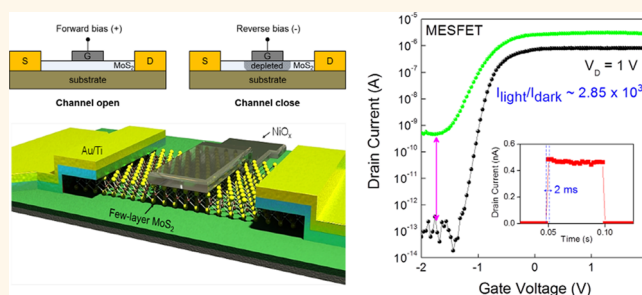


Metal Semiconductor Field-Effect Transistor with MoS₂/Conducting NiO_x van der Waals Schottky Interface for Intrinsic High Mobility and Photoswitching Speed

Hee Sung Lee,[†] Seung Su Baik,[‡] Kimoon Lee,[§] Sung-Wook Min,[†] Pyo Jin Jeon,[†] Jin Sung Kim,[†] Kyujin Choi,[†] Hyoung Joon Choi,[‡] Jae Hoon Kim,[†] and Seongil Im^{*,†}

[†]Institute of Physics and Applied Physics, Yonsei University, Seoul 120-749, Korea, [‡]Center for Computational Studies of Advanced Electronic Material Properties, Yonsei University, Seoul 120-749, Korea, and [§]Department of Physics, Kunsan National University, 558 daehak-ro, Gunsan, Jeonbuk 573-701, Korea

ABSTRACT Molybdenum disulfide (MoS₂) nanosheet, one of two-dimensional (2D) semiconductors, has recently been regarded as a promising material to break through the limit of present semiconductors. With an apparent energy band gap, it certainly provides a high carrier mobility, superior subthreshold swing, and ON/OFF ratio in field-effect transistors (FETs). However, its potential in carrier mobility has still been depreciated since the field-effect mobilities have only been measured from metal–insulator–semiconductor (MIS) FETs, where the transport behavior of conducting carriers located at the insulator/MoS₂ interface is unavoidably interfered by the interface traps and gate voltage. Moreover, thin MoS₂ MISFETs have always shown large hysteresis with unpredictable negative threshold voltages. Here, we for the first time report MoS₂-based metal semiconductor field-effect transistors (MESFETs) using NiO_x Schottky electrode which makes van der Waals interface with MoS₂. We thus expect that the maximum mobilities or carrier transport behavior of the Schottky devices may hardly be interfered by interface traps or an on-state gate field. Our MESFETs with a few and ~10 layer MoS₂ demonstrate intrinsic-like high mobilities of 500–1200 cm²/(V s) at a certain low threshold voltage between –1 and –2 V without much hysteresis. Moreover, they work as a high speed and highly sensitive phototransistor with 2 ms switching and ~5000 A/W, respectively, supporting their high intrinsic mobility results.



KEYWORDS: 2D nanosheet MoS₂ · NiO_x · MESFET · Hall measurement · Schottky junction · van der Waals interface

Until recent years, molybdenum disulfide (MoS₂) nanosheet among many two-dimensional (2D) transition metal dichalcogenide semiconductors^{1–22} has been regarded as one of the most promising materials to break through the limit of present semiconductors.^{3–10} With an apparent energy band gap, of which the range is 1.2–1.8 eV depending on the MoS₂ thickness,^{2,7} it certainly provides a high carrier mobility, superior subthreshold swing (SS), and ON/OFF current ratio in metal insulator semiconductor field-effect transistors (MISFETs).^{2–11} The highest mobility has been mostly achieved from few layer MoS₂

MISFETs with a top-gate high-k dielectric layer.^{3–5,7,8,10,11} However, their field-effect mobility is in a wide range from 10 to 1000 cm²/(V s) and their threshold voltages are not reproducibly consistent, either, in reports because those values should be seriously dependent upon the dielectric/MoS₂ interface and bulk trap states in a MISFET where in particular the mobility is influenced by the gate voltage as well.^{23,24} As a result, the potential or intrinsic carrier mobility of MoS₂ has not been properly appreciated yet since the field-effect mobilities have mostly been measured from MISFET structures,^{25,26} where the interface/bulk traps and gate

* Address correspondence to semicon@yonsei.ac.kr.

Received for review May 8, 2015 and accepted July 14, 2015.

Published online July 14, 2015
10.1021/acsnano.5b02785

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voltage unavoidably interfered with the transport behavior of conducting carriers located at the insulator/MoS₂ interface.^{23,24} Moreover, the top-gate high mobility MoS₂ MISFETs have often shown unpredictable and large negative threshold voltages.^{8,11,27,28}

To resolve such issues in MoS₂ FETs, we report, for the first time, metal semiconductor field-effect transistors (MESFETs)^{29,30} with MoS₂–NiO_x van der Waals Schottky junction interface, where the maximum mobilities or carrier transport behavior of the Schottky devices may hardly be interfered with by interface traps or an on-state gate field.^{30,31} Our MESFETs with a few and ~10 layer MoS₂ demonstrate high mobilities of 500–1200 cm²/(V s) approaching their intrinsic room temperature mobilities at a certain low threshold voltage between –1 and –2 V with little hysteresis. Moreover, they work as a high speed and highly sensitive phototransistor with 2 ms switching and ~5000 A/W, respectively, supporting their high intrinsic mobility results.

