Characterization of Domain Wall Dynamics in $PbZr_{1-x}Ti_xO_3$ Using X-ray Photon Correlation Spectroscopy

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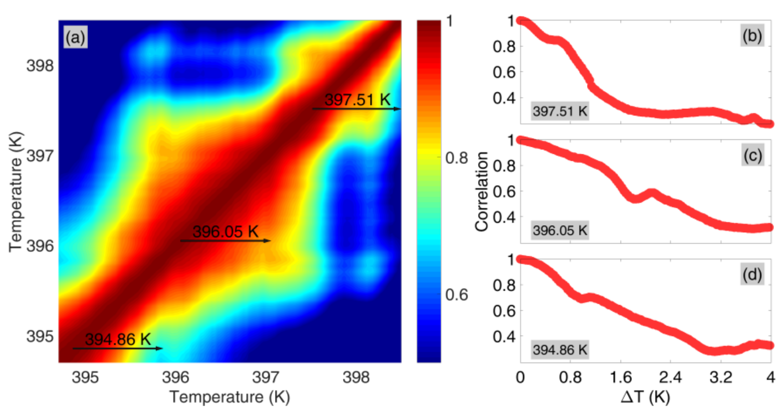
Ferroelectric domain patterns may be easily rearranged by thermal, electrical or mechanical influences. Such rearrangements greatly contribute to the remarkable pyroelectric, dielectric, piezoelectric or elastic properties of ferroic materials. Direct observation of ferroic domain patterns is often challenging: such existing techniques as optical birefringence, electron, confocal Raman or piezo-response-force microscopies do not access the mesoscopic dynamics (domain wall motion, domain nucleation and growth). Therefore, the physical laws governing domain patterns dynamics often remain unclear.

We present X-ray Photon Correlation Spectroscopy (XPCS) for the investigation of temperature-driven rearrangements of notoriously complex domain microstructures in $PbZr_{1-x}Ti_xO_3$ (PZT) close to the morphotropic phase boundary composition (x~0.46). XPCS is the recent coherent beam synchrotron diffraction technique, which (unlike the ones above) accesses the length-scale specific and the motion-specific information. It involves sequential collection of speckles - extremely detailed two-dimensional patterns of scattering of coherent synchrotron X-ray beam at a mesoscopic (typically sub-micrometer) domain patterns. Although any direct conversion of 2D speckle patterns to the 3D real-space domain patterns is hard, the correlations between different speckles can be readily calculated. It is furthermore assumed that the time depended of these correlations (correlation decay) reflect the dynamics of the real space domain patterns.

We collected the set of ~11 000 speckles around 001 Bragg reflection at the P10 beamline of the PETRAIII storage ring (Hamburg, Germany), as the PZT46 single crystal was gradually cooled down from 650 K to 350 K, passing through the phases of tetragonal (>600 K) and monoclinic (< 550 K) symmetry [1]. Figure 1(a) shows the correlation map between the speckle patterns, collected at different temperatures, Figures 1(b-d) show the decay of the correlation as the temperature interval between the speckle patterns increases. The analysis of the correlation decays shows that the temperature intervals with "stable" domain configuration (slower correlation decay) are alternated by the temperature intervals with "unstable" domain configuration (faster correlation decay).



We have furthermore developed original statistical analysis of the correlation coefficients, which shows that monoclinic domain rearrangements can be described as series of overlapping modes, where each mode represents the series of **countable** movements / jerks.

[1] A. A. Bokov, X. Long, and Z.-G. Ye Optically isotropic and monoclinic ferroelectric phases in PbZr/TiO3(PZT) single crystals near morphotropic phase boundary. Phys. Rev. B 81, 172103 (2010).