Supplementary Data

Comparative studies of optoelectrical properties of prominent PV materials: Halide Perovskite, CdTe, and GaAs

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1. Effect of numerical aperture in confocal PL measurement

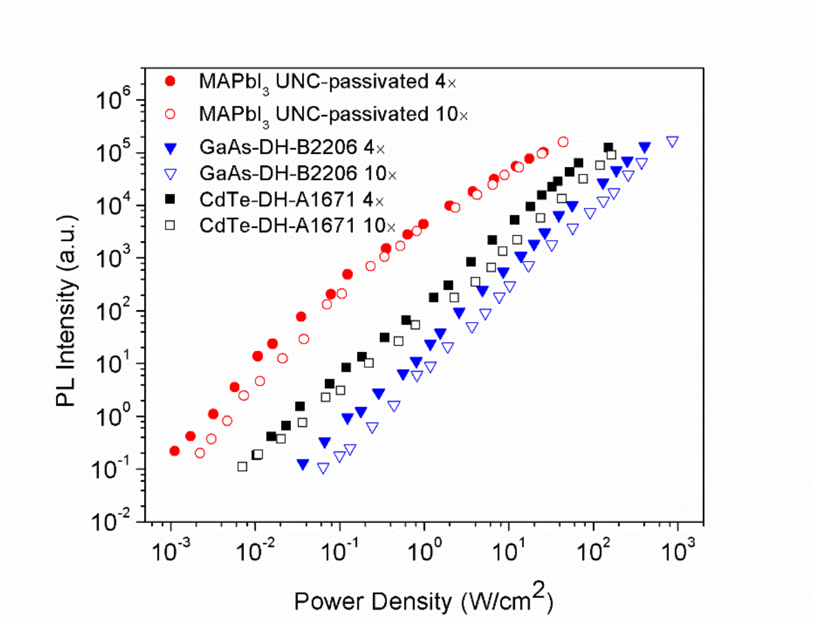

Figure S1. PL intensity vs. excitation density using two microscope lenses: 4× with NA = 0.1, and 10× with NA = 0.25. Results are shown for three samples: $MAPbI_3$ (UNC-passivated), CdTe (CdTe-DH-A1671), and GaAs (GaAs-DH-B2206).

2. PL intensity time dependence

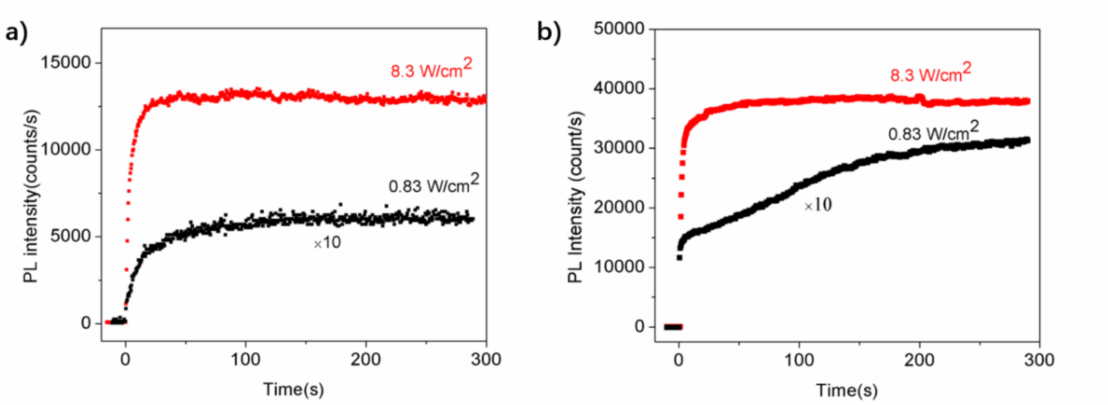

Figure S2. PL intensity time map of thin film perovskite under 10× lens. (a) $MAPbI_3$-LANL at 0.83 W/cm^2 and 8.3 W/cm^2; (b) $MAPbI_3$-UNC-passivated at 0.83 W/cm^2 and 8.3 W/cm^2.

