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Plasma oxidation as key mechanism for stoichiometry in Pulsed Laser Deposition grown oxide films

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Pulsed Laser Deposition (PLD) has been established in recent years as a versatile thin film deposition technique. PLD utilises the transient particle flow of a laser-induced plasma to achieve a fully controlled (also crystalline) growth of thin films of complex materials, including complex oxides. These materials exhibit a large variety of interesting physical properties, which are highly sensitive to slight deviation from ideal stoichiometry. Key mechanisms involved in optimized PLD thin film growth conditions haven't been fully understood, and better insight in the relation between growth parameters, plasma plume characteristics and film characteristics is necessary in obtaining full control over stoichiometry, doping, defect density of PLD grown thin films and herewith improved thin film properties.

We present a unique overview on the influence of growth parameters on the characteristics of the PLD plasma plume using Optical Self-Emission (OSE) imaging and spectroscopy, supported with Laser Induced Fluorescence (LIF) measurements. It is shown that in a relatively small background gas pressure regime, from 10^{-2} mbar to 10^{-1} mbar oxygen pressure, a transition from nonstoichiometric to stoichiometric growth of SrTiO₃ films occurs as measured with X-ray Diffraction (XRD). In this pressure regime, OSE spectroscopy and LIF measurements also show a transition from incomplete to full oxidation of species in the plasma plume. This suggests that the oxidation of species in the plasma is a crucial mechanism for the stoichiometric reconstruction of the synthesised oxide thin films.