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Schottky barrier contrasts in single and bi-layer graphene contacts for MoS₂ field-effect transistors

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We have investigated single- and bi-layer graphene as source-drain electrodes for n-type MoS₂ transistors. Ti-MoS₂-graphene heterojunction transistors using both single-layer MoS₂ (1M) and 4-layer MoS₂ (4M) were fabricated in order to compare graphene electrodes with commonly used Ti electrodes. MoS₂-graphene Schottky barrier provided electron injection efficiency up to 130 times higher in the subthreshold regime when compared with MoS₂-Ti, which resulted in V_DS polarity dependence of device parameters such as threshold voltage (V_TH) and subthreshold swing (SS). Comparing single-layer graphene (SG) with bi-layer graphene (BG) in 4M devices, SG electrodes exhibited enhanced device performance with higher on/off ratio and increased field-effect mobility (μFE) due to more sensitive Fermi level shift by gate voltage. Meanwhile, in the strongly accumulated regime, we observed opposing behavior depending on MoS₂ thickness for both SG and BG contacts. Differential conductance (σ_d) of 1M increases with V_DS irrespective of V_DS polarity, while σ_d of 4M ceases monotonic growth at positive V_DS values transitioning to ohmic-like contact formation. Nevertheless, the low absolute value of σ_d saturation of the 4M-graphene junction demonstrates that graphene electrode could be unfavorable for high current carrying transistors.

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Previous in-depth studies on two-dimensional (2D) MoS₂ transistors have demonstrated excellent field-effect mobility with high on-off ratio at room temperature.¹–³ However, Schottky barrier (SB) formation⁴ or wetting issues of metal-MoS₂ have limited device performance.⁵ Even the most commonly studied MoS₂-Ti contact has been the subject of much debate regarding the formation of ohmic or Schottky contacts.¹ Similar controversy exists for graphene-MoS₂ contacts.⁶,⁷ Previously, improved MoS₂ device properties were reported by utilizing graphene rather than metal contacts which resulted in an unexpectedly lower SB height in comparison with MoS₂-metal junctions.⁷,⁸ However, due to high sheet resistance of intrinsic monolayer graphene, doping or multi-layer graphene has been introduced."³

We fabricated three back-gated MoS₂ field-effect transistors (FETs) with Ti and graphene asymmetric electrodes in each device for direct comparisons in transistor performance. Three different FETs: single-layer MoS₂/single-layer graphene (1M-SG), 4-layer MoS₂/single-layer graphene (4M-SG), and 4-layer MoS₂/bi-layer graphene (4M-BG) were fabricated. The channel length/width (L/W) of each device was 2.6 μm/6.2 μm, 3.4 μm/3.2 μm, and 2.8 μm/5.5 μm for 1M-SG, 4M-SG, and 4M-BG, respectively. The characterization of thickness for MoS₂ and graphene including Raman spectrum and atomic force microscope imaging, as well as detailed transistor fabrication processes are described in the supplementary material.¹⁰

The transfer curves (I_DS-V_BG) and output curves (I_DS-V_DS) for our three devices are presented in Figs. 1(d)–1(f) and 1(g)–1(i), respectively, where I_DS is source-drain channel current and V_BG and V_DS are back gate voltage and source-drain voltage, respectively. V_DS is applied at the Ti electrode with grounded graphene electrode. All three devices demonstrate n-channel transistor behavior with on/off ratio of 10⁶–10⁷.

Nonlinear and asymmetric output characteristics in Figs. 1(j)–1(l) demonstrate back-to-back SB formation in fabricated MoS₂ transistors. Photocurrent data at V_BG = 0 V also support the existence of SBs.¹⁰ In contrast to conventional Si transistors, output curves show non-linearity rather than initial linear regimes and subsequent saturation with the increase in V_DS.

4M devices (4M-SG and 4M-BG) show larger off (I_OFF) and on current (I_ON) than 1M device (1M-SG). The higher I_DS in 1M devices can be explained by a few factors. First, the band gap of MoS₂ shows a transition from indirect to direct gap with monotonic increase from 1.3 eV for bulk to 1.9 eV for single-layer, while electron affinity also diminishes from 4.4 eV to 3.7 eV.¹¹ Therefore, thickness decrease

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accompanies the decrease in intrinsic conductivity in MoS2 and the increase in SB height in MoS2-graphene junction. Second, the depth of the surface channel known to be 6–10 nm in 2D MoS2 affects transport properties in both 1M and 4M. Consequently, in the strongly accumulated regime carrier, scatterings due to electron-electron interaction are reinforced in the narrow and dense channel of 1M devices.

