

Ultrahigh sensitive near-infrared photodetectors based on MoTe₂/ germanium heterostructure

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ABSTRACT

The efficient near-infrared light detection of the MoTe₂/germanium (Ge) heterojunction has been demonstrated. The fabricated MoTe₂/Ge van der Waals heterojunction shows excellent photoresponse performances under the illumination of a 915 nm laser. The photoresponsivity and specific detectivity can reach to 12,460 A/W and 3.3×10^{12} Jones, respectively. And the photoresponse time is 5 ms. However, the MoTe₂/Ge heterojunction suffers from a large reverse current at dark due to the low barrier between MoTe₂ and Ge. Therefore, to reduce the reverse current, an ultrathin GeO₂ layer deposited by ozone oxidation has been introduced to the MoTe₂/Ge heterojunction. The reverse current of the MoTe₂/GeO₂/Ge heterojunction at dark was suppressed from 0.44 μ A/ μ m² to 0.03 nA/ μ m², being reduced by more than four orders of magnitude. The MoTe₂/Ge heterojunction with the GeO₂ layer also exhibits good photoresponse performances, with a high responsivity of 15.6 A/W, short response time of 5 ms, and good specific detectivity of 4.86 × 10¹¹ Jones. These properties suggest that MoTe₂/Ge heterostructure is one of the promising structures for the development of high performance near-infrared photodetectors.

KEYWORDS

heterojunction, photodetector, MoTe₂, Ge, near-infrared

1 Introduction

Over the past few years, near-infrared photodetectors have been widely studied for their great practical importance, such as target detection, telecommunication, and thermal imaging [1-5]. Transition-metal dichalcogenides (TMDCs), a type of two dimensional (2D) materials, have been attracted extensive interest in photodetectors due to their excellent features including ultrathin structure, tunable band structures with different layers, free of dangling bonds and high light absorbance [6–9]. However, most of the photodetectors based on TMDCs such as molybdenum disulfide (MoS2) and tungsten diselenide (WSe2) are operated in the visible-wavelength range because of their relative large bandgaps [10-12]. As a new member of TMDCs, molybdenum ditelluride (MoTe₂) consists of Te-Mo-Te layers bonded by van der Waals forces. The bandgap of MoTe₂ ranges from the indirect bandgap of 0.83 eV for bulk to the direct bandgap of 1.1 eV for monolayer, which is comparably smaller than those of other TMDCs such as MoS₂ and WSe₂ [13]. The small bandgap extends the light absorption spectrum of TMDCs from the visible to the near-infrared, rendering MoTe2 promising for realizing high performance near-infrared photodetectors. The broad spectral range photodetection (0.6–1.55 μ m) based on MoTe₂ material has been demonstrated [14]. Furthermore, MoTe₂-based photodetectors with the structures of metalsemiconductor-metal (MSM) and heterojunction have been intensively investigated [15-18]. For practical photodetectors,

fast response speed is particularly important. However, the transit time of carriers between two contact electrodes in the photodetectors based on MSM structure would limit the response speed [19]. As an alternative, photodetectors based on heterojunctions usually rely on the interlayer built-in electric field, which can immediately separate photogenerated electron-hole pairs and result in high speed [20]. In the literatures, many materials have been used to form heterojunction photodetectors with MoTe₂, such as MoS₂ [21], graphene [22], and InGaZnO [23]. Germanium (Ge), a semiconductor material of group IV, has the bandgap of 0.67 eV. Owing to its small bandgap induced large absorption coefficient at near-infrared frequencies and excellent compatibility of parallel processing with silicon technology, Ge is also regarded as a good candidate for designing near-infrared photodetectors [24, 25]. Photodetectors based on Ge material with different structures such as MSM, p-i-n, and heterojunction have been widely investigated [26-28]. Furthermore, 2D materials are easily to form heterojunctions with semiconductors. The high performance near-infrared photodetector based on graphene/Ge heterojunction has been demonstrated [29]. However, the combination of MoTe₂ and Ge has been not investigated yet, which is desirable to obtain high-performance near-infrared photodetectors.

