

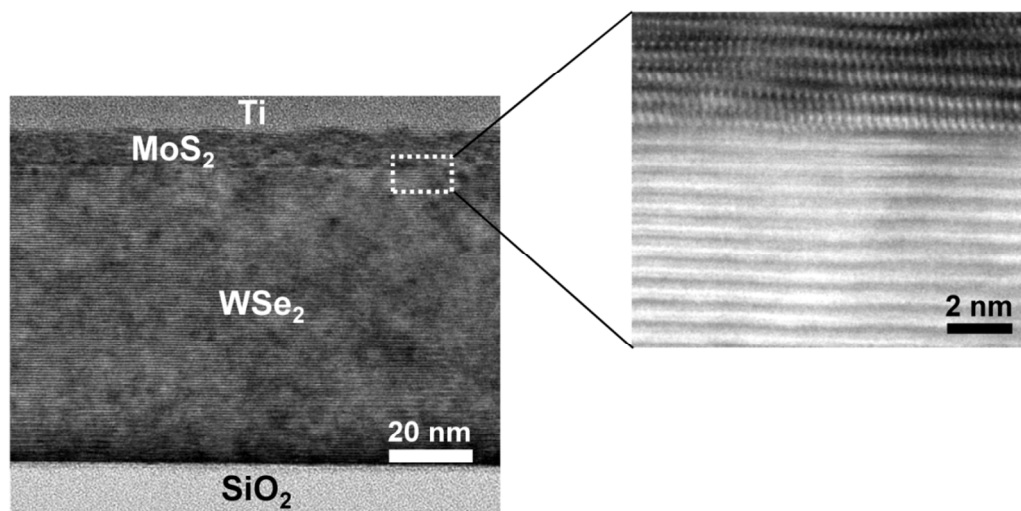
## SUPPLEMENTARY INFORMATION

# Charge Transport in MoS<sub>2</sub>/WSe<sub>2</sub> van der Waals Heterostructure with Tunable Inversion Layer

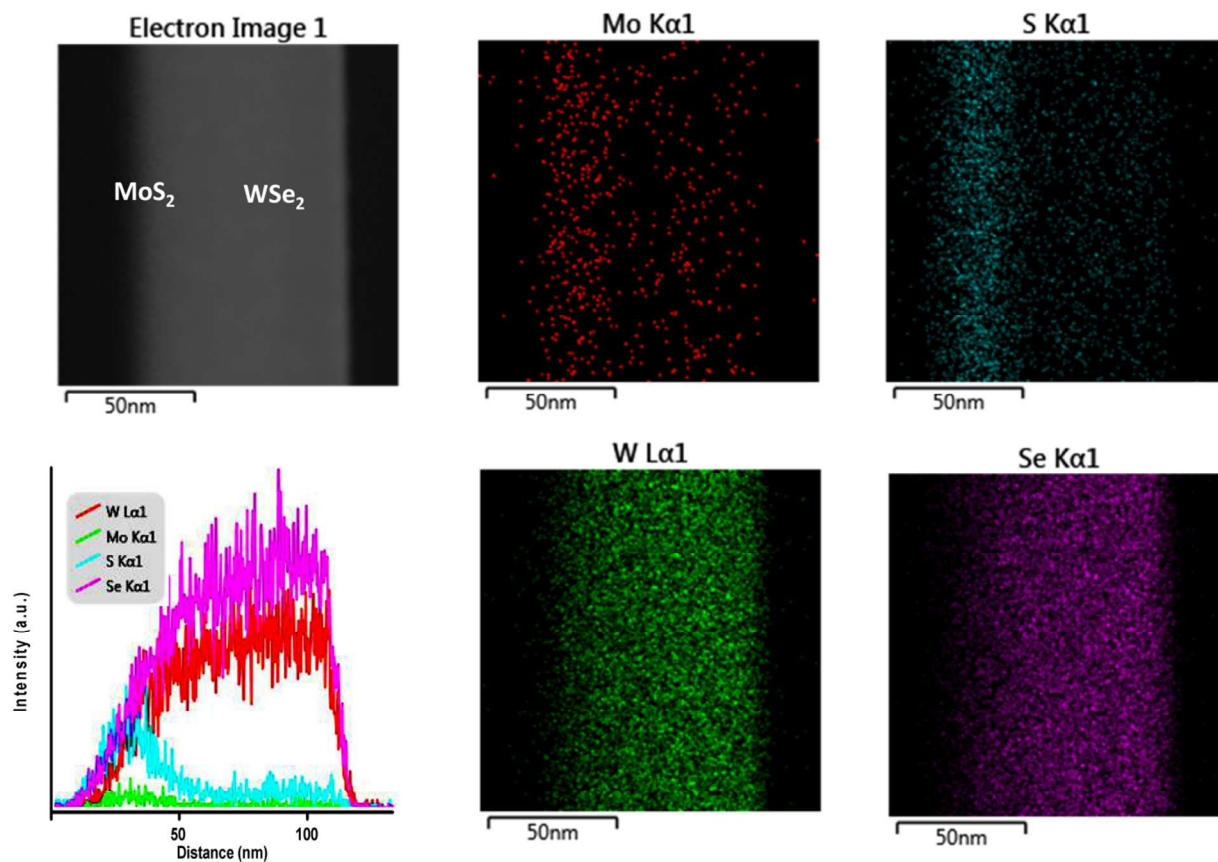
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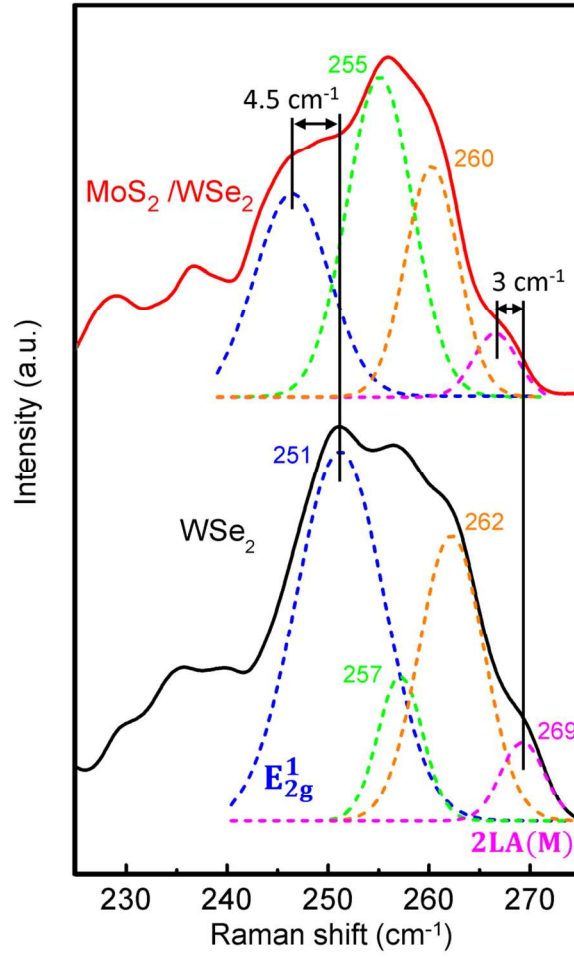
