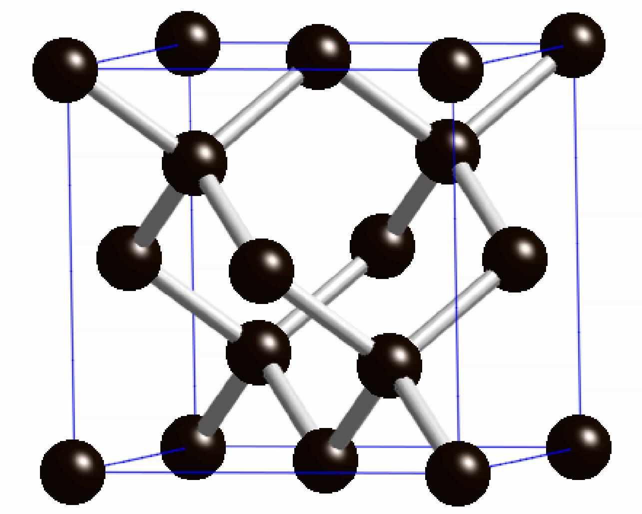
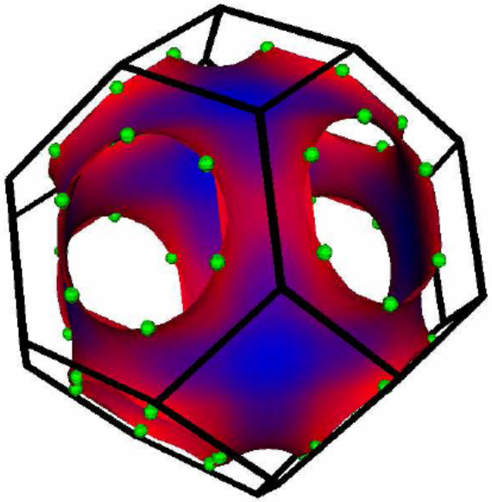
Frustrated diamond lattice antiferromagnets





Jason Alicea (Caltech)

Doron Bergman (Yale)

Leon Balents (UCSB)

Emanuel Gull (ETH Zurich)

Simon Trebst (Station Q)

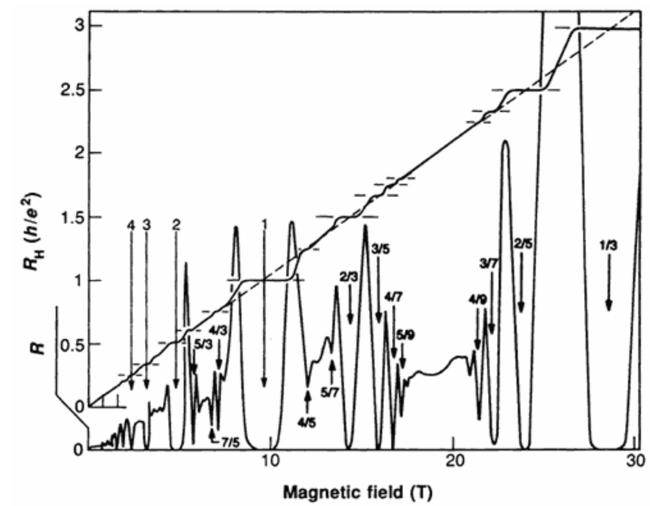
Bergman *et al*., Nature Physics 3, 487 (2007).

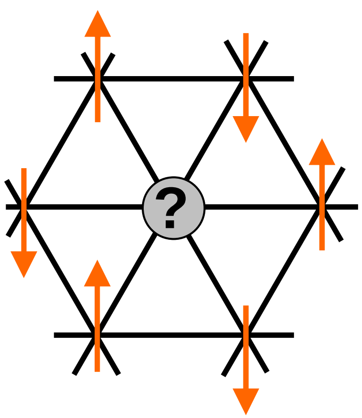
Outline

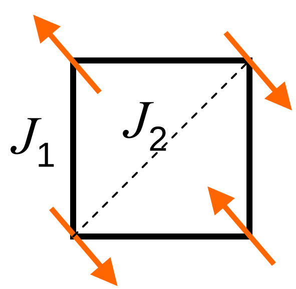
- Introduction
  - Frustration, degeneracy, & emergent phenomena
  - Diamond lattice antiferromagnets
  - Overview of experiments
- Theory of frustrated diamond lattice antiferromagnets
  - Ground states (highly degenerate spirals)
  - Stability & order-by-disorder
  - Monte Carlo simulations
  - Spiral 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)**

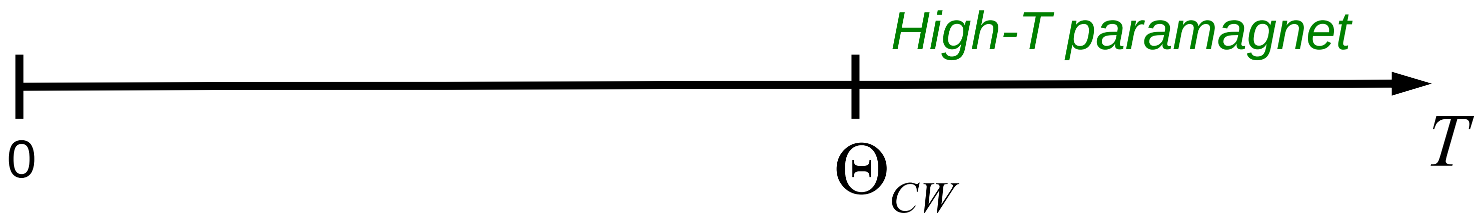






$\Rightarrow$ Highly degenerate ground states (pyrochlore, kagome, FCC, etc.)
- High sensitivity to perturbations
- Spin-glass behavior
- Spin-liquid physics
- Order-by-disorder

Experimental signatures of frustration



At high temperatures,
Curie-Weiss law holds.

$$\chi \sim \frac{1}{T - \Theta_{CW}}; \qquad \Theta_{CW} \sim -(\text{Exchange } J\text{'s})$$

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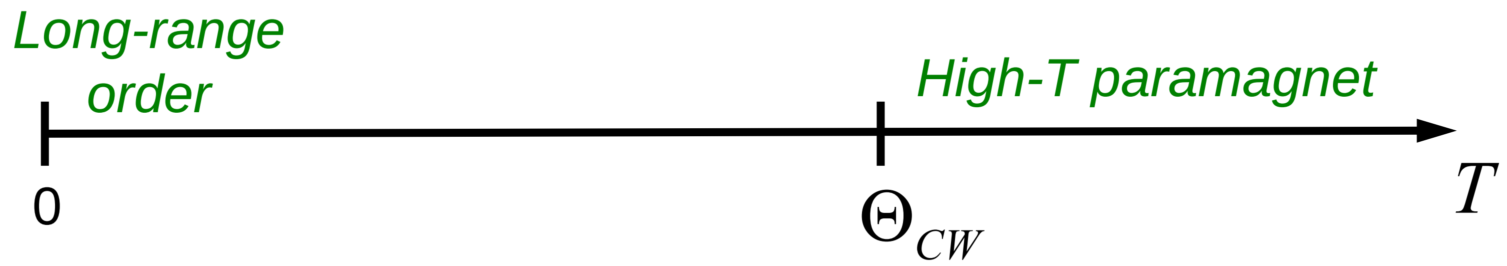

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At low temperatures, systems typically order.

Useful diagnostic: "frustration parameter" $f = |\Theta_{CW}|/T_c$

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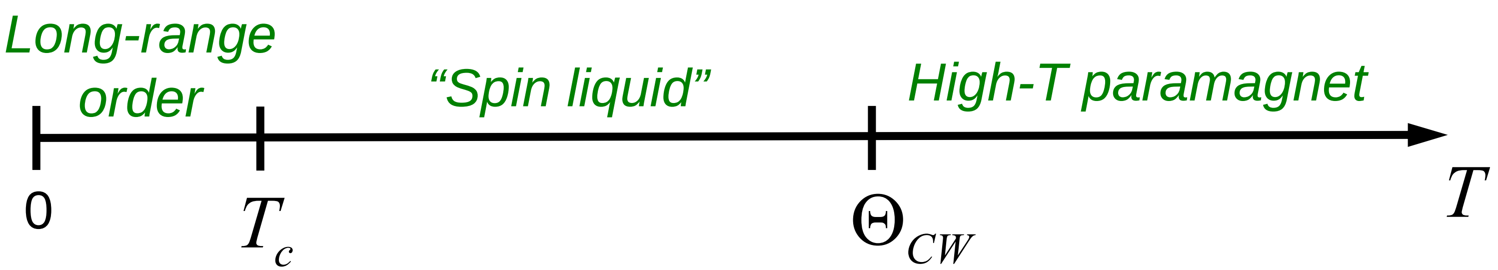

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At low temperatures, systems typically order.

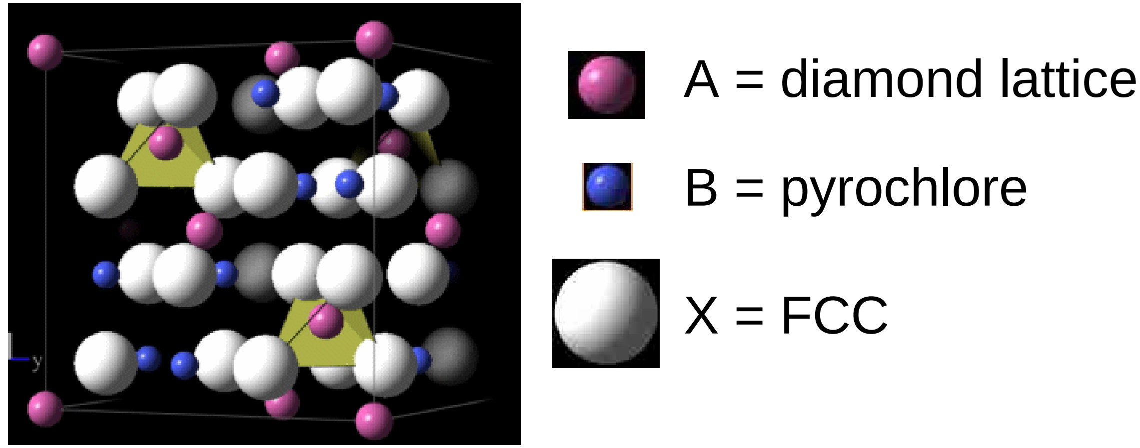
Useful diagnostic: "frustration parameter" $f = |\Theta_{CW}| / T_c$

Highly frustrated systems $\Rightarrow$ $f > 5\text{-}10$

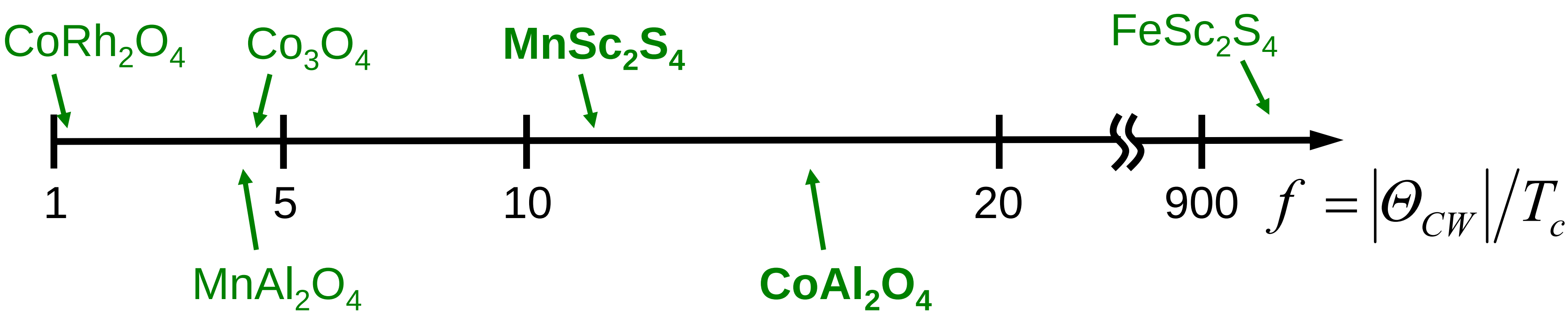
- Key challenges: Low-T ordering mechanisms & Characterizing "spin liquid" correlations

