

## Supporting Information

# Ultrafast growth of submillimeter-scale single-crystal MoSe<sub>2</sub> by pre-alloying CVD

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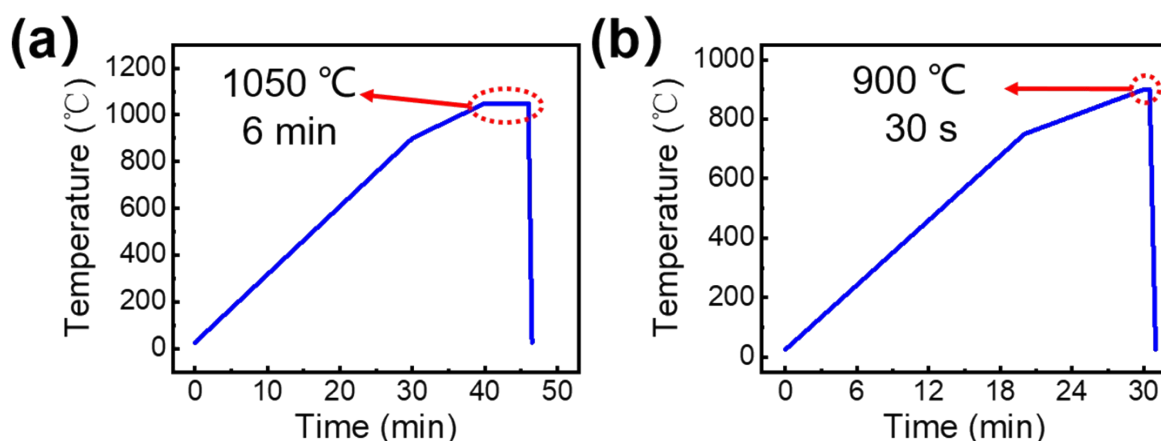
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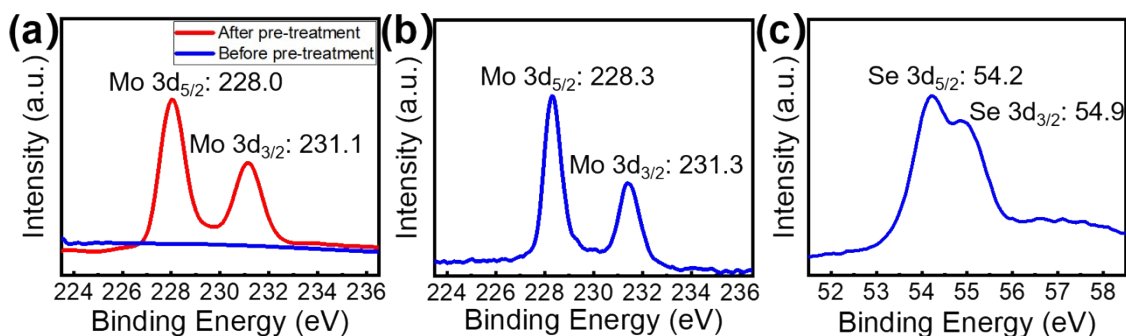
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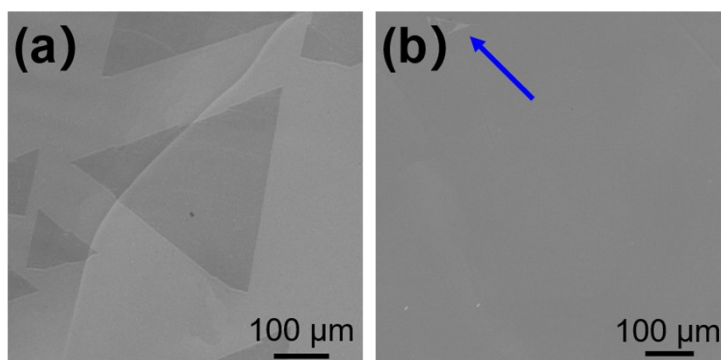


**Figure S1.** Schematic of (a) pre-alloying process for Au foils and (b) CVD process to grow large-scale monolayer MoSe<sub>2</sub> single crystals on pre-alloyed Au foils.

