Large-scale chemical assembly of atomically thin transistors and circuits

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Next-generation electronics calls for new materials beyond silicon, aiming at increased functionality, performance and scaling in integrated circuits. In this respect, two-dimensional gapless graphene and semiconducting transition-metal dichalcogenides have emerged as promising candidates due to their atomic thickness and chemical stability. However, difficulties with precise spatial control during their assembly currently impede actual integration into devices. Here, we report on the large-scale, spatially controlled synthesis of heterostructures made of single-layer semiconducting molybdenum disulfide contacting conductive graphene. Transmission electron microscopy studies reveal that the single-layer molybdenum disulfide nucleates at the graphene edges. We demonstrate that such chemically assembled atomic transistors exhibit high transconductance (10 μ S), on-off ratio (~10⁶) and mobility (~17 cm² V⁻¹ s⁻¹). The precise site selectivity from atomically thin conducting and semiconducting crystals enables us to exploit these heterostructures to assemble two-dimensional logic circuits, such as an NMOS inverter with high voltage gain (up to 70).

oore's Law projects that integrated electronics will soon require sub-10 nm transistors. With silicon, this will be extremely challenging, because the thickness of the transistor's channel will become greater than the channel length, ultimately leading to difficult electrostatic control via the transistor gate¹. The chemical syntheses of nanomaterials such as inorganic nanowires²⁻⁴ and carbon nanotubes⁵⁻⁷ have been aimed at addressing this issue, but using such materials in electrical devices requires precise placement and orientation using complex fabrication techniques^{1,4,7,8}. Although great strides have been made, for example in the demonstration of a carbon nanotube computer⁹, the fabrication required to create sophisticated electronic circuitry using these materials remains difficult or impractical. Recently, crystalline two-dimensional materials, with their ultimate atomic thickness, have shown promise as an elegant solution to the problematic scaling of silicon transistors. Graphene, the first widely studied twodimensional crystal, is a semi-metal with a massless carrier dispersion, high mobility and easily tunable Fermi level. Single-layer graphene lacks an electronic bandgap, rendering it unsuitable for a transistor channel (as it cannot be turned 'off'easily), but its excellent conductive properties make it ideal for the interconnections and wiring of next-generation devices¹⁰⁻¹³.

Semiconducting transition-metal dichalcogenides (TMDCs) such as molybdenum disulfide (MoS₂) have recently found success in a single-layer transistor^{14,15}. While early works using single-layer TMDCs relied on the 'Scotch tape' method of micro-mechanical exfoliation, considerable efforts have established the chemical vapour deposition (CVD) of a range of TMDCs^{16–20}. For example, CVD lateral heterostructures that include two different TMDCs have been used to create atomically sharp p–n junctions^{21,22}. Lateral heterostructures using wide-gap insulators and conductors, hexagonal boron nitride (h-BN) and graphene have also been grown^{23,24}, but the spatially controlled synthesis of conductor–semiconductor heterostructures—a necessary step

towards achieving full atomically thin circuitry-has not yet been demonstrated.

Here, we demonstrate the chemical assembly of heterojunctions using graphene– MoS_2 -graphene heterostructures, as shown in Fig. 1a. Unlike previous reports that rely on transferring and physically assembling transistors using these two-dimensional crystals^{25–29}, we chemically grow these transistors on a large scale. By effectively injecting current from the graphene through the MoS_2 , we demonstrate an NMOS inverter for logic operations using such heterostructure transistors.

