Definitive Evidence for Order-by-Quantum-Disorder

Lucile Savary

Hamilton, June 4th, 2012
Collaborators

Leon Balents (KITP)
Kate Ross
Bruce Gaulin (Argonne)
Jacob Ruff

experiments, McMaster
What is order-by-disorder?

- 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

$$H = J_1 \sum_{\langle i,j \rangle} S_i \cdot S_j + J_2 \sum_{\langle \langle i,j \rangle \rangle} S_i \cdot S_j$$

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

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

"U(1)" degeneracy
Order-by-disorder

- those states have the same energy
- but different "environment"
Order-by-disorder: an example

- picture

\[
T = 0 \quad H_{\text{eff}} = 0 \quad T > 0 \quad H_{\text{eff}} \neq 0
\]

- thermal fluctuations: entropy

\[
F = E - TS(\phi) = F(\phi) \text{ minimized for } \phi = 0 \text{ or } \pi
\]

The degeneracy is lifted in the free energy by the entropy
Order-by-disorder: an example

*quantum fluctuations: zero-point energy*

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

\[ \hbar \omega_k = \sqrt{\frac{A_k}{m}} \]

\[ E_{\text{zero-point}} = \sum_k \frac{\hbar \omega_k}{2} \sim \frac{1}{\sqrt{2m}} \sum_k \sqrt{A_k} \]

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!

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

We show definitive evidence for quantum ObD in Er$_2$Ti$_2$O$_7$. 
Outline

- Er$_2$Ti$_2$O$_7$
- Review of "older" literature
- Hamiltonian of Er$_2$Ti$_2$O$_7$
- Degeneracy
- Comparison with experiments
- Verifiable consequences

based on arXiv 1204.1320 (to appear in PRL)
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

The Allnighter

The Zone!

Existential Crisis! What am I doing with my life??

Take our time to finish

Take our time to finish

I don’t want to do this

Resignation

See Leon’s email: "We are too slow"

Realize you’re not sleeping tonight

Email party with Kate

Cue Hard Rock Music Montage

Meet with Leon

Submission... Oh my!!

Scrounge for food.

The Post Allnighter High. Life is awesome!! You want to kiss everyone you meet!

11:00pm

1:30pm

5:30am

1pm

arXiv Deadline

Leon’s Pep Email

The crash

You’re not as young as you used to be.

Maybe you should have started earlier.

ZZZZZZZZ...
The Story

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THE ALLNIGHTER

Definitive Evidence for Order–by–Quantum–Disorder in Er2Ti2O7
Lucile Savary, Kate A. Ross, Bruce D. Gaulin, Jacob P. C. Ruff, Leon Balents
(Submitted on 5 Apr 2012)

arXiv 1204.0595

>>originally published 4/6/2012

arXiv 1204.1320

725 papers
Er$_2$Ti$_2$O$_7$

- **Er$^{3+}$ pyrochlore**
- **local z-axes**
- **local XY-plane**

- rare-earth pyrochlore family: Ho$_2$Ti$_2$O$_7$, Dy$_2$Ti$_2$O$_7$, Ho$_2$Sn$_2$O$_7$, Dy$_2$Sn$_2$O$_7$, Er$_2$Ti$_2$O$_7$, Yb$_2$Ti$_2$O$_7$, Tb$_2$Ti$_2$O$_7$, Er$_2$Sn$_2$O$_7$, Tb$_2$Sn$_2$O$_7$, Pr$_2$Sn$_2$O$_7$, Nd$_2$Sn$_2$O$_7$, Gd$_2$Sn$_2$O$_7$, ...

- 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

**behaviors:**
- spin ices
- quantum AFM
- quantum spin liquids?
Er$_2$Ti$_2$O$_7$: Previous studies

- $k = 0$ order
- compatible with $\psi_1$ or $\psi_2$ states
- specific heat: $T^3$ behavior - Goldstone?
- Goldstone mode
**Er₂Ti₂O₇: Previous studies**

- **model:**
  
  \[ H = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + D \left( \sum_i \mathbf{S}_i \cdot \mathbf{\hat{e}}_i \right)^2 \]

- has a huge degeneracy

- order-by-disorder suggested

- dipolar interactions invoked

- effect of multi-spin terms

- anisotropy necessary

- not 1\textsuperscript{st} order transition?

