Room-Temperature Solution-Processed NiO\textsubscript{x}:PbI\textsubscript{2} Nanocomposite Structures for Realizing High-Performance Perovskite Photodetectors

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ABSTRACT: While methylammonium lead iodide (MAPbI\textsubscript{3}) with interesting properties, such as a direct band gap, high and well-balanced electron/hole mobilities, as well as long electron/hole diffusion length, is a potential candidate to become the light absorbers in photodetectors, the challenges for realizing efficient perovskite photodetectors are to suppress dark current, to increase linear dynamic range, and to achieve high specific detectivity and fast response speed. Here, we demonstrate NiO\textsubscript{x}:PbI\textsubscript{2} nanocomposite structures, which can offer dual roles of functioning as an efficient hole extraction layer and favoring the formation of high-quality MAPbI\textsubscript{3} to address these challenges. We introduce a room-temperature solution process to form the NiO\textsubscript{x}:PbI\textsubscript{2} nanocomposite structures. The nanocomposite structures facilitate the growth of the compact and ordered MAPbI\textsubscript{3} crystalline films, which is essential for efficient photodetectors. Furthermore, the nanocomposite structures work as an effective hole extraction layer, which provides a large electron injection barrier and favorable hole extraction as well as passivates the surface of the perovskite, leading to suppressed dark current and enhanced photocurrent. By optimizing the NiO\textsubscript{x}:PbI\textsubscript{2} nanocomposite structures, a low dark current density of 2 × 10\textsuperscript{−10} A/cm\textsuperscript{2} at −200 mV and a large linear dynamic range of 112 dB are achieved. Meanwhile, a high responsivity in the visible spectral range of 450−750 nm, a large measured specific detectivity approaching 10\textsuperscript{13} Jones, and a fast fall time of 168 ns are demonstrated. The high-performance perovskite photodetectors demonstrated here offer a promising candidate for low-cost and high-performance near-ultraviolet−visible photodetection.

KEYWORDS: perovskites, photovoltaic photodetectors, nanocomposite, low dark current, high detectivity

Photodetectors (PDs) that convert incident light into electrical signals have been broadly applied in scientific measurements, industrial manufacturing, optical communications, medical/chemical sensing, and light imaging. In order to achieve high-performance PDs for practical applications (with low dark current density, large responsivity, high specific detectivity, and fast response time), their light-absorbing materials should meet several requirements: (i) high absorption coefficient; (ii) large carrier mobility; (iii) favorable energy-level alignment with the electrode materials. Much effort has been paid to fabricate high-performance inorganic semiconductor-based PDs, such as silicon, gallium phosphide, and gallium arsenide phosphide, which typically require ultrahigh vacuum, high temperature, expensive substrates, and rigorous fabrication processing. In order to simplify the fabrication and reduce the cost of PDs, low-temperature solution-processed strategies that mainly exploit organic materials and inorganic semiconductor nanomaterials, such as nanowires and quantum dots, have been explored. Even though some competitive figures of merit of the PDs employing these materials have been demonstrated, the majority of these PDs still have issues of large dark current, small responsivity, low specific detectivity, and slow response time.

Received: April 10, 2016
Accepted: June 24, 2016
Published: June 24, 2016
Recently, hybrid organic—inorganic perovskites such as methylammonium lead iodide (MAPbI3) became a focus of research due to the outstanding optical and electrical properties including a high absorption coefficient (10^5 cm⁻¹ covering 300–750 nm),¹⁴,¹⁵ large carrier mobility,¹⁶ small excitonic binding energy at room temperature (meaning easy generation of free carriers following photoabsorption),¹⁷ and long electron/hole diffusion length.¹⁸,¹⁹ These excellent optoelectronic properties together with low-temperature and solution-processed formation of the polycrystalline films fueled attention to MAPbI3 perovskite-based PDs.²⁰⁻²⁴ Further improvements in the performance of perovskite PDs is expected from (i) designing suitable hole extraction layers (HELs) with favorable energy-level alignment with MAPbI3 for simultaneously enhancing photocurrent extraction and suppressing dark current and (ii) optimizing HELs and selecting suitable electron extraction layers (EELs) to achieve large responsivity, high specifc detectivity, and fast response time of perovskite PDs.

