High-Responsivity Multilayer MoSe$_2$ Phototransistors with Fast Response Time

Hyejoo Lee$^1$, Jongtae Ahn$^2$, Seongil Im$^2$, Jiyoung Kim$^3$ & Woong Choi$^1$

There is a great interest in phototransistors based on transition metal dichalcogenides because of their interesting optoelectronic properties. However, most emphasis has been put on MoS$_2$ and little attention has been given to MoSe$_2$, which has higher optical absorbance. Here, we present a compelling case for multilayer MoSe$_2$ phototransistors fabricated in a bottom-gate thin-film transistor configuration on SiO$_2$/Si substrates. Under 650-nm-laser, our MoSe$_2$ phototransistor exhibited the best performance among MoSe$_2$ phototransistors in literature, including the highest responsivity ($1.4 \times 10^5$ AW$^{-1}$), the highest specific detectivity ($5.5 \times 10^{13}$ jones), and the fastest response time (1.7 ms). We also present a qualitative model to describe the device operation based on the combination of photoconductive and photogating effects. These results demonstrate the feasibility of achieving high performance in multilayer MoSe$_2$ phototransistors, suggesting the possibility of further enhancement in the performance of MoSe$_2$ phototransistors with proper device engineering.

There is a great interest in transition metal dichalcogenides (TMDs), which are composed of vertically stacked layers held together by van der Waals interactions, because of their interesting electronic, optical, and chemical properties$^{1,2}$. Unlike graphene, the existence of bandgaps in TMDs$^{3,4}$ such as MoS$_2$ or MoSe$_2$ offers an attractive possibility of using these layered materials in various device applications. Field-effect transistors (FETs) based on single or multilayer MoS$_2$ exhibit outstanding performance metrics, including high on/off-current ratio ($\sim 10^9$), high mobility ($\sim 100$ cm$^2$V$^{-1}$s$^{-1}$) and low subthreshold swing ($\sim 70$ mV decade$^{-1}$)$^{5,6}$. As the band structure of TMDs depends on their physical thickness$^{3,4}$, FETs based on TMDs are especially promising for optoelectronic devices such as phototransistors. As the optoelectronic properties of early MoS$_2$ phototransistors improved$^{7–10}$, high responsivity ($\sim 10^5$ AW$^{-1}$) and fast response time ($\sim 1$ ms) were obtained in MoS$_2$ phototransistors with device engineering such as HfO$_2$ encapsulation or ferroelectric gate dielectrics$^{11–13}$.

While MoS$_2$ has been the most extensively investigated TMD for device applications, the higher optical absorbance of MoSe$_2$ suggests that MoSe$_2$ could be more suitable than MoS$_2$ for the application of phototransistors. However, little attention has been given to the optoelectronic properties of MoSe$_2$ phototransistors, which has been less impressive than those of MoS$_2$ phototransistors (responsivity: 0.01–238 AW$^{-1}$, response time: 5–400 ms)$^{13,15–19}$. Therefore, in this study, we explore the optoelectronic properties of MoSe$_2$ phototransistors fabricated with mechanically-exfoliated multilayer flakes on SiO$_2$/Si substrates. Our best-performance MoSe$_2$ phototransistor in a simple bottom-gate thin-film transistor configuration exhibits high responsivity ($\sim 1.4 \times 10^5$ AW$^{-1}$) and fast response time ($\sim 1.7$ ms) under 650-nm-laser surpassing previously reported MoSe$_2$ phototransistors. We also investigate the dependence of photocurrent on gate voltage and optical power density to describe the device operation based on photoconductive and photogating effects. These results demonstrate the feasibility of achieving high performance in MoSe$_2$ phototransistors without complicated device structures, suggesting that the performance of MoSe$_2$ phototransistors could be further enhanced by the combination of optimized device architecture and processing.

