

Supporting Information

Boosting External Quantum Efficiency in Perovskite Light-Emitting Diodes by an Exciton Retrieving Layer

*Chunhong Gao,^{‡*ac} Ziyang Xiong,^{‡^a} Ziqian He,^b Yajie Dong,^{bcd} Fuxing Yu,^a Xingjuan Ma,^a Yue Zhang,^a Dongying Zhou,^e Zuhong Xiong^{*a}*

^aSchool of Physical Science and Technology, MOE Key Laboratory on Luminescence and Real Time Analysis, Southwest University, Chongqing, 400715, China
E-mail: gch0122@swu.edu.cn; zhxiong@swu.edu.cn

^bCollege of Optics and Photonics, University of Central Florida, Orlando, FL, 32816, USA

^cNanoscience Technology Center, University of Central Florida, Orlando, FL 32826, USA

^dDepartment of Materials Science & Engineering, University of Central Florida, Orlando, FL, 32816, USA

^eSoochow Institute for Energy and Materials InnovationS (SIEMIS), College of Physics, Optoelectronics and Energy, Soochow University, Suzhou, 215006, China

Experimental Section

Materials: Cesium bromide (CsBr, > 99.999%) was purchased from Alfa-Aesar. Lead bromide (PbBr₂, > 99.99%), dimethyl sulfoxide (DMSO, 99.9%) and chlorobenzene (99.8%), were purchased from Sigma-Aldrich. Poly(9-vinylcarbazole) (PVK, M.w.=135600) is purchased from Acros. Phenethylammonium bromide (PEABr, > 99.5%) is purchased from Xi'an Polymer Light Technology Corp.. Firpic, TmPyPB and 8-Hydroxyquinolinolato-lithium (Liq) were purchased from Suzhou Fangsheng Light Technology Corp. All these materials were purchased from commercial sources and were used as received.

Device Fabrication: The CsPbBr₃ precursor solution for the 3D CsPbBr₃ PeLEDs was prepared by dissolving CsBr and PbBr₂ with 1.2:1 molar ratio in DMSO to obtain a concentration of ~10 wt%, and stirred at room temperature over 12 hrs. The 0.2 mol/L CsPbBr₃:PEABr precursor solution for the quasi-2D PeLEDs was prepared by dissolving CsBr, PbBr₂ and PEABr with 1:1:0.4 molar ratio in DMSO. The indium tin oxide (ITO) glass substrates (15 Ω square⁻¹) were cleaned successively by ethanol, acetone, and detergent water in an ultrasonic bath, and then dried in an oven at 120 °C for 10 min. Before device fabrication, the ITO substrates were treated with ultraviolet (UV) ozone plasma (120 W) for 5 min. To fabricate 3D PeLEDs, the PEDOT:PSS solution was spin-coated (4500 rpm, 40 s) onto the ITO substrates and baked at 120 °C for 20 min under ambient condition. After the substrates cooled down, the CsPbBr₃ precursor solution was spin-coated (4000 rpm, 60 s) onto the samples in a glovebox (H₂O and O₂ ≤ 0.1 ppm). For quasi-2D PeLEDs, the PVK solution (7 mg/ml in chlorobenzene) was spin-coated (5000 rpm, 30 s) onto the ITO substrates and baked at 150 °C for 15 min in glovebox. Then, the CsPbBr₃:PEABr precursor solution was spin-coated (4000 rpm, 60 s) onto the samples and baked at 70 °C for 5 min. Then, the samples (both 3D and quasi-2D ones) were transferred to the glovebox transfer chamber with ~10⁻¹ bar for 20 min to remove residual DMSO solvent. Subsequently, the samples were transferred into a deposition chamber

(Shenyang Vacuum Technology Research Institute) connected to the glovebox where the entire process was not exposed to atmosphere. Functional layers of TmPyPB:FIrpic, TmPyPB, Liq, Al were deposited sequentially by the thermal evaporator at pressure of $\sim 5 \times 10^{-4}$ Pa. The TmPyPB:FIrpic composite layer was prepared by co-evaporating FIrpic and TmPyPB from two individual pockets. After evaporations, the devices were encapsulated in the glovebox. The device active area was $\sim 2 \times 3$ mm².

