ESRF	**Experiment title:** Evolution of ferroelectric super-domain structure with temperature and electric field	**Experiment number**: HC-2912
Beamline: ID01	**Date of experiment**: from: 12 July 2017 to: 18 July 2017	**Date of report**: 26.09.2017
Shifts: 18	**Local contact(s)**: Dr. Steven Leake	*Received at ESRF:*

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Report:

Ferroelectric thin films display domain structures which depend on the electrostatic and elastic boundary conditions they are subjected to. The latter can be tailored by depositing the film epitaxially onto a substrate of appropriate lattice constants and symmetry, as well as by varying the film chemistry and thickness. Tuning these parameters changes the strain state of the film, promoting the formation of a particular domain structure. When $PbTiO_3$ (PTO) is tensile strained to approx. 0.85% by deposition on $KTaO_3$ (KTO), its calculated phase diagram predicts a mixed domain population, having the polarisation axis either in-plane or out-of-plane with respect to the main crystallographic axes of PTO.

We prepared PTO//KTO samples by RF magnetron sputtering and performed laboratory X-ray diffraction as well as Piezoresponse Force Microscopy (PFM) to investigate their domain structure. We found that a hierarchal arrangement is formed, whereby individual domains organise in distinct bundles, or "superdomains". Within each superdomain the domains are periodic in a specific direction. Each superdomain thus gives a well-defined signal in reciprocal space. Domain periods are of 10-30nm, while superdomain characteristic sizes are 200-600nm, both increasing with film thickness. Of interest for this experiment was to investigate what would happen to the morphology and arrangement of the superdomains under the application of temperature and electric field.

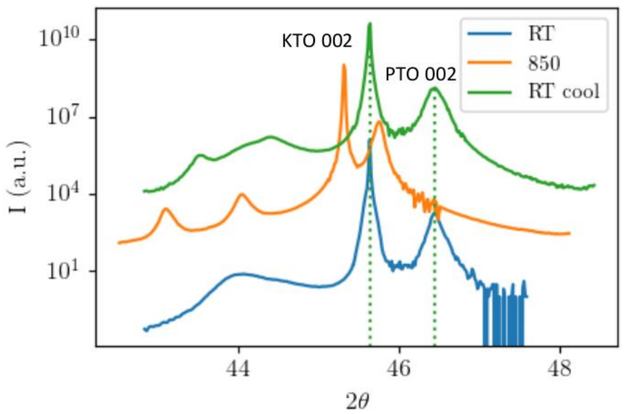

Figure 1 - Intensity of the 002 peak of a 200nm PTO//KTO film integrated in the axial direction; at room temperature (RT), 850C, and back to room temperature after having heated ('RT cool').

The ID01 beamline offers the possibility of performing so-called K-maps, i.e. measurements where a nano-beam is scanned across the sample while intensity of a Bragg reflection is recorded. Originally we had planned to collect K-maps on films of different thicknesses at several temperatures, and up to the ferroelectric transition temperature. Unfortunately the ID01 furnace stage had multiple problems in interfacing with the beamline; few shifts were spent trying to fix these issues which eventually were solved. In the limited time remaining, only the 002 Crystal Truncation Rod (CTR) from a 200nm PTO//KTO film was measured as a function of temperature, and no K-maps could be collected. Despite the ferroelectric transition temperature of PTO//KTO films at that thickness is expected to be around 600°C, no transition was observed up to 850°C (Fig. 1 - the onset of the transition is marked by the merging of the film and substrate peaks). In addition, two low-angle peaks appeared and grew in intensity with temperature, corresponding to the formation of two new (potentially parasitic) phases appearing. The temperature could not be increased above 850°C due to the limitations of the furnace. Further work is needed to reach higher temperatures and observe the ferroelectric transition, as well as collecting the K-maps on a range of samples as originally intended.

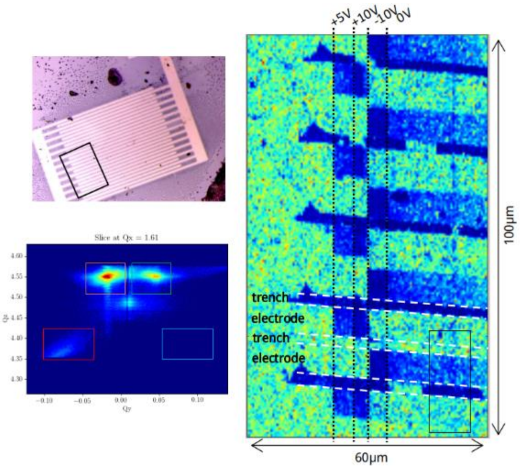

Figure 2 – Top left, optical microscopy image of the surface of the film, displaying the interdigitated electrode pattern. Bottom left, a detector image collected at the 103 PTO//KTO reflection displaying the regions of interest used for K-mapping (coloured rectangles). Right, a K-map showing the effect of voltage switching on the in-plane $a_1$domain population.

