Enhanced photoresponsivity of multilayer MoS$_2$ transistors using high work function MoO$_x$ overlayer

Geonwook Yoo, Seongin Hong, Junseok Heo, and Sunkook Kim

Citation: Appl. Phys. Lett. 110, 053112 (2017); doi: 10.1063/1.4975626
View online: http://dx.doi.org/10.1063/1.4975626
View Table of Contents: http://aip.scitation.org/toc/apl/110/5
Published by the American Institute of Physics

Articles you may be interested in

Improved integration of ultra-thin high-k dielectrics in few-layer MoS$_2$ FET by remote forming gas plasma pretreatment
Appl. Phys. Lett. 110, 05310053110 (2017); 10.1063/1.4975627

Chemical vapor deposition of monolayer MoS$_2$ directly on ultrathin Al$_2$O$_3$ for low-power electronics
Appl. Phys. Lett. 110, 053101053101 (2017); 10.1063/1.4975064

Encapsulation of graphene in Parylene
Appl. Phys. Lett. 110, 053504053504 (2017); 10.1063/1.4975491

Ultrafast photocurrent measurements of a black phosphorus photodetector
Appl. Phys. Lett. 110, 051102051102 (2017); 10.1063/1.4975360
Enhanced photoresponsivity of multilayer MoS$_2$ transistors using high work function MoO$_x$ overlayer

Geonwook Yoo,$^{1,8}$ Seongin Hong,$^2$ Junseok Heo,$^3$ and Sunkook Kim$^{2,9}$

$^1$School of Electronic Engineering, Soongsil University, Seoul 06938, South Korea
$^2$School of Advanced Materials Science & Engineering, Sungkyunkwan University, 300, Chunchun-dong, Jangan-gu, Suwon, 16419, South Korea
$^3$Department of Electrical and Computer Engineering, Ajou University, Suwon 16499, South Korea

(Received 1 December 2016; accepted 24 January 2017; published online 3 February 2017)

Using thin sub-stoichiometric molybdenum trioxide (MoO$_x$, $x<3$) overlayer, we demonstrate over 20-folds enhanced photoresponsivity of multilayer MoS$_2$ field-effect transistor. The fabricated device exhibits field-effect mobility ($\mu_{FE}$) of up to 41.4 cm$^2$/V s and threshold voltage ($V_{TH}$) of $-9.3$ V, which is also modulated by the MoO$_x$ overlayer. The MoO$_x$ layer ($\sim$25 nm), commonly known for a high work function ($\sim$6.8 eV) material with a band gap of $\sim$3 eV, is evaporated on top of the MoS$_2$ channel and confirmed by the transmission electron microscope analysis. The electrical and optical modulation effects are associated with interfacial charge transfer and thus an induced built-in electric field at the MoS$_2$/MoO$_x$ interface. The results show that high work function MoO$_x$ can be a promising heterostructure material in order to enhance the photoresponse characteristics of MoS$_2$-based devices. Published by AIP Publishing.

Molybdenum disulfide (MoS$_2$), a two-dimensional transition metal dichalcogenide, has been considered a promising candidate for emerging optoelectronic devices. MoS$_2$ possesses unique and advantageous properties such as controllable bandgap energy from a direct (1.2 eV) to indirect band gap (1.8 eV) depending on the number of layers, high mobility ($\sim$200 cm$^2$/V s) with a high-k dielectric layer, and high absorptivity. Especially, the existing bandgap allows low-dark current by modulating MoS$_2$ channel to depletion, which is suitable for a phototransistor mode. Furthermore, its outstanding mechanical property enlarges the application to flexible and conformal photodetectors for wearable devices, and the recent advances in scalable and large-area process of TMDCs (Transition Metal Dichalcogenides) make these unique features more compelling toward image sensor.

In this sense, intensive research efforts have been devoted to improve figure-of-merits (e.g., responsivity and spectral response) of MoS$_2$-based photodetectors by adopting a surface plasmonic nanostructure, a unique device structure, and various organic and inorganic overlayers. For example, the p-type PbS quantum-dots, p-type organic semiconductors (e.g., copper phthalocyanine, rubrene), organic dye, and hafnium oxide were investigated as a stacking layer on the top of MoS$_2$, resulting in enhanced photoresponsivity ($R$). Meanwhile, various p-n junctions based on MoS$_2$ heterostructure are also suggested for enhanced photodetection characteristics.

Considering desirable properties for an overlayer, molybdenum trioxide (MoO$_x$) can be a good alternative in that MoO$_x$ possess a large bandgap ($\sim$3 eV) and is more stable compared with organic materials in ambient conditions. In addition, a high work function of evaporated MoO$_x$ (6.6–6.8 eV) layer allows modulating of electronic property of MoS$_2$ field-effect transistor (FET).

