

## RESEARCH ARTICLE

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# An eco-friendly self-powered X-ray detector based on a multi-layered double perovskite ferroelectric

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Self-powered X-ray detectors based on lead halide perovskites (LHPs) have recently gained increased traction due to their low energy consumption and easy fabrication. However, the toxicity of lead in LHPs threatens human and environmental safety, which hinders their commercialization. Herein, an eco-friendly self-powered X-ray detector has been successfully fabricated using a multilayered double perovskite ferroelectric (C<sub>6</sub>H<sub>5</sub>CH<sub>2</sub>NH<sub>3</sub>)<sub>2</sub>CsAgBiBr<sub>7</sub> (**1**). The intrinsic ferroelectric spontaneous polarization property ( $P_s = 3.19 \mu\text{C cm}^{-2}$ ) in **1** generates an impressive polar photovoltage of 0.5 V, which provides an ideal driving force for exciton dissociation within the material, enabling X-ray detection without an external power source. The X-ray detector based on **1** shows a high sensitivity of  $47.2 \mu\text{C Gy}^{-1} \text{cm}^{-2}$  and an extremely low X-ray detection limit of  $716 \text{ nGy s}^{-1}$  at 0 V bias. Additionally, under an applied bias of 50 V, the sensitivity increases to  $1154.8 \mu\text{C Gy}^{-1} \text{cm}^{-2}$ , surpassing that of most halide double perovskite detectors. This work sheds light on the great potential of multilayered double perovskite ferroelectrics for cost-effective "green" self-powered X-ray detectors.

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## 1. Introduction

X-ray detectors which can convert X-rays into electrical signals play an indispensable role in many important fields, such as medical diagnosis, industrial production, and aerospace research.<sup>1–3</sup> Currently, commercial X-ray detectors mainly rely on pure inorganic semiconductors, such as Si,  $\alpha$ -Se, CdTe, *etc.*<sup>4–6</sup> These materials still face some problems such as low X-ray absorption coefficients and huge energy costs for high-temperature fabrication processing, which stimulates the exploration of new material systems for high-performance X-ray detection. Recently, two-dimensional (2D) lead halide perovskites (LHPs) have gained a lot of attention as prominent candidate materials for X-ray detection due to their ease of fabrication, superior X-ray absorption, and exceptional optoelectronic properties.<sup>7–9</sup> For instance, (DGA)PbI<sub>4</sub> (DGA = dimethylbiguanide) achieves a prominent sensitivity of  $4869 \mu\text{C Gy}^{-1} \text{cm}^{-2}$  with an ultralow dark current drift ( $I_{\text{drift}}$ ) of  $5.97 \times 10^{-7} \text{ nA cm}^{-1} \text{ s}^{-1} \text{ V}^{-1}$  by applying an external electric field of  $1200 \text{ V mm}^{-1}$ .<sup>10</sup> However, conventional detectors generally require

external electric field application during operation, resulting in high energy consumption and complicated circuit systems that limit their practical application.<sup>11,12</sup> Therefore, exploring self-powered devices for high-performance X-ray detection without external bias is currently a research hotspot.<sup>13–18</sup>

Ferroelectric materials, which exhibit a bulk photovoltaic effect (BPVE) due to their inherent spontaneous polarization ( $P_s$ ), are considered ideal candidates for self-powered X-ray detectors. Compared to materials with a central structure, this BPVE endows the material with a unique intrinsic electric field that promotes the independent separation of photo-excited charge carriers, thereby enabling X-ray detection without an external power source.<sup>19–25</sup> For example, the 2D perovskite ferroelectric (CH<sub>3</sub>OC<sub>3</sub>H<sub>9</sub>N)<sub>2</sub>CsPb<sub>2</sub>Br<sub>7</sub> exhibited a notable sensitivity of  $410 \mu\text{C Gy}^{-1} \text{cm}^{-2}$  under zero bias.<sup>24</sup> Despite the widespread recognition of lead-based ferroelectrics for their excellent optoelectronic properties, the inherent toxicity of Pb seriously threatens the human body and ecosystem. Therefore, exploring lead-free perovskite ferroelectrics with a strong BPVE is of significant importance for achieving prominent and eco-friendly self-powered X-ray detectors. The successful application of the double perovskite (R-MPA)<sub>4</sub>AgBiI<sub>8</sub> (R-MPA = R- $\beta$ -methylphenethylammonium) on self-driven X-ray detectors suggests the huge potential of lead-free halide double perovskites (HDPs) for passive X-ray detectors.<sup>19</sup> At present, there are only a few reports on lead-free HDP ferroelectrics that achieve self-driven X-ray detectors, especially in multilayered lead-free HDPs. In general, the charge transport properties increase with the number of inorganic layers. Therefore, it is necessary to