RESULTS AND DISCUSSION

Figure 1a displays the top view optical microscopy image of the MESFET with 10-layer MoS₂ channel contacting 150 nm-thick thermally evaporated NiO_x gate (G) on 285 nm-thick SiO₂/p⁺-Si substrate. According to the image, the MoS₂ flake shape is a little oblique to the source (S)-to-drain (D). NiO_x is known to have quite a deep work function of more than 5.1–5.2 eV as a Ni-rich semitransparent conducting oxide ($x \sim 0.9$).^{32,33} For a S/D ohmic contact, we utilized an Au/Ti bilayer, so that Ti might contact our MoS₂ nanosheet, as shown in the schematic three-dimensional (3-D) view of our MESFET device of Figure 1c. The channel thickness of our 10-layer MoS₂ on 285 nm-thick SiO₂/p⁺-Si was confirmed by atomic force microscope (AFM) scan in Figure 1b and its inset topological 3-D image.

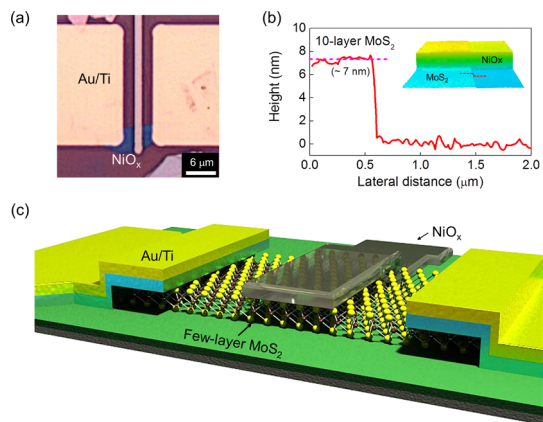


Figure 1. (a) Top views of our MESFET with 10-layer MoS₂, NiO_x top-gate, and Au/Ti source/drain in an optical microscopy. (b) AFM topography results from 10-layer thin MoS₂ channel in MESFET (thickness was measured to be ~7 nm). (c) Schematic 3D view of MoS₂-based MESFET with 6 μm-long channel, 3 μm-long NiO_x gate, and S/D region in contact with Ti.

(Other optical microscopies of thick MoS₂ nanosheets are also displayed in Supporting Information Figure S1.)

To achieve any typical operation of MESFET, the following two conditions should be met. First, Schottky barrier should be clearly formed between the gate and channel semiconductor nanosheet, so that the G-to-S or G-to-D Schottky diode operates as a rectifier. According to the current–voltage (I – V) characteristics from the 10- and 60-layer MoS₂/NiO_x diode junction in Figure 2a, the 40 nm-thick MoS₂ (with 60-layer) was not able to properly form any good Schottky junction with NiO_x showing very small ON/OFF ratio of less than 10. On the other hand, 10-layer MoS₂ sheets appear to form good Schottky barriers with respect to NiO_x, showing 2.49 as an ideality factor. It is also recognized that the reverse leakage current becomes larger with the sheet thickness, indicating that some lowering of physical barrier takes place with a thick MoS₂. The Schottky barrier lowering is certainly related to the thickness-induced band gap reduction in MoS₂; the conduction band edge of MoS₂ might decrease with the thickness increase as its bandgap does decrease.^{2,7} (Schematic illustrations and Schottky equations in Supporting Information Figures S2 and S3 help understanding these phenomenon.)

Second, the MESFET should turn on before the Schottky diode starts to operate, otherwise drain current (I_D) would be consumed by or leaked to the gate (joining the G-to-S leakage current, I_{GS}). As a matter of fact, we also fabricated Schottky pure metal (Pd and Pt)-driven MESFETs which did not work as an FET, although they operated well as a Schottky diode, because such deep work function metal gate device could not meet the second condition. (See Supporting Information Figure S4a,b.) It is interesting to note that unlike such cases of pure metal Schottky gate device, conducting NiO_x Schottky gate device operated very well as a MESFET. We suppose that this difference comes from a unique interface between thermally evaporated NiO_x and MoS₂, which would be much different from that between pure metal/MoS₂. In general, the pure metal contaminates the surface of MoS₂ due to an intimate chemical binding between metal and MoS₂ (Supporting Information Figure S4b),³⁴ while NiO_x would make only van der Waals interface^{14–16,35,36} to MoS₂ so as not to cause gate leakage during FET operation. Our density-functional-theory calculations show that the NiO_x/MoS₂ interface has a large NiO_x–MoS₂ distance of 3.31 Å with no significant influence on the electric structure of MoS₂, while Ti–MoS₂ and Au–MoS₂ interfaces have Ti–MoS₂ and Au–MoS₂ distances of 1.60 and 2.60 Å, respectively, perturbing greatly the electronic structure of MoS₂.

