Frustrated quantum magnetism

Emergent gauge fields, fractional moments, critical phases...

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SINP Colloquium July 31 2019

Electron waves in crystals

Band theory

- Band-structure $\epsilon_\mu(\mathbf{k})$: Eigenstates in periodic crystal potential.
- Pauli principle and Fermi distribution: e^- occupy $|\epsilon_\mu(\mathbf{k})\rangle$ with $\epsilon_\mu(\mathbf{k}) < \epsilon_F$
- Insulators and metals: Completely filled last band ☞ Insulator. Partially filled last band ☞ metal

Landau Fermi liquid theory

- What about e^-—e^- interactions? Landau: low energy 'quasiparticles' with charge e^- and spin $1/2$

  No qualitative change from band picture

Electron particles in crystals

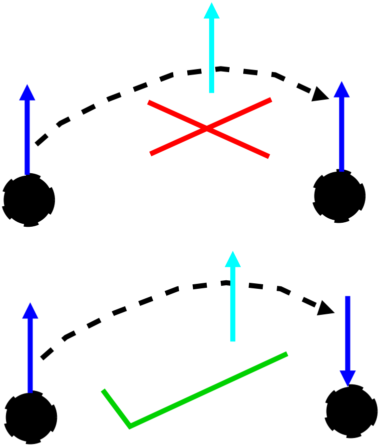
Mott insulators

- Band theory: metal with a half-filled conduction band.
- But: $e^- — e^-$ interactions dominate.
- Charge frozen (gapped) $\rightarrow$ Insulator

Local moments in Motterials

- Interactions localize 1 electron in each ‘conduction band’ orbital
- Low energy physics: Spin of localized electrons
- *Virtual* hopping of charge $\rightarrow$ Low-energy effective Hamiltonian

Recap: Antiferromagnetic exchange



Generalities on exchange interactions

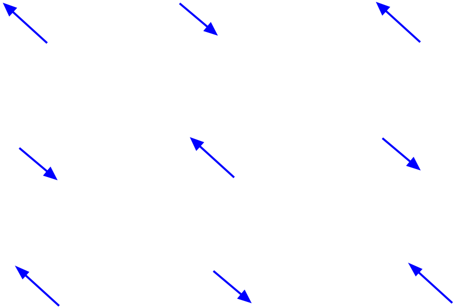
A Goodenough description

- Without spin-orbit: Isotropic exchange interactions.
  $E = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j \quad J > 0$
  When is $J > 0$, large?
  Are nearest neighbour interactions dominant?
  Difficult (quauntum chemistry) questions
  Thumb-rule answers: Goodenough-Kanamori-Anderson rules
  J.B. Goodenough, *Magnetism and the Chemical Bond (1963)*

Complications

- Spin-orbit coupling λ
- Orbital degeneracy
  Interplay between orbital structure and spin physics

Néel order

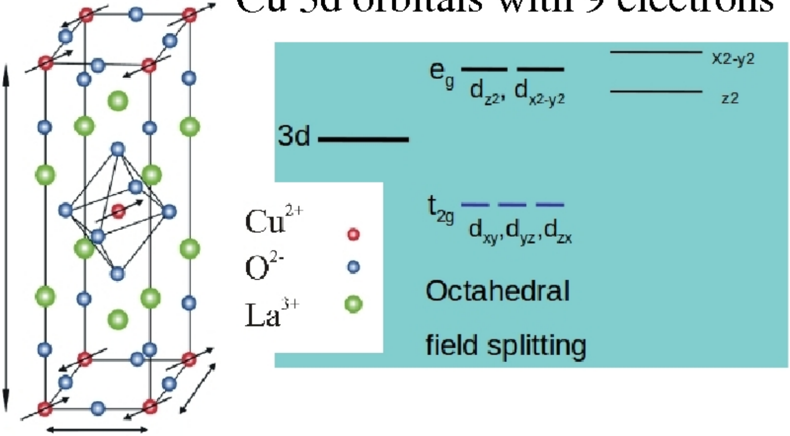


- Bipartite lattice and nearest neighbour $J > 0$
  Néel order: $\mathbf{n}$ (spontaneously chosen) and $\langle \vec{S}_{\vec{r}} \rangle = (-1)^{\vec{r}} \mathbf{n}$

A (famous) example

Cu-O planes in high T_c parent compound: La_2CuO_4

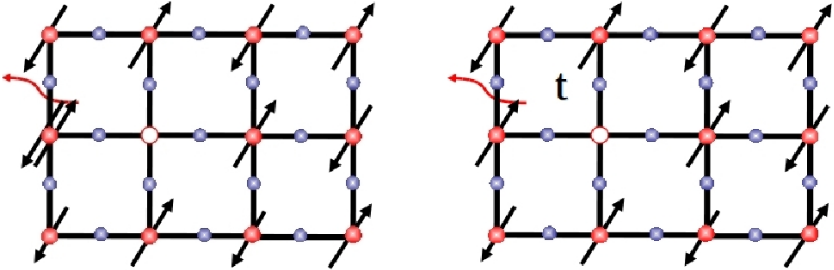
Cu 3d orbitals with 9 electrons

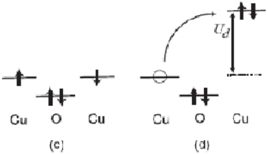


Strong e^{-}-e^{-} interactions

Cu-O planes with strong interactions

Fermi liquid prediction: n = 1+x (holes)





CuO_2 plane with doped holes:

$La^{3+} \rightarrow Sr^{2+}$: $La_{2-x}Sr_xCuO_4$

Doped Mott Insulator: n = x (holes)

Effective Hamiltonian for Cu-O plane

Effective Hamiltonian for Cu-O plane

- Written in terms of operators $c^{\dagger}_{j\sigma}$ that create a hole in the Cu $d_{x^2-y^2}$ orbital:
  $H = -t_{eff} \sum_{\langle ij \rangle \sigma} (c^{\dagger}_{i\sigma} c_{j\sigma} + h.c.) + U \sum_i n_{i\uparrow} n_{i\downarrow}$

Large U limit at $x = 0$: $S = 1/2$ spin Hamiltonian

- Second order perturbation theory at $x = 0$ gives:
  $H_{spin} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$ with $J = 4t^2/U$.

Antiferromagnetism at small x

Antiferromagnetic long-range order at $x = 0$

Classical ground state: Collinear, antiparallel neighbours

Neel order breaks global $SU(2)$ symmetry: Axis **n** chosen

Exact for large spin length S

Numerical evidence for $S = 1/2$:

Stable to quantum mechanical fluctuations on square lattice

(Large-S expansion qualitatively correct even at $S = 1/2$)

The $t - J$ model for hole motion

$H_{tJ} =$

Hole motion scrambles up antiferromagnetic background

At small x, unusual correlated metal with antiferromagnetic order

'Small' Fermi surface: Area $\propto x$.

For $x > x_c$, becomes a high-temperature superconductor(!)

Quasiparticle fractionalization?

Spin-charge separation in one-dimensional metals

- e^- breaks up into spin and charge carrying parts
  Spin and charge move with different velocities.
- No sharp quasiparticle peak in spectral function $A(\mathbf{k}, \omega)$
  $A(\mathbf{k}, \omega)$: Probability of finding electron occupying state with momentum $\hbar\mathbf{k}$ & energy $\epsilon = \epsilon_F - \hbar\omega$

Speculation in cuprate superconductors

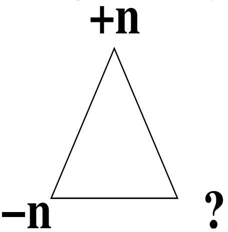
- Does this happen to holes doped into Mott insulator?
  No sharp peaks in spectral function $A(k, \omega)$ for small x(?)
- Materials with quasiparticle fractionalization?
  'Emergent' excitations: 'fractions' of elementary constituents

The story so far . . .

- Breakdown of band-theory
- Mott insulators with low energy spin degrees of freedom
- Antiferromagnetic exchange interactions between spins
- Neel ordered antiferromagnets on bipartite lattices
- Doped Mott insulators: Unusual, correlated metals.

Geometric frustration of exchange interactions

Triangles on my mind...

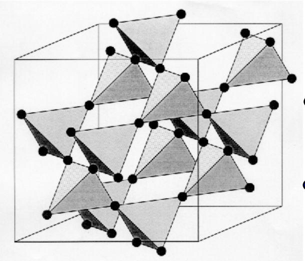


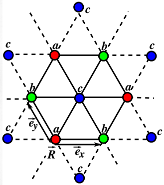
- Triangles *frustrate* Néel order
- Geometry ☞ *competition* between leading exchange interactions

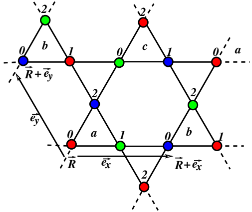
Frustration spawns novel states

- Quenching of leading *J* ☞ *J cannot pick ground state at classical level*
- Sub-dominant interactions & quantum fluctuations ☞ *Variety of novel low temperature states*

Lattices with triangles...







Pyrochlore (Bramwell & Harris); Triangular and Kagome

Frustrated magnets: Plethora of materials

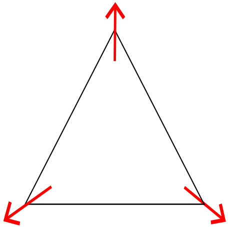
- Triangular lattice: $S = 1$ $AgNiO_2$ (Ni^{2+}), $S = 1/2$ Cs_2CuCl_4 (Cu^{2+})...
  Wheeler *et. al.* 2009; Coldea *et. al.* 2001...
- Kagome: $S = 5/2$ Fe jarosite (Fe^{3+}), $S = 1/2$ *Herbertsmithite* $ZnCu_3(OH)_6Cl_2$ (Cu^{2+})... (Han *et. al.* 2012; Fak *et. al.* 2007...)
- Pyrochlore spin ice $Ho_2Ti_2O_7$, pyrochlore-slab $S = 3/2$ $SrCr_{9p}Ga_{12-9p}O_{19}$ Cr^{3+} (SCGO)...
  (Harris *et. al.* 97; Limot *et. al.* 01......)

