

Electronic Supporting Information
**Colloidal engineering for monolayer $\text{CH}_3\text{NH}_3\text{PbI}_3$ films toward high
performance perovskite solar cells**

Bo Li^a, Mengjie Li^a, Chengbin Fei^b, Guozhong Cao^{a,c}, Jianjun Tian^{a*}

^a Institute of Advanced Materials and Technology, University of Science and
Technology Beijing, 100083, China.

^b Department of physics, University of Miami, Coral Gables, 33146, USA.

^c Department of Materials and Engineering, University of Washington, Seattle, WA
98195-2120, USA.

Email: tianjianjun@mater.ustb.edu.cn

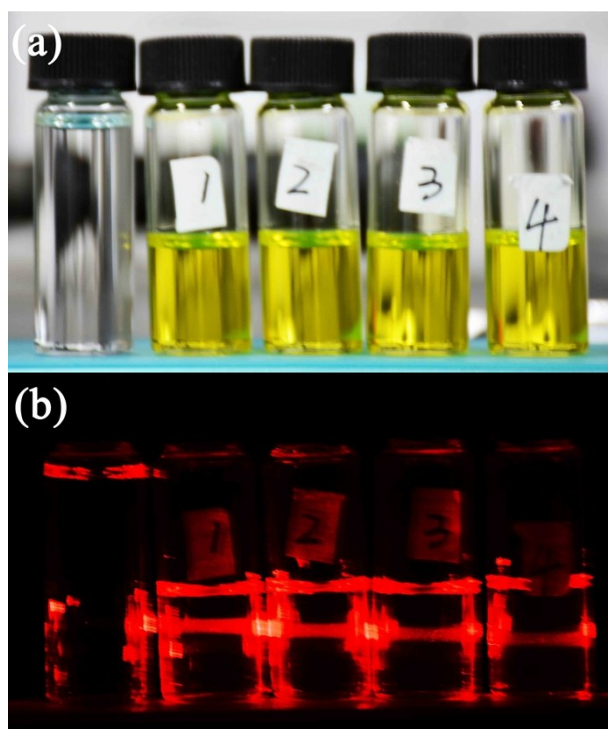


Figure S1 Photographs (a) of perovskite precursor without and with additives, (from left to right: pure DMF, standard perovskite precursor, DMSO added precursor, MACl added precursor, DMSO+MACl added precursor); Tyndall effect (b) in the dark.

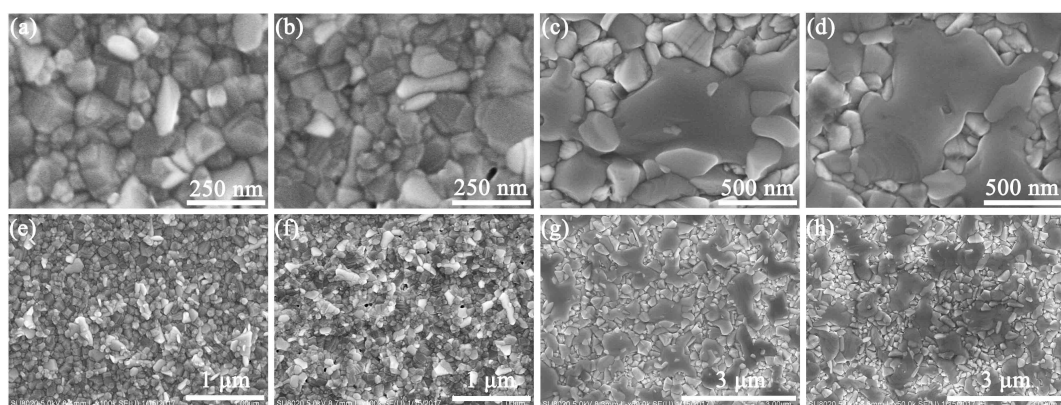


Figure S2 Surface SEM images of perovskite films prepared from different perovskite precursors on FTO substrates, the concentration of perovskite precursors was 0.25 M. The standard perovskite precursor (a, e); With the additive of DMSO (b, f), MACl (c, g) and DMSO+MACl (d, h).

