Growth of Ruddlesden–Popper $Ca_3Mn_2O_7$ thin films by Pulsed-Laser Deposition

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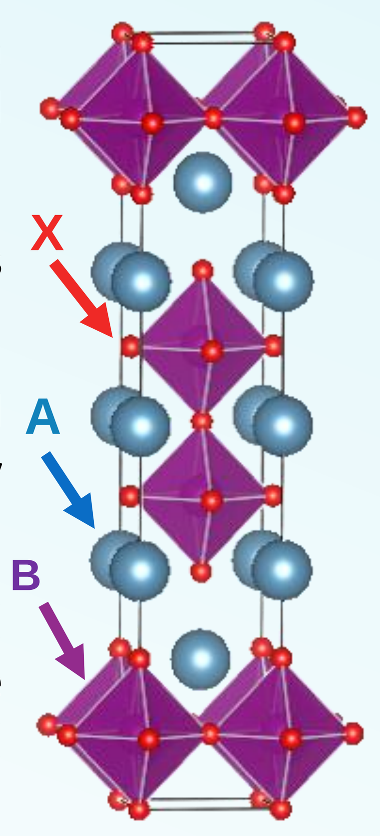
Motivation

- **Magnetoelectric multiferroic materials** have a coupling between magnetic and electrical degrees of freedom;
- Room temperature magnetoelectric compounds are rare and costly to produce;
- Improper ferroelectrics, magnetically induced by symmetry breaking due to rotation instability have a low electrical polarization;
- In multiferroic materials with great electrical polarization, there is a polar instability in the lattice, however it does not break the temporal symmetry and does not activate magnetization.



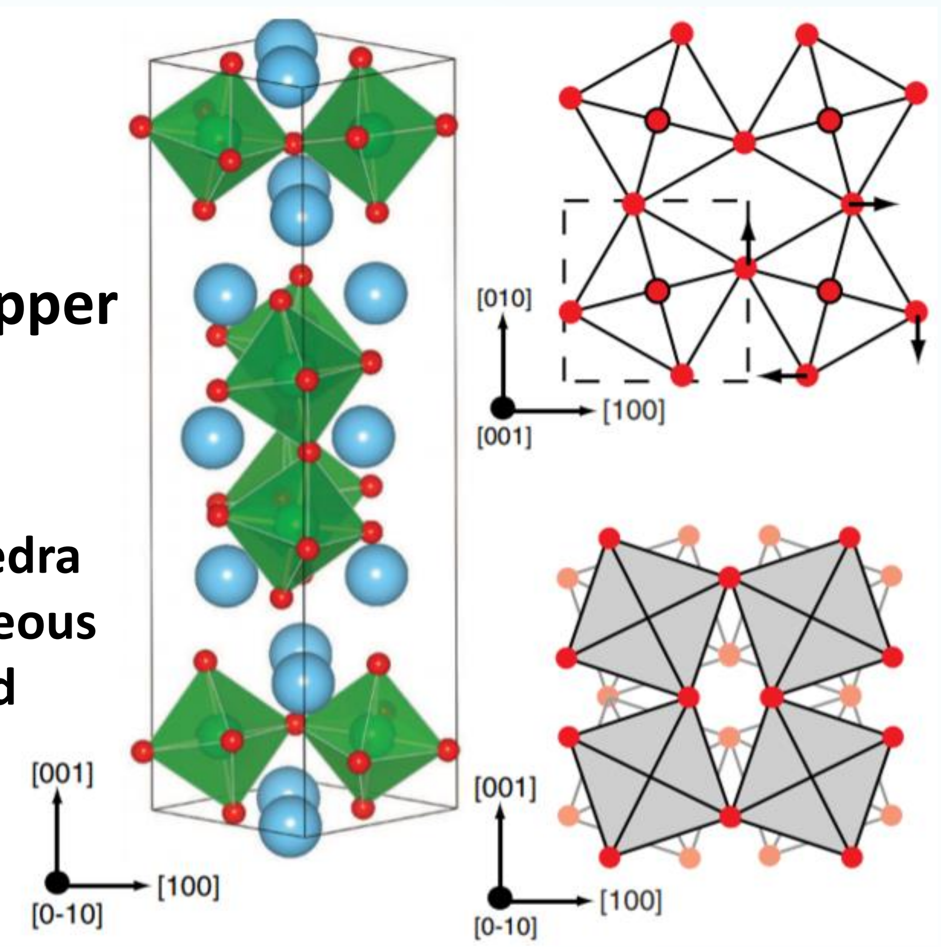
Look for mechanisms in which ferroelectricity and magnetism are defined by the same lattice instability

