Experimental report

16/09/2019

Proposal:	**5-54-274**	**Council:**	10/2018

Title: Pressure effect on spiral spin orientation in multiferroic pyroxene: SrMnGe2O6

Research area: Physics

This proposal is a new proposal

Main proposer: **Claire COLIN**

Experimental team: Claire COLIN
Noriki TERADA

Local contacts: Anne STUNAULT
Navid QURESHI

Samples: SrMnGe2O6

Instrument	Requested days	Allocated days	From	To
D3 CPA	7	0		
IN20	0	7	09/09/2019	16/09/2019

Abstract:

Using combination with CryoPAD and Hybrid Anvil pressure Cell, we propose to investigate pressure effect on spiral spin orientation in the new multiferroic pyroxene SrMnGe2O6. We ask for 7 days on D3 to determine the spiral plane orientation under pressure. In complement with electrical measurement that we have already done, the proposed experiment should allow us to clarify the microscopic mechanisms leading to multiferroïcity in SrMnGe2O6

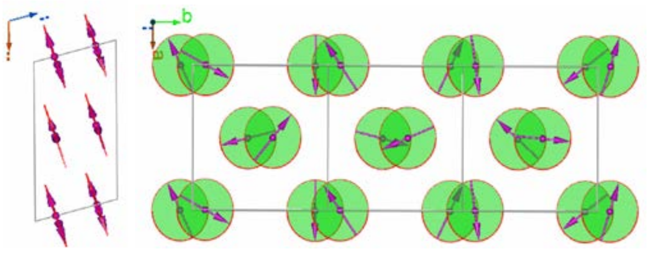
Experimental Report: cycle 192; IN20, 5-54-274

Pressure effect on spiral spin orientation in multiferroic pyroxene: $SrMnGe_2O_6$

Claire V. Colin, Céline Darie, Pierre Bordet, Pascal Lejay, Institut Néel, Grenoble
Noriki Terada, NIMS, Tsukuba, Japan
Navid Queshri, ILL, Grenoble, France

Scientific background: As a non-trivial magnetic system, pyroxenes with chemical formulas AMT_2O_6 (A = mono- or divalent metal, M = di- or trivalent transition metal, T = Si or Ge) containing magnetic cations have recently attracted revitalized interest because of their intriguing quasi-one-dimensional magnetic and multiferroic properties [1]. These properties are associated with the special arrangement of the pyroxene crystal structure, where almost isolated zig-zag chains of edge-sharing MO_6 octahedra are bridged by corner linked TO_4 tetrahedra chains. We investigated the crystal structure and magnetic properties of a new clinopyroxene compound $SrMnGe_2O_6$ (*C2/c*), by means of neutron powder diffraction and magneto-electric measurements [2]. $SrMnGe_2O_6$ undergoes two AFM phase transitions at T_{N1}=4.3 K and T_{N2}=4.0K, the second one being with incommensurate (propagation vector [0 q_y 0], q_y = 0.424) and associated with the appearance of spontaneous polarization. Recently, we investigated the magnetic and dielectric properties under a magnetic field by using both bulk measurements and neutron diffraction experiments by using newly grown single crystals. At zero field, below $T_{N2,}$ a cycloidal magnetic structure was determined for which the magnetic moments rotate along the *b* axis within a direction in the (*a*, *c*) plane. (Fig. 1a) The ordered magnetic moment of Mn^{2+} was refined to be 4.0 μ_B. Considering the crystal symmetry and cycloidal spin configurations (belonging to the magnetic point group m1'), polarization is allowed in any direction perpendicular to *b*. Two mechanism can be invoked to explain the appearance of the polarization: the "classical" inverse DM mechanism $\mathbf{p1} \propto \mathbf{e}_{ij} \times (\mathbf{S}_i \times \mathbf{S}_j)$ and the extended one $\mathbf{p2} \propto (\mathbf{S}_i \times \mathbf{S}_j)$. From the zero field magnetic structure, both mechanism can be invoked to explain the polarization direction and no one can be ruled out. More recently, we investigated the pressure effect on electric polarization in $SrMnGe_2O_6$, which shows drastic change in the polarization direction under pressure (see figure 1b). We anticipate that the polarization change is associated with varying spiral plane direction. However, so far, the magnetic structure under pressure have not been investigated, due to the experimental difficulty. The aim of the study is to to clarify the pressure dependence of spiral plane direction by using Spherical Neutron Polarimetry experiment with recently developed Hybrid-Anvil-Cell (HAC), [3] in order to understand how electric polarization components, p1 and p2, correspond to spiral plane direction. It is essential to use CryoPAD and Hybrid-Anvil-Cell to determine details of magnetic structure including precise spiral plane direction under high pressure.

