

Supporting Information

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Braiding Lateral Morphotropic Grain Boundaries in Homogenetic Oxides

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Supplementary Materials for

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This PDF file includes:

Figs. S1 to S19 References (34–38)



Fig. S1. Synthesis of LSCO hybrid structures. Process schematics for (a) (001)-oriented SAO/STO growth on (001)-oriented LSAT substrates, (b) water-solution of SAO layer and release ultrathin STO membranes, (c) transfer and (d) thermally release the STO membrane on different target substrates, for instance, the (110)- and (111)-oriented STO, LAO, and KTO substrates. (e) Fabrication of the LSCO hybrid structures on surface-engineered substrates by PLD technique. More experimental details see <u>Materials and Methods</u>. (f) Optical microscope image of an ultrathin STO membrane with a thickness of ~ 8-unit cells (u. c.) transferred onto the representative (110)-oriented STO substrates. The transferred STO membrane maintains its original shape and clear boundaries with minimal damage within the STO membranes. The typical size of the STO membranes is $3 \times 3 \text{ mm}^2$. (g) Atomic force microscopy (AFM) image for a STO/SAO bilayer grown on LSAT substrates. The clear terrace-and-step structures demonstrate the films are grown in a step flow mode and the sample quality is superior with unit-cell flat surface.



Fig. S2. Epitaxial growth of LSCO hybrid structures. XRD 2θ-ω scans of (a) a LSCO hybrid structure grown on a (110)-oriented STO substrate, (b) a LSCO hybrid structure grown on a (111)-oriented STO substrate, and (c) a (001)-oriented LSCO single film grown on STO substrates. The STO substrates' reflections are indicated with "*". The LSCO hybrids grown on (110)- and (111)-oriented STO substrates exhibit the clear (00*l*) reflections besides the typical reflections with identical orientations to the substrates. (d) Reciprocal space map (RSM) of a LSCO hybrid structure around 013 reflection of a (110)-oriented STO substrate. The results reveal that a 30 nm-thick LSCO film is coherently grown on the STO substrate. (e) RSM around 013 reflection of the (001)-oriented LSCO layers grown on the freestanding (FS) STO membranes. We find that the LSCO layers directly grown on STO substrates are tensile-strained. The out-of-lattice parameter (c) of LSCO layers is ~ 3.773 Å, whereas the c of FS-LSCO layer grown on FS-STO membranes is ~ 3.818 Å. Please note that this FS-LSCO layer is not fully relaxed to its bulk form ($c_{pc} \sim 3.838$ Å) but slightly tensile-strained by the FS-STO membranes. The macroscopic structural characterizations demonstrate the highly epitaxial growth of all LSCO layers on both STO substrates and (001)-oriented FS-STO membranes, forming a laterally LSCO hybrid structure with different crystallographic orientations.



Fig. S3. Microscopic structural characterization of a LSCO hybrid structure grown on (110)-oriented STO substrates. (a) Low-magnified cross-sectional HAADF-STEM image of a LSCO hybrid structure. The FS-STO membrane is closely adhered to the (110)-oriented STO substrates and is continuous with homogenous layer thickness. High-magnified STEM images from the colored rectangles marked in (a) indicate three representative interface regions: (b) (001)-LSCO/(001)-FS-STO/(110)-STO region, (c) grain boundary (GB) region, and (d) (110)-LSCO/(110)-STO region. The brighter layers are LSCO, and the darker layers are STO, because the stronger electron scattering by heavier elements with larger atomic number (La > Sr). STEM results demonstrate that the interfaces between LSCO layers and STO substrates/membranes are atomically sharp, and the interface roughness is less than one-u. c.-thick. The white dashed lines indicate the positions of grain boundaries (GB) between different oriented LSCO layers. We note that there is a black region between the FS-STO membranes and (110)-oriented STO substrates. The gap is unbelievable narrow with an averaged thickness less than 1 nm. The naturally formed gap allows the FS-STO membrane unbonded to the (110)-oriented STO substrates, thus the LSCO layer follows the crystallographic orientation of FS-STO membranes.



