Supplementary Information

For

Epitaxy growth of $MAPbBr_xCl_{3-x}$ single-crystalline perovskite films toward spectral selective detection in both broadband and narrowband range

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Keywords: epitaxial growth, mixed-halide perovskite, photoelectric characteristics, tunable spectral response, dual-mode detection

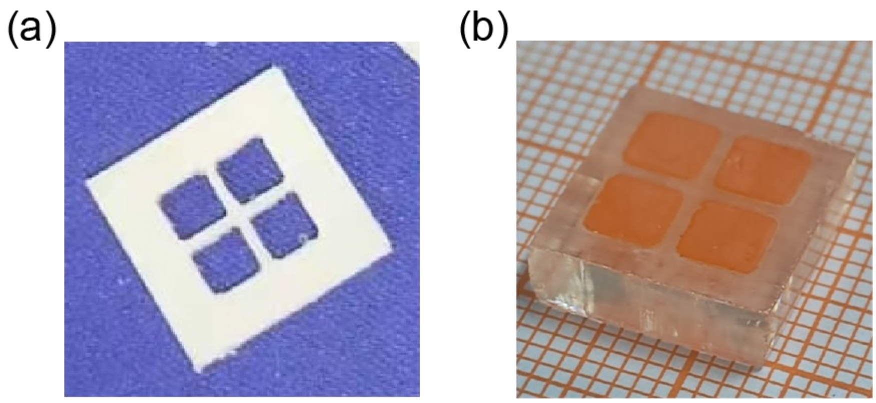

Figure S1 (a) Photograph of the used Teflon mould with a square of 15 × 15 mm^2 and thickness of 1 mm. (b) Photograph of the epitaxial 2×2 $MAPbBr_3$ single-crystalline arrays on $MAPbCl_3$ crystals with single pixel area of 4 × 4 mm^2.

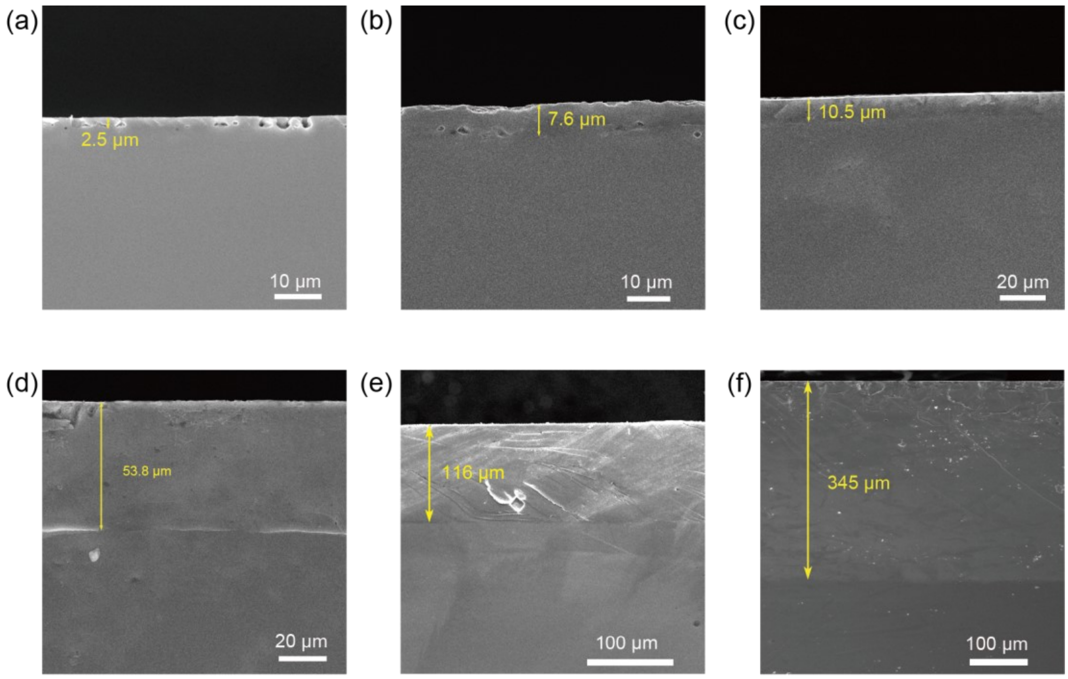

Figure S2 Cross-sectional SEM images of the formed heterojunction interfaces with the duration of $MAPbCl_3$ crystals dipping into hot $MAPbBr_3$ solution as (a) 20 seconds, (b) 50 seconds, (c) 2 minutes, (d) 10 minutes, (e) 1 hours to (f) 4 hours, respectively.

