Experimental report

19/02/2018

Proposal:	**5-31-2527**	**Council:**	10/2016
Title:	Pressure effects on the zigzag ladder antiferromagnet b-CaCr2O4		
Research area:	Physics		

This proposal is a resubmission of 5-31-2500

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Samples: b-CaCr2O4

Instrument	Requested days	Allocated days	From	To
D20	7	5	22/02/2017	27/02/2017

Abstract:

The zigzag antiferromagnet beta-CaCr2O4 exhibits an excitation spectrum characterized by an unusual pseudo-gapped signal above TN, thought to arise from the contribution of a two-spinon continuum. A distribution in the Cr-Cr distances can account for this pseudo-gap. As applying an external pressure on the system would allow to tune the Cr-Cr distances, we would like to study the effect of high pressure on the crystallographic and magnetic structure of b-CaCr2O4.

Pressure effects on the zigzag ladder antiferromagnet β-$CaCr_2O_4$
Report for experiment 5-31-2527

We addressed in this study the case of the chromium oxide β-$CaCr_2O_4$, which exhibits a tunnel-like structure (orthorhombic *Pbnm*, a = 10.6203(3)Å, b = 9.0801(3)Å, c = 2.9681(1)Å) akin to $CaFe_2O_4$. In this system, the magnetic Cr^{3+} ions (S = 3/2) are located on two different crystallographic sites. On one hand, the CrO_6 octahedra share edges to form triangular ladders running along *c* and on the other hand, they are connected by corners to form large cavities occupied by Ca^{2+} in prismatic coordination in the *(a,b)* plane.

β-$CaCr_2O_4$ exhibits long range incommensurate cycloidal magnetic order below T_N = 21 K ; above T_N, however, and up to ~ 100 K, the magnetic susceptibility is characteristic of a one dimensional magnetic compound, and the inelastic neutron scattering spectrum shows a magnetic excitation, which is thought to arise from the contribution of a two-spinon continuum, in agreement with theoretical views on quantum frustrated chains within the $J_2 >> J_1$ limit. This magnetic excitation is actually characterized by a (pseudo) gapped signal centered around Q = 1.25 $Å^{-1}$.

Preliminary results with inelastic neutron scattering showed a significant reduction of the ordered magnetic moment under pressure along with an increase of the 1D excitations, for an applied pressure up to 1 GPa. We thus wanted to further investigate the effect of external pressure on the magnetic order and the crystallographic structure in β-$CaCr_2O_4$. In particular, the aim of this experiment was two-fold: we first wanted to follow the magnetic Bragg peak located at Q = 1.15 $Å^{-1}$ and in parallel study the evolution of Cr-Cr distances as a function of pressure. For this we recorded several diffractograms using λ= 1.54 Å for the structural part and λ= 2.41 Å for the magnetic part, for pressure values from 1.8 to 9.5 GPa at 300K, 80K, 20K, and 6K.

From a structural point of view, Rietveld refinements show that the Cr(2) ladders are reinforced under pressure to the detriment of the Cr(1) ladders, as Cr(2)-Cr(2) distance decrease faster than the Cr(1)-Cr(1) distance under pressure (see figure 1). The one-dimensional character of the Cr(2)-Cr(2) chain thus seems to be enhanced with pressure.

