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Supporting Information

Scaffold-Directed Growth of Metal Halide Perovskite Hopper Crystals

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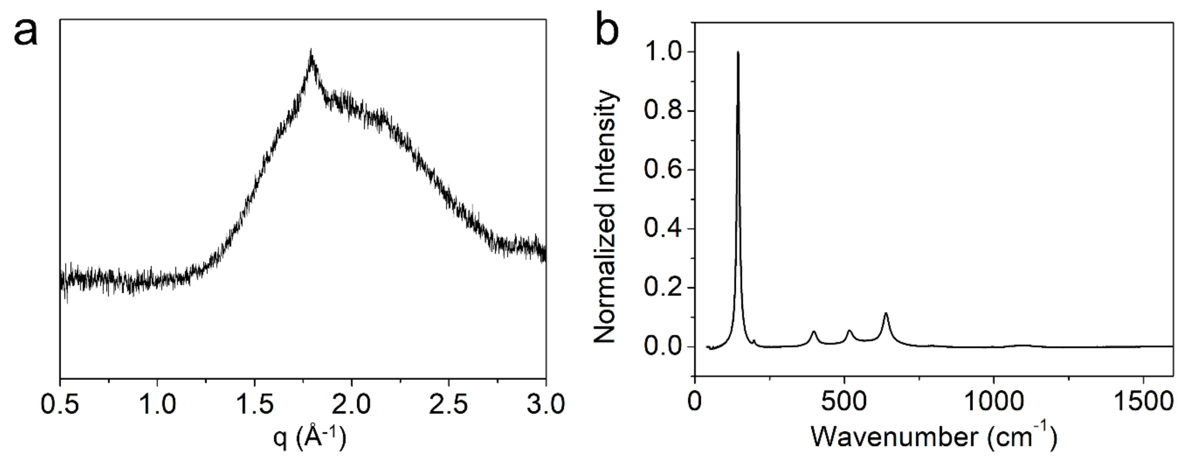


Figure S1. a) XRD pattern of a TiO_2 colloidal monolayer. The sharp peak at 1.79 $Å^{-1}$ corresponds to the 101 reflection of anatase TiO_2.[1] The broad peak is associated with diffuse scattering from the underlying glass substrate.[2] b) Raman spectrum of a TiO_2 colloidal monolayer. Peaks at 144, 400, 515, 641 cm^{-1} match the anatase phase.[1]