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)

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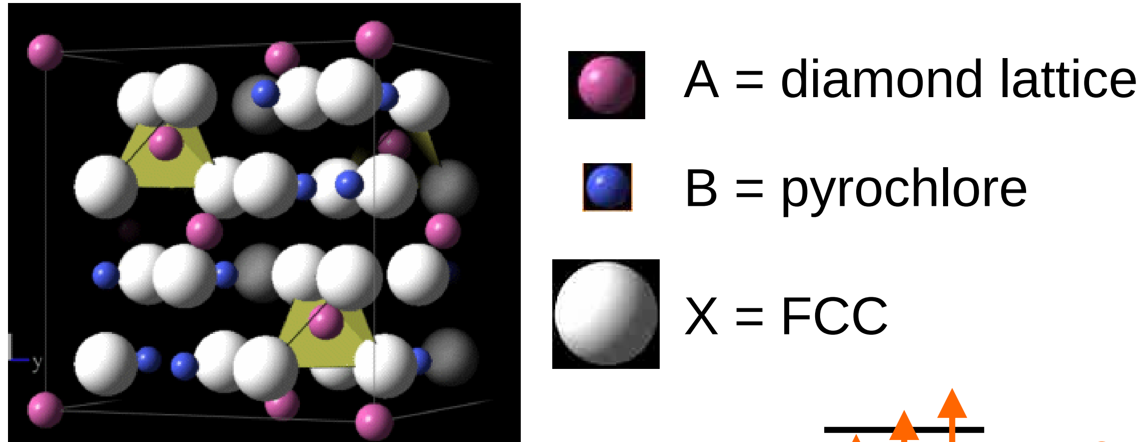

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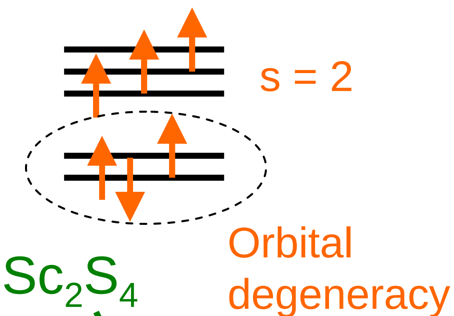

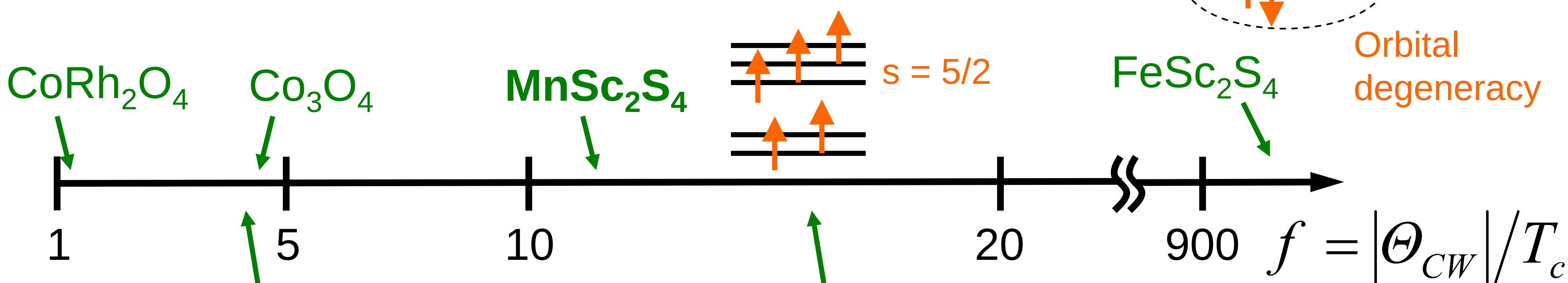

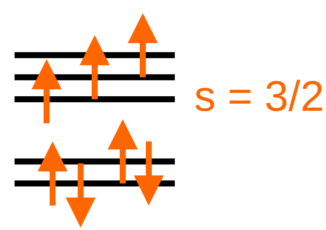

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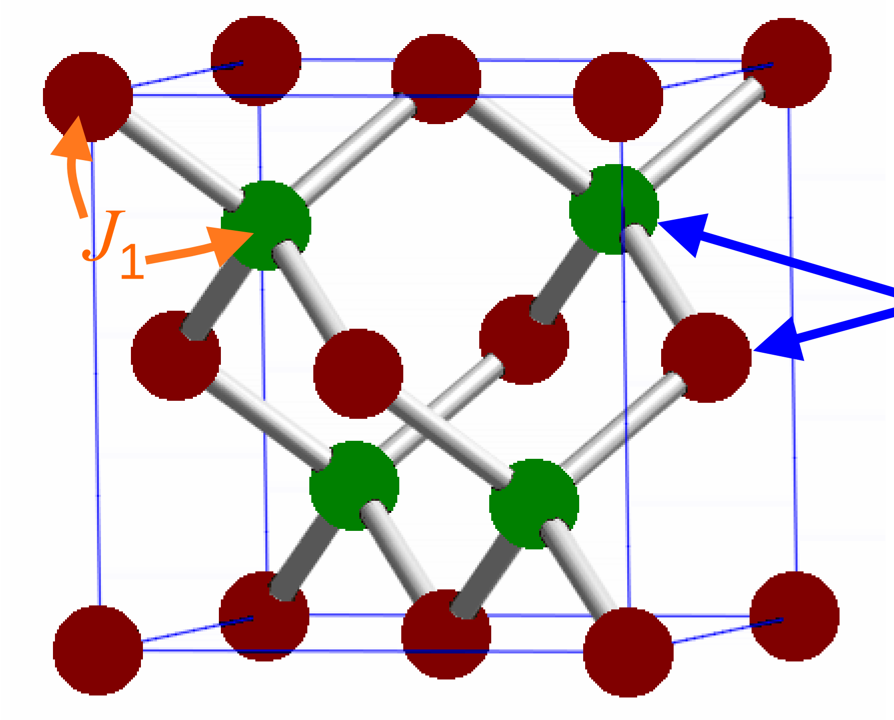
Frustration on the bipartite diamond lattice??

Naïve Hamiltonian:

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

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



Diamond lattice = 2 FCC sublattices coupled via J_1

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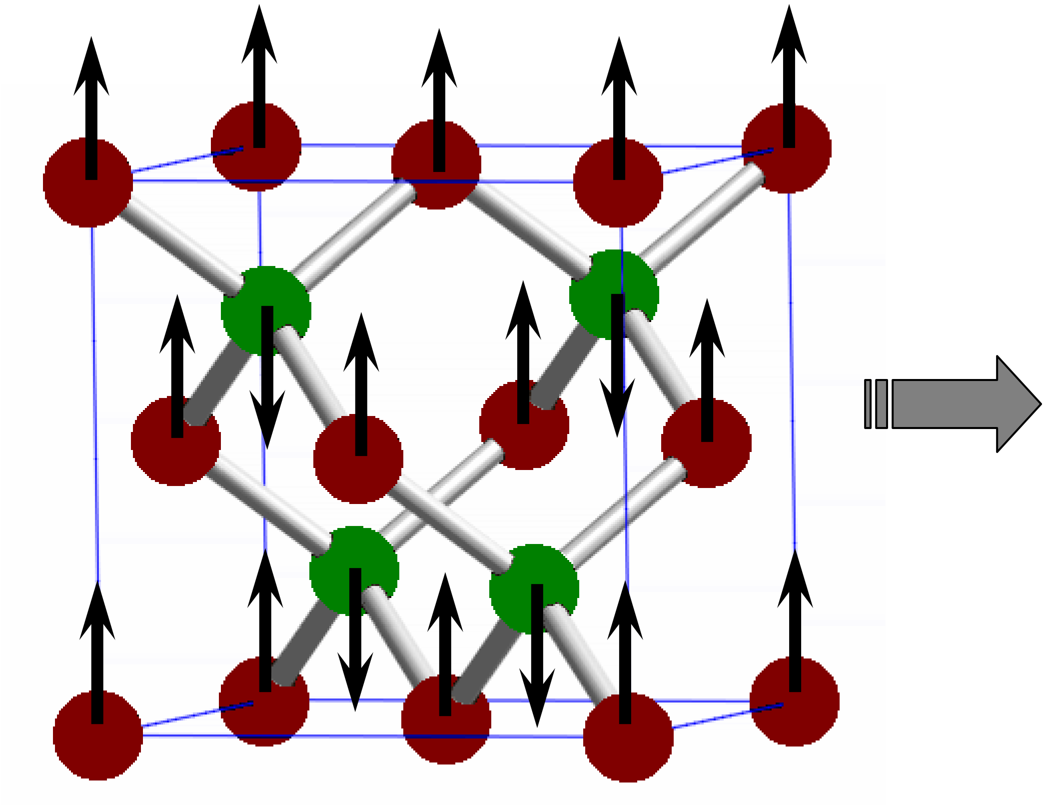