In this study, we present multilayer MoS$_2$ phototransistor with an MoO$_x$ overlayer deposited by thermal evaporation. The interface between MoS$_2$ and MoO$_x$ is characterized using transmission electron microscope (TEM) with energy dispersive spectroscopy (EDS) analysis. We investigate electrical performance and photoresponsivity characteristics of multilayer MoS$_2$ FET before and after stacking an MoO$_x$ overlayer onto the MoS$_2$ channel. It is found that the photoresponsivity of the MoS$_2$ phototransistor with the thin MoO$_x$ overlayer is enhanced by more than 20 times, which is associated with electron charge transfer due to the high work function of MoO$_x$. Finally, energy band diagram model is used to discuss the results.

Figure 1(a) shows a schematic illustration of multilayer MoS$_2$ transistor with an MoO$_x$ overlayer. Mechanically exfoliated MoS$_2$ flakes from bulk MoS$_2$ crystals (Graphene Market, USA) by a conventional scotch-tape method were transferred onto a heavily doped p-type Si substrate with thermally grown 200 nm SiO$_2$ layer. For the source and drain (S/D) electrodes, Au ($\sim$70 nm) was evaporated, followed by patterning using the conventional lift-off technique. Then, a thin MoO$_x$ layer ($\sim$25 nm) was deposited by thermal evaporation, followed by opening electrical contact using conventional photolithography and wet chemical etching. Finally, the fabricated device was annealed at 573 K in N$_2$ for 10 min using a rapid thermal annealing to remove any absorbed organic residues and to improve contact resistance. Figures 1(b) and 1(c) show a cross sectional TEM image of the SiO$_2$/MoS$_2$/MoO$_x$ stack and its enlarged image at the MoS$_2$/MoO$_x$ interface with EDS analysis, respectively. The transition layer was $\sim$3.1 nm thick, in which the atomic % ratio changes clearly show the sub-stoichiometric MoO$_x$ ($x<3$) layer deposited on top of the multilayer MoS$_2$ flake. The transition layer formed due to interface mixing during thermal evaporation used in this work.

Figure 2 shows typical electrical characteristics of the fabricated device. Measured transfer curves of $I_{DS}=V_{GS}$ for...
The charge concentration ($n$) using the parallel-capacitor model with $n = Q/e = C_{OX} (V_G - V_{TH})/e$, where $C_{OX}$ is capacitance of SiO$_2$, $V_G$ is gate bias, $V_{TH}$ is threshold voltage, and $e$ is elementary charge. The reduction of $\mu_{eff}$ originated from the scattering by impurities at the intermixed interface between MoO$_x$ and MoS$_2$, and from the depleted charge concentration in the channel. The increased $SS$ indicates that interface trap density at the intermixed interface increased as well. More details with energy band diagram will be discussed in the following.

We investigated photoresponsive characteristics of the multilayer MoS$_2$ phototransistor before (w/o MoO$_x$) and after (w/ MoO$_x$) stacking the MoO$_2$ layer. Figure 3(a) shows power-dependent photoresponsivity ($R$) for various irradiances (8.6, 17.2, 34.4, 68.7, 137.5, 274.9 mW/cm$^2$) as well as gate-bias ($V_{GS} = -30$ to $-10$ V, 5 V step) conditions under an incident light wavelength ($\lambda$) of 532 nm. The photoresponsivity was calculated from $R = I_{ph}/P_{inc}$. Two typical characteristics were observed for both bare (w/o MoO$_x$) and MoO$_x$ overlaid MoS$_2$ (w/ MoO$_x$) devices; photoresponsivity was reduced with increasing irradiance as well as gate modulation ($V_{GS}$) due to a trap-dominated process.

In order to characterize photostimulating behavior of the devices, we measured temporal photoresponses at $V_{DS} = 1$ V with a pulsed laser at 638 nm. Photocurrents generated and recombined in accordance with the incident light, turning on and off at a period of 20 s. The observed temporal responses throughout the multiple cycles demonstrate a stable and constant

### Table I. Electrical parameters and photoresponsivity ($R$) of the fabricated MoS$_2$ FETs before and after stacking MoO$_2$ layer.

