Title: Ba3Yb2Zn5O11: A model system for anisotropic exchange on the breathing pyrochlore lattice

Date: May 25, 2017 05:00 PM

URL: http://pirsa.org/17050092

Abstract: In this talk we present a study of the â€œbreathingâ€• pyrochlore compound Ba3Yb2Zn5O11. Due to the nearly decoupled nature of its tetrahedral units, this compound serves as an ideal testbed for exploring the nature of anisotropic exchange in a theoretically and experimentally tractable rare-earth system. The relevant low-energy model of the Yb3+ tetrahedra is parametrized by four anisotropic exchange constants and is capable of reproducing the inelastic neutron scattering data, specific heat, and magnetic susceptibility with high fidelity. Using this model, we predict the appearance of an unusual non-Kramers octupolar paramagnet at low temperatures. We further speculate on possible collective, inter-tetrahedron physics of these non-Kramers doublets and discuss applications to about anisotropic exchange in other rare-earth magnets.

The breathing pyrochlore $Ba_3Yb_2Zn_5O_{11}$

A model system for anisotropic exchange

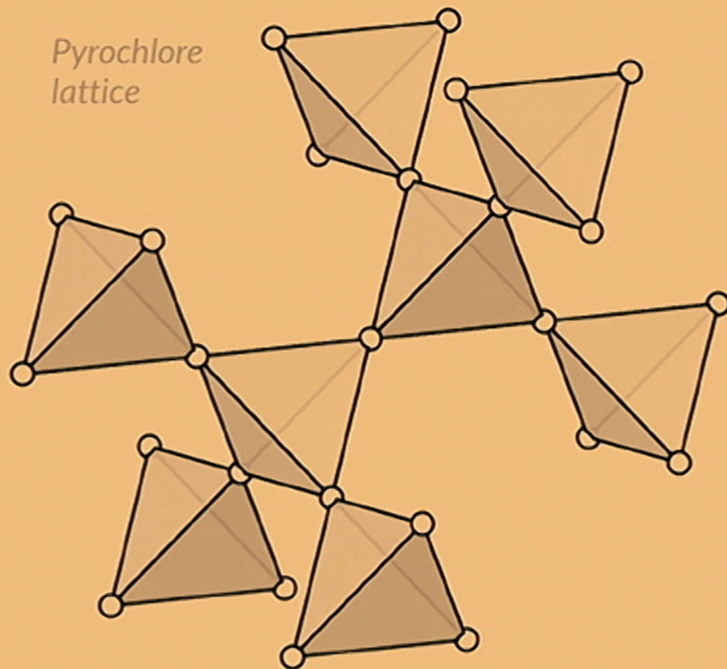
Jeffrey G. Rau
University of Waterloo

Four Corners Meeting (May 25th, 2017)

Pyrochlore magnets

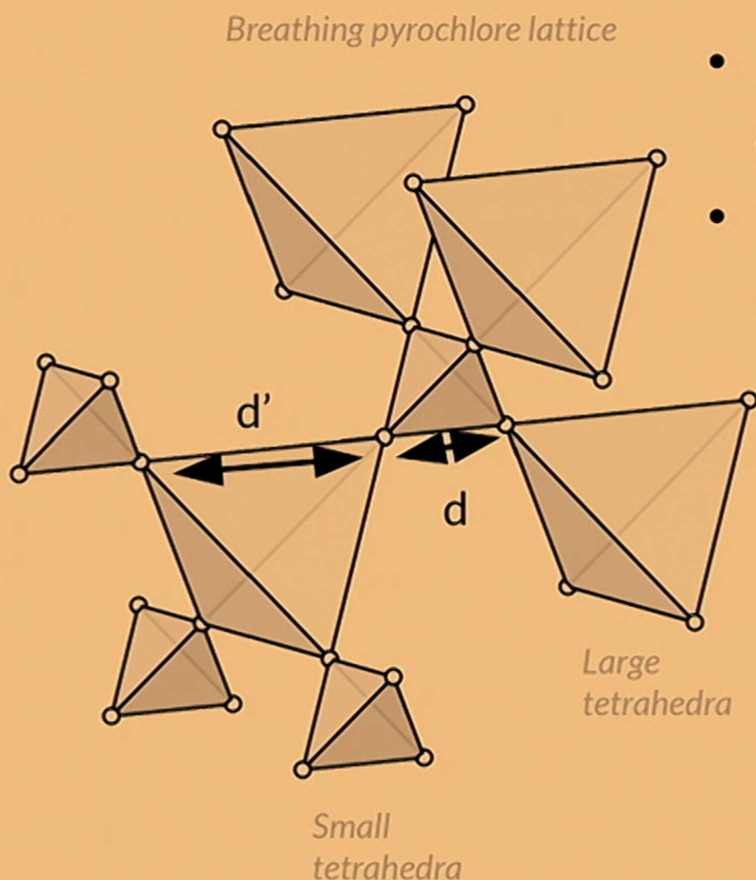
- 3D Frustrated lattice
- E.g. $R_2M_2O_7$, AR_2X_4, ...
- Lots of interesting physics
  - Classical/quantum spin ice
  - Order by disorder
  - Partial order
  - Multi-phase competition
  - ...

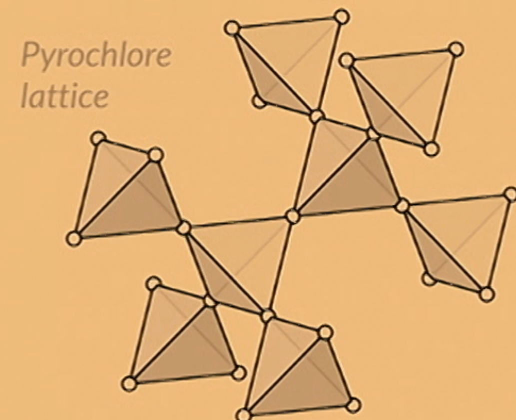
Breathing pyrochlore?



