

Experimental report

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Proposal: 5-54-153

Council: 4/2014

Title: Magnetic structure determination in o-TbMnO₃ and o-LuMnO₃ thin films by combining both neutron diffraction and spherical neutron polarimetry

Research area: Materials

This proposal is a new proposal

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Samples: TbMnO₃
LuMnO₃

Instrument	Requested days	Allocated days	From	To
D10	8	8	06/10/2014	14/10/2014
D3 CPA	8	7	17/04/2015 09/09/2015	18/04/2015 16/09/2015

Abstract:

Abstract: We propose two separate experiments to determine the magnetic structures of multiferroic and strained o-ReMnO₃ (Re=Lu,Tb) thin films grown on YAlO₃ substrates. In the first experiment, we request to use D10 in order to both identify and measure the strongest Bragg peaks due to the long-range symmetry-breaking antiferromagnetic (AFM) order in each film. Using this information, we request to subsequently use D3 with CryoPAD in order to conduct a spherical neutron polarimetry study of the strongest AFM peaks in each film. This second experiment is only made possible by recent pioneering technical developments at ILL where a unique zero-field four-circle device for CryoPAD provides in-situ control of the sample orientation, and hence of the magnetic interaction vector. From these experiments we aim to determine unambiguously both the magnetic structures in the strained thin films, and also the intrinsic magnetic domain populations. The latter result will show how the strain in the films may emerge as an effective tuning parameter for the magnetic and electric properties of the films.

Experimental Report

Magnetic structure determination in o-TbMnO₃ and o-LuMnO₃ thin films by combining both neutron diffraction and spherical neutron polarimetry

Experimental team

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Rare-earth manganite (RE₂MnO₇; RE= Tb, Lu) epitaxial films of thicknesses 80nm each, deposited on orthorhombic YAlO₃ (110) were used to perform these experiments. Both films when studied using X-ray diffraction and the reciprocal space mapping demonstrated twin free, epitaxial and highly strained growth. Neutron diffraction results on o-LuMnO₃ [110] oriented 80nm thick film demonstrated incommensurate magnetic phase ($0 \text{ qk} \sim 0.485 \text{ 1}$) unlike the commensurate E-type phase in bulk [1,2]. Growth induced strain in LuMnO₃ and TbMnO₃ thin films was attributed to play a crucial role in tuning their multiferroic properties. In contrast, [100] oriented TbMnO₃ film measured on D10, ILL demonstrated bulk like incommensurability ($0 \text{ qk} \sim 0.29 \text{ 1}$) similar to the bulk TbMnO₃ magnetic phase [3,4]. Hence we planned this experiment to resolve the magnetic structure precisely of the [110] oriented LuMnO₃ and TbMnO₃ films using polarimetry.

In the first section we performed neutron diffraction on both LuMnO₃ and TbMnO₃ films with D10. Incommensurate magnetic phase was identified for the LuMnO₃ ($0 \text{ qk} \sim 0.485 \text{ 1}$) (Fig.1) film comparable to previously reported results on [110] oriented films [1]. Temperature dependent study of the superlattice Bragg peak intensity indicates that the Néel temperature T_N is around 40K [Fig.1:Inset]. We managed to measure the following magnetic reflections within the available time frame : ($0 \text{ qk} \text{ 1}$), ($1 \text{ -1+qk} \text{ 1}$), ($2 \text{ qk} \text{ 1}$), ($1 \text{ qk} \text{ 1}$), ($0 \text{ qk} \text{ 3}$) and ($1 \text{ qk} \text{ 3}$) [Fig.2]. From these measurements it was clear that ($0 \text{ qk} \text{ 1}$) is the strongest magnetic reflection confirming the predicted intensity calculated using magnetic form factor for the incommensurate phase. Hence we decided to perform neutron polarimetry on the ($0 \text{ qk} \text{ 1}$) reflection for LuMnO₃ film and ultimately refine the magnetic structure combining these results. Similar measurements were performed on TbMnO₃ film but due to possible inhomogeneity the magnetic signal was very weak and we failed to measure unambiguously any magnetic reflection.

