Supporting Information

High-brightness green InP-based QLEDs enabled by in-situ passivating core surface with zinc myristate

Yuanbin Cheng†, Qian Li†, Mengyuan Chen, Fei Chen*, Zhenghui Wu* and Huaibin Shen

†*These authors contributed equally to this work.*

Key Laboratory for Special Functional Materials of Ministry of Education, National & Local Joint Engineering Research Center for High-efficiency Display and Lighting Technology

Henan University, Kaifeng 475004, China

E-mail: chenfei.henu@henu.edu.cnand wuzhenghuihk@henu.edu.cn

Chemicals

Zinc acetate ($Zn(Ac)_2$, 99.99%), indium acetate ($In(Ac)_3$, 99.99%), selenium (Se, 99.99%, powder), myristic acid (MA, 99%), sulfur (S, 99.5%, powder), zinc stearate ($Zn(St)_2$, 12.5~14.0% ZnO), trioctylphosphine (TOP, 97%), $(TMS)_3P$ (98%), and 1-octadecene (ODE, 90%), tetramethylammonium hydroxide (TMAH, 99%), magnesium acetate tetrahydrate ($Mg(OAc)_2{\cdot}4H_2O$, 99.98%), zinc(II) acetate dihydrate ($Zn(OAc)_2{\cdot}2H_2O$, 99.99%) were purchased from Shanghai Aldrich Reagent Company. Poly((9,9-dioctylfluorenyl-2,7-diyl)-alt-(9-(2-ethylhexyl)-carbazole-3,6-diyl)) (PF8Cz, MW≈80000) were purchased from volt-amp optoelectronics tech. co., LTD. Ethyl alcohol (HPLC) and dimethyl sulfoxide (DMSO, 99.7%) were provided by Acros Reagent Company. Chlorobenzene, n-octane, hexanes, and ethanol were purchased from Beijing Chemical Reagent Ltd., China.

Preparation of precursors

Phosphorus precursors: 0.14 mmol of $(TMS)_3P$ was mixed in 1 mL of TOP.

Se precursors (Se-ODE): 2 mmol of Se was dissolved in 10 mL of ODE. The concentration is 0.2 mol L^{-1}.

S precursors (S-TOP-ODE): 5 mmol of S was dissolved in 5 mL of TOP and 5 mL ODE. The concentration is 0.5 mol L^{-1}.

$ZnMy_2$ precursors: the mixture of 1.25 mmol $Zn(Ac)_2$, 1.25 mmol MA and 10 mL ODE was degassed at 150 °C for 30 min, and then cooled to 100 °C and stored in an N_2-filled flask.

Device fabrication

The indium tin oxide (ITO) glass substrates were thoroughly cleaned with deionized water, acetone, and isopropanol, respectively, and then treated under UV-ozone for 15 min. For the hole injection layer (HIL), poly(3,4-ethyle nedioxythiophene):polystyrenesulfonate (PEDOT:PSS) (AI 4083) was spin-coated onto the ITO substrates and annealed at 140 °C for 15 min. Then, these substrates were swiftly transferred into nitrogen-filled glove box for spin-coating the following layer. PF8Cz (8 mg mL^{-1} in chlorobenzene) was spin-coated and annealed at 150 °C for 30 min for use as hole transport layer (HTL) material. In turn, InP/ZnSe/ZnS (20 mg mL^{-1} in n-octane) and ZnMgO (30 mg mL^{-1} in ethanol, were spin-coated at 2000 and 2500 rpm for 20 s, respectively, and followed by baking at 60 °C for 30 min. Finally, an Al anode was deposited via thermal evaporation under a high vacuum of 4×10^{-6} Torr, and the effective area is 4 mm^2.