Corresponding to the results of Figure 2a, the drain current–gate voltage (I_D – V_{GS}) transfer characteristics are, respectively, achieved from the two MoS₂

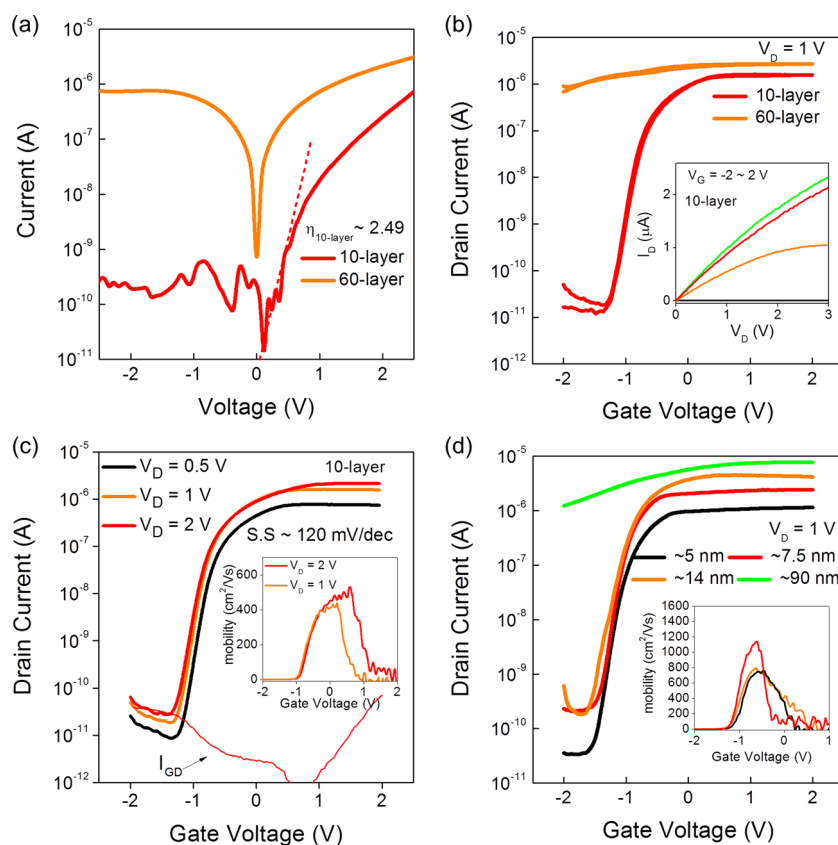


Figure 2. (a) I – V characteristics of Schottky diodes with 10- and 60-layer MoS₂. Diode behavior was clearly observed from 10-layer MoS₂ with NiO_x and Au/Ti electrodes, while the Schottky behavior of the thick MoS₂ diode appears very weak along with large reverse current. (b) I_D – V_G transfer curves of our MEFETs with 10- and 60-layer MoS₂ obtained at a V_D of 1 V; the device with 60-layer thick MoS₂ operates as an almost simple resistor. The inset shows output curves of 10-layer MoS₂ MEFET. (c) I_D – V_G transfer and I_{GD} – V_G gate leakage curves of the MEFET with 10-layer MoS₂, as measured with V_D increase; SS was ~ 120 mV/dec and inset curves show the saturation mobility plots. (d) Transfer curves of MEFETs with several MoS₂ thicknesses obtained at a V_D of 1 V; the device with 90 nm-thick MoS₂ operates as an almost simple resistor. The inset shows saturation mobility plots of 5 nm (7-layer), 7.5 nm (11-layer), and 14 nm (20-layer)-thick MoS₂ MEFETs.

nanosheet-based MEFETs as shown in Figure 2b, where MEFET operations are nicely observed from 10-layer MoS₂ devices as induced by the V_{GS} -dependent charge-depletion-modulation (which is not just to the thickness direction but also to in-plane direction).³⁷ According to the transfer curve of 10-layer MoS₂ MEFET achieved at a drain voltage (V_{DS}) of 1 V, its ON-current I_D appeared to be 1.6 μ A along with ON/OFF ratio, OFF current, and SS properties of $\sim 10^5$, ~ 30 pA, and 120 mV/dec, respectively. Inset output curves of Figure 2b display that our 10-layer MoS₂ MEFET does not show perfect linear but somewhat saturation behavior as V_{DS} increases. The saturation I_D behavior probably originates from the fact that easy pinch-off is possible in MEFET with thin MoS₂ unlike MISFET with the same MoS₂ as we compare both FETs in later discussion. On the one hand, the 60-layer MoS₂ MEFET displays a resistor type behavior in Figure 2b, where its I_D appears almost independent of V_{GS} due to the thick MoS₂ channel. This resistor type behavior is understandable, since the thick MoS₂ induces only a little Schottky barrier which would hardly cause any sufficient channel depletion even under quite a large negative gate bias.

On the basis of above results, it is regarded that a larger Schottky barrier induces more outstanding SS and ON/OFF (charge conduction/depletion) switching performance along with the sheet thickness.

