

Supporting Information

X-ray Detector with Ultra-low Detection Limit based on Bulk Two-dimensional Perovskite $\text{PEA}_2\text{PbBr}_4$ Single Crystals Grown in HBr Solution

Hao Dong^{a,b†}, Xin Liu^{b,c†}, Hu Wang^{b,c*}, Zhilong Chen^{b,d}, Fenghua Li^{b,c},
Pengxiang Wang^{b,c}, Jie Fu^{a,b}, and Yuchuan Shao^{a,b,c,e*}

^aSchool of Microelectronics, Shanghai University, Shanghai 201899, China

^bLaboratory of Thin Film Optics, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800, China

^cCenter of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China

^dKey Laboratory for Ultrafine Materials of Ministry of Education, Shanghai Engineering Research Center of Hierarchical Nanomaterials, School of Materials Science and Engineering, East China University of Science and Technology, Shanghai 200237, China

^eSchool of Physics and Optoelectronic Engineering, Hangzhou Institute for Advanced Study, UCAS, Hangzhou 310024, China

*Corresponding authors: {wanghu@siom.ac.cn; shaoyuchuan@siom.ac.cn}

†Contributed equally.

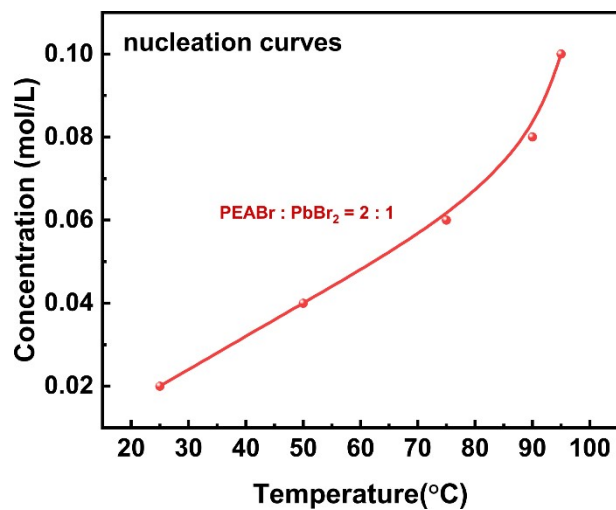
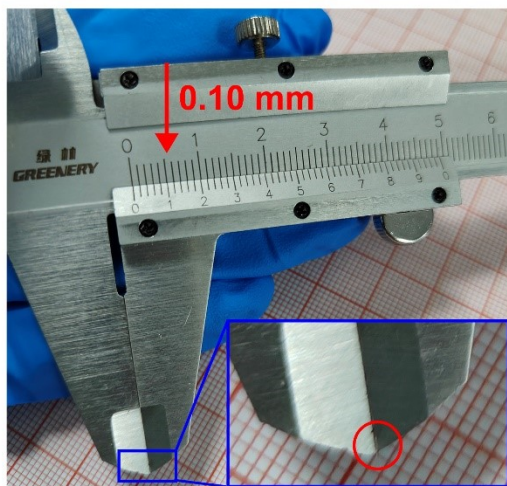
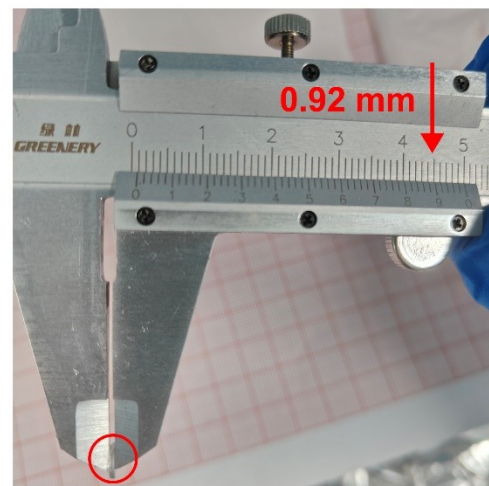


Fig. S1. Nucleation curve of $\text{PEA}_2\text{PbBr}_4$ precursor with solute ratios of $\text{PEABr}:\text{PbBr}_2 = 2:1$ in HBr .



fragile $\text{PEA}_2\text{PbBr}_4$



bulk $\text{PEA}_2\text{PbBr}_4$

Fig. S2. Crystal thickness comparison of fragile and bulk $\text{PEA}_2\text{PbBr}_4$. The fragile $\text{PEA}_2\text{PbBr}_4$ are grown from the precursor solution with stoichiometric ratio of $\text{PEABr}:\text{PbBr}_2 = 2:1$. The bulk $\text{PEA}_2\text{PbBr}_4$ are grown from the precursor solution with stoichiometric ratio of $\text{PEABr}:\text{PbBr}_2 = 2:3$.

