Classical and Quantum Frustrated Magnets

Yong Baek Kim
University of Toronto

University of Virginia, November 1, 2007

Collaborators:

J. Hopkinson (Toronto), S. Isakov (Toronto), M. Lawler (Toronto), H.Y. Kee (Toronto)
F. Wang (Berkeley), A. Vishwanath (Berkeley),
S. Sachdev (Harvard), L. Friz (Harvard)

Canada Research Chairs

NSERC CRSNG

CIAR

Outline

Introduction to Frustrated Magnets

Spin-1/2 Quantum Magnets; Real Materials

Review of the Heisenberg model
on the Kagome lattice

Distorted Kagome (Volborthites) Lattices

Hyper-Kagome ($Na_4Ir_3O_8$) Lattice

Zn-Paratacamite Lattice

Introduction to Frustrated Magnets

Geometric Frustration:

the arrangement of spins on a lattice precludes (fully) satisfying all interactions at the same time

Modern: large degeneracy of the (classical) ground state manifold $\sim e^{\alpha N}$

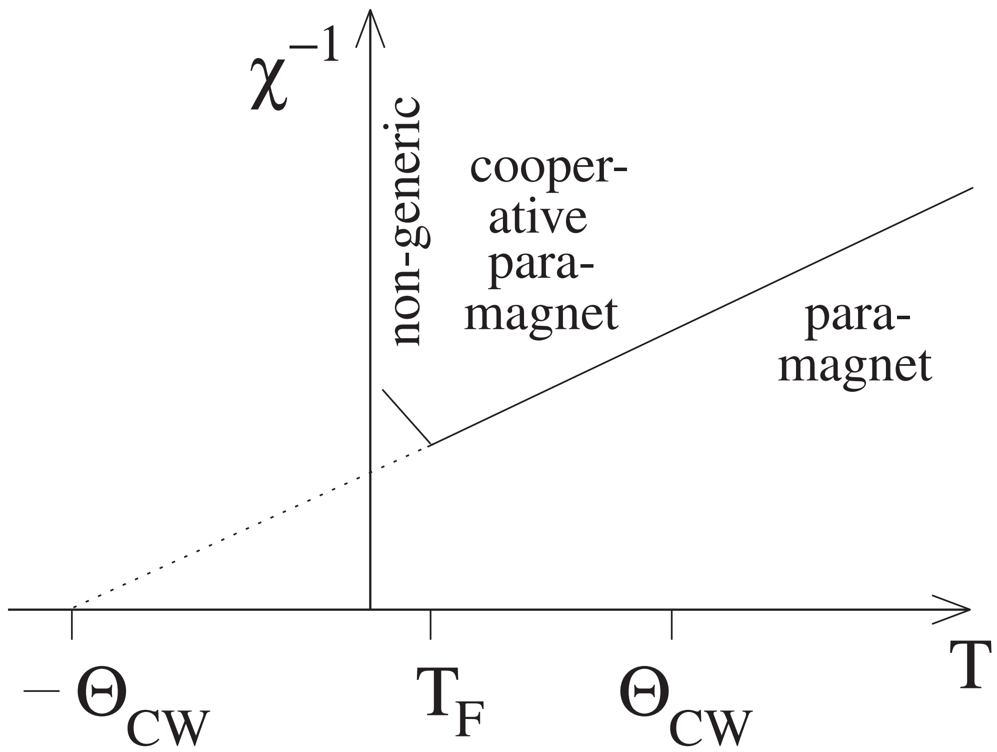
Consequence:

No energy scale of its own; any perturbation is strong (reminiscent of the lowest Landau level physics)

Mother of the conventional and exotic phases

Introduction to Frustrated Magnets

Susceptibility 'fingerprint':



Θ_{CW} Curie-Weiss temperature
mean-field ordering temp.
interaction energy scale

$T_F/\Theta_{\mathrm{CW}} \ll 1$ strong frustration

$T_F < T < \Theta_{CW}$

Cooperative paramagnet:
correlations remain weak
more universal

$T < T_F$

Magnetically ordered ?
Spin liquid ? Glassy ?
not universal

Origin of Classical Ground State Degeneracy

Classical nearest-neighbor antiferromagnetic Heisenberg model on lattices with corner-sharing simplexes (simplex = triangle, tetrahedron)

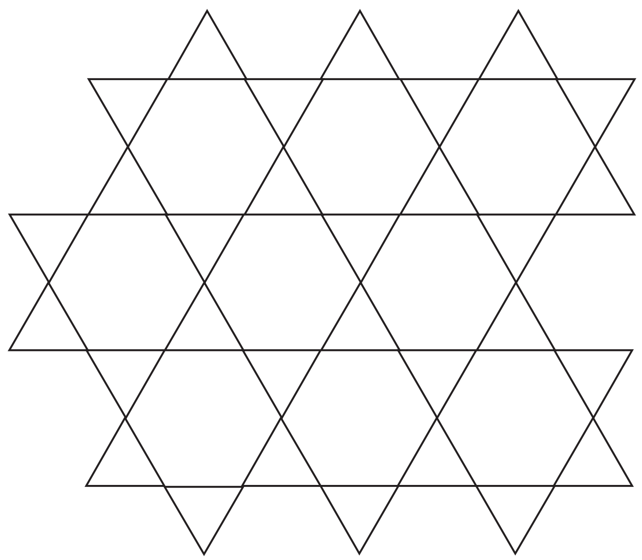
$$H = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j = \frac{J}{2} \sum_{\text{simplex}} \left(\sum_{i \,\epsilon\, \text{simplex}} \mathbf{S}_i \right)^2$$

$\mathbf{S}_i$ is a vector with a fixed length

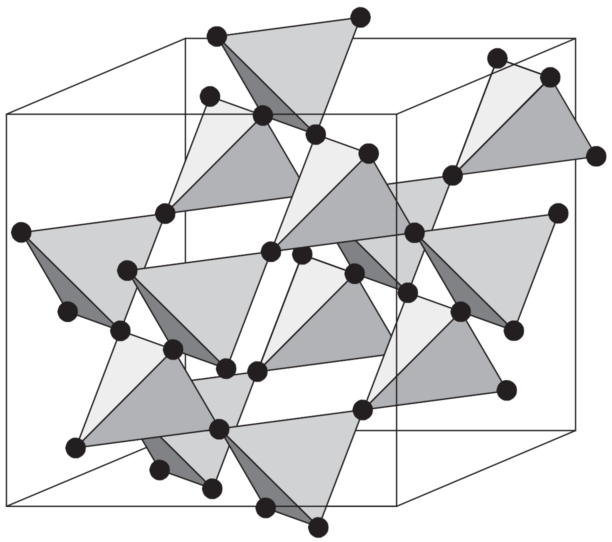
Classical ground state should satisfy $\sum_{i \,\epsilon\, \text{simplex}} \mathbf{S}_i = 0$

These constraints are not independent; counting is subtle

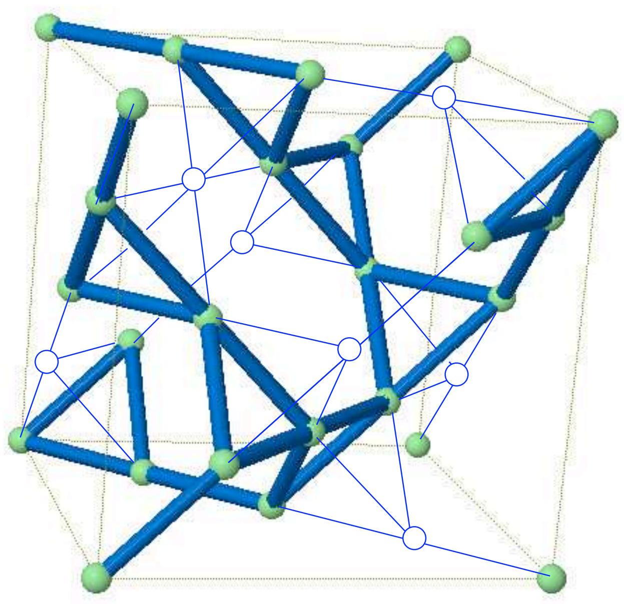
Nonetheless there exists macroscopic degeneracy



Kagome



Pyrochlore



Hyper-Kagome

Order by Disorder

Order by Disorder via Thermal Fluctuations:

Different entropic weighting to each ground state

Softer the fluctuations around a particular ground state, more likely this ground state will be entropically favored.

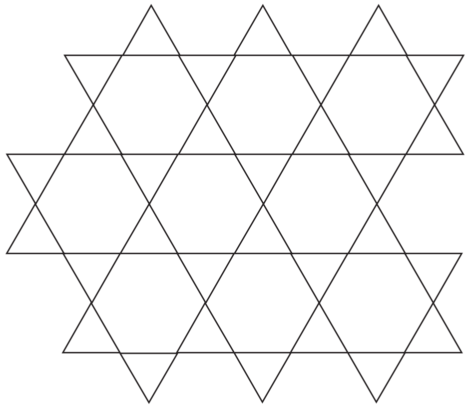
Order by Disorder via Quantum Fluctuations:

Quantum zero point energy and anharmonic contributions may select an ordered ground state.

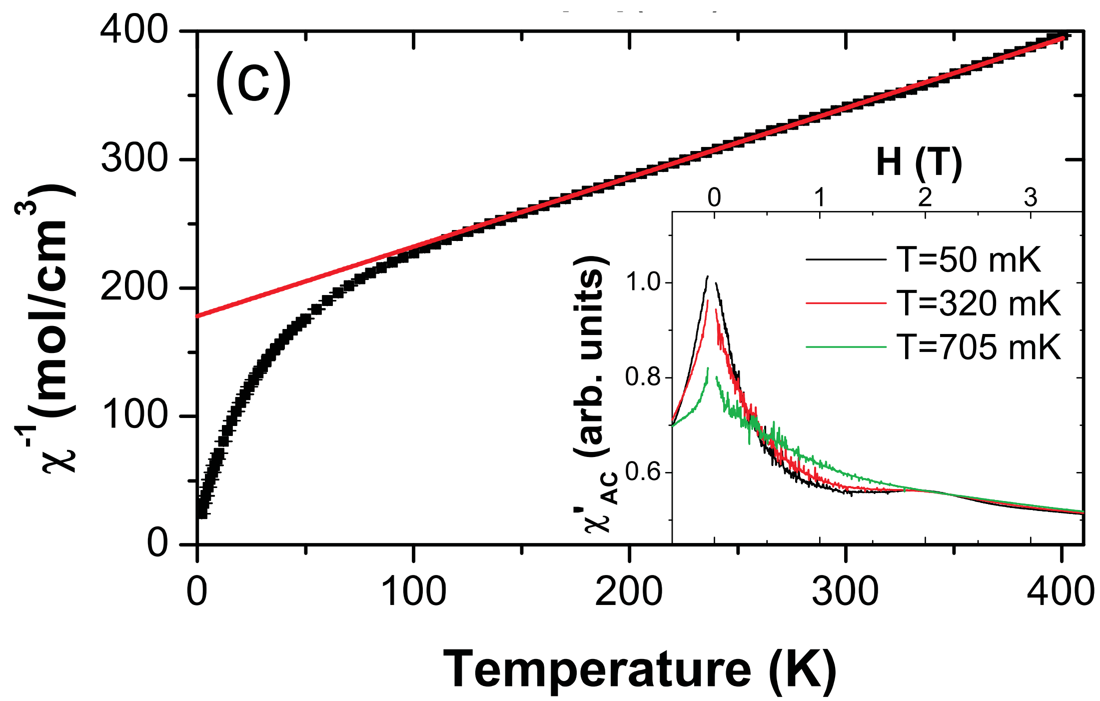
Sufficiently strong quantum fluctuations (S=1/2 for example), however, may destabilize any ordered phase; possible quantum spin liquid - Disorder by Disorder

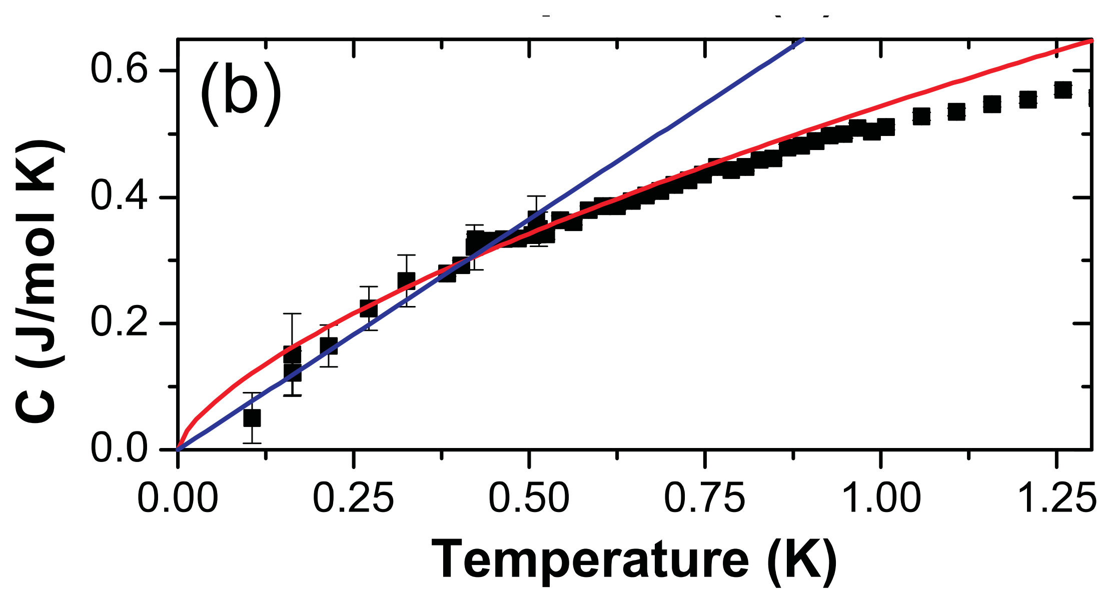
S=1/2 Frustrated Magnets; Real Materials

Ideal Kagome lattice



Herbertsmithite $\mathrm{ZnCu_3(OH)_6Cl_2}$





$\Theta_{\mathrm{CW}} = -300 \pm 20K \quad J \approx 197K$ (fit to series expansion)

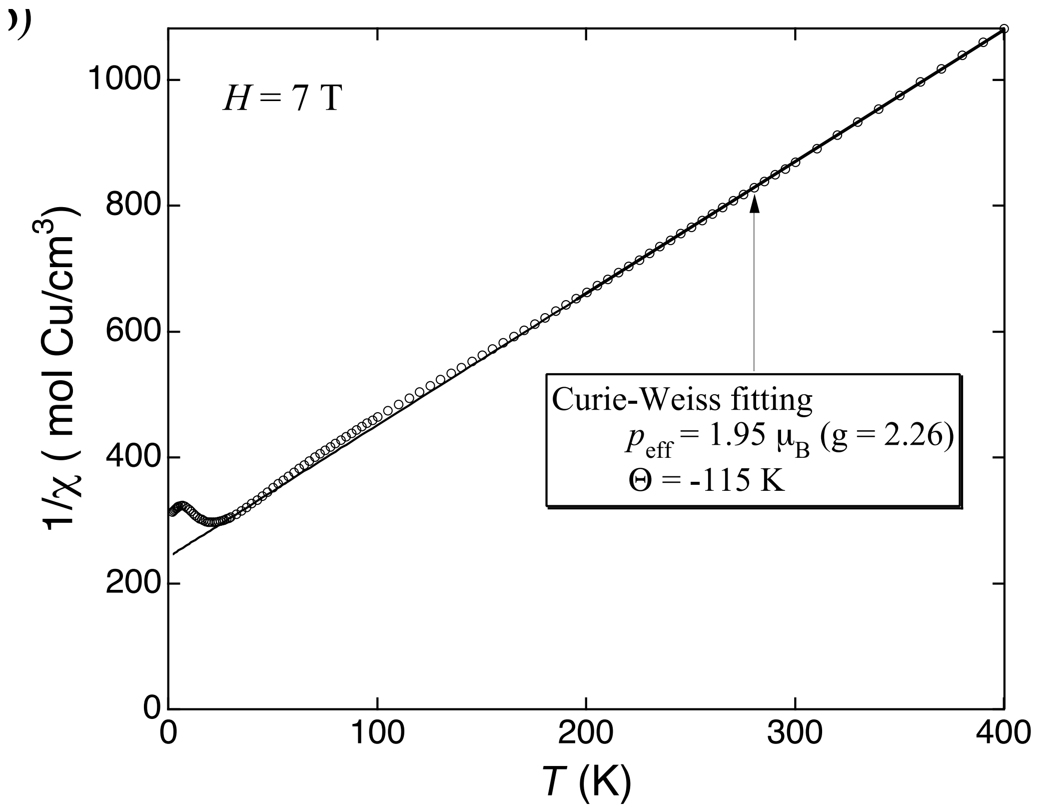
J. S. Helton et al, PRL 98, 107204 (2007)

