Supporting Information

High-Performance QLED Research Based on Direct Photo-crosslinking of PEDOT:PSS

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Supporting Information

For the optimization of photosensitizer content, a series of concentration optimizations (5 wt%, 10 wt%, 15 wt%, 20 wt%, 30 wt%, 40 wt%) were carried out to determine the optimal content of photosensitizer from two perspectives of film roughness and device performance. HOD content optimization of photosensitizer showed the same trend after PEDOT 4083 and PEDOT PH1000 photo-crosslinking. When the amount of HOD was 10 wt%, 20 wt%, 30 wt% and 40 wt%, the roughness of the film also increased with the increase of the amount of photosensitizer. This is shown in Figure S1 and S2. In terms of devices, the experiments were carried out under the conditions of adding 5 wt%, 10 wt% and 15 wt% of photosensitizer. It can be seen from Figure S3 that the device performance is the best when the amount of photosensitizer is 10 wt%. Therefore, the optimal amount of HOD was determined to be 10 wt% through the photosensitizer optimization experiment.

With the increase of HOD content, the roughness of PH1000/HOD films increased from 1.34 nm (10 wt% HOD) to 1.53 nm (40 wt% HOD), and the roughness of 4083/HOD films also showed an increase. Increased from 1.12 nm (10 wt% HOD) to 1.31 nm (40 wt% HOD). **Figure S1** shows the impact of different amounts of the photosensitive agent HOD (10 wt%, 20 wt%, 30 wt%, 40 wt%) on the roughness after photo-crosslinking in PEDOT PH1000.

