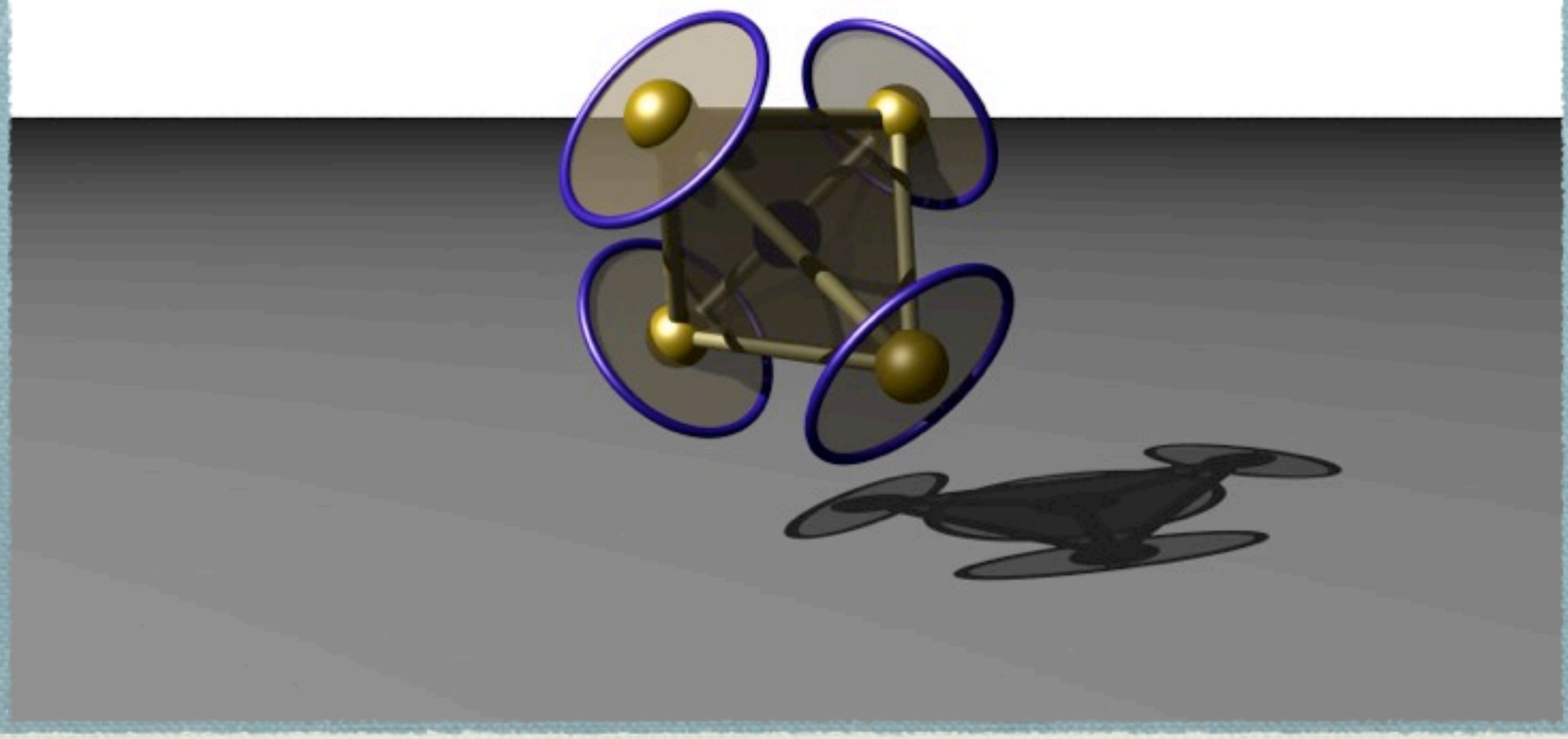


Definitive Evidence for Order-by-Quantum-Disorder

Lucile Savary

Hamilton, June 4th, 2012

Collaborators

Leon Balents
(KITP)

Kate Ross

Bruce Gaulin

Jacob Ruff
(Argonne)

experiments, McMaster

What is order-by-disorder?

- old idea: Villain 1980, Shender 1982, Henley 1989
- so far not definitively exhibited in experiment
- would be nice to do so!

What is order-by-disorder?

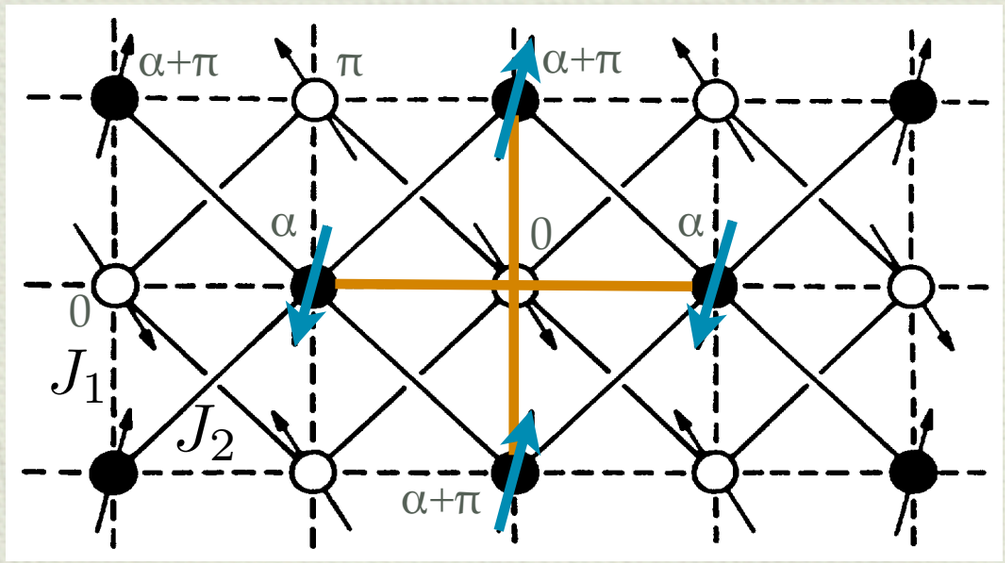
- Look at spin system

$$H = \sum_{i,j} \sum_{\mu,\nu} J_{ij}^{\mu\nu} S_i^\mu S_j^\nu$$

Order-by-disorder: an example

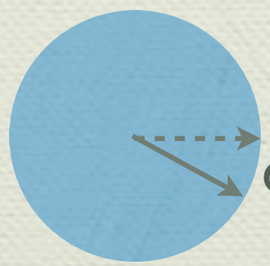
- Henley 1989: J_1-J_2 square lattice

$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$



$\mathbf{H_{eff} = 0}$

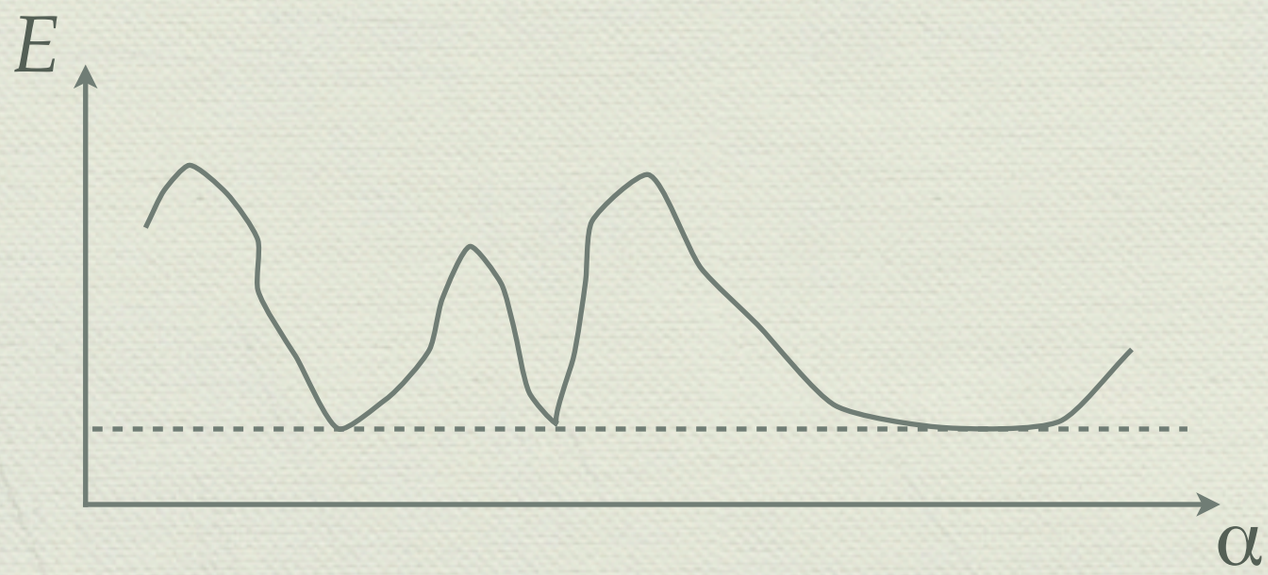
- classical XY unit vectors
- $J_2 > J_1/2 > 0$
- two interpenetrating decoupled Néel states



"U(1)" degeneracy

Order-by-disorder

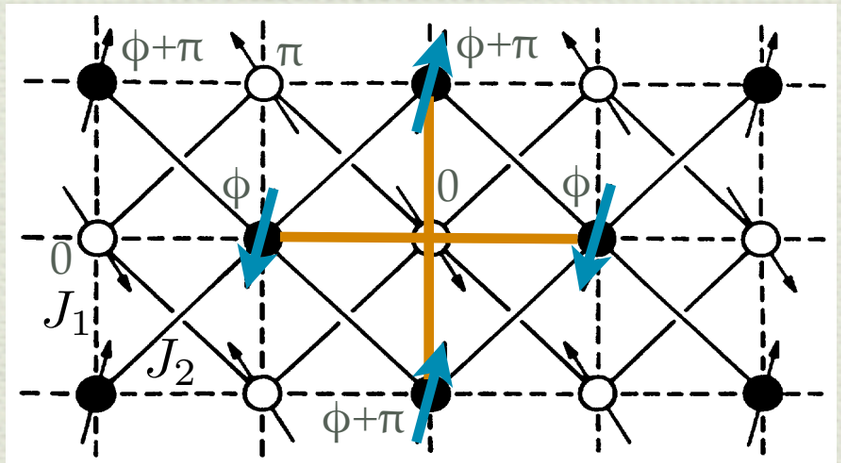
- those states have the same energy
- but different "environment"



Order-by-disorder: an example

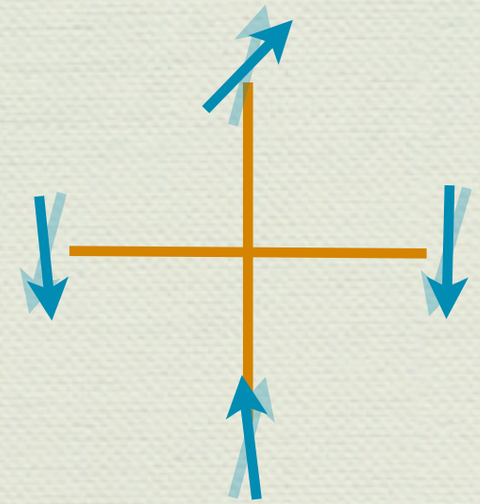
- picture

$T = 0$



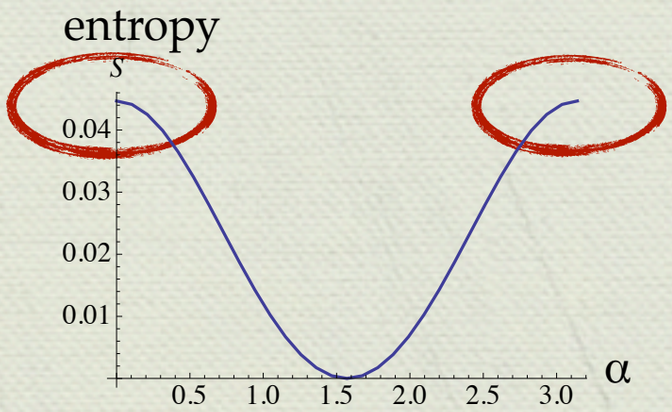
$\mathbf{H_{eff} = 0}$

$T > 0$

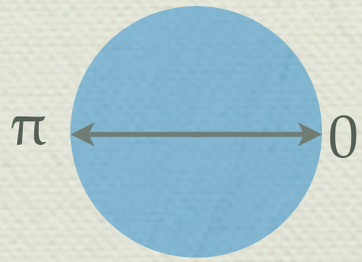


$\mathbf{H_{eff} \neq 0}$

- thermal fluctuations: entropy



$F = E - TS(\phi) = F(\phi)$ minimized for $\phi = 0$ or π



The degeneracy is lifted in the free energy by the entropy

Henley 1989

Order-by-disorder: an example

- quantum fluctuations: zero-point energy

$$E = E_0 + E_{\text{zero-point}}$$

$$\hbar\omega_{\mathbf{k}} = \sqrt{\frac{A_{\mathbf{k}}}{m}}$$

$$E_{\text{zero-point}} = \sum_{\mathbf{k}} \frac{\hbar\omega_{\mathbf{k}}}{2} \sim \frac{1}{\sqrt{2m}} \sum_{\mathbf{k}} \sqrt{A_{\mathbf{k}}}$$

classical degeneracy lifted by the zero-point energy

What is order-by-disorder?

