Experimental Report

23/01/2015

Proposal: **5-31-2320** **Council:** 4/2014

Title: New commensurate structure inducedby pressure in YMn_2O_5 multiferroic

This proposal is a new proposal

Researh Area: Physics

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Samples: YMn_2O_5

Instrument	Req. Days	All. Days	From	To
D20	3	3	21/11/2014	24/11/2014

Abstract:

Among the multiferroics, YMn_2O_5 was studied in great details; with decreasing temperature, its magnetic phase sequence shows a transition from paramagnetic to an incommensurate (IC) spin structure at 44K, then a transition to a commensurate (CM) magnetic phase at 39K, finally a transition to another IC structure, accompanied by a sudden drop of the polarization, occurs at 23K. A previous study under pressure shows strong changes of the polarization, with a change of sign in the low temperature IC phase at 1GPa, suggesting that a CM phase is being stabilized by pressure. The nature of this phase is however unknown. To study it, we have started high pressure neutron diffraction measurements on G6.1 (LLB) up to 3GPa. Surprisingly, the IC modulation persists at high pressures, well above the change of sign of the polarization (1Gpa). Moreover, we observe a new CM phase with propagation vector (K=0,0,0.5) instead of (0.5,0, 0.25) at ambient pressure. We propose to study this new phase in details on D20 and to apply higher pressures to see if it can be obtained as single phase and better characterized.

Among the multiferroics (MF), RMn2O5 compounds (R= rare earth, Bi or Y) received a lot of attention because of their complex magnetic structures, some of which at the origin of their ferroelectricity. With decreasing temperature, their common phase sequence first shows a transition from paramagnetic to an incommensurate spin structure at TN1, with an incommensurate (IC) modulation along *(a,b),* the structure remaining para-electric. Then a transition to a commensurate (CM) magnetic phase (k=0.5, 0, 0.25) occurs at TC1, accompanied with the onset of ferroelectricity. Finally a transition back to an incommensurate structure occurs at TC2. The unlocking of the q vector at TC2 is accompanied by a sudden drop of the polarization, which can even change sign. YMn2O5 (with TN= 44K, TC1=39K and TC2=23K) was studied in great details [1-3], since the analysis of the magnetic structure is simplified by the non-magnetic Y.

In multiferroics, applying pressure is an original way to check interactions schemes, since pressure can change the magnetic order and possibly the polarization, by changing the balance between magnetic interactions. In our previous studies recently performed on G61 at LLB [4] we observed pressure induced transitions such as INC-CM in TbMnO3, F-AF in BiMnO3, or a decrease of the ordered magnetism in YMnO3 or LuFe2O4.

Under pressure, YMn2O5 shows strong changes of the polarization [5] with a change of sign at low T induced by pressure (Fig. 1, suggesting that a CM phase is being stabilized by pressure. The nature of this phase is however unknown. To study it, we first started high pressure neutron diffraction measurements on G6.1 spectrometer (Fig. 2) up to 3GPa. We observed **a new CM phase (CM2)** with propagation vector (K=0.5, 0, 0.5) instead of (0.5, 0, 0.25) at ambient pressure (figure 2).

We have studied this new phase on D20, which allowed us to increase both the pressure range and Q-range. Two sets of diffraction patterns and their thermal evolutions were measured at 4.2±0.3 GPa and 6.3±0.3GPa, thanks to the Paris-Edinburg pressure cell. For 4.2GPa, we measured at 5 temperatures (2 to 3 hours counting each) in the range of 5K-50K to check the thermal evolution of the new phase.

Due to the weakness of the signal when the pressure was increased we measured only three temperature at 6.3GPa with a very long counting time to get a good signal to noise ratio (12h at 5K, 4h at 30K and 12h at 55K in the paramagnetic phase to perform a subtraction and have access to the magnetic signal only).

About 10 hours were necessary to decrease the temperature and increase it again at 300K to change the pressure in a hydrostatic way, when the ethanol/methanol transmitting medium is liquid.

Three important results were obtained thanks to this experiment:

- The first one is the confirmation of the **coexistence of the new phase CM2** with the incommensurate (IC) and the commensurate (CM1) phases, ***in a very large pressure range (from 0.5 GPa up to the maximum studied pressure of 6.3 GPa)***.
- The second result is the ***linear growth of the new phase peak under pressure***. The pressure at which the new CM structure should be a single phase as been extrapolated between 9GPa and 16GPa depending on the chosen model (see Fig. 4).
- The third result is the availability to ***check a potential spin structure*** for the new phase in ***an extended Q-range***. Our new results **confirm the propagation vector** (k=0.5,0,0.5), postulated from G61 patterns with a single Bragg peak. Refinements of the D20 and G61 patterns show that the structure analogous to the CM1 one but with another piling of the Mn sheets (+-+- instead of ++--) along the *c* axis can fit the data.

Problem encountered during the experiment: We also tried to check the EOS at 300K, unfortunately a huge 2θ-dependent decrease of signal was observed when we increased pressure at ambient temperature (likely coming from experimental setup, but still unexplained). This problem limited our study to 6.3 Gpa and forced us to increase the counting time. Fortunately we partly recovered the signal at low temperature (below 100K) allowing us to measure good diffraction pattern in the magnetic phase.

The observation and characterization of this new phase is an important result which must be taken into account to explain the variation of the electric polarization with pressure in this model family of multiferroics. A paper is currently being written on these measurements, combining ILL and LLB data [7].

