# Electronic Supplemental Information (ESI):
# Improvement of the hole mobility of SnO epitaxial films grown by pulsed laser deposition

Makoto Minohara[1]*, Naoto Kikuchi[1], Yoshiyuki Yoshida[1], Hiroshi Kumigashira[2,3], and Yoshihiro Aiura[1]

[1]*Electronics and Photonics Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki 305-8568, Japan*

[2]*Photon Factory, Institute of Materials Structure Science (IMSS), High Energy Accelerator Research Organization (KEK), Tsukuba, Ibaraki 305-0801, Japan*

[3]*Institute of Multidisciplinary Research for Advanced Materials (IMRAM), Tohoku University, Sendai 980-8577, Japan*

*Author to whom correspondence should be addressed; m-minohara@aist.go.jp

**This PDF file includes:**

Figures S1, S2, S3, S4 and S5

Reference S1

**Survey of the optimum growth conditions for SnO film grown on $LaAlO_3$ (001) substrate**

Figure S1 shows the growth condition dependence in XRD patterns of the grown films. At $T_g$ = 500 ºC, which is close to the conventional growth temperature of 575 ºC, lots of peaks can be observed, indicating there is no optimum condition for the growth of SnO single phase. The intensity of (00*l*) Bragg peaks of SnO become stronger by decreasing $T_g$ together with $P_{O2}$, however, other SnO Bragg peaks and other phases are still observed. Eventually, the SnO single phase can be obtained at $T_g$ = 350 ºC with $P_{O2}$ in the range between $2\times10^{-2}$ to $8\times10^{-2}$ Pa, while the $\beta$-Sn exists at $P_{O2} = 1\times10^{-2}$ Pa. Among them, the SnO film grown at $P_{O2} = 4\times10^{-2}$ Pa showed sufficiently low resistance at room temperature for reliable measurements using electric transport option with four-probe method attached on to the Physical Property Measurement System (Quantum Design, Dynacool). Therefore, we chose $T_g$ = 350 ºC and $P_{O2} = 4\times10^{-2}$ Pa as the optimum condition for single phase SnO epitaxial film.