A 100nm PTO//KTO sample with interdigitated electrodes was then measured (see Fig. 2, top left). These are placed so that when voltage is applied, an in-plane field exists within the film trenches in between each electrode. The in-plane and out-of-plane superdomains give distinct signals in reciprocal space around the 103 PTO//KTO reflection, a detector image of which is shown in Fig. 2 (bottom, left). The top and bottom satellites identified by coloured rectangles in the detector image correspond to the in-plane and out-of plane superdomains, respectively. The 60x100nm focused beam is scanned across the sample while the intensity in the coloured squares is recorded, i.e. K-maps are collected. The right side of Fig. 2 shows a K-map performed in the area corresponding to the black square in the top left picture of Fig. 2. The signal tracked in this case is that of the orange rectangle, i.e. the left in-plane superdomain peak (a_1). Application of positive and negative voltage stimulates the formation of out-of-plane domains just under the electrodes where the field is in such direction, which is seen as a reduction of intensity in the orange rectangle -- hence a low intensity area in the K-map.

Figure 3 shows this in more detail. The K-maps corresponding to the four squares in the detector image of Fig. 2 are shown, the probed area roughly corresponding to the black rectangle in the K-map of Fig. 2. The individual superdomains are resolved at this scale. Switching the voltage polarity results in: i) partial switching of the a_1 and a_2 in-plane superdomain variant present under the electrodes and in the trenches respectively; ii) the formation of out-of-plane a_1c and a_2c superdomains beneath the electrode with negative or positive polarity, respectively. Additional data analysis (ongoing) as well as the repetition of the experiment on a range on sample thicknesses are required in order to understand the details of this process.

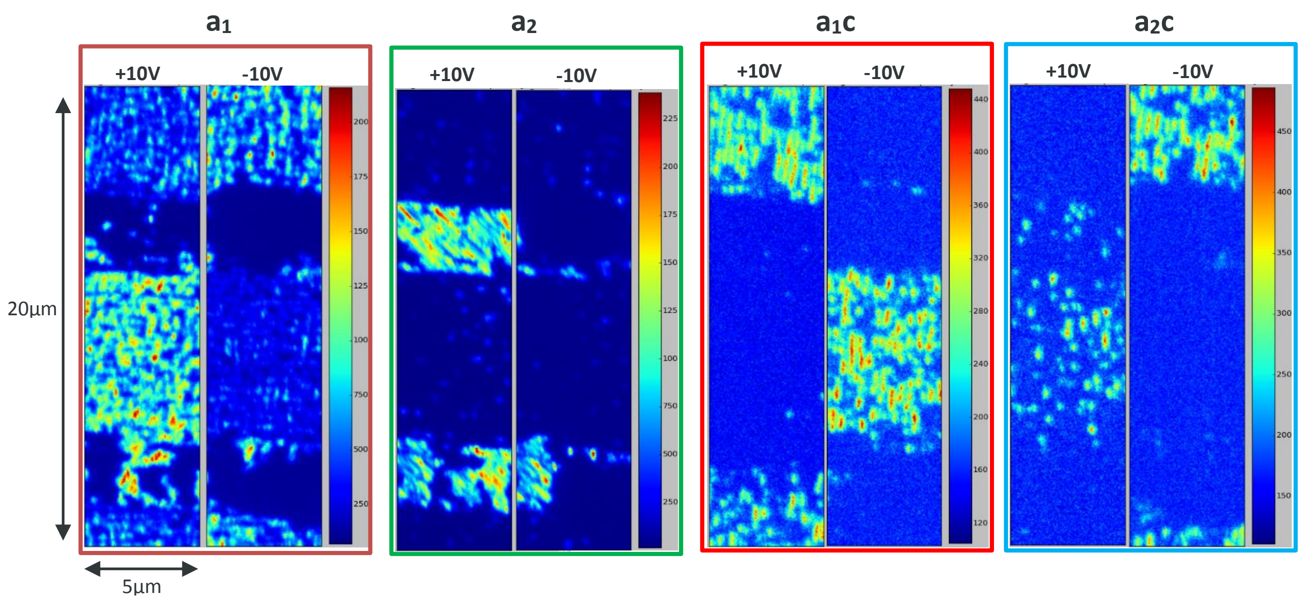

Figure 3 – K-maps of the region shown in Fig. 2 as black rectangle on the bottom right. Each couple of K-maps tracks a signal due to a specific set of superdomains. The effect of voltage switching on each kind of superdomain is visible.