Support Information

Growth of $MoS_{2(1-x)}Se_{2x}$ (x = 0.41 - 1.00) Monolayer Alloys with Controlled Morphology by Physical Vapor Deposition

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KEYWORDS: Physical vapor deposition, $MoS_{2(1-x)}Se_{2x}$, Alloy, Morphology, Tunable band gap, Raman Spectrum

Mass Ratio	T_{Se} (℃)	T_1(℃)	T_2 (℃)	T_3 (℃)	PL/ nm	morphology
$m_{MoS_2/MoSe_2}=1:3$	300	960	960	350	730 ± 2	Tringle
		950	960	350	745 ± 3	Tringle
$m_{MoS_2/MoSe_2}=1:5$		950	960	350	760 ± 3	Tringle
$m_{MoS_2/MoSe_2}=1:8$		950	960	350	775 ± 2	Tringle
$m_{MoS_2/MoSe_2}=1:9$		950	960	350	785 ± 3	Tringle
$m_{MoS_2/MoSe_2}=1:9$		940	970	350	790 ± 3	Tringle
Only $MoSe_2$			960	350	800 ± 2	Tringle

Figure S1. Summary of A-exciton emission energy for all synthesized $MoS_{2(1-x)}Se_{2x}$ (x: 0.4-1) monolayers.

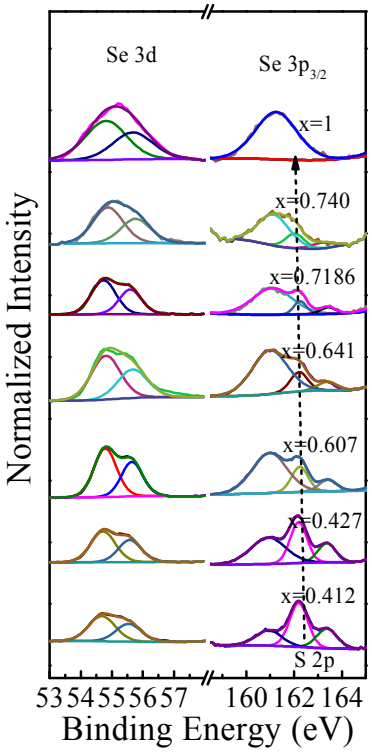

Figure S2. The XPS Spectra of As- Synthesized Monolayer $MoS_{2(1-x)}Se_{2x}$ alloy.

As shown in Figure S2, the intensity of Se_{3d} peak was adopted as normalized intensity, and

the area of S_{2p}, Se $_{3d}$ peaks was used to quantify. The S_{2p} peak intensity decreased with the Se composition increase, the $Se_{3p1/2}$ peak intensity increase against the Se composition increase.

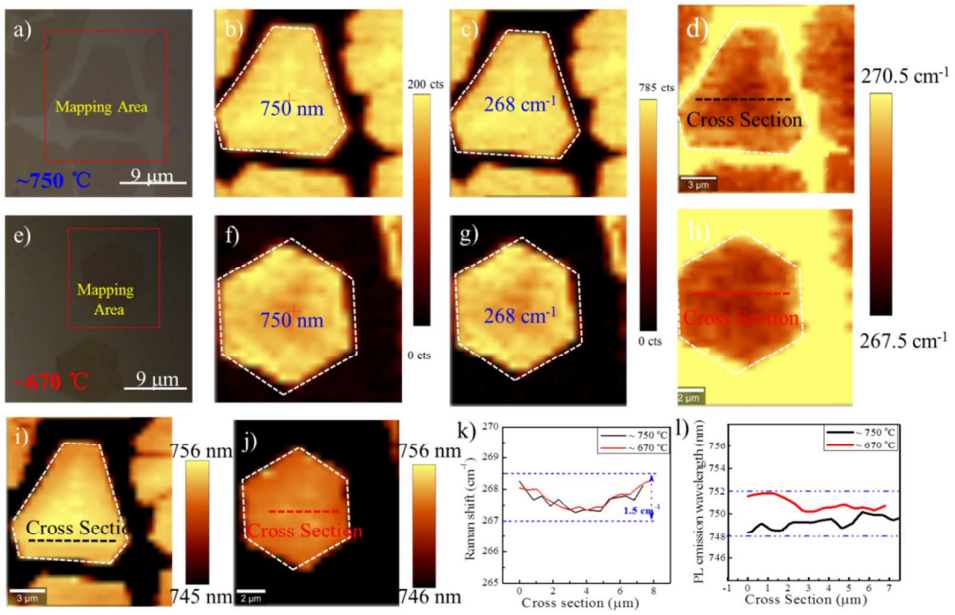

Figure S3. Different morphology of $MoS_{0.78}Se_{1.22}$ monolayer alloys. (a) Optical images (b) PL intensity map, (c) Raman intensity map, (d) Raman frequency map and (i) PL peak wavelength map of domains grown at 750 ^{o}C. (e) Optical images (f) PL intensity map, (g) Raman intensity map, (h) Raman frequency map and (j) PL peak wavelength map of domains grown at 670 ^{o}C. Cross-section of (k) Raman frequency profile and (l) PL peak wavelength profile along lines indicated in panel (i) and (j).