Single ion anisotropy can be large

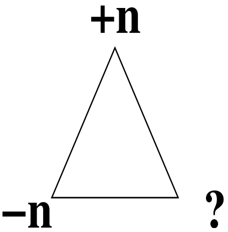
- Single ion anisotropy $-D(\mathbf{S}\cdot\mathbf{n})^2$can dominate over J
- Pyrochlore *spin ice* $Ho_2Ti_2O_7$ (Ho^{3+}, $(L+S)=8$)
  Easy axes **n** point outward from center of each tetrahedron
  $D \sim 50K$, $J \sim 1K$
  Harris *et. al.*, Phys. Rev. Lett. 79, 2554 (1997)
- Kagome Nd-langasite $Nd_3Ga_5SiO_{14}$ (Nd^{3+}, $(L+S)=9/2$)
  Easy axis perpendicular to lattice plane, $J \sim 2K$, $D \sim 10K$
  Robert *et. al.*, Physica B 2006
- $J \ll D$ is classical
  ☞ study leading quantum effects in a J/D expansion

Anisotropy amplies frustration

- Isotropic spins on a triangle



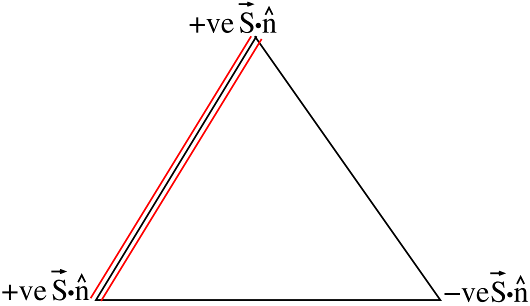
- Easy-axis **n** and triangular motifs...



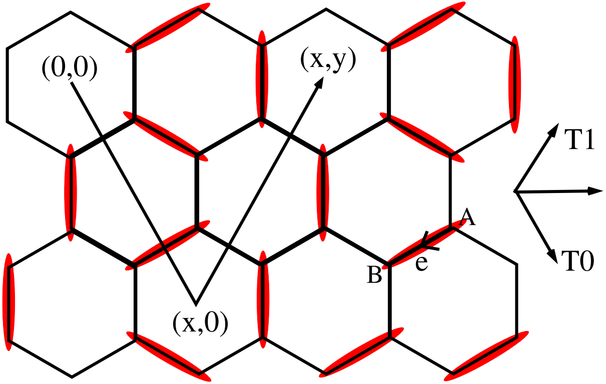
Wannier's triangular lattice model

- $H = J\sum_{\langle ij\rangle} \mathbf{S}_i \cdot \mathbf{S}_j - D\sum_i (S_i^z)^2$, with $D >> J$ on the triangular lattice.
- To leading order $S_i^z = \pm S \rightarrow \sigma = \pm 1$
  $H \approx JS^2 \sum_{\langle ij\rangle} \sigma_i \sigma_j$
- Minimum energy configurations?

Minimally frustrated configurations



- One frustrated bond per triangle
- Honeycomb lattice dimer model (one dimer touching each honeycomb site)



Honeycomb lattice dimer model: One dimer touching each honeycomb vertex

Classic problem in graph-theory/combinatorics/statistical mechanics

Ising 'liquid' in $T \to 0$ limit

- Calculation of Stephenson (64) gives

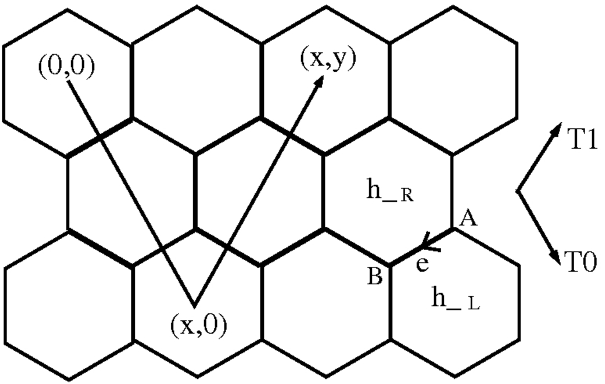
$$\langle \sigma(\mathbf{r})\sigma(0)\rangle \quad \sim \quad \frac{A}{r^{9/2}} + \frac{B\cos\left(2\pi(x+y)/3\right)}{\sqrt{r}}$$

- Spins neither freeze, nor fluctuate independently.
- Instead, form highly correlated "spin liquid".

Understanding this result:

- Dimers, heights, and Ising models of frustration
- (Obvious) connection to odd Ising gauge theories
- Connection to Kosterlitz-Thouless theory

Spins to dimers to electric fields

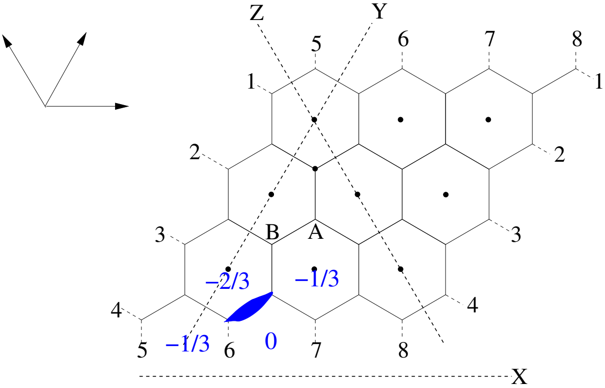


"Electric field" $\mathbf{e}_{\mathrm{A}\to\mathrm{B}} = n_{AB} - 1/3$
dimer constraint: Gauss law (!).
Youngblood and Axe (1980)

From dimers to microscopic heights $H(R)$

$$e_l = H_{L(l)} - H_{R(l)} \tag{1}$$

height field H on the original triangular lattice sites R



Henley 1990s

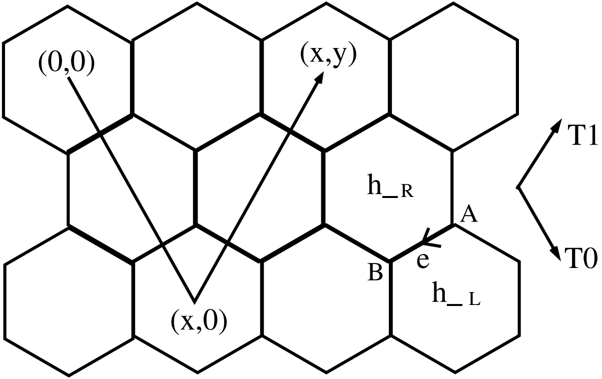
From microscopic $H(R)$ to coarse-grained $h(r)$

- Coarse-grain: Average over local rearrangements
- Locality: What happens "outside" cannot affect what happens "inside". $h(r) \to h(r) + 1$
  (Field theorists: "compactification radius")
- Lattice translations and $2\pi/6$ rotation ☞
  $h(r) \to h(r) + 1/3$, $h(r) \to -h(r)$

Ising spins in terms of $h(r)$

$3H$ jumps by odd (even) number whenever one crosses an unfrustrated (frustrated) bond

$\sigma(R) = \exp(-3\pi i H(R))$ (if $\sigma(R=0) = +1$, and $H(R=0) = 0$)



Dimer crossed $\rightarrow$ spin unchanged; empty link $\rightarrow$ spin flipped

$\sigma(R) = \exp\left(i\pi \sum_l (1 - n_l)\right) \sigma(0)$

$\sigma(R) = \exp(\frac{2\pi}{3} i(X + Y) - i\pi H(R))$

Fradkin et al (2004), KD (2009)

Effective action and operators

- Fewer flippable plaquettes $\rightarrow$ larger "tilt"
  $\mathcal{S}_{\text{eff}} = \frac{\pi}{g}(\nabla h)^2 + \lambda_6 \cos(6\pi h) + \ldots$
  Coarse-grained representation of spins:
  $\sigma(r) \sim Ae^{i\mathbf{Q}\cdot r}e^{-i\pi h(r)} + Be^{-3i\pi h(r)} + h.c.$
  Three-sublattice order parameter $\psi \sim e^{i\pi h} \equiv e^{i\theta}$ (!)

$T > 0$: Odd Ising gauge theory and Kosterlitz-Thouless vortices

- Nonzero temperature: Fully frustrated triangle $\rightarrow$ three dimers touching honeycomb site.
- "Electric field $E_{\mathrm{A}\rightarrow\mathrm{B}} = n_{AB} - 1/3$ no longer divergence-free
  But violations are 0 mod 2
  Field-theory language: Configuration space of odd-Ising gauge theory
- Heights no longer single valued
  Three dimers touching honeycomb site $\rightarrow$ vortex/antivortex in $\theta = \pi h$
- $T = 0$: Vortex-free *xy* model for θ with 6-fold anisotropy
  $T > 0$: Vortices allowed

Picture for $T = 0$ power-law ordered phase

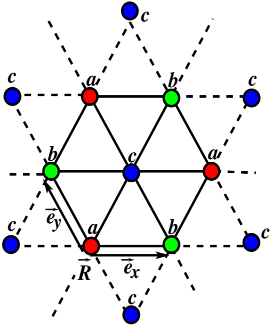
- In state with long-range three-sublattice order, θ feels $\lambda_6 \cos(6\theta)$ potential.
  Locks into values $2\pi m/6$ (resp. $(2m+1)\pi/6$) in ferri (resp. antiferro) three-sublattice ordered state
- In power-law three-sublattice ordered state λ_6 does not pin phase θ
  θ spread uniformly $(0, 2\pi)$
- But vortices absent.

