1. Introduction

The terahertz (THz) wave has become one of the international research hotspots in recent years due to its distinctive properties compared to other bands in the electromagnetic wave spectrum [1]. THz technologies are in demand in various fields such as astronomy, security, biomedicine and communications [2–5]. However, the practical application of THz technologies is strongly impeded by the lack of high-performance sources and detectors. Conventional THz detection can be largely classified into coherent and incoherent technologies. Coherent technologies include heterodyne mixing, ultrafast laser-based photoconductive and electro-optic sampling [1].

Schottky diodes, Golay cells, field-effect transistors (FETs), bolometers and pyroelectric detectors involve typical incoherent technologies [6]. These existing methods generally have some limitations in terms of sensitivity, bandwidth, cost, speed, operation temperature or integrability, and thus, novel technologies are highly desired for THz detection.

Carbon nanomaterials, including graphene and carbon nanotubes (CNTs), have emerged as extraordinary low-dimensional materials due to their unique electronic and photonic properties benefiting from the unusual crystal and electronic band structure [7]. The gapless nature of graphene, as well as its ultra-high carrier mobility and ultra-wide spectral absorption range, make it a promising candidate for the active elements in photodetectors from visible to THz frequencies [8]. CNTs, especially single-walled CNTs (SWCNTs), have chirality and therefore can be either metallic or semiconducting.

Keywords: graphene, photothermoelectric effect, THz photodetector, heterojunction

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Abstract

We report on the photoresponse of a graphene-metal contact device under terahertz (THz) illumination. The device has an extremely simple structure consisting of a large-area monolayer graphene stripe contacted with two gold electrodes. A significant position-dependent photovoltage is observed across the device by THz excitation, exhibiting a linear relationship with the incident beam power. Experimental results show that the graphene channel length and the substrate thermal conductivity have obvious influence on the photovoltage amplitude and response time, which is consistent with the photothermoelectric mechanism. This compact and powerless device is expected to have a promising application in THz detection. Our work provides theoretical and experimental evidence for the development of high-performance graphene-based THz photodetectors.

(Some figures may appear in colour only in the online journal)
This, coupled with orientation originating from the 1D nature, leads to different optical properties compared to graphene [9]. Recently, THz detectors based on carbon nanomaterials have been investigated and their responses upon THz excitation are dominated by various mechanisms such as photothermoelectric (PTE) effect, bolometric effect and plasma-wave-assisted mechanism [10–16]. When conductors or semiconductors, with different Seebeck coefficients, such as differently doped CNT films, graphene and metal are contacting and illuminated, the photo-induced temperature rise can produce photovoltage (PV) between the two materials due to the PTE effect. He et al developed a CNT PTE THz detector based on doped SWCNT film, demonstrating that the photoresponse originated from light-induced heating along the film [10]. Cai et al presented a sensitive room-temperature graphene PTE THz detector realized by creating a temperature gradient with asymmetric contact metals [11]. The bolometric effect is also of thermal origin, but is related to the change in transport conductance of the material when heated by the incident radiation. A typical example of the bolometric detector was made by coupling CNTs with a log-periodic toothed antenna, while the responsivity of such a bolometer depended strongly on the ambient temperature [12]. Besides, the epitaxial graphene quantum dot exhibited high performance as the THz bolometer at low temperature [13]. Benefiting from the ultra-high carrier mobility at room temperature, graphene can serve as the channel of an FET and support plasma wave oscillation launched by the incoming radiation. Vicarelli et al proposed graphene FETs with gate-tunable efficient plasmonic response stemming from THz excitation of plasma waves in the micrometer-scale graphene channel [14]. For the FET architecture, the plasmonic effect may not be the only detection mechanism and usually coexists with the PTE [15] or bolometric contributions [16].

However, the existing studies on graphene-based THz detectors either reported on devices which are complicated in structure and difficult to fabricate, or investigated the photoresponses of graphene-metal contact structures only in the visible and infrared ranges. Different from previous works, here we consider the photoresponse of graphene-metal contact structure under THz excitation and systematically investigate the photodetection mechanism, which has not been addressed, to the best of our knowledge. The merits of our devices are that they are structurally simple, compact, and easy to fabricate. We fabricated devices with heterostructure formed by large-area graphene stripe and Au electrodes, and experimentally characterized the influence of the illumination position, graphene channel length and substrate material on the response properties. The experimental results and theoretical analysis demonstrate that the PTE mechanism dominates the PV generation process in the devices.

