Defect-induced fractional spins in SCGO

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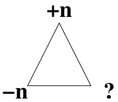
Discussions:
D. Dhar, C. Henley, & P. Mendels
Reference- Phys Rev Lett. **106**, 127203 (2011).

Frustrated magnets and spin liquids

- Antiferromagnetic exchange interactions of magnetic ions in insulators:
  $E = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j \qquad J > 0$
- When is J>0, large? Difficult (quauntum chemistry) question, with thumb-rule answer: Goodenough-Kanamori-Anderson rules
  J.B. Goodenough, *Magnetism and the Chemical Bond (1963)*
  (exceptions known, *e.g.* Oles *et. al.* 2006, Stuttgart group)
- Triangles $\rightarrow$ *Frustrated* antiferromagnetism

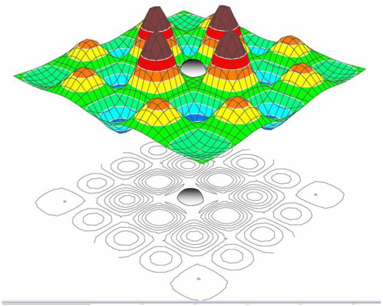


Competing interactions frustrate Neel order

Spin liquids

- 'Quenching' of exchange allows new physics to take center-stage: Spin liquids
- Macroscopic degeneracy of *classical* minimum energy configurations.
- At intermediate $T < J$, spin correlations reflect this macroscopic degeneracy:
  No Bragg peaks in structure factor $\rightarrow$ correlated liquid state

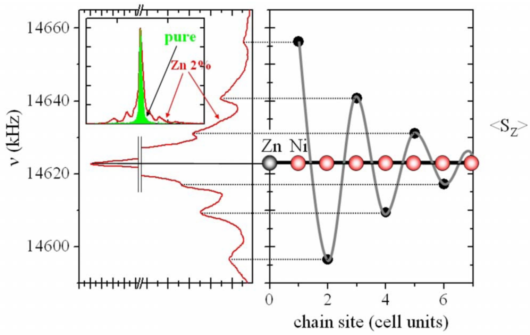
Impurities as probes



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

- Impurities can be useful probes of interesting low temperature states of matter—Zn doping in cuprates

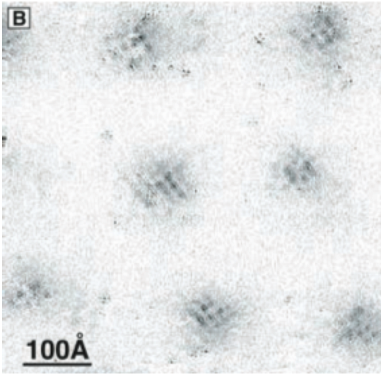
Impurities as probes



From Tedoldi et. al. 1999

- Non-magnetic impurities that cut spin-chains in quasi-1dimensional systems.

Impurities as probes



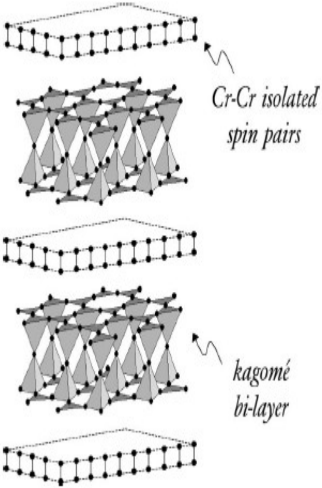
Checkerboard around vortex—from Seamus Davis group

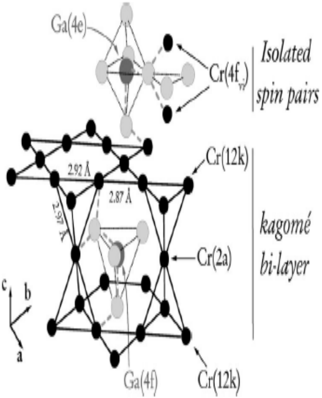
- Impurities change the state of system in immediate vicinity—Changes can be picked up by local probes such as STM
- Particularly interesting if system has 'nearby' competing ground-states Impurities can locally 'seed' a competing ground state with different ordering and symmetry properties

Our focus: SCGO

- Particularly interesting:
  Impurities can pick up ‘hidden’ correlations of the low temperature state and encode them as intricate charge/spin textures
- In this talk: Non-magnetic Ga impurities in pyrochlore slab magnet SCGO
  $\rightarrow$ Defect-induced spin textures encoding correlations of a classical spin liquid

Cast of characters: SCGO and its Galling defects





Idealized $SrCr_9Ga_3O_{19}$ unrealizable. $\rightarrow$ Instead: $SrCr_{9p}Ga_{12-9p}O_{19}$

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

Some chemistry: 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*)

What was seen?

