A Surface Modifier Enhances Performance in All-Inorganic $CsPbI_2Br$ Perovskite Solar Cells with Efficiencies Approaching 15%

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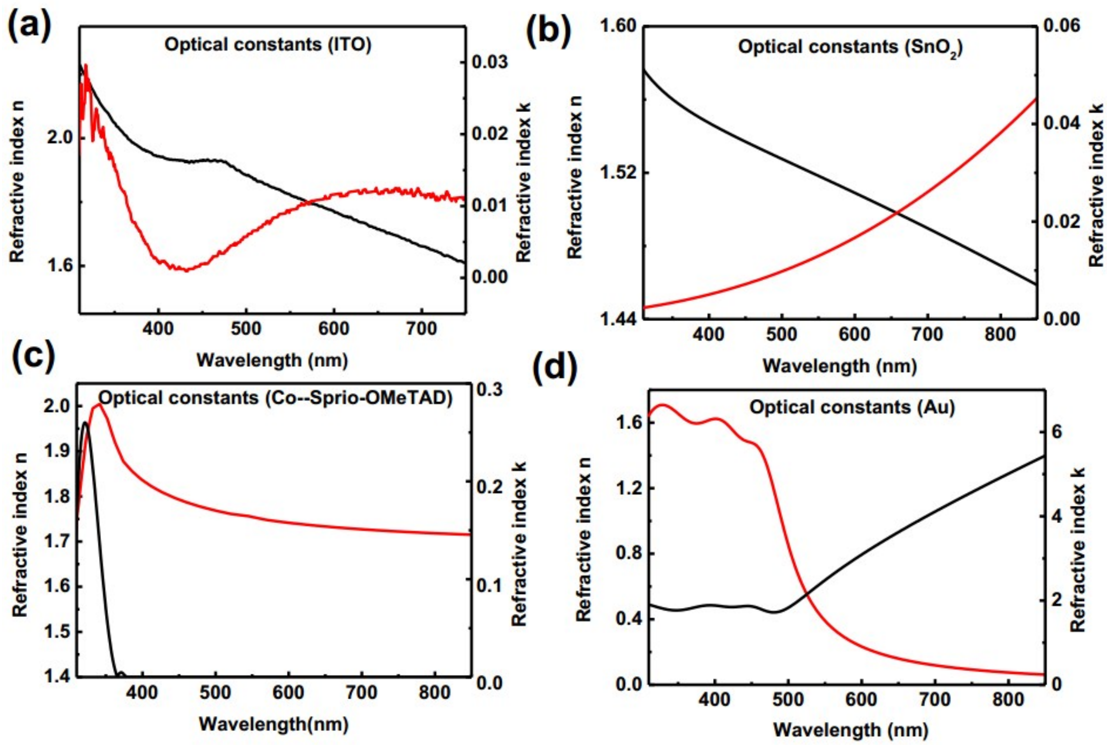
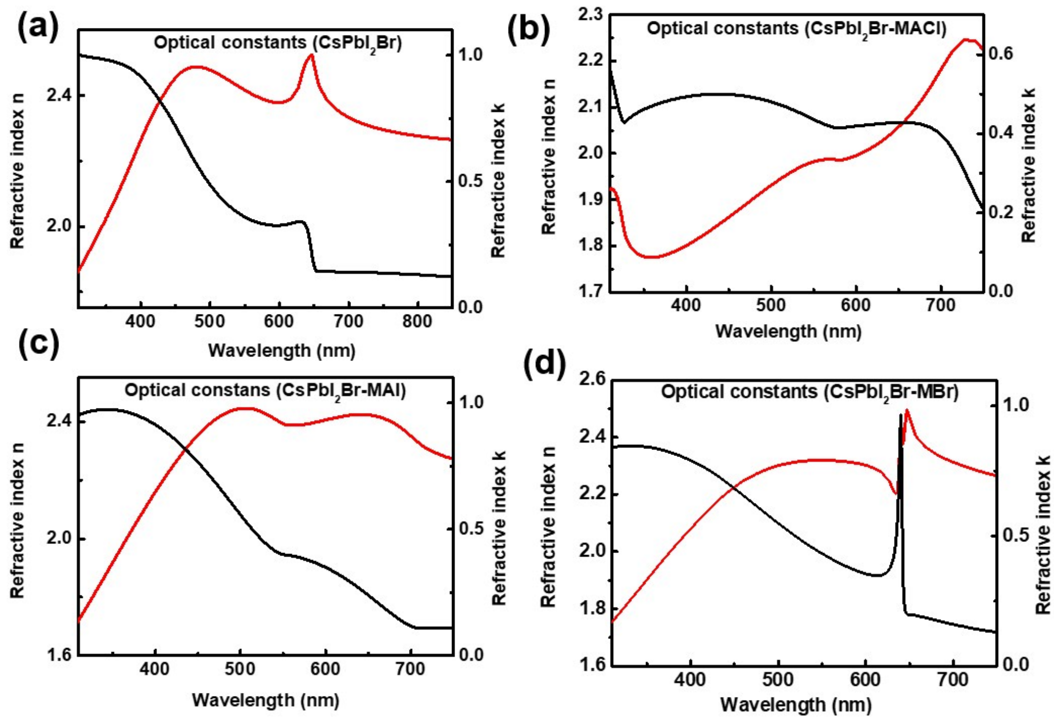