2. Experiments

Figure 1(a) shows the geometry of our device. It has quite a simple structure consisting of a monolayer graphene stripe, two gold electrodes and a wafer severed as the substrate. The fabrication of the device can be described as follows. First, we put the trivial transfer graphene (TTG, ACS material) into deionized water to release the large-area monolayer graphene. In order to prevent the graphene from curling during
the releasing process, the TTG had to be wetted all around by deionized water in advance. The wet floating graphene was then extracted by a piece of filter paper and cut into stripes with a size of $5 \times 15 \text{ mm}$ with a sharp knife blade. After the stripe dried, it was released into deionized water again and removed by the substrate. Then, the substrate was immersed into acetone to get rid of the PMMA on the graphene. After a 30 min drying process in an air oven, a rectangular graphene stripe had been transferred onto the substrate. Finally, high-purity Au was deposited at the two ends of the stripe as electrodes using a vacuum-evaporation method. We fabricated our first device on a Teflon substrate with a channel length of 8 mm as a conceptual demonstration. To ensure the reliability of the whole fabricating process, we characterized the device with scanning electron microscopy (SEM) and Raman spectroscopy. As can be seen in the SEM image (figure 1(b)), both graphene and Au are uniform and a regular heterojunction formed at their interface. In addition, the graphene stripe adheres well to the substrate, differing from our previous work in which the CNT bundle was suspended [17]. Figure 1(c) shows the Raman spectrum of the graphene stripe. The G and 2D peaks lie at 1585 and 2675 cm$^{-1}$, respectively, and the 2D/G intensity ratio is 2.34, which confirms the monolayer nature of the graphene [18]. In our experiments, continuous-wave THz radiation generated from a far-infrared gas laser (FIRL 100, Edinburgh Instruments Ltd) was focused onto the device, with a frequency of 2.52 THz and a spot diameter of about 1.5 mm. The THz radiation emitted from the laser with a power of approximately 90 mW might be too strong for the device and therefore it was attenuated to 30% during the experiments. A sourcemeter (Keithley 2602B) was used to measure the current–voltage ($I$–$V$) characteristics of our device as well as the photocurrent it generated under THz illumination. The photocurrent combined with the resistance calculated from the $I$–$V$ relationship yields the PV in the device, which will be shown in the following section. All the experiments were performed at room temperature in ambient air.

