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Broadband Phototransistor Based on CH$_3$NH$_3$PbI$_3$ Perovskite and PbSe Quantum Dot Heterojunction

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ABSTRACT: Organic lead halide perovskites have received a huge amount of interest since emergence, because of tremendous potential applications in optoelectronic devices. Here field effect phototransistors (FEpTs) based on CH$_3$NH$_3$PbI$_3$ perovskite / PbSe colloidal quantum dot heterostructure are demonstrated. The high light absorption and optoelectric conversion efficiency, due to the combination of perovskite and quantum dots, maintain the responsivities in a high level, especially at 460 nm up to 1.2 A/W. The phototransistor exhibits bipolar behaviors and the carrier mobilities are determined to be 0.147 cm$^2$V$^{-1}$s$^{-1}$ for holes and 0.16 cm$^2$V$^{-1}$s$^{-1}$ for electrons. The device has a wide spectral response spectrum ranging from 300 nm to 1500 nm. A short photoresponse time is less than 3 ms due to the assistance of heterojunction on the transfer of photoexcitons. The excellent performances presented in the device especially emphasize CH$_3$NH$_3$PbI$_3$ perovskite-PbSe quantum dot as a promising material for future photoelectronic applications.
The broadband photodetectors have recently aroused substantial interests in plenty of fields, such as day and night monitoring, chemical/biological sensing, environmental monitoring, and optical communications. Traditionally, broadband detection spectrum ranges from the ultraviolet (UV)-visible to the near infrared (NIR) wavelength. Commercial photodetectors are fabricated with gallium nitride (GaN), silicon (Si) and indium-gallium-arsenide (InGaAs) for sensing UV, visible and NIR light, respectively. Due to lacking suitable materials with the ability to absorb incident light over a wide range of wavelengths, the development of broadband photodetectors is restricted.

As one type of promising materials for broadband photodetection, organic lead halide perovskites (CH$_3$NH$_3$PbX$_3$, X=Cl, Br, I) have been investigated due to its ability to absorb light over a wide wavelength ranging from UV to visible and their suitable, direct bandgap with large absorption coefficients, as well as long electron-hole diffusion lengths and carrier lifetimes. Furthermore, on the basis of simple and cost-effective considerations, perovskites can be synthesized by a chemical solution-based self-assembly method. Various types of hybrid semiconductors combined of perovskite have been investigated as channel materials. For instance, the broad range of wavelengths (254 ~ 800 nm) detection, fast response (2.5 ns rise/ 30 ns fall based on a 337.1 nm irradiance laser), and device stability were reported through a perovskite photodetector by spin-coating with a Perfluorinated cyclic polymers (CYTOP) film. A flexible phototransistor fabricated by the active layer of CH$_3$NH$_3$PbI$_3$-graphene heterojunction achieved a wide...
photoresponse of 400 ~ 1000 nm\textsuperscript{18}. Although perovskite photodetectors presented high responsivity, short photoresponse time, the detection wavelength regions are still narrow—only from UV to visible, can’t involve infrared (IR).

To expand the wavelength range of operation, efforts should be devoted to design the photodetectors through the combination of perovskite and other materials with wide spectral sensitivity. Exhibiting tunable bandgap from 0.5 to 1.7 eV\textsuperscript{19} matching perovskite’s energy level and high air stability with organic ligands, PbSe colloidal quantum dots (CQDs)\textsuperscript{20-23} have a significant absorption capacity for NIR and short-wavelength light, which perovskites can’t be access to. Ning et al.\textsuperscript{24} implemented the excellent photo-carrier diffusion process in the perovskites, and the bright light can be emitted from infrared-bandgap quantum-tuned materials. An effective and solution-processable perovskite-PbSe phototransistor will exhibit excellent performances\textsuperscript{25-28} extending to NIR wavelength due to the combination of strong absorption efficiency of PbSe quantum dots and long distance carrier transmission in perovskite matrix. As our knowledge, phototransistors based on CH\textsubscript{3}NH\textsubscript{3}PbI\textsubscript{3} perovskite / PbSe colloidal quantum dots heterostructure have never been reported.