Function layer	Material	Thickness d (nm)
Electrode	ITO	75nm
ELFs	SnO_2	28 nm
Electrode	Au	60 nm
HLFs	Spiro-OMETAD	30 nm

Fig. S1. Optical constants used for the simulation of perovskite solar cells (PSC). (a) ITO, (b) SnO_2, and (c) Spiro-OMETAD (Co), and (d) Au used as input for the optical analysis. Also shown in the associated table are the thicknesses of different functional layers determined by profilometry.



	Material	Thickness d (nm)
	$CsPbI_2Br$	439 nm
$CsPbI_2Br$	$CsPbI_2Br$-MABr	441 nm
-MAX (Cl, I, Br)	$CsPbI_2Br$-MACl	449 nm
	$CsPbI_2Br$-MAI	441 nm

Fig. S2. Angle-varied spectroscopic ellipsometry measurements used for the simulation of perovskite solar cells. Optical constants of $CsPbI_2Br$ PSCs with different passivation layers used as input parameters for the optical analysis and full simulation of solar cells. (a) $CsPbI_2Br$, (b) $CsPbI_2Br$-MABr, (c) $CsPbI_2Br$-MACl, and (d) $CsPbI_2Br$-MAI. In the associated table, thicknesses of active layer and MAX measured from a step profiler are summarized.

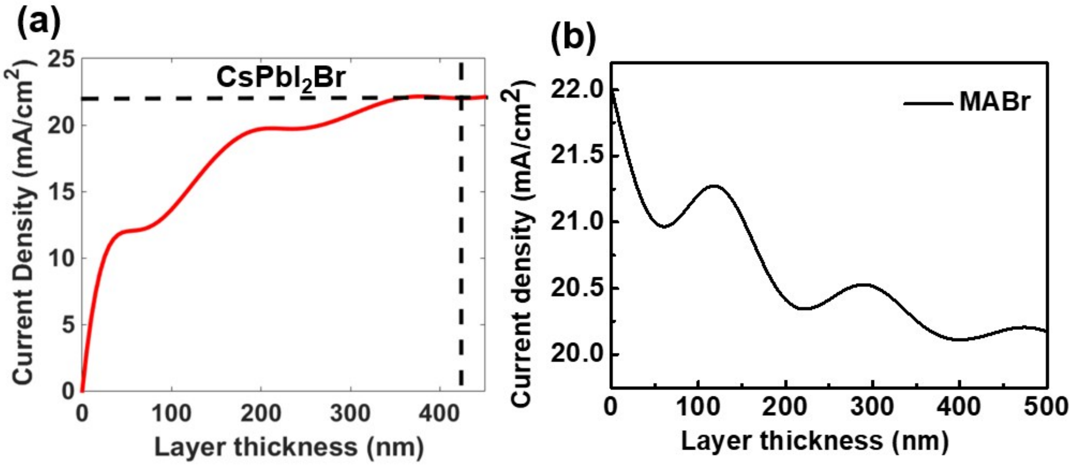


Fig. S3. Calculated photocurrent in different positions inside the $CsPbI_2Br$ perovskite solar cells (a) without and (b) with MABr passivation layer (Current density obtained by from 100% IQE).

