Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office using the **Electronic Report Submission Application:**

http://193.49.43.2:8080/smis/servlet/UserUtils?start

Reports supporting requests for additional beam time

Reports can now be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title: An investigation of the orbital and oxygen octahedral tilt ordering in multiferroic $TbMnO_3$.**	**Experiment number**: 28 01 820
Beamline: XmaS-BM28	**Date of experiment**: from: 19/03/08 to: 26/03/08	**Date of report**: 03/09/08
Shifts: 21	**Local contact(s)**: Dr. Laurence BOUCHENOIRE	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Mr Thomas Forrest*, Dr Helen Walker*, Professor Desmond McMorrow (Dept of Physics and Astronomy, and London Centre for Nanotechnology, University College London, UK)

Professor Andrew Boothroyd (Dept of Physics and Astronomy, University of Oxford, UK)

Professor Peter Hatton, Dr Thomas Beale (Dept of Physics and Astronomy, Durham University, UK)

Dr Stuart Wilkins (Brookhaven national laboratory, USA)

Dr Danny Mannix (CNRS, Grenoble, France)

Report: $TbMnO_3$ is a well known magnetoelectric multiferroic [1]; a material which posses both long range magnetic and ferroelectric ordering in a single phase [2]. $TbMnO_3$ has an orthorhombically distorted perovskite crystal structure (*Pbmn* space group) and exists in the following magnetic phases:

- **T>42K**, **Paramagnetic, Paraelectric** phase.
- **28K<T<42K**, **Collinear** phase: The magnitude of the Mn magnetic moments are sinusoidally modulated along the b-axis, with an incommensurate propagation vector of (0 q_{Mn} 1), where q_{Mn} ~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} ~0.28 [r.l.u.] at 28K. $TbMnO_3$ is still paraelectric in this phase.
- **T<28K**, **Cycloidal** phase: The Mn moments form a spiral magnetic structure in the b-c plane. This phase also has an incommensurate wave vector of (0 q_{Mn} 1) where q_{Mn}~0.28 [r.l.u.] upon a further decreases of temperature the magnitude of q_{Mn} increase slightly. This magnetic spiral structure breaks both time-reversal and inversion symmetry, allowing for an electric polarisation along the c axis. [3]
- **T<7K**, Tb moments order with a separate incommensurate propagation vector of (0 ~0.42 1) [r.l.u.].

These spiral magnetoelectric multiferroics ($TbMnO_3$, $DyMnO_3$, $Ni_3V_2O_8$ etc) display their multiferroic properties only at low temperatures. Because of this, the development of a microscopic theory of the coupling mechanism is clearly of vital importance to these materials. The unique properties of X-ray resonant scattering (XRS), including its elemental specificity, and exquisite sensitivity to the multipolar order parameters that are predicted to characterise the combined ferroelectric/magnetic state [4], make it ideal for working towards such a goal.

Our research group has already conducted several detailed investigations into the incommensurate magnetic ordering of $TbMnO_3$ [5]. The purpose of this experiment was to perform a detailed investigation of the orbital and oxygen octahedral tilt ordering in multiferroic $TbMnO_3$. This can be achieved by studying the (010) Templeton peak using X-ray resonant scattering with photon energies tuned to Mn K and Tb L_3 absorption edges respectively. The motivation for this proposal is our observation of a surprising correlation between the intensity of the (010) peak measured at the Tb M edges and the transitions between the various low temperature phases displayed by $TbMnO_3$ (see Fig. 2).

A single crystal sample of $TbMnO_3$ cut with a b-face as the surface normal was prepared by Professor Boothroyd's group at the University of Oxford. The sample was initially mounted with the crystallographic c-axis pointing down the beamline, giving scattering in b-c plane. After the UB matrix was established at room temperature, the low temperature Joule-Thomson cryostat was mounted and the sample was cooled to a base temperature of 5K. Initial scans of the (010) reflection were made with incident X-ray photon energies in the vicinity of the Tb L_3 absorption edge (7.514 keV), to perform polarisation analysis of the scattered beam at these energies a Au(222) crystal was used.