In this work, we first investigate the $MoTe_2$ and Ge heterojunction for near-infrared photodetector applications. The $MoTe_2$ and Ge heterojunction photodetector shows a high responsivity of 12,460 A/W, a short response time of 5 ms,

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and a good specific detectivity of 3.3×10^{12} Jones. These results suggest that the MoTe₂ and Ge heterojunction shows significant applicability in future high-performance near-infrared photodetectors.

2 Result and discussion

2.1 Characterization of the MoTe₂/Ge heterojunction

Bulk 2H-MoTe₂ crystals are purchased from HQ Graphene. Figure 1(a) shows the schematic diagram of the MoTe₂/Ge heterojunction photodetector. The Ge substrate is intrinsic state without doping. Firstly, a 30-nm-thick Al₂O₃ is deposited on Ge substrate by atomic layer deposition (ALD) at 200 °C and patterned into square matrix with 400 μ m × 400 μ m using ultra-violet lithography. Then, few-layer MoTe₂ materials are produced by the mechanical exfoliation method and transferred to the Ge substrate with patterned Al₂O₃ windows. Before transfer process, the Ge substrate is soaked in 5% HCl solution to remove the surface oxides. And the transfer process is conducted through the poly(dimethylsiloxane) (PDMS)-assisted transfer method, which allows the precise determination of the effective device area located at the window position. Finally, a 40-nm-thick Pt electrode is deposited at the edge of the window area to form the top contact with MoTe₂, while a 100-nm-thick Al film serves as the back contact with Ge. The optical image of the as-fabricated device is shown in Fig. 1(b). The MoTe₂ film remains continuous and complete after being transferred to the Ge substrate. The atomic force microscopy (AFM) image of the MoTe₂/Ge heterojunction is shown in Fig. 1(c). According to the AFM results, the thickness of MoTe₂ is approximately 16.2 nm. Figure 1(d) depicts the Raman spectra of the MoTe₂/Ge heterojunction, MoTe₂ film, and Ge. Three typical Raman peaks and one typical Raman peak are observed on MoTe2 film and Ge, respectively. And these four Raman peaks are both observed on the MoTe₂/Ge heterojunction, indicating the interlayer coupling between MoTe₂ and Ge. The electrical and optical properties of MoTe₂/Ge heterojunction device are characterized with a Keithley 4200-SCS parameter analyzer under a probe station in ambient air at room temperature. The photoresponse is measured under illumination of a laser with the wavelength of 915 nm. And the radius of the laser spot is 0.2 mm, which is large enough to cover the heterojunction. Furthermore, except the MoTe₂/Ge heterojunction region, the most area of Ge window is covered with a 100-nm-thick Pt film, which is used to serve as the light blocking layer of the Ge window.



Figure 1 (a) Schematic of the developed MoTe₂/Ge heterojunction with patterned window. (b) Optical image of the fabricated MoTe₂/Ge heterojunction device. (c) AFM image of the MoTe₂/Ge heterojunction. (d) Raman spectra of the MoTe₂/Ge heterojunction, Ge and MoTe₂.