Materials

Candidate

Ruddlesden-Popper
$Ca_3Mn_2O_7$

Rotation of octahedra generates spontaneous polarization and magnetization



[1] Benedek N A and Fennie C J 2011 Hybrid improper ferroelectricity: A mechanism for controllable polarization-magnetization coupling Phys. Rev. Lett. 106

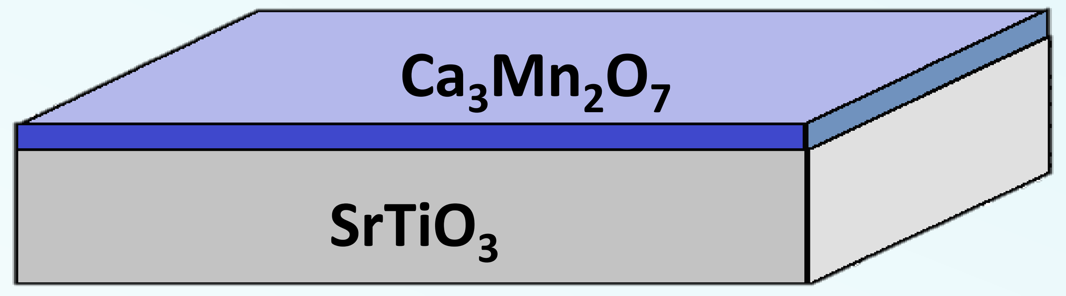
Methodology

Perovskites layered on a crystalline substrate

Interfacial deformation

Octahedrons rotation

Hybrid Improper Ferroelectric Polarization

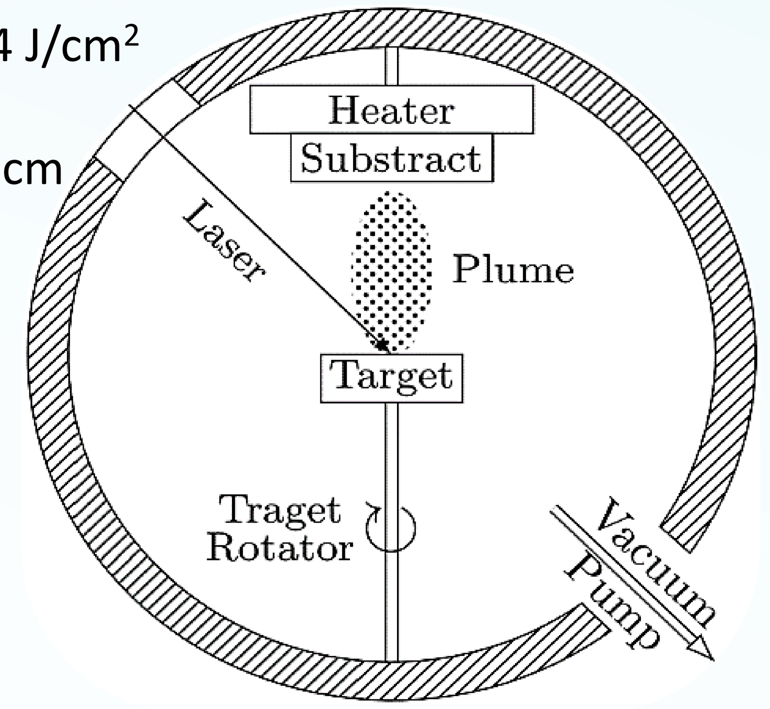


Deposition Conditions

Pulsed Laser Deposition:

- Target: $Ca_3Mn_2O_7$
- T = 700 ºC
- O_2 Pressure = 0.01 mbar
- Laser fluence: 3.4 J/cm²
- 10 Hz ($CoFe_2O_4$)
- $d_{target-substrate}$ = 6 cm

Laser ablation system:

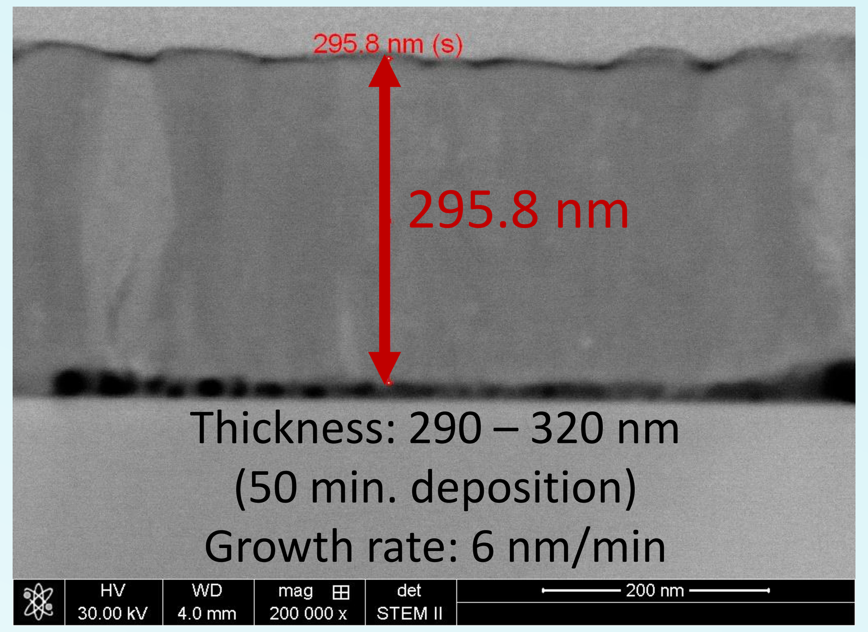


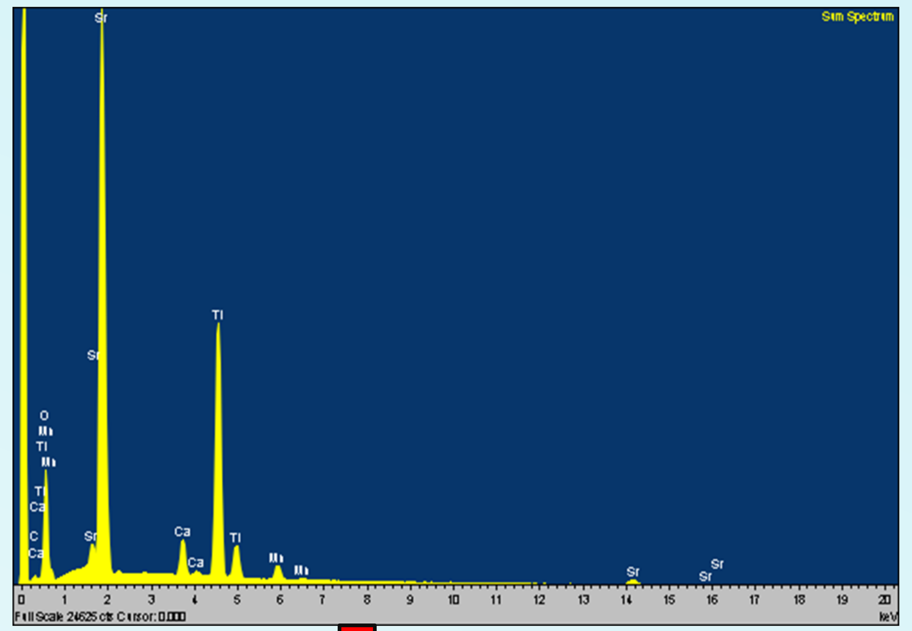
Post-annealing:

- T = 900 ºC
- t = 120 min
- Pressure: atm

[2] Oliveira, J, "Modelação de espectros raios-X em films finos", Master's thesis, Universidade do Minho, 2017.

