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High-sensitivity broadband colloidal quantum dot heterojunction photodetector for night-sky radiation

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ABSTRACT

Being with excellent photosensibility, bandgap tunability and solution processibility, lead sulfide (PbS) colloidal quantum dot (CQD) gets one of the most attracting nanomaterials for photodetectors and light-harvesting devices. Usually short ligands are used to passivate the surface of CQDs to enhance the carrier mobility in QD films, and different types of ligands show distinct influence on the physical/electrical properties on the passivated CQD films. In this paper, a high-sensitivity broadband photodetector based on PbS-EDT/PbS-TBAI heterojunction by band-alignment engineering was presented. After optimizing the thickness of PbS CQD films, we found that the maximum specific detectivity of the photodiode ITO/ZnO(40 nm)/PbS-TBAI(160 nm)/PbS-EDT(40 nm)/Au reached to 3.93×10^{13} Jones and 5.52×10^{13} Jones under 2.0 μW/cm^2 910 nm and 1.9 μW/cm^2 500 nm illuminations, respectively, showing excellent stability in air and high sensitivity to weak signal (i.e. night-sky radiation). Further, the device performance of photodiode ITO/ZnO/PbS-TBAI/PbS-EDT/Au is compared with that of the control device ITO/ZnO/PbS-TBAI/Au, and the underneath mechanism for the enhanced performance is discussed.

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1. Introduction

In the never-ending quest of novel materials and nanoscale structures for optoelectronic devices such as solar cells [1,2], light-emitting diodes [3,4] and photodetectors [5,6], semiconductor colloidal quantum dots (CQDs) have attracted much attention for their combined advantages of photosensibility and solution processability. The commonly used II-VI and III-V semiconductors, such as $Hg_{1-x}Cd_xTe$ and GaAs, can cover infrared region above 3000 nm but they have to be fabricated by the expensive epitaxial growth. On the other hand, Si-based photodetectors barely reach spectrum above 1000 nm [7]. Possessing the excellent light absorption features in the UV, VIS and NIR spectrum, and the multiple exciton generation (MEG) effect, narrow-bandgap IV-VI CQDs (e.g. PbS and PbSe) become the ideal candidates for solar cells and photodetectors [8–10].

As an IV-VI semiconductor, PbS exhibits a large exciton Bohr radius (18 nm) and a narrow tunable direct bandgap energy (0.41 eV @ 300 K) [11,12], which ensures its competence for photodetection in visible and near-IR regions. Due to the large surface-area-to-volume ratio of PbS CQDs, the atoms on the PbS CQDs surface mainly dominate their physical and electrical properties, especially, their carrier mobility and their conductivity [13].

Therefore, in this paper, we focus on the active surface energy band engineering by combining the application of organic-inorganic hybrid ligands, forming heterojunction between tetrabutylammonium iodide (TBAI)-ligated PbS CQD (i.e. PbS-TBAI) film and 1,2-ethanedithiol (EDT)-ligated PbS CQD (i.e. PbS-EDT) film. After optimizing the thickness of PbS-TBAI and PbS-EDT, a remarkable improvement of device performance was obtained from ITO/ZnO/PbS-TBAI/PbS-EDT/Au with responsivity (R) and specific detectivity (D^*) of 385 A/W and 3.90×10^{13} Jones under 2 μW/cm^2 910 nm illumination, 444 A/W and 4.52×10^{13} Jones under 1.9 μW/cm^2 500 nm illumination, respectively. The devices also showed good air-stability and time-responsibility. After 30 days of storing in air, no evident decrements were observed for the photocurrent.

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2. Experimental section

2.1. Materials

Lead oxide yellow (PbO, 99.9%), Oleic acid (OA, 90%), 1-octadecene (ODE, 90%), bis(trimethylsilyl) sulfide ($(TMS)_2S$, analytical reagent), 1,2-ethanedithiol (EDT, >98%) and tetrabutylammonium iodide (TBAI, >98%) were purchased from Sigma-Aldrich. Hexane (99.9%), acetone (99.98%), methanol (99.99%), and toluene (98%) were purchased from the Beijing Chemical Works. All chemicals were used as received.

2.2. PbS CQDs synthesis

PbS CQDs were synthesized following the procedure developed by Hines and Scholes [14] and performed under inert conditions with standard Schlenk-line techniques.

2.3. ZnO film synthesis

ZnO films were synthesized following the method reported by White et al. [15].

2.4. Device fabrication and characterization

The pre-patterned ITO glass substrate was cleaned in ultrasonic bath by de-ionized water, isopropyl alcohol, acetone and isopropanol for 15 min sequentially, and then it was dried with N_2. The ZnO film was deposited on the substrate by spin-coating for 60 s at speed of 3000 rpm, after that the annealing process was applied at the 280 °C for 2 h.

The optimized multilayer CQD photodiode consists of 8 layers of PbS-TBAI CQD films and 2 layers of PbS-EDT CQD films, with a total thickness of about 200 ± 20 nm. The devices were stored in air for ten hours and then transferred into nitrogen glove box for Au anode evaporation to complete the device fabrication. The thermal evaporation of Au electrodes was performed at an evaporation rate of 0.1–0.3 nm/s in a vacuum of 4×10^{-4} Pa, reaching a thickness of ~100 nm. The PbS-TBAI were deposited for different times (i.e. 6, 8, 10, 12 times) to fabricate the active layer in different thicknesses.