In this work, we introduce room-temperature solution-processed NiOₓ:PbI₂ nanocomposite structures to achieve highly efficient MAPbI₃ perovskite PDs. The nanocomposite structures, on one hand, facilitate the growth of high-quality MAPbI₃ crystalline films. On the other hand, the NiOₓ:PbI₂ nanocomposite structures function as the effective HEL, which suppresses the dark current through the large electron injection barrier at the interface of ITO/HEL, achieves efficient hole extraction due to the suitable energy-level alignment with the perovskite, and passivates the surface of the perovskite. Through optimizing the thicknesses of NiOₓ and PbI₂ layers, the carrier recombination lifetime of MAPbI₃ is increased and the capacitance of perovskite PDs is reduced. Consequently, the perovskite PDs achieve a low dark current density of 2 × 10⁻¹⁰ A/cm² at −200 mV and a large linear dynamic range of 112 dB. A measured specifc detectivity approaching 10¹³ Jones covering the whole visible region and a fast fall time of 168 ns are achieved. With these excellent performances, the perovskite PDs offer a promising candidate for low-cost and high-performance near-ultraviolet—visible photodetection.

RESULTS AND DISCUSSION

Optimization of Device Structure and Dark Current. The device structure of our perovskite PDs is indium tin oxide (ITO)/NiOₓ:PbI₂/MAPbI₃/C₆₀/bathocuproine (BCP)/Ag. Different from the typical perovskite PDs using poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) as the HEL in the literature,²⁰⁻²² NiOₓ:PbI₂ nanocomposite structures can not only function as effective HEL but also favor the formation of high-quality perovskite films (shown in Figure 1). The nanocomposite structures are formed by a simple room-temperature solution process. We first form NiOₓ nanostructures by spin-coating the NiOₓ...
nanoparticles dispersed in deionized water at various speeds, followed by the vacuum processing at $10^{-2}$ Torr for 30 min to remove the residual deionized water. The NiO$_2$-PbI$_2$ nanocomposite structures are then formed by the PbI$_2$ nanostructure deposited from PbI$_2$ dissolved in dimethylsulfoxide (DMSO) and dimethylformamide (DMF) blend solution as described in the Methods.

A sub-micrometer-sized MAPbI$_3$ polycrystalline thin film is formed on the NiO$_2$-PbI$_2$ HEL via a sequential deposition as detailed in the Methods. It shows a wide absorption ranging from near-ultraviolet to all over the visible spectral range, with a photoluminescence (PL) peak at 780 nm, as shown in Figure S1. Figure 1b provides the comparison between cross-sectional images of MAPbI$_3$ films grown on the surface of NiO$_2$-PbI$_2$ and PEDOT:PSS. The perovskite layer formed on the NiO$_2$-PbI$_2$ nanocomposite structures exhibits more compact and ordered morphological features than the one grown on PEDOT:PSS, which is beneficial for enhancing device performances.

Suppressing the dark current density and enhancing photocurrent of PDs is of importance for achieving the large linear dynamic range (LDR) and high specific detectivity. This enables PDs to measure the low-level intensity of incident light in such applications as low-level laser therapy and fluorescence imaging. As shown schematically in Figure 1d, owing to the conduction band position of NiO$_2$ being at 1.84 eV, the electron injection barrier between ITO and MAPbI$_3$ increases significantly from 0.8 to 2.86 eV using NiO$_2$-PbI$_2$ nanocomposite structures as the HEL. This considerably suppresses the electron injection in the dark from the external circuit and thus reduces the dark current. On the cathode side of perovskite PDs, BCP is introduced between C$_{60}$ and the cathode to create a large hole injection barrier of 2.5 eV at the Ag/BCP interface (see Figure 1d) and thus contributes to suppression of the dark current, as well. As shown in Figure 1c, the dark current density of NiO$_2$-PbI$_2$-based perovskite PDs is about 2 orders of magnitude lower than that of PEDOT:PSS-based devices.

