

Ultrasensitive and high gain solution-processed perovskite photodetectors by $\text{CH}_3\text{NH}_3\text{PbI}_{2.55}\text{Br}_{0.45}:\text{Zn}_2\text{SnO}_4$ bulk heterojunction composite

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Ultrasensitive and high gain solution-processed perovskite photodetectors by $\text{CH}_3\text{NH}_3\text{PbI}_{2.55}\text{Br}_{0.45}:\text{Zn}_2\text{SnO}_4$ bulk heterojunction composite

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Abstract

Hybrid perovskite photodetectors, the novel alternative devices transform incident light into electrical signal, have been rapidly developed in the past years. However, intrinsic unbalanced charge carrier transport within hybrid perovskite materials, restricting further boosting device performance of perovskite photodetectors, has rarely been addressed. In this study, we report room temperature-operated solution-processed bulk heterojunction perovskite photodetectors with ultrahigh sensitivity and great photo-gain. It is found that the introduction of Zn_2SnO_4 nanoparticles into $\text{CH}_3\text{NH}_3\text{PbI}_{2.55}\text{Br}_{0.45}$ perovskites results in enlarged photocurrent and suppressed dark current for bulk heterojunction perovskite photodetectors. As a result, a responsivity of over 1500 mA W^{-1} and projected detectivity of approximately 10^{14} Jones ($1 \text{ Jones} = \text{cm Hz}^{1/2} \text{ W}^{-1}$) from 380 to 760 nm are observed from bulk heterojunction perovskite photodetectors. In addition, bulk heterojunction perovskite photodetectors exhibit an excellent linear dynamic range of 124 dB and a great photo-gain of 535. All these results indicate that high-performance perovskite photodetectors could be realized through novel bulk heterojunction device structure.

Keywords Perovskite photodetectors · Zn_2SnO_4 nanoparticles · High gain · High responsivity and detectivity

1 Introduction

Photodetectors (PDs) that transform incident light into electrical signal show great potential applications of light-signal detection including environmental monitoring, medical sensing, optical fiber communication, day and night surveillance, and light imaging [1–5]. Nowadays, various inorganic and organic

semiconductors have been greatly developed for fabrication of PDs. However, the high cost of inorganic-based PDs and their requirements to be operated at low temperature and high voltage for receiving reasonable detectivities restricted their large applications [6, 7]. Organic-based PDs possess cost-effective processability and room-temperature operational feature, but most of them exhibit low sensitivity due to inefficient charge transfer and low charge carrier mobilities, and poor stability owing to intrinsic instability of organic semiconductors [8–13].

Recently, organic-inorganic hybrid halide perovskites have been demonstrated to be the photoactive layer for photovoltaics (solar cells and photodetectors) due to its superior optoelectronic properties and solution processability [14–19]. In 2014, Dou et al. reported solution-processed perovskite PDs with an inverted device structure [20]. In 2015, we reported high-performance solution-processed perovskite PDs with a conventional device structure [21]. Recently, ultraviolet (UV)-visible to near-infrared (NIR) photoresponsivity from perovskite PDs, where perovskite materials were incorporated with either low-bandgap conjugated polymers or inorganic quantum dots (QDs), was reported [16, 22–24]. For example, by forming the double-layer structure of $\text{CH}_3\text{NH}_3\text{PbI}_3/\text{NDI}$ -

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DPP (where NDI-DPP is poly[(*N,N'*-bis(2-octyldodecyl)-1,4,5,8-naphthalene diimide-2,6-diyl) (2,5-dioctyl-3,6-di(thiophen-2-yl)pyrrolo [3,4-*c*]pyrrole-1,4-dione-5,5'-diyl)], an n-type narrow bandgap conjugated polymer) as the photoactive layer, solution-processed perovskite PDs with responsivity (*R*) of 150 mA W⁻¹ and projected detectivity (*D*^{*}) over 10¹² Jones (1 Jones = cm Hz^{1/2} W⁻¹) in the NIR region were observed [16]. Perovskite PDs have been rapidly developed [20–26]; however, intrinsic unbalanced charge carrier transport within perovskite materials [27, 28], which restricts further boosting device performance of perovskite PDs, has not been fully addressed so far. In order to address unbalanced charge carrier transport within perovskite materials, bulk heterojunction (BHJ) device structure was firstly reported in our group for approaching efficient perovskite solar cells (PSCs) [29, 30]. As alternatives to construct BHJ composite with perovskite, metal oxide nanoparticles (NPs) exhibit unique optoelectrical properties [31–40]. More recently, we reported high-performance PSCs, where perovskite materials were incorporated with n-type Zn₂SnO₄ NPs, which possess superior charge carrier mobility, as the photoactive layer [41].

