TOPICAL REVIEW
Two-dimensional materials applied for room-temperature thermoelectric photodetectors

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Abstract
Due to the practical demand in many fields, room-temperature photodetectors in mid/long-wavelength and terahertz ranges have attracted much attention. Photothermoelectric (PTE) detectors based on photothermal conversion and thermoelectric effect can realize ultra-broadband detection of a photon without external bias. In recent years, two-dimensional (2D) materials open up revolutionary opportunities in rapid and sensitive photodetection by virtue of their remarkable electronic and optical properties. Here, we provide a brief review of state-of-the-art photodetectors based on PTE effect and 2D materials. It is worth noting that emerging PTE detectors based on 2D materials, including graphene, transition metal dichalcogenides (TMDCs), black phosphorus (BP) and MXenes, are proposed systematically. Next, we will discuss the existing challenges and prospects in PTE detectors, followed by a conclusion of this review.

1. Introduction

Due to the emergence of multi-dimensional materials and integration technologies, high-performance photodetectors operating at room temperature show huge commercial demand in practical applications, such as wearable health monitors [1], biosensors [2], gas or humidity sensors [3] and energy harvesting and storage [4]. However, photodetectors still suffer from a plethora of problems. For example, the bandgap of the materials may cause an impact for the spectral response [5]. Typically, traditional mercury cadmium telluride (HgCdTe) is challenging to fabricate and has high toxicity [6–8]. The commonly used quantum-well photodetectors require a cryogenic operation situation. Herein, with both advantages of room-temperature working range and no required cryogenic cooling unit, photothermoelectric (PTE) detectors are fast-emerging. The requirements for PTE detectors have also been dramatically increasing according to their responsivity, response time and noise level, as well as flexibility, transparency and reliability.

PTE effect mainly relies on photothermal conversion and thermoelectric effect (i.e. Seebeck effect) [9]. PTE mechanism converts absorbed light energy into temperature difference via different material electrodes. Then, an electric potential is established across a temperature gradient due to the movement and scattering of charge carriers within the semiconductors from the hot end to the cold end. For photothermal conversion, light-induced temperature change can be denoted as,

\[ \sum \Delta T = E_{\text{abs}} - E_{\text{loss}} \tag{1} \]

where \( E_{\text{abs}} \) is the absorbed energy into the materials, \( E_{\text{loss}} \) is the loss of the energy (heat flows to the surroundings), \( t \) is the duration of photothermal conversion, \( m \) and \( C_i \) are the mass and specific heat capacities of different components [10]. Hence, for high PTE detectors, the materials should obtain a higher photon absorption coefficient and smaller thermal conductivity.

As for the thermoelectric effect can be expressed with Seebeck coefficient \( S \), the temperature difference between the two electrodes \( \Delta T \) and the voltage difference \( \Delta U \),
\[
\Delta U = -S \times \Delta T
\]
(2)

\[
S = -\frac{\pi^2 k_B T}{3e} \left( \frac{d \ln \sigma}{dE} \right)_{E=E_f}
\]
(3)

\[
S = \frac{8L}{h^2 \hbar} \ln^{*} + 4 \left( \frac{\pi}{3n} \right)^{4/3}
\]
(4)

Seebeck coefficient is expressed via the Mott relationship in degenerate systems for metal and doped semiconductors, where \(E_f\) is Fermi energy, \(k_B\) is Boltzmann constant, \(\sigma\) is the electrical conductivity of materials, \(m^*\) is the effective mass of the carrier and \(n\) stands for the carrier concentration. Mott’s relationship reveals the relationship between electrical conductivity and its variation with Fermi energy. As for the materials with strong electron-phonon coupling, the phonon drag effect contributes to increasing the Seebeck coefficient by phonons’ diffusion.

