Tuning Schottky Barrier of Single-Layer MoS$_2$ Field-Effect Transistors with Graphene Electrodes

A-Rang Jang

Division of Electrical, Electronic and Control Engineering, Kongju National University, Cheonan 31080, Korea; arjang@kongju.ac.kr

Abstract: Two–dimensional materials have the potential to be applied in flexible and transparent electronics. In this study, single-layer MoS$_2$ field-effect transistors (FETs) with Au/Ti–graphene heteroelectrodes were fabricated to examine the effect of the electrodes on the electrical properties of the MoS$_2$ FETs. The contact barrier potential was tuned using an electric field. Asymmetrical gate behavior was observed owing to the difference between the MoS$_2$ FETs, specifically between the MoS$_2$ FETs with Au/Ti electrodes and those with graphene electrodes. The contact barrier of the MoS$_2$ FETs with Au/Ti electrodes did not change with the electric field. However, the contact barrier at the MoS$_2$–graphene interface could be modulated. The MoS$_2$ FETs with Au/Ti–graphene electrodes exhibited enhanced on/off ratios (~10$^2$ times) and electron mobility (~2.5 times) compared to the MoS$_2$ FETs with Au/Ti electrodes. These results could improve the understanding of desirable contact formation for high-performance MoS$_2$ FETs and provide a facile route for viable electronic applications.

Keywords: graphene; molybdenum disulfide; Schottky barrier

1. Introduction

Two–dimensional (2D) materials have attracted significant attention as potential candidates for next-generation electronics [1,2]. Graphene is considered to be one of the most promising 2D materials because of its unique electrical, mechanical, and optical properties. However, the widespread use of graphene in viable electronic device applications is limited by its zero-bandgap property, which considerably decreases the on/off ratio [3–6]. To overcome this limitation, graphene nanoribbons [7,8], bilayer graphene [9–12], and modified device architectures, such as vertical tunneling transistors, have been developed [13,14]. Although these devices have improved the on/off ratio, other desirable properties, such as mobility and current density, have deteriorated. Thus, there is an urgent requirement for 2D materials, including transition metal dichalcogenides (TMDs), with an appropriate bandgap and reasonable mobility to replace graphene. MoS$_2$ is one of the most promising TMDs because its bandgap is 1.3–1.8 eV depending on the number of layers. Single-layer MoS$_2$ films have a direct bandgap of 1.8 eV, whereas multilayer MoS$_2$ films have an indirect bandgap of 1.2 eV [15–17]. Owing to these unique properties, MoS$_2$ has been intensively studied for electronic and optoelectronic applications. In recent years, it has become possible to synthesize large-area single-layer MoS$_2$ via chemical vapor deposition (CVD) [18–22]. This has provided a major opportunity for next-generation electronic device applications. However, the contact barrier issue must be studied for electronic device applications of 2D materials [23,24]. Moreover, the performance of MoS$_2$ field-effect transistors (FETs) is lower than the theoretically predicted performance [25]. This discrepancy has been explained on the basis of charged impurities and localized states in MoS$_2$ [26–28]. Dominant scattering processes decrease carrier mobility. In addition, the contact at MoS$_2$–metal electrode interfaces is a critical issue. A tunneling barrier that is formed at the interface of a metal contact in an MoS$_2$ device [29] significantly reduces carrier mobility in single–layer MoS$_2$. This is one of the main reasons for the poor performance of single-layer MoS$_2$ FETs.
Sulfur atoms mediate the hybridization between a contact metal and Mo atoms, resulting in the tuning of the bandgap [30]. Furthermore, the bandgap of single-layer MoS$_2$ can be determined by the strength of the Mo–S covalent bonding [31]. Therefore, a systematic study of the effects of electrode materials on the performance of MoS$_2$ FETs can help resolve this critical issue and find a reliable method of improving the electrical properties of MoS$_2$ FETs. A charge accumulation region forms at metal–MoS$_2$ interfaces when a metal contact is used. This generally leads to the formation of an interface electric dipole, which modifies the interface band alignment [30]. This results in poor contact and an unexpected contact barrier between the metal and MoS$_2$. Owing to the challenges associated with metal electrodes, graphene has been considered as a suitable electrode material for MoS$_2$ FETs. Graphene and single-layer MoS$_2$ bond via van der Waals (vdW) forces, thereby creating a pristine interface. Furthermore, the contact barrier between graphene and MoS$_2$ can be controlled by tuning the work function of graphene (4.5 eV), which is quite similar to that of MoS$_2$ [32]. As the work function of graphene can be readily tuned by applying an electric field, graphene-based heterostructures have recently been studied in electronic devices [33–36]. For instance, the Schottky barrier formed between graphene and silicon can be tuned by approximately 200 meV as a function of the gate voltage [13]. Therefore, the contact barrier between graphene and MoS$_2$ can be tuned by applying an electric field.

