Experimental report

31/03/2017

Proposal: 4-05-640 **Council:** 4/2016

Title: Spin dynamics in Pr2Zr2O7: interplay of quadrupolar and dipolar degrees of freedom in spin ice

Research area: Physics

This proposal is a resubmission of 4-01-1448

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Samples: Pr2Zr2O7

Instrument	Requested days	Allocated days	From	To
IN5	7	6	08/12/2016	14/12/2016

Abstract:

The proposed experiment is part of our systematic experimental studies of geometrical frustration in various pyrochlore R2M2O7 (R = Rare earth, M an element like Ti, Sn, Zr). Here, we propose to study the spin excitations in Pr2Zr2O7 for H//111 at IN5. In combination with our previous measurements, this new dataset would provide a complete study of non-Kramers rare earth (like Pr) based pyrochlore which are likely governed by dominant effective coupling between quadrupoles on top of weaker interactions between dipoles. This experiment would stand as a test of our scenario.

Experimental report on Exp. 4-05-640

Rare earth pyrochlore magnets $R_2T_2O_7$ are model systems in the field of geometrical frustration. The case of non Kramers R-ions has especially attracted much attention, as it might realize a “quantum spin ice” with strongly correlated moments still fluctuating at 50 mK [1,2,3]. In contrast with Kramers ions, their ground state crystal field doublet is not protected from perturbations, especially from distortions which couple to quadrupoles and lift the degeneracy of those doublets. In this context, $Pr_2Zr_2O_7$ is precious (see [4]). Indeed, like Pr3+ is a non Kramers ion, and its magnetic anisotropy is found to be Ising-like (along the <111> axes). Despite a negative Curie Weiss temperature, reflecting antiferromagnetic interactions, it does not order down to 50 mK. Xac measurements show evidence for a freezing at very low temperatures.

Our neutron experiments at IN5 have provided some new insight into this issue, and shown that effective interactions between quadrupoles on the top of exchange interactions are at play [4,5]. They have been published in a recent PRB paper (https://doi.org/10.1103/PhysRevB.94.165153).

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Antiferroquadrupolar correlations in the quantum spin ice candidate $Pr_2Zr_2O_7$

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We present an experimental study of the quantum spin ice candidate pyrochlore compound $Pr_2Zr_2O_7$ by means of magnetization measurements, specific heat, and neutron scattering up to 12 T and down to 60 mK. When the field is applied along the [111] and [1$\bar{1}$0] directions, $\mathbf{k} = 0$ field-induced structures settle in. We find that the ordered moment rises slowly, even at very low temperature, in agreement with macroscopic magnetization. Interestingly, for $H \parallel [1\bar{1}0]$, the ordered moment appears on the so-called α chains only. The spin excitation spectrum is essentially *inelastic* and consists in a broad flat mode centered at about 0.4 meV with a magnetic structure factor which resembles the spin ice pattern. For $H \parallel [1\bar{1}0]$ (at least up to 2.5 T), we find that a well-defined mode forms from this broad response, whose energy increases with H, in the same way as the temperature of the specific-heat anomaly. We finally discuss these results in the light of mean field calculations and propose an interpretation where quadrupolar interactions play a major role, overcoming the magnetic exchange. In this picture, the spin ice pattern appears shifted up to finite energy because of those interactions. We then propose a range of acceptable parameters for $Pr_2Zr_2O_7$ that allow to reproduce several experimental features observed under field. With these parameters, the actual ground state of this material would be an antiferroquadrupolar liquid with spin-ice-like excitations.

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