Fractional spin textures and their interactions in $SrCr_{9p}Ga_{12-9p}O_{19}$ (SCGO)

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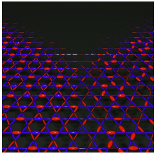

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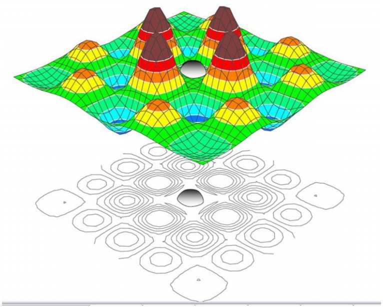
Impurities as probes



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

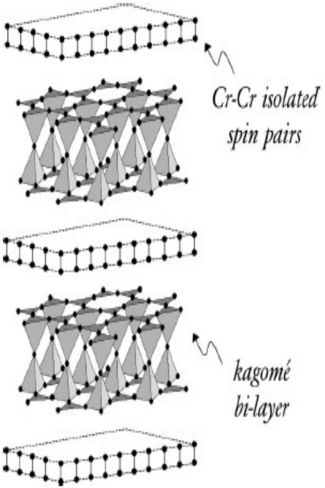
General idea

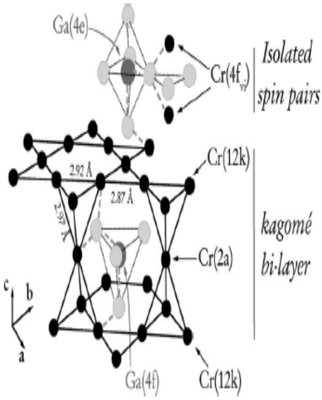
- Impurities disturb the system locally
  Host response characteristic of correlations of the low temperature state
- Correlations encoded in intricate charge/spin textures seeded by impurities
- Picked up by local probes like NMR and STM

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 $p_{max} \approx 0.95$

$J_{\text{bilayer}} \approx 80K$ $J_{\text{dimers}} \approx 200K$ *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 $T < T_f$ 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?

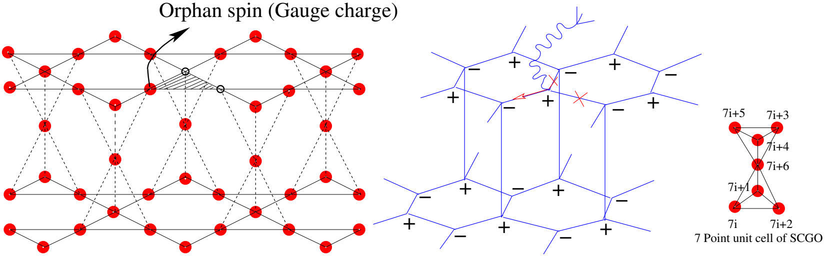
Some theory: $T = 0$ Simplex satisfaction

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

- 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)

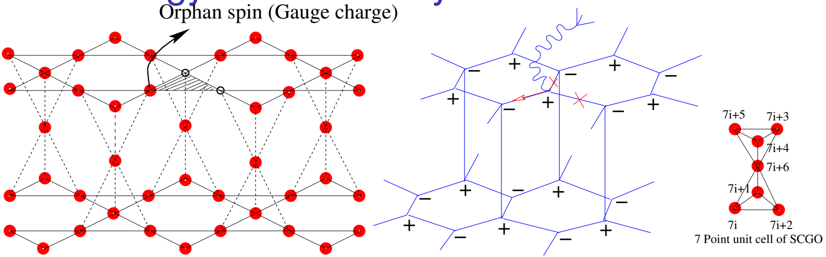
Relies on constructing states that also satisfy $\vec{S}_i^2 = S^2$ for h not-to-large.