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Fig. S4. HAADF-STEM image and EELS map collected from a respective grain boundary region in LSCO hybrid structures. (a) High-magnified STEM image around a grain boundary. The colored panels show the integrated EELS intensities of (b) Co $L_{3,2}$ - and (c) Ti $L_{3,2}$ -edges, respectively. We find that the interfaces between (001)-LSCO/(001)-FS-STO membranes and (110)-LSCO/(110)-STO substrates do not exhibit significant chemical intermixing. We determine that the chemical intermixing at both interfaces is approximately one-u. c.-thick.



Fig. S5. High-magnified HAADF-STEM image around interface region. (a) Low-magnified STEM image of a LSCO hybrid structure grown on (110)-oriented STO substrates. (b) Atomic-resolved STEM image from a blue rectangle region in (a). The STEM results reveal that the averaged thickness of (001)-FS-STO membrane is ~ 8 u. c. (approximately 3 nm). The zoom-in STEM image illustrates that the thickness of gap is homogeneous and no more than 1 nm. The yellow dashed rectangle regions reveal that the possible formation of chemical bonding between the (001)-FS-STO membrane and (110)-STO substrates. We believe that the formation of these bonding at the interfaces can be attributed to the identical chemical composition and short distance between gangling bonds. However, please note that this chemical bonding is discontinuous and unstable due to the polar catastrophe (*37*). The (110) plane of STO substrate is charged, whereas the (001)-FS-STO is charge neutral. Thus, it prohibits forming the stable chemical bonding between (001)-FS-STO membranes and (110)-STO substrates. The naturally formed gap at the interface region allows the FS-STO membranes to keep their own lattice parameters and crystallographic orientations, serving as independent templates for the epitaxial thin film growth.



Fig. S6. Atomic-resolved grain boundary in LSCO hybrid structures. (a) HAADF-STEM image of a representative grain boundary (GB) between two LSCO domains with different orientations. The atom positions are acquired by fitting the intensity peaks with Gaussian function and are embed in the STEM image. The atomic structures around GB are shown in (b). The dashed lines represent the precise position of GB. We believe that the orientation of GB is possibly determined by minimizing misfit strain and thermally stabilization energy between LSCO domains with different orientations.



Fig. S7. HAADF-STEM and geometric phase analysis (GPA) of LSCO hybrid structures. (a) HAADF-STEM image of the (001)-LSCO/(001)-FS-STO/(110)-STO interface region. (b) Inplane and (c) out-of-plane strain distribution within the interface region in (a). The GPA results reveal that the (001)-LSCO layers possess nearly identical in-plane lattice constant of FS-STO membrane and (110)-STO substrates, demonstrating the as-grown (001)-LSCO layers still suffer from the in-plane tensile strain of FS-STO membrane, even though the thickness of membrane is only ~ 8 u. c. The out-of-plane lattice constant of (001)-LSCO layers is smaller than that of FS-STO membranes. (d) HAADF-STEM image of the (110)-LSCO/(110)-STO interface region. (e) In-plane and (f) out-of-plane strain distribution within the interface region in (d). The GPA results indicate that the (110)-LSCO layers are coherently grown on the (110)-STO substates and have the same in-plane lattice constants. The (110)-LSCO layers are tensile-strained, thus the out-of-plane lattice constant of films is smaller than that of (110)-STO substrates. However, the difference between the out-of-plane lattice constants of (110)-LSCO ($c_{LSCO110} \sim 2.68$ Å) and (110)-STO ($c_{\text{STO110}} \sim 2.76$ Å) is small. Therefore, the GPA is insensitive to such small variation when the LSCO films grown along the (110) orientation compared to the films grown along the (001) orientation.



