Frustrated diamond lattice antiferromagnets

Jason Alicea (Caltech)
Doron Bergman (Yale)
Leon Balents (UCSB)
Emanuel Gull (ETH Zurich)
Simon Trebst (Station Q)

• Introduction
  – Frustration, degeneracy, & emergent phenomena
  – Diamond lattice antiferromagnets
  – Overview of experiments
• Theory of frustrated diamond lattice antiferromagnets
  – Ground states (highly degenerate spirals)
  – Stability & order-by-disorder
  – Monte Carlo simulations
  – Spiral spin liquid
• Comparison to experiment
• Summary & future directions
Frustration, degeneracy, & emergent phenomena

- Frustration = all interactions not satisfiable simultaneously

- General principle: The presence of many competing states often leads to interesting physics
  - Quantum Hall effect
  - High-$T_c$ superconductors
  - Frustrated magnets (Mott insulators)

$\Rightarrow$ Highly degenerate ground states (pyrochlore, kagome, FCC, etc.)
  - High sensitivity to perturbations
  - Spin-glass behavior
  - Spin-liquid physics
  - Order-by-disorder
At high temperatures, Curie-Weiss law holds. \[ \chi \sim \frac{1}{T - \Theta_{CW}}; \quad \Theta_{CW} \sim -(\text{Exchange } J's) \]
Experimental signatures of frustration

At high temperatures, Curie-Weiss law holds.

At low temperatures, systems typically order.

Useful diagnostic: “frustration parameter” \( f = \frac{\Theta_{CW}}{T_c} \)
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Useful diagnostic: “frustration parameter”

\[ f = \frac{|\Theta_{CW}|}{T_c} \]

Highly frustrated systems \( \Rightarrow \) \( f > 5 - 10 \)

- Key challenges: Low-\( T \) ordering mechanisms & Characterizing “spin liquid” correlations
Frustrated diamond lattice antiferromagnets: Materials

Many materials take on the normal spinel structure: $AB_2X_4$

Focus: spinels with magnetic A-sites (only)

Very limited theoretical understanding…

V. Fritsch et al. PRL 92, 116401 (2004); N. Tristan et al. PRB 72, 174404 (2005); T. Suzuki et al. (unpublished)
Frustrated diamond lattice antiferromagnets: Materials

Many materials take on the normal spinel structure: $AB_2X_4$

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CoRh$_2$O$_4$  Co$_3$O$_4$  MnSc$_2$S$_4$  FeSc$_2$S$_4$

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Frustration on the bipartite diamond lattice?

Naïve Hamiltonian:

\[ H = J_1 \sum_{\langle ij \rangle} S_i \cdot S_j \]

Classical spins (s = 3/2, 5/2 for materials of interest)

Diamond lattice = 2 FCC sublattices coupled via \( J_1 \)
Frustration on the bipartite diamond lattice??

Naïve Hamiltonian:

\[ H = J_1 \sum_{\langle ij \rangle} S_i \cdot S_j \]

No Frustration
Frustration on the **diamond lattice**

Remedy:

$2^{nd}$ neighbor exchange

$H = J_1 \sum_{\langle ij \rangle} S_i \cdot S_j + J_2 \sum_{\langle\langle ij \rangle\rangle} S_i \cdot S_j$

$J_1$ and $J_2$ expected to be comparable due to similarity in exchange paths

$T = 0$ physics: ground states

$$H = J_1 \sum_{\langle ij \rangle} S_i \cdot S_j + J_2 \sum_{\langle\langle ij \rangle\rangle} S_i \cdot S_j$$

Neel

0 1/8

$J_2 / J_1$

Useful rewrite of Hamiltonian:

$$H = J_1 \sum_t [S_0 + (S_1 + S_2 + S_3 + S_4)/4]^2$$

$$+ (J_2 - J_1/8) \sum_{\langle\langle ij \rangle\rangle} S_i \cdot S_j$$
$T = 0$ physics: ground states

$$H = J_1 \sum_{\langle ij \rangle} S_i \cdot S_j + J_2 \sum_{\langle\langle ij \rangle\rangle} S_i \cdot S_j$$

Highly degenerate coplanar spirals

Direction & pitch of spirals characterized by a wavevector residing on a surface in momentum space!

