Supporting Information

# A Facile Strategy for the Growth of High-Quality Tungsten Disulfide Crystals Mediated by Oxygen-Deficient Epitaxial Oxide Precursors

Denys I. Miakota<sup>1</sup>, Raymond R. Unocic<sup>2</sup>, Fabian Bertoldo<sup>3</sup>, Ganesh Ghimire<sup>1</sup>, Sara Engberg<sup>1</sup>, David Geohegan<sup>2</sup>, Kristian S. Thygesen<sup>3</sup>, and Stela Canulescu<sup>1</sup>

<sup>1</sup>Department of Photonics Engineering, Technical University of Denmark, DK-4000 Roskilde, Denmark

<sup>2</sup>Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge,

Tennessee 37831, United States;

<sup>3</sup>CAMD and Center for Nanostructured Graphene (CNG), Department of Physics, Technical University of Denmark, 2800 Kgs. Lyngby, Denmark

KEYWORDS: transition-metal dichalcogenides, WS2 monolayers

# X-ray photoelectron spectroscopy (XPS) of WOx films

**Table S1.** The XPS spectra fitting of the PLD-grown  $WO_{3-x}$  thin films.  $W4f_{7/2}$ ,  $W4f_{5/2}$ , and  $W5p_{3/2}$  peak parameters are presented for the corresponding doublets. BE denotes the binding energy and FWHM denotes the full width of half maximum.

XPS peak parameters		O2 background gas				Argon background gas			
		Film growth temperature				Film growth temperature			
Tungsten oxidation state	XPS peak	500° C	600º C	700º C	800° C	500° C	600º C	700º C	800° C
W <sup>6+</sup>	BE W4f7/2 (eV)	35.90	35.90	35.90	35.90	35.90	35.90	35.90	35.90
	BE W4f5/2 (eV)	38.05	38.03	38.02	38.05	37.97	38.00	37.97	38.04
	FWHM (eV)	1.33	1.55	1.53	1.54	1.55	1.71	1.76	1.78
	BE W5p3/2 (eV)	41.74	41.60	41.60	41.60	41.60	41.60	41.60	41.60
	FWHM (eV)	2.02	2.40	2.21	2.40	2.40	2.13	2.21	2.40
W <sup>5+</sup>	BE W4f7/2 (eV)	34.66	34.25	34.29	34.11	34.07	34.08	34.04	34.00
	BE W4f5/2 (eV)	36.73	36.32	36.36	36.32	36.25	36.28	36.11	36.17
	FWHM (eV)	1.74	1.39	1.25	1.30	1.44	1.07	1.12	1.18
	BE W5p3/2 (eV)	40.36	39.95	39.99	40.01	39.94	39.98	39.94	39.78
	FWHM (eV)	1.53	1.44	1.64	1.44	1.44	1.25	1.32	1.76
W <sup>4+</sup>	BE W4f7/2 (eV)	-	-	33.37	33.24	32.96	32.86	32.69	32.55
	BE W4f5/2 (eV)	-	-	35.44	35.31	35.03	34.97	34.79	34.67
	FWHM (eV)	-	-	1.25	1.59	1.48	1.79	1.75	1.66
	BE W5p3/2 (eV)	-	-	39.27	38.94	38.86	38.76	38.59	38.45
	FWHM (eV)	-	-	1.64	1.64	1.48	1.48	2.01	2.50

Table S1 shows the parameters extracted from the XPS peak fitting of the W 4f spectra presented in Figure 1a-h.

## XPS spectra of WS<sub>2</sub>

Table S2. The results of the W 4f and S 2p XPS spectra fitting of WS<sub>2</sub> films.

