Supporting Information

Epitaxially Grown Single-Crystalline $SrTiO_3$ Membranes Using a Solution-Processed, Amorphous $SrCa_2Al_2O_6$ Sacrificial Layer

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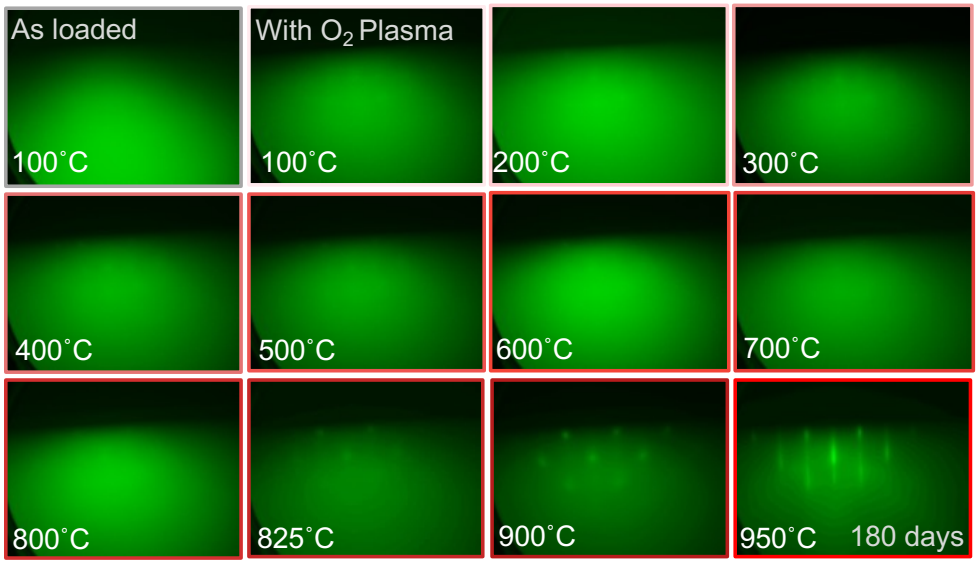

Figure S1: RHEED evolution as a function of substrate temperature during oxygen plasma annealing of $SrCa_2Al_2O_6$ on STO (001) substrate. The substrate temperature at which the image is taken is indicated on the image. All images are taken along [100] azimuthal direction.

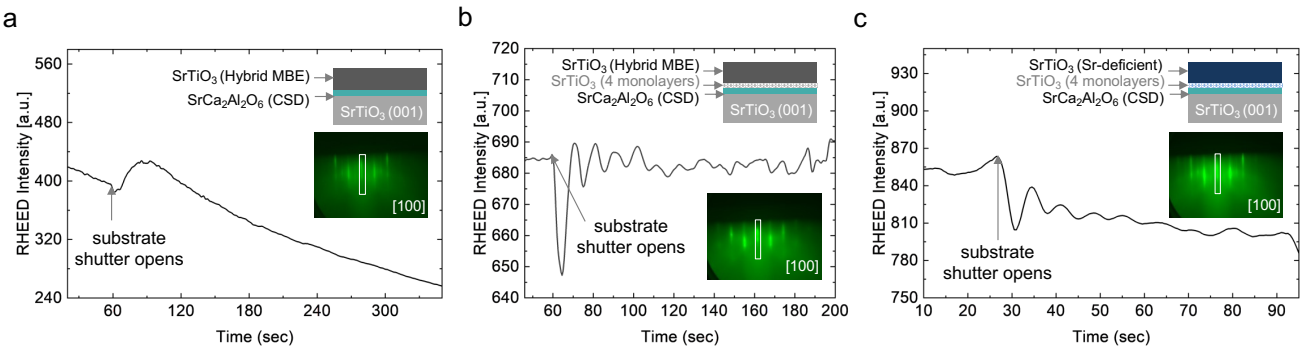

Figure S2: Time-dependent RHEED intensity profile during STO growth. (a) when STO is grown directly on annealed $SrCa_2Al_2O_6$. (b) when STO is grown on a 4-monolayers of STO on $SrCa_2Al_2O_6$. (c) when Sr-deficient STO is grown on a 4-monolayers of STO on $SrCa_2Al_2O_6$. Inset of (a, b, c) show a sample schematic and the RHEED pattern with a box where time-dependent intensity was recorded.

