SPIN LIQUIDS

FROM THEORY TO EXPERIMENT: FINDING NEW MATERIALS

A central aim in condensed matter physics is to understand and classify possible phases of matter. Everyday examples of such phases are gases, liquids and solids, and we know that nearly everything becomes solid at sufficiently low temperature. The sole exceptions to this rule are the two isotopes of helium, in which quantum fluctuations are so large that a crystalline lattice cannot form at ordinary pressures, even at the absolute zero of temperature.

The features that distinguish a solid from a liquid or gas arise microscopically from the fact that its atoms have a fixed arrangement in space. Alternative phases of matter can be characterised by degrees of freedom other than the positions of atoms. In magnetic materials, the relevant degrees of freedom are the orientations of the atomic magnetic moments, or *spins*. At sufficiently high temperatures, spins in these systems are thermally disordered and the materials are in the paramagnetic phase – the magnetic equivalent of a gas. At low temperature, many magnetic materials have ordered arrangements of spin orientations, analogous to the regular positions of atoms in a crystal. For example, the macroscopic magnetism of a fridge magnet is due to a net alignment of its atomic magnetic moments.

What happens in a magnetic material when quantum fluctuations are large? During the last few years some answers have taken shape to this question, which has loomed over condensed matter physics for almost half a century. As hinted at by the analogies, quantum fluctuations can be large enough in some magnetic systems to maintain a liquid-like state of spins even at zero temperature, and we are now beginning to understand the properties of these spin liquid phases. Progress has been made by theorists using pencil, paper and computers to study simplified models, and by experimentalists who synthesise and characterise new materials.

Spin liquids turn out to be even *more* interesting than the remarkable superfluid phases of liquid helium (at least in the doting eyes of condensed matter physicists!). In particular, we expect some of them (we now know that there are many different types) to have what are known as *fractionalised excitations*. By this we mean that the microscopic constituents of the material – spins or electrons – correlate their motion with one another so strongly that one can no longer describe the collective behaviour in terms of individual electrons or spins. In effect, the original particles dissolve into a quantum soup and are lost. In their place, when we probe the material (for example, using magnetic neutron scattering to make excitations) we find new particles with quantum numbers corresponding to fractions of the original constituents.

At present, physicists have only a few fully certified examples of phases of matter that show fractionalisation. Perhaps the most spectacular occurs in the so-called *fractional quantum Hall effect*. In that instance, electrons (which are indivisible particles from the viewpoint of high-energy physics) break into pieces, each with a fraction (one third in the simplest case) of the elementary charge. A second example arises in magnetic materials that have one-dimensional arrangements of spins. The geometry makes this the easiest case to visualise, as explained in the box below. We hope that spin liquids will give us many more examples of fractionalisation. The search for spin liquids is very much curiosity-driven basic physics. It sets out to employ the enormous scope offered by the periodic table of elements and materials science, as a way of realising some of the astonishing consequences of quantum mechanics. Oxford physicists have been at the centre of these international efforts. Prof Radu Coldea's group has been concerned with finding and studying new materials that are candidate spin liquids [1, 2], while Prof John Chalker, Dr Dmitry Kovrizhin and collaborators have done calculations to predict what should be seen in experiments on these phases [3].

[1] R Coldea, D A Tennant and Z Tylczynski, *Phys Rev B* **68**, 134424 (2003).

[2] A Biffin, R D Johnson, I Kimchi, R Morris, A Bombardi, J G Analytis, A Vishwanath and R Coldea, *Phys Rev Lett* **113**, 197201 (2014).

[3] J Knolle, D L Kovrizhin, J T Chalker and R Moessner, *Phys Rev Lett* **112**, 207203 (2014).