XPS peak parameters		O2 background gas				Argon background gas			
		Film growth temperature				Film growth temperature			
Oxidation state	XPS peak	500ºC	600ºC	700°C	800°C	500°C	600°C	700⁰C	800°C
W <sup>6+</sup>	BE W4f7/2 (eV)	-	-	35.90	35.90	35.90	35.90	35.90	35.90
	BE W4f5/2 (eV)	-	-	38.01	37.97	38.04	38.06	38.02	38.01
	FWHM (eV)	-	-	1.47	1.48	1.42	1.47	1.50	1.51
	BE W5p3/2 (eV)	-	-	41.60	41.60	41.60	41.00	41.60	41.60
	FWHM (eV)	-	-	2.14	1.92	2.40	2.40	2.40	1.92
$\mathbf{W}^{4+}$	BE W4f7/2 (eV)	-	-	32.75	32.97	32.62	32.59	32.56	32.67
	BE W4f5/2 (eV)	-	-	34.88	35.13	34.76	34.72	34.70	34.81
	FWHM (eV)	-	-	0.81	0.85	0.71	0.70	0.70	0.76
	BE W5p3/2 (eV)	-	-	38.65	38.67	38.52	38.49	38.46	38.57
	FWHM (eV)	-	-	1.99	2.02	2.09	2.40	1.83	2.07
W <sup>2x+</sup>	BE W4f7/2 (eV)	-	-	32.07	31.99	32.15	32.03	31.94	31.71
	BE W4f5/2 (eV)	-	-	34.14	34.36	34.22	34.10	34.01	33.78
	FWHM (eV)	-	-	1.25	1.25	1.64	1.64	1.44	1.48
	BE W5p3/2 (eV)	-	-	-	-	-	-	-	-
	FWHM (eV)	-	-	-	-	-	-	-	-
S <sup>2-</sup>	BE S 2p3/2 (eV)	-	-	162.39	162.59	162.31	162.26	162.27	162.34
	BE S2p1/2 (eV)	-	-	163.58	163.79	163.50	163.46	163.46	163.53
	FWHM (eV)	-	-	0.80	0.91	0.70	0.68	0.69	0.73
def S <sup>2-</sup>	BE S 2p3/2 (eV)	-	-	161.69	161.31	161.82	161.79	161.71	161.47
	BE S2p1/2 (eV)	-	-	162.96	162.41	163.12	163.09	163.01	162.77
	FWHM (eV)	-	-	1.15	1.44	1.16	1.30	1.42	1.15

Table S2 summarizes the fitting results of the XPS spectra shown in Figures 2 and S1. The XPS spectra of WS<sub>2</sub> synthesized from Ar-grown WO<sub>3-x</sub> films show narrow W 4f peaks (FWHM of W  $4f_{7/2} \sim 0.7$  eV, BE  $\sim 32.7$  eV) along with a stoichiometric 2H-WS<sub>2</sub> phase. Residual WO<sub>3</sub> found in the sulfurized films is evidenced by the presence of W<sup>6+</sup> doublet with W  $4f_{7/2}$  BE  $\sim 35.9$  eV. The

assignment of W components corresponding to WS<sub>2</sub> and WO<sub>3</sub> is straightforward and in good agreement with the literature<sup>1–3</sup>. We note that an additional low-intensity W<sup>2x+</sup> peak (where 0 < x < 2) doublet with BE ~31.9 eV must be taken into account to achieve the best fitting results of the W 4f spectra. These components may be attributed to the sulfur-deficient WS<sub>2</sub> phase (denoted as WS<sub>x</sub>), which may originate from grain boundaries, as well as various intrinsic point defects<sup>4</sup>. The broad contributions at BE of ~38.8 eV and ~41.6 eV (with a color corresponding to a doublet) are due to W 5p<sub>3/2</sub> satellite peaks. W5p<sub>3/2</sub> contribution originating from W<sup>2x+</sup> was intentionally neglected due to extremely low XPS signal contribution (Figure 2a-d) <sup>1–3</sup>.



Figure S1. S 2p core-level XPS spectra of WS<sub>2</sub> monolayers obtained upon sulfurization at 900 °C of tungsten oxide (40 shots) thin-film precursor grown in Ar at a) 500 °C, b) 600 °C, c) 700 °C, d) 800 °C. The XPS spectra peaks were attributed to sulfur valence states S<sup>2-</sup> in WS<sub>2</sub> (magenta lines) and S<sup>2-</sup> in WS<sub>x</sub> (orange lines).

Figure S1 shows the S 2p XPS spectra of WS<sub>2</sub> films obtained by WO<sub>3-x</sub> thin films synthesized in Ar and sulfurized at 900°C.



**Figure S2**. S 2p core-level XPS spectra of WS<sub>2</sub> were obtained upon sulfurization at 900 °C of WO<sub>3-x</sub> thin-film precursor grown in O<sub>2</sub> at a) 700 °C, b) 800 °C. The XPS spectra peaks were attributed to sulfur valence states S<sup>2-</sup> in WS<sub>2</sub> (magenta lines), S<sup>2-</sup> in WS<sub>x</sub> (orange lines), and S<sup>2-</sup> in WO<sub>x</sub>S<sub>y</sub> (green lines). All films were grown using 40 laser pulses.

