

Supplementary Information For Atomically-Thin Ohmic Edge Contacts Between Two-Dimensional Materials

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contacts

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1: Additional Information on the heterostructure growth

Graphene growth: Copper foil (Nilaco Corporation, #CU-113213, 99.9% purity) was placed in a quartz boat and annealed in a 1 inch quartz tube hot wall furnace for 4 hours under hydrogen flow of 137 sccm at 1040 °C. Then diluted methane (1%, balanced with hydrogen) was introduced to the furnace under the flow rate of 4.5 sccm for one hour to grow a continuous graphene film (partially grown graphene can be obtained by reducing the growth time). The furnace was cooled down to room temperature with hydrogen flowing after the growth was complete. Figure S1a shows an optical micrograph of a nearly continuous graphene film transferred to a Si/SiO₂ used to measure the typical graphene grain size of approximately 20 μm.

Graphene transfer: The as-grown graphene on the Cu substrate was spin coated with PMMA A4 at 3000 RPM for one minute followed by etching in Cu etchant (CE-200, Transene Company INC). The resulting film was sequentially rinsed in DI water for 15 minutes, 4% HCl for one minute, and DI water for one minute. The film was then transferred to a SiO₂/Si substrate and baked at 170 °C until it was completely dried. After that, the substrate was soaked in hot acetone (90 °C) to remove PMMA. Finally the substrate was annealed in an ultra-high vacuum (10⁻⁷ Torr) furnace at 350 °C for 5 hours to increase the adhesion between the graphene and the substrate and to further remove any polymer residues.

Graphene patterning: After being placed on the SiO₂/Si substrate, the graphene was patterned using photolithography and plasma etching. Briefly, positive photoresist S1805 (MicroChem) was spin coated on the substrate at 3000 RPM for one minute followed by baking at 115 °C for one minute. A contact aligner was used to expose the pattern using 365 nm light for 3 seconds. Then the pattern was developed using MIF 726 developer (MicroChem) for one minute and rinsed by isopropyl alcohol. The exposed graphene region was etched away using an oxygen plasma.

TMD growth: The substrate with patterned graphene was placed on a quartz plate and inserted into TMD growth furnace. The synthesis of monolayer TMD was carried out in a hot-wall quartz tube furnace with 4.3 inch inner diameter according to our previous report.¹ Molybdenum hexacarbonyl (Mo(CO)₆, MHC, Sigma Aldrich 577766), tungsten hexacarbonyl (W(CO)₆, THC, Sigma Aldrich 472956) and diethyl sulphide (C₄H₁₀S, DES, Sigma Aldrich 107247) were the

chemical precursors for Mo, W, and S, respectively. The growth was performed at 500 °C for 30 hours. The flow rate of precursors, regulated by individual mass flow controllers, were 0.01 sccm for MHC or THC, 0.3 sccm for DES, 1 sccm for H₂, and 150 sccm for Ar. NaCl was loaded as a desiccant in the upstream region to dehydrate the growth chamber. Figure S1b shows dark-field transmission electron microscopy (DF-TEM) images of individual MoS₂ grains that range from 500 nm to 1 μm in size.