- Broad spin liquid regime down to $T \sim \Theta_{CW}/100$ ($\Theta_{CW} \approx 500K$)
- Macroscopic susceptibility has 'defect contribution' $\chi_{def} = C_d/T$, with $C_d \propto (1-p) \equiv x$
  Attributed to 'orphan-spin population', Schiffer-Daruka (97)
- Broad, apparently symmetric Ga NMR line, 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)

Some theory: Single-unit analysis

- Correlations beyond near-neighbours can perhaps be ignored in a short-ranged spin liquid
  $\rightarrow$ Single-unit approximation.
- Defective simplices (with all but one spin removed) give Curie tail; no other simplices contribute to Curie tail.
- Identify the 'orphan population' of Schiffer and Daruka with defective simplices in diluted lattice

Moessner-Berlinksy (1999)

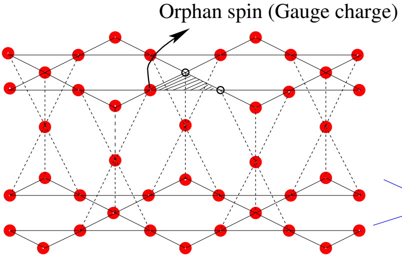
Some theory: $T = 0$ Simplex satisfaction

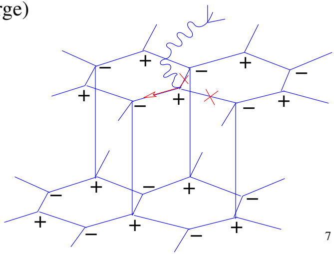
$$H = \frac{J}{2}\sum_{\boxtimes}(\sum_{i\in\boxtimes}\vec{S}_i - \frac{\mathbf{h}}{2J})^2 + \frac{J}{2}\sum_{\triangle}(\sum_{i\in\triangle}\vec{S}_i - \frac{\mathbf{h}}{2J})^2$$

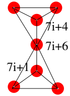
- Absolute minimum of energy is achievable:
  If no symmetry breaking: $S^z_{Kag} = h/6J$, $S^z_{apical} = 0$
  (for $\mathbf{h} = h\hat{z}$)
  Henley (2000)

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

Some theory: Half-orphans

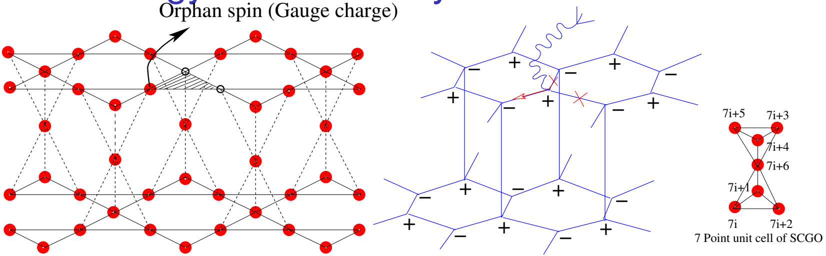






- Single Ga on any simplex $\rightarrow$ no problem with simplex satisfaction
- If two Ga in one $\triangle \rightarrow \triangle$ has only one spin
  $\langle S^z_{\mathrm{tot}} \rangle = \frac{1}{2} \sum_{\mathrm{simplices}} \langle S^z_{\mathrm{simplices}} \rangle = S/2 = 3/4!$ (at $T = 0$, $h/J \rightarrow 0$)
  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: $(\nabla \cdot \mathbf{E}^\alpha)_\triangle = S^\alpha_{\text{orphan}} \hat{e}_{\text{orphan}}$
- But $T = 0$ Gauss law $\rightarrow 1/\vec{r}$ decay of $T = 0$ induced spin-texture.

We ask: Are there “really” fractional spins at $T > 0$?

