



Bellaterra, 4th June 2019

The Thesis written by Mr. Zahradnik reports on structural, magnetic, optical and magneto-optical characterization of $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ (LSMO) and SrRuO_3 (SRO) thin films. In a first part, the magnetic properties of LSMO thin films are studied as a function of the epitaxial strain imposed by different substrates. The Thesis analyzes a possible influence of oxygen octahedra rotations (OOR) on the magnetic properties of the films, beyond the effect of the epitaxial constraint. The possible role of OOR in functional properties of thin film heterostructures is an important topic nowadays, where interfaces between materials of different crystal symmetries can drive localized distortions of the lattice around the interfaces, changing the orbital bonds and, thereby, modulating physical properties. The work by Mr. Zahradnik enables concluding that the observed magnetic properties cannot be described by the effect of OOR, but to the strain-driven changes in the electronic band structure.

Beyond static-strain, the Thesis also analyzes the effects of dynamic strain by coupling the LSMO films to piezoelectric PZT layers and applying electric fields. The study finds a modulation of the magneto-optical properties that can be understood in terms of changes in the thickness of the films due to electrically driven changes of strain.

Finally, a last part of the Thesis analyzes the impact of substrate vicinity on the structural and magnetic properties of SRO thin films.

A first chapter is devoted to an Introduction to the fundamentals of the physics of colossal magnetoresistance materials (among which LSMO is included), emphasizing the role of changes in bonds and angles in modulating transport and magnetic properties and the effect of epitaxial strain on the structural properties of the thin films. This introduction is necessary to understand the possible role that OOR could have on physical properties. The introduction also puts emphasis on the interest of understanding of dynamic strain control of magnetic properties, making the connection with recent results showing electric-field control of domain wall motion in magnetic nanostructures. In addition, a quick introduction to the properties and interest of SRO is given in the Introduction, with emphasis on the interplay of structure and magnetic properties.

Chapter 2 describes the theoretical framework necessary to understand how information about the polarization of light can be extracted in experiments involving different optical elements, including the effect of the sample itself on polarization. Also, the chapter explains how the permittivity tensor can be extracted from experiments and also reviews the microscopic theory of magneto-optical responses. Important to understand the content and scope of the Thesis, a clear discussion is done about the types of magneto-optical responses, i.e., diamagnetic and paramagnetic transitions. The text is clearly written and makes a useful introduction to basic concepts.



The next chapter describes the experimental techniques for thin film growth, x-ray diffraction, SQUID magnetometry, spectroscopic ellipsometry and magneto-optical spectroscopy and microscopy. The description of the different experimental methods is concise and well written, enabling a quick grasp on basic concepts.

Subsequently, chapter 4 gives a quick overview on the analyzed samples and devices. This is very useful, as it facilitates the comprehension of the results that are described in the following chapters.

Chapter 5 discusses the effect of strain on the structural properties of LSMO films. The analysis of x-ray diffraction is extensive and clear, and the conclusions robust. Especially relevant in this chapter is the discussion of the optical properties by spectroscopic ellipsometry and magneto-optical spectroscopy, which enables to have an insight into the electronic band structure and the electronic transitions at the origin of the spectral response. This study enables to see one particular transition, which presence or absence depends on the degree of strain imposed on the thin films. This is one of the most important conclusions of the Thesis. There is an extensive and exhaustive discussion about the origin of the emergence or disappearance of this transition on varying the strain. The arguments to discard OOR are convincing, mostly based on the experimental data displayed in Figures 5.18-21 and the summary tabulated in Table 5.6. At the end, the conclusion is that the involved transitions are Mn d-d and strain drives changes in the energies of these electronic states. It is remarkable that the ab-initio calculations included in the Thesis support well the arguments drawn after the experimental observations and reproduce well the spectroscopic features, as shown in Figure 5.23. Maybe, a deeper discussion about the transitions inferred from spectroscopic ellipsometry and how they relate to the transitions extracted from magneto-optical spectroscopy would have been desirable for completeness. Overall, the conclusions extracted from this analysis are important in the context of structural properties of thin film heterostructures and the specific role of interfaces and provide relevant guidelines for future work. Along these lines, the Thesis reports clear results that changes in electronic structure are driven by epitaxial strain, excluding convincingly any effect from OOR.

A second part of chapter 5 describes experiments reported in LSMO/PZT samples, where PZT is piezoelectric. The experiments have enabled to shed light on the effects of dynamic strain on the optical and magneto-optical responses of LSMO films. The conclusion is that the modulation of these responses are driven by changes in thickness caused by piezoelectricity. Again, I find the study thorough and carefully done and the conclusions robust. I deeply appreciate the efforts to model the role of optical interference as the thickness of LSMO is changed by the piezoelectric effect on the optical response (Figure 5.29), which gives important clues to understand the effect of dynamic strain. These results may provide insights into magnetoelectric devices in hybrid magnetic/PZT structures.

Finally, the influence of substrate vicinality on the magnetic properties of SRO films is analyzed in Chapter 6. I appreciate the carefulness of the results reporting on the time



evolution of the magnetization, describing its relaxation with time. Often, this sort of phenomena are overlooked, and it is noteworthy that attention is dedicated to this issue. I also appreciate positively the analysis of magnetic domains by AFM (Figures 6.7-11), which enables a discussion about the nucleation of magnetic domains and the effect of miscut angle on nucleation. In my view, the conclusions about magnetic domain nucleation and microscopic structural properties are well founded.

In summary, the Thesis presented by Mr. Martin Zahradnik is an original work, which comes timely in the context of the physics of thin film heterostructures involving perovskite oxides. It presents important results that may guide future work, especially regarding the responsivity of magnetoelectric devices using magnetic/piezoelectric hybrid structures. At the same time, the work provides important insights into the optical responses of these materials and their relation to the electronic band structure. Mr. Martin Zahradnik has presented a rigorous work, in which he demonstrates a deep understanding of the subject and may serve as further stimulus for further works in the field. In view of this, I think that the candidate fully deserves the title of Doctor in Physics.