Experimental report

18/02/2022

Proposal:	**5-42-537**	**Council:**	10/2019
Title:	Polarization Analysis of the Dynamic Quantum Spin Ice Correlations inthe Ground State of a Pyrochlore Magnet		
Research area:	Physics		

This proposal is a new proposal

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Samples: Ce2Zr2O7

Instrument	Requested days	Allocated days	From	To
D7	6	5	25/03/2021	30/03/2021

Abstract:

The rare-earth pyrochlore magnets, R2B2O7, continue to attract great interest as they can combine quantum magnetism with geometrical frustration due to the architecture of the crystal sub-lattice that the magnetic rare-earth ions reside on. The corner-sharing tetrahedral geometry of this sub-lattice allows for the possibility of strong magnetic frustration and the typical effect of this is the promotion of exotic magnetic ground states over conventional ordered ground states. This family of magnetic materials is responsible for many quantum spin liquids. This proposal focuses on the rare type of quantum spin liquid called "quantum spin ice" (QSI) with associated emergent quantum electrodynamics and exotic excitations that behave like photons and monopoles (both electric and magnetic). We've reported inelastic measurements on Ce2Zr2O7 that give compelling evidence for a QSI ground state in this material and are proposing the first (to our knowledge) polarization analysis of the diffuse magnetic scattering arising from an integration of the low-energy excitations present in the QSI phase.

We have carried out new polarized diffraction measurements on single crystal $Ce_2Zr_2O_7$ using the D7 diffractometer at the Institute Laue Langevin. This diffractometer employs a spin polarized monochromatic incident beam, which was $E_i = 3.47$ meV for this experiment. This configuration effectively integrates over $\Delta E \sim$0.16 meV during the course of a diffraction measurement. A single polarization direction, perpendicular to the $[HHL]$ scattering plane, was employed, and as such the spin flip (SF) and non-spin flip (NSF) diffuse scattering profiles can be independently measured. The diffuse scattering associated with these two cross sections, SF and NSF, are shown in the $[HHL]$ scattering plane for $Ce_2Zr_2O_7$ in Fig. 1(a) and 1(b), respectively for the temperature-difference data set $T = 0.045$ K - $T = 10$ K. For comparison, the corresponding SF and NSF diffuse scattering patterns as measured on single crystal $Ho_2Ti_2O_7$ at $T = 1.7$ K are shown in Fig. 1(c) and 1(d), respectively [1]. These earlier spin polarized diffuse scattering measurements on $Ho_2Ti_2O_7$ (Ref. [1]) played a formative role in the development of classical spin ice physics, as they drew clear attention to "pinch point" scattering within the SF cross section at (0,0,2) and (1,1,1) and equivalent wavevectors, due to the presence of a classical Coulomb phase at low temperature. These measurements on $Ho_2Ti_2O_7$ also observed zone-boundary diffuse scattering in the NSF channel, which was later attributed to the long range dipolar interactions relevant to the large Ho^{3+} dipole moments.

