

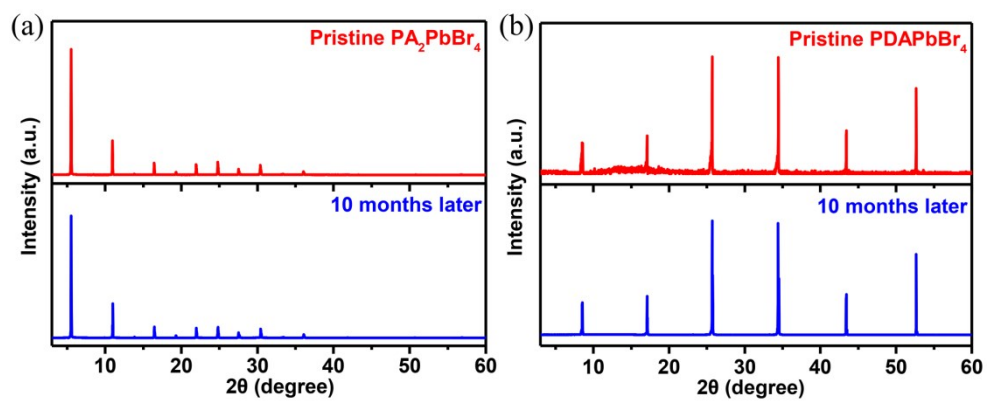
## Supporting Information

### **Diamine Tailored Smooth and Continuous Perovskite Single Crystal with Enhanced Photoconductivity**

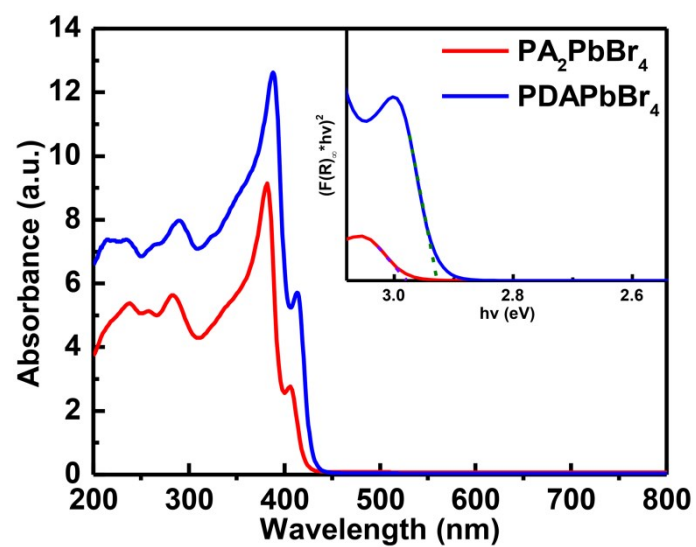
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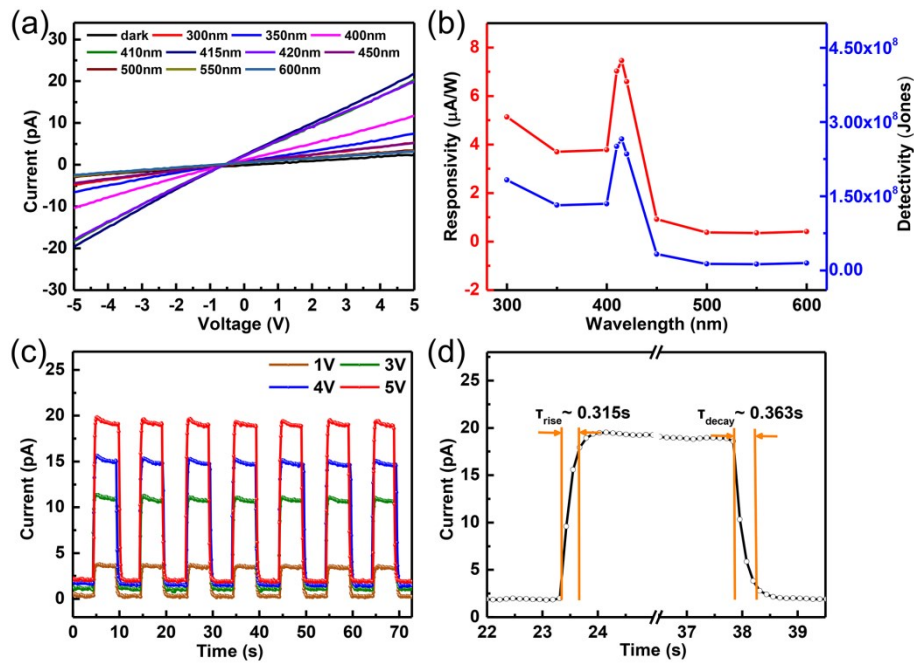
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**Fig. S1** Time-dependent XRD of (a)  $\text{PA}_2\text{PbBr}_4$  and (b)  $\text{PDAPbBr}_4$  single crystals. The XRD patterns of  $\text{PA}_2\text{PbBr}_4$  and  $\text{PDAPbBr}_4$  single crystals exhibit no obvious change when stored in the ambient atmosphere for ten months, indicating good stability.