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
  $\langle S^z_{\text{tot}} \rangle = \frac{1}{2} \sum_{\text{simplices}} \langle S^z_{\text{simplices}} \rangle = S/2 = 3/4!$ (at $T = 0$, $h/J \rightarrow 0$)
  Half-Orphan spins
  Henley (2000)

Aside: Analogy with electrodynamics



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

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

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

Lone-spins on defective triangles/tetrahedra:

$$\delta(\phi_{\vec{r}}^{\alpha} - Sn_{\vec{r}}^{\alpha}) = \frac{1}{2\pi}\int d\mu_{\vec{r}}^{\alpha}\exp(i\mu_{\vec{r}}^{\alpha}(\phi_{\vec{r}}^{\alpha} - Sn_{\vec{r}}^{\alpha}))$$

Combined notation:

$$\Lambda_{\vec{r}}^{\alpha} = \delta_{\vec{r},\vec{r}_v}\lambda_{\vec{r}_v}^{\alpha} + \delta_{\vec{r},\vec{r}_o}\mu_{\vec{r}_o}^{\alpha}$$

Action for μ, λ, $\vec{n}$

$$
\begin{aligned}
Z_{\text{eff}} \quad \propto \quad & \int \mathcal{D}\vec{n} \int \mathcal{D}\vec{\lambda} \int \mathcal{D}\vec{\mu} \\
& \exp\left(+ \frac{1}{2} \sum_{\vec{r}\vec{r}'\,\alpha} (\beta h^{\alpha} + i\Lambda^{\alpha}_{\vec{r}}) C_{\vec{r}\vec{r}'} (\beta h^{\alpha} + i\Lambda^{\alpha}_{\vec{r}'}) - i \sum_{\vec{r}_o \alpha} \mu^{\alpha}_{\vec{r}_o} n^{\alpha}_{\vec{r}_o} \right)
\end{aligned}
$$

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

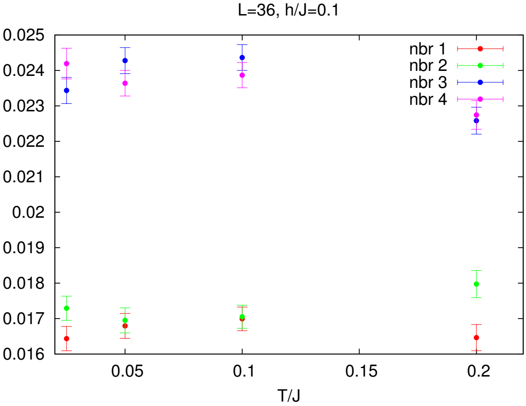
$$
\langle \phi^{\alpha}_{\vec{r}} \phi^{\beta}_{\vec{r}'} \rangle \quad \equiv \quad C_{\vec{r}\vec{r}'} \delta_{\alpha\beta}
$$

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



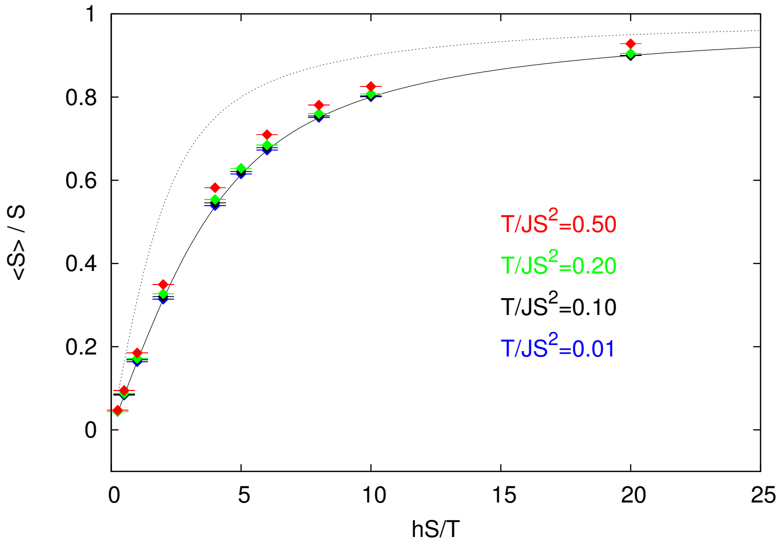
- Reproduced within effective theory (Easy to check)