Graphene-MoS₂ growth and optical characterization

To chemically assemble our heterostructures, single-layer graphene was first transferred onto a silica substrate (large-scale growth and transfer techniques for graphene are now common^{10,11}). Patterns of channels (we have chosen a grid) were defined using oxygen plasma. The graphene on silica was then placed into a quartz tube for seed-promoted CVD growth of single-layer MoS₂. The fabrication on graphene before this growth results in dangling bonds and lithographic residues, causing a high density of MoS₂ nucleation at the edges. The lack of new chemical bonding and crystalline mismatch between these two crystals results in nanometre-scale overlap junctions between the two atomic crystals. Preferential growth within the SiO₂ channels results in the merging of individual domains, which form a continuous, polycrystalline single layer of MoS₂, consistent with the observations made with large-area chemical vapour growth on bare substrates^{16,19,20} (see Methods for additional growth details).

Using solid precursors, we successfully grew heterostructures with millimetre-scale coverage, as shown in Fig. 1b, where the uniform single-layer MoS_2 can be observed within defined channels in the etched graphene. With optical microscopy, triangular MoS_2 grains are observed along the graphene, indicating that the edge serves as a site for crystal nucleation (see Supplementary Fig. 1 for

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Figure 1 | Growth scheme and optical characterizations of the graphene-MoS₂ heterostructure. a, Scheme of the heterostructure growth process. Graphene is first etched into channels and MoS₂ begins to nucleate around the edges and within the channel. On the edges, MoS₂ forms a thicker overlap junction with the graphene. Finally, further growth results in MoS₂ completely filling the channels. **b**, Scanning electron microscope image of the chemically grown MoS₂ between the edges of the graphene. The image shows a large scale of coverage, on a millimetre scale. Typical triangular crystal domains grow in areas without graphene, as seen on the right side of the image. **c**, Optical image of the heterostructure. Within the narrow channel, the MoS₂ areas (marked by the black arrows). The dashed box is the area used for the spectral mapping in **d-f. d**, Raman mapping using integrated peak intensities from 1,500 to 1,700 cm⁻¹. Graphene's Raman signature is the G peak centred near 1,600 cm⁻¹. The mapping shows that only the contact areas are graphene. The same results are observed from integrating the intensities around graphene's 2D peak, centred near 2,700 cm⁻¹. **e**, Raman mapping using the integrated peak intensities from 380 to 415 cm⁻¹. MoS₂ has two distinct Raman peaks, E_{2g} and A_{1g}, centred near 385 and 410 cm⁻¹, respectively. These Raman modes support the finding that the crystalline region inside the grown region is indeed MoS₂. **f**, Photoluminescence mapping of emission from MoS₂ centred at 660 nm. MoS₂ transitions from an indirect to direct bandgap semiconductor when it is thinned to a single layer, showing high emission only in areas without graphene.

additional optical images). Figure 1c also shows an optical image of the heterostructure, with brighter nucleation regions (black arrows in Fig. 1c) along the junction between the graphene and MoS_2 , consistent with previous reports^{18,19}.

To confirm the existence of the proposed heterostructure we performed Raman spectroscopy mapping. Graphene areas preserve their Raman signature after growth (Fig. 1d), as seen from either the G or 2D peak³⁰, indicating that the growth process is nondestructive to graphene. Using the E_{2g} and A_{1g} modes³¹ of MoS₂, the boundaries of MoS₂ growth are also clearly delineated (Fig. 1e). Within the channel regions, the Raman spectra were used to map out the thickness of the sample (Supplementary Fig. 3). Based on the frequency difference between the two modes corresponding to different layer numbers, we found the channel to be predominately single-layer MoS₂. Larger peak differences and higher intensities correspond to thicker nucleation centres (black arrows in Fig. 1e). These findings are confirmed by photoluminescence mapping of the channel, as shown in Fig. 1f. Because single-layer MoS₂ possesses a direct bandgap, strong emission centred at ~1.85 eV was detected, in agreement with the findings observed from the Raman spectra. The weaker areas of the photoluminescence signal also correspond well to the higher intensities in the MoS₂ Raman signals, which indicate the thicker MoS₂ regions (black arrows in Fig. 1e,f). The photoluminescence energy and peak width, as well as the Raman spectra of the MoS₂ within our heterostructure, are consistent with reports of CVD MoS₂ on SiO₂ substrates, indicating that the presence of graphene does not influence the relative strain and doping levels of the MoS_2 (Supplementary Fig. 3). Additionally, the MoS_2 within the channel is a uniform single layer, with the exception of sparse multilayer patches that occur near the nucleation centres in the channel and around the edges. The optical spectroscopy results agree well with the microscopy image, showing the same thicker areas. Representative full spectra are provided in Supplementary Fig. 2.

