High-quality infrared imaging with graphene photodetectors at room temperature†

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Graphene, a two-dimensional material, is expected to enable broad-spectrum and high-speed photodetection because of its gapless band structure, ultrafast carrier dynamics and high mobility. We demonstrate a multispectral active infrared imaging by using a graphene photodetector based on hybrid response mechanisms at room temperature. The high-quality images with optical resolutions of 418 nm, 657 nm and 877 nm and close-to-theoretical-limit Michelson contrasts of 0.997, 0.994, and 0.996 have been acquired for 565 nm, 1550 nm, and 1815 nm light imaging measurements by using an unbiased graphene photodetector, respectively. Importantly, by carefully analyzing the results of Raman mapping and numerical simulations for the response process, the formation of hybrid photocurrents in graphene detectors is attributed to the synergistic action of photovoltaic and photo-thermoelectric effects. The initial application to infrared imaging will help promote the development of high performance graphene-based infrared multispectral detectors.

Introduction

An infrared (IR) detector, as the core component of an IR imaging system, has achieved great improvement in sensing capability during the last century because of its important applications in the military, civilian and astronomy fields.1–3 Previously, a variety of IR detectors have been developed by using narrow band gap semiconductors such as ternary alloys, and group III–V and group IV semiconductors. Although the advanced technologies of material growth and fabrication processing have made rapid progress in the study of IR detectors,1,2 how to achieve broadband absorption and fast response is still the main problem limiting system performance.4 In the recent decade, the advent of new materials, especially nano-materials having distinct physical and optical characters,5–10 offers a new opportunity to develop and improve the next-generation IR photodetector system.

As an attractive two-dimensional (2D) material, graphene has drawn a lot of attention in the study of optoelectronic devices due to its unique properties including broadband photon absorption,7,8 ultrafast carrier dynamics9 and high mobility10 in comparison with other 2D crystals. Diverse prototype photodetectors based on different response mechanisms, such as the photovoltaic (PV) effect,11–14 photo-thermoelectric (PTE) effect,15–17 photogating effect18,19 and bolometric effect,20 have been investigated to develop high-performance graphene detectors. So far, high photoconductive gain and fast photoresponse have been demonstrated in graphene-based hybrid phototransistors,18,19,21 waveguide integration,22,23 2D van der Waals heterostructures24–26 and other device configurations. Although most of the research studies are devoted to the device design, test and mechanism analysis, the demand for implementation of high-resolution imaging from graphene photodetectors is gaining importance, especially in the infrared spectral region.

In this work, we demonstrate an active infrared imaging by using a multilayer graphene photodetector at room temperature. Based on hybrid response mechanisms, high-resolution optical images with close-to-theoretical-limit Michelson contrasts have been acquired without any bias from a graphene detector in the visible and short-wavelength infrared spectral range. The study, assisted with Raman measurement and numerical simulation, has revealed that the photovoltaic and photo-thermoelectric effects are the main basis for the hybrid...
photocurrents which contribute to high-quality images. The reported imaging results will pave the way for developing a high-performance and compact graphene-based infrared multispectral detector.

**Experimental**

Graphene used in this work was mechanically exfoliated from a graphite flake onto a 300 nm SiO$_2$/Si substrate. The source/drain (S/D) electrodes (15 nm Cr and 50 nm Au) were prepared by electron-beam lithography (JEOL 6510 with an NPGS System), metallization, and a lift-off process. The optical-electronic properties of the fabricated devices were characterized by scanning photocurrent microscopy (SPM). For SPM measurements, a femtosecond OPO laser (Spectra-Physics Inspire ultrafast OPO system with a tunable wavelength of 345–2500 nm) was used as the excitation source. The graphene device is mounted onto a piezo-driven sample stage, and then illuminated by a focused laser spot using a ×100/NA-0.95 objective. The laser beam is modulated at a frequency of ∼893 Hz with a mechanical chopper used as the reference to a lock-in amplifier from which the photocurrent signal can be extracted. Simultaneously, the reflection map is obtained through collecting the reflected light by using a silicon avalanche photodiode (APD). For active imaging measurements, we replace the APD with the graphene device as a point-like photodetector to collect the reflected light of the illuminated objects (STIP & FUDAN). The objects made of metal were patterned on the silicon substrate which was mounted onto the piezo-driven sample stage. The scan speed is 25.2 ms per pixel and there are 5 pixels per micron. A ×100/NA-0.95 objective was used to focus the laser spot on the patterns. Simultaneously, a ×20/NA-0.4 objective was positioned in front of the graphene photodetector for better light collection. In addition, Raman spectra were measured by using a Nanofinder 30 (TII Tokyo Instruments, Inc.) at room temperature. All spectra were recorded by using a solid state laser (λ = 532 nm) with the same laser excitation power of 0.5 mW, and calibrated by the Raman shift of the single crystal silicon at 520.4 cm$^{-1}$. The incident laser was focused on the sample with a ×100/NA-0.7 objective.

