Experimental report

25/01/2022

Proposal:	**5-54-371**	**Council:**	4/2021
Title:	Neutron diffraction in epitaxial antiferromagnetic oxide films for application in spintronics : effect of the thickness of the magnetic laye		
Research area:	Physics		

This proposal is a new proposal

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Samples: SrTiO3 ((LaMnO3)3 (BaTiO3)2)14

Instrument	Requested days	Allocated days	From	To
D10	10	8	31/08/2021	03/09/2021
			24/09/2021	29/09/2021

Abstract:

The search of new antiferromagnetic materials for spintronics is an important issue of this decade. The strains induced by the substrate control the orientation of the orbitals during the film deposition and control the nature and the orientation of the spins in the ordered phase. This control of the orbital ordering allows to fabricate artificial structures with create antiferromagnetic ordering along the c axis (perpendicular to the substrate) of ferromagnetic monolayers with a quite high critical temperature. We propose here to study the influence of the thickness of the magnetic layer of the properties of the magnetic film. The experiment will be to measure one sample on D10 the antiferromagnetic structure at low temperatures (30K) were the magnetic structure is well established. For one sample and based on our previous experience, we ask for 10 days. We need a four circles set up with a displex cryostat and the triple axis option to reduce the background. This experiment will be part of PhD thesis of the PhD program of ILL.

Experiment - August 2021 (5-54-371) – D10

During the 3 days of beamtime in August 2021, we measured the 3-2 sample i.e. $(La_{2/3}Sr_{1/3}MnO_3)_3(BaTiO_3)_2$ repeated 29 times, deposited on a lattice matched $SrTiO_3$ (001)-oriented substrate. The sample size is 5x5 mm^2 with a total thickness of typically 60 nm.

The goal of this experiment was to measure diffraction peaks to solve for the nuclear structure of the superlattice. The configuration 3-2 was chosen because we had already collected data for the 3-3 and 2-3 sample (experiment 5-54-341), and this experiment will help us better understand the role of LSMO and BTO layers in the material's magnetic behavior. From the theoretical calculations for this sample, we had already identified the diffraction peaks that contain the most useful information and should be measured with good statistics in this experiment. We have used 1 million monitor for most of the reflections, taking about 11 minutes to measure one point. We have focused mainly on the measurement of nuclear peaks of the film at 30 K as it is not possible to measure the paramagnetic phase, which is above 650 K.

The film was held in a cryostat on the D10 diffractometer. Incident wavelength of 1.53 Å from Cu (2 0 0) monochromator was used. The energy analyzer was used to reduce the signal coming from the substrate. Orientation of the film was done at each temperature on two independent peaks of the substrate. The lattice parameters (in Å) obtained are as follows:

Film - RT: 3.905   3.905   20.1 (from XRD),

LT: 3.896   3.896   20.08 (calculated using 0 0 10, 0 2 10 and 1 1 15 measured film peaks)

In total, 9 film reflections were measured using Q-scan. The measured peaks are shown in the following figures, in which, the red dashed line represents theoretically calculated position of the peak. Few preliminary conclusions:

- As evident from the graphs, we only found peaks for the reflections with L multiple of 5 and no intensity for other reflections: (0 0 9), (1 1 14), (0 2 9) and (1 1 16).
- Peaks (0 2 10) and (1 1 5) are very close to the substrate peaks.

For the 5 days of experiment in September 2021, we measured an $La_{2/3}Sr_{1/3}MnO_3$, 40 nm thin film deposited on Si substrate with layer of STO in the middle. From the TEM measurements, we confirm that the film maintains epitaxy with the STO layer, which has two domains of STO of thickness 8 nm and 12 nm. The STO and LSMO peaks are very close to each other in the XRD pattern but since STO is only few nm in thickness we are able to distinguish between the two types of peaks.

SQUID magnetic measurements suggest that the material has Curie temperature of about 270 K and is canted ferromagnetic in nature. There seems to be interlayer antiferromagnetic coupling along the c-direction. [1 1 0] is the easy axis of magnetization.

Theoretical intensities were calculated using Vesta software for the I 4/m geometry, best guess from the XRD measurements.

