Electronic Supplementary Information

Incorporating Mixed Cations in Quasi-2D Perovskites for High-Performance and Flexible Photodetectors

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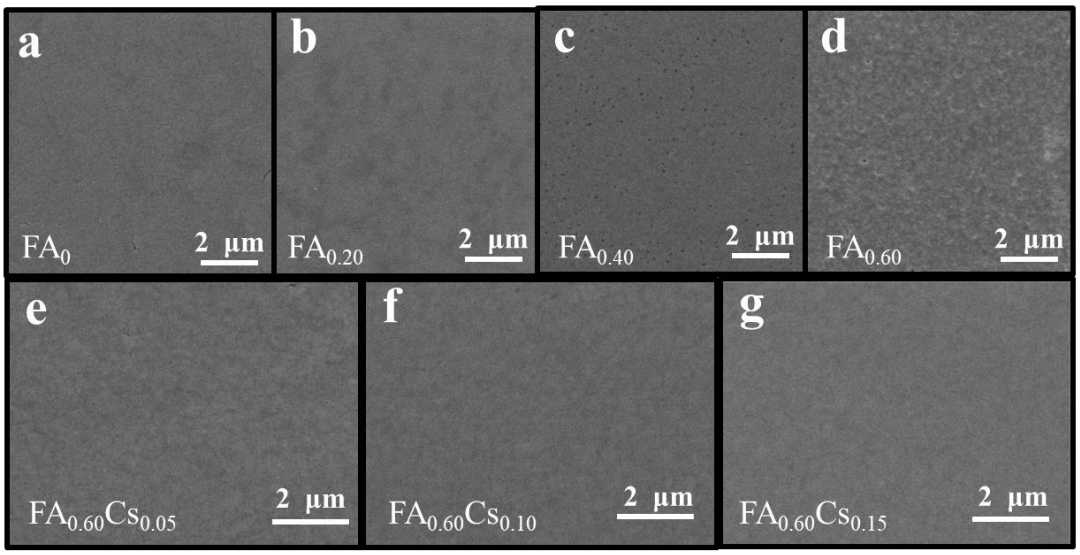

Fig. S1 SEM images of (a)-(d) samples FA_x (x = 0, 0.20, 0.40 and 0.60) and (e)-(g) samples $FA_{0.6}Cs_y$ (y = 0, 0.05, 0.10 and 0.15).

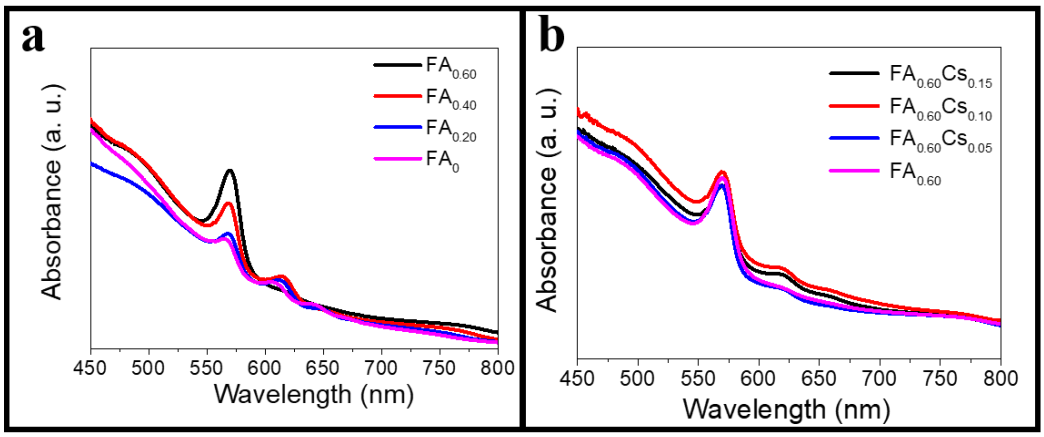

Fig. S2 Absorption spectra of the samples FA_x (x = 0, 0.20, 0.40 and 0.60) and $FA_{0.60}Cs_y$ (y = 0, 5%, 10% and 15%).