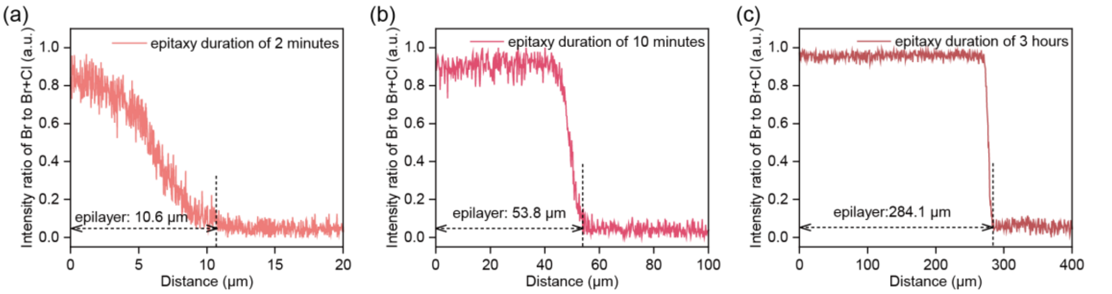

Figure S3 EDX line scanning results images of the Br/(Br+Cl) atom intensity ratio across the epitaxial interface of formed heterojunctions with the duration of $MAPbCl_3$ crystals dipping into hot $MAPbBr_3$ solution as (a) 2 minutes, (b) 10 minutes, and (c) 3 hours, respectively.

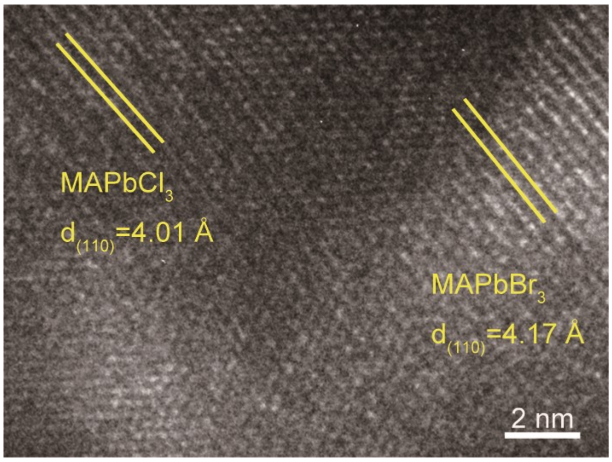

Figure S4 HRTEM image of the interface region of Bi-$MAPbCl_3$/$MAPbBr_3$ single-crystalline heterojunction.

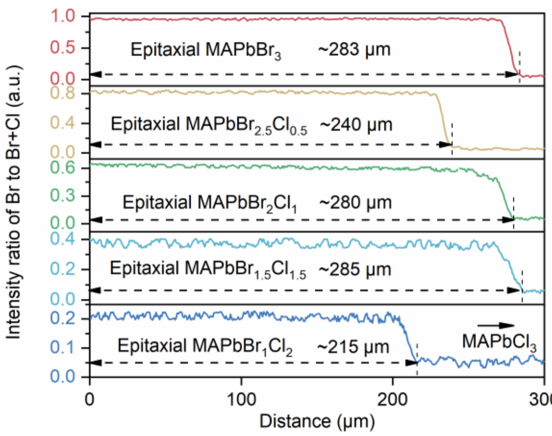

Figure S5 EDX line scanning results images of the Br/(Br+Cl) atom intensity ratio form epilayer $MAPbBr_xCl_{3-x}$ surface to the $MAPbCl_3$ substrate.

