Defect-induced fractional spins in SCGO

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Impurities yield information about host materials

- Impurities can be useful probes of interesting low temperature states of matter
  Alloul et. al. Rev. Mod. Phys. **81**, 45 (2009).
  e.g Zn and Ni doping in CuO_2 planes of high-T_c superconductors
  non-magnetic impurities that cut spin-chains in quasi-1dimensional systems.
- Impurities change the state of system in immediate vicinity
  Changes can be picked up by local probes such as NMR

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
  → Defect-induced spin textures encoding correlations of a classical spin liquid

Cast of characters: SCGO and its Galling defects

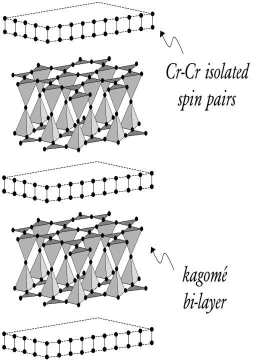


FIG. 1: The magnetic lattice of SCGO is a stacking of kagomé bi-layers of Cr^{3+} ions ($\approx 78\%$ of the Cr sites), separated by Cr-Cr isolated spin pairs ($\approx 22\%$ of the Cr sites).

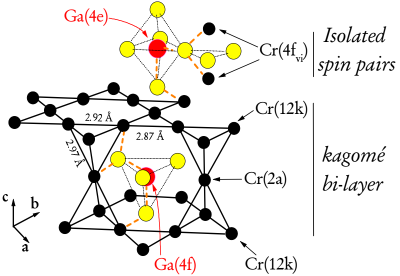


FIG. 2: The crystal structure of ideal $SrCr_9Ga_3O_{19}$. The light grey circles represent the oxygen atoms (we have restrained their number for clarity). The Sr atoms are not represented. The thick dashed lines show the typical hyperfine coupling paths of the gallium nuclei to various Cr sites through the oxygen ions.

Fig. from Limot et al PRB (2002)

S=3/2 moments nearly classical
p=0.95 for best samples

Idealized $SrCr_9Ga_3O_{19}$ unrealizable. $\longrightarrow$ 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 $C_d \propto 1 - p$
  Attributed to 'orphan-spin population', Schiffer-Daruka (97)
- Broad, apparently symmetric Ga NMR line, with broadening $\Delta H \propto 1/T$
  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)

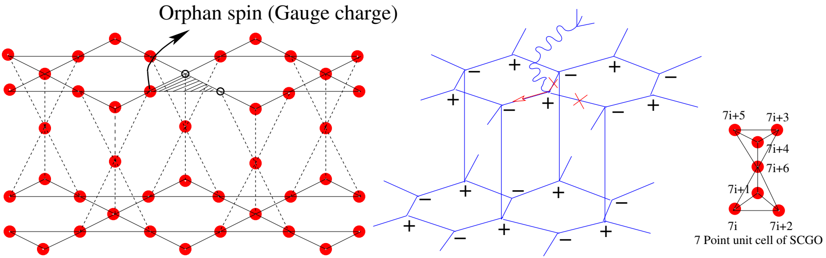
Some theory: $T = 0$ Simplex satisfaction

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

- 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)

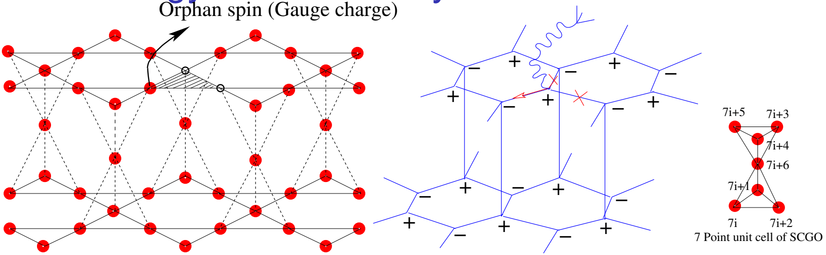
Relies on constructing states that also satisfy $\vec{S}_i^2 = S^2$ for h not-to-large.