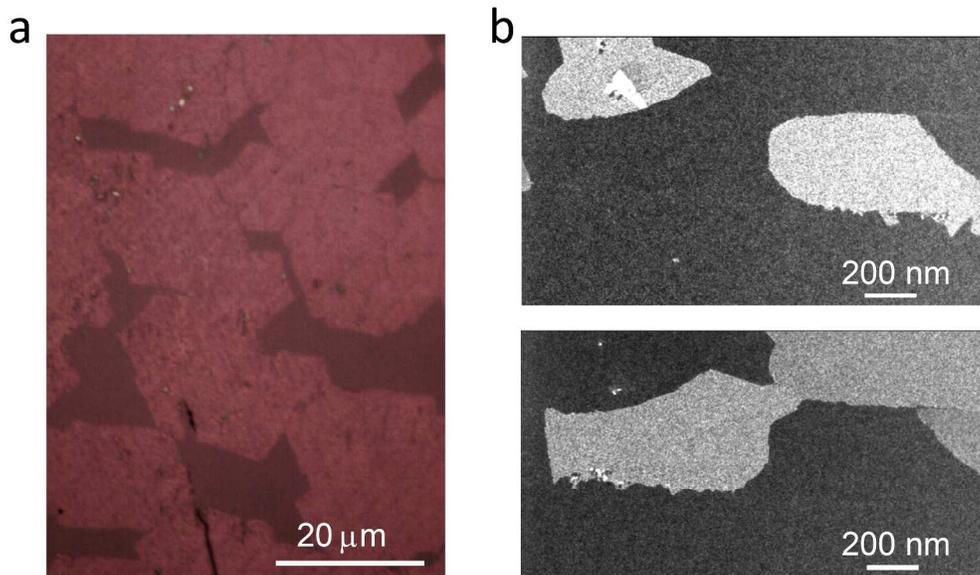


Figure S1: (a) Optical micrograph of a nearly continuous graphene film transferred to a Si/SiO₂ substrate showing a typical graphene grain size of about 20 μm. (b) DF-TEM image of individual MoS₂ grains. The grain size for MoS₂ ranges from 500 nm to 1 μm.

Optimized conditions: To create laterally-stitched graphene/TMD heterostructures we use the conditions as described above. A key parameter is the pressure of the chamber containing the Mo or W precursor (MHC or THC). We found that the ideal conditions are obtained in a steady flow with the internal pressure of the precursor vessel = 0.53 PSI, resulting in a partial pressure below 0.7 mTorr. The resulting film obtained using these parameters is homogenous without multilayer TMDs or overlapped junctions as shown in Fig. S2.

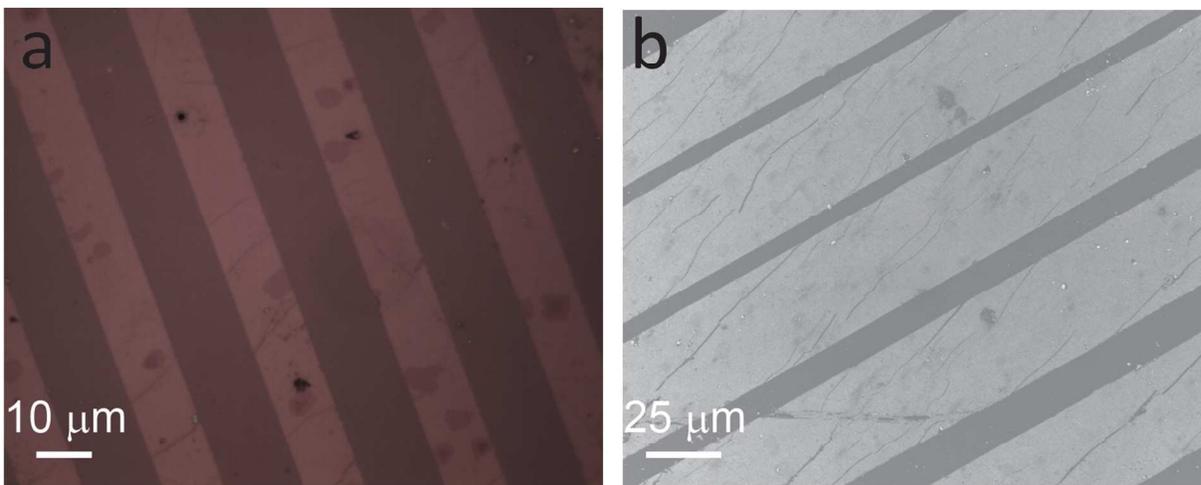


Figure S2: (a) Optical micrograph showing a heterostructure film grown under optimized conditions. Light pink: graphene, Dark Pink: MoS₂ (b) SEM image of the heterostructure film. White: graphene. Dark: MoS₂.

Increasing precursor pressure: With an increase of the pressure in the precursor chamber, the resulting film becomes non-homogenous with multilayer TMDs and with TMD nucleation observed on graphene surface. We consistently found overgrowth of the TMD layer and TMD nucleation on graphene for partial pressures of the metal precursor above 1 mTorr (Fig. S3).

