Photocurrent Switching of Monolayer MoS$_2$ Using a Metal–Insulator Transition

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**Supporting Information**

**ABSTRACT:** We achieve switching on/off the photocurrent of monolayer molybdenum disulfide (MoS$_2$) by controlling the metal–insulator transition (MIT). N-type semiconducting MoS$_2$ under a large negative gate bias generates a photocurrent attributed to the increase of excess carriers in the conduction band by optical excitation. However, under a large positive gate bias, a phase shift from semiconducting to metallic MoS$_2$ is caused, and the photocurrent by excess carriers in the conduction band induced by the laser disappears due to enhanced electron–electron scattering. Thus, no photocurrent is detected in metallic MoS$_2$. Our results indicate that the photocurrent of MoS$_2$ can be switched on/off by appropriately controlling the MIT transition by means of gate bias.

**KEYWORDS:** MoS$_2$, metal–insulator transition, photocurrent switching, optical phonon scattering, electron–electron scattering

Semiconducting transition metal dichalcogenides (s-TMDs), including MoS$_2$, WSe$_2$, and WS$_2$, have attracted worldwide attention as alternative future electronic materials. The single atomic thickness and the absence of dangling bonds of s-TMDs is expected to benefit future submicroelectronics since the surface scattering of electrons in the channel materials—an important limiting factor of Si devices as the device size scales down—is suppressed with s-TMDs. Furthermore, unlike graphene, the optical band gap of s-TMDs broadens their application area to optoelectronic devices such as photodetectors and solar cells. 

The modulation of the electrical properties of MoS$_2$ has recently been achieved by controlling the gate bias. Depending on the carrier concentration $n$, MoS$_2$ changes from an insulator to a metal. This constitutes the so-called metal–insulator transition (MIT).$^9,10$ The dramatic change in the electrical behavior of MoS$_2$ significantly affects its optical properties. For instance, at low carrier concentrations, MoS$_2$ illuminated by a laser with $E_l > E_g$ forms electron–hole pairs, excitons, owing to a strong Coulomb interaction such that luminescence is dominated by exciton states. However, at high carrier concentrations, excitons bind with electrons, forming negative trions.$^{11–13}$ Then, the luminescence peak shifts to a lower energy than the exciton binding energy.

Generally, the photocurrent of MoS$_2$ can be generated due to a few different causes. In addition, its modulation is demonstrated by controlling the barrier height or band alignment using the gate bias$^{14}$ and by exposing it to gas adsorbates.$^{15}$ In the channel of MoS$_2$ FET, a strong photovoltaic effect is observed.$^{14,16}$ In addition, a large and tunable thermoelectric effect is observed from the layered materials.$^{17,18}$ Therefore, the metal–MoS$_2$ junction can exhibit intense photothermoelectric effects owing to the different Seebeck coefficients of the layers. MoS$_2$ has shown various ways of optical response to the light, which are correlated with different physical aspects of MoS$_2$. Similarly, the photocurrent of MoS$_2$ is expected to be affected by the metallicity of the MoS$_2$ channel, the latter being controlled by the gate bias. However, an MIT-based photocurrent modulation has not been reported yet.

In this study, we characterize the photocurrent as a function of the back gate bias, which transforms insulating MoS$_2$ to metallic MoS$_2$. At the insulator state, the photocurrent significantly increases in response to optical input, resulting from an increase in the amount of excess carriers. However, at the metallic state, the photocurrent is not generated under illumination. The absence of a photocurrent in the metallic state is attributed to increased carrier–carrier scattering that nullifies the generation of excess carriers in the MoS$_2$ conduction band. Our results indicate that the photocurrent can be switched on/off using the MIT transition.

A monolayer MoS$_2$ was synthesized using chemical vapor deposition.$^{20}$ Monolayer flakes grew into triangles with each side length of about 30–50 μm. They were randomly located.
MoS2 FET was additionally fabricated on an exfoliated, barrier height (SBH) on the photocurrent, the monolayer positive gate bias. The n-type behavior of MoS2 is reportedly occurs at approximately speci.

