Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information

Van der Waals interaction-induced photoluminescence weakening and multilayer growth in epitaxially aligned WS_2

Hyun Goo Ji,[a] Mina Maruyama,[b] Adha Sukma Aji,[a] Susumu Okada,[b] Kazunari Matsuda,[c] and Hiroki Ago[a,d,*]

[a] *Interdisciplinary Graduate School of Engineering Sciences, Kyushu University, Fukuoka 816-8580, Japan*
[b] *Graduate School of Pure and Applied Sciences, University of Tsukuba, Ibaraki 305-8571, Japan*
[c] *Institute of Advanced Energy, Kyoto University, Uji, Kyoto, 611-0011, Japan*
[d] *Global Innovation Center (GIC), Kyushu University, Fukuoka 816-8580, Japan*

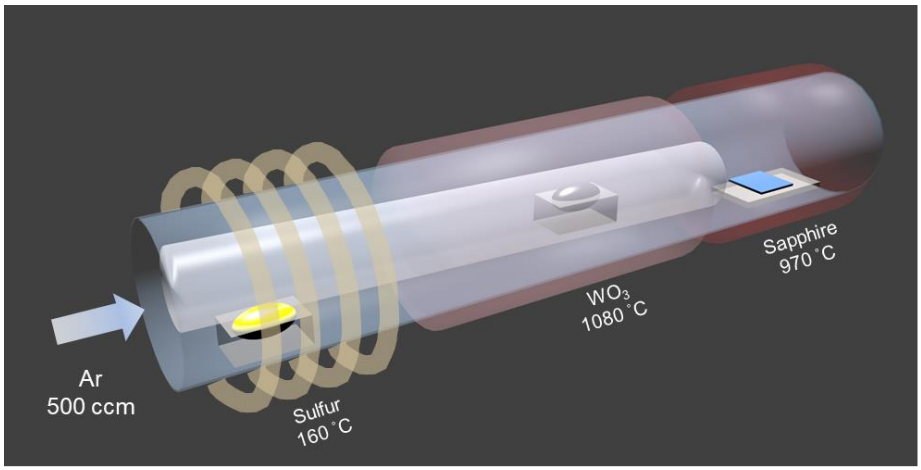

Figure S1. CVD condition used for WS_2 growth on sapphire.