Materials and devices characterization

UV-vis absorption and photoluminescence (PL) spectra were measured by Ocean Optics spectrophotometer (model PC2000-ISA). X-ray photoelectron spectroscopy (XPS) was recorded by a

VG ESCALAB 220i-XL spectrometer with a 300 W Al Kα radiation source, and all binding energies for different elements were calibrated with respect to the C 1s line at 284.8 eV. PL QY data was collected by JY HORIBA FluoroLog-3 fluorescence spectrometer coupled with an integrating sphere. A JEOL JEM-2010 electron microscope operating at 200 kV was used to obtain transmission electron microscopy (TEM) studies. X-ray diffraction (XRD) patterns were recorded on a Bruker D8 Advance diffraction meter using a Cu Ka radiation source (λ = 1.54056 Å). An Edinburgh F900 steady/transient state fluorescence spectrometer was used to record the time-resolved PL spectra. The current density-luminance-voltage (J-L-V) characteristics of QLEDs were analyzed using an Agilent 4155C semiconductor parameter analyzer with a calibrated Newport silicon diode. The combination of an Ocean Optics (USB 2000) spectrometer and a Keithley 2400 source meter was used to record the EL spectra. All the measurements were performed at room temperature.

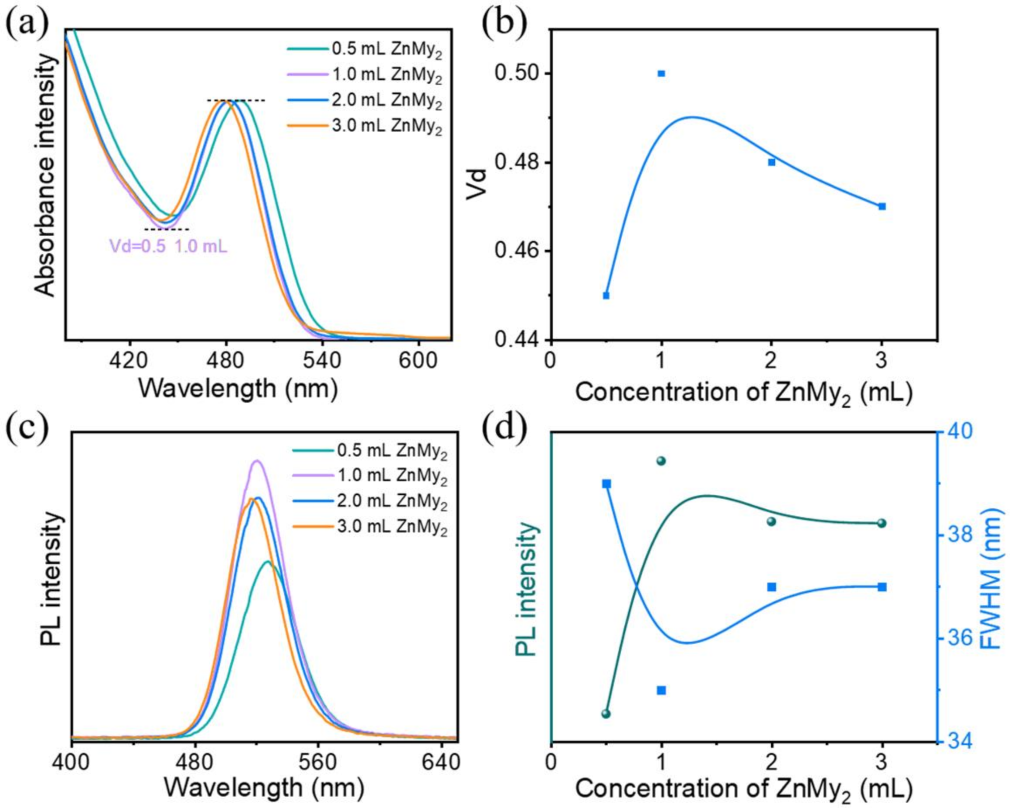

Figure S1. (a) UV-vis absorption spectra of InP cores with $ZnMy_2$ at different concentration. (b) Vd as a function of $ZnMy_2$ concentration. (c) PL spectra of InP cores with $ZnMy_2$ at different concentration. (d) PL intensity and FWHM as a function of $ZnMy_2$ concentration.