**Results and discussion**

The multilayer graphene identified by optical contrast and Raman spectroscopy, as the channel material, has a length of ∼13 μm. To investigate the photoresponse mechanism of a graphene photodetector, we characterize the device by SPM. It should be noted that all the photocurrent (PC) maps are taken at zero source–drain bias. Fig. 1a and b show an illustration of the graphene photodetector and correlated spatially resolved PC map under 1550 nm light illumination. It can be seen that, by scanning the focused laser spot over the device, four red-and-blue PC ribbons appear across the graphene channel. The colors red and blue represent the opposite polarity of PC. As a function of distance, the PC along the white arrow marked in Fig. 1b is shown in Fig. 1c. It is obvious that there are two PC maxima near the S/D contacts, while there are two PC extrema in the middle of the conductive channel. These four PC values have the same order of magnitude. And, the infrared PCs increase linearly with optical power (see Fig. 1d). As reported by many groups, the PCs occurring in close proximity to the contacts (corresponding to points A and D) are attributed to the separation of photogenerated electron–hole pairs by the built-in electric field (PV effect) or PTE effect.\(^{27,28}\) For the homogeneous graphene channel, PCs away from contacts were typically weak under zero applied bias. In the following discussion, we will focus on the positive and negative PC pairs located at the channel center (corresponding to points B and C). In addition, the fact that the $I_{ds}$ monotonously decreases with the increase of gate voltage $V_{gs}$ as shown in Fig. 1e proves that the graphene has a p-type characteristic.

In order to study the PC formation mechanism at points B and C, Raman microspectroscopy was used to characterize the properties of graphene. Fig. 2a and b present the Raman G(2D) band intensity maps and G(2D) band frequency maps. The Raman intensity maps have a contrast profile similar to that of the optical reflection morphology as shown in Fig. 1b. Except for the portion of graphene near the source electrode, which is a little thicker than the other parts, the uniform graphene channel was verified by the homogeneous G/2D band intensity Raman maps, especially for the PC region marked by a rectangle. Additionally, distinct blue shifts of the G and 2D bands can be observed in Raman frequency maps, suggesting that the marked graphene area is more heavily p-doped compared to its surroundings.\(^{29–31}\) And, this shift could be due to an unintentional doping by species absorbed from the surroundings or residual polymers used during device fabrication.\(^{31}\) Fig. 2c displays the microzone Raman spectra taken from the points labeled by circles in Fig. 2a. The basically same high intensity ratio $I_{G}/I_{2D}$ in p and p$^{+}$ regions further proves that the marked graphene region has a uniform multilayer structure.\(^{32–34}\) Therefore, the generation of positive and negative PC pairs located at points B and C could be a result of the p–p$^{+}$–p junction.

The PV or PTE effect is responsible for the PC generation at the graphene–metal contact as mentioned above. Due to the identical PC polarity and comparable PC amplitude induced by these two effects, it has been difficult to distinguish which one would be the dominant response mechanism.\(^{15,17,20}\) The same situation also arises in a graphene p–n junction. However, for the unipolar junction regimes n–n+ and p–p+, the two effects present opposite PC polarity.\(^{15,20}\) Here, unlike the experimental methods previously reported, we explore the origin of the PC response in the p–p$^{+}$–p junction by numerical simulation. For simplifying the simulation, only the PV effect is considered as the main factor responsible for PCs at the graphene–metal junction.