Fig. S8. Microscopic structural characterization of LSCO hybrid structures on (111)oriented STO substrates. (a) Schematic of lateral homogeneitic LSCO hybrid structures grown on (111)-oriented STO substrates. (b) Low-magnified HAADF-STEM image of a LSCO hybrid structure grown on (111)-oriented STO substrates. The dark layer is the (001)-oriented FS-STO membranes, which provides a growth template for the (001)-oriented LSCO layers. (c) Crosssectional STEM image of (111)-LSCO/(111)-STO interface region. The interface between LSCO layers and STO substrates is atomically sharp. The (111)-oriented LSCO layers are structurally uniform. Cross-sectional STEM images of grain boundary (GB) region in a LSCO hybrid structure are shown in (d) and (e). The (001)-FS-STO membrane is twisted unconsciously in a small angle with respect to the STO substrates, i. e. the [100] orientation of FS-STO membranes is not perfectly aligned to the $[11\overline{2}]$ or $[\overline{1}10]$ orientations of (111)-STO substrates. The twisted structures can be arbitrarily controlled through the alignment process during the FS-STO transfer onto the substrates. The imperfect alignment between FS-STO membranes and (111)-STO substrates prevents us acquiring the atomic-resolved STEM images from both interface regions simultaneously. The twisted angle is determined to be ~ 10 degrees by rotating the sample stick stepwise. The STEM image indicates that the uniformed structure within the (001)-LSCO layers and the (001)-LSCO/(001)-FS-STO interface is atomically sharp. From (e), the GB between (001)- and (111)-oriented LSCO layers can be identified clearly. The orientation of GB is determined by the minimum thermally stabilized energy between two different oriented LSCO domains.







Fig. S10. Strain-mediated physical properties of LSCO thin films. (a) XRD 2θ - ω scans around 002 reflections of 30-nm-thick LSCO single films grown on LAO, STO, and KTO substrates. Clear Laue oscillations suggest that all LSCO films are epitaxially grown with high crystallinity. (b) RSMs around 103 reflections of strained LSCO single films. The LSCO films are coherently strained by LAO and STO substrates, yielding to the out-of-plane lattice parameters (*c*) of LSCO films are 3.922 and 3.775 Å, respectively. The *a* and *c* of LSCO films on KTO is 3.891 Å and 3.773 Å, respectively, demonstrating the strain relaxation of LSCO films. Temperature dependent (c) resistivity (ρ) and (d) magnetization (*M*) of LSCO films reveal the

ferromagnetic character and spin-glass behavior at low temperatures. The LSCO films under tensile strains exhibit an insulating behavior, whereas the compressive-strained LSCO film undergoes an insulator-to-metal transition around $T_{\rm C}$ and shows a resistivity upturn at ~100 K. Inset of (d) shows the first derivatives of ρ for LSCO films grown on STO and LAO, yielding to clear slop changes at ~ 175 K, corresponding to $T_{\rm C}$ of films. (e) Magnetoresistance (MR) and (f) *M* of LSCO films as a function of magnetic field at 10 K. MR of LSCO films on KTO is an order of magnitude larger than that of LSCO layers on LAO. We observe a strong reduction of ordered magnetic moment with growing in-plane strain, in consistent with early work (*38*).



Fig. S11. Uniformly distributed magnetization within strained LSCO films probed by PNR. (a) XRR curve of a LSCO film grown on a LAO substrate. The solid line is the best fitting to the experimental data (open circles). The thickness of LSCO film is 30.6 ± 0.4 nm and the surface/interface roughness is ~ 5 Å. Inset of (a) shows the X-ray scattering length density (SLD) depth profile of a LSCO film grown on a LAO substrate. The chemical composition and structural parameters are used for the polarized neutron reflectivity (PNR) fitting. (b) Spin asymmetry (SA) as function of wave vector $q (= 4\pi \sin\theta/\lambda)$, where θ is the incident angle and λ is the wavelength of neutron beam. The SA is derived from the different neutron reflectivities for spin-up (R^+) and spin-down (R^-) polarized neutrons and calculated by $(R^+-R^-)/(R^++R^-)$. The solid red line is the best fit to the experimental data. (c) Magnetization (M) and nuclear SLD (nSLD) depth profile of a LSCO film grown on a LAO substrate. The magnetization (~ 123 emu/cm³) is uniformly distributed within the LSCO film, except for a slightly reduced magnetization at the surface and interface due to the different boundary conditions. This result agrees well with the magnetometry result (~110 emu/cm³ at 1 T) on LSCO films grown on LAO substrates.