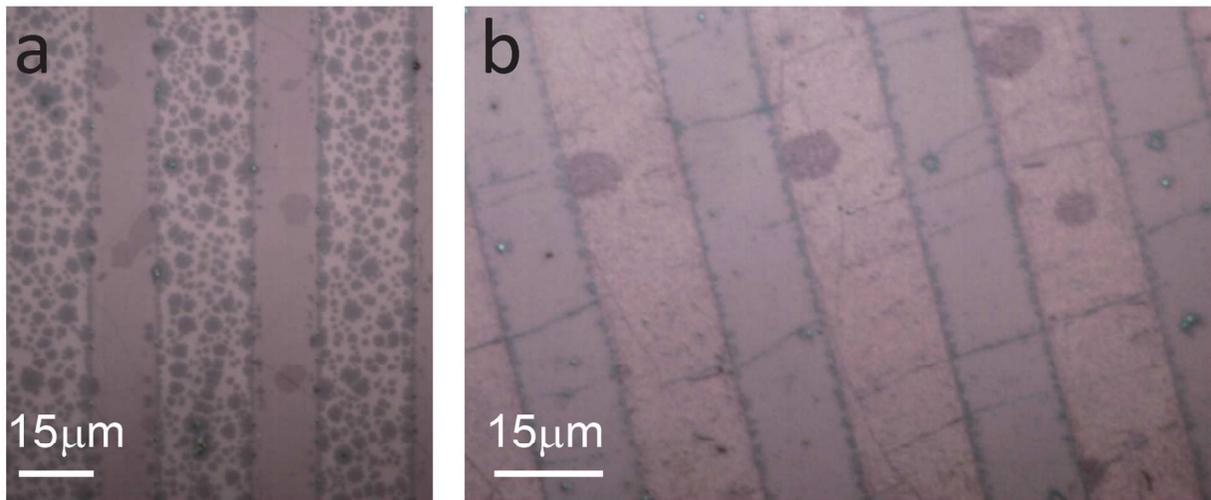


Figure S3: (a) Optical micrograph showing the nucleation at the initial stage when the TMD growth is performed under non-optimized growth conditions. (b) Optical micrograph showing the resulting heterostructure film after the growth is complete using non-optimized growth conditions.

TMD nucleation: TMD nucleation occurs at the graphene edge during the initial growth stage. We observed that the graphene edge becomes fully covered by MoS₂ early in the growth (Fig. S4). For a patterned heterostructure growth, for samples with a greater distance between the graphene stripes the average coverage of MoS₂ is decreased, indicating that the nucleation of MoS₂ occurs preferentially at graphene edges. This can be explained by a diffusion of the precursors off of the graphene film and onto the SiO₂ surface.

To explore the possibility of miniaturization of our devices we have fabricated lateral heterostructures with channel lengths as small as 200 nm (Fig. S4f). To obtain such small scale channels, the growth conditions had to be optimized (reduction of the growth time).

