

Supplementary Information for

Stoichiometric control of electron mobility and 2D superconductivity at $\text{LaAlO}_3\text{-SrTiO}_3$ interfaces

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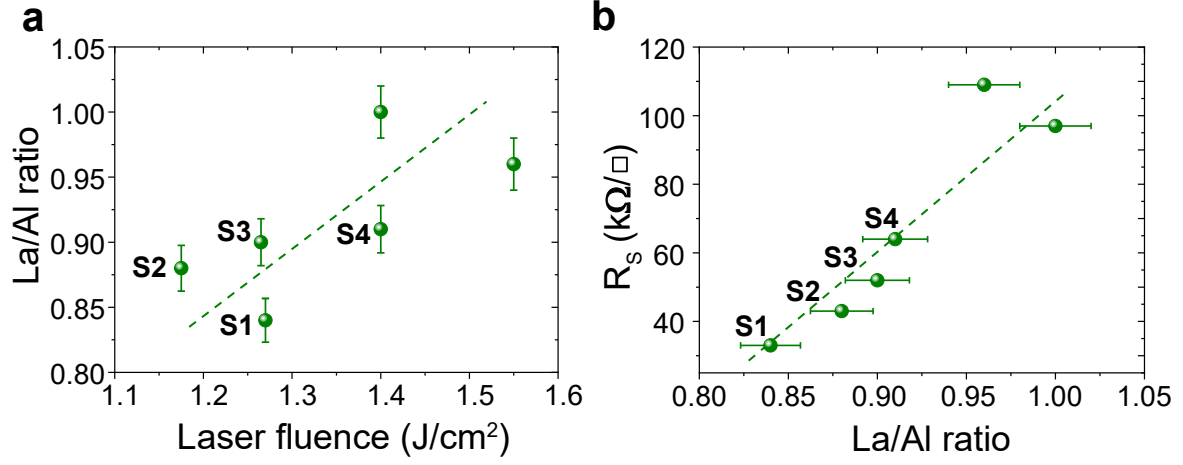
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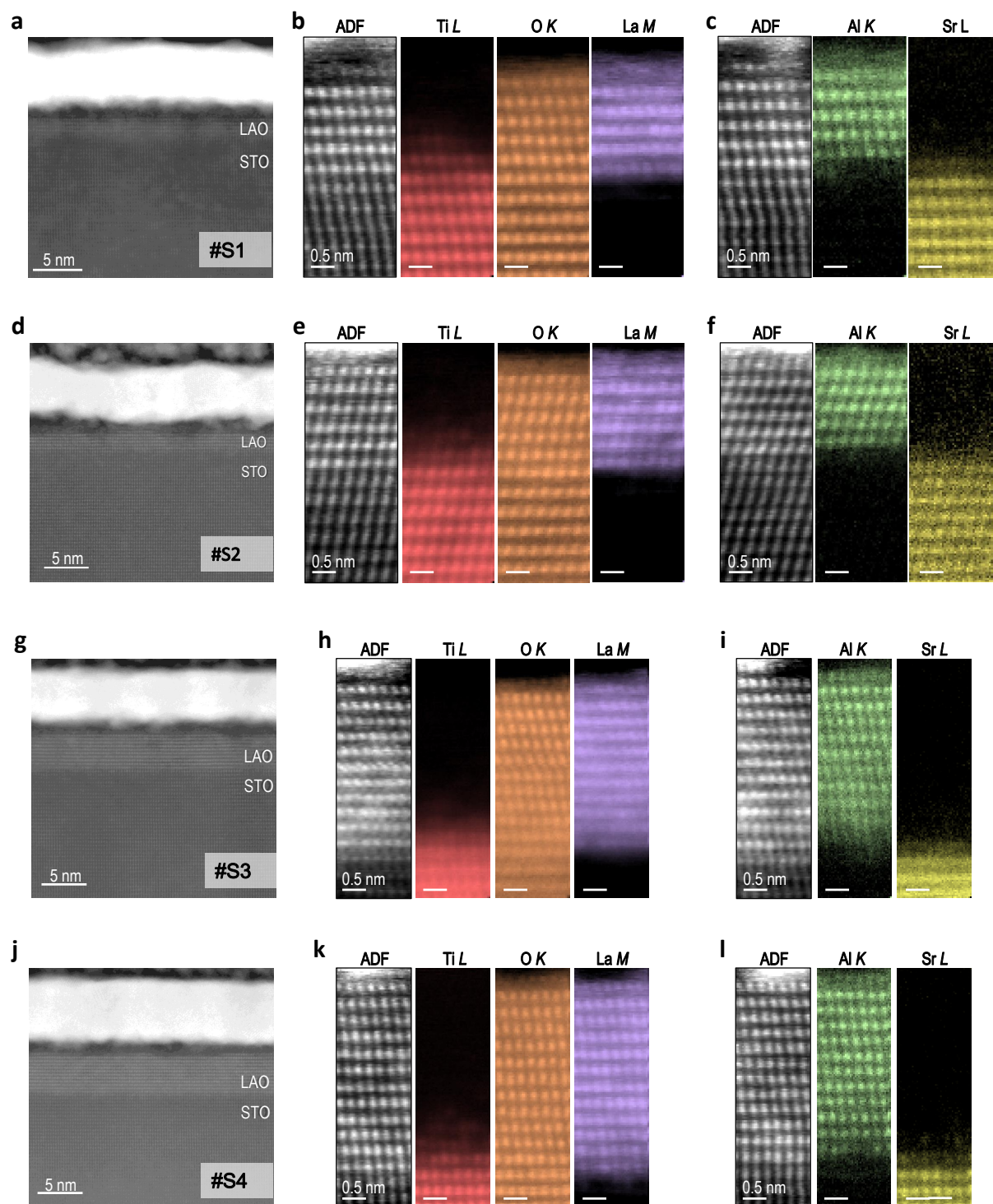
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Supplementary Note 1: Effect of laser fluence on stoichiometry