Another important aspect for suppressing the dark current is the controllable formation of PbI$_2$ nanocrystals in the NiO$_2$-PbI$_2$ nanocomposite HEL. We first verify the formation of PbI$_2$ nanocrystals, and then its contribution on suppressing dark current will be demonstrated. As shown by X-ray diffraction (XRD) patterns in Figure 2a, by adjusting the methylammonium iodide (MAI) concentration during the sequential deposition, the PbI$_2$ content in the MAPbI$_3$ layer experiences an increase upon decreasing MAI concentration. From time-resolved PL spectra (Figure 2b) measured on the structure of NiO$_2$-PbI$_2$/MAPbI$_3$ from the front and back (close to the NiO$_2$-PbI$_2$) sides using a 405 nm pulse laser as an excitation source, a longer PL decay lifetime was observed at the NiO$_2$-PbI$_2$ side, suggesting a different carrier recombination dynamics at these two interfaces of MAPbI$_3$. The longer recombination lifetime measured at the back side corresponds to the reduced carrier recombination at the interface of NiO$_2$-PbI$_2$/MAPbI$_3$, indicating that the NiO$_2$-PbI$_2$ nanocomposite HEL effectively passivates the surface of the perovskite. We further investigate the dark current density of perovskite PDs as a function of different concentrations of MAI (i.e., different PbI$_2$ thicknesses). As shown in Figure 2c, the increase of MAI concentration from 50 to 66 mg/mL induces the gradual increase of dark current density of perovskite PDs because of the reduction of PbI$_2$ content, as confirmed by XRD data of Figure 2a. Interestingly, the responsivity (the ratio of photocurrent density to the intensity of incident light) of the perovskite PDs improves by introducing the PbI$_2$ in the NiO$_2$-PbI$_2$ nanocomposite HEL as shown in Figure S2, particularly when MAI concentration is reduced from 66 to 60 mg/mL because of reduced nonradiative recombination centers. The high responsivity of 0.36 A/W can be maintained over a range of PbI$_2$ content (from MAI of 50–60 mg/mL). We conclude that the formation of PbI$_2$ can not only efficiently reduce dark current but also offer high responsivity over a wide range contents of PbI$_2$ in the NiO$_2$-PbI$_2$-based perovskite PDs.

We optimized the thickness of NiO$_2$ nanostructures in the nanocomposite HEL by studying performances of the devices with four different thicknesses of 18, 15, 14, and 13 nm (denoted as NiO$_2$-1, NiO$_2$-2, NiO$_2$-3, and NiO$_2$-4, respectively), as shown in Figure 3a. The dark current density at the bias of $-200$ mV was reduced from $1 \times 10^{-9}$ to $2 \times 10^{-10}$ A/cm$^2$ when the NiO$_2$ thickness decreased from 18 to 13 nm. For perovskite PDs operating at $-10$ mV, the measured dark
Transient photocurrent response of perovskite PDs using NiO$_x$-1, NiO$_x$-3, and NiO$_x$-4 HELs are shown in Figure 3c. NiO$_x$-4-based perovskite PDs achieved a dark current comparable to that of NiO$_x$-3-based perovskite PDs, while the fall time was longer for NiO$_x$-4-based perovskite PDs, which can be related to their larger capacitance (16.1 nF for NiO$_x$-3-based PDs vs 20.1 nF for NiO$_x$-4-based PDs). It should be noted that since the NiO$_x$ has a deeper valence band of 5.48 eV compared with the 5.0 eV of PEDOT:PSS, more favorable energy-level alignment of NiO$_x$ with MAPbI$_3$ is realized for enhancing hole extraction and thus photocurrent, as demonstrated in Figure 1d. To demonstrate the photocurrent improvement, we measured the spectral responsivity, as shown in Figure S6. The responsivity of the optimized NiO$_x$:PbI$_2$-based perovskite PDs was better than that of the optimized PEDOT:PSS-based perovskite PDs in the whole wavelength region, except for the small region around 700 nm, probably due to different optical-field distribution in the device. Consequently, by optimizing the thicknesses of NiO$_x$ nanostructures and PbI$_2$ nanocrystals in the NiO$_x$:PbI$_2$ nanocomposite HEL, a low dark current density of $2 \times 10^{-10}$ A/cm$^2$ among the vertical perovskite PDs was achieved.