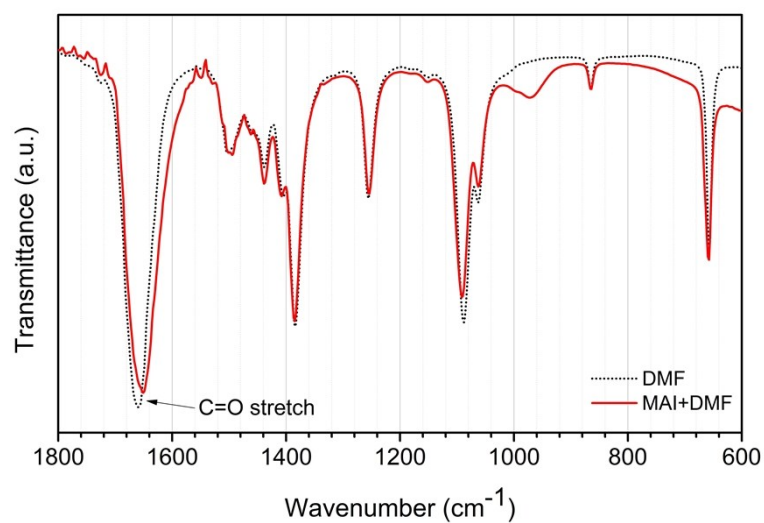


Figure S3 FTIR spectra of pure DMF and MAI in DMF in the range of 1800~600 cm^{-1} .



Figure S4 Photographs precursor solutions with different molar ratio, from left to right:

MAI:PbI₂:DMSO:MAcI=1:0:1:1, MAI:PbI₂:DMSO:MAcI=1:1:1:2 and
MAI:PbI₂:DMSO:MAcI=1:1:1:2.5