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

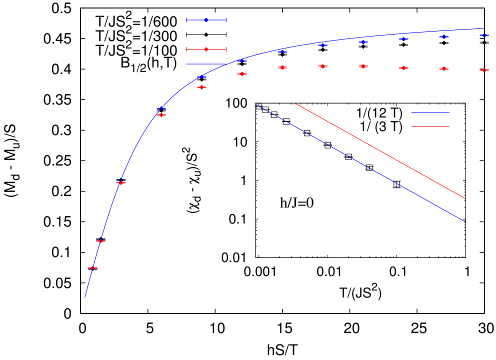


Effective theory works well at low temperature

Spin texture

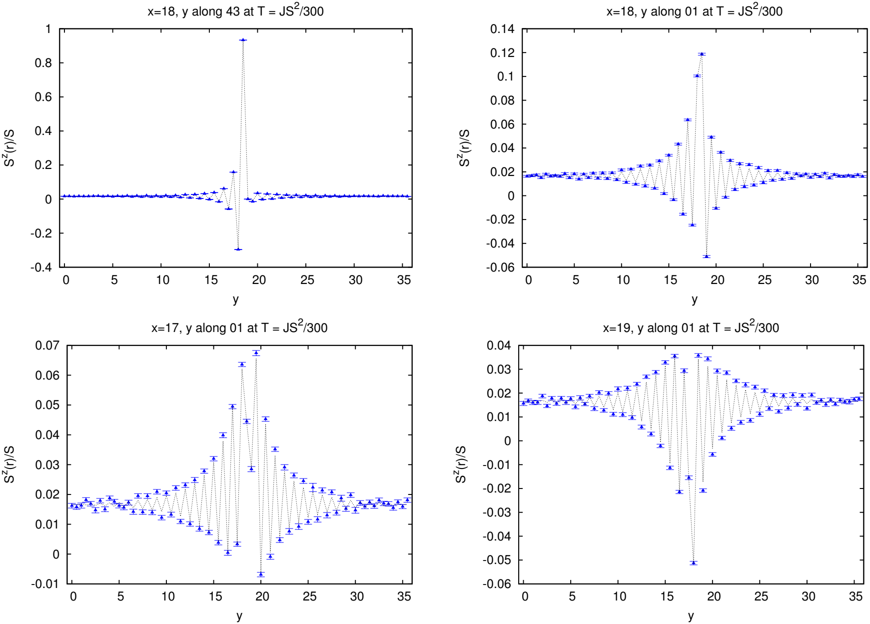
- The lone-spin polarization $S\mathcal{B}(hS/2T)$ serves as the 'source' for $\vec{\phi}_i$.
- Effective theory gives prediction for defect induced spin-texture $\langle S_i^z \rangle(h, T) = \langle \phi_i^z \rangle(h, T)$ and defect-induced impurity moment M_{imp}
- Effective theory also gives impurity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



- $\chi_{\text{imp}}(T)$ fits Curie law $S_{\text{eff}}^2/3T$ with $S_{\text{eff}} = S/2$
- 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 $J_{\rm eff}$.
- Sign of $J_{\rm eff}$ is positive (antiferromagnetic) if two orphans are in the same Kagome layer. Else it is ferromagnetic

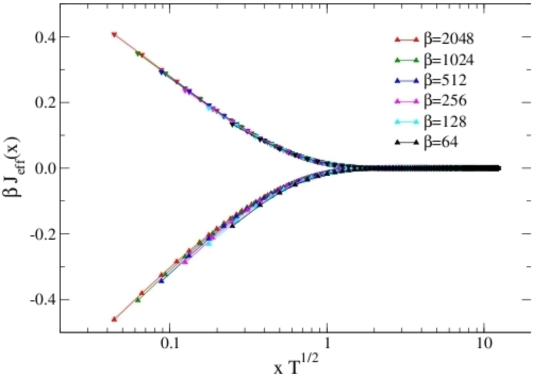
$$J_{eff}(\vec{r}_1 - \vec{r}_2, T) \quad = \quad \eta(\vec{r}_1)\eta(\vec{r}_2) T \mathcal{J}(\sqrt{T}(\vec{r}_1 - \vec{r}_2))$$

