Lateral epitaxial growth of two-dimensional layered semiconductor heterojunctions

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1. Methods

The synthesis of WS$_2$-WSe$_2$ lateral heterostructures. The synthesis of WS$_2$-WSe$_2$ lateral heterostructures was carried in a home built CVD system that can allow in situ switch of solid chemical sources (Supplementary Fig. 1). Two quartz boats with 0.4 g WS$_2$ powder (Alfa Aesar, 99.8%) or 0.6 g WSe$_2$ powder (Alfa Aesar, 99.8%) were loaded into a horizontal tube furnace with 1-inch quartz tube. A clean Si/SiO$_2$ substrate ($\sim$1×5 cm) was placed at downstream end of the furnace as the growth substrate. The system was pump-purged with ultra-high purity argon (Ar) gas (Rizhen, $\sim$99.999%) then ramped to the desired growth temperature for the growth of WS$_2$ nanosheets at 1057 $^\circ$C for 20 minutes under ambient pressure and a constant flow of 70 sccm Ar as the carrier gas. The furnace was then cooled down to 800 $^\circ$C, and the boat with WS$_2$ powder was pushed out of the hot zone without breaking the vacuum, while the boat with WSe$_2$ powder was simultaneously pushed into the hot zone of the tube in situ. The temperature was then ramped to 1190 $^\circ$C for the lateral epitaxial growth of WSe$_2$ for about 20 minutes under 5000 sccm Ar flow. Then the growth was terminated by shutting off the power of the furnace, and the sample was naturally cooled down to ambient temperature under 50 sccm Ar flow.

The synthesis of MoS$_2$-MoSe$_2$ lateral heterostructures. The reaction was carried out in a 12-inch horizontal tube furnace in a 1-inch quartz tube using a home-built CVD system (Supplementary Fig. 2). 0.7 g MoO$_3$ powder (Sigma Aldrich, 99%) was placed in a 5 mL alumina boat and placed in the center of the quartz tube in the furnace. Four pieces of Si/SiO$_2$(300 nm) substrate was cut into $\sim$1.5× 1.5 cm squares and placed on top of the alumina boat containing MoO$_3$ precursor powder with the polished-side facing downwards. Two boats were placed upstream containing 0.4 g of S powder and...
0.6 g of Se powder. The boat containing S powder is closer to the MoO₃ powder than the Se. The quartz tube was then sealed and pump-purged several times with 100 sccm Ar flow, and brought back to atmospheric pressure. The furnace was then ramped to 750 °C at a rate of 25 °C/min. The sulfur in the first boat was introduced into the heated furnace by mechanically moving the quartz tube into the furnace just enough so that the selenium is left outside of the heating zone. After S was completely evaporated in about 5 minutes, the furnace was cooled to 700 °C and an additional 5 sccm of H₂ was introduced, which was previously found to be the most ideal for MoSe₂ growth. The quartz tube was further shifted to move Se boat into the heated zone to provide Se vapour for lateral epitaxial growth of MoSe₂. The reaction proceeded for 15 minutes until the furnace was turned off and naturally cooled to room temperature.

**Microscopic, optical, electrical and optoelectrical characterizations.** The microstructures and morphologies of the heterostructures are characterized by an optical microscope, JEOL 6700 scanning electron microscope (SEM), and an FEI Titan transmission electron microscope (TEM). The confocal micro-PL and micro-Raman measurements were conducted using a Horiba LabRAM HR Evolution confocal Raman system with 600 g/mm and 1800 g/mm grating, 50× diffraction-limited objective (N.A.=0.75), with an Ar ion laser (514 nm) excitation. To fabricate WS₂-WSe₂ heterojunction device, the metal electrodes (for probe contact or wire bonding purposes) were patterned using electron-beam lithography followed by electron-beam deposition of metal thin films. Ti/Au (5/50 nm) was used as the contact electrodes for WS₂ and Au (50 nm) for WSe₂. The D.C. electrical transport measurements were conducted in a Lakeshore probe station (Model TTP4), with a computer-controlled analogue-to-digital converter (National Instruments model 6030E) and a low noise current pre-amplifier.
(Stanford Research Systems SR570). The scanning photocurrent measurements were conducted with the same Horiba LabRAM HR Evolution confocal Raman system combined with the same electrical measurement system.

2. Schematic illustration of the home-built CVD system for the growth of WS$_2$-WSe$_2$ lateral heterostructures.

Supplementary Fig. 1| Schematic illustration of the home-built CVD system for the growth of WS$_2$-WSe$_2$ lateral heterostructures. The WS$_2$ powder and WSe$_2$ powder was directly used as the solid source. Two magnets (one inside the CVD chamber and one outside) was used to mechanically push the WS$_2$ solid source out of and WSe$_2$ solid source into the centre hot zone without breaking vacuum. This *in situ* switch of chemical source is essential for retaining the active edge growth front for sequential epitaxial growth of WS$_2$-WSe$_2$ lateral heterostructures.
3. Schematic illustration of the home-built CVD system for the growth of MoS₂-MoSe₂ lateral heterostructures.

Supplementary Fig. 2 | Schematic illustration of the home-built CVD system for the growth of MoS₂-MoSe₂ lateral heterostructures. The MoO₃ powder and elemental S and Se powders were used as the solid source. The MoO₃ powder is kept in the centre hot zone, and the S and Se vapour was sequentially introduced into the CVD reaction by shifting quartz tube rightward to move the S and Se source into the furnace without breaking vacuum, which is essential for retaining the active edge growth front for continued epitaxial growth of MoS₂-MoSe₂ lateral heterostructures.

4. Histogram of domain thickness distribution.

Supplementary Fig. 3 | Histogram of domain thickness distribution. The nanosheet thickness is rather sensitive to the growth conditions (e.g. temperature, flow rate and position of the substrate). With the optimized condition for WS₂/WSe₂ system, ~43% of domain are single layers, 33% are bilayers and the rest are multilayers.
5. Transfer characteristics and carrier mobility of the synthetic WS$_2$ and WSe$_2$.

![Graphs showing transfer characteristics of WS$_2$ and WSe$_2$.](image)

**Supplementary Fig. 4** | Transfer characteristics and carrier mobility of the synthetic WS$_2$ and WSe$_2$.  

**a**, Transfer curve ($I_{sd}$-$V_{BG}$) of a back-gated WS$_2$ device, with which an electron mobility of 16 cm$^2$/Vs can be derived.  

**b**, Transfer curve ($I_{sd}$-$V_{BG}$) of a back-gated WSe$_2$ device, with which a hole mobility of 82 cm$^2$/Vs can be derived.