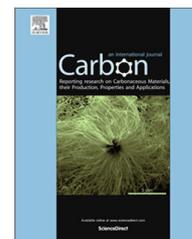


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A self-powered graphene–MoS₂ hybrid phototransistor with fast response rate and high on–off ratio



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ABSTRACT

Here, we present a self-powered graphene–MoS₂ hybrid phototransistor that simultaneously exhibits high responsivity, fast response rate and high on–off ratio. In this phototransistor, fewer-layer MoS₂ serves as a sensitizer to absorb light, and high mobility graphene serves as an expressway for carrier transport, an asymmetric metal contact (titanium and palladium) design is adopted to provide a strong internal electric-field in the phototransistor channel. Under light illumination, a photovoltage arises at the two asymmetric graphene/electrode interface with photodiode mechanism, which act as a bias voltage driving the large quantity of photocarriers injected from MoS₂ layer to transport in graphene channel with photoconductor mechanism. With this ingenious design, the hybrid phototransistor achieves a response rate of 0.13 ms and on–off ratio up to 1428 together with responsivity exceeding 3.0 A/W under zero bias operation. The hybrid phototransistor takes full the advantage of fast response rate of the photodiode and high responsivity of the photoconductor, which makes it has great potential for wide applications in optoelectronics, such as optical logic, optical computing and optical communication.

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1. Introduction

The high responsivity phototransistor with large on–off ratio and fast response rate is highly desirable for various demanding applications, such as optical communication, imaging, and sensing [1]. Recently, phototransistors based on silicon semiconductors, organic semiconductors, quantum dots and carbon nanotubes have been extensively studied [2–5]. The two main types of phototransistors are photodiodes and

photoconductors [4]. Photodiodes respond fast but have low gains that are usually less than 1. Photoconductors, in contrast, are capable of high gain because one type of charge carrier is able to circulate through an external circuit many times before it recombines with its opposite carrier, and thus responses slowly. Moreover, the materials with suitable band gap and high mobility are favorable for high responsivity photoconductors, in which more photocarriers can be excited, and it can recirculate more times through an external

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circuit. The types of phototransistor are usually determined by the materials which are used in the transistor channel.

Two-dimensional (2D) atomic crystal, such as graphene, MoS₂, black phosphorus, etc., has attracted great attention in recent years because of their superior electrical and optical properties [6–13]. The 2D phototransistor represented by graphene and MoS₂ phototransistors are becoming the focus of the research topic, and are expected to be used in next generation ultrathin and flexible optoelectronic devices [14–20]. Graphene phototransistors, which work based on the photodiode mechanism, have been used as ultra-fast photodetectors for high-speed optical communications because of their ultrafast carrier dynamics [8,21]. However, the weak optical absorption of graphene (~2.3%) due to the zero band gap limits its photoresponsivity (<6.1 mA/W) [8]. In addition, the zero band gap and the extremely short exciton life-time (in the ps range [22]) of graphene make its phototransistor cannot work with the photoconductor mechanism. At the same time, MoS₂ phototransistors, which work based on the photoconductor mechanism, have also been widely studied for their great potential in visible light photodetectors [23–25]. The thickness-dependent band gap of MoS₂ makes its phototransistor show selective photodetection capabilities to red and green light with triple, double and single layer thickness [25]. However, the responsivity of MoS₂ phototransistor is still low due to its low carrier mobility [26,27]. Therefore, for graphene and MoS₂, each material has its spotlight merit, but also has fatal weakness, for example, graphene has high carrier mobility but zero band gap, MoS₂ has large band gap but low carrier mobility, which limit their potential application in optoelectronics.

Until recently, our previous work and several other group's work have addressed this issue by integrating graphene and MoS₂ into one device to complement their merit and the weakness [26–28]. In fact, all these work are based on the idea of implementation of photoconductive gain – the ability to generate multiple charge carriers by absorption of single incident photon, which was considered as the key to ultrasensitive graphene-based photodetection [29]. In the previous work, we constructed a graphene–MoS₂ hybrid phototransistor, in which MoS₂ acts as a sensitizer to absorb light and graphene acts as an expressway for carrier transport. The photocarriers generated in MoS₂ layer are transferred to graphene layer derived by a built-in electrical field at graphene–MoS₂ heterojunction, where they recirculate many times due to the high carrier mobility of graphene. In this case, strong light absorption and fast charge-transport processes are decoupled in one device with two materials, which make the responsivity of this hybrid phototransistor (up to 10⁸ A/W) is much larger than that of individual graphene or MoS₂ based phototransistor [26–28].