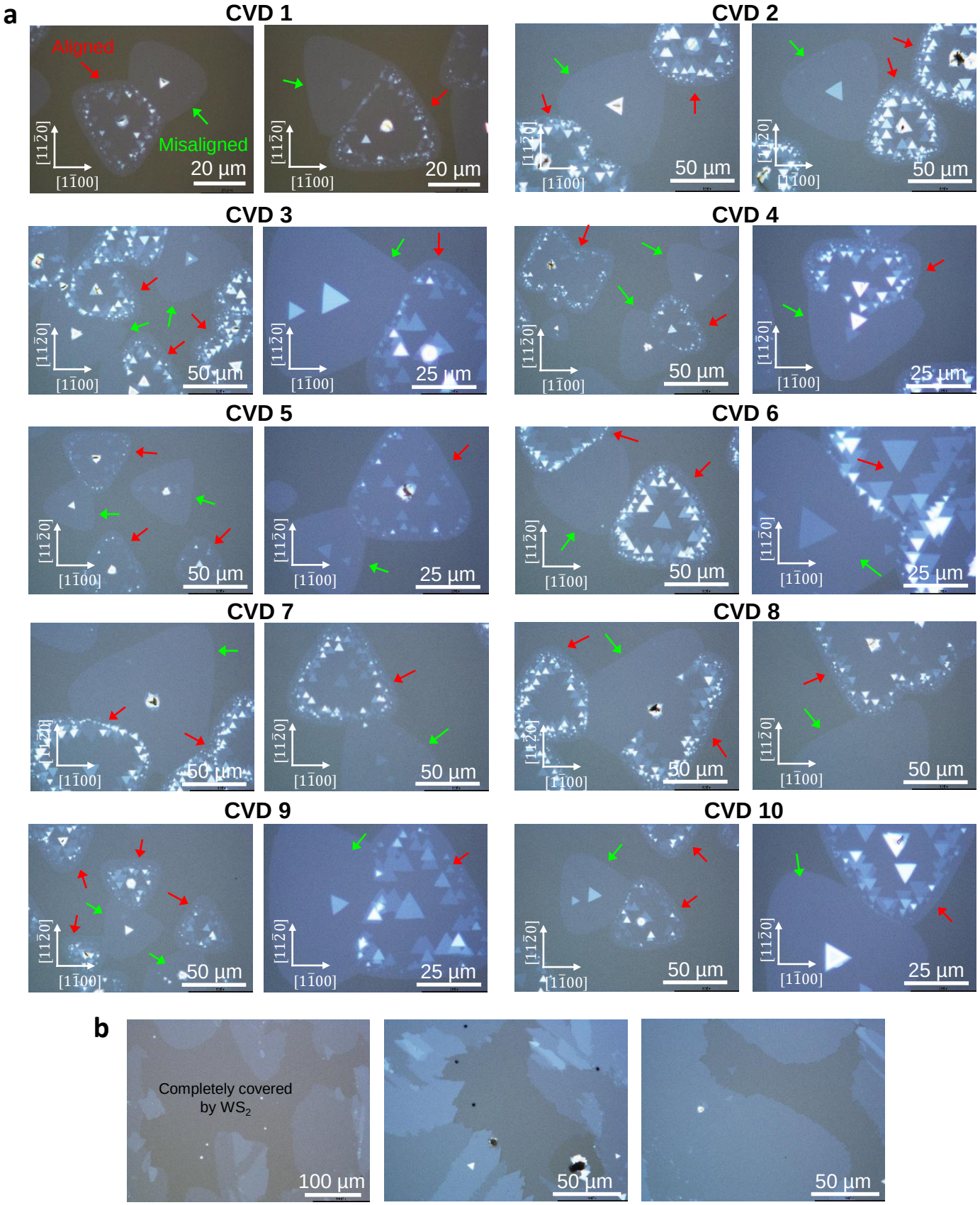

Figure S2. Optical micrographs of as-grown WS_2 grains on sapphire substrates. (a) The preferential overlayer deposition was observed on the aligned grains. (b) In a few cases, the sapphire surface was fully covered with WS_2 together with large multilayer grains.

Table S1. CVD conditions used for sample (A) ~ (G) shown in Figure S3 and the observed growth modes.

	Growth parameters				Growth mode	
	Gas flow rate	WO_3	S	sapphire	Epitaxially aligned grains	Preferential multilayer growth
A	300 ccm	1080 °C	160 °C	970 °C	X	X
B	500 ccm				O	O
C	700 ccm				O	O
D	500 ccm	1075 °C	160 °C	770 °C	X	X
E				870 °C	X	X
F				970 °C	O	X
G				1070 °C	O	O