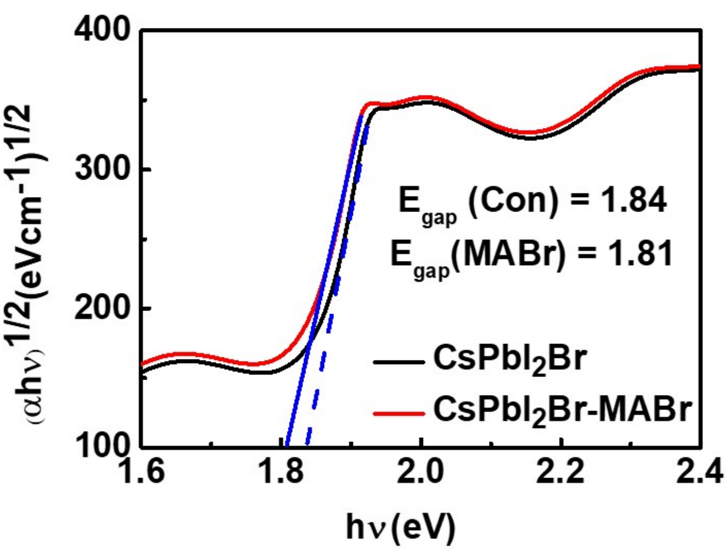


Fig. S4. UV-Vis absorption spectra of the $CsPbI_2Br$ and $CsPbI_2Br$-MABr film (Tauc method). $hv = 1240/\lambda$**, absorption coefficient spectrum:** $\alpha = \frac{1}{d} \times In(1/T)$**, d is thickness of active layer. The picture of Tauc,** $(\alpha hv)^{1/2} \sim hv$**, the unit of** α **is** cm^{-1} **and the unit of** hv **is eV.**

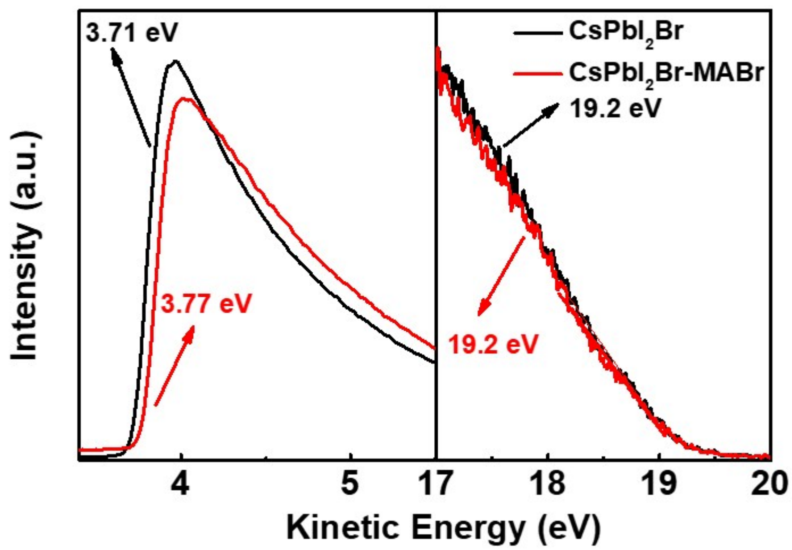


Fig. S5. UPS spectra measured on $CsPbI_2Br$ and $CsPbI_2Br$-MABr films. Work function ϕ: $h\nu - \phi = E_{feimi} - E_{cutoff}$, hν = 21.2eV and the value of E_{feimi} from the Au.

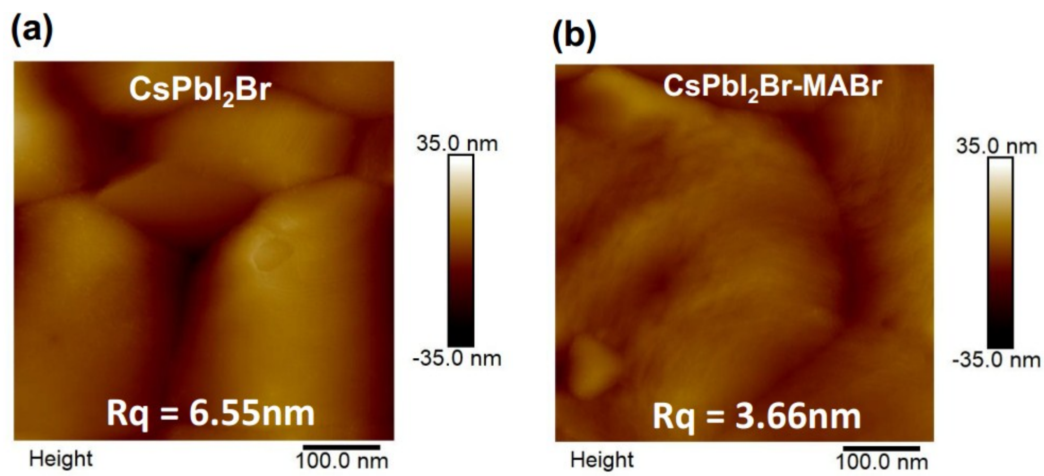


Fig. S6. Topographic images of (a) $CsPbI_2Br$ and (b) $CsPbI_2Br$-MABr films measured at the grain boundaries with atomic force microscopy.