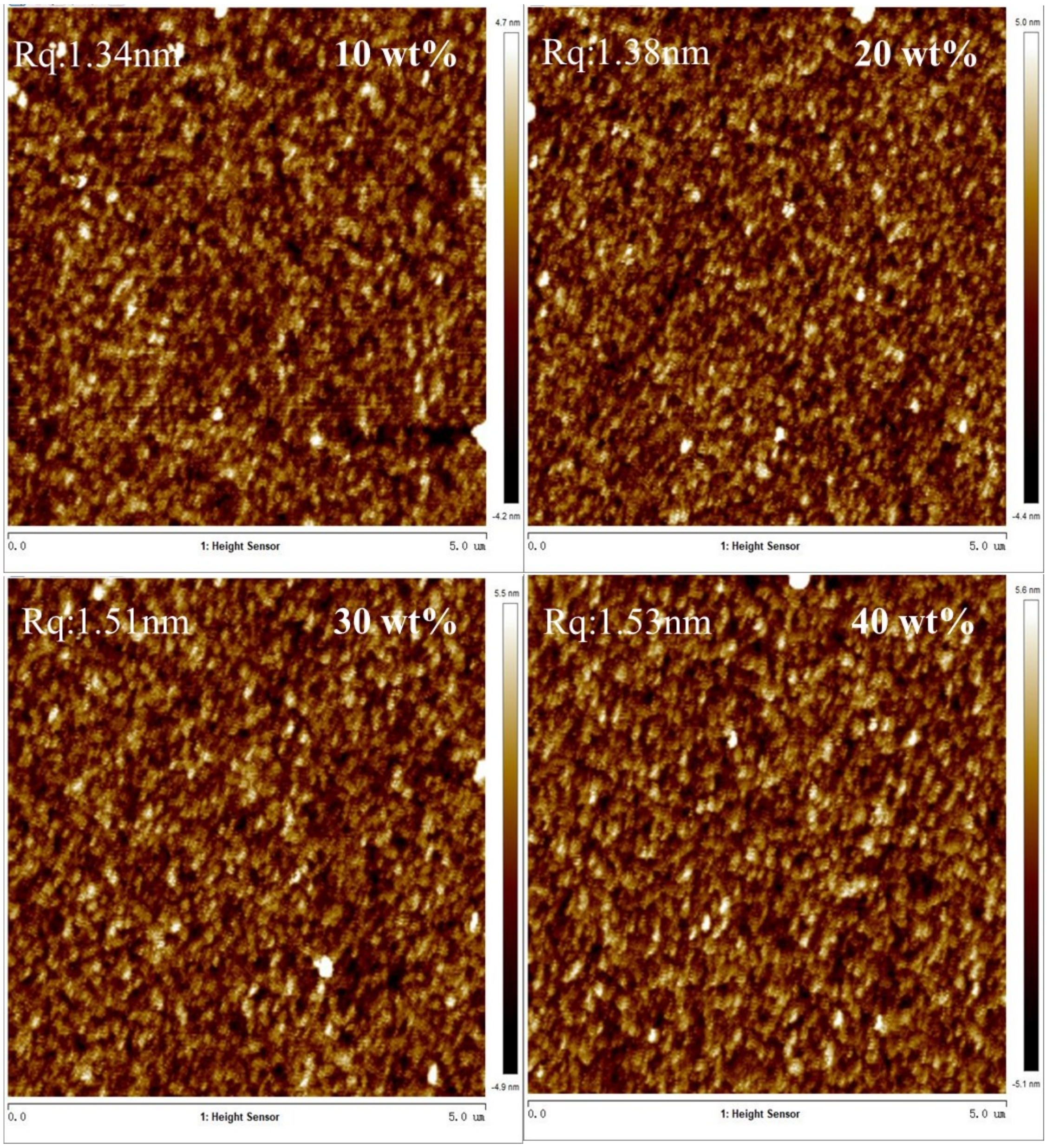

Figure S1. The roughness of the film after photo-crosslinking with different content of photosensitizer added to PEDOT PH1000 (10 wt%, 20 wt%, 30 wt%, 40 wt%).

