Supporting Information

Spontaneous cationic ordering in chemical solution grown La_2CoMnO_6 double perovskite thin films

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S1. X-ray reflectivity spectra of epitaxial LCMO/STO thin films

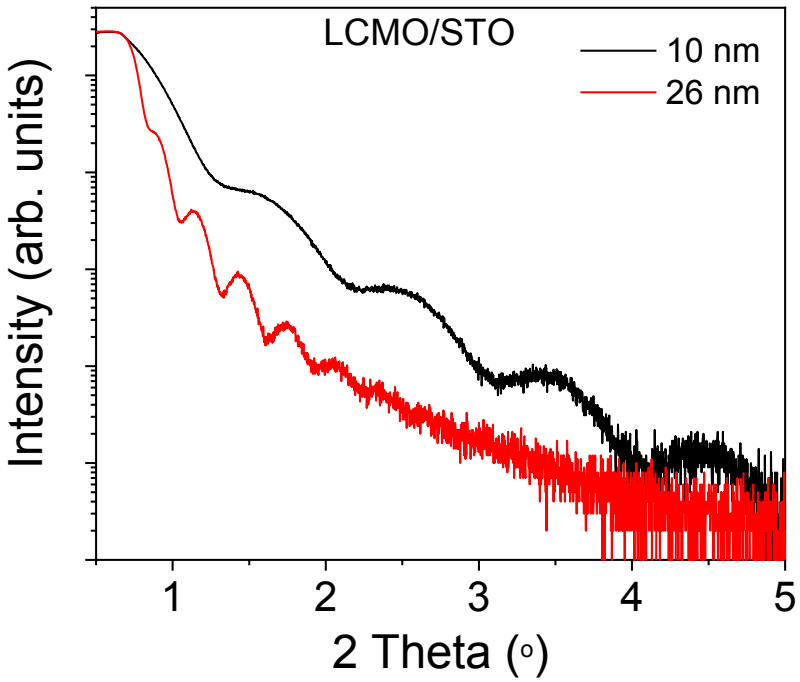

Figure S1: X-ray reflectivity spectra of two LCMO/STO films of thickness 10 nm (black line) and 26 nm (red line) respectively

S2. 3D reciprocal space tomography of LCMO/STO films

3D reciprocal space tomography was done by collecting diffracted intensity using a Bruker D8 advance diffractometer equipped with a 2D GADDS detector (four-circles). Phi-scans where performed at small steps ($\Delta\phi$= 0.5^o) for two different values of Ψ angle (28 ^{o}, 63^o) and two different values of α angle ($\alpha \equiv 2\theta$ at the center of the detector, 25^o, 45^o). Figure S2 shows the collected intensity at two cuts of the reciprocal space, perpendicular to (001) direction at values of $q_z \sim 1/a_{STO}$ (S2(a)) and $q_z \sim 1/2a_{STO}$ (S2(b)).

In figure S2(a) all diffraction peaks have integer indices, no peaks indexed as (h/2k1) (h=odd) or (hk/21) (k=odd) can be seen (indices are referred to primitive cubic reciprocal lattice). Besides, diffraction peaks indexed as (hk½) can be seen in figure S2(b) but not in figure S2(a). This shows that LCMO is orientated with *c* perpendicular to the film.[1]