In this paper, we fabricate CH\textsubscript{3}NH\textsubscript{3}PbI\textsubscript{3} perovskite-PbSe QDs hybrid phototransistors through solution-processed strategy. Due to the appropriate bandgap and large absorption coefficient, CH\textsubscript{3}NH\textsubscript{3}PbI\textsubscript{3}-PbSe QDs with a wide absorption wavelength from UV to infrared light are desirable for fabricating broadband photodetectors. The CH\textsubscript{3}NH\textsubscript{3}PbI\textsubscript{3}-PbSe phototransistor exhibits wide photodetection
region ranging from 300 nm to 1500 nm, high sensitivity \((R = 1.2 \text{ A/W})\), high carrier mobility \((\mu_n = 0.147 \text{ cm}^2\text{V}^{-1}\text{s}^{-1} \text{ and } \mu_p = 0.16 \text{ cm}^2\text{V}^{-1}\text{s}^{-1})\), a fast response times (rise time: 2.5 ms and fall time: 3 ms), and excellent stability and reproducibility. Therefore, these high performances indicate that solution-based process perovskite-PbSe colloidal quantum dots heterostructures have great potential for ultra-broadband photodetection applications, especially in the IR region.

The phototransistor architecture was fabricated with the configuration shown in Figure 1a. For the bottom-gate configuration, a silicon wafer \((n^+ \text{ Si})\) with a 300 nm thick SiO\(_2\) layer (capacitance \(C_{ox}\) of 11.5 nFcm\(^{-2}\)) was used as gate electrode. A perovskite thin film with an active channel was made by two step spin-coating solution (see Supporting Information (SI) for details). Source and drain electrodes were thermally evaporated through a sophisticated shadow mask with the channel length \((L = 0.1 \text{ mm})\) and channel width \((W = 2.5 \text{ mm})\), respectively. Finally, a spin-coated film of PbSe Quantum dots was covered on the top. The preparation and characterization of PbSe QDs are referred to SI. Figure 1b depicts the cross-sectional scanning electron microscopy (SEM) image of the device with the scale of 500 nm. It described that the thickness of SiO\(_2\) is 300 nm, then the perovskite film grown on SiO\(_2\) has the average thickness of ~ 75 nm and the thickness of PbSe QDs film spinning on perovskite is ~ 177 nm. It is observed that the interface between CH\(_3\)NH\(_3\)PbI\(_3\) perovskite and PbSe QDs is clear and no intermediate layer, which is essential to obtain excellent photoelectric properties. Notably, the thickness of active layer is a key parameter. If the layer is too thin (40 nm ~70 nm for our devices) to
effectively absorb light \(^{29}\). Meanwhile, thin perovskite films are prone to produce pinholes, leading to inhomogeneous conduction in the channel. Furthermore, if the film is too thick (> 80 nm for our devices), and the channel are not efficaciously modulated by the bottom gate. Therefore, we utilized the optimized thickness (~75 nm) of the CH\(_3\)NH\(_3\)PbI\(_3\) film. The X-ray diffraction (XRD) spectrum of the perovskite film synthesized on a glass substrate is depicted in Figure 1c. From XRD spectrum, four characteristic peaks centered at 14.09°, 28.53°, 31.9°, 43.04° are assigned to

![Figure 1](image-url)

**Figure 1.** (a) Device schematic diagram. (b) Cross-sectional SEM image of the device with the scale of 500 nm. (c) X-ray diffraction spectrum of the
CH$_3$NH$_3$PbI$_3$ film, (inset: the SEM image top-view of perovskite on silicon substrate). (d) Optical absorption spectrum of hybrid including perovskite and PbSe quantum dots on glass substrate.

