A possible quantum spin liquid in a spin-orbital entangled 2d magnet

Spin-Orbital Entangled Quantum Liquid on Honeycomb Lattice
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Localized magnetic moments in Mott insulators typically order into long range magnetism. It has long been recognized that strong quantum fluctuations - such as found in low dimensional and/or frustrated magnets - may inhibit such ordering and give rise to a paramagnetic ground state. A special class of such quantum paramagnets - known as quantum spin liquids - have been attracting tremendous theoretical and experimental attention in the last 25 or so years[1].

The concept of a quantum spin liquid was conceived theoretically by Phil Anderson in 1973 and revived by him in 1987 in the context of the cuprate high temperature superconductors. A fascinating aspect of a quantum spin liquid is that it supports excitations with novel fractional statistics and/or fractional quantum numbers. From a modern perspective quantum spin liquid ground states are characterized by long range quantum entanglement between the local degrees of freedom (analogous to the celebrated fractional quantum Hall states). This distinguishes them from other simpler quantum paramagnets (such as ,e.g., a dimerized state that spontaneously breaks lattice translation symmetry).

Theoretically there has been tremendous progress[1] in our understanding of such quantum spin liquid phases in diverse systems in the last 3 decades. An infinite variety of quantum spin liquids are known to be possible - they are distinguished by the nature of their low energy excitations. A crude but experimentally relevant distinction is whether the spectrum of excitations is gapped or gapless.

Experimentally a number of diverse systems have been discussed as candidate quantum spin liquids in the last 15 years. These include a number of quasi-two dimensional organic salts, inorganic spin-1/2 Kagome magnets, and some quantum magnets on frustrated three dimensional lattices.

Quantum spin liquid states have also been sought in a class of quasi-two dimensional Iridium oxides of the form A_2IrO_3 for a number of years. Such materials form a spin-orbit entangled Mott insulator with effective J = 1/2 moments on the Ir sites. Each Ir ion is octahedrally coordinated with O ions and neighboring octahedra share a common edge. Further the Ir ions form layers of honeycomb lattices which are only weakly coupled

magnetically with each other. It was proposed by Jackeli and Khaliullin[2] in 2009 that the dominant interactions between the local moments in such materials are of the type:

$$H_K = -K \left(\sum_{x-bonds} S_{ix}S_{jx} + \sum_{y-bonds} S_{iy}S_{jy} + \sum_{z-bonds} S_{iz}S_{jz} \right) \qquad (1)$$

(The x, y, z-bonds label the three distinct bonds on the honeycomb latice). This Hamiltonian was solved exactly by Kitaev in a beautiful theoretical paper in 2006. The exact solution shows that the ground state is a quantum spin liquid with an emergent Z_2 gauge field and gapless propagating Majorana fermions with linear dispersion. Excitations (known as visons) that carry the flux of the Z_2 gauge field are gapped and dispersionless. The vison gap is however small - a few percent of the exchange K. Half of the magnetic entropy per spin ($\frac{1}{2}\ln 2$) is released only below this vison gap. Numerical calculations[3] show a corresponding big peak in the specific heat at temperatures corresponding to the vison gap. At lower temperatures, the gapless Majorana fermions make a T^2 contribution to the heat capacity due to their linear dispersion. A peculiarity of the exact solution is that despite the gapless spectrum the NMR relaxation rate is exponentially small at low temperature with an activation gap set by the vison gap. The model remains exactly solvable when the interaction strengths in the different directions are different. If the exchanges are sufficiently anisotropic the Majorana fermions are gapped out and the heat capacity is exponentially small at the lowest temperatures.

The Jackeli-Khaliullin proposal has lead to intense experimental scrutiny[4] of the 2-1-3 Iridates and of $\alpha - RuCl_3$ which have come to be known as Kitaev materials. However the best studied iridate Kitaev materials Na_2IrO_3 and Li_2IrO3 seem to order magnetically rather than form a spin liquid. Indeed any material realization will have additional interactions[5] (such as the ordinary Heisenberg exchange) beyond the proposed Kitaev exchange and these seem efficient in favoring magnetic ordering over the spin liquid state.

In the paper selected for this review, Takagi and co-workers synthesize a new member of the honeycomb iridate family: $H_3LiIr_2O_6$. Compared to Li_2IrO3, in this material, all the Li in between the honeycomb layers are replaced by H. However the structure of the basic honeycomb layer is unchanged. $H_3LiIr_2O_6$ is insulating, and has a Curie-Weiss spin susceptibility at high T with a Curie-Weiss constant of about $-100K$. However no magnetic ordering is observed in either the susceptibility, or NMR experiments down to a fraction of 1 K, or in heat capacity measurements down to 0.05 K. The absence of magnetic ordering down to temperature scales much smaller than the Curie-Weiss temperature makes it hopeful that this material realizes a quantum spin liquid.

Several unusual properties are seen in the very low temperature limit $T < 1K$. The heat capacity $C(T) \sim \sqrt{T}$ but application of an external magnetic field $B > 1$ Tesla converts this to a T^2 temperature dependence with a coefficient that decreases as $B^{-\frac{3}{2}}$. Further the NMR relaxation rate $\frac{1}{T_1}$ is roughly proportional to T at low fields but $\frac{1}{T_1 T}$ decreases below a characteristic temperature scale $T^* \sim \frac{\mu_B B}{k_B}$ as the field is increased. In contrast to these observations the Knight shift is constant at low-T and only has a weak field dependance.

Is this the long sought Kitaev spin liquid? How should one understand these various measurements? Several comments are pertinent. The low temperature behavior does not directly fit the properties of the exactly solved model. For instance the low T heat capacity

accounts for only about 1-2 per cent of the $\ln 2$ spin entropy in contrast to the 50 per cent expected below the vison gap in the exactly solved model. The material will surely not be described precisely[5] by the Kitaev model. For instance the mechanism which generates the Kitaev exchange naturally leads to a ferromagnetic sign while the observed Cure-Weiss constant is antiferromagnetic. Thus there must clearly be other important interactions.

Taken together a suggestive picture emerges of the phenomenology from these facts: the majority of spins are rendered inert (i.e are gapped) at low temperatures and do not contribute significantly to the thermodynamics or NMR relaxation. The low T anomalies are instead due to a small fraction of "defect" spins. One important issue then is to understand the physics of the "silent majority" of spins: do they form a gapped quantum spin liquid? Such a state is not expected in the pristine Kitaev model but can arise if there is some distortion leading to different bond strengths in different directions. It remains to be seen if such a distortion is present in the material. Alternately the possibility that the majority spins form a spin liquid sharply distinct from that in the exactly solved model should be kept in mind. A different issue is to understand the mechanism by which a few defect spins (the "rebels") can lead to the observed low-T anomalies.

In any case as the first honeycomb Kitaev material that stays paramagnetic down to low T, $H_3LiIr_2O_6$ is an exciting new system which will stimulate much further experimental and theoretical studies.

References

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