Figure S2 shows the S 2p XPS spectra of WS<sub>2</sub> films obtained from WO<sub>3-x</sub> thin films synthesized in O<sub>2</sub> and sulfurized at 900°C. A low fraction of residual WO<sub>x</sub>S<sub>y</sub> is evidenced by the presence of S-O bonds in the S 2p XPS spectra and the presence of the peaks in W 4f spectra at higher BE (with respect to the WS<sub>2</sub> phase). The BE of the S 2p3/2 peak is ~162.5 eV for WS<sub>2</sub> and ~161.7 eV for defective WS<sub>2</sub>. Similar values were obtained for WS<sub>2</sub> made from Ar-grown precursors (not shown).



**Figure S3**. W 4f core-level XPS spectra of WS<sub>2</sub> films obtained upon sulfurization at 900 °C of WO<sub>3-x</sub> precursors grown in O<sub>2</sub> at a) 700 °C, b) 800 °C. The XPS spectra peaks attributed to three sets of major doublets corresponding to W<sup>6+</sup> (blue lines), W<sup>4+</sup> (magenta lines), and W<sup>x+</sup> (orange lines) valence states, and a corresponding satellite W5p<sub>3/2</sub> peak (with the same color as the main doublet). In b) W 4f an additional doublet corresponding to W chemical state in WO<sub>x</sub>S<sub>y</sub> is also present (light green lines).

Figure S3 shows the W 4f XPS spectra of WS<sub>2</sub> films obtained from the WO<sub>3-x</sub> thin films grown in O<sub>2</sub> and sulfurized at 900°C. The XPS peaks are assigned to tungsten in distinct chemical states, i.e., W<sup>4+</sup> valence state with W4f<sub>7/2</sub> BE of 32.8 eV in hexagonal WS<sub>2</sub>, W<sup>6+</sup> state with W 4f7/2 BE of 35.90 eV for fully coordinated W in WO<sub>3</sub>, and W<sup>4+</sup> state with W4f7/2 BE of 32.0 eV in defective WS<sub>2</sub> (WS<sub>x</sub>). Additionally, W state with W4f<sub>7/2</sub> BE of 33.4 eV evidence defective residual WO<sub>x</sub>S<sub>y</sub>, oxysulfide where S-O bonds are present. In this case, the assignment is made considering that: 1) the position of the W<sup>4+</sup> peaks is shifted to lower binding energy by only 0.7 eV from the WS<sub>2</sub> peaks and not 1 eV, as expected for the 1T-WS<sub>2</sub>; 2) the stoichiometric ratio between the S 2p def and the W 4f def components is ~1 and not 2, as expected for both the 1T-WS<sub>2</sub> and vacancies <sup>1,5,6</sup>. The sulfurization reaction appears to be very different from the films deposited in Ar. Indeed, a much larger fraction of WO<sub>3</sub> is observed in WS<sub>2</sub> sulfurized from precursors grown in O2 as compared to Ar. This is due to the presence of lower oxidation states in the precursor film.



**Figure S4**. (left)  $\theta - 2\theta$  XRD patterns of WO<sub>x</sub> films on (0001) Al<sub>2</sub>O<sub>3</sub> substrates. The sapphire peaks are marked with asterisks. The films were grown using 1500 laser pulses and substrate temperatures of 500 °C (black curves), 600 °C (blue curves), 700 °C (green curves), and 800 °C (red curves).

A higher crystallinity of the WO<sub>x</sub> films is observed with increasing substrate temperature. Films grown at 500°C have a very broad diffraction peak which stems from a poor crystallinity. At 600 °C and 700 °C, the XRD patterns show peaks at 20 of 35.78° and 36.58°, respectively which are assigned to the (020) planes of monoclinic WO<sub>2</sub>. Films grown at 800 °C in Ar exhibit a significant peak shift to 37.42°, which may be assigned to a change in the film orientation to (002) WO<sub>2</sub>. Lastly, the XRD peaks at 20.58°, 41.70°, 64.56°, 86.08°, and 90.82° were assigned to the (0003), (0006), (0009), K<sub>β</sub> ,(00012), and (00012) reflections of the Al2O3 (0001) substrate.



**Figure S5**. High-resolution SEM images of WS<sub>2</sub> were obtained upon sulfurization of WO<sub>3-x</sub> films grown in Ar at a) 500 °C; b) 600 °C; c) 700 °C; d) 800 °C. All images were recorded on a Zeiss Merlin microscope using an In-Lens detector, an acceleration voltage of 1 to 1.5 kV, and a short working distance (3 mm or less).