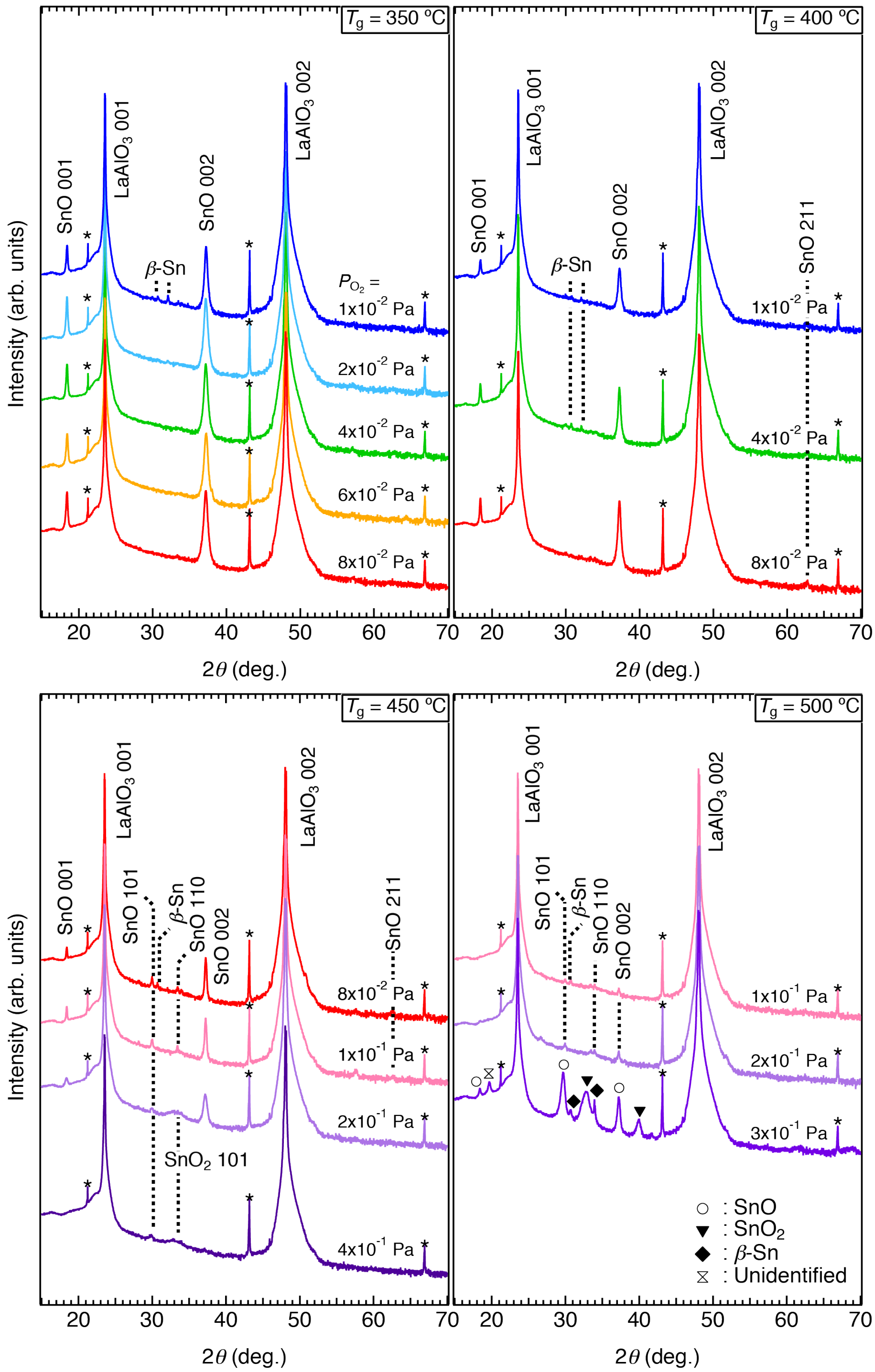


**Figure S1.** XRD patterns of the grown films on a $LaAlO_3$ (001) substrate at various $T_g$ and $P_{O2}$. The asterisks indicate the peaks originated by Cu Kβ radiation.

**Angular dependence of PES spectra for a Sn-3*d* core level**

Figure S2 shows angular dependent PES spectra from the SnO film for a Sn-3$d_{5/2}$ core level. Since the intensity of the peaks at higher (lower) binding energies becomes stronger (weaker) with increasing photoelectron emission angle $\theta$, the higher (lower) peaks are assigned to the surface (bulk) components.

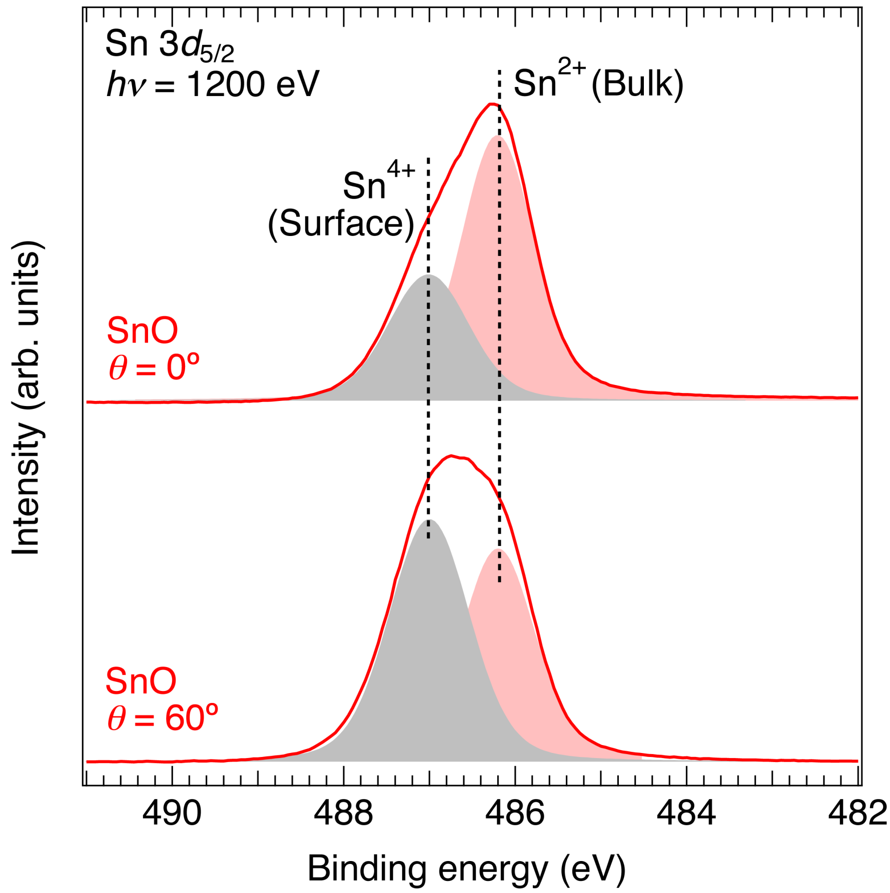


**Figure S2.** Angular (surface-sensitivity) dependent PES spectra from the SnO film for a Sn-3$d_{5/2}$ core level. The empirical spectrum can be fitted using two curves shaded with gray and red.

