Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

Damage-free and rapid transfer of CVD-grown two-dimensional transition metal dichalcogenides by dissolving sacrificial water-soluble layers

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Section S1. The schematic setup for the synthesis of mono- and few-layer MoS_2 flakes and films.

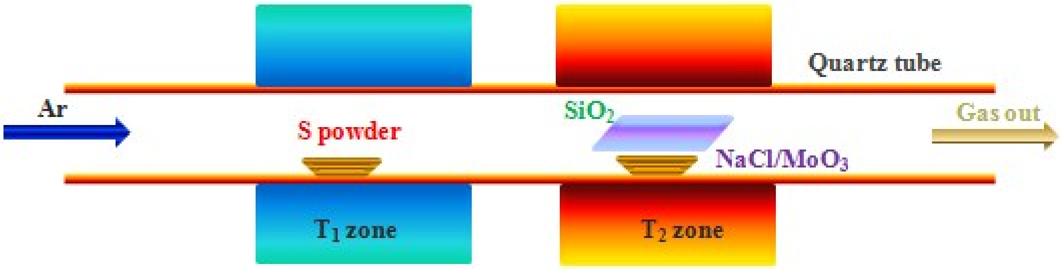

Fig. S1 The schematic setup for the growth of two-dimensional MoS_2.

Section S2. Optical images of the as-grown and transferred MoS_2 flakes and films.

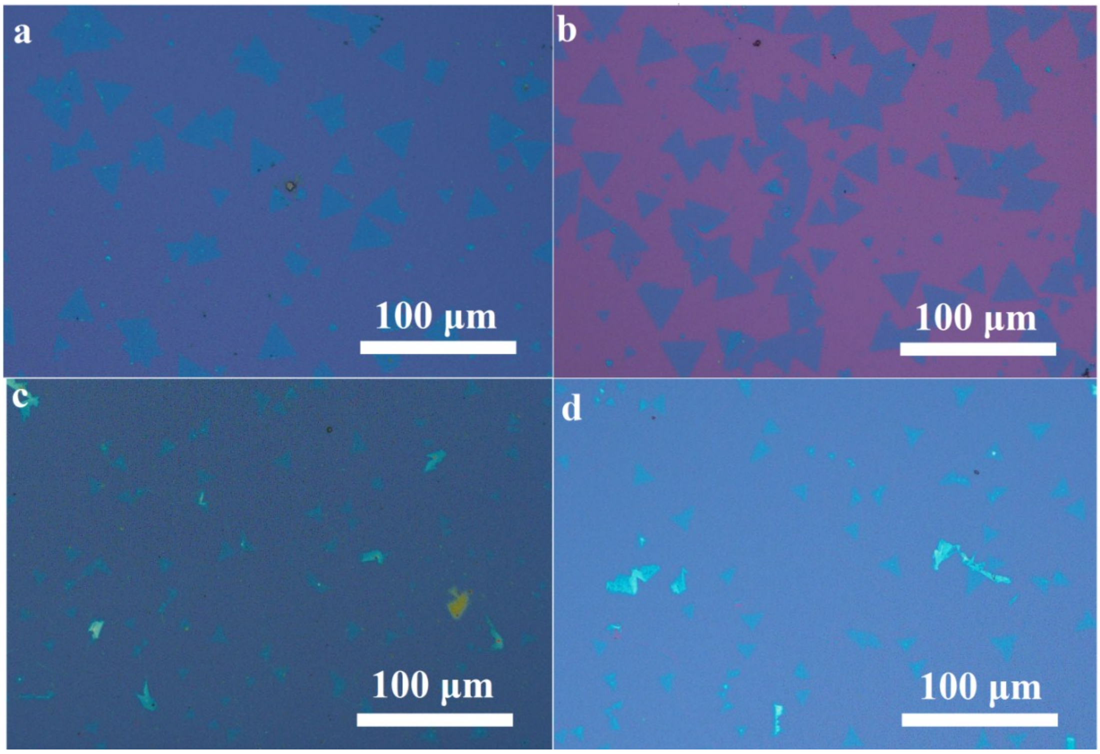

Fig. S2 Comparison of the as-grown and transferred MoS_2 flakes and films. Optical images of (a) as-grown and (b) transferred MoS_2 flakes; optical images of (c) as-grown and (d) transferred MoS_2 films. Note: (b, d) Transferred MoS_2 flakes and films are not the same area as the as-grown ones in (a, c), but on the same growth substrate.

Section S3. Additional XPS spectra of the transferred MoS_2 flakes.

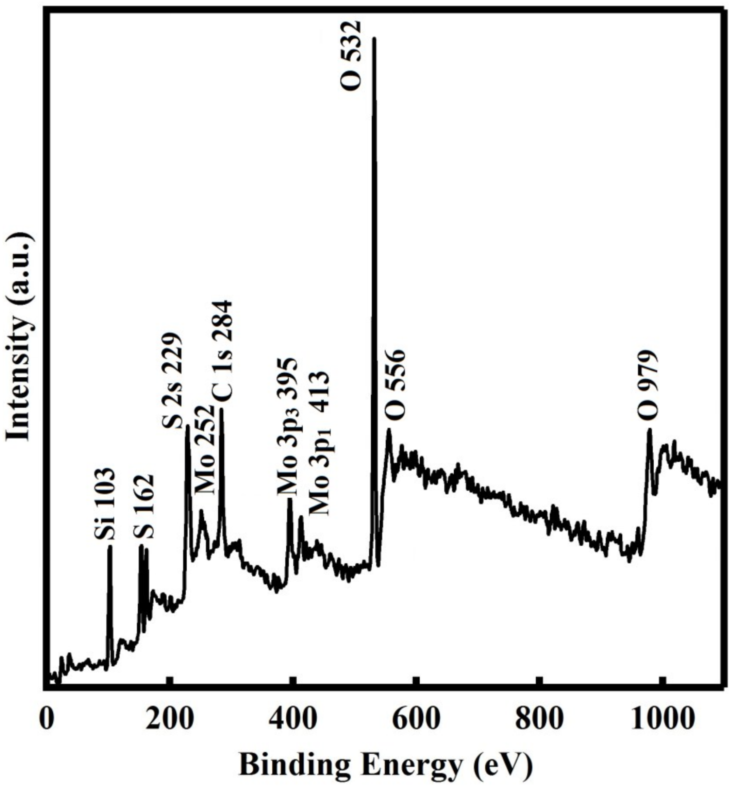

Fig. S3 XPS spectra of the transferred MoS_2 flakes on SiO_2/Si substrate. No residual Na and Cl elements were detected.

Section S4. EDS analysis of water-soluble substance on growth substrate and underneath MoS_2 flakes.

