Order by disorder and spiral spin liquid in frustrated diamond lattice antiferromagnets

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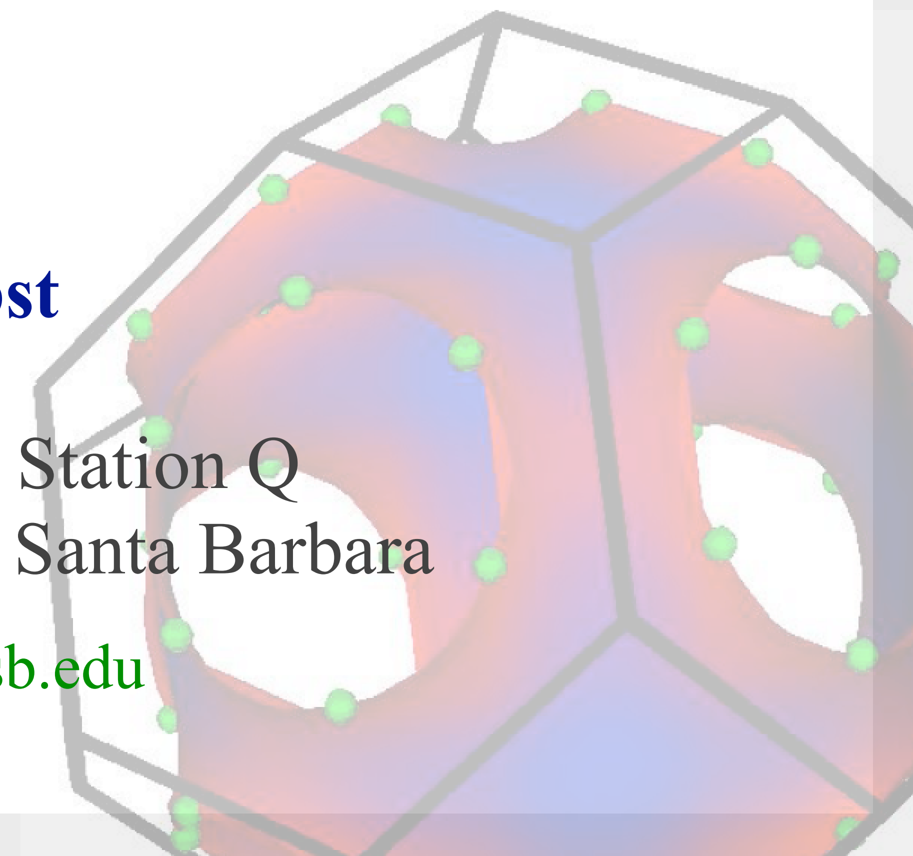
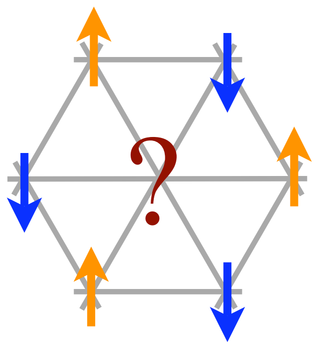


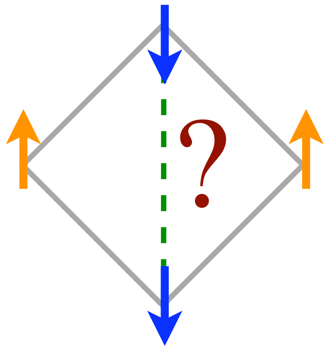
Outline

- **Motivation**
  - Frustrated magnetism, degeneracies & emergent phenomena
  - Experimental observations
- **Theory of frustrated diamond lattice AFMs**
  - Ground-states, stability and order-by-disorder
  - Numerical simulations
  - The spiral spin liquid regime
- **Comparison to experiments**

Frustrated magnetism & degeneracies

Frustration = not all interactions satisfiable simultaneously.





Microscopic interactions can produce
many nearly **degenerate low-energy states**.

Weak residual effects can give rise to
remarkable **emergent phenomena**.

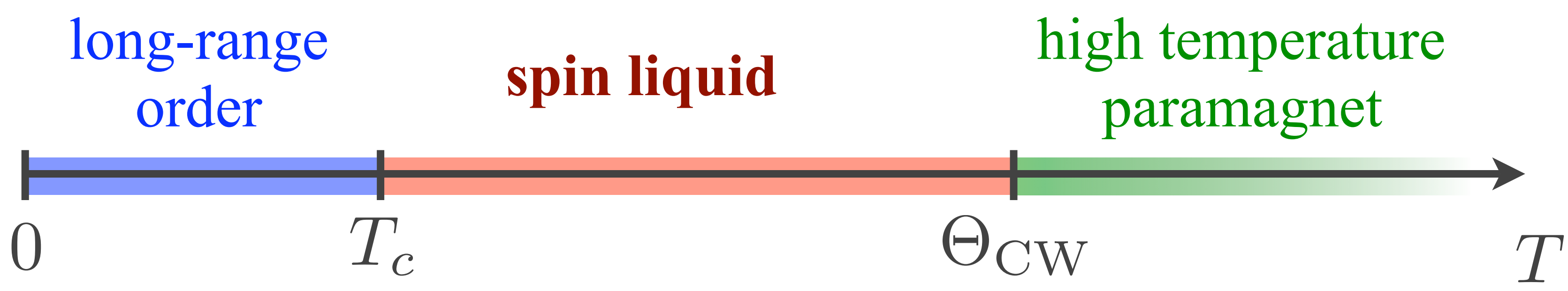
2D electron systems

partial Landau-level occupation
exotic quantum Hall liquids

cuprates

several competing orders
high-T_c superconductivity

Experimental signatures of frustration



frustration parameter

$$f = \frac{\Theta_{CW}}{T_c}$$

"highly frustrated"

$$f > 5 - 10$$

spin liquid

system **fluctuates** amongst low-energy configurations, but **no** long-range order

Curie-Weiss law

$$\chi \sim \frac{1}{T - \Theta_{CW}}$$

Key challenges

Low temperature ordering mechanisms

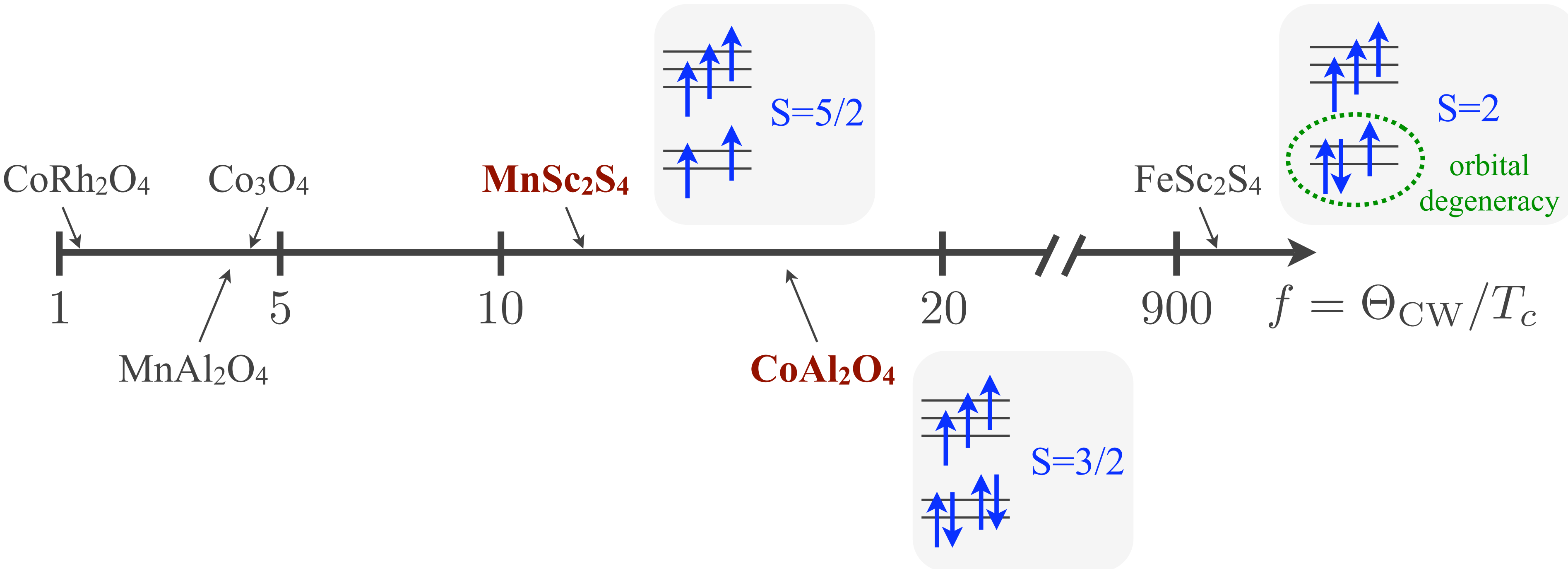
Characterizing "spin liquid" correlations

Diamond lattice antiferromagnets: Materials

V. Fritsch *et al.*, PRL **92**, 116401 (2004); N. Tristan *et al.*, PRB **72**, 174404 (2005); T. Suzuki *et al.* (2007)

Many materials take on the normal spinel structure AB_2X_4.

