Investigation of the Pb depletion in Single and Dual Pulsed Laser Deposited Epitaxial PZT Thin films and their Structural characterization

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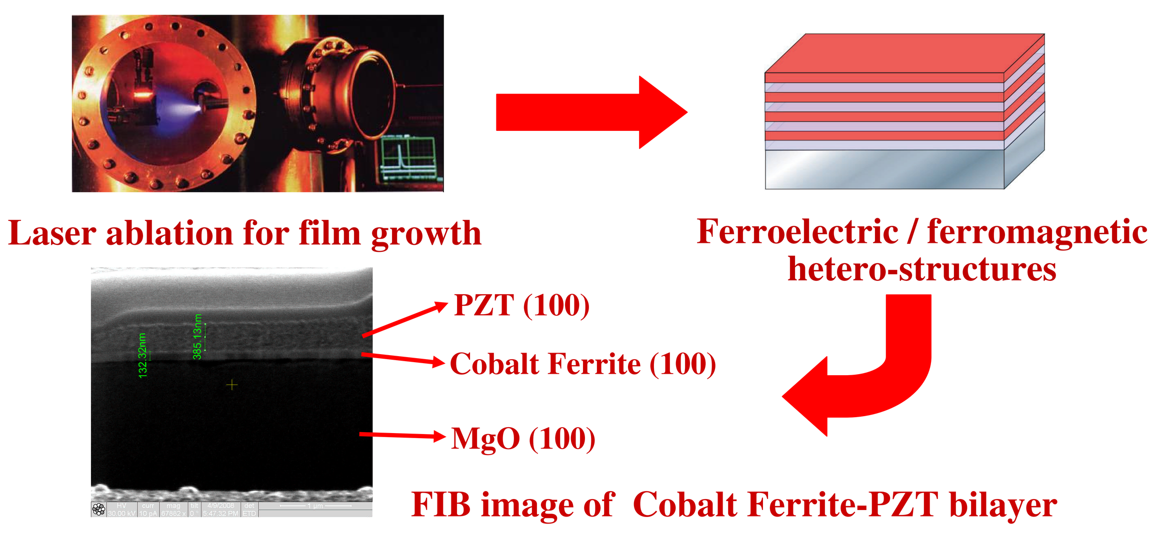
Introduction

Motivation: Increasing demands for high-quality defect- free epitaxial lead zirconium titanate $PbZr_{0.52}Ti_{0.48}O_3$ (PZT) thin films in present market

Applications : PZT thin films are successfully used in

- non-volatile random access memory devices (FeRAM)
- micro actuators and ultrasonic sensors

NEW Pulsed laser deposition (PLD) to grow multi- layered PZT based hetero-structures for multi-ferroic device applications



Advantages of Pulsed Laser Deposition technique :

