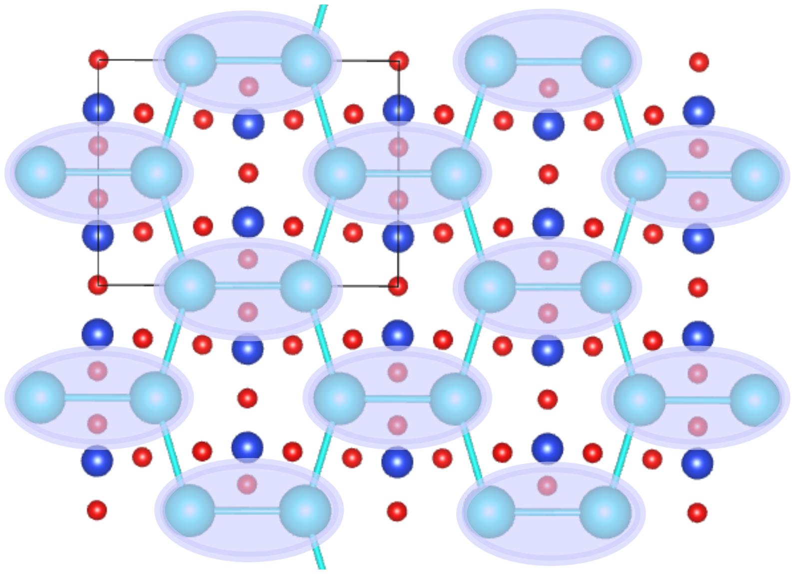
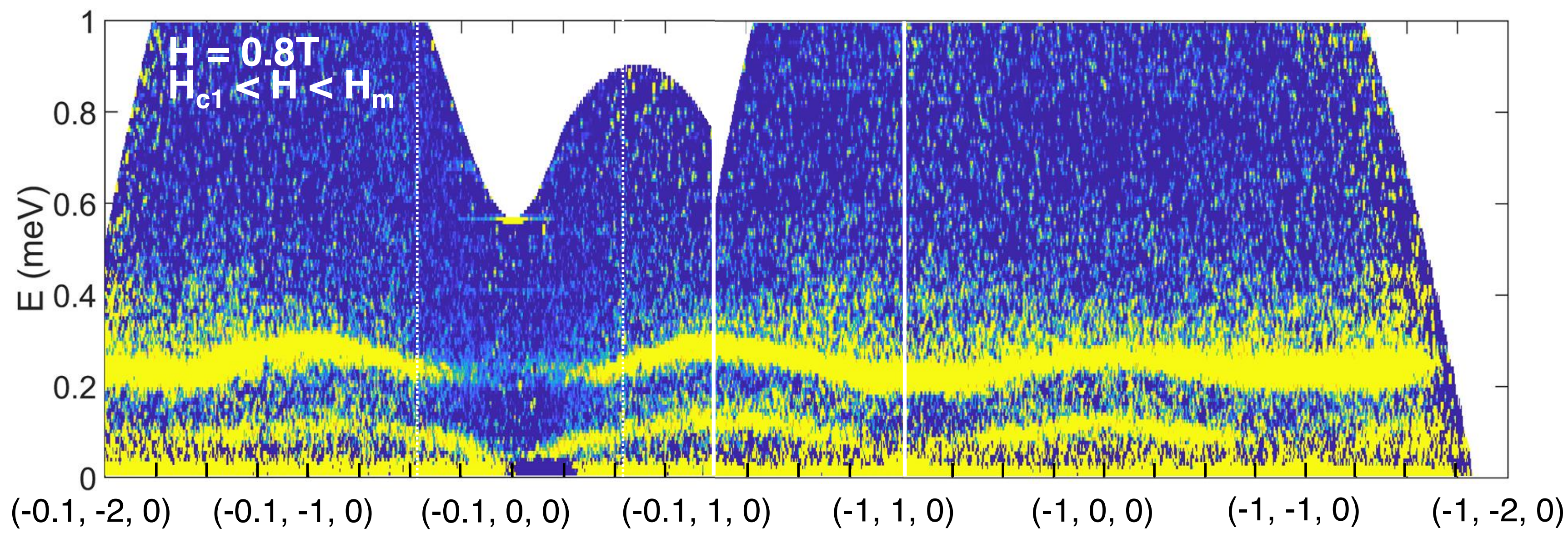
KITP Workshop
October 27, 2020

Using S_{eff} =1/2 for Quantum Magnets: dimer state and BEC dome in $Yb_2Si_2O_7$

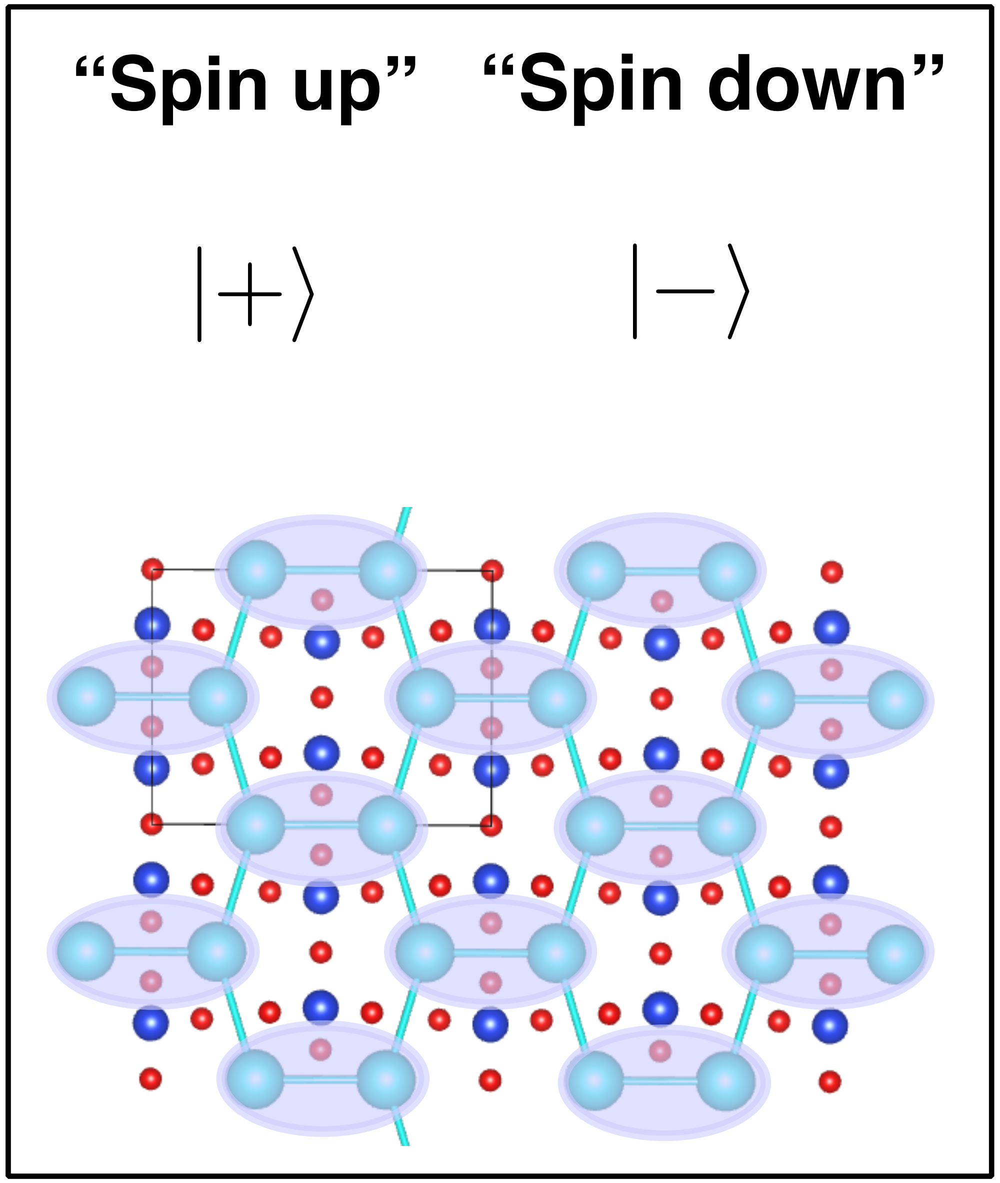
Kate A. Ross
Colorado State University

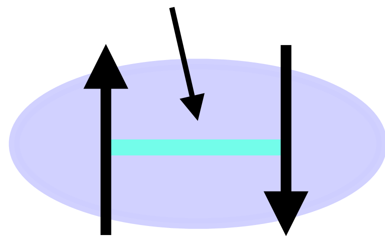




Outline

- Quantum Dimer Magnets and BEC
- Yb^{3+} as an S_{eff} = 1/2 quantum ion
- $Yb_2Si_2O_7$: A new QDM
    - Thermodynamics
    - Inelastic Neutron Scattering
- Open questions: "Mystery Field"





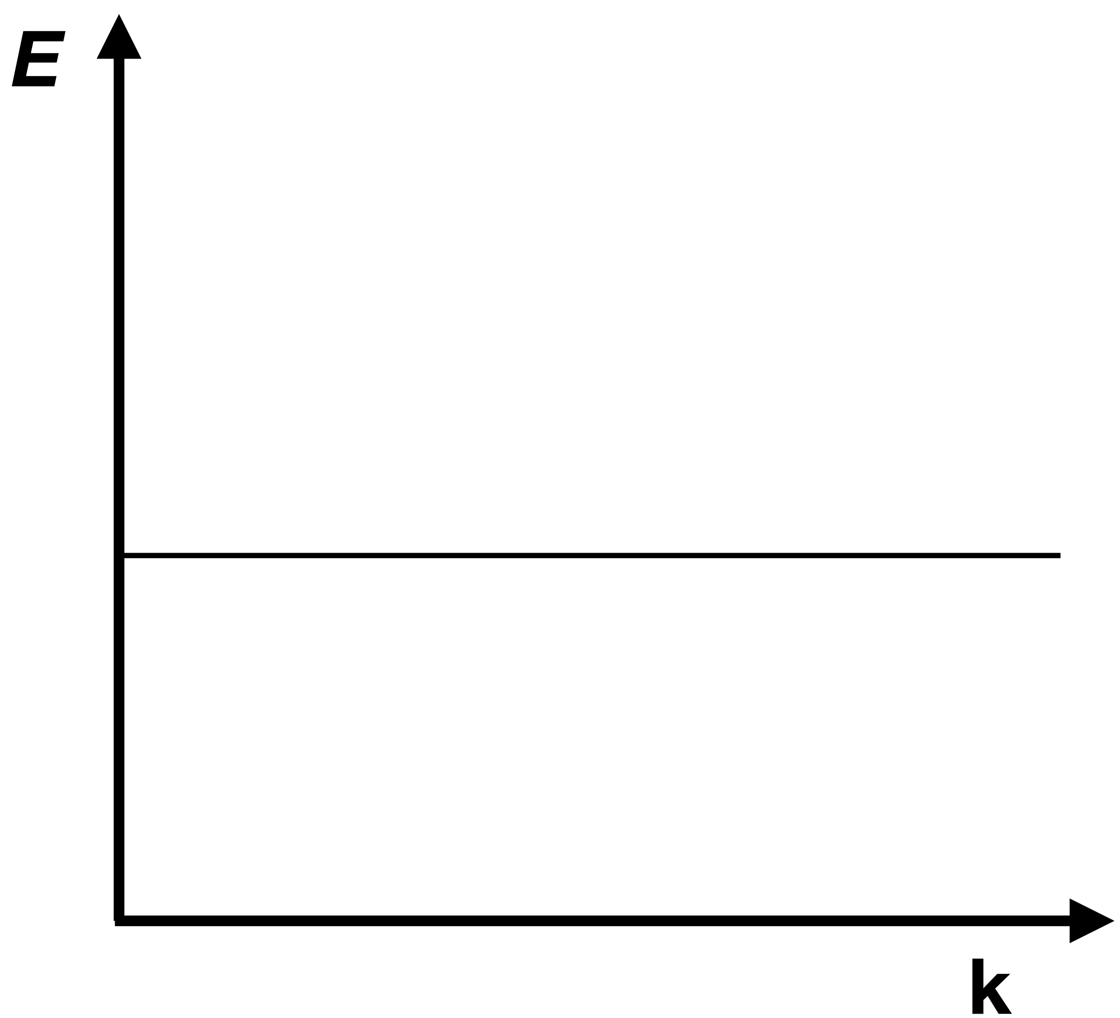
$|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$ $S_{tot} = 0$ Ground state

$|\uparrow\uparrow\rangle$

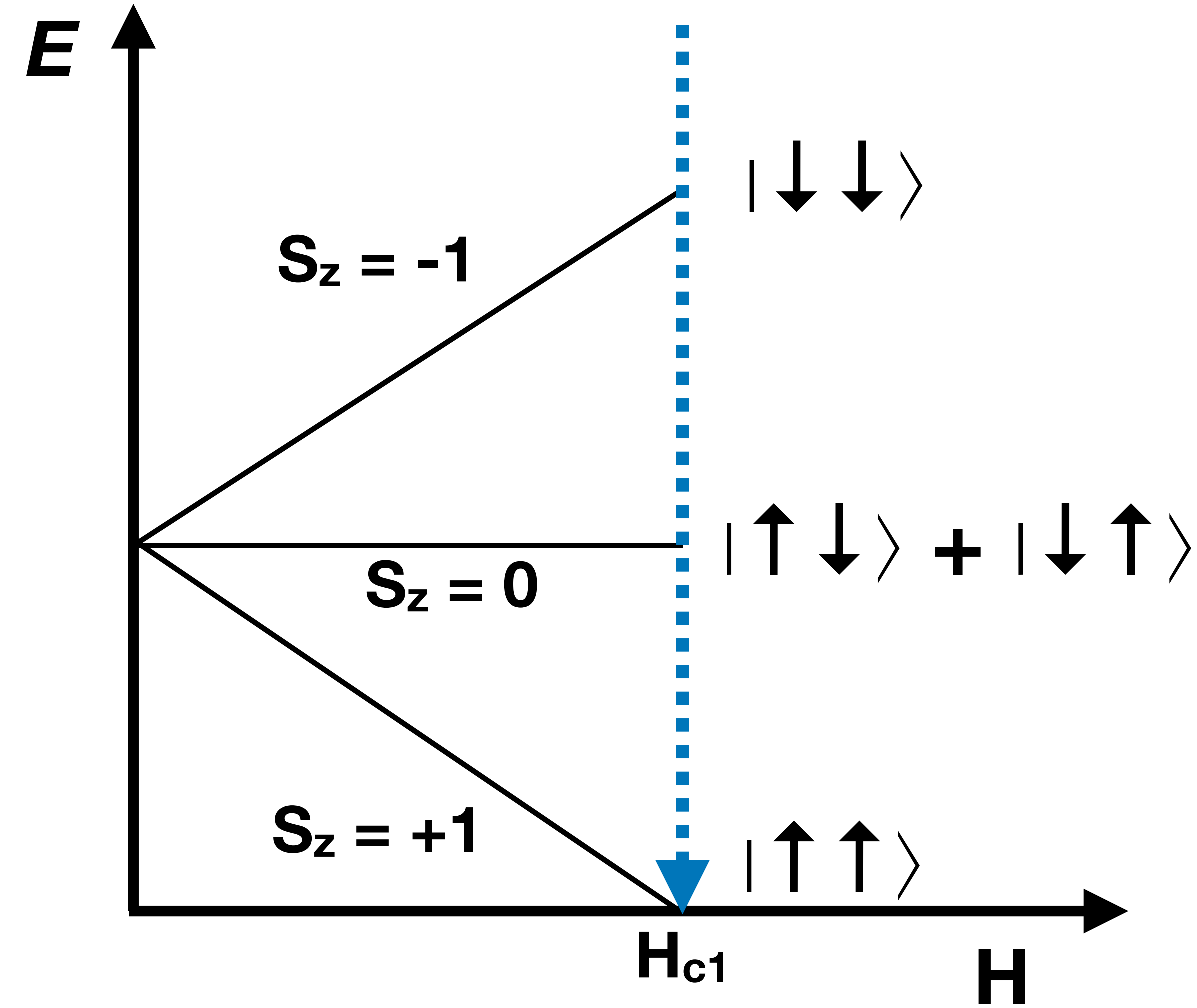
$|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle$ $S_{tot} = 1$ Excited Triplet states

$|\downarrow\downarrow\rangle$

Triplet mode for isolated dimer is flat in momentum space

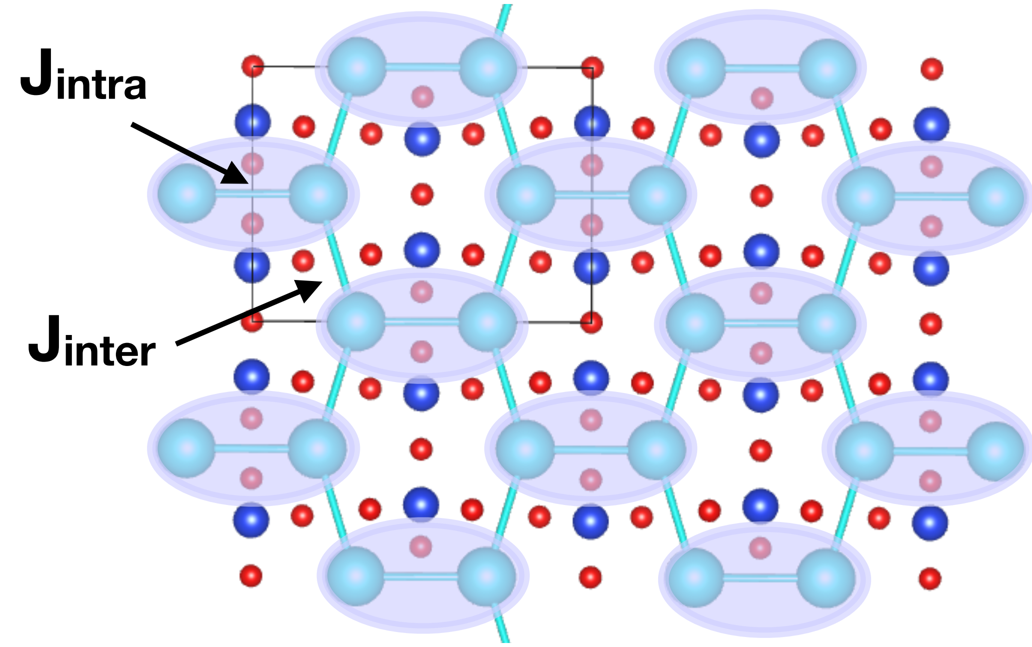


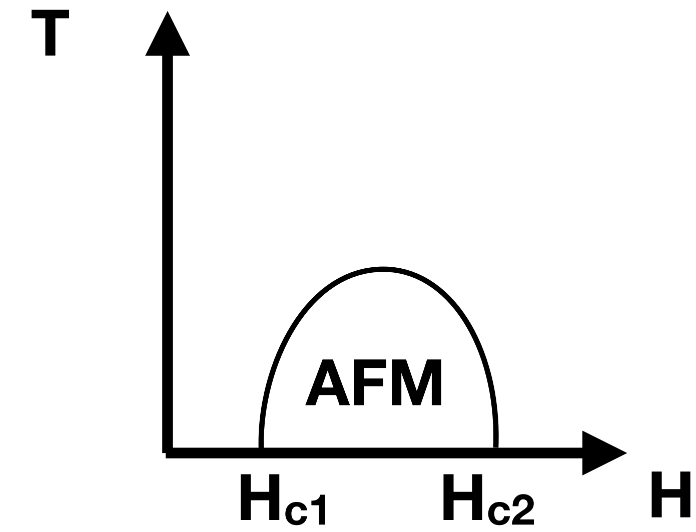
Triplet mode splits in a magnetic field: Transition occurs when $S_z = +1$ mode crosses the ground state energy