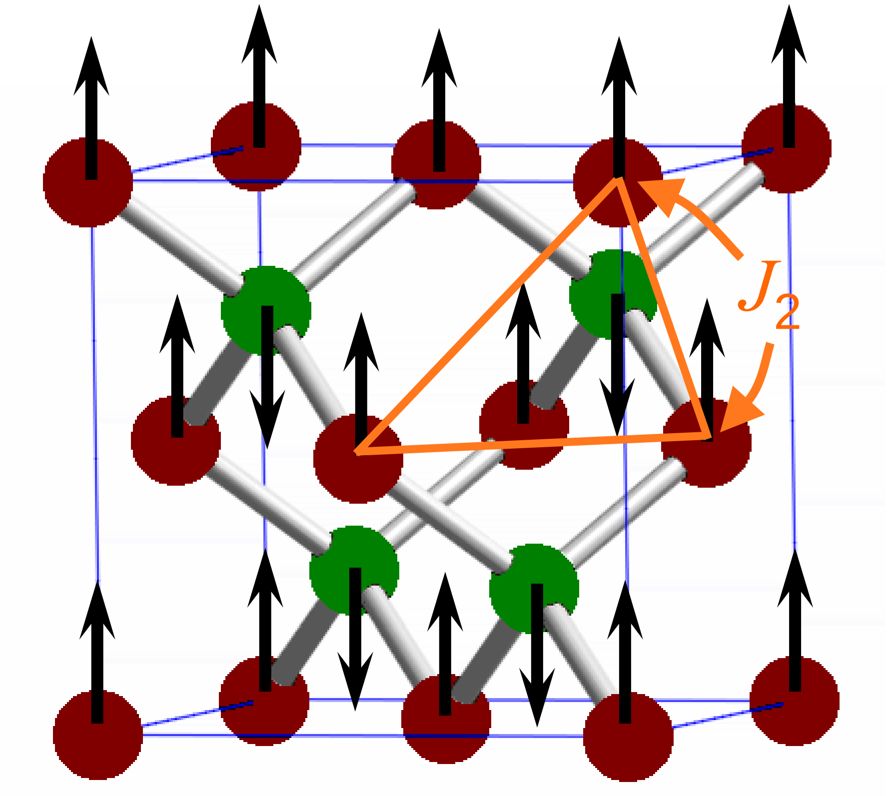
No Frustration

Frustration on the **diamond lattice**

Remedy:
2nd neighbor exchange

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

Generates strong frustration

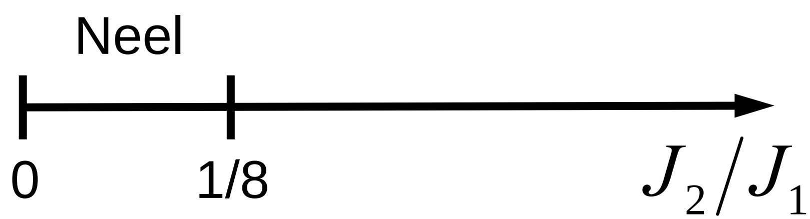


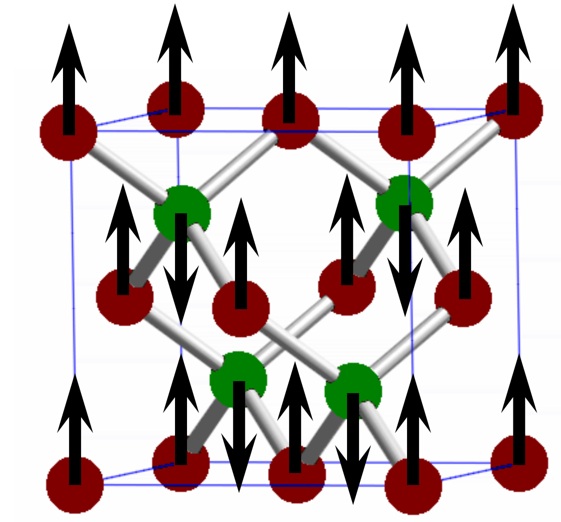
J_1 and J_2 expected to be comparable due to similarity in exchange paths

W. L. Roth, J. Phys. (Paris) **25**, 507 (1964)

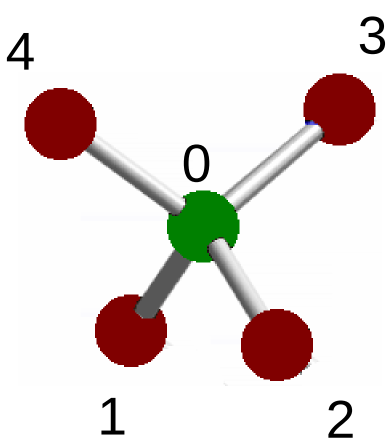
$T = 0$ physics: ground states

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





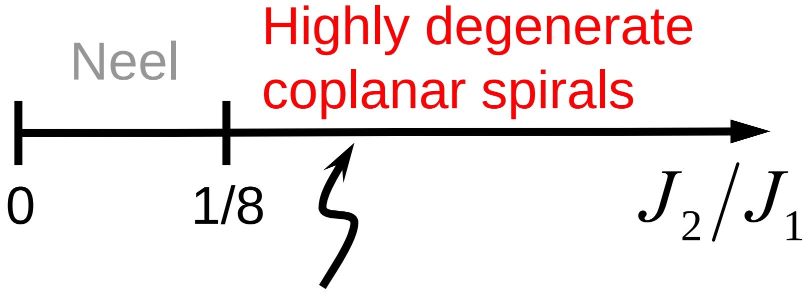
Useful rewrite of Hamiltonian:

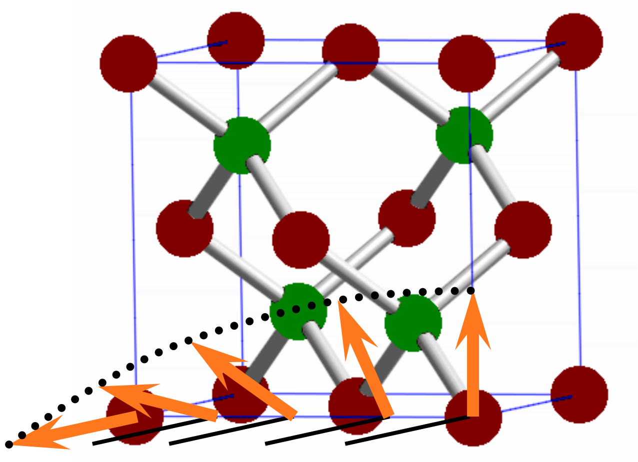


$$H = J_1 \sum_t [\mathbf{S}_0 + (\mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3 + \mathbf{S}_4)/4]^2$$
$$+ (J_2 - J_1/8) \sum_{\langle\langle ij \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

$T = 0$ physics: ground states

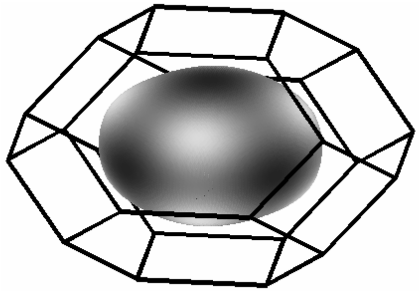
$$H = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle ij \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$





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

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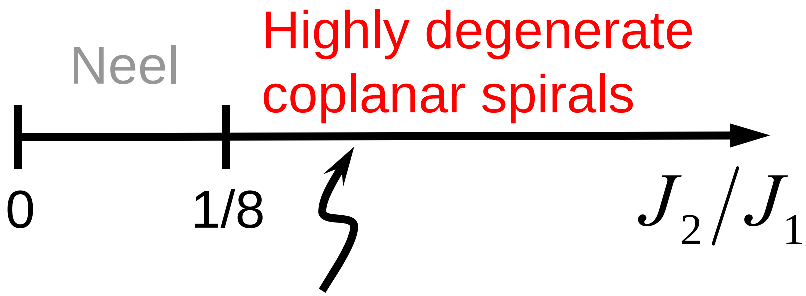

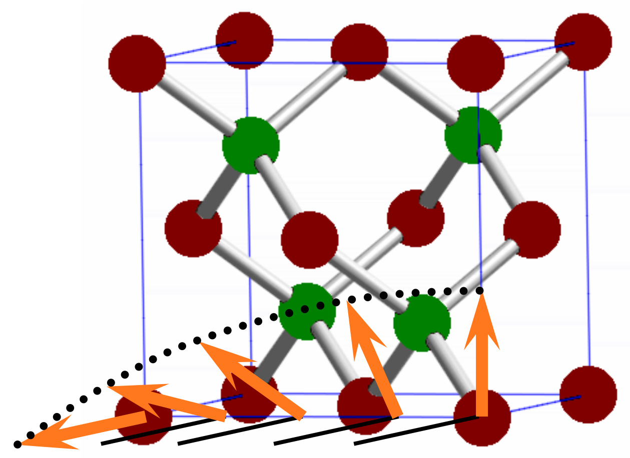

Direction & pitch of spirals characterized by a wavevector residing on a surface in momentum space!

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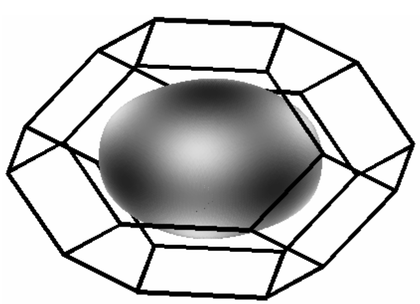

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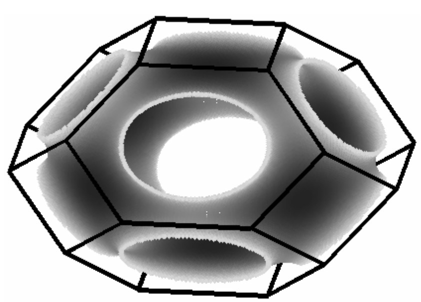
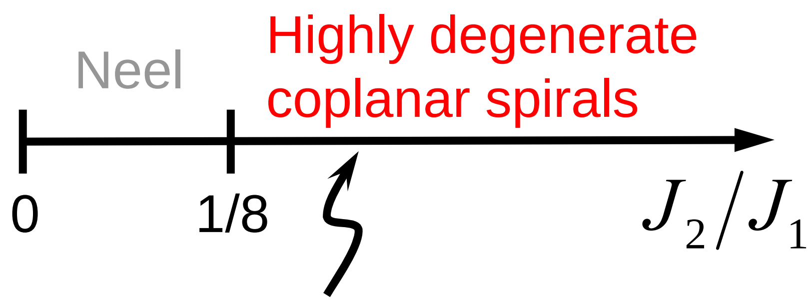
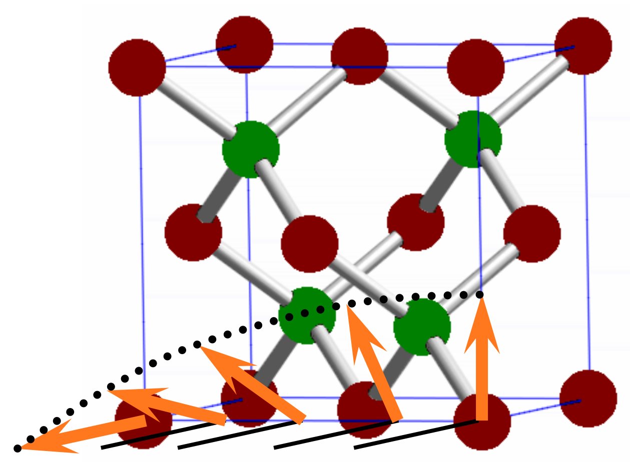


$J_2/J_1 = 0.4$

$T = 0$ 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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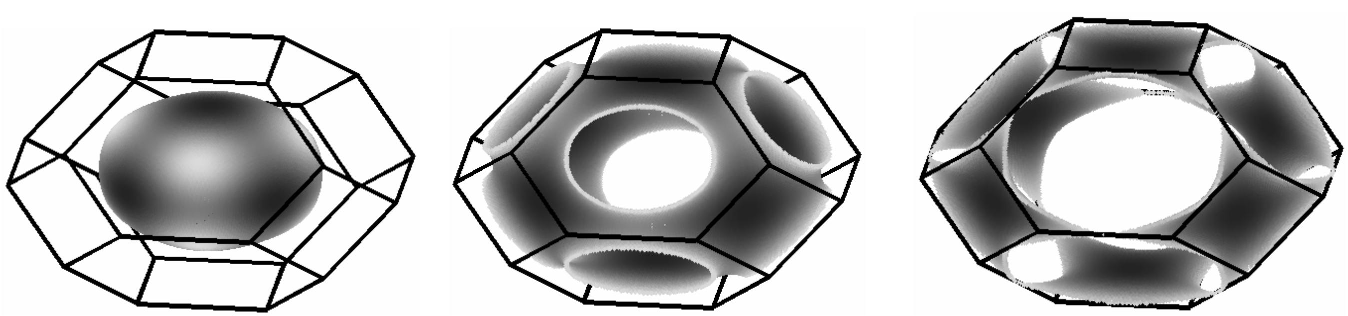

$J_2/J_1 = 0.2$ $J_2/J_1 = 0.4$ $J_2/J_1 = 0.85$

$T = 0$ physics: ground states

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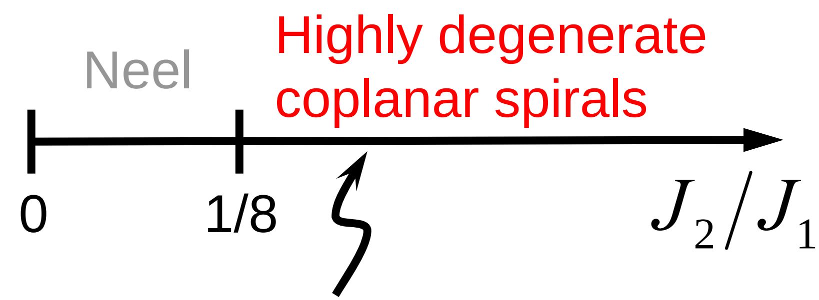

