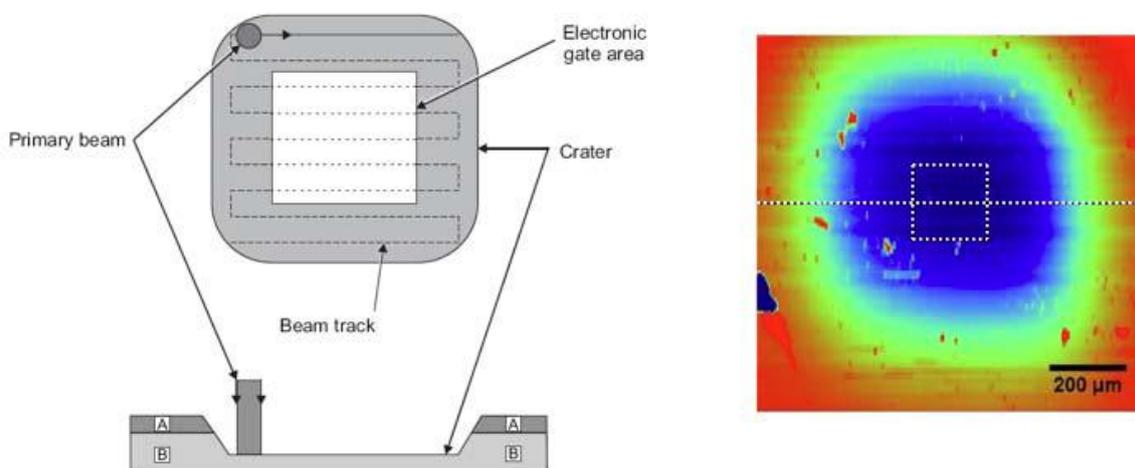


## The origin of oxygen in oxide thin films: Role of the substrate

During the growth of oxide thin films by pulsed laser deposition, there is always the question from which source oxygen is incorporated in the as-grown thin film: Target, background or substrate. This is an essential question since the optimization of the deposition conditions will depend on it. Using  $^{18}\text{O}$  exchanged  $\text{SrTiO}_3$  and  $\text{LaAlO}_3$  as substrates, a strong oxygen substrate-to-film transfer has been experimentally observed for epitaxially grown  $\text{SrTiO}_3$  and  $\text{LaAlO}_3$  thin films by SIMS depth profiling. This oxygen transfer effect can seriously change the respective thin film properties.

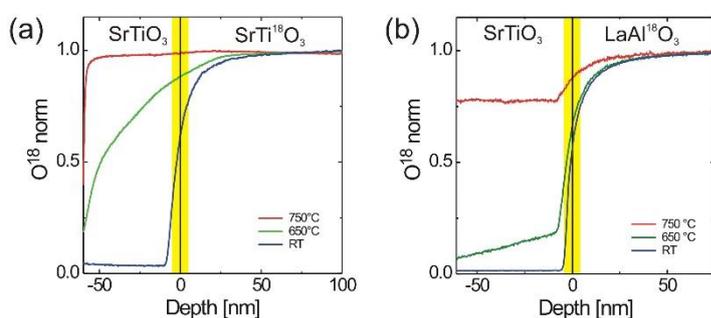
The preparation of high quality complex oxides thin films and heterostructures with atomic precision has considerably advanced in recent years considerably with a crystalline quality comparable to semiconductors and oxide semiconductors structures. The challenge associated with growing oxides is to provide and control the amount of oxygen during the growth to obtain the desired oxygen stoichiometry and hence appropriate physical properties of the respective oxide. For a deposition using pulsed laser deposition (PLD) it often seems to be sufficient to create the correct amount of plasma species within an oxygen background to form the chosen compound as a thin film on a suitable substrate, and to supply the missing oxygen with a subsequent annealing step.

To investigate the role of oxygen supplied by the substrate during a deposition, the oxygen diffusion properties of  $\text{SrTiO}_3$  and  $\text{LaAlO}_3$  thin films grown on  $^{18}\text{O}$  isotope exchanged  $\text{SrTi}^{18}\text{O}_3$  and  $\text{LaAl}^{18}\text{O}_3$  substrates were studied by dynamic secondary ion mass spectrometry (D-SIMS) which yields elemental depth profiles.  $\text{SrTiO}_3$  and  $\text{LaAlO}_3$  thin films have been prepared at three different deposition temperatures (nominal room temperature,  $650^\circ\text{C}$  and  $750^\circ\text{C}$ ) at a background pressure  $p=1.5\times 10^{-5}$  mbar and a laser fluence  $F=4\text{ Jcm}^{-2}$ . SIMS spectra were recorded using a Hidden Analytical EQS quadrupole mass spectrometer operated with a 2.5 keV Ar ion beam focused to 150  $\mu\text{m}$  diameter rastering over a square of  $1\times 1\text{mm}$  with an effective sampling area of  $500\times 500\ \mu\text{m}$ . The etched area is subsequently measured with a Dektak 8 profilometer to convert etching time into depth. (see Fig. 1). In addition a kinetic energy selection scheme is used to separate species with the same mass.



**Fig. 1:** Schematic representation of the raster crater (left) and a real crater measured with a profilometer (right). The different colours correspond to different depth levels.

The  $^{18}\text{O}$  diffusion from  $\text{SrTi}^{18}\text{O}_3$  into  $\text{SrTiO}_3$  shows a pronounced dependence on the deposition temperature (Fig. 2a). Whereas for a room temperature deposition, no traceable  $^{18}\text{O}$  diffusion into the film has been measured, the situation changes dramatically for elevated deposition temperatures. At  $650^\circ\text{C}$ , there is considerable  $^{18}\text{O}$  diffusion from the substrate into the film, while at  $T_s=750^\circ\text{C}$  there is no significant difference with respect to the amount of  $^{18}\text{O}$  measured in the film and substrate. In the case of  $\text{SrTiO}_3$  grown on  $\text{LaAl}^{18}\text{O}_3$  a significant and homogeneous oxygen contribution is measured for the  $\text{SrTiO}_3$  film prepared at  $T_s=750^\circ\text{C}$  (Fig. 2b). Even at  $650^\circ\text{C}$ , there is still a significant  $^{18}\text{O}$  intake which can be detected up to the film surface. Measuring Sr and Ti species from the substrate and film simultaneously with  $^{16}\text{O}$  and  $^{18}\text{O}$ , the elemental composition of film and substrate are very similar. This implies that oxygen in the  $\text{SrTiO}_3$  thin film is supplied by the substrate and the oxygen provided by the target seems to play a minor role for this system. The results also indicate that the initially formed film is oxygen deficient and a chemical gradient is in favour of supplying oxygen via the substrate.



**Fig. 2:** a)  $^{18}\text{O}$  SIMS depth profile of  $\text{SrTiO}_3$  on  $\text{SrTi}^{18}\text{O}_3$  grown at  $T_s=750^\circ\text{C}$ ,  $650^\circ\text{C}$  and room temperature. The sharp drop of the  $^{18}\text{O}$  signal near the  $\text{SrTiO}_3$  surface for the film grown at  $T_s=750^\circ\text{C}$  could be related to a back-exchange of  $^{16}\text{O}$  at room temperature. b)  $^{18}\text{O}$  SIMS depth profile of  $\text{SrTiO}_3$  on  $\text{LaAl}^{18}\text{O}_3$  grown at  $T_s=750^\circ\text{C}$ ,  $650^\circ\text{C}$  and room temperature.

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