Research article

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Highly responsive near-infrared photodetector with low dark current using graphene/germanium Schottky junction with Al₂O₃ interfacial layer

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Abstract: The performance of a graphene/Ge Schottky junction near-infrared photodetector is significantly enhanced by inserting a thin Al₂O₃ interfacial layer between graphene and Ge. Dark current is reduced by two orders of magnitudes, and the specific detectivity is improved to $1.9 \times 10^{10} \text{ cm} \cdot \text{Hz}^{1/2} \text{W}^{-1}$. The responsivity is improved to 1.2 AW^{-1} with an interfacial layer from 0.5 AW^{-1} of the reference devices. The normalized photo-to-dark current ratio is improved to $4.3 \times 10^7 \text{ W}^{-1}$ at a wavelength of 1550 nm, which is 10–100 times higher than those of other Ge photodetectors.

Keywords: graphene; graphene/germanium heterostructure; interfacial oxide layer; photodetector; Schottky junction.

1 Introduction

Infrared (IR) photodetectors are being used in diverse applications, including optical communications, image

sensing, thermal detector, and distance sensors [1–3]. For high-performance IR photodetector applications, a variety of IR photodetectors have been developed using narrow bandgap semiconductors to absorb long-wavelength lights, such as ternary semiconductors (HgCdTe, CdZnTe) and compound semiconductors (InSb, GaAs) [4–8]. In general, process cost and complexity for these devices are less competitive compared to the photodetectors used for the visible wavelength range. Graphene, which has excellent electrical conductivity and optical transparency, is a semi-metal with a controllable work function [9–11]. Various graphene/semiconductor Schottky junction devices have been investigated to combine these unique properties [12–17]. For IR detection, the graphene/Ge Schottky junction photodetector is gaining attention owing to its simple structure, excellent performance, and direct compatibility with high-speed integrated circuits.

Zeng et al. [18] demonstrated a monolayer graphene/Ge Schottky junction IR photodetector. The responsivity of this device approaches 51.8 mAW^{-1} at a zero bias. Chang et al. [19] proposed a gate-modulated graphene/Ge Schottky junction photodetector and reported a responsivity of 750 mAW^{-1} . However, the dark current of the graphene/Ge photodetectors is worse than that of commercial IR photodetectors, which is in the order of nA. As the high dark current degrades the detectivity and increases the power consumption of large-scale optoelectronic systems, it should be suppressed further, preferably to less than $1 \mu\text{A}$ per device.

In the case of a Ge-based photodetector, the origin of leakage current has been attributed to threading dislocations in Ge [20, 21]. Moreover, the quality of the graphene/Ge interface has a strong impact on the dark current of heterojunction photodetectors. The dangling bonds at the Ge surface behave as charge carrier recombination sites and degrade the overall photodetector performance. Even though various studies have been performed to solve this problem [19, 22, 23], the best case dark current of Ge-based photodetectors is extremely high for commercial applications.

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Alternatively, several approaches to modulate the Schottky barrier height in a graphene/Ge heterojunction have been tried as an indirect method to suppress the dark current, including graphene doping and surface texturing [22, 24]. Yet, these approaches have limitations in terms of process stability and versatility.

In this work, a thin Al_2O_3 interfacial layer is introduced between graphene and Ge to suppress the dark current by increasing the tunneling distance for low energy carriers and reducing the Fermi level pinning effect at the interface. After the introduction of the Al_2O_3 interfacial layer, a normalized photo-to-dark current ratio ($NPDR$) of $4.3 \times 10^7 \text{ W}^{-1}$ and specific detectivity of up to $1.8 \times 10^{10} \text{ cm} \cdot \text{Hz}^{1/2} \text{ W}^{-1}$ were attained at a wavelength of 1550 nm at room temperature. In addition, the responsivity of graphene/Ge heterojunction photodetector has been improved to 1.2 AW^{-1} , which is 1.6–23 times higher than the previously reported responsivity of a graphene/Ge photodetector [18, 19].

2 Experimental methods

Figure 1A shows the schematic of the fabricated graphene/Ge heterojunction device having a thin Al_2O_3 interfacial layer between graphene and Ge. For device fabrication, a 100 nm thick SiO_2 layer was deposited on an n-type Ge substrate (the resistivity of 2.5–2.7 $\Omega \text{ cm}$), using the plasma-enhanced chemical vapor deposition (PECVD) process. Because the native oxide of Ge could affect the interface quality, DI water treatment was employed at room temperature for 24 h to remove the native oxide before depositing 100 nm SiO_2 on Ge substrate [25, 26]. Subsequently, $16 \times 16 \mu\text{m}^2$ oxide windows were patterned to expose the Ge surface using photolithography and buffered oxide etch (BOE). Immediately afterward, a thin Al_2O_3 layer with a different

