

## Nanostructuration in insulating $\text{La}_2\text{Ni}_{1-x}\text{Mn}_{1+x}\text{O}_6$

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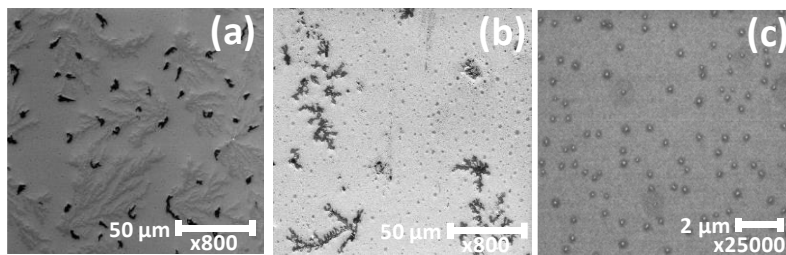
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The tendency of some functional perovskite oxides toward self-organized growth and spontaneous nanostructuration, offers enormous potential for the implementation of new nanodevices. Among them, double perovskite  $\text{La}_2\text{NiMnO}_6$  is one of few ferromagnetic materials with semiconducting character [1]. In this work, we report the epitaxial growth of non-stoichiometric  $\text{La}_2\text{Ni}_{1-x}\text{Mn}_{1+x}\text{O}_6$  thin films on  $\text{SrTiO}_3$  (001) substrates by RF magnetron sputtering using a stoichiometric ceramic  $\text{La}_2\text{NiMnO}_6$  target prepared by sol-gel method. Structural and magnetic properties of non-stoichiometric  $\text{La}_2\text{Ni}_{0.4}\text{Mn}_{1.6}\text{O}_6$  thin films, prepared at high temperature ( $T = 800\text{--}900\text{ }^\circ\text{C}$ ) and low oxygen pressure ( $P_{\text{O}_2} = 130\text{--}150\text{ mTorr}$ ), are studied as a function of annealing conditions (Fig.1). Irrespective to the growth conditions used, films grow fully strained showing insulating behavior. While Atomic Force Microscopy (AFM) show flat surfaces with low roughness, Scanning Electron Microscopy (SEM) images indicate the presence of ramified clusters, randomly and uniformly distributed throughout the surface, associated to low adatom mobility during film growth, as typically observed in metal depositions (Fig.1(a)). High temperature annealing treatment after film growth, as typically used to improve magnetic properties, promotes closing of ramified clusters and the appearance of monolayer vacancy islands (Fig.1(b)). Increasing annealing temperature up to  $900\text{ }^\circ\text{C}$ , ramified clusters disappear completely leaving only round vacancy islands (Fig.1(c)). In addition, the spontaneous formation of nanometric cubes can be observed inside of vacancy islands, probably related with the formation of nickel oxide clusters previously reported in [2]. X-ray absorption spectroscopy (XAS) experiments reveal that these formations in the films are likely due to  $\text{Mn}^{2+}$  inclusions on the surface.

### References

- [1] M. Bernal-Salamanca, Z. Konstantinović, Ll. Balcells, E. Pannunzio-Miner, F. Sandiumenge, L. Lopez-Mir, B. Bozzo, J. Herrero-Martín, A. Pomar, C. Frontera, B. Martínez, *Cryst. Growth Design* (2019)
- [2] S. R. Spurgeon et al., *Chem. Mater.* 28 (2016): 3814–3822.

### Figures



**Figure 1:** SEM images of LNMO thin films grown on STO (001) substrate under low oxygen pressures ( $P_{\text{O}_2} = 140\text{ mTorr}$ ) at high temperature (a) without annealing process and with different annealing under  $420\text{ Torr O}_2$  (b) at  $850\text{ }^\circ\text{C}$  and (c) at  $900\text{ }^\circ\text{C}$ .