Spectral Responsivity Characterization. The spectral responsivity of optimized perovskite PDs with the structure of ITO/NiO$_x$ (14 nm):PbI$_2$/MAPbI$_3$ (250 nm)/C$_{60}$ (50 nm)/BCP (10 nm)/Ag (120 nm) is shown in Figure 4a. Remarkably, they exhibit high responsivity in the visible spectral range of 450–750 nm, corresponding to the high EQE between 70 and 90%, which exceeds the value of commercial Si and GaAsP PDs. The high spectral response of perovskite PDs that cover red, green, and blue regions, but does not extend its response to the near-infrared region (thus excess infrared noise will be significantly reduced), can enable their applications in visible light imaging. The comprehensive studies of other characteristics of the optimized perovskite PDs are described below.

LDR Characterization. LDR value, denoting the range where the photocurrent density of perovskite PDs is linearly proportional to the incident light intensity, is an important parameter for characterizing the detectable linear light intensity of perovskite PDs. Theoretically, the lower limit of LDR is determined by the noise current, which originates from the thermal noise (Johnson–Nyquist noise), the shot noise, and 1/ $f$ noise. Typically, shunt resistance largely contributes to the thermal noise (see details in the Supporting Information). NiO$_x$:PbI$_2$-based perovskite PDs achieve a shunt resistance of 14 GΩ, which is about 2 orders of magnitude larger than 0.16 GΩ for PEDOT:PSS-based perovskite PDs, suggesting that shunt resistance can be effectively suppressed by NiO$_x$:PbI$_2$ nanocomposite HEL. Reverse leakage current originating from the reverse-bias-induced charge injection from the external circuit makes a large contribution to the shot noise, which is efficiently suppressed in our perovskite PDs due to the large carrier injection barrier of 2.86 eV at the anode interface and 2.5 eV at the cathode interface, as shown in Figure 1d. Moreover, the introduction of C$_{60}$ EEL was found to efficiently passivate the surface traps of perovskites, which can effectively suppress 1/ $f$ noise that originated from the carrier trapping–detrapping process. In order to obtain the accurate value of the total noise, we directly measured the noise, as shown in Figure S7. The noise current was found to be $\sim20 \text{ fA Hz}^{-1/2}$, corresponding to a noise equivalent power of $\sim56 \text{ fW Hz}^{-1/2}$. 

Figure 3. (a) Dark current density of NiO$_x$:PbI$_2$-based perovskite PDs as a function of four different thicknesses of NiO$_x$ HELs. Thicknesses of NiO$_x$-1, NiO$_x$-2, NiO$_x$-3, and NiO$_x$-4 films are 18, 15, 14, and 13 nm, respectively. (b) Time-resolved PL decay curves of MAPbI$_3$ films formed on NiO$_x$-1, NiO$_x$-3, and NiO$_x$-4 films. The PL decay data were fitted with a multiexponential function. The average carrier recombination lifetimes of MAPbI$_3$ formed on NiO$_x$-1, NiO$_x$-3, and NiO$_x$-4 are 21, 25, and 25.3 ns, respectively. (c) Transient photocurrent response of NiO$_x$:PbI$_2$-3, and NiO$_x$:PbI$_2$-4-based perovskite PDs.
The upper limit of LDR is governed by the saturation current when the incident light intensity reaches a certain level, \( P_{\text{sat}} \), and thus photocurrent cannot be efficiently extracted due to the space charge buildup. \(^{35,36} \) The lowest charge carrier mobility within perovskite PDs determines the saturation current (i.e., the maximum detectable light intensity, \( P_{\text{sat}} \)) and also is a limiting parameter for achieving high-speed response perovskite PDs that will be discussed below. In order to achieve a large \( P_{\text{sat}} \), \( C_{60} \) that has been widely applied in perovskite PDs and solar cells is intentionally selected as the EEL of our perovskite PDs. Using the capacitance of perovskite PDs obtained from RC (resistor-capacitor) time-constant-limited transient response (discussed below) and the \( C_{60} \) electron mobility of \( 1.11 \times 10^{-3} \) cm\(^2\)/Vs (extracted from the space-charge-limited current shown in Figure S8), \(^{37} \) the calculated saturation current is \( 0.36 \) A/cm\(^2\) and the upper limit of LDR reaches \( 1.067 \) W/cm\(^2\).

