Gate-tunable photodetector and ambipolar transistor implemented using a graphene/MoSe₂ barristor

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Abstract

Next-generation electronic and optoelectronic devices require a high-quality channel layer. Graphene is a good candidate because of its high carrier mobility and unique ambipolar transport characteristics. However, the on/off ratio and photoresponsivity of graphene are typically low. Transition metal dichalcogenides (e.g., MoSe₂) are semiconductors with high photoresponsivity but lower mobility than that of graphene. Here, we propose a graphene/MoSe₂ barristor with a high-k ion-gel gate dielectric. It shows a high on/off ratio (3.3 × 10⁴) and ambipolar behavior that is controlled by an external bias. The barristor exhibits very high external quantum efficiency (EQE, 66.3%) and photoresponsivity (285.0 mA/W). We demonstrate that an electric field applied to the gate electrode substantially modulates the photocurrent of the barristor, resulting in a high gate tuning ratio (1.50 μA/V). Therefore, this barristor shows potential for use as an ambipolar transistor with a high on/off ratio and a gate-tunable photodetector with a high EQE and responsivity.

Introduction

Graphene, a two-dimensional (2D) carbon atomic crystal, has attracted substantial interest for electronic applications owing to its high intrinsic carrier mobility, excellent mechanical flexibility, optical transparency, and unique ambipolar transport characteristics. In particular, its ambipolarity suggests that carriers can be tuned continuously between electrons and holes by supplying the required gate biases, enabling a wide variety of applications, including memory, frequency multipliers, high-frequency oscillators up to the THz range, and fast switches. However, graphene-based transistors have not yet been implemented in real applications because of their low on/off ratio, which arises from graphene’s gapless band structure. A high on/off ratio has been obtained by introducing a barristor, that is, a gated graphene/Si junction, because the high junction resistance can provide a sufficiently low off-state current. However, these barristors usually do not have the typical advantages of graphene, such as mechanical flexibility, optical transparency, and ambipolar transport properties, because Si and a low-k dielectric are used. Recently reported barristor structures, such as graphene/MoS₂ and graphene/WS₂, have shown very low mobility (40–60 cm²/V s) compared to that of graphene.

Graphene photodetectors are being developed. The high mobility of graphene enables high-speed extraction of the photogenerated carriers. However, graphene photodetectors, which typically use the local potential gradient near the graphene–metal junctions, have shown low external quantum efficiency (EQE) and responsivity owing to poor absorption and low built-in potential. To overcome this problem, graphene photodetectors require extensive junctions rather than local junctions, as well as high mobility and a high built-in potential. Unlike graphene-based lateral photodetectors, which have a rather small photosensing active area near the gate...
graphene–metal contact, the vertical barristor device generates a broad region of photocurrent throughout the vertical stack area\textsuperscript{15–17}. In addition, barristor devices based on bonding with a high-absorption material may exhibit high absorption resulting from the high built-in potential. However, the 2D barristor devices have revealed limited optoelectronic performances due to their low carrier mobilities and poor gate tuning ratios\textsuperscript{12,14}.

Here, we propose a field-effect device with a graphene/MoSe\textsubscript{2} channel layer and a high-\textit{k} ion-gel gate dielectric. The device shows a high on/off ratio (3.3 × 10\textsuperscript{4}) and ambipolar behavior that is controlled by an applied gate voltage. Modulation of the Fermi level (\(E_F\)) of graphene by applying a gate voltage (\(V_{SG}\)) is confirmed by the change in the Schottky barrier (SB) height (\(\Phi_b\)) at the graphene/MoSe\textsubscript{2} junction. These field effects, including ambipolar behavior, are locally investigated using scanning photocurrent microscopy (SPCM). It is further demonstrated that an external electric field can be used to modulate the amplitude or even completely reverse the polarity of the photocurrent in the vertical junction of the graphene/MoSe\textsubscript{2} barristor device. The strong gating effect of the device results in a higher EQE (66.3\%), responsivity (285.0 mA/W), and gate tuning ratio (1.50 \(\mu\)A/V) compared to those of pristine devices. Therefore, our graphene/MoSe\textsubscript{2} barristor with an ion-gel gate dielectric is a suitable candidate for use in ambipolar transistors with a high on/off ratio and gate-tunable broad-area photodetectors with a high EQE and responsivity.