In this study, we report a simple and versatile solution method for approaching ultrasensitive solution-processed perovskite PDs with BHJ device structure. Operated at room temperature, it is found that BHJ perovskite PDs exhibit a responsivity of over 1500 mA W⁻¹ and projected detectivity of approximately 10¹⁴ Jones from 380 to 760 nm. In addition, BHJ perovskite PDs possess excellent linear dynamic range of 124 dB and a great photo-gain of 535.

2 Results and discussion

Perovskite PDs with a BHJ device structure of ITO/PTAA/CH₃NH₃PbI_{2.55}Br_{0.45}:Zn₂SnO₄/PC₆₁BM/Al, and a planar heterojunction (PHJ) device structure of ITO/PTAA/CH₃NH₃PbI_{2.55}Br_{0.45}/PC₆₁BM/Al, where ITO is indium tin oxide, PTAA is polytriarylamine and acts as the hole extraction layer (HEL), PC₆₁BM is [6,6]-phenyl-C61-butyric acid methyl ester and acts as the electron extraction layer (EEL), and Al is aluminum, respectively, were fabricated and characterized. The device structures for perovskite PDs under this study are the same as the device structures for PSCs reported previously [41]. Based on the band alignment of the materials and the electrodes used for fabrication BHJ perovskite PDs [41], the PTAA HEL is favorable for holes being extracted from CH₃NH₃PbI_{2.55}Br_{0.45}:Zn₂SnO₄ BHJ composite to the ITO anode, which enhances the photocurrent density. The conduction band of Zn₂SnO₄ NPs is −4.10 eV, which is located at between the lowest unoccupied molecular orbit (LUMO) energy level (−3.79–3.93 eV) of CH₃NH₃PbI_{2.55}Br_{0.45} and the LUMO energy (−4.20 eV) of the PC₆₁BM EEL, forming the gradient energy level difference, and further

reducing the direct energy loss. Owing to the deep valence band maximum (−7.90 eV) of Zn₂SnO₄ NPs, it markedly increases the hole injection barrier between the Al cathode and CH₃NH₃PbI_{2.55}Br_{0.45}:Zn₂SnO₄ BHJ composite from 1.20 to 3.60 eV, which is more efficient to block holes being injected into CH₃NH₃PbI_{2.55}Br_{0.45}:Zn₂SnO₄ BHJ composite in dark from the external circuit, and thus, reduced dark current density is expected. Therefore, boosted photocurrent density and suppressed dark current density are expected to be observed from perovskite PDs with a BHJ device structure.

Operated at room temperature, the current density versus voltage (*J*-*V*) characteristics of perovskite PDs with either BHJ or PHJ device structures measured in dark and under a monochromatic light of 500 nm with a light intensity of 0.28 mW/cm² are shown in Fig. 1a. It is obvious that perovskite PDs with a BHJ device structure exhibit dramatically reduced dark current density at reverse bias compared with