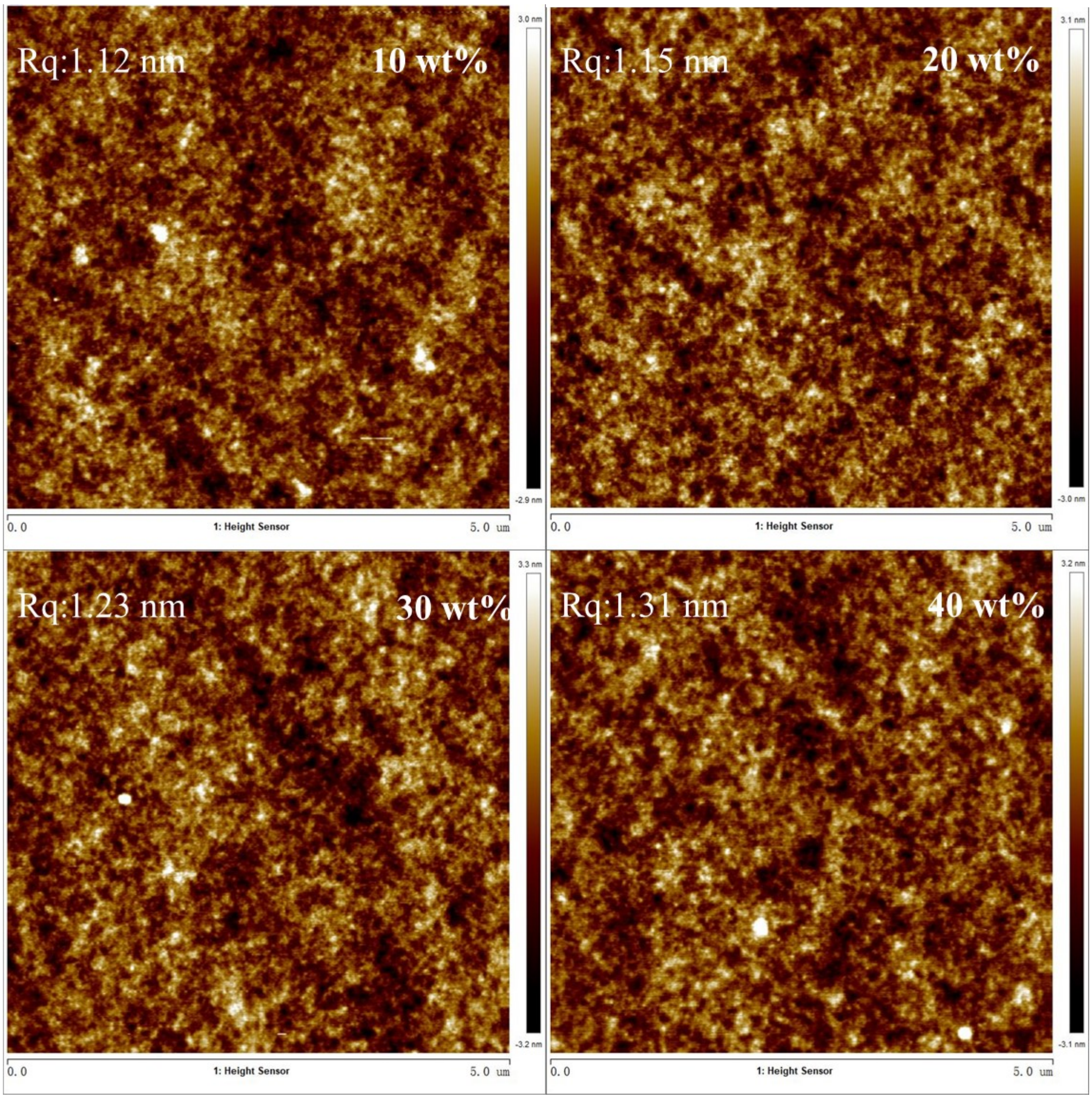

Figure S2. The roughness of the film after photo-crosslinking with different content of photosensitizer added to PEDOT 4083 (10 wt%, 20 wt%, 30 wt%, 40 wt%).

Figure S3 shows the extractor-device quantum efficiency diagram of QLED prepared with 4083/HOD and PH1000/HOD as HIL under different addition amounts of HOD (0 wt%, 5 wt%, 10 wt%, 15 wt%). The content of HOD for optimal device performance in PEDOT 4083 and PEDOT PH000 can be found to be 10 wt%. The EQE of QLED devices with PH1000/HOD-10 wt% reached 18.1%, which is higher than the performance of devices with other HOD additions. It is also much higher than 4083/HOD-10 wt% devices (EQE=14.2%).

