

## Supporting information

### Revealing the Unfavorable Role of Superfluous $\text{CH}_3\text{NH}_3\text{PbI}_3$ Grain

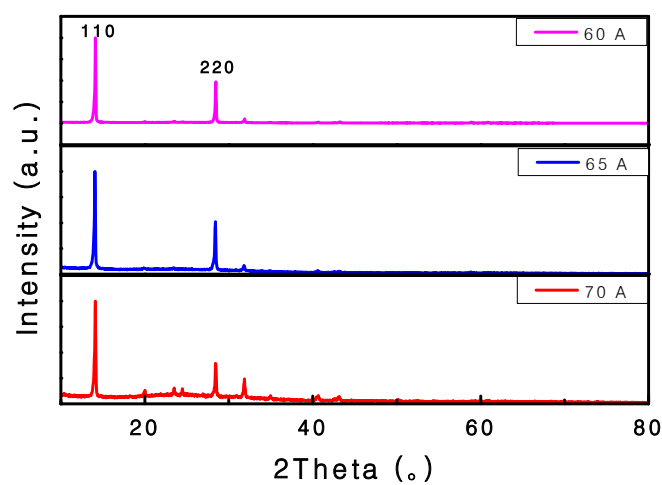
#### Boundary Traps in Perovskite Solar Cells on Carriers Collection

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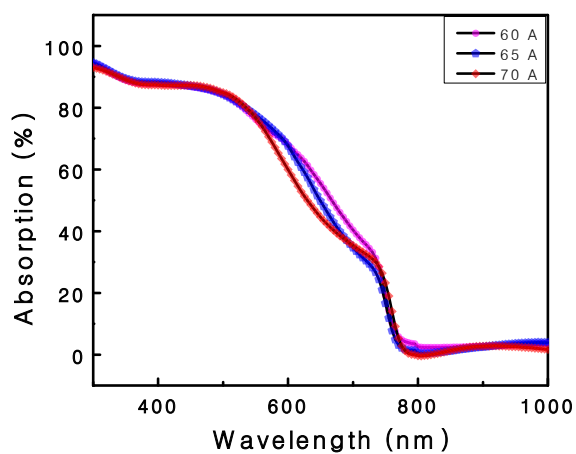
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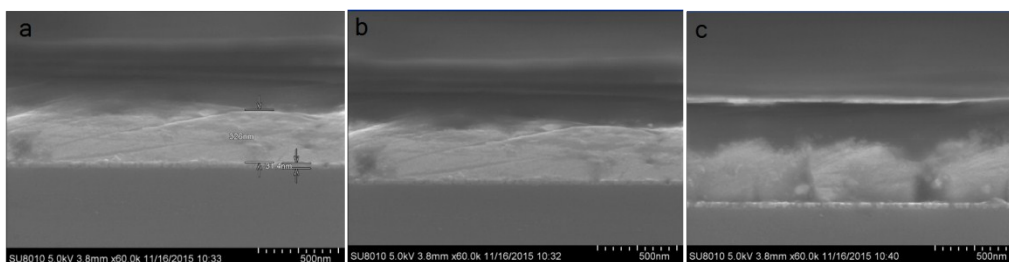
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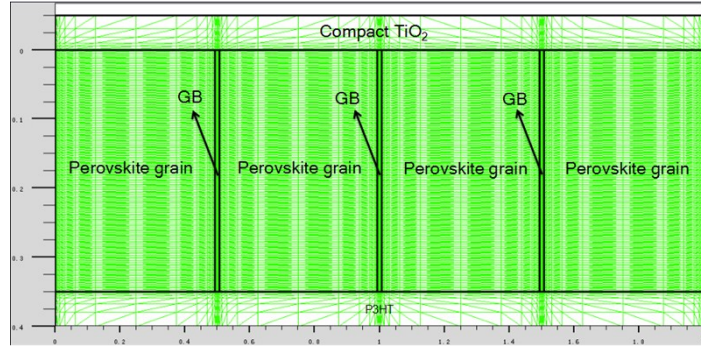
**Fig. S1**  $\text{CH}_3\text{NH}_3\text{PbI}_3$  films XRD evolution pattern as the function of evaporation current.



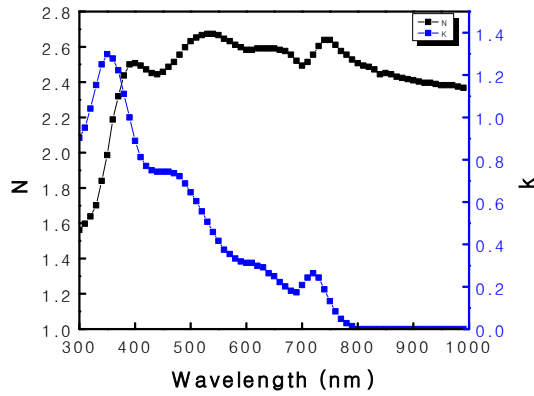
**Fig. S2**  $\text{CH}_3\text{NH}_3\text{PbI}_3$  absorption spectrum prepared by STELV with different evaporation current.



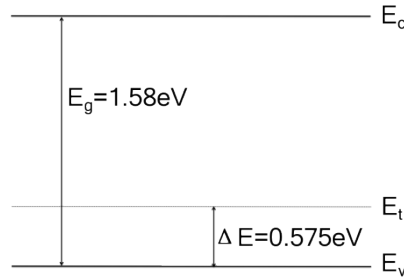
**Fig. S3** Cross section SEM image of  $\text{CH}_3\text{NH}_3\text{PbI}_3$  films prepared by the STELV procedure with different value of evaporation current keeping identical thickness, which can be obtained with the same amount of  $\text{PbI}_2$  power. (a) 70 A; (b) 65 A; (c) 60 A.