**Materials and methods**

Single-layer graphene and few-layer MoSe\textsubscript{2} were fabricated by mechanical exfoliation on a SiO\textsubscript{2} (300 nm)/Si substrate. The SiO\textsubscript{2}/Si substrate was cleaned with hot piranha solution (H\textsubscript{2}SO\textsubscript{4}/H\textsubscript{2}O\textsubscript{2} = 4:1) to remove organic matter from its surface\textsuperscript{18}. The electrodes were fabricated using electron beam lithography (Tescan Mira 2 and Raith Elphy Quantum Plus) and electron beam evaporation (Daeki-Hi-tech DKEB-02-04). Poly(methyl methacrylate) (PMMA) C\textsubscript{4} solution was spin-coated on the layers at 4500 rpm, followed by baking at 180 °C for 2 min. Electron beam lithography at a dose of ~280 \(\mu\)C/cm\textsuperscript{2} was used to define patterns on the spin-coated PMMA layer. Then, the Au (50 nm) source, drain, and gate electrodes were deposited on the graphene, MoSe\textsubscript{2}, and SiO\textsubscript{2}/Si substrate, respectively, by electron beam evaporation at a deposition rate of 0.4 \(\AA\)/s and a pressure of 10\textsuperscript{−6} Torr. Graphene with two source electrodes was transferred to the sample on which MoSe\textsubscript{2}, two drain electrodes, and one gate electrode were deposited. The graphene/MoSe\textsubscript{2} junction was formed between the source and drain electrodes. A polydimethylsiloxane well was located between the side gate and the graphene/MoSe\textsubscript{2} channel and filled with an ion-gel dielectric for the SPCM measurements\textsuperscript{19,20}.

Electrodes and markers were patterned on exfoliated graphene and MoSe\textsubscript{2} layers over SiO\textsubscript{2}/Si substrates using electron beam lithography and electron beam evaporation systems. An ion gel was fabricated by gelation of a triblock copolymer in an ionic liquid\textsuperscript{21}. An ultraviolet (UV) cross-linkable polyelectrolyte ion-gel dielectric layer was deposited on the graphene/MoSe\textsubscript{2} heterostructure as a gate dielectric. The ionic liquid 1-ethyl-3-methylimidazolium bis (trifluoromethylsulfonyl)imide, the monomer poly(ethylene-glycol) diacrylate (M\textsubscript{w} = 575 g/mol), and the UV cross-linking initiator 2-hydroxy-2-methylpropiophenone were mixed at a weight-ratio of 88:8:4, and the mixed solution was dropped and spread on the graphene/MoSe\textsubscript{2} structure. The dropped solution was solidified by UV exposure (365 nm, 100 mW/cm\textsuperscript{2}) for 10 s. Finally, a side-gate graphene/MoSe\textsubscript{2} device with channel dimensions of 10 \(\mu\)m \times 30 \(\mu\)m was obtained.

**Results and discussion**

Figure 1a shows a schematic diagram of the graphene/MoSe\textsubscript{2} barristor structure. High-quality single-layer graphene and few-layer MoSe\textsubscript{2} samples were prepared by mechanical exfoliation on a SiO\textsubscript{2} (300 nm thick)/Si substrate. The Au (50 nm) source, drain, and gate electrodes were deposited on the graphene, MoSe\textsubscript{2}, and SiO\textsubscript{2}/Si substrate, respectively, by electron beam evaporation. The device fabrication is described in detail in the “Materials and methods” section and Supplementary Information (Fig. S1).

