Supporting Information

Meso-Superstructured Perovskite Solar Cells: Revealing the Role of the Mesoporous Layer

Daniel Ramirez,[a] Kelly Schutt,[b] Juan Felipe Montoya,[a] Santiago Mesa,[a] JongChul Lim,[b] Henry J. Snaith,[b] and Franklin Jaramillo,[a*]

a. Centro de Investigación, Innovación y Desarrollo de Materiales – CIDEMAT, Facultad de Ingeniería, Universidad de Antioquia UdeA, Calle 70 No. 52-21, Medellín, Colombia.

b. Department of Physics, University of Oxford, Clarendon Laboratory, Oxford, United Kingdom.

Details of KPFM

For the case of the KPFM, in order to obtain a thin layer of perovskite capping layer, the perovskite precursor solution was diluted and deposited by spin coating at the same conditions that were used for the device fabrication. This lead to a perovskite capping layer of around 50 nm on top of the mesoporous Al_2O_3. Then, surface potential (KPFM) and surface photovoltage (SPV) measurements where performed employing a MFP-3D AFM (Asylum Research) using dual-pass technique. Topography is mapped using tapping mode AFM during the first pass, which is then traced at a set lift height above the surface performing the surface potential measurement. During the second pass of KPFM, the mechanical drive to the cantilever is disabled and an AC bias voltage (V_{AC} = 1 V) is applied to the probe at the mechanical resonance of the cantilever. The V_{AC} causes the cantilever to oscillate due to the attractive and repulsive electrostatic interaction between the probe and the sample. A feedback loop monitors and maintains a constant amplitude of the cantilever oscillations by applying a compensating V_{DC} to the probe to cancel the probe-sample electrostatic forces. The tips used were Silicon Ti-Ir coated (Asyelec-01) with nominal spring constant of 2.89 N/m and resonance frequency of 71.7 KHz. The scan size was 5 µm at a scan rate of 0.5 Hz. The probes were calibrated using evaporated gold substrate in 20% relative humidity and 25 °C. The surface potential between the AFM tip and the perovskite surface was determined at different light intensities (3 mW/cm^2), in order to determine the surface photovoltage (SPV), defined as the difference between Surface potential$_{LIGHT}$ and Surface potential$_{DARK}$.