Fig. S12. Structural analysis of LSCO hybrid structures on LAO substrates. (a) Schematic of twisted ultrathin FS-STO membranes transferred on (001)-oriented LAO substrates. The twisted angle between [010] orientation of FS-STO membranes and [010]_{pc} orientation of LAO substrates is $\sim 16^{\circ}$, determined by sample rotation in the microscopy (b) HAADF-STEM image of LSCO hybrid structures on LAO substrates. The brighter regions in the STEM image are LAO and LSCO due to the stronger electron scattering from heavier elements (La). An atomicresolution STEM image at the interface region is present in (c). We plot the intensity line profiles along (d) the out-of-plane direction, (e) LSCO film region, and (f) STO membrane region. The averaged in-plane lattice parameter of (001)-oriented LSCO layer is ~ 3.86 Å, which is slightly smaller than that of FS-STO membrane (~ 3.87 Å). Although the in-plane strain is slightly relaxed, the LSCO layers still suffer from the tensile strain of STO membranes. This case is different from a LSCO single film with identical thickness coherently strained to the STO single crystal substrates. The thickness of STO membranes (~ 8 u. c.) is an order of magnitude smaller than the thickness of LSCO layers (~ 80 u. c.). The lattice structure of STO membranes may deforms slightly to comply the elastic misfit strain between STO and LSCO. A small lattice elongation of FS-STO membrane is observed along the out-of-plane direction. In addition, we note that the gap between FS-STO and LAO substrates is ~ 2 nm in thickness. There is no apparent chemical bonding between FS-STO and LAO.



Fig. S13. Strain distribution within an LSCO layer grown directly on LAO substrates. (a) Schematic of sample configuration. (b) A HAADF-STEM image of the interface region between LSCO layers and LAO substrates. Since the heavy elements in LSCO and LAO are identical (La), it is difficult to distinguish the interfaces between those two materials. We perform the geometric phase analysis (GPA) at the same region. (c) and (d) show the in-plane and out-of-plane strain distributions at the interface region, respectively. The LSCO layers are coherently compressive-strained to LAO substrates. The in-plane lattice constant of LSCO layers is identical to that of LAO substrates. Thus, the LSCO layers is elongated along the out-of-plane direction. We determine that the out-of-plane lattice constant of LSCO layers is approximately 3.92 Å, which is significantly larger than that of LAO substrates (~ 3.79 Å).



Fig. S14. Regional dependent electronic states of LSCO hybrid structures. (a) and (b) XAS at Co L_{3,2}-edges measured at the edge and center regions of LSCO hybrid structures on LAO substrates, respectively. The edge region of LSCO hybrid structure is the direct growth of (001)-LSCO layers on (001)-oriented LAO substrates. The LSCO layers is compressively strained, similar to the LSCO single films grown on the LAO substrates. The center region of the LSCO hybrid structure possesses the (001)-LSCO layers grown on the FS-STO membranes. In this case, the (001)-LSCO layers at the center region is slightly tensile-strained to the FS-STO membranes. The distinct strain states of LSCO layers determine the opposite XLD at different regions [(c)]. In the center region of LSCO hybrid structure, the electrons prefer occupying the $d_{x^2-v^2}$ orbitals, whereas the electron occupancy in the $d_{3z^2-r^2}$ orbitals is larger at the edge region of LSCO hybrid structure. The control experiments were conducted on a LSCO hybrid structure grown on KTO substrates [(d) and (e)]. Both FS-STO membranes and KTO substrates apply the in-plane tensile strain to the LSCO layers. The signs of XLD are the same for the edge and center regions of LSCO hybrid structures. The amplitude of XLD at the edge region (LSCO/KTO) is larger than that of XLD measured at the center region ((001-)LSCO/FS-STO) because the LSCO layers suffer a larger tensile strain from the KTO substrates. Thus, more electrons occupy the $d_{x^2-y^2}$ orbitals at the edge regions compared to the center regions.



