Proposal to use neutron scattering to access scalar spin chirality fluctuations in kagome lattices

Patrick A. Lee and Naoto Nagaosa

Phys. Rev. B **87**, 064423 — Published 28 February 2013

DOI: 10.1103/PhysRevB.87.064423

A Proposal to Use Neutron Scattering to Access Scalar Spin Chirality Fluctuations in Kagome Lattices

Patrick A. Lee
Department of Physics, MIT, Cambridge, MA 02139, USA

Naoto Nagaosa
Department of Applied Physics, The University of Tokyo, 7-3-1, Hongo, Bunkyo-ku, Tokyo 113-8656, Japan and Cross-Correlated Materials Research Group (CMRG), and Correlated Electron Research Group (CERG), RIKEN-ASI, Wako, Saitama 351-0198, Japan

In the theory of quantum spin liquid, gauge fluctuation is an emergent excitation at low energy. The gauge magnetic field is proportional to the scalar spin chirality $\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 \times \boldsymbol{S}_3$. It is therefore highly desirable to measure the fluctuation spectrum of the scalar spin chirality. We show that in the Kagome lattice with a Dzyaloshinskii-Moriya term, the fluctuation in S_z which is readily measured by neutron scattering contains a piece which is proportional to the chirality fluctuation.

PACS numbers:

It has long been suspected that the spin 1/2 antiferromagnetic Heisenberg model on the Kagome lattice may support a spin liquid ground state, i.e., a singlet ground state which has no Neel order due to quantum fluctuations.[1,2] Several years ago the compound $ZnCu_3(OH)_6Cl_2$ (called Herbertsmithite) where the Cu^{2+} ions form $S = 1/2$ local moments on a Kagome lattice was synthesized.[3,4] Despite an exchange constant J estimated to be ~ 200 K, no magnetic order was detected down to 30 mK. Recent neutron scattering shows that the spin excitations are gapless and form a broad continuum.[5] Thus Herbertsmithite has emerged as a strong candidate for the spin liquid state. However, for reasons described below, much remains unknown about this material and the connection with theory is tenuous at best. There is thus a strong need for more experimental probes to help establish the nature of this state of matter.

Theoretically it has been proposed by Ran *et al.*[6] based on projected fermionic wavefunctions that the ground state is a U(1) spin liquid, with spinons which exhibit a gapless Dirac spectrum. On the other hand, recent DMRG calculations on finite size cylinders show strong evidence that the ground state is a Z_2 spin liquid, with a substantial triplet gap.[7] However, the nearest-neighbor Heisenberg model appears to be a very delicate point, because a small ferromagnetic next-nearest neighbor exchange $J_2 \approx -0.01J$ is sufficient to destabilize the Z_2 state.[8] Meanwhile, more detailed projected wavefunction calculations show that the Dirac state is surprisingly stable. Furthermore, the application of a couple of Lanczos steps produces an energy quite competitive with the energy of the Z_2 state obtained by DMRG.[9] Thus while there is general agreement that this ground state is a spin liquid, the precise nature of the spin liquid remains somewhat unsettled.

Experimentally it is known that about 15% of the Zn ($S = 0$) ions which are located between the Kagome planes are replaced by $S = 1/2$ Cu ions. It has been argued that there is not much Zn substitution for Cu in the Kagome planes,[10] so that the disturbance of the Kagome structure may be minimal. However, much of the low energy excitations measured by thermodynamic probes such as specific heat and spin susceptibility are dominated by the local moments between planes. Furthermore, due to spin orbit coupling, we expect deviation from the Heisenberg model. To first order in the spin orbit coupling constant λ, we expect Dzyaloshinskii-Moriya (DM) terms of the form

$$\mathcal{H}_{\rm DM} = \sum_{\langle ij \rangle} \boldsymbol{D}_{ij} \cdot \boldsymbol{S}_i \times \boldsymbol{S}_j \tag{1}$$

where the DM vector $\boldsymbol{D}_{ij}$ is located on bond $\langle ij \rangle$. Since $\boldsymbol{D}_{ij} = -\boldsymbol{D}_{ji}$, the $\boldsymbol{D}_{ij}$ vectors depend on the convention of the bond orientation.[11–14] For a given convention the DM vectors are shown in Fig. 1. The out-of-plane DM term (D^z) has been estimated to be about 8% of J. Due to the delicate nature of the ground state of the Heisenberg model explained above, it is not at all clear that the nearest-neighbor Heisenberg result applies to the Herbertsmithite.

