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High-mobility junction field-effect transistor via graphene/MoS₂ heterointerface

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Monolayer molybdenum disulfide (MoS₂) possesses a desirable direct bandgap with moderate carrier mobility, whereas graphene (Gr) exhibits a zero bandgap and excellent carrier mobility. Numerous approaches have been suggested for concomitantly realizing high on/off current ratio and high carrier mobility in field-effect transistors, but little is known to date about the effect of two-dimensional layered materials. Herein, we propose a Gr/MoS₂ heterojunction platform, i.e., junction field-effect transistor (JFET), that enhances the carrier mobility by a factor of ~10 (~100 cm² V⁻¹ s⁻¹) compared to that of monolayer MoS₂, while retaining a high on/off current ratio of ~10⁸ at room temperature. The Fermi level of Gr can be tuned by the wide back-gate bias (V_{BG}) to modulate the effective Schottky barrier height (SBH) at the Gr/MoS₂ heterointerface from 528 meV (n -MoS₂/ p -Gr) to 116 meV (n -MoS₂/ n -Gr), consequently enhancing the carrier mobility. The double humps in the transconductance derivative profile clearly reveal the carrier transport mechanism of Gr/MoS₂, where the barrier height is controlled by electrostatic doping.

Graphene (Gr), which consists of carbon atoms in a planar two-dimensional (2D) array, provides a platform for a new era of 2D electronics to replace mainstream silicon-driven semiconductors owing to its excellent mobility of up to 200,000 cm² V⁻¹ s⁻¹ at room temperature^{1,2}. Yet, the poor switching due to the zero bandgap of Gr remains a critical issue hindering practical applications, despite extensive efforts such as functionalization³⁻⁵, chemical doping⁶, graphene nanoribbon formation⁷⁻¹⁰, and the development of bilayer graphene with a dual-gate structure¹¹⁻¹³. Hexagonal molybdenum disulfide (MoS₂) comprising one molybdenum atom with two surrounding sulfur atoms exhibits n -type semiconducting behavior with a high on/off current ratio exceeding 10⁸ [ref. 14,16]. Nevertheless, the moderate carrier mobility (0.1 to 10 cm² V⁻¹ s⁻¹)^{14,17-19}, is several orders of magnitude lower than that of Gr, thereby limiting the potential of MoS₂ for high-speed switching device applications.

Thus far, numerous approaches have been introduced to enhance electrical performance of MoS₂ based device, for example, gate dielectric and contact resistance engineering have been suggested for enhancing the carrier mobility of MoS₂ to achieve high switching performance. High- κ (e.g., Al₂O₃, HfO₂) dielectrics suppress the surface reaction and enhance the dielectric screening effect²⁰⁻²², thereby enhancing the carrier mobility of MoS₂ (reaching 81 cm² V⁻¹ s⁻¹)²³. Sub-stoichiometric high- κ dielectrics generate more carriers in the thin MoS₂ layer, effectively screening out various Coulomb scattering sources derived from polymer residues, charged impurities, and interface states^{15,24}. Encapsulating h -BN is a prospectively ideal approach²⁵, but mechanical exfoliation is impractical because of the scalability of the cleaved film. Contact resistance engineering by utilizing a low work-function metal such as titanium²⁶ or scandium²⁷ is one alternative approach for achieving high carrier mobility. Despite lowering the Schottky barrier height and improving the carrier injection of multilayer MoS₂ significantly, the choice of contact metals for the monolayer in this approach is very limited due to Fermi level pinning depending on the surface states or defect sites of the metal/MoS₂ interface. Recently, it has been successfully demonstrated that monolayer Gr enables to suppress the Schottky barrier height sufficiently at Gr/MoS₂ heterointerface as an ideal contact material for 2D electronic materials²⁸, which facilitates diverse

