Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

Spin-phonon coupling in epitaxial $SrRuO_3$ heterostructures

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Supplementary Note 1 | Raman tensor calculation of SRO for the relative polarization dependencies.

The intensity of each Raman mode is proportional to $\left|<\hat{e}_s\,|\,R\,|\,\hat{e}_i>\right|^2$, where $\hat{e}_i$ ($\hat{e}_s$) is the polarization of the incident (scattered) photons, and R is the Raman tensor of each mode. The Raman tensors can be written as

$$R\,(A_g)=\begin{pmatrix} a & 0 & 0 \\ 0 & b & 0 \\ 0 & 0 & c \end{pmatrix},\qquad R\,(B_{1g})=\begin{pmatrix} 0 & d & 0 \\ d & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

$$R\,(B_{2g})=\begin{pmatrix} 0 & 0 & e \\ 0 & 0 & 0 \\ e & 0 & 0 \end{pmatrix},\qquad R\,(A_g)=\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & f \\ 0 & f & 0 \end{pmatrix}, \tag{S1}$$

where a, b, c, d, e, and f are constants. Therefore, the Raman intensities depending on the polarization angle of the A_g mode, for example, can be calculated as

$$I(A_g) \propto \left| \begin{pmatrix} \frac{1}{\sqrt{2}}\cos\theta_s & \sin\theta_s & -\frac{1}{\sqrt{2}}\cos\theta_s \end{pmatrix} \begin{pmatrix} a & 0 & 0 \\ 0 & b & 0 \\ 0 & 0 & c \end{pmatrix} \begin{pmatrix} \frac{1}{\sqrt{2}}\cos\theta_i \\ \sin\theta_i \\ -\frac{1}{\sqrt{2}}\cos\theta_i \end{pmatrix} \right|^2 . \quad \text{(S2)}$$

$$= \left| \frac{1}{2}(a+c)\cos\theta_s \cos\theta_i + b\sin\theta_s \sin\theta_i) \right|^2$$

In the case of relative polarization dependence, the relative polarization θ_r should be equal to the value of $\theta_i + \theta_r$, where θ_i and θ_s are the polarizations of the incident and scattered photons, respectively. Therefore, the intensity of the A_g mode is proportional to $\left|\frac{1}{2}(a+c)\cos\theta_r\right|^2$. Similarly, the intensity depending on the relative polarization angle of the B_{2g} mode is proportional to $\left|\frac{1}{2}e\cos\theta_r\right|^2$, and the intensities of the B_{1g} and B_{3g} modes are proportional to $\left|\frac{1}{\sqrt{2}}d\sin\theta_r\right|^2$ and $\left|\frac{1}{\sqrt{2}}f\sin\theta_r\right|^2$, respectively.

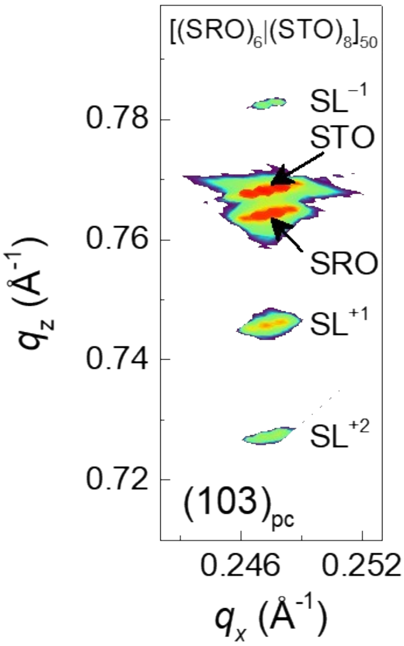

Fig. S1 | XRD RSM of the SRO/STO SL.

RSM of the $[(SRO)_6|(STO)_8]_{50}$ SL over a wider q_z range. The Bragg peaks of the SL ($SL^{\pm n}$) indicate a fully strained state.