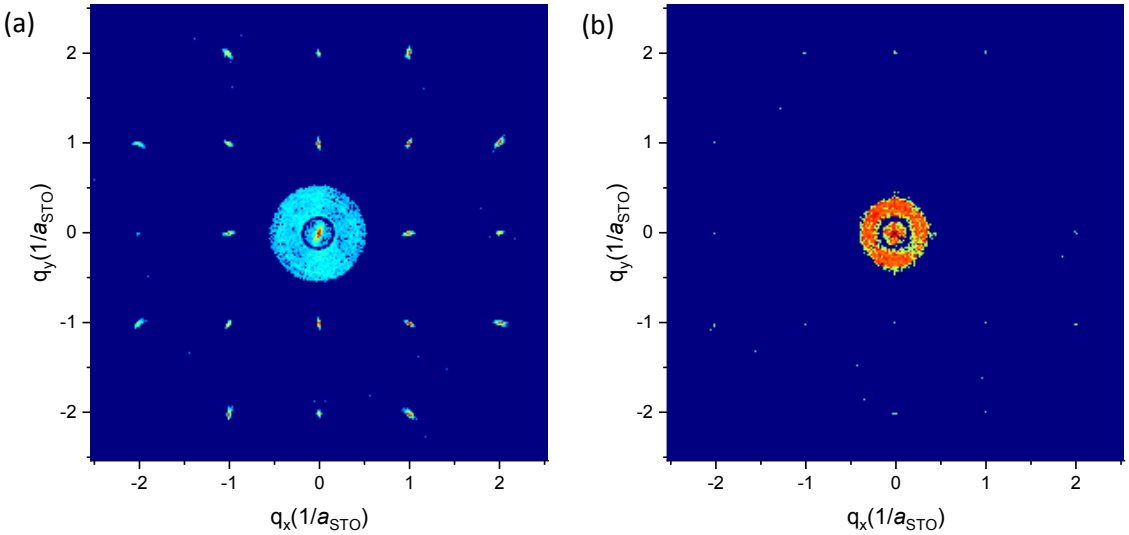

Figure S2: Cuts of the reciprocal space at around (a) l=1 (q_z~ $1/a_{STO}$) and (b) l= ½ (q_z~ $1/2a_{STO}$).

S3. Reciprocal space maps of samples grown under different conditions

Reciprocal space maps of two 26 nm thick samples prepared under different annealing conditions are shown in Figure S3. Sample (a) exhibits very good magnetization properties and has been prepared with a long annealing time at high oxygen flow, while sample (b) has been annealed for a short time under low oxygen flow. In both cases LCMO films grow epitaxial cube-on-cube and fully strained with the substrate. XRD spectra of different samples indicate that the degree of crystallinity and the microstructural film's quality improves with increasing annealing time and oxygen flow.

