All-Perovskite Photodetector with Fast Response

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Abstract
Perovskites have attracted substantial attention on account of their excellent physical properties and simple preparation process. Here we demonstrated an improved photodetector based on solution-processing organic-inorganic hybrid perovskite CH$_3$NH$_3$PbI$_3$$_x$Cl$_{1-x}$ layer decorated with CsPbBr$_3$ perovskite quantum dots. The CH$_3$NH$_3$PbI$_3$$_x$Cl$_{1-x}$-CsPbBr$_3$ photodetector was operated in a visible light region, which appeared high responsivity ($R = 0.39$ A/W), detectivity ($D^* = 5.43 \times 10^9$ Jones), carrier mobility ($\mu_p = 172$ cm$^2$ V$^{-1}$ s$^{-1}$ and $\mu_n = 216$ cm$^2$ V$^{-1}$ s$^{-1}$), and fast response (rise time $121 \mu$s and fall time $107 \mu$s). The CH$_3$NH$_3$PbI$_3$$_x$Cl$_{1-x}$-CsPbBr$_3$ heterostructure is anticipated to find comprehensive applications in future high-performance photoelectronic devices.

Keywords: Perovskite, CsPbBr$_3$ quantum dots, Photodetector, Fast response

Background
Photodetectors (PDs), which transduce the optical signal into electric information, are one of the key semiconductor devices in many fields, such as optical image sensors, environmental surveillance, electrocommunication and remote sensing technology, etc. [1–4]. Three kinds of devices, i.e., photodiodes, photoconductors, and photo-FETs (field-effect transistors), are commonly adopted to detect optical signals. Especially, photo-FETs are considered as a promising architecture for photodetectors due to their capability to balance high gain and low dark current in comparison with photodiodes and photoconductors.

The photo-FETs have been explored extensively by many groups [5–11]. Generally, to achieve low dark current, a thin active layer is favorable, which renders as depletion layer and can be tuned readily by electric field applied from a gate electrode. However, the thinner the thickness of the active layer is, the lower the optical absorption level is, which leads to low sensitivity. The materials to form an active layer of photo-FETs, therefore, should have high photo-electric conversion efficiency (PECE). Varieties of materials, such as quantum dots (QDs) [12], carbon nanotubes [13], graphene [14], transition metal dichalcogenides (TMDCs) [15], black phosphorus [16], organic molecules, [17] etc., have been employed as active layers for high optical performance of photo-FETs. Until now, halide perovskite has been used widespread as photoactive materials for developing high-performance optoelectronic devices due to its high optical absorption, conversion efficiency, and readily prepared method. Recently, the halide perovskite was also found applications in the high-performance photo-FETs [18–27].

However, even with high PECE material (such as organic/inorganic hybrid perovskite) used as a depletion layer, the light absorption cannot satisfy the practical applications of photo-FETs for efficient gate control. To address the issue, i.e., achieving high gain with low dark current, many solutions have been developed, such as doped with high absorption materials and noble metal nanoparticles for plasmonic enhancement. Among them, the architecture with dyesensitizer layer prepared on the active layer renders as a promising solution. This architecture can decouple the absorption (in the sensitizer) and charge transport (in the channel) and allows operation of the thin channel layer in full depletion with high optical absorption. Accordingly, a strong absorbing semiconductor is a favorable sensitizer to prepare the high-performance photo-FETs. The QDs, such as PbSe [28], PbS [29], and CdSe [30], have attracted much attention due to the peculiar properties (high quantum yield...
efficiency, size-sensitive absorption spectrum, etc.) and have been employed in a diversity of high-performance optoelectronic devices.