### Analysis of the angular dependence of PES spectra for a Sn-3*d* core level

In general, the total core-level photoemission signal $I$ can be written as the summation of a core-level spectrum emitted from each atomic layer:

$$I = \sum_{n=0}^{\infty} \exp\left(-nl/\lambda \cos\theta\right), \tag{1}$$

where $n$ is number of the $n$-th layer from the surface and $l$ is the distance between the neighboring atomic layers, the height of a single unit cell of assumed crystal structure. $\lambda$ is the characteristic probing depth based on the theoretical study of Tanuma *et al.*,[S1] and $\theta$ is the photoelectron emission angle with respect to the surface normal. In order to evaluate the thickness of the surface layer, the equation (1) is extended as

$$I_S/I_B = \left[a\left\{\sum_{n=0}^{d} \exp\left(\frac{-nl_S}{\lambda_S \cos\theta}\right)\right\}\right] / \left[(1-a)\left\{\sum_{n=d+1}^{\infty} \exp\left(\frac{-nl_B}{\lambda_B \cos\theta}\right)\right\}\right], \tag{2}$$

Subscript $S$ and $B$ means to surface and bulk layer, respectively. $a$ and $d$ correspond to the coverage and the thickness of the surface layer, respectively. By changing $a$ and $d$ as a free parameter for the simulation, we estimated at most 2-unit cells for the surface $Sn^{4+}$ region.

### The schematics for the characterization setup for the electrical measurement

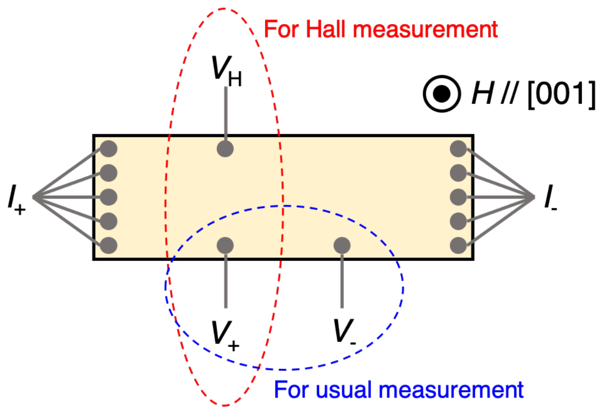


**Figure S3.** Schematic picture for the Hall measurements.

**XRD patterns of SnO film grown on YSZ (001) substrate for various growth temperature**

Figure S4a shows $2\theta$-$\theta$ XRD pattern of SnO film grown on YSZ (001) substrate. The lattice constant, which is estimated from (00*l*) Bragg peaks, as a function of the growth temperature is shown in Fig. S4b. The lattice constant is close to the bulk value at $T_g$ = 350 ºC. Above (below) this temperature, the lattice constant becomes smaller (bigger). This strongly suggests that the amount of defect (such as Sn or O vacancy) changes by changing the growth temperature.