Breathing pyrochlore lattice

- Expand *half* of tetrahedra
- Removes *inversion symmetry* of lattice
- New control parameter → **Breathing ratio** = d'/d



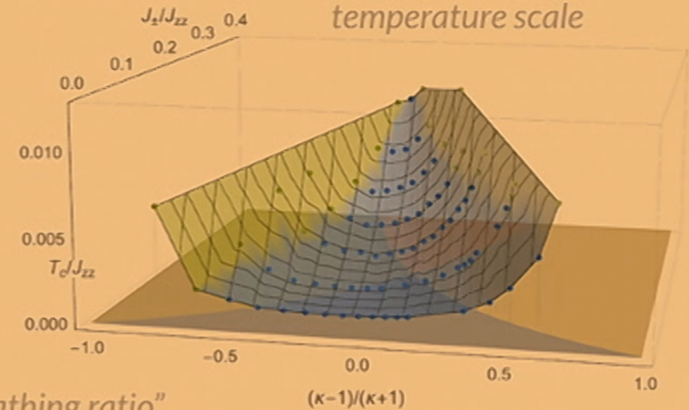


What's so interesting?

Why could it be interesting?

- **Breathing quantum spin ice:**
  - Stronger quantum effects?
- **Inversion breaking**
  - Topological excitations (Weyl magnons)
- **Controlled limit:**
  - Nearly decoupled tetrahedra

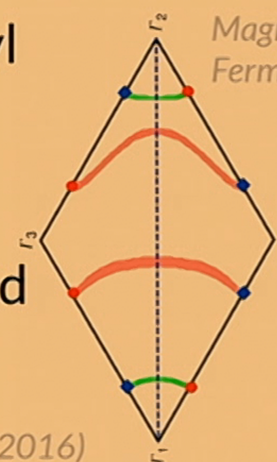
Characteristic temperature scale



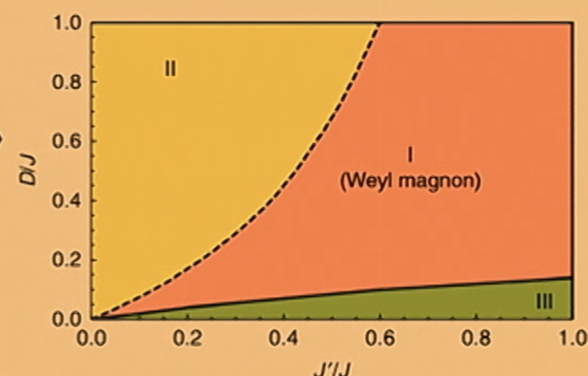
"Breathing ratio"

$$J_{\rm ring} = 6(\kappa^3 + \kappa^{-2})\frac{J_\pm^3}{J_{zz}^2}$$

Magnon Fermi-arcs

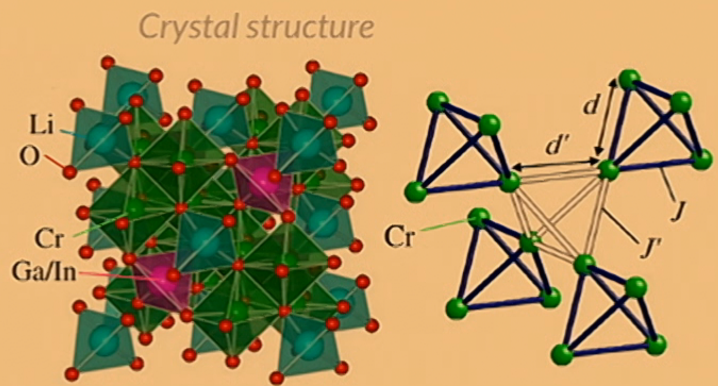


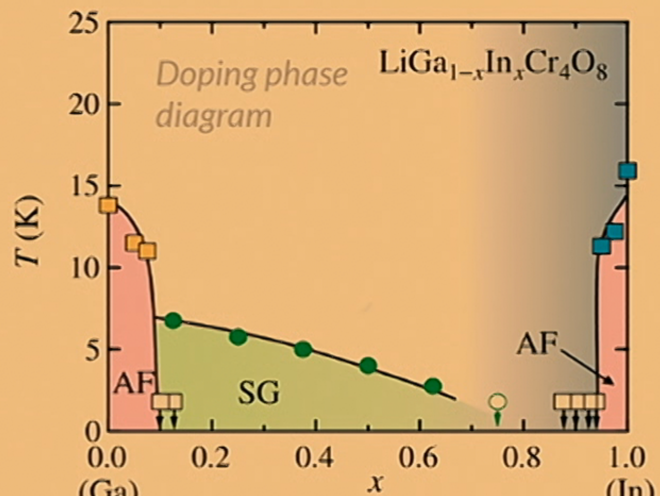
Phase diagram



Savary et al, Phys. Rev. B ***94****, 075146 (2016)*
Li et al, Nat. Comm. *7, 12691 (2016)*

Materials





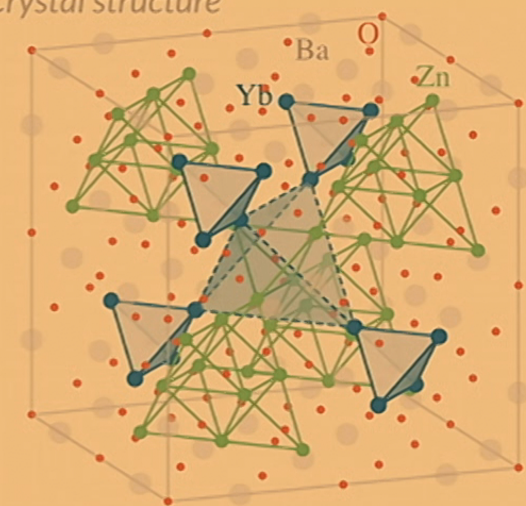
- **Example:** Li(Ga,In)Cr_4O_8
  - Breathing ratio not large ~ 3%-5%
  - Ordering
  - In-Ga mixing?
- **Example:** GaV_4S_8
  - Larger breathing ratio
  - Not exactly localized

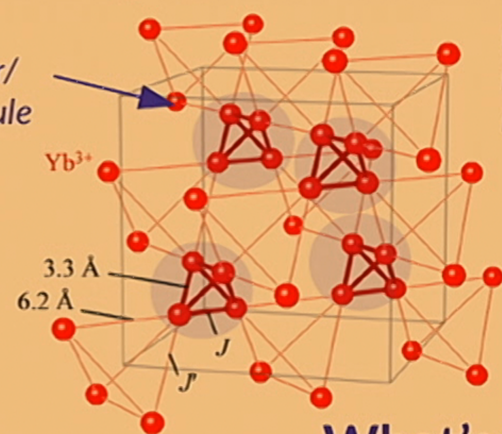
$Ba_3Yb_2Zn_5O_{11}$?