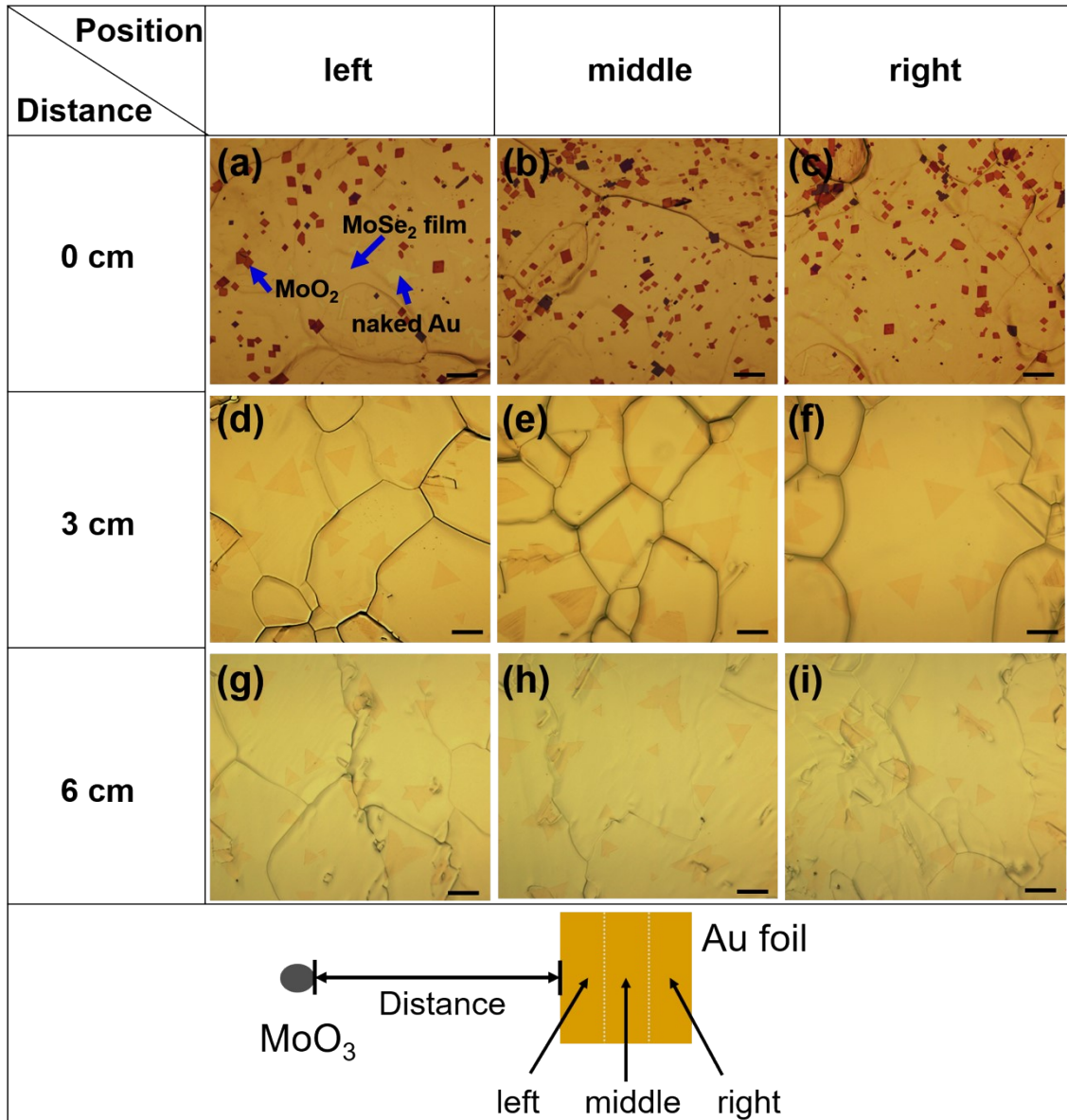


**Figure S2.** XPS spectra of elemental changes on Au foils. (a) The variation of Mo content in Au foils before and after pre-treatment with MoO<sub>3</sub>. The XPS spectra of (b) Mo and (c) Se in the pre-alloyed Au foils detected after the MoSe<sub>2</sub> growth.

Au foils were pre-treated by H<sub>2</sub> reduction of MoO<sub>3</sub> to form pre-alloyed Au. Figure S2a shows that no Mo peak exists on the Au surface before pre-alloying. After pre-alloying, the peaks of 228.0 eV and 231.1 eV appear, which are consistent with Mo 3d<sub>5/2</sub> and Mo 3d<sub>3/2</sub> peaks, indicating the formation of Au-Mo alloys. Figure S2b and S2c exhibit that Mo 3d<sub>5/2</sub> and 3d<sub>3/2</sub> peaks are located at ~228.3 and 231.3 eV, while Se 3d<sub>5/2</sub> and 3d<sub>3/2</sub> peaks are located at ~54.2 and 54.9 eV, respectively, illustrating the formation of MoSe<sub>2</sub> on pre-alloyed Au foils. Mo in Au-Mo alloys or MoSe<sub>2</sub> can be distinguished by the position of Mo peaks in XPS spectra.



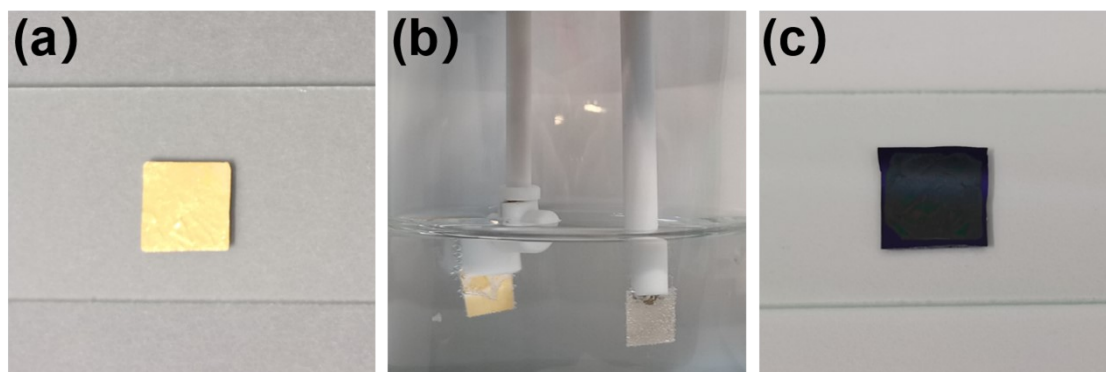
**Figure S3.** The SEM images of MoSe<sub>2</sub> (a) single crystals and (b) a continuous film. The blue arrow in (b) shows the naked Au area without MoSe<sub>2</sub>. MoSe<sub>2</sub> film over a large area represents uniformly monolayer without any multilayer.



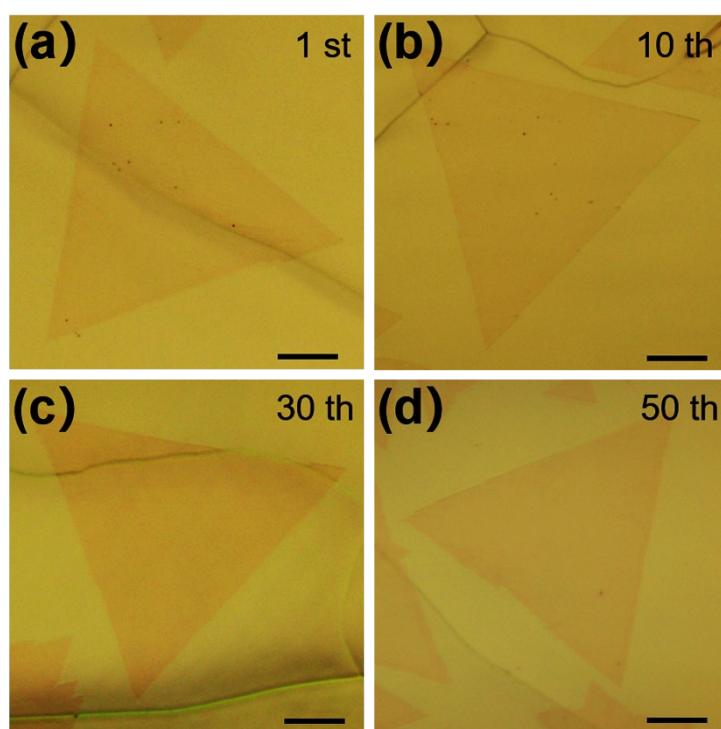
**Figure S4.** Optical images of MoSe<sub>2</sub> grown on the different areas (left, middle, right) of pre-alloyed Au foils with changing the distance (D) between Au foils and MoO<sub>3</sub> powders for (a-c) D = ~0 cm, (d-f) D = ~3 cm, and (g-i) D = ~6 cm. Au foils were pre-alloyed at 1050 °C for 4 times. The scale bars in all images are 100 μm.

We have explored morphology evolution of MoSe<sub>2</sub> with changing the distance (D) between Au foils and MoO<sub>3</sub> powders in Figure S4. When Au foil was very close to MoO<sub>3</sub> (D=0 cm, Figure S4a-c,) MoSe<sub>2</sub> domains join together to form a film and parallelogram-like MoO<sub>2</sub> also appeared. When D was increased to ~3 cm, the individual triangle MoSe<sub>2</sub> single crystals were obtained with the disappearance of MoO<sub>2</sub> flakes (Figure S4d-f). With further increasing D to ~6 cm (Figure S4g-i), MoSe<sub>2</sub> domains become smaller and a little irregular because of the insufficient Mo source supply. The above results indicate that the morphology of MoSe<sub>2</sub> depended on D due to the MoO<sub>3</sub> with point-like distribution.

