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Supporting Information

Fully Solution-Processed Red Tandem Quantum Dot Light-Emitting Diodes with an EQE Exceeding 35%

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■ EXPERIMENTAL SECTION

Synthesis of ZnMgO NPs (using 15% doping concentration as an example): Dissolve 2.56mmol zinc acetate dihydrate and 0.45mmol magnesium acetate tetrahydrate in 30mL dimethyl sulfoxide. Combine 5mmol of TMAH with 5mL of ethanol, and continuously add into the zinc acetate solution at constant speed, then stir for 20 minutes at room temperature. Centrifuge the ZnMgO NPs and rinse twice with ethyl acetate, then dispense them in ethanol to yield a clear solution of 30mg mL^{-1}.

Device manufacturing: Tandem QLEDs are fabricated by means of solution spin coating, where Ag is deposited by thermal evaporation. Patterned ITO substrates ($Rs \approx 30\ \Omega\ \square^{-1}$) are first sequentially cleaned with deionized water, acetone, ethanol, and isopropanol in ultrasonic regime for 15 minutes, then treated with UV ozone for 20 minutes, after which the solution deposition of functional layers takes place. Firstly, PEDOT:PSS (CLEVIOS P VP AI 4083, Heraeus) is spin-coated onto the ITO substrate at 1500rpm in an air glove box, and then thermally treated at 150℃ for 15 minutes. The substrate is then transferred to an glove box with nitrogen, TFB (dissolved in chlorobenzene, 8mg mL^{-1}) is spin-coated at 3000rpm as a hole-transporting layer, and then annealed at 150°C for 30 minutes. QD is procured from Hefei Fullnano Technology Co., Ltd. The red QD (20mg ml^{-1}, solvent is n-octane) is spin-coated at 3000rpm without annealing. ZnMgO nanoparticles are spin-coated at a speed of 1500rpm, then annealed at 80°C for 30 minutes. Then, the substrate is moved back into the air glove box, PEDOT:PSS is spun at 5000rpm, annealed at 100°C for 5min, and then transferred back to the glove box with nitrogen for annealing for 10min. After that, TFB is spun at 3500rpm and annealed at 100°C for 30 minutes. The QD layer fabrication is the same as before. Then, ZnMgO nanoparticles are spin-coated at a speed of 2000rpm and annealed at 80°C for 30 minutes. Subsequently, the sample is transferred to a high vacuum evaporation chamber to evaporate the Ag cathode (100nm). The TQLED has the same effective area of 2.0 mm×2.0 mm,

defined by the overlapping area of the anode and cathode. The top and bottom devices are single junction QLEDs and are fabricated using the process parameters specific to each unit of the tandem device.

Device Characterization: UV-visible absorption spectra were obtained by Perkin-Elmer Lambda 950. The cross-sectional TEM images of the tandem QLEDs were obtained by a Talos F200X with 200 kV acceleration voltage. The work function of each layer and the energy level alignment of the ZnMgO with different doping concentrations were explored by ultraviolet photoelectron spectroscopy (AXIS SUPRA+). Photoluminescence (PL) spectra were obtained by Horiba FL3-111 with an excitation source at 400 nm. Time-resolved PL (TRPL) decay spectra were obtained by Horiba FL3-111. Electroluminescence spectra were obtained using an Ocean Optics USB 2000+ spectrometer with the devices driven at a constant current with a Keithley 2400 source meter. The C-V curve of the ICL were were measured by a BioLogic SP-50e electrochemical work station. The J-L-V characteristics of the devices were taken under ambient conditions with a Keithley 2400 source meter measuring the sweeping voltages and currents and a Keithley 6485 Picoammeter together with a calibrated silicon detector (Edmund) measuring light intensities. Luminance was calibrated using a photometer (Spectra Scan PR655) with the assumption of the Lambertian emission pattern of all devices. The operational lifetime test was conducted under ambient conditions at room temperature (22±2℃) using a commercialized lifetime test system (Guangzhou Jinghe Equipment Co., Ltd). The devices were encapsulated with Nagase UV epoxy resin XNR5516Z(C)-SA1 and capping glasses.