The overall efficiency of a thermal electric materials can be characterized by a dimensionless figure of merit ZT value,

\[
ZT = \frac{S^2 \sigma T}{k}
\]
(5)

Where \(k\) is thermal conductivity, to achieve a higher ZT value, materials are expected to obtain a high Seebeck coefficient, a large electrical conductivity, and a low thermal conductivity to retain sufficient temperature gradient. Current commonly used materials of thermoelectric detector may have a relatively high ZT value but most of them have a relatively low photothermal conversion. Additionally, they need extra voltages to make these detectors work. Besides bio-inspired materials may have high photothermal conversion but low ZT values hinder their use on PTE devices.

The emergence of PTE-related materials starts from graphene. Since the advent of first monolayer graphene, graphene has become a competitive two-dimensional (2D) material in optoelectronic devices because it exhibits excellent mechanical, optical, thermal and electronic properties [11]. For instance, phototransistors [12], organic light-emitting diodes (OLED) [13], optical modulators [14], transparent conductive films [15] and phototheranostic devices [16] have been increasingly investigated due to the development of graphene. As a kind of popular optoelectrical devices, photodetectors draw much attention based on the exploitation of various properties of graphene and other materials. Carbon-based graphene is composed of carbon atoms arranged in a hexagonal honeycomb lattice with zero bandgap. Such a gapless band structure of graphene helps activate charge carriers by photon absorption over an ultrabroad electromagnetic spectrum from ultraviolet (UV) to terahertz spectral regimes. Besides, graphene has high charge carrier mobility \((10^5 \sim 10^6 \text{ cm}^2/\text{Vs})\) [17, 18], which is beneficial to convert optical signals to electrical signals at a relatively fast speed. Furthermore, high thermal conductivity up to \(5 \text{ kW mK}^{-1}\) for suspended monolayer graphene at ambient temperature and phototunable properties via the doping process are equally essential properties for graphene as active materials among optoelectronic devices, such as PTE detectors [19–21]. In the last decade, graphene-based optoelectronic devices are sparkling and gradually commercialized because of their reduced price and integration on a large scale. Common semiconductor materials, due to the energy and momentum conservation, are strictly difficult to release relaxation and hard to fulfill at the same time. By virtue of zero-band gap semiconductor model, and the linear band structure, graphene will allow the carrier multiplication via auger-type processes and impact ionization, thus leading to the carrier dynamics for energy transportation and bridge the conduction and valance band [22–24]. Therefore, strong electron interactions are established through photon excitation with multiple carriers, which enhance PTE effects. Moreover, to seek for more promising 2D materials with small bandgap, various materials with direct or indirect bandgap, such as transition metal dichalcogenides (TMDs), black phosphorus (BP), single-element 2D materials (Xenes), nitrides and carbonitrides (MXenes), and their derivatives are being widely investigated. These common layered 2D materials show weak interlayer bonding by van der Waals interaction and strong covalent in-plane bonding. A typical example utilizing such a structure is that TMDs can control layers and adjust the bandgap by controlling layers to achieve light detection at various wavelengths [25]. According to the layered structure, most atoms after exfoliation are uncovered on the surface \(\sim \sim\\), and the surface areas of these materials will expand, which is beneficial to activate their physical and chemical properties and impact the quantum confinement effect [26]. At present, a promising configuration is to combine graphene and related 2D materials to fabricate functional, high-performance photodetectors.

In this paper, we mainly refer to the current PTE detectors based on 2D thermoelectric materials only warmed by incident photons and ignore those materials that require additional photothermal conversion. Such a structure of PTE detectors exhibits some advantages that include the utilization of hot-carrier PTE effect, the formation of high-compact arrays of PTE detectors and the introduction of nonequilibrium dynamic effect [27].
We will summarize different 2D materials used in PTE detectors, followed by some existing challenges in the PTE photodetection platform and potential strategies to optimize the functions (Figure 1).

