Neutron Scattering from Corner-sharing Simplexes

Collin Broholm

For Christopher Henley on his 59th Birthday

Supported by U.S. DoE
Basic Energy Sciences,
Materials Sciences & Engineering
DE-FG02-08ER46544
Corner-sharing simplexes:

- **Kagome**: Highly constrained yet highly degenerate
- **Pyrochlore**: Balents et al.
- **Hyper Kagome**: Takagi et al.
Magnetic Neutron Scattering

\[ Q = k_i - k_f \]

\[ \hbar \omega = E_i - E_f \]

\[
\frac{d^2 \sigma}{d\Omega dE} = \frac{k_f}{k_i} N r_0^2 \left| \frac{g}{2} F(Q) \right|^2 e^{-2W(Q)} \sum_{\alpha\beta} \left( \delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta \right) S^{\alpha\beta}(Q\omega)
\]

\[
S^{\alpha\beta}(Q,\omega) = \frac{1}{2\pi \hbar} \int dt \ e^{-i\omega t} \frac{1}{N} \sum_{\mathbf{R} \mathbf{R}'} e^{iQ(\mathbf{R}-\mathbf{R}')} < S^\alpha_\mathbf{R}(0) S^\beta_{\mathbf{R}'}(t) >
\]
User facilities for neutron scattering

If your proposal is accepted!
A godfather in neutron scattering?

Reviewer: [redacted]
Rating: F (Fair)
I am a theorist comparatively un-expert concerning magnetization plateaus, but I know a wide variety of materials exhibit them. So the unanswered question is, why *this* material?

Reviewer: [redacted]
Rating: G (Good)
As a theorist, I am somewhat prejudiced against "stuffing" as

Reviewer: [redacted]
Rating: E (Excellent)
I commend this proposal for being so clear and informative. (Perhaps, being a theorist, I just liked it because it is somewhat theory-laden!)

Reviewer: [redacted]
Rating: E-VG
Herbertsmithite has been taking its turn as the latest and so far best approximation of the theorist's ideal spin-1/2 Kagome antiferromagnet. This appears to be gapless and to lack spin order; but theory really does not understand gapless spin liquids.

<p>
Till now these experiments relied on powder samples. As the proposal says it would be wonderful to find out just where in the zone the (gapless) action is, so I would give one or both of these experiments a priority.
Overview

• The theme: corner-sharing simplexes

• Experimental variations
  – No forward scattering in SCGO
  – Resilient form factor in ZnCr2O4
  – Quantum monopoles in Pr$_2$Zr$_2$O$_7$
  – Continuum scattering in Herbertsmithite

• Observations
## Main Collaborators

<table>
<thead>
<tr>
<th>Name</th>
<th>Institution</th>
</tr>
</thead>
<tbody>
<tr>
<td>Satoru Nakatsuji</td>
<td>ISSP</td>
</tr>
<tr>
<td>Kenta Kimura</td>
<td>ISSP/Osaka</td>
</tr>
<tr>
<td>Seunghun Lee</td>
<td>U. Virginia</td>
</tr>
<tr>
<td>Gabriel Aeppli</td>
<td>PSI</td>
</tr>
<tr>
<td>Young Lee</td>
<td>MIT/Stanford</td>
</tr>
<tr>
<td>Tianheng Han</td>
<td>Argonne</td>
</tr>
<tr>
<td>Jiajia Wen</td>
<td>JHU</td>
</tr>
<tr>
<td>Tyrel M. McQueen</td>
<td>JHU</td>
</tr>
<tr>
<td>Seyed Koohpayeh</td>
<td>JHU</td>
</tr>
<tr>
<td>Martin Mourigal</td>
<td>JHU</td>
</tr>
<tr>
<td>Arthur P. Ramirez</td>
<td>U. Santa Cruz</td>
</tr>
<tr>
<td>Jose A. Rodriguez</td>
<td>NIST</td>
</tr>
<tr>
<td>G. Granroth</td>
<td>SNS</td>
</tr>
<tr>
<td>Matt Stone</td>
<td>SNS</td>
</tr>
</tbody>
</table>
A Kagome Sandwich in SCGO

