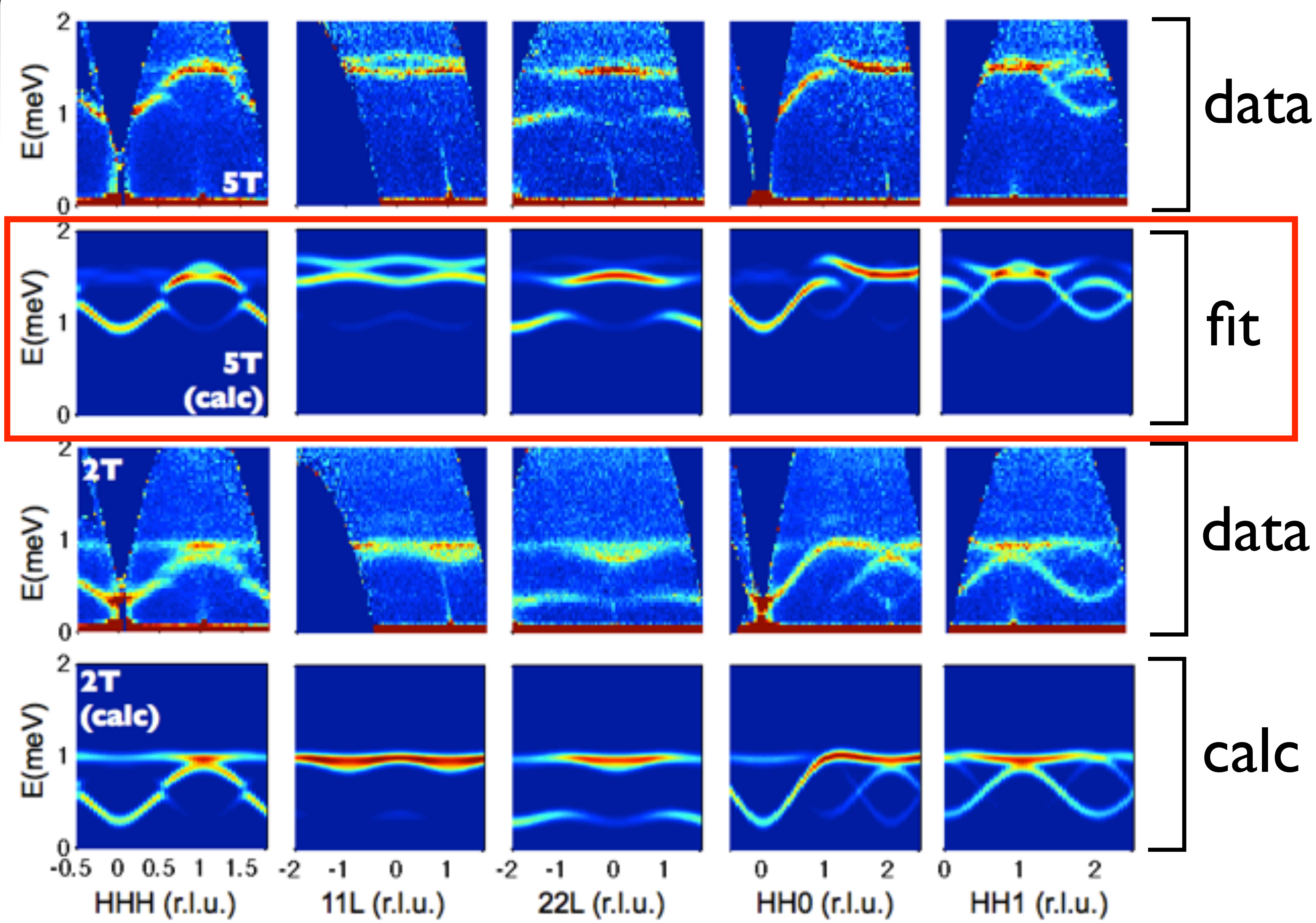


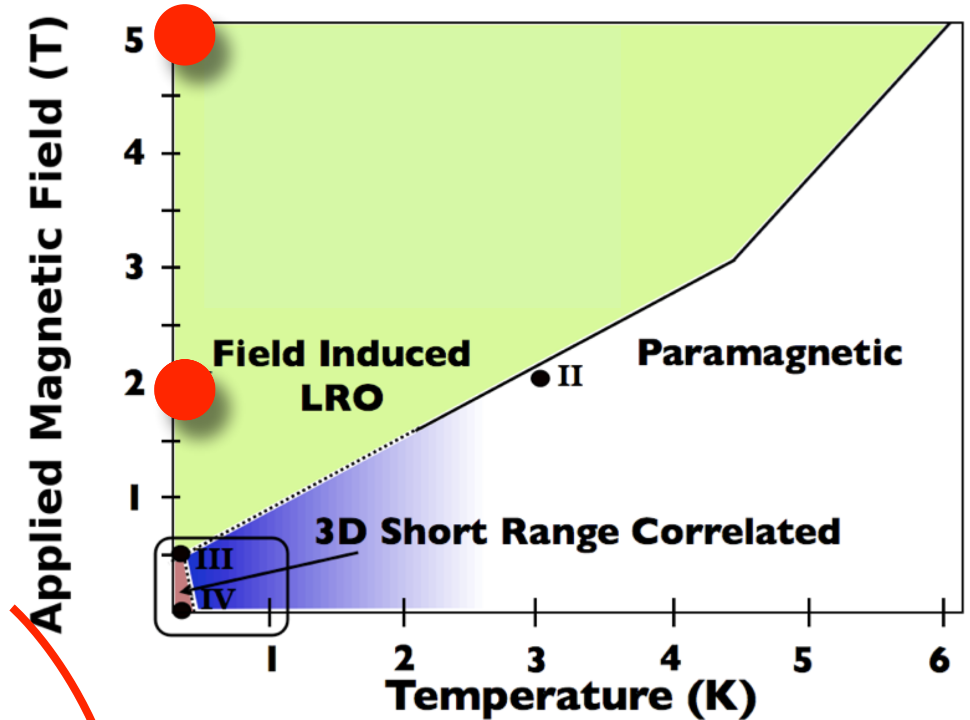
$$H = \sum_{\langle ij\rangle}\Big\{J_{zz}\mathsf{S}_i^z\mathsf{S}_j^z - J_{\pm}(\mathsf{S}_i^+\mathsf{S}_j^- + \mathsf{S}_i^-\mathsf{S}_j^+) + J_{++}\left[\gamma_{ij}\mathsf{S}_i^+\mathsf{S}_j^+ + \gamma_{ij}^*\mathsf{S}_i^-\mathsf{S}_j^-\right] + J_{z\pm}\left[\mathsf{S}_i^z(\zeta_{ij}\mathsf{S}_j^+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

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

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

Spin waves in $Yb_2Ti_2O_7$'s field polarized state





H along [1-10]

See also:
J. Robert et al.,
Phys. Rev. B 92, 064425 (2015)

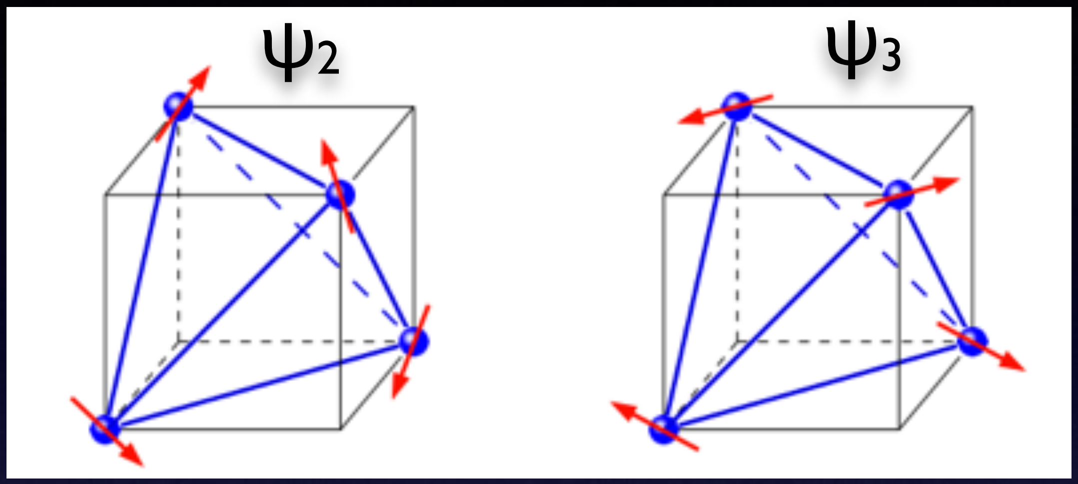
J.D.Thompson et al, arXiv:1703.04506R

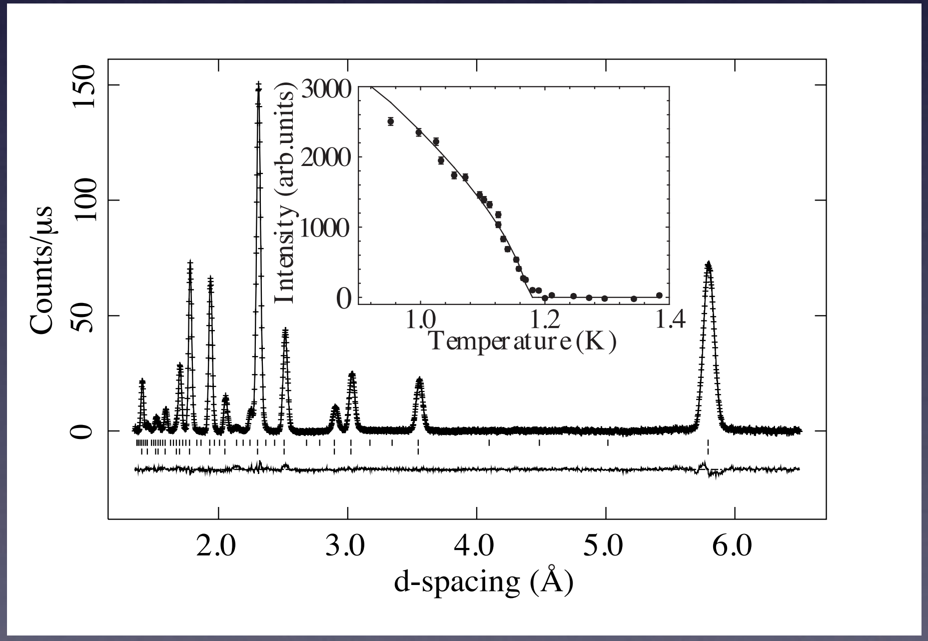
"Quantum Spin Ice"

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

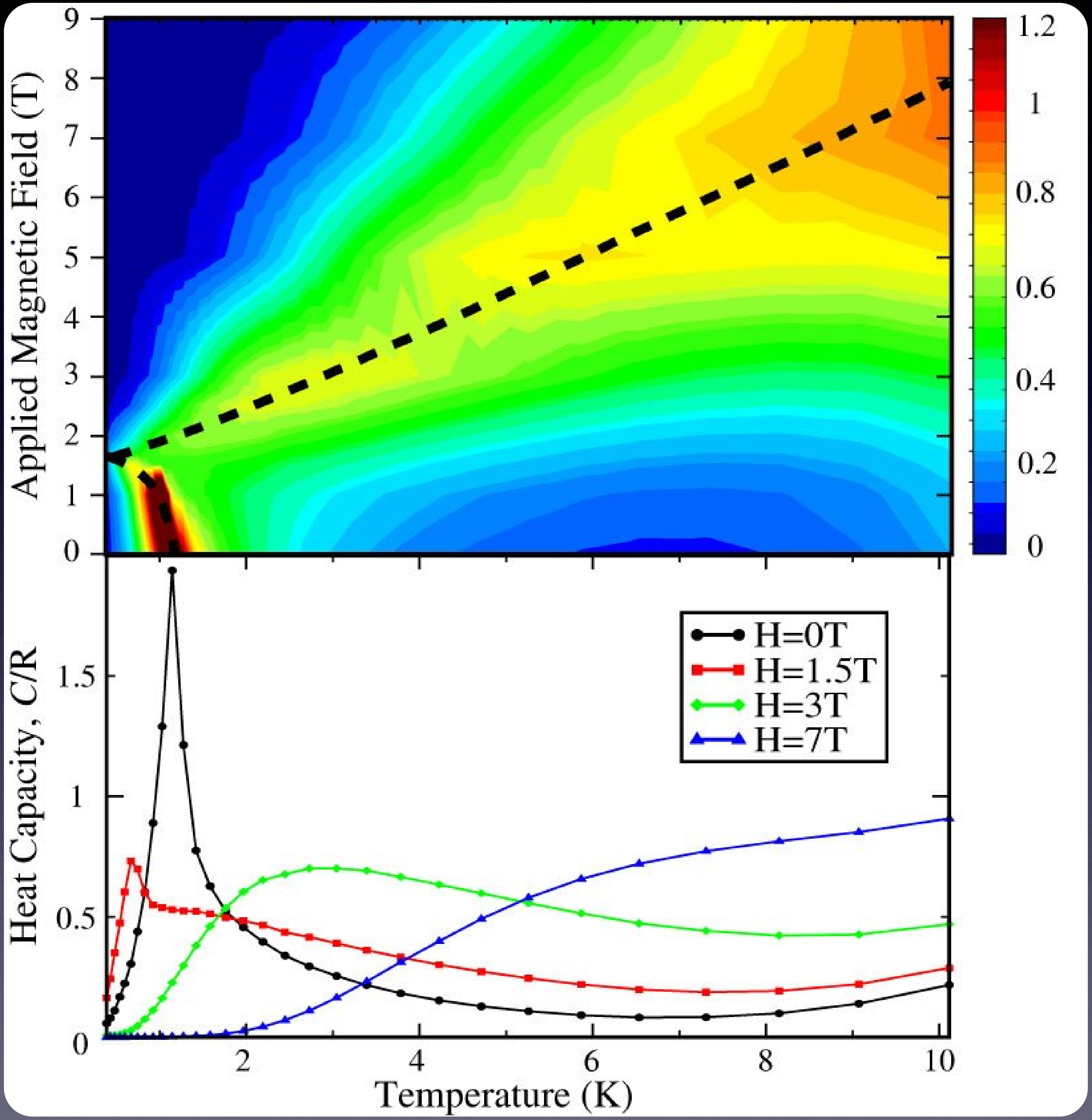
K.A. Ross, L. Savary, B.D. Gaulin, and L. Balents, PRX, 1, 021002 (2011)

S_{eff} = 1/2 XY AF Pyrochlore : $Er_2Ti_2O_7$ $\Theta_{CW} \sim$ -22 K



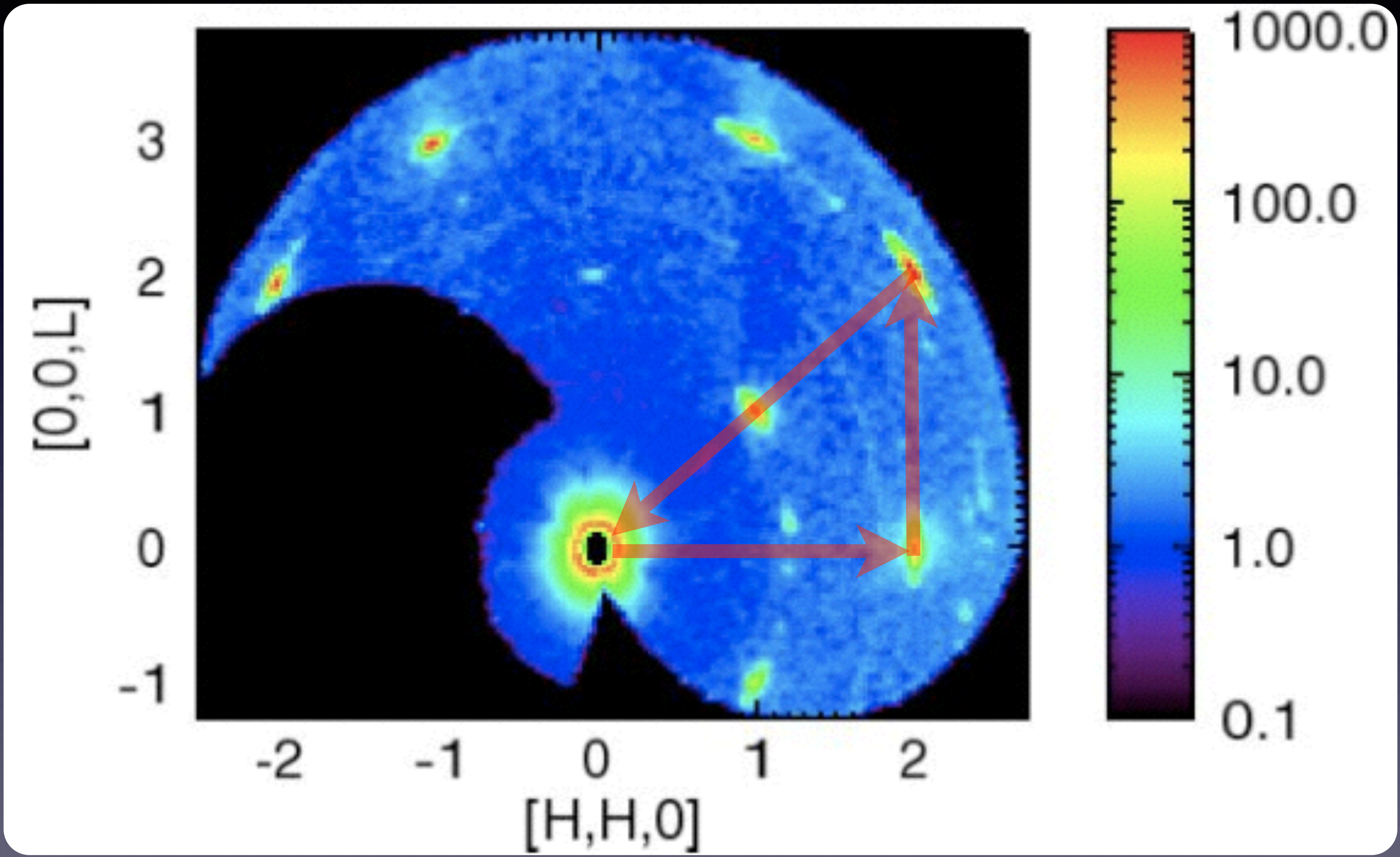


Champion et al, PRB 68, 020401 (2003)



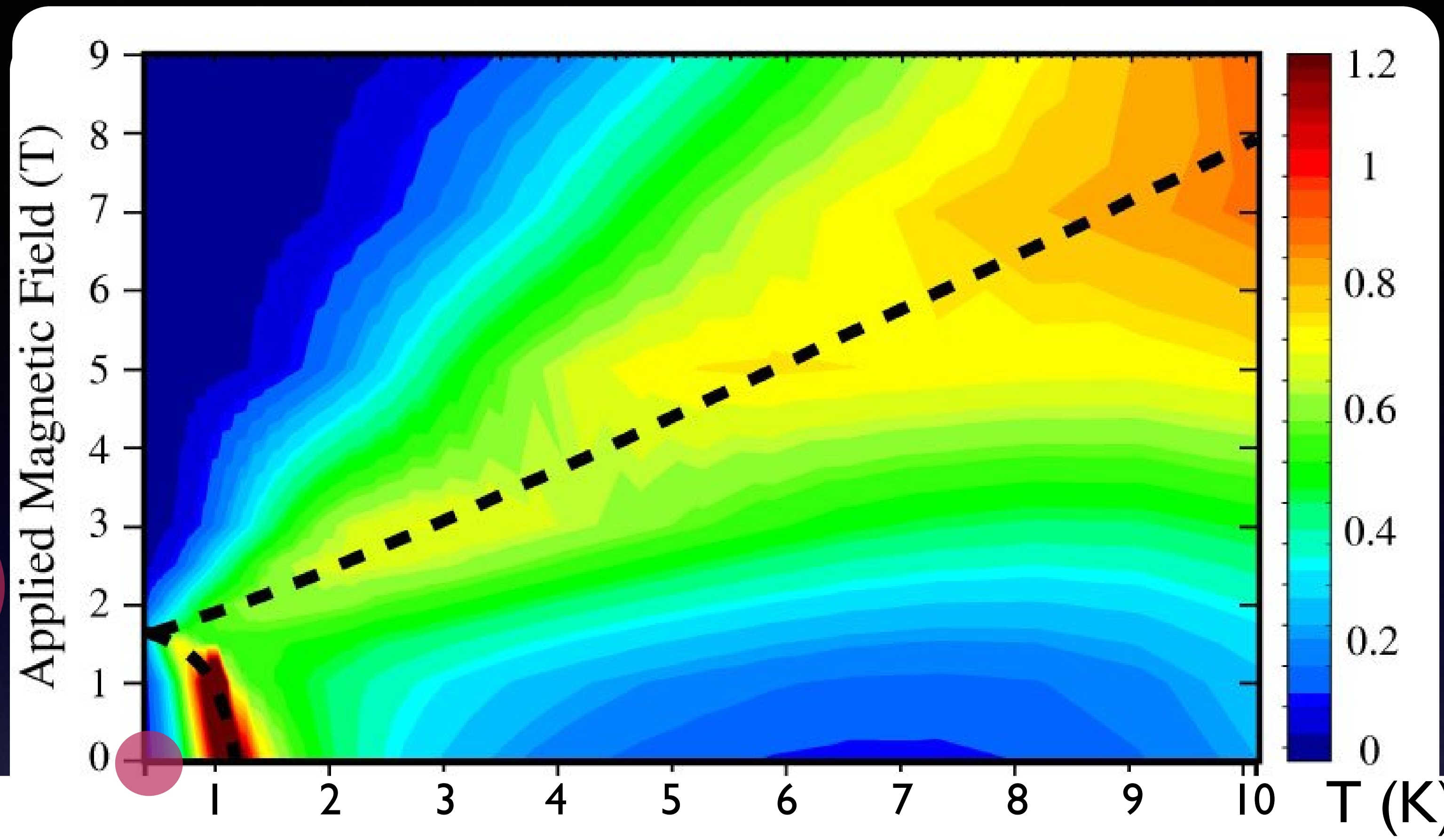
A 9 year mystery (2003-2012)
as to what selects ψ_2 ground state in $Er_2Ti_2O_7$

