## Phase diagrams and interfacial properties exploration using combinatorial thin film synthesis

J. Wolfman<sup>1</sup>, A. Ruyter<sup>1</sup>, B. Negulescu<sup>1</sup>, N. Jaber<sup>1</sup>, C. Daumont<sup>1</sup>, C. Autret-Lambert<sup>1</sup>, P. Andreazza<sup>2</sup>, X. Wallart<sup>3</sup>

- 1. GREMAN, UMR7347 CNRS, Université François Rabelais de Tours
- 2. ICMN, UMR 7374 CNRS, Université d'Orléans
- 3. IEMN, UMR CNRS 8520, Villeneuve d'Ascq

Transition metal oxides with perovskite structure present a large panel of physical properties driven by a subtle equilibrium of competing interactions between charges, spins and crystalline structure. This results in an extreme sensitivity of physical properties to cationic substitutions, making desirable the fine and systematic exploration of phase diagrams to identify the compounds presenting the most interesting properties. During the 90s, a combinatorial synthesis aiming at producing samples with a composition continuum was proposed to address this need. The idea was to use multi-target Pulsed Laser Deposition (PLD) to synthetize films with lateral composition gradient. A single sample then allows to characterise a complete solid solution. Although PLD is known to promote stoichiometric transfer from the target to the film, it is a highly directional deposition technique. Consequently growing homogeneous multi-cationic films from a single target in terms of thickness and composition is already challenging. The control of a lateral composition gradient using several targets appears in turn as a very complex task. Numerous work have unfortunately been published without demonstrating this control, leading to a not totally undeserved scepticism within the community regarding Combinatorial PLD (CPLD).

We have revisited the CPLD synthesis concept, modified some aspects and developed some others. In particular, a statistical approach to the compounds libraries characterisation was introduced, which comforts its statements. We have set-up a pool of local physical, chemical and structural characterisation technics allowing the reliable exploration of binary and ternary phase diagrams. Finally, we have recently implemented the CPLD for the combinatorial exploration of heterostructures interfacial properties. Various studies dealing with Ga-doped BiFeO<sub>3</sub>, the Ba<sub>1-x</sub>Ca<sub>x</sub>Ti<sub>1-y</sub>Zr<sub>y</sub>O<sub>3</sub> system and the La<sub>1-x</sub>Sr<sub>x</sub>MnO<sub>3</sub>/SrTiO<sub>3</sub> interface will illustrate the effectiveness of CPLD approach.



Figure 1: Example of a combinatorial interface sample and its work function evolution vs interface composition.

Reference :

[1] J. Wolfman, B. Negulescu et al., 'Interface combinatorial pulsed laser deposition to enhance heterostructures functional properties' in Laser Ablation, ed. D. Yang, IntechOpen Ltd, Londres, 2020, <u>http://dx.doi.org/10.5772/intechopen.94415</u> ISBN 978-1-83968-304-6