- when a system displays *classical* accidental degeneracy which is (at least partially) lifted by fluctuations

But:

- There usually are other degeneracy breaking terms in the Hamiltonian:
  - further neighbor interactions (dipolar, exchange)
  - spin-orbit coupling & crystal fields
  - spin-phonon coupling
  - multiple-spin terms
  - etc.

So, how can we know?

How can we be sure that ObD is at play?

- need a *robust* classical degeneracy of the ground state
  - protected by symmetry?
  - but should still be allowed to lift it!

} hard

- **so far none was exhibited for sure**
  - look in frustrated magnets!

We show definitive evidence for quantum ObD in $Er_2Ti_2O_7$

Outline

- $Er_2Ti_2O_7$
- Review of "older" literature
- Hamiltonian of $Er_2Ti_2O_7$
- Degeneracy
- Comparison with experiments
- Verifiable consequences

based on arXiv 1204.1320 (to appear in PRL)

The Story

Leon Balents balents@kitp.ucsb.edu via gmail.com 3 avr.

à moi, Bruce, Kate

Bad news - we are too slow...can we rush it out in a day or two?

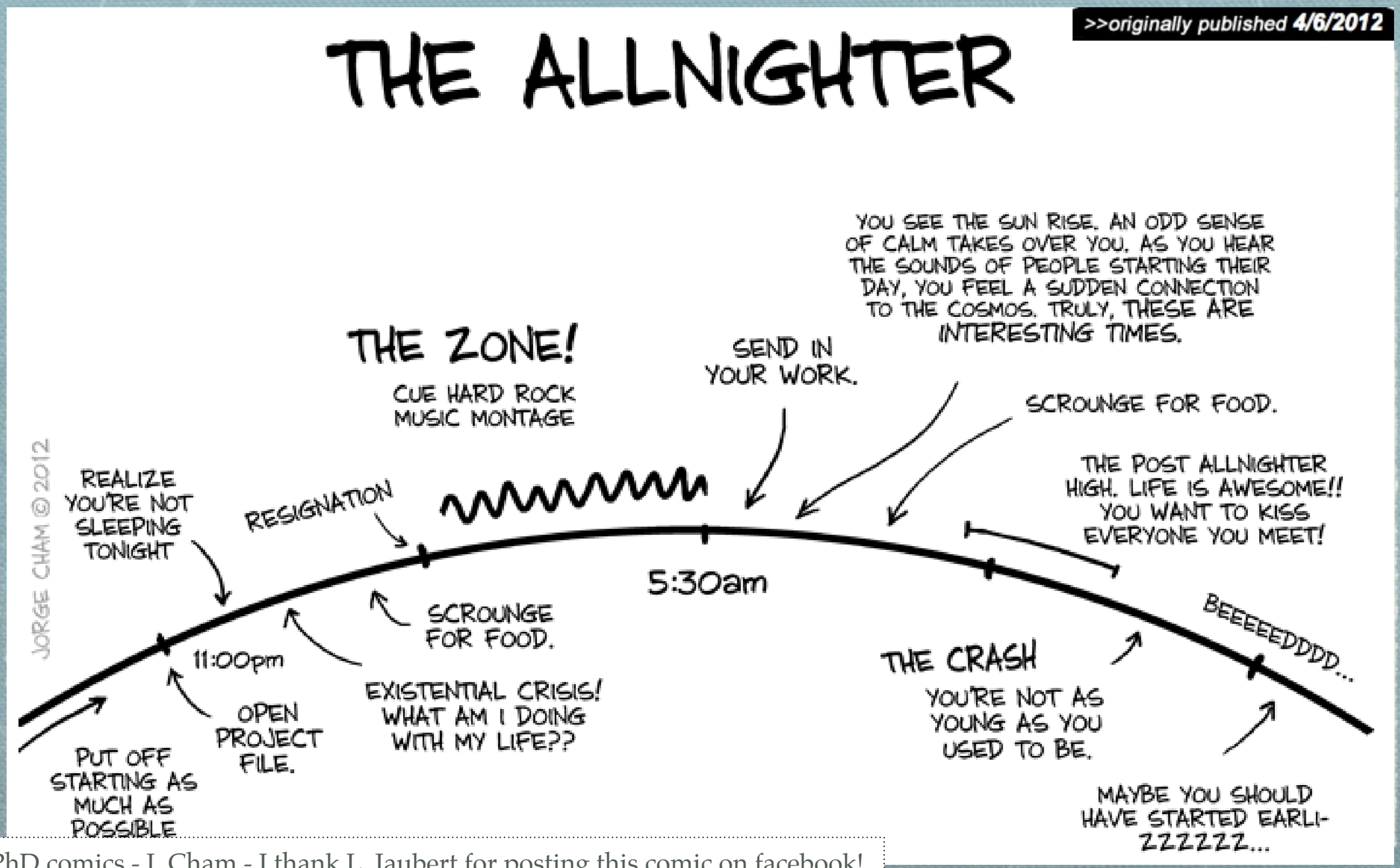
Leon

Quantum order by disorder and accidental soft mode Er2Ti2O7

M. E. Zhitomirsky, M. V. Gvozdikova, P. C. W. Holdsworth, R. Moessner

(Submitted on 3 Apr 2012)

The Story



PhD comics - J. Cham - I thank L. Jaubert for posting this comic on facebook!

The Story

>>originally published **4/6/2012**

THE ALLNIGHTER

Leon .. Bruce, Kate, moi (86) Boîte de réception Yb2Ti2O7 project Quantum order by disorder and accidental soft mode Er2Ti2O7. (arXi

THE ZONE!

CUE HARD ROCK MUSIC MONTAGE

EMAIL PARTY WITH KATE

MEET WITH LEON

SUBMISSION... OH MY!!

SCROUNGE FOR FOOD.

"I DON'T WANT TO DO THIS"

EXISTENTIAL CRISIS! WHAT AM I DOING WITH MY LIFE??

RESIGNATION

THE POST ALLNIGHTER HIGH. LIFE IS AWESOME!! YOU WANT TO KISS EVERYONE YOU MEET!

5:30am

12:30pm

1pm ARVIX DEADLINE

LEON'S PEP EMAIL

11:00pm

REALIZE YOU'RE NOT SLEEPING TONIGHT

SEE LEON'S EMAIL: "WE ARE TOO SLOW"

TAKE OUR TIME TO FINISH

THE CRASH

YOU'RE NOT AS YOUNG AS YOU USED TO BE.

BEEEEEDDDD...

MAYBE YOU SHOULD HAVE STARTED EARLI-

ZZZZZZ...

PhD comics - J. Cham - I thank L. Jaubert for posting this comic on facebook!

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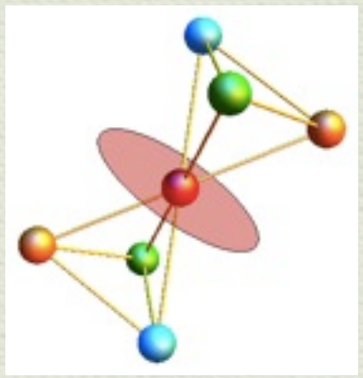
(Submitted on 5 Apr 2012)

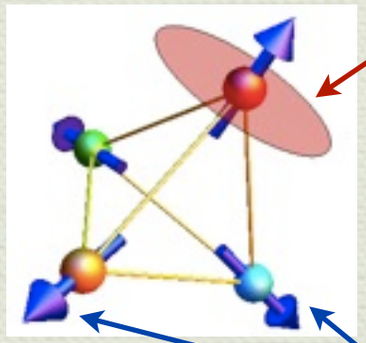
arXiv 1204.1320

725 papers

$Er_2Ti_2O_7$

- Er^{3+} pyrochlore





behaviors:

spin ices

quantum AFM

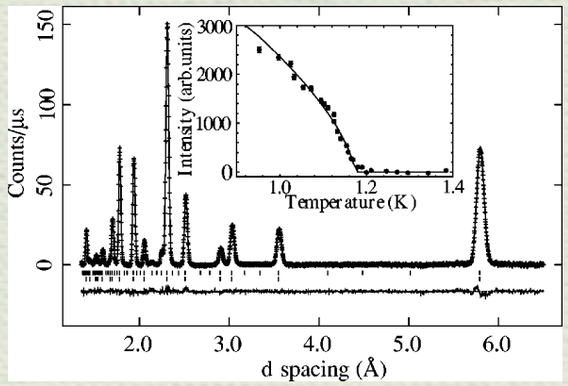
quantum spin liquids ?

- rare-earth pyrochlore family: $Ho_2Ti_2O_7$, $Dy_2Ti_2O_7$, $Ho_2Sn_2O_7$, $Dy_2Sn_2O_7$, $Er_2Ti_2O_7$, $Yb_2Ti_2O_7$, $Tb_2Ti_2O_7$, $Er_2Sn_2O_7$, $Tb_2Sn_2O_7$, $Pr_2Sn_2O_7$, $Nd_2Sn_2O_7$, $Gd_2Sn_2O_7$, ...

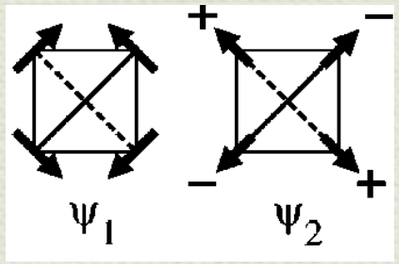
- many presentations this week: Ross, Onoda, Singh, Broholm, Lee, Petit, Bonville, Gukasov, Wan, Tomiyasu, Benton, Kadowaki, Liu, Lhotel, Wiebe, Henley, Fennell, Ryzhkin, Holdsworth, Dunsiger, Bovo, Matthews, Pan, Ishizuka, Powell, McClarty, Toews, Kycia, Jaubert, Tchernyshyov, Aczel, Matsuhira, Storchak, Tachibana, White, Ishikawa, Shinaoka, Yamauchi, Stewart, Shinaoka, Hallas, Silverstein, Clancy, Khemani, MacDougall, Clark, Yamaura

$Er_2Ti_2O_7$: Previous studies

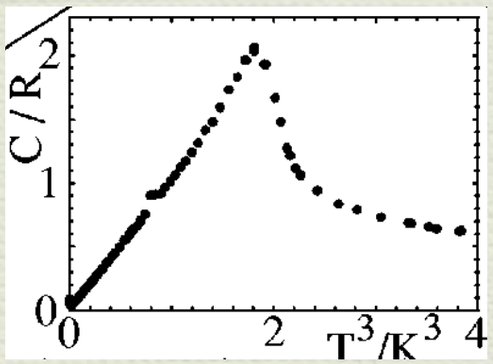
- **k = 0** order



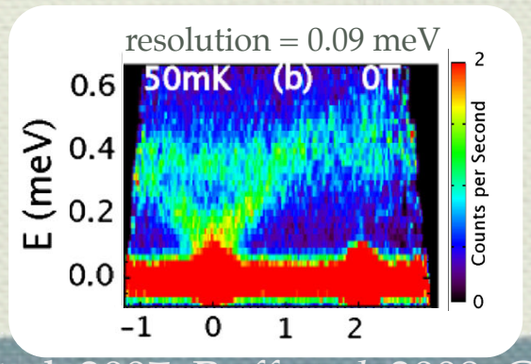
- compatible with "ψ_1"or "ψ_2" states



- specific heat: T^3 behavior - Goldstone?



