Experimental report

30/09/2016

Proposal: 5-54-212 **Council:** 4/2016

Title: To study the incommensurate magnetic phase in strained (010) orientedo-LuMnO3 thin films using spherical neutron polarimetry.

Research area: Materials

This proposal is a new proposal

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Samples: LuMnO3

Instrument	Requested days	Allocated days	From	To
IN20 CPA	9	16	24/08/2016	02/09/2016
			03/10/2016	10/10/2016
IN3	0	1	02/11/2016	03/11/2016

Abstract:

We want to determine the magnetic structure of multiferroic and strained orthorhombic (o-) LuMnO3 thin films deposited on (010) oriented o-YAlO3 substrates. Recent neutron diffraction experiments on the triple axis spectrometer RITA-II, SINQ (PSI), suggests the presence of an incommensurate (IC) magnetic state at (0 qk 1) with qk~ 0.49 (r.l.u). This IC-phase persists up to TN ~38K. We will employ polarised neutron analysis using the CryoPAD set up on IN20 to resolve the intrinsic magnetic structure and determine the magnetic domain population. The sample to be measured is a 100 nm thick (010) oriented o-LuMnO3 film.

To study the incommensurate magnetic phase in strained (010) oriented o-$LuMnO_3$ thin films using spherical neutron polarimetry. (**Report on experiment: 5-54-212**)

In this experiment, a 100 nm $LuMnO_3$ thin film deposited on (010) oriented $YAlO_3$ substrate is used to resolve the symmetry of magnetic order using neutron diffraction with polarised neutron and performing full 3-dimesional polarization analysis at IN20, ILL (France) with CryoPAD setup. The measurements were conducted on the magnetic reflection (0 qk 1) at two different temperatures T1 = 1.5 K and T2 = 32.5 K. These temperatures were specifically selected due to the observed magnetic transitions to incommensurate magnetic phase followed by a mixed state with both commensurate and incommensurate order coexisting below 37 K and 30 K, respectively at RITA-II (SINQ) PSI (Fig. 1). In the incommensurate phase 30 K < T < 37 K the modulation wave-vector varied in the range qk = 0.478-0.482 (r.l.u). And below T = 30 K two clear peaks at qk ~ 0.483 and at a commensurate position with qk ~ 0.5 was observed confirming the existence of the mixed state. The Mn^{3+} ion moments were observed to be lying in the *bc*-plane with the dominant component along the *b*-axis and a small canting towards the *c*-axis. In fact it is predicted that moment components along a-axis is also present but never proved experimentally. So, in this experiment one of major objective was to study the existence of the *a*-axis canting of moments out of the *bc*-plane. Hence, confirm the details of the spin structure in the commensurate phase. The other open question is if the pure incommensurate phase has the same component.

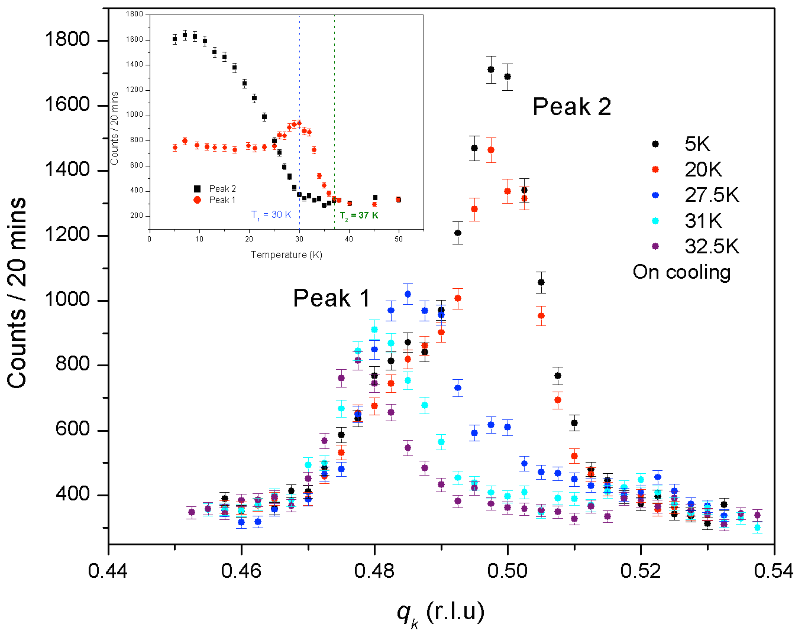

Fig. 1: The (0 qk 1) reflection measured at RITA-II (SINQ, PSI) at different temperatures. (Inset) Showing a temperature dependence of the peak amplitude.

To understand better the symmetry of the spin structure of the magnetic order both the incommensurate and commensurate phase we employed the polarised neutron studies at IN20 with the CryoPAD set up. To access the (0 qk 1) reflection the (0 K 0) – (0 0 L) scattering plane of sample was aligned first at IN3 using the nuclear reflections (004), (040),(022) and (023). Next the sample was mounted at IN20 and the cooling was performed using Orange cryostat. The orientation geometry of the polarization axes in the laboratory frame was fixed such that the scattering vector Q ǁ Px (polarization along x-axis), Q ⊥ Py (in 0 K L plane) and Pz ⊥ (0 K L) plane i.e here it is the *bc*-plane of the sample crystallographic geometry. To estimate the incident and final polarization efficiency we measured the nuclear reflection (0 0 -4) with different combinations of incident and scattered polarised neutrons. The sample was cooled slowly below 60 K to ensure gradually growth of the magnetic domains since the phase transition from paramagnetic to the incommensurate antiferromagnetic phase is a first-order phase transition confirmed by the hysteresis of the (0 qk 1) peak intensity observed.

