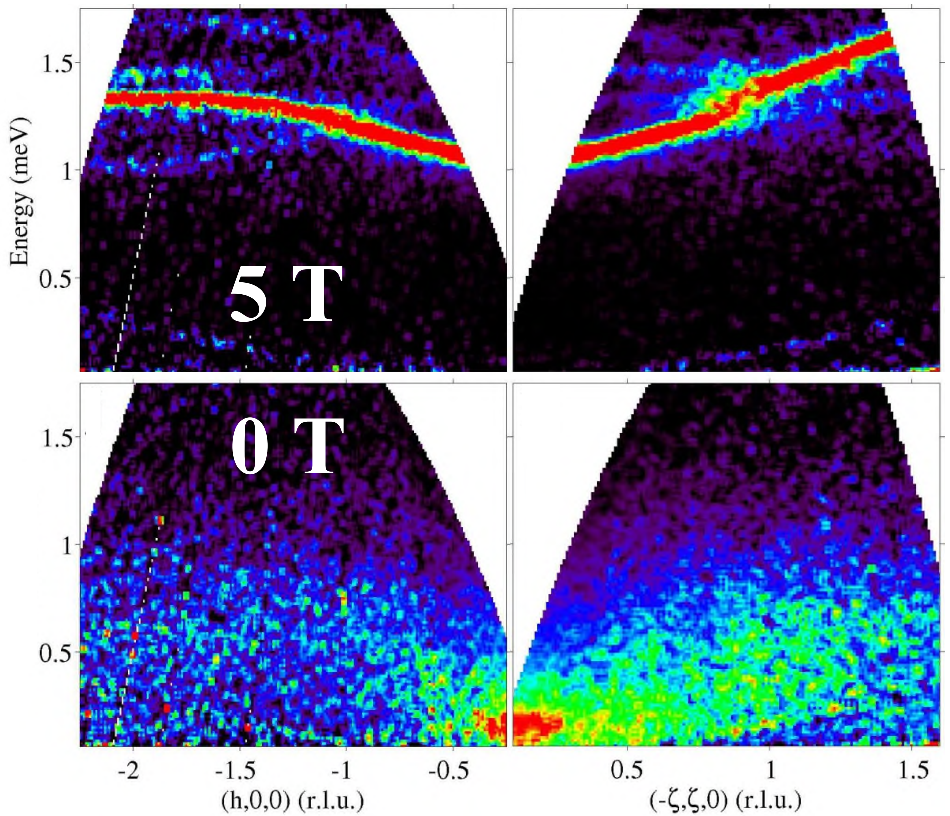
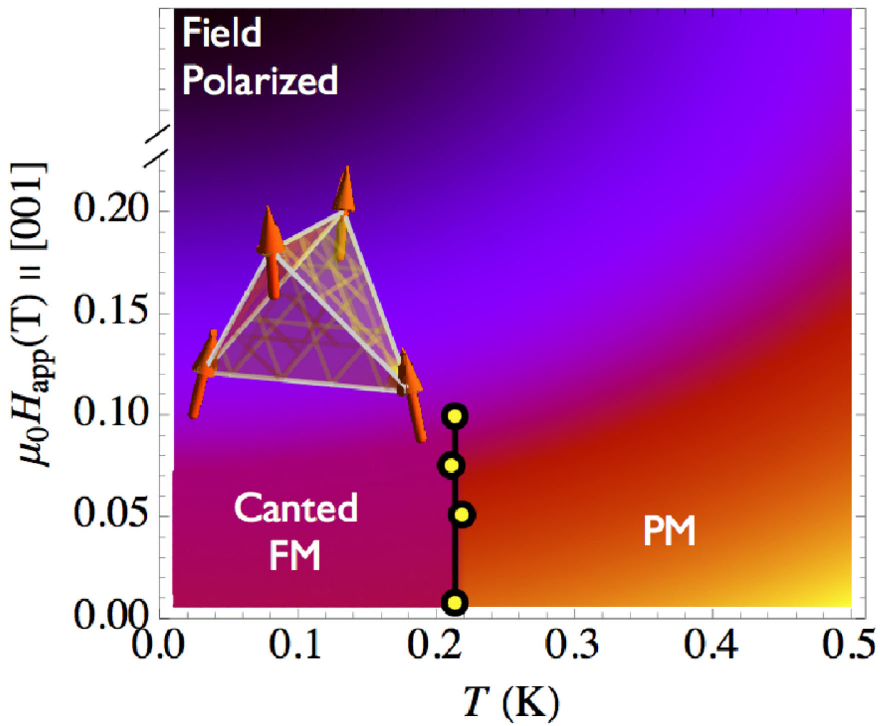
Anomalous spin dynamics in the frustrated quantum pyrochlore $Yb_2Ti_2O_7$ in magnetic field

Radu Coldea (Oxford and KITP)





Collaborators

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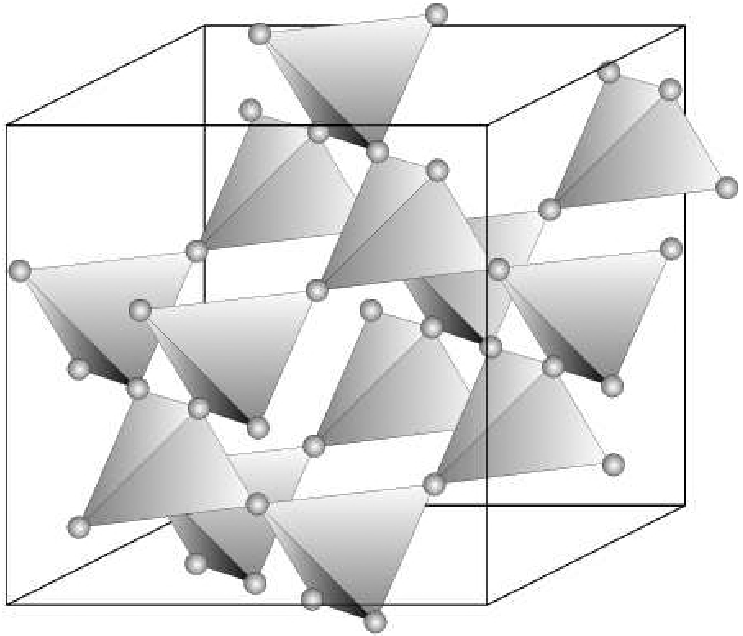
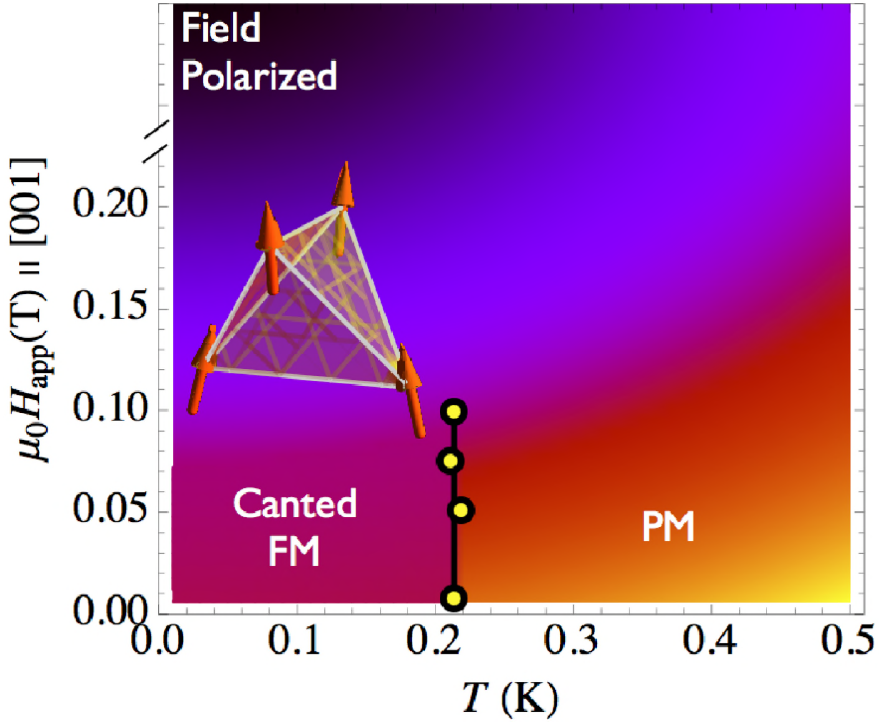
Inelastic Neutron Scatering on LET spectrometer @ ISIS Facility



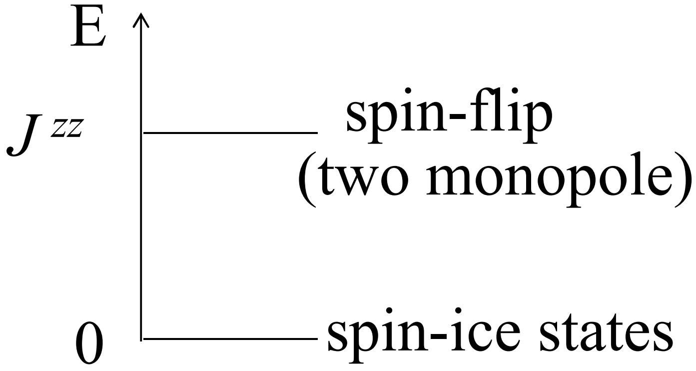


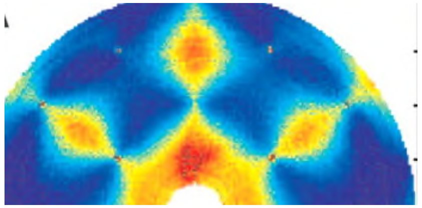
Outline

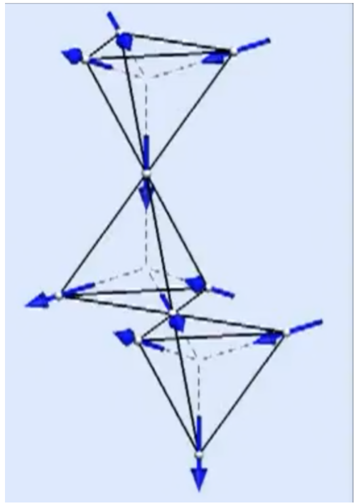
1. $Yb_2Ti_2O_7$ candidate for novel spin dynamics
2. Spin excitations at high field, one and two-magnons
3. Spin dynamics at low field, dominant continuum lineshapes
4. Specific heat & phase diagram in field

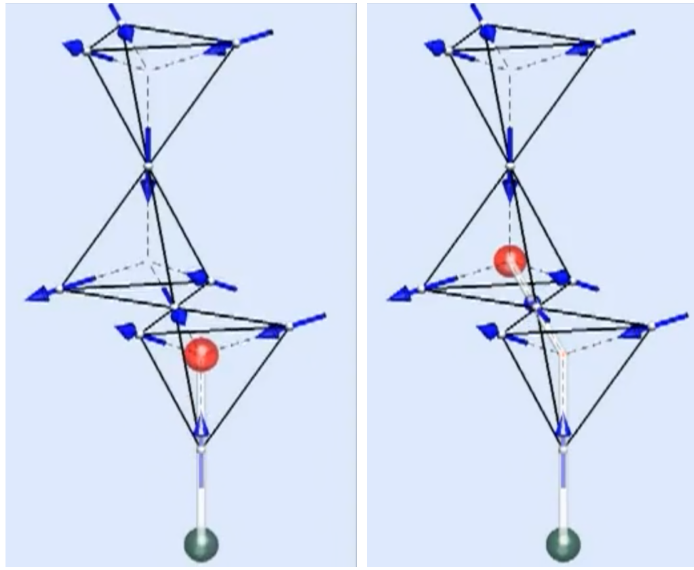
Spin ice physics on the pyrochlore lattice

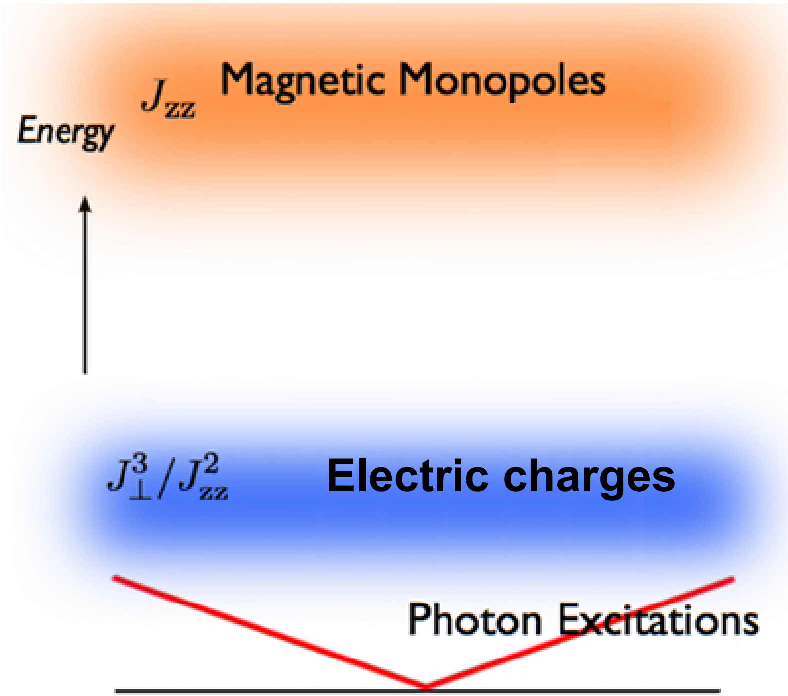
- corner-shared tetrahedra, Ising spins (local 111 axis) coupled FM many degenerate states => Spin Ice, 2 in 2 out, $Ho_2Ti_2O_7$





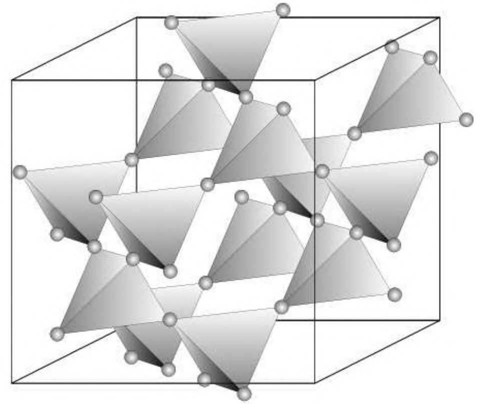


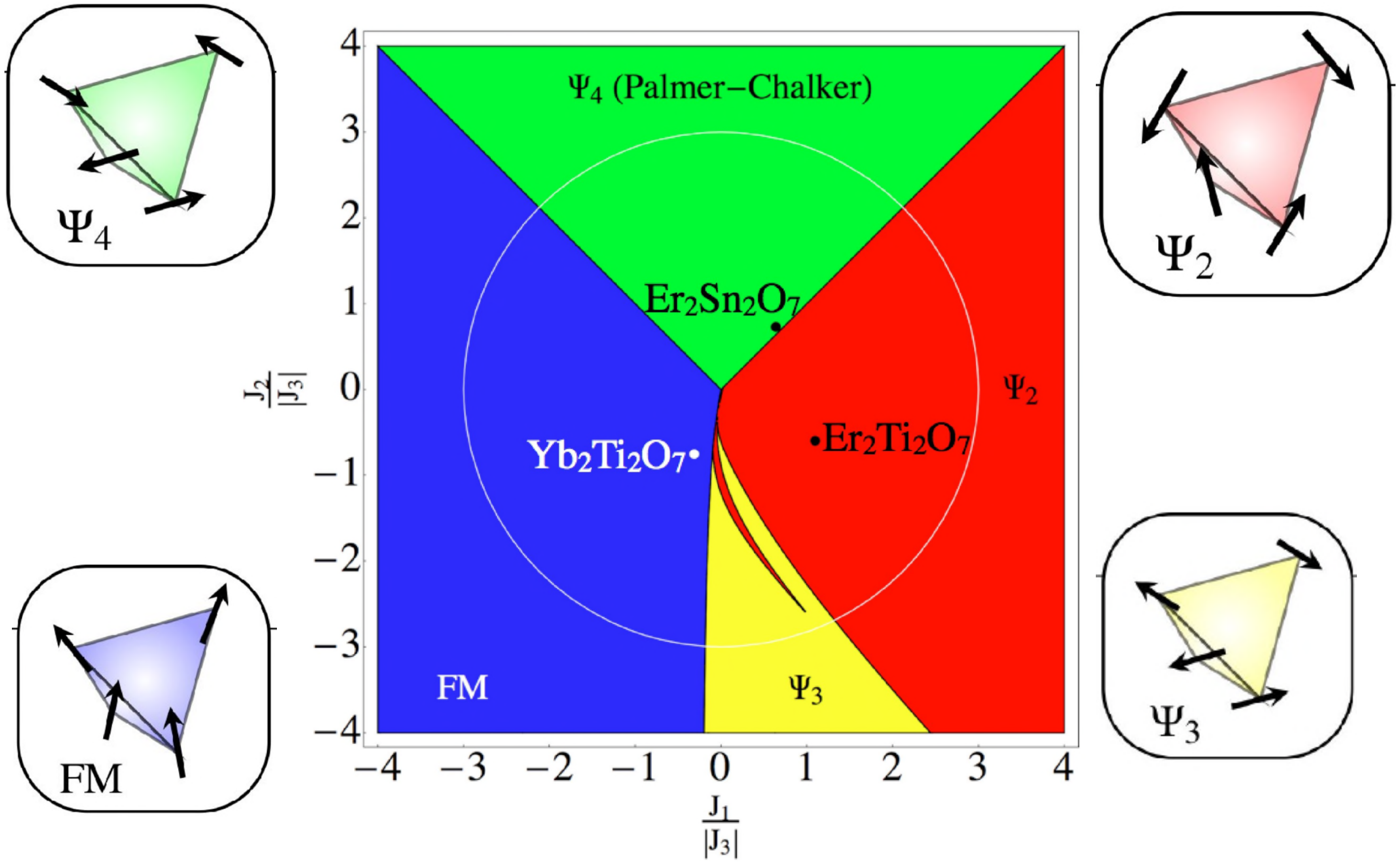




- additional "transverse exchange" terms -> "**quantum spin ice**" (photon + propagating monopoles)

Semi-classical phase diagram of ordered phases on the pyrochlore lattice



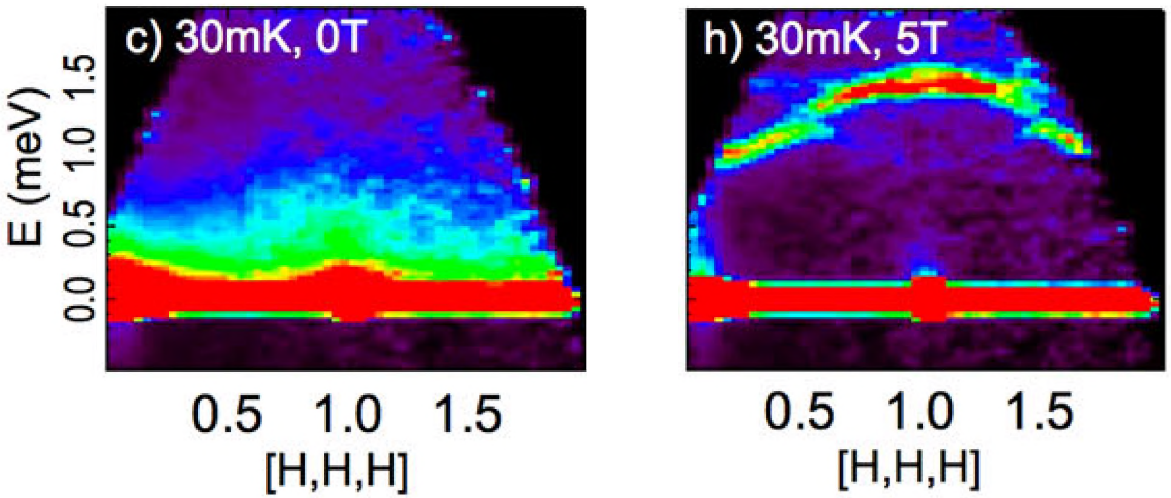


$$\mathbf{J}_{01} = \begin{pmatrix} J_2 & J_4 & J_4 \\ -J_4 & J_1 & J_3 \\ -J_4 & J_3 & J_1 \end{pmatrix}$$

Yan, …Shannon arXiv:1311.3501 (2013)

- quantum dynamics effects ?