# **Optical images**



**Figure S6**. **(a-c)** Optical images of WS<sub>2</sub> films obtained by sulfurization of the WO<sub>x</sub> thin films grown at 800 °C in Ar. The oxide films were grown using 40 laser pulses. The optical images were taken from different regions of the films. (d) SEM images reveal single- and multi-layer regions with dominant AA (A...) crystal orientation. **(f, g)** Comparison between the PL spectra of PLD-WS<sub>2</sub> and CVD-WS<sub>2</sub> (purchased from 2dsemiconductors). Note that the PL emission of the PLD-WS<sub>2</sub> most likely originates from 1 to 3 layers of WS<sub>2</sub>.

#### **Raman and PL studies**

WS<sub>2</sub> optical characterization was performed by photoluminescence (PL) and Raman spectroscopy. Raman spectroscopy is an important tool to determine the number of layers and their stacking sequence<sup>7–9</sup>, inter-layer coupling<sup>9</sup>, the external field, and molecular doping effects, defects, or disorder in 2D materials<sup>8,10</sup> and strain<sup>7,11</sup>. The variations of Raman peaks position, width, and intensities contain information regarding the electron-electron, electron-phonon interactions <sup>7,8,12</sup> or exciton-phonon coupling <sup>8,13</sup>.

Hexagonal WS<sub>2</sub> bulk crystal belongs to P63/mmc ( $D_{6h}$ <sup>14</sup>) point group and the group theory suggests the following vibrational modes at the center of the Brillouin zone ( $\Gamma$  point):

$$\Gamma = A_{1g} \oplus 2A_{2u} \oplus B_{1u} \oplus 2B_{2g} \oplus E_{1g} \oplus 2E_{1u} \oplus E_{2u} \oplus 2E_{2g} \quad (1)$$

Among these, there are four Raman-active modes, i.e.,  $A_{1g}(\Gamma)$ ,  $E_{1g}(\Gamma)$ ,  $E_{2g}(\Gamma)$ , and  $E^{2}_{2g}(\Gamma)$ . However, due to the lack of translational symmetry along the *z*-axis, there is a reduction in symmetry in few-layers WS<sub>2</sub> (FL WS<sub>2</sub>), Moreover, in the WS<sub>2</sub> monolayer (1L), the E<sup>2</sup><sub>2g</sub> mode is absent due to the lack of inversion symmetry along the z-axis, which is perpendicular to the basal XY-plane. Odd few-layers WS<sub>2</sub> crystal belongs to D<sub>3h</sub> point group <sup>15</sup> symmetry because of the presence of the horizontal reflection plane that passes through the transition metal atom and exhibits 9 phonon branches. In particular, the unit cell of 1L-WS<sub>2</sub> is composed of three atoms with 9 normal vibrational modes at  $\Gamma$  point, which is expressed based on D<sub>3h</sub> as <sup>15</sup>  $\Gamma$  = 2A<sup>"</sup><sub>2</sub> +A<sup>'</sup><sub>1</sub>+2E<sup>'</sup>+E<sup>"</sup>, where one  $A''_2$  and one E' are acoustic modes, another  $A''_2$  is IR active,  $A'_1$  and E'' are Raman active, and another E' is both Raman and IR active and three Brillouin zone-center ( $\mathbf{q} \approx \mathbf{0}$ ) modes A'<sub>1</sub>, E' and E" are Raman-active. However, in the conventional back-scattering geometry where the incident light direction is perpendicular to XY-plane, only two A'<sub>1</sub> and E' Raman active first-order bands are observed. For a perfect crystalline material, the vibrational normal modes have an infinite spatial correlation. Therefore, Raman active phonons can be described as plane waves with a finite wave vector  $\mathbf{q} \approx \mathbf{0}$ . The fundamental Raman selection rule for the phonons ( $\mathbf{q} \approx \mathbf{0}$ ) is relaxed for a finite crystalline domain size  $d (\Delta q \approx 1/d)$  or if defects are introduced, that perturbs the spatial translational invariance of the system. As a consequence, the phonon correlation length L<sub>c</sub> becomes finite, and the fundamental selection rule for the Raman process is relaxed <sup>16</sup>. In particular, the unit cell of hexagonal 1L-WS<sub>2</sub> has three Brillouin zone-center ( $\mathbf{q} \approx \mathbf{0}$ ) Raman-active modes  $A'_1, E'$ , and E". In the conventional back-scattering geometry only two A'<sub>1</sub> and E' Raman-active first-order