the 110, 220, 310 and 330 planes of a tetragonal perovskite crystal structure with lattice parameter of a=b=8.849 Å, c=12.64 Å, which is consistent with literatures reported$^{13,30-33}$. Meanwhile, it is obvious that CH$_3$NH$_3$PbI$_3$ exhibits well-defined cuboid shapes and the surface morphologies are homogenous in the inset of Figure 1c, and few distinct pores exist in the grain boundary benefitting that light penetrates through the whole film$^{34}$. The absorption region ranges from visible to near-infrared wavelength, depicted in figure 1d, indicating that this device can be used for broadband spectrum detection. The hybrid shows two absorption peaks —‘547 nm’ and ‘1160 nm’—between 300nm and 1500 nm. For electric measurements, drain and source electrodes (ground connection) were connected with tow output ports of Keithley 2400 forming bias voltage ($V_{DS}$). The drain source current ($I_{DS}$) was also measured by Keithley 2400. And the gate electrode was connected with a constant voltage source HP6030A. Based on this system, the photoelectrical measurements were also performed under illumination of 405 nm, 532 nm and 808 nm lasers.

Figure 2a describes the $I$-$V$ characteristics of different gate voltages (0 V, ±1 V, ±2 V, ±3 V, ±4 V, ±5 V) under 808 nm laser with irradiance of 845 mW cm$^{-2}$ (solid lines) and without irradiation (dashed lines). If $V_{GS}$ and $V_{DS}$ are both negative, the device operates in the hole-enhancement mode, while both are positive, the device operates in the electron-enhancement mode. Specially, at low $|V_{GS}|$ (Off-state), $I_{DS}$
increases rapidly with the increase of $V_{DS}$, indicating that a Schottky barrier forms in the device. However, at high $|V_{GS}|$ (ON-state), similar to traditional field effect transistors (FETs) with linear-to-saturation current-voltage characteristics, the device exhibits unipolar transport properties. Comparing with the condition with no irradiation, the values of $|I_{DS}|$ become larger. It can be understood that the photo-induced carriers can be generated under the condition of light irradiation. Therefore the enhancements of $I_{DS}$ are ascribed to the increase of the electron and hole concentrations. It can be indicated that the thermionic and tunneling currents dominate the vital role. Figure 2b shows transfer characteristics of a bottom-gate CH$_3$NH$_3$PbI$_3$ phototransistor. The device exhibits typically ambipolar characteristics, and a “V” shape is particularly presented in the transfer curves, manifesting either electrons or holes transport in the n-type or p-type channels of the device, respectively. The mechanism can be interpreted that owing to the difference of electron potentials, some electrons are transferred from CH$_3$NH$_3$PbI$_3$ to PbSe, resulting in the formation of a built-in-electric field in the surface that points toward PbSe. Accordingly, holes separated from photoexcitons generated in heterojunction tend to reside in the poverskite layer due to the assistance of the built-in electric field. Therefore, the shift toward positive $V_{GS}$ (shown in figure 2b) can be ascribed to the efficient photoexcitons separation and holes transport at the interface. Meanwhile, in the linear region, the relationship between field-effect mobility and gate voltage can be extracted with the equation of

$$\mu = \frac{L}{V_{DS} C_{ox} W} \frac{\partial I_{DS}}{\partial V_{GS}},$$

(1)
where $L$ and $W$ are the length and width of the channel, respectively, $C_{ox}$ is the capacitance per area. Therefore, both the electron and hole mobilities can be calculated as $0.147 \, \text{cm}^2\text{V}^{-1}\text{s}^{-1}$ and $0.16 \, \text{cm}^2\text{V}^{-1}\text{s}^{-1}$, which are larger one order of magnitude than prior studies in pristine PbSe QDs phototransistor ($0.01$ and $0.03 \, \text{cm}^2\text{V}^{-1}\text{s}^{-1}$) \cite{38}, respectively. It is attributed to the ‘quantum-dot-in-perovskite’ \cite{24} at the interface, which PbSe QDs are effectively passivated by perovskite instead of conventional organic ligands (i.e. ethanedithiol, formic acid, hydrazine), providing a superior diffusion length.