Focus: Spinels with magnetic A-sites (only).



Very limited theoretical understanding...

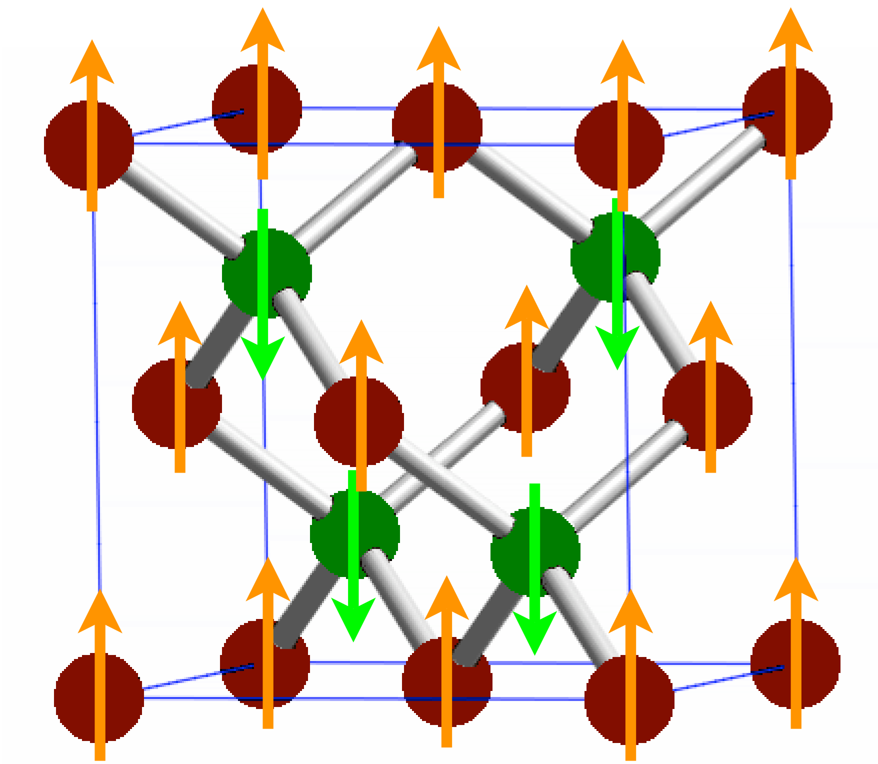
Frustration in the diamond lattice

Naive Hamiltonian $H = J_1 \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j$

antiferromagnetic

classical spins
S=3/2, S=5/2

diamond lattice
two FCC lattices
coupled via J_1



bipartite lattice
no frustration

Frustration in the diamond lattice

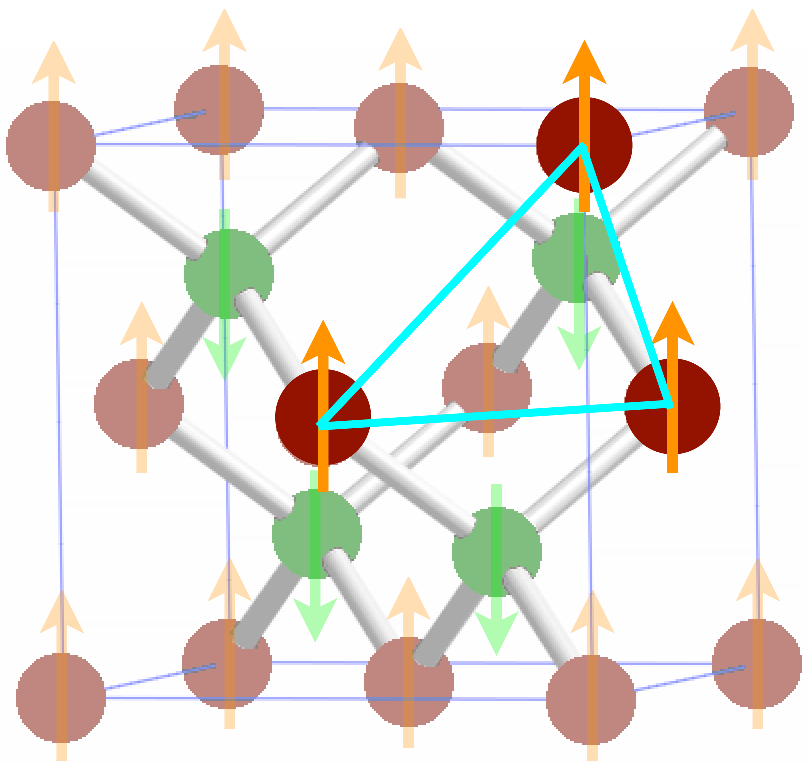
2nd neighbor exchange

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

$J_1 \approx J_2$

similar exchange path

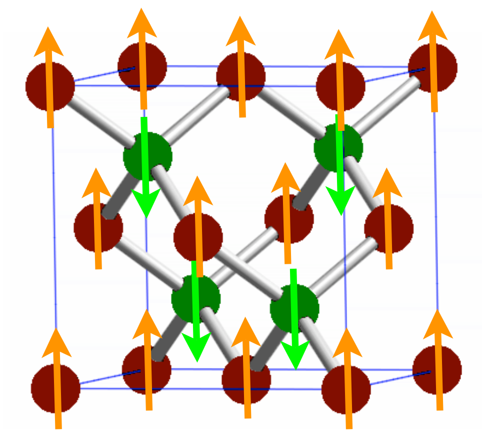
W. L. Roth, J. Phys. **25**, 507 (1964)

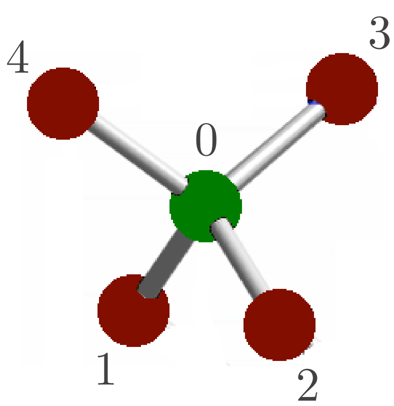


J_2 generates **strong** frustration

Stability of the Néel-ground state

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



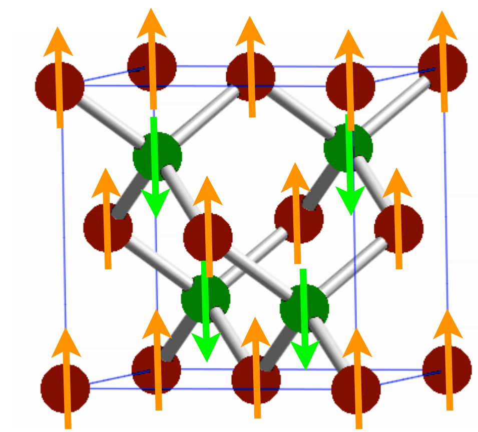


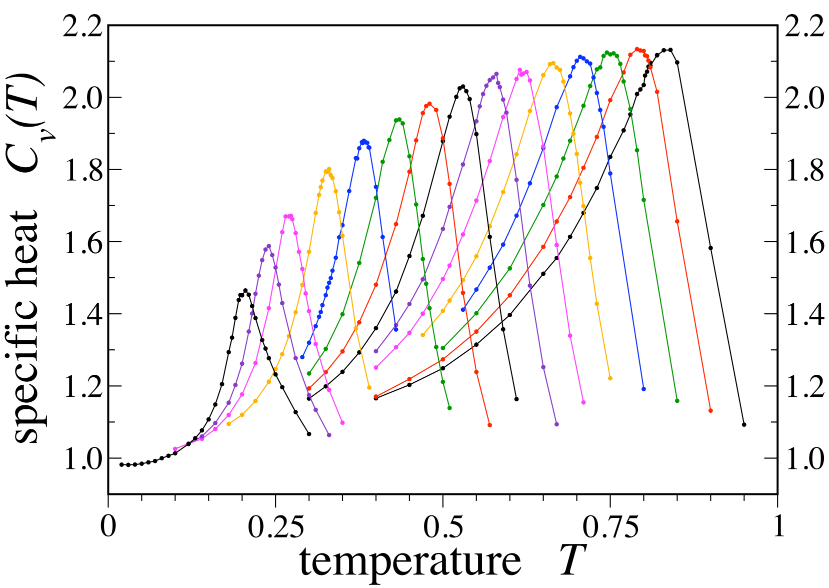
$$H = J_1 \sum_t \left[\vec{S}_0 + \left(\vec{S}_1 + \vec{S}_2 + \vec{S}_3 + \vec{S}_4 \right)/4 \right]^2 + (J_2 - J_1/8) \sum_{\langle\langle ij \rangle\rangle} \vec{S}_i \cdot \vec{S}_j$$

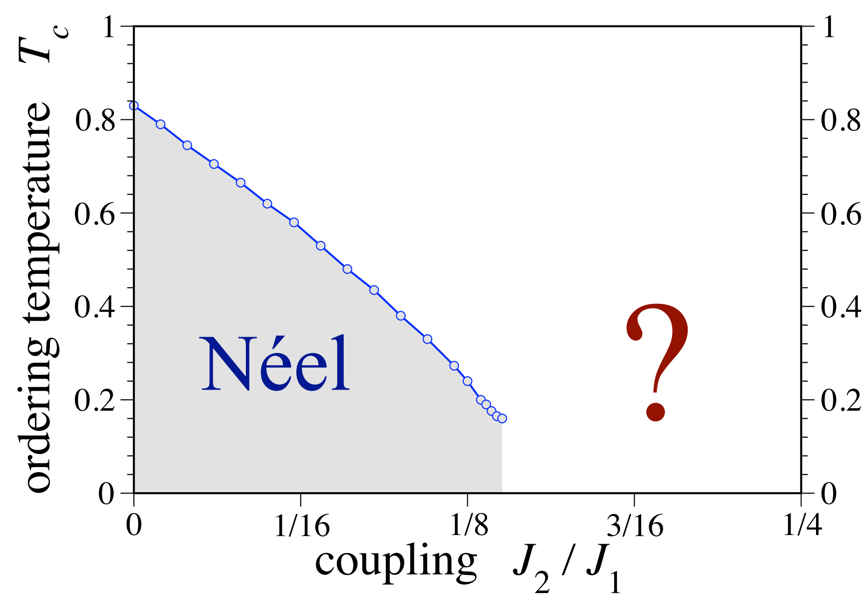
$J_2/J_1 < 1/8$ ➠ Néel state is favored.

