



	Experiment title: Study of the TbMnO ₃ local distortions by x-ray resonant scattering at the Mn K-edge and Tb L ₃ -edge	Experiment number: HE 2761
Beamline: ID 20	Date of experiment: from: May 13, 2008 to: May 20, 2008	Date of report: 1/09/08
Shifts: 18	Local contact(s): Javier Herrero-Martín	<i>Received at ESRF:</i>
Names and affiliations of applicants (* indicates experimentalists): Dr. Joaquín García*, Dra. Gloria Subías*, Dr. Javier Blasco* and Vera Cuartero* Instituto de Ciencia de Materiales de Aragón, CSIC – Univ. de Zaragoza, Zaragoza (Spain).		

Report:

TbMnO₃ perovskite is the representative of materials where antiferromagnetism (AFM) and ferroelectricity coexist at low temperatures. Neutron diffraction experiments have shown that Mn³⁺ magnetic moments go through an incommensurate AFM sine order below 40 K, described by a wave vector $\mathbf{k}_{\text{Mn}} = (0, 0.29, 0)$ in the orthorhombic unit cell $Pbnm$. The \mathbf{k}_{Mn} vector decreases with temperature down to a *quench* at $\mathbf{k}_{\text{Mn}} = (0, 0.28, 0)$ with a $T_{\text{quenching}} \sim 28\text{K}$. At this temperature a magnetization component caused by Mn sublattice along \mathbf{c} axis is developed. TbMnO₃ shows then a ferroelectric order at $T_C \sim T_{\text{quenching}} \sim 28\text{K}$. [1] In this phase, the Tb moments also order with the same wave vector as the Mn, but they are transversally polarized along the \mathbf{a} axis. [2] Moreover, below 7 K there is an AFM order of Tb³⁺ magnetic moments, showing a magnetization component along \mathbf{a} axis, with a wave vector $\mathbf{k}_{\text{Tb}} = (0, 0.415, 0)$. However, the microscopic origin of the magnetically induced ferroelectricity is still unknown.

A resonant x-ray scattering study at the Mn K-edge and Tb L₃-edge of TbMnO₃ was performed to investigate the local distortions responsible for the ferroelectricity. Forbidden reflections $(0\ k\ 0)$, $(0\ 0\ l)$ with $k, l = 3, 5$ were measured in single-crystals cut with $[010]$ and $[001]$ surfaces normal at room temperature. The dependence of the intensity for these reflections on temperature and azimuthal angle was investigated in detail. A Cu (220) crystal was used to analyze the polarization of the scattered beam: perpendicular (σ') or parallel (π') to the scattering plane. We also observed some incommensurate reflections of the type: $(0, 2+q_{\text{Mn}}, 0)$, $(0, 4-q_{\text{Mn}}, 0)$, $(0, 4-q_{\text{Mn}}, 1)$ in both edges and $(0, 3-q_{\text{Mn}}, 0)$, $(0, 1-q_{\text{Mn}}, 3)$ and $(0, \pm q_{\text{Mn}}, 3)$ reflections only in Tb L₃ edge, being $q_{\text{Mn}} = 0.28$.

We can see in figure 1 (a) the intensity of (030) forbidden reflection as a function of the photon energy at different azimuthal angles in the σ - π' channel, at room temperature. A strong resonance is observed at 6552 eV, just at the Mn K-absorption edge. Scattering intensity is only observed in the σ - π' polarization channel for all forbidden reflections studied and when the crystal is rotated around its scattering vector, the σ - π' intensity of these reflections show a $\cos^2\phi$ dependence. There is a $\pi/2$ periodicity of the resonant intensity on the azimuthal angle ϕ for $(0,k,0)$ and $(0,0,l)$ reflections.

We have also measured the temperature evolution so that in figure 1 (b) we can see the integrated intensity of the forbidden reflections evolving with temperature. Here, we could not find any significant intensity variation either around the Néel temperature, $T_N \sim 40$ K, or at $T_C \sim 28$ K.