3. Results and discussion

First, we test the $I$–$V$ characteristics of our device (Teflon substrate, 8 mm channel length) with the bias voltage ranging from $-10$ to $10$ mV, as shown in figure 2 (only the data between $-2$ and 2 mV are given here for clarity). The black line in figure 2(a) shows that the device has a nearly linear $I$–$V$ relationship without THz illumination, which means that the graphene stripe has an ohmic contact with the electrodes. The resistance of the device calculated from this measurement is about 3.5 kΩ. The red and blue lines exhibit obvious upward and downward shift with respect to the dark $I$–$V$ curve when the right junction and the left one were exposed to the THz beam with a power of about 23 mW, respectively. It is observed that a short-circuit photocurrent ($I_{sc}$) and an open-circuit PV ($V_{oc}$) are produced in response to the THz radiation. We determine that $I_{sc}$ is $\sim 70$ nA and $V_{oc}$ is $\sim 245$ µV for the right junction. Similar experimental phenomena have been reported in the CNT THz photodetectors in which the junctions were formed by the connection of p-doped and n-doped SWCNT films [10] or CNT-metal contacts [17]. Note that the resistance of the channel material may change obviously upon illumination due to bolometric effect, especially at liquid helium temperatures [19]. As the slope of the $I$–$V$ curve changes quite slightly in our case, we ignore the contribution of the bolometric effect to the photoreponse here. Figure 2(b) shows the power dependence of the PV within the range of our experimental conditions. The linear relationship between $V_{oc}$ and the incident power makes this simple structure a promising candidate as a THz detector, with a PV responsivity of about 8.4 mV W$^{-1}$ extracted from the curve. Compared to some devices based on aligned CNTs, the THz response of graphene-metal contact exhibits no obvious dependence on the polarization of the incident radiation, as shown in figure 2(c). This may not be surprising, because the graphene stripe does not show specific orientation as highly aligned CNTs do. It is possible to grasp the polarization information by modifying the current device. For instance, some subwavelength periodic patterns could be induced for the electrode structure or the graphene channel to enable polarization-dependent coupling or absorption of the THz beam [15, 20]. In order to understand the origin of the photoreponse, we performed scanning photocurrent measurement by moving the THz spot from the left junction to the right one along the graphene channel. Figure 2(d) illustrates the position-dependent $V_{oc}$ profile. The fact that the PV achieves its maximum with opposite signs at two junctions, combined with a value close to zero with THz spot focused at the middle of the channel, coincides well with the behavior of the PTE mechanism. Previous study on suspended and unsuspended CNT samples in the visible and THz range [17, 21, 22] as well as graphene devices in the visible range [23] also show similar position-dependent phenomena, as a significant evidence of the PTE effect in corresponding experiments. In the PTE process, it first undergoes light absorption and conversion to heat. When graphene is illuminated by visible light, interband transitions occur, induced by incident photons, leading to the generation of electron–hole pairs. However, intraband transitions dominate graphene absorption of THz radiation, i.e. it would be absorbed by free carriers instead of electron–hole pair excitation [24–27]. The incident photons will heat the carriers in graphene via strong electron–electron interactions. The hot carriers and the lattice finally reach thermal equilibrium via the scattering process between carriers and phonons [28]. The temperature gradient of the hot carriers produces a potential gradient which is given by [29]

\[
\nabla V = -S \cdot \nabla T
\]

(1)

where $S$ is the Seebeck coefficient and $T$ is the carrier temperature. The total voltage can be calculated by integrating the potential gradient over the device length, given by

\[
V_{PTE} = \int S \cdot \nabla T \, dx
\]

(2)

The Seebeck coefficient $S$ (also called thermoelectric power) is dependent on the electrical properties of the material according to the Mott formula [30]:

\[
S = \frac{k_B T}{e} \ln \left( \frac{m^*}{m_0} \right)
\]

where $k_B$ is the Boltzmann constant, $T$ is the temperature, $e$ is the elementary charge, $m^*$ is the effective mass, and $m_0$ is the free electron mass.
where \( e \) is the electron charge, \( k_B \) is the Boltzmann constant, \( \sigma \) is the electrical conductivity, \( \varepsilon \) is the electron energy, and the derivative must be evaluated at the Fermi energy. Note that we have to introduce some asymmetry into either the Seebeck coefficient or the carrier temperature distribution upon illumination to obtain a non-zero integral voltage value, in other words, a detectable PV along the device, according to formula (2), and several attempts have been made from different aspects in previous studies. In the asymmetric CNT device developed by Fedorov’s group, a logarithmic spiral antenna was coupled with the CNT film as the electrodes, half of the CNT film between the electrodes was sticking out of the catalyst islands, thus suspended, while the other half was attached closely to the substrate [19]. There was an asymmetric thermal distribution along the device when it was entirely illuminated by incident beam due to the asymmetric thermal contact between the CNT film and the substrate, and therefore PV arises due to the PTE effect. Moreover, contact asymmetries were applied in the room-temperature graphene THz detector reported by Cai et al in which the Au electrode was totally and partially overlapped with the Cr electrode, respectively, at the two ends of the graphene flake [11]. Consequently, the Fermi energy profile and therefore the Seebeck coefficient are asymmetric along the device, giving rise to obvious photoreponse upon incident THz wave. The remarkable design of the device structure, which utilizes asymmetries of both Seebeck coefficients and temperature profile, will enhance the device performance significantly, such as the above-mentioned p–n junction CNT detector [10]. As the p-doped and the n-doped films have Seebeck coefficients of opposite sign, the PV originates from individual contributions of each film. For the graphene-metal structure considered here, the asymmetry is introduced from the perspective of temperature distribution. Owing to the macro-scale of our device, we are able to focus the THz beam on one of the junctions to generate an asymmetric temperature profile, which means a distinct temperature difference at the two junctions. Within the PTE model, the device with our geometry can be simply considered as a thermocouple, with one arm corresponding to the graphene stripe and the other corresponding to the metal. The two junctions formed by the graphene and electrodes serve as two contact points of the arms. When the THz illumination was focused on a single junction, PV arises due to the temperature difference between the two junctions, which is given by [29]

