

Electronic Supplementary Information (ESI)

A Sandwich-Like Structural Model Revealed for Quasi-2D Perovskite Films

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Supplementary Text:

The crystal parameters (a , c , b) of the $\text{BA}_2\text{MA}_{n-1}\text{Pb}_n\text{I}_{3n+1}$ RPPs with $n=3, 4, 5$ are taken from literature reports and listed in **Table S1**.^{1,2} For large- n ($n=L$), but not 3D, RPPs, the a , c values are expected to be identical to those for small- n RPPs and the b value can be approximated to be infinity, as b has little influence on the peak position of the $(\bar{1}1\bar{1})$ diffraction feature for large- n RPPs. The q values for $(\bar{1}1\bar{1})$ diffraction peaks are calculated by the following equation:

$$q_{(\bar{1}1\bar{1})} = 2\pi \sqrt{\frac{1}{a^2} + \frac{1}{b^2} + \frac{1}{c^2}} \quad (\text{S-1})$$

Table S1. The crystal parameters (a , c , b) and the calculated $q_{(\bar{1}1\bar{1})}$ values for $\text{BA}_2\text{MA}_{n-1}\text{Pb}_n\text{I}_{3n+1}$ RPPs with $n=3, 4, 5$ and $n=L$.

	a (Å)	c (Å)	b (Å)	$q_{(\bar{1}1\bar{1})}$ (nm ⁻¹)
$n=3$	8.93	8.88	51.96	10.06
$n=4$	8.93	8.88	64.38	10.02
$n=5$	8.93	8.91	77.01	9.99
$n=L$	8.93	8.91	∞	9.95

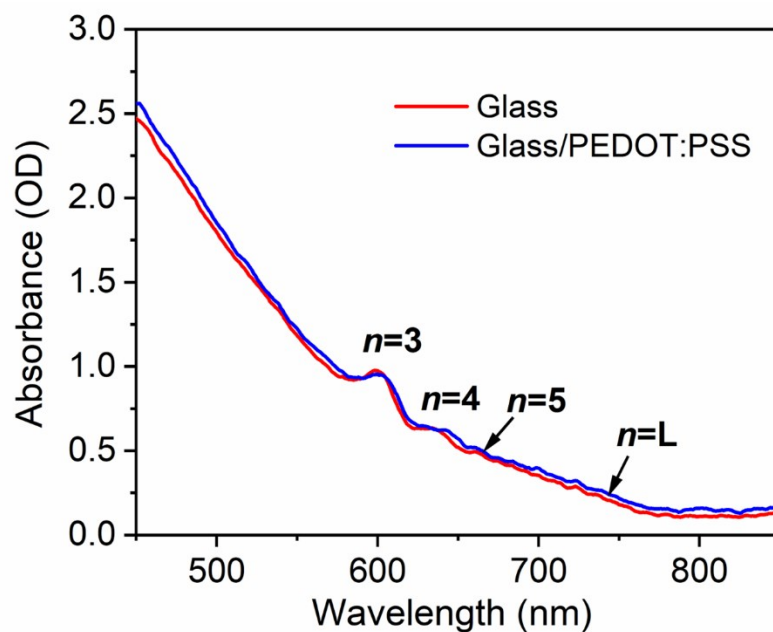


Fig. S1 UV-Vis absorption spectra of the $\text{BA}_2\text{MA}_{n-1}\text{Pb}_n\text{I}_{3n+1}$ ($\langle n \rangle = 5$) quasi-2D perovskite films drop-cast on bare glass and PEDOT:PSS coated glass substrates.