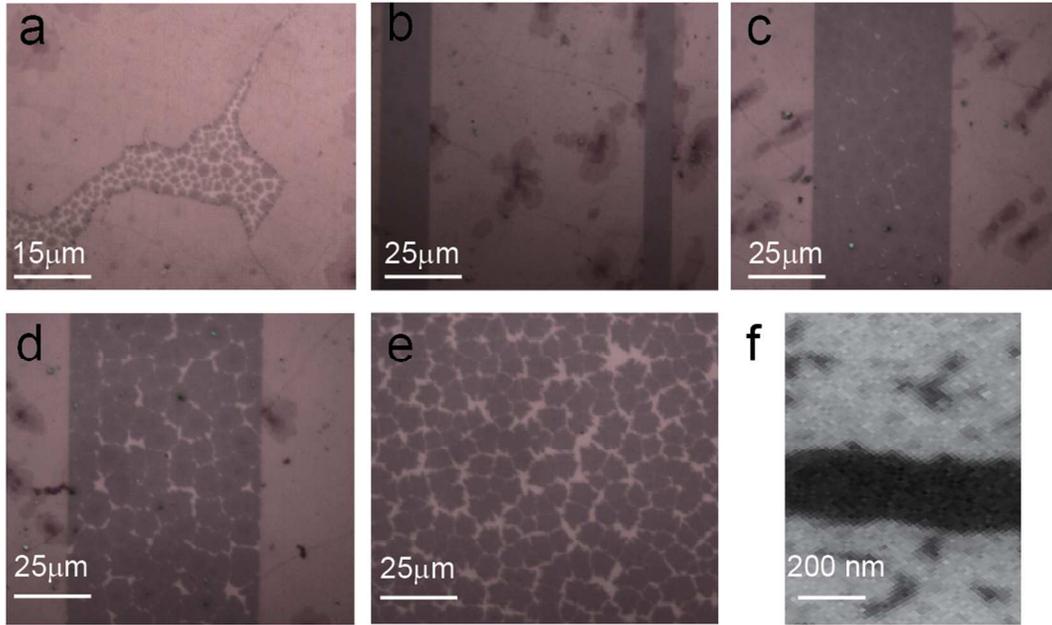


Figure S4: (a) Optical micrograph of a heterostructure film grown using non-continuous graphene at the initial growth stage, showing a graphene edge covered by MoS₂ grains. (b), (c), and (d) are the optical images showing coverage of the MoS₂ channel between graphene stripes with different channel length. (e) Optical image of the same substrate as (b), (c) and (d) showing coverage of the MoS₂ for a region containing no graphene stripes. (f) SEM image of a 200 nm WS₂ channel in between two graphene stripes.

2: Photoluminescence and Raman scattering

Photoluminescence: Photoluminescence (PL) measurements were performed under ambient conditions using a laboratory-built apparatus with a 532 nm excitation laser (Fig. S5a). The spectra were obtained by a spectrometer with a CCD camera and the images were directly taken using band pass filter.

Raman: Raman spectra (Fig. S5b) were taken under ambient conditions with a 532 nm excitation laser.

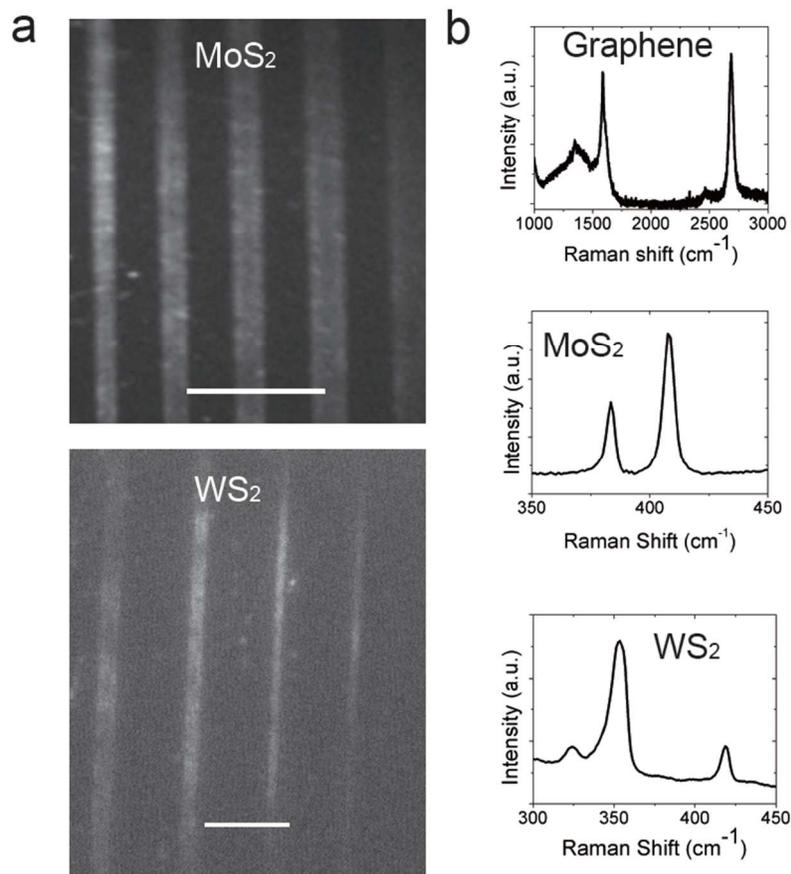


Figure S5: (a) Photoluminescence (PL) intensity mapping for MoS₂ or WS₂ on MoS₂/graphene (top) and WS₂/graphene (bottom) heterostructure films. Bright regions correspond to high intensity. The scale bar is 50 μm. (b) Raman spectra for graphene (top), MoS₂ (middle) and WS₂ (bottom) on the heterostructure film.

3: Additional DF-TEM images

TEM Sample preparation: A monolayer heterostructure film grown on a SiO₂/Si substrate was spin coated with PMMA A2 at 4000 RPM for one minute. Then the substrate was etched in 1M KOH solution at 60 °C until the film is delaminated from the substrate. The film was rinsed in deionized water three times before being transferred to a TEM grid. The chip was then annealed in an ultra-high vacuum (10^{-7} Torr) furnace at 350°C for 5 hours to remove the PMMA.

