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Supporting Information

Boosting Efficiency and Stability of Green InP Quantum Dot Light-Emitting Diodes by Interface Dipole Modulation

Yimin Wang[1], *Qianqian Wu*[2], *Lin Wang*[2,]*, *Zhongjiang Sun*[2], *Fan Cao*[2], *Lingmei Kong*[2], *Lufa Li*[2], *Chengxi Zhang*[2], *Sheng Wang*[2], *Zhijun Zhang*[1] *and Xuyong Yang*[2,]*

[1] School of Materials Science and Engineering, Shanghai University, 149 Yanchang Road, Shanghai 200072, China

[2] Key Laboratory of Advanced Display and System Application of Ministry of Education, Shanghai University, Shanghai 200072, China

*E-mail: lin_wang@shu.edu.cn; yangxy@shu.edu.cn

Experimental Section

Chemicals: Indium(III) acetate (99.999%), zinc undecylenate (98%), palmitic acid (98%, PA), zinc stearate (10% Zn basis), 1-octadecene (90%, ODE), oleic acid (90%, OA), selenium (>99.5%), zinc oxide (99.9%), 1-octanethiol (>98%, OT), methylammonium bromine (98%, MABr), phenylethylammonium bromide (98%, PEABr) were purchased from Sigma-Aldrich. Tris(trimethylsilyl)-phosphine (98%, $(TMS)_3P$) and trioctylphosphine (97%, TOP) were purchased from Stream Chemicals. n-butanol (HPLC, 99.9%) and ethanol (HPLC, 99.5%) were purchased from Innochem. Poly[(9,9-dioctylfluorenyl-2,7-diyl)-co-(4,4'-(N-(pbutylphenyl))diphenylamine)] (99.5%, TFB) was purchased from J&K Chemical Ltd. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) was purchased from Xi'an Polymer Light Technology Corp. All chemicals were used without further purification.

Synthesis of InP/ZnSe/ZnS QDs. The QDs was synthesized by modifying the procedures reported in our previous work.[1] Typically, indium acetate (0.15 mmol), zinc undecylenate (0.15 mmol) and palmitic acid (0.45 mmol) and 10 mL of ODE were mixed in a three-neck flask, which was heated to 120 °C under vacuum for 30 min and then cooled to 50 °C refilled under N_2 atmosphere. Subsequently, 0.13 mmol $(TMS)_3P$ mixed with 1 mL of TOP were injected quickly into the flask and the temperature was raised to 280 °C and maintained for 3 min. Then, 0.1 mmol zinc stearate in 1 mL ODE and 0.1 mmol TOP-Se in 1 mL ODE was injected in sequece and maintained at 280 °C for 2 min and 5 min, respectively, which was repeated for twice. Next, the mixture was cooled down to 200 °C, followed by 7.5 mL $Zn(OA)_2$ was added drop-wise into the flask and kept for 30 min. Subsequently, the mixture was heated to 300 °C then the 0.5 mmol OT was injected and maintained for 15 min. These steps were repeated for five times and maintained at 300 °C for another 1 h. The mixture was cooled down to room temperature and the QDs were precipitated by centrifugation and washed with ethanol for three times. Finally, the QDs were re-dispersed in octane and stored in a vial in a N_2 filled glovebox.

Synthesis of colloidal $Zn_{0.9}Mg_{0.1}O$ *(MZO) nanoparticles.* The QDs was synthesized by modifying the procedures in the reported literature.[2] Typically, a dimethyl sulfoxide (DMSO) solution (30 mL) of zinc acetate hydrate (2.7 mmol) and magnesium acetate (0.3 mmol) were mixed with an ethanol solution (10 mL) of tetramethylammonium hydroxide (5 mmol). The mixture was stirred for 1 h under ambient conditions to generate colloidal MZO nanoparticles. The resulting MZO nanoparticles were precipitated and washed by ethyl acetate. The final precipitate of MZO nanoparticles was re-dispersed in ethanol and stored in a refrigerator (4 °C). The solutions were filtered through 0.22 μm PTFE filters before use.

Preparation of PEABr:MABr solution. The appropriate amount of PEABr and MABr was dissolved in DMSO respectively, to get the solution with concentrations of 0.25, 0.5, 0.75 and 1.0 mg mL^{-1}. The PEABr:MABr solution were prepared by mixing the PEABr solution (0.5 mg mL^{-1}) and MABr solution (0.5 mg mL^{-1}) in volume ratios of 8:2, 7:3, 6:4, 5:5, respectively.

