Quantum Effects in Spin Ice

1. Thermodynamic properties of Yb2Ti2O7 pyrochlore as a function of temperature and magnetic field: Validation of a quantum spin ice exchange Hamiltonian.

Authors: N. R. Hayre, K. A. Ross, R. Applegate, T. Lin, R. R. P. Singh, B. D. Gaulin, M. J. P. Gingras.
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2. Crystal-field states of Pr3+ in the candidate quantum spin ice Pr2Sn2O7.

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Recommended, with a commentary by Sunil Sinha, University of California San Diego.

The continuing interest in condensed matter physics in the properties of quantum systems which exhibit exotic properties such as emergent hidden or topological order, quantum entanglement or fractionalized excitations has made the study of quantum spin liquids and the so-called "spin ice" materials an area of considerable activity over the last decade.
This has also coincided with increasing interest in the dynamics of disordered systems which exhibit no obvious long-range order.

Spin liquids are magnetic systems in which the existence of frustrated magnetic interactions between spins, which can lead to highly degenerate ground states and, sometimes together with the existence of large quantum fluctuations, can prevent the formation of long-range magnetic order. Examples are materials with the kagome lattice structure such as herbertsmithite, and the three dimensional magnetic oxides with the pyrochlore structure and the chemical formulae $RE_2M_2O_7$, where RE stands for a trivalent magnetic rare earth ion and M for a tetravalent transition metal ion. The latter constitute the spin ice compounds, of which the canonical ones which have been intensively studied are $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$. The lattice consists of corner-sharing tetrahedra, with the corners occupied by the RE ions. Their magnetic moments interact via short-range antiferromagnetic interactions and long-range dipolar interactions. The crystal field anisotropy requires the moments to point parallel or antiparallel to the <111> directions,

i.e. toward or away from the centers of the tetrahedra. The effective Hamiltonian transformed along the appropriate axes constitutes an Ising-like Hamiltonian. These moments also obey the so-called ice rules, formulated a long time ago for ordinary ice by Pauling, which require two protons to point in to the oxygen and two to point out, hence the name. These systems exhibit a variety of interesting properties, such as large zero temperature entropy, fractionalized excitations which are magnetic (Dirac) monopoles, Dirac strings, interesting singularities in their neutron scattering signatures in reciprocal space etc., all of which have been studied theoretically and experimentally [1].

Interesting as these systems are, they constitute so-called classical spin liquids. Thus there continues to be an active search for the occurrence in these types of compounds for signatures of quantum spin liquids, for which very interesting predictions of emergent U(1) gauge symmetry, accompanied by topological order and the presence of gapless "photon-like" excitations have been made [2,3]. The present articles suggest that suitable candidates for such materials may have been found in the pyrochlore family of compounds.

The article by Hayre et al. points out that interesting quantum dynamics can be realized in such systems with ions where the crystal field ground state is a Kramers doublet in which the interactions between the ions has non-vanishing matrix elements between the component wavefunctions, and show that this is true for the compound $Yb_2Ti_2O_7$, thus contradicting the common expectation of classical spin liquid behavior in the case of systems containing ions with large J values. In order to establish this, they use results from an earlier paper by Ross et al. which measured the spin wave spectra in the spin-oriented paramagnet $Yb_2Ti_2O_7$ induced by an applied magnetic field. The model of Ross et al. yields a good fit to the spin wave excitations with nearest neighbor anisotropic exchange interactions and anisotropic g-factors. Hayre et al. use these parameters to calculate a variety of experimental thermodynamic properties. which compare very well with their measured experimental data over a wide range of fields and temperatures, and conclude that this validates their model of representing $Yb_2Ti_2O_7$ as an effective spin-1/2 system with anisotropic exchange and thus capable of quantum fluctuations.

The paper by Princep et al. discusses a study of the crystal field states of the Pr^{+3} ions in the compound $Pr_2Sn_2O_7$ using time-of-flight neutron

spectroscopy, motivated by the large spin fluctuations and zero-point entropy observed in this compound down to the lowest temperatures. They point out that such zero-point fluctuations can exist if the non-Kramers doublet ground state contains a significant admixture of $| M_J \neq \pm J>$ states mixed in by the crystal field. These authors fit their crystal field parameters to the experimentally observed energies and intensities of the peaks observed in their inelastic spectra, and obtain a good fit to the neutron data and also to the powder-averaged magnetic susceptibility as a function of temperature. Their calculated ground state is indeed a doublet containing admixtures of various M_J values, confirming that this system is a good candidate for quantum spin liquid effects.

These papers point to the next interesting step in this area, which would seem to be a detailed study of the wave-vector dependent low-temperature spin dynamics of these materials to compare with the interesting theoretical predictions referred to above. This will require challenging inelastic and quasielastic neutron scattering experiments or possibly resonant x-ray magnetic scattering inelastic or photon correlation spectroscopy experiments, as various time scales are studied.

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[2] Michael Hermele, Matthew P. A. Fisher, and Leon Balents, Phys. Rev. B 69, 064404 (2004)
[3] SungBin Lee, Shigeki Onoda, and Leon Balents, Phys. Rev. B 86, 104412 (2012)