bands are observed. When the crystalline domain size is finite or if defects are introduced, the fundamental Raman selection rule for the phonons ( $q \approx 0$ ) is relaxed. Since the momentum conservation condition can be satisfied by phonon scattering from a defect, this enables the observation of several first-order zone-edge (M point) phonons away from the  $\Gamma$  point for samples with high densities of defects <sup>7</sup>. A zone-edge mode, which is identified as the longitudinal acoustic mode at M point, LA(M), is also known to be Raman active in the WS2<sup>8,13</sup>. The symbol (M) refers to the M-point of the Brillouin zone with the specific momentum q of the phonon. Along with the first-order Raman modes at  $\Gamma$  point, several features have been identified in the WS<sub>2</sub> spectrum and they are associated with second-order processes, such as multi-phonon combinations or overtones of phonons with finite wavevectors ( $q \neq 0$ ). They are usually located at the corners and edges of the Brillouin zone<sup>8,13</sup>. The PL and Raman spectra for the WS<sub>2</sub> monolayers sulfurized using Argrown WO<sub>x</sub> precursor are presented in Figure 3 in t Each spectrum was deconvoluted with different phonon modes, as presented in Figure 3 (b,d,f,h). The Raman peak at ~177 cm<sup>-1</sup> is assigned to LA(M), a first-order Raman-active mode related to the longitudinal acoustic (LA) phonons at the edge of the Brillouin zone (the M point)<sup>7</sup>. The prominent peak at  $\sim$ 353 cm<sup>-1</sup> is discussed in the main text. Although the second-order longitudinal acoustic mode at the M point 2LA(M) strongly attenuates the first-order  $E^{1}_{2g}(\Gamma)$  mode and overlaps with the first-order  $E^{1}_{2g}(M)$ mode, multi-peak Lorentzian fitting correlates their contributions. Second-order Raman contributions can be observed, and the most prominent one is ascribed to the 2LA(M) band, originating from a second-order double resonant process involving two longitudinal acoustic (LA) phonons close to the M point of the Brillouin zone. The intensity of the 2LA(M) mode can be strongly enhanced with decreasing number of WS<sub>2</sub> monolayers <sup>7</sup>, if the energy of the incident laser photon is in resonance with X<sub>A</sub>, X<sub>B</sub>, and X<sub>C</sub> excitations states with the reported energies of ca. 2.0 eV, 2.4 eV and 2.8 eV, respectively <sup>8,17</sup>. In this work, the excitation at 532 nm is close to the B exciton, resulting in an enhanced 2LA(M) Raman mode and this makes the second-order phonon mode 2LA(M) much more pronounced than the in-plane phonon mode  $E^{1}_{2g}(\Gamma)$ . The other Raman features in the range of 300 to 350 cm<sup>-1</sup> can be tentatively assigned to three peaks with the following peak centers:  $2LA(M)-2E^{2}_{2g}(\Gamma)$  at ~299.5 cm<sup>-1</sup>, LA(M)+ TA(M) at ~314.2 cm<sup>-1</sup> and  $2LA(M) - E^{2}_{2g}(\Gamma)$  at ~326.8 cm<sup>-1</sup> following the reports of Berkdemir et al.<sup>7</sup> and E.del Corro et.al.<sup>8</sup>. Raman bands at ~419.3 cm<sup>-1</sup> indicate the  $A_{1g}(\Gamma)$  out-of-plane mode, which is known to have a strong electron-phonon coupling and is also sensitive to electron doping <sup>18</sup>.

## Formation Energies of Oxygen Vacancies in non-stoichiometric tungsten oxides



**Figure S7.** (a) DFT-calculated formation energy ( $E^F$ ) of oxygen vacancies in different supercells for different  $WO_{3-x}$  oxides (0<x<1) as a function of the oxygen chemical potential ( $\mu_O$ ). The left side of the x-axis corresponds to the oxygen-poor limit whereas the right side corresponds to the oxygen-rich limit. (b) The formation energies of oxygen vacancies for various WOx as a function of oxygen vacancy content.

Figure S7 visualizes the formation energies of oxygen vacancies for different oxides of  $WO_{3-x}$  (0<x<1) as a function of the oxygen chemical potential. The DFT calculations reveal that the formation energy is the lowest for oxides close to stoichiometric  $WO_3$  (E<sup>F</sup> of  $WO_{2.944}$  is 1.055 eV for O-poor conditions). One can also observe that with decreasing concentration of oxygen vacancies, i.e. higher x values on the X-axis of Figure S7b, the formation energy decreases. This

can be rationalized by the fact that a lower vacancy concentration corresponds to a smaller disturbance of the pristine material. As expected, the formation energy increases with increasing the oxygen chemical potential of oxygen.



