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Fractional spin textures and their interactions in a classical spin liquid

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ICTP2012: Innovations in Strongly Correlated Electron Systems

Anatomy and behaviour of a (dys)functional material: $SrCr_{9p}Ga_{12-9p}O_{19}$

A. Sen (TIFR ☞BU ☞MPIPKS) & R. Moessner (Oxford ☞MPIPKS)

Ref- PRL. **106**, 127203 (2011) & arXiv:1204.4970

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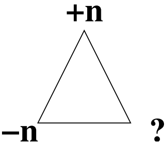
Antiferromagnetism in Mott insulators:

- Antiferromagnetic exchange interactions of magnetic ions in insulators:
  $E = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j \quad J > 0$
- 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)
- Sometimes possible to "measure" *J*: Inelastic neutron scattering in high field.
  e.g. $Yb_2Ti_2O_7$ Ross *et al.* PRX 2011

Triangles on my mind: Frustration and spin liquid behaviour

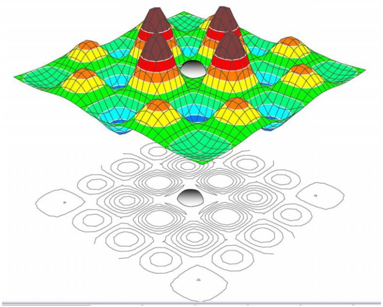
- Triangles $\rightarrow$ *frustrated* antiferromagnetism



  Competing interactions frustrate Neel order

- ‘Quenching’ of exchange allows new physics to take center-stage: Spin liquids
- Macroscopic degeneracy of *classical* minimum energy configurations.
- At intermediate $T_f < T < JS^2$, spin correlations reflect this macroscopic degeneracy:
  No Bragg peaks in structure factor $\rightarrow$ correlated liquid state

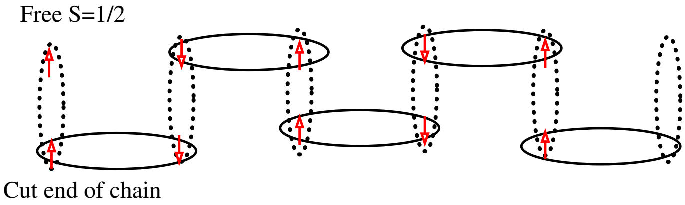
Impurities as probes



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

- 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

Impurities as probes: “Cutting” a Haldane-gapped chain



- Cut-end of $S = 1$ AF chain hosts free $S = 1/2$ moment
- Characteristic of “topological order” in Haldane state

Impurities as probes: Probing cut chains with NMR

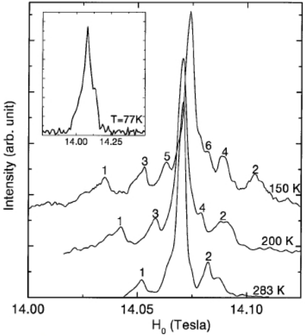
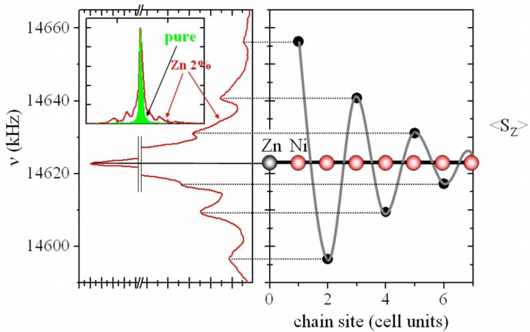


FIG. 1. ^{89}Y NMR spectra in $Y_2BaNi_{0.95}Mg_{0.05}O_5$ recorded at fixed frequency $\nu_{RF} = 29.4$ MHz by sweeping magnetic field. Resolved satellite peaks are labeled with the index I, following the decreasing magnitude of their shift (measured from the central line). In the inset, all of the peaks are shown to be smeared in a single wide line when the temperature is lowered.