2. Photothermoelectric detectors based on two-dimensional materials

2.1. Graphene
Photons can be absorbed with banding energy inside graphene from ultraviolet to terahertz, and even to microwave radiation regime, possibly because of the interband transitions of carriers and gapless band structure. Due to its inherent thinness, single-layer graphene shows a limited capacity of optical absorption, and this may restrict the responsivity of the device (ca. 0.5 mA W$^{-1}$) [28]. However, the heat-carrier-assisted phenomenon may help improve the responsivity. After the generation of electron-hole pairs, the interaction between electrons triggers the relaxation of the absorbed energy among charge carriers, and then these carriers are heated. Without involving the photothermal transport, the hot-carrier-assisted PTE effect can happen in graphene, as a result of the ultrafast speed and high responsivity [27]. Generally, the electron temperature is the same as the lattice temperature, but the equilibrium effect can be broken in graphene under photoexcitation. During this process, the most crucial parameter in this response is the cooling length. If the cooling length of carriers is much smaller than the channel length of graphene, the lattice temperature difference dominates and drives the diffusion of carriers instead of the hot-electron-assisted mechanism [29, 30]. Graphene, as a typical zero-band gap material, its structure improves the multiple-excitation in the absorption of a single photon by enhancing an abundance of hot carriers. However, for higher THz waves, the absorption efficiency of graphene is reduced because of the Pauli blocking. Additionally, disorders still plays the role of the transition in doped graphene [31].

2.2. Transition metal dichalcogenides
Due to the fact that graphene has no bandgap, some graphene-based digital optoelectronic devices are difficult to fabricate. With the advantage of tunable bandgap, TMDCs can bridge the gap. For instance, the bulk molybdenum disulfide (MoS$_2$) exhibits an indirect bandgap of 1.2 eV. In contrast, the monolayer MoS$_2$ shows a large direct bandgap of 1.8 eV, resulting in a large room-temperature Seebeck coefficient of monolayer MoS$_2$ up to 30 mV K$^{-1}$. With the mechanical flexibility and simple processing, TMDCs can provide additional advantages compared with the conventional direct-semiconductor device. Furthermore, allowing for the strong interband absorption, TMDC-based photon detectors are expected to be optimized.

With the remarkable mechanical property, single- and few-layer MoS$_2$ and tungsten diselenide (WSe$_2$) are most widely investigated among TMDCs in optoelectronic devices. In single-layer MoS$_2$ photodetector, the evident PTE response was reported between MoS$_2$ and the titanium electrode [32]. The responsivities at 532 nm above-band excitation and 750 nm sub-band excitation are 9.6 and 1 V W$^{-1}$. During the gradient from low-resistance to
high-resistance state, an increase of PTE voltage by two orders of magnitude caused an improved Seebeck coefficient (ca. $3 \times 10^2 \mu V K^{-1}$). Zhang et al [33] demonstrated the PTE effects in multilayer MoS2 with Ti/Au electrodes. They used scanning photocurrent microscopy to prove the mechanism of PTE appearing in the MoS2-metal interface, which dominated in the accumulation regime. Besides, the anomalously large Seebeck coefficient was observed in multilayer MoS2, and explained as hot photo-excited carriers to increase the thermoelectric transitions. For WSe2, the double-gated WSe2 homojunction interface was created due to its ambipolar behavior [34]. According to different illumination regions, the PTE current can be demonstrated and dominates in the typical region, such as the p-p region. Presently, the centrosymmetric distribution of PTE voltage is also observed in WTe2 nanoflakes [35]. TMDCs are promising materials because these new materials have a strong possibility to tune down the bandgap with other metal/semiconductor nanomaterials and most of them obtain a relatively high ZT value. However, TMDCs are not quite sensitive with light, comparing to other 2D PTE materials, the photon-thermal transition is smaller than 5–10 times and therefore challenges still wait for improvements.

2.3. Black phosphorus
As an allotrope of phosphorus, BP has the graphene-like configuration. The bandgap of this materials depends on the thickness about 0.3 eV of bulk and about 2 eV of monolayer that is larger than monolayer graphene and akin to TMDs (1.2–1.8 eV) [37]. By virtue of the sp3 hybridization within the layer, BP exhibits a different puckered hexagonal arrangement for single-layer and bilayer, along with the armchair and zigzag direction, respectively (figure 2), which leads to the anisotropic of band structure and that of in-plane properties. BP can be applied into high-speed photonic applications because BP shows high mobility up to 1000 cm$^2$ V$^{-1}$ s$^{-1}$ compared with TMDCs ($\sim$200 cm$^2$ V$^{-1}$ s$^{-1}$) [38]. Besides, BP shows a large Seebeck coefficient about 355 $\mu V K^{-1}$ at room temperature [39]. Accordingly, BP can become a promising candidate in PTE detectors due to the abovementioned properties.