Fractionalisation: splitting the electron

In some types of quantum matter, new excitations or *quasiparticles* can emerge, which are quite different from the original building blocks. This is most easily pictured for a one-dimensional quantum antiferromagnet. Magnetic materials host atomic magnetic moments, which arise from orbital currents of electrons and from the intrinsic magnetic moments of electrons. In a one-dimensional version of such a material these moments – known as spins – are arranged in well-separated chains. Interactions between neighbouring spins within a chain favour opposite alignment, and interactions between spins in different chains are negligible. The ground state for a system of this type is shown (schematically!) as the top row of fig 1a. Adding energy to the system, we can reverse one of the spins, as depicted in the middle row of fig 1a. Contrary to the obvious expectation, this process generates not one excitation, but two independent ones. These excitations, known as *spinons*, are not the reversed spin (coloured red in fig 1a), but rather the domain walls between parallel spins (pink planes in fig 1a). Two of these are created when a single spin is reversed; by inserting a segment of reversed spins, the two spinons can be separated without energy cost, as illustrated in the bottom row of fig 1a. These excitations have been observed in the material Cs$_2$CuCl$_4$ using inelastic neutron scattering. Some of the resulting data appear in fig 1b, which shows the response of the material as a function of the energy and momentum transferred to it.

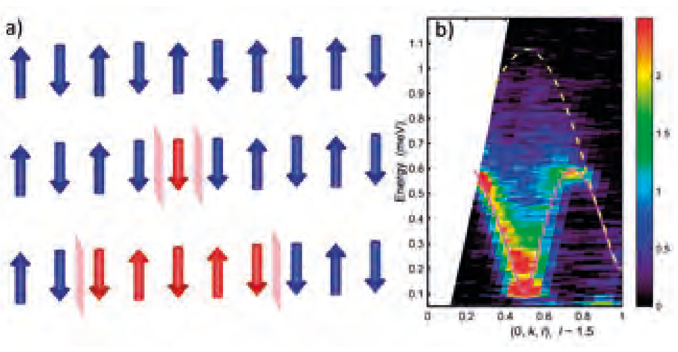

Fig 1: a) Ground and excited states of a spin-1/2 antiferromagnetic chain. b) Inelastic neutron scattering from pairs of spin-1/2 spinons in Cs$_2$CuCl$_4$ [1].

Prof John Chalker

Prof Radu Coldea

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Designer materials

What is a good strategy for finding spin liquid materials? We should avoid systems in which interactions favour a limited number of regular, solid-like spin configurations. Instead we require *frustration*. Then there may be many classical configurations that provide equally good compromise solutions to the problem of minimising the energy of the system, and a quantum ground state that is a superposition of these different configurations.

A fruitful route to frustration is shown in fig 2b. Consider first the interaction between a pair of spins, and suppose that energy is minimised only when both are aligned along a specified axis, as shown in fig 2b (top). Then either spoiling the co-alignment (fig 2b middle) or aligning along a different axis to the specified one (fig 2b bottom) will cost energy. Frustration arises if we take a lattice of such spins, with interactions favouring different axes for different pairs. An example is the honeycomb lattice shown in fig 2a, where the red, green and blue links indicate three competing preferred directions.

Models of this type were suggested by the Caltech physicist Alexei Kitaev and they turned out to be exactly solvable. At first, they seemed simply to be theorists' toys, amenable to a beautiful mathematical analysis, but far from reality. Remarkably, however, it turns out that transition metal ions with strong spin orbit interaction may have the key ingredients to realise such physics.

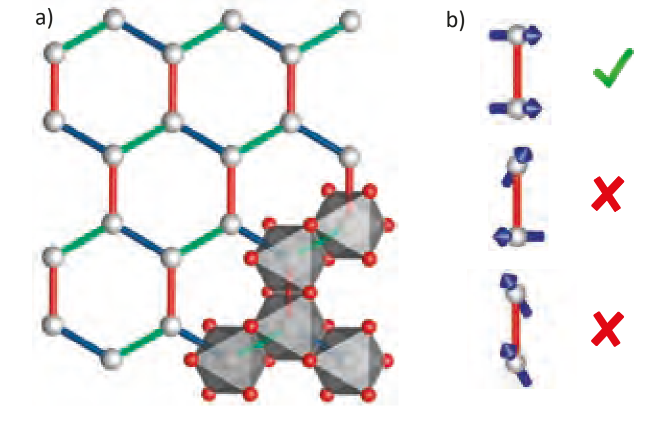

Fig 2: Kitaev model on the honeycomb lattice as described in the text.