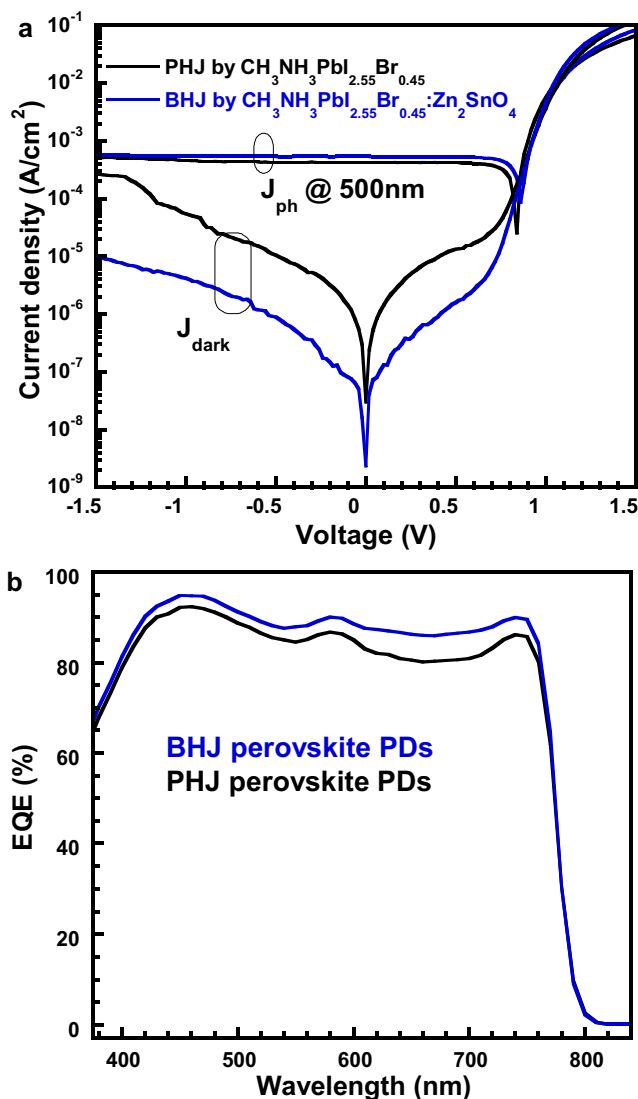


Fig. 1 a The *J*-*V* characteristics and b EQE spectra of both BHJ and PHJ perovskite photodetectors

those from perovskite PDs with a PHJ device structure, i.e., biased at -0.5 V; a dark current density of $8.97 \times 10^{-7} \text{ A cm}^{-2}$ observed from BHJ perovskite PDs is much smaller than that of $1.05 \times 10^{-5} \text{ A cm}^{-2}$ observed from PHJ perovskite PDs, indicating the small amount of Zn_2SnO_4 NPs would suppress leakage current. Such suppressed dark current density is attributed to the high-quality thin film morphology with diminished pinholes and grain boundaries of solution-processed $\text{CH}_3\text{NH}_3\text{PbI}_{2.55}\text{Br}_{0.45}:\text{Zn}_2\text{SnO}_4$ BHJ composite thin film as compared with that of solution-processed pristine $\text{CH}_3\text{NH}_3\text{PbI}_{2.55}\text{Br}_{0.45}$ thin film [41], and inhibition of charge injection from electrodes as discussed above. On the other hand, under monochromatic light of 500 nm , the photocurrent density of BHJ perovskite PDs is slightly higher than that from PHJ perovskite PDs, indicating that Zn_2SnO_4 NPs is functionalized as the electron acceptor, boosting separated charge carrier to be efficiently transferred to the Al cathode [41]. Enhanced photocurrent and suppressed dark current indicate that BHJ perovskite PDs possess boosted detectivity [2].

The external quantum efficiency (EQE) spectra of both PHJ and BHJ perovskite PDs are shown in Fig. 1b. These EQE spectra are nearly the same as the EQE spectra of PSCs reported previously [41]. Both PHJ and BHJ perovskite PDs possess the identical photoresponse since Zn_2SnO_4 NPs is a wide bandgap semiconductor and it has no contribution to the photocurrent over the wavelength (λ) of 375 nm . However, as indicated in Fig. 1b, an enhancement in EQE is observed from 380 to 760 nm from BHJ perovskite PDs as compared with that of PHJ perovskite PDs, indicating that BHJ perovskite PDs generate more photocurrent than that of PHJ perovskite PDs. These observations are in good agreement with the J-V characteristics (Fig. 1a).