Herein, we report high-performance single–layer MoS$_2$ FETs with graphene electrodes that exhibit a considerable enhancement in the on/off ratio (~10$^2$ times) and electron mobility (~2.5 times) compared to the MoS$_2$ FETs with Au/Ti electrodes. We show that the contact barrier potential of the MoS$_2$ FETs with graphene electrodes can be effectively tuned by applying an electric field. The work function of graphene becomes higher than that of MoS$_2$ at a negative bias voltage, resulting in the formation of a Schottky barrier. Similarly, the work function of graphene becomes lower than that of MoS$_2$ at a positive bias voltage, resulting in the formation of an ohmic barrier. The contact barrier between MoS$_2$ and graphene can be easily tuned using graphene electrodes. Thus, the on/off ratio and electron mobility of the MoS$_2$ FET can be improved by tuning the contact barrier.

2. Materials and Methods

2.1. Graphene Growth and Transfer

Graphene was synthesized on a copper foil (99.8% purity, 0.025 mm thick, Alfa Aesar, Haverhill, MA, USA) using CVD at a growth temperature of 1050 °C with 10 sccm of H$_2$ and 15 sccm of CH$_4$ [37]. Then, the full side of the foil that faced upwards during synthesis was covered with poly(methyl methacrylate) (PMMA) (AR–N 7500.18, Allresist, Strausberg, Germany) via spin coating (4000 rpm for 60 s). The remaining graphene on the Cu foil that faced downwards during the synthesis was removed using O$_2$ plasma (Femto, Diener, Ebhausen, Germany). The Cu foil was completely etched using 0.1 M ammonia persulfate (Sigma Aldrich, St. Louis, MO, USA). The PMMA/graphene layer was washed several times with fresh deionized water. Finally, the PMMA/graphene layer floated on the surface of the water, and it was transferred to a SiO$_2$ substrate. The transferred PMMA/graphene layer was patterned using electron beam lithography (Nanobeam nB4, NBL, Cambridge, UK) as shown in Figure S1a.

2.2. Fabrication of the MoS$_2$ Field-Effect Transistor

As shown in Figure S1b, single-layer MoS$_2$ was prepared via mechanical exfoliation from a bulk MoS$_2$ flake (429ML–AB, SPI Supplies, West Chester, PA, USA). To fabricate MoS$_2$ FET with graphene electrode, a dry transfer process was employed [38]. Patterned graphene was transferred onto single–layer MoS$_2$ flake after the alignment position using a micromanipulator (NMO–203, Narishige, Tokyo, Japan) (Figure 1a). Au/Ti electrodes were patterned using electron beam lithography with a positive electron beam resist (ARP 671.04, Allresist, Strausberg, Germany). This was followed by metal deposition (Ti (5 nm)/Au (45 nm)) and a lift-off process.
were patterned using electron beam lithography with a positive electron beam resist (AR–P 671.04, Allresist, Strausberg, Germany). This was followed by metal deposition (Ti (5 nm)/Au (45 nm)) and a lift-off process.

Figure 1. (a) Schematics illustration of the fabrication process for MoS$_2$ FET with graphene electrode. Raman spectroscopy of mechanically exfoliated single-layer MoS$_2$ (red), chemical vapor deposition (CVD)-grown graphene on single-layer MoS$_2$ (blue), and CVD-grown graphene (green). (b) MoS$_2$ region, and (c) graphene region of the Raman spectrum (the insert of (b) shows the Raman analysis position by cross mark).