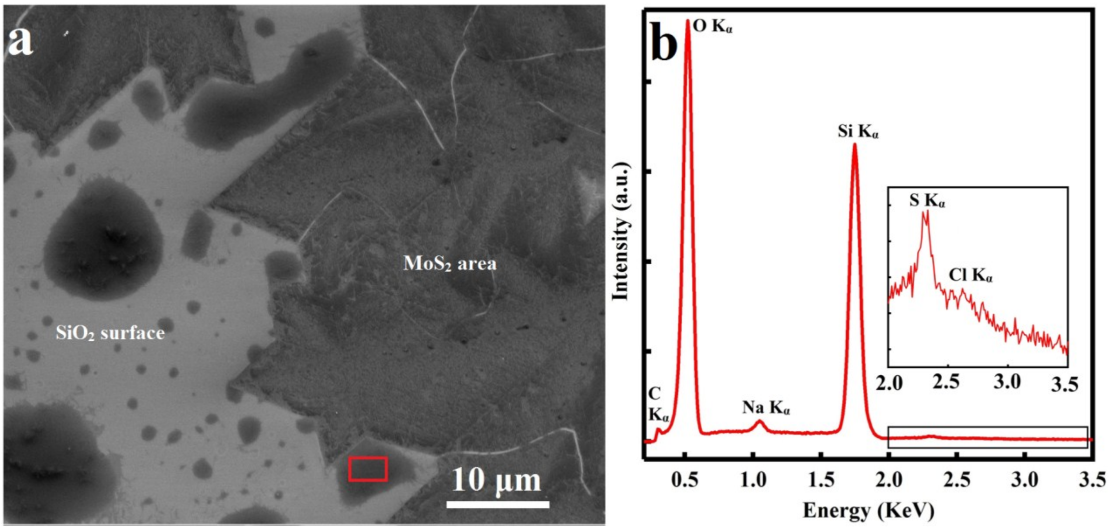

Fig. S4 Characterization of water-soluble substance. (a) SEM image of the substance existed on SiO_2 surface and underneath MoS_2 flakes. (b) EDS analysis of the substance elected in red rectangle area in (a).

Section S5. XPS data for the plasma etched MoS_2 film.

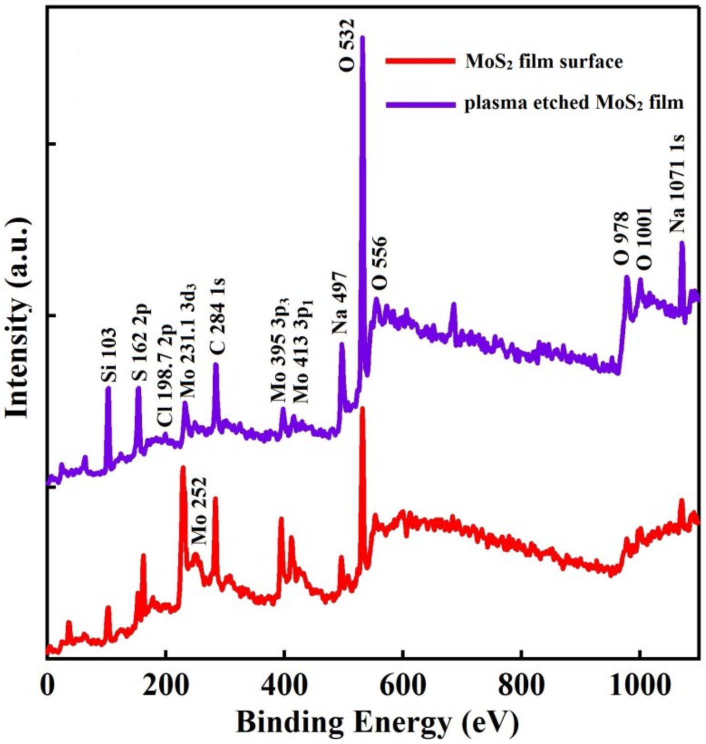

Fig. S5 XPS spectra of the as-grown and plasma etched MoS_2 film on SiO_2/Si substrate (plasma treated conditions: 300sccm Ar, 250sccm O_2, 250W, 12minutes).

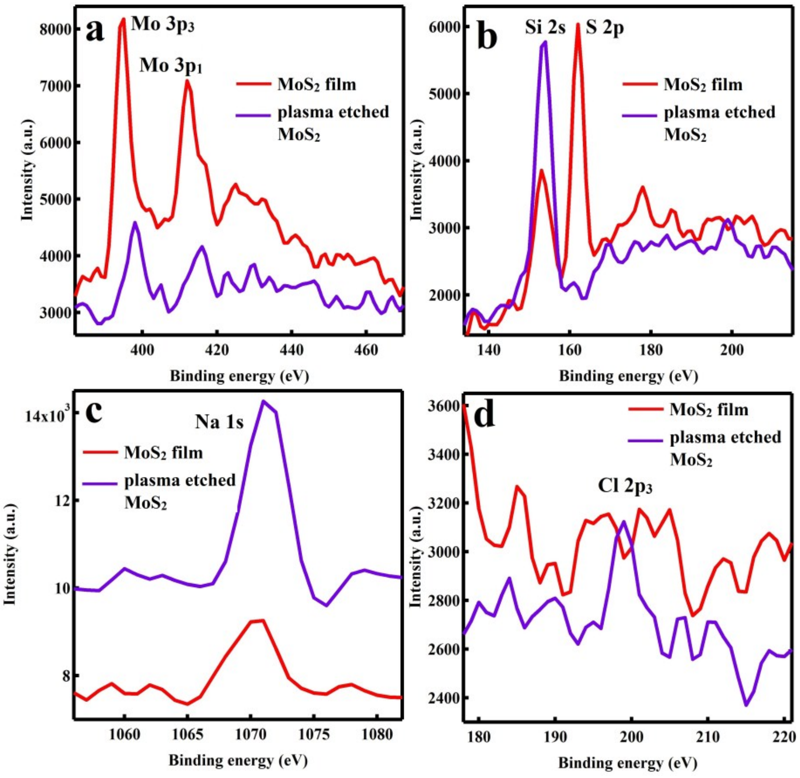

Fig. S6 XPS characterization of as-grown and plasma etched MoS_2 films. (a) Mo $3p_3$ and Mo $3p_1$, (b) S 2p, (c) Na 1s and (d) Cl $2p_3$ core-level XPS of as-grown and plasma etched MoS_2 films.

Plasma has been desmonstrated to be capable of etching MoS_2 layers.[1] We introduced plasma irradiation to etch a MoS_2 sheet and characterized the water-soluble substance underneath it. XPS measurement results clearly show that, after thinning MoS_2, the signalsof Na and Cl elements got enhanced while the one of Mo element got supressed, suggesting Na and Cl are indeed two major elemental compositions for the water-soluble layer.

Section S6. Controlled thickness and area of the sacrificial layer.