Device Fabrication: The ITO substrates (sheet resistance ≈ 20 Ω sq^{-1}) were sonicated in deionized water, acetone, and isopropanol for 40 min. The ITO substrates were dried in an oven for 30 min and treated by a UV-ozone cleaner for 30 min. The PEDOT:PSS was then deposited by spin-coating at 4000 rpm for 40 s and baked at 150 °C for 15 min on a hot plate. Then the substrates were transferred to the nitrogen glove box. The solution of TFB was spin-coated and annealed at the temperature of 120 °C for 30 min, and the solution of InP QDs was spin-coated and baked at 60 °C for 10 min sequentially. Next, the solution of MZO nanoparticles was spin-coated and baked at 90 °C for 30 min, the solution of MABr was spin-coated and annealed at the temperature of 60 °C for 10 min in sequence. Thereafter, the substrates were loaded into a vacuum chamber (≈1.0×10^{-4} Pa) for the deposition of aluminum (100 nm). The effective light-emitting area of QLEDs is 4.0 mm^2.

Characterizations: The PL spectra were measured by using an Edinburgh FLS920 PL spectrometer. The cross-sectional scanning electron microscopic (SEM) image of the device was characterized by the Hitachi S-4800 scanning electron microscope. The XPS and UPS measurements were performed by using a Thermo Scientific Escalab 250Xi. He (I) ultraviolet radiation source (21.22 eV) from a He discharge lamp was used in UPS measurements. The surface morphology of the films was measured by using atomic force microscope Seiko SPA 400 AFM. The *J-V-L* and *CE-L-EQE* characteristics were acquired by using a fiber integration sphere (FOIS-1) coupled with a QE Pro650 spectrometer and the Keithley 2400 source meter. EL spectra of QLEDs were measured on a QE Pro650 spectrometer. The device lifetime tests of QLEDs were performed on a ZJZCL-1 OLED ageing lifespan test instrument. All measurements were performed at room temperature.

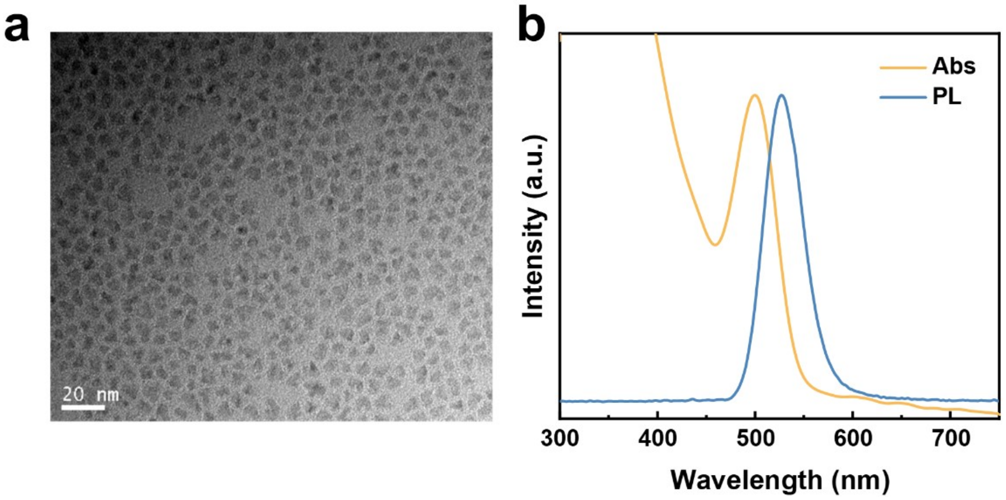

Figure S1 (a) TEM image of InP/ZnSe/ZnS QDs, (b) UV-vis absorption and PL spectra of the QDs. The PLQY of the utilized InP QDs is ~90%.

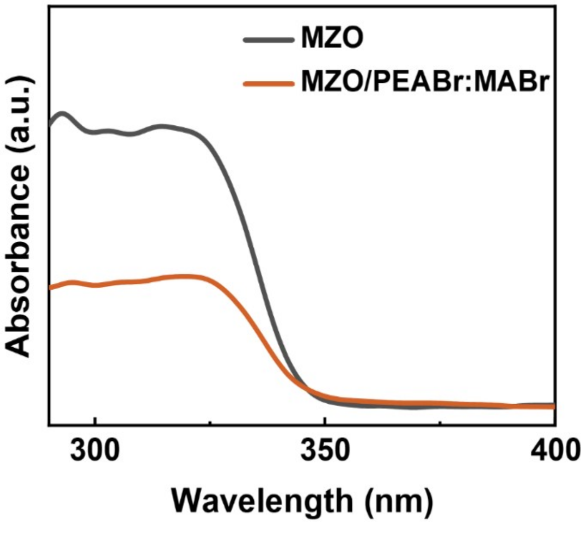

Figure S2 UV-vis absorption spectra of MZO and MZO/PEABr:MABr films.