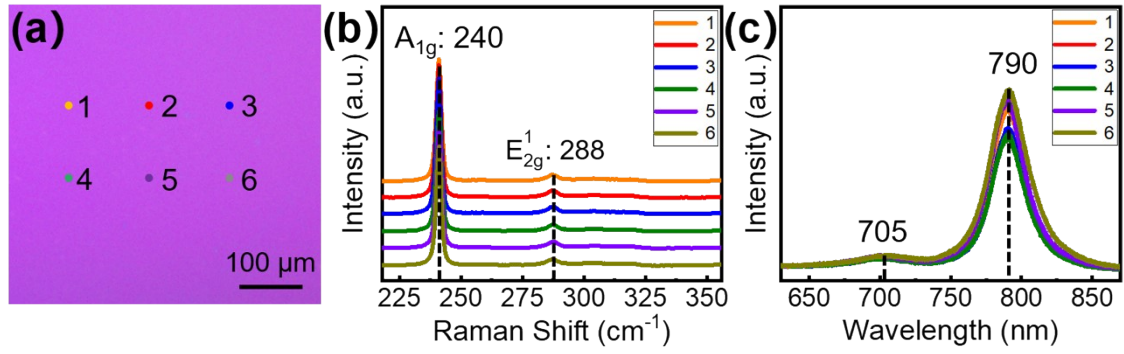
However, in our experiments, the size of Au foils was  $1\text{ cm} \times 1\text{ cm}$ . When D was fixed at 3 cm, the morphology of  $\text{MoSe}_2$  on different areas of Au foils (left, middle, right) was very similar and  $\text{MoSe}_2$  domains distribute on the whole Au surface uniformly.



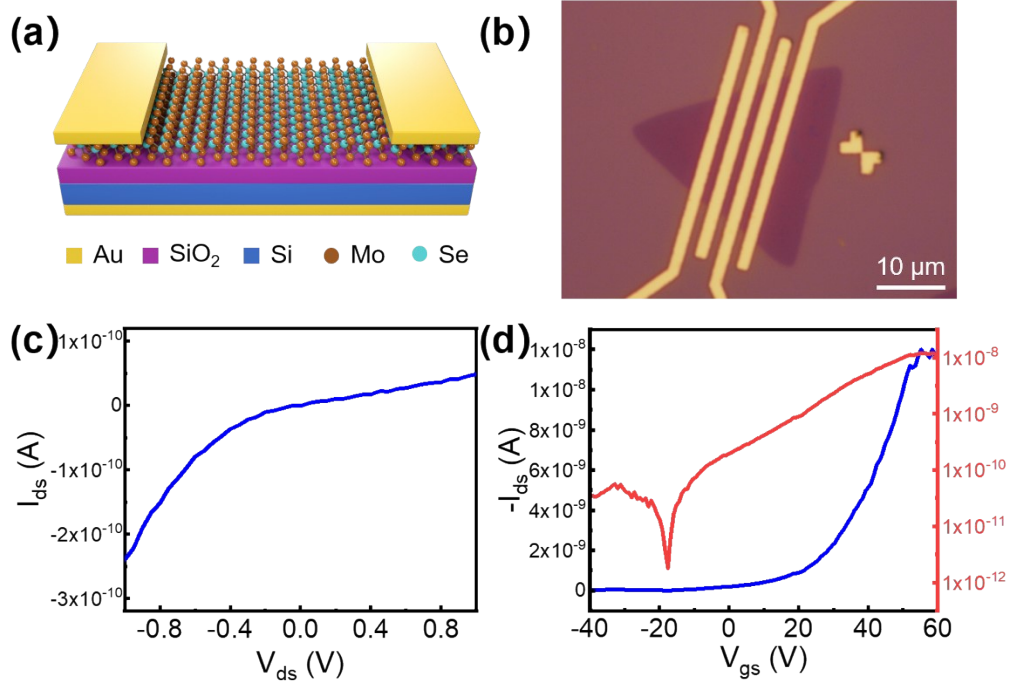
**Figure S5.** Schematic diagram of the electrochemical bubbling method for transferring  $\text{MoSe}_2$  single crystals and their films. Photographs of (a)  $\text{MoSe}_2/\text{Au}$  after spin-coated with PMMA, (b) the electrolysis process and (c)  $\text{PMMA}/\text{MoSe}_2$  transferred onto the  $\text{SiO}_2/\text{Si}$  substrate.



**Figure S6.** The optical images of single-crystal  $\text{MoSe}_2$  domains grown on the same pre-alloyed Au foil for different times. All the  $\text{MoSe}_2$  domains show the similar morphology. Before the  $\text{MoSe}_2$  growth, Au foil was pre-alloyed at  $1050\text{ }^\circ\text{C}$  for 20 times (see Experimental, Figure 1a). Scale bar in all images is  $20\text{ }\mu\text{m}$ .