$Er_2Ti_2O_7$ @ 50 mK

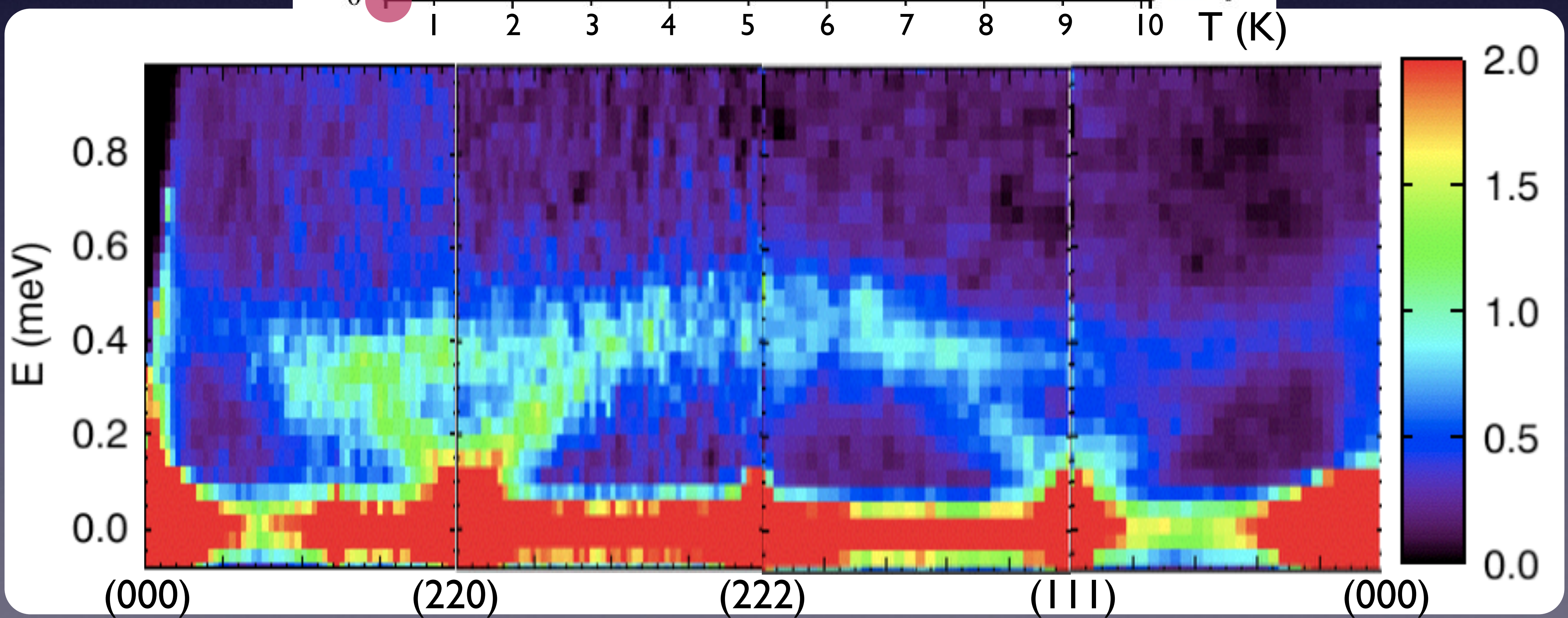


J.P.C. Ruff, J.P. Clancy, A. Bourque, M.A. White, M. Ramazanoglu, J.S. Gardner, Y. Qiu, J.R.D. Copley, M.B. Johnson, H.A. Dabkowska and B.D. Gaulin, Phys. Rev. Lett., 101, 147205 (2008)

50 mK, 0 T

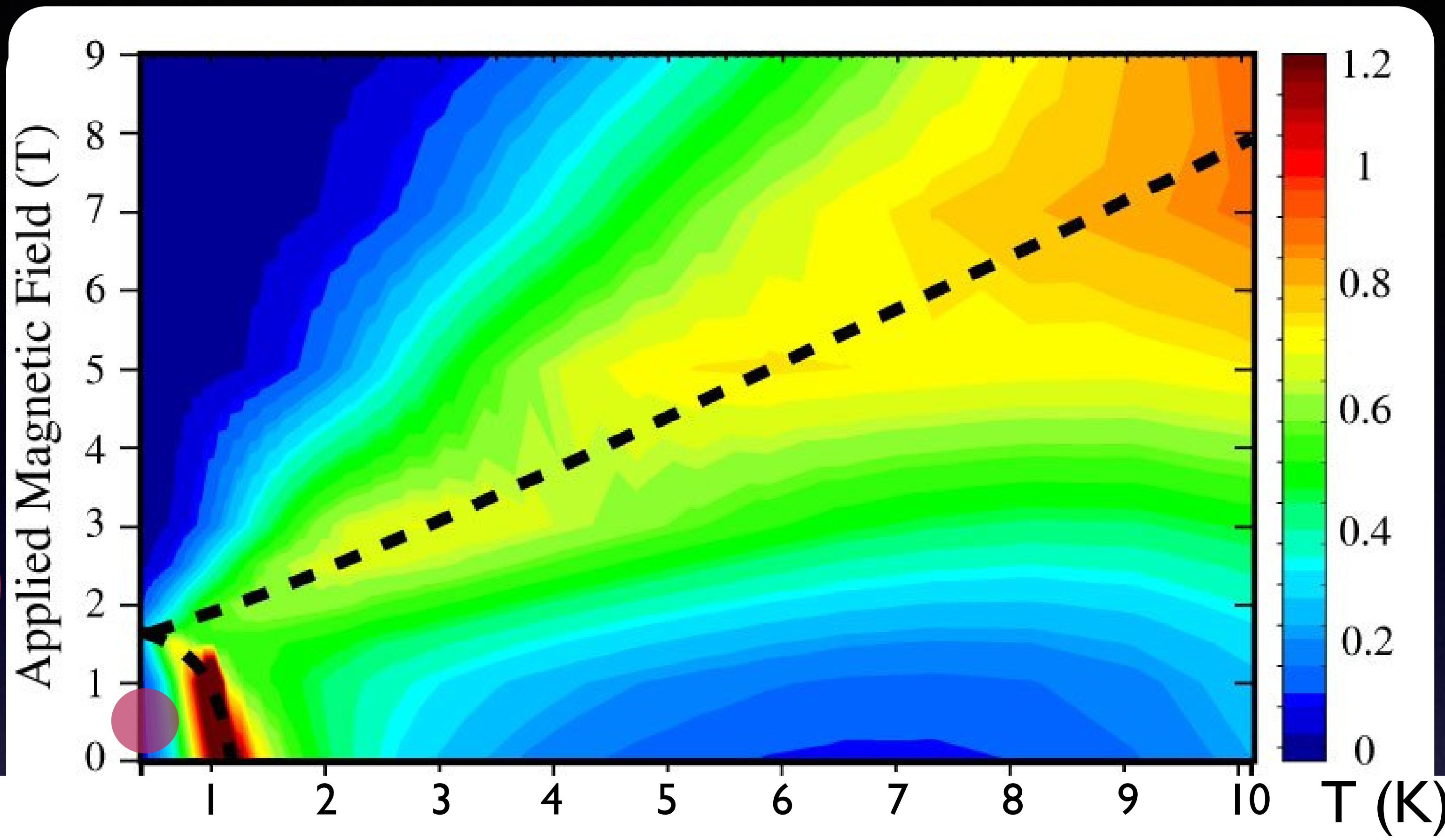


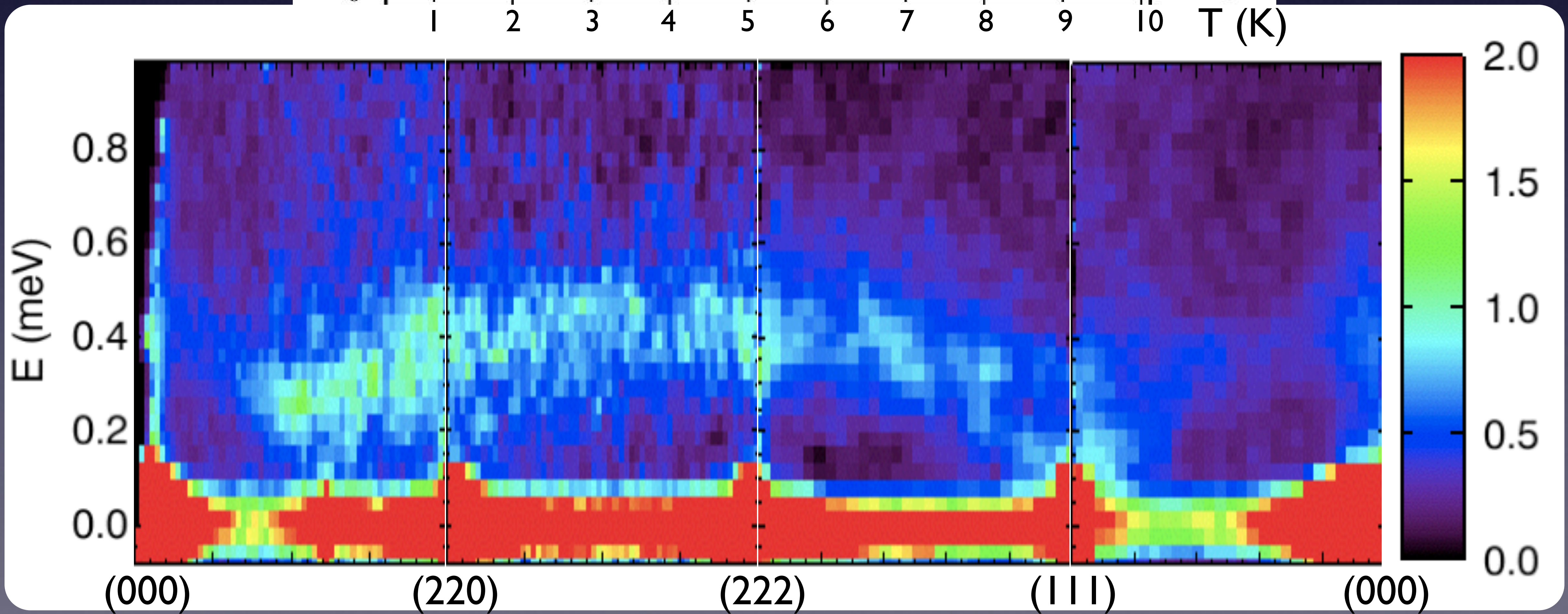
Applied Magnetic Field (T)
9
8
7
6
5
4
3
2
1
0
1.2
1
0.8
0.6
0.4
0.2
0
1
2
3
4
5
6
7
8
9
10
T (K)



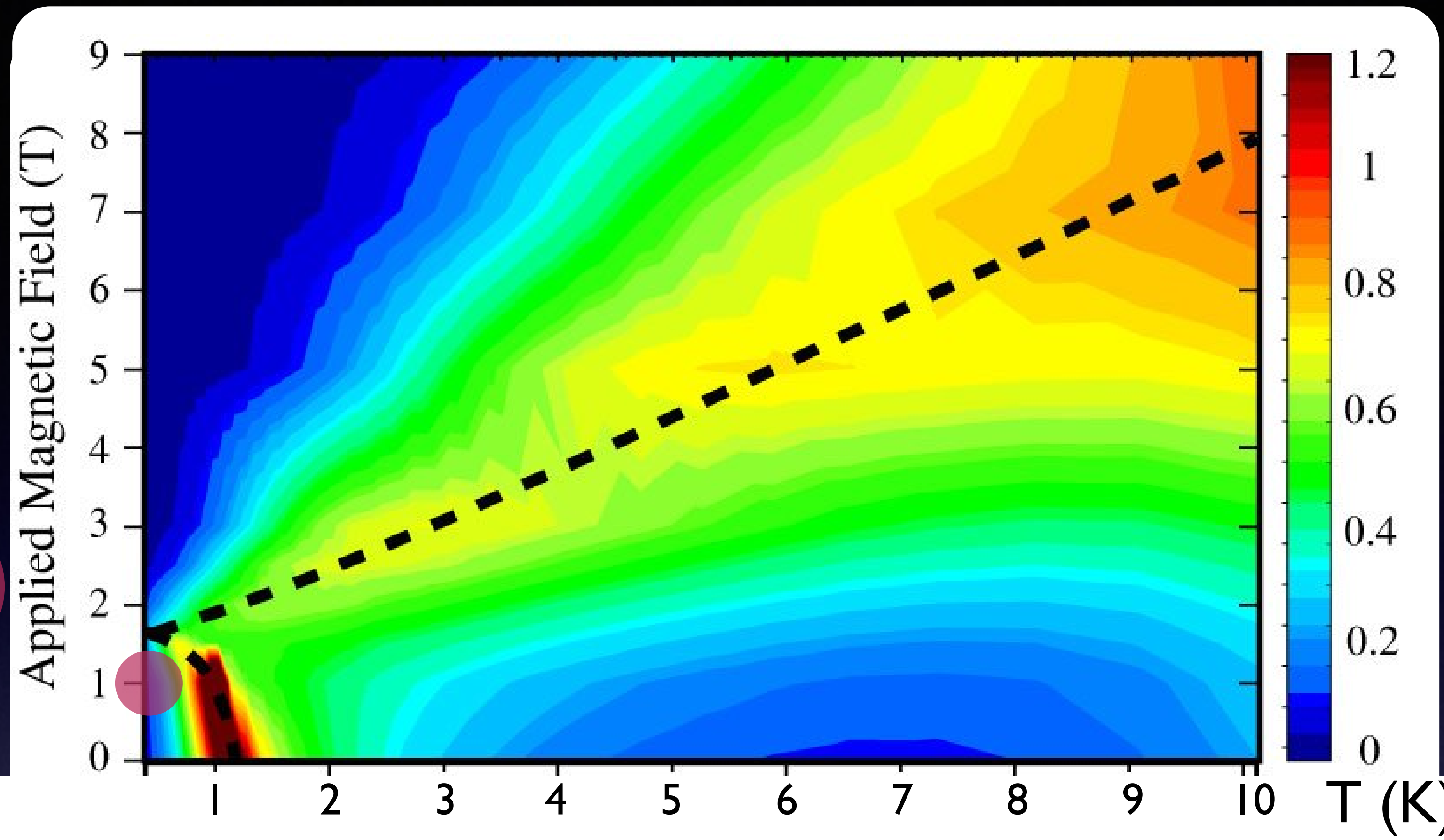
E (meV)
0.8
0.6
0.4
0.2
0.0
2.0
1.5
1.0
0.5
0.0
(000)
(220)
(222)
(111)
(000)