- somewhat ad hoc

- insightful!

- "Palmer-Chalker state" not experimental ground state

- very small

- puzzle

- Champion \textit{et al.}

- McClarty \textit{et al.}

- Stasiak \textit{et al.}

- Cao \textit{et al.}

- McClarty \textit{et al.}

- Stasiak \textit{et al.}
Er$_2$Ti$_2$O$_7$: Previous studies

- model:  
  $H = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + D \left( \sum_i \mathbf{S}_i \cdot \hat{e}_i \right)^2$
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  - order-by-disorder suggested
  - dipolar interactions invoked
  - effect of multi-spin terms
  - anisotropy necessary

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- somewhat ad hoc
- insightful!
- "Palmer-Chalker state" not experimental ground state
- very small

Need a more accurate Hamiltonian
Er$_2$Ti$_2$O$_7$: towards a Hamiltonian for rare-earth oxides with intrinsic strong spin-orbit coupling.

A$_2$B$_2$O$_7$: strong crystal fields with discrete cubic symmetries only.

Space group: Fd-3m, i.e. #227.
Er$_2$Ti$_2$O$_7$: towards a Hamiltonian

rare-earths : intrinsic strong spin-orbit coupling

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C$_3$
mirror
inversion
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mirror

inversion

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C$_3$

mirror

inversion

time reversal
Er$_2$Ti$_2$O$_7$: most general NN effective spin-1/2 Hamiltonian

- strong SOC & crystal fields $\Rightarrow J = L+S = 15/2, \Delta \sim 75$ K

- spin-1/2 Hamiltonian:

$$H = \sum_{\langle i,j \rangle} \left[ J_{zz} S_i^z S_j^z ight. $$

$$- J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+)$$

$$+ J_{Z\pm} \left[ S_i^z (\zeta_{ij} S_j^+ + \zeta_{ij}^* S_j^-) + i \leftrightarrow j \right] $$

$$+ J_{\pm\pm} \left[ \gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^- \right] $$

What are the parameters for Er$_2$Ti$_2$O$_7$?
Fits to experiments: Er$_2$Ti$_2$O$_7$

parameters

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

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

recall Kate's talk on Yb$_2$Ti$_2$O$_7$ this morning

$H = 3$ T $\mathbf{H}//110$

inelastic neutron scattering expt

spin wave theory

E (meV)

E (meV)
Phase diagrams of pyrochlores

What are the ground states of \( \text{Er}_2\text{Ti}_2\text{O}_7 \)?

SungBin Lee
poster tomorrow

Ross, Savary, Gaulin and Balents 2011; Savary and Balents 2012; Lee, Onoda and Balents 2012
The H=0 ground states

- semi-classical \( k = 0 \) ordered states
- minimize classical energy: find \( E_{\text{min}} \) for:

\[
S_i = \frac{1}{2} \left( \cos \alpha \, \hat{a}_i + \sin \alpha \, \hat{b}_i \right)
\]

\( U(1) \) degeneracy

Goldstone mode in neutron scattering
\( T^3 \) behavior of \( C_V \)
order-by-disorder

Is this degeneracy robust?
Proof of the robustness

- Ansatz: \( S_i(\alpha) = \frac{1}{2} \text{Re} \left[ e^{-i\alpha} \left( \hat{a}_i + i\hat{b}_i \right) \right] \)
- Define: \( \Phi = \frac{1}{2} e^{i\alpha} \)
- Quadratic Hamiltonian: \( \Rightarrow \quad E[\Phi] = a\Phi^2 + \alpha^*\Phi^2 + b|\Phi|^2 \)

- \( C_3 \) rotations:
  \[ \Phi \rightarrow e^{2i\pi/3}\Phi \quad \Rightarrow \quad a = 0 \]

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 ($< 6^{\text{th}}$ order = negligible), spin-lattice couplings
- But: "environment" effects = order-by-disorder (!) can do the job
Really just two symmetry-allowed possibilities: 

$$ E = b|\Phi|^2 + \ldots $$

Leading-order term allowed by symmetry:

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

- **The sign of c** determines how the degeneracy is lifted

  - **c > 0 :** $\alpha = n \pi/3$ - "$\psi_2$" states
  - **c < 0 :** $\alpha = (n + 1/2) \pi/3$ - "$\psi_1$" states