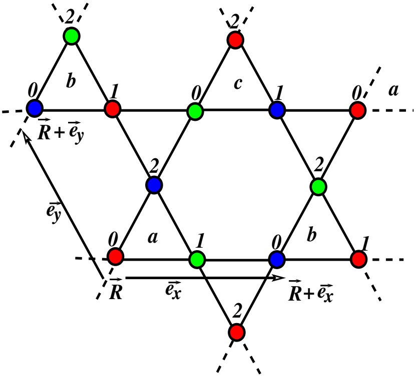
RG description

- Fixed point action: $S = \frac{1}{4\pi g}(\nabla\theta)^2$
- For $g > \frac{1}{9}$
  $\lambda_6 \cos(6\theta)$ *irrelevant* along fixed line
  ☞ $\langle\psi^*(r)\psi(0)\rangle \sim \frac{1}{r^{\eta(T)}}$ with $\eta = g$
  Relies on absence of vortices at $T = 0$

Jose, Kadanoff, Kirkpatrick, Nelson (1977)

Easy-axis antiferromagnets on triangular lattices

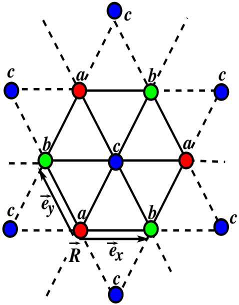




Natural tripartite structure ☞

Perturbations/quantum fluctuations easily stabilize this order...

Three-sublattice order on the triangular lattice



θ	● (red)	● (green)	● (blue)
0	–S	+S	+S
$\frac{\pi}{6}$	–S	0	+S

$\psi = |\psi| e^{i\theta} = -\sum_{\vec{R}} e^{i\mathbf{Q}\cdot\vec{R}} S^z_{\vec{R}}$

Ferri vs antiferro order distinguished by the choice of phase θ

Ferri: $\theta = 2\pi m/6$, Antiferro: $\theta = (2m+1)\pi/6$ ($m = 0, 1, 2 \ldots 5$)

Very recent: $TmMgGaO_4$ (Thulium [Xe] $4f^{13}$ $6s^2$) Tm^{3+} $J_z = \pm 6$

Princeton, Augsburg, Fudan (2017-18)

Prototypical example of order-by-(quantum) disorder

- $H_{\text{TFIM}} = J \sum_{\langle ij \rangle} \sigma_i^z \sigma_j^z - \Gamma \sum_i \sigma_i^x$ on the triangular lattice
  small Γ ☞ Long-range order at three-sublattice wavevector **Q**
- Ordering of "antiferro" type $\rightarrow (+, -, 0)$
  antiferro order provides maximum "room" for quantum fluctuations
  Moessner, Sondhi, Chandra (2001), Isakov & Moessner (2003)

Another example: $S = 1$ with easy-axis single-ion anisotropy

- $H_{\mathrm{AF}} = J\sum_{\langle ij\rangle} \vec{S}_i \cdot \vec{S}_j - D\sum_i (S_i^z)^2$ on triangular lattice
  Closely related to effective model for AgNiO2
  (Seabra & Shannon '11)
- Low-energy physics for $D \gg J$:
  $H_b = -\frac{J^2}{D}\sum_{\langle ij\rangle}(b_i^\dagger b_j + h.c.) + J\sum_{\langle ij\rangle}(n_i - \frac{1}{2})(n_j - \frac{1}{2}) + \ldots$
  KD & Senthil '06
- Low-temperature state for $D \gg J$: "supersolid" state of hard-core bosons at half-filling.
  Auerbach & Murthy (97), Heidarian & KD, Melko, Wessel...(05)
- Implies: ferri three-sublattice order in S^z + "ferro-nematic" order in $\vec{S}_\perp^2$
  (Simple easy-axis version of Chandra-Coleman (1991) "spin-nematic" ideas)

Symmetry breaking transitions: Generalities

- Symmetry-breaking state characterized by long-range correlations of "order-parameter" $\hat{O}$
- phenomenological Landau free energy density $\mathcal{F}[\hat{O}]$
  Expanding $\mathcal{F}$ in powers of $\hat{O}$ (symmetry allowed terms)
- Neglecting spatial variation & fluctuations:
  phase transition $\rightarrow$ change in minimum of $\mathcal{F}$

Fluctuation effects at continuous transitions:

- More complete description of long-wavelength physics:
  Include (symmetry allowed) gradient terms in $\mathcal{F}$
  Integrate over all possible order parameter configurations
- In most cases: Corrections to mean-field exponents

Symmetries are (usually) decisive:

- Transformation properties of $\hat{O}$ determine nature of continuous transition

Landau-theory for melting of three-sublattice order

- $\mathcal{F} = K|\nabla\psi|^2 + r|\psi|^2 + u|\psi|^4 + \lambda_6(\psi^6 + \psi*^6) + \dots$
  Connection with six-state clock models
  $Z = \sum_{\{p_i\}} \exp[\sum_{\langle ij \rangle} V(\frac{2\pi}{6}(p_i - p_j))]$
  Each $p_i = 0, 1, 2, ...5$
  $V(x) = K_1 \cos(x) + K_2 \cos(2x) + K_3 \cos(3x)$
  Cardy (1980)

Simplest lattice model

$$H_{\rm xy} \quad = \quad -J_{\rm xy} \sum_{\langle \vec{r}\vec{r}' \rangle} \cos(\theta_{\vec{r}} - \theta_{\vec{r}'}) - h_6 \sum_{\vec{r}} \cos(6\theta_{\vec{r}}) \; .$$

(higher harmonics $J^{(p)}$ ($p = 2, 3$) left out of $H_{\rm xy}$ for simplicity)

Melting scenarios for three-sublattice order

- Analysis (Cardy 1980) of generalized six-state clock models
  $\rightarrow$ Three generic possibilities of relevance here:
  Two-step melting, with power-law ordered intermediate phase
  OR
  3-state Potts transition to ferromagnetic phase followed by loss of ferromagnetism via Ising transition at higher temperature..
  or vice-versa...
  OR
  First-order transition (always possible!)

Melting of three-sublattice order in various examples

- Antiferro three-sublattice order in triangular lattice transverse field Ising model
  Two-step melting
  (Isakov & Moessner ’01)
- Ferrimagn. three-sublattice order in triangular lattice-gas models of monolayer films
  Two-step melting
  D.P. Landau ’83
- Ferri. three-sublattice order in Kagome Ising antiferromagnets
  With second-neighbour ferro couplings: Two step melting
  Wolf & Schotte ’88
  With long-range dipolar couplings: Three-state Potts transition
  Moller & Moessner ’09, Chern, Mellado, Tchernyshyov ’11

Detecting power-law order?

Need scattering experiment to detect power-law version of Bragg peaks

Or

Real-space data by scanning some local probe $+$ Lots of image-processing

Alternate thermodynamic signature(!)

- Singular thermodynamic susceptibility to *uniform* easy-axis field B:
  $\chi_u(B) \sim \frac{1}{|B|^{p(T)}}$
- $p(T) = \frac{4-18\eta(T)}{4-9\eta(T)}$ for $\eta(T) \in (\frac{1}{9}, \frac{2}{9})$
  So $p(T)$ varies from $2/3$ to 0 as T increases from T_{c1} to just below T_{c2}

(KD '15)

Recall: picture for power-law ordered phase

- In state with long-range three-sublattice order, θ feels $\lambda_6 \cos(6\theta)$ potential.
  Locks into values $2\pi m/6$ (resp. $(2m+1)\pi/6$) in ferri (resp. antiferro) three-sublattice ordered state for $T < T_{c1}$
- In power-law three-sublattice ordered state for $T \in (T_{c1}, T_{c2})$, λ_6 does not pin phase θ
  θ spread uniformly $(0, 2\pi)$
- But vortices continue to be irrelevant
  Distinction between ferri and antiferro three-sublattice order lost for $T \in (T_{c1}, T_{c2})$
  Ferromagnetic response part of the time...

Recall: More formally

- Fixed point free-energy density: $\frac{\mathcal{F}_{\mathrm{KT}}}{k_B T} = \frac{1}{4\pi g}(\nabla\theta)^2$
  with $g(T) \in (\frac{1}{9}, \frac{1}{4})$ corresponding to $T \in (T_1, T_2)$
- $\lambda_6 \cos(6\theta)$ *irrelevant* along fixed line
- $\langle \psi^*(r)\psi(0)\rangle \sim \frac{1}{r^{\eta(T)}}$
  with $\eta(T) = g(T)$

Jose, Kadanoff, Kirkpatrick, Nelson (1977)

General argument—I

Starting point: Ferrimagnetic three-sublattice order also involves uniform magnetization m

More complete theory should treat m and ψ on equal footing

- Symmetries allow coupling term $\tilde{\lambda}_3 m(\psi^3 + \psi*^3)$
  augment $\frac{\mathcal{F}_{\rm KT}}{k_B T}$ with gapped free-energy density $\mathcal{F}_{\rm ferro}(m)$:
  $\mathcal{F}_{\rm ferro}(m) + \lambda_3 m \cos(3\theta)$
- λ_3 formally irrelevant along fixed line $\mathcal{F}_{\rm KT}$
  $\rightarrow$
  Physics of two-step melting unaffected—m "goes for a ride..."

But ...