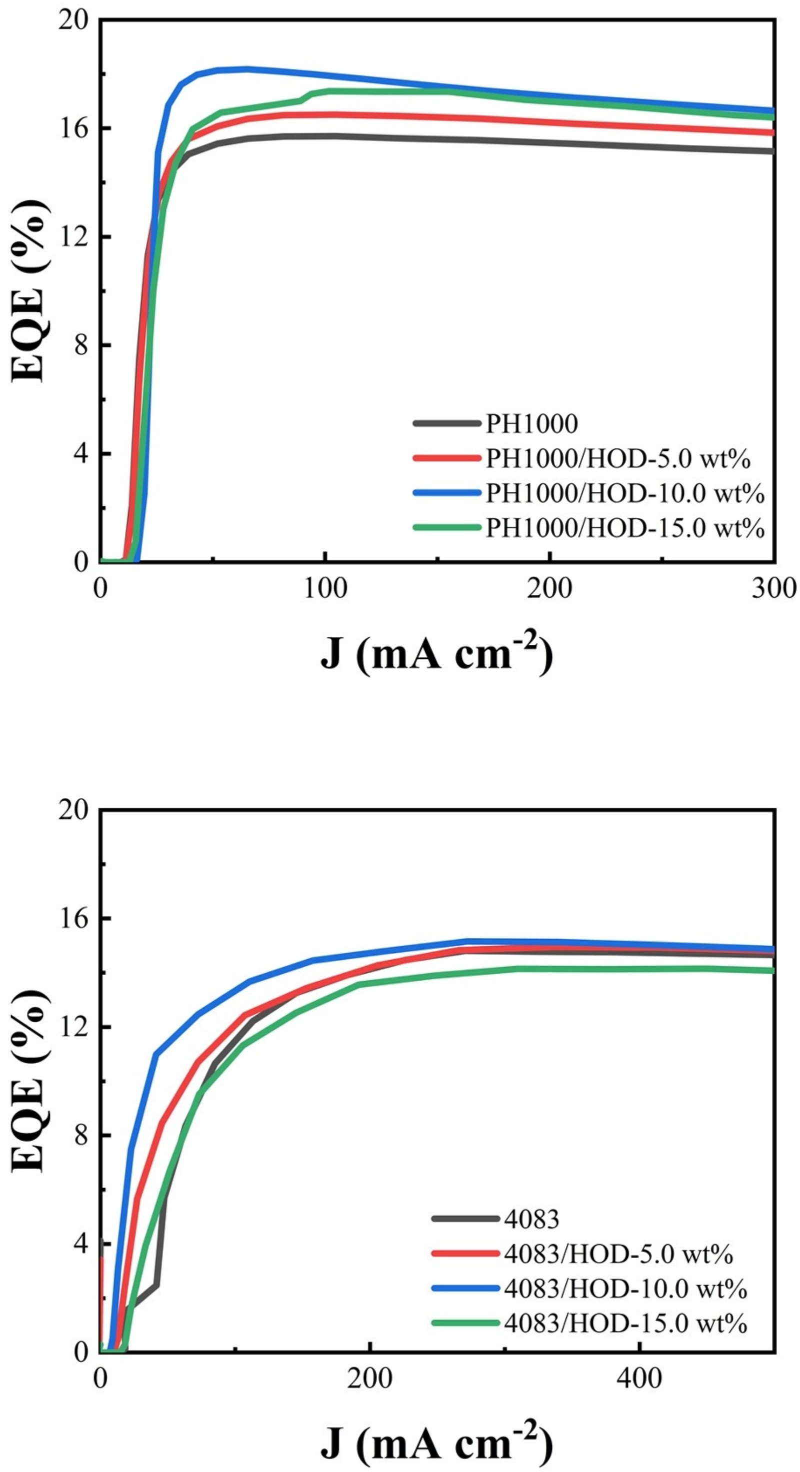

Figure S3. External quantum efficiency performance of QLED devices after adding different amounts of photosensitizers (0 wt%, 5 wt%, 10 wt%, 15 wt%) to 4083 and PH1000 photo-crosslinking.

Comparison of photo-crosslinking with the peak position and full width at half maximum

(FWHM) of original devices, red QLEDs prepared with original PH1000 and PH1000/HOD photo-crosslinking as the hole injection layer (HIL) exhibit consistent peak position and FWHM, with a peak position of 625 nm and FWHM of 23 nm, as shown in **Figure S4.**

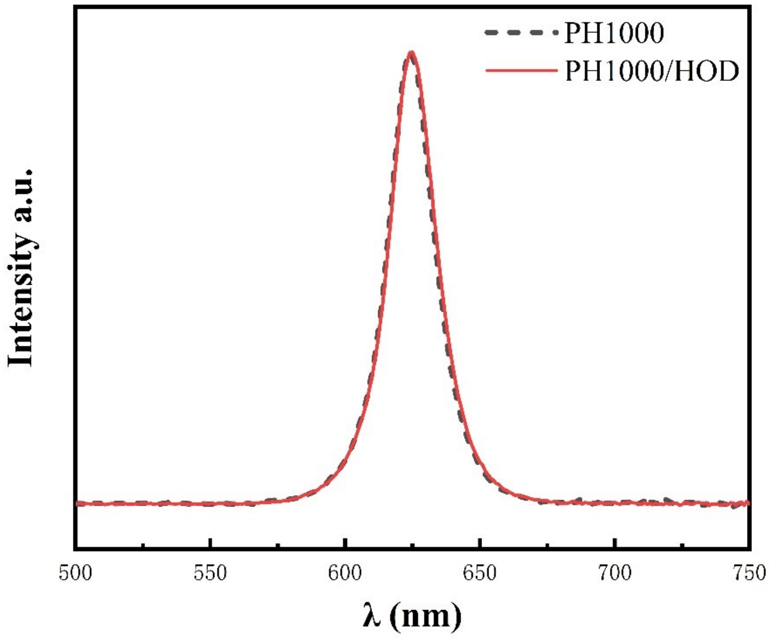

Figure S4. PEDOT PH1000 and PH1000/HOD photo-crosslinking as HIL red QLED devices for peak and Semi-maximum Total width (FWHM).

For the reference sample, At an initial brightness of 1000 nit, the operational lifetime (T_{95}) of the original PH1000 device is 164 hours, as shown in **Figure S5**.