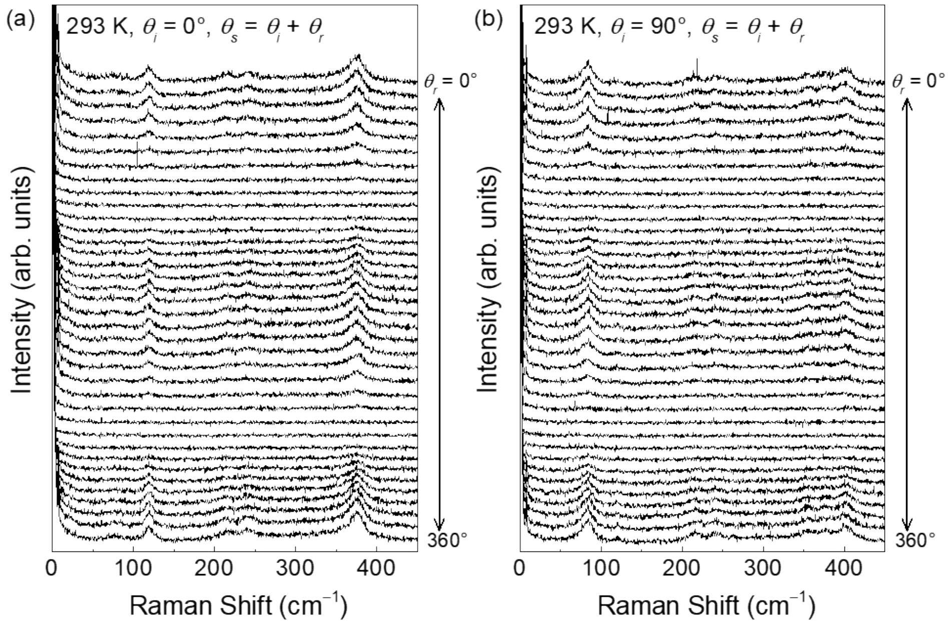

Fig. S2 | Polarized Raman spectra of SRO for different relative polarizations between incident and scattered photons.

Angle-resolved polarized Raman spectra of the SRO single film measured at 293 K as a function of the relative polarization θ_r between the incident (θ_i) and scattered (θ_s) photon polarizations ($\theta_s = \theta_i + \theta_r$) for θ_i = (a) 0° and (b) 90°. In the relative polarization configuration, both $I\,(A_g)$ and $I\,(B_{2g})$ are proportional to $|\cos^2\theta_s|$. This indicates that all observed SRO phonon modes can be attributed to either A_g or B_{2g} symmetry.

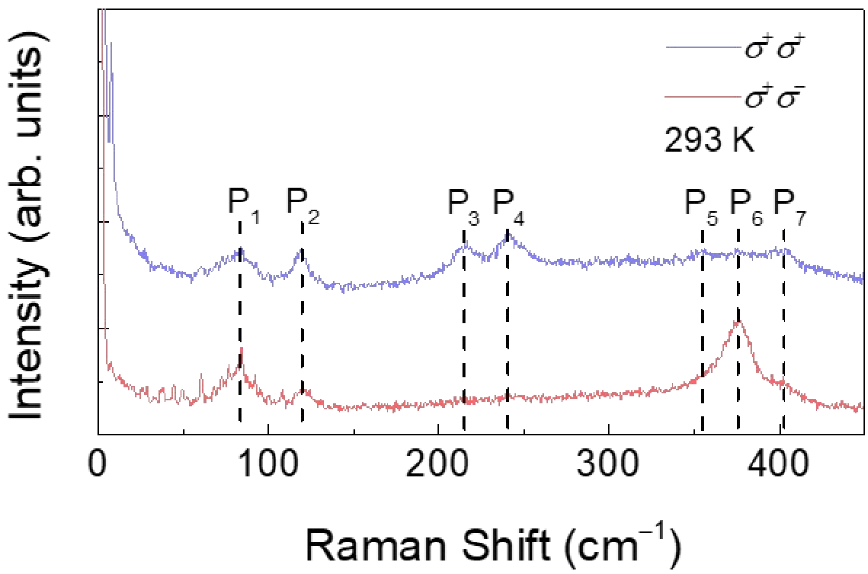

Fig. S3 | Circularly polarized Raman spectra for SRO.