Experimental report: The experiment was initially planed on D3 but was transferred to the IN20 beamline and performed from September 9 to September 16 (today, the deadline of the proposal round!). The sample was mounted with *a*-axis vertical to provide access to [0KL] to measure magnetic Bragg reflections, such as (0,q,0) and (0,q,2). We started by measuring a large sample in order to check the polarization matrix at ambient pressure with k_i=5.5A^{-1}. We faced already several technical problem due to some irregularity in the marble floor of IN20: some area in 2theta were not accessible (the motors failed to reach the good position) and furthermore the vibration due to these attempts moved away the cryopad and the nutator and affected the polarization measurement. We decided to change the incident wave vector for 4.1A^{-1} for the rest of the experiment. It took then us 4 days to align properly the small sample (0.5 * 0.5 * 0.2 mm) inside the HAC. It is very difficult tack because of the very low signal due to the sample size and the restricted accessible area in Q-space due to the pillar of the cell. Finally we succeeded in measuring Pxx, Pyz, Pzz on one magnetic reflection at 3GPa (see figure 2a and b) but we were not able to go at higher pressure. Therefore we ask for a continuation proposal.



Zero magnetic field

Figure 1a: Scheme of the cycloidal magnetic structures of $SrMnGe_2O_6$ at 2K projected onto the a–c plane (left) and the (a, b) plane (right) for zero field (Top) and 10T applied along the a direction(bottom).

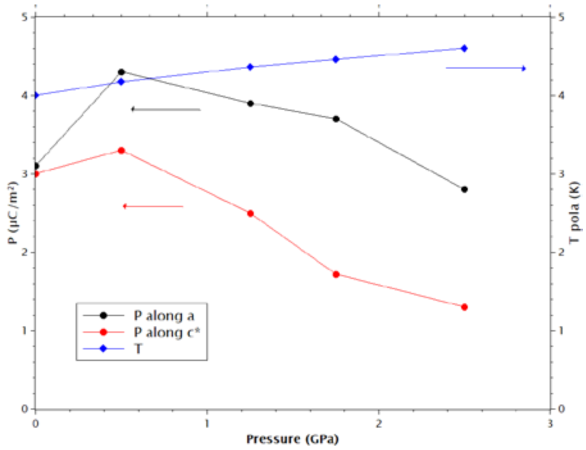


Fig 1b: Evolution of polarization and transition temperature with the applied pressure

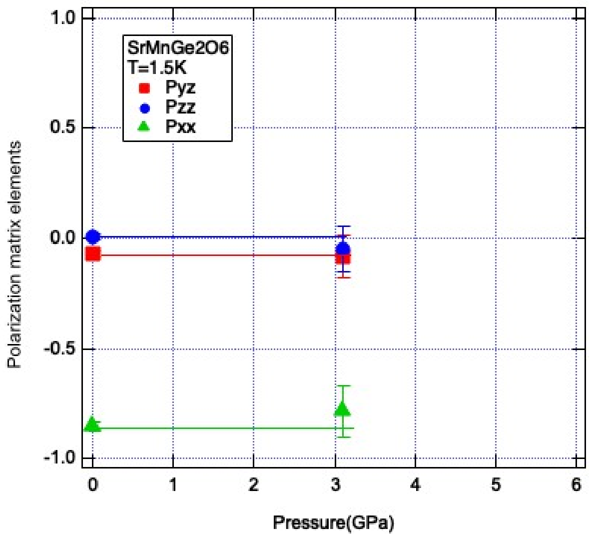


Fig. 2a: Evolution of the polarization matrix elements measured on IN20 up to 3 GPa

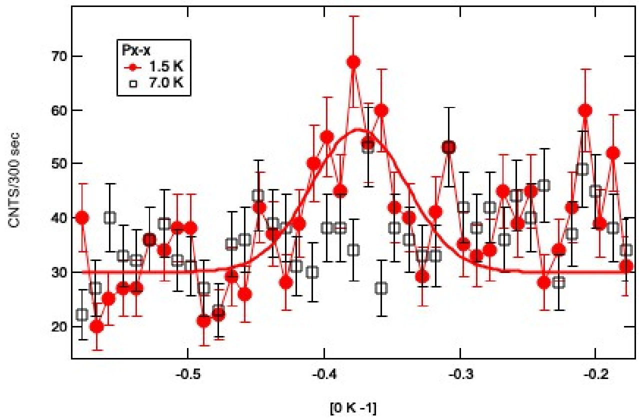


Fig 2b : Q-scan of the measured (0 0 1) – magnetic satellite at 3GPa

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[2] L. Ding, C. V. Colin, C. Darie, and P. Bordet, "$SrMGe_2O_6$ (M = Mn, Co): a family of pyroxene compounds displaying multiferroicity," *J. Mater. Chem. C*, vol. 4, pp. 4236–4245, 2016

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