Figure S8. Equilibrium oxygen vacancy concentration as a function of temperature.

Figure S8 shows the equilibrium concentration of oxygen vacancies as a function of temperature. As expected, the concentration increases with the increasing temperature to values up to  $10^{19}$  cm<sup>-3</sup>. The concentrations were calculated according to a Boltzmann distribution:



**Figure S9.** Visualization of sulfur interstitial ( $S_i$ ) and sulfur on oxygen site ( $S_o$ ) in WO<sub>3</sub> (both shown by the yellow spheres). The grey spheres denote the W atoms and the red circles the O atoms. Table showing the formation energy (eV) for  $S_i$  and  $S_o$ .

#### Slab Calculations for WO3 and the Sulphurisation Process

In addition to the DFT calculations on bulk, we have also conducted slab calculations for  $WO_3$ . For this purpose, we set up a pristine (001)- $WO_3$  surface using a slab model (Figure S11, left), with an oxygen vacancy,  $V_0$  (Figure S11, middle), and sulfur on the oxygen site,  $S_0$  (Figure S11, right). We calculate the adsorption energy for a sulfur atom on top of a surface with an existing oxygen vacancy in the following way:

$$E_{ads} = E(slab with S_0) - (E(slab with V_0) - E(S))$$

Our calculations suggest that the incorporation of S on the WO<sub>3</sub> surface with an oxygen vacancy present yields adsorption energy of -1.256 eV. It is therefore energetically favorable to incorporate sulfur into the structure when oxygen vacancies are present.



**Figure S10**: Visualization of systems in the slab calculations. Left: pristine WO<sub>3</sub> slab along the (001) surface. Middle: Oxygen vacancy ( $V_0$ ) within the slab. Right: sulfur on oxygen site ( $S_0$ ).



**Figure S11**. Intrinsic point defects of monolayer  $WS_2$ . Atomic resolution images show point defects in monolayer  $WS_2$ , namely single W vacancy ( $V_W$ ) and double W vacancy ( $V_{2W}$ ). The upper and lower rows are the original and processed images, respectively.

STEM images in Figure S11 show intrinsic point defects in monolayer  $WS_2$ , namely single W vacancy (V<sub>W</sub>) and double W vacancy (V<sub>2W</sub>).<sup>19</sup>

All images were processed using Gaussian filtering, then a fast Fourier transform (FFT) filtering was applied to the original images, and lastly, the Gaussian and FFT-filtered images were multiplied. The procedure was applied to reduce the background due to the electron probe tail.



**Figure S12**. (a) Atomic structure of  $20.5^{\circ}$  grain boundary in monolayer WS<sub>2</sub>. The arrows highlight the commonly seen intrinsic point defects in monolayer WS<sub>2</sub>, namely single W vacancy (V<sub>W</sub>) and double W vacancy (V<sub>2W</sub>). (b) Zoom-in view of the grain boundary region highlighted by the yellow rectangle in (a) showing domains with  $20.5^{\circ}$  tilt angle and two parallel displaced cluster defect rings. The overlaid triangles denote  $20.5^{\circ}$  rotated domains.



**Figure S13**. Low-resolution STEM image showing a 30.5<sup>0</sup> monolayer (ML) grain boundary (GB). (b) Fast Fourier (FFT) filter image showing two sets of 6-fold diffraction spots. (c), and (d) FFT-filtered images indicating the lateral extension of the 2H/3R bilayer domains.



Figure S14. Atomic structure of twisted bilayer WS2. (a) Original ADF-STEM image. (b) Processed STEM image. (c) FFT power spectrum showing 3 sets of 6-fold diffraction spots which stem from three distinct grains. (d-f) FFT-filtered images show the spatial extension of the monolayers and bilayers shown in (a). The FFT legends marked in yellow, purple, and green correspond to the FFT-filtered images from the spot areas highlighted in (c). The contour lines extracted from FFT-filtered images in (d-f) are overlapped in (b).



Figure S15. a) AFM line profiles of single-and multilayer WS<sub>2</sub> crystals. The blue dots in (a) are used to extract a thickness of ~0.65 nm. b) AFM image and line profile of single-layer crystals revealing a thickness of ~0.75 nm.

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