One of the most important is probably how to determine the mobility (μ) of the 10-layer MoS₂ MEFET. According to the output curves, we want to use the saturation regime method for the mobility estimation as more general method; we can calculate the channel mobilities using transconductance (g_m) and the following equations,^{30,38,39} since we have g_m plots depending on V_{DS} . (See Supporting Information Figure S5, for g_m plots.)

$$g_m = \frac{\partial I_D}{\partial V_G} = \frac{qN_d\mu tW}{L}$$

So, the maximum channel mobility can be calculated as follows,

$$\mu = \frac{Lg_{\text{Max}}}{qN_d tW}$$

where g_{Max} is a maximum value of g_m plot as a function of V_{GS} . N_d is carrier density as number per cm^3 ; q is an

electronic charge; and t , W , and L are the thickness, width, and length of the channel, respectively. Now, the most wanted information to determine the mobility is the electron carrier density, n , or unintentional doping concentration, N_d , according to above equations. N_d is reported to be in a range from $\sim 5 \times 10^{15}$ to $5 \times 10^{16} \text{ cm}^{-3}$ in general MoS₂ bulk crystals;^{9,40–42} however, we here estimated the values of our own MoS₂ sheets by using two methods: the Padovani–Stratton parameter method^{34,43} on 10-layer MoS₂ on SiO₂/p⁺-Si (Supporting Information Figure S6 and Table S1) and the physical property measurement system (PPMS)-adopting AC current Hall measurements on 16 nm-thin (~ 23 -layer) MoS₂ on glass.^{44,45} We thus obtained respective n values of 1.42×10^{16} and $2.52 \times 10^{16} \text{ cm}^{-3}$. (The electron density values of 10- and 23-layer MoS₂ are $\sim 1 \times 10^{10}$ and $4.1 \times 10^{10} \text{ cm}^{-2}$ in area density, which is about 2 orders of magnitude lower than that of gate charging state in MISFET.)^{25,26} The electron density values are comparable to not only those reported for thick bulk MoS₂ in literature,^{9,40–42} but also the previously reported value from Padovani–Stratton method for 12 nm-thin (~ 17 -layer) MoS₂ on SiO₂/p⁺-Si.⁴³ Nonetheless, we chose the number density of $2.52 \times 10^{16} \text{ cm}^{-3}$ as the most reliable room temperature N_d value, since it was more directly obtained by high magnetic field (maximum 10 T) Hall measurements. Figure 2c shows the transfer curves obtained from 10-layer MoS₂ MESFET at V_{D5} of 0.5, 1, and 2 V, while its inset displays maximum saturation mobility of the device, to be $\sim 520 \text{ cm}^2/(\text{V s})$. The leakage current (I_{GD}) is also plotted in Figure 2c. The maximum I_{GD} is about 100 pA, which is quite smaller than I_D by more than 4 orders of magnitude, confirming the high quality of the NiO_x Schottky gate for the MoS₂ MESFET. A few other similarly thick MoS₂ MESFETs were fabricated to confirm this result of 10-layer MoS₂ MESFET. According to the respective transfer curves and saturation mobilities in Figure 2d and inset plots, 5 nm (7-layer), 7.5 nm (11-layer), and 14 nm (20-layer)-thick MoS₂ MESFETs display a range of high mobility between 760 and 1200 $\text{cm}^2/(\text{V s})$. (These are peak mobilities extracted by considering transconductance g_m plots, and the zero mobility beyond peak value in the plot is not much meaningful.) When the layer was too thick (90 nm), Schottky gate did not work. In this regard, our MESFET device with ~ 10 nm-thick MoS₂ channel demonstrates its intrinsic potential approaching to the intrinsic room temperature mobility of $\sim 1000 \text{ cm}^2/(\text{V s})$. The mobility difference among all the MESFETs with 5–14 nm probably come from their respective S/D contact resistances which can be different depending on the contact area. From the transfer curves of Figure 2c,d, it is also worth to note that our all MESFETs have low operational or threshold voltages between -1 and -2 V with little gate hysteresis, since the FET gate opening in the present device is based on

Schottky-driven operation on thin channels. The threshold voltage value is thus always predictable unlike the reported cases of MoS₂ MISFET,^{8,11,27,28} to be marked as an important benefit of MESFET. All the electrical properties of our MESFET devices and MoS₂ thickness profiles are summarized in detail in Supporting Information Figure S7 and Table S2.

In the present study, we even implemented the temperature-dependent Hall measurements on 16 nm-thin MoS₂ on glass in the range of 200–300 K to confirm any reliability of our measurements on the electron number density or intrinsic doping density (n or N_d). Figure 3a shows an optical image of our 4-probe MoS₂ sample for Hall measurement, while AFM topography in Figure 3b indicates the sample thickness and surface morphology. To measure the Hall resistance (R_H) of our 16 nm-thin MoS₂ sample on glass substrate, we applied AC current of 0.4 Hz and 0.1 mA as input signal frequency and amplitude, respectively, under the magnetic field (H) sweep from -10 to 10 T. As shown in Figure 3c, linear dependence of R_H on H was clearly observed with various temperatures. (The inset plot of Figure 3d again shows a magnified view of linear R_H variation under H sweep at 300 K). Since the R_H can be determined to be,⁴⁴

$$R_H = \frac{V_H}{I} = -\frac{H}{n_s q}$$

where, I is input current, V_H is Hall voltage, n_s is sheet carrier concentration (cm^{-2}), and q is electric charge, n_s can be estimated from the slope of R_H – H curve as below.