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:

- Intuition for $T \ll J$ (from large-N theory)
  Entropy $\mathcal{S} \propto \left(-\frac{\rho_1}{2}\sum_{i\in\mathrm{Kagome}}\vec{\phi}_i^2 - \frac{\rho_2}{2}\sum_{i\in\mathrm{apical}}\vec{\phi}_i^2\right)$
- 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
  $E = \frac{J}{2}\sum_{\boxtimes}(\sum_{i\in\boxtimes}\vec{\phi}_i - \frac{\mathbf{h}}{2J})^2 + \frac{J}{2}\sum_{\triangle}(\sum_{i\in\triangle}\vec{\phi}_i - \frac{\mathbf{h}}{2J})^2$

ρ_1 and ρ_2 phenomenological parameters

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

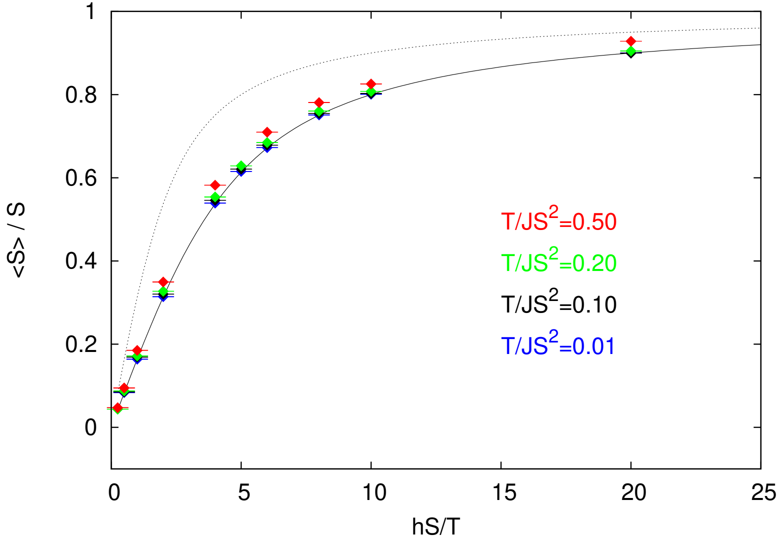
(Gaussian theory→Independent effective action for each spin component)

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

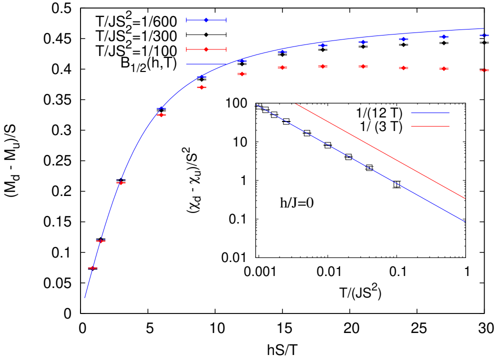


Effective theory works well at low temperature

Spin texture

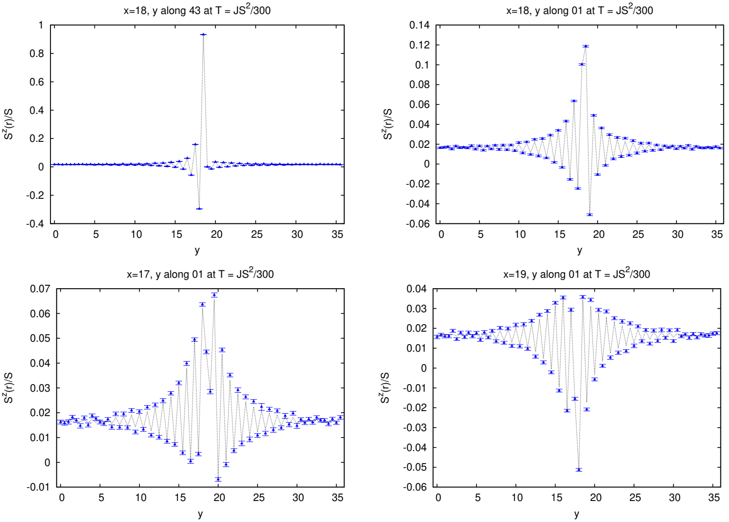
- The lone-spin polarization $S\mathcal{B}(hS/2T)$ serves as the 'source' for $\vec{\phi}_i$.
- Effective theory gives prediction for defect induced spin-texture $\langle S_i^z \rangle(h,T) = \langle \phi_i^z \rangle(h,T)$ and defect-induced impurity moment M_{imp}
- Effective theory also gives impurity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



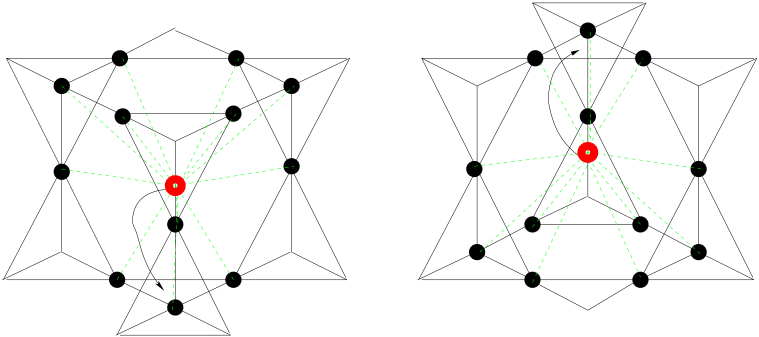
- $\chi_{\rm imp}(T)$ fits Curie law $S_{\rm eff}^2/3T$ with $S_{\rm eff} = S/2$
- Full magnetization curve of impurity-induced magnetization predicted correctly.