Circularly polarized Raman spectra of the SRO single film measured at 293 K. The P_2 and P_6 modes have different intensities in the two polarization configurations, meaning these modes should be attributed to A_g symmetry. The $(\sigma^+\sigma^+)$ $[(\sigma^+\sigma^-)]$ configuration indicates the same (opposite) circular polarization between the incident and scattered photons.

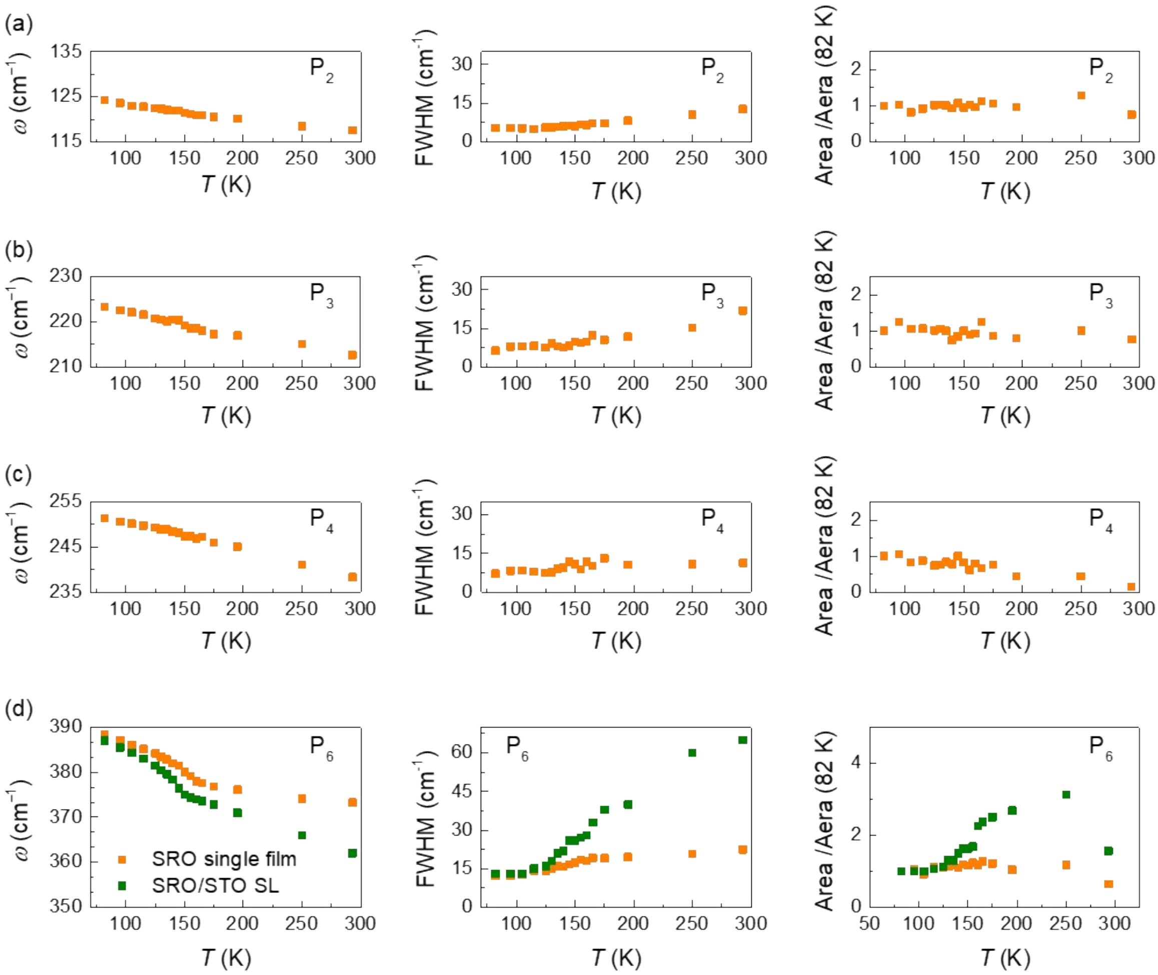

Fig. S4 | Lorentzian fitting parameter for SRO phonon modes.