3. Absorption spectra of thin-film perovskite $MAPbI_3$

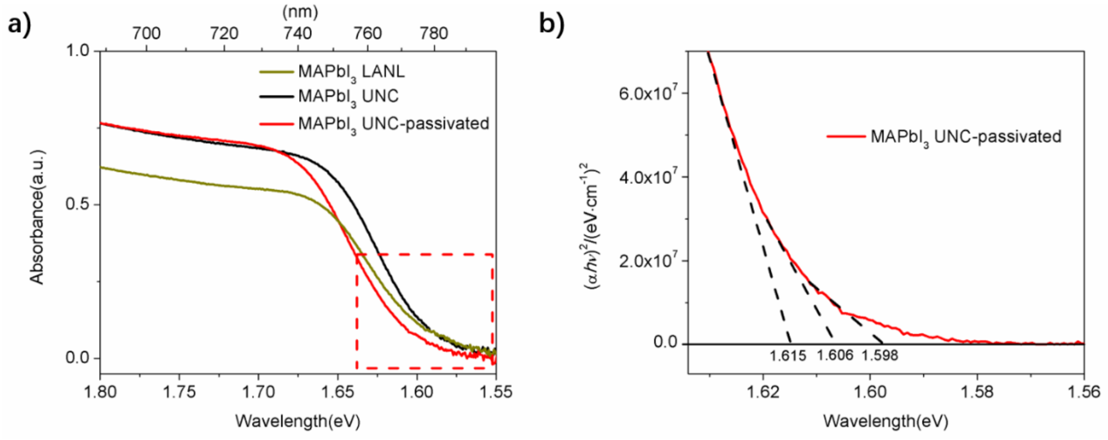

Figure S3. (a) Absorption spectra of thin film perovskite measured by 10× lens. Reflection has been considered. (b) Tauc plot of UNC-passivated sample, corresponding to red dashed box in Figure S3(a). Dashed lines show possible extrapolation.