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Results and Discussion

Before fabricating MoSe₂ transistors, we first measure the optical absorbance of MoSe₂ crystals and mechanically exfoliated flakes on sapphire substrates across visible and near-infrared spectral ranges (Fig. 1(a)). The MoSe₂ crystal is thicker than 100 μm and the thickness of exfoliated MoSe₂ flakes are in the range of 20–80 nm. Both samples show two excitonic absorbance peaks A and B at 1.55 eV and 1.82 eV, respectively, which is consistent with literature20. Next, multilayer MoSe₂ transistors are fabricated on SiO₂/Si substrates. Figure 1(b) shows the optical microscopy image of a completed MoSe₂ transistor along with its schematic cross-section. The measured transfer curve of an MoSe₂ transistor in Fig. 1(c) shows asymmetric ambipolar behavior with strong n-type characteristic (MoSe₂ thickness (t) = 25 nm). For electron transport without light, the MoSe₂ transistor exhibits on/off-current ratio (I_on/I_off) of 10⁵ and field-effect mobility (μ_FE) of 50.6 cm²V⁻¹s⁻¹ extracted from μ_FE = L(dI/dVg)/WCoxVd, where L is the channel length (5 μm), I_d is drain current, V_g is gate voltage, W is the channel width (27 μm), Cox is the oxide capacitance, and V_d is the drain voltage (1 V). For hole transport without light, I_on/I_off of 10⁴ and μ_FE of 2.8 cm²V⁻¹s⁻¹ are obtained. The n-type-dominant ambipolar behavior of MoSe₂ transistors with Ti/Au electrodes was also observed in literature21,22. The output curves in Fig. 1(d) show linear region at low V_d suggesting decent contact properties. The transfer and output characteristics of an MoSe₂ transistor in Fig. 1(c,d) show the increase of I_d with the power density of incident light.

The photocurrent (I_ph) of phototransistors based on transition metal dichalcogenides such as MoSe₂ is known to be dominated by photovoltaic effect and photogating effect23. In photovoltaic effect, photogenerated excess carriers increase conductivity resulting in increased current. The photocurrent component flowing between two electrodes by photoconductive effect is given by 24

\[ I_{ph} = (\Delta \sigma)EWD = (\Delta n)\mu EWD = q(nP_d/\hbar\nu)(\mu E/L), \]

where Δn, E, D, Δm, q, μ, τ, P_d, h, ν, and τ are change in conductivity, electric field, depth of absorption region, change in carrier concentration, unit charge, carrier mobility, quantum efficiency, incident optical power, Planck constant, frequency of incident light, and carrier lifetime, respectively. The photovoltaic component of I_ph is proportional to areal power density of incident light P_d and weakly depends on V_g23,24. In photogating effect, one type of photogenerated carriers (electrons or holes) is trapped in localized states and the other type of carriers flows in the channel unrecombined. As this is equivalent to doping by the other type of carriers, photogating effect accompanies a shift of threshold voltage (V_th)25. As V_th shifts, the drain current changes from I_d to I_d + ΔI_d and it follows that 26

\[ I_{ph} = I_d(V_g - V_th + \Delta V_th) - I_d(V_g - V_th) \approx g_m \Delta V_th = g_m(kT/q)ln(1 + nq\lambda P_d/\hbar c V_th), \]

where g_m, k, T, λ, I_d, and c are transconductance, Boltzmann constant, temperature, wavelength of incident light, dark
current, speed of light, respectively. Thus, the photogating component of $I_{ph}$ shows logarithmic dependence on $P_{in}$ and is roughly proportional to transconductance ($g_m$)\textsuperscript{23,26}.