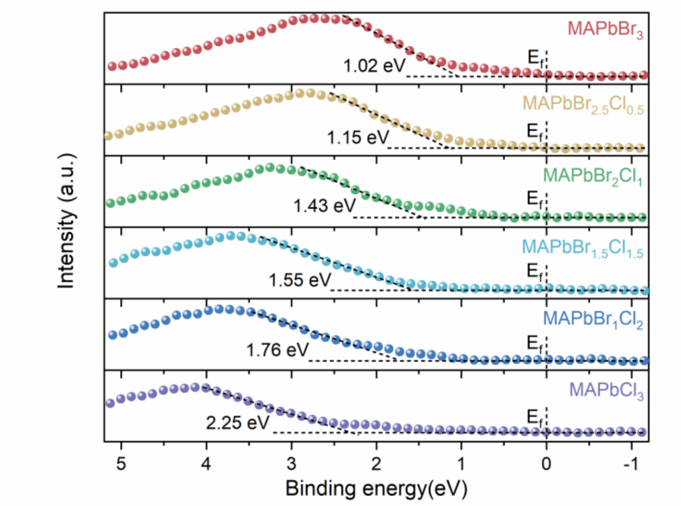

Figure S6 XPS valence spectra of fabricated single-crystalline $MAPbBr_xCl_{3-x}$ samples with x=0, 1, 1.5, 2, 2.5, 3, respectively.

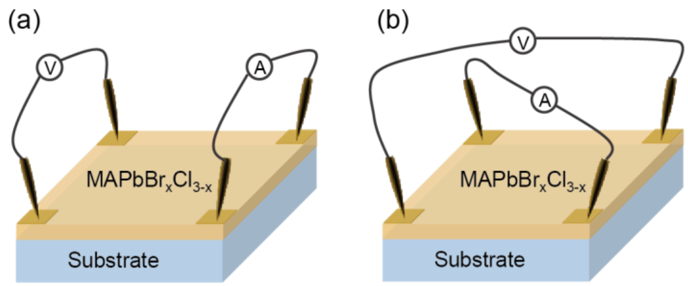

Figure S7 Schematic diagram of (a) sheet resistivity measurement and (b) hall effect measurement for the epitaxial MAPbBrxCl3-x single-crystalline layers by four contacts van der Pauw method.

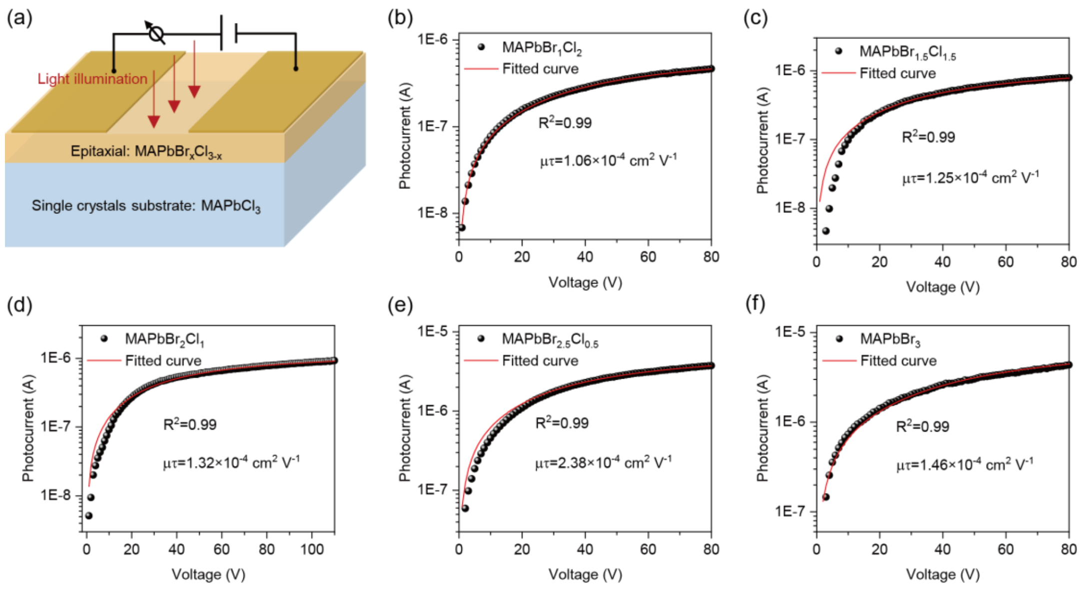

Figure S8 (a) Schematic diagram of the voltage-dependent photocurrent measurement for the epitaxial $MAPbBr_xCl_{3-x}$ single-crystalline planar-type devices. (b-f) The results of voltage-dependent photocurrent for different epitaxial $MAPbBr_xCl_{3-x}$ layers. The red solid line represents a mobility-lifetime fitting to the data according to the Hecht equation.