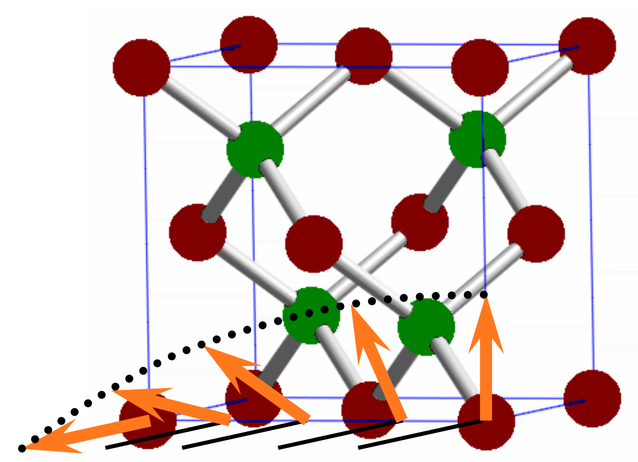

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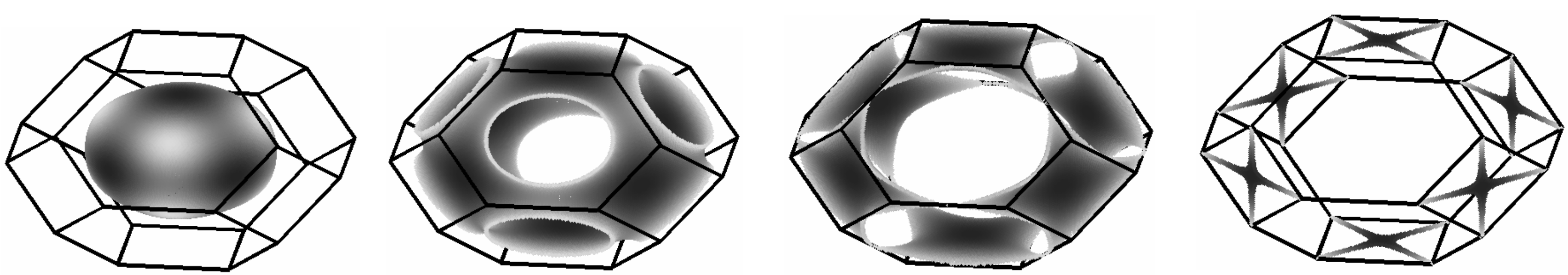


$J_2/J_1 = 0.2$ $J_2/J_1 = 0.4$ $J_2/J_1 = 0.85$ $J_2/J_1 = 20$

Low-T physics: ***Can long-range order occur?***

Stability nontrivial due to massive spiral degeneracy.

- Expand Hamiltonian in **fluctuations**:

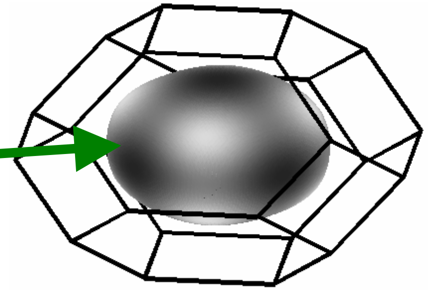
$$\delta \mathbf{S}_i = \mathbf{S}_i - \langle \mathbf{S}_i \rangle$$

Arbitrary ground state order

- At ***T* = 0**, branch of normal modes has **infinite # of zeros**!

$$\omega_0(\mathbf{q}) = 0$$

For all $\mathbf{q}$ on surface



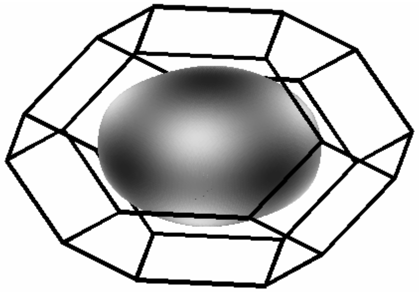
Naively, fluctuations ***diverge***

$$\langle \delta \mathbf{S}_i^2 \rangle \sim T \int \frac{d^3\mathbf{q}}{\omega_0^2(\mathbf{q})} \to \infty$$

"Order-by-disorder" **stabilization**

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

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



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

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

(Fluctuations small at low T)

- **Non-analytic *T*-dependence** ⇒ unconventional thermodynamic behavior, e.g.,

$$C_v = A + BT^{1/3}$$

Aside on self-consistent approach

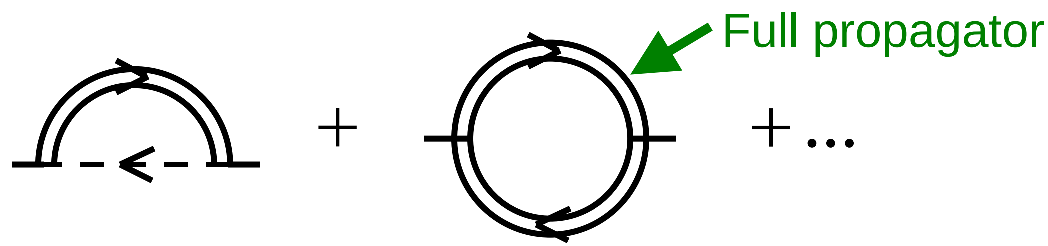
- Expand Hamiltonian in fluctuations:

"Interaction" terms

$$\delta\mathbf{S}_i = \mathbf{S}_i - \langle \mathbf{S}_i \rangle; \qquad H = H_2 + \underbrace{H_3 + H_4 + \ldots}_{}$$

- Get self-energy self-consistently for divergent mode

$$\bar{\Sigma}(\mathbf{q}) =$$



$$\Rightarrow \quad \bar{\Sigma}(\mathbf{q}) = T\int_{\mathbf{k}} \Gamma(\mathbf{q},\mathbf{k}) G(\mathbf{k}); \qquad G(\mathbf{k}) = \frac{1}{\omega_0^2(\mathbf{k}) + \bar{\Sigma}(\mathbf{k})}$$

- For $\mathbf{q}$ on surface, assume $\bar{\Sigma}(\mathbf{q}) \sim T^{\alpha}\Sigma(\mathbf{q})$

$$\Rightarrow \quad \boxed{\omega_T^2(\mathbf{q}) = \omega_0^2(\mathbf{q}) + T^{2/3}\Sigma(\mathbf{q})}$$

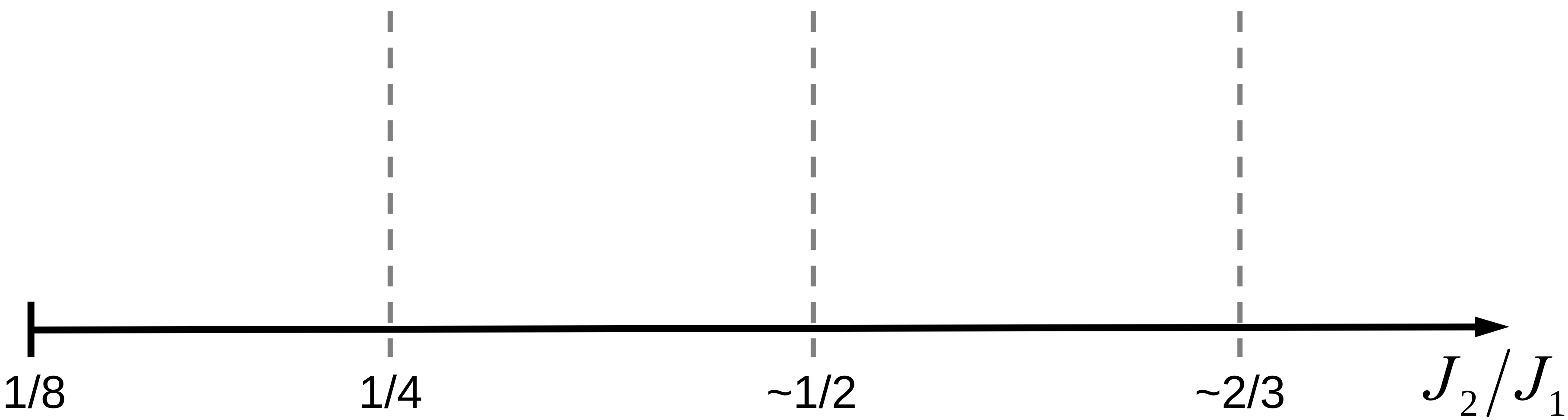
“Order-by-disorder” **selection**

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

- Need Free Energy for all $\mathbf{Q}$ on spiral surface

$$F(\mathbf{Q}) = E - TS(\mathbf{Q})$$

- Entropy favors states with highest density of nearby low-energy states
- Complex phase structure emerges:



Green = Free energy minima, red = low, blue = high

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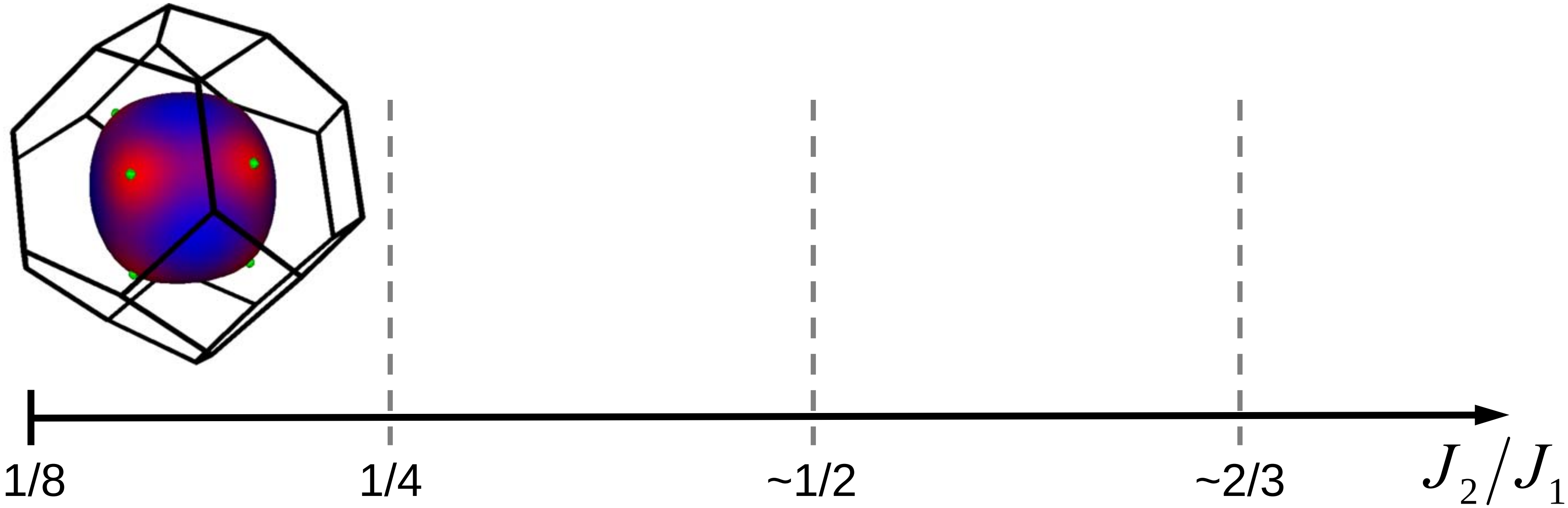