Figure 2(a) shows $I_{ph}$ and $g_m$ as a function of $V_g$ for the same device in Fig. 1(c). The calculation of $I_{ph}$ ($I_{ph} = I_{light} - I_{dark}$, where $I_{light}$ is $I_g$ in a detector with light), and $g_m$ ($g_m = dI_g/dV_g$) is based on the data in Fig. 1(c) at $P_{in} = 18$ mWcm$^{-2}$. The similarity between $I_{ph}$ and $g_m$ suggests that photogating effect dominates the photoresponse of MoSe$_2$ transistors. In the inset of Fig. 2(a), the change in $V_{th}$ ($\Delta V_{th}$) for electrons and holes is shown as a function of $P_{in}$. The increasing change of $V_{th}$ with increasing $P_{in}$ also suggests the dominant role of photogating effect in our MoSe$_2$ phototransistors. However, the dependence of $I_{ph}$ on $V_g$ in Fig. 2(b) through (d) suggests that each effect dominates $I_{ph}$ at different range of $V_g$. Figure 2(b) through (d) show $I_{ph}$ of our MoSe$_2$ phototransistor at different on-state for electrons (at $V_g = 40$ V), an off-state (at $V_g = 0$ V), and an on-state for holes (at $V_g = -40$ V) as a function of $P_{in}$ in sequence. $I_{ph}$ is calculated based on the data in Fig. 1(c). The $I_{ph}$ in an on-state for electrons (at $V_g = 40$ V) and for holes (at $V_g = -40$ V) shows logarithmic dependence on $P_{in}$ suggesting the dominant role of photogating effect. However, the linear dependence of $I_{ph}$ in an off-state suggests the dominant role of photocductive effect. Such a distinct dependence of $I_{ph}$ on $V_g$ regime was also observed in phototransistors based on MoS$_2$\textsuperscript{10}, MoTe$_2$\textsuperscript{26}, compound semiconductors\textsuperscript{28}, and organic semiconductors\textsuperscript{29}.

The observed dependence of $I_{ph}$ on $V_g$ can be understood by the simplified energy band diagrams of an MoSe$_2$ phototransistor under a bias ($V_g$) at different $V_g$ in Fig. 3. For mechanically exfoliated MoS$_2$ flakes and chemical vapor deposited MoS$_2$ films, the existence of trap state was reported in literature\textsuperscript{13,29,30} as a result of structural defects at the surface and inside MoSe$_2$. Similarly, we assume that electron traps and hole traps exist in the energy bandgap of MoSe$_2$ by structural defects at the surface and inside MoSe$_2$. In Fig. 3(a) (at $V_g = 40$ V), the Fermi level ($E_F$) is located close to the conduction band edge and the majority of electron traps are filled. Without light, $I_{dark}$ flows by the thermionic emission or tunneling of electrons. With light, the photogenerated holes fill hole traps and additional current $I_{ph}$ flows by the uncombined photogenerated electrons. In Fig. 3(b) (at $V_g = 0$ V), $E_F$ moves toward midgap and the majority of electron traps and hole traps become unfilled. Without light, $I_{dark}$ is negligible as the high barrier height at the contact allows negligible injection of electrons and holes. With light, $I_{ph}$ is less than that in Fig. 3(a) as the photogenerated electrons and holes recombine or fill the trap states. In Fig. 3(c) (at $V_g = -40$ V), $E_F$ is close to the valence band edge and the majority of hole traps are filled. Without light, $I_{dark}$ flows by the thermionic emission or tunneling of holes. With light, the photogenerated electrons fill electron traps and additional current $I_{ph}$ flows by the uncombined photogenerated holes.