In the transfer curve, $I_{DS}$ is larger at positive (solid lines) $V_{DS}$ than negative (dotted lines) $V_{DS}$ especially in the subthreshold regime. We have redrawn transfer curves at $V_{DS} = \pm 3$ V with the ratio $R_{AI} = (I_{DS}(+3V) - I_{DS}(-3V))/I_{DS}(-3V)$ to compare $V_{DS}$ polarity quantitatively in the inset of Fig. 1(f). Positive $R_{AI}$ indicates higher $I_{DS}$ at positive $V_{DS}$. Substantial distinction was mainly observed below zero $V_{BG}$ with the largest $R_{AI}$ of 130 in 4M-BG. Normalized output curves by $I_{DS}(+3V)$ manifest diode-like nonlinear behavior in the off regime and evolve into more symmetric (though still nonlinear) ones up to $V_{BG} = 20$ V as shown in Figs. 1(j)–1(l).

When applying positive (negative) $V_{DS}$ on Ti contact, graphene (Ti) plays the role of source junction in n-type MoS2 transistors. The overall observed data, such as diode-like behavior in normalized $I_{DS}$-$V_{DS}$, higher $I_{DS}$ in positive $V_{DS}$, positive $R_{AI}$, confirm the enhanced injection efficiency from the graphene contact rather than the Ti contact, particularly in the subthreshold regime. The increased van der Waals interaction and electron transfer from MoS2 to graphene shown in Raman10 are accountable for barrier lowering and hence injection efficiency in our fabricated devices.

We extracted device parameters from transfer curves: cut-off voltage ($V_{OFF}$), threshold voltage ($V_{TH}$), subthreshold swing (SS), and field-effect mobility ($\mu_{FE}$). $V_{OFF}$, the gate voltage where channel surface potential crosses Fermi level ($E_F$) of MoS2, is obtained from the $V_{BG}$ of minimum current and shown in Fig. 2(a). $V_{OFF}$ decreases with $V_{DS}$ magnitude but no distinction can be drawn with $V_{DS}$ polarity (positive $V_{DS}$ shown in closed circles and negative $V_{DS}$ shown in open circles). $V_{TH}$ shown in Fig. 2(b) also decreases with $V_{DS}$ but in contrast to $V_{OFF}$, $V_{TH}$ is lower at positive $V_{DS}$ (closed circle) indicating polarity dependence. $V_{DS}$ dependent parameter variation in Fig. 2 is ascribed to non-ohmic SB behavior.

$V_{DS}$ dependence of $V_{TH}$ in our transistor is not consistent with the results in conventional transistors. If the second order term of $V_{DS}$, which is generally neglected, is included in channel current, $V_{TH}$ should increase as a function of $V_{DS}$ as shown in following equation. $I_{DS}(V_{GS}) = L/W\mu_{FE}C_{OX} [(V_{GS} - V_{TH}) V_{DS} - V_{DS}^2/2] = L/W\mu_{FE}C_{OX}[V_{GS} - (V_{TH} + V_{DS}/2)] V_{DS}$, where $C_{OX}$ and $V_{GS}$ are gate oxide capacitance and gate-source voltage, respectively.13 We should note here that the lowest $V_{OFF}$ was observed in 4M-BG, while the lowest $V_{TH}$ was observed in 4M-SG.