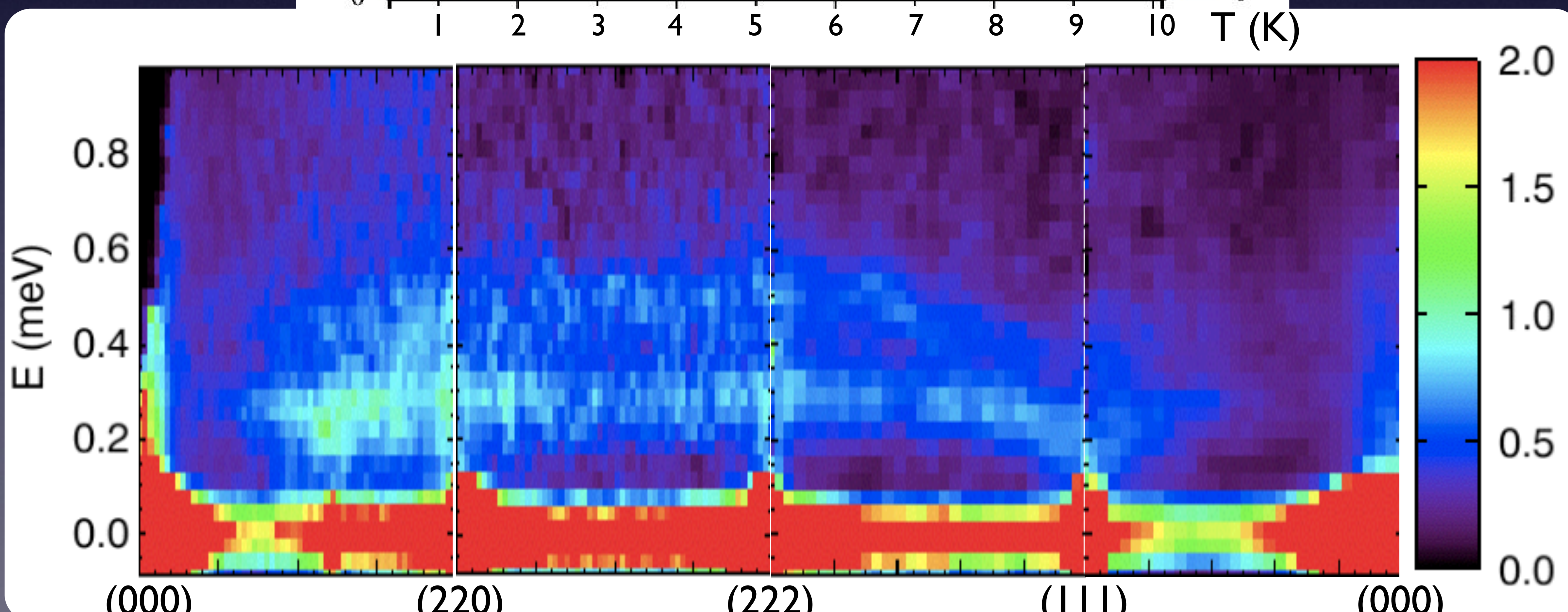
50 mK, 0.5 T


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

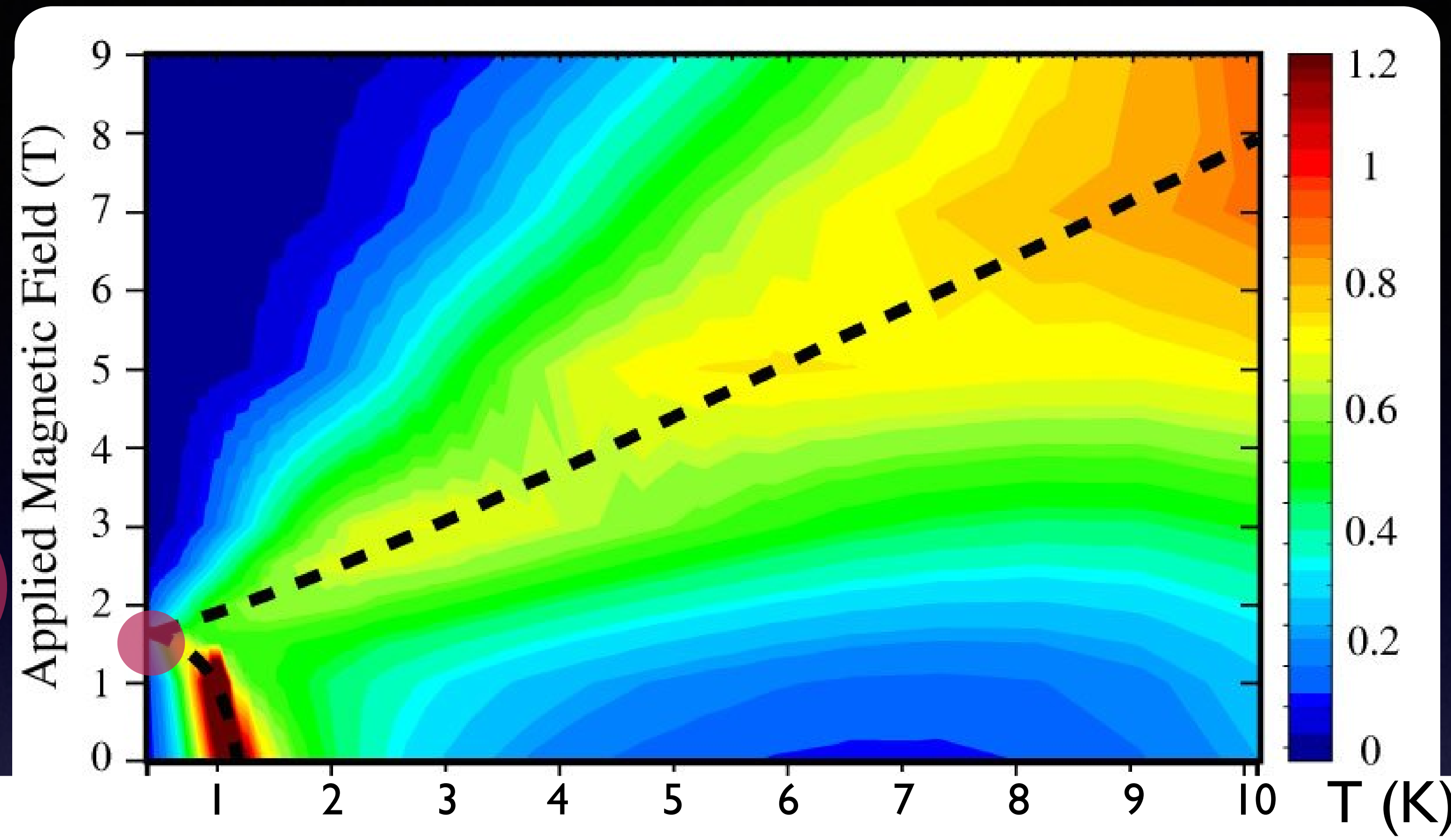

E (meV)
0.0
0.2
0.4
0.6
0.8
(000)
(220)
(222)
(111)
(000)
0.0
0.5
1.0
1.5
2.0

50 mK, 1.0 T

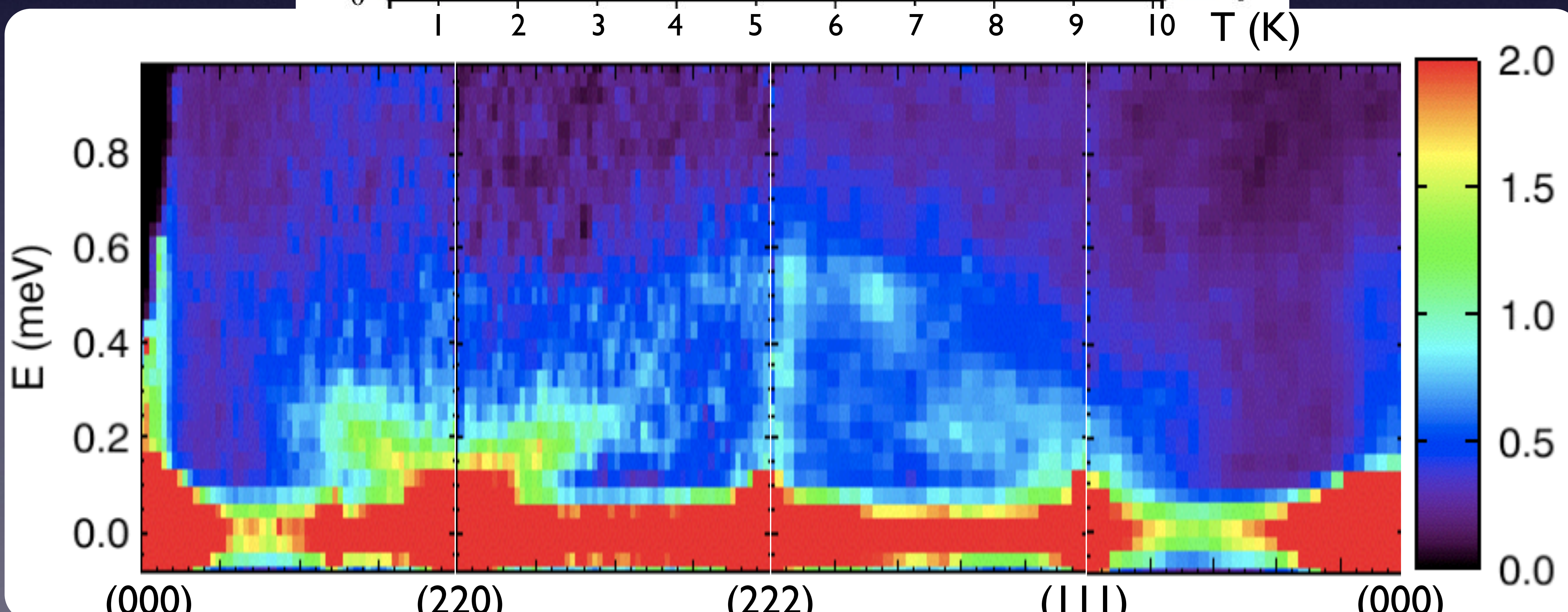




50 mK, 1.5 T

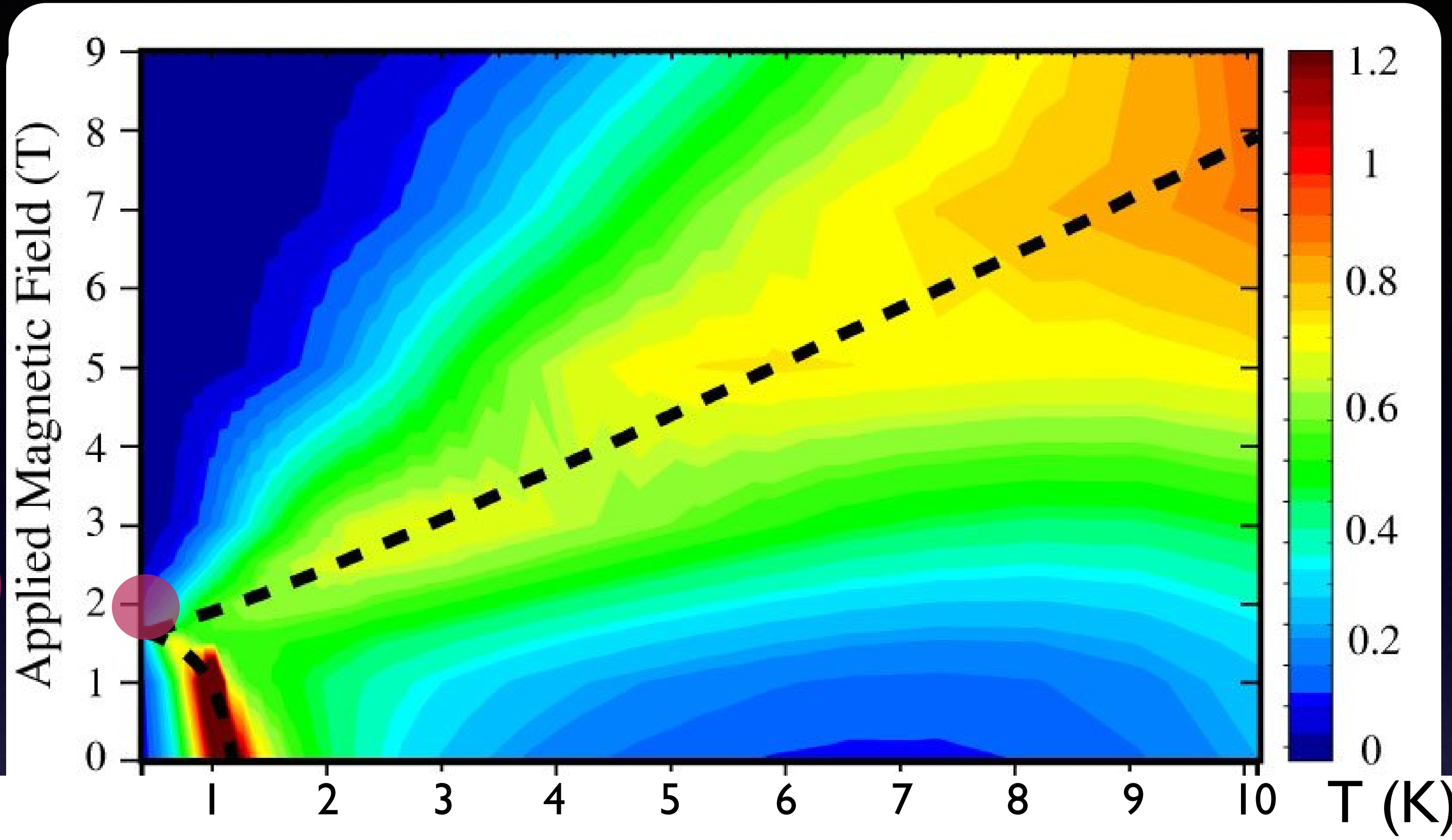


Applied Magnetic Field (T)
0
1
2
3
4
5
6
7
8
9
T (K)
1
2
3
4
5
6
7
8
9
10
0
0.2
0.4
0.6
0.8
1
1.2

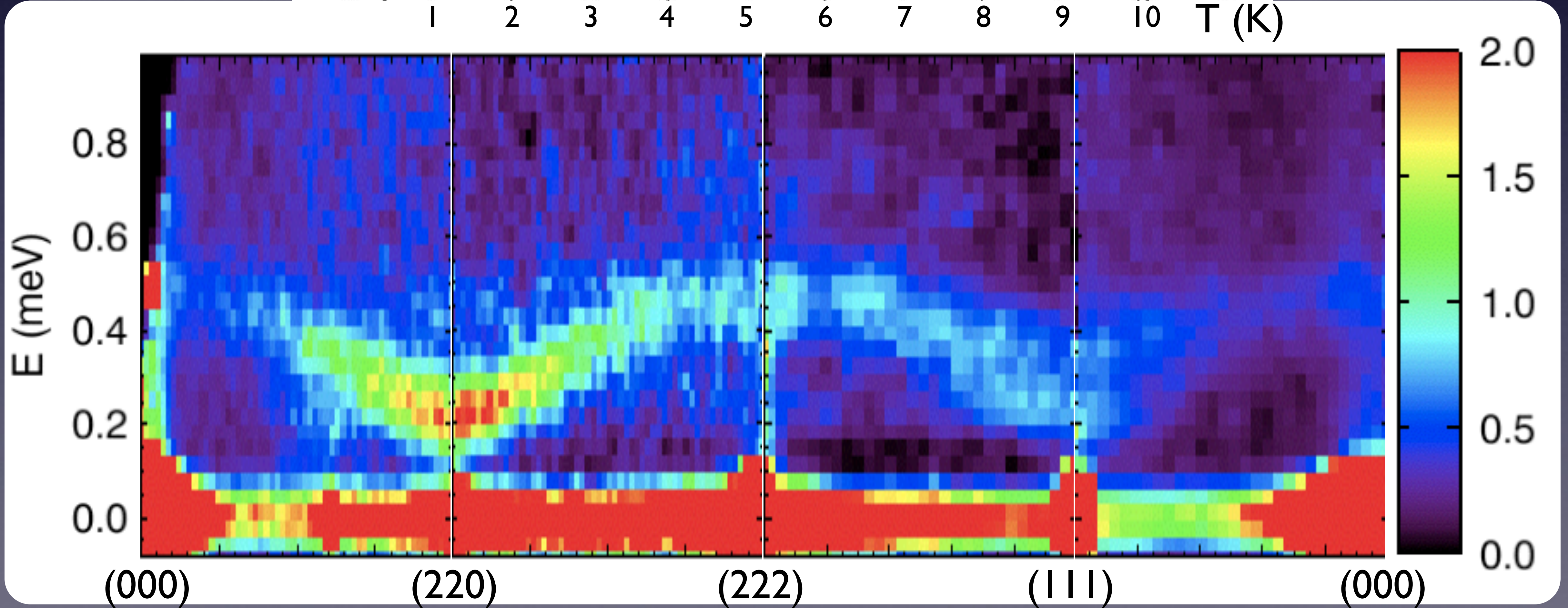


E (meV)
0.0
0.2
0.4
0.6
0.8
(000)
(220)
(222)
(111)
(000)
0.0
0.5
1.0
1.5
2.0

50 mK, 2.0 T

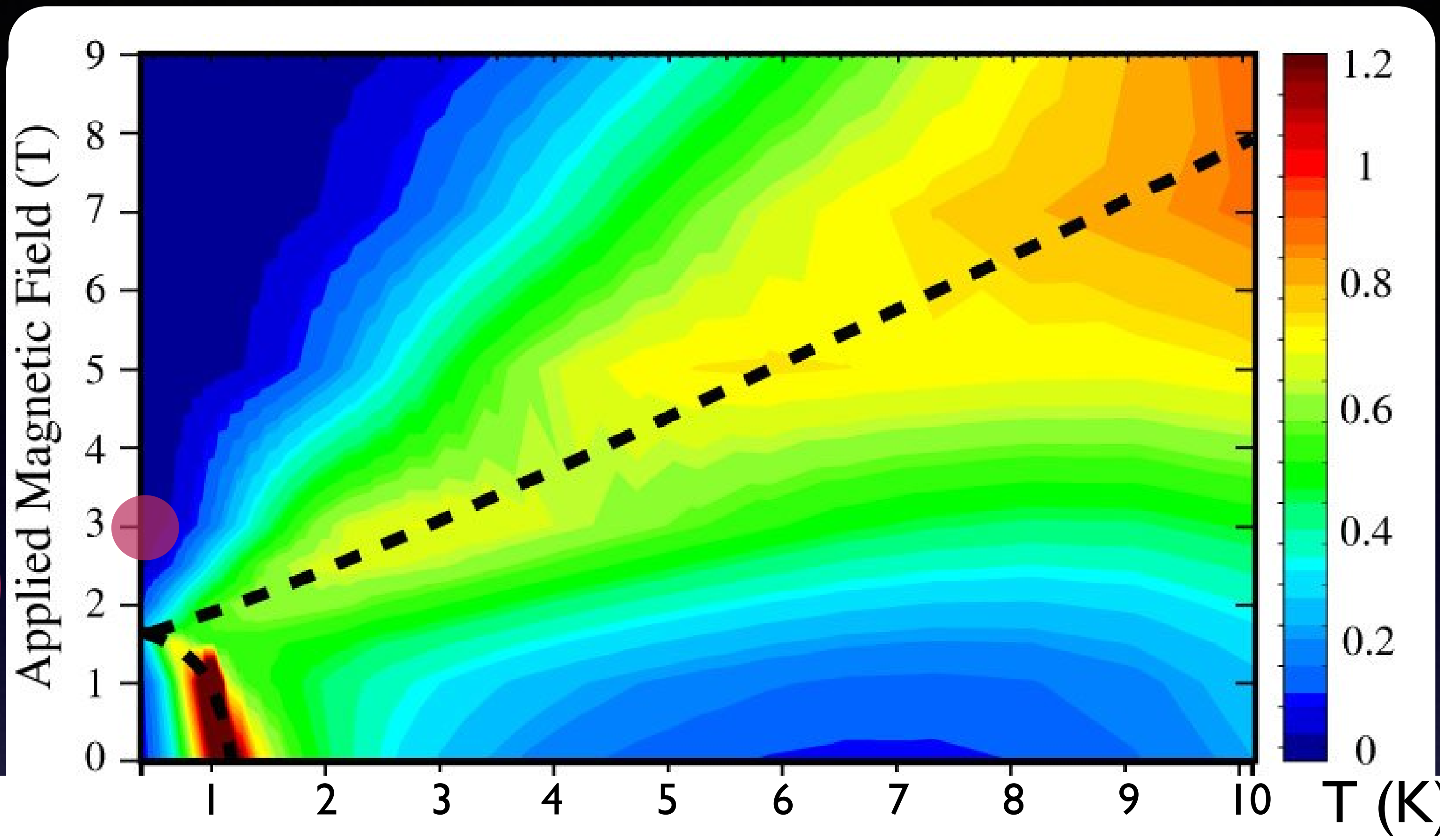


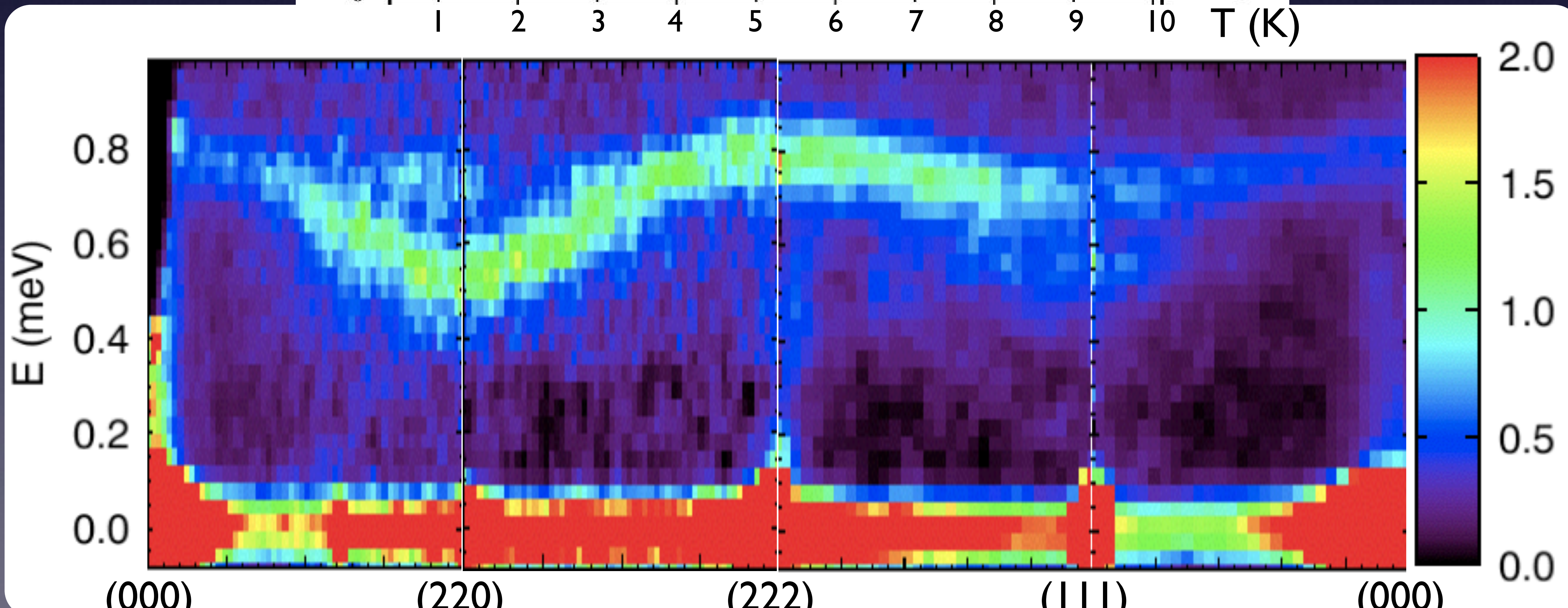
Applied Magnetic Field (T)
9
8
7
6
5
4
3
2
1
0
1
2
3
4
5
6
7
8
9
10
T (K)
1.2
1
0.8
0.6
0.4
0.2
0



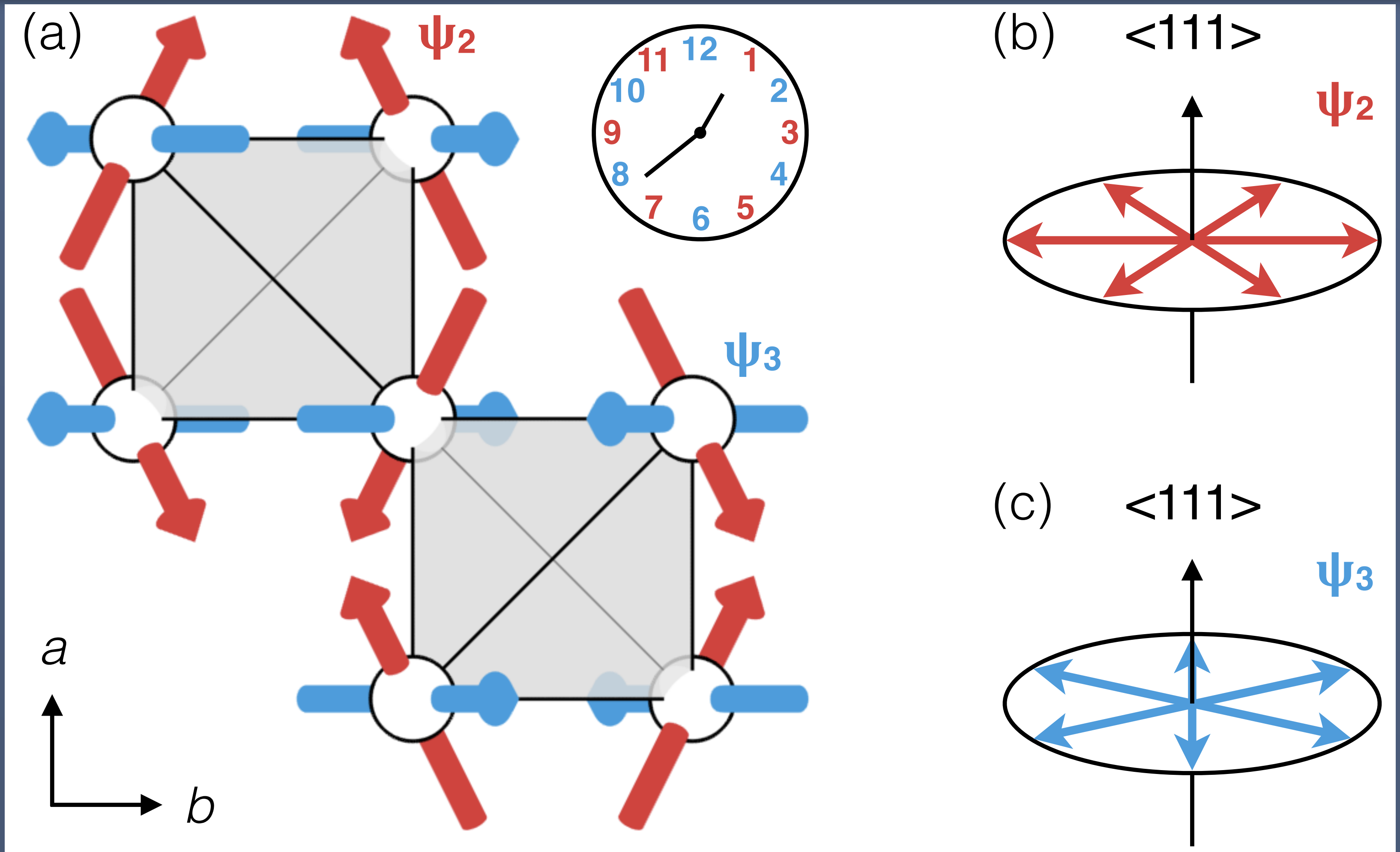
E (meV)
0.8
0.6
0.4
0.2
0.0
(000)
(220)
(222)
(111)
(000)
2.0
1.5
1.0
0.5
0.0

50 mK, 3.0 T

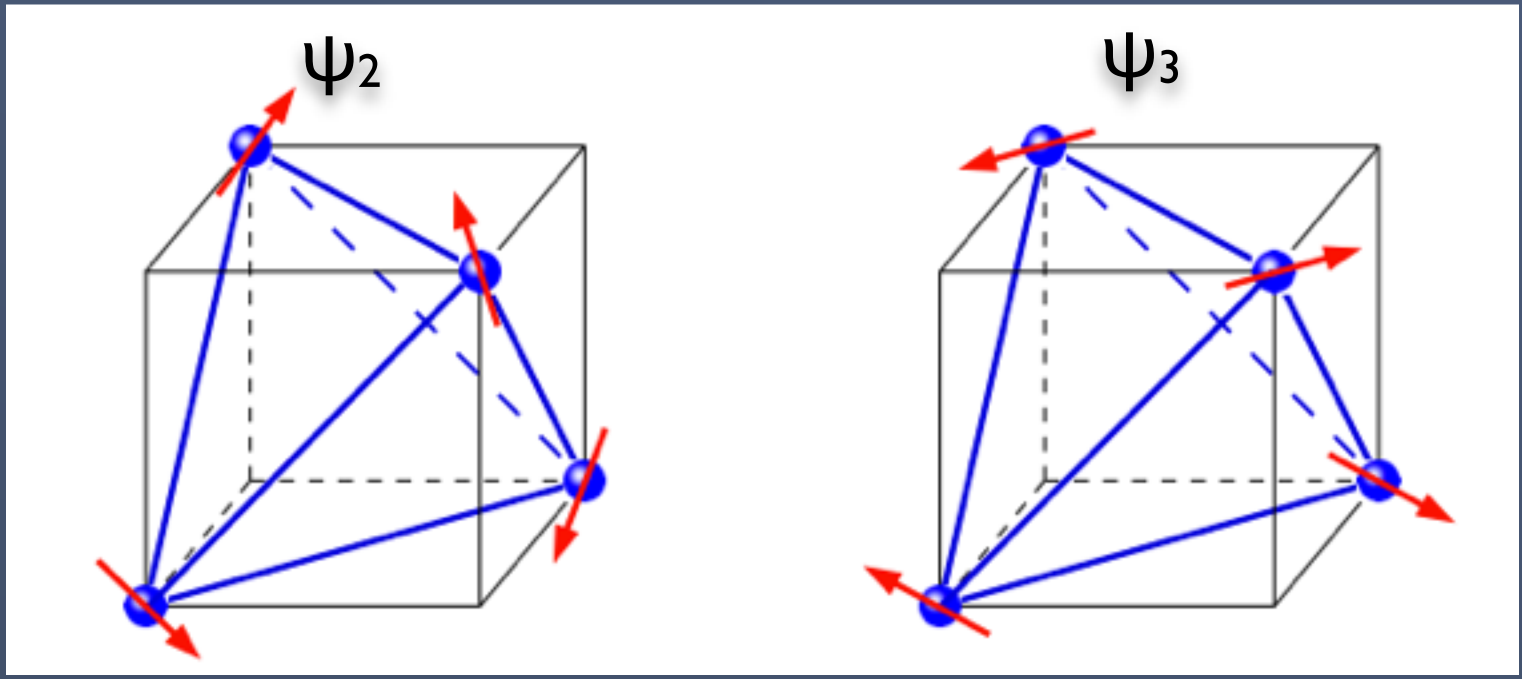

Applied Magnetic Field (T)
0
1
2
3
4
5
6
7
8
9
0
0.2
0.4
0.6
0.8
1
1.2
1
2
3
4
5
6
7
8
9
10
T (K)