- Goldstone mode



Champion *et al.* 2003, Poole et al. 2007, Ruff *et al.* 2008, Cao *et al.* 2009

$Er_2Ti_2O_7$: Previous studies

- model:

$$H = \overbrace{J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j}^{\text{isotropic interactions}} + D \left(\sum_i \mathbf{S}_i \cdot \hat{\mathbf{e}}_i \right)^2$$

somewhat ad hoc

- has a huge degeneracy
- order-by-disorder suggested — insightful! — Champion *et al.*
- dipolar interactions invoked — "Palmer-Chalker state" not experimental ground state
- effect of multi-spin terms — very small — McClarty *et al.*, Stasiak *et al.*
- anisotropy necessary — Cao *et al.*, McClarty *et al.*, Stasiak *et al.*
- not 1st order transition?

puzzle

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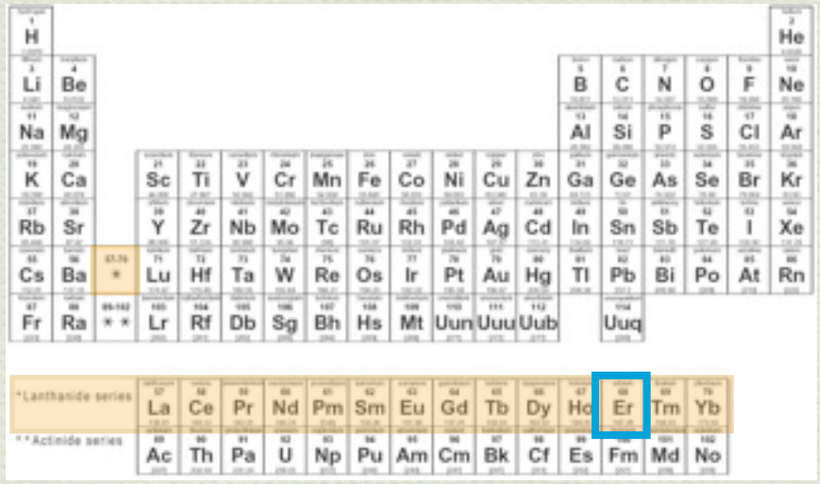
- not 1st order transition?

Need a more accurate Hamiltonian

$Er_2Ti_2O_7$: towards a Hamiltonian

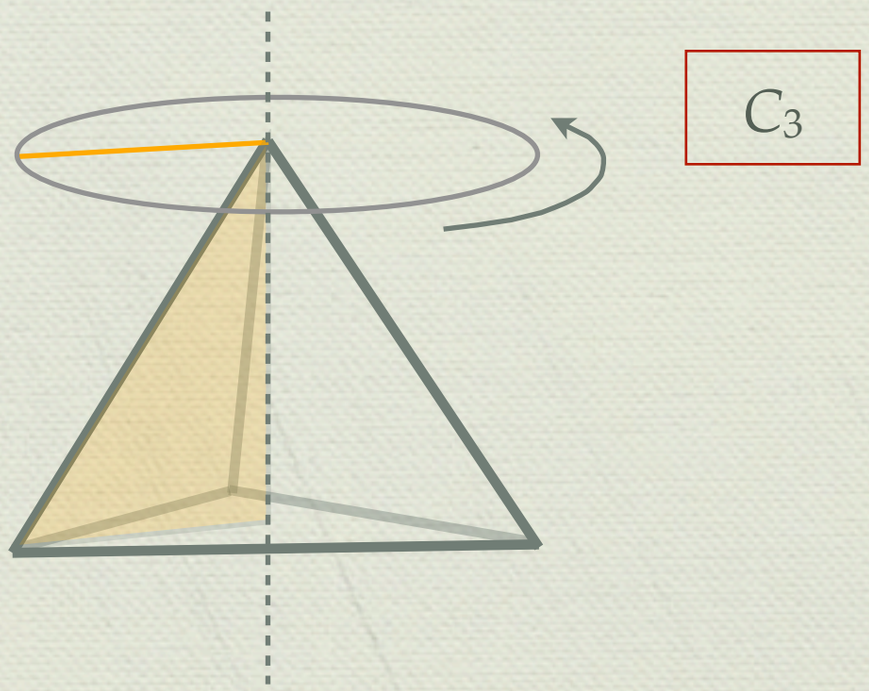


rare-earths : intrinsic strong spin-orbit coupling

$A_2B_2O_7$: strong crystal fields

⟶ *discrete* cubic symmetries *only*

space group: Fd-3m, i.e. #227



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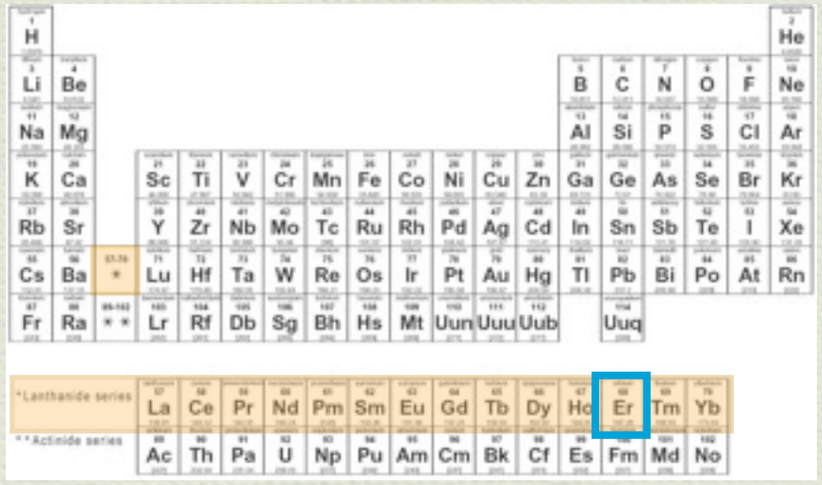

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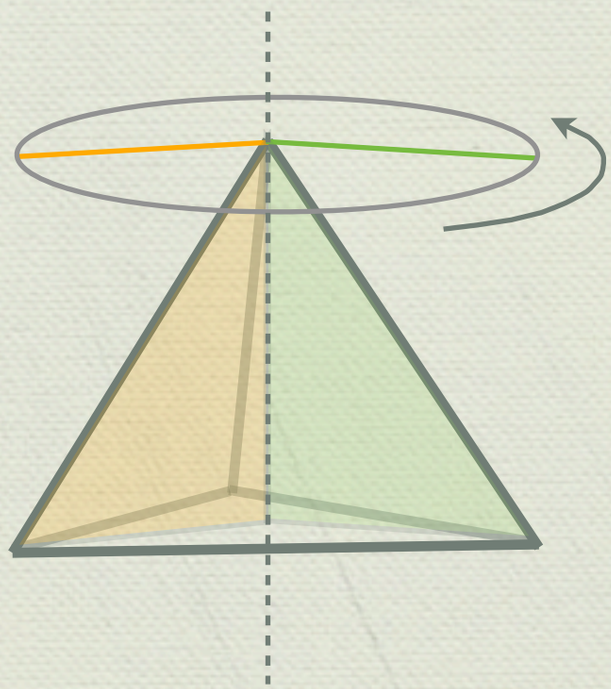
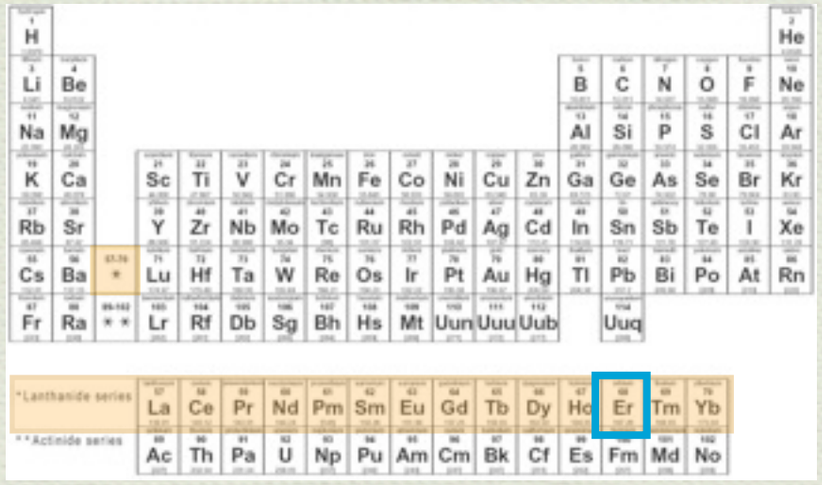


C_3

$Er_2Ti_2O_7$: towards a Hamiltonian



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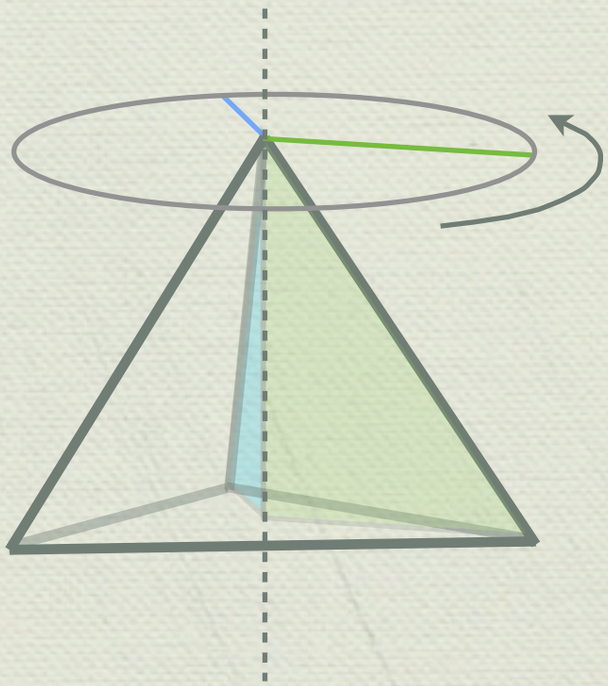

C_3

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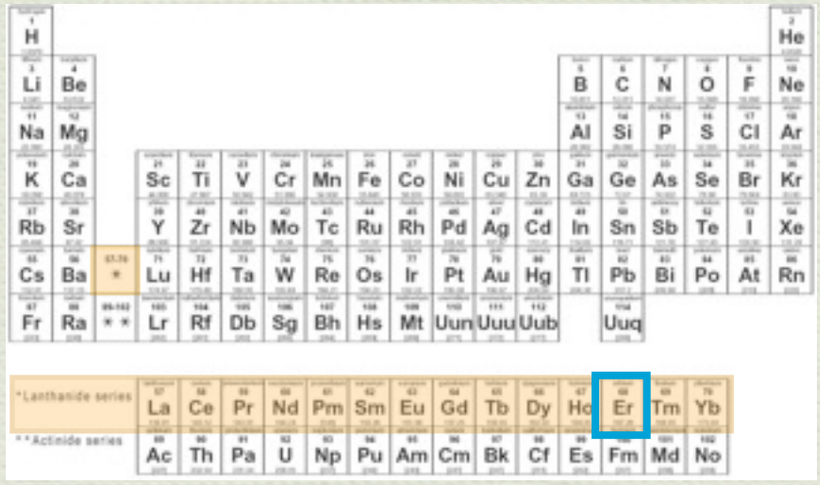

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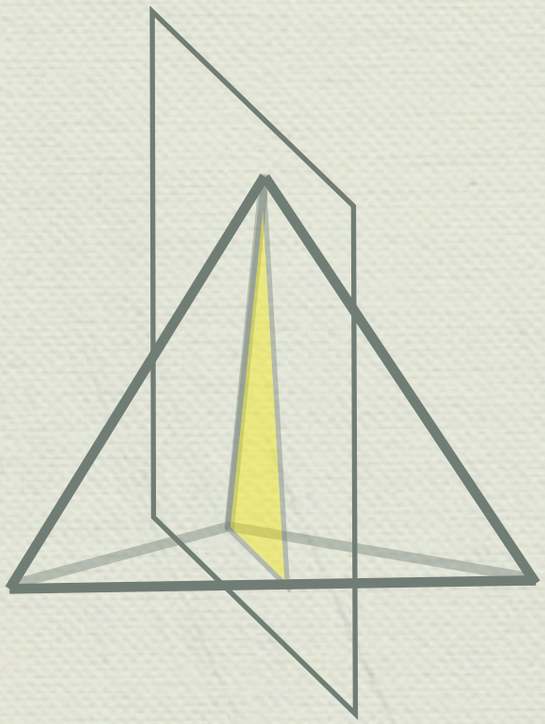
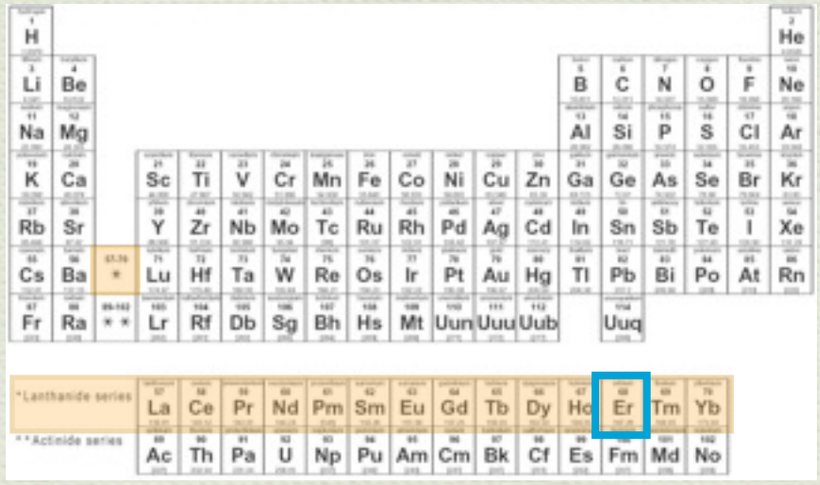


