Near-Infrared Heterojunction Field Modulated Phototransistors with Distinct Photodetection/Photostorage Switching Feature for Artificial Visuals

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Abstract

With the rising demand of recording, computing and image capture, advanced optoelectronic detection, storage and logic devices are highly pursued. Nevertheless, multi-functional vision chip based on infrared detection and memory switching has never been demonstrated. Here, by utilizing the electronic extraction layer ZnO and face-on orientation of D-A, we exhibit the broadband visible to near-infrared photo-response and photo-storage characters on graphene phototransistor. Functions as photodetection and photo-storage can be switched with the variation of gate voltage. The device demonstrates high photo-responsivity up to $1.88 \times 10^6$ A/W at 895 nm corresponding detectivity of $4.8 \times 10^{12}$ Jones. Importantly, the rewritable and switching infrared optoelectronic memory function can be achieved with good retention over $10^4$ s. The both retinomorphic vision and memorial preprocessing in artificial visual are simultaneously realized by photodetection/photostorage switching property. Such nearly all-solution processes in our phototransistors may open up the path for the large-scale and easy manufacturing infrared multifunctional bio-optoelectronic device.

Main text

The exotic optoelectronic and quantum properties, such as ultrahigh mobility, tunable Fermi level, superconductivity feature and modified electronic spectra, are explored in graphene system. To probe all these phenomena, graphene field-effect transistor framework offers the vital platform for devising multi-functional optoelectronic devices. For the limitation of broadband absorption in monolayer graphene (2.3%), researches initially integrate graphene with single component semiconductor for high performance phototransistors utilizing photo-gating effect. Nevertheless, most of photoconductive gating devices suffer low operation time (typically millisecond) due to the prolonged lifetime of carriers. In addition, thick light-harvesting layer may promote the absorption proportion but suffer inferior transfer efficiency due to restriction of nanometer range in effective exciton length. To settle this trade-off relations, ultra-thin light-harvesting layer and valid intrinsic dissociation electric field are needed. Therefore, the ultra-thin type-II Donor-Acceptor (D-A) heterojunctions, such as PCBM/perovskite, PTCDA/pentacene and C$_{60}$/pentacene, are proposed onto graphene for improving the quantum efficiency (QE) up to around 10% compared with the single component enhanced devices. Since the absent of blocking layer may cause recombination energy losses in graphene/Acceptor interface and molecular packing orientation for efficient charge transfer, suggesting that there is still space to improve.

As we enter the areas like big data and cloud computing, more compatibility and complexity memory cells are urgently needed to exploit. With the emerge of technique in 2D materials, the high performance three-terminal semi-floating gate, floating gate and two-terminal ferroelectric tunnel junction memory structures are manufactured without dangling bonds. And optoelectronic memory promotes the data transport without limitation of short interconnections. Nevertheless, infrared optoelectronic memory has acquired far less attention, which is vital for image capture and optical communication. Infrared optical communication requires fast optoelectronic conversion-relaxation, photo-storage and logical operation. Especially, the
multi-functional high performance optoelectronic devices are highly pursued, where the image sensing and processing functions in separated vision chips can integrate together for promoting the efficiency of vision system.

Here, by utilizing electronic extraction layer ZnO and face-on orientation of D-A molecular packing into D-A/graphene system, we explore the graphene/ZnO/PTB7-Th:IEICO-4F bulk heterojunction (BHJ) field-effect transistor with prominent infrared broadband photo-response (488-1064 nm) and photo-storage phenomena. We can operate both photo-response and photo-storage in one device simultaneously. And four optoelectronic states can be switched by gate voltage. The electronic extraction and deep hole trapping state provided by ZnO, remarkably promote the speed of photo-generated electrons into channel with fastest 178 μs, while the ZnO layer prevents the holes from graphene injection into D-A hybrid with recombination energy losses, resulting in high EQE up to $2.6 \times 10^5$ %. Rely on the conspicuous infrared absorption of PTB7-Th:IEICO-4F heterojunction and novel structure, we perform a high detectivity of $4.8 \times 10^{12}$ Jones at 895 nm and photo-responsivity up to $1.88 \times 10^6$ A/W. Furthermore, we find that deep P-type graphene controlled by gate (operated at -30 V) with appropriate input light power can achieve good retention (over $10^4$ s) infrared photo-storage. The gate-tunable rewritable and infrared optoelectronic switching features suggest that phototransistor can be applied to retinomorphic vision and memorial preprocessing in artificial visual. The infrared responsivity and memory dual functional device paves the new path for fabricating non-floating gate advanced neural network phototransistor in future.