Tedoldi *et. al.* 1999

- Non-magnetic Mg^{2+} impurities in $S = 1$ (Ni^{2+}) chain Y_2BaNiO_5 cut chain.
- ^{89}Y NMR (Knight-shift)
  Snapshot of free $S = 1/2$ moments localized near cut end

Probing cut chains with NMR—II



Das *et. al.* 1999

- More quantitative, lower temperature studies—comparison against QMC data possible

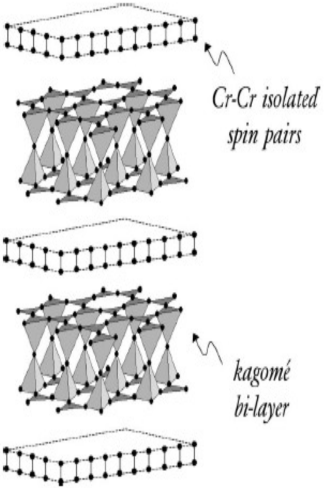
General idea

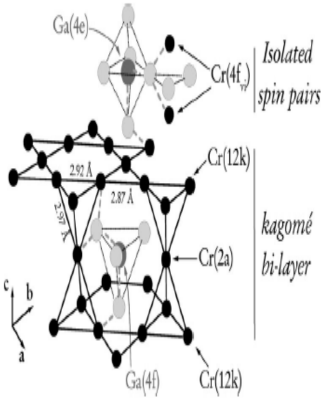
- Impurities disturb the system locally
  Host response characteristic of correlations of the low temperature state
- Correlations encoded in intricate charge/spin textures seeded by impurities
- Picked up by local probes like NMR and STM

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 4*f* sites
  Break isolated dimers
- Close runners-up are 12*k* sites
  And substitute into upper or lower Kagome layers
- Significantly lower probability of going to the 2*a* 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 $T < T_g$ 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?

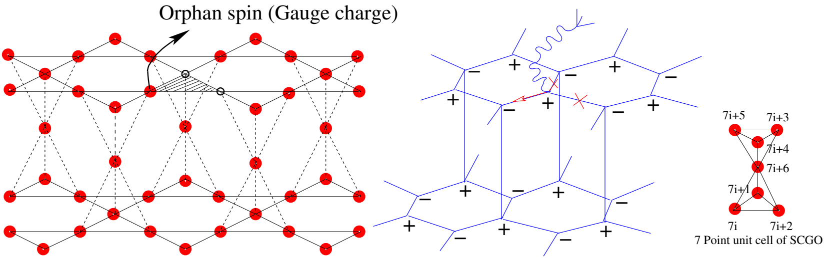
Some theory: $T = 0$ Simplex satisfaction

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

- 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)

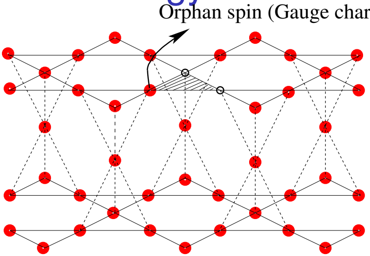
Relies on constructing states that also satisfy $\vec{S}_i^2 = S^2$ for h not-to-large.

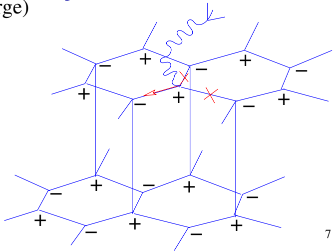
Some theory: Half-orphans

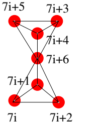


- Single Ga on any simplex $\rightarrow$ no problem with simplex satisfaction
- If two Ga in one $\triangle \rightarrow \triangle$ has only one spin
  $\langle S^z_{\text{tot}} \rangle = \frac{1}{2} \sum_{\text{simplices}} \langle S^z_{\text{simplices}} \rangle = S/2 = 3/4!$ (at $T = 0$, $h/J \rightarrow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

Aside: Analogy with electrodynamics







7 Point unit cell of SCGO

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

- $\mathbf{E}_i^\alpha = S_i^\alpha \hat{e}_i$,
  (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
But: curious property of a single tetrahedron/triangle

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

Real question: What about correlations (long-range) between simplices?