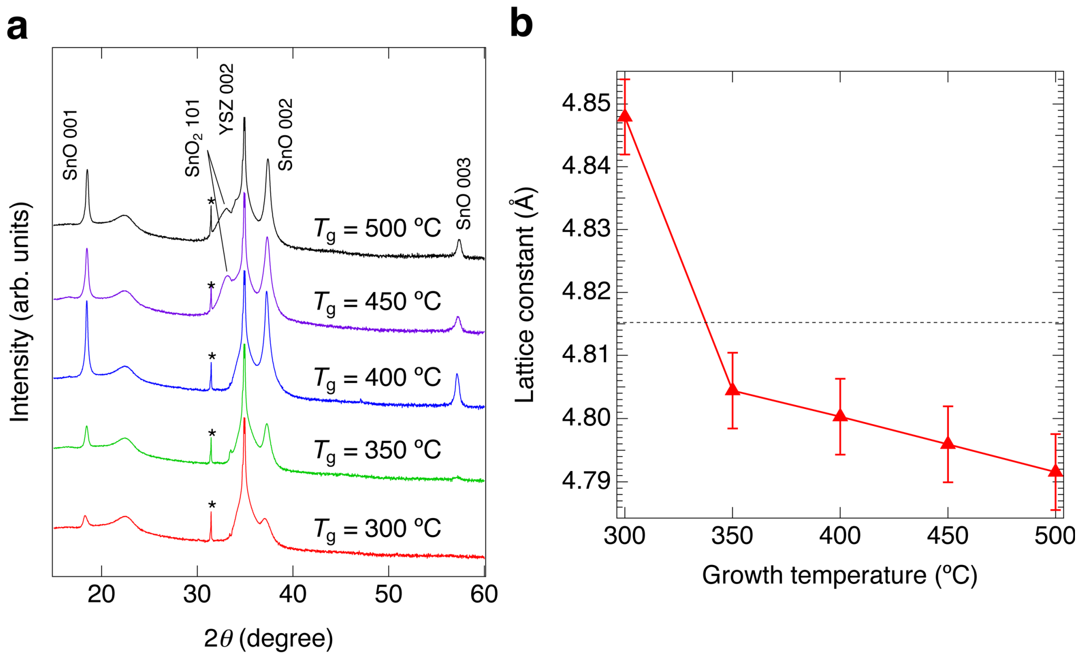


**Figure S4.** (a) XRD patterns of the grown films on a YSZ (001) substrate at various growth temperature $T_g$. The asterisks indicate the peaks originated by Cu Kβ radiation. (b) Estimated lattice constant from (00*l*) Bragg peaks in Fig. S4a. Broken line corresponds to the reported lattice constant of bulk SnO.

**The effect of growth temperature on the carrier density of SnO films**

Figure S5 shows the carrier density of single-phase SnO films which are grown on YSZ substrate as a function of the growth temperature (see also Fig. S4). The carrier density clearly decreases with decreasing the growth temperature. Therefore, we are convincing that the growth temperature plays an important role to suppress the carrier density. Considering the hole carrier density increases with increasing the growth temperature and the facts that the $SnO_2$ appears at a higher growth temperature range ($T_g \geq$ 450 ºC), we can deduce that Sn concentration becomes relatively poor (rich) with increasing (decreasing) the growth temperature.

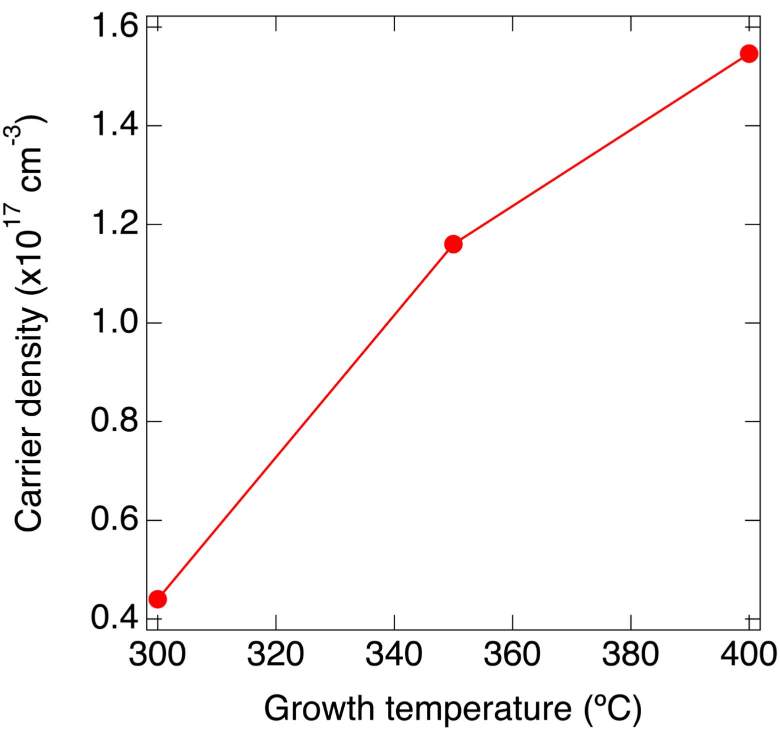


**Figure S5.** Carrier density of single-phase SnO films grown on YSZ substrate as a function of growth temperature.

**References**

R1 S. Tanuma, C. J. Powell, and D. R. Penn, *Surf. Interface Anal.*, 2003, **35**, 268.