$Yb_2Ti_2O_7$ spin dynamics : open questions



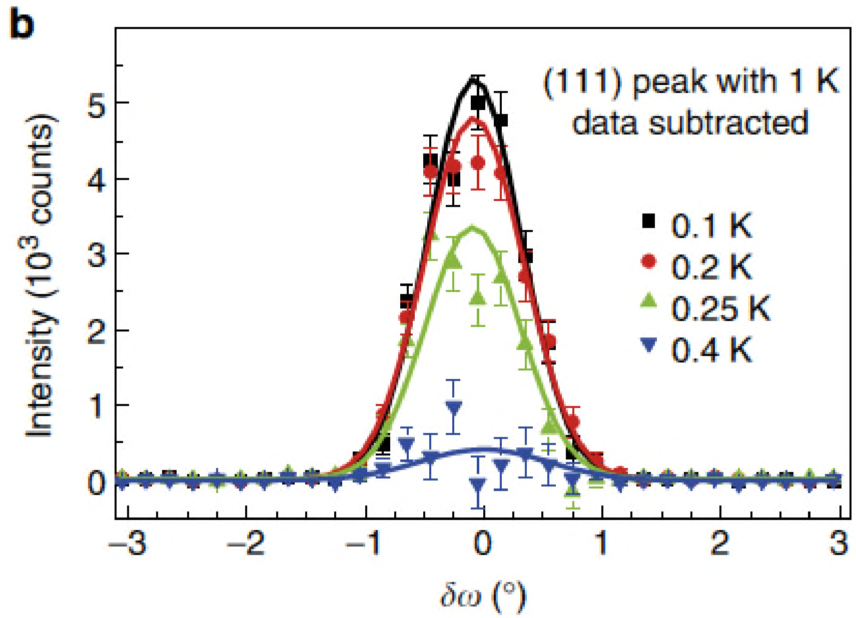
B=0 broad scattering reported

Field-polarized state
sharp magnon modes

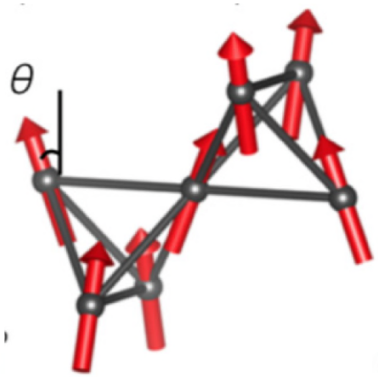
Ross et al. (*2009,2011*)

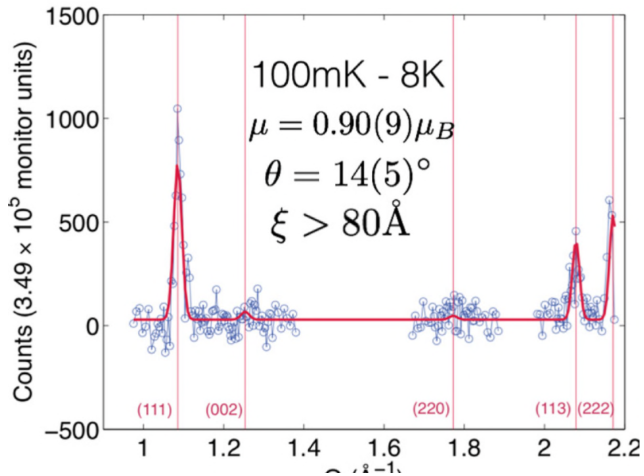
- **how do magnons disappear upon lowering B**?
- if broad scattering at B=0 is due to quantum fluctuations, are fluctuations still present at high field, what is their manifestation, **how do fluctuations evolve upon lowering field** (gradually or with a sharp onset below a critical field) ?
- **what is B=0 magnetic excitations spectrum** (lineshapes, ***Q***-modulations), is a quasiparticle description possible, what is the physical picture?
- if continuum due to magnetic monopoles of a Quantum Spin Ice expect characteristic field behaviour

$Yb_2Ti_2O_7$ low temperature phase: canted ferromagnet



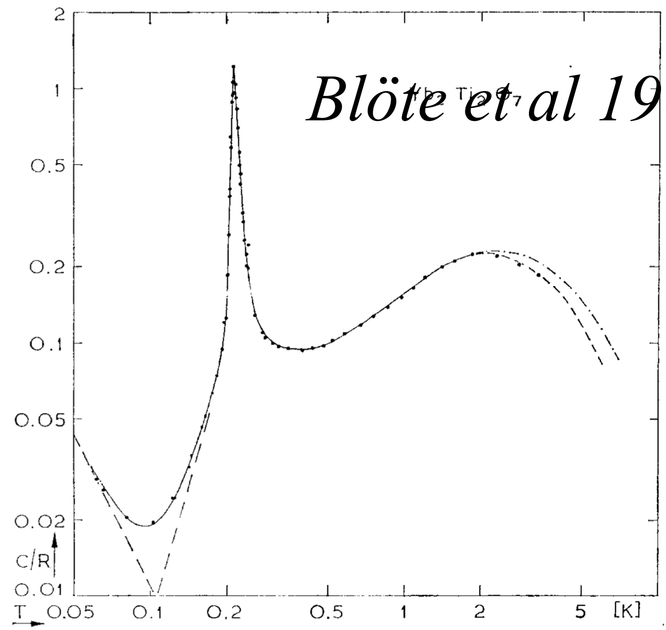
Chang et al (2012)





Gaudet et al (2016)

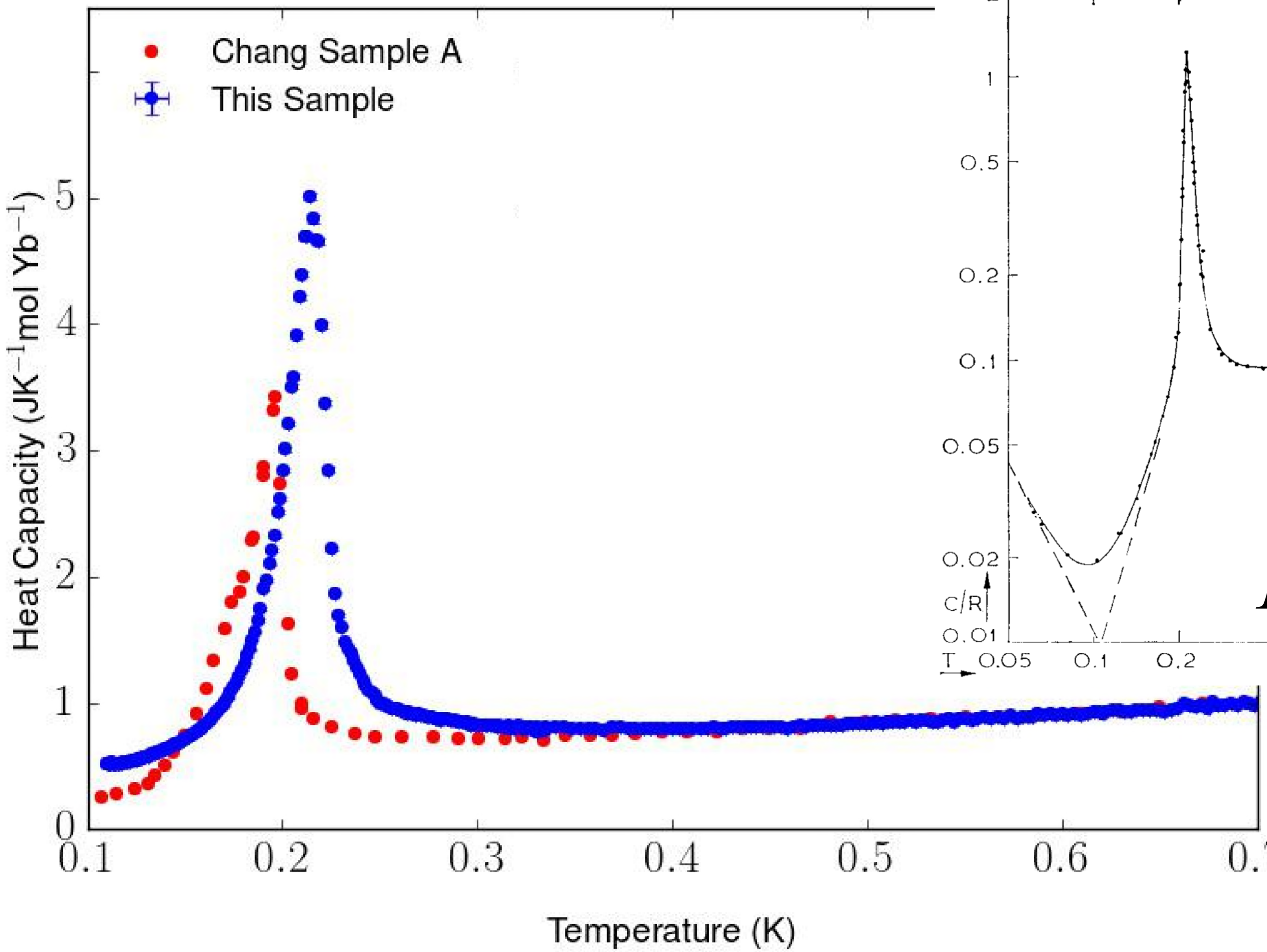
- polarized single crystal neutron diffraction & powder neutron diffraction confirm spontaneous ferromagnetic order below peak in heat capacity 0.21-0.26 K (1st order transition, 6 domains)

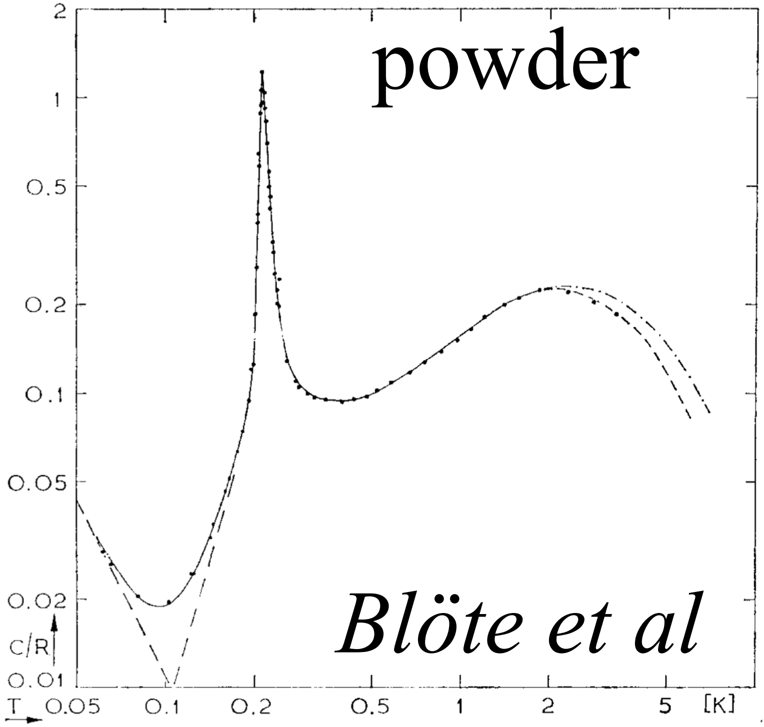


Blöte et al 1969

- we probe magnetic excitations in [001] field (single magnetic domain + simpler phase diagram)

$Yb_2Ti_2O_7$ single crystal specific heat

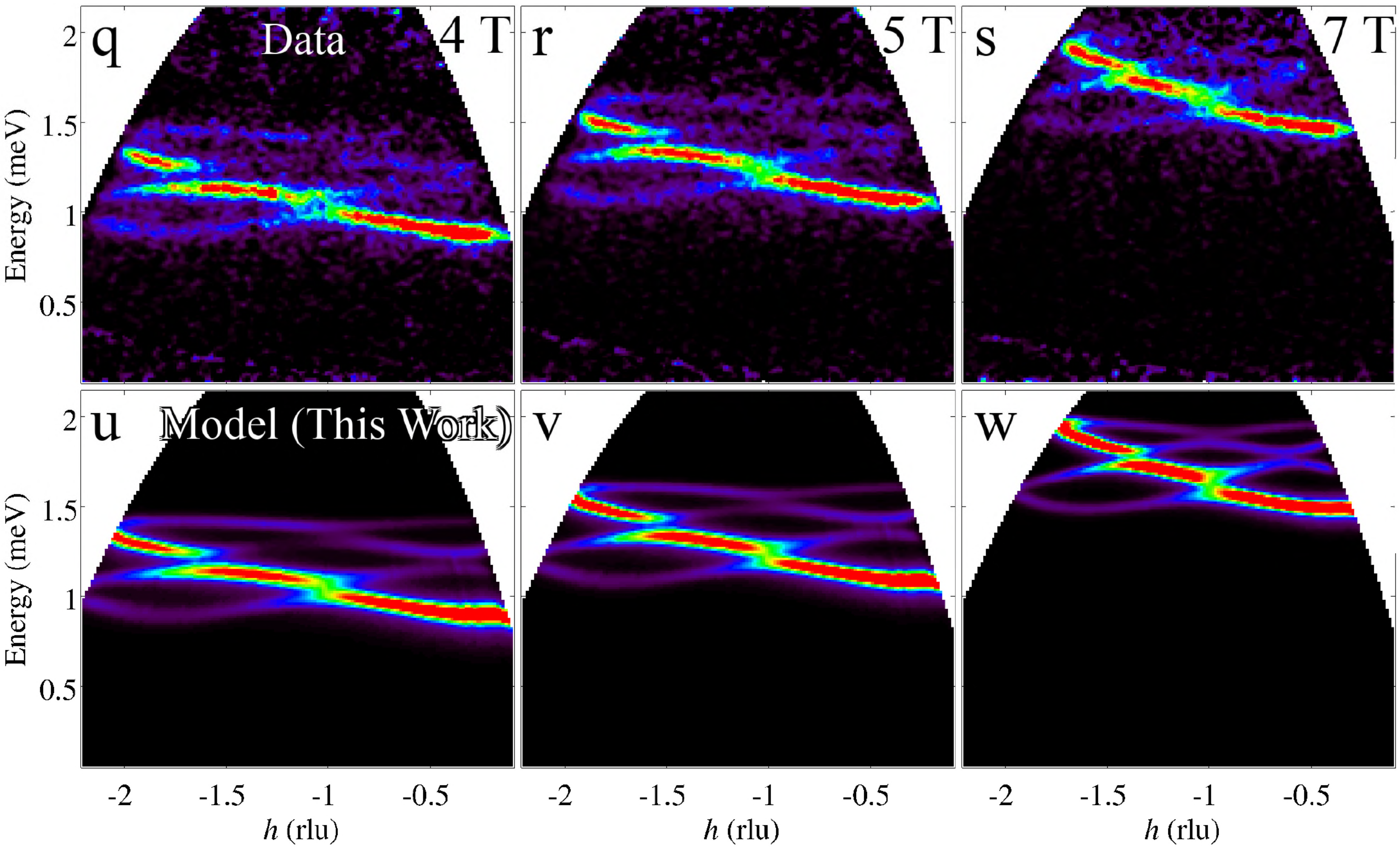


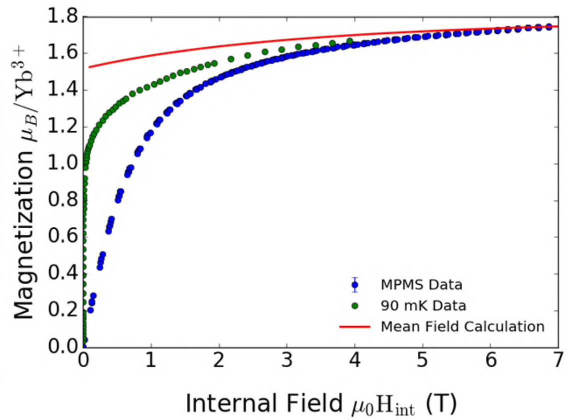


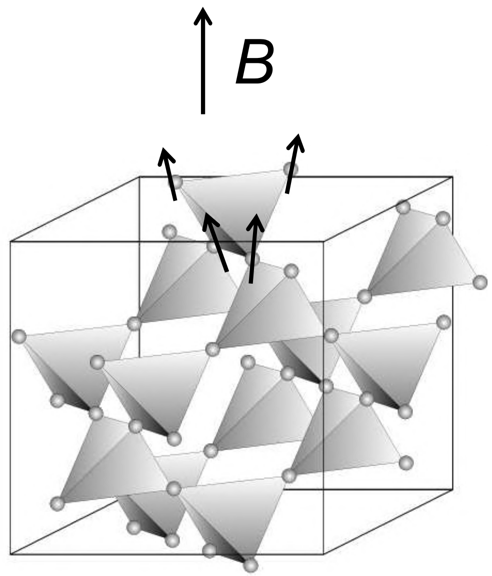
- our image furnace "slow"-grown & annealed single crystal shows a single, sharp specific heat peak at 0.214 K -> similar in behaviour to high purity limit (stoichiometric powders have 1st order transition 0.24-0.26 K)

Chang et al (2012) *Arpino …McQueen (2017)*

Spin dynamics at high magnetic field // [001] 0.15 K







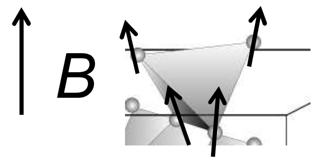
- observe sharp modes with gap increasing in field, as seen by *Ross et al*. in B//[-1,1,0]
- coherently-propagating spin-flips on 4 sublattices

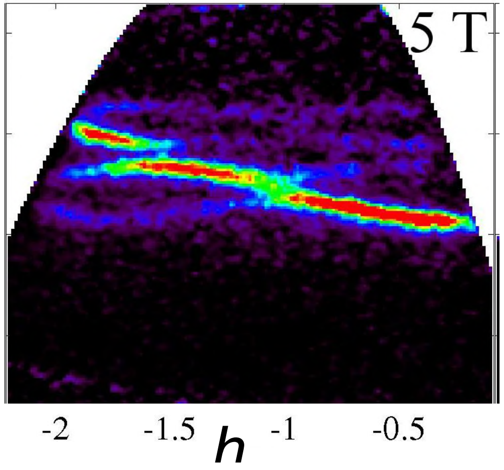
Parameterization by spin waves of a nn Hamiltonian

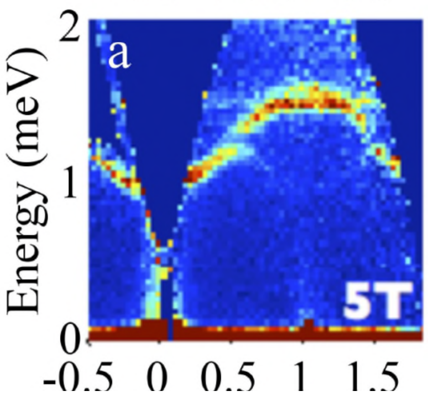
$$\mathcal{H}_{\text{Exchange}} = \sum_{\langle ij \rangle} \{ J_{zz} \mathsf{S}_i^z \mathsf{S}_j^z - J_{\pm} \left(\mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+ \right)$$
$$J_{\pm\pm} \left(\gamma_{ij} \mathsf{S}_i^+ \mathsf{S}_j^+ + \gamma_{ij}^* \mathsf{S}_i^- \mathsf{S}_j^- \right)$$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

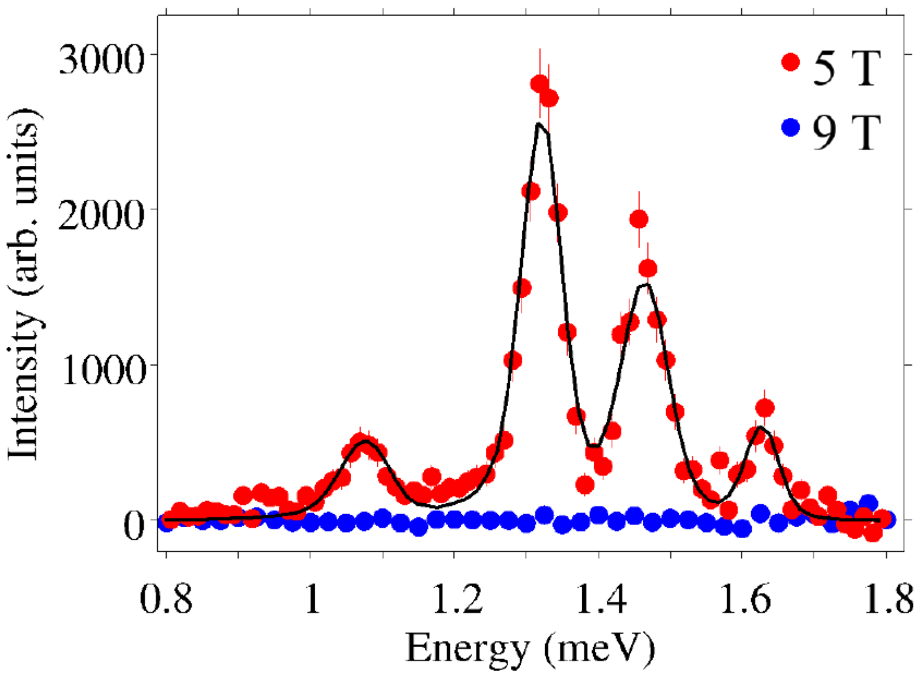
$$\mathbf{g} = \begin{pmatrix} g_\perp & 0 & 0 \\ 0 & g_\perp & 0 \\ 0 & 0 & g_\parallel \end{pmatrix}$$