Consequently, LDR defined as
\[
\text{LDR} = 10 \log \left( \frac{P_{\text{sat}}}{P_{\text{noise}}} \right)
\]
where \( P_{\text{noise}} \) is the lowest detectable light intensity governed by the total noise current. Our perovskite PDs achieved a large LDR of about 120 dB when perovskite PDs were operated.

In order to experimentally characterize LDR of perovskite PDs, we measured the photocurrent density as a function of the wide-range incident light intensities, as shown in Figure 4b. The lowest light intensity of \( 2.5 \times 10^{-12} \) W/cm\(^2\) and the highest light intensity of \( 0.41 \) W/cm\(^2\) were achieved for our perovskite PDs, corresponding to the LDR of 112 dB, which is the highest value among reported perovskite PDs, \(^{20-22,25,42} \) comparable with the value of commercial visible Si PDs (120 dB), \(^{30} \) GaAsP PDs (110 dB), \(^{39} \) and state-of-the-art organic-material-based PDs (110 dB). \(^{25,40,41} \) It should be mentioned that the measured lowest light intensity approaches the value of device noise, as shown in Figure 4c, suggesting efficient trap passivation by \( C_{60} \) EEL as well as high-quality perovskites fabricated on the nanostructured NiO\(_x\):PbI\(_2\) structure. The highest light intensity of 0.41 W/cm\(^2\) achieved in our perovskite PDs approached the theoretical value of 1.067 W/cm\(^2\), which probably resulted from the lower hole mobility of NiO\(_x\) and the lower electron mobility of BCP compared with that of MAPbI\(_3\) perovskite and \( C_{60} \). \(^{25,40,42} \)

**Detectivity Characterization.** Specific detectivity \( D^* \) is one of the most important figures of merit for PDs, defined as
\[
D^* = (A\Delta f)^{1/2}R/i_{\text{measure}}
\]
where \( A \) is the device area, \( \Delta f \) is the bandwidth, \( R \) is the responsivity, and \( i_{\text{measure}} \) is the measured noise (Figure S7). \( D^* \) normalizes variations in device working areas and bandwidth, thus enabling comparison among different PDs. Specific detectivity \( D^* \) of our optimized perovskite PDs as a function of spectral wavelength is presented in Figure 4d. Values of \( D^* \) exceeding \( 1.0 \times 10^{12} \) Jones in the near-ultraviolet region (340–400 nm) and \( 4.0 \times 10^{12} \) Jones covering the whole visible region (450–750 nm) are achieved, which are among the highest reported for perovskite-based PDs \(^{11,22,38,43-46} \) and organic-material-based PDs (~\( 1 \times 10^{12} \) Jones) \(^{40,47,48} \) and comparable to the theoretical value of commercial Si (~\( 4.5 \times 10^{13} \) Jones), GaAsP (3.0 \times 10^{13} Jones), and GaP PDs (2.3 \times 10^{12} Jones). \(^{7,29} \)