Fig. 2(c) plots SS ($dV_{GS}/d(logI_{DS})$) which is the gate voltage required to increase channel current by a decade in...
dependence of SS variation. In Fig. 2(d), the field effect mobility
positive (negative) \( V_{DS} \) in 1M-SG, 4M-SG, and 4M-BG corre-
gmax represents the maximum of transconductance
fixed \( V_{DS} \)
metal-channel overlapped area should explain the \( V_{DS} \) de-
states, the contribution by depletion capacitance in channel or
the subthreshold regime. SS variation results from the serially
connected capacitance to \( C_{OX} \).13 With negligible inter-
state states, the contribution by depletion capacitance in channel or
the channel material is equivalently 4M, the contact
contribution determines \( \sigma_d \) evolution as a function of \( V_{DS} \).
Recalling earlier that \( \mu_{FE} \) of 4M-SG becomes more symmetric
with increased \( V_{BG} \). This is from the relatively reduced
effect of asymmetric SB at high gate bias. Second, \( \sigma_d \)
increased 6 orders for 1M-SG, while it increased only 3
orders in 4M devices as \( V_{BG} \) increased from \(-60 \) to \( 80 \) V.
Third, for 4M devices \( \sigma_d \) in 4M-BG than 4M-SG starts
higher at \( V_{BG} = -60 \) V, however, the trend reverses and \( \sigma_d \)
of 4M-BG is lower at \( V_{BG} = 0 \) V, and the ratio of \( \sigma_d \) for 4M-
Sg to 4M-BG remains approximately 1.5 above \( V_{BG} = 20 \) V.
\( \sigma_d \) includes terms from channel and contact contributions.
Since the channel material is equivalently 4M, the contact
In Fig. 2(d), the field effect mobility is calculated by the equation
\( \mu_{FE} = \frac{g_{max}}{V_{DS}} \) and is obtained around \( V_{BG} = 60 - 80 \) V. The extracted \( \mu_{FE} \) at
positive (negative) \( V_{DS} \) in 1M-SG, 4M-SG, and 4M-BG corre-
spond to 4.4 ± 0.33 (4.4 ± 1.1), 42.9 ± 1.5 (49.9 ± 0.6), and
27.4 ± 2.1 (32.5 ± 2.9) cm²/V s, respectively.

to analyze the details of 4M-SG and 4M-BG transistors,
the previous transfer curves were replotted in Fig. 3(a) for
fixed \( V_{DS} = 0.1 \) V, 1 V, and 3 V. 4M-SG demonstrated higher
\( V_{OFF} \) and lower \( V_{TH} \) than 4M-BG implying improved gate
control due to the SB modulation of graphene-MoS₂ junction
during transitioning from complete depletion to flat band of the
surface channel.

The band diagram for gate modulation was illustrated in
Fig. 3(b). Negative \( V_{BG} \) down-shifts \( E_F \) by p-doping
graphene, while positive \( V_{BG} \) up-shifts \( E_F \) by n-doping graphene.
Respective up- and down-shifts of graphene \( E_F \) with n-type
MoS₂ induce the increase and the decrease in SB height. The \( E_F \)
variation of SG and BG can be described by the change of car-
derrier density (n) induced by gate voltage, i.e., \( E_F = \frac{\hbar v_F n}{2\pi m^*} \)
for SG and \( E_F = \frac{\hbar v_F n}{2\pi m^*} \) for BG, where \( v_F = 1 \times 10^6 \) m/s is the
Fermi velocity of SG, and \( m^* = 0.033 m_e \) is the effective mass
of carrier in BG relative to the bare electron mass \( m_e \).9 This was
otherwise also observed by work function measurement, which showed an
excellent agreement.14 With 300 nm SiO₂ gate oxide, the range
of \( E_F \) shift corresponds to \( \pm 200 \) meV and \( \pm 300 \) meV with
\( V_{BG} = 80 \) V for SG and BG, respectively, as shown in Fig.
3(c). Therefore, in 4M-SG devices, \( I_{OFF} \) is suppressed by larger
SB height, while \( I_{ON} \) is conversely enhanced by lower SB height.
As we noted in Fig. 2, the lower \( V_{OFF} \) in 4M-BG along with the lower \( V_{TH} \) in 4M-SG makes it obvious that SS and
\( \mu_{FE} \) are also enhanced in 4M-SG. During transistor opera-
tion, higher on/off ratio of 4M-SG originates from both lower
\( I_{OFF} \) and higher \( I_{ON} \).

