

## **Oxide electronics: Increasing trends and future challenges**

Guus Rijnders.

*MESA+ Institute for Nanotechnology (Inorganic materials science), University of Twente, P.O.Box 217, 7500 AE, Enschede, the Netherlands*

**Abstract** Oxide materials exhibit a wide range of functional properties, such as isolators, superconductors, ferromagnets and ferroelectrics. This diversity in materials properties opened the very active research area of “oxide electronics”. Nowadays, it is possible to control the growth of oxide thin films with atomic precision and these oxide films start to play a very important role in electronic devices.

Recently, a new “degree of freedom” has been recognised. By atomic control of interfaces between oxide materials, i.e., the chemical composition and crystalline structure, new phenomena can be observed. One example is the electronically conducting interface between  $\text{SrTiO}_3$  and  $\text{LaAlO}_3$ , both insulating oxides.

In this paper, the recent developments of oxide thin film growth and future challenges in the field of oxide electronics are addressed.

Oxide materials start to play a very important role in electronic devices. Nowadays, it is possible to control the electronic properties of these materials with nanoscale precision [1, 2, 3]. This class of oxide compounds with perovskite type crystal structure exhibit a broad range of functional properties, such as high dielectric permittivity, piezoelectricity and ferroelectricity, superconductivity, colossal magnetoresistance and ferromagnetism. Many of these phenomena occur in perovskite oxides that are lattice-matched within a few percent of one another. This enables fabrication of heteroepitaxial structures, in which the multiple degrees of freedom can be accessed, making a myriad of devices with novel functionalities possible [4]. The properties of oxide interfaces in such heterostructures can be exploited for applications in electronic devices. Specific oxide devices relying on interface properties have already been realized and employed, such as Josephson junctions and SQUIDs, magnetic tunnel junctions based on manganites and FeRAMS. However, a completely new class of (nano) devices can be envisaged and engineered exploiting the functional properties of oxides. A real breakthrough in this field can be obtained by controlling and tailoring the physical properties at the interfaces between different oxide materials. Interfaces and surfaces in such highly correlated systems offer far more application possibilities than interfaces involving only conventional metals and semiconductors, but are more complex. Interfaces alter the bulk electronic system sometimes with dramatic consequences; interfaces break the translational and the

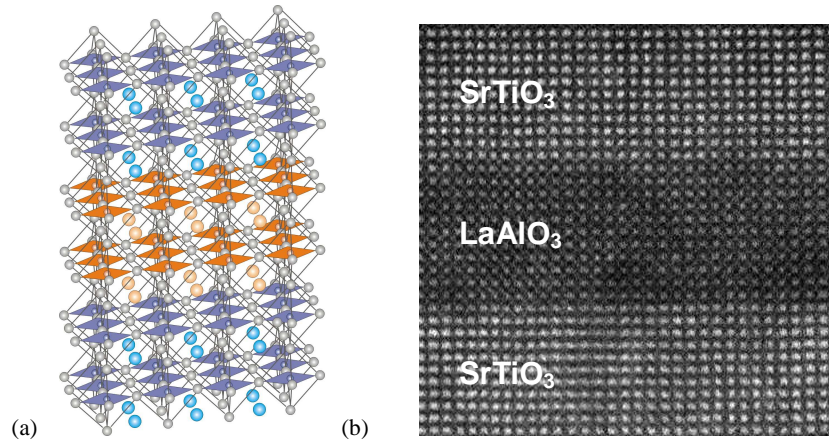


Figure 1: Schematic view of a SrTiO<sub>3</sub>/LaAlO<sub>3</sub>/SrTiO<sub>3</sub> heterostructure (a) Cross section high-angle annular dark field (HAADF) TEM image of the LaAlO<sub>3</sub>/SrTiO<sub>3</sub> superlattice along the [001] zone axis (b).

rotational symmetry, induce stress or strain, consequently altering the distances and bonds between the ions, giving rise to shift and distortion of the electronic states and energy levels and modifying the bands. By altering the electronic states, interfaces modify also electronic correlations. Because the correlations control the electronic behaviour of the material, their modification can induce remarkable changes of the collective electronic and magnetic properties.