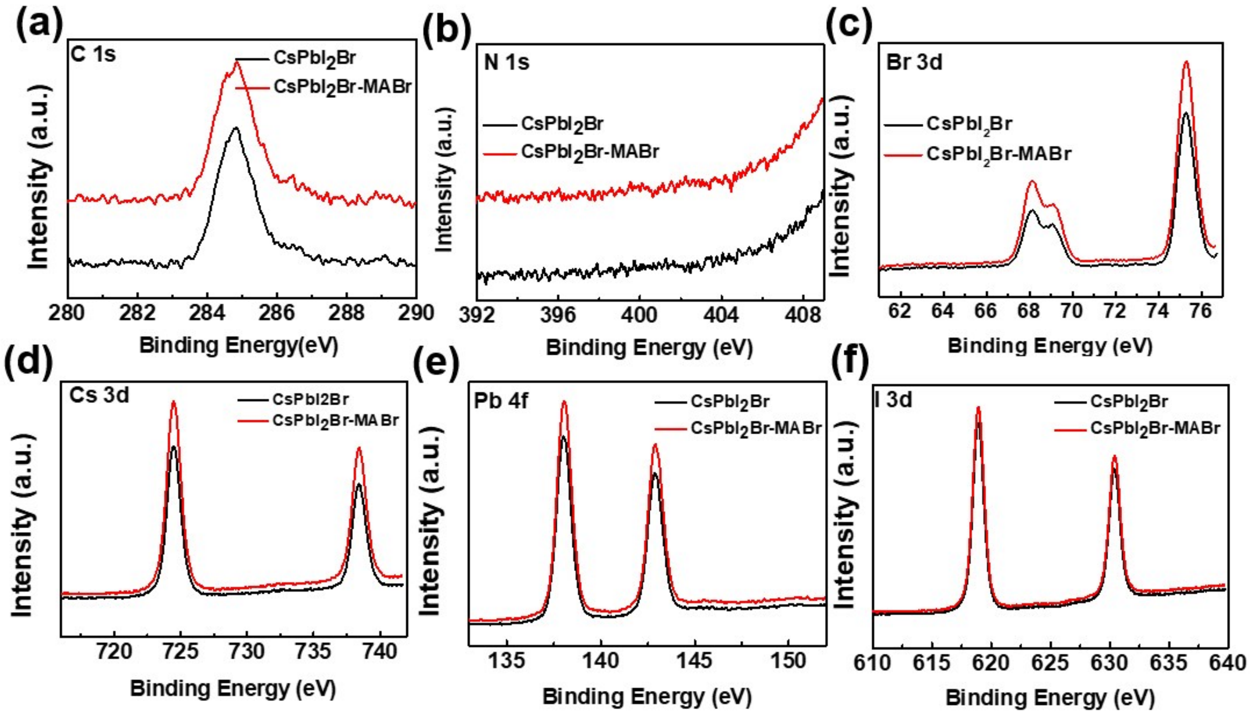


Figure. S7. High resolution XPS of $CsPbI_2Br$ and $CsPbI_2Br$-MABr probed on core-level (a) C 1s, (b) N 1s, (c) Br 3d, (d) Cs 3d, (e) Pb 4f and (f) I 3d.

Table. S1. The statistical analysis of element contents on the surface of $CsPbI_2Br$-MABr thin films.

	Element	Br 3d	Pb 4f	C 1s	I 3d	Cs 3d
$CsPbI_2Br$	wt.%	13.6	13.3	22.2	29.5	17.6
$CsPbI_2Br$-MABr	wt.%	16.3	13.0	21.2	27.0	19.0

Table. S2. Device parameters along statistics of $CsPbI_2Br$ solar cells prepared by passivation methods measured in the reverse scan directions under AM 1.5 G solar illumination.