We measured the entire magnetic reflection (0 qk 1) with the following orientations of the neutron beam polarizations: $S_{x,-x}$, $S_{-x,x}$, $S_{x,x}$, $S_{y,-y}$, $S_{y,y}$, $S_{z,-z}$, $S_{z,-z}$, $S_{y,-z}$, $S_{y,z}$, $S_{z,-y}$ and $S_{z,y}$, where the first and second subscripts are the incident and scattered polarization axis respectively. The negative sign refers to the spin flip measurements. With this convention we measured the scattered intensities ($S_{i,j}$) to extract the peak amplitudes (Fig. 3). From these peak amplitudes we estimated the polarization matrix elements at different peak positions in the pure incommensurate phase at T2 = 32.5 K and in the mixed state at T1 =1.5 K.

At, T1 = 1.5 K we observed that consistent with the non-polarised neutron diffraction results the signature of the mixed state was present and clear two peak feature at qk ~ 0.477 and qk ~ 0.5 was present. We therefore computed the polarization elements respectively at these peak positions. To resolve the contribution due to the overlap of the peaks we measured the entire reflection and fitted the (0 qk 1) spectra using double-Gaussian function to obtain the peak amplitudes. The polarization matrices at T1 =1.5 K are given in Tab. 1.

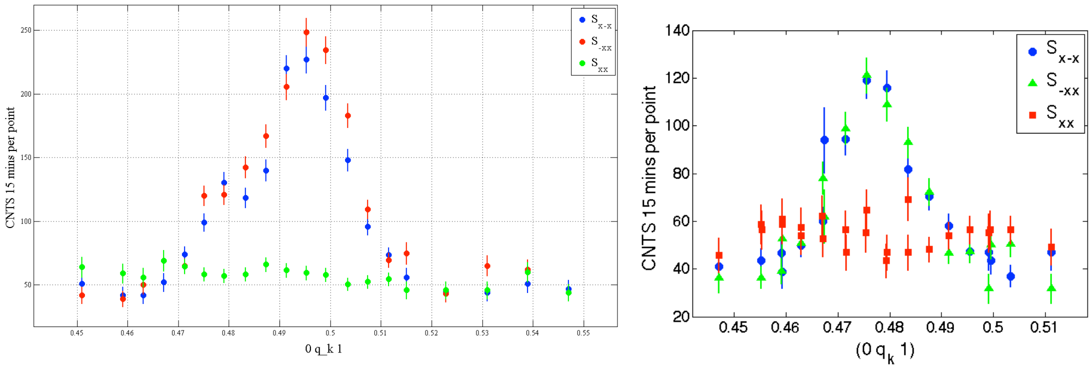

Fig. 3: Shown, here the (0 qk 1) reflections measured at T1 = 1.5 K (left image) and T2 = 32.5 K (right image) with polarization along x-axis for all the spin flip (x ⟶ -x) and non-spin flip (x ⟶ x) channels on IN20 (ILL, Grenoble).

	Pi||x	Pi||y	Pi||z	Pi||x	Pi||y	Pi||z
Phase		IC (T = 1.5 K) (qk ~0.477)			CM (T = 1.5 K) (qk ~0.5)	
Pf||x	-1	0	0	-1	0	0
Pf||y	-0.028(0.077)	0.8717(0.13)	-0.0794(0.06)	-0.0726(0.03)	0.8639(0.04)	-0.0212(0.02)
Pf||z	-0.028(0.077)	-0.0794(0.06)	-0.8717(0.13)	-0.0726(0.03)	-0.0212(0.02)	-0.8639(0.04)

Table 1: Table with elements described by incident polarization (Pi) and scattered polarization (Pf) axis.

In the table the elements Pyy and Pzz are solely sensitive to the a-axis canting of the moments out of the *bc*-plane. So, in order to verify the proposed spin model with canting of moments towards a-axis in the E-type commensurate phase of $LuMnO_3$ we estimated values of these elements. Comparing the measured elements for the mixed phase at qk ~ 0.477 and qk ~ 0.5 with the calculated values for a spin structure with moments aligned in the *bc*-plane and small canting towards a-axis we obtained that in the mixed state we obtained a finite canting of the moments of around 10 degrees out of the *bc*-plane towards a-axis. Interestingly, using this information we can also compute the domain population fraction in the commensurate E-type phase which stabilizes into two identical domains E1 and E2 defined by the change in sublattice magnetization along the crystallographic axes. So, the terms Pxy, Pxz are sensitive to any imbalance of E1/E2 domain population. Therefore, comparing the measured Pxy, Pxz with calculated values for E-type with moments canted towards a-axis by 10 degrees we obtained a domain fraction ratio of 3:2 for E1:E2 domains. This indicated that under strain we managed to tune the magnetic domain population.

In pure incommensurate phase the canting angle decreased down to 4 degrees.