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IV. SUPPLEMENTARY INFORMATION: FILM CHARACTERIZATION BY XRD, AFM, MUTUAL INDUCTANCE AND RESISTIVITY MEASUREMENTS

We have characterized all samples by X-ray diffraction and low-angle X-ray reflectance using an X-pert PRO PANalytical four-circle diffractometer with a Cu tube. A typical X-ray diffractogram for the $x = 0.1$ sample is shown in Figure 5. Out-of-plane lattice constants c_0 are determined using multiple Bragg diffractions and applying the Nelson-Riley algorithm (1). For the various samples, the nominal thickness is known by virtue of our digital (atomic-layer-by-layer) molecular beam epitaxy technique. This nominal thickness was in agreement to within ±5% with the thickness calculated from the finite-thickness oscillations (so-called Kiesig fringes) seen in the diffractograms as satellite peaks on both sides of the strong Bragg peaks (see the inset to Figure 5), as well as in low-angle X-ray reflectance. The error in thickness determination (typically of the order of 5-10 Å) depends on the accuracy that one can measure the period of oscillations. The large number of well-resolved satellite peaks in Fig. 5 indicates that the surface roughness of the film is small.

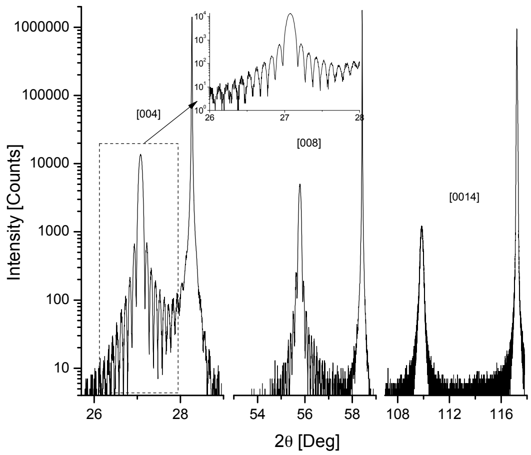

FIG. 5 **X-ray diffractogram of the LSCO $x = 0.1$ film.** An accurate value of the lattice constant c_0 can be retrieved by means of the Nelson-Riley algorithm using a number of observed diffraction peaks. Inset: Magnified view of the (004) Bragg peak, showing pronounced finite-thickness oscillations.

This small surface roughness was confirmed with Atomic Force Microscopy (AFM). For the $10 \times 10\ \mu m^2$ field of view shown in Fig. 6 for the $x = 0.1$ sample, the typical rms surface roughness (2) varied between 0.2 and 0.9 nm; in all cases it was less than one unit cell height ($c_0 \approx 1.3$ nm).

Every sample was also characterized using a two-coil transmission mutual inductance (MI) technique to check film homogeneity. The diamagnetic MI signal onsets when the resistance of the sample reaches zero. The in-phase (dissipative) MI shows a peak below T_c, the width of which is sensitive to both in-plane and out-of-plane inhomogeneity up to the scale of the coil diameter (≈ 1 mm). As seen in Fig. 7a for a optimally doped sample the measured FWHM values for the peaks were in the range between 0.2-0.5 K, which demonstrates the high homogeneity of the films and their excellent quality. Any variation in stoichiometry, either in-plane or out-of-plane, would result in broadening of the dissipative peak, or even in the appearance of multiple peaks.

Measurements of resistance as a function of temperature provide another figure of merit attesting to the high quality of the films, the Residual Resistivity Ratio (RRR). As seen in Fig. 7c, for an optimally doped LSCO film the resistivity ρ(T) is typically linear in T down to temperatures near T_c. This linear dependence can be extrapolated down to $T = 0$, to define the so-called Residual Resistivity ρ_0. The ratio ρ_0/ρ(T=300 K) can be used as a measure of film quality. In general a RRR of 5-10 % is considered very good. In this particular sample the RRR $\ll$ 1%.

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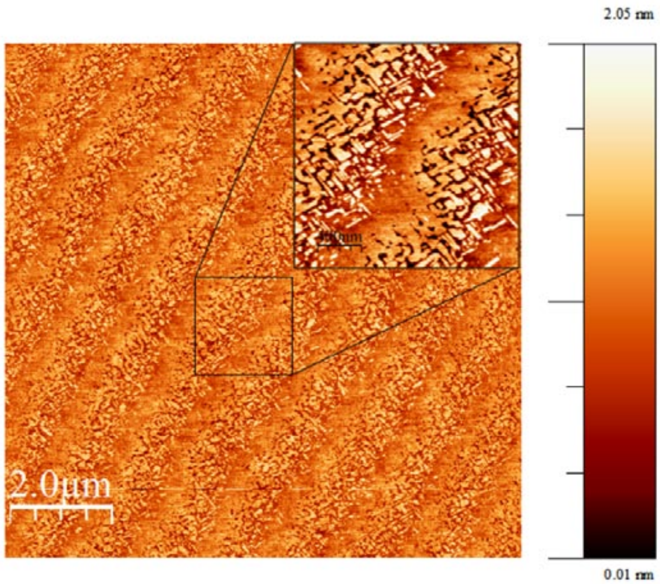

FIG. 6 **Typical AFM image of the $x = 0.1$ LSCO films.** The surface of this sample show terraces that are one mono-layer (1/2 unit cell) high. The rms roughness extracted from this 10×10 μm image is 2.74 Å

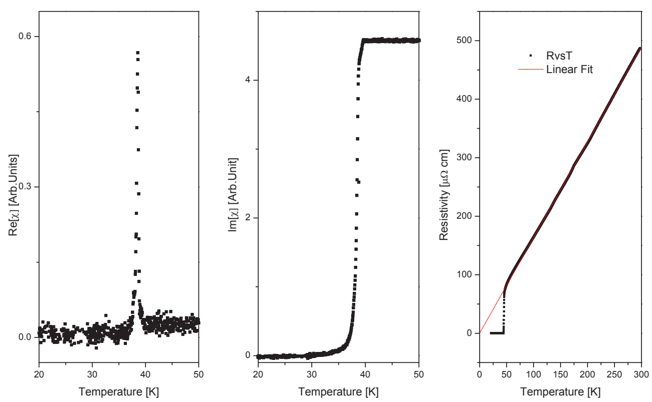

FIG. 7 **Transport in LSCO thin films.** (a) The in-phase (dissipative) and (b) the out-of-phase (inductive) components of mutual inductance measured by two-coil technique. (c) Resistivity vs. temperature measured in an optimally doped LSCO film. The red straight line is the linear fit from the room temperature down to few degrees above T_c.

Supplementary Information References

[1] Lipson, H. & Steeple, H *Interpretation of X-ray Powder Diffraction Patterns* (Macmillan, London, 1970).
[2] Horcas, I. et al. Wsxm: A software for scanning probe microscopy and a tool for nanotechnology. *Review of Scientic Instruments* **78**, 013705 (2007).