Figure 1b shows an optical microscope image of a graphene/MoSe\textsubscript{2} barristor device. The blue and green areas surrounded by white dashed lines are graphene and MoSe\textsubscript{2}, respectively. Two source and drain electrodes were used to check the electrical characteristics of graphene and MoSe\textsubscript{2}, respectively. The electrical characteristics of the graphene/MoSe\textsubscript{2} barristor device were checked between one source and one drain electrode. The SPCM setup, which includes a transport measurement system, is illustrated in Fig. 1c and the Supplementary Information (Fig. S2). The graphene/MoSe\textsubscript{2} barristor device was illuminated by a diffraction-limited laser (spot diameter, ~500 nm; wavelength, 532 nm) while the device conductance was recorded as a function of the laser spot position. The small spot size (Supplementary Information, Fig. S3) enabled us to record the photoinduced electronic signal originating from light illumination on a specific part of the graphene/MoSe\textsubscript{2} barristor. Because the photocurrent is influenced by the potential profile, an SPCM image can provide information on the local potential profile. We simultaneously obtained a reflected light image to determine the position of the laser spot. Figure 1d shows a photocurrent image of the graphene/MoSe\textsubscript{2} barristor obtained at \(V_{SG} = −1.0\) V. The yellow areas denote the source and drain electrodes. The color scale
refers to the photocurrent measured between the source and drain electrodes at zero bias. The red area near the graphene/MoSe$_2$ junction corresponds to photocurrent flowing to the drain. The blue area near the MoSe$_2$/drain electrode junction corresponds to photocurrent flowing to the source. The generated photocurrent signals are attributed to band bending and the resultant local electric field at the graphene/MoSe$_2$ and MoSe$_2$/drain electrode junctions. The opposite photocurrent directions indicate the opposite directions of the local electric fields at the graphene/MoSe$_2$ and MoSe$_2$/drain electrode junctions.

Figure 2a shows the source–drain current amplitude ($|I_{SD}|$) vs. the source–drain voltage ($V_{bias}$) measured as $V_{SG}$ increases from $-1.25$ to $0.5$ V (right panel) and from $0.5$ to $1.25$ V (left panel) in $0.25$ V steps. The orange and blue arrows indicate directions of changes in $|I_{SD}|$ with increasing $V_{SG}$ in the $p$-type and $n$-type regions, respectively. The right panel shows typical $p$-type behavior, in which the current increases as $V_{SG}$ increases from $0.5$ to $1.25$ V. We can confirm the ambipolar behavior ($p$-type and $n$-type characteristics) for $-1.25 < V_{SG} < 1.25$ V. Note that we can control the type of graphene/MoSe$_2$ barristor device by changing the applied $V_{SG}$, as in an ambipolar graphene channel device.

Figure 2b shows the transfer characteristics of the graphene/MoSe$_2$ barristor device, which were measured as a function of $V_{SG}$ at a fixed $V_{bias}$ of $10$ mV. These characteristics are different from those of graphene and MoSe$_2$ devices (Supplementary Information, Figs. S4 and S5, respectively). Our device shows a high on/off ratio of $3.3 \times 10^4$, which substantially exceeds those reported for pure graphene transistors ($2$–$20$). The barristor exhibits distinct ambipolar behavior and remarkably high carrier mobility values, specifically, an electron mobility of $247$ cm$^2$/V s and a hole mobility of $182$ cm$^2$/V s at room temperature. These values are determined using the following expression for the field-effect mobility: \[ \mu = \frac{1}{C_{SG}} \times \frac{d\sigma}{dV_{SG}} \], where $C_{SG}$ is the side-gate dielectric
The conductivity is defined as \( \sigma = \frac{I_{SD}}{V_{bias} \times L/W} \), where \( L \) is the length and \( W \) is the width of the barristor channel. Because this device does not have well-defined channel dimension, the mobility was calculated from the minimum value of \( L \) and the maximum value of \( W \) for identifying the lower bound of the mobility. The obtained mobility values have similar orders of magnitude to those reported for other barristor devices (40–60 cm²/V s), such as graphene/MoS₂ and graphene/WS₂², and for MoSe₂ devices (50–160 cm²/V s).²²–²⁵