C_3

mirror

$Er_2Ti_2O_7$: towards a Hamiltonian



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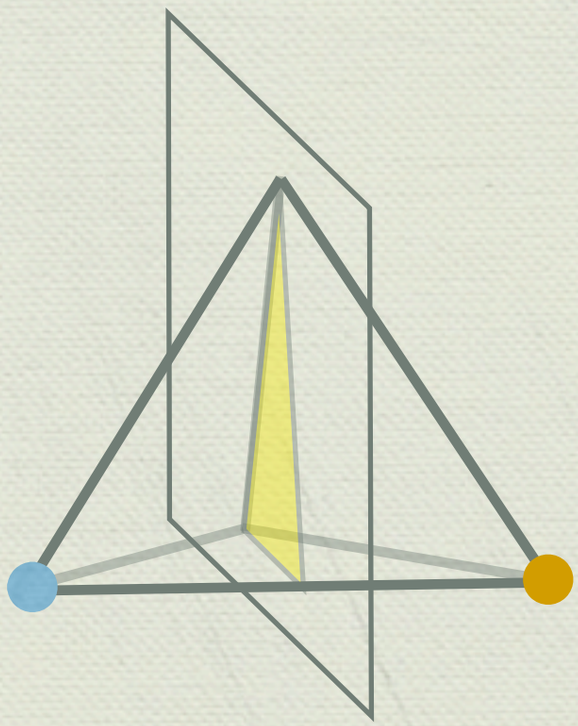

C_3

mirror

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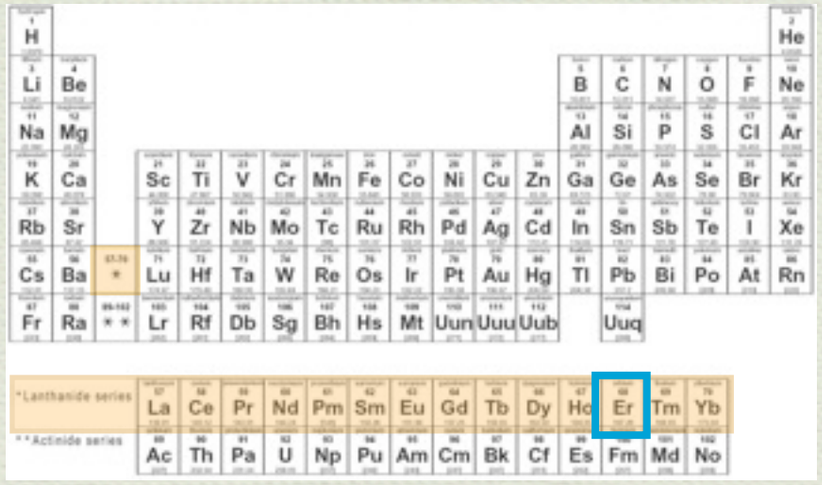

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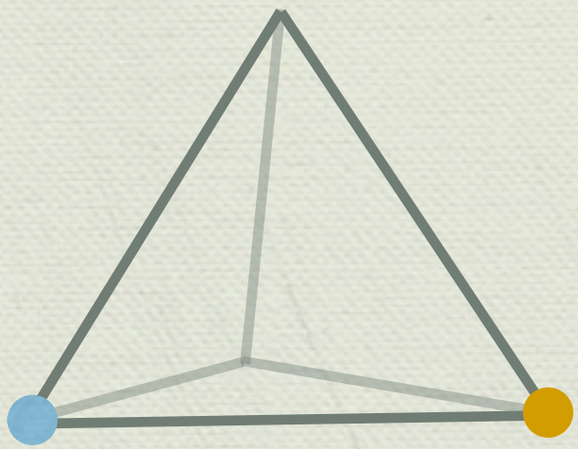
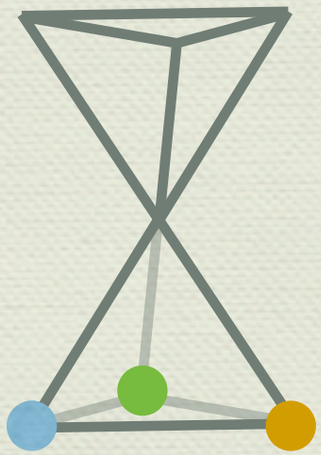


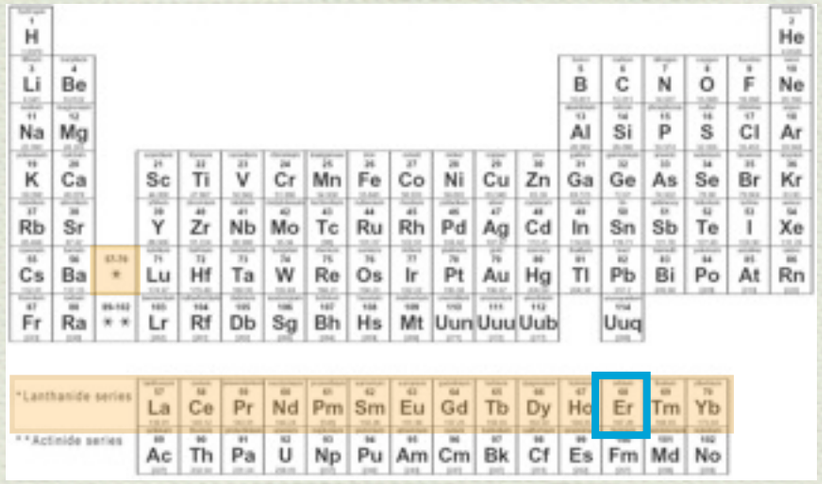
C_3

mirror

inversion



$Er_2Ti_2O_7$: towards a Hamiltonian



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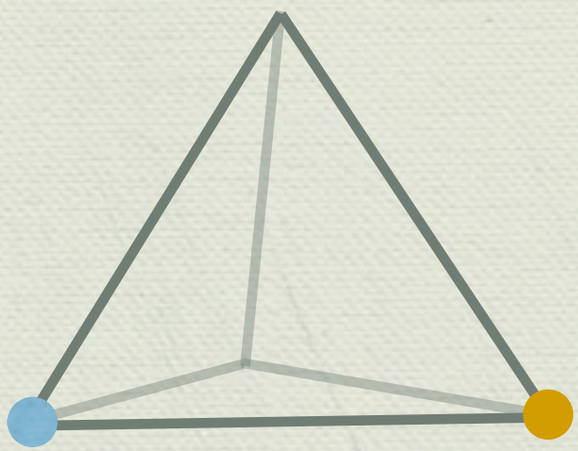
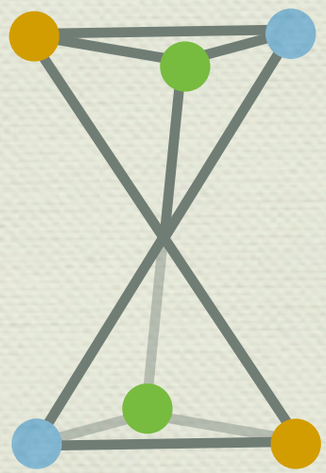


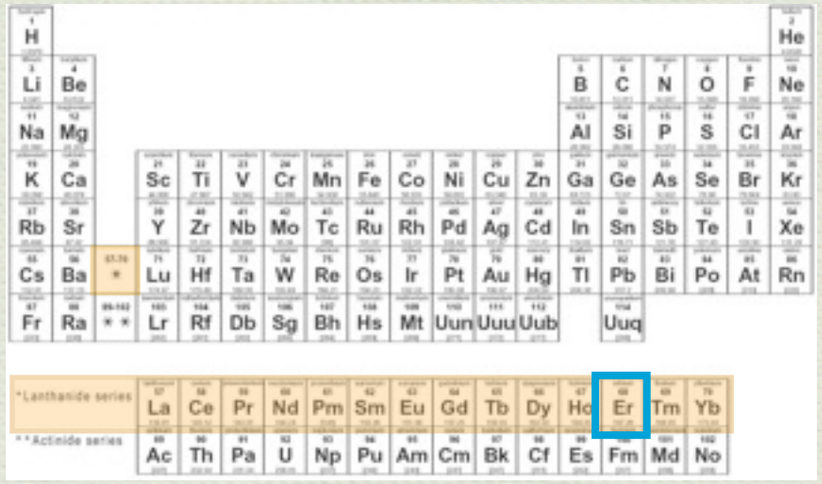
C_3

mirror

inversion



$Er_2Ti_2O_7$: towards a Hamiltonian



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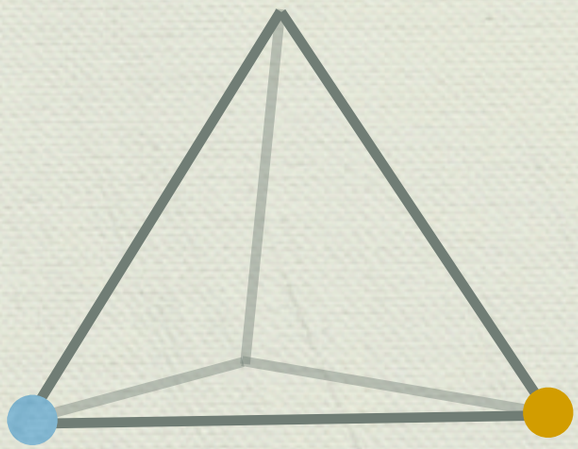
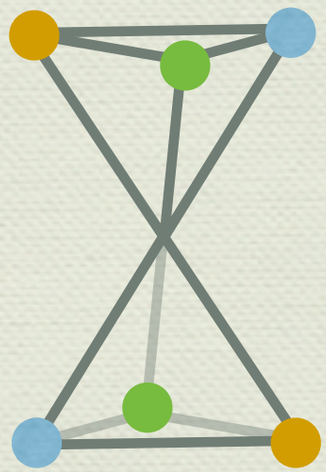


C_3

mirror

inversion

time reversal



$Er_2Ti_2O_7$: most general NN effective spin-1/2 Hamiltonian

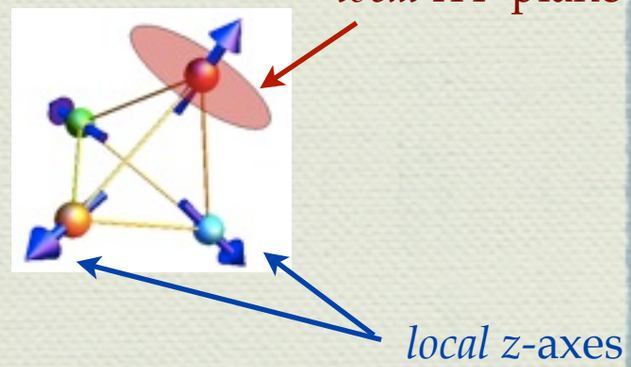
- strong SOC & crystal fields => J = L+S = 15/2, $\Delta \sim 75$ K
- spin-1/2 Hamiltonian:

$$H = \sum_{\langle ij \rangle} \Big[\quad J_{zz} \mathsf{S}_i^z \mathsf{S}_j^z$$

$$- J_{\pm} (\mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+)$$

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

$$+ J_{\pm\pm} \left[\gamma_{ij} \mathsf{S}_i^+ \mathsf{S}_j^+ + \gamma_{ij}^* \mathsf{S}_i^- \mathsf{S}_j^- \right] \Big]$$



What are the parameters for $Er_2Ti_2O_7$?

