ESRF	**Experiment title:** XMCD study of the pressure-induced magnetoelectric phase transition on multiferoic $TbMnO_3$	**Experiment number**: HC2654
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Report:

The application of high pressure on $TbMnO_3$ recently opened a breakthrough on the field of multiferroic magnetoelectric materials, showing magnetic ordering and a spontaneus electric polarization of the order of standard ferroelectrics for the first time [1]. In particular, the orthorhombic oxide $TbMnO_3$ is a widely studied multiferroic material, where a complex non-collinear magnetic ordering breaks the inversion symmetry of the system and triggers the appearance of spontaneous electric polarization P_s (~ 0.07 μC/cm^2) parallel to the **c** axis, below 27 K. The application of physical pressure induces a pressure-induced polarization-flop transition from **c** to **a** axis around 4.5 GPa and below 20 K [1]. The value of P_s under these conditions is the largest ever reported on a magnetic-driven ferroelectric, that is 1 μC/cm^2, comparable with standard ferroelectrics polarization values. Besides that, in the high pressure ***P***||**a** phase, P_s is strongly affected by magnetic fields and at 5 K, 8 T and 4.2 GPa it reaches 1.8 μC/cm^2, the same order of magnitude of many conventional FE.

In this experiment on ID12 beamline, the pressure induced magnetoelectric phase transition on $TbMnO_3$ (P_c= 4.2 GPa, T <15K, under H > H_c= 2 T along **a** direction) [1] has been explored by X-ray absorption spectroscopy (XAS) and X-ray magnetic circular dichroism (XMCD) at Tb L_3 edge and Mn K edge. For this purpose, we used a $TbMnO_3$ single crystal (**a**-axis perpendicular to sample surface and parallel to H direction) in total fluorescence yield at T = 7 K using a diamond anvil cell (600 μm cullet) with Si oil as a pressure transmitting medium. Three pressure points have been investigated, below (P=1 GPa) and above (P=5, 14 GPa) the magnetoelectric transition. It is worth noting that the region P = 14 GPa at low T under magnetic fields was explored for the first time on this experiment.

Tb L_3 XMCD signal intensity decreases as pressure increases above P_c, as it is shown in figure 1. This intensity reduction above P_c is in agreement with a 21% decrease on F_x component of Tb magnetic moment, being this value slightly below the one found by neutron diffraction [2]. Besides that, the intensity of the white line at the XANES region decreases with pressure as well, which might indicate an electronic transition on Tb ion at higher pressures.

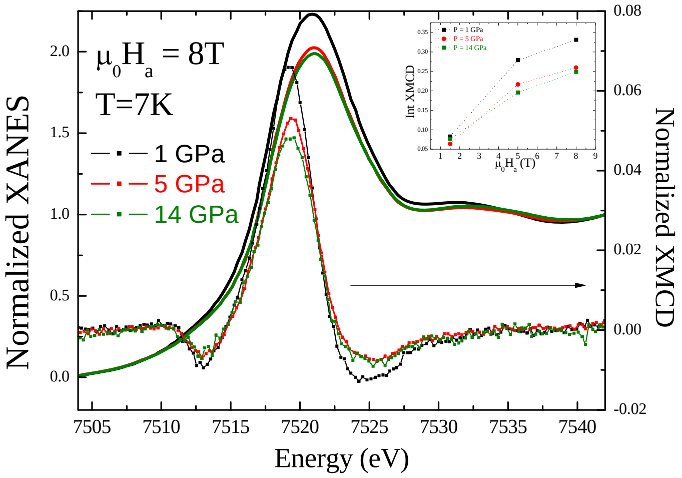

Fig. 1. Tb L_3 edge normalized XANES (left y-axis) and XMCD (right y-axis) at μ_0Ha=8T and T=7 K for three different pressure points below (1GPa) and above (5, 14 GPa) the magnetoelectric transition. Inset: Integrated XMCD signal as a function of H.

For P = 1GPa < P_c, Mn K edge shows XMCD signal (0.4% of XAS signal for H=8 T, not shown here), which disappears at 5 GPa. This is in agreement with an antiferromagnetic arrangement of Mn moments at 5 GPa under magnetic field, predicted theoretically [1] and more recently proposed by neutron diffraction [2]. However, as shown on figure 2, there is a re-entrant XMCD signal of 0.6% of XAS signal at 14 GPa and H=1.5 T (below H_c). This indicates that at pressures above 5 GPa a new ordering of Mn moments take place, with a considerable ferromagnetic component along **a** axis. The disappearance of the antiferromagnetic arrangement of Mn sublattice at higher pressures might have important implications on the recently found high pressure multiferroic phase of $TbMnO_3$.[1]

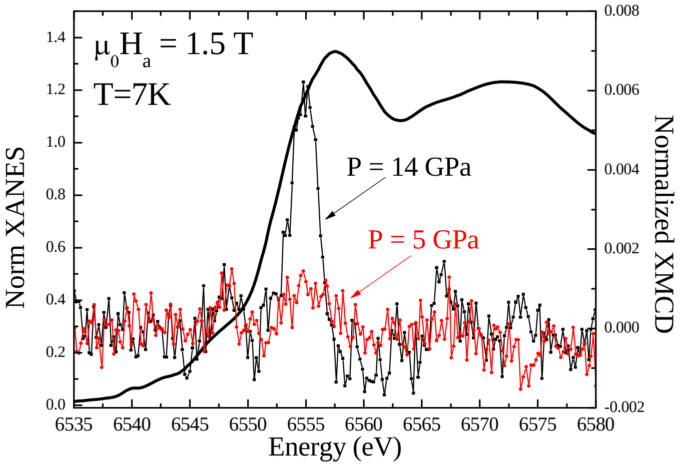

Fig. 2. Mn K edge normalized XANES (left y-axis) and XMCD (right y-axis) at μ_0Ha=1.5T and T=7 K for P=5 GPa (red) and 14 GPa (black).

[1] T. Aoyama et al., Nature Comm **5**, 4927 (2014)

[2] N. Terada et al., Phys. Rev. B **93**, 081104(R) (2016)