



**Experiment title: Magnetolectric order parameters in multiferroic DyMnO<sub>3</sub>**

**Experiment number:**  
HE2606

**Beamline:**

ID20

**Date of experiment:**

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15

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**Report:** *Magnetolectric multiferroics* form a class of materials in which magnetism and ferroelectricity coexist and interact. This endows them with a range of intriguing properties that - from a fundamental point of view - are a considerable challenge to understand at the microscopic level, and suggests a number of potential practical applications in the fields of spintronics, data storage, etc. [1]. Typically the chemical requirements for ferroelectricity and magnetism are mutually exclusive. Recently, however, a number of new routes to multiferroicity have been discovered, including the breaking of inversion symmetry (required for ferroelectricity) through the formation of complex magnetic structures [2]. The distorted perovskites REMnO<sub>3</sub> (RE=Tb and Dy) are a family of such multiferroics, in which ferroelectricity develops at the phase transition from a collinear to a spiral magnetic structure, with a concomitantly gigantic magnetolectric effect. This allows, for example, the electric polarization in DyMnO<sub>3</sub> to be switched by applied magnetic fields [3]. The magnetolectric ferroelectrics known so far display these properties only at low temperatures. The development of a microscopic theory of the coupling mechanism is clearly of vital importance to the technological advancement of 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. *In this experiment we continued our previous successful experiment on DyMnO<sub>3</sub> (HE2294) concentrating this time on the ordering of the manganese sublattice.*

A single crystal sample was prepared by the group of Tokura in Japan, and cut with a *b*-face normal. Measurements were made on the ID20 beamline using the low temperature displax cryostat to enable azimuthal measurements, at the manganese *K*-edge and the dysprosium *L<sub>III</sub>*-edge, in zero applied magnetic or electric field. ID20 is unique with its high flux and experimental set up in allowing us to investigate the manganese sublattice, since the resonant enhancement at the Mn *K*-edge is small. To gain an understanding of the magnetolectric order parameters it was essential to perform polarisation analysis and this was done using Cu (220) and Pt (222) analyser crystals.

A careful study of the manganese sublattice incommensurate peaks in the cycloidal phase was performed allowing us to identify the A, C, F and G-type reflections. These reflections were further investigated by measuring their energy, temperature and azimuthal dependence, where appropriate. A and F-type peaks were also observed at base temperature in the dysprosium ordered phase, at the manganese ordering wave vector but at the dysprosium  $L_{III}$ -edge.

The A and F-type peaks in sigma-pi have been identified as associated with the manganese ordering, while the peaks which appear to be second order C and G-type are temperature independent and hence unassociated with the magnetic ordering, see Figure. Second harmonic A and F-type peaks have also been observed in sigma-sigma, and these display the same temperature dependence as the first order magnetic peaks, disappearing above 33 K. There is still some discrepancy between the temperature dependence observed using XRS and the ordering temperature as reported from other techniques at  $T_N \sim 39$  K.

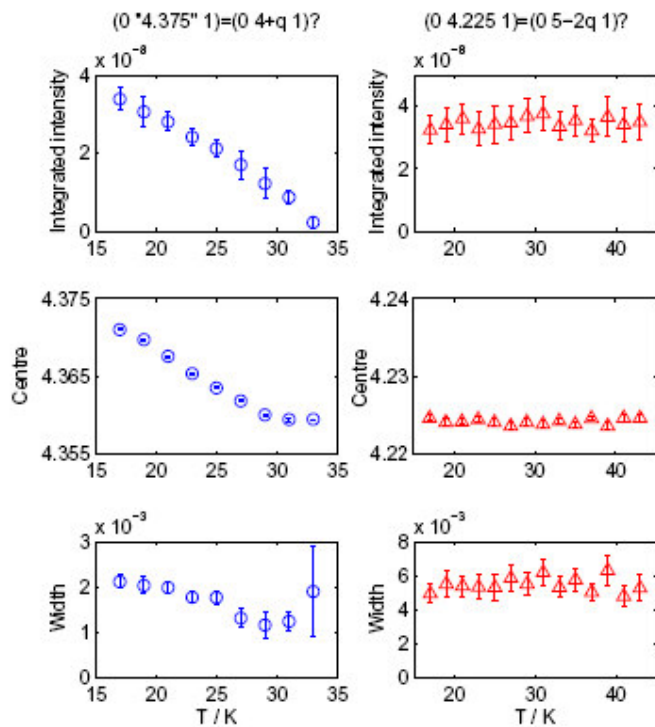


Figure: Temperature dependence of the “A”-type and “G”-type peaks in the sigma-pi channel at  $E=6.5492$  keV. The “A”-type peak disappears above  $T_{Mn}$  and shows the expected evolution of the incommensurate wave-vector, while the “G”-type peak intensity and position are independent of temperature.

The different roles of the A and F-type peaks within the cycloidal phase, at  $T = 15$  K, were investigated by means of energy and azimuthal measurements, which show distinct different behaviours for the two peaks. It will be interesting to investigate this point further by use of the phase plate to scatter from the two reflections with circularly polarised incident light, which has already proved informative for probing  $TbMnO_3$ , which displays similar magnetoelectric properties.

Further investigation will be necessary to investigate the azimuthal dependence of the C and G-type peaks to try and identify the order parameters with which they are associated. A complete study of the superlattice reflections must also be performed in the collinear phase,  $19 < T < 39$  K, in which  $DyMnO_3$  is paraelectric to gain a full insight of the magnetoelectric properties. It will also be interesting to apply magnetic and electric fields to further investigate the rich magnetoelectric phase diagram.

## References:

- [1] M. Fiebig, J. Phys. D 38, R123 (05)
- [2] H. Katsura et al., Phys. Rev. Lett. 95, 057205 (05)
- [3] T. Kimura et al., Phys. Rev. B 71, 224425 (05)
- [4] T. Arima et al., J. Phys. Soc. Jpn 74, 1419 (05)