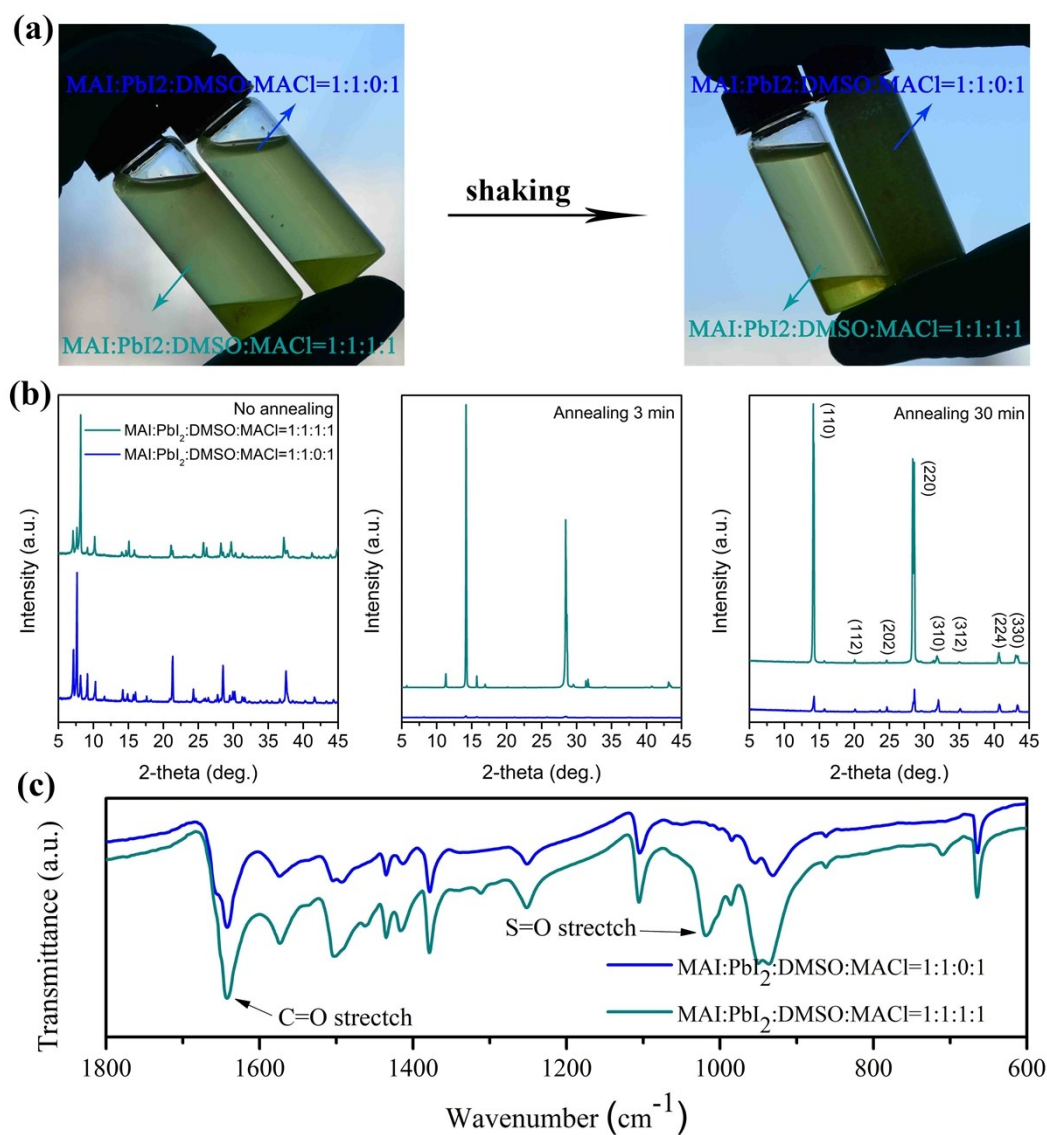


Figure S5 Photographs (a) of colloidal perovskite precursor solution extracted with ethyl acetate before and after shaking; In situ XRD patterns (b) of the different annealing time of gel complexes extracted from colloidal perovskite precursors with the additive of MACl and DMSO+MACl. FTIR spectra (c) of gel complexes extracted from colloidal perovskite precursors with the additive of MACl and DMSO+MACl.

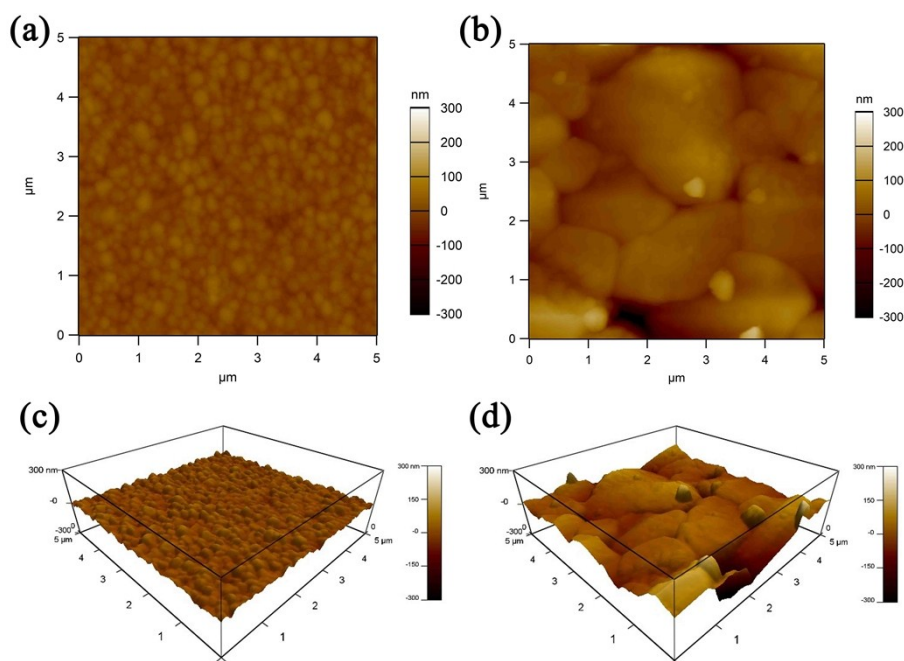


Figure S6 Topography and three-dimensional atomic force microscopy (AFM) images of the perovskite films prepared from the standard perovskite precursors (a, c) and with additive of DMSO+ MACl (b, d).

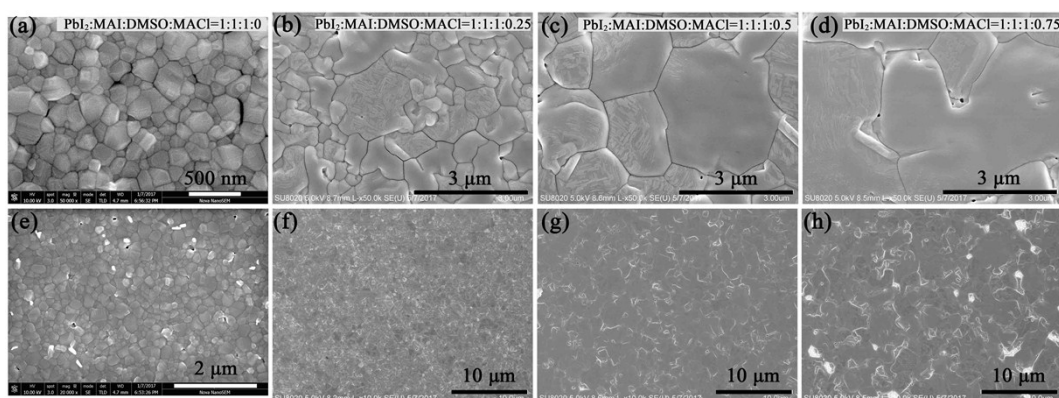


Figure S7 Surface SEM of perovskite films prepared from DMSO+MACl colloidal perovskite precursor with different amount of MACl on FTO substrates. 0 M (a, e); 0.25 M (b, f); 0.5 M (c, g); 0.75 M (d, h).