Stability of the Néel-ground state

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

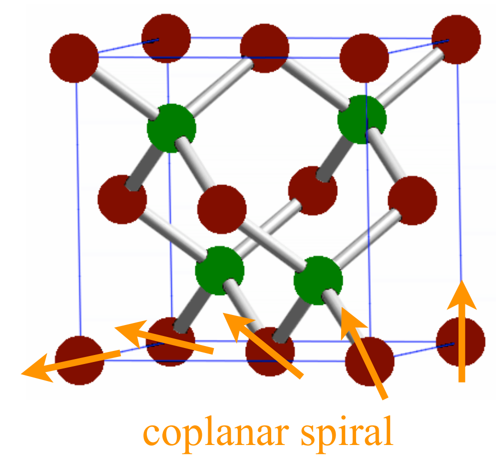






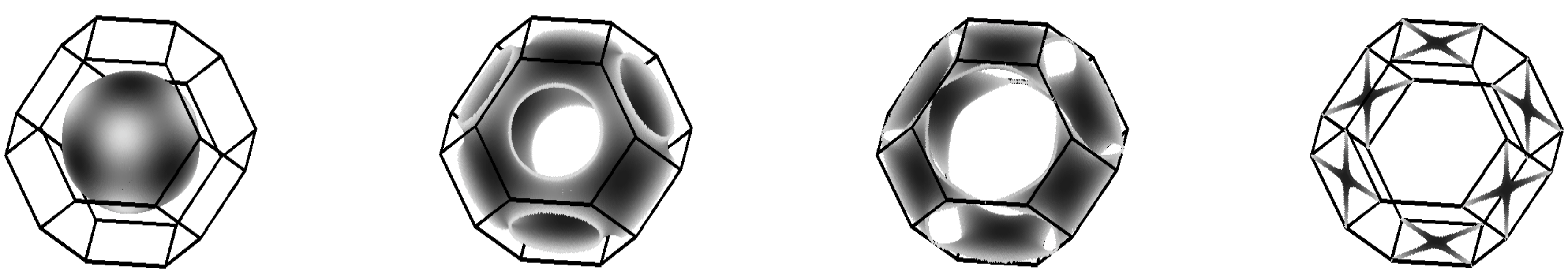
Spiral ground states

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



Direction & pitch of spirals is characterized by a **wavevector** residing on a **surface** in momentum space.

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



Low-T physics: Long-range order?

At **finite temperature** stability non-trivial due to massive spiral degeneracy.

Expand in small fluctuations around spiral ground states

$$\delta\vec{S}_i = \vec{S}_i - \langle\vec{S}_i\rangle$$ ← arbitrary ground-state order

Entropy stabilizes **long-range order** at finite temperature by lifting the degeneracy in the free energy along the spiral surface.

$$\Delta \sim T^{2/3}$$ thermally induced splitting

$$C_v(T) \sim T^{1/3}$$ unconventional thermodynamic behavior

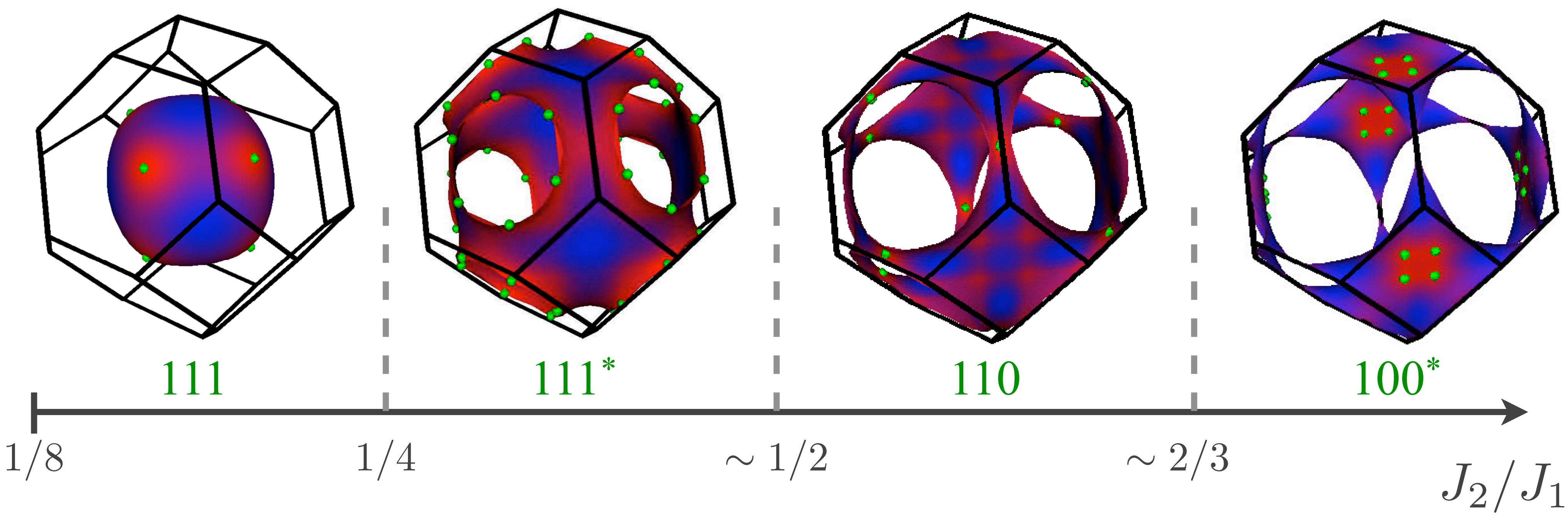
The system undergoes a **thermal order-by-disorder transition**.

Order-by-disorder **selection**

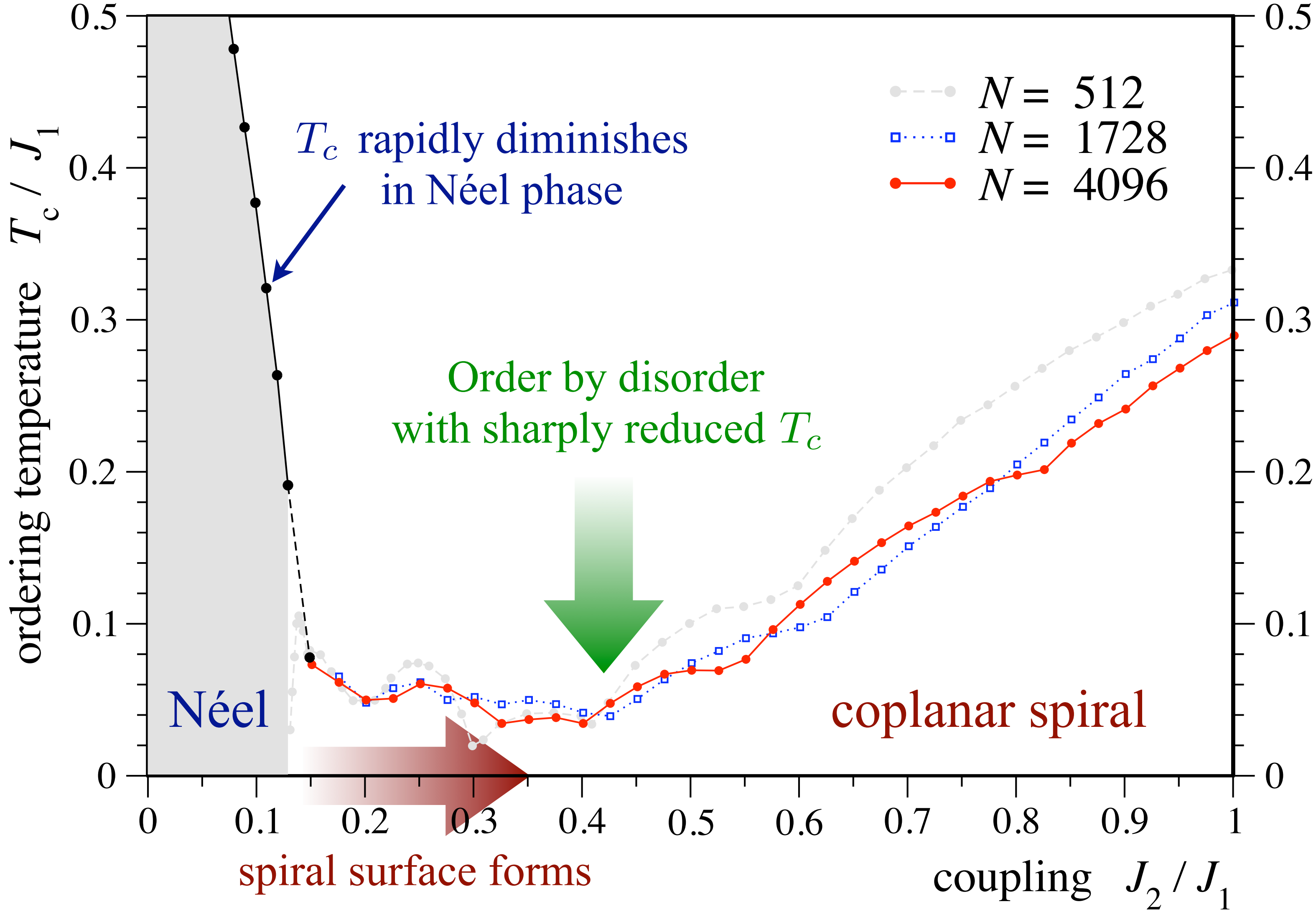
At low temperatures, which state does entropy select?