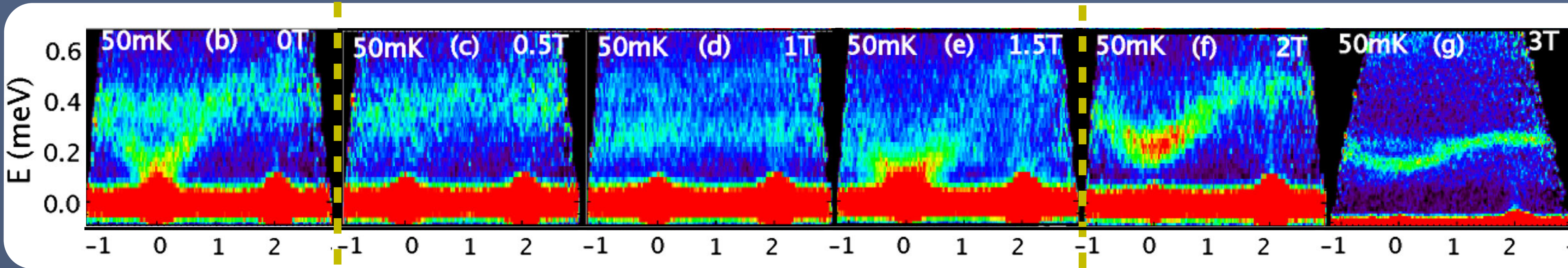

E (meV)
0.0
0.2
0.4
0.6
0.8
2.0
1.5
1.0
0.5
0.0
(000)
(220)
(222)
(111)
(000)

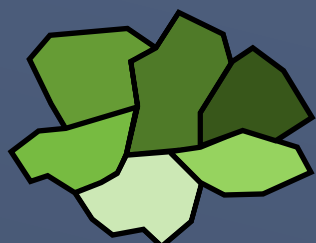
AF interactions and local XY anisotropy

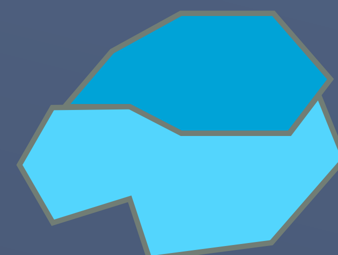


- **continuous degeneracy in mean field**
- **6 ψ_2 and 6 ψ_3 domains**
- **ψ_2 selected by ObD**







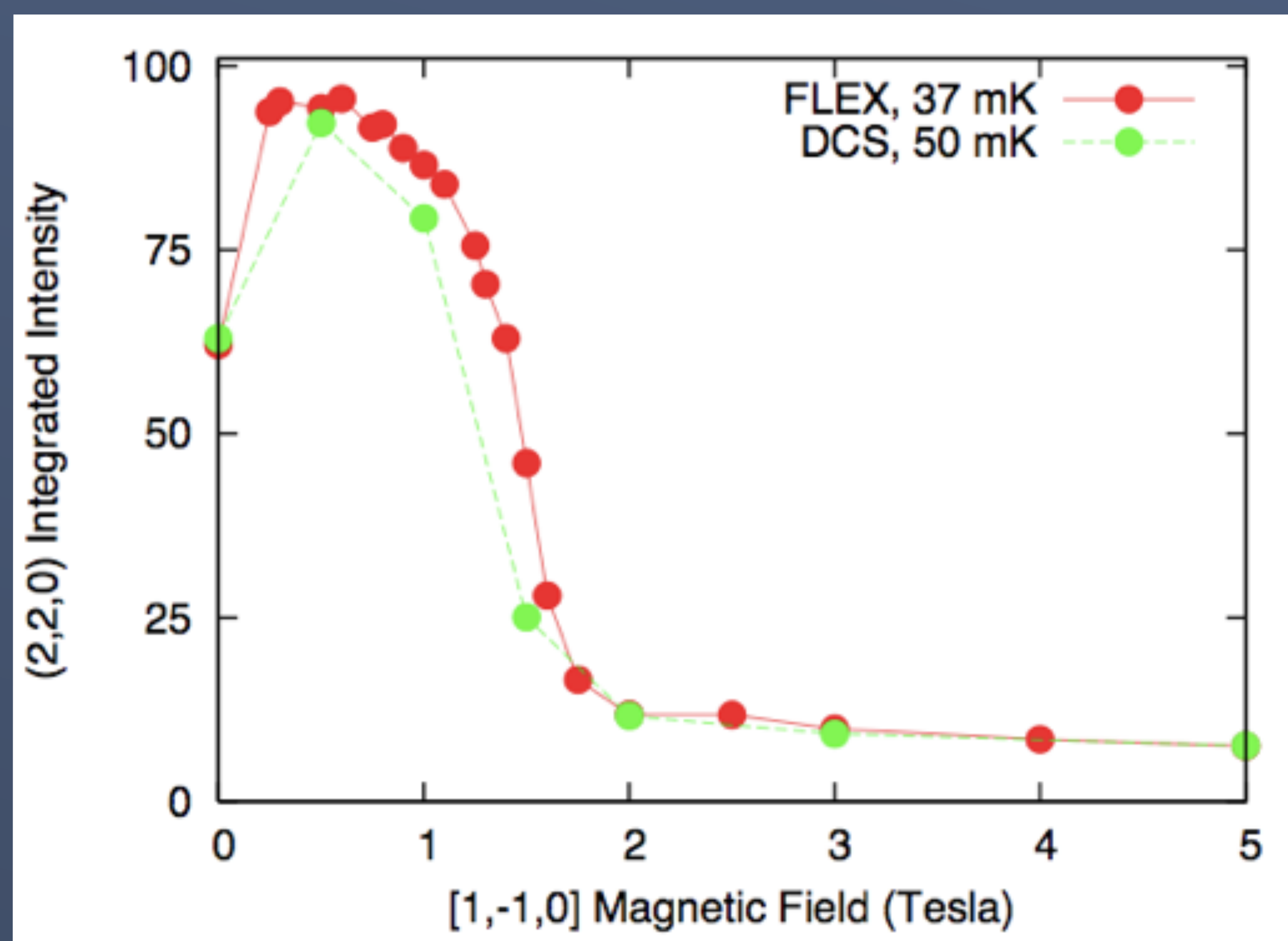


$H_c = 1.74$ T

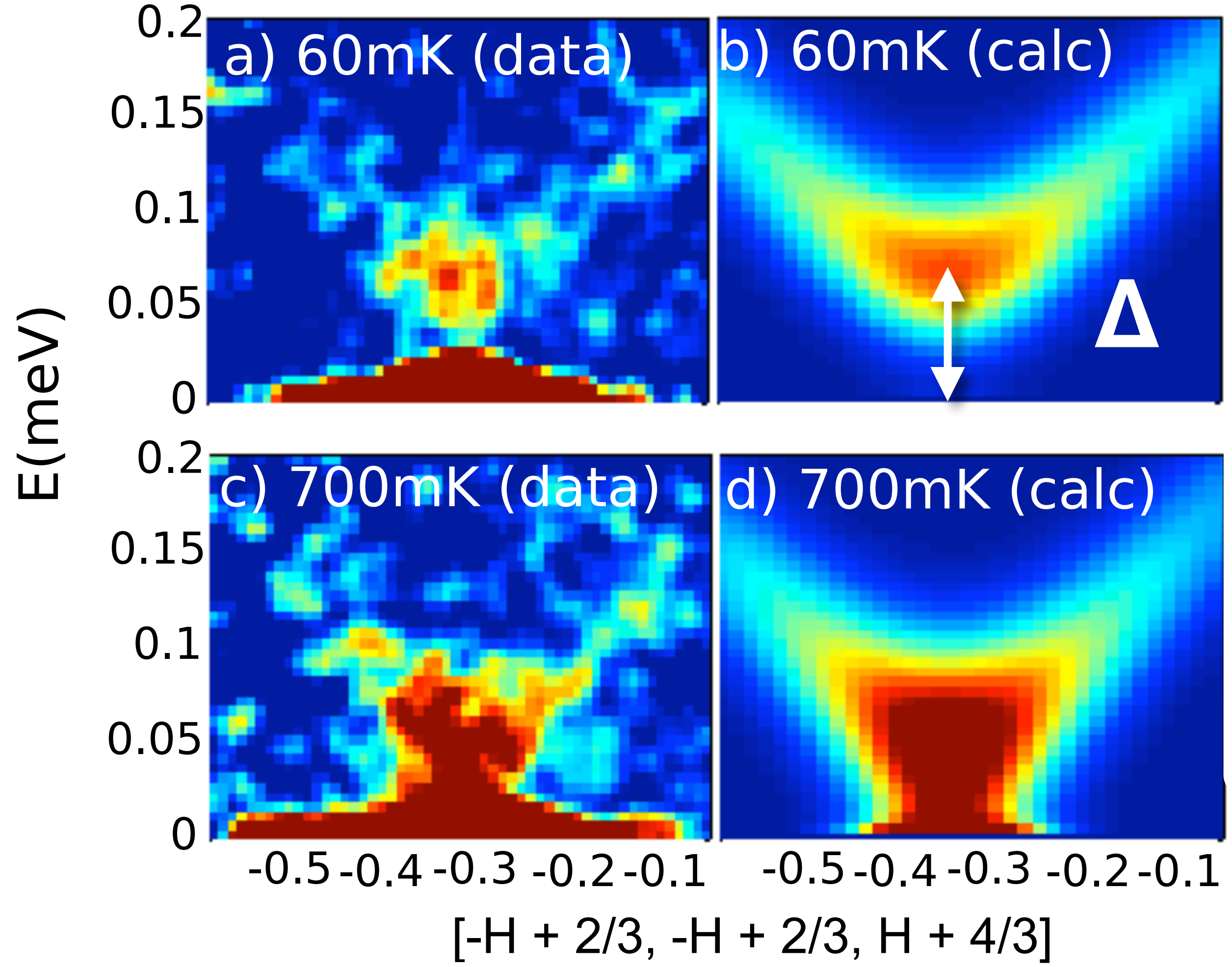
6 ψ2 domains

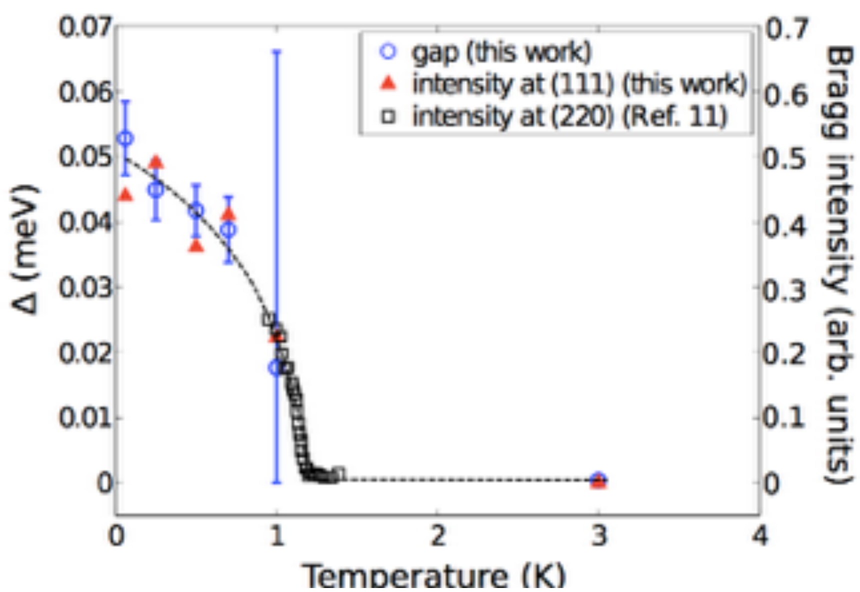
2 ψ2 domains

1 polarized domain

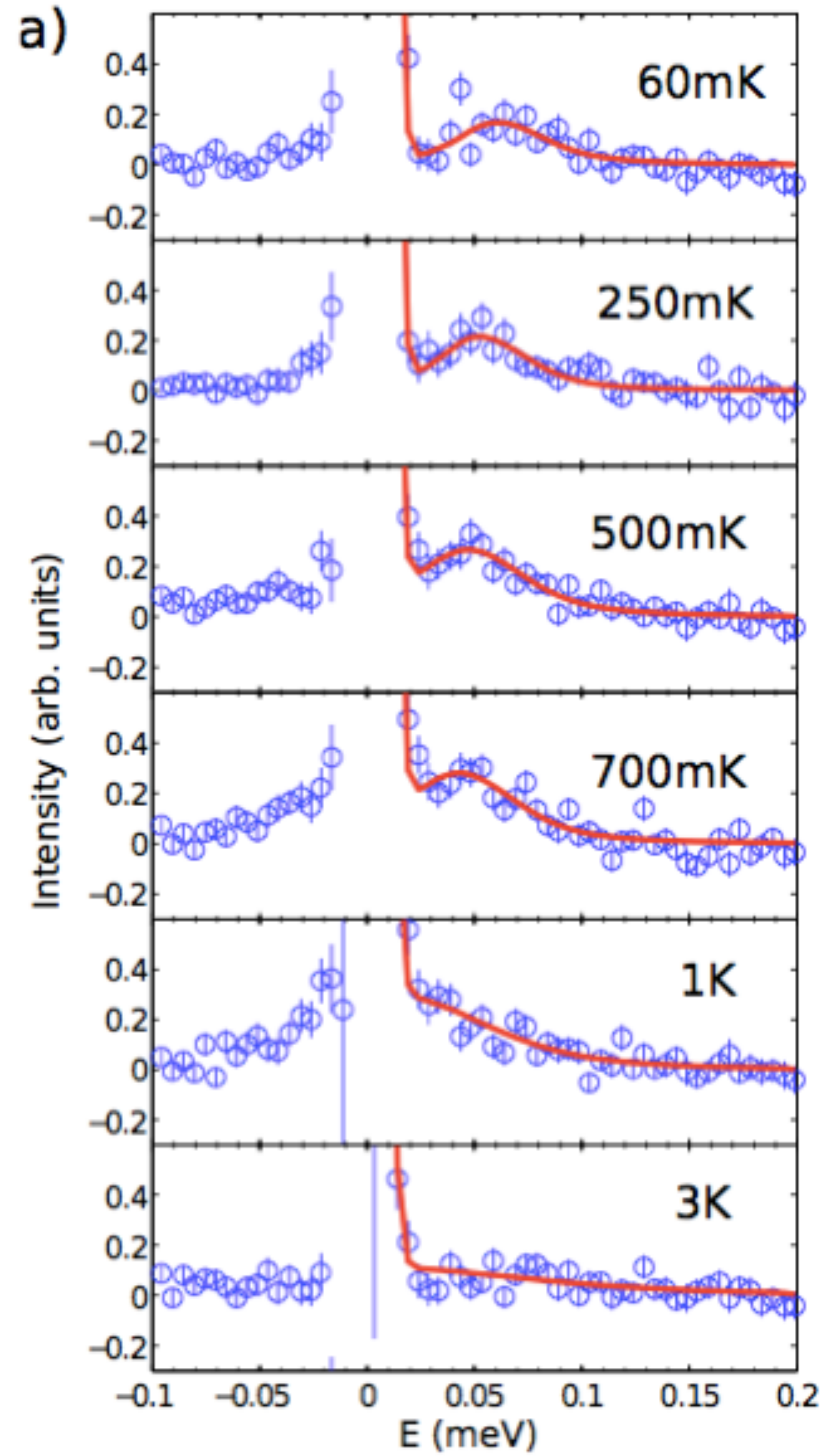


A very small gap exists - It stabilizes ψ_2 vs ψ_3 !





$\Delta \sim (OP)^2$



K.A. Ross, Y. Qiu, J.R.D. Copley, H.A. Dabkowska, and B.D. Gaulin, Phys. Rev. Lett., 112, 057201, 2014

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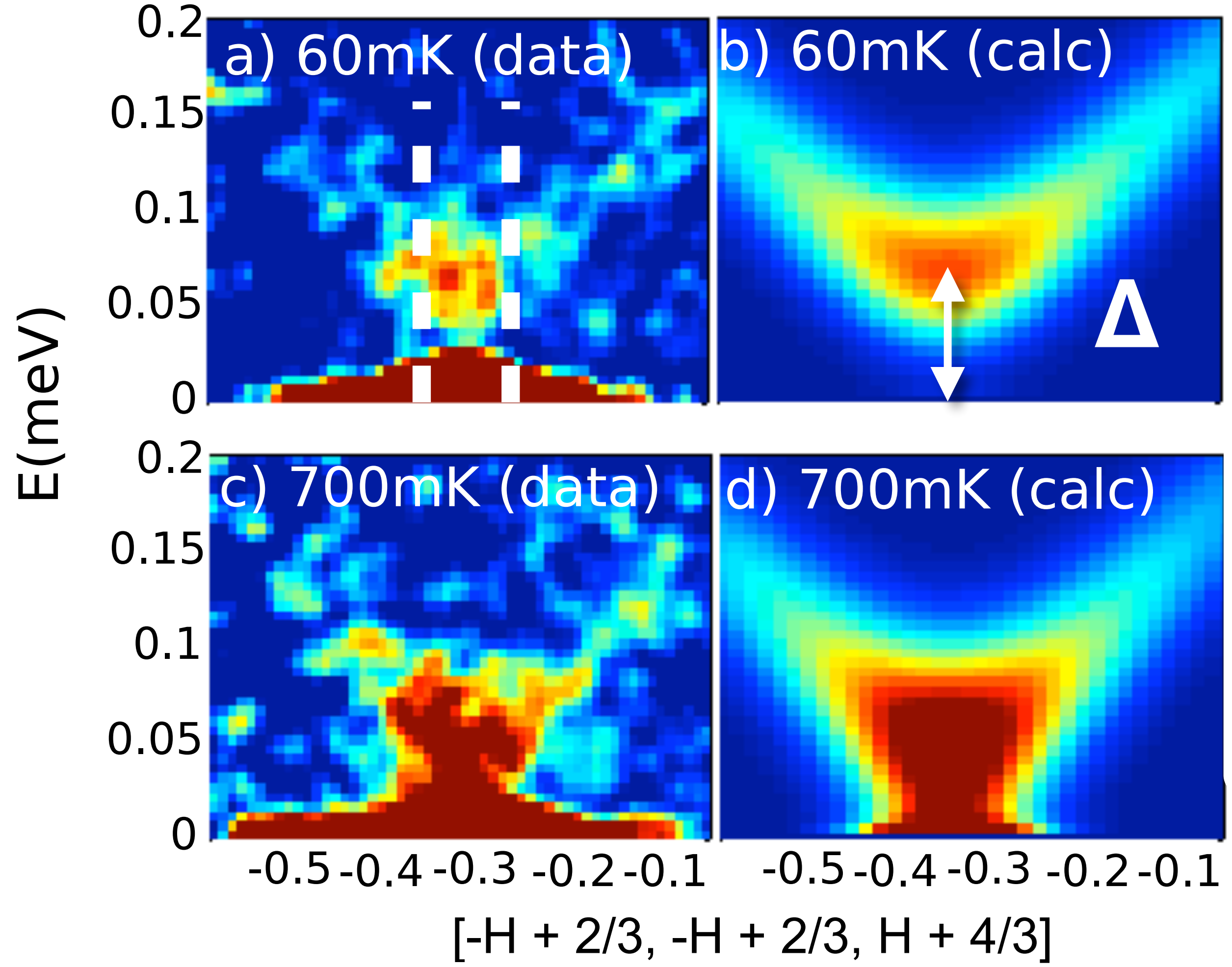
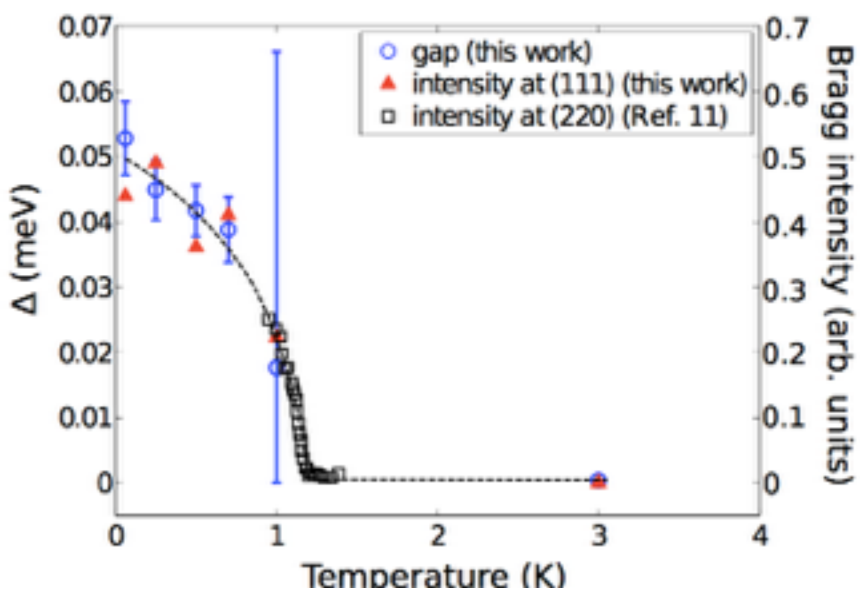
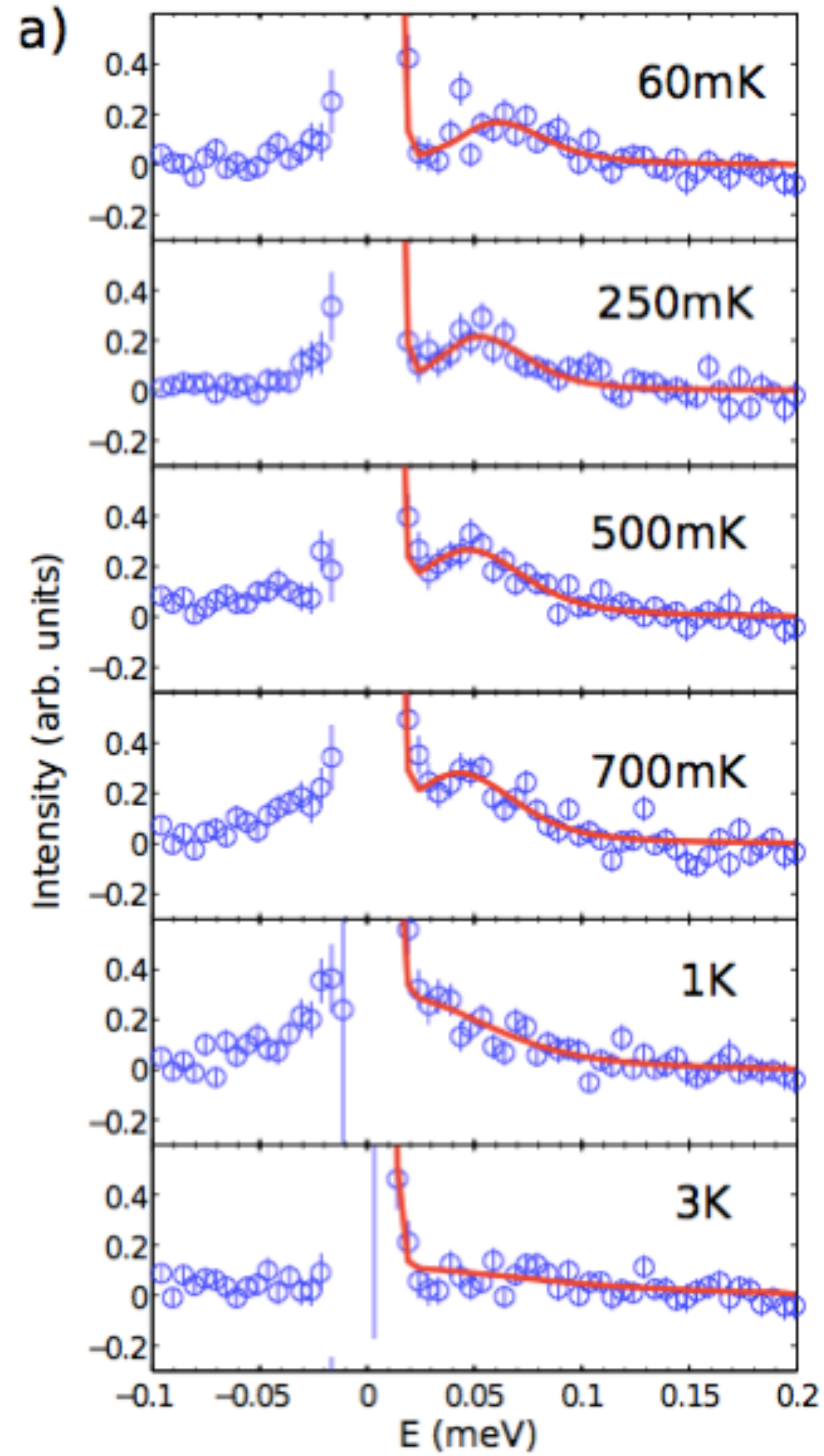




