

Ozone-assisted Molecular Beam Epitaxy for the growth of complex oxides systems

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Transition metal oxides attract an increasing interest driven by their wide range of physical properties and the opportunity to functionalize them in nanometric heterostructures for new electronic devices. Among these, the rare earth (RE) vanadates-based nanosystems have been highlighted as potential new multiferroic materials [1]. *Ab initio* calculations namely predict (i) the emergence of hybrid improper ferroelectricity in layered structures combining different cations, and (ii) the coupling of the polarization to Jahn-Teller distortions leading to a direct and strong coupling between polarization and magnetism [1]. Our objective is therefore to synthesize a multiferroic material in combining strains and interfaces engineering in $AVO_3/A'VO_3$ superlattices. In order to achieve an ultimate control of the interfaces where the ferroelectricity should emerge, the systems are grown in a Molecular Beam Epitaxy chamber with controlled ozone pressure.

The scope of this presentation is first to give an extensive presentation of this ozone-assisted MBE equipment (Fig. 1 (a)). This technique is namely rather unusual in the field of oxides growth, widely dominated by Pulsed Laser Ablation. We will emphasize the specificities of ozone MBE for the growth of oxides, compared to conventional MBE for metals and detail our specific MBE (DCA) connected to the UHV Daum instrument at IJL.

In a second step, we will present our recent results obtained on $LaVO_3$ and $PrVO_3$ thin films [3], and $LaVO_3/PrVO_3$ superlattices. The growth window (deposition temperature and ozone pressure) has been optimized to obtain highly controlled 2D growth and stoichiometry. Electronic microscopy experiments allowed the accurate determination of the atomic positions [2,3] (Fig.1 (b)) and enabled the observation of the RE atoms displacements required for the appearance of ferroelectricity. $(LaVO_3)_n/(PrVO_3)_m$ superlattices of high quality have been grown, ultimately as a stacking of one unit cell individual layers, for which first electric characterization suggests a ferroelectric behavior.

The last part of the presentation will be devoted to other examples of systems synthesized by ozone-MBE. In these later, the authors take full benefit of the Atomic-Layer-by-Layer growth by MBE to design materials at the atomic level and explore new magnetic, supraconductive, multiferroic or thermoelectric properties [4-7].



Fig. 1: (a) Picture of the IJL ozone-MBE connected to the Daum equipment at IJL (b) Local displacement of RE atoms extracted from HRTEM observation of a $LaVO_3$ epitaxial film.

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- [1] J. Varignon et al., Scientific Reports **5**, 15364 (2015)
 - [2] H. Meley et al., APL Materials **6**, 046102 (2018)
 - [3] G. Masset et al., Phys. Rev. Mat. **4**, 064417 (2020)
 - [4] T.S Santos et al., Phys. Rev. B **80**, 155114 (2009)
 - [5] A.T. Bollinger et al, Supercond. Sci. and Technol. **29**, 103001 (2016)
 - [6] J.A. Mundy et al., Nature **537**, 523 (2016)
 - [7] P. Kaya et al., ACS Appl. Mater. Interfaces **10**, 22786 (2018)