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 $\langle\vec{\phi}_i^2\rangle = S^2$

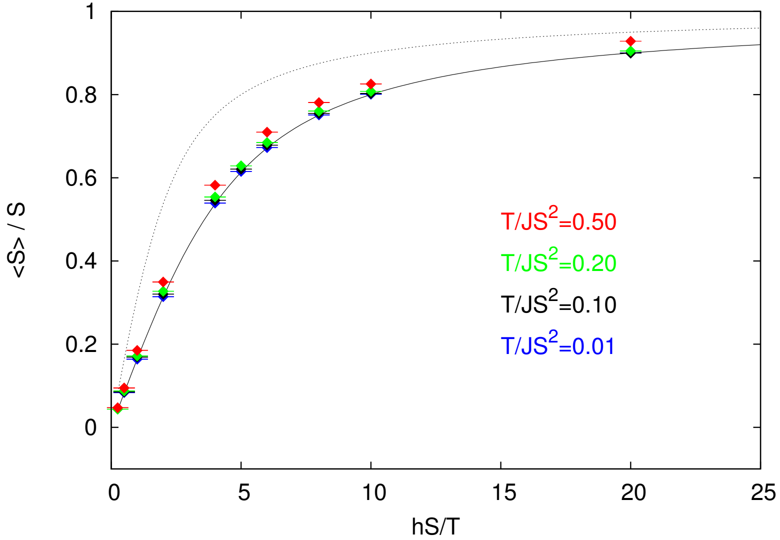
(Gaussian theory$\rightarrow$Independent effective action for each spin component)

Modeling the half-orphans in effective field theory

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

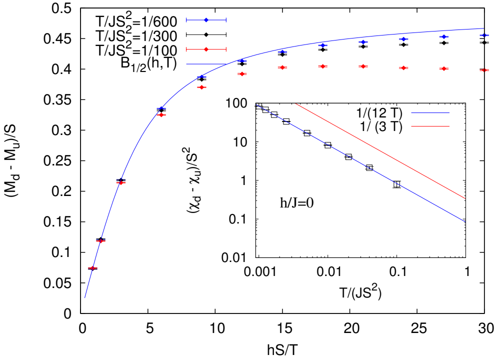


Effective theory works well at low temperature

Spin texture

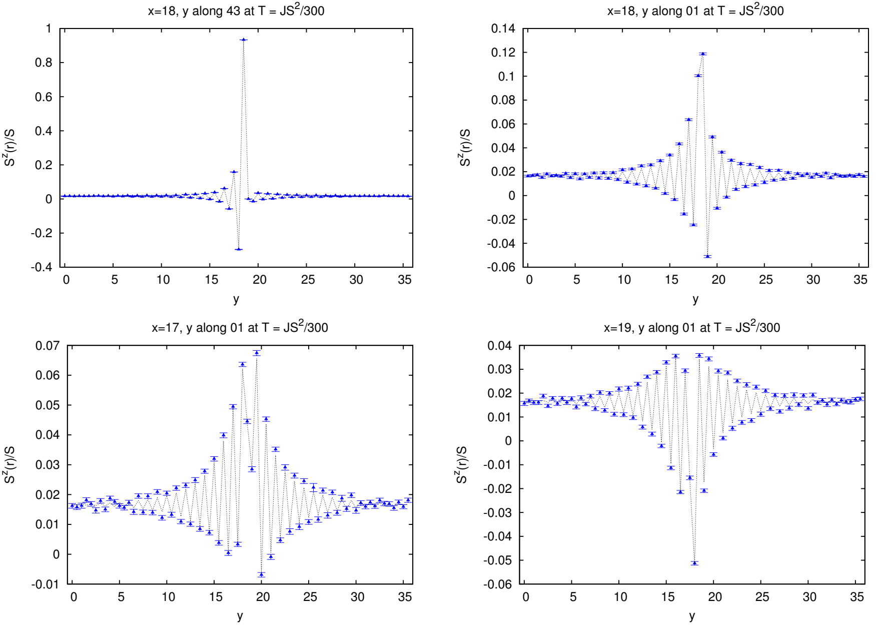
- The lone-spin polarization $S\mathcal{B}(hS/2T)$ serves as the 'source' for $\vec{\phi}_i$.
- Effective theory gives prediction for defect induced spin-texture $\langle S_i^z \rangle(h, T) = \langle \phi_i^z \rangle(h, T)$ and defect-induced impurity moment M_{imp}
- Effective theory also gives impurity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

