Experimental report

21/02/2020

Proposal: **5-31-2538** **Council:** 4/2017

Title: Magnetic spiral in ZnCr2Se4 under pressure up to 10GPa and low T.

Research area: Physics

This proposal is a new proposal

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Local contacts: Thomas HANSEN

Samples: spinel ZnCr2Se4

Instrument	Requested days	Allocated days	From	To
D20	4	4	06/04/2018	10/04/2018

Abstract:

The ZnCr2Se4 compound with the spinel structure is of interest as one of objects with magnetoelectric effect [1], in which at 21K occurs a weak tetragonal distortion [2] and below this temperature the DM interaction produces a simple magnetic spiral structure of the SS type (period is 22,5A) [3], whose period decreases synchronously with the increase of the spontaneous strain [4]. To determine the influence of high pressure on the magnetic spiral, we would like to perform a neutron diffraction study at T = 1.5-50K and pressures P = 0-120kbar using high pressure cell of Paris-Edinburg type. The lattice parameter is close to the value of 10A that period of magnetic spiral at a pressure 75kbar virtually coincides with it, and the magnetic structure becomes commensurate.

[1] H. Murakawa, Y. Onose, K. Ohgushi, S. Ishiwata, and Y. Tokura, J. Phys. Soc. Jap. 77, 043709 (2008)
[2]. R. Kleinberger and R. de Kouchkovsky. CR Acad. Sc.Paris.t.262, Ser.B (1966).
[3]. R. J. Plumier, J. Appl. Phys.V.37, No.3, p.964 (1966).
[4]. Jun Akimitsu at all. J. Phys. Soc. Japan, V.44, No.1, p.172, (1978).

Report on the experiment 5-31-2538: Magnetic spiral in $ZnCr_2Se_4$ under high pressure and low T.

Experimental team: R. Sadykov, I. Mirebeau. Local contact: T. Hansen. Data treatment: N. Martin and M. Deutsch.

Experimental goal:

Our previous (unpublished) measurements on ZnCr2Se4 were performed at PSI (DCS, up to 2.8 Gpa) and LLB (G61, up to 5.6 GPa). They showed that the helical structure is sensitive to pressure: T_N increases from 20K at P=1 bar up to 78K at P= 5.6 GPa, whereas the helical pitch decreases with increasing pressure. These results suggested a transition from the helical to a commensurate collinear phase could occur around 7 GPa. The experiment on D20 was performed up to 8.8 GPa to check this scenario.

Experiment:

A $ZnCr_2Se_4$ sample was loaded in the Paris-Edimburgh pressure cell, with deuterated ethanol-methanol as transmitting medium, and inserted in cryogenerator. The experiment was performed in 5 steps:

1) Increase of pressure at 300K up to P= 6.1 GPa (He, 1040 bars)
2) P=6.1 GPa : measure in the T range 300K- 8K
3) At 303 K increase pressure up to 7.6 GPa (He, 1250 bars) : measure in T range 303K-10K
4) At 300 K, increase pressure up to 8.8 GPa (He, 1550 bars) : measure in T range 302K-78K
5) At 295K, decrease pressure down to P=1.8 GPa (He, 85 bars) : measure at 295K and 78K

Results

The experiment on D20 confirmed the results obtained at lower pressures, but it also yielded unexpected results which we summarize by showing the pressure and temperature dependence of the zero order helimagnetic satellite (Fig. 1). Unexpectedly, we observe that its intensity splits into two contributions I_1 and I_2. The first intensity I_1 disappears around 6.5 GPa at T_N= 80K as expected, but the second one I_2 persists up to the highest pressure with a very high transition temperature, above 300K. This second contribution is clearly observed on D20, thanks to high intensity and extended temperature and pressure range.

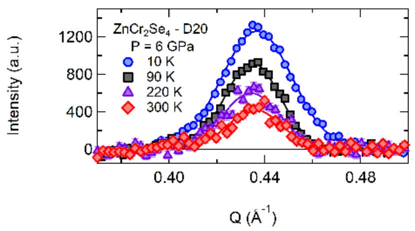

Fig1a: T-dependence of the zero satellite measured on D20 at 6 GPa

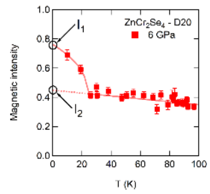

Fig 1b: Integrated intensity of the 00+ satellite, scaled to the intensity of the 400 Brag peak versus temperature at 6. Gpa.