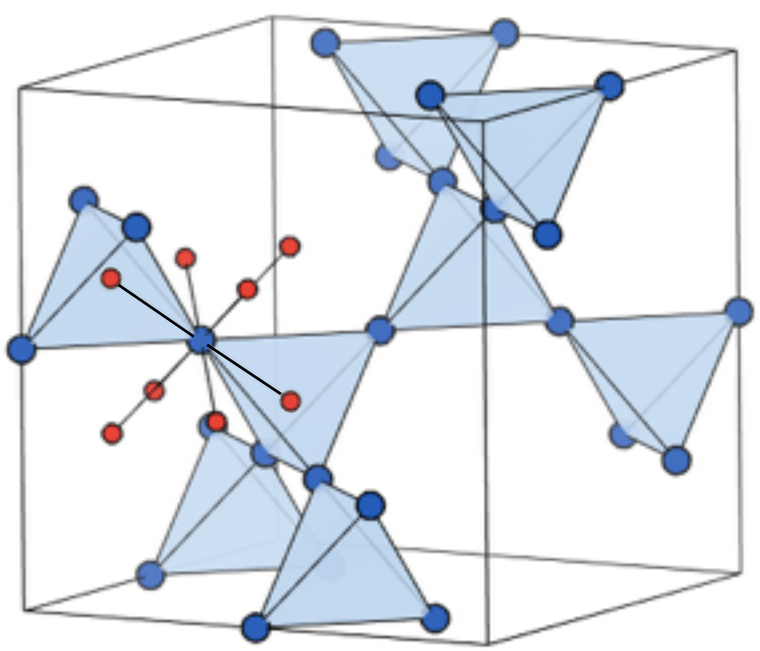
$\Delta \sim (\mathrm{OP})^2$



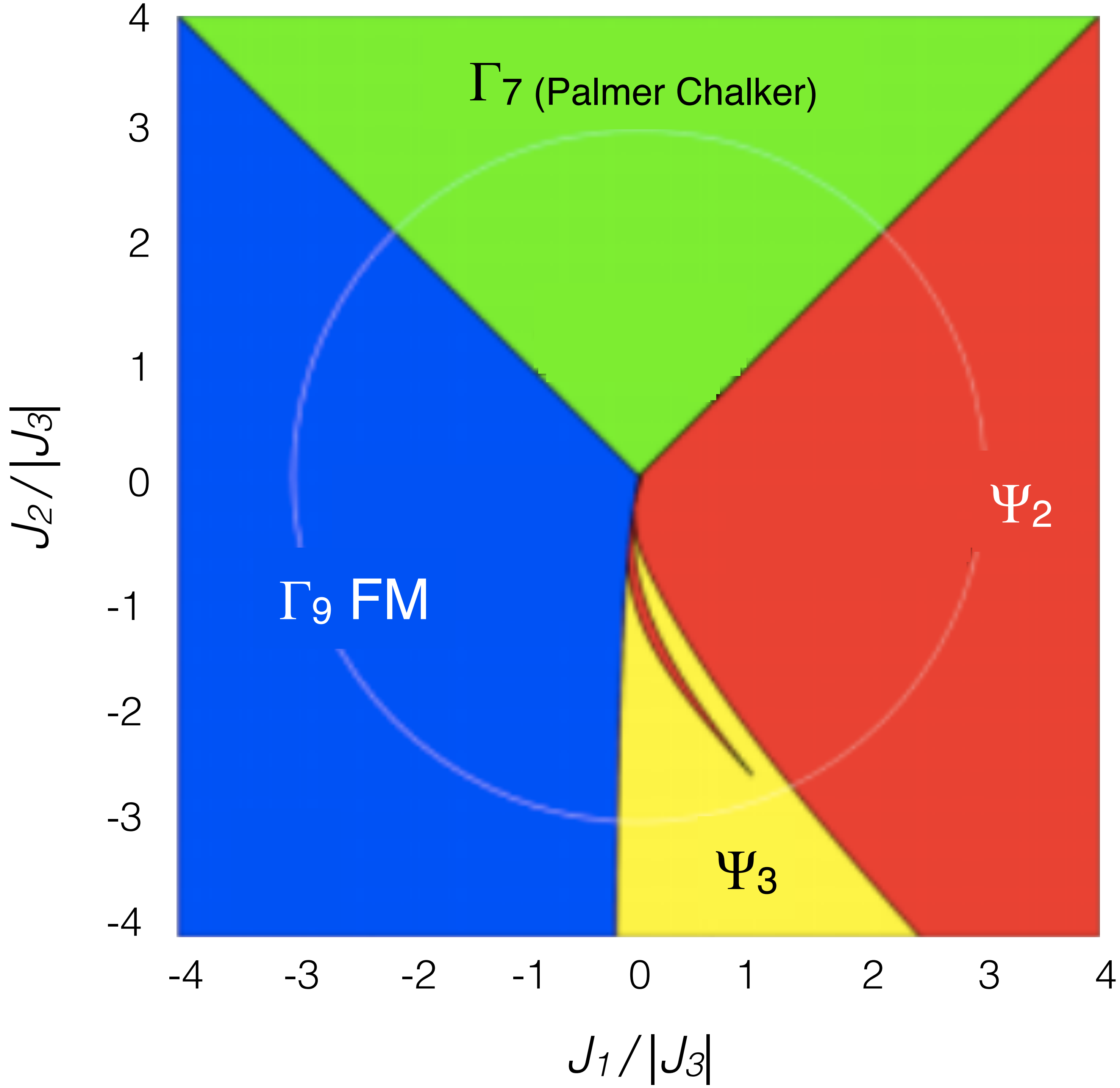
K.A. Ross, Y. Qiu, J.R.D. Copley, H.A. Dabkowska, and B.D. Gaulin, Phys. Rev. Lett., 112, 057201, 2014

The anisotropic exchange Hamiltonian predicts a rich phase diagram for the XY pyrochlores

$$\mathcal{H} = \sum_{<ij>} \vec{S}_i \vec{J}_{ij} \vec{S}_j$$

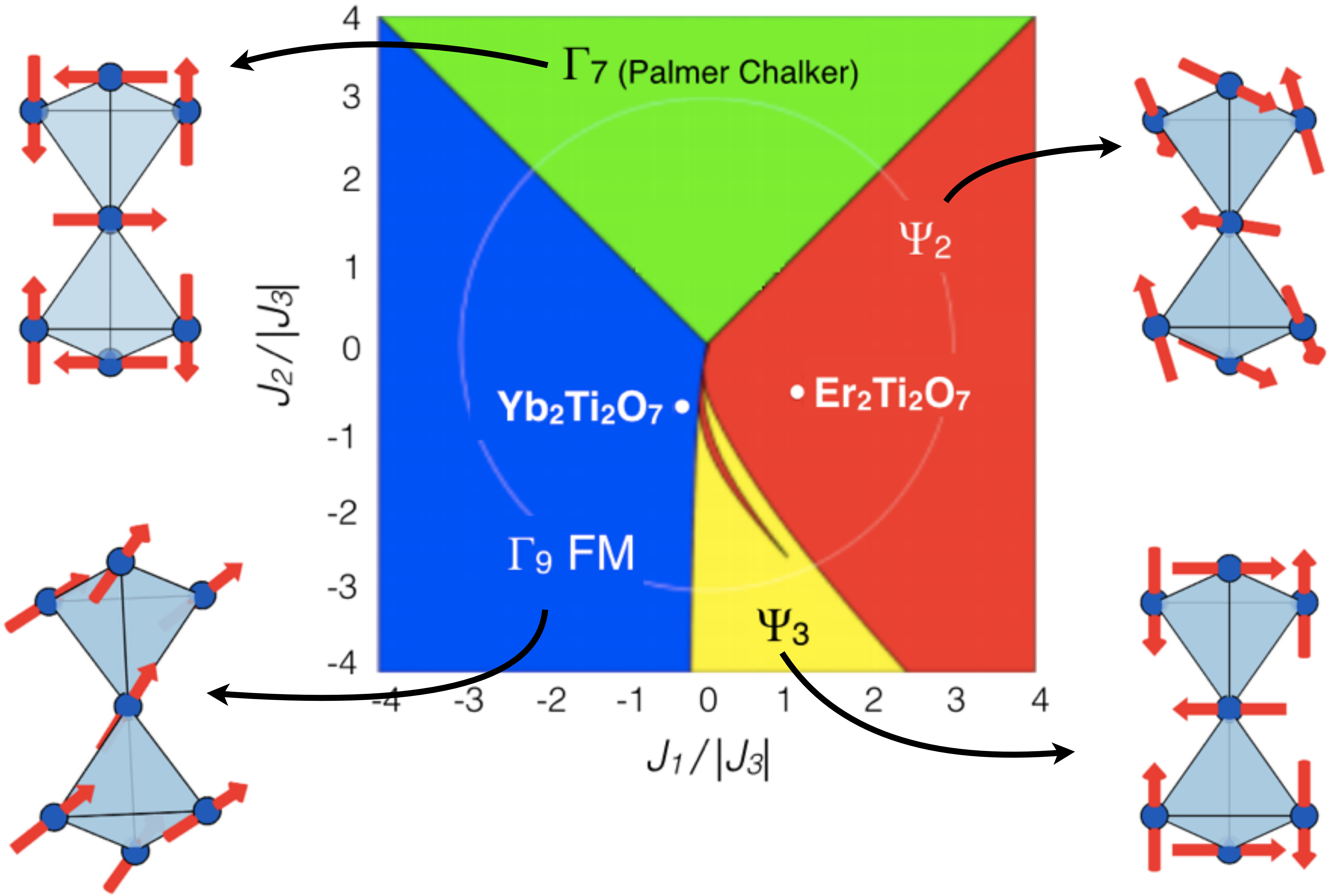


J_1 = XY
J_2 = Ising
J_3 = Symmetric off-diagonal
J_4 = Dzyaloshinskii-Moriya



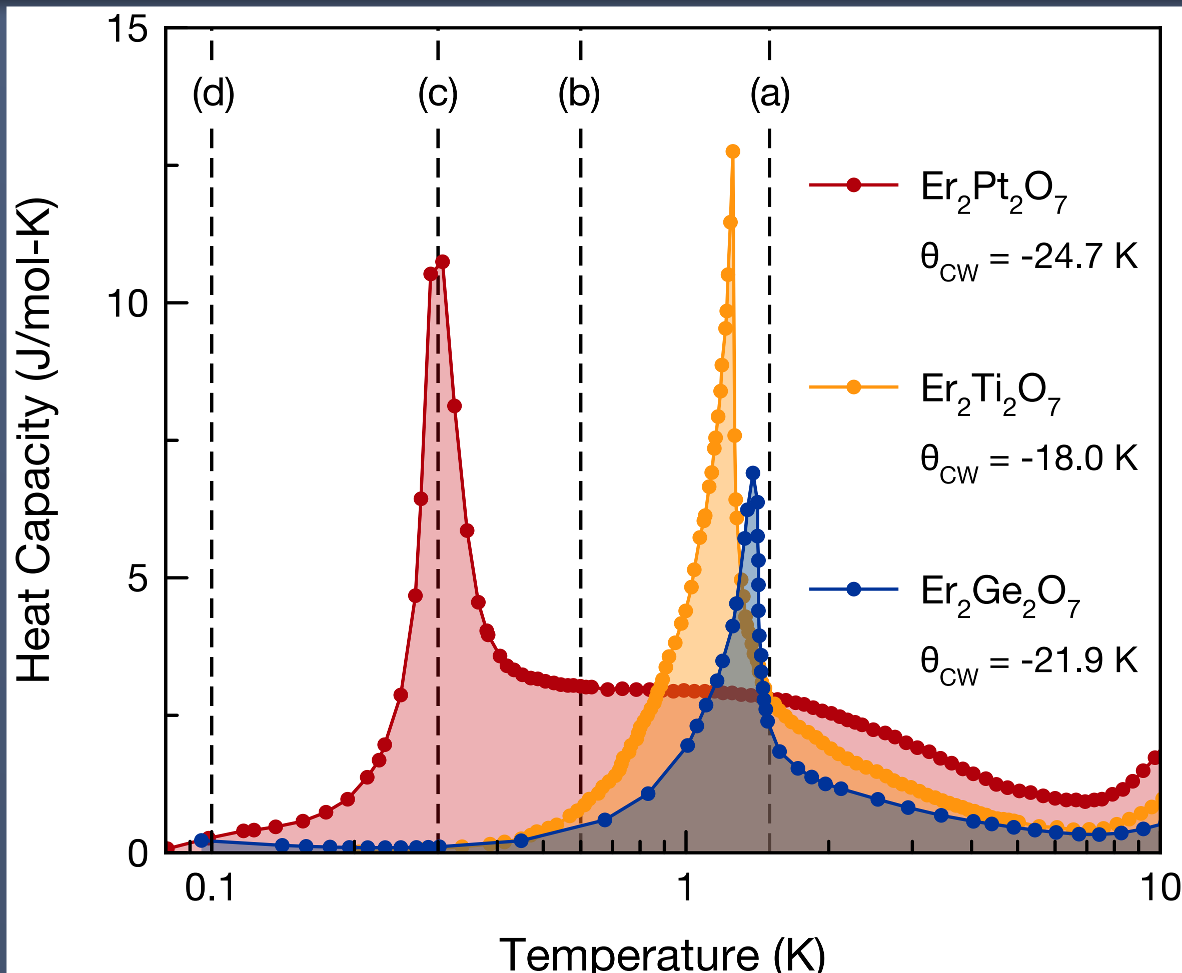
Yan *et al.*, Phys. Rev. B 95, 094422 (2017)

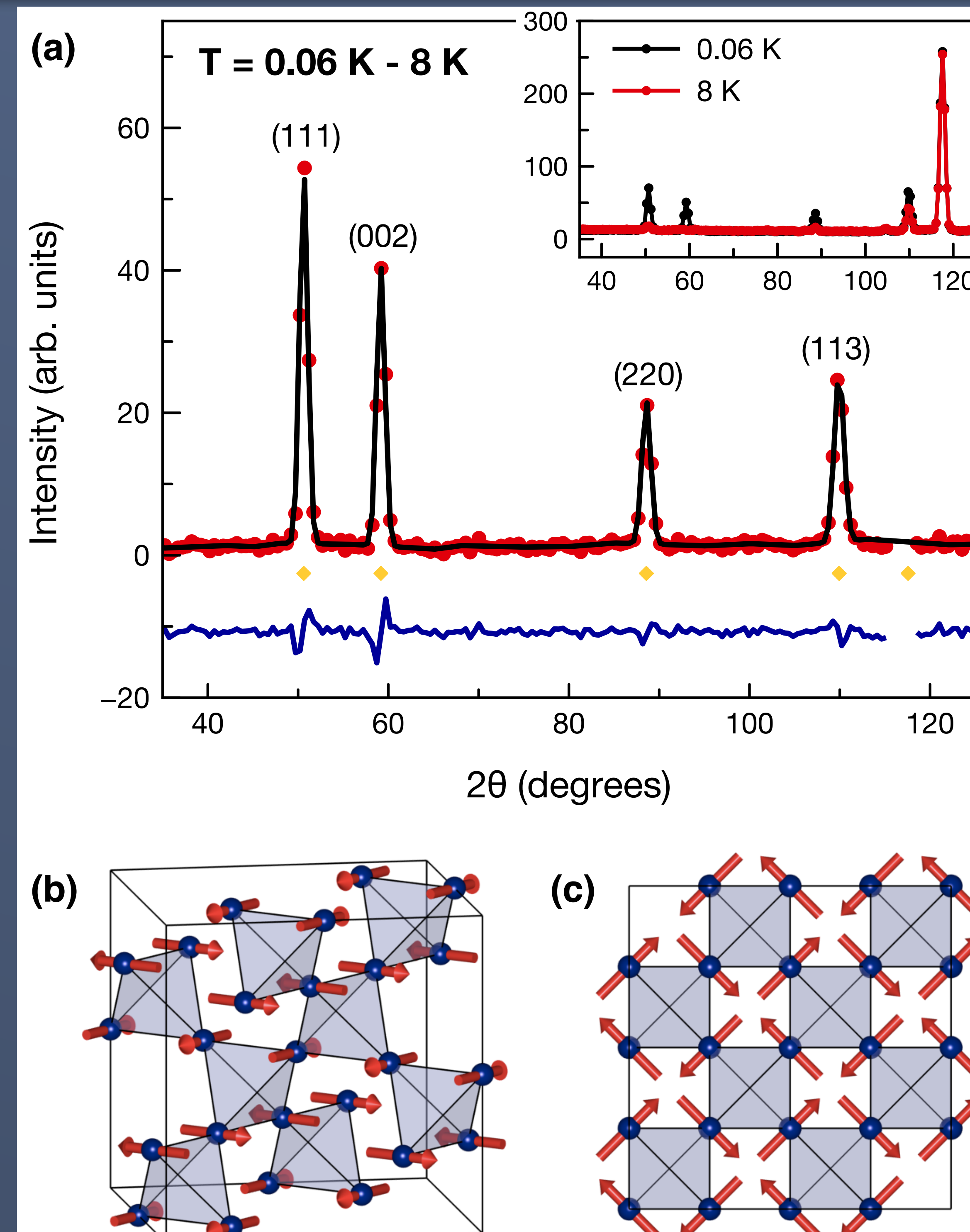
The anisotropic exchange Hamiltonian predicts a rich phase diagram for the XY pyrochlores



Yan *et al.*, Phys. Rev. B 95, 094422 (2017)

$Er_2Pt_2O_7$:
Palmer-Chalker (Γ_7) Phase at much reduced T_N

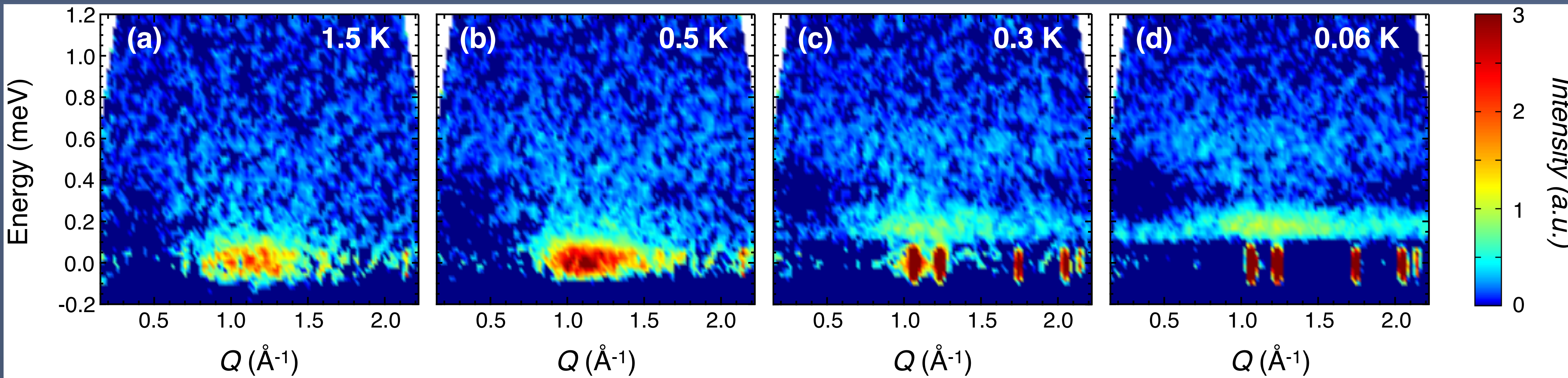




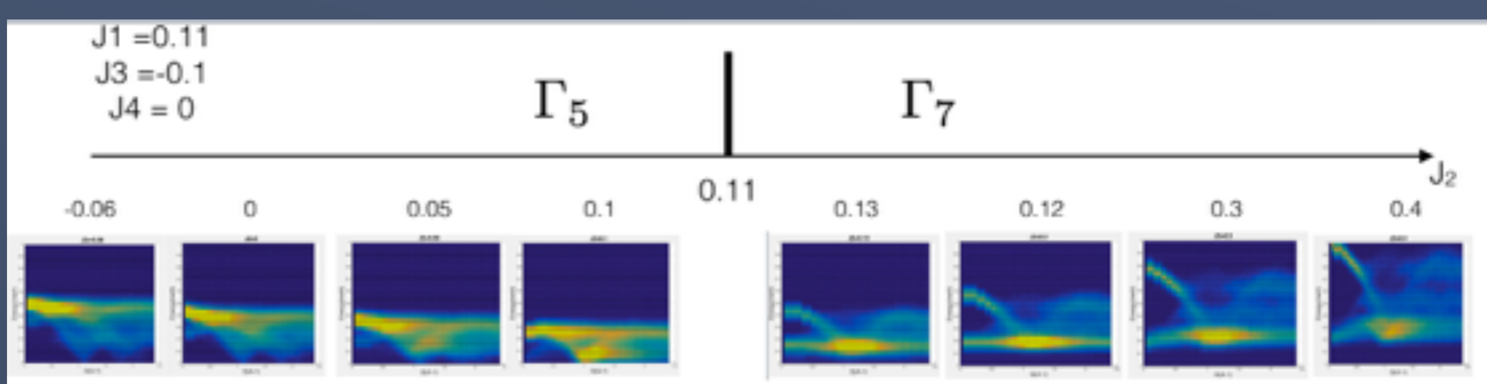
Evidence that small changes in anisotropic exchange can easily induce different ordered phase(s): $\Gamma_5 \longrightarrow \Gamma_7$