Very recently, a novel class QDs, i.e., perovskite QDs, have been successfully developed and used in various fields, such as solar cells [31], LEDs [32], and single photon emitters [33]. Considering the requirements of photodetectors, perovskite QDs, i.e., CsPbX3 (X = Cl, Br, I), is also a suitable sensitizer to enhance the light absorption. As aforementioned, organic-inorganic hybrid perovskite materials have been proved a promising solution for high-performance photo-FETs. In view of the figure of merit of inorganic perovskite quantum dot, we anticipate the all-perovskite device composed of solution-processed CH3NH3PbI3-xClx depletion layer and CsPbBr3 QDs sensitizer layer will exhibit excellent performances in responsivity and detectivity. To our knowledge, this composed perovskite photo-FET has not been fully explored before.

In this paper, CH3NH3PbI3-xClx perovskite-CsPbBr3 QDs hybrid photodetector (CCPD) is prepared with solution-processed strategy. The fabricated photodetector exhibits a wide spectrum spanning from 400 to 800 nm, high responsivity (0.39 A/W), detectivity (5.43 × 109 Jones), carrier mobility (μp = 172 cm2 V−1 s−1 and μn = 216 cm2 V−1 s−1), fast response (rise time 121 μs and fall time 107 μs), and good reproducibility. Solution-based CH3NH3PbI3-xClx-CsPbBr3 heterostructures pave a way for the high-performance optoelectronic device in the UV-visible light region.

Materials and Methods

Device Fabrication

First, on the substrate, a commercial silicon wafer (n+ Si) with a 300-nm-thick SiO2 layer (Suzhou Crystal Silicon Electronic & Technology Co., Ltd), active layer (organic-inorganic hybrid perovskite CH3NH3PbI3-xClx) was deposited via spin-coating followed with 90 min post-annealing to resin the film. Subsequently, the sensitized layer, CsPbBr3 QDs, was spin-coated layer by layer for three times at 1500 rpm and dried in 60 °C for 15 min on a hot plate after each spin-coating. The source and drain electrodes were thermally evaporated through a sophisticated shadow mask with a channel length (L) of 0.1 mm and a channel width (W) of 2.5 mm.

Results and Discussion

As shown in Fig. 1a, the photodetectors are composed of a gate electrode, a silicon wafer (n+ Si) with a 300-nm-thick SiO2 layer (capacitance Cox of 11.5 nF cm−2), active layer (organic-inorganic hybrid perovskite thin film prepared by one step spin-coating solution processing), decorated layer (CsPbBr3 QDs), and source and drain electrodes (thermally evaporated through masks). Figure 1b describes the cross-sectional scanning electron microscopy (SEM) image of the device. The thickness of SiO2 dielectric layer is 300 nm, CH3NH3PbI3_xClx perovskite active layer is about 102 nm, and the decorated CsPbBr3 QDs layer film is about 97 nm. The diagram shows clearly that the interface between CH3NH3PbI3-xClx perovskite and CsPbBr3 QDs is clear and has no intermediate layer, manifesting optimized photovoltaic properties. As aforementioned, in photo-FETs, the thickness of semiconducting channel plays a crucial role. Firstly, a thinner active layer is required for tuning the behavior effectively. The thinner perovskite films, however, are prone to produce pinholes, leading to inhomogeneous conduction in the channel. Meanwhile, the thinner active layer means low photon absorption as well. The optimized thickness of the CH3NH3PbI3-xClx film in our device is at about 102 nm. To enhance light-matter interaction in a thinner perovskite device, 97-nm CsPbBr3 QD layer, optimum sensitizer with strong absorption is prepared. TEM image of CsPbBr3 QDs, in Fig. 1c, shows the uniform particle size and rectangle shape. The inset of Fig. 1c shows the X-ray diffraction (XRD) peaks. The peaks show a typical cubic structure (ICPDS No. 54-0752), which is coincided with the TEM results. Furthermore, to investigate the crystallinity of the CH3NH3PbI3-xClx film, X-ray diffraction (XRD) spectrum of the perovskite film synthesized on a glass substrate chalk up. Figure 1d presents the XRD spectrum, and four characteristic peaks centered at 14.2°, 28.6°, 31.02°, and 43.38° are assigned to the (110), (220), (310), and (330) planes, respectively, indicating that the halide perovskite films possess the expected orthorhombic crystal structure with high crystallinity, which is consistent with the reported literature [34–38].