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

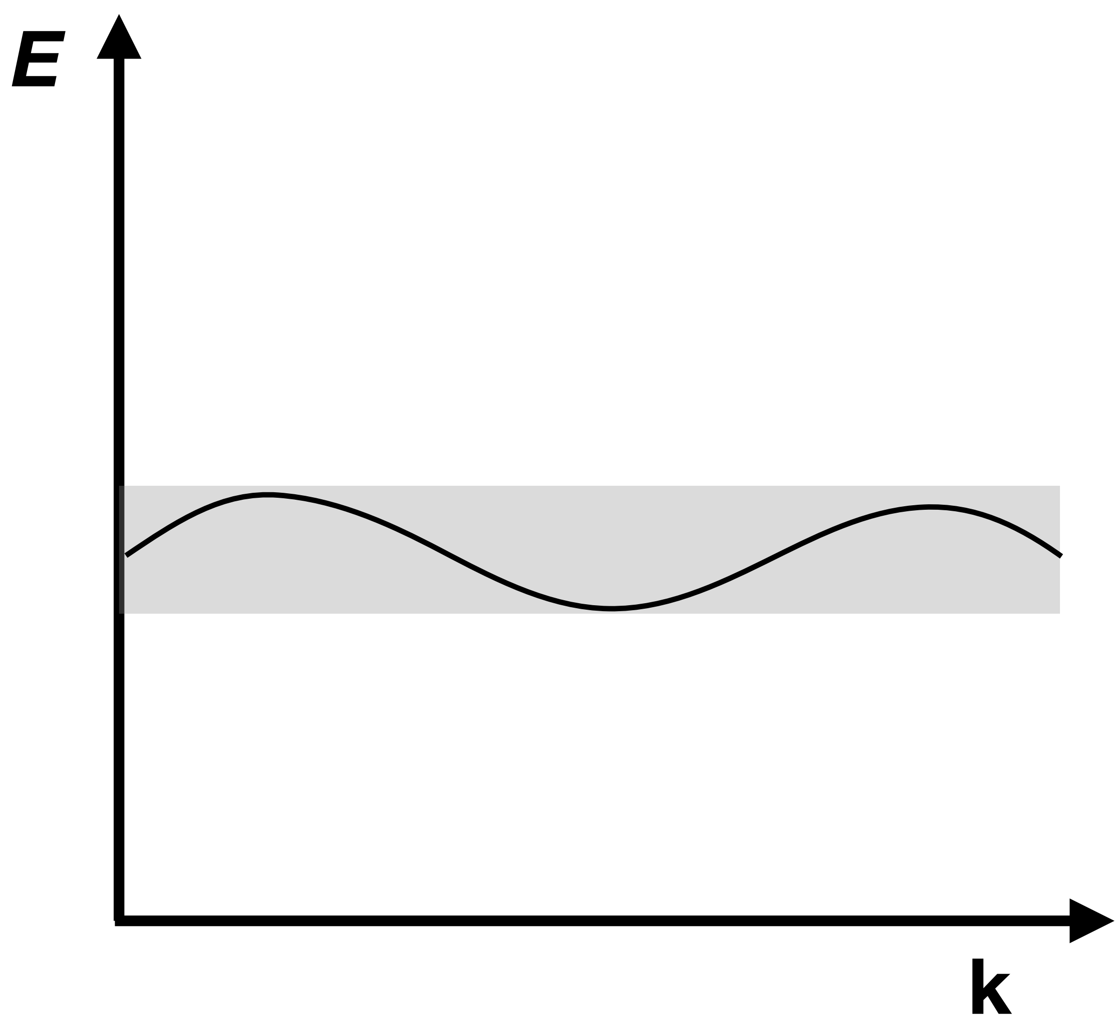
$S_{tot} = 0$



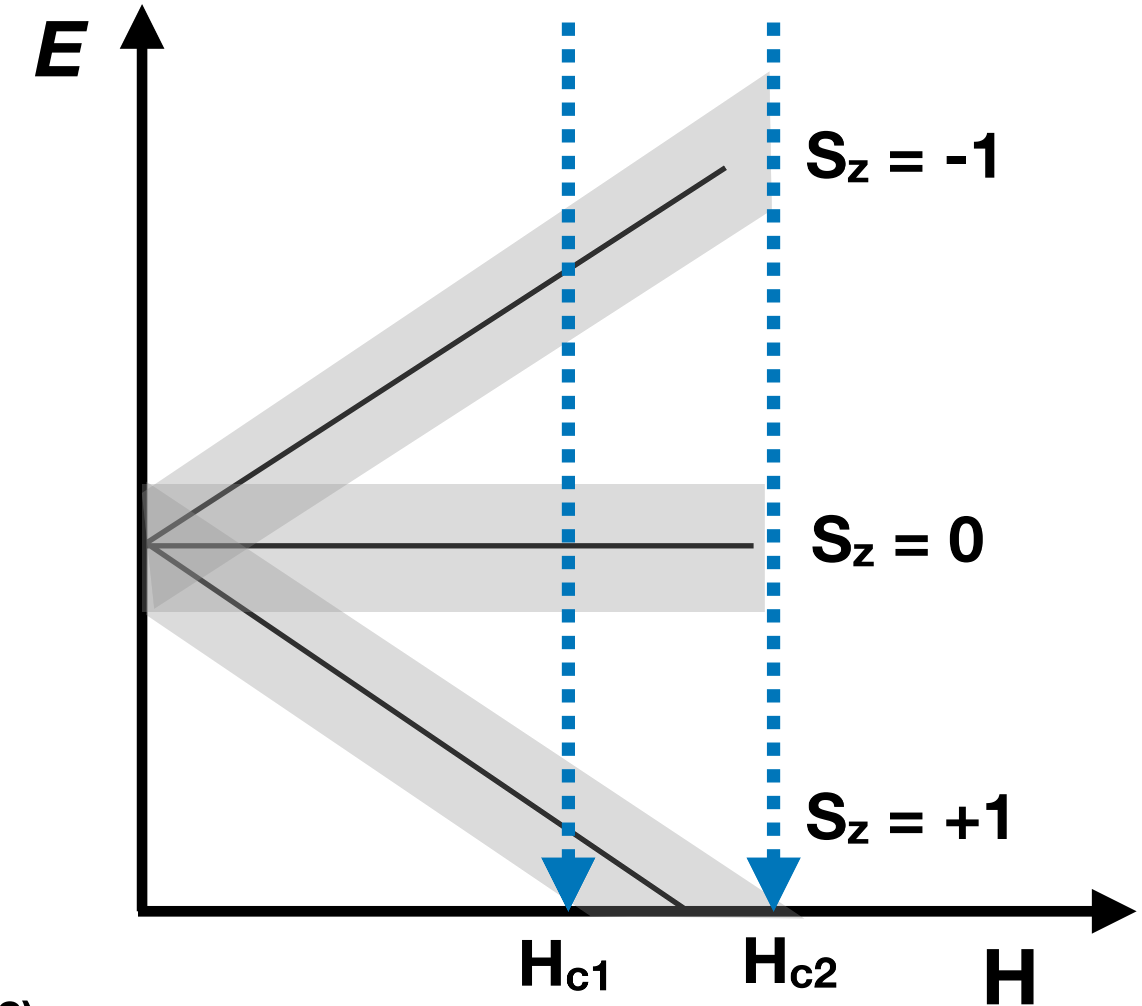


$J_{inter} \neq 0$

Triplet modes gain dispersion: "Triplons"



Transitions at crossing of lower and upper part of Triplon bands



T. Giamarchi, C. Rüegg, O. Tchernyshyov, Nat. Phys 4, 198–204 (2008)

U(1) symmetry and BEC

Matsubara and Matsuda transformation (XXZ Hamiltonian - valid near H_{c1} and H_{c2})

$$\mathcal{H}_{XXZ} = J\sum_{\mathbf{r},\nu}(S^x_{\mathbf{r}}S^x_{\mathbf{r}+\mathbf{e}_\nu} + S^y_{\mathbf{r}}S^y_{\mathbf{r}+\mathbf{e}_\nu} + \gamma S^z_{\mathbf{r}}S^z_{\mathbf{r}+\mathbf{e}_\nu}) - g_{zz}\mu_B H\sum_{\mathbf{r}} S^z_{\mathbf{r}}.$$

$$\boxed{S^+_j = b^\dagger_j, \qquad S^-_j = b_j, \qquad S^z_j = b^\dagger_j b_j - \tfrac{1}{2}.}$$

Zapf, Jaime, Batista, Rev. Mod. Phys. 86 (2014)

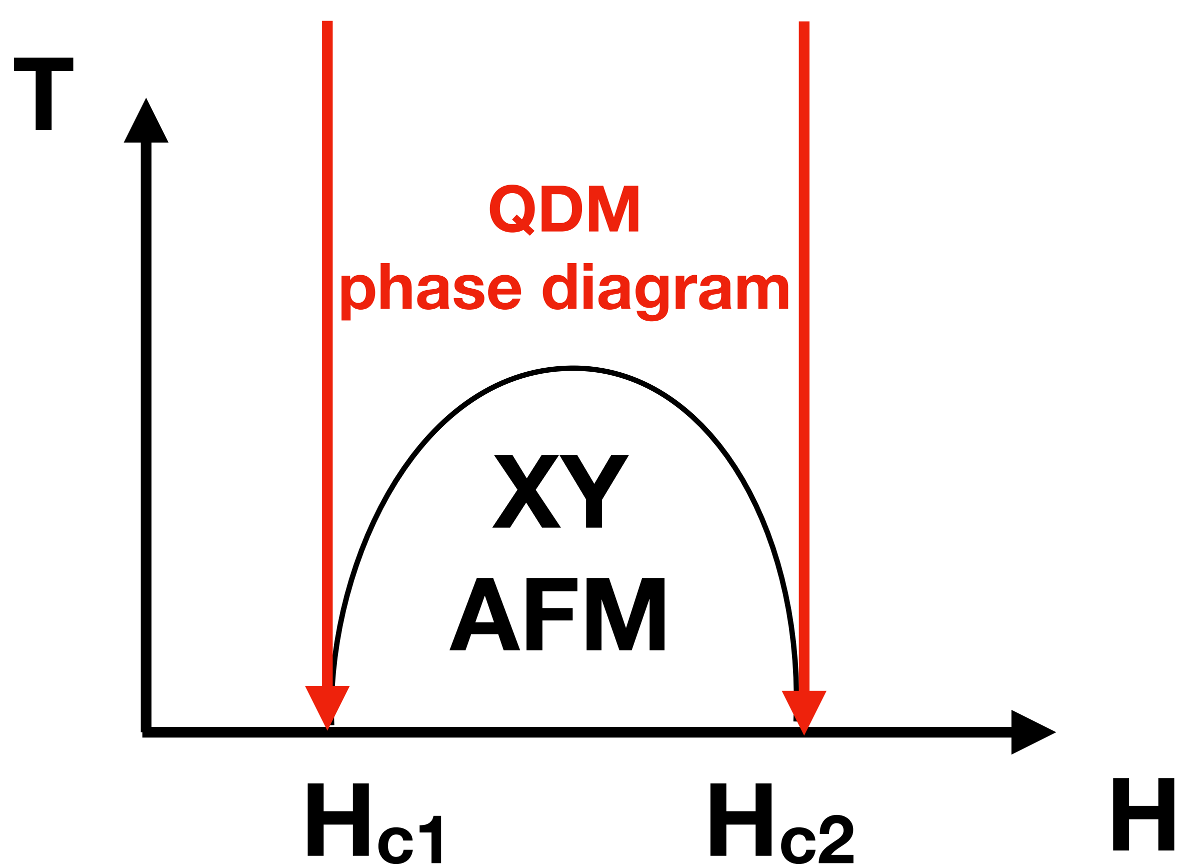
U(1) symmetry and BEC

Matsubara and Matsuda transformation (XXZ Hamiltonian - valid near H_{c1} and H_{c2})

$$\mathcal{H}_{XXZ} = J\sum_{\mathbf{r},\nu}(S^x_{\mathbf{r}}S^x_{\mathbf{r}+\mathbf{e}_\nu} + S^y_{\mathbf{r}}S^y_{\mathbf{r}+\mathbf{e}_\nu} + \gamma S^z_{\mathbf{r}}S^z_{\mathbf{r}+\mathbf{e}_\nu}) - g_{zz}\mu_B H\sum_{\mathbf{r}} S^z_{\mathbf{r}}.$$

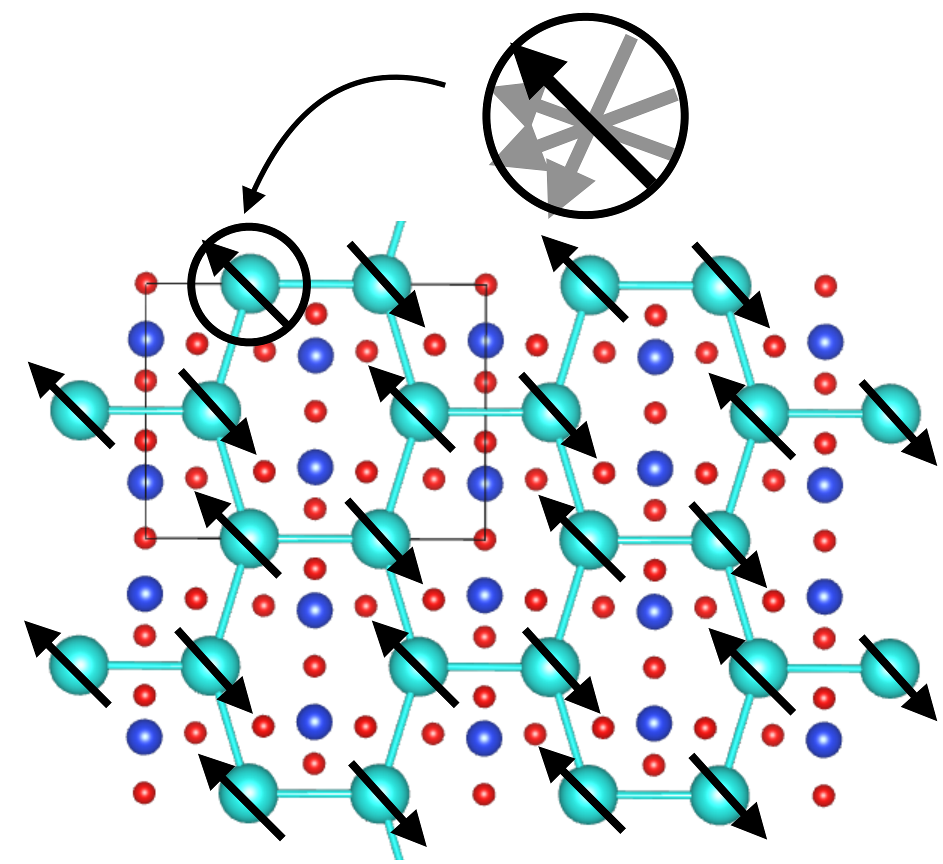
$$\mathcal{H}_{XXZ} = \frac{J}{2}\sum_{\mathbf{r},\nu}(b^\dagger_{\mathbf{r}}b_{\mathbf{r}+\mathbf{e}_\nu} + b^\dagger_{\mathbf{r}+\mathbf{e}_\nu}b_{\mathbf{r}}) + \gamma J\sum_{\mathbf{r}}(n^b_{\mathbf{r}} - 1/2)(n^b_{\mathbf{r}+\mathbf{e}_\nu} - 1/2) - \mu\sum_{\mathbf{r}} n^b_{\mathbf{r}}.$$

XY AFM transition corresponding to BEC transition in boson language



T. Giamarchi, A.M. Tsvelik, PRB 59 (1999)

Spontaneously breaks U(1) symmetry in XY plane

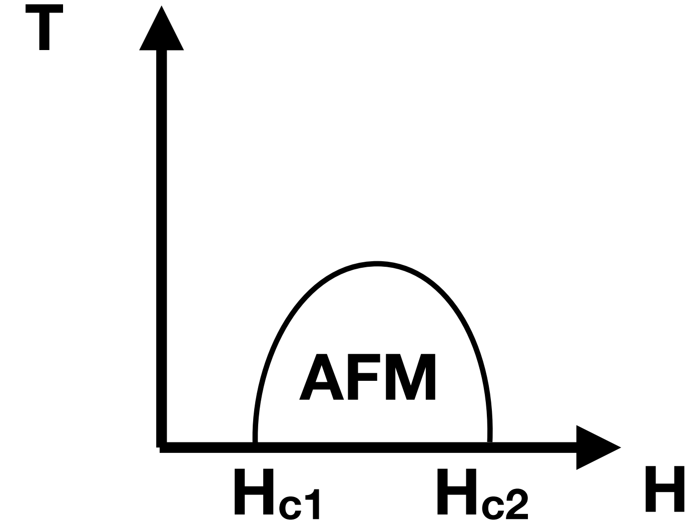


Zapf, Jaime, Batista, Rev. Mod. Phys. 86 (2014)