Check: Fractional spin is real



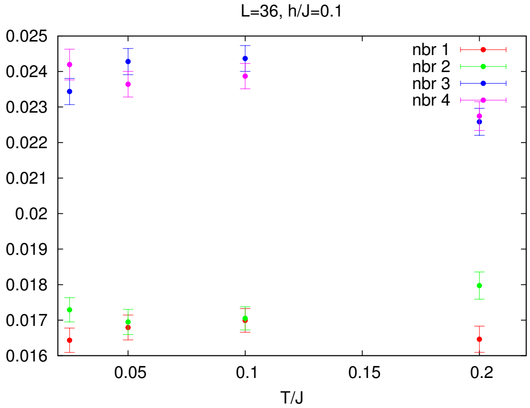
- $\chi_{\rm imp}(T)$ fits Curie law $S_{\rm eff}^2/3T$ with $S_{\rm eff} = S/2$
- Full magnetization curve of impurity-induced magnetization predicted correctly.

Spin texture: Theory vs "experiment"



Isolated vacancies to not contribute to Curie term

Susceptibility of sites around a single missing spin



- 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

Entropic interactions between orphan spins

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

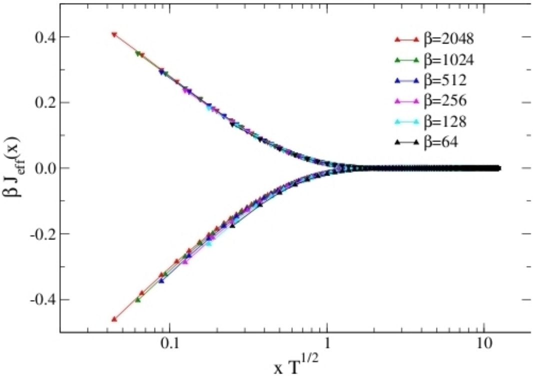
$$J_{eff}(\vec{r}_1 - \vec{r}_2, T) \quad = \quad \eta(\vec{r}_1)\eta(\vec{r}_2) T \mathcal{J}(\sqrt{T}(\vec{r}_1 - \vec{r}_2))$$

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

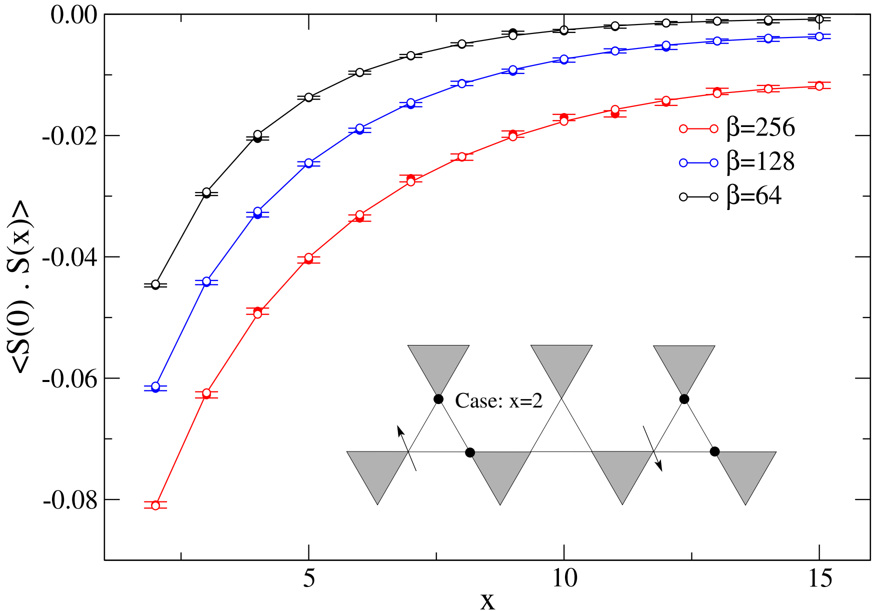
$J_{\rm eff}$ between two orphans in the same layer (upper curve) and different layers (lower curve).