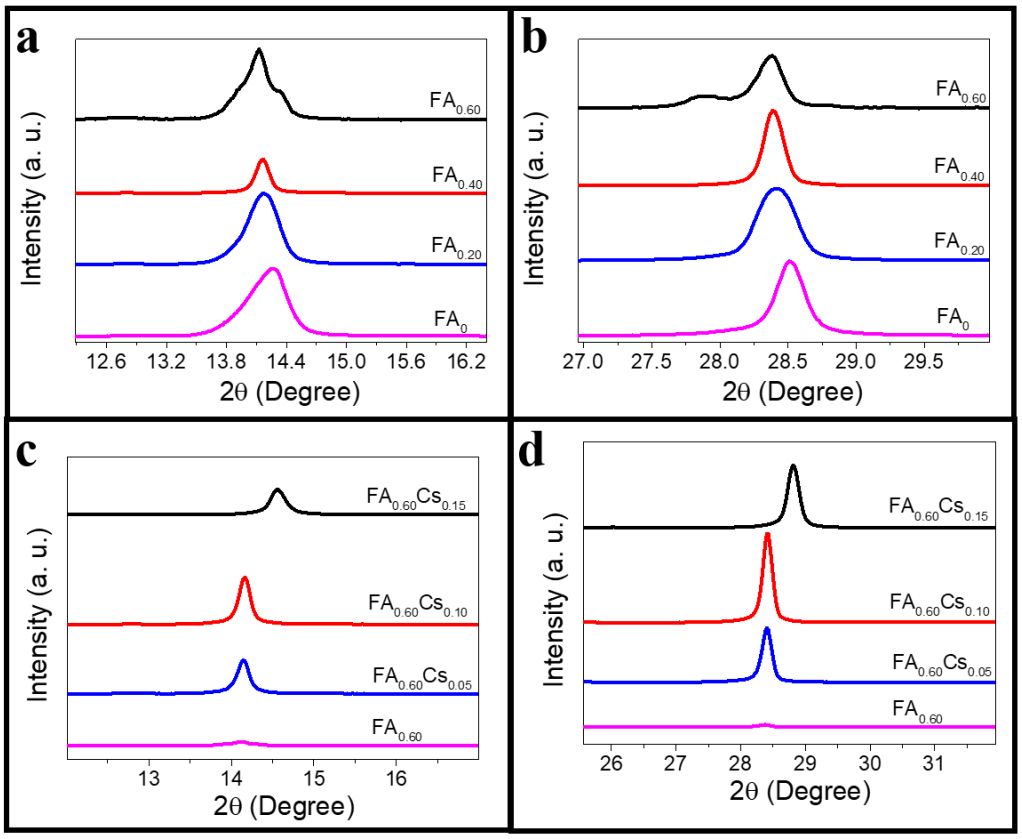

Fig. S3 The high-resolution XRD patterns of all the samples. (a) (111) and (b) (220) crystallographic plane of samples FA_x (x = 0, 0.20, 0.40 and 0.60). (c) (111) and (d) (220) crystallographic plane of samples $FA_{0.60}Cs_y$ (y = 0, 5%, 10% and 15%).

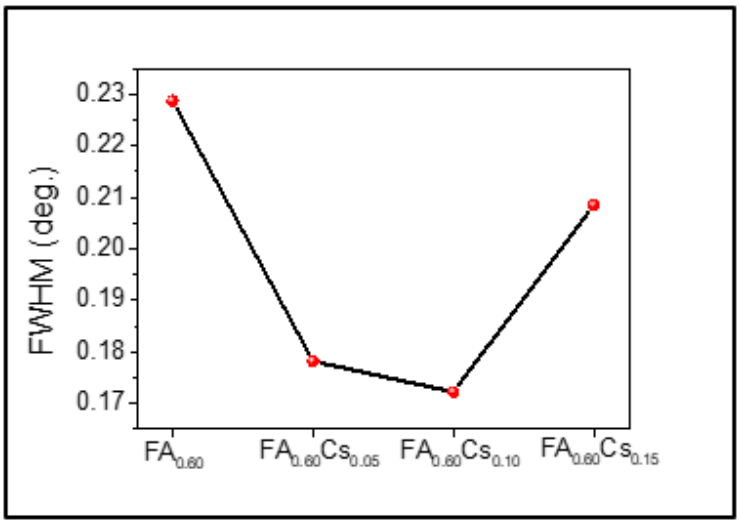

Fig. S4 Full width at half-maximum (FWHM) of the (220) reflection as a function of precursor solvent used for thin film fabrication.