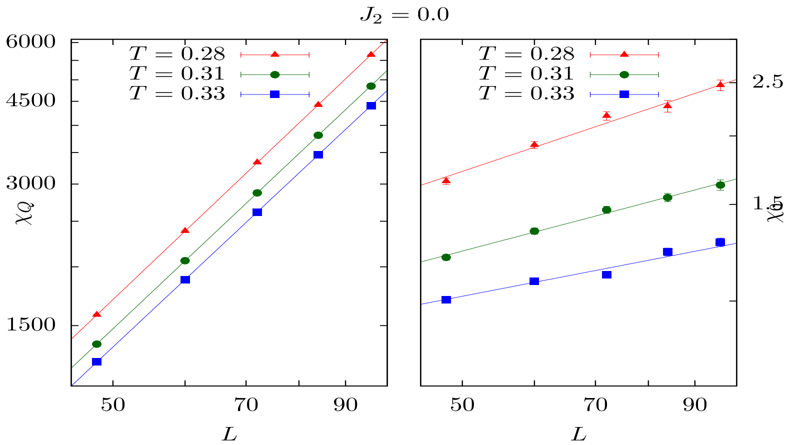
General argument—II

- m "inherits" power-law correlations of $\cos(3\theta)$:
  $C_m(r) = \langle m(r) m(0) \rangle \sim \frac{1}{r^{9\eta(T)}}$
- $\chi_L \sim \int^L d^2 r C_m(r)$ in a finite-size system at $B = 0$
- $\chi_L = \chi_{\rm reg} + b L^{2-9\eta(T)}$ for $\eta(T) \in (\frac{1}{9}, \frac{2}{9})$
  Diverges with system size at $B = 0$

General argument—III

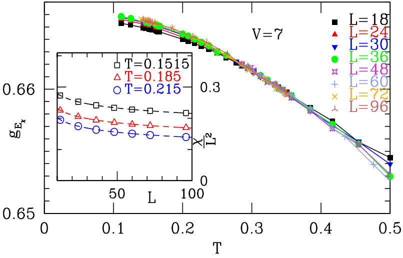
- Uniform field $B > 0 \rightarrow$ additional term $h_3 \cos(3\theta)$ in $\mathcal{F}_{\mathrm{KT}}$
- Strongly relevant along fixed line, with RG eigenvalue $2 - 9g/2$
- Implies finite correlation length $\xi(B) \sim |B|^{-\frac{2}{4-9\eta}}$
- $\chi_u(B) \sim |B|^{-\frac{4-18\eta}{4-9\eta}}$ for $\eta(T) \in (\frac{1}{9}, \frac{2}{9})$

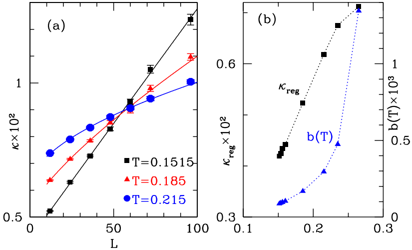
Test in prototypical example



In power-law ordered phase of H_{TFIM} on triangular lattice (Biswas & KD '18)

Test in KT phase of easy-axis $S = 1$ triangular lattice AFM





In power-law ordered phase of H_b
(Heidarian & KD '17)

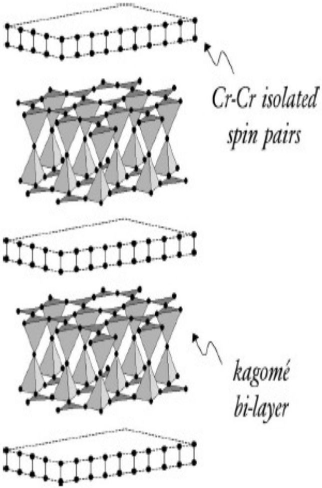
Also interesting:—Multicritical melting

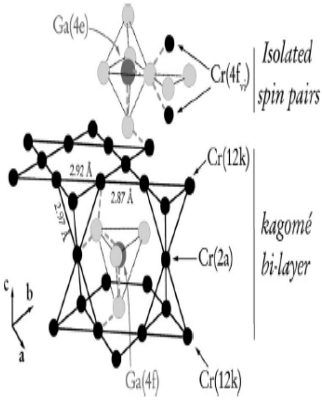
- KT phase can
  Pinch-off at multicritical point $\mathcal{M}_?$, giving way to three-state Potts criticality. $c_{\mathcal{M}_?} = ?$
  OR
  Pinch-off at multicritical point $\mathcal{M}_{\rm Clock}$, giving way to first-order transition line.
  (KD '15)
- $\mathcal{M}_{\rm Clock}$ previously known, not $\mathcal{M}_?$
  Note: Conjecture (Dorey-Tateo-Thompson '96) relates $\mathcal{M}_{\rm Clock}$ to self-dual Z_6 $c = 1.25$ CFT (Zamolodchikov-Fateev '85)
  $\rightarrow c_{\mathcal{M}_{\rm Clock}} = 1.25$
  (KD '15)

Also interesting: Multicritical melting

- How does the KT phase pinch-off for specific cases?
  - Evidence for $\mathcal{M}_{\mathrm{Clock}}$ on the triangular lattice
    (Rakala, Shivam, & KD '19)
  - Similar results on Kagome lattice systems
    Conjecture for $\mathcal{M}_{?}$ in triangular bilayers
    (Rakala & KD unpublished)

Also interesting: Fractional moments in SCGO

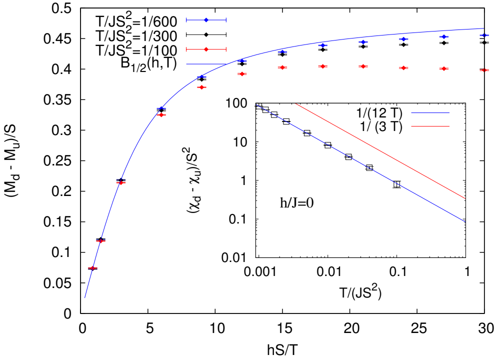




Idealized $SrCr_9Ga_3O_{19}$ unrealizable. $\rightarrow$ Instead: $SrCr_{9p}Ga_{12-9p}O_{19}$ with $p_{max} \approx 0.95$

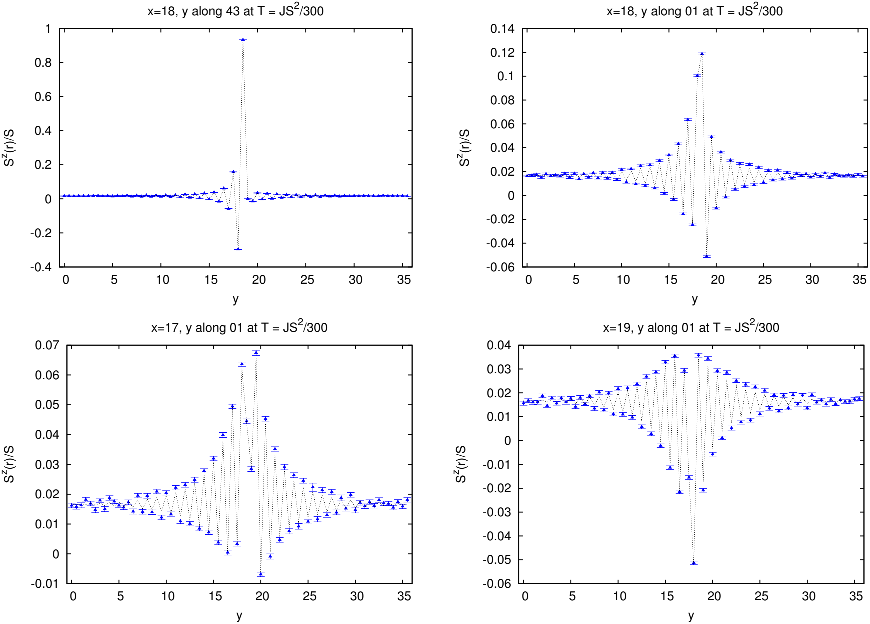
$J_{\text{bilayer}} \approx 80K$ $J_{\text{dimers}} \approx 200K$ *Limot et al PRB 02*

Also interesting: Fractional moments

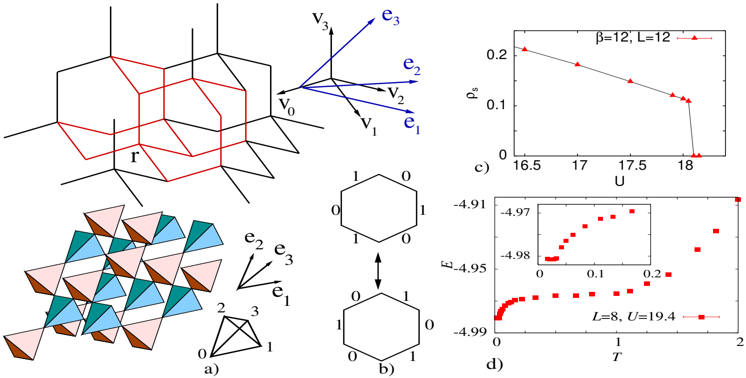


- $\chi_{\rm imp}(T)$ fits Curie law $S_{\rm eff}^2/3T$ with $S_{\rm eff} = S/2$
- Full magnetization curve of impurity-induced magnetization predicted correctly.

Spin texture: Theory vs “experiment”



Pyrochlore lattice: Emergent electrodynamics



$H_{XXZ} = U \sum_{\text{tetra}} (\sigma^z_{\text{tetra}})^2 - t \sum_{\text{links}} (\sigma_i^+ \sigma_j^- + h.c.)$ Coulomb spin liquid for $t \ll U$

Banerjee, Isakov, KD, YB Kim (2008)

Acknowledgements and references

Students: D. Heidarian, A. Banerjee, A. Sen, P. Dutt, K. Ramola, G. Rakala, S. Biswas, S. Shivam, N. Desai

Resources: Computational cluster of DTP, Fell-Fund Oxford, ICTS TIFR, ARCUS (Orsay), MPIPKS (Dresden), CEFIPRA...

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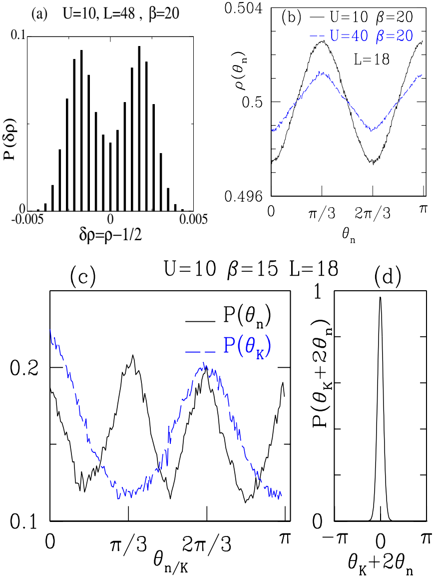
Some loose ends—I

Some loose ends...Three sublattice order and its melting in S=1 easy axis triangular antiferromagnet, and in classical Ising models on the triangular lattice

Is three-sublattice ordering of S^z in H_{AF} ferri or antiferro?