According to light absorption curves of the CH3NH3PbI3-xClx perovskite (blue line) and perovskite decorated by CsPbBr3 QDs (pink line) as shown in Fig. 1e, the decorated CsPbBr3 QDs can only enhance the absorption for a narrow range (400–500 nm) in comparison with CH3NH3PbI3-xClx layer solely. Furthermore, we also calculated the bandgap of QDs according to the Tauc equation [39–44] as shown in Additional file 1: Figure S1. The bandgap is about 2.38 eV. The photoluminescence (PL) spectrum of QDs is also shown in Additional file 1: Figure S2, the central wavelength of PL is almost equal to the absorption edge.

Next, the electrical properties of the devices were explored. Figure 2a describes the I–V characteristics of photodetectors with varied gate voltages (0 V, ± 0.2 V, ±

Materials

N,N-dimethylformamide (DMF, 99.5%), oleic acid (OA, 90%), 1-octadecene (ODE, 90%), oleylamine (OLA, 90%), PbCl2 (99.99%), PbBr2 (AR, 99.0%), and CH3NH3I (98.0%) were purchased from Aladdin.

The details about synthesis of CH3NH3PbI3-xClx perovskite, fabrication of CsPbBr3 QDs, and the instrument model are placed at Additional file 1.
0.4 V, ± 0.6 V, ± 0.8 V, ± 1.0 V) in the dark. There are two states in detectors, according to Fig. 2a. At OFF-state (|VGS| = 0), the spectral lines are linear, and IDS increases rapidly with the increase of VDS, indicating that a Schottky barrier forms in the device. While at ON-state (|VGS| ≥ 0.4 V), linear-to-saturation current-voltage characteristics appear as the voltage increase, similar to traditional FETs. On account of the excitons remain in the trap states [45] of perovskite which cannot be converted to the photocurrent, leading to the saturation of photocurrent.

The ambipolar performance can be concluded from the transfer characteristics (Fig. 2b) under dark and light illumination, i.e., for both negative VGS and VDS the device operates as hole-enhancement mode and, to the contrary, the device operates in the electron-enhancement mode with both positive VGS and VDS. Owing to the difference of electron potentials, holes separated from photoexcitons generated in heterojunction tend to reside in the perovskite layer. By increasing the incident power density, the transfer rate of holes is higher than electrons. The curve shift towards to positive VGS in Fig. 2b indicates the heterojunction tend to be p-type in this device. Meanwhile, in the linear region, the relationship between field-effect mobility and gate voltage can be extracted with the equation of

\[ \mu = \frac{L}{V_{DS}C_{ox}W} \frac{\partial I_{DS}}{\partial V_{GS}} \]  

where L and W are the length and width of the channel, respectively, and C_{ox} is the capacitance per area. Therefore, the mobility for both holes and electrons can be calculated as 172 cm² V⁻¹ s⁻¹ and 216 cm² V⁻¹ s⁻¹. This balanced hole and electron mobility further explains the ambipolar behavior of the device under light illumination.
Figure 2c and d describe the photoelectric properties of the fabricated device. Figure 2c shows the curve of photo-detectors as a function of negative gate-source voltage at $V_{DS} = -1$ V with varying incidence optical powers. It is obvious that the device exhibits $n$-type doping behavior. Built-in field at the heterojunction promotes more electron-hole pairs separation and accelerated holes injection into the perovskite channel for negative $V_{GS}$ and $V_{DS}$.

Figure 2d exhibits the $R$ of the device with the relation of irradiance ($E_e$), in which the wavelength of incident light is 405 nm. As can be seen, the $R$ decreases linearly with $E_e$ at irradiant power under 200 mW/cm$^2$, while it deviates from linearity at power irradiation greater than 200 mW/cm$^2$.