2.3. Characterization of the MoS$_2$ Thin Film and Field–Effect Transistor

Mechanical exfoliation was employed to extract high-quality single-layer MoS$_2$ from bulk MoS$_2$ [3]. Then, single-layer MoS$_2$ was transferred onto a silicon wafer with a 300 nm thick SiO$_2$ layer. Raman spectroscopy (Alpha 300R, WiTec, Ulm, Germany) was used to determine the number of layers of MoS$_2$ [39]. The Raman spectrum of MoS$_2$ revealed a peak spacing of less than 20 cm$^{-1}$ between the E$_{2g}$ and A$_{1g}$ modes, indicating that single-layer MoS$_2$ was formed. A 532 nm laser with a power of 1 mW was used as an excitation source. The exposure time was 1 s, and calibration was performed using a reference Si peak position of 520 cm$^{-1}$. The fabricated MoS$_2$ FETs were loaded into a vacuum chamber.
3. Results and Discussion

Figure 1b,c show the Raman spectra of single-layer MoS$_2$, graphene on single-layer MoS$_2$, and graphene, respectively. The MoS$_2$ and graphene/MoS$_2$ layers exhibit typical single-layer Raman active modes (~18.27 cm$^{-1}$ of frequency difference between E$_{2g}$ and A$_{1g}$), and the 2D/G ratio of graphene is about 4.06. Therefore, it can be noted that exfoliated MoS$_2$ flake has a formation of single layer. The Dirac point of intrinsic graphene is at zero gate voltage, the work function of which is approximately 4.5 eV [32]. As shown in Figure S2, the Dirac point of the CVD-grown graphene electrode was measured at 22.5 V, owing to the hole doping originated from both coupling with dielectric layer of SiO$_2$ and exposure to oxygen and moisture [40]. The schematic of the band structure of graphene and MoS$_2$ is shown in Figure 2. Graphene and single-layer MoS$_2$ were bonded via weak vdW forces. However, MoS$_2$ and the metal interfaces formed covalent interactions, causing a change in the electronic structure [30]. This led to unexpected contact resistance. Three different types of single-layer MoS$_2$ FETs were fabricated to investigate the effects of the graphene electrode. The first was a single-layer MoS$_2$ FET with a Au/Ti–graphene heteroelectrode, as shown in Figure 3a. Highly boron-doped Si (resistance of 0.001 $\Omega$) with a 300 nm thick SiO$_2$ layer was used as the substrate. The channel length and width of the mechanically exfoliated MoS$_2$ used in the single-layer MoS$_2$ FET were ~2 $\mu$m and ~4 $\mu$m, respectively. Figure 3b shows the asymmetric $I_{DS}$–$V_{DS}$ output characteristics of the single-layer MoS$_2$ FET with the Au/Ti–graphene heteroelectrode without the gate voltage. Different contact barriers were generated according to the contact material. An ohmic contact was formed between single-layer MoS$_2$ and Au/Ti. A Schottky contact was formed between single-layer MoS$_2$ and graphene. Figure 3c shows the $I_{DS}$–$V_{GS}$ transfer characteristics for a positive source–drain voltage ($V_{DS}$). The on/off ratio and electron mobility (graphene in the heteroelectrode) were >10$^5$ and ~3.2 cm$^2$/V·s, respectively. Figure 3d shows the $I_{DS}$–$V_{GS}$ transfer characteristics for a negative drain voltage. The on/off ratio and electron mobility (Au/Ti in the heteroelectrode) were >10$^5$ and ~1.2 cm$^2$/V·s, respectively. These results indicated that graphene could be used as an ideal electrode in a single-layer MoS$_2$ FET. Mobility was calculated using the following equation: $\mu = g_m \times L / C_g \times V_D \times W$; where $g_m$ is the transconductance, $V_D$ is the source–drain voltage, $L$ is the channel length, $W$ is the channel width, and $C_g$ is the capacitance of 300 nm thick SiO$_2$. The MoS$_2$ FET with the Au/Ti electrodes exhibited ohmic contact behavior, whereas the MoS$_2$ FET with the graphene electrodes exhibited Schottky contact behavior. Multilayer MoS$_2$ FETs with exfoliated graphene electrodes also showed ohmic contact behavior [41]. The work function of graphene was approximately 4.5 eV because mechanically exfoliated graphene was almost pure with no doping. Therefore, the single-layer MoS$_2$ FET with the graphene electrodes exhibited a Schottky barrier without a gate bias voltage. However, the work function of graphene was electrostatically adjusted to approximately 300 meV for single-layer graphene by tuning the Fermi level ($E_F$) by changing the gate voltage by 50 V [32]. The work function of graphene decreased at a positive gate bias voltage. Figure 4 shows the $I_{DS}$–$V_{DS}$ characteristics of the single-layer MoS$_2$ FET as a function of the back-gate voltage. The Schottky barrier between graphene and single-layer MoS$_2$ was enhanced at a negative gate voltage; thus, current could not flow in the negative gate voltage direction (Figure 4a). As the gate was positively biased, the Schottky barrier between graphene and single-layer MoS$_2$ decreased, and the contact barrier between single-layer MoS$_2$ and Au/Ti did not change. The $I_{DS}$–$V_{DS}$ output characteristics of the single-layer MoS$_2$ FET with the Au/Ti–graphene heteroelectrode (green solid line) showed almost similar with linear (red dashed line) at a gate voltage of 20 V because the work function of graphene became similar to that of single-layer MoS$_2$ (Figure 4c). As the gate voltage exceeded 20 V, the current level
The electrical properties of the single-layer MoS$_2$ FET were enhanced using the graphene electrodes. A Schottky barrier was formed at the interface of graphene and MoS$_2$ in the current-off region; thus, there was no leakage current. However, an ohmic barrier was formed at the interface between graphene and MoS$_2$ in the current-on region. Therefore, the on/off ratio and electron mobility of single-layer MoS$_2$ were high. The on/off ratio and electron mobility of single-layer MoS$_2$ were compared with those of homogeneous electrodes. A single-layer MoS$_2$ FET with the graphene electrodes was fabricated, and its electrical properties were measured. Figure 5a shows the schematic of the single-layer MoS$_2$ FET with the graphene electrodes, and Figure 5b shows its $I_{DS}$-$V_g$ transfer characteristics. The $I_{DS}$-$V_{DS}$ output characteristics shown in Figure 5c confirmed that a Schottky barrier was formed. When an increasingly positive back-gate bias was applied to the single-layer MoS$_2$ FET with the graphene electrodes, the Schottky barrier was slightly modified into a clear ohmic contact, as shown in Figure 5d. The on/off ratio and electron mobility were $>10^5$ and $\sim 2.3$ cm$^2$/V·s, respectively. A single-layer MoS$_2$ FET with the Au/Ti electrodes was fabricated, and its electrical properties were measured for comparison. Figure S2a shows the schematic of the single-layer MoS$_2$ FET with the Au/Ti electrodes, and Figure S2b shows its $I_{DS}$-$V_g$ transfer characteristics. The on/off ratio and electron mobility were $>10^3$ and $\sim 0.9$ cm$^2$/V·s, respectively. The on/off ratio and electron mobility of the single-layer MoS$_2$ FET with the graphene electrodes were $\sim 10^2$ and $\sim 2.5$ times higher than those of the single-layer MoS$_2$ FET with the Au/Ti electrodes, respectively.

