Four circles X-ray diffractometry and piezoelectric force microscopy for the study of the local switching properties of PZT thin films

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$PbZr_{0.53}Ti_{0.47}O_3$ (PZT) thin films (90-nm thick) have been grown on $TiO_x/Pt/TiO_2/SiO_2/Si$ and $TiO_x/Pt/MgO$ substrates by radio-frequency magnetron sputtering. The temperature of deposition was 500°C and the gas atmosphere was a mixture of oxygen (20%) and argon (80%). Atomic force microscopy study evidences the existence of a dense granular-like surface for the two kinds of films. *D-E* hysteresis loops performed at the "macroscopic" scale, i.e. considering many grains and grain boundaries, show that the average coercive field is 120 kV/cm for the film grown on $TiO_x/Pt/TiO_2/SiO_2/Si$ and 210 kV/cm for the one deposited on $TiO_x/Pt/MgO$. Piezoelectric force microscopy imaging indicates that there is no one-to-one correlation between the topography and piezoelectric images.

In order to get information about the switching properties of the films at a submicron scale level, local "in-field" piezoelectric hysteresis loops performed by atomic force microscopy have been recorded at the nanometer scale by positioning the tip at the center of the grains. Fifty loops were recorded for each film.

Whatever the substrate, a bimodal distribution of coercive voltage V_c was obtained. In the case of the films grown on $TiO_x/Pt/TiO_2/SiO_2/Si$, 70% of the Vc-values were included between 0.7V and 0.9V while 30% were between 0.9V and 2.0V. For the films deposited on

TiO_x/Pt/MgO, 88% of the values were included between 1.3V and 2.4V and 12% between 2.6V and 2.9V.

In order to explain these values, texture analysis of the films were carried out using four circles X-ray diffraction. For the film grown on TiO_x/Pt/TiO_2/SiO_2/Si substrate, quantitative texture analysis (QTA) reveals that 70% of the sample volume is oriented with the ***c-*** axes perpendicular to the sample plane and 30% of the sample is oriented with the ***a-*** axes perpendicular. On TiO_x/Pt/MgO substrate, an upper limit of around 10% for the sample volume oriented with the ***a-***axis perpendicular to the sample plane is obtained.

The combination of local piezoelectric hysteresis loops and QTA, shows that the percentage of (001)- and (100)-oriented grains obtained for each kind of films corresponds to the percentage of low and high coercive voltages values, respectively. The lower coercive voltages for the (001)-oriented grains are explained by taking into account the different orientations of the polar axis in the grains. Also, when considering the switching properties of grains having the same orientation in the two kinds of films, we show that the larger coercive voltages are obtained for the films having the larger c/a ratio (film grown on TiO_x/Pt/MgO substrate). This is attributed to structural strains imposed to the film.

We conclude that four circles X-ray diffraction and piezoelectric force microscopy are two complementary powerful techniques which permit to relate the local switching properties of PZT thin films to their local structural properties.