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Supporting Information

Hybrid Perovskites: Prospects for Concentrator Solar Cells

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1. Experimental details

Preparation of perovskite films. $MAPbI_3$ solution: 1 M perovskite precursor solution was prepared using a 4 : 1 (v : v) mixed solvent from anhydrous dimethylformamide (DMF) and dimethyl sulfoxide (DMSO) from Sigma-Aldrich with desired composition using precursor salts: methylammonium iodide (MAI; Dyesol), lead iodide (PbI_2; TCI), lead bromide ($PbBr_2$; TCI). $FA_{0.83}Cs_{0.17}PbI_{2.7}Br_{0.3}$ solution: 0.6 M perovskite precursor solution was prepared using a 4 : 1 (v : v) mixed solvent from anhydrous DMF and DMSO (Sigma-Aldrich) with desired composition using precursor salts: formamidinium iodide (FAI; Dyesol), cesium iodide (CsI; Alfa Aesar), lead iodide (PbI_2; TCI), lead bromide ($PbBr_2$; TCI). $FA_{0.7}MA_{0.25}Cs_{0.05}PbI_{2.7}Br_{0.3}$ solution: 0.6 M perovskite precursor solution was prepared using a 4 : 1 (v : v) mixed solvent from anhydrous DMF and DMSO (Sigma-Aldrich) with desired composition using precursor salts: formamidinium iodide (FAI; Dyesol), methylammonium iodide (MAI; Dyesol), cesium iodide (CsI; Alfa Aesar), lead iodide (PbI_2; TCI), lead bromide ($PbBr_2$; TCI). All the solutions were made in a nitrogen-filled glovebox and kept stirring overnight at room temperature. The $MAPbI_3$ films were deposited through a two-step spin coating program (10 s at 1000 rpm and 15 s at 6000 rpm) with dripping of anisole (Sigma-Aldrich) as anti-solvent during the second step, 10 s before the end. The films were then transferred to a hotplate immediately afterwards and annealed at 100 °C for 10 min. For films containing the FA-Cs and FA-MA-Cs mixtures, the precursor solutions were spin-coated through a two-step spin coating program (10 s at 1000 rpm and 35 s at 6000 rpm) with dripping of anisole (Sigma-Aldrich) as anti-solvent during the second step, 10 s before the end. The films were then annealed at 100 °C for 60 min. All the films were spin-coated in a drybox with relative humidity below 20%.

Preparation of quencher layers on perovskite films. [6,6]-phenyl C61 butyric acid methyl ester (PC61BM, Solenne) was dissolved in Chloroform at 30 mg mL^{-1} and spin-coated at

1000 rpm for 30 s. Poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS, Heraeus) was spin-coated at 2000 rpm for 60s, and N^2,N^2,$N^{2'}$,$N^{2'}$,N^7,N^7,$N^{7'}$,$N^{7'}$-octakis(4-methoxyphenyl)-9,9'-spirobi[9H-fluorene]-2,2',7,7'-tetramine (Spiro-MeOTAD) solution was spin-coated at 2500 rpm for 40s. To obtain a spiro-OMeTAD solution, we dissolved 85.7mg spiro-OMeTAD (Borun Technology) in 1ml anhydrous chlorobenzene with additives of 28.8 μl tert-butylpyridine (tBP) and 20 μl lithium bis(trifluoromethylsulfonyl) imide (Li-TFSI) salt in acetonitrile (520 mg ml^{-1}).

Characterization of PL lifetime. PL spectra were acquired using a time-resolved single photon-counting set-up (FluoTime 300, PicoQuant). Samples were photo-excited using a 507 nm laser head (LDH-P-C-510, PicoQuant) with pulse duration of 117 ps, fluence of ~30 nJ cm^{-2} per pulse and a repetition rate of 1 MHz. The PL was collected using a monochromator and hybrid photomultiplier detector assembly (PMA Hybrid 40, PicoQuant GmbH).

2. Supporting tables and figures

Table S1. Comparison of the record efficiencies of standard (one-sun) and concentrator solar cells.

Classification	One-sun solar cell	Concentrator solar cell	
	Efficiency [%]	Efficiency [%]	Solar Concentration [Suns]
Silicon	25.63[a]	27.6[b]	92
GaAs	28.8 [b]	29.1[b]	117
CIGS	22.6[a]	23.3[b]	15
Multi-junction cells	38.8 [c]	46.2[b]	508

a data taken from Ref. S1; **b** data taken from Ref. S2; **c** data taken from Ref. S3.

Table S2. Typical recombination rate parameters for hybrid organic-inorganic metal halide perovskites.

