

## Supporting Information

### **Scalable, epitaxy-preserving transfer of freestanding perovskite and layered oxide membranes with tunable strain**

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## Lattice parameter and strain calculations

The out-of-plane lattice parameter ( $c$ ) of the films was determined from the d-spacings of symmetric (00 $l$ ) reflections. Specifically,  $c_{\text{film}}$  values calculated from the (004) and (008) peak positions were averaged. The in-plane lattice parameter ( $a$ ) was determined from reciprocal space maps (RSMs) by fitting the peak-center positions ( $q_x$  and  $q_z$ ) for epitaxially related  $hkl$  reflections from both the substrate and the film. The extracted  $q_x$  values were used to calculate  $a_{\text{film}}$  according to

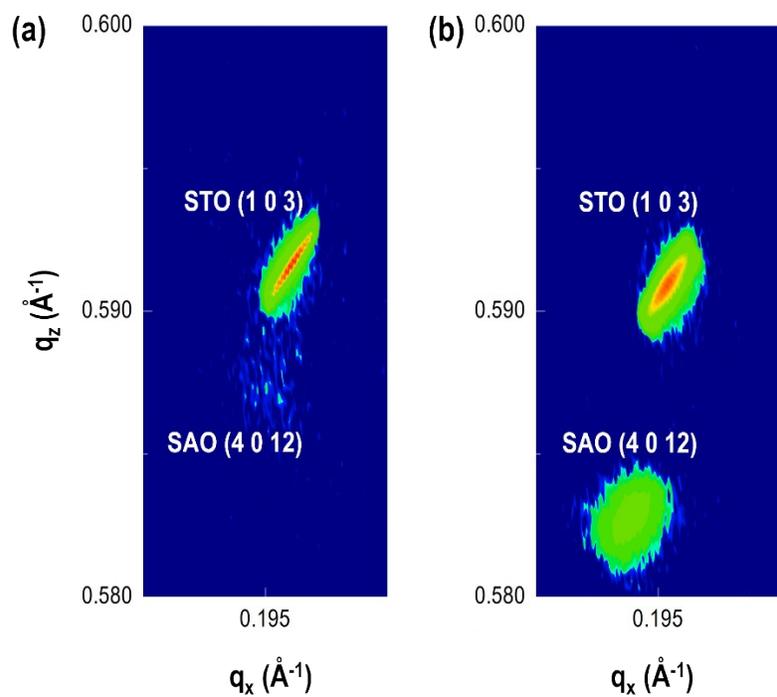
$$a_{\text{film}} = \frac{\left[ \frac{q_x(\text{sub})}{h_{\text{sub}}} \right]}{\left[ \frac{q_x(\text{film})}{h_{\text{film}}} \right]} \times a_{\text{sub}} \quad (1)$$

, which reduces to  $a_{\text{film}} = (q_{x,\text{sub}} / q_{x,\text{film}}) \times a_{\text{sub}}$  when the same in-plane index is used for the film and substrate peak.