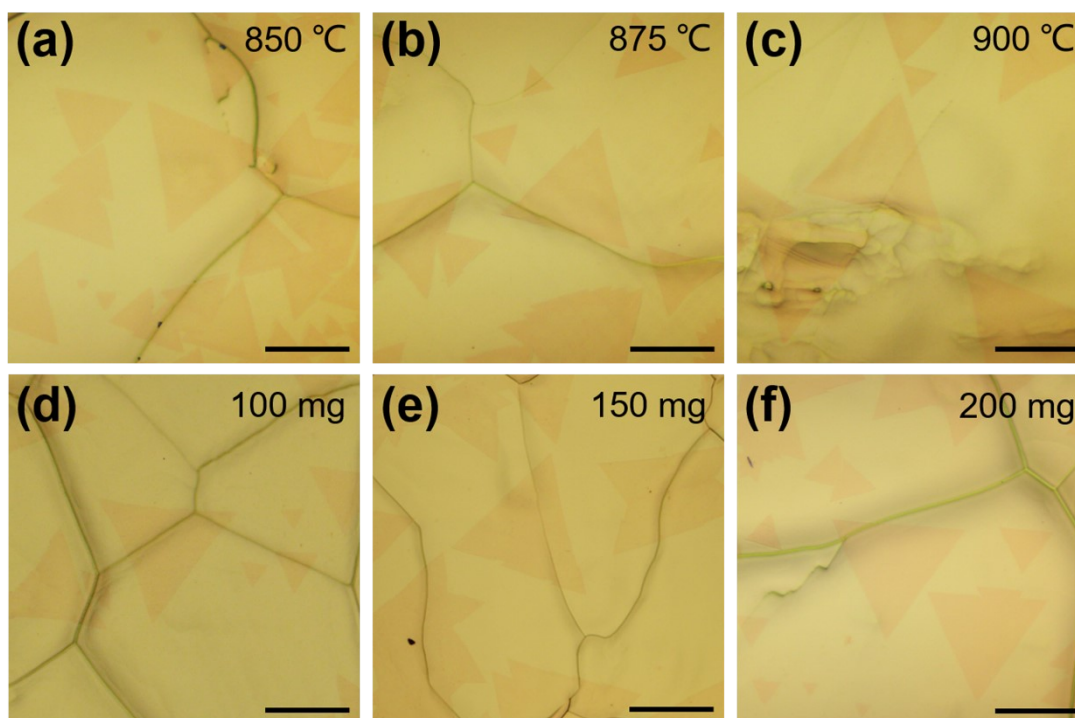


**Figure S7.** (a) Optical image of monolayer MoSe<sub>2</sub> film transferred on the SiO<sub>2</sub>/Si substrate. (b) Raman and (c) PL spectra taken from 6 positions marked with different colors in (a), showing the characteristic peaks of monolayer MoSe<sub>2</sub>. Considering that mapping a large area is very time-consuming, Raman and PL spectra were randomly taken in the MoSe<sub>2</sub> film.

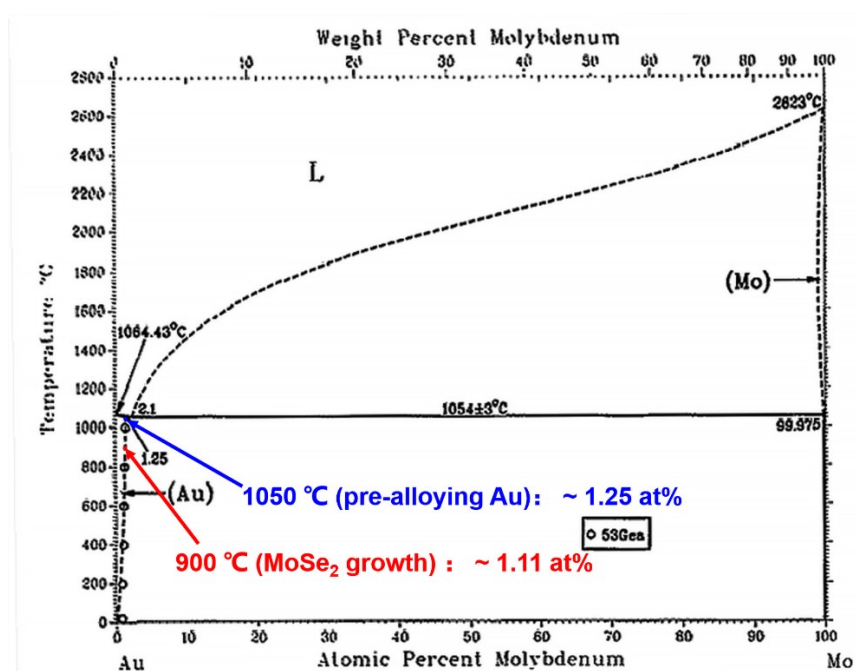


**Figure S8.** Electrical properties of monolayer single-crystal MoSe<sub>2</sub>. (a) Schematic diagram and (b) the optical image of the back-gate FET device based on the single-crystal MoSe<sub>2</sub> domain. (c) Output characteristic and (d) transfer characteristic of MoSe<sub>2</sub> FET device. The non-linear output characteristic in (c) presents a Schottky contact between the MoSe<sub>2</sub> and Au electrodes which may be caused by the inevitable contaminations during the fabrication of devices. The transfer characteristic in (d) indicates that as-grown MoSe<sub>2</sub> is a typical n-type semiconductor. The carrier mobility and ON/OFF ratio evaluated under  $V_{ds}$  of 0.1 V from (d) is 11.6 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> and  $\sim 10^3$ , respectively.

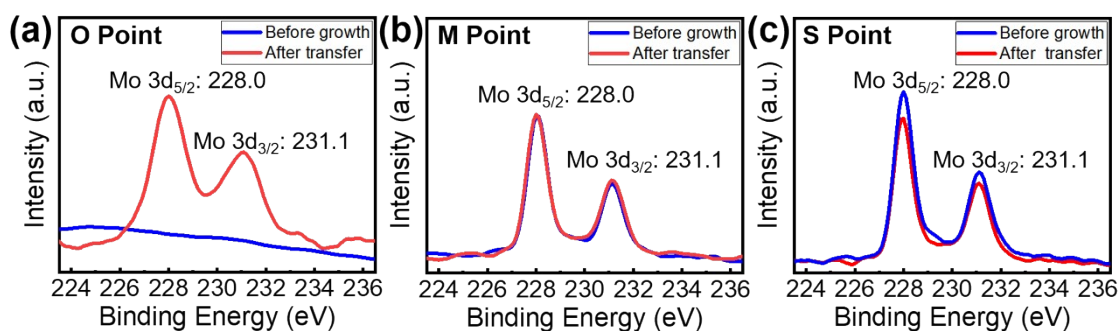




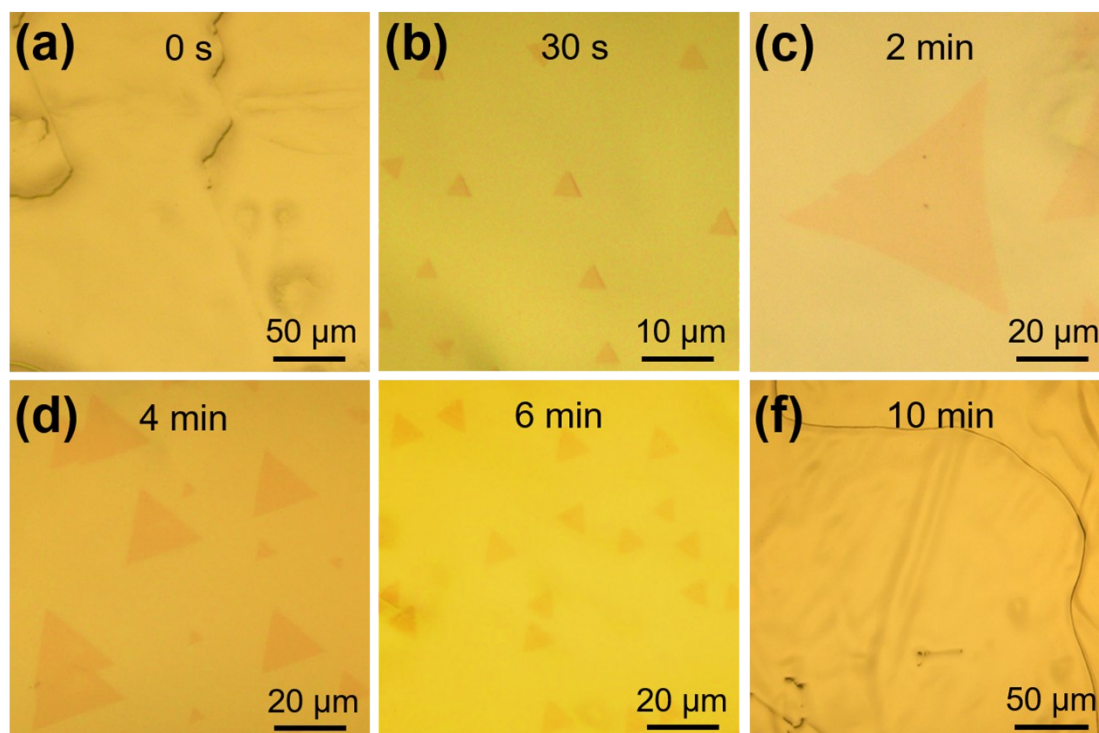
**Figure S9.** Optical images of MoSe<sub>2</sub> grown on the original Au foils under (a-c) different growth temperature and (d-f) different Se content. Scale bar in all images is 50  $\mu\text{m}$ .