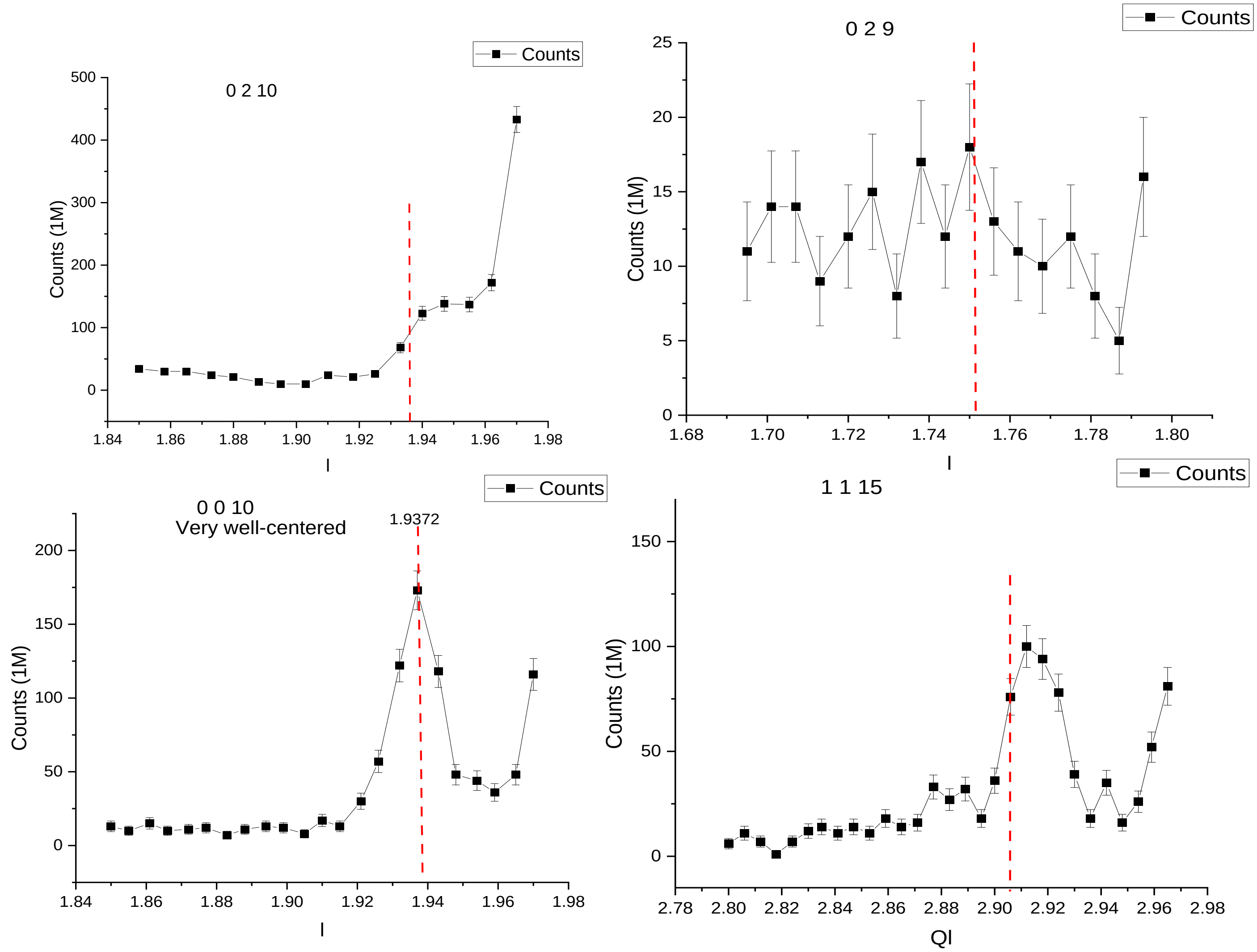

Fig. 1: Few of the measured peaks of the 3-2 sample at 30 K

Experiment:

The film was glued on the sample holder, which is screwed on the goniometer head and held in a cryostat on the D10 diffractometer. Incident wavelength of 2.36 Å from pyrolytic graphite monochromator was used. The energy analyzer was used to reduce the signal coming from the substrate. Initial orientation of the film was done at each temperature on independent peaks of the substrate Si and the film orientation matrix was calculated later. The lattice parameters (in Å) obtained are as follows:

Substrate (at RT): 5.43   5.43   5.43

Film - RT: 3.896   3.896   3.861,

LT (20 K): 3.898   3.898   3.847

The goal of this experiment was to measure nuclear and magnetic phases to solve for the magnetic structure. For the nuclear peaks, omega-2theta and Q-scans were performed at room temperature. Same set of reflections were measured at 20 K as well, in the magnetically ordered phase. Few of the reflections were also followed in temperature with Q-scans at 90, 160, 200 and 250 K. We see double peaks for some of the reflections. First analysis suggests that the second peak is from STO. Figure 2 shows few of the measured peaks at different temperatures. For the (0 0 2) peak, the STO peak on the left can be seen clearly. A clear increase in the intensity can also be observed for all the measured peaks as we go to lower temperatures, which is consistent with the SQUID magnetic measurements.

