

Supporting Information

High-performance Self-powered Photodetector Based on Concentric Annular α -FAPbI₃/MAPbI₃ Single Crystals Lateral Heterojunction with Broadband Detectivity

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To obtain FAPbI₃ single crystals, a concentration of 1.2 M is chosen in present work, then the proper temperature range for crystal growth should be carefully considered. The solubility curve of FAPbI₃ in γ -GBL is tested as following figure. In order to obtain FAPbI₃ single crystals via inverse temperature crystallization, the temperature should be at least \sim 65°C. To obtain FAPbI₃ single crystal seeds in a shorter time, we choose 105°C for growth temperature of FAPbI₃ single crystal seeds. However, after obtaining FAPbI₃ single crystal seeds, the temperature should be lower than 105°C but higher than 65°C for subsequent quality FAPbI₃ single crystal wafer growth. In our work, we found the temperature values ranging from 75°C to 85°C is proper. For liquid phase epitaxy of MAPbI₃ single crystal, the same solvent (γ -GBL) and concentration (1.2 M) are chosen, while the temperature is ranging from 88°C to 95°C.

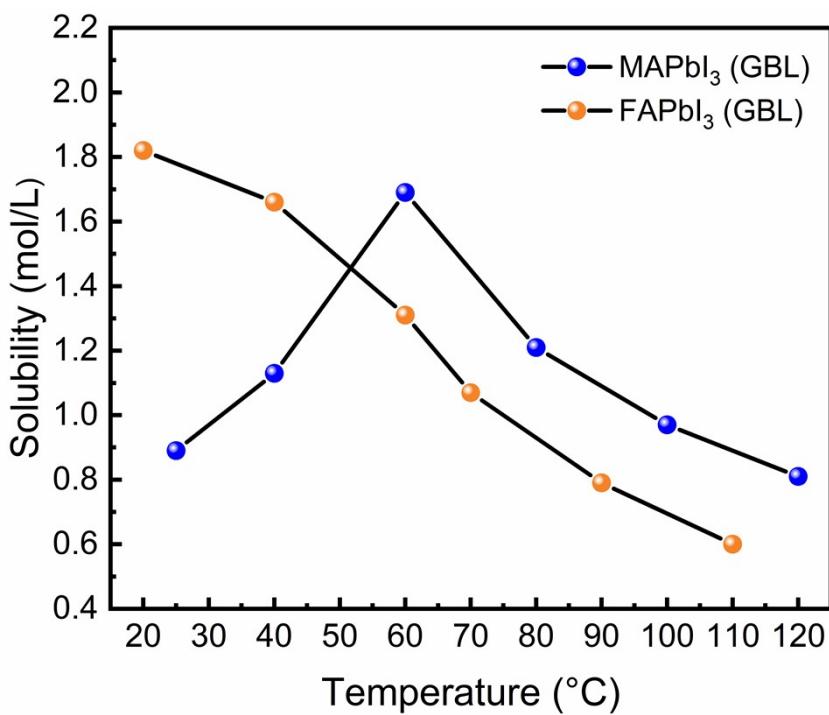


Fig. S1 The solubility of FAPbI₃ and MAPbI₃ in γ -GBL.

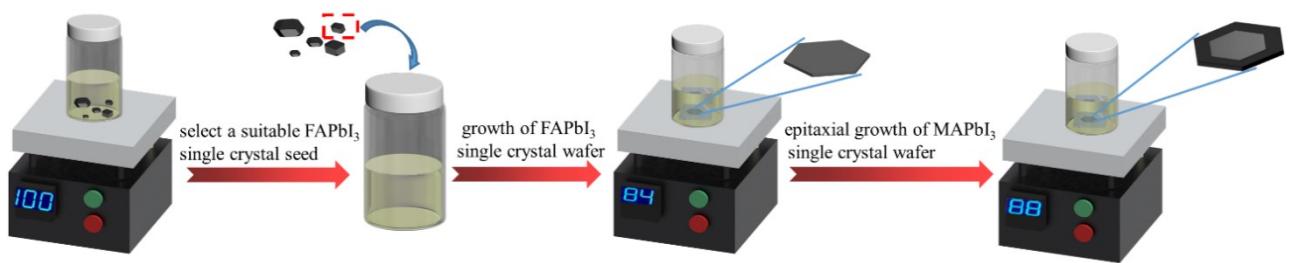


Fig. S2 Flow diagram of epitaxial growth of FAPbI₃/MAPbI₃ SCs concentric annular lateral heterojunction.