- Natural expectation: Quantum fluctuations induce antiferro order (like in the transverse field Ising model)
  $\rightarrow$
  Initial confusion: Ordering will be antiferro three-sublattice order
  e. g. Melko *et. al.* (2005)

Actual state has ferrimagnetic three-sublattice order

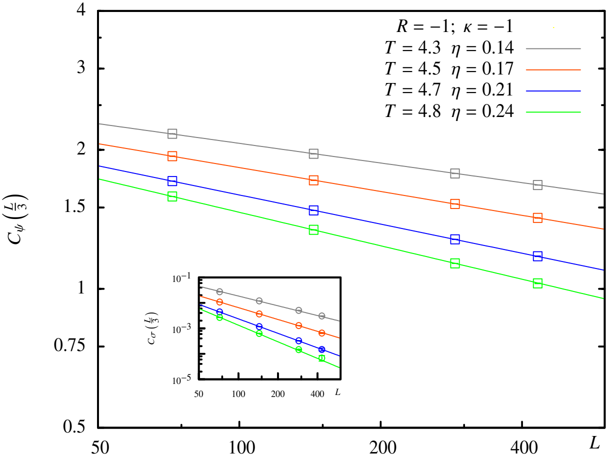


Heidarian & KD '05

Early work: Triangular lattice-gas models for monolayer films on graphite

- Three-sublattice long-range order of noble-gas monolayers on graphite
  Birgeneau, Bretz, Chan, Vilches, Wiechert...(1970—1990)
  $H_{\mathrm{J_1J_2}} = J\sum_{\langle ij\rangle}\sigma_i^z\sigma_j^z - J_1\sum_{\langle\langle ij\rangle\rangle}\sigma_i^z\sigma_j^z - J_2\cdots - B\sum_i\sigma_i^z$
  Long-range three-sublattice ordering (wavevector **Q**) at low temperature
  D. P. Landau (1983)

Test in J1-J2model

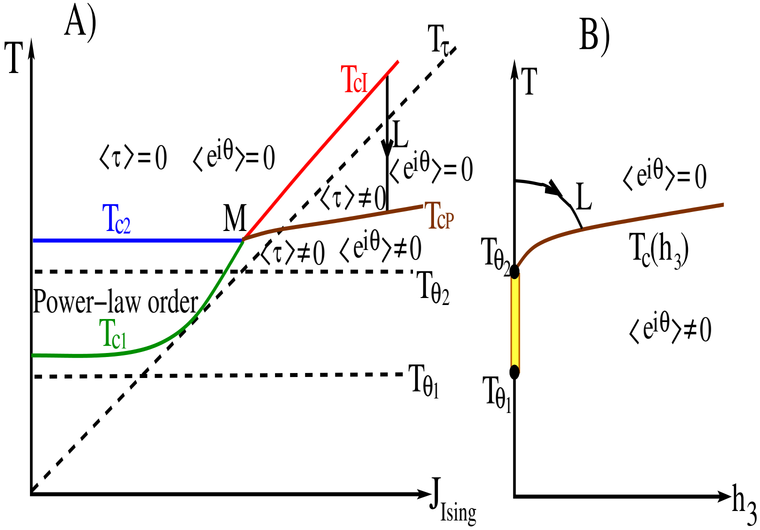


In power-law ordered phase of $H_{J_1J_2}$
$(R = -(J_1 + J_2)/J$ and $\kappa = (J_2 - J_1)/J)$
(Geet Rakala & KD in prep.)

Some loose ends—II

Some loose ends...multicritical melting

Multicritical melting of three-sublattice order



(KD ’15)

More complete coarse-grained description

$$
\begin{aligned}
H_{\rm eff} &= H_{\rm xy} + H_{\rm Ising} - J_{\theta\tau} \sum_{\vec{r}} \tau_{\vec{r}} \cos(3\theta_{\vec{r}}) \,, \\
\text{where} \quad H_{\rm Ising} &= -J_{\rm Ising} \sum_{\langle \vec{r}\vec{r}' \rangle} \tau_{\vec{r}} \tau_{\vec{r}'} - h \sum_{\vec{r}} \tau_{\vec{r}} \,, \\
H_{\rm xy} &= -J_{\rm xy} \sum_{\langle \vec{r}\vec{r}' \rangle} \cos(\theta_{\vec{r}} - \theta_{\vec{r}'}) - h_6 \sum_{\vec{r}} \cos(6\theta_{\vec{r}}) \,,
\end{aligned}
$$

with $h \propto B$.
(KD '15)

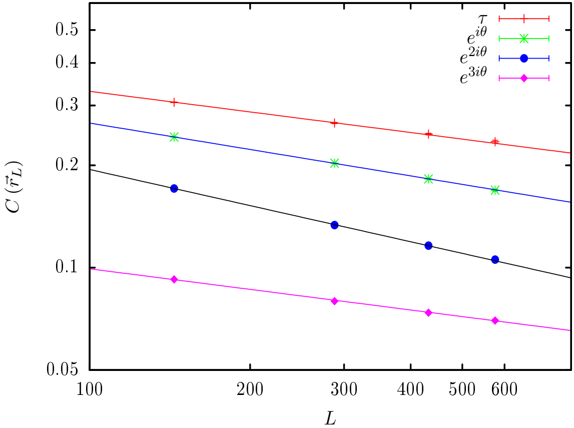
The argument...

- Start with known phase diagrams of $H_{\rm xy}$ and $H_{\rm Ising}$ and build in effects of $J_{\theta\tau}$
- When τ orders, $H_{\rm xy}$ sees effective three-fold symmetric perturbation $h_{\rm 3eff}\cos(3\theta_{\vec{r}})$ with $h_{\rm 3eff} \sim \langle\tau\rangle$
- When $e^{i\theta}$ orders, $H_{\rm Ising}$ sees effective field $h_{\rm eff}\tau_{\vec{r}}$ with $h_{\rm eff} \sim \langle\cos(3\theta)\rangle$

The "new" multicritical point $\mathcal{M}_?$

- c-theorem argument: $1 \leq c \leq \frac{3}{2}$
- To search:
  $J_{\text{xy}} = h_6 = 1.0$, $J_{\theta\tau} = 0.25$
  Parametrize: $J_{\text{Ising}} = f_{xy} T_{\theta_1} / T_\tau$ and $T = f_I f_{xy} T_{\theta 1}$ [with $T_{\theta 1} = 1.04$ and $T_\tau = 3.6409$]

Multicritical melting at $\mathcal{M}_?$



$[f_{\mathrm{xy}}^{\mathcal{M}_?}, f_I^{\mathcal{M}_?}] \approx [1.5570(8), 1.0061(5)]$

$C_{2\theta}$ [$C_{3\theta}$] rescaled by a factor of 7 [factor of 10]

$\eta_{3\theta} = \eta_\tau = 0.201(20)$, $\eta_\theta = 0.258(5)$, and $\eta_{2\theta} = 0.353(6)$.

(KD '15)

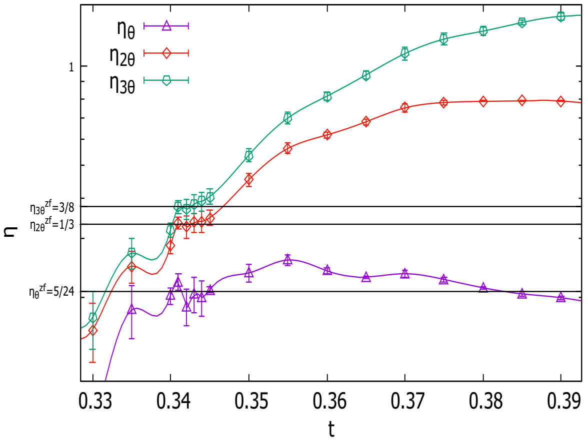
Speculation (aka wishful thinking?)

- If relative strength of first/second neighbour exchange tunable relative to long-range dipolar part in artificial kagome-ice: Could tune melting to multicritical point $\mathcal{M}_?$...
- Computations challenging due to long-range interactions

$\mathcal{M}_?$ vs $\mathcal{M}_{\rm clock}$

- Conjecture (Dorey '96): $\mathcal{M}_{\rm clock}$ corresponds to $c = 1.25$ self-dual Z_6 CFT constructed by Zamolodchikov-Fateev ('85).
- Conjecture yields exponents at $\mathcal{M}_{\rm clock}$: $\eta_{3\theta} = 3/8$, $\eta_{2\theta} = 1/3$, and $\eta_\theta = 5/24$.
  $\eta_{2\theta}$ and $\eta_{3\theta}$ very different from values at $\mathcal{M}_?$
  Recall: at $\mathcal{M}_?$, $\eta_{3\theta} = \eta_\tau = 0.201(20)$, $\eta_\theta = 0.258(5)$, and $\eta_{2\theta} = 0.353(6)$.

