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Photoluminescence quantum yield exceeding 80% in low dimensional perovskite thin-films by passivation control

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Materials preparation

Glass substrates with an indium-doped tin-oxide (ITO) coating were subsequently cleaned with a detergent solution, Millipore water, isopropyl alcohol, and then transferred to a UV-ozone chamber for 20 minutes of treatment. Hence they were coated with a 70 nm thick poly(3,4-ethylenedioxythiophene) doped with poly(styrenesulfonate) (PEDOT:PSS, Clevios PVP AI 4083 from Heraeus) film in air, and annealed at 150 ºC for 15 minutes. The substrates were then transferred to a nitrogen filled glovebox for the perovskite deposition. The perovskites precursor solution was prepared by dissolving BABr (Dyesol), MABr (Dyesol) and $PbBr_2$ (Sigma Aldrich, 99.999% trace metals basis) in dimethyl sulfoxide (DMSO) solution with 200 mg/mL concentration. The solution was stirred at 60 ºC overnight and was filtered using a PTFE syringe filter (0.22 μm) before deposition. We prepared different quasi-2D stoichiometries varying the amount of BABr, MABr and $PbBr_2$ in dimethyl sulfoxide (DMSO) solutions, maintaining the overall concentration at 200 mg/mL. A table with the exact composition of the precursors is reported below.

$BABr:MABr:PbBr_2$ molar ratio	In 1 mL	BABr	MABr	$PbBr_2$
2:4:5	mass (mg)	23.8	34.6	141.6
	mole (mol)	0.154	0.309	0.386
3:3:5	mass (mg)	35.1	25.5	139.4
	mole (mol)	0.228	0.228	0.380
4:2:5	mass (mg)	46.1	16.7	137.2
	mole (mol)	0.299	0.150	0.374

The deposition was performed via a consecutive two-steps spin coating process at 1000 and 3000 rpm for 5 and 60 seconds, respectively. When solvent engineering was used, during the second spin-coating step 300 μL of chloroform were dripped onto the substrate, and the resulting films were annealed at 90 ºC for 5 minutes. In order to improve the PEDOT:PSS stability and its adhesion to the metal oxide substrate, we added to its suspensions a small amounts (1 v/v %) of 3-glycidoxypropyltrimethoxysilane (GOPS) as a cross-linking agent. The cross-linking of PEDOT:PSS with GOPS takes place at the same temperature we used for the annealing of PEDOT:PSS neat films (150 ºC).

Characterization and device preparation

The photoluminescence (PL) characteristics were studied using a Xe lamp coupled to a monochromator as the excitation source and an integrated sphere coupled to a spectrometer (Hamamatsu C9920-02 with a Hamamatsu PMA-11 optical detector) in order to quantitatively determine the PLQY. UV-visible absorption spectra of the films were collected using a fiber-optics based Avantes Avaspec2048 spectrophotometer. TRPL decays were measured using a

Compact fluorescence lifetime spectrometer C11367, Quantaurus- Tau, with LED light source. Fluorescence lifetime measurement software U11487 was used to register the data. All the data of PL decay of perovskite films were acquired under an excitation wavelength of 340 nm.

TheThe AFM images were collected with a Digital Instrument Veeco Nanoscope IVa AFM microscope in tapping mode, using silicon tips with natural resonance frequency of 320 kHz and with an equivalent constant force of 42 N m^{-1}.

The electron transport (hole blocking) layer (BmPyPhB) and the top metal electrode (5 nm Ba capped with 100 nm Ag) were thermally evaporated in a high vacuum chamber with a base pressure of 10^{-6} mbar. Devices were not encapsulated and were characterized in the nitrogen filled glove box. The current density and luminance versus voltage characteristics were measured using a Keithley 2400 Source-Meter and a photodiode coupled to a Keithley 6485 pico-ammeter.