Characterizations: The current density-luminance-voltage (*J-L-V*) characteristics of PeLEDs were measured by Keithley2400 source Meter and silicon photodiode (calibrated by PR670 Spectra Colorimeter). The EL spectra and photoluminescence (PL) spectra are collected by PR670 Spectra Colorimeter and Hitachi F-2500 spectrometer, respectively. The absorption spectra of the films were measured by UV-2600 spectrophotometer (Shimadzu Corp.). All device measurements were carried out at room temperature in ambient air.

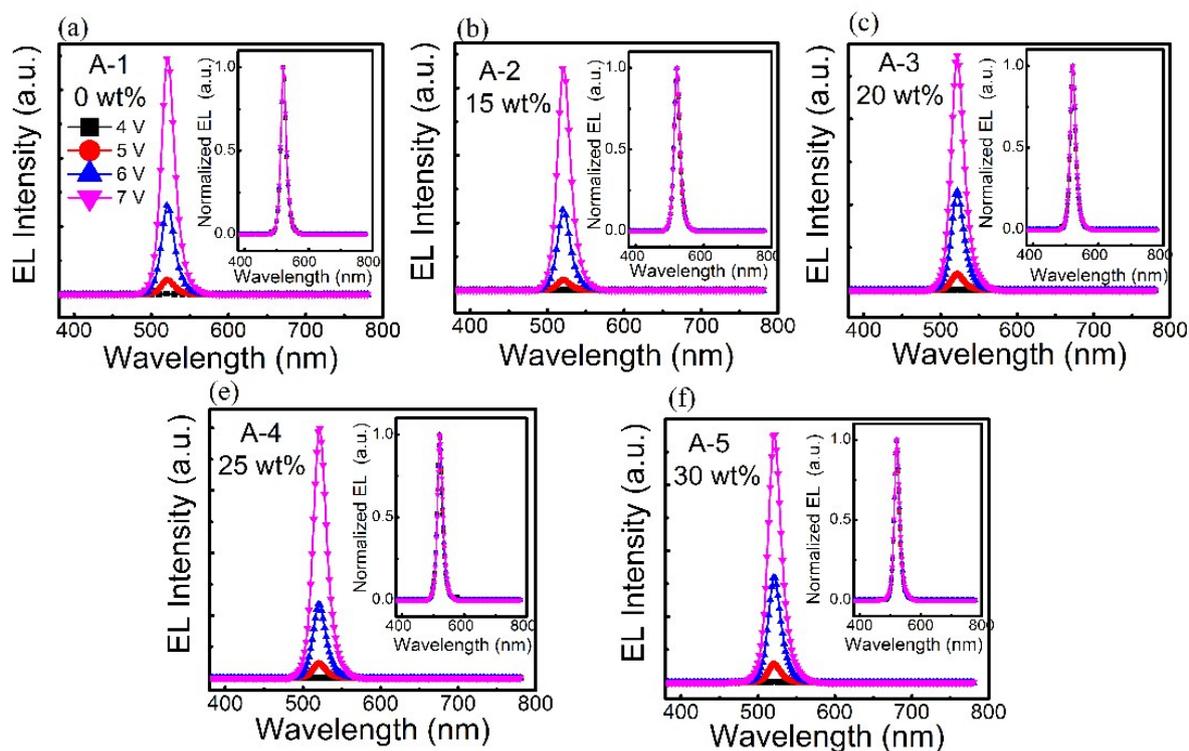


Figure S1. EL spectra of the 3D PeLEDs based on the different exciton retrieving layers (ERLs) of TmPyPB:FIrpic at different weight ratio of 0 wt%, 15 wt%, 20 wt%, 25 wt% and 30 wt% in devices A-1, A-2, A-3, A-4, and A-5, respectively, under different driving voltages of 4, 5, 6 and 7 V. The insets show the normalized EL spectra of these PeLEDs under different driving voltages of 4, 5, 6 and 7 V (right inset). (a) Device A-1 with 0 wt% TmPyPB:FIrpic, (b) Device A-2 with 15 wt% TmPyPB:FIrpic, (c) Device A-3 with 20 wt% TmPyPB:FIrpic, (d) Device A-4 with 25 wt% TmPyPB:FIrpic, (e) Device A-5 with 30 wt% TmPyPB:FIrpic.