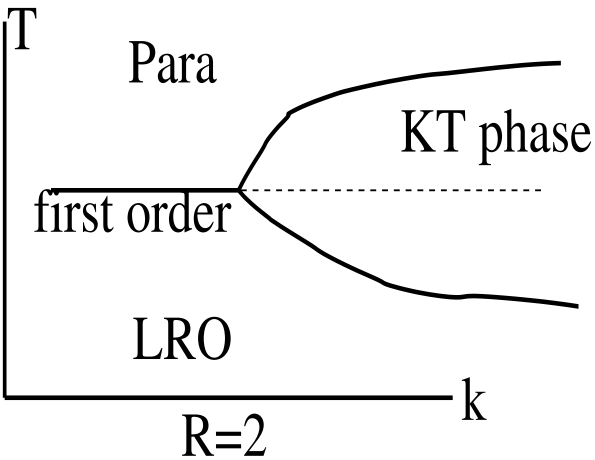
Test of conjectured exponents for $\mathcal{M}_{\text{clock}}$



Results on Cardy's six-state clock model

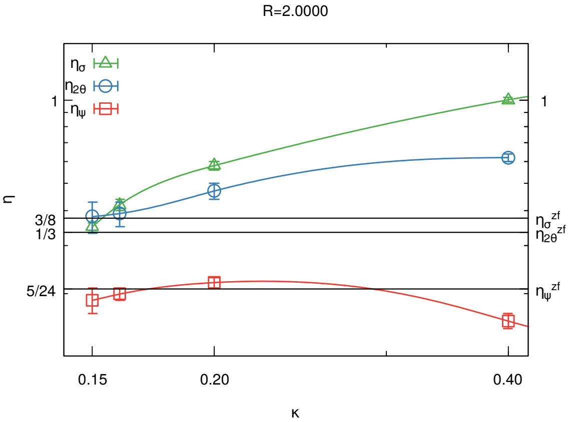
(Rakala, Shivam, & KD in prep.)

Schematic of pinch-off in triangular lattice Ising AFM



$J_1 = 1$, $R = J_2 + J_3$, $\kappa = J_2 - J_3$

Evidence for $\mathcal{M}_{\text{clock}}$ in triangular Ising AFM



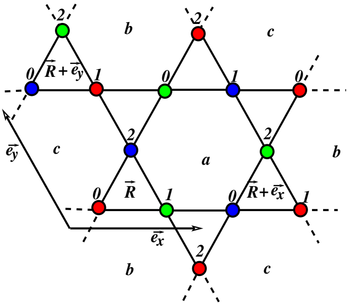
(Rakala, Shivam, & KD in prep.)

Some loose ends—III

Some loose ends...previous work on Kagome systems

Three-sublattice order on the Kagome lattice

θ	red	green	blue
0	−S	+S	+S
$\frac{\pi}{6}$	−S	0	+S



$$\psi = |\psi| e^{i\theta} = -\sum_{\vec{R}} \sum_{\alpha=0,1,2} e^{i\mathbf{Q}\cdot\vec{R} - 2\pi i \frac{\alpha}{3}} S^z_{\vec{R},\alpha}$$

Again: Ferri vs antiferro distinguished by the choice of phase θ

Ferri: $\theta = 2\pi m/6$, Antiferro: $\theta = (2m+1)\pi/6$ ($m = 0, 1, 2 \ldots 5$)

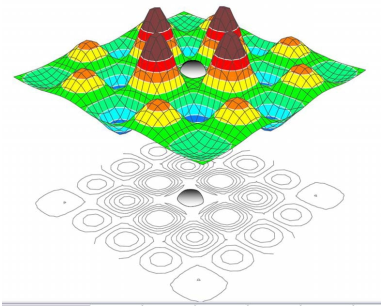
Ising models for “Artificial Kagome-ice”

- $H_{\text{Kagome}} = J \sum_{\langle ij \rangle} \sigma_i^z \sigma_j^z - J_1 \sum_{\langle\langle ij \rangle\rangle} \sigma_i^z \sigma_j^z - J_2 \dots$
- Only nearest-neighbour couplings $\rightarrow$ classical short-range spin liquid (Kano & Naya 1950)
- Second-neighbour ferromag. couplings destabilize spin liquid (Wolf & Schotte 88)
  Ferrimagnetic three-sublattice order at low T.
- “Artificial Kagome-ice: Moments $\mathbf{M}_i = \sigma_i^z \mathbf{n}_i$
  ($\mathbf{n}_i$ at different sites non-collinear)
  Expt: Tanaka *et. al.* (2006), Qi *et. al.* (2008), Ladak *et. al.* (2010,11)
  Theory: Moller, Moessner (2009), Chern *et. al.* (2011)

Some loose ends—IV

Some loose ends...quick introduction to SCGO and its Galling(!) defects

Impurities as probes



Alloul *et. al.* Rev. Mod. Phys. **81**, 45 (2009).

- Vacancy defect (Zn substition at Cu site in cuprate AF insulators) ☞characteristic response in local susceptibility.
- Picked up by local probes like NMR:
  ☞NMR line position shift (Knight shift) measures local spin-polarization of spin system (via hyperfine coupling to nuclear moment).
  ☞Measures histogram of local susceptibility at various distances from impurity

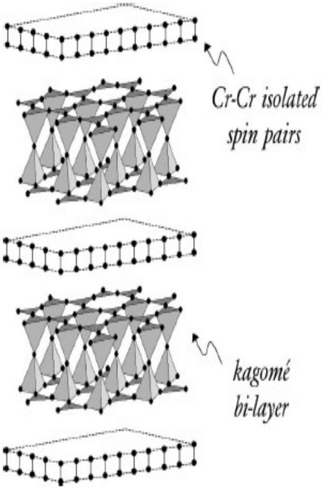
General idea

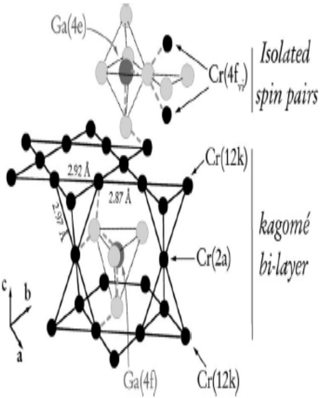
- Impurities disturb the system locally
  Host response characteristic of correlations of the low temperature state
- Correlations encoded in intricate charge/spin textures seeded by impurities
- Picked up by local probes like NMR and STM

Our focus: $SrCr_9Ga_3O_{19}$ (SCGO)

- In this talk: Non-magnetic Ga impurities in pyrochlore slab magnet SCGO
  Insulating magnet: Cr^{3+} ☞ $S = 3/2$ moments.
  No significant anisotropy (exchange or single-ion).
  $\rightarrow$ Vacancy-defect induced spin textures and their interactions in a classical spin liquid

Anatomy: SCGO and its Galling defects





Idealized $SrCr_9Ga_3O_{19}$ unrealizable. $\rightarrow$ Instead: $SrCr_{9p}Ga_{12-9p}O_{19}$ with $p_{max} \approx 0.95$

$J_{\mathrm{bilayer}} \approx 80K$ $J_{\mathrm{dimers}} \approx 200K$ *Limot et al PRB 02*

Anatomy: Where do the Ga go?

- Slight bias towards $4f$ sites
  Break isolated dimers
- Close runners-up are $12k$ sites
  And substitute into upper or lower Kagome layers
- Significantly lower probability of going to the $2a$ sites
  Rarely substitute for ‘apical’ spins

(neutron diffraction, quoted in *Limot et. al. 2002*)

Behaviour—Macroscopic susceptibility

- High temperature χ fits Curie-Weiss form, with $\Theta_{CW} \approx 500$—$600K$.
  [from extrapolation of linear behaviour for χ^{-1}]
- But: No sign of any magnetic ordering down to $T_f \sim 3$—$5K$
- At $T = T_f$, some kind of freezing transition.
  [cusp in susceptibility]
- (Spin) glassy behaviour for $T < T_f$.
  [hysterisis between field-cooled vs zerofield cooled data]
- Nature of phase for $T < T_f$ not clear at present
  [Not our focus here]

Magnetic susceptibility in spin liquid regime

- Macroscopic susceptibility measurements have interesting "two-fluid" phenomenology:
  An "intrinsic part", well-behaved and finite until the freezing transition is approached.
  A "defect contribution" $\chi_{def} = C_d/T$, with $C_d \propto (1-p) \equiv x$
  Attributed to "orphan-spin population", Schiffer-Daruka (97)

NMR in spin liquid regime

- Broad, apparently symmetric Ga NMR line (field-swept), with broadening $\Delta H \propto \mathcal{A}(x)/T$ and $\mathcal{A}(x) \sim x$ for not-too-small x.
  Attributed to a short-ranged oscillating spin density near defects, Limot *et. al.* (2000,2002). Orphan spins of Schiffer-Daruka?

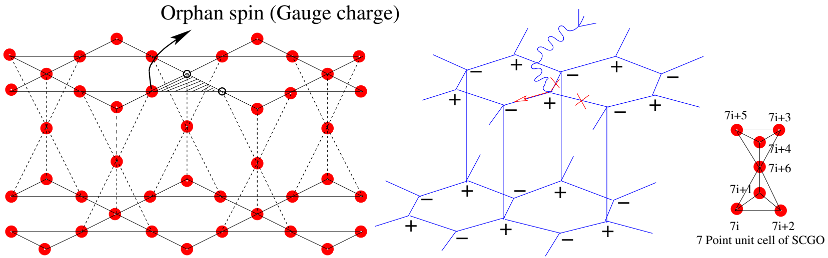
Some theory: $T = 0$ Simplex satisfaction

$$H = \frac{J}{2}\sum_{\boxtimes}(\sum_{i\in\boxtimes}\vec{S}_i - \frac{\mathbf{h}}{2J})^2 + \frac{J}{2}\sum_{\triangle}(\sum_{i\in\triangle}\vec{S}_i - \frac{\mathbf{h}}{2J})^2$$

- Absolute minimum of energy is achievable:
  If no symmetry breaking: $S^z_{Kag} = h/6J$, $S^z_{apical} = 0$
  (for $\mathbf{h} = h\hat{z}$)
  Henley (2000)

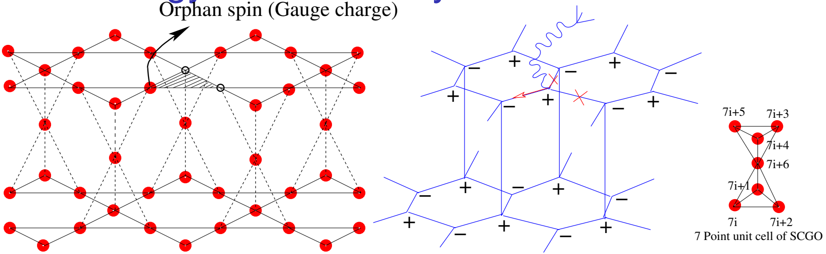
Relies on constructing states that also satisfy $\vec{S}_i^2 = S^2$ for h not-to-large.