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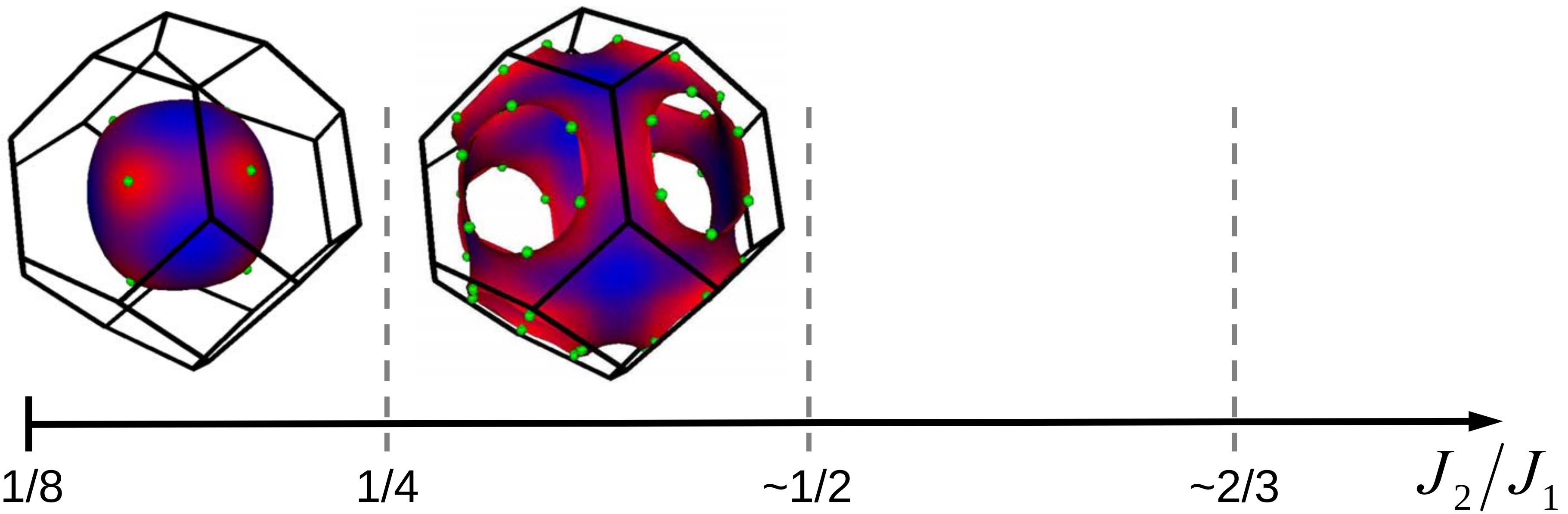

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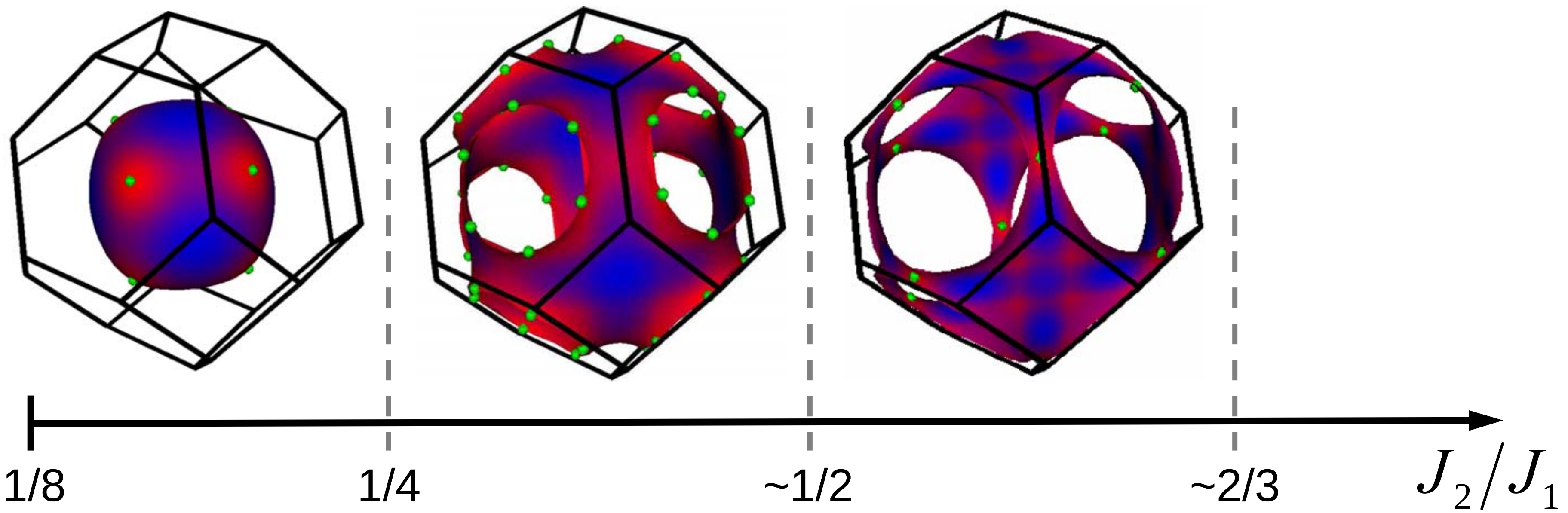

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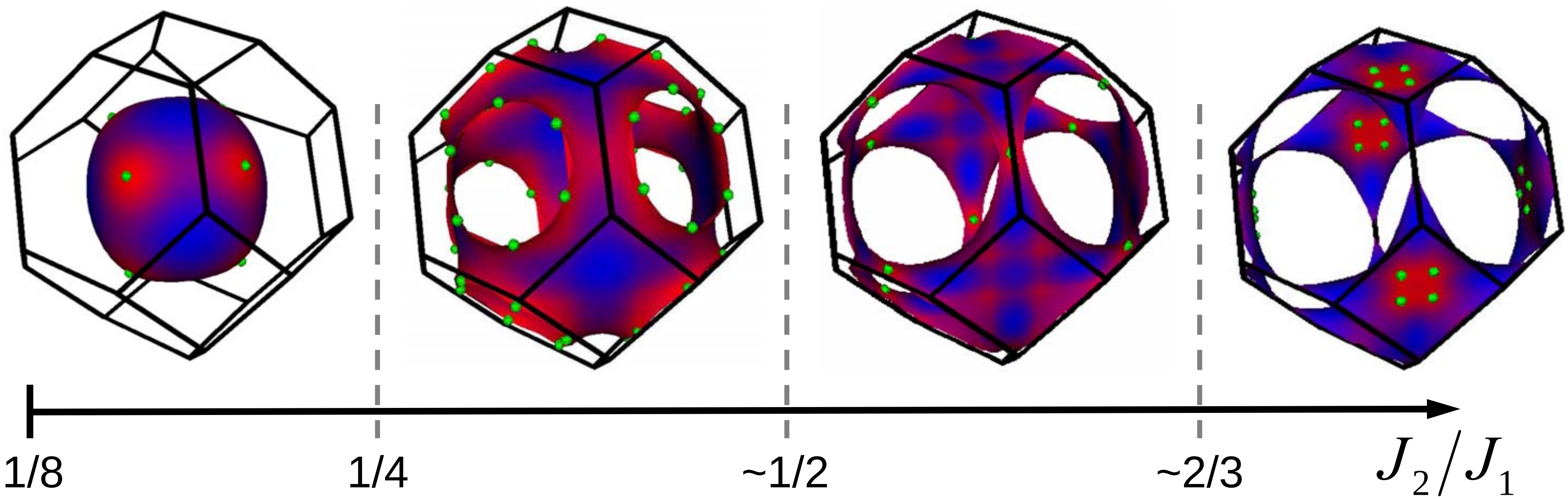

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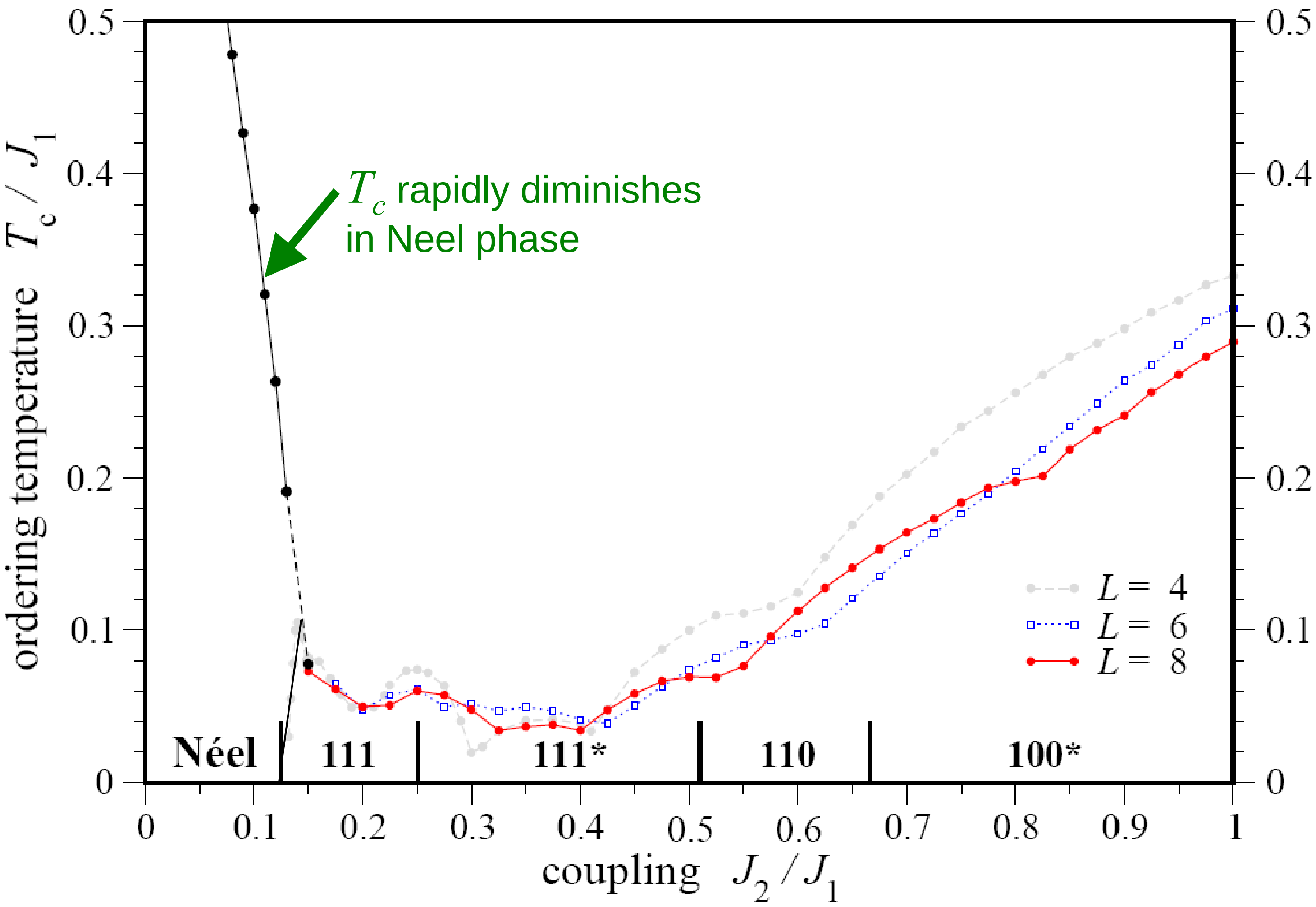
Monte Carlo simulations

- Parallel tempering algorithm employed to dramatically improve thermal equilibration