T-dependent phonon dynamics of the SRO single film (Fig. 3(a)) obtained by fitting Lorentzian oscillators. The peak positions (ω), FWHM values, and normalized intensities of (a) P_2, (b) P_3, (c) P_4, and (d) P_6 are displayed as functions of T, respectively. To investigate the spin-phonon coupling in the SL, we further analyzed the parameters of P_6 for the SL in (d).

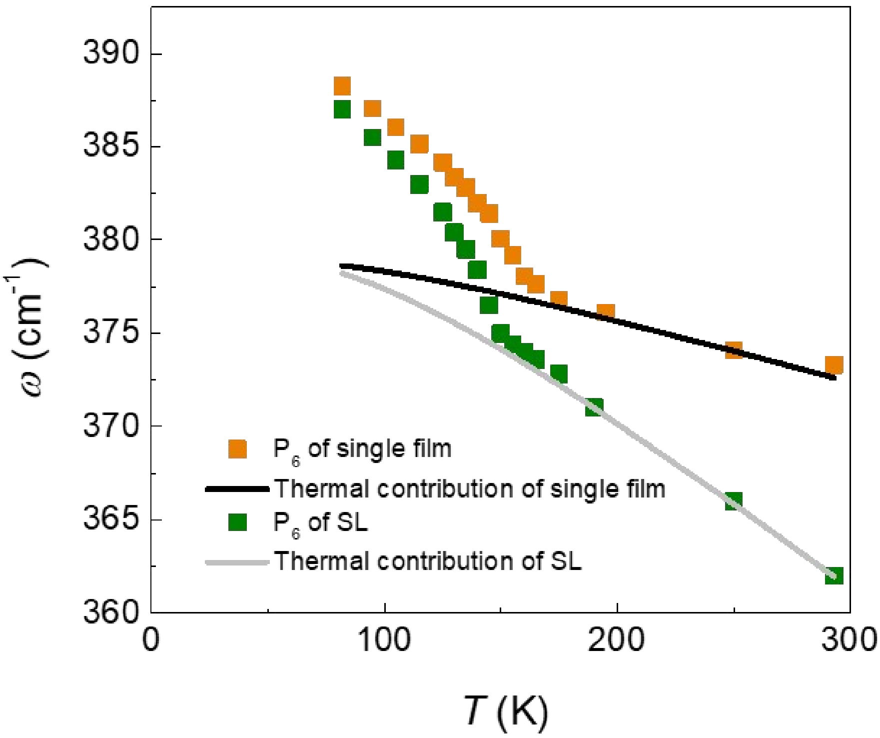

Fig. S5 | Anharmonic contribution of the P_6 mode.

An anharmonic phonon scattering model was used to remove the thermal contributions.[S1] The anharmonic contribution assuming three phonon processes can be described as,

$$\omega(T) = \omega_0 - A\left[1 + \frac{2}{e^{\frac{\hbar\omega_0}{2k_BT}} - 1}\right], \tag{S3}$$

where $\hbar$ and k_B are the Planck and Boltzman constants, respectively, and A and ω_0 are fitting parameters. We subtracted the anharmonic contributions (solid lines) from the T-dependent phonon frequencies ω (symbols) of the P_6 mode for the SRO single film and SL.

Reference

S1. M. Balkanski, R. F. Wallis and E. Haro, *Phys. Rev. B*, 1983, **28**, 1928-1934.

