

Pulsed laser deposition chamber for *in situ* X-ray diffraction

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A sample chamber has been constructed for studying the growth of thin films by pulsed laser deposition *in situ* with surface X-ray diffraction. The achievable temperature ranges from room temperature to 1073 K in a controlled oxygen environment. The partial pressure of the oxygen background gas covers the range from 0.1 to 10^5 Pa. The first results, showing intensity oscillations in the diffracted signal during homoepitaxial deposition of SrTiO₃, are presented.

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1. Introduction

Pulsed laser deposition (PLD) is a very successful technique that is ever more used for the growth of thin complex oxidic films (Chrisey & Hubler, 1994). It makes use of a laser to ablate target material, which is deposited on a heated substrate, where a thin film is grown. Since with each laser pulse only a fraction of a monolayer is deposited, the thickness of the film can be controlled very accurately. Until recently (Eres *et al.*, 2002), *in situ* monitoring of the growing interface has been performed mainly using electron diffraction techniques, even at relatively high background pressure (Rijnders *et al.*, 1997). Unfortunately, quantitative analysis of the electron scattering is hampered by the fact that the kinematical description does not apply, unlike in the case of X-rays. The study of crystal growth using surface-sensitive X-ray scattering techniques is a field that has gained much interest over the past years (Renaud, 1998; Vlieg, 2002). Combining the high-brilliance synchrotron X-ray beams, nowadays readily available at third-generation sources, with sample chambers, allowing for grazing-incidence geometry, opens the possibility to study the growing interface or surface on an atomic scale. Here we present the design of a chamber for studying *in situ* the PLD process using synchrotron X-rays.

2. Requirements and present design

Three different parameters play an important role in the PLD process: substrate temperature, oxygen background pressure and the laser fluence. In the case of growth of complex oxidic materials, like ferroelectrics and high- T_c superconductors, the substrate temperature is typically 1073 K and the oxygen background pressure is around 10 Pa. Since in some cases the oxygen content of the grown materials is not ideal at these high temperatures, the oxygen pressure in the chamber is increased to 10^5 Pa during cool-down to room temperature, in order to obtain the desired phase. The laser beam, having typically a wavelength of 248 nm, is focused by a lens, resulting in a fluence of approximately 1.5 J cm^{-2} in the focal spot on the target. In the present design, shown in Fig. 1, the laser enters the chamber through a fused silica window that is fixed to the aluminium chamber wall, thereby hitting the target at an angle of 45° . SXRD measurements are typically performed at a fixed angle of the order of 1° of the X-ray beam with respect to the substrate surface. Since the

measurements are performed with one point detector, during deposition only one reciprocal space point is being monitored at a time. Therefore deposition is interrupted after a number of monolayers, thereby allowing for movement of the diffractometer angles and detailed measurements of the grown film. The X-rays pass through the middle aluminium part, where a cylindrically shaped strip of height 27 mm is thinned to a thickness of 0.5 mm, in order to minimize absorption. The opening angles achievable in this way are 0 to 20° for the incoming and outgoing beam with respect to the surface and 310° for rotation around the surface normal. The choice of geometry when monitoring the diffracted intensity during deposition is free, *i.e.* both in-plane or out-of-plane reflections can be