2.2 Electrical and optical properties of the MoTe₂/Ge heterojunction

The current-voltage (I-V) curves of the MoTe₂/Ge heterojunction under different light intensities are shown in Fig. 2(a). At dark state, a rectification ratio (the ratio of forward current to reverse current, $|I_{fwd}|/|I_{rev}|$) of 133 is obtained. At illuminated states, an obvious photocurrent is observed under reverse bias voltage and the photocurrent increases significantly with the increase of light intensity. However, the photocurrent under forward bias voltage is negligible. The different results between reverse and forward bias voltage arise from that the heterojunction barrier at the MoTe₂/Ge interface under reverse bias voltage is larger than that under forward bias voltage and thus a more effective separation of photogenerated electron-hole pairs can be attained under reverse bias voltage. Hence, the photoresponse under reverse bias voltage is much faster than that under forward bias voltage. Moreover, it can also be observed that the minimum current point of the *I*-*V* curves shifts toward the forward voltage under illumination. This behavior results from the photovoltaic activity, which indicates that the device can operate without an external power source [30, 31]. Photocurrent $(I_{\rm ph})$ at V = -2 V and $|I_{\rm fwd}|/|I_{\rm rev}|$ at $V = \pm 2$ V of the MoTe₂/Ge heterojunction are plotted as a function of light intensity as shown in Fig. 2(b). It can be found that I_{ph} at V =-2 V is increased almost linearly with the increase of light intensity and the $I_{\rm ph}$ of 44.5 μ A is achieved when the power of the incident light is 66.65 mW/cm². The exponent θ can be calculated by $I_{\rm ph} \approx P^{\theta}$ [32–34], where $I_{\rm ph} = I_{\rm light} - I_{\rm dark}$ represents the photocurrent, Ilight and Idark represent the current at illuminated and dark states, respectively, and P is the light intensity. Through the curve-fitting method with the power function, the θ is estimated to be around 0.93. Furthermore, the $|I_{\rm fwd}|/|I_{\rm rev}|$ decreases significantly with the light intensity rising, which results from the significant increase of $|I_{rev}|$ with light intensity. Photoresponsivity (R) and specific detectivity (D^{*}) are crucial factors in photodetector performance, which can be calculated by the formula $R = I_{\rm ph}/PS$ and $D^* = RS^{1/2}/(2qI_{\rm dark})^{1/2}$, respectively, where S is the effective illumination area and q is the electron charge [33]. Figure 2(c) plots R and D^* of the MoTe₂/Ge heterojunction as a function of light intensity at V = -2 V. Both R and D^* increase with the increase of light intensity and show relatively high values of 12,460 A/W and 3.3×10^{12} Jones when the power of the incident light is 1.09 mW/cm², respectively. Moreover, the time-dependent photoresponse properties of the MoTe₂/Ge heterojunction are investigated by modulating the laser with a square wave and recording the photocurrent. As shown in Fig. 2(d), the device exhibits good photoswitching stability and high photosensitivity. The rise time (t_R) and the decay time (t_D) are defined as the time taken to go from 10% to 90% and from 90% to 10% of the total photocurrent, respectively. The regions where current rises and falls with time are enlarged as shown in Figs. 2(e) and 2(f), respectively. Both $t_{\rm R}$ and $t_{\rm D}$ are around 5 ms, indicating that electron-hole pairs are effectively generated, separated, and recombined in the MoTe₂/Ge heterojunction.

To better understand the photoresponse mechanism of the MoTe₂/Ge heterojunction, the band alignment of the heterojunction is displayed in Fig. 3(a). Since the MoTe₂ with Pt contacts is p-type conduction while the Ge is intrinsic state without doping [35, 36], Fermi level of MoTe₂ is lower than that of Ge. Therefore, electrons would diffuse from Ge to MoTe₂, while holes would diffuse from MoTe₂ to Ge. As a result, a built-in electric field is formed at the MoTe₂/Ge interface at the equilibrium state as shown in Fig. 3(b). The barrier could



Figure 2 (a) I-V curves of the MoTe₂/Ge heterojunction under 915 nm laser illumination with different light intensities. (b) The photocurrents at -2 V and $|I_{\text{fwd}}|/|I_{\text{rev}}|$ at ± 2 V of the MoTe₂/Ge heterojunction depend on the light intensity. (c) Photoresponsivity and specific detectivity of the MoTe₂/Ge heterojunction as a function of light intensity. (d) The continuous current-time cycles of the MoTe₂/Ge heterojunction. The enlarged regions where current (e) rises and (f) falls with time.



Figure 3 (a) Band alignments of the MoTe₂/Ge heterojunction. (b) Energy band diagrams of the MoTe₂/Ge heterojunction at the equilibrium state.

be adjusted by the bias voltage, which will be increased under reverse bias voltage while decreased under forward bias voltage. Furthermore, the Schottky barriers between semiconductors (MoTe₂ and Ge) and contact metals (Al and Pt) are formed [35, 36]. When the photodetector is under illumination, Ge and MoTe₂ both absorb photons to generate electron–hole pairs, then the photoexcited electron–hole pairs will be separated by the built-in electric field at the MoTe₂/Ge interface and the Schottky barriers between semiconductors (MoTe₂ and Ge) and contact metals (Al and Pt). And this process leads to the generation of photocurrent in the device.