To study the barrier height of the MoS$_2$ FET with graphene electrodes, current-voltage characteristics (Figure 6a) and $I_{DS}$-$V_g$ transfer characteristics (Figure 6b) were measured at different temperatures. The 2D thermionic emission equation was used to describe the electrical transport behavior of Schottky contacted MoS$_2$ devices [41,42].

$$I_{DS} = A A^{*}_{2D} T^{3/2} \exp \left[ \frac{q}{k_B T} \left( \Phi_B - \frac{V_{DS}}{n} \right) \right]$$

where $A$ is the contact area of the junction, $A^{*}_{2D}$ is the two–dimensional equivalent Richardson constant, $q$ is the magnitude of the electron charge, $\Phi_B$ is the Schottky barrier height, $k_B$ is the Boltzmann constant, $n$ is the ideality factor, and $V_{DS}$ is the drain-source bias. Instead of the typical Arrenhius plot, $\ln(I_d/T^2)$ versus $1000/T$ for three-dimensional semiconductors, $\ln(I_d/T^{3/2})$ versus $1000/T$ was used because here the semiconducting channel is two-dimensional. The $\ln(I_d/T^{3/2})$ versus $1000/T$ of MoS$_2$ FET with graphene electrodes for various values of $V_g$ is shown in Figure 6c. Based on Equation (1), the height of the Schottky barrier can be deduced as Equation (2):