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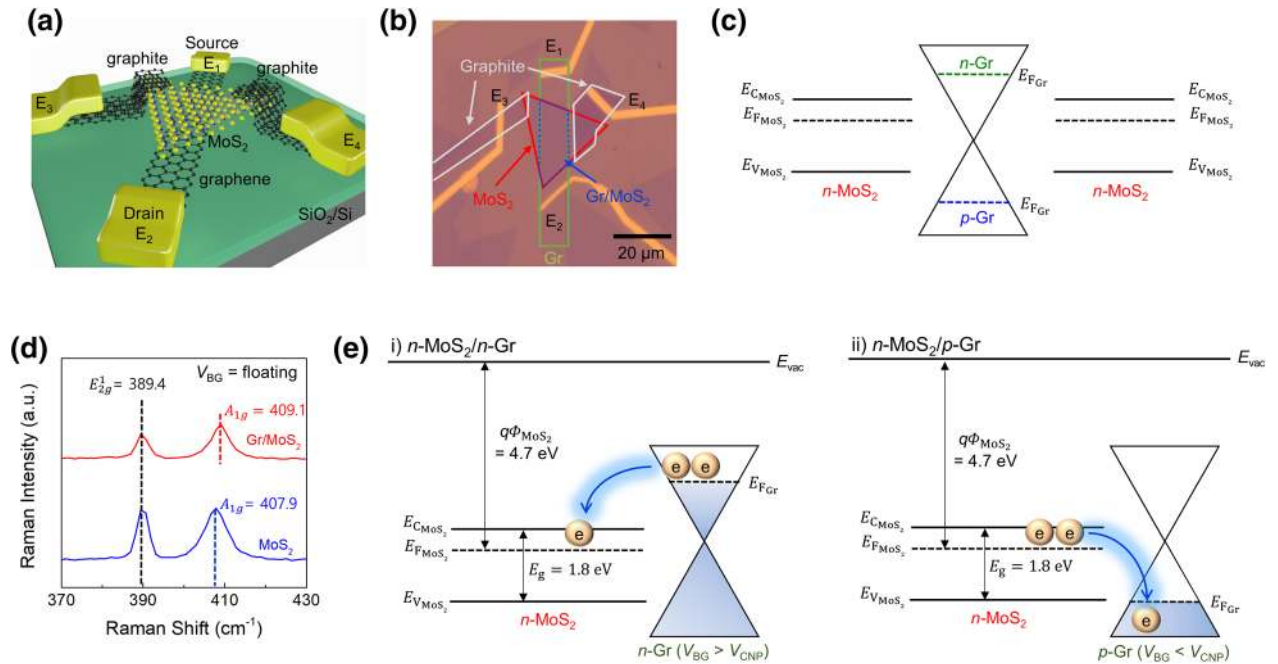


Figure 1. Device structure and Raman characterization of the Gr/MoS₂ heterostructure. **(a)** Schematic of a two-terminal device with the Gr/MoS₂ heterostructure with graphite as the electrical contact. **(b)** Optical microscopic image of the heterostructure device. Electrode E₁ and E₂ are the source/drain contacts for the bottom Gr (green rectangular) layer, and E₃ and E₄ are electrically connected to the upper MoS₂ layer via the graphite electrode. **(c)** Simplified band diagram of the Gr/MoS₂ heterostructure in the lateral direction with graphite contacts (E₃ and E₄). **(d)** Optical Raman spectra of MoS₂ and the Gr/MoS₂ heterostructure. Blue-shift of A_{1g} peak indicates charge transfer from MoS₂ to Gr. **(e)** Band diagram of charge transfer direction at the Gr/MoS₂ heterointerface under different gate bias conditions. E_{vac}, qΦ_{MoS₂}, E_{C,MoS₂}, E_{F,MoS₂}, E_{V,MoS₂}, E_g, and E_{F,Gr} indicate the vacuum level, work function of MoS₂, conduction band of MoS₂, Fermi level of MoS₂, valence band of MoS₂, band-gap, and Fermi level of Gr, respectively.

2D heterostructures²⁹. Nevertheless, the limited carrier mobility of 2D materials still constrains their practical electronic applications, requiring different approaches.

We propose an ideal device platform based on a junction field-effect transistor (JFET) architecture featuring a Gr/MoS₂ heterointerface, where the carrier mobility of MoS₂ is enhanced by a factor of 10, while maintaining a high on/off current ratio of up to 10⁸ at room temperature. The Schottky barrier height (SBH) governs the carrier injection and carrier mobility in the Gr/MoS₂ heterojunction device. The low SBH regime at *n*-Gr/*n*-MoS₂ provides an additional Gr conduction path for MoS₂, leading to μ_{FE} ~ 100 cm² V⁻¹ s⁻¹, whereas the high SBH regime at *p*-Gr/*n*-MoS₂ blocks the contribution of Gr to MoS₂, leading to μ_{FE} ~ 10 cm² V⁻¹ s⁻¹, similar to that of the pure MoS₂-based device.

Results and discussions

To simultaneously achieve high carrier mobility and on/off current ratio, chemical vapor deposited (CVD) monolayer MoS₂ was intentionally stacked on top of a monolayer Gr strip that was also grown by CVD. Properly chosen mechanically exfoliated graphite flakes were employed as the source and drain contacts to protect the MoS₂ channel during metal deposition, thereby circumventing the Fermi level pinning effect³⁰. Fig. 1 presents a schematic of the conceptual Gr/MoS₂ heterostructure device with the graphite contact (Fig. 1a), the corresponding optical image (Fig. 1b), and the simplified band diagram (Fig. 1c) in the lateral direction with graphite contacts (E₃ and E₄), respectively. The detailed device fabrication procedure is described in the Methods section.

