ESRF	**Experiment title:** Ferroelectric domain melting in epitaxial oxide superlattices	**Experiment number**: HC-2615
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Names and affiliations of applicants (* indicates experimentalists):

S. Leake* (ESRF, Grenoble)

E. Zatterin (ESRF, Grenoble / UCL, United Kingdom)

P. Zubko, (UCL, United Kingdom)

M. Hadjimichael (UCL, United Kingdom)

Report:

Ferroelectric superlattices are an extremely rich group of artificially engineered heterostructures, which have recently gained a lot of attention, due to the possibility of engineering materials with enhanced functional properties, and because of the novel emergent phenomena observed in these materials. The properties of these superlattices are controlled by ferroelectric domains which have a well-defined periodicity, making them perfect for X-ray diffraction (XRD) studies. We have recently discovered that during the ferroelectric to paraelectric phase transition, the diffracted intensity due to the periodic domain satellites disappears ~50K below the anomaly in the tetragonality of the film, the conventional mark of the transition temperature. First-principles calculations by our collaborators predict that this temperature window is characterised by dynamic fluctuations of the ferroelectric domain structure.

To get more insight into what happens to the ferroelectric domains near the phase transition we first used a full beam to map the intensity of the domain satellites in 3D reciprocal space as a function of temperature. We discovered a transition during which the domain walls change their orientation, from walls aligned along the <100> crystallographic axes, to walls oriented along <110> at higher temperatures (see Figure 1). Through a detailed analysis of the temperature dependence of the intensity due to each domain orientation, we conclude that this change in intensity is not due to the reduction of polarisation, but an actual gradual increase in the volume fraction of domains oriented along <110>. This is further confirmed by measuring the in-plane coherence length of the <110> satellites. We observe that it increases at higher temperature, marking the onset of order for this orientation.

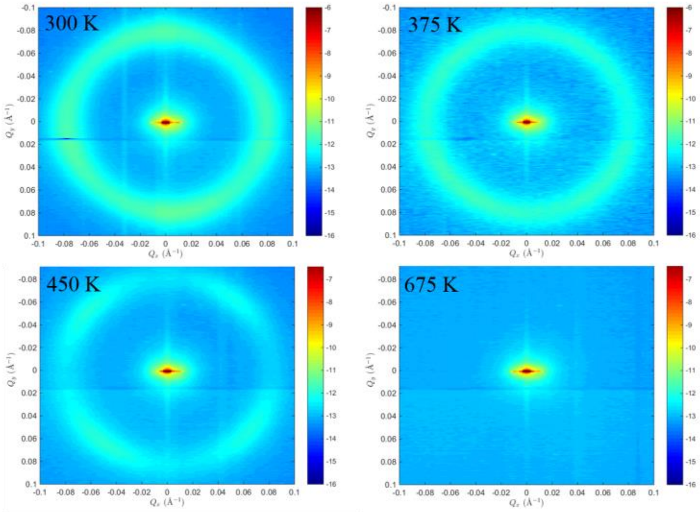

Figure 1. In-plane reciprocal space maps for a superlattice with domain period ~ 5.0 nm as a function of temperature, showing a change in dominant domain orientation at 450K.

After mapping the domain intensity as a function of temperature, we went to coherent beam mode to look at the speckle patterns around the domain satellites (Figure 2). By looking at the autocorrelation functions of these patterns as a function of time we can extract some characteristic timescales for the fluctuations. The results of the preliminary analysis are shown in Figure 2. A complex temperature dependence of the extracted timescales is observed, and further work is required to deconvolute the effect of the stability of the experimental setup from that of fluctuations.