**Transient Photocurrent Response Characterization.** The photocurrent response time determines the ability of PDs...
to truthfully record a high-frequency incident light signal. The upper limit of response time $T_r$ of PDs is mainly governed by the carrier transit time.\(^\text{30}\) For our perovskite PDs, $T_r$ is the sum of the carrier transit across the perovskite and the $C_{60}$ layers; it is estimated to be 23 ns (see details in the Supporting Information). It is limited by the electron mobility of $C_{60}$, which is lower than that of MAPbI$_3$ perovskite.\(^\text{16}\) The transient photocurrent response of optimized perovskite PDs (under 532 nm light with the intensity of about 3 $\mu$W/cm$^2$) is shown in Figure S11, with a rise time of 58 ns and a fall time of 168 ns. By reducing the size of perovskite PDs and thus the RC time constant, the photocurrent response is expected to approach the theoretical limit. In order to confirm this assumption, the RC time constant of the perovskite PD system was calculated (see details in Figure S9). Using the integrated Q of 0.9578 nC, the transient voltage $V$ of 0.0595 V, and the load resistor $R$ with 10 $\Omega$, RC time constant ($RC = RV/Q$) was calculated to be 161 ns, which is close to our measured fall time, suggesting that the transient photocurrent response of our perovskite PDs is governed by RC time constant. This is further confirmed in Figure S10, showing that measured fall times of perovskite PDs increase with the raised resistance of load resistors. It should be noted that transient photocurrent response (rise time and fall time) is a function of the intensity of incident light. Based on the results shown in Figure S11, the rise time and fall time of our perovskite PDs obeyed this trend, as they increased slowly with the increased intensity of incident light, suggesting that the capacitance of perovskite PDs increases with the increased incident light power, which is attributed to the enhanced dielectric permittivity of perovskites under illumination.\(^\text{39}\) The photocurrent response as a function of modulation frequency of incident light is shown in Figure Sb. Perovskite PDs with a working area of 6 mm$^2$ achieved a 3 dB cutoff frequency of 2 MHz, which is comparable to the value of 2.9 MHz for the previously reported perovskite PDs with a much smaller working area of 1 mm$^2$.\(^\text{20}\) It should be noted that the transient photocurrent response of our perovskite PDs is limited by the RC time constant. Through reducing the working area of perovskite PDs, our perovskite PDs achieved a 3 dB cutoff frequency of 7 MHz (see Figure S12). Consequently, the 3 dB cutoff frequency of 24 MHz can be expected if carrier transit time determines the transient photocurrent behavior. In addition to the large-bandwidth photocurrent response, our perovskite PDs are repeatable (Figure S13) and stable (Figure S14), which are beneficial for practical applications.

**CONCLUSIONS**

We have introduced a room-temperature solution-processed NiO$_2$:PbI$_2$ nanocomposite HEL into MAPbI$_3$ perovskite-based PDs and demonstrated their excellent performance characteristics. The NiO$_2$:PbI$_2$ layer favors the subsequent formation of high-quality crystalline MAPbI$_3$ films and passivates the surface of the perovskite. Moreover, the NiO$_2$:PbI$_2$ nanocomposite structures offer a large electron injection barrier at the ITO interface for suppressing dark current and the favorable energy-level alignment with perovskite film for enhancing the hole extraction. After the thicknesses of the NiO$_2$:PbI$_2$ nanostructure were optimized, an ultralow dark current density of $2.0 \times 10^{-10}$ A/cm$^2$ at $-200$ mV was achieved. Besides, a high high LDR of measured 112 dB, and a fast fall time of 168 ns limited by RC time constant were demonstrated. These excellent figures of merit of our perovskite PDs make them highly promising for a wide range of applications, such as light imaging, medical/chemical sensing, optical communications, and smoke/fire monitoring.

