ESRF	Experiment title: Resonant Soft X-ray scattering from TbMnO <sub>3</sub> .	Experiment number: HE 2367
<b>Beamline</b> : ID08	<b>Date of experiment</b> : from: 04/04/07 to: 10/04/07	<b>Date of report</b> : 17/05/07
Shifts: 18	Local contact(s): F. Yakhou-Harris	Received at ESRF:
<b>Names and affiliations of applicants</b> (* indicates experimentalists):		

**T. Forrest**\*, **D. McMorrow**\*, **H. Walker**\* (Dept. of Physics & Astronomy, University College London, UK)

**P. Hatton, T. Beale\*, S. Bland\*** (Dept. of Physics, University of Durham)

S. Wilkins (Brookhaven National Laboratory, New Jersey, USA)

**D. Mannix** (Xmas, CRG beamline, ESRF)

Report:

TbMnO<sub>3</sub> is a magnetoelectric multiferroic; a class of materials in which ferroelectricity and magnetism coexist [1]. TbMnO<sub>3</sub> can be further sub-categorised as an improper ferroelectric [2], this means that the ferroelectricty is induced by TbMnO<sub>3</sub>'s complex magnetic structure, thus the electric and magnetic order parameters are closely linked, and the electric polarisation is highly sensitive to an external magnetic field [3]. This property is of great interest as it might provide useful applications for the spintronics industry. However, before any practical applications can be constructed, a microscopic theory of the coupling mechanism in this type of multiferroic is needed.

TbMnO<sub>3</sub> exists in the following magnetic phases:

- **T>42K**, TbMnO<sub>3</sub> has a orthorhombically distorted perovskite structure (*Pbmn* space group), **Paramagnetic** phase.
- **28K**<**T**<**42K**, **Collinear** phase: The Mn magnetic moments are sinusoidally modulated along the baxis, with a incommensurate propagation vector of  $(0 q_{Mn} 1)$ , where  $q_{Mn} \sim 0.29$  [r.l.u.] at 42K. Upon a further decrease in temperature the propagation vector  $q_{Mn}$  decreases in value until it reaches a minimum value of  $q_{Mn} \sim 0.28$  [r.l.u.] at 28K.
- T<28K, Cycloidal phase: The Mn magnetic moments form a spiral magnetic structure in the b-c plane. This phase also has an incommensurate wave vector of (0 q<sub>Mn</sub> 1) where q<sub>Mn</sub>~0.28 [r.l.u.] and is constant upon further decreases of temperature. In addition to breaking time-reversal symmetry, this magnetic spiral structure also breaks inversion symmetry, allowing for an electric polarisation along the c axis. [2]
- **T**<**7K**, In addition to the ordering on the Mn sublattice, at temperatures below 7K, the Tb magnetic moments order with an incommensurate propagation vector of (0 ~0.42 1) [r.l.u.].

TbMnO<sub>3</sub> has been extensively studied with neutrons [4,5] and by Mannix et al [6] using resonant scattering (RXS) of x-rays with energies equal to the Mn K (6589 eV) and Tb  $L_3$  (7514 eV) absorption edges on the ID20 ESRF beamline and the CRG XmaS beamline.

However, both neutrons and high energy RXS have their limitations:

Firstly, neutrons cannot separate the magnetic ordering of the different sublattices and cannot see other forms of ordering such as orbital ordering, which may play an important role in the multiferroic properties of these crystals.

Secondly, because of the electric dipolar selection rules, the E1 transition used in the RXS performed by Mannix et al [6] did not probe the Mn 3*d* and Tb 4*f* electronic shells, which are directly involved with the magnetic ordering of this crystal.

RXS of soft x-rays is both sensitive to the magnetic ordering on the different sublattices, can detect other forms of ordering and can directly probe the E1-E1 transition to the Mn 3d (L<sub>2</sub> & L<sub>3</sub> absorption edges) and the Tb 4f (M<sub>4</sub> & M<sub>5</sub> absorption edges) electronic shells.

Measurements were made on the ID08 beamline's 5-circle diffractometer, using a single crystal prepared in Oxford by D. Prabhakaran and cut with a [010] orientation. Because the diffractometer has a base temperature of ~20K, it was not possible to study the low temperature ordering of the Tb Magnetic moments. Previous experiments conducted on ID08 and the 5. U1 station at the SRS, (Daresbury, UK) have extensively studied the Manganese sublattice in the cycloidal phase. For this experiment, we turned our attention to the collinear phase, where azimuthal scans of the (0 q<sub>Mn</sub> 0) peak were taken. (See figure 1).

Figure 1: An azimuthal scan of the  $(0 q_{Mn} 0)$  peak at the Mn L<sub>2</sub> & L<sub>3</sub> absorption edges. The scans are fitted with a  $\sin^2(\Psi)$  function. The insert is an energy resonance scan of the peak.



In addition to azimuthal scans of the (0  $q_{Mn}$  0), (0  $2q_{Mn}$  0) and (0  $1-2q_{Mn}$  0) satellites, high resolution energy dependence scans were taken in the collinear phase (an example can be seen in the insert of figure 1). The second half of the experiment was concerned with studying the sample with x-rays equal in energy to the M<sub>4</sub> & M<sub>5</sub> absorption edges. Previous experiments had only detected the (010) Templeton peak at these energies with horizontally polarised incident x-rays. Therefore it was decided to switch to vertically polarised x-rays. Surprisingly, 2 peaks corresponding to the (0  $q_{Mn}$  0) and (0  $1-q_{Mn}$  0) where seen at the Tb M<sub>5</sub> absorption edge with vertically polarised incident x-rays, energy dependence scans suggested that there was no resonant enhancement at the Tb M<sub>4</sub> edge. The temperature dependence of the (0  $1-q_{Mn}$  0) peak at the Tb M<sub>4</sub> absorption edge was recorded, results showed that the peak disappears a T~25K. Suggesting that this peak was due to the polarisation of the Tb 4*f* electrons by the Mn sublattice? An azimuthal scan of this peak in the cycloidal phase was also recorded, however because of the peak's relative weakness no clear dependence was shown.

Analysis of the data is ongoing, we plan to perform polarisation analysis of the the 3 peaks seen at the Manganese L-edges at the SLS this March. We also plan to continue the experiment by studying a single crystal that is cut with a [0 0.28 1] orientation, allowing us observe the A-type (0  $q_{Mn}$  1) peaks at the Tb  $M_4$  & $M_5$  absorption edges.

## **References:**

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