Gavin Hester

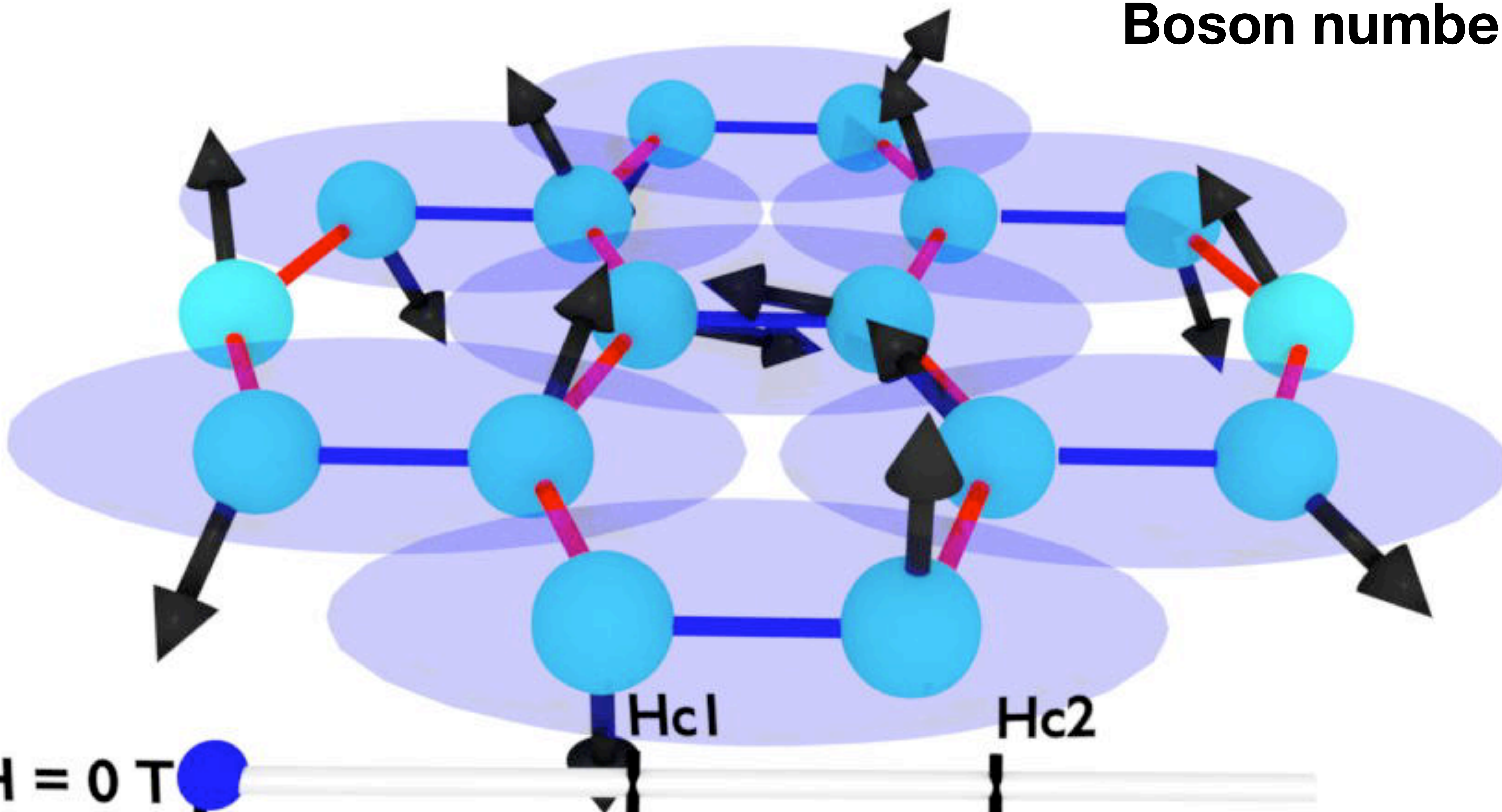
$= |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$

$S_{tot} = 0$



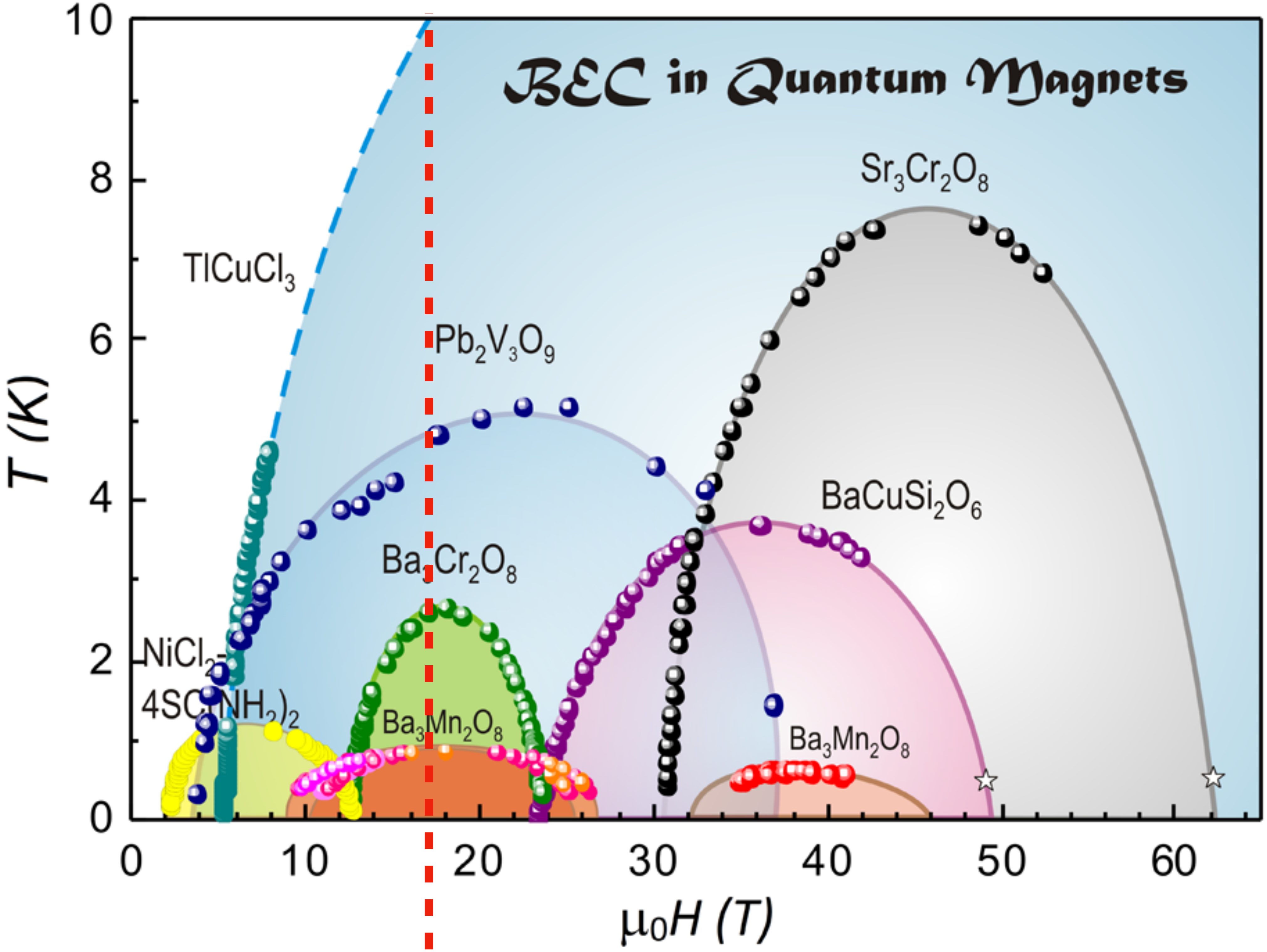
Quantum Paramagnet

Boson number = |Sz|



BEC in Quantum magnets

Zapf, Jaime, Batista, Rev. Mod. Phys. 86 (2014)

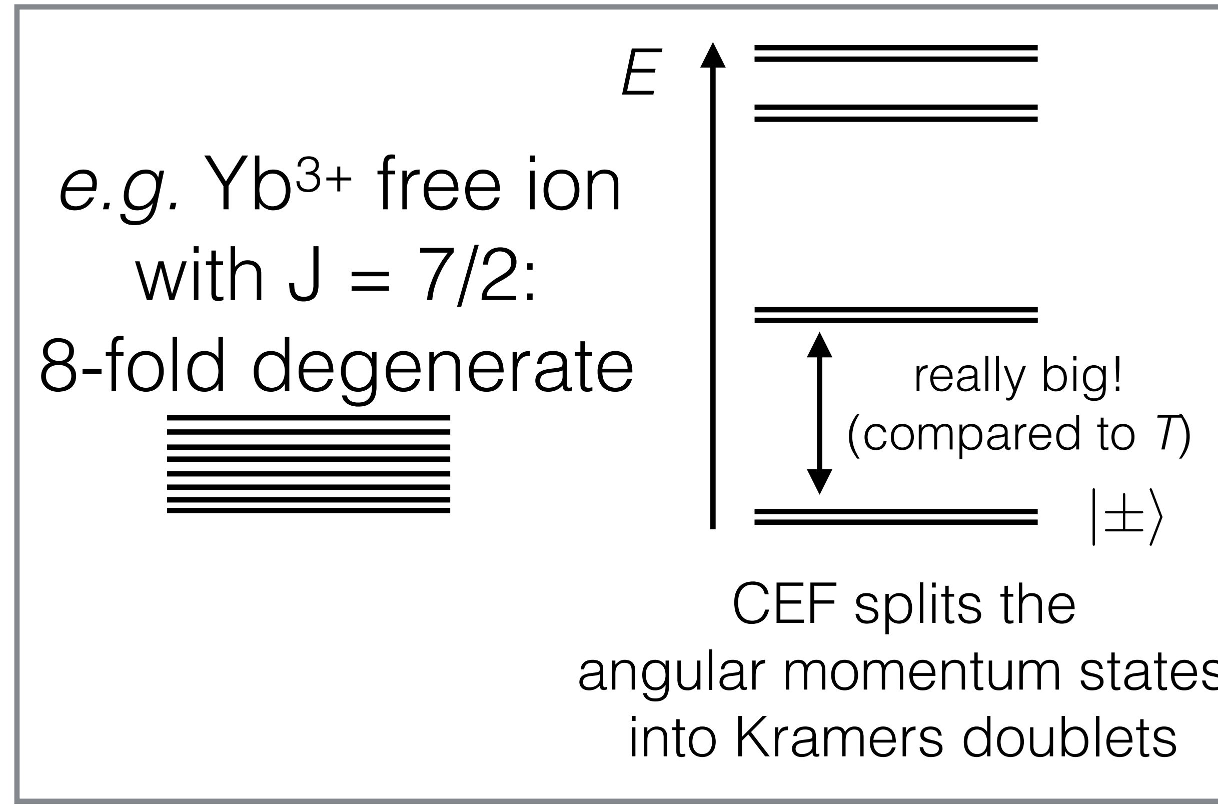


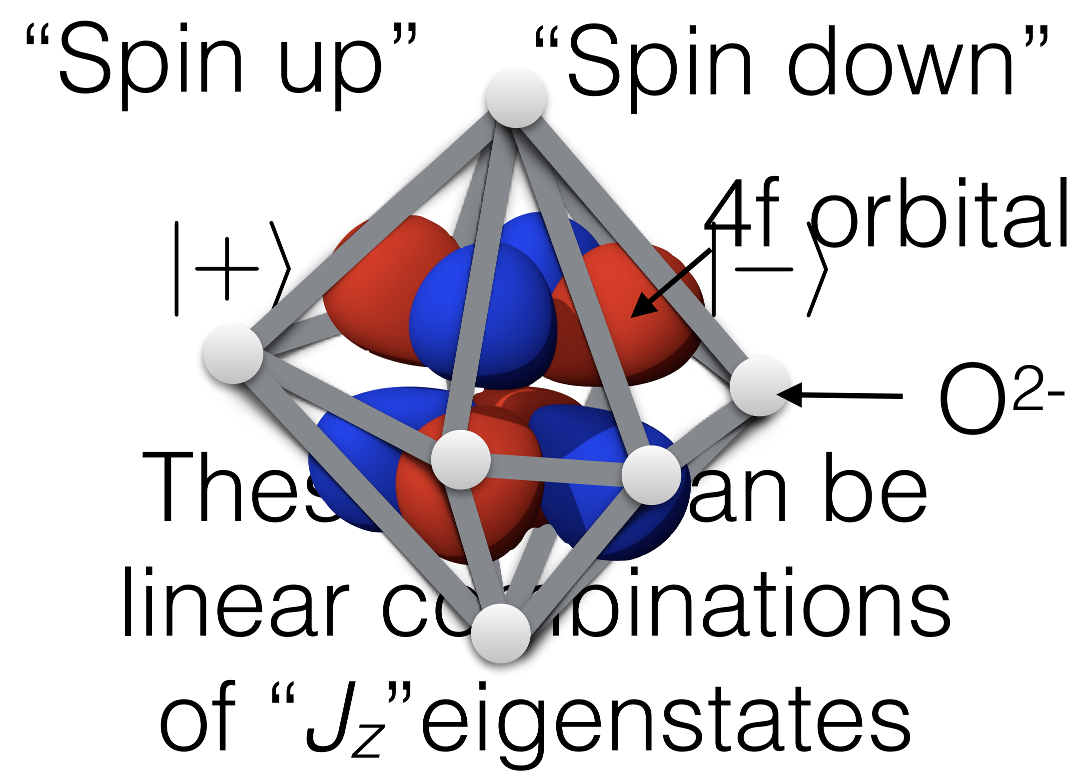
superconducting magnet max (neutron scattering)

$Yb_2Si_2O_7$

Quantum spins from large angular momentum ions

- Crystal electric field plus spin orbit coupling
- *“well isolated”* **doublets** (compared to interaction energy scale)**. These doublets can be mapped onto spin 1/2 formalism**
- However, depending on coupling between larger J moments, **Exchange Hamiltonian can in principle become highly *anisotropic***
  - e.g. Pyrochlore “Quantum Spin ice”

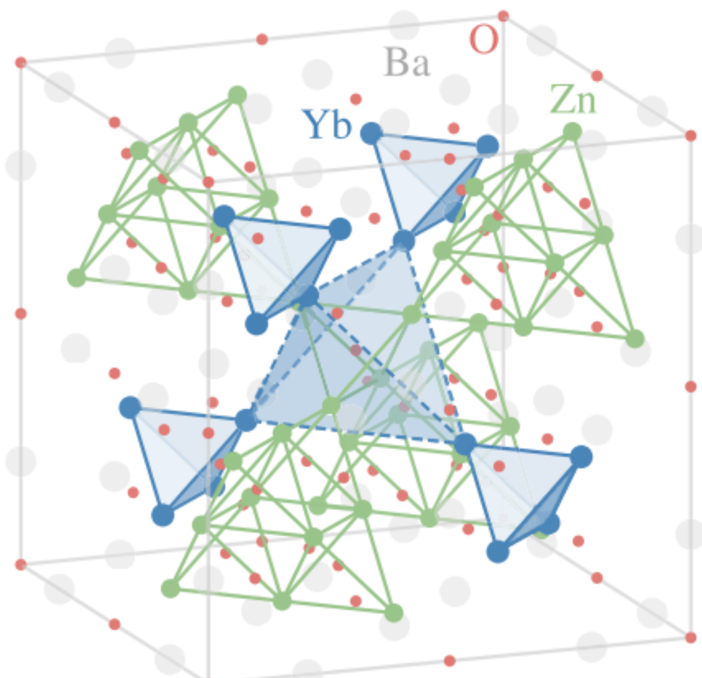




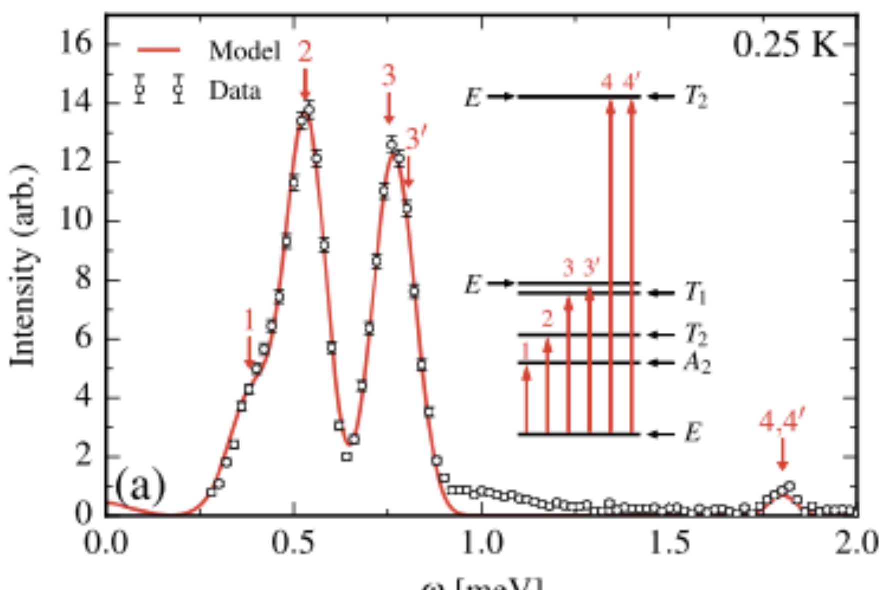
Yb^{3+} quantum magnets: "Ytterbium is the new Copper"

$BaYb_2Zn_5O_{11}$

quantum tetramers



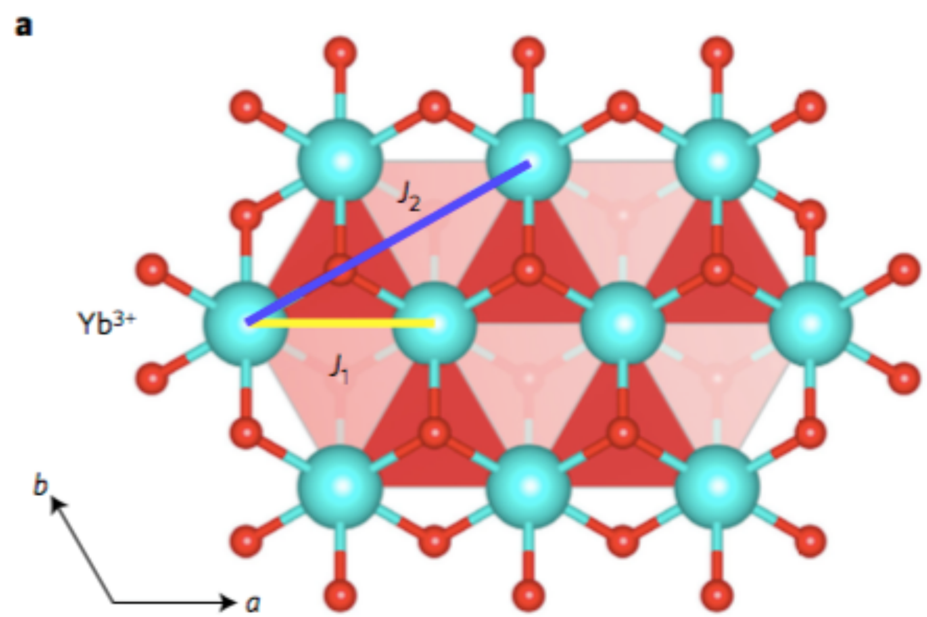
Rau et al, PRL 116 257204 (2016)



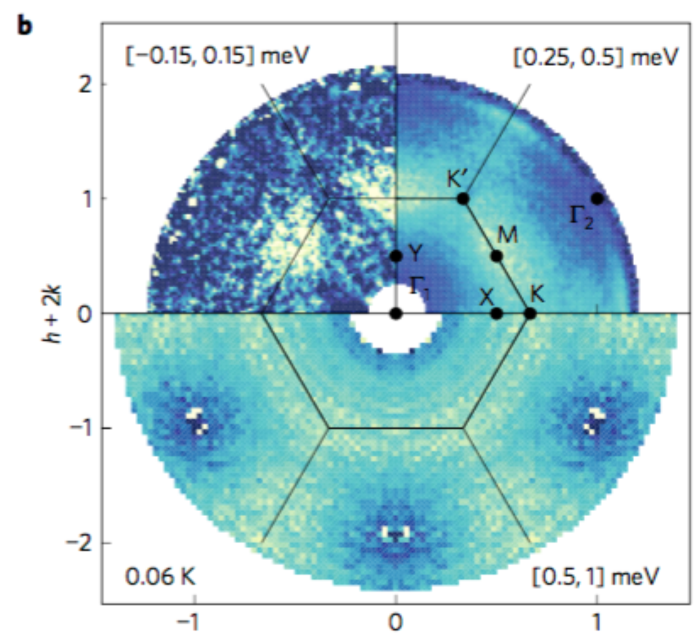
$YbMgGaO_4$

QSL?

