Supplementary Information

Tailoring the crystal growth of quartz on silicon for patterning piezoelectric films

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Table S1. Correspondence between the molarity of the SrCl\textsubscript{2} aqueous solution and the Sr to SiO\textsubscript{2} molar ratio in the final solution used in the dip-coating process.

| Concentration of the SrCl\textsubscript{2} aqueous solution (Solution B) | Sr/Si molar ratio in the final solution (Solution C) |
|-------------------------------------------------|---------------------------------------------------|
| 0.4 M                                           | 0.02                                              |
| 0.5 M                                           | 0.025                                             |
| 0.6 M                                           | 0.03                                              |
| 0.65 M                                          | 0.0325 (shown as 0.033 in paper)                   |
| 0.7 M                                           | 0.035                                             |
| 0.75 M                                          | 0.0375 (shown as 0.038 in paper)                   |
| 1 M                                             | 0.05                                              |
| 1.5 M                                           | 0.075                                             |
| 2 M                                             | 0.1                                               |
**Figure S1.** SEM image in secondary electrons mode of a film without Sr, showing the presence of cracks and pores.

**Figure S2.** XPS overview of $R_{Sr} = 0.05$ film to show the present of Sr components.
inside film.

Figure S3. Topography study for different $R_{Sr}$: (a) The relationship between with $R_{Sr}$ and film roughness ($R_{rms}$) (b) Topographic AFM image of the porous film ($R_{Sr}=0.033$) with the lowest $R_{rms}$, (c) Topographic AFM image of a dense film (with $R_{Sr}=0.1$) displaying the largest $R_{rms}$.

Figure S4. Pole figure of (101) $\alpha$-quartz for two characteristic microstructures: (a) porous film ($R_{Sr} = 0.033$), (b) dense film ($R_{Sr} = 0.05$).
Figure S5. Optical images of partially crystallized films when $R_{Sr}$ is (a) 0.02, (b) 0.025, (c) 0.03. In these images, only the light color areas are crystallized to $\alpha$-quartz phase, the remaining violet and brown areas are still amorphous. (d) is the SEM image of one selected zone of $R_{Sr} = 0.02$ condition film which containing crystalline structure (dark area) and amorphous surface (light area) for EBSD test. (e) is the EBSD result of point A in (d) with $\alpha$-quartz phase signals. (f) is the EBSD result of point B in (d) without any signal.
**Figure S6.** SEM images in secondary electrons mode of $R_{sc} = 0.035$ films crystallized with different times at 1000°C: (a) 40 min. (b) 50 min. (c) 60 min.
Figure S7. SEM images in secondary electrons mode of $R_{Sr} = 0.05$ films crystallized with different annealing times at 1000°C: (a) 20 min. (b) 30 min. (c) 60 min.
Figure S8. Topographic AFM images showing the influence of annealing time at 1000°C for (a) porous microstructure (obtained with $R_{Sr} = 0.035$), (b) dense microstructure (obtained with $R_{Sr} = 0.05$).

Figure S9. SEM pictures of gel films with $R_{Sr} = 0.05$ (upper panel) and the corresponding annealed films for 300 min at 1000°C (lower panel) with optical images in as an inset using three different three surfactants: (a) Brij-58, (b) F127, (c) CTAB.
Figure S10. SEM pictures of films with $R_{Sr} = 0.05$ prepared at three different relative humidities indicated in the axis next to the images: (a) gel films (b) after annealing for 300 min at 1000°C.
Figure S11. Thickness measurement of gel films with $R_{Sr} = 0.05$ and prepared using different withdrawal speeds showing the increasing gel film thickness with $U_w$: and the different regimes: the viscous drag regime for $U_w > 4 \text{ mm/s}$ and the mixed regime, for $U_w < 4 \text{ mm/s}$. 

The linear relationship is given by $\lg T = 1.851 + 0.643 \times \lg U_w$ with $R^2 = 0.9987$. 

![Graph showing thickness measurement and linear relationship](image-url)
**Figure S12.** The FWHM analysis of $R_{Sr} = 0.05$ films made by different withdraw rates is shown as (a). Meanwhile, the raw data of rocking curves at $\alpha$-quartz (100) peak are shown as (b).

**Figure S13.** A 2 inch epitaxial quartz thin film on silicon substrate fabricated by 5 multilayer deposition, reaching a final thickness of 800 nm (a). Heat map of peak intensity of (100) quartz reflection (b). Heat map of evaluated peak width (FWHM) of rocking curves around quartz (100) reflection (c).
**Figure S14.** (a) Optical image of Si (100) masters used along this work obtained by using LIL lithography. (b) FEG-SEM image of printed Sr-doped silica columns with a diameter of 1 µm and height of 2 µm on silicon. The inset pictures shows FEG-SEM images at higher magnification of pillars. (c) 3D AFM images of the nanostructured silica films prepared by NIL lithography.