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Growing oxide thin films in a low-energy electron microscope

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Introduction

1.1 Introduction

Silicon is the workhorse for modern electronics. The performance of Si-based devices is mainly boosted by their miniaturization, which will soon be limited by quantum mechanical obstacles. A way to overcome this is to find other materials than the covalent semiconductors, that can be used in electronics. Silicon is a conventional semiconductor where the conductance or metallicity derives from exciting electrons over the band gap, or doping the material with either electrons or holes. In all cases, the number of charge carriers is low, with in particular long screening lengths for electric fields as a consequence. In silicon, the charge carriers behave as independent particles, with little interaction between them selves or with the lattice. In contrast, in transition metal oxides (TMO) the metallicity is determined by details of the electron-electron interaction between the 3d electrons of the transition metal ion. The carrier concentration can be very high, in the order of an electron per atom, and screening lengths small. Also, the interactions can lead to complex phenomena such as high-temperature superconductivity or colossal magnetoresistance. Special properties and phenomena can also be achieved at the interface between different oxides. As Herbert Kroemer said during his Nobel lecture: "Often, it may be said that the interface is the device"¹. Already many devices based on conventional semiconductors exploit interfaces, for example transistors, lasers, memory and solar cells. The interfaces between TMOs, however open up a new world of interesting physics.

Research of TMO devices was long held back by the difficulties in growing TMO multilayers. Developments in the last decades have improved this. The

1 ability to obtain well-defined interfaces by fabricating surfaces consisting of only a single well-defined ionic or atomic plane, the so-called singly-terminated surface, has been a key step to obtain sharp interfaces². Furthermore, the ability to grow clean multilayers by molecular-beam epitaxy (MBE)³, and pulsed laser deposition (PLD)⁴ has been of utmost importance. Moreover, the development of in-situ reflection high-energy electron diffraction (RHEED)⁵ for PLD at elevated pressures has led to thickness control down to the unit cell range.

To further optimize the material properties more information during the growth process would be of great value. Especially the structural and electronic properties are important for the resulting film. Basic structural information can be obtained from the diffraction pattern obtained by RHEED but, electronic information is mostly missing. Other analysis techniques have been hampered by the extreme conditions, i.e. high temperature and oxygen background pressures. The requirements of surface sensitivity and non-invasive, non-contact techniques limit the analysis to electron optical techniques.

As most widely used system to grow TMOs we focus on PLD. We extend the possibility for in-situ film growth analysis by introducing low-energy electron microscopy (LEEM), with in-situ PLD system, as a new technique in this field. RHEED does not allow for spatial resolution and is more difficult to analyze than low-energy electron diffraction (LEED). Low-energy electron microscopy (LEEM) is able to combine spatial resolution with LEED. New developments, partially in our lab, allow one to investigate the electronic structure of a surface or thin film⁶. This extends LEEM to a versatile set-up for material science, combining structural and electronic properties with lateral resolution and fast investigation.

An ideal device for electronics would be a two dimensional electron gas (2-DEG) at the interface between two insulators, with high electron mobility, tunable by a gate and working at room temperature. The 2-DEG found between the TMO band-insulators LaAlO_3 and SrTiO_3 was a remarkable discovery⁷, which comes close to this ideal device. However, the exact origin of this 2-DEG is still not understood. In this thesis we will use LEEM as a method to investigate the film properties of the hetero-structure during growth.

1.2 Thesis outline

The main material system to be investigated is the $\text{LaAlO}_3/\text{SrTiO}_3$ interface. This will be introduced in chapter 2 and the method used for studying the interface, LEEM with its full set of subtechniques, in chapter 3. While the SrTiO_3 building block has already been studied in LEEM by Hesselberth *et al.*⁸ we continue with the other building block, LaAlO_3 , in chapter 4. For the growth studies we require a PLD system. Its development is described in chapter 5. Also its abilities are demonstrated by the growth of homoepitaxial SrTiO_3 . With the building blocks and equipment in place we study the formation of the $\text{LaAlO}_3/\text{SrTiO}_3$ hetero-structure in chapter 6. Finally, we use our knowledge about the $\text{LaAlO}_3/\text{SrTiO}_3$ hetero-structure to investigate $\text{LaAlO}_3/\text{SrTiO}_3$ on $\text{Ca}_2\text{Nb}_3\text{O}_{10}$ nanosheets

deposited on Si, in chapter 7. In this chapter everything comes together and the spatial resolution of the LEEM is of great importance.

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