$$n_s = -\frac{1}{q} \left(\frac{\partial R_H}{\partial H} \right)^{-1}$$

On the basis of above equation, we could measure n_s values as $4.03 \times 10^{10} \text{ cm}^{-2}$ ($2.52 \times 10^{16} \text{ cm}^{-3}$) at 300 K. At 200 K, the n_s appeared to be an order of magnitude smaller than that at 300 K. From the fact that n_s is suppressed as the temperature (T) decreases as shown in Figure 3d, our MoS₂ sample exhibits a typical semi-conducting behavior with a finite activation energy value. (After room temperature sheet resistance measurements, our Hall mobility could also be extracted out, to be $\sim 200 \text{ cm}^2/(\text{V s})$, which is a few times lower than that of MESFET with NiO_x gate and it is attributed to the fact that our Hall sample cannot have the NiO_x gate which may help electron scattering reduction.)⁴⁶

For our experiment, which was intended to clearly distinguish the properties between MISFET and MESFET, we fabricated 4L-MoS₂ MESFET with NiO_x (inset optical image in Figure 4a; $W/L = 7.5/6 \mu\text{m}$) and 4-layer MoS₂ MISFET with 50 nm-thin atomic layer deposited (ALD) Al₂O₃ top gate insulator (inset in Figure 4b; $W/L = 10/5 \mu\text{m}$), while those two devices were fabricated on separate glass substrates. (The layer thickness, 4-layer was confirmed by Raman spectroscopy and AFM topography

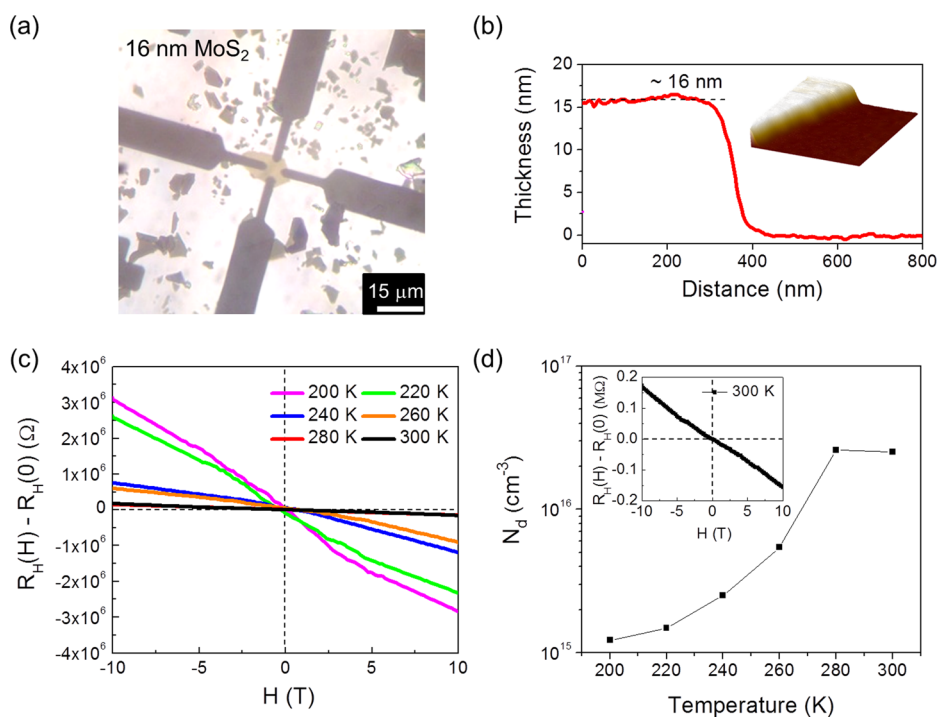


Figure 3. (a) The optical image and (b) thickness profile of 16 nm-thick MoS₂ sample for 4-probe Hall measurement. (c) Linear R_H variation under H sweep with various temperatures (T). (d) Plot of N_D versus temperature as achieved from the H sweep at different temperatures. The inset shows linear R_H variation under H sweep at 300 K.