**Results and discussions**

The CVD mono-layer graphene was transferred onto the SiO$_2$/Si++ substrate by solution process with prepared bottom gate. After the Au electrode onto the graphene, ZnO and BHJ layer was formed by solution spin-coating method with the thickness of 15 nm and 35 nm (Fig. 1a), respectively. ZnO nanoparticle was used as electronic extraction layer for its high electron mobility, matched energy levels with graphene and remarkable environmental stability (Fig. S1). PTB7-Th: IEICO-4F was utilized as photosensitive layer, which is due to the broad response wave band (488-1064 nm) as shown in Fig. S2a. After spin coating, each component is confirmed by Raman spectroscopy (Fig. S2b and more peaks characterization in Fig. S3). The D-A hybrid exhibited bi-continuous interpenetrating network structure with appropriate phase separation size (Fig. S2c), which is beneficial for exciton dissociation and charge transport. Meanwhile, the unique BHJ morphology allows the phototransistor with thicker photosensitive layer than traditional plane structure, and then guarantee a high response current. AFM image of the BHJ layer showed smooth film surface with a small root-mean-square (RMS) roughness of 2.65 nm (Fig. S2d), implied satisfactory interfacial contact between BHJ and ZnO layers. Moreover, demonstrated via Grazing-incidence wide-angle X-ray scattering (GIWAXS) measurement in Fig. S2e, the combination of PTB7-Th and IEICO-4F displayed ordered and compact molecular packing in face-on orientation, which was conductive to achieve efficient charge transport in vertical direction.

Next, we further perform the intrinsic optical responsivity of hybrid phototransistor. Interestingly, we observe the four different optoelectronic switching states with the variation gate (Fig. 1b and Fig. S2f). When the applied gate voltage is about -20 V, photo-memory behavior predominates in our device (Fig. 1c). As the increasing of gate voltage, device switched from photo-
memory state to no-memory response state and off-state (Fig. 1d to f).

The gate-control graphene device shows an asymmetry bi-polar photocurrents with different input power in Fig. 2a, indicating a distinct band alignment and mobility for carriers. With the different wavelengths of light excitation onto the device, all exhibit negative photocurrents from 488 nm to 1064 nm, which are highly reproducible and stable under the Vg of 10 V as plotted in Fig. S4. Since the IEICO-4F is a non-fullerene acceptor with the LUMO (HUMO) of 4.16 eV (5.44 eV) and PTB7-Th is a donor with LUMO (HUMO) of 3.55 eV (5.27 eV) for formation of type-II heterojunction for exciton separation and charge transfer.33,34 In this configuration, graphene acts as collective channel, PTB7-Th:IEICO-4F bulk heterojunction work as mainly absorption layer in visible-infrared regime (on the basic of the absorption spectra in Fig. S2a). However, the carrier mobility of blend structure PTB7-Th:IEICO-4F is relatively low which leads the carrier recombination before collected by graphene channel.35 Thus, we restrict the thickness of active blend heterojunction into 35 nm scale for facilitating the photo-response. The control device without ZnO layer still exhibits inferior performance as shown in Fig. S5, suggests that photo-generated holes and electrons recombination in graphene. With the strong electron affinity of ZnO due to energy band alignment, implies the intensive electrons extraction from D-A system PTB7-Th:IEICO-4F to graphene channel (Fig. 2b). Under the illumination of light, photo-generated hole-electron pairs dissociated and electrons are extracted into graphene causing a N-doping, leading the negative photocurrents (Fig. 2c). The intrinsic graphene is hole dominated characterized by the transfer curve in dark condition with a Dirac point at 27.6 V (Fig. 2c). The 5 mW/cm² 895 nm light induced a N-doping negative shift with new Dirac point at 7.2 V, which induce carriers variation can be computed by light-doping expression: \( \Delta n = C_{ox} |V_g - V_{Dirac}|/e - \sigma/e\mu \). As the consequence, a concentration of \( 1.39 \times 10^{12}/\text{cm}^2 \) photo-generated electrons transfer into graphene.