The transition of MoS2 from semiconductor to metal is attributed to S orbital vacancies. When the transition occurs, attributed to increased electron mean free path by the Schottky barrier height reduction, the Schottky barrier height gradually decreases with the increase in the carrier mobility. The same procedure was applied for the device on the h-BN substrate.

As shown in Figure 2a, the temperature-dependent transport behavior of MoS2 reveals that the \( I_{ds} \) increases with temperature when \( V_{gs} \) increases. The transition of MoS2 from semiconductor to metal is assumed to occur when the Fermi wavelength \( \lambda_F \) of electrons is longer than the electron mean free path \( L_m \). Therefore, the transition is theoretically predicted to occur at the critical charge concentration of \( n_c > 1 \times 10^{27} \text{cm}^{-3} \). The transition occurs at approximately \( V_{MIT} = 60 \text{ V} \) from a device on the SiO2 substrate, as shown in Figure 2a. Both the n-type behavior and MIT are reproduced in the MoS2 FET on the h-BN substrate, as shown in Figure 2b, although \( V_{MIT} \) for the device on h-BN is about 45 V.

Figure 2c shows the Schottky barrier heights (SBHs) obtained only from Au-MoS2 junctions, both on SiO2 and h-BN substrates at 300 K. The SBH gradually reduces with increasing \( V_{gs} \). A higher SBH is observed when the device is on the SiO2 substrate rather than on the h-BN substrate. In Figure 2c, the SBHs of MoS2 with two terminals are approximately 68 and 40 meV at \( V_{gs} = -60 \text{ V} \) for SiO2 and h-BN, respectively. For a comparison, MoS2 devices with four electrodes were prepared on SiO2 and h-BN substrates as well. The theoretical model and procedure to evaluate SBHs and their values from four-electrode devices are described in Supporting Information 3.

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MoS₂ junction occurs. Thus, our narrow beam diameter prevents the simultaneous occurrence of both photothermal and photovoltatic effects at the junction. This is due to the different Fermi levels and Seebeck coefficients of the Au electrode and the MoS₂ contacts.

The laser is absorbed not only by MoS₂, but mostly by the Si substrate after passing through the SiO₂ or h-BN gate oxide. It can ionize trap sites at the SiO₂/MoS₂ interface resulting in an electric field at the channel in addition to \( V_{gs} \). The photoinduced gating effect is different from the electrostatic Si back gate \( V_{gs} \). In addition to \( I_{ds} \) caused by \( V_{gs} \), photoinduced gating increased the current that originated from the metal electrode, and not due to interband transition from the valence band of MoS₂. In order to separate the photoinduced gating current \( I_{ds-pg} \) from the photocurrent by excess carriers \( I_{ds-pc} \), we compiled a characterization system outlined in Supporting Information 4. Since the photoinduced gating effect is caused by trapped charges, it has a considerably long time constant\(^{26,28}\) (Supporting Information 5). Therefore, using chopped light with a frequency of 100–300 Hz, \( I_{ds-pg} \) can be selectively filtered out from the photocurrent measurement.\(^{26}\)

The inset of Figure 3a is the optical image of the MoS₂ FET patterned using the reactive-ion etching (RIE) method using SF₆ gases. The optical response of MoS₂ on the SiO₂ substrate in the labeled area of the inset was investigated using different \( V_{gs} \) and laser powers. The areal photocurrent distribution is shown in Figures 3a and b. Under \( V_{gs} < V_{th} < V_{MIT} \) and a power of 1 \( \mu \)W, and wavelength of 532 nm, we observed a high \( I_{ds-pc} \) through the MoS₂ FET on a SiO₂ substrate, as shown in Figure 3a. The channel demonstrates considerably higher \( I_{ds-pc} \) than that at the MoS₂–metal junction. However, under \( V_{gs} > V_{MIT} > V_{th} \) and a power of 10 \( \mu \)W, the \( I_{ds-pc} \) from the channel significantly drops, as shown in Figure 3b. These results indicate that the photocurrent switching using \( V_{gs} \) results from the optical excitation inside the MoS₂ channel, rather than from the SBH variation.

