ESRF	**Experiment title:** Investigation of ferroelectric switching mechanism in ultrathin barium titanate films with Resonant Elastic X-ray Scattering	**Experiment number:** MA 1887
Beamline: ID01	**Date of experiment:** from: 25 Okt 2013 to: 29 Okt 2013	**Date of report:** 15.02.2014
Shifts: 12	**Local contact(s):** Dr. J.Hilhorst	*Received at ESRF:* *16.03.2017*

Names and affiliations of applicants (* indicates experimentalists):

* Dr. A.V.Zenkevich, NRC Kurchatov Institute, Moscow, Russia

* Dr. D.V.Novikov, DESY, Hamburg, Germany

* C.Richter, TU Freiberg, Freiberg, Germany

Report: The aim of the project was investigating the switching effects in ultrathin ferroelectric $BaTiO_3$ (BTO) films, concentrating on the polarization and relaxation dynamics under external electrical field. In particular, the experiment was focused on the mechanism of homogeneous polarization switching. This mechanism is not fully understood so far and must function without domain formation, in contrast to the well known Kolmogorov-Avrami-Ishibashi (KAI) model, which is typical for thicker (> 10 nm) films and bulk crystals.

The experimental setup relied on the cutting-edge hardware environment available at the ID01 beamline. The measurement procedure combined micrometer large focused beams with resonant Bragg diffraction measurements at specially chosen X-ray energies, at which the scattered beam intensity is most sensitive to the polarization of the BTO lattice. To implement polarization switching, the samples had a specially prepared patterned structure of top electrodes, while the external voltage was applied through the cantilever of an atomic force microscope that was fixed to a quartz tuning fork (Fig.1)

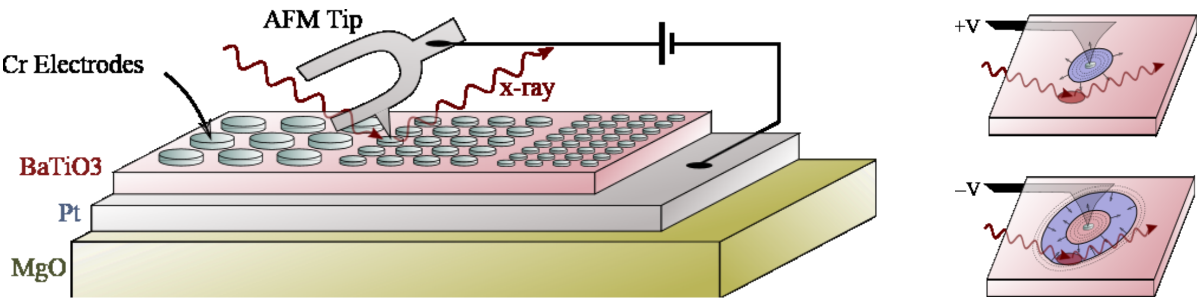

Figure 1: Sketch of the sample with patterned electrodes and the in-situ AFM (left) which was used at beamline ID01 for imaging of the electrodes as well as for voltage application. On the right the formation of different domain shapes as a result of alternating voltage is shown.

The main aim of the project could not be reached for reasons beyond our control: the vertical drive of the AFM appeared to be less precise than necessary for the task and perforated the samples with sub-10 nm thickness.

However, the results obtained during the calibration measurements on thicker films demonstrated that the approach can be reliably used as a non-invasive tool for measurement of ferroelectric switching in ultra-thin films: after landing the AFM tip on an electrode, we monitored the Bragg intensity of the 001 reflection at an energy of 5000 eV where a relatively large contrast of 8% for polarization reversal was expected. In parallel, a square wave-shaped, periodic, alternating voltage was applied between AFM tip and platinum buffer layer with different voltages in the range of 1…4 V which corresponds to a field of 33…133 MV/m for the 30 nm film thickness. The intensity was accumulated over a large number of electric field cycles and the average is shown for different voltages in Fig. 2.