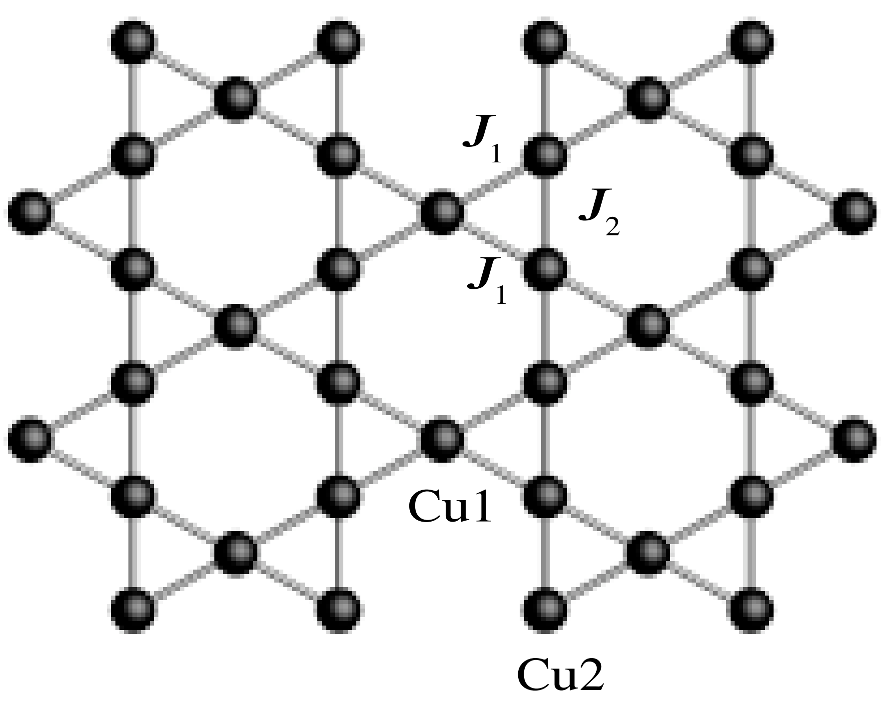
S=1/2 Frustrated Magnets; Real Materials

Volborthite

$$Cu_3V_2O_7(OH)_2 . 2H_2O$$

Distorted Kagome lattice

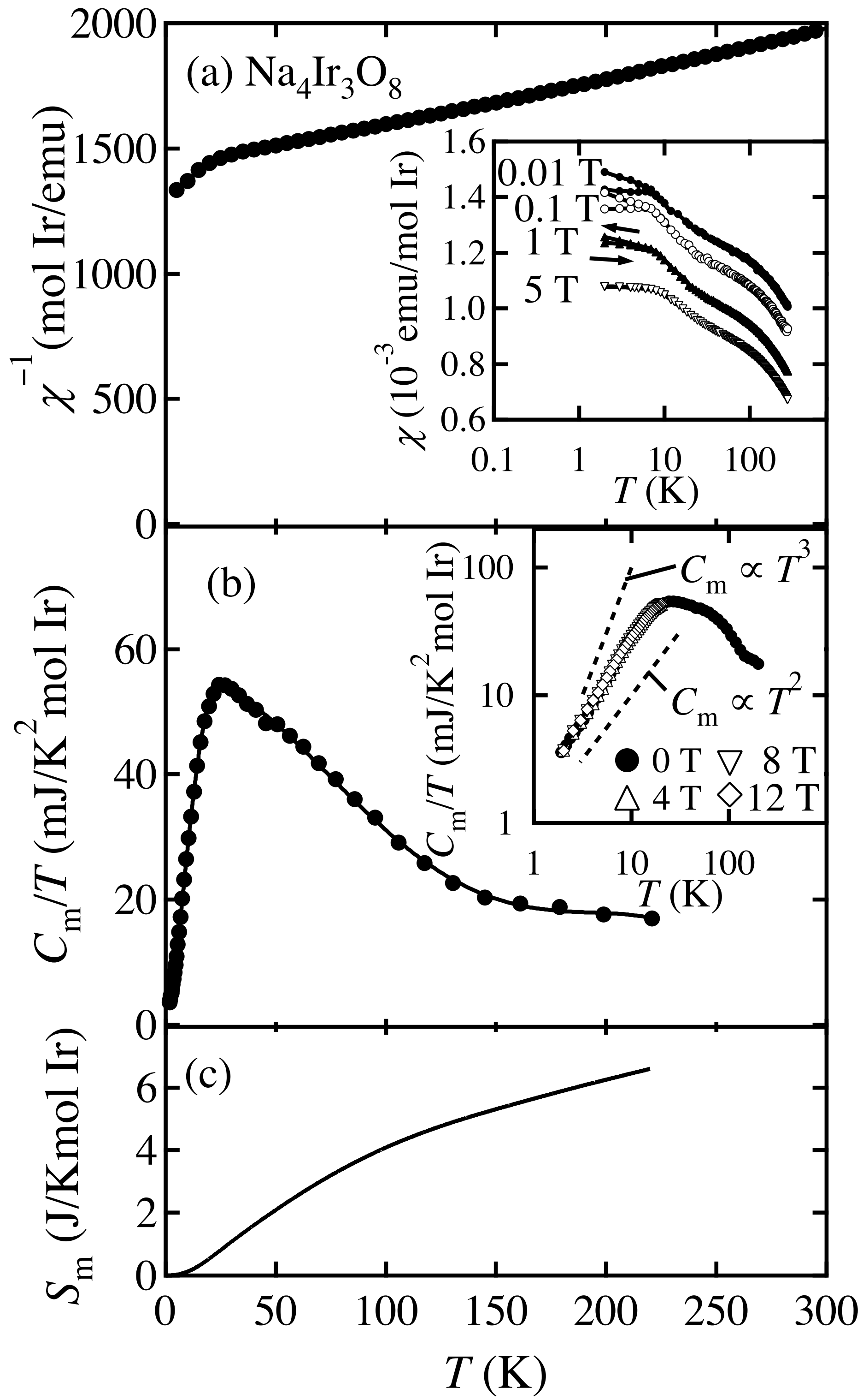




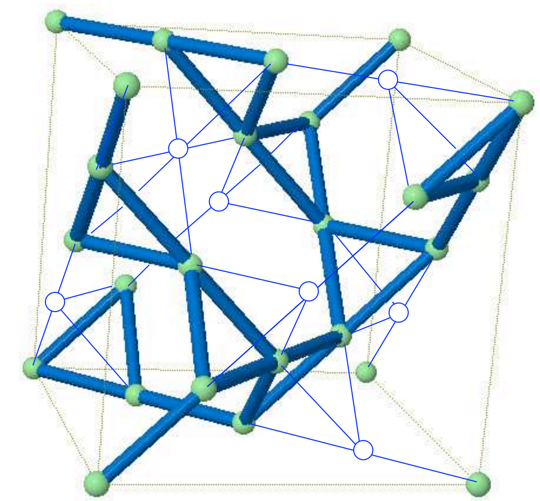
$$\Theta_{CW} = -115K$$

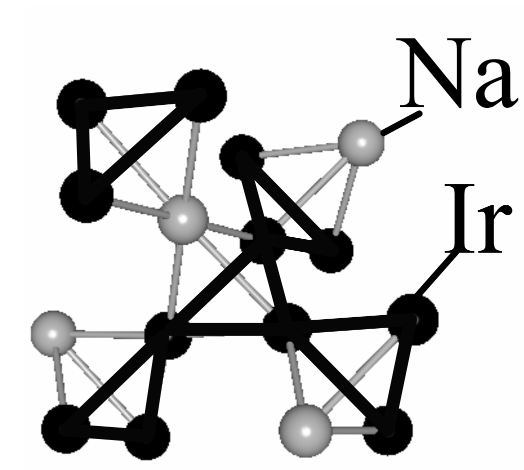
Z. Hiroi et al, JPSJ 70, 3377 (2001); F. Bert et al, PRL 95, 087203 (2005)

S=1/2 Frustrated Magnets; Real Materials



Hyper-Kagome $Na_4Ir_3O_8$



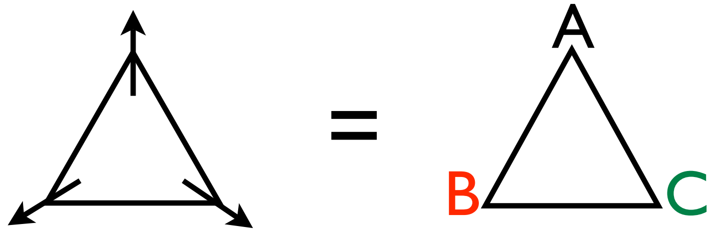


$$\Theta_{CW} = -650K$$

Y. Okamoto, M. Nohara, H. Agura-Katrori, and H. Takagi, arXiv:0705.2821 (2007)

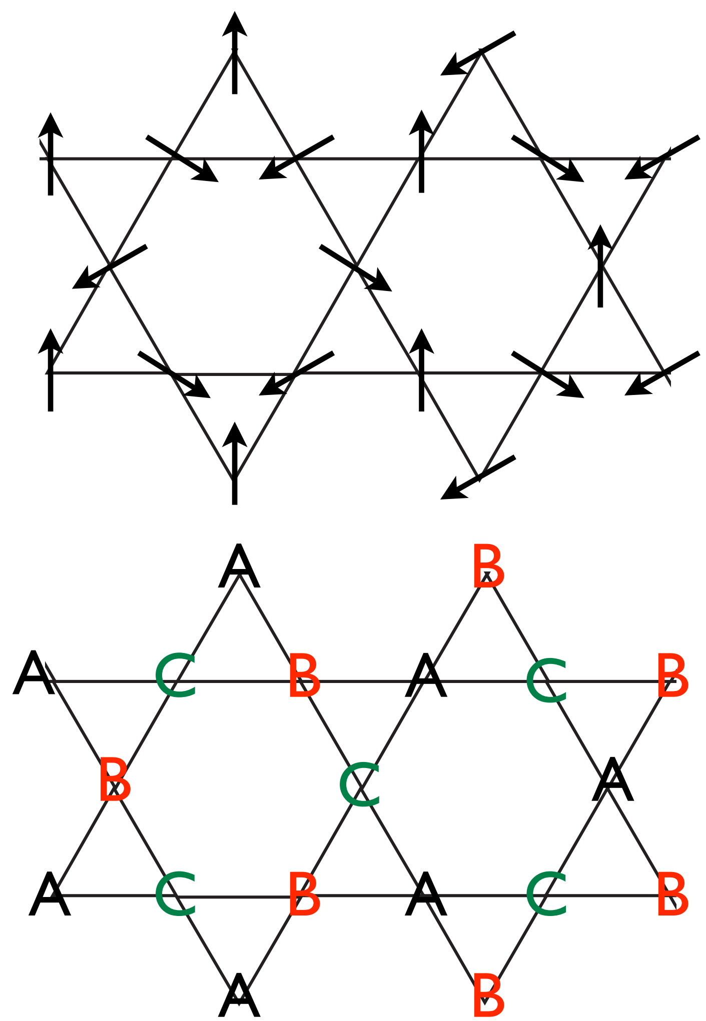
Classical Heisenberg Model on Kagome Lattice

Consider first co-planar states $\sum_{i \,\epsilon\, \triangle} \mathbf{S}_i = 0$

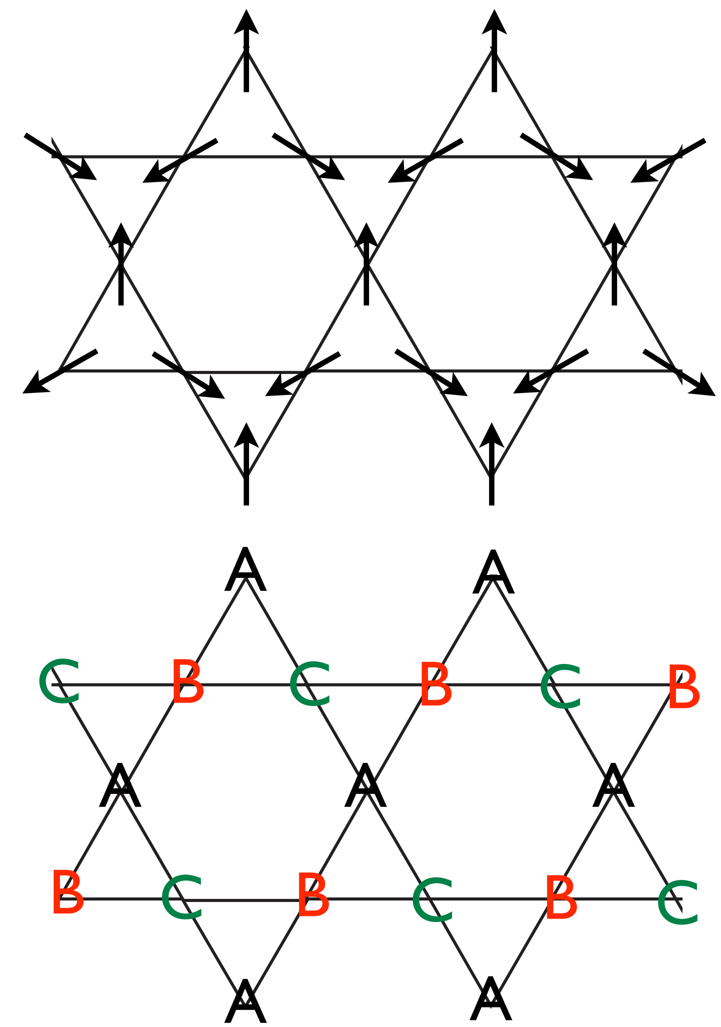


3-state Potts spins

$\sqrt{3} \times \sqrt{3}$



$q = 0$

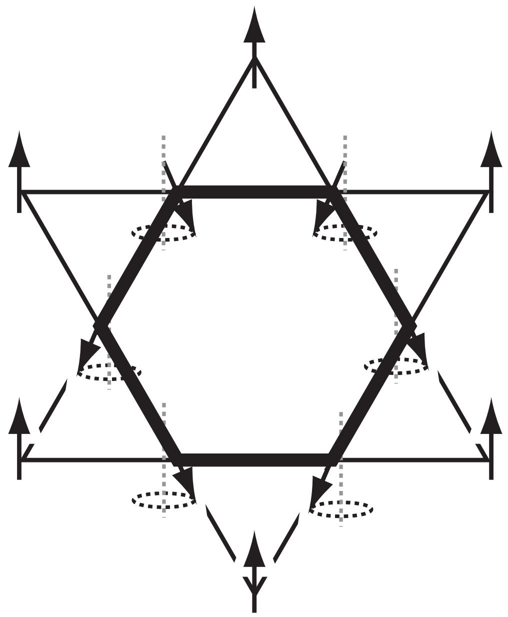


Degeneracy of the co-planar states is extensive;

