DUBBLE	**Experiment title:** *In-situ* monitoring of the PLD process by synchrotron X-rays: PTO on $DyScO_3$	**Experiment number:** 26-02-460
Beamline: BM26	**Date(s) of experiment:** From : 10-11-08 To : 17-11-08	**Date of report:** 05-01-09
Shifts: 18	**Local contact(s):** G. Portale	
Names and affiliations of applicants (* indicates experimentalists):		

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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 ferro-magnets, 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.
The preferred technique for the growth of these thin films is 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, PLD and surface diffraction is combined 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 26-02-271, 292, 309 and 405.

Previous experiments showed the possibility to study layer-by-layer growth of several complex oxides ($PbTiO_3$ on $SrTiO_3$, $SrTiO_3$ on $DyScO_3$), using X-rays.
During this experiment the system $PbTiO_3$ (PTO) on $DyScO_3$(110) (DSO) was investigated. The novel substrate DSO has special properties (e.g. a larger in-plane lattice constant) as compared to other commonly used substrates, creating the possibility to obtain for example tensilely strained ferroelectric phases of extremely thin films.
The thin film needs to be grown at elevated temperature (550^{o}C). Due to the difference in thermal expansion coefficient, the crystal lattice of the substrate and film material at this temperature match and a good epitaxial film is grown. When the substrate and film are cooled down, the difference in thermal expansion coefficient creates strain in the film.

The first 3 shifts in this experiment were used to align the beam and start-up (including maintenance and reparation) of the diffractometer. The following MDT day was used for mounting/aligning the PLD chamber and 2 more shifts were used for the alignment of the diffactometer.

During the subsequent measurements 5 PTO thin films, ranging from ½ to 8 monolayers where deposited and Crystal Truncation Rods were measured. These structural datasets were taken at deposition and room temperature.

The growth of the thin film was monitored by measuring scattered intensity at the “anti-Bragg” position (0 0 ½). Growth oscillations can be seen in figure 1 and 3: each half oscillation corresponds to one monolayer of PTO. The constant signal, after the laser is stopped, indicates a stable thin film. Subsequent measurements of e.g. the specular rod (fig. 2) show an epitaxially strained film, the one extra fringe in the rod profile confirms the deposition of one monolayer of PTO.

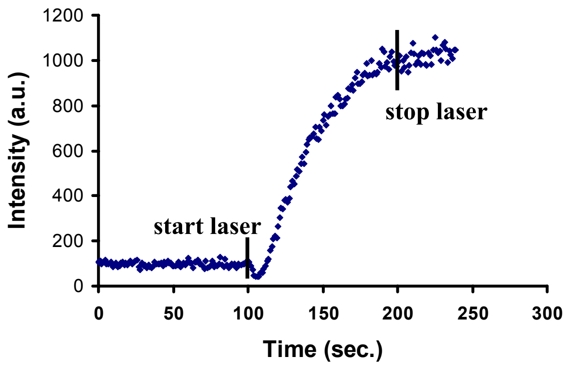

Fig. 1: Left, intensity oscillation at hkl (0 0 ½). The stabilization of the scattered intensity just before laser is stopped indicates the formation of the first monolayer.

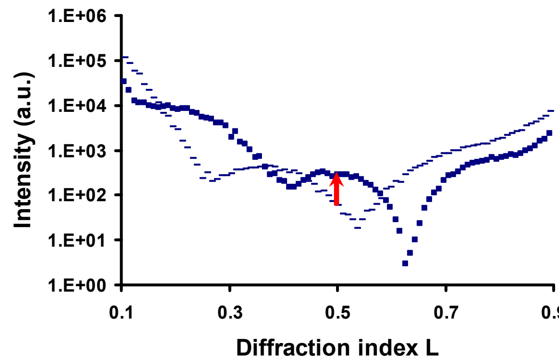

Fig 2: Specular rod of the bare DSO substrate (dashed) and the substrate with the deposited PTO thin film of one monolayer (squares). The arrow indicates the increase of scattered intensity seen in figure 1.

During deposition of one of the films a rather unusual oscillation pattern was observed (fig 3). The signal does not drop down at the completion of the 3rd monolayer, but continues to rise and reaches a maximum at the formation of the 4th monolayer. The measurement of the specular rod (fig 4) confirms that 4 monolayers have been grown on the substrate in an orderly fashion. The oscillation pattern of figure 3 could be due to a change in growth mode, which causes a change in oscillation pattern.

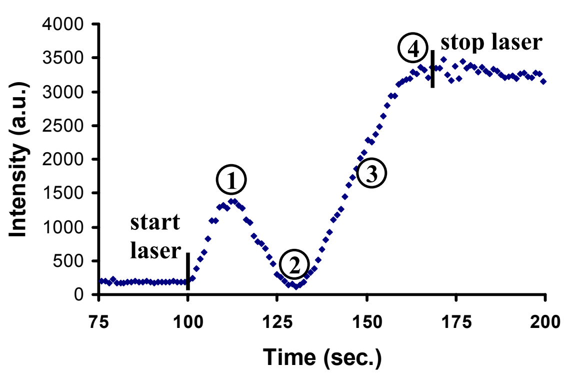

Fig 3: Intensity oscillation observed during the growth of 4 monolayers of PTO on DSO. The numbers indicate the number of monolayers completed.

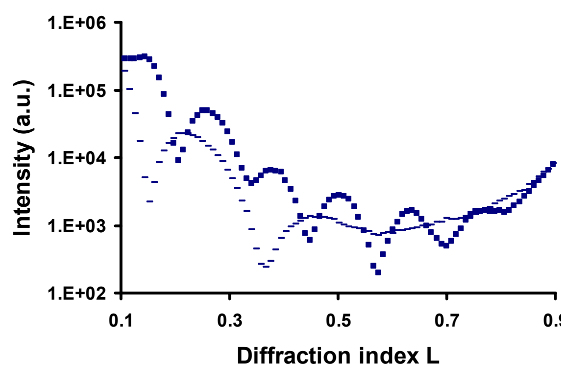

Fig 4: Specular rod of the bare substrate (dashed) and the substrate with film (squares). The presence of clearly distinguishable fringes indicates a well grown smooth film.

Despite the identical preparation procedure of the substrates, not all bare substrates have the same scattering pattern (dashed lines, fig. 2 and 4). They fall into two categories: 4 or 5 monolayers of relaxed bulk material and/or oxide layers. The collected data (crystal truncation rods) are being analysed at this moment.