2.3 Electrical and optical properties of the MoTe₂/ GeO₂/Ge heterojunction

As shown in Fig. 3(a), the ΔE_c between MoTe₂ and Ge is 0.33 eV And the ΔE_v between MoTe₂ and Ge is almost negligible. Hence, the hot carriers generated and transported between MoTe₂ and Ge are large, leading to a non-negligible reverse current at dark (I_{leak}). The I_{leak} of the MoTe₂/Ge heterojunction is 0.44 μ A/ μ m². The large I_{leak} may limit the further performance improvements and increase the power consumption of the photodetectors. It has been reported that inserting an interfacial oxide layer would reduce the *I*_{leak} in the graphene/Si heterojunction photodetector [37]. Therefore, in order to effectively suppress the *I*_{leak} of the MoTe₂/Ge heterojunction, an ultrathin interfacial GeO₂ layer is inserted between MoTe₂ and Ge in this work. Before the transfer of the MoTe₂ film, the GeO₂ layer is grown on the patterned Ge substrate by ozone oxidation at 300 °C for 30 min using the same ALD system. The schematic diagram is shown in Fig. 4(a), where a GeO₂ layer is sandwiched between MoTe₂ and Ge. The band diagram of the MoTe₂/GeO₂/Ge heterojunction is shown in Fig. 4(b). The thin GeO₂ layer serves as a blocking layer to reduce hot carriers transporting between MoTe₂ and Ge and thus suppress the I_{leak} of the MoTe₂/Ge heterojunction. The MoTe₂/GeO₂/Ge heterojunction is characterized by transmission electron microscope (TEM) as shown in Fig. 4(c). A 2-nm-thick GeO_2 layer is formed on the surface of Ge after ozone oxidation. And the MoTe₂ is around 21 layers with the thickness of around 16 nm, which is similar with that of the MoTe₂/Ge heterojunction. The I-V curves under dark of the MoTe₂/Ge heterojunction with and without



Figure 4 (a) The schematic diagram of the MoTe₂/GeO₂/Ge heterojunction. (b) Energy band diagrams of the MoTe₂/GeO₂/Ge heterojunction at the equilibrium state. (b) The TEM image of the MoTe₂/GeO₂/Ge heterojunction. (d) The *I*–*V* curves under dark of the MoTe₂/Ge heterojunction with and without GeO₂ layer.

GeO₂ layer are plotted in Fig. 4(d). After the insertion of GeO₂ layer, the I_{leak} is reduced by more than four orders of magnitude, reaching to a very small value of 0.03 nA/µm². Furthermore, the forward current at dark is also reduced due to the existence of the GeO₂ layer, which results from that the GeO₂ layer would also block the transport of carriers under forward bias voltage. However, the reduction under reverse current is more obvious than that under forward current, leading to the increase of the

rectification ratio after inserting the GeO_2 layer. A rectification ratio of 654 is obtained after the insertion of the GeO_2 layer, compared to that of 133 without the GeO_2 layer.

After optimizing the Ileak of the MoTe2/Ge heterojunction by inserting a GeO₂ layer, the optoelectronic properties of the MoTe₂/GeO₂/Ge heterojunction are systematically investigated. The I-V characteristics of the MoTe₂/GeO₂/Ge heterojunction under 915 nm laser illumination at different light intensities are shown in Fig. 5(a). Consistent with the MoTe₂/Ge heterojunction without the GeO₂ layer, an obvious photocurrent can be observed under reverse bias voltage while negligible photocurrent is attained under forward bias voltage at illuminated states. The current increases significantly under reverse bias voltages as the light intensity increases. Furthermore, the minimum current point of the I-V curve also shifts toward the forward voltage under illumination, which is like the results of the MoTe₂/Ge heterojunction without the GeO₂ layer. And the shift is more obvious in the MoTe₂/GeO₂/Ge heterojunction, which indicates that the photovoltaic activity is more obvious after the insertion of the GeO₂ layer. Figure 5(b) shows the $I_{\rm ph}$ at -2 V and $|I_{\text{fwd}}|/|I_{\text{rev}}|$ at ± 2 V as a function of light intensity for the MoTe₂/GeO₂/Ge heterojunction. The I_{ph} increases and the $|I_{\rm fwd}|/|I_{\rm rev}|$ decreases significantly with the light intensity rising. And the θ of $I_{\rm ph}$ and light intensity is estimated to be around 1. When the power of the incident light is 66.65 mW/cm^2 , the $I_{\rm ph}$ of 0.73 μ A is achieved. The value is smaller than that of the MoTe₂/Ge heterojunction without the GeO₂ layer, which results from the existence of the GeO₂ block layer. R and D^* of the MoTe₂/GeO₂/Ge heterojunction as a function of light intensity