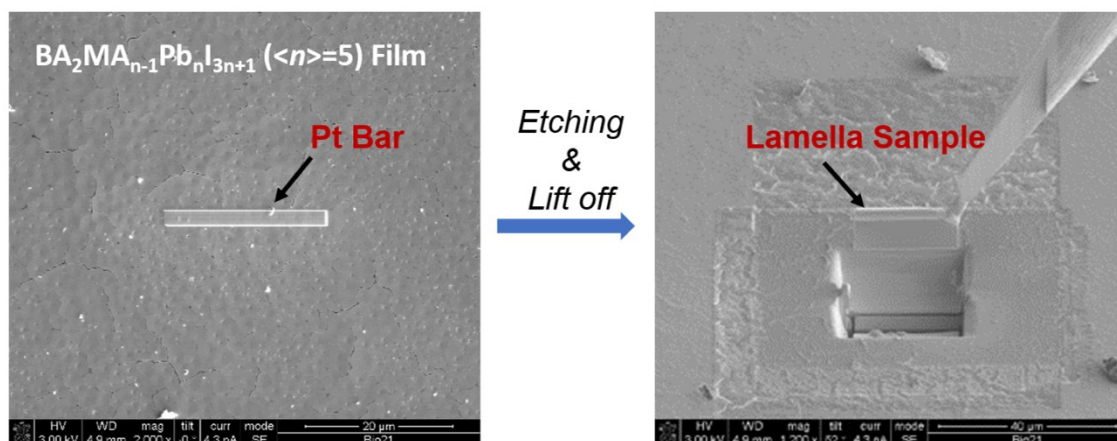


Fig. S2 The procedure for preparing the $\text{Glass}/\text{BA}_2\text{MA}_{n-1}\text{Pb}_n\text{I}_{3n+1}$ ($\langle n \rangle = 5$) Quasi-2D Perovskite/Carbon/Pt lamella sample using the lift-off FIB technique.

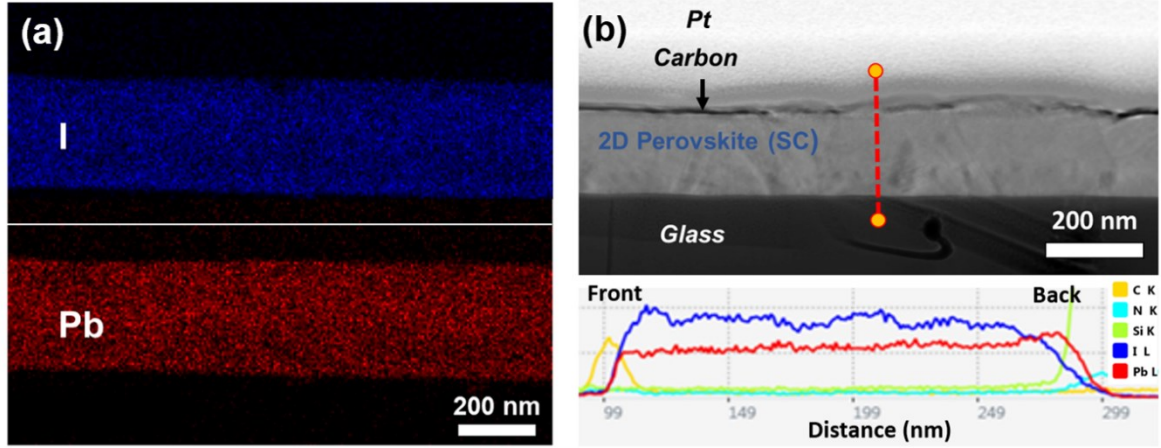


Fig. S3 (a) STEM-EDAX elemental mapping of iodine (I) (upper panel) and lead (Pb) (lower panel) in the drop-cast $\text{BA}_2\text{MA}_{n-1}\text{Pb}_n\text{I}_{3n+1}$ ($\langle n \rangle = 5$) Quasi-2D perovskite film lamella within the rectangular area indicated in Fig. 1e; (b) The high-angle annular dark-field (HAADF) STEM image of the spin-coated (SC) $\text{BA}_2\text{MA}_{n-1}\text{Pb}_n\text{I}_{3n+1}$ ($\langle n \rangle = 5$) quasi-2D perovskite film lamella (upper panel) and the corresponding EDAX line scan profile of I and Pb along the labeled red dashed line (lower panel).