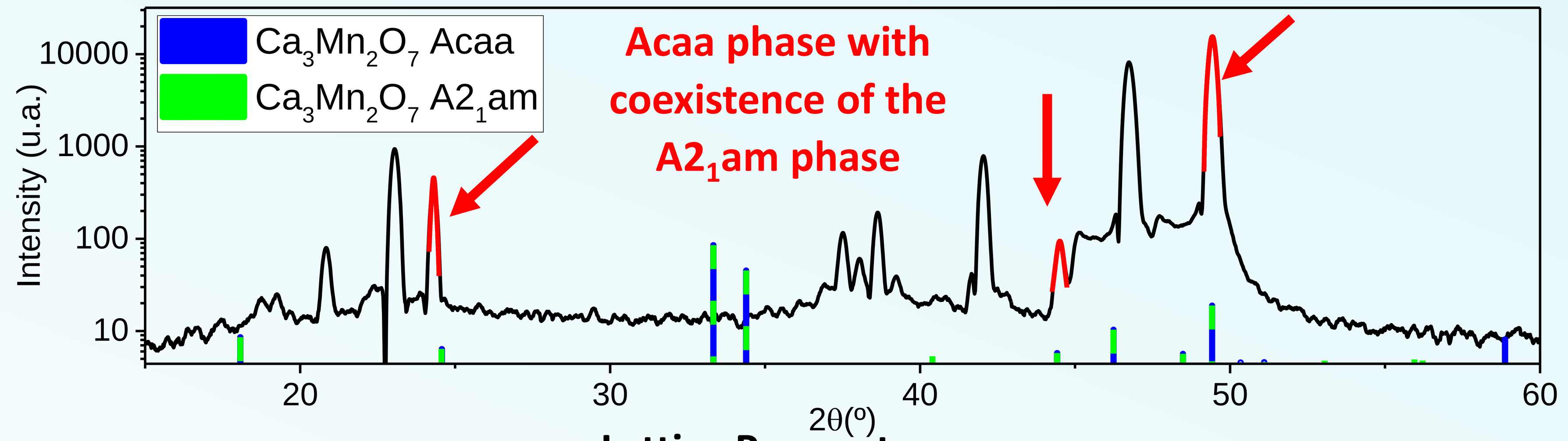
Structural analysis - SEM/EDX





Element	Atomic obtained (%)	Expected atomic (%)
Ca	60.2%	60%
Mn	39.8%	40%
Stoichiometry	Ca_3Mn_2	Ca_3Mn_2