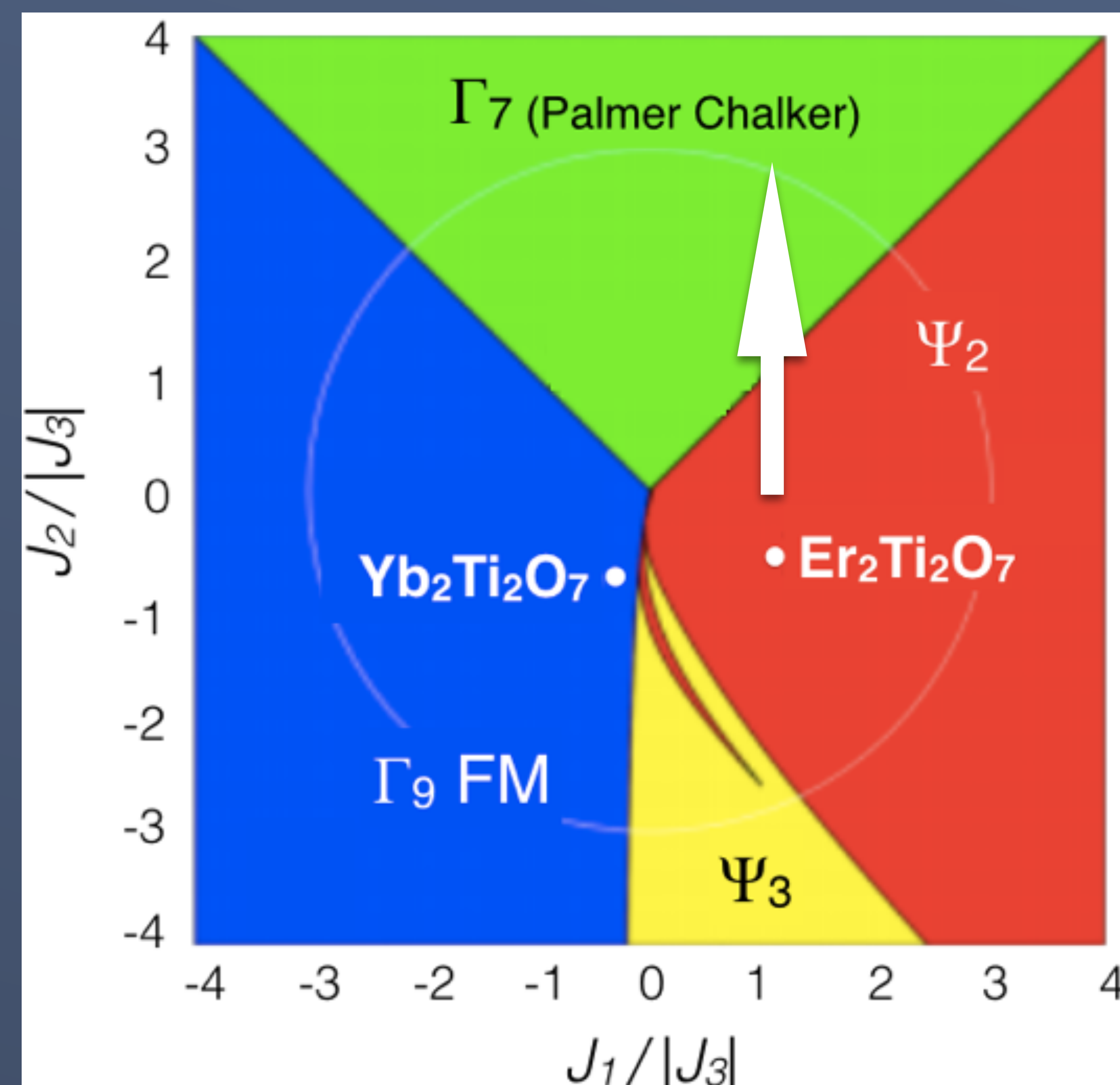
A.M. Hallas, J. Gaudet et al, arXiv:1705.06680

$Er_2Pt_2O_7$: Inelastic Scattering

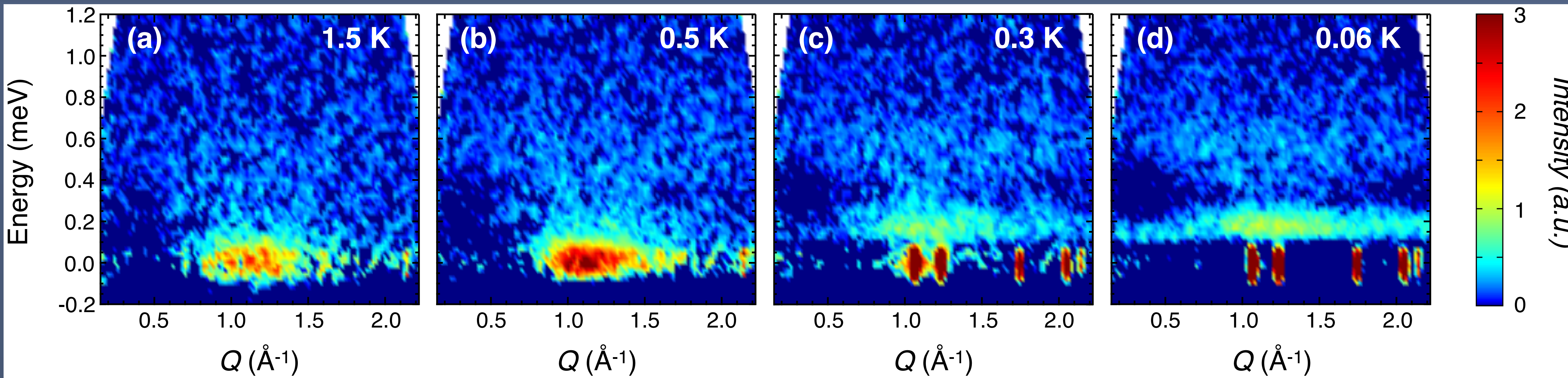


A.M. Hallas, J. Gaudet et al, arXiv:1705.06680

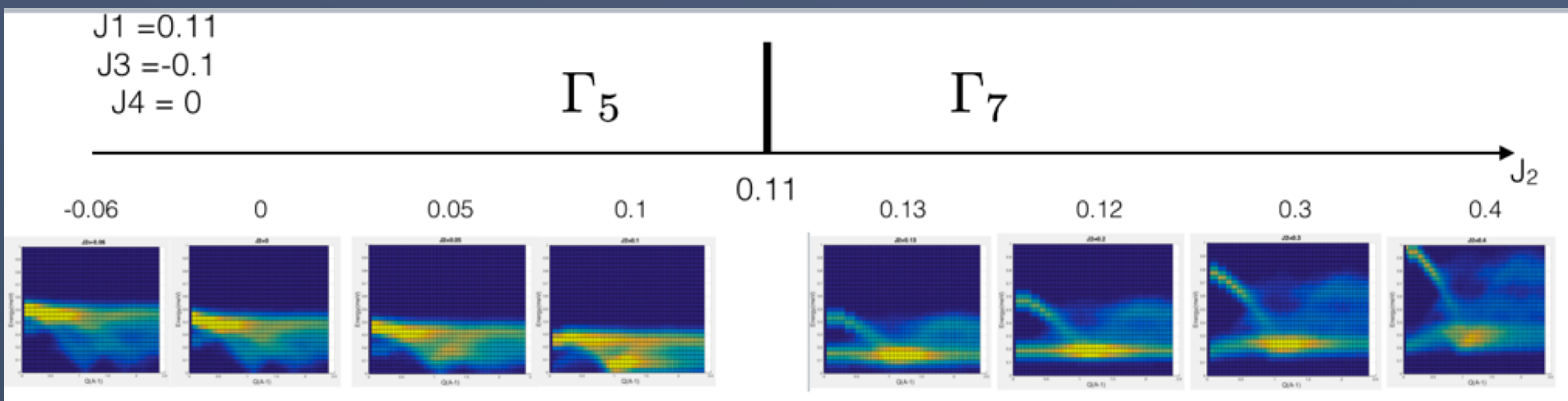




$Er_2Pt_2O_7$: Inelastic Scattering

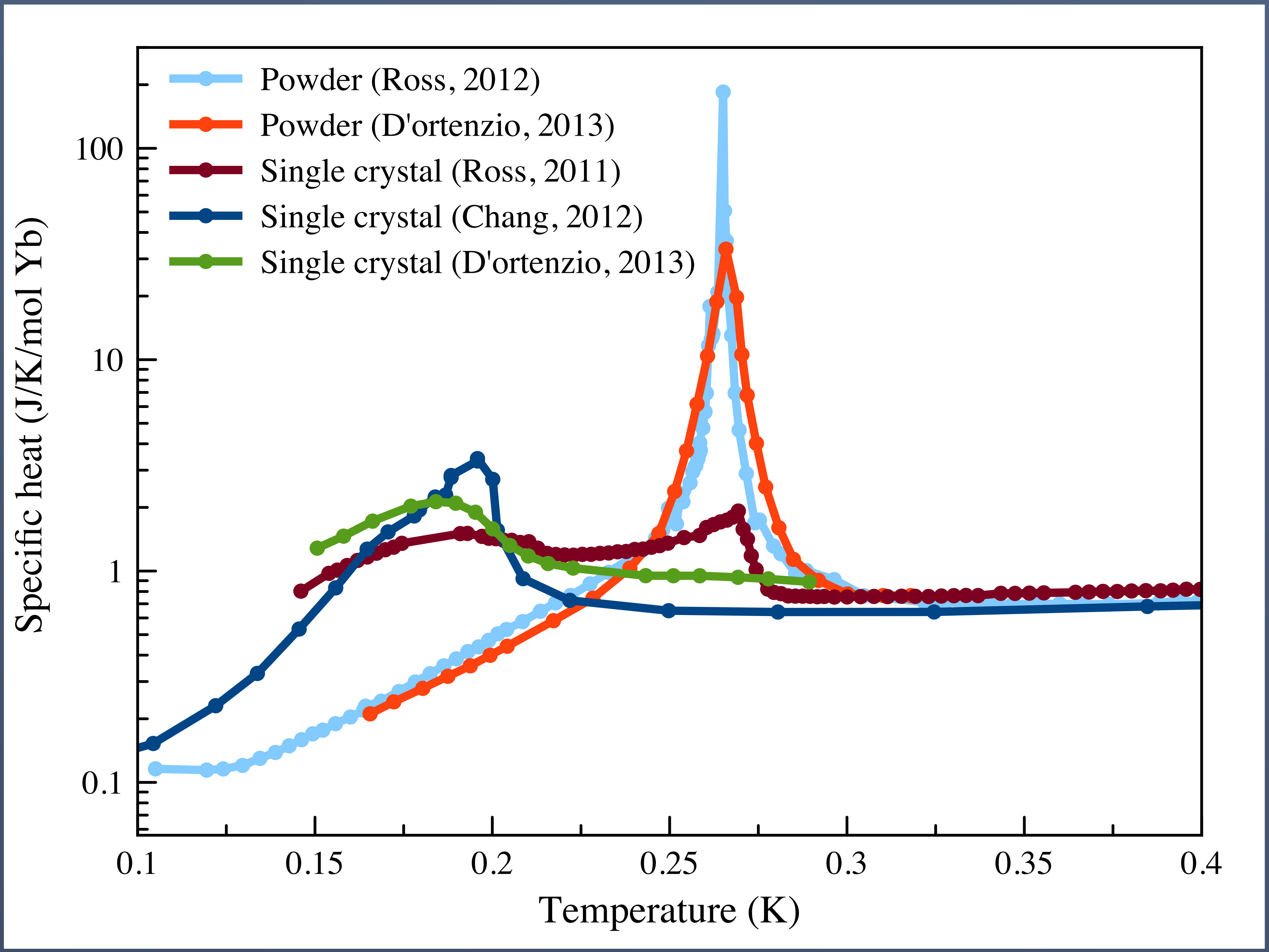


A.M. Hallas, J. Gaudet et al, arXiv:1705.06680



Low energy spin excitation is gapped and dispersion less near Γ_7 to Γ_5 phase boundary

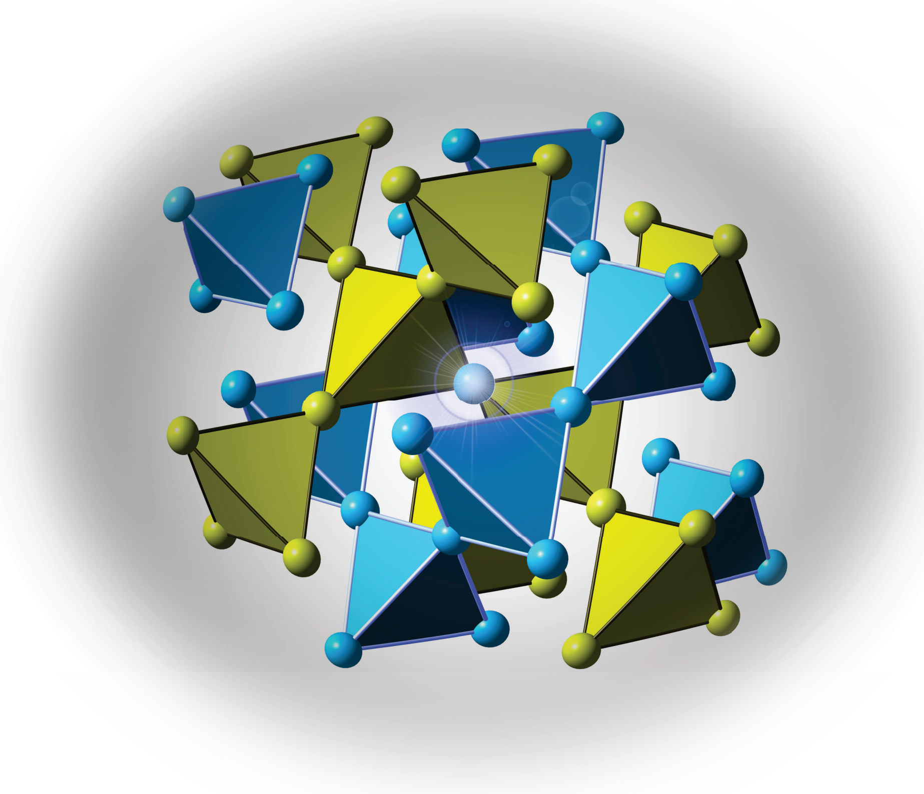
$Yb_2Ti_2O_7$: Sample dependent ground state properties

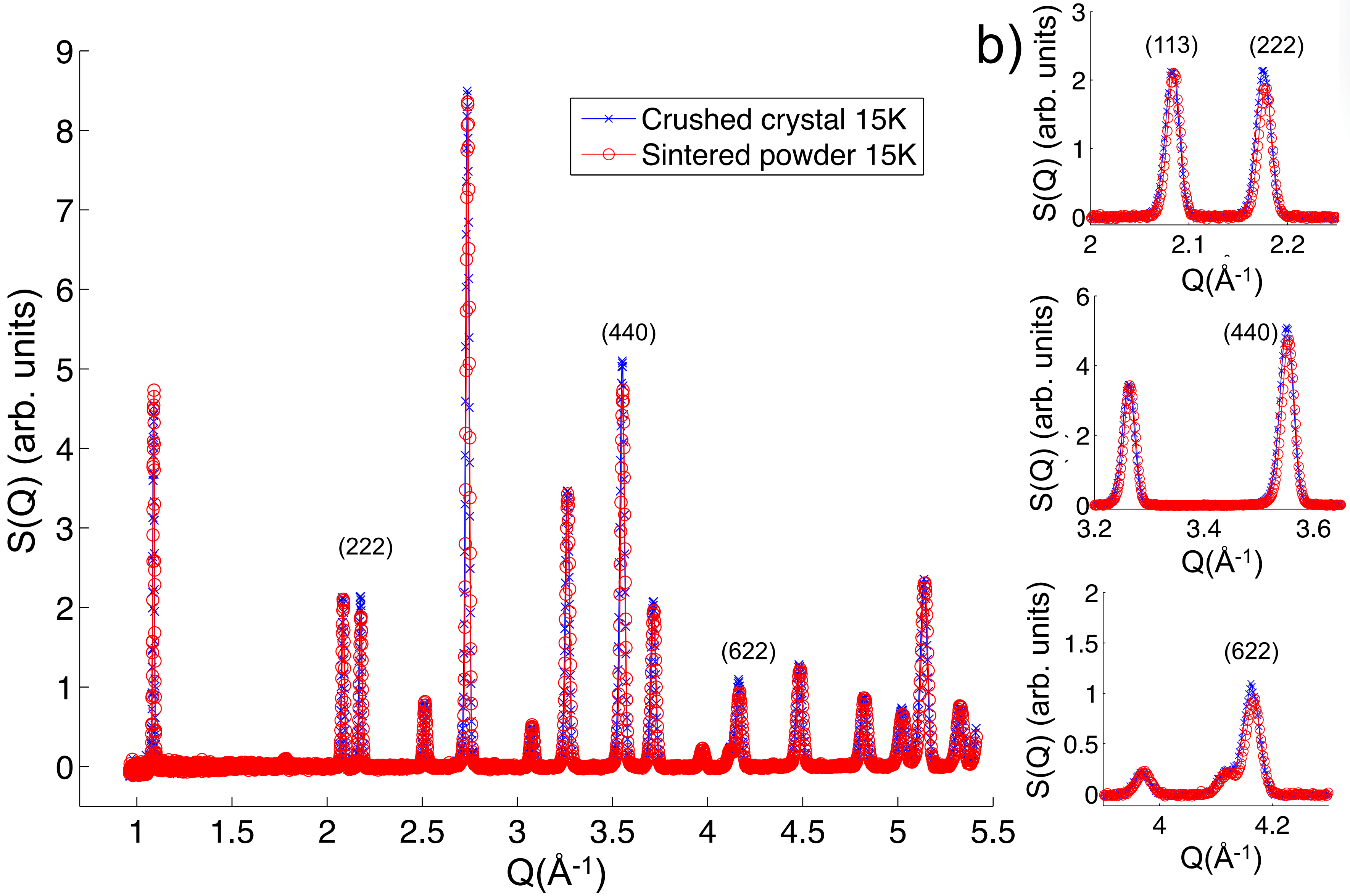


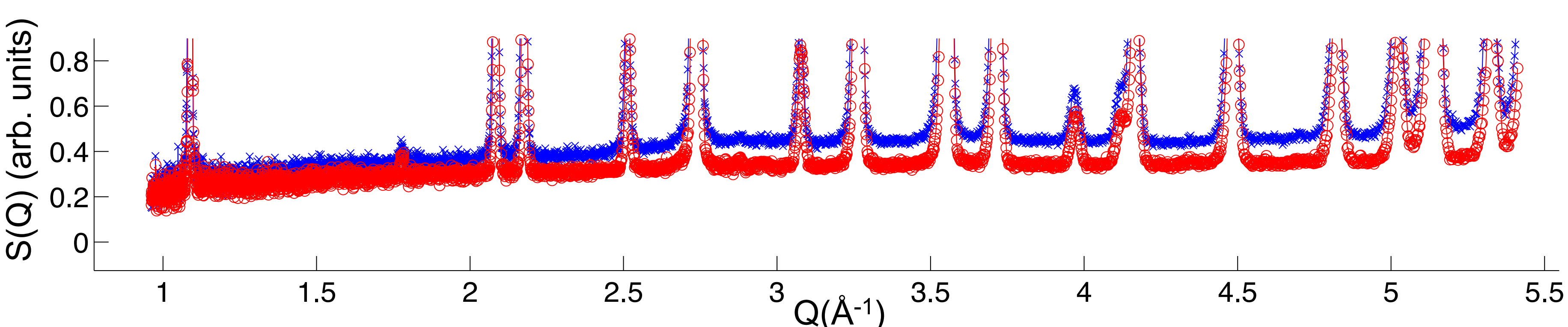
Powder samples grown from solid state synthesis tend to have T_C ~ 0.26 and very sharp C_P anomalies

Single crystal samples grown using FZ techniques tend to have lower T_Cs (0.15 - 0.22 K) and broader, sometimes multiple, and weaker C_P anomalies

What is different between powder and single crystal $Yb_2Ti_2O_7$?