$e^{0.379N}$ (from 3-state Potts antiferromagnet)

D.A. Huse, A. Rutenberg, PRB 45, 7536 (1992)

Non-planar states can be generated by continuous distortions of a planar state



Weathervane loop

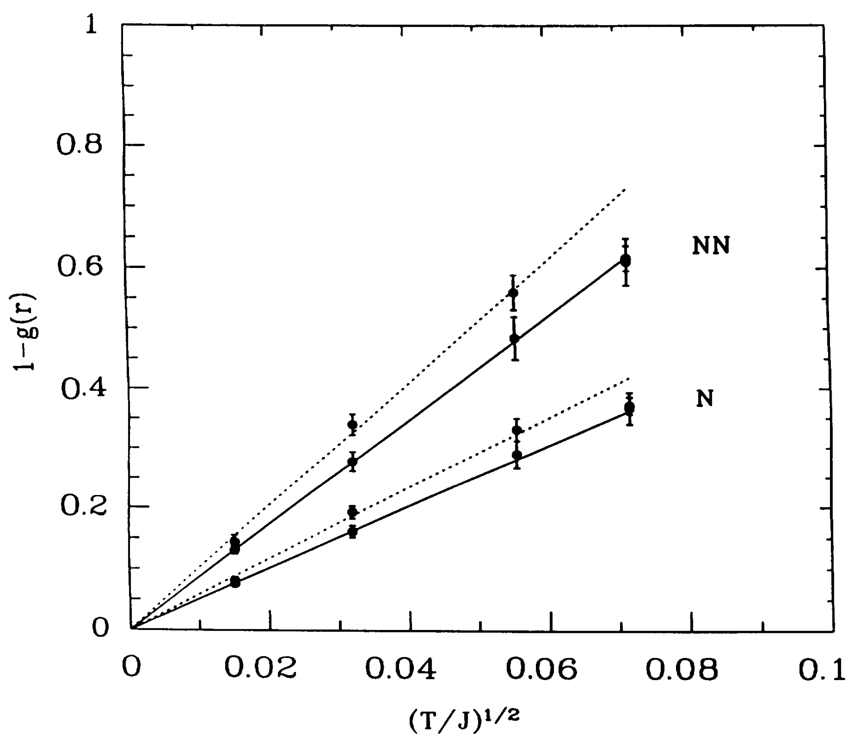
All ground states can be generated by repeated introduction of ‘defects’ into the different parent planar states

Planar ground states have more soft modes
(introduction of 'defect' removes certain soft modes)
Planar ground states are favored at small temperatures

Expect growing correlation of nematic order
(broken spin-rotation symmetry);
Nematic long range order as $T \to 0$

$$g(\mathbf{r}_a - \mathbf{r}_b) = \frac{3}{2}\langle(\mathbf{n}_a \cdot \mathbf{n}_b)^2\rangle - \frac{1}{2};$$

$$\mathbf{n}_a = \frac{2}{3\sqrt{3}}(\mathbf{S}_1 \times \mathbf{S}_2 + \mathbf{S}_2 \times \mathbf{S}_3 + \mathbf{S}_3 \times \mathbf{S}_1)$$



J.T. Chalker, P. C. W. Holdsworth, E. F. Shender, PRL 68, 855 (1992)

D. A. Huse, A. Rutenberg, PRB 45, 7536 (1992)

$g(r) \to 1$ for coplanar ground states

N --- nearest-neighbor triangles

NN --- next-nearest-neighbor triangles

Small distortions from an arbitrary coplanar ground state

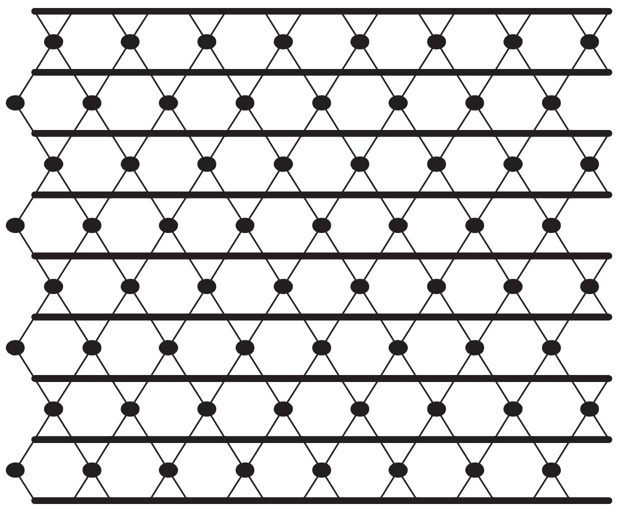
$$H = H_0 + \sum_n H_n(\epsilon^n)$$

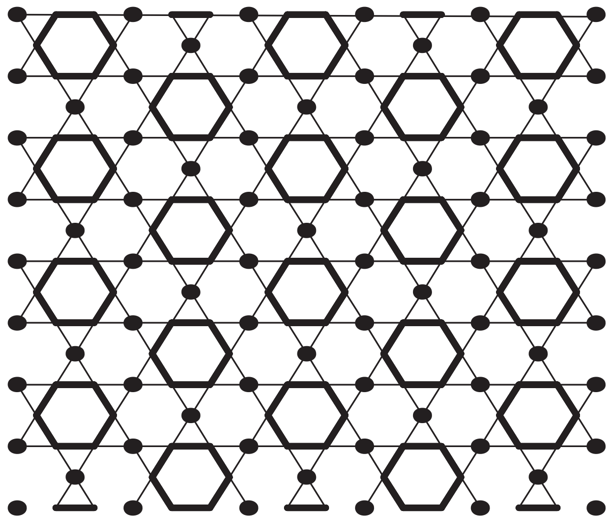
H_2 : quadratic potential; identical for all coplanar states
zero mode (for all q)

$H_3 + H_4$: Quartic potential not the same for different coplanar states

Boltzman weight not the same; $\sqrt{3} \times \sqrt{3}$ favored as $T \to 0$

D.A. Huse, A. Rutenberg, PRB 45, 7536 (1992)   A. Chubukov, PRL 69, 832 (1992)





Quantum Heisenberg Model on Kagome Lattice

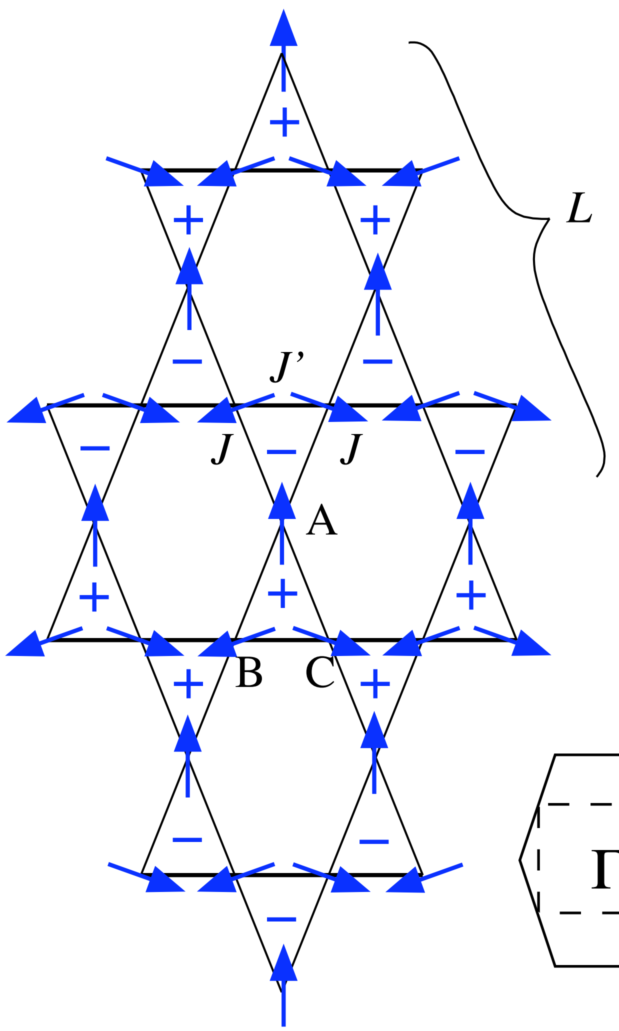
What about spin-1/2 quantum model ?

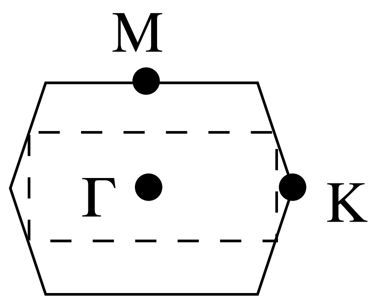
Exact Diagonalization	Singlet Ground State ?
Effective Field Theory	Spin Liquid ?
Series Expansion	Valence Bond Solid ?

Nature of the ground state not understood ...

Distorted Kagome (Volborthite) Lattice

$$J_{\mathrm{AB}} = J_{\mathrm{CA}} \neq J_{\mathrm{BC}}$$





$$H = \sum_{\text{triangles}} (\mathbf{S}_{\mathrm{A}} \cdot \mathbf{S}_{\mathrm{B}} + \alpha \mathbf{S}_{\mathrm{B}} \cdot \mathbf{S}_{\mathrm{C}} + \mathbf{S}_{\mathrm{C}} \cdot \mathbf{S}_{\mathrm{A}})$$

$$= \frac{\alpha}{2} \sum_{\text{triangles}} [(1/\alpha)\mathbf{S}_{\mathrm{A}} + \mathbf{S}_{\mathrm{B}} + \mathbf{S}_{\mathrm{C}}]^2 - \text{constant}$$

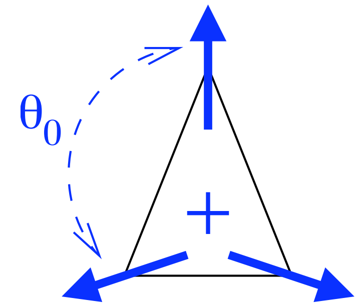
$$J_{\mathrm{AB}} = J_{\mathrm{CA}} = 1 \qquad J_{\mathrm{BC}} = \alpha.$$

Volborthite; $\alpha > 1$

Constraint on classical ground states $(1/\alpha)\mathbf{S}_{\mathrm{A}} + \mathbf{S}_{\mathrm{B}} + \mathbf{S}_{\mathrm{C}} = 0$

Classical Heisenberg Model

Single triangle

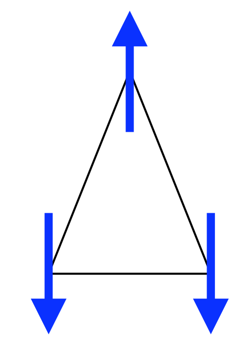


$$\theta_0 = \arccos(-1/2\alpha)$$

(3-state Potts does not apply)

$\alpha \leq 1/2.$ 'cluster spin' cannot be zero

collinear ground state; no degeneracy; ferrimagnet

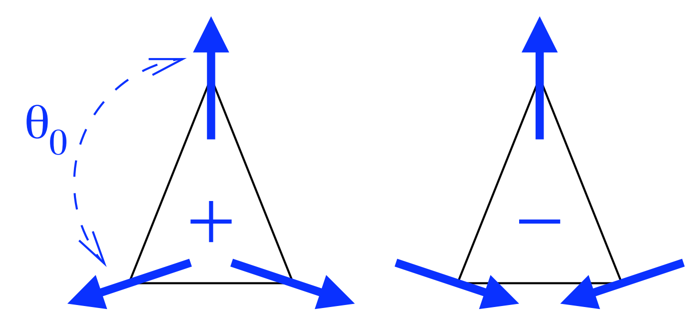


$\alpha \leq 1/2$

$\alpha > 1/2$ coplanar ground states

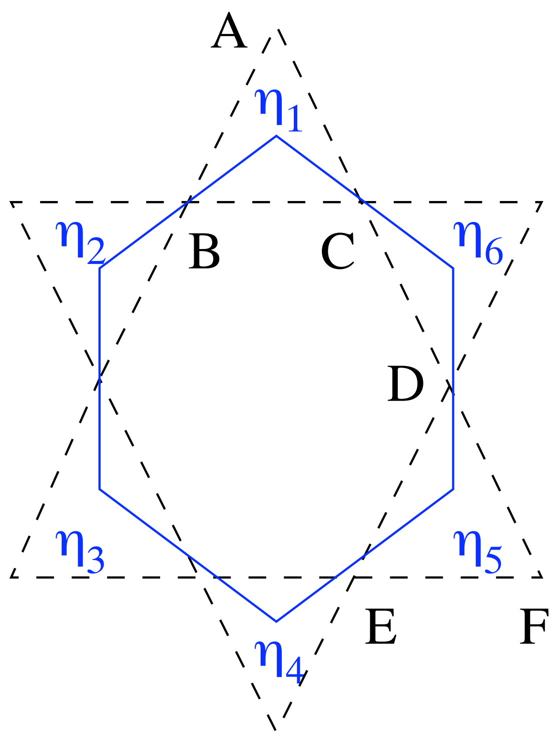
chirality variables are more useful

$$\eta = \pm 1$$



Classical Ground State Degeneracy

Constraints on the chirality variables



Isotropic Kagome

$$\sum_{i=1}^{6} \eta_i = \pm 6 \text{ or } 0.$$

Volborthite Kagome

$$\sum_{i=1}^{6} \eta_i = \pm 6 \text{ or},$$

$$\sum_{i=1}^{6} \eta_i = 0 \text{ and } \eta_1 + \eta_4 = 0.$$

Degeneracy of coplanar ground states

Isotropic Kagome $\exp(0.379N)$

Volborthite Kagome $\exp(2.2L)$ Sub-extensive !