To better understand the negative or positive photo-response dynamic, we further investigate the bi-directional time-resolved photocurrents measurements with the variation of Vg in Fig. S6a and 3a. With the Vg growth, the intrinsic negative infrared 940 nm photo-responses significantly converts into positive photocurrents (Fig. S6a). When \( V_g < V_{Dirac}=27.6 \text{ V} \), the hole predominates in graphene channel (Fig. 2b and c). The photo-generated electrons inject into graphene, giving a negative response (Fig. S6a top panel). And at deep-hole condition (such as \( V_g=-13 \text{ V} \)), the bending energy band can remarkably promote the electrons injection with direct photocurrent of 3.23-fold improvement compared to \( V_g=10 \text{ V} \) (Fig. 2c). In the electron branch with \( V_g \) increasing (\( V_g > V_{Dirac}=27.6 \text{ V} \)), we observe the positive photocurrents with increasing and degradation which is slightly distinct with hole branch. The main reason accounts for this inhibition photocurrents when \( V_g > 40 \text{ V} \) is that the Fermi level of electron-dominated graphene may approaches or exceed LUMO level of ZnO (4.4 eV), causing the carrier quenching. For further acquisition of optimized photo-responsivity, we characterize the photocurrent as the function of Vg sweeping from -20 V to 65 V with the variation of input optical power density (Fig. 2a). The asymmetry bidirectional photocurrents imply that deep-hole graphene can induce the higher photo-gating effect at sacrifice of the response time (Fig. 1c and d). And positive responses start degradation when the Vg exceeding 42.8 V (the corresponding work function is nearly 4.43 eV which can be determined by \( E_f = \hbar v_F \sqrt{\pi C_{ox} |V_g - V_{Dirac}|/e} \)). As we can see in Fig. 2c, the transfer curves under the irradiation of light show a bending near the Vg approaches 40 V, revealing validity of the photocurrents inhibition
theory proposed before. Moreover, our graphene heterojunction photoconductor exhibits linear photocurrent with bias voltage at various $V_g$ (Fig. S6b).

When the $V_g$ approaches 7.5 V (hole branch of graphene), the device shows no photo-memory due to the shallow trapping state for electron. By utilizing the gate modulation tendency from Fig. 2a, we present the detailed no memory optimized photo-responsivity ($R$) measurements with the constant $V_g=10$ V and $V_g=50$ V. The power-dependent time-resolved responses are measured in Fig. 2d, demonstrating the prominent photo-sensitive capacity even at ultra-low input optical power (895 nm, 14.4 nW/cm$^2$, $V_g=10$ V and 50 V). And the highest infrared $R$ of $1.88 \times 10^6$ A/W in negative responses and $1.3 \times 10^4$ A/W at positive responses, which is comparable with infrared detection performance of graphene based phototransistors.$^{10,37}$ The Fig. 2e shows a logarithm linear relation in most excitation wavelength and $R$ decreases with the light irradiation with the variation tendency: $R = \frac{AC}{\mu} \frac{V_{ds}}{wl} p^{\beta - 1}$. Especially for the 895nm photo-responses as illustrated inset the Fig. 2e, the fitting curve show linear photo-gating hole trapping state with the $\beta = 0.417$, whereas for 1064 nm, the linear relation of $R$-Power shows a saturation around 1.16 $\mu$W/cm$^2$ and the (Fig. 2e) corresponding to the cut-off edge of D-A absorption around 1 $\mu$m. Compared with previous bilayer and bulk heterojunction enhanced graphene, our device exhibits nearly 2 to 100 fold improvement in $R$.$^{18-20}$

In sensitized photo-gating phototransistors, the prolonged recombination time of trapping carriers and another type of carriers recycled in channel, yielding high gain responses. Therefore, the gain (recycled times) is associated with recombination time and computed by $G = \frac{\tau_{transit}}{\tau_{eff}}$, where the $\tau_{eff}$ is the life time of carriers and the $\tau_{transit}$ is the transit can be estimated by $\tau_{transit} = \frac{l^2}{\mu V_{ds}}$ (the $\mu$ is obtain by Fig. S7). The relative long recovery time always originates inferior operation time of device. Consequently, there is a trade-off relation between the $R$ and operation bandwidth. The transient responses of device with different gate biases are measured in Fig. S8a. The response and recover time are respectively 178 $\mu$s and 1.41 ms at $V_{ds}$ of 1 V with 15 V $V_g$ (Fig. S8a top panel), which is superior with the majority of previous graphene photoconductive works.$^{15-12,17-20}$ For higher responsivity, the recovery time has been prolonged to 32 ms at 10 V $V_g$ (Fig. S8a bottom panel). Thus, the gain of negative infrared responses reaches up to $5.01 \times 10^6$ with EQE up to $2.6 \times 10^8$ % under 10 V $V_g$.

