Supplementary Information (SI) for Nanoscale.

Supplementary Materials

Nanoscale engineering of electronic and magnetic modulations in gradient functional oxide heterostructures

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Dynamic Metalorganic Aerosol Depositon (dyna-MAD)

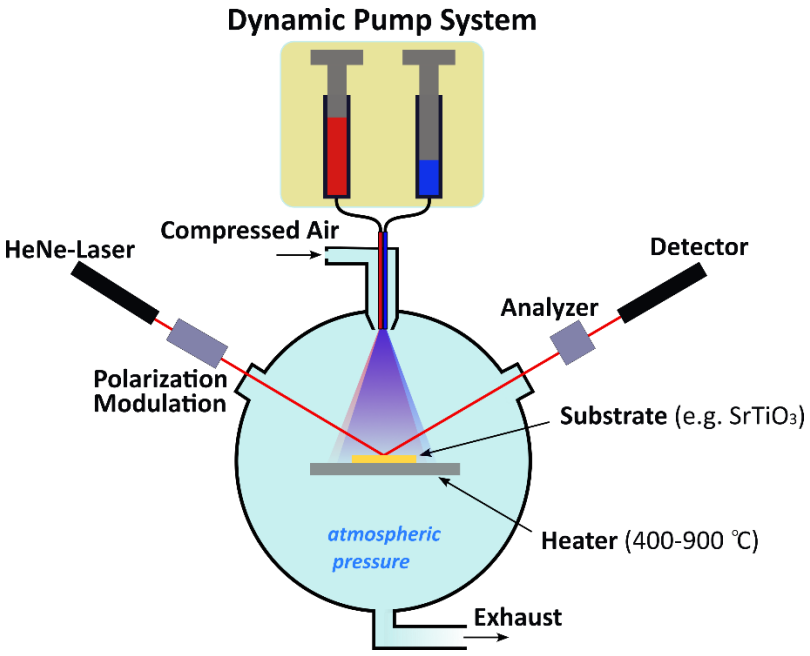

Figure SM-1. (a) Principle of dynamic metalorganic aerosol deposition technique (dyna-MAD), from [1], [2]. The optical monitoring setup consist of a Helium-Neon laser with a wavelength of 633 nm (E=1.96 eV), a polarizer, photoelastic modulator and analyzer.

Electron Energy Loss Analysis of Chemical Composition

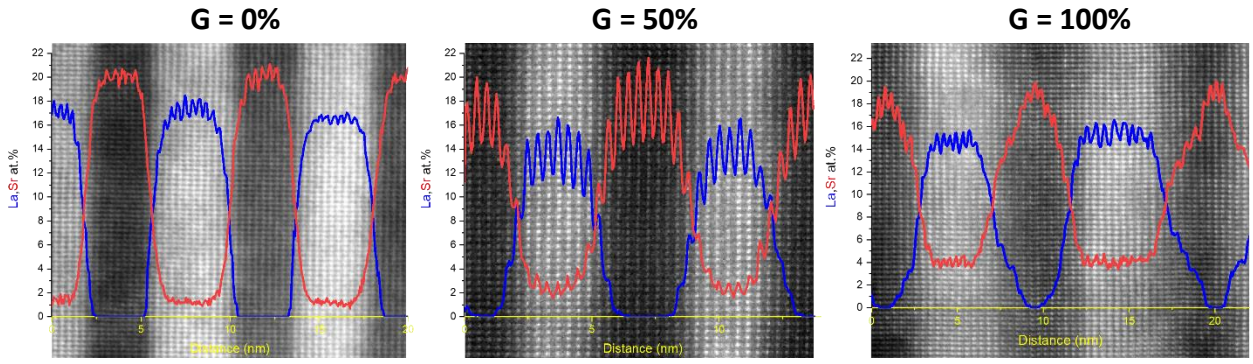

Figure SM-2. EELS analysis. Individual EELS spectra are quantified via the Hartree-Slater model cross-section method [3]. The two-dimensional maps of relative composition are then profile-averaged and plotted along the layer growth (horizontal) direction. Shown are relative La content (blue) and Sr content (red). Mn and Oxygen were included in the analysis but are omitted here for clarity. Overlaid are the corresponding ADF-STEM images.

The limit of sensitivity in this method is about 2 atomic %; residual Sr signal at or below this value results from fit errors of the background model for large energy ranges. Of particular note is the GL100 result: the SMO layers are fully graded as desired, while the LMO layers plateau at a finite Sr concentration, interpreted in the main manuscript as $La_{0.825}Sr_{0.175}MnO_3$. The layer growth is slightly asymmetric, which also becomes apparent in the fit of the XRR data (see Figure 5 in the main manuscript).

Atomic Force Microscopy Images

All samples exhibit a smooth surface morphology typical for a layer-by-layer epitaxial growth., The calculated mean-square-roughness varies in the range of RMS=0.3-0.5 nm for all studied samples [1].