thickness in the range of 0–3 nm was deposited as an interfacial layer using the atomic layer deposition (ALD) process (Lucida D100, NCD tech). The growth rate measured with spectroscopic ellipsometry was approximately 1 $\text{\AA}/\text{cycle}$. The monolayer graphene sheet, grown on a copper foil using the thermal chemical vapor deposition (TCVD) method, was wet-transferred onto the Al_2O_3 layer using a polymethylmethacrylate (PMMA) sacrificial layer. Thus, the graphene/Ge junction was formed with minimal native oxide. The optical microscope image of the fabricated device is shown in Figure 1B. Atomic force microscopy (AFM) measurements were conducted to determine the surface roughness of the deposited Al_2O_3 films. As shown in Figure 1C, the average roughness of the bare germanium substrate was 0.83 nm, while the roughness for the thin Al_2O_3 layers was less than 0.4 nm and appeared to be smooth without any observable defects.

After the transfer and cleaning process, graphene channel patterns were formed using a 30 nm Au hard mask and O_2 plasma etch. The Raman spectra of graphene transferred on SiO_2 , thin Al_2O_3 , and Ge are shown in Figure 1D. The presence of a high-quality graphene film is established by the presence of a 2D band peak at 2680 cm^{-1} , a G band peak at 1580 cm^{-1} , and a D band peak at 1350 cm^{-1} . Further, the intensity ratio of the 2D to G band (I_{2D}/I_G) is more than two, which confirms the graphene used in this work is monolayer.

Following this, a 100 nm Au source electrode was formed on the graphene channel using the e-beam evaporator and the photolithography process. For the drain, a 45 nm AgSb alloy (Ag with 1% Sb) was formed on the Ge region using a thermal evaporator and the lift-off process, followed by the rapid thermal annealing process for 5 min in a N_2 atmosphere at 450°C . After the AgSb drain contact formation, the contact resistance was measured using a circular transmission line method (CTLM), and a specific contact resistivity of $4.7 \times 10^{-7} \Omega \cdot \text{cm}^2$ was obtained (Supporting information S1) [27]. This is a relatively low contact resistance for moderately doped n-Ge, considering that highly doped n-Ge ($N_D > 1 \times 10^{19} \text{ cm}^{-3}$) was used to obtain a low specific contact resistivity of $1.68 \times 10^{-7} \Omega \cdot \text{cm}^2$ using a NiGe Ohmic contact [28]. Finally, a passivation anneal was performed at 300°C for 1 h in a vacuum chamber to eliminate residual water

Figure 1: (A) Schematic diagram of graphene/n-type Ge Schottky junction photodetector with a thin Al_2O_3 interfacial layer. (B) Optical image of the fabricated graphene/Ge Schottky junction photodetector. (C) Surface roughness vs. thickness of Al_2O_3 layer grown on Ge surface. (D) Raman spectrum of monolayer graphene on SiO_2 , thin Al_2O_3 , and the Ge substrate.

molecules from the interface of graphene and Ge, and to densify the Al_2O_3 layer.

The current–voltage characteristics of the photodetector were measured using a semiconductor parameter analyzer (Keithley 4200). All the characterizations were performed at room temperature. The photoresponse characteristics were measured using solid-state laser diodes at wavelengths of 520, 660, 850, 1310, 1550, and 1625 nm. The incident power was monitored using the Newport Model 1918-C hand-held optical power meter and 919P-003-10 thermopile sensor. Further details of the experimental setup used to characterize photoresponsivity are provided in Figure S2 in Supporting Information.

3 Results and discussion

Figures 2A shows the dark current of graphene/Ge Schottky junction photodetectors with different interfacial Al_2O_3 thicknesses. Both graphene/Ge photodetectors with and without the Al_2O_3 interfacial layer show typical characteristics of rectifying photodiode. The rectification ratios at ± 2 V are higher than three orders of magnitude, except for a photodetector with a 3 nm Al_2O_3 interfacial layer having a rectification ratio of 36. As the thickness of the Al_2O_3 interfacial layer increases, the dark current is effectively reduced by two orders of magnitude from 1.7×10^{-6} A to 2.7×10^{-8} A at a 2 V bias.

According to the thermionic emission theory [29], the diode current flowing through an oxide can be expressed as

$$I = AA^* T^2 \exp(-\sqrt{\chi_{\text{ox}}} \delta) \exp\left(-\frac{\Phi_{\text{B}}}{kT}\right) \left[\exp\left(\frac{qV}{kT}\right) - 1 \right] \quad (1)$$

where A is the area of the active region, A^* is the effective Richardson constant, T is the absolute temperature, Φ_{B} is the Schottky barrier height, k is the Boltzmann constant, and q is electronic charge. The transmission coefficient across the interfacial oxide layer can be described by the prefactor $\exp(-\sqrt{\chi_{\text{ox}}} \delta)$, where χ_{ox} is the mean barrier height of the oxide layer, and δ is the thickness of the oxide layer. Thus, the drastic decrease in the dark current can be explained by the presence of the ultrathin interfacial oxide layer.