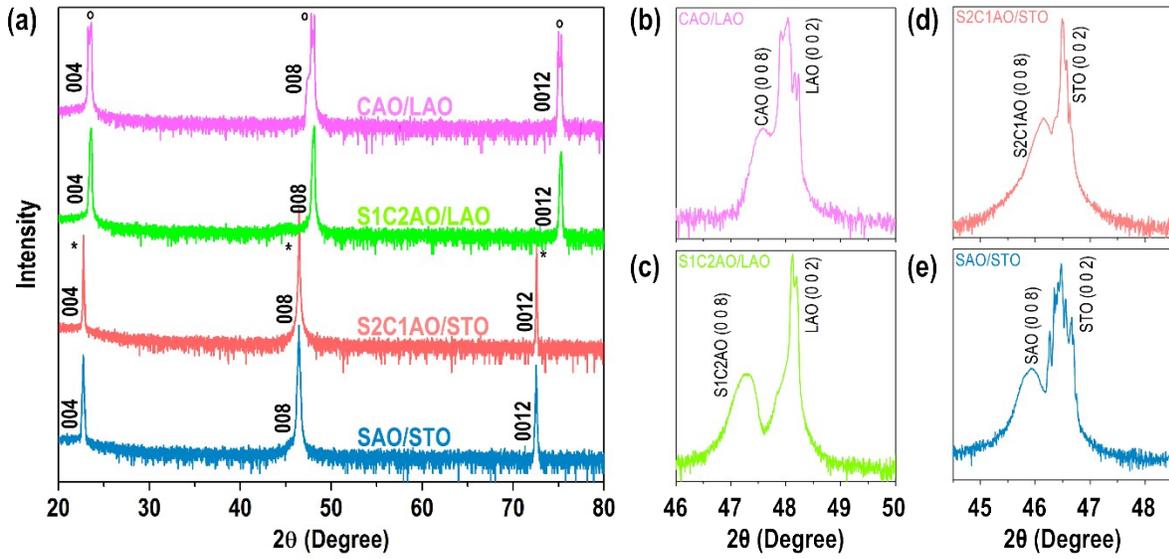
The in-plane strain was calculated as

$$\varepsilon = \frac{a_{\text{film}} - a_{\text{bulk}}}{a_{\text{bulk}}} \times 100\% \quad (2)$$

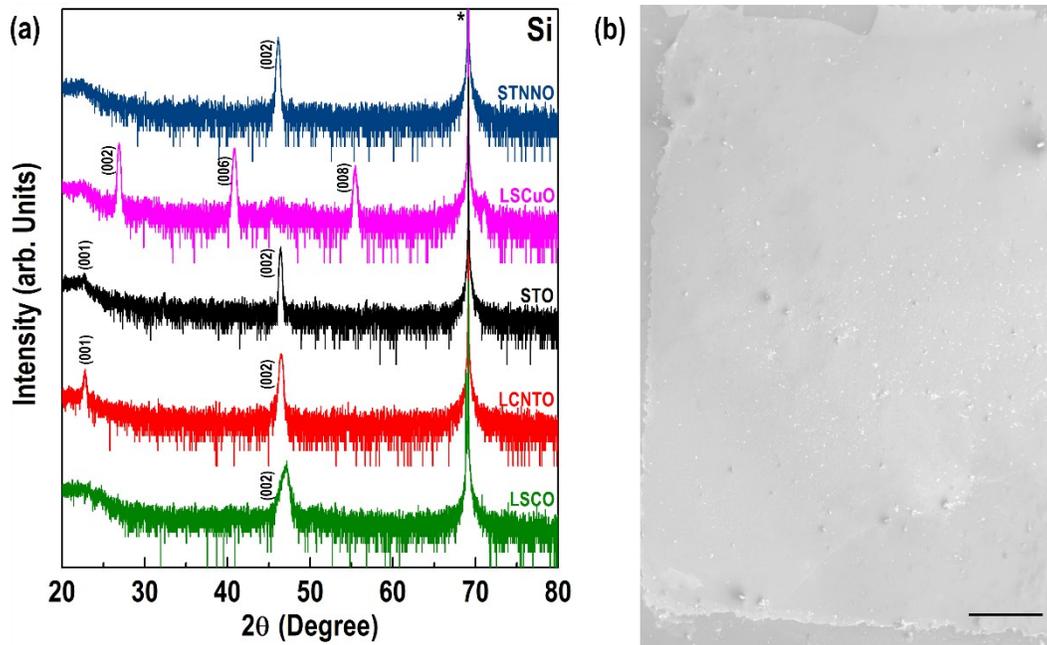
The same procedure was used to determine the in-plane lattice parameter of the SCAO sacrificial layer,  $a_{\text{SCAO}}$ , enabling identification of whether the film was coherently strained to the sacrificial layer ( $q_{x,\text{film}} \approx q_{x,\text{SCAO}}$ ) and whether the sacrificial layer remained strained to the substrate ( $q_{x,\text{SCAO}} \approx q_{x,\text{sub}}$ ).