"Spiral surfaces"

$J_2 / J_1 = 0.2$
$T = 0$ physics: ground states

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

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$J_2 / J_1 = 0.2$

$J_2 / J_1 = 0.4$
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Highly degenerate coplanar spirals

Neel

Direction & pitch of spirals characterized by a wavevector residing on a surface in momentum space!

“Spiral surfaces”

$J_2/J_1 = 0.2$  $J_2/J_1 = 0.4$  $J_2/J_1 = 0.85$
\[ H = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle ij \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j \]

Direction & pitch of spirals characterized by a wavevector residing on a surface in momentum space!

“Spiral surfaces”
Low-\(T\) physics: *Can long-range order occur?*

Stability nontrivial due to massive spiral degeneracy.

- Expand Hamiltonian in fluctuations:

\[
\delta S_i = S_i - \langle S_i \rangle
\]

\((\text{Arbitrary ground state order})\)

- At \(T = 0\), branch of normal modes has infinite # of zeros!

\[
\omega_0(q) = 0\quad \text{For all } q\text{ on surface}
\]

\[
\langle \delta S_i^2 \rangle \sim T \int \frac{d^3q}{\omega_0^2(q)} \rightarrow \infty
\]

Naively, fluctuations *diverge*
"Order-by-disorder" stabilization

- **Key ideas:**
  - Only symmetry-required zeros in $\omega_0(q)$ are the "Goldstone modes"
  - Thermal fluctuations lifts the remaining "accidental" zeros $\Rightarrow$ entropy stabilizes long-range order!

- **Needed:** finite-$T$ corrections to $\omega_0(q)$ on "spiral surface"
  - Perturbation theory insufficient
  - Use self-consistent approach instead

- **Answer:**
  $$\omega_T^2(q) = \omega_0^2(q) + T^{2/3} \Sigma(q)$$

  $$\left\langle S^2_i \right\rangle \sim T \int \frac{d^3q}{\omega_T^2(q)} \sim T^{1/3}$$

- **Non-analytic $T$-dependence $\Rightarrow$ unconventional thermodynamic behavior, e.g.,**
  $$C_v = A + BT^{1/3}$$
Aside on self-consistent approach

- Expand Hamiltonian in fluctuations:
  \[ \delta S_i = S_i - \langle S_i \rangle; \quad H = H_2 + H_3 + H_4 + \ldots \]
  
  “Interaction” terms

- Get self-energy self-consistently for divergent mode
  \[ \Sigma(q) = \text{Full propagator} \]

- For \( q \) on surface, assume
  \[ \Sigma(q) \sim T^\alpha \Sigma(q) \]

\[ \omega_T^2(q) = \omega_0^2(q) + T^{2/3} \Sigma(q) \]
“Order-by-disorder” selection

Long-range order occurs—but which state does entropy select?

- Need Free Energy for all \( Q \) on spiral surface
  \[
  F(Q) = E - TS(Q)
  \]
- Entropy favors states with highest density of nearby low-energy states
- Complex phase structure emerges:

![Diagram showing various states and energy levels with labels for entropy and free energy minima, and color-coding for minimum, low, and high energy states.](image-url)
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\(1/8\) \(1/4\) \(~1/2\) \(~2/3\)

Green = Free energy minima, red = low, blue = high
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Monte Carlo simulations

- Parallel tempering algorithm employed to dramatically improve thermal equilibration

$T_c$ rapidly diminishes in Neel phase
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Spiral surface develops
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\[ T_c \] rapidly diminishes in Neel phase

Spiral surface develops

Reentrant Neel
Monte Carlo simulations

- **Parallel tempering** algorithm employed to dramatically improve thermal equilibration

![Graph showing ordering temperature vs. coupling ratio with annotations on Neel and Spiral surface phases](image)

- $T_c$ rapidly diminishes in Neel phase
- "Order-by-disorder", with sharply reduced $T_c$
- Spiral surface develops
- Reentrant Neel
Spin liquid physics

- Order-by-disorder occurs at low temperatures
- Broad spin liquid regime emerges due to low $T_c$
- Can probe this physics experimentally via neutron scattering

Spin structure factor directly images “spiral surface” & entropic free energy corrections!
$J_2/J_1 = 0.85$

MnSc$_2$S$_4$

- Free energy corrections visible for $T_c < T < 1.3 \, T_c$
- “Spiral surface” more robust: persists for $T_c < T < 3 \, T_c$

