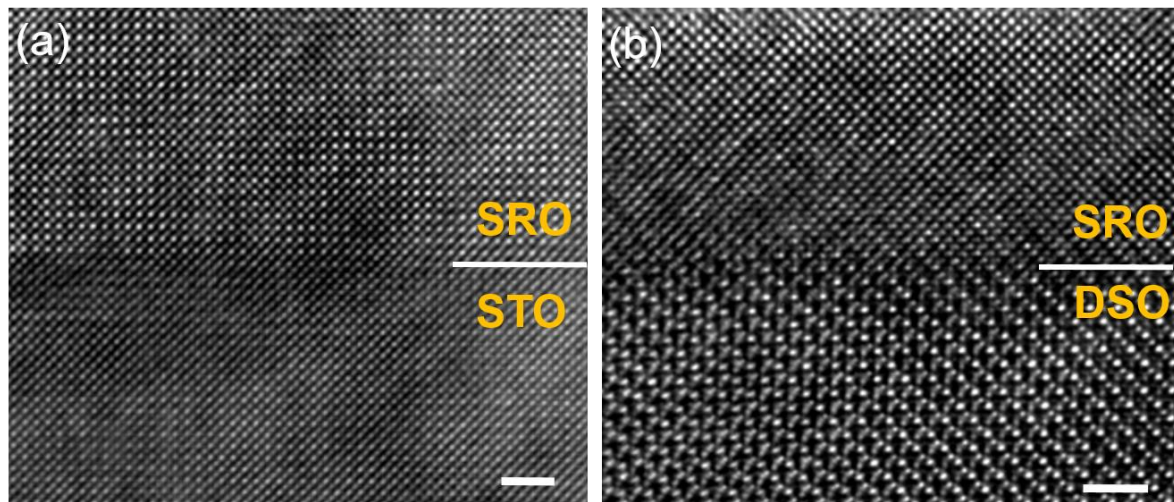
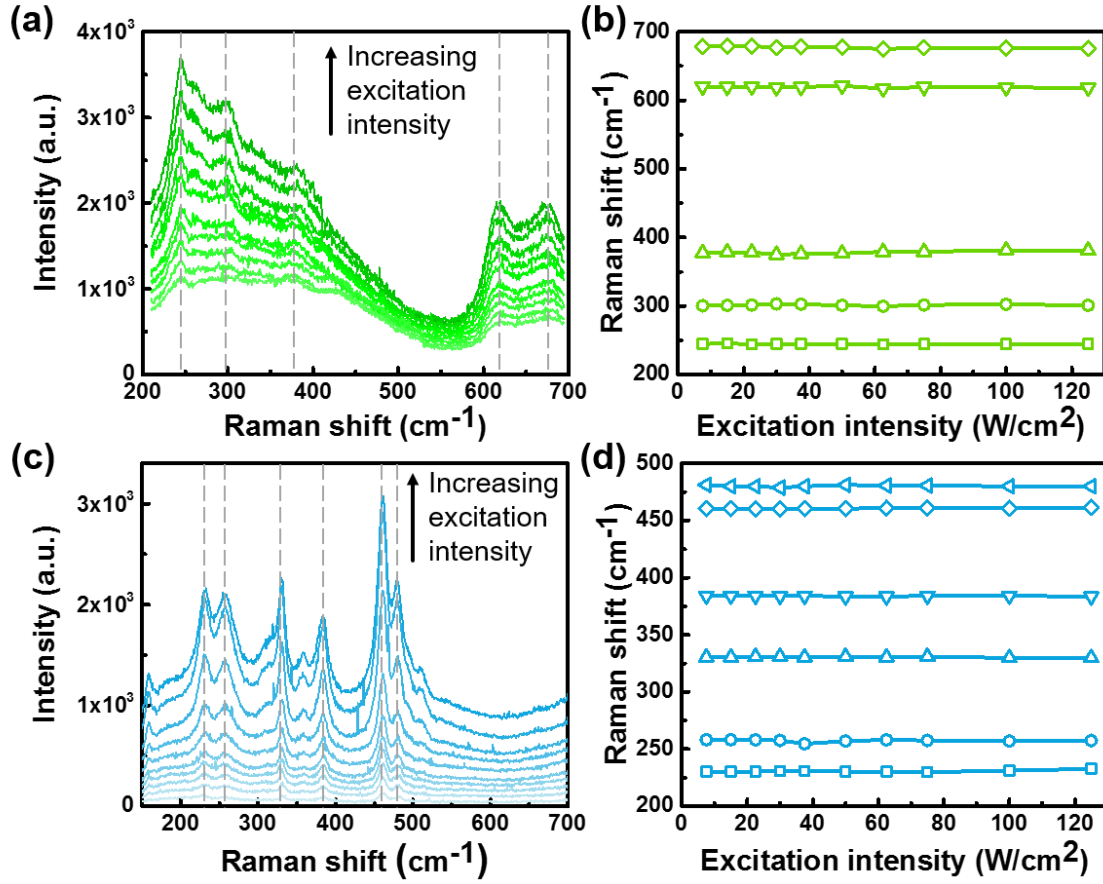
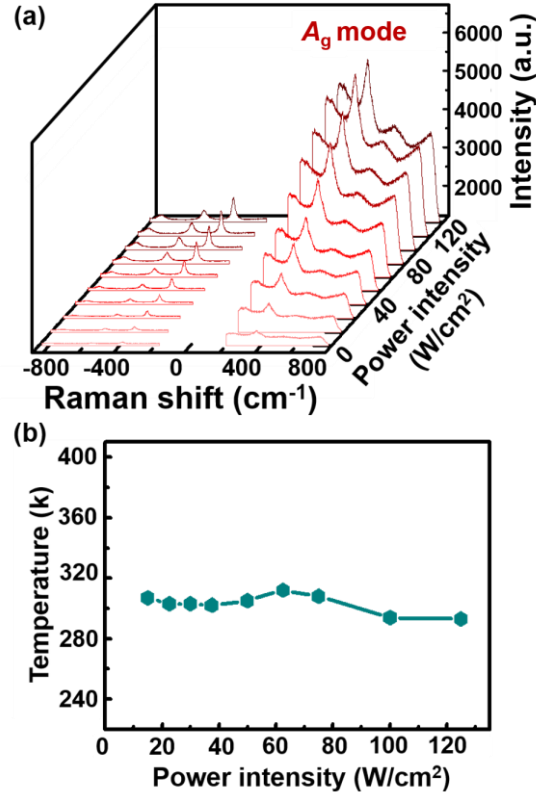