The strong interplay of the crystal degree of freedom as well as the correlated character of electronic behaviour in perovskites allows for new microscopic physical mechanisms as well as new macroscopic physical properties. This is, for instance, best illustrated by recent work by Harold Hwang and our Laboratory [1, 5] on interfaces between epitaxial heterostructures of SrTiO<sub>3</sub> and LaAlO<sub>3</sub>, both band insulators. In Figure 1, a schematic view and cross-section transmission electron micrograph (TEM) are shown of a SrTiO<sub>3</sub>/LaAlO<sub>3</sub>/SrTiO<sub>3</sub> heterostructure, fabricated by pulsed laser deposition (PLD). Reflection high-energy electron diffraction (RHEED) is used to control the growth, which provides heterostructures with sharp interfaces. Because of the valence discontinuity at the interface, electronic reconstruction takes place between the two insulating oxides which gives rise to conductivity at the interface. To understand this interface activity, it is instructive to describe the perovskites in terms of their constituting AO and BO<sub>2</sub> layering sequence. For example, whereas the two band insulators SrTiO<sub>3</sub> and LaAlO<sub>3</sub> are seemingly similar, the SrO and TiO<sub>2</sub> layers are charge-neutral, while the charge states in the LaAlO<sub>3</sub> are (LaO)<sup>+</sup> and (AlO<sub>2</sub>)<sup>-</sup>, respectively. In heterostructures, the AO-BO<sub>2</sub> stacking sequence is maintained, and consequently a polarity discontinuity arises at the LaAlO<sub>3</sub>-SrTiO<sub>3</sub> interface. It has been shown [1] that due to this, the LaO:TiO<sub>2</sub> interface becomes conducting, with the conduction governed by electron-transfer from LaAlO<sub>3</sub> into the TiO<sub>2</sub> bonds of the SrTiO<sub>3</sub>. At the AlO<sub>2</sub>:SrO interface the AlO<sub>2</sub> acts as an electron acceptor, resulting in a p-type interface. Such conducting interfaces are analogous to two-dimensional electron (hole) gasses (2-DEGs) in semiconductors, which find

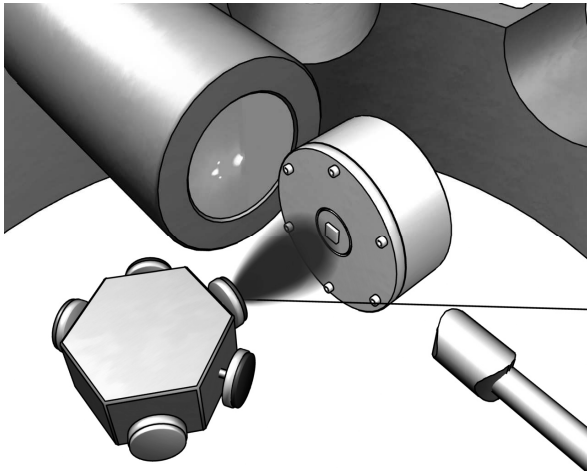


Figure 2: Schematic view of pulsed laser deposition with RHEED. The black line represents the trajectory of the laser beam. With every laser pulse, material is evaporated from a target and deposited on a substrate. With RHEED, the growth of this material can be controlled on an atomic level.

applications in e.g., opto-electronic, high-power RF and magnetoelectronic devices. This example

shows that perfect control of the atomic stacking at interfaces between perovskite oxides allows for tuning of the electronic structure of such interfaces.

The application of epitaxial<sup>1</sup> oxide thin films in devices, relying on multilayer technology, requires (atomically) smooth film surfaces and interfaces. Understanding of the different mechanisms affecting the growth mode is, therefore, necessary to control the surface morphology during thin film growth.

The fabrication of epitaxial complex oxide thin films involves deposition as well as subsequent growth. Here, growth is the incorporation of the adsorbed atoms (adatoms) at the film surface. Many surface processes during deposition and growth determine the thin film properties such as crystallinity in-plane residual stress and surface morphology and are, therefore, subject to in many studies. Reflection high-energy electron diffraction (RHEED) is most often used to study the mechanisms and film surface processes during growth, whereas scanning probe microscopy (SPM) is usually employed to study the surface morphology after deposition.