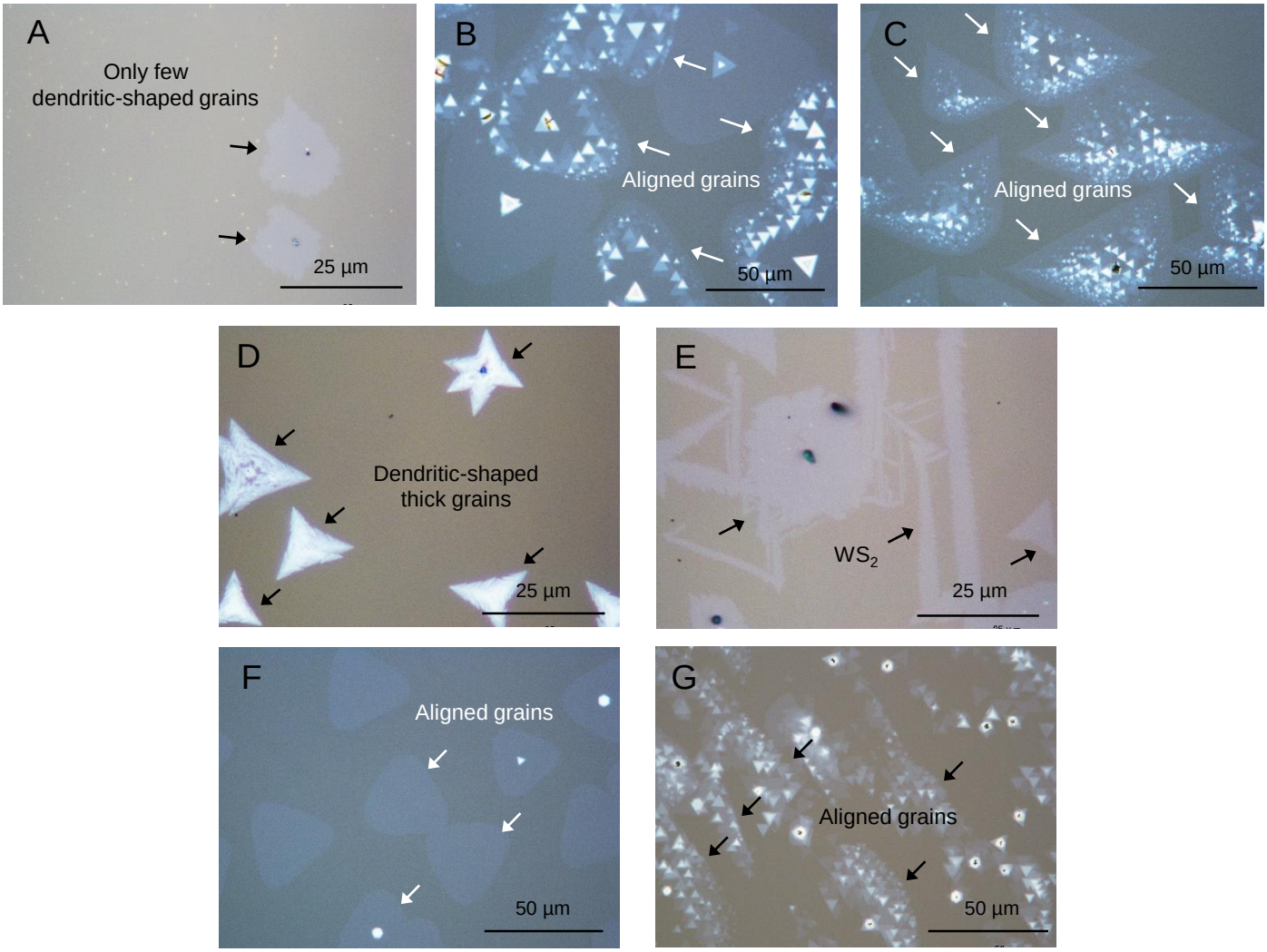

Figure S3. Optical images of as-grown WS_2 on c-plane sapphire synthesized using the growth conditions shown in Table S1.

