Hunting of the Spin-Liquids

1. **Unconventional Magnetic Oscillations in Kagome Mott Insulators**
   Authors: Guoxin Zheng, Yuan Zhu, Kuan-Wen Chen, Byungmin Kang, Dechen Zhang, Kaila Jenkins, Aaron Chan, Zhenyuan Zeng, Aini Xu, Oscar A. Valenzuela, Joanna Blawat, John Singleton, Patrick A. Lee, Shiliang Li, Lu Li
   arXiv:2310.07989

2. **Dirac quantum spin liquid emerging in a kagome lattice**
   Authors: Zhenyuan Zeng, Chengkang Zhou, Honglin Zhou, Lankun Han, Runze Chi, Kuo Li, Maiko Kofu, Kenji Nakajima, Yuan Wei, Wenliang Zhang, D. G. Mazzone, Zi Yang Meng, Shiliang Li
   arXiv:2310.11646

Recommended with a Commentary by Chandra Varma,
University of California, Berkeley - Visiting Scholar

The idea that quantum fluctuations in $d = 2$ with $S = 1/2$ spins, especially with sufficient frustration due to competing interactions, leads to states which do not have conventional order even at $T = 0$ is unquestionably true. Such states are being called spin-liquids. Despite nearly thirty years of their intensive investigation, both theoretically and experimentally, it has not been clear what we should expect of their thermodynamics and correlation functions.

Recent advances in the theory of spin-liquids was the subject of a commentary by Joel Moore. The occasion for the present comments are the two experimental papers highlighted above on a recently fabricated compound in which the spins sit on a Kagome lattice - the most frustrated two-dimensional lattice we know. The first includes magneto-oscillations in torque experiments while the second presents results from neutron scattering to obtain the excitation spectra at zero applied field. The results in the two paper are truly intriguing, and there is a valiant effort to understand them in the first paper above.

The low temperature properties of most of the compounds that do not order even far below their antiferromagnetic Curie-Weiss temperature are dominated by their defects; so that they are not good candidates to study spin-liquids even though several among them show curious thermodynamic and transport properties. The compound studied in the papers above appears not to be plagued by defects down at least to 0.3 K, while the measured Curie-Weiss temperature is about 80 K. The compound is a variant of the much studied Herbertsmithite ($ZnCu_3(OH)_6Cl_2$) in which as much as as 5% Zn disorder on the Cu site on a Kagome lattice vitiates the properties. The variant is $YCu_3(OH)_6Br_2[Br_x(OH)_{1-x}]$. This may also not be ideal because the random location of Br and OH makes for disorder in the exchange energies between the Cu's on the Kagome lattice.

Earlier specific heat measurements show a specific heat fitted to be $\propto T^2$ between 0.3 K

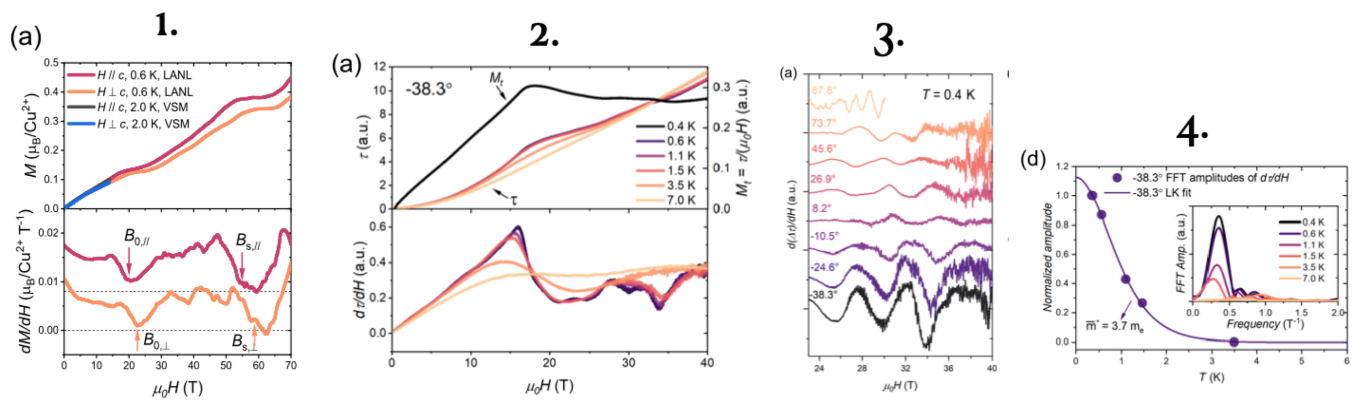

Figure 1: 1. The magnetization and its derivative are shown as a function of the field applied. 2. gives the torque as a function of field and its derivative as a function of field. 3. shows the derivative of the torque at various angles of the field from which it is deduced that the oscillations depends importantly on the field perpendicular to the planes. 4. shows that the amplitude of the oscillations in this very good insulator follows the Kosevich-Lifshitz formula really well. The figures are taken from Zheng et al., the first paper noted above.