Crystallinity and nucleation at the edge

We evaluated the crystallinity of the grown single-layer MoS₂ as well as the junction between the MoS₂ and graphene using transmission electron microscopy (TEM)^{17,18,32}. The dark-field TEM (DF-TEM) images in Fig. 2a were used to create a false-colour map of the MoS₂ grains in the heterostructure using the diffraction pattern (Fig. 2b). The selected area contains a single grain of graphene, as indicated by graphene's diffraction spots in Fig. 2b. The colourmapped grains of MoS₂ in Fig. 2a indicate that the growth of MoS₂ along the graphene edge is independent of the graphene lattice orientation. This is not unexpected, as the graphene fabrication results in the edges being random and containing many defects. Coupled with optical microscopy images (Supplementary Fig. 1), the MoS₂ grain size along the edges of the graphene was found to be ~1 µm.

Annular dark field (ADF) scanning transmission electron microscopy (STEM) was used to provide high-resolution images of the graphene– MoS_2 junction (Fig. 2c). Using an intensity plot from a line profile in the image (Fig. 2c inset; for additional



Figure 2 | Electron microscopy of the graphene-MoS₂ **structure and compositional map. a**, False-colour DF-TEM overlay showing the MoS₂ is continuous and polycrystalline at the graphene-MoS₂ junction. The MoS₂ grains show random orientations with respect to the graphene. A magnified DF-TEM overlay of both graphene and MoS₂ is shown in Supplementary Fig. 4. b, Corresponding diffraction pattern from the region in **a** indicates the graphene is single crystal. The four coloured circles indicate distinct grain orientations of the MoS₂ used to create the false-colour map in **a**. **c**, ADF-STEM image of the graphene-MoS₂ junction, showing an overlap of ~100-200 nm. The graphene edge was identified by the image intensity, which is proportional to Z^r, where Z is the atomic number and $1.3 < \gamma < 2$. Inset: Corresponding intensity line profile from the white dashed line across the junction. A detailed 8-bit image grey value calibration is shown in Supplementary Fig. 4. Gr, graphene; 1L, single layer. **d**, EELS spectra of graphene, MoS₂ and the junction show S and C edges. The graphene signature in the σ^* peak shows up on the graphene and the junction, while only amorphous carbon (π^*) from the polymer residue shows up on the MoS₂. **e**, Elemental EELS map of graphene (yellow) and MoS₂ (red), confirming the overlapped junction. The graphene and MoS₂ maps are integrated from the yellow and red regions, respectively, of the spectra in **d** using multiple linear least-squares (MLLS) fitting.

information see Supplementary Fig. 4) it was possible to elucidate that the junction is a $\sim 100 \text{ nm}$ overlap of single-layer MoS₂ on top of graphene. In addition, further growths of secondary and tertiary patches of MoS₂ were observed close to the graphene edge, though never growing within the graphene surface. This confirms that nucleation of the MoS₂ occurs at the edges of graphene, with defects of the graphene-MoS₂ junction allowing multilayer patches to grow on top. The overlapped region retains the crystalline nature of MoS₂, as observed in an atomic-resolution high-angle ADF-STEM image of the junction in Supplementary Fig. 4f. Although the junction in Fig. 2c is an overlap junction and not atomically sharp, there is still a significant barrier to further growth of the MoS₂ on graphene. This is clearly seen in Fig. 2a, where larger grains terminate near the boundary between the graphene and MoS₂, and multilayer patches do not grow into the graphene region (Fig. 2c).