Solid lines: low *T* scaling form.

Points: full effective field theory results

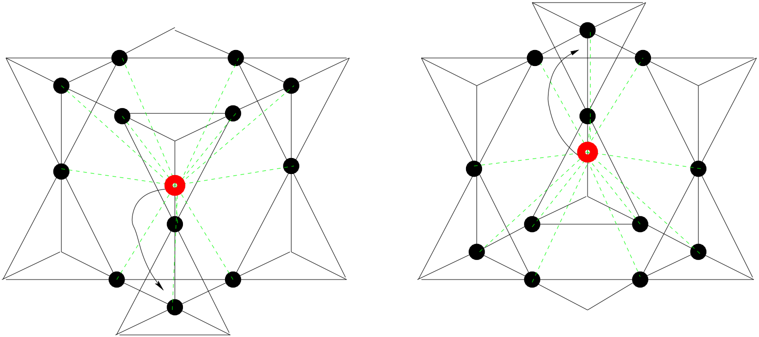
Check against Monte-Carlo simulations



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.

Finally: Modeling the Ga(4f) NMR line

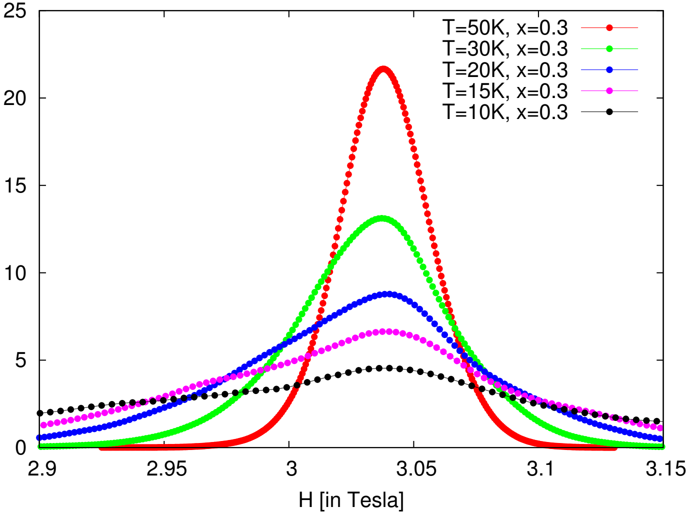


Averaging over 12 Cr spins 'loses information'

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

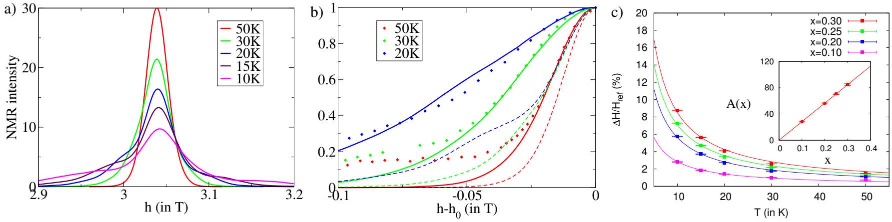
All parameters known from experiment

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?

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  Mumbai ☞Dresden: MPIPKS