Experimental report

15/02/2018

Proposal: 4-05-641 **Council:** 4/2016

Title: Time-of-flight inelastic neutron scattering investigation of the Quantum Spin Ice and Quantum Kagome Icephases in Pr2Hf2O7

Research area: Physics

This proposal is a new proposal

Main proposer: **Romain SIBILLE**

Experimental team: Nicolas GAUTHIER
Michel KENZELMANN
Romain SIBILLE

Local contacts: Jacques OLLIVIER

Samples: Pr2Hf2O7

Instrument	Requested days	Allocated days	From	To
IN5	8	8	14/12/2016	22/12/2016

Abstract:

We have recently reported the low temperature magnetic properties of the pyrochlore Pr2Hf2O7. Polycrystalline and single-crystal samples were investigated using time-of-flight neutron spectroscopy and macroscopic measurements, respectively. The crystal field splitting produces a non-Kramers ground state doublet for Pr3+, with Ising-like anisotropy. Below 0.5 K, the system enters a correlated state with spin-ice like configurations. We have observed the development of elastic magnetic scattering and of a discrete inelastic excitation in the neutron spectra within this regime. We want to characterize the momentum dependence of both contributions using IN5 and a single crystal sample. In addition, the transition at 2.4 T is reminiscent of the kagome ice state observed in classical spin ices for fields applied along the [111] direction. We want to characterize this other phase using the same method.

Experimental report for experiment 4-05-641 on IN5

"Time-of-flight inelastic neutron scattering investigation of the Quantum Spin Ice and Quantum Kagome Ice phases in $Pr_2Hf_2O_7$"

- First part of the experiment: Quantum Spin Ice in $Pr_2Hf_2O_7$

We have measured the scattering from a single-crystal of the candidate quantum spin ice material $Pr_2Hf_2O_7$ on IN5, at temperature of 0.05 K and 50 K, using an incident wavelength of 5.5 Å. The resolution at the elastic position was 0.050 meV. This setting together with a careful subtraction of the high temperature data and absorption correction, have produced very high quality data that allow us to make new statements and confirm the quantum spin ice (QSI) ground state of this material. The results are submitted to Nature Physics (arXiv:1706.03604)
Our results indicate that the correlated regime appearing at T < 0.5 K in macroscopic measurements is characterized by the development of distinctive features, on both powder and single-crystal samples: *(i)* a quasi-elastic contribution whose structure factor conforms with spin ice correlations (Fig. 1**a**), and *(ii)* inelastic scattering that we attribute to the monopole excitations of the QSI (Fig. 2).
A close look at the quasi-elastic signal measured at 0.05 K on IN5 (Fig. 1**a** and data points with error bars on Fig. 1**c**) indicates that the pinch-points are broadened compared to a classical near-neighbor spin ice model (dashed red line on Fig. 1**c**). This is expected in the quantum-coherent regime of the QSI at low-temperature. Instead, our data are successfully accounted for by the lattice field theory of a QSI (Fig. 1**b** and solid blue line on Fig. 1**c**). The fit to our experimental data shown on Fig. 1**c** gives a strong case that $Pr_2Hf_2O_7$ is a QSI, and allows an estimate for the speed of light associated with magnetic photon excitations (c ~ 3.6 m/s).

Figure 1: Quasi-elastic scattering obs. in $Pr_2Hf_2O_7$ (**a**) and calc. using a QSI model (**b**) **[6]**. Panel **c** shows a cut in panel **a** (grey points with error bars) compared to calculations for the QSI model (solid blue line) and classical near-neighbor spin ice model (dashed red line).

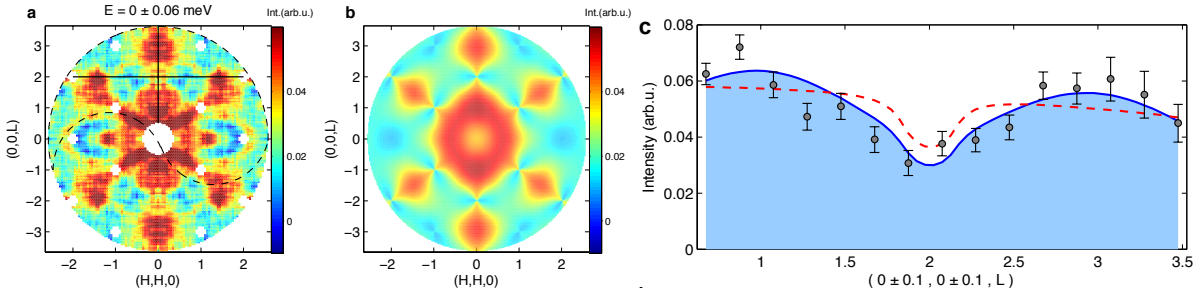
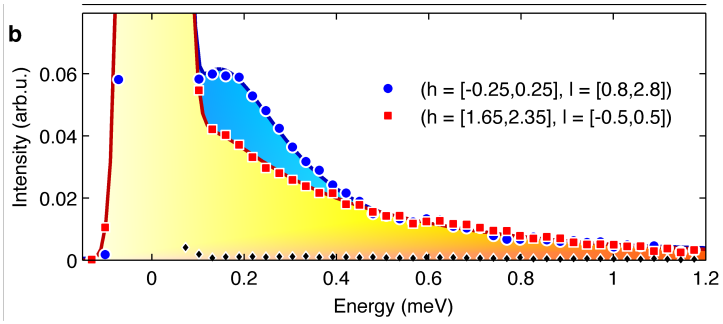


Figure 2: Inelastic cut through the IN5 data measured on $Pr_2Hf_2O_7$.



- Second part of the experiment: Quantum Kagome Ice in $Pr_2Hf_2O_7$

We have measured, using the same setting of the instrument as above, another sample of $Pr_2Hf_2O_7$, mounted with the <111> direction vertical, inside a vertical cryomagnet.
We are currently working on the analysis of the data – an overview is shown on Figure 3.

Figure 3: IN5 data measured on $Pr_2Hf_2O_7$ at 0.05 K and 2.4 T // [111].