Fig. S3 Grow FAPbI₃ SC a) without using the seeds, the inset shows needle-like yellow δ-FAPbI₃ perovskites. b) using the seeds. c) using the SCITC method.

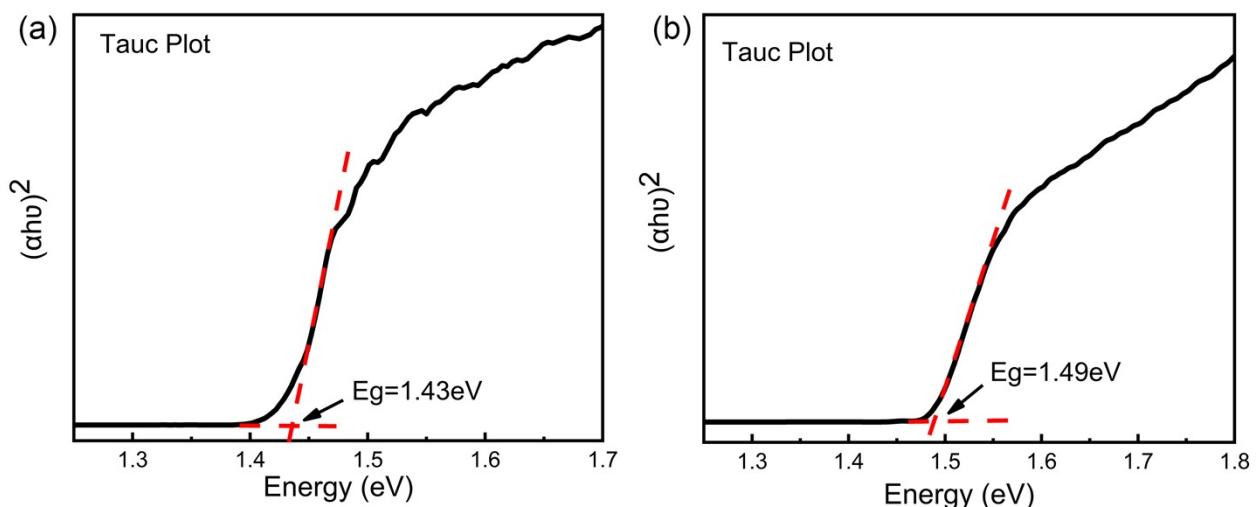


Fig. S4 Tauc plots of c) FAPbI₃ SC and d) MAPbI₃ SC obtained from their absorbance spectra.

