Chin. Phys. Lett. **39**, 050102 (2022) **Viewpoint**

Material Search for Quantum Spin Liquids on the Simplest Frustrated Lattice

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The past few decades have witnessed a great endeavor in the search for quantum spin liquids (QSLs).[1–8] This phase of matter, which features long-ranged quantum entanglement and fractionalized spin excitations, is beyond Landau's symmetry-breaking paradigm and is naturally associated with the celebrated idea of resonating valence bond (RVB).[1] What is more, high-temperature superconductivity was proposed to emerge from doping such an RVB state.[2,9]

While so far there is still lack of a smoking-gun experimental evidence for identifying a QSL, this exotic phase has been well established theoretically via exactly solvable models together with other analytical and numerical analyses.[6] It is widely accepted that quantum spin fluctuations play a crucial role in the formation of a QSL, and there are several routes to enhance the fluctuations, namely, (i) a small spin quanta S, (ii) geometric frustration,[10–13] (iii) competing spin interactions that include nearest-neighbor (NN) anisotropic spin coupling and isotropic (or anisotropic) spin couplings on longer bonds, (iv) charge fluctuations in the vicinity of a Mott transition, and (v) extra degeneracy due to orbital degrees of freedom.

The simplest example satisfying (i) and (ii) is the $S = 1/2$ antiferromagnet on the triangular lattice, which was proposed as the first QSL candidate by Anderson in 1973.[1] Unfortunately, numerical studies on the antiferromagnetic (AFM) Heisenberg model with NN couplings on a triangular lattice strongly suggested a long-range 120° AFM order,[6] thereby falsifying Anderson's original proposal. However, the door to triangular-lattice QSLs is still open because of the last three routes: (iii) competing spin interactions, (iv) charge fluctuations, and (v) orbital degeneracy.

On the experimental side, the successful synthesis of the rare-earth compound $YbMgGaO_4$ has aroused a lot of interest in triangular lattices antiferromagnets.[14–26] This insulating compound has triangular layers formed by Yb^{3+} ions, exhibiting spatially isotropic $R\bar{3}m$ symmetry in crystallography.[14] While temperature dependent magnetic susceptibility yields a small AFM Curie–Weiss constant $\theta_w \sim -4\,K$,[14] various measurements, including specific heat, thermal transport, electron spin resonance, and muon spin relaxation, suggest the absence of spin ordering or freezing down to $T \sim 100\,mK$.[14–17] In particular, a continuum of spin excitations has been observed by neutron scattering experiments, broadly distributed in momentum-energy space (0.1–2.0 meV) and indicative of short-range spin correlations.[18] Meanwhile, the Yb^{3+} ions in this compound are slightly displaced by the Mg^{2+}–Ga^{3+} mixing in the interleaved double layers. This lattice distortion will introduce considerable bond randomness. Therefore the possibility of a bond glass (or a random spin-singlet state) cannot be ruled out.[21,23,27,28]

In comparison with $YbMgGaO_4$, a family of newly discovered rare-earth chalcogenides, $AReCh_2$ (A = alkali, Re = rare earth, and Ch = O, S, Se), provides a more promising platform for searching QSLs on triangular lattices.[29,30] This family shares the same $R\bar{3}m$ space group with $YbMgGaO_4$, but has a simpler crystal structure, so it has more chances to avoid the crucial random disorder issue. Furthermore, the relatively larger NN AFM exchange in the sub-family $NaYbCh_2$, $\theta_w \sim -10\,K$, is expected to conquer unavoidable lattice disorder, at least partially. In consistence with these considerations, magnetic susceptibility and specific heat measurements on the representative member, $NaYbS_2$, display no magnetic transition

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down to $T \sim 50$ mK, implying a QSL state. Moreover, the rich diversity of this family allows us to carry out comparative studies.

In addition to the long-sought QSL, the exciting advantages stem from the family of rare-earth chalcogenides $AReCh_2$, also facilitate the studies of other phenomena. Indeed, an insulator to metal phase transition has been observed in $NaYbSe_2$ under a critical pressure ~59 GPa.[31] In the insulating phase, the low temperature resistance can be fitted by Mott's variable range hopping model, $R(T) = R_0 \exp[(T_0/T)^{1/(d+1)}]$, with the dimensionality $d = 2$. As the pressure increases, the QSL candidate becomes a complete metallic state at ~75 GPa, and a non-Fermi liquid is indicated by the transport $R(T) = R_0 + AT^n$ with $n \sim 1$ under this pressure. Above 75 GPa, the power n gradually increases with pressure and reaches the Fermi liquid value $n = 2$ at 126 GPa. Interestingly, when the pressure exceeds ~103 GPa, a sudden drop of $R(T)$ takes place at low temperatures. For instance, a maximum drop by ~6% occurs at ~8 K under 126 GPa, the highest pressure realized in the experiment. This small fraction of the $R(T)$ drop was attributed to the emergence of superconductivity. Certainly, it would be a fragile superconductor, if its existence was confirmed.

Finally, we would like to make a few comments on other vital ingredients to QSLs, namely, (iii) competing spin couplings, (iv) charge fluctuations, and (v) orbital degeneracy, as mentioned above. First, we believe that competing spin couplings widely exist in all the rare-earth compounds, which is indispensable for an $S = 1/2$ (or $J = 1/2$) QSL on triangular lattices. Second, charge fluctuations will manifest and give rise to significant multiple-spin exchanges in the vicinity of a Mott transition. Third, orbital degeneracy is known to provide a mechanism to enhance quantum fluctuations,[32] and recent theoretical studies suggest a nematic spin-orbital liquid state on triangular lattice.[33] With all these essential ingredients in mind, one can draw up a "road map" for the future QSL material searching.

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