[1] L. Chapon *et al* PRL **96**, 097601, (2006)

[2] Kim *et al* PRB **78,** 245115, (2008)

[3] P. G. Radaelli *et al* PRB **79**, 020404R(2009)

[4] O. Makarova *et al* PRB RC (2011) ; APL 103, 082907 (2013) ; D. Kozlenko et al PRB (2010) ; PRB (2009)

[5] Chaudury *et al* PRB **77**, 220104R, (2008)

[6] G. R. Blake *et al* PRB **71**, 214402, (2005).

[7] M. Deutsch *et al* to be submitted to Phys. Rev. B Rapid comm. (2015)

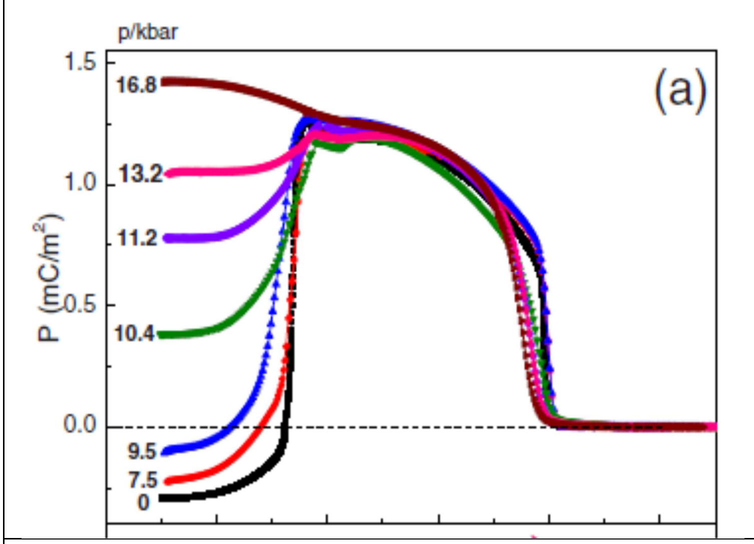

Fig. 1: Temperature dependence of the polarization **P** for different pressures, from [5]

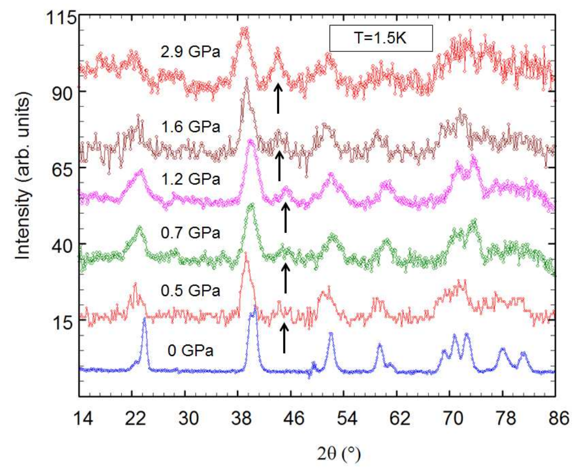

Fig 2: Magnetic pattern in YMn2O5 for different pressures, measured on G61 (λ=4.74A); arrows show the new CM structure.

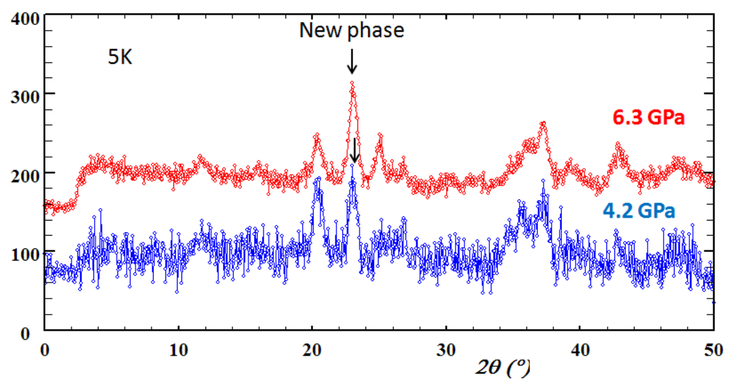

Fig. 3: Magnetic pattern in YMn2O5 for the two pressures measured on D20 (λ=2.42A); arrows show the new CM structure.

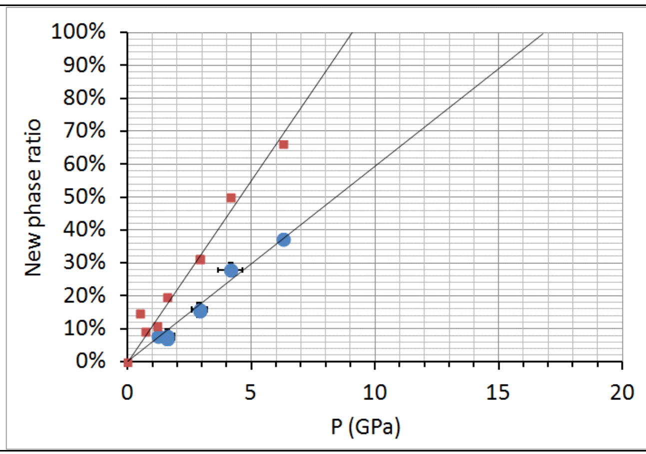

Fig4: ratio of the new phase CM2. The ratio is based either on the integrated intensities (red squares), or on refined scale factors (blue dots), assuming a CM2 structure identical to CM1 (but with another k vector)