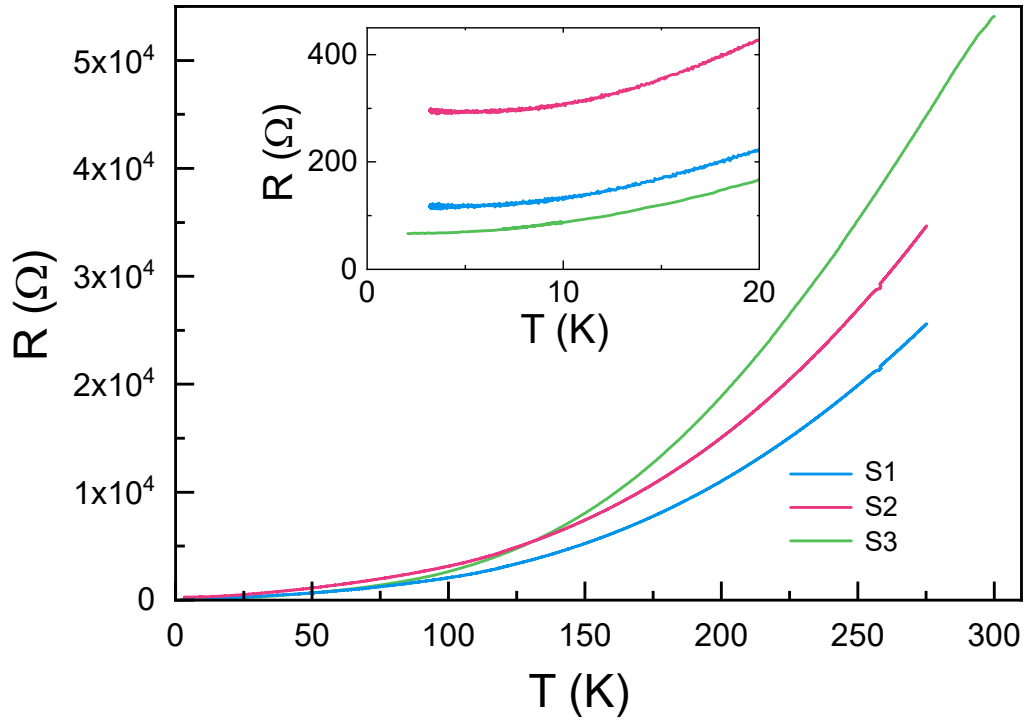
Pulsed laser deposition is a very complicated process involving various parameters and mechanisms. The resulting thin film stoichiometry depends mainly on the distribution of species in the plasma plume, which in turn is primarily governed by the laser fluence on the target, partial background gas pressure, and target substrate distance. Other parameters like position of the substrate in the plume and substrate temperature may also affect the film composition. At the oxygen background pressure below 10^{-3} mbar, early reports found that increasing laser energy density results in the decreased La/Al ratio ([1, 2]). We observed a similar trend in our films, see Supplementary Figure 1a. However, there is a large spread of film composition as a function of laser fluence, which may be caused by small variations of sample position in the plume. We emphasize that the sheet resistance at $T = 300$ K systematically increases in samples with higher La/Al ratio, Supplementary Figure 1b.