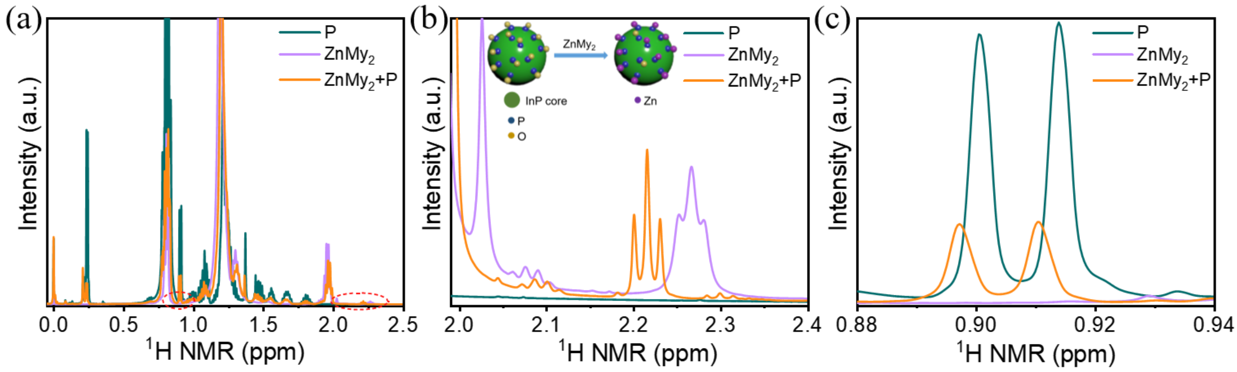

Figure S2. ^{1}H NMR spectra of phosphorus precursors, $ZnMy_2$ and the mixture of phosphorus precursors and $ZnMy_2$ in chloroform-d. Inset is the schematic illustration of the $ZnMy_2$-treated InP core.

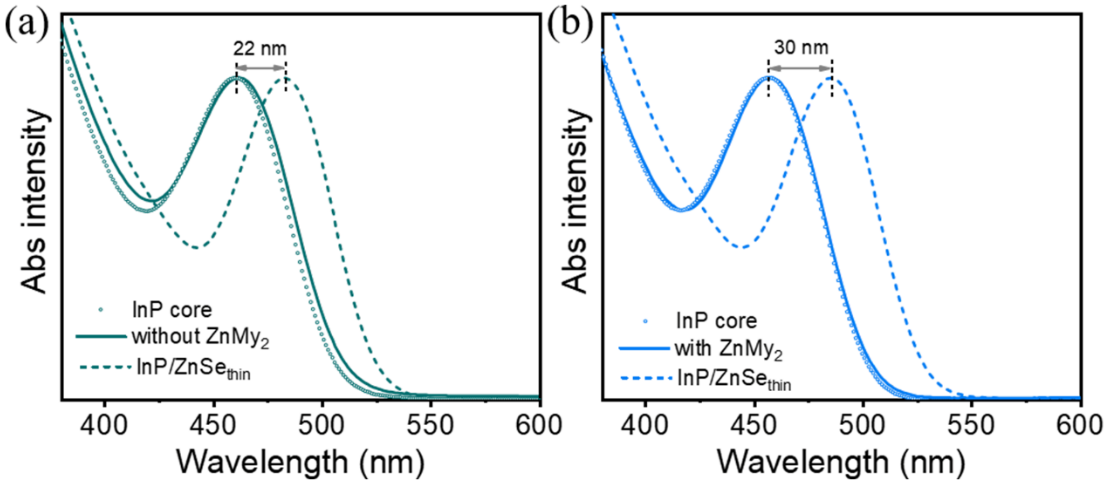

Figure S3. UV-vis absorption spectra of InP/$ZnSe_{thin}$ cores (a) without and (b) with $ZnMy_2$.