Processing	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
$CsPbI_2Br$	1.20 (± 0.2)	14.1 (± 0.3)	68.4 (± 2)	12.8 (± 1)
$CsPbI_2Br$-MABr	1.23 (± 0.1)	14.9 (± 0.2)	79.2 (± 3)	14.0 (± 0.8)

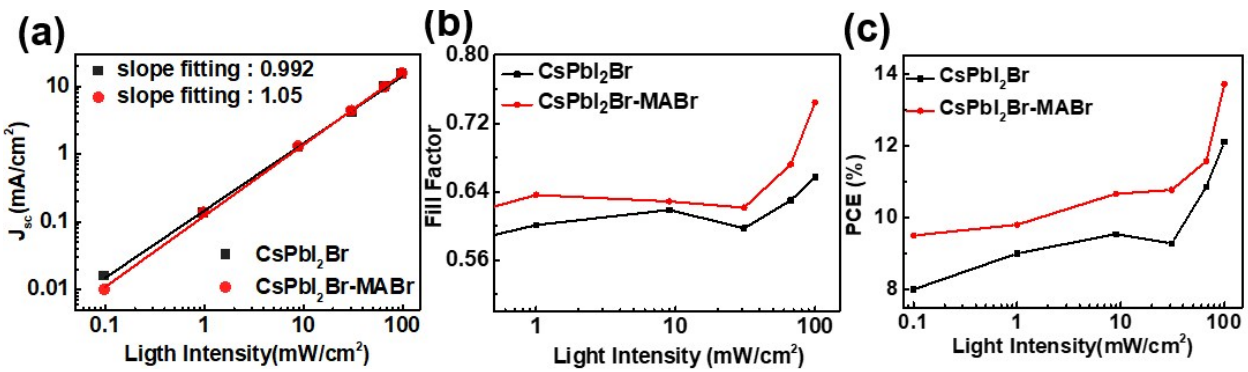


Fig. S8. Light intensity-dependent (a) Short-circuit current, (b) Fill factor, (c) Power conversion efficiency of $CsPbI_2Br$ and $CsPbI_2Br$-MABr solar cells.

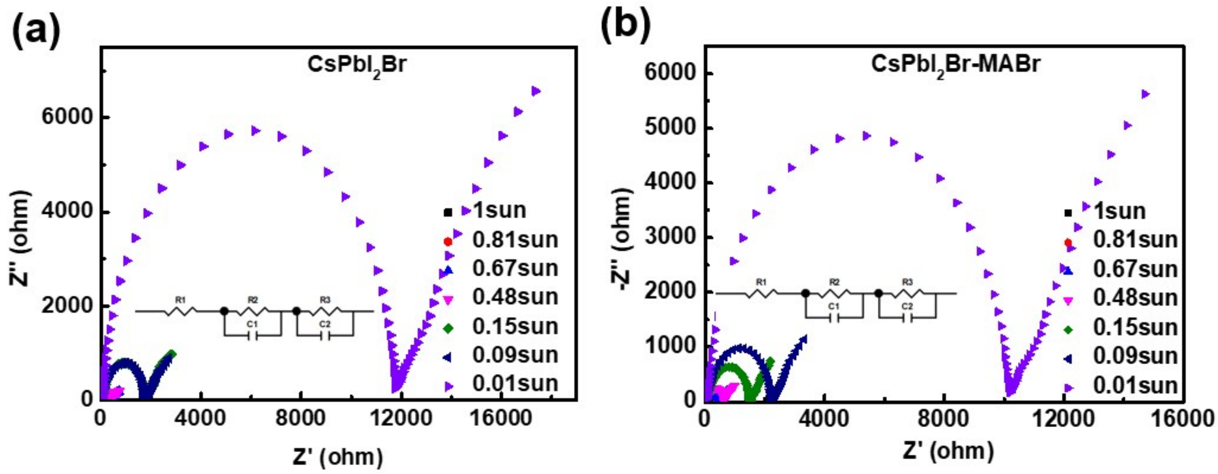


Fig. S9. Impedance spectra of $CsPbI_2Br$ solar cells with and without MABr passivation layers measured at different light intensities (P_{light}).

Table. S3. Device parameters of impedance spectroscopy of $CsPbI_2Br$ and $CsPbI_2Br$-MABr solar cells measured under 1 sun at V_{oc} together.

