High performance mid-wave infrared photodetector based on graphene/black phosphorus heterojunction

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Abstract
Black phosphorus (BP) as a promising candidate for mid-wave infrared (MWIR) detection has attracted much attention. However, the high-speed photoresponse at the MWIR is yet to be a challenge. In this paper, we report a BP-graphene heterostructure photodetector with fast photoresponse at MWIR range using molybdenum electrode as a contact to realizing a low Schottky barrier. The device exhibits a fast photoresponse in a broad-spectrum range from visible to MWIR (0.67–4.2 μm). A high photovoltaic responsivity up to 183 mA W\(^{-1}\) and EQE up to 35.6% were realized at the visible range of 0.637 nm and respectively 7.9 mA W\(^{-1}\) and 0.31% at MWIR 3098 nm. The specific detectivity \(D^* = 6.69 \times 10^8\) Jones is obtained at 1 mV bias under the illumination of the MWIR 4.25 μm in the ambient condition. Our work may open a new way to realizing fast MWIR photoresponse at the low light level.

1. Introduction

The photodetector is the core of various technologies such as video imaging [1], optical communication [2], night vision [3], remote sensing [4], and biomedical imaging [5]. Two dimensional (2D) materials including graphene, black phosphorus (BP), and Emerging transition metal dichalcogenides (TMDs), are the research focus in recent years [6, 7]. Due to unique physics properties such as strong light–matter interactions [8], high performance at device applications [9], and easily stacked to form heterostructures [10], make those 2D materials to be the most promising candidates for channel materials of the next-generation optoelectronic device [11, 12]. Graphene is one of the most frequently studied 2D materials for broad spectra photodetection from visible to long-wave infrared and even to THz range due to its gapless band structure [13, 14]. However, the photoresponsivities of those graphene base photodetectors are low only a few milliamperes per watt to few tens of milliamperes per watt. The low responsivity could be ascribed to the short lifetime of the photogenerated carriers and low light absorption (2.3% for monolayer graphene) [15]. The ultrahigh photoresponsivity photodetectors can be realized by hybrid graphene with PbS quantum dots [16], fabricating heterostructures such as G-Silicon heterostructure [17], G-MoS\(_2\) heterostructures [18], and reduced graphene-oxide-MoS\(_2\) heterostructures [19, 20]. However, the high photoresponse is obtained at the cost of the speed of the photoresponse [21]. Another issue of the graphene-based photodetector is large dark current and high noise current power which could be ascribed to the gapless bandstructure [15]. Narrow bandgap 2D semiconductor BP with high hole mobility, direct bandgap 0.3 eV is excellent for mid-wave IR detection [22, 23]. The bandgap...
could be tuned by alloying with arsenic and an external electric field which can be used as long-wave IR detection [24]. However, black phosphorus is unstable in the ambient air. Owing to the development of the transfer technique, various van der Waals (vdW) heterostructures could be fabricated without limited by lattice matching [25]. The novelty of the structure and properties of the BP and graphene (BP-G) vdW heterostructures provides broad prospects in a variety of applications for photodetectors [26–30]. But up till now, there are few studies on ultra-wide wavelength photodetectors using graphene and BP. Here, we report a Mo-BP-G heterostructure photodetector, using a molybdenum electrode as a mirror to enhance the light absorption. The photocarrier could be effective collecting as the low Schottky barrier between the Mo electrode and BP the vertical device structure. Ultra-broadband photoresponse from visible to MWIR (0.63–4.2 μm) with fast speed is realized.

2. Experimental

The electrode pattern (typically 5 nm Ti/25 nm Mo) was fabricated using ultraviolet lithography and electron beam evaporation on a silicon wafer coated with silicon dioxide (300 nm). The BP flacks and graphene films were fabricated and transferred to the corresponding Mo and Mo electrode using the standard mechanical exfoliation method. The thicknesses of BP and graphene were further verified by Raman spectroscopy (HORIBA-JY) using a 532 nm excitation laser. The microstructures of BP and graphene were further verified by Raman spectroscopy, as shown in figure 1(a) and 1(b) (see support information). The vertical atomic thin heterostructures were obtained using a polymer-free assembly technique and transfer to the corresponding Mo electrode. The microstructures of BP and graphene were further verified by Raman spectroscopy (HORIBA-JY) using a 532 nm excitation laser. The Electrical transport measurements were implemented using the Keithley 2636 A dual-channel digital source meter. A semiconductor laser with a spot size of ~60 μm was used to detect the light response related to the mid-wavelength, and the beam diameter is ~3 mm.