Some theory: Half-orphans



- Single Ga on any simplex $\rightarrow$ no problem with simplex satisfaction
- If two Ga in one $\triangle$ $\rightarrow$ $\triangle$ has only one spin
  $\langle S^z_{\mathrm{tot}} \rangle = \frac{1}{2}\sum_{\mathrm{simplices}} \langle S^z_{\mathrm{simplices}} \rangle = S/2 = 3/4!$ (at $T = 0$, $h/J \rightarrow 0$)
  Half-Orphan spins
  Henley (2000)

Aside: Analogy with electrodynamics



$$\sum_{i \in \boxtimes} S_i^\alpha = \frac{h^\alpha}{2J} \quad \text{and} \quad \sum_{i \in \triangle} S_i^\alpha = \frac{h^\alpha}{2J}$$

- $\mathbf{E}_i^\alpha = S_i^\alpha \hat{e}_i$,
  (Unit vector $\hat{e}_i$ points along the dual bond from dual $+$ sublattice to dual $-$ sublattice.)
- Simplex satisfaction at $h = 0 \rightarrow \nabla \cdot \mathbf{E}^\alpha = 0$ at $T = 0$.
- On defective simplex: $(\nabla \cdot \mathbf{E}^\alpha)_\triangle = S^\alpha_{\text{orphan}}$
- But $T = 0$ Gauss law $\rightarrow 1/\vec{r}$ decay of $T = 0$ induced spin-texture.

What happens at $T > 0$?

Simplex satisfaction *a la* Henley is inherently a $T = 0$ statement
What about $T > 0$?
Answer not obvious...

- But, curiously:
  Defective tetrahedron/triangle (with all but one spin removed) give Curie tail; no other simplices contribute to Curie tail. (Moessner-Berlinsky 99)

Real issue: Need to incorporate correlations (long-range as $T \to 0$) between spins on equal footing with thermal fluctuations.

Are there “really” fractional half-orphan spins at $T > 0$?

Our approach

Putting entropic effects on same footing as energetics:

- In pure problem: Large N theory known to be very accurate
  Garanin & Canals, 1999; Isakov *et. al.* 2004
- Effective field theory $Z \propto \int \mathcal{D}\vec{\phi} \exp\left(-\mathcal{F}/T\right)$
  Free-energy functional $\mathcal{F} = E - T\mathcal{S}$ with
  $E = \frac{J}{2}\sum_{\boxtimes}(\sum_{i\in\boxtimes}\vec{\phi}_i - \frac{\mathbf{h}}{2J})^2 + \frac{J}{2}\sum_{\triangle}(\sum_{i\in\triangle}\vec{\phi}_i - \frac{\mathbf{h}}{2J})^2$
  statistical weight $\mathcal{S} \propto \left(-\frac{\rho_1}{2}\sum_{i\in\mathrm{Kagome}}\vec{\phi}_i^2 - \frac{\rho_2}{2}\sum_{i\in\mathrm{apical}}\vec{\phi}_i^2\right)$

ρ_1 and ρ_2 phenomenological parameters

Use values that satisfy $\langle\vec{\phi}_i^2\rangle = S^2$

(Gaussian theory$\rightarrow$Independent effective action for each spin component)

Modeling the half-orphans in effective field theory

- Ga substitution implies constraint
  $\vec{\phi}_{\mathrm{Ga}} = 0$
- Lone spin on defective triangle needs to be handled carefully: Retain as a classical spin S variable $S\vec{n}$ (with $\vec{n}$ a unit vector).

General framework

Vacancies:

$$\delta(\phi^{\alpha}_{\vec{r}}) = \frac{1}{2\pi}\int d\lambda^{\alpha}_{\vec{r}} \exp(i\lambda^{\alpha}_{\vec{r}}\phi^{\alpha}_{\vec{r}})$$

Lone-spins on defective triangles/tetrahedra:

$$\delta(\phi^{\alpha}_{\vec{r}} - Sn^{\alpha}_{\vec{r}}) = \frac{1}{2\pi}\int d\mu^{\alpha}_{\vec{r}} \exp(i\mu^{\alpha}_{\vec{r}}(\phi^{\alpha}_{\vec{r}} - Sn^{\alpha}_{\vec{r}}))$$

Combined notation:

$$\Lambda^{\alpha}_{\vec{r}} = \delta_{\vec{r},\vec{r}_v}\lambda^{\alpha}_{\vec{r}_v} + \delta_{\vec{r},\vec{r}_o}\mu^{\alpha}_{\vec{r}_o}$$

Action for μ, λ, $\vec{n}$

$$Z_{\mathrm{eff}} \propto \int \mathcal{D}\vec{n} \int \mathcal{D}\vec{\lambda} \int \mathcal{D}\vec{\mu}$$

$$\exp\left(+\frac{1}{2} \sum_{\vec{r}\vec{r}'\,\alpha} (\beta h^{\alpha} + i\Lambda^{\alpha}_{\vec{r}}) C_{\vec{r}\vec{r}'} (\beta h^{\alpha} + i\Lambda^{\alpha}_{\vec{r}'}) - i \sum_{\vec{r}_o\alpha} \mu^{\alpha}_{\vec{r}_o} n^{\alpha}_{\vec{r}_o} \right)$$

C: Matrix of zero-field correlations in pure large-N theory

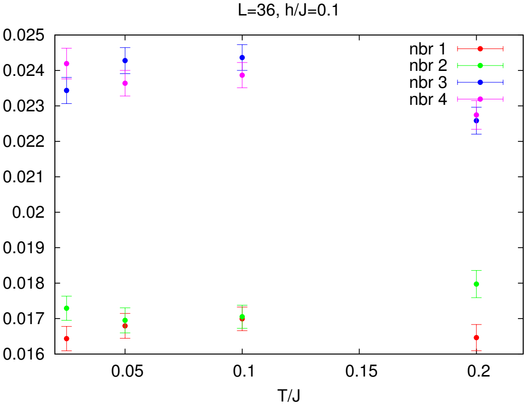
$$\langle \phi^{\alpha}_{\vec{r}} \phi^{\beta}_{\vec{r}'} \rangle \equiv C_{\vec{r}\vec{r}'} \delta_{\alpha\beta}$$

General approach

- Do integrals over λ and μ *exactly*.
- Get effective theory for orphan spins (unit vectors $\vec{n}$) coupled to each other and to external magnetic field
- Analytically tractable for one or two or three defective triangles

Isolated vacancies to not contribute to Curie term

Susceptibility of sites around a single missing spin



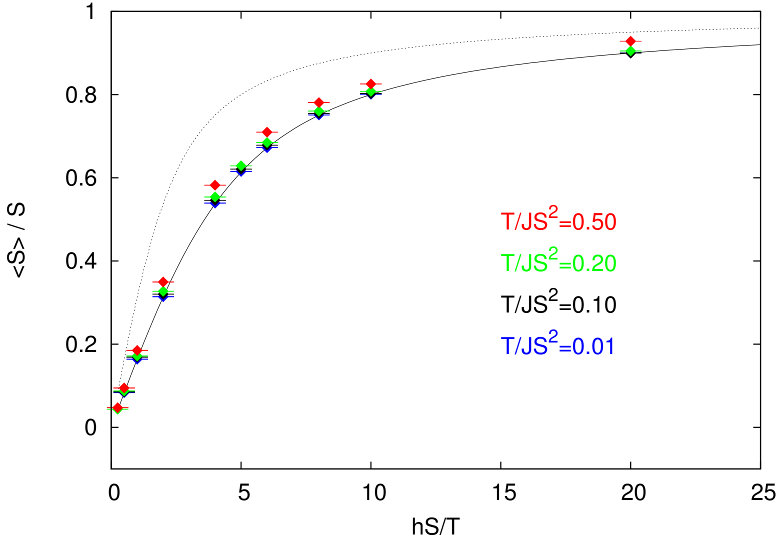
- Reproduced within effective theory (Easy to check)

Two vacancies on triangle: Orphan spin magnetization curve

- Integrate out other fields and derive magnetization curve of $S\vec{n}$ with field $\mathbf{h} = h\hat{z}$.
  For for $h \ll JS$, $T \ll JS^2$ but arbitrary hS/T, prediction:
  $S\langle n^z \rangle(h, T) = S\mathcal{B}(hS/2T)$

($S\mathcal{B}(hS/2T)$ is the classical magnetization curve of single spin S in field $h/2$)
Test: Can compare classical monte-carlo "experiment" with effective field theory prediction.