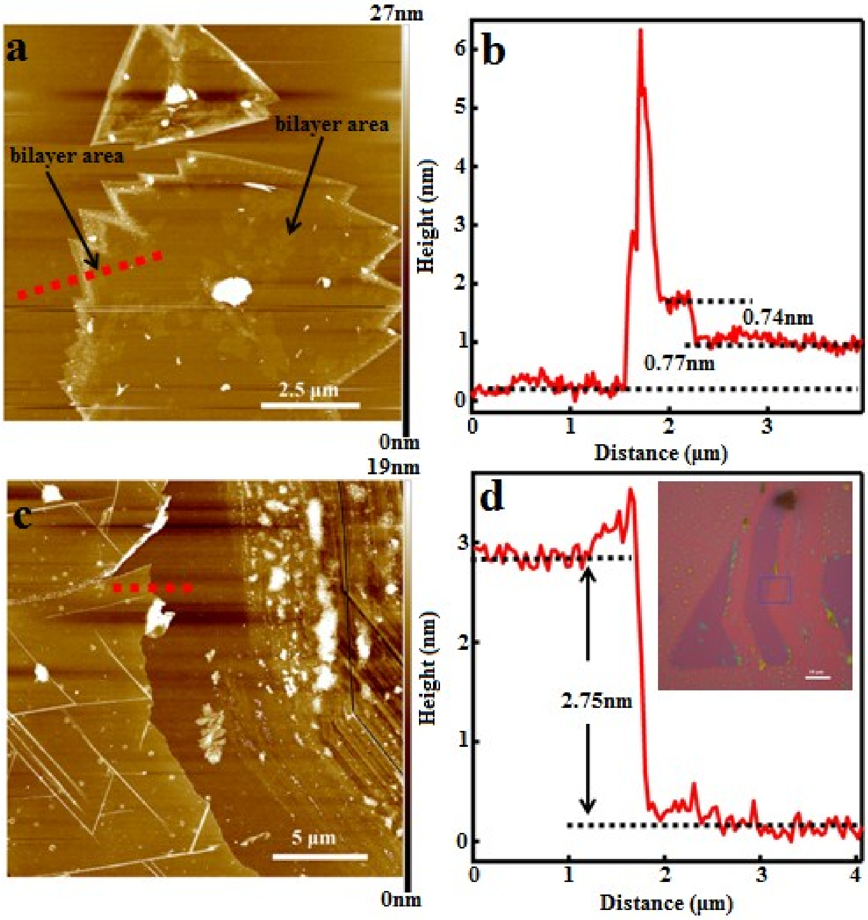

Fig. S7 AFM images of MoS_2 flakes synthesized at (a) 500°C and (c) 650°C. (b, d) The AFM cross-sectional profile along the red lines indicated in (a) and (c), respectively, and the blue square area in the inset in d is where the AFM characterization was conducted in c.

We found that 650°C is the optimized growth temperature and the thickness of the sacrificial layer is ~2nm based on our AFM characterizations. We measured the samples synthesized at 500°C and found the thickness of single layer MoS_2 flakes is ~0.75nm. This thickness was calculated from the height difference between the first layer flake and the growth substrate, as well as the height difference between the first and the second layer flakes as observed in Fig. S7b. We then scratched a MoS_2 flake synthesized at 650°C together with the sacrificial layer underneath it. The thickness is ~2.75nm as observed in Fig. S7d. This thickness minus the thickness of single layer MoS_2 flake (~0.75nm) is about 2nm, which is estimated to be the thickness of the sacrificial layer.

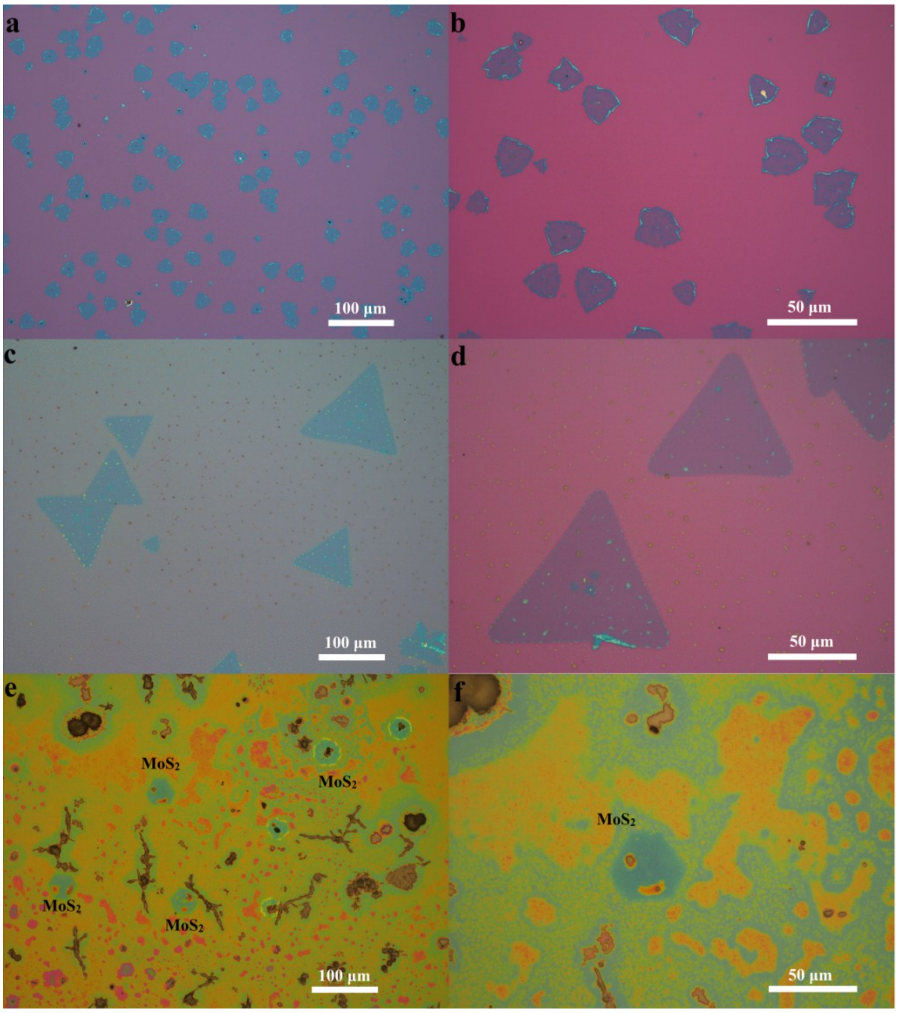

Fig. S8 Optical images of samples synthesized at (a, b) 500°C, (c, d) 650°C and (e, f) 800°C.

As for the controllability for the thickness and area of the sacrificial layer, we found that it is closely related with the growth temperatures. At 500°C, no sacrificial layer was introduced as observed in Fig. S8a and Figs. S8a, S8b. At 650°C, the sacrificial layer with 2nm thickness appeared underneath the MoS_2 flakes as shown in Figs. S7c, S7d and Figs. S8c, S8d. At 800°C, thick water-soluble films and many irregular particles or aggregates were grown on the substrate surface as observed in Figs.S8e and S8f. Thus, generally speaking, the thickness and the area of the sacrificial layer increase gradually with increasing growth temperatures, while the optimized growth temperature for the synthesis of MoS_2 flakes was found to be around 650°C.