Perovskite formula	Monomolecular recombination rate [s^{-1}]	Bimolecular recombination rate constant [$cm^3\ s^{-1}$]	Auger recombination rate constant [$cm^6\ s^{-1}$]	
$CH_3NH_3PbI_3$	15×10^6	0.6×10^{-10}	1.6×10^{-28}	Ref. S4
$CH_3NH_3PbI_{3-x}Cl_x$	5×10^6	0.9×10^{-10}	1×10^{-28}	Ref. S5
$CH(NH)_2PbI_3$	7×10^6	1.1×10^{-10}	0.2×10^{-28}	Ref. S6
$CH_3NH_3SnI_3$	8×10^9	14×10^{-10}		Ref. S7
$CH_3NH_3PbI_3$ (calculated)		$0.1\sim1 \times 10^{10}$		Ref. S8
$CH_3NH_3PbI_3(Cl)$	$\sim1 \times 10^6$			Ref. S9

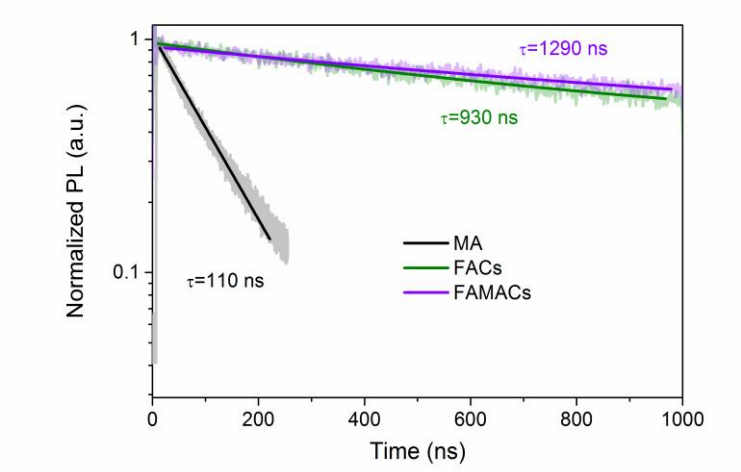

Figure S1. Photoluminescence transients for thin films of various hybrid organic-inorganic perovskites, including a methylammonium (MA) perovskite ($MAPbI_3$), a formamidinium cesium (FACs) double-cation perovskite ($FA_{0.83}Cs_{0.17}PbI_{2.7}Br_{0.3}$) and a formamidinium – methylammonium-cesium (FAMACs) triple cation perovskite ($FA_{0.7}MA_{0.25}Cs_{0.05}PbI_{2.7}Br_{0.3}$). Data were taken using the TCSPC technique, following low-fluence pulsed excitation at a wavelength of 507nm. The lifetime values indicated for τ are derived from mono-exponential fits to the transients.

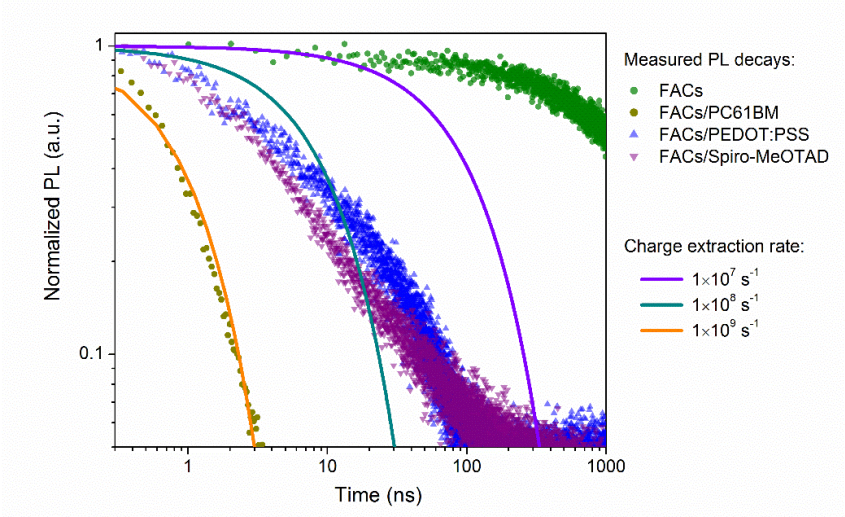

Figure S2. PL quenching transients measured for a thin film of the formamidinium cesium (FACs) double cation perovskite ($FA_{0.83}Cs_{0.17}PbI_{2.7}Br_{0.3}$) covered with a layer of various

transport materials, shown together with calculated curves based on various constant (time-independent) charge extraction rates (solid lines).