Lone spin magnetization

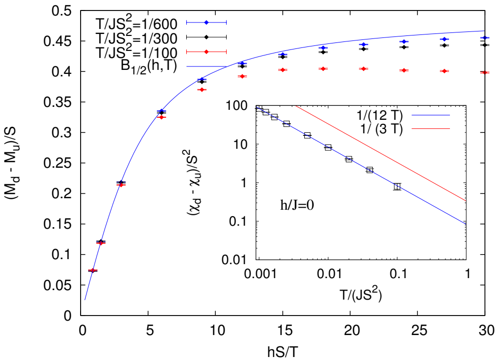


Effective theory works well at low temperature

Spin texture

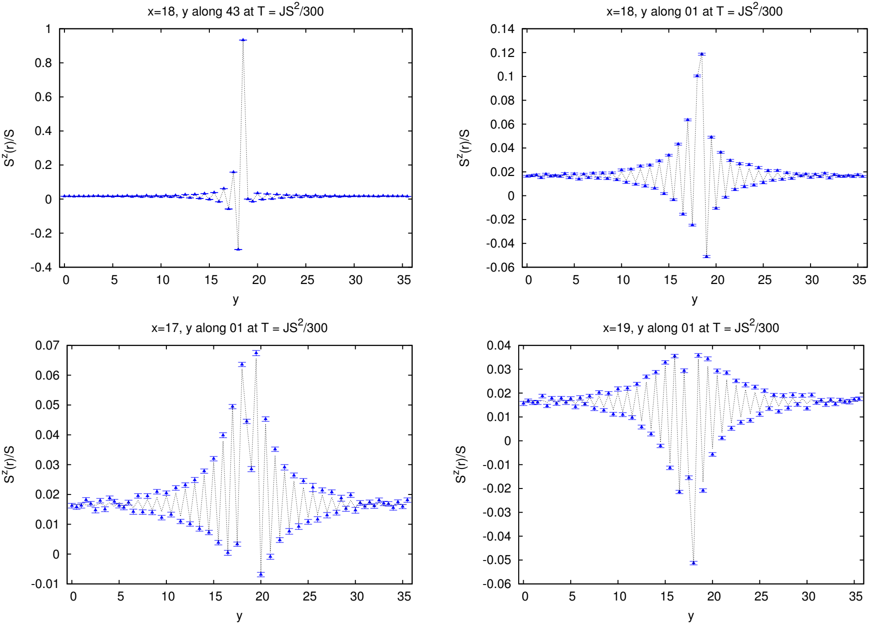
- The lone-spin polarization $S\mathcal{B}(hS/2T)$ serves as the 'source' for $\vec{\phi}_i$.
- Effective theory gives prediction for defect induced spin-texture $\langle S_i^z \rangle(h, T) = \langle \phi_i^z \rangle(h, T)$ and defect-induced impurity moment M_{imp}
- Effective theory also gives impurity susceptibility $\chi_{\rm imp} = \frac{dM_{imp}}{dh}$
  Prediction $\chi_{\rm imp} = (S/2)^2/3T$, *i.e.* fractional spin $S/2$ "really" exists!
  Can test against Monte-Carlo "experiment"

Check: Fractional spin is real



- $\chi_{\rm imp}(T)$ fits Curie law $S_{\rm eff}^2/3T$ with $S_{\rm eff} = S/2$
- Full magnetization curve of impurity-induced magnetization predicted correctly.

Spin texture: Theory vs "experiment"



Entropic interactions between orphan spins

- Tractable computation within effective field theory
- Result: Orphan spins have only two-body (bilinear) exchange interactions J_{eff}.
- Sign of J_{eff} is positive (antiferromagnetic) if two orphans are in the same Kagome layer. Else it is ferromagnetic

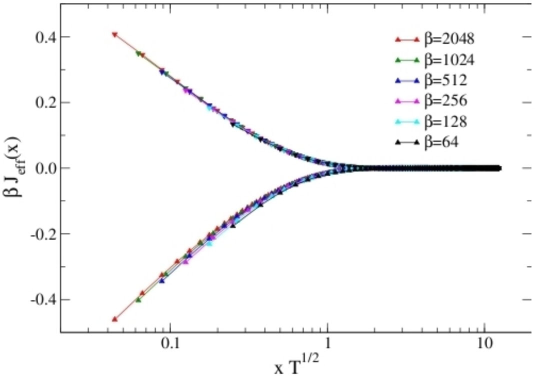
$$J_{eff}(\vec{r}_1 - \vec{r}_2, T) = \eta(\vec{r}_1)\eta(\vec{r}_2) T \mathcal{J}(\sqrt{T}(\vec{r}_1 - \vec{r}_2))$$

with

$$\begin{aligned} \mathcal{J}(\vec{y}) &\sim \log(1/|\vec{y}|) \text{ for } |\vec{y}| \ll 1 \\ \mathcal{J}(\vec{y}) &\sim \exp(-|\vec{y}|) \text{ for } |\vec{y}| \gg 1 \end{aligned}$$

Form of interaction

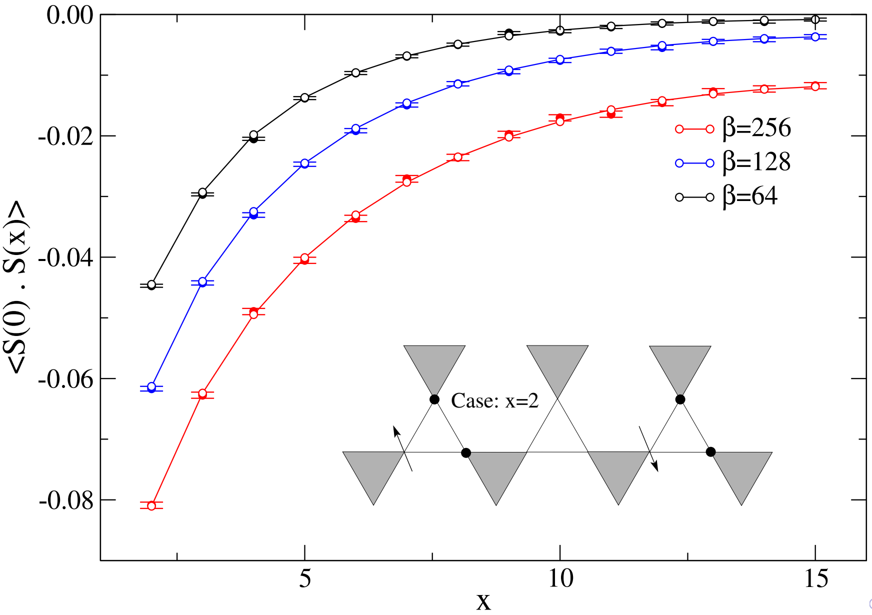
$J_{\rm eff}$ between two orphans in the same layer (upper curve) and different layers (lower curve).



Solid lines: low T scaling form.

Points: full effective field theory results

Check against Monte-Carlo simulations



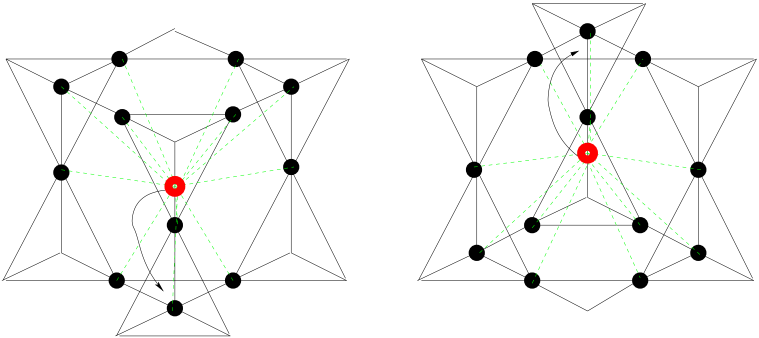
Further checks of theory

Prediction of absence of three-body and higher order terms is confirmed by monte-carlo studies of a system with three and four orphans.

Origins of NMR broadening

- Isolated vacancies have no associated Curie response.
  Cannot account for NMR line broadening $\Delta H \propto 1/T$
- At small x, NMR line broadening reflects response to defective triangles produced by vacancy-pairs

Finally: Modeling the Ga(4f) NMR line

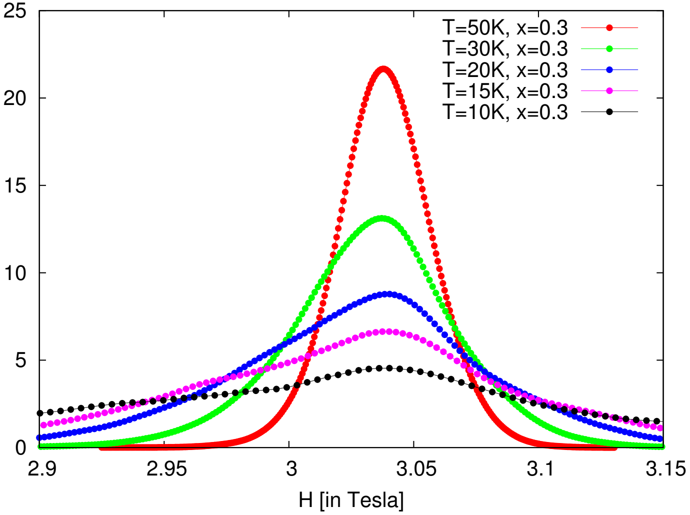


Averaging over 12 Cr spins 'loses information'

Field swept NMR line gives histogram of h satisfying $\gamma_N(h + \mathcal{A} g_L \mu_B \sum_{i \in Ga(4f)} \langle S_i^z \rangle) = \omega_{NMR}$ for each Ga(4f) nucleus in lattice

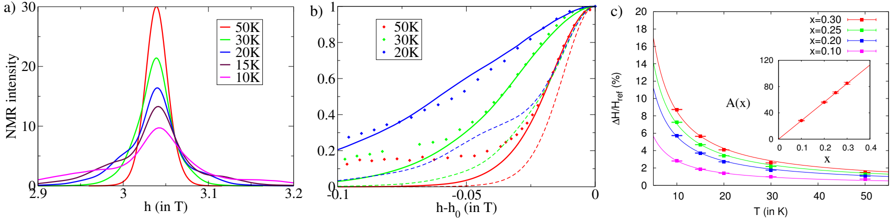
All parameters known from experiment

Ga NMR lineshape



Finite vacancy density $x = 0.3 \rightarrow$ Incorporate interactions between spin textures via Monte-Carlo simulation

Comparison with experiment



Theory ($x = 0.2$ dashed, $x = 0.3$ solid) vs experiment ($x = 0.19$ dots, Limot 2002)

$\Delta H \sim \mathcal{A}(x)/T$ captured correctly

$\mathcal{A}(x) \sim x$ for not-too-small x captured correctly(!)

But independent dilution produces too few defective triangles ($\mathcal{O}(x^2)$ for small enough x)

Verdict(?)

- Detailed understanding of the physics of spin-textures in SCGO, a spin liquid with power-law spin correlations.
- Reliable description of defect-induced fractional moments
- But: Disorder modeling too simplistic.
  Correlations between vacancies, bond-disorder...?

Outlook

Can we understand the freezing transition by thinking of a system of randomly positioned orphan spins interacting with long-range couplings?