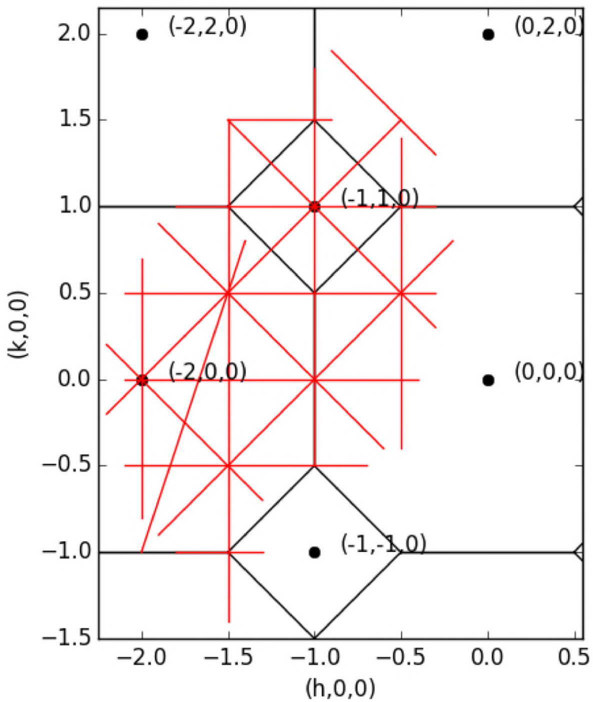
Ross, Savary, Gaulin
Balents (2011)









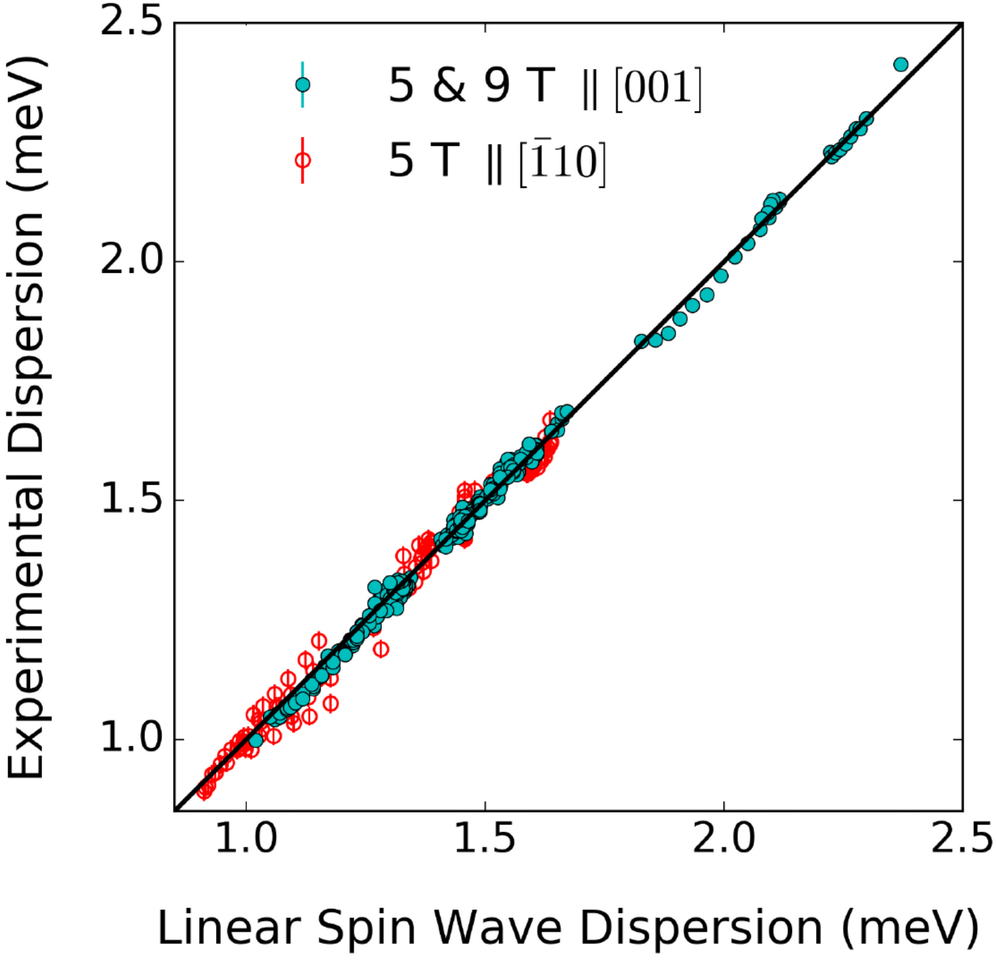


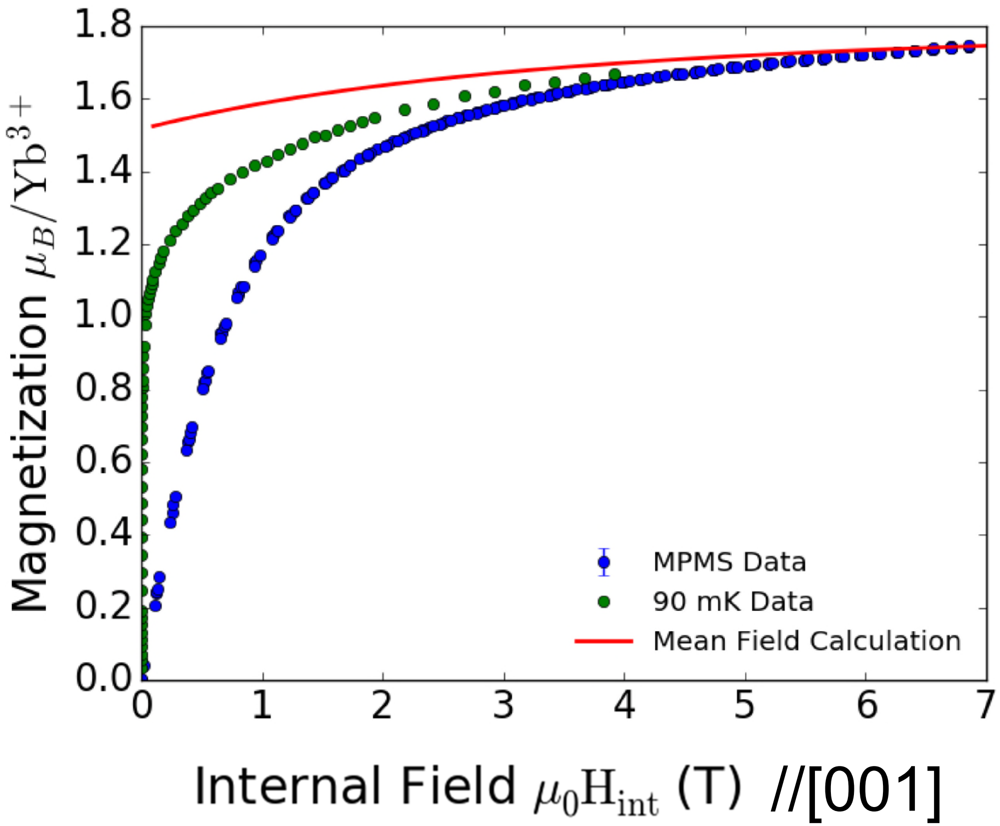
- pick dispersion points throughout the full volume of data (17 directions total) get (h,k,l,E) and mode index 1-4

- **fit 6 parameters** (4 symmetry-allowed nn exchange terms) + g tensor (2 terms) to data from **<u>both</u>** neutron experiments B//[001] and published B//[-1,1,0]

Parameterization by spin waves of a nn Hamiltonian

dispersion points fit



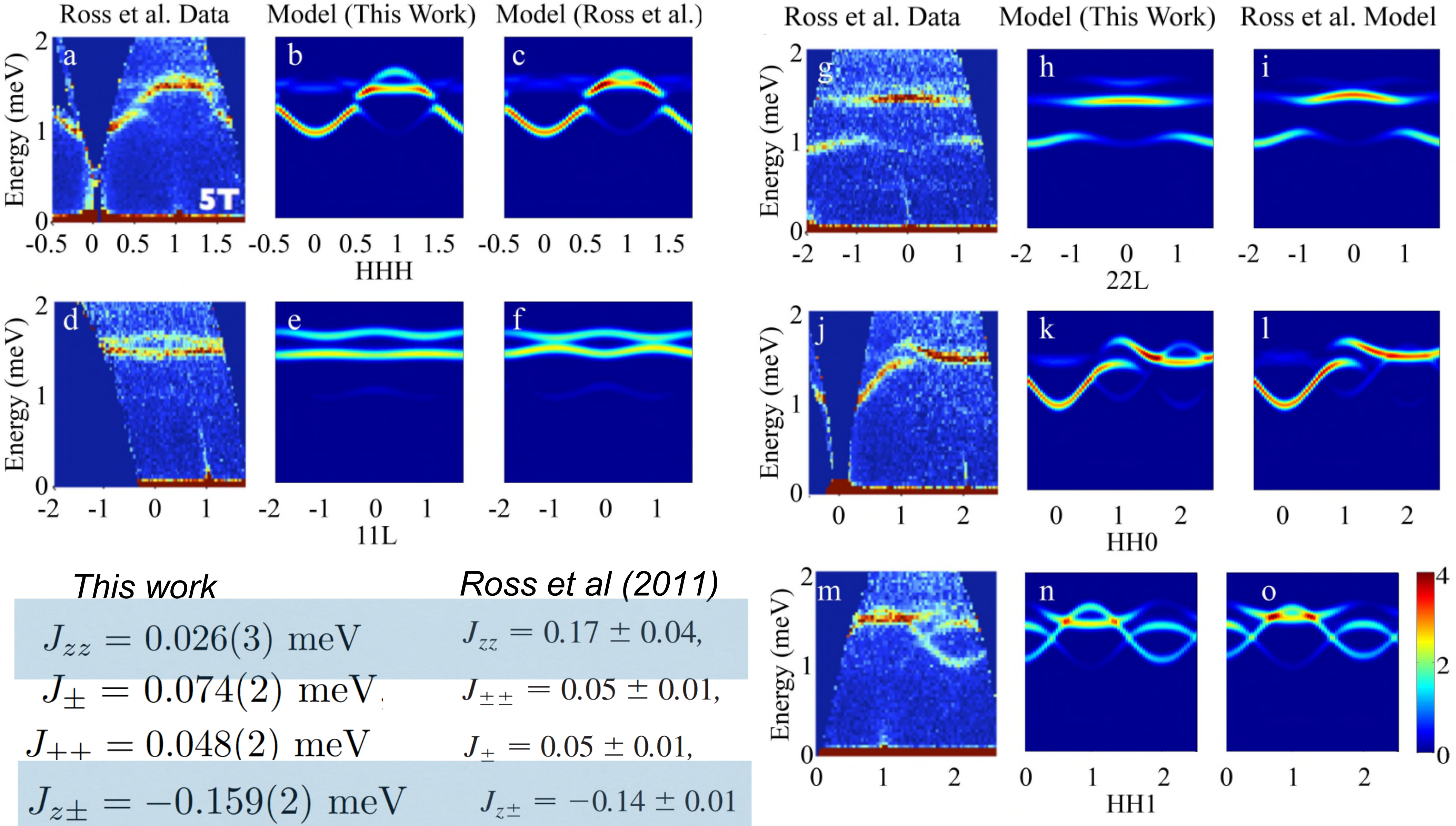


//[001]

- to fit all Hamiltonian parameters without any constraints use neutron data for the **two field orientations** (over 550 dispersion points) and **magnetization near saturation** (7 T)

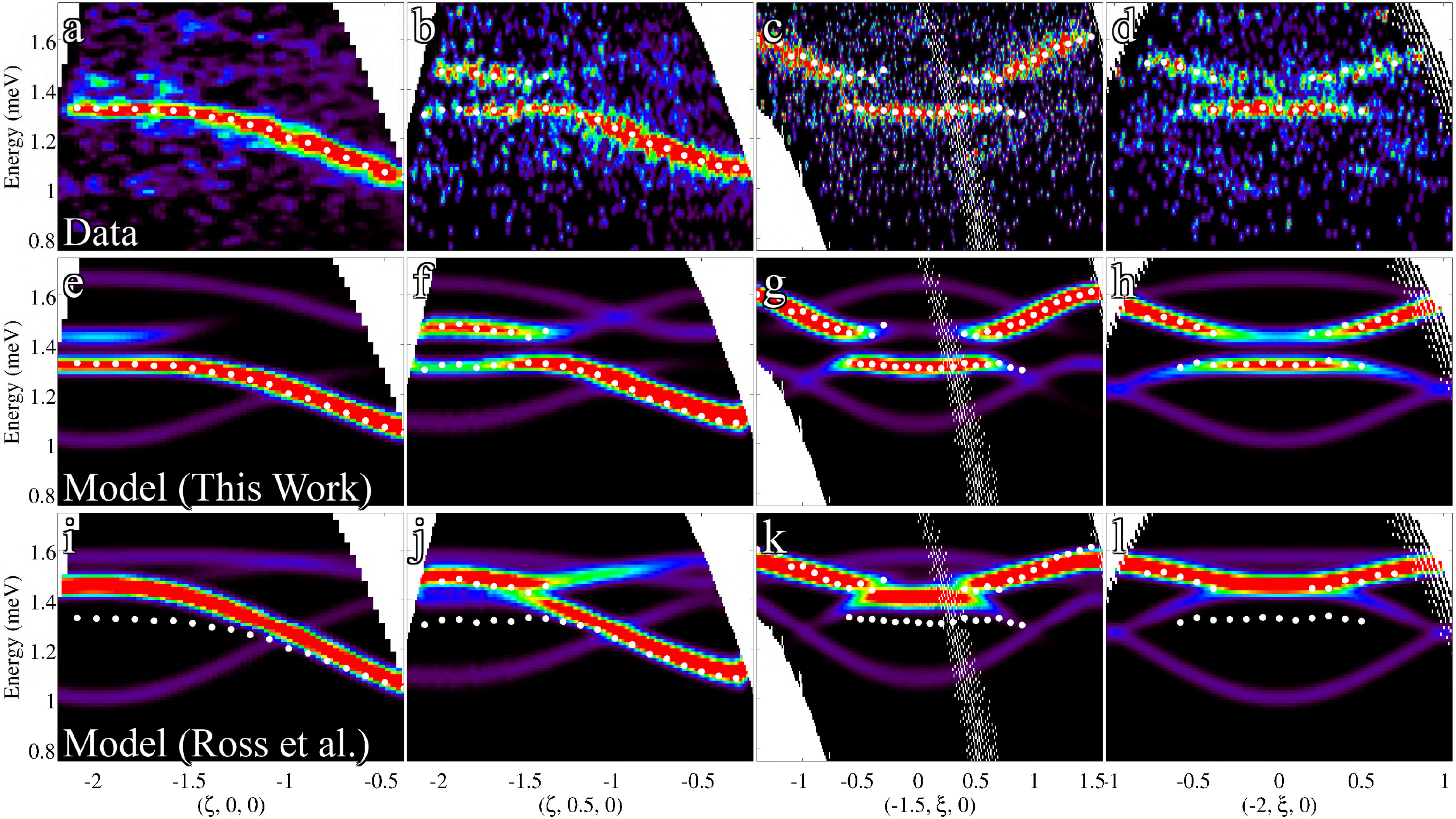
$$J_{zz} = 0.026(3)\ \mathrm{meV}$$
$$J_{\pm} = 0.074(2)\ \mathrm{meV}$$
$$J_{\pm\pm} = 0.048(2)\ \mathrm{meV}$$
$$J_{z\pm} = -0.159(2)\ \mathrm{meV}$$
$$g_{\|} = 2.14(3)$$
$$g_{\perp} = 4.17(2).$$

Parameterization of dispersions at B=5T // [-1,1,0]



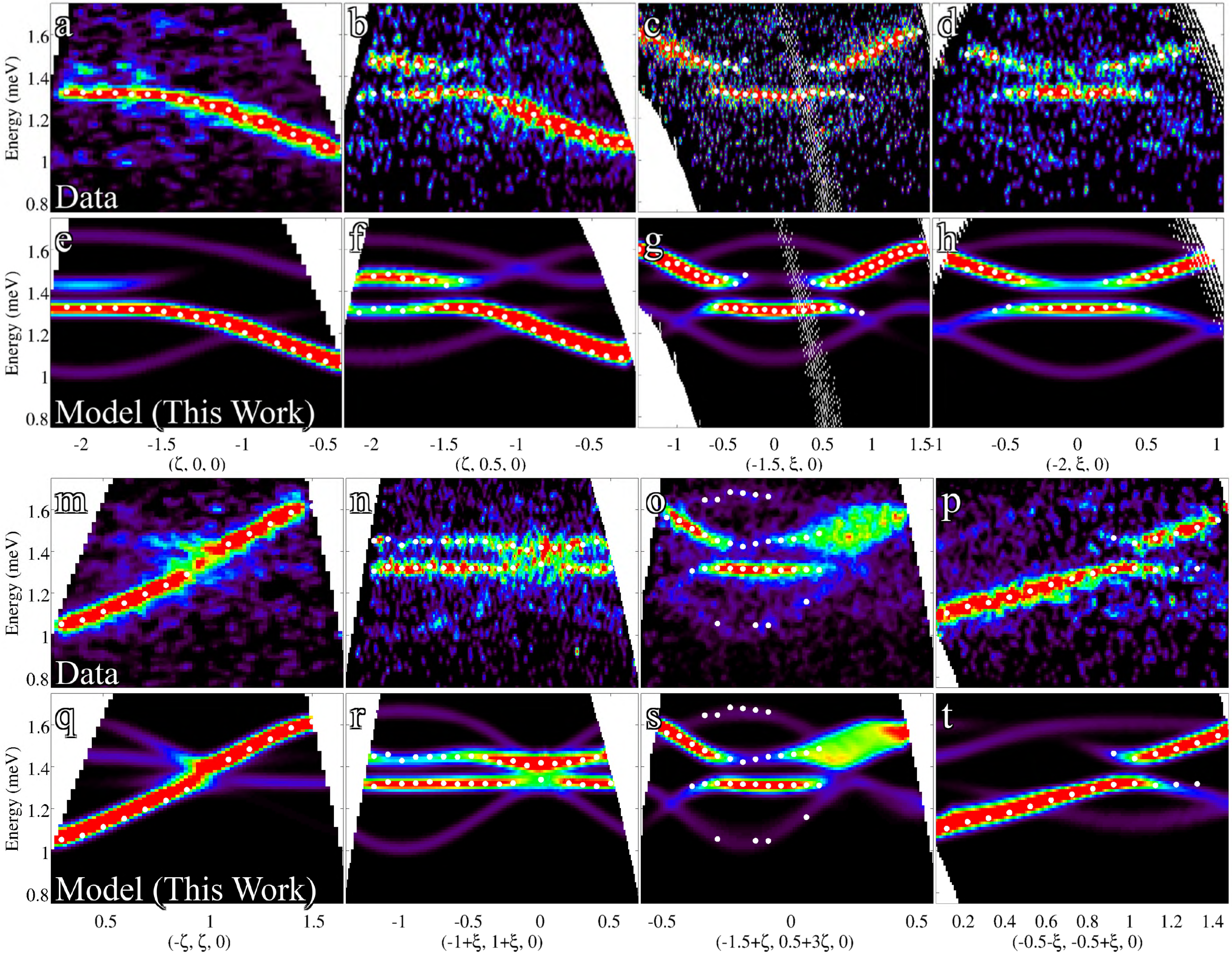
This work	*Ross et al (2011)*
$J_{zz} = 0.026(3)$ meV	$J_{zz} = 0.17 \pm 0.04,$
$J_{\pm} = 0.074(2)$ meV	$J_{\pm\pm} = 0.05 \pm 0.01,$
$J_{++} = 0.048(2)$ meV	$J_{\pm} = 0.05 \pm 0.01,$
$J_{z\pm} = -0.159(2)$ meV	$J_{z\pm} = -0.14 \pm 0.01$
$g_{\|} = 2.14(3)$	$g_z = 1.80$
$g_{\perp} = 4.17(2).$	$g_{xy} = 4.32$
(see also Robert et al. PRB 2015)	$g_{xy}/g_z = 2.4$ fixed