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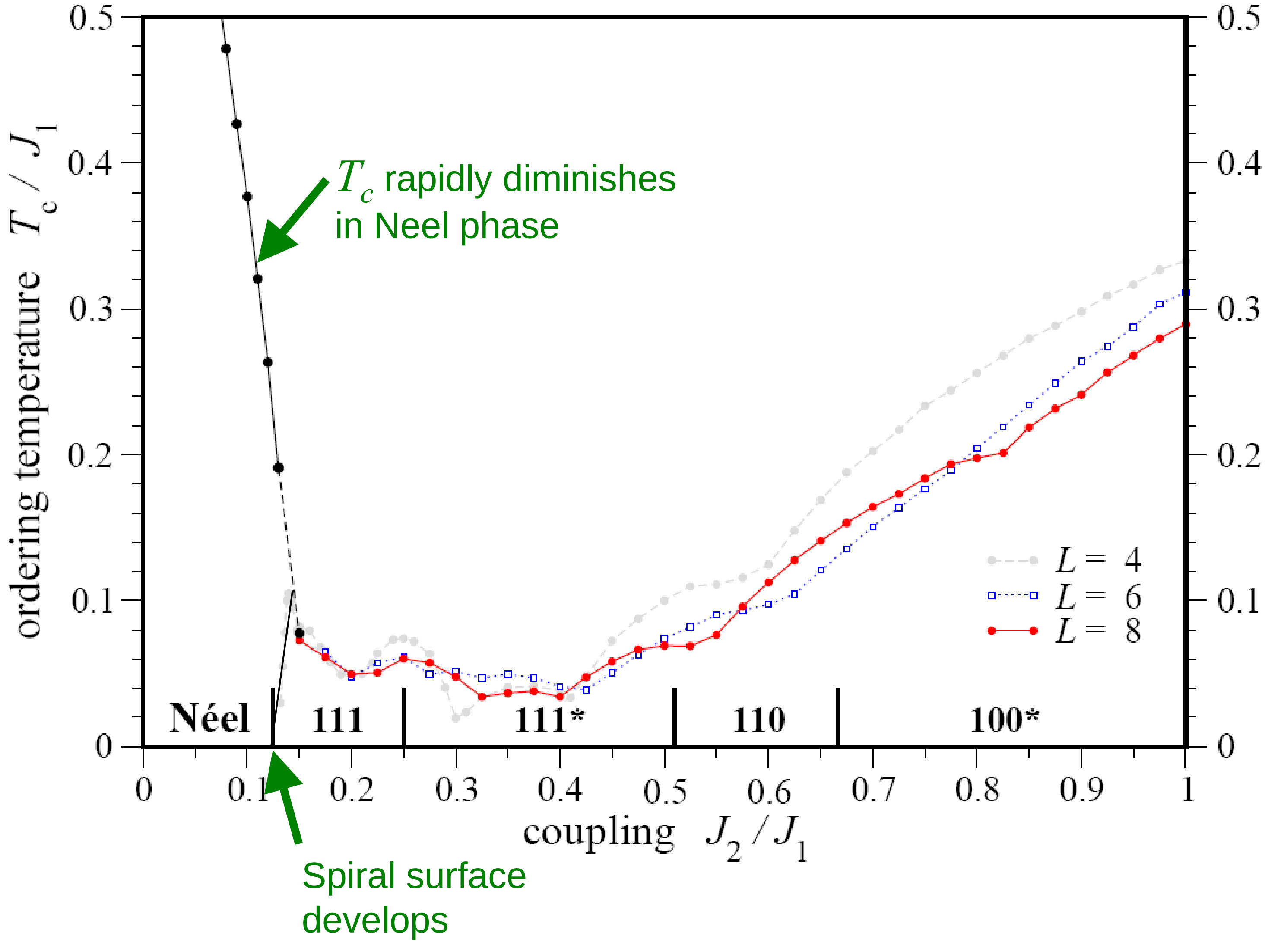

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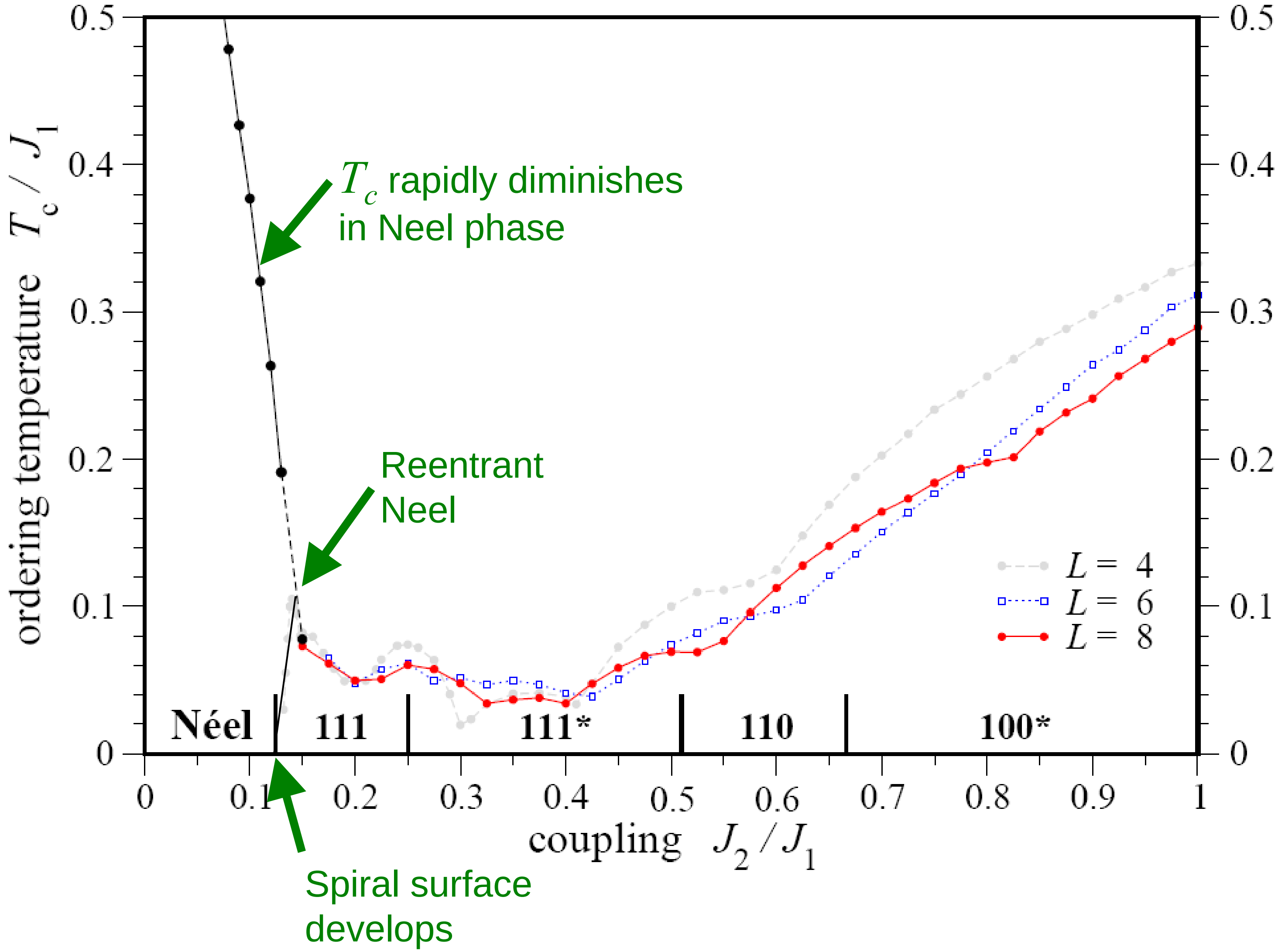

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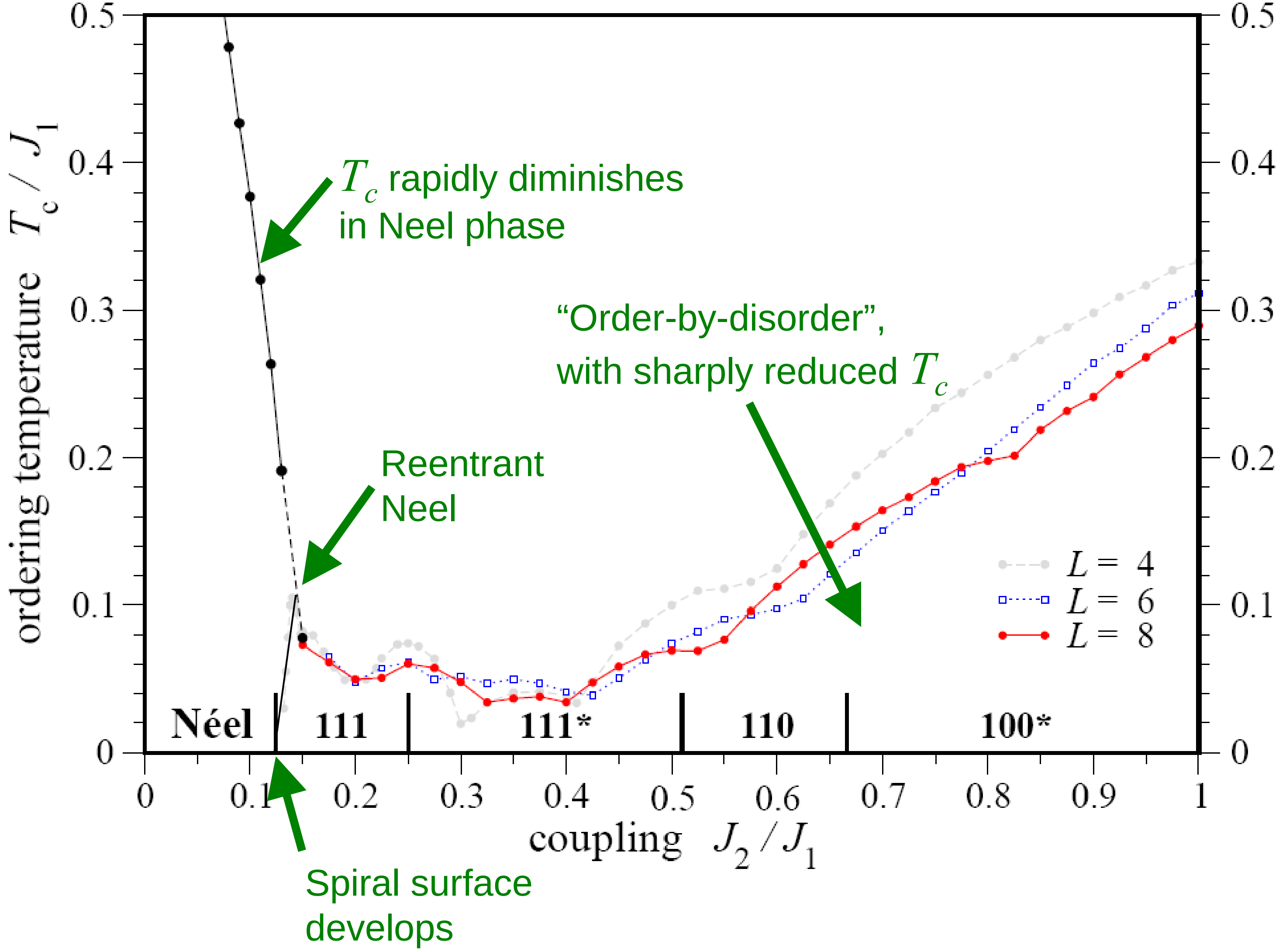
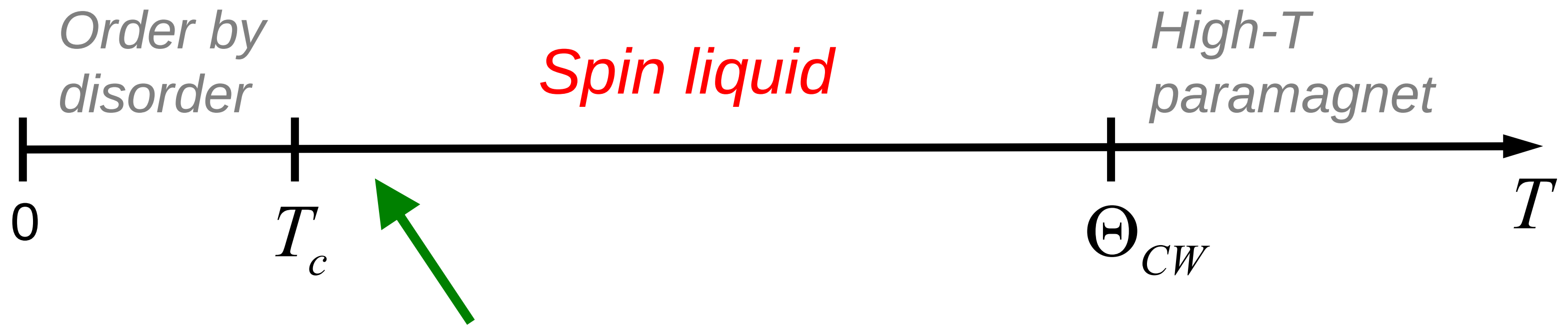