In order to insight the superior performance of the CCPD. A series of comparisons is necessary. Figure 3a shows the comparison of $R$ about the device with the relation of irradiance ($E_e$), in which the wavelength of incident light is 405 nm. As can be seen, the $R$ decreases linearly with $E_e$ at irradiant power under 200 mW/cm$^2$, while it deviates from linearity at power irradiation greater than 200 mW/cm$^2$.

The $R$, as a figure of merit in photodetector, can be calculated from the formula of

$$ R = \frac{I_p}{W \times L \times E_e} \quad (2) $$

where $L$ is the channel length (0.1 mm), $W$ is the channel width (2.5 mm), and $I_p$ is the difference value between light photocurrent and dark photocurrent measured at $V_{DS} = 1$ V in the output curve. The maximum $R$ is calculated to be 0.39 A/W (in CCPD), obviously larger than that of 0.22 A/W (in CPD). The enhanced responsivity of CCPD is attributed to the CsPbBr$_3$ QDs sensitizer with high light absorption and efficient carrier injection in the perovskite layer.

Detectivity ($D^*$) is another key parameter for evaluating the performance of photodetectors. Based on pre-existing responsivity numerical value, the $D^*$ versus irradiance ($E_e$) can be estimated by the following equation:

$$ D^* = \frac{RA^2}{(2eI_{DS})^\frac{1}{2}} \quad (3) $$

Where $R$, $A$, $e$, and $I_{DS}$ are the responsivity, available channel area of the devices, charge of an electron, and dark current, respectively. As shown in Fig. 3b, it is clear that $D^*$ of CCPD (5.43 $\times$ 10$^9$ Jones) is notably higher than that of CPD (1.25 $\times$ 10$^9$ Jones). Further proving sensitized channel material with strongly absorbing CsPbBr$_3$ QDs can improve device performance.

Other key parameters representation photodetector's performance, such as the noise equivalent power (NEP), and the gain ($G$) can be given as [46]
where $R$, $A$, $e$, and $I_{DS}$ have the same meaning as the previous one. Particularly, when the maximum $R$ of CCPD is 0.39 A/W, the $D^*$ reached $5.43 \times 10^9$ Jones. In this condition, the $NEP$ and $G$ of this device can be received at an incredibly high value of $9.21 \times 10^{-12}$ W/Hz and 1.197, respectively.

Responsibility to optical signals is an important index about efficient carrier transport and collection. Figure 4a shows the drain current with on-off light cycles at a time interval of 20 ms and biased $V_{DS} = 1$ V, $V_{GS} = 1$ V. As can be seen, the current rises rapidly as soon as the light turned on and reduces quickly while the light turned off, suggesting the good stability and reproducibility in the progress of on-off cycles with light irradiance of 648 mW/cm$^2$ at 405 nm. However, a time interval of 20 ms is too long to express the photocurrent response of the device. To calculate the response time of the device, a 4000-Hz pulsed light source is used to irradiate the device. Figure 4b depicts the temporal photocurrent response of the image. The rise and fall times of the photocurrent are $\sim 121$ and $\sim 107$ $\mu$s, respectively, indicating an ultrafast response speed than previous reports, as shown in Table 1.

The working principle and interfacial processes of CCPD are schematically showed in Fig. 5. The fabricated detector was excited with 405 nm (3.06 eV) laser, which photon energy is larger than both the hybrid perovskite (1.5 eV) and CsPbBr$_3$ (2.4 eV) to ensure the generation of exciton in both layers. As the discrepancy of Fermi energy ($E_F$) of CsPbBr$_3$ and hybrid perovskite, the heterojunction would be formed at the interfaces of the two layers, which would mediate or suppress the diffusion of the carriers. Fortunately, the $E_F$ of CsPbBr$_3$ is higher than that of hybrid perovskite and leads to an energy configuration as shown in Fig. 5. According to this energy level configuration, the interface can mediate the transport of both carriers from sensitizer layer to

