Supplementary information

Carrier Recombination Dynamics in $[MAPbCl_3]_x[CsPbBr_3]_{1-x}$ Shell Passivated $CsPbBr_3$ Single Crystals

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S1. Revealing the $MAPbCl_3$ and $CsPbBr_3$ content in $[MAPbCl_3]_x[CsPbBr_3]_{1-x}$ shell by the correlation between PL peak energy of $MAPbCl_3$ and $CsPbBr_3$ single crystal

Many studies have shown that the bandgap of mixed perovskite is approximately linear with the composition of the corresponding components.[1-4] Assuming the bandgap of mixed crystal $[MAPbCl_3]_x[CsPbBr_3]_{1-x}$ follow a linear correlation between the bandgap and x composition, which was determined by the bandgaps (PL peak energy) of pure $MAPbCl_3$ and $CsPbBr_3$ single crystals. Then, the composition x for $[MAPbCl_3]_x[CsPbBr_3]_{1-x}$ shell can be determined as x = 0.34 for $CsPbBr_3$ grown without CB, see Figure S1.

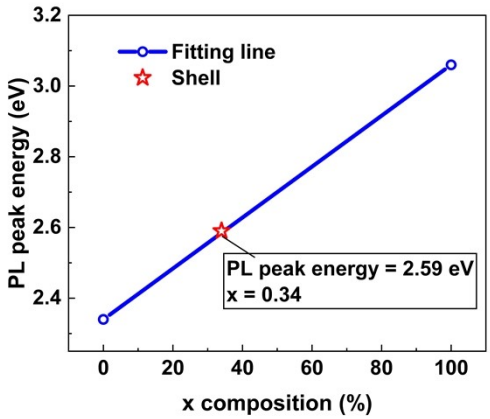

Fig. S1 The correlation between the content of $MAPbCl_3$ (x) and PL emission energy in mixed $[MAPbCl_3]_x[CsPbBr_3]_{1-x}$ single crystal.

S2. TRPL kinetics of $CsPbBr_3$ single crystals grown with and without choline bromide (CB)

Table S1. Fitting parameters for TRPL kinetics of $CsPbBr_3$ single crystals with and without CB additives. TRPL measurements were conducted upon 485 nm photoexcitation with an excitation fluence of 4.9×10^8 pulse$^{-1}\cdot$cm^{-2}.

Sample	**Decay**				
	A_1	τ_1 **[ns]**	A_2	τ_2 **[ns]**	τ_A **[ns]**
With CB	0.70	2.3	0.30	31.5	11.1
Without CB	0.70	4.0	0.30	37.8	14.1

S3. TRPL and TRMC kinetics of $CsPbBr_3$ (with CB) single crystals covered with and without shell

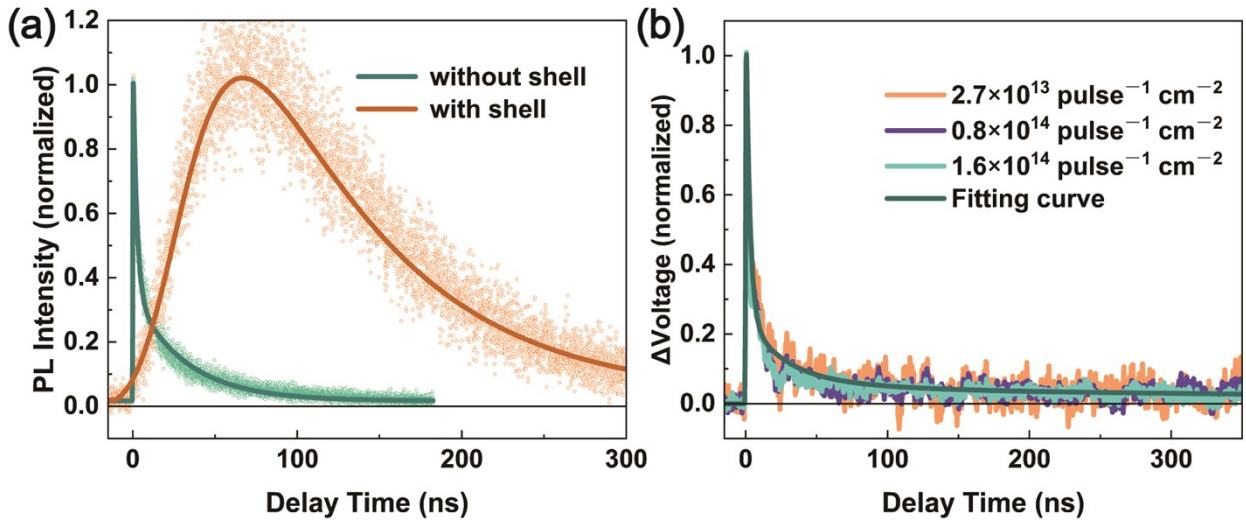

Fig. S2 (a) Comparison of TRPL kinetics between bare $CsPbBr_3$ (with CB) single crystals (SCs) and $CsPbBr_3$ (with CB) SCs covered with $[MAPbCl_3]_x[CsPbBr_3]_{1-x}$ shell. TRPL measurements were conducted upon 485 nm photoexcitation with an excitation fluence of 4.9×10^8 $pulse^{-1} \cdot cm^{-2}$. (b) TRMC kinetics of $CsPbBr_3$ (with CB) SCs covered with shell after 400 nm photoexcitation under the indicated excitation fluences. The solid lines are fitting curves based on multi-exponential functions, the detailed fitting parameters are shown in Table S2.

S4. TRPL and TRMC kinetics of $CsPbBr_3$ (with and without CB) single crystals covered with $[MAPbCl_3]_x[CsPbBr_3]_{1-x}$ shells

Table S2. Fitting parameters for TRPL and TRMC kinetics of $CsPbBr_3$ (with and without CB) covered by $[MAPbCl_3]_x[CsPbBr_3]_{1-x}$ shells.

Sample	TRPL				TRMC								k_{trap} [$\times 10^6$ s^{-1}]	k_{trap} [$\times 10^6$ s^{-1}]
	A_1	τ_t [ns]	A_2	τ_d [ns]	A_1	τ_1 [ns]	A_2	τ_2 [ns]	A_3	τ_3 [ns]	A_4	τ_4 [ns]		
Without CB	−1	21	1	73.2	0.65	IRF	0.17	21	0.11	73	0.07	1334	0.7	13.6
With CB	−1	25	1	82.9	0.75	IRF	0.18	25	0.04	82	0.03	899	1.1	12.1

IRF: Instrumental response function; τ_t and τ_d are the rise lifetime and decay lifetime for TRPL kinetics, representing charge transfer from shell to core and carrier recombination processes in the core, respectively.

After photoexcitation at 485 nm, carriers may be generated in shell and core. For photogenerated carriers in the shell, both electrons and hole can be trapped by surface traps of shell or transferred from shell to core. Considering the proportional correlation between photoconductivity and carrier mobility, as well as the higher carrier mobility of $MAPbCl_3$ (179 $cm^2 \cdot V^{-1} \cdot s^{-1}$) than $CsPbBr_3$ (52 $cm^2 \cdot V^{-1} \cdot s^{-1}$), the transfer of carriers from shell to core may induce the decay of photoconductivity. Therefore, both surface trapping of charges in the shell or charge transfer from shell to core may induce the decay of photoconductivity. For carriers in the core, hole trapping and electron trapping would also induce the decay of photoconductivity. Therefore, TRMC kinetics of core shell samples were fitted with four exponential decay functions, representing electron and hole photoconductivity decays in the shell and in the core respectively.

References

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