DF-TEM: DF-TEM images with the corresponding electron diffraction patterns were taken using an FEI Tecnai T12 Spirit TEM operating at 80 keV. The line profiles in Fig. S6a were determined using an averaging window 15 nm in width. We can extract the resolution of our DF-TEM images (~8 nm) from such profiles by the change in intensity at the edge of different MoS₂ and graphene grains. We observe that the transitions between graphene and MoS₂ and between different MoS₂ grains show the same resolution-limited abruptness, indicating lateral stitching of the MoS₂ and graphene.

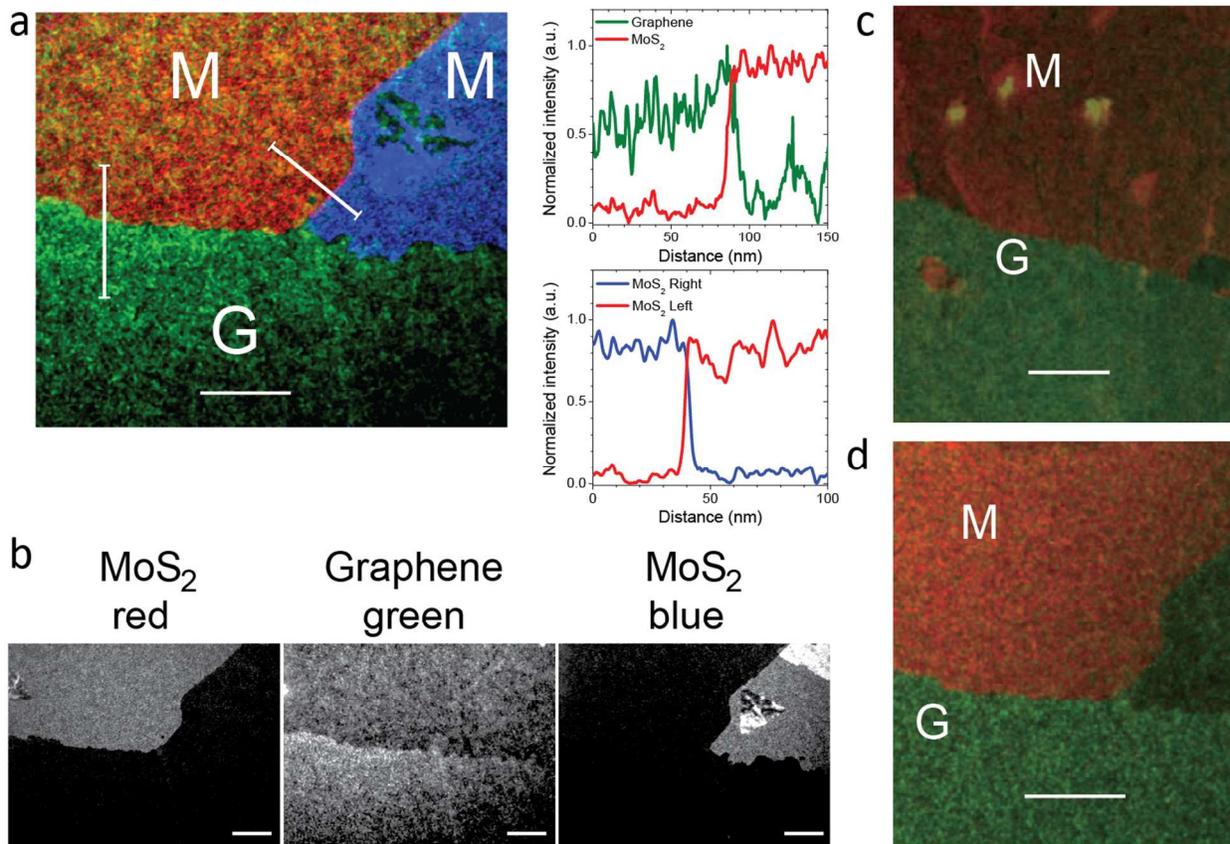


Figure S6: DF-TEM images taken from three different samples showing no overlapped junctions within the resolution of the TEM. The scale bars are 100 nm. M = MoS₂, G = Graphene. (a) *Left:* DF-TEM image of a graphene/MoS₂ junction. *Right:* Line profiles taken at a boundary between graphene and MoS₂ and at a boundary between two different MoS₂ grains. (b) Individual DF-TEM images used to obtain the colored image shown in (a). Two additional images from different randomly selected junctions are shown in (c) and (d).