The observed ambipolar behavior controlled by \( V_{SG} \) can be explained by using the schematic band diagrams of the graphene/MoSe₂ barristor at \( V_{SG} < 0 \) V, \( V_{SG} > 0 \) V, and \( V_{SG} \gg 0 \) V (Fig. 2c–e, respectively). \( \Phi_{B1} \) and \( \Phi_{B2} \) indicate the barrier heights for electrons and holes, respectively, at the graphene/MoSe₂ interface. Because of the observed p-type behavior at \( V_{SG} < 0 \) V, we can assume that \( \Phi_{B1} > \Phi_{B2} \) at \( V_{SG} < 0 \) V (Fig. 2c). If the \( E_F \) of graphene is near the valence band of MoSe₂, holes become the majority carriers, so a p-type SB forms at the graphene/MoSe₂ junction. As the gate voltage increases, \( \Phi_{SG} \) also increases, and hole transport becomes more difficult. Conversely, as the gate voltage decreases, \( \Phi_{B2} \) also decreases, and hole transport becomes easier. The \( E_F \) of graphene can be increased by the applied positive \( V_{SG} \) so that \( \Phi_{B1} = \Phi_{B2} \) and even \( \Phi_{B1} < \Phi_{B2} \), as shown in Fig. 2d, e, respectively. When \( \Phi_{B1} < \Phi_{B2} \) at \( V_{SG} \gg 0 \) V, electrons become the majority carriers, and the graphene/MoSe₂ barristor exhibits n-type behavior, as shown in Fig. 2e. As the gate voltage increases, \( \Phi_{B1} \) decreases, and electron transport becomes easier. Conversely, as the gate voltage decreases, \( \Phi_{B1} \) increases, and electron transport becomes more difficult. The substantial change in the \( E_F \) of graphene is attributed to the small area of the Fermi surface and the introduction of the ion-gel gate dielectric. In addition, because MoSe₂ has a small band gap and similarly high-mobility values (50–160 cm²/V s) for electrons and holes, the relative sizes of \( \Phi_{B1} \) and \( \Phi_{B2} \) can be controlled by modulating the \( E_F \) of graphene.

To investigate the local field effect, we performed SPCM measurements of the graphene/MoSe₂ barristor shown in Fig. 3a at various gate voltages. Figure 3b–d show photocurrent images of the barristor at various gate voltages between −1.0 and 1.0 V. Because of the side-gate structure and transparent ion-gel dielectric, the illumination light reaches the barristor device without substantial loss. The blue dashed line, red dashed line, and yellow areas
denote graphene, MoSe2, and the electrodes, respectively. Unlike a graphene-based lateral photodetector, which has a rather small photosensing active area near the graphene–metal contact, our vertical barristor device clearly shows a broad area of photocurrent generation throughout the junction of the vertical graphene/MoSe2 stack. When a $V_{SG}$ of $-1.0$ V is applied from the side gate (Fig. 3b), the SPCM images show a red area corresponding to photocurrent flowing to the drain electrode at the graphene/MoSe2 and a blue area corresponding to photocurrent flowing to the source electrode at the MoSe2/drain electrode junction. Under a $V_{SG}$ of 0.25 V (Fig. 3c), the photocurrent contrast signal at the graphene/MoSe2 and MoSe2/drain electrode junctions disappears. As $V_{SG}$ increases to 1.0 V, the photocurrent contrast at the MoSe2/drain electrode junction changes to red, whereas blue and red photocurrents are presented on the graphene/MoSe2 heterojunction (Fig. 3d). It seems that the blue and red photocurrents on the graphene/MoSe2 heterojunction may be caused by the spatial inhomogeneity of the graphene/MoSe2 device. Because we used the side gate configuration and a liquid ion gel, the field effect on the graphene/MoSe2 junction may depend on the location of the 2D planar device. The changes in photocurrent direction at both junctions demonstrate manipulation of the local potential profile and inversion of the band bending at each junction, which are obtained by varying $V_{SG}$ in this vertically stacked device.