Competing mechanisms between PV and PTE was investigated for BP-based field-effect transistor (FET) [41]. This experiment measured the photocurrent based on the spatial, polarized, gate, and bias-dependent properties. As shown in the photocurrent response near the electrode contacts, PTE effect dominates in the on state but PV dominates in the off state (figure 3). Then, Wang et al [42] designed a BP-based photodetector utilizing the electromagnetic diffraction and infrared photon transition with the antenna-integrated sensitive element, which shows a high responsivity in the near-infrared (1–450 V W$^{-1}$) and in the terahertz regime (1–300 V W$^{-1}$).

A BP-based PTE detector was fabricated in the THz regime by Guo et al [40] in a channel about 30 nm in length. The PTE effect is amplified in the tiny photoactive area with the intensity improvement of largely localized electric-field beyond the skin-depth limit. Due to the use of evaporation technology, the asymmetric fabrication contacts helped to enhance the PTE effect. With the above fabrication, photon absorption was enhanced, and preferential hot carrier flow without bias could be realized. This design also can achieve a broadband detection (figure 3) for other 2D materials for a thermal imaging setup with excellent responsivity up to a 297 V W$^{-1}$. Besides, this device exhibited a low power consumption and NEP below 58 pW Hz$^{-1/2}$, and response time is under 0.8 ms, which shows a better performance than room-temperature thermal detectors.

The main reason for restricting BP-based applications is ambient instability. When uncovered to oxygen or water for a period, the phosphorus atoms may chemically degrade to phosphorus oxides. Several strategies to enhance stability have been widely verified by protective layer or ionophore coating [43, 44], physical encapsulation and chemical doping modulation [45, 46]. Furthermore, Thurakkal et al [37] summarized recent progress in the chemical functionalization of 2D BP nanosheets, which has been verified as a useful approach, such as efficient passivation.
2.4. MXenes

Depending on the surface functionalization types, MXenes show tunable metal or semiconductor properties [47, 48]. Presently, the most widely investigated MXene is Ti\(_3\)C\(_2\)\(\text{Tx}\), which shows high electrical conductivity and broad optical absorption response [49, 50]. The spectral absorption region ranges from visible to near-infrared [51]. Zuo et al. [52] proposed a Ti\(_3\)C\(_2\)\(\text{Tx}\)-based optical sensor (figure 4), which can show an evident temperature profile. Besides, the device illustrates a fast response of 23.4 \(\mu\)s. Moreover, the photothermal conversion efficiency of Ti\(_3\)C\(_2\)\(\text{Tx}\) is high in the visible and near-infrared (NIR) regions [53], i.e. the absorbed photons can be efficiently converted to heat energy. Additionally, even if without any additional encapsulation, Ti\(_3\)C\(_2\)\(\text{Tx}\) can still remain ambiently stable for about one month [47]. With the excellent thermal stability in the air, MXenes have also been explored as a promising thermoelectric material [48, 54, 55], such as the thermoelectric figure of merit (ZT) value of 0.112 via doping, which demonstrates the viability of MXenes applied into PTE detectors. According to the abovementioned properties of MXenes, MXenes are potential as a sort of photoactive material in PTE detectors. At present, a wide range of theoretical studies of stacked Ti\(_3\)C\(_2\) flakes have been proposed [56–60], and more related experiments are expected to be performed. MXenes possess a relatively high photothermal and ZT value and broadband light absorption. However, most current MXenes procedures may require strict ambient surroundings during sample preparation. A large amount of sedimentary cannot dispersion well and drop down in the solvents without continuous stirring. Therefore, MXenes fabrication process should be optimized in the future.

