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Experiment Report Form

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Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** Ferroelectric-Ferroelastic Domain Switching and the Blocking Stress in Piezoelectric Ceramics	**Experiment number**: MA1727
Beamline: ID11	**Date of experiment**: from: 25/04/2013 to: 29/04/2013	**Date of report**: 3/9/2013
Shifts: 9	**Local contact(s)**: Andrew King	*Received at ESRF:*

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Report:

The aim of the experiment was to determine the changes in lattice strain and ferroelectric-ferroelastic domain fractions in piezoelectric ceramics under combined electrical and mechanical loading conditions. More precisely, the purpose was to establish how differently-oriented grain families respond to produce a macroscopic average strain of zero under the blocking stress conditions and hence to estimate the magnitude of the residual inter-granular stresses. The tests have been successfully conducted on two polycrystalline piezoelectric materials, a tetragonal PZT51-49 and a rhombohedral PZT55-45.

The experiments were conducted using small square-ended rod specimens with dimensions of 1 mm x 1 mm x 3 mm, machined from larger ceramic disks. XRD spectra were measured in transmission, using a CCD detector with an x-ray photon energy of 78 keV. A mechanical compressive stress was applied in-situ using the ESRF mini-stress rig. An electric field up to 4 kV/mm was also applied in-situ using a high voltage amplifier, insulated from the mechanical testing apparatus by means of alumina spacers. The test fixture was immersed in insulating silicone oil to prevent electric breakdown during the high voltage tests. User safety was assured by using a magnetic cut-out switch to the high voltage amplifier, attached to the door of the experimental hutch.

The experimental procedure was as follows: the sample was first poled, or repoled, under an electric field of 4 kV/mm and then subjected to a static electric field in the range 0.5 to 2.5 kV/mm, inducing a tensile piezoelectric strain along the electric field axis. A compressive stress was then progressively applied in the direction of the electric field so as to approximately cancel the piezoelectric strain. The stress was finally released, and a new cycle started again for another value of the electric field. By this means it was possible to construct blocking stress curves similar to those measured from macroscopic tests, but defined from a local point of view in terms of the lattice strain for grain families having specific crystallographic orientations. The variations in the applied electric field and compressive stress recorded over time for a typical experiment on a rhombohedral PZT55-45 ceramic are illustrated in Fig. 1.

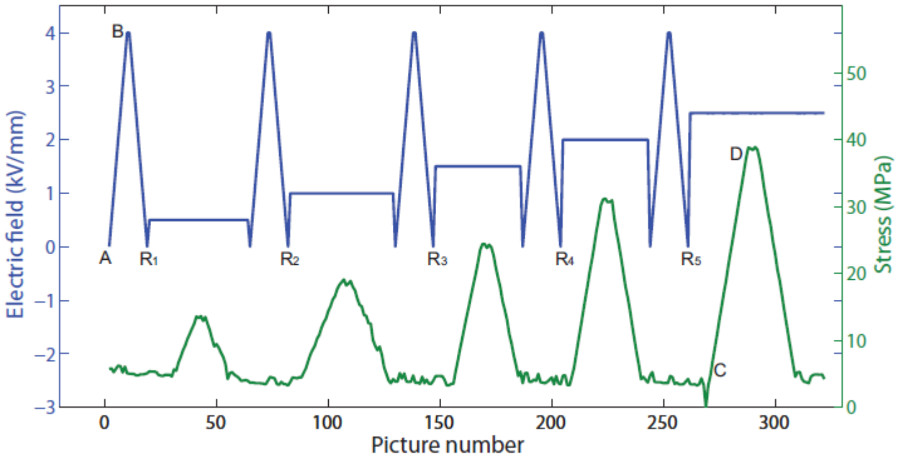

Fig. 1. Changes in applied electric field (top) and compressive stress (bottom) during the in-situ loading experiment on a rhombohedral PZT55-45 ceramic.

Selected regions of the diffraction patterns obtained during the initial electric field cycle are presented in Fig. 2. It is evident that the (200) peak exhibited a shift to lower angles for ψ = 0, indicating the development of a tensile strain along the electric field direction, and a shift to higher angles for ψ = 90°, corresponding to a compressive strain in the transverse directions. This was accompanied by an enhancement of the (111) peak intensity relative to that of the (-111) peak for ψ = 0 due to ferroelectric domain switching along the direction of the applied electric field.