An electronic band configuration of the graphene device based on the results of Raman measurements has been shown.
in Fig. 2d. The model used here is based on drift–diffusion approximation, where the Poisson’s equation and continuity equations for electrons and holes are solved self-consistently by the finite element method. For simplicity, the multilayer graphene was regarded as a narrow-gap material with a bandgap of 0.2 eV and its dimension was enlarged for convergence during the simulation. The Schottky barrier at the graphene–metal junction was set to 0.2 eV. The p region doping concentration of $3.08 \times 10^{13} \text{cm}^{-2}$ can be obtained from the $I_{ds}$–$V_{gs}$ characteristic in Fig. 1e at $V_{gs} = 0$ V. However, it is hard to define the doping concentration accurately for the p+ region. Therefore, we adopt one approximate approach by using the hysteresis behavior of transfer properties in a graphene transistor resulting in a slight p-type doping to acquire the $I_{ds}$–$V_{gs}$ characteristic in Fig. 1e at $V_{gs} = 0$ V. However, it is hard to define the doping concentration accurately for the p+ region. Therefore, we adopt one approximate approach by using the hysteresis behavior of transfer properties in a graphene transistor resulting in a slight p-type doping to acquire the p+ region doping concentration of $3.8 \times 10^{13} \text{cm}^{-2}$. This approximation will be explained below in detail. For the PTE effect, a temperature difference induced by the laser spot results in a net thermoelectric voltage across the graphene channel. After a fast initial relaxation process, the photoexcited carriers form a hot Fermion distribution. Under the action of the temperature gradient, the hot carriers lead to a thermal current that corresponds to the thermoelectric current.

During the simulation, the thermodynamic model has been used to account for the PTE effect. The drift–diffusion model was extended by including the temperature gradient as a driving term. Fig. 2e and f present the simulated results of the electric field distribution along the conductive channel and corresponding generated PC induced by the PV effect ($I_{PV}$). Obviously, the directions of $I_{PV}$ at the p–p+p–p junction are opposite to that of the PC obtained from experiment ($I_{Exp}$). So, the PV effect is not the main reason for the generation of $I_{Exp}$ at the p–p+p–p junction. Fig. 2g shows the simulated distribution of the generated PC induced by the PTE effect ($I_{PTE}$). It can be seen that the direction of $I_{PTE}$ is consistent with that of $I_{Exp}$. Moreover, Fig. S1† shows the simulated $I_{PTE}$ for the temperature difference ranging from 2 to 6 K. A good linear dependence of $\Delta T$ was obtained. Therefore, the PTE mechanism should dominate the PC generation at the p–p+p–p junction in comparison with the PV effect.

To understand how the strength of the PC changed with $V_{gs}$ modulation, the photoresponse properties of this graphene device under different back-gate voltages were investigated by using SPM. Fig. S2† presents the PC maps of another device in our work. It appears that the 565 nm map gives more PC specifics than the 1550 nm map due to the smaller diffraction-limited light spot. So, as shown in Fig. 3a, 565 nm light was used to obtain relatively high-resolution PC maps at $V_{gs}$ of −60, 0 and 60 V. As a result, the PC distribution and

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**Fig. 1** Photoresponse measurement of the graphene photodetector. (a) A schematic illustration of the graphene photodetector. (b) Optical reflection map taken under 633 nm laser illumination and the scanning photocurrent map taken under 1550 nm laser illumination with a light power of 22.5 μW. Scale bar, 3 μm. (c) Photocurrent line traces along the white arrow in b. The shaded area indicates the contour of the source/drain electrode. (d) Photocurrents vs. incident light power taken at the points A, B, C, and D in b. (e) $I_{ds}$–$V_{gs}$ characteristic of the graphene transistor acquired at $V_{ds} = 0.1$ V.
morphology are basically the same as that shown in Fig. 1b. 
$I_{PV}$ and $I_{PTE}$ taken at points A, B, C and D with the change of $V_{gs}$ have been presented in Fig. 3b and c, respectively. For $I_{PV}$, the current amplitude decreases with an increase of $V_{gs}$. This is because the Fermi level gradually reaches the Dirac point to weaken the strength of the built-in electric field at the graphene–metal junction (see the inset of Fig. 3b). For $I_{PTE}$ at points B and C, however, there is a maximum value with increasing $V_{gs}$. Usually, the direction and magnitude of $I_{PTE}$ depend on the thermoelectric power (Seebeck coefficient) in each part of the graphene channel and can be expressed as:

$$I_{PTE} = \frac{S_p - S_{p+}}{R} \Delta T$$

where $S$ is the Seebeck coefficient in the p and p+ regions, and $\Delta T$ is the temperature difference resulting from laser illumination. From the Mott relation, the Seebeck coefficient can be written as:

$$S = \frac{\pi^2 k_b^2 T^2}{3e} \frac{dR}{dV_g} \frac{dE}{dE} \bigg|_{E=E_F}$$

where $k_b$ is the Boltzmann constant, $e$ is the electron charge, $T$ is the sample temperature, $R$ is the resistance, $dR/dV_g$ can be derived from the transport characteristic measurements, and $E_F$ is the Fermi energy. $E_d(n)$ dependence of multilayer graphene in this work is approximated to be that of bilayer graphene, which is formulated as:

$$E_d = \frac{1}{2} \sqrt{(2\hbar\nu_F)^2 \pi n + 2\gamma_1 \nu_F^2 - 2\sqrt{(2\hbar\nu_F)^2 \pi n + \gamma_1^2} \nu_F}$$

where $\gamma_1$ is the interlayer coupling strength and set to be 0.4 eV, $\hbar$ is Planck’s constant, and the Fermi velocity $\nu_F = 10^6$ m s$^{-1}$. To explain the phenomenon shown in Fig. 3c, eqn (1)–(3) should be solved together. However, it is hard to extract $S_p$ and $S_{p+}$ accurately for the p and p+ regions in the same conductive channel. Therefore, one approximate approach was adopted to obtain p- and p+-type graphene on the same device, in which the sweeping direction of the gate bias was changed from forward to backward to form a slight hysteresis in transfer properties. Hysteresis behavior in a graphene transistor has been explored by many groups. Charge injection into the trap sites on a dielectric substrate and charge transfer from/to neighboring adsorbates have been considered as the main reasons for graphene doping to result in hysteresis. Fig. 3d shows the transfer curve of another device in our work. The black and red dotted lines reveal the p- and p+-type graphene characteristics, respectively. Here, the transport characteristic in the $V_{gs}$...
range of −50 to 30 V, marked with a shaded area, was chosen to approximately analyze the $I_{PTE}$ change with $V_{gs}$ because this portion of the line shape is similar to that in Fig. 1e. Fig. 3e presents the calculated $S_p$ and $S_{p+}$ (the inset) and the difference ($S_p - S_{p+}$). It is obvious that, compared with $I_{PTE}$ in Fig. 3c, the difference in the Seebeck coefficient has a consistent variation tendency with $V_{gs}$ modulation. It should be noted that only the amplitude of $I_{PTE}$ changes with increasing $V_{gs}$. No polarity reversal was observed in Fig. 3a, which is opposite to the calculated results (unshaded area in the inset of Fig. 3e) predicting a pronounced change in the $I_{PTE}$ sign, i.e. the sign of ($S_p - S_{p+}$), at high $V_{gs}$. This may be due to the reversal in the $I_{PTE}$ sign that happened in the process of $V_{gs}$ approaching the Dirac point as shown in the inset of Fig. 3e. However, in the case of Fig. 3a, the graphene is more p-doped than that in Fig. 3d, and $V_{gs}$ is not high enough to tune the Fermi level efficiently.