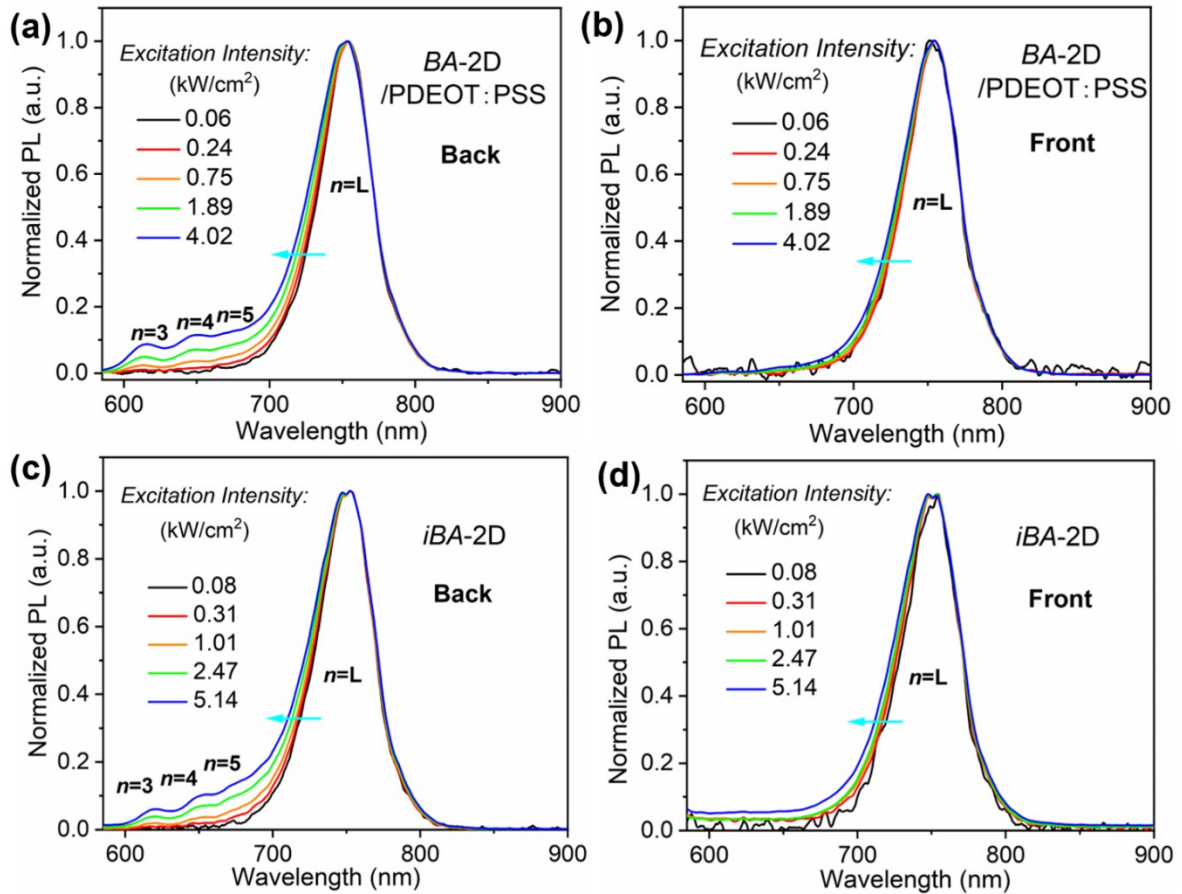


Fig. S4. Excitation intensity-dependent PL spectra collected from (a) the back-side and (b) the front-side of the $\text{BA}_2\text{MA}_{n-1}\text{Pb}_n\text{I}_{3n+1}$ ($\langle n \rangle = 5$) quasi-2D perovskite films drop-cast on the PEDOT:PSS coated glass substrate; (Excitation laser: 400 nm, 5.4 MHz.) Excitation intensity-dependent PL spectra collected from (c) the back-side and (d) the front-side of the $\text{iBA}_2\text{MA}_{n-1}\text{Pb}_n\text{I}_{3n+1}$ ($\langle n \rangle = 5$) quasi-2D perovskite films drop-cast on the glass substrate. (Excitation laser: 450 nm, 5.4 MHz.)