The optical Raman spectrum of the Gr/MoS₂ heterostructure (Fig. 1d) was acquired under argon atmosphere to prevent unintended oxidation or degradation. The E_{2g}¹ peaks (~389.4 cm⁻¹) of bare MoS₂ and the Gr/MoS₂ heterostructure are similarly positioned, implying negligible strain in the developed device. The blue-shift of the A_{1g} peak for the heterostructure with respect to that of MoS₂ indicates electron charge transfer from MoS₂ to Gr, as reported previously^{31,32}. The relevant band diagram depending on the carrier type of Gr (Fig. 1e) illustrates the vertical charge transfer direction based on the gate bias. In the *n*-Gr/*n*-MoS₂ regime, the electron population in *n*-Gr is transferred to *n*-MoS₂, leading to a negligible vertical SBH, whereas in the *p*-Gr/*n*-MoS₂ regime, electrons in *n*-MoS₂ are transferred to *p*-Gr, resulting in a large vertical SBH, as discussed later. This charge transfer model explains the key carrier transport mechanism in the developed heterostructure device associated with the vertical SBH at the Gr/MoS₂ heterojunction.

We first examined the back-gate bias (V_{BG})-dependent drain current (I_{DS}) of graphene (Device #1: Gr-Gr/MoS₂-Gr using metal electrodes E₁ and E₂) in the Gr/MoS₂ heterostructure as displayed in Fig. 2a. The transfer curves based on the drain-source voltage (V_{DS}) (Fig. 2b) demonstrate the ambipolar characteristics of the Gr in