(below which a nuclear Schottky anomaly dominates) and 0.7 K, not much of a range to really draw conclusions. The most prominent features of the data in the two papers above are reproduced in Figs. (1) and (2). Magnetization as a function of field shows a plateau near $B = B_0 \approx 20$ Tesla, with a value $\approx 1/9\mu_B$ and another at $B \approx 65$ Tesla with a value close to $1/3\mu_B$. Such plateaus have also been observed by Suetsugu *et al.* and Joen *et al.* The data shows a rather noisy magnetization above 20 Tesla without any discernible oscillations. The measured anisotropy in magnetization gives an anisotropy in g factor of about 1.2; the inevitable spin-orbit interactions and Dzialoshinskii-Moriya interaction in a Kagome lattice lead to magnetization perpendicular to field applied which is measured through torque. Above B_0 there is a well defined region of oscillations in the magnetic torque which is shown up to about 40 Tesla. The amplitude of the torque as a function of temperature follows that for a theory of ordinary fermions, i.e. given purely by the Fermi-Dirac distribution. Their period (six of them are shown) is related to the component of the field $B\cos\theta$ perpendicular to the c-axis as for orbitally induced oscillations generated through Landau levels in two dimensions. But that is not the complete B dependence. The oscillations have been fitted above B_0 to

$$\propto \frac{(B - B_0)^2}{B\cos\theta}. \tag{1}$$

It is surprising that magnetization does not show oscillations while torque does. This may be due to the relative accuracy with which torque is measurable compared to magnetization.

But if more refined measurements rule out oscillations in magnetization, ideas different from those proposed are required.

Numerical calculations for the Heisenberg model in a field give a 1/3 plateau and a 1/9 plateau at about the right value of B/J. The 1/3 plateau appears to be due to a geometric order of the z-component of 1/3 of the spins. The physical picture of the 1/9 plateau is less clear in such calculations.

Only a brief summary of the theory employed in the first paper above to understand the results can be given here. It is based on the very popular theory in which fermions at 1/2 filling in a Hubbard model at large U/t are turned into Dirac fermions in a gauge field b to handle the no double occupancy constraint treated in a mean-field approximation. The fermion band-width is J of $\mathrm{O}(t^2/U) \approx 80$ K, There is an additional gauge field $\propto \alpha B$ with α of $\mathrm{O}(t/U)$, where B is the applied field. The zero-field flux due to the gauge field is assumed to form a periodic pattern on the Kagome lattice giving rise to a $\sqrt{3} \times \sqrt{3}$ structure at $B = 0$. This gives 9 Dirac bands of which $4 + 1/2$ are filled at $B = 0$. B leads also to the Zeeman splitting of the fermions. The explanation of the data rests on some further adjustments. It is envisaged that at $B = B_0$, the chemical potential is such that the down spin Fermi-surface lies at the Dirac point of zero density of states while the up-spin Fermi-surface lies in semi-metallic bands of much higher density of states. For further increase in field, the chemical potential is assumed such that the up-spin Fermi-surface is nearly stuck at the large density of states while the down spin Fermi-surface moves rapidly into the low density Dirac band with an area of the Fermi-surface $\propto (B - B_0)^2$. Landau levels are formed due to the gauge field $b = \alpha B \cos\theta$. This accounts for the oscillations following Eq. (1) but surprisingly the data requires $\alpha \approx 1$. The amplitude of oscillations follows Kosevich-Lifshitz theory which is one of the clearest results in the experiments. From the same data one gets an effective mass which is well fitted to be proportional to $(B - B_0)$ which the same model gives with a coefficient in reasonable agreement with the experiment. This is also consistent with the change with field of the Fermi-surface which is also an explanation of the V-shaped magnetization around 20 Tesla.

As the authors clearly state, the explanation of the the data near and above 20 Tesla which is not much smaller in energy than J has little to say about the zero-field state, where this kind of spin-liquid state, one can argue, is most likely to be found theoretically because the fluctuations are not gapped out in any direction. In fact, their state at 20 Tesla has the band-structure with $\sqrt{3} \times \sqrt{3}$ structure with chemical potential at a Dirac point, which appears to be consistent with the $S = 1$ gapless spin-wave excitations spectra and linear dispersion emanating from the six K-K' points, measured by neutron scattering at zero field. This is illustrated by the schematic reproduced from the second paper above in Fig. (2) at zero field. That is more consistent with an ordered magnetic structure with a broken translational symmetry, not withstanding the title of that paper. (But there is no thermodynamic evidence for a transition at any temperature.) In contrast, the model to explain oscillations and other properties at high fields has a large Fermi-surface at zero field, which does not appear compatible with the spin-wave dispersion.

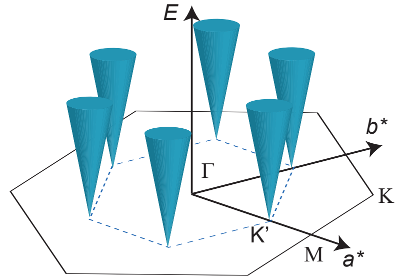

Figure 2: Schematic representation of the dispersion of the spin-fluctuations spectra obtained, taken from the second paper noted above. The detailed data in the paper shows the interesting fact that the bottom of the spectra at any momentum is much better defined compared to the top, reminiscent of spectra expected and observed in one-dimensional spin-1/2 systems. The data goes up to 1 meV in energy.

The sample and the data both on the magnetic torque and the neutron scattering appear sound enough that these papers should inspire more experiments and theory which understands principal aspects of all their properties. Understanding spin-liquids is a truly important problem.

I wish to thank Patrick Lee for a detailed discussion of the theory proposed for these experiments.