To further investigate the signal-noise character of negative infrared responses, we perform gate-dependent noise spectral in Fig. S8b. At the $V_g=20$ V, the noise spectral density can reach lowest at $1.23 \times 10^{-9}$ (100 Hz) due to direct suppression of dark current near the Dirac point. We use a simple but precise method to evaluate the detectivity by formula: $D^* = R(Sf)\frac{l_n}{2}$, where the $f$ represents operation frequency, $S$ stands for active area and $l_n$ is the noise current. With the measured noise characterization (Fig. S8b) and responsivity-frequency relation (Fig. S9), the color map of detectivity ($D^*$) can be plotted as depicted in Fig. 2f. At the bias of 1 V, the values of $D^*$ in low frequency regime (1-10 Hz) fluctuates from $10^{10}$-10$^{12}$ Jones due to the affection of flicker noise. When the $f$ approaches the bandwidth (127 Hz) of device, the $D^*$ shows over $1.4 \times 10^{12}$ Jones mainly in region of infrared band (780-940 nm). The maximum of $D^*$ can be achieved up to $4.8 \times 10^{12}$ Jones at frequency of 100 Hz with the applied gate 10 V.

After systematic investigation on crucial parameters of no-memory photo-response, the long-
term infrared photo-memory characters are exhibited in deep-hole branch graphene condition. We first carefully explore the infrared photo-induced memory case. Fig. 3a depicts the stable and erasable near-infrared storage in 3 cycles when graphene is heavily hole-doped (Vg work at -30 V). We find that the hysteresis phenomenon appears in variation of \( I_{ds} \) with \( V_g \) relation in Fig. 3b. Owing to the prolonged hole trapping state induced by ZnO, the optoelectronic memory function can be tuned and erased by Vg. There is also a conspicuous reference erasing Vg voltage which, as in bending point of transfer curve (arrow in Fig. 3b), are implied so as to ensure the value of Vg pulse is adequate for the entirely resettable. The low erasing Vg pulse proved unequal to the reset task (Fig. S10a). When the Fermi level of graphene approaches or surpass the LUMO level of ZnO, corresponding Vg transforms graphene into N-doping. And the N-type graphene can supply abundant electrons into active layer to neutralize hole trapping near ZnO interface for erasing. Therefore, by applying a +60 V Vg pulse, we can rapidly erase the memorial photo-response and reset in nearly 5 seconds (Fig. 3a). Note that fast downward pulse in Fig. 3a, which is ascribed to transient resistance change of graphene due to Vg pulse.

By utilizing the bi-directional photocurrents switching feature (Fig. 2a and Fig. 3b), there implies a threshold voltage (V\(_T\)) that can modulate the on-off state in photo-induced storage. V\(_T\) can be obtained from transfer curve at the node of dark \( I_{ds} \) and light \( I_{ds} \) branches, which is 13.7 V. The distinct color traces in Fig. 3c represent different Vg working state. At the programming occasion (Fig. 3c top panel), the photocurrent comes to nearly 574 \( \mu \)A with 151 \( \mu \)A memory current (Vg = -10 V) The deep P-doping graphene facilitates the photo-generated electrons injection into graphene giving a strongly memorial response. With Vg approaching the V\(_T\), few of photo-generated electrons neutralize hole in graphene and corresponding holes trapping into active layer resulting in inferior photo-storage or no-memory photo-response condition. However, the practical measurements show that V\(_T\) is roughly 20 V due to electric drift effect (Fig. 3c). As the Vg reaches 10 V, the photocurrent has slumped to around 99 \( \mu \)A with the no photo-memory. Higher Vg (20 V) attempt to further inhibition of response is plotted in Fig. 3c (blue point), the suppression ratio has increased by 95.3 % in terms of Vg=-10 V.