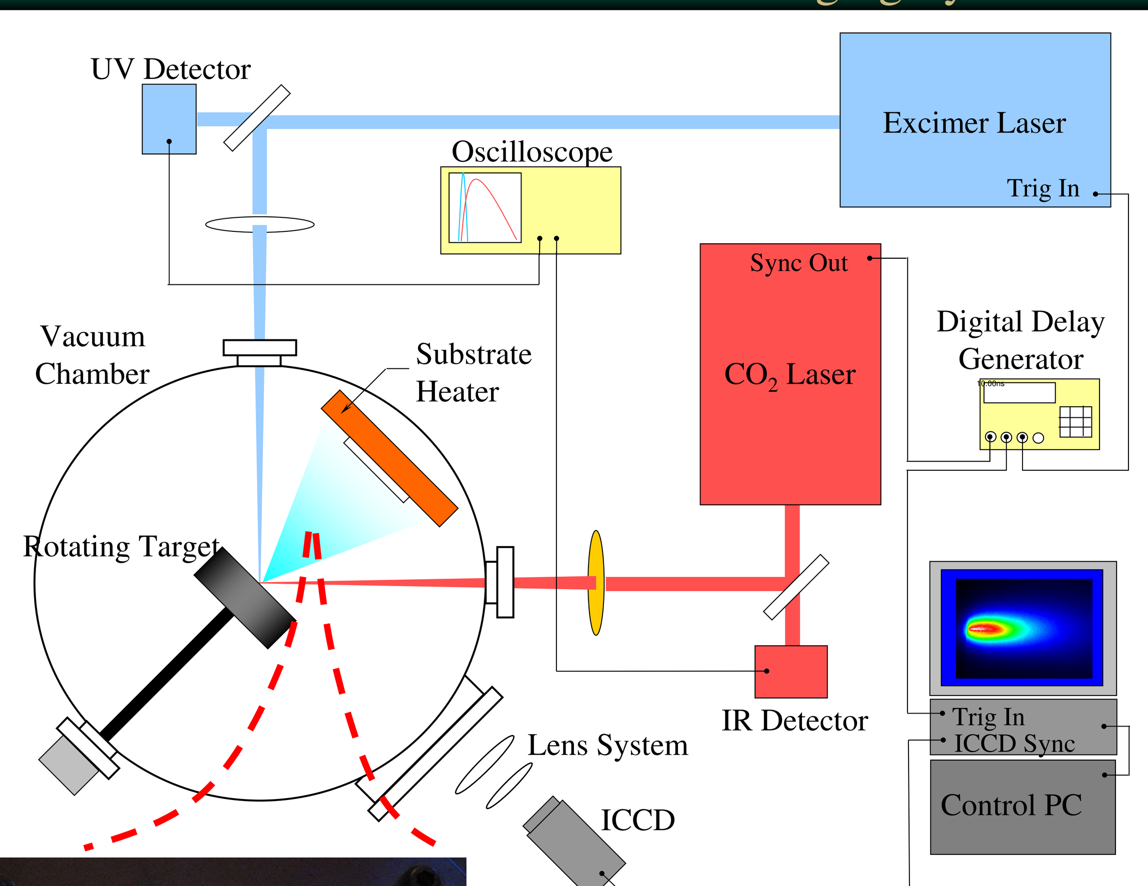
- Stoichiometric transfer of materials from target to films
- Fast and versatile method for in-situ hetero-structure growth
- Plume of highly energetic materials have sufficient ion mobility for the growth of epitaxial thin films

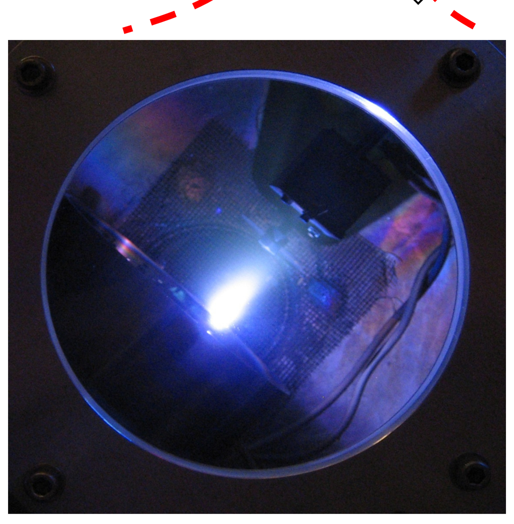
Problem !!
High volatility of Pb in PZT leads to Pb depletion in deposited films lowering ferroelectric properties.

Solution !!
Dual laser ablation to produce high Pb content films with less particulates using low excimer energy.

