

## Electronic Supplementary Information

# Highly Efficient CsPbBr<sub>3</sub> Perovskite Nanocrystals Induced by Structure Transformation between CsPbBr<sub>3</sub> and Cs<sub>4</sub>PbBr<sub>6</sub> Phase

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## 1. Experimental section

### 1.1 Chemicals and Reagents.

Cesium carbonate (Cs<sub>2</sub>CO<sub>3</sub>, 99.9%), oleic acid (OA, ≥99%), 1-octadecene (ODE, 90%), oleylamine (OAm, ≥98%), lead bromide (PbBr<sub>2</sub>, 99.99%), zinc bromide (ZnBr<sub>2</sub>, 99.99%) were purchased from Sigma-Aldrich. Toluene (≥95%) and hexane (≥95%) were purchased from Beijing Chemical Works. All the reagents were used without further purification.

### 1.2 Preparation of Cesium Oleate Precursors.

First, Cs<sub>2</sub>CO<sub>3</sub> (0.326 g, 1 mmol), OA (1.27 mL, 4 mmol), and ODE (18.73 mL) were mixed into a 50 mL 3-neck flask. After an hour's stirring under the flowing of Ar at 120°C, this addition was heated to 160°C for Cs<sub>2</sub>CO<sub>3</sub> salt dissolving sufficiently. After about 30 min reaction, Cs<sub>2</sub>CO<sub>3</sub> was fully reacted with OA and the cesium oleate precursors can be prepared. Notably, cesium oleate must be preheated to above 80°C before injection to avoid them separating out of ODE at room temperature.

### 1.3 Synthesis of the perovskite NCs in different molar ratios.

In typical, 0.3 mmol XBr<sub>2</sub> (80% refers to 0.022 g PbBr<sub>2</sub> (0.06 mmol) and 0.054 g ZnBr<sub>2</sub> (0.24 mmol), etc.) and ODE (18 mL) was put into a 50 mL three-neck flask. After a 30-min stirring under the

flowing of Ar at room temperature, this addition was heated to 120°C for another 30 min. Then OAm (2.25 mL) and OA (0.75 mL) were injected into the reaction bottle through a syringe needle. After a few seconds, PbBr<sub>2</sub> and ZnBr<sub>2</sub> could be completely solubilized and come into a clear solution. Then, the temperature was raised to 180°C and the cesium oleate precursor (1.5 mL) was swiftly injected. After a two minutes' reaction time, the reaction mixture was cooled down by an ice-water bath to terminate the growth of the NPs for the next purification process. Furtherly, the acquired NPs were centrifuged with toluene at 9000 rpm (a centrifugal force of 9000 g) for 10 min. After centrifugation, the supernatant was discarded and the precipitations adhered to the bottom of the centrifuge tube were redispersed in hexane. This purification process was repeated twice. After the purification, NPs were put into hexane or chloroform as dispersion.

#### **1.4 Preparation of the Green Light-Emitting Diode based on the fluorescence enhanced CsPbBr<sub>3</sub> PQDs**

ITO substrates were cleaned sequentially in ultrasonic solvent baths of deionized water, acetone and isopropyl alcohol for 15 min. Then, the substrates were blow-dried with nitrogen and then treated with oxygen-plasma for 8 min. PEDOT: PSS was spin-coated onto the ITO substrates at 3000 rpm for 60 s and then baked at 130°C for 20 min. For the perovskite layer, the as-prepared CsPbBr<sub>3</sub> QDs were first dispersed in tetrahydrofuran (THF) at a concentration of 2 mg/mL under continuous stirring for 1 hour. Then, the solution was treated with static settlement over 48 hours in a glove box and consequently, distinct delamination was observed. We took a dip of upper clear part onto the treated substrates and spin-coated at 2000 rpm for 40 s. Then the substrates were transferred to a high vacuum thermal evaporation chamber for the deposition of 30nm TPBi, 0.5 nm LiF and 100 nm Aluminum with the rate of 0.5 Å/s, 0.03 Å/s and 2~5 Å/s.

#### **1.5 Characterization.**

The energy-dispersive X-ray results of the fluorescence enhanced CsPbBr<sub>3</sub> PQDs were obtained using a field emission scanning electron microscopy (FE-SEM, Hitachi, SACS 4800), and the voltage here is 5 kV. The fluorescent lifetime was performed on a fluorescence spectrometer. (Edinburgh Instrument, FLS 980). Nikon microscopy apparatus was applied to take the PL images of the powders. TEM and HRTEM analyses were carried out using a Tecnai-G2-F20 TEM. Absorption and PL spectra were evaluated at indoor temperature by a UV-3101 spectrophotometer and Hitachi F-7000 fluorescence spectrofluorimeter, respectively. X-ray diffraction (XRD) patterns were characterized by D8 Focus X-ray diffractometer purchased from Bruker Company. Current density-voltage and electroluminescence (EL) properties of PeLEDs were measured using a semiconductor

characterization analyzer (Keithley 2634B) and Maya Spectrometer. Luminance-current density values were recorded by using a ST 86LA luminance meter. Results of EQE were obtained from the system of integrating sphere combined with Ocean Optics spectrum-photometer.

