## **Supporting Information**

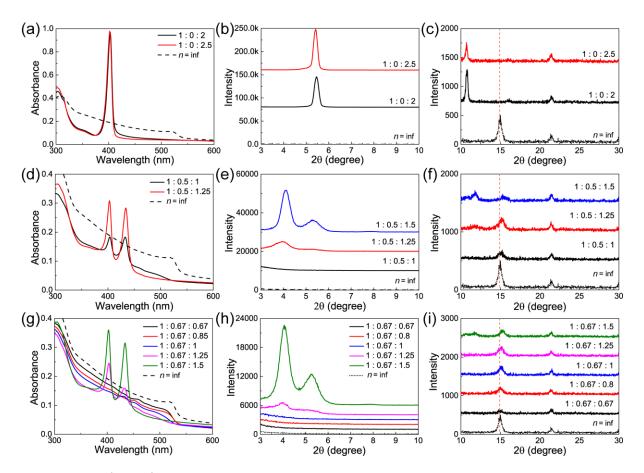
## Phase Control of Quasi-2D Perovskites and Improved Light-Emitting Performance by Excess Organic Cations and Nanoparticle Intercalation

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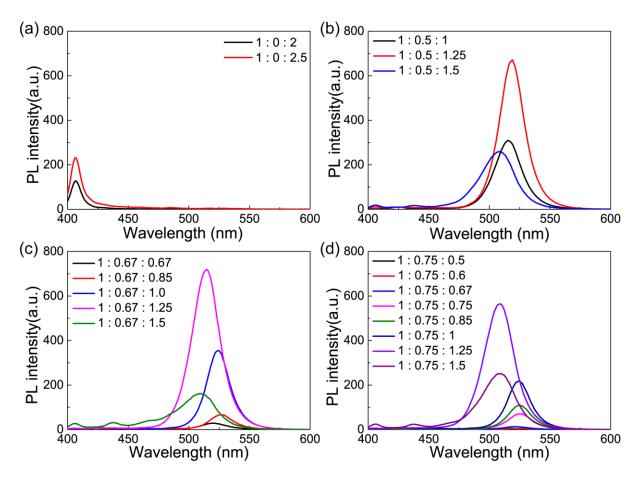
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**Figure S1.** Absorption spectra of  $(PEA)_2MA_{n-1}Pb_nBr_{3n+1}$  films prepared with a Pb:MA:PEA molar ratio of (a) 1:0:*x*, (d) 1:0.5:*x* and (g) 1:0.67:*x*. X-ray diffraction patterns of  $(PEA)_2MA_{n-1}Pb_nBr_{3n+1}$  films prepared with a Pb:MA:PEA molar ratio of (b, c) 1:0:*x*, (e, f) 1:0.5:*x* and (h, i) 1:0.67:*x* at low and high angles. The dashed curve represents the absorption spectrum and the XRD patterns of 3D perovskites. The vertical dashed line drawn in (c, f, i) represents the XRD diffraction peak for (100) plane of 3D perovskites.



**Figure S2.** Photoluminescence of  $(PEA)_2MA_{n-1}Pb_nBr_{3n+1}$  films with excess PEA cations; (a) without MA, (b) MA:Pb = 0.5:1, (c) MA:Pb = 0.67:1 and (d) MA:Pb = 0.75:1 molar ratios. The ratio shown in each subfigure represents a Pb:MA:PEA molar ratio.

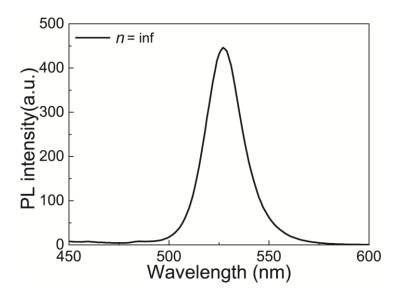


Figure S3. Photoluminescence of MAPbBr<sub>3</sub> 3D perovskite films.

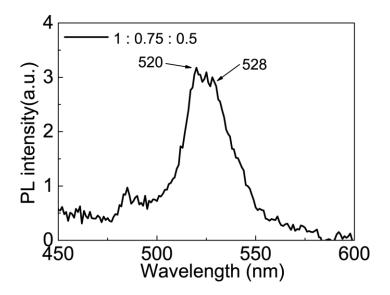
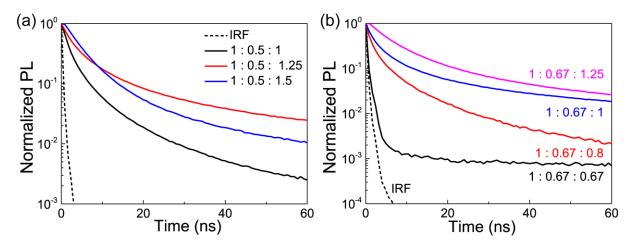
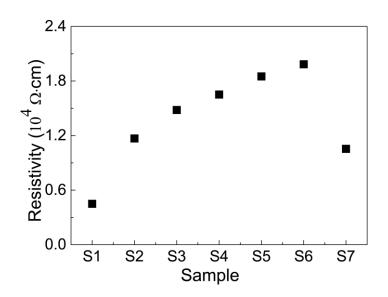