Simplex satisfaction *a la* Henley is inherently a $T = 0$ statement

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 $Z \propto \int \mathcal{D}\vec{\phi} \exp(-\mathcal{F}/T)$
  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 $\mathcal{S} \propto \left(-\frac{\rho_1}{2} \sum_{i \in \text{Kagome}} \vec{\phi}_i^2 - \frac{\rho_2}{2} \sum_{i \in \text{apical}} \vec{\phi}_i^2\right)$

ρ_1 and ρ_2 phenomenological parameters

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

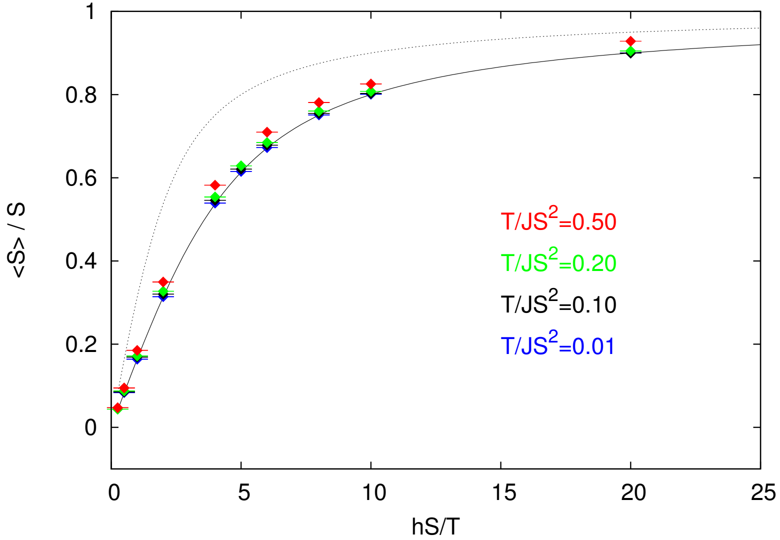
(Gaussian theory$\rightarrow$Independent effective action for each spin component)

Modeling the half-orphans in effective field theory

- Ga substitution implies constraint
  $\vec{\phi}_{\rm Ga} = 0$
- 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).
- 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

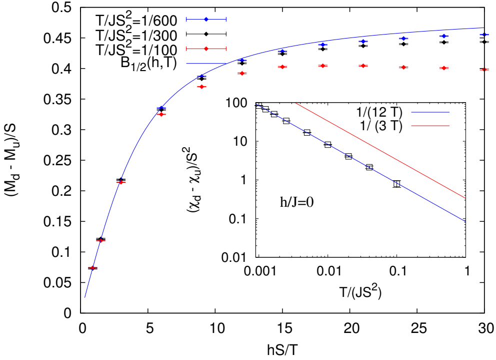


Effective theory works well at low temperature

Spin texture

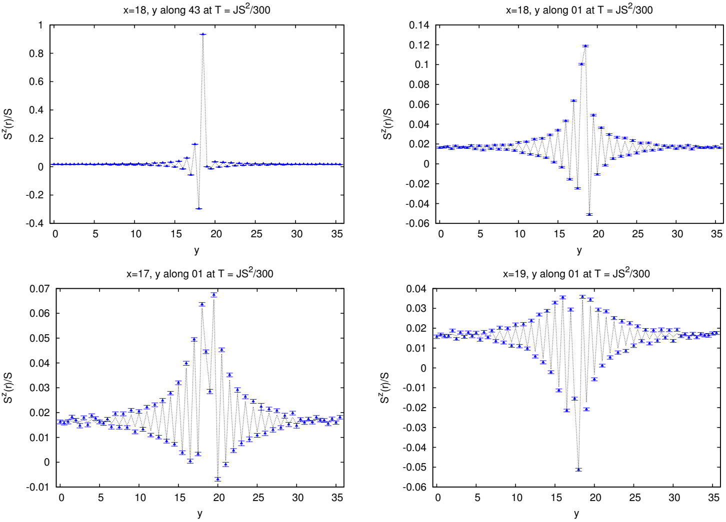
- The lone-spin polarization $S\mathcal{B}(hS/2T)$ serves as the 'source' for $\vec{\phi}_i$.
- Effective theory gives prediction for defect induced spin-texture $\langle S_i^z\rangle(h,T) = \langle \phi_i^z\rangle(h,T)$ and defect-induced impurity moment M_{imp}
- Effective theory also gives impurity 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



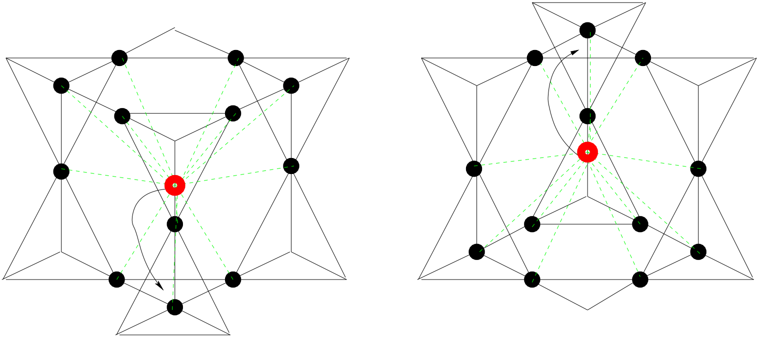
- $\chi_{\mathrm{imp}}(T)$ fits Curie law $S_{\mathrm{eff}}^2/3T$ with $S_{\mathrm{eff}} = S/2$
- Full magnetization curve of impurity-induced magnetization predicted correctly.

