

Supporting information

Phase Distribution Regulation of Formamidinium-based Quasi-2D Perovskites through Solution Engineering

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Table S1. Orthogonal experiments using different solvent combinations and excess doping fractions for quasi-2D perovskites (PDMA)(FA)₄Pb₅I₁₆.

	Without additive	10 mol% MACl	10 mol% NH ₄ SCN	10 mol% MACl + NH ₄ SCN	30 mol% MACl
DMF:DMSO = 4:1	δ-phase	Low surface coverage	δ-phase	Low surface coverage	-
DMF:NMP = 9:1	Minor δ-phase	α-phase	Minor δ-phase	α-phase with peak split	α-phase with peak split

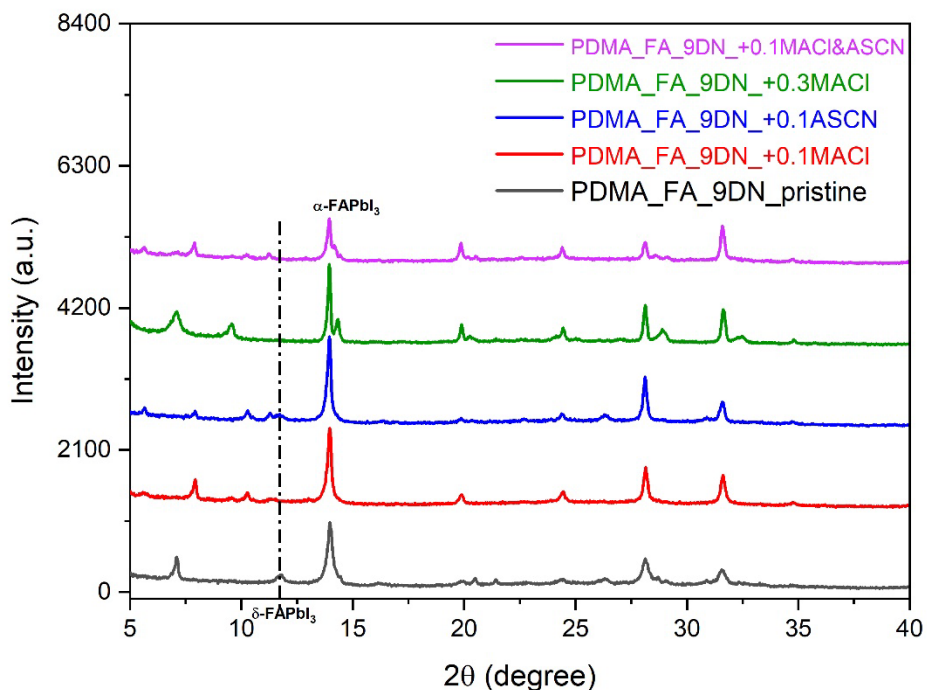


Figure S1. Power X-ray diffractograms of quasi-2D perovskites (PDMA)(FA)₄Pb₅I₁₆ with fixed solvent combination of 9:1 (v/v) DMF:NMP and different additive doping ratios.

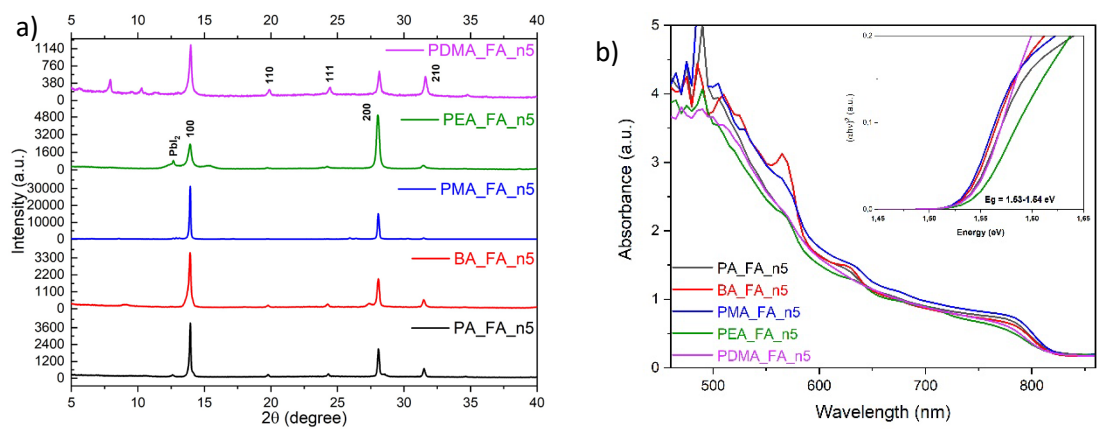


Figure S2. Crystal orientation and optical properties of quasi-2D perovskites $(L)_m(\text{FA})_4\text{Pb}_5\text{I}_{16}$, $L = \text{PA}, \text{BA}, \text{PMA}, \text{PEA}, \text{PDMA}$. (a) Power X-ray diffractograms. (b) UV-Vis absorption spectra (Tauc plot in the inset).