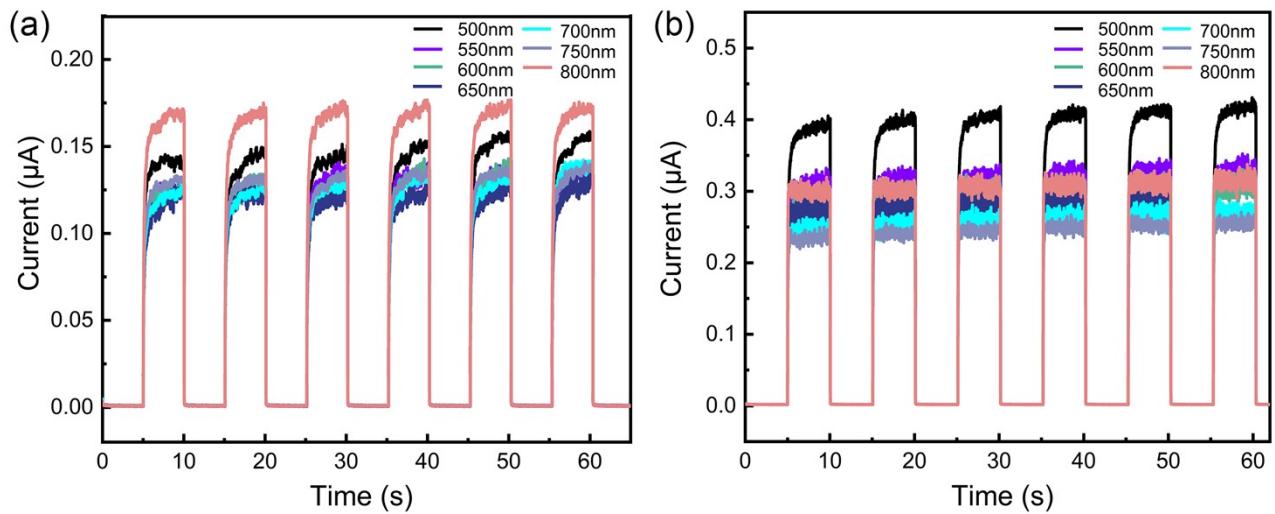


Fig. S5 Time-dependent photoresponse under light illumination (0.19 mW cm^{-2}) ranging from 500 nm to 800 nm at a) -1 V bias. b) -2 V bias.

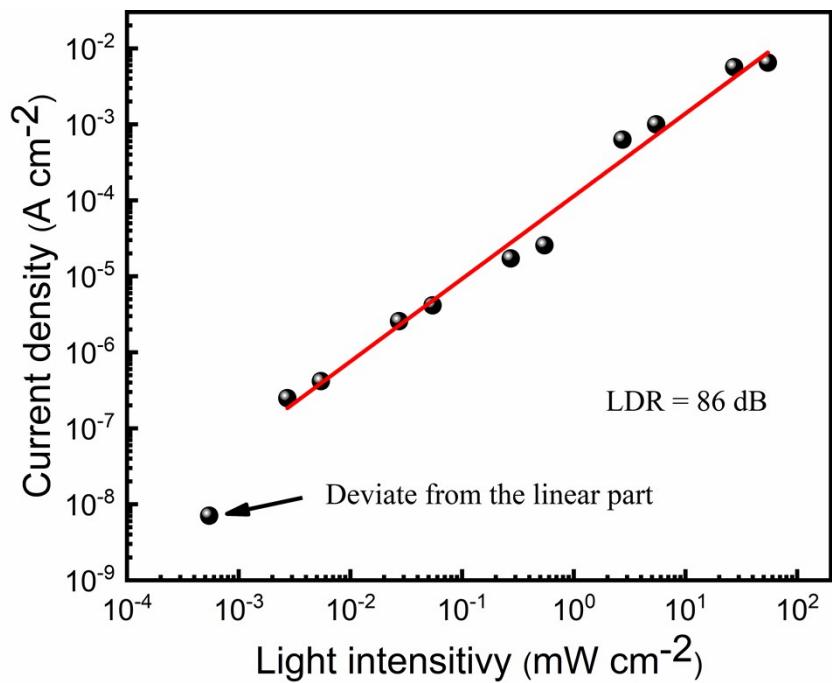


Fig. S6 LDR of the device tested under 530 nm illumination at a bias of -3 V .



Fig. S7 Comparison of stability between pristine FAPbI₃ SC and α -FAPbI₃/MAPbI₃ SC lateral heterojunction.

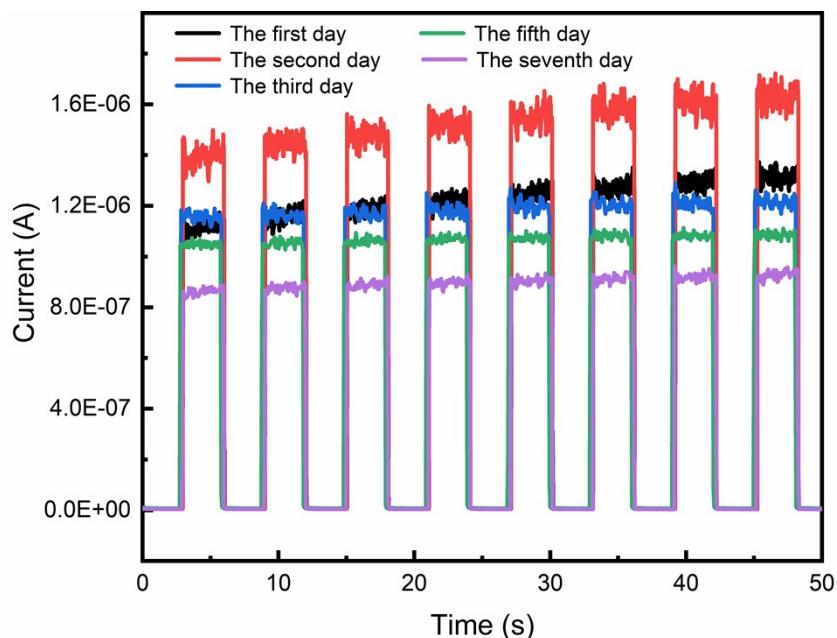


Fig. S8 The stability test of the unencapsulated device.

