Spin ice: a new concept in statistical physics

Studies of the pyrochlore compound $Ho_2Ti_2O_7$ are shedding new light on long-standing problems concerning spin-glass like behaviour in chemically-ordered compounds. A model of magnetic interactions analogous to proton ordering in ice has been developed to describe its behaviour. Evidence for this 'spin ice' description has been provided by recent neutron scattering results taken on PRISMA operating in diffraction mode.

The pyrochlore lattice antiferromagnet is the paradigm example of a system containing strongly-competing interactions. Its tetrahedral geometry leads to intense frustration of the magnetic bonds, and a number of unusual effects that present challenges to the modern understanding of co-operative phenomena. Most notably, many pyrochlore compounds display spin-glass transitions in the apparent absence of chemical disorder, in direct contradiction to well-established theory.

In contrast, the pyrochlore *ferromagnet* would seem at first sight unfrustrated and conventional. However, we have discovered that magnetic anisotropy can have the surprising effect of reversing the roles of ferro- and antiferromagnetic exchange couplings with regard to frustration, such that the ferromagnet is highly frustrated and the antiferromagnet is not. The frustrated ferromagnet is closely related to the problem of proton ordering in ice considered by Pauling in his classic text *The Nature of the Chemical Bond*; for brevity we refer to the model of the frustrated ferromagnet as the 'spin ice' model. Perhaps most significantly, it undergoes spin freezing as a natural consequence, and thus shows a way to resolve the some of the outstanding contradictions between theory and experiment in this area.

In the inset to figure F14.1, we show the ground state of a single tetrahedron from the pyrochlore lattice, with ferromagnetic coupling and anisotropy constraining the spins to point along the <111>-type directions. These are the directions which connect a spin with the centre of its tetrahedron. The resulting 'two spins in, two spins out' configuration is topologically equivalent to the ice rules, so this is the spin ice ground state. Bulk magnetisation, μSR, and neutron scattering work has demonstrated that the pyrochlore $Ho_2Ti_2O_7$ is a good realisation of the spin ice model. In this article, we concentrate on the most convincing results, obtained recently using PRISMA.

In the main part of figure F14.1, we show the inverse susceptibility of $Ho_2Ti_2O_7$ compared to the results of a Monte Carlo calculation for the spin ice model, which is shown as the full line. The dashed line shows a fit to the data of the simple Curie-Weiss law, appropriate for a conventional (mean field) ferromagnet; the extracted Curie-Weiss temperature is +1.9 K, indicating ferromagnetic exchange of the order of J~1 K. The spin ice inverse susceptibility is linear at high temperatures with a positive intercept (reflecting ferromagnetic nearest-neighbour coupling), but deviates for temperatures below 10 K, so as to extrapolate to zero as $T \rightarrow 0$, as if the system was a paramagnet with no coupling. No phase transition to an ordered state is observed in the simulations at any temperature. Now comparing the simulation with the experimental data from $Ho_2Ti_2O_7$, the agreement is clearly excellent over the

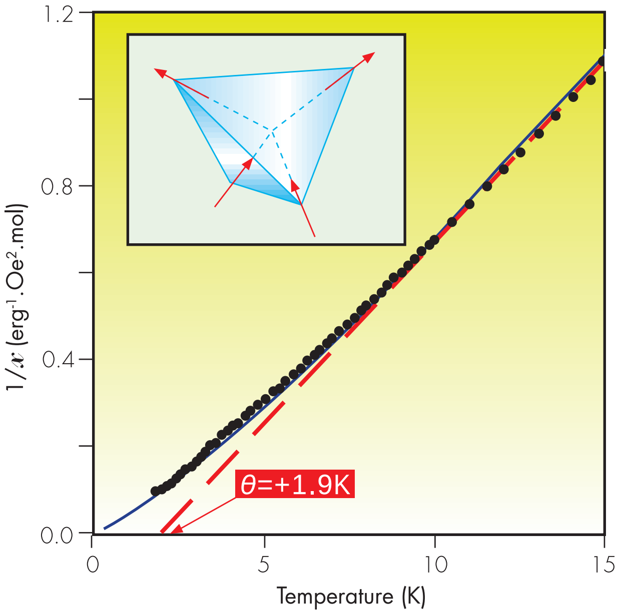

Figure F14.1. The inverse susceptibility of $Ho_2Ti_2O_7$, with a Monte Carlo simulation of the spin ice model shown as a blue line, and the standard Curie-Weiss law as a red line. The inset shows the ground state of a single tetrahedron of spins with ferromagnetic coupling and <111> anisotropy.

