

Supporting Information

High-Performance Photodetector based on ZnO/CsPbBr₃ Quantum-dot-level-contact Hybrid Sandwich Structure

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Part I: Experiments and calculations

1. Materials

Cesium carbonate (Cs_2CO_3 , 99%), lead bromide (PbBr_2 , 99.9%), 1-octadecene (ODE, 95%), oleic acid (OA, 85%), oleyl amine (OLA, 85%), octane (C_8H_{18} , 96%), and ethyl acetate ($\text{C}_4\text{H}_8\text{O}_2$, 98%) were purchased from Aladdin Reagent Company. Chloroform (CHCl_3 , 99.9%) was purchased from Fisher Chemical. Zinc acetate dihydrate ($\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$, 99%), anhydrous ethanol ($\text{C}_2\text{H}_6\text{O}$, 99.7%), potassium hydroxide (KOH, 85%) and n-hexane (C_6H_{14} , 99%) were all purchased from China National Pharmaceutical Group Chemical Reagent Co., Ltd (purity AR). All chemicals were used as received without further purification.

2. Material synthesis

2.1 Preparation of CsPbBr_3 quantum dots

In a typical synthesis of CsPbBr_3 quantum dots. Firstly, Cs_2CO_3 (0.407g), ODE (20 ml), and OA (1.25 ml) were added a 50 ml three-necked-flask, dry with N_2 at 120 °C for one hour, heat to 150 °C and keep for 30 minutes until the mixture turns into a transparent Cs-oleate solution, the solution is kept at 120 °C under N_2 for subsequent experiments. And then add PbBr_2 (0.752 mmol, 0.276 g) and octadecene (ODE, 20 mL) to a 50 ml three necked flask, dry with N_2 at 120 °C for one hour, add oleic acid (OA, 2 mL) and oleyl amine (OLA, 2 mL), and heat up to 150 °C for one hour under N_2 atmosphere. Subsequently, the Cs-oleate solution of 1.6ml was rapidly inject into the reaction system, and after 5 seconds, cool the reaction mixture with an ice water bath. To remove excess surface ligands, ethyl acetate (EA) was added to the crude solution

(crude solution: EA=1:3 volume ratio) and centrifuged at 10000 rpm for 10 minutes. Repeat the purification procedure once. Finally, the precipitate was dispersed in 4ml of octane solution to obtain a clear solution.

2.2 Preparation of ZnO quantum dots

ZnO quantum dots were synthesized by classical wet-chemical method. Firstly, zinc acetate dihydrate was added 35 ml anhydrous ethanol and stir under reflux for 2 hours on an 80 °C digital intelligent temperature controlled magnetic stirrer, store it in the refrigerator at 0-4 °C. And then heat 10 ml of precursor solution of zinc acetate on a digital intelligent temperature controlled magnetic stirrer to 50 °C using three-necked-flask, the newly prepared anhydrous ethanol solution of KOH with concentrations of 0.45 M, 1 M, and 2 M were rapidly inject into the reaction solution through a neck on the side of the flask using a glass syringe. After 3 minutes of reaction, immediately cool the reaction solution to room temperature using an ice water bath. Subsequently, add excess n-hexane to the reaction solution and centrifuge to precipitate. Remove the centrifuge supernatant and dissolve the precipitate with an appropriate amount of anhydrous ethanol, repeat this process twice. Finally, the product is dissolved in chloroform.

3. Characterization

The X-ray diffraction (XRD) patterns of the CsPbBr₃ QDs film and ZnO-x:CsPbBr₃ nanocomposite films were characterized using the XRD instrument (XRD-6100, Shimadzu, Japan). The scanning electron microscope (SEM) images were recorded by an electron microscope (JEOL, JSM-7800F, Japan). The transmission

electron microscopy (TEM) images were obtained on the JEOL-JEM 2100 F microscope in Japan. Absorption spectra was recorded by a UV–VIS–NIR spectrophotometer (UV-3600, Shimadzu, Japan). Photoluminescence (PL) spectra and Time-resolved photoluminescence (TRPL) of the films were obtained by a fluorescence spectrophotometer (Cary Eclipse, Agilent, USA). Current-voltage (I–V) characteristics and photo-response curves were measured using the Keithley 4200 electrometer. The time resolution of the platform for current-time (I-T) measurement is about 7.85 ms. Incident light was provided by the blue laser with a center wavelength of 450 nm. All tests were performed in air and at room temperature.