Though, the graphene–MoS₂ hybrid phototransistor achieves a extremely high responsivity, graphene's high carrier concentration nature inevitably results in the large dark current (up to 50–100 μ A level) and low on–off ratio (less than 2) in this photoconductor based phototransistor [26,27]. In addition, the response rate of the hybrid phototransistors is much slow (exceeding 3 s), which is the typical feature of the photoconductors [26]. Of course, it is the reality that the

responsivity and response rate is contradictory in the photoconductor based phototransistor, the higher the responsivity the slower the response rate [4]. Anyway, these serious problems of the graphene–MoS₂ hybrid phototransistor hinder its potential applications in future optoelectronics, such as optical logic, optical computing and optical communication.

In this work, we demonstrate a new type graphene–MoS₂ hybrid phototransistor (Fig. 1a) that simultaneously exhibits high speed (0.13 ms), high on–off ratio (up to 1428) and high responsivity (exceeding 3.0 A/W) under the zero-bias operation. In this new type hybrid phototransistor, an asymmetric metal contact design (titanium (Ti) and palladium (Pd)) is designed used to break the mirror symmetry of the internal (built-in) electric-field profile in the traditional phototransistor channel, and thus provides a strong internal electric-field in the new hybrid phototransistor channel. In this case, a photovoltage arises in the phototransistor channel under the light illumination at the two asymmetric graphene/electrode interfaces, which acts as a bias-voltage driving the large quantity of photo-carriers injected from MoS₂ layer to transport in the graphene channel. It is the photovoltage that make the phototransistor operated self-powered operation, and have very small dark current and large on–off ratio. In addition, the response rate of the hybrid phototransistor is very fast, because it is determined with the response rate of the photovoltage in the channel. With this ingenious design, the new type hybrid phototransistor takes full the advantage of high response speed of the photodiode and high responsivity of the photoconductor. This masterly design and superior properties make this new type hybrid phototransistor a great promise for future applications in optoelectronic functional devices.

2. Experiment section

2.1. Materials preparation

Few-layer MoS₂ flake was deposited on the Si/SiO₂ (300 nm) substrate by using scotch-tape based mechanical exfoliation of bulk MoS₂ (purchased from smart-elements), its optical microscopy (OM) image and Raman spectroscopy are shown in Fig. S1. Single layer graphene was produced by using the chemical vapor deposition (CVD) method using Cu foils (Alpha aser) as catalytic substrates. First, the Cu foil was located at the center position of the CVD furnace and was heated up to 1000 °C under a 10 sccm H₂ flow at 50 mTorr. The chamber was maintained under the same flow conditions for 30 min at 1000 °C to anneal the Cu foil. After 30 min at 1000 °C, the mixture of 2 sccm CH₄ and 10 sccm H₂ gases was flowed for another 30 min at 400 mTorr. The sample was then cooled rapidly to room temperature with flowing 10 sccm H₂ at the pressure below 50 mTorr. After the growth of graphene on Cu foils, polymethylmetacrylate (PMMA) were coated onto the graphene film grown Cu foil by spin coating. The backside graphene film was removed by reactive ion etching with O₂ plasma. After etching the Cu layer with wet etchants (saturated solution of FeCl₃), the remaining graphene films on the polymer support were