Direct enumeration

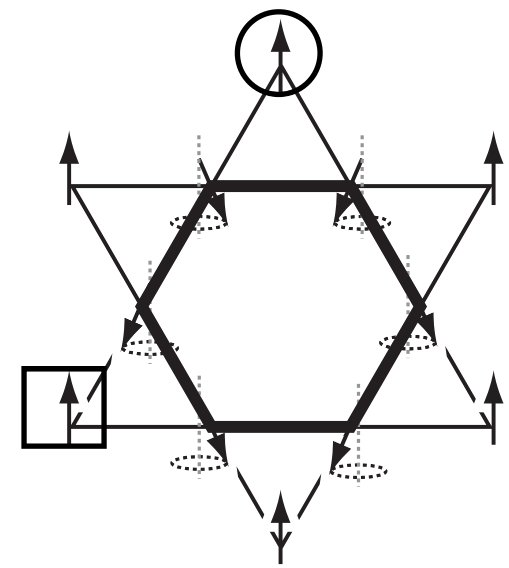
Transfer Matrix Method

Consequence of Sub-extensive Degeneracy

No local weather-vane mode

○ □ not equivalent

cannot be in the same direction



Non-local weather-vane modes exist - the number do not scale as the area of the system

Classical ground state manifold of the Volborthite Kagome is much less connected than the isotropic case

requires moving an infinite number of spins;
large kinetic barriers;
may expect freezing at low temperatures

Application to Volborthtite

Low temperature NMR experiments on Volborthite;
spin freezing below 1.5 K (J/60)
(isotropic Herbertsmithite; no freezing observed)

^{51}V NMR; V atoms at the hexagon centers
$1/T_1$ rises rapidly through the glass transition temperature
two distinct local environments for the ^{51}V
higher static field - 20% lower static field - 80%

F. Bert et al, Phys. Rev. Lett. 95, 087203 (2005)

Assume that the glassy state locally resembles certain classical ground state

Volume average of a local quantity in the glassy state
= ensemble average over classical ground states

$\alpha \approx 1$ case: three different field values are possible

H_{Cu} : the field from a single spin

$H \approx 3H_{cu}$ $\quad$ $H \approx \sqrt{3}H_{Cu}$ $\quad$ $H \approx 0$

$(\sqrt{3} \times \sqrt{3})$

In the experiment, assume $H \approx 3H_{cu}$ for high static field

copper moment per site = $0.4\ \mu_B$

Constraint on the classical ground states (via transfer matrix method) leads to

$H \approx \sqrt{3}H_{Cu}$ - 25% $\qquad$ $H \approx 0$ - 75%

copper moment per site = $\sqrt{3} \times 0.4\ \mu_B = 0.7\ \mu_B$

Summary

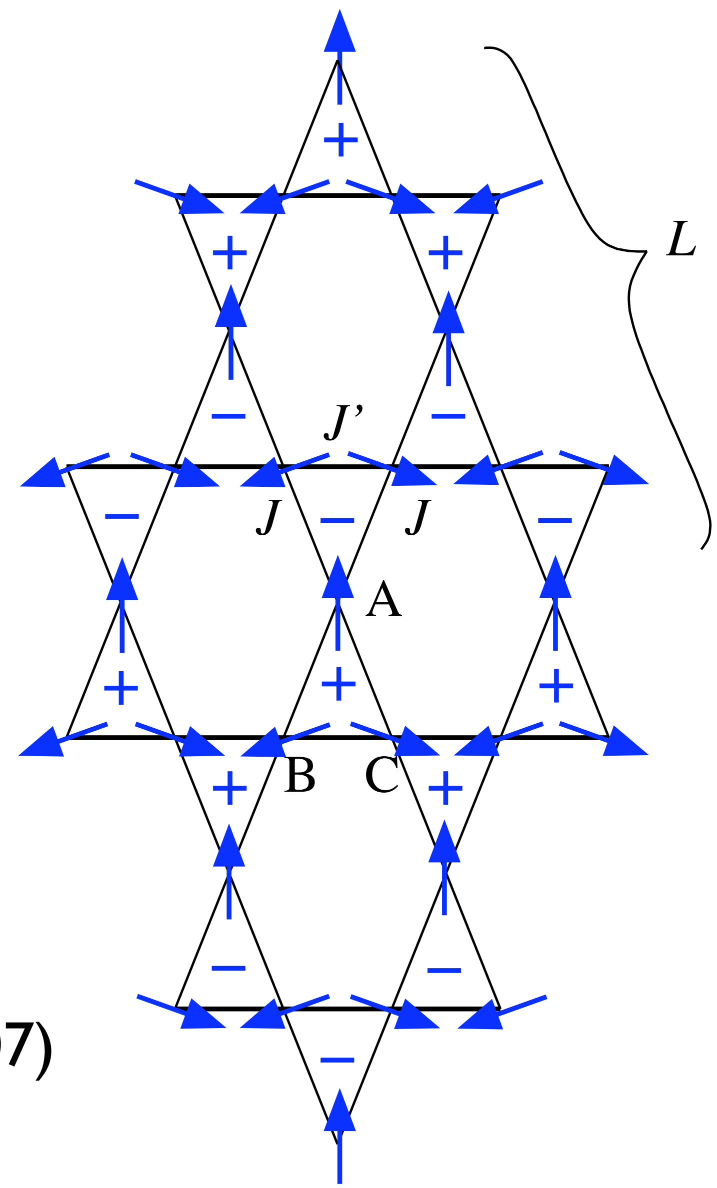
Distorted Kagome Lattice

Classical Heisenberg model:

sub-extensive degeneracy of the classical ground states

much less connected than the isotropic case; glassy behavior ?

thermal fluctuations favor
Chirality Stripe state



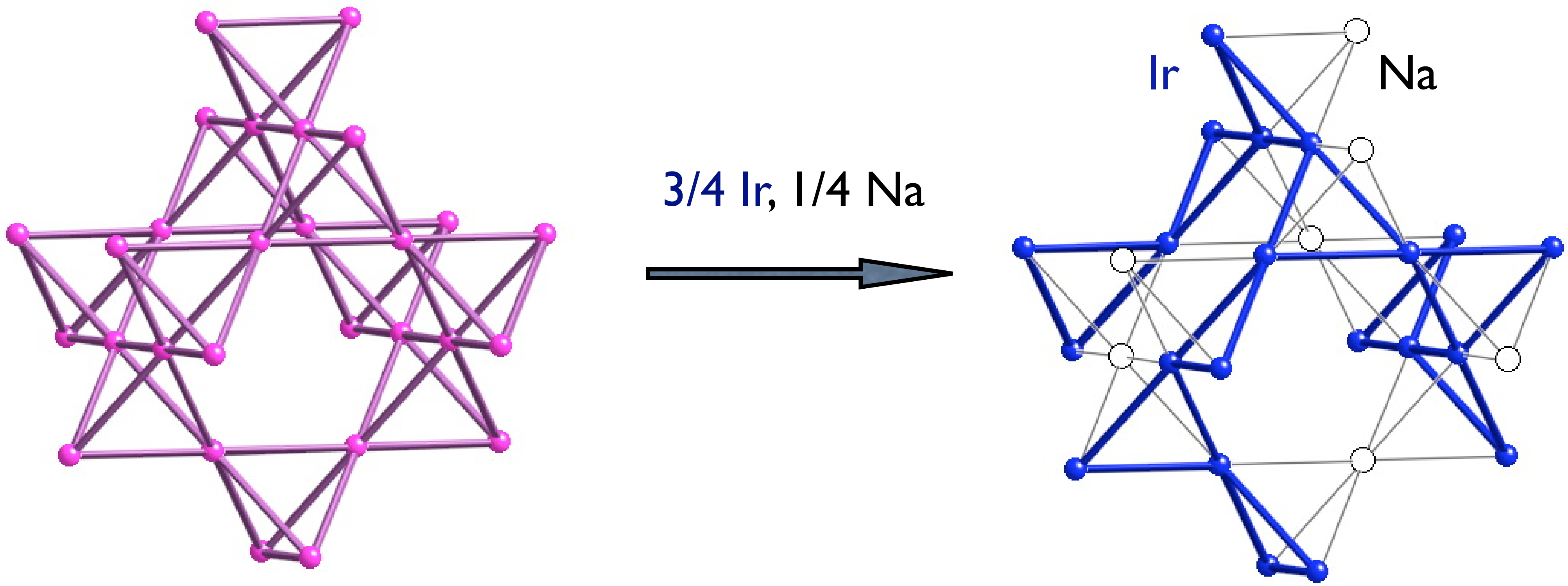
Quantum spin-1/2 Heisenberg model

singlet ground state with a spin gap;
spin liquid ?

F. Wang, A. Vishwanath, Y. B. Kim, Phys. Rev. B 76, 094421 (2007)

Three-dimensional S=1/2 Frustrated Magnet

$Na_4Ir_3O_8$ has a Hyper-Kagome sublattice of Ir ions



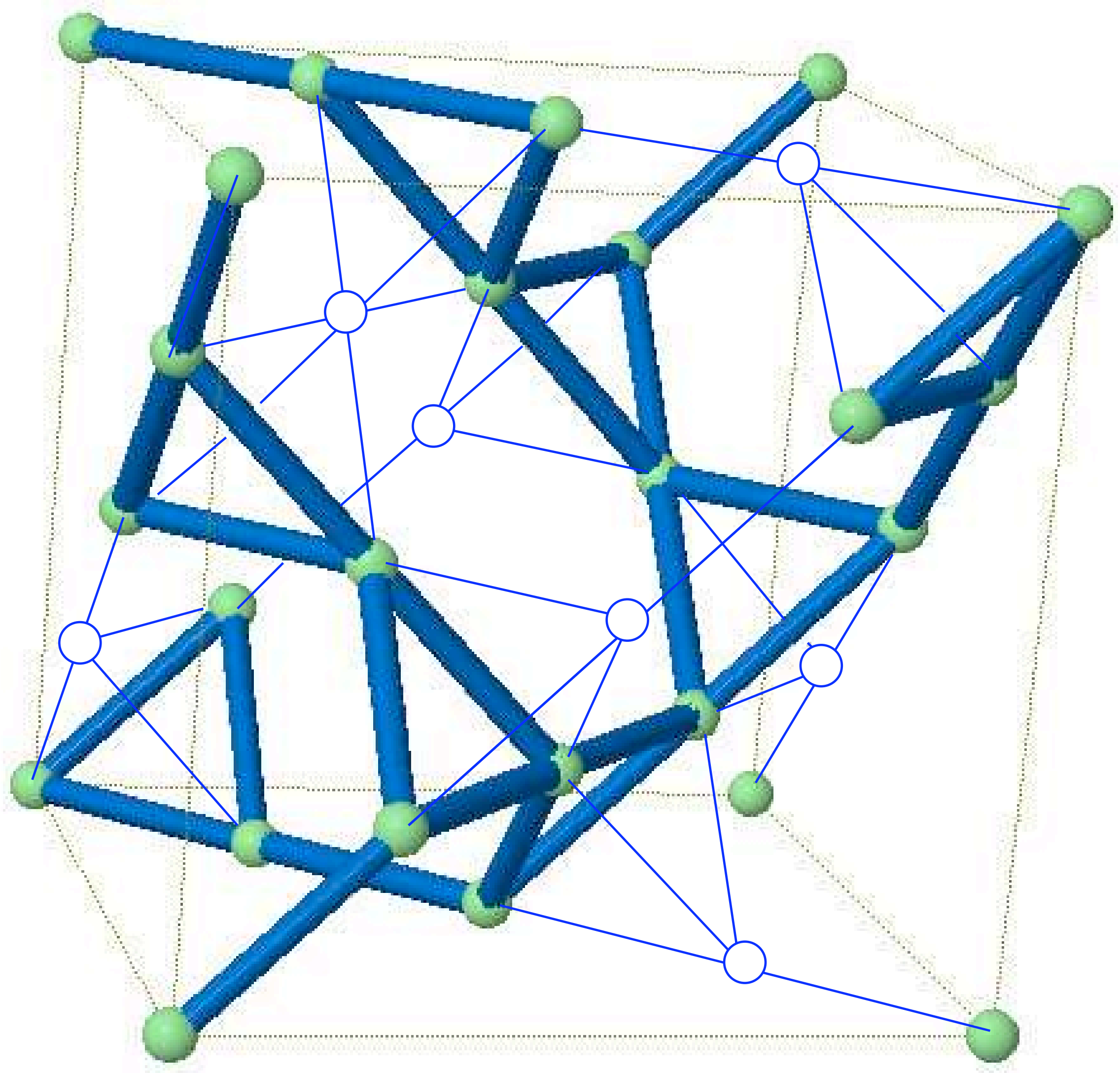
Pyrochlore   Hyper-Kagome

All Ir-Ir bonds are equivalent

Ir^{4+} ($5d^5$) carries S=1/2 moment (low spin state)

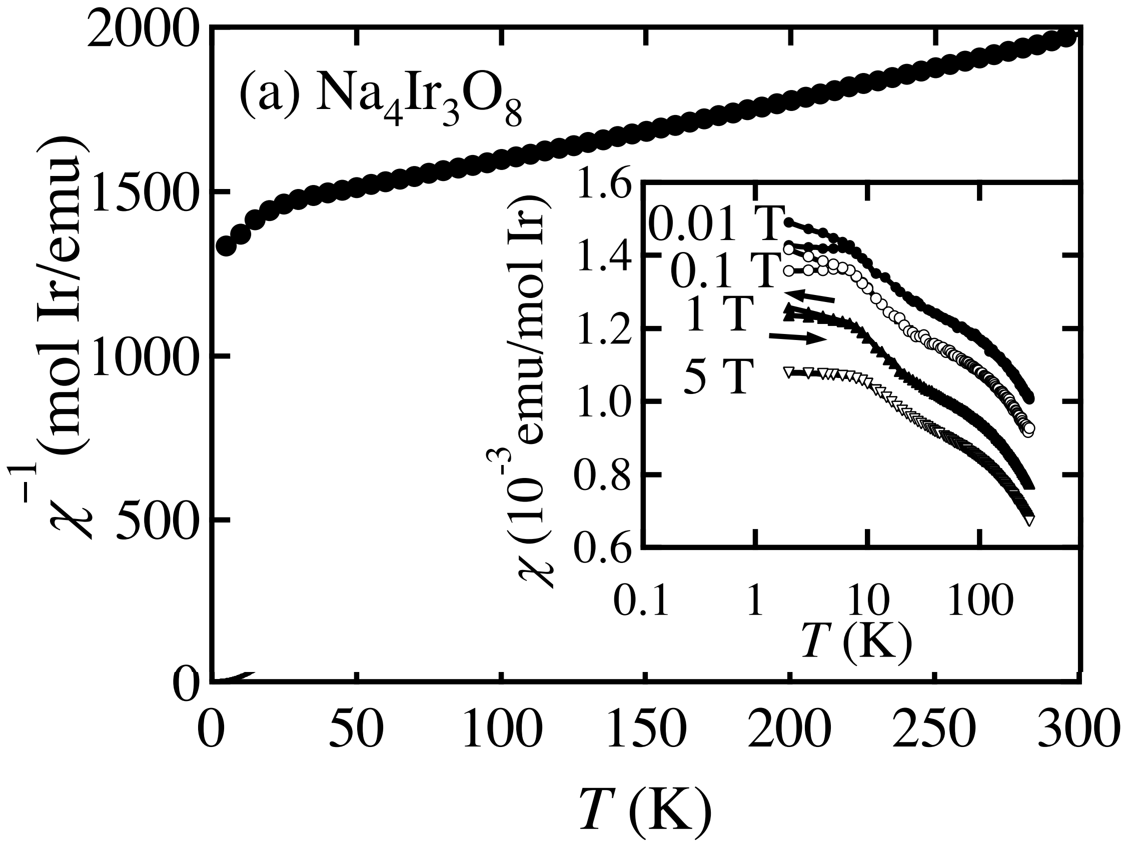
Y. Okamoto, M. Nohara, H. Agura-Katrori, and H. Takagi, arXiv:0705.2821 (2007)

Hyper-Kagome Lattice



Inverse Spin Susceptibility; Strong Spin Frustration

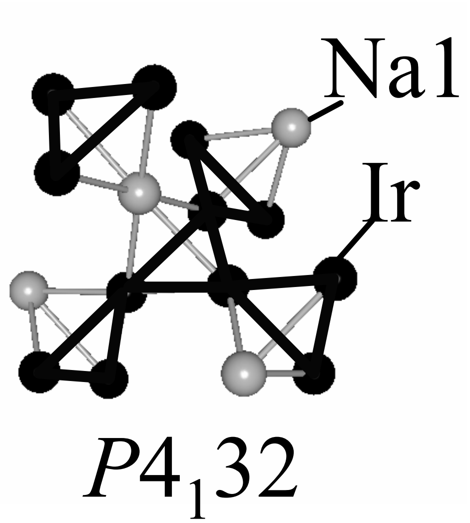
Y. Okamoto, M. Nohara, H. Agura-Katrori, and H. Takagi, arXiv:0705.2821 (2007)