Free energy on spiral surface $F(Q) = E - TS(Q)$

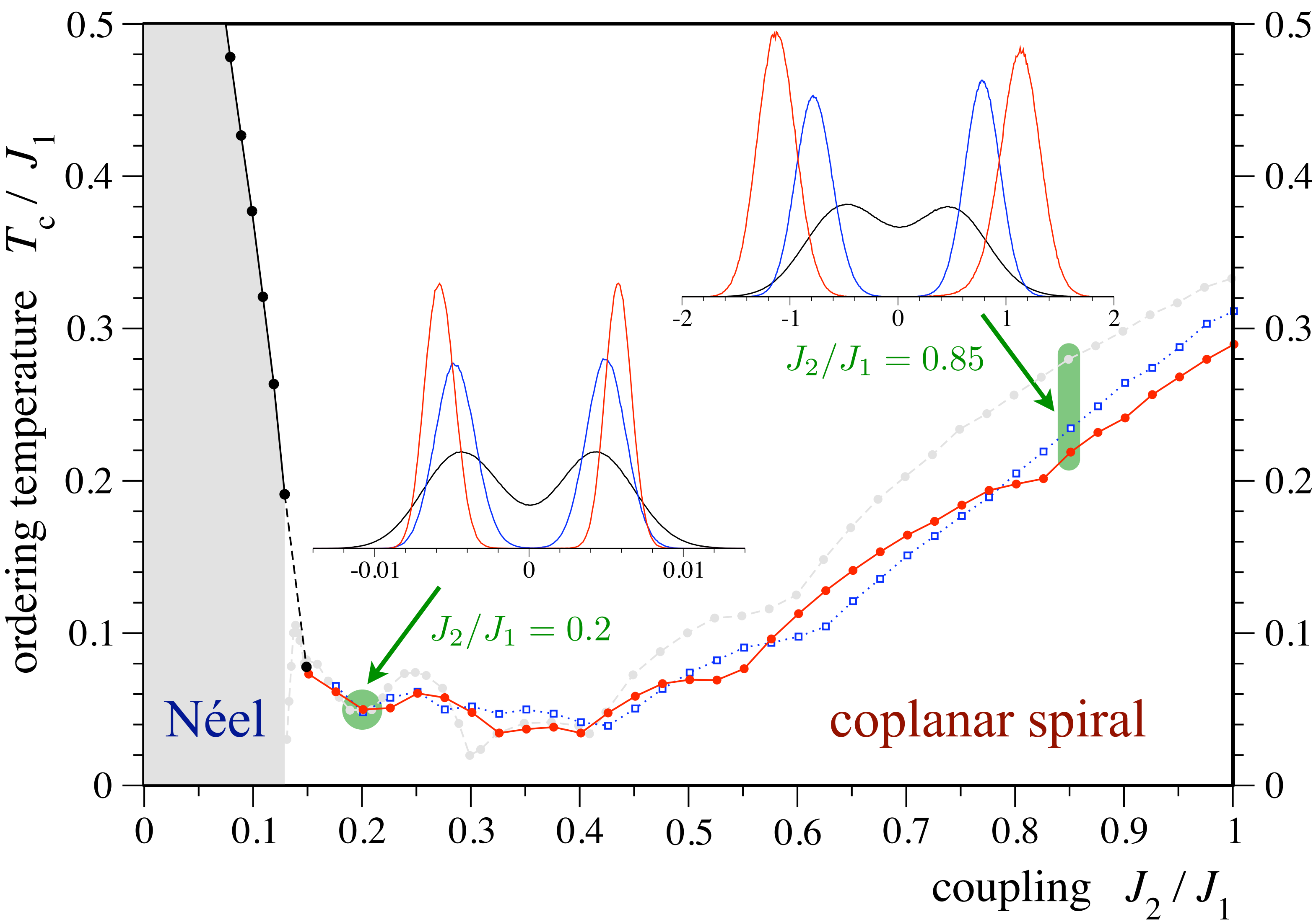
Entropy favors states with **highest density** of nearby low-energy states.



Ordering temperature



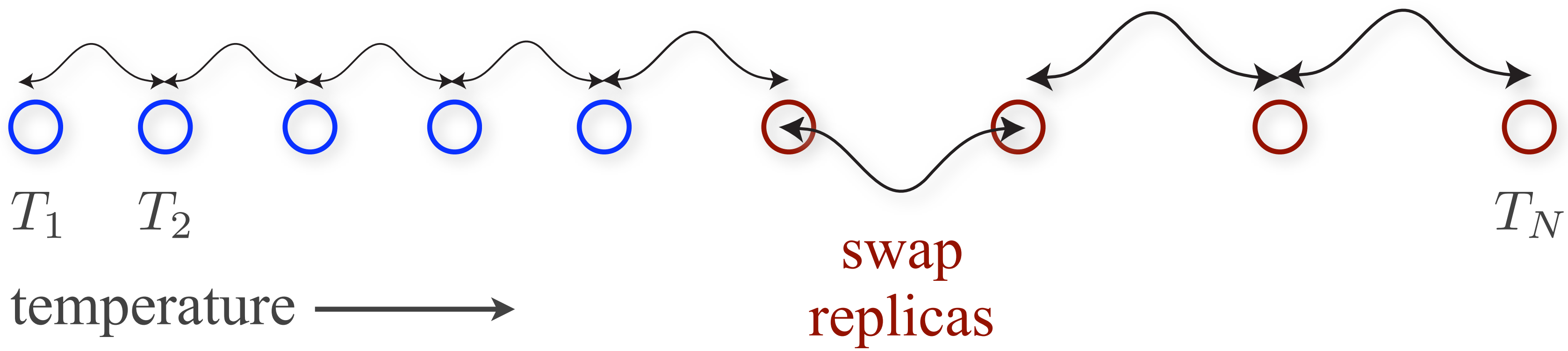
First-order transitions



Parallel tempering

K. Hukushima and Y. Nemoto, J. Phys. Soc. Jpn. **65**, 1604 (1996)

Simulate **multiple replicas** of the system at various temperatures.



$$p(E_i, T_i \rightarrow E_{i+1}, T_{i+1}) = \min(1, \exp(\Delta\beta\Delta E))$$

Single replica performs **random walk** in temperature space.

How do we choose the temperature points?

Ensemble optimization

S. Trebst, D.A. Huse, M. Troyer, PRE **70**, 046701 (2004)
H.G. Katzgraber, S. Trebst, D.A. Huse, M. Troyer, JSTAT P03018 (2006)

Feedback algorithm

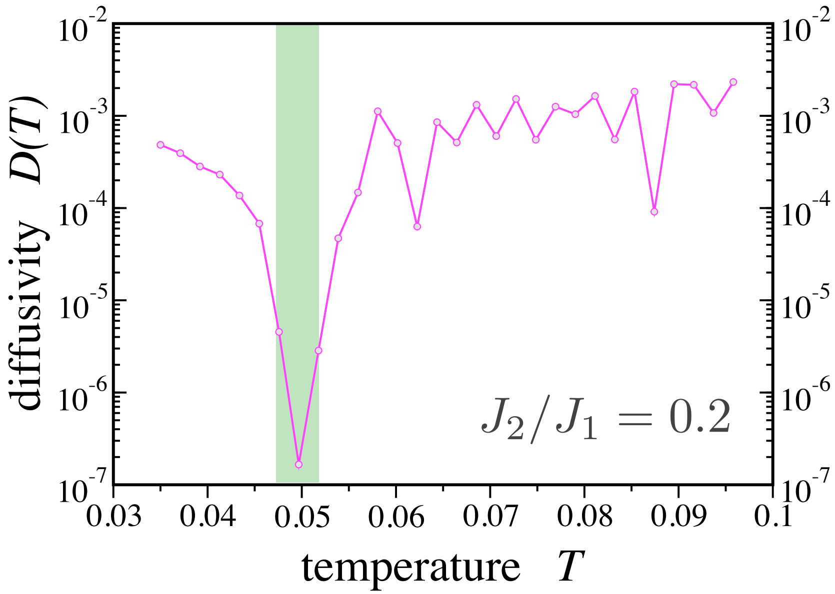
Measure **local diffusivity** $D(T)$ of current in temperature space.

