The Search for the Quantum Spin Liquid in Kagome Antiferromagnets

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A quantum spin liquid (QSL) is an exotic quantum ground state that does not break conventional symmetries and where the spins in the system remain dynamic down to zero temperature. Unlike a trivial paramagnetic state, it features long-range quantum entanglement and supports fractionalized excitations.[1] Since Anderson's seminal proposal in 1973,[2] QSLs have been vigorously studied both theoretically and experimentally. Frustrated magnets have been the most fruitful playground for the QSL research.[3] These are materials with competing exchange interactions, which typically arise from triangle-based lattices, leading to macroscopic classical ground state degeneracy. This type of frustration is a key ingredient in discovering quantum disordered ground states.[3]

The spin-1/2 Heisenberg model on the kagome lattice, a two-dimensional lattice formed by corner sharing triangles, is an intensively studied frustrated model. From early on it was recognized that the ground state of the nearest neighbor kagome lattice antiferromagnet is a non-magnetic state, although it is not clear whether it is a QSL[4] or a valence-bond-solid[5] which breaks the translation symmetry. Recent state-of-the-art numerical studies have converged on the ground state being a QSL, yet the nature of the QSL remains an open question with evidences for both a gapped Z_2 QSL[6−8] and a gapless $U(1)$ QSL.[9,10]

Experiments on kagome lattice antiferromagnets are equally challenging. One of the difficulties arises from the rarity of magnetic materials that contain perfect kagome lattices. The situation changed when the successful synthesis of herbertsmithite was reported in 2005.[11] Herbertsmithite is the full Zn end member of Zn-paratacamite with general chemical formula $Zn_xCu_{4-x}(OH)_6Cl_2$ $(0 \leq x \leq 1)$, where perfect kagome layers of spin-1/2 Cu^{2+} are separated from each other by the non-magnetic Zn layers. Since then, extensive characterization has been carried out on herbertsmithite, and all signs point to a quantum disordered ground state consistent with a QSL.[12,13] In particular, inelastic neutron scattering measurements on herbertsmithite single crystals revealed a continuum of magnetic excitations that is characteristic of the fractionalized spinons.[14] Analogous to the situation in theoretical studies, however, it has been difficult to resolve whether or not the putative QSL is gapped. This is due to the complexity that even in the best herbertsmithite single crystal synthesized so far, the Zn substitution is not perfect: while the kagome layers remain fully occupied by Cu, ∼15% of the Zn sites are occupied by Cu.[15] These "impurity" spins are expected to be weakly interacting and contribute mainly to the low energy magnetic response, and therefore hinder the direct probe of the intrinsic gap size of the kagome layer spins. Only recently has careful analysis of NMR and inelastic neutron scattering measurements that took into account of the impurity spin contributions found evidence of a gapped QSL in herbertsmithite.[16,17] The possibility of a gapped QSL is further supported by recent NMR work on the kagome QSL candidate Zn-barlowite.[18,19]

The discovery of a new kagome QSL candidate material Zn-claringbullite $[Cu_3Zn(OH)_6FCl]$[20] brings an interesting new addition to the field. Like herbertsmithite, Zn-claringbullite contains well-separated perfect kagome layers, which makes it an ideal platform to explore the kagome QSL.[20] Because of the different coordination environment of the Zn ion, which is trigonal prismatic compared to octahedral in herbertsmithite, the kagome layers in Zn-claringbullite are stacked in an AA pattern instead of ABC stacking, which is similar to Zn-barlowite.[18,21] In fact, the physical properties of the claringbullite family appear to be rather similar to the barlowite family.[20]

The absence of a magnetic transition in Zn-claringbullite is a promising indication of a QSL.[20] This is the tip of the iceberg, and continued studies would further illuminate the novel magnetic ground state in Zn-claringbullite, such as resolving the extent of Zn substitution into the kagome layer Cu sites,

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probing the effects of the impurity Cu spins that sit on the Zn sites, and ultimately determining whether the ground state is gapped. It would also be interesting to see if sizable single crystals of Zn-claringbullite can be grown to facilitate more detailed experimental studies such as inelastic neutron scattering.

With the discovery of new and promising kagome QSL candidate materials, we can expect more clues will be uncovered in the near future to help resolve the long-standing kagome antiferromagnet problem. This important experimental work will also provide new insights regarding topological order and quantum entanglement as manifested in quantum spin liquids in real materials.

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