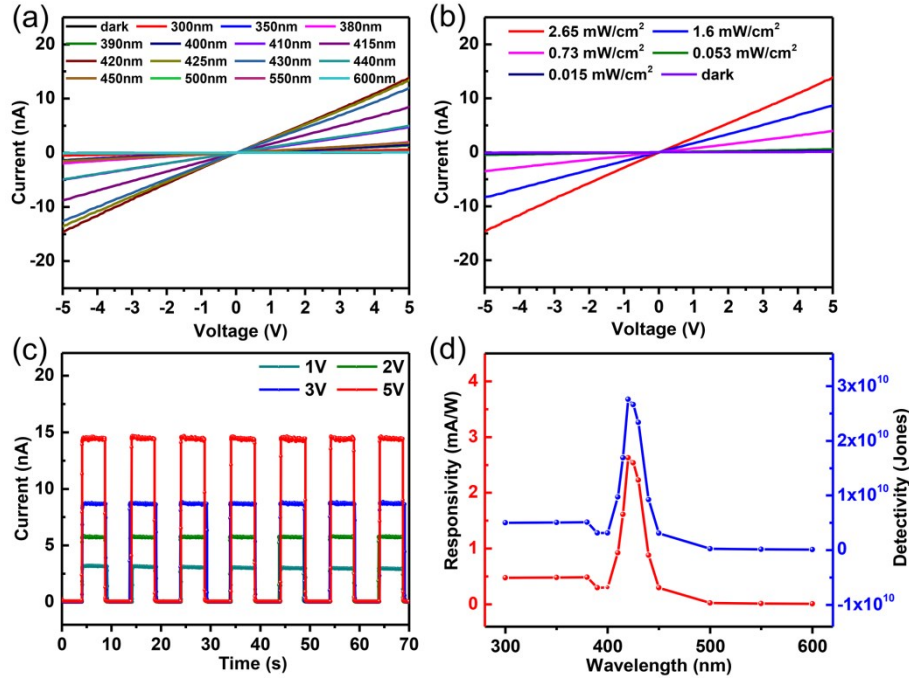


**Fig. S2** Absorption spectra of  $\text{PA}_2\text{PbBr}_4$  and  $\text{PDAPbBr}_4$  perovskites at room temperature; the inset shows the band gaps determined from a Tauc plot. The sharp absorption edges of  $\text{PA}_2\text{PbBr}_4$  and  $\text{PDAPbBr}_4$  locate at 416 nm and 423 nm, affording the bandgap ( $E_g$ ) of 2.98 eV and 2.93 eV by extrapolating the high-energy edge to the energy axis.