The nature of this overlap can be probed using electron energy loss spectroscopy (EELS), which provides compositional and bonding information for the heterostructure. From the spectrum in Fig. 2d, the carbon K-edge is used to identify graphene, while distinct molybdenum and sulfur edges can be used to identify the MoS_2 . Using the distinct graphene signature (σ^*) and the sulfur edges for MoS_2 , a compositional map can be created (Fig. 2e). Within the overlapped region, the lack of additional new peaks as well as the retention of graphene and MoS_2 signatures confirms that the overlap is a van der Waals heterojunction without the formation of additional covalent bonding in the energies probed.

The role of graphene as a growth mask is surprising, as the nucleation of MoS₂ (a three-atom-thick crystal) occurs from graphene's one-atom-thick edges. Thus far, lateral heterostructures between van der Waals crystals have shared similar lattice constants, or chemistries. Heterostructures grown using two different TMDCs show that there is typically a preference for lateral epitaxy accompanied by vertical growth as time increases³³. In the case of graphene and MoS₂, the lattice mismatch (in- and out-of-plane) as well as distinct chemical differences inhibit in-plane bonding and epitaxy, leading to the formation of an overlap junction. The results indicate a significant reduction in the nucleation energy barrier of MoS₂ at the edges of graphene compared with its surface. Lithographically opening up channels on graphene causes defects and dangling bonds, while leaving residues at the edges. This leads to increased nucleation accompanied by slight surface growth of MoS₂, which forms a van der Waals overlapped junction. Similar reports have been made for CVD graphene, as an overlap of similar width forms during the growth of graphene-graphene junctions^{34,35}. Growth on the graphene is limited due to difficulties with the vertical epitaxy of MoS₂ on graphene, as large crystalline areas require unique growth conditions that are not present in this particular growth³⁶. Additionally, the hydrophilic SiO₂ surface (resulting from the oxygen plasma treatment used to etch graphene) has a higher affinity for transport of the growth seed, especially compared with the hydrophobic graphene surface²⁰. Thus, we summarize the growth process as edge nucleation and the formation of an overlap junction



Figure 3 | Room-temperature electrical transport measurements of the graphene-MoS₂ heterostructure transistor. **a**, Optical image of a FET device array fabricated from one growth. **b**, Typical source-drain current-voltage curves measured at differing top-gate voltages. Below a source-drain bias of 1 V, linear ohmic behaviour is observed, suggesting efficient electrical contact between the graphene and MoS₂. Current saturation is achieved at a bias voltage of 1.5 V. **c**, A typical source-drain current curve measured by altering the top-gate voltage for a source-drain voltage of 1 V. From the curve, an on-off ratio for the heterostructure transistor is estimated to be ~1 × 10⁶. Inset: Transconductance of the heterostructure under V_{DS} = 1 V, reaching a peak of 10 µS. From the curve, the room-temperature field-effect mobility is estimated to be ~20 cm² V⁻¹ s⁻¹ for this device. **d**, Histogram of device mobility performed on 100 devices, yielding a mobility of 17.2 ± 3.9 cm² V⁻¹ s⁻¹. **e**, Field-effect mobilities from **d** as a function of transistor length. Two growths were performed to give the red and blue data points. The mobilities across different growths both hover in the 15-20 cm² V⁻¹ s⁻¹ range, showing that neither the channel length nor growth conditions have a large impact on the field-effect mobility. **f**, Contact resistance of the heterostructure device and graphene-only control devices (after the growth and fabrication processes) extracted using the transfer length method. Inset: Typical transfer length method for both the complete heterostructure (V_{DS} = 0.2 V) and graphene (V_{DS} = 0.01 V) at V_{TG} – V_{Th} = 4 V. The total contact resistance of ~10 kΩ µm. Error bars in **e** and **f** are due to sample to sample variations from the growth locations.

encouraged by defects in the graphene, combined with ${\rm MoS}_2$ 'filling' through seeded growth on hydrophilic areas.