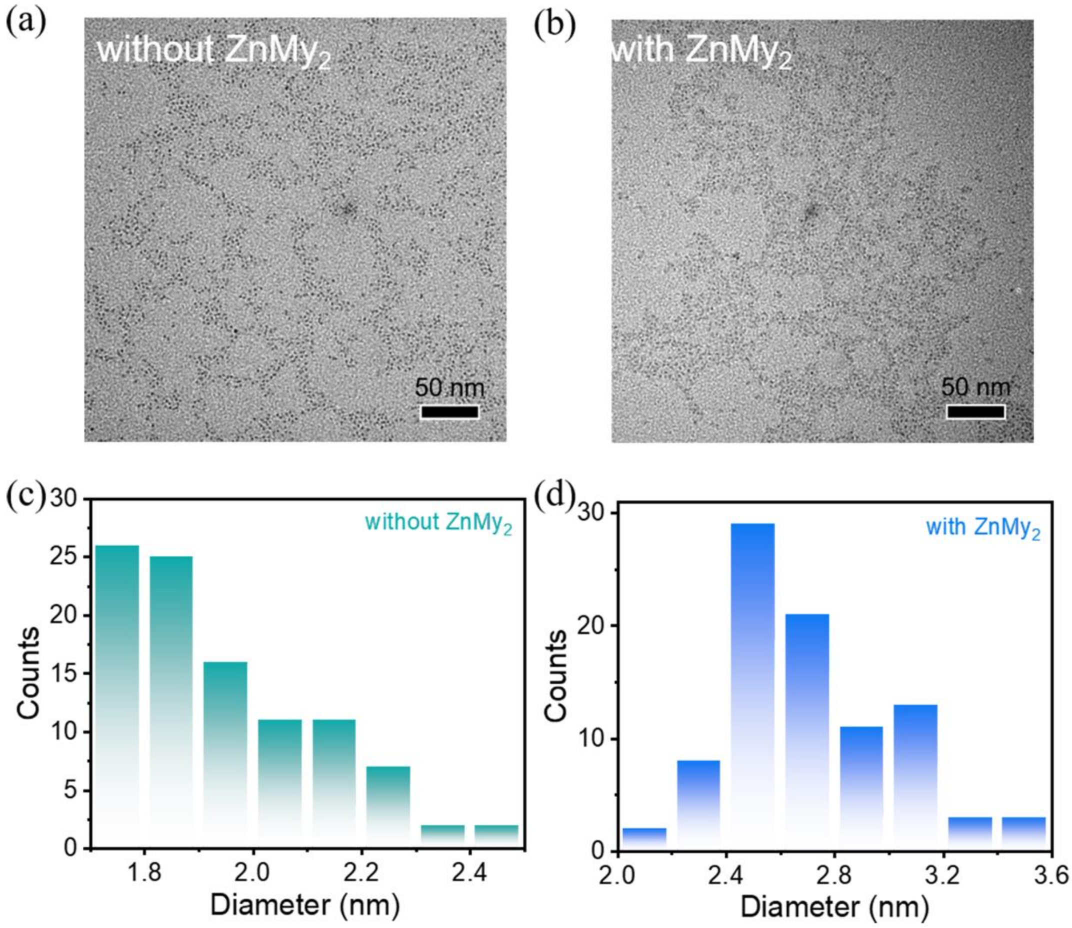

Figure S4. TEM pictures of InP/$ZnSe_{thin}$ core synthesized (a) without and (b) with $ZnMy_2$. The orresponding size distribution histograms of InP/$ZnSe_{thin}$ cores synthesized (c) without and (d) with $ZnMy_2$.