To investigate the imaging ability of the graphene photodetector, a confocal laser scanning microscope setup has been refitted. The graphene device is configured as a point-like detector to collect the reflected light (see Fig. 4a). Through the current pre-amplifier and lock-in amplifier, signal PCs are extracted from the graphene device to form images. For achieving the best performance, a ×100 objective lens having a numerical aperture of 0.95 is used to obtain a close-to-diffraction-limited light spot on the objects. And, the graphene detec-
tor is then slightly moved in three dimensional directions near the focus of the objective lens 2 (×20/NA-0.4) to achieve maximum signal currents. These currents consist of $I_{\text{PTE}}$ and $I_{\text{PV}}$ originating from the hybrid response mechanisms, i.e. the synergistic action of PTE and PV effects. Fig. 4b gives a scanning electron microscopy image of metallic objects (SITP) with a line width of 2 μm patterned on a silicon substrate. The corresponding images are acquired with the graphene detector by applying the focused laser spot (565 nm, 1550 nm and 1815 nm) scanning across the patterns (see Fig. 4c–e). Simultaneously, the signal PCs from a pattern edge cross section in the images have been extracted (see Fig. 4f). It can be seen that, though these images are all well-resolved, the resolution of the 565 nm image is still somewhat higher than that of the other two images (1550 nm and 1815 nm) which have a similar profile. This is because, for the active infrared imaging system in our work, the main factor influencing the resolution depends on the ability of the objective lens 1. When a short wavelength light passes through an objective, a small diffraction-limited light spot will be focused onto the objects, resulting in a relatively clear image outline compared with that using long wavelength light. In order to obtain the image resolution, the model of point spread function based on the convolution of a step function and a Gaussian function has been used to fit the data in Fig. 4f. As shown in Fig. S3, the optical resolutions of 418 nm, 657 nm and 877 nm are extracted from the fitting functions for the 565 nm, 1550 nm and 1815 nm light imaging measurements, respectively. Additionally, Fig. S4 presents the imaging results of metallic objects with a line width of 5 μm under 1550 nm illumination. The basically same optical resolution of 676 nm is obtained. The results demonstrate that a graphene photodetector can be effectively used in broadband high-resolution imaging applications.

Furthermore, to characterize the image quality, the Michelson contrast $C$ is calculated with the formula

$$C = \frac{L_{\text{max}} - L_{\text{min}}}{L_{\text{max}} + L_{\text{min}}}$$

Fig. 4  Active imaging by using a graphene photodetector. (a) Schematic of the active imaging setup. (b) Scanning electron microscopy image of metallic objects (SITP) with a line width of 2 μm patterned on a silicon substrate. Scale bar, 4 μm. (c–e) Images acquired from the graphene photodetector at λ of 565 nm, 1550 nm and 1815 nm. (f) The signal PCs taken from a pattern edge cross section in the images. The shaded area indicates the contour of the pattern as shown in the inset.
where $L_{\text{max}}$ ($L_{\text{min}}$) is the PC maximum (minimum) of the images. As a result, ultrahigh contrasts of 0.997, 0.994 and 0.996 are obtained for 565 nm, 1550 nm and 1815 nm images, respectively. The graphene photodetector shows good performance for imaging with a close-to-theoretical-limit contrast in the visible and short-wavelength infrared spectral range. Fig. S5† gives the 1550 nm imaging results for the “FUDAN” patterns. The same clear and high-resolution image was also obtained.

Conclusions

We have demonstrated an active infrared imaging by using multilayer graphene photodetectors based on hybrid response mechanisms at room temperature. The high optical resolutions of 418 nm, 657 nm and 877 nm and close-to-theoretical-limit Michelson contrasts of 0.997, 0.994, and 0.996 have been acquired from the graphene detector in the visible (565 nm) and short-wavelength infrared (1550 nm and 1815 nm) spectral range, respectively. Detailed studies based on the Raman measurements and numerical simulation of the response process reveal that the formation of hybrid photocurrents in the multilayer graphene detector is attributed to the synergistic action of photovoltaic and photo-thermoelectric effects. The initial implementation of imaging will help promote the development of high performance graphene-based infrared multispectral detectors.

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Notes and references

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