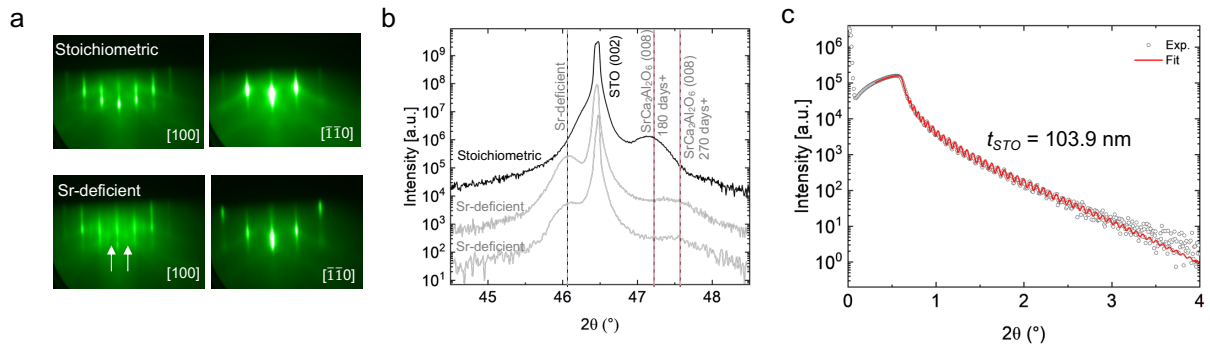

Figure S3: RHEED and X-ray diffraction of Sr-deficient and stoichiometric STO / $SrCa_2Al_2O_6$/ STO (001). (a) RHEED images after STO growth taken at 100 ˚C along [100] and [$\bar{1}\bar{1}$0] directions. (b) 2θ-ω coupled scan of as-grown films. The film with TTIP/Sr ratio of 103.84 is stochiometric, the out-of-plane lattice parameter extracted from STO (002) peak is 3.905 ± 0.002 Å and from $Sr_xCa_{3-x}Al_2O_6$ (008) is 15.39 ± 0.002 Å (÷4 = 3.849 Å), indicating x ~ 0.9 (assuming relaxed lattice parameter). The film with TTIP/Sr ratio of 134.47 is Sr-deficient, the lattice parameter extracted from STO (002) peak is 3.936 ± 0.002 Å and from $Sr_xCa_{3-x}Al_2O_6$ (008) is 15.30 ± 0.002 Å (÷4 = 3.825 Å), indicating x ~ 0.3 (assuming relaxed lattice parameter). This film is grown after 270+ days of $SrCa_2Al_2O_6$ sample preparation, and nominal deviation from peak position is expected in hygroscopic samples. The reduction in intensity of SCAO suggests change in crystallinity over the period of time. (c) x-ray reflectivity scan of 100 nm STO/ 20 nm $SrCa_2Al_2O_6$/ STO (001) substrate with a fit done using GenX software, indicating a thickness of 103.9 nm.

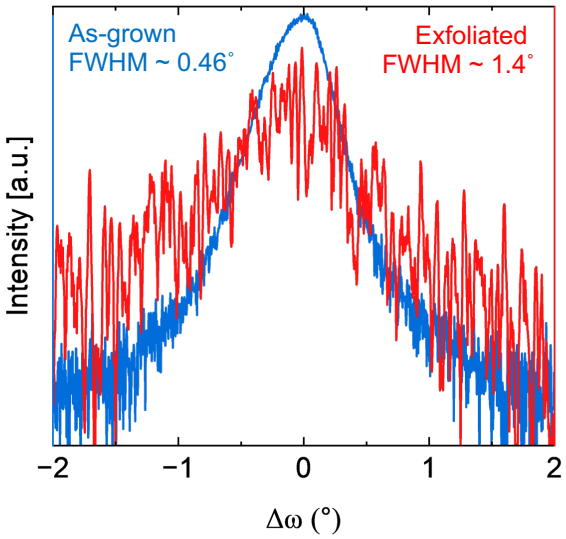

Figure S4: Rocking curve of as-grown STO film and after exfoliation and transfer onto a Au-coated Si substrate. The FWHM of the film has changed from 0.46˚ to 1.40˚.