Heterostructure transistors and contact resistance

Critically, the heterostructure uses graphene as a metallic electrode to inject current into the MoS₂. Transistors fabricated using physically transferred graphene–MoS₂ heterostructures have demonstrated that graphene is an efficient electrical contact material^{25–29}. Owing to the tuning of graphene's Fermi level, graphene is able to reduce the contact barrier and form an ohmic contact with MoS₂. An inherent challenge in the use of single-layer MoS₂ arises because of the typical Schottky-limited transport³⁷, as the CVD MoS₂ results in the contact resistance (R_c) ranging from ~50 to 200 k Ω µm (refs 19, 38). Hence, the electrical properties that result from nucleation along the graphene edges are expected to play a crucial role in the electrical transport characteristics of these heterostructure transistors.

The field-effect transistor (FET) performance of a typical graphene– MoS_2 -graphene structure was measured at room temperature (see Fig. 3a for optical images of devices). The *I*-*V* curves show linear behaviour at small source–drain voltages, confirming the ohmic contact between the graphene and the single-layer MoS_2 film (Fig. 3b). In addition, the source–drain current saturates at a larger source–drain voltage (1.5 V), a crucial parameter for reaching the maximum possible operating speeds and maximizing the intrinsic

transistor gain¹². Through electrical transport measurements, the heterostructure exhibits typical n-type channel characteristics. Figure 3c shows the top-gate dependence of the heterostructure FET under bias voltages of 1 V. The turn-on voltage is measured at around -1.5 V, indicative of a relatively high electron-doping concentration. These measurements show that the heterostructure exhibits a high on-off current ratio of $\sim 1 \times 10^6$, peak transconductance close to 10 μS , and a corresponding carrier concentration of $5.6\times 10^{12}~cm^{-2}.$ We measured 100 devices and extracted a fieldeffect mobility of $17.2 \pm 3.9 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ (Fig. 3d), comparable to the mobilities reported for physically transferred heterostructures²⁹. In addition, we notice that the mobility across various channel lengths yields similar values of 15–20 cm² V^{-1} s⁻¹ (Fig. 3e), indicating that the mobility does not vary with channel length. Additional optical images and transistor performance are provided in Supplementary Fig. 5, while graphene-only device characteristics are presented in Supplementary Fig. 6.

To determine the contact resistance R_c between the graphene and MoS₂, we used the transfer length method to extract the lengthdependent resistance in both the complete heterostructure and our graphene samples after growth and fabrication (Fig. 3f, inset). The total R_c (metal–graphene–MoS₂) is plotted versus gate voltage in Fig. 3f, showing that the minimum R_c at high gate voltages when the device turns on is ~11 k Ω µm, with the graphene–metal R_c corresponding to ~1 k Ω µm. Thus, we extract an R_c of ~10 k Ω µm



Figure 4 | Demonstrating logic through a heterostructure inverter. a, Illustration of an inverter circuit. Inset: Circuit diagram. Using two n-type transistors to create an inverter, the bottom transistor (load) is subject to a driving voltage while the top transistor (driver) is grounded. The load gate and common electrode are read through as the output voltage. The input voltage is applied through the driver gate. At low input voltages, the driver is turned off and the output voltage is read as the driving voltage ('1'). At higher input voltages, the system inverts and the output voltage becomes zero, as the driver turns on ('0'). **b**, Inverter transfer characteristics for driving voltages of 1, 2 and 4 V, showing the inversion behaviour at a threshold voltage of around –1.7 V. As described in **a**, low input voltages yield an output voltage equal to the driving voltage. After crossing the threshold, the output voltage is read as 0. **c**, The absolute differential of the inverter curves gives the voltage gain. At a driving voltage of 4 V, an extremely high voltage gain of 70 is achieved, among the highest in TMDC inverters.