Figure 4a–f depicts the photocurrent density \( J_{ds-pc} \) of MoS₂ on SiO₂ dielectrics as a function of \( V_{gs} \). Under a large negative gate bias, MoS₂ exhibits semiconducting behavior, under which

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**Figure 3.** Photocurrent mapping images of the MoS₂ FET under (a) \( V_{gs} < V_{th} < V_{MIT} \) and (b) \( V_{gs} > V_{MIT} > V_{th} \). The inset shows the optical image of the MoS₂ FET. The dashed lines indicate the edge of the MoS₂ channel. The unit of the color scale is ampere (A).

**Figure 4.** \( J_{ds-pc} \) versus \( V_{gs} \) of the MoS₂ FET on the SiO₂ substrate (a) before annealing and (b) after annealing at 130 °C for 2 h in a vacuum system. The schematic diagrams of scattering of excess carriers when (c) \( V_{gs} < V_{MIT} \) and (d) \( V_{gs} > V_{MIT} \). (e) \( J_{ds-pc} \) versus \( V_{gs} \) of the MoS₂ FET on the h-BN substrate without annealing. (f) \( J_{ds-pc} \) versus \( T \) of the MoS₂ FET on the SiO₂ substrate measured at \( V_{gs} = 60 \) V (black circles) and \( V_{gs} = -75 \) V (red circles).
a significant generation of $I_{ds-pc}$ is observed, as shown in Figure 4a. As the gate bias is swept toward the positive side, the photocurrent of MoS$_2$ fluctuates but gradually decreases. At $V_{gs} > 45$ V, $I_{ds-pc}$ becomes zero. It is also possible that the laser heats up and reduces the resistance of MoS$_2$, resulting in an increase in the photocurrent. However, the absorption of 532 nm by MoS$_2$ is not affected by $V_{gs}$. Therefore, no enhanced photoconductivity is caused by the laser from the photothermal heating of MoS$_2$. Thus, the absence of $I_{ds-pc}$ above $V_{MIT}$ stems from other mechanisms, rather than absorption changes. In addition to the absence of $I_{ds-pc}$ in metallic MoS$_2$, the photocurrent fluctuates during the gate bias sweep in Figure 4a.

The photocurrent $I_{ds-pc}$ of MoS$_2$ is expressed as $I_{ds-pc} = (W/L)V_{gs}\sigma_{pc}$, where $W$ and $L$ are the width and length of the channel, respectively, and $\sigma_{pc}$ is the electrical conductivity by excess charges. The only parameter depending on $V_{gs}$ in the above expression of $I_{ds-pc}$ is the electrical conductivity. The change in the electrical conductivity can be further described by $\sigma_{pc} = q(\mu_\nu + \mu_\rho)\Delta n_{ex}$ where $\mu_\nu$ ($\mu_\rho$) is the electron (hole) mobility and $\Delta n_{ex}$ ($=\Delta \rho_{ex}$) is the number of excess electrons (holes) that is expressed with the number of absorbed photons $\eta$ and nonradiative carrier lifetime $\tau_\eta$. Since some amount of excess carriers recombine before they are attracted to the electrodes and are trapped in the defect states, the net photoconductivity $\sigma_{pc}$ of electron excess carriers can be expressed as $\sigma_{pc} = q\mu_\nu(\Delta n_{ex} - p_\rho)$, where $p_\rho$ is the trapped electron concentration. MoS$_2$ contains numerous trap sites with different densities and energy levels. Therefore, the fluctuating $I_{ds-pc}$ shown in Figure 4a is expected to reflect the trap site density $N_T$ at different energy levels inside MoS$_2$. However, as $V_{gs}$ increases to a positive value, more trap sites are occupied by channel electrons. In such a case, $I_{ds-pc}$ should not reduce at high positive $V_{gs}$.

