Electronic Supplementary Information for

## Photo-Assisted Epitaxial Growth from Nanoparticles to Enhance Multi-

## **Materialization for Advanced Surface Functionalization**

Masayuki Fukuda,1\* Yuuki Kitanaka,1 and Tomohiko Nakajima1

1. Advanced Manufacturing Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba Central 5, 1-1-1 Higashi, Tsukuba, Ibaraki 305-8565, Japan

Email:

\* m-fukuda@aist.go.jp

	<i>R</i> (at 248 nm)	lpha / cm <sup>-1</sup>	$\kappa / \mathrm{W} \mathrm{cm}^{-1} \mathrm{K}^{-1}$	$ ho$ / g cm $^{-3}$	$C / \mathrm{J} \; \mathrm{g}^{-1} \; \mathrm{K}^{-1}$
CaTiO <sub>3</sub> (CTO)			0.0421	4.10 <sup>2</sup>	0.721 <sup>3</sup>
BaTiO <sub>3</sub> (BTO)			$0.027^4$	6.06 <sup>2</sup>	0.4505
SrTiO <sub>3</sub> (STO)	0.3266	$700000^{6}$	0.11 <sup>7</sup>	5.17 <sup>2</sup>	0.5368
Film (a-1)	0.165	260000	$0.04^{a}$	4.63 <sup><i>a</i></sup>	0.625 <sup><i>a</i></sup>
Film (b-2)	0.104	280000	$0.04^{a}$	4.41 <sup><i>a</i></sup>	0.663 <sup><i>a</i></sup>

Table S1. The optical and thermal parameters for numerical simulations

<sup>*a*</sup>The parameters were determined using the values for CTO and BTO and their respective ratio in each film. The values in the table represent the following properties: *R* (reflectance),  $\alpha$  (optical absorption coefficient),  $\kappa$  (thermal conductivity),  $\rho$  (mass density), and *C* (specific heat capacity). They were substituted into the following heat diffusion equation, which is simplified into one-dimensional heat flow and includes *T* (temperature), *t* (time), *z* (depth), and *I*(*z*, *t*) (laser intensity),  $\rho C(\partial T/\partial t) = \kappa (\partial^2 T/\partial z^2) + \alpha I(z, t)$ .



**Fig. S1**. X-ray diffraction (XRD) pattern  $(2\theta/\omega)$  for film (a-1) with the intense STO peaks derived from the Cu K $\beta$  and W L $\alpha$  lines hindering the observation of the epitaxial growth of CTO from STO single-crystal (STO<sub>SC</sub>) substrate.



**Fig. S2**. Grazing incidence X-ray diffraction ( $\omega = 0.5^{\circ}$ ) patterns for films on glass coated with BTO nanoparticle (BTO<sub>NP</sub>) colloidal dispersion with (upper side) and without (lower side) 60 mJ cm<sup>-2</sup>–laser irradiation, indicating that laser irradiation does not change the peak intensity of BTO<sub>NP</sub>. The laser irradiation conditions for the two films were set following those for CTO-BTO films (a-1) and (b-1) in the main text: with a KrF pulse laser with a wavelength of 248 nm, pulse duration of 26 ns, and frequency of 50 Hz at 300 °C at 30 mJ cm<sup>-2</sup> for 500 pulses. Subsequently, only one of the film was irradiated with the laser at the same wavelength, pulse duration, frequency, and temperature conditions with fluence of 60 mJ cm<sup>-2</sup> for 500 pulses.



## Precursor film (b)



Fig. S3. Cross-sectional scanning electron microscopy images for precursor films (a) and (b).



**Fig. S4**. Energy-dispersive X-ray spectroscopy (EDS) mappings for the CTO-BTO films (a-1), (a-2), (b-1), and (b-2). The magenta, green, and blue colors represent Ca, Ba, and Sr atoms, respectively.



**Fig. S5**. Enlarged Cross-sectional transmission electron microscopy (X-TEM) image overlaid with the EDS mapping for  $BTO_{NP}$  in film (a-1), presented in Fig. 3. The interface between CTO and  $BTO_{NP}$  shows a continuous lattice image, highlighting the epitaxial growth of CTO from  $BTO_{NP}$ .



Fig. S6. X-TEM images of electron beam induced crystallization at the STO<sub>SC</sub> interface in the film (b-1).



**Fig. S7**. Enlarged view of X-TEM images for NPs from  $[100]_{pc}$  direction in the (b-2) film: (left) CTO<sub>NP</sub> derived from crystal nucleation growth with partial multi-domain, (right) BTO<sub>NP</sub> surrounded by CTOs with discontinuous lattice images, indicating no epitaxial growth.

## References

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