Parameterization of dispersions at B = 5T //[001]

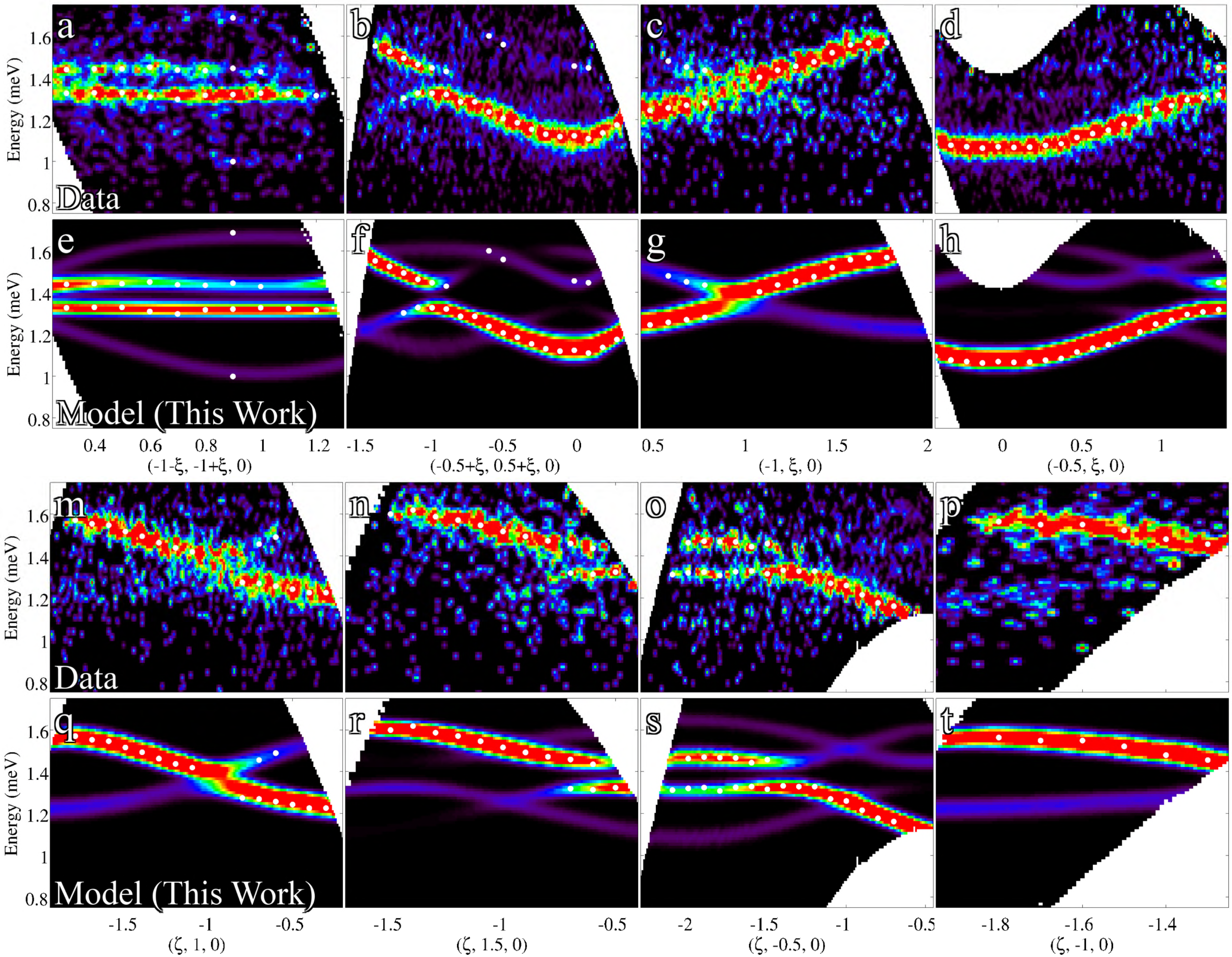


- earlier parameterization does not fit data in [001] field

Parameterization of dispersions at B=5T //[001]

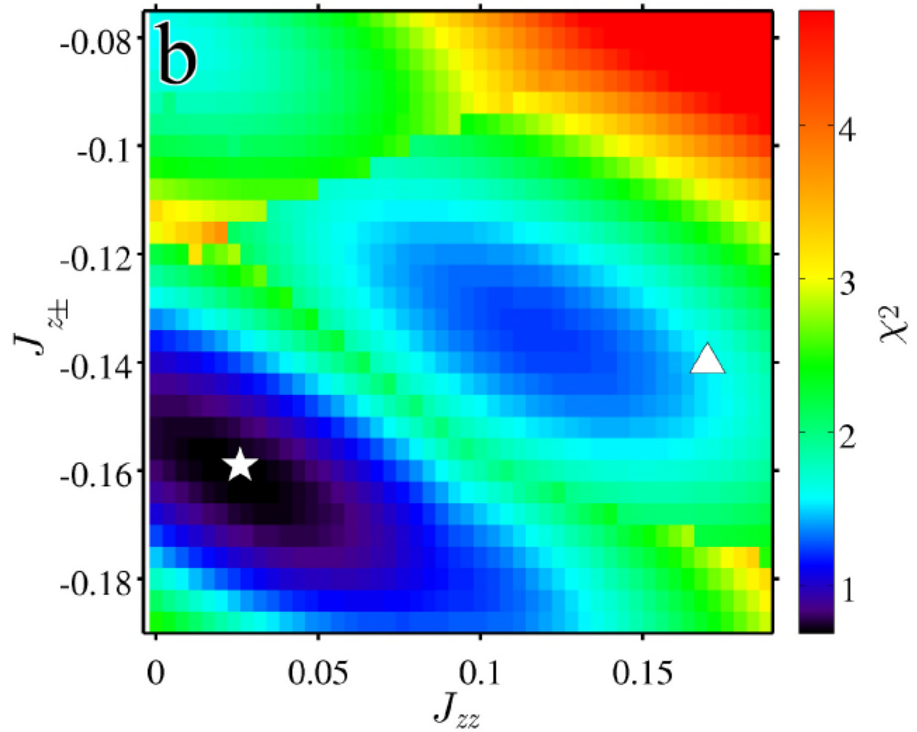


Parameterization of dispersions at B=5T //[001]



Convergence of refinement of Hamiltonian parameters

This work	*Ross et al (2011)*
$J_{zz} = 0.026(3)$	$J_{zz} = 0.17 \pm 0.04,$
$J_{\pm} = 0.074(2)$	$J_{\pm} = 0.05 \pm 0.01,$
$J_{\pm\pm} = 0.048(2)$	$J_{\pm\pm} = 0.05 \pm 0.01,$
$J_{z\pm} = -0.159(2)$	$J_{z\pm} = -0.14 \pm 0.01$
$g_{\parallel} = 2.14(3)$	$g_z = 1.80$
$g_{\perp} = 4.17(2).$	$g_{xy} = 4.32$
$g_{\perp} / g_{\parallel} = 1.95(3)$	$g_{xy}/g_z = 2.4.$ fixed

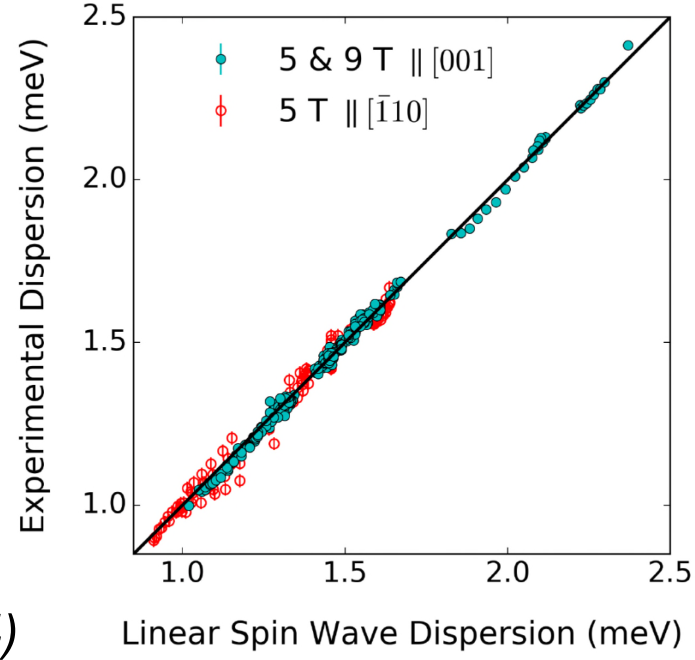


- unique solution explains **all existing dispersions data (2 field orientations) + saturation magnetization**

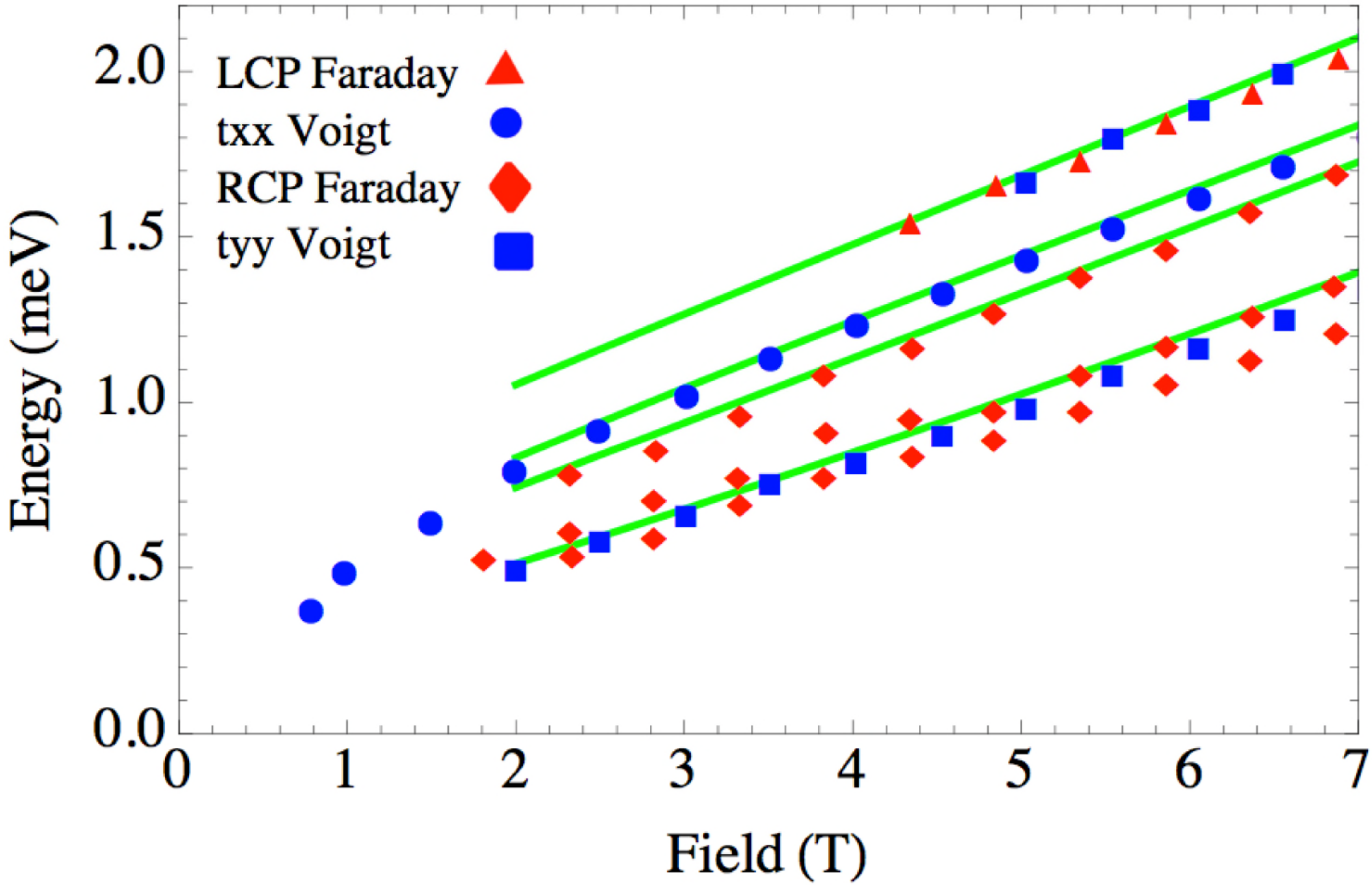
- refined g-factor ratio agrees with recent crystal field parameterization 1.96 +/- 0.13
  J. Gaudet et al (2015)

- agrees with parameterization of diffuse scattering at 0.4 K Roberts et al PRB (2015)
  DiLong et al (2014)

- agrees with THz data (energies &polarization)



Field-dependence of THz data in (001) field (q=0 excitations)



- revised parameters also fit well the THz data in [001] field (energy & polarization)

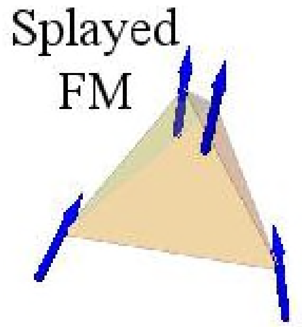
DiLong et al (2014)

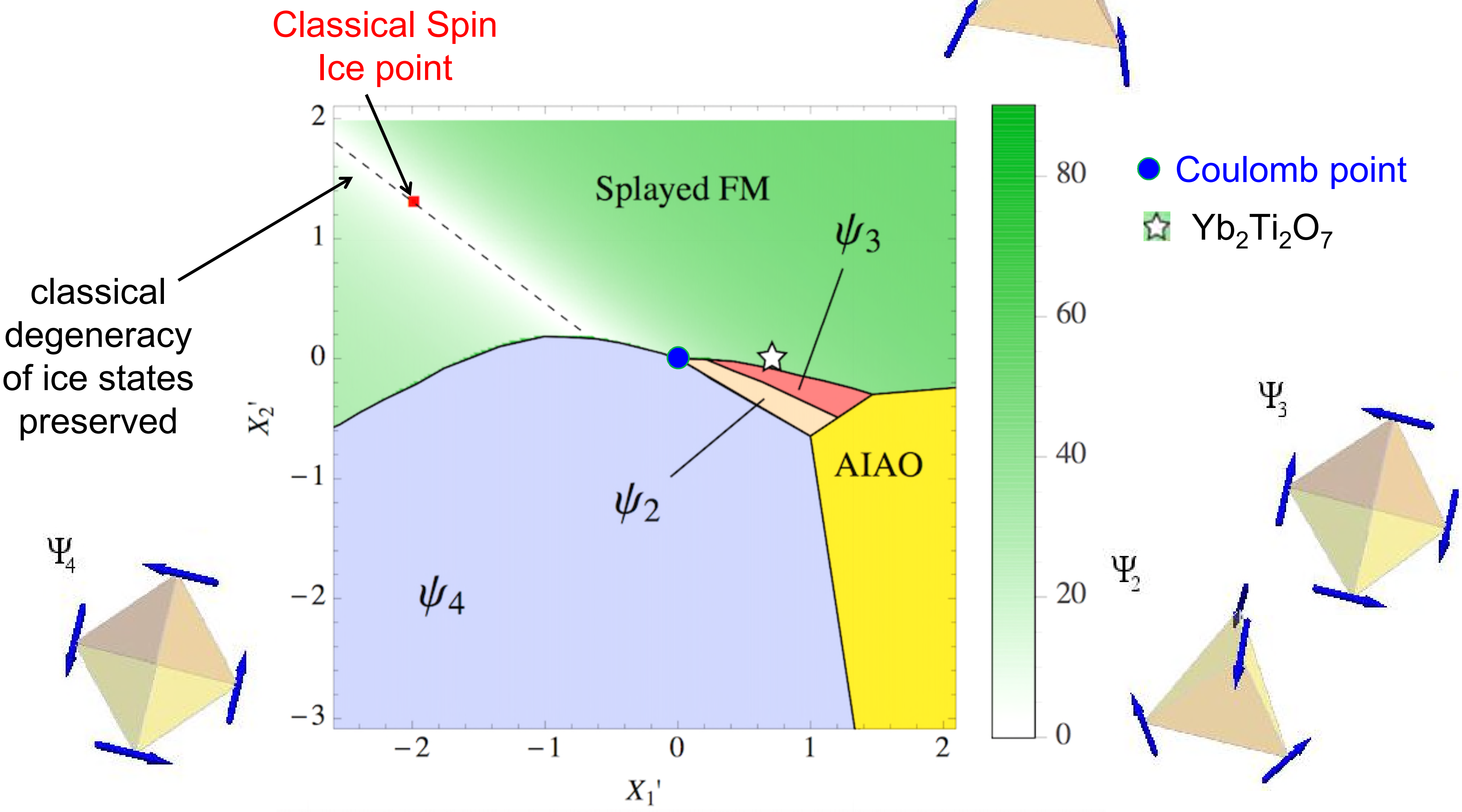
Semi-classical Phase Diagram

- revised parameters put system **almost on phase boundary** Splayed FM – AFM $\Psi_{2,3}$

=> strongly frustrated

- mean-field T_C ~ 3 K >> actual Tc = 0.21-26 K





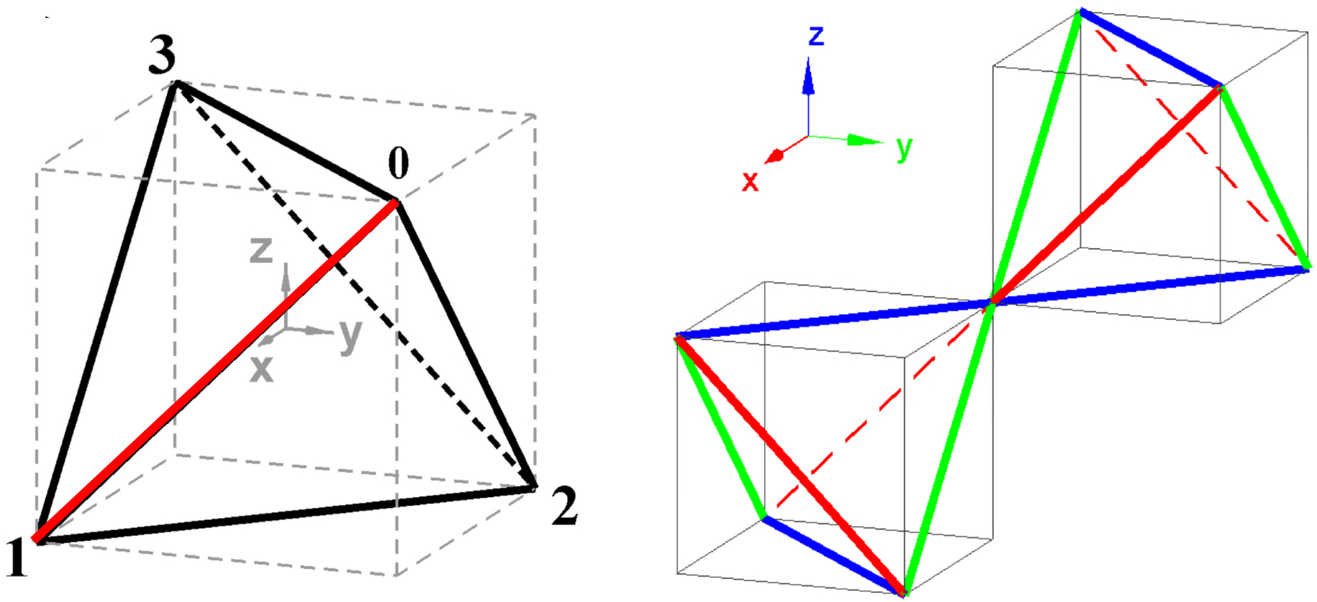
Exchange Hamiltonian – global cubic axes

$$\mathbf{J}_{01} = \begin{pmatrix} J_2 & J_4 & J_4 \\ -J_4 & J_1 & J_3 \\ -J_4 & J_3 & J_1 \end{pmatrix}$$