3. Results and discussion

Figure 1(a) shows a schematic diagram of the Mo-BP-G vertical heterojunction photodetector. The bottom Mo electrode works as a mirror to enhance the light absorption. The top graphene layer can effectively prevent the degradation of BP. To protect the device, a PMMA layer was spin-coated in a glove box to prevent the device from exposing itself in the air. The data of stability for this Mo-BP-G heterostructure photodetector is presented in figure 1(c). After the device was exposed to air for more than two months, the photoresponse doesn’t show any decrease (see support information). As shown in figure 1(b), the microscope image illustrates that the morphology of the BP-G film is uniform. Then we study the electrical transport properties and photoresponse of this Mo-BP-G heterostructure device. The I-V curve of a typical Mo-BP-G device is plotted in figure 1(c). The good linear I-V curve indicates that good contact is obtained. A width range of the I-V curve from −1 V to 1 V is shown in the inset of figure 1(c). A low Schottky barrier of φb ≈ 0.093 eV was obtained from figures s2(a) and s2(b) (see support information). The details are described in supporting materials. The photocarrier could be collected effectively for the low barrier at the Mo-BP interface and high mobility graphene. The microstructures of BP and graphene were further verified by Raman spectroscopy, as shown in figure 1(d). Under 532 nm laser excitation, the G peak at 1586 cm$^{-1}$ and 2D peak at 2663 cm$^{-1}$ are more obvious, $I_{2D}/I_G$ is ~ 0.5, and FWHM$_{2D}$ is ~72.7 cm$^{-1}$, which indicated obvious characteristics of multilayer graphene. The low-frequency Raman peaks of BP film at 358, 435, and 462 cm$^{-1}$ corresponded to the $A_1^\text{g}$, $B_{2g}$, and $A_2^\text{g}$ vibration modes of BP lattice, respectively.

To study the photoresponse of this Mo-BP-G device, we measure the temporal photoresponse at various incidence light power as shown in figure 2(a). The incidence laser wavelength is 637 nm with a spot size of ~60 μm. To operate at a low dark current, the bias voltage was set at zero. As the laser incidence on, the current increases sharply, while switch of the light, the current decreased rapidly. Then we extract the photocurrent and calculated the photoresponsivity (R) of the external quantum efficiency (EQE). The photoresponsivity is defined as the ratio of photocurrent to incident power $R = I_p/I_0$ [9]. The EQE is one of the key figures of merit for photodetector, which is the ratio of the number of photoexcited charge carriers to the number of incident photons and can be expressed as $\text{EQE} = (h c R/e \lambda)$, where $h$ is the Planck constant, $c$ is the speed of light, and $\lambda$ is the wavelength of the incident laser [9]. The incident light power dependence R and EQE is shown in figure 1(b). For the photovoltaic response, the measured R and EQE at 637 nm is up to 0.183 A W$^{-1}$ and 35.63% respectively. The EQE is higher than that of the multilayer p-n junction of MoS$_2$-WSe$_2$ (34%) [31]. This could be ascribed to the mirror structure Mo electrode which could enhance the light absorption and fast carrier collection. As the incidence of light power increasing, the $R$ and $\text{EQE}$ were shown slightly decreasing. As the incidence of light power increasing, the density of the photogenerated carrier is increased considerably which enhances the interaction between the photocarrier and causes the decrease in the lifetime of photocarriers. The
recombination rate of the photocarriers increased which caused the decrease of the photoresponsivity. Then we
study the photoresponse at short wave infrared range. We measured the temporal photoresponse under a
1310 nm laser. The bias was set at 1 mV. As shown in figure 2(c), the photocurrent increased as the incidence
light increasing. When the light power increased to 2.4 μW, the photocurrent is up to 72.67 nA. The calculated R
and EQE as a function of incidence light power are plotted in figure 2(d). At the low intensity of the incidence
light power, the R and EQE become saturated. As the light power increasing, the R and EQE decreased, which
could be attributed to the limited density of the trap centers [32, 33]. At the higher light intensity, the number of
available trap centers is reduced, leading to a reduction of photoresponsivity. The photoresponsivity up to
51.5 mA W⁻¹ and EQE up to 10.1% are realized under the illumination of 1310 nm at 1 mV bias. The temporal
photovoltaic response at MWIR is present in figure s4(a). While at somewhat higher bias $V_{ds} = 10$ mV, the
response becomes slow, but a much higher photocurrent was realized, as shown in figure s4(b) (see support
information).

