Quantum materials

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Melted spin ice

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Spin ices have magnetic moments arranged on a lattice with many possible ground-state configurations. Quantum effects can 'melt' the spin ice into a liquid that fails to form static order even at absolute zero despite strong interactions.

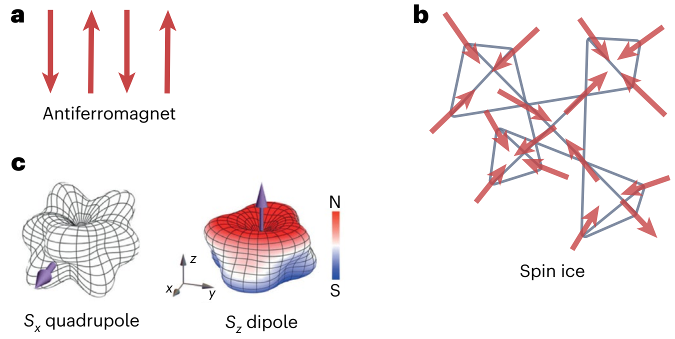

Fig. 1 | Spins in an antiferromagnet and the spin ice $Pr_2Zr_2O_7$. a, Dipole spins forming antiferromagnetism (spins depicted as arrows). **b**, Dipole spins in a classical spin ice 2-in-2-out configuration on a pyrochlore lattice where the lattice forms corner-sharing tetrahedra and the spins point towards or away from the centre of the tetrahedra (*z* axis). There are six ways to satisfy 2-in-2-out for each tetrahedron. The top-left tetrahedron illustrates a low-level 3-in-1-out excitation. **c**, Orbital shape of S_x quadrupolar component and S_z dipolar components of the praseodymium spins, with the colour scale indicating the direction of the magnetic dipole and the arrows indicating the quantization axes. Panel **c** reproduced from ref. [1], Springer Nature Ltd.

In magnetic crystalline materials, the interactions between localized magnetic moments usually cause them to form ordered patterns below a critical temperature. For example, in an antiferromagnet (Fig. 1a), spins on neighbouring atoms point in opposite directions. Typically, there are only a few spin configurations that have the lowest energy. In certain crystal lattices such as spin ices, however, the interactions between spins do not fully constrain the magnetic pattern, resulting in many possible ground states (Fig. 1b). Predictions suggested that, at low temperatures, quantum effects can drive the spins to move freely between different spin configurations and/or form superpositions, producing a so-called quantum spin liquid. Now, writing in *Nature Physics*, Nan Tang and co-workers have found evidence in favour of a quantum liquid phase down to very low temperatures in $Pr_2Zr_2O_7$ (ref. [1]).

Spin liquids are different from molecular liquids like water in that they persist down to absolute zero temperature despite strong interactions between spins. Some of the most interesting predictions for spin liquids are quantum in nature. Spin liquids can be a superposition of different ordering patterns — in some cases creating entanglement between spins that are far apart. The spin liquid states can be dynamic and have topological properties. These phases are predicted to have exotic emergent properties and can form unconventional excitations with properties distinct from the underlying electrons. The number of predicted excitations with different symmetry is vast and may have applications to data storage and quantum technologies[2–5].

Unfortunately, the theory of spin liquids currently outpaces experiments. In real magnets, spin liquid states are often prevented by disorder or weaker interactions that favour ordered ground states or glassy states. As a result, there are only a few candidate spin liquids available in the laboratory, and all are subject to debate as to whether they are true spin liquids. To argue for a spin liquid phase[5,6], experiments must prove a lack of long-range order at low temperatures and find signatures of behaviour that is predicted to be unique to spin liquids. Unconventional excitations can be found with inelastic neutron scattering and other spectroscopic techniques. Indirect evidence can be obtained by comparing unusual thermodynamic quantities to theory and by observing how the spin liquids evolve and are destroyed as a function of temperature, magnetic field, and other tuning parameters.

Spin ices such as the pyrochlore compound $Pr_2Zr_2O_7$ studied by Tang and co-workers (Fig. 1b,c) have been identified as candidate materials to host a spin liquid phase at low temperatures[7,8]. The praseodymium moments in $Pr_2Zr_2O_7$ are not simple magnetic dipoles. Instead the magnetic moments form non-Kramers' doublets due to the coupling between the two electrons that are localized at each praseodymium ion, the magnetism produced by their orbits, and the electric fields of the surrounding ions in the crystal.

The non-Kramers' doublet on each Pr^{3+} ion behaves like a magnetic dipole in one direction (pointing in and out of the tetrahedral along the *z*-axis as illustrated in the right panel of Fig. 1c) and the compound would behave like a straightforward spin ice (Fig. 1b) if that were the end of the story. However, in the transverse directions (here *x* and *y*) the ions form an orbital quadrupole moment that does not couple to magnetic fields but does couple linearly to lattice distortions (illustrated in the left panel of Fig. 1c). Thus, the dipole moments along the *z* axis couple to their neighbours through normal magnetic exchange, while the transverse quadrupole components *x* and *y* couple to the crystal lattice and also to each other via these lattice distortions.

Spin liquid behaviour comes about because the non-Kramers' state is a quantum-mechanical doublet and only the *x*, *y*, or *z* state can be known at any one time. Thus, any interaction that affects one component (like the magnetic dipole) will quantum mechanically scramble the other components (such as the transverse quadrupole moments) and vice versa. These quantum fluctuations make it easier for the system to transit between different states of the spin ice, and also form quantum superpositions between them. These quantum effects can 'melt' the spin ice into what is termed a spin-orbital liquid in recognition of the role of electronic orbital effects. An earlier theoretical investigation[8] identified thirty different spin-orbital liquids that can form from ions with non-Kramers' states on certain crystal lattices.

Tang and co-workers found evidence for a spin-orbital liquid in $Pr_2Zr_2O_7$ by measuring its influence on the length change, sound velocity, and dielectric constant of the crystal. Their observation required crystals that are much purer than those studied previously. This purity is particularly important because non-Kramers' doublets with their linear coupling to lattice distortions are very sensitive to defects in the crystal. Tang and co-workers found no static long-range order in their samples down to extremely low temperatures, as is expected for spin liquids. They also studied how the proposed spin-orbital liquid state is suppressed by increasing temperature and magnetic field. Consistent with a spin-orbital liquid, the observed suppression by magnetic field is accompanied by a liquid-gas-type transition without change in symmetry.

The measurements by Tang and co-workers offer strong preliminary evidence of a spin-orbital liquid. They set the stage for future investigations of $Pr_2Zr_2O_7$ such as spectroscopic measurements to further support the claim of a spin-orbital liquid state. We expect this work will also motivate further investigations into the rich possibilities of spin-orbital liquid phases in other compounds with non-Kramers' ions.

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Competing interests

The authors declare no competing interests.