Thus, Zn_2SnO_4 in the $\text{CH}_3\text{NH}_3\text{PbI}_{2.55}\text{Br}_{0.45}:\text{Zn}_2\text{SnO}_4$ BHJ composite thin film plays two functionalities. Firstly, Zn_2SnO_4 NPs serve as the electron acceptor to facilitate the charge carrier separation and transport in PDs under illumination. The n-type Zn_2SnO_4 NPs and p-type perovskite could form p-n junction at the interface and benefit the separation of photogenerated electron-hole pairs, resulting in more extracted charge density as well as boosted electron mobility [41]. Thus, the boosted photocurrent is observed in PDs by $\text{CH}_3\text{NH}_3\text{PbI}_{2.55}\text{Br}_{0.45}:\text{Zn}_2\text{SnO}_4$ BHJ composite thin film. Secondly, Zn_2SnO_4 NPs act as nucleation center for the growth of perovskite crystal and help to improve the quality of resultant thin film with better uniformity, enlarged grain size, and suppressed pinholes and grain boundaries [41]. Such optimal thin film morphology delivers the reduced dark current in corresponding PDs.

Responsivity (R), one of the most important parameters for evaluating PDs performance, is described as [2, 42]:

$$R = \frac{J_{\text{ph}}}{L_{\text{light}}} \text{ or } R = \text{EQE} \times \frac{e}{h\nu} = \text{EQE} \times \frac{\lambda}{1240} \quad (1)$$

where J_{ph} is photocurrent density, L_{light} is the light intensity, e is the elementary charge, h is the Planck's constant, ν is the frequency of optical signal, and λ is wavelength. Thus, biased at -0.5 V and under illumination at 500 nm with the light intensity of 0.28 mW/cm^2 , the estimated R of 1632 mA/W is for PHJ perovskite PDs, whereas R of 1847 mA/W is for BHJ perovskite PDs.

Detectivity is another most important parameter for evaluating PD performance [2, 28]. Detectivity is dependent on the noise current, which includes shot noise, Johnson noise, and thermal fluctuation "flicker" noise [2, 42, 43]. If only, take the shot noise from the dark current density (J_d) into consideration, the projected detectivity (D^*) is described by [2]:

$$D^* = \frac{\text{EQE} \cdot \lambda}{1240 \sqrt{2eJ_d}} = \frac{R}{\sqrt{2eJ_d}} \quad (2)$$

Thus, biased at -0.5 V and under λ of 500 nm , D^* of $9.09 \times 10^{12} \text{ Jones}$ is estimated for PHJ perovskite PDs. Under the exact conditions, D^* of $6.08 \times 10^{13} \text{ Jones}$ is estimated for BHJ perovskite PDs, which is one of the highest value of PDs based on perovskite materials as listed in Table S1 [19, 21, 23, 44–48].

Based on Eqs. (1) and (2) and the EQE spectra of perovskite PDs (Fig. 1b), R and D^* versus wavelength for both BHJ and PHJ perovskite PDs could be estimated and the results are shown in Fig. 2. As compared with PHJ perovskite PDs, the enlarged R observed from BHJ perovskite PDs is attributed to boosted J_{ph} due to photoinduced charge transfer among $\text{CH}_3\text{NH}_3\text{PbI}_{2.55}\text{Br}_{0.45}:\text{Zn}_2\text{SnO}_4$ BHJ composite thin film [41]; the enhanced D^* observed from BHJ perovskite PDs is attributed to boosted J_{ph} and suppressed J_d as well. The suppressed J_d is originated from superior film morphology induced by introduction the additional Zn_2SnO_4 into $\text{CH}_3\text{NH}_3\text{PbI}_{2.55}\text{Br}_{0.45}$ thin film [41]. Overall, BHJ perovskite PDs with average R of over 1500 mA/W and D^* of