Spin liquid physics

- 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



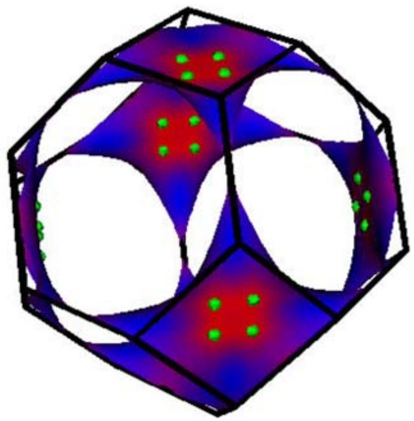
Spin structure factor directly images “spiral surface” & entropic free energy corrections!

Structure factor in spiral spin liquid regime

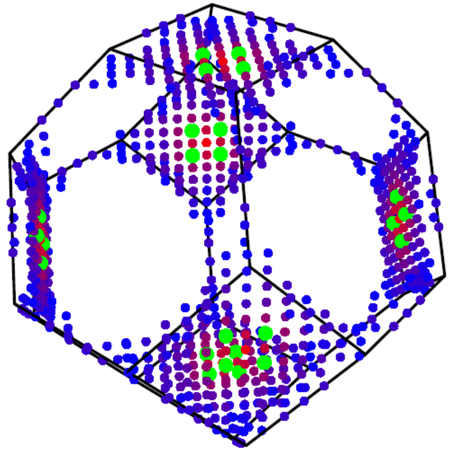
$J_2/J_1 = 0.85$

$MnSc_2S_4$

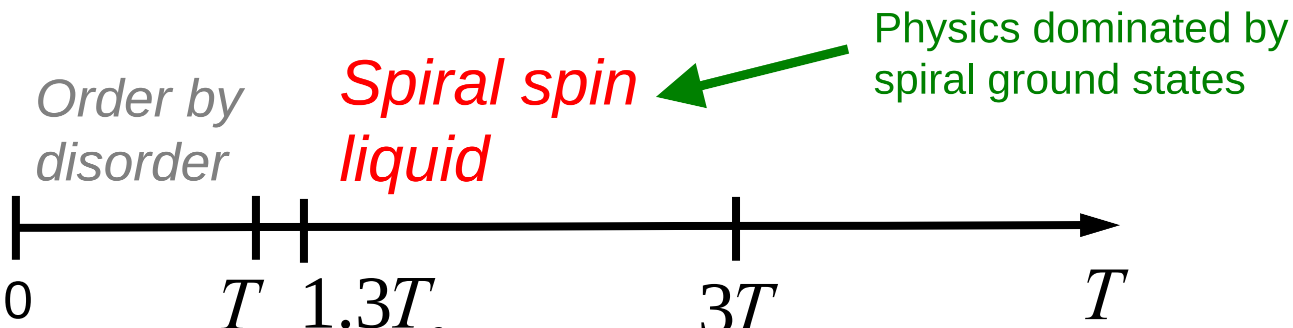
Analytic free energy



Numerical structure factor



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



Spin liquid correlations analytically

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

$$\mathbf{S}_j^2 = 1 \rightarrow \sum_j \mathbf{S}_j^2 = N$$

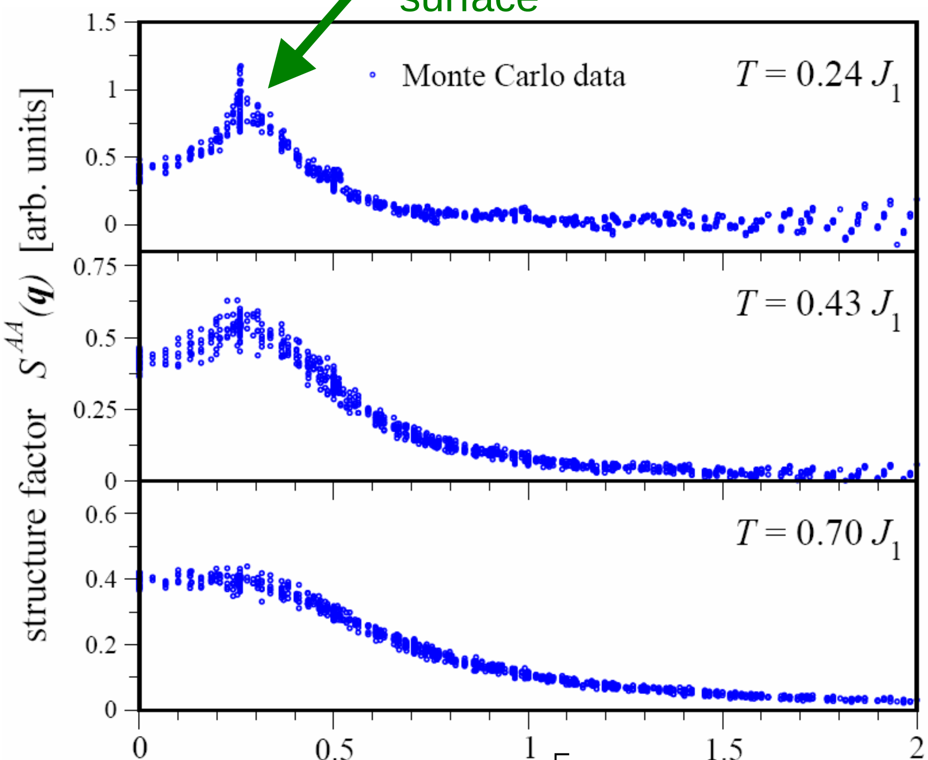
# of sites

$J_2/J_1 = 0.85$

$MnSc_2S_4$

Peaked near surface

Structure factor for one FCC sublattice



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

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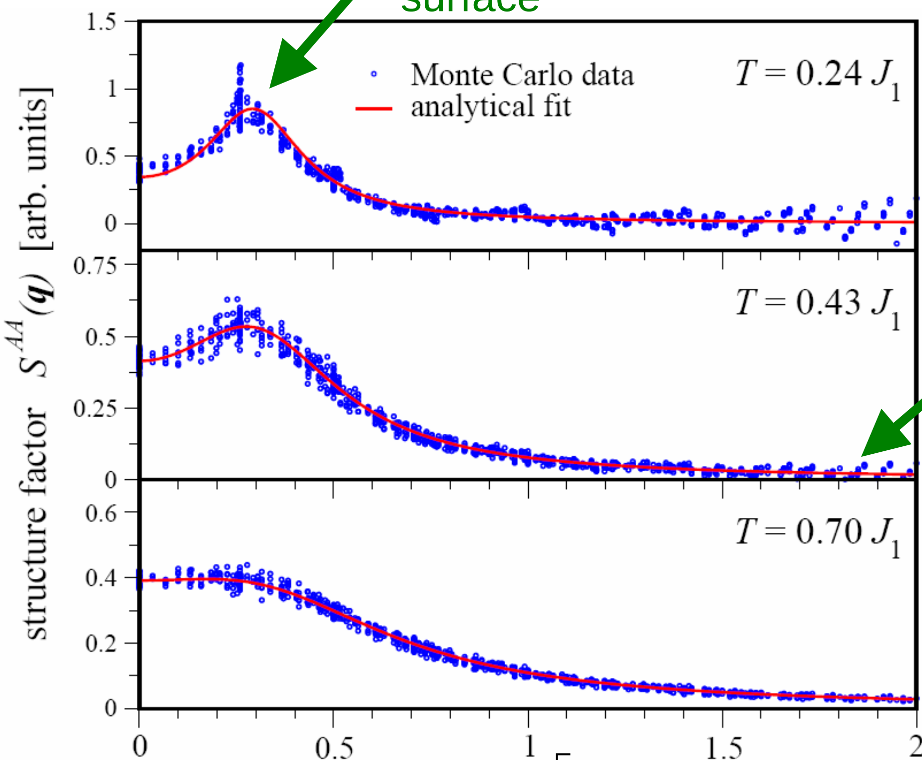

Quantitative agreement! (except very near T_c)

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Nontrivial experimental test, but need single crystals…

What can we expect for experiments?

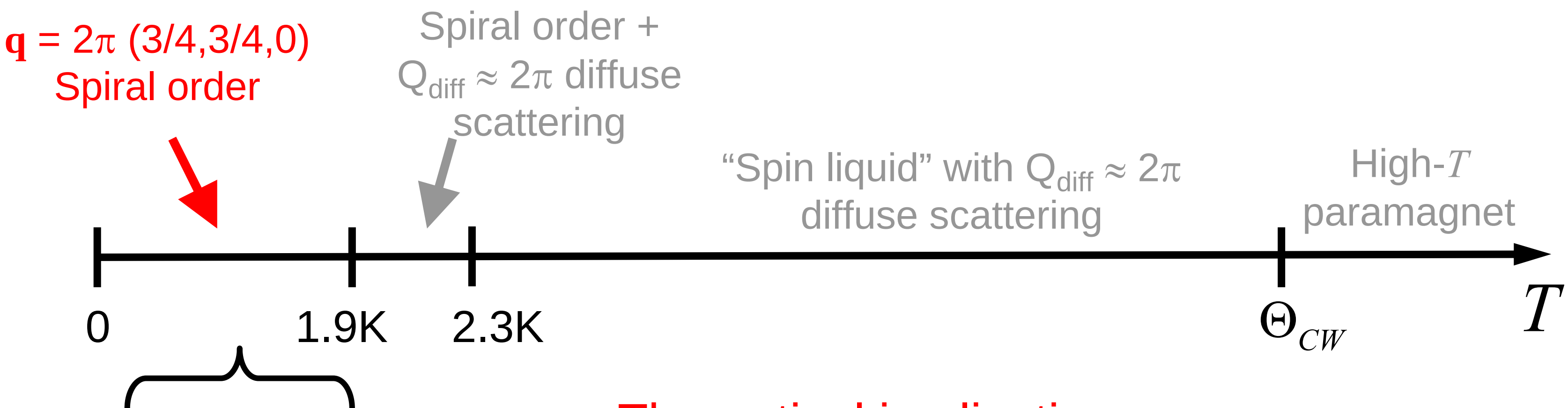
Realistic Hamiltonian:

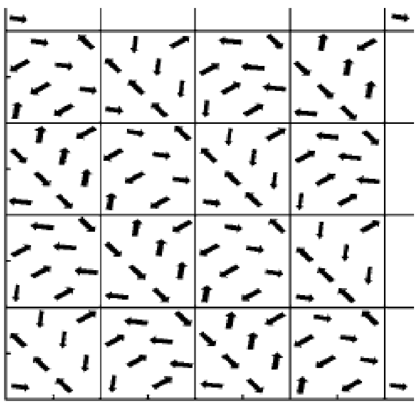
$$H = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle ij \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + \delta H$$

