Experimental report

04/04/2016

Proposal: 5-53-255 **Council:** 4/2015

Title: Diffuse scattering and possible pinch points in the Ising antiferromagnet Nd2Zr2O7

Research area: Physics

This proposal is a new proposal

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Samples: Nd2Zr2O7

Instrument	Requested days	Allocated days	From	To
D7	8	8	17/11/2015	25/11/2015

Abstract:

Recent combined magnetization and neutron diffraction experiments demonstrate that the Ising AF pyrochlore antiferromagnet Nd2Zr2O7 orders in an all-in all-out configuration. In principle, this corresponds to an un-frustrated situation, in contrast with other remarkable members of the family like Ho2Ti2O7 or Tb2Ti2O7. However, our findings point to the predominance of quantum fluctuations in this material and show that this case is a lot more complicated than it first appears. To shed light on this issue, we plan to study the diffuse scattering in Nd2Zr2O7 on the time of flight spectrometer D7 using polarized neutrons and equipped with the dilution insert.

Experimental report on Exp. 5-53-255

Diffuse scattering and possible pinch points in the Ising antiferromagnet $Nd_2Zr_2O_7$

Following conventional ideas [1,2,3], antiferromagnetic interactions between Ising spins on the pyrochlore lattice correspond to a "simple" canonical un-frustrated case. The energy is minimized for a single configuration where the 4 spins point towards or outwards the center of a tetrahedron, so called "all-in / all-out" structure. Recently, we have investigated single crystals of $Nd_2Zr_2O_7$ and conclude that this compound can be indeed considered as a model system of this case. The Nd^{3+} magnetic moments exhibit a strong local <111> Ising anisotropy with an effective value of about $2.4\mu_B$ [4]. Magnetization measurements performed along the [100], [110] and [111] axes of a crystal down to 70 mK, together with powder neutron diffraction experiments, reveal that $Nd_2Zr_2O_7$ undergoes an antiferromagnetic transition at 285 mK towards an all-in / all-out state [5].

Commentaire [ML1]: I think we should say what these directions are rather than just saying all the three main axes

Surprisingly, these experiments demonstrate that the magnetic moment is only partially ordered ($0.8\mu_B$), while the full moment is $2.4\mu_B$. These findings point to the predominance of quantum fluctuations and show that this case is a lot more complicated than it first appears: a large part of the moment is fluctuating. To address this issue and shed light on the enigmatic role of quantum fluctuations in these 3D systems, we propose to measure the diffuse scattering at different temperatures on D7, using polarized neutrons.

This experiment on $Nd_2Zr_2O_7$ was quite successful, as we have discovered in this particular material a novel fractionalization mechanism called "fragmentation". The results have been accepted for publication in Nature Physics (DOI **10.1038/nphys3710**):

Observation of magnetic fragmentation in spin ice

Authors: S. Petit, E. Lhotel, B. Canals, M. Ciomaga-Hatnean, J. Ollivier, H. Mutka, E. Ressouche, A.R. Wildes, M.R. Lees, G. Balakrishnan
(accepted in Nature Physics, see also http://arxiv.org/abs/1603.05008)

Abstract: Fractionalised excitations that emerge from a many body system have revealed rich physics and concepts, from composite fermions in two-dimensional electron systems, revealed through the fractional quantum Hall effect, to spinons in antiferromagnetic chains and, more recently, fractionalisation of Dirac electrons in graphene and magnetic monopoles in spin ice. Even more surprising is the fragmentation of the degrees of freedom themselves, leading to coexisting and *a priori* independent ground states. This puzzling phenomenon was recently put forward in the context of spin ice, in which the magnetic moment field can fragment, resulting in a dual ground state consisting of a fluctuating spin liquid, a so-called Coulomb phase, on top of a magnetic monopole crystal. Here we show, by means of neutron scattering measurements, that such fragmentation occurs in the spin ice candidate $Nd_2Zr_2O_7$. We observe the spectacular coexistence of an antiferromagnetic order induced by the monopole crystallisation and a fluctuating state with ferromagnetic correlations. Experimentally, this fragmentation manifests itself via the superposition of magnetic Bragg peaks, characteristic of the ordered phase, and a pinch point pattern, characteristic of the Coulomb phase. These results highlight the relevance of the fragmentation concept to describe the physics of systems that are simultaneously ordered and fluctuating.

The maps performed on D7 allow to observe both the spin-ice pattern along with the "all-in / all-out" Bragg peaks, especially (220) and (113) (Figure 1). Figure 2 shows the magnetic neutron intensity integrated over a small region around (220) as well as over a sector delineating the arm along (hhh)

(i.e. the direction of the (111) pinch point), between 35 and 55 degrees. These data show that the Bragg intensity disappears at T_N, between 300 and 450 mK, while the spin-ice pattern persists above T_N, up to 600 mK and becomes barely observable only above 750 mK. Interestingly, the intensity of the arms seems to be maximum around 300 mK. Further analysis is in progress to confirm this effect, and understand it.

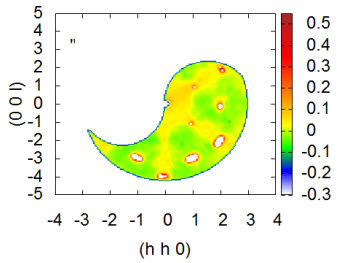

Figure 1: Diffraction data obtained at 50 mK

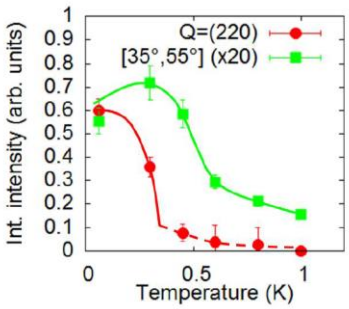

Figure2: Red dots: Averaged neutron intensity around the (220) Bragg peak as a function of temperature. Green squares: Averaged neutron intensity in a sector delineating the arm along (hhh) of the spin-ice pattern. This sector is defined in the range 35 to 55 degrees.

[1] J.S. Gardner et al., Rev. Mod. Phys. **82**, 53 (2010).
[2] Introduction to Frustrated Magnetism, edited by C. Lacroix, P. Mendels, and F. Mila (Springer-Verlag, Berlin, 2011.
[3] S. T. Bramwell and M. J. P. Gingras, Science 294, 1495 (2000).
[4] M. Ciomaga Hatnean et al., submitted to Phys. Rev. B
[5] E. Lhotel et al, Phys. Rev. Lett. 115, 197202 (2015).