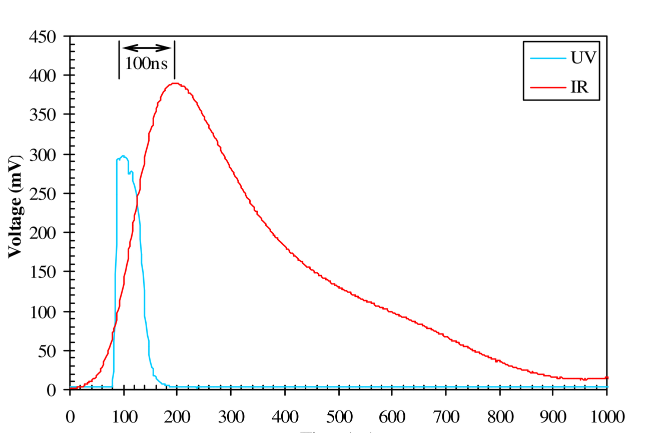
Experimental Details

Dual-Laser PLD with the ICCD Imaging System





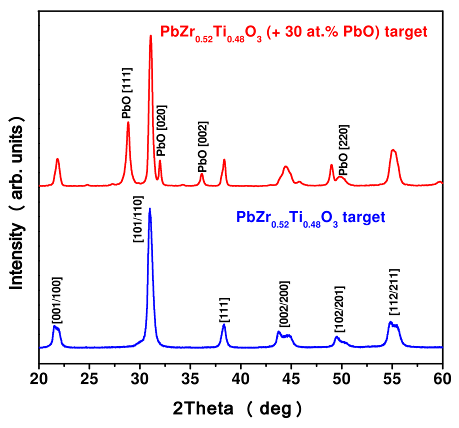
Photograph of laser ablated plume

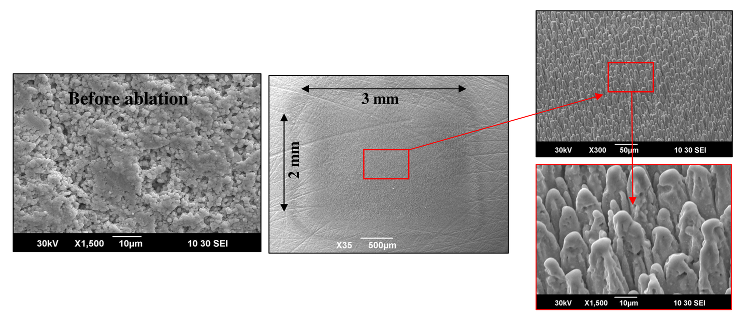


The time-resolved oscilloscope traces of the excimer and CO_2 laser pulses from the UV and IR detectors.