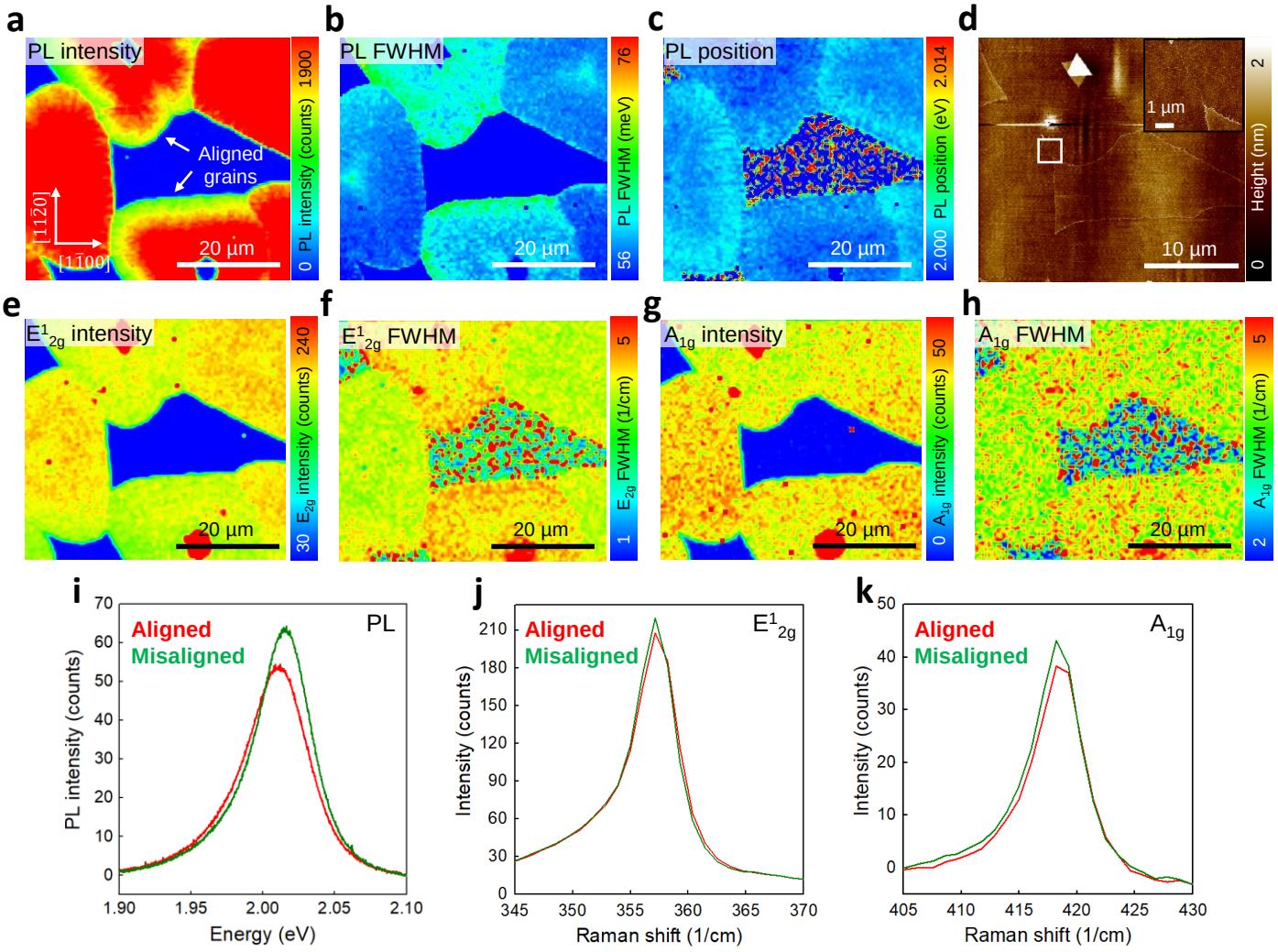

Figure S4. PL mapping images of the peak intensity (a), FWHM (b), and position (c) of monolayer WS_2 grains. (d) AFM height image. The inset shows a magnified image of the marked area. Raman E^1_{2g} mappings with intensity (e) and FWHM (f), and A_{1g} mappings with intensity (g) and FWHM (h). (i) PL, (j) E^1_{2g}, and (k) A_{1g} spectra taken from as-grown aligned and misaligned grains.