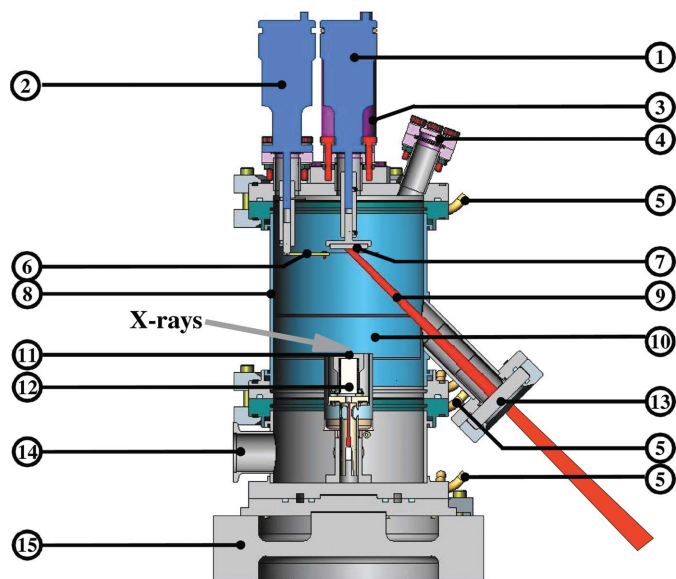


Figure 1
Cross-sectional view of the sample chamber. The height from base plate to top lid is 260 mm, and the diameter of the middle part is 140 mm. 1, Feedthrough for target spindle; 2, feedthrough for pre-ablation shutter; 3, electro-motor for target spindle; 4, viewport; 5, water-cooling connections; 6, pre-ablation shutter; 7, target; 8, aluminium chamber wall; 9, laser beam; 10, thinned part of aluminium wall (0.5 mm); 11, substrate position; 12, resistive wire heater embedded in nickel heater block; 13, fused silica laser window; 14, vacuum pump connection; 15, base plate ($d = 150$ mm).

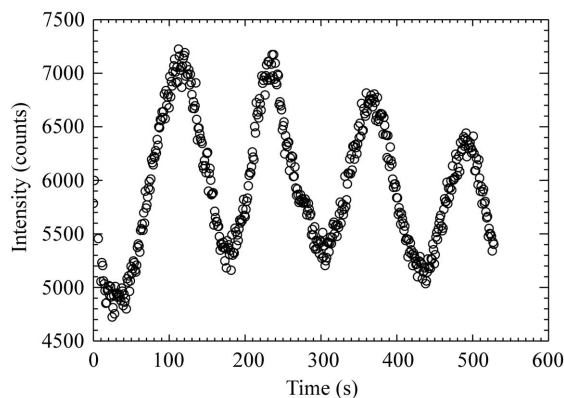


Figure 2

Intensity oscillations for the homoepitaxial deposition of SrTiO₃. Clearly growth proceeds in a layer-by-layer fashion. Upon completion of a monolayer the intensity goes to a maximum. Owing to surface roughening, the intensity decreases.

chosen. By use of two rotary feedthroughs in the cover lid, the target spindle and shutter axes are led into the chamber. The target is rotated during deposition in order to avoid hitting the same position with each laser pulse. Prior to deposition the shutter is placed between the substrate and target in order to clean the top-most layer of the target by firing the laser a number of times without depositing material on the substrate. The heating element consists of a coil-shaped thermocoax resistor wire embedded in a Ni heater block, which is non-reactive in an oxygen environment. The substrate is attached to the heater, using Cerastil ceramic glue. At 1073 K and ambient pressure the total power used by the heater is 85 W. Under these demanding conditions the chamber was tested for thermal loss, showing that the cover lid reaches a temperature of 333 K while the other walls only heat up to 313 K.

3. Applications

The chamber has been used in combination with an excimer laser (KrF 248 nm, Lambda Physik EMG 103) for the homoepitaxial

deposition of SrTiO₃. The laser beam is focused down to about 3 mm² by a lens, resulting in a laser fluence of 2 J cm⁻² on the target. X-ray diffraction experiments, using a wavelength of 0.775 Å, were performed *in situ* by mounting the chamber on a 2+3-type surface diffractometer (Vlieg, 1998) on BM26 (Dubble) (Borsboom *et al.*, 1998) at the European Synchrotron Radiation Facility. Fig. 2 shows the measured intensity oscillations of the specularly reflected beam at an angle of incidence of 1°. Each maximum in Fig. 2 corresponds to the completion of another monolayer of the material, and this kind of oscillation is a clear signature for layer-by-layer growth.

4. Conclusion

A sample chamber for the purpose of studying the PLD process *in situ* with surface-sensitive X-ray scattering has been successfully tested and implemented at a synchrotron beamline. The very first results show that it is possible to follow the evolution of the SrTiO₃(001) surface, during layer-by-layer growth up to many monolayers. This opens the possibilities to carry out detailed studies of crystal growth using PLD and to profit from the resulting quantifiable data using a kinematical description of scattering.

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