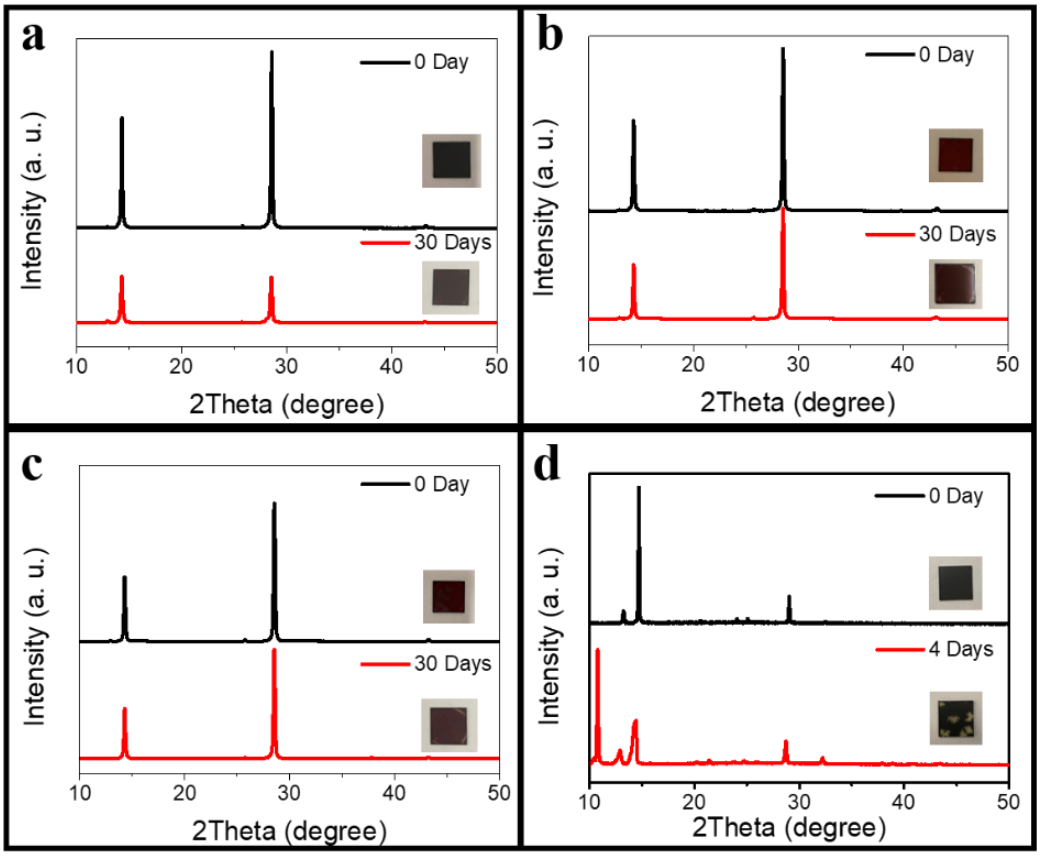

Fig. S5 XRD patterns of all the samples showing the stability of the 2D layered perovskite films and 3D bulk perovskite film. (a) $FA_{0.60}Cs_{0.05}$. (b) $FA_{0.60}Cs_{0.10}$. (c) $FA_{0.60}Cs_{0.15}$. (d) $MAPbI_3$. Inset: optical images of fresh and aged samples after exposure in humidity environments for 30 days or 4 days.