$$y_{intercept} = -\frac{q}{1000k_B} \Phi_B$$

In the MoS$_2$ FET with graphene electrodes, the Schottky barrier is decreased dramatically—from 51.5 meV to 0 meV—with the back gate voltage changing from $-7.5$ to $12.5$ V, as shown in Figure 6d. The change of the Schottky barrier in the MoS$_2$ FET with graphene electrodes comes from changes in work function of graphene.
MoS2 FET were enhanced using the graphene electrodes. A Schottky barrier was formed at the interface of graphene and MoS2 in the current-off region; thus, there was no leakage current. However, an ohmic barrier was formed at the interface between graphene and MoS2 in the current-on region. Therefore, the on/off ratio and electron mobility of single-layer MoS2 were high. The on/off ratio and electron mobility of single-layer MoS2 were compared with those of homogeneous electrodes. A single-layer MoS2 FET with the graphene electrodes was fabricated, and its electrical properties were measured. Figure 5a shows the schematic of the single-layer MoS2 FET with the graphene electrodes, and Figure 5b shows its I DS–Vg transfer characteristics. The I DS–VDS output characteristics shown in Figure 5c confirmed that a Schottky barrier was formed. When an increasingly positive back-gate bias was applied to the single-layer MoS2 FET with the graphene electrodes, the Schottky barrier was slightly modified into a clear ohmic contact, as shown in Figure 5d. The on/off ratio and electron mobility were >10^5 and ~2.3 cm^2/V∙s, respectively. A single-layer MoS2 FET with the Au/Ti electrodes was fabricated, and its electrical properties were measured for comparison. Figure S 2a shows the schematic of the single-layer MoS2 FET with the Au/Ti electrodes, and Figure S 2b shows its I DS–Vg transfer characteristics. The on/off ratio and electron mobility were >10^3 and ~0.9 cm^2/V∙s, respectively. The on/off ratio and electron mobility of the single-layer MoS2 FET with the graphene electrodes were ~10^2 and ~2.5 times higher than those of the single-layer MoS2 FET with the Au/Ti electrodes, respectively. To study the barrier height of the MoS2 FET with graphene electrode, current-voltage characteristics (Figure 6a) and I DS–Vg transfer characteristics (Figure 6b) were measured at different temperatures. The 2D thermionic emission equation was used to describe the electrical transport behavior of Schottky contacted MoS2 devices [41,42].

**Figure 2.** Schematic band diagram of intrinsic graphene, CVD-grown graphene, and single-layer MoS2.

**Figure 3.** Schematic and electrical properties of MoS2 field-effect transistor (FET) with hetero-electrodes. (a) Schematic of MoS2 FET with hetero-electrodes; (b) I DS–VDS output characteristics; (c) I DS–Vg transfer characteristics at VDS = 0.5 V; (d) I DS–Vg transfer characteristics at VDS = −0.5 V.
Figure 4. Band diagrams and electrical properties of the MoS$_2$ FET with Au–graphene heteroelectrode at different gate voltages ((a) $-20$ V, (b) 0 V, (c) 20 V, and (d) 40 V).
Figure 5. Schematic and electrical properties of MoS$_2$ FET with graphene electrodes. (a) Schematic of MoS$_2$ FET with graphene electrodes; (b) $I_{DS}$–$V_g$ transfer characteristics at $V_{DS} = 0.5$ V; (c) $I_{DS}$–$V_{DS}$ output characteristics; (d) $I_{DS}$–$V_{DS}$ characteristics at different gate bias voltages.

Figure 6. Temperature-dependent electrical transport of the MoS$_2$ FET with graphene electrode. (a) Current voltage characteristics and (b) $I_{DS}$–$V_g$ transfer characteristics from 30 K to 230 K, for source-drain bias voltage of 0.1 V. (c) Linear fit of the Arrhenius plot, $\ln \left( \frac{I_D}{T^{3/2}} \right)$ vs. 1000/$T$ as function of $V_g$. (d) The Schottky barrier of MoS$_2$ FET with graphene electrode depends on the gate voltage.
4. Conclusions

This work demonstrates the enhancement of the electrical properties of an MoS\textsubscript{2} FET with graphene electrodes by tuning the contact barrier using an electric field. The MoS\textsubscript{2} FET with a Au/Ti-graphene heterocontact shows a clear change in the contact barrier between MoS\textsubscript{2} and graphene. A Schottky barrier and ohmic barrier exist in the off and on states of the MoS\textsubscript{2} FET with the graphene electrodes. The on/off ratio and electron mobility of the MoS\textsubscript{2} FET with the graphene electrodes are 10\textsuperscript{2} and 2.5 times higher than those of the MoS\textsubscript{2} FET with the Au/Ti electrodes, respectively. The Schottky barrier between MoS\textsubscript{2} and graphene is decreased from 51.5 to 0 meV by the back gate voltage. The implication of these results could be of great importance in better understanding the desirable contact formation for high performance MoS\textsubscript{2} FETs. This FET may be promising for electronic device applications based on next-generation 2D materials.

**Supplementary Materials:** The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/nano12173038/s1, Figure S1: Schematic illustration of sample preparation process; Figure S2: Electrical properties of chemical-vapor-deposition-grown graphene; Figure S3: Schematic and electrical properties of MoS\textsubscript{2} field-effect transistor with Au/Ti electrodes.