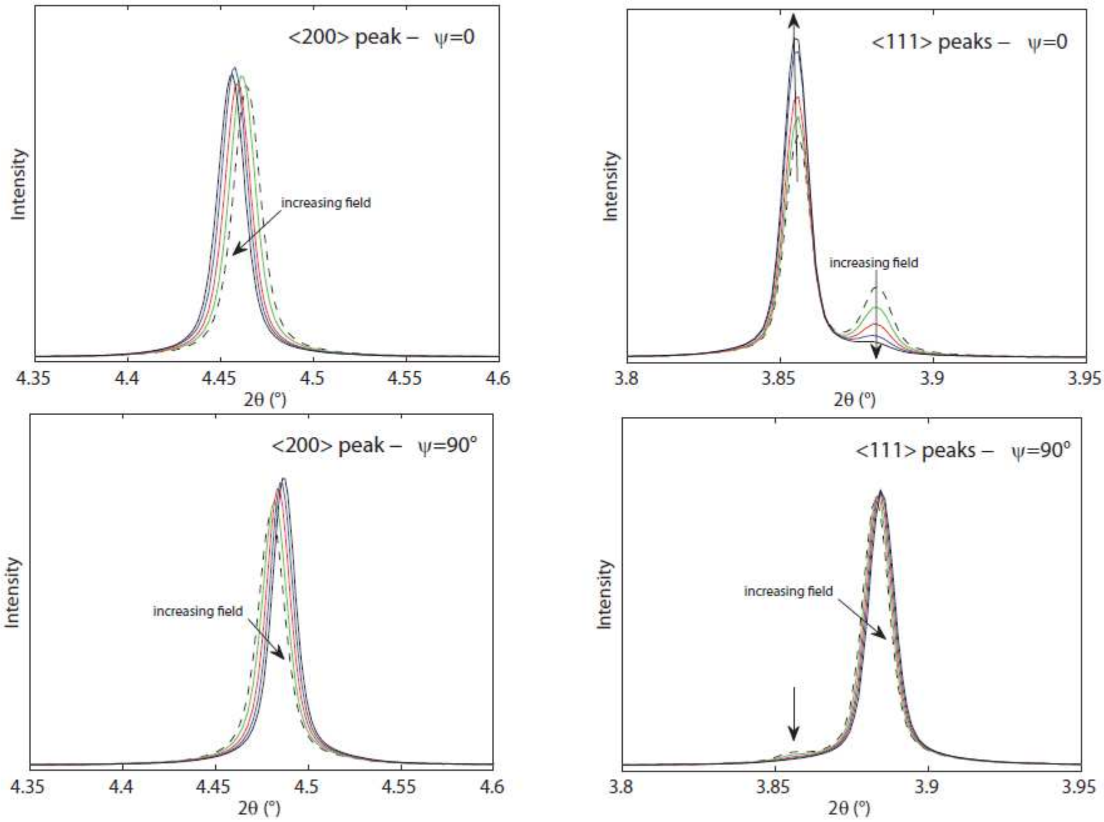

Fig. 2. Selected regions of the diffraction patterns obtained during the initial electric field cycle (between points A and B in Fig. 1).

A similar set of results was obtained during compressive loading under a static electric field of 2.5 kV/mm, as illustrated in Fig. 3. However, in this case the opposite trends were observed, indicating that the applied compressive stress acted to cancel out the piezoelectric strain induced by the static electric field.