The absorption spectra of the samples were measured with an ultraviolet–visible–near infrared (UV–vis–NIR) spectrophotometer (Shimadzu, UV-3600). The thickness of the film was measured by an XP-2 high resolution surface profiler. All electrical and optical characteristics of the devices were measured with a Keithley semiconductor characterization system (Model 4200-SCS) in dark and under illumination. The transmission electron microscopy (TEM) images of the CQDs were taken with FEI Tecnai G^2 F20 S-TWIN. The X-ray diffraction (XRD) pattern of the CQDs was characterized by Rigaku RM-2000. The external quantum efficiency (EQE) was performed using Oriel IQE-200TM. And Newport 6279 NS Solar Simulator was used as the illumination light source. All the characterization of the devices was performed at room temperature in air.

3. Results and discussion

PbS CQDs were obtained by following the hot injection method developed by Hines and Scholes [14] after optimizing the synthesis conditions. Fig. 1(a) shows TEM image of the as-synthesized PbS CQDs, certifying the high quality of nanoparticles with a size of ~4 nm in average. Fig. 1(b) shows the exciton absorption peak in the mono-sized CQDs in n-hexane solution, and its photoluminescence under the excitation wavelength of 325 nm, demonstrating its quantum confinement properties. Fig. 1(c) shows the XRD patterns

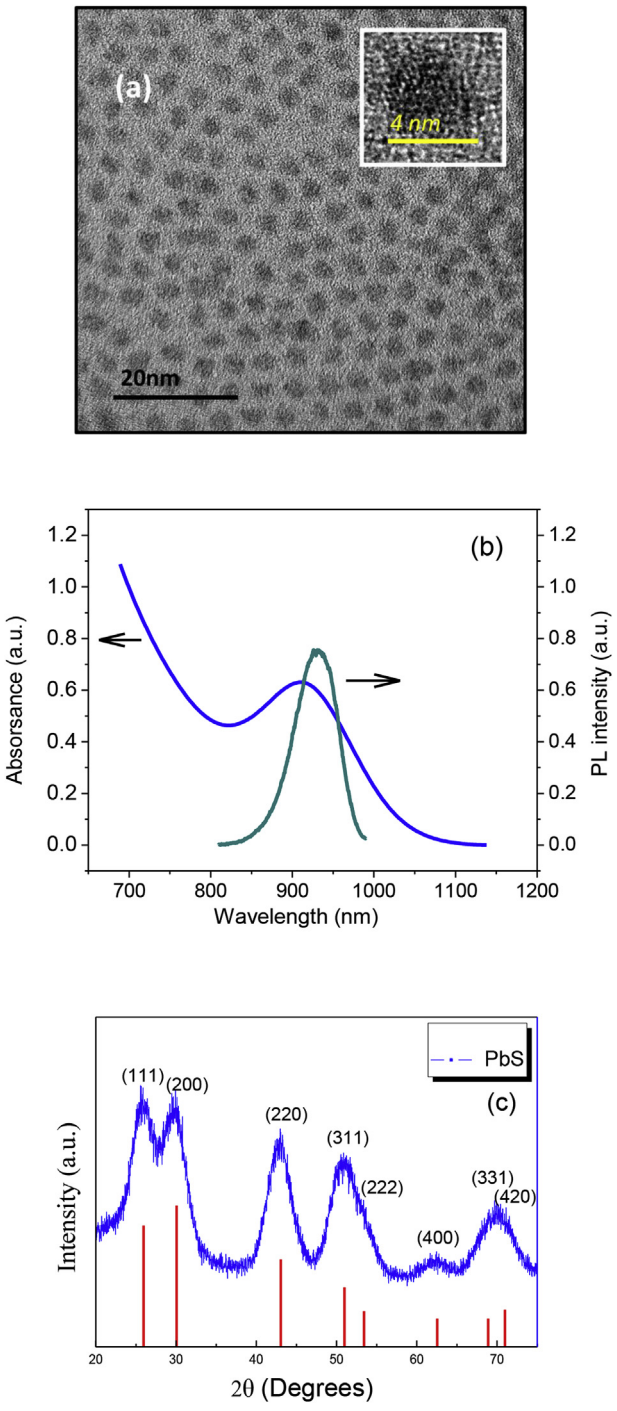

Fig. 1. (a) TEM image of the as-synthesized PbS CQD, the inset shows its HR-TEM image, (b) absorption and PL spectra of PbS CQDs in n-hexane solution, and (c) XRD patterns of PbS nanocrystals.

of the PbS CQDs, one can see that PbS CQDs is really in nanometer scale.

Surface passivation has to be performed for CQD based devices, and many achievements have been reported in this field [16–18]. In our experiments, ligand exchange is performed by using TBAI to replace the insulting long alkyl chains (e.g. oleic acid) used for the synthesis of CQDs, to increase the interdot carrier transportability [19–22], and to reduce defect density [23–25].

The schematic of photodiode ITO/ZnO/PbS-TBAI/PbS-EDT/Au are

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