Frequently, physical vapour deposition (PVD) techniques, such as sputter deposition, molecular beam epitaxy (MBE) and pulsed laser deposition (PLD), are used for the fabrication of thin films. Among these techniques, PLD has emerged as a unique method to fabricate epitaxial and nearly single crystalline thin films of complex oxides, such as superconducting high- $T_c$  cuprates, metallic, ferroelectric, ferromagnetic, and dielectric oxides. Conceptually and operationally, PLD is a simple technique. A pulsed, highly energetic laser beam is focused on a target, resulting in ablation of material. This material is deposited on a substrate, placed opposite to the target, resulting in thin film growth.

---

<sup>1</sup> In epitaxial growth, a film with preferred crystallographic orientation is formed on a crystalline surface as the result of deposition of material onto that surface.

A unique feature of PLD is the modulated deposition. Short pulses (~100  $\mu\text{sec}$ ) with high deposition rate are alternated by a relatively long interval where no deposition takes place. Because of the instantaneous deposition, the two basic processes, i.e., random deposition and growth through rearrangement are separated in time, which is unique for PLD. Since the diffusivity is directly coupled to the surface properties, i.e. structural, chemical and physical, these can be studied during the initial growth. A good example is the observed self-organized conversion [6] of the terminating atomic layer from  $\text{RuO}_2$  to  $\text{SrO}$  during the initial growth of the ferromagnetic oxide  $\text{SrRuO}_3$  on  $\text{TiO}_2$ -terminated  $\text{SrTiO}_3$ . This conversion induces an abrupt change in growth mode from layer-by-layer to growth by step advancement, indicating a large enhancement of the surface diffusivity. This growth mode enables the growth of single crystalline and single domain thin films. We have shown that both conversion and resulting growth mode enables control of the interface properties in heteroepitaxial multilayer structures on an atomic level.

To summarize, perovskite ( $\text{ABO}_3$ ) oxides, interfaces and epitaxial heterostructures of these oxides exhibit a large amount of exceptional physical properties, providing the basis for novel concepts of oxide-electronic devices. For a given parent compound, a rich phase diagram is coloured-in by substitution of the A or B cations, and/or a change in the oxygen stoichiometry. The ionic character of the chemical bonds and the consequent possibility of electronic reconstruction often render interfaces in these materials strongly electronically active. These remarkable interface characteristics even increased the interest in these materials, especially in the field of conducting interfaces and artificial multiferroics. Perfect control of the crystalline structure, chemical bonding and the phase formed at the interface are a prerequisite to access the mentioned multiple degrees of freedom. However, there are many open questions which will be addressed by research groups in the near future. These questions relate to the following topics:

- the atomistic initial growth processes during heteroepitaxial growth;
- control of the interface structure, i.e., crystallinity and atomic stacking sequence;
- the relation between the interface structure and physical properties, such as electronic structure;
- the relation between the electronic structure of the growth surface and initial growth behaviour.

Furthermore, increasing trends in the field of oxide electronics are observable.

These are, amongst others:

- Materials by design; theoretical and computational work will drive the experimental groups to “design” materials with enhanced or new properties;
- Complex oxide heterostructures will be integrated with silicon technology. Integration of these technology platforms will enable devices with new functionalities. An example is the emerging field of piezo-MEMS technology;

- Major facilities (such as the ESRF in Grenoble) will play a important role in characterization.

To conclude, the application of complex oxides in electronic devices will enable new possibilities, provided that the fabrication can be performed with atomic precision. Recent progress in fabrication technologies enables researchers to obtain this precision and an even greater progress can be expected in the active research field of oxide electronics in the near future.

## References

- 1 Ohtomo A, and Hwang HY, *Nature* **427** (2004) 423.
- 2 Muller DA, Nakagawa N, Ohtomo A, et al., *Nature* **430** (2004) 657.
- 3 Mannhart J, and Schlom DG, *Nature* 430 (2004) 620.
- 4 G. Rijnders and D.H.A. Blank, "Build your own superlattice", News & Views, *Nature* **433** (2005) 369-370.
- 5 M. Huijben, G. Rijnders, D.H.A. Blank, S. Bals, S. Van Aert, J. Verbeeck, G. Van Tendeloo, A. Brinkman, H. Hilgenkamp, *Nature Materials* **5** (2006) 556-560.
- 6 G. Rijnders, D.H.A. Blank, J. Choi, C.B. Eom, *Applied Physic Letters* **84** (2004) 505.