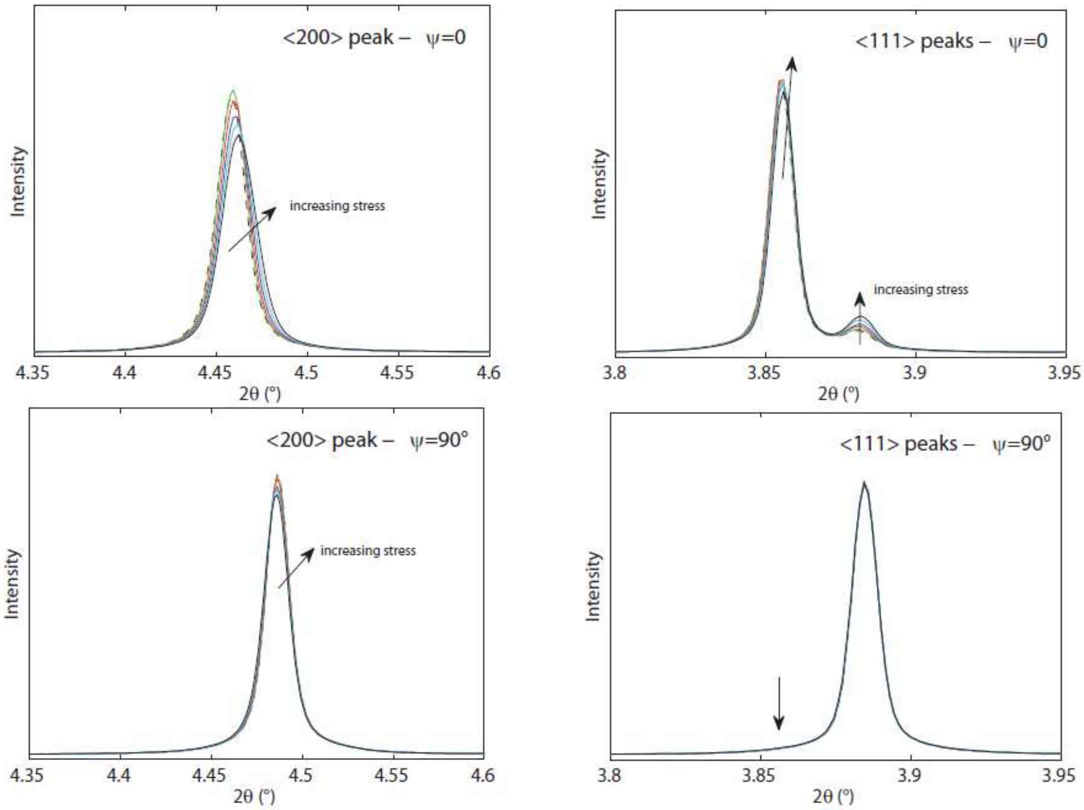

Fig. 3. Selected regions of the diffraction patterns obtained during the final compressive loading cycle under a static electric field of 2.5 kV/mm (between points C and D in Fig. 1.).

Analysis of the (200) diffraction peak profiles during the experiment yielded the results for lattice spacing, d_{200}, presented in Fig.4. By comparing these data with the loading profiles shown in Fig. 1, it is possible to track the evolution of the lattice spacing under combined electro-mechanical loading.

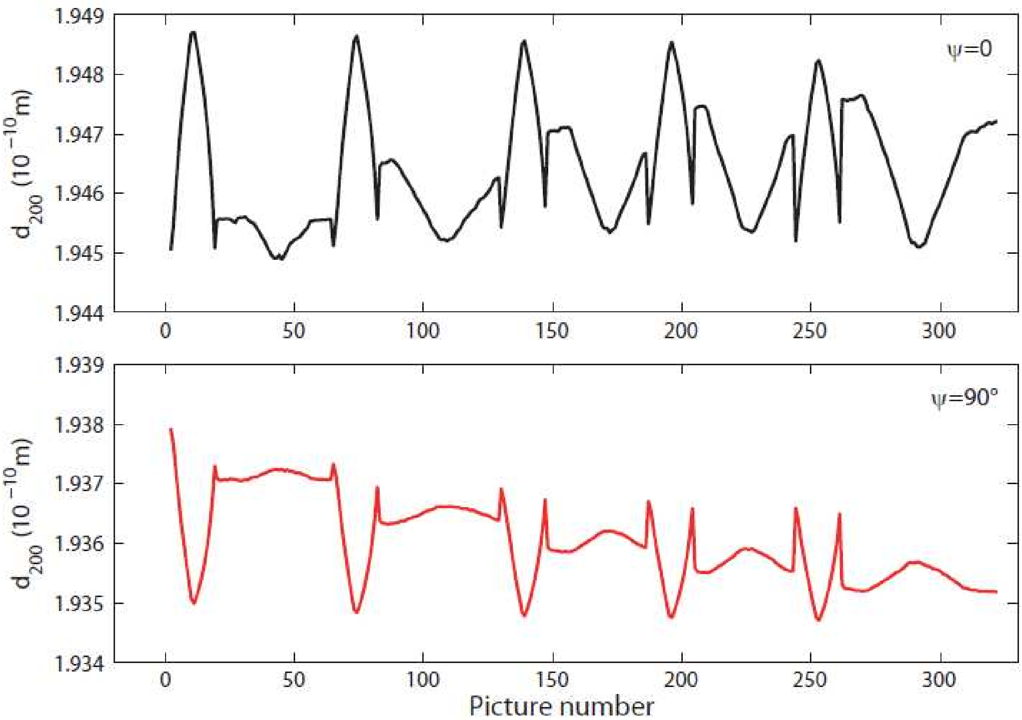

Fig. 4. Evolution of lattice spacing d_{200} during the loading experiment on a rhombohedral PZT55-45 ceramic.