Random Valence Bonds?



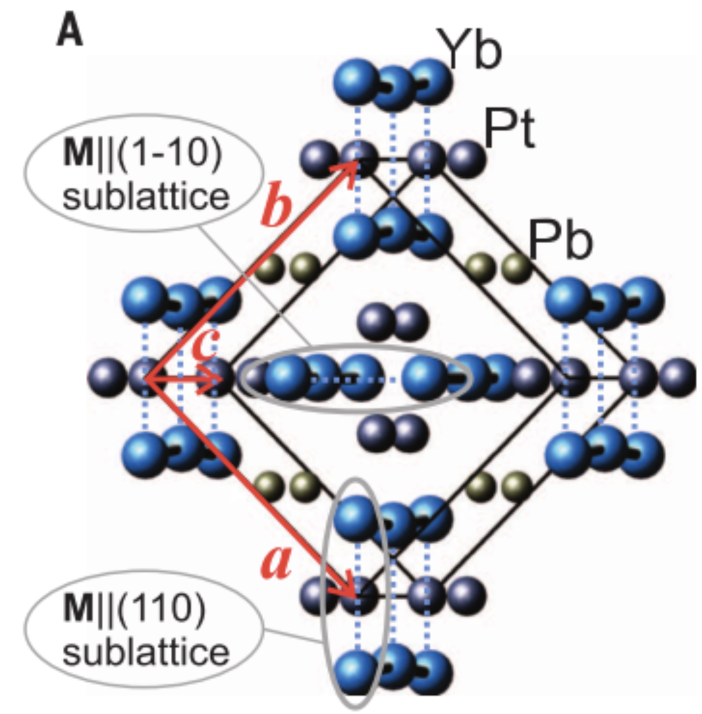
Paddison et al, Nat. Phys, (2017)



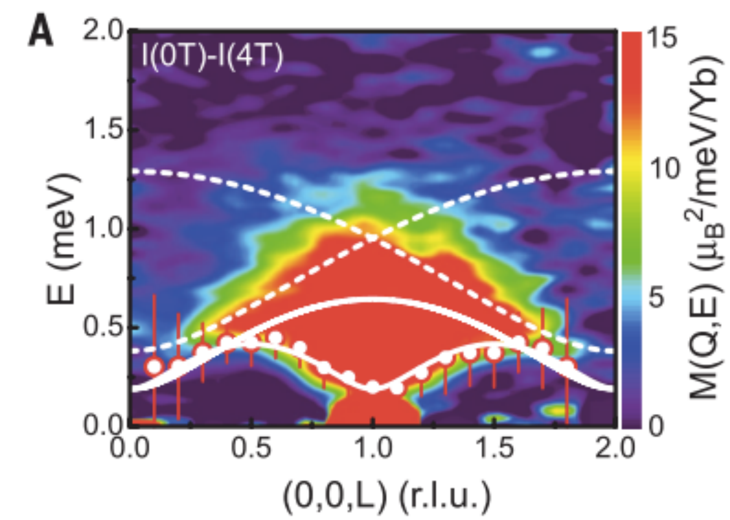
Yb_2Pt_2Pb

XXZ Chains

Spinons

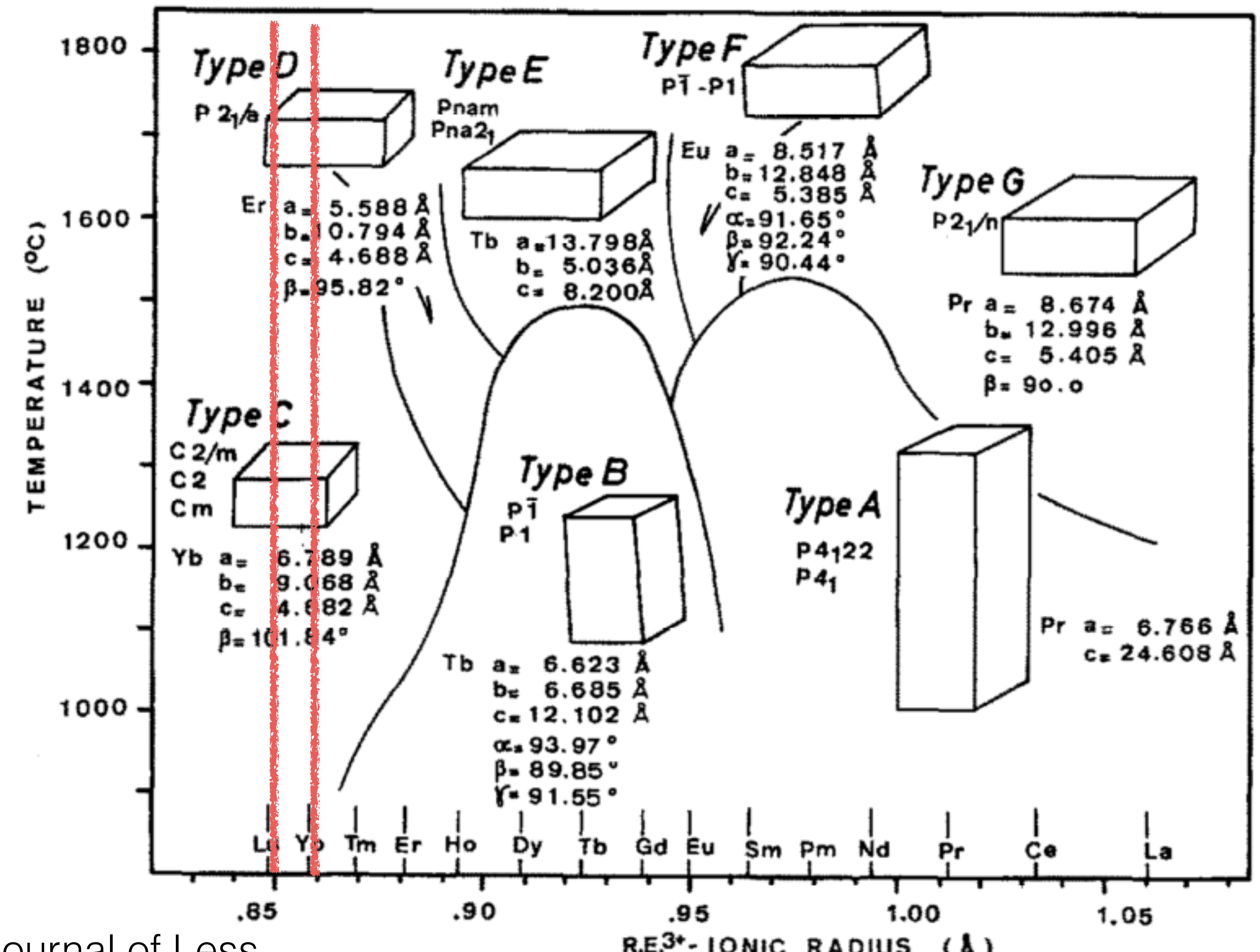


Wu et al, Science, 352 (2016)



Honeycomb compounds with Seff = 1/2

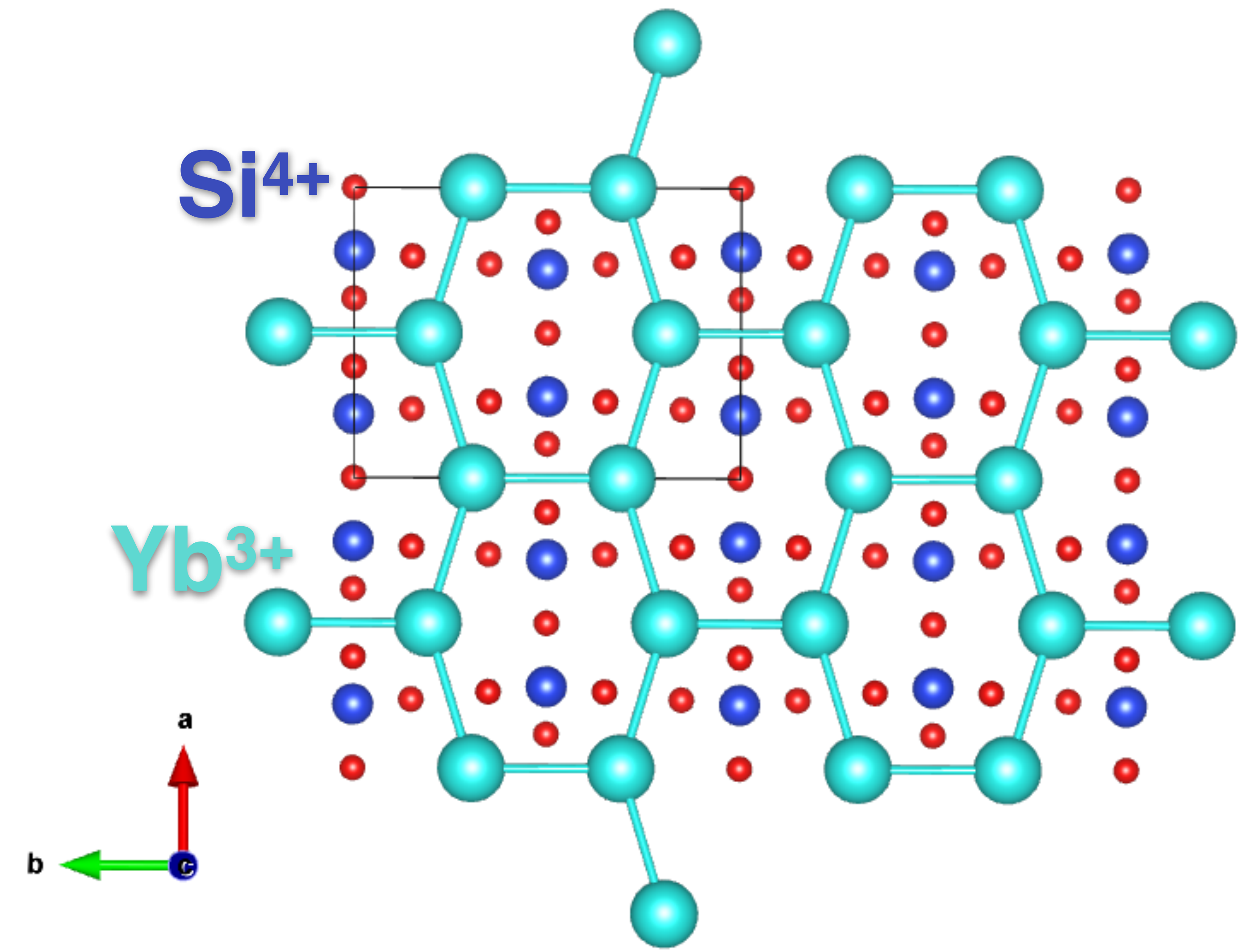
- A potentially useful series: $R_2Si_2O_7$



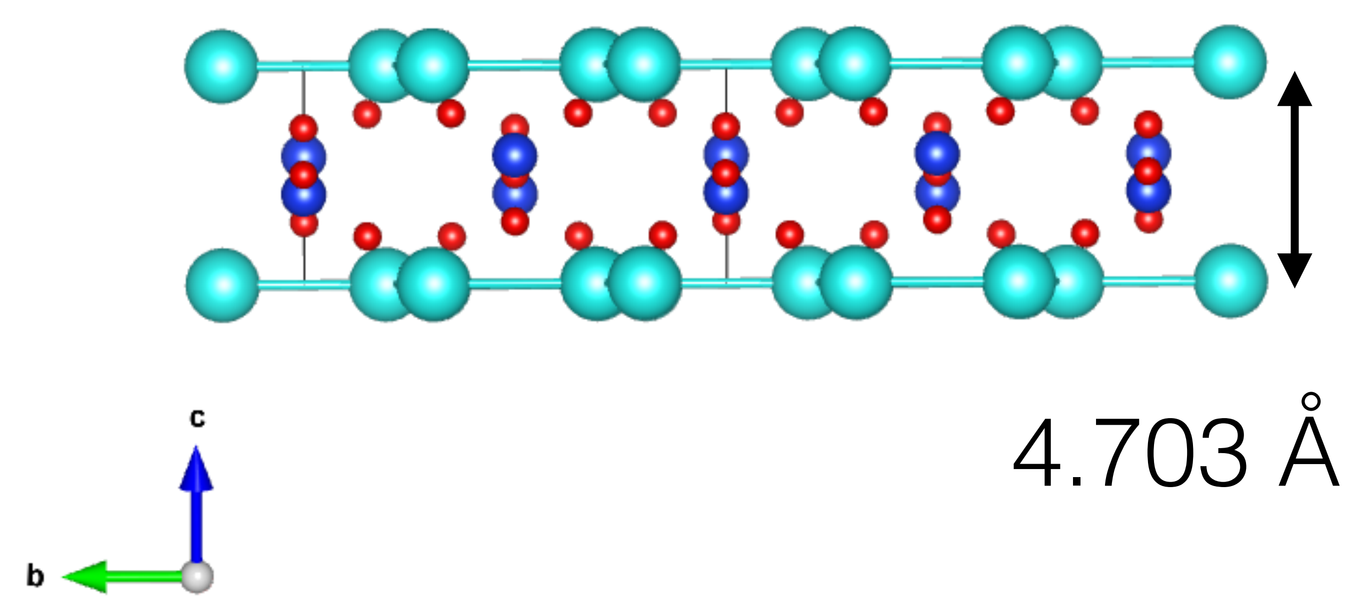
J. Felsche, Journal of Less-Common Metals, 21 (1970)