Okamoto et al, Phys. Rev. Lett. **110***, 097203 (2013)*
Okamoto et al, J. Phys. Soc. Jpn. **84***, 043707 (2015)*

Materials (cont.)

- Much more "breathing"
  - **Factor of two** in inter- and intra-tetrahedraon distance
  - Spin "molecules"
- **Rare-earth based**
  - Highly localized magnetic degrees of freedom





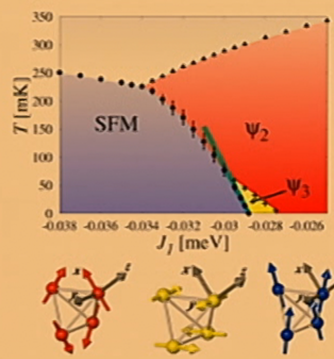
Kimura et al, Phys. Rev. B 90, 060414(R) (2014)

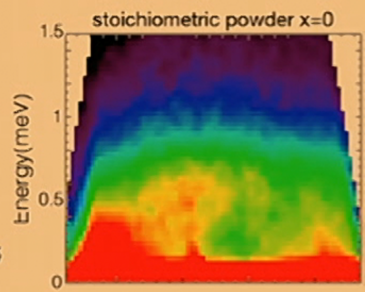
What's the goal?

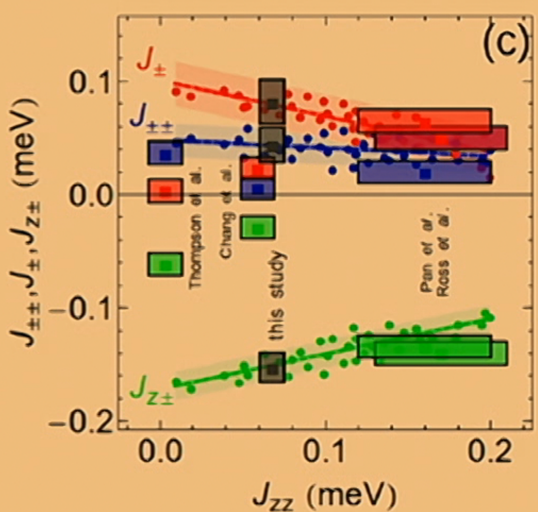
Things to understand

- **Controlled starting point:**
  - Both model *and* solution of model can be worked out (essentially) exactly
- **Better understanding of rare-earth exchange:**
  - Can we understand different **anisotropic exchange** regimes in other rare-earth magnets?
- **Possible collective inter-tetrahedron physics:**
  - New **cluster** degrees of freedom?
  - Interesting **collective** physics?

Example: Rare-earth exchange





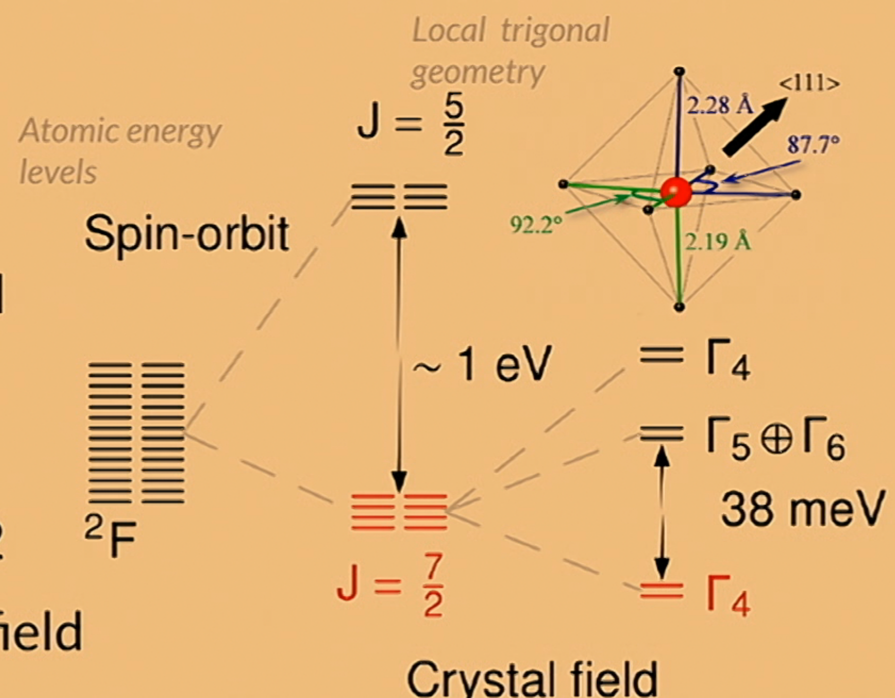


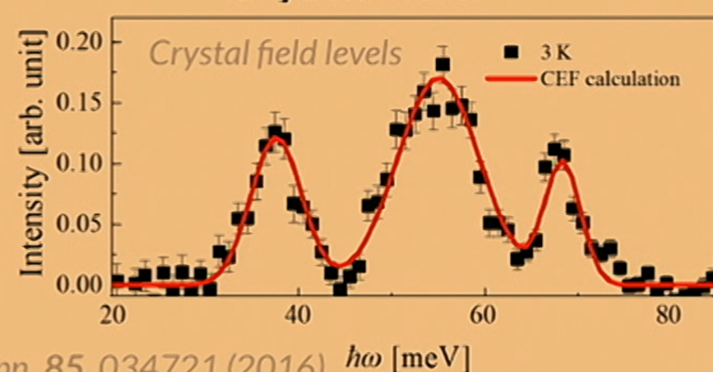
- $Yb_2Ti_2O_7$ serves as a good example
- Very interesting low temperature physics
- **Difficult to pin down model**
- Several, *very different*, proposed exchange parameter sets

Ross et al, Phys. Rev. X 1, 021002 (2011)
Robert et al, Phys. Rev. B 92, 064425 (2015)
*Gaudet et al, Phys. Rev. B **93**, 064406 (2016)*
Thompson et al, arxiv:1703.04506