In order to reduce the effect of trap sites on $I_{ds-pc}$, our device in Figure 4a was annealed at 130 $^\circ$C for 2 h in a vacuum chamber under a base pressure of $5 \times 10^{-6}$ Torr. After annealing, $I_{ds-pc}$ in Figure 4b increased by an order of magnitude compared to that shown in Figure 4a. In addition, the fluctuations of $I_{ds-pc}$ were reduced. The enhancement in $I_{ds-pc}$ is attributed to the removal of trapped impurities $p_\rho$ of MoS$_2$. After annealing, the correlation between $I_{ds-pc}$ and $V_{gs}$ was clearly manifested; i.e., a complete elimination of $I_{ds-pc}$ was achieved when the metallicity of MoS$_2$ shifted from semiconductor to metal. In the expression for the net photoconductivity $\sigma_{pc} = q\mu_\nu(\Delta n_{ex} - p_\rho)$, even after the annealing, we still have some trap sites, and $p_\rho$ depends on $V_{gs}$ because the number of active trap sites can change depending on $E_p$. From the Shockley–Read–Hall recombination process, $\tau_\rho$ is inversely proportional to $N_T$, $\tau_\rho \approx 1/N_T$. Thus, the number of trap sites should decrease as $V_{gs}$ increases, resulting in an increase of $\tau_\rho$. Then, $\Delta \sigma_{pc}$ should increase since $\Delta n_{ex}$ is proportional to $\tau_\rho$. Unfortunately, we did not observe such behavior from the gate-dependent photocurrent.

In $\sigma_{pc} = q\mu_\nu(\Delta n_{ex} - p_\rho)$, we assume that $\mu_\nu$ remains constant regardless of metallicity of MoS$_2$. However, our results lead us to speculate on the variation of $\mu_\nu$ of excess carriers depending on the gate bias. One of the possible scenarios is the enhanced carrier scattering rate when MoS$_2$ switches from semiconductor to metal using the gate bias. Therefore, the increase in $I_{ds-pc}$ by excess carrier $\Delta n_{ex}$ can be suppressed by the increase of carrier scattering in the MoS$_2$ channel. This will reduce the mobility of excess carriers since $\mu_\nu$ depends on the scattering time $\tau_\nu$, $\mu_\nu = q\tau_\nu/m_\nu$, where $\tau$ is the carrier scattering time and $m_\nu$ is the electron mass. In such a case, the photocurrent caused by excess carriers $I_{ds-pc}$ can drop to zero. For instance, when $n > n_c$, the carrier scattering is dominantly mediated by electron–electron scattering. The scattering time of electron scales with carrier concentration as $n^{-1/3}$.

A similar behavior of the photocurrent has been observed for graphene. When graphene was heavily doped to resemble a metal, negative photoconductivity was observed owing to increased carrier scattering. However, lightly doped graphene resembling a semiconductor showed positive photoconductivity from the increase in the number of excess carriers. Another possibility is that, with the increase in the number of excess carriers $>10^{13}$/cm$^2$, pre-existing excitons at low $n$ start to form trions with the same charge and mass three times larger than the electron mass. In order to emphasize on the photocurrent switching mechanism using MIT, schematics shown in Figure 4c and d were drawn. When $V_{gs} < V_{MIT}$ in Figure 4c, the photocurrent generation is attributed to the increase in the number of excess carriers by optical excitation. However, when $V_{gs} > V_{MIT}$ in Figure 4d, the excess carriers are severely scattered by the increased channel current, and no photocurrent is observed.

