Disorder in a classical spin liquid: topological spin glass, cluster algorithm and dynamical Griffiths phase

Arnab Sen Department of Theoretical Physics, Indian Association for the Cultivation of Science, Kolkata Novel Phases of Quantum Matter, ICTS

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#### Collaborators:

#### Tushar Kanti Bose (IACS, Kolkata) Roderich Moessner (MPIPKS, Dresden)





#### also thanks to

Shivaji Sondhi (Princeton, USA) Vojtech Kaiser (MPI-CBG, Germany) Reference:

Bose, Moessner and Sen, Phys. Rev. B **100**, 064425 (2019), Sen and Moessner, Phys. Rev. Lett. **114**, 247207 (2015), and ongoing work

### What is a spin liquid?

Spin solid — e.g. A ferromagnet or an antiferromagnet



- Fig shows dispersion in La<sub>2</sub>CuO<sub>4</sub> [from Coldea et. al, PRL 86, 5377 (2000)] –shows existence of magnons
- Spin gas e.g. an uncorrelated paramagnet
- Spin liquid—strongly "correlated" paramagnet

### Phases of matter with fractionalized excitations



- These break no symmetries of the underlying Hamiltonian but possess *completely different* excitations from conventional ordered states.
- $H = J \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j$  with S = 1/2 spins in 1D
- Excitations not magnons (S = 1) but spinons (S = 1/2)

### (Weak) Diagnostics



- No magnetic ordering even well below |⊖<sub>CW</sub>| unlike unfrustrated magnets
- $T_f \ll T \ll |\Theta_{CW}|$ —spin liquid regime (Ramirez)

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#### Glassiness in spin liquids?

SPIN DYNAMICS IN THE  $S = \frac{1}{3}$  QUANTUM KAGOME ....



PHYSICAL REVIEW B 83, 180416(R) (2011)

#### Spin dynamics in the $S = \frac{1}{2}$ quantum kagome compound vesignieite, $Cu_3Ba(VO_5H)_2$

R. H. Colman, <sup>1</sup>. E Bert, <sup>2</sup>. D. Boldrin, <sup>1</sup> A. D. Hillier, <sup>3</sup> P. Manuel, <sup>3</sup> P. Mendels,<sup>3</sup> and A. S. Wills,<sup>1,\*</sup> <sup>1</sup>Department of Chemistry, University College London, 20 Gondon Street, London, WCHH 0MJ, United Kingdom <sup>2</sup>Laboratione de Physique des Solides, UMR CNRS 802, Université Patris-Sud, 94/43 Orono, France <sup>3</sup>ISIS Faculty, STFC, Rutherford Appleton Laboratory, Chilton, Ofondaire 0X11 0QX, United Kingdom (Received 20 January 2011; reside annascript received 13 April 2011; Junibiasd 31 May 2011)

We report the study of high-quality samples of the frustrated  $S = \frac{1}{2}$  kagome antiferromagnet vesignitie. CubBaVO-HJy, Neuton provder diffraction measurement veidence the scalence of the kagome latice and show no sign of a transition to magnetic long-range order. A kink in the susceptibility below T = 9K is matched to a reduction in paramagnetic-like correlations in the diffraction data and a slowing of the spin dynamical and small forzem moments  $\sim 0.1 \mu_B$ . We propose that this novel quantum ground state is stabilized by a large Dzyalobihs/N-bydray interaction.

- Exp in Cu<sub>3</sub>Ba(VO<sub>5</sub>H)<sub>2</sub> which is S = 1/2 kagome antiferromagnet.
- SrCr<sub>9p</sub>Ga<sub>12-9p</sub>O<sub>19</sub> (SCGO) has a spin-glass transition at  $T_g \approx 3.5$  K but persistent spin dynamics down to 100 mK (from  $\mu$ SR probes)
- No sign of transition to magnetic long range order
- Coexistence of both dynamical (for majority of spins) and small frozen moments, no phase seperation

### Spin ice



 $H = rac{J}{3} \sum_{\langle ij \rangle} S_i S_j + Da^3 \sum_{(ij)} \left( rac{\hat{e}_i \cdot \hat{e}_j}{|r_{ij}|^3} - rac{3(\hat{e}_i \cdot \mathbf{r}_{ij})(\hat{e}_j \cdot \mathbf{r}_{ij})}{|r_{ij}|^5} 
ight) S_i S_j$ 

 Experimentally relevant: Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>, Ho<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>, Er<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>, SrCr<sub>8</sub>Ga<sub>4</sub>O<sub>19</sub>

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#### Short and long loops



- $S(T=0) \approx k_B \ln \left(2^N \left(\frac{6}{16}^{N/2}\right)\right) = \frac{N}{2} k_B \ln \left(\frac{3}{2}\right)$  [Pauling, 1935]
- Power-law spin correlations gives rise to pinch points in momentum space [Isakov, Gregor, Moessner, Sondhi (2004), Henley (2005)]

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#### Spin ice and Dumbbell model

- Represent the "point" dipoles by dumbbells that live on the ends of the dual diamond lattice.
- $H_d = \frac{2\sqrt{2}}{3\sqrt{3}} Da_d \sum_{i>j} \frac{Q_i Q_j}{r_{ij}} + \Delta \sum_i (Q_i/2)^2$  where  $Q_i = \eta_i (S_{\boxtimes})_i$ and  $\Delta = \frac{2J}{3} + \frac{8}{3} \left(1 + \sqrt{23}\right) D$