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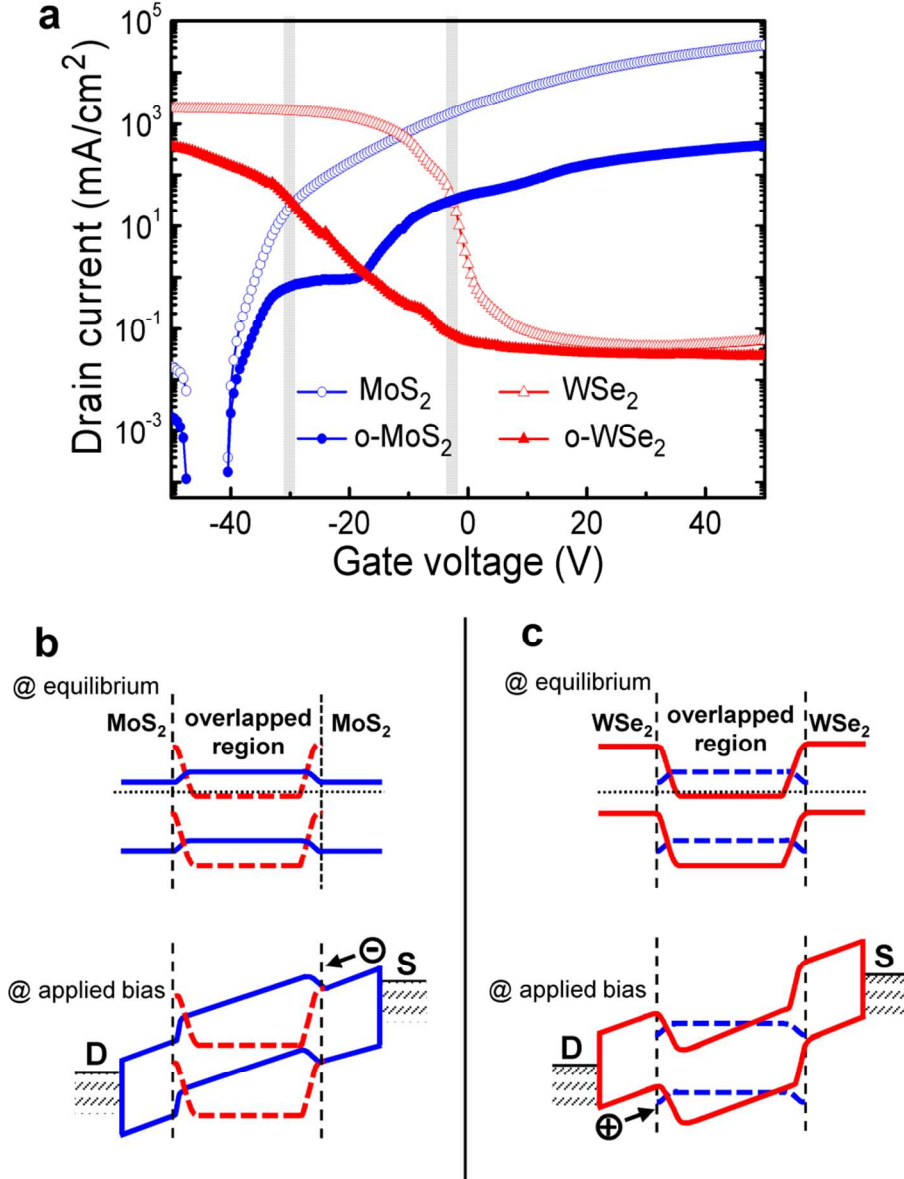
**Figure S1.** Cross-sectional TEM images of the MoS<sub>2</sub>/WSe<sub>2</sub> interface in small and large magnification observation (left and right panels, respectively). The atomically contacted interface is visible as show in the left image.