The resulting 'blocking stress' curves obtained for the lattice spacing of the {200} grain family in a rhombohedral PZT 55-45 ceramic with the scattering vector along the electric field direction (ψ=0°) are shown in Fig.5(a). The observed trends are similar in form to typical macroscopic data[1] but they provide a measure of the strains experienced by specific grain 'families' on the local microscopic scale. It has notably been possible to analyse the responses of grain families having a range of orientations relative to the direction of the electric field/compressive stress axis. This is shown in Fig.5(b), where the blocking stress curves for a given electric field level (2.5 kV/mm) are plotted for different grain orientations (ψ=0 to 90°).

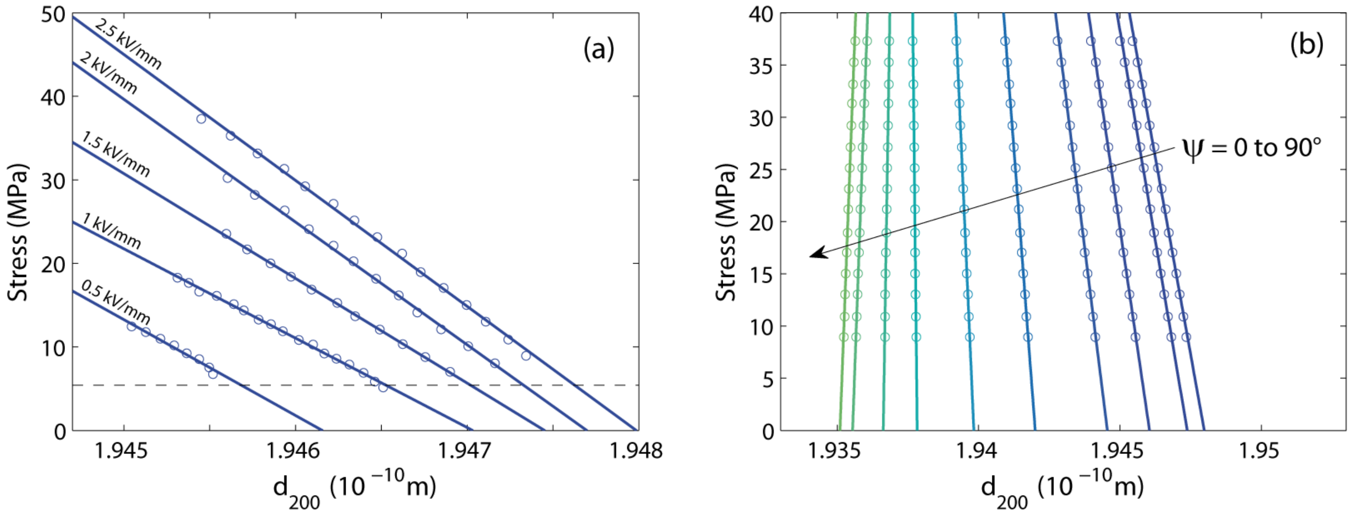

Fig. 5. In-situ blocking stress curves for rhombohedral PZT 55-45. Measurements conducted under (a) different static electric field levels (ψ=0°) and (b) different grain orientations (E=2.5 kV/mm, ψ=0 to 90°).

A micromechanical modelling approach was used to further evaluate the results of this experiment. By this means, it has been possible to interpret the evolution of the slopes of the blocking stress curves as a function of crystallographic orientation and single crystal anisotropy. It has been shown that the local elastic anisotropy is very significant in this material, with an elastic anisotropy ratio ($2C_{44}/(C_{11}-C_{12})$) as high as 3.3 being obtained. In other words, the Young modulus of the grains can vary by a factor from 1 to 4 according to the considered crystallographic direction. Such results have not been presented before and are the subject of a paper in preparation. It is of particular importance since single crystal elastic anisotropy plays a major role in the development of internal stresses in ferroelectric materials[2] and is thus key to durability of piezoelectric devices.

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[1] K.G. Webber, E. Aulbach and J. Rodel, J. Phys. D: Appl. Phys. 43, 365401 (2010).

[2] L. Daniel, D.A. Hall, P.J. Withers, "Analysis of the contribution of elastic anisotropy to internal stresses in ferroelectric materials using a multiscale modelling approach", Proceedings of the Electroceramics for End-users VII conference (PIEZO 2013), Les Arcs, France (2013).