## **Supporting Information for**

## Bulk Heterojunction Quasi-Two-Dimensional Perovskite Solar Cell with 1.18 V High Photovoltage

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**Figure S1.** (a) UV absorption spectra, (b) its logarithmic plots of Q-2D perovskites with 33.3% PMA to 100% PMA molar ratio. The inset shows the optical image.



**Figure S2.** Power dependent TRPL spectrum for 33.3% PMA Q-2D perovskite film at (a) 576 nm, (b) 626 nm, (c) 740 nm, (d) 770 nm and 40% PMA Q-2D perovskite film at 60 K at (e) 578 nm (f) 740 nm with varying excitation power from 0.32 nJ/cm<sup>2</sup> to 15.68 nJ/cm<sup>2</sup>.



**Figure S3.** (a) 2D pseudocolor transient absorption spectrum for 33.3% PMA Q-2DP thin film illuminated from (a) front side and (b) back side with power density  $1.5\mu$ J/cm<sup>2</sup>. (c) (d) time slices showing the bleach rise and decay kinetics at the bleaching peaks corresponding to (a) and (b) with power density  $1.5\mu$ J/cm<sup>2</sup>.



**Figure S4.** (a) Temperature dependent steady state PL spectrum for 33.3%PMA Q-2D perovskite film at wavelength range from 540 nm to 700 nm (b) from 700 nm to 900nm (c) Normalized steady state PL spectrum at wavelength range 700 nm to 900nm.



**Figure S5.** TRPL for 40%PMA Q-2DP. (a) PL spectra at different temperature. (b-f) Decay profiles in time scale at (b) 570 nm, (c) 613 nm, (d) 640 nm, (e) 670 nm and (f) 752nm.



**Figure S6.** Steady state PL spectrum of BHJ perovskite bare film on glass substrate illuminated from front side, i.e. perovskite side (a) and also from back side, i.e. glass side (b) with different excitation power from 0.01 to 3.2 mW/cm<sup>2</sup>.



**Figure S7.** The polar intensity profiles along the ring in the  $q_r$  range of 0.96 to 1.02 Å<sup>-1</sup> for 33.3%PMA (a) BHJ film (b) Planar film.



Figure S8. Azimuthally integrated intensity plots for 33.3%PMA BHJ film and Planar film.



Figure S9. Device statistics on the PCE for planar and BHJ 33.3%PMA Q-2D PSCs.



Figure S10. Forward and back JV scan characteristics for BHJ and planar 33.3% PMA Q-2D solar cells , at scanning rate 0.4V/s

Device	Scan Direction	$J_{SC}$ at $0.8V_{OC}$	Hysteresis index
		(mA/cm <sup>2</sup> )	
BHJ	Reverse	10.76	0.167
	Forward	8.96	
Planar	Reverse	6.49	0.764
	Forward	1.53	

Table S1. Summary of JV characteristic for BHJ and Planar 33.3%PMA Q-2D PSC.

Hysteresis index (HI) =  $\frac{J(0.8Voc) - JFS(0.8Voc)}{JRS(0.8Voc)}$ 



**Figure S11.** Steady state current density and PCE over time for 33.3%PMA BHJ and planar Q-2D PSC.



**Figure S12.** Device statistics of dripping time effect on performance metrics for 33.3% PMA BHJ Q-2D PSC. (a)  $V_{OC}$  (b)  $J_{SC}$  (c) FF (d) PCE.



Figure S13. Solvent engineering for 33.3% PMA Q-2D PSC. (a)  $V_{OC}$  (b)  $J_{SC}$  (c) FF (d)

PCE.



Figure S14. Humidity stability test for 33.3% PMA Q-2D PSC under 65% RH without encapsulation.