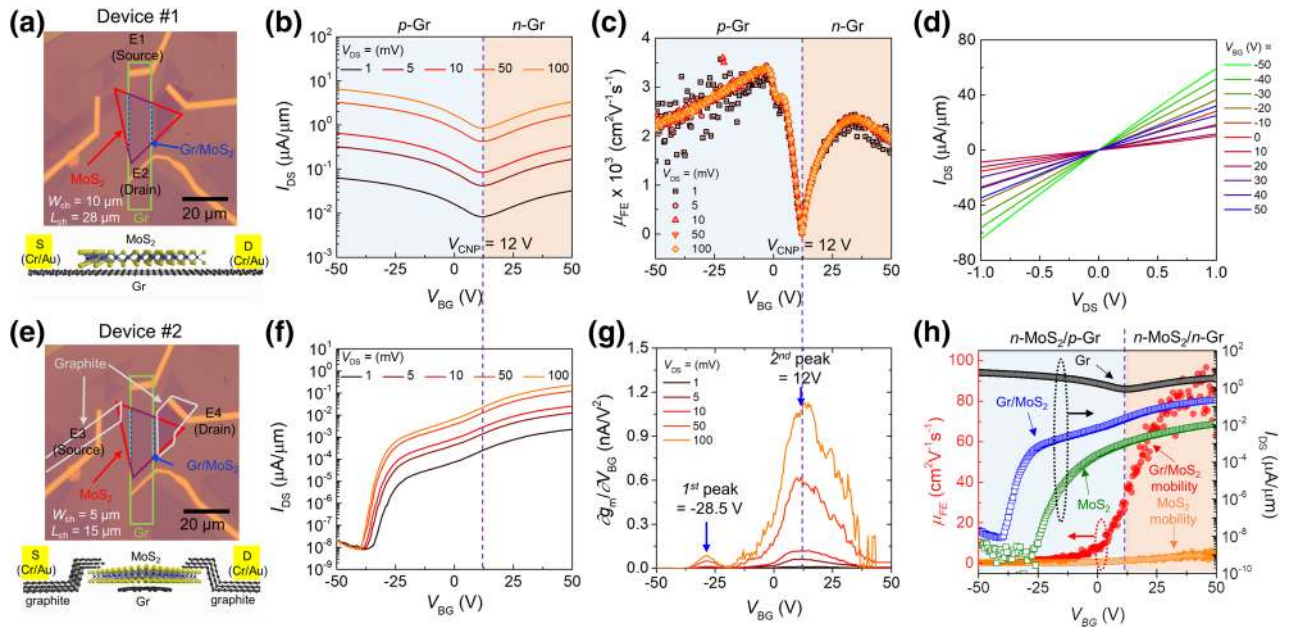


Figure 2. Electrical characteristics of Gr/MoS₂ heterostructure device. **(a–d)** Gr/MoS₂ heterostructure device employing Gr contact with E₁ (source) and E₂ (drain) (Device #1: Gr-Gr/MoS₂-Gr, $L/W = 28 \mu\text{m}/10 \mu\text{m}$). **(a)** Optical device image, V_{DS} -dependent **(b)** $I_{\text{DS}}-V_{\text{BG}}$ transfer curves ($V_{\text{CNP}} = 12 \text{ V}$) and **(c)** μ_{FE} , respectively. **(d)** V_{BG} -dependent $I_{\text{DS}}-V_{\text{DS}}$ output characteristic curves. **(e–g)** Gr/MoS₂ heterostructure device employing MoS₂ contact with E₃ and E₄ (Device #2: graphite-MoS₂-Gr/MoS₂-MoS₂-graphite, $L/W = 15 \mu\text{m}/5 \mu\text{m}$). **(e)** Optical device image, V_{DS} -dependent **(f)** transfer curves and **(g)** corresponding derivative of $g_{\text{m}} (= \partial I_{\text{DS}}/\partial V_{\text{BG}})$ curves. The 1st and 2nd peak positions of V_{BG} indicate the V_{FB} of MoS₂ and V_{CNP} of Gr. **(h)** Comparison of transfer curves of the three devices (Gr, MoS₂, Device #2) (right y-axis) and the μ_{FE} (left y-axis) of MoS₂ and Device #2 at $V_{\text{DS}} = 0.1 \text{ V}$. An enhancement of μ_{FE} ($\sim 100 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) is clearly observed in the $n\text{-MoS}_2/n\text{-Gr}$ regime.

Device #1. The charge neutrality point (V_{CNP}) is found at $V_{\text{BG}} = 12 \text{ V}$ regardless of V_{DS} . This positive V_{CNP} indicates dominant electron transfer from Gr to the positive fixed oxide traps in the SiO₂/Si substrate^{31,33,34}. The field-effect mobility is described as $\mu_{\text{FE}} = g_{\text{m}}(L/W) C_{\text{OX}}^{-1} V_{\text{DS}}^{-1}$, where g_{m} , L/W , and C_{OX} denote the transconductance ($= \partial I_{\text{DS}}/\partial V_{\text{BG}}$), channel length-to-width ratio, and oxide capacitance per unit area, respectively. The maximum μ_{FE} is $\sim 2,500 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for $n\text{-Gr}$ and $\sim 3,500 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for $p\text{-Gr}$ (Fig. 2c). The μ_{FE} range is similar to that of the bare Gr device on SiO₂³⁵. The collapse of the V_{DS} -dependent μ_{FE} clearly mirrors the negligible SBH effect. Further, super-linear $I_{\text{DS}}-V_{\text{DS}}$ output characteristic curves were obtained because of the high tunability of the Fermi-level of Gr (Fig. 2d), confirming the low SBH or nearly ohmic contact.

Investigation of the Gr/MoS₂ heterostructure device with the MoS₂ contact in the two graphite electrodes (Device #2: graphite-MoS₂-Gr/MoS₂-MoS₂-graphite using metal electrodes E₃ and E₄) (see Fig. 2e), shows a distinct current path difference comparing to Device #1. Because the conductivity of MoS₂ is much lower than that of Gr or the Gr/MoS₂ heterojunction in this case, the bare MoS₂ dominates the carrier transport of Device #2, in contrast with Device #1. The $I_{\text{DS}}-V_{\text{BG}}$ transfer curves (Fig. 2f) show clear switching behavior in the negative V_{BG} regime, originating primarily from the bare MoS₂ component in Device #2. Notably, in the positive V_{BG} regime, a hump associated with a rapid enhancement of I_{DS} appears, independent of V_{DS} . This strongly implies the presence of two different conduction mechanisms or conducting paths, similar to double-gate Si and junctionless Si transistors^{36,37}.

To gain insight into the carrier transport mechanism of Device #2 in detail, the flat-band voltage (V_{FB}) was determined from the second-derivative of the current (Fig. 2g)³⁸. The first peak at $V_{\text{BG}} = -28.5 \text{ V}$ is attributed to the turn-on voltage of MoS₂. The second peak at $V_{\text{BG}} = 12 \text{ V}$ is ascribed to the underlying Gr in Device #2, as confirmed by the identical V_{CNP} of Gr (Fig. 2c). The coincident position of the second hump for V_{FB} and V_{CNP} further rationalizes the presence of second conducting path Device #2. Since it has theoretically and experimentally been suggested that the vertical SBH of Gr/MoS₂ can be modulated by electrostatic doping^{39–42}, the bare MoS₂ consequently governs the carrier transport of the device exclusively when $V_{\text{BG}} \leq V_{\text{CNP}}$ (*i.e.*, $p\text{-Gr}/n\text{-MoS}_2$) at high vertical SBH. Another current path involving Gr in addition to MoS₂ is provided at $V_{\text{BG}} \geq V_{\text{CNP}}$ (*i.e.*, $n\text{-Gr}/n\text{-MoS}_2$) with low vertical SBH, leading to μ_{FE} for Device #2.