$[J_1\, J_2\, J_3\, J_4] = [\ -0.028 \quad \mathbf{-0.326} \quad \mathbf{-0.272} \quad 0.049]$ meV

FM Ising coupling $J_2\, S_x S_x$ ("Kitaev"-type) *K*-term

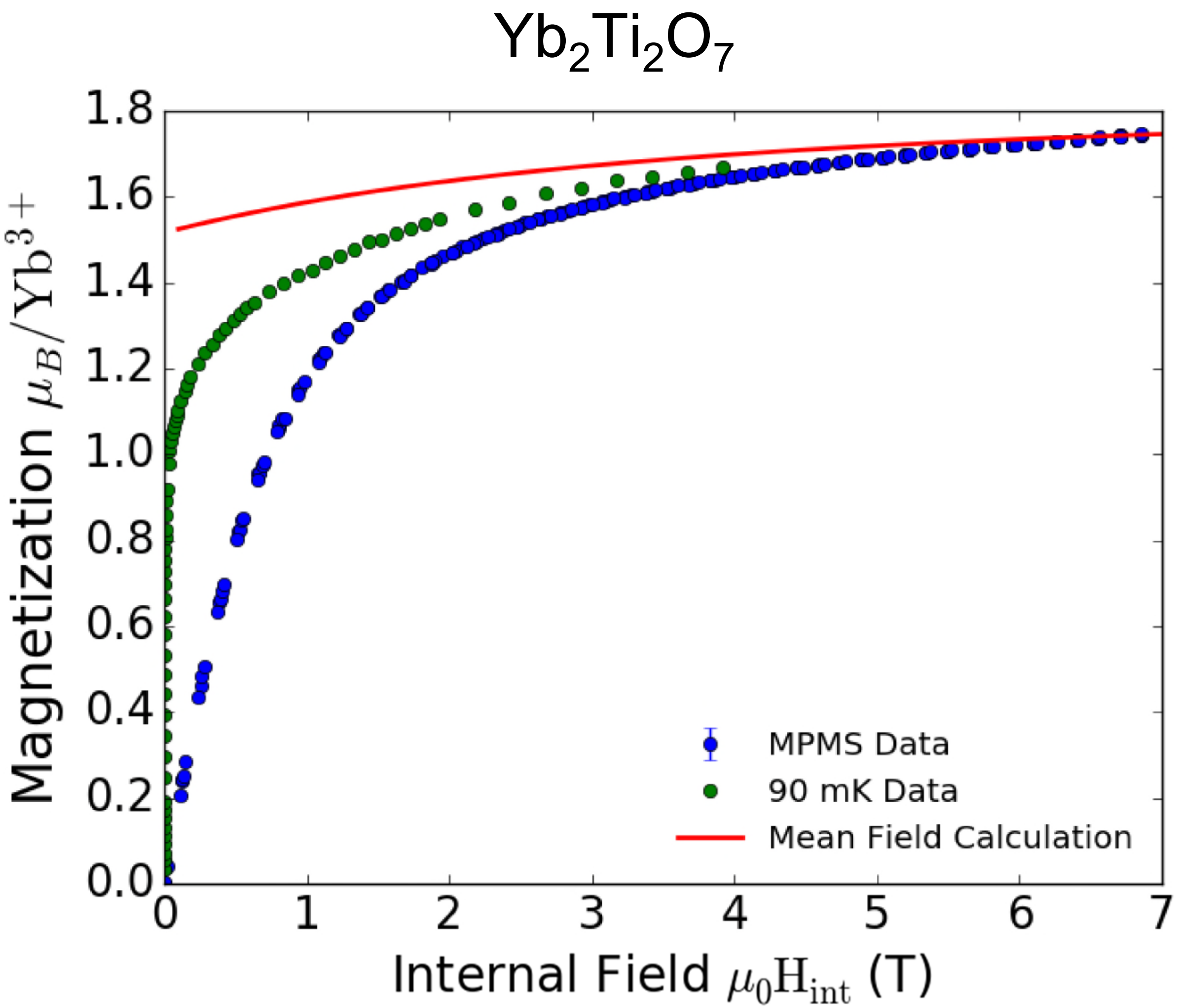
+ "pseudo-dipolar" symmetric exchange $J_3(\, S_y S_z + S_z S_y)$ Γ-term



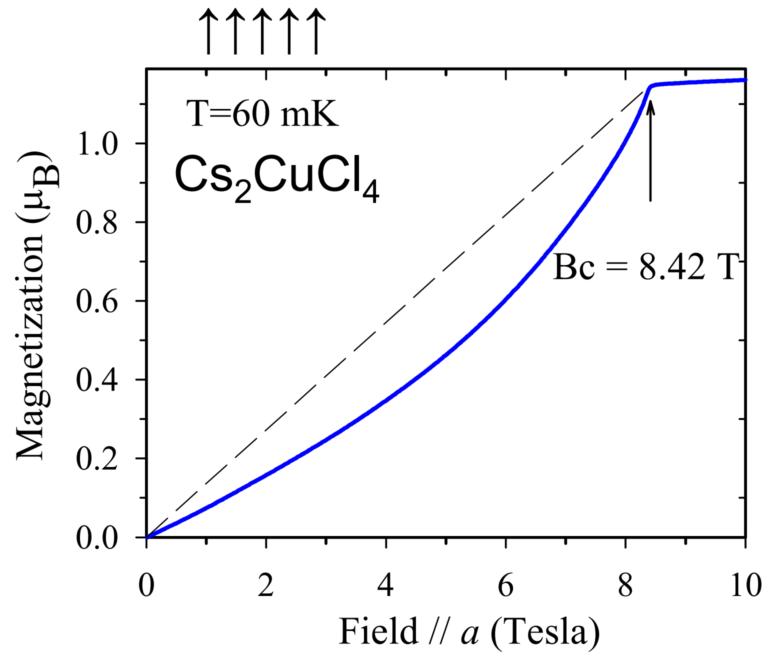
- for **S**//z 4/6 Kitaev bonds + 6 Γ bonds create quantum dynamics

=> **strongly frustrated quantum Hamiltonian**

Longitudinal magnetization in field // [001]



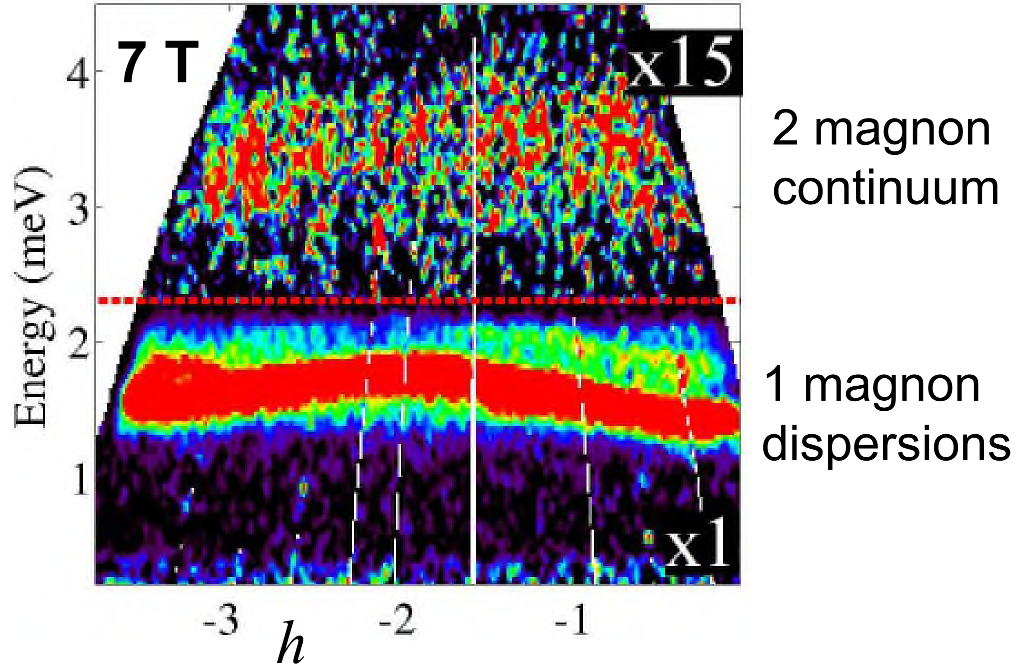
contrast with saturation plateau (exact eigenstate)

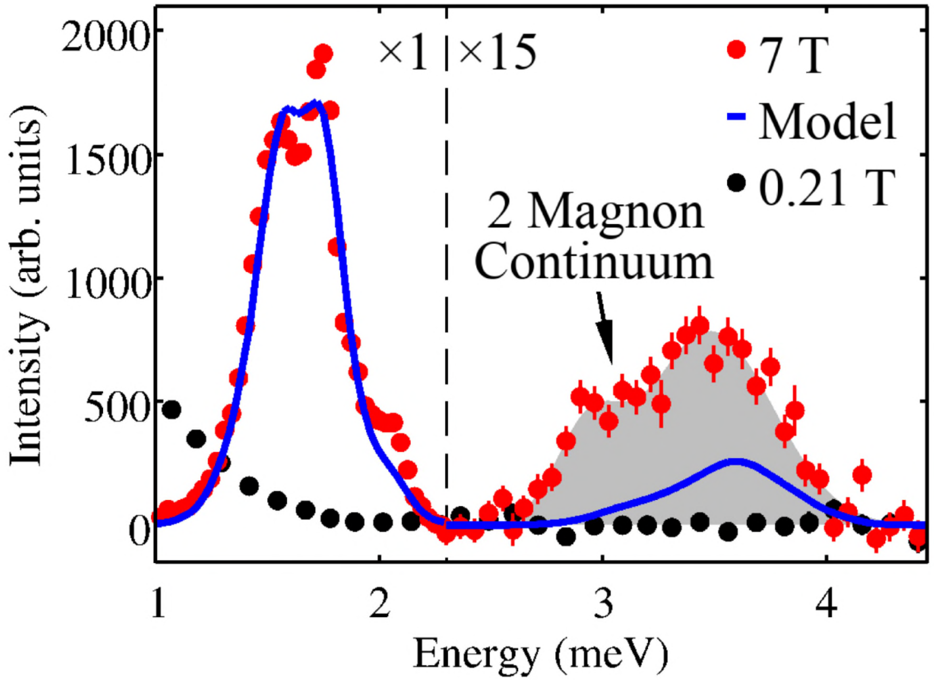


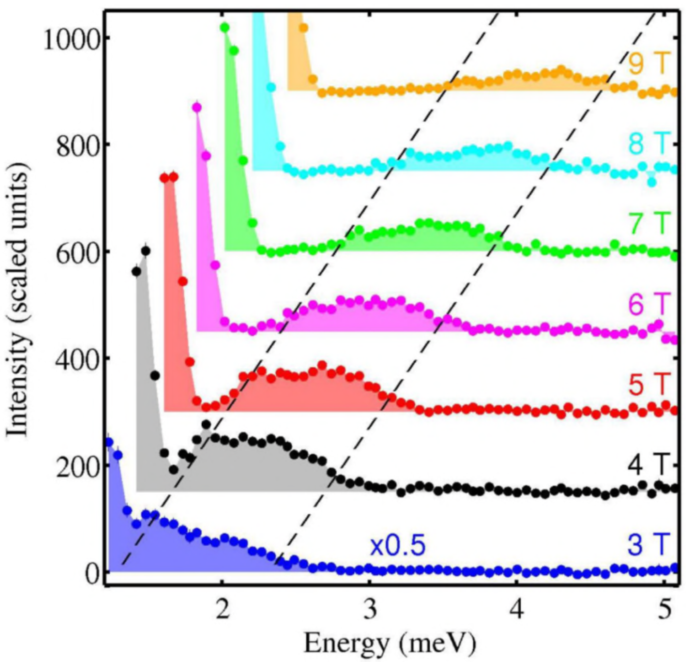
- saturation reached asymptotically (not a plateau), S_z not conserved, characteristic of anisotropic interactions $[\mathcal{H}, S_z] \neq 0$
- **quantum fluctuations suppressed gradually** as field increases
- expect two-magnon continuum in addition to one-magnon in INS

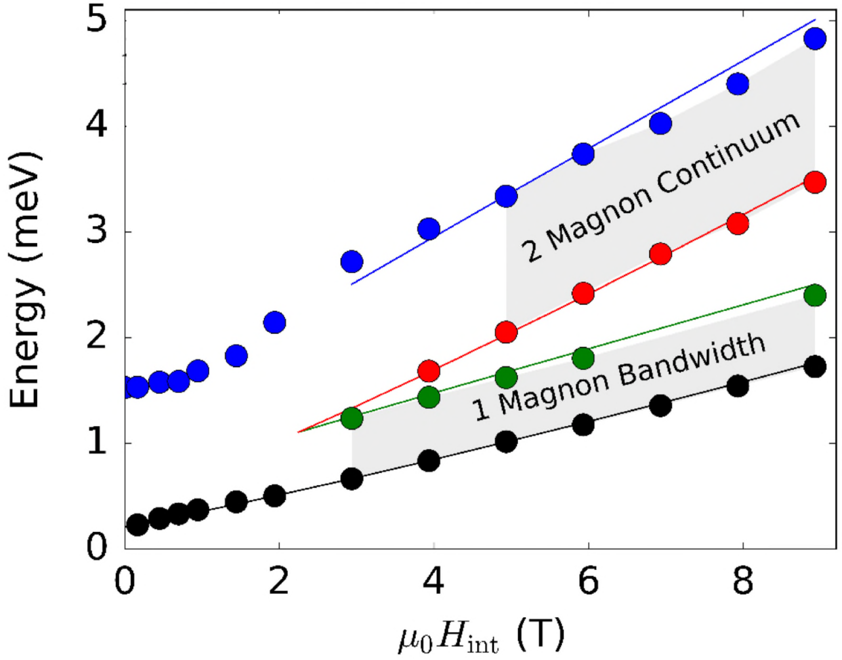
Two-magnon scattering continuum

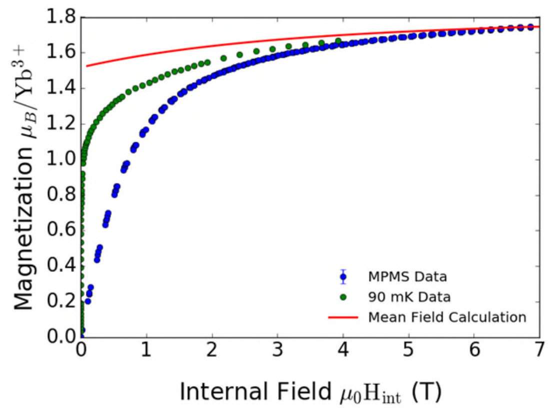
- at higher energies see additional weak continuum scattering (1-2% of one-magnon weight)



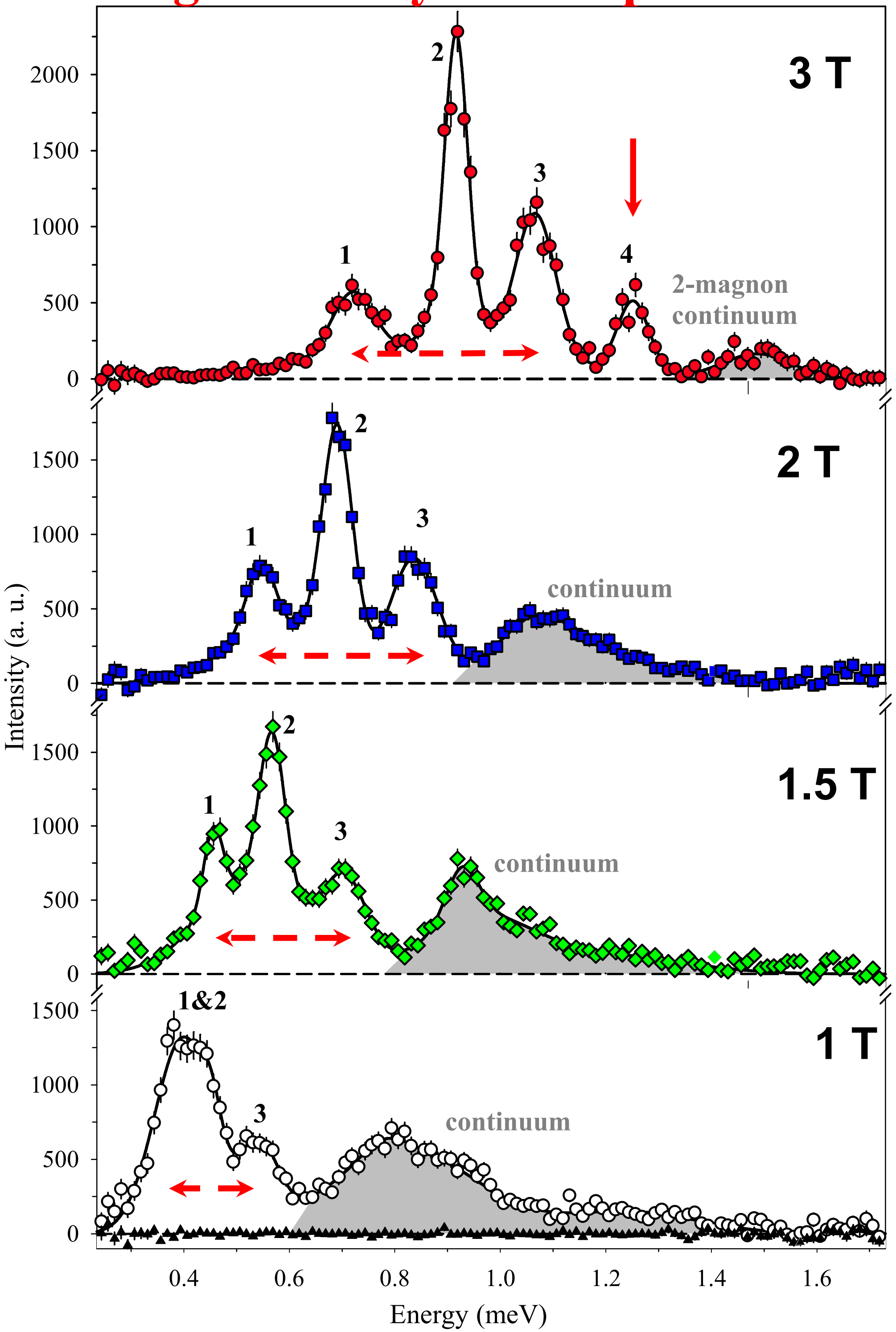


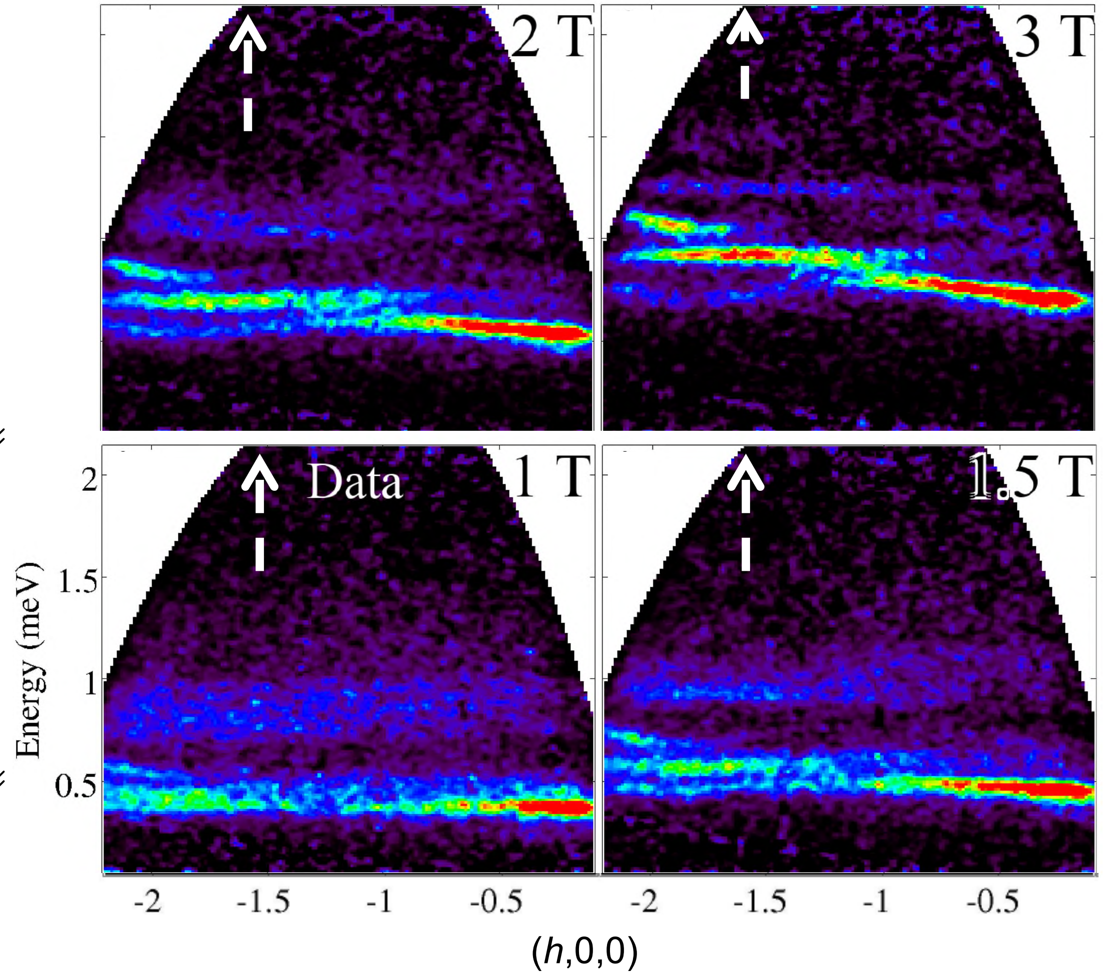






Magnon decay and dispersion renormalization





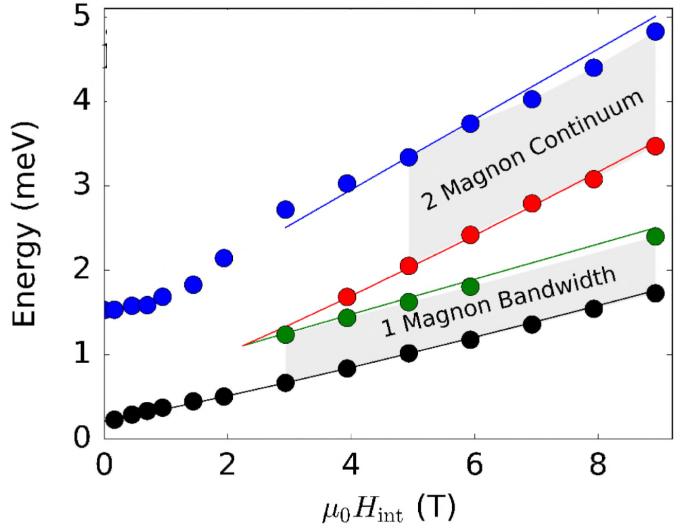
- 2-magnon excitations become progressively stronger with weight comparable to 1-magnon at low field

