Mysterious Order for Spins on a Triangular Lattice

Unconventional spin freezing and fluctuations in the frustrated antiferromagnet NiGa2S4
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**Recommended with a Commentary by Chandra Varma,
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While the curious search for theoretical models for and experimental realization of spin-liquids remains unfulfilled, there are some genuine recent discoveries in two-dimensional spin systems which are nourishing food for thought as well as recall some older mysteries.

$NiGa_2S_4$ crystallizes with very well separated stacks of triangular $Ni^{2+}, \mathbf{S=1}$ ions. The high temperature magnetic susceptibility is isotropic to the best resolution showing negligible effects of crystal field or spin-orbit induced anisotropies. The simplest model for it is a $\mathbf{S=1}$ Heisenberg model on a triangular lattice which one might expect to order with a 120 degrees triad of spins to form a $\sqrt{3}\times\sqrt{3}$ antiferromagnetic structure at $T \to 0$. Instead, the experiments show that the magnetic susceptibility which has a Curie-Weiss form for $T \gtrsim 150K$ with a Weiss constant $\approx -80K$ shows a change in slope at $8.5K$ (and has a finite value for $T \to 0$, just a few percent below its value at $8.5K$). Moreover, the specific heat appears quite smooth across $8.5K$. After judiciously subtracting the lattice specific heat, C_M/T, the "magnetic" specific heat divided by temperature, shows *two* bumps: one peaked at about $10K$ and the other at $T \approx 100K$ where the susceptibility begins to decrease from the Curie-Weiss value. Integration of C_M/T to get the entropy shows a plateau at $(1/3ln3)k_B/spin$ at about $10K$ before saturating to $k_B(ln3)$/spin at high temperature.

Careful neutron scattering shows only short range spin correlations (correlation length $\approx 25\AA$) around an incommensurate wave-vector. There is also evidence that the bulk of the material has no spin-glass freezing. All this is accompanied by the delicious fact that the low temperature specific heat is $\propto T^2$ implying well-defined excitations with $\omega = vk$, expected for instance in a 2d Antiferromagnet. The specific heat is however quite insensitive to a field up to 7 $Tesla$ indicating that these are not spin-waves.

All this was known about three years ago and is discovered or summarized in the second of the two papers above. The T^2 low temperature specific heat is reminiscent of the $\mathbf{S} = 3/2, Co^{2+}$ on a Kagome lattice discovered many years ago[1] and which also shows no specific heat singularity or long-range order discoverable by neutron scattering. The Co-compound however has a singularity in the non-linear susceptibility $\partial^3 M/\partial H^3$, while no such singularity[2] is in evidence in $NiGa_2S_4$. The properties of the Co-Kagome lattice compound continues to be a mystery.

The new development is the discovery of muon oscillations in the first paper highlighted above and in the references in that paper, setting in at $8.5K$, the same temperature as the change in slope of the magnetic susceptibility, as well as an apparent singular change in the muon relaxation rate at the same temperature. For high temperatures the muon relaxation follows what is expected from the fluctuations of a $\mathbf{S} = \mathbf{1}$ paramanget. The muon relaxation rate in the "ordered state" is quite field dependent in striking contrast to the specific heat.

Traditionally the muon oscillations are associated with the setting in of a local magnetic-field $H(0)$ at the muon site. The $H(0)$ deduced has a dramatic change: from a value of essentially zero at $8.5K$ to about 1 kilo-Gauss within about 0.2 K below it. It saturates at a value at low temperatures of about 2.5 kilo-Gauss. Such a field corresponds to a frozen magnetic moment (frozen at least at time scales less than about 10^{-6} secs.) of almost the full Ni^{2+} value at a distance of a few angstroms!

The contrast between the muon results and the neutron scattering and specific heat results below $8.5K$ is so dramatic that one is forced to ask if (the charged) muon disrupts the state locally and uncovers a frozen magnetic moment in its vicinity. But it is hard to think that a frozen moment is possible unless some other subtle order parameter sets in globally at this temperature or some correlation length grows very very rapidly near $8.5K$.

A proposal for a subtle order comes from Monte-Carlo simulations [3] of a classical Heisenberg Hamiltonian supplemented by a biquadratic term which has also been investigated by other methods[4]:

$$H = -\sum_{<ij>} J\left(\mathbf{S}_i \cdot \mathbf{S}_j\right) - K\left(\mathbf{S}_i \cdot \mathbf{S}_j\right)^2 \tag{1}$$

with J antiferromagnetic and K may have either sign. The biquadratic term occurs naturally in the spin-1 representation of the permutation operator. Even for $K = 0$, the model appears to have a topological transition at finite temperatures in which the chirality ($\pm$ or $Z(2)$) defects in the ordered triads of $S = 1$ spins in a $\sqrt{3} \times \sqrt{3}$ ordered structure expected for the ground state proliferate. For finite $|K|$ comparable to $|J|$, there appear to be other topological transitions to phases which are of the quadrupolar or spin-nematic form. There would appear to be no specific heat singularity, just as in the xy model in 2D, and a low temperature phase with T^2 specific heat due to the oscillations of the quadrupoles which is insensitive to magnetic fields.

The Montecarlo calculations ought to stimulate analytic derivations of the topological transitions. At the moment, we do not even understand why the entropy in the low temperature phase exhausts 1/3 of the available entropy and certainly not what is the source of the singularity in the internal magnetic field that the muon spins feel.

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2. S. Naktsuji, Private Communication.
3. H. Kawamura and A. Yamamoto, J. Phys. Soc. Japan **76**, 073704-1 (2007)
4. A. Läuchli. F. Mila and K. Penc, Phys. Rev. Lett. **97**, 087205 (2006).