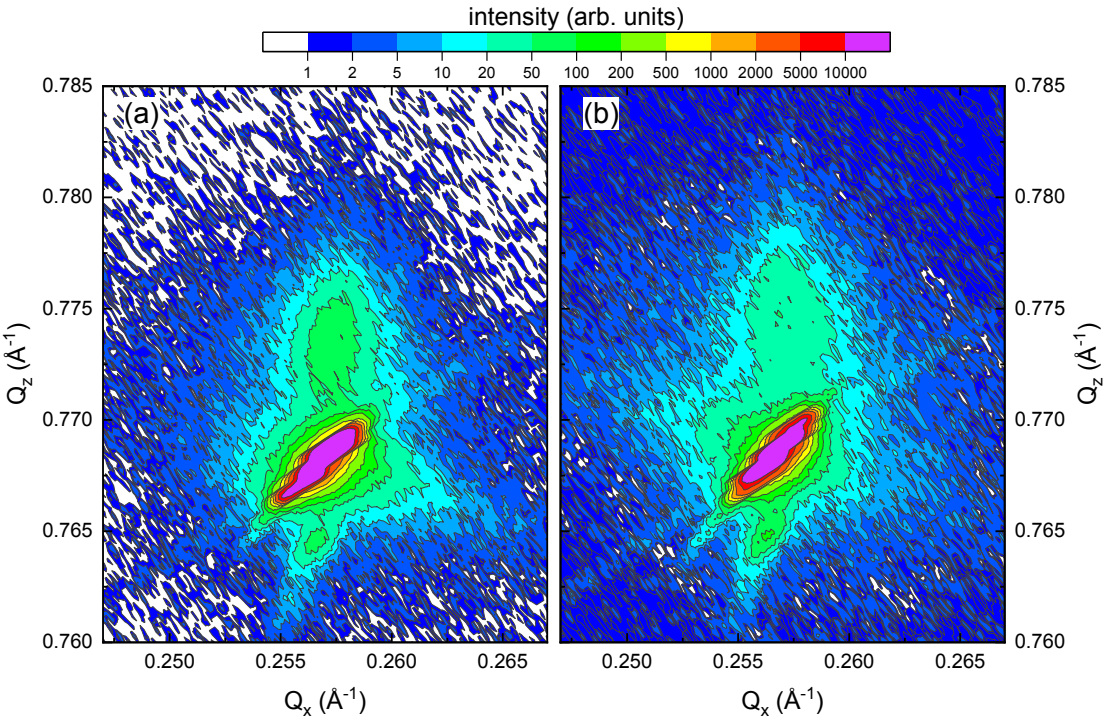

Figure S3: Reciprocal space maps around STO-(103) reflection for two samples prepared at: (a) 900 °C, with long annealing time (60 min) and high oxygen flow (0.3 l/min); and (b) 900 °C, with short annealing time (15 min) and low oxygen flow (0.1 l/min).

S4. Magnetization Vs. temperature of films grown under different conditions

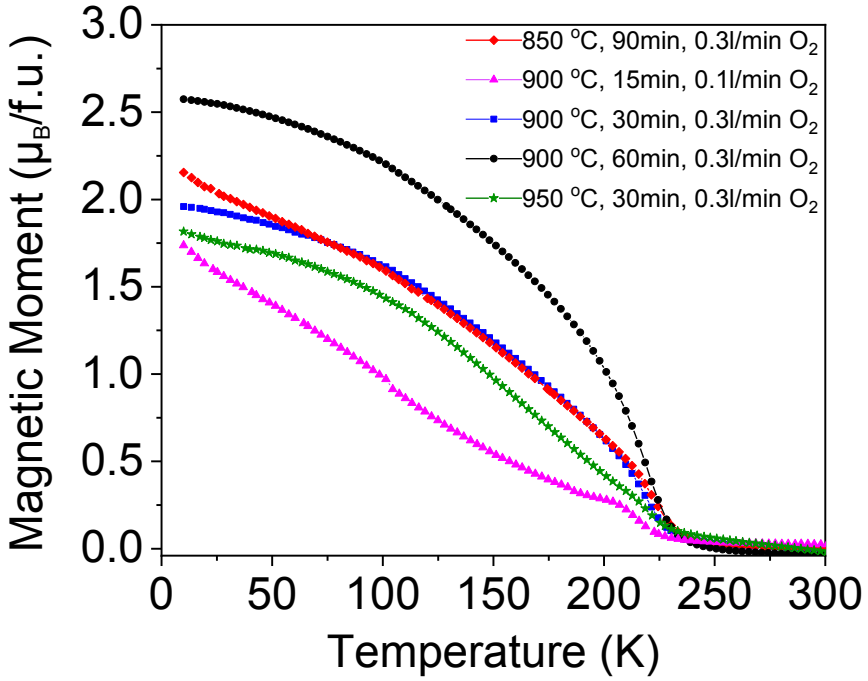

Figure S4: Magnetization vs. temperature of different films grown under different conditions, measured under an applied magnetic field *H*=1 kOe.

From figure S4, it is evidenced that to obtain good magnetic ordering it is necessary to attain high enough temperatures (900 °C), long annealing times (60 min) and high oxygen flow (0.3 l/min). Samples grown under lower oxygen flow (0.1 l/min) or shorter annealing times show lower magnetization values. In concomitance with the improvement in magnetic properties, the degree of crystallinity and the micro-structural film's quality also improves with increasing annealing time and oxygen flux, as it can be appreciated from Figure S3.