Results

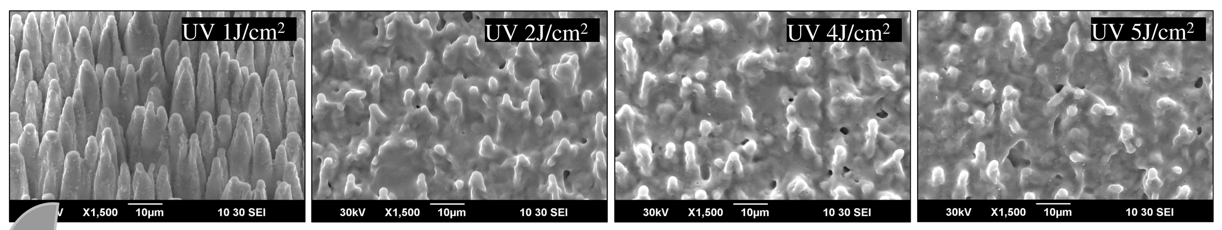
Laser-Target Interactions



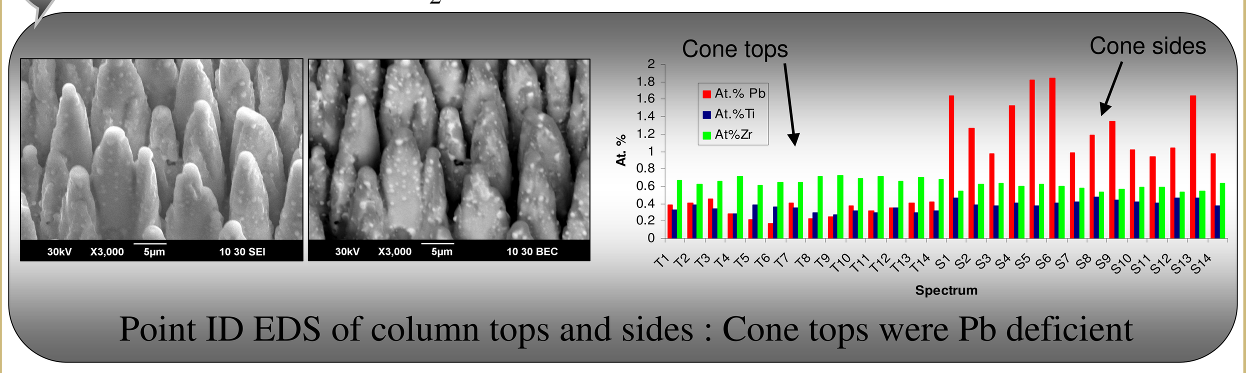


(Left) XRD data for the PZT targets.
(Top) SEM images of typical PZT target surface before/after irradiation by laser.

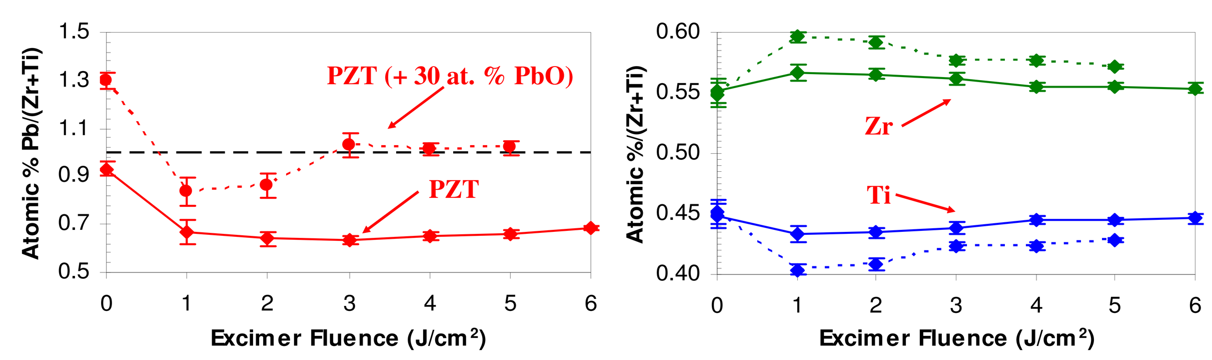
Single Laser Ablation



SEM images of a PZT target surface after irradiation by 1000 pulses of a KrF laser beam in 500 mT O_2 ambient at different UV fluences.