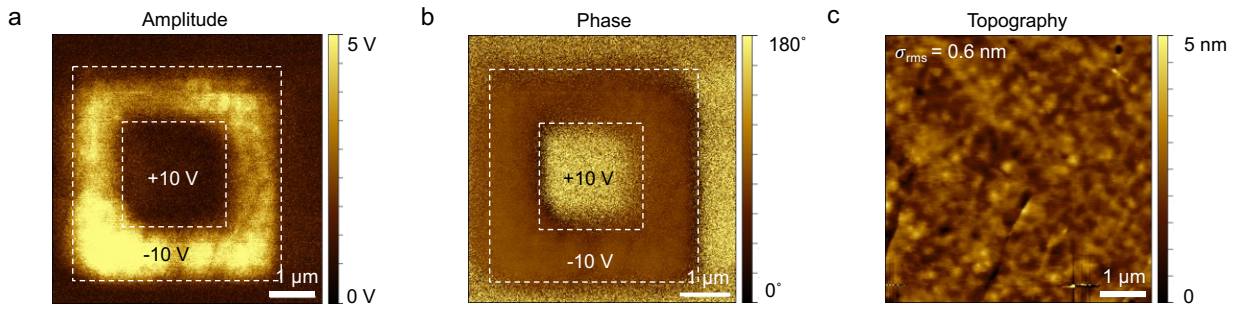

Figure S5: Box-pattern on Sr-deficient STO membrane on a different region than shown in Figure 4. First, a 4 × 4 μm^2 square area was scanned with -10 V tip bias with bottom electrode grounded. A 2 × 2 μm^2 square area that was scanned with +10 V tip bias. Finally, a 6 × 6 μm^2 area was scanned with 0 V tip bias. The 6 × 6 μm^2 area scan with 0 V also captures the pristine state of the sample. And the -10 V and +10 V region represents the polled state of the sample. (a) Amplitude demonstrating contrast between regions that were scanned with -10 V and 10 V. (b) Phase demonstrating contrast of nearby 180˚ between the region that was scanned with -10 V and 10 V. (c) Topography scan of the membrane done during 0 V scan. This scan further reveals ferroelectric-like switching on a different region on Sr-deficient membrane.

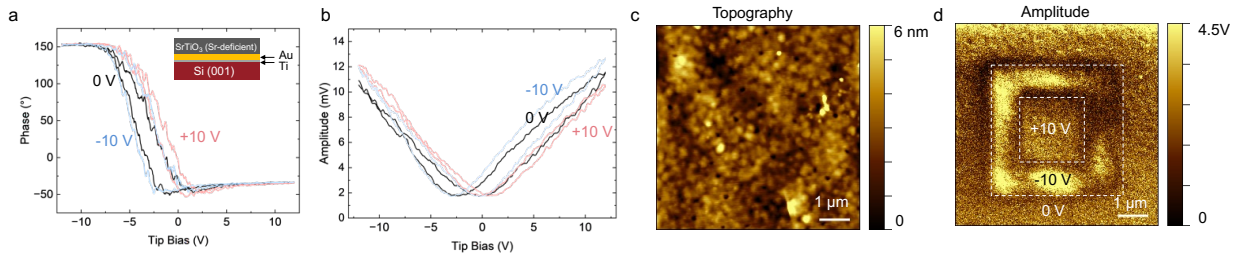

Figure S6: PFM hysteresis loops, topography, and amplitude on Sr-deficient STO membrane on the region shown in Figure 4a. (a) Phase and amplitude hysteresis loops obtained on Sr-deficient STO membrane on a surface polled to -10 V, +10 V, and pristine surface (0 V). The scans on the polled region were done after 30 minutes of polling process. A 180˚ flipping is observed in phase angle in all three areas of the sample, -10 V (polled), +10 V (polled), and 0 V (pristine). These loops demonstrate switching in the sample at various regions. (b) Amplitude and bias butterfly loops done on three areas of the sample as discussed in Figure S6a. (c-d) Topography and amplitude of membrane after box-writing shown in Figure 4a. A minimal contrast is observed in amplitude when +10 V and 0 V compared since the pristine state of the sample could have dipoles in the out-of-plane direction.