Experimental report

16/09/2016

Proposal: 5-53-253 **Council:** 4/2015

Title: Diffuse Scattering in the New Rare Earth Pyrochlores Pr2Hf2O7 and Nd2Hf2O7

Research area: Physics

This proposal is a new proposal

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Samples: Nd2Hf2O7
Pr2Hf2O7

Instrument	Requested days	Allocated days	From	To
D7	12	12	20/10/2015	01/11/2015

Abstract:

The aim of the proposed experiment is to explore the magnetic behavior of the new rare earth Pyrochlores Pr2Hf2O7 and Nd2Hf2O7 by using D7 to measure the diffuse magnetic scattering. Both compounds promise interesting new physics. Pr2Hf2O7 may realise the long-sought-after quantum spin ice state expected for Pr3+ on a Pyrochlore lattice. This compound shows complete absence of long-range magnetic order despite substantial spin correlations revealed by AC susceptibility which verify a spin liquid ground state. In contrast, Nd2Hf2O7 develops long-range magnetic order with an all-in-all-out magnetic structure revealing that the magnetic interactions are antiferromagnetic rather than ferromagnetic as found in spin ice. Here strong diffuse scattering is also expected due to the highly suppressed ordered moment.

Title: Measurement of single crystals $Pr_2Hf_2O_7$ and $Nd_2Hf_2O_7$ at D7 Spectometer of ILL_ 20.10.2015-02.11.2015

Background:

The Rare Earth pyrochlore oxides $RE_2^{3+}B_2^{4+}O_7$ where RE is a Rare Earth ion are extensively studied due to the exotic ground states occurred from geometrical frustration. The RE site forms a network of corner-sharing tetrahedra which is the quintessential lattice for geometrical frustration, and the natural tendency for long-range magnetic order is suppressed resulting in new physics. Here we investigate two new pyrochlore systems with Hf^{4+} the nonmagnetic ion, $Nd_2Hf_2O_7$ and $Pr_2Hf_2O_7$ which have shown by macroscopic measurements of powder samples that they are governed by high anisotropic magnetic moments. There are no published magnetic studies, by neutron scattering, of $Nd_2Hf_2O_7$ and $Pr_2Hf_2O_7$ single crystals.

$Nd_2Hf_2O_7$

So far studies have been performed only to powder samples and have been mainly focused on the crystallographic and magnetic bulk properties. The crystal field has trigonal symmetry about [111] (or equivalent) and we therefore expect the magnetic moment to aligned along this direction. AC susceptibility shows an AFM transition at T_N=0.55K confirmed by neutron powder diffraction. The refinement of neutron diffraction pattern reveals an all-in all-out spin configuration. The analysis of χ_{dc}(T) data reveal a positive Curie Weiss temperature, suggesting FM spin interactions.

$Pr_2Hf_2O_7$

$Pr_2Hf_2O_7$ powder sample investigation with macroscopic and diffraction techniques reveal also high anisotropy with spins aligned along the [111] direction. The absence of ordering down to base temperature combined with the weakening of Ising anisotropy suggests that quantum tunneling is allowed which introduces spin dynamics. This system is proposed as a new quantum spin ice candidate.

The aim of this experiment

The neutron diffraction measurement of these single crystals, with polarized neutrons, would shed light onto the spin correlations at base temperature, while it is possible the qualitative estimation of the correlation lengths. Also it would reveal possible magnetic transitions that were not possible to be identified so far.

Experimental Setup:

The crystals have been aligned by Laue diffractometer with the symmetry axis [1-10] vertical in the white neutron beam. The energy of the incident neutron beam was =3.47 meV, and the temperature range T= 25-0.06K in zero magnetic field. The XYZ-polarization analysis technique provides separation of the magnetic cross section. The samples have been covered with a thin, oxygen free Cu foil and mounted on a Cu sampleholder in order to allow thermal conductivity with the dilution stick.

Results:

Nd2Hf2O7

In Fig. 1 the magnetic scattering cross section, after the XYZ analysis, at om= 180.1-280.1° for both tth positions of the detectors, at T=60mK for the Z flipper On, is presented. The background seems to mask the diffuse scattering which we expect to be weak due to the strongly reduced value of the magnetic moment. The T=15K data have been used as a background. Both high and the low temperature data have been normalized to the NSI compound of high temperature data after we have erased from the high temperature data all the contaminated by Bragg peaks data. That is the best result, between many background subtraction techniques, that we have succeeded.