**Funding:** This research was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIT) (2021R1A4A1031900, 2021R1F1A3049729) and Nano-Material Technology Development Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Science, ICT and Future Planning (N220212001).

**Institutional Review Board Statement:** Not applicable.

**Informed Consent Statement:** Not applicable.

**Data Availability Statement:** Not applicable.

**Conflicts of Interest:** The author declare no conflict of interest.

**References**

1. Glavin, N.R.; Rao, R.; Varshney, V.; Bianco, E.; Apte, A.; Roy, A.; Ringe, E.; Ajayan, P.M. 2D materials: Emerging applications of elementary 2D materials. *Adv. Mater.*, 2020, 32, 1904302. [CrossRef] [PubMed]

2. Cheng, Z.; Cao, R.; Wei, K.; Yao, Y.; Liu, X.; Kang, J.; Dong, J.; Shi, Z.; Zhang, H.; Zhang, X. 2D materials enabled next-generation integrated optoelectronics: From fabrication to applications. *Adv. Sci.*, 2021, 8, 2003834. [CrossRef]

3. Novoselov, K.S.; Geim, A.K.; Morozov, S.V.; Jiang, D.; Zhang, Y.; Dubonos, S.V.; Grigorieva, I.V.; Firsov, A.A. Electric field effect in atomically thin carbon films. *Science*, 2004, 306, 666–669. [CrossRef] [PubMed]

4. Novoselov, K.S.; Geim, A.K.; Morozov, S.V.; Jiang, D.; Katsnelson, M.I.; Grigorieva, I.V.; Dubonos, S.V.; Firsov, A.A. Two-dimensional gas of massless Dirac fermions in graphene. *Nature*, 2005, 438, 197–200. [CrossRef] [PubMed]

5. Zhang, Y.; Tan, Y.-W.; Sromer, H.L.; Kim, P. Experimental observation of the quantum Hall effect and Berry’s phase in graphene. *Nature*, 2005, 438, 201–204. [CrossRef]

6. Meric, I.; Han, M.Y.; Young, A.F.; Ozylmaz, B.; Kim, P.; Shepard, K.L. Current saturation in zero-bandgap, top-gated graphene field-effect transistors. *Nat. Nanotechnol.*, 2008, 3, 654–659. [CrossRef]

7. Li, X.; Wang, X.; Zhang, L.; Lee, S.; Dai, H. Chemically Derived. Ultrasmooth graphene nanoribbon semiconductors. *Science*, 2008, 319, 1229–1232. [CrossRef]

8. Wang, X.; Ouyang, Y.; Li, X.; Wang, H.; Guo, J.; Dai, H. Room-temperature all-semiconducting sub-10-nm graphene nanoribbon field-effect-transistors. *Phys. Rev. Lett.*, 2008, 100, 206803. [CrossRef]

9. Jing, L.; Velasco, J.; Kratz, P.; Liu, G.; Bao, W.; Bockrath, M.; Lau, C.N. Quantum transport and field-induced insulating states in bilayer graphene pnp junctions. *Nano Lett.*, 2010, 10, 4000–4004. [CrossRef]

10. Castro, E.V.; Novoselov, K.S.; Morozov, S.V.; Peres, N.M.R.; Santos, J.M.B.L.; Nilsson, J.; Guinea, F.; Geim, A.K.; Neto, A.H.C. Biased bilayer graphene: Semiconductor with a gap tunable by the electric field effect. *Phys. Rev. Lett.*, 2007, 99, 216802. [CrossRef]

11. Taychatanapat, T.; Jarillo-Herrero, P. Electronic transport in Dual-gated bilayer graphene at large displacement fields. *Phys. Rev. Lett.*, 2010, 105, 166601. [CrossRef] [PubMed]

12. Xia, F.; Farmer, D.B.; Lin, Y.-M.; Avouris, P. Graphene field-effect transistors with high On/Off current ratio and large transport band gap at room temperature. *Nano Lett.*, 2010, 10, 715–718. [CrossRef] [PubMed]