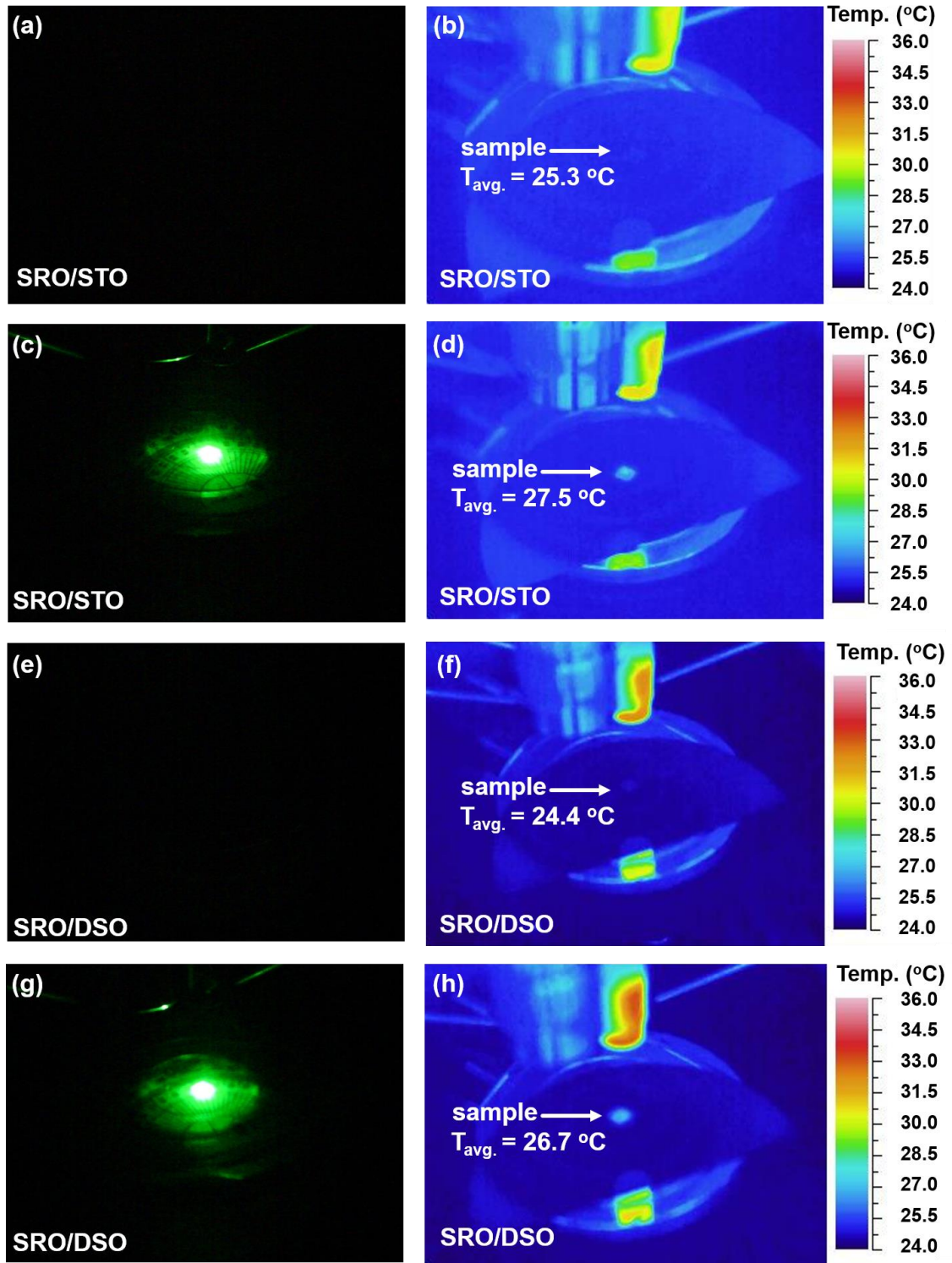
Supplementary Figure 1. Cross-sectional high-resolution transmission electron microscopy (HR-TEM) images. Using HR-TEM imaging, we were able to observe the apparent crystallinity of the **(a)** $\text{SrRuO}_3/\text{SrTiO}_3$ and **(b)** $\text{SrRuO}_3/\text{DyScO}_3$ cross-sectional interfaces (scale bars, 2 nm (**a** and **b**)). We emphasize the location of each interface with the added white line. These images reveal superior crystallinity of SrRuO_3 and its clear lattice structure. The $\text{SrRuO}_3/\text{SrTiO}_3$ and $\text{SrRuO}_3/\text{DyScO}_3$ materials feature sharp interfaces without observable interdiffusion of the species across these boundaries. Therefore, we can rule out the possibility that the crystal structure and crystal defects of the SrRuO_3 play a role in the observed photostriction.