The defining character of a quantum spin liquid is the emergence of exotic particles such as spinons which carry $S = 1/2$ and the associated gauge fields.[15] In the U(1) spin liquid, the gauge field is gapless whereas in the Z_2 spin liquid the gauge field is gapped. The gauge field is defined by the phase a_{ij} of the spinon hopping matrix element $te^{ia_{ij}}$ on link ij. It is a compact gauge field and the spin liquid corresponds to the deconfined phase of the gauge field, so that the compactness may be ignored in the long wavelength limit and a_{ij} may be replaced by a continuum field $\boldsymbol{a}(\boldsymbol{r})$. The gauge invariant quantities are the gauge field $\boldsymbol{b} = \boldsymbol{\nabla} \times \boldsymbol{a}$ which is in the $\hat{z}$ direction and the gauge electric field $\boldsymbol{e} = -\boldsymbol{\nabla} a_0 + \frac{d\boldsymbol{a}}{dt}$ which lies in the plane. The physical meaning of the magnetic flux has been extensively discussed.[16] If Φ is the flux through a plaquette, $\sin \Phi$ is one half of the solid angle subtended by the spins along the

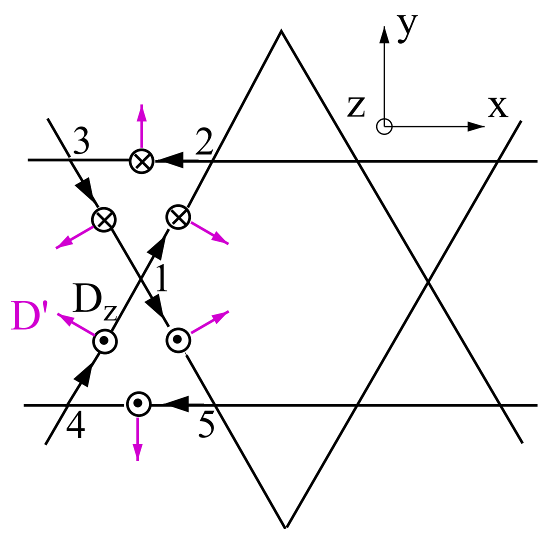

FIG. 1: The DM vectors on the Kagome lattice for Herbertsmithite. The arrow specifies the order of the operator $\boldsymbol{S}_i \times \boldsymbol{S}_j$. (adapted from Ref. 11) D_z is the z component while D' is the in-plane component.

plaquette. For a three site triangle, we have

$$\sin\Phi = \frac{1}{2}\mathbf{S_1}\cdot(\mathbf{S_2}\times\mathbf{S_3}) \tag{2}$$

The quantity $\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 \times \boldsymbol{S}_3$ is known as the scalar spin chirality. Thus the fluctuation spectrum of the gauge magnetic field is proportional to the fluctuation of the spin chirality. It is highly desirable to measure this correlation function because it gives information on the gauge fluctuation and can help to distinguish different spin liquids. However the measurement of a correlation of a product of 3 spin operators is a daunting task. A method to measure the vector chirality $\boldsymbol{S}_1 \times \boldsymbol{S}_2$ has been suggested by Maleev,[17] but that does not apply to scalar chirality. Shastry and Shraiman[18] have suggested measuring chirality fluctuations using Raman scattering, but that contains information only for very small q. A proposal to measure this using resonant X-ray scattering has been made.[19] However, the energy resolution of this technique is currently limited to 20 meV or so, which is on the scale of J.

It turns out that in the Kagome lattice we can turn the DM term to our advantage and achieve a simpler measurement of the chirality correlation. The hint comes from a recent paper by Savary and Balents,[20] who pointed out that in a certain pyrochore spin-ice material, the gauge field (in their case the electric field[21]) is proportional to S_z and its fluctuation can be directly measured by neutron scattering.[22] This system is treated in the strong spin-orbit coupling where J rather than spin S is a good quantum number, but this work raises the possibility of finding the same proportionality in the presence of weak spin-orbit coupling. We find that it is indeed the case for the Kagome lattice with the DM vectors shown in Fig. 1. In the Kagome lattice each site is connected to two triangles. Let us consider the site labeled 1. The total chirality through the two attached triangles is

$$\chi_1 = \boldsymbol{S}_1 \cdot \boldsymbol{S}_2 \times \boldsymbol{S}_3 + \boldsymbol{S}_1 \cdot \boldsymbol{S}_4 \times \boldsymbol{S}_5 g \tag{3}$$