<table>
<thead>
<tr>
<th>$V_{TH}$ (V)</th>
<th>$\mu_{eff}$ (cm$^2$/V s)</th>
<th>$SS$ (V/dec)</th>
<th>$R$ (A/W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Before</td>
<td>$-17.3$</td>
<td>62.1</td>
<td>1.8</td>
</tr>
<tr>
<td>After</td>
<td>$-9.3$</td>
<td>41.4</td>
<td>3.2</td>
</tr>
</tbody>
</table>

$V_{DS} = 1$ V are presented in Fig. 2(a), in which the effect of the MoO$_x$ overlayer on $V_{TH}$ shift ($\Delta V_{TH} \approx 8.0$ V) is clearly shown. The field-effect mobility ($\mu_{eff}$) in a linear operation regime was extracted from $\mu_{eff} = L/2W(C_{OX} V_{DS})$, where $W$ is channel width, $L$ is channel length, $C_{OX}$ is capacitance of SiO$_2$, and $V_{DS}$ is drain bias of 1 V. Threshold voltage ($V_{TH}$) was calculated using a linear extrapolation method in a linear regime ($V_{DS} = 1$ V); it was found from the intercept of a tangent at the minimum $g_m$ with $V_{GS}$ axis. Subthreshold slope was calculated as $SS = [d(\log_{10} I_d)/dV_{GS}]^{-1}$. Figure 2(b) represents output characteristics ($I_{DS}$-$V_{DS}$) for $V_{GS} = 0$, 5, 10, 15, 20 V, exhibiting an ohmic-like linear behavior at low $V_{DS}$ and saturation at high $V_{DS}$ bias conditions. Electrical parameters are summarized in Table I. The positive $\Delta V_{TH}$ is attributed to the significant electron transferring from MoS$_2$ to MoO x, resulting in the depletion of electrons in the MoS$_2$ channel. The average $\Delta V_{TH}$ was $\sim 7.6 \pm 0.9$ V, and its corresponding amount of depleted charge concentration after depositing MoO$_x$ overlayer was estimated to $\Delta n \sim (0.8 \pm 0.1) \times 10^{12}$ cm$^{-2}$ (at $V_G = 20$ V).
photoswitching performance of the device. In particular, the response times (i.e., rise and fall time) were calculated from 10% to 90%, and 90% to 10% of the maximum photocurrent after light was turned on and off, respectively. After stacking MoO$_x$ layer, the rise time ($t_{rise}$) decreased from 6.65 s to 3.52 s while the fall time ($t_{fall}$) remained similar. It is to be noted that the photosresponsivity is enhanced by more than 20-folds with enhanced switching characteristics for the measured irradiance and $V_{GS}$ ranges.

Aforementioned $V_{TH}$ shift and enhanced photosresponsivity by stacking thin MoO$_x$ overlayer can be explained using the energy-band diagram. It is commonly considered that evaporated MoO$_x$ films have a very high work function of 6.6–6.8 eV and a bandgap of ~3 eV. Note that the incident light wavelengths (532 nm and 638 nm) are not absorbed in the MoO$_x$ overlayer due to its large bandgap (~3 eV). As presented in Fig. 4a, when MoS$_2$ and MoO$_x$ form a physical contact, a significant electron charge transfer from MoS$_2$ to MoO$_x$ because the energy levels of MoO$_x$, including Fermi level ($E_F$) and conduction band (CB) locate below the valence band (VB) of MoS$_2$. Consequently, significant interfacial charge transfer occurs and electron charges deplete in MoS$_2$ (i.e., less accumulated charges), and thus $V_{TH}$ shift toward positive direction. Meanwhile, the interfacial charge transfer can induce upward band-bending in MoS$_2$ and downward in MoO$_x$ at equilibrium, as shown in Fig. 4b. Thus, an induced built-in electric field can effectively separate photo-generated electron-hole pairs in MoS$_2$ and contribute to the holes to be trapped at the interface trap sites instead of electron-hole recombination. Therefore, photosresponsivity of the multilayer MoS$_2$ phototransistor could be enhanced by more than 20 times at $V_{GS} = -30$ V by stacking the MoO$_x$ overlayer.

In conclusion, we have reported on electrical and photosresponsive characteristics of the multilayer MoS$_2$ FET with the evaporated thin MoO$_x$ overlayer. The fabricated device exhibits $J_{th}$ of up to 41.4 A cm$^{-2}$ and the threshold voltage of ~9.3 V, which was positive-shifted from ~17.3 V. Furthermore, the MoO$_x$ overlayer results in enhanced photosresponsivity of more than 20 times (65.2 A/W) at $V_{GS} = -30$ V under the illumination of 532 nm and 8.5 mW/cm$^2$. The modulation of electrical as well as optical properties is associated with the high work function of evaporated MoO$_x$ layer contributing to the significant interfacial electron charge transfer from the MoS$_2$ channel to MoO$_x$ and thus induced built-in electric field at the MoS$_2$/MoO$_x$ interface. The large bandgap of MoO$_x$ can be an
additional merit as an overlay to be transparent to visible light. The results suggest that photoresponsive characteristics of the multilayer MoS$_2$ phototransistor can be greatly improved by integrating an inorganic layer, MoO$_x$, with high work function, and can further be a promising heterostructure for high performance phototransistors based on MoS$_2$ and other 2-D materials.

This research was supported in part by the National Research Foundation of Korea (NRF) funded by the Ministry of Science, ICT, and Future Planning (Grant No. NRF-2014M3A9D7070732).