SUPPLEMENTARY RESULTS

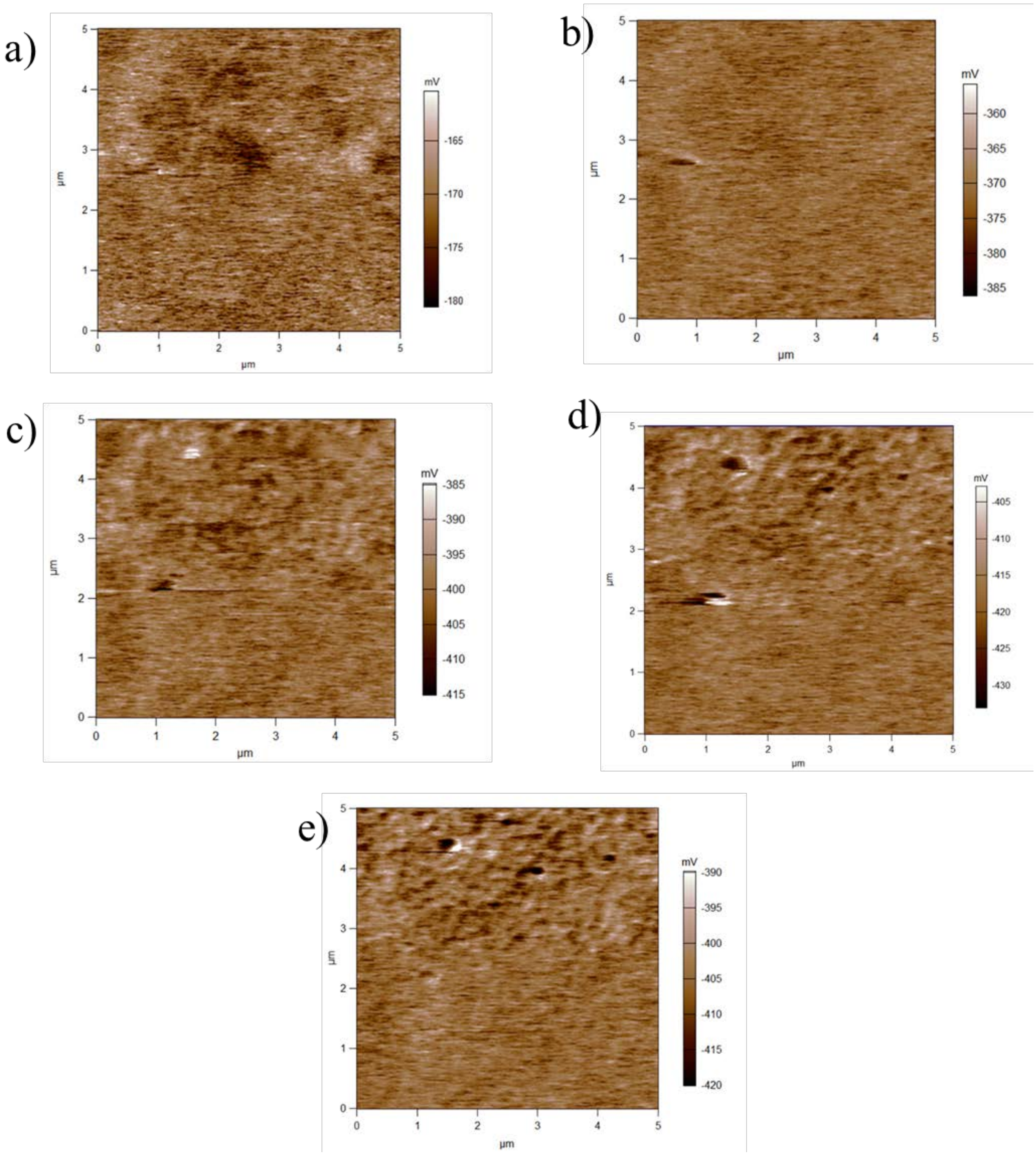


Figure S1. Illuminated KPFM of p-type/PVKT: a) 0%, b) 25%, c) 50%, d) 75%, e) 100%

