ESRF	**Experiment title:** **Investigation of Surface- and Interface Structure in $K_xNa_{1-x}NbO_3$||$SrTiO_3$/$LaAlO_3$ and $CoFe_2O_4$||MgO Thin Films**	**Experiment number**: **HC-3220**
Beamline: ID03	**Date of experiment**: from: 05 Dec to: 09 Dec	**Date of report**: Feb 27th 2018
Shifts: 11	**Local contact(s)**: Raja Znaiguia	*Received at ESRF:*

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Report:

The idea behind this experiment was to understand the surface- and interface structure of piezoelectric and ferromagnetic composite thin film stacks. This is part of a project trying to investigate magnetoelectric coupling in such composite stacks, in the hope of producing high performance hybrid multiferroics that can outperform the current state-of-the-art single phase multiferroic materials. Prior to the experiment we had proof of highly oriented thin films of $NiFe_2O_4$ (as deposited by atomic layer deposition) and $K_xNa_{1-x}NbO_3$ (upon annealing after atomic layer deposition). Since the magnetoelectric coupling is highly affected by the epitaxial relationships for substrate||film and film||films in multilayer systems, it is crucial to understand the surface- and interface structures to be able de design high-performance samples.

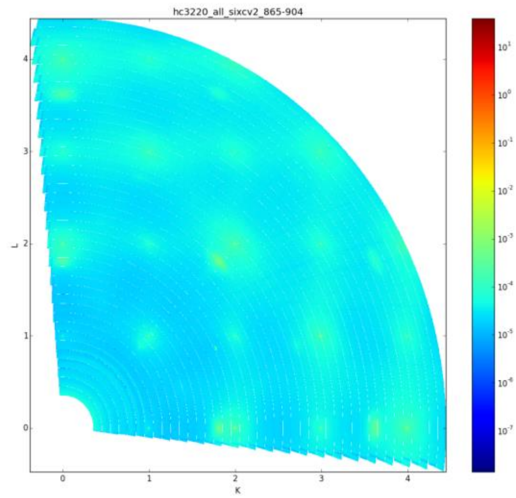
In the original proposal, we wanted to study single layer systems (ex. $NiFe_2O_4$||$LaAlO_3$ and $K_xNa_{1-x}NbO_3$||$LaAlO_3$) and multilayer systems with mixes of the ferromagnetic and ferroelectric layers. It was immediately clear that the time frame set for the experiment did not allow for all this. First of all this was the first time anyone from our scientific group used

ID03, meaning there was significant time lost in learning to use the infrastructure. Furthermore, the chemical complexity of our samples was at a level at which the beam line staff does not typically work. Last, but most important, investigating the samples proved to be more tedious than originally planned, and there were a lot of features that had to be studied for each sample that we simply could not have planned for.

As a result of this, the experiment ended up being a surface- and interface structural study of $NiFe_2O_4$||$LaAlO_3$(100), with different thin film thickness to deconvolute surface-, interface- and bulk effects. Even though the amount of samples studied became less than intended, the data obtained proved to be extremely useful:

1. The $NiFe_2O_4$ thin films proved to be highly epitaxial (in-plane and out-of-plane oriented), as deposited by atomic layer deposition at 250 ºC. This can be seen from fig 1, showing a 0.8 k l reciprocal space map of a selected sample. Obtaining high quality ferromagnetic complex oxide thin films at this low temperature is unique, and extremely uncommon.
2. Surface diffraction enabled us to understand the driving mechanism behind this epitaxy. As can be seen from fig 2, an intermediary layer between the substrate and the thin film is formed, having a lattice with parameters in between $LaAlO_3$(100) and $NiFe_2O_4$. The relative intenisty of the diffraction from this layer as a function of layer thickness suggests that this mitigating layer is a single unit cell. The lattice parameters match with that of Fe_2O_3. This may be a result of the fact that iron is pulsed first in the atomic layer deposition process. It would be very interesting to se if the same epitaxial relation is achieved upon pulsing nickel first.
3. A large set of crystal truncation rods (21 per sample) was collected to understand more about the surface- and interface structures in different crystallopgraphic directions. Initial investigation of these CTR’s suggest a reconstruction of the lattice (unsuprisingly) both at the film||substrate-interface and at the surface. This information is very important when continuing with multilayer systems, which is deposited over several runs. Modelling the reconstruction to fit with the observed data is still in progress.

All-in-all the experiment proved to be an immediate success that ended up raising more questions than what it answered (As It should!). The infrastructure proved very useful for the type of samples that are important for our project. More time to analyze the data is needed, and more experimental time is called for to understand more about the mechanisms in play. With that said, the initial results are already worked to be published as part of a paper on as deposited epitaxial films of $NiFe_2O_4$ that will be submitted medio 2018.



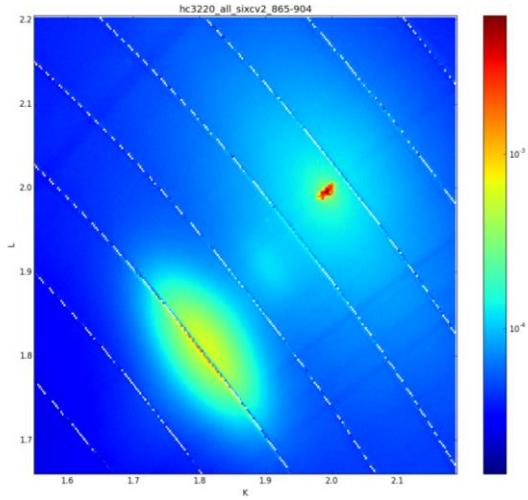


Fig 1, left. RSM (k-l-plane) of $NiFe_2O_4$ on $LaAlO_3$ at h = 0.8, showing the high in-plane ordering and epitaxial relationship of the NFO||LAO-samples. Fig 2, right. (0.8 2 2)-reflection of $LaAlO_3$ (most intense) together with the (0.8 4 4)-reflection of NFO (most broad). In between a highly defined peak corresponding to an intermediary structure that mitigates the as deposited epitaxy can be observed.