Experimental report

29/06/2022

Proposal:	**4-05-813**	**Council:**	10/2020
Title:	Exploration of Excitations in Cerium Hafnate Pyrochlore Oxide.		
Research area:	Physics		

This proposal is a new proposal

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Samples: $Ce_2Hf_2O_7$

Instrument	Requested days	Allocated days	From	To
IN5	8	5	30/06/2021	06/07/2021

Abstract:

A theoretically predicted Octupolar QSI was found in the $Ce_2Sn_2O_7$ pyrochlore oxide. The dipole-octupole nature of the ground state doublet explains remarkably well macroscopic measurements. Furthermore, the octupolar nature of the correlations has been observed, taking the shape of a broad hump located at high momentum transfer in neutron diffraction patterns. Unfortunately, the chemistry of $Ce_2Sn_2O_7$ makes it challenging to grow single crystals to further elucidate the nature of its ground state. However, it is possible for its parent compound, $Ce_2Hf_2O_7$. Its chemical and physical similarities with $Ce_2Sn_2O_7$ motivate its investigation as it could withstand undoubtable fingerprints of this Octupolar QSI. We propose to explore the low-lying continuum of excitations present in $Ce_2Hf_2O_7$ and to track their evolution when entering the correlated regime as well as under the application of a magnetic field.

Experiment # 4-05-813: Exploration of Excitations in Cerium Hafnate Pyrochlore Oxide.

A large powder sample of $Ce_2Hf_2O_7$ (~30g), placed in a copper can, was mounted on a dilution fridge. The can was filled with few bars of He in order to have as efficient cooling as possible without bringing additional difficulties to the experiment. Once base temperature of the dilution fridge reached, additional time was needed so as to thermalize the sample. This step took a few hours due to the powder nature of the sample. The actual temperature of the sample was estimated base on fits performed on the inelastic spectrum of the sample using various models (single Lorentzian, double Lorentzian, damped harmonic oscillator). Meanwhile, an incident wavelength of 10 Angstrom was chosen to carry out the main part of experiment. A few additional measurements were carried out using 8 Angstrom incident wavelength. After complete stabilization of the inelastic signal, the sample's temperature was found to be about 100 mK.

After measuring at this base temperature, the remaining time was divided into six segments including measurements at five temperatures (200 mK, 400 mK, 800 mK and 5K) as well as an empty copper can. This was performed in such a way that different data sets will have similar statistics, allowing for a meaningful data analysis involving subtraction of data sets.

After careful study of the raw data, selected data sets of each temperature were carefully reduced through a Mantid routine. Figure 1.a) and 2.a) show the evolution of the inelastic signal upon temperature change, which, as expected for this compound, takes the shape of a continuum of excitations. Data sets were then subtracted to each other, taking into account the impact of the Bose factor. Resulting signals in the positive energy channel were subsequently fitted using a Lorentzian, as can be seen in Figure 1.b) and 2.b), providing useful information such as the energy gap and the "width" of the signal. Constant energy cuts in the 10 Angstrom data in the elastic line and in the excitation are displayed in Figure 1.c) and 1.d), with the former spoting an interesting Q-dependence, reminiscent of the magnetic diffuse signal in dipolar spin ices. Finally, Figure 1.e) shows a Q-E map of the spectrum obtained at the lowest temperature and with an incident wavelength of 10 Angstrom. The 5 K data were used to subtract the paramagnetic background.

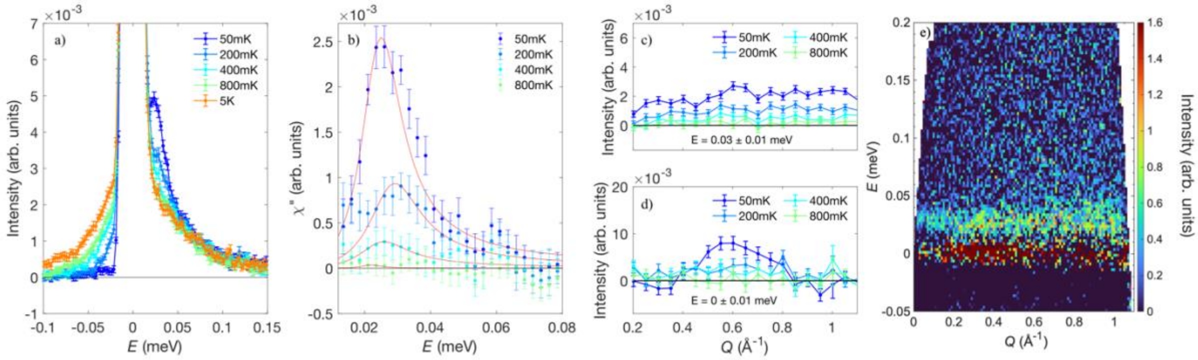

Figure 1: a) Background subtracted spectra collected using the incident wavelength of 10 Å at various temperatures. b) The imaginary part of the dynamics susceptibility extracted from the spectra displayed in a) by subtraction of the 5 K data (considered as high temperature background). The red lines represent the Lorentzian fits to the data, allowing to estimate the excitation's position, width and intensity evolution with respect to temperature. c) Q-dependence of the magnetic diffuse intensity in the elastic line. d) Q-dependence of the broad continuum of excitation centered around 0.03 meV. e) Difference map obtained by subtracting the 5 K data from the 100 mK data, showing a broad continuum of excitation at about 0.03 meV, as well as a Q-dependent signal at zero energy transfer.

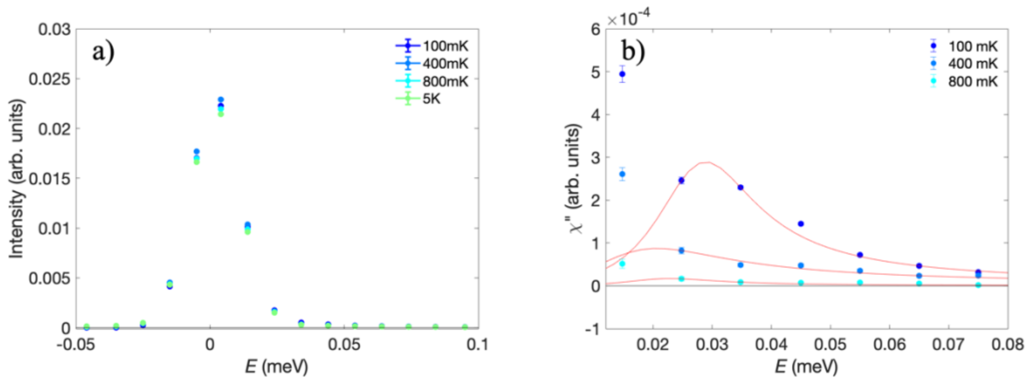

Figure 2: a) Background subtracted spectra collected using the incident wavelength of 8 Å at various temperatures. b) The imaginary part of the dynamics susceptibility extracted from the spectra displayed in a) by subtraction of the 5 K data (considered as high temperature background). The red lines represent the Lorentzian fits to the data, allowing to estimate the excitation's position, width and intensity evolution with respect to temperature.