Furthermore, the photoresponse of this BP-G device at the mid-wave infrared range was studied. We extract
the time-varying photocurrent at 1 mV bias from visible to MIR (0.637–4.3 μm). At the wavelength of 637 nm,
as shown in figure 3(a), the photocurrent can be effectively modulated by periodically turning the light on and
off in seconds. A stable photocurrent of about 4.5 μA can be observed. The temporal photoresponse under the
switch illumination of 2611 nm, 3098 nm, and 3662 nm are respectively plotted in figures 3(b)–(d). As shown in
figure 3(b), the photocurrent is up to 16.8 nA under the illumination of 8.28 μW. Figure 3(c) shows the
photocurrent curves of 3098 nm at 1 mV. the photocurrent is dived to 4.6 nA. It can be seen that the response
was weak but stable. In figure 3(d), at 1 mV bias under the illumination of 3662 nm laser, the photocurrent of
16.9 nA is obtained at the exciting power of 8.9 μW. The results indicated that the photoresponse was fast at the
mid-wave infrared range with a low bias of 1 mV.
Then we extract the photoresponsivity and EQE at 1 mV bias from visible to MIR (0.637–4.3 μm). The R and EQE as a function of wavelength are plotted in figure 4(a). From 0.637 μm to 3.662 μm the R and EQE decrease as the wavelength increases, while from 3.662 μm to 4.25 μm, the R and EQE show an increasing tendency. This could be attributed to the high light absorption of the BP flake (0.3 eV direct bandgap, corresponding to 4.13 μm at this range).