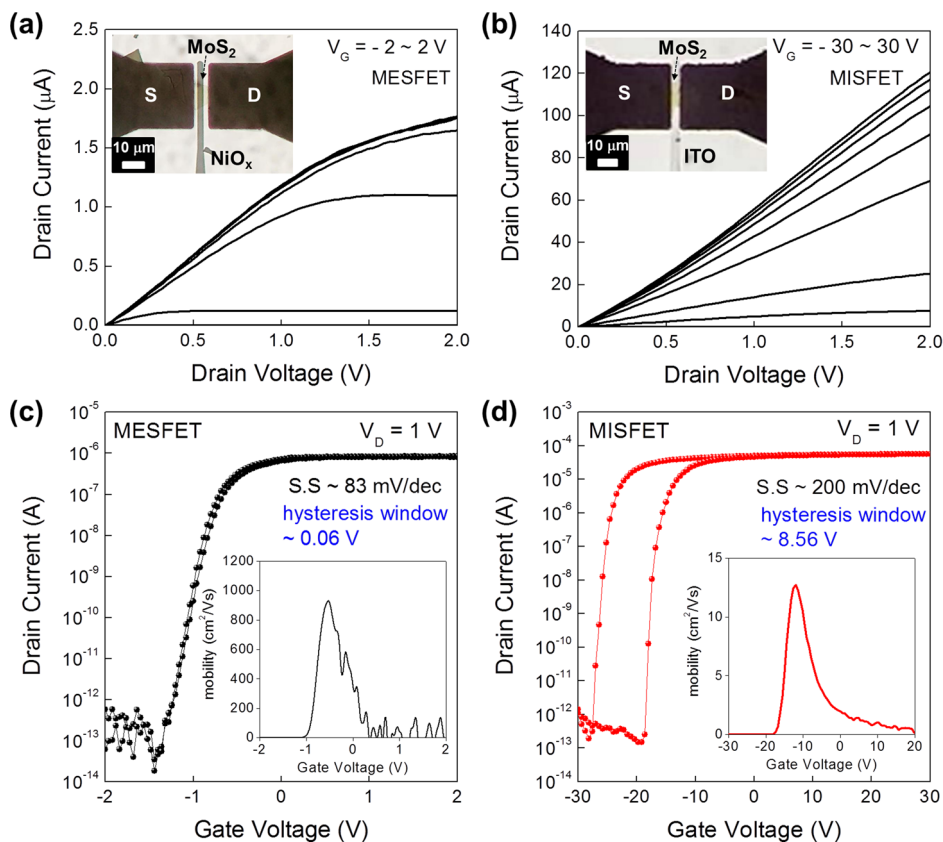


Figure 4. (a) Saturation I_D – V_{DS} behavior of our MEFET with 4L MoS₂. (b) Linear output behavior of MISFET with 4-layer MoS₂. (c) Transfer curves of our MEFET device show SS of only 83 mV/dec, 60 mV hysteresis window, on/off ratio of 1×10^7 , and $\sim 950 \text{ cm}^2/\text{V s}$ for saturation mobility (inset). (d) Transfer curves of MISFET with ITO gate display 8.56 V hysteresis window, 200 mV SS, on/off ratio of 6×10^7 , and its linear mobility of $12.5 \text{ cm}^2/\text{V s}$.

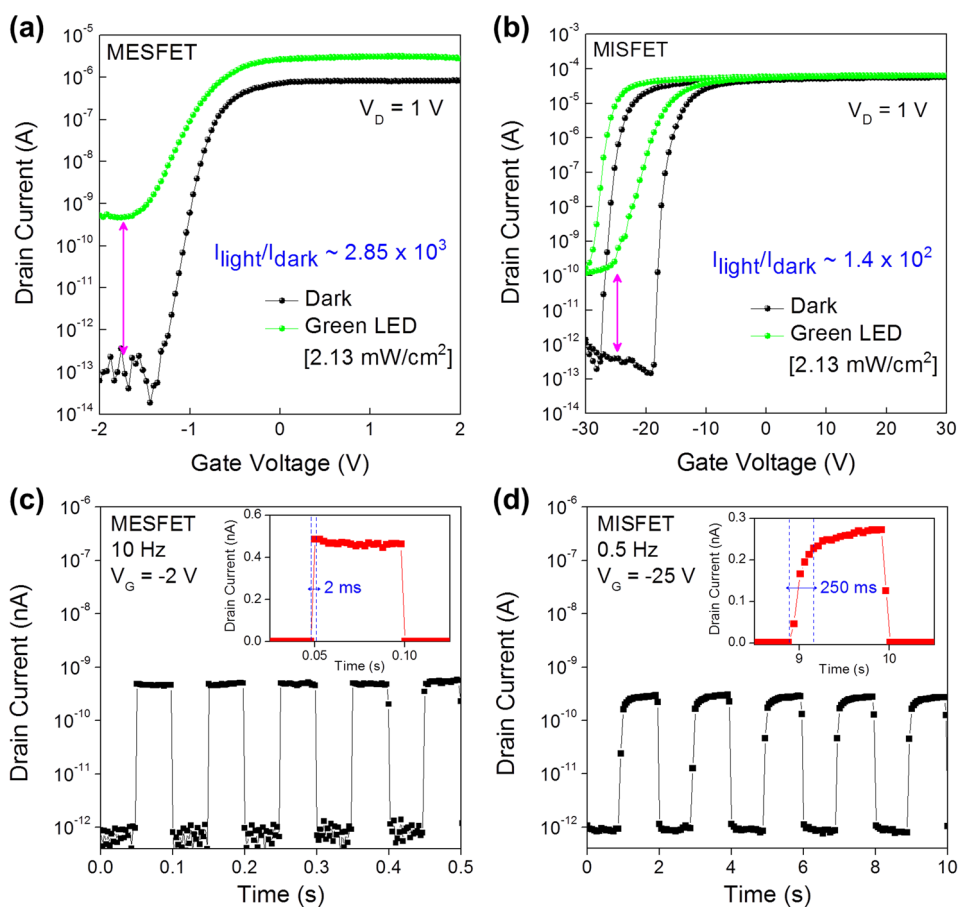


Figure 5. Photoinduced transfer curves of our (a) MEFET and (b) MISFET devices with transparent gate electrode, as obtained under green LED. Photo-to-dark ratio of MEFET ($I_{\text{light}}/I_{\text{dark}}$) appears an order of magnitude higher than that of MISFET device. Dynamic photoswitching behavior of our (c) MEFET and (d) MISFET devices; according to the respective insets, MEFET displays 2 orders of magnitude higher switching speed than MISFET along with slightly higher photocurrent.