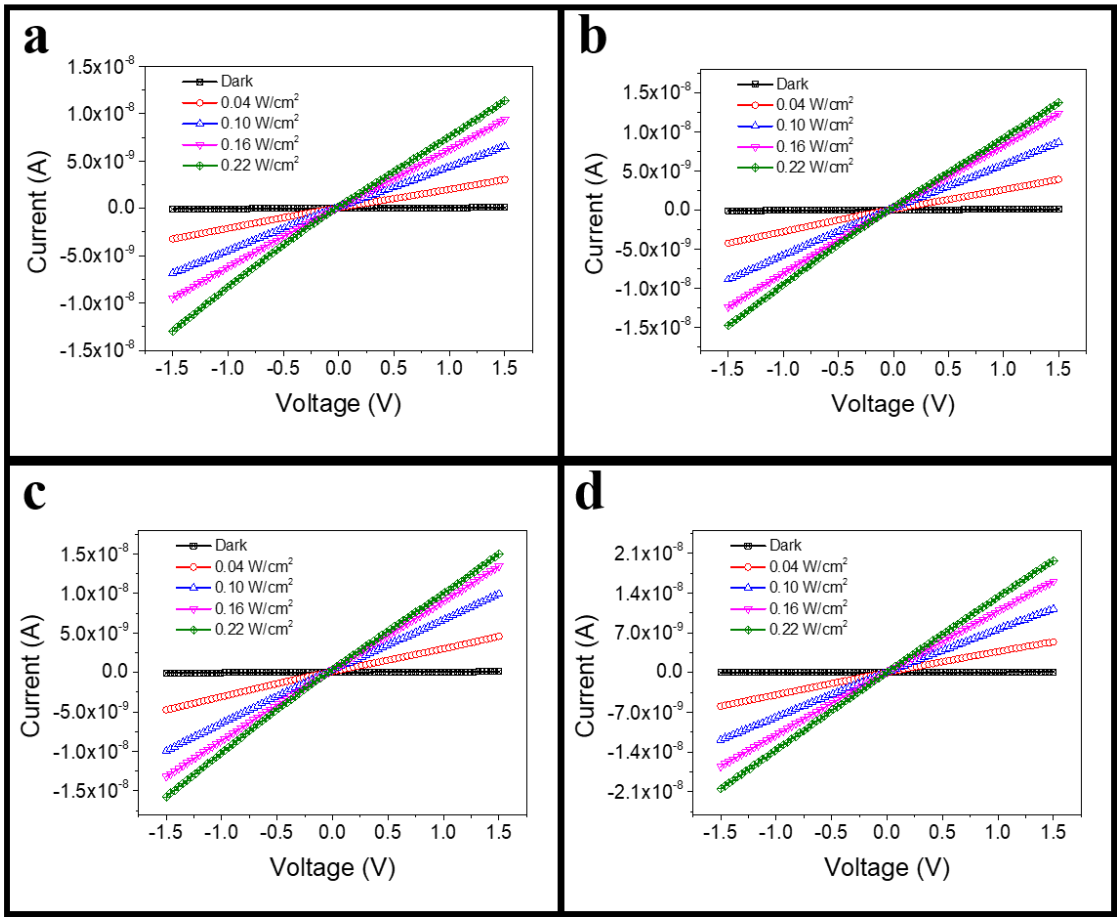

Fig. S6 *I-V* curves of all the samples with and without light illumination. (a) FA_0. (b) $FA_{0.20}$. (c) $FA_{0.40}$. (d) $FA_{0.60}$.