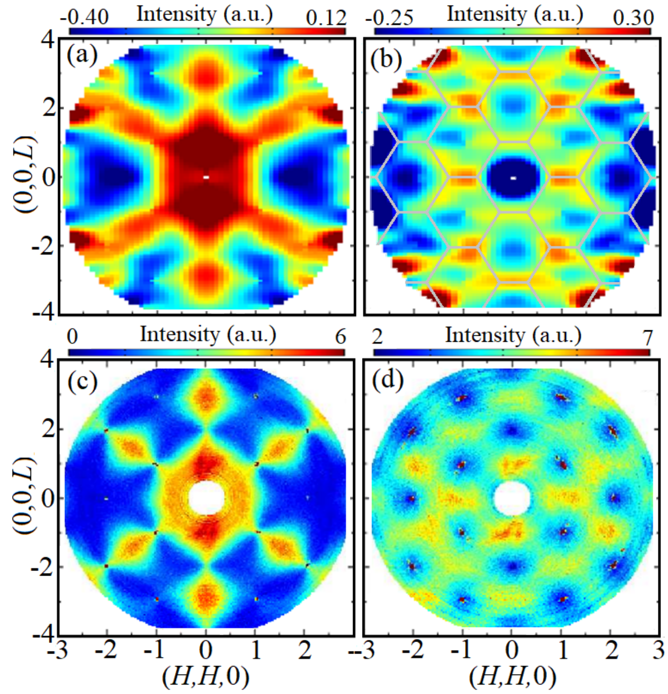

FIG. 1. The symmetrized $T = 45$ mK - $T = 10$ K temperature-difference neutron signal measured in the (a) SF and (b) NSF channels of our polarized neutron diffraction experiment on $Ce_2Zr_2O_7$. The (c) SF and (d) NSF scattering signals in the $[HHL]$ plane measured in a polarized neutron scattering experiment on $Ho_2Ti_2O_7$ at $T = 1.7$ K [1]. The data in this figure is shown in arbitrary units.

The comparison between the spin polarized diffuse scattering from $Ce_2Zr_2O_7$ and $Ho_2Ti_2O_7$ in Fig. 1 is interesting both in what is similar and where the discrepancies between the two materials lie. One may note however, that the comparison is made at quite different temperatures, 0.045 K for $Ce_2Zr_2O_7$ but only 1.7 K for $Ho_2Ti_2O_7$. In fact, the large Ho^{3+} moments and effective ferromagnetic coupling cause $Ho_2Ti_2O_7$ to depolarize the beam at lower temperatures, whereas no such issue is present for $Ce_2Zr_2O_7$ due to its much smaller Ce^{3+} moments. Quasi-pinch point SF scattering is observed near (0,0,2) Bragg positions in $Ce_2Zr_2O_7$, but it is not as constricted as that observed at (0,0,2) in $Ho_2Ti_2O_7$, even though the earlier measurements in $Ho_2Ti_2O_7$ were taken at much higher temperature. Furthermore while diffuse SF scattering extends out in (1,1,1) and equivalent directions in a snow flake-like pattern in $Ce_2Zr_2O_7$, pinch points appear to be absent in these directions.

In contrast, and somewhat surprisingly, the observed NSF diffuse scattering in $Ce_2Zr_2O_7$ is quite similar to that seen in $Ho_2Ti_2O_7$. In both cases the diffuse scattering tends to follow the face-centred cubic Brillouin zone boundaries, outlined in grey in Fig. 1(b). In $Ho_2Ti_2O_7$, this was ascribed to interactions beyond near neighbour [1], which was not surprising, given that dipolar interactions are expected to dominate over exchange interactions even for near neighbours in $Ho_2Ti_2O_7$. However, the Ce^{3+} moments are $\sim$8 times smaller than those of Ho^{3+} and hence dipolar interactions are expected to be $\sim$64 times smaller in $Ce_2Zr_2O_7$.

We also use this new polarized neutron diffraction data to compare the measured SF and NSF signals with NLC calculations using the near-neighbour exchange parameters yielded in a recent work (Ref. [2]). These best-fitting exchange parameters correspond to a quantum spin ice phase in the near-neighbour exchange ground state phase diagram relevant for $Ce_2Zr_2O_7$ and other dipolar octupolar pyrochlores [2]. The calculations are carried out at $T = 0.5$ K (see Ref. [2]), as that is the lowest temperature for which the third-order NLC calculation converges, while the new polarized neutron diffraction measurements were performed at lower temperatures, $T = 0.045$ K. Nonetheless, we assume that this calculation will capture most of the features at lower temperatures, as the ground state is disordered. In this recent work (Ref. [2]), we use the resulting near neighbour exchange parameters to calculate the equal-time spin flip (SF) and non-spin flip (NSF) structure factors in the $[HHL]$ scattering plane. This calculation resembles the new polarized neutron diffraction measurements in the SF channel from single crystal $Ce_2Zr_2O_7$, but cannot account for the observed zone-boundary diffuse scattering in the NSF channel. We attribute this discrepancy to interactions beyond near-neighbour in the Hamiltonian, which are expected to be small, and a full study of which is beyond the scope of the recent work (Ref. [2]). The same discrepancy exists for