13. Yang, H.; Heo, J.; Park, S.; Song, H.J.; Seo, D.H.; Byun, K.-E.; Kim, P.; Yoo, I.; Chung, H.-J.; Kim, K. Graphene barrister, a triode device with a gate-controlled Schottky barrier. *Science*, 2012, 336, 1140–1143. [CrossRef] [PubMed]
14. Britnell, L.; Gorbachev, R.V.; Jalil, R.; Bello, B.D.; Schedin, F.; Mishchenko, A.; Georgiou, T.; Katsnelson, M.I.; Eaves, L.; Morozov, S.V.; et al. Field-Effect Tunneling Transistor Based on Vertical Graphene Heterostructures. *Science* 2012, 335, 947–950. [CrossRef] [PubMed]

15. Mak, K.F.; Lee, C.; Hone, J.; Shan, J.; Heinz, T.F. Atomically thin MoS$_2$: A new direct-gap semiconductor. *Phys. Rev. Lett.* 2010, 105, 136805. [CrossRef]

16. Eda, G.; Yamaguchi, H.; Voiry, D.; Fujita, T.; Chen, M.; Chhowalla, M. Photoluminescence from Chemically Exfoliated MoS$_2$. *Nano Lett.* 2011, 11, 5111–5116. [CrossRef]

17. Lee, H.S.; Min, S.-W.; Chang, Y.-G.; Park, M.K.; Nam, T.; Kim, H.; Kim, J.H.; Ryu, S.; Im, S. MoS$_2$ Nanosheet Phototransistors with Thickness-Modulated Optical Energy Gap. *Nano Lett.* 2012, 12, 3695–3700. [CrossRef]

18. Najmaei, S.; Liu, T.; Zhou, W.; Zou, X.; Shi, G.; Lei, S.; Yakobson, B.I.; Idrobo, J.-C.; Ajayan, P.M.; Lou, J. Vapour phase growth and grain boundary structure of molybdenum disulphide atomic layers. *Nat. Mater.* 2013, 12, 754–759. [CrossRef]

19. Lee, Y.-H.; Zhang, X.-Q.; Zhang, W.; Chang, M.-T.; Lin, C.-T.; Chang, K.-D.; Yu, Y.-C.; Wang, J.T.-W.; Chang, C.-S.; Li, L.-J.; et al. Synthesis of large-area MoS$_2$ atomic layers with chemical vapor deposition. *Adv. Mater.* 2012, 24, 2320–2325. [CrossRef]

20. Lee, Y.-H.; Yu, L.; Wang, H.; Fang, W.; Ling, X.; Shi, Y.; Lin, C.-T.; Huang, J.-K.; Chang, M.-T.; Chang, C.-S.; et al. Synthesis and Transfer of Single-Layer Transition Metal Disulfides on Diverse Surfaces. *Nano Lett.* 2013, 13, 1852–1857. [CrossRef]

21. Ling, X.; Lee, Y.-H.; Lin, Y.; Fang, W.; Yu, L.; Dresselhaus, M.S.; Kong, J. Role of the Seeding Promoter in MoS$_2$ Growth by Chemical Vapor Deposition. *Nano Lett.* 2014, 14, 464–472. [CrossRef] [PubMed]

22. Schmidt, H.; Wang, S.; Chu, L.; Toh, M.; Kumar, R.; Zhao, W.; Neto, A.H.C.; Martin, J.; Adam, S.; Adam, S.; et al. Transport Properties of Monolayer MoS$_2$ Grown by Chemical Vapor Deposition. *Nano Lett.* 2014, 14, 1909–1913. [CrossRef]

23. Schulman, D.S.; Arnold, A.J.; Das, S. Contact engineering for 2D materials and devices. *Chem. Soc. Rev.* 2018, 47, 3037–3058. [CrossRef]

24. Kaasbjerg, K.; Thygesen, K.S.; Jacobsen, K.W. Phonon-limited mobility in n-type single-layer MoS$_2$ from first principles. *Phys. Rev. B* 2012, 85, 115317. [CrossRef]

25. Radisavljevic, B.; Kis, A. Mobility engineering and a metal-insulator transition in monolayer MoS$_2$. *Nat. Mater.* 2013, 12, 815–820. [CrossRef]

26. Jariwala, D.; Sangwan, V.K.; Late, D.; Johns, J.E.; Dravid, V.P.; Marks, T.J.; Lauhon, L.J.; Hersam, M.C. Band-like transport in high mobility unencapsulated single-layer MoS$_2$ transistors. *Appl. Phys. Lett.* 2013, 102, 173107. [CrossRef]

27. Qiu, H.; Xu, T.; Wang, Z.; Ren, W.; Nan, H.; Ni, Z.; Chen, Q.; Yuan, S.; Miao, F.; Song, F.; et al. Hopping transport through defect-induced localized states in molybdenum disulphide. *Nat. Commun.* 2013, 4, 2642. [CrossRef]