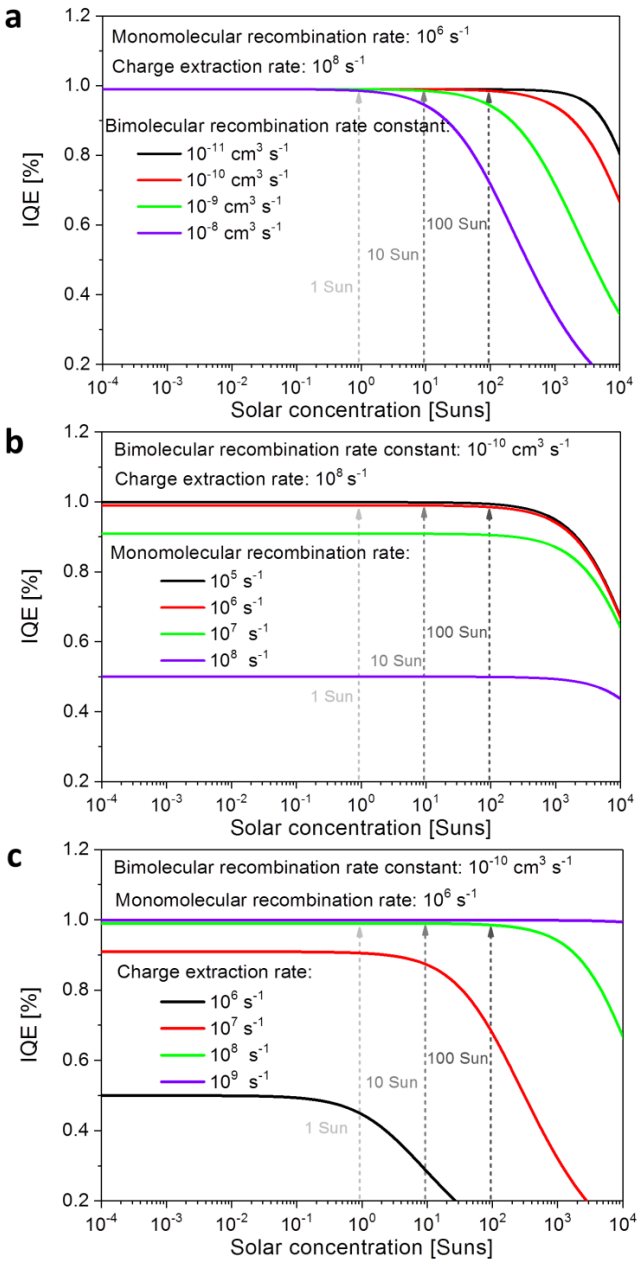

Figure S3. Internal quantum efficiency (IQE) calculated as a function of solar concentration level for a typical single-junction perovskite solar cells (1.6eV bandgap), for variation of (**a**) the bimolecular charge-carrier recombination rate constant k_2, (**b**) the monomolecular (trap-mediated) recombination rate k_1 and (**c**) the charge extraction rate c_{ext}. Unvaried parameters were set to $k_1=10^6\ s^{-1}$, $k_2=10^{-10}\ cm^3 s^{-1}$, $k_3=10^{-28}\ cm^6 s^{-1}$ and $c_{ext}=10^8\ s^{-1}$ for the calculations.

