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Overview

Periodic domains arise in ferroelectric materials as a compromise in energy associated with various external forces such as depolarizing field, substrate-induced strain and applied electric field. The occurrence of domains, their topologies and dynamic behaviour in response to an external electric field are issues of crucial importance for the design and application of these materials in electromechanical and optoelectronic devices.

The coercive field E_c required to reverse the polarization and create a monodomain state increases with decreasing film thickness d as $E_c(d) \propto d^{-2/3}$. For a 100 nm film of $PbTiO_3$ the local coercive field is of the order 200 kV/cm. In the present work we have studied several epitaxial films ranging in thickness from 100 unit cells (uc) to 12 uc, both under zero field and under weak fields $E << E_c$ in order to investigate possible influence of field strength and direction on the domain structure. The diffuse scattered intensity near the Bragg reflections gives information both on the average domain periodicity and orientation.

Experimental

Our samples in these studies are epitaxial films of $PbTiO_3$ (PTO) deposited by RF magnetron sputtering onto (001) oriented insulating $SrTiO_3$ (STO) single crystal plates of dimension $a \times a \times 0.5$ mm, a = 5–7 mm. The nearly perfect lattice match between cubic STO and the *ab*-plane of tetragonal PTO ensures that the polar *c*-axis of PTO will be well aligned in the growth direction of the film, favouring the formation of *c*-oriented domains. In the present work PTO films in the thickness range 100 uc to 12 uc, corresponding to $d \sim 41.5$ nm to 4.95 nm, were studied by SR X-ray scattering, using a specially designed sample holder for applying an electric field along the polar axis. [1] The 500 μm thick substrate tablet with film was fixed to the lower capacitor plate, a 0.5 mm thick copper plate with corresponding lateral dimensions. The upper capacitor plate was a 50 μm thick Kapton foil with a thin layer of gold deposited on the surface facing the sample. The distance between the capacitor plates was 2 mm. A single reflection, for most samples the 1 0 3 reflection, and the associated diffuse scattering were examined both by step scans in ω with step lengths in the range 0.01 – 0.05°, and by continuous scans over a range typically 3°. Scans were made without field and under nominal applied fields up to ~ 1.4 kV/cm through the PTO film. Scattered intensities were recorded using a CCD detector with pixel size 60.3 μm placed orthogonal to the X-ray beam at a distance of 360 mm from the sample. The data were binned 2x2 giving an angular resolution ~ 0.02°. Rectilinear reconstructions of reciprocal space were made with new locally developed software. A wavelength 0.9700 Å was used to avoid fluorescence from the heavy elements of the sample.

Results and brief discussion

In a previous experiment, 01-02-839, we studied a PTO film of thickness 50 uc (*d* ~ 20.7 nm) under variable electric fields by cycling the applied voltage in steps of 200 V between +1200 V and –1200 V. This work showed quite unexpectedly an asymmetric hysteretic response in domain size and order to changes in magnitude and polarity of the applied voltage. We refer to Experiment Report 01-02-839 for a definition of positive field direction and a more detailed description of the hysteresis. A publication based on this work is under preparation.

A major purpose of the present study was to clarify if the field-driven domain hysteresis is a general phenomenon in epitaxial films in this thickness range. We also wanted to search for possible new effects on the domain structure by employing stronger fields within the limit imposed by the the dielectric breakdown in dry air. A further aim was to compare various scan techniques for the collection of imaga data.

Between the capacitor plates there are three dielectrics, a 0.5 mm slab of STO, a PTO film of thickness *d* nm, and a 1.5 mm air gap. For the dielectric constant ε at room temperature we can use the values 295 for single crystal STO [2], and 20 for PTO film [3]. For *d* = 20 nm (near 50 uc) and an applied voltage of 2000 V we obtain ~ 13.3 kV/cm, ~ 0.67 kV/cm and ~ 45 V/cm for the nominal fields through the air gap, the PTO film and the STO slab, respectively. Compared to the estimated coercive field for this film thickness the applied field is of the order 0.1 %.

Five different PTO films of thickness 100, 50, 27, 16 and 12 uc were examined. Applied voltages were in general varied between +2000 and –2000 V, in one case voltages up to 4200 V were applied, this voltage generates fields of ~ 1.4 kV/cm and ~ 28 kV/cm through the film and the air gap, respectively. For the 50 uc thick film we studied the reflection 0 0 3 in order to compare with the results obtained previously for 1 0 3 in Exper. 01-02-839.

The data that have been processed so far

- support the observation that domain size and order exhibit an asymmetric hysteretic response to changes in magnitude and polarity of an applied electric field E of the order 0.1 % of the estimated E_c

- confirm that the direction of the hysteresis is uniquely linked to the field direction, and independent of the initial direction in stepping the voltage from zero

- indicate that for positive potential differences increasing above the magnitude favouring a smaller domain size with *d*-specific average period there is a beginning reversal from this state towards one with more disorder and larger domains

The present work has been very valuable in confirming previous observations, and suggesting new features in the field-induced dynamics of domains. It was inevitable that it became to a large extent an exploration of various measurement techniques and a prestudy of domain behaviour. Systematic studies of some selected film samples, in particular films with *d* < 50 uc, must be carried out also under stronger fields than used in Exper. 01-02-839. Data must be collected in a manner enabling a 3D reconstuction of the diffuse intensity as seen both along the Bragg truncation rod and orthogonal to this rod.

References

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