4: Additional TEM Studies

In addition to DF-TEM imaging we also performed high-angle annular dark-field (HAADF) image and electron energy loss spectroscopy (EELS) analysis of a graphene/MoS₂ junction. Figure S7a shows the HAADF image, which is highly sensitive to the atomic and shows heavier elements in bright contrast. We observe a sharp transition in the HAADF contrast at the junction.

A similar abrupt transition is observed in the chemical analysis extracted by EELS (Figure S7b). In order to improve the signal-to-noise ratio of the EELS line profile, we averaged 22 spectra over 80 nm parallel to the junction.

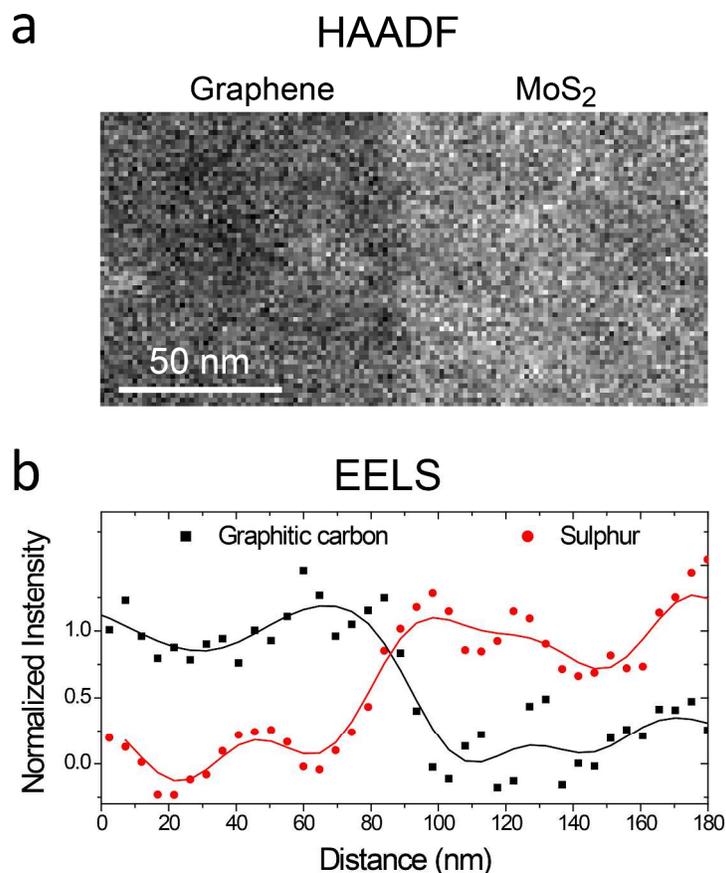


Figure S7: (a) HAADF image of a graphene/MoS₂ junction. Low atomic number (graphene) is shown in dark and high atomic number (MoS₂) is shown in bright contrast. (b) Normalized line profile for the integrated graphitic carbon peak (black) and the Sulphur peak (red) obtained by EELS for the same region shown in (a).

5: Device fabrication

We start our device fabrication by transferring chemical-vapor-deposition (CVD) grown graphene onto a heavily doped Si substrate with a thin (300 nm) SiO₂ layer to be used as back gate electrode and dielectric. The graphene is then patterned into stripes by optical lithography and oxygen plasma etching using a reactive ion etching (RIE) tool. The desired TMD (MoS₂ or WS₂) is grown using the optimal conditions described above in order to avoid overlapped regions between the graphene and the TMD.

To contact the graphene (or TMD) sheet, Ti/Au (5/50 nm) metal electrodes are fabricated using conventional optical and electron beam lithography methods followed by metal deposition using an electron beam evaporator at high vacuum (10⁻⁷ Torr). The lift-off step is done by soaking the chip in acetone for several (> 5) hours and rinsing with isopropyl alcohol. The conducting channel is defined by subsequent lithography and RIE steps. For the top gate dielectric we deposit an Al layer (1 nm) to be used as a seeding layer for HfO₂ deposited by atomic layer deposition (30 – 60 nm). The top gate electrode is defined by an optical lithography step followed by metal deposition (Ti/Au 5/50 nm) in an electron beam evaporator.