Importantly, because of the finite density of states in graphene and the weak electrostatic screening effect, the graphene/MoSe2 SB height can be effectively modulated by applying an external field through the side-gate electrode$^{26-28}$. Figure 3e–g show schematic band diagrams of the graphene/MoSe2 and MoSe2/drain electrode junctions at various $V_{SG}$ values. Modulation of the $E_F$ of graphene and MoSe2 can directly affect the band bending and photocurrent characteristics at the junctions. Because of the quantum capacitance and partial electrostatic transparency of graphene, the applied $V_{SG}$ not only modulates the $E_F$ of graphene but also penetrates graphene to accumulate/invert space charges within MoSe2$^{28}$. In graphene/MoSe2 barristor devices, the change in $E_F$ of graphene is very important. Therefore, we calculated $\Delta E_F$ using $\Delta E_F = h\nu_T \sqrt{n}$. Here, $h$ is the Dirac constant, $\nu_T$ is the Fermi velocity of graphene ($1.0 \times 10^6$ m/s), and $n$ is the carrier density$^{11,30}$. The hole carrier density at $V_{SG} = -1.50$ V and electron carrier density at $V_{SG} = 1.45$ V of our graphene/MoSe2 barristor device are determined to be $1.03 \times 10^{13}$ and $4.88 \times 10^{12}$ cm$^{-2}$, respectively. The calculated $\Delta E_F$ is $-0.37$ eV at $V_{SG} = -1.50$ V and 0.26 eV at $V_{SG} = 1.45$ V. The amount of change in $E_F$ of 0.63 eV at the graphene/MoSe2 barristor device obtained when $V_{SG}$ varies between $-1.50$ and $1.45$ V is larger than that of other barristor devices$^{11,30}$. We define $V_F$ as the applied $V_{SG}$ at which $\Phi_{B1} = \Phi_{B2}$, which results in a flat band in
MoSe$_2$. When the $E_F$ value of graphene is reduced at $V_{SG} < V_F$, holes become the majority carriers, resulting in a $p$-type SB with downward band bending at the graphene/MoSe$_2$ junction. Then, the photoexcited electrons near the graphene/MoSe$_2$ junction drift toward the graphene to produce a positive photocurrent to the drain electrode (Fig. 3e). Similarly, $p$-type band bending causes photoexcited electrons near the MoSe$_2$/drain electrode to drift toward the drain electrode, resulting in a negative photocurrent to the source electrode. When the $E_F$ values of graphene and MoSe$_2$ are increased at $V_{SG} = V_F$, the band bending of MoSe$_2$ at the interfaces becomes negligible, resulting in easy recombination of photoexcited electrons and holes before the photocurrent forms (Fig. 3f). When the $E_F$ value of graphene is higher at $V_{SG} > V_F$, electrons become the majority carriers, resulting in an $n$-type SB with upward band bending at the graphene/MoSe$_2$ junction. Then, the photoexcited holes near the graphene/MoSe$_2$ junction drift toward the graphene to produce a negative photocurrent to the source electrode (Fig. 3g). Similarly, $n$-type band bending causes the photoexcited holes near the MoSe$_2$/drain electrode to drift toward the drain electrode, resulting in a positive photocurrent to the drain electrode. Various graphene/MoSe$_2$ devices showed similar photocurrent images obtained at $V_{SG} = 0$ V (Supplementary Information, Fig. S7).