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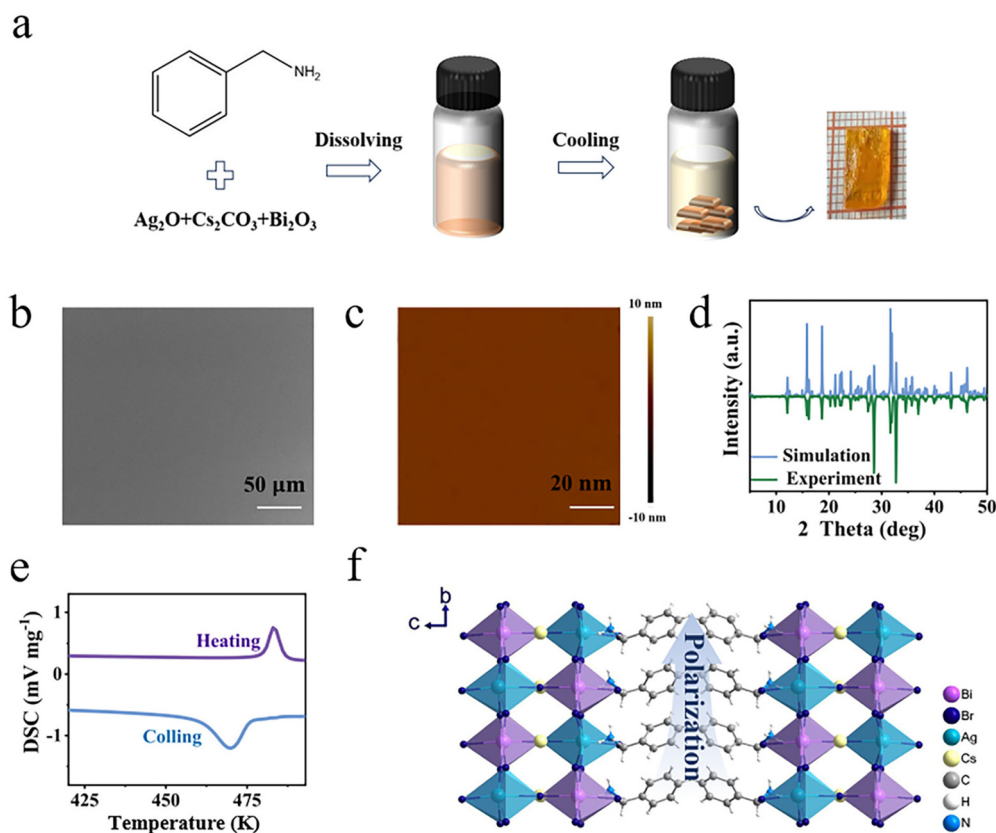
explore the potential of multilayered lead-free HDPs in application in self-driven X-ray detectors.

Herein, we successfully constructed an eco-friendly self-powered X-ray detector using the multilayered hybrid perovskite ferroelectric  $(\text{C}_6\text{H}_5\text{CH}_2\text{NH}_3)_2\text{CsAgBiBr}_7$  (**1**). Owing to the remarkable spontaneous polarization in **1**, the BPVE was induced to separate the charge carriers, yielding a polar photovoltage of approximately 0.5 V. This built-in field gradient facilitates efficient photocarrier dissociation and directional transport, enabling a remarkable sensitivity of  $47.2 \mu\text{C Gy}^{-1} \text{cm}^{-2}$  and a low limit of detection (LoD) of  $716 \text{ nGy s}^{-1}$  without external bias. External voltage enhances charge collection efficiency, further improving sensitivity to  $1154.8 \mu\text{C Gy}^{-1} \text{cm}^{-2}$  at 50 V. This work demonstrated the great potential of multilayered lead-free layered perovskites for manufacturing X-ray detectors and provided a reliable pathway for fabricating eco-friendly self-powered X-ray detectors.