4. Calculations

In order to evaluate the photoresponse performance of ZnO-1/P-ZnO-1/P photodetectors, four key parameters, namely on/off ratio, R, EQE and D*, were calculated. The on/off ratio is the ratio of photocurrent to dark current. R represents the ability of the photodetector to convert light signals into electrical signals. EQE is the ratio of the number of photogenerated electrons to the number of incident photons. And D* represents the sensitivity of the device to weak light signals. The above four key parameters can be calculated using the following formula:

$$\text{on/off ratio} = \frac{I_p}{I_d} \quad (1)$$

$$R = \frac{J_p - J_d}{P_i n} \quad (2)$$

$$\text{EQE} = \frac{R h c}{e \lambda} \quad (3)$$

$$D^* = \frac{R}{\sqrt{2} J_d} \quad (4)$$

In the formula, I_p is the photocurrent, I_d is the dark current, J_p is the photocurrent density, J_d is the dark current density, P_{in} is the illumination power density. h is Planck's constant, c is the speed of light, λ is the wavelength of incident light, and e is the fundamental charge.

Part II: Supplementary Table and Figures

Table S1. Summarization of decay lifetime for each material in TRPL measurements.

CsPbBr₃ QDs film and P-ZnO-x nanocomposite films with different-sizes ZnO QDs.

Using multi-exponential decay fitting: $\tau_{avg} = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)$

Table S1

	CsPbBr ₃	P-ZnO- 0.45	P-ZnO-1	P-ZnO-2
τ_1 (ns)	1.003	1.470	1.530	1.098
τ_2 (ns)	4.648	6.303	8.210	6.525
A_1 (%)	6981.429	253.223	324.988	2876.642
A_2 (%)	0.336	0.658	0.520	0.672
τ_{avg} (ns)	1.004	1.523	1.587	1.106

