Enhanced X-ray Sensitivity of MAPbBr₃ Detector by Tailoring the Interface-States Density

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Supporting Information

ABSTRACT: An important factor for the high performance of light-harvesting devices is the presence of surface trappings. Therefore, understanding and controlling the carrier recombination of the organic–inorganic hybrid perovskite surface is critical for the device design and optimization. Here, we report the use of aluminum zinc oxide (AZO) as the anode to construct a p–n junction structure MAPbBr₃ nuclear radiation detector. The AZO/MAPbBr₃/Au detector can tolerate an electrical field of 500 V·cm^{−1} and exhibit a very low leakage current of ~9 nA, which is 1 order of magnitude lower than that of the standard ohmic contact device. The interface state density of AZO/MAPbBr₃ contact was reduced from 2.17 × 10¹⁰ to 8.7 × 10⁸ cm^{−2} by annealing at 100 °C under an Ar atmosphere. Consequently, a photocurrent to dark current ratio of 190 was realized when exposed to a green light-emitting diode with a wavelength of 520 nm (~200 mW·cm^{−2}). Simultaneously, a high X-ray sensitivity of ~529 μC·Gy_{air}^{−1} cm^{−2} was achieved under 80 kVp X-ray at an electric field of 50 V·cm^{−1}. These results demonstrate the use of surface engineering to further optimize the performance of MAPbBr₃ detectors, which have many potential applications in medical and security detection with low radiation dose brought to the human body.

KEYWORDS: p–n junction, X-ray detector, interface state density, MAPbBr₃ PSC, annealing

INTRODUCTION

X/γ-ray detection is of great interest for a range of applications, such as medical imaging, industrial inspection, astronomical telescopes, and scientific research.^{1–3} Among the semiconductor materials explored for direct X/γ-ray detection, the hybrid lead halide perovskite materials are particularly attractive as they combine low-cost fabrication processes and remarkable electro-optical properties.^{4,5} MAPbX₃ (X = Br, I) (MA = CH₃NH₃⁺) perovskite single crystals (PSC) grown by the solution method have a large charge drift length of 175 μm⁶ and a low trap density of 10⁹ cm^{−3},⁷ which shows great potential for applications in solar cells, light-emitting diodes (LEDs), lasers, scintillators, and radiation detection.^{8–17}

Recently, many achievements in MAPbBr₃ single crystal growth have been developed by tailoring the solvent types, precursor concentration, growth temperature, and so forth.^{18–20} The photoresponse and charge collection efficiency (CCE) of MAPbBr₃ crystal-based devices have been improved by defect engineering.^{21,22} However, to approach higher sensitivity and efficiency for soft and hard X-ray detection, which will require lower radiation doses in many medical and security check applications, the effects of the contact properties

between the electrode and MAPbBr₃ crystal should be also seriously considered.

Several ways to optimize the device interface have been reported. Murali et al.²³ revealed that the surface reorganization of the aged surface can improve the photoelectric performance of the device. Sun et al.²⁴ used the plasmonic effect of gold nanoparticles which can effectively enhance the near-field performance of perovskites and thus facilitate the light-harvesting. Seo et al.²⁵ employed LiF to adjust the dipole moment to reduce the metal work function, thereby increasing the contact barrier. In addition, electron transport layers, hole transport layers, and passivation layers are also adopted to regulate carriers transport at the interface. Therefore, the device stability and the CCE have been greatly improved. However, these methods usually demanded complex processes and potentially high costs. In this work, we report the development of cost-effective surface processing and a simple

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electrode structure for MAPbBr₃ perovskite nuclear radiation detectors.

Generally, C₆₀, BCP, and PCBM are used to fabricate MAPbBr₃ PSC nuclear radiation detectors because the surface traps can be passivated by C₆₀.^{26,27} For MAPbBr₃ PSC nuclear radiation detectors, Wei et al.¹¹ reported an X-ray sensitivity of 80 $\mu\text{C Gy}_{\text{air}}^{-1} \text{cm}^{-2}$ based on the Au/BCP/C₆₀/MAPbBr₃/Au structure X-ray detector at -0.1 V bias. Xu et al.²⁷ fabricated a Cr/MAPbBr₃/C₆₀/BCP/Cr detector and obtained ²⁴¹Am alpha particle response with an applied bias voltage of 180–550 V. Wei et al.⁴ prepared a detector of Cr/MAPbBr_{2.94}Cl_{0.06}/C₆₀/BCP/Cr structure, which can get a distinguishable ¹³⁷Cs energy peak. C₆₀ and BCP in the device promote the charge transfer and improve charge collection.²⁸ However, there are still some shortcomings that need to be issued before their commercialization. First, these raw materials are difficult to purify, and the resulting synthesis methods are complicated and costly. Second, the performance of the device deteriorates easily due to water denaturation of the BCP and C₆₀ layers.^{29,30}