- Defects produce non-interacting “half orphan” spins
- Screening pushes their contribution to scattering to very low $Q$

$\Delta S(Q) \rightarrow 0$
Effective Hamiltonians and dilution effects in Kagome and related anti-ferromagnets

Christopher L. Henley


\[ H = J \sum_{\langle ij \rangle} s_i \cdot s_j - B \cdot \sum_i s_i = \sum_\alpha \frac{J}{2} \left| L_\alpha - \frac{\lambda}{J} B \right|^2 + E_0 \] (1.2)

Thus, the defect creates a slowly-varying spin twist with a pseudo-dipolar spatial dependence. (It has the angular dependence of a dipole field and decays with distance as \(1/|r|^d\), where \(d\) is the spatial dimension.)

4.4. Divergence theorem

I now introduce a sort of “Gauss law”, which is handy for revealing the nonlocal effects of defects.

Acknowledgements

I thank E.F. Shender, V.B. Cherepanov, C. Broholm, P. Mendels, and I. Mirebeau for stimulating discussions, and I thank Johns Hopkins University for hospitality (in 1994) when parts of this work were begun. This work was supported by NSF Grant No. DMR-9981744, and used the Cornell Center for Materials Research computing facilities, supported by NSF MRSEC DMR-9632275.
AFM Isotropic spins: Cooperative Paramagnet

\[ \mathcal{H} = |J| \sum_{ij} S_i \cdot S_j \]

\[
= \frac{1}{2} |J| \sum S_i^2 + cst
\]

<table>
<thead>
<tr>
<th>Compound</th>
<th>Spin</th>
<th>( \Theta_{CW} ) (K)</th>
<th>Order</th>
<th>( T_C ) (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MgTi_2O_4</td>
<td>( \frac{1}{2} )</td>
<td>-</td>
<td>Singlet?</td>
<td>260</td>
</tr>
<tr>
<td>MgV_2O_4</td>
<td>1</td>
<td>-750</td>
<td>Orbital/AFM</td>
<td>-/45</td>
</tr>
<tr>
<td>ZnV_2O_4</td>
<td>1</td>
<td>-600</td>
<td>Orbital/AFM</td>
<td>-/40</td>
</tr>
<tr>
<td>CdCr_2O_4</td>
<td>( \frac{3}{2} )</td>
<td>-83</td>
<td>AFM</td>
<td>9</td>
</tr>
<tr>
<td>MgCr_2O_4</td>
<td>( \frac{3}{2} )</td>
<td>-350</td>
<td>AFM</td>
<td>15</td>
</tr>
<tr>
<td>ZnCr_2O_4</td>
<td>( \frac{3}{2} )</td>
<td>-392</td>
<td>AFM</td>
<td>12.5</td>
</tr>
</tbody>
</table>

\( \Theta_{CW} \)
Average form factor for AFM hexagons

\[ I(Q) = \sum_{\hat{n}=(111)} |F_{\hat{n}}(Q)|^2 \]

\[ \propto \left\{ \sin \frac{\pi}{2} h \left( \cos \frac{\pi}{2} k - \cos \frac{\pi}{2} l \right) \right\}^2 + \left\{ \sin \frac{\pi}{2} k \left( \cos \frac{\pi}{2} l - \cos \frac{\pi}{2} h \right) \right\}^2 + \left\{ \sin \frac{\pi}{2} l \left( \cos \frac{\pi}{2} h - \cos \frac{\pi}{2} k \right) \right\}^2 \]

Tchernyshyov et al. PRL (2001)

INSTITUTE FOR QUANTUM MATTER

iqm.jhu.edu
Straining to order

Edge sharing n-n exchange in ZnCr$_2$O$_4$ depends on Cr-Cr distance, $r$.

\[
\frac{\partial |J|}{\partial r} \approx -40 \text{ meV/Å}
\]

