

Supplementary Material

for

Size matters: quantum confinement-driven dynamics in CsPbI₃ quantum dot light-emitting diodes

Shuo Li, Wenxu Yin, Weitao Zheng, Xiaoyu Zhang*

School of Materials Science and Engineering, Jilin University, Changchun 130012, China

* Corresponding author: XY Zhang, zhangxiaoyu@jlu.edu.cn

Materials

Cs₂CO₃(99.995%), PbI₂(99.999%), Oleic acid (OA, 90%), octadecene (ODE, 90%), poly (Maleic Anhydridealt-1-octadecene) (PMA) were bought from Sigma-Aldrich. ZnI₂ (99.99%) and oleylamine (OLA, 70%) were purchased from Aladdin. Poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) (4083), poly (N, N'-bis(4-butylphenyl)-N, N'-bis- (phenyl)benzidine) (poly-TPD), 2,4,6-tris[3-(diphenylphosphinyl)phenyl]-1,3,5-triazine (PO-T2T) and LiF were purchased from Xi'an Yuri Solar Co. Ltd. N-hexane (99.5%) and ethanol (99%) were obtained from Beijing Chemical Factory. N-octane (99%) and methyl acetate (99%) were purchased from J&K Chemicals. All chemicals were used directly without further purification.

Methods

Preparation of the cesium precursor. 0.814 g Cs₂CO₃, 30mL ODE, 2.6 mL OA were added to a 50 ml three-neck flask and keep at 120°C under vacuum for one hour.

It is filled with nitrogen and heated to 150°C until all solids are dissolved. It needs to be preheated to 90 °C to form a clarified solution before injection.

Synthesis of CsPbI₃ NCs. 0.173 g PbI₂, 0.346 g PMA, 0.12 g ZnI₂ and 10 mL ODE were loaded into a 50 ml three-neck flask and kept at 120°C under vacuum for 1 h. The flask is then filled with N₂. Then, 2 mL OLA and OA are injected into the flask. After all the solid substances are dissolved, the temperature is heated to 170°C, and 1.5 mL of the cesium precursor is rapidly injected. The reaction is stopped immediately in the ice water bath after 10 s.

Purification of CsPbI₃ NCs. 20 mL of ethyl acetate was added to the reaction mixture, and the solution was transferred to a centrifuge cylinder at 10000 rpm for 10 min. After centrifugation, the supernatant was discarded and 2 mL of n-hexane was added to the precipitation for dispersion. Add 4 mL methyl acetate and centrifuge at 8000 rpm for 5 min, then disperse the precipitate with 1 mL n-octane. The mixture is subsequently centrifuged at a speed of 12,000 per minute for five minutes to segregate the supernatant from the precipitate. Thereupon, the precipitate is dispersed with 500 microliters of n-octane as a large-size nanocrystal solution, whereas the supernatant constitutes a small-size quantum dot solution.

LED fabrication. The indium tin oxide (ITO)-coated glasses were cleaned using deionized water, ethanol, chloroform, acetone, and isopropanol and subjected to UV–ozone treatment for 15 min. Then PEDOT:PSS was spin-coated onto the ITO glass at 4000 rpm for 40 s, followed by annealing at 140°C for 15 min. The ITO glass is then transferred to the inside of a N₂-filled glovebox. Poly-TPD (8 mg · mL⁻¹ in

chlorobenzene) was spin-coated onto the surface of PEDOT:PSS at 4000 rpm for 40 s followed by annealing at 130°C for 15 min. The quantum dot solution was spin-coated onto the surface of poly-TPD at 3000 rpm for 40 s. The concentration of the three colloidal solutions used for the preparation of the LED devices is 25 mg mL⁻¹. Subsequently, an electron-transporting layer (PO-T2T, ~30 nm) and LiF/Al electrodes (1 nm/ 100 nm) were deposited by thermal evaporation in a vacuum deposition chamber ($\sim 5 \times 10^{-4}$ Pa).

Characterizations. Absorption spectra were measured with a UV-2600 spectrophotometer (Shimadzu). Photoluminescence (PL) spectra were collected on a QE Pro spectrometer (Ocean Optics) with 365 nm excitation. XRD patterns were recorded on a X-ray diffraction (XRD, D8-Advance X-ray diffractometer) with Cu K α radiation ($\lambda = 1.5406$ Å). Transmission electron microscopy (TEM) images were obtained on a JEM-2100 high-resolution transmission electron microscopy (HRTEM) using an accelerating voltage of 200 kV. PL QY and time-resolved PL measurements were measured on a FLS920P Edinburgh spectrometer with 365 nm excitation. Ultraviolet photoelectron spectra (UPS) were collected using the Thermo Fisher Scientific ESCALAB250 XI system. Electrical characteristics and LED performance were recorded in a glovebox filled with N₂. The electrical curves were acquired using a Keithley 2400 System Source Meter, while the light output measurements were made using a fiber optic integration sphere in conjunction with a QE-Pro spectrometer.

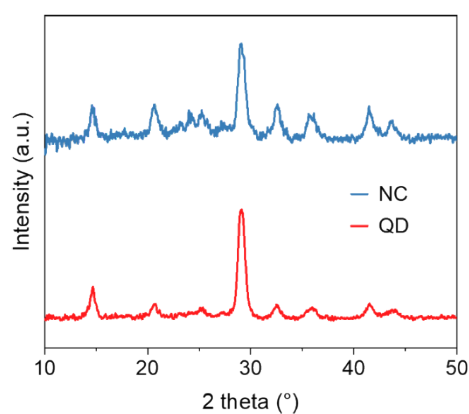


Fig. S1. (color online) XRD patterns for NC and QD films.

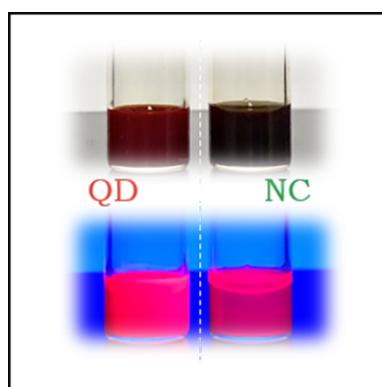


Fig. S2. (color online) Photograph of NC and QD solutions under daylight and ultraviolet light (365 nm) conditions.

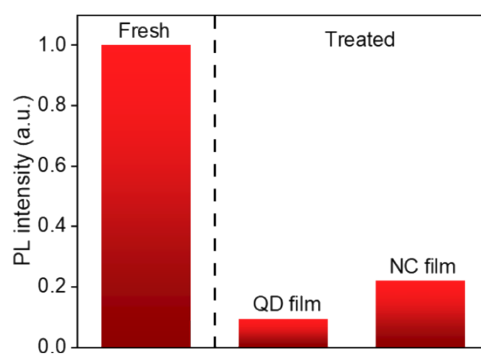


Fig. S3. (color online) Comparison of the PL intensity attenuation of QD and NC films before and after thermal treatment.

	f_1	τ_1 (ns)	f_2	τ_2 (ns)	τ_{ave} (ns)
NC	0.18	34	0.82	127	110
QD	0.35	35	0.65	103	79

Table S1. The fitting of parameters to the time-resolved PL curves of NC and QD.