Fig. S15. Electronic states of strained LSCO single films. (a) Schematic of electronic states under different strain states. The X-ray beam with linearly polarization incidents on the sample surface. The X-ray beam is parallel to the surface normal or has an incident angle of 60° with respect to the surface normal. The electronic state of Co 3d orbitals is probed by measuring the resonance X-ray absorption spectra. When the X-ray beam is parallel to the surface normal, the XAS (I_0°) reflects the orbital occupancy of Co $d_{x^2-y^2}$ orbital directly. When the X-ray beam incidents on the sample surface with an angle of 60° , the XAS ($I_{60^{\circ}}$) contains both orbital information from Co $d_{x^2-v^2}$ and Co $d_{3z^2-r^2}$ orbitals. (b), (d), and (f) XAS at Co $L_{3,2}$ -edges for LSCO films grown on (001)-oriented LAO, STO, and KTO substrates, respectively. Apparently, the XAS signals from Co^{3+} ions dominate the entire XAS spectra due to the 80% Co ions is +3 and only 20% Co ions is +4. The black and red curves represent the XAS $(I_{0^{\circ}})$ and XAS $(I_{60^{\circ}})$, respectively. In LSCO/LAO, the intensity and peak energy of XAS (I_{60}°) are smaller than those of XAS (I_0°) , indicating that the most of holes occupy the $d_{x^2-y^2}$ orbitals and the free electrons occupy the $d_{3z^2-r^2}$ orbitals. By contrast, the electrons will occupy the $d_{x^2-v^2}$ orbitals when the LSCO films are tensile-strained. (c), (e), and (g) X-ray linear dichroism (XLD) for the LSCO films grown on (001)-oriented LAO, STO, and KTO substrates, respectively. The XLDs are simply calculated by $I_{0^{\circ}} - I_{60^{\circ}}$. The orbital polarization of LSCO films varies with the strain states. XLD results reinforce the predominant $d_{x^2-y^2}$ (or $d_{3z^2-r^2}$) hole character in compressively(or tensile) strained LSCO films.



Fig. S16. Transport measurements of LSCO hybrid structures. (a) Schematic setups for transport measurements at edge and center regions of LSCO hybrid structures. (b) Temperature dependent resistivity (ρ) of a (001)-oriented LSCO single film, the edge and center regions of LSCO hybrid structures on LAO substrates. The transport measurements were performed in van der Paw method with applying the out-of-plane magnetic fields. The ρ of LSCO hybrid structures is larger than that of a (001)-oriented LSCO single film grown on LAO because the LSCO layers under tensile strain states exhibit an insulating behavior in contrast to the metallic conductivity of a compressively strained LSCO layers. (c) Magnetoresistance $[MR = (\rho_{7T} - \rho_{0T}) \times 100\% / \rho_{0T}]$ of a (001)-oriented LSCO single film, the edge and center regions of LSCO hybrid structures on LAO substrates at different temperatures. The MR of LSCO hybrid structures is a few times larger than that of a LSCO film grown on LAO substrates at low temperatures. This fact agrees well with the reduction of ordered magnetic domains as increasing in-plane tensile strain. The difference between MRs of (001)-LSCO/LAO and LSCO hybrid structures becomes small at high temperatures. The physical picture is further reinforced by performing transport measurements on a LSCO hybrid structure grown on KTO substrates [(d) and (e)]. The ρ of LSCO hybrid structures reduces by less than an order of magnitude compared to that of a LSCO single film grown on KTO substrates. Meanwhile, the MR of LSCO hybrid structure is slightly smaller than that of a LSCO single film on KTO. These results support the strain-driven suppression of the ferromagnetic phase in LSCO layers.



Fig. S17. Anomalous Hall resistivities of LSCO single films and hybrid structures. Schematic measurement configurations for (a) LSCO single films grown on (001)-LAO substrates (LSCO/LAO), (b) the edge region and (c) center region of LSCO hybrid structure grown on LAO substrates. (d), (e), and (f) Field-dependent Hall conductivities at different temperatures for a LSCO/LAO, the edge and center regions of LSCO hybrid structures, respectively. The R_0H is subtracted from ρ_{xy} by linearly fitting the data at high magnetic field region. The (ρ_{xy} - R_0H) at 7 T for the LSCO hybrid (edge) and LSCO hybrid (center) are summarized in Figs. 4C and 4E of main text.