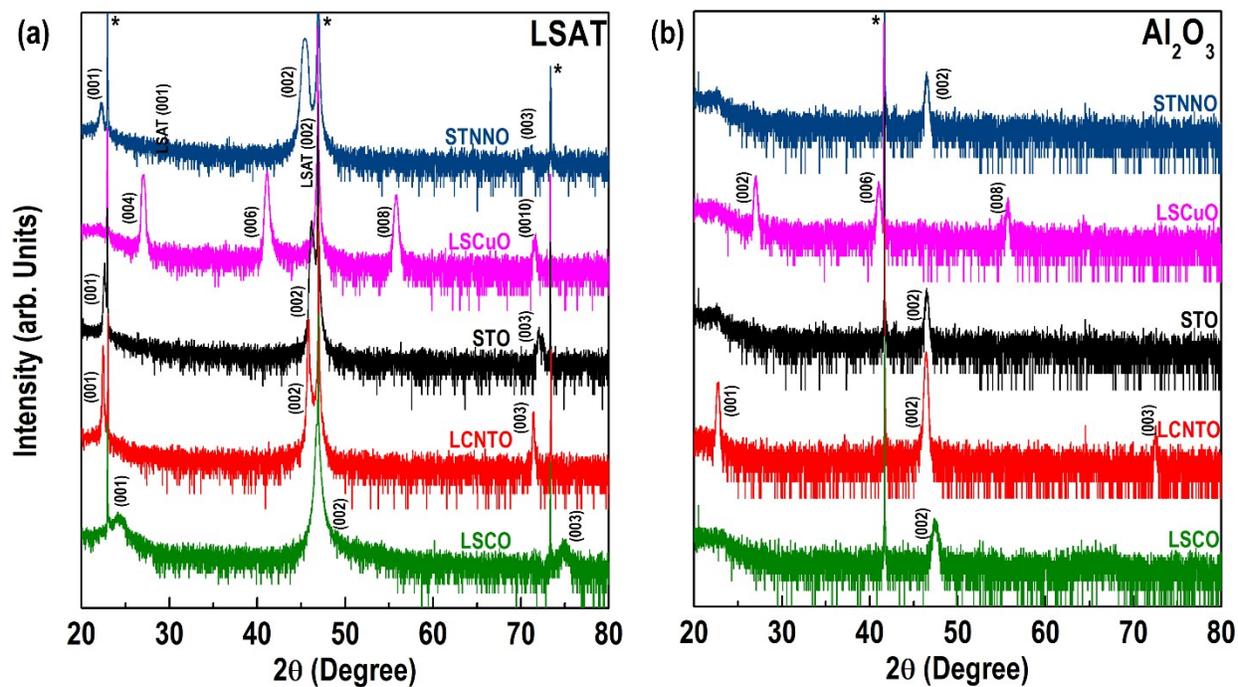
**Figure S1. Representative XRD RSMs around the substrate STO (103) reflection for SAO layers grown epitaxially on STO. (a) 30 nm-thick SAO and (b) 120 nm-thick SAO.**



**Figure S2. X-ray diffraction analysis of epitaxial SCAO sacrificial layers.** (a) Out-of-plane  $\theta$ - $2\theta$  scans of SCAO sacrificial layers grown epitaxially on STO and LAO substrates. The \* and  $\circ$  symbols denote the STO and LAO (00*l*) peaks, respectively. (b-d) Magnified  $\theta$ - $2\theta$  scans around the substrate (002).



