Supplementary Information for

## A multiple-coordination framework for CsPbI2Br perovskite solar cells

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Fig. S1. a) The SEM images and b) corresponding energy disperse spectrum image of





Fig. S2. The AFM images of a) ETL0 and b) ETL3.



Fig. S3. a) The schematic diagram and b) the cross-section image of CsPbI<sub>2</sub>Br PSCs.



Fig. S4. The J-V characteristics of CsPbI<sub>2</sub>Br PSCs based on different ETLs.



**Fig. S5**. The representative structure models: (a) unpassivated Pb custer, (b) passivated Pb custer, (c) unpassivated Pb-I antisite defect, (d) passivated Pb-I antisite defect.



Fig. S6. The mirror-liked CsPbI<sub>2</sub>Br film.



Fig. S7. The current distribution of  $CsPbI_2Br$  film.



Fig. S8. Steady-state photoluminescence (PL) spectrum of  $CsPbI_2Br$  films deposited on ITO/ETL substrate, the excitation light incidents from the ITO side.



Fig. S9. Normalized TRPL decay profiles.



Fig. S10. The TRPL decay curves derived from FLIM.



Fig. S11. SCLC measurements of hole-only devices.



Fig. S12. EQE spectra of ETL3/CsPbI<sub>2</sub>Br-ChCl PSCs.

PSCs	A <sub>1</sub>	$\tau_1$ (ns)	A <sub>2</sub>	$\tau_{2}$ (ns)	$\tau_{ave}$ (ns)
ETL0	0.142	2.974	0.863	7.513	7.236
ETL3	0.263	1.423	0.749	3.898	3.617
ETL3-ChCl	0.192	0.758	0.816	2.322	2.210

 Table S1. Fitting results of the TRPL spectra.