Experimental Report

Proposal: **5-32-796** **Council:** 4/2014

Title: Short-range magnetic correlations in the "large-D" 1D system Sr3NitO6

This proposal is a new proposal

Researh Area: Physics

Main proposer: **LEFRANCOIS Emilie**

Experimental Team: CHAPON Laurent
SIMONET Virginie
LEFRANCOIS Emilie
BALLOU Rafik
LHOTEL Elsa

Local Contact: NILSEN Goran

Samples: Sr3NiPtO6

Instrument	Req. Days	All. Days	From	To
D7	4	4	13/10/2014	18/10/2014

Abstract:

We propose to study the magnetic short-range correlations in the frustrated Sr3NiPtO6 compound, theoretically predicted to lie in a regime where quantum fluctuations enhanced by a large D/J ratio, suppress long-range magnetic ordering. Determining the exact nature of the magnetic ground state is key to probe the supposedly large-D phase predicted in this easy plane one-dimensional system.

Oxides of general formula $A_3MM'O_6$ (M and M' are transition metal ions) have attracted considerable interest in the last two decades, because their peculiar crystal structure leads to unconventional magnetic properties. A famous prototype is the strongly frustrated Ising magnet $Ca_3Co_2O_6$ (M=M'=Co), which displays plateaus in the magnetization [1] and lies close to a magnetic instability, best evidenced by the slow percolation of a metastable magnetic phase under certain conditions [2]. The crystal structure of $A_3MM'O_6$ consists of chains of alternating face-sharing octahedra (MO_6) and trigonal prisms ($M'O_6$) linked by their faces and arranged in an hexagonal motif. In most compounds, where M, M' or both are magnetic, a strong magnetic frustration arises as a consequence of the competition between first-neighbour magnetic interactions (J1) within chains and antiferromagnetic interactions between adjacent chains (J2 and J3) mediated by super-super-exchange interactions [3].

We are interested in the compound Sr_3NiPtO_6, where the only magnetic species are the $M=Ni^{2+}$ (S=1) ions of the NiO_6 octahedra. Its magnetic properties were recently revisited [4]. A strong easy plane magnetocrystalline anisotropy was evidenced from susceptibility measurements on a single crystal, in strong contrast to the easy-axis magnetism found in the prototype $Ca_3Co_2O_6$. An helical magnetic structure whose pitch is related to the exchange ratios J1/(J2, J3) was classically awaited. No signature of a long-range order however was found down to 1.5 K, neither from the magnetic susceptibility nor from the specific heat, leading to propose that Sr_3NiPtO_6 is an experimental realization of the so-called "large-D" phase. We were able to prepare single crystals of small size, on which we confirmed the existence of the large easy-plane magnetocristalline anisotropy by magnetization measurements. No sign of a magnetic order was detected down to 0.05 K. The large-D phase should be awaited in easy-plane 1D magnet where quantum fluctuations destroy magnetic order but also for which the ratio D/J1 of the magnetocrystalline anisotropy D over the intra-chain exchange J1 is large enough to destabilize the Haldane phase [5]. The large-D phase is in principle characterized by a gap in the excitation spectrum like the Haldane phase, but, in contrast, should not show antiferromagnetic short-range correlations. This picture may hold to a certain extent, but the system is far from an ideal configuration of isolated chains, which prompted us to study the low-temperature magnetic state in details.

The diffuse neutron scattering experiment with tri-axial polarization analysis on the D7 spectrometer was performed on a poly-crystalline sample. The scattering data were collected at different temperatures from 0.05 K to 300 K. After calibration they were corrected from the empty can contribution, measured beforehand, and renormalized with respect to previously recorded vanadium scattering intensities. Unambiguous separation of the magnetic, nuclear and spin incoherent scattering was achieved by the tri-axial polarization analysis [5].

No magnetic signal is detected at the lowest temperature whereas a paramagnetic diffuse scattering gradually emerges as the temperature is increased (see figure 1). The magnetic peak observed at 1.3 Å comes from NiO impurity. We thus confirmed that the S=1 quantum spin chains of alternating face-sharing octahedra (NiO_6) in Sr_3NiPtO_6 are indeed in the large-D phase at the lowest temperature. Of interest would be to probe by inelastic experiment the associated excitation spectrum, in particular to determine the excitation gap, to access to the exciton-antiexciton continuum and possible bound states [6] **and to examine whether soft modes inherent to interchain interactions occur [7]. A surprise was to find out that the diffuse nuclear scattering substantially increases also as the temperature is increased up to 100 K (see figure 2). Then this signal decreases as the temperature is further increased up to 300 K. We do not understand the origin of this mysterious nuclear response. An inelastic neutron scattering experiment should help to get some insights.**