Fig. S18. Diamond NV magnetometry on LSCO hybrid structures. Nanodiamonds (NDs) with ensemble NV centers were dispersed randomly on the surface of LSCO hybrid structures on LAO substrates. (a) and (b) Optical microscopy images of NDs at the edge and center regions of LSCO hybrid structures. Assisted from motorized precision translation stages at the microscale, we could select specific NDs at the edge and center regions of LSCO hybrid structures and investigate their temperature dependencies. (c) and (d) Zero-field optically detected magnetic resonance (ODMR) spectra of selected NDs at the edge and center regions of LSCO hybrid structures when *T* increases from 50 to 250 K. As increasing temperature, the splitting (2γ B) of ODMR spectra reduces. The temperature dependences of 2γ B at edge and center regions of LSCO hybrid structures are plotted in Fig. 4I of main text.



Fig. S19. Fabrication of LSCO hybrid structures on arbitrary substrates. We followed the same protocol as the (001)-LSCO hybrid structures grown on other single crystalline substrates. Firstly, we fabricated the (110)- and (111)-oriented STO/SAO bilayers on the relevant substrates. Then, we dissolved the SAO and support the ultrathin STO membranes using thermally-release tapes. The (110)- and (111)-oriented FS-STO membranes were transferred to the (001)-oriented STO substrates. Finally, we deposited the LSCO layers on the modified (001)-oriented STO substrates using PLD technique. (a) and (b) RSMs around 002 reflections of (110)- and (111)oriented 30-nm-thick LSCO hybrid structures grown on the (001)-oriented STO substrates, respectively. The (110)- and (111)-reflections from FS-LSCO/STO membranes are observed except for (002) reflections of LSCO layers and STO substrates, suggesting that different oriented LSCO layers can be successfully fabricated on the (001)-oriented STO substrates. Another example is the fabrication of epitaxial (001)-LSCO layers on the SiO₂/Si substrates. Typically, it is challenging to grow a functional oxide film directly on the silicon or silicon oxide substrates due to the enormous misfit strain. Using a freestanding STO membrane, we were able to transfer it onto the SiO₂/Si substrates and then fabricated high-quality single-crystalline functional oxides. (c) XRD 2θ-ω scan of a LSCO hybrid structure on SiO₂/Si substrates. A XRD 2θ - ω curve of A (001)-oriented LSCO single film is plotted as a direct comparison. Only (00*l*) reflections of LSCO layers are observed, suggesting the epitaxial growth of LSCO layers. These results indicate that the method in present work is readily applicable to any arbitrary substrates.



Fig. S20. Control of crystallographic orientation of freestanding membranes with respect to the substrates. (a) Schematics and (b) optical images of (001)-oriented STO freestanding membranes attached to (110)-oriented STO substrates, respectively. The (100) orientation of freestanding membranes rotates gradually by an angle (α) of 0°, 30°, 45°, 60°, and 90°, with respect to the $[1\overline{1}0]$ orientation of substrates. (c) XRD θ -2 θ scans of LSCO hybrids grown on above modified substrates. Both (110) and (002) diffraction peaks from LSCO layers can be observed, indicating the robust methodology for fabrication LSCO hybrid structures with different orientations. Phi scans are performed at different LSCO hybrid structures to determine the orientation of LSCO layers grown on freestanding membranes. For (110)-oriented LSCO layers, the 013 diffraction peaks show two-fold symmetry and appear at the fixed phi angles (0°) and 180°). However, for (001)-oriented LSCO layers, the 013 diffraction peaks exhibit four-fold symmetry. The diffraction peaks separate with an integral of 90° in each phi scan. We find the four peaks are left-shifted by 30°, 45°, 60°, and 90° with respect to those of LSCO hybrid structures ($\alpha = 0^{\circ}$). The diffraction patterns are identical for LSCO hybrid structures ($\alpha = 0^{\circ}$ and $\alpha = 90^{\circ}$). The structural characterizations illustrate clearly the successful control of crystallographic orientation of freestanding membranes (and the layer grown on top) with respect to the substrates.

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