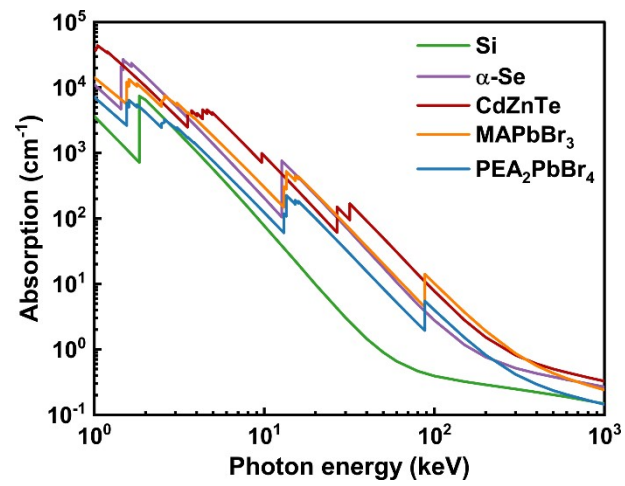


Fig. S3. Linear attenuation coefficients (absorption) of PEA₂PbBr₄, Si, α-Se, CdZnTe and MAPbBr₃ versus different X-ray photons energy.

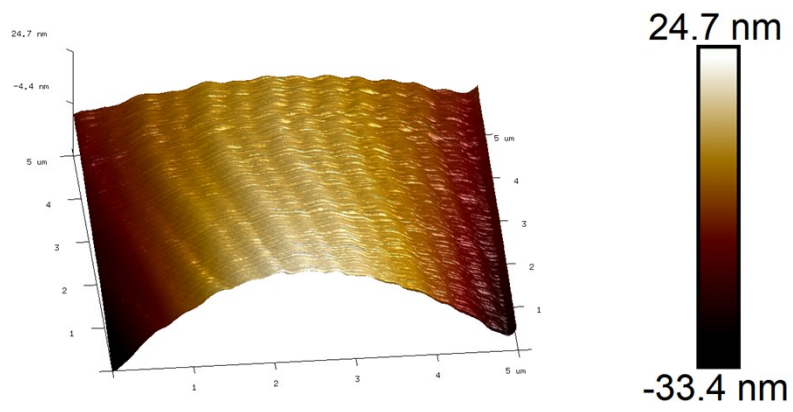


Fig. S4. AFM morphology images of bulk PEA₂PbBr₄ single crystal

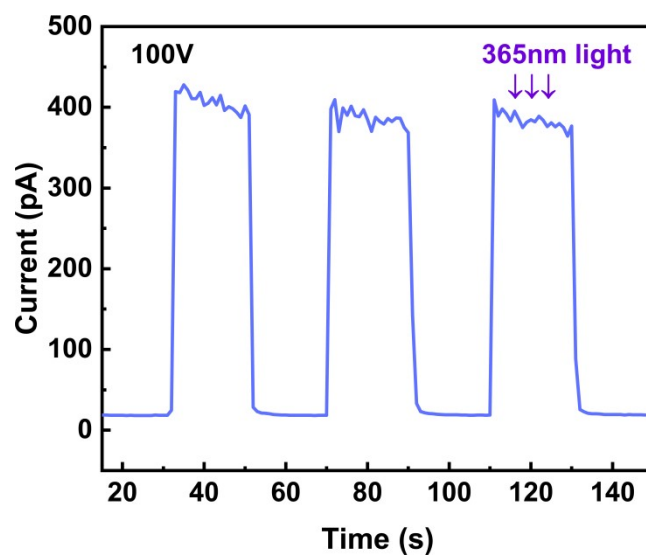


Fig. S5. 365 nm ultraviolet light responses of Au/PEA₂PbBr₄/Au. This measurement serves as a preliminary assessment to determine whether the device exhibits a valid photo response.