Structural analysis - XRD



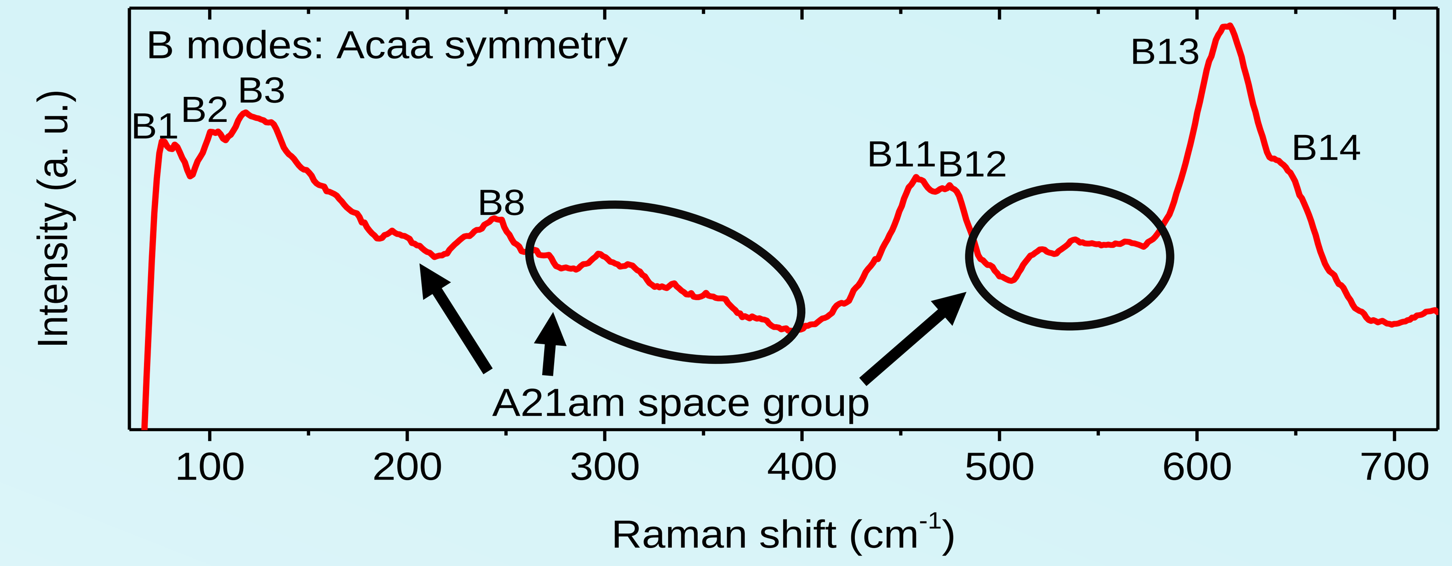
Lattice Parameters

		Obtained (Å)	Deformation along growth direction	
$Ca_3Mn_2O_7$	a	5.214	0.04%	expansion
	b	5.214	0.04%	expansion
	c	19.528	-0.35%	contraction

Interfacial substrate/film strain promotes the formation of the polar distortion

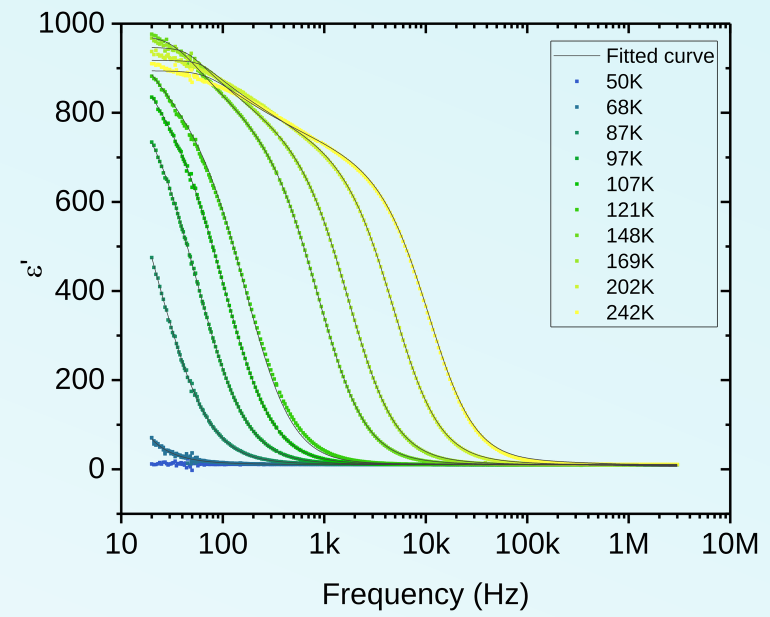
Stabilization of the $Ca_3Mn_2O_7$ ferroelectric phase

Structural analysis - Raman Spectroscopy



Coexistence of the ferroelectric low-T A2$_1$am and the intermediate-T orthorhombic Acaa phases at room temperature

Impedance Spectroscopy



Two distinct regimes two relaxations at low and high frequencies

Havriliak-Negami model function:

$$\varepsilon(\omega) = \varepsilon_\infty + \sum_{j=1}^{2} \frac{\Delta\varepsilon_j}{\left[1 + (i\omega\tau_j)^{\beta_j}\right]^{\gamma_j}}$$

$$\tau(T) = \tau_0 e^{\left(-\frac{E_a}{k_B T}\right)}$$