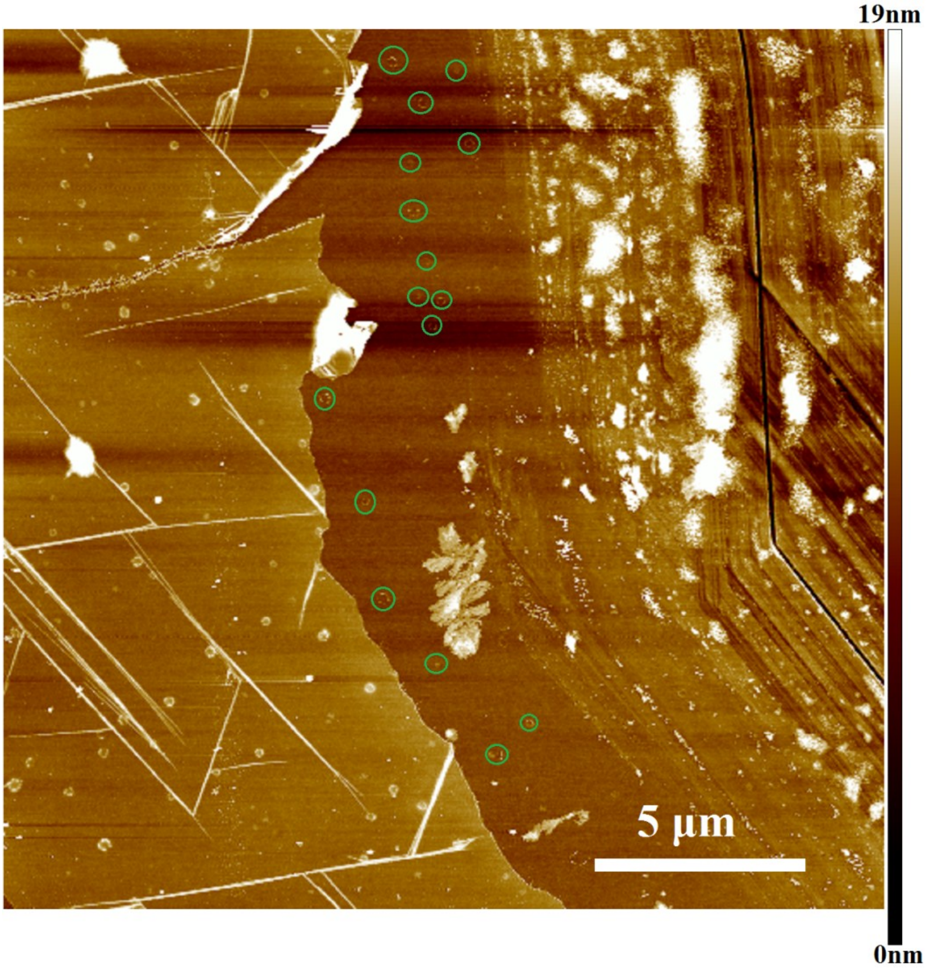

Fig. S9 AFM images of a scratched MoS_2 flake.

The sacrificial layer underneath the MoS_2 flakes is very difficult to characterize directly. However, according to our AFM characterizations on the MoS_2 flakes on top of the sacrificial layers, we found the thickness is quite uniform as displayed in Fig. S7c and S7d, indicating that the sacrificial layer should be quasi-continuous. The dark islands in Fig. 3a in the manuscript correspond to the scratched residues. We performed AFM characterizations on the scratched area, with a typical image shown in Fig. S9. We indeed observed some residues left after scratch as highlighted in green circles in Fig. S9, which could be related to the nucleation sites with stronger adhesion to the substrate.

Section S7. Log scale transfer curves of a transferred MoS_2 flake.

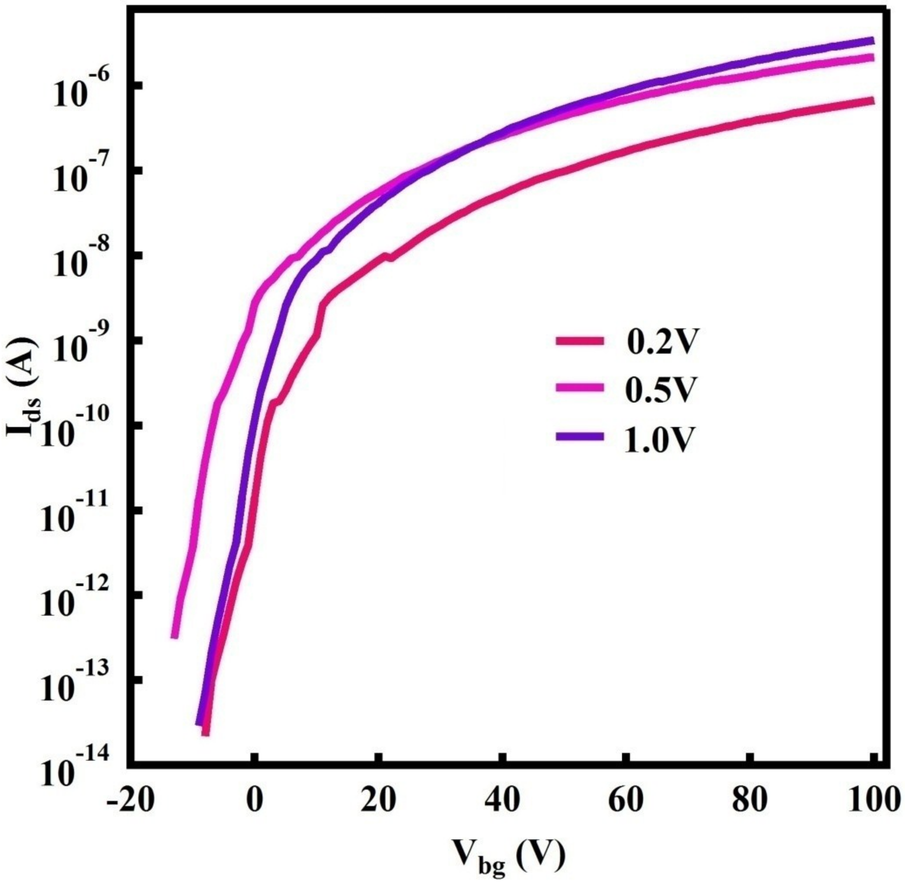

Fig. S10 Transfer curves of a transferred MoS_2 flake in log scale.

Section S8. Synthesis and transfer of $MoSe_2$ flakes.

Growth of $MoSe_2$ flakes: $MoSe_2$ flakes were synthesized in a two-temperature zone tube furnace. In a typical procedure, mixed NaCl and MoO_3 powders (30mg) with a weight ratio of 9:1 in a quartz boat were placed in the high-temperature zone. Another quartz boat holding 50mg pure selenium was placed in the upwind low-temperature zone. The distance between two quartz boats was fixed at 18cm. The fresh SiO_2/Si substrate was faced down above the quartz boat containing the mixed NaCl/MoO_3 powders. Before the experiments, high purity (99.99%) Ar gas was passed through the tube with a flow rate of 400 standard-state cubic centimeter per minute (sccm) for 10min to flush. The temperatures of two boats were then gradually increased from room temperature (RT) to target temperatures(650°C for mixed NaCl and MoO_3 powders, and 240°C for sulfur powders) within 25min and kept for 10min with 200sccm Ar flow, followed by a cooling down process (cooled down to RT within 2 hours).

Transfer of MoSe$_2$ flakes: Identical to main text methods, Transfer of MoS$_2$ flakes.