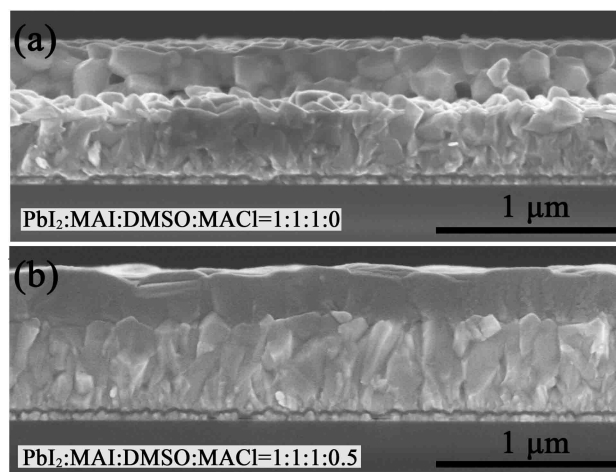


Figure S8 Cross SEM images of multilayer perovskite films (a) and monolayer perovskite films (b) on FTO substrates.

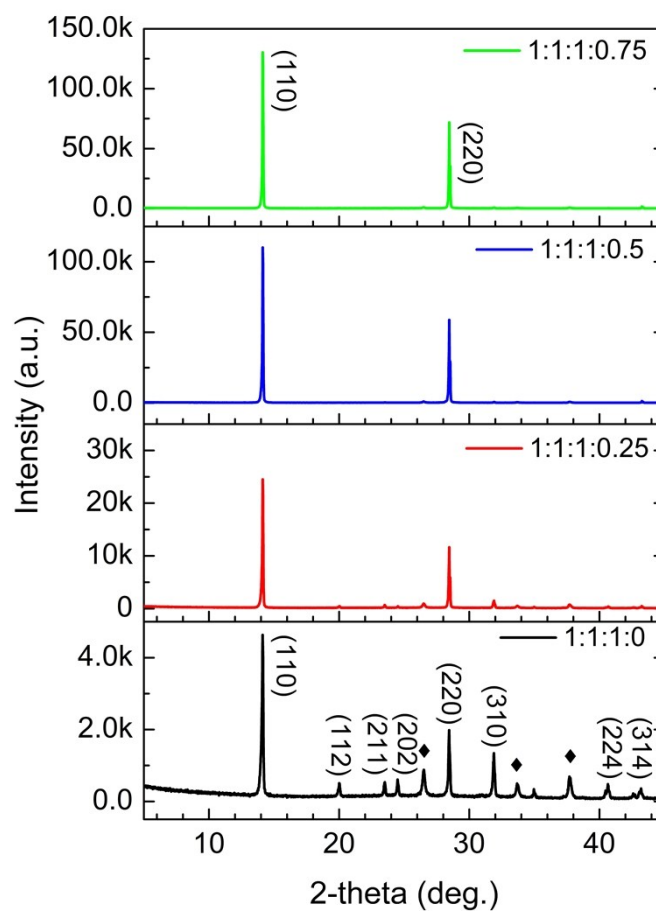


Figure S9 XRD patterns of perovskite films prepared from colloidal perovskite

precursors with different amount of MACl. From bottom to top, the molar ratio of MAI:PbI₂:DMSO:MACl = 1:1:1:0, 1:1:1:0.25, 1:1:1:0.5 and 1:1:1:0.75. The diamond indicates the FTO diffraction peaks.

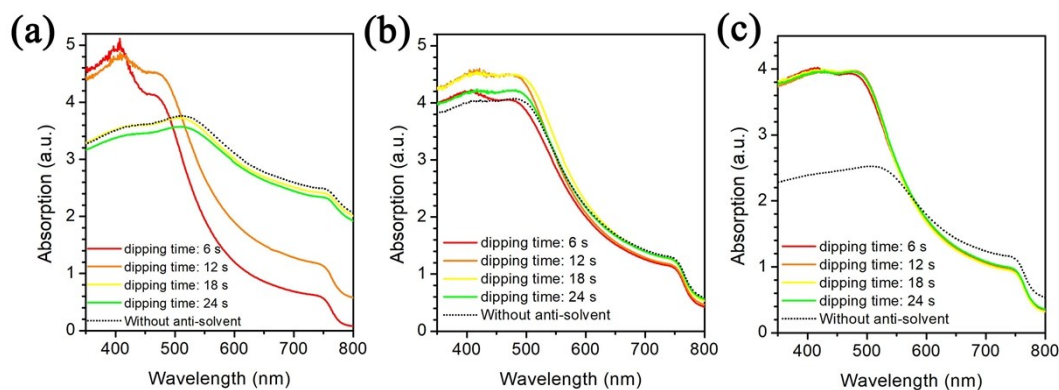


Figure S10 UV-vis absorption spectra of perovskite film prepared from different colloidal perovskite precursor with different anti-solvent dipping time. (a) DMSO added; (b) MACl added; (c) DMSO+MACl added.