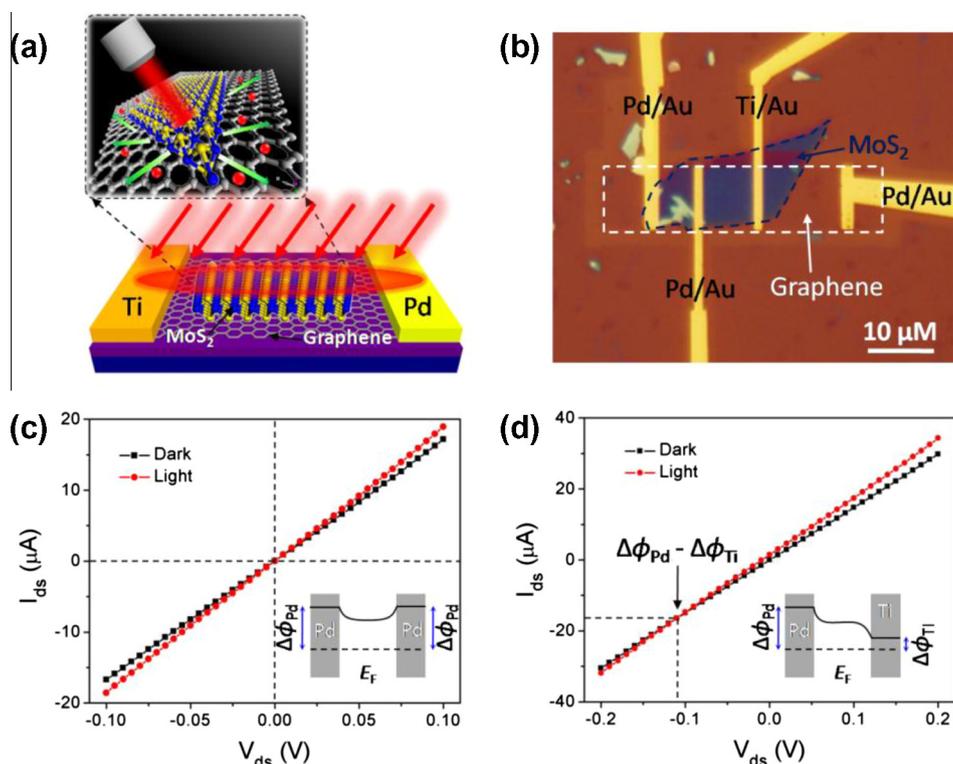


Fig. 1 – (a) Schematic diagram of a self-powered graphene–MoS₂ hybrid phototransistor with asymmetry metal contact under light irradiation. (b) Optical image of the graphene–MoS₂ hybrid phototransistor. Source–drain bias voltage dependence of current in the hybrid phototransistor with (c) symmetry and (d) asymmetry metal contact under dark and light illumination, at an excitation wavelength of 632.8 nm (incident power = 0.82 μW). Δφ_{Pd} and Δφ_{Ti} represent the difference between the Dirac point energy and the Fermi level in palladium- and titanium-doped graphenes, respectively. (A colour version of this figure can be viewed online.)

washed with deionized water and transferred onto the target substrate.

2.2. Device fabrication

First, the CVD grown graphene was transferred onto the MoS₂ flake which has been deposited on Si/SiO₂ substrate, and then annealed at 380 °C under the protection of H₂ (300 sccm) and Ar (300 sccm). The single-layer graphene was used in our work, which is distinguished by using both (OM) image and Raman spectroscopy as shown in Fig. S2. After that, a three-step electron beam lithography (EBL) with the metal evaporation and lift-off techniques were used to construct the hybrid device. The individual graphene–MoS₂ stack layer separated from the large area CVD graphene layer was obtained by using the first EBL step followed with O₂ plasma lithography treatment; The two asymmetry contact electrodes Ti/Au (10 nm/50 nm) and Pd/Au (10 nm/50 nm) were fabricated using the second and the third EBL steps, respectively.

2.3. Photoelectric measurement

The photocurrent measurement was performed on a homemade photoelectric measurement platform which are composed of Horiba HR800 Raman spectrometer and

Keithley 4200 semiconductor analyzer. The 632.8 nm laser beam was used as the incident light, the spot size of the laser was adjust to about 30 μm to ensure the phototransistor channel was completely covered.

3. Results and discussion

Fig. 1b presents an optical image of one graphene–MoS₂ hybrid phototransistor, the corresponding SEM image is shown in Fig. S3. The device contains two types of phototransistors, one is formed with symmetry metal contact electrodes (the left two Pd/Au electrode), the other is formed with asymmetry metal contact electrodes (the middle Ti/Au and Pd/Au electrode). All these electrodes are directly contacted to the graphene surface, which can induce doping to graphene at their interface. It has been shown that a weak internal (built-in) electric field generated at the graphene/electrode interface allows high-speed and efficient photocarrier separation. Fig. 1c presents the drain–source bias (V_{ds}) dependence of the current (I_{ds}) in the hybrid phototransistor with symmetry metal contact electrodes. The black and red curves show the dark current (I_{dark}) and the total current I ($I = I_{dark} + I_{light}$) under light illumination, respectively. It can be seen that the two curves always cross at $V_{ds} = 0$ mV, indicating that the built-in electric field in the channel between two symmetric contact is zero, which can