for the graphene–MoS₂ contact. Although multilayer MoS₂ and graphene may have a much lower contact resistance³⁹ due to the electronic structure changes leading to improved electrical properties, the values we find are markedly reduced compared with the contact resistances of physically transferred CVD monolayer MoS₂ and graphene, which report $R_c = 100 \text{ k}\Omega \text{ }\mu\text{m}$ (ref. 29). Hence, we have demonstrated that the chemically synthesized graphene–MoS₂ structure reduces the contact resistance compared with traditional metal-contacted CVD monolayer MoS₂, while preserving the mobilities achieved from the physically transferred heterostructures, in a scalable method.

Demonstrating device logic

To demonstrate the additional applicability of our structure, the two-dimensional atomic logic circuitry of an inverter (a NOT gate; Fig. 4a) was assembled using our heterostructures. Figure 4b shows the static voltage transfer characteristics of the heterostructure NMOS inverter with driving voltages of 1, 2 and 4 V, demonstrating clear inverting behaviour. At low $V_{\rm in}$, starting at -2.5 V, $V_{\rm out}$ is consistently at the level of $V_{\rm DD}$. Once $V_{\rm in}$ reaches a threshold of around -1.7 V, $V_{\rm out}$ quickly switches to zero and maintains this voltage up to $V_{\rm in} = 0$ V. The heterostructure inverter also yields an extremely high voltage gain, reaching 70 for $V_{\rm DD} = 4$ V (Fig. 4c), one of the highest reported of all inverter gates made from TMDC materials^{40–42}. Having now demonstrated a logic application, it is clear that control over arbitrarily designed patterns will lay the foundation for chemically grown atomic computing.

During the review process, we became aware of an independent work (submitted at nearly the same time as this work) that also reported the growth of similar heterostructures using graphene and MoS₂ (ref. 43). However, there is a key difference, in the lower contact resistance reported here (10 k Ω µm), compared with that in ref. 43 (300 k Ω µm). This ultimately leads to an order of magnitude higher inverter gain (70, compared with 6 in ref. 43). Our DF-TEM results (Fig. 2a) confirm that the MoS₂ grains have no orientation preference to graphene, and the EELS (Fig. 2d,e) results definitively indicate the junction to be of a van der Waals nature without new bonding. Finally, our device statistics prove this method's scalability and repeatability, which will be necessary for future applications.

Conclusion

We have successfully demonstrated chemically assembled heterostructures composed of single layers of conductive graphene and semiconducting MoS₂. Large-area coverage (over a millimetre in size) and functionality is achieved due to the dual nature of graphene as a growth template and electrical contact. Using over 100 heterostructure FETs, we show these devices have on-off ratios of 1×10^6 and mobilities centred around $17 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and up to $\sim 30 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. These transistors are suitable for inverters that have voltage gains up to 70. It is envisioned that these heterostructures will be able to form ballistic transistors in the short-channel limit with high performance, enabling us to develop alternatives to silicon technologies. Coupled with the industrial wafer-scale compatibility of graphene and MoS₂ growth, spatial control over the synthesis of two-dimensional conductor–semiconductor heterostructures will pave the way for next-generation electronics and computing.

Methods

Methods and any associated references are available in the online version of the paper.

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Author contributions

M.Z., Y.Y. and X.Z. conceived the project. Graphene fabrication was done by Y.Y. and Y.X. M.Z. grew the heterostructure. M.Z. and H.Z. performed the optical characterizations and analysis. Y.Y., S.W. and Y.X. fabricated electrical devices. M.Z. and Y.Y. performed the electrical measurements and analysis. Y.H. and D.A.M. performed the TEM measurements and analysis. All authors contributed to discussions and writing of the manuscript. X.Z. and Y.W. guided the research.

Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to X.Z.

Competing financial interests

The authors have filed a patent based on this work.

