## FERROELECTRIC BEHAVIOR OF POLYCRYSTALLINE ULTRATHIN LEAD TITANATE FILMS

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Ferroelectric lead titanate based thin films have extensively been studied for their applications as active elements in non-volatile ferroelectric random access memories (NV-FeRAM) and microelectromechanical systems (MEMS). The miniaturization trends followed by the electronics industry results in a drastic reduction of the dimensions of the devices and their elements, which has stimulated an increasing number of studies on the preparation of ferroelectric nanostructures and their properties. For example, regarding the Nanoelectromechanical Systems (NEMS), one of the major challenges they face now is related to the search of alternative transduction elements [1], as most of the techniques used in MEMS reach their limits in the nanoscale. A promising solution is the use of piezoelectric transduction with ferroelectric films. To this aim, high quality ultrathin capacitors need to be prepared. Among the different methods used for the fabrication of films, Chemical Solution Deposition methods have the advantages of their low cost, good compositional control and large deposition areas, but it has traditionally regarded as useless for the preparation of very thin films. Recently, we have shown that lead titanate based continuous films down to 13-18 nm thickness can successfully be prepared by CSD [2,3], by a careful control of the precursor solutions and deposition parameters. Deposits are made on Si-based substrates, on which most of the current microelectronics technology is based. Even the thinnest films show significant piezoelectric activity (Fig. 1), which made them promising for applications as transducer elements in the complex structures of NEMS.

The following step towards their integration in real devices is that the capacitors obtained are macroscopically addressable. Electrical characterization of ferroelectric ultrathin films in most studies is carried out by scanning probe microscopy, with only few recent exceptions [4]. We present results of our dielectric, ferroelectric results on capacitors down to  $\approx$ 40 nm thick, and the strategies we are following to push down this limit, mainly focused on the improvement of the microstructural quality of the films.

Properties are of course affected by the size reduction. The use of polycrystalline Pt bottom electrodes, easily integrated on Si-based substrates and resistant to the thermal treatments that ferroelectrics usually need during processing, results in the fabrication of films polycrystalline in nature. Although there is extensive work devoted in the literature to study the effect of grain size on the ferroelectric domain configuration of ferroelectrics, these studies have mostly been carried out in bulk ceramics. Alternatively, thickness effects that include electrode charge screening, parasitic interfacial layers or strain gradients have primarily been studied in epitaxial films. In this work we try to elucidate which of these two types of size effects is more important for the different thicknesses. We observe an abrupt decrease of the remnant polarization (Fig. 2) which we attribute to the disappearance of 90°-type ferroelectric domains with grain size reduction, as observed by Piezoresponse Force Microscopy (Fig. 3). However, the piezoelectric behaviour of the thinnest films observed on their local piezoelectric loops (Fig. 1) is strongly dependent on the strain gradients produced by the clamping of the substrate. From the results obtained we draw conclusions for the improvement of the properties of these ferroelectric ultrathin films.

## **References:**

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**Figures:** 



**Fig. 1.** Local piezoelectric loops of an ultrathin lead titanate film.

**Fig. 2.** Corrected P-E hysteresis loop of films with decreasing thickness.



**Fig. 3.** Phase piezoresponse force microscopy images that show the sense of the out-of plane polarization vector of films with (a) 132 nm and (b) 40 nm thickness. Banded twin domains corresponding to 90°-type ferroelectric domains are observed in (a).