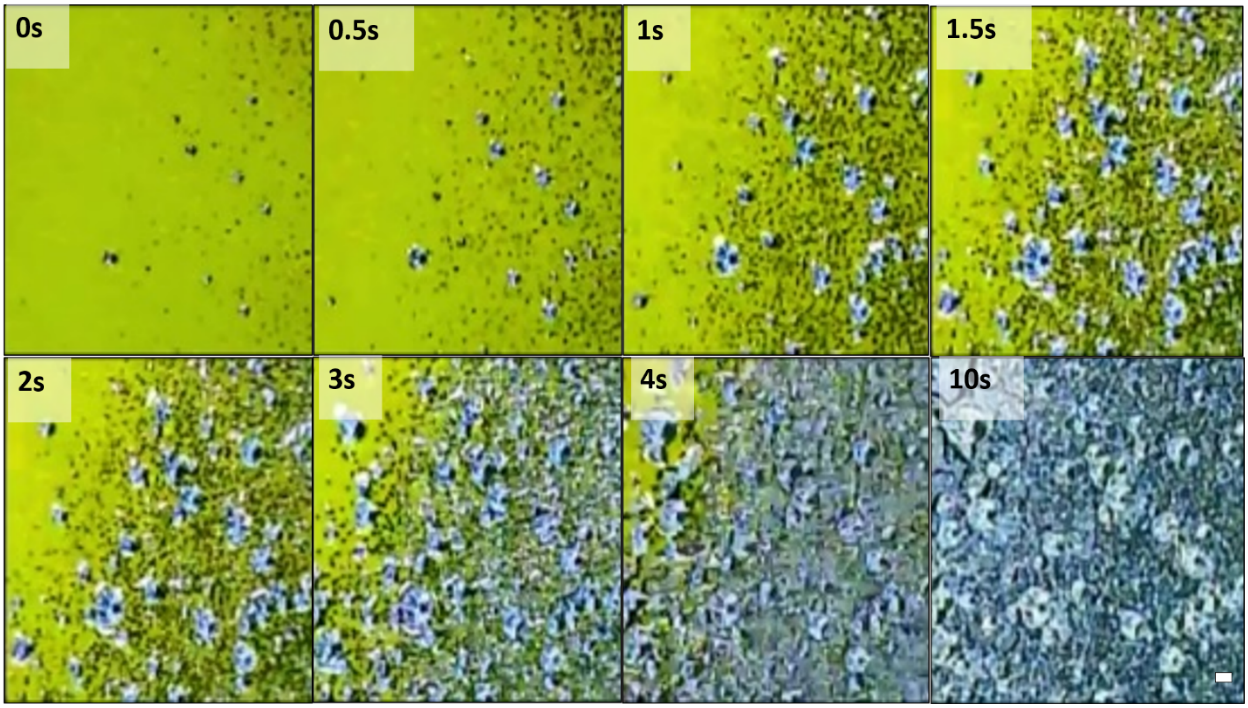


Figure S2. *In situ* optical micrographs of a $FA_{0.5}MA_{0.5}PbI_3$ film drying. Solution appears as green-yellow while crystals acquire a blue color. t = 0 s corresponds to the time at which crystal nuclei were first observed. Flower stem growth was observed first. Subsequently, flower petals began to crystallize starting as early as t = 0.5 s. At later times, a continuous perovskite film crystallized around the flowers, with crystallization completing around t = 10 s. Scale bar = 100 µm.