Check: Intricate spin texture



Simple effective theory works well

From texture to Ga(4f) NMR line

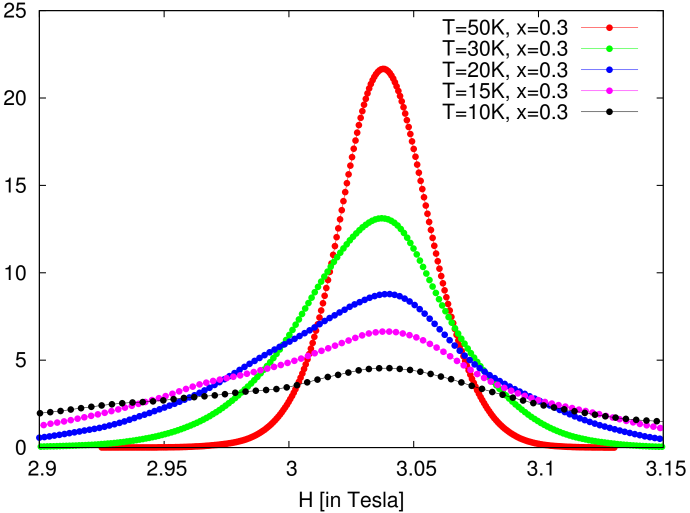


Averaging over 12 Cr spins ‘loses information’

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

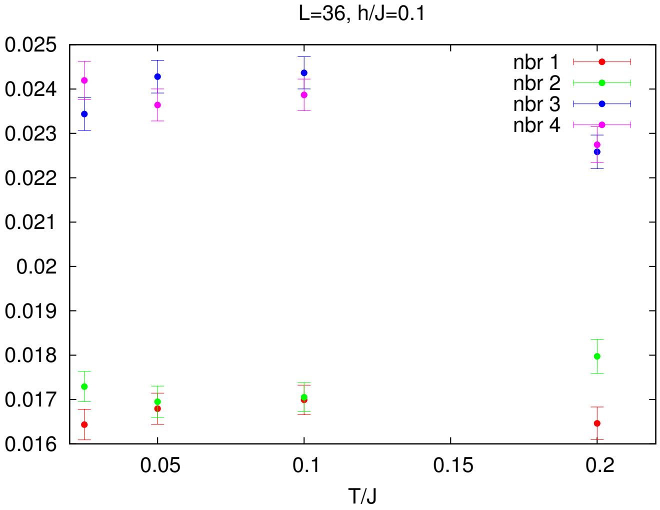
All parameters known from experiment

Ga NMR lineshape



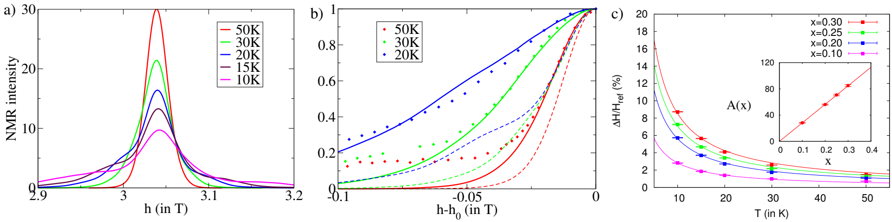
Finite vacancy density $x = 0.3$→Incorporate interactions between spin textures via Monte-Carlo simulation

Isolated vacancies to not contribute to Curie term



Isolated vacancies cannot account for $\Delta H \propto 1/T$

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.
- Reliable description of defect-induced fractional moments
- But: Disorder modeling too simplistic.
  Correlations between vacancies, bond-disorder...?

The road ahead: Natural ‘spin’-offs.

Entropic interactions between ‘orphan’ spins
Collective behaviour of finite density of ‘orphan’ textures
Glassy low temperature state?

Uphill task: Putting in quantum effects (with S. Sanyal)

Mean field theory
Series expansions
Mapping to “Kondo” physics (???)

Support

- Computational resources of TIFR.
- DST (India) grant funding.
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