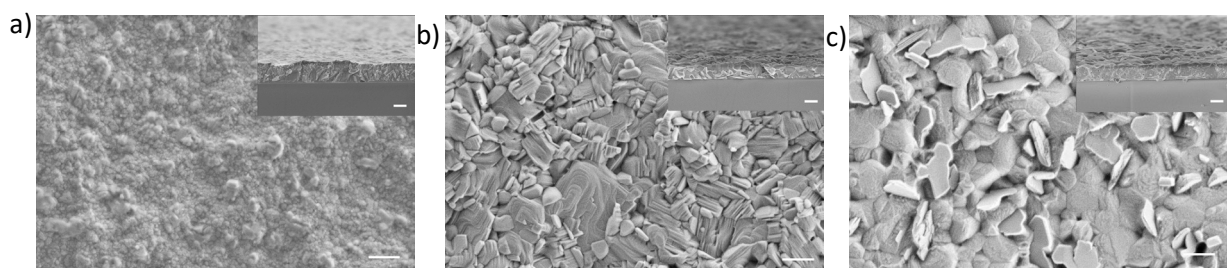


Figure S3. Surface morphologies of the films of $(\text{PDMA})(\text{FA})_4\text{Pb}_5\text{I}_{16}$, $(\text{PA})_2(\text{FA})_4\text{Pb}_5\text{I}_{16}$, $(\text{ALA})_2(\text{FA})_4\text{Pb}_5\text{I}_{16}$, respectively, with solvent ratios of 9:1 (v/v) DMF:NMP, and 10 mol% excess MAcl doping. Insets are the cross-sectional views, all scale bars are 500 nm.

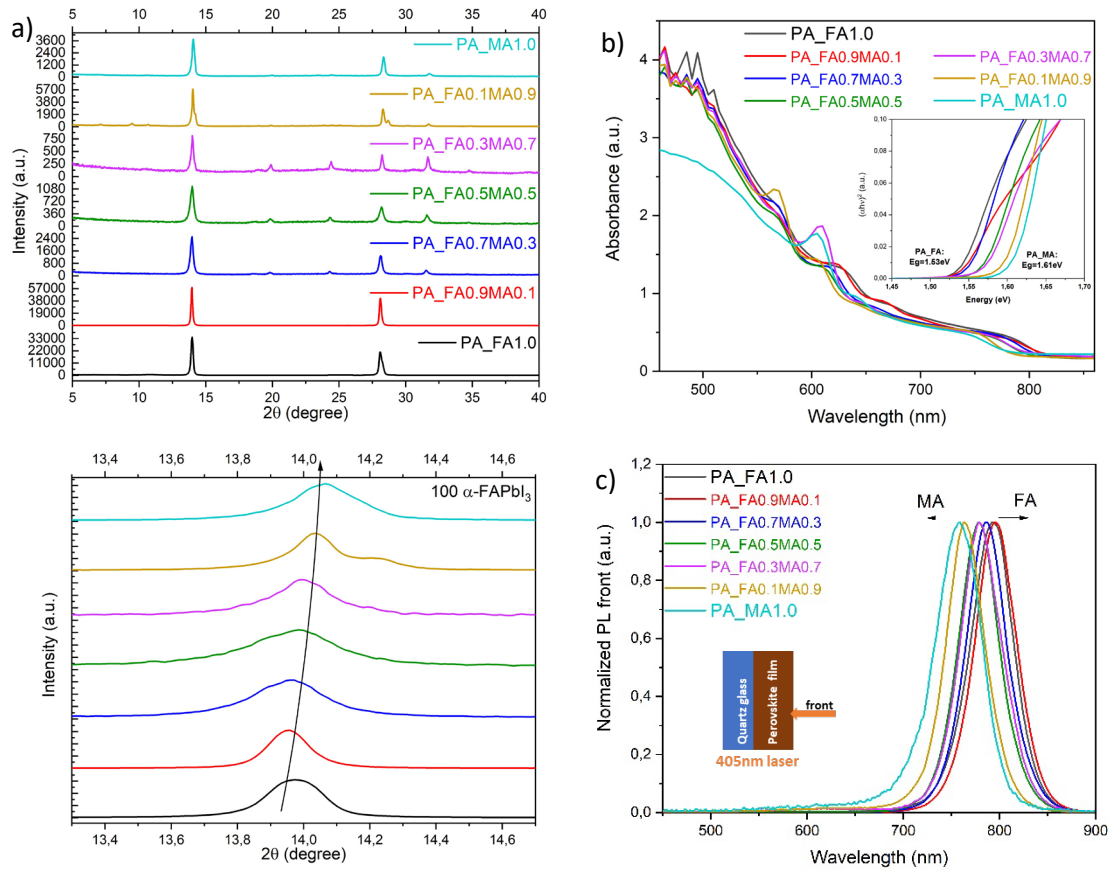


Figure S4. Crystalline orientation, optical properties and phase distribution of quasi-2D perovskites $(PA)_2(FA)_4Pb_5I_{16}$ with A-site cations (FA and MA) mixing. (a) Powder X-ray diffractograms (upper part), and magnified 100 peak (lower part). (b) UV-Vis absorption spectra (Tauc plot in the inset). (c) Steady-state PL from front (perovskite) with a near-UV laser of 405 nm.