For Ir^{4+} ions inside edge-sharing cubic octahedra of oxygen ions, as shown in fig 2a, the strong spin-orbit coupling at the Ir site ensures that the magnetic exchange interaction is firmly linked to the direction of the electronic orbitals making up the Ir-O-Ir bond. Edge-sharing octahedra forming a honeycomb arrangement represent a very stable structural framework, found in several layered honeycomb materials, including Na$_2$IrO$_3$, α-Li$_2$IrO$_3$ and α-RuCl$_3$, where in the latter the magnetic ion is Ru^{3+}.

The materials studied so far develop solid-like magnetic order at low temperatures, thought to be stabilised by additional interactions that co-exist with a dominant frustrated exchange. The ordering patterns are rather unusual: see fig 3a. They give direct evidence that Kitaev interactions indeed exist in nature and stabilise unconventional magnetism. If we can modify the materials or the experimental conditions so that additional interactions no longer interfere, we expect a spin liquid with fractionalised quasiparticles as theoretically predicted. Realising such a phase experimentally would open up completely new avenues to explore and manipulate fractionalised phases of quantum matter. The current research frontier in the field is the exploration of the phase diagram of candidate Kitaev materials as a function of pressure-tuning, bond-angle deformation via doping or inter-layer intercalations, or magnetic field tuning, hunting for signatures of such a spin liquid.

Fig 3 (below): a) Counter-rotating spiral magnetic order in the 3D honeycomb lattice in γ-Li$_2$IrO$_3$ stabilised by Kitaev interactions [2]. b) X-ray diffraction data on two different magnetic Bragg peaks used to deduce the magnetic structure in a) (solid lines are fits to the model).

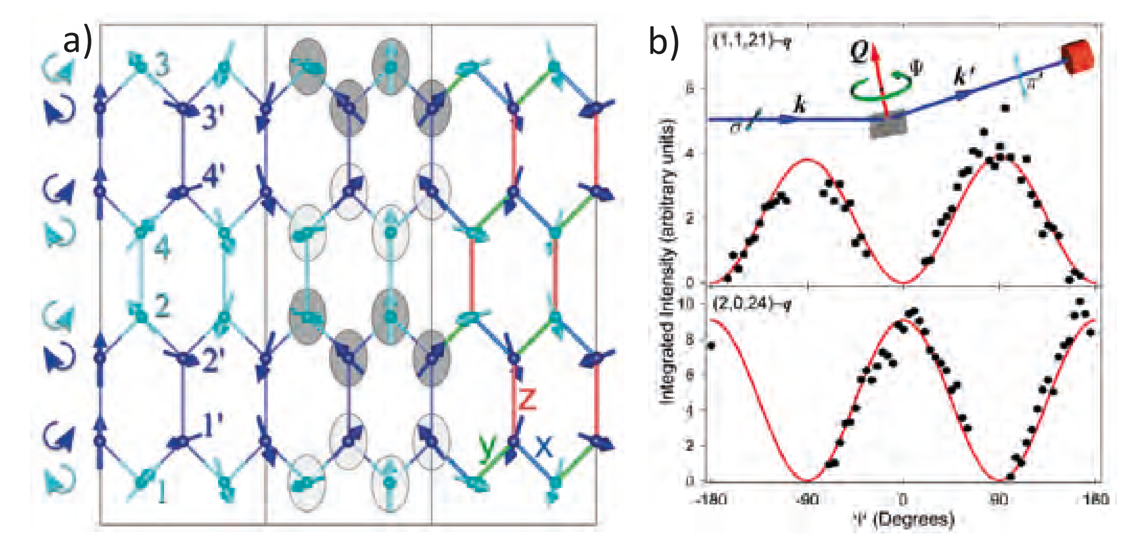

Fig 3 data were collected using instrumentation at the Diamond synchrotron at the Rutherford Appleton Laboratory.