Figure 5 (a) I-V curves of the MoTe₂/GeO₂/Ge heterojunction under 915 nm laser illumination with different light intensities. (b) The photocurrents at -2 V and $|I_{\text{fwd}}|/|I_{\text{rev}}|$ at ± 2 V of the MoTe₂/GeO₂/Ge heterojunction depend on the light intensity. (c) Photoresponsivity and specific detectivity of the MoTe₂/GeO₂/Ge heterojunction as a function of light intensity. (d) The continuous current-time cycles of the MoTe₂/GeO₂/Ge heterojunction. The enlarged regions where current (e) rises and (f) falls with time.

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at -2 V are displayed in Fig. 5(c). The maximum values of R and D^* are 15.6 A/W and 4.86 × 10¹¹ Jones, respectively when the power of the incident light is 1.09 mW/cm². The reduced *R* and D^* in the MoTe₂/GeO₂/Ge heterojunction result from the reduced $I_{\rm ph}$. However, they are still compared to those of other reported MoTe₂-based photodetectors [14, 35, 38, 39]. The time-dependent photoresponse properties of the MoTe₂/GeO₂/Ge heterojunction are also investigated. As shown in Fig. 5(d), the good photoswitching stability is well-preserved after the insertion of the GeO₂ layer. The regions where current rises and falls with time are enlarged as shown in Figs. 5(e) and 5(f), respectively. Both t_R and t_D are 5 ms, which is consistent with the MoTe₂/Ge heterojunction without the GeO₂ layer. It indicates that the effect of the generation, separation and recombination of electronhole pairs are still effective in the MoTe₂/Ge heterojunction after inserting the GeO₂ layer. Table 1 presents the characteristic comparison of the MoTe₂/Ge heterojunction with/without a GeO₂ layer with other MoTe₂-based photodetectors in previous reports. The heterostructures in this work display good comprehensive photodetection performance, which indicates the MoTe₂/Ge heterostructure has great potential in applications for future photodetectors.

 Table 1
 Performance comparison of the photodetectors in this work with other reported MoTe₂-based photodetectors

Materials	Wavelength (nm)	Responsivity (A/W)	Response time (ms)	Detectivity (Jones)	Ref.
MoTe ₂ /Ge	915	12,460	5	$3.3 imes 10^{12}$	This work
MoTe ₂ /GeO ₂ /Ge	915	15.6	5	$4.86 imes 10^{11}$	This work
MoTe ₂	1,060	0.024	1.3	$1.3 imes 10^9$	[14]
MoTe ₂	680	6	0.16	—	[35]
MoTe ₂ /graphene	1,064	970	78	1.55×10^{11}	[38]
MoTe ₂ /MoS ₂	633	0.073	68	—	[39]

3 Conclusions

In summary, a high-performance near-infrared photodetector is obtained based on the structure of MoTe₂/Ge heterojunction. The optoelectronic properties of the MoTe₂/Ge heterojunction under 915 nm laser illumination have been systematically studied. The MoTe₂/Ge heterojunction photodetector shows good optoelectronic performances with photoresponsivity and specific detectivity of as high as 12,460 A/W and 3.3×10^{12} Jones, respectively. And the photoresponse time of 5 ms is attained. Moreover, in order to reduce the reverse current at dark, a thin GeO₂ layer is introduced to the MoTe₂/Ge heterojunction. After inserting the GeO₂ layer, the reverse current at dark of the MoTe₂/Ge heterojunction is reduced by more than four orders of magnitude. And the MoTe₂/Ge heterojunction with a GeO₂ layer also exhibits good optoelectronic performances with high responsivity (15.6 A/W), short response time (5 ms), and good specific detectivity (4.86 \times 10¹¹ Jones), which is compared to those of other reported MoTe2-based photodetectors. All these results make MoTe₂/Ge heterojunction a promising structure for next generation high-performance photodetectors.

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