## 2. Results and discussion

High-quality orange single crystals (SCs) of compound **1** were prepared from saturated hydrobromic solutions by adding a certain proportion of  $\text{Cs}_2\text{CO}_3$ ,  $\text{Ag}_2\text{O}$ ,  $\text{Bi}_2\text{O}_3$  and benzylamine

via a slow cooling process (Fig. 1a). As shown in Fig. 1b and c, the scanning electron microscopy (SEM) and atomic force microscopy (AFM) images of **1** SC show a very flat and smooth morphology, which is important for carrier transport in self-powered X-ray detectors. As shown in Fig. 1d, the powder X-ray diffraction (PXRD) pattern of **1** is highly consistent with the simulated PXRD pattern from the crystal structure (CCDC 2010350),<sup>26</sup> verifying the phase purity. A pair of reversible peaks were observed in differential scanning calorimetry (DSC) measurements, indicating that a reversible phase transition occurred at 475 K for **1** (Fig. 1e). Furthermore, the thermodynamic stability of **1** was investigated by thermogravimetric analysis (TG), which revealed excellent thermal stability with a decomposition temperature as high as 570 K (Fig. S1). Fig. 1f shows that **1** adopts a typical Ruddlesden–Popper structure where alternating corner-sharing  $[\text{AgBr}_6]^{5-}$  and  $[\text{BiBr}_6]^{3-}$  octahedral units form the inorganic layers and the organic cations are firmly linked with the inorganic layers via N–H...Br interactions (2.89–3.12 Å). The spontaneous polarization was calculated to be  $3.19 \mu\text{C cm}^{-2}$  along the polar axis (Fig. S2), resulting in the generation of the BPVE. The powerful BPVE provides further support for self-powered X-ray detection. Meanwhile, the piezoelectric response of **1** SC was tested along the direction perpendicular to the polar axis, and the test results



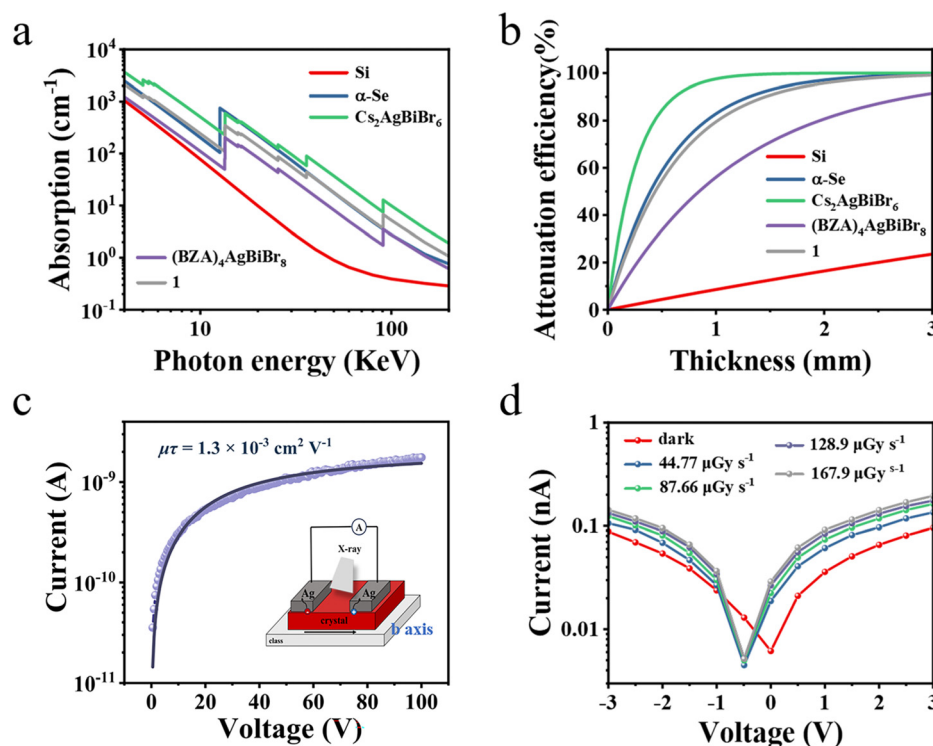
**Fig. 1** (a) Schematic for the growth and a photograph of **1** SC. (b and c) SEM and AFM images of the surface of **1** SC, respectively. (d) The simulated and experimental powder XRD patterns of **1**. (e) DSC curve of **1**. (f) Crystal structure of **1** at room temperature, showing a spontaneous electric polarization along the polar *b*-axis.