To evaluate the sensitivity of this BP-G heterostructure device, the current noise density spectra were measured at different bias as shown in figure 4(b). At the low-frequency part (1–1 kHz), the 1/f noise dominated the contribution. When the frequency larger than 1 kHz, the current noise density spectrum becomes frequency independent. At the high-frequency range, the shot noise is the major originate of the noise which could be evaluated by $i_n^2 = 2e\langle i_d^2 \Delta f \rangle$. At 1 mV bias, the $i_d = 0.2 \mu A$ and $\Delta f = 1$ Hz, the $\langle i_n^2 \rangle = 6.4 \times 10^{-26}$ A$^2$ Hz$^{-1}$, which is consistent well with the experimental result at the frequency larger than 1 kHz. In the low-frequency range ($f < 1$ kHz), the 1/f noise is the dominant contribution, where the noise current is much higher than that estimated by shot noise. At this frequency range, the noise current should be calculated by $\langle i_n^2 (\omega) \rangle = [2e\langle i_d \rangle + i_n^2 (\omega)] \Delta f$ [40]. Another important figure of merit for photodetector, the noise equivalent power (NEP), other, is defined as NEP = $i_n/R$ [9]. From the measure noise density spectrum, we calculated the $\langle i_n \rangle$ using the formula of $\langle i_n^2 \rangle = \frac{1}{B} \int_0^B \langle i_n \rangle^2 df$, where the B is the measuring bandwidth [33, 41]. The NEP as a function of wavelength is presented in figure 4(c). The room temperature operation NEP of a typical BP-G heterostructure device is 2.1 pW Hz$^{-1/2}$ at MWIR 4.25 μm and 0.43 pW Hz$^{-1/2}$ at 0.637 μm. One of the most important figure-of-merits is the specific detectivity, $D^* = (AB)^{1/2}$/NEP, which determines the minimum illumination light power that a detector can distinguish from the noise [9]. In figure 4(d), the red solid circle line is the $D^*$ of a typical BP-G heterostructure. From visible to MWIR (0.637–4.25 μm), the $D^*$ is larger than that of...
the uncooled bolometer $10^8$ Jones. At the 4.24 \(\mu\)m, the \(D^*\) is up to $6.69 \times 10^8$ Jones, which is better than the b-AsP phototransistor [24] and uncooled commercial PbSe detector and is comparable to the PdSe$_2$ phototransistor [38]. The \(D^*\) of this Mo-BP-G heterostructure could be enhanced by inducing a hole barrier and applied at a higher bias which could enhance the photon gain. Another important figure-of-merit is response speed. We measured the response time of a typical device at 1 mV bias under a 637 nm laser. The rise/decay time is defined as the time that forms 10/90\% to 90/10\% of the stable photocurrent after turning the laser on/off. The rise time \(\tau_r = 12.6 \mu s\) and decay time \(\tau_d = 13.2 \mu s\) is obtained as shown in figures 4(e) and 4(f), which corresponds to the −3 dB response bandwidth of 3.87 kHz according to the previous work [42]. The performances of broadband photodetection based on graphene and topological insulator material heterostructures are summarized in table 1 for comparison.

To illustrate the photoresponse mechanism of the Mo-BP-G photodetector, the photocurrent mapping measurement was carried out by scanning the laser spot on typical devices. The optical image of a typical Mo-BP-G photodetector device is present in figure 5(a). The scale bar is 5 \(\mu\)m. Figure 5(b) shows the photocurrent mapping of the device as present in figure 5(a). The wavelength of the exciting laser is 830 nm and the bias is set at 0. As shown in figure 5(b), the strongest positive photoresponse appears in the junction region of Mo-BP-G, which is highlighted by a white solid line. And the high negative photoresponse appears at the area of the Mo-BP junction, highlighting by the red solid line. This could be explained as the orientation built-in electric fields are different. The bandstructure aliment is shown in figure 5(c). The work function of Mo is 4.6 eV [43] and graphene is 4.5 eV [44]. Conduction and valence band edges of BP based on the reported electron affinity values 4.4 eV in the literature [45]. The Fermi level of Mo is a bit (less than 0.1 eV) higher than that of BP, the electron transfer from Mo to the BP. The orientation of the built-in field from Mo to BP is formed as shown in the upper
For the graphene contact to the BP, the Fermi level of graphene is 4.5 eV. More electrons are transferred from graphene to BP during contact. The built-in field at the interface between BP and graphene is larger than that at the interface between Mo and BP. The orientation of the built-in field at the interface between BP and graphene is from graphene to BP, which is in the opposite direction with that of at the interface of Mo-BP. At zero bias, the photogenerated carriers transfer to the opposite direction at those two regions as shown in the down panel of figure 5(d).

4. Conclusions

In summary, we report a Mo-BP-G heterostructure photodetector with Mo as a bottom electrode. Ultrabroad band photodetection from under visible to MWIR (0.637 μm to 4.25 μm) was demonstrated based on this Mo-BP-G heterostructure. The high photovoltaic responsivity up to 0.183 A W⁻¹ and EQE up to 35.6% were realized at a visible range of 0.637 μm light. The specific detectivity \( D^* = 6.69 \times 10^8 \) Jones is obtained at room temperature under the illumination of the MWIR 4.25 μm. The bottom Mo electrode plays a role of a mirror to enhance light absorption. Moreover, a low Schottky barrier is formed at the Mo-BP interface, which is helpful for photocarrier collecting. This work may pave a way for low energy consumption, high sensitivity, and fast MWIR detection.
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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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