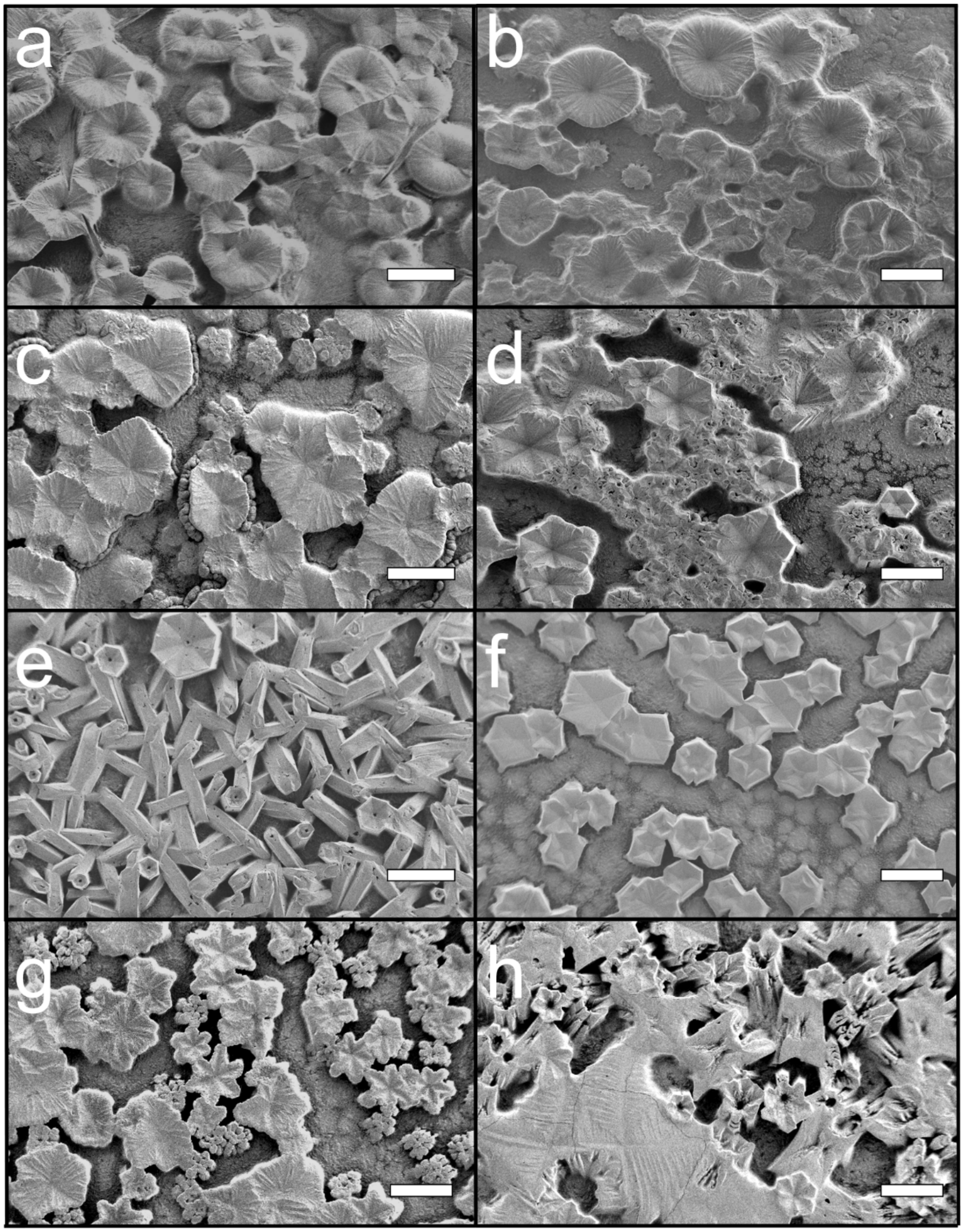
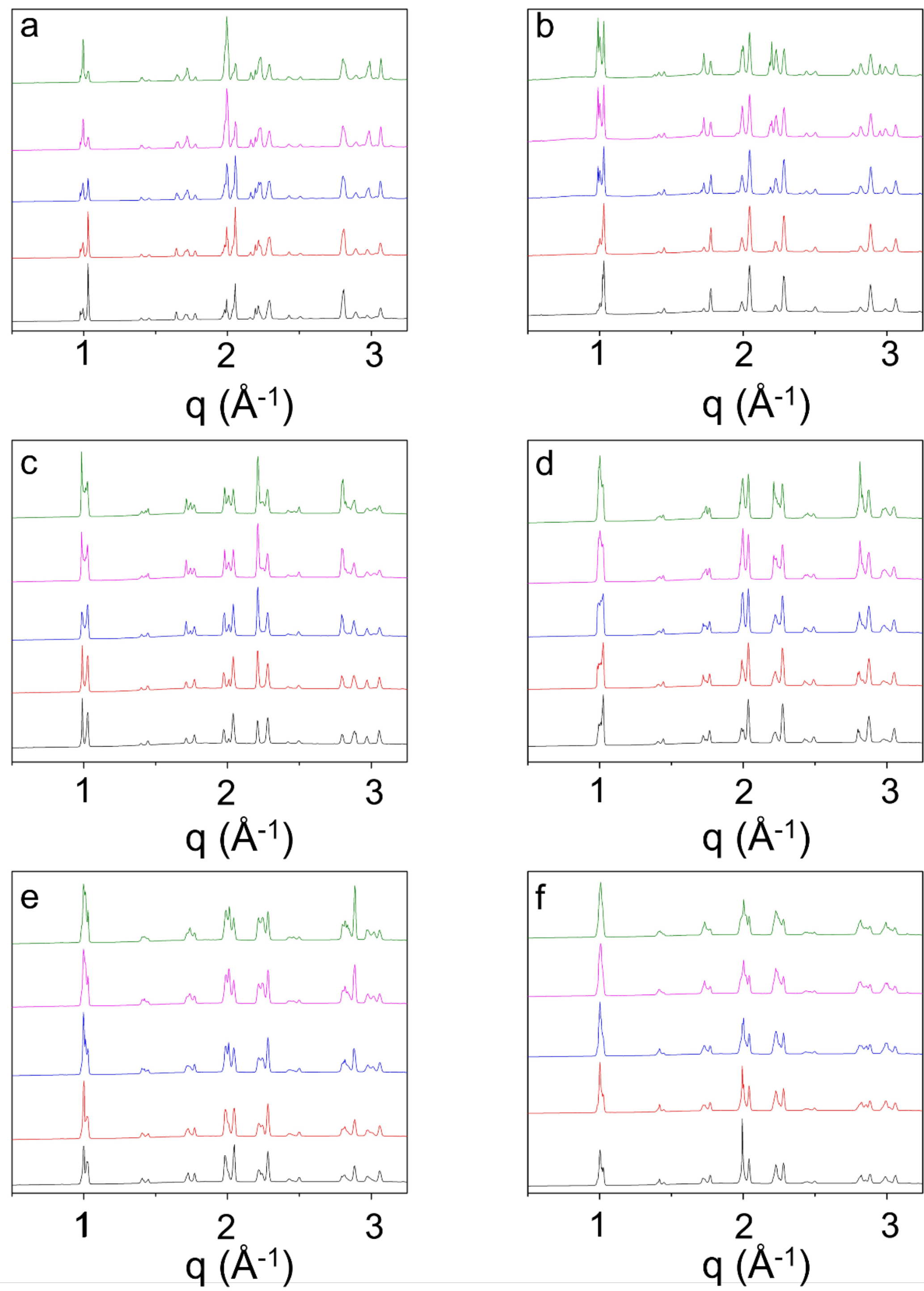