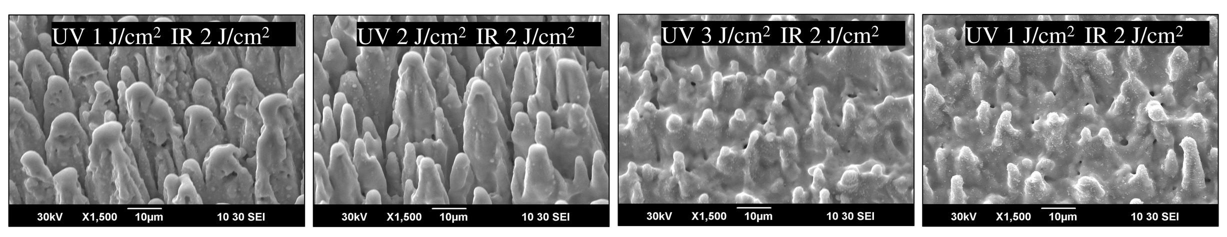


Point ID EDS of column tops and sides : Cone tops were Pb deficient

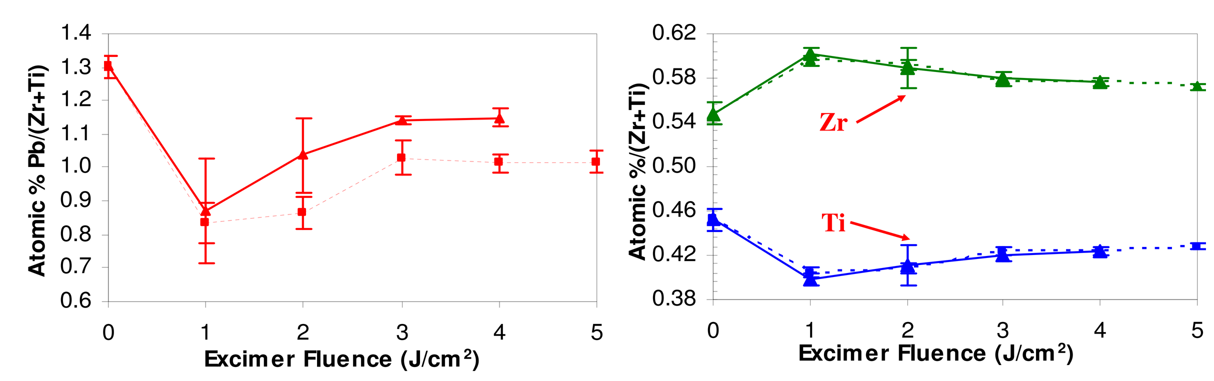


Chemical compositions using EDS analysis of the ablated target surfaces versus fluence. **(Single laser ablation)**