be easily understood from its band profiles (the insert in Fig. 1c). In contrast, that exhibits great difference in the graphene–MoS₂ hybrid phototransistor with asymmetry metal contact electrodes as shown in Fig. 1d. The two curves cross at $V_{ds} = -110$ mV, which means the photocurrent turns to zero at this bias voltage. This result exactly corresponds with our design and can be easily understood from the band profiles of the device (the insert in Fig. 1d). It means that at $V_{ds} = -110$ mV, the external bias just offsets the built-in potential, making the band profile flat and the photocurrent become zero. For this device, the height of the potential within the graphene channel induced by the doping of Ti and Pd is ~ 110 meV, which can be estimated from the above experiment result. Therefore, the large work function difference between Ti and Pd electrodes brings a large built-in potential profile in the hybrid phototransistor with asymmetry metal contacts.

Fig. 2a shows the drain–source voltage (V_{ds}) dependence of the phototransistor current under light illumination with different power. The photocurrent increases with the optical power increasing, which is consistent with previous reported work [26]. However, the obvious difference is a photovoltage generated in the phototransistor. The black arrows indicate the photovoltage of the device under different optical power illumination, which were achieved from the V_{ds} at $I_{ds} = 0$ A. The photovoltage arises from the photocurrent at the two asymmetry graphene/electrode contacts with a photodiode mechanism. Thus, the photovoltage can be simply given by the Ohm's law: $V_{ph} = I_{ph} * R_g$, where R_g is the resistance of graphene. It can be seen that the photovoltage in the phototransistor channel decreases as the optical power decreases (Fig. 2b), a 12 mV photovoltage can be obtained under $8.2 \mu\text{W}$ light illumination, and it decrease to 0.2 mV as the optical power decrease to $0.0082 \mu\text{W}$. This is reasonable because the photocurrent generated from the two asymmetry graphene/electrode decreases as the optical power decreases. In fact, there is also photovoltage arising at each graphene/electrode interface in the phototransistor with symmetry metal contact, however, the opposite direction of them leads the total photovoltage in the phototransistor channel to be zero. Therefore, the large internal electrical

field due to asymmetry band bending allows photocarrier separation at graphene/electrode interface to arise a photocurrent, which induces a photovoltage in the phototransistor channel.

Based on our design, the photovoltage generated at the graphene/electrode interface is expected to replace the external bias voltage which is needed in traditional phototransistor work with photoconductor mechanism, driving the entire graphene–MoS₂ phototransistor to work with photoconductor mechanism as shown in Fig. 3a and b. Fig. 3c shows the optical power dependence of photoresponse of the graphene–MoS₂ phototransistor. It can be seen that the photocurrent monotonically decreases as the optical power decreases due to the decline of excitation efficiency. It needs to be noted that the photocurrent exhibits a non-linear decrease with the optical power decreasing (Fig. 3d), it decreases dramatically in the lower optical power range. Moreover, the responsivity also shows a non-linear increase as the optical power decreases (Fig. 3d), which is different from that in the traditional phototransistor where it shows a linear relationship. This phenomenon is attributed to the photoresponse mechanism in our hybrid phototransistor. The photocurrent (PC) of our graphene–MoS₂ phototransistor under light irradiation is given by:

$$PC = \frac{WC_i\mu\Delta V_G V_{PV}}{L} \quad (1)$$

where C_i is the capacitance of the gate dielectric per unit area, μ is the carrier mobility of graphene, V_{ph} is the photovoltage in the channel, W and L are the width and the length of the channel, respectively. The ΔV_G can be considered as the photogate, which is proportional to the quantum efficiency of the device [27]. So the responsivity (R) of the device can be calculated by:

$$R = \frac{PC}{WLE} = \frac{C_i\mu\Delta V_G V_{PV}}{L^2 E} \quad (2)$$

In our hybrid phototransistor, the quantity of the photocarriers in the channel contributed from MoS₂ layer is decreases as the optical power decreasing. In addition, the photovoltage arising from the asymmetry metal contact, which serves as the bias voltage driving the photocarriers to