**Figure S10.** Au-Mo phase diagram.<sup>41</sup>

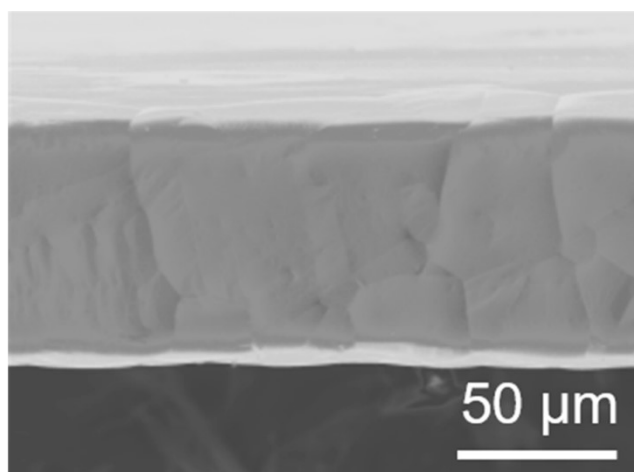


**Figure S11.** XPS spectra collected on the Au foils before and after MoSe<sub>2</sub> growth (Then MoSe<sub>2</sub> were totally transferred from Au foils). (a) At O Point, Mo content in Au increases after MoSe<sub>2</sub> growth, due to the formation of Au-Mo alloys along with the MoSe<sub>2</sub> growth on the pristine Au. (b) At M Point, the Mo content in the pre-alloyed Au remains unchanged because Mo was saturated at the MoSe<sub>2</sub> growth temperature (900 °C). (c) At S Point, the Mo content in the pre-alloyed Au is oversaturated at 900 °C. The excess Mo atoms diffuse onto the Au surface, so that the Mo content in pre-alloyed Au slightly decreases after MoSe<sub>2</sub> growth.

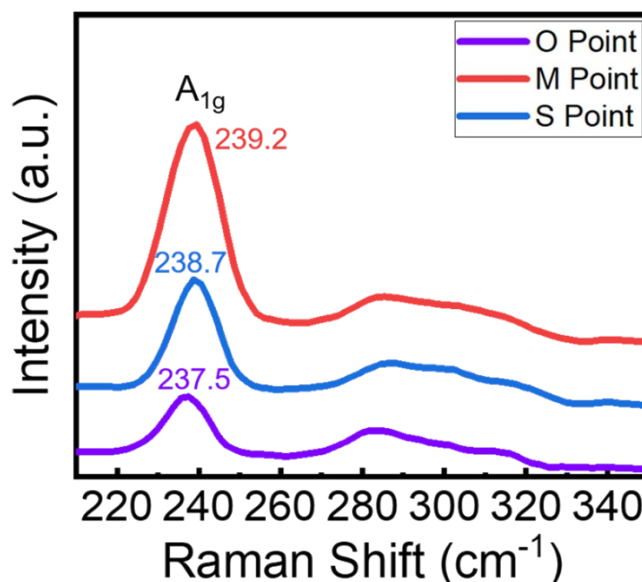


**Figure S12.** Optical images of MoSe<sub>2</sub> single crystals synthesized on pre-alloyed Au foils without MoO<sub>3</sub> precursor supply. The Au foil has been pre-alloyed for 25 times (see Experimental). The growth time in (a-f) varied from 0 to 12 min while keeping all the other parameters constant. We can see that the largest domains can be obtained within 2 min. The Mo content in the pre-alloyed Au is not enough to grow MoSe<sub>2</sub> for more than 10 min without the continuous Mo source supply.

Figure S12b and Figure 4a (the last figure) show MoSe<sub>2</sub> growth on pre-alloyed Au in 30 s without and with extra MoO<sub>3</sub> supply. Many small MoSe<sub>2</sub> nucleus ( $\sim 4\ \mu\text{m}$ ) were formed without MoO<sub>3</sub> supply because of precipitation of pre-deposited Mo (Figure S12b). However, MoSe<sub>2</sub> domains can be increased 27 times larger ( $107\ \mu\text{m}$ ) after introducing extra MoO<sub>3</sub> supply (Figure 4a). The phenomenon proves that although the pre-deposited Mo can be act as Mo source for the MoSe<sub>2</sub> growth, it has a slight effect on promoting the grain size of MoSe<sub>2</sub>. The main Mo source for MoSe<sub>2</sub> growth come from the extra MoO<sub>3</sub> powders but not pre-deposited Mo.