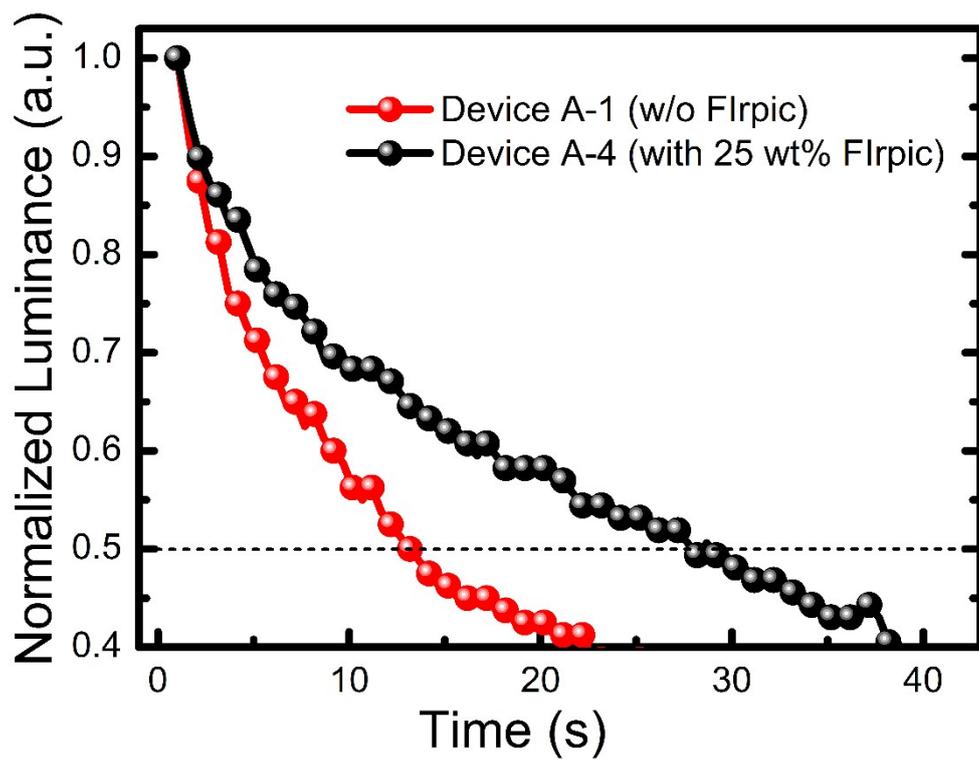


Figure S2. The stability of PeLEDs without ERL (Device A-1) and with ERL of TmPyPB:FIrpic 25 wt% (Device A-4).

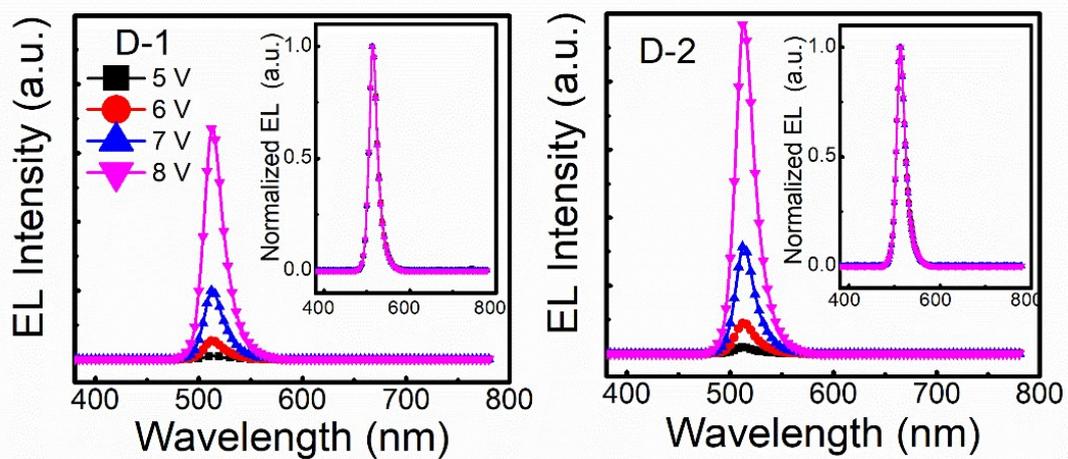


Figure S3. EL spectra of (a) Device D-1 with 0 wt% TmPyPB:FIrpic and (b) Device D-2 with 25 wt% TmPyPB:FIrpic under different driving voltages of 5, 6, 7 and 8 V. The insets show the normalized EL spectra of these PeLEDs under different such driving voltages (right inset).