Experimental report

28/04/2022

Proposal: **4-05-784** **Council:** 10/2019

Title: Probing Emergent Photon Excitations in a Pyrochlore Quantum Spin Ice Candidate

Research area: Physics

This proposal is a new proposal

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Samples: $Ce_2Zr_2O_7$

Instrument	Requested days	Allocated days	From	To
IN5	7	6	01/06/2021	07/06/2021

Abstract:

The rare-earth pyrochlore magnets, $R_2B_2O_7$, 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 measurements on $Ce_2Zr_2O_7$ that give compelling evidence for a QSI ground state in this material and are proposing to perform the first (to our knowledge) experimental investigation of the photon excitations that are predicted to occur in the QSI phase. An experimental confirmation of these photon excitations would be of extreme impact to the field of quantum materials research.

We have performed inelastic neutron scattering measurements on a single crystal sample of $Ce_2Zr_2O_7$ using the IN5 instrument at the Institut Laue-Langevin in an attempt to improve on existing measurements of the magnetic diffuse scattering from $Ce_2Zr_2O_7$ at low temperature.

Our 1.5g single crystal sample of $Ce_2Zr_2O_7$ was aligned in the [*HHL*] scattering plane within a copper sample mount. The 8 Å wavelength setting (E_i = 1.278 meV) was used with 8000 rpm chopper frequency in order to provide a suitable combination of flux, *Q*-coverage, and energy resolution. The corresponding energy resolution was ΔE = 0.03 meV at the elastic line and the maximum accessible *Q* corresponding to this instrumental set-up is Q_{max} = 1.435 Å^{-1}, which gives Q_{max} = 2.445 r.l.u for $Ce_2Zr_2O_7$. The experiment was conducted with zero magnetic field and a dilution refrigerator sample environment, allowing a base temperature of 50 mK. The sample was rotated within the [*HHL*] scattering plane in steps of 1° with a total of 200° covered and data was measured at temperatures of 50 mK, 0.7 K and 10 K.

Unfortunately, we were unable to resolve the expected snowflake-like pattern of scattering in the [*HHL*] plane that has been measured from $Ce_2Zr_2O_7$ in previous experiments [1, 2]. This previously measured snowflake-like pattern of scattering is shown in Fig. 1(a), along with the simulated scattering patterns for a classical spin ice and quantum spin ice in Fig. 1(b, c) that were used as comparisons with this previously measured data in Ref. [1]. Although we did not detect this clear snowflake-like pattern of scattering due to experimental noise, we did detect a diamond-shaped ring at the centre of the measured signal with a decrease in scattering towards $Q = 0$ as shown in Fig. 2, which is generally consistent with this previously detected pattern. Furthermore, cuts along particular directions of reciprocal space (not shown) yield general consistency with the expected scattering but ultimately the noise level dictates that these new measurements, despite having a better energy-resolution, do not improve on the existing measurements which show significantly less noise.

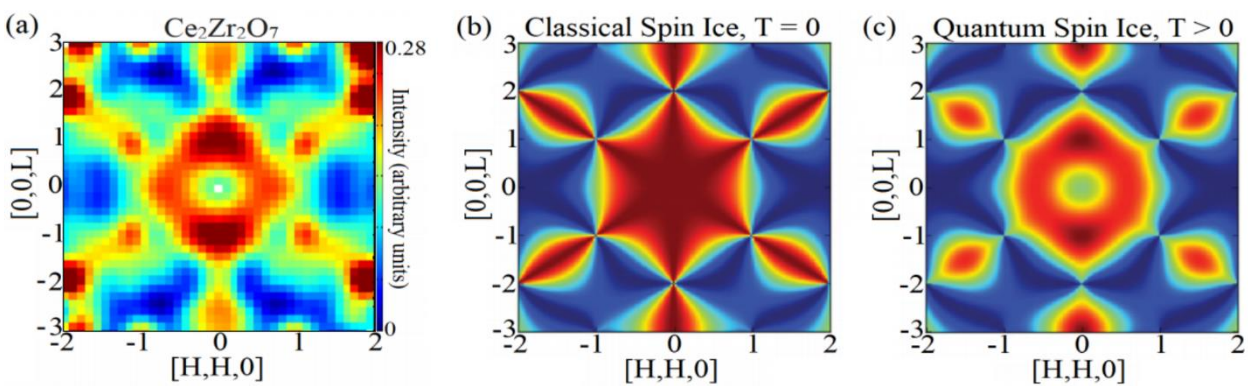

Fig. 1: (a) The snowflake-like pattern of scattering measured from $Ce_2Zr_2O_7$ in Ref. [1]. The data is shown in its symmetrized form for a T = 0.06 K – T = 2 K temperature subtraction with integration in energy-transfer over E = [0, 0.15] meV. (b) and (c) show the simulated scattering signals for a classical spin ice at zero temperature and a quantum spin ice at finite temperature, respectively [3]. This figure has been adapted from Ref. [1] with permission of the authors; the simulated signals shown here were compared with the measured signal in Ref. [1] in order to draw a conclusion of quantum spin ice behaviour in $Ce_2Zr_2O_7$ at low temperature.