Curnoe 2008, Ross Savary Gaulin Balents 2011

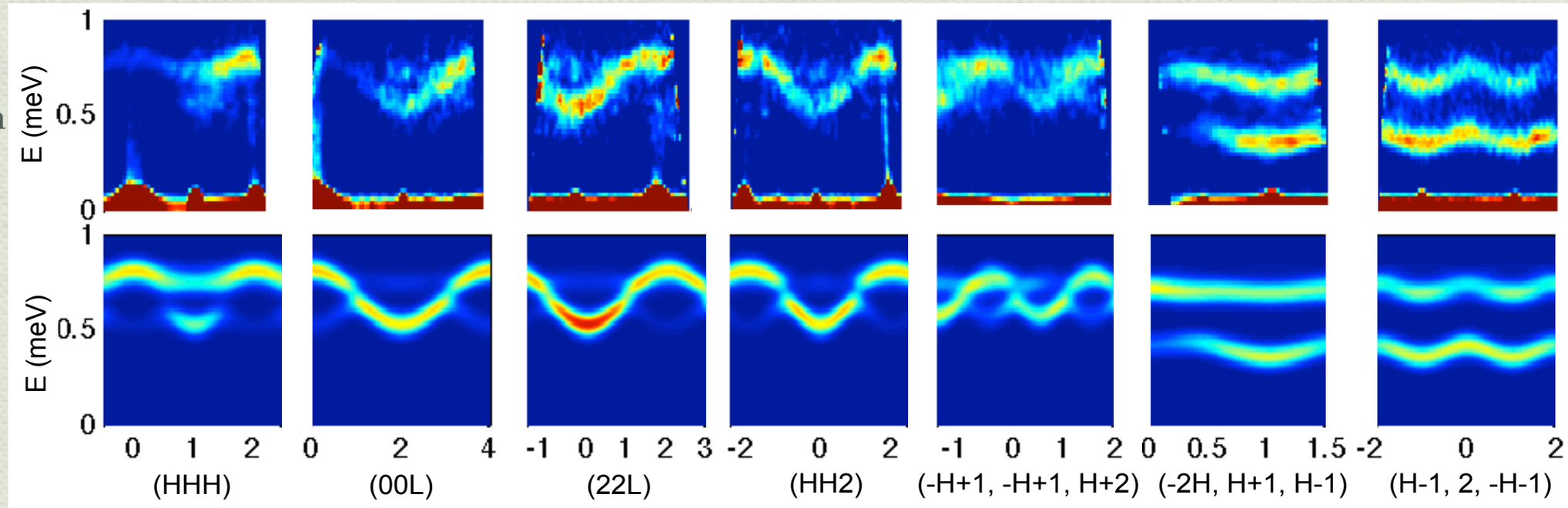
Fits to experiments: $Er_2Ti_2O_7$ parameters

recall Kate's talk on $Yb_2Ti_2O_7$ this morning

$H = 3$ T **H**//110

inelastic **neutron scattering** expt

spin wave theory



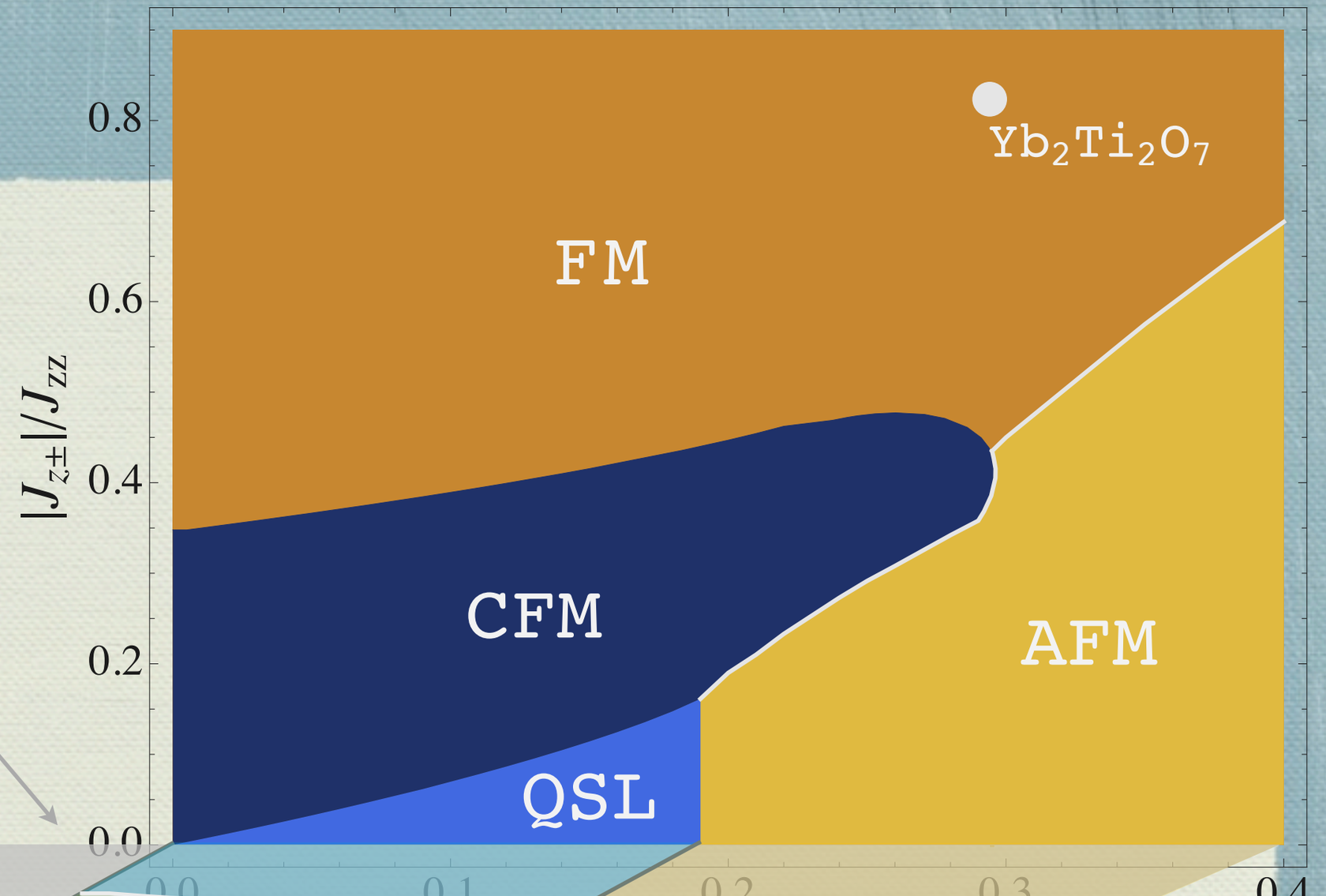
$$J_{zz} = -2.5\,10^{-2},\quad J_{\pm} = 6.5\,10^{-2},\quad J_{z\pm} = -0.88\,10^{-2},\quad J_{\pm\pm} = 4.2\,10^{-2}\ \text{meV}$$

What are the zero-field ground states of this Hamiltonian?

Phase diagrams of pyrochlores

SungBin Lee
poster tomorrow

$Er_2Ti_2O_7$



What are the ground states of $Er_2Ti_2O_7$?

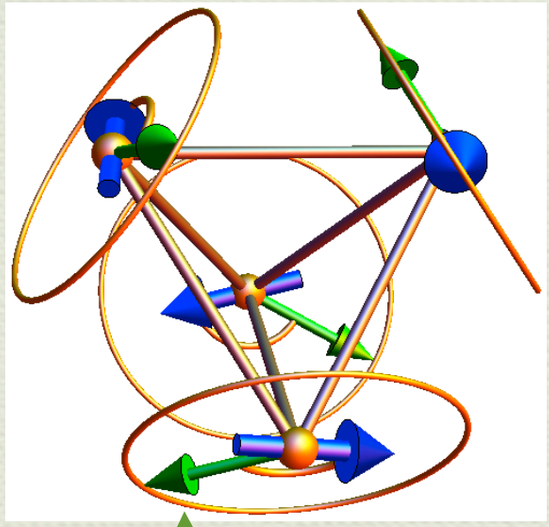
Ross, Savary, Gaulin and Balents 2011; Savary and Balents 2012; Lee, Onoda and Balents 2012

The H=0 ground states

- semi-classical $\mathbf{k} = \mathbf{0}$ *ordered* states
- minimize *classical* energy: find E_{min} for:

$$\mathbf{S}_i = \frac{1}{2}\left(\cos\alpha\,\hat{\mathbf{a}}_i + \sin\alpha\,\hat{\mathbf{b}}_i\right)$$

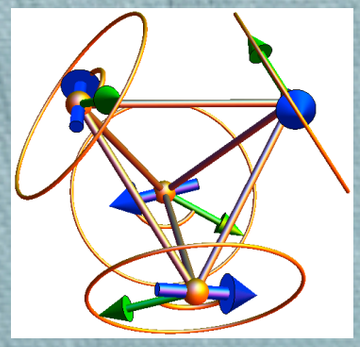
U(1) degeneracy



{ Goldstone mode in neutron scattering
{ T^3 behavior of C_V
order-by-disorder

→ Is this degeneracy robust?

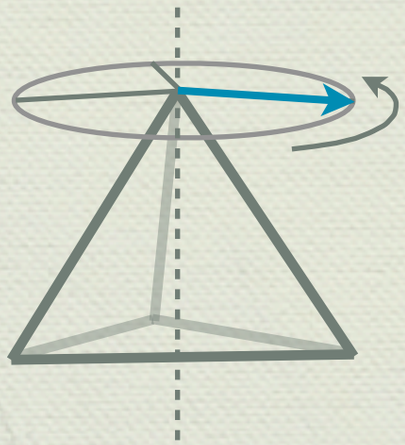
Proof of the robustness



- Ansatz $\mathbf{S}_i(\alpha) = \frac{1}{2}\mathrm{Re}\left[e^{-i\alpha}\left(\hat{\mathbf{a}}_i + i\hat{\mathbf{b}}_i\right)\right]$
- Define $\Phi = \frac{1}{2}e^{i\alpha}$
- quadratic Hamiltonian => $E[\Phi] = a\Phi^2 + a^*\Phi^2 + b|\Phi|^2$

independent of α

- C_3 rotations



$$\Phi \to e^{2i\pi/3}\Phi \quad \Rightarrow \quad a = 0$$

This uses the discrete symmetries of *H* only

QED

What exactly we showed

- classically, the ground-state degeneracy cannot be lifted
  - includes: long-range (dipolar) interactions, multi-spin terms (< 6^{th} order = negligible), spin-lattice couplings

What exactly we showed

- classically, the ground-state degeneracy cannot be lifted
  - includes: long-range (dipolar) interactions, multi-spin terms (< 6^{th} order = negligible), spin-lattice couplings
- But: "environment" effects = order-by-disorder (!) can do the job

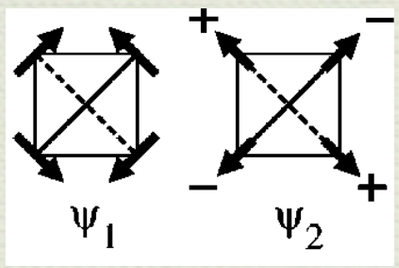
Really just two symmetry-allowed possibilities $E = b|\Phi|^2 + ...$

Leading-order term allowed by symmetry:

$$E_6 = -c\,(\Phi^6 + (\Phi^*)^6) \sim \pm\cos 6\alpha$$

six-fold degeneracy

- The *sign* of c determines how the degeneracy is lifted
  - $c > 0 : \alpha = n\,\pi/3$ - "ψ_2" states
  - $c < 0 : \alpha = (n + 1/2)\,\pi/3$ - "ψ_1" states



- The *magnitude* of c determines the gap (later...)

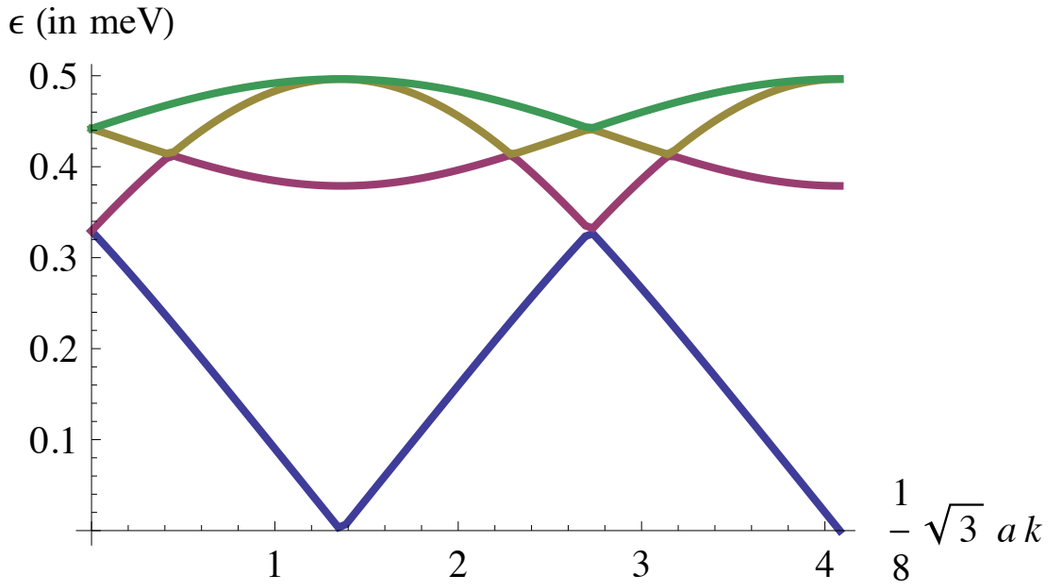
Let's look at all of this!

