Experimental report

04/05/2022

Proposal: 5-53-295 **Council:** 4/2020

Title: Role of strain in inducing quantumfluctuations in Pr2ScNbO7

Research area: Physics

This proposal is a new proposal

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Samples: Pr2ScNbO7

Instrument	Requested days	Allocated days	From	To
D7	7	7	27/05/2021	03/06/2021
IN3	1	1	21/05/2021	22/05/2021

Abstract:

The most compelling evidence to date of quantum spin-ice correlations has been provided by studies of Pr2Zr2O7 and Pr2Hf2O7, and structural disorder is believed to play a crucial role. Here we propose to study Pr2ScNbO7, where the replacement of tetravalent ions by an equal number of trivalent and pentavalent ions is expected to introduce strains without the complication of introducing charge-compensating oxygen vacancies. We propose to separate the structural diffuse scattering from the magnetic diffuse scattering at low temperature using XYZ polarisation analysis on D7. The structural diffuse scattering will determine the defect structures, giving the magnitudes of ionic displacements and the nature of the strain distribution. The magnetic diffuse scattering will tell us to what extent the presence of uncorrelated spins from quantum fluctuations is due to the structural disorder. In this way, we expect to unambiguously distinguish between quantum and classical spin ice behaviour.

Scientific background
Disorder is often regarded as a nuisance in the study of spin liquids, and much effort has been devoted in the past to reduce it as much as possible. However, recent theoretical predictions suggest the presence of structural disorder can be used to stabilise classical and quantum spin liquids, and it can lead to new magnetic degrees of freedom, the formation of topological spin glasses [1] and the formation of entirely novel quantum spin liquids [2,3].

The most compelling evidence to date of quantum spin-ice correlations has been provided by studies of $Pr_2Zr_2O_7$ [4] where the presence of structural disorder comes into play. In these systems quasi-elastic pinch-point scattering, which is the signature of classical spin ice, is rather weak. Characteristic star-fish-like inelastic scattering is observed at low energy transfer, as figure 1(b) shows. No modulation is observed at higher energy transfers, as figure 1(a) shows, and this continuum of excitations has been attributed to the fractionalized excitations of a quantum spin ice ground state

Experimental report
We proposed to study highly disordered systems in order to separate the contribution from structural disorder and that from anisotropic exchange. For this, a large 5g single crystal of Pr_2ScNbO_7 was grown. The single crystal was aligned with the [1, -1, 0] direction vertical using the Laue diffractometer OrientExpress.

To study its diffuse neutron scattering, the wide-angle multi-detector diffractometer D7 was used. The sample was measured for 8 days at 50mK using XYZ polarization analysis (PA). Such long periods of measurements were needed due to the small magnetic moment of Pr. XYZ PA was needed to isolate the magnetic diffuse scattering from the structural diffuse scattering (which was used to determine the defect structure). Furthermore, because the quantum fluctuations are expected to lead to a contribution to the magnetic scattering from uncorrelated spins, we need XYZ PA to distinguish between our magnetic scattering and the spin-incoherent background.

The purely magnetic diffuse scattering is shown in figure 1(c). This measurement shows that the magnetic scattering is independent of Q. As such, the star-fish modulation in the scattering observed at low energy transfers in figure 1(b) for $Pr_2Zr_2O_7$ is absent. The scattering is instead consistent with either a paramagnet or the continuum from the quantum spin dynamics in figure 1(a).