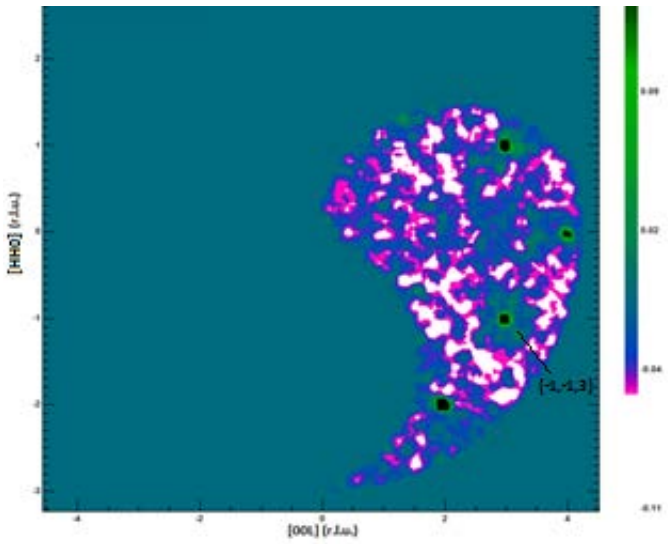

Fig. 1: Q-map of diffuse scattering experiment of NHO in the (hhl) plane for Z-spin-flip polarization, measured at T=700mK after high temperature background subtraction.

The intensity of the (113) and (-1-13) peaks have increased in comparison to the data at $T>T_N$, which proves that they have also magnetic origin. The comparison with $Nd_2Zr_2O_7$ results suggest that they identify the all-in all-out spin configuration. Hints of diffuse scattering along the (00L) direction is reminiscent of the spin ice pattern.

Pr2Hf2O7

Figure 2 present the experimental neutron scattering pattern of $Pr_2Hf_2O_7$ measured at T=0.085K, in the (hhl) plane of reciprocal space with Z flipper On, normalized by the NSI component. The data measured at T=15K have been subtracted as background after they have also been normalized by the NSI component. Diffuse scattering is presented along the (h00) and the (hhh) direction with hints of pinch points at the (00-2) and (111) positions. The diffuse scattering pattern qualitatively confirms the spin ice model, although it is not sharp as in a classical spin ice. Also, the pinch point at (11-1) is much broader, suggesting an increased amount of tetrahedral that violate the two –in two-out rule.

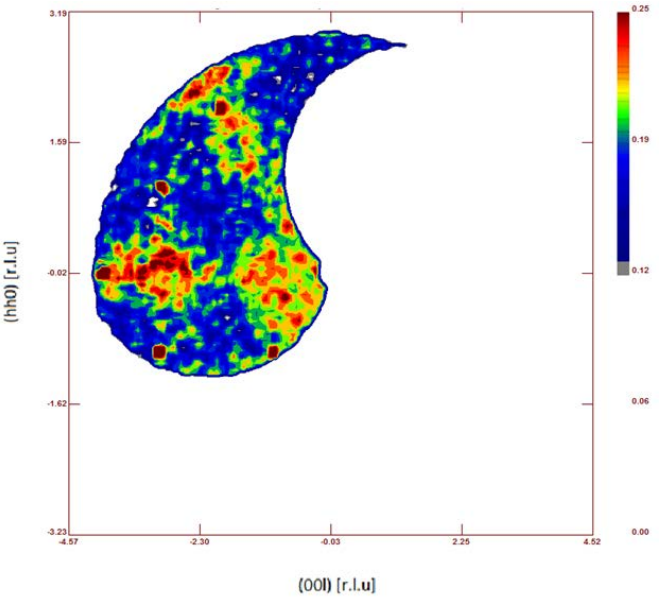

Fig. 2: Q-map of diffuse scattering experiment of PHO in the (hhl) plane for Z-spin-flip polarization, measured at T=85mK with the high temperature (T=15K) data subtracted as background