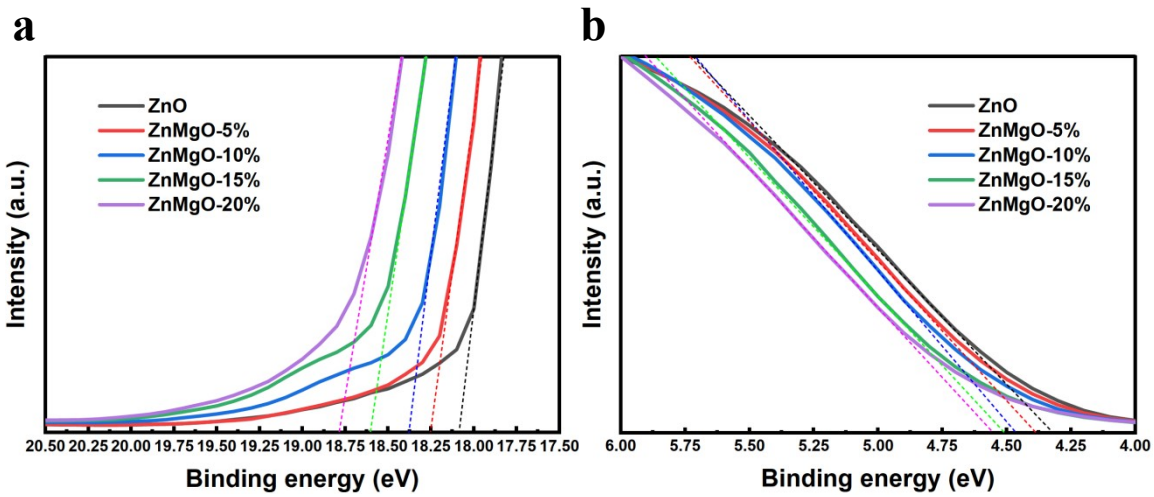

Figure S1. (a) The Ecut-off region of ZnO NP films and ZnMgO NP films with different Mg doping levels. (b) Valence band edge regions of ZnO NP films and ZnMgO NP films.

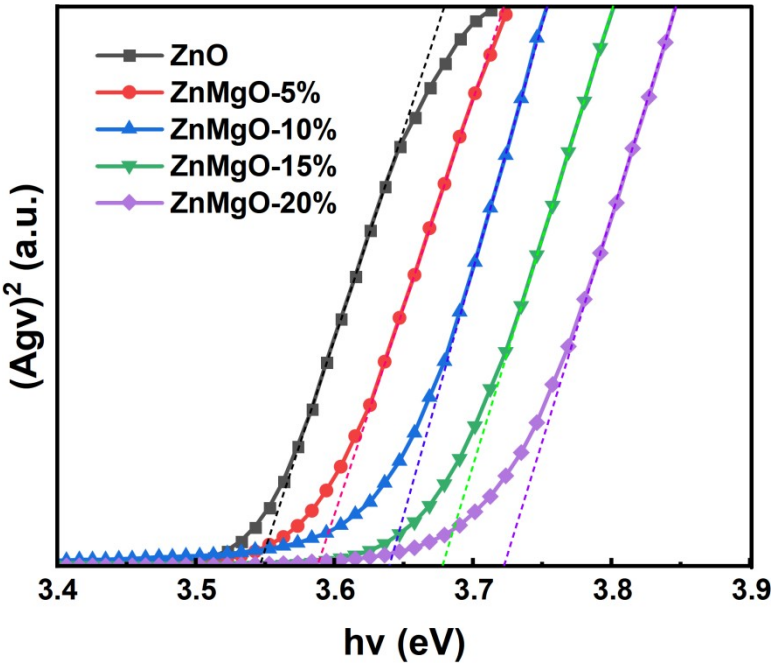

Figure S2. Adsorption spectrum of ZnMgO NP films with different Mg doping levels.

