ESRF	**Experiment title:** Investigation of individual domains and domain walls in ferroelectric $K_{0.75}Na_{0.25}NbO_3$ films grown on (110) $TbScO_3$	**Experiment number:** **MA-2851**
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Report:

In experiment MA-2851 we have investigated the ferroelectric domain structure of two $K_{0.75}Na_{0.25}NbO_3$ thin films (19 nm and 23 nm thickness) by using a micro-focused x-ray beam at ID01 beamline. Our experiment has been quite successful.

In our experiment a 300 µm diameter gold Fresnel zone plate (FZP) with an outermost zone width of 70 nm has been employed at 8 keV x-ray energy. The resulting angular divergences in the 90 nm (v) x 150 nm (h) spot at the sample were about 2.2 mrad (v) x 2.2 mrad (h). The samples have been mounted on an x-y-z scanning piezoelectric stage with a resolution of 2 nm in all three directions. A fast readout two-dimensional detector (MAXIPIX) consisting of 516 × 516 pixels with 55 µm pixel size was placed at a distance of 986 mm from the sample. The experimental setup allows for (i) three-dimensional reciprocal space mapping at a selected fixed position in real space (with a resolution in reciprocal space which is limited by the divergence of the incident beam), and (ii) scanning x-ray micro-diffraction at a selected fixed position in reciprocal space with a spatial resolution which is comparable to the focal size of the primary x-ray beam.

Owing to the strong anisotropy of misfit strains in the $K_{0.75}Na_{0.25}NbO_3$ epitaxial layers (the epitaxial strain is highly compressive in one in-plane direction and weakly tensile in the corresponding orthogonal direction) a one dimensional, highly periodic ferroelectric domain pattern is formed which extends over several tens of micrometers. The domains consist of the inclined monoclinic M_A phase [1] which is associated with both a strong vertical and horizontal electrical polarization component. Beside the prominent one dimensional domain pattern which is aligned along the lateral $[-112]_{TSO}$ direction of the underlying $TbScO_3$ substrate – and which we refer to as the '0° variant' – a structural variant of a 90° rotated M_A domain pattern is also observed, however, with significantly lower probability (see schematic view of domains in Fig.1a). In this 90° variant the domain pattern is aligned along the lateral $[1-12]_{TSO}$ direction. The lateral piezoresponse force micrograph (LPFM) of the 23 nm sample (Fig.1a) shows both variants. One specific aim of our experiment was the independent structural investigation of both domain variants by using a micro-focused x-ray beam.

A selected detector frame (Fig.1c) recorded in the vicinity of the $(-113)_{pc}$ reciprocal lattice vector of the 23 nm $K_{0.75}Na_{0.25}NbO_3$ film exhibits strong satellite peaks P1 and P2 which are caused by the periodicity of the domain pattern aligned along the $[-112]_{TSO}$ direction (0° domain variant). Furthermore, we could also resolve a needle shaped area A on the sample (Fig.1b) where the satellite peaks P1 and P2 vanish (Fig.1d). The $(113)_{pc}$ reflection (not shown due to restricted place) which is sensitive to domains aligned along $[1-12]_{TSO}$