We extracted differential conductance (\( \sigma_d \)) from
\( I_{DS}-V_{DS} \) at constant \( V_{BG} \) by differentiating \( I_{DS} \) with respect to
\( V_{DS} \) as shown in Fig. 4. The \( \sigma_d \) curves reveal several distinct
features not easily observed from the \( I_{DS}-V_{DS} \) curves.
First, highly asymmetric curves of \( \sigma_d \) become more symmetric
with increased \( V_{BG} \). This is from the relatively reduced
effect of asymmetric SB at high gate bias. Second, \( \sigma_d \)
increased 6 orders for 1M-SG, while it increased only 3
orders in 4M devices as \( V_{BG} \) increased from \(-60 \) to \( 80 \) V.
Third, for 4M devices \( \sigma_d \) in 4M-BG than 4M-SG starts
higher at \( V_{BG} = -60 \) V, however, the trend reverses and \( \sigma_d \)
of 4M-BG is lower at \( V_{BG} = 0 \) V, and the ratio of \( \sigma_d \) for 4M-
Sg to 4M-BG remains approximately 1.5 above \( V_{BG} = 20 \) V.

\( \sigma_d \) includes terms from channel and contact contributions.
Since the channel material is equivalently 4M, the contact
contribution determines \( \sigma_d \) evolution as a function of \( V_{DS} \).
Recalling earlier that \( \mu_{FE} \) of 4M-SG was 1.5 times that of
4M-BG, similar to the \( \sigma_d \) ratios between 4M-SG to 4M-BG.
Our results show some evidence that the mobility enhance-
ment of 4M-SG may be connected to improved contact resis-
tance in the accumulation regime. Both \( I_{DS}-V_{BG} \) and \( \sigma_d \)
establish that SG might be favorable as an electrode, demonstrat-
ating raised on/off ratio, improved SS, and \( \mu_{FE} \) than BG.

The saturation of \( \sigma_d \) and eventual decrease were
observed at positive \( V_{DS} \) in the accumulation regime of both
4M-SG and 4M-BG as shown in Fig. 4 from \( V_{BG} = 20 \) V to
80 V, but not for 1M devices. As previously mentioned, if \( \sigma_d \)
changes are related with contact resistance, then the ohmic-
lke 4M-graphene junction can explain this behavior. In fact,
at \( V_{BG} > 20 \) V pulling the conduction band close to \( E_F \) in
n-type MoS₂ considerably drops SB width, which allows for
efficient carrier injection by tunneling through a transparent
SB of nearly negligible width. However, the eventual satura-
tion level of \( \sigma_d \) is lower than that of the 4M-Ti junction
which shows monotonic growth at negative \( V_{DS} \). High gra-
phene sheet resistance could be a major culprit in limiting

FIG. 2. \( V_{DS} \) dependent (a) \( V_{OFF} \), (b) threshold voltage (\( V_{TH} \)) extracted by
conventional linear extrapolation method, (c) Subthreshold swing (SS), and
(d) field-effect mobility (\( \mu_{FE} \)).

FIG. 3. (a) Transfer curves of 4M-SG (lines) and 4M-BG (dotted lines) at
\( V_{DS} = 0.1 \) V (black), 1 V (red), and 3 V (green). (b) The comparison of band
diagram for 4M-SG and 4M-BG. (c) Calculated \( E_F \) variation of SG and BG
by gate voltage.
conductance.\textsuperscript{15} In contrast to the improved performance in the subthreshold regime, $\mu_{FE}$ decline in graphene source electrode devices seems to be associated with limited $\sigma_d$ at a rather low saturation level. From the point of view of applications, when high concentrations of charge carriers must be delivered, we can speculate that graphene could be a poor choice of source electrode.

In conclusion, transport properties on metal-MoS$_2$-graphene transistors revealed enhanced performance with graphene as source electrode particularly in the subthreshold regime. On the contrary, $\mu_{FE}$ measurement showed the opposite results. $V_{DS}$ polarity dependence of device parameters, such as SS and $V_{TH}$, was ascribed to the asymmetric Schottky barrier height. We confirmed that single-layer graphene as source electrode provides improved transistor properties than that of bi-layer as demonstrated with lower SS and larger $\mu_{FE}$. However, in spite of improved charge carrier injection, graphene can still be unfavorable in the strongly accumulated regime if employed for high current delivery without further improved sheet resistance.

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\textsuperscript{10}See supplementary material at http://dx.doi.org/10.1063/1.4937266 for the followings: Schematic illustration of fabrication process; thickness analysis of 2D materials by AFM and Raman; Raman spectra of MoS$_2$ and MoS$_2$/graphene overlapped area; and photocurrent mapping image $V_{BG}$=0V.