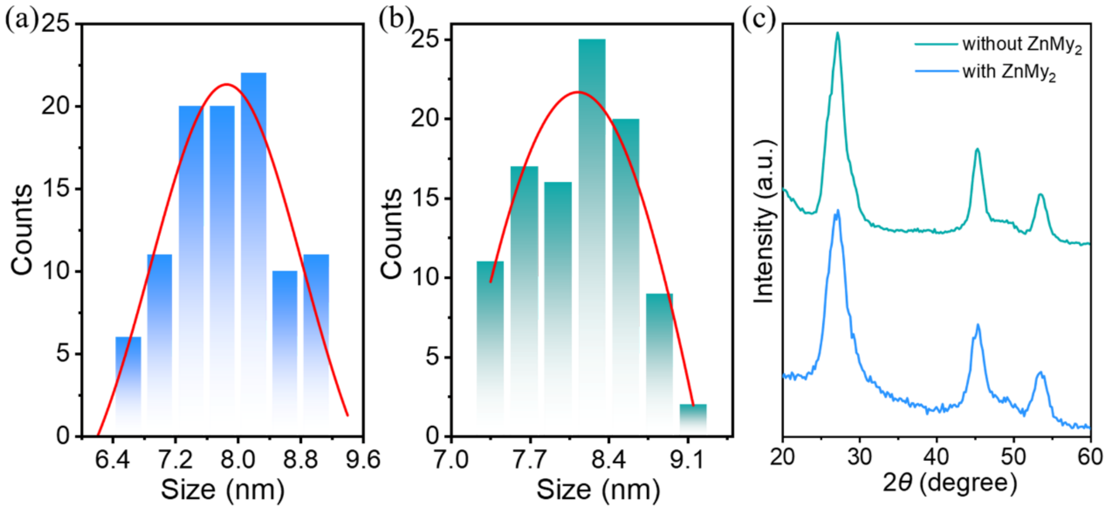

Figure S5. Size distribution histograms of InP/ZnSe/ZnS QDs synthesized (a) without and (b) with $ZnMy_2$. (c) The corresponding XRD patterns.

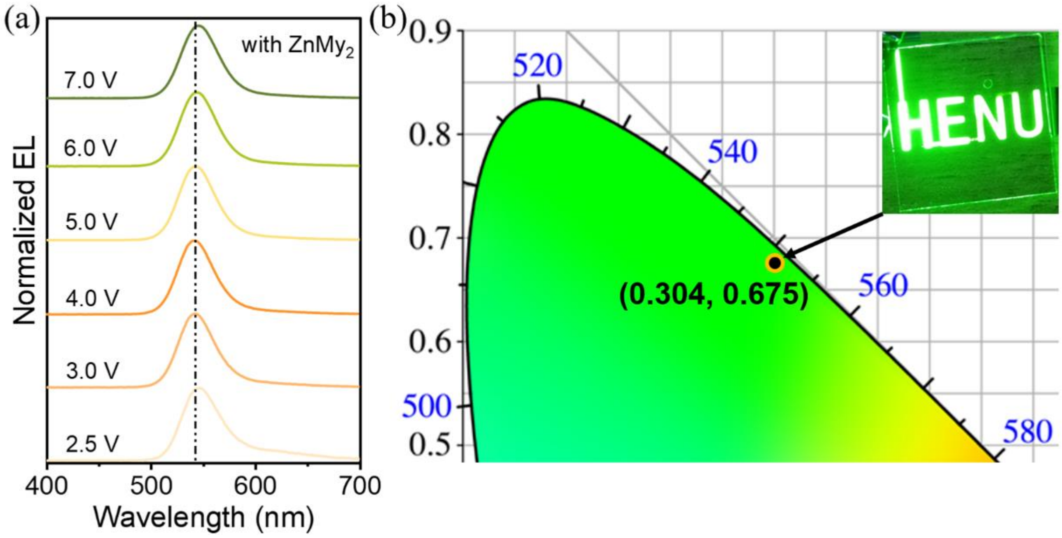

Figure S6. (a) Electroluminescence (EL) spectra of QLEDs under different driving voltages. (b) CIE chromatic coordinates of our QLED, and the inset shows the photograph of EL emission from the device operated at 5 V.

