

Supplementary information: Raman fingerprint of interlayer coupling in 2D TMDCs

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1 Sample preparation

1.1 WSe₂/WSe₂ homo-bilayer

WSe₂/WSe₂ homo-bilayer is prepared by a combination of a deterministic all-dry transfer technique and tear-and-stack method [castellanos2014deterministic, Pan2022signature]. As shown in Figure. ??, a large monolayer WSe₂ is mechanically exfoliated on PDMS. The monolayer is then partially transferred on a 300 nm SiO₂/Si substrate. The substrate is mounted on a rotation stage and twisted by 60°. The remaining WSe₂ monolayer is aligned and transferred. This will lead to the formation of an artificially-stacked 2H phase bilayer WSe₂.

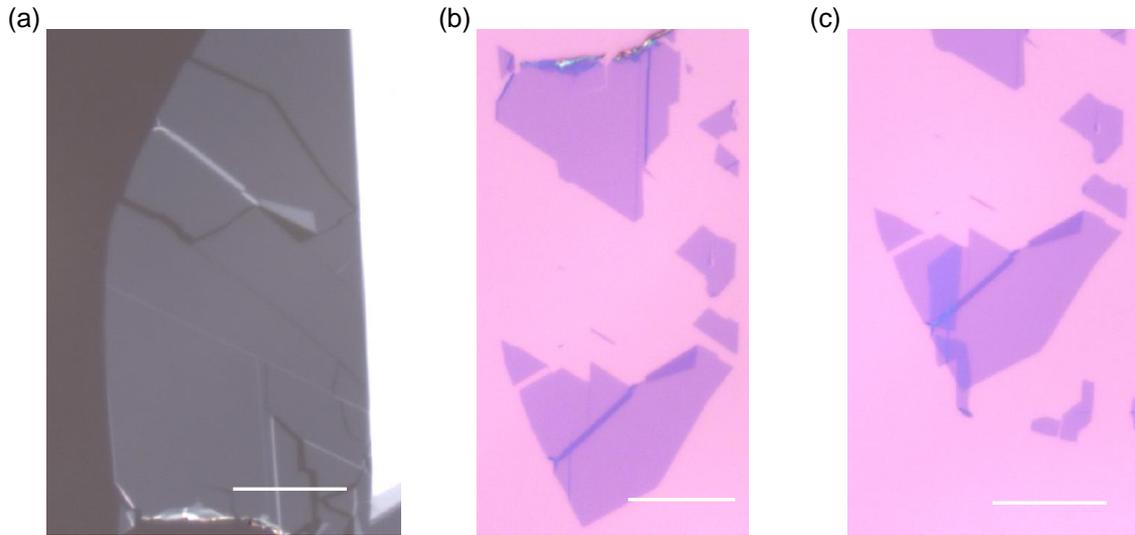


Figure S1: Sample preparation of WSe₂/WSe₂ homo-bilayer. (a) Monolayer WSe₂ on PDMS. (b) Partially transferred monolayer WSe₂ on 300 nm SiO₂/Si. (c) Second transfer with a 60° twisting angle. The scale bar is 15 μm.

1.2 MoSe₂/WSe₂ hetero-bilayer

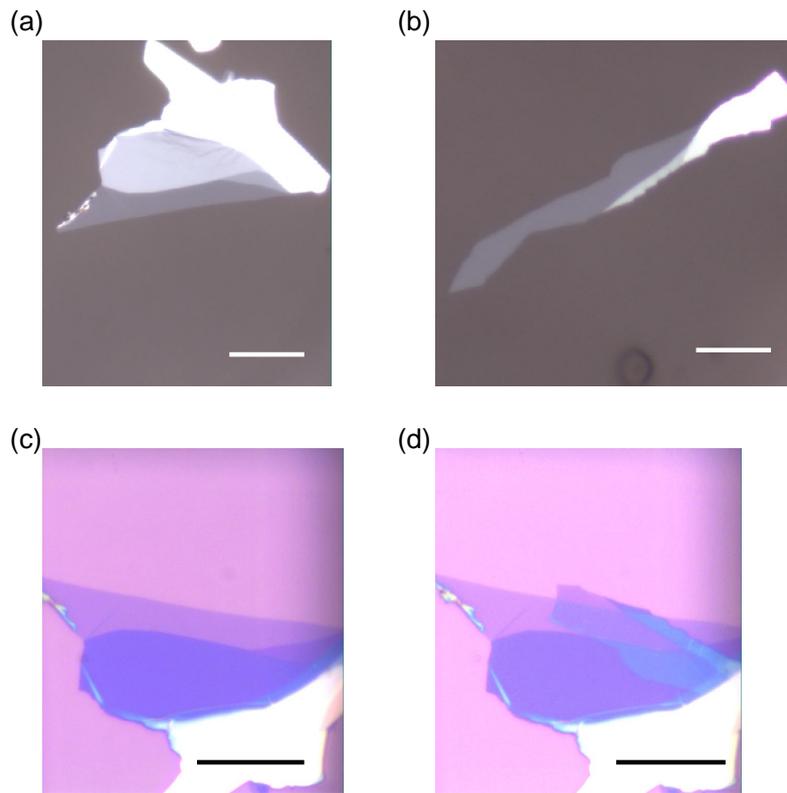


Figure S2: Sample preparation of MoSe₂/WSe₂ hetero-bilayer. Monolayer (a) WSe₂ and (b) MoSe₂ on PDMS. (c) Monolayer WSe₂ transferred on 300 nm SiO₂/Si. (d) Monolayer MoSe₂ transferred on WSe₂. The scale bar is 15 μm.