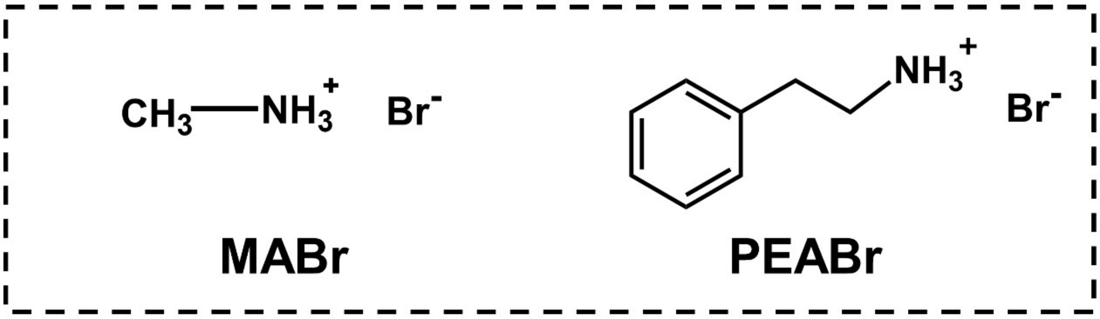

Figure S3 Molecular formulas and molecular structures of PEABr and MABr.

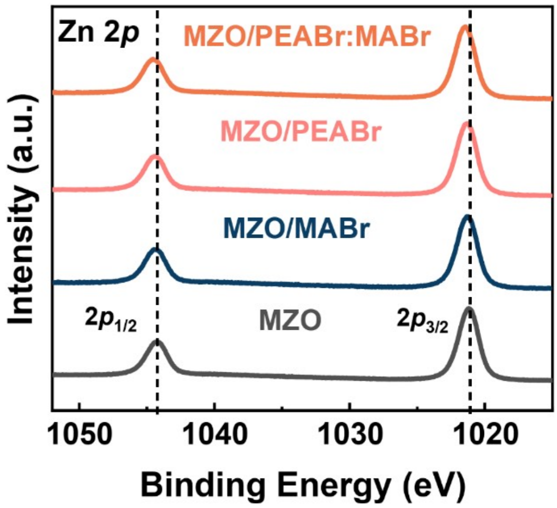

Figure S4 High resolution XPS spectra of Zn 2*p* for MZO, MZO/MABr, MZO/PEABr and MZO/PEABr:MABr, respectively.

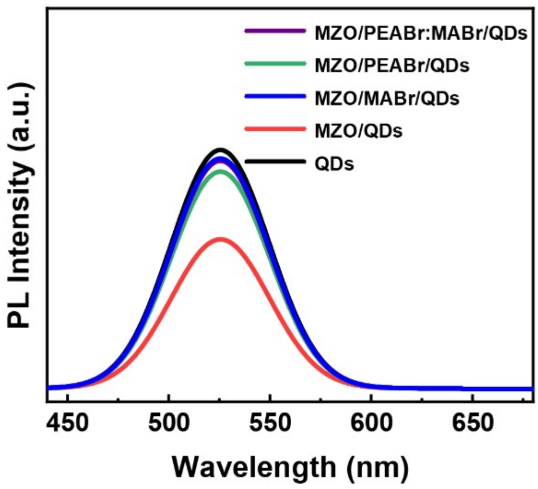

Figure S5 Steady-state PL spectra of samples with structures of QDs, MZO/QDs, MZO/MABr/QDs, MZO/PEABr/QDs and MZO/PEABr:MABr /QDs.