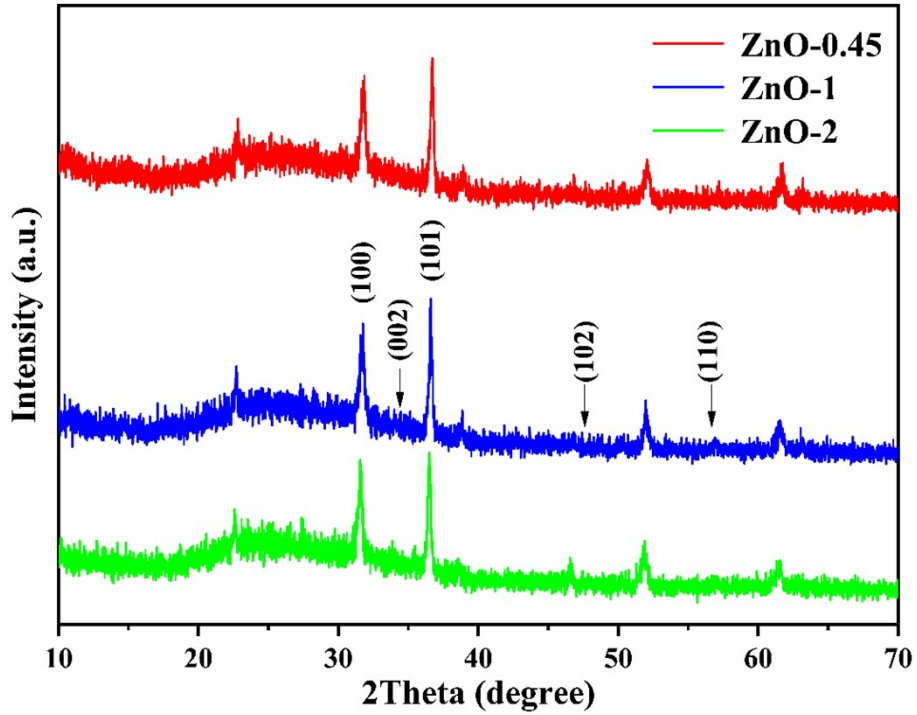


Figure S1. XRD patterns of ZnO-x.

As shown in Figure S1, all XRD patterns of ZnO-x have five diffraction peaks at 31.8° , 34.5° , 36.3° , 47.5° and 56.7° , corresponding to the ZnO hexagonal wurtzite structure on the (100) (002) (101) (102) and (110) crystallographic planes, respectively.

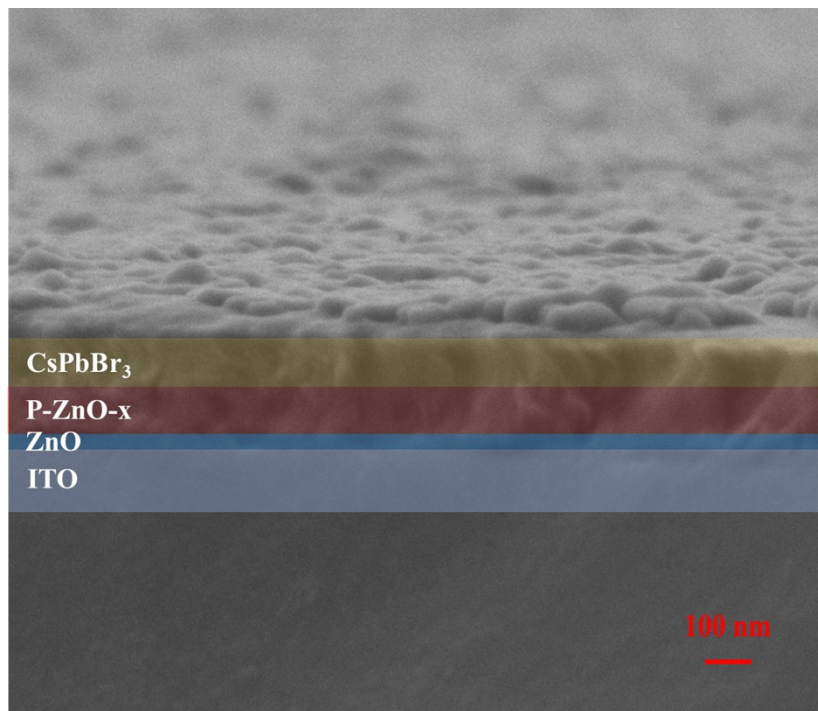


Figure S2. Cross-sectional SEM image of ZnO-1/P-ZnO-1/P photodetector.