Basics

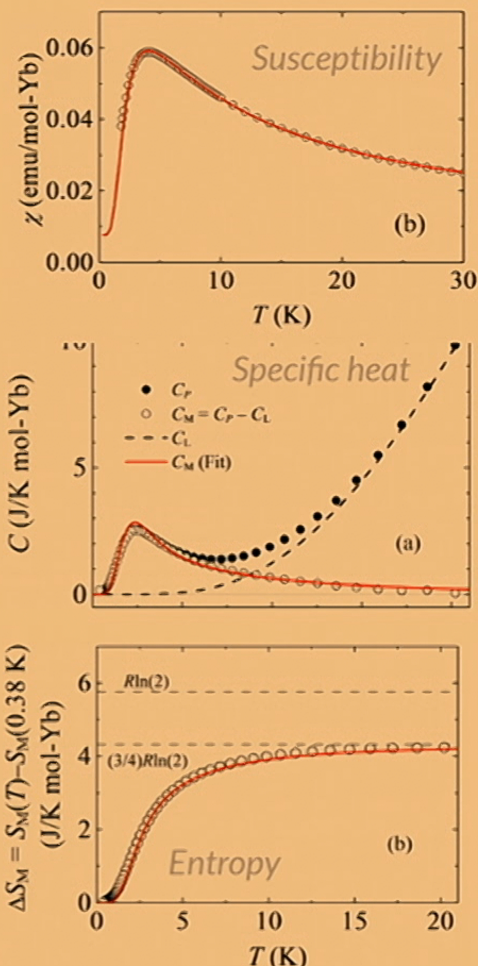
- $Yb^{3+} \rightarrow 4f^{13}$
- **One hole** in 4f-shell
- **Large** spin-orbit coupling
- L=3, S=1/2 → J=7/2
- Split by C_{3v} crystal field
  - Four Kramers' doublets
- **Well-separated** pseudo-spin doublet (~38 meV)





Haku et al, J. Phys. Soc. Jpn. **85**, 034721 (2016)

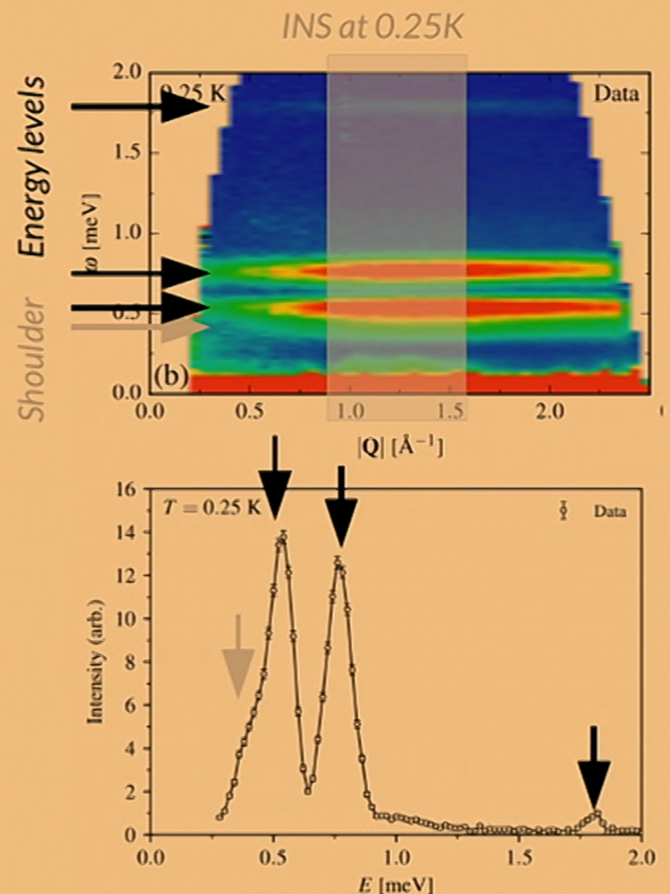
Basics (cont.)



- Broad features in specific heat, susceptibility
- Susceptibility not cleanly going to zero
- **No indications of any transition**
- Residual (magnetic) entropy
  - ~¼ log 2 at ~0.3K
- One doublet degree of freedom per tetrahedron?

Spectrum?

Spectroscopy



Cut integrated over
1A<|Q|<1.5A

- Powder samples
- **Inelastic neutron scattering** (ORNL)
- Flat, dispersionless modes → **decoupled tetrahedra**
- Many distinct levels
  - Two main modes
  - One weak, one shoulder
- **Strong spin-orbit**

Rau et al, Phys. Rev. Lett. **116** *257204 (2016)*

Model

- Write down *symmetry allowed* model
- **Six** parameters
  - Four exchanges
  - Two g-factors

General tetrahedron model with two spin interactions

$$H_{\text{eff}} \equiv \sum_{i=1}^{4} \sum_{j<i} \{J_{zz} S_i^z S_j^z - J_{\pm}(S_i^+ S_j^- + S_i^- S_j^+) + J_{\pm\pm}(\gamma_{ij} S_i^+ S_j^+ + \text{H.c.}) + J_{z\pm}[\zeta_{ij}(S_i^z S_j^+ + S_i^+ S_j^z) + \text{H.c.}]\},$$