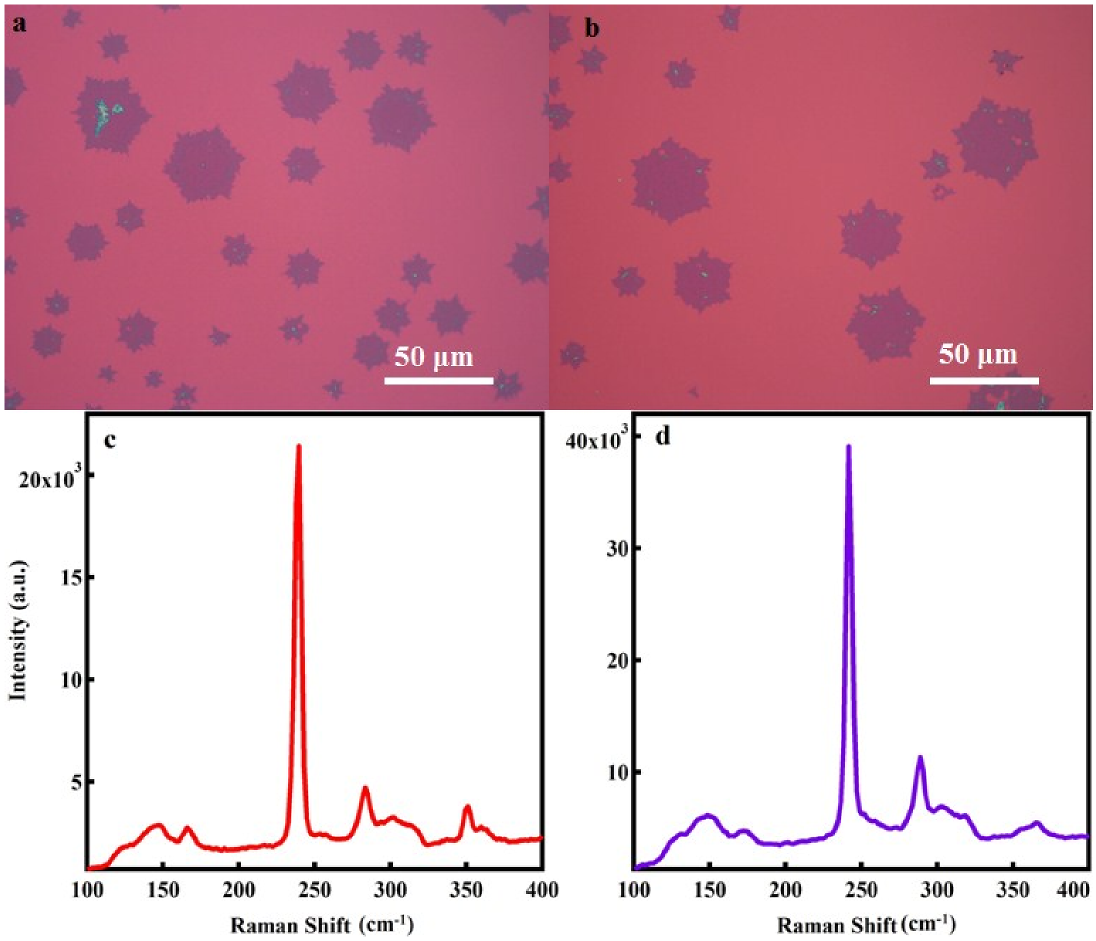

Fig. S11 Optical images of (a) as-grown and (b) transferred MoSe$_2$ flakes. Raman spectra of (c) as-grown and (d) transferred MoSe$_2$ flakes.

Fig. S11c and d show the Raman spectra of as-grown and transferred monolayer MoSe$_2$ flakes, and the Raman modes A_{1g} and E_{2g}^{1} of MoSe$_2$ were indentified at 241.7 and 288.8 cm^{-1}, respectively.[3, 4]

Section S9. Growth and transfer of MoS$_2$ flakes using different halide salts.

Growth of MoS$_2$ flakes using NaBr/MoO$_3$ as precursors: Monolayer MoS$_2$ flakes were synthesized in a two-temperature zone tube furnace. In a typical procedure, mixed NaBr and MoO$_3$ powder (13mg) with a weight ratio of 9:1 in a quartz boat was placed in the high-temperature zone. Another quartz boat holding 115mg pure sulfur was placed in the upwind low-temperature zone. The distance between two quartz boats was fixed at 18cm. The fresh SiO_2/Si substrate was faced down above the quartz boat containing the mixed NaBr/MoO_3 powders. Before the experiments, high purity (99.99%) Ar gas was passed through the tube with a flow rate of 400sccm for 10min to flush. The temperatures of two boats were then gradually increased from RT to target temperatures (650^{o}C for mixed NaCl and MoO_3 powders, and 240^{o}C for sulfur powders) within 25min and kept for 10min with 200sccm Ar flow, followed by a cooling down process (cooled down to RT within 2 hours).

Transfer of MoS_2 flakes: Identical to main text methods, Transfer of MoS_2 flakes.

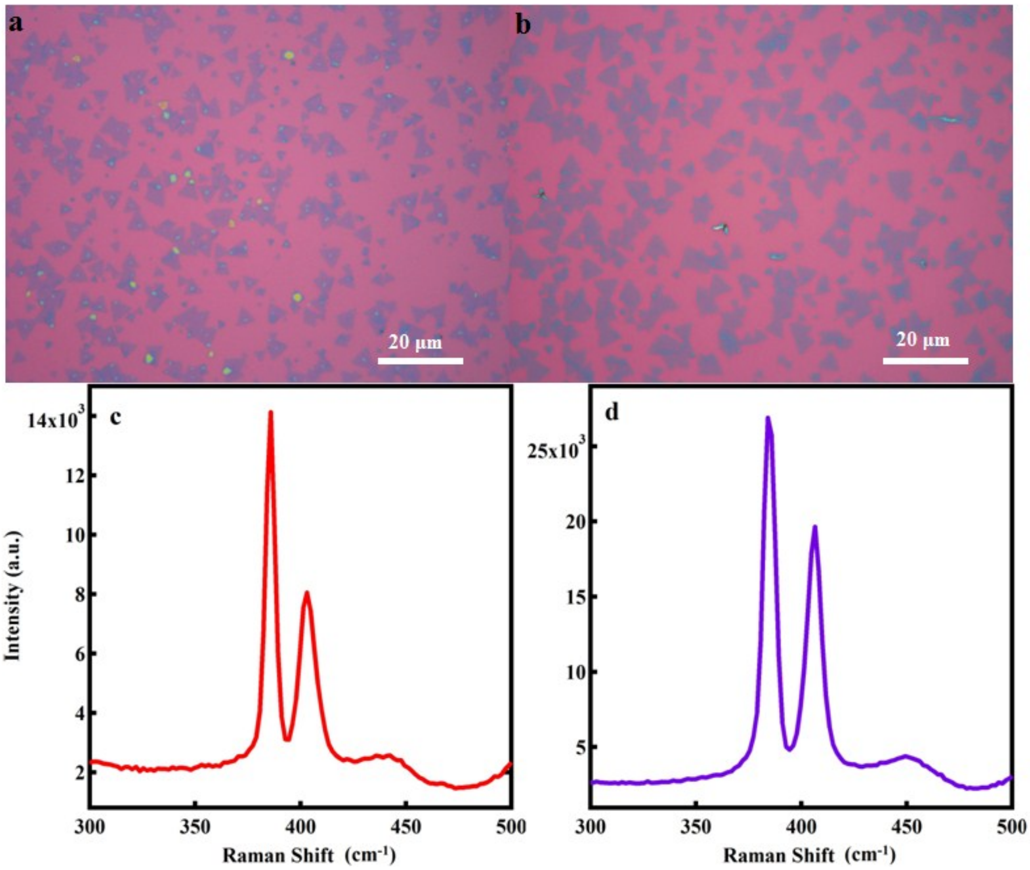

Fig. S12 Growth and transfer of MoS_2 flakes using NaBr/MoO_3 as precursors. Optical images of (a) as-grown and (b) transferred MoS_2 flakes. Raman spectra of (c) as-grown and (d) transferred MoS_2 flakes.