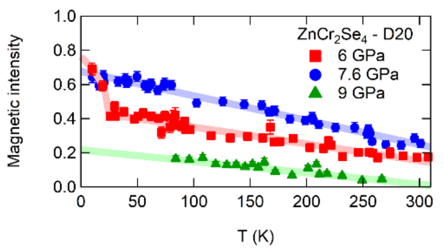

Fig 1c: T-dependence of the Intensity of the 00+ satellite measured on D20 at several pressures, scaled to the intensity of the 400 Bragg peak.

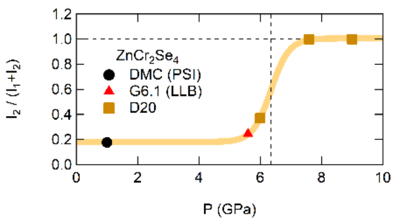

Fig 1d: relative spectral contribution of the high pressure signal I_2 to the total magnetic signal I_1+I_2

Moreover, we do not see any onset of commensurate order, the position of the zero satellite (yielding the magnetic period) becoming independent of pressure and temperature in the high pressure range, with a value of about 1.41 times the lattice constant (Figure 2)

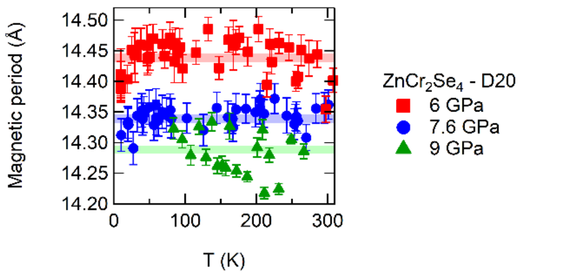

Fig. 2a: Temperature dependence of the magnetic period versus temperature

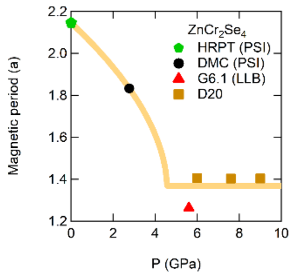

Fig. 2b: pressure dependence of the magnetic period at low temperature.

Interpretation

$ZnCr_2X_4$ spinels (X=O, S, Se) are known to be highly frustrated materials, with a frustration ratio $\theta cw/T_N$~5 for $ZnCr_2Se_4$ at ambient pressure, and quasi-static magnetic fluctuations highly coupled to the crystal lattice. The interaction scheme involves up to 5 near-neighbor interactions, where both direct and indirect super-exchange contribute. Due to this intricate situation, the ambient pressure ground state (moment value), the occurrence of several magneto-structural transitions, and the presence of diffuse scattering at ambient pressure, are sample dependent. It suggests that the magnetic frustration may be relieved either by spontaneous strains, induced by small non-stoechiometries or site inversion, or by impurities, pinning the lattice period to peculiar values.

Influence of the sample state at ambient pressure

To check this scenario, a precise investigation of the ambient pressure state is necessary for each sample synthesis. It is also crucial to perform Fullprof refinements of the high-pressure patterns measured on D20, considering the numerous phases involved (up to 5, including NaCl and Pb pressure calibrants). We are now completing this ambient pressure study in the two samples studied under pressure, by combining high-resolution PND patterns, magnetization and X-rays. The D20-sample shows the presence of an impurity (Figure 3), whereas another sample studied at PSI-LLB, where impurity phases are in negligible amount, reveals a phase separation between two spinel-phases with slightly different lattice constants and propagation vectors. So both mechanisms could be at play to prevent the onset of a commensurate order. To complete this comparative study and finalize the publication of our pressure results, we have asked for beam time at D1B.

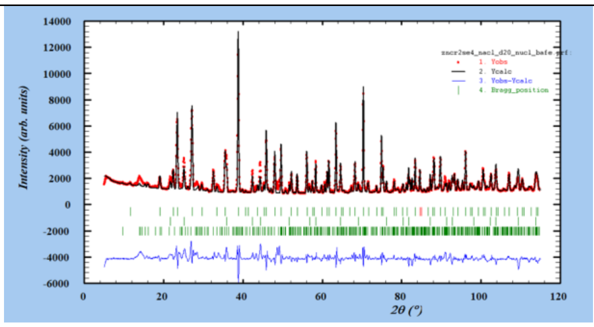

Fig: 3 : High resolution PND pattern obtained at 3T2-LLB. The $ZnCr_2Se_4$ sample belongs to the same synthesis as measured on D20 under pressure. An impurity phase isostructural to $(Ba_{1-x}Sr_x)_2Zn_2Fe_{12}O_{22}$ is included in the refinement.