Relation of magnetic moment to pseudo-spins

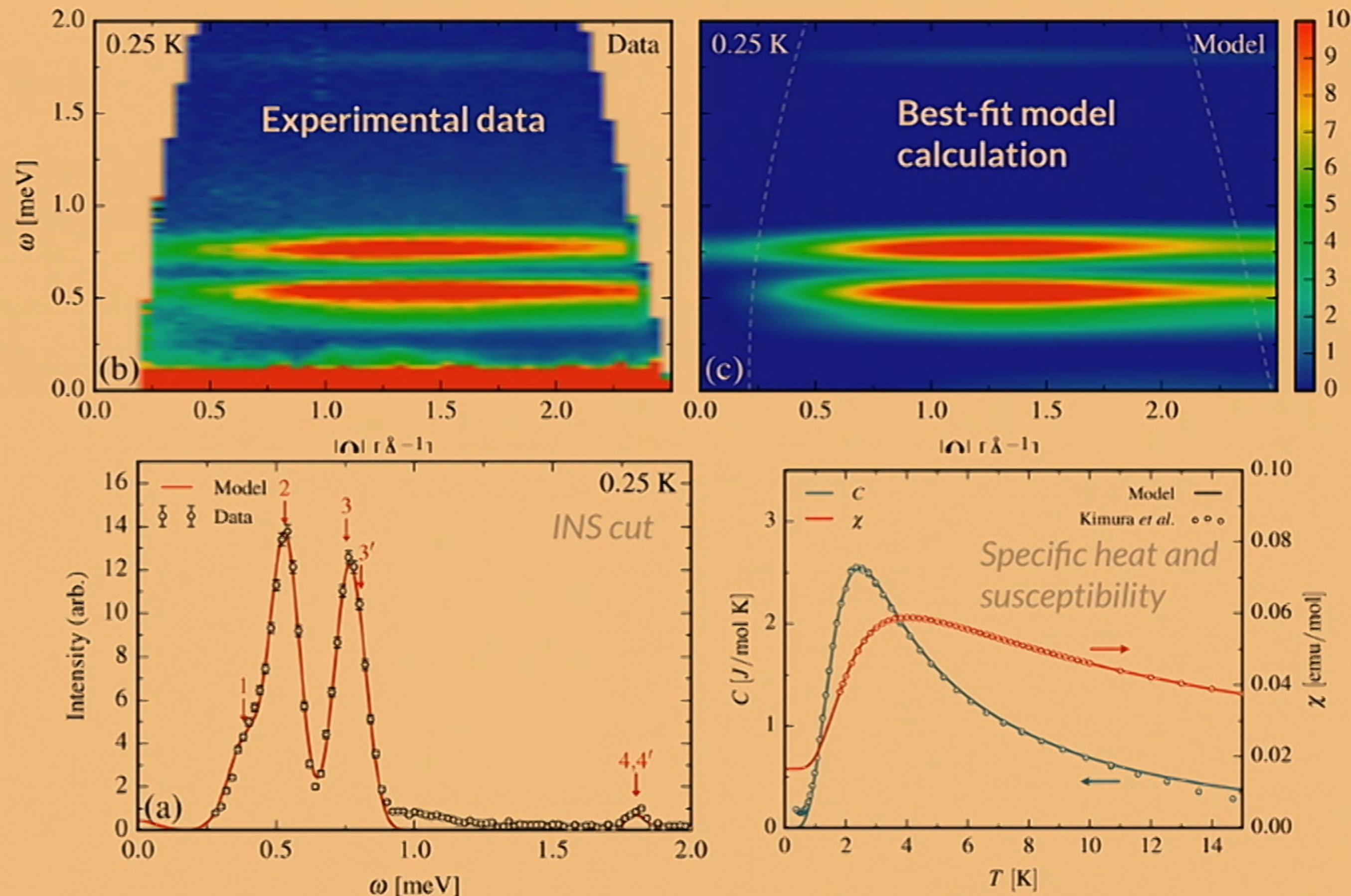
$$\boldsymbol{\mu}_i \equiv \mu_B [g_{\pm}(\hat{\mathbf{x}}_i S_i^x + \hat{\mathbf{y}}_i S_i^y) + g_z \hat{\mathbf{z}}_i S_i^z],$$

Best fit (INS, specific heat & susceptibility)

$$J_{zz} = -0.037 \text{ meV}, \quad J_{\pm} = +0.141 \text{ meV},$$
$$J_{\pm\pm} = +0.158 \text{ meV}, \quad J_{z\pm} = +0.298 \text{ meV},$$
$$g_{\pm} = 2.36, \quad g_z = 3.07.$$

Model (cont.)

- Quantitative reproduction of **all** experimental data



Model (cont.)

- Model more illuminating in **global basis**
- **Four** types of interaction: Heisenberg, Kitaev, symmetric off-diagonal, Dzyaloshinkskii-Moriya

$$H = \sum_{\langle ij \rangle} \bar{\mathbf{S}}_i^{\intercal} \bar{\mathbf{J}}_{ij} \bar{\mathbf{S}}_j$$

Equivalent to usual local exchanges

$$\bar{\mathbf{J}}_{12} = \begin{pmatrix} J+K & +\frac{D}{\sqrt{2}} & +\frac{D}{\sqrt{2}} \\ -\frac{D}{\sqrt{2}} & J & \Gamma \\ -\frac{D}{\sqrt{2}} & \Gamma & J \end{pmatrix}$$

Kitaev exchange

Dzyaloshinskii-Moriya (DM)

Heisenberg exchange

Symmetric off-diagonal

$J = +0.587$ meV

$D = -0.164$ meV

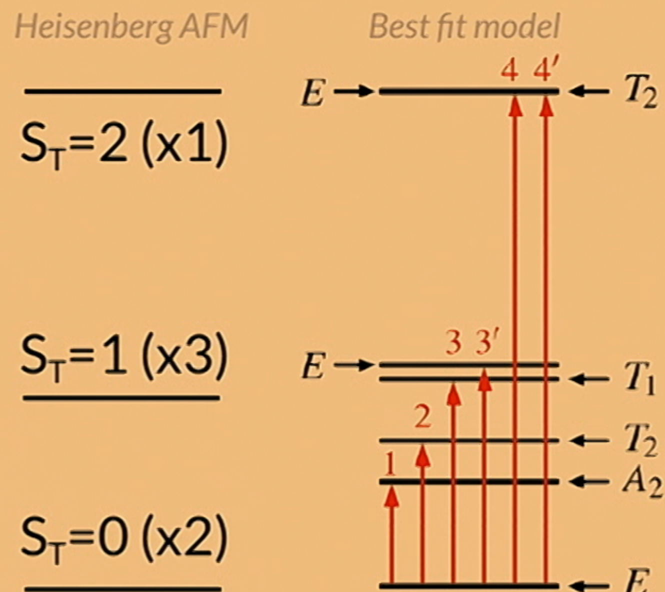
$K = -0.014$ meV

$\Gamma = -0.01$ meV

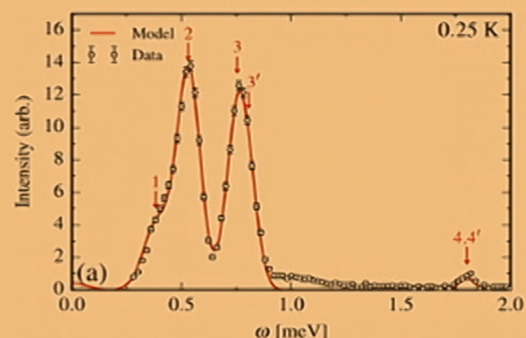
Lots of structure:

- Strong AF Heisenberg
- Indirect DM (D/J ~ -0.28)
- **Negligible** symmetric anisotropies

Model (cont.)