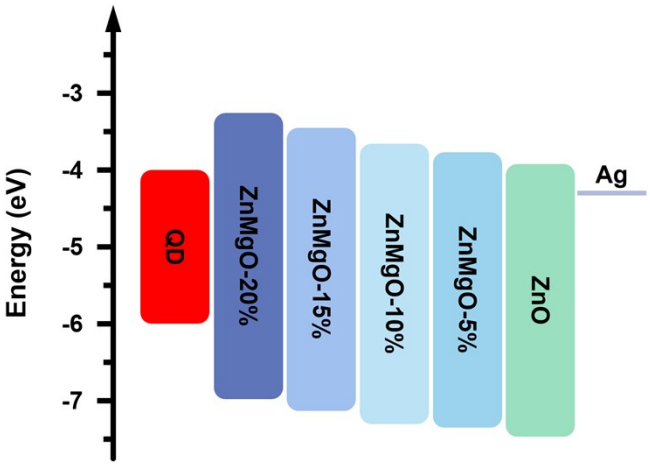

Figure S3. Energy band diagram of ZnMgO with different Mg doping concentrations.

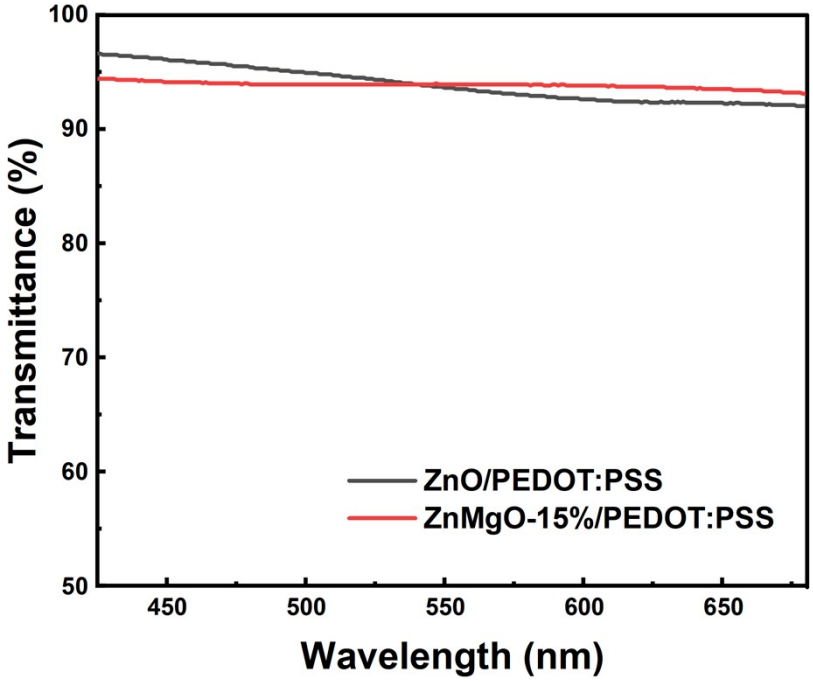

Figure S4. Transmittance characteristics of the ICL with structures of ZnO/PEDOT:PSS and ZnMgO-15%/PEDOT:PSS.