![Fig. 3 Key parameters of CCPD.](image)

![Fig. 4 Photoresponse characteristics of CCPD.](image)
active layer, which will enhance the performance of the device. On the other hand, the pristine perovskite has a low density of surface states [49], which lead the band’s easy bending to light absorber layer when the two layers form a heterojunction. This energy level alignment plays an important role in the diffusion of electrons from the sensitizer absorption layer to the perovskite transport layer. The energetic level configuration can accelerate the holes injecting from the CsPbBr3 sensitized absorption layer to hybrid perovskite transfer layer, which is coincident with the significant current increases in negative VGS upon light illumination (shown in Fig. 2b). Meanwhile, the heterojunction in hybrid perovskite/CsPbBr3 depletion layer accelerates the separation rate of electron-hole pairs and reduces the separation time, leading to a fast response of the hundred microseconds order.

**Conclusion**

In conclusion, we demonstrated highly photosensitive perovskite photodetectors decorated by perovskite QDs. This novel photodetector is operated in a visible light region, which appears high responsivity ($R = 0.39 \text{ A/W}$), detectivity ($D^* = 5.43 \times 10^9$ Jones), and carrier mobility ($\mu_p = 172 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $\mu_n = 216 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$). Meanwhile, the devices also show fast response (rise time 121 $\mu$s and fall time 107 $\mu$s) and better on-off stability and reproducibility under 405 nm illumination. However, on the one hand, the wide electrode span (hundreds of micrometers) lowers the performances of devices such as photocurrent-related responsivity. Efforts need to be paid to reduce the electrode spacing width for efficient charge transport with less recombination. On the other hand, the short lifetime (few days) of the CCPD remains the severe bottleneck in commercial application. In order to improve the lifetime, further studies will focus on improving the stability and reproducibility of the CCPD.

| Active materials | Photoresponsivity (A W$^{-1}$) | Rise time (ms) | Fall time (ms) | Ref. |
|------------------|--------------------------------|----------------|----------------|-----|
| CH$_3$NH$_3$PbI$_{3-x}$Cl$_x$ | 620                            | 200            | 100            | [47]|
| CsPbBr$_3$ QDs   | 0.005                          | 0.20           | 1.30           | [48]|
| CH$_3$NH$_3$PbI$_{3-x}$Cl$_x$/CsPbBr$_3$ | 0.39                          | 0.121          | 0.107          | This work |

**Table 1** Comparison of the device performances of CH$_3$NH$_3$PbI$_{3-x}$Cl$_x$/CsPbBr$_3$ device with its single counterparts

![Fig. 5 Schematic of the band diagram of hybrid perovskite/CsPbBr3 heterostructure](image)
on the understanding of ligand effects in the hybrid perovskite-quantum dot devices.

Additional file

**Additional file 1:** Figure S1. Tauc equation plot. Figure S2. PL intensity and absorption curve of CsPbBr3 QDs. (DOCX 421 kb)

**Abbreviations**

PDs: Photodetectors; CPD: CH3NH3PbI3−xClx; perovskite photodetector; CCGP: CH3NH3PbI3−xClx–CsPbBr3 photodetector; QDs: Quantum dots; FETs: Field-effect transistors; TEM: Transmission electron microscopy; SEM: Cross-sectional scanning electron morphology; XRD: X-ray diffraction; NEP: Noise equivalent power; G: Gain

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**Authors’ Contributions**

YY and XZ worked on device fabrication. YZ, DL, KW, and FY performed the measurements and analyzed the data presented in the paper. HD, XWS, and JY initiated the research. YY wrote the manuscript. All authors read and approved the final manuscript.

**Availability of Data and Materials**

The conclusions made in this manuscript are based on the data (main text and figures) presented and shown in this paper.

**Competing Interests**

The authors declare that they have no competing interests.

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