In addition, the indium-free, cost-effective aluminum zinc oxide (AZO) anode has been proved to be an alternative electrode in MAPbBr₃ devices. AZO is an n-type semiconductor³¹ with high electrical conductivity.^{32,33} The adhesion between a magnetron-sputtered AZO film and the MAPbBr₃ PSC is relatively high, which is easy to standardize and scale up in comparison to C₆₀, or BCP.^{11,27,34} A device with AZO has a simple structure, and there are no issues related to multiple interface matching. Moreover, the charge recombination at the perovskite interface can be suppressed and the CCE can be improved by AZO films.³⁵ Therefore, AZO electrodes have a potential to replace the conventional metal electrodes in MAPbBr₃ nuclear radiation detector fabrication.

However, in order to reduce the carrier recombination further and improve the photoresponse characteristics, modification of the interface state density is still needed. Simpson et al.³⁶ reported that the electron trap density of perovskite thin films can be reduced by thermal annealing for 2 h. Huang et al.³⁷ used multistep annealing to improve the stability of a CH₃NH₃PbI₃ device and achieved a power conversion efficiency of 13.58%, which was attributed to the reduction of physical defects or bulk traps. Pan et al.³⁸ showed that Ag⁺/Bi³⁺ disordering can be eliminated by thermal annealing and surface treatment, which resulted in an improvement of the resistivity and X-ray detection limit of Cs₂AgBiBr₆ single crystal.

In this work, we report the fabrication and characterization of MAPbBr₃ PSC X-ray detectors based on the bulk crystals grown by the solution method. The p–n junction structure MAPbBr₃ (p-type) detectors using AZO (n-type) and Au as the anode and cathode, respectively, have been developed. Moreover, the interface state density of MAPbBr₃ and AZO is identified by both capacitance–voltage and conductance–voltage measurements and optimized by wafer annealing under an Ar atmosphere, which is further certified by the enhanced optical response. Simultaneously, an X-ray sensitivity evaluation is systematically conducted to investigate the effect of the interface state density.

EXPERIMENTAL SECTION

Crystal Growth and Wafer Processing of MAPbBr₃ Crystals.

A modified antisolvent vapor-assisted crystallization method was employed to grow MAPbBr₃ single crystals.³⁹ An equimolar mixture

of the MABr (99.9%) and PbBr₂ (99.9%) was dissolved in dimethylformamide (DMF) for 30 min using magnetic stirring. The solution concentration was controlled in the range of 0.7–1.4 M. To control the MAPbBr₃ growth rate, the antisolvent mixture of dichloromethane and DMF was volatilized into the precursor solution to reduce the solubility of MAPbBr₃ in the solution. Finally, centimeter-sized well-defined cubic MAPbBr₃ single crystals with six (100) facets, as in Figure S1, were obtained at room temperature. Then, the MAPbBr₃ wafers with the dimension of $4 \times 4 \times 2 \text{ mm}^3$ were cut from the as-grown MAPbBr₃ crystals and mechanically polished using fine diamond paste. Finally, the wafer was chemically etched with DMF for 20 s prior to deposition of electrodes.

Electrodes Preparation and Annealing. The Au electrode with a thickness of 80 nm was deposited onto one face of the MAPbBr₃ wafer by thermal evaporation. The AZO film $\sim 300 \text{ nm}$ was prepared on the opposite face by MSP-400 RF magnetron-sputtering method using ZnO–Al₂O₃ (98:2 wt %) ceramic as the sputtering target. The diameters of both Au and AZO electrodes are 2 mm, as shown in Figure 1b,c. High purity Ar (5 N) was adopted as the atmosphere,

Figure 1. (a) Structure of the MAPbBr₃ device with Au and AZO electrodes, (b) MAPbBr₃ wafer coated with Au electrode. (c) MAPbBr₃ wafer sputtered with AZO electrode. (d) AFM image of AZO film on Si wafer. (e) Cross-sectional SEM image of AZO film on MAPbBr₃ wafer.

with a sputtering pressure of 0.4 Pa. The sputtering power was set in the range of 50–100 W, which was finally optimized at 60 W to minimize the leakage current at higher bias (Figure S3). Immediately after deposition, the PSC/AZO devices were further modified by annealing at a temperature of 100 °C. The devices were annealed in an Ar atmosphere over a time range of 30–120 min.

