Supporting Information

3D architectures of single-crystalline complex oxides

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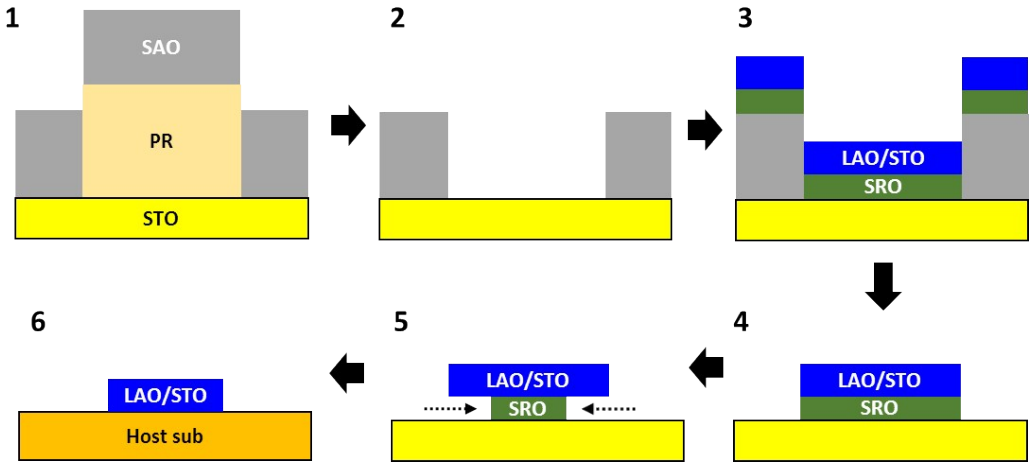

Figure S1. Free-standing membrane fabrication flow. $Sr_3Al_2O_6$ (SAO) hard mask is deposited at room temperature to define the pattern of LAO/STO membranes (1-2). The epitaxial SRO, STO films are deposited by pulsed laser deposition (PLD) under 100 mTorr with a laser energy density of 1.5 J/cm^2 and a frequency of 2 Hz at 700 ^{o}C. The epitaxial $LaAlO_3$ films are deposited under 1 mTorr with the same laser conditions and temperature (3). SAO hard mask is removed by water (4). The membrane of LAO/STO structure is released by selectively etching the SRO thin film in $NaIO_4$ solution (4). When the substrate is transferred to DI water and the membrane is released, the membrane floats on the surface of the water and can be transferred to another host substrate (6).

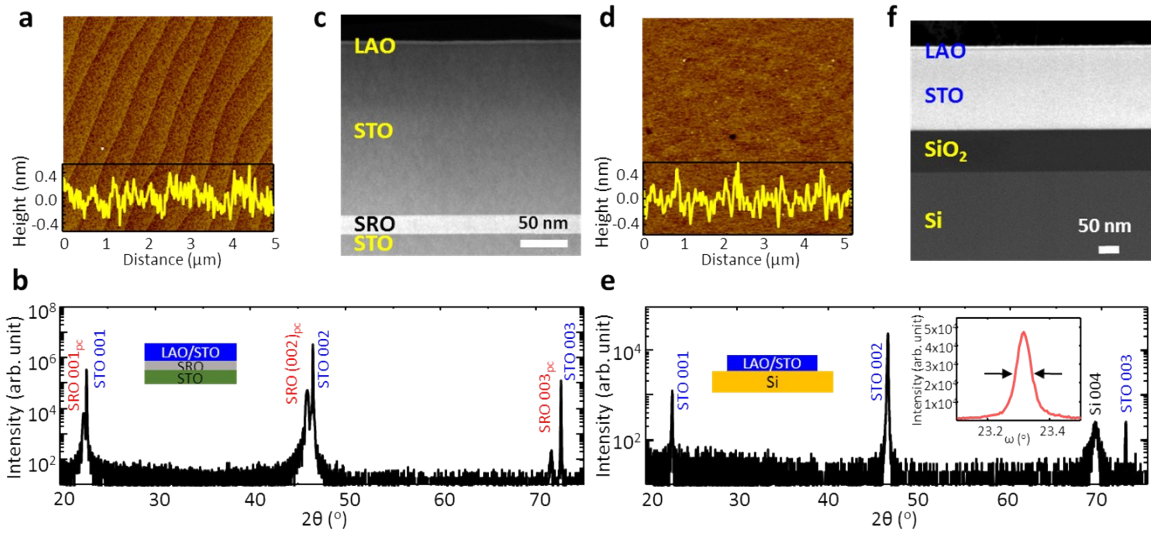

Figure S2. Structural characterization of LAO/STO/SRO/STO and free-standing LAO/STO. The surface morphology of the (a) LAO/STO/SRO/STO and (d) free-standing LAO/STO membrane measured by AFM. X-ray diffraction out-of-plane θ-2θ scan of the (b) LAO/STO/SRO/STO and (e) free-standing LAO/STO membrane. Cross-sectional STEM images of the (c) LAO/STO/SRO/STO and (f) free-standing LAO/STO membrane. The full width at half maximum (FWHM) of rocking curve (RC) of STO 002 peak is 0.058° as shown in the inset of Fig. S2e. Considered the possible micro-wrinkles in the membrane, the actual FWHM of the STO 002 peak of the membrane may be smaller than 0.058°.