Quantum order-by-disorder

- zero-point energy

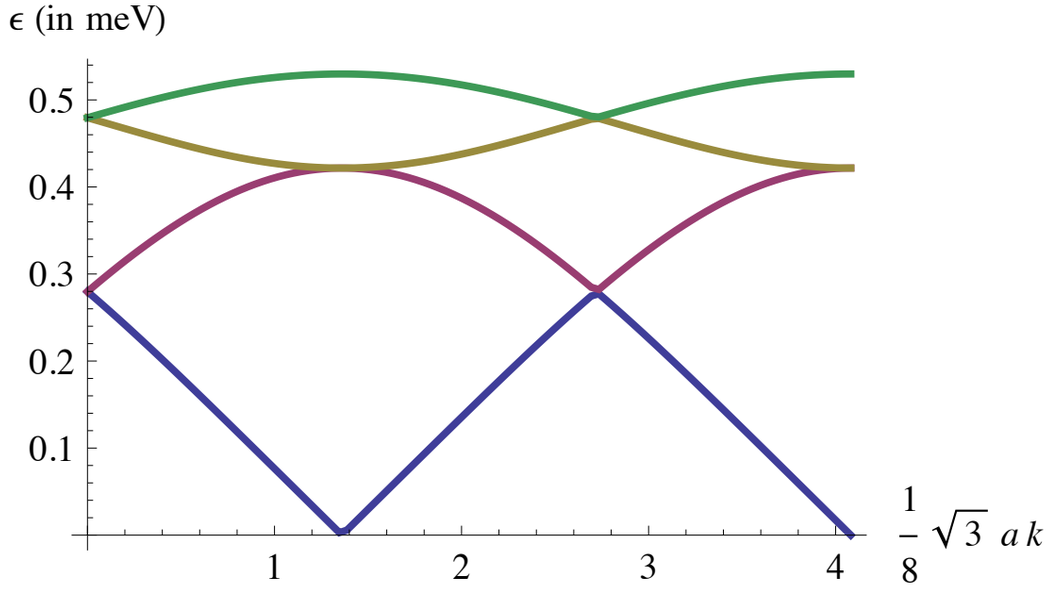
$$\epsilon_0^{sw} = \frac{1}{V_{\mathrm{BZ}}} \sum_{i=1}^{4} \int_{\mathbf{k}\in \mathrm{BZ}} \frac{\omega_{\mathbf{k}}^i}{2}$$

$\left\{\left\{\frac{3\sqrt{3}\,\pi}{4}, \frac{3\sqrt{3}\,\pi}{4}, k\right\}, \alpha = 0\right\}$



ψ_2

$\left\{\left\{\frac{3\sqrt{3}\,\pi}{4}, \frac{3\sqrt{3}\,\pi}{4}, k\right\}, \alpha = \frac{\pi}{6}\right\}$



ψ_1

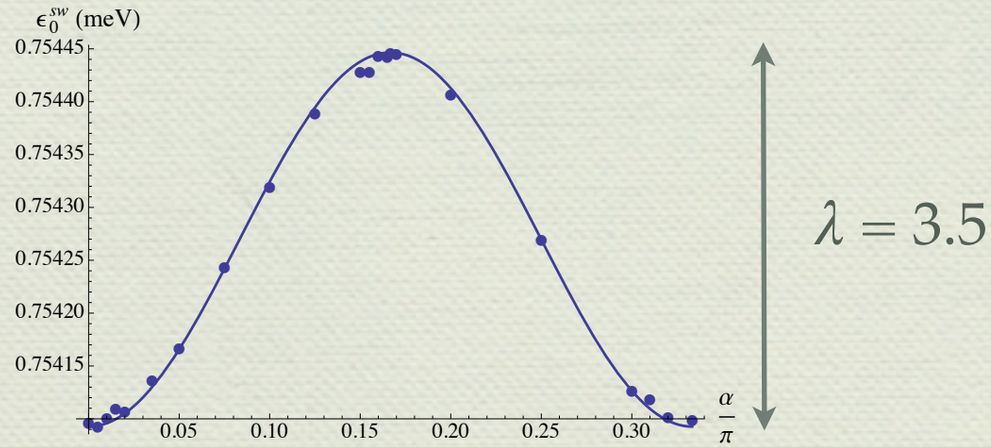
Quantum order-by-disorder

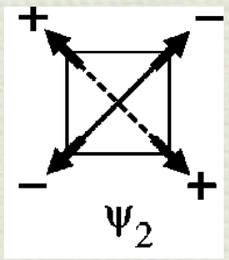
- zero-point energy

$$\epsilon_0^{sw} = \frac{1}{V_{\mathrm{BZ}}} \sum_{i=1}^{4} \int_{\mathbf{k}\in \mathrm{BZ}} \frac{\omega_{\mathbf{k}}^i}{2}$$

$$\epsilon_0^{sw} \sim -\frac{\lambda}{2} \cos 6\alpha$$

six-fold degeneracy, $\lambda > 0$



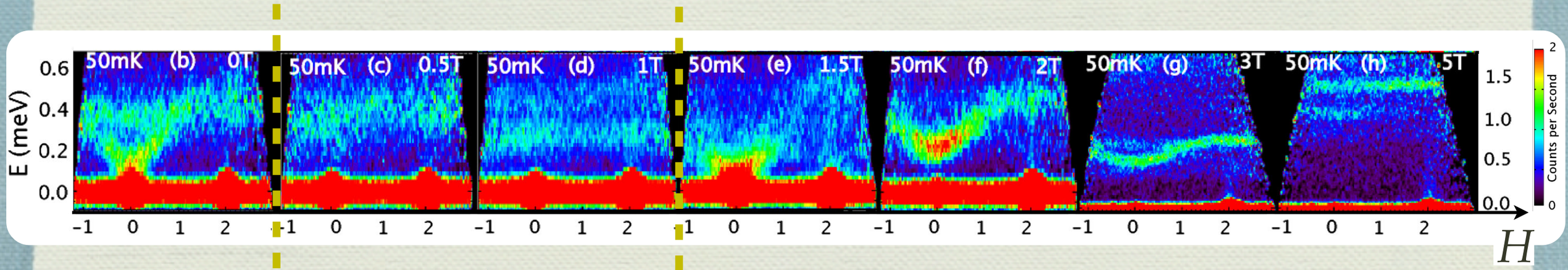


- recall $E_6 = -c\left(\Phi^6 + (\Phi^*)^6\right)$ $c = 32 N_{u.c.} \lambda$

- note: *non*-field-cooled sample => domain (6 types) formation

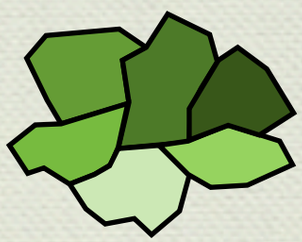
Evolution of the ground state degeneracy with a field

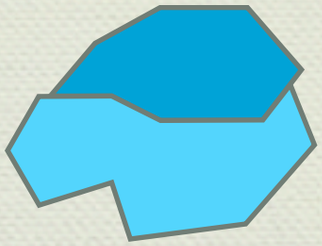
H//110



6 ground states — 2 ground states — 1 ground state

$H_c = 1.74$ T



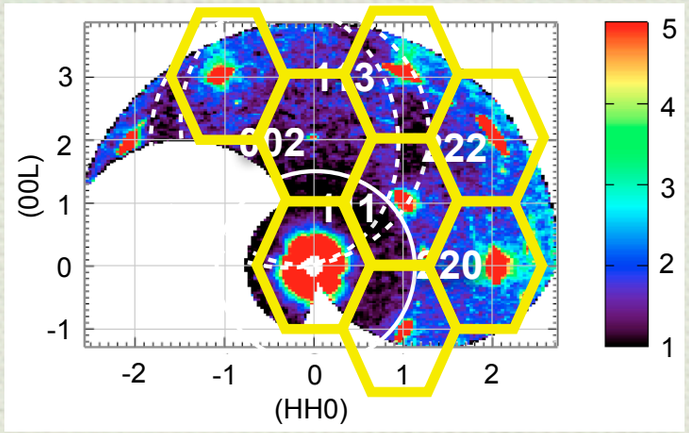


domain formation in non-field-cooled samples

Bragg peak intensity a priori depends on the domain

Confirmation of quantum order-by-disorder

- Bragg peaks:

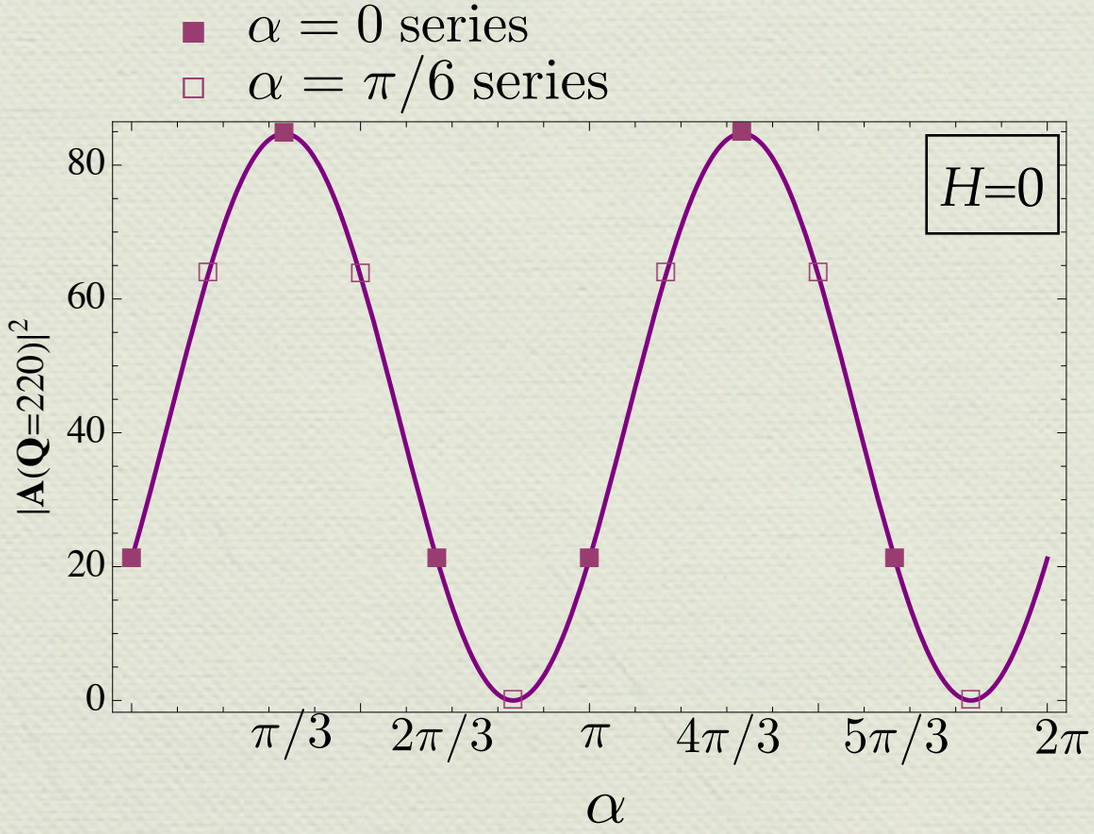


- elastic intensity:

$$\mathcal{I}(\mathbf{k},\omega=0) \propto \left|\hat{\mathbf{k}} \times \left(\hat{\mathbf{k}} \times \mathbf{A}_{\mathbf{k}}\right)\right|^2$$

$$\mathbf{A}_{\mathbf{k}} = \left\langle 0 \left| \sum_{a=0}^{3} \mathbf{M}_a(\mathbf{k}) \right| 0 \right\rangle$$

