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Efficient photovoltaic and electroluminescent perovskite devices

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Supporting Information

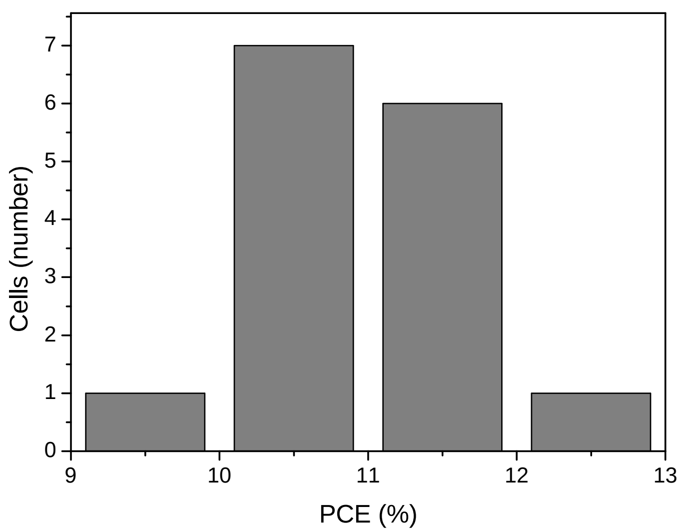

Figure S1. Distribution of the power conversion efficiency (PCE) of the different cells measured as solar cells.

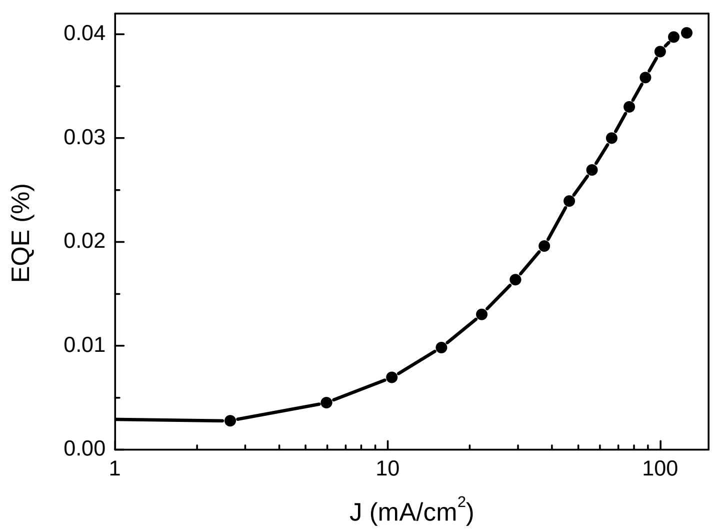

Figure S2. External quantum efficiency (EQE) versus current density for a dc- (symbols) and a pulsed current- driven (line and symbols) device.

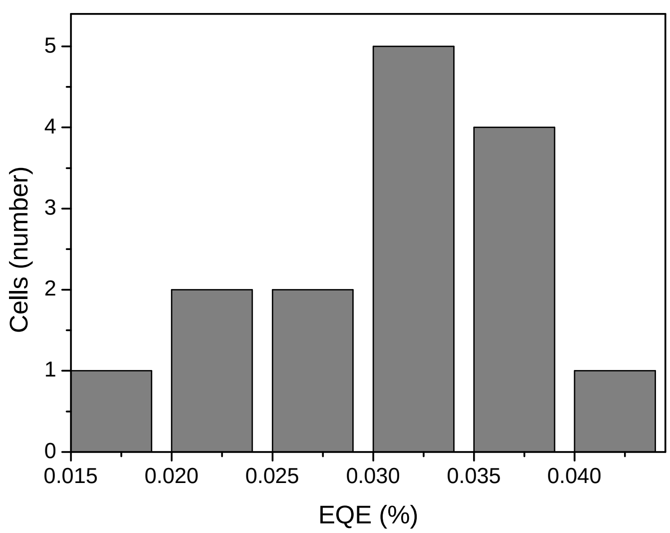

Figure S3. Distribution of the external quantum efficiency (EQE) of the different cells measured as light-emitting devices.