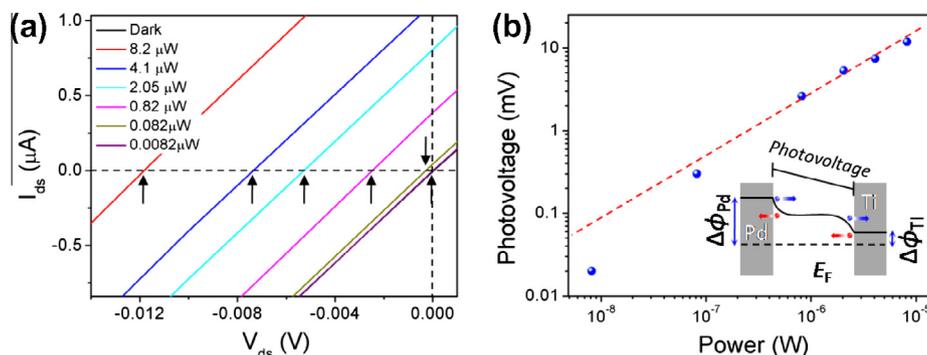


Fig. 2 – (a) Photoresponse as a function of drain–source voltage (V_{ds}) of the graphene–MoS₂ hybrid phototransistor with asymmetry metal contact under different optical power illumination, the whole curves in the range from -0.2 to 0.2 V are shown in Fig. S4. The arrows indicate the position of photovoltage at each optical power irradiation. (b) The photovoltage in the phototransistor channel as a function of optical power. The insert is the schematic diagram of the arising process of the photovoltage. (A colour version of this figure can be viewed online.)

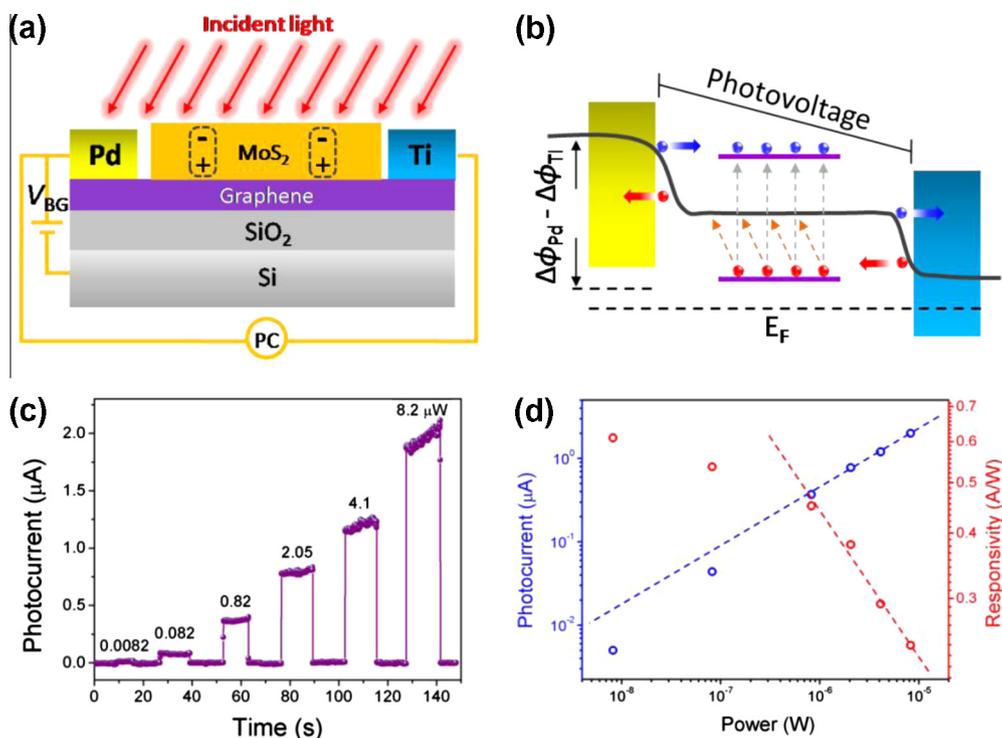


Fig. 3 – Schematic diagram of (a) the charge generation and separation process and (b) the corresponding energy diagram of the graphene–MoS₂ hybrid phototransistor with asymmetry metal contact under light irradiation. (c) Photoresponse of the graphene–MoS₂ hybrid phototransistor as a function of time under different optical power illumination at zero bias voltage. (d) The photocurrent and the responsivity of the phototransistor as a function of optical power. (A colour version of this figure can be viewed online.)