**Fig. S3** Optoelectronic performance of  $\text{PA}_2\text{PbBr}_4$  perovskite-based photodetector. (a)  $I$ - $V$  characteristics of the  $\text{PA}_2\text{PbBr}_4$ -based device in the dark, and under light illumination at different wavelengths. The corresponding optical power values are 0.57, 1.35, 2.45, 2.55, 2.60, 2.65, 3.07, 2.78, 2.37, and 1.98  $\text{mW cm}^{-2}$  for the given wavelengths of 300, 350, 400, 410, 415, 420, 450, 500, 550, and 600 nm, respectively. (b) Photoresponsivity and detectivity of  $\text{PA}_2\text{PbBr}_4$ -based device as a function of incident light wavelength. (c) Time-resolved photoresponse of the  $\text{PA}_2\text{PbBr}_4$ -based device under 415 nm light with 2.6  $\text{mW cm}^{-2}$  light intensity. (d) Rise and decay curves of  $\text{PA}_2\text{PbBr}_4$ -based device. The optimum irradiation wavelength of the  $\text{PA}_2\text{PbBr}_4$  photodetector was determined to be 415 nm via characterizing the wavelength-dependent photocurrent, which agrees well with the light absorption. The current of  $\text{PA}_2\text{PbBr}_4$  perovskite is extremely low ( $2.5 \times 10^{-12}$  A) in the dark, and could be increased to only 0.022 nA under light intensity of 2.6  $\text{mW cm}^{-2}$  at a bias of 5V. Hence, a lower on-off current ratio of 9 was obtained for a  $\text{PA}_2\text{PbBr}_4$  photodetector. The device showed the highest responsivity of  $7.45 \times 10^{-3}$   $\text{mA W}^{-1}$  and detectivity of  $2.66 \times 10^8$  Jones under 415 nm light with 2.6  $\text{mW cm}^{-2}$  light intensity. Moreover, the cycling behavior of

PA<sub>2</sub>PbBr<sub>4</sub> perovskite also indicates good stability. The response time of the device was determined to be 315 ms and 363 ms for rise and decay times, respectively.



**Fig. S4** Optoelectronic performance of PDAPbBr<sub>4</sub> perovskite-based photodetector. (a) *I-V* characteristics of the PDAPbBr<sub>4</sub>-based device in the dark, and under light illumination at different wavelengths. The corresponding optical power values are 0.57, 1.36, 1.96, 2.22, 2.45, 2.55, 2.6, 2.65, 2.66, 2.69, 2.81, 3.07, 2.78, 2.37, and 1.98 mW cm<sup>-2</sup> for the given wavelengths (unit of 300, 350, 380, 390, 400, 410, 415, 420, 425, 430, 440, 450, 500, 550, and 600 nm, respectively). (b) *I-V* characteristics of the PDAPbBr<sub>4</sub>-based device in the dark and under 420 nm light illumination at different intensities. (c) Time-resolved photoresponse of the PDAPbBr<sub>4</sub>-based device using 420 nm light at an intensity of 2.65 mW cm<sup>-2</sup> with different bias. (d) Photoresponsivity and detectivity of PDAPbBr<sub>4</sub>-based device as a function of incident light wavelength. The optimum irradiation wavelength of the PDAPbBr<sub>4</sub> photodetector was determined to be 420 nm via characterizing the wavelength-dependent photocurrent, which agreed well with the light absorption. The current of PDAPbBr<sub>4</sub> perovskite is low ( $57 \times 10^{-12}$  A) in the dark, and could be enhanced to 13.99 nA under light intensity of 2.65 mW cm<sup>-2</sup> at a bias of 5V. Hence, a higher on-off current ratio of 246 was obtained for the PDAPbBr<sub>4</sub> photodetector. The device showed responsivity of 2.63 mA W<sup>-1</sup> and

detectivity of  $2.76 \times 10^{10}$  Jones under 420 nm light with intensity of  $2.65 \text{ mW cm}^{-2}$ . The cycling behavior of  $\text{PDPbBr}_4$  perovskite also indicates good stability.