Materials and methods

Photolithographically patterned ITO covered glass substrates were purchased from Naranjo-Substrates. Aqueous dispersions of poly(3,4-ethylenedioxythiophene) doped with poly(styrenesulfonate) (PEDOT:PSS, CLEVIOS PVP Al 4083) were obtained from Heraeus Holding GmbH and used as received. Poly[N,N'-bis(4-butylphenyl)-N,N'-bis(phenyl)benzidine] (poly-TPD) was purchased from ADS Dye Source. [6,6]-Phenyl-C_{61}-butyric acid methyl ester ($PCBM_{60}$, >99.5 % purity) was obtained from Solenne BV. PbI_2 was purchased from Sigma-Aldrich while CH_3NH_3I was obtained from Solaronix SA, both materials were used without any further purification.

Device preparation and characterization

PEDOT:PSS was spin coated on top of clean ITO substrates resulting in a 80 nm thick layer, annealed at 120 ºC for 15 minutes. On top of this layer a thin film of polyTPD was deposited from a chlorobenzene solution (10 mg ml^{-1}) using a meniscus coater at a coating speed of 2.5 mm s^{-1}. Subsequently the substrates were transferred to a vacuum chamber integrated into a Nitrogen filled glovebox and evacuated to a pressure of 1×10^{-6} mbar. Temperature controlled evaporation sources fitted with ceramic crucibles were employed to sublime the CH_3NH_3I and the PbI_2. Co-evaporation of the ammonium and metal iodides was performed heating the crucibles at 90 ºC and 250 ºC, respectively. The film thickness of the perovskite film was monitored using a quartz sensor in the vicinity of the substrate holder. The $PCBM_{60}$ layer was deposited using a chlorobenzene solution of 10 mg ml^{-1} in ambient conditions using a meniscus coater at a coating speed of 10 mm s^{-1}. The device was completed by the thermal evaporation of the top metal cathode (10 nm Ba followed by 100 nm Ag) under a base pressure of 2×10^{-6} mbar. The thickness of the films was determined with an Ambios XP-1 profilometer. The devices were then encapsulated with a glass cover using a UV curable epoxy sealant (Ossila E131 Encapsulation Epoxy), UV exposure 5min.

GIXRD measurements were performed at room temperature in the 2θ range 5–50º on an Empyrean PANalytical powder diffractometer, using Cu Kα1 radiation. Pawley refinements were performed using the TOPAS software and revealed an excellent fit to a one-phase model with a tetragonal cell (a = 8.80(2) Å, c = 12.57(2) Å) and space group I4/mcm. Simulation of the crystal structure of $CH_3NH_3PbI_3$ perovskite was performed using as starting model the isostructural Sn analogue (CCDC refcode: ZZZBWS02) modifying the cell parameters to those obtained in the Pawley refinement.

Characterization of the photovoltaic performances was obtained using two methods. IPCE measurements were performed using a white light halogen lamp in combination with interference filters (MiniSun simulator by ECN, the Netherlands). Current-voltage (J-V) characteristics were measured using a Keithley 2400 source measure unit with and without illumination. The light source was a 10500 solar simulator by Abet Technologies. Before each measurement, the exact light intensity was determined using a calibrated Si reference solar cell.

Current density and electroluminescence were measured by using a Keithley 2400 source meter and a photodiode coupled to the Keithley 6485 pico-amperometer calibrated. The irradiance and EQE were determined using an integrated sphere coupled to an UDT instruments

S370 Optometer. An Avantes luminance spectrometer was used to measure the electroluminescent spectrum. The pulsed current driving was achieved using a Test System designed by BoTEST (Botest OLT OLED Lifetime-Test System).

Transient luminance signals were obtained by applying a pulsed current and measuring the photocurrent generated by a photometry-precision Si-photodiode (Hamamatsu ST3368BK) coupled to a four channel Tektronix Oscilloscope (TDS2024B).