	R_1 (Ω)	R_2 (Ω)	R_3 (Ω)	C_1 (F)	C_2 (F)
$CsPbI_2Br$	48.6 (± 4)	207 (± 1)	19.3 (± 6)	7.63 (± 0.3) x10^{-9}	0.0023 (± 0.001)
$CsPbI_2Br$-MABr	64.9 (± 3)	194 (± 6)	251 (± 1)	5.16 (± 0.3) x10^{-9}	0.0030 (± 0.001)

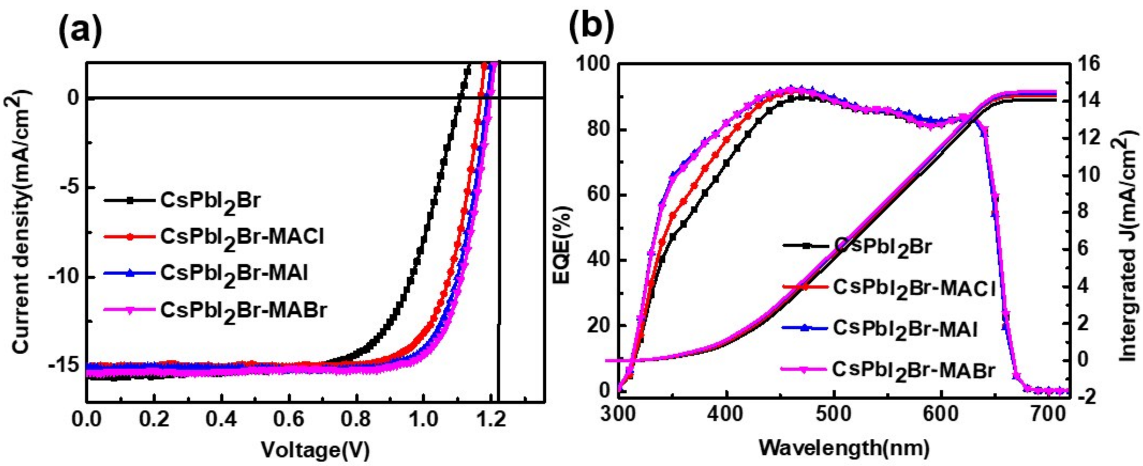


Fig. S10. (a) J-V characteristics of best $CsPbI_2Br$ and $CsPbI_2Br$-MAX (X = Cl, Br, and I) solar under simulated AM 1.5G illumination (100 mW/cm^2), (b) EQE spectra of according devices.

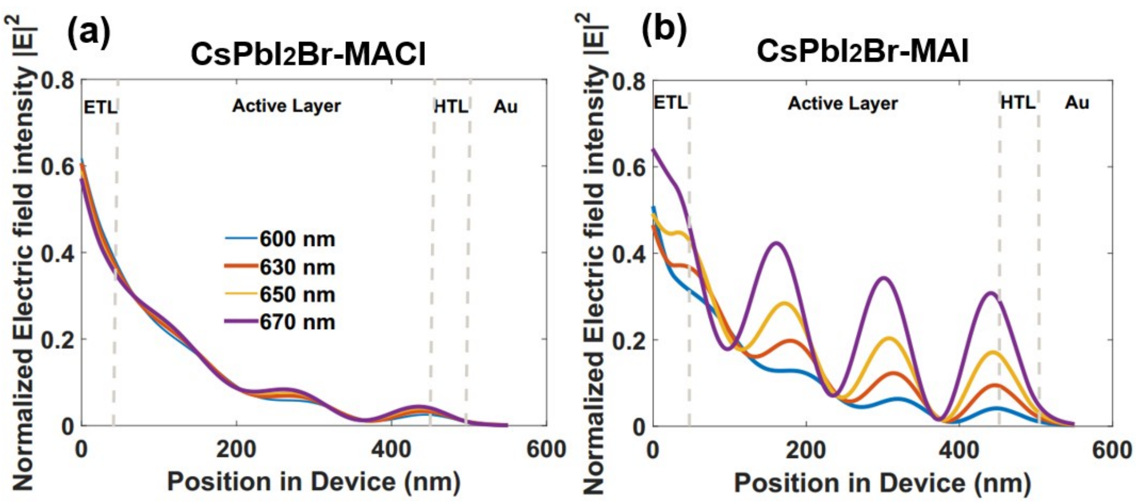


Fig. S11 (a-b) Simulated electric field intensities induced by propagation of various wavelengths of light through (a) $CsPbI_2Br$-MACl, (b) $CsPbI_2Br$-MAI.