further confirmed the polar axis to be in the *b*-axis direction (Fig. S3). Ultraviolet-visible (UV-vis) absorption spectroscopy reveals that the obvious absorption edge of **1** was observed at 533 nm (Fig. S4), where the optical bandgap of **1** was determined to be approximately 2.42 eV by using the *Tauc* plot. The current density–voltage curve along the *b*-axis further demonstrates that the bulk resistivity ( $\rho$ ) of **1** is as high as  $5.66 \times 10^{10} \Omega \text{ cm}$  (Fig. S5). This value is comparable to that of other 2D HDPs, such as  $(\text{BA})_2\text{CsAgBiBr}_7$  ( $1.5 \times 10^{11} \Omega \text{ cm}$ ),<sup>27</sup>  $(\text{I-C}_4\text{H}_8\text{NH}_3)_4\text{AgBiI}_8$  ( $3.04 \times 10^{10} \Omega \text{ cm}$ )<sup>28</sup> and commercial CdZnTe ( $10^{10} \Omega \text{ cm}$ ).<sup>29</sup> Notably, this resistivity is more than two orders of magnitude higher than that of conventional 3D MAPbX<sub>3</sub> (X = Cl, Br, I) perovskite SCs ( $10^7$ – $10^8 \Omega \text{ cm}$ ).<sup>30–33</sup> The exceptionally high resistivity of **1** contributes to its remarkably low dark current, suggesting that it holds significant potential as an effective material for X-ray detection.

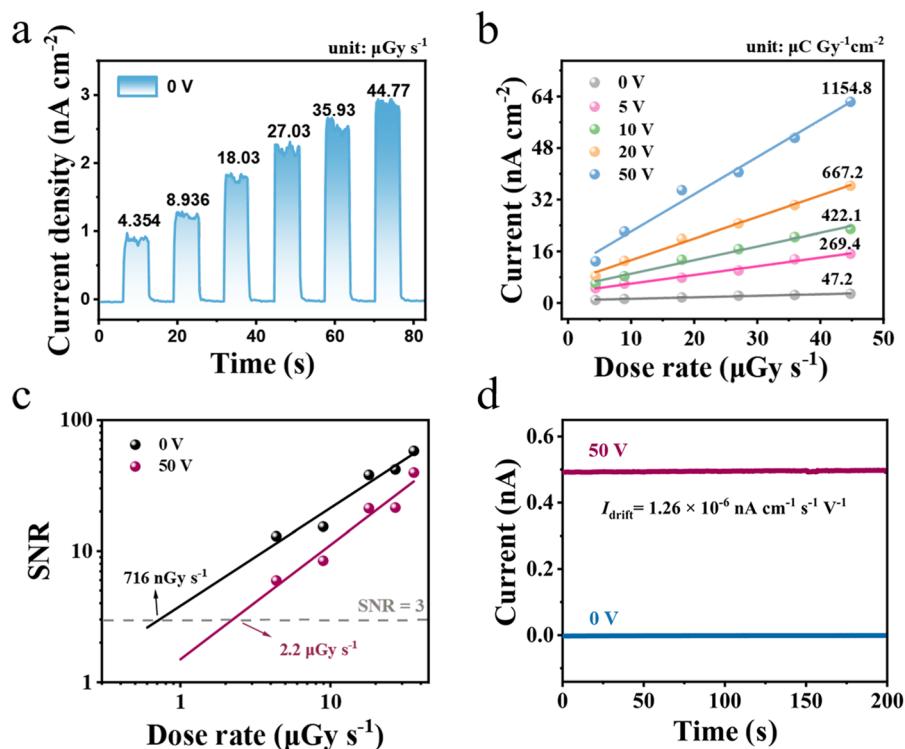
X-ray absorption is an important parameter for X-ray detection. Therefore, we investigate our detector's X-ray absorption capacity by simulating the absorption efficiency across a broad photon energy range based on a photon cross-section database.<sup>34</sup> As depicted in Fig. 2a, **1** exhibits strong X-ray absorption across a wide photon energy range, surpassing that of Si and closely matching that of  $\alpha$ -Se. This high absorption capability stems from the presence of heavy atoms such as Ag, Bi and Br. Due to density differences, the absorption coefficient of **1** ( $2.920 \text{ g cm}^{-3}$ ) is greater than that of  $(\text{BZA})_4\text{AgBiBr}_8$  ( $2.159 \text{ g cm}^{-3}$ ) but slightly lower than that of  $\text{Cs}_2\text{AgBiBr}_6$  ( $4.93 \text{ g cm}^{-3}$ ).<sup>35,36</sup> Fig. 2b presents the relationship between

