

Experimental report

19/03/2019

Proposal: 4-05-715

Council: 4/2018

Title: Nd₂Zr₂-xTi_xO₇: a new quantum spin ice candidate

Research area: Physics

This proposal is a new proposal

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Samples: Nd₂Zr₂O₇

Instrument	Requested days	Allocated days	From	To
IN5	7	7	19/10/2018	26/10/2018

Abstract:

According to recent INS measurements carried out at IN5, the pyrochlore magnet Nd₂Zr₂O₇ appears to be close to a U(1) liquid phase in parameter space. This phase is the long sought quantum spin ice phase in the context of spin ice physics. By replacing part of the Zr by Ti, we seek to slightly tune the effective coupling constants and move the system to this U(1) phase. This is the aim of this proposal to study the spin excitation spectra in a 20 % doped single crystal.

Experimental report on 4-05-715

$\text{Nd}_2\text{Zr}_{2-x}\text{Ti}_x\text{O}_7$: a new quantum spin ice candidate

In this proposal we asked for beam time on the IN5 time-of-flight spectrometer to study mechanism behind the magnetic moment fragmentation [1]. This exotic phenomenon manifests as a dual ground state consisting of a fluctuating spin liquid fragment, a so-called Coulomb phase, on top of a magnetic monopole crystal. Using a combination of elastic and inelastic neutron scattering measurements performed on D23, D7 and IN5, we proposed that such a phenomenon likely occurs in $\text{Nd}_2\text{Zr}_2\text{O}_7$ [2,3,4]. We indeed observed the spectacular coexistence of an antiferromagnetic “All-in All-out” order ($T_N=285$ mK) induced by the monopole crystallization and of a pinch point pattern typical of a Coulomb phase (see figure 1). Furthermore, we could show that the spin ice pattern is not elastic as in classical spin ice, but an inelastic feature, which suggests that quantum effects are at play in this material. Dispersing features akin to spin waves are also observed above the dynamic spin ice mode (figure 2).

Interestingly, the whole spectrum is well reproduced in the framework of the XYZ Hamiltonian [3,5]. In this context, the softening of the spin ice flat band occurs at a critical ratio between effective exchange parameters of the model $J_x/|J_z|\approx 3$. Beyond this value, a $U(1)$ spin-liquid phase is stabilized, the celebrated “quantum spin ice” which has been long sought among pyrochlores magnets. To test this prediction, we have initiated the study of doped compounds, where a small amount of the Zr is replaced by Ti. This doping is expected to slightly change the exchange paths (and the crystal field environment of the Nd ion).

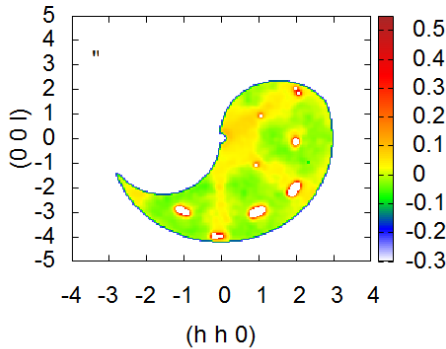


Fig 1: Diffuse scattering map of the spin-flip component (with Pz polarization along (1-10) measured at 60 mK on D7, obtained after subtraction of the 10 K data. Bragg peaks at (220) and (113) are characteristic of the antiferromagnetic “all in - all out” ordering.

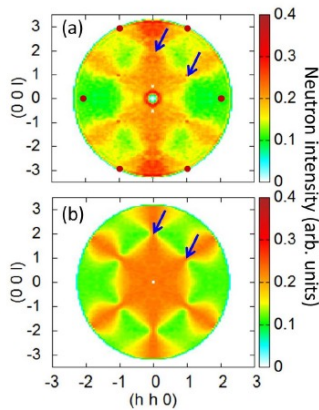


Fig 2: INS maps taken at 0.05 meV (top), along with the corresponding RPA calculations (bottom). Blue arrows indicate the pinch point position. Red spots denote the all-in all-out Bragg peaks (from D23 and [3]).

Our INS experiment has been carried out on IN5 in a standard set-up involving a cryo-magnet and a dilution insert. The measurements were performed with a wavelength of 6 Å and 8.5 Å to ensure a good energy resolution required to resolve the small energy gap to the spin ice mode (about 70 μeV). We varied the temperature from 45 mK up to 1 K.

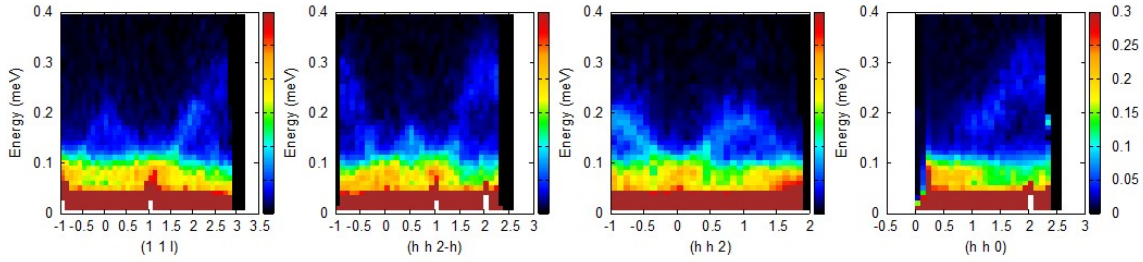


Fig 3: Magnetic excitations spectra in the 5% doped sample ($T_N=375$ mK) measured at 45 mK.

The spin ice dynamics has been fully determined at low temperature (figure 3), yielding slightly different effective parameters of the XYZ Hamiltonian compared to the pure sample. Further analyses are on-going to understand these results as well as their impact on the fragmentation mechanism.

- [1] Brooks-Bartlett et al., Phys. Rev. X 4, 011007 (2014).
- [2] Elsa Lhotel et al, Phys. Rev. Lett. 115, 197202 (2015).
- [3] Sylvain Petit et al., Nature Phys. 12, 746 (2016).
- [4] Elsa Lhotel et al, Nature Communications volume 9, Article number: 3786 (2018)
- [5] Owen Benton, Phys. Rev. B 94, 104430 (2016).