6: 2-Probe mobility as a function of channel length for 1DG contacts

Figure S8 shows the 2-probe field-effect mobility as a function of the channel length for MoS₂-based devices with 1DG contacts at room temperature. We observe mobilities between 10 and 30 cm²/Vs, consistent with our previous work¹.

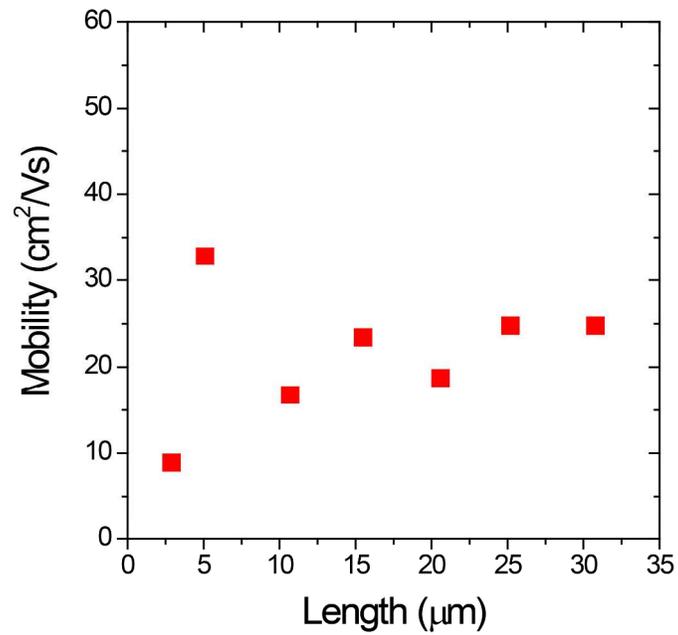


Figure S8: Room temperature 2-probe field-effect mobility as a function of channel length for MoS₂-based devices with 1DG contacts.

7: Contact resistance from 4-probe measurements

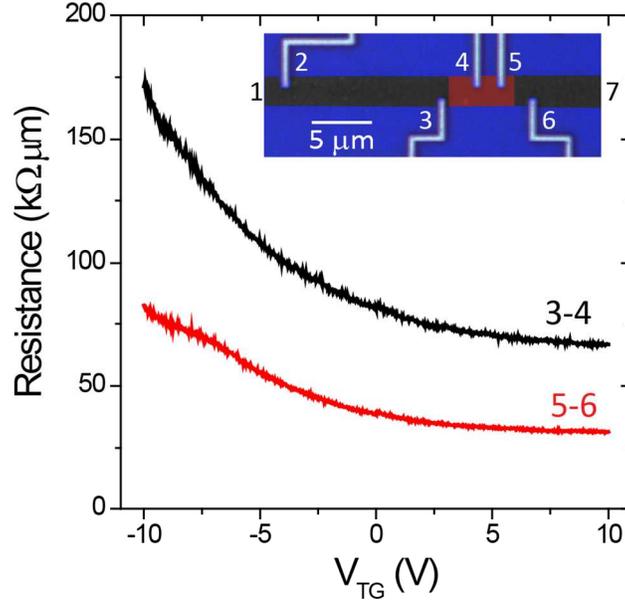


Figure S9: As-measured resistance (R_{34} and R_{56}) for the junctions in between electrodes 3 and 4 (black) and 5 and 6 (red) as a function of the top gate voltage (V_{TG}). *Inset:* Optical micrograph of the device indicating the current electrodes (1 and 7, outside the field of view), and several voltage probes, numbered 2 to 6.

For the 4-probe measurements of the graphene/TMD contact resistance, we drive a current $I_{17} = 100 - 200$ nA between two electrodes (1 and 7, out of the field of view in Fig. S9) and measure the voltage between all the other adjacent contacts simultaneously (V_{23} , V_{34} , V_{45} , V_{56}).

The sheet resistance for graphene is given by: $R_g = \frac{V_{23}}{I_{17}} \frac{W}{L_{23}}$, where $W = 2.37$ μm is the channel width, and $L_{23} = 12.37$ μm is the distance between electrodes 2 and 3. Analogously, the sheet resistance for MoS₂ is given by: $R_{\text{MoS}_2} = \frac{V_{45}}{I_{17}} \frac{W}{L_{45}}$, where $L_{45} = 2.02$ μm .