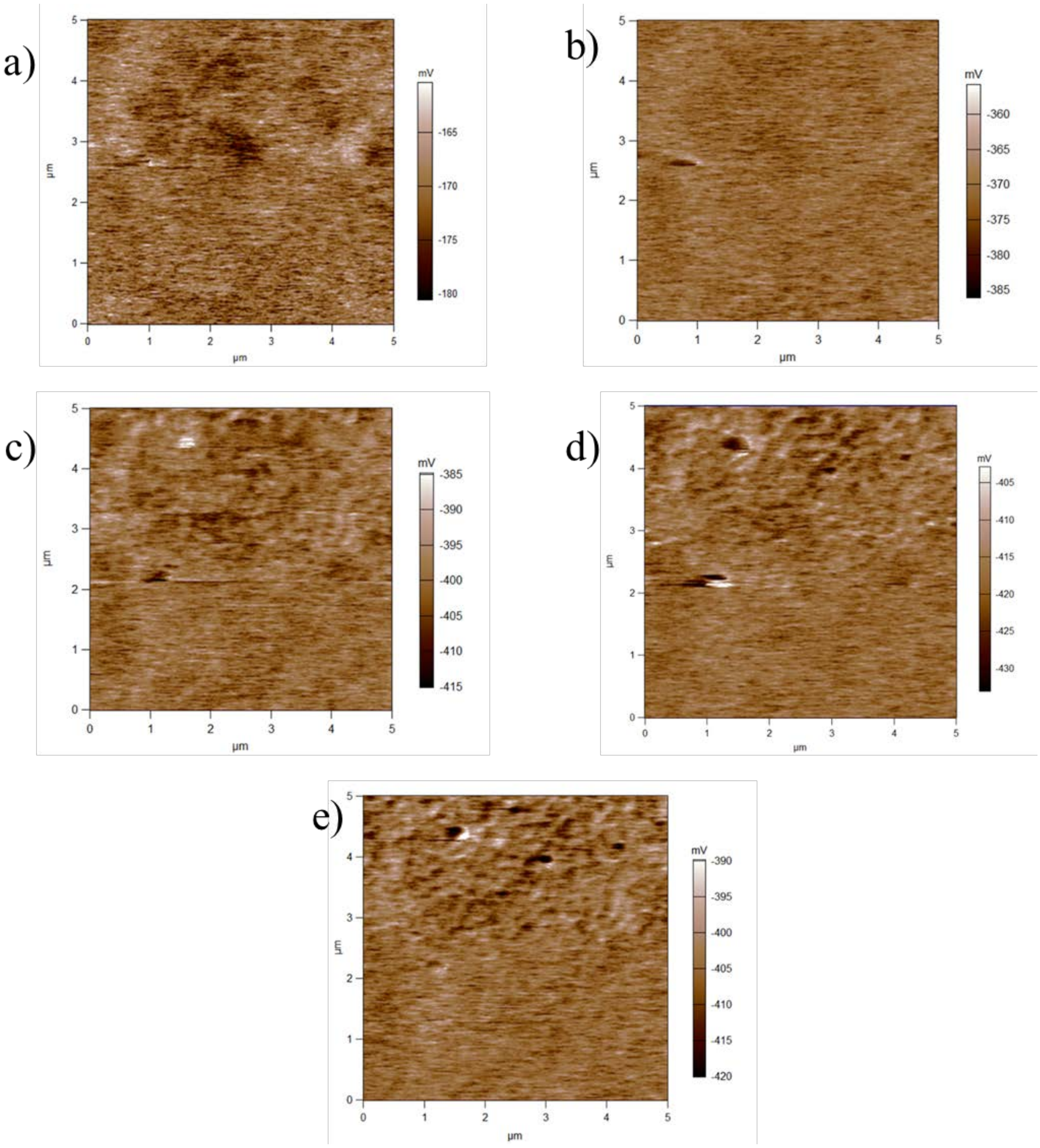


Figure S2. Illuminated KPFM of p-type/Al_2O_3/PVKT: a) 0%, b) 25%, c) 50%, d) 75%, e) 100%