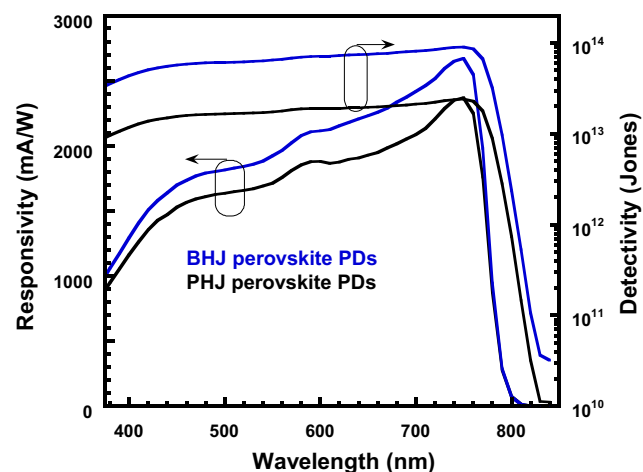


Fig. 2 Responsivity and detectivity versus wavelength for both BHJ and PHJ perovskite photodetectors

approximately 10^{14} Jones are observed in λ ranging from 375 to 760 nm. These device performance parameters are compatible to those Si-based PDs operated at low temperature [49–51].

Linear dynamic region (LDR) is a crucial figure of merit for evaluating PDs. The LDR (typically quoted in dB) is described as [2]:

$$\text{LDR} = 20 \log \frac{J_{\text{ph}}^*}{J_{\text{d}}} \quad (3)$$

where J_{ph}^* is the photocurrent density measured at the light intensity of 1 mW/cm^2 . Figure 3 shows the photocurrent densities versus the light intensities for both PHJ and BHJ perovskite PDs. The LDR for PHJ perovskite PDs is 108 dB, whereas the LDR for BHJ perovskite PDs is 124 dB. Such high LDR is one of the highest values among the reported perovskite PDs [20, 52–55]. Moreover, this value is comparable with commercial Si PDs (120 dB) and higher than other types of PDs such as organic PDs [56, 57].

Fast and reliable responses to light illumination are important to high-performance PDs [42, 43]. The response times, which include rising response to light on and decaying response to light off, are strongly related to the charge transport and collection and describe how sensitive of the PDs towards light

switching on and off. The temporal response of the photocurrent of perovskite PDs is characterized by using an optical chopper controlled at $\lambda = 532\text{-nm}$ laser pulse at a frequency of 1 kHz. The rise time and decay time are defined as the time required for output signals to increase from 10 to 90% of the peak photocurrent and decrease from 90 to 10% of the peak photocurrent [22]. Figure 4 displays the transient photocurrent of both PHJ and BHJ perovskite PDs. The rise times of $11.9 \mu\text{s}$ and $4.2 \mu\text{s}$ are observed from PHJ perovskite PDs and BHJ perovskite PDs, respectively. However, a biexponential photocurrent decay with time constant of $10.4 \mu\text{s}$ for PHJ perovskite PDs is obtained. In contrast, BHJ perovskite PDs exhibit longer decay time of $3.2 \mu\text{s}$. The faster rise time and decay time in PDs by BHJ perovskite illustrate the sensitive response to light switching owing to the improved charge transport and facile charge collection after incorporating with Zn_2SnO_4 NPs.

The photo-gain (PG) of PDs is determined by the ratio recombination lifetime (τ_{life}) to charge transmit time (τ_{transit}), given by [58, 59]:

$$\text{PG} = \frac{\tau_{\text{life}}}{\tau_{\text{transit}}} = \frac{\tau_{\text{life}}}{d^2/\mu V} \quad (4)$$

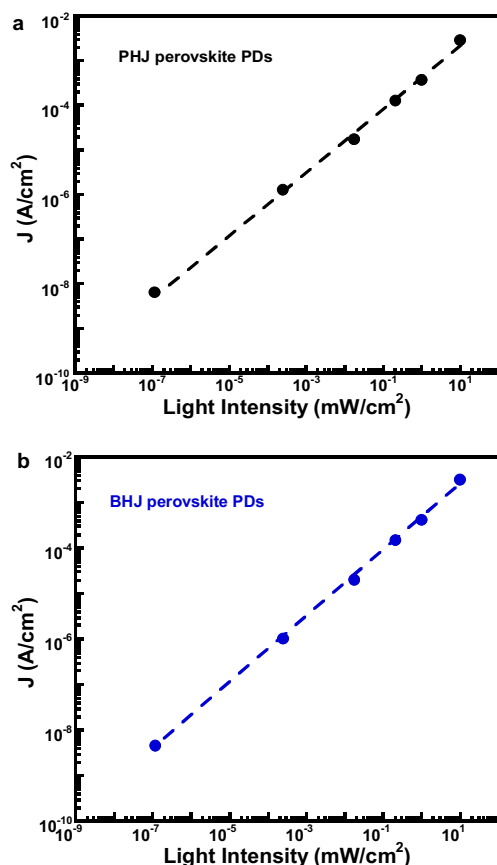


Fig. 3 The photocurrent densities versus the light intensities for **a** PHJ and **b** BHJ perovskite photodetectors