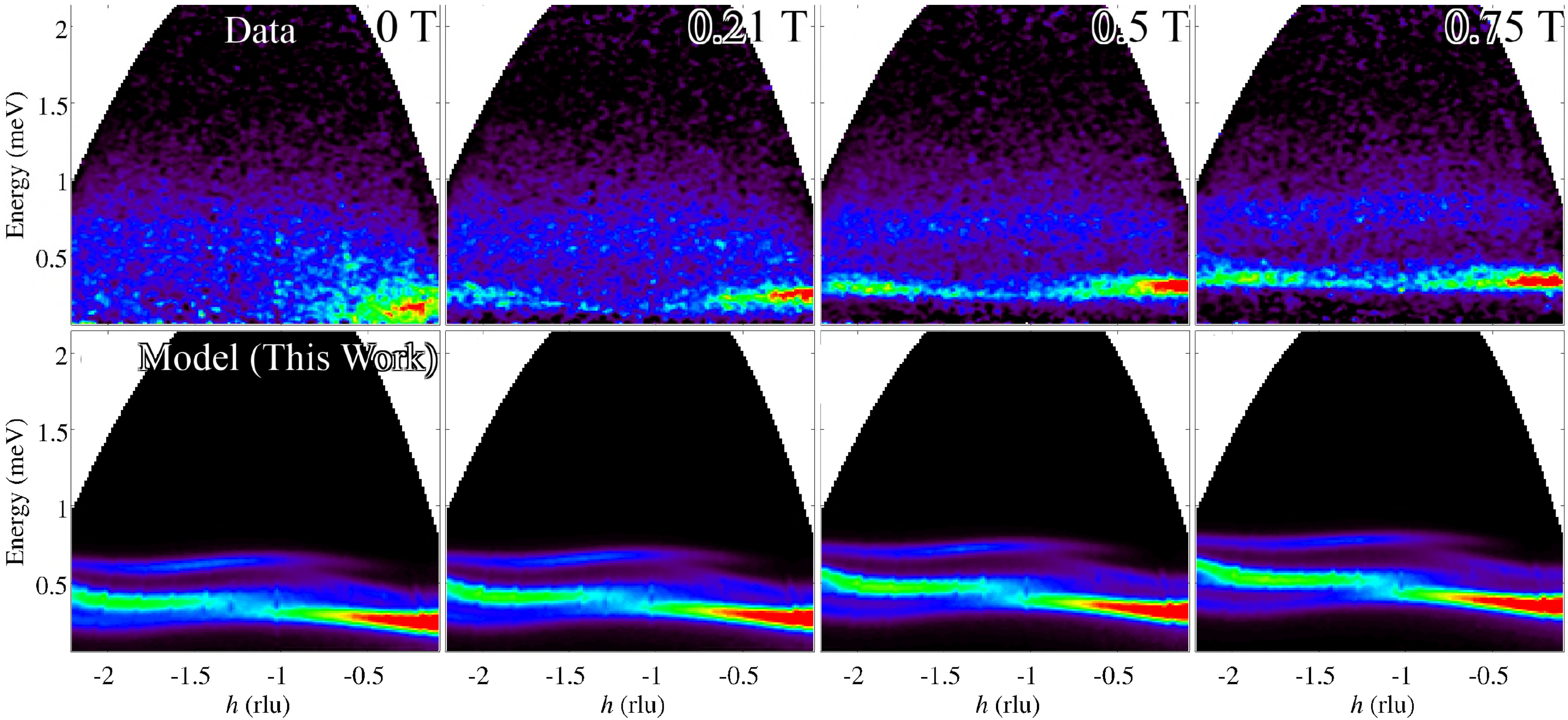
- 1-magnon decay and dispersion renormalization -> strong coupling of 1 & 2 magnon states

- suppression of magnon bandwidth due to increased quantum fluctuations at low field

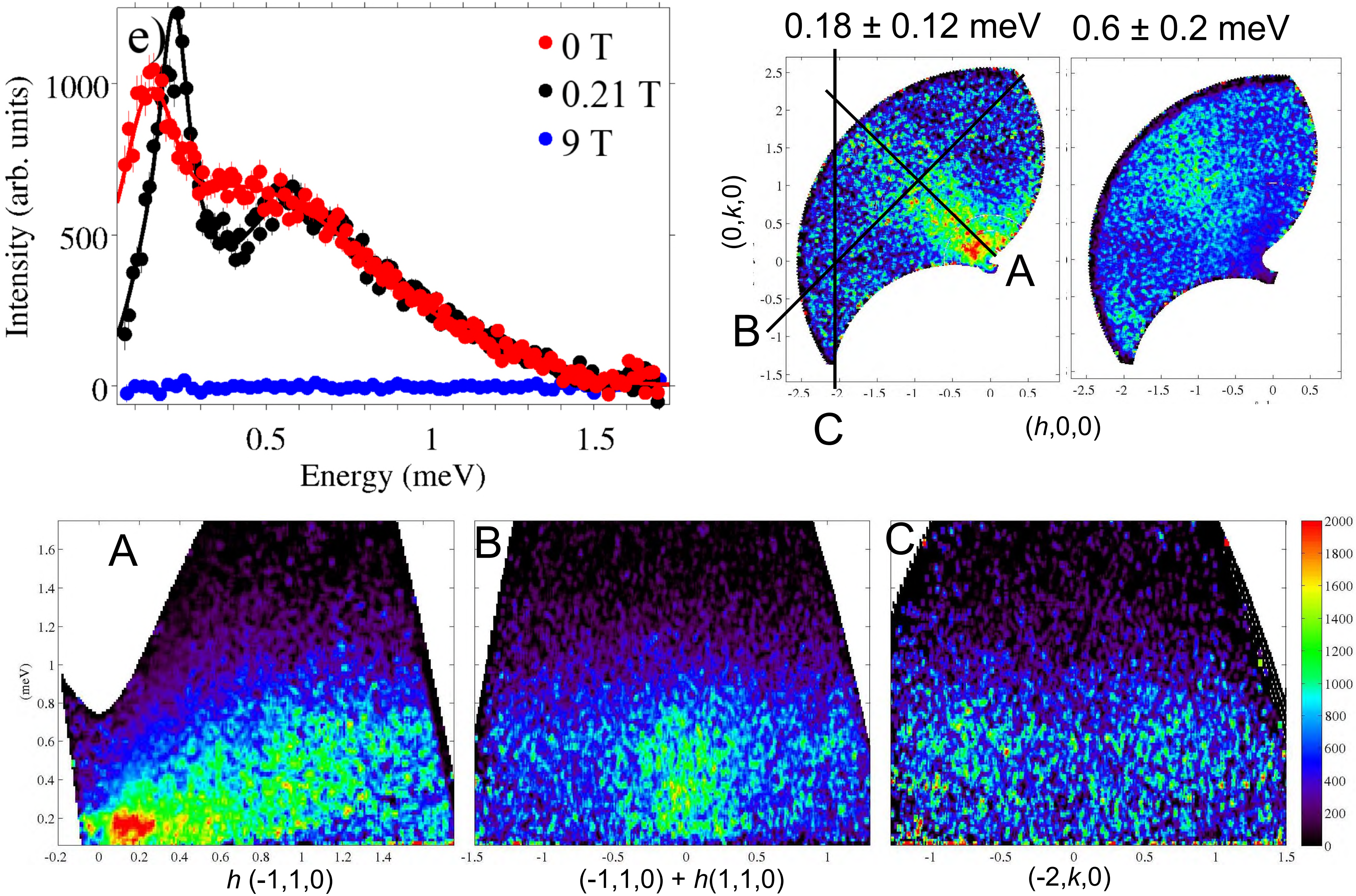
Spectrum at low field

- at 0.75 T single sharp mode observed, almost non-dispersive (localized) + gapped higher energy broad continuum
- at 0.21 T low energy sharp mode + extended continuum
- at 0 T extended continuum over the full predicted spin-wave range





Zero field excitations – continuum scattering with intensity modulations

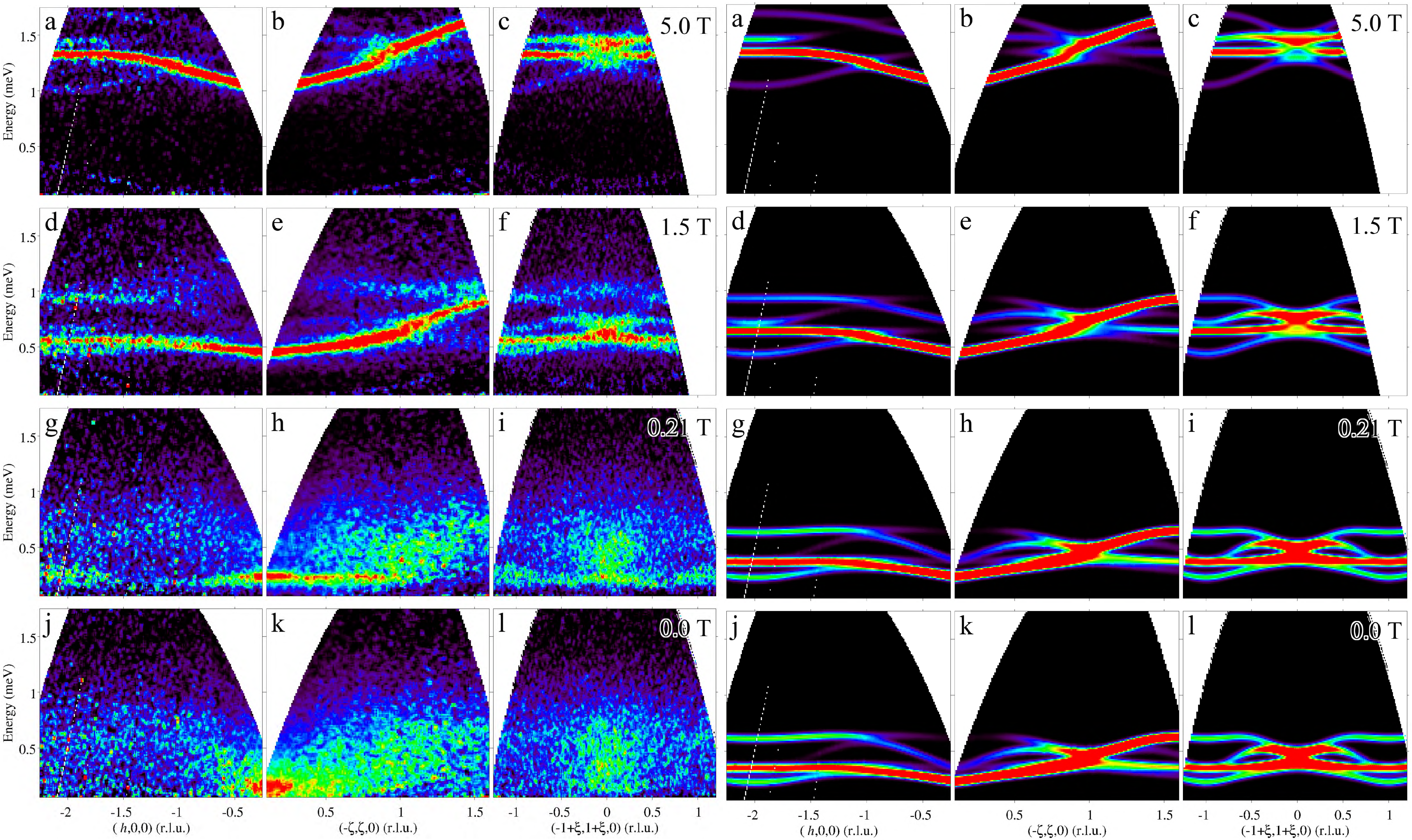


- extended continuum scattering over with intensity modulations, largest along (-1,1,0) diagonals