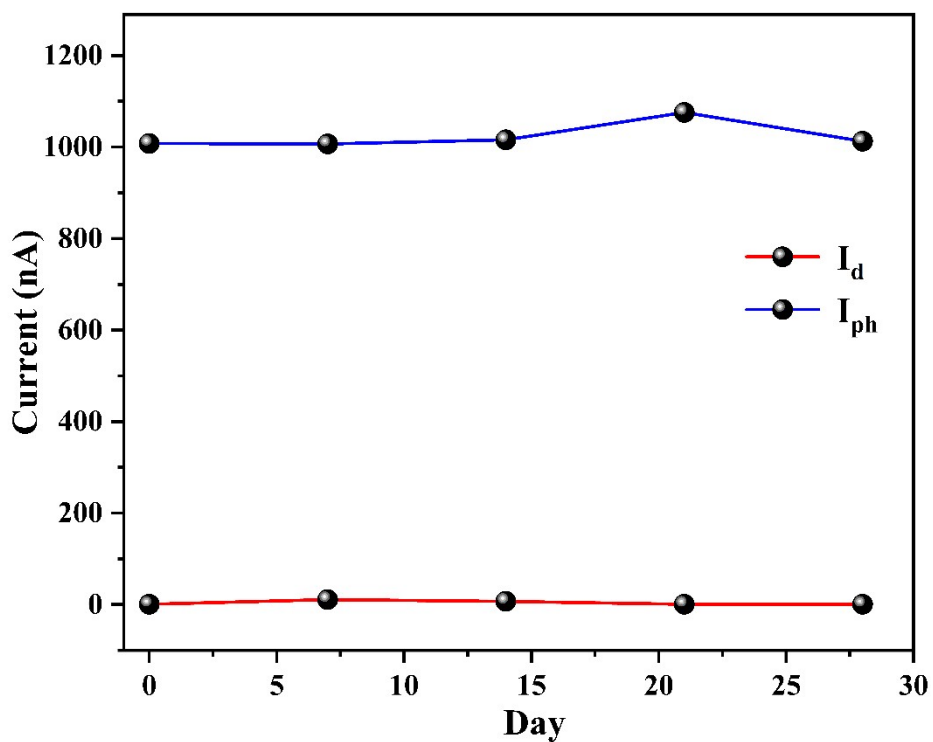


Figure S3. The long-term stability of the ZnO-1/P-ZnO-1/P photodetector over 1 months.

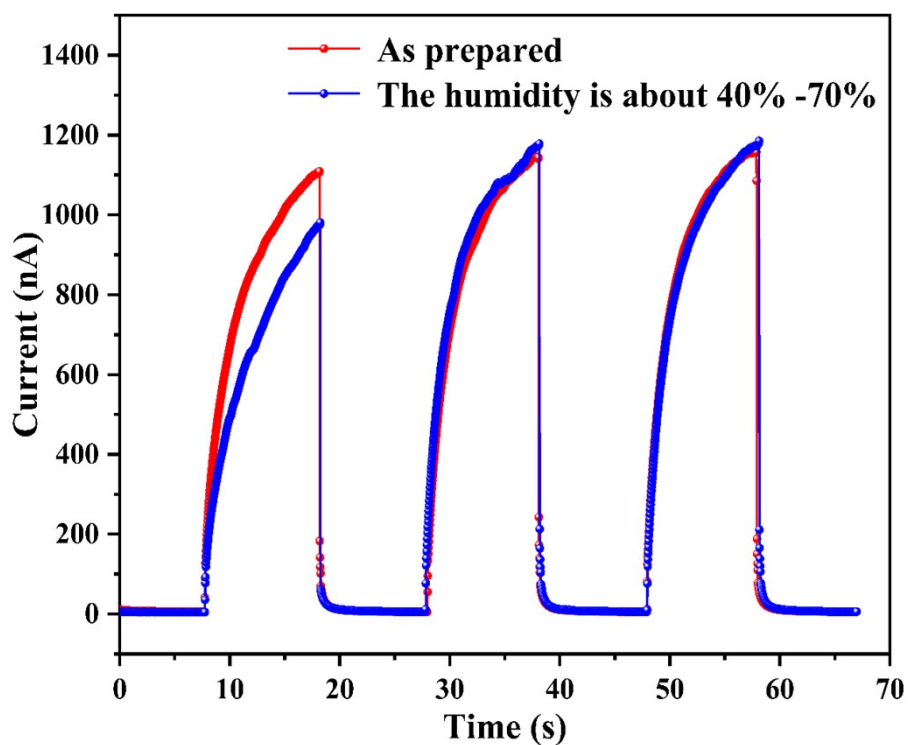


Figure S4. Photoresponse curve of ZnO-1/P-ZnO-1/P photodetector after storage for one month at a humidity of about 40% -70%.