### **1.6 Calculation of QY**

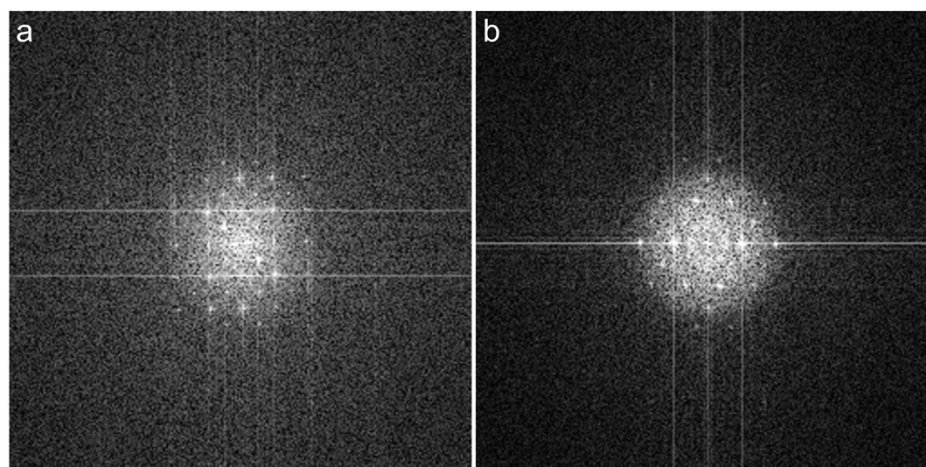
PL QY was estimated by compared the samples with the standard fluorescent dye according to the following formula:  $Q_x=Q_r[A_r/A_x] [I_r/I_x] [n_x^2/n_r^2] [D_x/D_r]$ , where Q refers to the QY, A refers to the absorption intensity at the excitation wavelength, I refers to the intensity of the exciting light, n is the refractive index, D denotes the fluorescent integral area, and the subscripts r and x respectively represent standard and sample, and the excitation wavelength is 360 nm.

## 2. The EDS results of the perovskite NPs samples with different ZnBr<sub>2</sub>.

Sample	Cs (At%)	Pb (At%)	Br (At%)	Zn (At%)
10%	19.86	19.06	60.2	0.88
30%	20.58	18.46	60.09	0.87
50%	27.9	14.31	56.99	0.8
70%	30.42	12.7	57.94	0.74
80%	32.17	8.76	58.38	0.69

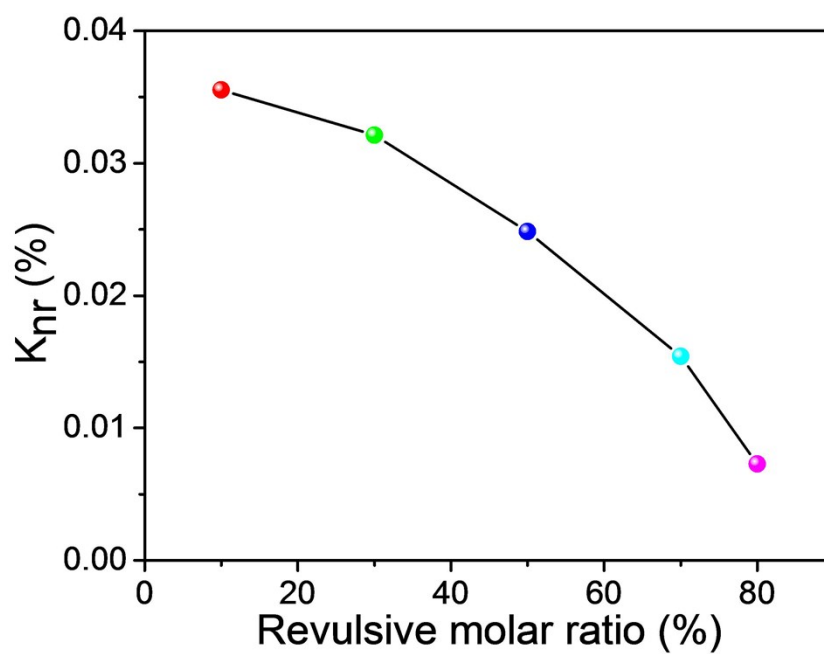
**Table S1** Element composition of Cs, Pb and Br atoms of the perovskite NPs samples with different ZnBr<sub>2</sub> calculated by the EDS results from Figure 2.

## 3. The FFT patterns of perovskite NPs with the amount of 10% and 80% ZnBr<sub>2</sub>.



**Figure S1.** (a)FFT image of 113-structure CsPbBr<sub>3</sub> PQDs with 10% ZnBr<sub>2</sub> revulsive, (b) FFT image of 416-structure Cs<sub>4</sub>PbBr<sub>6</sub> PNCs with 80% ZnBr<sub>2</sub> revulsive.

#### 4. The image of the nonradiative decay rate.



**Figure S2.** the image of the nonradiative decay rate about the perovskite NPs samples with different  $ZnBr_2$ .