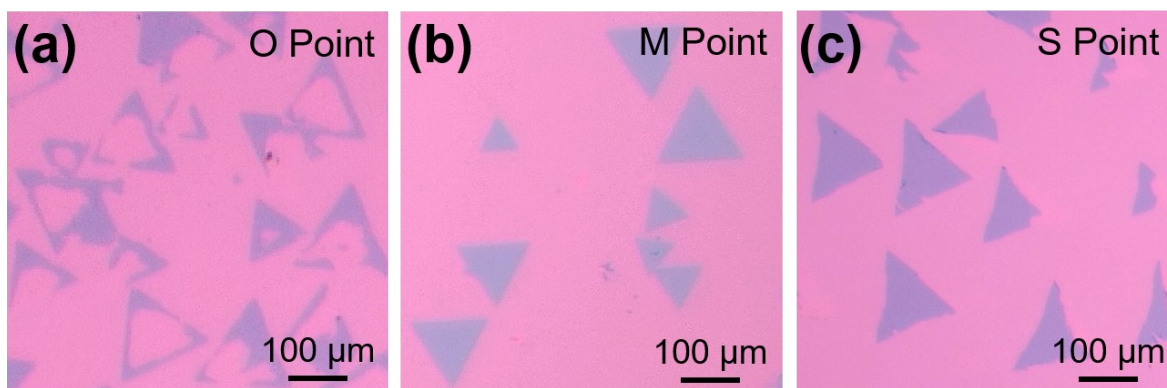


**Figure S13.** The SEM image of the cross section of pre-alloyed Au foil.

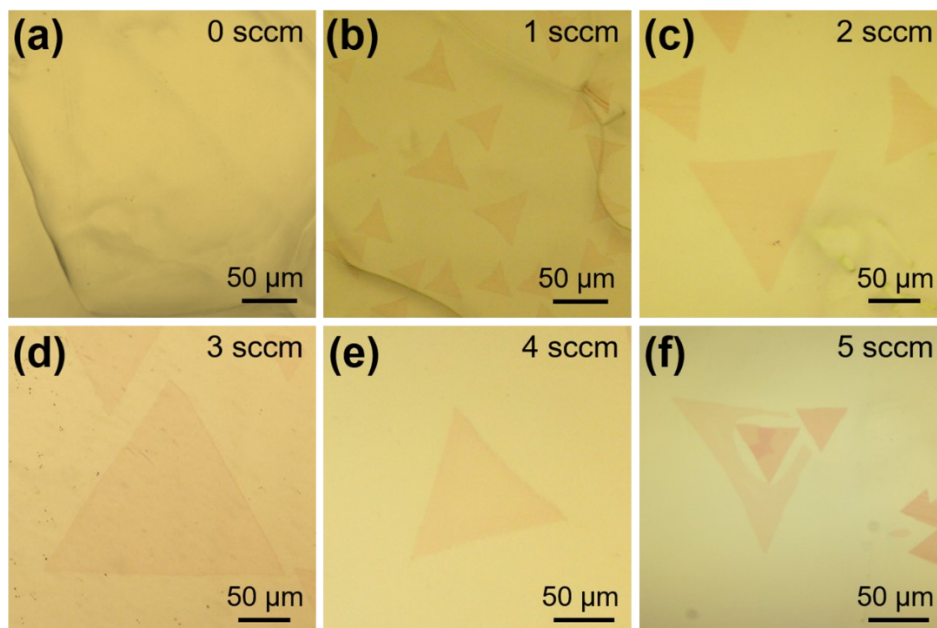


**Figure S14.** Raman spectra of MoSe<sub>2</sub> collected on Au foils with different Mo content corresponding to O Point, M Point and S Point in Figure 4c. The A<sub>1g</sub> peaks at M and S Point have a blue shift than that at O Point, indicating a relative weak interaction between MoSe<sub>2</sub> and the pre-alloyed Au.



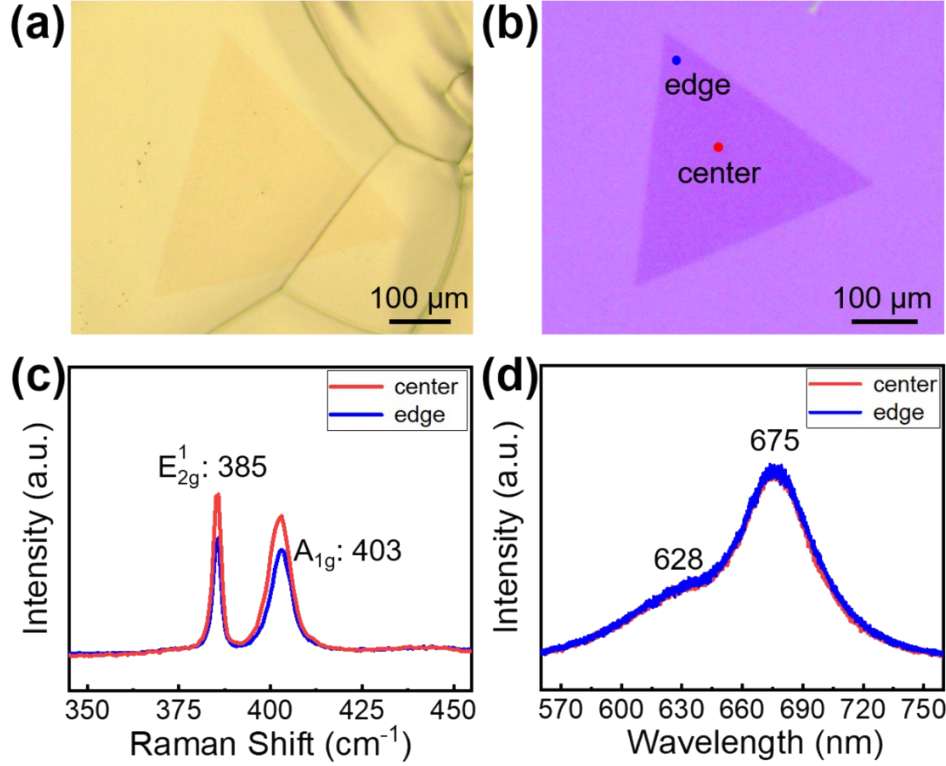


**Figure S15.** Optical images of MoSe<sub>2</sub> domains transferred from Au foils onto SiO<sub>2</sub>/Si substrates. MoSe<sub>2</sub> domains were obtained on (a) the original Au without pre-alloying (O Point), the pre-alloyed Au at (b) M Point and (c) S Point. MoSe<sub>2</sub> domains were seriously damaged in (a), while the whole triangle was completely transferred in (b).



**Figure S16.** Optical images of MoSe<sub>2</sub> grown on pre-alloyed Au foils with different H<sub>2</sub> flow rate. Before the MoSe<sub>2</sub> growth, Au foils were pre-alloyed at 1050 °C for 5 times. Except for changing the H<sub>2</sub> flow rate, we kept all the other CVD parameters constant to eliminate the uncertain factors.