$Yb_2Si_2O_7$

C2/m
a = 6.80 Å
b = 8.87 Å
c = 4.70 Å
β = 102.12

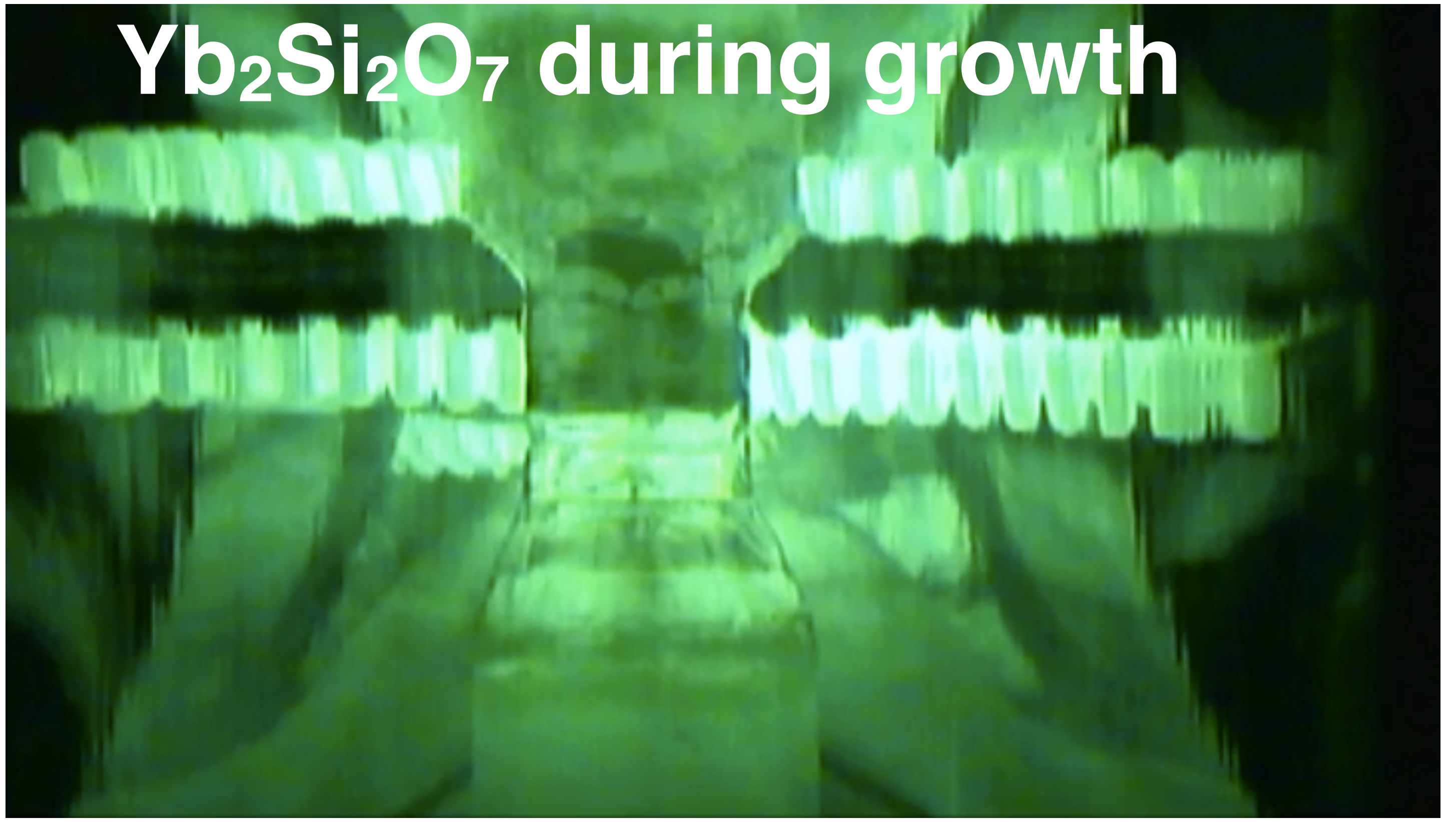


"honeycomb" lattice compressed along *b*

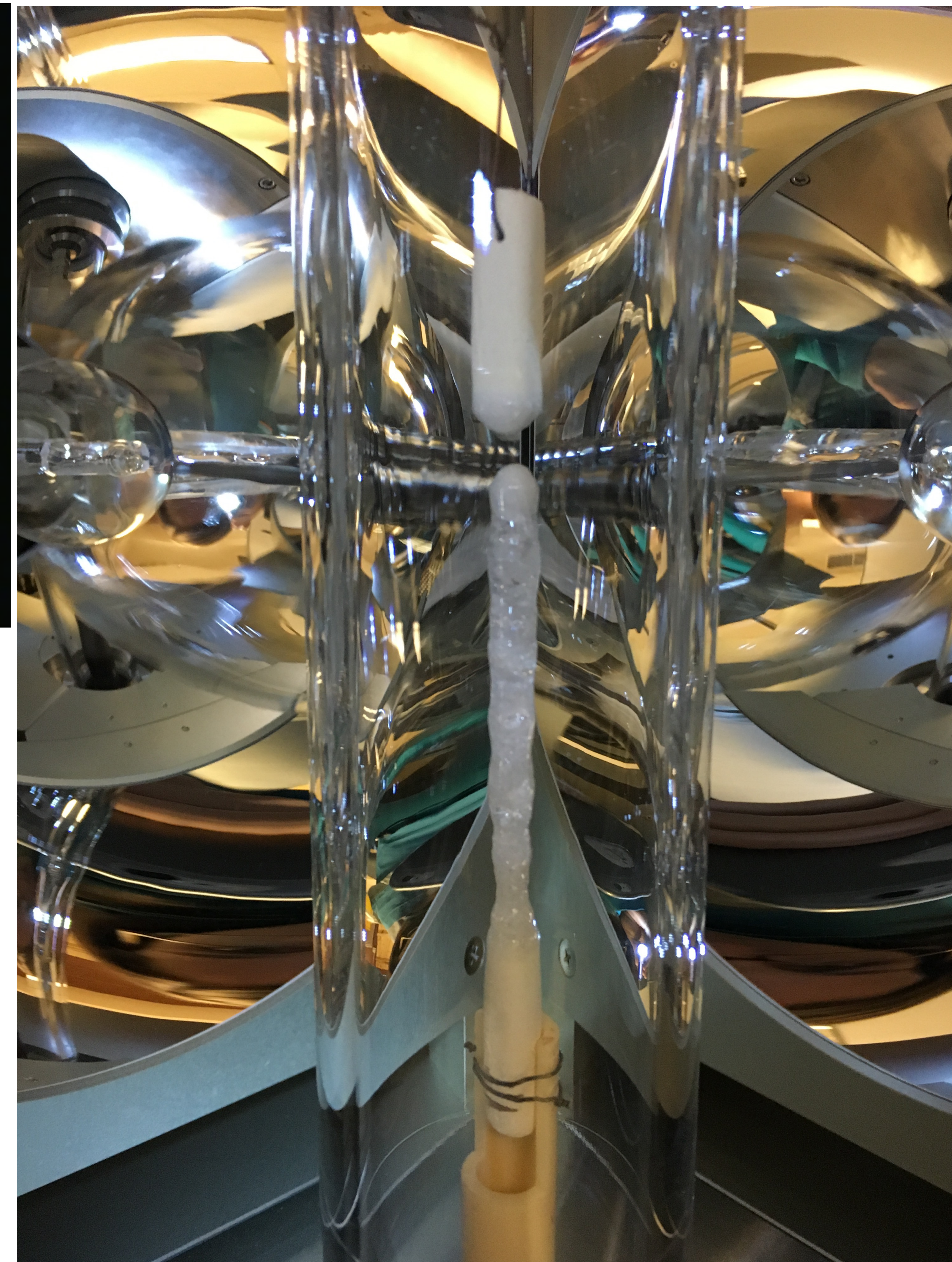


3.4% difference for in-plane Yb-Yb distances:
3.428 Å
3.548 Å

$Yb_2Si_2O_7$ Crystal Growth



Grown by Dr. Harikrishnan Nair at CSU (now Assistant Prof. at UTEP)

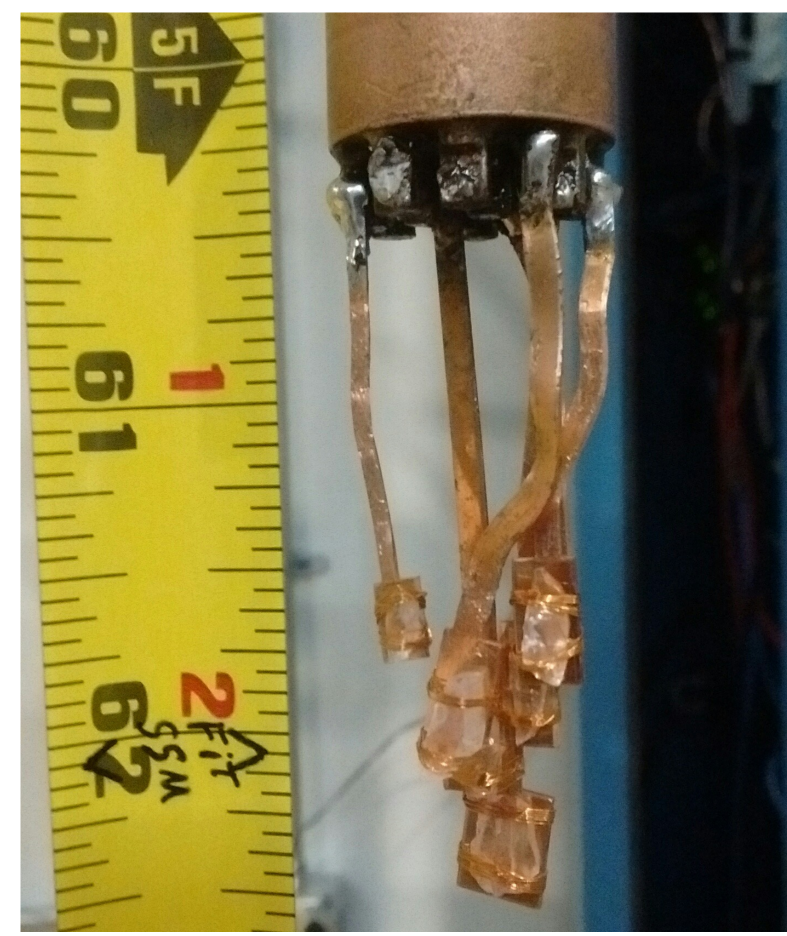


H. S. Nair, T. DeLazzer, T. Reeder, A. Sikorski, G. Hester, and KAR. Crystals 9(4), 196 (2019)

Co-alignment of crystal pieces

cracking of growth leads to ~0.6 cm size crystals

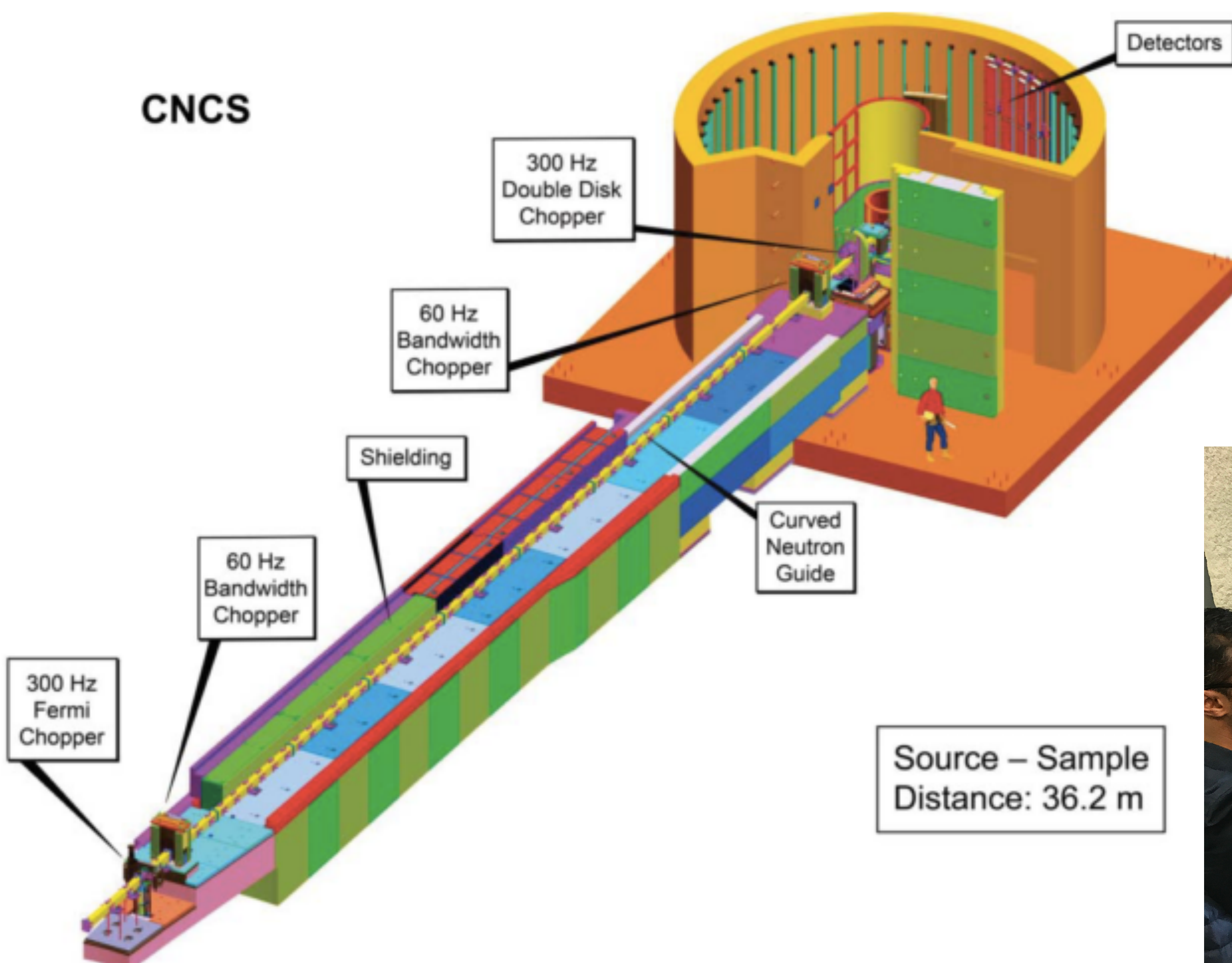
Tim Reeder
(UG researcher, now at JHU): co-alignment of crystal pieces



Neutron scattering: CNCS

Cold Neutron Chopper Spectrometer (CNCS)

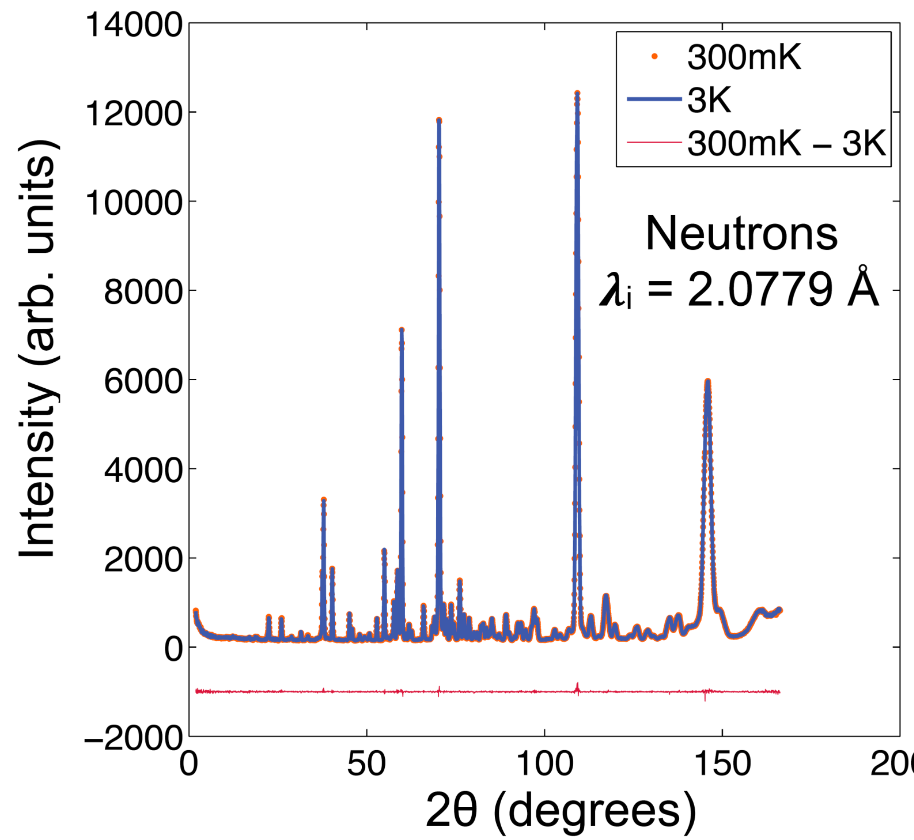
High energy resolution (0.037 meV) - map out elastic and inelastic scattering in a plane of reciprocal space

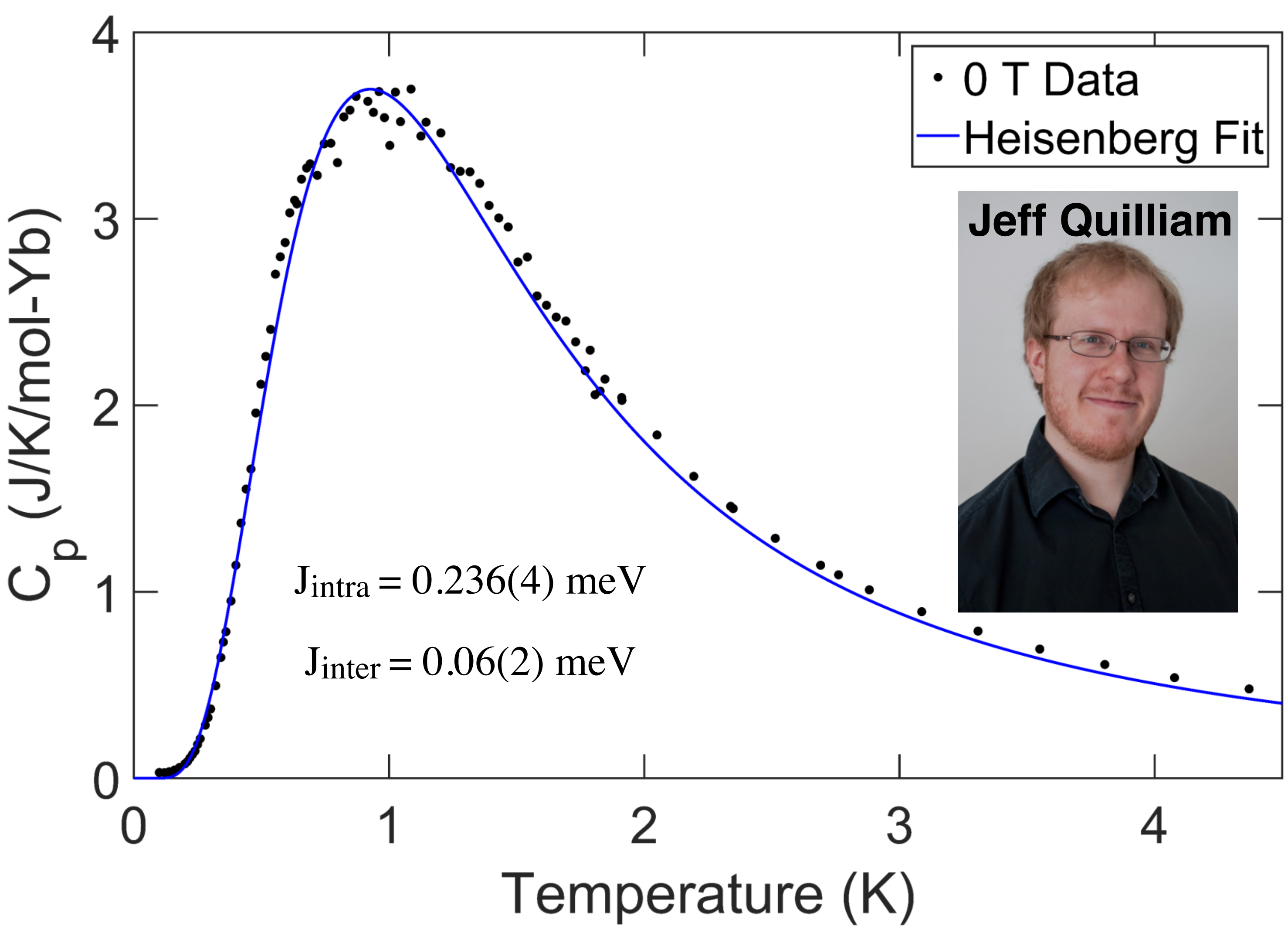


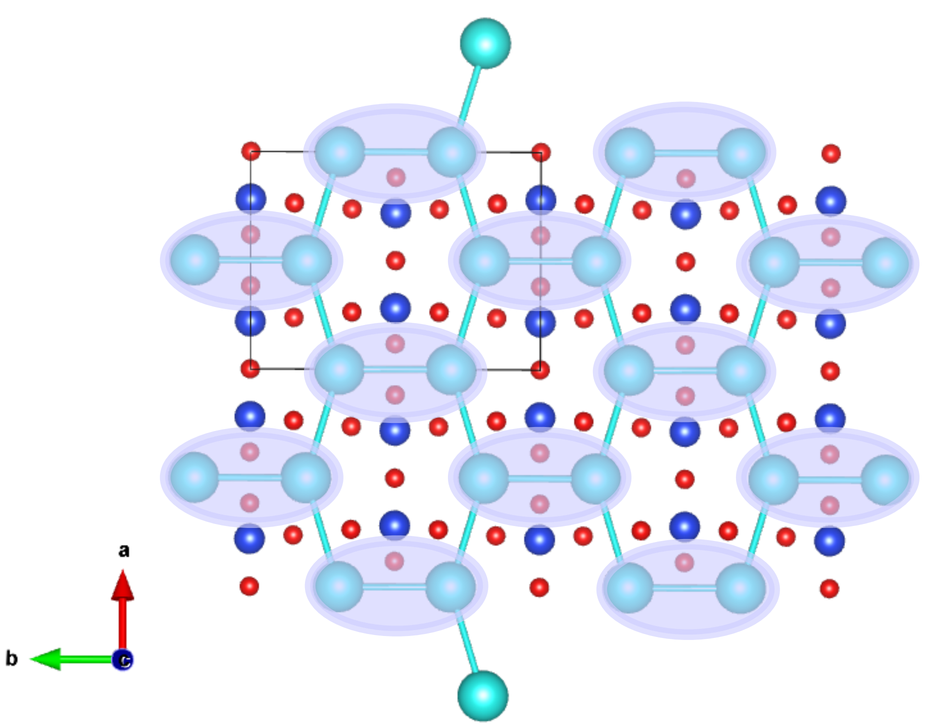
Gavin Hester

$Yb_2Si_2O_7$ Quantum Dimers in Zero Field

- No magnetic Bragg Peaks at 300 mK
- Schottky form of Cp vs. T, releases all *R*ln2/mol Yb
- Fits well to Heisenberg model