Unfortunately, due to a number of technical problems, (the most serious of these was the failure of the water pump which cools the beamline's monochromator crystal, leading to the monochromator needing to be replaced) our experimental time was severely limited, we lost approximately 4 days of experimental time. In fact it was not possible to perform any measurements with Mn K edge incident photons (6.539 keV).

The measurements that were taken (at the Tb L_3 edge) include a temperature, azimuthal (at 5K) and several incident photon energy scans (at various temperatures) of the (010) reflection. The results indicate that the ordering of the Tb moments at a temperature of 7K might have an effect on the octahedral tilt ordering in TbMnO3, as the peak's intensity clearly drops for temperatures less than 10K. However, there does not seem to be any significant change in the peak's intensity near the magnetic phase transitions, at T=28K and T=42K, (as observed at the Tb M edges).

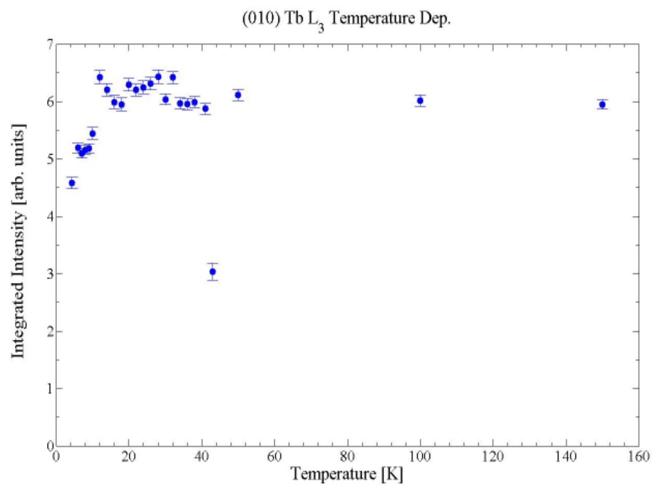

Figure 1: The intensity of the (010) Templeton peak at the Tb L_3 absorption edge as a function of temperature. The data point at T~42K is belived to be incorrect (due to experimental error).

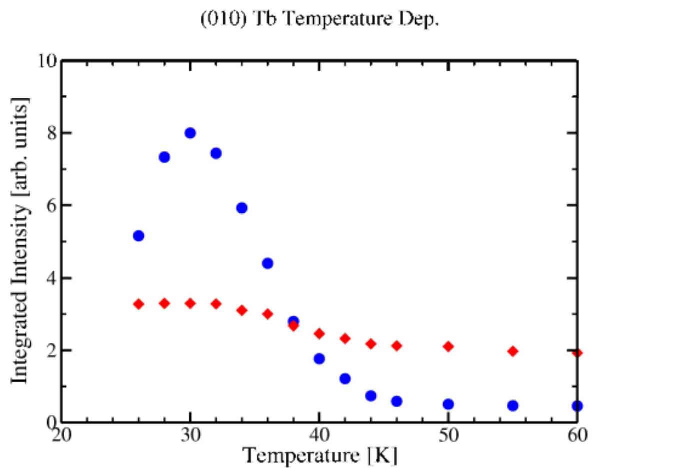

Figure 2: The intensity of the (010) Templeton peak at the Tb M_4 (red) & M_5 (blue) absorption edges as a function of temperature

Although the results obtain from this experiment are disappointing, they clearly show that there is no connection between the Manganese magnetic phases and the tilt ordering of the MnO_6 octahedral.

References:

[1] T. Kimura et al., Nature 426, 55 (2003)

[2] W. Eerenstein et al., Nature, 442, 759 (2006)

[3] M. Kenzelmann et al., Phys. Rev. Lett. 95(8), 087206 (2006)

[4] T. Arima et al., J. Phys. Soc. Jpn 74, 1419 (2005)

[5] D. Mannix et al, Phys. Rev. B, 76, 184420 (2007)