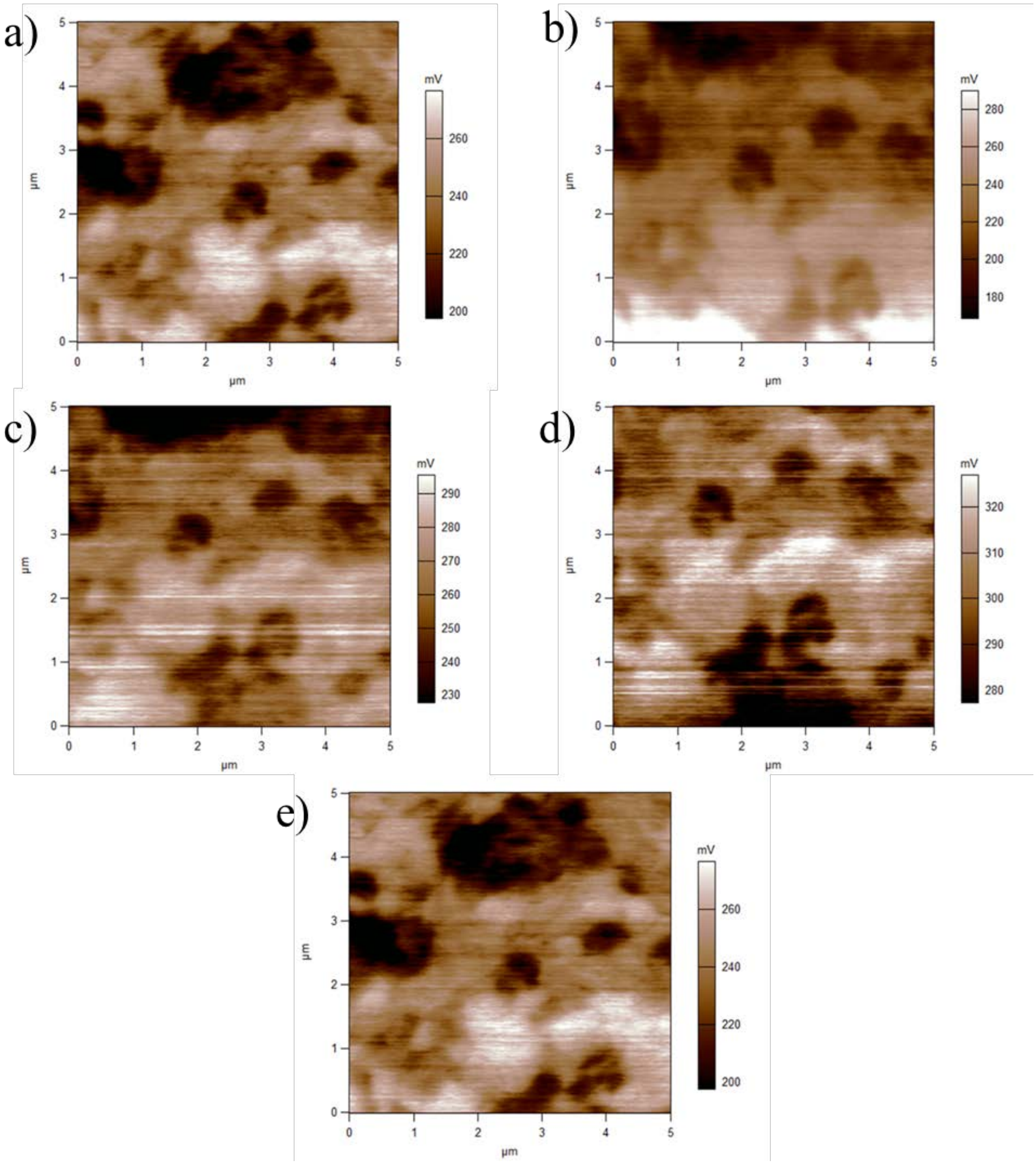


Figure S3. Illuminated KPFM of n-type/PVKT: a) 0%, b) 25%, c) 50%, d) 75%, e) 100%