**Figure S3. Structural characterization of transferred oxide membranes on Si.** (a) Out-of-plane  $\theta$ - $2\theta$  scans of LSCO, LCNTO, STO, LSCuO, and STNNO membranes transferred onto Si. Well-defined (00 $l$ ) reflections from each membrane confirm retention of the epitaxial out-of-plane orientation after transfer. (b) Representative SEM image of a transferred STO membrane on Si, demonstrating large-area transfer with high uniformity and minimal visible surface defects. Scale bar: 500  $\mu\text{m}$ .



**Figure S4. X-ray diffraction analysis of epitaxial reference films and transferred oxide membranes.** (a) Out-of-plane  $\theta$ - $2\theta$  scans of relaxed epitaxial STNNO, LSCuO, STO, LCNTO, and LSCO layers grown on LSAT substrates. (b) Out-of-plane  $\theta$ - $2\theta$  scans of STNNO, LSCuO, STO, LCNTO, and LSCO membranes transferred onto sapphire. The \* symbol indicates substrate (00 $l$ ) reflections.

**Table S1.** Lattice mismatch between the sacrificial layers and the substrate materials.

Sacrificial layer	$a_{\text{bulk}}$ (Å)	Thickness (nm)	Substrate (Å)	Lattice mismatch ( $a$ - $b$ plane)	In-plane strain (%)
$\text{Sr}_3\text{Al}_2\text{O}_6$ (SAO) <sup>1</sup>	3.961	120	STO, $a = 3.904$ <sup>2</sup>	$\sim -1.5\%$	0
	3.961	30	STO, $a = 3.904$	$\sim -1.5\%$	-1.46
$\text{Sr}_2\text{CaAl}_2\text{O}_6$ (S2C1AO) <sup>3</sup>	3.907	120	STO, $a = 3.904$	$\sim 0.1\%$	0.07
$\text{SrCa}_2\text{Al}_2\text{O}_6$ (S1C2AO) <sup>4</sup>	3.852	30	LAO, $a = 3.780$ <sup>5</sup>	$\sim -1.9\%$	-1.89
$\text{Ca}_3\text{Al}_2\text{O}_6$ (CAO) <sup>6</sup>	3.819	120	LAO, $a = 3.780$	$\sim -1.1\%$	-1.03

**Table S2.** Lattice mismatch between the films and strain-controlled sacrificial layers.

Materials	$a_{\text{bulk}}$ (Å)	$c_{\text{bulk}}$ (Å)	Sacrificial layer (Å)	Sub.	In-plane strain (%)
$\text{Sr}_{0.95}\text{Ti}_{0.76}\text{Ni}_{0.05}\text{Nb}_{0.19}\text{O}_3$ (STNNO) <sup>7, 8</sup>	3.941	3.941	Relaxed SAO $a = 3.961$	STO	0.5
$\text{La}_{0.7}\text{Ca}_{0.2}\text{Ni}_{0.25}\text{Ti}_{0.75}\text{O}_3$ (LCNTO) <sup>9, 10</sup>	3.924	3.924	Relaxed S2C1AO $a = 3.907$	STO	-0.5
$\text{SrTiO}_3$ (STO) <sup>2</sup>	3.904	3.904	Strained SAO $a = 3.904$	STO	0
$\text{La}_{0.8}\text{Sr}_{0.2}\text{CoO}_3$ (LSCO) <sup>11</sup>	3.851	3.851	Strained S1C2AO $a = 3.780$	LAO	-1.87
$\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ (LSCuO) <sup>12</sup>	3.770	13.232	Strained CAO $a = 3.780$	LAO	0.26