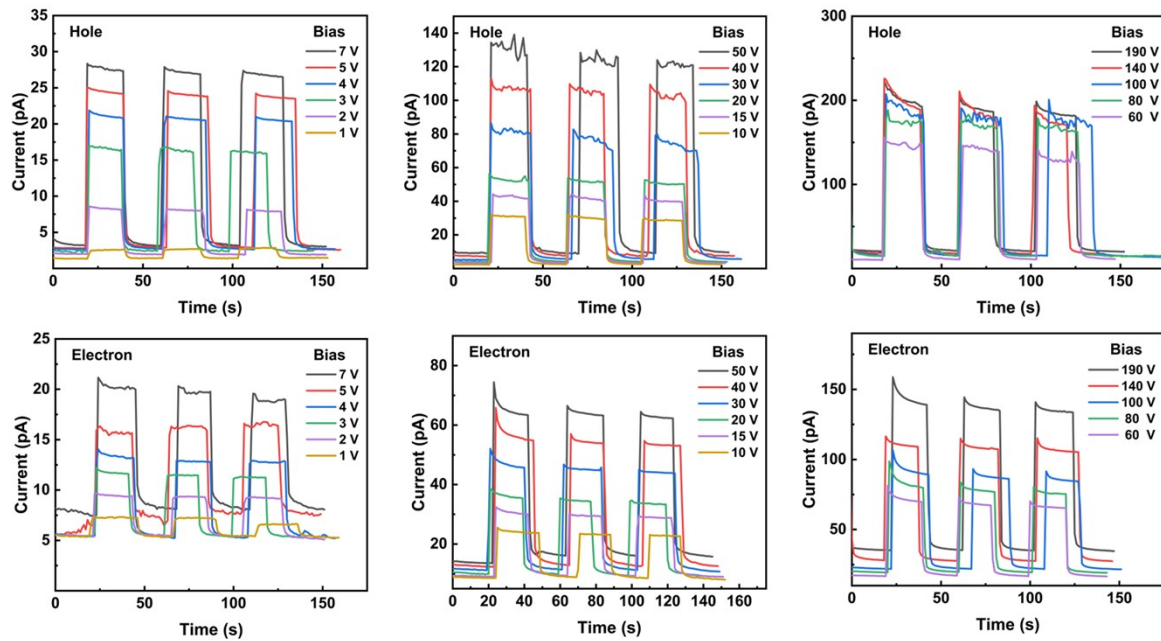


Fig. S6. 365 nm ultraviolet light responses (0.13 mW cm^{-2}) of Au/PEA₂PbBr₄/Au

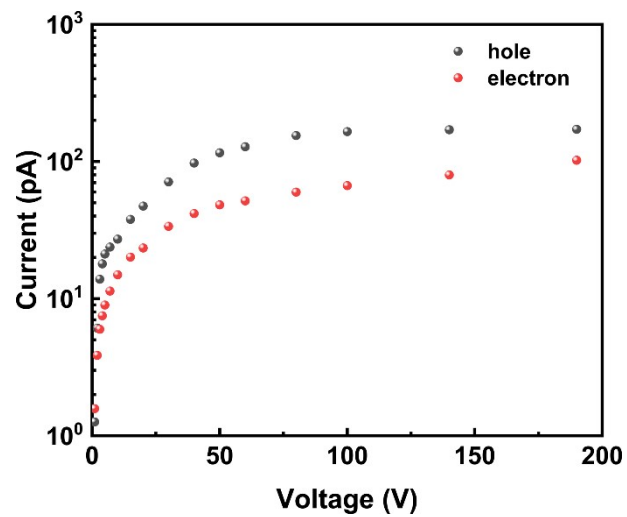


Fig. S7. hole and electron signal currents of Au/PEA₂PbBr₄/Au under 365 nm ultraviolet light

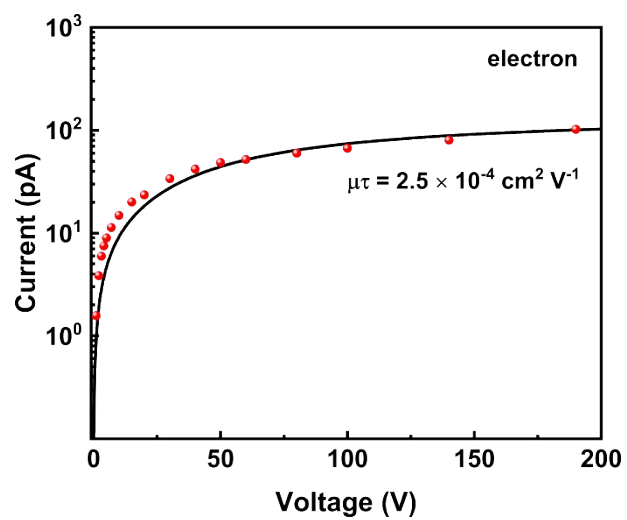


Fig. S8. Photoconductivity measurement for electron under 0.13 mW cm^{-2} ultraviolet LED (365 nm) illumination