Methods

Graphene fabrication. Wafer-scale wet-transferred graphene was purchased from Graphenea. Photolithography was used to define a range of channel lengths in the graphene, using poly(methyl methacrylate) (PMMA) and i-Line as photoresist. Oxygen plasma was then used to etch away the opened channels in the graphene. The photoresist was washed away using acetone and then annealed in an Ar/H₂ environment for 2 h at 300 °C.

Heterostructure growth. Heterostructure growth was performed following a procedure similar to reports of seeded CVD MoS_2 growth²⁰. The chemical seed perylene-3,4,9,10-tetracarboxylic acid tetrapotassium salt (PTAS) was purchased from 2D Semiconductors and dispersed in water to create a solution corresponding to ~20 μ M, then 5 μ l of the solution was placed onto the substrate with graphene and blown off immediately using nitrogen. Approximately 18 mg of solid molybdenum trioxide powder was placed in an alumina crucible at the centre of the furnace and 16 mg of solid sulfur was placed upstream at a lower temperature. The substrate containing the graphene patterns was placed directly above the MoO₃ powder. The temperature was ramped to 650 °C at a rate of 15 °C min⁻¹ and kept at 650 °C for 5 min. High-purity argon was used as a carrier gas to first flush the tube for 5 min and a rate of 5 s.c.c.m. was used during growth. Following this growth, the furnace was opened and argon was flushed at a high flow rate to rapidly cool the sample. Samples were optically characterized using a Raman microscope set-up (Horiba LabRAM HR Evolution) with a 473 nm excitation laser.

The heterostructures covered ~20–30% of the chip exposed to the MoO₃ powder, typically further away from the centre, where unwanted bulk over-growths occur. The length of the growth region was limited by the chip and precursor length within the crucible, typically resulting in total heterostructure areas with a length of ~2 cm and width of 1–2 mm. The heterostructure growths were also performed on different chips with different channel lengths. Given a stable growth of single-layer

 MoS_2 on SiO_2 with high coverage, the MoS_2 is typically able to successfully grow within the graphene channels. The data presented are representative of proper growth of the graphene– MoS_2 heterostructure.

TEM sample preparation. The sample was coated with polypropylene carbonate (PPC). The Si/SiO₂ substrate was etched with 1 M KOH solution at 90 °C. The film supported by PPC was rinsed in deionized water three times (10 min each) and transferred to a QUANTIFOIL holey carbon TEM grid. Afterwards, the sample was baked in vacuum (1×10^{-7} torr) at 350 °C for 5 h to remove the PPC. The sample was baked again in an ultrahigh-vacuum bake-out system for 8 h at 130 °C to further remove polymer residue before loading into the STEM.

TEM, STEM and EELS. Bright-field (BF) and DF-TEM images were taken using an FEI Tecnai T12 Spirit transmission electron microscope operated at 60 keV. ADF-STEM images were taken using a Nion Ultra STEM 100 operated at 60 keV. The beam convergence angle was 35 mrad and the probe current was ~60 pA. The acquisition time was 32 µs per pixel. The EELS spectrum and map were acquired using a Gatan Quefina dual-EELS spectrometer with an energy dispersion of 0.1 eV per channel. The EELS false-colour elemental map was created by integrating the C-K edge and S-L_{2,3} edge. The imaging and EELS mapping conditions were similar to those used in other reports^{18,19,44,45}. All EELS analysis was performed with open-source Cornell Spectrum Imager software⁴⁶.

Electrical measurements. For the fabrication of field-effect transistors, Pd/Au (10/80 nm) contacts on the graphene (for current injection) were defined by electron-beam lithography, followed by electron-beam evaporation. Atomic layer deposition (ALD) of 20 nm ZrO₂ was used for the top-gate dielectric, and an additional top-gate electrode (Pd/Au) was subsequently defined on top of the MoS_2 channel.