S5. *M(H)* hysteresis loops of epitaxial LCMO/STO epitaxial thin films of different thickness

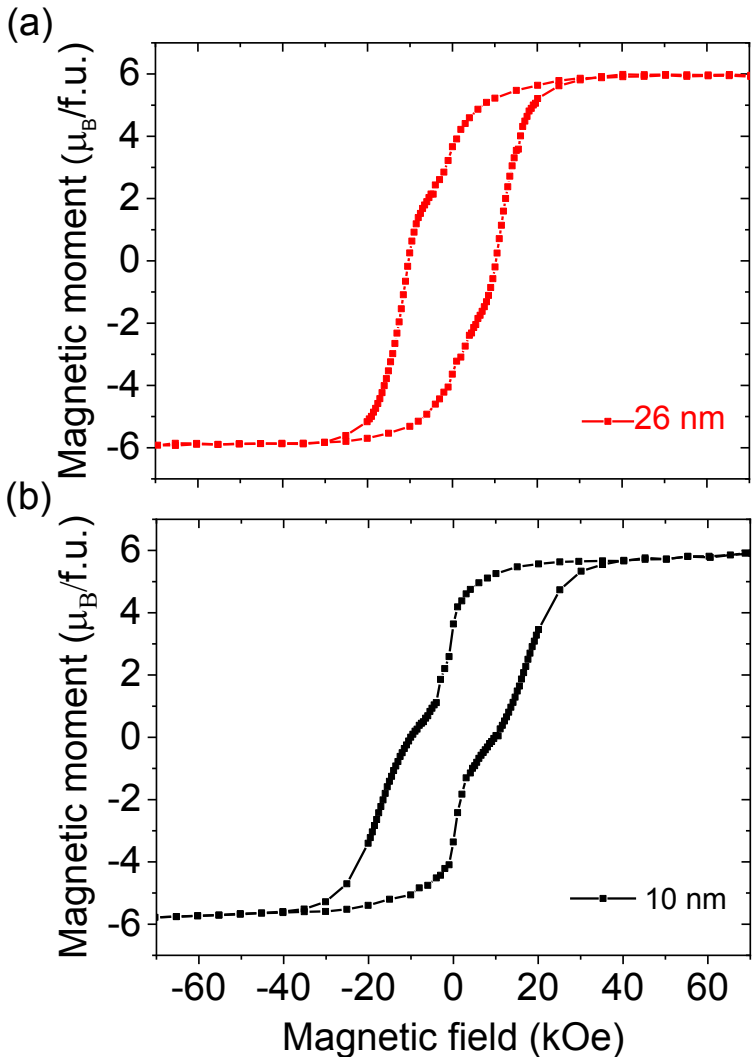

Figure S5: Out of plane *M(H)* hysteresis loops at 10K for two LCMO films grown on STO of different thickness (a) 26 nm, (b) 10 nm.

M(H) loops in Figure S5 show the typical biloop shape. The origin of this sudden drop of the remanent magnetization at H=0, is due to the existence of antiphase boundaries. Antiphase boundaries appear because nucleation of Co^{2+} and Mn^{4+} cationic ordering may occur at different positions in different regions of the sample,

when regions with inverted Co^{2+} and Mn^{4+} positions relative to the other merge together an antiphase boundary is formed. As pointed out by Dass and Goodenough,[2] at the antiphase interface Co^{2+}-O-Co^{2+} or Mn^{4+}-O-Mn^{4+} AF interactions occur, on lowering field from saturation, at H=0, any antiphase regions would revert back to an antiparallel orientation with a 180° change of spin orientation across an antiphase boundary, which would produce the observed sudden drop of the remanent magnetization. This drop is more pronounced in thinner films indicating a larger contribution of antiphase boundaries on the magnetic hysteresis loops.

References:

1. Galceran, R; Frontera, C; Balcells, Ll.; Cisneros-Fernandez, J; Lopez-Mir, L; Roqueta, J; Santiso, J; Bagues, N; Bozzo, B; Pomar, A; Sandiumenge, F; and Martınez, B; Engineering the microstructure and magnetism of $La_2CoMnO_{6-\delta}$ thin films by tailoring oxygen stoichiometry *Appl. Phys. Lett.* (2014) *105*, 242401.
2. Dass R.I. and Goodenough J.B.; Multiple magnetic phases of La_2CoMnO_6, *Phys. Rev. B: Condens. Matter* (2003) *67*, 014401.