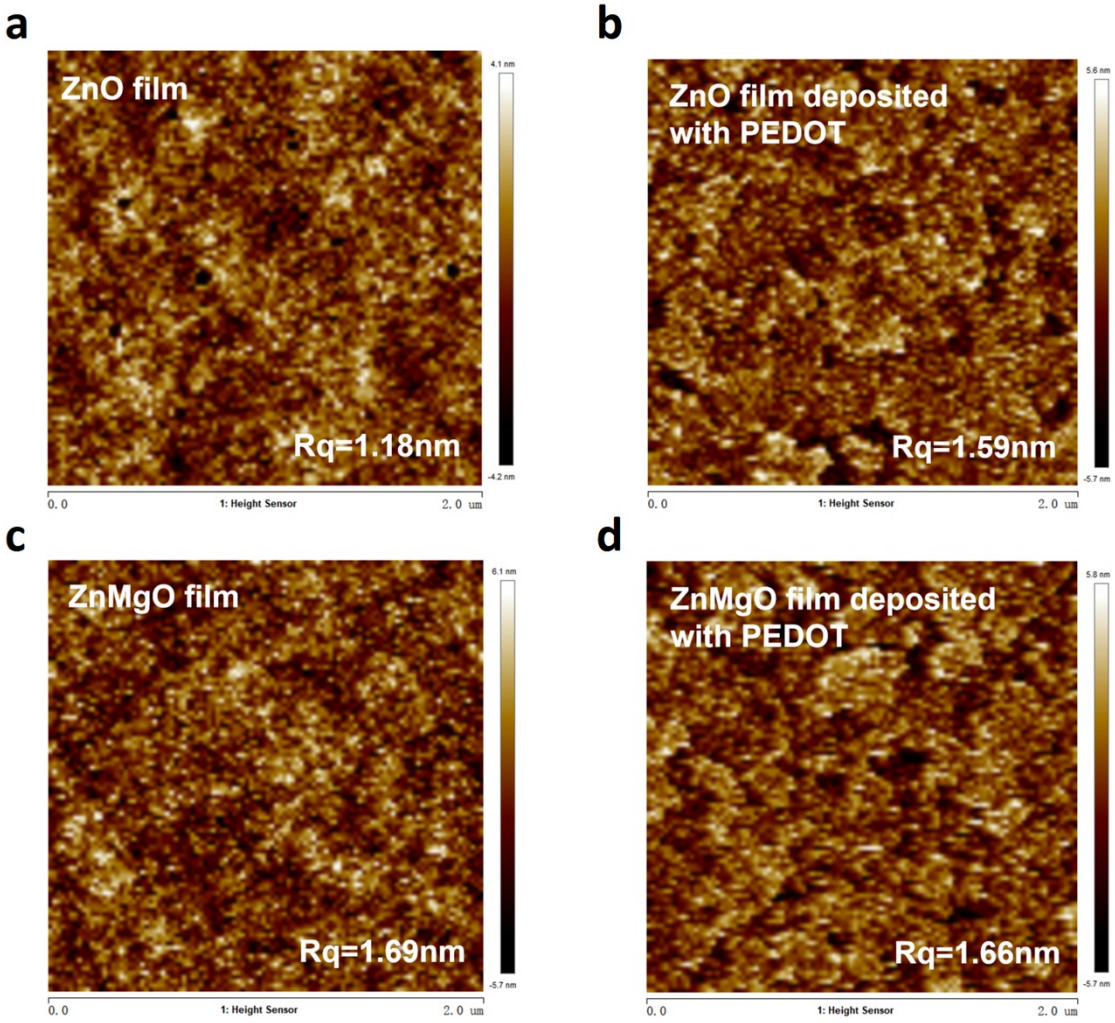

Figure S5. (a) ZnO film, (b) ZnO film deposited and cleaned by PEDOT:PSS, (c)ZnMgO flim, (d) ZnMgO film deposited and cleaned by PEDOT:PSS.

