How to avoid atomic sandpaper

When growing thin films of novel materials, smooth surfaces are a must. How else could one stack them layer by layer, as needed in optical coatings, sensors or conductors? One method known to produce atomically smooth films is Pulsed Laser Deposition (PLD).

In PLD, a pulsed laser beam hits a bulk target. With every pulse it creates a jet (or “plume”) of high energy atoms from the target. When these condense on a smooth substrate they recreate the target material, but now as a thin film. Per laser pulse only about 1/100th of an atomic layer is deposited on the substrate.

Although the fact that PLD produces atomically smooth films has long been known, the reasons proposed for this were somewhat speculative. A recent experiment measured the roughness of PLD films, carefully adding one laser pulse after another. This provided the clue to the puzzle: When the first atoms land on the smooth substrate they form small islands of the material. Subsequent pulses lead either to formation of more small islands if the particles land on an uncovered part of the monolayer, or, importantly, cause already-formed islands to break up if the energetic particles land on top of them. This avoids growth of "islands on top of islands", which would gradually result in rough films - that is, atomic sandpaper.

Now that the process in understood one can optimally tune PLD for growing thin films of materials which are then used in devices.

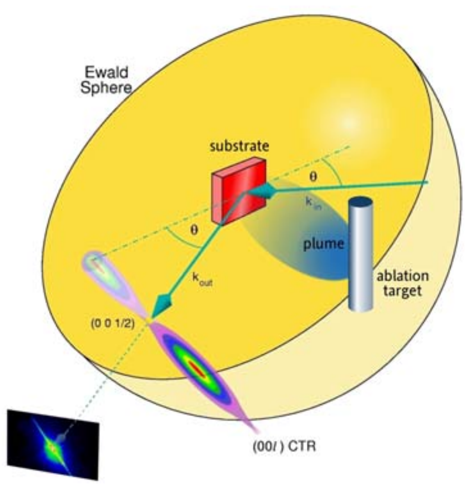

Figure 1: *Sketch showing the ablation target with the plume recondensing on the substrate. The incoming x-ray beam is reflected by 2θ and produces a crystal truncation rod (CTR) when intersecting the Ewald sphere.*

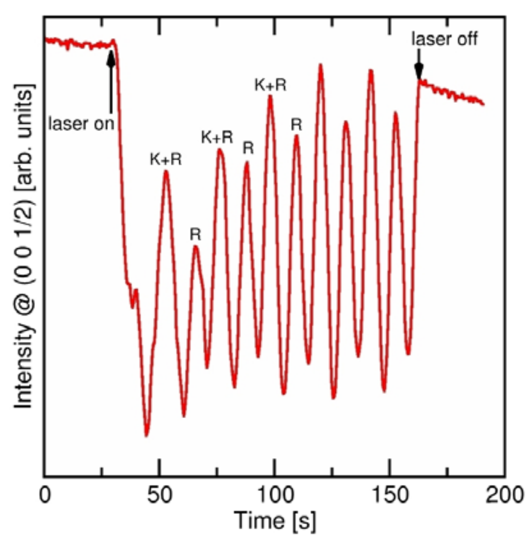

Figure 2: *Intensity of the(0 0 ½) CTR as a function of time. The oscillations are due to changes in the roughness of the growing film (R) and interference between substrate and surface (K).*

The experiments were carried out at the Surface Diffraction station of the Materials Science (MS) beamline of the Swiss Light Source. The film roughness can be measured while continuously sending laser pulses as in Figure 2. This causes oscillations of the signal which can be used to count the number of atomic layers deposited.

When shooting only controlled bursts of laser pulses (17 pulses followed by several seconds dwell time) as in Figure 3, one observes the formation of a new film layer. After about 200 s half an atomic layer has formed and the reflected light reaches a minimum. After 600 s the full layer has been deposited and the reflectivity reaches the old value. Between two bursts of laser pulses the atoms rearrange on the surface causing a change of the reflectivity (red lines in Figure 3).

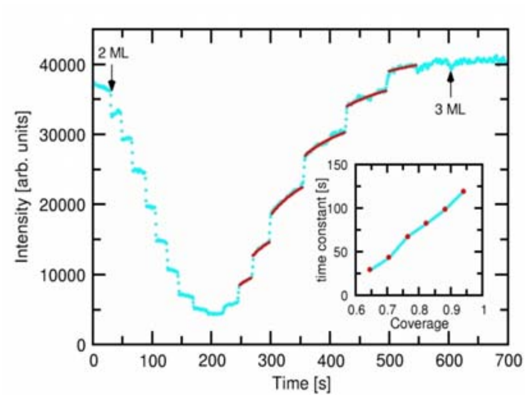

Figure 3: *Evolution of the reflected x-ray intensity during 17 laser bursts, showing maximum roughness (Time = 200s) and subsequent smoothing. Between two laser bursts the atoms diffuse across the growing surface to the edges of growing islands (red solid lines), thereby reducing the roughness. The change in the diffusion time constant as a function of monolayer coverage is shown in the inset.*

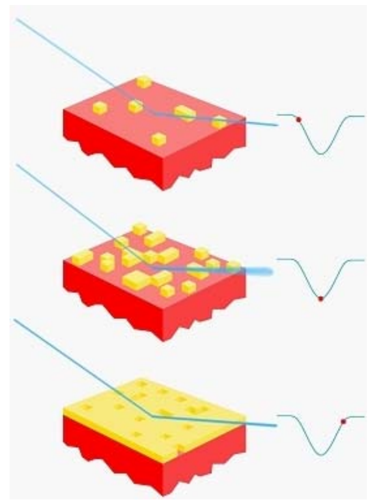

Figure 3: *Sketch showing the film (yellow) growing on the substrate (red). At about 50% coverage the roughness is at a maximum, thus the reflected x-ray intensity is minimal.*

Analyzing this relaxation with a mathematical model shows that the high particle energy in PLD breaks up islands in the film once they exceed a certain size. This inhibits growth of film material on top of an island and thus any roughness which would result in atomic sandpaper.

Publications

- **Energetic smoothing studies of complex metal-oxide thin films**
  P.R. Willmott, R. Herger, C.M. Schlepuetz, D. Martoccia, and B.D. Patterson
  Phys. Rev. Lett. 96, 176102 (2006)
  doi: 10.1103/PhysRevLett.96.176102

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