Figure 4a shows the gate dependence of the local photocurrent measured along the black line in the inset SPCM image in Fig. 4a as $V_{SG}$ is swept from $-1.0$ to $1.0$ V in 0.01 V steps. We measured the photocurrent by applying very low laser power (0.407 $\mu$W/cm$^2$) without a bias voltage. At $V_{SG} < V_F$, positive (red contrast) and negative (blue contrast) photocurrents are generated at the graphene/MoSe$_2$ (1) and MoSe$_2$/drain electrode (2) junctions, respectively. This photocurrent polarity is a result of $p$-type band bending, in which the $E_F$ of graphene becomes closer to the valence band of MoSe$_2$, as shown in the band diagram in Fig. 3e. This result implies that the photogenerated electrons at the MoSe$_2$/drain electrode and graphene/MoSe$_2$ junctions are collected by the drain and source electrodes, respectively, as reported in previous studies of semiconducting devices with $n$-type bending$^{34}$. The graphene/MoSe$_2$ barristor device shows ambipolar behavior with $p$- and $n$-type characteristics at $V_{SG} < V_F$ and $V_{SG} > V_F$, respectively. Because the intensity of the photocurrent was roughly proportional to the local electric field or the slope of the electrostatic potential, we qualitatively examined the electrostatic potential along the device by integrating the photocurrent line profile at $V_{SG}$ values ranging from $-1.0$ to $1.0$ V in 0.25 V steps.

Figure 4b presents the resulting electrostatic potential profile measured along the black line in the inset SPCM image in Fig. 4a. The measured photocurrent signal $[I_{ph}(x)]$ (Fig. 4b, inset) is proportional to the potential gradient as follows$^{34}$:

$$I_{ph}(x) \propto -\frac{d\varphi(x)}{dx}$$

where $x$ denotes the position along the device channel, and $\varphi(x)$ is the electrostatic potential. The potential changes dramatically near the graphene/MoSe$_2$ (1) and MoSe$_2$/drain electrode (2) contact regions, indicating SB formation at the graphene/MoSe$_2$ junction (1) and MoSe$_2$/drain electrode (2). Note that the change in potential at the graphene/MoSe$_2$ junction with changing $V_{SG}$ is 13-fold larger than that at the MoSe$_2$/drain electrode. This finding implies that the graphene/MoSe$_2$ junction can become a potential element for tunable electronic or optoelectronic devices.

Figure 4c, d show photocurrent vs. $V_{SG}$ data obtained at the graphene/MoSe$_2$ and MoSe$_2$/drain electrode junctions, respectively. At the graphene/MoSe$_2$ junction, the positive photocurrent decreases as $V_{SG}$ increases from $-1.0$ to 0.4 V, and the photocurrent becomes negative at $V_{SG} = 0.4$ V. When $V_{SG}$ is $-0.98$ V, we obtain a very high gate tuning ratio of $\sim 76.6$ nA/V, which is defined as the ratio of the change in photocurrent amplitude to the change in $V_{SG}$. At the MoSe$_2$/drain electrode junction, negative and positive photocurrents are generated at $-1.0$ V < $V_{SG}$ < 0.15 V and 0.15 V < $V_{SG}$ < 1.0 V, respectively. When $V_{SG}$ is $-0.71$ V, we obtain a high gate tuning ratio of $\sim 18.2$ nA/V. We could obtain a higher gate tuning ratio of 1.50 $\mu$A/V at a graphene/MoSe$_2$ junction in another barristor device (Supplementary Information, Fig. S8). These gate tuning ratios, which were measured at a very low laser power (0.407 $\mu$W/cm$^2$), are much higher than those of other graphene-based devices (1–5 nA/V)$^{25}$. In addition, the very high gating effect of our graphene/MoSe$_2$ barristor structure with a high-k ion-gel gate dielectric dramatically changes the polarity of the photocurrent. The high gate-tunability of the photocurrent is significant for the facile modulation of sensitivity for optical sensors. In dark spaces (such as a tunnel and cinema) where the physical
environment is limited, photosensitivity should be amplified so that sufficient photocurrent can be secured. In bright spaces (such as outdoors on a sunny day), photosensitivity can be reduced and the value of photocurrent flowing through the device can be constantly adjusted. Therefore, the development of high-gate tuning elements in optical devices is very necessary.