Curie-Weiss fit

$$\Theta_{\rm CW} = -650K$$

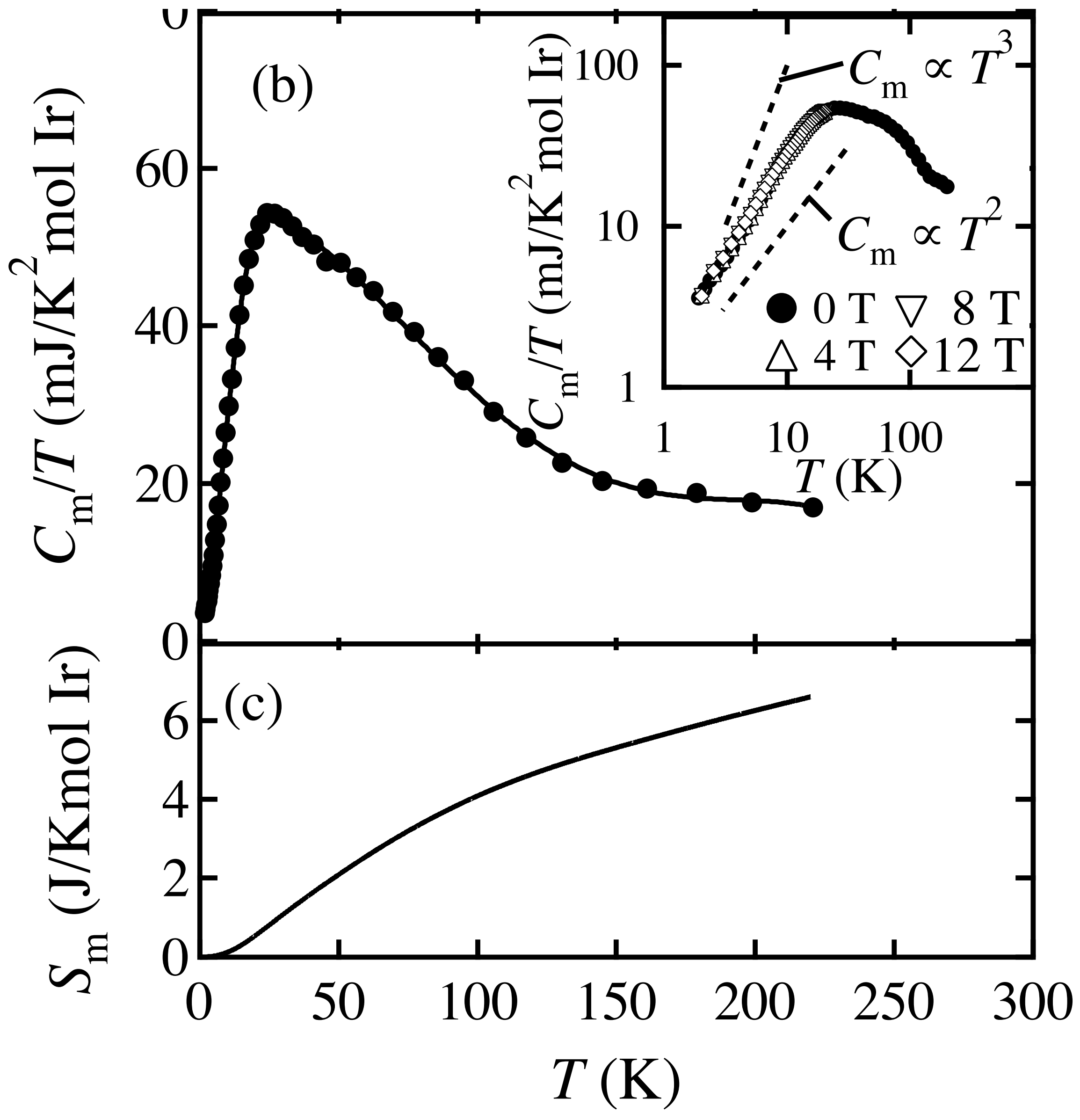
No magnetic ordering down to $|\Theta_{\rm CW}|/300$



Large Window of Cooperative Paramagnet

Specific Heat; Low Energy Excitations ?

Y. Okamoto, M. Nohara, H. Agura-Katrori, and H. Takagi, arXiv:0705.2821 (2007)



No Magnetic Ordering

Gapless Excitations
or Small Gap ?

Field-independent
up to 12T

Is the T=0 Ground State a Spin Liquid ?

Classical Model

Classical Antiferromagnetic O(N) Model

N-component spins with fixed length N

$$H = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j \qquad \mathbf{S}_i = (S_i^1, ..., S_i^N) \qquad \mathbf{S}_i \cdot \mathbf{S}_i = N$$

Large-N limit: $N \rightarrow \infty$

The lowest eigenvalue (4-fold) is independent of wavevector

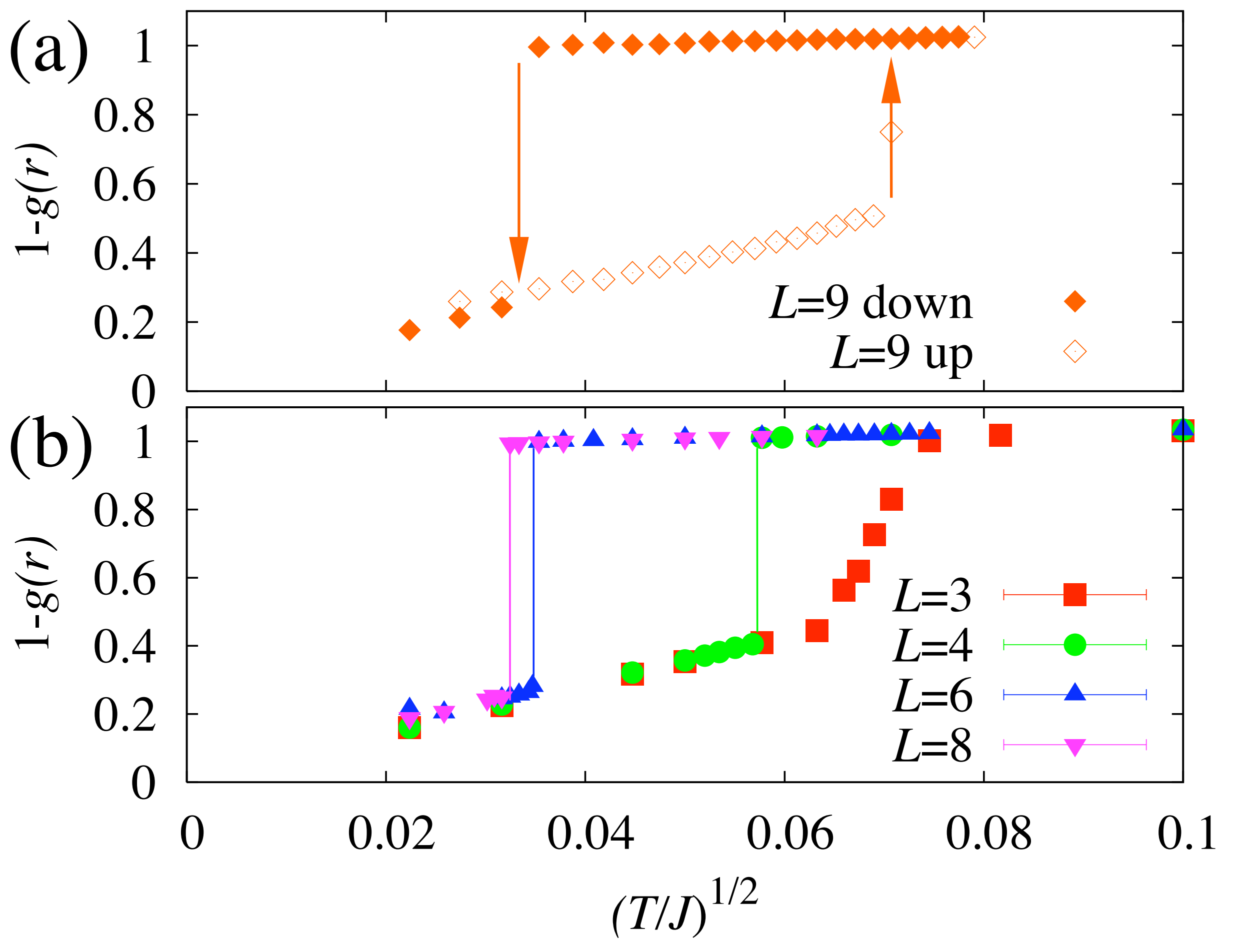
macroscopic degeneracy

Describes physics in the Cooperative Paramagnet regime

Thermal Order by Disorder; Monte Carlo

First order transition to a nematic order $T < 1 \sim 5 \times 10^{-3} J$

L x L x L x 12 lattice (L = 3, 4, 6, 8, 9)



$$g(\mathbf{r}_a - \mathbf{r}_b) = \frac{3}{2}\langle(\mathbf{n}_a \cdot \mathbf{n}_b)^2\rangle - \frac{1}{2};$$

$$\mathbf{n}_a = \frac{2}{3\sqrt{3}}(\mathbf{S}_1 \times \mathbf{S}_2 + \mathbf{S}_2 \times \mathbf{S}_3 + \mathbf{S}_3 \times \mathbf{S}_1)$$

$g(\mathbf{r}) = 1$ ideal coplanar state

$g(\mathbf{r}) = 0$ non-coplanar state

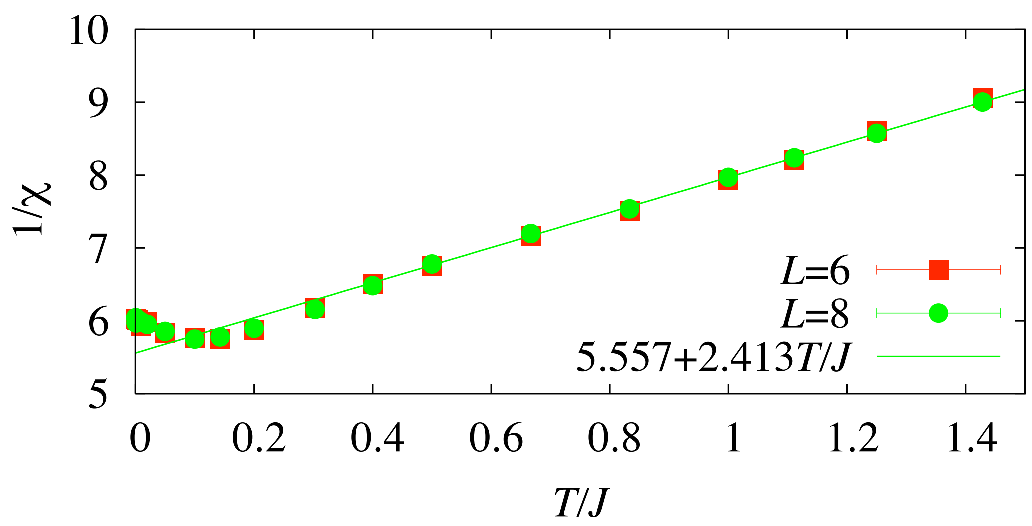
coplanar favored

Thermal Order by Disorder; Monte Carlo

Magnetic Order ?

not found, but cannot reliably be determined below T_n

Comparison with Experiment ?



$$\theta_{CW} = -2.303(5)J$$ Monte Carlo

$$\theta_{\mathrm{CW}}^{\mathrm{exp}} = -650K$$

$$J \approx 280K \qquad T_n \sim 0.3 - 1.4\ K$$

Experimental data exist only down to 2-3 K ...

Cooperative Paramagnetic Regime

$T > J/100 \sim 2 - 3K$

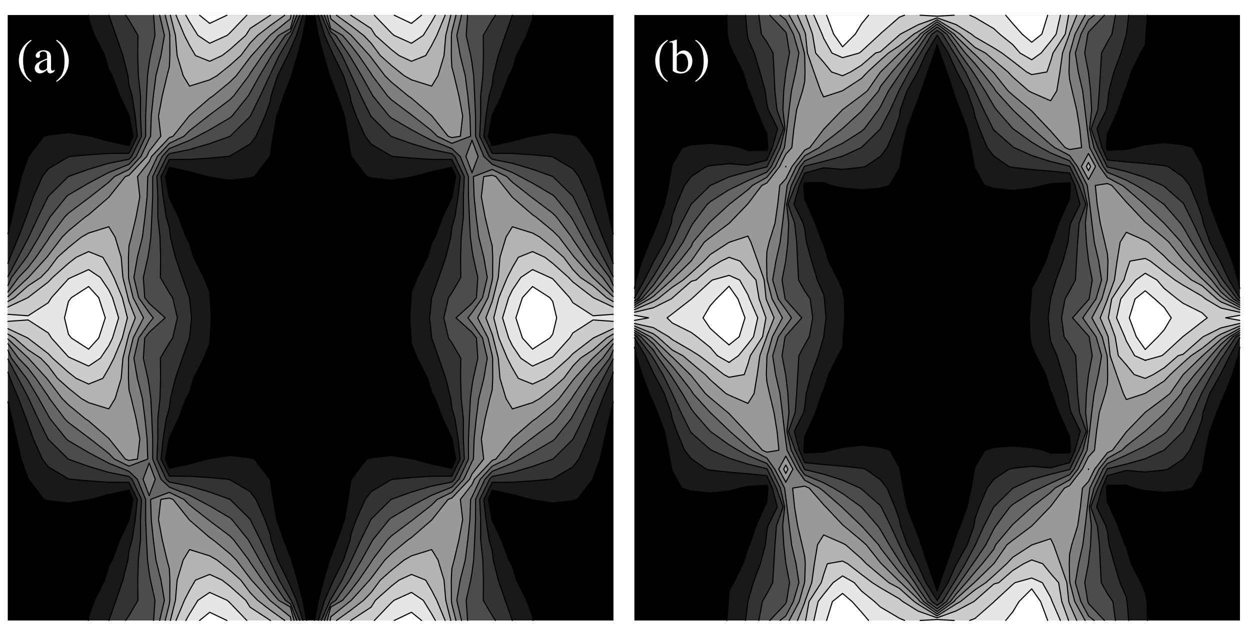
Physics is dominated by
Cooperative Paramagnet behavior

Behavior of the Large-N O(N) model

≈ O(3) Monte Carlo for $T > J/100 \sim 2 - 3K$

Spin Structure Factor in the [hhl] plane $S(\mathbf{q}) = \sum_{\mu\nu} \langle \mathbf{S}_{\mathbf{q},\mu} \cdot \mathbf{S}_{-\mathbf{q},\nu} \rangle$

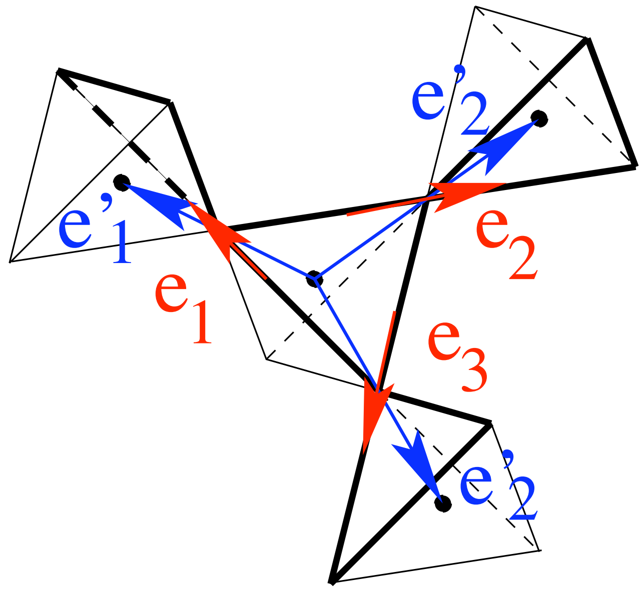
O(N)
large-N



O(3)
Monte Carlo

Dipolar Spin Correlations in the Cooperative Paramagnet Regime

$$\langle S_i^\alpha S_j^\beta \rangle \propto \delta_{\alpha\beta} \left[\frac{3(\mathbf{e}_i \cdot \mathbf{r}_{ij})(\mathbf{e}_j \cdot \mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^5} - \frac{\mathbf{e}_i \cdot \mathbf{e}_j}{|\mathbf{r}_{ij}|^3} \right]$$



c.f. Pyrochlore lattice: S.V. Isakov, K. Gregor, R. Moessner, S. L. Sondhi, PRL 93, 167204 (2004)

Quantum Heisenberg Model

SU(2) Heisenberg Model $\mathbf{S}_i = \frac{1}{2} b_i^{\dagger\alpha} \sigma_\alpha^\beta b_{i\beta}$ $\alpha, \beta = \uparrow, \downarrow$

$$H = -\frac{1}{2} \sum_{ij} J_{ij} (\epsilon_{\alpha\beta} b_i^{\dagger\alpha} b_j^{\dagger\beta})(\epsilon^{\gamma\delta} b_{i\gamma} b_{j\delta}) \qquad n_b = b_i^{\dagger\alpha} b_{i\alpha} = 2S$$

$\epsilon_{\alpha\beta}$ antisymmetic tensor of SU(2)

Quantum Heisenberg Model

SU(2) Heisenberg Model $\mathbf{S}_i = \frac{1}{2} b_i^{\dagger\alpha} \sigma_\alpha^\beta b_{i\beta}$ $\alpha, \beta = \uparrow, \downarrow$

$$H = -\frac{1}{2} \sum_{ij} J_{ij} (\epsilon_{\alpha\beta} b_i^{\dagger\alpha} b_j^{\dagger\beta})(\epsilon^{\gamma\delta} b_{i\gamma} b_{j\delta}) \qquad n_b = b_i^{\dagger\alpha} b_{i\alpha} = 2S$$

$\epsilon_{\alpha\beta}$ antisymmetic tensor of SU(2)

Sp(N) generalized model; N flavors of bosons on each site

$b_{i\alpha} \quad \alpha = 1, ..., 2N$

$$H = -\frac{1}{2N} \sum_{ij} J_{ij} (\mathcal{J}_{\alpha\beta} b_i^{\dagger\alpha} b_j^{\dagger\beta})(\mathcal{J}^{\gamma\delta} b_{i\gamma} b_{j\delta}) \qquad n_b = b_i^{\dagger\alpha} b_{i\alpha} = 2NS$$

$\mathcal{J}^{\alpha\beta} = \mathcal{J}_{\alpha\beta} = -\mathcal{J}_{\beta\alpha}$ $2N \times 2N$ matrix = $\mathrm{blockdiag}[\epsilon, \epsilon, ...]$

N=1 is the physical limit (S = half-integer); Sp(1)=SU(2)

Large-N limit: $N \to \infty$ with fixed $n_b/N = 2S = \kappa$.