3. Challenges and prospects of 2D materials-based PTE detectors

Self-powered, no extra voltages or magnet field-assisted PTE detectors are promising detectors. High photothermal and high thermoelectric conversion can be applied for many optical devices, communication components and wearable or flexible equipment. Considerable achievements are made for the exploration of 2D PTE materials and a great deal of progress has been accomplished during the seeking for suitable PTE materials. However, challenges have remained for improvements. Three major issues worth thinking about for future designs. First, both high photothermal and high thermoelectric conversion is hard to find on current materials. Most applied 2D PTE...
materials are former thermoelectric materials or photothermal materials. They may have a high ZT value or photon to heat conversion, but the other side is not as good as their advantage. Therefore, the overall PTE conversion percentage is not satisfied. Second, the detailed conversions in microscale are still not fully uncovered, especially for photothermal conversion. Therefore, the strategies for raising photon excitations are limited. Finally, the fabrication process is complex and needs to simplify. These nanomaterials are hard to dispersion well in samples and degradation is a major impact. Moisture and oxygen issues continue as placing PTE devices under ambient surroundings for several months, around 20%–30% degradation. Metal electrodes oxidation and polymer degradation may also speed up the degradation of PTE detectors. The long-term reliability of PTE detectors needs to be improved.

At present, 2D materials have attracted much attention, and many important results, especially for 2D materials-based PTE detectors, have been verified. Many strategies are considered to apply, and the performance of PTE detectors seems to be enhanced. The main methods focus on changing the single- or few-layer photoactive/thermoelectric materials to improve the performance of the PTE detectors [61]. A deep understanding or exploration of electric or photon transport mechanism is also required, which can essentially help design and guide the novel structures. In addition, facile and scalable fabrication process are encouraged. Given the requirements for the large responsivity of PTE detectors, materials are encouraged with excellent photon absorptivity, low heat capacity, high Seebeck coefficient, and proper size. Moreover, reasonable structure design can help decrease the heat dissipation to the environment and improve light absorption and responsivity [27], which can be optimized by adjusting light–matter interaction, including various polaritons [62–65], the cavity or waveguide integrated into photodetectors [66–69], antennas-based functional structures [70–74].

Due to the excellent photon and electronic properties, 2D materials-based PTE detectors are potential as portable wearable devices. Zhang et al [75] designed a device to detect the response of infrared radiation from the human body, ranging from mid- to long-wavelength. They placed one finger close to the array (about 2 mm) and detected a remarkable photocurrent. This demonstrates the feasibility of applying PTE detectors into wearable devices. Contrary to traditional photodetectors or other sensors, this device also exhibits advantages with self-powered and integration miniaturization. Given the flexibility and transparency of the detector array, this device also provides potential applications in many fields, such as acting as an assistant tool to help optimize the

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**Figure 4.** Overview of MXene-based optical device guidance for PTE detectors. (a) A typical SEM image of MXene flakes. (b) It shows the working temperature profile of the device. (c) It shows different working lengths of special 2D materials. (d) It shows the control efficiency and response time of the control efficiency and response time of different 2D materials. Reproduced from [32], © 2020 De Gruyter.
performance of the autonomous vehicles (Figure 5). In the environmental area, the device can sense the changes of light intensities via their matched photocurrent. Additionally, due to the different temperature regions inside the eyeball, it can make sense for ophthalmological devices under dark conditions.

4. Conclusions

In conclusion, by virtue of strong thermal, optical and electronic properties, 2D materials have achieved considerable progress in PTE detectors. Typically, with the introduction of the hot-carrier-assisted mechanism, the response time and speed have been significantly improved. However, their researches are still in the academic and laboratory stage. Herein, the first priority is to develop room-temperature low-cost ultrabroadband PTE detectors without external bias. Furthermore, an in-depth understanding of the fundamentals of performance enhancement is essential. It is expected that 2D materials-based PTE detectors can obtain notable accomplishments in the wearable display and related optoelectronic applications.

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