**Figure 2.** Electrical properties of the perovskite phototransistor. (a) Output characteristic ($I_{DS}$ versus $V_{DS}$ at different $V_{GS}$) under 808 nm laser with irradiance of 845 mW/cm$^2$ (solid lines) and in the darkness (dashed lines). (b) Transfer characteristics ($I_{DS}$ versus $V_{GS}$) of the device under light condition or not at $V_{DS} = -0.5 \, \text{V}$, respectively.

In order to investigate wide spectral response, the dark currents and photocurrents are measured by varying the wide range of wavelengths under the same irradiation. Photoresponsivity ($R$) is calculated by
\[ R = \frac{\Delta I_{DS}}{P} = \frac{I_{ill - I_{dark}}}{E_e \times S}, \]  

(2)

where \( I_{ill} \) and \( I_{dark} \) represent the drain current under illumination and in the darkness. \( P, E_e \) and \( S \) are power density, irradiance and the effective channel area, respectively.

The photoresponsivity and detectivity with the relationship of wavelength in the device is shown in figure 3a. The device exhibits a broadband photoresponse (300 nm ~ 1500 nm) and is suitable for visible light to near-infrared light photodetection, which is determined by the absorption curve of hybrid film in Figure 1 d. The maximum \( R \) is calculated to 1.2 A/W at 300 nm, and other key parameters characterizing photodetector’s performance, such as the detectivity (D*), the noise equivalent power (NEP), the gain (G) can be given as

\[ D^* = RA^{1/2}/(2eI_{DS})^{1/2}, \]

\[ NEP = A^{1/2}/D^* \]

and \[ G = \frac{hv}{e} R, \]

respectively, where \( R, A, e, I_{DS} \) are the responsivity, the channel area of the device, the charge of an electron, and the dark current, respectively. Particularly, if maximum photoresponsivity (\( R \)) is set to 1.2 A/W at 300 nm, and the detectivity has been obtained and impressive high value of \( 3.3 \times 10^7 \) Jones, indicating that the proposed device is extremely sensitive to incident illumination. The \( NEP \) and \( G \) of the device (shown in Figure S5) are \( 1.5 \times 10^{-11} \) W/Hz and 4.96, respectively. Meanwhile, the phototransistor exhibits stability and reproducibility in the progress of on-off cycles shown in the inset of Figure 3a.

Comparing with different photocurrents excited by different wavelength lights at the same power density, the photocurrents excited by 405 nm light, 532 nm and 808 nm are ~ 5 \( \mu \)A, ~ 2.3 \( \mu \)A, and ~ 1 \( \mu \)A, respectively. It is reasonable since the photocurrent generation needs to match the basic condition, that is, the incident photo energy must
be larger than the energy gap. Only these incident photos with enough photon energy can excite electrons from the valance band maximum (VBM) to the conduction band minimum (CBM), generating the photocurrent if the $V_{DS}$ is applied. Therefore, more electrons and photoexcitons can be excited with larger incident photo energy lights in the short wavelength and contribute more to photocurrent. It indicates that electrons must pass through the Schottky barrier to transport. Under the irradiation of larger energy light in the shorter wavelength, Schottky barrier for built-in potential is reduced, leading to a huge enhancement in the density of free carrier, which in turn results in easier carriers transport and tunneling, thus, to greatly enhanced photocurrents.

Figure 3b exhibits the photocurrent and responsivity ($R$) of the device with the relation of irradiance ($E_e$), in which the photocurrent increases linearly with $E_e$ at low light irradiance, while it deviates below this linearity at higher light irradiation. It is concluded that with larger irradiance more photoexcitons and electrons can be excited, leading to the decrease of built in potential of the Schottky barrier, which have a higher probability to overpass the Schottky barrier, and then linearly enhanced photocurrents are generated during the process of the carriers flow to the external circuit. However, for higher light irradiances, some photoexcitons remained in the trap states $^{11}$ of perovskite, thus cannot be converted to the photocurrent, leading to the saturation of photocurrent. As for $R$, the $R$ decreases if $E_e$ increases in a reciprocal function of $R = a/\left(1 + (E_e/b)^n\right)$, where a and b are constants and n is a logistic fitting parameter. Based on the theory $^{40-42}$
where $e$ is the charge of single electron, $h\nu$ is the single photon energy, $T_0$ is the carrier lifetime, $T_r$ is the carrier transfer time, $P_0$ is the excitation intensity where the surface states are fully filled, and $n$ is a phenomenological fitting parameter. The red solid line in the inset of figure 3b is the best fit to the data obtained by equation (3) from which $P_0 = 2.42 \times 10^{-6}$ W, and $n = 0.54$ have been deduced.