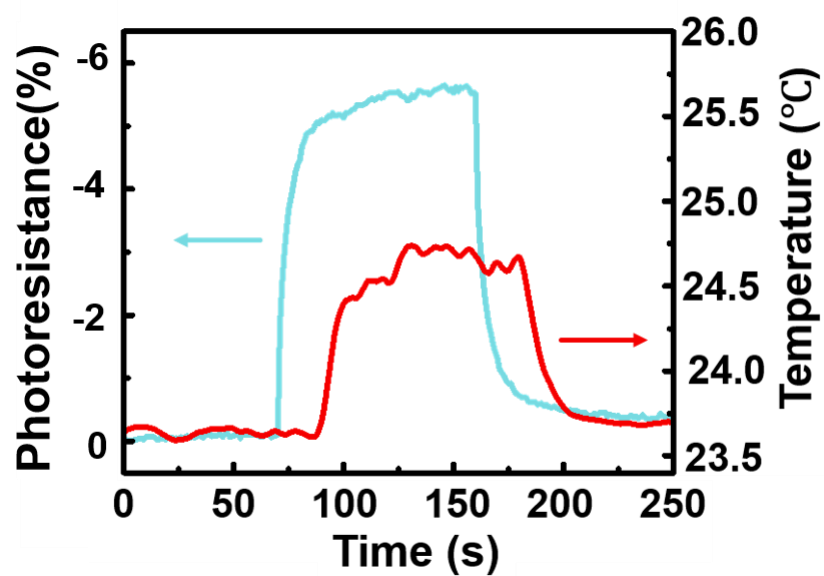
Supplementary Figure 2. Laser intensity-dependent Raman scattering spectra. The 532 nm laser intensity-dependent (a) Raman scattering spectra and (b) phonon modes of the SrTiO₃ substrate. The 532 nm laser intensity-dependent (c) Raman scattering spectra and (d) phonon modes of the DyScO₃ substrate. The vertical dashed lines indicate the positions of the phonon modes of the SrTiO₃ and DyScO₃ substrates.