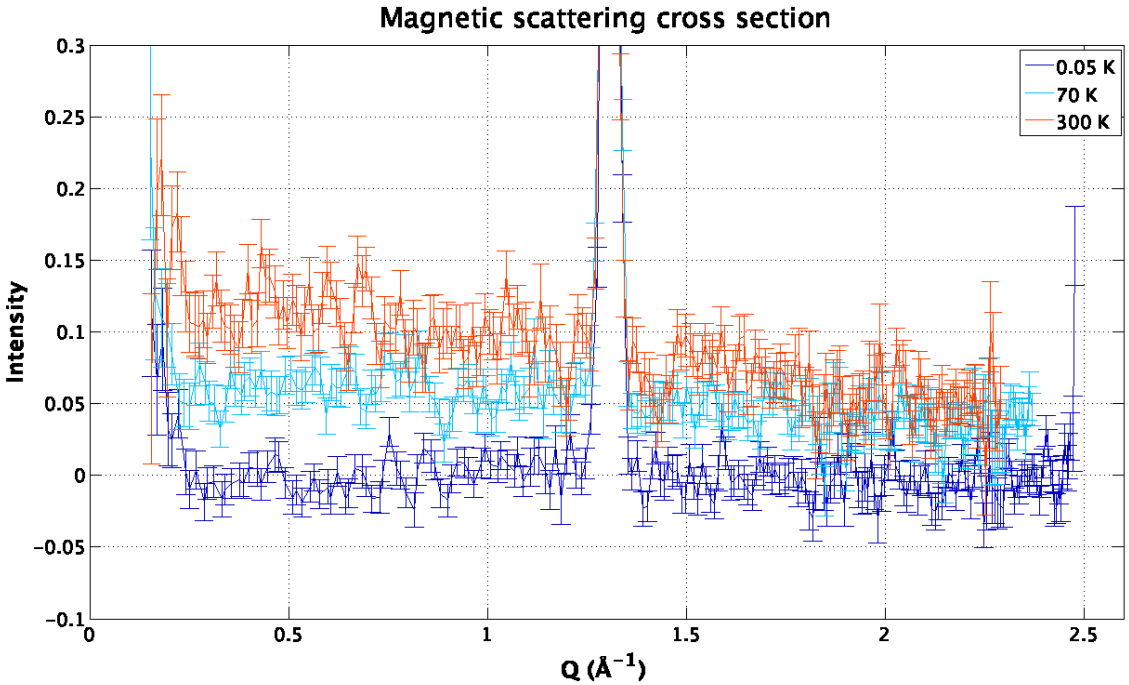

Figure 1: Magnetic scattering extracted from diffuse neutron scattering with tri-axial polarization analysis on Sr_3NiPtO_6 polycrystals at different temperatures from 0.05 K to 5 K.

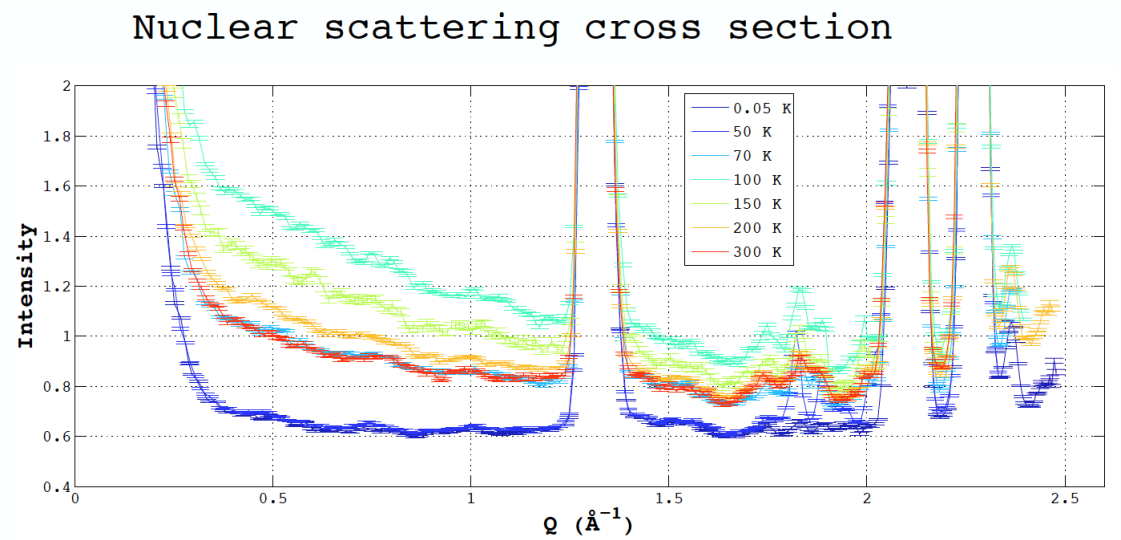

Figure 2: Nuclear scattering extracted from diffuse neutron scattering with tri-axial polarization analysis on Sr_3NiPtO_6 polycrystals at different temperatures from 0.05 K to 5 K

References

[1] A. Maignan, et al., Eur. Phys. J. B 15, 657 (2000)
[2] S. Agrestini et al., Phys. Rev. Lett. 106, 197204 (2011)
[3] L. C. Chapon, PRB, 80, 172205 (2009)
[4] S. Chattopadhyay, PRB, 82, 094431 (2010)
[5] J. R. Stewart, et al., J. Appl. Cryst. 42, 69–84 (2009)
[6] N Papanicolaout and P Spathis, J. Phys.: Condens. Matter 1**,** 5555 (1989)
[7] P.-A. Lindgard, Physica 120B, 190 (1983)