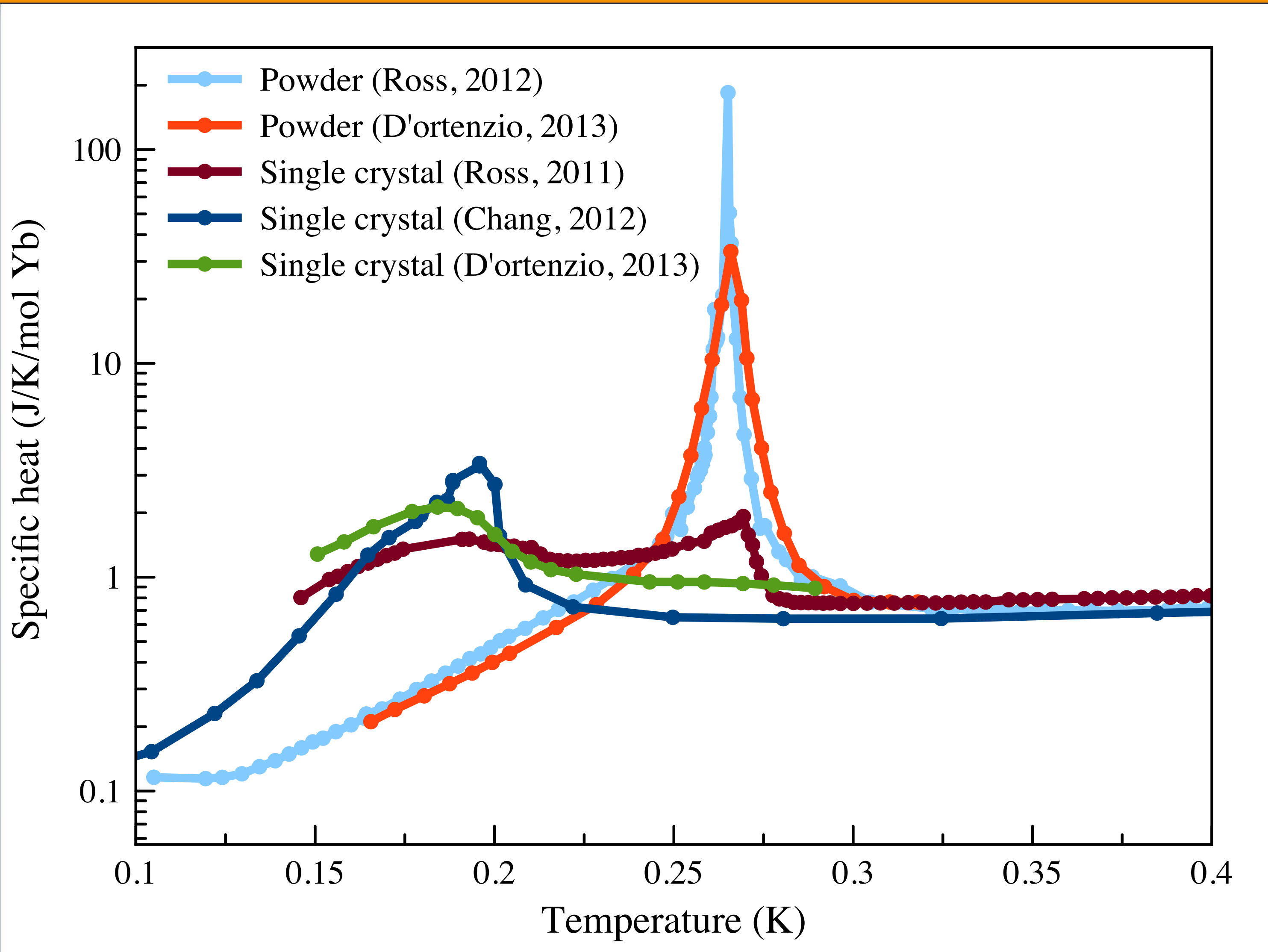
$Yb_{2+x}Ti_{2-x}O_{7-y}$

Our powders are stoichiometric: x=0

Our floating zone grown single crystals tend to be "lightly stuffed" x ~ 0.04

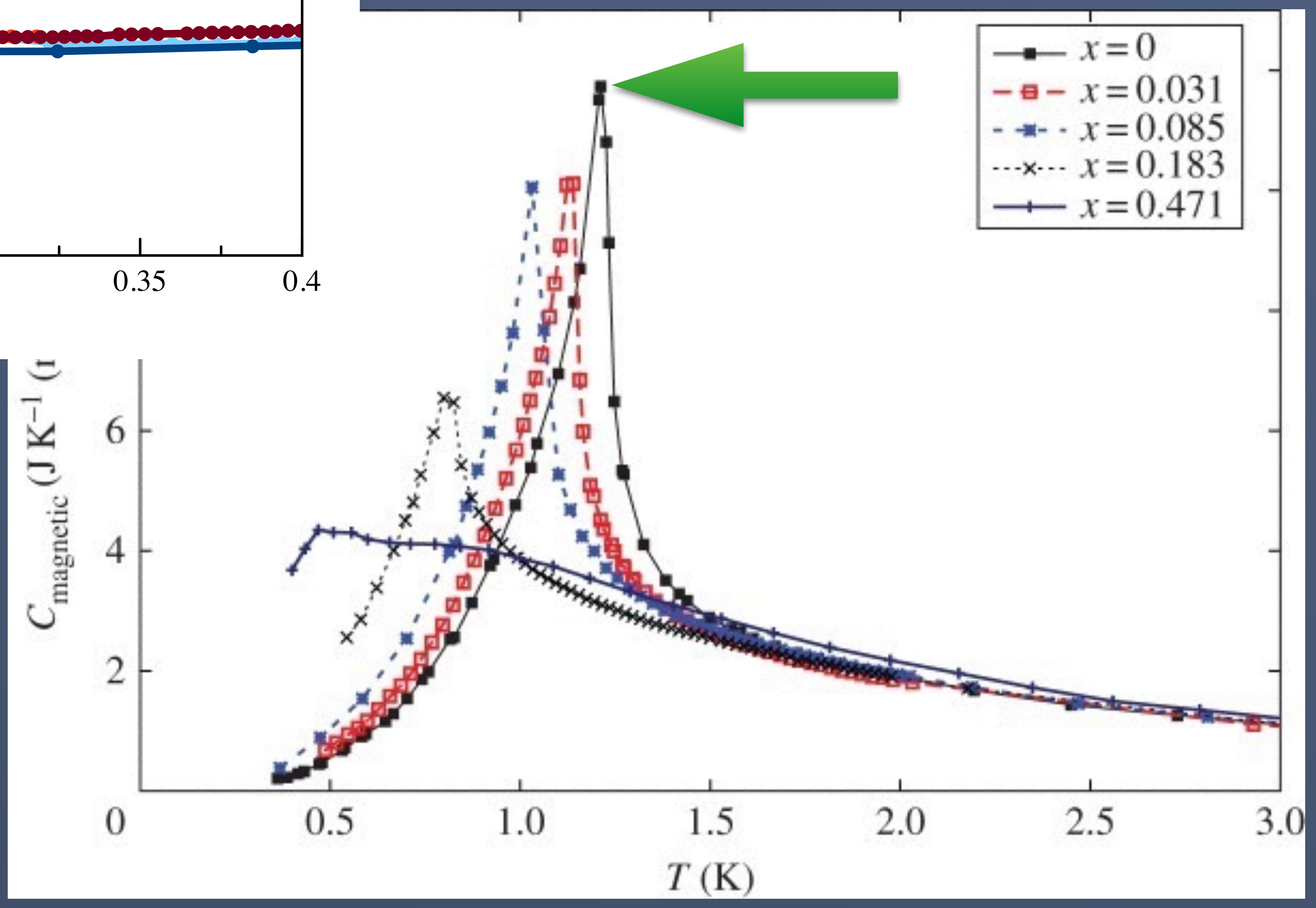
K.A. Ross et al., Phys. Rev. B 86, 174424 (2012)

In contrast, $Er_2Ti_2O_7$ shows little or no sample variability



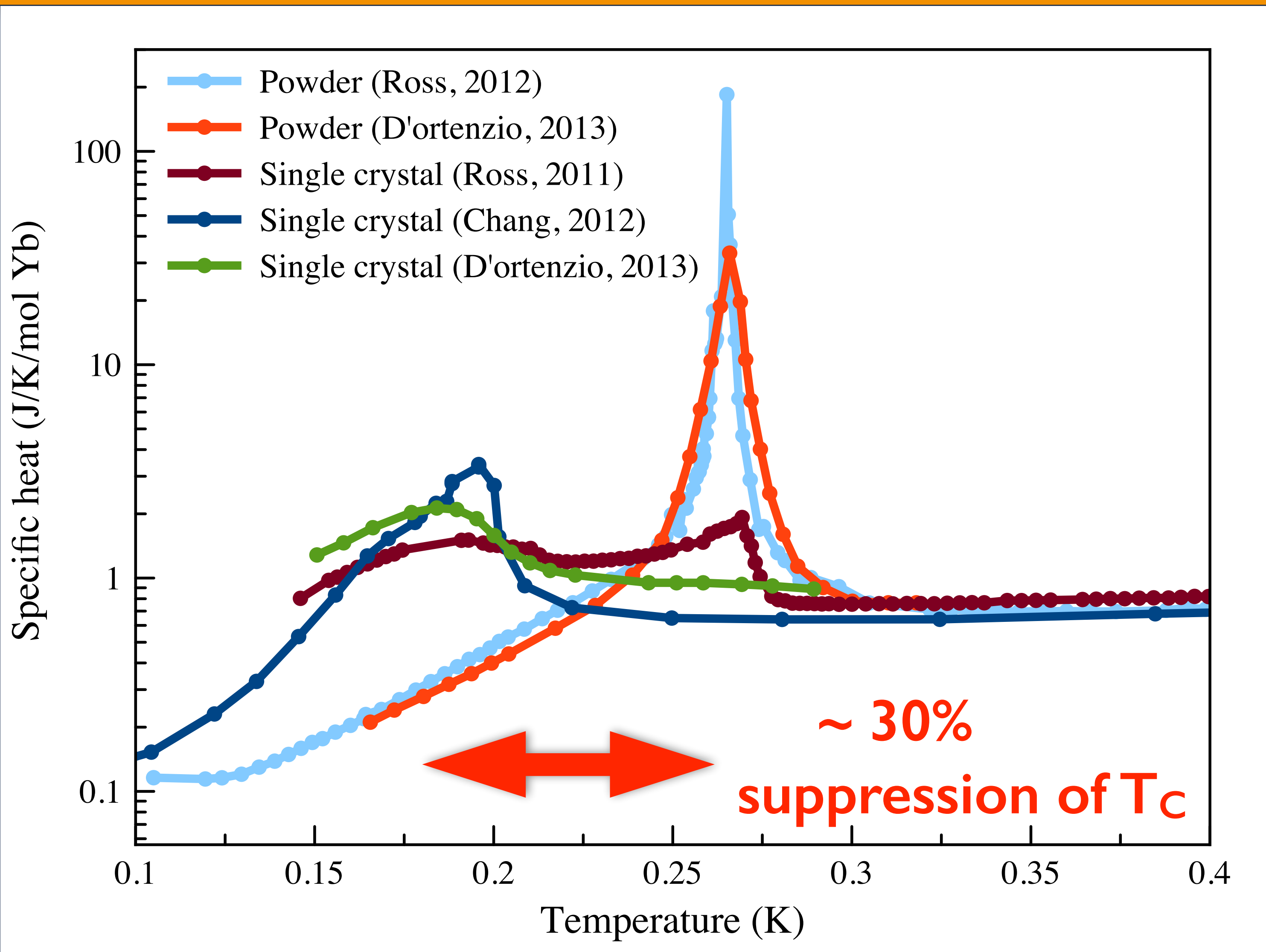
$Er_{(2-x)}Y_{(x)}Ti_2O_7$

- **Consistent with 3D percolation**



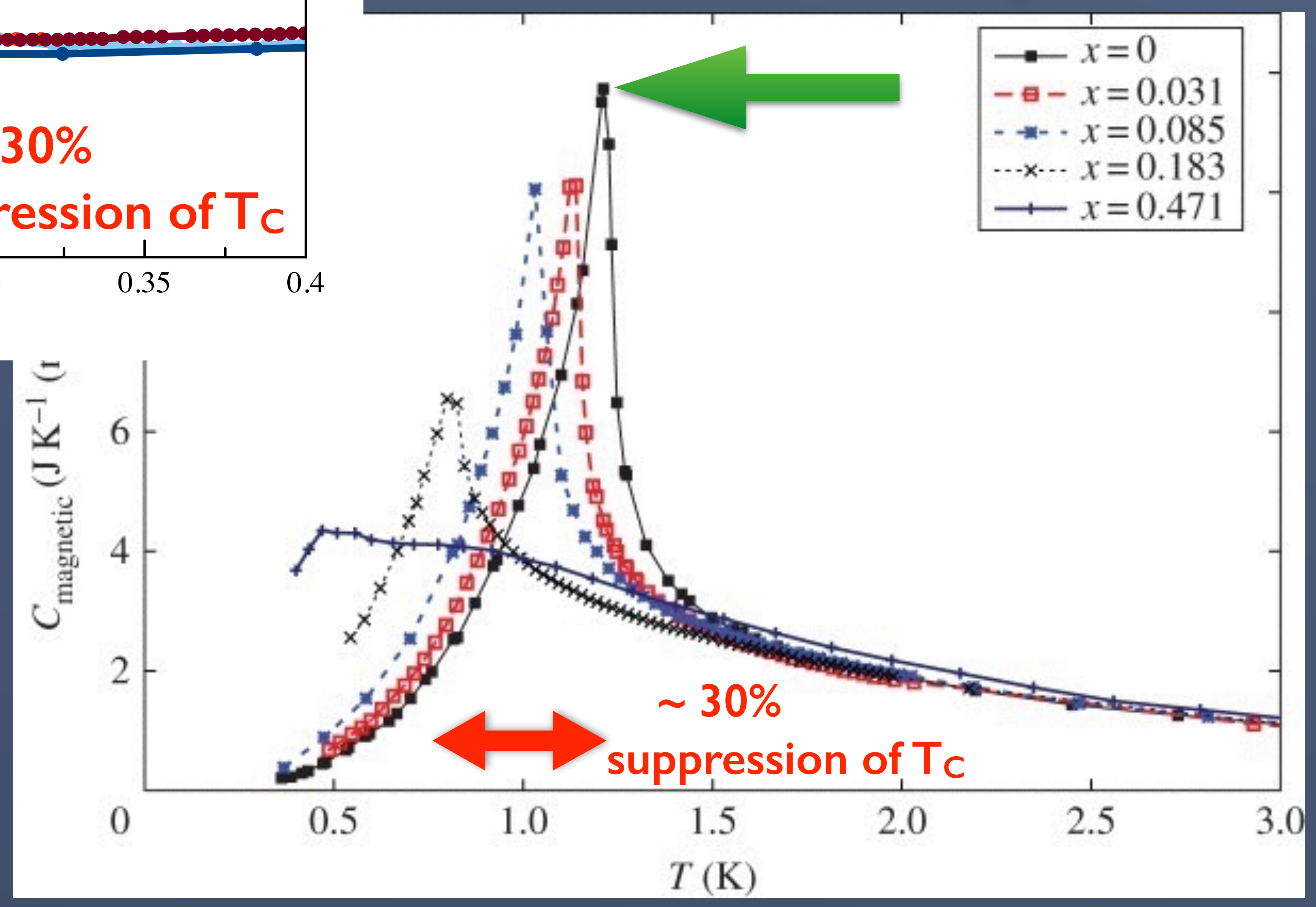
Niven et al,
Proc. R. Soc. A. 470, 2171 (2014)

In contrast, $Er_2Ti_2O_7$ shows little or no sample variability



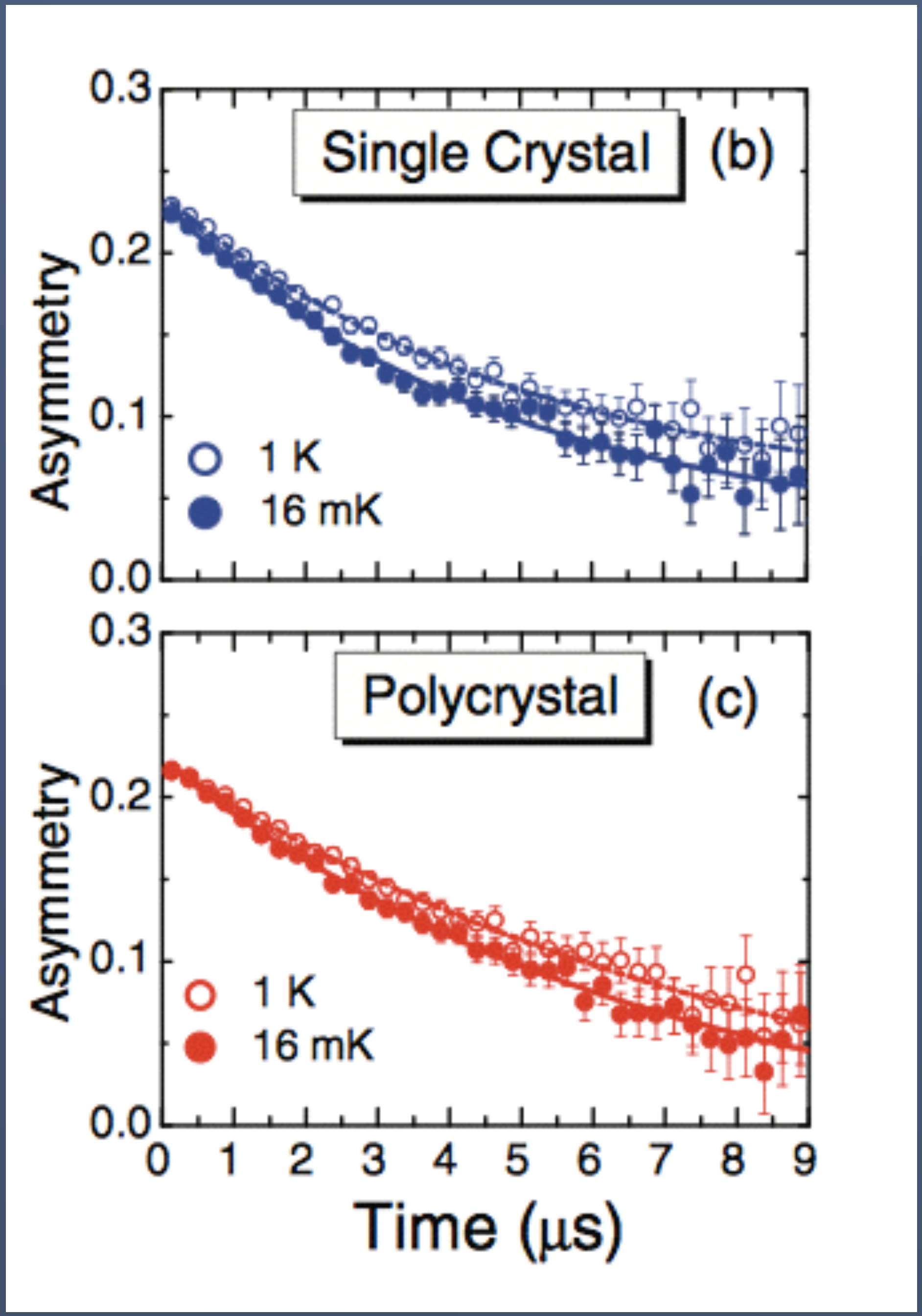
$Er_{(2-x)}Y_{(x)}Ti_2O_7$

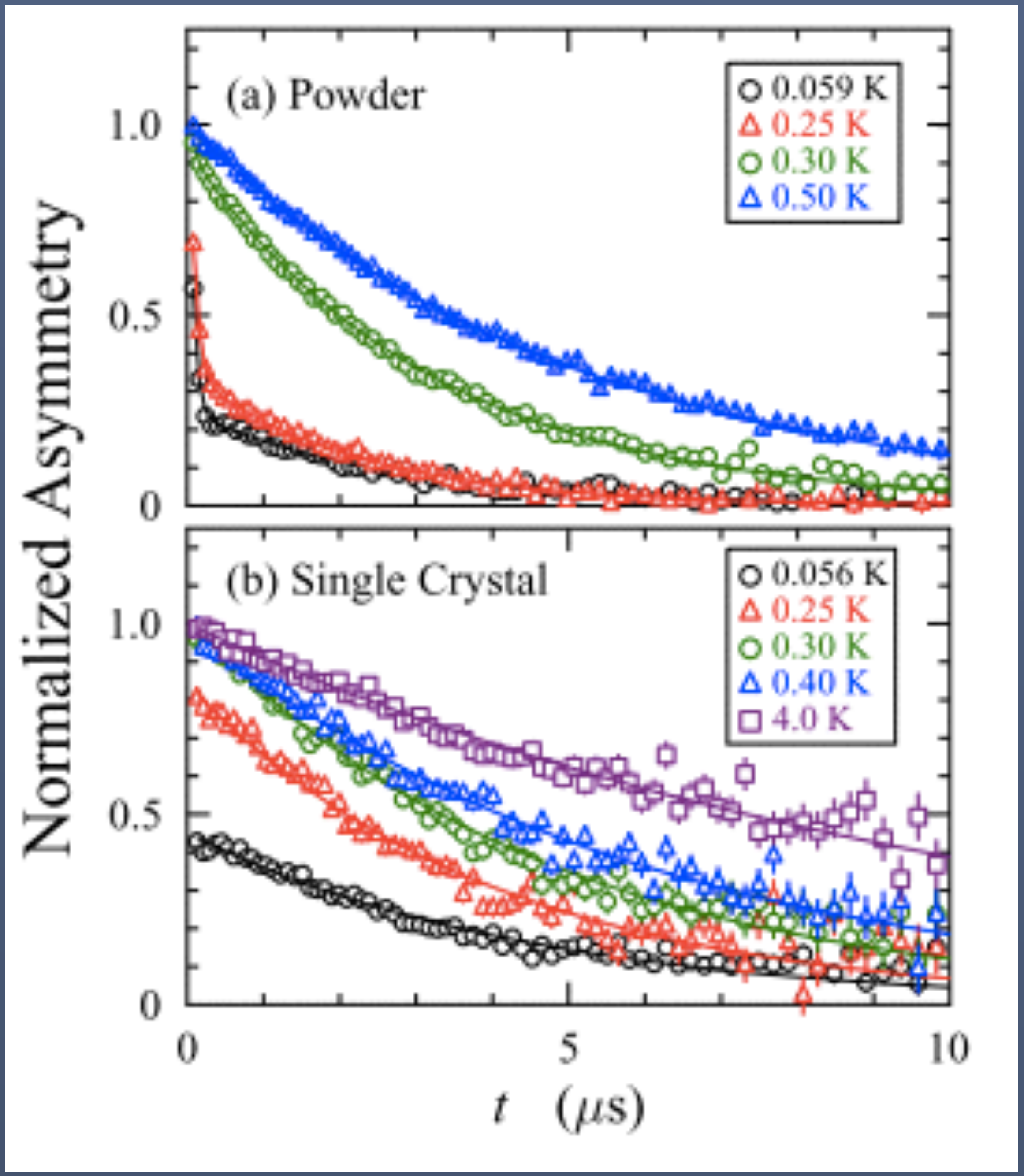
- **Consistent with 3D percolation**



Niven et al,
Proc. R. Soc. A. 470, 2171 (2014)

Magnetic properties (muons, neutrons ..) also show sample dependence about T_C ~ 0.15 - 0.26 K