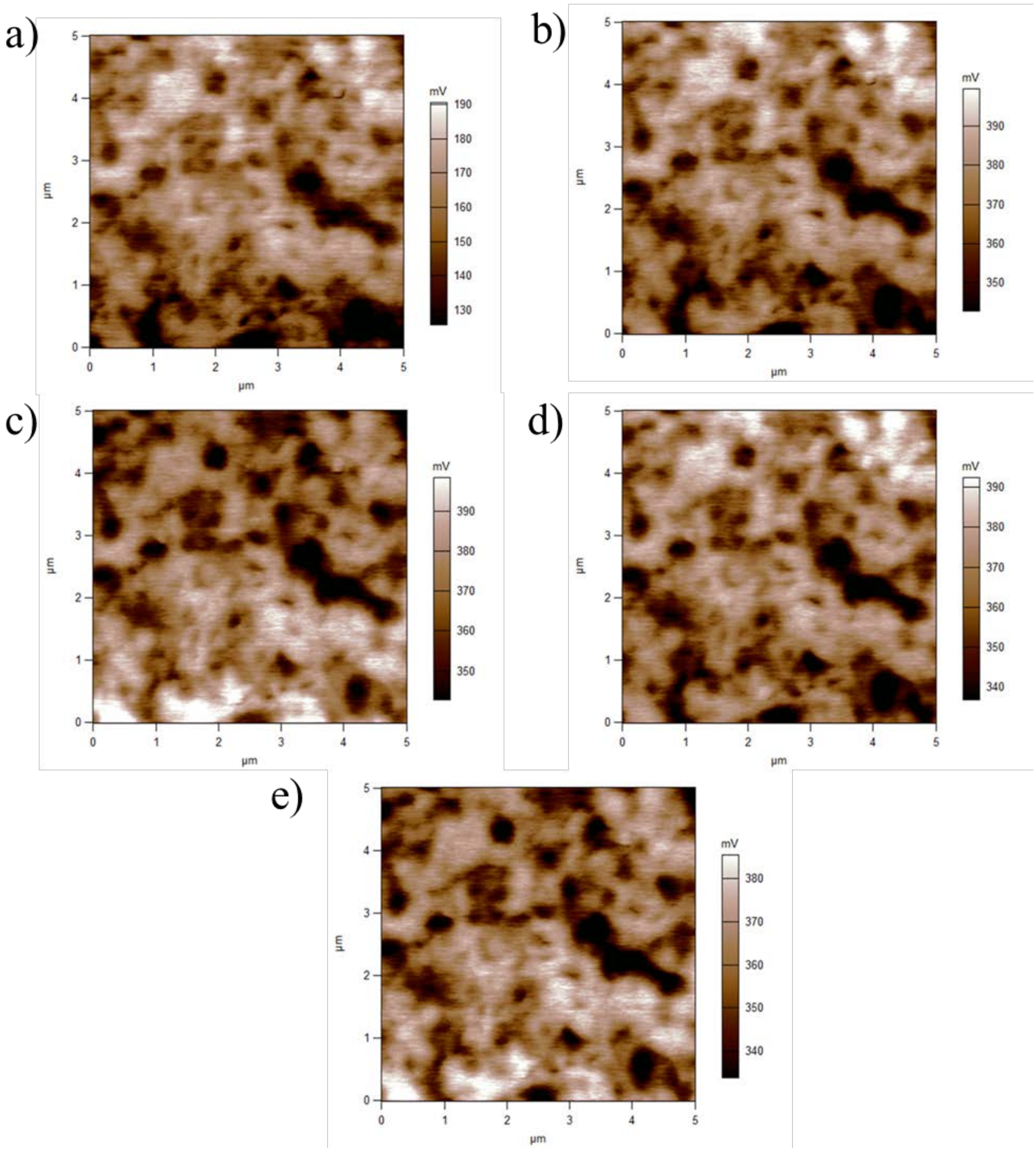


Figure S4. Illuminated KPFM of n-type/Al_2O_3/PVKT: a) 0%, b) 25%, c) 50%, d) 75%, e) 100%