The performance of an MoSe$_2$ transistor as a photodetector can be evaluated by responsivity (a measure of the electrical response to light) and specific detectivity (a measure of detector sensitivity)\textsuperscript{31}. Responsivity ($R$) is given by $R = (I_{light} - I_{dark})/(P_{in}A)$, where $A$ is the area of the detector. Under the assumption that shot noise from $I_{dark}$ is the major contributor to the total noise, specific detectivity ($D^*$) is given by\textsuperscript{32} $D^* = RA^{1/2}/(2\Delta f_{dark})^{1/2}$. Figure 4(a,b) show the calculated $R$ and $D^*$ of the MoSe$_2$ phototransistor at different $P_{in}$ and $V_g$. Maximum $R$ of $1.4 \times 10^7$ AW$^{-1}$...
and \(D^*\) of \(5.5 \times 10^{13}\) jones are obtained at \(P_m = 27\mu\text{Wcm}^{-2}\) and \(V_f = 40\text{ V}\). These are the highest values of \(R\) and \(D^*\) among MoSe\(_2\) phototransistors reported in literature so far (\(R = 0.01–238\text{ AW}^{-1}\) and \(D^* = 1.0 \times 10^{11}–7.6 \times 10^{11}\) jones at \(P_m = 10–100\text{ mWcm}^{-2}\))\(^{13,15–19}\). As \(R\) and \(D^*\) increase with decreasing \(P_m\), the enhancement of \(R\) and \(D^*\) in this work may be due to the low \(P_m\) compared to that in literature. However, even at comparable \(P_m\) in the range of 18–54\text{ mWcm}^{-2}, the maximum \(R\) and \(D^*\) in this work (\(R = 519\text{ AW}^{-1}\) and \(D^* = 1.3 \times 10^{12}\) jones) are about twice as high as those in literature. In Fig. 4(a,b), the overall dependence of \(R\) and \(D^*\) on \(P_m\) and \(V_g\) is consistent with literature\(^{13}\). \(R\) increases as \(P_m\) decreases or \(V_g\) increases, while \(D^*\) increases as \(P_m\) or \(V_g\) decreases. As \(P_m\) increases, more holes fill shallow trap states where lifetime is short. This results in faster recombination hence \(R\) decreases. When \(P_m\) increases, \(D^*\) also decreases as \(R\) decreases and \(I_{\text{dark}}\) remains unchanged. When \(V_g\) increases, electrical doping at higher \(V_g\) reduces contact resistance resulting in higher photocurrent and \(R\). However, as \(V_g\) increases, \(I_{\text{dark}}\) also increases, which degrades \(D^*\).

It needs to be mentioned that our MoSe\(_2\) transistors show wide device-to-device variation of \(\mu_{\text{FE}}, R,\) and \(D^*\) (Table S1 in Supplementary Information). Such wide device-to-device variation is commonly observed in the transistors based on transition metal dichalcogenides such as MoSe\(_2\), presumably because of the variation of intrinsic defects in crystals\(^3\). While it is very difficult to pinpoint the origin of high performance in the best device, the correlation between \(R\) and \(\mu_{\text{FE}}\) in this work (Fig. S1 in Supplementary Information) suggests that the enhanced optoelectronic properties may be related to the enhanced electrical performance of our MoSe\(_2\) device. It is also supported by the fact that our MoSe\(_2\) device shows the highest mobility among MoSe\(_2\) transistors in Table 1.

We also note the negligible correlation between the optoelectronic properties of MoSe\(_2\) devices and MoSe\(_2\) thickness. This may seem counterintuitive because the width of energy bandgap changes for thin MoS\(_2\) crystals (<~4 nm in thickness)\(^4\) and light absorption depends on MoSe\(_2\) thickness. Yet, because the thickness of our...
MoSe₂ flakes ranges from 20 nm to 80 nm, we expect negligible differences in energy bandgap in our MoSe₂ devices. On the other hand, we expect higher responsivity for devices with thicker MoSe₂ as more light is absorbed in thicker MoSe₂. However, the responsivity shows negligible correlation with thickness of MoSe₂ flakes in this investigation (Fig. S2 in Supplementary Information). This may be due to the variation of intrinsic materials quality overshadowing the effect of thickness. The mobility and detectivity in Fig. S2 also show negligible correlation with the thickness of MoSe₂ flakes, supporting this argument.

To explore the response time of our MoSe₂ phototransistors, we measure the time-resolved photoresponse of our MoSe₂ phototransistors for multiple illumination cycles. Figure 4(c) shows the result for the same device in Fig. 1(c). The nearly identical response for multiple cycles suggests the overall robustness and reproducibility of our MoSe₂ phototransistors. From a zoomed-in region in Fig. 4(d), we obtain rise time of 1.7 ms and fall time of 2.2 ms. (Rise time is calculated as the time taken by current to increase from 10% to 90% of the maximum current. Fall time is calculated as the time taken by current to decrease from 90% to 10% of the maximum current.) This is the fastest response time of MoSe₂ phototransistors ever reported in literature, which ranges from 5 ms to 400 ms. It is intriguing that our MoSe₂ phototransistors exhibit high responsivity and fast response time. Because the long lifetime of carriers in photogating effect suggests slow response to light, the fast response time in our MoSe₂ device may be related to the characteristics of trap states. One possibility is that trap states in our MoSe₂ device may have shorter lifetime and higher density than those in literature.
while the shorter lifetime of trap states could provide fast response, the higher density of trap states could provide higher doping enhancing responsivity. However, we may only speculate at this stage and further investigation is needed on the characteristics of trap states including the distribution of trap energy, trap density, trap lifetime, and carrier capture probability.