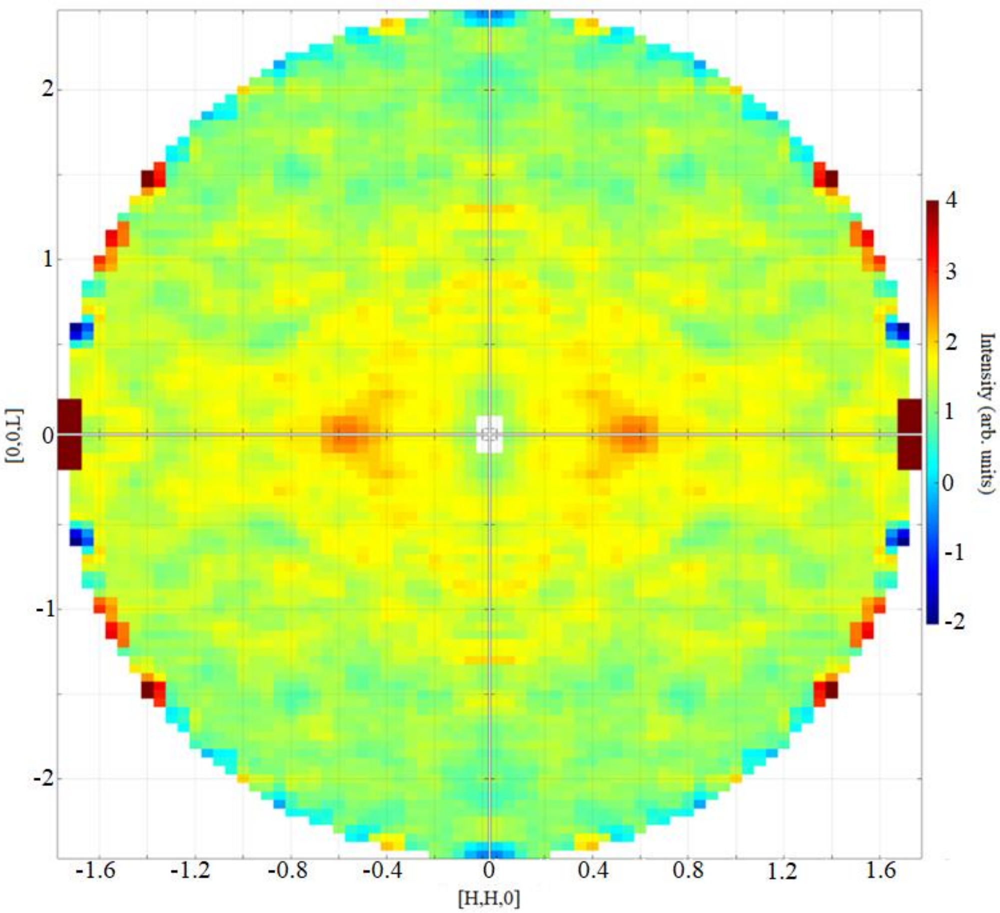

Fig. 2: The diamond-like pattern of scattering measured from $Ce_2Zr_2O_7$ in our IN5 experiment. The data is shown in its symmetrized form for a $T = 0.05$ K – $T = 10$ K temperature subtraction with integration in energy-transfer over $E = [0.05, 0.15]$ meV. This integration was chosen to enclose the dominant portion of the measured inelastic scattering while avoiding contamination from noise in the elastic channel.

References:

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[2] B. Gao, T. Chen, D. Tam, C.-L. Huang, K. Sasmal, D. Adroja, F. Ye, H. Cao, G. Sala, M. Stone, C. Baines, J. Barker, H. Hu, J.-H. Chung, X. Xu, S.-W. Cheong, M. Nallaiyan, S. Spagna, M. Maple, and P. Dai, *Experimental Signatures of a Three-dimensional Quantum Spin Liquid in Effective Spin-1/2* $Ce_2Zr_2O_7$ *Pyrochlore*, Nat. Phys. 15, 1052–1057 (2019).

[3] O. Benton, O. Sikora, and N. Shannon, *Seeing the Light: Experimental Signatures of Emergent Electromagnetism in a Quantum Spin Ice*, Phys. Rev. B 86, 075154 (2012).