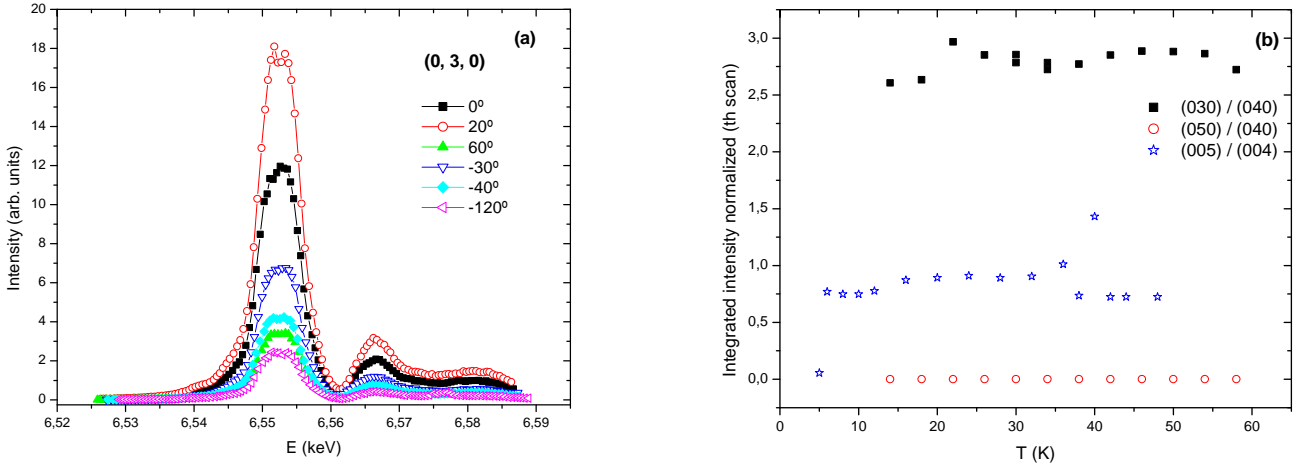


Figure 1. (a) Energy dependence of the forbidden (0, 3, 0) reflection measured at different azimuth angles, in the $\sigma \rightarrow \pi'$ channel at the Mn K absorption edge. (b) Integrated intensity of the (0k0) and (00l) (θ scan) of forbidden reflections as a function of temperature.

On the other hand, figure 2 shows the energy dependence of the observed incommensurate reflections in the σ - π' channel at the Mn K edge. It is worth noting that the resonance observed at the (0, $4-q_{Mn}$, 1) reflection is slightly shifted to lower energies with respect to the (0, $2+q_{Mn}$, 0) and (0, $4-q_{Mn}$, 0) reflections. This indicates that the resonant signal at these (0, $2+q_{Mn}$, 0) and (0, $4-q_{Mn}$, 0) reflections could have a different origin, taking into account that the (0, $4-q_{Mn}$, 1) reflection has the hallmarks of a resonant x-ray magnetic scattering (A-type AFM).

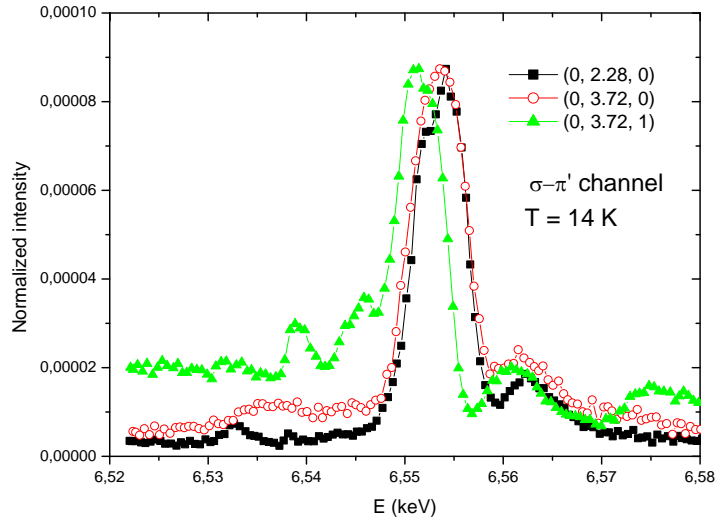


Figure 2. Energy dependence of the incommensurate reflections, polarization-resolved ($\sigma \rightarrow \pi'$) at the Mn K absorption edge at $T = 14$ K. The first two are normalized to the maximum of intensity of the third.

Finally, the behavior of the studied forbidden reflections closely resembles to LaMnO_3 [3], which suggests that in this case they are also originated by anisotropic tensor scattering.

- [1] S. Quezel *et al.* Physica B+C **86-88**, (1977), 916-918
- [2] M. Kenzelmann *et al.* Phys. Rev. Lett. **95**, 087206 (2005).
- [3] G. Subías *et al.* Phys. Rev. B **75**, 235101 (2007)