28. Popov, I.; Seifert, G.; Tomanek, D. Designing Electrical Contacts to MoS$_2$ Monolayers: A Computational Study. *Phys. Rev. Lett.* 2012, 108, 156802. [CrossRef]

29. Gong, C.; Colombo, L.; Wallace, R.M.; Cho, K. The Unusual Mechanism of Partial Fermi Level Pinning at Metal-MoS$_2$ Interfaces. *Nano Lett.* 2014, 14, 1714–1720. [CrossRef]

30. Gong, C.; Zhang, H.; Wang, W.; Colombo, L.; Wallace, R.M.; Cho, K. Band alignment of two-dimension transition metal dichalcogenides: Application in tunnel field effect transistors. *Appl. Phys. Lett.* 2013, 103, 053513. [CrossRef]

31. Yu, Y.-J.; Zhao, Y.; Ryu, S.; Brus, L.; Kim, K.S.; Kim, P. Tuning the Graphene Work Function by Electric Field Effect. *Nano Lett.* 2009, 9, 3430–3434. [CrossRef] [PubMed]

32. Huang, H.; Xu, W.; Chen, T.; Chang, R.-J.; Sheng, Y.; Zhang, Q.; Hou, L.; Warner, J.H. High-Performance Two-Dimensional Schottky Diodes Utilizing Chemical Vapor Deposition-Grown Graphene-MoS$_2$ Heterojunctions. *ACS Appl. Mater. Interfaces* 2018, 10, 37258–37266. [CrossRef]

33. Baik, S.S.; Im, S.; Choi, H.J. Work Function Tuning in Two-Dimensional MoS$_2$ Field-Effect-Transistors with Graphene and Titanium Source-Drain Contacts. *Sci. Rep.* 2017, 7, 45546. [CrossRef] [PubMed]

34. Tian, H.; Tan, Z.; Wu, C.; Wang, X.; Mohammad, M.A.; Xie, D.; Yang, Y.; Wang, J.; Li, L.-J.; Xu, J.; et al. Novel Field-Effect Schottky Barrier Transistors Based on Graphene-MoS$_2$ Heterojunctions. *Sci. Rep.* 2014, 4, 5951. [CrossRef] [PubMed]

35. Shin, C.-J.; Wang, Q.H.; Son, Y.; Jin, Z.; Blankschtein, D.; Strano, M.S. Tuning On-Off Current Ratio and Field-Effect Mobility in a MoS$_2$-Graphene Heterostructure via Schottky Barrier Modulation. *ACS Nano* 2014, 8, 5790–5798. [CrossRef]

36. Li, X.; Cai, W.; An, J.; Kim, S.; Nah, J.; Yang, D.; Piner, R.; Velamakanni, A.; Jung, I.; Tutuc, E.; et al. Large-Area Synthesis of High-Quality and Uniform Graphene Films on Copper Foils. *Science* 2009, 324, 1312–1314. [CrossRef]

37. Dean, C.R.; Young, A.F.; Meric, I.; Lee, C.; Wang, L.; Sorgenfrei, S.; Watanabe, K.; Taniguchi, T.; Kim, P.; Shepard, K.L.; et al. Boron nitride substrates for high-quality graphene electronics. *Nat. Nanotechnol.* 2010, 5, 722–726. [CrossRef]

38. Ryu, S.; Liu, L.; Berclaud, S.; Yu, Y.-J.; Liu, H.; Kim, P.; Flynn, G.W.; Brus, L.E. Atmospheric Oxygen Binding and Hole Doping in Deformed Graphene on a SiO$_2$ Substrate. *Nano Lett.* 2010, 10, 4944–4951. [CrossRef]
41. Yoon, J.; Park, W.; Bae, G.-Y.; Kim, Y.; Jang, H.S.; Hyun, Y.; Lim, S.K.; Kahng, Y.H.; Hong, W.-K.; Lee, B.H.; et al. Highly Flexible and Transparent Multilayer MoS$_2$ Transistors with Graphene Electrodes. Small 2013, 9, 3295–3300. [CrossRef] [PubMed]

42. Chen, J.-R.; Odenthal, P.M.; Swartz, A.G.; Floyd, G.C.; Wen, H.; Luo, K.Y.; Kawakami, R.K. Control of Schottky Barriers in Single Layer MoS$_2$ Transistors with Ferromagnetic Contacts. Nano Lett. 2013, 13, 3106–3110. [PubMed]