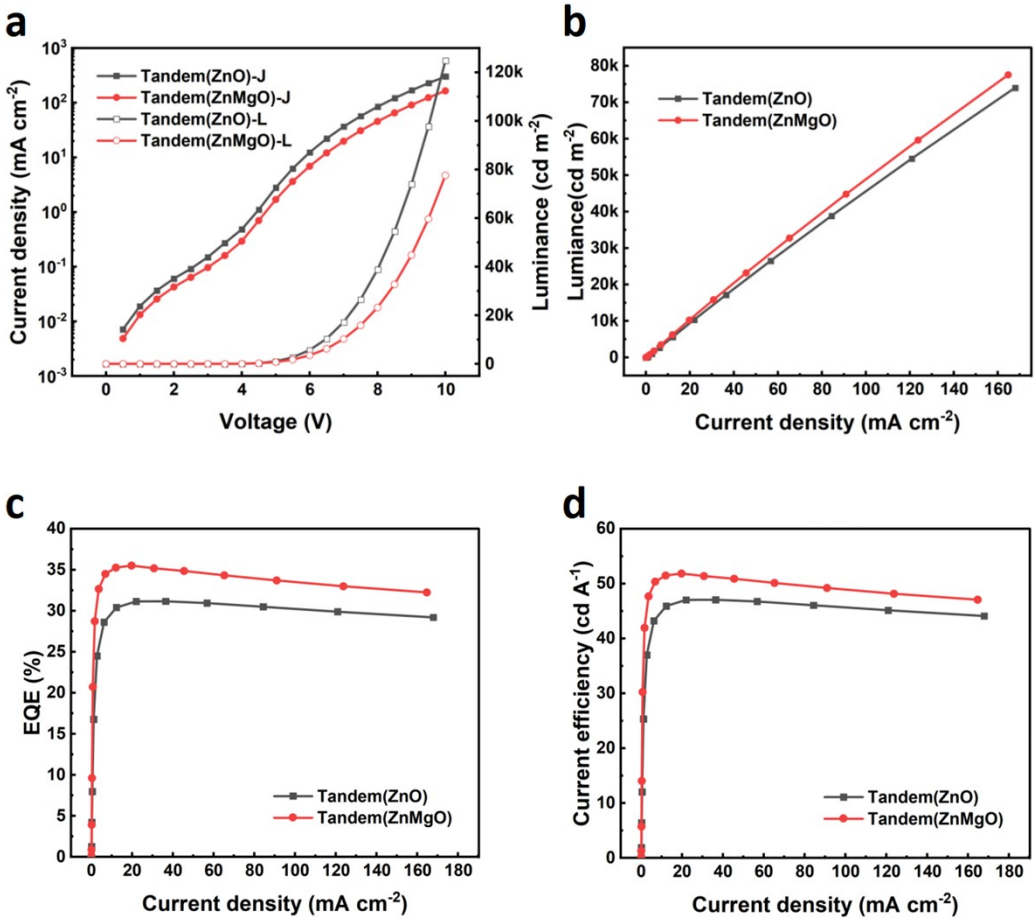

Figure S6. (a) J-V-L, (b) J-L, (c) EQE-J, (d) CE-J curves of T-QLEDs using ICL with ZnO and ZMO

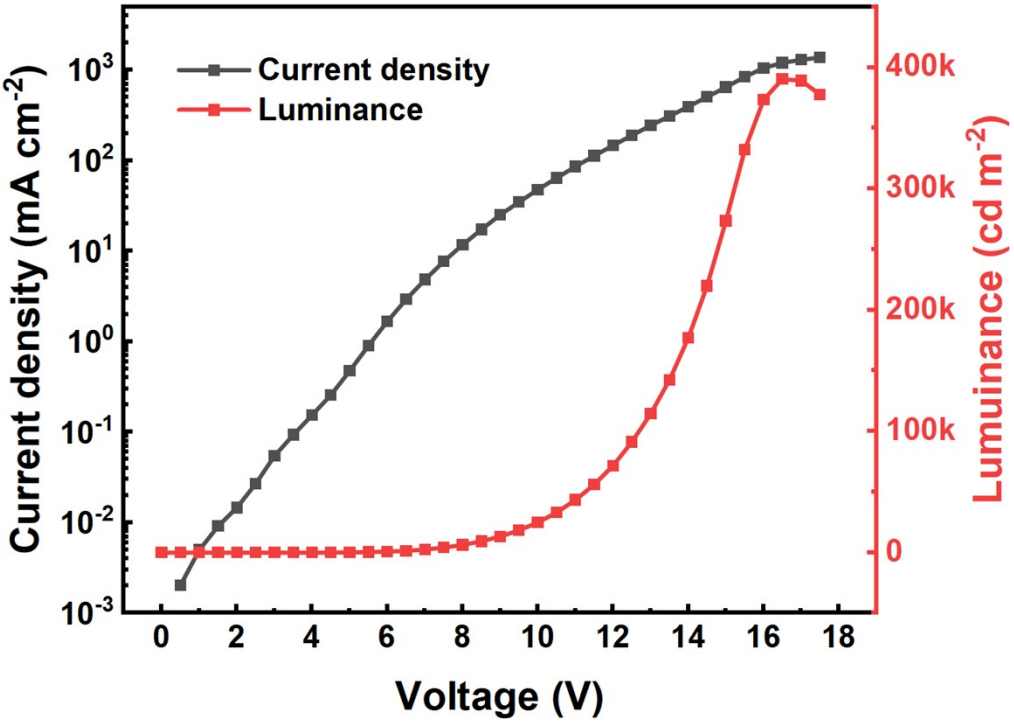

Figure S7. Current density-Voltage-Luminance (J-V-L) curves for red tandem QLED.

Table S1. Energy band data of ZnMgO with different Mg doping concentrations.

Material	$E_{cut-off}$	E_{onset}	E_g	VBM	CBM
ZnO	4.33	18.08	3.55	7.47	3.92
ZnMgO-5%	4.39	18.26	3.58	7.35	3.77
ZnMgO-10%	4.46	18.38	3.64	7.30	3.66
ZnMgO-15%	4.51	18.60	3.68	7.13	3.45
ZnMgO-20%	4.55	18.79	3.72	6.98	3.26