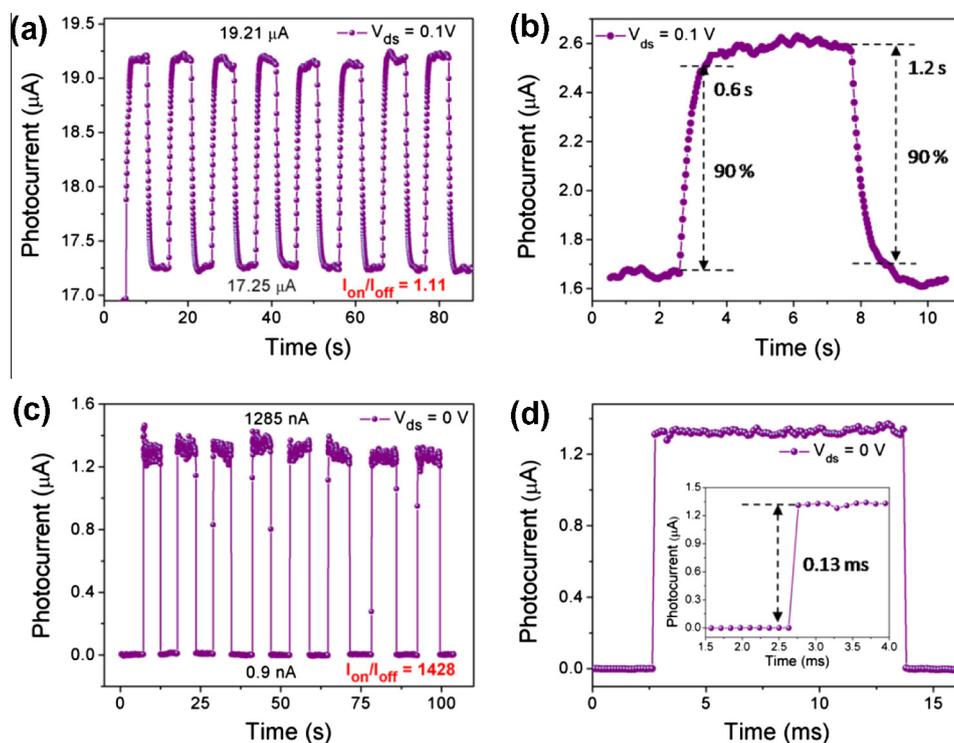


Fig. 4 – Photocurrent response of graphene–MoS₂ phototransistor with (a and b) symmetry metal contact and (c and d) asymmetry metal contact. (b) and (d) are the corresponding enlarged figures of (a) and (c). The irradiation light at wavelength of 632.8 nm, optical power = 0.82 μW. The phototransistors with symmetry and asymmetry metal contact are operated at 0.1 V and 0 V bias voltage, respectively. (A colour version of this figure can be viewed online.)

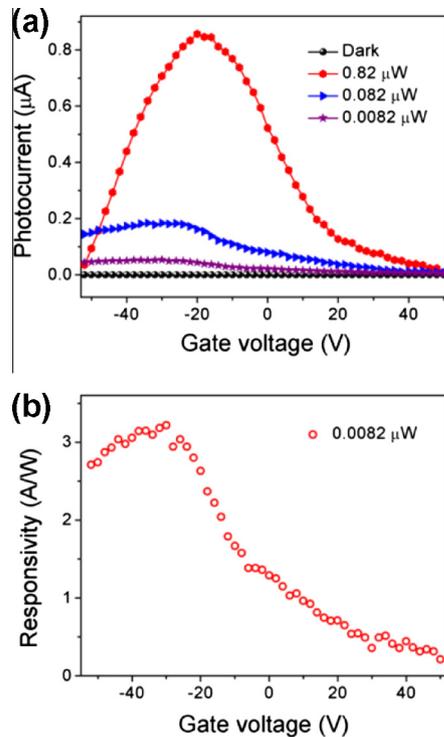


Fig. 5 – (a) Photocurrent as a function of back-gate voltage for the self-powered graphene–MoS₂ transistor with different irradiation power. (b) Responsivity as a function of back-gate voltage with 0.0082 μW light irradiation at $V_{ds} = 0$ V, showing that the responsivity of the phototransistor can be directly tuned with the applied back-gate voltage. (A colour version of this figure can be viewed online.)

transport in the channel, is also decreasing as the optical power decreases. So, from these two equations we can see that the photocurrent and responsivity are proportional to both ΔV_G and V_{PV} which are proportional to the optical power, thus they show a non-linear variation with the optical power. In spite of the drastically decreasing of the photocurrent with the optical power decreases, the responsivity as high as 0.6 A/W (photocurrent 5 nA and on-off ratio 10) can be obtained even at a very weak optical power (0.0082 μW). In fact, this value is relative smaller than that of the traditional graphene–MoS₂ hybrid phototransistor [26,27]. However, the smaller sacrifice in the responsivity is worthwhile as illustrated below.