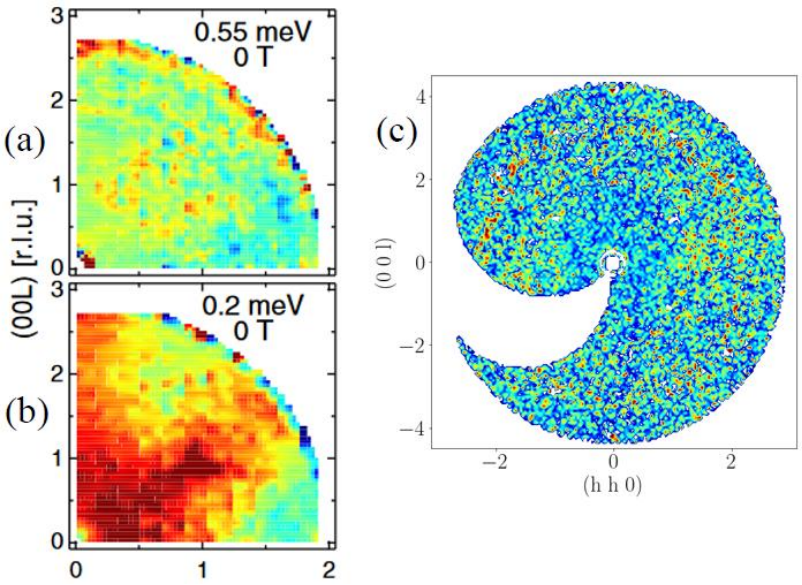

Figure 1. The inelastic magnetic diffuse scattering measured from $Pr_2Zr_2O_7$. Modulation in Q for scattering intensity in the (hhl) plane at fixed energy transfers (a) $\Delta E \sim 0.55$ meV and (b) $\Delta E \sim 0.2$ meV at T ~ 50 mK [5]. (c) Magnetic diffuse scattering for single-crystal Pr_2ScNbO_7 at T = 50 mK measured using XYZ PA and without energy analysis on D7.

The purely structural diffuse scattering is shown in figure 2(a). These results are compared with *ab initio* DFT calculations for different arrangements of Sc and Nb ions on the pyrochlore B sites. The calculated scattering from the lowest energy configuration in figure 2(b) is in good qualitative agreement with the data.

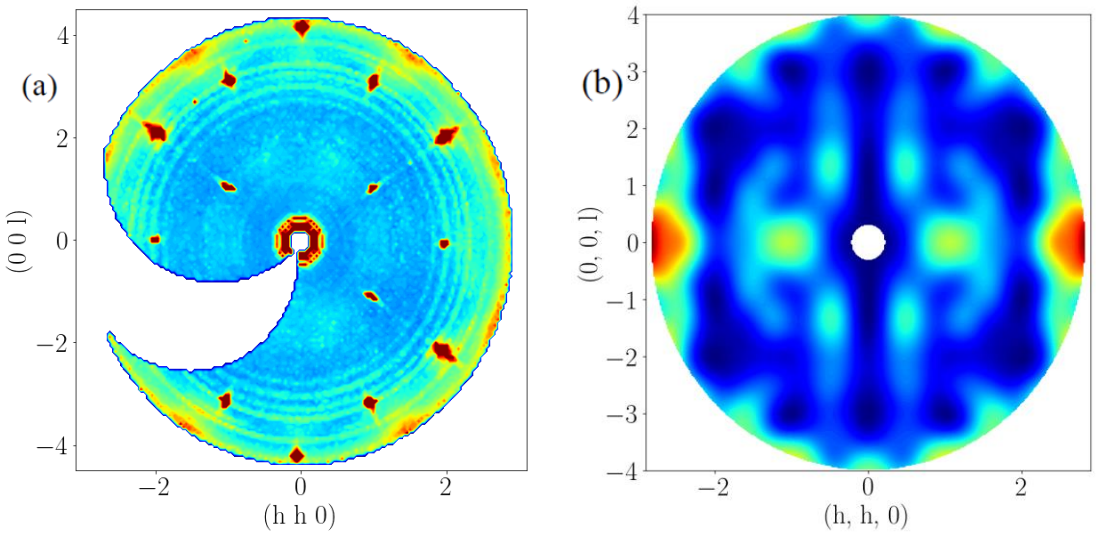

Figure 2. (a) Structural diffuse scattering for single-crystal Pr_2ScNbO_7 at T = 50 mK measured using XYZ PA. (b) Structural diffuse scattering for the lowest energy Sc-Nb configuration simulated with DFT.

Reference

[1] A. Sen et al. Phys. Rev. Lett. 114, 247207 (2015).
[2] L. Savary et al. Phys. Rev. Lett. 118, 087203 (2017).
[3] O. Benton Phys. Rev. Lett. 121, 037203 (2018).
[4] K. Kimura et al. Nat. Commun. 4, 1934 (2013).