To have a better picture, μ_{FE} of heterostructure (Device #2) and bare MoS₂ (Device #5 with metal electrodes E₉ and E₁₀, see Supplementary Note 1 and Fig. S1) at $V_{\text{DS}} = 0.1 \text{ V}$ is compared together in Fig. 2h. In principle, the current onset voltage for Device #2 and Device #5 should be identical because the bare MoS₂ in both of them limits the carrier transport in the subthreshold regime. We ascribed this mismatched current onset voltage in Fig. 2h between the Gr/MoS₂ (Device #2) and MoS₂ (Device #5) to the different effective SBHs. While the MoS₂ (Device #5) has a Cr/Au contact to MoS₂, the MoS₂/Gr (Device #2) possesses a graphite contact to MoS₂, respectively. The maximum μ_{FE} of Device #5 ranges from 8 to 10 $\text{cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at $V_{\text{BG}} = 50 \text{ V}$, which reaches to 16–20 $\text{cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ after contact resistance correction (see Supplementary Fig. S2). Meanwhile, the μ_{FE} of Device #2 is

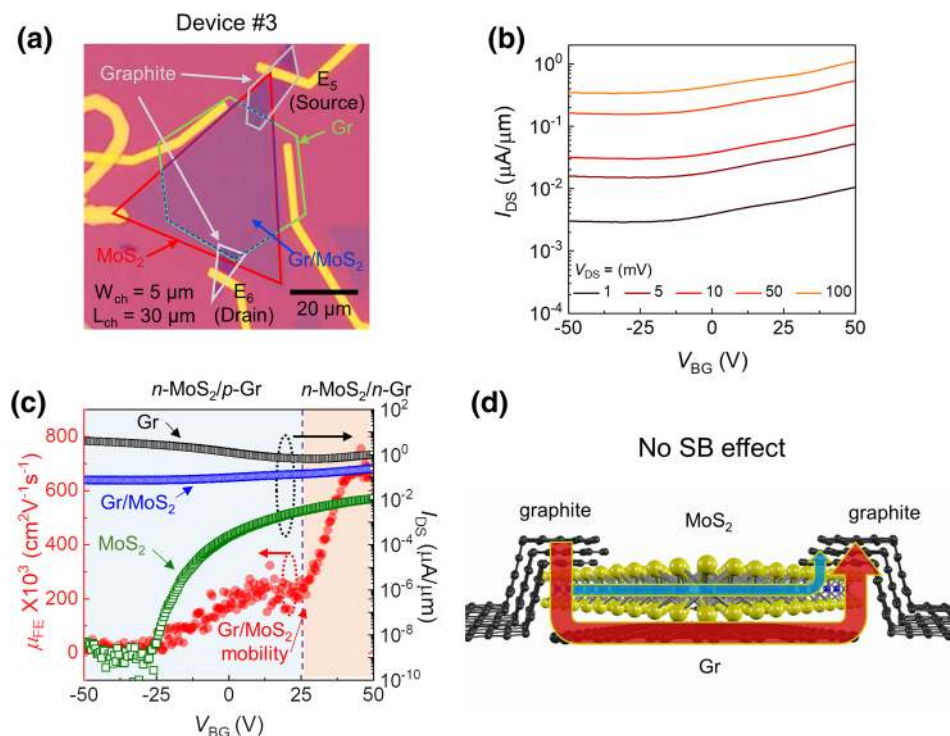


Figure 3. Vertical Gr/MoS₂ heterointerface device (the bottom Gr, MoS₂, and top graphite layers all overlapped). (a) Optical image of the vertical Gr/MoS₂ heterointerface device employing graphite contact with E₅ (source) and E₆ (drain) (Device #3: graphite-Gr/MoS₂-graphite, $L/W = 30 \mu\text{m}/5 \mu\text{m}$). (b) V_{DS} -dependent transfer curve shows low on/off current ratio. (c) Comparison of transfer curves of the three devices (right y-axis) and V_{BG} -dependent μ_{FE} (left y-axis) of Device #3 at $V_{\text{DS}} = 0.1 \text{ V}$. (d) Current path marked by arrows for this device structure. Direct tunneling (DT) from graphite to graphene through monolayer MoS₂ is suggested, as indicated by the red arrow. A small portion of the current flow along the MoS₂ layer is represented by the blue arrow.

slowly developed after the turn-on voltage of MoS₂ at $V_{\text{BG}} = -28.5 \text{ V}$ which is very similar to μ_{FE} of Device #5. This μ_{FE} behavior reflects clearly that the bare MoS₂ part in Device #2 limits the overall carrier transport in this V_{BG} regime. But, μ_{FE} rapidly increases after V_{FB} or V_{CNP} reaching μ_{FE} saturation ($\sim 100 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) at $V_{\text{BG}} = 50 \text{ V}$. This mobility enhancement is mainly ascribed to Gr with low SBH in the central $n\text{-Gr}/n\text{-MoS}_2$ heterojunction regime in Device #2. The high on/off current ratio of the device is further attributed to the bare MoS₂ region in the device. As a consequence, provided that the portion of bare MoS₂ in Device #2 shrinks as short as possible without losing the on/off current ratio, a further μ_{FE} improvement would expect via a device layout optimization. All devices were annealed at $T = 150 \text{ }^\circ\text{C}$ for 2 h in high vacuum chamber before the electrical measurement to eliminate effect of adsorbates and interface trap sites between 2D materials and dielectrics (See Supplementary Fig. S3). Hysteresis at Gr/MoS₂ interface was negligible from that of MoS₂ device.