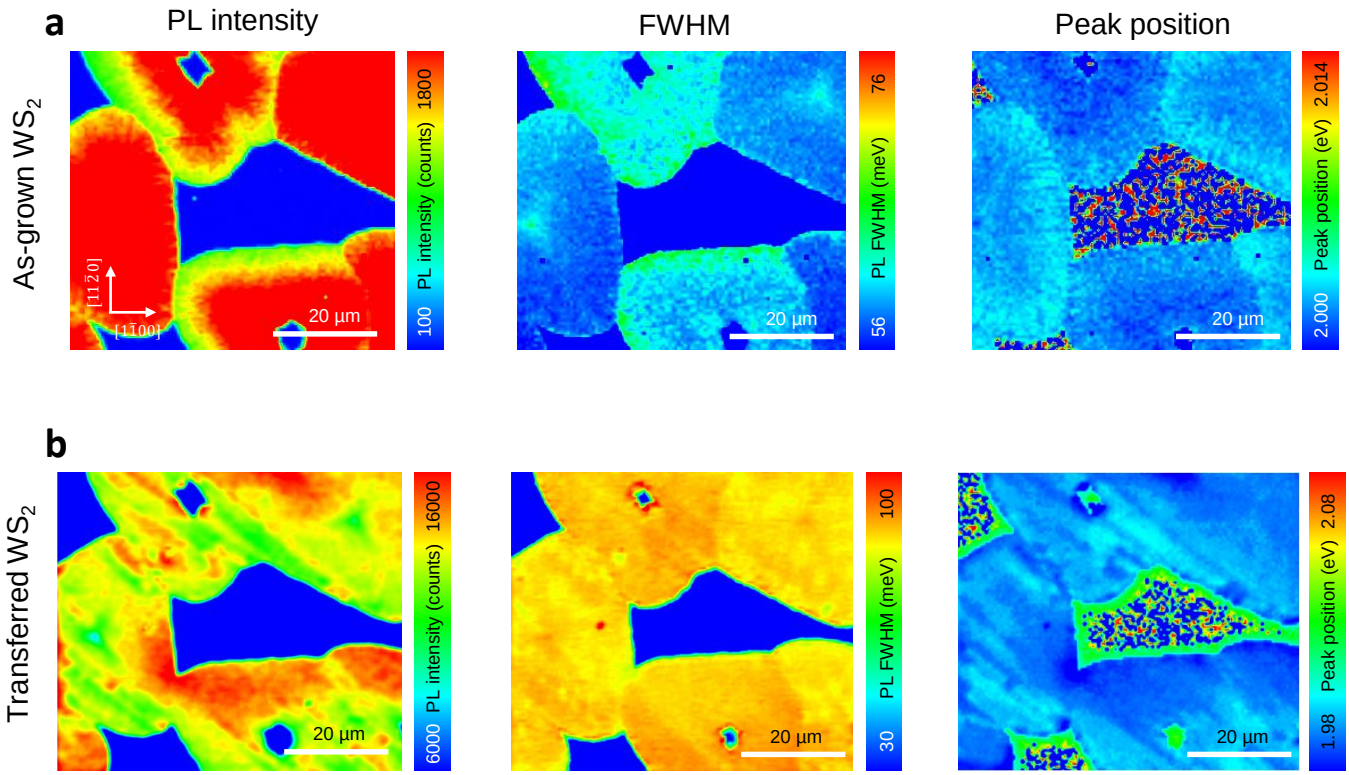

Figure S5. PL mapping images of identical WS_2 grains before (a) and after (b) transfer on SiO_2/Si substrate. The difference in PL intensity observed between aligned- and misaligned-grains in as-grown WS_2 disappeared after the transfer process. The wavy patterns observed in the PL intensity of the transferred WS_2 (b) are from wrinkles induced during transfer process.