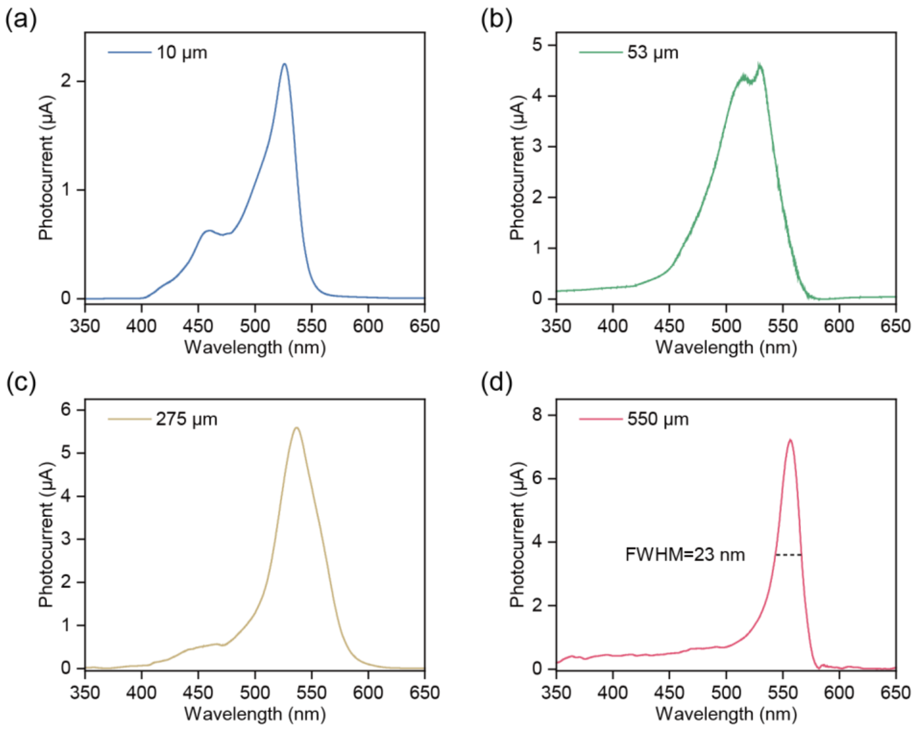

Figure S9 Photo response spectra of the epitaxial planar-type $MAPbBr_3$ photodetectors with different thickness upon bottom illumination at an external electric field of 30 V mm^{-1}.

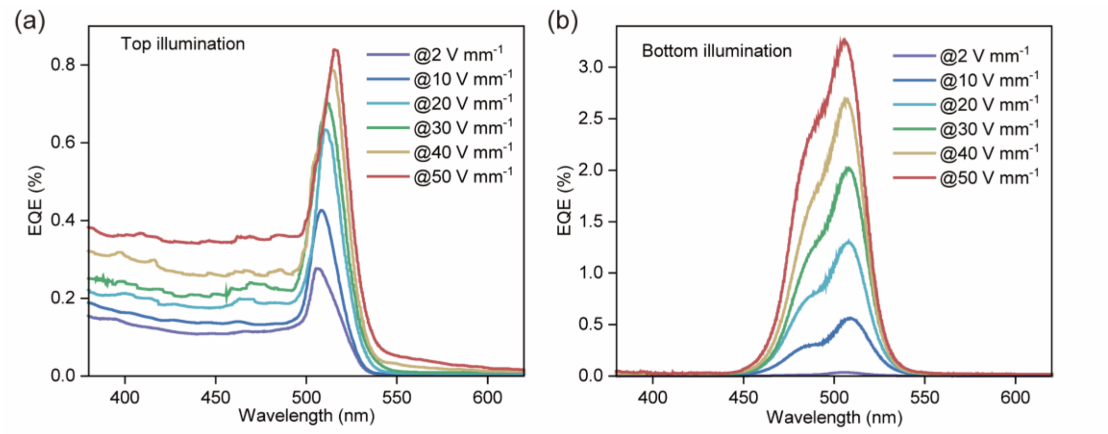

Figure S10 EQE spectra of the planar-type $MAPbBr_{2.5}Cl_{0.5}$ photodetector under various external electric field upon (a) top illumination and (b) bottom illumination.