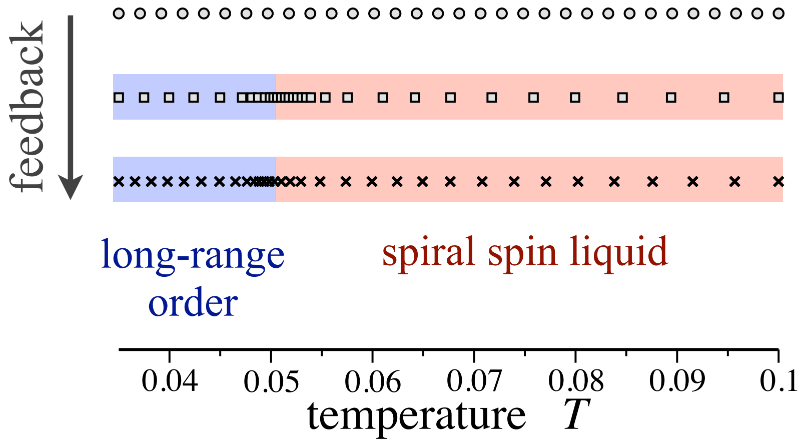
Optimal choice of temperatures

$$\eta^{\text{opt}}(T) \sim \frac{1}{\sqrt{D(T)}}$$

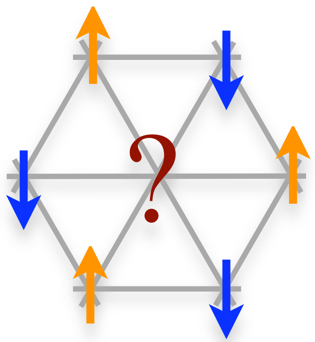
density of T-points

Iterate feedback of diffusivity.



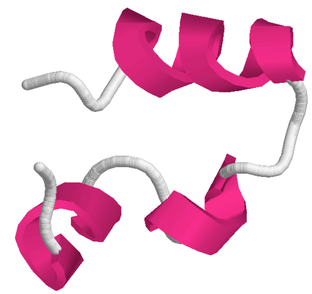


Ensemble optimization



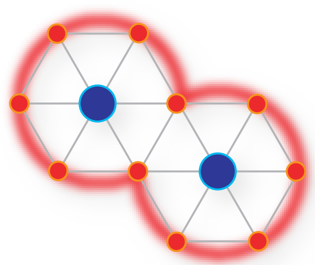
frustrated magnets

S. Trebst *et al.*, Phys. Rev. E (2004)



small proteins

S. Trebst *et al.*, J. Chem. Phys. (2006)



dense liquids

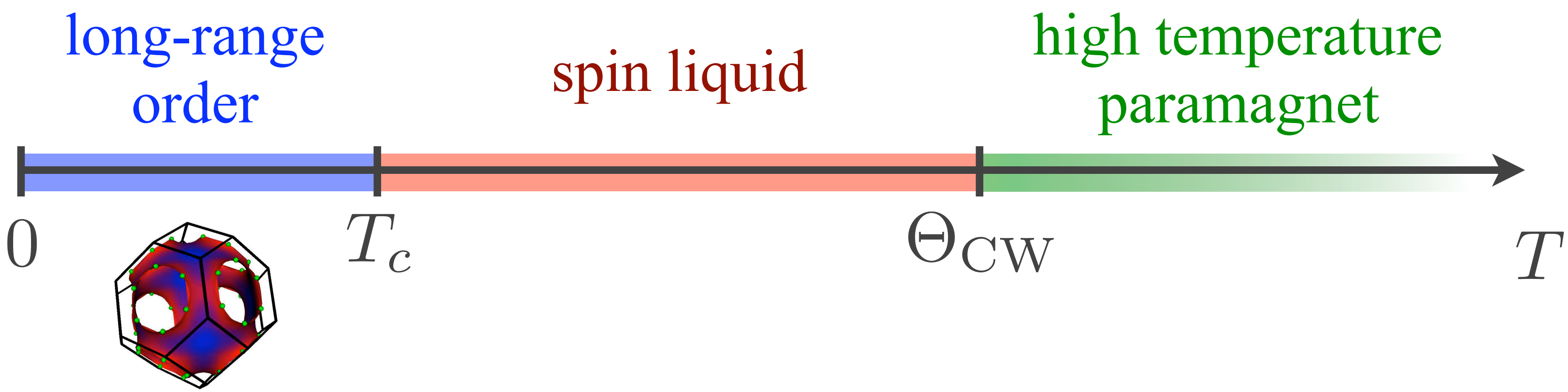
S. Trebst *et al.*, J. Chem. Phys. (2005)

$$Z = \mathrm{Tr} e^{-\beta H} = \sum_{n=0}^{\infty} g(n) \beta^n$$

quantum systems

S. Wessel *et al.*, JSTAT (2007)

Spin liquid physics



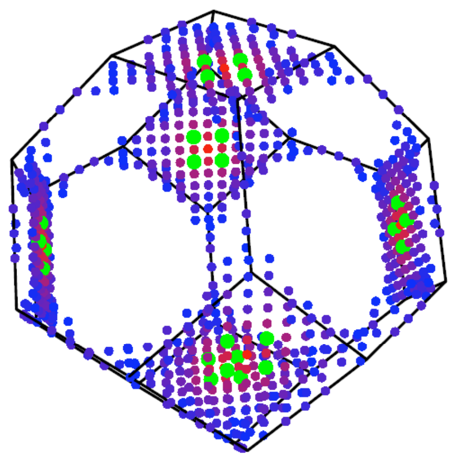
Order-by-disorder selects low temperature coplanar spiral state.

Broad **spin liquid regime** emerges due to low T_c.

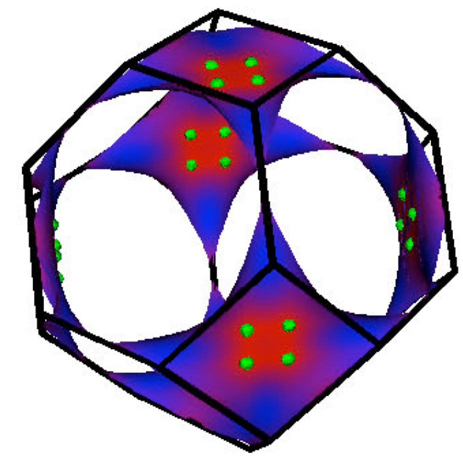
How can we characterize the spin liquid regime?
Is there an experimental probe to observe the spin liquid?

Magnetic correlations in spin liquid

numerical **structure factor**



analytic **free energy**



$J_2/J_1 = 0.85$

$MnSc_2S_4$

Spin structure factor directly images **"spiral surface"**.

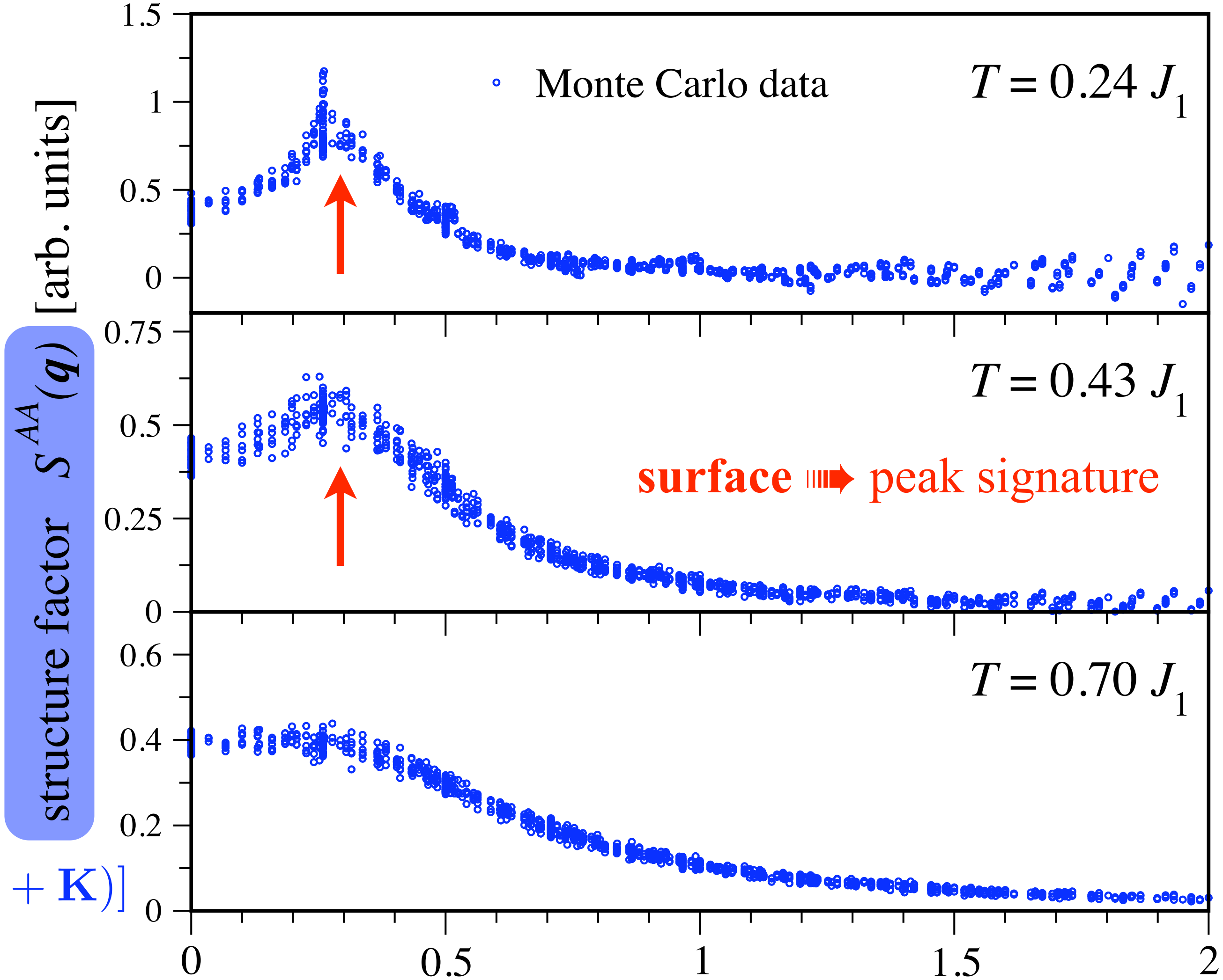
Free energy corrections visible for $T_c < T < 1.3T_c$