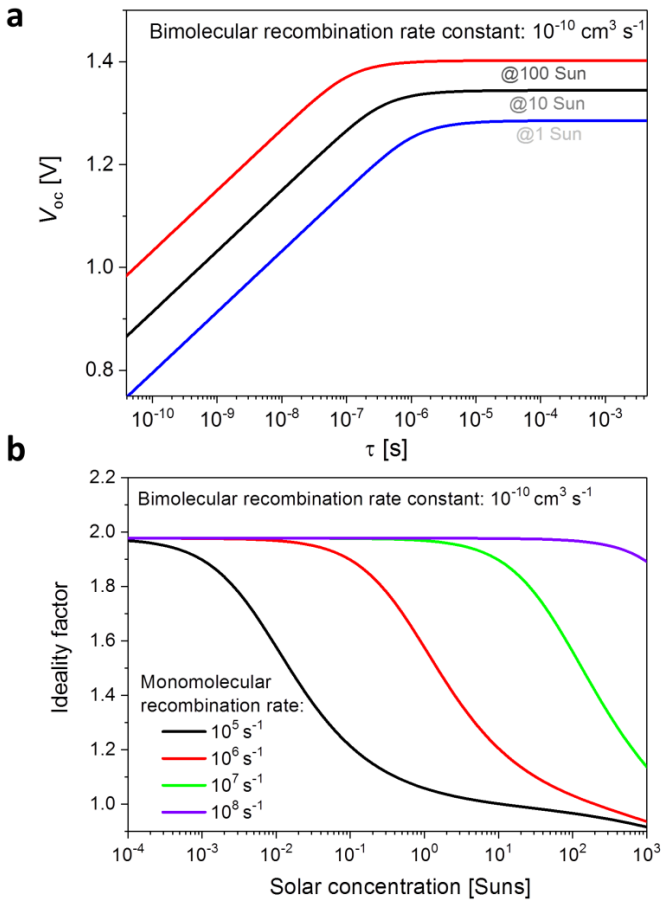

Figure S4. (**a**) Open circuit voltage as a function of charge-carrier lifetime $\tau=k_1^{-1}$ for a typical single-junction perovskite solar cell, calculated for a number of different solar concentration levels. (**b**) Ideality factor *m*, for a range of different monomolecular recombination rates k_1, as derived from the slope of the curves shown in **Figure 3b** that relate the open-circuit voltage V_{oc} to the Solar concentration (Suns), where $m= (q/k_BT)*d(V_{oc})/d(\ln(Suns))$,[S10] and *Suns* is the illumination intensity normalized to that under standard solar (AM1.5) conditions. The Auger rate constant was set to $k_3=10^{-28}$ cm^6s^{-1} for all calculations.

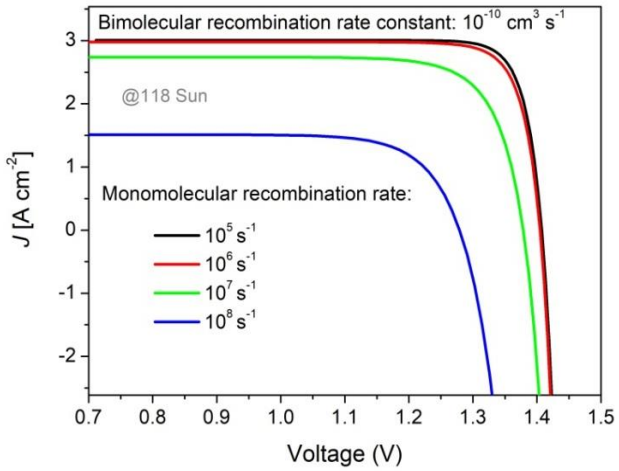

Figure S5. Calculated *J*-*V* curves based on the Suns-V_{oc} method at an illumination of 118 Sun, for variation of the monomolecular recombination rate k_1 for fixed values of k_2=10^{-10} cm^3s^{-1} and k_3=10^{-28} cm^6s^{-1}.

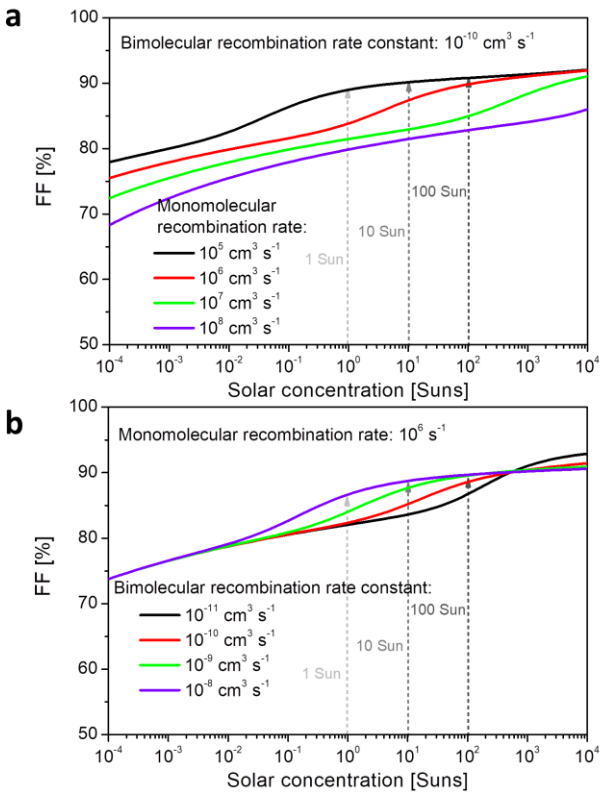

Figure S6. Fill factor as a function of solar concentration level calculated based on empirical relationship between FF and V_{oc},[S11] for variation of (**a**) the bimolecular charge-carrier recombination rate constant k_2 at a fixed value of k_1=10^6 s^{-1}, and (**b**) the monomolecular (trap-

mediated) recombination rate k_1 for a fixed value of $k_2=10^{-10}\,cm^3s^{-1}$. The Auger rate constant was set to $k_3=10^{-28}\,cm^6s^{-1}$ for all calculations.

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