

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

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#### **ABSTRACT**

The efficient near-infrared light detection of the MoTe<sub>2</sub>/germanium (Ge) heterojunction has been demonstrated. The fabricated MoTe<sub>2</sub>/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  $3.3 \times 10^{12}$  Jones, respectively. And the photoresponse time is 5 ms. However, the MoTe<sub>2</sub>/Ge heterojunction suffers from a large reverse current at dark due to the low barrier between MoTe<sub>2</sub> and Ge. Therefore, to reduce the reverse current, an ultrathin GeO<sub>2</sub> layer deposited by ozone oxidation has been introduced to the MoTe<sub>2</sub>/Ge heterojunction. The reverse current of the MoTe<sub>2</sub>/GeO<sub>2</sub>/Ge heterojunction at dark was suppressed from 0.44  $\mu$ A/ $\mu$ m<sup>2</sup> to 0.03 nA/ $\mu$ m<sup>2</sup>, being reduced by more than four orders of magnitude. The MoTe<sub>2</sub>/Ge heterojunction with the GeO<sub>2</sub> 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  $\times$  10<sup>11</sup> Jones. These properties suggest that MoTe<sub>2</sub>/Ge heterostructure is one of the promising structures for the development of high performance near-infrared photodetectors.

#### **KEYWORDS**

heterojunction, photodetector, MoTe<sub>2</sub>, 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 (MoS<sub>2</sub>) and tungsten diselenide (WSe<sub>2</sub>) are operated in the visible-wavelength range because of their relative large bandgaps [10–12]. As a new member of TMDCs, molybdenum ditelluride (MoTe<sub>2</sub>) consists of Te-Mo-Te layers bonded by van der Waals forces. The bandgap of MoTe<sub>2</sub> 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  $MoS<sub>2</sub>$  and WSe<sub>2</sub> [13]. The small bandgap extends the light absorption spectrum of TMDCs from the visible to the near-infrared, rendering MoTe<sub>2</sub> promising for realizing high performance near-infrared photodetectors. The broad spectral range photodetection (0.6–1.55 μm) 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 MoTe<sub>2</sub>, such as MoS<sub>2</sub> [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  $MoTe<sub>2</sub>$  and  $Ge$ heterojunction for near-infrared photodetector applications. The MoTe<sub>2</sub> 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  $3.3 \times 10^{12}$  Jones. These results suggest that the MoTe<sub>2</sub> and Ge heterojunction shows significant applicability in future high-performance near-infrared photodetectors.

## **2 Result and discussion**

#### **2.1 Characterization of the MoTe<sub>2</sub>/Ge heterojunction**

Bulk 2H-MoTe<sub>2</sub> crystals are purchased from HQ Graphene. Figure 1(a) shows the schematic diagram of the  $MoTe<sub>2</sub>/Ge$ heterojunction photodetector. The Ge substrate is intrinsic state without doping. Firstly, a  $30$ -nm-thick  $Al_2O_3$  is deposited on Ge substrate by atomic layer deposition (ALD) at 200 °C and patterned into square matrix with 400  $\mu$ m  $\times$  400  $\mu$ m using ultra-violet lithography. Then, few-layer MoTe<sub>2</sub> materials are produced by the mechanical exfoliation method and transferred to the Ge substrate with patterned  $Al_2O_3$  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 MoTe<sub>2</sub>, 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  $MoTe<sub>2</sub>$  is approximately 16.2 nm. Figure 1(d) depicts the Raman spectra of the  $MoTe_{2}/Ge$ heterojunction, MoTe<sub>2</sub> 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 MoTe<sub>2</sub>/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 MoTe<sub>2</sub>/Ge heterojunction with patterned window. (b) Optical image of the fabricated MoTe2/Ge heterojunction device. (c) AFM image of the MoTe<sub>2</sub>/Ge heterojunction. (d) Raman spectra of the MoTe2/Ge heterojunction, Ge and MoTe2.