**Figure S2.** Energy dispersive spectroscopy (EDS) mappings of the  $\text{MoS}_2/\text{WSe}_2$  interface after stacking and annealing. The buried interface was formed, which is similar with the observation in graphene/h-BN heterostructure fabricated by dry transfer and thermal annealing.<sup>1</sup>



**Figure S3.** Raman peaks of WSe<sub>2</sub> at the positions with and without MoS<sub>2</sub> stacking. A broad peak appeared in the both positions. The peak of pristine WSe<sub>2</sub> was deconvoluted into three peaks; E<sub>2g</sub><sup>1</sup> near 251 cm<sup>-1</sup>, A<sub>1g</sub> near 260 cm<sup>-1</sup>, and 2LA(M) near 269 cm<sup>-1</sup>.<sup>2,3</sup> The split of A<sub>1g</sub> into two peaks of 257 and 262 cm<sup>-1</sup> is ascribed to the contribution from SiO<sub>2</sub> substrate where p-doped WSe<sub>2</sub> is further blueshifted by losing charges to the substrate (brown color), in other words, phonon stiffening.<sup>4</sup> The E<sub>2g</sub><sup>1</sup> peak at the overlapped region is redshifted by 4.5 cm<sup>-1</sup>, indicating a tensile strain.<sup>4</sup> The intensity is reduced by the strain and interlayer coupling with MoS<sub>2</sub>. The redshift of the A<sub>1g</sub> peak indicates phonon softening by dedoped WSe<sub>2</sub> (charge compensation) due to electrons transferred from MoS<sub>2</sub>, which is consistent with the Raman peak shift of MoS<sub>2</sub> discussed in the main text. The 2LA(M) peak position is also redshifted by the phonon softening without altering intensity. The SiO<sub>2</sub> related peak near 262 cm<sup>-1</sup> is also redshifted, indicating that the charges transferred from MoS<sub>2</sub> is widely spread to the substrate.



**Figure S4. Interlayer tunneling at the overlapped MoS<sub>2</sub> and WSe<sub>2</sub> channels.** (a) Transfer curves of the pristine and overlapped channels. The on-currents were reduced, the threshold voltages were shifted, and the additional shoulders appeared in the overlapped channels due to interfacial charge transfer and interlayer tunneling. (b, c) Band diagrams of the overlapped MoS<sub>2</sub> and WSe<sub>2</sub> channels under equilibrium and an applied source-drain bias. The shoulders observed in the transfer curves correspond to the gate voltages in which the conduction (valence) band of MoS<sub>2</sub> (WSe<sub>2</sub>) is aligned with the one of WSe<sub>2</sub> (MoS<sub>2</sub>) as marked by the two broad lines in (a).

## Supplementary references

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