Adsorption-Controlled Growth of Perovskite Oxides for Optimized Opto-Electric Properties

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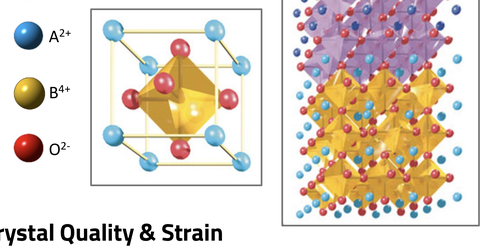
Introduction

Perovskite Oxides

- Exhibit tunable opto-electric properties
- Useful for many applications such as quantum computing and solar cells[1,2]

ABO_3 Perovskite Oxides[3]



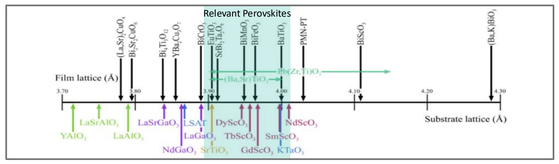
Crystal Quality & Strain

- High crystal quality is essential for leveraging these properties
- Films are grown on crystalline substrates to induce ordered growth
- Film strain critical in optimizing properties
- Film strain controlled by adjusting film and substrate lattice mismatch

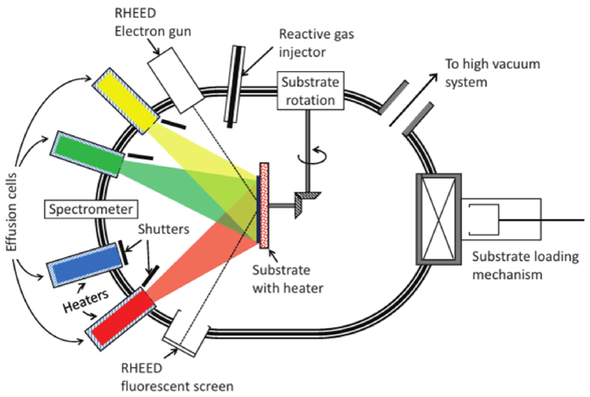
Barium Strontium Titanate - $Ba_xSr_{1-x}TiO_3$ (BST)

- Cubic perovskite with tunable lattice constants (3.9-4.0 Å) based on Ba:Sr ratio (x)
- Useful as a pseudo substrate for fine-tuning strain in perovskite films
- Can reduce costs of high-quality perovskite film production by replacing expensive substrates

Substrates Suitable for Films Based on Lattice Match[4]



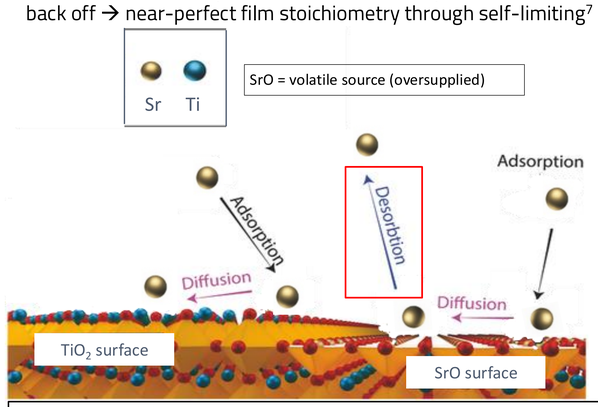
Molecular Beam Epitaxy[5]



Molecular Beam Epitaxy (MBE) Growth in Adsorption-Controlled Windows

Adsorption-Controlled Growth Mechanism[6]

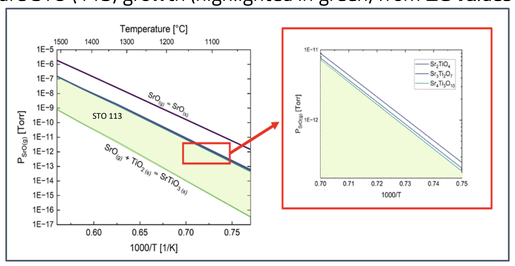
- One material more volatile than the rest
- Volatile material is oversupplied
- Volatile material incorporates stoichiometrically & excess desorbs back off → near-perfect film stoichiometry through self-limiting[7]



SrTiO3 (STO) (113) was grown as a baseline for BST growth (BST with x = 0)

Adsorption-Controlled Growth Window for $SrTiO_3$ (STO)

Calculated window of temperature-pressure conditions for phase-pure STO (113) growth (highlighted in green) from ΔG values[8,9]