Dual Laser Ablation

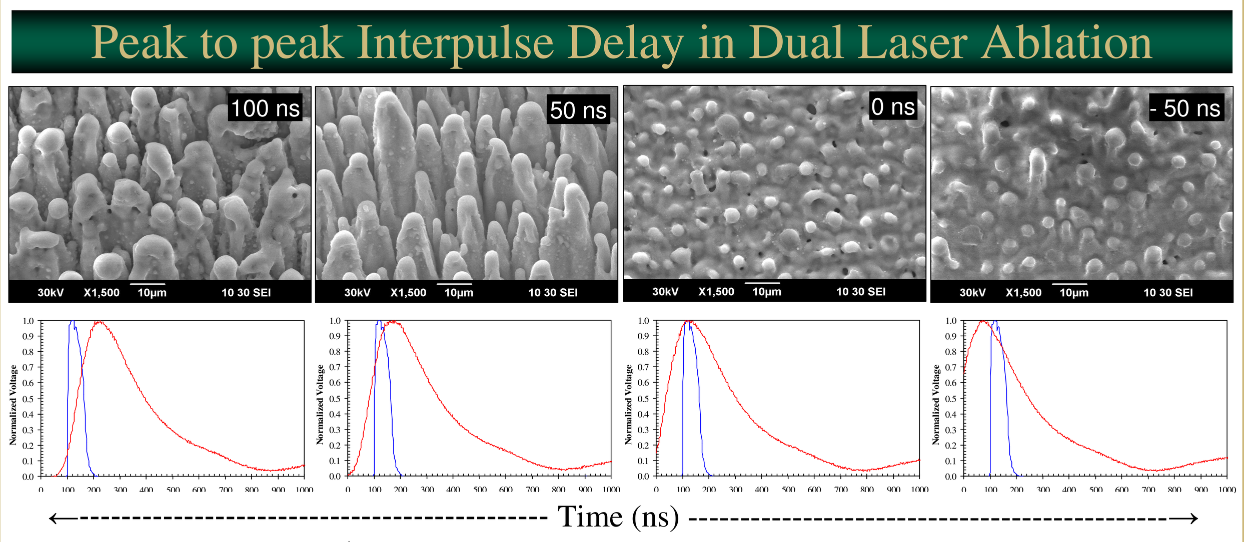


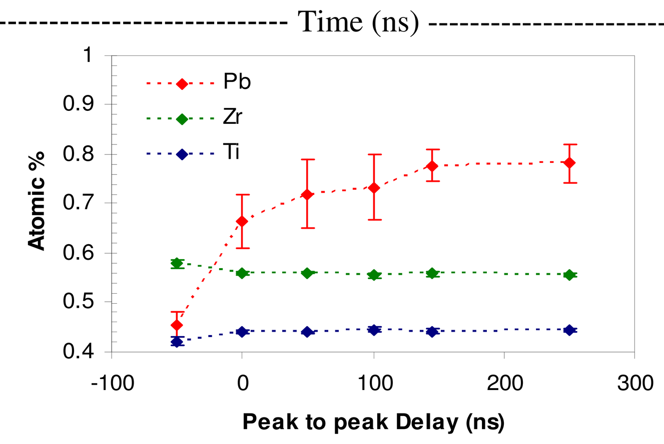
SEM images of irradiated target surfaces after 1000 pulses using dual laser ablation for different excimer fluences and 2 J/cm² CO_2 laser and **100 ns** peak to peak interpulse delay in 500 mT O_2 ambient.



Chemical compositions using EDS analysis of the ablated target surfaces versus fluence. **(Dual laser ablation)**

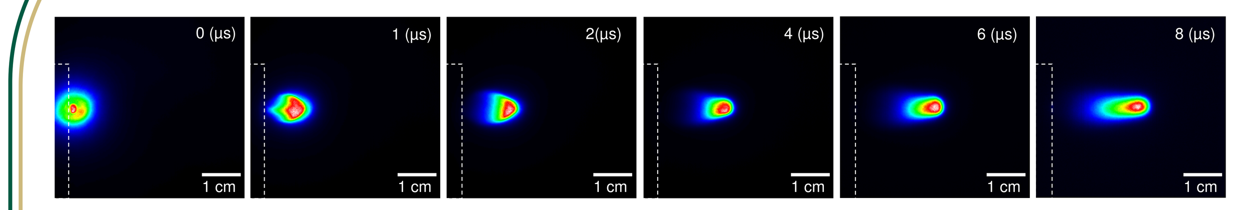
Peak to peak Interpulse Delay in Dual Laser Ablation



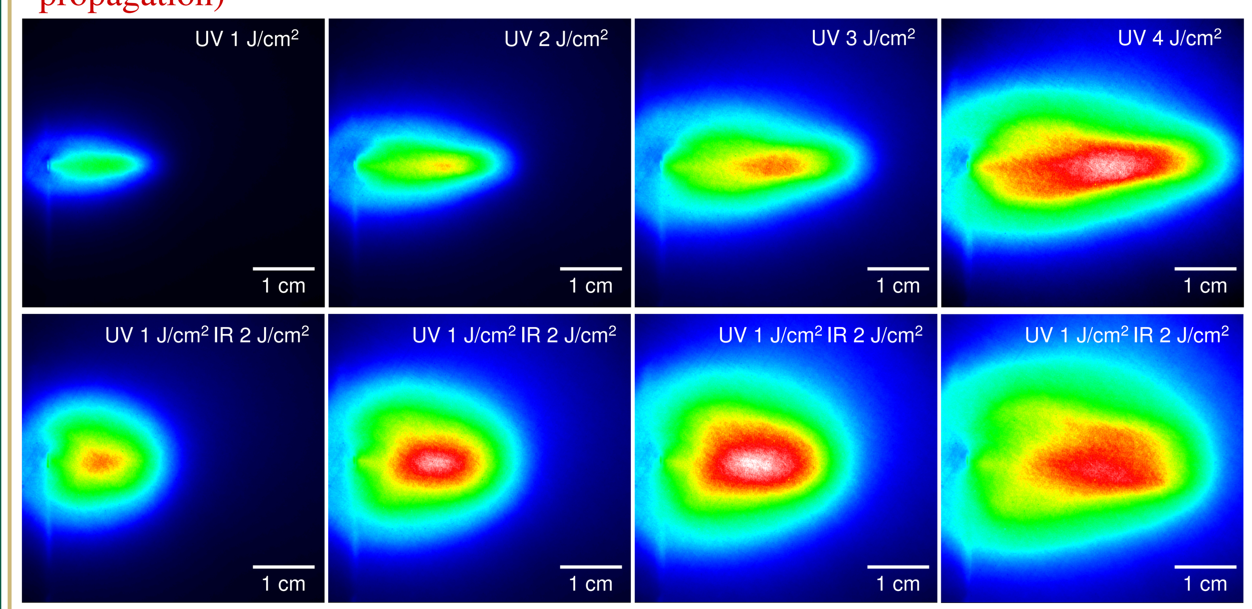


Chemical compositions of the ablated target surfaces versus peak to peak interpulse delay.