in Supporting Information Figure S8.) According to the respective optical images, MEFET channel with semi-transparent NiO_x gate appears quite transparent on glass, while MISFET top-gate was covered by fully transparent indium–tin-oxide (ITO) electrode (back illumination was taken for photograph). Their respective output curves are shown in Figure 4, panels a and b, along with their inset device images for comparison; MEFET shows some saturation behavior unlike MISFET, which does appear almost completely linear. Such saturation behavior in MEFET with 4-layer MoS_2 comes from the easy channel-depletion (pinch-off) in drain side with V_{DS} increase which would cause even negative V_{GD} , while the on-state gate voltage of MISFET with the same 4-layer always provides the channel with charging-induced high density electron carriers to seldom cause such pinch-off in drain side channel. The transfer curves of the two devices are displayed in Figure 4c,d, where MISFET shows ~ 60 times higher drain current than MEFET under 1 V drain voltage. So, MISFET would be more advantageous in practical electronic applications requesting high current if reasonable threshold voltages are reproducibly controlled. However, according to the mobility plots in Figure 4d

and its inset, the linear mobility of MISFET was limited to only $\sim 12.5 \text{ cm}^2/(\text{V s})$ along with a large swing ($SS = \sim 200 \text{ mV/dec}$), large hysteresis (8.56 V) and too much negative threshold voltages ($\sim -20 \text{ V}$), while MEFET shows $\sim 950 \text{ cm}^2/(\text{V s})$ as a saturation mobility, little hysteresis (0.06 V), and sharp swing ($SS = 83 \text{ mV/dec}$) near -2 V (Figure 4c and inset). For the linear mobility (μ_{lin}) of MISFET with Al_2O_3 dielectric, conventional equation was used as $\mu_{\text{lin}} = (1/V_{\text{D}}C_{\text{ox}})(L/W)(\partial I_{\text{D}}/\partial V_{\text{G}})$, where C_{ox} is the dielectric capacitance.¹⁰ All the differences originate from the fact that the 4-layer channel of MISFET has two ~ 3 orders of magnitude higher carrier density ($\sim 10^{19} \text{ cm}^{-3}$ or $\sim 10^{12} \text{ cm}^{-2}$) drawn by gate charging,^{25,26} which would however cause serious trap-induced interface scattering during operation. Moreover, we also suspect that ALD-process for high-k dielectric induces H atoms to MoS_2 channel by diffusion, and that the diffused H impurities play as scattering center.^{47–49} Again, the high mobilities from our MEFETs are explained with the low densities of electron carrier and scattering center in the thin MoS_2 channel itself, while the MEFET operation also allows scattering-minimized transport without gate charging-induced interferences.^{30,31}

Such a high intrinsic mobility in MoS₂ MESFET was supported by implementing phototransistor measurements, where 520 nm-wavelength green light emitting diode (LED) was used for light-detection performance comparison between the MISFET and MESFET (of Figure 5a,b). According to dynamic photoswitching behavior in Figure 5c,d (respectively with 10 and 0.5 Hz LED switching), the fast response time of 2 ms is obtained in MESFET with 4-layer MoS₂ at low gate voltages of -2 V, while more than 250 ms appears for 4-layer MoS₂ MISFET with ALD Al₂O₃ top-gate dielectric at high gate voltages of -25 V. Moreover, their respective phototo-dark I_D ratios are estimated to be 1.4×10^2 and 2.85×10^3 for MISFET and MESFET devices, which means MESFET photodetector is 100 times faster and 10 times more sensitive, although in fact 250 ms in our MoS₂ MISFET phototransistor is still quite good among many MoS₂ photo FET reports as summarized in Table 1.^{50–53} We regard that the reason for the fast photoresponse is definitely related to 50–100 times higher mobility of MESFET than that of MISFET. (Responsivity of our MESFET under green LED was estimated to be 5000 and 1.1 A/W for ON and OFF states, respectively.)

As our final experimentation, we also attempted to demonstrate organic LED (OLED) switching using our MESFET. Despite the small I_D current of our MESFET, OLED switching was nicely operated by using the MESFET under low voltages ($V_{IN} = V_{DD} = 2$ V) (related circuits and demonstration details are found in Supporting Information Figure S9 and OLED operation video.avi file). This indicates that our MoS₂ nanosheet-based MESFET device is quite useful in electronics and

TABLE 1. Summary Table of Dynamic Switching Properties and Photosensitivity Measured from MoS₂-Based Phototransistor in Air Ambient

	$I_{\text{light}}/I_{\text{dark}}$ ratio	response time	light power	wavelength
Our MESFET	2.85×10^3	~ 2 ms	2.13 mW/cm ²	520 nm
Our MISFET	1.4×10^2	~ 250 ms	2.13 mW/cm ²	520 nm
ref47	$\sim 3 \times 10^2$	~ 1 s	6 mW/cm ²	532 nm
ref48	$\sim 5 \times 10^2$	~ 1 s	50 mW/cm ²	633 nm
ref49	Unknown	50 ms	80 μ W	550 nm
ref50	~ 30	4 s	4.25 μ W	561 nm

optoelectronic applications, particularly for low voltage electronics.