**METHODS**

**NiO$_2$:PbI$_2$ Nanocomposite Structures.** The NiO$_2$:PbI$_2$ nanocomposite structure was produced by a simple and fast room-temperature and solution-based process on ITO/glass substrates with the square of resistive of $\sim 17 \, \Omega \cdot \square$. First, NiO$_2$ nanoparticles were synthesized by a chemical precipitation method using Ni(NO$_3$)$_2$·6H$_2$O and NaOH. Briefly, Ni(NO$_3$)$_2$·6H$_2$O (0.5 mol) was dispersed in 100 mL of deionized water to obtain a dark green solution. The pH of the solution was adjusted to 10 by adding a NaOH solution (10 mol L$^{-1}$). After being stirred for 5 min, the colloidal precipitation was thoroughly washed with deionized water twice and dried at 80 °C for 6 h. The obtained green powder was then calcined at 270 °C for 2 h to obtain a dark black NiO$_2$ nanoparticle powder. Then, the nanoparticles were dispersed in the deionized water to obtain the desired concentrations. NiO$_2$ nanocostrucures were formed by spin-coating the nanoparticles at various speeds, followed by the vacuum processing at $10^{-2}$ Torr for 30 min to remove the residual deionized water. To form the NiO$_2$:PbI$_2$ nanocomposite structure, a solution of 0.5 g/mL PbI$_2$ (Sigma-Aldrich 99% PbI$_2$) in a DMSO/DMF mixture (4:6 by volume) was spin-coated on the NiO$_2$ nanoparticle structures at 5000 rpm for 50 s.

**Device Fabrication.** The device structure of perovskite PDs is ITO/NiO$_2$:PbI$_2$/MAPbI$_3$/C$_60$/BCP/Ag. The cleaning process of ITO substrates can be found elsewhere.\(^\text{40}\) Starting from the NiO$_2$:PbI$_2$/MAPbI$_3$ nanocomposites, the samples were spin-coated with different concentrations of MAI (50, 55, 60, and 66 mg/mL from isopropyl...
alcohol solvent). The samples were then heated in nitrogen atmosphere on a hot plate at 100 °C for 10 min to obtain the MAPbI3 polycrystalline films. To form the EEL, 0.02 wt % C60 in 1,2-dichlorobenzene was directly spin-coated on the MAPbI3 polycrystalline films. Cross-sectional SEM images of the perovskite films grown on NiO, PEDOT:PSS substrates were measured by a LEO 1530 FEG SEM. The thicknesses of the NiO, nanostructures were determined by a J.A. Woollam Co. ellipsometer. The optical transmittance of MAPbI3 polycrystalline films was measured by the goniometer. The absorbance spectra were determined from the optical transmittance. The PL spectrum of MAPbI3 was measured on a home-built spectrometer using an excitation source of 325 nm laser. Time-resolved PL spectra (TCSPC) of MAPbI3 were measured on an Edinburgh Instruments FLS920 spectrophotometer equipped with a 405 nm, 5 mW picosecond pulsed diode laser (EPL-405S) as an excitation source. PL decay data were fitted using a multiparameter exponential function. Lifetime was calculated by taking the weighted average across the exponential components.

**Film Characterization.** The top morphology images of perovskites grown on NiO/PbI2, nanocomposite structures were characterized by NT-MDT atomic force microscopy. Cross-sectional SEM images of the perovskite films grown on NiO, and PEDOT:PSS substrates were measured by a LEO 1530 FEG SEM. The thicknesses of the NiO, nanostructures were determined by a J.A. Woollam Co. ellipsometer. The optical transmittance of MAPbI3 polycrystalline films was measured by the goniometer. The absorbance spectra were determined from the optical transmittance. The PL spectrum of MAPbI3 was measured on a home-built spectrometer using an excitation source of 325 nm laser. Time-resolved PL spectra (TCSPC) of MAPbI3 were measured on an Edinburgh Instruments FLS920 spectrophotometer equipped with a 405 nm, 5 mW picosecond pulsed diode laser (EPL-405S) as an excitation source. PL decay data were fitted using a multiparameter exponential function. Lifetime was calculated by taking the weighted average across the exponential components.

**Device Characterization.** Current density as a function of working bias and of the different wavelength (responsivity) was measured on a Keithley 2635 source meter with a light source setup of 1000 W xenon arc lamp integrated with an Acton SpectraPro 2150i monochromator. Dynamic linear range measurements were performed under different intensities of monochromatic light using ThorLabs metallic-coated neutral density filters. The intensity of monochromatic light was measured by facility calibrated (Newport) Si photodetectors. The transient photocurrent measurements have been done with a 532 nm 6 ps pulse width laser (130 μJ) taking the weighted average across the exponential components.

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