Experimental report

02/09/2022

Proposal: **5-53-309** **Council:** 4/2021

Title: Correlations of disorder-induced quantum spin ice Ho2(Ti0.9Hf0.1)2O7

Research area: Physics

This proposal is a resubmission of 5-53-306

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Samples: Ho2ScTaO7

Instrument	Requested days	Allocated days	From	To
D7	8	6	02/09/2021	08/09/2021

Abstract:

Following a theoretical study, we propose to investigate possible disorder-induced Quantum Spin Liquids. To do so, a series of non-Kramers rare-earth pyrochlore was synthesized and characterized using XRD, NPD and magnetic susceptibility. In the set of samples, the density of Frenkel pair defect is tuned through the admixing of different quadrivalent cation on the pyrophore B site. Experimental observations corroborate the persistence of spin ice correlations along with strengthened fluctuations due to disorder-induced transverse fields. The aim of this experiment is to precisely measure the spin ice scattering in Ho2(Ti0.9Hf0.1)2O7 in order to compare with Ho2Ti2O7 classical spin ice and theoretical expectations for quantum spin ice.

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Correlations of disorder-induced spin ice $Ho_2(Ti_{0.9}Hf_{0.1})2O_7$

Note:
The quality of the sample of $Ho_2(Ti_{0.9}Hf_{0.1})2O_7$ that we intended to use for this experiment was found not to be good enough (due to the presence of several crystallites) just before the D7 experiment.
For this reason, a last minute sample change has been made with the approval of Andrew Wildes (local contact) and Mark Johnson.
The studied sample is another B-site disordered holmium pyrochlore, $Ho_2(Sc/Ta)_2O_7$ in which the physics at play is very similar (effect of B-site disorder on the magnetism of an holmium spin ice, and possible interplay with correlated structural disorder).
The experimental plan remained the same.

Spin ice, $Ho_2Ti_2O_7$, is a well-studied example of frustrated magnet. At low temperatures, it is a Coulomb phase with emergent algebraic correlations while its low-energy excitations are shown to be magnetic monopoles. The diffuse scattering spectrometer D7 has be used in the past to measure such correlations [1]. In the case of the classical spin ice the Ho^{3+} spins are anisotropic with their easy-axis pointing along the <111> local axis. It is proposed that structural disorder may add a transverse component to the spin (by modifying the crystal electric field potential) which in turn could enhance the quantum excitations and realize the quantum spin ice (QSI) state [2].
Here we used the diffuse scattering spectrometer D7 at ILL with the polarization analysis with two polarization channels: non-spin flip (NSF) and spin flip (SF) with the incident polarization vertical (z). The sample was a single crystal of Ho_2ScTaO_7 and was mounted on a copper finger and cooled in a dilution refrigerator in a standard Orange cryostat. The crystal was aligned with the [h,-h,0] axis vertical to provide a scattering plane containing (h, h, l) wavevectors. Reciprocal space was surveyed by rotating the crystal about the vertical axis. The efficiencies of the polarization elements were corrected using a standard silica glass ("quartz") sample; and a vanadium sample was used to correct for detector efficiencies. Weak Bragg peaks are visible in the spin flip scattering. These are due to analyzer inefficiency at very intense Bragg scattering positions which cannot be corrected by the quartz. The angular dependence of the sample transmission and its modification of the background scattering prevents us from correctly subtracting the instrumental background so the data are not normalized to absolute units.
The chosen geometry is particularly favorable in measuring anisotropic pyrochlores such as spin ice, as the anisotropy of the spins means that two spins on each tetrahedron lie in the scattering plane. The spin flip scattering is due to spin components perpendicular to both the scattering vector and the polarization. At a general scattering vector in the plane, all four spins on a tetrahedron contribute to

the spin flip scattering which therefore contains contributions from correlations between all four sublattices. The non-spin flip scattering is due to spin components perpendicular to the scattering vector but parallel to the polarization, so only two spins per tetrahedron contribute in this channel. The contribution due to the ice rule correlations is separated from the total diffuse scattering.

Compared to $Ho_2Ti_2O_7$, the crystal field excitations of Ho_2ScTaO_7 (HSTO) are at lower energies, the lowest crystal field level is almost an order of magnitude lower in energy; this could lead to mixture of ground state and the excited states giving rise to a transverse spin component (like the case of QSI candidate $Tb_2Ti_2O_7$).