## 2.2 Electrical and optical properties of the MoTe<sub>2</sub>/Ge **heterojunction**

The current–voltage  $(I-V)$  curves of the MoTe<sub>2</sub>/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 MoTe<sub>2</sub>/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  $(I_{ph})$  at  $V = -2$  V and  $|I_{fwd}|/|I_{rev}|$  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*ph of 44.5 μA is achieved when the power of the incident light is 66.65 mW/cm2 . The exponent *θ* can be calculated by  $I_{ph} \approx P^{\theta}$  [32–34], where  $I_{ph} = I_{light} - I_{dark}$  represents the photocurrent, *I*light and *I*dark 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*fwd|/|*I*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  $R = I_{ph}/PS$  and  $D^* = RS^{1/2}/(2qI_{dark})^{1/2}$ , respectively, where *S* is the effective illumination area and *q* is the electron charge [33]. Figure 2(c) plots *R* and *D*<sup>\*</sup> of the MoTe<sub>2</sub>/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  $3.3 \times 10^{12}$  Jones when the power of the incident light is 1.09 mW/cm<sup>2</sup>, respectively. Moreover, the time-dependent photoresponse properties of the MoTe<sub>2</sub>/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  $(t<sub>R</sub>)$  and the decay time  $(t<sub>D</sub>)$  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  $t<sub>R</sub>$  and  $t<sub>D</sub>$  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 MoTe<sub>2</sub> with Pt contacts is p-type conduction while the Ge is intrinsic state without doping  $[35, 36]$ , Fermi level of MoTe<sub>2</sub> is lower than that of Ge. Therefore, electrons would diffuse from Ge to MoTe<sub>2</sub>, while holes would diffuse from MoTe<sub>2</sub> 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  $|I_{fwd}|/|I_{rwd}|$  at  $\pm 2$  V of the MoTe<sub>2</sub>/Ge heterojunction depend on the light intensity. (c) Photoresponsivity and specific detectivity of the MoTe<sub>2</sub>/Ge heterojunction as a function of light intensity. (d) The continuous current-time cycles of the MoTe<sub>2</sub>/Ge heterojunction. The enlarged regions where current (e) rises and (f) falls with time.



Figure 3 (a) Band alignments of the MoTe<sub>2</sub>/Ge heterojunction. (b) Energy band diagrams of the MoTe<sub>2</sub>/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 (MoTe<sub>2</sub> 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 MoTe<sub>2</sub>/ GeO<sub>2</sub>/Ge heterojunction

As shown in Fig. 3(a), the  $\Delta E_c$  between MoTe<sub>2</sub> and Ge is 0.33 eV And the Δ*E*v between MoTe2 and Ge is almost negligible. Hence, the hot carriers generated and transported between MoTe<sub>2</sub> and Ge are large, leading to a non-negligible reverse current at dark ( $I_{\text{leak}}$ ). The  $I_{\text{leak}}$  of the MoTe<sub>2</sub>/Ge heterojunction is 0.44  $\mu$ A/ $\mu$ m<sup>2</sup>. 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 *Ileak* of the MoTe<sub>2</sub>/Ge heterojunction, an ultrathin interfacial  $GeO<sub>2</sub>$  layer is inserted between MoTe<sub>2</sub> and Ge in this work. Before the transfer of the MoTe<sub>2</sub> film, the GeO<sub>2</sub> 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 GeO<sub>2</sub> layer is sandwiched between MoTe<sub>2</sub> and Ge. The band diagram of the MoTe<sub>2</sub>/GeO<sub>2</sub>/Ge heterojunction is shown in Fig. 4(b). The thin  $GeO<sub>2</sub>$  layer serves as a blocking layer to reduce hot carriers transporting between MoTe2 and Ge and thus suppress the *I*leak of the MoTe<sub>2</sub>/Ge heterojunction. The MoTe<sub>2</sub>/GeO<sub>2</sub>/Ge heterojunction is characterized by transmission electron microscope (TEM) as shown in Fig.  $4(c)$ . A 2-nm-thick GeO<sub>2</sub> layer is formed on the surface of Ge after ozone oxidation. And the MoTe<sub>2</sub> is around 21 layers with the thickness of around 16 nm, which is similar with that of the MoTe<sub>2</sub>/Ge heterojunction. The *I*-*V* curves under dark of the MoTe2/Ge heterojunction with and without



**Figure 4** (a) The schematic diagram of the MoTe<sub>2</sub>/GeO<sub>2</sub>/Ge heterojunction. (b) Energy band diagrams of the MoTe<sub>2</sub>/GeO<sub>2</sub>/Ge heterojunction at the equilibrium state. (b) The TEM image of the MoTe<sub>2</sub>/GeO<sub>2</sub>/Ge heterojunction. (d) The *I*–*V* curves under dark of the MoTe2/Ge heterojunction with and without GeO<sub>2</sub> layer.

 $GeO<sub>2</sub>$  layer are plotted in Fig. 4(d). After the insertion of  $GeO<sub>2</sub>$ layer, the *I*leak is reduced by more than four orders of magnitude, reaching to a very small value of  $0.03 \text{ nA}/\mu\text{m}^2$ . Furthermore, the forward current at dark is also reduced due to the existence of the  $GeO<sub>2</sub>$  layer, which results from that the  $GeO<sub>2</sub>$  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  $GeO<sub>2</sub>$  layer. A rectification ratio of 654 is obtained after the insertion of the  $GeO<sub>2</sub>$  layer, compared to that of 133 without the  $GeO<sub>2</sub>$  layer.