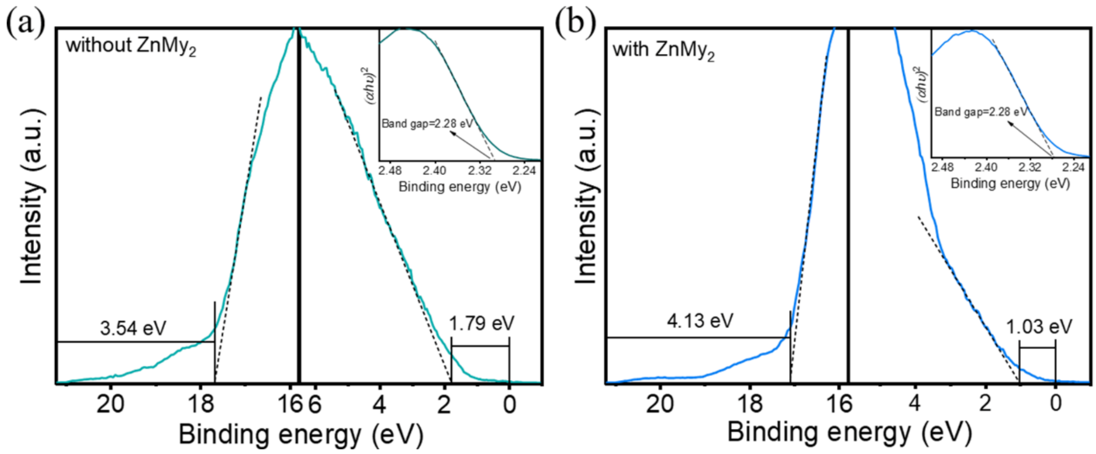

Figure S7. The ultraviolet photoelectron spectroscopy (UPS) spectra of the high-binding energy secondary electron cutoff regions and the valence-band edge regions of InP/ZnSe/ZnS QDs synthesized (a) without and (b) with $ZnMy_2$.

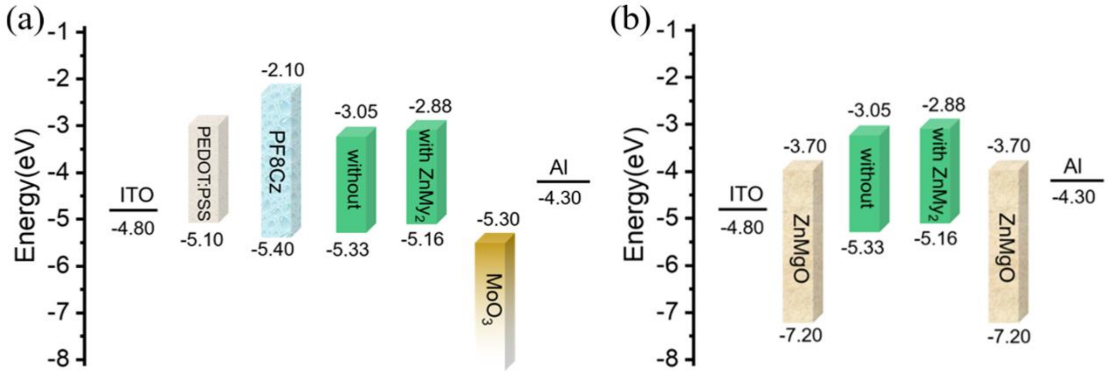

Figure S8. Energy level diagrams of (a) HOD with ITO/PEDOT:PSS/TFB/QDs/MoO_3/Al and (b) EOD with ITO/ZnMgO/QDs/ZnMgO/Al.