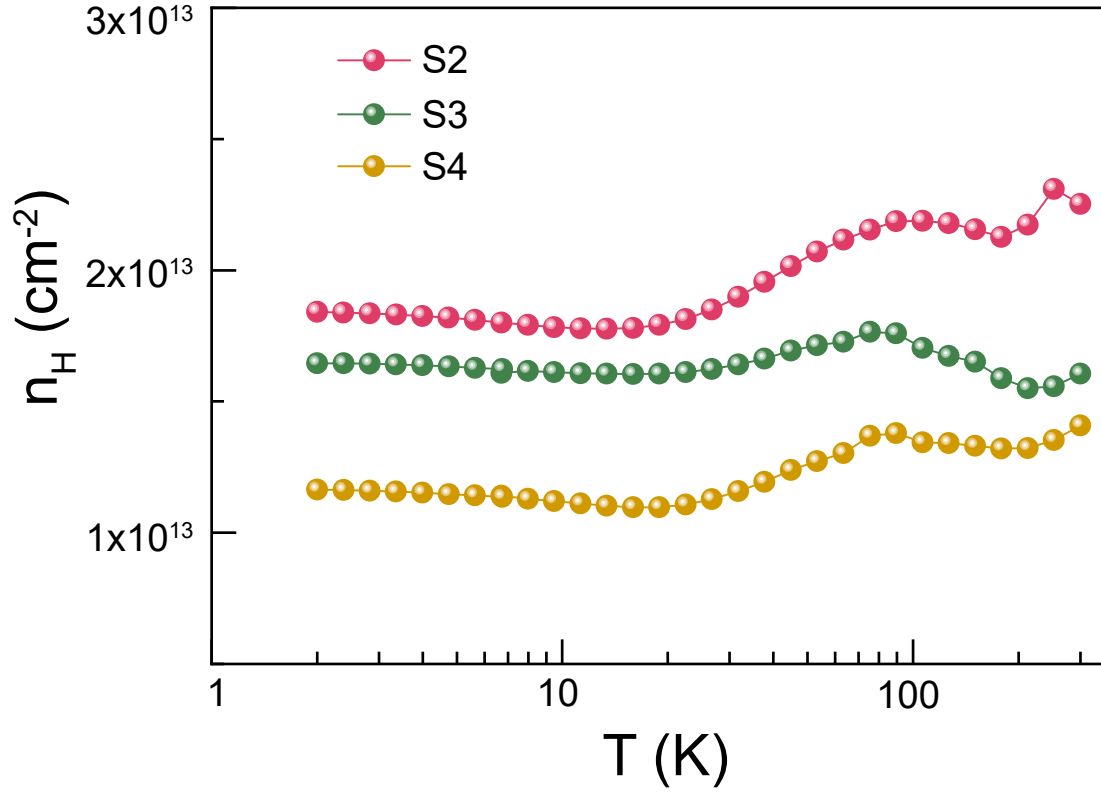
Supplementary Figure 1. (a) Dependence of La/Al ratio obtained from XPS measurements on the laser fluence during pulsed laser deposition. (b) Room temperature sheet resistance of different LAO/STO samples as a function of La/Al ratio.