The map of reciprocal lattice from the SF and NSF channels are shown in figure 1, the presence of pinch points in the SF map suggests the existence of spin ice correlations, however the broadness of the pinch points could suggest the enhancement of excitations. The cut through the (0,0,2) position is shown in the figure 2, the signal in the SF and NSF channels is fitted with a Gaussian convoluted Lorentzian. As seen in the total scattering cut, there is a very subtle signature of the pinch point in the unpolarised data.

Data analysis is ongoing, the absorption correction could help smoothing up the data especially the NSF scattering, modelling and finding the Hamiltonian are the next steps.

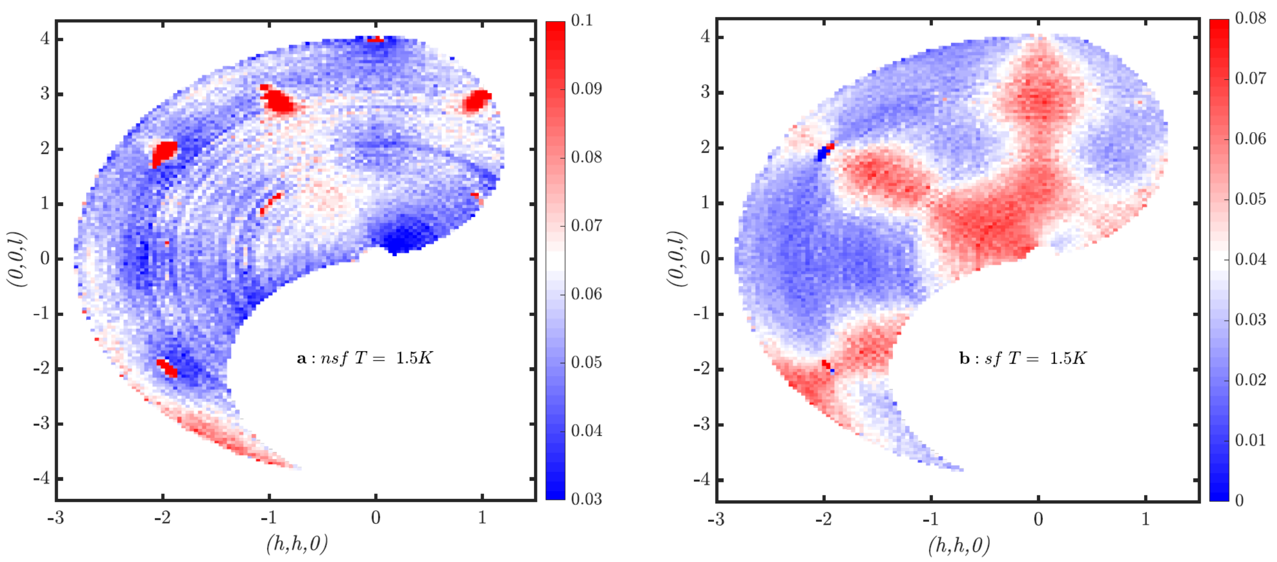

Figure 1: a) Non spin flip scattering in HSTO at 1.5 K, showing similar features observed in $Ho_2Ti_2O_7$ which are due to long-range dipolar interactions. b) Spin flip scattering shows the broadened pinch points at (1,1,1) and (0,0,2) positions.

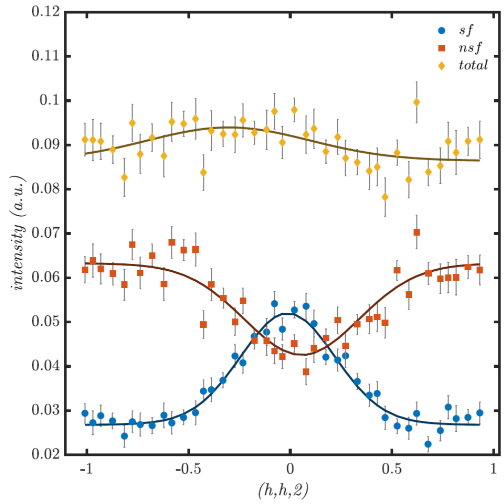

Figure 2: Cuts through the (0,0,2) pinch point in different polarization channels (T= 1.5 K).

Reference:

[1] T. Fennell, P. P. Deen, A. R. Wildes, K. Schmalzl, D. Prabhakaran, A. T. Boothroyd, R. J. Aldus, D. F. McMorrow, and S. T. Bramwell, *Magnetic Coulomb Phase in the Spin Ice Ho2Ti2O7*, Science **326**, 415 (2009).

[2] L. Savary and L. Balents, *Disorder-Induced Quantum Spin Liquid in Spin Ice Pyrochlores*, Phys. Rev. Lett. **118**, 087203 (2017).