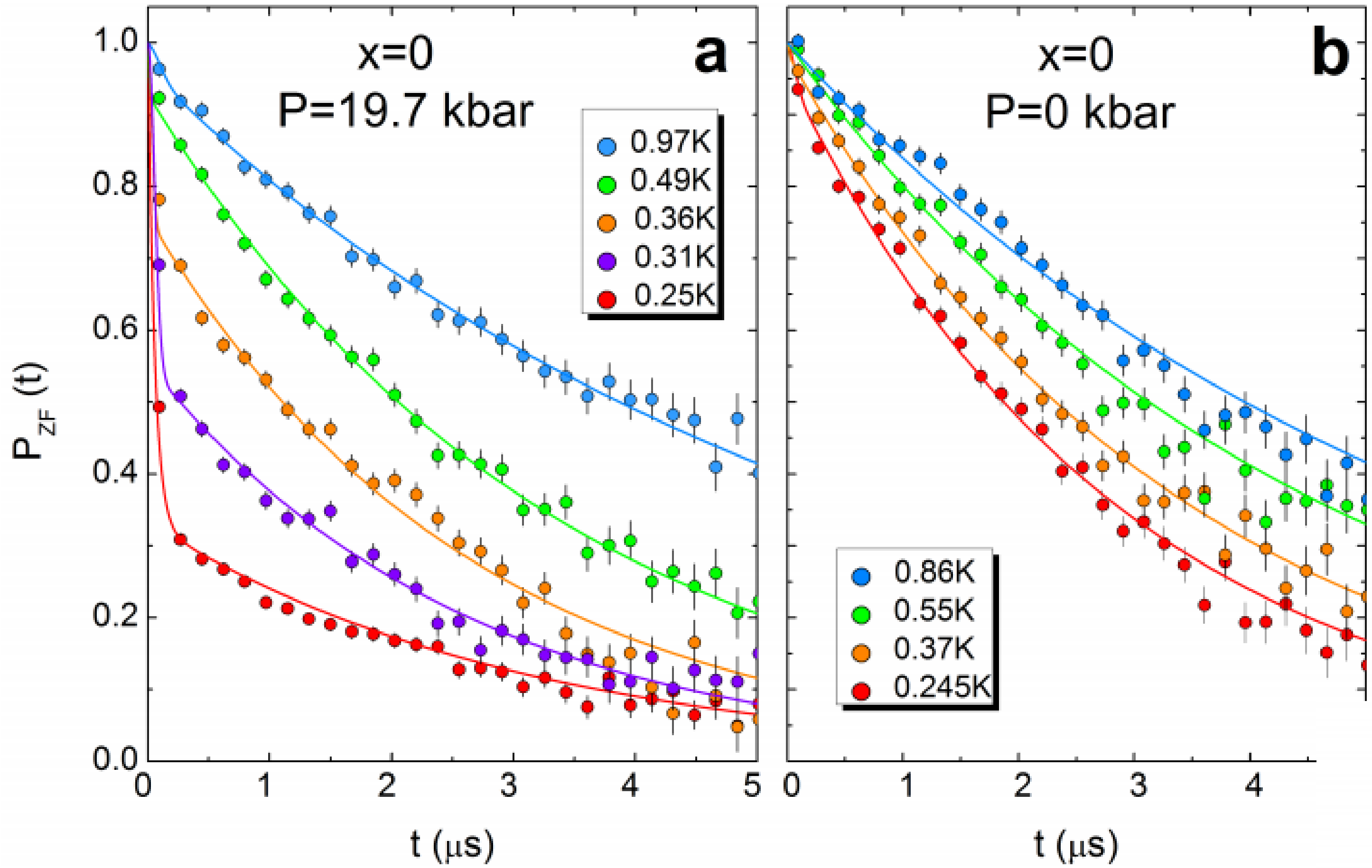




Disorder at 16 mK; no phase transition
D'ortenzio et al, Phys. Rev. B 88, 134428 (2013)

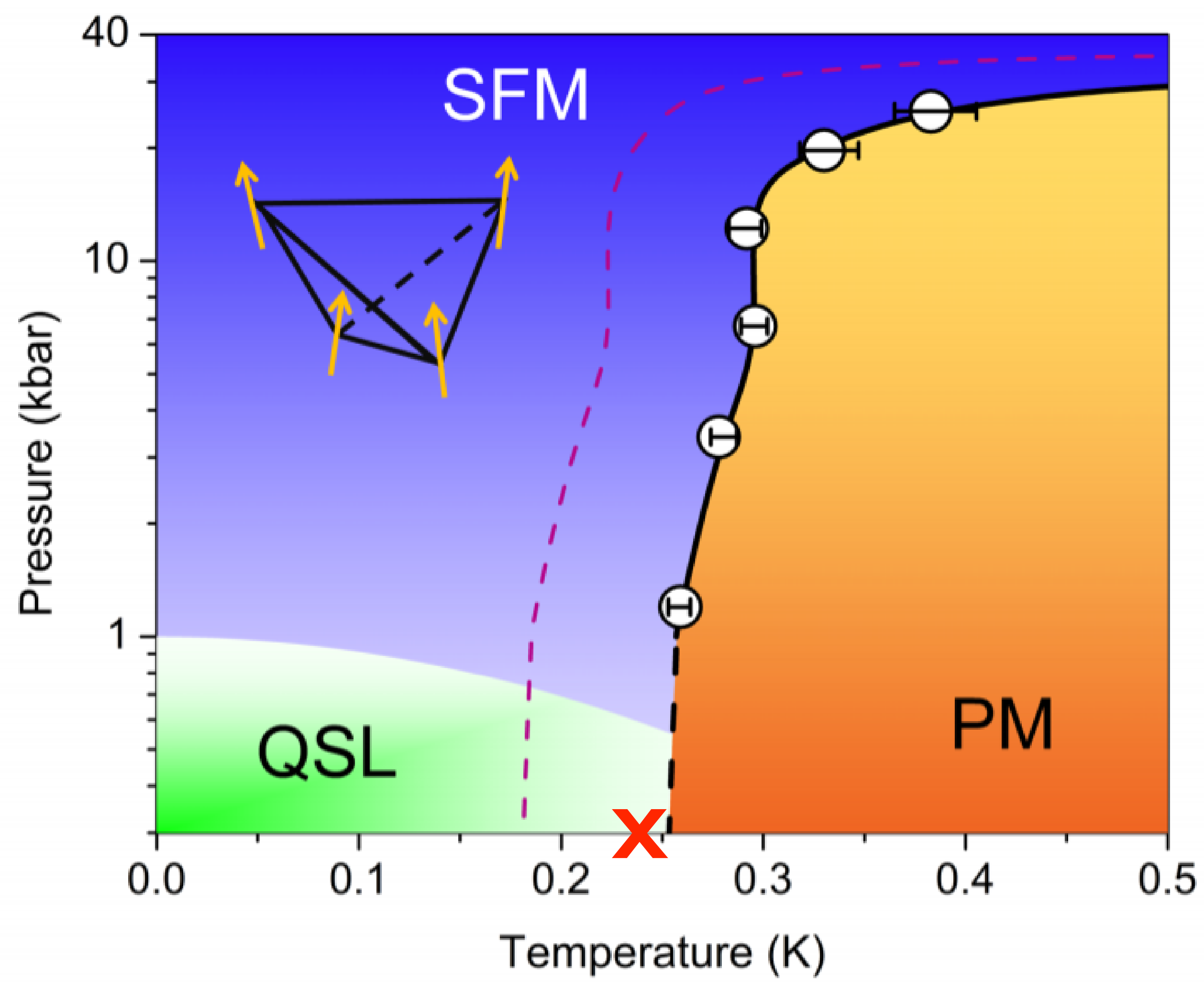
Order at 50 mK; T_C ~ 0.25 K
Chang et al, Phys. Rev. B 89, 184416 (2014)

Stoichiometric $Yb_2Ti_2O_7$ does order *under pressure*!



Fraction of magnetic order ~ fast drop off in P_{zf} (t) at small t

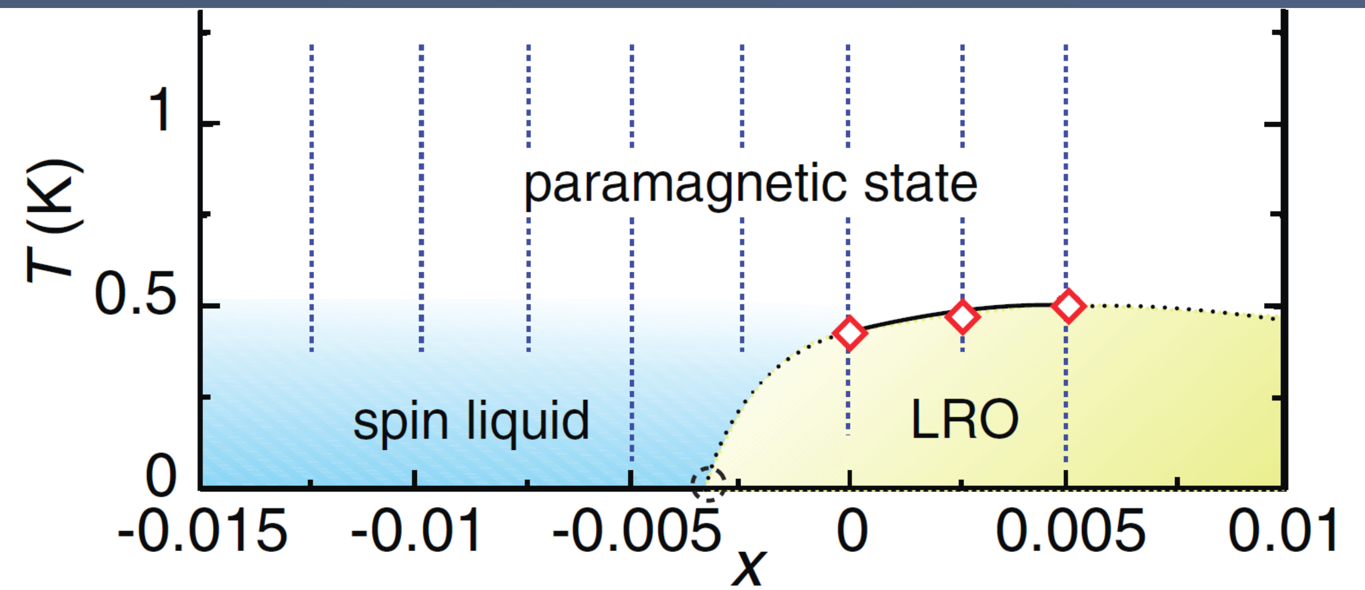
$$\mathrm{P}_{\mathrm{zf}}(t) = f\left(\frac{2}{3}e^{-\Delta^2 t^2/2} + \frac{1}{3}e^{-\lambda t}\right) + (1-f)e^{-\lambda t}$$



E. Kermarrec, J. Gaudet et al,
Nature Communications, 8, 14810 (2017)

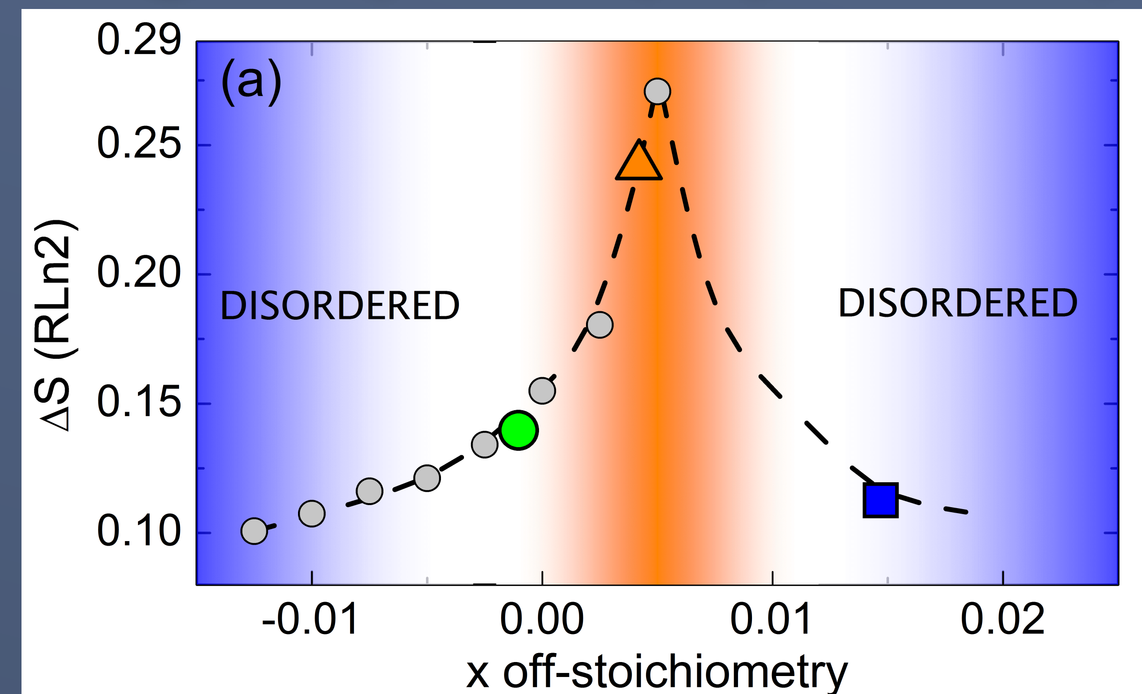
$Tb_2Ti_2O_7$, another QSI candidate, also displays extreme sensitivity to stoichiometry

powders

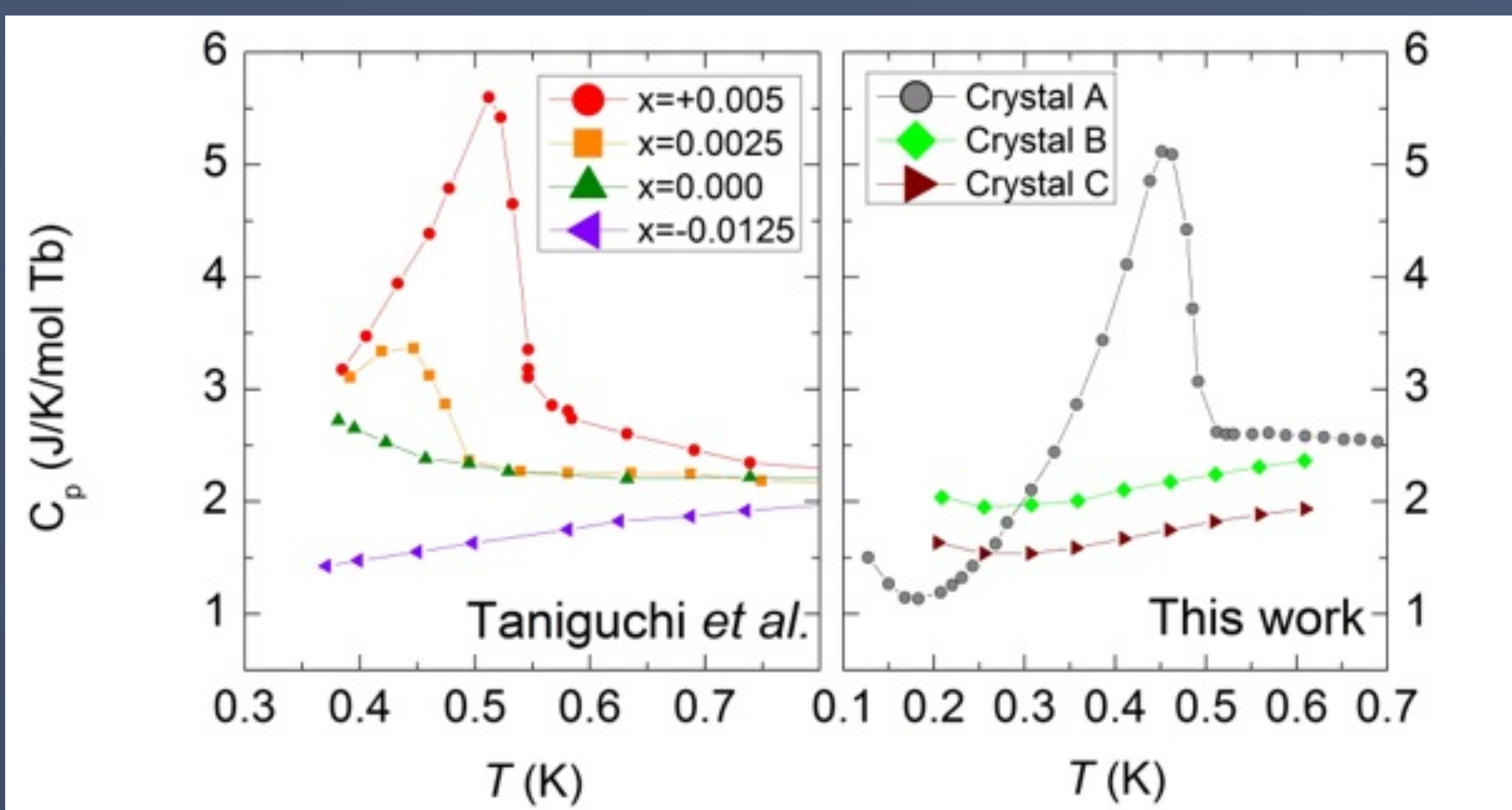


Taniguchi et al, PRB, 87, 060408(R), 2013.

single crystals (& powders)



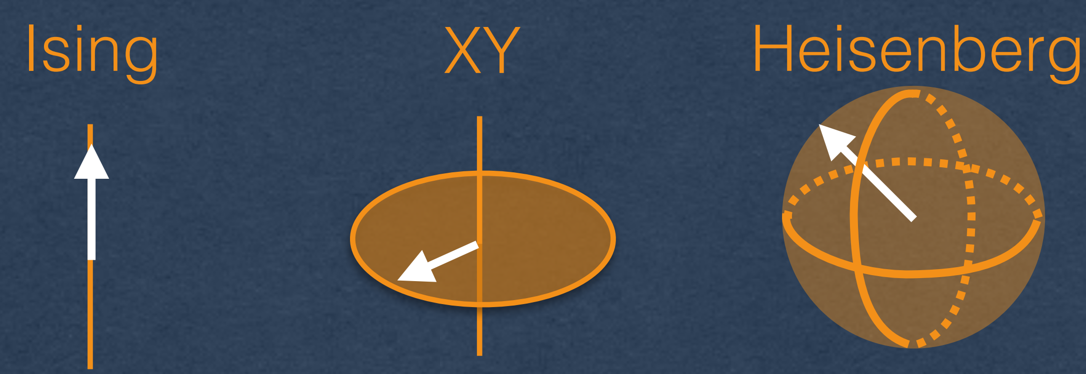
Kermarrec et al, PRB, 92, 245114 (2015)

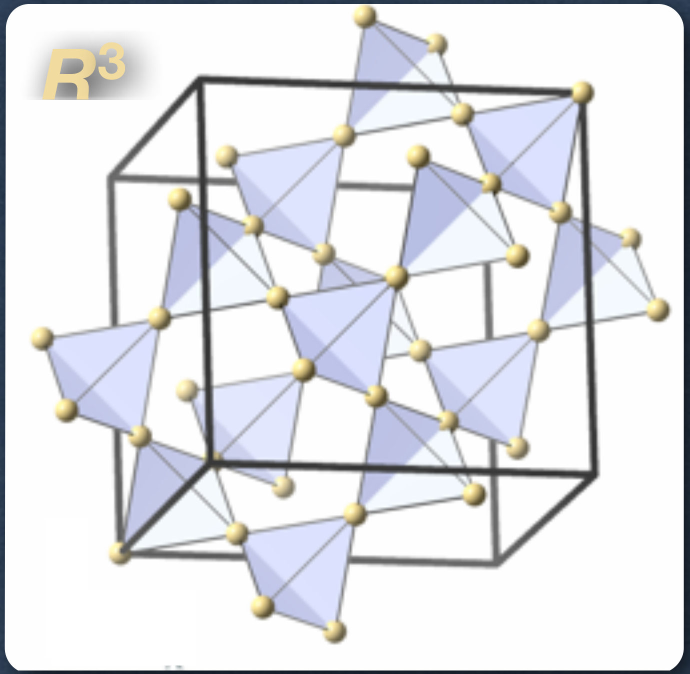


$Tb_{(2+x)}Ti_{(2-x)}O_{7-y}$

Sensitivity to disorder: Its not a bug, its a feature!

$R_2Ti_2O_7$ "Rare earth titanates"

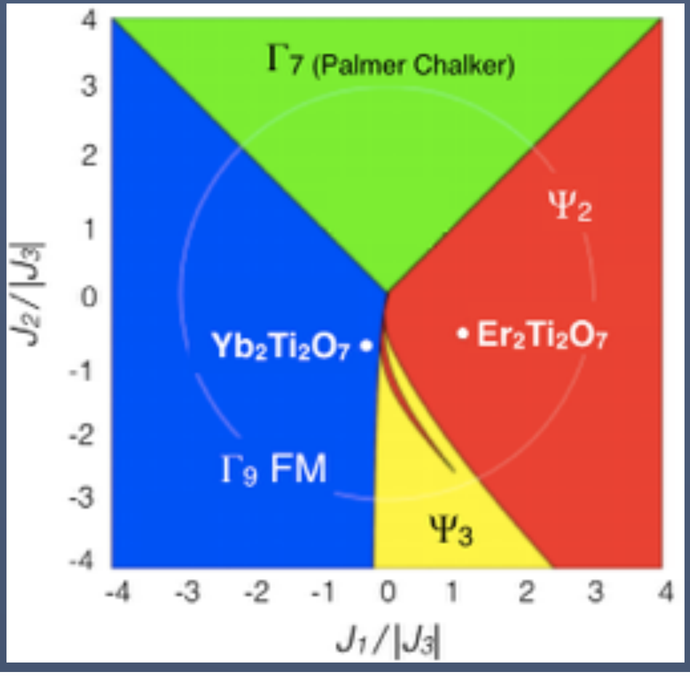




	Anisotropy	Interactions	Ground state	Sensitivity to disorder
Ho, Dy	Ising	FM	spin ice	-
Tb	Ising	AFM	**quantum spin ice**	**very high**
Gd	Heisenberg	AFM	partial order	-
Er	XY	AFM	**Neel state via ObD**	**little**
Yb	XY	FM	**quantum spin ice**	**very high**

Conclusions:

- Pyrochore magnets can exhibit exotic quantum ground states despite J=large magnetic moments
- Local XY anisotropy gives Seff=1/2 moments in $Yb_2Ti_2O_7$ and $Er_2Ti_2O_7$.
- Anisotropic exchange couples these moments to give a disordered ground state, possibly QSI, in $Yb_2Ti_2O_7$. ObD selects a ψ_2 non-coplanar Neel state in $Er_2Ti_2O_7$; Palmer - Chalker Γ_7 state in $Er_2Pt_2O_7$
- Modern neutron scattering techniques offer remarkable advances for comprehensive, high resolution measurements



Proximity to other ordered phases?

Bruce D. Gaulin
McMaster University