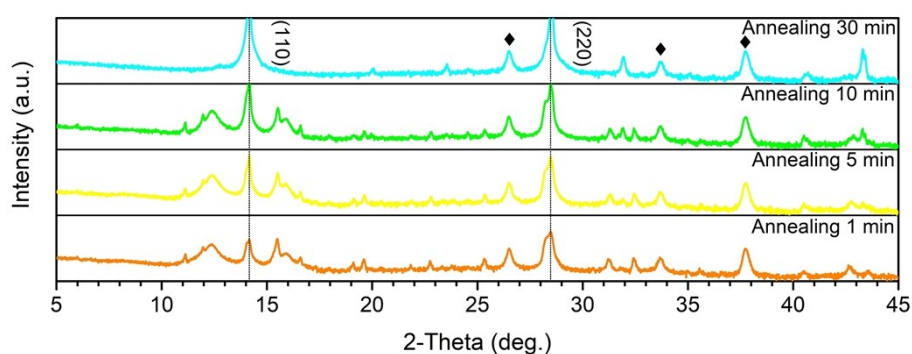


Figure S11 XRD patterns of crystallization evolution as function of annealing time on a logarithmic scale for perovskite films prepared from the perovskite precursor with additive of MACl. The diamond indicates the FTO diffraction peaks.

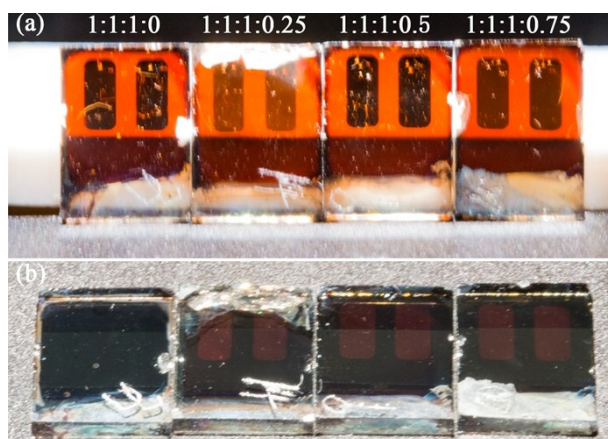


Figure S12 Photographs of perovskite solar cells prepared from different perovskite precursor under parallel illumination (a) and vertical illumination (b). From left to right, the molar ratio of MAI:PbI₂:DMSO:MACl = 1:1:1:0, 1:1:1:0.25, 1:1:1:0.5 and 1:1:1:0.75.

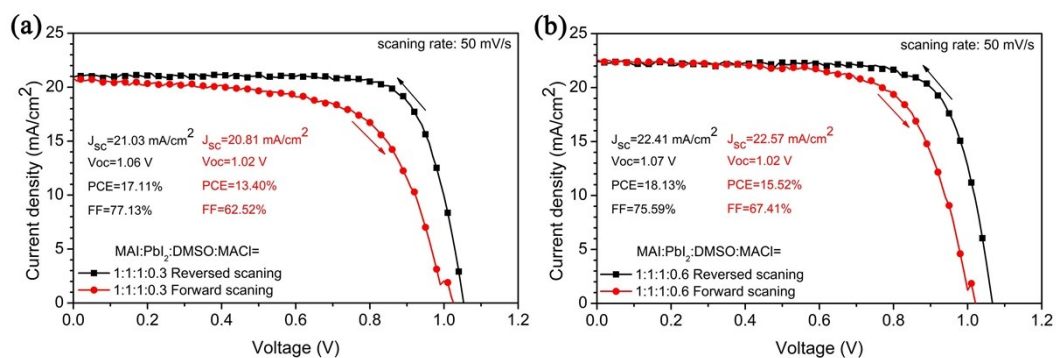


Figure S13 the forward scanning and reverse scanning of J-V curves of PSCs prepared colloidal perovskite precursor with different amount of MACl, 1:1:1:0.3 (a); 1:1:1:0.6 (b).