INS cut showing levels



- Heisenberg-AFM good starting point
- Total spin S_T almost good quantum number

$$H_{\mathrm{H-AFM}} \propto \left| \sum_{i=1}^{4} \mathbf{S}_i \right|^2 \equiv |\mathbf{S}_T|^2$$

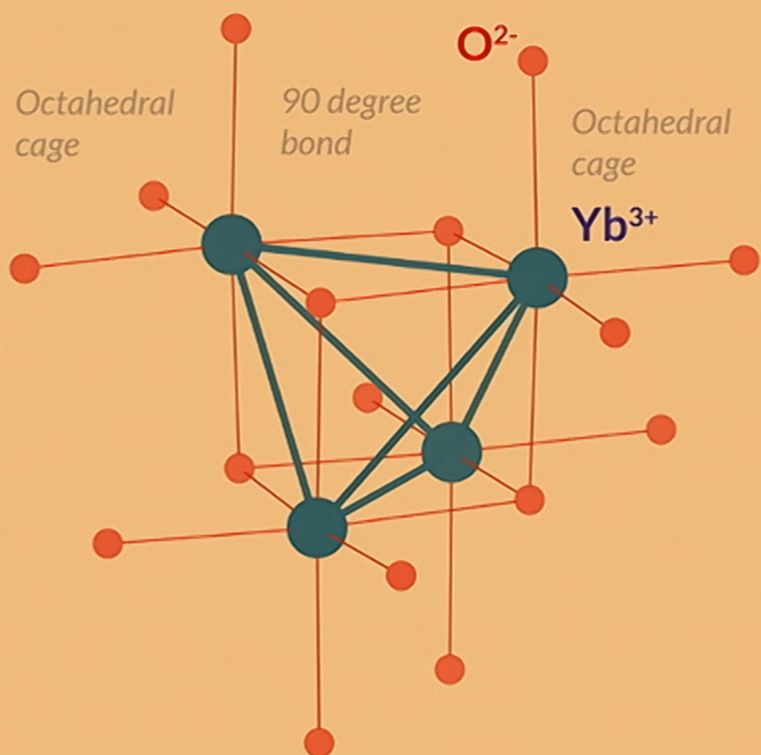
Level structure

- Two S_T=0 singlets
- Three S_T=1 triplets
- One S_T=2 quintet

- DM splits S_T=1 states, but **not** S_T=0 or 2

Origins of exchange?

Exchange physics



Super-exchange processes?

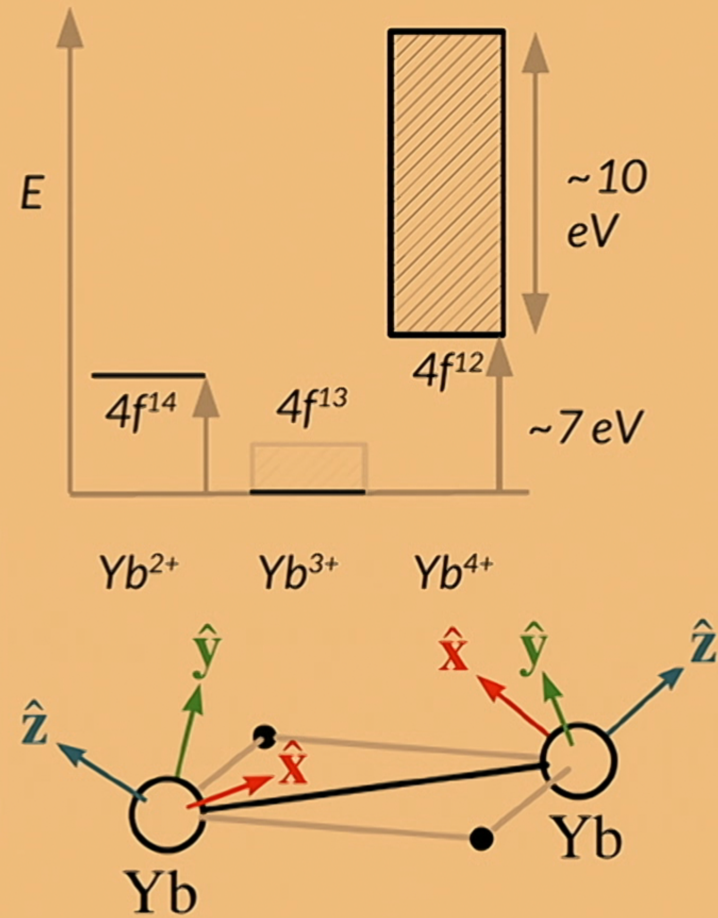
- **Exchange interaction isn't generic!**
- Why?
- No clear exchange "regime" in many other pyrochlore magnets
- Yb-Yb bond
  - **Edge-shared** octahedra
  - **Two** exchange paths

Exchange physics (cont.)

- Can try and calculate
  - **It works!**
- Reproduces exchange almost quantitatively
- **Robust** to details of calculation

Computed from g-factors:
$D/J \sim -0.23, \ K/J \sim \Gamma/J \sim -0.01$

Fitted from experiment:
$D/J \sim -0.28, \ K/J \sim \Gamma/J \sim -0.01$



Rau et al, Phys. Rev. Lett. ***116*** *257204 (2016)*
Park et al, Nat. Comm. ***7*** *12912 (2016)*

Exchange physics (cont.)

- *Why* does it work?
- **Simplest picture:** only lowest orbital, **perfect** octahedral cage

Creates/destroys hole in CEF doublet

$$-t_{\text{eff}} \sum_{\langle ij \rangle \sigma} c^{\dagger}_{i\sigma} c_{j\sigma} + U_{\text{eff}} \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

Symmetry forces NN hopping to be pseudo-spin independent

Only on-site interaction for pseudo-spin 1/2

- **Robust** anti-ferromagnetic Heisenberg exhange

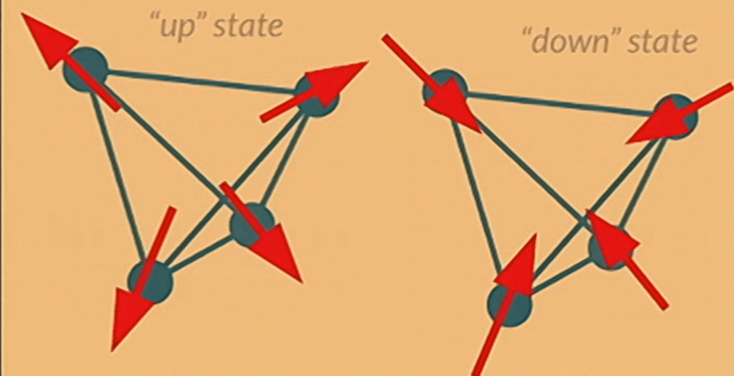
Real material: distorted octahedral cage

- Pseudo-spin dependent hopping → DM interaction

- Emergent *weak* anisotropy

Low energy physics?