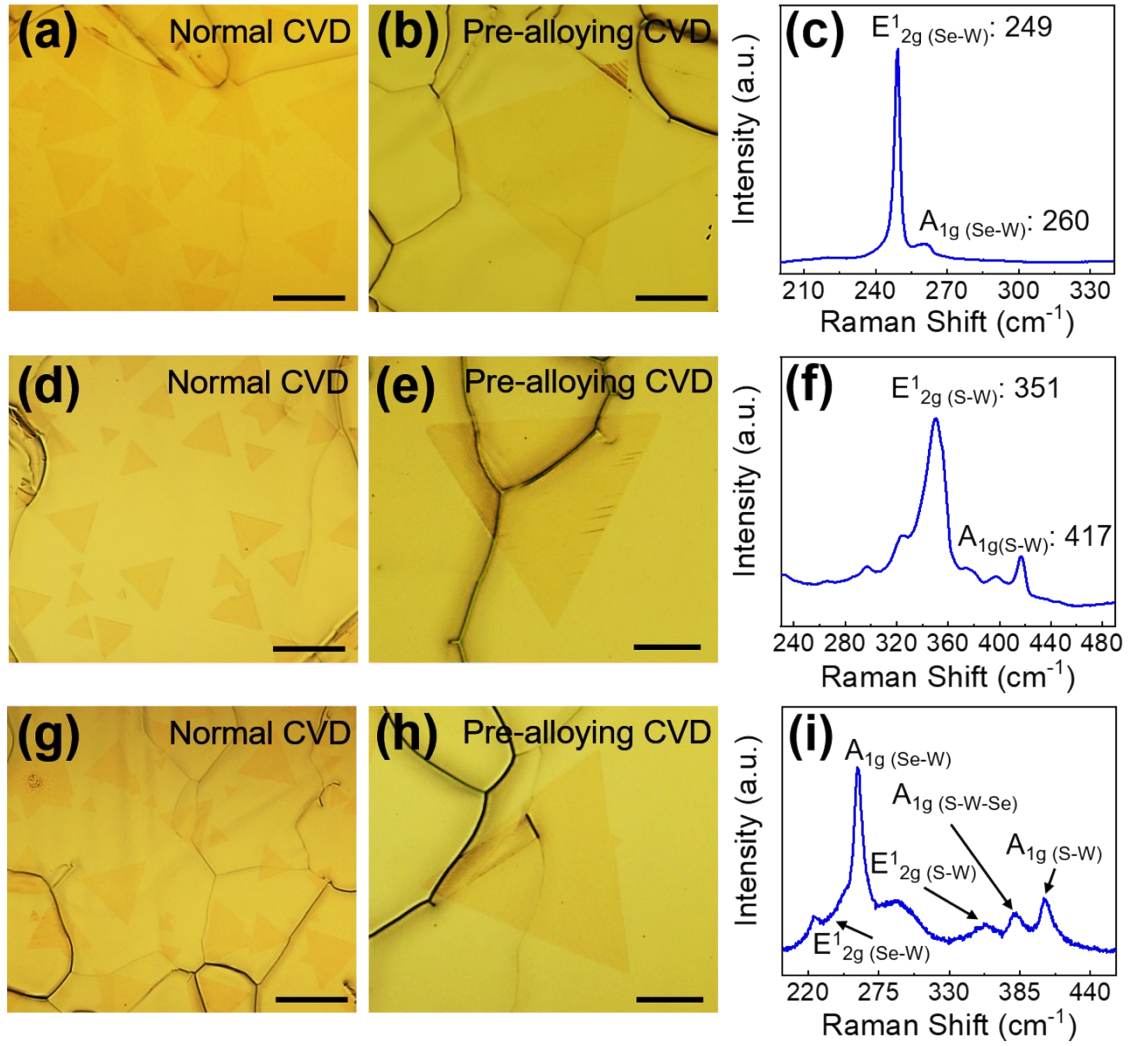
The grain size of monolayer MoSe<sub>2</sub> single crystals increases along with the H<sub>2</sub> concentration increasing from 0 to 3 sccm. Submillimeter-scale single-crystal MoSe<sub>2</sub> domain was achieved with a moderate H<sub>2</sub> flow of 3 sccm. Then after, grain size decreases and MoSe<sub>2</sub> multilayers appear at a higher H<sub>2</sub> flow rate.



**Figure S17.** (a) An optical image of  $\sim 450\ \mu\text{m}$ -sized  $\text{MoS}_2$  single crystal grown by pre-alloying CVD. (b)  $\text{MoS}_2$  transferred to the  $\text{SiO}_2/\text{Si}$  substrate by electrochemical bubbling method. (c) Raman and (d) PL spectra taken from the center and edge of single-crystal  $\text{MoS}_2$  domain in (b).

Monolayer  $\text{MoS}_2$  were obtained on pre-alloyed Au by changing the growth precursors. The 155 mg S powder (99.5 wt%) was separately heated to  $\sim 200\ ^\circ\text{C}$  at the upstream of the pre-alloyed Au foil outside the furnace. The tube furnace was heated from room temperature to  $750\ ^\circ\text{C}$  within 20 min and then to  $900\ ^\circ\text{C}$  in 10 min under an Ar flow of 100 sccm. Then after, 3 sccm  $\text{H}_2$  was turned on for 2 min.

Figure S17 shows that  $\sim 450\ \mu\text{m}$ -sized triangular  $\text{MoS}_2$  was synthesized within 2 min, with a growth rate of  $\sim 3.75\ \mu\text{m}\ \text{s}^{-1}$ . Obviously, the value is much lower than the growth rate of  $\text{MoSe}_2$  ( $\sim 18.7\ \mu\text{m}\ \text{s}^{-1}$ ). Although the growth mechanism is similar, the growth dynamics are different between  $\text{MoS}_2$  and  $\text{MoSe}_2$ . In our previous reports,<sup>13</sup> we have demonstrated that  $\text{S}_2$  dimer dissociate into S atoms firstly and then S atoms diffuse to form  $\text{WS}_2$ . Such two processes need to overcome 0.22 eV barriers for  $\text{S}_2$  dissociation and 0.58 eV barriers for S atom diffusion. By contrast, there exists only one process with a relatively smaller energy barrier (0.27 eV) for Se atoms diffusion to form  $\text{WSe}_2$ .<sup>20</sup> Therefore, the growth rate of  $\text{MoSe}_2$  is higher than  $\text{MoS}_2$ .



**Figure S18.** Comparison of (a-c)  $\text{WSe}_2$ , (d-f)  $\text{WS}_2$  and (g-i)  $\text{WS}_{2x}\text{Se}_{2(1-x)}$  obtained by normal and pre-alloying CVD. Figure c, f and i are the corresponding Raman spectra. Au foils were pre-alloyed at 1050 °C for 9 times. All scale bars are 100  $\mu\text{m}$ .  $\text{WSe}_2$  was grown at 900 °C for 2 min with an Ar and  $\text{H}_2$  flow of 100 and 3 sccm.  $\text{WS}_2$  was grown at 850 °C for 5 min with an Ar and  $\text{H}_2$  flow of 100 and 2 sccm.  $\text{WS}_{2x}\text{Se}_{2(1-x)}$  was obtained at 875 °C for 5 min with an Ar and  $\text{H}_2$  flow of 120 and 3 sccm. The samples obtained by pre-alloying CVD have much lower nucleation density and larger grain size.