Spin liquid correlations

$J_2/J_1 = 0.85$

$MnSc_2S_4$

structure factor for
one FCC sublattice



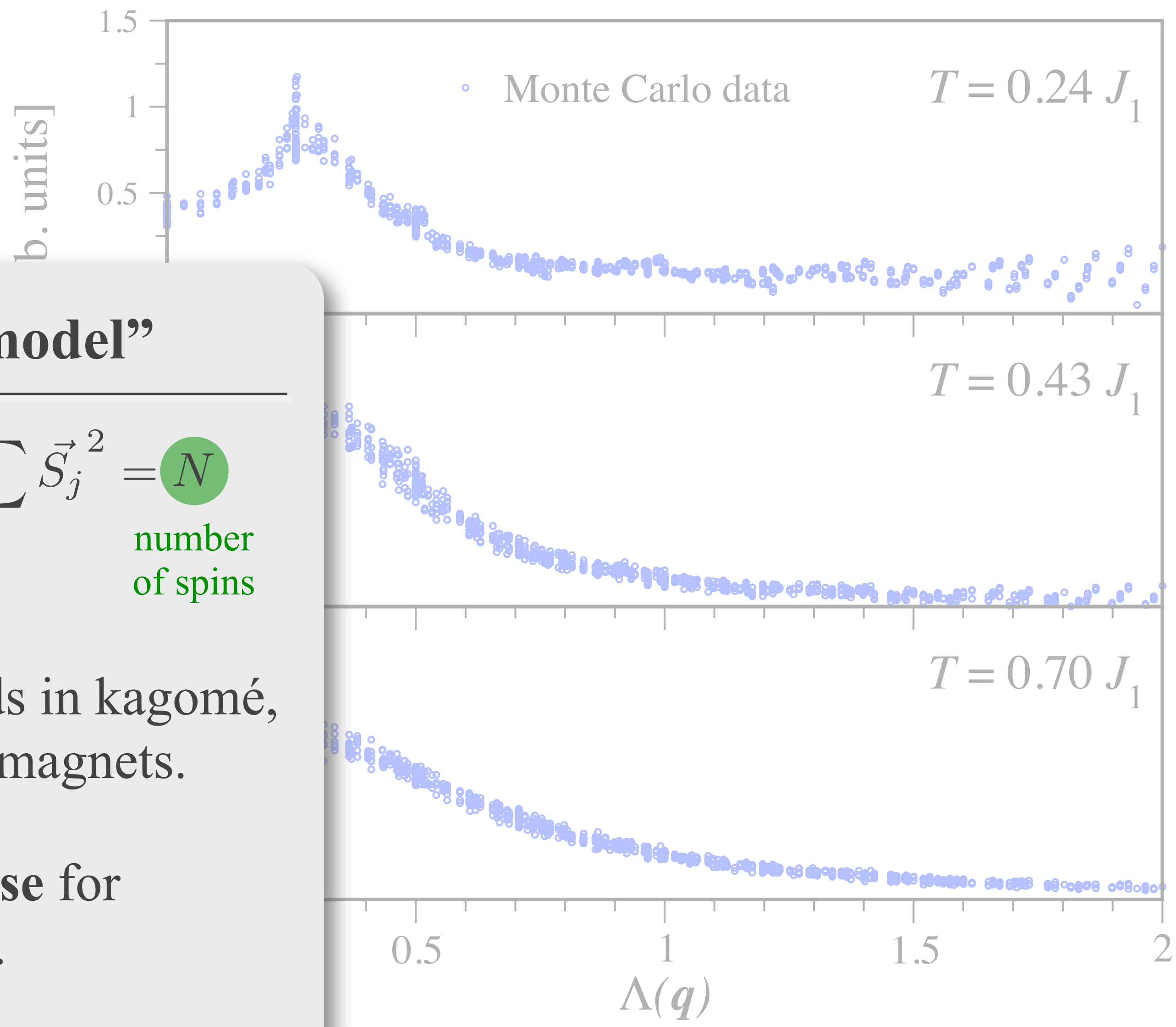
$$S^{AA}(\mathbf{q}) = \frac{1}{2}\left[S(\mathbf{q}) + S(\mathbf{q}+\mathbf{K})\right]$$

$$\mathbf{K} = 4\pi(1,0,0)$$

$$\Lambda(\mathbf{q}) = 2\sqrt{\cos^2\frac{q_x}{4}\cos^2\frac{q_y}{4}\cos^2\frac{q_z}{4} + \sin^2\frac{q_x}{4}\sin^2\frac{q_y}{4}\sin^2\frac{q_z}{4}}$$

"surface function"

Spin liquid correlations



"Spherical model"

$$\vec{S}_j^{\,2} = 1 \quad \rightarrow \quad \sum_j \vec{S}_j^{\,2} = N$$

number of spins

describes spin liquids in kagomé, pyrochlore antiferromagnets.

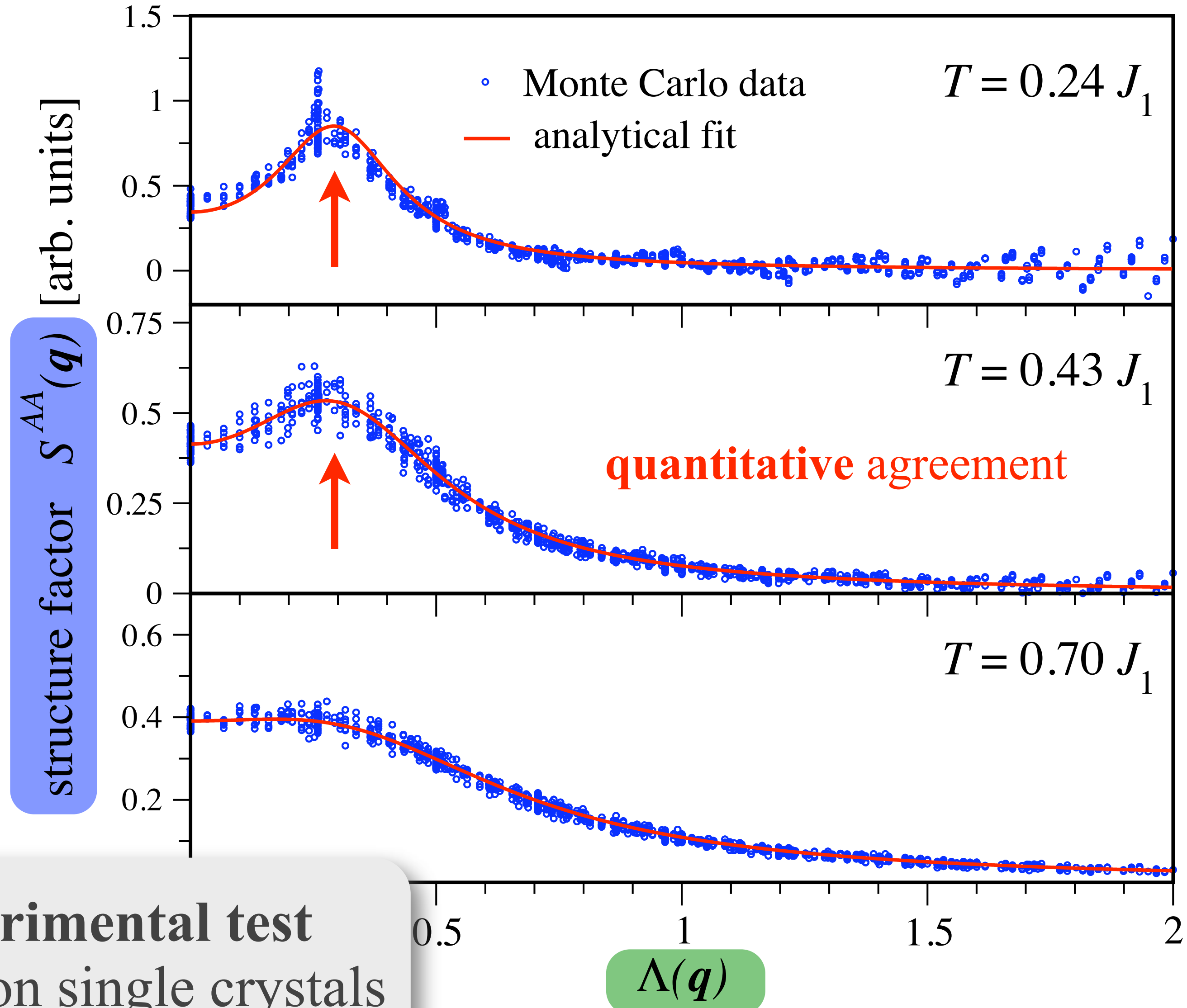
predicts **data collapse** for structure factor data.

