Experimental report

27/03/2020

Proposal: 4-05-748 **Council:** 4/2019

Title: Nd2Zr2-xTixO7: spin dynamics in a new quantum spin ice candidate. Influence of the magnetic field along 001

Research area: Physics

This proposal is a new proposal

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Samples: Nd(ZrTi)2O7

Instrument	Requested days	Allocated days	From	To
IN3	2	2	22/07/2019	24/07/2019
IN5	7	6	29/08/2019	04/09/2019

Abstract:

This proposal aims at studying in details the spin dynamics associated with a field induced transition observed in the doped quantum spin ice candidate Nd2Zr2O7. We plan to apply the field along [001] where the metamagnetic transition is markedly influenced by the doping (according to low temperature magnetization measurements). We especially would like to follow the dynamic spin ice mode accross the transition.

Experimental report on 4-05-748 at IN5
$Nd_2Zr_{2-x}Ti_xO_7$: Unexpected magnetic transition in a new quantum spin ice candidate

In this proposal, we asked for beam time at the IN5 time-of-flight spectrometer to investigate the physics of $Nd_2Zr_2O_7$. This pyrochlore material shows below T_N= 285 mK an all-in all-out magnetic structure characterized by a strong reduction of the ordered magnetic moment: only 0.8 µB out of 2.4 µB for the free Nd ion. Strikingly, the spectrum of the excited states consists in a flat band showing a structure factor typical of the diffuse scattering observed in classical spin ice, along with dispersing modes akin to spin waves [1,2,3].

Interestingly, these features can be understood in the framework of an XYZ Hamiltonian [2,3,4] written for a pseudo spin τ. The z component of this pseudo spin corresponds to the magnetic moment while the x and y components correspond to different octupolar moments. The apparent reduction of the ordered moment is then interpreted as a mixed octupolar and dipolar (all in all out) ordered phase: the pseudo spin is fully ordered but tilted away from the z axis, hence the apparent reduction of the dipolar ordered moment. Furthermore, the gap to the "spin ice" band characterizes the proximity of the spin ice phase. It is worth emphasizing that the softening of the spin ice flat band would signal a transition from the all in all out to a U(1) spin-liquid phase (the celebrated "quantum spin ice" which has been long sought among pyrochlores magnets [4]).

To further investigate this peculiar ordered state along with its unconventional excitations, we have started the study of doped compounds, where Ti replaces a small amount of the Zr. This should slightly change the exchange paths as well as the crystal field environment of the Nd ion. Our first results on the 2.5% sample are quite unexpected and extremely encouraging: instead of a weakening of the properties of the pure compound, inelastic scattering measurements performed on IN5 on the doped compound show an extremely clean and robust dynamic spin ice band.
Furthermore, magnetization measurements (Fig. 1) show that a marked field induced transition occurs when the field is applied along the [001] direction. At 95 mK, the transition is expected at about H_c=0.15 T. Concomitantly our magnetization and diffraction measurements (D23) have shown that the all in all out antiferromagnetic transition is reinforced with a higher transition temperature (T_N=400 mK) and a larger ordered magnetic moment.

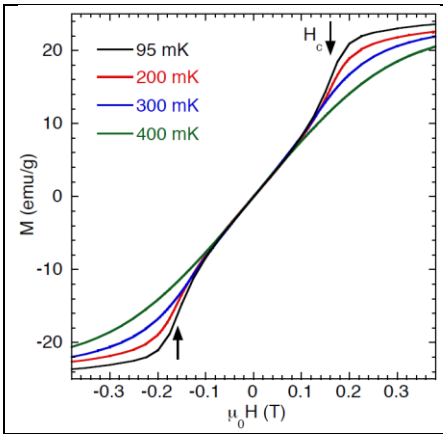

Fig 1: M(H) curves measured up to 400 mK in the 5% doped sample with H// [001]. The upturn between 0.1 and 0.2 T is the signature of the anomalous field induced transition.

The INS measurements at IN5 were performed with a wavelength of 6 Å for H=0 and for 8 and 10 Å (to ensure a good energy resolution) for H=0, 0.1, 0.2 and 0.3T // [001]. Figure 2 summarizes our main results. We find that the spectrum is very close to the one observed in the pure sample, indicating quite similar exchange parameters.