Non-perturbative in the coupling constant and S

Mean-field theory for $S = \kappa/2$ well controlled

$\langle Q_{ij} \rangle = \frac{1}{N} \left\langle \mathcal{J}^{\alpha\beta} b^\dagger_{i\alpha} b^\dagger_{j\beta} \right\rangle$ Valence Bond Singlet

$\langle b^\alpha_i \rangle = x^\alpha_i$. Magnetic Order

Large κ Magnetic Order

Small κ Disordered (Spin Liquid, Valence Bond Solid)

Finite-N fluctuations; Compact U(1) gauge theory

Sp(N) model - quantum spin $“S” = \kappa/2$

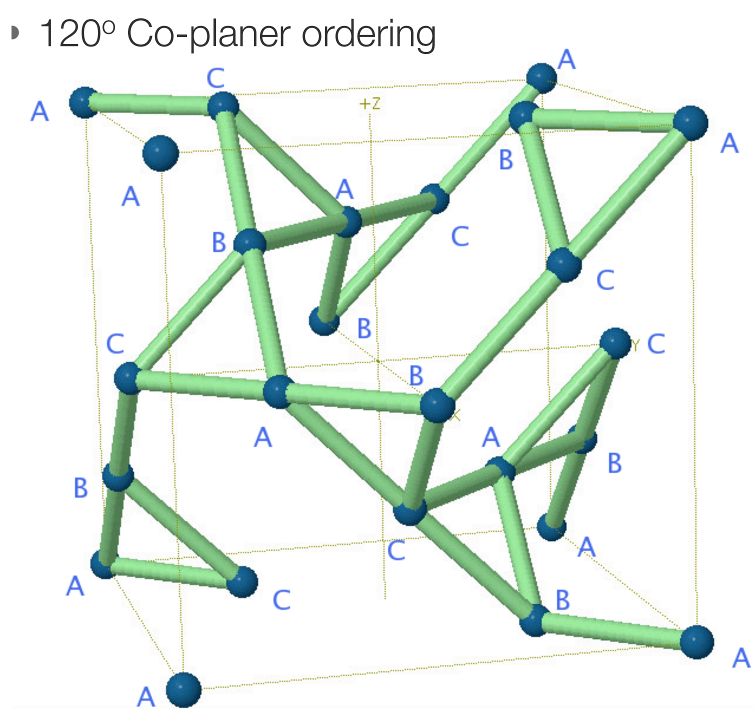
Large-N limit

$\kappa < \kappa_c = 0.4$ Z2 spin liquid (finite spin gap)

8-fold degenerate ground states with p.b.c.

Topological order

$\kappa > \kappa_c = 0.4$ Coplanar magnetically ordered state

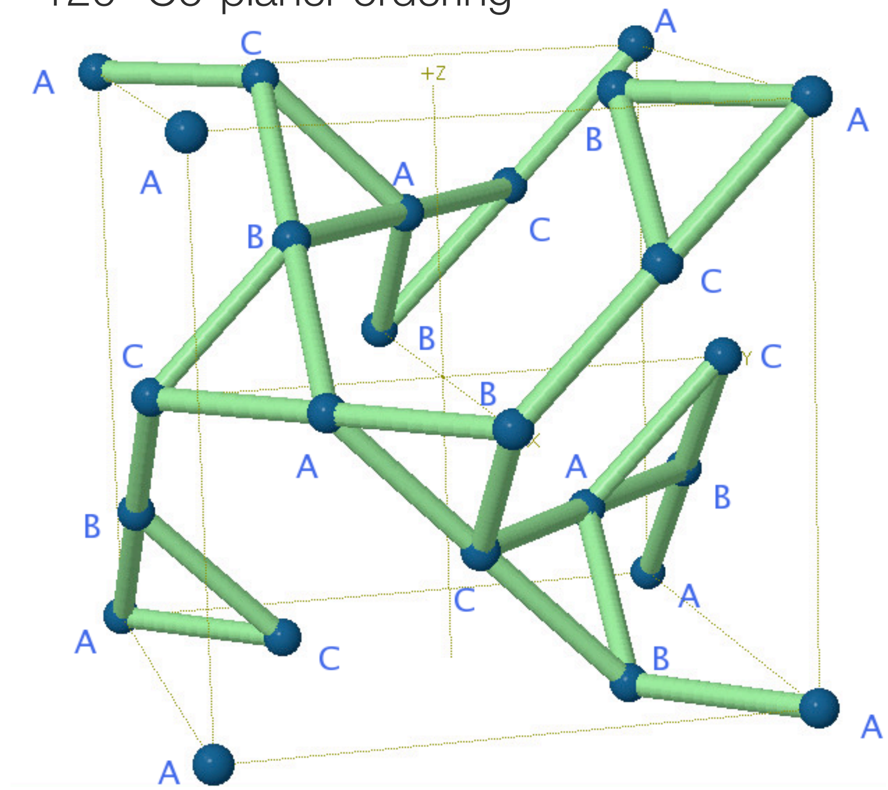


no local weather-vane mode

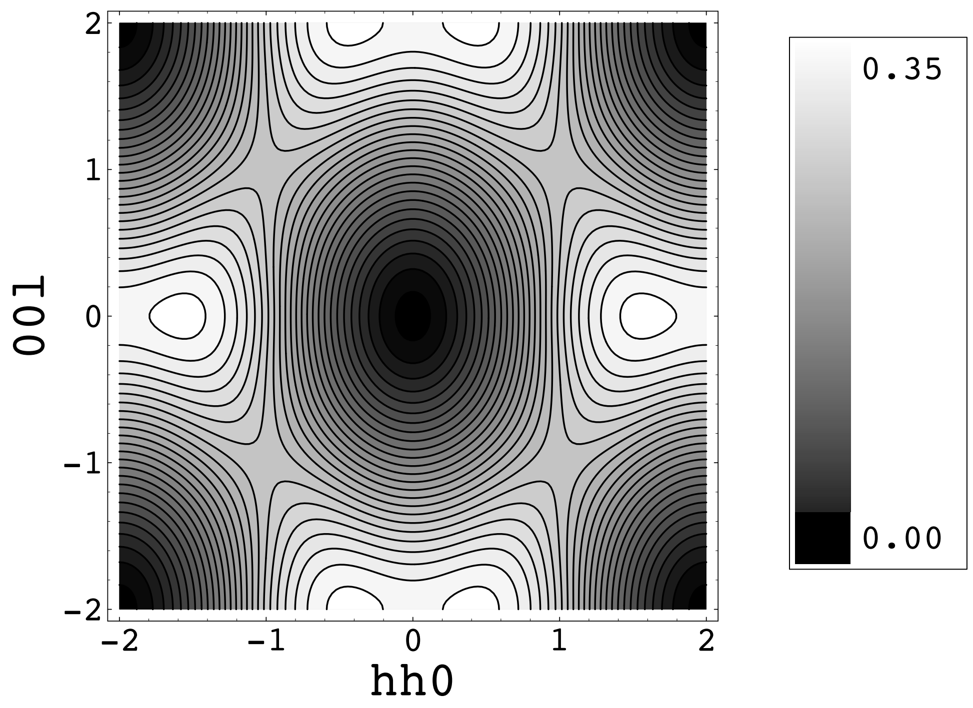
fluctuations can only occur along an infinitely long thread with pattern BCBC...

thermal fluctuations may not select this state

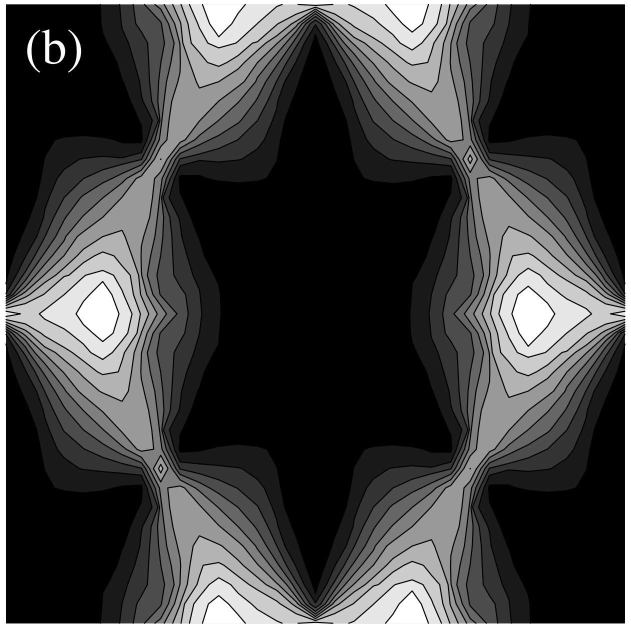
- 120º Co-planer ordering



Spin Structure Factor for Z2 Spin Liquid is very different from that of Cooperative Paramagnet



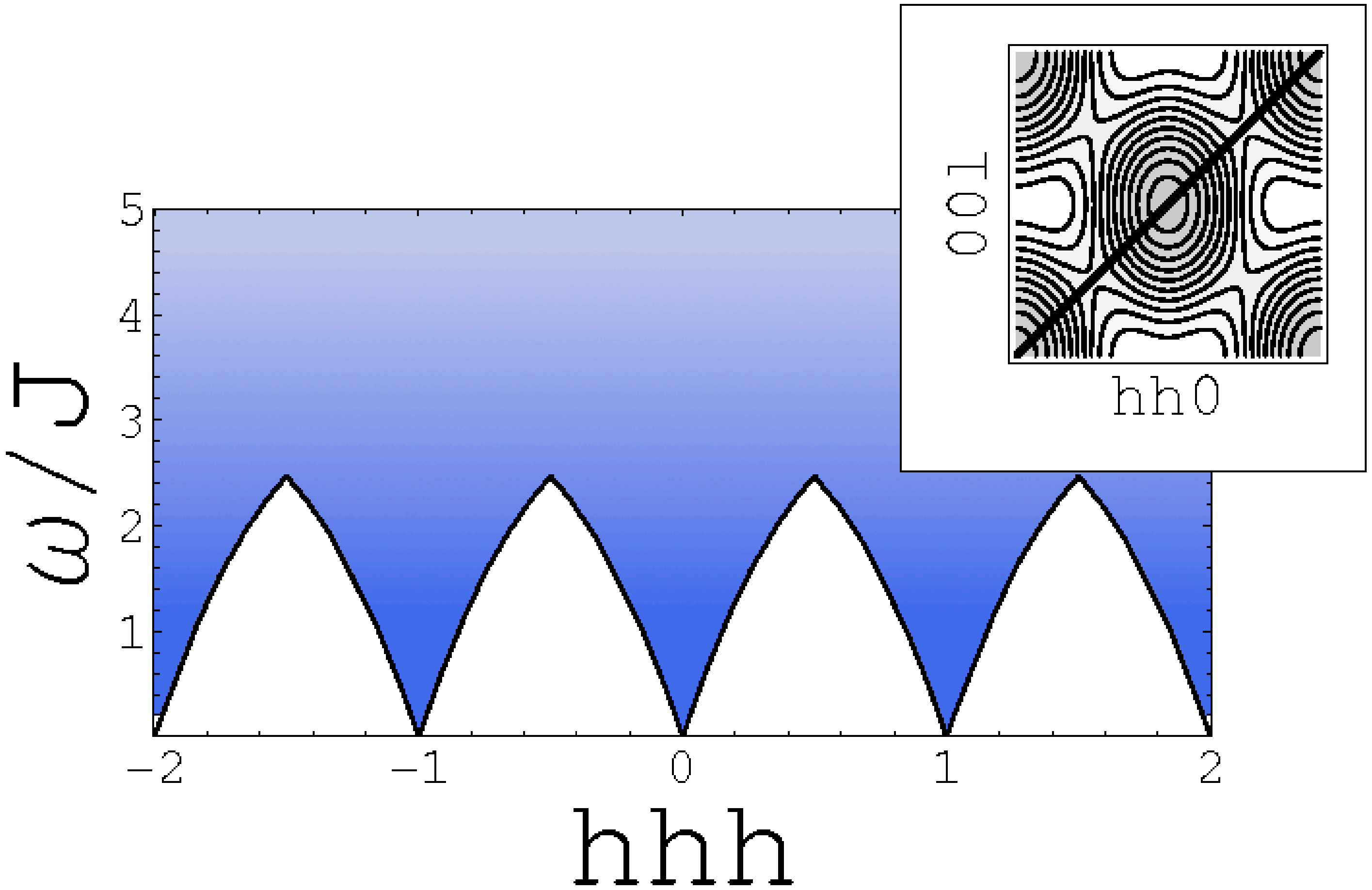
(a)hhl-plane



Quantum Spin Liquid at T=0

Cooperative Paramagnet for T > J/100

Two-Spinon Continuum in the Z2 Spin Liquid



Exact Diagonalization on a unit cell (12, 24 sites)

High temperature results for susceptibility and specific heat with J = 300K compare well with experimental data

Small spin gap ...

Alternative Approaches/Possibilities

Gapless U(1) spin liquid with fermionic spinons

Projected Variational Wavefunction (in progress)

Role of DM interaction ?