Growth of MoS_2 flakes using NaI/MoO_3 as precursors: Monolayer MoS_2 flakes were synthesized in a two-temperature zone tube furnace. In a typical procedure, mixed NaI and MoO_3 powder (18mg) with a weight ratio of 19:1 in a quartz boat was placed in the high-temperature zone. Another quartz boat holding 80mg pure sulfur was placed in the upwind low-temperature zone. The distance between two quartz boatswas fixed at 18cm. The fresh SiO_2/Si substrate was faced down above the quartz boat containing the mixed NaI/MoO_3 powders. Before the experiments, high purity

(99.99%) Ar gas was passed through the tube with a flow rate of 400 sccm for 10min to flush. The temperatures of two boats were then gradually increased from room temperature to target temperatures (600^{o}C for mixed NaI and MoO_3 powders, and 240^{o}C for sulfur powders) within 23min and kept for 10min with 200sccm Ar flow, followed byacooling down process (cooled down to RT within 2 hours).

Transfer of MoS_2 flakes: Identical to main text methods, Transfer of MoS_2 flakes.

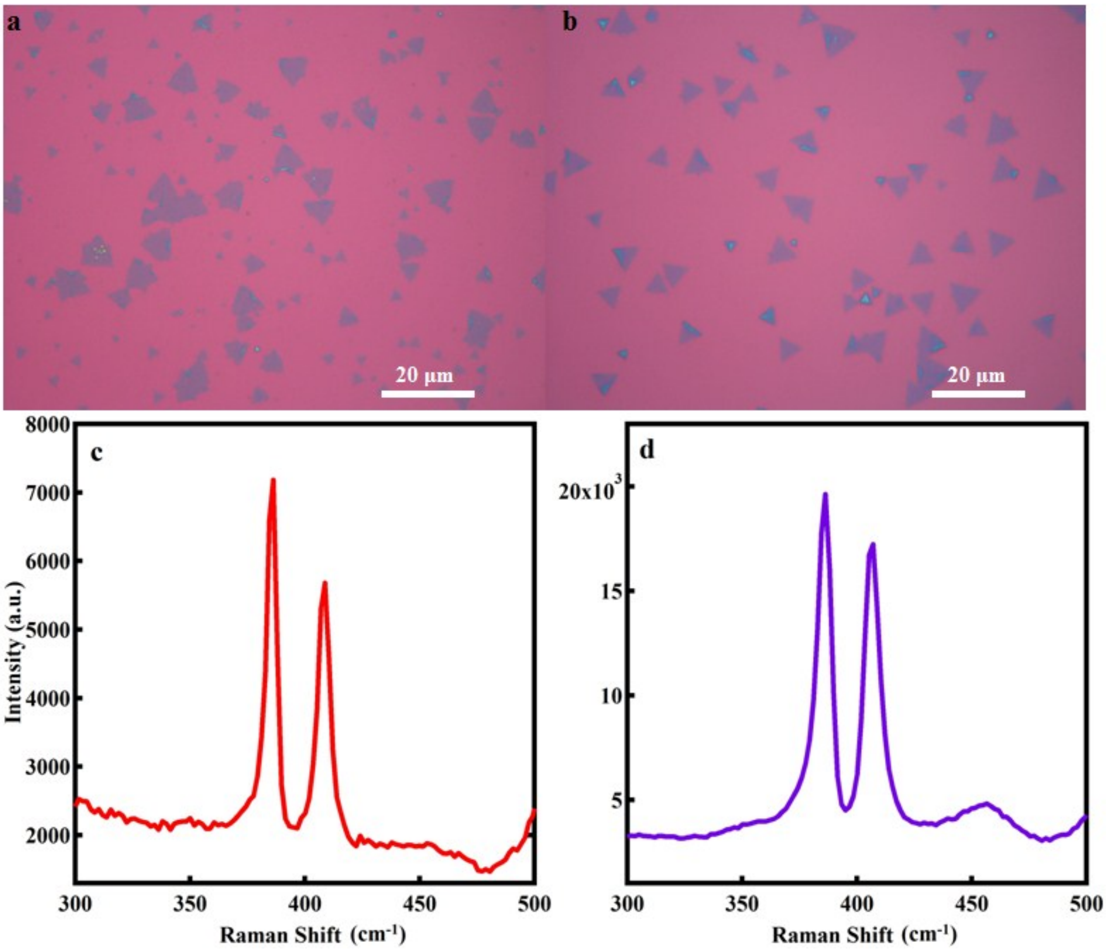

Fig. S13 Growth and transfer of MoS_2 flakes using NaI/MoO_3 as precursors. Optical images of (a) as-grown and (b) transferred MoS_2 flakes. Raman spectra of (c) as-grown and (d) transferred MoS_2 flakes.

Growth of MoS_2 flakes using KCl/MoO_3 as precursors: Monolayer MoS_2 flakes were synthesized in a two-temperature zone tube furnace. In a typical procedure, mixed KCl and MoO_3 powder (16mg) with a weight ratio of 19:1 in a quartz boat was placed in the high-temperature zone. Another quartz boat holding 150mg pure sulfur was placed in the upwind low-temperature zone. The distance between two quartz boatswas fixed at 18cm. The fresh SiO_2/Si substrate was faced down above the quartz boat containing the mixed KCl/MoO_3 powders. Before the experiments, high purity

(99.99%) Ar gas was passed through the tube with a flow rate of 400 sccm for 10min to flush. The temperatures of two boats were then gradually increased from RT to target temperatures (650^{o}C for mixed KCl and MoO_3 powders, and 240^{o}C for sulfur powders) within 25min and kept for 10min with 200sccm Ar flow, followed by a cooling down process (cooled down to RT within 2 hours).

Transfer of MoS_2 flakes: Identical to main text methods, Transfer of MoS_2 flakes.