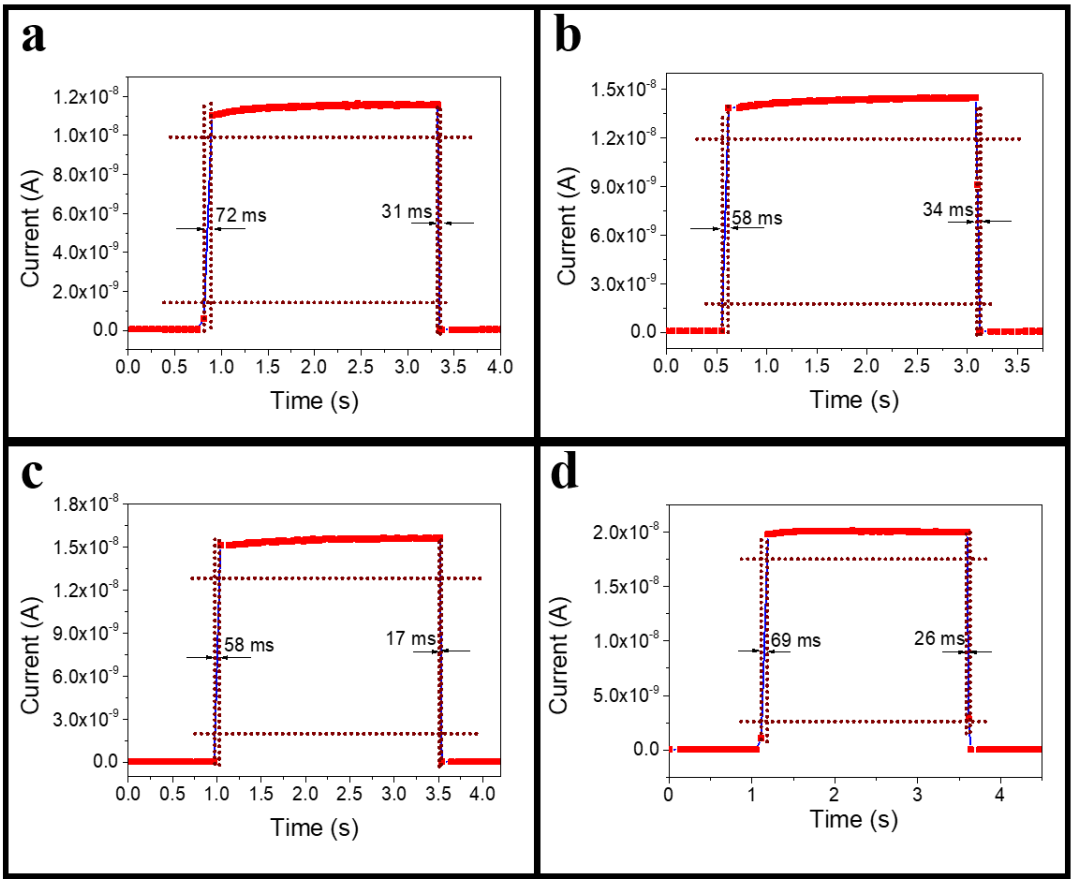

Fig. S7 High resolution current *vs*. time under modulated incident light to determine response times. (a) FA_0. (b) $FA_{0.20}$. (c) $FA_{0.40}$. (d) $FA_{0.60}$.

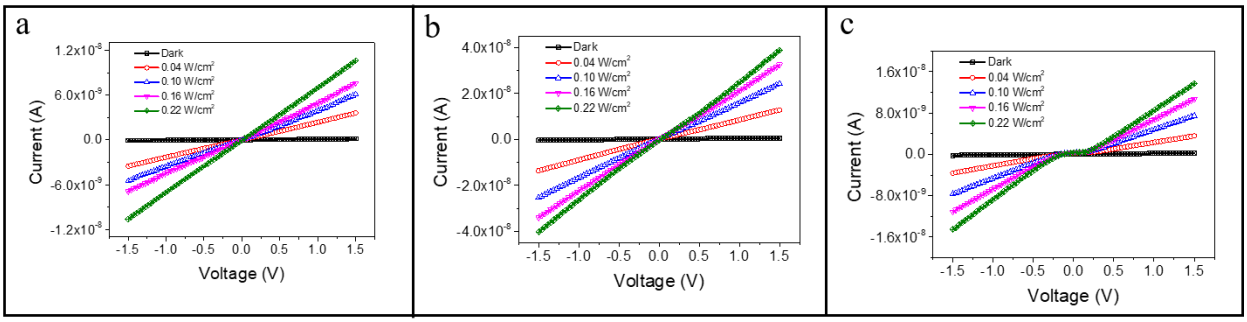

Fig. S8 *I-V* curves of all the samples with and without light illumination. (a) $FA_{0.60}Cs_{0.05}$. (b) $FA_{0.60}Cs_{0.10}$. (c) $FA_{0.60}Cs_{0.15}$.

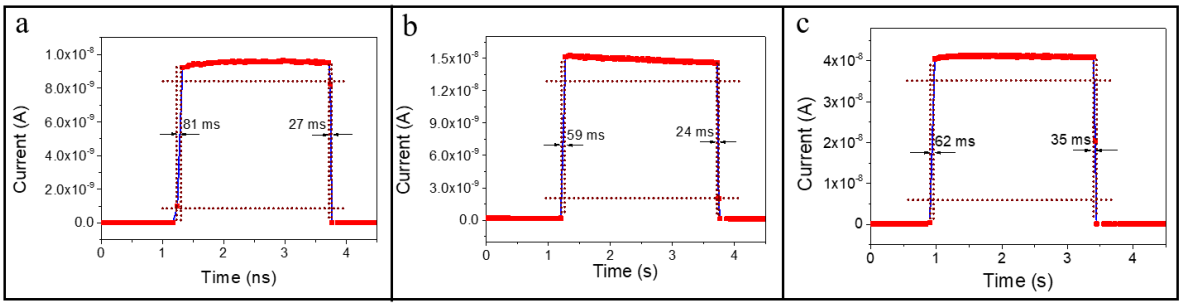

Fig. S9 High resolution current *vs*. time under modulated incident light to determine response times. (a) $FA_{0.60}Cs_{0.05.}$ (b) $FA_{0.60}Cs_{0.10.}$ (c) $FA_{0.60}Cs_{0.15.}$