\[ R = \left( \frac{e}{h \nu} \frac{T_0}{T_r} \right) \frac{1}{1 + \left( \frac{P}{P_0} \right)^n} \]  

(3)

Figure 3. (a) The photoresponsivity ($R$) and detectivity ($D^*$) of the device with the relationship of wavelength ranging from 300 nm to 1500 nm at $V_{DS} = 2$ V and $V_{GS} = 0$ V. (Inset: Switching behavior of the perovskite phototransistor at the irradiance of 50 mW/cm$^2$ under various illuminations of 405 nm, 532 nm, and 808 nm.) (b) $R$ and photocurrent with the relationship of $E_e$, and the red solid line accounts for the reciprocal function fitting.

Photoresponse speed determining the capability of a photodetector to follow a fast-switching optical signal is another key factor for photodetector. In order to highlight the photoresponse of the device during IR spectrum, initially we exploit
pump laser with an illumination wavelength of 808 nm — below the absorption edge of PbSe (shown in figure 1 d). Figure 4a shows drain current response of the device with light irradiance of 845 mW/cm², exhibiting stable and reproducible photoresponse. The drain current quickly increases rapidly once the light is turned on and then decreases as the light switch is off. It indicates that the increased charge intensity will lower the effective barrier height upon illumination, which allows easier charge tunneling and transportation than that of device in darkness.

Figure 4b depicts the time-photocurrent response of the device. The rise and decay times of the photocurrent are ~ 2.5 ms and 3 ms, respectively. Recently, using pristine perovskite, Li et al. reported switching times of phototransistors are ~ 6.5 and 6.0 μs. It is reasonable that the speed is probably confined by the lower charger mobility of the PbSe QDs layer (carrier mobilities ~ 0.03 (hole) cm²V⁻¹s⁻¹ and ~ 0.01 (electron) cm²V⁻¹s⁻¹) and some photo carriers remain trapped in the perovskite channel, leading to the increasement of carrier lifetime. To demonstrate the groundbreaking improvements obtained by this hybrid, we fabricated another two kinds of phototransistors, with similar method and CH₃NH₃PbI₃ perovskite as the only active layer (Figure S3, Supporting Information) or PbSe quantum dots as the active layer (Figure S4, Supporting Information). To compare with characteristics based on CH₃NH₃PbI₃ perovskite and PbSe QDs alone, several key parameters are list in Table 1.
Figure 4. Photoresponse characteristics of perovskite phototransistor under irradiation with red light (λ=808 nm). (a) Current response of the device with a red light irradiance of 845 mW/cm². (b) Time dependent photocurrent responses, indicating that a rise time of 2.5 ms and a fall time of 3 ms.

Table 1 Comparison in device performance of CH$_3$NH$_3$PbI$_3$ / PbSe hybrid FEpTs with its single counterparts.

<table>
<thead>
<tr>
<th>Device</th>
<th>Charge mobility [cm$^2$V$^{-1}$s$^{-1}$]</th>
<th>Spectral coverage [nm]</th>
<th>Rise / fall time to 808 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH$_3$NH$_3$PbI$_3$</td>
<td>0.29 / 0.38</td>
<td>300–730</td>
<td>—</td>
</tr>
<tr>
<td>PbSe</td>
<td>0.01 / 0.03</td>
<td>300–1500</td>
<td>10.9 s / 8.3 s</td>
</tr>
<tr>
<td>CH$_3$NH$_3$PbI$_3$ / PbSe</td>
<td>0.147 / 0.16</td>
<td>300–1500</td>
<td>2.5 ms / 3ms</td>
</tr>
</tbody>
</table>