Order by disorder

Spiral spin liquid

Physics dominated by spiral ground states
Spin liquid correlations analytically

- “Spherical model”
  - Describes spin liquids in kagome, pyrochlore antiferromagnets
  - Predicts structure factor data collapse

\[
S_j^2 = 1 \rightarrow \sum_j S_j^2 = N
\]

\[
J_2/J_1 = 0.85
\]

\[
\text{MnSc}_2\text{S}_4
\]

Structure factor for one FCC sublattice

\[
\Lambda(q) = 2 \left[ \cos^2 \frac{q_x}{4} \cos^2 \frac{q_y}{4} \cos^2 \frac{q_z}{4} + \sin^2 \frac{q_x}{4} \sin^2 \frac{q_y}{4} \sin^2 \frac{q_z}{4} \right]^{1/2}
\]
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\]

\[J_2/J_1 = 0.85\]

\[\text{MnSc}_2\text{S}_4\]

Structure factor for one FCC sublattice

Peaked near surface

Quantitative agreement! (except very near \(T_c\))

\[\Lambda(q) = 2 \left[ \cos^2 \frac{q_x}{4} \cos^2 \frac{q_y}{4} \cos^2 \frac{q_z}{4} + \sin^2 \frac{q_x}{4} \sin^2 \frac{q_y}{4} \sin^2 \frac{q_z}{4} \right]^{1/2}\]
What can we expect for experiments?

Realistic Hamiltonian:

\[ H = J_1 \sum_{\langle ij \rangle} S_i \cdot S_j + J_2 \sum_{\langle\langle ij \rangle\rangle} S_i \cdot S_j + \delta H \]

- **Entropic** free energy corrections vanish as \( T \to 0 \)

- **Energetic** corrections from \( \delta H \) inevitably dominate at lowest \( T \)

- If \( \delta H \) small enough, expect order-by-disorder phase to appear at higher \( T \)
Comparison with experiment: MnSc$_2$S$_4$

$q = 2\pi (3/4,3/4,0)$

Spiral order

Spiral order + $Q_{\text{diff}} \approx 2\pi$ diffuse scattering

"Spin liquid" with $Q_{\text{diff}} \approx 2\pi$ diffuse scattering

High-$T$ paramagnet

$0 \quad 1.9K \quad 2.3K$  

Theoretical implications

- $J_1$ is ferromagnetic here
- $J_2 / |J_1| \approx 0.85$
- Lowest-$T$ order determined energetically, not entropically

Entropy favors ~100 order

Experiment sees 110 order (favored by AFM $J_3$)

A. Krimmel et al. PRB 73, 014413 (2006); M. Mucksch et al. (unpublished)
Comparison with experiment: MnSc$_2$S$_4$ (cont’d)

$q = 2\pi (3/4,3/4,0)$  
Spiral order

Spiral order + 
$Q_{\text{diff}} \approx 2\pi$ diffuse scattering

Spin liquid with $Q_{\text{diff}} \approx 2\pi$ diffuse scattering

0 1.9K 2.3K

High-$T$ paramagnet

$\Theta_{CW}$

• Intensity shifts from $|q|$ to “spiral surface” as $T$ washes out $J_3$
• Consistent with “spiral spin liquid”

A. Krimmel et al. PRB 73, 014413 (2006); M. Mucksch et al. (unpublished)
Comparison with experiment: CoAl$_2$O$_4$

- Much less known here
  - Strong frustration, sample dependent
  - No sharp transition observed yet

- Powder neutron data + frustration suggest $J_2 / J_1 \approx 1/8$ for this material

Summary

• Many spinels constitute *frustrated diamond lattice antiferromagnets*
  – MnSc$_2$S$_4$, CoAl$_2$O$_4$, etc.

• Simple $J_1$-$J_2$ model captures essential physics
  – Continuous spiral ground state degeneracy
  – Important ordering mechanism is order-by-disorder
  – Spin correlations in “spiral spin liquid” reveals surface + entropic effects

• Theoretical predictions consistent with existing experiments
Future Directions

- Single crystals wanted
  - Allow for more direct comparison
  - Concrete experimental realization of order-by-disorder??
- Explore spin dynamics for inelastic neutron scattering?
- Effects of disorder?
- Details of low-$T$ order in MnSc$_2$S$_4$? Commensurate lock-in?
- Physics of spin + orbitally frustrated FeSc$_2$S$_4$? Exotic quantum ground state?

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