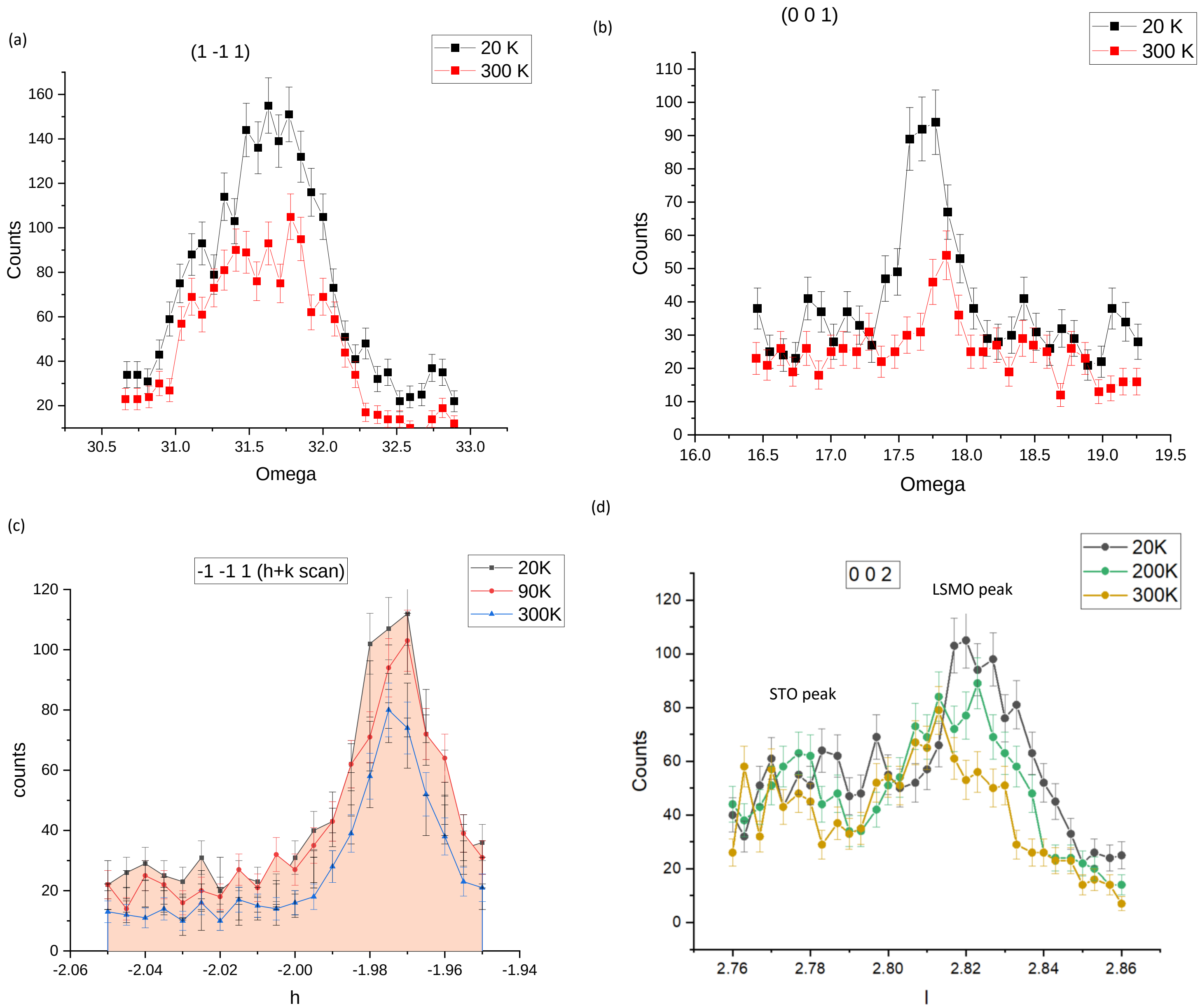

Fig. 2: Few of the measured peaks of LSMO thin film at different temperatures