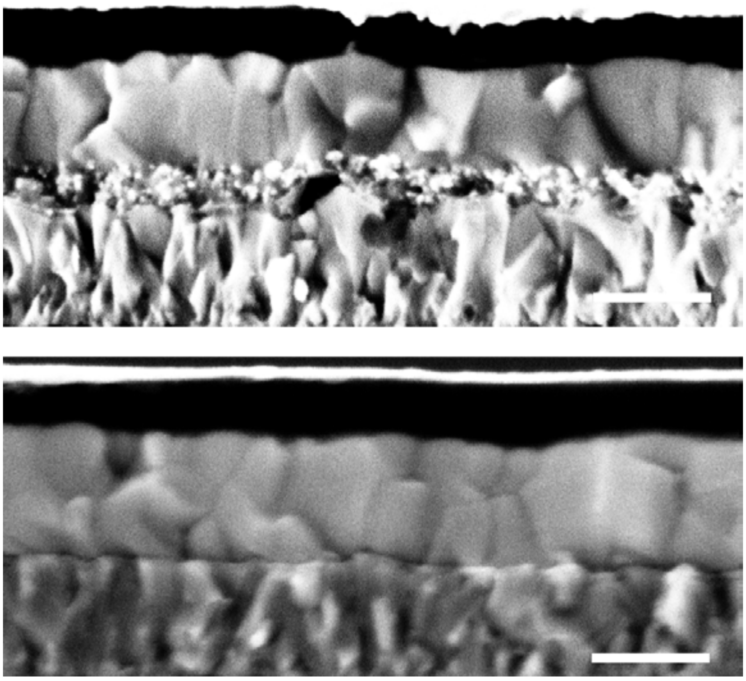


Figure S5. n-i-p Meso-superstructured (top) and planar (bottom) devices. Scale bar is 500 nm.

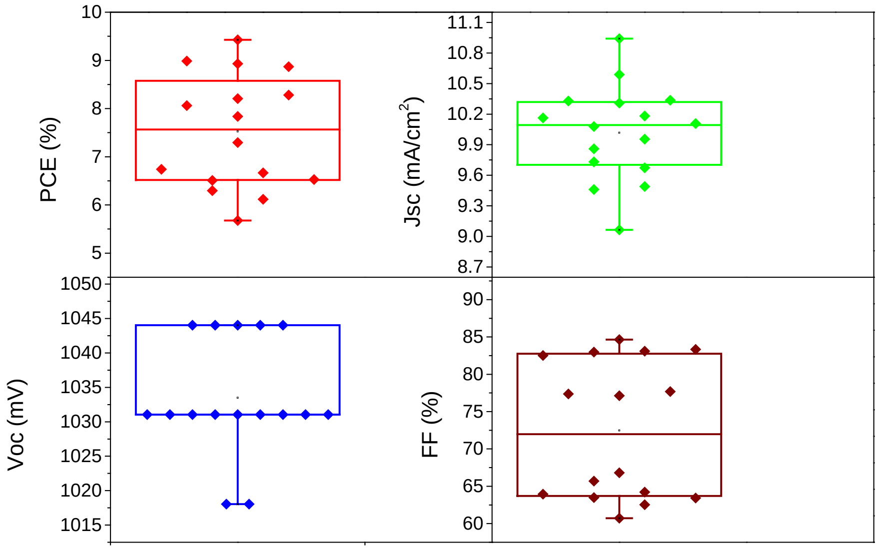


Figure S6. Photovoltaic parameters of p-i-n devices with a 400 nm Al_2O_3 mesoporous layer.

Reflectance

Increasing the thickness of the alumina layer reduced the obtained J_{sc}. In order to check the origin of such reduction, absolute reflectance measurements were performed. As shown in Figure S3, all films exhibited similar specular reflectance (even those deposited on top of a 400 nm mesoporous layer), which indicates that the loss in current is related to the electronic properties of the thicker perovskite layer instead of an inherent optical loss.