attenuation efficiency and material thickness when the energy of X-ray photons reaches 50 keV. At a thickness of 2 mm, ferroelectric **1** attenuates approximately 95.8% of incident X-ray photons, significantly outperforming Si (23.4%). Based on its outstanding semiconducting properties and high X-ray absorption efficiency, **1** is believed to be a promising candidate for X-ray detectors. A two-terminal device was fabricated by depositing Ag electrodes on the surface of **1** along the direction of polarization (*b* axis), as illustrated in the inset of Fig. 2c. The mobility-lifetime product ( $\mu\tau$ ), which reflects the efficiency of charge collection, is another key merit for X-ray detectors. The  $\mu\tau$  is estimated by fitting current–voltage (*I*–*V*) characteristics under the exposure of X-rays using Hecht's equation. As shown in Fig. 2c, the  $\mu\tau$  of **1** is calculated to be  $1.3 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1}$ , which is  $10^4$  times higher than that of  $\alpha$ -Se ( $\sim 10^{-7} \text{ cm}^2 \text{ V}^{-1}$ ) and higher than the reported values of  $(\text{BZA})_2(\text{R/S-PPA})\text{BiI}_6$  ( $5.94 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1}$ ),  $(\text{R-MPA})_4\text{AgBiI}_8$  ( $2.2 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1}$ ), and  $(\text{HIA})_2\text{AgBiBr}_8$  ( $1.23 \times 10^{-3}$ ).<sup>19,21,37</sup> A pronounced BPVE along the direction of polarization is observed during *I*–*V* measurements because of the spontaneous polarization of **1** at room temperature. It can be observed that **1** shows an open-circuit voltage of 0.5 V under X-ray irradiation in Fig. 2d. Considering its strong X-ray absorption, excellent charge transport properties and significant BPVE, we conclude that **1** holds promising prospects for next-generation eco-friendly self-powered X-ray detectors.

To further investigate the X-ray detection performance of the **1** SC-based detectors, the photocurrent under X-ray



**Fig. 2** (a) X-ray absorption spectra and (b) corresponding attenuation efficiencies of Si,  $\alpha$ -Se,  $\text{Cs}_2\text{AgBiBr}_6$ ,  $(\text{BZA})_4\text{AgBiBr}_8$ , and **1**. (c) Bias-dependent X-ray conductivity of the **1** SC device. (d) *I*–*V* curves of the **1** device along the *b*-axis in the dark and under X-ray irradiation.



**Fig. 3** (a) Current density–time curves of the **1** SC detector under increased X-ray dose rates at 0 V bias. (b) X-ray-induced current densities as a function of dose rates with external voltages increasing from 0 to 50 V. The slope of the fitting line represents the sensitivity. (c) SNRs of the **1** detector under different dose rates at 0 and 50 V bias. (d) Dark current drift under 0 and 50 V bias, respectively.

irradiation at different external voltages was measured. Notably, due to the pronounced BPVE generated by spontaneous polarization, the device demonstrates a strong response to X-rays even in the absence of an external bias (Fig. 3a). Notably, **1** exhibited a low dark current density ( $J_d$ ) of approximately  $10^{-11}$  A cm<sup>-2</sup>, resulting from the high bulk resistivity. Such low  $J_d$  lays a solid foundation for high-performance X-ray detectors. As illustrated in Fig. 3a, the photocurrent density ( $J_{ph}$ ) exhibits a nearly perfect linear increase as the X-ray dose rate rises from 4.35 to 44.77 μGy s<sup>-1</sup>. The sensitivity was calculated to be 47.2 μC Gy<sup>-1</sup> cm<sup>-2</sup> under 0 V bias by fitting the relationship between the current density ( $J_{ph}-J_d$ ) and dose rate (Fig. 3b). This value not only competes with commercial α-Se (20 μC Gy<sup>-1</sup> cm<sup>-2</sup>), which needs an exceptionally high electric field of 10 μm V<sup>-1</sup>, but also significantly surpasses many lead-free perovskites operating at an applied voltage, such as (CPA)<sub>4</sub>AgBiBr<sub>8</sub> (0.8 μC Gy<sup>-1</sup> cm<sup>-2</sup>, 10 V),<sup>38</sup> (BA)<sub>2</sub>CsAgBiBr<sub>7</sub> (4.2 μC Gy<sup>-1</sup> cm<sup>-2</sup>, 10 V),<sup>27</sup> and (HIA)<sub>2</sub>AgBiI<sub>8</sub> (118 μC Gy<sup>-1</sup> cm<sup>-2</sup>, 10 V).<sup>9</sup> Additionally, the X-ray responses to the dose rate of **1** under varying bias from 5 to 50 V were investigated (Fig. S6–S9). As expected, the sensitivity increases with increasing external electric fields due to enhanced charge collection efficiency, reaching a peak value of 1154.8 μC Gy<sup>-1</sup> cm<sup>-2</sup> at 50 V (Fig. 3b and Table S1). This value surpasses those of most of the HDP detectors and is even comparable to that of the widely known Cs<sub>2</sub>AgBiBr<sub>6</sub> single-crystal detector.<sup>39</sup> LoD