4. Photon-recycling effect

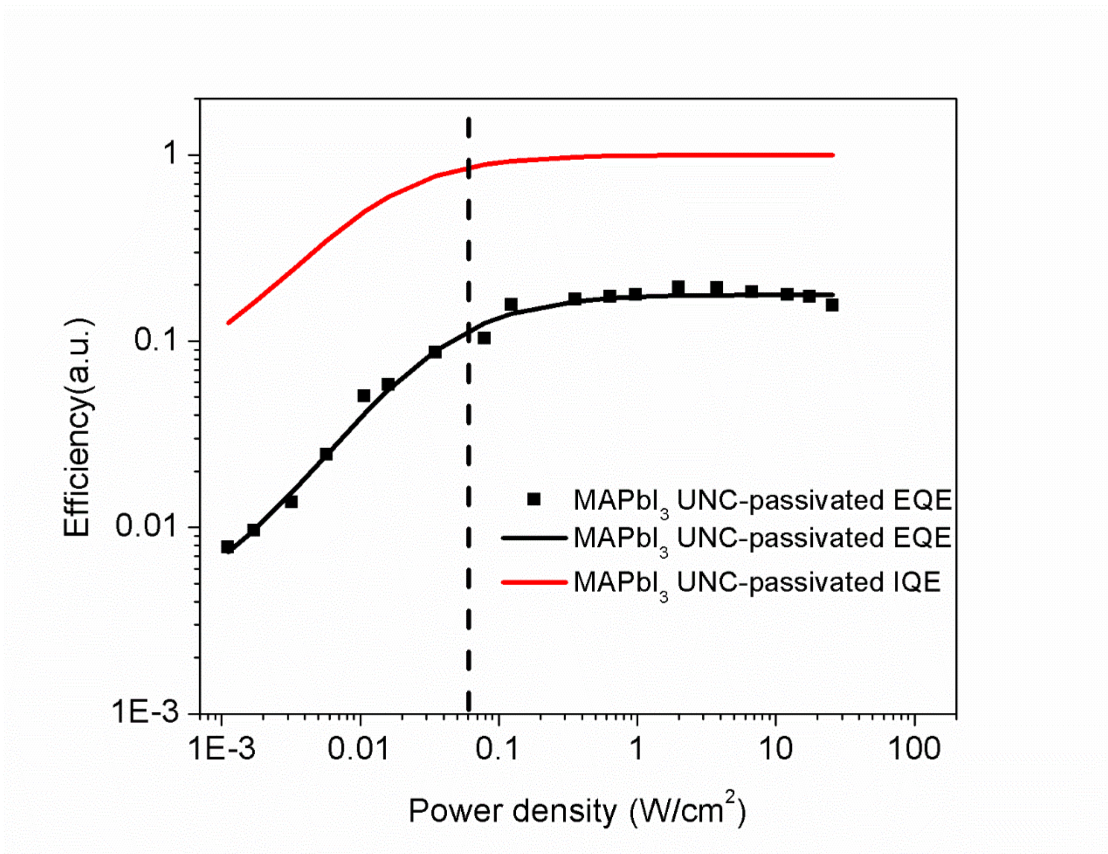

Figure S4. Excitation density dependent external quantum efficiency for $MAPbI_3$ sample UNC-passivated. Black solid lines are fitted curve and vertical dashed line shows one sun illumination. Red line is the internal quantum efficiency for the same data.

We use UNC-passivated as an example to examine the possible effect of photo-recycling on the extracted internal quantum efficiency value.

$$\eta_{IQE} = \frac{1}{2}\left(1 - \beta_0'\left(1 + dx^{\zeta}\right)\frac{u(1+dP^{\zeta})+1}{P} + \sqrt{(1 + \beta_0'(1 + dP^{\zeta})\frac{u(1+dP^{\zeta})+1}{P})^2 - \frac{4\beta_0'(1+dP^{\zeta})}{P}}\right) \quad \text{(S1)}$$

$$\eta_{EQE} = \frac{\frac{C}{2{n_r}^2}\eta_{IQE}}{\frac{1}{2{n_r}^2}\eta_{IQE}+1-\eta_{IQE}+\frac{L}{4\alpha_0 d_0}} \quad \text{(S2)}$$

Internal quantum efficiency η_{IQE} given by Eq. (S1) is the same formula as Eq. (4) in main text for relative external quantum efficiency η_{EQE}, except for removing the constant C from Eq. (5). Measured relative external efficiency η_{EQE} can be linked to η_{IQE} through Eq. (S2) that adopts the relationship between IQE and absolute EQE proposed in Ref. [1], where C is a scaling factor because we do not measure the absolute EQE. n_r is the average refractive index at the perovskite PL peak position, α_0 is the average band-edge absorption coefficient over the perovskite emission band, d_0 is the absorber thickness, L is the loss factor, defined as L = 1−Reflectivity. Eq. (S2) can be used to fit the experimental data of relative EQE. The absolute EQE can then be calculated by dividing fitting curve of Eq. (S2) and the experimental data with C. Taking the same value of n_r = 2.65, and the same α_0 value as in Ref. [2], but adjusted for the thickness difference (in our case d_0 = 500 nm), yielding $\alpha_0 d_0$ = 0.6, and L = 0.796. The fitting results of the IQE and absolute EQE curve are shown in Figure S4. Not only the relative EQE curves are very similar between this work and that in Ref. [2], if fitted with Eq. (S2), our data would also give rise to a similar IQE (85%) vs. 92% there under 1 Sun equivalent (60 mW/cm^2) [2].

[1] I. Schnitzer, E. Yablonovitch, C. Caneau, T.J. Gmitter, Ultrahigh spontaneous emission quantum efficiency, 99.7% internally and 72% externally, from AlGaAs/GaAs/AlGaAs double heterostructures, Applied Physics Letters, 62 (1993) 131-133.

[2] I.L. Braly, D.W. deQuilettes, L.M. Pazos-Outón, S. Burke, M.E. Ziffer, D.S. Ginger, H.W. Hillhouse, Hybrid perovskite films approaching the radiative limit with over 90% photoluminescence quantum efficiency, Nature Photonics, 12 (2018) 355-361.