CONCLUSION

In summary, we have fabricated MoS₂-based metal semiconductor field-effect transistors (MESFETs) with NiO_x Schottky electrode, where the maximum mobilities or carrier transport behavior of the Schottky devices may hardly be interfered by interface traps or an on-state gate field. Our MESFETs with 4–10 layer MoS₂ demonstrate high intrinsic-like mobilities of 500–1200 cm²/(V s) at a certain low threshold voltage between -1 and -2 V with little gate bias-induced hysteresis, while they also exhibit very fast photoswitching in 2 ms playing as phototransistors with semitransparent NiO_x window. We conclude that our MESFET with van der Waals interface between NiO_x gate and MoS₂ channel would be a promising model nano FET with high speed, predictable low threshold voltage, and sharp swing behavior, to be applied for future nanoelectronics and nano-optoelectronics.

METHODS

Device Fabrication. All the MoS₂ nanosheets were mechanically exfoliated from bulk MoS₂ crystals (SPI supplies, natural molybdenite) and transferred to 285 nm SiO₂ grown on heavily doped p-type silicon substrate (285 nm-thick SiO₂/p⁺-Si) or to glass substrate (Eagle 2000) by using a standard scotch tape method. For the source (S) and drain (D) ohmic contact electrodes Au/Ti (70/30 nm) were patterned (deposited at room temperature) on top of MoS₂ flakes by using photolithography, lift-off, and DC-sputter deposition processes. As the first layer, the Lift-Off-Layer (LOL: using a solution, LOL 2000, Micro Chemical) was coated, followed by thermal curing at 115 °C for 2 min. Then, photoresist (PR: SPR 3612, Micro Chemical) as the second layer was coated and baked at 95 °C for 2 min. The samples were exposed to UV light for 5 s under photomask aligner for S/D pattern. The samples were subsequently patterned with metal-ion-free (MIF) developer solution, and sequential DC-magnetron sputter-deposition of 30 nm-thin Ti and a 70 nm thin Au layers (Au/Ti) were carried out. For the liftoff process to finally define S/D electrode, acetone and LOL remover solvents were used. After that, the device was annealed at 250 °C with N₂ flow in rapid thermal annealing (RTA) system, to remove polymer residue and simultaneously to reduce contact resistance. A 150 nm-thick NiO_x Schottky contact gate electrode for MESFET (or Pt and Pd Schottky electrodes for diode) was patterned (deposited at room temperature) between the S/D electrodes by the same lithography and liftoff

methods as performed for S/D contact, but for the deposition of NiO_x, we used the thermal evaporation of NiO_x powder while Pt and Pd films were deposited by DC-magnetron sputtering. The channel length, L of our MESFET was 6 μ m and the gate length of NiO_x, Pt, and Pd was 3 μ m in channel direction. NiO_x evaporation would hardly contaminate MoS₂ according to I – V curves in Supporting Information Figure S10, which was achieved with and without NiO_x electrode on top of MoS₂.

Measurements and Characterizations. The thickness of MoS₂ flakes were first detected by optical microscope, then the exact layer number of each MoS₂ was characterized by AFM (XE-100, psia) and Raman spectroscopy. SEM was also necessary for the top view of our MESFET. All current–voltage (I – V) characteristics were measured by using a semiconductor parameter analyzer (HP 4155C, Agilent Technologies) in ambient air and room temperature. The Hall measurements for electron density determination of 23L-thin MoS₂ on glass were implemented at various temperatures in the range of 200–300 K by applying AC current of 0.4 Hz and 0.1 mA as input signal frequency and amplitude, respectively, under the magnetic field (H) sweep from -10 to 10 T. We used the physical property measurement system (PPMS) for Hall measurements. Photoinduced static and dynamic signals were measured by using 520 nm-wavelength green LED (2.13 mW/cm²), which was pulsed in square wave for the dynamic photoswitching by pulse generator (AFG3022B, Tektronix). For the OLED operation, we used the conventional OLED devices (Courtesy from Samsung Display Co.).

Conflict of Interest: The authors declare no competing financial interest.

Supporting Information Available: Optical images of several MoS₂ nanosheets which play as the channel of MESFET, schematic energy band diagram, schematic illustrations for MESFET operations, comparison between NiO_x and pure metal contact for MESFET, transconductance plots of 10-layer MoS₂ MESFET, Padovani–Stratton parameter for electron density estimation, reproducibility of MESFET, AFM profile and Raman spectra obtained from 4-layer MoS₂, OLED operation using our MESFET. The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nano.5b02785.

Acknowledgment. The authors acknowledge the financial support from NRF (NRL program: Grant No. 2014R1A2A1A0-1004815), Nano-Materials Technology Development Program (Grant No. 2012M3A7B4034985), NRF of Korea (Grant No. 2011-00183306), KISTI supercomputing center (Project No. KSC-2013-C3-008) and Brain Korea 21 plus Program.

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