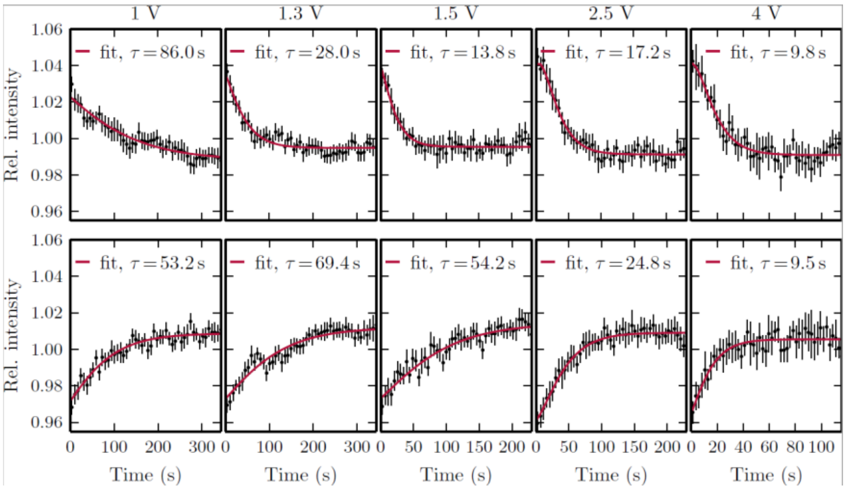

Figure 2: Intensities measured during a large number of alternating, square wave-shaped electric field cycles averaged for intervals of constant voltages.

The observed curves can be directly attributed the effect of domain wall propagation, as described in the KAI theory. The overlap O of a circularly growing domain and a elliptic beam footprint with normally distributed intensity has been modeled numerically and, later, approximated with the function $O = 1- [(t-t0)/\tau +1]*\exp(-(t-t_0)/ \tau)$, where $t > t_0$ is the time t_0 is the time of voltage reversal and τ is the characteristic switching time. The intensity is then obtained via $I(t) = I(0)+[I(1) - I(0)]\cdot O(t)$. The switching times are shown as a function of absolute applied voltage and classified as slow and fast component in Fig. 3. The asymmetric behavior can be understood since one polarization state is more stable depending on the initial (monodomain) state of the BTO film.

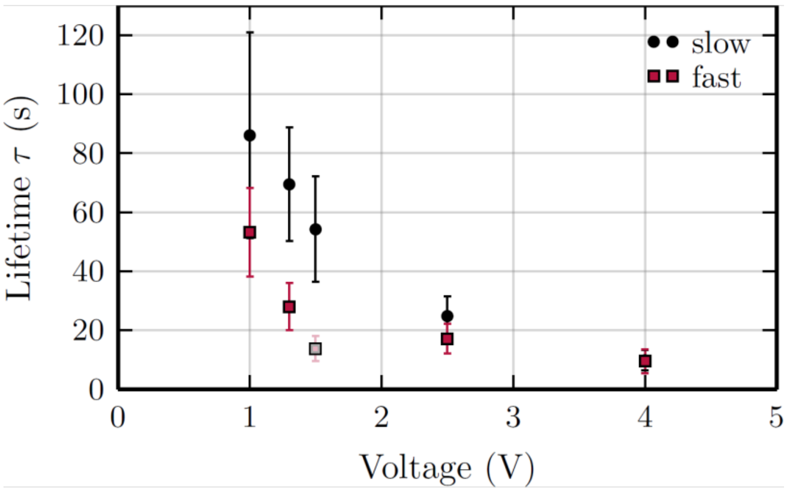

Figure 3: Characteristic decay times as obtained by fitting the averaged time series shown in Fig. 2 sorted for fast and slow direction of switching, i.e. in the direction of initial lattice polarization and against it.

Since the beam size was approximately 1.8 µm, a rough estimate for the speed of domain wall motion can be computed as 1.8 µm/4τ (the overlap grows beyond 90% after 4τ) . The values can be obtained with a much better precision in the frame of a specially designed experiment.

The approach developed in this project proves to be very effective for observation and precise characterization of lattice polarity switching in ultra-thin single crystalline ferroelectrics. Quantitative information on the domain wall propagation velocity and its dependence on film thickness and applied voltage can be used to obtain main thermodynamic parameters that control the switching dynamics in thin ferroelectric oxide films.