Figure S3. Low magnification SEM images of a) $MAPbI_3$; b) $FA_{0.5}MA_{0.5}PbI_3$; c) $FA_{0.75}MA_{0.25}PbI_3$; d) $FA_{0.8}MA_{0.2}PbI_3$; e) $FAPbI_3$; f) $MAPb_{0.8}I_{2.6}$; g) $FA_{0.75}MA_{0.25}Pb_{0.8}I_{2.6}$; h) $FAPb_{0.8}I_{2.6}$ crystals. Scale bar = 100 µm.


a
b
c
d
e
f
1
2
3
q (Å-1)

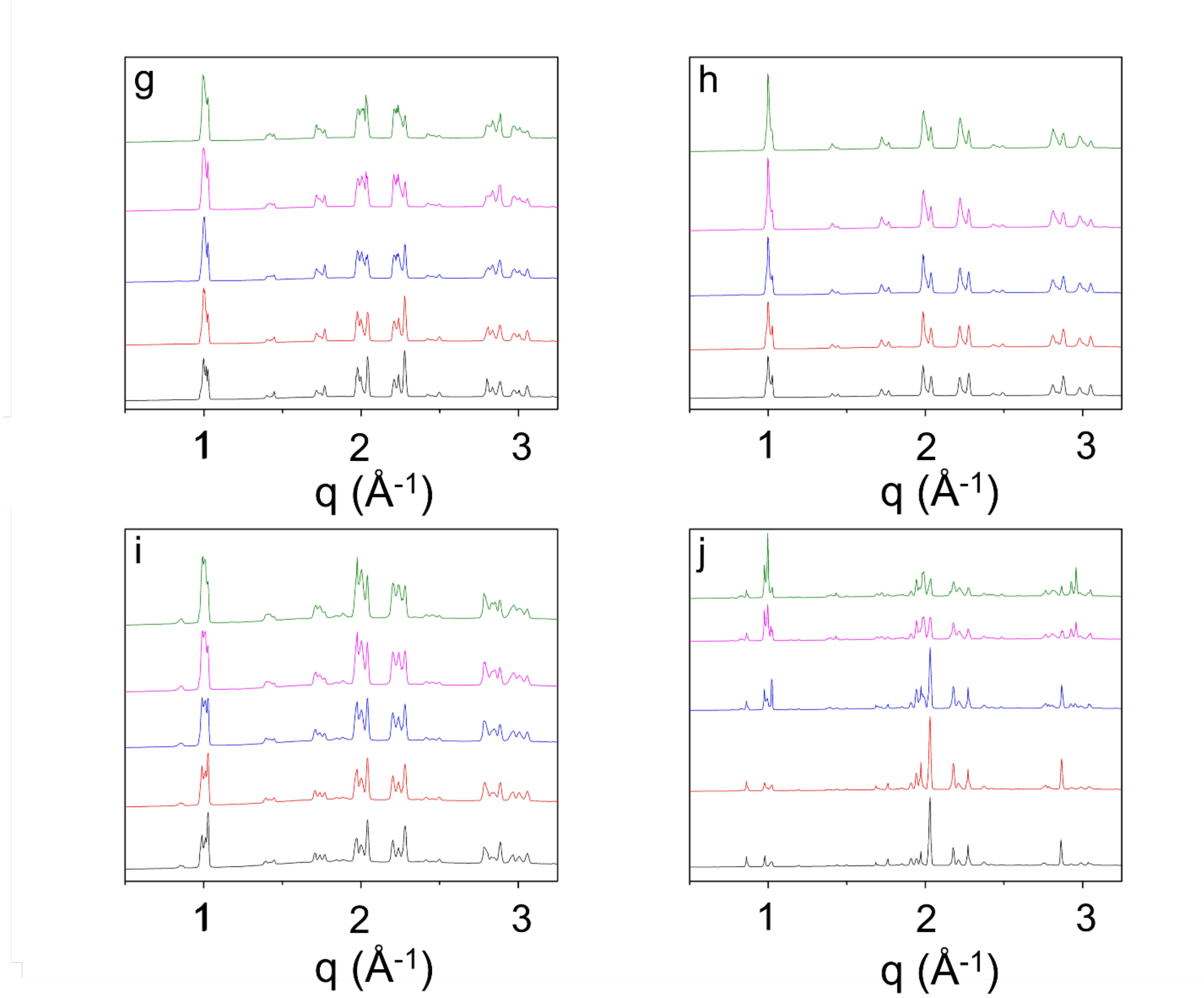


Figure S4. GIWAXS diffraction patterns at different incident angles for the following perovskite compositions: a) $MAPbI_3$, b) $FA_{0.2}MA_{0.8}PbI_3$, c) $FA_{0.4}MA_{0.6}PbI_3$, d) $FA_{0.5}MA_{0.5}PbI_3$, e) $FA_{0.6}MA_{0.4}PbI_3$, f) $FA_{0.65}MA_{0.35}PbI_3$, g) $FA_{0.7}MA_{0.3}PbI_3$, h) $FA_{0.75}MA_{0.25}PbI_3$, i) $FA_{0.8}MA_{0.2}PbI_3$, and j) $FAPbI_3$. Diffraction patterns were collected at incident angles of 0.100 ° (green), 0.120 ° (magenta), 0.150° (blue), 0.200 ° (red), 0.250 ° (black).

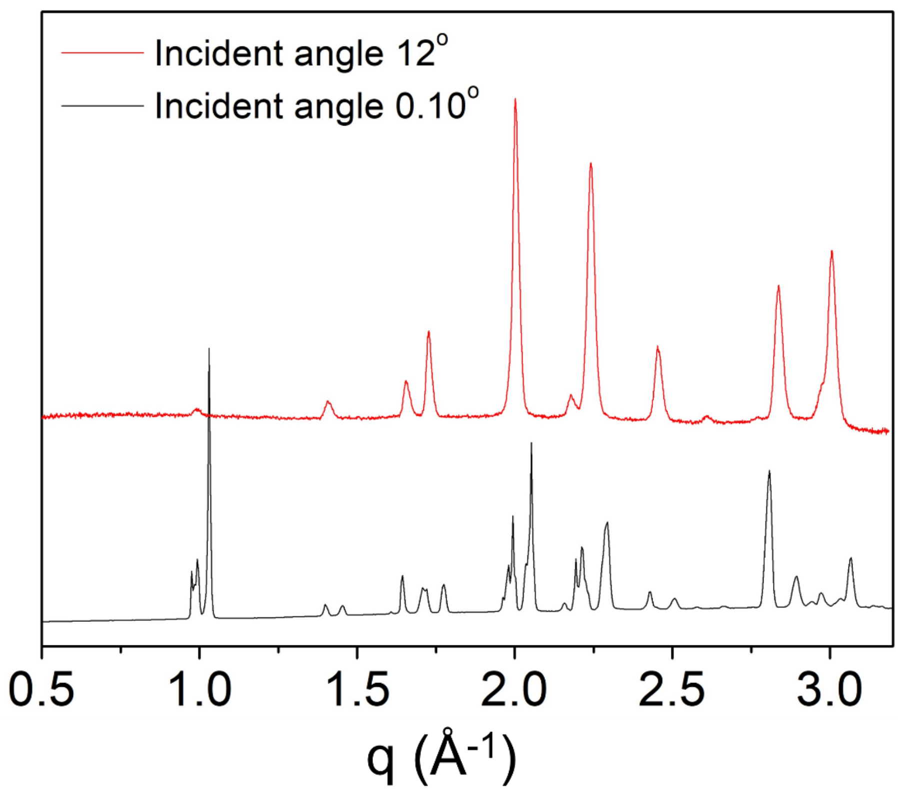


Figure S5. Comparison of XRD patterns collected on a $MAPbI_3$ flower at incident angles of 0.10° and 12°.