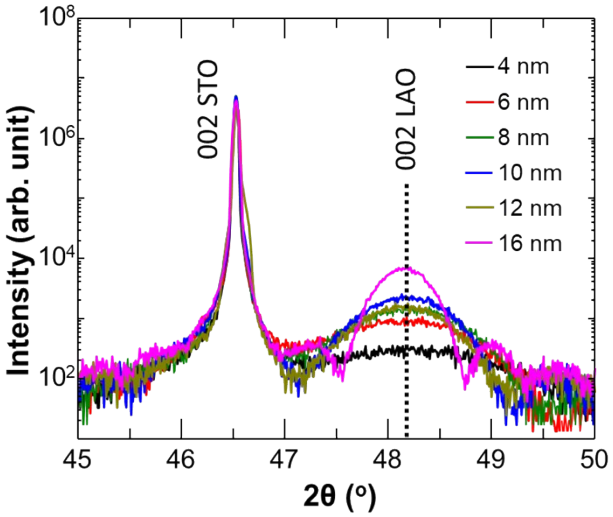

Figure S3. *θ-2θ* scan of LAO/STO with various LAO thicknesses.

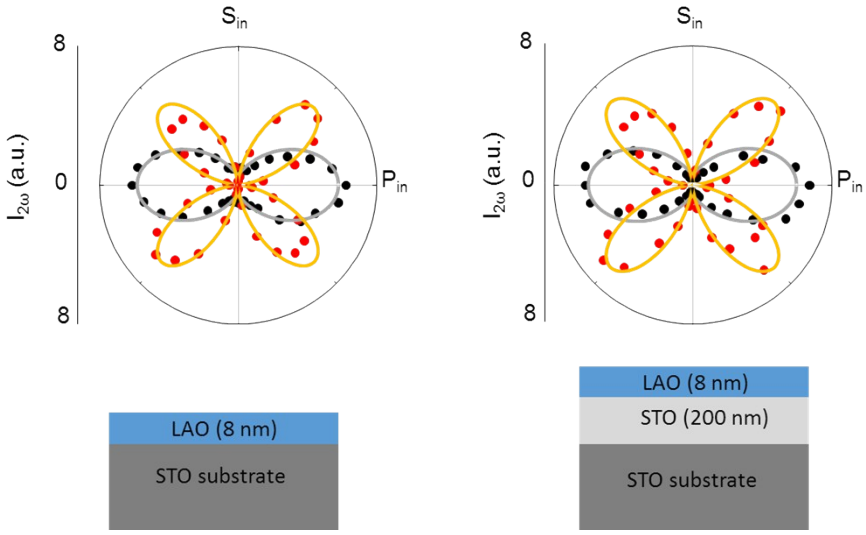

Figure S4. SHG measurements on control samples: LAO (8 nm)/STO substrate and LAO (8 nm)/STO (200 nm)/STO substrate.

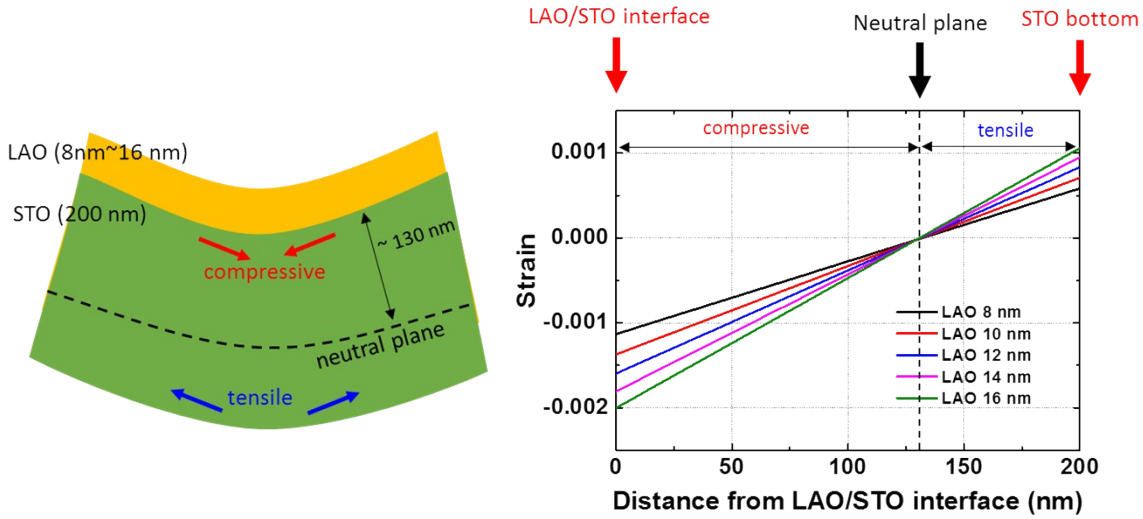

Figure S5. Lateral strain of the STO layer in the free-standing membrane of LAO (x nm)/STO (200 nm) estimated by Finite Element Analysis.

	$LaAlO_3$	$SrTiO_3$
Density	6520 kg/m^3	6810 kg/m^3
Young's modulus	256.07 Gpa [a]	277.44 Gpa [b]
Poisson's ratio	0.24962 [a]	0.23807 [c]

Table S1. Materials parameters of LAO and STO used for computational calculation. ([a]*J. Appl. Phys*. 104, 073518 (2008), [b]*Europhys. Lett*, 50, 41 (2000), [c]*Appl. Phys. Lett*. 100, 061904 (2012))