Optical-plume diagonostics

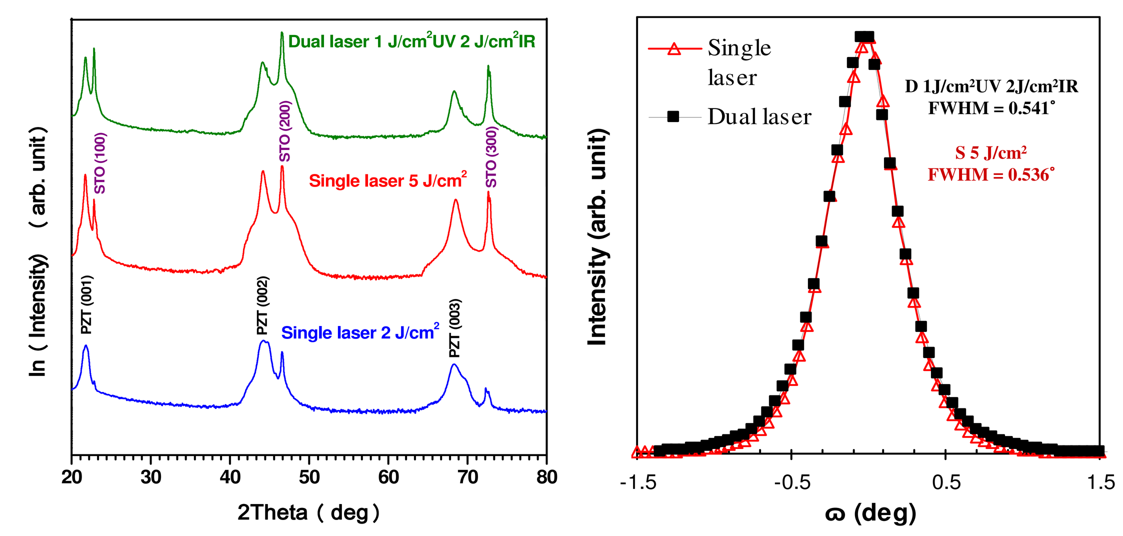


Time gated ICCD images using 200 ns exposure time of single laser ablated plumes from PZT target using 2 J/cm² fluence under 500 mT O_2 gas. (Plume propagation)

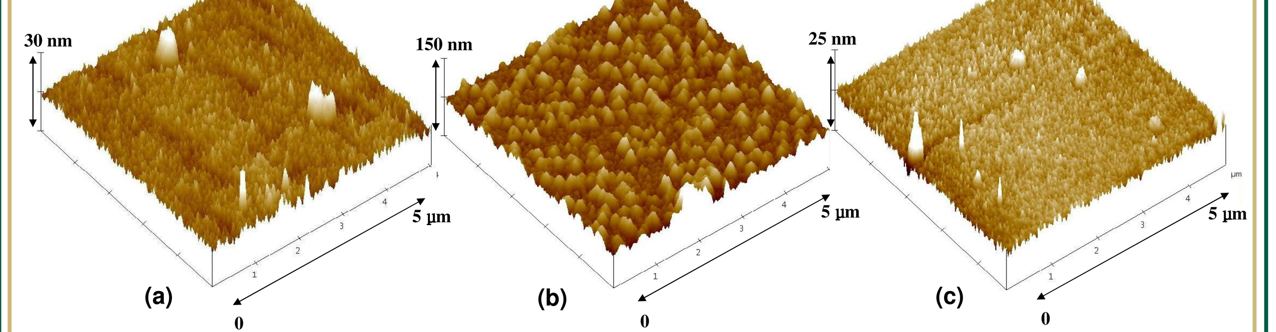


ICCD images of total visible emission spectra of single laser plumes (top row) varying the excimer (UV) fluences from 1 to 4 J/cm² and dual laser plumes (bottom row) varying excimer fluences keeping 2 J/cm² CO_2 (IR) fluence and 100 ns peak to peak inter-pulse delay