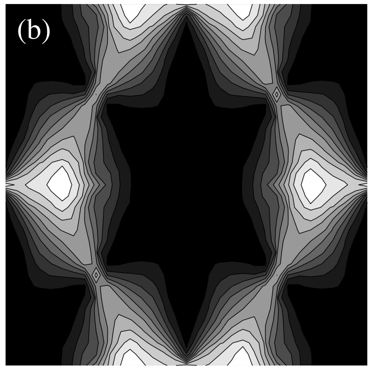
Summary

Spin-1/2 Hyper-Kagome Lattice - Na4Ir3O8 $J \approx 280\ K$

$J/1000 - J/200$ $J/100$

? $0.3 - 1.5K$ $2 - 3K$ $|\Theta_{\mathrm{CW}}| = 650K$

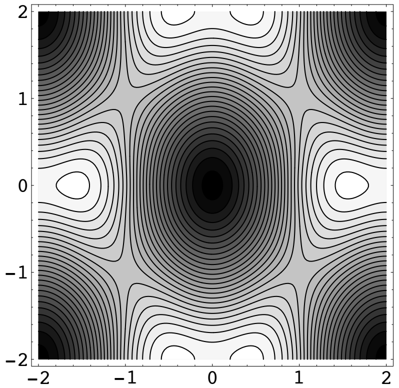
Cooperative Paramagnet



Nematic Order

Thermal order by disorder

Z2 Spin Liquid or Coplanar order



J. M. Hopkinson, S.V. Isakov, H.-Y. Kee, Y. B. Kim, Phys. Rev. Lett. 99, 037201 (2007)
M. J. Lawler, H.-Y. Kee, Y. B. Kim, A. Vishwanath, arXiv:0705.0990

Zn-paratacamite $Zn_xCu_{4-x}(OH)_6Cl_2$

Cu^{2+} spin-1/2 moment

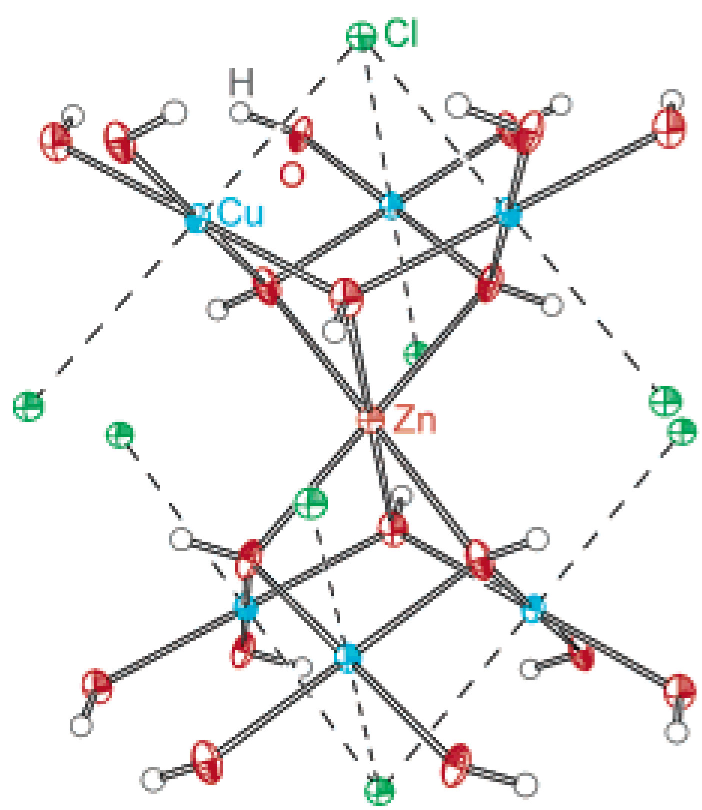
$x < 0.33$ monoclinic

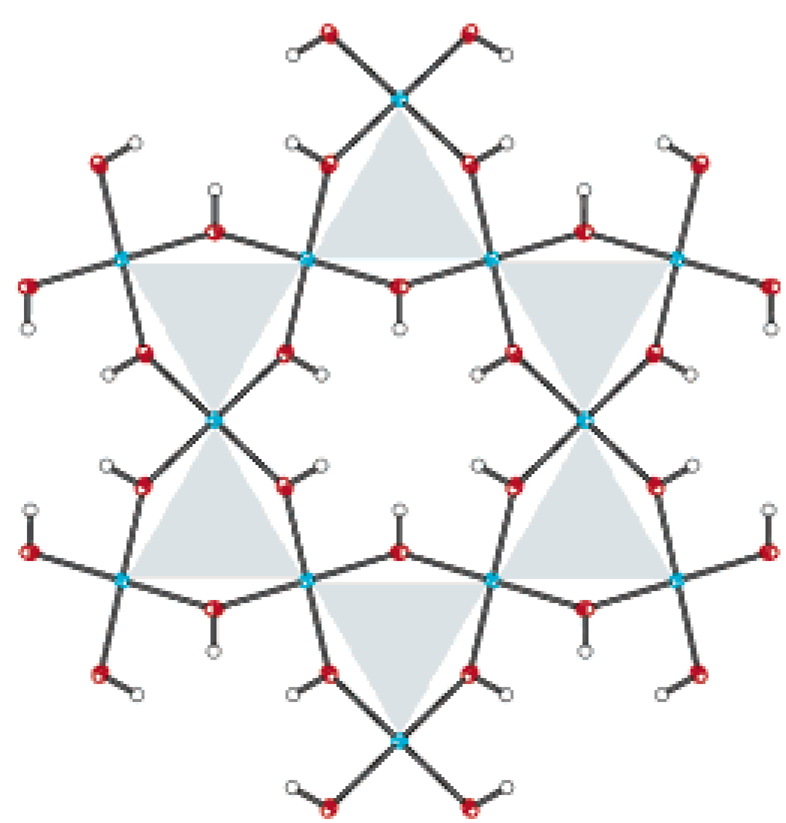
distorted kagome lattices
coupled by triangular sites

$x > 0.33$ rhombohedral

ideal kagome lattice

Zn mostly goes
to triangular sites





herbertsmithite: $x = 1$

$\Theta_{\rm CW} \sim -300\ K$

J. S. Helton et al, arXiv:0610539

no magnetic ordering
or spin freezing
down to $50\ mK$
$(\mu SR, NMR)$

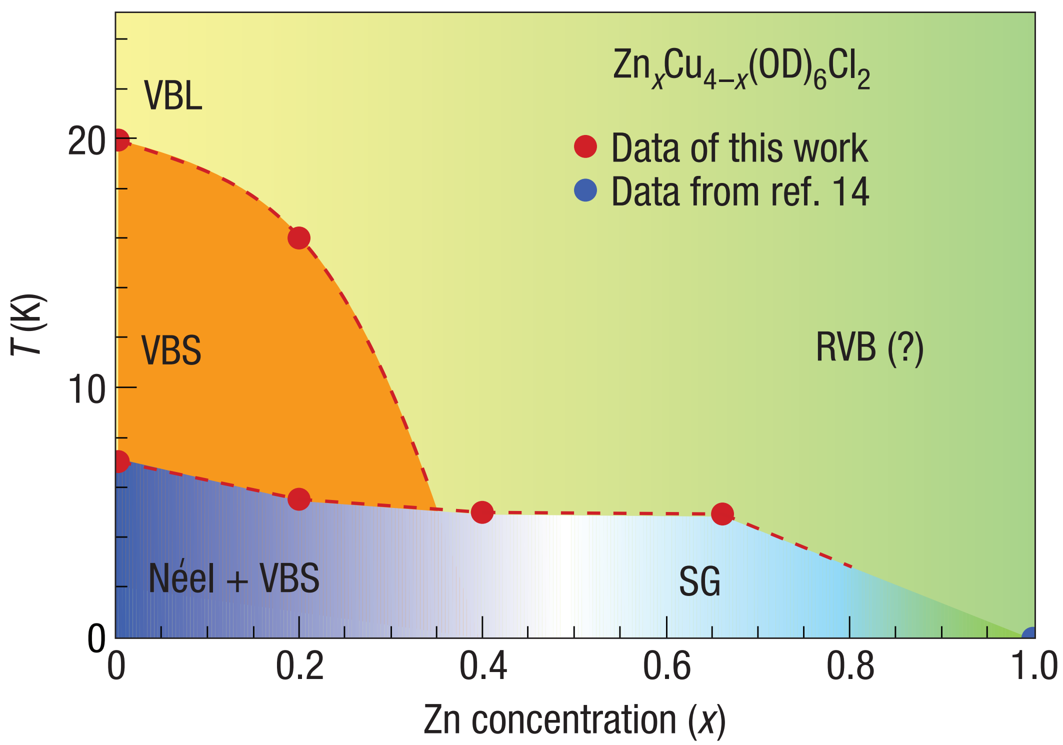
P. Mendels et al, arXiv:0610565
O. Ofer et al, arXiv:0610540

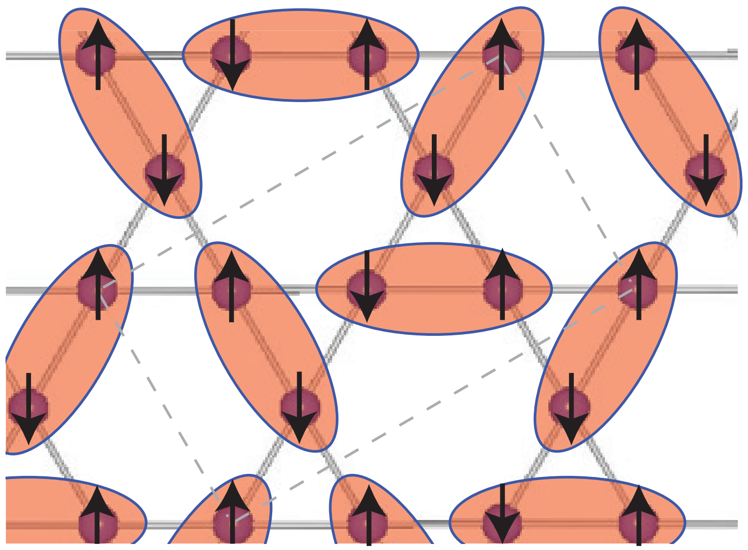
$C_m \sim T^\alpha$
gapless excitations ?

J. S. Helton et al, arXiv:0610539
M. A. de Vries et al, arXiv:0705.0654

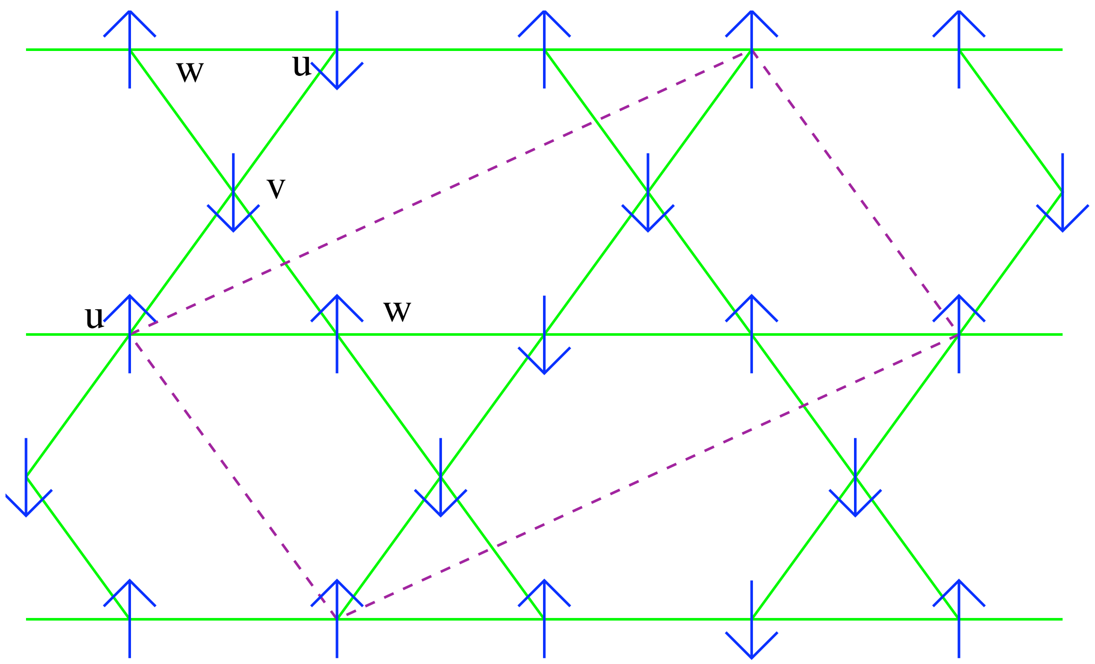
Neutron Scattering: S.-H. Lee et al, Nature Materials, Aug 26, 2007 (arXiv:0705.2279)

Suggested Phase Diagram





Valence Bond Solid



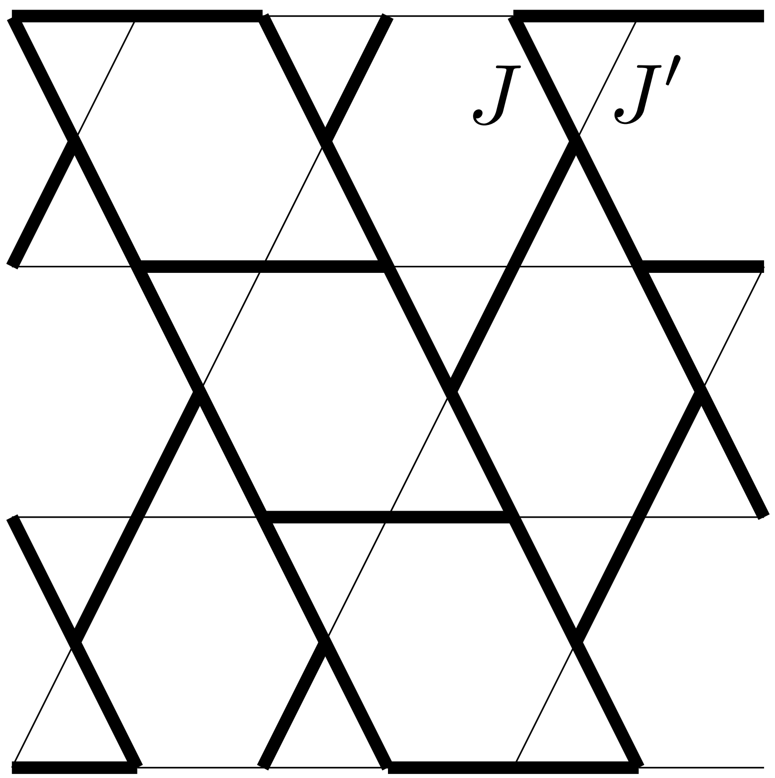
Collinear Magnetic Order

Order pattern (|<S>|=m):