After optimizing the *I*<sub>leak</sub> of the MoTe<sub>2</sub>/Ge heterojunction by inserting a GeO<sub>2</sub> layer, the optoelectronic properties of the MoTe2/GeO2/Ge heterojunction are systematically investigated. The  $I-V$  characteristics of the  $MoTe_{2}/GeO_{2}/Ge$  heterojunction under 915 nm laser illumination at different light intensities are shown in Fig.  $5(a)$ . Consistent with the MoTe<sub>2</sub>/Ge heterojunction without the  $GeO<sub>2</sub>$  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 MoTe<sub>2</sub>/Ge heterojunction without the GeO<sub>2</sub> layer. And the shift is more obvious in the  $MoTe<sub>2</sub>/GeO<sub>2</sub>/Ge$  heterojunction, which indicates that the photovoltaic activity is more obvious after the insertion of the GeO<sub>2</sub> layer. Figure 5(b) shows the  $I_{ph}$ at −2 V and |*I*fwd|/|*I*rev| at ±2 V as a function of light intensity for the MoTe2/GeO2/Ge heterojunction. The *I*ph increases and the |*I*fwd|/|*I*rev| decreases significantly with the light intensity rising. And the  $\theta$  of  $I_{ph}$  and light intensity is estimated to be around 1. When the power of the incident light is 66.65 mW/cm<sup>2</sup>, the *I*ph of 0.73 μA is achieved. The value is smaller than that of the MoTe<sub>2</sub>/Ge heterojunction without the GeO<sub>2</sub> layer, which results from the existence of the GeO<sub>2</sub> block layer. *R* and *D*<sup>\*</sup> of the MoTe<sub>2</sub>/GeO<sub>2</sub>/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*fwd|/|*I*rev| at ±2 V of the MoTe2/GeO2/Ge heterojunction depend on the light intensity. (c) Photoresponsivity and specific detectivity of the MoTe<sub>2</sub>/GeO<sub>2</sub>/Ge heterojunction as a function of light intensity. (d) The continuous current–time cycles of the MoTe<sub>2</sub>/GeO<sub>2</sub>/Ge heterojunction. The enlarged regions where current (e) rises and (f) falls with time.

**TSINGHUA 12) Springer** | www.editorialmanager.com/nare/default.asp at −2 V are displayed in Fig. 5(c). The maximum values of *R* and  $D^*$  are 15.6 A/W and  $4.86 \times 10^{11}$  Jones, respectively when the power of the incident light is 1.09 mW/cm<sup>2</sup>. The reduced *R* and *D*<sup>\*</sup> in the MoTe<sub>2</sub>/GeO<sub>2</sub>/Ge heterojunction result from the reduced *I*ph. However, they are still compared to those of other reported MoTe<sub>2</sub>-based photodetectors [14, 35, 38, 39]. The time-dependent photoresponse properties of the MoTe<sub>2</sub>/GeO<sub>2</sub>/Ge heterojunction are also investigated. As shown in Fig. 5(d), the good photoswitching stability is well-preserved after the insertion of the  $GeO<sub>2</sub>$  layer. The regions where current rises and falls with time are enlarged as shown in Figs. 5(e) and 5(f), respectively. Both  $t<sub>R</sub>$  and  $t<sub>D</sub>$  are 5 ms, which is consistent with the MoTe<sub>2</sub>/Ge heterojunction without the  $GeO<sub>2</sub>$  layer. It indicates that the effect of the generation, separation and recombination of electron– hole pairs are still effective in the MoTe<sub>2</sub>/Ge heterojunction after inserting the  $GeO<sub>2</sub>$  layer. Table 1 presents the characteristic comparison of the MoTe<sub>2</sub>/Ge heterojunction with/without a GeO<sub>2</sub> layer with other MoTe<sub>2</sub>-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	(nm)	Wavelength Responsivity Response Detectivity (A/W)	time (ms)	(Jones)	Ref.
MoTe <sub>2</sub> /Ge	915	12,460	5	$3.3 \times 10^{12}$	This work
MoTe <sub>2</sub> /GeO <sub>2</sub> /Ge	915	15.6	5	$4.86 \times 10^{11}$	This work
MoTe <sub>2</sub>	1,060	0.024	1.3	$1.3 \times 10^{9}$	[14]
MoTe <sub>2</sub>	680	6	0.16		$[35]$
MoTe <sub>2</sub> /graphene	1,064	970	78	$1.55 \times 10^{11}$	$[38]$
MoTe <sub>2</sub> /MoS <sub>2</sub>	633	0.073	68		[39]

## **3 Conclusions**

In summary, a high-performance near-infrared photodetector is obtained based on the structure of MoTe<sub>2</sub>/Ge heterojunction. The optoelectronic properties of the MoTe2/Ge heterojunction under 915 nm laser illumination have been systematically studied. The MoTe<sub>2</sub>/Ge heterojunction photodetector shows good optoelectronic performances with photoresponsivity and specific detectivity of as high as 12,460 A/W and  $3.3 \times 10^{12}$  Jones, respectively. And the photoresponse time of 5 ms is attained. Moreover, in order to reduce the reverse current at dark, a thin GeO<sub>2</sub> layer is introduced to the MoTe<sub>2</sub>/Ge heterojunction. After inserting the  $GeO<sub>2</sub>$  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  $\times$  10<sup>11</sup> Jones), which is compared to those of other reported MoTe<sub>2</sub>-based photodetectors. All these results make MoTe<sub>2</sub>/Ge heterojunction a promising structure for next generation high-performance photodetectors.

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