Electrical and Photo-Electric Response Measurement. The current–voltage (I – V) and current–time (I – t) curves were measured at room temperature using a Keithley 6517B picoammeter/voltage supply under both low and high bias. Photoconductivity was measured using a custom-made setup. The MAPbBr₃ device was placed into a dark box and exposed to a 520 nm LED ($\sim 200 \text{ mW}\cdot\text{cm}^{-2}$) with an ON–OFF state. The capacitance–voltage (C – V) and conductance–voltage (G – V) curves were measured at room temperature using an Agilent B1500A with the frequency of 1 kHz, from -15 to 15 V .

Characterization of X-ray Detection Performance. For X-ray detection performance, a tungsten target X-ray tube (SPELLMAN XRB011) was used as the source. The X-ray source was operated with a tube voltage of 80 kVp, and the tube current varied from 1 to 100 μA . The X-ray dose rate was in direct proportion to the tube current and was carefully calibrated by using both the Geiger counter and the thermoluminescence dosimetry. The MAPbBr₃ device was placed in a dark shielded box to prevent disturbance from ambient light. The bias voltage was applied on the device and the induced photocurrent was recorded by a Keithley 6517B.

RESULTS AND DISCUSSION

Design of Asymmetric Electrodes. As-grown MAPbBr₃ PSC shows p-type conductivity according to the Hall measurement at room temperature, seen in Table S1. Therefore, to fabricate MAPbBr₃ nuclear radiation detectors operated at high bias, an asymmetric electrode configuration was proposed, where one electrode is n-type semiconductor, such as AZO, ITO, or electronic transport layer such as BCP, C₆₀, and the opposite electrode is metal with high work function, such as Au and Pt.⁴⁰ Figure 1a shows the p–n junction MAPbBr₃ device with AZO and Au electrodes, AZO and Au contact to MAPbBr₃ to form a p–n junction and an ohmic contact, respectively. To reveal the morphology and optimize the film, first, the amorphous AZO was deposited on the polished (100) Si wafer to avoid the influence of roughness. A typical AFM image of AZO films is shown in Figure 1d, with an area of 300 × 300 nm² and the surface roughness of ~1.6 nm, which indicates the smooth amorphous AZO film. Figure 1e shows a typical cross-sectional scanning electron microscopy (SEM) image of the AZO/MAPbBr₃ PSC interface, revealing that the AZO film has a thickness of ~300 nm and is densely packed, free from pin-holes and cracks.

To elucidate the effect of the interface and band structure on the charge transport behavior, a band diagram of AZO/MAPbBr₃/Au is illustrated in Figure 2a. The minimum value of

Figure 2. (a) Band diagram of AZO/MAPbBr₃/Au; (b) schematic diagram of the chemical bonding between AZO and MAPbBr₃. (c) Room-temperature *I*–*V* curves for MAPbBr₃ devices with different annealing times. (d) ON–OFF responses to 520 nm light (~200 mW·cm^{−2}) at a bias of 100 V at room temperature.

the conduction band and the maximum value of the valence band in MAPbBr₃ are −3.3 and −5.6 eV, respectively. When AZO is prepared as the anode, the barrier height of holes at the AZO/MAPbBr₃ interface is increased in a reverse bias state; thus, the reverse dark current is reduced. When carriers transport through the electrode, they are easily captured and recombined at the interface. Subsequently, the electron–hole recombination will reduce the CCE of the device, which deteriorates X/γ-ray spectroscopy response. Therefore, in addition to improving the performance of crystals, it is also important to modify the device interface states. After polishing and etching, there is a high concentration of MA⁺ and Pb²⁺ dangling bonds on the (100) MAPbBr₃ PSC. The deposition AZO on MAPbBr₃ tends to neutralize and fill the dangling bonds because O^{2−} ions in AZO (ZnO or Al₂O₃) would react with MA⁺ and Pb²⁺, as shown in Figure 2b. Thus, AZO/

MAPbBr₃ interface state density can be reduced. However, the reaction needs to be promoted by thermal annealing.