$$m_u = -\bar{m}e^{i(\pi,0)\cdot\mathbf{R}}$$

$$m_v = -\bar{m},\ m_w = \bar{m}$$

Distorted Kagome Lattice



bond length

$3.41\mathring{A}$ $3.42\mathring{A}$

Goodenough-Kanamori rule

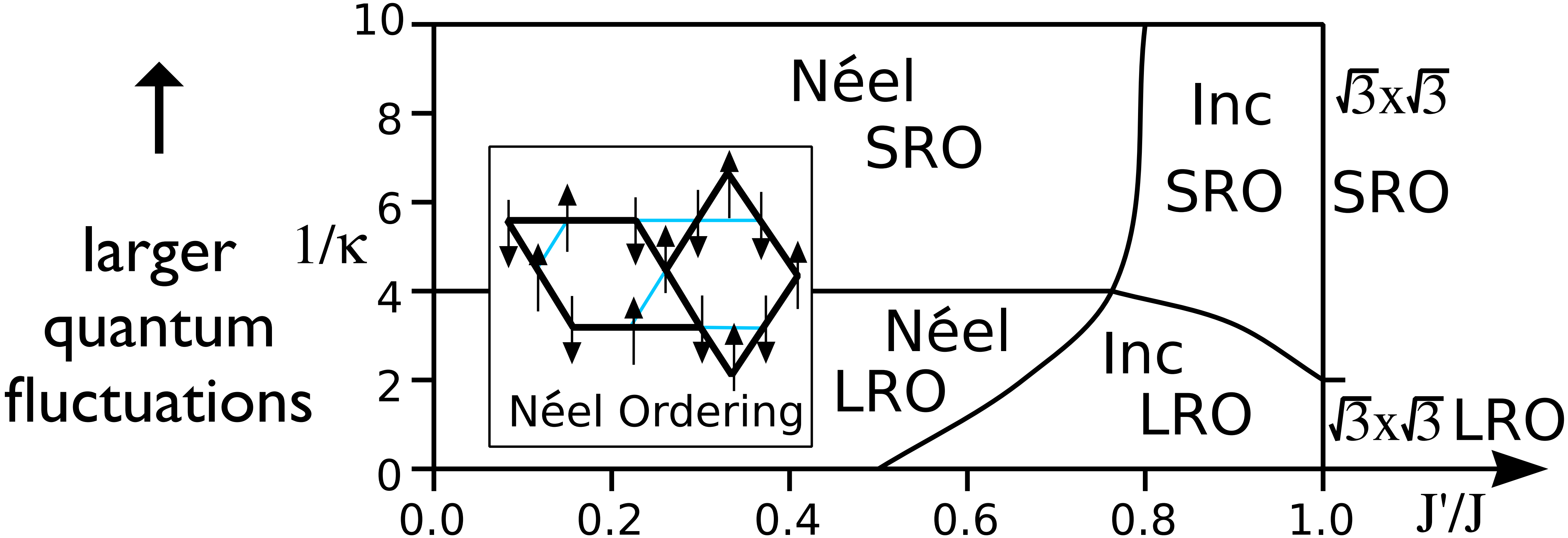
$J'/J \approx 1/3$

Classical O(N) model; Large-N limit

collinear magnetic ordering for $J'/J < 0.5$

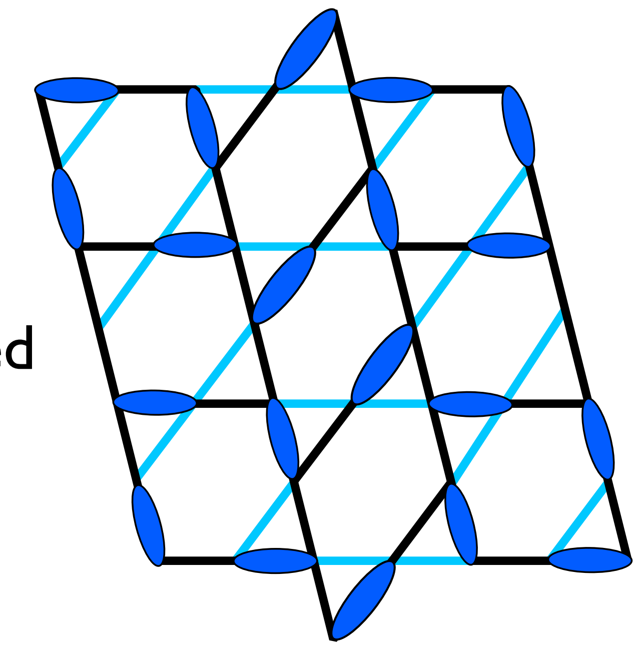
can be stabilized up to $J'/J \sim 1$ with moderate $J_3 < 0$

Quantum Sp(N) model; Large-N limit

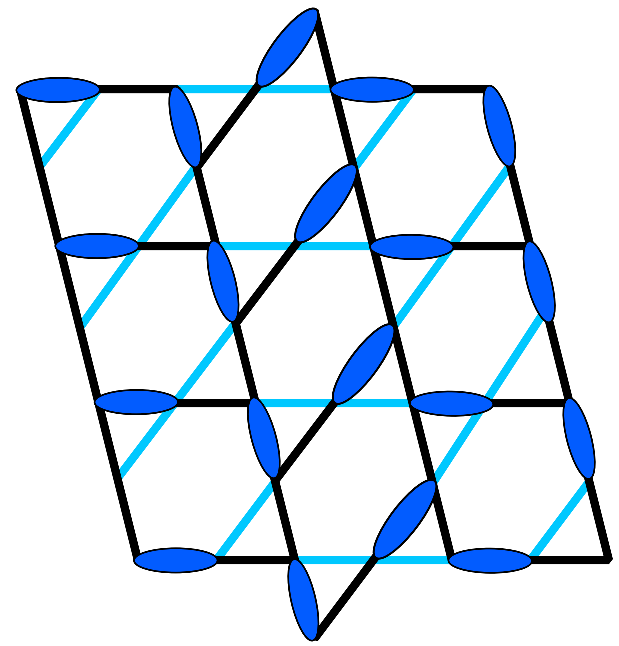


Quantum Sp(N) model; Instanton (via Berry phase) analysis

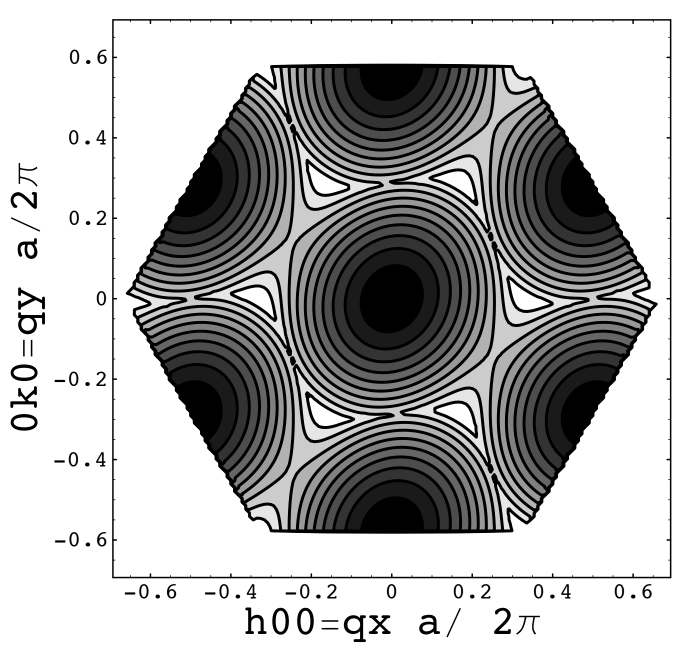
favored



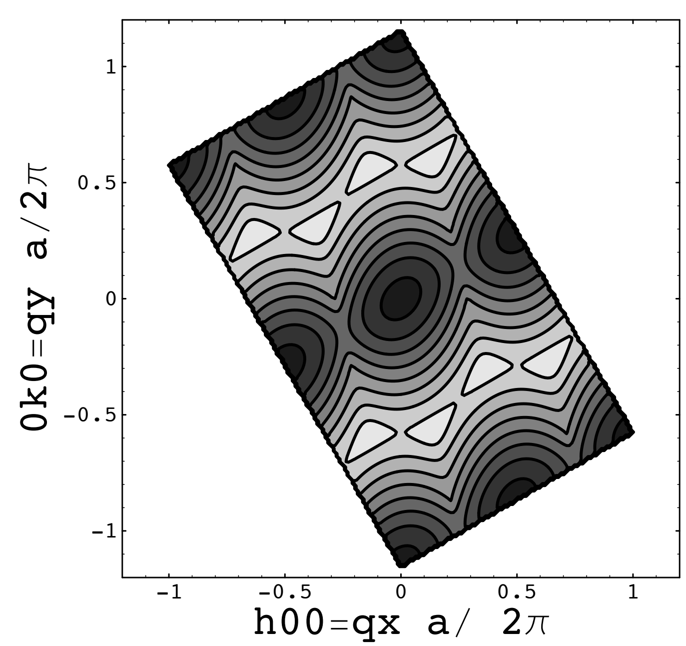
(a)Pin-wheel state



(b)Columnar state



(c)Pin-wheel triplons



(d)Columnar triplons

Lattice Distortion and X-Ray Scattering

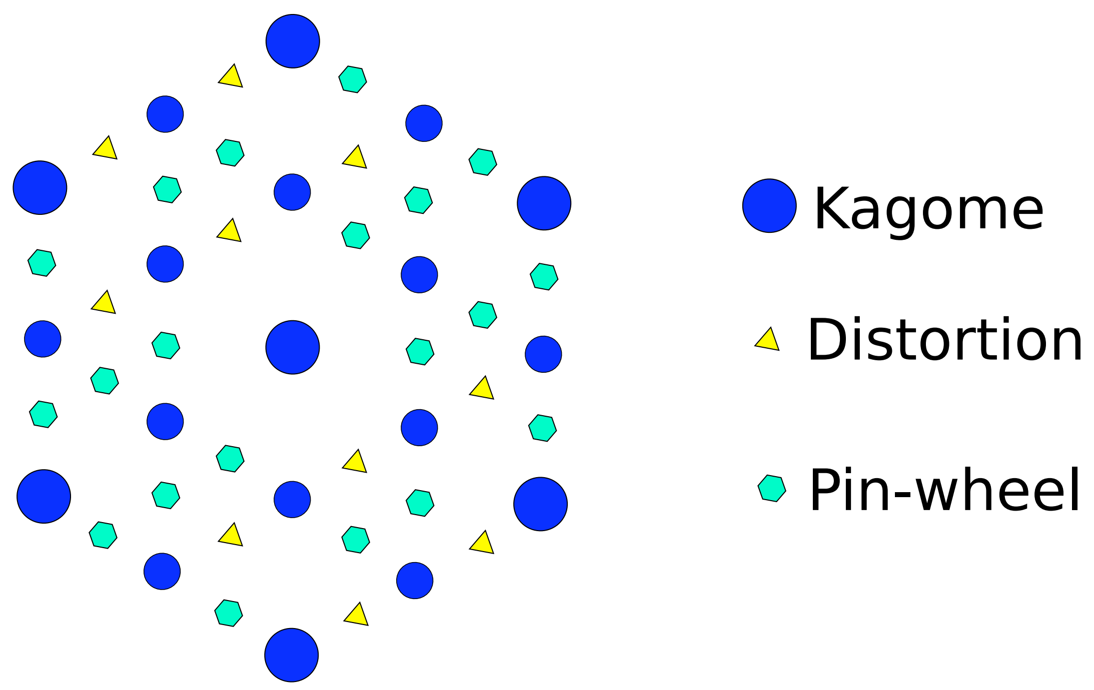
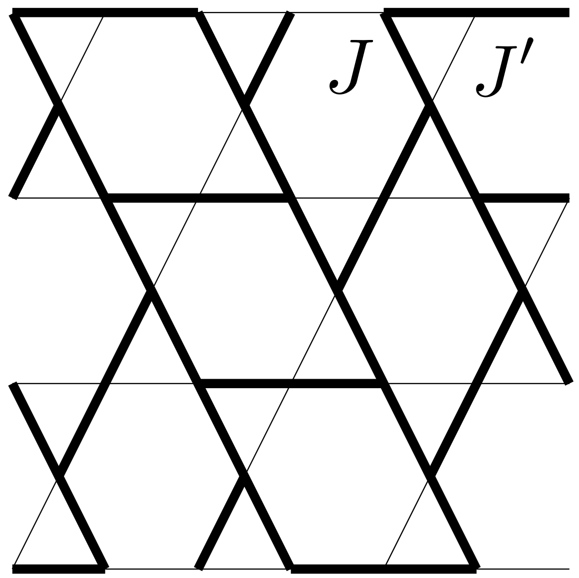


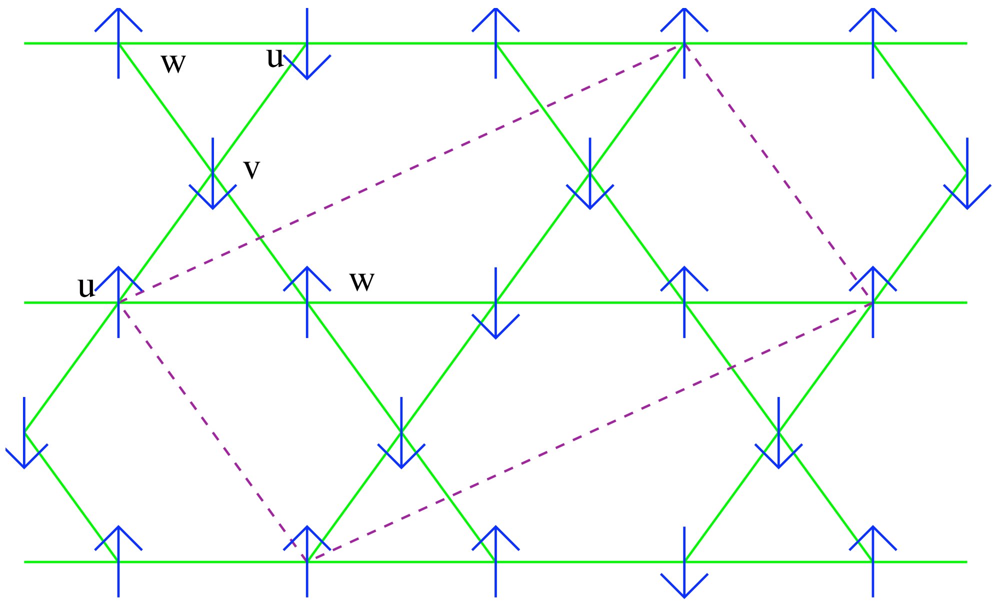
FIG. 4: X-ray structure factor: circles represent Bragg peaks of the ideal kagome lattice; triangles arise from the structural distortion shown in Fig. 1. These are the only Bragg peaks in the columnar state. In the pin-wheel state, additional Bragg peaks (hexagons) appear due to further lattice distortion.

Summary

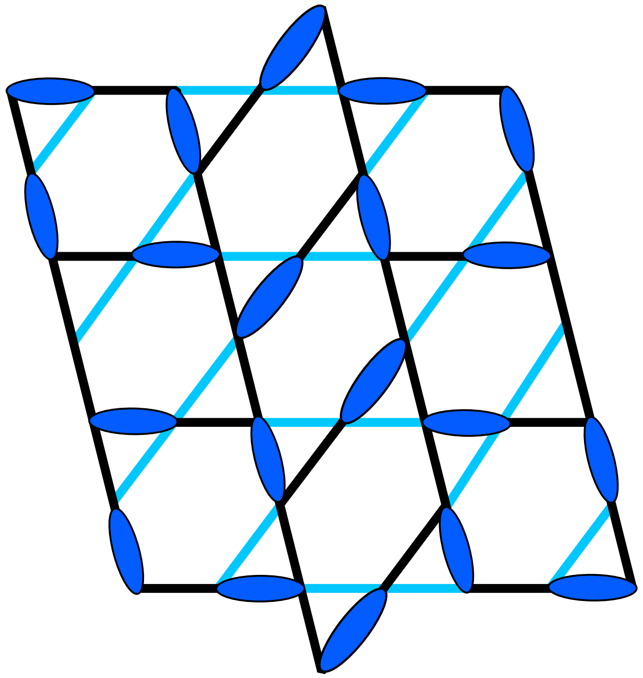
Zn-paratacamite $\mathrm{Zn}_x\mathrm{Cu}_{4-x}(\mathrm{OH})_6\mathrm{Cl}_2 \quad x < 0.33$



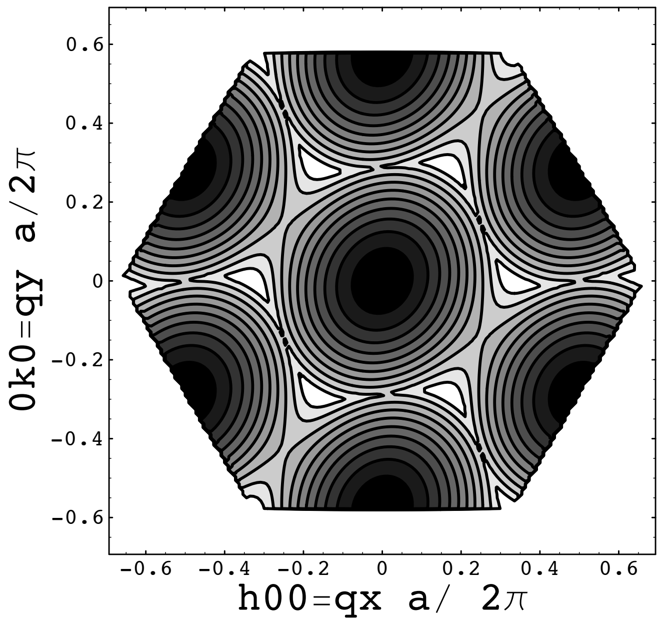
distorted kagome lattice



collinear order



VBS phase



triplon dispersion

M. J. Lawler, L. Fritz, Y. B. Kim, S. Sachdev, arXiv:0709.4489 (2007)