with

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

Form of interaction

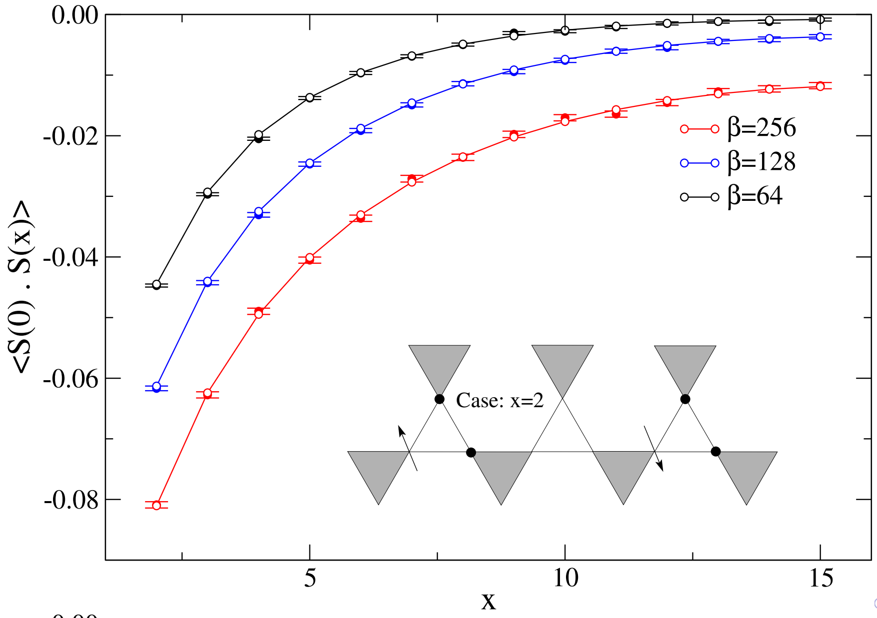
J_{eff} between two orphans in the same layer (upper curve) and different layers (lower curve).



Solid lines: low T scaling form.

Points: full effective field theory results

Check against Monte-Carlo simulations



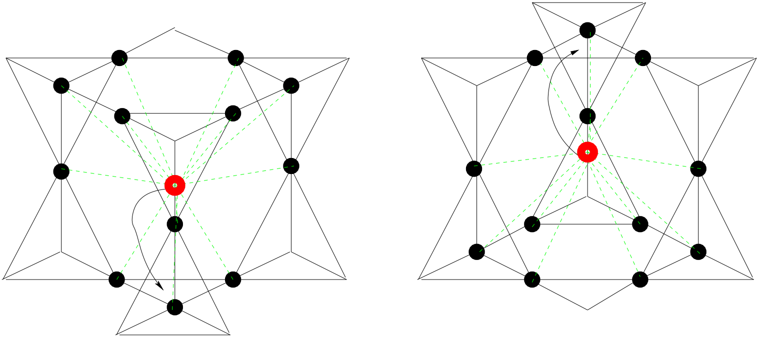
Further checks of theory

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

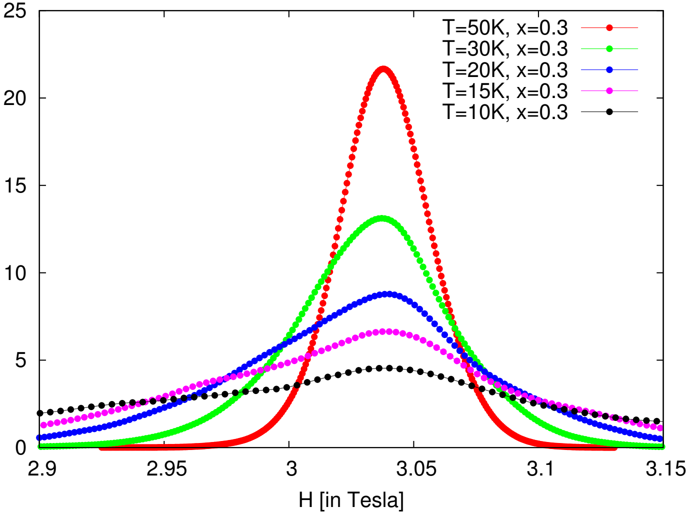


Averaging over 12 Cr spins 'loses information'

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

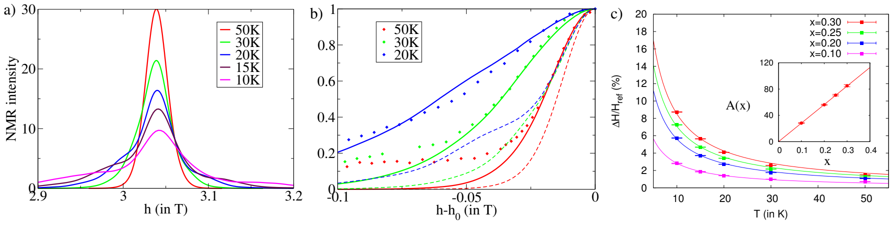
All parameters known from experiment

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?

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  Mumbai ☞Dresden: MPIPKS