\[
PV = (S_{Au} - S_G)(T_L - T_R)
\]

Figure 2. (a) \( I-V \) characteristics of the device (Teflon substrate, 8 mm channel length) in dark condition (black line) and when the THz beam with a power of about 23 mW was focused on the right junction (red line) and the left one (blue line). (b) Power dependence of the PV with a responsivity of approximately 8.4 mV W \(^{-1}\) extracted from the linear fit. (c) Variation of the normalized PV with the polarization of the incident beam changing continuously. (0° corresponds to the polarization parallel to the long edge of the graphene stripe). (d) Scanning photocurrent measurement of the device. The THz spot moved gradually from the left electrode to the right one.
graphene can be doped into p-type by the metal it contacted with, as well as gas and water molecules in the atmosphere [31–33]. Because the work function of graphene is generally different from those of metal surfaces, electrons are transferred at the interface to equilibrate the Fermi levels when graphene is contacted with metal. The Fermi level of intrinsic graphene coincides with the conical point (Dirac point), and it will shift downwards into the valence band when graphene is contacted with Au, making the graphene stripe p-doped [31]. Besides, when impurities adhere to the surface of graphene, they will influence the carrier density of graphene by electron transfer without breaking the chemical bond and lattices in it [32]. Gas and water molecules from the atmosphere adhering to the surface of graphene or even lying between graphene and the substrate would also dope graphene into p-type [32, 33].

In order to further investigate our device with the PTE model, the temperature profile along the device channel upon THz illumination was extracted from a heat conduction equation. In the equation, the incident circular Gaussian laser beam was modeled as a point heat source at the center of the beam. If we take the heating point as the origin and establish a coordinate axis along the graphene channel, the temperature profile under localized THz illumination can be given by [29]

\[ T(x) = T_{\max} e^{-\frac{|x|}{\lambda}} \]  

The maximum temperature is

\[ T_{\max} = \frac{P}{\sqrt{G \kappa_G W}} \]  

while the temperature decays exponentially along the channel with the thermal length scale given by

\[ \lambda = \sqrt{\frac{\kappa_G h}{G}} \]  

Moreover, we can also estimate how the temperature profile decays with time by a time-dependent heat equation, and the time scale given by this equation is

\[ \tau = \frac{\rho C_p}{G} \]  

In the expressions above, \( \kappa_G \) is the thermal conductivity of the graphene stripe, \( h \) is the thickness of the stripe, \( W \) is the stripe width, \( P \) is the optical power absorbed by the film, \( G \) is the thermal conductance between graphene and the substrate, \( \rho \) is the film mass density, and \( C_p \) is the heat capacity. According to the PTE theory, the device parameters such as the channel length and the substrate thermal conductivity will have a significant impact on the response characteristics such as the PV amplitude and the response time. So, more tests have been done to confirm the PTE nature of the device.

To evaluate the influence of the channel length on the photoresponse, another six devices also on the Teflon wafer with different channel lengths ranging from 0.2–8 mm were fabricated and tested. We periodically illuminated the devices with THz beam and obtained their temporal PV curves, which are shown in figure 3(a). Each time period was taken to be 120 s so that all the devices could have enough time to respond to the incident beam. One can clearly find that for the device with a channel length of 8 mm, when it is illuminated, the
PV increases most slowly, but finally reaches the maximum, which is the highest value among all these devices. However, the PV of the device with a channel length of 0.2 mm gets saturated in a very short period of time with a minimum voltage value. Comparing all the curves in figure 3(a), it is evident that the device channel length surely has a significant influence on its photoresponse properties, i.e. a longer channel length leads to a stronger but slower response in general. This may be easily understood considering that the longer the stripe is, the larger the junction temperature difference is when the device reaches thermal equilibrium, so that a larger PV occurs, at the same time as a longer response time due to a slower equilibrium process. Note that the THz spot was fixed at the right junction in all the tests here and below, and the PV can be formulated as a variable of the channel length $L$ by substituting (5) into (4)

$$PV = T_{\text{max}}(S_G - S_{\text{th}})(1 - e^{-\frac{L}{\tau}})$$

(9)