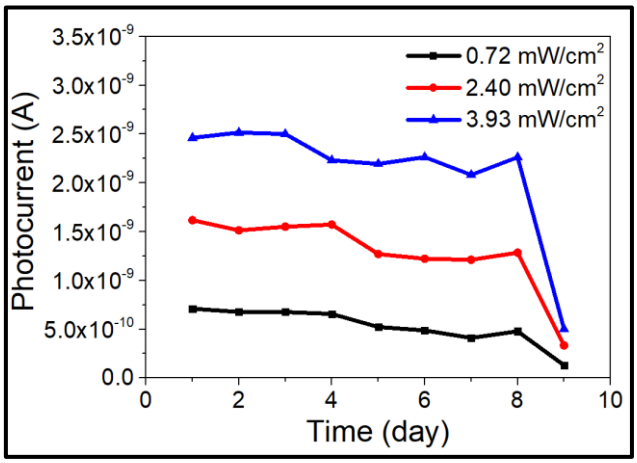

Fig. S10 Operation stability test. Photocurrent as a function of time monitored in humid environments for 9 days (50 % humidity).

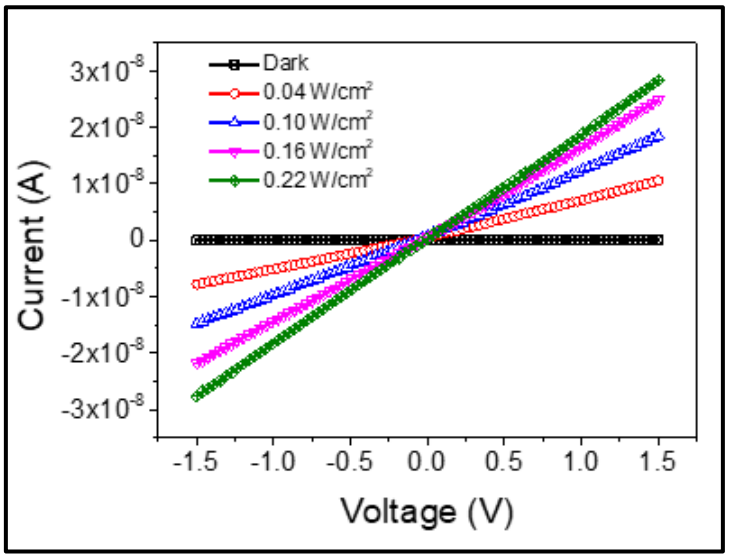

Fig. S11 *I-V* curves of the sample $FA_{0.60}Cs_{0.10}$ on PI substrate with and without light illumination.

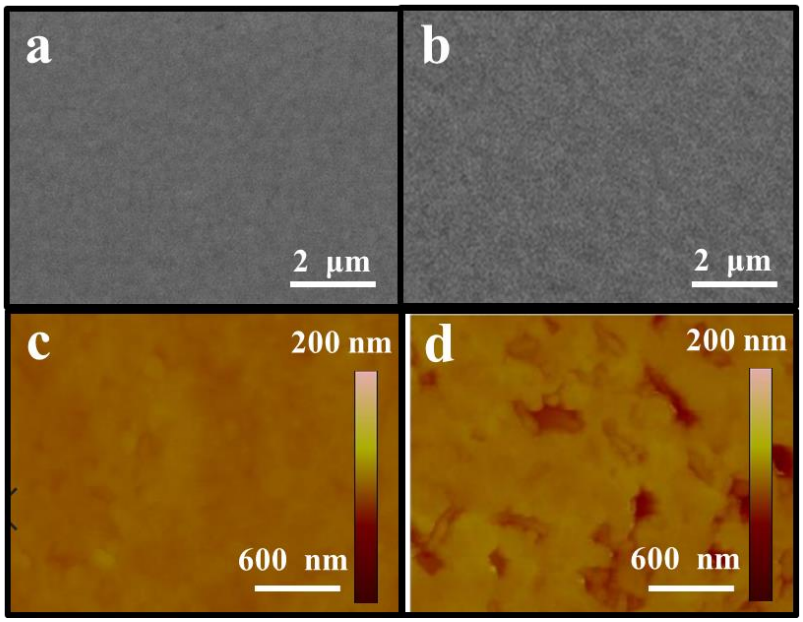

Fig. S12 Characterization of the morphology of $FA_{0.60}Cs_{0.10}$ on glass and PI substrates. (a)-(b) SEM images. (c)-(d) AFM images.