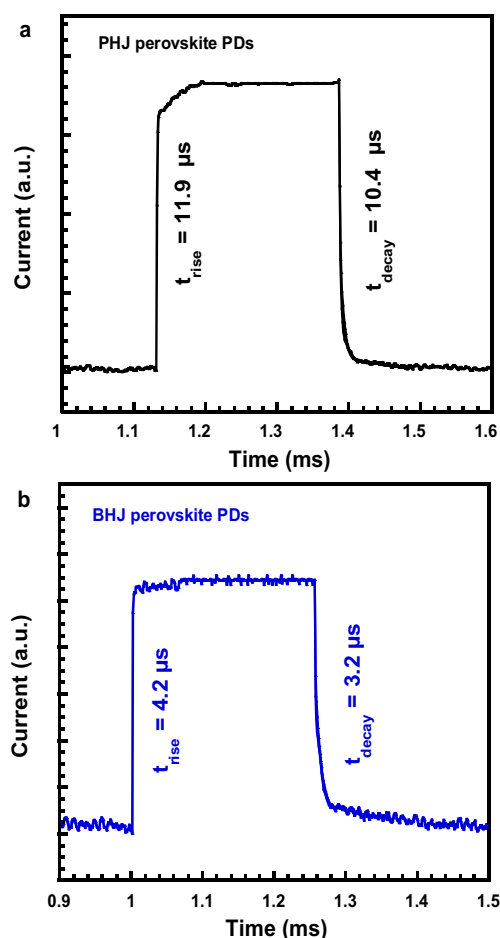


Fig. 4 The transient photocurrent of **a** PHJ and **b** BHJ perovskite photodetectors

where d is the thickness of perovskite thin film, μ is the charge carrier mobility, and V is the applied bias. For PHJ perovskite PDs, at $V = 0.5$ V and $d = 340$ nm, the charge carrier recombination lifetime is estimated to be 8.1 μ s, and the hole mobility is estimated to be 1.16×10^{-2} cm² V⁻¹ s⁻¹ [41]. For BHJ perovskite PDs, at $V = 0.5$ V and $d = 360$ nm, the charge carrier recombination lifetime is estimated to be 16.8 μ s, and the hole mobility is estimated to be 8.26×10^{-1} cm² V⁻¹ s⁻¹ [41]. Thus, the estimated PG values are 43 and 535 for PHJ perovskite PDs and BHJ perovskite PDs, respectively. These results demonstrate that BHJ perovskite PDs could get more PG as compared with that of PHJ perovskite PDs.

In summary, we reported room temperature-operated solution-processed bulk heterojunction (BHJ) perovskite photodetectors (PDs) by CH₃NH₃PbI_{2.55}Br_{0.45} blended with Zn₂SnO₄ NPs. Zn₂SnO₄ NPs acted as the electron acceptors for boosting photocurrent in BHJ perovskite PDs. Zn₂SnO₄ NPs also promotes the formation of high-quality perovskite thin film and generates photoinduced charge transfer, resulting in enlarged photocurrent and suppressed dark current for BHJ perovskite PDs. A responsivity of over 1500 mA W⁻¹ and projected detectivity of approximately 10¹⁴ Jones from 380 to 760 nm, which are compatible to Si-based PDs and higher than those from other perovskite PDs, were observed from BHJ perovskite PDs. In addition, BHJ perovskite PDs exhibit excellent linear dynamic range of 124 dB and great photo-gain of 535. All these device performance parameters are better than those of perovskite PDs with a planar heterojunction device structure. Our discovery reported in this study demonstrates that fabrication of BHJ perovskite PDs is an appropriate approach to realize high-performance perovskite PDs.