To systematically identify the transport mechanism in the Gr/MoS₂ heterostructure, we constructed a device (Device #3: graphite-Gr/MoS₂-graphite using metal electrodes E₅ and E₆) with the two graphite electrodes overlapping the lower graphene layer, intentionally excluding the bare MoS₂ region (see Fig. 3a). It is worthy to underline that device structure of Device #3 is in contrast with that of Device #2 in particular for a spatial distance between graphite and underlying Gr (see Fig. 2e and Fig. 3d). The V_{DS} -dependent transfer curves (Fig. 3b) were obtained from the two graphite electrodes (E₅ and E₆). The lack of a high on/off current ratio is ascribed to the direct tunneling (DT) current across the atomically thin monolayer MoS₂ to the underlying graphene. The screening effect of the underlying graphene is another plausible underlying factor, resulting in weak gate modulation in the MoS₂ channel. From the transfer curves of individual Gr, MoS₂, and the Gr/MoS₂ heterostructure (Fig. 3c), the charge neutrality point of Gr was found at $V_{\text{BG}} = 27 \text{ V}$. General μ_{FE} behavior of Device #3 is very similar to that of Device #1 as shown in Fig. 2h. The high μ_{FE} ($\sim 800 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) of Device #3 in the $V_{\text{BG}} \geq V_{\text{CNP}}$ confirms clearly that the origin of high μ_{FE} is the underlying Gr. Further relevant electrical characteristics of Gr in Device #3 is discussed in Supplementary Fig. S4. In such a structure, even if the SBH at the Gr/MoS₂ interface is modulated by V_{BG} , the Schottky barrier effect is largely suppressed by direct tunneling at the interlayer distance, while a small portion of the total current flows along the MoS₂ channel based on the conductivity ratio between Gr and MoS₂ (Fig. 3d).

Now, we turn to evaluate the V_{BG} -dependent vertical SBH in the Gr/MoS₂ heterojunction at a given V_{BG} using Richardson's equation⁴³:

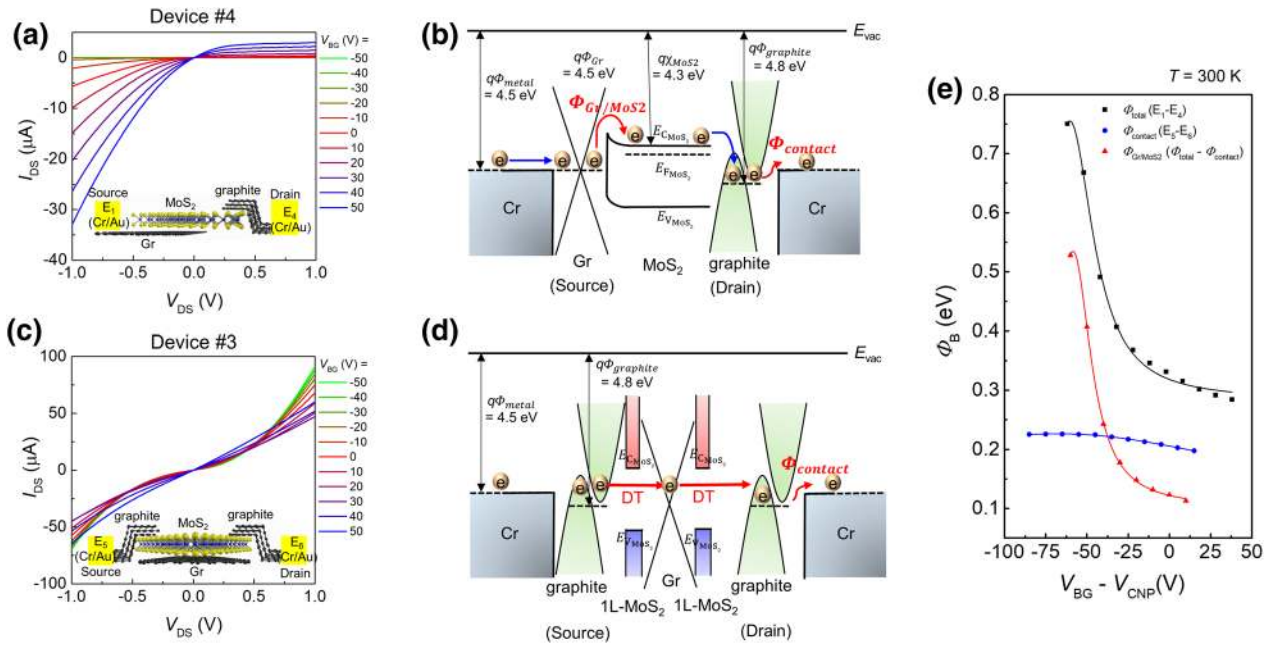


Figure 4. Determination of Schottky barrier height at Gr/MoS₂ heterointerface and band diagram. (a) V_{BG} -dependent output characteristic curves of Gr/MoS₂ heterostructure device contact with E_1 and E_4 (Device #4: Gr-Gr/MoS₂-graphite) and (b) corresponding band diagram. (c) V_{BG} -dependent output characteristic curves of Device #3 and (d) corresponding band diagram. (e) Effective SBH for Gr/MoS₂ heterointerface (Φ_{Gr/MoS_2}) as a function of $V_{BG} - V_{CNP}$.

$$I_{DS} = I_0 \exp(qV_{DS}/k_B T) \times \left[1 - \exp\left(-\frac{qV_{DS}}{k_B T}\right) \right] \quad (1)$$

where $I_0 = AA^*T^{3/2} \exp(-\frac{q\Phi_B}{k_B T})$, A is the junction area, A^* ($= 54 \text{ A K}^{-2} \text{ cm}^{-2}$) is the Richardson constant⁴⁴, n is the ideality factor, k_B is the Boltzmann constant, q is the electrical unit charge, Φ_B is the effective SBH, and T is the absolute temperature. Rearranging Eq. 1 gives:

$$\ln \left[\frac{I_{DS} \exp(qV_{DS}/k_B T)}{\exp(qV_{DS}/k_B T) - 1} \right] = \ln(I_0) + \frac{qV_{DS}}{nk_B T} \quad (2)$$

where n and Φ_B can be respectively determined from the slope and y -intercept of the $\ln[I_{DS} \times \exp(qV_{DS}/k_B T) / (\exp(qV_{DS}/k_B T) - 1)]$ versus the negative V_{DS} plot, (see Supplementary Note 2 and Fig. S5–S6 for the detailed procedures for SBH estimation).

The V_{BG} -dependent $I_{DS}-V_{DS}$ curves obtained from Device #4 (Gr-Gr/MoS₂-graphite using metal electrodes E_1 and E_4) are plotted in Fig. 4a. The asymmetric $I_{DS}-V_{DS}$ curve is mainly derived from the asymmetric contact barrier (graphene contact vs. graphite contact). The band alignment is illustrated, along with the two dominating Schottky barriers that contribute to the total SBH (Fig. 4b). One barrier is the SB at the Gr/MoS₂ heterojunction (Φ_{Gr/MoS_2}) and the other is the SB at the graphite/Cr contact junction ($\Phi_{contact}$). At the source terminal, the contact resistance at the Gr/Cr junction is negligible due to the low SB or nearly ohmic contact⁴², which is consistent with the linear output characteristics illustrated in Fig. 2d. The SB was clearly formed at the Gr/MoS₂ junction due to the difference in the work function of the two materials. Thereafter, electrons flow naturally from MoS₂ to graphite because the energy state of the conduction band of MoS₂ is higher (~ 4.3 eV) than that of graphite (~ 4.8 eV)³⁰. This implies that the SB at the MoS₂/graphite interface can be excluded from the total SBH estimation. Electrons are then thermally emitted to Cr by overcoming the SB at the graphite/Cr interface, where the contact barrier is formed. Therefore, the total SBH (Φ_{total}) obtained from this structure is attributed to the SB at the Gr/MoS₂ (Φ_{Gr/MoS_2}) heterojunction and graphite/Cr contact ($\Phi_{contact}$), where the total SBH can be expressed as: $\Phi_{total} = \Phi_{Gr/MoS_2} + \Phi_{contact}$. The $I_{DS}-V_{DS}$ curves of Device #3 present non-linear behavior in the V_{BG} regimes below V_{CNP} ($-50 \text{ V} \leq V_{BG} \leq 20 \text{ V}$) (see Fig. 4c). The low on/off current ratio of this device (see Fig. 3b) provides clear evidence of DT, where the electrons flow through the atomically thin monolayer MoS₂ to the underlying Gr, leading to a negligible contribution of Gr/MoS₂ SB to the total SBH. Accordingly, $\Phi_{contact}$ at the graphite/Cr interface could be regarded as Φ_{total} for this device structure, where $\Phi_{total} \simeq \Phi_{contact}$ (see Fig. 4d). Consequently, Φ_{Gr/MoS_2} can be deduced by subtracting $\Phi_{contact}$ from Φ_{total} . The V_{BG} -dependent Φ_{total} , $\Phi_{contact}$, and effective Φ_{Gr/MoS_2} at $T = 300$ K are directly compared in Fig. 4e. Increasing V_{BG} reduced Φ_{Gr/MoS_2} , when the value of V_{BG} was positive in the range of 528–116 meV, clearly indicating the large Fermi-level tunability of Gr via electrostatic gating.