We shall use $\chi_1(\boldsymbol{r})$ to denote this operator at the lattice position $\boldsymbol{r}$. Due to the DM term,

$$\begin{aligned} \langle \boldsymbol{S}_2 \times \boldsymbol{S}_3 \rangle &= \alpha \boldsymbol{D}_{23} \\ \langle \boldsymbol{S}_4 \times \boldsymbol{S}_5 \rangle &= \alpha \boldsymbol{D}_{45} = -\alpha \boldsymbol{D}_{54} = \alpha \boldsymbol{D}_{23} \end{aligned} \tag{4}$$

where α is a constant and the last step is by inspection from Fig. 1. For $|\boldsymbol{D}_{23}| \ll J$, we expect $\alpha \sim \frac{1}{J}$. The important point is the contributions from the two triangles add and we find a linear coupling between χ_1 and $\boldsymbol{S}_1 \cdot \boldsymbol{D}_{23}$. If we average over corners of the triangle 123 and define $\bar{\chi} = \frac{1}{3}(\chi_1 + \chi_2 + \chi_3)$, it is clear that the in-plane component of the $\boldsymbol{D}$ vectors cancel and a local fluctuation in $S_z(\boldsymbol{r}, t)$ induces a local fluctuation in $\bar{\chi}(\boldsymbol{r}, t)$ with a proportionality constant of $(2\alpha D_{23}^z)$. This suggests that a measurement of the $\langle S_z(\boldsymbol{r}, t) S_z(0, 0)\rangle$ correlation function will contain a piece which is proportional to the chirality correlation $\langle \bar{\chi}(\boldsymbol{r}, t)\bar{\chi}(0)\rangle$. A more formal argument proceeds as follows. Let $|\alpha\rangle$ and $E(\alpha)$ denote the exact eigenstates and energies of the Hamiltonian H_0 without the DM term and we perturb in H_{DM}. We are interested in a subset of the excited states, denoted by $|\alpha_\chi\rangle$ which are connected to the ground state $|0\rangle$ by the operator χ_1, i.e., $\langle \alpha_\chi | \chi_1 | 0\rangle \neq 0$. On the other hand, the operator $S_z(\boldsymbol{r})$ has no matrix element to these states, i.e., $\langle\alpha_\chi|S_z(\boldsymbol{r})|0\rangle = 0$. The gauge magnetic field spectral function is proportional to the chirality spectral function

$$S_\chi(q,\omega) = \sum_{\alpha_\chi}\int d\boldsymbol{r}e^{i\boldsymbol{q}\cdot\boldsymbol{r}}\langle 0|\chi_1(\boldsymbol{r})|\alpha_x\rangle\langle\alpha_x|\chi_1(0)|0\rangle\delta(\omega - E(\alpha_x) - E(0)) \tag{5}$$

Now we turn on the Dirac terms. The neutron scattering cross-section is proportional to

$$S(q,\omega) = \sum_f \int d\boldsymbol{r}e^{i\boldsymbol{q}\cdot\boldsymbol{r}}\langle i|S_z(\boldsymbol{r})|f\rangle\langle f|S_z(0)|i\rangle\delta(\omega - (E_f - E_i)) \tag{6}$$

where $|f\rangle$, $E(f)$ are the exact eigenstate and energy of the total Hamiltonian. We now compute the correction to the matrix element $\langle f|S_z|i\rangle$ to first order in $H_{\rm DM}$ for the state $|f_\chi\rangle$ which derives from the state $|\alpha_\chi\rangle$, i.e.,

$$|f_\chi\rangle = |\alpha_\chi\rangle + \sum_{\alpha\neq\alpha_\chi}\frac{\langle\alpha|H_{\rm DM}|\alpha_\chi\rangle}{E(\alpha_\chi) - E(\alpha)}|\alpha\rangle \tag{7}$$

and similarly for $|i\rangle$. Since the zeroth order term vanishes, we find

$$\langle f_\chi|S_z(\boldsymbol{r})|i\rangle = \sum_\alpha\left[\frac{\langle\alpha_\chi|H_{\rm DM}|\alpha\rangle\langle\alpha|S_z(\boldsymbol{r})|0\rangle}{E(\alpha_\chi) - E(\alpha)} + \frac{\langle\alpha_\chi|S_z(\boldsymbol{r})|\alpha\rangle\langle\alpha|H_{\rm DM}|0\rangle}{E(0) - E(\alpha)}\right] \tag{8}$$