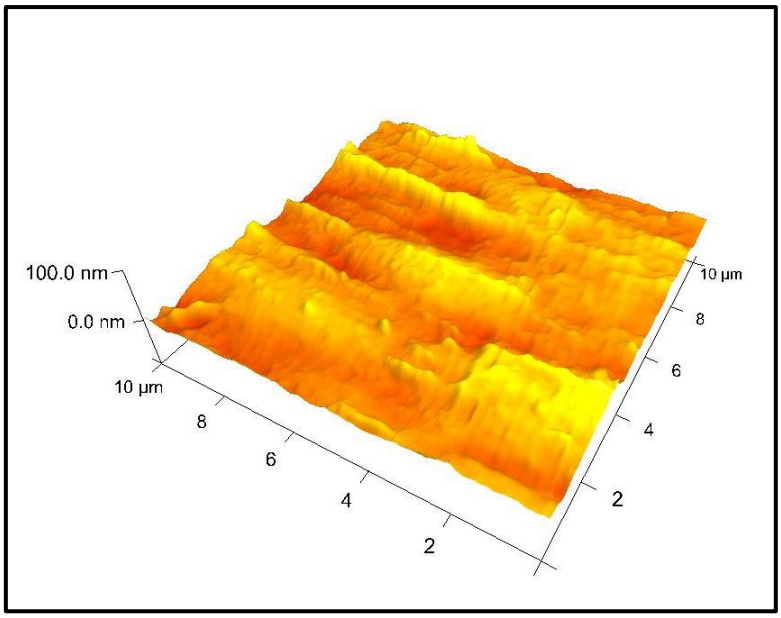

Fig. S13 Atomic force microscopy (AFM) image of the flexible PI substrate, showing the relatively rough surface of the substrate.

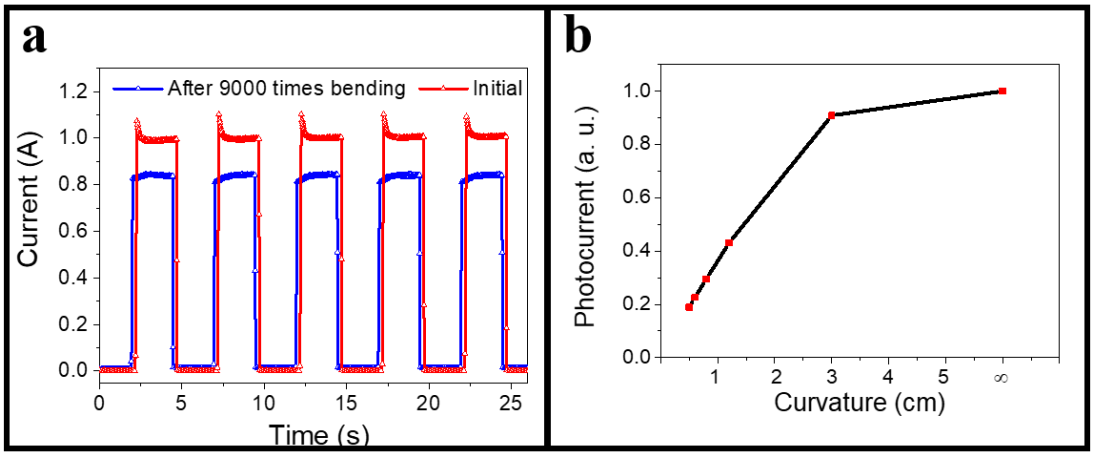

Fig. S14 Photodetection measurements under mechanical bending. (a) Normalized photocurrent as a function of time under chopped light illumination. The photocurrent is normalized to the photocurrent of the initial state without any bending. (b) Normalized photocurrent as a function of bending curvatures after 9000 bending circles. The photocurrent is normalized to the photocurrent without any bending.

Table S1. The time-resolved photoluminescence (TRPL) decay lifetimes of FA_x and $FA_{0.60}Cs_y$ 2D perovskite films.

FA_x	**x = 0**	**x = 0.20**	**x = 0.40**	**x = 0.60**
τ (ns)	4.05	117.9	91.8	144.6
$FA_{0.60}Cs_y$	**y = 0.05**	**y = 0.10**	**y = 0.15**	
τ (ns)	133.2	210.5	180.5	

Table S2. Comparison of various figure of merits of different 2D layered perovskites based photodetectors.