Ground state



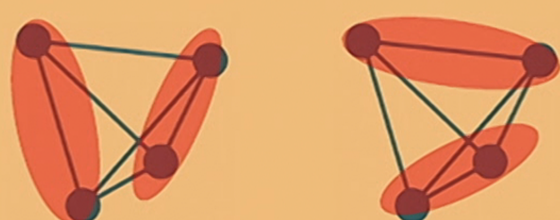
$$\langle \pm | \tilde{\mathbf{S}}_i | \pm \rangle = \pm \lambda \hat{\mathbf{z}}_i$$

DM induced AIAO moment

$$\langle \pm | \tilde{\mathbf{S}}_i | \mp \rangle = 0 \qquad \lambda \sim 0.13$$

Large spin chirality

$$\langle \pm | \tilde{\mathbf{S}}_i \cdot (\tilde{\mathbf{S}}_j \times \tilde{\mathbf{S}}_k) | \pm \rangle \sim \pm 0.4$$



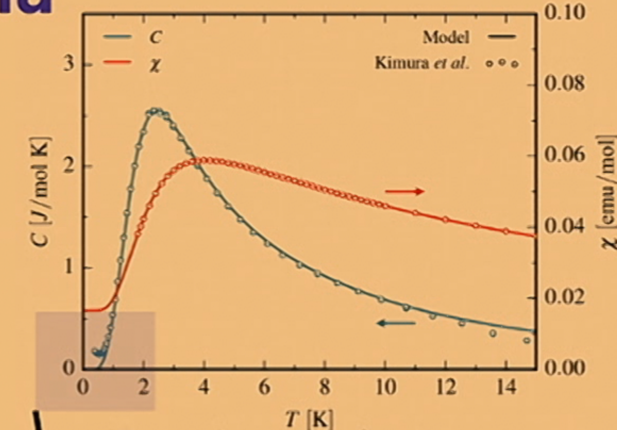
In-plane moments are VBS states – "quadrupolar"

- **Doublet** ground state on tetrahedron
  - Non-Kramers
- Smoothly connected to AF Heisenberg ground state
- Low temperature state is **octupolar paramagnet**
  - AIAO for z
  - VB states for x/y
- "Pauli" behaviour in non-linear susceptibility

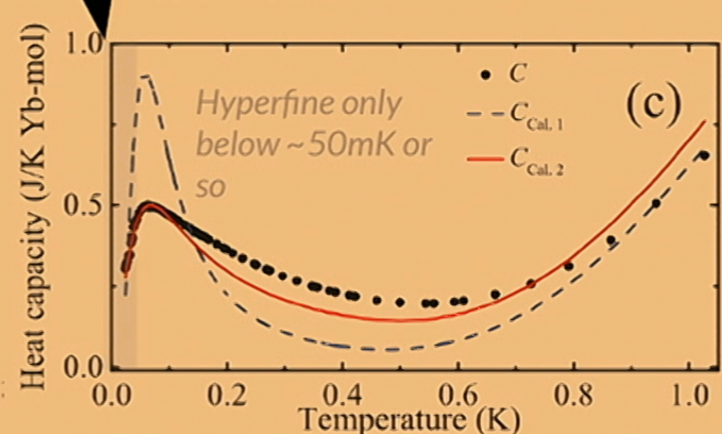
Interactions?

Collective phenomena

- How do we resolve this doublet degeneracy?
- Further neighbour exchanges ~ 100 mK
- Interactions between E-doublets could become important below ~0.5K
- **Interacting non-Kramers doublets on FCC lattice**
- *Some* evidence for this



Upturn in specific heat below ~0.5K



Haku et al, Phys. Rev. B **93**, 220407(R) (2016)

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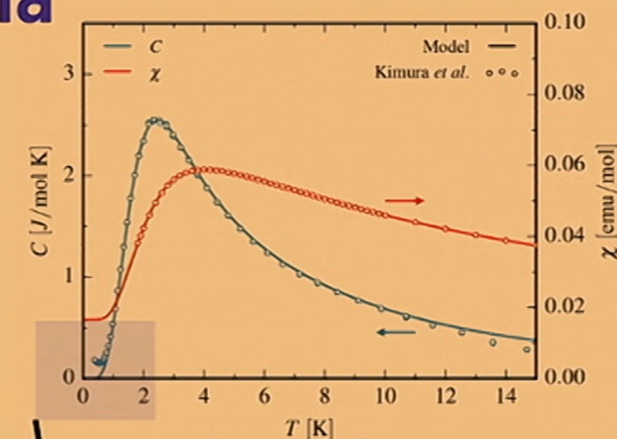

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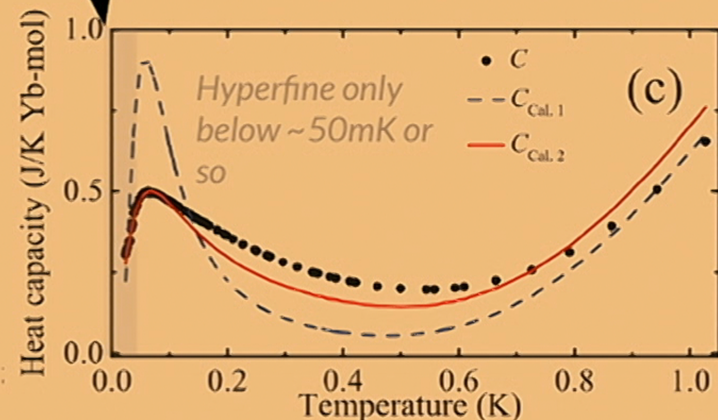