The *I*–*V* curves of the AZO/MAPbBr₃/Au detector were measured at room temperature before and after annealing at 100 °C in Ar, for different times. AZO is grounded and a voltage is applied to the gold electrode, which is facing up. The results are shown in Figure 2c, which exhibit typical diode-like shapes owing to the excessive injection under the forward bias.⁴¹ The leakage current under reverse bias decreases gradually as the annealing time increases. The lowest leakage current was achieved as ~9 nA under an electric field of 500 V·cm^{−1} for the sample with annealing time of 90 min. The α particle-induced pulse height spectra was obtained at a reverse bias from 100 to 500 V using a 0.8 μ Ci ²⁴¹Am @ 5.49 MeV source, as seen in Figure S4. An electron mobility lifetime ($\mu\tau$)_e product of 2.6 × 10^{−4} cm²·V^{−1} was achieved by fitting the pulse height spectra versus the electrical field, using the single charge carrier Hecht equation.^{42,43} Comparing to the reported $\mu\tau$ values using α pulse height spectra fitting by the Hecht equation, Liu et al.³⁹ revealed that the ($\mu\tau$)_e in MAPbBr₃ crystal is 2.2 × 10^{−4} cm²/V by using Au/MAPbBr₃/Au devices. Xu²⁷ obtained a ($\mu\tau$)_h of 4 × 10^{−4} cm²/V.

The ON–OFF state photocurrent response under 500 V·cm^{−1} electric field was recorded as a function of time, when illuminated by an LED light with a wavelength of 520 nm (~200 mW·cm^{−2}), as shown in Figure 2d. In order to certify the reduction of the AZO/MAPbBr₃ interface state density by annealing, the LED light should be incident from the AZO side. Thus, a positive voltage was applied to the AZO electrode to obtain the rectification. The ON–OFF current response is instantaneous without significant exponential decay. The leakage current was reduced after annealing. The ratios of photocurrent to dark current was 190 after 90 min annealing, which is 7 times higher than that before annealing. Because of the short penetration depth of 520 nm light, most of the photogenerated carriers were located near the surface rather than in the bulk. Therefore, these results suggested that the surface recombination was reduced by the annealing process. Further X-ray photoelectron spectroscopy (XPS) measurements on the AZO/MAPbBr₃ interface before and after annealing also confirmed the chemical reaction between O^{2−} and Pb²⁺ and MA⁺ on the (100) plane, as shown in Figure S5 and Table S2. In addition, the PL spectra of MAPbBr₃ PSC were compared before and after annealing, as seen in Figure S6. No significant differences were observed, which reflected that the annealing did not deteriorate the properties of the bulk crystal and introduce more defects.

Interface State Density of AZO/MAPbBr₃. Capacitance–voltage (*C*–*V*) and conductance–voltage (*G*–*V*) measurements on AZO/MAPbBr₃/Au devices were conducted at room temperature. The voltage is applied to the Au electrode, and the AZO electrode is grounded. Figure 3a shows the *C*–*V* curves of AZO/MAPbBr₃/Au devices after annealing. The barrier height of AZO/MAPbBr₃ contact can be calculated using the formula⁴⁴

$$C(V) = A \sqrt{\frac{\epsilon \epsilon_0 q^2 N_d}{2(\phi_0 - qV)}} \quad (1)$$

where *A* is the area of the diode, ϵ is the relative permittivity of the semiconductor, ϵ_0 is the permittivity in vacuum, *q* is the electron charge, *N_d* is the concentration of uncompensated acceptors, and ϕ_0 is the barrier height from the semiconductor

Figure 3. (a) C – V , (b) $1/C^2$ – V , and (c) G – V curves of AZO/MAPbBr₃/Au devices with annealing time in the time range of 30–120 min. (d) AZO/MAPbBr₃ interfaces state densities as a function of the annealing time.

side. Assuming the whole space charge region was homogeneous, linear $1/C^2$ – V plots are obtained under the reverse bias, as shown in Figure 3b, which is in agreement with published data.⁴⁵ The resulting barrier heights (ϕ_0) are listed in Table 1.

Table 1. Contact Barriers of AZO Electrodes Deposited on MAPbBr₃ Crystals

annealing time (min)	barrier height (eV)
30	0.76
60	0.77
90	0.85
120	0.79

The barrier heights are gradually increased from 0.76 to 0.85 eV with the annealing time from 30 to 90 min, but decreased to 0.79 eV under the annealing time of 120 min. The increased barrier height reduces the possibility for electrons to cross the interface layer, which is consistent with the observed I – V characteristics in Figure 2c.