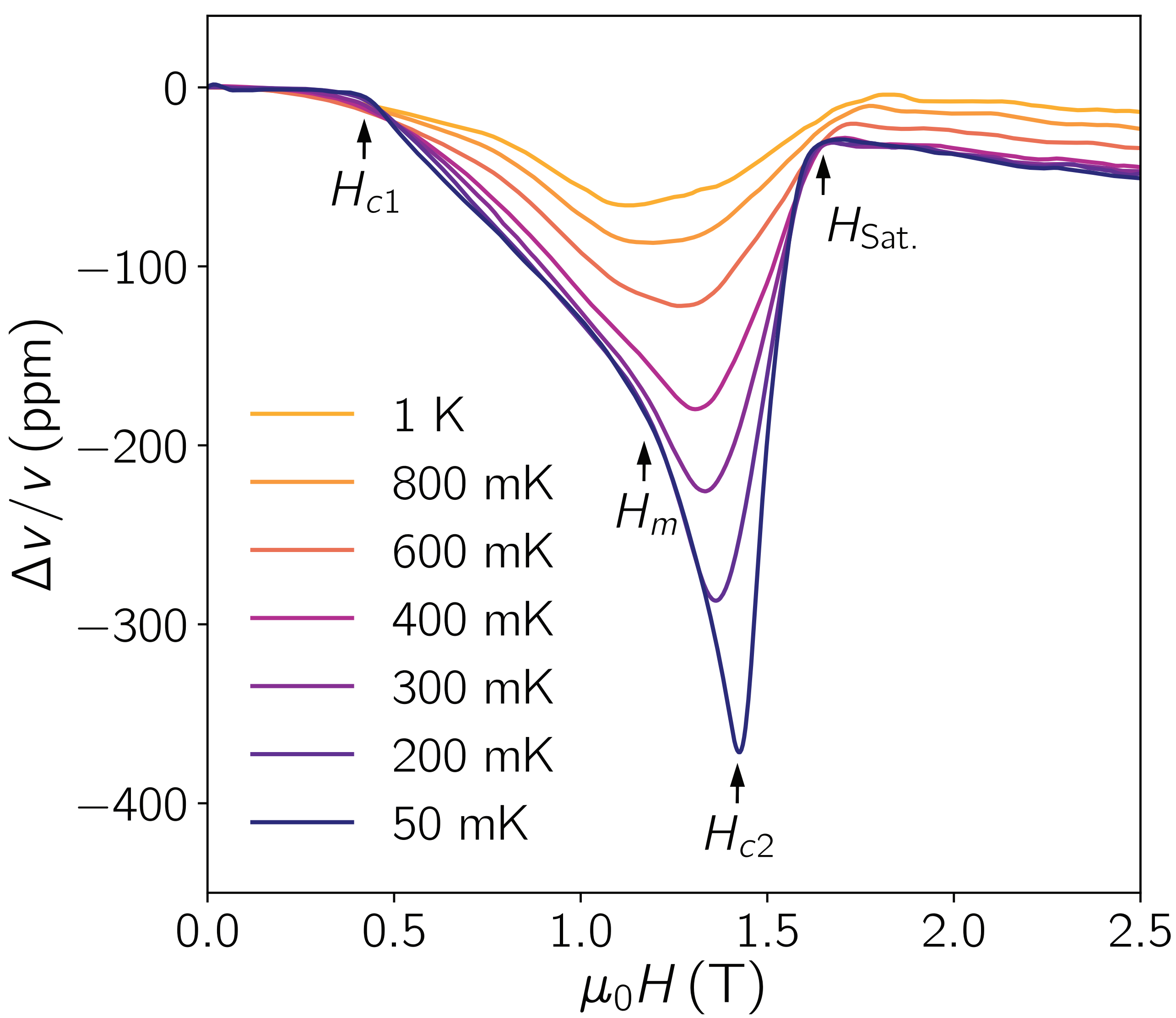




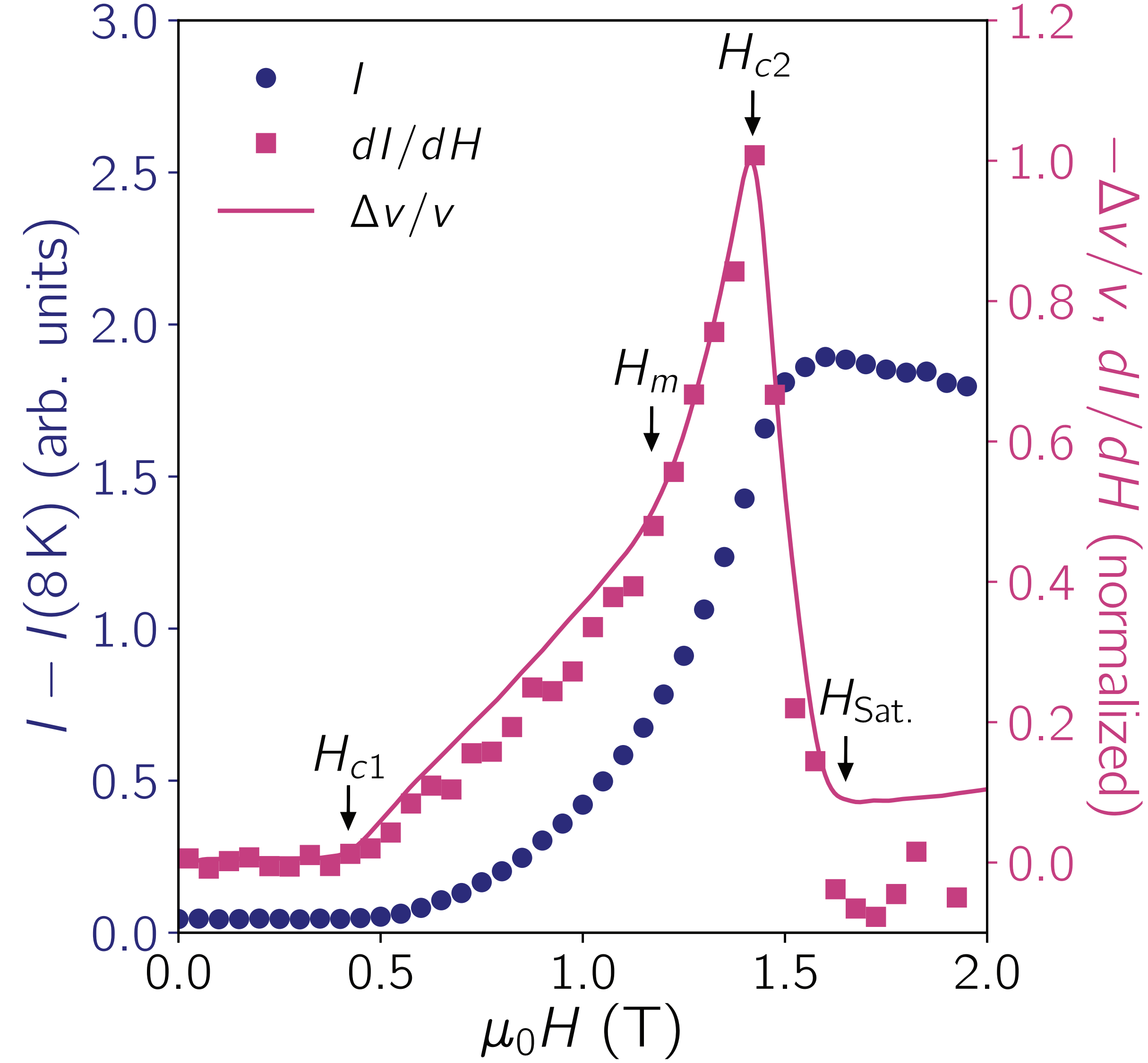


Field Induced Magnetic Order

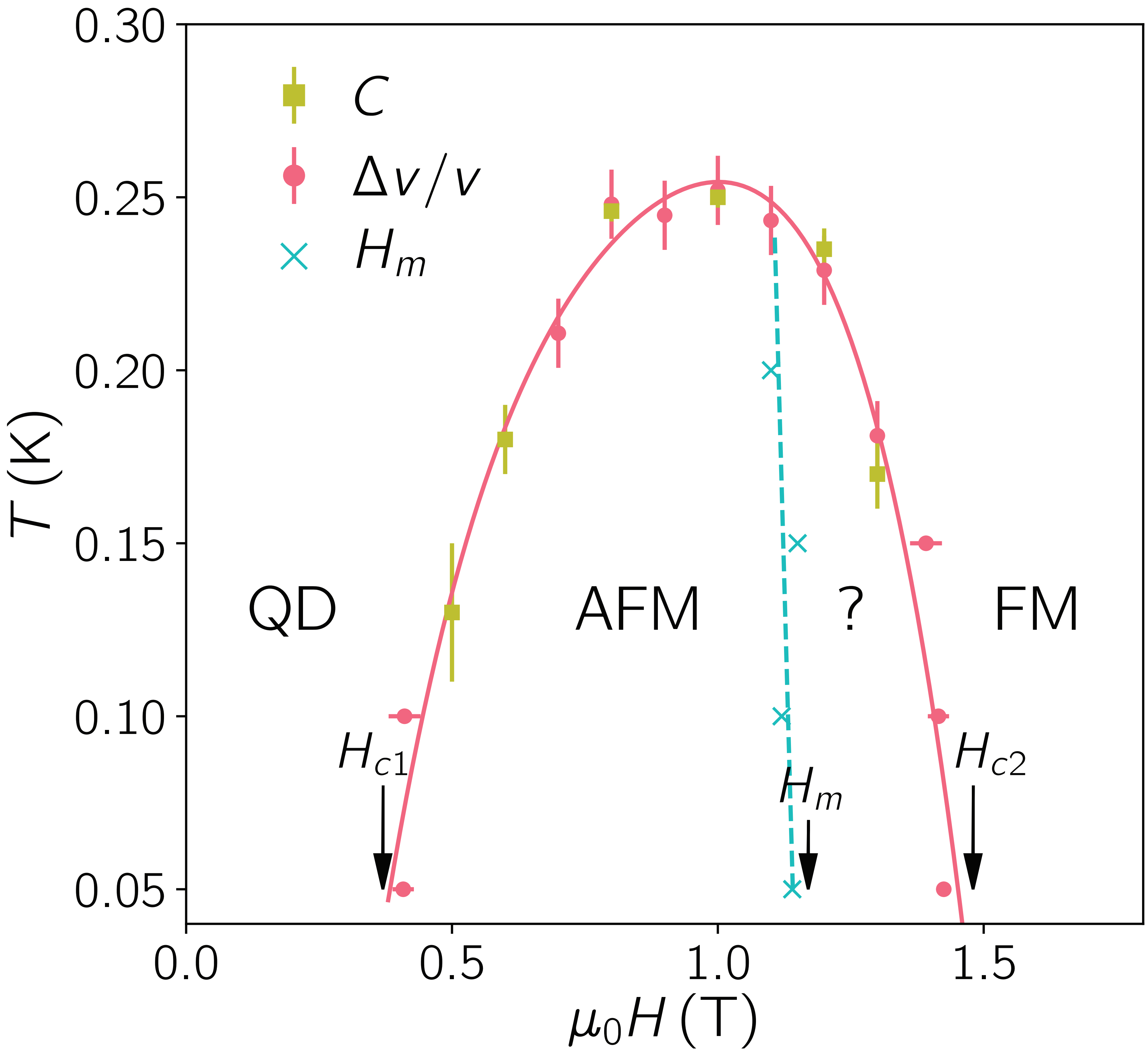
Ultrasound Velocity (Quilliam lab)

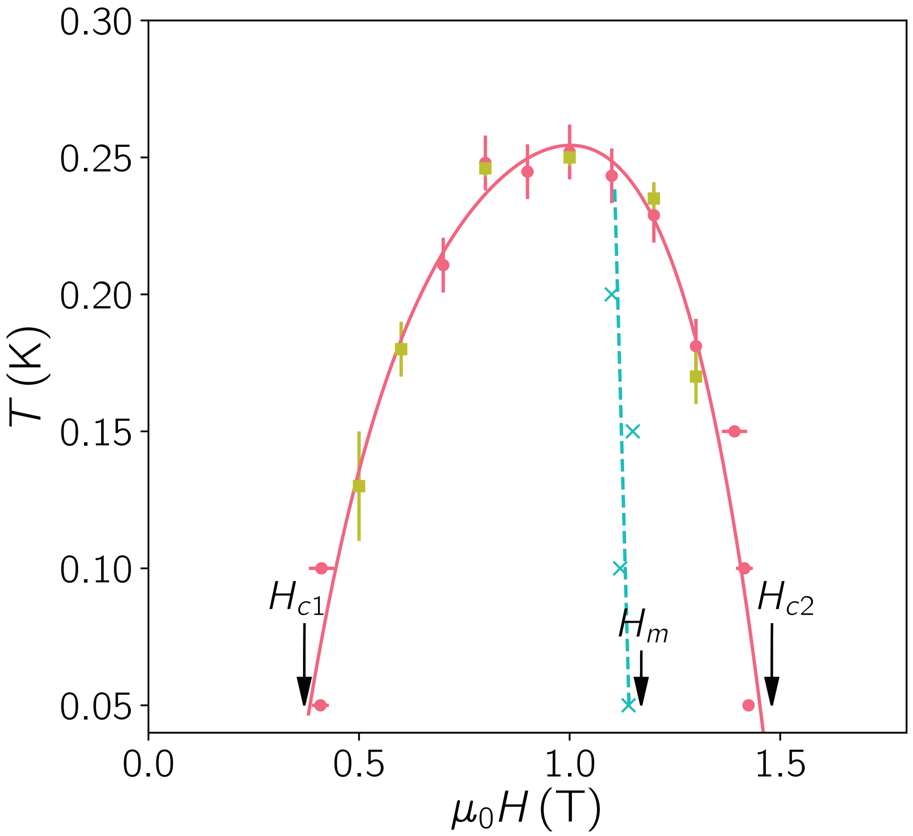


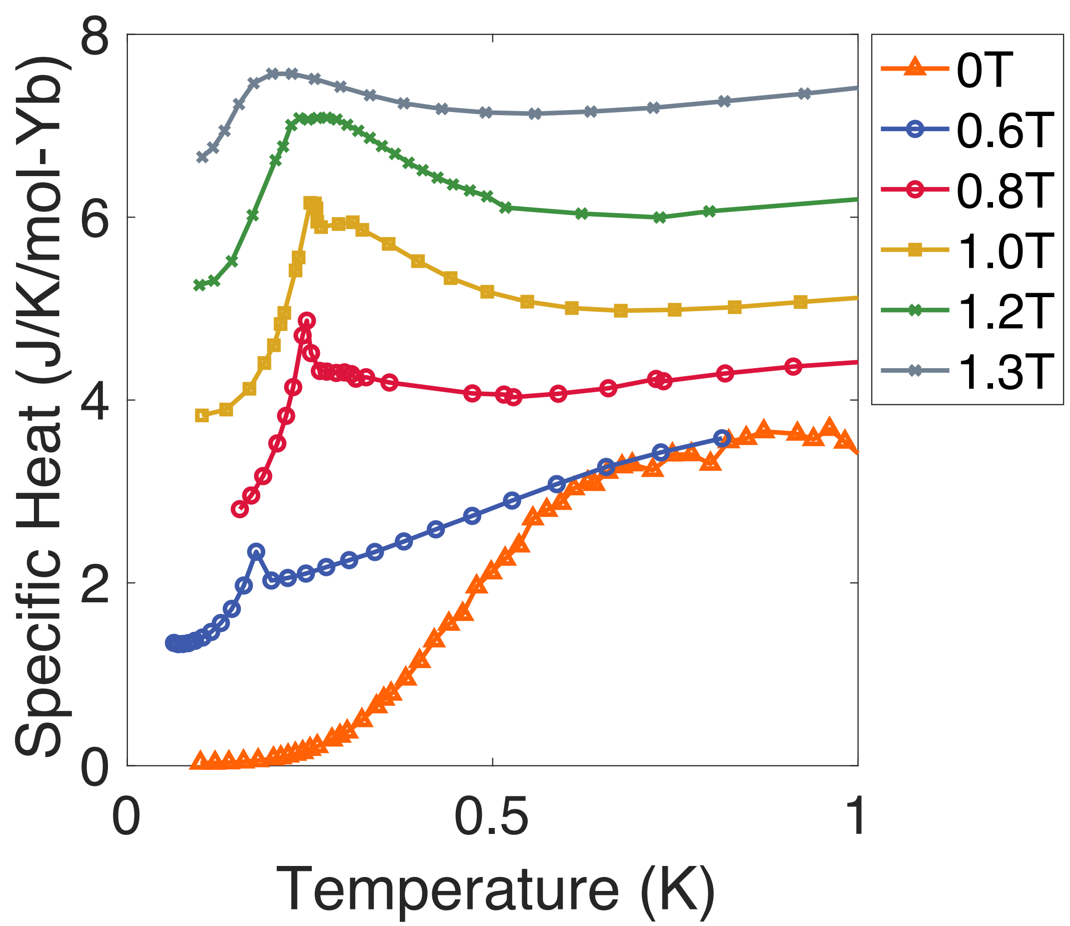
(2,0,0) Magnetic Bragg Peak (T = 100 mK)

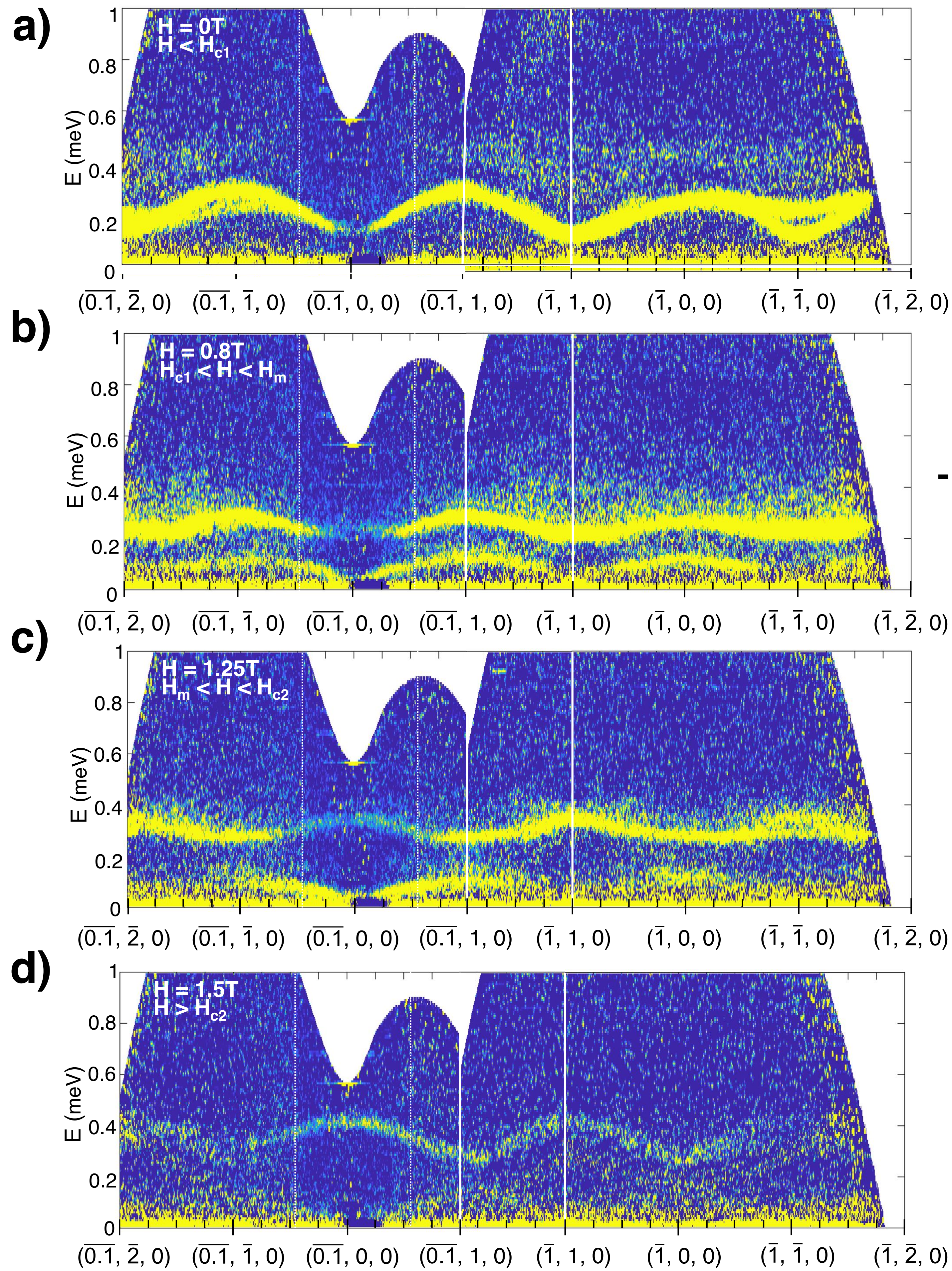


Field Induced Magnetic Order: BEC Dome?