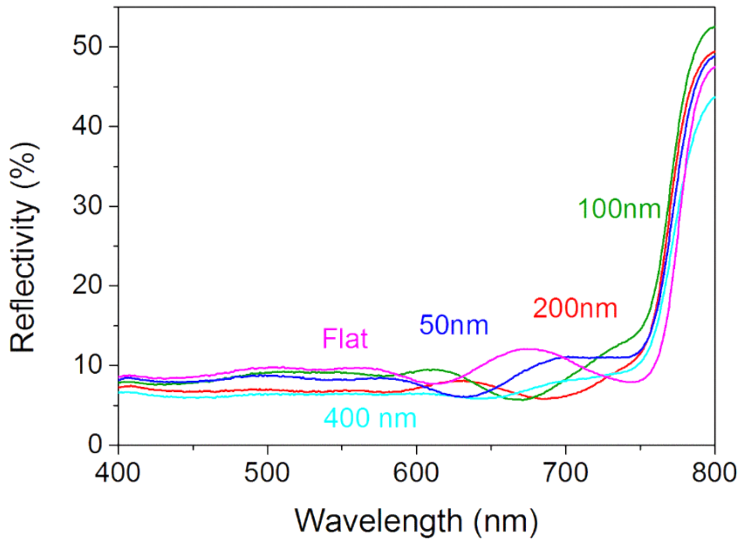
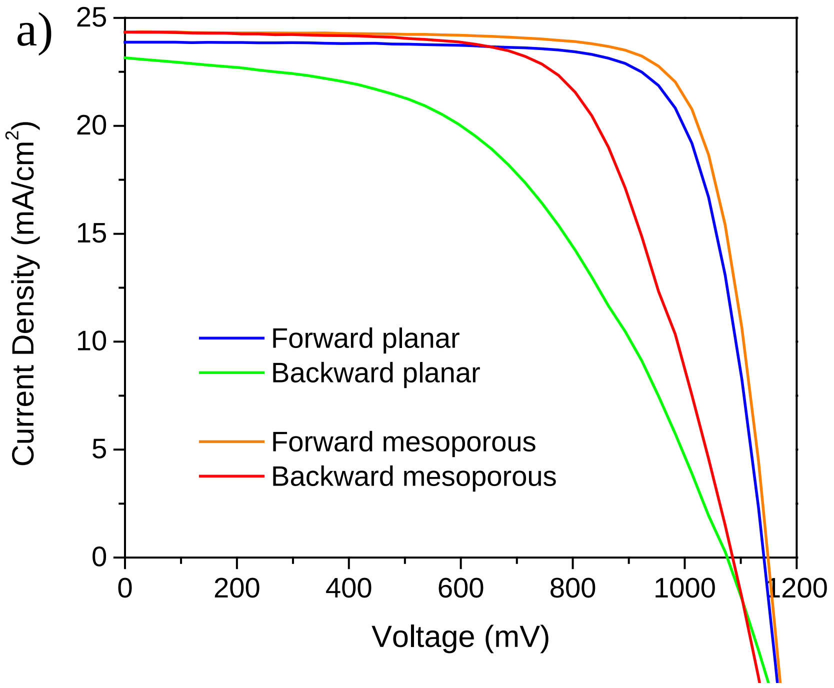


Figure S7. Specular reflectance of the planar and mesoporous p-i-n devices.

Table S1. Total thickness of the active layer including infiltrated perovskite and capping layer.

Al_2O_3 thickness (nm)	Perovskite capping layer thickess (nm)	Total thickness (nm)
0	434	434
49	386	435
97	346	444
211	351	561
399	224	623



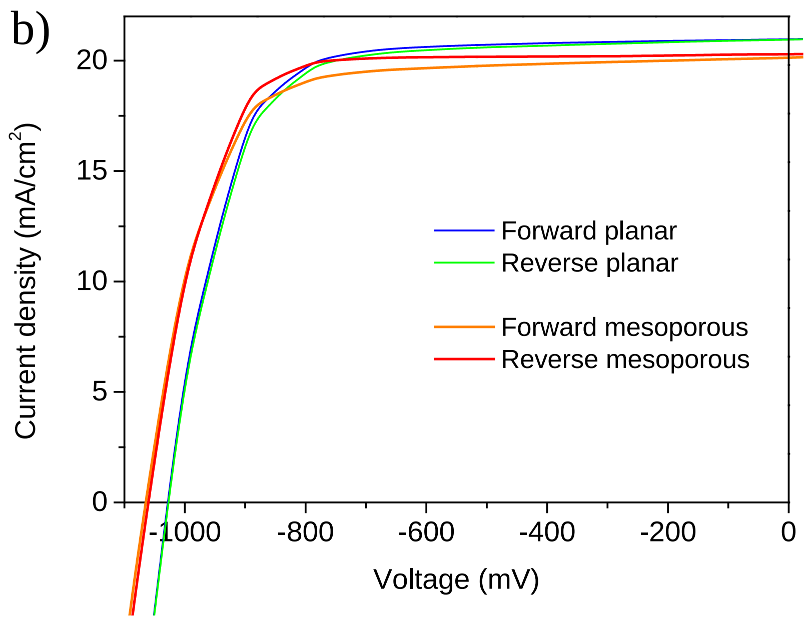


Figure S8. Comparative JV curves showing both, forward and reverse scans for a) n-i-p and b) p-i-n devices.