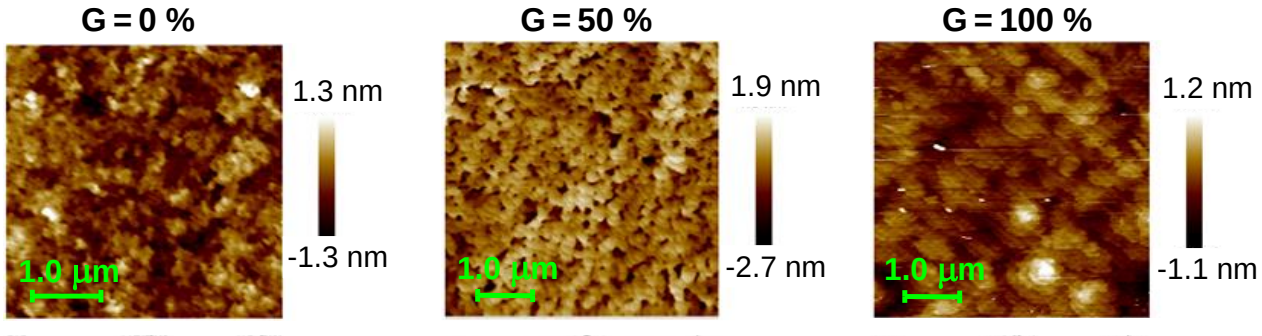

Figure SM-3. Atomic force microscopy images for 0 - 100 % gradient samples.

Electrical Resistance Measurements

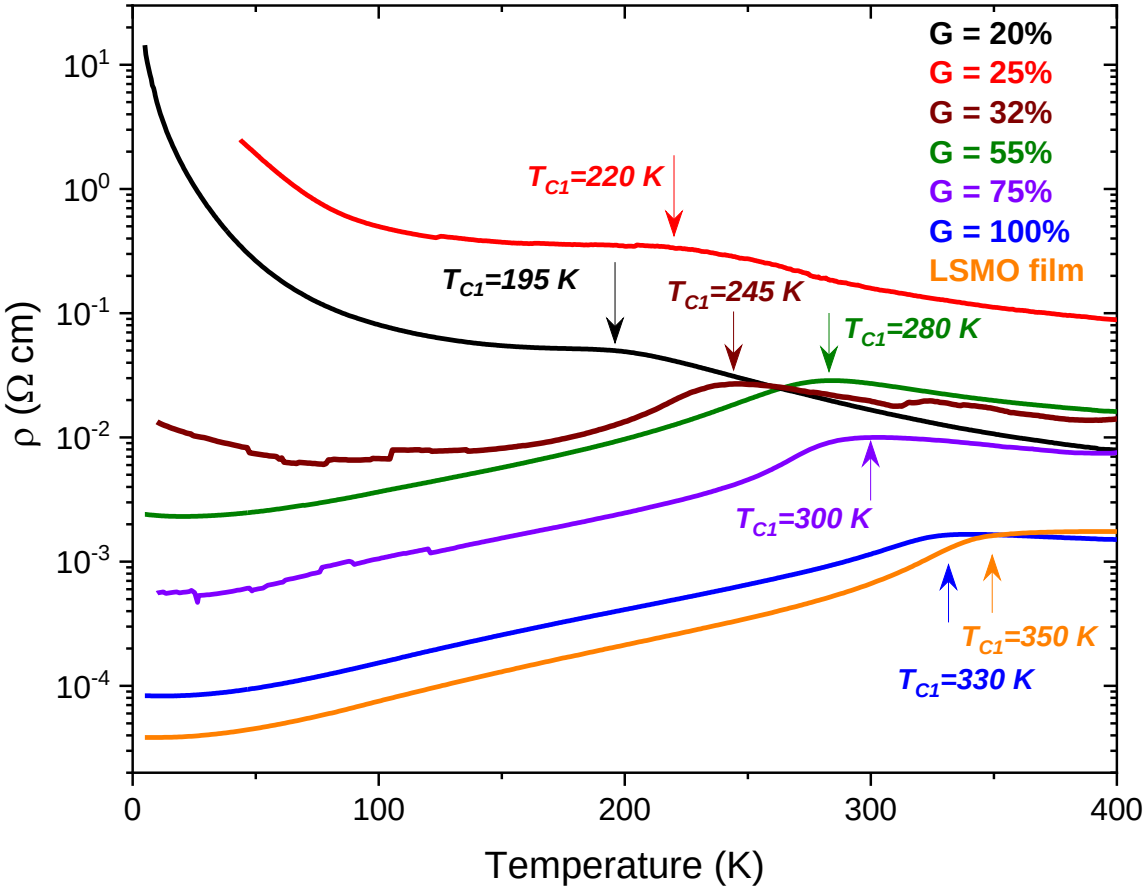

Figure SM-4. Electrical resistivity curves of an LSMO film and of LMO/SMO superlattices with various amounts of interface gradient ***G***, as determined from XRR measurements.

References

[1] F. Lyzwa, L. Schüler and V. Moshnyaga, "Dynamic growth control of complex oxide heterostructures," *Submitted,* 2024.

[2] M. Jungbauer, "Atomic Layer Design of Electronic Correlations in Perovskite Heterostructures," 2016.

[3] G. Bertoni and J. Verbeeck, "Accuracy and precision in model based EELS quantification," *Ultramicroscopy,* vol. 108, pp. 782-790, 2008.