Quantum Fluctuations Induce Stable Magnetic Arrangements in Layered Materials

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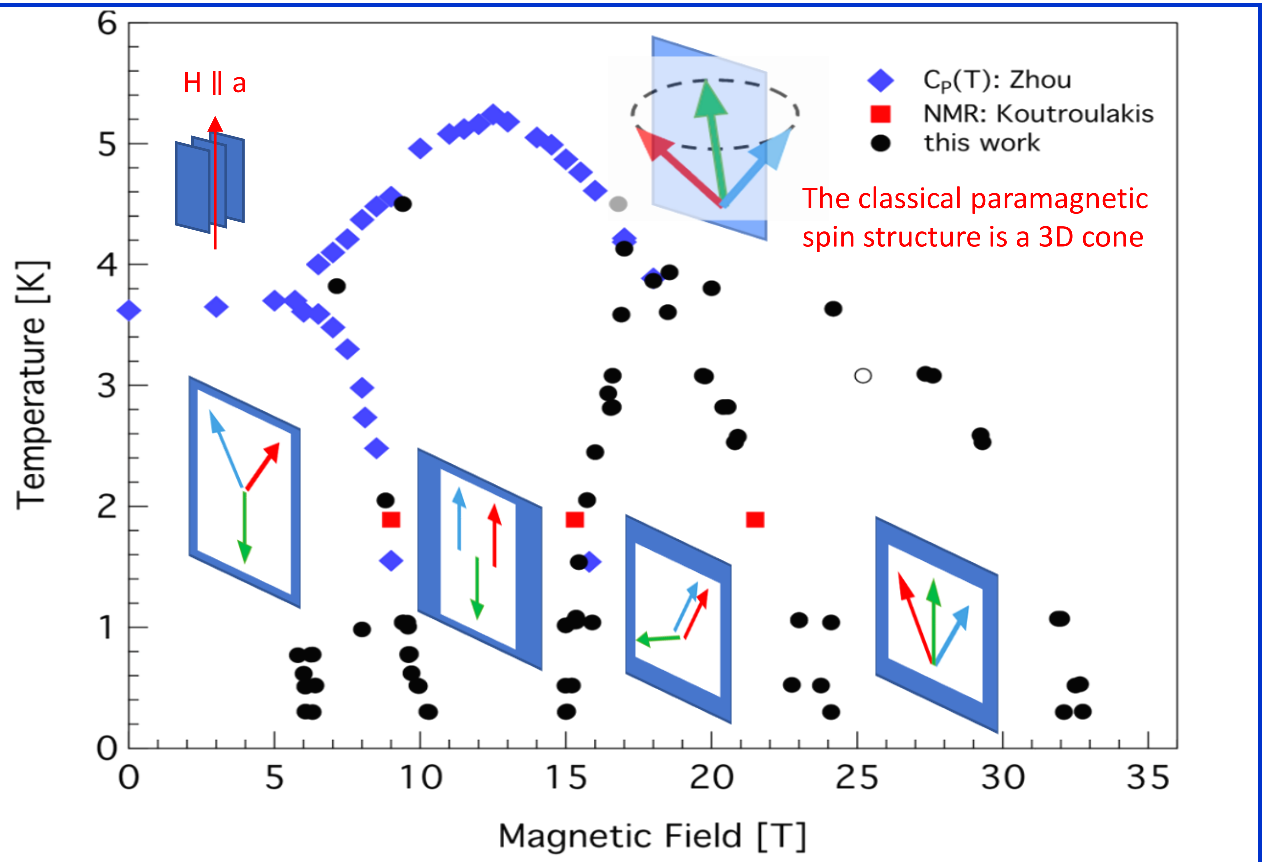
Quantum fluctuations can lift the classical degeneracy of the antiferromagnetic ground state in a magnetic field, producing a series of novel spin structures. This was demonstrated in the effective spin-1/2, layered triangular-lattice quantum Heisenberg antiferromagnet $Ba_3CoSb_2O_9$ when magnetic fields were applied within the crystallographic *ab* plane. One of the four states arising from quantum fluctuations was found to be the celebrated collinear “up-up-down” (UUD) ordering, with a magnetization equal to 1/3 of the saturation magnetization. This collaboration, led by a MagLab user from an undergraduate institution, employed specific heat, neutron diffraction, thermal conductivity, and magnetic torque measurements to map the phase diagram as a function of magnetic field intensity, temperature, and magnetic field orientation.

The results find that a theoretically unexpected magnetic-field-induced phase exists below the UUD phase at temperatures below 1K, as well as the discovery of an unexpected tetracritical point in the phase diagram at high field for H // a. This feature — and the second order nature of the phase transition — eliminates the theoretically predicted spin structure for the phase that exists between 24 T and 33T and indicates instead the so-called “Ψ” phase, so denoted due to the co-planar trident arrangement of the three spins.

The existence of a tetracritical point further reveals that weak interlayer coupling plays an essential role in the magnetic ordering of ‘2D’ triangular lattice quantum antiferromagnets, doubling the period of the magnetic structure along the direction perpendicular to the layers. This allows the spins to change directions in alternating layers, and, in so doing, alters which spin arrangement minimizes magnetic energy in a particular high field phase.

The MagLab’s unique combination of a 35 T resistive magnet, dilution refrigerator, and single-axis rotator made it possible for users to calorimetrically map out the complete phase diagram from zero field to beyond the saturation field of 33 T.



Calorimetrically determined magnetic phase diagram for $Ba_3CoSb_2O_9$ for in-plane magnetic field (H ∥ a). Insets show the classical ground state (a 3D cone), which contrasts with the observed series of four co-planar (2D) spin arrangements that are stabilized by quantum fluctuations. Note the still-unexplained phase below 1K for magnetic fields between 6T and 11T, as well as the tetracritical point at 4K and 18T, where four states meet at a single point.

Facilities and instrumentation used: DC field Facility. Heat capacity measurements shown in black done in 35 T resistive magnet and dilution refrigerator. Blue points done using SCM1.
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