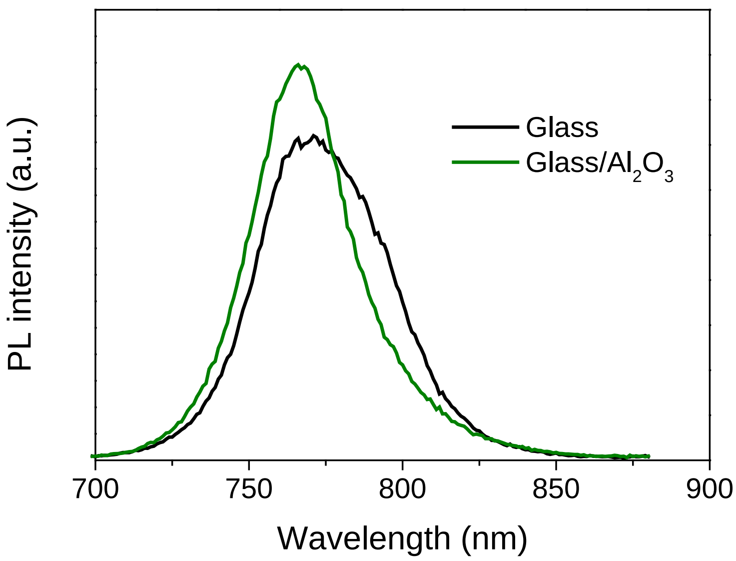


Figure S9. Comparative steady state PL of a planar and meso-superstructured perovskite deposited on top of glass.

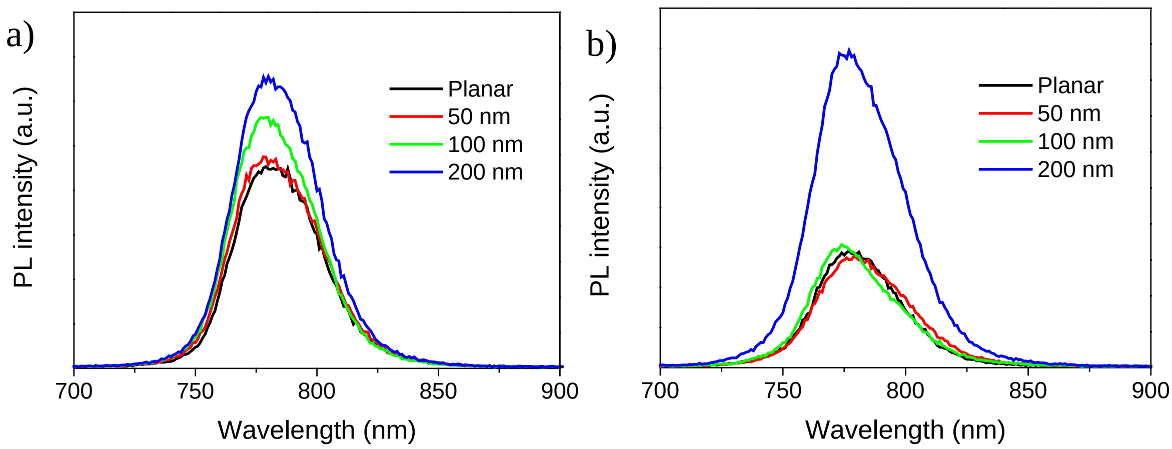


Figure S9. Al_2O_3 Thickness dependent state PL of meso-superstructured perovskites deposited on top of a) SnO_2 and b) NiO_x.