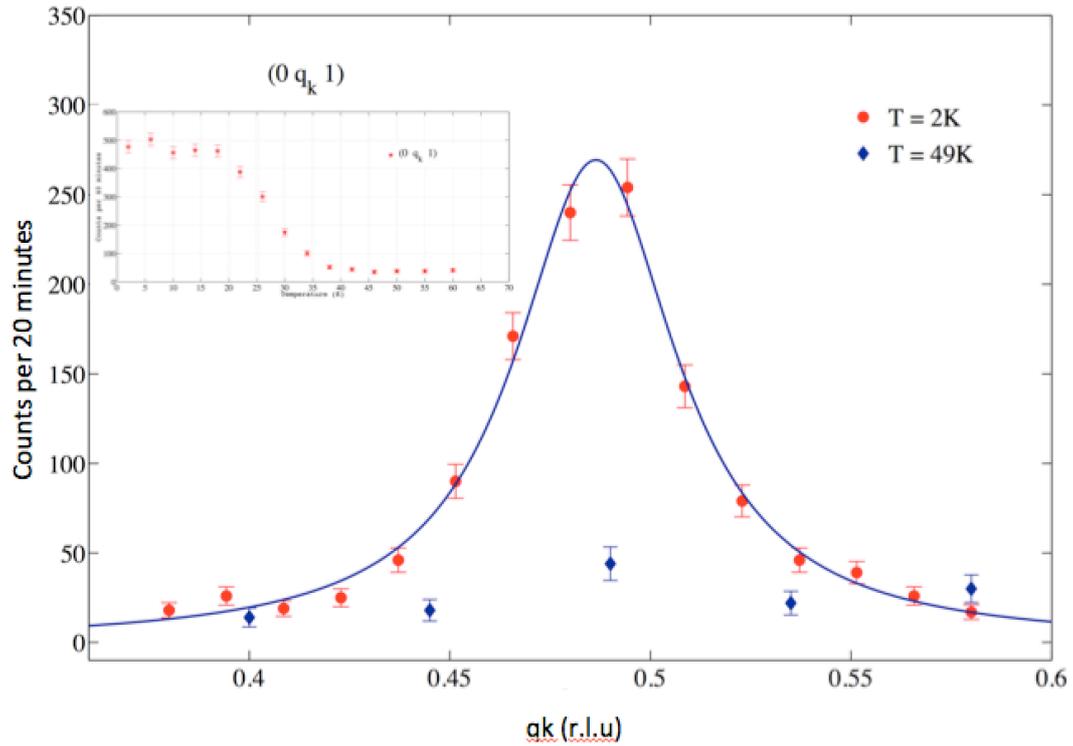


Fig.1: Superlattice Bragg peak $(0\ q_k\ 1)$ at 2K and 49K on LuMnO_3 thin film.
 Inset : Measured intensity at peak position ($q_k \sim 0.485$) as a function of temperature.

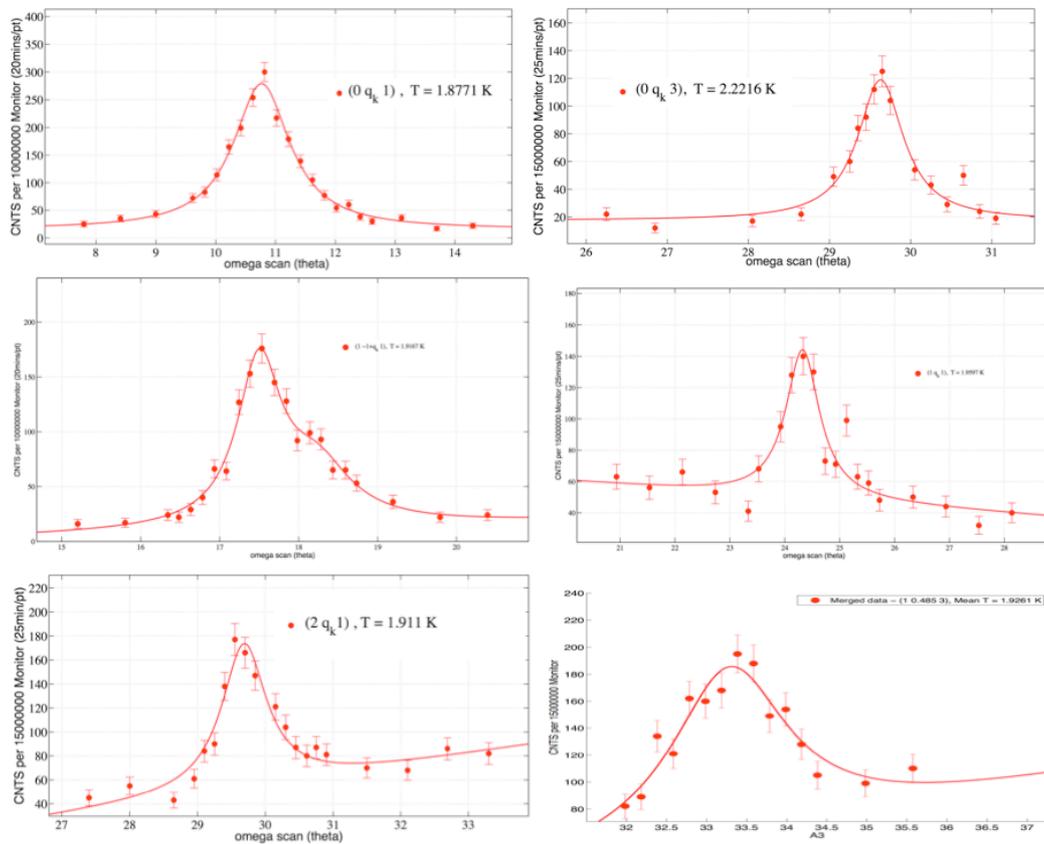


Fig.2: Rocking scans of magnetic reflections measured on LuMnO_3 thin films at 2K.

