Magnetic and non-magnetic phases of a quantum spin liquid

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For over two decades, there have been intensive searches for experimentally realized quantum spin liquid phases in two and higher dimensions. Theoretical studies point to two possible routes to such exotic spin liquid behavior – macroscopic degeneracy of the classical ground state, such as occurs for Heisenberg spins on a Kagome lattice, and geometric frustration with substantial magnetic ring exchanges, such as one has for the Hubbard model sufficiently close to the metal-insulator transition. The material -(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ is an example of the latter: a Mott insulator which is nearly metallic, with spin 1/2 BEDT-TTF dimers on an almost isotropic triangular lattice. No magnetic order has been observed in this material down to 20mK in zero magnetic field, and several earlier experiments have been taken as evidence for spin liquid behavior in this system.[1] While there is some uncertainty about whether the proposed spin liquid phase is gapless or gapped, the experiments by Pratt et al. discussed here, point toward a small gap. Gapped spin liquids are an example of topological order and may support exotic excitations, such as "visons", so there is considerable interest in understanding this system more thoroughly.

Earlier NMR studies on -(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, for fields in the range of 2 – 8 T, had shown the existence of a phase with field and temperature dependent inhomogeneous staggered moments, conjectured to be due to impurities and other defects embedded in an underlying spin liquid. The present work, based on muon spin resonance (MuSR), extends these results to much lower fields, revealing a rich variety of detail in the low temperature and low field phase diagram.

The ability to study much lower fields than is possible for NMR, is a consequence of the much larger moment of the muon compared to that of nuclear spins. What is found is consistent with a uniform gapped spin liquid phase in the millikelvin range for zero and very low fields. In this phase there is no relaxation of the MuSR line, as expected for electronic moments that are paired into singlets in the spin liquid. As the field is increased, the gap decreases, vanishing at about H_c = 5 mT, in an apparently second order transition into a state with weak staggered local moments. The field and temperature dependence of the homogeneous and inhomogeneous relaxation rates of the muon spin resonance are measured and these grow up continuously, following power laws in the difference ($H – H_c$) and in T. The transition, from a low field non-magnetic phase in which the spins are hidden into a magnetic phase which occurs at H_c, may be thought of as a Bose-Einstein Condensation (BEC) of bosonic excitations from the spin liquid phase.

The weak antiferromagnetic moment phase (WAF) exists in a broad region of the phase diagram from the millikelvin range up to over 10 K and from 5 mT to over 10T. This region contains low and high field regions (WAF_L and WAF_H) that exhibit different behaviors as a function of temperature, reflecting behavior also seen in the low-field liquid/paramagnetic phase.

These detailed observations provide the opportunity to compare to theories of spin liquid phases and how they connect to other magnetic phases in a field. The observation that the transition seems to be continuous places a constraint on the nature of the two phases. The low field phase boundary Tc(H), follows a roughly linear behavior, starting from zero at H_0 0.5 mT. Field sweeps, starting at the phase boundary, measuring the increase in the static width of the MuSR line, determine an order parameter exponent, , which characterizes the growth of the weak moments. Similarly, measurements of the temperature dependence of the spin fluctuation rate in different regimes of field and temperature, determine an exponent, w, which for a quantum spin liquid is equal to , the exponent associated with the wavevector dependence of critical fluctuations. These exponents are discussed in the context of possible models for conjectured quantum liquids and other possible states of the system.

Pratt et al. extract the critical exponents and from the data of the two WAF phases and compare to a variety of theoretical scenarios describing behavior close to a QCP. They conclude that models with a weakly gapped spin liquid are favored over those for gapless spin liquids or for models based on orbital currents or impurity-induced moments. The models contain a number of parameters and the high-field phase is not directly adjacent to the spin-liquid phase, so the comparisons between theory and experiment are not completely conclusive. Nevertheless, this paper has mapped out a very interesting phase diagram of -(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ with two quantum critical points, one between a possible spin-liquid phase and the low-field WAF state and the other between the low and high field WAF states. The detailed comparisons to various spin liquid theories suggest a gapped spin liquid phase and it will be of great interest to see if future work confirms this picture.

[1] See, for example, R.S. Manna et al., Phys. Rev. Lett. 104, 016403 (2010) and references therein.