The working principle of this phototransistor is schematically depicted in Figure 5. Upon light illumination, the photocurrents can be generated under the condition that incident photon energy must be larger than bandgap of perovskite, which is 1.5 eV$^{43}$, that is, 826 nm in wavelength. While for PbSe, the bandgap is 0.95 eV, that is, 1.3 μm
in wavelength. Only these incident photons with larger energy \( (h\nu > 1.5 \text{ eV}) \), that is, wavelength < 826 nm) can excite electrons from the valance band maximum (VBM) to the conduction band minimum (CBM) in the perovskite, generating the photocurrent if the drain voltage is applied. Given the CBM of PbSe is lower than that of perovskite, photo-generated electrons can roll down to the CBM of PbSe. However, the VBM of perovskite is larger than that of PbSe, so, the photo-generated holes in PbSe can be energetically transferred to perovskite. Due to the larger hole mobility in perovskite, the holes drift faster in perovskite than that in PbSe, and can be rapidly collected by the electrodes. And the charges transfer at the PbSe / perovskite interface, leading to the formation of depletion layer. And the depletion layer assists in the charge separation. Meanwhile, three occasions need to discuss due to different energy gaps \( (E_g) \) of perovskite and PbSe. If \( h\nu \) is larger than 1.5 eV, shown in figure 5a, electron-hole pairs are generated in perovskite and PbSe films, and the electrons (holes) are transferred to PbSe (perovskite) film, which lowers (raises) the Fermi level of PbSe (perovskite) and reduces the Schottky barriers, resulting in the high photocurrent. While \( h\nu \) is larger than 0.95 eV smaller than 1.5 eV, shown in figure 5b, electron-hole pairs can be excited only in PbSe film. Therefore, the photocurrents become small. And if \( h\nu \) is smaller than 0.95 eV, none electron-hole pairs are generated so the photocurrent approximately equals dark current.
In summary, broadband phototransistors based on CH$_3$NH$_3$PbI$_3$ perovskite-PbSe colloidal quantum dots heterostructures were manufactured in a cost-effective, solution-processed strategy. The phototransistor exhibits bipolar behaviors and a wide spectral response ranging from 300 nm to 1500 nm. Particularly, the phototransistor is photoresponsive to the UV (405 nm), VIS (532 nm) and NIR (808 nm) irradiations, and presents stability and reproducibility in the progress of ON-OFF cycles. Carrier mobilities of CH$_3$NH$_3$PbI$_3$-PbSe heterojunction photodetector are measured as 0.147 cm$^2$V$^{-1}$s$^{-1}$ for holes and 0.16 cm$^2$V$^{-1}$s$^{-1}$ for electrons. It is worth mentioning that the fast rise and decay times of the photocurrent are $\sim$ 2.5 ms and 3 ms, respectively. Therefore, these results indicate that solution-based process perovskite-PbSe colloidal quantum dots heterostructures have great potential applications for ultra-broadband photodetection, especially in the IR region.
Phototransistor Fabrication: The CH$_3$NH$_3$PbI$_3$ perovskite thin film with an active channel made by two step spin-coating solution (see Supporting Information (SI) for details) was deposited onto a silicon wafer ($n^+$ Si) covered with a 300 nm thick SiO$_2$ layer. As source and drain electrodes, Au was thermally evaporated through a sophisticated shadow mask at a rate of 1 Å s$^{-1}$ in the pressure of $3.8 \times 10^{-4}$ Pa. The detail process of spin-coated film of PbSe QDs was referred to Ref [44]. All the measurements were performed in air at room temperature.

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Notes

The authors declare no competing financial interest.

ASSOCIATED CONTENT

Supporting Information. The characterizations and synthesis of CH$_3$NH$_3$PbI$_3$ perovskite and PbSe QDs, characteristics of CH$_3$NH$_3$PbI$_3$ perovskite and PbSe QDs phototransistsors, and stability of the CH$_3$NH$_3$PbI$_3$ perovskite / PbSe QDs FEpTs.

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