Figure 2B illustrates the photocurrent curves of photodetectors comprising different Al_2O_3 interfacial layers measured at a wavelength of 1550 nm with a fixed intensity of 8.2 μW . Without the Al_2O_3 interfacial layer, the photocurrent increases rapidly at near zero-bias and saturates to 5.8 μA at 2 V. This photovoltaic behavior is similar to that of a typical p – n junction or metal–semiconductor Schottky photodetectors, where the diffusion of minority carrier is limited. With a 1 nm Al_2O_3 interfacial layer, the photocurrent increased to 9.8 μA at a 2 V bias, which is an increase of

approximately 69%. The mechanism of this increase is explained below. As the thickness of Al_2O_3 increased further (more than 2 nm), the photocurrent decreased rapidly due to the increased tunneling distance.

To further investigate the effect of the Al_2O_3 interfacial layer on the quality of the interface and the performance of the graphene/Ge heterojunction, a quantitative analysis of open-circuit voltage was performed. Figure 2C shows the open-circuit voltage (V_{oc}) and excess carrier density (Δn) calculated from V_{oc} of graphene/Ge Schottky junction photodetectors with different Al_2O_3 thicknesses. Equation (2) describes the relationship between the open-circuit voltage V_{oc} , excess carrier density Δn , and doping density N [30].

$$\Delta n(\Delta n + N) = n_i^2 \exp\left(\frac{qV_{\text{oc}}}{kT}\right) \xrightarrow{\Delta n \ll N} \Delta n = \frac{n_i^2}{N} \exp\left(\frac{qV_{\text{oc}}}{kT}\right) \quad (2)$$

where n_i is the intrinsic carrier concentration and k is the Boltzmann constant. As the temperature T and doping density N are accurately known, excess carrier density Δn is correlated with the open-circuit voltage V_{oc} . Thus, under the low injection conditions ($\Delta n \ll N$), the experimentally measured V_{oc} can be used to extract Δn . The Δn tends to increase up to 1.5 nm Al_2O_3 case and then rapidly decreases with increasing Al_2O_3 thickness. In general, a high density of recombination sources close to the junction promotes the influx of carriers and reduces V_{oc} by the recombination at the interface. An increase in excess carrier density with thin Al_2O_3 interfacial layers indicates that Al_2O_3 can effectively passivate the Ge surface with the interfacial layer. This result reveals that the Al_2O_3 interfacial layer plays an important role in improving the interface of graphene and Ge while working as a tunneling barrier for dark current.

Figure 2D shows the normalized photo-to-dark current ratio ($NPDR$) defined as

$$NPDR = \frac{I_{\text{photo}}/I_{\text{dark}}}{P_{\text{in}}} \quad (3)$$

where I_{photo} and I_{dark} are the photocurrent and dark current, respectively, and P_{in} is the incident optical power. The $NPDR$ of the graphene/Ge photodetector is measured at the bias voltage of 0.5, 1, and 2 V while varying the thickness of the Al_2O_3 interfacial layers. Under a 2 V bias condition, the $NPDR$ values are significantly improved at the optimal Al_2O_3 thickness of approximately 2 nm owing to the effective suppression of dark current and the increase in responsivity. The $NPDR$ of graphene/Ge photodetector with a 2 nm Al_2O_3 interfacial layer is $4.3 \times 10^7 \text{ W}^{-1}$ at a 2 V bias under the illumination intensity of 8.2 μW . This result is about two orders of magnitude higher than that of the graphene/Ge without the Al_2O_3 interfacial layer. Furthermore,

Figure 2: Current–voltage characteristics of graphene/Ge Schottky junction photodetectors with varying Al_2O_3 interfacial layers (A) in the dark, (B) under illumination, and (C) Excess carrier density calculated from an open-circuit voltage. (D) Normalized photo-to-dark current ratio (NPDR). (E) Responsivity and (F) detectivity measured at a wavelength of 1550 nm with a fixed intensity of 8.2 μW .

this value is higher than those of other Ge photodiodes and similar to the best result reported previously [31–34].