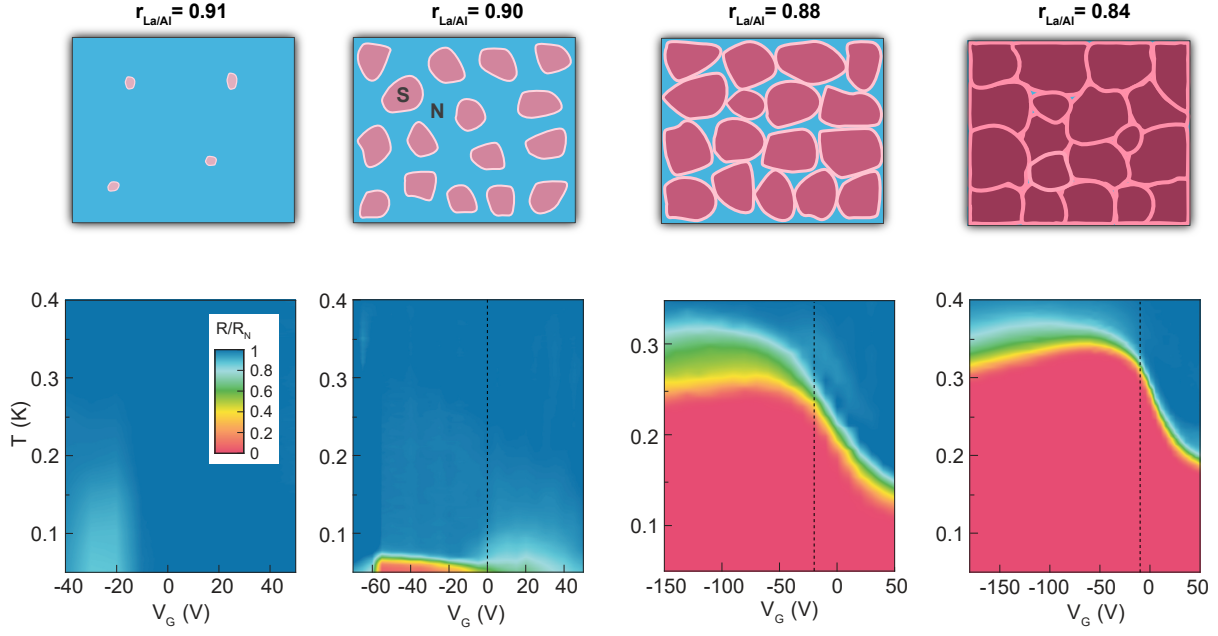
Supplementary Figure 2. Atomic-resolution elemental maps of the rest of the cations using STEM-EELS. Since the energy range of the spectrometer is limited, two different sets of spectrum images were acquired to obtain the elemental maps of all cations of the heterostructure. Each row contains the atomic-resolution elemental maps of each sample of S1, S2, S3, and S4. The figures, from left to right, show the HAADF image (a,d,g,j) from where the spectrum images were acquired, the simultaneous ADF image of the spectrum used to obtain the Ti L (456 eV) edge, the O K edge (530 eV) and the La M (830 eV) edge elemental maps (b,e,h,k), and finally a second simultaneous ADF image of the spectrum image used to obtain the Al K-edge (1560 eV) and the Sr L-edge (1940 eV) elemental maps (c,f,i,l).



Supplementary Figure 3. The sheet resistance as a function of temperature measured between 2-300K for samples S1, S2, S3. The inset shows a zoom below 20K.



Supplementary Figure 4. The Hall carrier density n_H as a function of temperature between 2K-300K for samples S2, S3 and S4. The n_H estimated from R_H for magnetic field limit of $B < 1\text{T}$.



Supplementary Figure 5. Upper panels show the sketch of superconducting puddles (in red) embedded in the weakly metallic matrix (in blue) for samples with $r_{La/Al} = 0.91, 0.9, 0.88, 0.84$. In the lower panel, the corresponding measured superconducting phase diagram is presented.

Supplementary References

- [1] Breckenfeld, E., Wilson, R. B., & Martin, L. W. Effect of growth induced (non)stoichiometry on the thermal conductivity, permittivity, and dielectric loss of LaAlO_3 films. *Appl. Phys. Lett.* **103**, 082901 (2013).
- [2] Park, D.-S. et al. The emergence of magnetic ordering at complex oxide interfaces tuned by defects. *Nature Commun.* **11**, 3650 (2020).