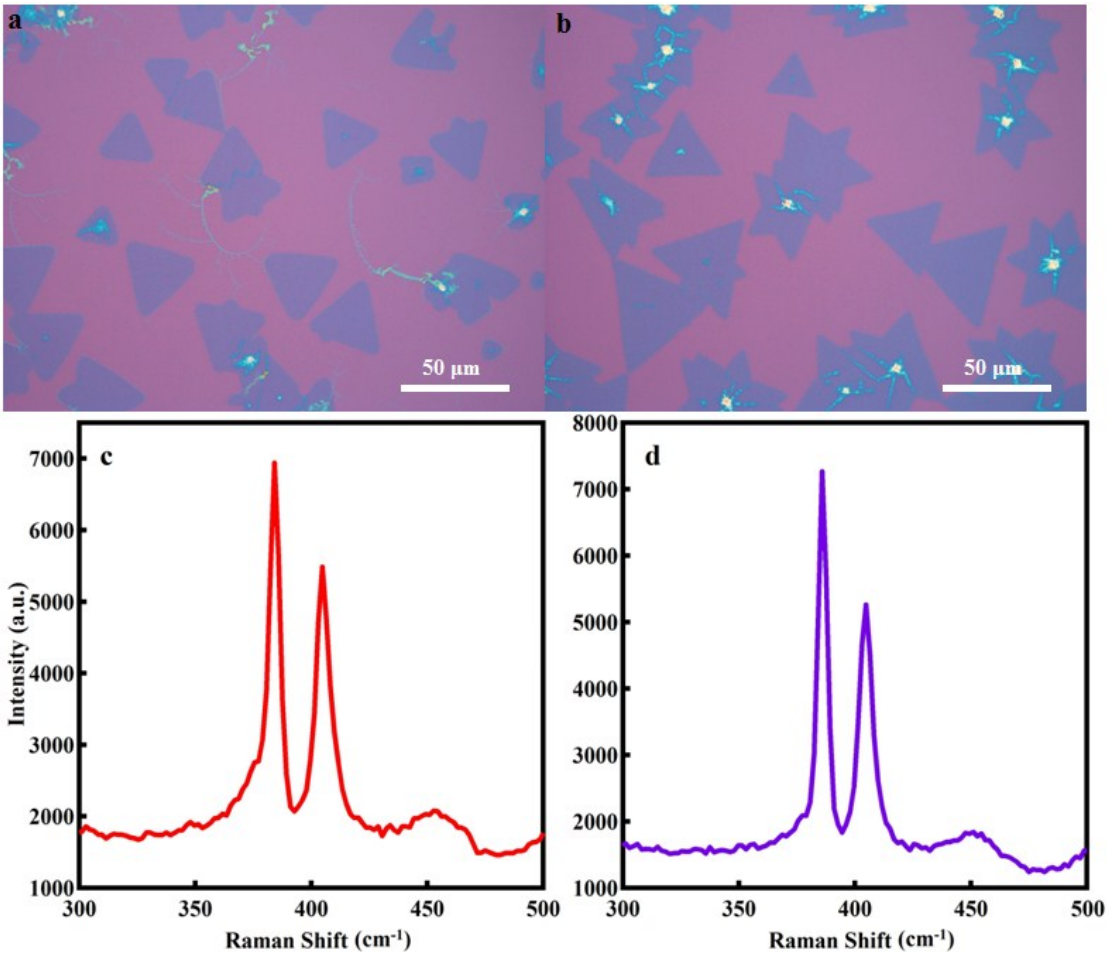

Fig. S14 Growth and transfer of MoS_2 flakes using KCl/MoO_3 as precursors. Optical images of (a) as-grown and (b) transferred MoS_2 flakes. Raman spectra of (c) as-grown and (d) transferred MoS_2 flakes.

Growth of MoS_2 flakes using KI/MoO_3 as precursors: Monolayer MoS_2 flakes were synthesized in a two-temperature zone tube furnace. In a typical procedure, mixed KI and MoO_3 powder (17mg) with a weight ratio of 19:1 in a quartz boat was placed in the high-temperature zone. Another quartz boat holding 100mg pure sulfur was placed in the upwind low-temperature zone. The distance between two quartz boatswas fixed at 18cm. The fresh SiO_2/Si substrate was faced down above the quartz boat containing the mixed KI/MoO_3 powders. Before the experiments, high purity

(99.99%) Ar gas was passed through the tube with a flow rate of 400 sccmfor 10min to flush. The temperatures of two boatswere then gradually increased from RT to target temperatures (650°C for mixed KI and MoO_3 powders, and 240°C for sulfur powders) within 25min and kept for 10min with 200sccm Ar flow, followed by a cooling down process (cooled down to RT within 2 hours).

Transfer of MoS_2 flakes: Identical to main text methods, Transfer of MoS_2 flakes.

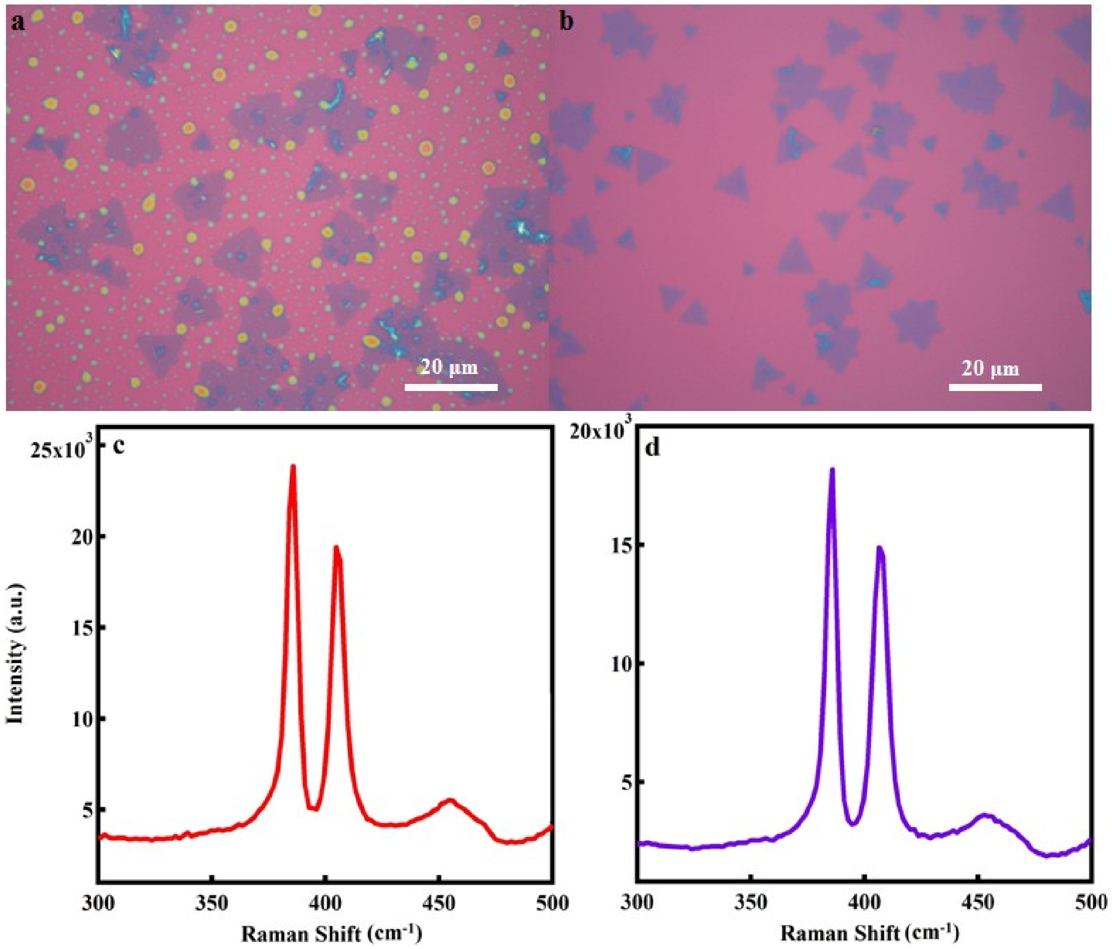

Fig. S15 Growth and transfer of MoS_2 flakes using KI/MoO_3 as precursors. Optical images of (a) as-grown and (b) transferred MoS_2 flakes. Raman spectra of (c) as-grown and (d) transferred MoS_2 flakes.

Section S10. Growth of MoS_2 flakes on sapphire and glass substrates.

Growth of MoS_2 flakes on sapphire substrate: Monolayer MoS_2 flakes were synthesized in a two-temperature zone tube furnace. In a typical procedure, mixed NaCl and MoO_3 powder (13mg) with a weight ratio of 9:1 in a quartz boat was placed in the high-temperature zone. Another quartz boat holding 90mg pure sulfur was placed in the upwind low-temperature zone. The distance between two quartz boatswas fixed at 18cm. The fresh SiO_2/Si substrate was faced down above the quartz boat containing the mixed KI/MoO_3 powders. Before the experiments, high purity (99.99%) Ar gas was passed through the tube with a flow rate of 400 sccm for 10min to flush. The temperatures of two boats were then gradually increased from RT to target temperatures (700°C for mixed NaCl and MoO_3 powders, and 240°C for sulfur powders) within 27min and kept for 10min with 200sccm Ar flow, followed by a cooling down process(cooled down to RT within 2 hours).