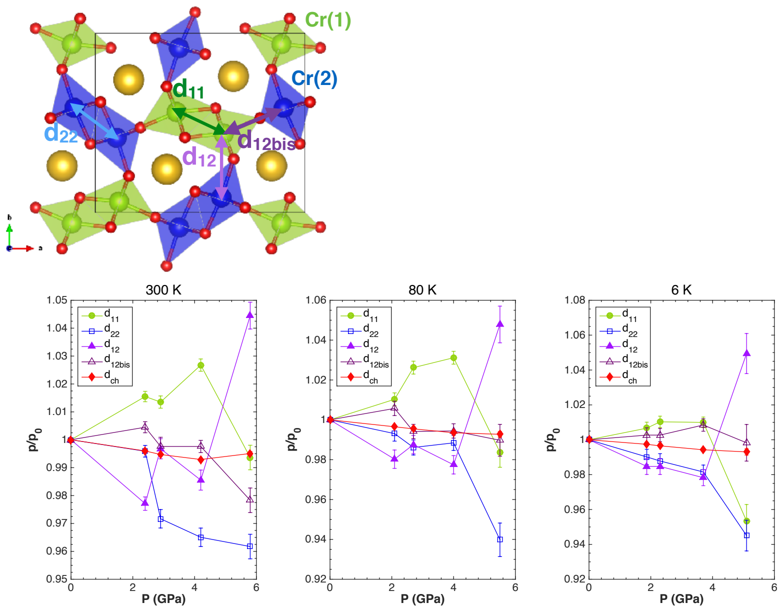

Fig.1 Evolution of the relative Cr-Cr distances as a function of pressure at T = 300K, 80K and 6K. P_0 is the Cr-Cr distance refined at ambiant pressure. The different nearest and next nearest-neighbour distances between chromium atoms are illustrated by the colored arrows.

For the magnetic part, some data was lost for a few pressure values and temperatures, due to a problem in programming the sequence. However, the data for P = 1.87 GPa and P = 3.7 GPa at T = 6K and 20K could be analysed and shows that the magnetic moment seems to disappear for an intermediate pressure value between 1.9 and 3.7 GPa. Indeed figure 2 shows that the magnetic Bragg peak located at Q = 1.15 $Å^{-1}$ is still clearly present at 1.87 GPa but seems to transform into a diffuse signal at 3.7 GPa. This diffuse scattering could come from a contribution of the inelastic part integrated in energy by the diffractometer.

This study hence shows a potential cancellation of the Néel temperature around 2 GPa, for which the system would fall into a one-dimensional regime. It would be necessary to confirm the effect of pressure on the magnetic excitations, by performing an inelastic study under applied pressure ranging from 1.5 to 4 GPa.

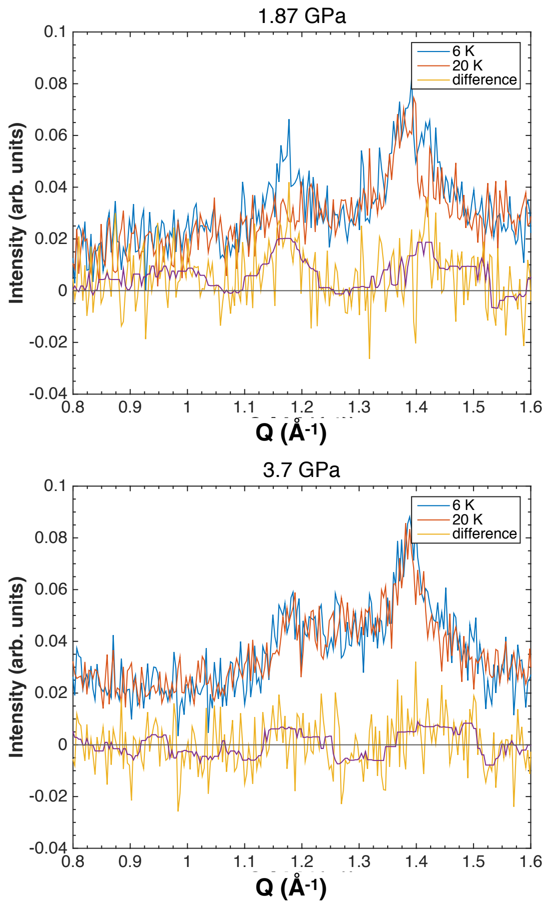

Fig.2 Magnetic Bragg peak located at Q = 1.15 Å^{-1} measured at 1.87 GPa and 3.7 GPa. The blue line represent the data at 6K, the orange line shows the data at 20 K (1D regime) and the yellow line show the difference between the two temperatures. The purple line is the averaged difference