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**Quasiparticle condensation and breakdown in a quantum spin liquid**[1]

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Piperazinium hexachlorodicuprate (PHCC) is a frustrated bilayer antiferromagnet with a disordered quantum spin-liquid (QSL) ground state at zero field [1] and a diverse magnetic field versus temperature phase diagram which includes two field-induced quantum critical points [2]. The spin excitations in PHCC have a spectral gap of $\Delta \approx 1$ meV above which they follow a nearly 2D-isotropic dispersion with a bandwidth slightly larger than $\Delta$. Field dependent neutron scattering and thermodynamic measurements reveal a lower critical field, $H_{c1} = 7.5$ T, separating the QSL phase from a three dimensional spin-ordered state and an upper critical field, $H_{c2} = 37$ T, marking the onset of a saturated ferromagnetic phase. The two-dimensional antiferromagnet supports a field induced long range ordered phase well described as a Bose-Einstein condensate (BEC) embedded within a gapless quasi-two-dimensional paramagnetic regime. Inelastic neutron scattering experiments also reveal a peculiar type of hybridization of magnetic excitations in PHCC with their two- particle continuum [3], similar to the post-roton regime in superfluid helium. The excitations at this point become broadened and diffuse, no longer describable as quasiparticles. Although such effects are expected to be strongest in one- dimensional systems with gapped spectra [4], such as Haldane chains, direct observation therein is difficult due to a weak scattering structure factor in the vicinity of the quasiparticle breakdown point [5,6]. The dimer-dominated magnetism in PHCC, on the other hand, is favorable for investigating changes in quasiparticle spectra in the vicinity of their breakdown point. Our results have implications for a variety of condensed matter systems, in particular for other QSLs, where spin excitations have a bandwidth greater than the gap energy.

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[1]Work performed in collaboration with I. Zaliznyak, D. H. Reich, C. L. Broholm, O. Tchernyshyov, P. Vorderwisch, T. Hong, and N. Harrison