More measurements are raised on optimized factors for implement of long-term memory. The pulsed illumination of constant input power at 940 nm with 3 cycles in Fig. 3d shows an on-off state at the higher Vg which is ascribed to the different value of Dirac shift induced by 940 nm. The shallow-hole graphene (Vg= 10, 20 V) implies obvious relaxation (over 25 % of current losses in 20 s). Furthermore, the power-dependent ‘light write-reset’ cycles are depicted in Fig. 3e. We notice that for stronger input light power (input light>12.5 \( \mu W/cm^2 \), light pulse width=0.5 s), the retention current after light pulse can hardly maintain but quick release to recover, the photocurrent of 91 \( \mu W/cm^2 \) and 12.5 \( \mu W/cm^2 \) input light has fallen to 85.4 % and 89.9 % respectively within nearly 20 s. The releasing current fluctuation stay close to the 3.3 % in the same time range under the 1.3 \( \mu W/cm^2 \) power. And even retention current becomes nearly constant (0.27 % fluctuation) at the power of 0.32 \( \mu W/cm^2 \). The lower Fermi level of graphene gains much photo-generated electrons traction into channel for lager photocurrents, which may make more contribution to prolong the retention time. Fig. 3f shows the deep-hole Vg can promote the retention time limited extent (nearly 2-fold prolong time in Fig. 3f top panel and middle panel). And the retention time is also found less correlation with light pulse width (Fig. S10b). Thus, we achieve retention time up to 10\(^4\) s with relative small input power (0.32 \( \mu W/cm^2 \)) and deep-hole at higher Vg=-30 V (Fig. 4i bottom panel). The exponential fitting of \( e^{-t/\tau} \) gives a life time \( \tau=11999 \) s with 1 s light pulse (Fig. 3i bottom
Furthermore, we demonstrate 3×4 array for assessment of the infrared imaging performance (Fig. 4a). In this test, each detection unit stands for one pixel. The letter “H” pattern is mapping onto the detection array under the 940 nm light irradiation. Two working mode are shown. The receptive field is a part of biological image preprocessing systems, including two types of bipolar cells, on/off cells (When receiving light stimuli, the response of on cells is positive while the response of off cells is negative). Our devices function similarly to these cells and can work as a proper simulation of visual processing (Fig. 4a and b). Based on the device performance, we constructed a 3×3 artificial receptive field, which consists of a central device and 8 surrounding devices. The center device works in the state of negative photocurrent. While the surrounding units work in the state of positive photocurrent. The processing function is demonstrated by the greyscale “Leave” image, shown in Fig. 4c, and the process are provided in Fig. 4b. Also, by controlling the Vg, more functions were achieved (Fig. 4b, e and f), which means the reconfigurability of the device array has more potential functions. As an example, two additional functions, embossing, and blur are demonstrated in Fig. 4e and f. Traditional visual circus processes the image by transporting the image from the camera to the processor, leading to energy waste and efficiency loss on transporting. Different from the traditional visual circus, real-time sensing, and processing greatly avoid the interference of miscellaneous information and improve efficiency. Especially, to deserve to be mentioned that, it is the first time that infrared light was applied in the preprocessing by Van der Waals heterojunctions. Moreover, we mimic memorial preprocessing and forgetting process of human visual systems when the Vg approaches -20 V (Fig. 4g). The infrared image memorizations of “H” pattern are shown in Fig. 4h with just 1 optical pulse. After the light stimuli, the dimmer “H” pattern is observed with memory effect. By further forgetting process, “H” pattern vanishes. All this processes show the capability of multi-functional infrared artificial visual based on ORRAM array.

This on-off state switching phototransistor demonstrates prominent performance both in photo-storage and photo-detection. The state-of-the-art of sensitized graphene phototransistors with similar framework and photo-memory devices are summarized in Table 1. Graphene based photoconductor have mainly two outlets for high-performance design, like single layer enhanced and bilayer or bulk heterojunction enhanced strategies (Table 1). It is notable that the majority devices have averaged over millisecond response time and low photo-responsivity in infrared regime. Our phototransistor exhibits high optical sensitivity in fast response time with simultaneous long-term gate-control photo-memory feature due to high-efficiency multi-layer heterojunction and effective trapping state.

Conclusions

With the system so well-designed, we show a photo-storage and photo-detection switching characteristics in graphene/ZnO/PTB7-Th:IEICO-4F framework phototransistor. The devices show over 10⁶ A/W infrared photo-responsivity with fast response time down to 178 μs and the corresponding D* is up to 4.8×10¹² Jones. And a long-term (10⁴ s) infrared photo-memory with writing and erasing functions can be achieved. The reset Vg reference value is further validated by bending point of transfer curve. The systematic investigations have been done in tunable infrared photo-memory phenomenon. The infrared retinomorphic vision and memorial preprocessing functions in artificial visual can operate simultaneously. The high performance infrared photoresponses and multi-functional artificial visual applications are highly beneficial for devising future
switching bio-optoelectronic applications with fast speed and high photo-responsivity.