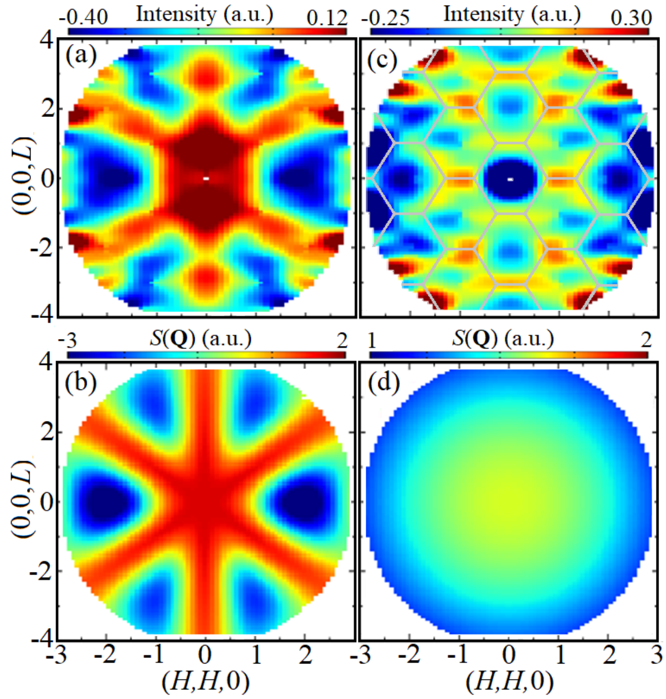

FIG. 2. (a) The symmetrized $T = 45$ mK - $T = 10$ K temperature-difference neutron signal measured in the SF channel of our polarized neutron diffraction experiment. (b) The NLC-calculated equal-time structure factor for SF scattering in the $[HHL]$ plane at $T = 0.5$ K with a $T = 10$ K temperature subtraction. (c) The symmetrized $T = 45$ mK - $T = 10$ K temperature-difference neutron signal measured in the NSF channel of our polarized neutron diffraction experiment. The grey lines show the Brillouin zone boundaries. (d) The NLC-calculated equal-time structure factor for NSF scattering in the $[HHL]$ plane at $T = 0.5$ K with a $T = 10$ K temperature subtraction. Both (b) and (d) are calculated using the experimental estimates for the A near-neighbour exchange parameters yielded in this work (see main text).

spin-polarized neutron diffraction from $Ho_2Ti_2O_7$, where it was ascribed to expected long range dipolar interactions [1].

The measured (NLC-calculated) SF scattering in the $[HHL]$ scattering plane is shown in Fig. 2(a) (2(b)) and the measured (NLC-calculated) NSF scattering in the $[HHL]$ scattering plane is shown in Fig. 2(c) (2(d)). The comparison between measurement and theory for the SF channel in Fig. 2(a) and 2(b) is good, although sharper features are present in the lower temperature, SF polarized diffraction, such as the broad pinch point scattering near (0,0,2). The measured NSF structure factor in the $[HHL]$ scattering plane (Fig. 2(c)) shows intensity that is maximal along Brillouin zone boundaries (shown as grey lines in Fig. 2(c)) and minimal at zone centres. As discussed above, this zone boundary scattering is similar to that measured in the NSF channel of polarized neutron diffraction measurements on $Ho_2Ti_2O_7$, shown in Fig. 1(d) [1], and associated with interactions beyond the nearest-neighbour. The calculated NSF structure factor is featureless for the near-neighbour-only XYZ spin Hamiltonian employed here, with a **Q**-dependence originating from the Ce^{3+} magnetic form factor only, as Fig. 2(d) illustrates.

[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* $Ho_2Ti_2O_7$, Science **326**, 415 (2009).

[2] E. M. Smith, O. Benton, D. R. Yahne, B. Placke, R. Schäfer, J. Gaudet, J. Dudemaine, A. Fitterman, J. Beare, A. R. Wildes, S. Bhattacharya, T. DeLazzer, C. R. C. Buhariwalla, N. P. Butch, R. Movshovich, J. D. Garrett, C. A. Marjerrison, J. P. Clancy, E. Kermarrec, G. M. Luke, A. D. Bianchi, K. A. Ross, and B. D. Gaulin, *The case for a* $U(1)_\pi$ *Quantum Spin Liquid Ground State in the Dipole-Octupole Pyrochlore* $Ce_2Zr_2O_7$ (2021), arXiv:2108.01217 [cond-mat.str-el].