The implication is that there are forces between Cr$^{3+}$ atoms

\[
F_{12} = -\nabla \left( J_{12} \langle S_1 \cdot S_2 \rangle \right) = -\hat{r}_{12} \frac{\partial J_{12}}{\partial r} \langle S_1 \cdot S_2 \rangle
\]

\[Tchernyshyov et al. PRL (2001) and PRB (2002)\]
Magneto-elastic first order transition
SEQVOIA Time of Flight Spectrometer

Fermi Chopper

Detector
The structure factor has pseudo-dipolar singularities:

\[ \langle |\vec{r}(\mathbf{K}_{200} + \mathbf{q})|^2 \rangle \propto \frac{q_\perp^2}{q_{\parallel}^2 + q_\perp^2}. \]

At finite $T$:

\[ q^2 \rightarrow q^2 + \xi^{-2}. \]
Pinch points and monopoles in spin ice

Fennell et al. Science (2009)
Ising FM : Spin-Ice

\[ S_i = \sigma_i \mathbf{d}_k \text{ where } \mathbf{d}_k \cdot \mathbf{d}_{k'} = -\frac{1}{3} \]

\[ \mathcal{H} = -|J| \sum_{\langle ij \rangle} S_i \cdot S_j = \frac{1}{3} |J| \sum_{\langle ij \rangle} \sigma_i \sigma_j \]

\[ = \frac{1}{6} |J| \sum \sigma^2 + cst \]

Ground state: \[ \sigma = \sum_{i \in \downarrow} \sigma_i = 0 \]

6 out of 16 states have \( \sigma = 0 \)

\[ S_{\text{Pauling}} = k_B \ln \left( 2^N \left( \frac{6}{16} \right)^{\frac{N}{2}} \right) = \frac{1}{2} R \ln \frac{3}{2} \]
Emergent Magnetricity

A manifold of ice-rule states

From dipoles to monopoles

Ising spin

Ice rules: two in two out

Castelnovo, Bramwell, Moessner et al.
Quantum Spin Ice in Pr$_2$Zr$_2$O$_7$?

\[ H = \sum_{\langle ij \rangle} \left[ J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \right. \\
\left. + J_{\pm\pm} (\gamma_{ij} S_i^+ S_j^+ + \gamma^*_{ij} S_i^- S_j^-) \right. \\
\left. + J_z (\zeta_{ij} S_i^+ S_j^- + \zeta_{ij} S_i^- S_j^+) \right] \]

Lee, Onoda, Balents

Non-kramers
Gauge-Mean Field phase diagram

Lee, Onoda, Balents, arXiv:1204.2262v2
Elastic scattering $|\hbar \omega| < 0.2$ meV

$\mathcal{I}(T = 0.1 \text{ K}) - \mathcal{I}(T = 20 \text{ K})$
Quantum Fluctuations in Ice-like state

(a) $T=0.1K$, (b) $T=2.0K$, (c) $T=1.0K$, (d) $T=0.1K$

Intensity [10$^2$ counts per 10$^6$ monitor ~ 1.4 min]

Most spectral weight lies in quantum fluctuations
The monopole creation energy of 1.7 K is not resolved
The spectrum broadens on warming:

$$\Gamma(T) = \sqrt{\Gamma_0^2 + (Ck_BT)^2}$$

$$S(\omega) = \frac{1}{1 - e^{-\beta\hbar\omega}} \frac{\chi_0 \Gamma \omega}{\Gamma^2 + \omega^2}$$
No pinch points for inelastic scattering

\[ \nabla \cdot \mathbf{M} \neq 0 \]
Quantum Monopoles in $\text{Pr}_2\text{Zr}_2\text{O}_7$

$\nabla \cdot \mathbf{M} \approx 0$

$\nabla \cdot \mathbf{M} \neq 0$

Calculation by S. Onoda for $\mathbf{J}$ Inferred from CF levels and Anderson super exchange
Long-range order in the classical kagome antiferromagnet: Effective Hamiltonian approach

Christopher L. Henley

Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14853-2501, USA

(Received 5 January 2009; revised manuscript received 9 October 2009; published 2 November 2009; corrected 4 November 2009)

