Experimental report

20/10/2016

Proposal: **5-31-2446** **Council:** 4/2015

Title: neutron study of the new pressure induced commensurate phase in RMn2O5 multiferroics

Research area: Physics

This proposal is a new proposal

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Samples: TbMn2O5
DyMn2O5 with isotopic 163Dy

Instrument	Requested days	Allocated days	From	To
D20	5	5	25/05/2016	30/05/2016

Abstract:

RMn2O5 multiferroics show complex magnetic structures, highly sensitive to pressure with strong consequences on the ferroelectric properties. The magnetism and chemical pressure of the R3+ ion also have a strong impact on these properties. In YMn2O5 where Y is non magnetic, we recently discovered new pressure induced commensurate phase by high pressure neutron diffraction (up to 4 GPa and 7 GPa on G61 and D20 respectively). We propose to study this new phase under very high pressure in 2 compounds with magnetic rare earth: TbMn2O5, where our preliminary data show that this phase indeed exists, and DyMn2O5, where it could perhaps be identified to the X para electric phase previously detected by measurements of the electric polarisation.

Among RMn_2O_5 multiferroic compounds (R= rare earth, Bi or Y) a lot of magnetic structures and different behaviors appear, depending on the rare earth ion. Even with the same rare earth ion these compounds undergo with temperature a cascade of magnetic transitions giving rise to commensurate or incommensurate magnetic structures [1,2]. With decreasing temperature, the "classical" phase sequence is the following

1. Transition from paramagnetic to an incommensurate spin structure at TN1, with an incommensurate (ICM) modulation along (a,b), the structure remaining para-electric.
2. Transition to a commensurate (CM) magnetic phase (k= (½ 0 ¼)) at TC1, accompanied with the onset of ferro-electricity.
3. Transition back to an incommensurate structure occurs at TC2 (k= (½±δ 0 ¼+ε)). The unlocking of the q vector at TC2 is accompanied by a sudden drop of the polarization, which can even change sign.

However, the diversity of behavior, even on the propagation vector, is huge for this family:

- (½ 0 ¼) YMn2O5, TbMn2O5, HoMn2O5, ErMn2O5 (between ~20K and ~38K)
- (0 0 ½) LaMn2O5, PrMn2O5
- (½ 0 0) DyMn2O5* below 8K, EuMN2O5 below 5K
- (½ 0 ½) BiMn2O5 at all temperatures (no CM-ICM transition)

Even for the most studied compounds (YMn2O5 and TbMn2O5) the detailed magnetic structure was highly debated [3] and pressure was used to unveil the mechanisms at the origin of these behaviors at a macroscopic scale [4,5] or microscopic one [6].

Our previous studies on YMn2O5 allow us to observe a **new pressure induced commensurate phase (PCM)**, which appears at low pressure and progressively develops at the expense of the known CM and ICM phases with increasing pressure [6]. Refinements yield a propagation vector k= (½ 0 ½) for this new phase as in BiMn2O5 compound. We suspect that this new phase plays a crucial role in the surprising variation of the electric polarization and dielectric constant with temperature, which strongly evolve under pressure, and also depends on the R ion.

Our main objective was to confirm the generality of this new PCM phase in the RMn2O5 family by observing it in TbMn2O5 and DyMn2O5, two potential candidates since there macroscopic behavior under pressure show some similarities with YMn2O5 [4].

Several diffraction spectra at low temperatures and two pressures were measured for each compound during these five days (4.4 and 5.6 GPa for TbM2O5, 2.4 and 6.6 GPa for DyMn2O5). A Pb shave inserted in the powder was used as pressure probe. The pressure was increased at ambient temperature when the pressure transmitting medium (ethanol/methanol) is liquid and provides an isotropic pressure afterward the temperature was cooled down.

Four major results were obtained thanks to this experiment:

- The PCM phase is observed in both compounds (see figure 1 (a) and 2) confirming the generic nature of this phase.
- In TbMn2O5 at the highest pressure the PCM phase was observed as a single phase allowing a precise refinement of its magnetic structure (figure 1 (b)).
- The PCM phase is not observed in DyMn2O5 at the lowest pressure (2.2GPa). It only appears at the highest pressure (6.6Gpa).

- As already known, at ambient pressure DyMn2O5 shows two magnetic phases: below 32K an ICM1 (k = (0.49,0,0.25)) phase appears and then a transition to a commensurate phase CM2 k = (0.5,0,0) occurs below 8K with an overlapping of the two phases around 8K.
  Our experiment shows that the PCM phase grows at the expense of the ICM1 and not at the expense of CM2, which suggests that the CM2 phase is more stable under pressure than the ICM1.