Indeed, the PVs extracted from figure 3(a) show a negative exponential relationship with the channel lengths, as fitted in figure 3(b), in good agreement with equation (9). Clearly, the PV of a device may finally reach saturation as the length of the graphene channel increases, indicating that further lengthening the stripe will not efficiently enhance the device performance. In fact, similar tests had been done in the visible range by changing the length of a SWCNT film gradually between two metal electrodes [21]. Although the PVs were not fitted with the film lengths in that work, the negative exponential relationship between them could be obviously observed through the data points that were shown. Also, the heat capacity of the graphene stripe $C_p$ is approximately proportional to the channel length, thus $\tau$ should also scale with it as concluded from equation (8). The response times extracted by exponential fitting of the curves in figure 3(a) are plotted in figure 3(c), exhibiting a good linear relationship with the channel lengths. Besides, the spot diameter may also influence the PV amplitude in a similar way to the channel lengths. Without changing the incident beam power, when the spot diameter increases, the temperature profile along the channel flattens gradually with the maximum temperature decreasing, owing to the decrease in optical power density. As a consequence, the temperature difference between the two junctions decreases, leading to a smaller PV.

As the thermal condition may significantly dominate the response characteristics of a heat sensitive device, two other kinds of substrates, quartz and sapphire, are chosen as alternatives to Teflon with the graphene channel length fixed at 8 mm. The temporal PV curves obtained with the same method described above are illustrated in figure 4(a). Taking into account that Teflon has the minimum thermal conductivity among the substrates, while sapphire has the maximum, we can find that a smaller substrate thermal conductivity generally leads to a stronger but slower response. This can be explained qualitatively that the substrate with a low thermal conductivity leads to a slow thermal equilibrium process with relatively high maximum temperature, and as a consequence, relatively large PV along the device. The thermal conductance $G$ in equations (6)–(8) is expected to be proportional to the substrate thermal conductivity $\kappa_{\text{sub}}$ thus implying $T_{\text{max}}$, and therefore, the PV should scale with $\kappa_{\text{sub}}^{-1/2}$ according to (6) and (9). At the same time, $\tau$ should scale with $\kappa_{\text{sub}}^{-1}$ according to (8).
These have been confirmed by the results extracted from the temporal PV experiments, as shown in figures 4(b) and (c), where values $\kappa_{\text{Teflon}} = 0.25 \text{ W/m} \cdot \text{K}$, $\kappa_{\text{quartz}} = 1.46 \text{ W/m} \cdot \text{K}$, and $\kappa_{\text{sapphire}} = 25 \text{ W/m} \cdot \text{K}$ are used [34–36]. A previous study on the PTE photodetector in the visible range drew similar conclusions to ours on how substrate thermal properties influence the photoresponse of the device [34]. He et al also found that the thermal environment had a significant impact on the voltage responsivity of the CNT THz detector [10]. To date, sufficient evidence has been produced from several aspects to confirm the PTE nature of our device.

4. Conclusion

In conclusion, we have investigated a graphene-metal contact structure upon THz excitation, which shows obvious photoresponse at room temperature. The linear relationship between the PV and the incident beam power makes it available for THz detection. The PV and response time extracted from the measurement results correspond with the PTE mechanism, indicating that the photoresponse originates from the asymmetric thermal distribution along the graphene channel. Furthermore, we conclude that a longer channel leads to a stronger but slower response, while a substrate with a larger thermal conductivity leads to a weaker but faster response, both from experiments and the PTE model. The compact and powerless properties of our device are expected to be useful for large-scale array integration, and our work provides ways to optimize graphene-based THz photodetectors with a similar structure.

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