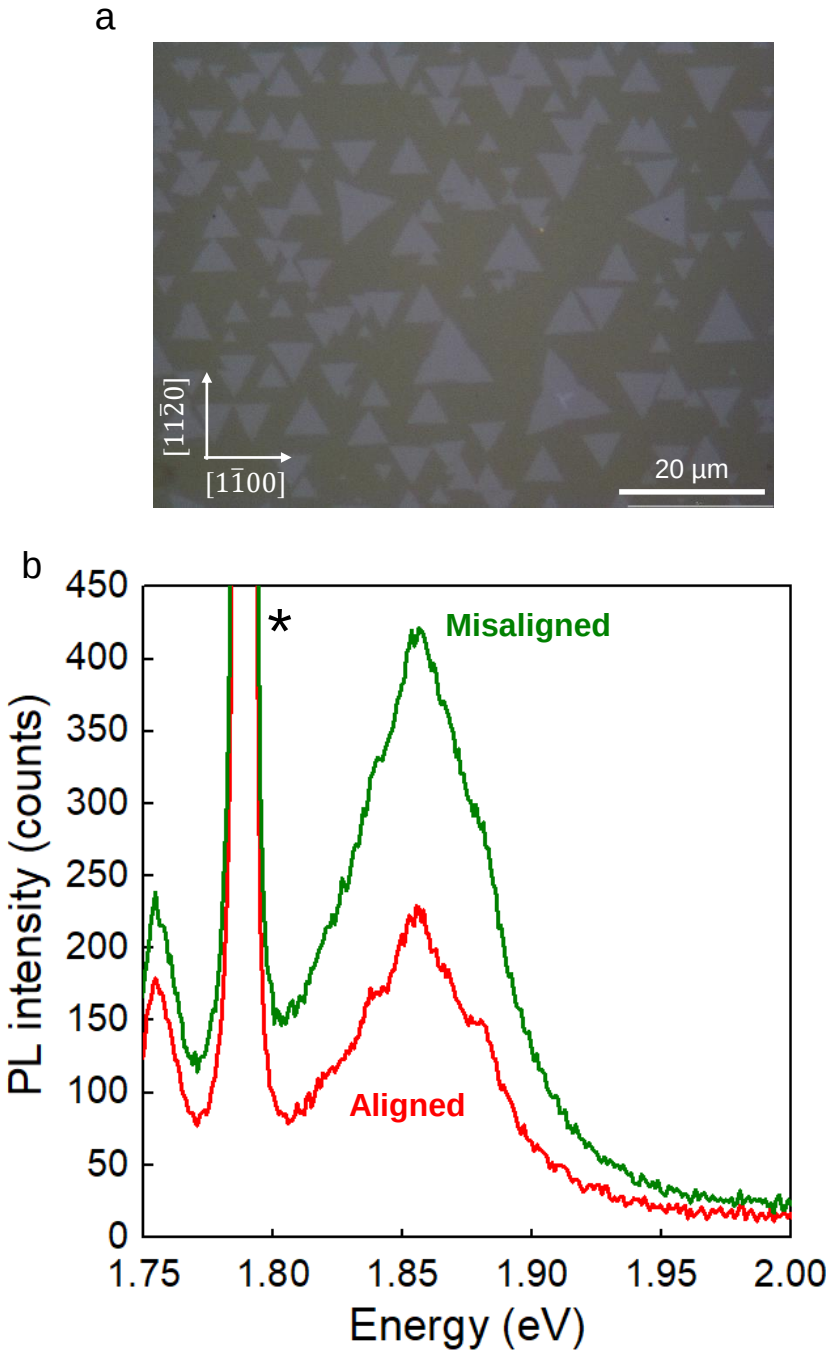

Figure S6. OM image and PL spectra of MoS_2 grown on c-plane sapphire. In the OM image (a), oriented grains are aligned along [1$\bar{1}$00] axis of sapphire substrate. PL spectra are shown in (b) where red and green spectra correspond to PL from aligned- and misaligned-grains, respectively. Asterisk (*) indicates a peak from a sapphire substrate. The peak at ~1.76 eV corresponds to bound exciton in MoS_2.