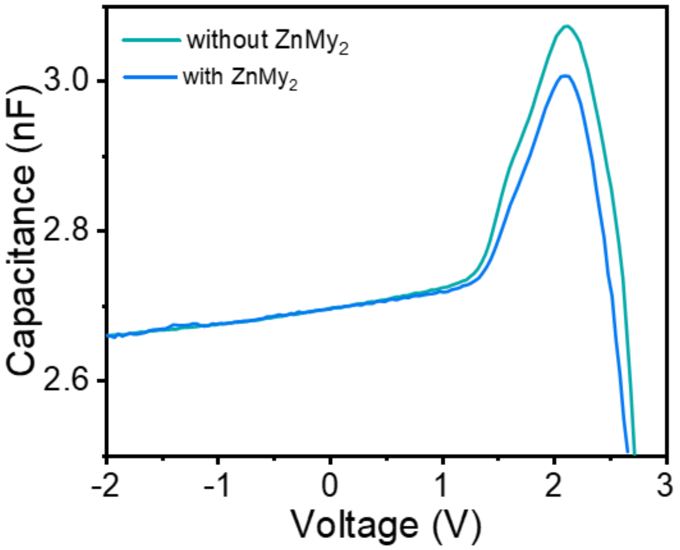

Figure S9. The capacitance-voltage characteristics of QLEDs based on InP/ZnSe/ZnS QDs synthesized without and with $ZnMy_2$.

Table S1. Comparison of the device performance in this work and from other works reported previously.

Year	PL (nm)	FWHM (nm)	PL QY (%)	Device structure	EQE (%)	η_A (cd A^{-1})	L (cd m^{-2})	Von (V)	T_{50}@ 100 cd m^{-2} (h)	Ref.
2013	518	64	80	ITO/ZnO/PFN/InP@ZnSeS/TCTA/MoO_3/Al	3.46	10.9	3900	2.2		1
2019	516	53	67	ITO/PEDOT:PSS/TFB/InP/GaP/ZnS/ZnO/Al	6.3	13.7	2938	2.6		2
2019	531	34	82	ITO/PEDOT:PSS/poly-TPD:PVK/InP QD/ZnMgO/Al	13.6		13900	2.5		3
2019	529	41	86	Glass/Ag/ZnO/PFN/InP/ZnSeS/CzSi/TCTA/MoO_x/Ag		15.3	38800			4
2020	525	40	81	ITO/ZnO/TmPPPyTz/InP QD/TCTA/MoO_3/Al	10			2.4		5
2021	545		86	ITO/ZnMgO/InP/ZnSe/ZnS/TCTA/MoO_3/Al	16.3	57.5	12646.3	2	1033.4	6
2022	526	35	97	ITO/PEDOT:PSS/TFB/InP QD/$ZnSe_xS_{1-x}$/ZnS/ZnMgO/Al	15.2		2300@ 4V	2.1		7
2022	535	43	54	ITO/PEDOT:PSS/PVK/InP/ZnSe/ZnS/PO-T2T/Al	15		10010	3.1	1430	8
2022	528	38	89	ITO/ZnO@ZnS/In(Zn)P/ZnSeS/ZnS/DBTA/PCBBiF/HATCN/Al	10.8	37.5	1756	2.4	60255	9
2022	510	36	91	ITO/PEDOT:PSS/TFB/PVP/InP/ZnSe/ZnS/ZnO/Al	10.6	40.7	15606	1.8	5642	10
2023	533			ITO/PEDOT:PSS/MoO_3/PVK/InP/ZnS/ZnO/Al	7.39		52730	2.5	104.09	11
2023	529		80	Ag/ZnMgO/InP/ZnSe/ZnS/TADF-EHL/TCTA/MoO_3/Ag		68	40700			12
2023	532	36	90	ITO/PEDOT:PSS/PTAA/InP/ZnSe/ZnS/ZnMgO@NaCl/Al	13.8	52.2	16788	2.2	5944	13
2023	535	33.7	95	ITO/PEDOT:PSS/TFB/InP/ZnSeS/ZnS/ZnMgO/Al	14.3	39	11920	2.2		14
2024	534	44	91	ITO/PEDOT:PSS/PF8Cz/InP/ZnSe/ZnS/ZnMgO/Al	12.74	53.31	175084	2.0	20044	This work

Supplementary References

[1] Lim J, Park M, Bae W K, Lee D, Lee S, Lee C and Char K 2013 Highly Efficient Cadmium-Free Quantum Dot Light-Emitting Diodes Enabled by the Direct Formation of Excitons within InP@ZnSeS Quantum Dots *ACS Nano* **7**, 9019-9026.