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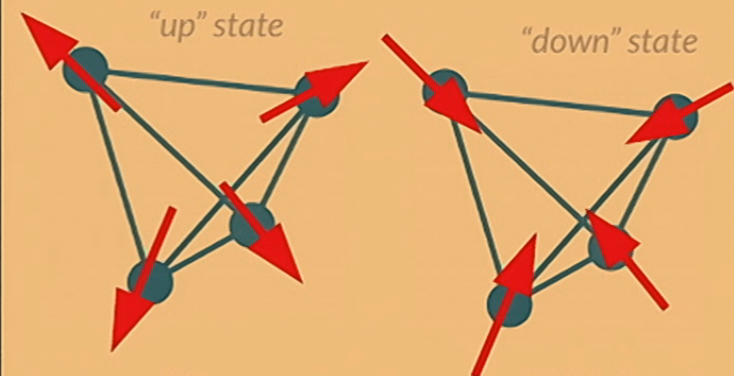
Ground state



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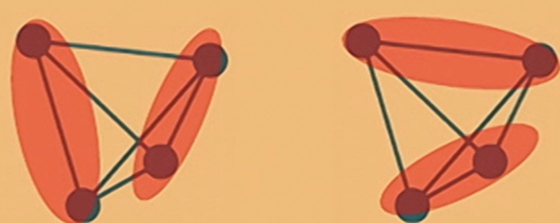

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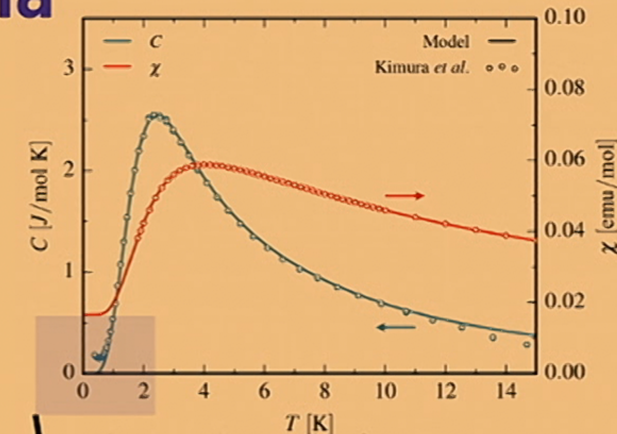

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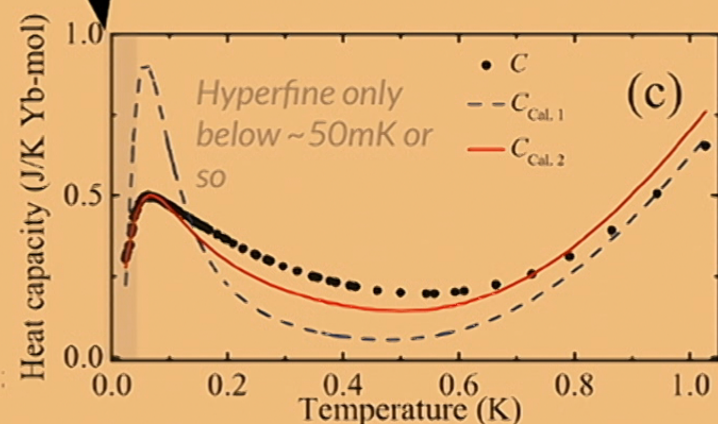
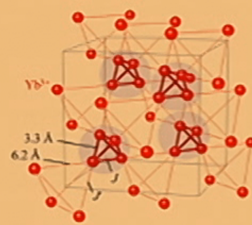


Haku et al, Phys. Rev. B **93**, 220407(R) (2016)

Collective phenomena (cont.)



- General inter-tetrahedron exchange model projects to **Ising-only interactions**

$$P\mathbf{S}_i P = \lambda T_I^z \hat{\mathbf{z}}_i$$

- Virtual intra-tetrahedron exchange **competitive**
  - Generates "transverse" terms

General: Anistropic exchange on FCC lattice
- Three symmetry allowed couplings

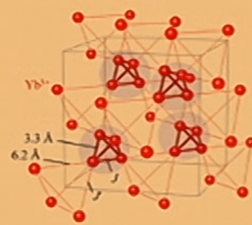
$$\sum_{\langle IJ \rangle} \left[Q_{zz} T_I^z T_J^z + Q_{\pm} \left(T_I^+ T_J^- + \text{h.c.} \right) + Q_{\pm\pm} \left(\xi_{IJ} T_I^+ T_J^+ + \text{h.c.} \right) \right]$$

Disorder

- Another possible route to resolve degeneracy
- **Structural disorder can split doublet**
- Unclear disorder scale, distribution or origins

Key (open) questions:

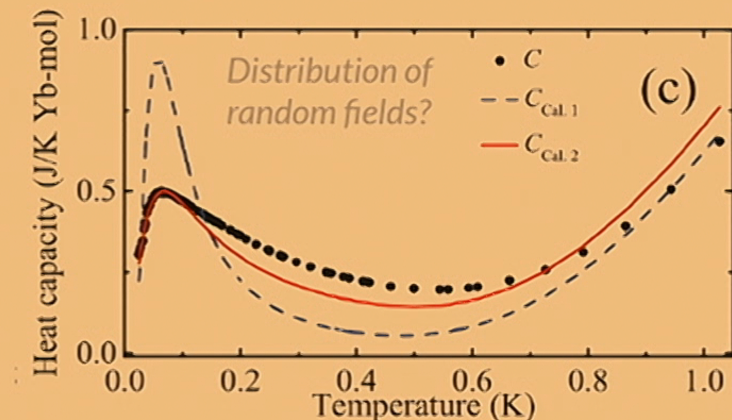
- Disorder?
- Interactions?
- Both?



Random transverse fields

$$\sum_l \left[\Gamma_l T_l^+ + \Gamma_l^* T_l^-\right]$$

Favours random VB states in real space?



*Haku et al, Phys. Rev. B **93**, 220407(R) (2016)*

Conclusions

- **Model system** to understand frustrated rare-earth models
- **Validates** *some* of our understanding of rare-earth exchange more generally

Interesting possible **collective / disorder physics** at very low energies

- Octupolar paramagnet
- **Non-Kramers doublets on FCC lattice**
- Competitive with **disorder** physics?

Rau et al, Phys. Rev. Lett. ***116*** *257204 (2016)*
See also related works:
Park et al, Nat. Comm. ***7*** *12912 (2016)*
Haku et al, Phys. Rev. B ***93****, 220407(R) (2016)*

Thank you for your attention