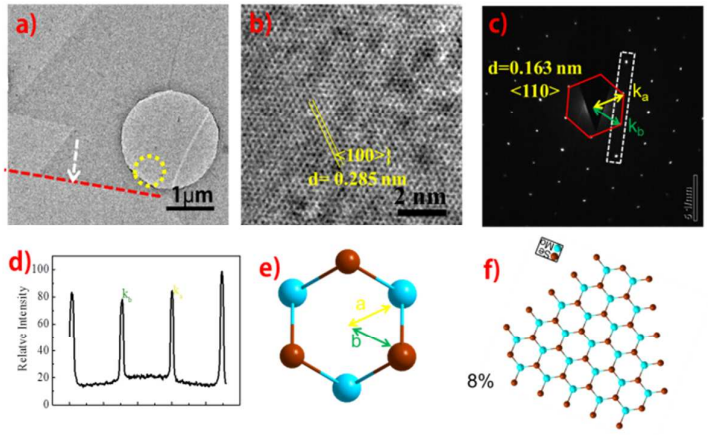

Figure S4. The Crystal Identification of As- Synthesized Monolayer $MoSe_2$

To identify crystal structure and edge orientation, the HRTEM and SAED pattern were conducted. The SAED pattern was measured at yellow cycle in image a, we can found that SAED pattern shows only one hexagon spot, and the lattice space is consistent with $MoSe_2$, demonstrated the monolayer $MoSe_2$ is single crystal. And the higher diffraction point intensity was related to Mo atom, and the weaker is Se atom. The angle between this direction and the triangle edge is almost 90 degree (perpendicular). This result demonstrated that this edge of triangle is zigzag. And the edge of the triangle is straight, not inward curving. According to the recent work, this edge would be Mo-zigzag orientation.

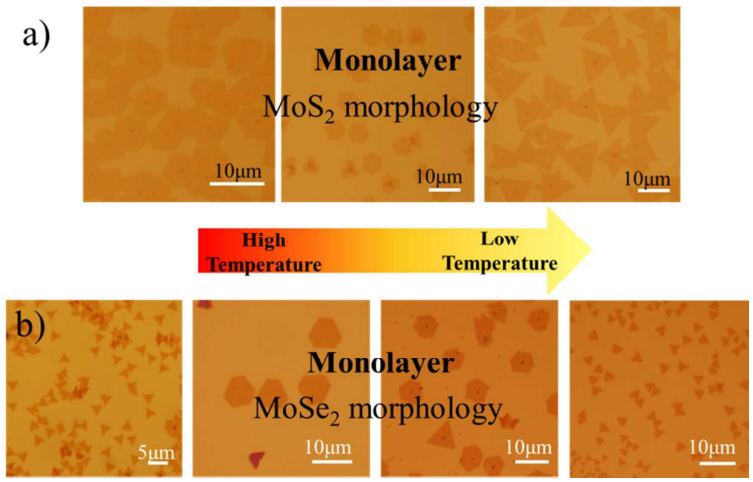

Figure S5. The morphology of monolayer MoS_2, $MoSe_2$ (under Se-rich condition)

dependent on the deposition temperature in the cooling zone.

As shown in Figure S5a-b, the subtriangular with long inward curving edge (similar to Figure 5a) at high temperature in the cooling zone, and transform to perfect regular hexagon (similar to Figure 5c) when temperature decreased to middle, and then change to regular triangle with straight edge at lower temperature (similar to Figure 5d) finally. The transformed process should be bi-directional from regular hexagon, as demonstrated in Figure 5e.

As shown in the Figure S 6a, the edge of long side is inward curving obviously, while other edge is striaght line along the edge. Otherhand, the edge of hexagon is very shape in figure S 6b. In addition, the intensity of raman mapping and PL mapping are also homogeneous over a large area.

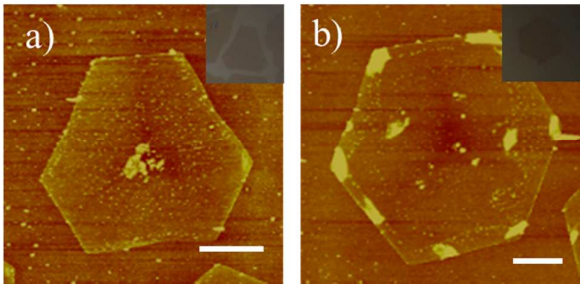

Figure S6. The AFM images of different morphology of monolayer $MoS_{0.78}Se_{1.22}$ alloys deposited at (a) 750 ^{o}C, (b) 670 ^{o}C.

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