# Experimental report

15/02/2018

**Proposal:** 4-03-1716 **Council:** 10/2014

**Title:** Crystal electronic field excitations of pyrochlore antiferromagnet Ce2Sn2O7

**Research area:** Physics

**This proposal is a new proposal**


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**Samples:** La2Sn2O7
Ce2Sn2O7 powder
Pr2Hf2O7
La2Hf2O7

| Instrument | Requested days | Allocated days | From | To |
|---|---|---|---|---|
| IN4 | 3 | 4 | 02/07/2015 | 06/07/2015 |


**Abstract:**

We have recently discovered an exotic magnetic state developing at subKelvin temperatures in the Ce3+ pyrochlore stannate. The spin system of this compound is characterized by a thermally isolated Jz=1/2 Kramers' doublet at low temperature, providing effective spin 1/2 moments on a pyrochlore lattice. We have observed that these spins are characterized by a strong Ising <111> anisotropy. The system does not order down to the lowest investigated temperature (70 mK). However, it develops a correlated regime for temperatures below 1 K, due to antiferromagnetic interactions one order of magnitude larger than the expected ferromagnetic dipolar couplings. We propose to explicitly measure the CEF scheme of Ce2Sn2O7 by inelastic neutron spectroscopy in order to confirm the picture of a thermally isolated Jz=1/2 Kramers' doublet at low temperature.

**Experimental report for experiment 4-03-1716 on IN4**

**"Crystal electronic field excitations of pyrochlore antiferromagnet $Ce_2Sn_2O_7$"**

- First part of the experiment: crystal-electric field of $Ce^{3+}$ in $Ce_2Sn_2O_7$

The aim of this experiment was to measure the crystal-electric field excitations of the quantum spin liquid candidate on $Ce_2Sn_2O_7$ IN4, where we can measure energy transfers up to 100-150 meV. This range of energy transfers covered the first, and eventually the second, crystal-field level positions that were estimated from our fit of the magnetic susceptibility prior to the experiment (c.f. proposal).

Complementary measurements were conducted using MAPS at ISIS in order to also access the intermultiplet transitions, which can play an important role in the composition of the ground doublet wavefunction – the final goal of our experiment being the determine it precisely. These measurements proved to cover the entire range of energy transfers that we needed, i.e. all crystal-field levels could be measured (see figure below). Nonetheless, we have used IN4 in order to check the absence of crystal-field level at the energy transfers that fall too close to the elastic line of our MAPS measurements (second figure below).

Figure 1: INS measurements from MAPS at ISIS, covering all CEF transitions in $Ce_2Sn_2O_7$.

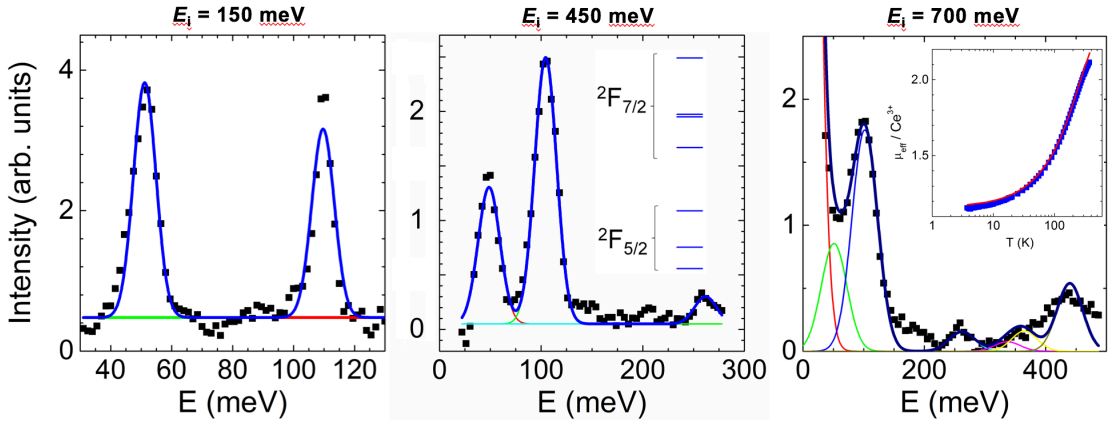
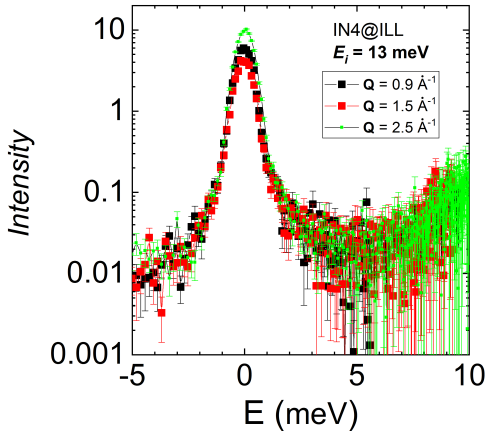



Figure 2: INS measurements (IN4), showing the absence of CEF level below that at 50 meV.




Taken together, the data from IN4 and MAPS allowed us to determine the wavefunction with a good level of confidence, and a paper is in preparation.

- Second part of the experiment: crystal-electric field of $Pr^{3+}$ in $Pr_2Hf_2O_7$

Given the short time needed for the measurements on $Ce_2Sn_2O_7$, we have consulted the college 4 secretaries about the possibility to measure a related compound during our beamtime on IN4. Our demand was positively received and therefore we have additionally measured samples of the candidate quantum spin ice $Pr_2Hf_2O_7$, and $La_2Hf_2O_7$ as a non-magnetic blank.

The results are described in R. Sibille et al. Phys. Rev. B. **94**, 024436 (2016).

The measurements on IN4 have allowed us to determine the CEF scheme of this interesting material. We could fit the data using a crystal field Hamiltonian and deduce the wavefunction of the ground-state doublet. Importantly, this measurement has demonstrated the admixture of quadrupolar components in the ground state wavefunction, which is an important ingredient for promoting quantum fluctuations in spin ice materials.

Figure 2: INS measurements (IN4) on $Pr_2Hf_2O_7$ (subtracted from $La_2Hf_2O_7$).