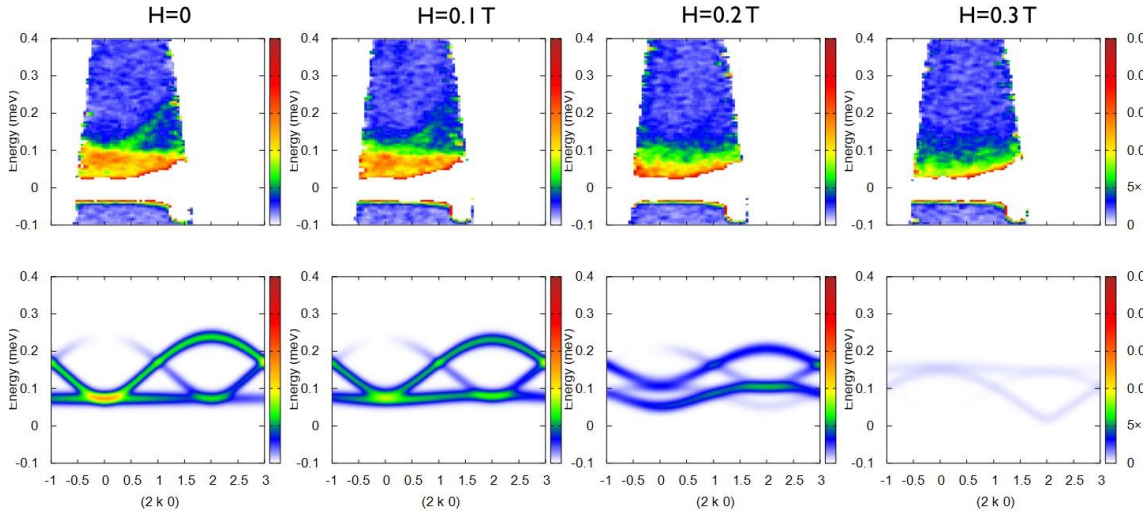

Figure 2: (Top) measurements performed at T=60 mK and H=0, 0.1, 0.2 and 0.3 T. IN5 was operated with $\lambda = 8$ Å. (Bottom) calculations carried out in the XYZ model with the exchange parameters of the pure $Nd_2Zr_2O_7$. The transition is expected at 0.25 T in those calculations.

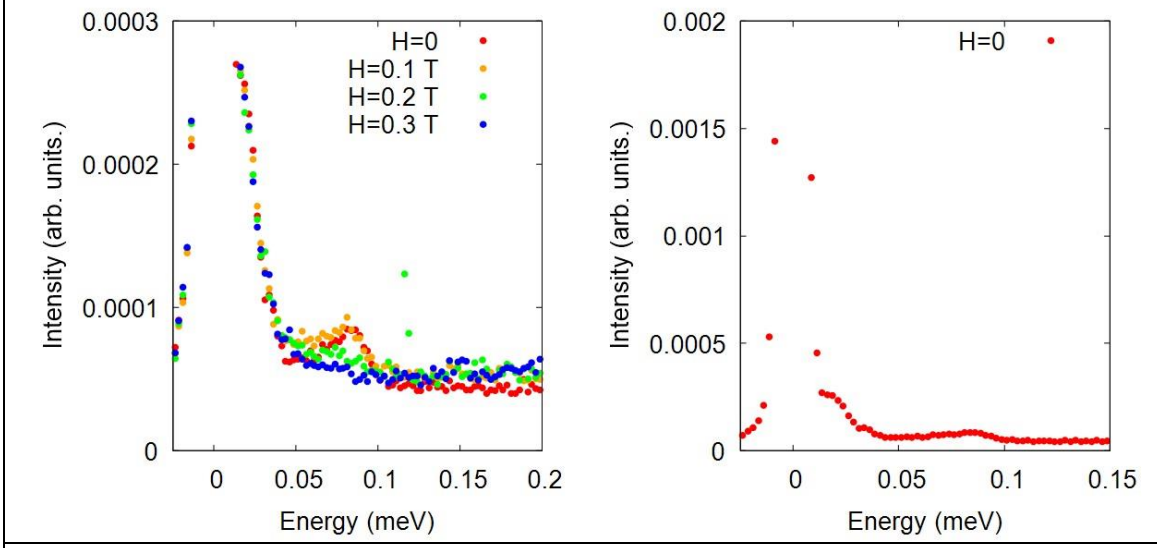

Figure 3: (Left) Softening of the flat band with increasing field. (Right): Total (Q integrated) signal at H=0 (same as left with a different scale). Note the bump at low energy close to the incoherent response.

We find that with increasing the field, the flat band seems to soften (see also Figure 3). Note that according to our D23 diffraction results, the high field is an ordered "two in two out" phase (this phase has a net magnetic moment along the field). This gives a clue to the interpretation of our results and may explain the nature of the transition: closing the gap to the spin ice band brings back to the ground state ordered "two in two out" configurations. The field selects, however, those which are compatible with the [001] direction.

The evolution of the spin ice band not only vs field but also vs temperature is likely a key information. According to a recent scenario [5], the AIAO phase might be stabilized out of a high temperature spin-ice like state. The prediction is that the spin ice flat band should detach from the zero energy line at T_N

and that its energy should increase with decreasing temperature. Such an observation would definitely shed light on the physics at play in this material.

It is also worth noting the bump at low energy close to the incoherent response. The origin of this feature remains unclear at this step. We speculate that it could be the signature of the splitting of the Nd nuclear multiplet by the hyperfine coupling. It could be also an “in-gap” response specific of the Ti-impurities. An experiment is planned at ILL to test this issue.

Finally, above the transition, no clear spin waves could be identified. We know from the calculations that the signal should be very weak, however, observation of dispersive features would help to determine accurately the exchange parameters. Further analysis is ongoing to propose a model able to predict the meta-magnetic at the true critical field.

References

[1] Elsa Lhotel et al, Phys. Rev. Lett. 115, 197202 (2015).
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[3] Elsa Lhotel et al, Nature Communications volume 9, Article number: 3786 (2018)
[4] Owen Benton, Phys. Rev. B 94, 104430 (2016).
[5] J Xu, O. Benton et al, arXiv:1907.11056v1 (2020)