For the traditional graphene–MoS₂ hybrid phototransistor with symmetry metal contact, where an external bias voltage (here $V_{ds} = 0.1$ V) is needed for the device operation with a photoconductor mechanism. Fig. 4a shows the photoresponse of the phototransistor as a function of time. It can be seen that a large dark current up to 17.25 μA and a small on-off ratio less than 2 were obtained due to the nature of graphene's high carrier concentration. In addition, the phototransistor exhibits a extremely slow photoresponse rate (rise time of ~0.6 s and fall time of ~1.2 s) as shown in Fig. 4b. The slow response rate, large dark current and small on-off ratio are the typical features of the graphene–MoS₂ hybrid phototransistor with symmetry metal contact,[26] which

make the device be not benefit to be used in fast optical communication, imaging and optical logic, etc. In contrast, for graphene–MoS₂ hybrid phototransistor with asymmetry metal contact, the photovoltage serves as a bias voltage (replace the external bias voltage) driving the device operation. It can be seen that this phototransistor has a very small dark current (0.9 nA) and a large on-off ratio (1428) as shown in Fig. 4c. This is because that the photovoltage is zero under the dark state, which is different from the traditional phototransistor where an external bias voltage is always applied. At the same time, the on-off photo-switching of this phototransistor is quite fast (0.13 ms) as shown in Fig. 4d. This can be explained as the fast on-off rate of the photovoltage in the channel, which raises at the two asymmetry metal contact with photodiode mechanism served as a bias voltage. Therefore, for the hybrid phototransistor with asymmetry metal contact, the smaller sacrifice in the responsivity can retrieve the fast response rate and large on-off ratio with zero bias operation, which is useful for the application of some special electronic devices where not much high responsivity but high speed and large on-off ratio is needed.

Fig. 5a shows the gate voltage dependent photoresponse of our graphene–MoS₂ hybrid phototransistor with dark and different optical power illumination under zero bias voltage operation. It can be seen that the device reveals an obvious gate tunability of the photocurrent response under light illumination, while the current is always zero in the whole gate voltage range under dark condition. The responsivity can up to 3 A/W at $V_{BG} = -30$ V as shown in Fig. 5b, where the position of graphene Fermi level is at the optimum location for the photo-excited holes to inject from MoS₂ layer to graphene layer. By tuning the Fermi level close to the Dirac point at $V_{BG} = 50$ V, the responsivity falls near to zero, where the photo-excited holes are just completely recombined by the electrons induced from back gate voltage. Moreover, by tuning the Fermi level far away from the Dirac point ($V_{BG} > -30$ V), the responsivity also declines, where the Fermi level of graphene is too low to match with the energy level of MoS₂, and thus the photo-excited holes transfer is forbidden. This feature demonstrates the potential of this device as a gate-tunable phototransistor. This tunability is of great importance in phototransistor because it allows adjustment of the required state (on-off) as well as gain of the device, depending on the light power detected.

4. Conclusion

In summary, based on the idea of takes full the advantage of fast response rate of the photodiode and high responsivity of the photoconductor, we present a self-powered graphene–MoS₂ hybrid phototransistor with an asymmetry metal contact. Through this ingenious design, a photovoltage in the channel, which generates at the asymmetry graphene/electrode interface with the photodiode mechanism, drives the hybrid phototransistor operating with photoconductor mechanism. The photovoltage serves as an bias voltage, which has a fast switch effect to light on-off, making the hybrid phototransistor exhibit fast response rate (0.13 ms), small dark current (less than 1 nA) and high on-off ratio (up

to 1428) at a zero external bias voltage (self-powered operation). At the same time, through combining the virtues of strong light absorption of MoS₂ which plays a role of sensitizer to absorb light and the high mobility of graphene which can be used as expressway for photocarrier transport, the high responsivity can be guaranteed in this hybrid phototransistor. This study opens new avenues for developing 2D nano-material-based optoelectronics for imaging techniques and optical communications as well as future memory storage and optoelectronic circuits.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.carbon.2015.03.064>.

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