3 Experimental

Materials Lead iodide (PbI₂, 99.999%), poly(triaryl amine) (PTAA), methylammonium bromide (MABr), anhydrous *N,N*-dimethylformamide (DMF, 99.8%), anhydrous ethanol (>99.5%), anhydrous toluene (99.8%), anhydrous chlorobenzene (CB, 99.8%), zinc chloride (ZnCl₂), tin (IV) chloride pentahydrate (SnCl₄·5H₂O), zinc acetate dehydrate, and all other reagents were purchased from Sigma-Aldrich and used as received without further purification. [6,6]-Phenyl-C61-butyric acid methyl ester (PC₆₁BM, 99.5%) was purchased from Solenne BV and used as received without further purification. Methylammonium iodide (CH₃NH₃I (MAI)) was synthesized by using hydroiodic acid and methylamine in our laboratory.

Preparation of Zn₂SnO₄ NPs The detailed preparation procedures are described in other literatures [41].

Perovskite precursor preparation PbI₂ was dissolved into DMF solvent with a concentration of 400 mg mL⁻¹, followed by magnetically stirred at 70 °C for ~12 hours (h) until the cloudy solution yields to transparent yellow solution. For BHJ perovskite PDs fabricated by CH₃NH₃PbI_{2.55}Br_{0.45}:Zn₂SnO₄ thin film, PbI₂ was dissolved in mix solution of Zn₂SnO₄ DMF solution where DMF is with a concentration of 400 mg mL⁻¹. Mixture MAI:MABr (85:15 by molar ratio) was dissolved into ethanol solvent to make a concentration of total 35 mg mL⁻¹ solutions.

Fabrication of perovskite photodetectors Approximately 8-nm PTAA layer is spin-coated onto pre-cleaned and UV-Ozone-treated ITO-coated glass substrates from PTAA toluene solution with the concentration of 2 mg mL⁻¹ at 6000 RPM for 40 seconds (s), followed with thermal annealing at 100 °C for 10 min (min) on the hotplate in the glovebox with a nitrogen atmosphere. PbI₂ layer is spin-casted on the top of the pre-heated (85 °C) ITO/PTAA substrates from the warmed (85 °C) PbI₂ solution at a spin speed of 6000 RPM for 20 s with an acceleration time of 5 s, followed with thermal annealing at 85 °C for 10 min on the hotplate, and then cooling down to room temperature. Afterward, the mixture MAI:MABr solution is spin-coated on the top of PbI₂ layer at a spin speed of 6000 RPM for 40 s with an acceleration time of 5 s, followed by thermal annealing at 100 °C for 2 h on the hotplate to form CH₃NH₃PbI_{2.55}Br_{0.45} thin film. For CH₃NH₃PbI_{2.55}Br_{0.45}:Zn₂SnO₄ thin film, PbI₂ is mixed with Zn₂SnO₄ DMF solution. The thickness of CH₃NH₃PbI_{2.55}Br_{0.45} and CH₃NH₃PbI_{2.55}Br_{0.45}:Zn₂SnO₄ thin films are measured to be ~340 nm and ~360 nm, respectively. After the perovskite layer was cooled down to room temperature, ~60 nm PC₆₁BM is spin-coated on the top of perovskite layer at a spin speed of 1800 RPM for 30 s with an acceleration time of 2 s from the PC₆₁BM chlorobenzene solution with the concentration of 20 mg mL⁻¹. Lastly, a 100-nm-thick Al is thermally deposited through a shadow mask in the vacuum with the press of 4×10^{-6} mbar. The active device area is measured to be 0.045 cm².

Characterization of perovskite photodetectors The *J-V* characteristics of perovskite PDs were measured in dark and under monochromatic light of 500 nm with a light intensity of 0.28 mW/cm² from a Xenon lamp on a Keithley model 2400 source measure unit. The EQE is obtained by using the solar cell quantum efficiency measurement system (QEX10) from PV measurements with a 300-W steady-state xenon lamp as the source light. The transient photocurrent measurements were performed by using an optical chopper controlled at $\lambda = 532$ -nm laser pulse at a frequency of 2 kHz.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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