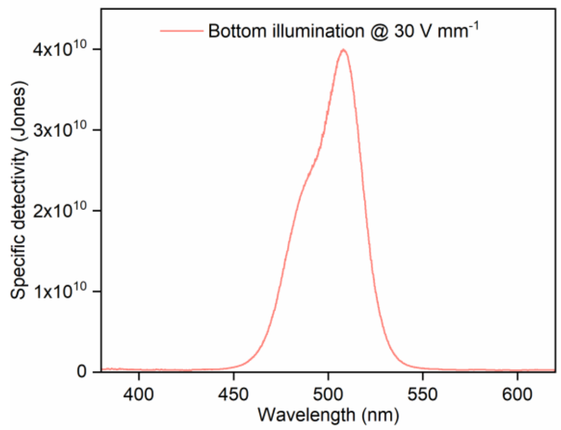

Figure S11 Specific detectivity (D*) of the planar-type $MAPbBr_{2.5}Cl_{0.5}$ photodetector under 30 V mm^{-1} upon bottom illumination.

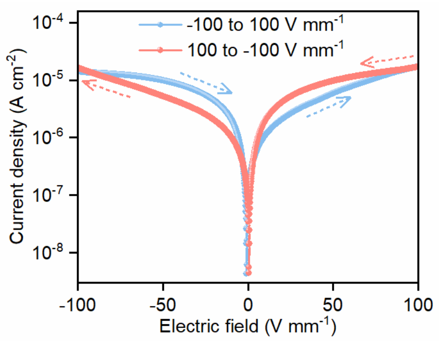

Figure S12 Current density-Electric field hysteresis loop for the epitaxial $MAPbBr_{2.5}Cl_{0.5}$ planar-type device in darkness.

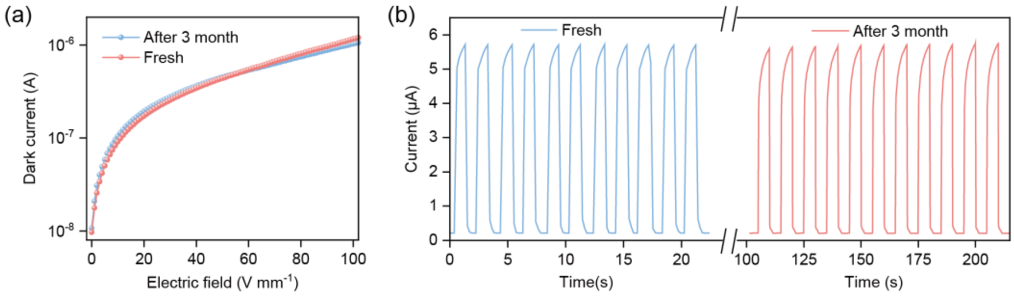

Figure S13 Long-time stability of the epitaxial $MAPbBr_{2.5}Cl_{0.5}$ photodetector: (a) I-V stability in darkness and (b) photo response stability after 3 month in air condition without any encapsulation.

Table S1 The key optoelectronic parameters of $MAPbBr_xCl_{3-x}$ perovskite crystals and films reported in literatures.

Material	**Morphology**	**Mobility ($cm^2 V^{-1} s^{-1}$)**	**Carrier lifetime (ns)**	**Diffusion length (μm)**	**Ref.**
$MAPbBr_xCl_{3-x}$	**single-crystalline film**	**110~213**	**620~1360**	**16.6~24.8**	**This work**
$MAPbCl_3$	single-crystal	42 ± 9	662	3.0–8.5	1
$MAPbCl_3$	single-crystal	26	31	NA	2
$MAPbCl_3$	Polycrystalline film	NA	6	NA	2
$MAPbBr_3$	single-crystal	81 ± 5	899 ± 127	5.4–14.2	3
$MAPbBr_3$	single-crystalline film	60	10–60	1.5–3	4
$MAPbBr_{2.94}Cl_{0.06}$	single-crystal	560	5000	NA	5
$MAPbBr_3$	single-crystalline film	60	189±10	5	6
$MAPbBr_3$	single-crystalline film	261.94	60.26	NA	7
$MAPbBr_3$	single-crystalline film	40.7(e); 27.3(h)	390	6.4(e); 5.2(h)	8
$MAPbBr_3$	Polycrystalline film	0.26	168	0.33	6
$MAPbBr_3$	Polycrystalline film	8.6(e); 9.0(h)	51	1.058 ± 0.048	9

Reference

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