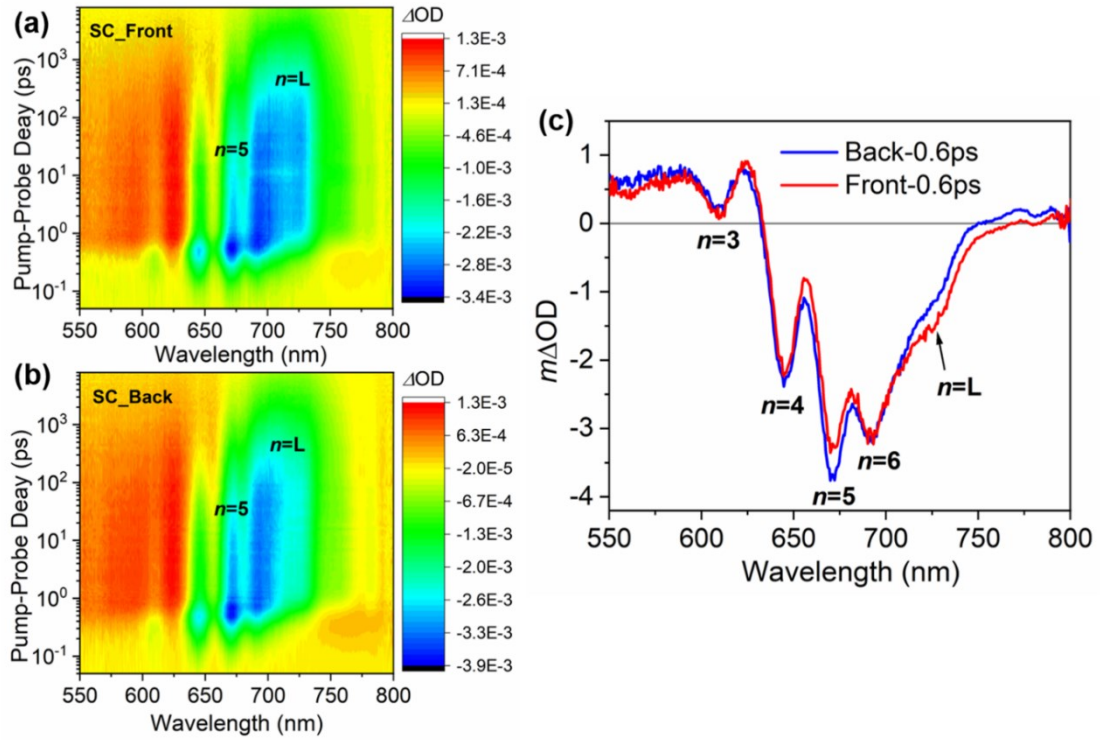


Fig. S5. Pseudo-color 2D plots of TA spectra (ΔOD) of the spin-coated (SC) $BA_2MA_{n-1}Pb_nI_{3n+1}$ ($\langle n \rangle = 5$) quasi-2D perovskite film obtained under (a) front- and (b) back-side excitation; (c) TA spectra of the (SC) quasi-2D perovskite film as a function of probe wavelength recorded at 0.6 ps pump-probe delay.