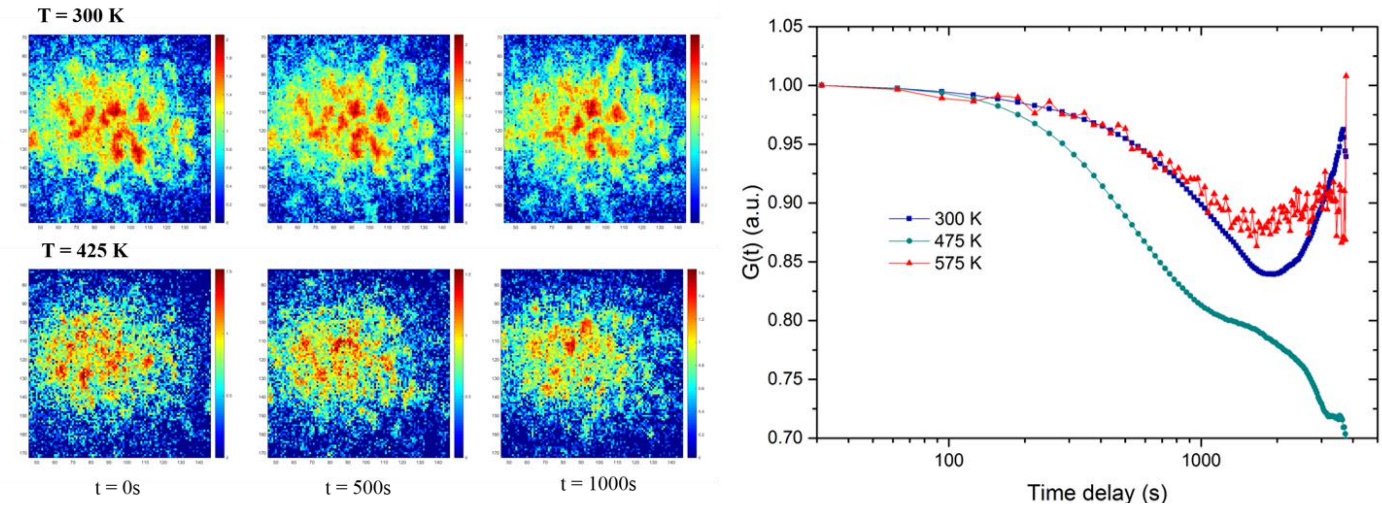

Figure 2. Speckle patterns (left) around the domain peak of the superlattice at two different temperatures, as a function of time. We see that at 425 K the speckle patterns change significantly with time. Normalised autocorrelation function (right) of the images as a function of time delay, showing a characteristic decay time at 300 K, and perhaps two timescales at 475 K.

To understand the domain patterns further, we investigated their real-space distributions using a nanofocused beam and k-mapping. We mapped the intensity of the domain satellite peaks for different domain orientations to see if there are any preferential alignments and if they correlate with any features of the sample. By mapping the domain orientation intensities around a defect, we find that the domain walls align along characteristic features in topography (Figure 3), features which were later confirmed with atomic force microscopy. This is in contrast to what happens in the rest of the film, where the domain wall alignment is random.

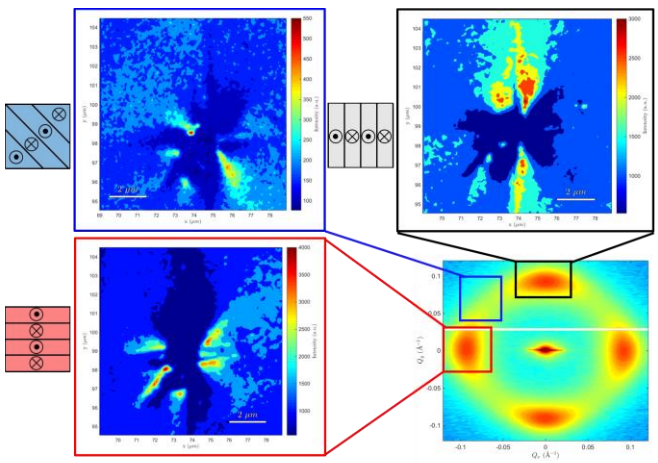

Figure 3. Real-space maps of the variations in intensity of satellites due to different domain orientations around a dislocation. The schematics show the orientation of the domain walls for the relevant intensity which is mapped.