2D Perovskites	Light	Bias	Dark current	Responsivity	Detectivity	Rise time	Decay Time	Ref
FA_0 film	532 nm	1.5V	9.07×10^{-11} A	72 mA/W	3.61×10^{10} Jones	72 ms	31 ms	Our work
$FA_{0.20}$ film			1.18×10^{-10} A	94 mA/W	4.13×10^{10} Jones	58 ms	34 ms	
$FA_{0.40}$ film			9.66×10^{-11} A	150 mA/W	7.29×10^{10} Jones	58 ms	17 ms	
$FA_{0.60}$ film			6.28×10^{-11} A	217 mA/W	1.31×10^{11} Jones	69 ms	26 ms	
$FA_{0.60}Cs_{0.05}$ film			1.44×10^{-10} A	223 mA/W	8.88×10^{10} Jones	81 ms	27 ms	
$FA_{0.60}Cs_{0.10}$ film on rigid glass substrate			6.72×10^{-10} A	351 mA/W	8.36×10^{10} Jones	59 ms	24 ms	
$FA_{0.60}Cs_{0.15}$ film			2.85×10^{-10} A	212 mA/W	6.00×10^{10} Jones	62 ms	35 ms	
$FA_{0.60}Cs_{0.10}$ film on flexible PI substrate			1.27×10^{-12} A	397 mA/W	1.68×10^{12} Jones	43 ms	22 ms	
$(iBA)_2(MA)_3Pb_4I_{13}$ film (Hot casting treated)	532 nm	1.5 V	---	117.09 mA/W	---	16 ms	15 ms	[27]
$(BA)_2PbI_4$ film	White light	30 V	1×10^{-12} to 1×10^{-11} A	3 mA/W	---	28.4 ms	27.5 ms	[24]
$(BA)_2(MA)Pb_2I_7$ film				7.31 mA/W	---	8.4 ms	7.5 ms	
$(BA)_2(MA)_2Pb_3I_{10}$ film				12.78 mA/W	---	10.0 ms	7.5 ms	
$(C_4H_9NH_3)_2PbBr_4$ nanobelt	365 nm	5 V	1.51×10^{-9} A	1.1×10^{-5} A/W	---	49 ms	53 ms	[25]
	405 nm			2.3×10^{-5} A/W	---	34 ms	50 ms	
$(C_4H_9NH_3)_2PbBr_4$ microplatelet	365 nm		5.21×10^{-9} A	1.7×10^{-6} A/W	---	31 ms	33 ms	

	405 nm			4.5×10^{-6} A/W	---	28 ms	55 ms	
PEA_2PbI_4· $(MAPbI_3)_{n-1}$ (n=1, 2, 3) single crystal	500 nm	5 V	1×10^{-11} A to 1×10^{-8}A	less than 200 mA/W	10^{13} Jones	---	---	[40]
2DH $(BA)_2(MA)_{n-1}Pb_nBr_{3n+1}$ ($1<n<\infty$) crystal	500 nm	1 V	2.8×10^{-12} A	190 mA/W	---	210 ms	240 ms	[16]
$(C_4H_9NH_3)_2(CH_3NH_3)_2Pb_3Br_{10}$ single crystal	420 nm	5 V	2×10^{-12} A	---	3.6×10^{10} Jones	150 μs	570 μs	[41]
$(OA)_2FA_{n-1}Pb_nBr_{3n+1}$ microplatelet	442 nm	9 V	---	32 A/W	---	0.25 ms	1.45 ms	[42]
$(C_4H_9NH_3)_2PbBr_4$ single crystal	470 nm	0.5 V	10^{-10} A	2100 A/W	---	---	---	[23]
$(PEA)_2PbBr_4$ Single Crystal	365 nm	10 V	2.5×10^{-13} A	31.48 mA/W	1.55×10^{13}	0.41 ms	0.37 ms	[43]
$(C_6H_5C_2H_4NH_3)_2PbI_4$ Single Crystal	solar light	5 V	<1 pA	5.4 mA/W	1.07×10^{13}	---	---	[44]
$(BA)_2(MA)_{n-1}Pb_nBr_{3n+1}$ n = 4 nanowire	530 nm	5 V	below 10^{-12} A	1.5×10^{4} A/W	7×10^{15} jones	27.6 μ s	24.5 μ s	[45]
BA_2PbI_4 Single Crystal	Xenon lamp	0.8 V	6×10^{-13} A	---	---	1.7 μs	3.9 μs	[46]