Since $|0\rangle$ and $|\alpha_\chi\rangle$ are total spin singlets, we argue that by choosing a different spin quantization axis, S_z can rotated to S_x in Eq.(8) and $|\alpha\rangle$ are spin triplet states. If the spin triplets have a large gap Δ_t we may replace $E(0) - E(\alpha)$ in the second term by $-\Delta_t$. If we are interested in low energy modes of the chirality fluctuations such that $\omega = E(\alpha_\chi) - E(0) \ll E(\alpha) - E(0) \approx \Delta_t$, we may likewise replace that energy denominator in the first term by $-\Delta_t$. Then the sum over $|\alpha\rangle$ can be done and

$$\langle f_\chi|S_z(\boldsymbol{r}_1)|i\rangle = -\sum_{jk}\frac{2D_{jk}}{\Delta_t}\langle\alpha_\chi|S_z(\boldsymbol{r}_1)\hat{z}\cdot\boldsymbol{S}(\boldsymbol{r}_j)\times\boldsymbol{S}(\boldsymbol{r}_k)|0\rangle. \tag{9}$$

Putting this into Eq.(6) and focusing on terms in Eq.(9) where j,k are connected to site 1 as corners of a triangle, i.e., $(j,k) = (2,3)$ or $(4,5)$ in Fig.1, we see indeed that $S(q,\omega)$ has a piece which couples to the chirality fluctuation $S_\chi(q,\omega)$ given by Eq.(5) with a form factor given by $\sum_{\langle jk\rangle}|2D_{ij}/\Delta_t|^2$. The remaining terms couple to correlators of the operator $\boldsymbol{S}_1\cdot\boldsymbol{S}_j\times\boldsymbol{S}_k$ where $1,j,k$ do not form triangles. These are expected to contribute to a smooth background rather than coherent spectra and will be ignored. Note that because $|0\rangle$ and $|\alpha_\chi\rangle$ are eigenstates of the system without $H_{\rm DM}$, what is being measured is the chirality correlation of the system without $H_{\rm DM}$. However, since we are working to leading order in $H_{\rm DM}$, the modification of the full system due to the presence of $H_{\rm DM}$ is small and the measured property can be considered a good approximation of that of the full system itself.

We expect the coupling of $S(\boldsymbol{q},\omega)$ and the chirality correlation $S_\chi(\boldsymbol{q},\omega)$ will continue to hold even if the assumption of a large Δ_t fails, as in the Dirac spin liquid case, but the form factor may acquire some quantitative differences and perhaps some $\boldsymbol{q}$ and ω dependence. This will be the case if the contribution to the sum in Eq(8) from low energy (near gapless) triplet excitations are reduced due to restricted phase space and we can approximately replace the energy denominator by an average triplet excitation energy.

As an example, if the ground state is described by the U(1) Dirac spin liquid, the correlation of the gauge magnetic field is expected to be (in the RPA approximation)[23]

$$\langle|b(q,\omega)|^2\rangle \sim \frac{q^2\theta(\omega - vq)}{(\omega^2 - v^2q^2)^{1/2}} \tag{10}$$

where θ is the step function and v is spinon velocity. The neutron scattering intensity which couples to the S_z channel is expected to have a piece given by Eq.(10), with the intensity reduced by $(2\alpha D^z_{ij})^2$. If the ground state is a Z_2 gapped spin liquid, the gauge fluctuation is gapped and one may expect to see a gapped mode instead of Eq.(10). As pointed out by Savary and Balents,[20] the q^2 dependence in Eq.(10) implies that the gauge fluctuation structure factor vanishes as $q \to 0$, in contrast with the spin wave structure factor which diverges as $1/q$ in this limit. While this helps identify the gauge fluctuation, it also makes its detection more difficult.

The linear relationship between chirality fluctuation and S_z fluctuation is rather special to the Kagome lattice. Such a coupling does not exist for the square lattice, for instance, for slowly varying chirality fluctuations. Consider

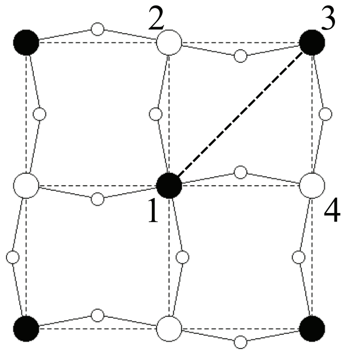

FIG. 2: The distortion of the Ir (solid) and oxygen (open) ions in the Ir-0 plane of Sr_2IrO_4. (adapted from Ref. 23)

magnetic ions on a square lattice each surrounded by corner sharing oxygen cages which rotate about the $\hat{z}$ axis in a staggered manner. (Sr_2IrO_4 is such an example[24] shown in Fig.2.) In this case each magnetic ion is connected to 8 triangles whose opposite side is a nearest-neighbor bond with a DM vector in the $\hat{z}$ direction. The gauge magnetic flux through each square is given in terms of the scalar chirality of the triangles which split up the square.[16] It is easy to see that the contributions from the two triangles 123 and 134 which split up a square cancel each other and there is no linear coupling between the gauge flux and S_z in this case.