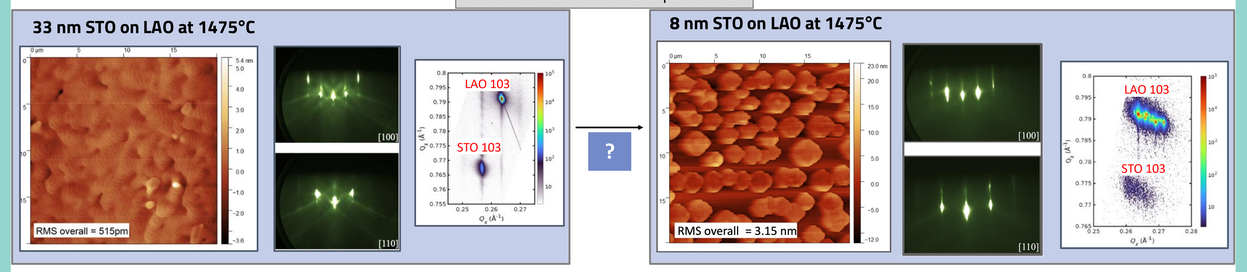


Conventional substrate heaters: up to ~1000°C
PARADIM laser heater: up to 2000°C

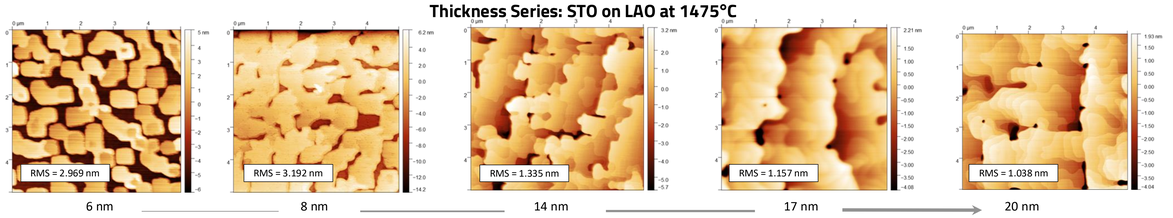
- Phase-pure STO (113) observed between 1425-1525 °C
- Best samples at 1450 °C & 1475 °C
- Impurities observed outside or on edge of highlighted growth window

Effects of Film Thickness and Growth Temperature on STO Nucleation

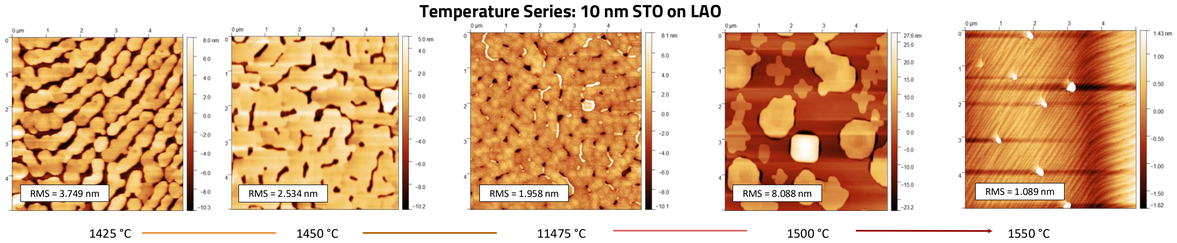
Sr:Ti flux ratio 5:1 for all experiments



Thickness Series: STO on LAO at 1475°C



Temperature Series: 10 nm STO on LAO



Conclusions & Future Work

STO Nucleation

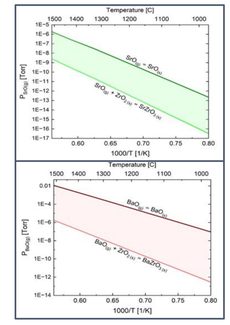
- STO nucleates from islands, merging around 1475°C.
- Steps form when the film thickness reaches approximately 14 nm.
- Impurities observed outside or on edge of highlighted growth window
- Further research: explore STO nucleation at different growth rates

BST Pseudo Substrates

- A combined understanding of STO and BaTiO3 (BTO) can be applied for BST pseudo-substrate growth.

Predictive Modeling

- Ellingham-esque diagrams successfully predict the phase-pure growth window for STO (113).
- Similar calculations applied to zirconates and lead zirconium titanate (PZT).
- This approach can be expanded to other systems for experimental exploration.



Works Cited

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[3] Figure adapted from Brahlek, M, et al. Adv. Funct. Mater. 2018, 28 (9), 1702772.
[4] Figure adapted from Schlom, D. G.; Chen, L.-Q.; Pan, X.; Schmehl, A.; Zurbuchen, M. A. J. Am. Ceram. Soc. 2008, 91 (8), 2429–2454.
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