Spin liquid correlations

$J_2/J_1 = 0.85$

$MnSc_2S_4$

structure factor for one FCC sublattice



"surface function"

Non-trivial eperimental test
neutron scattering on single crystals

Multistage ordering

Realistic Hamiltonian

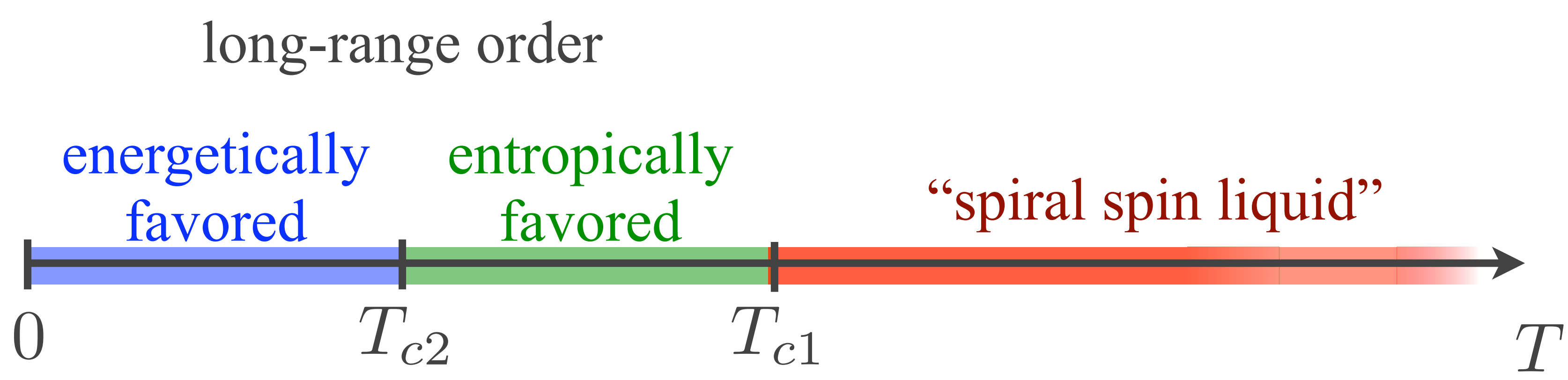
$$H = J_1 \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{\langle\langle ij \rangle\rangle} \vec{S}_i \cdot \vec{S}_j + \delta H$$

$$\delta H = J_3 \sum_{\langle\langle\langle ij \rangle\rangle\rangle} \vec{S}_i \cdot \vec{S}_j$$

degeneracy breaking perturbations

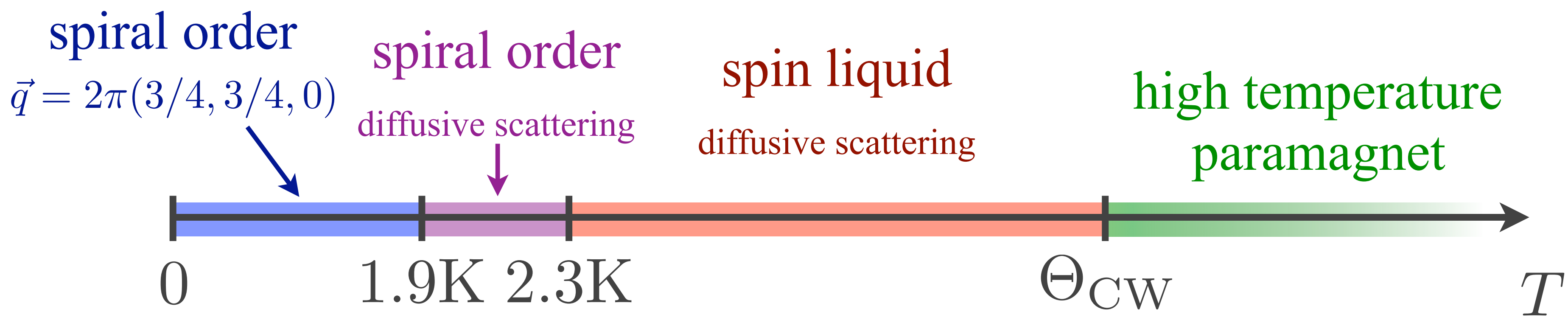
Entropic corrections vanish as $T \to 0$. (free energy $F = E - TS$)

Energetic corrections from δH inevitably dominate at lowest T.



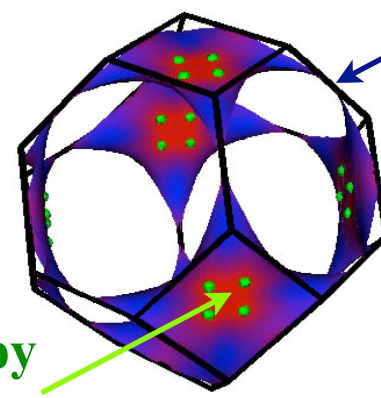
$MnSc_2S_4$

A. Krimmel *et al.*, PRB **73**, 014413 (2006); M. Müksch *et al.* (2007)



Theoretical implications

- J_1 is **ferromagnetic**
- $J_2/|J_1| \approx 0.85$
- $\Theta_{CW} \approx -22.1\text{K}$ gives
  $J_1 \approx 1.2\text{K} \quad J_2 \approx 1.0\text{K} \quad T_c \approx 2.4\text{K}$
- low-T order determined **energetically**, not entropically