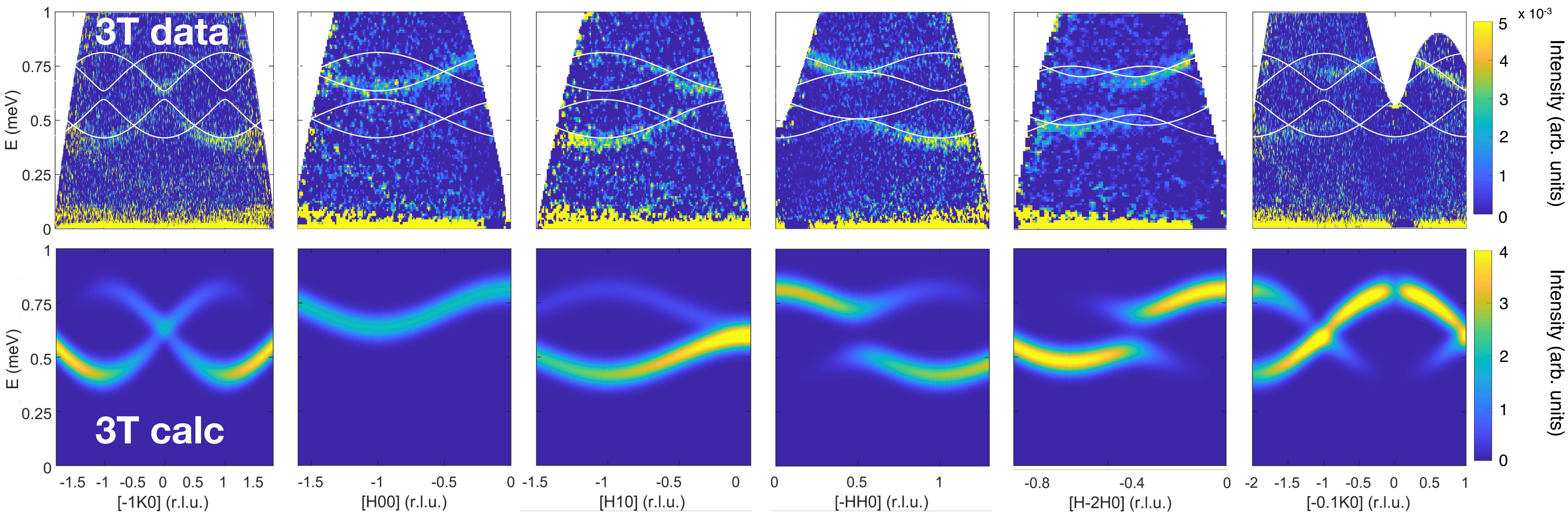



0.30
0.25
0.20
0.15
0.10
0.05
T (K)
H_{c1}
H_m
H_{c2}
0.0
0.5
1.0
1.5
$\mu_0 H$ (T)


Specific Heat (J/K/mol-Yb)
Temperature (K)
0T
0.6T
0.8T
1.0T
1.2T
1.3T


a)
H = 0T
H < H_{c1}
b)
H = 0.8T
H_{c1} < H < H_m
c)
H = 1.25T
H_m < H < H_{c2}
d)
H = 1.5T
H > H_{c2}
E (meV)
$(\overline{0.1}, \bar{2}, 0)$
$(\overline{0.1}, \bar{1}, 0)$
$(\overline{0.1}, 0, 0)$
$(\overline{0.1}, 1, 0)$
$(\bar{1}, 1, 0)$
$(\bar{1}, 0, 0)$
$(\bar{1}, \bar{1}, 0)$
$(\bar{1}, \bar{2}, 0)$

Field polarized data: fit to extract interactions



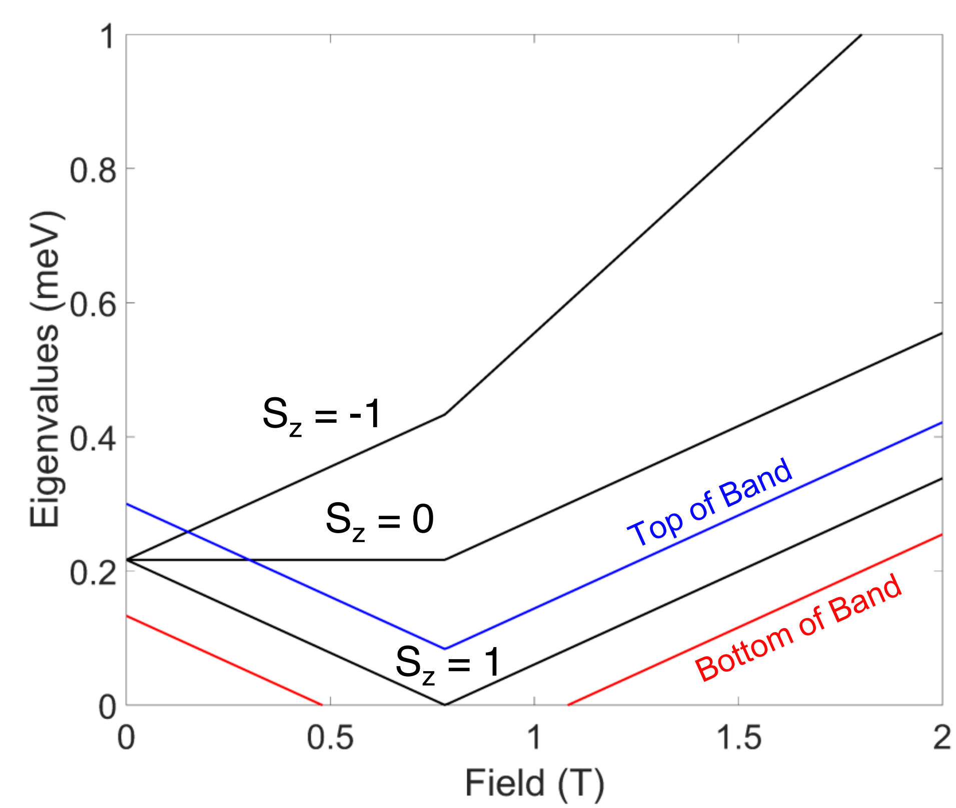
Spin Waves

J_{intra} = 0.217(3) meV
J_{inter} = 0.089(1) meV

Heat Capacity

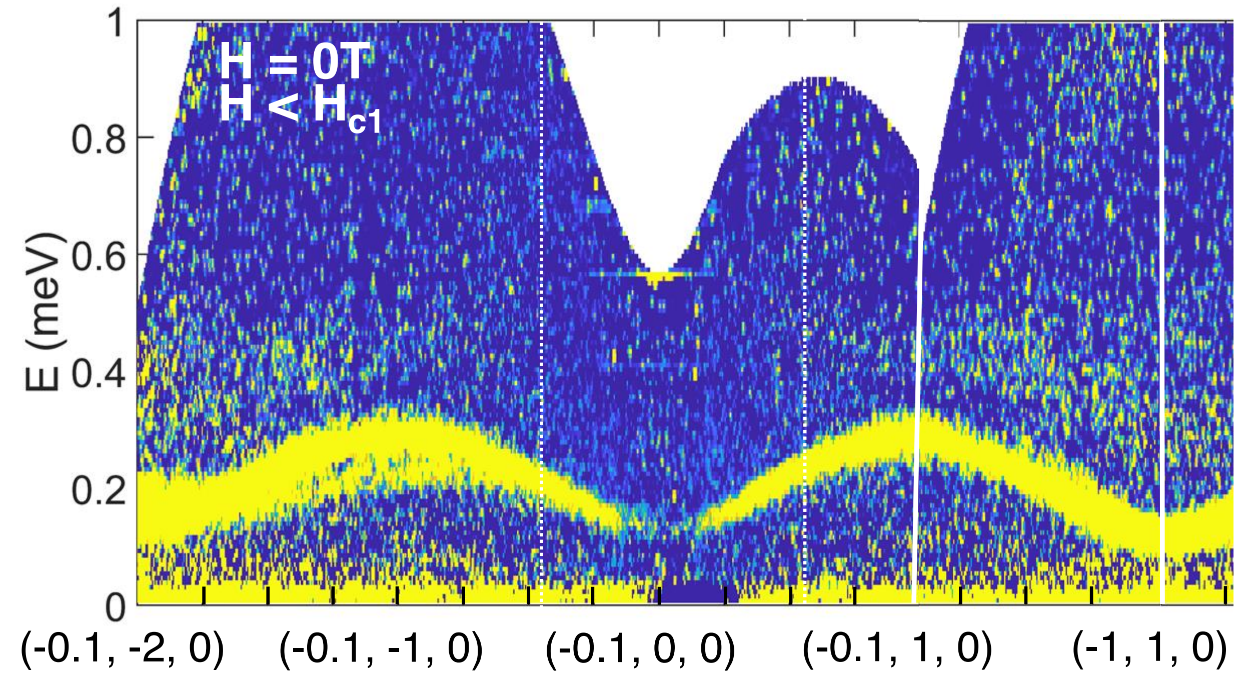
J_{intra} = 0.236(4) meV
J_{inter} = 0.06(2) meV

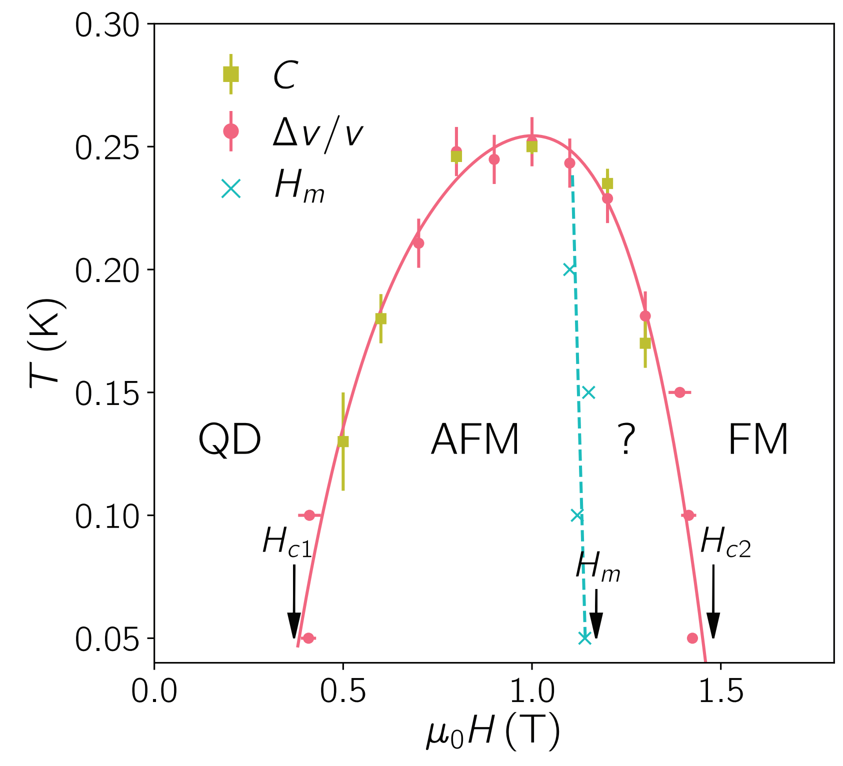
Heisenberg model with intra- and inter-dimer interactions: gets H_{c1} and H_{c2} approximately correct, reasonable fit to 3T spin waves



How good is the Heisenberg model for $Yb_2Si_2O_7$?

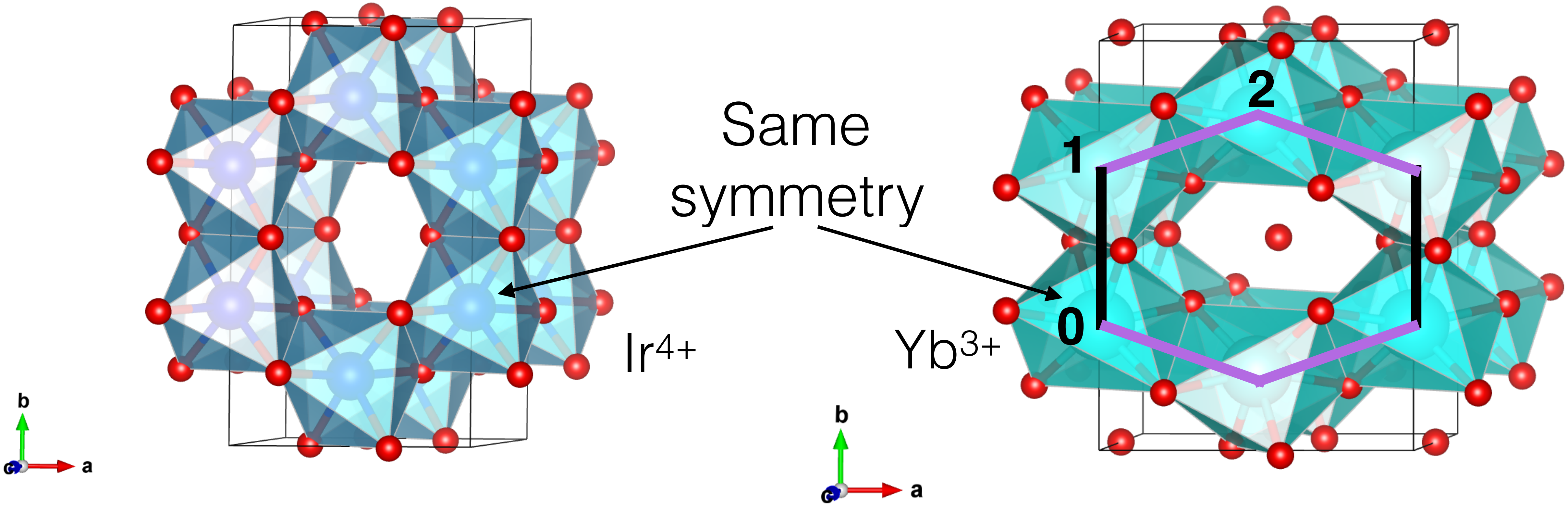
- Goldstone mode is gapless *to within the energy resolution of the instrument,* which is 16% of J_{intra}
- Need some other ingredient beyond Heisenberg exchange, to account for the *mystery phase?*
- *On the other hand* - reason to think Heisenberg is not so bad in some Yb materials…





J. G. Rau, M.J. P. Gingras, Phys. Rev. B 98, 054408 (2018)
L.S. Wu et al, Science, 352 (2016)

Na_2IrO_3 (C2/m) $Yb_2Si_2O_7$ (C2/m)



("Kitaev" honeycomb material)

(Quantum Dimer Magnet - relevance of Kitaev exchange?)