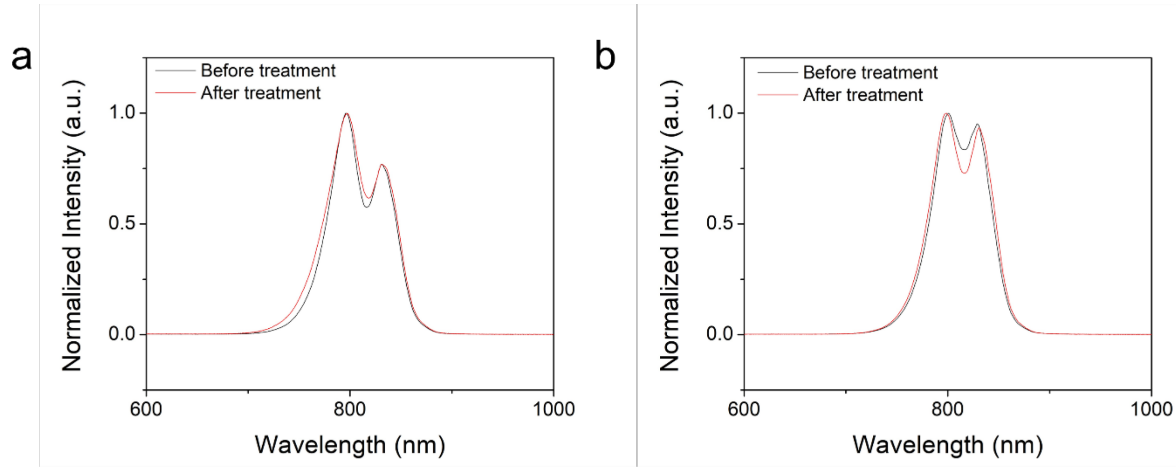


Figure S6. PL spectra (λ_{ex} = 546 nm) of $FA_{0.75}MA_{0.25}PbI_3$ flowers after surface passivation with a) thiophene and b) 10 wt% benzothiadiazole dissolved in dichlorobenzene.[3] Passivation was performed by drop casting thiophene and benzothiadiazole/dichlorobenzene solutions onto perovskite films and allowing the solutions to sit for 10 min. Excess solution was subsequently removed by spin coating the samples at 2000 rpm. After spin coating, some residual solution remained.