Check: Intricate spin texture



Simple effective theory works well

From texture to Ga(4f) NMR line

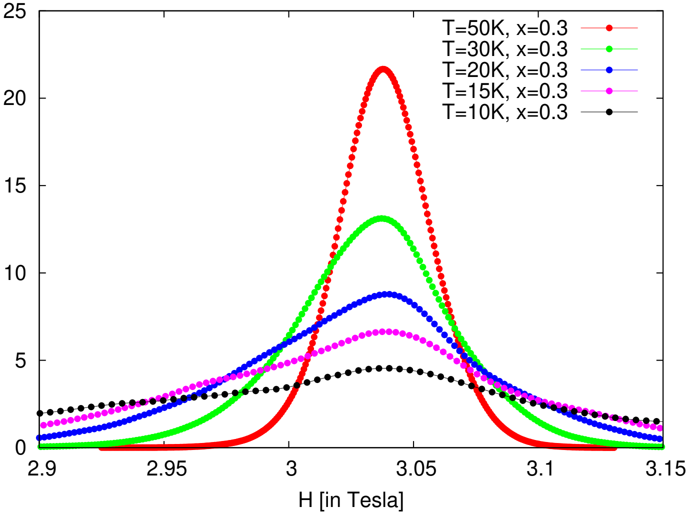


Averaging over 12 Cr spins 'loses information'

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

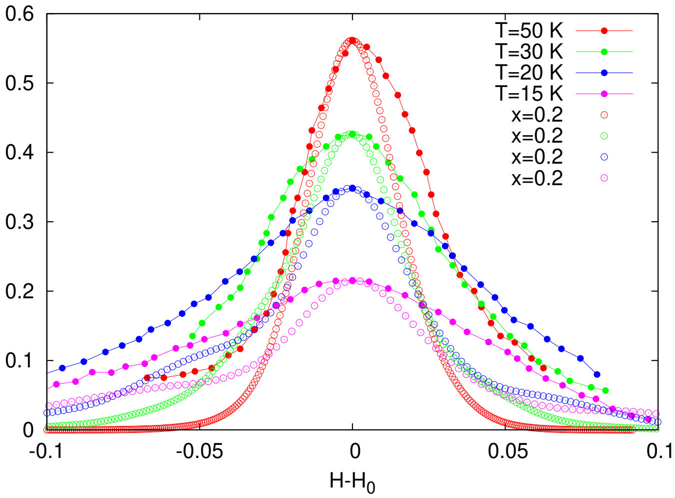
All parameters known from experiment

Ga NMR lineshape



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

Comparison with experiment



Theory ($p = 0.8$) vs experiment ($p = 0.81$, Limot 2002)

$\Delta H \sim 1/T$ captured correctly

But independent dilution produces too few defective triangles

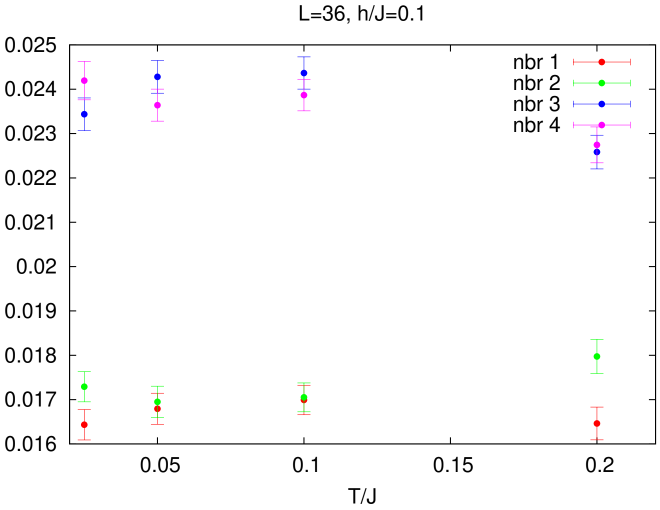
Verdict(?)

- 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...?

Support

- Computational resources of TIFR.
- DST (India) grant funding.
- Fell-Fund (Oxford), ICTS (Mumbai), ARCUS (Orsay), and MPIPKS (Dresden) support for collaborative visits.

Single vacancy



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