Figure 3c shows the G – V curves of AZO/MAPbBr₃/Au devices after annealing. The peak shapes become sharper as the annealing time increasing, which reflects the decrease of the interface state density. The interface state densities (N_{ss}) can be derived from the Hill–Coleman method⁴⁶ based on the high-frequency C – V and G – V results, whose formula is as follows

$$N_{ss} = \frac{2}{qA} \frac{(G_m/\omega)_{\max}}{[(G_m/\omega)_{\max} C_{ox}]^2 + (C_m/C_{ox})^2} \quad (2)$$

where $(G/\omega)_{\max}$ is the maximum measured conductance, C_{ox} is the capacitance of insulator layer in strong accumulation region, and C_m is the capacitance that corresponds to $(G/\omega)_{\max}$.

According to G – V curves, the interface state density of AZO/MAPbBr₃ was obtained, which decreases with the annealing time up to 90 min, as seen in Figure 3d. A minimum value of $8.7 \times 10^8 \text{ cm}^{-2}$ was approached, which is

4% of AZO/MAPbBr₃ contact ($2.17 \times 10^{10} \text{ cm}^{-2}$, seen in Supporting Information Figure S7) without annealing. Generally, Br vacancies tend to be generated in the MAPbBr₃ perovskite surface due to the loss of bromide, which results in a net positive charge residing on Pb and MA atoms at the crystal surface (such as Pb^{2+} and MA^+). Such defects have been observed in low-temperature scanning tunneling microscopy studies of MAPbBr₃ crystals.^{47,48} Therefore, the carriers are easily trapped by Pb^{2+} and MA^+ cations when transporting through the interface.⁴⁹ Fang et al. reported that O_2 and H_2O adsorbed physically on the MAPbBr₃ surface provided electrons to Pb^{2+} cations, effectively neutralizing the excess positive charge, thus greatly regulating the surface recombination rate in MAPbBr₃ PSC.²¹ In this work, Ar atmosphere annealing promotes the interdiffusion between AZO and MAPbBr₃, which leads to O^{2-} in AZO combining with the dangling Pb^{2+} or MA^+ in MAPbBr₃ and decreasing the interface state density.

Device Performance of MAPbBr₃ X-ray Detectors. X-ray response and sensitivity of Au/MAPbBr₃/AZO detectors were investigated before and after annealing. A negative voltage is applied to the Au electrode, and X-rays are incident on the AZO surface. X-ray irradiation was performed with a tube voltage of 80 kV and a tube current from 1 to 100 μA . The time-dependent photocurrent of the detector was recorded by modulating the dose rate from 1 to 72 μGys^{-1} before and after annealing, as shown in Figure 4a,c. The average dark current density at -5 V bias is -7.6 and -5.7 nA cm^{-2} , respectively, before and after 100 $^\circ\text{C}$ annealing for 90 min. It is suggested

Figure 4. X-ray response and sensitivity, (a) time response of MAPbBr₃ sample under 80 kVp X-ray beam before annealing in the range of 0–100 μA at 25 V cm^{-1} , (b) X-ray response of MAPbBr₃ device before and after annealing, (c) time response of annealed sample under 25 V cm^{-1} , (d) X-ray photocurrents of annealed and unannealed sample as a function of dose rate under 25 V cm^{-1} , (e) time response of annealed sample under 50 V cm^{-1} , (f) X-ray photocurrent and CCE of annealed sample as a function of exposure dose rate under 50 V cm^{-1} .

that the annealed sample can detect a lower dose rate of X-ray with tube current threshold of 2 μA . However, the unannealed sample can only respond with a minimum tube current of 10 μA . This is determined by the signal-to-noise ratio of the device, which is driven by the dark current. The detection limit of the AZO/MAPbBr₃/Au detector was evaluated as 1.21 μGys^{-1} , seen in Figure S8.

Figure 4b demonstrates the photoresponse of the device by turning the X-ray source on and off with a dose rate of 380 μGys^{-1} . The ON–OFF ratio is 5.2 and 3.5, respectively, for the annealed and as-deposited Au/MAPbBr₃/AZO detectors at 25 $\text{V}\cdot\text{cm}^{-1}$. The enhancement of photocurrent response is attributed to the decrease of the interface state density after annealing. The carrier recombination in the interface is reduced. Therefore, more photogenerated carriers are collected in the annealed device. The photocurrent density of the annealed device is 2.7 nA/cm^2 , which is much higher than that of before annealing ($\sim 0.6 \text{ nA}/\text{cm}^2$), under the dose rate of 7 μGys^{-1} , as seen in Figure 4d.