Table 1. Comparison with previous graphene phototransistors and photo-memory device with high performance.

| Framework                              | Active materials                              | Responsivity [A/W] | Response time [ms] | Spectral range [nm] | Photo-memory Feature | Ref. |
|----------------------------------------|-----------------------------------------------|---------------------|--------------------|---------------------|----------------------|------|
| Single layer enhance                   | Graphene/SWNTs                                | 100                 | 0.1                | 400-1550            | NA                   | 10   |
|                                        | Graphene/C$_6$-BTBT                            | $10^4$              | 25                 | 355                 | NA                   | 17   |
|                                        | Graphene/Rubrene                              | $10^7$              | 100                | 400-600             | NA                   | 13   |
|                                        | Graphene/WS$_2$                               | $10^6$              | 0.13               | 400-700             | NA                   | 9    |
|                                        | Graphene/PbS QDs                              | $10^7$              | 10                 | 600-1450            | NA                   | 5    |
|                                        | Graphene/MA$PbBr_3$                           | $1.4 \times 10^8$   | 4.5$\times 10^3$   | 430-440             | NA                   | 7    |
|                                        | Graphene/COF                                  | $3.2 \times 10^7$   | 1.14               | 400-600             | NA                   | 8    |
| Bilayer or Bulk heterojunction enhance | Graphene/PCBM/perovskite                      | $8 \times 10^5$     | $10^6$             | 400-750             | NA                   | 18   |
|                                        | Graphene/PTCDA/Pentacene                      | $10^5$              | 0.03               | 400-700             | NA                   | 19   |
|                                        | Graphene/C$_{60}$/Pentacene                   | 9127                | 0.275              | 405-1550            | NA                   | 20   |
|                                        | Graphene/ZnO/PTB7-Th:IEICO-4F                 | 1.88$\times 10^6$   | 0.178              | 488-1064            | over 10$^4$s with switching states | This work |
| Photo-memory devices                   | MoS$_2$/PbS                                   | $10^7$              | NA                 | 808-1940            | over 10$^4$s         | 31   |
|                                        | ReS$_2$/h-BN/graphene                         | NA                  | NA                 | 532                 | over 10$^4$s         | 32   |
|                                        | Sol-PDI/C$_{10}$-DNTT                         | NA                  | NA                 | 405-640             | over 10$^4$s         | 40   |
|                                        | diF-TESADT                                    | $10^4$              | NA                 | 575                 | over 600s            | 41   |
|                                        | Perovskite/IGZO                               | $10^5$              | NA                 | 457-1064            | over 10$^4$s         | 42   |
|                                        | Graphene/MoS$_2$                              | $10^{10}$           | NA                 | visible             | over 10$^4$s         | 6    |

Materials and methods

Device Fabrication and Characterization: Single-layer CVD graphene was transferred to an n+ Si/SiO$_2$ substrate (SiO$_2$ 285 nm) by a solution method. Then the 90 nm gold electrode is deposited by thermal evaporation with symmetrical contact onto prepared graphene to form a channel (25 μm in length and 250 μm in width). ZnO
precursor solution was formed by dissolving 110 mg of zinc acetate dehydrate (Zn(CH$_3$COO)$_2$$\cdot$2H$_2$O) and 31 mg of ethanolamine in 1 ml of 2-methoxyethanol with stirring overnight, and then was spin-coated on graphene to form 15 nm thin films. PTB7-Th: IEICO-4F (1: 1.5) mixture was dissolved in in CB: CN (96:4, volume ratio) solution with a donor concentration of 8 mg/ml. The thickness of BHJ layer was tuned at 35 nm.