**Table S1.** Comparison of growth method, grain size of single-crystal TMDs.

Material	Substrate	Temperature (°C)	Time	Size	Rate ( $\mu\text{m/s}$ )	Property	References
MoS <sub>2</sub>	SiO <sub>2</sub> /Si	700	5 min	120 $\mu\text{m}$	0.40	—	1
	Sapphire	850	10 min	350 $\mu\text{m}$	0.58	$\mu=90 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF $10^7$	2
	SiO <sub>2</sub> /Si	850	10 min	305 $\mu\text{m}$	0.51	$\mu=28 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF $10^6$	3
	SiO <sub>2</sub> /Si	800	5 min	500 $\mu\text{m}$	1.67	—	4
	Sapphire	775	10 min	170 $\mu\text{m}$	0.28	$\mu=3 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	5
	Anyone	750	10 min	200 $\mu\text{m}$	0.33	$\mu=90 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF $10^7$	6
	SiO <sub>2</sub> /Si	770	15 min	500 $\mu\text{m}$	0.56	$\mu=48.7 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF $10^7$	7
	Au foil	680	60 min	50 $\mu\text{m}$	0.01	Tafel slope: 61 mV dec <sup>-1</sup> J: 38.1 $\mu\text{A cm}^{-2}$	8
	Au foil	680	60 min	81 $\mu\text{m}$	0.02	—	9

	Au foil	680	60 min	115 $\mu\text{m}$	0.03	—	10
	<b>Au foil</b>	<b>900</b>	<b>2 min</b>	<b>450 <math>\mu\text{m}</math></b>	<b>3.75</b>	—	<b>Our work</b>
WS <sub>2</sub>	SiO <sub>2</sub> /Si	750	5 min	178 $\mu\text{m}$	0.59	—	11
	Sapphire	900	60 min	135 $\mu\text{m}$	0.04	$\mu=4.1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	12
	Au foil	800	4 h	600 $\mu\text{m}$	0.04	$\mu=2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF 10 <sup>7</sup>	13
	Au foil	935	15 min	420 $\mu\text{m}$	0.47	$\mu=20 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF 10 <sup>8</sup>	14
	SiO <sub>2</sub> /Si	900	10 min	250 $\mu\text{m}$	0.42	$\mu=32.3 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	15
	SiO <sub>2</sub> /Si	950	5 min	360 $\mu\text{m}$	1.20	—	4
	SiO <sub>2</sub> /Si	1070	15 min	256 $\mu\text{m}$	0.28	$\mu=2.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	16
WSe <sub>2</sub>	Sapphire	850	15 min	50 $\mu\text{m}$	0.06	$\mu=90 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	17
	SiO <sub>2</sub> /Si	720	30 min	168 $\mu\text{m}$	0.09	$\mu=100 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF 10 <sup>7</sup>	18
	SiO <sub>2</sub> /Si	900	20 min	800 $\mu\text{m}$	0.67	—	19
	Au foil	950	30 s	780 $\mu\text{m}$	26.00	$\mu=143 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	20



						ON/OFF $9 \times 10^6$	
	SiO <sub>2</sub> /Si	1050	10 min	570 $\mu\text{m}$	0.95	$\mu < 0.1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	21
	Quartz	850	20 min	100 $\mu\text{m}$	0.08	PL QY $\sim 60\%$	22
	hBN	800	20 min	380 $\mu\text{m}$	0.32	$\mu = 4.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	23
MoSe <sub>2</sub>	SiO <sub>2</sub> /Si	750	10 min	30 $\mu\text{m}$	0.05	—	24
	SiO <sub>2</sub> /Si	820	15 min	40 $\mu\text{m}$	0.04	R=13 mA W <sup>-1</sup>	25
	SiO <sub>2</sub> /Si	750	20 min	135 $\mu\text{m}$	0.11	$\mu = 50 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF $> 10^6$	26
	SiO <sub>2</sub> /Si	750	10 min	50 $\mu\text{m}$	0.08	—	27
	SiO <sub>2</sub> /Si	750	20 min	1.3 mm	1.08	$\mu = 42 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF $10^6$	19
	Molten glass	1050	5 min	2.5 mm	8.33	$\mu = 95 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF $> 10^7$	28
	SiO <sub>2</sub> /Si Quartz	750	20 min	100 $\mu\text{m}$	0.08	—	29
	SiO <sub>2</sub> /Si	730	15 min	195 $\mu\text{m}$	0.22	—	30
	SiO <sub>2</sub> /Si	730	3 min	135 $\mu\text{m}$	0.75	—	31
	<b>Au foil</b>	<b>900</b>	<b>30 s</b>	<b>560 <math>\mu\text{m}</math></b>	<b>18.67</b>	<b><math>\mu = 11.6 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}</math></b>	<b>Our work</b>

**Table S2.** Comparison of single-crystal TMDs grown on Au.

Materials	Temperature (°C)	Time	Size	Rate ( $\mu\text{m/s}$ )	Property	References
ReSe <sub>2</sub>	750	15 min	Monolayer parallelogram: 20 $\mu\text{m}$	0.02	—	32
	750	15 min	—	—	—	33
	750	30 min	Monolayer: 30 $\mu\text{m}$	0.02	—	34
ReS <sub>2</sub>	800	5 min	—	—	$\mu=2.3 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ $R=178.75 \text{ A W}^{-1}$	35
PtTe <sub>2</sub>	800	10 min	5.6-50 nm Rectangular: 200 $\mu\text{m}$ ; triangular: 30 $\mu\text{m}$	Rectangular: 0.33 Triangular: 0.05	$R=250 \text{ mA W}^{-1}$	36
PtSe <sub>2</sub>	850	20 min	Irregular: 245 $\mu\text{m}$	0.20	Tafel slope: $\sim 33$ $\text{mV dec}^{-1}$ $J: \sim 215 \mu\text{A cm}^{-2}$	37
TaSe <sub>2</sub>	930	5-25 min	Different thickness: 5-20 $\mu\text{m}$	—	—	38
TaS <sub>2</sub>	750	10 min	Different thickness: 20 $\mu\text{m}$	0.03	Tafel slope: 33-42 $\text{mV dec}^{-1}$ $J: 100\text{-}179 \mu\text{A cm}^{-2}$	39
WS <sub>2</sub>	800	4 h	600 $\mu\text{m}$	0.04	$\mu=2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF $10^7$	13

	935	15 min	420 $\mu\text{m}$	0.47	$\mu=20 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF $10^8$	14
$\text{WSe}_2$	950	30 s	780 $\mu\text{m}$	26.00	$\mu=143 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF $9 \times 10^6$	20
$\text{MoS}_2$	680	60 min	50 $\mu\text{m}$	0.01	Tafel slope: 61 mV $\text{dec}^{-1}$ J: 38.1 $\mu\text{A cm}^{-2}$	8
	680	60 min	81 $\mu\text{m}$	0.02	—	9
	680	60 min	115 $\mu\text{m}$	0.03	—	10
	720	8 min	film	—	$\mu=11.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ON/OFF $7.7 \times 10^5$	40
	<b>900</b>	<b>2 min</b>	<b>450 <math>\mu\text{m}</math></b>	<b>3.75</b>	—	<b>Our work</b>
$\text{MoSe}_2$	<b>900</b>	<b>30 s</b>	<b>560 <math>\mu\text{m}</math></b>	<b>18.67</b>	$\mu=11.6 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	<b>Our work</b>

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