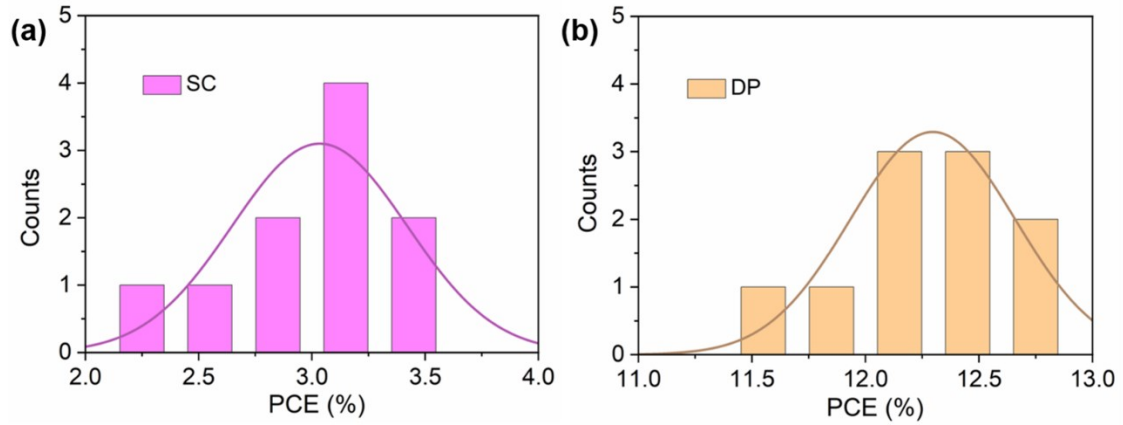


Fig. S6. The statistics of PCE distribution for (a) spin-coated (SC) PSCs and (b) drop-cast (DP) PSCs based on $BA_2MA_{n-1}Pb_nI_{3n+1}$ ($\langle n \rangle = 5$) quasi-2D perovskite films.

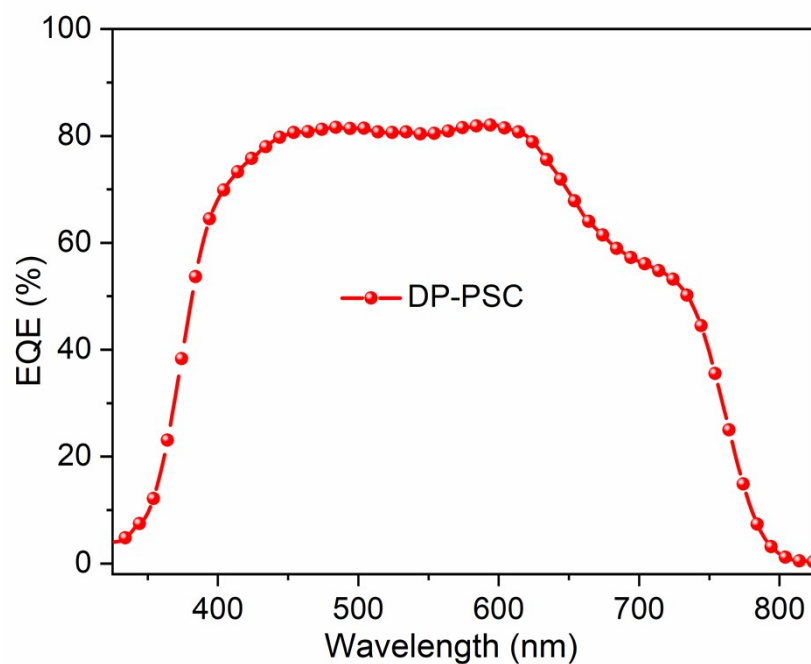


Fig. S7. The external quantum efficiency (EQE) spectrum of a perovskite solar cell based on the drop-cast $\text{BA}_2\text{MA}_{n-1}\text{Pb}_n\text{I}_{3n+1}$ ($\langle n \rangle = 5$) quasi-2D perovskite film (DP-PSC).

References

- 1 C. C. Stoumpos, D. H. Cao, D. J. Clark, J. Young, J. M. Rondinelli, J. I. Jang, J. T. Hupp, M. G. Kanatzidis, *Chem. Mater.*, 2016, **28**, 2852-2867.
- 2 C. C. Stoumpos, C. M. M. Soe, H. Tsai, W. Nie, J.-C. Blancon, D. H. Cao, F. Liu, B. Traoré, C. Katan, J. Even, A. D. Mohite, M. G. Kanatzidis, *Chem*, 2017, **2**, 427-440.