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Strain and composition effects in epitaxial ferroelectrics

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Motivation and Outline

Ferroelectric materials show a number of desirable properties such as a switchable polarization, high piezoelectric responses and high dielectric constants. For this reason they are widely used in devices such as memory elements, ultrasound generators, capacitors, gas igniters and many more. As the polarization, the defining property of a ferroelectric, is very sensitive to strain, strain can be efficiently used to enhance the properties of thin film ferroelectrics. Thin film ferroelectrics can be grown under epitaxial strain when using suitable crystalline substrates. This strain originates in the difference in lattice parameters between substrate and ferroelectric. Theory predicts rich phase diagrams with strain as a control parameter [1–3]. This strain tuning has been successfully applied by increasing the transition temperatures by hundreds of degrees or by transforming a paraelectric material, SrTiO_3 into a room temperature ferroelectric. [4, 5] Despite this progress, full control of the structure and properties of ferroelectric thin films under strain has not been achieved yet. This thesis aims to unveil novel aspects of the effect of epitaxial strain on the properties of ferroelectric thin films. Thin films close to boundaries between ferroelectric phases with different symmetry are particularly interesting, as large piezoelectric and dielectric responses are expected.

In chapter 1, a general introduction on ferroelectricity and thin films will be given. The properties of the main material under study, PbTiO_3 , will be described. Landau-Ginzburg theory is used throughout this thesis to aid interpretation of experimental data and as a guide to experiments. The phenomenological Landau-Ginzburg description of (strained) ferroelectrics will be introduced. The last part of chapter 1 will give an overview on the recent theoretical and experimental progress

in using strain to tune the properties of ferroelectrics as well as some notes on the experimental difficulties in applying strain tuning.

Growing fully strained ferroelectrics requires a high crystalline quality and thicknesses low enough to prevent strain relaxation. Molecular Beam Epitaxy (MBE) is a suitable technique to grow high quality epitaxial thin films. Chapter 2 shows the basics of MBE and thin film growth. The adsorption controlled growth of $\text{Pb}_x\text{Sr}_{1-x}\text{TiO}_3$ thin films will be described.

X-Ray diffraction (XRD) is the main technique we used to study both the structural and ferroelectric properties. Although hysteresis loop and dielectric measurements give the most direct access to the ferroelectric properties, they are difficult to implement because of the high leakage present at the low thickness required to prevent strain relaxation. Moreover, buffer electrode layers needed to perform electrical measurements will most likely modify the misfit strain imposed by the substrate and/or the electrical boundary conditions, adding an extra degree of freedom. That is why XRD is exploited to the maximum in the study of ferroelectric thin films. The basics of XRD will be described as well as an overview of ways to study the ferroelectric phases using diffraction. XRD techniques are capable of giving information about the magnitude and direction of the polarization, the transition temperature and domain structures, but also in some cases about the presence of dead layers and substrate/film termination.

Transition temperatures of ferroelectrics increase with increasing magnitude of the strain. In PbTiO_3 grown on SrTiO_3 this leads to a transition temperature that is higher than the growth temperature. In chapter 4 we study the implications of growing in the ferroelectric phase and its effect on the domain structure and properties.

Strain tuning is a very promising technique to enhance ferroelectric properties (increase polarization and T_C), but the limited number of suitable substrates hampers its applicability. An approach to overcome this issue is proposed in chapter 5. By combining epitaxial misfit strain with the effect of cation substitution on the lattice parameters, the misfit strain can be fine tuned. Thin films of $\text{Pb}_x\text{Sr}_{1-x}\text{TiO}_3$ of various compositions grown on DyScO_3 and SrTiO_3 substrates are used to

explore the experimental possibilities. In $\text{Pb}_x\text{Sr}_{1-x}\text{TiO}_3$ on DyScO_3 the polarization can be continuously tuned to be both either predominantly out-of-plane or in-plane and allows to chose highly responsive materials placed at the boundary between two phases.