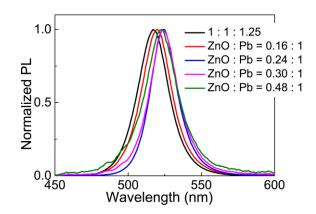
Figure S4. Photoluminescence of  $(PEA)_2MA_{n-1}Pb_nBr_{3n+1}$  films with a Pb:MA:PEA molar ratio of 1:0.75:0.5.



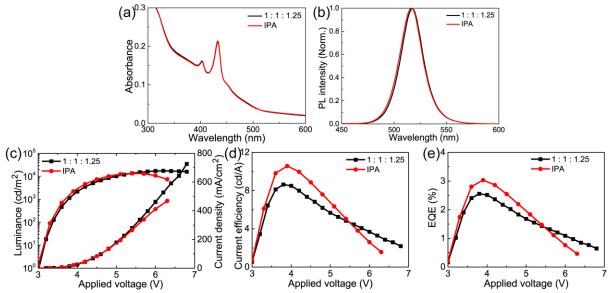
**Figure S5.** Time-resolved photoluminescence of  $(PEA)_2MA_{n-1}Pb_nBr_{3n+1}$  films with excess PEA cations; (a) MA:Pb = 0.5:1 and (b) MA:Pb = 0.67:1 molar ratios. The dashed curve represents the instrument response function (IRF). The ratio shown in each subfigure represents a Pb:MA:PEA molar ratio.



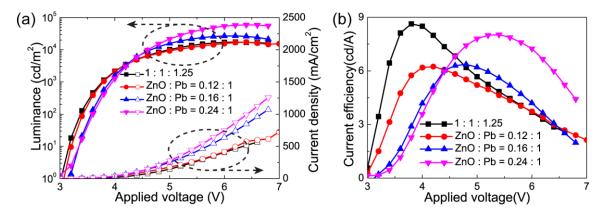
**Figure S6.** Resistivity of  $(PEA)_2MA_{n-1}Pb_nBr_{3n+1}$  films with a Pb:MA:PEA molar ratio of 1:0.75:0.5 (S1), 1:0.75:0.75 (S2), 1:0.75:1 (S3), 1:0.75:1.25 (S4), 1:0.75:1.5 (S5), 1:1:1.25 (S6), 1:1:1.25 with ZnO incorporation (S7, ZnO:Pb molar ratio is 0.24:1).



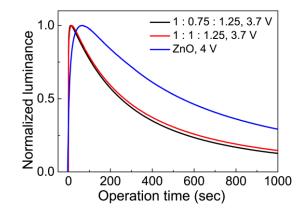
**Figure S7.** Normalized PL with different ZnO NP molar ratios. PL becomes red shift with increasing ZnO NPs and there is no further red shift beyond the ratio of 0.24:1.



**Figure S8.** (a) Absorption and (b) photoluminescence spectra of  $(PEA)_2MA_{n-1}Pb_nBr_{3n+1}$  films with (red) and without (black) IPA additive. (c) Current density-luminance-voltage characteristics, (d) current efficiency and (e) EQE as a function of applied bias for  $(PEA)_2MA_{n-1}Pb_nBr_{3n+1}$  based devices with a Pb:MA:PEA molar ratio of 1:1:1.25 with and without IPA additive.



**Figure S9.** (a) current density-luminance-voltage characteristics and (b) current efficiency (CE) versus voltage of quasi-2D perovskite light-emitting diodes based on blends of  $(PEA)_2MA_{n-1}Pb_nBr_{3n+1}$  and ZnO NPs dispersed in IPA.



**Figure S10.** Operational stability of  $(PEA)_2MA_{n-1}Pb_nBr_{3n+1}$  based device under a constant voltage of 3.7 V and 4V for the devices (a) without ZnO NPs and (b) with ZnO NPs.

Pb:MA:PEA Molar ratio	1:0.75:0.5	1:0.75:0.75	1:0.75:1.0	1:0.75:1.25	1:0.75:1.5	1:1:1.25	1:1:1.25 + ZnO
Thickness (nm)	55	64	83	92	113	112	113

**Table S1.** Thickness of quasi-2D perovskite films prepared on top of the PEDOT:PSS layer.