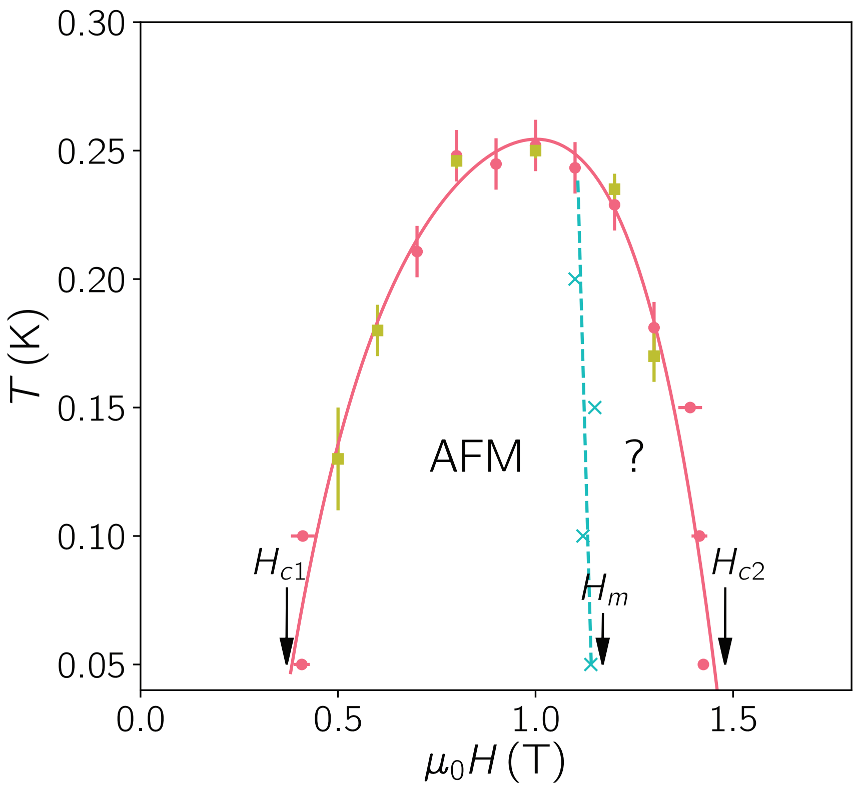
$$JS_0S_1 + KS_0^\gamma S_1^\gamma + \Gamma(S_0^\alpha S_1^\beta + S_0^\beta S_1^\alpha)$$

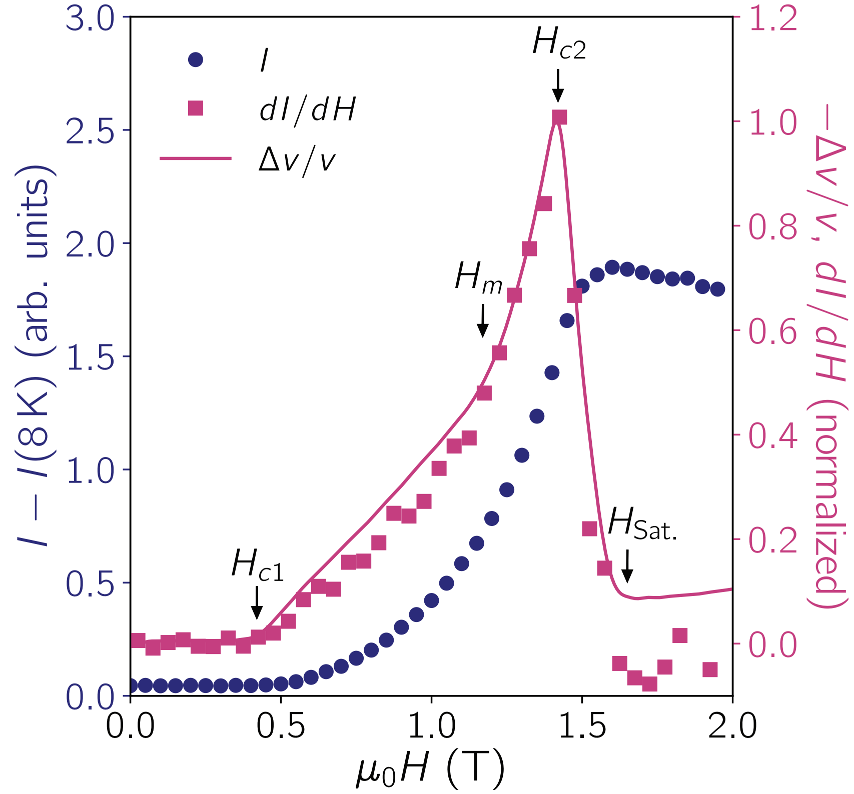
Symmetry-allowed effective exchange

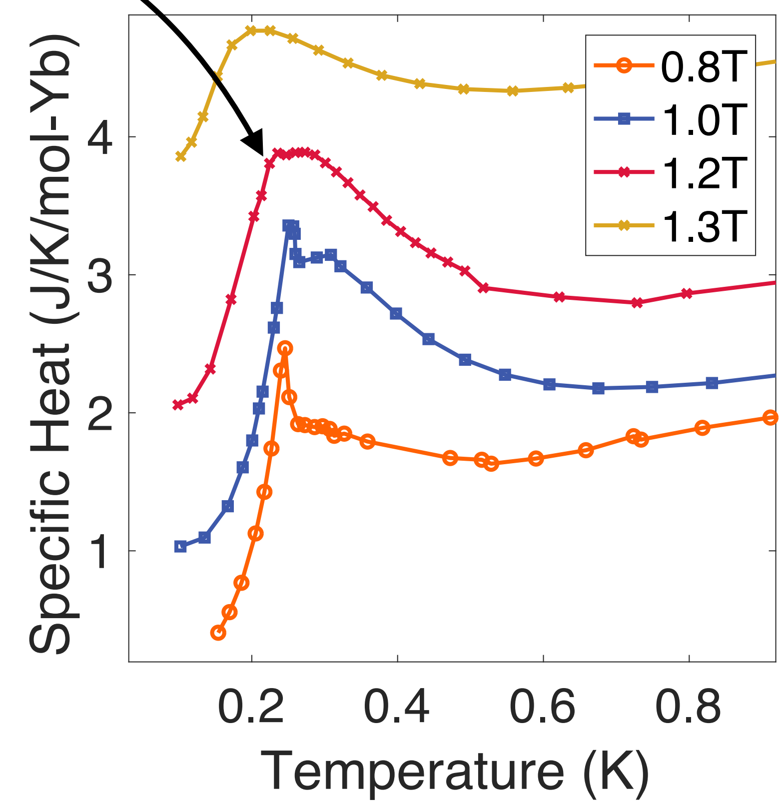
$$\mathcal{J}_{01} = \begin{bmatrix} J_{xx} & 0 & 0 \\ 0 & J_{yy} & J_{yz} \\ 0 & J_{yz} & J_{zz} \end{bmatrix} \qquad \mathcal{J}_{12} = \begin{bmatrix} J_1 & J_4 & J_5 \\ J_4 & J_2 & J_6 \\ J_5 & J_6 & J_3 \end{bmatrix}$$

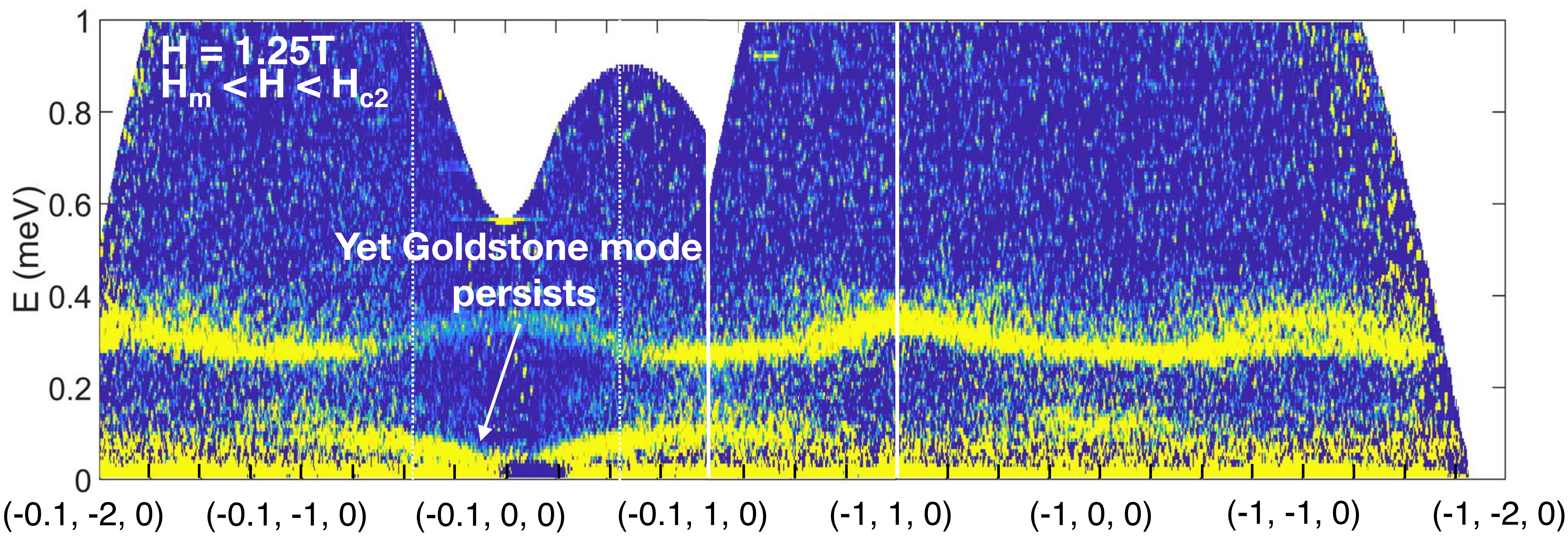
What is going on above H_m?

Signatures of symmetry-breaking vanish?









Potential explanation for H>Hm

On two phases inside the Bose condensation dome of $Yb_2Si_2O_7$

Michael O. Flynn,[1,*] Thomas E. Baker,[2] Siddharth Jindal,[3] and Rajiv R. P. Singh[1]

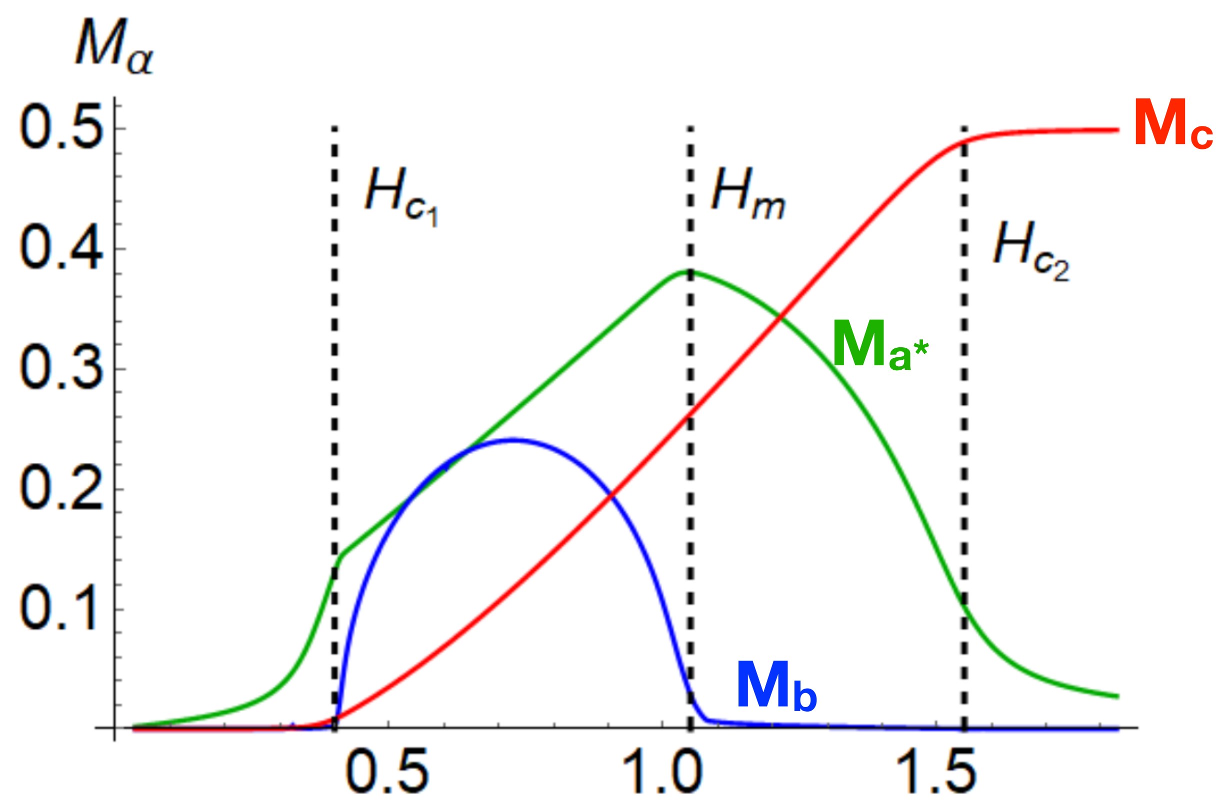
https://arxiv.org/pdf/2001.08219.pdf

Proposal: very weak exchange anisotropy in combination with a weak *field-induced staggered field* (off diagonal g-tensor component)

$$H = \sum_{\langle ij\rangle,\alpha} J^{\alpha}_{ij} S^{\alpha}_i S^{\alpha}_j - h \sum_{i,\alpha} g_{z\alpha} S^{\alpha}_i$$

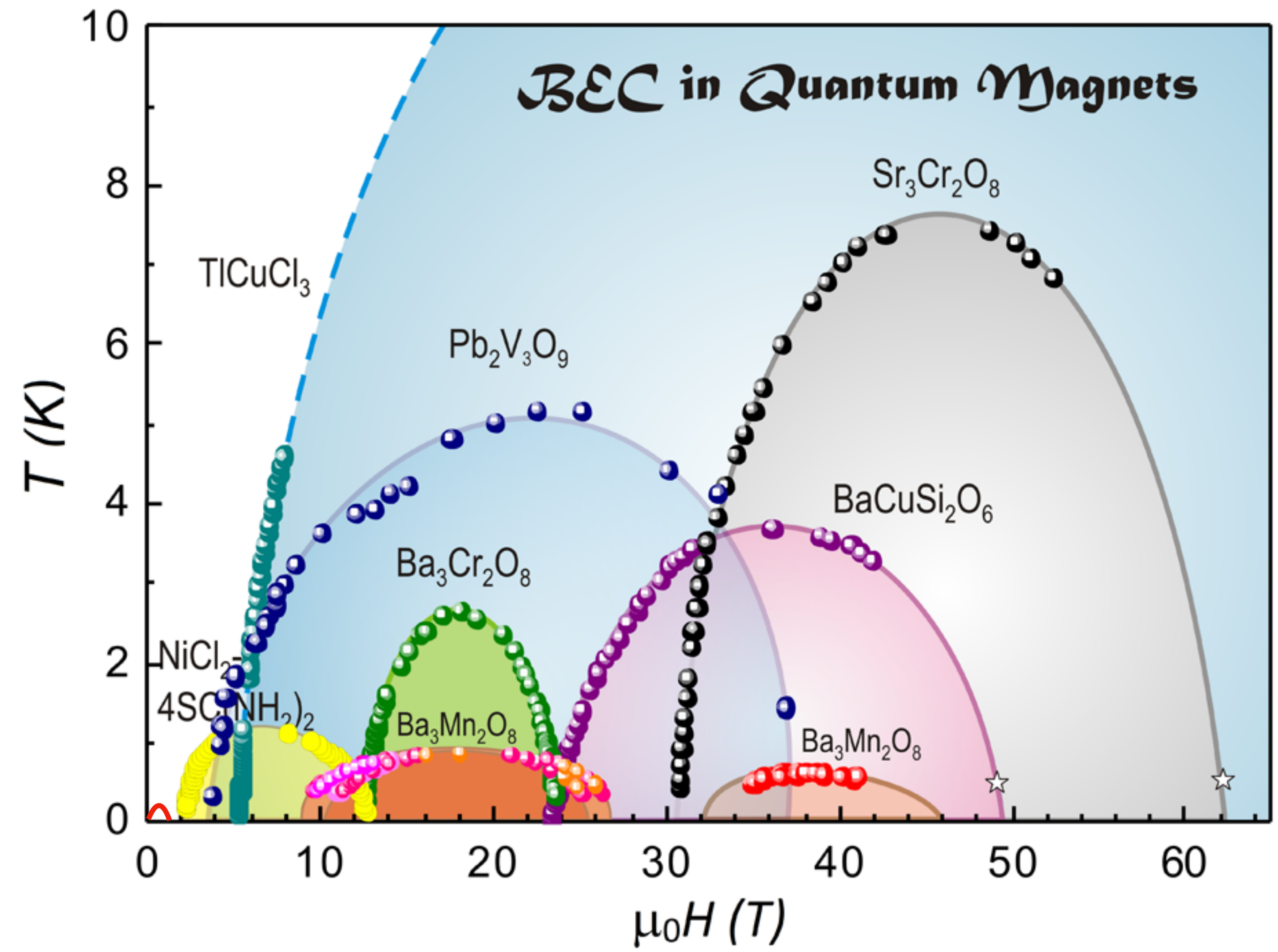
$$J^{y}_{ij} = (1+\lambda)\, J^{x}_{ij}$$

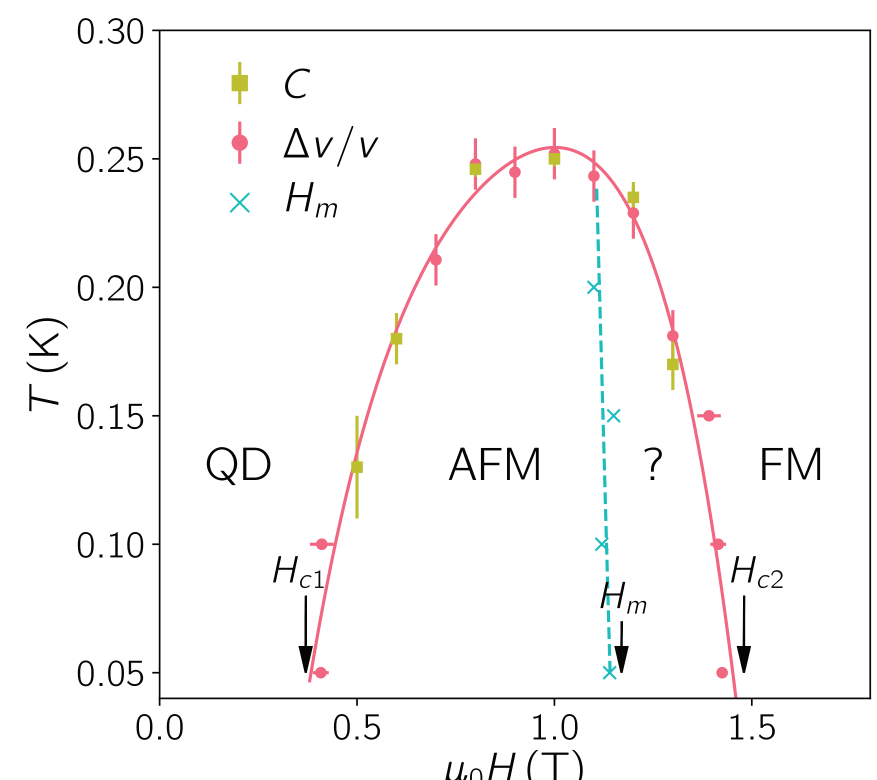
$$g_{zx} \ll g_{zz} \text{ with } g^{A}_{zx} = -g^{B}_{zx}$$



Summary

- $Yb_2Si_2O_7$ is the first Yb^{3+}-based QDM with BEC-like phase
- Low energy scale for interactions — very low critical fields
- Signatures of BEC (goldstone mode) to within at least 16% of largest Heisenberg interaction
- Strange phase in high field part of dome - result of modified weakly interacting boson model?





G. Hester, et al. *Novel Strongly Spin-Orbit Coupled Quantum Dimer Magnet: $Yb_2Si_2O_7$*. PRL, **123** 027201 (2019)

H. S. Nair, *et al*. *Crystal Growth of Quantum Magnets in the Rare-Earth Pyrosilicate Family R2Si2O7 (R = Yb, Er) Using the Optical Floating Zone Method*. Crystals 9(4), **196** (2019)

Acknowledgements

Gavin Hester
CSU

Hari Nair
CSU → UTEP

Tim Reeder
CSU → JHU

Jeff Quilliam
Sherbrooke U

- Danielle Yahne (CSU)
- Tim DeLazzer (CSU)
- Léo Berges (Sherbrooke)
- Djamel Ziat (Sherbrooke)
- Jamie Neilson (CSU)
- Adam Aczel (ORNL)
- Gabrielle Sala (ORNL)

NSF DMR-1611217

G. Hester, et al. *Novel Strongly Spin-Orbit Coupled Quantum Dimer Magnet:* $Yb_2Si_2O_7$. PRL, **123** 027201 (2019)

H. S. Nair, *et al*. *Crystal Growth of Quantum Magnets in the Rare-Earth Pyrosilicate Family R2Si2O7 (R = Yb, Er) Using the Optical Floating Zone Method*. Crystals 9(4), 196 (2019)