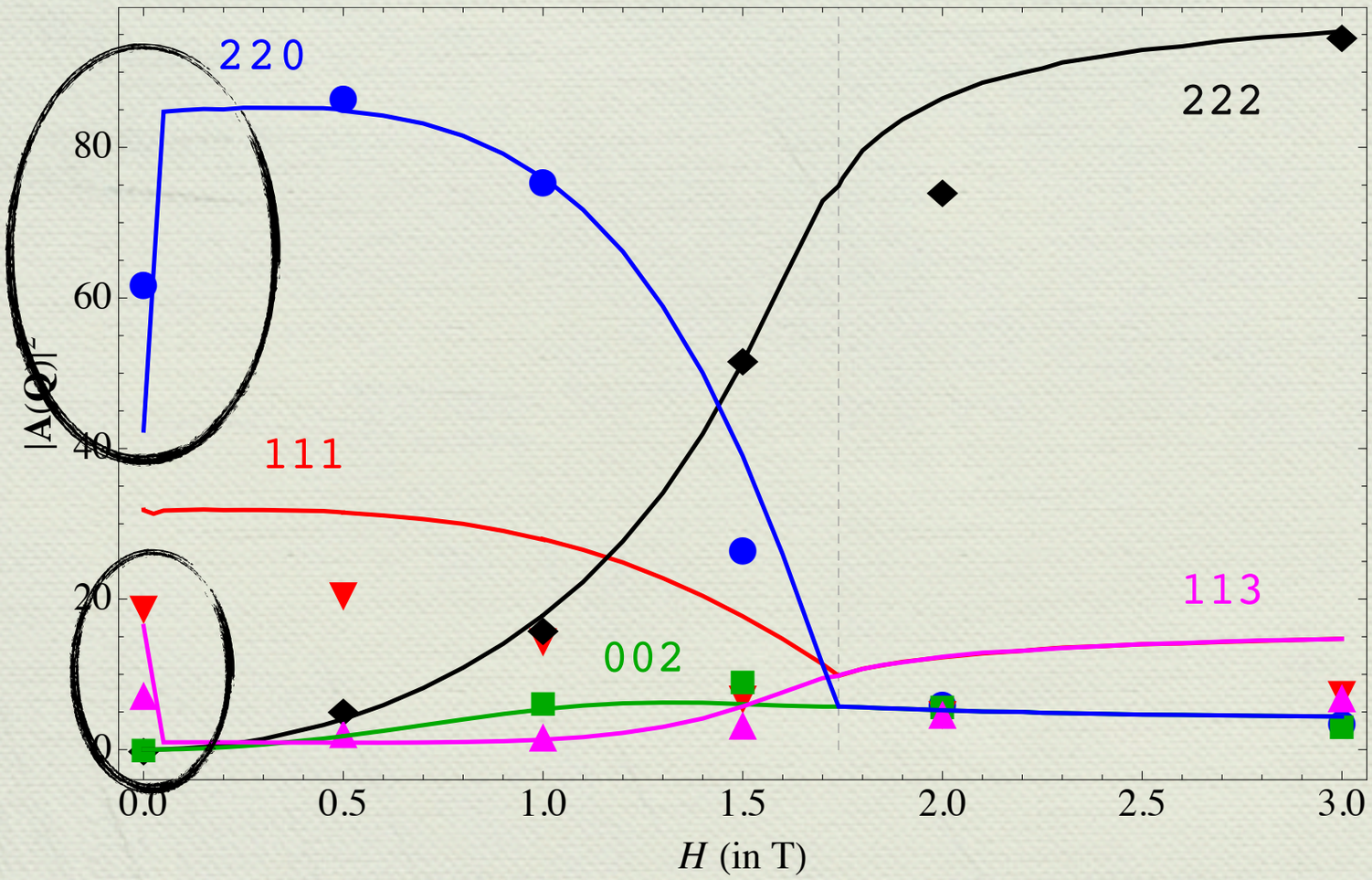
depends on α



I_0=2L+4S

Confirmation of quantum order-by-disorder

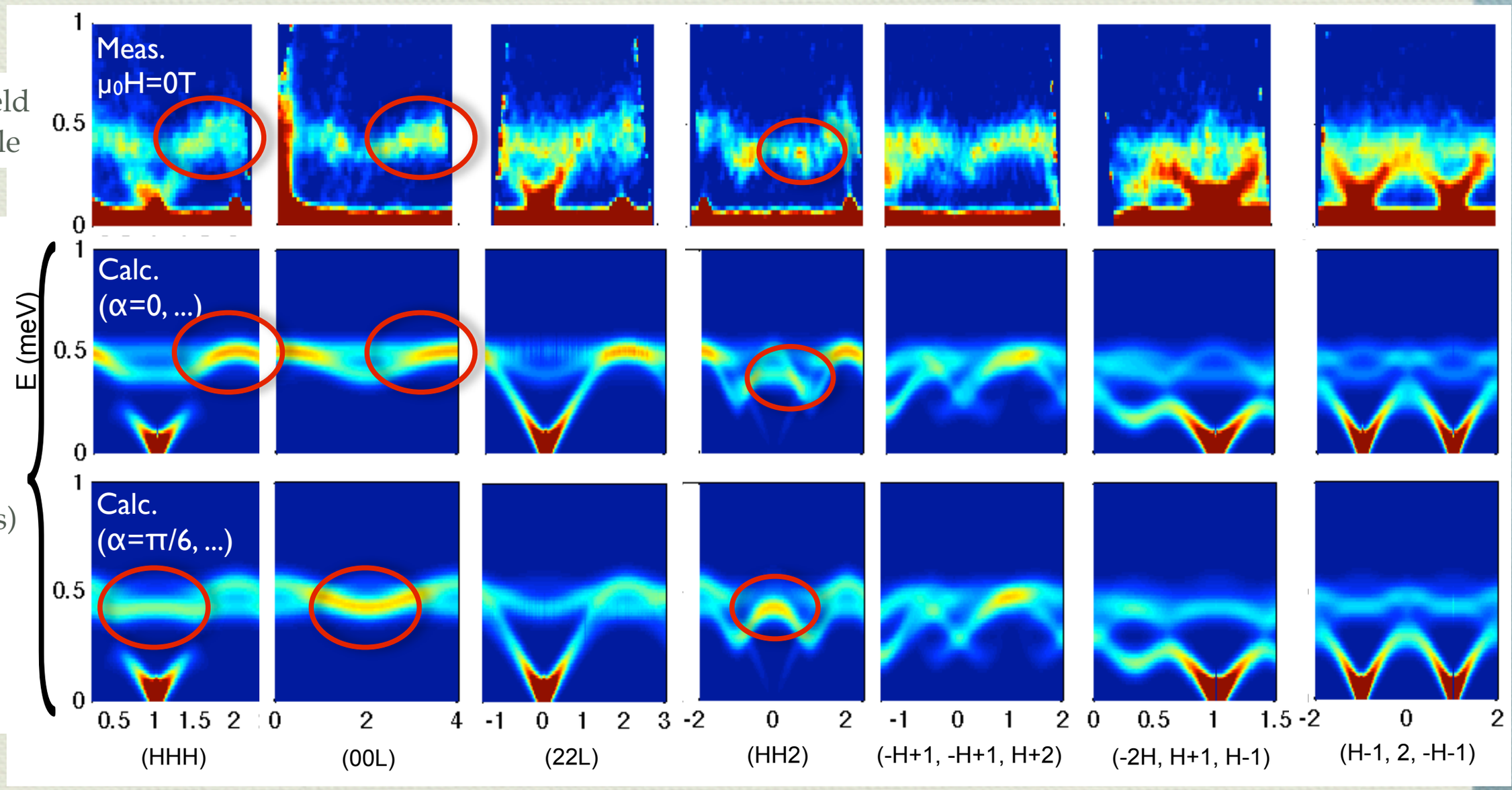
H//110

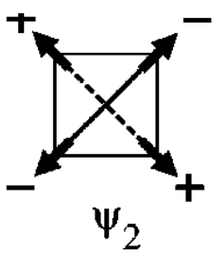


"Direct" inelastic structure factor comparison

$H = 0$

INS of zero-field -cooled sample





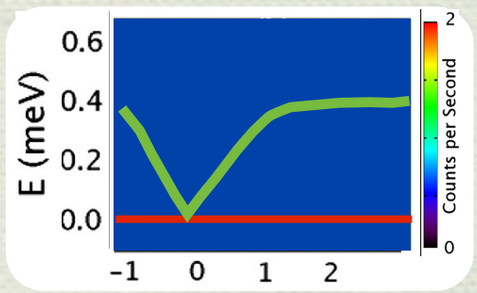
ψ_2

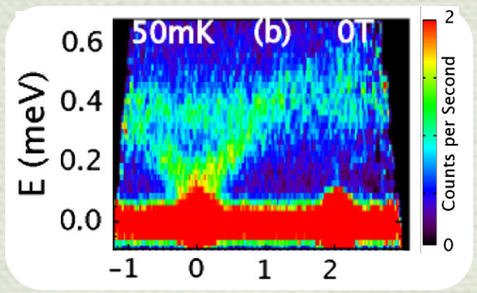
spin wave theory (signal from 6 domains)

ψ_1

continuous degeneracy (no fluctuations)

- gapless Goldstone-like mode





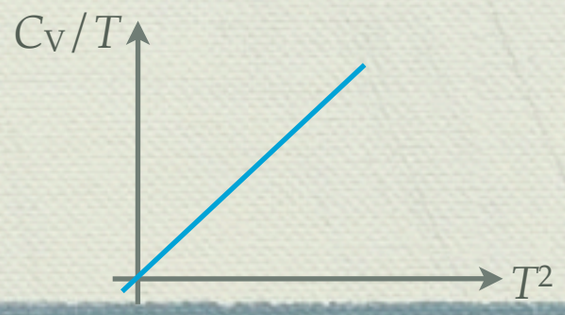
resolution = 0.09 meV

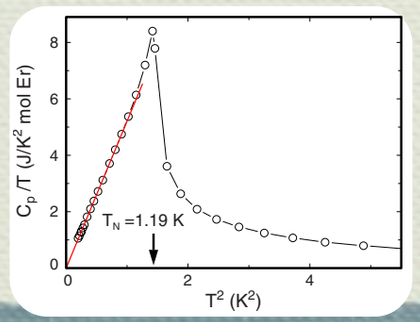
- low-energy theory:

$$\mathcal{S}_0 = \frac{1}{2}\int \frac{d^3r}{v_{u.c.}} d\tau \left[\kappa(\boldsymbol{\nabla}\alpha)^2 + \eta(\partial_\tau \alpha)^2\right]$$

$$\omega_{\mathbf{k}} = c\,k$$

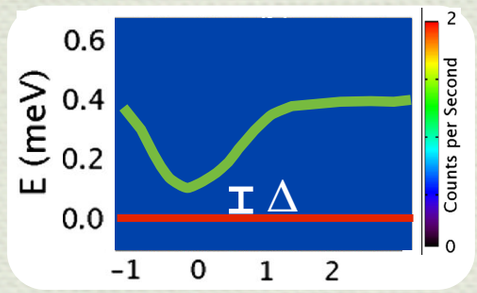
- T^3 specific heat





weakly-lifted C^0 degeneracy (order-by-disorder)