Following Huse and Rutenberg [Phys. Rev. B 45, 7536 (1992)], I argue the classical Heisenberg antiferromagnet on the kagome lattice has long-range spin order of the $\sqrt{3} \times \sqrt{3}$ type in the limit of zero temperature. I start from the effective quartic Hamiltonian for the soft (out of plane) spin-fluctuation modes and treat as a perturbation those terms which depend on the discrete coplanar state. Soft-mode expectations become the coefficients of a discrete effective Hamiltonian, which (after a coarse graining) has the sign favoring a locking transition in the interface representation of the discrete model.

Spin tunneling in the kagome antiferromagnet

Jan von Delft and Christopher L. Henley

Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14853

(Received 5 November 1992; revised manuscript received 12 February 1993)

The collective tunneling of a small cluster of spins between two degenerate ground-state configurations of the kagome-lattice quantum Heisenberg antiferromagnet is studied. The cluster consists of the six spins on a hexagon of the lattice. The resulting tunnel-splitting energy $\Delta$ is calculated in detail, including the prefactor to the exponential $\exp(-\delta_0/H)$. This is done by setting up a coherent-spin-state path integral in imaginary time and evaluating it by the method of steepest descent. The hexagon tunneling problem is mapped onto a much simpler tunneling problem, involving only one collective degree of freedom, which can be treated by known methods. It is found that for half-odd-integer spins, the tunneling amplitude and the tunnel-splitting energy are exactly zero, because of destructive interference between symmetry-related $(\pm)$ instanton and $(\mp)$ instanton tunneling paths. This destructive interference is shown to occur also for certain larger loops of spins on the kagome lattice. For small, integer spins, our results suggest that tunneling strongly competes with in-plane order-from-disorder selection effects; it constitutes a disordering mechanism that might drive the system into a partially disordered ground state, related to a spin nematic.
Herbertsmithite: \((\text{Zn}_{0.85}\text{Cu}_{0.15})\text{Cu}_3(\text{OD})_6\text{Cl}_2\)

Review: Christopher Henley
Rating: E-VG

Herbertsmithite has been taking its turn as the latest and so far best approximation of the theorist's ideal spin-1/2 Kagome antiferromagnet. This appears to be gapless and to lack spin order; but theory really does not understand gapless spin liquids.

Till now these experiments relied on powder samples. As the proposal says it would be wonderful to find out just where in the zone the (gapless) action is, so I would give one or both of these experiments a priority.
Correlations in quantum kagome magnet

- “Rigid” extended structures in momentum space
- Consistent with structure factor for spinon


Hao & Tchernyshyov, PRB (2010)
Continuum in 2D quantum magnet

Spinons on kagome

$S = \frac{1}{2}$ kagome AFM has a finite concentration of spinons in its ground state.

Spinons are solitons with spin $S = \frac{1}{2}$ and fermionic statistics.

Exchange-mediated attraction binds spinons into pairs with spin $S=0$.

The ground state appears to be a Z2 spin liquid.

Y. Wan and O. Tchernysnyov, ArXiv 1301.5008v1 (2013)
Topological excitations and the dynamic structure factor of spin liquids on the kagome lattice

Matthias Punk, Debanjan Chowdhury, and Subir Sachdev
Department of Physics, Harvard University, Cambridge MA 02138

Conclusions

• **Corner-sharing simplexes**
  – Individual spin highly constrained by simplex
  – Relevant degree of freedom is dynamic spin cluster
  – Emergent quasi-particles from violating constraints

• **Low T phases**
  – Order by disorder
  – Magneto-elastic order
  – Order from further neighbor interactions
  – Spin Ice with Pauling entropy
  – Quantum spin liquid

• **Outstanding challenges**
  – Detect “photons” within the quantum spin liquid
  – Detailed understanding of continuum scattering
  – Is there an application for frustrated magnetism?

• **Progress through**
  – Discovery of new materials
  – Detailed scattering studies
  – Imaginative clearly communicated theory: Thank you Chris!