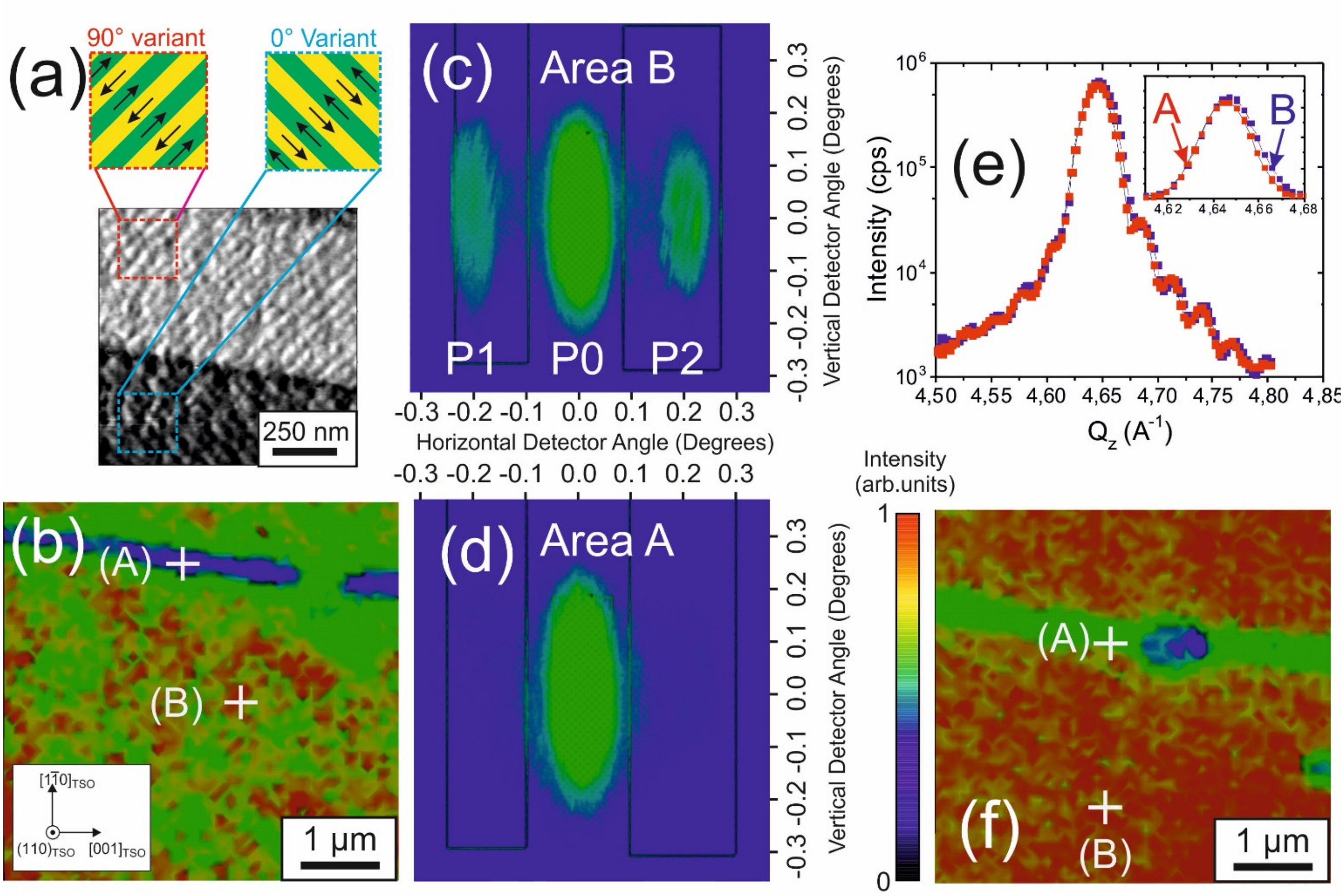

Fig.1: (a) Lateral PFM image of 23 nm sample showing two variants of stripe domains differing by 90° in stripe orientation. **(b)** Scanning x-ray micrograph of the integrated intensity of satellite peak P1 in the vicinity of $(-113)_{pc}$. Corresponding detector frames recorded in **(c)** region B and **(d)** region A. **(e)** Integrated intensity of the $(113)_{pc}$ CTR (Peak P0) as a function of the vertical scattering vector Q_z. The observed peak shift proves different vertical lattice parameters in **(f)** regions A (90° domain variant) and B (0° domain

exhibits a complementary behavior: Here, P1 and P2 are strong in area A and are vanishing in area B. Therefore, area A can be assigned to the 90° domain variant, and can be clearly distinguished from the 0° domain variant.

Surprisingly, also the central peak P0 – which is given by the intersection of the sample crystal truncation rod (CTR) with the 2D detector – shows contrast in the images (Fig.1f). However, the observed effect is much weaker than for the satellite peaks P1 and P2. While the contrast in P1 and P2 maps (Fig.1b) is produced by the different domain alignments of the 0° and 90° variants, the contrast of the central CTR is presumably caused by different vertical lattice parameters in the 0° and 90° variants. This is demonstrated in Fig.1f for the $(113)_{pc}$ Bragg reflection where again areas A and B can be resolved. The maximum of the CTR of the 90° domain variant appears at a slightly smaller Q_z-value than that of the 0° variant (Fig.1e) revealing a slightly enhanced vertical lattice parameter of about $(\Delta d/d)_{\perp} = (6 \pm 1)\cdot 10^{-4}$ for the 90° domain variant. This effect has been observed (i) for a variety of different Bragg reflections and (ii) for both samples under investigation. We interpret the observed vertical lattice parameter difference by the elastic anisotropy of the $K_{0.75}Na_{0.25}NbO_3$ film. Corresponding calculations using linear elasticity theory are in agreement with the experimental findings.

In summary, we have successfully demonstrated the independent investigation of different domain variants in ferroelectric $K_{0.75}Na_{0.25}NbO_3$ epitaxial films by using a micro-focused x-ray beam. As a first interesting result we have shown that the vertical lattice parameters in these variants are slightly different (a manuscript is currently in preparation). However, a single pair of domains (at samples with increased film thickness where the domain sizes are larger) could not be investigated owing to restricted time. Extended studies on samples with varying film thicknesses would thus be highly desirable.

References

[1] J. Schwarzkopf, D. Braun, M. Hanke, A. Kwasniewski, J. Sellmann, and M. Schmidbauer, J. Appl. Cryst. **49**, 375 (2016).