Table S1 Parameters of perovskites

Species	Lattice constant	Band gap	Crystal system
FAPbI ₃	6.40 Å [1]	1.43 eV	cubic
FAPbBr ₃	6.03 Å [2]	2.26 eV [3]	cubic
FAPbCl ₃	6.2 Å [4]	2.84 eV [4]	cubic
MAPbI ₃	6.276 Å [5]	1.49 eV	cubic
MAPbBr ₃	5.872 Å [6]	2.3 eV [7]	cubic
MAPbCl ₃	5.7 Å [8]	2.85 eV [9]	cubic
CsPbI ₃	6.18 Å [10]	1.67 eV [11]	cubic
CsPbBr ₃	5.87 Å [12]	2.25 eV [13]	cubic
CsPbCl ₃	5.61 Å [14]	3.1 eV [15]	cubic

The lattice misfits can be calculated by the following formula:

$$f(\text{epitaxial layer, substrate}) = [a(\text{epitaxial layer}) - a(\text{substrate})] / a(\text{substrate})$$

Where $a(\text{epitaxial layer})$ is the lattice constant of epitaxial layer, $a(\text{substrate})$ is the lattice constant of substrate, so the lattice misfits between different kinds of perovskites are calculated as follows:

$$f(\text{FAPbBr}_3, \text{FAPbI}_3) = -5.78,$$

$$f(\text{FAPbCl}_3, \text{FAPbI}_3) = -3.13,$$

$$f(\text{MAPbI}_3, \text{FAPbI}_3) = -1.94,$$

$$f(\text{MAPbBr}_3, \text{FAPbI}_3) = -8.25,$$

$$f(\text{MAPbCl}_3, \text{FAPbI}_3) = -10.94,$$

$$f(\text{CsPbI}_3, \text{FAPbI}_3) = -3.44,$$

$$f(\text{CsPbBr}_3, \text{FAPbI}_3) = -8.28,$$

$$f(\text{CsPbCl}_3, \text{FAPbI}_3) = -12.34,$$

$$f(\text{FAPbI}_3, \text{MAPbI}_3) = 1.98,$$

$$f(\text{FAPbBr}_3, \text{MAPbI}_3) = -3.92,$$

$$f(\text{FAPbCl}_3, \text{MAPbI}_3) = -1.21,$$

$$f(\text{MAPbBr}_3, \text{MAPbI}_3) = -6.44,$$

$$f(\text{MAPbCl}_3, \text{MAPbI}_3) = -9.18,$$

$$f(\text{CsPbI}_3, \text{MAPbI}_3) = -1.53,$$

$$f(\text{CsPbBr}_3, \text{MAPbI}_3) = -6.47,$$

$$f(\text{CsPbCl}_3, \text{MAPbI}_3) = -10.61.$$

Table S2 Performance comparison of perovskite-based photodetectors

Material structure	Light wavelength (nm)/power	R (mA W ⁻¹)
MAPbI ₃ microwires/Graphene	520 nm/13.5 mW cm ⁻²	2.2
CsPbI ₃ -CsPbBr ₃ anowire Array	650 nm/0.5 μW cm ⁻²	125
MAPbBr ₃ /Graphene	532 nm/0.66 mW cm ⁻²	1.017 × 10 ⁶
CsPbBr ₃ Monocrystalline Films	530 nm/216 μW cm ⁻²	—
CsPbBr ₃ /CuI	540 nm/216 μW cm ⁻²	1.4
MAPbICl ₂ /TiO ₂ /Si	800 nm/1.13 × 10 ⁻⁴ mW cm ⁻²	870
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D* (Jones)	Response speed	Ref.
1.78 × 10 ⁵	T _r =0.068 s	[16]
—	T _r =0.7 ms, T _d =0.8 ms	[17]
2.02 × 10 ¹³	T _r =50.9 ms, T _d =26.0 ms	[18]
—	T _r =0.4 ms, T _d = 9 ms	[19]
6.2 × 10 ¹⁰	T _r =0.04 ms, T _d =2.96 ms	[20]
6 × 10 ¹²	T _r =116 ms, T _d =50 ms	[21]