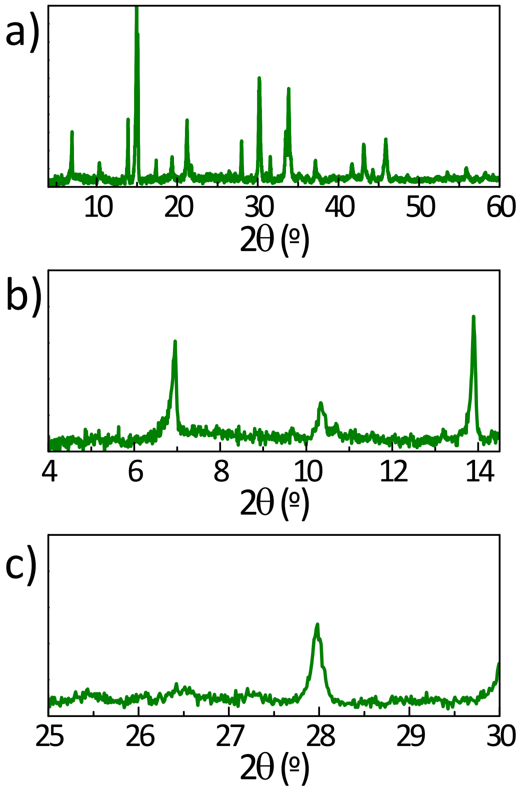

Fig. S1 X-ray diffraction patterns of the quasi-2D perovskite with molar ratio BA:MA = 3:3 a) Bulk powder diffraction and close-up views of the regions between b) 2θ = 4 − 15° and c) 2θ = 25 − 30°.

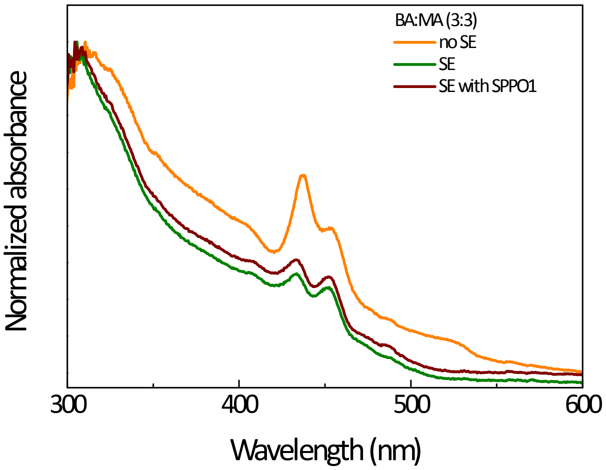

Fig. S2 Absorption spectra in thin-film of the quasi-2D perovskite with BA:MA 3:3 ratio without, with solvent engineering (SE) and with SE in the presence of SPPO1.

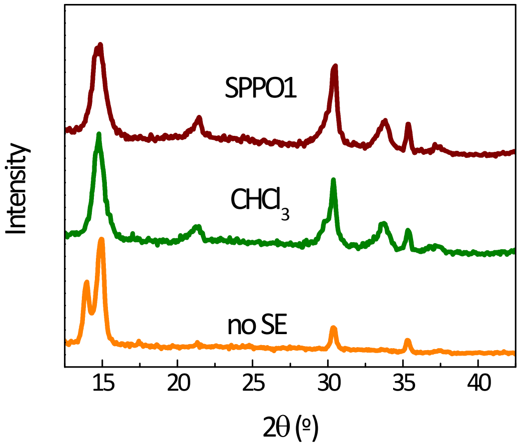

Fig. S3 Grazing incidence X-ray diffraction pattern in thin-film of the quasi-2D perovskite with BA:MA 3:3 ratio without, with solvent engineering (SE) and with SE in the presence of SPPO1.

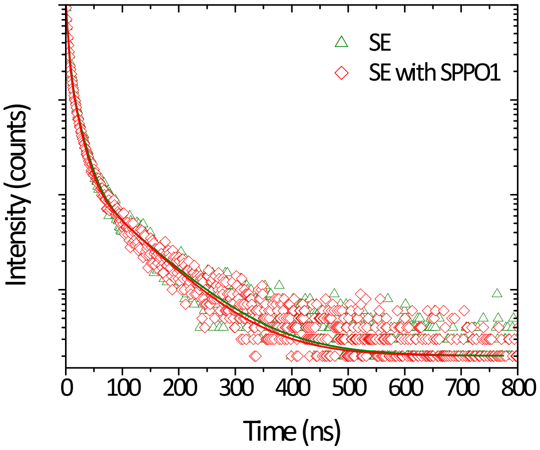

Fig. S4 Electroluminescence spectrum of the LED using quasi-2D perovskite as the emissive layer.

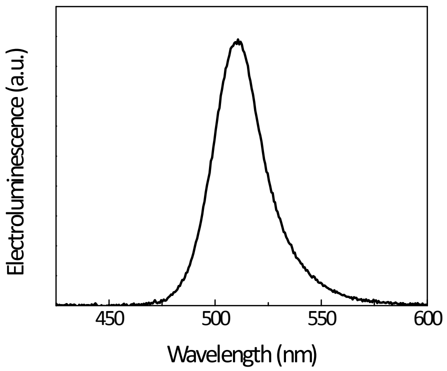

Fig. S5 Electroluminescence spectrum of the LED using quasi-2D perovskite as the emissive layer.