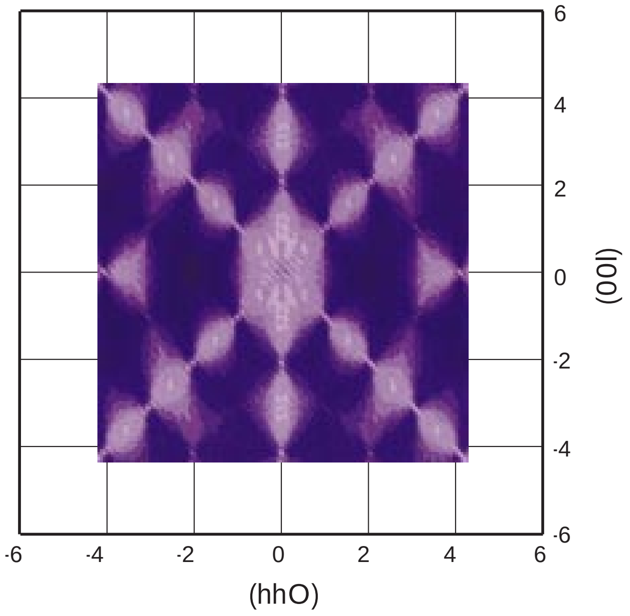

Figure F14.2. The expected magnetic neutron scattering from the spin ice model at a temperature of T/J=0.05.

temperature range covered. However, due to the fact that the lowest temperature accessible was only 1.8 K, this cannot be considered to be a true test of the ground state properties of the spin ice model.

A much more thorough test requires a neutron scattering measurement of the spin correlations at low temperatures. Since our Monte Carlo calculations predict that there is a Schottky anomaly in the heat capacity at $T \sim J/3$, the ground state properties will only be probed at temperatures considerably lower than this. The acquisition of a new dilution refrigerator by ISIS in March this year provided the ideal opportunity to do just this experiment.

In figure F14.2 we show the results of a Monte Carlo simulation of the magnetic neutron scattering from the spin ice model, for the (h,h,l) plane of reciprocal space at a temperature of $T \sim J/20$. The prominent features are maxima around the (0,0,0) and (1.5,1.5,1.5) points, and a series of connected maxima along the **c***-axis. No Bragg peaks are present, only diffuse scattering, reflecting the fact that the spin ice model is so strongly frustrated that no transition to long-range order is expected at any temperature.

In figure F14.3 we show the diffuse magnetic neutron scattering from $Ho_2Ti_2O_7$, measured using PRISMA operating in its diffraction mode. Unfortunately no sensor could be attached to the sample, but the temperature was estimated to be in the range 0.05 to 0.1 K, corresponding to $T \sim J/15$. The very intense spots of scattering are nuclear Bragg peaks, which showed no significant change in intensity on cooling from 300 K to base temperature. Thus there is no magnetic phase transition down to these temperatures. Instead of Bragg scattering, magnetic diffuse scattering is observed, very similar in form to the Monte Carlo calculation shown in figure F14.2. In particular, note the broad peaks at (0,0,3) and (1.5,1.5,1.5). There are some differences between theory and experiment, particularly in the way that the peak at (1.5,1.5,1.5) is more smeared out in the experimental data than in the prediction.

At present, we are attempting to resolve these small differences, but the excellent overall agreement between theory and experiment shows clearly that $Ho_2Ti_2O_7$ corresponds to an almost ideal realisation of the spin ice model. With this knowledge, we can now proceed to studies of the spin dynamics, which (we anticipate) should clear up some of the long-standing problems concerning spin-glassiness in chemically-ordered magnets.

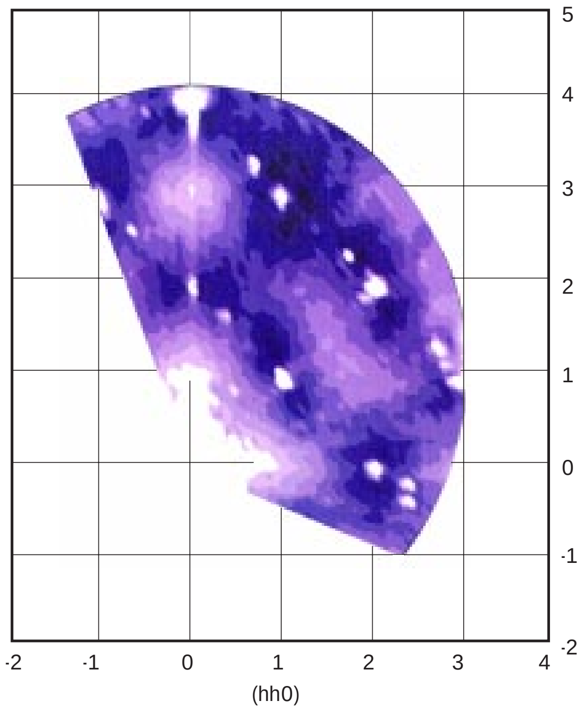

Figure F14.3. The neutron scattering observed from $Ho_2Ti_2O_7$, using PRISMA. The large diffuse features are magnetic scattering, and the sharp peaks are nuclear Bragg peaks.

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