Transfer of MoS_2 flakes: Identical to main text methods, Transfer of MoS_2 flakes.

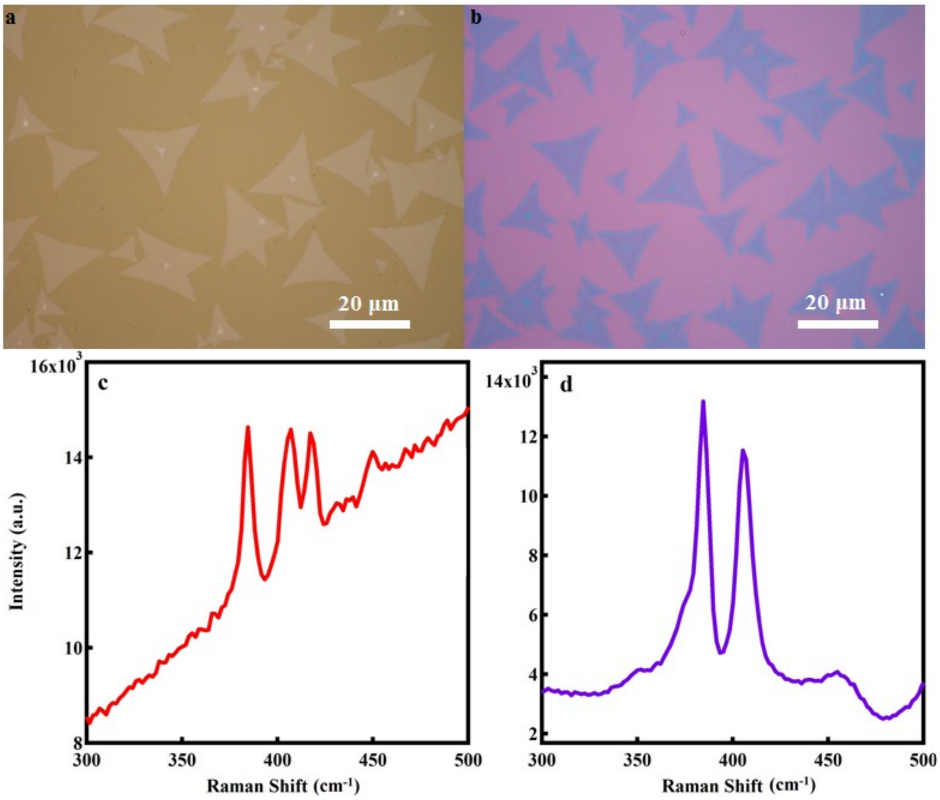

Fig. S16 Growth of MoS_2 flakes on sapphire substrate. Optical images of (a) as-grown MoS_2 flakes on sapphire substrate and (b) transferred MoS_2 flakes on SiO_2/Si substrate. Raman spectra of (c) as-grown and (d) transferred MoS_2 flakes.

Raman modes at 417cm^{-1} and 449.8cm^{-1} belong to sapphire (α-Al_2O_3) substrate in Fig. S16c.[5]

Growth of MoS_2 flakes on glass substrate: MoS_2 flakes were synthesized in a two-temperature zone tube furnace. In a typical procedure, mixed NaCl and MoO_3 powder (15mg) with a weight ratio of 9:1 in a quartz boat was placed in the high-temperature zone. Another quartz boat holding 100mg pure sulfur was placed in the upwind low-temperature zone. The distance between two quartz boats was fixed at 18cm. The fresh SiO_2/Si substrate was faced down above the quartz boat containing the mixed NaCl/MoO_3 powders. Before the experiments, high purity (99.99%) Ar gas was passed through the tube with a flow rate of 400 sccmfor 10min to flush. The temperatures of two boats were then gradually increased from RT to target temperatures (650^{o}C for mixed NaCl and MoO_3 powders, and 240^{o}C for sulfur powders) within 25min and kept for 10min with 200sccm Ar flow, followed by a cooling down process (cooled down to RTwithin 2 hours).

Transfer of MoS_2 flakes: Identical to main text methods, Transfer of MoS_2 flakes.

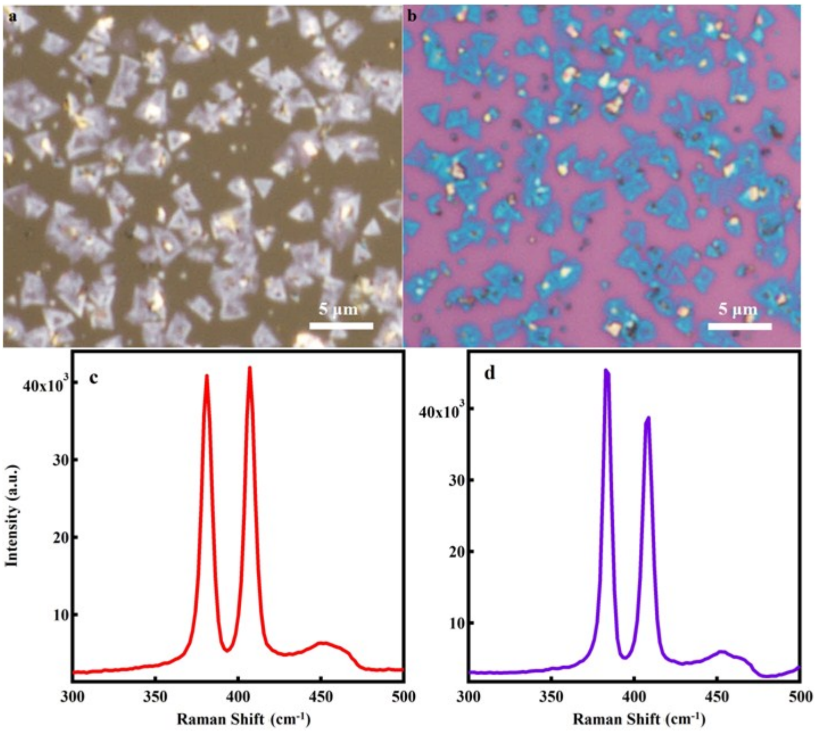

Fig. S17 Growth of MoS_2 flakes on glass substrate. Optical images of (a) as-grown MoS_2 flakes on glass substrate and (b) transferred MoS_2 flakes on SiO_2/Si substrate. Raman spectra of (c) as-grown and d) transferred MoS_2 flakes.

References:

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2. J. C. Shaw, H. Zhou, Y. Chen, N. O. Weiss, Y. Liu, Y. Huangand X. Duan, *Nano Res*, 2014, **7**, 511-517.
3. P. Tonndorf, R. Schmidt, P. Böttger, X. Zhang, J. Börner, A. Liebig, M. Albrecht, C. Kloc, O. Gordan. and D. R. Zahn, *Opt. Express* 2013, **21**, 4908-4916.
4. M. Ashkin, J. H. Parker and D. W. Feldman, *Solid State Commun*, 1968, **6**, 343-346.