	Experiment title:	**Experiment number:**
DUBBLE	**Towards *in-situ* monitoring of the PLD process by synchrotron X-rays** ***In-situ monitoring of the PLD process by synchrotron X-rays Study of the formation of thin layers by interface diffraction methods.***	**26-02-292**
Beamline: BM26	**Date(s) of experiment:** From: 08-11-2005 To: 15-11-2005	**Date of report:** 25-11-2005
Shifts: 18	**Local contact(s):** W. Bras	

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Report:

An important class of oxidic materials is formed by the perovskites: complex transition metal oxides. Depending on composition, this class of materials includes itinerant and local ferromagnets, high Tc superconductors, ferroelectrics, insulators, semiconductors and half-metallic magnets. In view of the technological importance of these compounds and especially of thin layers of these materials, they are extensively studied in our group.

$SrTiO_3$ (001) substrates are widely used in thin film growth of related oxide materials by Pulsed Laser Deposition (PLD).

The PLD process can be monitored by high pressure Reflection High Energy Diffraction (RHEED). The RHEED method, however, only probes the topmost layers. Furthermore, due to the strong interaction, the theoretical interpretation of the result is complicated. When using (synchrotron) X-rays the periodicity is probed on a much larger scale, making the method less sensitive for contaminations. The theoretical interpretation (kinematical theory) is much simpler. Therefore, we started a project to combine PLD and surface diffraction by means of synchrotron X-rays to *in-situ* monitor intensity oscillations during PLD and to study the thin (few unit cell) layers produced this way.

Earlier experiments of this project were 26-02-129,157, 224, 248 and 271.

In our previous runs, we have succesfully tested and implemented the sample chamber, which is designed especially for use on the Dubble interface diffractometer. Intensity oscillations in the specularly reflected X-ray beam were observed during the deposition of complex oxides , which indicates that with the present set-up it is possible to grow and study layer-by-layer growth. In the present proposal the main aim was to stop deposition at a well defined coverage and collect data in the form of several crystal truncation rods (CTR's), in order to get information on the interfacial atomic structure.

During the first day the set up was completed and the optics were aligned to obtain a suitable X-ray beam at the sample position. We obtained a positionally very stable 0.5x0.35 mm^2 beam, with a flux of about $5x10^{10}$ photons/s (which is about a factor of 5 better than in our last run!). Although the specifications of the optics should provide sufficient higher harmonic suppression, we still found non-negligible contamination of $\lambda/3$ and $\lambda/4$ in the diffracted signals. Apparently the angle of incidence of the X-ray beam with the mirror is too low.

This is quite a nuisance when searching for fractional order reflections, but fortunately we can work around the problem.

With the final set-up, five deposition runs using different substrates and targets were carried out. All substrates were single TiO_2-terminated STO single crystals. The targets were sintered pellets of either $LaAlO_3$ (LAO) or $PbTiO_3$ (PTO).

By carefully monitoring the specular intensity during deposition, it is possible to stop after one unit cell layer has been grown. Figure 1 shows the intensity of the reciprocal point (0,0,0.5) during the initial deposition of LAO on STO. The unchanged intensity in the first 180 seconds is coming from the bare STO surface before the laser is switched on. The intensity after completion of the monolayer (around 300 seconds) is higher , because a monolayer of LAO scatters more than a monolayer of STO. The parabolic shape inbetween the start of the deposition and completetion of the LAO monolayer is characteristic for layer-by-layer-growth, which is governed by island formation and subsequent coalesence of these islands. The solid line in figure 1 is a simulation of the scattered intensity of a TiO_2-terminated substrate, on which a LAO layer is grown, assuming a constant growth rate. The stacking at the interface is bulk-TiO_2-LaO-AlO_2 , which is also expected based on the bulk structres of both materials. Furthermore, in the simulation the LAO unit cell layer was extended by 11% in the growth-direction and a final coverage of 80% was used. The implications of these results are presently being worked out, based on the full data set consisting of several crystal truncation rods (CTR's) that were obtained from this interface structure.

In another experiment, we deposited PTO on STO. The initial growth showed a behaviour similar to that observed in the previous experiment. Again here a complete data set consisting of several CTR's was collected. In order to monitor the stability of the surface/interface, a surface sensitive reflection was measured several times during the final data collection that lasted about 8 hours. Figure 2 shows the peaks, measured at 570 °C, and since they are practically identical, the interface can be assumed stable and unchanged during the measurements.

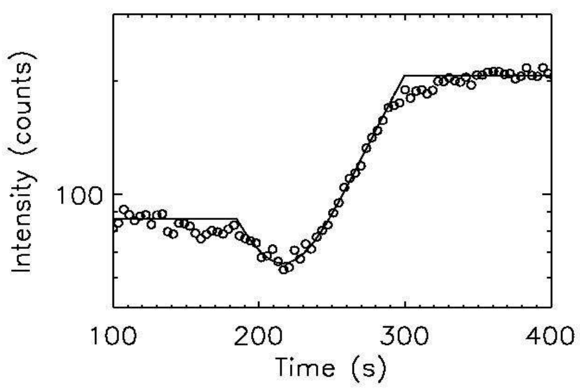

Fig.1 The intensity of the reciprocal point (0,0,0.5) during initial depostion of LAO on STO at 850 °C. After starting the laser for deposition, due to roughening the intensity decreases, and finally upon completion of the layer the surface is smooth again, hence a maximum. The solid line is a simulation, which is explained in the text.

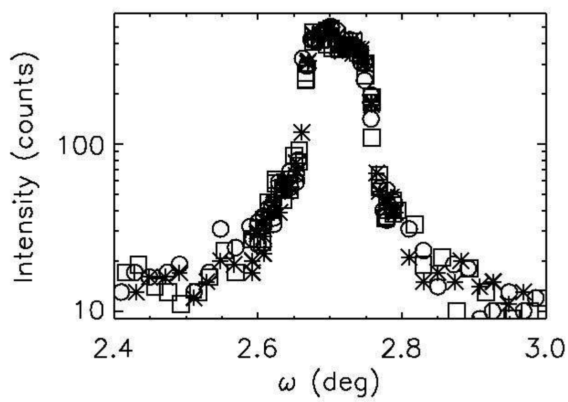

Fig. 2 The anti-Bragg peak (0,0,0.5) of a monolayer PTO on STO. Shown are three times the same peak (asterisks-circles-squares), measured a few hours apart during data collection at 570 °C of several CTR's. Clearly the peaks are almost identical, which is an indication that the surface/interface was stable during the data collection.

In conclusion, we have succeeded in obtaining full surface crystallographic data sets of the interface structure of the LAO/STO and PTO/STO systems. These will be very helpfull for clearifying the unknown atomic interface structures, which govern to a great extent the physical properties. From an instrumentation point of view, this run has been very succesful. The intensity and stability of the X-ray beam was excellent. The hexapod worked almost without any failure, and the systems under investigation proved stable at deposition conditions for several hours. This all resulted in 5 deposition runs that were performed on two different systems, the results of which are presently being processed.