In conclusion, we have shown that in the Kagome lattice, the DM term leads to a linear coupling between the S_z fluctuation with the spin chirality fluctuation. It will be interesting to see if neutron scattering can give information on the chirality fluctuation and shed light on the nature of the spin liquid in Herbertsmithite. While the size of the signal $(2\alpha D_{ij}^z)^2$ is small (a few percent) compared with the main signal, in the Kagome, we are helped by the unusual structure factor which greatly suppresses the latter in certain Brillouin zones. This was predicted theoretically[25] based on a model of nearest neighbor singlets and is consistent with the experimental observation[5]. If the chirality spectrum is coherent, they may show up in a careful search in these Brillouin zone where the background is small. Two tests can help support this interpretation: the signal should appear only in the $S_z - S_z$ channel, not $S_x - S_x$ or $S_y - S_y$ and it should not split in a magnetic field because it is a singlet excitation.

We thank Leon Balents, Young Lee and Andrew Potter for helpful discussions. We also thank the KITP for its hospitality and support through NSF grant number PHY11-25915. PAL acknowledges support by the NSF through DMR 1104498. NN is supported by Grant-in-Aid for Scientific Research (Grants No. 24224009) from the Ministry of Education, Culture, Sports, Science and Technology of Japan, Strategic International Cooperative Program (Joint Research Type) from Japan Science and Technology Agency, and Funding Program for World-Leading Innovative RD on Science and Technology (FIRST Program)

[1] P.W. Leung and V. Elser, Phys. Rev. B **47**, 5459 (1993).
[2] P. Lecheminant, B. Bernu, C. Lhuillier, L. Pierre and P. Sindzingre, Phys. Rev. B **56**, 2521 (1997).
[3] M.P. Shores *et al.*, Am. Chem. Soc. **127**, 13462 (2005).
[4] J.S. Helton *et al.*, Phys. Rev. Lett. **98**, 107204 (2007).
[5] Tianheng Han *et al.*, Nature, to be published.
[6] Y. Ran, M. Hermele, P.A. Lee and X.-G. Wen, Phys. Rev. Lett. **98**, 117205 (2007).
[7] S. Yan, D. Huse and S. White, Science **332**, 1173 (2011).
[8] S. White, private communication.
[9] Y. Igbol, F. Becca, S. Sorellla and D. Poilblanc, arXiv: 1209.1858.
[10] D.E. Freedman *et al.*, J. Am Chem. Soc. **132**, 16185 (2010).
[11] See O. Cepas *et al.*, Phys. Rev. B **78**, 140405 (2008).
[12] M. Elhajal, B. Canals and C. Lacroix, Phys. Rev. B **66**, 014422 (2002).
[13] A. Zorko *et al.* Phys. Rev Lett. **101**, 026405 (2008).
[14] S. El Shawish *et al.*, Phys. Rev. B**81**, 224421 (2010).
[15] X.-G. Wen, *Quantum Field Theory of Many-body Systems* (Oxford Univ. Press, 2004).
[16] X.-G. Wen, F. Wilczek and A. Zee, Phys. Rev. B **39**, 11413 (1989).
[17] S. V. Maleev, Phys Rev Lett. **75**, 5682 (1995).
[18] S. Shastry and B. Shraiman, Phys. Rev. Lett. **65**, 1068 (1990).
[19] Wing-ho Ko and P.A. Lee, Phys. Rev. B **84**, 125102 (2011).
[20] L. Savary and L. Balents, Phys. Rev. Lett. **108**, 037202 (2012).
[21] Due to duality the naming of electric and magnetic fields is a matter of choice. In the choice made in Ref. 15, the electric field is odd under time reversal and has the symmetry of the more conventional choice for magnetic field.
[22] O. Benton, O. Sikora and N. Shannon, Phys. Rev. B **867**, 075154 (2012).
[23] L. Ioffe and A. Larkin, Phys. Rev. B **39**, 8988 (1989).
[24] See F. Wang and T. Senthil, Phys. Rev. Lett. **106**, 136402 (2011).
[25] Z. Hao and O. Tchernyshyov, Phys. Rev. B **81**, 214445 (2010).