is defined as the lowest dose rate where the signal-to-noise ratio (SNR) equals 3, a critical metric for evaluating X-ray detectors' performance. The plot of SNR versus dose rate is presented in Fig. 3c. By linearly fitting the function of SNR and the dose rate, the LoD of **1** is calculated to be 716 nGy s<sup>-1</sup> at 0 V bias. It should be noticed that this value is much lower (about 7.6 times) than the standard dose rate (5.5 μGy s<sup>-1</sup>) used in medical imaging. When the external bias is enhanced to 50 V, the LoD increases to 2.2 μGy s<sup>-1</sup>, which is attributed to the amplified dark current and noise arising from significant ion migration under high electric fields. Ion migration was further assessed through current drift ( $I_{drift}$ ). Under self-powered operation, the dark current remains exceptionally stable due to negligible ion migration. In contrast, at 50 V bias,  $I_{drift}$  reaches  $1.26 \times 10^{-6}$  nA cm<sup>-1</sup> s<sup>-1</sup> V<sup>-1</sup>, highlighting the unique virtue of self-powered mode in operational stability (Fig. 3d). The radiation stability was further evaluated by measuring the photocurrent of the **1** SC-based detector under continuous X-ray exposure at a dose rate of 1.99 mGy s<sup>-1</sup>. It is remarkable that after a large amount of X-ray dose irradiation, both dark current and photocurrent at 0 V and 50 V bias remained ultra stable (Fig. S10), suggesting the excellent irradiation stability of the **1** SC device. Taking all these advantages into account, the **1** SC-based X-ray detector outperforms most reported HDP detectors and is even comparable to some reported devices, as summarized in Table S1.

### 3. Conclusion

In summary, we have successfully achieved eco-friendly self-powered X-ray detection based on a 2D multilayered double perovskite ferroelectric. Combining the strong BPVE induced by ferroelectric spontaneous polarization with excellent radiation absorption ability, the X-ray detectors based on 1 SC achieve a high sensitivity of  $47.2 \mu\text{C Gy}^{-1} \text{cm}^{-2}$ , an ultralow LoD of  $716 \text{ nGy s}^{-1}$  and remarkable operational stability under zero bias. Furthermore, when an external bias reaches 50 V, the sensitivity is significantly enhanced to a maximum of  $1154.8 \mu\text{C Gy}^{-1} \text{cm}^{-2}$ , surpassing most previously reported HDP detectors. This study not only demonstrates the huge potential of HDPs on self-powered X-ray detection but also provides valuable insights into the development of next-generation “green” self-powered X-ray detectors.

### Author contributions

Q. Gao, K. H. Li, and Y. Y. Zheng prepared the samples, performed data collation, and wrote the manuscript. Q. Gao and K. H. Li carried out the X-ray performance testing. Y. P. Yao and Z.-K. Zhu provided assistance with single crystal analysis. P. P. Yu and J. H. Luo designed and directed this project. All the authors discussed and commented on the manuscript.

### Conflicts of interest

There are no conflicts to declare.

### Data availability

The data supporting the findings of this study are available in the supplementary information (SI). Supplementary information including experimental section, extra experimental data, additional X-ray detection performance, and summary of some reported perovskite-based X-ray detectors is available. See DOI: <https://doi.org/10.1039/d5qi01949d>.

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