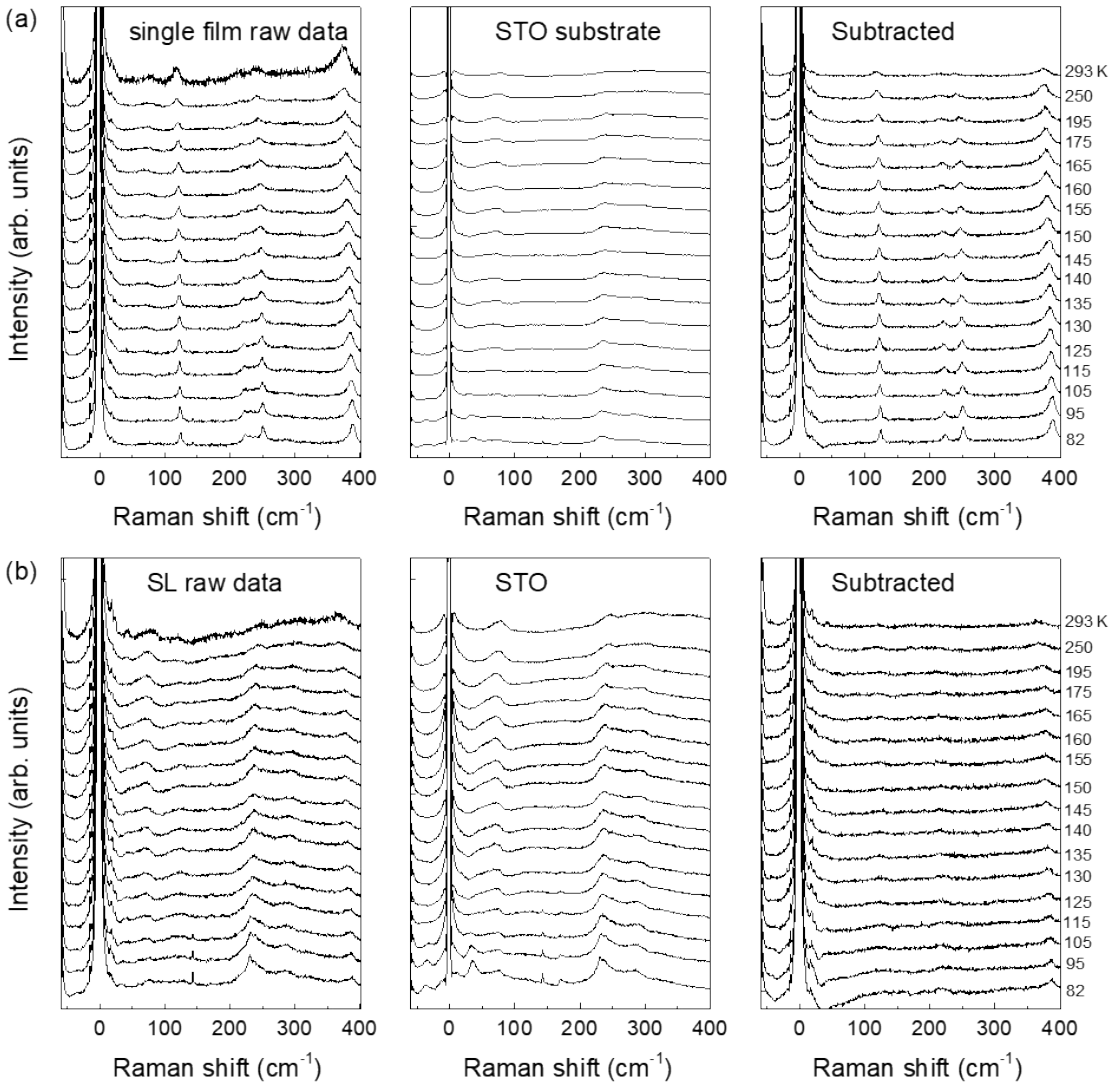

Fig. S6 | Subtraction of STO contributions from Raman spectra of SRO heterostructures. *T*-dependence of the Raman spectra of (a) SRO single film and (b) SL obtained by subtracting the STO contributions at the corresponding temperatures. The set of STO spectra in (a) and (b) are identically drawn in different *y*-scale to match the STO signals in each type of sample. Note that STO peaks in the SL include that from the STO layers within the SL in addition to the substrate.