- gapped pseudo-Goldstone mode



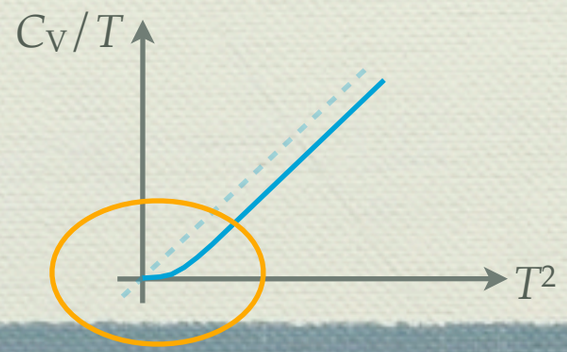
need high resolution

- low-energy theory:

$$\mathcal{S} = \frac{1}{2}\int \frac{d^3r}{v_{u.c.}} d\tau \left[\kappa(\boldsymbol{\nabla}\alpha)^2 + \eta(\partial_\tau \alpha)^2 + m^2\alpha^2\right]$$

$$\omega_{\mathbf{k}} = \sqrt{c^2\,k^2 + \Delta^2}$$

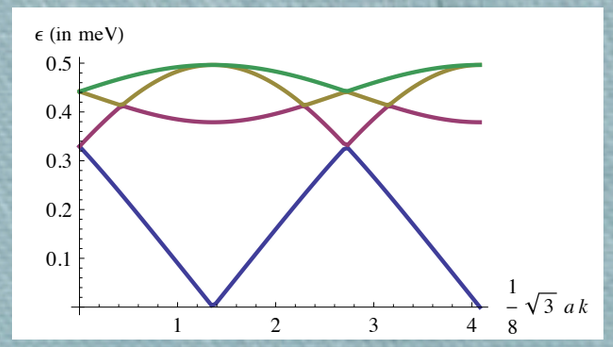
- no T^3 behavior very close to $T = 0$



need very low T data

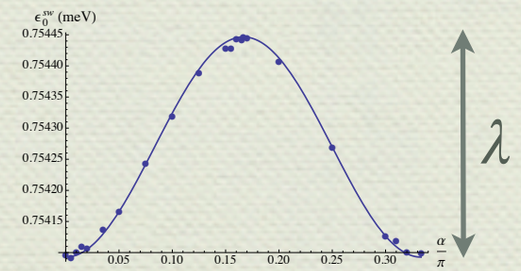
Ruff *et al.* 2008, Sosin *et al.* 2010

The gap: we know everything about the spin wave spectrum

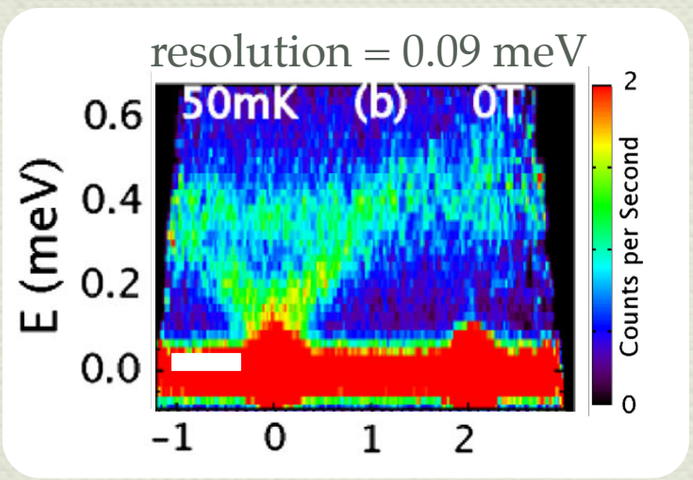


$$\mathcal{S} = \frac{1}{2}\int \frac{d^3r}{v_{u.c.}} d\tau \left[\kappa(\boldsymbol{\nabla}\alpha)^2 + \eta(\partial_\tau \alpha)^2 - \lambda\cos 6\alpha\right] \qquad \omega_{\mathbf{k}} = \sqrt{\frac{\kappa}{\eta}\mathbf{k}^2 + \Delta^2}$$

- η and $\varkappa$: extracted by expanding the spin-wave theory
- λ: extracted by calculating the zero-point energy from spin-wave theory



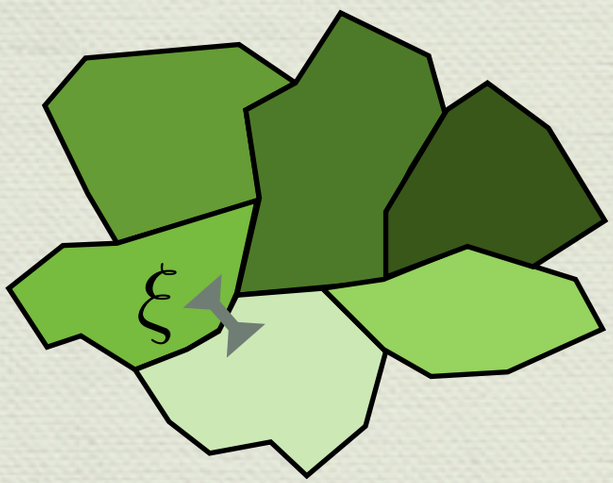
$$\Delta = \sqrt{\frac{18\lambda}{\eta}} \approx 0.02\,\text{meV} = 260\,\text{mK}$$



data from Ruff *et al.* 2008

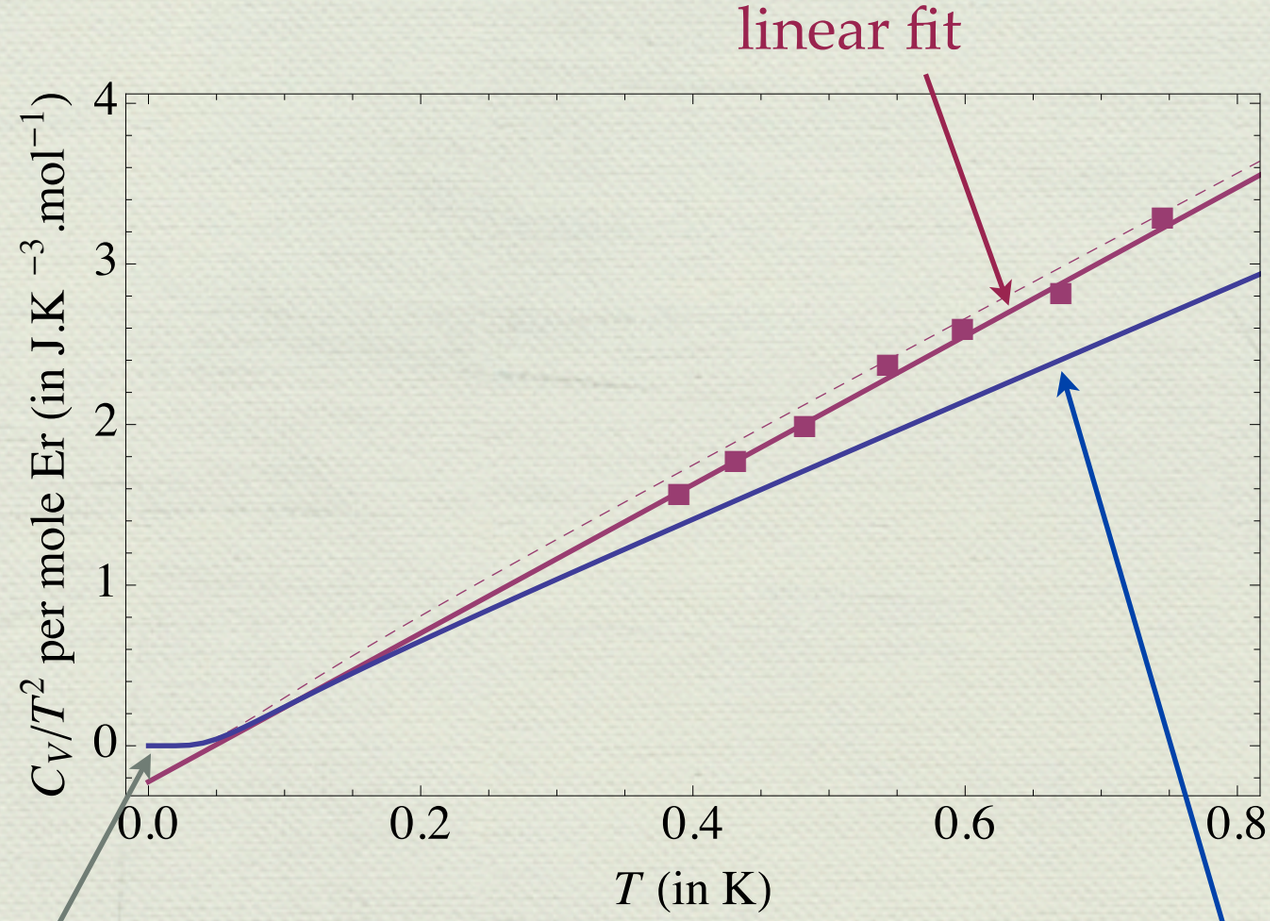
Domain wall width

$$\mathcal{S} = \frac{1}{2}\int \frac{d^3r}{v_{u.c.}} d\tau \left[\kappa(\boldsymbol{\nabla}\alpha)^2 + \eta(\partial_\tau \alpha)^2 - \lambda\cos 6\alpha\right]$$



$$\xi = \sqrt{\frac{\kappa}{18\lambda}} \approx 2a \approx 20\,\text{Å}$$

The specific heat



$$C_V \sim 4N_{u.c.}\sigma T^3 \qquad k_B T \gg \Delta$$

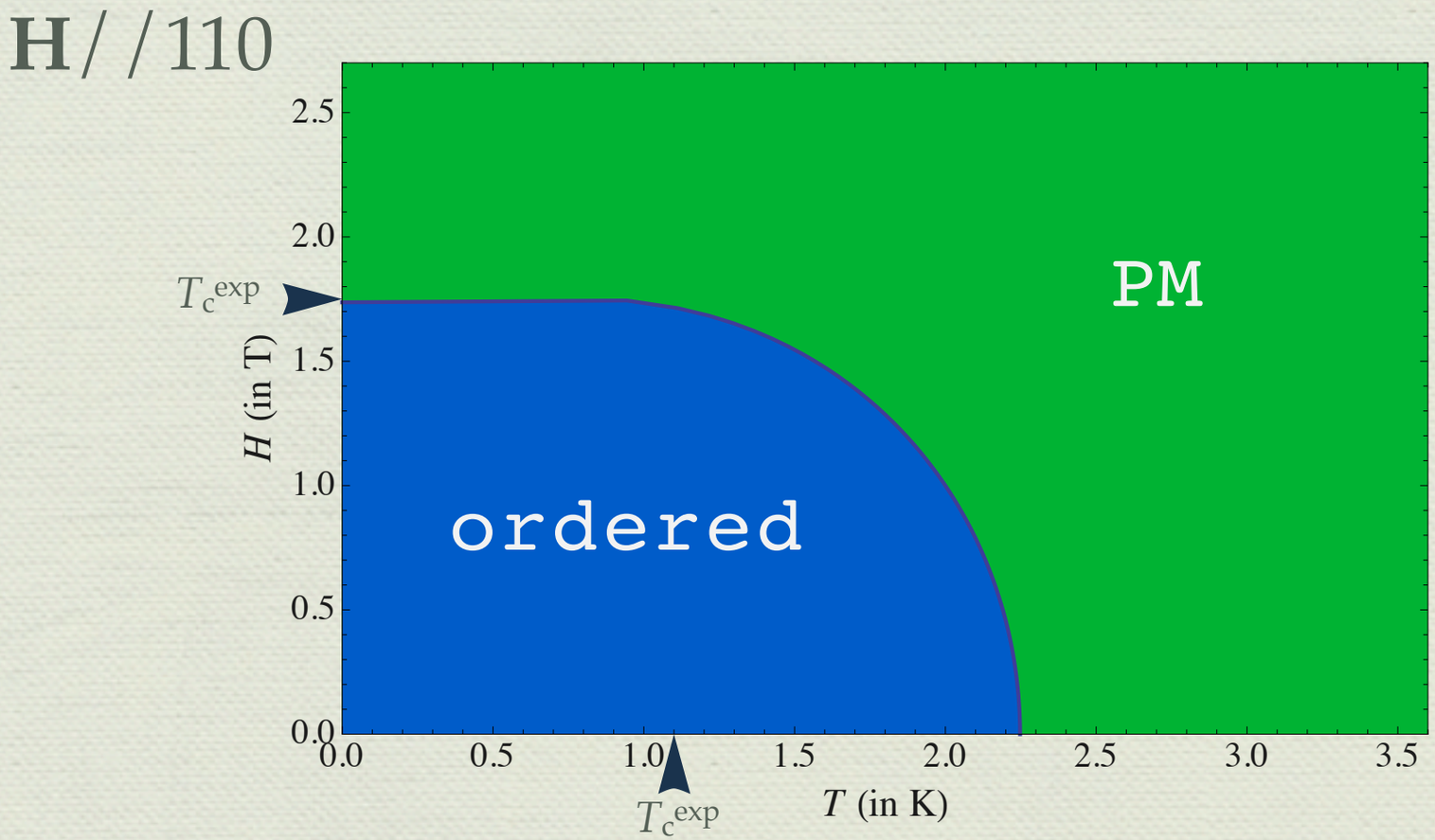
$$\sigma = \frac{k_B^4 \pi^2 a^3}{120\,\overline{v}^3}$$

obtained from spin wave theory

$$\sigma \approx 3.6\ \mathrm{J.K^{-4}.mol^{-1}}$$

data from Ruff *et al.* 2008

Further considerations: MFT



$H_c^{exp} = 1.75$ T — perfect agreement

$T_c^{exp} = 1.1$ K — $f = 2.1$ cf. MFT neglects fluctuations

- 110 field: 2nd order phase transition (Ising)
- 111 field: 1st order (Potts)
- etc.

Conclusions and perspectives

- first definitive proof of the experimental realization of order-by-disorder: so far examples where result could always be disputed, including in ETO - full Hamiltonian, no room for speculations
- conclusive determination of quantumness
- calculation of associated quantities
- other materials and lattices (kagomé?)
- needed experiments:
  - experiments on **field-cooled samples**
  - **lower-temperature specific heat**
  - more **field directions**
  - **gap: NMR?**
  - **gap**: **higher resolution** neutron scattering

Thank you for your attention