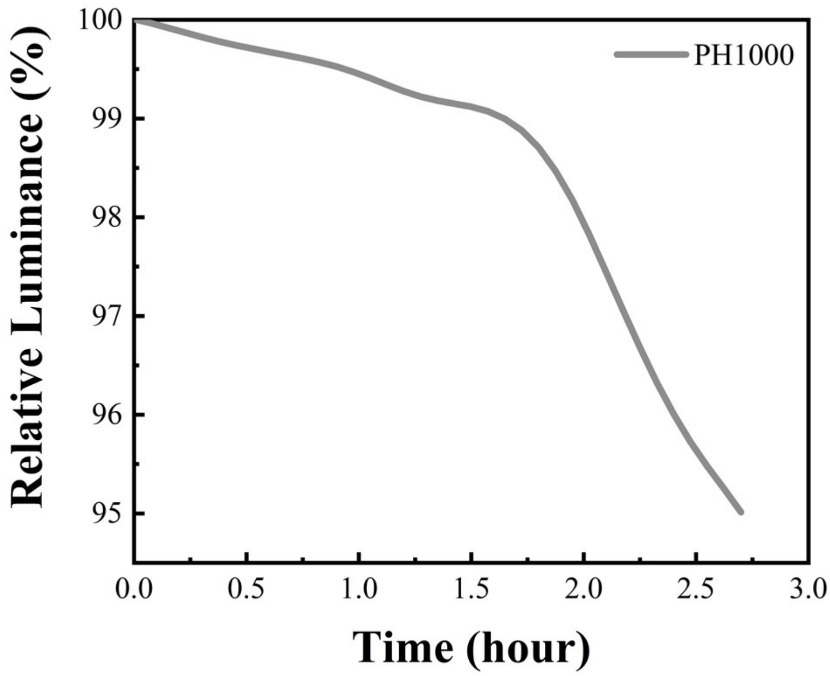

Figure S5. Lifetime (T_{95}) of PEDOT PH1000 device.

The EQE distribution characteristics based on PEDOT 4083 and PEDOT PH1000 and the treated PEDOT:PSS used as HIL to produce more than 20 QLED devices are shown in Figure S7. PH1000/HOD as a HIL QLED device has an average EQE of 17.9% and a relative standard deviation of 3.2%. Not only better than PEDOT PH1000 QLED devices, further than 4083/HOD devices.

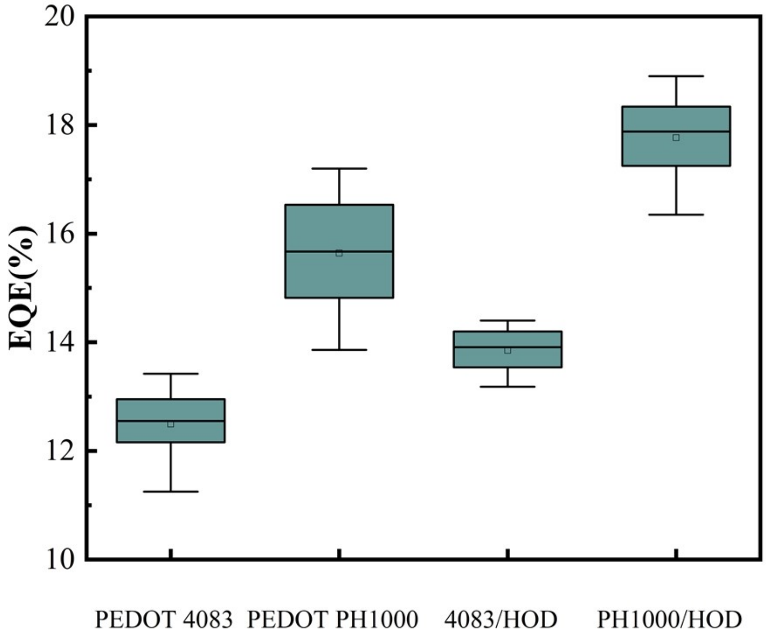

Figure S6. PEDOT 4083, PEDOT PH1000, 4083/HOD and PH1000/HOD were used as the EQE value distribution characteristics of QLED devices prepared by HIL.