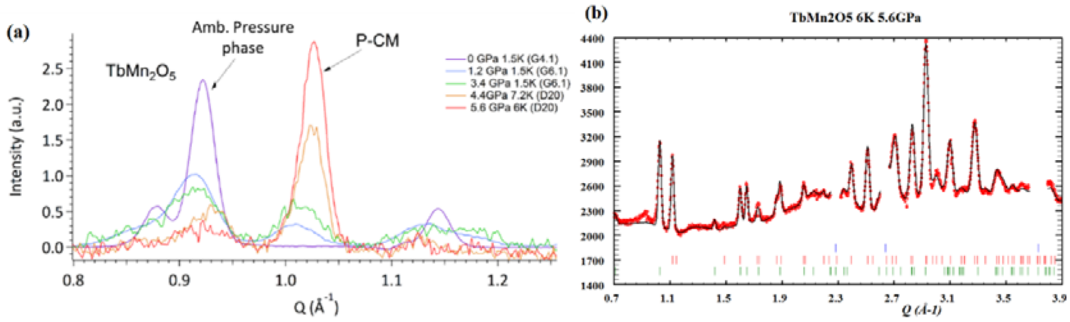

Figure 1 (a) $TbMn_2O_5$ at low temperature and different pressures. Data combine previous measurements (G6-1 at LLB) and measurement at higher pressure (D20-ILL). The growing of the PCM phase is confirmed at the expense of the ICM and CM phases, which fully disappear around 5.6 GPa. (b) refinement of the structural and the PCM magnetic phase obtained at 5.6GPa in $TbMn_2O_5$

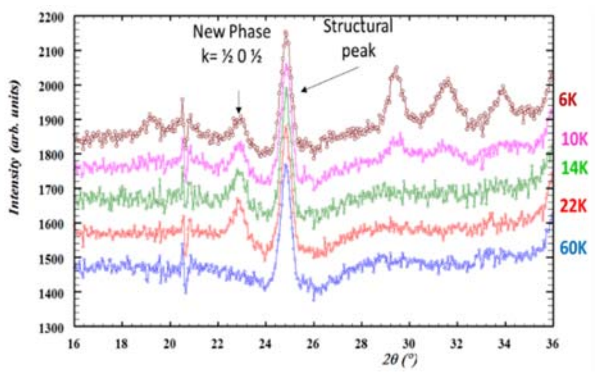

Figure 2 DyMn2O5 under pressure at 6.6 GPa: the new PCM phase is clearly observed and coexists with the CM1

In summary, the comparison between R=Y, Tb and Dy compounds suggests that stabilizing a pressure induced commensurate PCM phase with k_Z = ½ is a generic feature of the RMn_2O_5 family. Since Y is non-magnetic, the main effect of pressure is therefore a change in Mn-Mn interactions. For R=Tb or Dy the strong R magnetism allows us to refine the magnetic structures more precisely than for Y. We find that the PCM phase appears at the expense of ambient pressure phases with k_Z close to 0.25 (k_{ZCM} = ¼ or k_{ZICM} = ¼+ε), whereas phases with k_Z = 0 or ½ are more stable under pressure. The magnetic structure in the (a,b) plane seems to be quite stable. The whole study [to be published] contributes to a general understanding of magnetic interactions and pressure induced phase diagrams in RMn_2O_5 family, which strongly influences the electric polarization.

[1] Radelli *et al* J. Phys.: Condens. Matter 20 (2008) 434213

[2] Noda *et al* J. Phys.: Condens. Matter 20 (2008) 434206

[3] Chapon *et al* PRB 77, 134434 / J.-H. Kim *et al.*, Phys. Rev. B 78, 245115 (2008) / S.Wakimoto, *et al*, Phys. Rev. B 88, 140403 (2013)/ S. Partzsch *et al* Phys. Rev. Lett. 107, 057201 (2011)/ P. G. Radaelli, *et al* Phys. Rev. B 79, 020404 (2009)/ R. P. Chaudhury, et al, Phys. Rev. B 77, 220104(R)(2008). // Wilkinson *et al* PRB **84**, 224422 (2011) / Blake *et al* PRB 71, 214402

[4] C. R. dela Cruz, et al PRB 76 174106 (2007).

[5] R. P. Chaudhury, et al PRB 77, 20104(R)(2008).

[6] Deutsch et al PRB **92**, 060410(R) (2015) // Kozlenko *et al* PRB **92**, 134409 (2015)