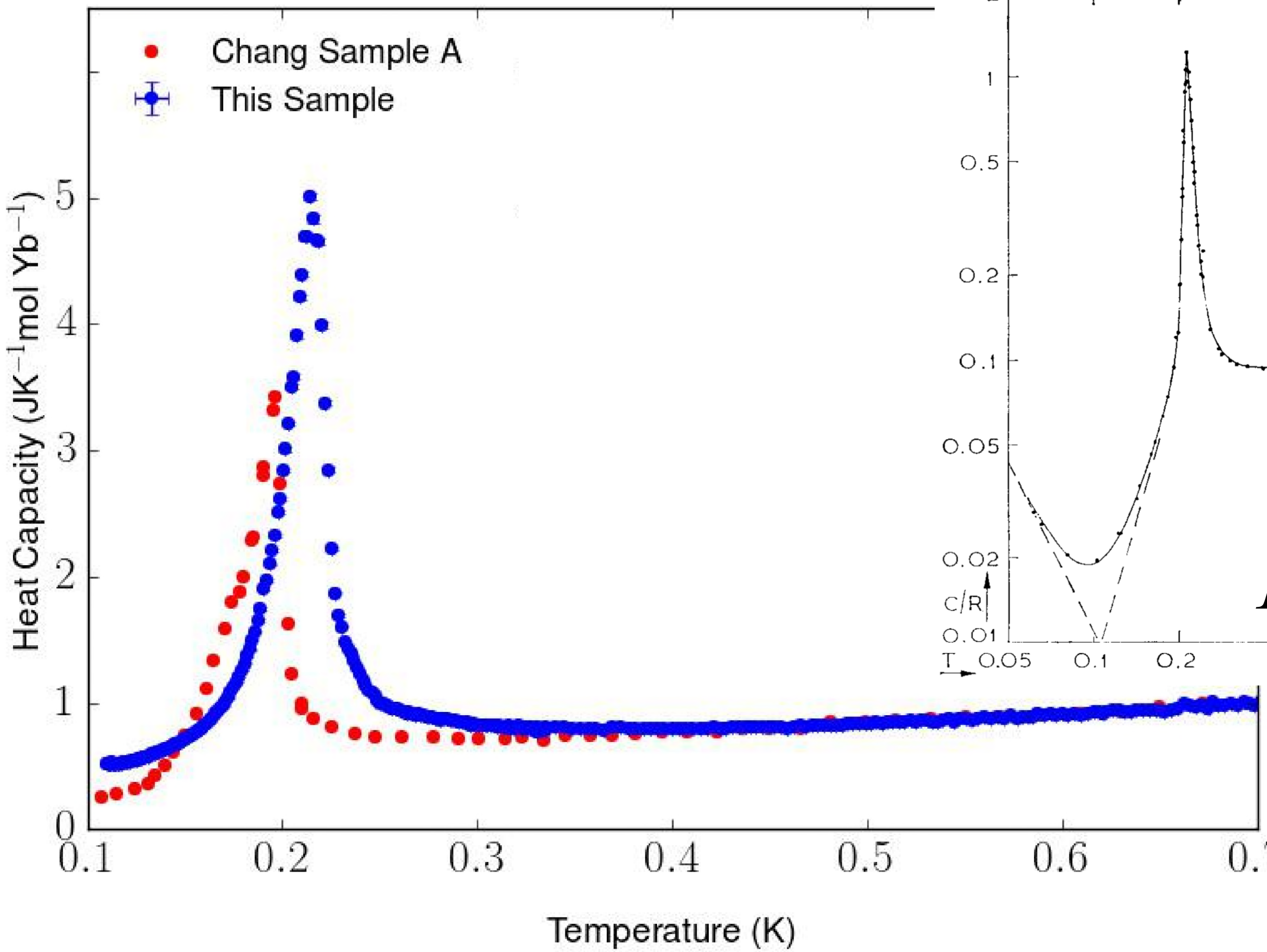
Spin dynamics as a function of field

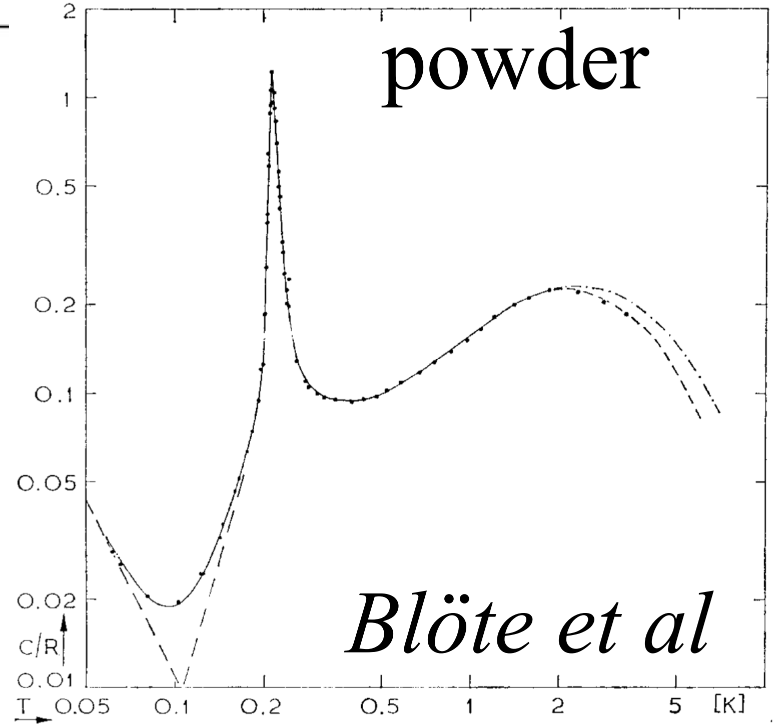
Data 0.15 K

Spin-wave Theory



$Yb_2Ti_2O_7$ single crystal specific heat

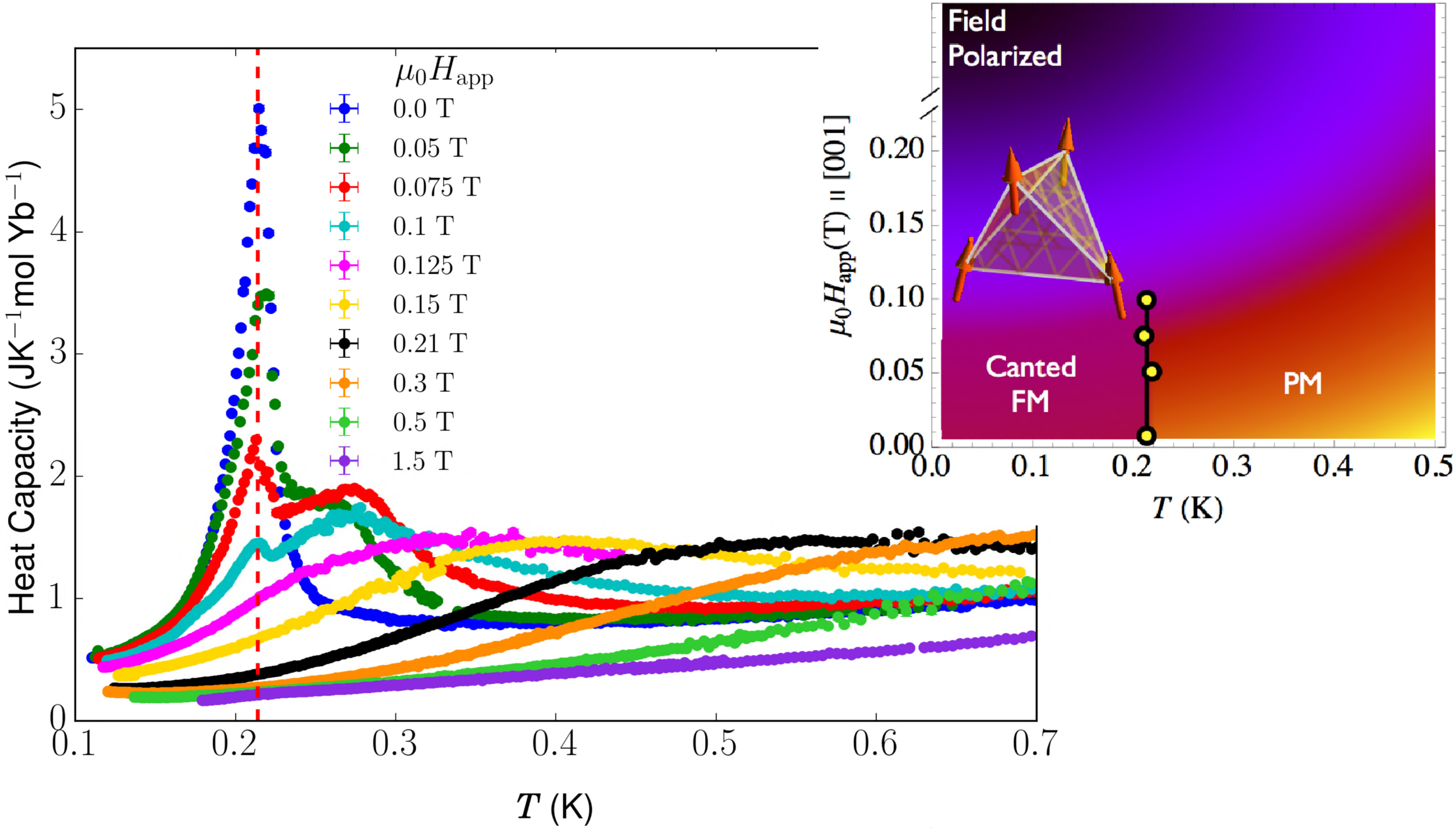




- our image furnace "slow"-grown & annealed single crystal shows a single, sharp specific heat peak at 0.214 K -> similar in behaviour to high purity limit (stoichiometric powders have 1st order transition 0.24-0.26 K)

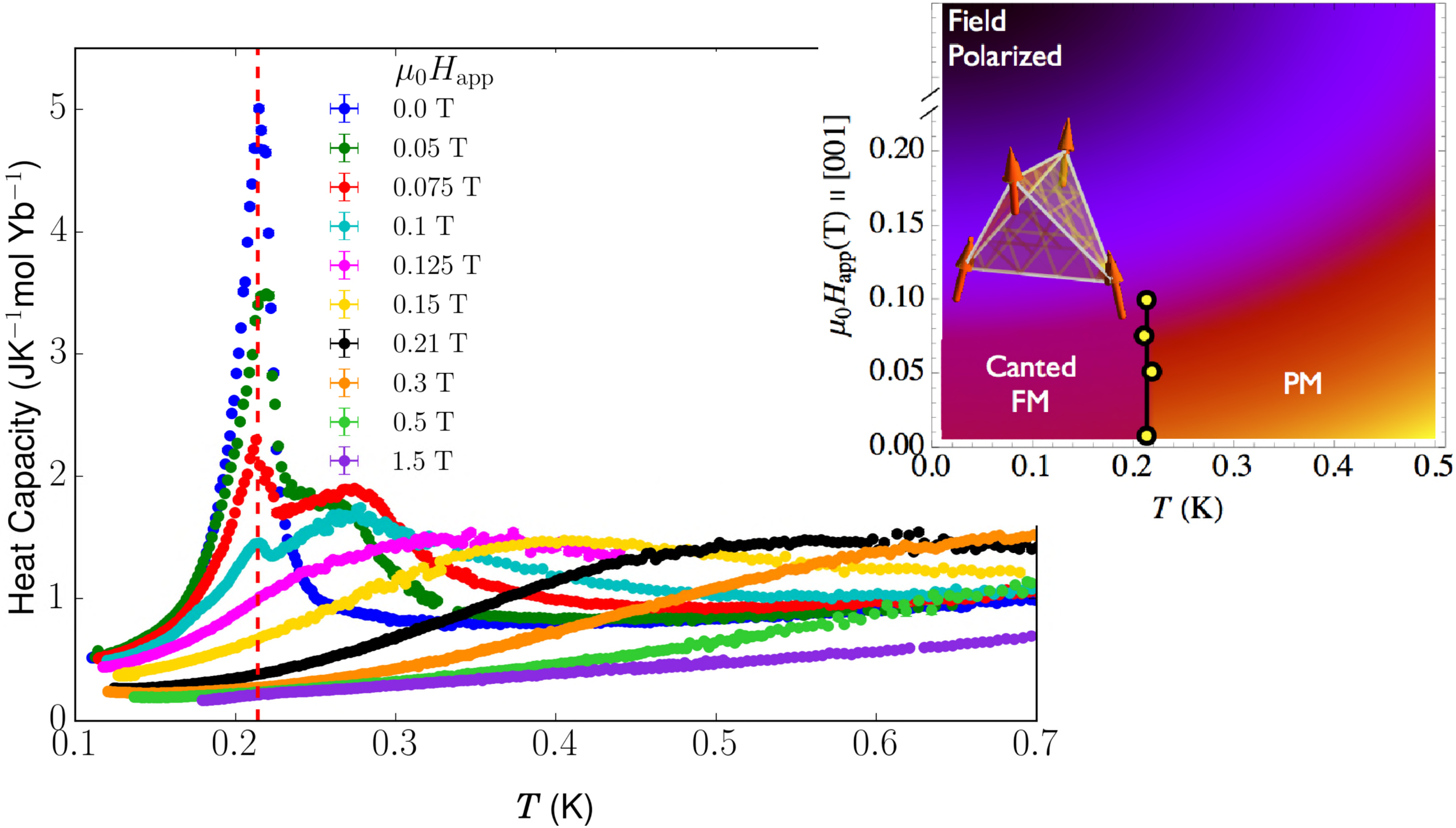
Chang et al (2012) *Arpino …McQueen (2017)*

Heat capacity in magnetic field // [001]



- sharp anomaly rapidly suppressed by small field 0.1 T
- in *finite* [001] field Canted FM, Paramagnet and Field Polarized have the SAME symmetry; *cross-over* from Canted FM -> Field Polarized, no sharp phase transition

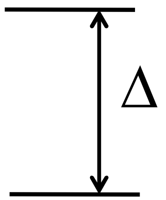
Heat capacity in magnetic field // [001]



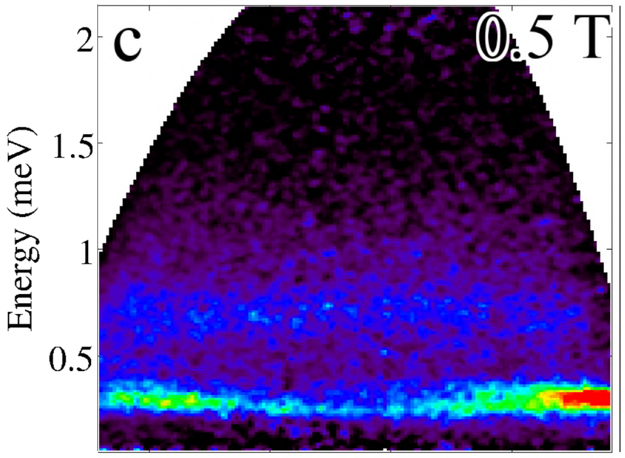
- broad Schottky anomaly appears at finite B and moves up to higher T upon increasing B -> rapid loss of low-energy spectral weight, gap increases rapidly in field

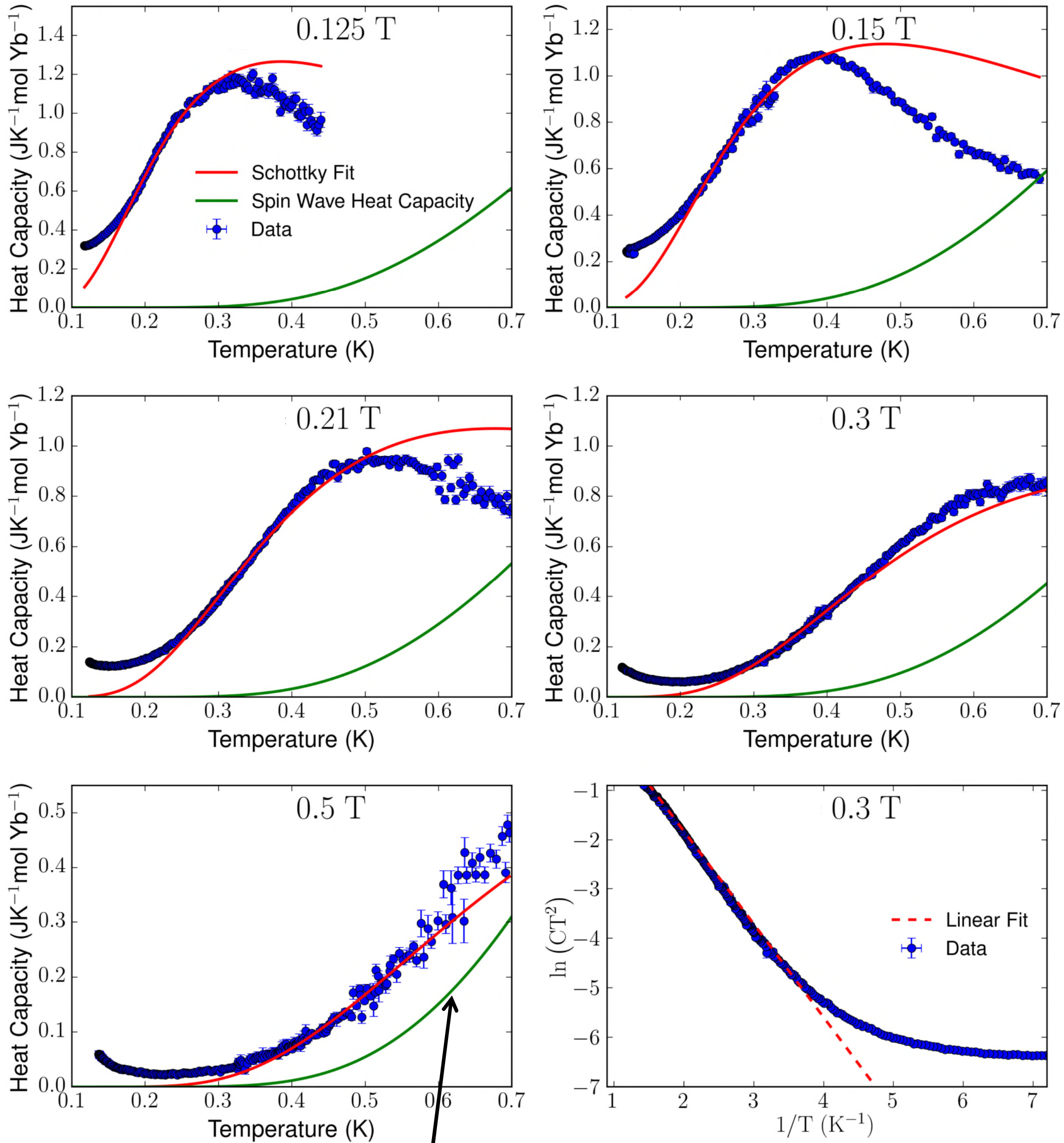
Heat capacity in magnetic field // [001]

- parameterize with form for gapped 2-level system



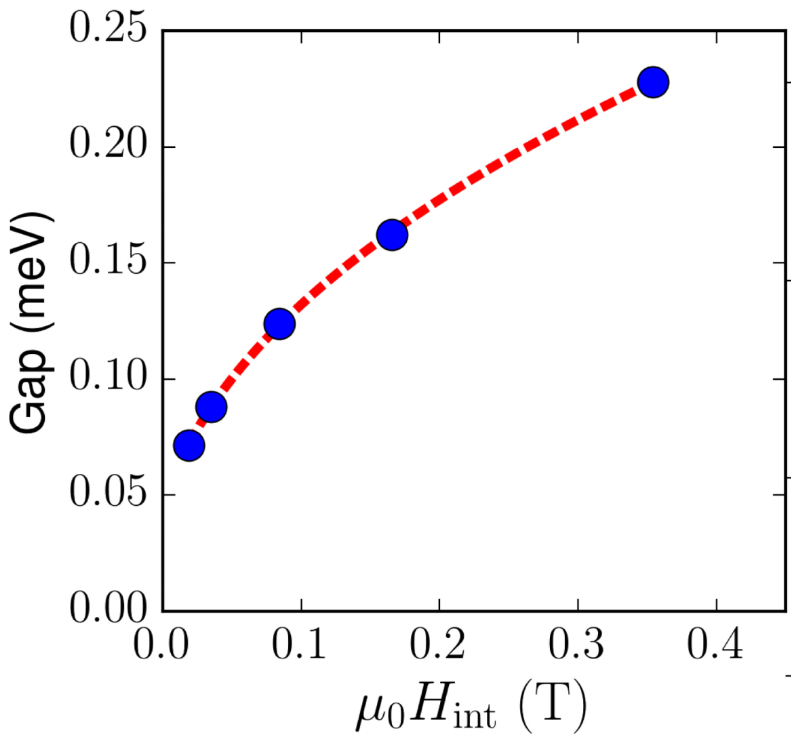
$$C(T) = R\left(\frac{\Delta}{k_{\mathrm{B}}T}\right)^2 \frac{e^{-\Delta/k_{\mathrm{B}}T}}{\left(1+e^{-\Delta/k_B T}\right)^2}$$

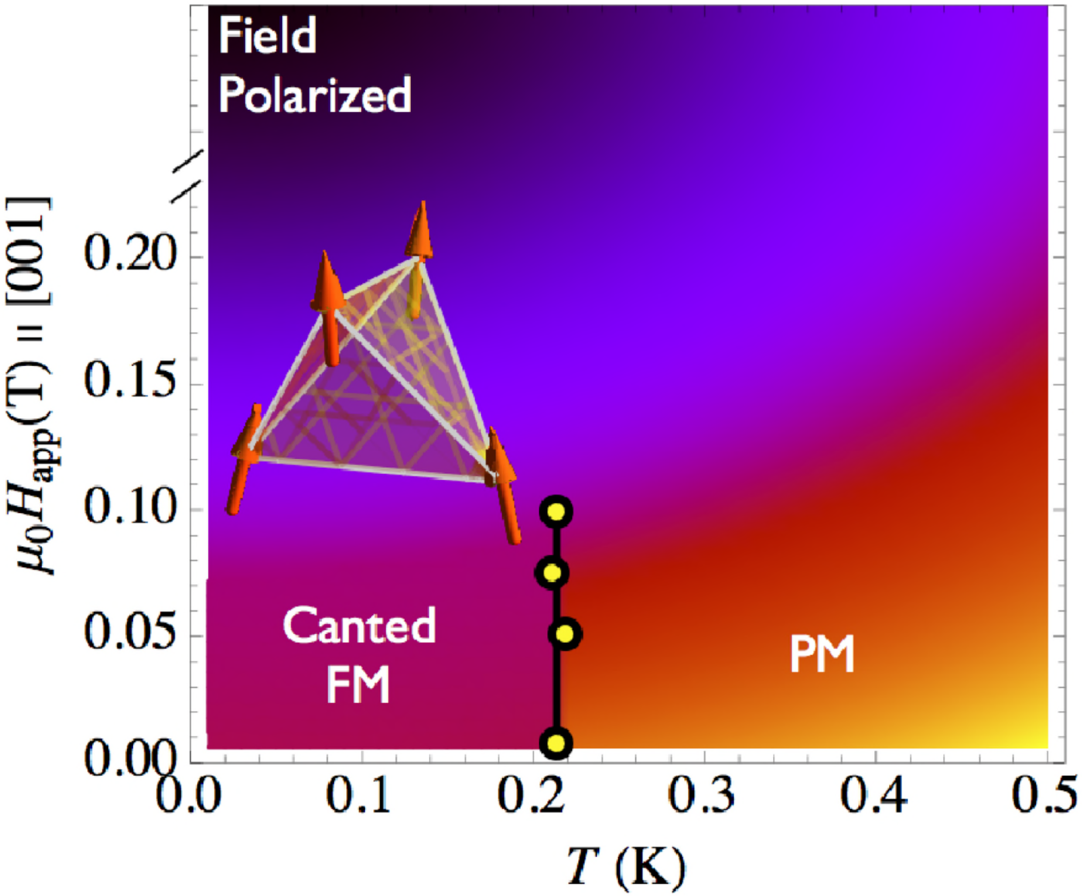


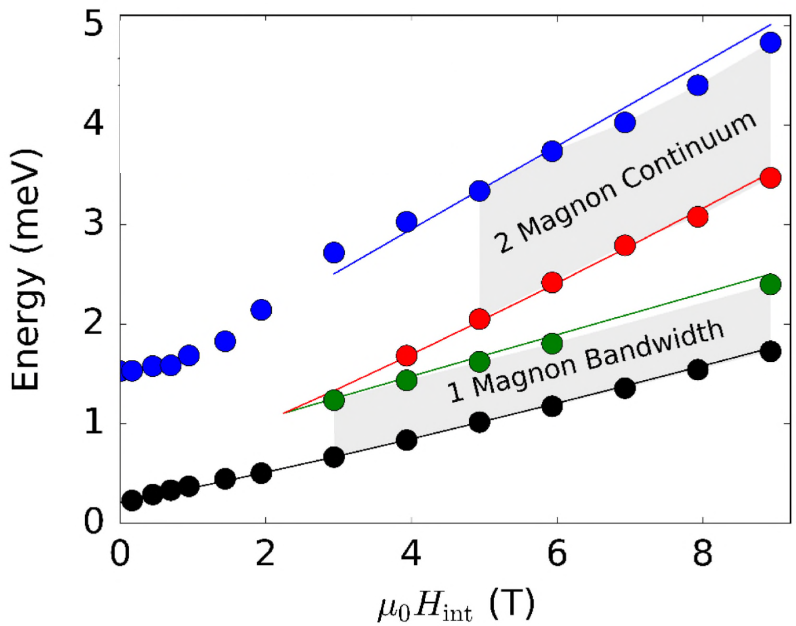


- spin-wave prediction becomes better at high B

Gap vs magnetic field // [001]