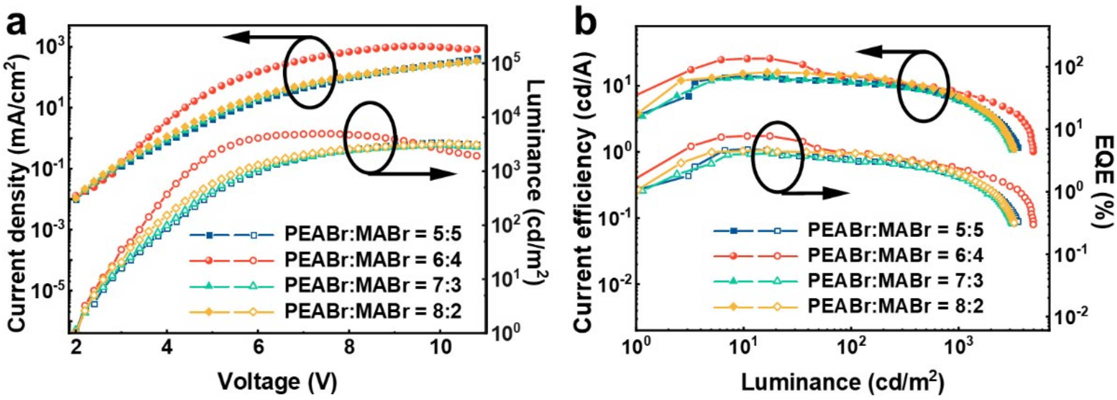

Figure S6 (a) J-L-V and (b) CE-EQE-J curves of InP QLEDs with the PEABr:MABr interlayer of different thickness. The thickness of the interlayer is regulated by adjusting the mixing mass ratio of the PEABr and MABr mixture solutions.

Table S1. Detailed performance parameters of QLEDs with different thick interlayer.

the mixing mass ratio of PEABr to MABr	V_{on} (V)	Max.Lum (cd m^{-2})	Max.CE (cd A^{-1})	Max.EQE (%)
5:5	2	3456	13.8	4.2
6:4	2	4955	25.8	7.8
7:3	2	3101	13.6	4.1
8:2	2	3279	16.0	4.7

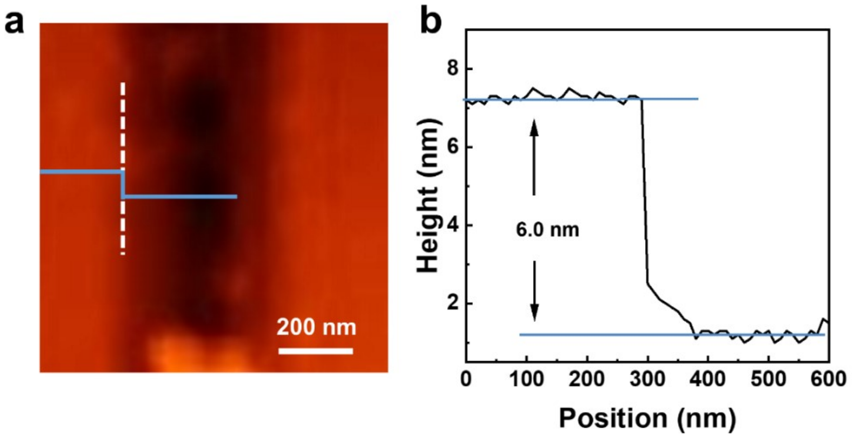

Figure S7 (a) AFM image and (b) the profile of film thickness for the PEABr: MABr interlayer with a mixing mass ratio of 6:4.

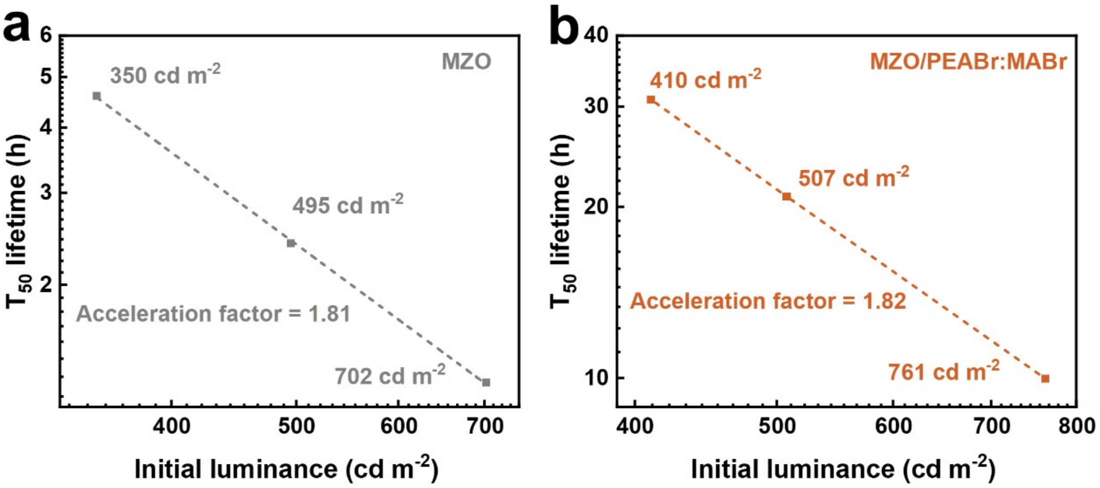

Figure S8 Lifetime measurements under different initial luminance for (a) MZO and (b) MZO/PEABr:MABr films.

References

1 F. Cao, S. Wang, F. Wang, Q. Wu, D. Zhao and X. Yang, *Chem. Mater.*, 2018, **30**, 8002-8007.

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