Supplementary Figure 3. Laser intensity-dependent Stokes and anti-Stokes Raman spectra of SrRuO₃ thin films. (a) The Raman frequency of the A_g phonon mode are shown as a function of excitation power intensity. (b) The local temperature at the laser focal point obtained from the Stokes and anti-Stokes intensity ratio (I_S/I_{AS}) as a function of the laser intensity.



Supplementary Figure 4. Non-contact temperature measurements of the SrRuO₃. (a), (c) Optical and (b), (d) thermal images of SrRuO₃ on SrTiO₃ substrate before and during the laser irradiation, respectively. (e), (g) Optical and (f), (h) thermal images of the SrRuO₃ thin film on the DyScO₃ substrate before and during the laser irradiation, respectively.



Supplementary Figure 5. The time dependent photoresistance and temperature of the **SrRuO₃/DyScO₃**. The change in photoresistance (left y-axis) and temperature (right y-axis) as a function of time under 532 nm illumination at 50 W cm⁻².

Supplementary Note 1: Estimate the effective local temperature at the laser focal point.

Supplementary Figure 3a shows the laser intensity-dependent Stokes and anti-Stokes spectra of the SrRuO₃ thin films (the peaks of 372 and -372 cm⁻¹, respectively). The frequency shifts of the Stokes and anti-Stokes scattering were symmetric, but the scattering intensity of the anti-Stokes transition was much lower than the Stokes transition. According to the quantum theory, the local temperature, T_{eff} , can be extracted by¹:

$$T_{\text{eff}} = \frac{\frac{\hbar\omega}{k_B}}{\ln\left(\frac{I_S}{I_{AS}}\right)} = \frac{1.43859 \times \omega}{\ln\left(\frac{I_S}{I_{AS}}\right)} \quad (1)$$

where ω is the Raman frequency, $\hbar$ is the reduced Planck constant, k_B is the Boltzmann constant, and I_S and I_{AS} are the intensities of the Stokes and anti-Stokes Raman scattering, respectively. T_{eff} as a function of incident laser intensity is shown in Supplementary Figure 3b. There was no significant laser intensity dependence, demonstrating that there was no measurable laser-induced thermal expansion in the material. In addition, the photostriction of SRO depends nonlinearly on the light intensity and saturates beyond 50 W cm⁻², which is also inconsistent with the light-induced heating effect².

Supplementary Note 2: Measured the temperature of the SRO thin films.

To further exclude laser-induced thermal expansion, we measured the temperature of the SrRuO₃ thin films using an InfReC Thermo Gear G100EXD thermal camera (Nippon Avionics Co., Ltd.) under laser illumination for 3 min. The excitation intensity of the laser for the thermal camera measurements was the same as that for the Raman experiments in this study. As shown in Supplementary Figure 3a-d, the temperature in the lab was constant (25.3 °C), while we observed that the surface temperature of the SrRuO₃/SrTiO₃ sample increased by 2.2 °C (from 25.3 °C to 27.5 °C) under laser illumination. Supplementary Figure 3e-h shows the thermal image of the SrRuO₃/DyScO₃ sample. We observed a similar change in the surface temperature (2.3 °C increase) under laser illumination. This increase in temperature is too small to result in a large lattice strain as compared to photon-induced strain demonstrated in this study. Researchers have found that the volume change of SrRuO₃ is only about 0.02% from 200 K to 300 K³. However, we measured the photon-induced strain of SrRuO₃ to be as high as 1.12%.

Supplementary References

1. Balkanski, M., Wallis, R. F. & Haro, E. Anharmonic effects in light scattering due to optical phonons in silicon. *Phys. Rev. B* **28**, 1928–1934 (1983).
2. Daranciang, D. *et al.* Ultrafast photovoltaic response in ferroelectric nanolayers. *Phys. Rev. Lett.* **108**, 087601 (2012).
3. Lee, S. Large in-plane deformation of RuO₆ octahedron and ferromagnetism of bulk SrRuO₃. *J. Phys. Condens. Matter* **25**, 465201 (2013).