Table 1 compares $\mu_{\text{FE}}$, $R$, $D^*$, and response time of MoSe$_2$ phototransistors in literature. Because the measurement conditions, such as $V_{GS}$, $V_{DS}$, $P_{in}$, and excitation energy, can influence the device performance, comparable measurement conditions with those in literature are used in this work. Our MoSe$_2$ phototransistors exhibit the best performance in terms of $\mu_{\text{FE}}$, $R$, $D^*$, and response time, demonstrating the feasibility of achieving high responsivity and fast response time in multilayer MoSe$_2$ phototransistors. Future work combining controlled growth of materials with optimized device architecture and processing will further enhance the performance of MoSe$_2$ phototransistors.

**Conclusions**

We report high-responsivity multilayer MoSe$_2$ phototransistors with fast response time fabricated with mechanically-exfoliated MoSe$_2$ flakes on SiO$_2$/Si substrates. Our MoSe$_2$ phototransistors exhibit asymmetric ambipolar behavior with strong $n$-type characteristic. Without light, high on/off-current ratio of $10^6$ and field-effect mobility of 50.6 cm$^2$/V·s are obtained for electrons. Under 650-nm-laser, our MoSe$_2$ phototransistor exhibits the best performance among MoSe$_2$ phototransistors in literature including high responsivity ($1.4 \times 10^3$ AW$^{-1}$), high specific detectivity ($5.5 \times 10^{11}$ Jones), fast rise time (1.7 ms) and fast fall time (2.2 ms). The dependence of photocurrent on gate voltage and optical power density suggest that photocurrent is dominated by photoinduced charge formation and minority carrier conductivity effect in off-state. These results demonstrate the feasibility of achieving high-performance multilayer MoSe$_2$ phototransistors, providing potentially important implications on using MoSe$_2$ phototransistors for a variety of applications including touch sensor panels, image sensors, solar cells, and communication devices.

**Methods**

**Device fabrication.** Multilayer MoSe$_2$ flakes were obtained by gold-mediated mechanical exfoliation from bulk MoSe$_2$ crystals (2D Semiconductors) and transferred to highly doped p-type Si wafer with thermally grown SiO$_2$ (300 nm). The thickness of MoSe$_2$ flakes measured by atomic force microscope (AFM, Park Systems XE-100) existed between 20 nm and 80 nm. To form source and drain electrodes (100 nm Ti and 50 nm Au) deposited by electron-beam evaporation were patterned using photolithography and etching. The device was then annealed at 200°C in a vacuum tube furnace for 2 hours (100 sccm Ar and 10 sccm H$_2$) to remove resist residue and to decrease contact resistance.

**Device characterization.** Optical absorbance of MoSe$_2$ was measured by UV-visible spectroscopy (Perkin-Elmer Lambda 35). Electrical characterizations were carried out with current-voltage ($I$-$V$) measurements (Agilent 4155 C Semiconductor Parameter Analyzer) at room temperature. The photoresponse of MoSe$_2$ phototransistors was measured with a 650-nm-laser (beam size of 3 mm) at different power densities (0.027, 18 and 54 mW cm$^{-2}$). Dynamic on/off switching was conducted using a function generator (Tektronix AFG310).

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Author Contributions
H.L. and W.C. designed the experiments. H.L. fabricated the devices. H.L., J.A., S.I. and J.K. characterized the devices. H.L. and W.C. wrote the manuscript. All authors reviewed the manuscript.

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