Using the photocurrent response and input laser power, we can determine the EQE and photoresponsivity. The photoresponsivity ($R_I$) is defined as the ratio of the electrical current response to the incident optical power ($R_I = I_{ph}/P_{opt}$). The EQE ($\eta$) is defined as the number of carriers produced per photon and is expressed as

$$\eta = \frac{I_{ph}}{q\Phi} = \frac{I_{ph}}{q} \left( \frac{h\nu}{P_{opt}} \right)$$

where $I_{ph}$ is the photocurrent, $\Phi$ is the photon flux ($=P_{opt}/h\nu$), $h$ is the Planck constant, $\nu$ is the frequency of the light, $q$ is the electron charge, and $P_{opt}$ is the optical power. At the graphene/MoSe$_2$ junction, the EQE increases from 3.3% (corresponding to a photoresponsivity of 14.2 mA/W) at $V_{SG} = 0$ V to 24.0% (corresponding to a photoresponsivity of 103.2 mA/W) at $V_{SG} = -0.98$ V under excitation at a laser power of 0.407 µW/cm$^2$. At the MoSe$_2$/drain electrode junction, the EQE increased from 0.5% (corresponding to a photoresponsivity of 2.2 mA/W) at $V_{SG} = 0$ V to 3.18% (corresponding to a photoresponsivity of 16.8 mA/W) at $V_{SG} = -0.71$ V. The EQE at the graphene/MoSe$_2$ junction is ~6.6-fold larger than that at the MoSe$_2$/drain electrode junction. The EQE (24%) observed at the vertical graphene/MoSe$_2$ junction at $V_{SG} = -0.98$ V is two orders of magnitude higher than those of lateral metal–graphene–metal
photodetectors (EQE = 0.1–0.2%)\textsuperscript{14,38,39}. We could obtain a higher EQE of 66.3% and photoresponsivity of 285.0 mA/W at a graphene/MoSe\textsubscript{2} junction in another barristor device (Supplementary Information, Fig. S8). This high EQE is due primarily to more efficient photon absorption in the broad area of the vertical barristor device and more efficient charge separation resulting from a much larger band offset and a much higher mobility\textsuperscript{16}.

**Conclusion**

In summary, we fabricated a graphene/MoSe\textsubscript{2} barristor with a high-k ion-gel gate dielectric. The graphene/MoSe\textsubscript{2} device showed a high on/off ratio (3.3 × 10\textsuperscript{6}) and ambipolar behavior that was controlled by an applied gate voltage. SPCM measurements revealed that the graphene/MoSe\textsubscript{2} barristor had a very high EQE (66.3%) and photoresponsivity (285.0 mA/W) values, making it suitable for highly efficient photocurrent generation and photodetection. We further demonstrated that an electric field applied to the gate electrode could substantially modulate the amplitude and polarity of the photocurrent at the graphene/MoSe\textsubscript{2} junction, resulting in a high gate tuning ratio (1.50 μA/V). Therefore, the graphene/MoSe\textsubscript{2} barristor with a high-k ion-gel gate dielectric is a suitable candidate for use in ambipolar transistors (with a high on/off ratio) and gate-tunable photodetectors (with a high EQE and responsivity).

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**Author contributions**

G.O. and B.H.P. planned the projects and designed the experiments. G.O. fabricated and characterized graphene/MoSe\textsubscript{2} barristors. Y.C.K., Y.H.A. and G.O. performed the SPCM experiment. G.O., J.J., and B.H.P. interpreted the results. All the authors participated in discussions and writing the manuscript.

**Conflict of interest**

The authors declare that they have no conflict of interest.

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**Supplementary Information**

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