Quest to U(1) quantum spin liquids, valence bond solids, novel ordered phases in pyrochlores and spinels: unconventional quasiparticles and interference effects

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Magnetic rare-earth pyrochlores have attracted great interest as materials realizing quantum variants of spin ice, dubbed quantum spin ice [1,2]. It potentially hosts a novel U(1) quantum spin liquid where spin-ice monopoles play roles of gapped deconfined fractionalized charge-1 quasiparticles that are coupled to linearly dispersive gapless charge-0 excitations analogous to photons [3]. Because of significantly large and complicated magnetic anisotropy allowed by the symmetry in the nearest-neighbor exchange interactions [2], quantum spin ice systems display a rich phase diagram [4,5]. It includes the U(1) quantum spin liquid around the nearest-neighbor classical spin ice point and long-range ordered phases with Bose-Einstein condensed, Higgs-confined spinons such as a ferromagnetic phase of the magnetic space group $I4_1/am'd'$, as in Yb₂Ti₂O₇ [6], and antiferromagnetic phases of $I4_1'/am'd$, $I4_1/amd$, and $I4_1'/amd'$. However, the U(1) quantum spin liquid has not yet been established in real magnetic rare-earth pyrochlores.

Here, we report a series of our recent theoretical works along this direction. (i) We uncover by means of unbiased extensive quantum Monte-Carlo simulations on a minimal quantum spin ice model that the U(1) quantum spin liquid can be achieved through a classical-to-quantum crossover on cooling from classical spin ice regime [7] and that it might be experimentally verifiable with high-accuracy low-energy inelastic neutron-scattering experiments, possibly on a candidate material such as $Pr_2Zr_2O_7$ but at rather low temperatures. (ii) The phase diagram and magnetic properties of the same model under the 111 magnetic field are also for the first time clarified numerically. A valence bond solid state at the kagome spin ice plateau and a monopole supersolid at the higher fields are found [8]. Furthermore, for more direct relevance to available experiments, we identify quasiparticle excitations in the Higgs-confining ferromagnetic phase of Yb₂Ti₂O₇, in terms of Higgs modes associated with Bose-Einstein condensed monopole-charge-1 spinons and charge-0 gauge excitations endowed with a mass, in favorable comparison with inelastic neutron-scattering spectra [9].

Actually, the Higgs-ferromagnetic phase should involve a macroscopic phase coherence of Bose-Einstein condensed spinons. We then demonstrate an analogous Josephson supercurrent of the bosonic spinons between two Higgs ferromagnets, e.g., Yb₂Ti₂O₇, weakly connected through a nanoscale U(1) quantum spin liquid material, possibly, Pr₂Zr₂O₇ [10]. Since the spinons carry quantum spin ice monopole charges, the effect yields a dissipationless longitudinal spin current, opening a route to potential spintronics applications.

Finally, we make a theoretical proposal for high-temperature quantum spin ice in the thin-film spinel iridate Ir_2O_4 on the basis of first-principles calculations [11], as the onset temperature of the spin ice physics increases up to room temperature, which is two orders of magnitude higher than in pyrochlores.



Fig. 1: Phase diagram of a minimal quantum spin ice model under the 111 field. (Taken from Ref. [8].)

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