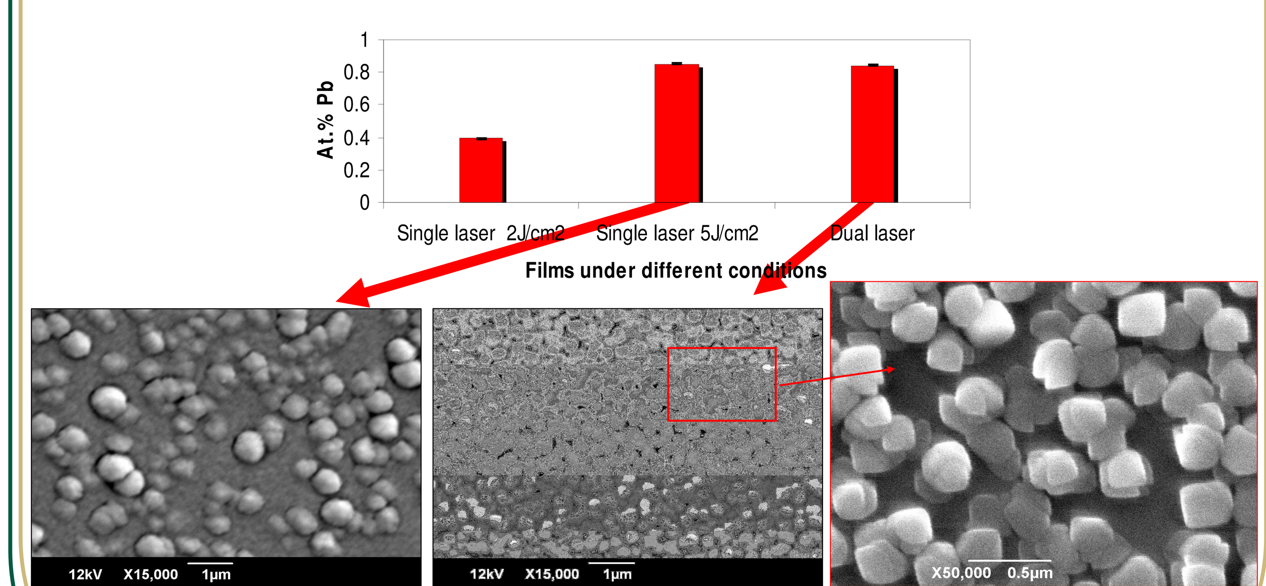
Thin film properties



XRD Θ-2Θ scans and rocking curves about PZT (200) plane for single and dual laser deposited films grown on $SrTiO_3$ (STO) (100) substrates.



AFM images of PZT films deposited using single laser fluences **(a)** 2J/cm² **(b)** 5J/cm² and **(c)** dual laser ablation. Surface roughnesses for **(a)**, **(b)** and **(c)** are 3 nm, 12 nm and 2 nm respectively.



SEM images and chemical compositions of the PZT films grown on STO substrates at 550°C and 500mT O_2 pressure using single and dual laser ablation. Film thicknesses about 350nm.

Conclusion

- Using a dual laser ablation process high Pb content, particulate free, smooth and epitaxial PZT films with the desired perovskite structure and no impurity phases were grown.
- This technique could be generalized to all multi-component thin film growth where high volatility of one of the elements leads to non-stoichiometric transfer of materials in the PLD process.

References

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ACKNOWLEDGMENTS

This work was supported in part by the National Science Foundation under the grants DMI-0217939, DMI-0078917 and by the Department of Defense under the grant W81XWH- 07-1-0708.

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