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

Let's look at all of this!
Quantum order-by-disorder

zero-point energy

\[ \epsilon_0^{sw} = \frac{1}{V_{BZ}} \sum_{i=1}^{4} \int_{k \in BZ} \frac{\omega_k}{2} \]

\[ \{ \left\{ \frac{3\sqrt{3}}{4}, \frac{3\sqrt{3}}{4}, k \right\}, \alpha = 0 \} \]

\[ \{ \left\{ \frac{3\sqrt{3}}{4}, \frac{3\sqrt{3}}{4}, k \right\}, \alpha = \frac{\pi}{6} \} \]
Quantum order-by-disorder

- **zero-point energy**

\[ \epsilon_0^{sw} = \frac{1}{V_{BZ}} \sum_{i=1}^{4} \int_{\mathbf{k} \in BZ} \frac{\omega_k^i}{2} \]

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

six-fold degeneracy, \( \lambda > 0 \)

- **recall**

\[ E_6 = -c (\Phi^6 + (\Phi^*)^6) \quad c = \frac{32N_{u.c.}}{\lambda} \]

- **note:** non-field-cooled sample => domain (6 types) formation
Evolution of the ground state degeneracy with a field

$H/\parallel 110$

6 ground states - 2 ground states $H_c = 1.74$ T - 1 ground state

domain formation in non-field-cooled samples

Bragg peak intensity a priori depends on the domain
Confirmation of quantum order-by-disorder

**Bragg peaks:**

\[ I(\mathbf{k}, \omega = 0) \propto \left| \mathbf{\hat{k}} \times (\mathbf{\hat{k}} \times \mathbf{A}_k) \right|^2 \]

**elastic intensity:**

\[ \mathbf{A}_k = \left\langle 0 \left| \sum_{\alpha=0}^{3} \mathbf{M}_\alpha(\mathbf{k}) \right| 0 \right\rangle \]

depends on \( \alpha \)

\[ I_0 = 2L + 4S \]
Confirmation of quantum order-by-disorder

$H \parallel 110$
"Direct" inelastic structure factor comparison

\[ H = 0 \]

INS of zero-field cooled sample

Spin wave theory (signal from 6 domains)
continuous degeneracy (no fluctuations)

- gapless Goldstone-like mode

\[ S_0 = \frac{1}{2} \int \frac{d^3r}{v_{u.c.}} d\tau \left[ \kappa (\nabla \alpha)^2 + \eta (\partial_\tau \alpha)^2 \right] \]

\[ \omega_k = c k \]

- low-energy theory:

- \( T^3 \) specific heat

\[ C_v/T \quad \sim \quad T^2 \]

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

- gapped pseudo-Goldstone mode

need high resolution

\[ \omega_k = \sqrt{c^2 k^2 + \Delta^2} \]

- low-energy theory:

- \( T^3 \) specific heat

\[ C_v/T \quad \sim \quad T^2 \]

- 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

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

\[ \omega_k = \sqrt{\frac{\kappa}{\eta} k^2 + \Delta^2} \]

- \( \kappa \) and \( \eta \): extracted by expanding the spin-wave theory
- \( \lambda \): extracted by calculating the zero-point energy from spin-wave theory

\[ \Delta = \sqrt{\frac{18\lambda}{\eta}} \approx 0.02 \text{ meV} = 260 \text{ mK} \]
Domain wall width

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

\[ \xi = \sqrt{\frac{\kappa}{18\lambda}} \approx 2a \approx 20 \text{ Å} \]
The specific heat

\[ C_V \sim 4N_{u.c.}\sigma T^3 \quad k_B T \gg \Delta \]

\[ \sigma = \frac{k_B^4 \pi^2 a^3}{120 \bar{v}^3} \]

\[ \sigma \approx 3.6 \text{ J.K}^{-4}.\text{mol}^{-1} \]

Data from Ruff et al. 2008

graph showing the specific heat per mole Er (in J.K^{-3}.mol^{-1}) as a function of T (in K) with linear fit and signature of the gap. Spin-wave theory curve + gap obtained from spin wave theory.
Further considerations: MFT

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

\[ H_c^{\text{exp}} = 1.75 \, \text{T} \]  
perfect agreement

\[ T_c^{\text{exp}} = 1.1 \, \text{K} \]  
\( f = 2.1 \) cf. MFT
neglects fluctuations
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