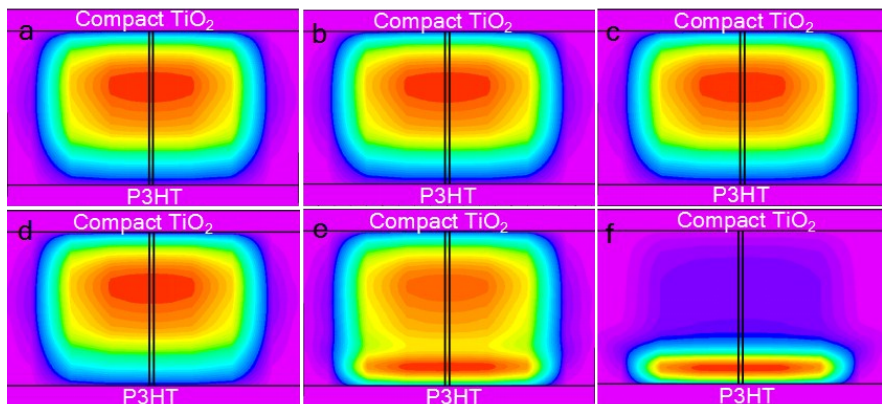
**Fig. S4** The mesh diagram for cross section PSCs covered the physical simulation domain.



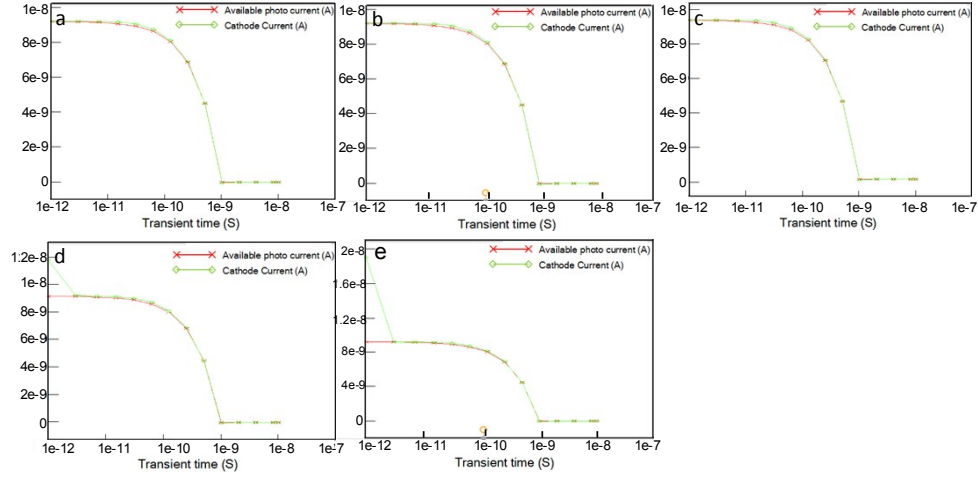
**Fig. S5** Refractive index (n) and extinction coefficient (k) spectra of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> film for simulation.



**Fig. S6** The specification of PGB trap energy level diagram.



**Fig. S7** Recombination rate distribution within PSCs in light illumination versus different bulk defects density. (a) without any defects; (b)  $1e11 \text{ cm}^{-3}$ ; (c)  $1e12 \text{ cm}^{-3}$ ; (d)  $1e13 \text{ cm}^{-3}$ ; (e)  $1e14 \text{ cm}^{-3}$ ; (f)  $1e15 \text{ cm}^{-3}$ .



**Fig. S8** Transient photo current collection efficiency versus the amount of bulk traps density. (a)  $1e11 \text{ cm}^{-3}$ ; (b)  $1e12 \text{ cm}^{-3}$ ; (c)  $1e13 \text{ cm}^{-3}$ ; (d)  $1e14 \text{ cm}^{-3}$ ; (e)  $1e15 \text{ cm}^{-3}$ ;

**Table S1.** Parameter set for the simulation of PSCs

Parameter and units	TiO <sub>2</sub>	Perovskite	P3HT
Thickness (nm)	50	350	50
Band gap (eV)	3.2	1.58	3.02
Electron affinity (eV)	4.0	3.75	2.18
Permittivity (F/cm)	46	70	3.4
Effective conduction band density (cm <sup>-3</sup> )	-	$1e21$	$2.5e19$
Effective valence band density (cm <sup>-3</sup> )	-	$1e21$	$2.5e19$
Electron mobility (cm <sup>2</sup> v <sup>-1</sup> s <sup>-1</sup> )	1	20	-
Hole mobility (cm <sup>2</sup> v <sup>-1</sup> s <sup>-1</sup> )	0.13	20	0.001
Electron Lifetime (mS)	20	100	-
Hole Lifetime (mS)	2.3	100	-
Doping concentration of acceptors (cm <sup>-3</sup> )	-	-	$2e17$
Doping concentration of donors (cm <sup>-3</sup> )	$2e16$	-	-
Band tail density of states (cm <sup>-3</sup> eV <sup>-1</sup> )	-	$1e11$ to $1e15$	-
Characteristic energy donors, acceptors	-	1,1	-
Capture cross section for donor states, e,h (cm <sup>2</sup> )	-	$1e-8$	-
Capture cross section for acceptors, e,h (cm <sup>2</sup> )	-	$1e-8$	-
Total Gaussian density of states N <sub>DB</sub> (cm <sup>-3</sup> )	-	$1e14$	-
Gaussian peak energy donors, acceptors	-	0.575	-
Characteristic decay energy of donors, acceptors	-	0.1, 1	-
Physical models	Phonon transition (SRH); Photon transition (OPTR)		