**Measurement Methods:** The absorption spectra were recorded by Hitachi U-3010 UV-VS. The thickness of BHJ layer was calibrated by a step profilometer (AMBIOS-XP2). Atomic force microscope (AFM, Asylum Research AFM system (MFP-3D-BIO)) and transmission electron microscopy (TEM, Hitachi TEM system) were used to investigate the film morphology. Cross-sectional TEM was acquired using Helios NanoLab 600i. Grazing-incidence wide-angle X-ray scattering (GIWAXS) experiments were performed at BL18U beam line of shanghai synchrotron radiation facility (SSRF) and utilized to analyze the molecule orientation and stacking property of BHJ. All samples were radiated at 12.67 keV X-ray with an incident angle of 0.12°, Pilatus 6M is the detector. All electrical and photo-response characteristics of graphene/organic semiconductor heterojunction phototransistors were measured by a Keithley 2636b source meter analyzer and PDA with different radiation wavelength. Raman spectra measurement was performed by a confocal microprobe Raman spectrometer (RENISHAW inVia Raman Microscope) under the illumination of a 514 nm helium-neon laser.
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Conflict of interests

The authors declare that they have no conflicts of interest.

Contributions

J.H., X.D., Z.Z. contribute equally to this work. J.W., S.T., W.H. conceived and supervised the project. J.H., X.D. and Z.H. prepared the device and performed the photocurrent measurements and characterizations. J.H. set up the experimental platform, and J.H., X.D., and Z.H. performed the data analysis. Z.Z. performed artificial visual content. J.H., X.D. and Z.Z. co-wrote this paper. All authors discussed the results and commented on the manuscript.
Figure Caption

**Fig. 1** | Morphology characterization and multifunctional switching of graphene/organic phototransistor in different Vg. **a**, Cross-sectional TEM image. **b**, Device structure of graphene/ZnO/PTB7-Th:IEICO-4F phototransistor. **c,d,e,f** The time resolved photocurrents under the different Vg. **c**, Vg=-20 V, long-term photo-memory state in negative photocurrent. **d**, Vg=7.5 V, no memory photo-response state in negative photocurrent. **e**, Vg=17 V, off state. **f**, Vg=40 V, no memory photo-response state in positive photocurrent.

**Fig. 2** | The transfer, no memory photo-responsive characteristics of graphene/organic phototransistor. **a**, Vg and Power dependent photocurrents intensity distribution colormap. **b**, The energy band alignment and charge transfer diagram of the multi-layer heterojunction under the light illumination at different Vg conditions. Top panel is deep-hole graphene circumstance and bottom panel corresponding to shallow-hole or electron branch graphene. **c**, Transfer characteristics of the graphene/organic phototransistor under different input wavelengths with the same power density= 60 mW/cm². **d**, The photocurrent variations of the different Vg values (Vg=10 V top panel, Vg=50 V bottom panel) with the reductive input light power. **e**, The photo-responsivity plotted as a function of input light power density. The inside is the logarithm linear fitting curve. **f**, The frequency and wavelength dependent color map of detectivity.

**Fig. 3** | The transfer, infrared photo-memory cycle and retention characteristics of graphene/organic phototransistor. **a**, The 3 writing and erasing cycles. **b**, The hysteresis transfer curve along with arrow direction and inside is transfer curve of device with or without light illumination. **c**, The gate-dependent photo-memory switching operation. **d**, Gate-related relaxation currents under the constant input light power with 1 s pulse width. For all traces, input light power=663 μW/cm², gate pulse width=0.25 s. **e**, The 3 programming-relaxation cycle measurements as the function of input light power. light pulse width=0.5 s, gate pulse width=0.25 s. **f**, The retention and persistent state of infrared photo-memory with different Vg (two of top panel). Optimized retention in bottom panel with proper input power and Vg.

**Fig. 4** | The planar array of graphene/organic phototransistors for reconfigurable retinomorphic vision and memorial preprocessing application. **a**, The schematic diagram of 3×4 array irradiated by infrared light irradiation metal through the shadow mask with ‘H’ shaped pattern and photoresponse of the negative and positive photoresponsive modes with the infrared 940 nm light (3.76 mW/cm²) for imaging. Inset is the 3×4 array picture. **b**, Simulations of reconfigurable retinomorphic vision. **c**, Original image. **d**, Edge detection. **e**, Emboss. **f**, Dispose of Blurring. **g**, Schematics of the human visual system. **h**, The illustrations of the image memory of the letter ‘H’ at memorial preprocessing working mode (Vg=-20 V, 940 nm light with 3.76 mW/cm², optical pulse width: 2s, electric pulse width: 1s).
Figures

Fig. 1.
Fig. 2.
Fig. 3.
Fig. 4.
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