Next we performed spherical neutron polarimetry on LuMnO₃ film with D3. To overcome the challenge of accessing different hkl scattering planes we used Cryocradle set up on D3. The Cryocradle set up constituted of a four-circle cradle mounted within the CryoPAD, which provided the unique facility to align different magnetic reflections in the horizontal scattering plane.

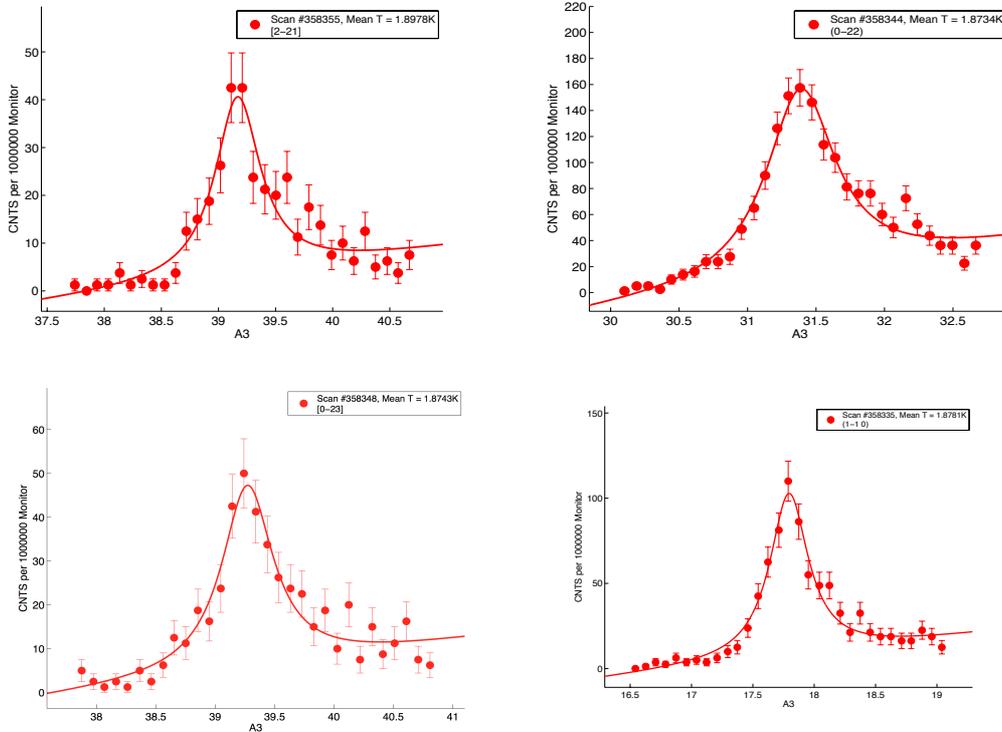


Fig.3: Few nuclear reflections measured using polarized neutrons on LuMnO₃ thin film using D3 with Cryocradle set up. Using the Cryocradle we were able to measure nuclear reflections of the [110] oriented LuMnO₃ film and refine the UB matrix [Fig.3]. Using, the refined UB matrix we managed to align the (0k0)-(00l) scattering plane and measurement of the (0 qk 1) magnetic reflection was attempted using polarized neutrons. Unfortunately, the signal-to-noise ratio in this measurement was very small which limited our attempt to measure any magnetic response. Ideal step forward will be to improve the sample quality with larger homogenous magnetic volume and the possible use of analysers to increase the signal-to-noise ratio. This may give us a better chance to resolve the incommensurate magnetic phase using polarized neutrons.

[1] J.S. White et al. Phys. Rev. Lett. 111, 037201 (2013).
 [2] H. Okamoto et al. Solid State Commun. 146, 152 (2008).
 [3] M.Kenzelmann et al. Phys. Rev. Lett. **95**, 087206 (2005).
 [4] S.Mukherjee et al. (in preparation).

Proposal-number: 5-54-153

Instrument: D10 and D3

Date of experiment: 05 - 11.11.2012

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