Degeneracy-breaking perturbations

- **Entropic** free energy corrections vanish as $T \to 0$
- **Energetic** corrections from δH inevitably dominate at lowest T
- If δH small enough, expect order-by-disorder phase to appear at higher T

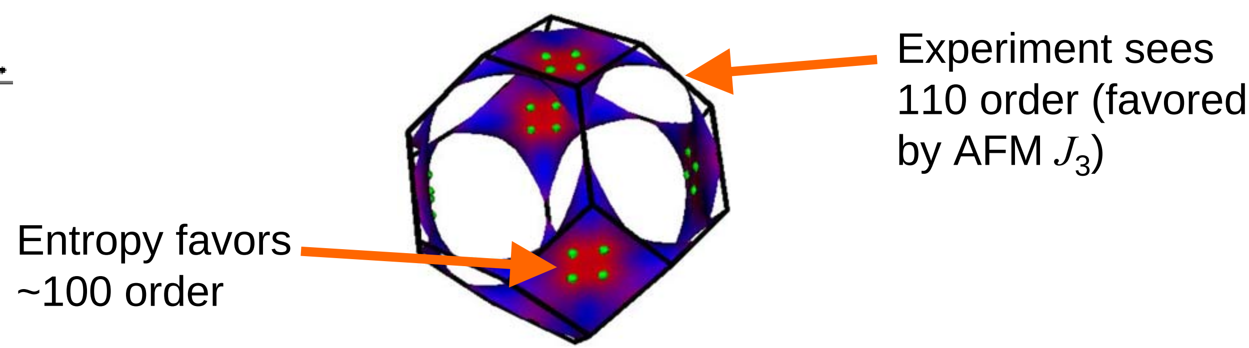
Comparison with experiment: $MnSc_2S_4$





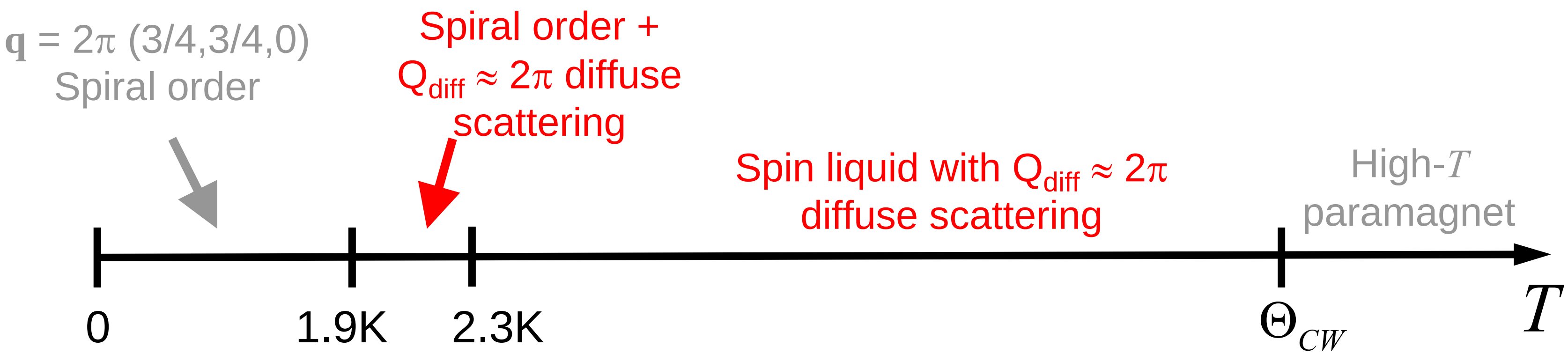
Theoretical implications

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

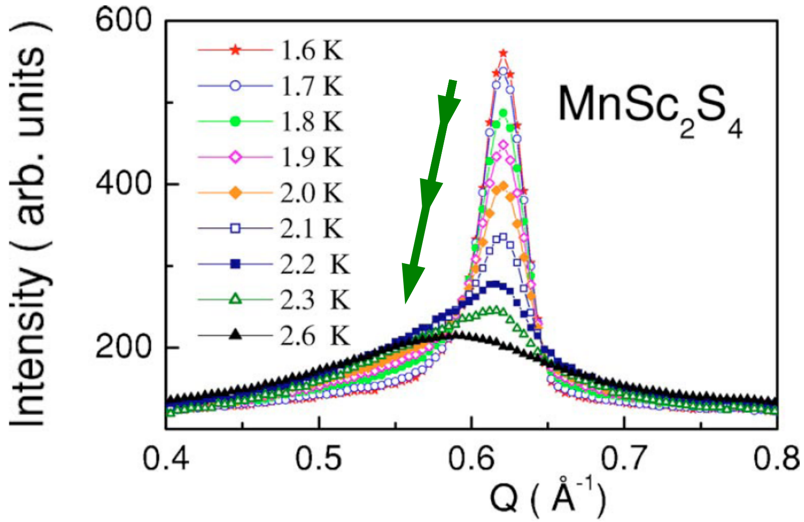


A. Krimmel et al. PRB **73**, 014413 (2006); M. Mucksch *et al*. (unpublished)

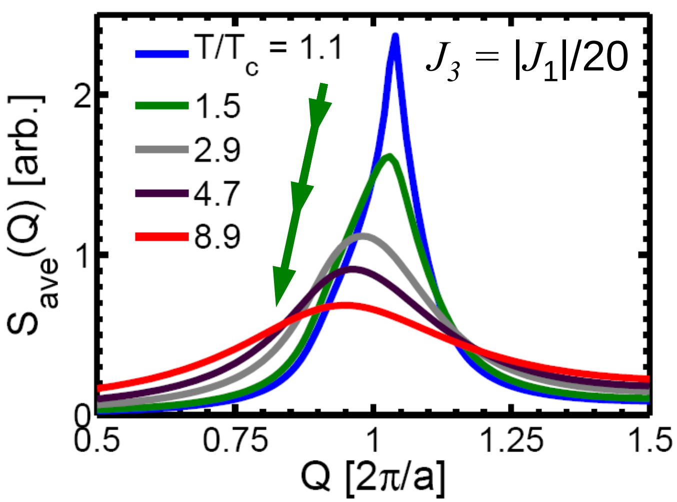
Comparison with experiment: $MnSc_2S_4$ (cont'd)



Experiment



Theory

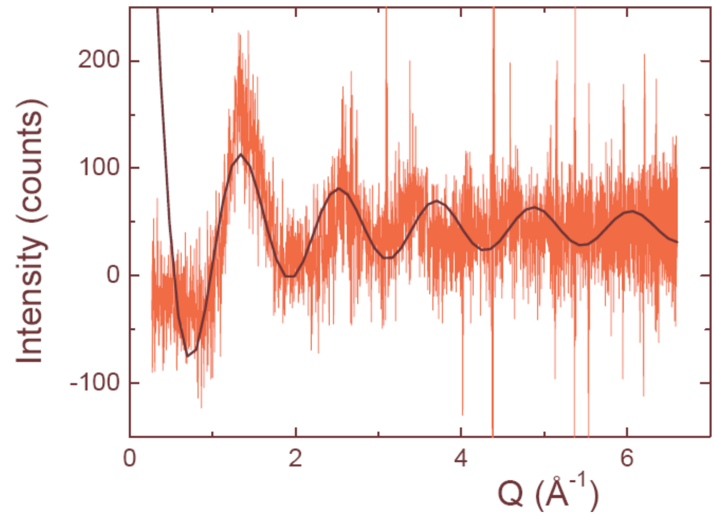


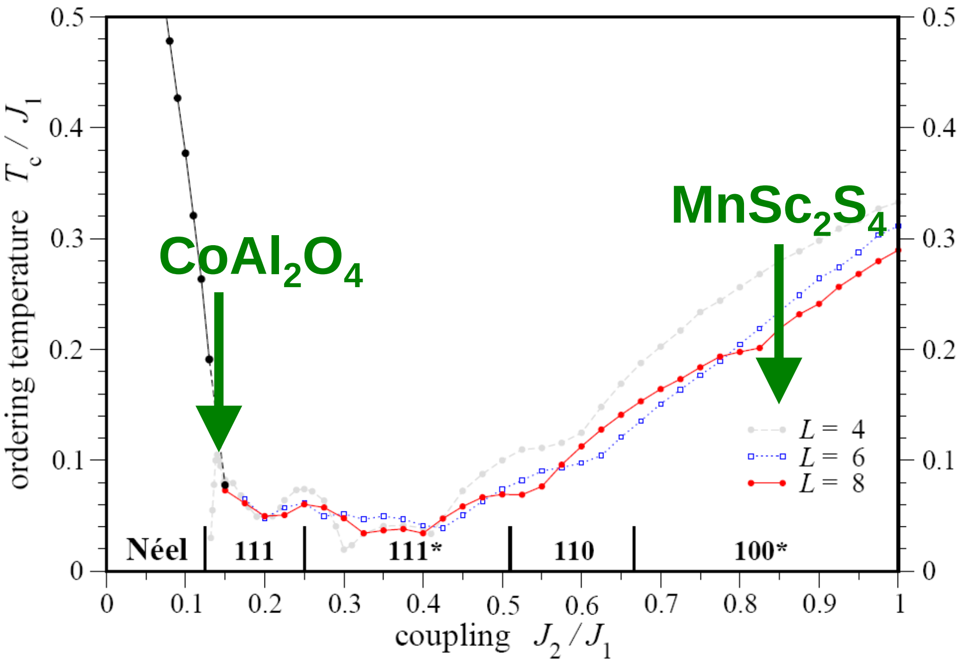
- Intensity shifts from |**q**| to "spiral surface" as T washes out J_3
- Consistent with "spiral spin liquid"

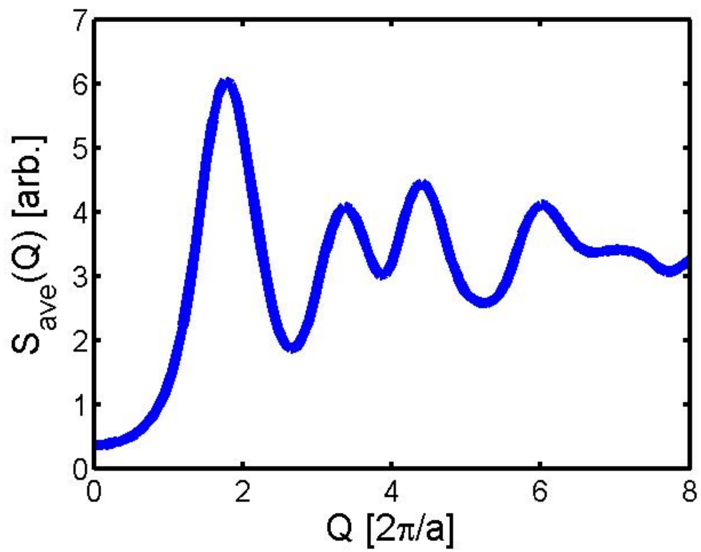
A. Krimmel et al. PRB **73**, 014413 (2006); M. Mucksch *et al*. (unpublished)

Comparison with experiment: $CoAl_2O_4$

- 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







N. Tristan et al. PRB **72,** 174404 (2005); A. Krimmel et al. Physica B **378-380**, 583 (2006); T. Suzuki *et al*. (unpublished)

Summary

- Many spinels constitute *frustrated diamond lattice antiferromagnets*
  - $MnSc_2S_4$, $CoAl_2O_4$, 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 $MnSc_2S_4$? Commensurate lock-in?
- Physics of spin + orbitally frustrated $FeSc_2S_4$? Exotic quantum ground state?

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