The resistance across a particular junction, e.g. between electrodes 5 and 6, can be described as:

$R_{56} = \frac{V_{56}}{I_{17}} = R_g \left(\frac{L_g^{56}}{W} \right) + R_c + R_{\text{MoS}_2} \left(\frac{L_{\text{MoS}_2}^{56}}{W} \right)$, where L_g^{56} is the length of the graphene region between electrodes 5 and 6, R_c is the contact resistance between the graphene and the MoS₂, and $L_{\text{MoS}_2}^{56}$ is the length of the MoS₂ region between electrodes 5 and 6. Both L_g^{56} and $L_{\text{MoS}_2}^{56}$ are

obtained from the optical image shown in the inset of Fig. S9. Finally, the value for R_c is obtained by subtracting the graphene and MoS₂ contributions to R_{56} .

8: IV curves for low carrier density at room temperature

As mentioned in the main text, the IV characteristics of the 1DG contacts are linear at room temperature for a wide range of V_{BG} values. Figure S8 shows a source-drain current *versus* voltage characteristics at 300 K for a sample containing two junctions in series for different values of V_{BG} .

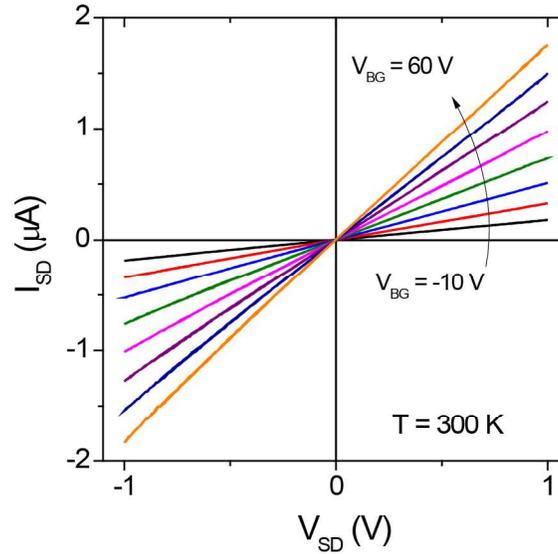


Figure S10: Source-drain current (I_{SD}) *versus* voltage (V_{SD}) for different values of back gate voltage (V_{BG}) at 300 K for low doping ($n < 3 \times 10^{12} \text{ cm}^{-2}$).

9: Arrhenius plots for measurements of the barrier height

To extract the barrier height we performed temperature dependence measurements of the junction resistance to determine an activation energy. Assuming a thermo-ionic emission model,² the source-drain current across the device is given by: $I_{SD} = AT^{3/2} \exp\left(\frac{-q\Phi_B}{k_B T}\right) \left[1 - \exp\left(\frac{qV_{SD}}{\eta k_B T}\right)\right]$, where I_{SD} is the source-drain current, A is the effective Richardson constant, T is the temperature, q is the elementary charge, k_B is the Boltzmann constant, $V_{SD} = 50 - 100$ mV is the source-drain bias, and η is the ideality factor. The ideality factor is related with tunneling at high carrier concentration at low temperatures and was obtained from a plot of the logarithm of I_{SD} as a function of V_{SD} at 4.2 K. The barrier height is obtained from the slope of the plot of $\ln\left(\frac{I_{SD}}{T^{3/2}}\right)$ versus $1/k_B T$ (Arrhenius plot). The slope is given by: $-q\Phi_B + qV_{SD}/\eta$.

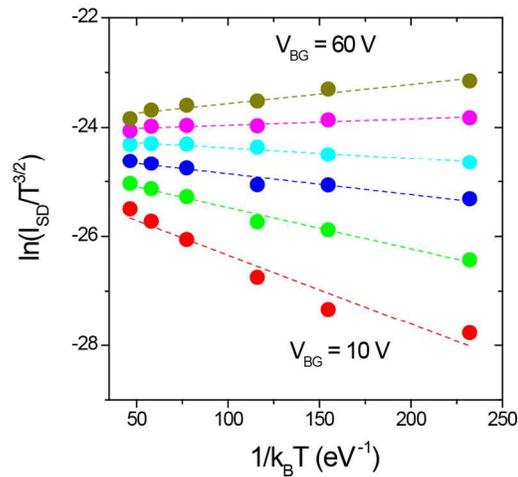


Figure S11: Arrhenius plot used to extract the barrier height in Figure 5b (main text).

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