To quantitatively assess the performance of our photodetector, the responsivity and detectivity are calculated using the following equations:

$$R = \frac{I_{\text{ph}}}{P_{\text{in}}}, \quad D = R \sqrt{\frac{A}{2qI_d}} \quad (4)$$

where I_{ph} is the photocurrent, P_{in} is the power of the incident light, I_d is the dark current, and A is the area of the device. It is clear from Figures 2E–F that 2 nm of Al_2O_3

interfacial layer is the optimal thickness showing the highest responsivity and detectivity of approximately 1.2 AW^{-1} and $1.8 \times 10^{10} \text{ cm} \cdot \text{Hz}^{1/2} \text{W}^{-1}$, respectively, at a wavelength of 1550 nm. The detectivity of the graphene/ Al_2O_3 /Ge photodetector is comparable with those of other commercial IR photodetectors [35, 36].

The schematic energy band diagrams of the graphene/Ge heterojunction shown in Figure 3 describe the changes in the photocurrent and dark current conduction mechanism modulated by the Al_2O_3 interfacial layer. As shown in Figure 3A, without an interfacial layer between graphene

Figure 3: The energy band diagram of a graphene/Ge Schottky junction under illumination in reverse biased condition.

(A) Without an Al_2O_3 between the graphene and germanium, many electrons and holes can move into the graphene, where they recombine. (B) With a thin Al_2O_3 layer of approximately 20 Å, electrons are blocked, but holes are transmitted. (C) With a thick Al_2O_3 (>20 Å) layer, both electrons and holes are blocked, and eventually recombine. Experimentally measured I - V curves for each case are shown together both in the dark and under illuminated conditions. Here, Φ_B , E_C , E_V , $E_{F,\text{Ge}}$, and $E_{F,g}$ represent the Schottky barrier height, conduction band edge, valence band edge, Fermi level of Ge, and Fermi level of graphene, respectively. The arrows indicate the direction and relative magnitudes of current depending on the Al_2O_3 interfacial layer.

and Ge, the photo-generated holes diffuse into graphene, and electrons are diffused into the Ge substrate. As the holes diffused into the graphene are immediately recombined, an additional hole flux from the substrate is necessary to maintain the charge neutrality in the depletion layer of Ge [17]. When there is no illumination, the Schottky barrier blocks the electron transfer from the graphene, which is a dominant source of dark current. In the case shown in Figure 3B, the thin interfacial layer between graphene and Ge results in an asymmetric tunneling barrier that blocks the electron transfer contributing to the dark current while allowing the hole transport via direct tunneling mechanism. Thus, when the thickness of the Al_2O_3 interfacial layer is moderate, the dark current

reduction can be maximized with a minimal impact on the photocurrent. However, when the Al_2O_3 interfacial layer becomes too thick as in the case shown in Figure 3C, both electron and hole transport are retarded by the interfacial layer, resulting in the degraded performance of graphene/ Al_2O_3 /Ge photodetector.

Figure 4A shows the photocurrent in the graphene/Ge photodetector with a 2 nm Al_2O_3 interfacial layer as a function of the light intensity in the range of 8.2–82.5 μW . It was observed that the photocurrent quickly rises from 5.6 to 101 μA . Moreover, because of the negligible recombination loss, the photocurrent is linearly dependent on the light intensity. The real-time photocurrent response of the graphene/Ge photodetector at a 2 V bias

Figure 4: (A) Incident light intensity-dependent photocurrent and (B) time-resolved photocurrent of the graphene/Ge photodetector with a 2 nm Al_2O_3 interfacial layer at a wavelength of 1550 nm. Wavelength-dependent (C) responsivity and (D) detectivity of the photodetector measured at a fixed light intensity of 25 μW .

under the illumination of 1550 nm is shown in Figure 4B. While the incident light is switched with 5 s period, consistent and repeatable photocurrent responses are observed without noticeable degradation. Figures 4C–D shows the responsivity and detectivity as a function of the incident light wavelength in the range of 520–1625 nm at a constant intensity of 25 μW . The photodetector exhibits peak sensitivity at 1310 nm, which corresponds to the intrinsic optical absorption wavelength of germanium. For the entire wavelength range, the photodetector with the thin Al_2O_3 interfacial layer exhibited higher responsivity and detectivity than the photodetector without an interfacial layer.

4 Conclusion

The merits of a graphene/n-Ge heterojunction photodetector with a thin Al_2O_3 interfacial layer are successfully demonstrated. The dark current could be reduced by two orders of magnitude at a bias of 2 V, and the specific detectivity was improved to $1.9 \times 10^{10} \text{ cm}\cdot\text{Hz}^{1/2}\text{W}^{-1}$. The responsivity was improved to 1.2 AW^{-1} , and a very high normalized photo-to-dark current ratio of $4.3 \times 10^7 \text{ W}^{-1}$ was obtained. These results indicate that a graphene/n-Ge heterojunction photodetector with a thin Al_2O_3 interfacial layer can be a very promising solution for infrared photodetector applications requiring low power consumption, low cost, and large area sensing simultaneously.

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