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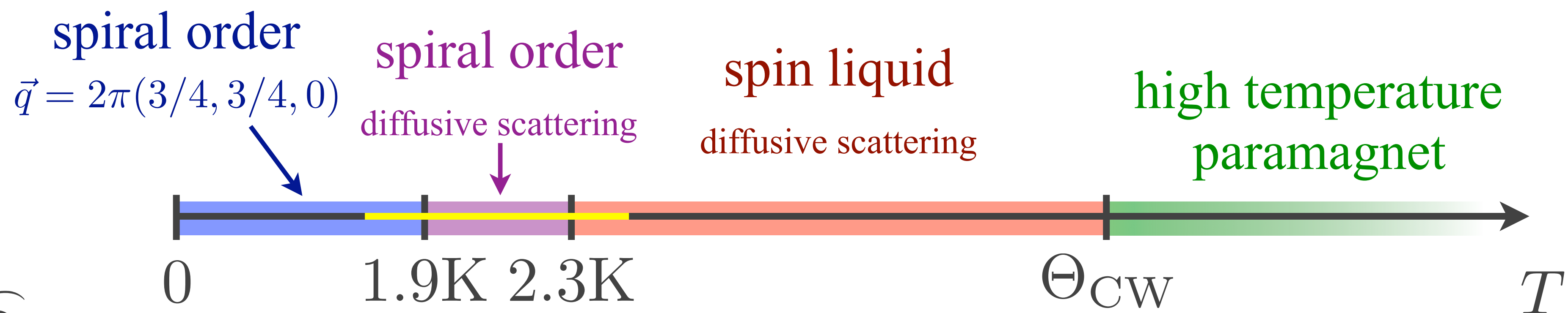
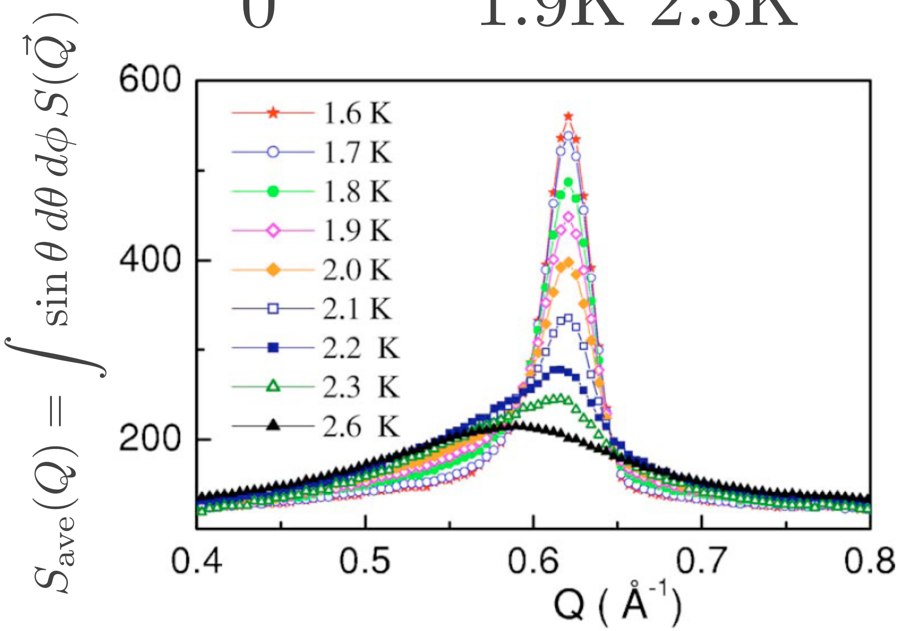
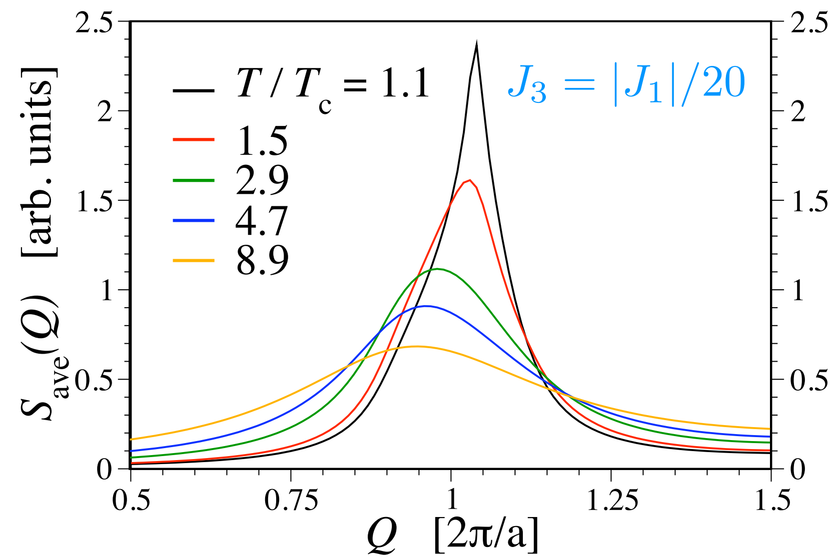






Intensity shifts from $|\vec{q}|$ to "spiral surface" as T washes out J_3.

Consistent with "spiral spin liquid".

$CoAl_2O_4$

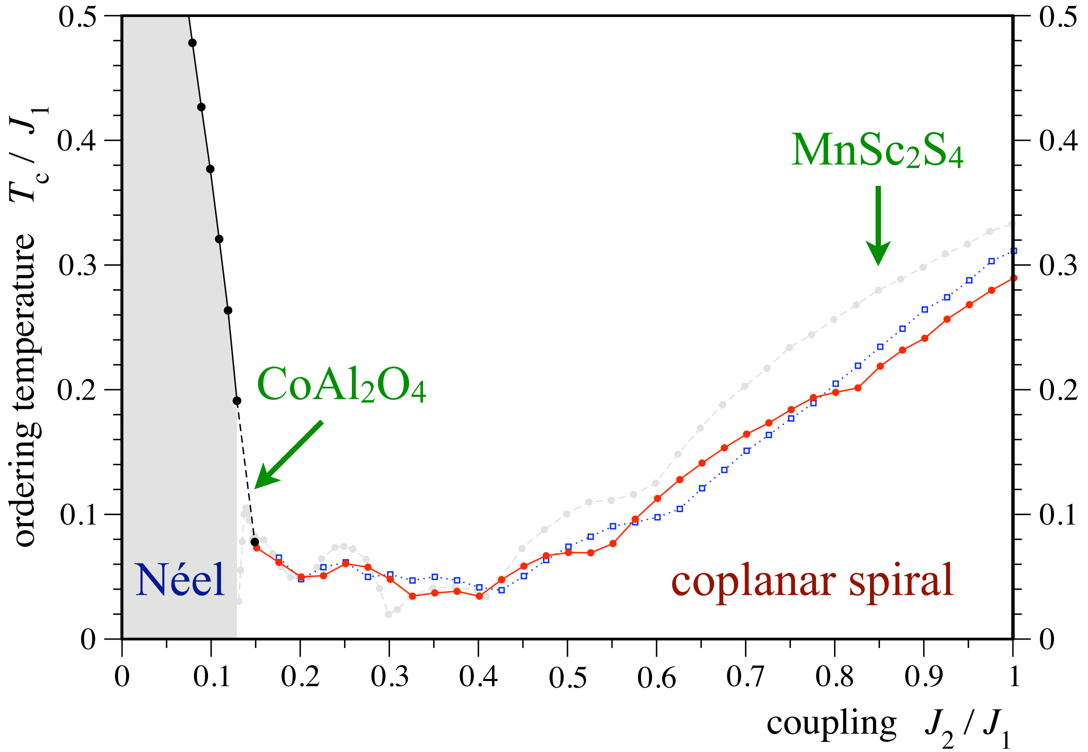
N. Tristan *et al.*, PRB **72**, 174404 (2005); A. Krimmel *et al.*, Physica B **378-380**, 583 (2006); T. Suzuki *et al.* (2007)

Experimental observations

- strong frustration, sample dependent
- no sharp transition observed yet

Theoretical implications

- powder neutron data + frustration suggest $J_2/J_1 \approx 1/8$



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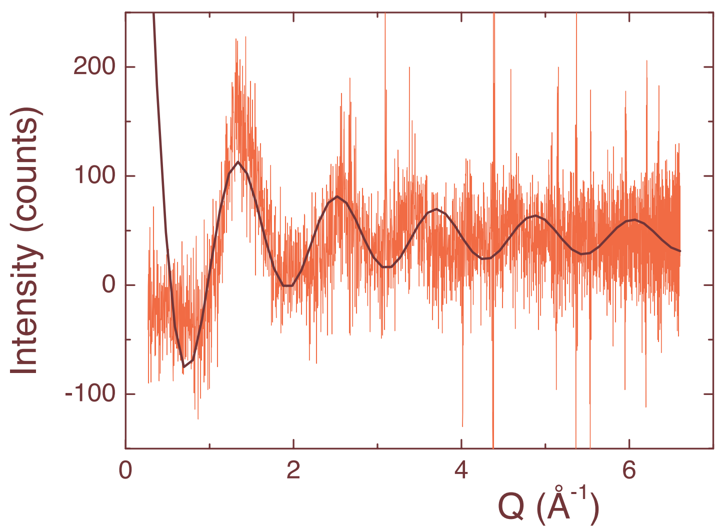
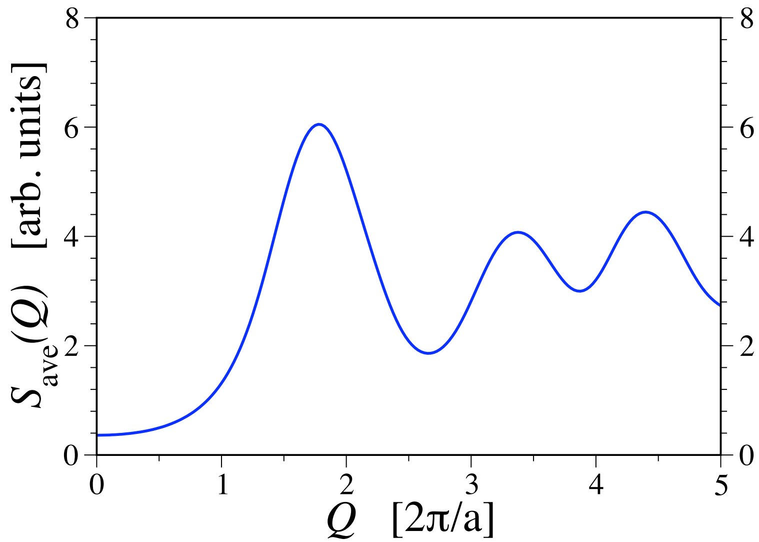
experiment

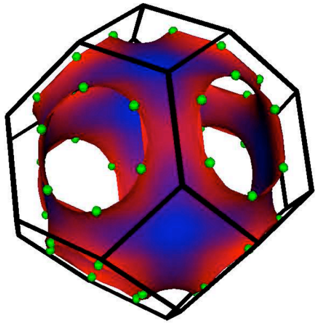


theory



Summary

- Many spinels constitute frustrated diamond lattice AFMs
  - $MnSc_2S_4$, $CoAl_2O_4$, etc.
- Simple J_1-J_2 model captures essential physics
  - Continuous spiral ground-state degeneracy
  - Manifestation of order-by-disorder physics
  - Spin correlations in spiral spin liquid regime reveal ordering surface and entropic effects



- Theoretical predictions consistent with experiments.

D. Bergman, J. Alicea, E. Gull, S. Trebst, and L. Balents   Nature Physics **3**, 487 (2007)

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