## References

1. Lu, D.; Baek, D. J.; Hong, S. S.; Kourkoutis, L. F.; Hikita, Y.; Hwang, H. Y., Synthesis of freestanding single-crystal perovskite films and heterostructures by etching of sacrificial water-soluble layers. *Nature materials* **2016**, *15* (12), 1255-1260.
2. Fuchs, D.; Adam, M.; Schweiss, P.; Gerhold, S.; Schuppler, S.; Schneider, R.; Obst, B., Structural properties of slightly off-stoichiometric homoepitaxial SrTi<sub>x</sub>O<sub>3-δ</sub> thin films. *Journal of Applied Physics* **2000**, *88* (4), 1844-1850.
3. Yun, S.; le Cozannet, T. E.; Christoffersen, C. H.; Brand, E.; Jespersen, T. S.; Pryds, N., Strain Engineering: Perfecting Freestanding Perovskite Oxide Fabrication. *Small* **2024**, 2310782.
4. Varshney, S.; Ramis, M.; Choo, S.; Coll, M.; Jalan, B., Epitaxially grown single-crystalline SrTiO<sub>3</sub> membranes using a solution-processed, amorphous SrCa<sub>2</sub>Al<sub>2</sub>O<sub>6</sub> sacrificial layer. *Journal of Materials Chemistry C* **2024**, *12* (35), 13809-13815.
5. Bořkov, Y. A.; Danilov, V., Electrical resistivity and magnetoresistance of nonuniformly mechanically strained films (30 nm) of the La<sub>0.67</sub>Ca<sub>0.33</sub>MnO<sub>3</sub> manganite. *Physics of the Solid State* **2006**, *48* (8), 1533-1538.
6. Chiabrera, F. M.; Yun, S.; Li, Y.; Dahm, R. T.; Zhang, H.; Kirchert, C. K.; Christensen, D. V.; Trier, F.; Jespersen, T. S.; Pryds, N., Freestanding perovskite oxide films: Synthesis, challenges, and properties. *Annalen der Physik* **2022**, *534* (9), 2200084.
7. Weber, M. L.; Wilhelm, M.; Jin, L.; Breuer, U.; Dittmann, R.; Waser, R.; Guillon, O.; Lenser, C.; Gunkel, F., Exsolution of embedded nanoparticles in defect engineered perovskite layers. *ACS nano* **2021**, *15* (3), 4546-4560.
8. Hirata, S.; Ohtaki, M.; Watanabe, K., Highly improved thermoelectric performance of Nb-doped SrTiO<sub>3</sub> due to significant suppression of phonon thermal conduction by synergetic effects of pores and metallic nanoparticles. *Ceramics International* **2020**, *46* (16, Part A), 25964-25969.
9. Kim, Y. H.; Kang, Y.; Jo, S.; Jeong, H.; Neagu, D.; Myung, J.-h., Shape-shifting nanoparticles on a perovskite oxide for highly stable and active heterogeneous catalysis. *Chemical Engineering Journal* **2022**, *441*, 136025.
10. El Loubani, M.; Yang, G.; Kouzehkanan, S. M. T.; Oh, T.-S.; Balijepalli, S. K.; Lee, D., Influence of redox engineering on the trade-off relationship between thermopower and electrical conductivity in lanthanum titanium based transition metal oxides. *Materials Advances* **2024**, *5* (22), 9007-9017.
11. Lee, D.; Lee, Y.-L.; Grimaud, A.; Hong, W. T.; Biegalski, M. D.; Morgan, D.; Shao-Horn, Y., Enhanced Oxygen Surface Exchange Kinetics and Stability on Epitaxial La<sub>0.8</sub>Sr<sub>0.2</sub>CoO<sub>3-δ</sub> Thin Films by La<sub>0.8</sub>Sr<sub>0.2</sub>MnO<sub>3-δ</sub> Decoration. *The Journal of Physical Chemistry C* **2014**, *118* (26), 14326-14334.
12. Zhu, J.; Zhao, Z.; Xiao, D.; Li, J.; Yang, X.; Wu, Y., Characterization and catalytic activity in NO decomposition of La<sub>2-*x*</sub>Sr<sub>*x*</sub>CuO<sub>4</sub> (0 ≤ *x* ≤ 1) compounds with T\* phase structure. *Materials Chemistry and Physics* **2005**, *94* (2-3), 257-260.