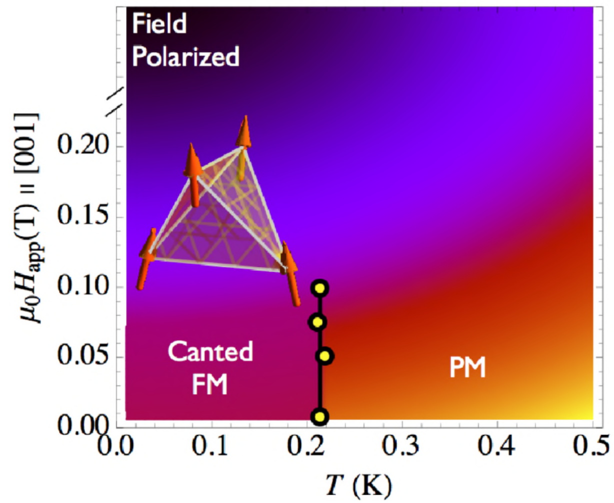




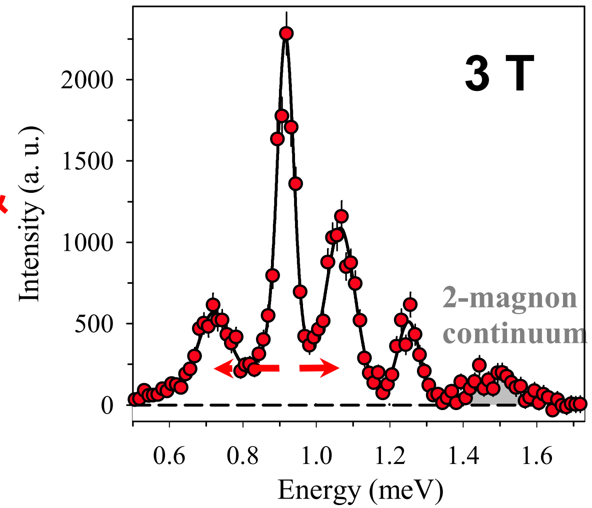
- Gap increases in field
- *cross-over* from Canted FM -> Field Polarized, no sharp phase transition

Conclusions

- in [001] field Canted FM and Field-Polarized are smoothly connected, no phase transition, **gap grows in field**

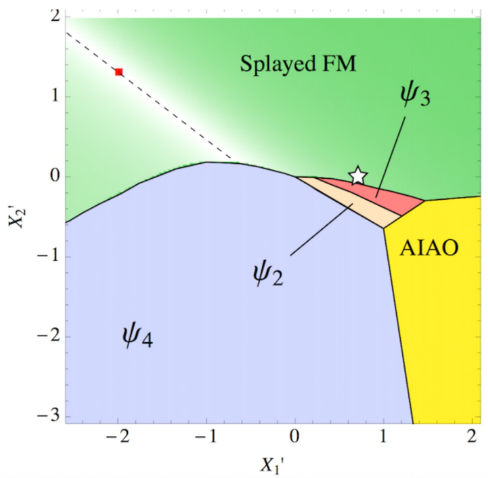


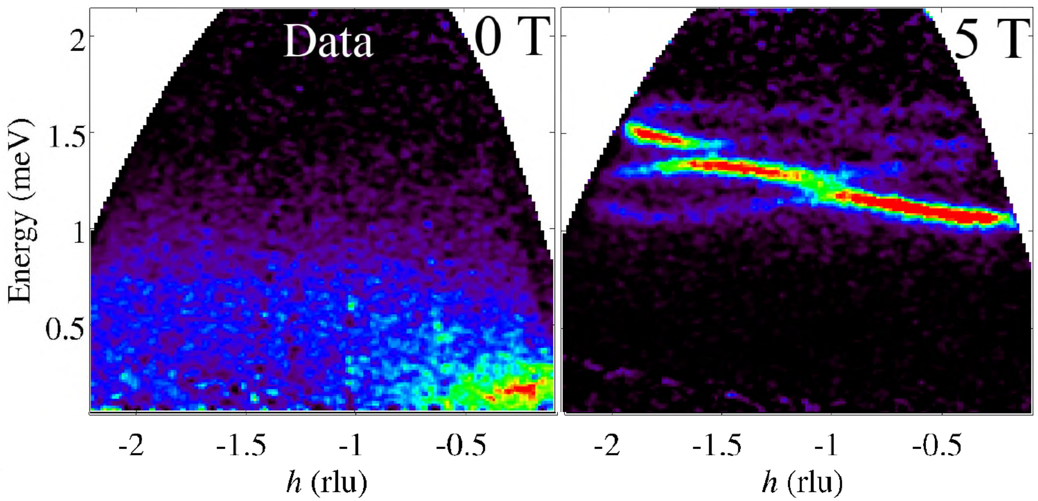
- at high field see sharp magnons + **2-magnon continuum** that grows rapidly upon lowering field, when overlap occurs top **magnon decays** and lower magnon dispersions are strongly renormalized; continuum dominates at zero field

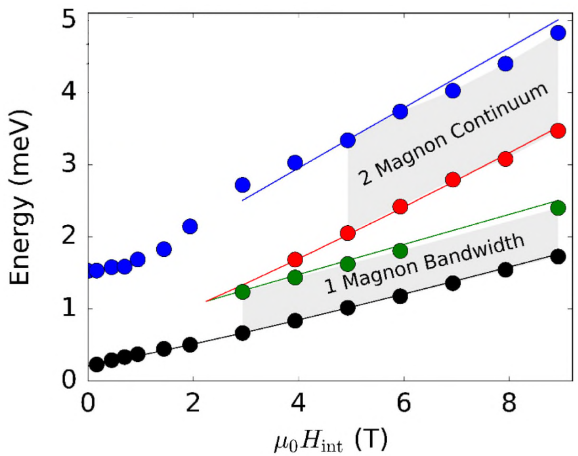


- at high fields sharp magnons captured well by spin waves of nn Hamiltonian with revised parameters, **negligible J_{zz}** & **dominant $J_{z\pm}$** almost on (mean-field) phase boundary between Canted FM and AFM $\Psi_{2,3}$, strongly frustrated

arxiv:1703:04506, 10.1103/PhysRevLett.119.057203

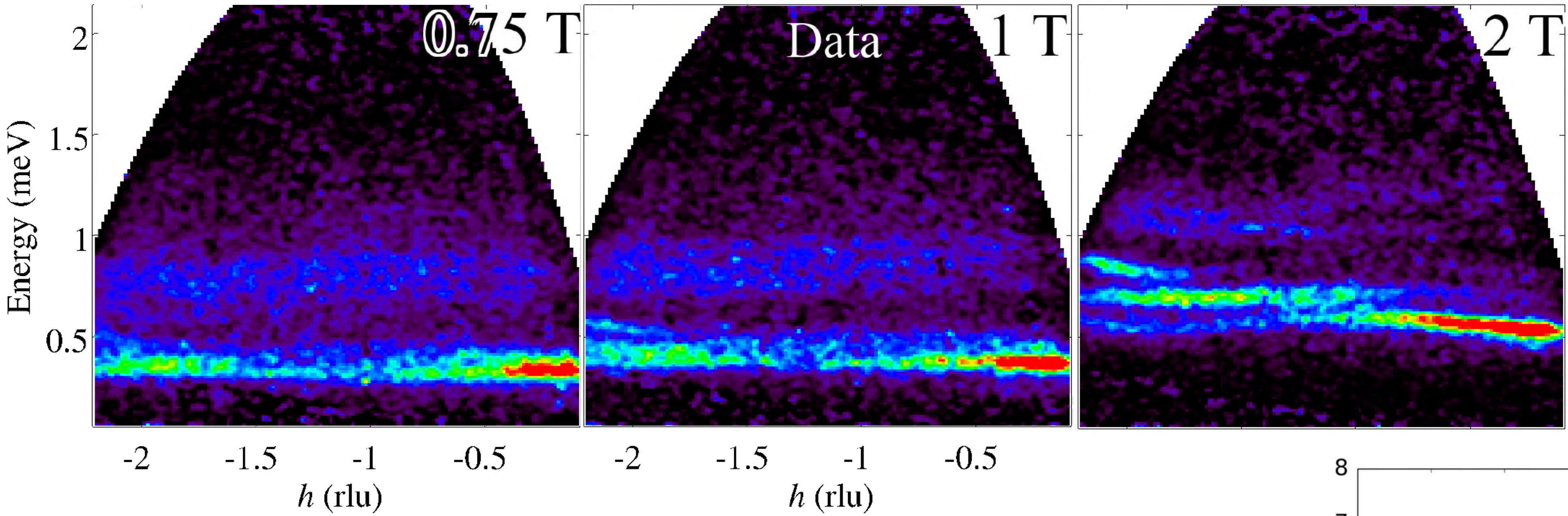




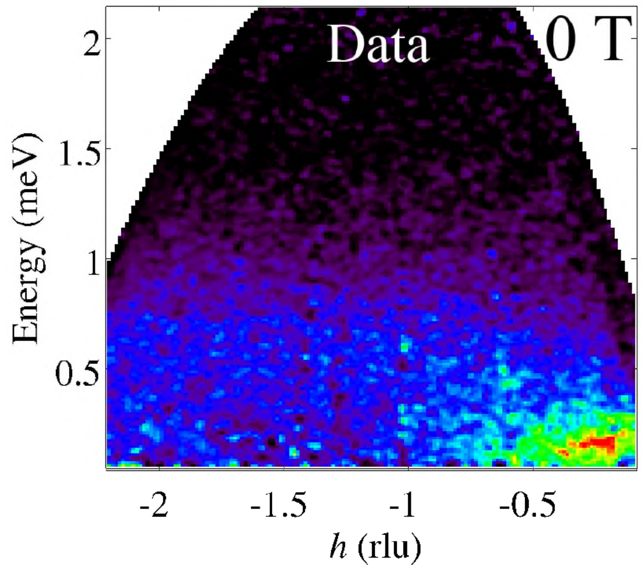


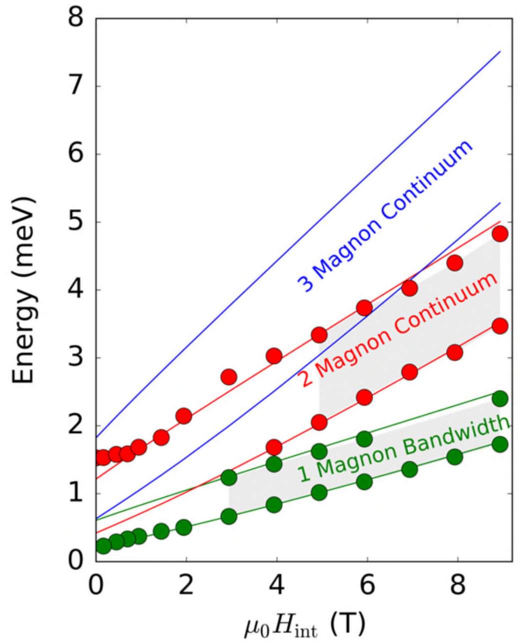
Open questions

- intermediate fields (1 - 2.5 T) how to describe coupling between 1 & 2 magnon excitations & **strong dispersion renormalization**?
- lower field (0.75 T) – physical picture of the **near-flat (non-propagating) mode**?



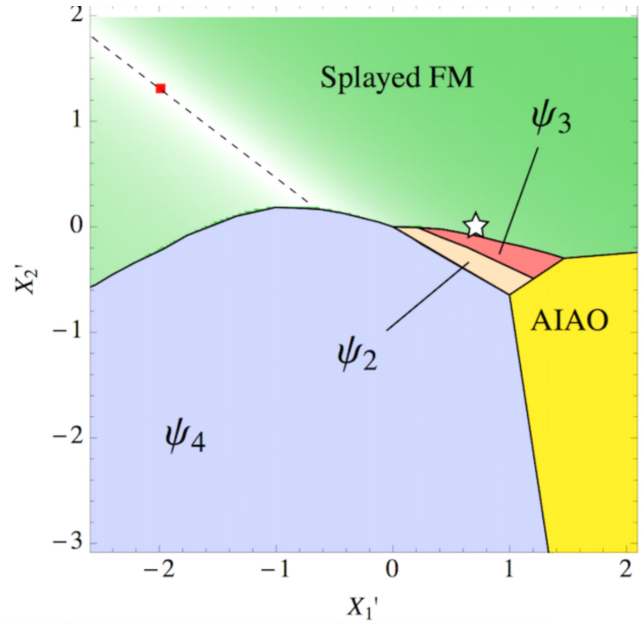
- zero-field, many multi-magnon states overlap, so 1 spin flip mixed with multiple spin flips, is a magnon description still a good starting point, is there a "simple" **physical picture of the continuum**?

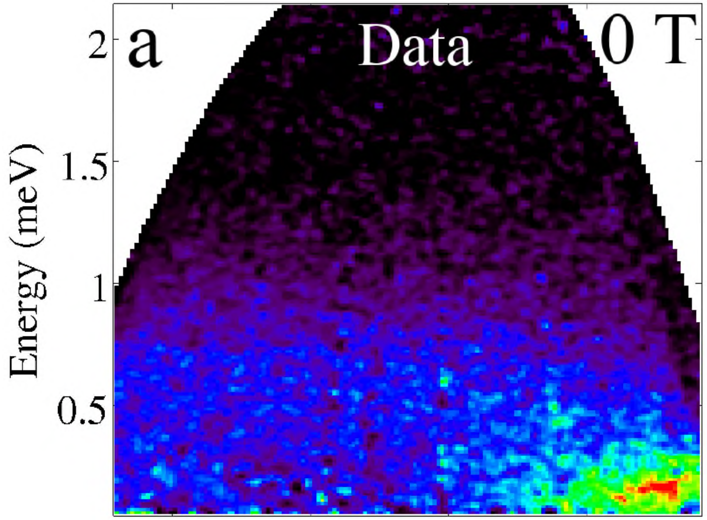




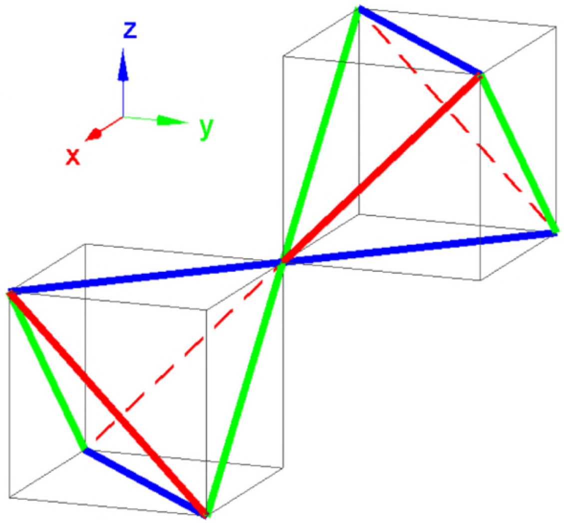
More open questions …

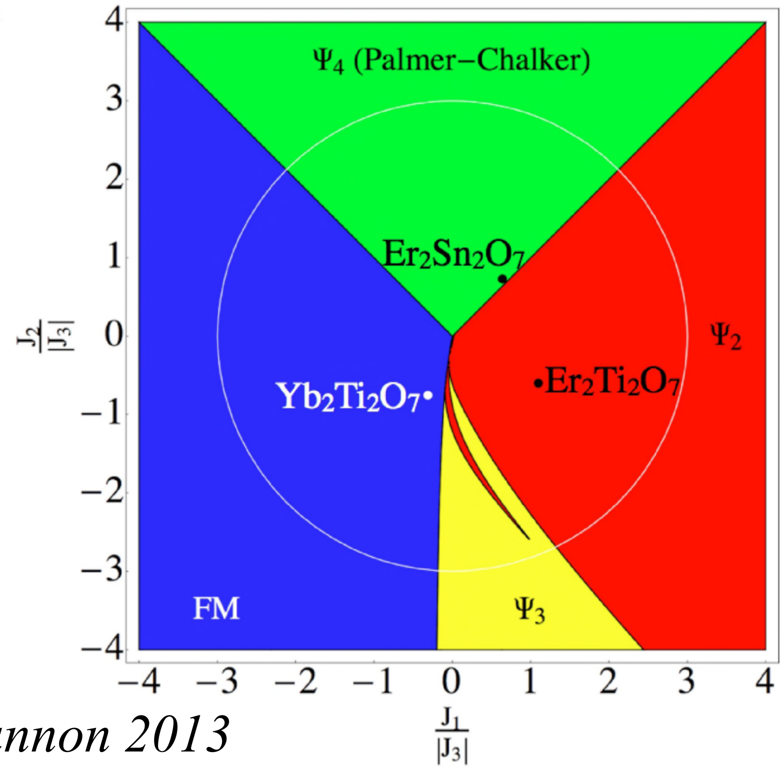
- how is proximity to (semi-classical) FM-AFM phase boundary relevant for the dynamics?





- **strongly quantum Hamiltonian** (Kitaev *K* and Γ, both <0) on pyrochlore lattice, what is quantum phase diagram + dynamics?





limiting cases :

Γ =-1 three phases meet

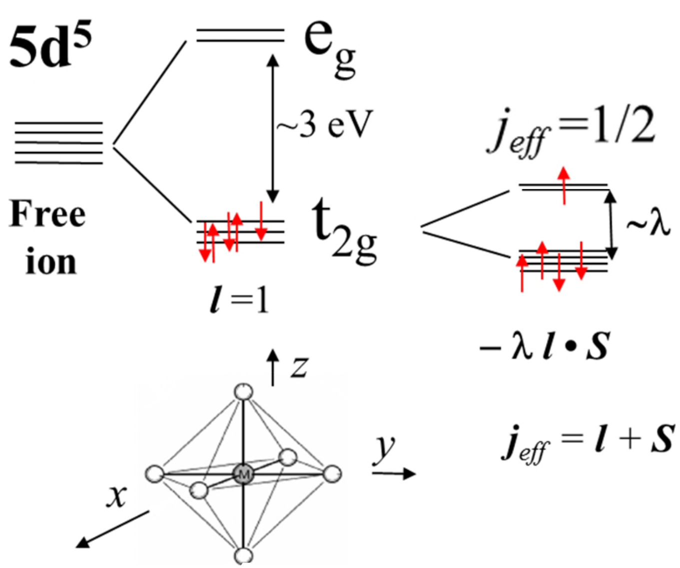
Benton 2016, Yan,Shannon 2013

K = -1 sub-extensive degeneracy

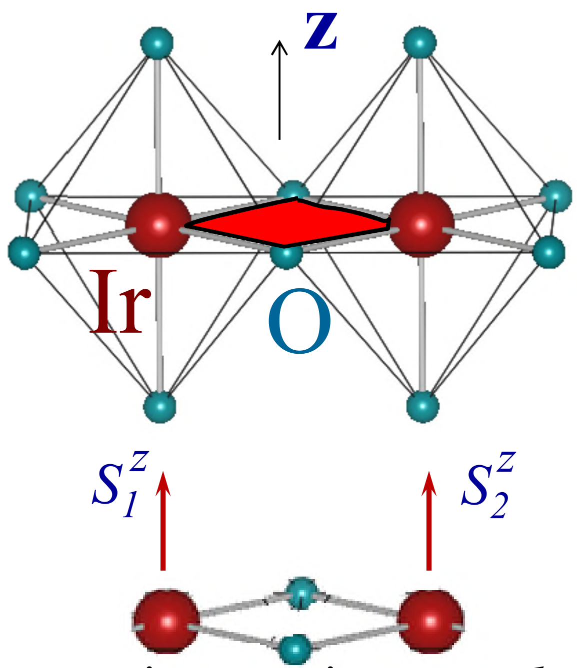
Kimchi, Vishwanath 2014

More open questions …

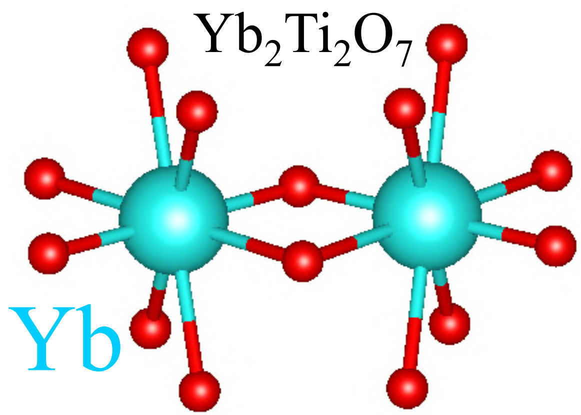
- Kitaev (bond-dependent Ising) exchange originally predicted in case of strong SO coupling for very specific $j_{eff}=1/2$ doublets and exchange paths through two near 90-deg M-O-M bonds, maybe is more general as Yb^{3+} has a different Kramers ground state doublet, to test by *ab initio* calculations why Kitaev exchange appears here?



Jackeli, Khaliullin (2009)



Kitaev axis *normal* to plane of bond



Kitaev axis *in plane* of bond