After photo-crosslinking PEDOT, the configuration changes, and the PEDOT chain changes from benzenoid structure to quinoid structure.

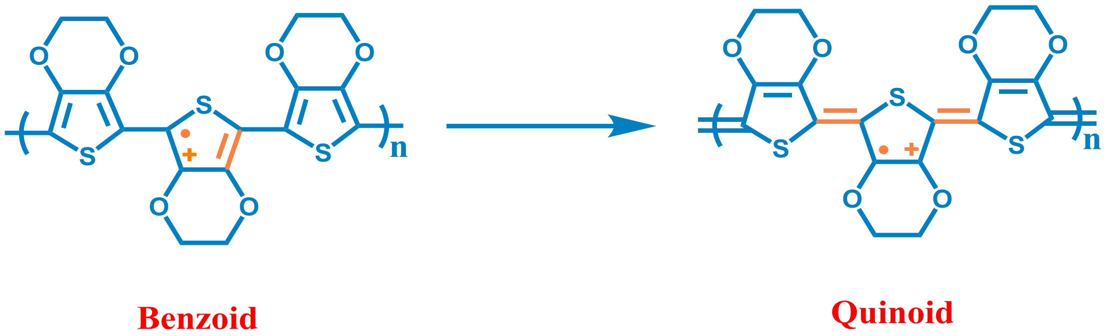

Figure S7. Configuration change of PEDOT before and after optical crosslinking (benzenoid structure to quinone structure).

Experimental Section

Materials: PEDOT:PSS aqueous solution (PH1000/4083) was received from Heraeus. Photosensitizer (2,4-diyne-1,6-hexanediol) was procured from Alfa Aesar (China). The TFB was obtained from LinkZill. The CdSe/ZnS QDs and ZnO nanoparticles (ZnO NPs) were ordered from Full Nanotech, Hefei, China. The ITO conductive glass with a surface resistance of 30 $\Omega \cdot sq^{-1}$ was purchased from CSG Holding Co. All chemicals were used as received without further purification.

Device Fabrication: The indium tin oxide (ITO) substrate was cleaned with deionized water, acetone and ethanol and subjected to ultrasonic treatment for 15 minutes each. After drying, the substrate is exposed to UV ozone for 15 minutes. In air, a photosensitive PEDOT:PSS solution (10 wt% HOD/ ml) was spun onto ITO at 3000 rpm and then exposed to UV for 15 min (254 nm). Note that the entire process was operated in a yellow light room. Then, bake at 150°C for 15 minutes. Then spin TFB (8 mg/ml chlorobenzene solvent) at 4000 rpm in a glove box and bake at 150°C for 30 minutes. The CdSe–ZnS QD (20 mg*ml^{-1}, n-octane) solution were spin-coated at 2000 rpm for 40 s. Next, ZnO layers were prepared by spin-coating the ZnO NP solution (30 mg ml^{-1} in alcohol) at 2000 rpm for 30 s on QD surface and subsequently dried at 80 °C for 30 min. 150 nm thick layer of silver (Ag) was deposited by vapor deposition as the electrode. The active area of the QLED devices was 4.0 mm^2. Finally, the QLED devices were packaged by curing glue from Nagase chemteX corporation.

Characterizations: The surface morphology of the films was detected by scanning electron microscopy (SEM). The morphology analyses were obtained using Kelvin probe force microscopy (Brook dimension icon). UPS measurements were carried out using a Kratos AXIS ULTRA DALD UPS system. Particle size analysis was performed using a dynamic Light scattering particle size analyzer (ZETA) (model: Zetasizer Nano ZS). The molecular structures of pristine and photosensitive PEDOT:PSS were investigated by Raman spectroscopy (Renishaw inVia Reflex). Electroluminescence spectra were collected using an Ocean Optics USB 2000+ spectrometer, which operated under the constant current drive of a Keithley 2400 source meter. Luminance is calibrated using a photometer (Spectra Scan PR655) and all devices were assumed to be in Lambert emission mode. The operating lifetime tests of QLED devices were measured through a commercialized lifetime test system (Guangzhou Jinghe Equipment Co., Ltd) under ambient conditions.