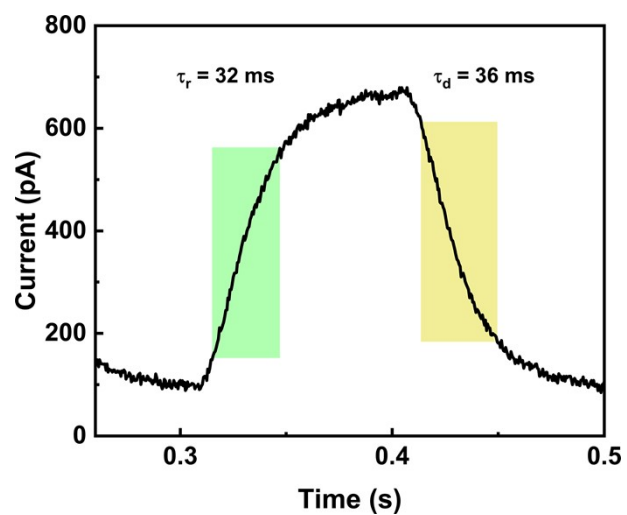


Fig. S9. Temporal response of Au/PEA₂PbBr₄/Au device under ultraviolet LED (365 nm) pulsed illumination

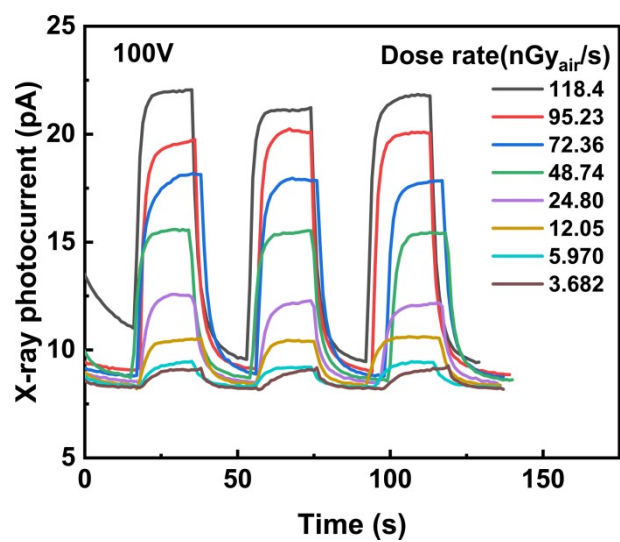


Fig. S10. X-ray responses of Au/PEA₂PbBr₄/Au with dose rates from 118.4 to 3.682 nGy_{air} s⁻¹ at 100V

Table S1. Summary of performance for X-ray detectors based on 2D perovskite single crystals.

Device structure	Crystal growth method	Crystal Thickness [mm]	Resistivity [Ω cm]	Noise current [nA Hz ^{-1/2}]	$\mu\tau$ product [cm ² V ⁻¹]	Sensitivity [μ C Gy _{air} ⁻¹ cm ⁻²]	Bias for sensitivity [V]	Detection limit [nGy _{air} s ⁻¹]	X-ray [keV]	Ref.
Au/ (F-PEA) ₂ PbI ₄ / C60/BCP/Cr	STL ^{a)} in GBL	2	1.36 × 10 ¹²	<0.25	5.1 × 10 ⁻⁴	3,402	200	23	120	1
Au/ [Cu(O ₂ C- (CH ₂) ₃ - (NH ₃) ₂]PbBr ₄ / C60/BCP/Cr	slow evaporation	1.2	8.85 × 10 ¹¹	10 ⁻⁴	/	114,000	960	56	120	2
Au/BA ₂ PbBr ₄ /Au	STL in HBr	0.16	8.51 × 10 ¹¹	/	1.1 × 10 ⁻⁵	726.18	150	8.2	50	3
planar Au/ (PA) ₄ AgBiBr ₈	slow evaporation	2.2	/	/	/	6.89	40	<5,000,00 0	40	4
Au/ (BDA)PbI ₄ / Au	STL in HI	/	/	/	4.43 × 10 ⁻⁴	242	10	430	40	5
Au/ BA ₂ EA ₂ Pb ₃ Br ₁₀ / Au	STL in HBr	2	4.5 × 10 ¹⁰	/	1.0 × 10 ⁻²	6,800	10	5,500	70	6
Au/ (BA) ₂ CsAgBiBr ₇ / Au	STL in HBr	3	1.5 × 10 ¹¹	/	1.21 × 10 ⁻³	4.2	10	/	70	7
planar Au/ (DGA)PbI ₄	STL in HI	/	/	/	4.12 × 10 ⁻³	4869	60	95.4	40	8
Au/ PEA ₂ PbBr ₄ /Au	STL in HBr	1	1.25 × 10 ¹²	10 ⁻⁵	1.0 × 10 ⁻³	2998	100	0.79	30	This work