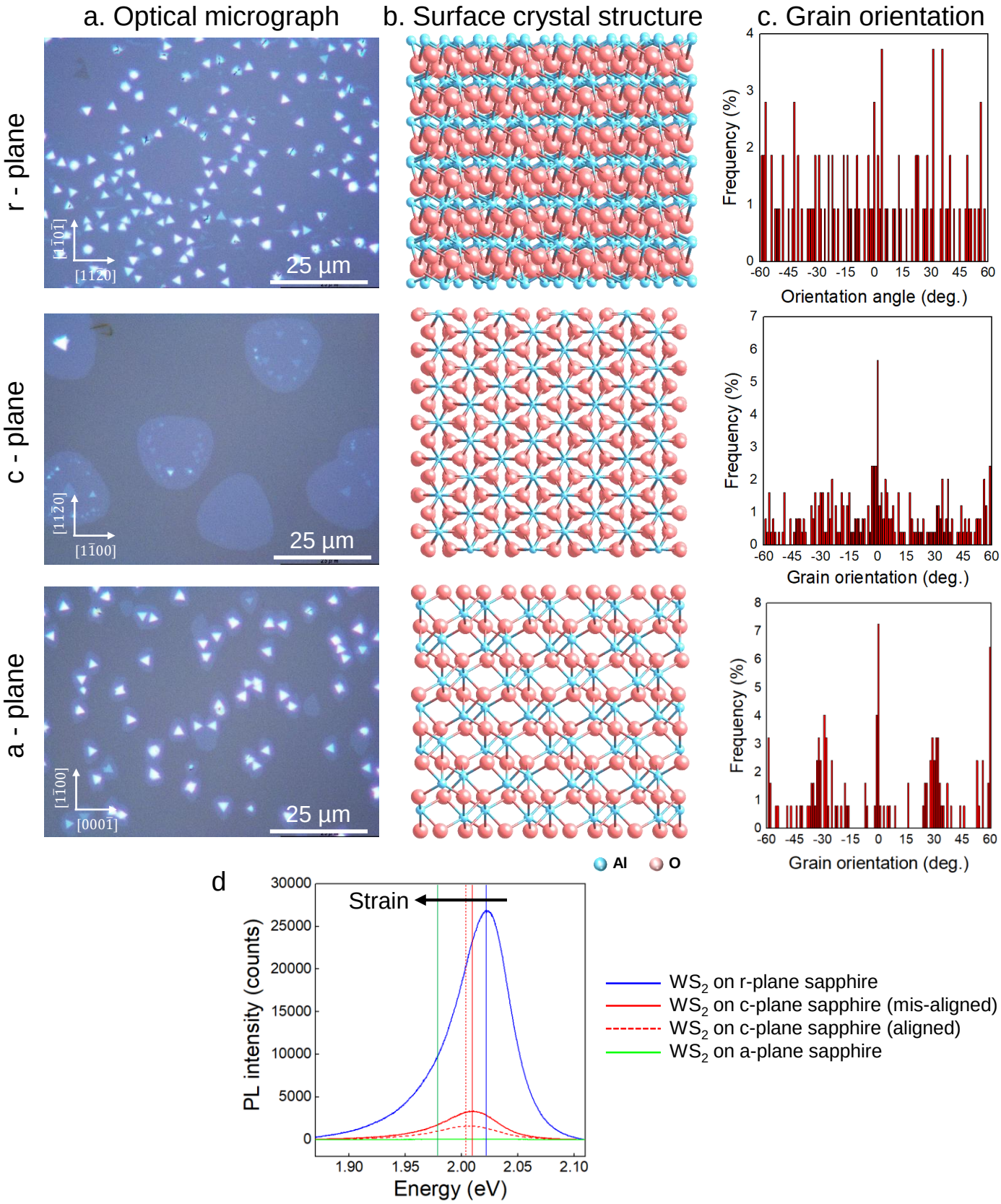

Figure S7. (a) OM images of CVD-grown WS_2 on different planes of sapphire substrates. (b) Surface crystal structure of each sapphire plane. (c) Grain orientation histogram of WS_2 grown on each sapphire plane. $[11\bar{2}0]$, $[1\bar{1}00]$, and $[000\bar{1}]$ axes are set as 0° for r-, c-, and a-plane sapphire, respectively. (d) PL spectra from the as-grown WS_2 grains.