In the case of MoS$_2$ FET on the h-BN layer, the dependence of the photocurrent on the back gate bias is observed in a considerably narrower voltage window, from $V_{gs} = -60$ to $40$ V, as shown in Figure 4e. The power dependence of the photocurrent in this $V_{gs}$ range indicates that the measured current is attributed to excess carriers. When $V_{gs} > -40$ V, no photocurrent is detected, regardless of the laser power. The peculiar phenomenon of the MoS$_2$ photocurrent on the h-BN substrate is associated with a lower SBH, which, via the photoinduced gating effect, enables the carrier concentration of MoS$_2$ to easily reach above $n_c$. Because of a relatively high SBH on SiO$_2$, the back-gate bias for MIT is slightly shifted by the photoinduced gating effect.

In order to understand the transport behavior of excess carriers of MoS$_2$, the photocurrent is characterized as a function of the temperature. Figure 4f shows the $I_{ds-pc}$ versus $T$ curve measured at $V_{gs} = -75$ V, at which MoS$_2$ behaves as the semiconductor. As the temperature decreases, the photocurrent increases. Since we probe only the temperature dependence of excess carriers, it should not follow the intrinsic behavior of a semiconductor, $J \sim J_0 \exp(-E_g/k_B T)$, where $J_0$ is the current density at a given temperature and $E_g$ is the bandgap of MoS$_2$. The $I_{ds-pc}$ versus $T$ curve should reflect the dependence of excess carrier scattering on temperature. The photocurrent decreasing with temperature, as shown in Figure 4f, is obtained under constant light power. Thus, decreasing $I_{ds-pc}$ with temperature indicates an increase in phonon scattering, when the temperature becomes higher. The power law fit of $I_{ds-pc}$ as a function of $T$ reveals a fitting exponent of 1.67. Since during the experiment, $\Delta \rho_{ex} \approx \tau_\rho$ and electric field $E$ do not change, the dependence of $I_{ds-pc}$ on $T$ is dominated by $\mu_\nu$. Thus, the dependence of $I_{ds-pc}$ on $T$ can be explained by the scattering of excess carriers by optical phonons. Unlike graphene, which has a large optical phonon energy of 190 meV, MoS$_2$ has a rather low energy of optical phonons, 48 meV ($\omega_{LO}/2\pi = 400$ cm$^{-1}$), which play as a dominate scattering centers in our temperature range. Therefore, excess carriers are interfered by optical phonons at around 300 K, rather than acoustic phonons. The contribution of acoustic phonons to the transport of excess carriers is expected to appear below 100 K after the suppression of optical phonons.
In contrast to the temperature dependence of $J_{\text{DPC}}$ at $V_g = -75$ V, the photocurrent density $J_{\text{DPC}}$ obtained at $V_g = 60$ V does not change with temperature. In this metallic region, electron–electron interaction is the dominating scattering source. Usually, electron–electron scattering does not exhibit temperature dependence. Below 10 K, electron–electron scattering is expected to show dependence on $T^2$ in highly crystalline samples. Since electron–phonon scattering and electron–impurity scattering are much stronger in our temperature range than electron–electron scattering, no temperature dependence is postulated to occur under our experimental conditions.

In summary, we studied the photoelectric behavior of MoS$_2$ and its electrical properties were modulated using the back gate bias. The photocurrent detected was attributed to excess carriers at the semiconducting state. In the metallic state, however, increased scattering of excess carriers completely eliminated the photocurrent. The photoc conductivity switching can form the basis for establishing logic gates under light illumination.

**ASSOCIATED CONTENT**

• Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.6b03689.

Characterization of CVD-grown monolayer MoS$_2$ using Raman spectroscopy, extraction of SBH, experimental setup, and the shift of threshold voltage due to the photogating effect (PDF)

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Author Contributions

J.H.L. and H.Z.G. contributed to this work equally. J.H.L. and H.Z.G. set up the experiment and acquired the data. H.K. grew the MoS$_2$ monolayer. B.H.M. analyzed the MoS$_2$ FET on the h-BN substrate. J.H.K. and H.C. fabricated the MoS$_2$ FETs. Y.H.L. and S.C.L. organized the data and prepared the manuscript.

Notes

The authors declare no competing financial interest.

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