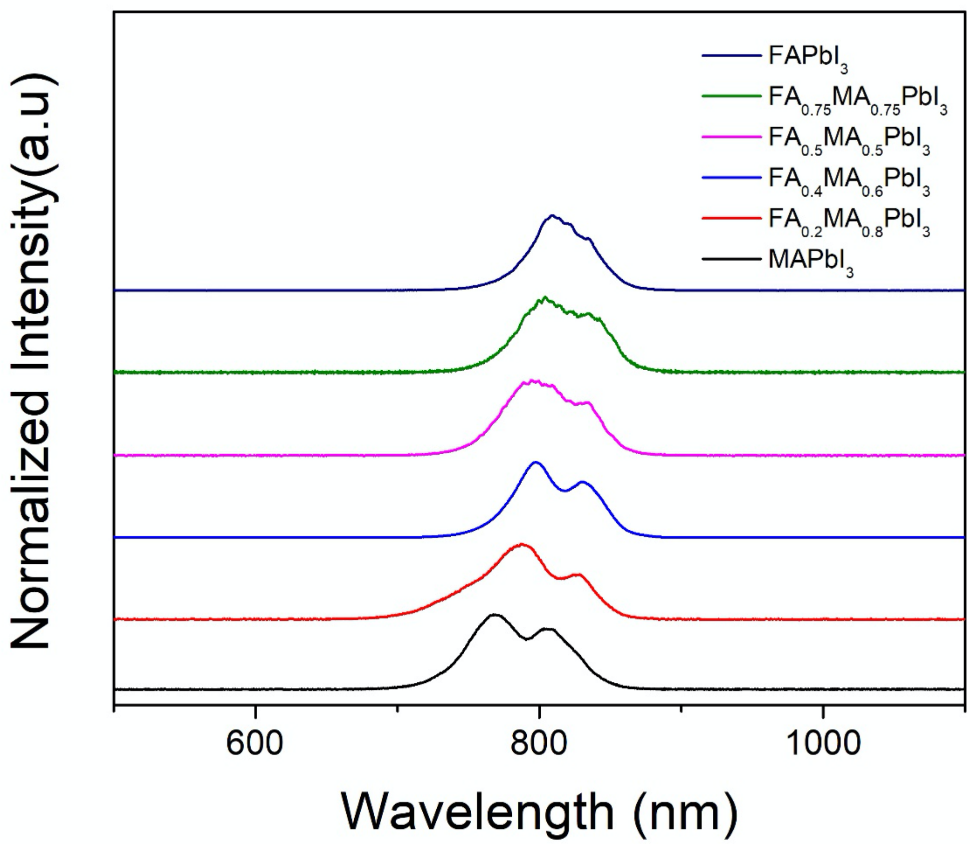


Figure S7. PL spectra of perovskite flower films (λ_{ex} = 546 nm) collected with a 73 μm^2 aperture to capture an entire flower for the following compositions: $MAPbI_3$, $FA_{0.2}MA_{0.8}PbI_3$, $FA_{0.4}MA_{0.6}PbI_3$, $FA_{0.5}MA_{0.5}PbI_3$, $FA_{0.75}MA_{0.25}PbI_3$, and $FAPbI_3$.

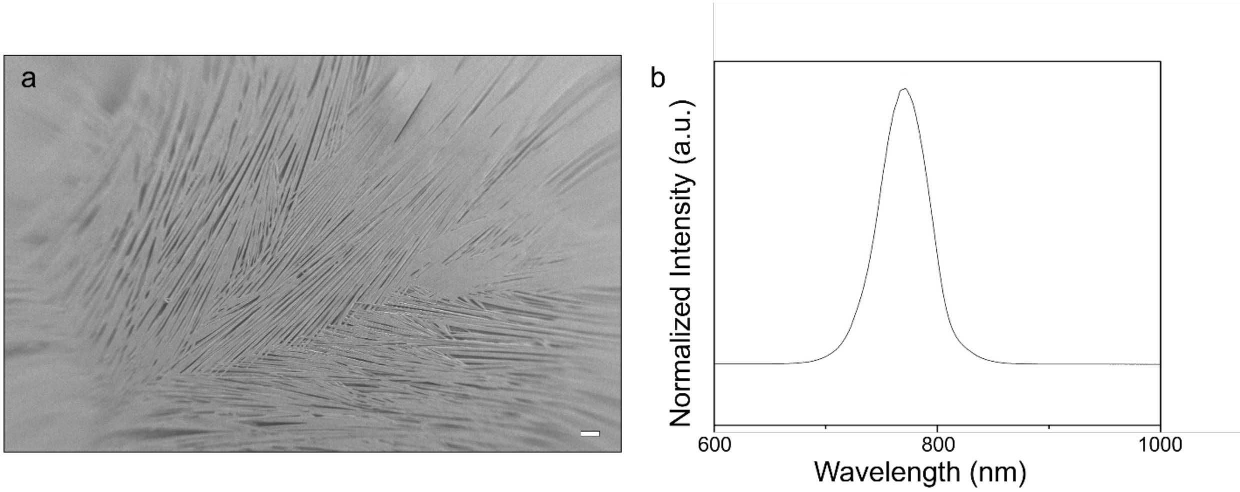


Figure S8. a) $MAPbI_3$ needle crystals formed by drop casting a precursor solution onto a TiO_2 colloidal monolayer at 20 ℃. Scale bar = 100 μm. b) PL spectrum collected on the film (λ_{ex} = 546 nm).

References

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