^{a)}Abbreviations: STL, Solution temperature-lowering.

Table S2. Summary of detection limit for typical 3D perovskite SC X-ray detectors.

Device structure	Detection limit [nGy _{air} s ⁻¹]	X-ray [keV]	Ref.
Au/Al/BCP/C ₆₀ /CsPbBr ₃ /Au	200	30	9
Au/MAPbI ₃ /Au	1.5	22	10
Au/MAPbBr ₃ /C ₆₀ /BCP/Ag or Au	500	22	11
Cr/MAPbBr _{2.94} Cl _{0.06} /C ₆₀ /BCP/C r	7.6	8	12
Au/Cs ₂ AgBiBr ₆ /Au	45.7	50	13

References

- 1 H. Li, J. Song, W. Pan, D. Xu, W. Zhu, H. Wei and B. Yang, *Adv. Mater.*, 2020, **32**, 2003790.
- 2 K. Guo, W. Li, Y. He, X. Feng, J. Song, W. Pan, W. Qu, B. Yang and H. Wei, *Angew. Chem. Int. Ed.*, 2023, **62**, e202303445.
- 3 X. Xu, Y. Wu, Y. Zhang, X. Li, F. Wang, X. Jiang, S. Wu and S. Wang, *ENERGY Environ. Mater.*, 2023, e12487.
- 4 P. Liu, Y. Xiao, Z. Yang, S. Yu and X. Meng, *Opt. Mater.*, 2022, **133**, 112972.
- 5 Y. Shen, Y. Liu, H. Ye, Y. Zheng, Q. Wei, Y. Xia, Y. Chen, K. Zhao, W. Huang and S. (Frank) Liu, *Angew. Chem. Int. Ed.*, 2020, **59**, 14896–14902.
- 6 C. Ji, S. Wang, Y. Wang, H. Chen, L. Li, Z. Sun, Y. Sui, S. Wang and J. Luo, *Adv. Funct. Mater.*, 2020, **30**, 1905529.
- 7 Z. Xu, X. Liu, Y. Li, X. Liu, T. Yang, C. Ji, S. Han, Y. Xu, J. Luo and Z. Sun, *Angew. Chem. Int. Ed.*, 2019, **58**, 15757–15761.
- 8 B. Zhang, T. Zheng, J. You, C. Ma, Y. Liu, L. Zhang, J. Xi, G. Dong, M. Liu and S. (Frank) Liu, *Adv. Mater.*, 2023, **35**, 2208875.
- 9 Z. Chen, H. Wang, F. Li, W. Zhang, Y. Shao and S. Yang, *ACS Appl. Mater. Interfaces*, 2023, **15**, 51370–51379.
- 10 Y. Song, L. Li, W. Bi, M. Hao, Y. Kang, A. Wang, Z. Wang, H. Li, X. Li, Y. Fang, D. Yang and Q. Dong, *Research*, 2020, **2020**, 2020/5958243.
- 11 H. Wei, D. DeSantis, W. Wei, Y. Deng, D. Guo, T. J. Savenije, L. Cao and J. Huang, *Nat. Mater.*, 2017, **16**, 826–833.
- 12 H. Wei, Y. Fang, P. Mulligan, W. Chuirazzi, H.-H. Fang, C. Wang, B. R. Ecker, Y. Gao, M. A. Loi, L. Cao and J. Huang, *Nat. Photonics*, 2016, **10**, 333–339.
- 13 L. Yin, H. Wu, W. Pan, B. Yang, P. Li, J. Luo, G. Niu and J. Tang, *Adv. Opt. Mater.*, 2019, **7**, 1900491.