References

- [1] L. Qiao, X. Sun and R. Long, *J. Phys. Chem. Lett.*, 2019, **10**, 672.
- [2] M. Ng and J. E. Halpert, *RSC Adv.*, 2020, **10**, 3832.
- [3] N. Arora, M. I. Dar, M. Abdi-Jalebi, F. Giordano, N. Pellet, G. Jacopin, R. H. Friend, S. M. Zakeeruddin and M. Gratzel, *Nano Lett.*, 2016, **16**, 7155.
- [4] S. Pachori, R. Agarwal, A. Shukla, U. Rani and A. S. Verma, *Int. J. Quantum Chem.*, 2021, **121**, e26671.
- [5] T. Baikie, Y. Fang, J. M. Kadro, M. Schreyer, F. Wei, S. G. Mhaisalkar, M. Graetzel and T. J. White, *J. Mater. Chem. A*, 2013, **1**, 5628.
- [6] L. Zhang, L. Liu, P. Zhang, R. Li, G. Zhang and X. Tao, *ACS Appl. Mater. Interfaces*, 2020, **12**, 39834.
- [7] F. Babbe and C. M. Sutter-Fella, *Adv. Energy Mater.*, 2020, **10**, 1903587.
- [8] A. Osherov, Y. Feldman, I. Kaplan-Ashiri, D. Cahen, G. Hodes and *Chem. Mater.*, 2020, **32**, 4223.
- [9] Z. Yuan, W. Huang, S. Ma, G. Ouyang, W. Hu, W. Zhang and *J. Mater. Chem. C*, 2019, **7**, 5442.
- [10] G. Yuan, S. Qin, X. Wu, H. Ding and A. Lu, *Phase Transitions*, 2017, **91**, 38.
- [11] K. Wang, D. Yang, C. Wu, M. Sanghadasa and S. Priya, *Prog. Mater. Sci.*, 2019, **106**, 100580.
- [12] S. Liu, A. R. DeFilippo, M. Balasubramanian, Z. Liu, S.G. Wang, Y.-S. Chen, S. Chariton, V. Prakapenka, X. Luo, L. Zhao, J. S. Martin, Y. Lin, Y. Yan, S. K. Ghose and T. A. Tyson, *Adv. Sci.*, 2021, **8**, 2003046.
- [13] P. Zhang, G. Zhang, L. Liu, D. Ju, L. Zhang, K. Cheng and X. Tao, *J. Phys. Chem. Lett.*, 2018, **9**, 5040.

- [14] M. Naseri, D. M. Hoat, R. Ponce-Pérez, J. F. Rivas-Silva and G. H. Cocoletzi, *Chem. Phys.*, 2020, **531**, 110654.
- [15] Q. Yao, J. Zhang, K. Wang, L. Jing, X. Cheng, C. Shang, J. Ding, W. Zhang, H. Sun and T. Zhou, *J. Mater. Chem. C*, 2021, **9**, 7374.
- [16] J. Ding, H. Fang, Z. Lian, Q. Lv, J. L. Sun and Q. Yan, *Nanoscale*, 2018, **10**, 10538.
- [17] M. Wang, W. Tian, F. Cao, M. Wang and L. Li, *Adv. Funct. Mater.*, 2020, **30**, 1909771.
- [18] Y. Zou, T. Zou, C. Zhao, B. Wang, J. Xing, Z. Yu, J. Cheng, W. Xin, J. Yang, W. Yu, H. Dong and C. Guo, *Small*, 2020, **16**, 2000733.
- [19] Z. Yang, Q. Xu, X. Wang, J. Lu, H. Wang, F. Li, L. Zhang, G. Hu and C. Pan, *Adv. Mater.*, 2018, **30**, 1802110.
- [20] Y. Zhang, S. Li, W. Yang, M. K. Joshi and X. Fang, *J. Phys. Chem. Lett.*, 2019, **10**, 2400.
- [21] F. Cao, Q. Liao, K. Deng, L. Chen, L. Li and Y. Zhang, *Nano Res.*, 2018, **11**, 1722.