Conclusion

In conclusion, we propose that the Gr/MoS₂ heterointerface can be employed as a high-performance electronic device. The high carrier mobility of 100 cm² V⁻¹ s⁻¹ of Gr/MoS₂ heterojunction device over 8–10 cm² V⁻¹ s⁻¹ of MoS₂ device is ascribed to the underlying Gr, which is activated when the low SBH is formed in the *n*-Gr/*n*-MoS₂ regime. On the other hand, the high on/off current ratio of ~10⁸ is attributed to the bare MoS₂ region in the heterostructure device. Furthermore, we demonstrate that the high tunability of the Fermi level of Gr allows to control the SBH at the Gr/MoS₂ interface, resulting in distinctive carrier conduction features of the Gr/MoS₂ heterojunction device, and confirming consequently two different conduction mechanisms.

Methods

Material synthesis. Monolayer Gr flakes were first synthesized on a 100 μm thick Cu foil (111) in a chemical vapor deposition (CVD) chamber under hydrogen with a low concentration of methane (0.1%, balance Ar gas) as a carbon source. The synthesized graphene with the polymer supporting layer was softly detached by bubble interception in sodium hydroxide solution and rinsed thrice with distilled water to eliminate contaminants during the chemical wet etching process. The graphene layer was then transferred onto a 300 nm-thick SiO₂ substrate. Reactive ion etching (RIE, AFS-4RT; O₂ plasma power of 30 mW; flow rate of 20 sccm at 10⁻³ torr) was used to define the channel area of Gr. Monolayer MoS₂ flakes were grown in a separate CVD chamber and transferred onto graphene strips by a water-assisted transfer method⁴⁵. Thereafter, the exfoliated graphite flakes (~5 nm) were placed on the Gr/MoS₂ heterostructure by the aligned transfer technique, in a glove box. Finally, the source/drain metal electrodes were patterned on the polymer resist via electron beam lithography, followed by metal deposition. All processes were completed within 24 h to maintain consistency in terms of the device reliability and reproducibility.

Device fabrication. The devices were fabricated by a series of wet- and dry-cooperative transfer methods. The channel materials (single-crystal graphene and MoS₂) were both prepared by CVD^{35,46}. The graphene synthesized on copper was firstly delaminated by electrochemical bubbling and then transferred onto the target 300 nm SiO₂/Si substrates⁴⁷. Thereafter, the MoS₂ grown on the SiO₂/Si substrate was detached by chemical etching⁴⁸. After rinsing copiously with deionized water, the MoS₂ flakes carried by the polymethyl methacrylate (PMMA) transfer film were assembled on the dry transfer holder. The two channels were aligned in the desired positions, and MoS₂ was brought into contact with the graphene and held at 140 °C for 5 min, thus isolating the PMMA film from the transfer holder. The electrical contact of MoS₂ was improved by using graphite, which was mechanically exfoliated onto the PMMA coated 300 nm SiO₂/Si substrates and then deposited on the designed positions inside or outside the overlapped graphene/MoS₂ by the conventional PMMA supporting layer method⁴⁹. Subsequently, the source and drain for the graphene channel and the contact to the graphite electrodes were patterned by e-beam lithography, followed by vapor deposition of 5/50 nm Cr/Au. For the strip graphene channel, an additional patterning step was performed by photolithography prior to O₂-plasma dry etching.

Optical and electrical characterization. The optical Raman spectrum of the developed Gr/MoS₂ heterointerface device was acquired under 532 nm laser excitation with a power of 12 μW (Witec Alpha 300) in an argon-filled glove box system, where the oxygen and moisture levels were kept below 1 ppm to prevent undesired oxidation and aging of the samples. Electrical measurements were carried out using a commercial source measurement unit (Keithley 4200-SCS) under low-vacuum conditions (~10⁻³ torr) in a vacuum probe station, at room temperature.

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Author contributions

T.K., S.F., Y.H.L., and M.-K.J. conceived the research and designed the experiment. T.K. and S.F. performed most of the experiments including device fabrication, characterization and data analysis. S.L. prepared the CVD graphene and MoS₂ samples. T.K., S.F., and M.-K.J. illustrated all the images and plots in main figures and supplementary information. T.K., S.F., M.-K.J., and Y.H.L. analyzed the results and wrote the manuscript. All authors discussed the results and commented on the manuscript.

Competing interests

The authors declare no competing financial interests.

Additional information

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