(Castelnovo, Moessner and Sondhi (2008))



### Pinch points at low T

Arnab Sen, R. Moessner, S. L. Sondhi, Phys. Rev. Lett. 110, 107202 (2013)+ unpublished work with V. Kaiser



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### Introducing disorder

#### E.g., $Dy_{2-x}Y_xTi_2O_7/Ho_{2-x}Y_xTi_2O_7$



- At defective tetrahedra,  $S_{\boxtimes} \neq 0$  even at T = 0.
- Cost of bulk  $Q = \pm 2$  monopoles ( $\Delta$ ) higher than impurity  $Q = \pm 2$  monopoles ( $\delta = \frac{4\sqrt{2D}}{3\sqrt{3}}$ )
- Impurity monopoles dominate below  $T_{\delta} \sim \frac{\delta \Delta}{\ln x}$

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#### Disorder effects

Arnab Sen, R. Moessner, Phys. Rev. Lett. 114, 247207 (2015).



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#### Particle-hole transformation



Spins on a random lattice

- Interaction between the "ghost spins" have two parts:
- $\left(D + \frac{3T}{\sqrt{2\pi}}\right) \left(\frac{a}{r_{ij}}\right)^3 (\hat{e}_i \cdot \hat{e}_j 3(\hat{e}_i \cdot \hat{r}_{ij})(\hat{e}_j \cdot \hat{r}_{ij}))$
- The T dependent renormalization is because of the background spin liquid.

### Spin glass of ghost spins

- High temperature :  $\langle S_i \rangle = 0$
- Low temperature:  $\langle S_i \rangle \neq 0$  but  $\frac{1}{N} \sum \langle S_i \rangle = 0$ ;  $q_{EA} = \frac{1}{N} \sum \langle S_i \rangle^2 \neq 0$
- We simulate the dilute system of ghost spins and "confirm a glass transition" [cluster algorithm needed].
- Dense dipoles with random orientations and dilute but collinear dipoles both studied earlier in 3D



#### Phase diagram of diluted spin ice



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### Below glass transition



- A small frozen moment q<sub>EA</sub> develops continuously with T below transition
- Sets up local fields. Resulting Zeeman energy ~ D<sub>\sqrt{qEA}x</sub> attempts to pin bulk spins along the local fields
- Competition with Pauling entropy of  $\frac{1}{2} \ln \frac{3}{2}$

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- Glass: Freezing of ghost spins below T<sub>c</sub>(x). Probes like nonlinear susceptibility, history dependent mag.
- Liquid: Pinch points persist below *T<sub>c</sub>*(*x*). Neutron scattering
- Interplay shows up in gradual but complete loss of Pauling entropy as T is lowered below  $T_c(x)$ . Probe specific heat

#### **Cluster algorithm**



FIG. 1. A particular disorder realization for L = 6 and x = 1/32. Colors at different sites in (a) and (b) represent different acceptance ratios of spin flips  $(R_i)$  using a conventional single spin-flip Metropolis algorithm. Sites with  $(0 < R < 10^{-4})$  are denoted by violet,  $(10^{-4} < R < 10^{-3})$  by blue,  $(10^{-3} < R < 10^{-2})$  by green,  $(10^{-2} < R < 0.1)$  by yellow, (0.1 < R < 0.25) by orange and (0.25 < R < 1) by red. With the chosen cluster parameters  $(a_s = 1.3125, b_s = 0.75)$  and  $C_L = N/5$ ), three clusters est  $C_0, C_1$  and  $C_2$  are obtained for this disorder realization. (c) shows the member sites of the clusters that belong to the set  $C_2$  (in violet). (d) shows the additional member sites of the clusters in  $C_0$  that are already not part of  $C_2$ ,  $C_1$ (in orange). The figures were generated using the graphics software QMGA [41].

#### with Tushar Kanti Bose (IACS)

- Problem with single spin flips –rare clusters of spins with  $|J_{ij}| \gg |J_{avg}|$  frozen
- Can simulate much larger number of dipoles reliably

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#### **Equilibration tests**



•  $q_{EA}^{\alpha\beta}(\mathbf{k}) = \frac{1}{N} \sum_{i} \mu_{i}^{\alpha(1)} \mu_{i}^{\beta(2)} \exp(i\mathbf{k} \cdot \mathbf{r}_{i})$  where  $N = 16L^{3}x$ ,  $\alpha, \beta = x, y, z$ 

• 
$$\chi_{SG}(\mathbf{k}) = N \sum_{\alpha,\beta} [\langle |q_{EA}^{\alpha\beta}|^2 \rangle]$$

### Universality (I)



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## Universality (II)



 Thermal exponent ν = 1.27(8) and anomalous exponent η = 0.228(35) extracted from the finite size scaling analysis

#### Dynamical Griffiths effects



#### with Tushar Kanti Bose (IACS)

- Spins in different clusters have different timescales for fluctuations
- Dynamical heterogeneity remain even for  $T \sim 4T_c$

#### **Clusters from dynamics**



- Spins with slow dynamics (*R* < 0.01) clearly show clustering</li>
- Inter-cluster correlations are also significant between certain clusters

#### Conclusions: disorder in spin liquids

- Plays many roles: may be a nuisance in some cases but may be really interesting in others
- Topological glass in spin ice
- Identification of glass-like and liquid-like degrees of freedom
- Novel cluster algorithm to simulate this spin glass
- Dynamical Griffiths effects for local dynamics

# Thanks for your attention

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