[2] Zhang H, Hu N, Zeng Z, Lin Q, Zhang F, Tang A, Jia Y, Li L S, Shen H, Teng F and Du Z 2019 High-Efficiency Green InP Quantum Dot-Based Electroluminescent Device Comprising Thick-Shell Quantum Dots *Adv. Optical Mater.* **7**, 1801602.

[3] Moon H, Lee W, Kim J, Lee D, Cha S, Shin S and Chae H 2019 Composition-tailored ZnMgO nanoparticles for electron transport layers of highly efficient and bright InP-based quantum dot light emitting diodes *Chem. Commun.* **55**, 13299-13302.

[4] Lee T, Hahm D, Kim K, Bae W K, Lee C, and Kwak J 2019 Highly Efficient and Bright Inverted Top-Emitting InP Quantum Dot Light-Emitting Diodes Introducing a Hole-Suppressing Interlayer *Small* **15**, 1905162.

[5] Iwasaki Y, Motomura G, Ogura K, and Tsuzuki T 2020 Efficient green InP quantum dot light-emitting diodes using suitable organic electron-transporting materials *Appl. Phys. Lett.* **117**, 111104.

[6] Chao W C, Chiang T H, Liu Y C, Huang Z X, Liao C C, Chu C H, Wang C H, Tseng H W, Hung W Y and Chou P T 2021 High efficiency green InP quantum dot light-emitting diodes by balancing electron and hole mobility *Commun. Mater.* **2**, 96.

[7] Yu P, Cao S, Shan Y, Bi Y, Hu Y, Zeng R, Zou B, Wang Y and Zhao J 2022 Highly efficient green InP-based quantum dot light-emitting diodes regulated by inner alloyed shell component *Light: Sci. Appl.* **11**, 162.

[8] Gao P, Zhang Y, Qi P, and Chen S 2022 Efficient InP Green Quantum-Dot Light-Emitting Diodes Based on Organic Electron Transport Layer *Adv. Optical Mater.* **10**, 2202066.

[9] Mude N N, Khan Y, Thuy T T, Walker B and Kwon J H 2022 Stable ZnS Electron Transport Layer for High-Performance Inverted Cadmium-Free Quantum Dot Light-Emitting Diodes *ACS Appl. Mater. Interfaces* **14**, 5592555932.

[10] Wu Q, Cao F, Wang S, Wang Y, Sun Z, Feng J, Liu Y, Wang L, Cao Q, Li Y, Wei B Wong W Y, and Yang X 2022 Quasi-Shell-Growth Strategy Achieves Stable and Efficient Green InP Quantum Dot Light-Emitting Diodes *Adv. Sci.* **9**, 2200959.

[11] Zhang T, Liu P, Zhao F, Tan Y, Sun J, Xiao X, Wang Z, Wang Q, Zheng F, Sun X W, Wu D, Xing G and Wang K 2023 Electric dipole modulation for boosting carrier recombination in green InP QLEDs under strong electron injection *Nanoscale Adv.* **5**, 385.

[12] Kim J, Hong A, Hahm D, Lee H, Bae W K, Lee T and Kwak J 2023 Realization of Highly Efficient InP Quantum Dot Light-Emitting Diodes through In-Depth Investigation of Exciton-Harvesting Layers *Adv. Optical Mater.* **11**, 2300088.

[13] Wu Q, Wang L, Cao F, Wang S, Li L, Jia G and Yang X 2023 Bridging Chloride Anions Enables Efficient and Stable InP Green Quantum-Dot Light-Emitting Diodes *Adv. Optical Mater.* **11**, 2300659.

[14] Shin S, Gwak N, Yoo H, Jang H, Lee M, Kang K, Kim S, Yeon S, Kim T A, Kim S, Hwang G W and Oh N 2023 Fluoride-free synthesis strategy for luminescent InP cores and effective shelling processes via combinational precursor chemistry *Chem. Eng. J.* **466**, 143223.