Supporting Information

**Strain effect on structural transition in SrRuO$_3$ epitaxial thin films**

Daisuke Kan* and Yuichi Shimakawa

Institute for Chemical Research, Kyoto University, Uji, Kyoto, 611-0011 Japan

*E-mail dkan@scl.kyoto-u.ac.jp

**X-ray structural analysis for SrRuO$_3$ (110)$_{orth}$ epitaxial thin films at room temperature**

To reproduce the observed x-ray 2theta-theta profiles, we performed calculations using a structural model depicted in Fig. S1a. We assume tetragonal SrRuO$_3$ unit cells ($a_0 \times a_0 \times c$) with an in-plane lattice parameter identical to that of the SrTiO$_3$ substrate ($a_0 \times a_0 \times a_0$; $a_0 = 0.3905$ nm). In this model, the atomic coordinates for the SrRuO$_3$ unit cell are Sr (1/2, 1/2, 1/2), Ru (0, 0, 0), O1(1/2, 0, 0), O2(0, 1/2, 0), and O3(0, 0, 1/2).

\[
F_{SRO}(00L) = \left\langle f_o \exp\left[-2\pi\tilde{a}'c\left(-c/2\right)\right] + f_n \left[2 + \exp\left[-2\pi\tilde{a}'c\left(-c/2\right)\right]\right]\sum_{L=1}^{N} \exp(-2\pi(k+1)La'c) \right\rangle
\]
The structure factor of the N unit cell thick SrRuO$_3$ film layer contributing to (00$L$)$_{pc}$ scattering, $F_{SRO}(00$L$)$_{pc}$, is written as

\begin{equation}
    F_{SRO}(00$L$)$_{pc}$ = f_{Sr} + f_{Ru} + f_{O}
\end{equation}

where $f_{Sr}$, $f_{Ru}$, and $f_{O}$ are atomic scattering factors for the Sr, Ru, and O atoms, respectively. The $a^*$ is the reciprocal lattice unit defined by $a^* = 1/a_0$. For the calculation, the origin of the z axis, is set at the Ti position at the interface between the SrRuO$_3$ layer ($Z > 0$) and SrTiO$_3$ substrate region ($Z \leq 0$).

For $Z \leq 0$, the structure factor, $F_{STO}(00$L$)$_{pc}$, is given by

\begin{equation}
    F_{STO}(00$L$)$_{pc} = \sum_{k \neq 0} \exp\left(-\frac{2\pi L a^*}{a_0} \right)
\end{equation}

Thus the scattering intensity $I(00$L$)$ is calculated to be

\begin{equation}
    I(00$L$) = A \cdot |F_{SRO}(00$L$)$_{pc} + F_{STO}(00$L$)$_{pc}|^2
\end{equation}

where the coefficient $A$ is a scale factor.

A typical example of the calculated 2theta-theta profiles for the 11.9nm-thick film is displayed in Fig. S1b together with the experimental results. The calculation reproduces the experimentally observed profiles (blue line) well although the orthorhombic unit cell is not taken into account. It is worthwhile to note that the
temperature-induced evolution of $d_{001\text{pc}}$ in Fig 1 in the main text is independent of the structural model (Fig. S1a) for the calculations.

We have also performed reciprocal space mappings (RSMs) around the SrTiO$_3$ (204)$_{pc}$ Bragg reflection at 30 °C. Figures S2 and S3 display the semilogarithmic counter plots of the RSM around SrTiO$_3$ (204)$_{pc}$ with the phi configurations of 0, 90, 180 and 270° for the 41.1 and 12.6 nm-thick films, respectively. Since the data were taken at 30 °C, the films are in the orthorhombic structural phases with the dimension of $\sqrt{2}a_0 \times \sqrt{2}a_0 \times 2a_0$. All reflections from the films appear in the same position in $Q_{100\text{pc}}$, indicating that in-plane lattice parameters $d_{001\text{ortho}}$ and $d_{1-10\text{ortho}}$ of the films are locked by the substrates. Because this orthorhombic phase grows with (110)$_{\text{ortho}}$ orientation on the SrTiO$_3$ (001)$_{pc}$ substrate, the difference in peak position along $Q_{001\text{pc}}$ depending on the phi setting is clearly seen. It should be noted that in the case for the thinner films, (444)$_{\text{ortho}}$ and (44-4)$_{\text{ortho}}$ reflections, of which positions are the same for the thicker films, are seen at slightly different positions in $Q_{001\text{pc}}$. The appearance of (444)$_{\text{ortho}}$ and (44-4)$_{\text{ortho}}$ reflections in the different $Q_{001\text{pc}}$ positions was also confirmed for the films thinner than 20 nm. Figure S4 shows examples of the (20$L$)$_{pc}$ line scan profiles observed for the 12.6 nm-thick film. These observations indicate that the orthorhombic unit cell...
for the thinner films is distorted along the [001]$_{\text{ortho}}$ direction as shown in Fig.4 in the main text.

Figure S1 : (a) Schematic of structural model for the x-ray 2theta-theta profile calculation. (b) Calculated x-ray 2theta-theta profile (red line) around the STO (002)$_{pc}$ Bragg reflection, together with the experimentally observed 2theta-theta profile at room temperature for the 11.9 nm-thick films grown on SrTiO$_3$ (001)$_{pc}$ substrate.
Figure S2: Reciprocal space mappings around the SrTiO\textsubscript{3} (204)\textsubscript{pc} Bragg reflection for 41.1 nm-thick SrRuO\textsubscript{3} film at 30 °C. The mappings were acquired with configurations of phi = 0, 90, 180 and 270 °.
Figure S3: Reciprocal space mappings around the SrTiO$_3$ (204)$_{pc}$ Bragg reflection for 12.6 nm-thick SrRuO$_3$ film at 30 °C. The mappings were acquired with configurations of phi = 0, 90, 180 and 270 °.
Figure S4 : $(20L)_{pc}$ line scans taken for 12.6 nm thick SRO thin films at 30 °C. The $(444)_{ortho}$ and $(44-4)_{ortho}$ reflections were observed by conducting the line scans around the STO $(204)_{pc}$ Bragg reflections with configurations of phi = 0, and 180 °.