2 WSe₂/WSe₂ homo-bilayer before and after annealing

The squared area rolled up along the wrinkle after annealing.

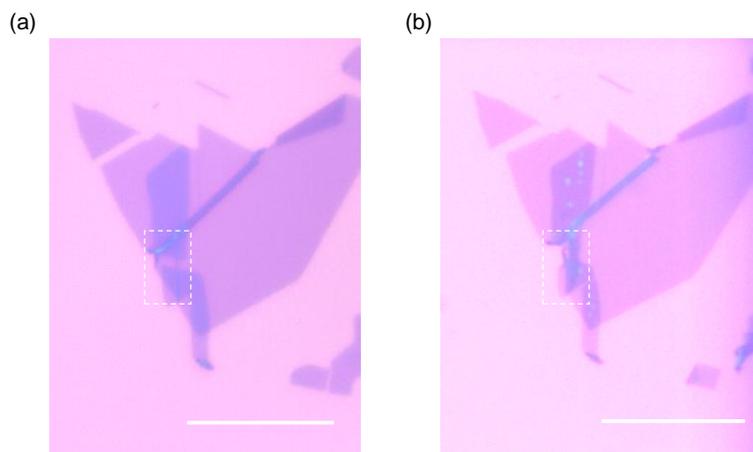


Figure S3: WSe₂/WSe₂ homo-bilayer (a) before and (b) after annealing. The scale bar is 15 μm.

3 Raman and PL map of MoSe₂/WSe₂ hetero-bilayer

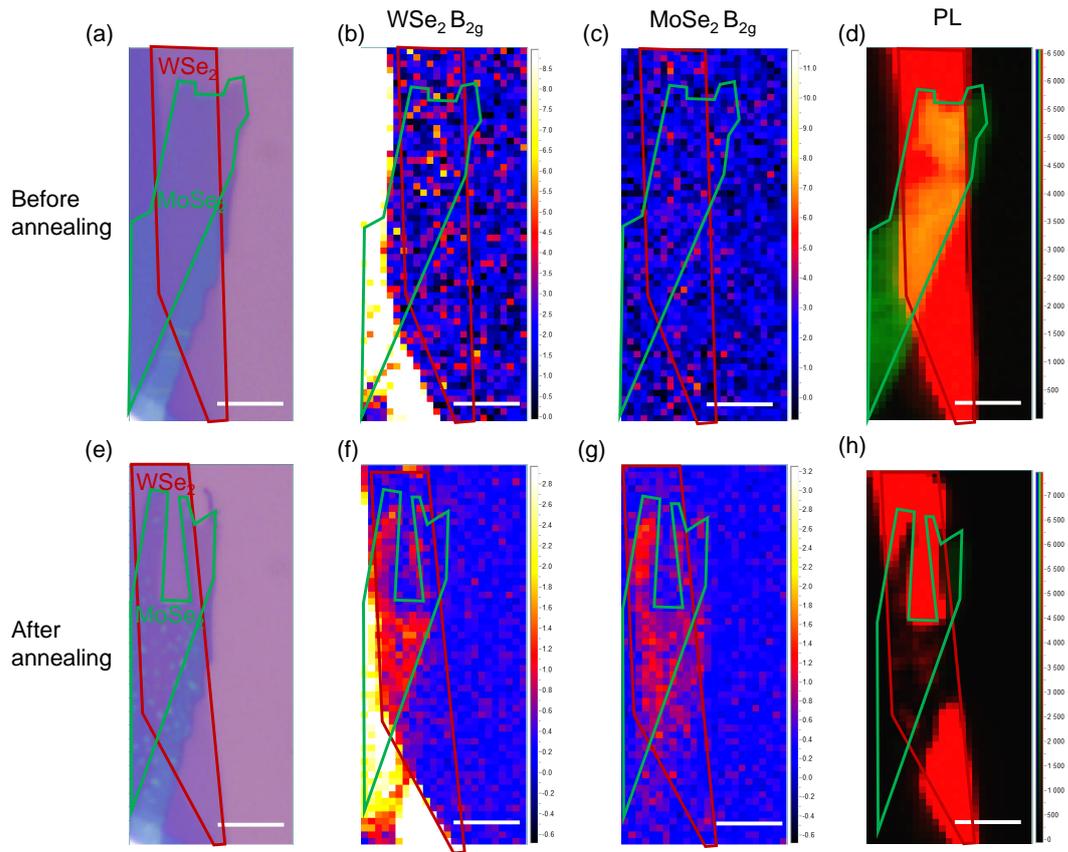


Figure S4: (a) Optical microscope image, (b) WSe₂ B_{2g} Raman intensity map, (c) MoSe₂ B_{2g} Raman intensity map, and (d) PL map before annealing. (e) Optical microscope image, (f) WSe₂ B_{2g} Raman intensity map, (g) MoSe₂ B_{2g} Raman intensity map, and (h) PL intensity map after annealing. The scale bar is 5 μm.