Besides, the X-ray sensitivity was examined by fitting the photocurrent density as a function of dose rate, as shown in Figure 4d. An X-ray sensitivity of 467 $\mu\text{C Gy}^{-1} \text{ cm}^{-2}$ is realized for the annealed detector at an electrical field of 25 $\text{V}\cdot\text{cm}^{-1}$, which is 3 times higher than that of the as-deposited device (155 $\mu\text{C Gy}^{-1} \text{ cm}^{-2}$). Comparing to the published results, this value is 23 times higher than the state-of-the-art α -Se detectors (20 $\mu\text{C Gy}^{-1} \text{ cm}^{-2}$ at 10 $\text{V}\cdot\mu\text{m}^{-1}$)⁵⁰ and more than 5 times higher than the Au/MAPbBr₃/C₆₀/BCP/Au detector under 50 kVp X-ray (80 $\mu\text{C Gy}^{-1} \text{ cm}^{-2}$),¹¹ although at relatively low electrical field.

To improve the X-ray sensitivity, a higher electrical field (50 $\text{V}\cdot\text{cm}^{-1}$) was performed on the annealed device, as shown in Figure 4e. The photogenerated electron–hole pairs are tended to be separated under high bias, which reduces the carrier recombination and results in higher CCE. The sensitivity was improved from 467 to 529 $\mu\text{C Gy}^{-1} \text{ cm}^{-2}$. The ON–OFF current response is instantaneous even when exposed to a higher X-ray dose rate of 72 μGys^{-1} . Figure 4f shows the photocurrent density and CCE under 50 $\text{V}\cdot\text{cm}^{-1}$. The CCE was calculated as $\text{CCE} = I_{\text{R}}/I_{\text{p}}$, where I_{R} is induced photocurrent and I_{p} is theoretical photocurrent induced by X-ray ionization, which is defined as⁵¹ $I_{\text{p}} = \varphi\beta e$, where e is the quantity of electric charge. The photon absorption rate φ can be defined as $\varphi = \varepsilon D m_{\text{s}}/E_{\text{ph}}$, where ε is the fraction of photons in the sample ($\sim 100\%$ for the 2 mm sample in this study, the attenuation efficiency to 50 keV X-ray for MAPbBr₃ SCS seen in Figure S2), D is the dose rate, m_{s} is the mass of the sample, and E_{ph} is the photon energy. β is the number of carriers generated by per X-ray photon, given by $\beta = E_{\text{ph}}/\Delta$, where Δ is calculated by the empirical model¹¹ of $\Delta = 1.43 + 2E_{\text{g}}$. The CCE of the detector declined from 66 to 52% in the dose rate range of 7–36 μGys^{-1} , as shown in Figure 4f. The reduction tendency is similar to the phenomenon in Cs₂AgBiBr₆ PSC detector.³⁸ This phenomenon is generally observed in photoconductive photodetectors. When the irradiation intensity increases, carriers tend to fill into shallower traps with smaller gain, which is known as the dynamic-range enhancing gain compression.^{52,53}

CONCLUSIONS

The p–n junction structure AZO/MAPbBr₃/Au detector was prepared by sputtering to suppress the leakage current. A low leakage current of $\sim 9 \text{ nA}$ was achieved under an electric field

of 500 V/cm . The interface state density of AZO/MAPbBr₃ was tailored by annealing under an Ar atmosphere at 100 $^{\circ}\text{C}$ in the time range of 30–120 min, which was reduced from 2.17×10^{10} to $8.7 \times 10^8 \text{ cm}^{-2}$ after 90 min annealing. Simultaneously, the charge transport behavior was modified, which resulting in an improvement by 3 times of the X-ray sensitivity. Finally, a high sensitivity of 529 $\mu\text{C Gy}^{-1} \text{ cm}^{-2}$ was realized under 80 kVp continual X-rays at an electric field of 50 $\text{V}\cdot\text{cm}^{-1}$. This high X-ray sensitivity suggests that MAPbBr₃ PSCs could be used as active components in X-ray detection applications with the device performance improved by interface state optimization.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.8b18598.

MAPbBr₃ crystal image; X-ray diffraction patterns of MAPbBr₃; attenuation efficiency to 50 keV X-ray for MAPbBr₃ PSC; Hall measurement results; I – V curves; α particles response of the MAPbBr₃ PSC device; XPS analysis; photoluminescence spectra; C – V and G – V results; and calculation of signal-to-noise ratio (PDF)

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Notes

The authors declare no competing financial interest.

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