SUPPORTING INFORMATION

Single Step Deposition of an Interacting Layer of Perovskite Matrix with Embedded Quantum Dots.

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Experimental Section:

TiO_2 compact layer: SnO_2:F (FTO) substrates were cleaned with soap, sonicated in distilled water, ethanol and a mixture of acetone/isopropanol (30:70 v/v ratio) for 15minutes and then treated with aUV−O_3lamp for 15minutes. The TiO_2 compact layer was deposited by spray pyrolysis at 450°C, consuming 28ml of a solution of titanium di-isopropoxide bis(acetylacetonate, 75% vol. 2-propanol) in absolute ethanol (1:39, v/v ratio); the films were annealed at 450°C for 30 minutes.

PbS/CdS QD synthesis and Ligand exchange: Core/shell QDs were synthesized according to procedure previously reported.[26]Briefly, for the synthesis of 2.3-nm PbS nanocrystals, a three-necked round-bottom flask containing0.9 g (4 mmol) of PbO, 2.0 g (7 mmol) of Oleic acid and 36 ml of 1-octadecene (ODE) was heated up to 150^{o}C under N_2 to form Pb-oleate moieties. The solution was degassed for 30 minutes under vacuum, and 3 ml (6.7 mmol)of trioctylphosphine (90%) was injected. Then the temperature was reduced to 110^{o}C and a mixture of 0.42 ml (2 mmol) of hexamethyldisilathiane (HMDS) and 4 ml ODE was quickly injected to the flask. The temperature of solution was left to decrease to room temperature. The reaction product was cleaned three times with ethanol/acetone (1:1, v/v), centrifuged (3000 rpm for 10 minutes) and dispersed in toluene (100 mg/ml).

For the growth of CdS shell, 0.44 g (3.4 mmol) of CdO and 2.4 g (8.5 mmol) ofOA and 40 ml of ODE was heated at 220°C under air to dissolve CdO, cooled down to 150°C and then degassed for 1 hour under vacuum and the temperature was then reduced to 70°C. Next, 5 ml of as-prepared PbS QDs (100 mg/ml) was rapidly injected and after 5 minutes of reaction at 70°C, the temperature was decreased by adding the nonsolvent mixture (ethanol/acetone, 1:1, v/v). The product was washed following the same washing steps as for PbS nanocrystals.

For the synthesis of 3-nm PbS/CdS core/shell QDs, an identical procedure was applied for the synthesis of 2.3 nm QDs, however 2.7 g (9.6 mmol)of OA was used for PbS formation and 0.34 g(2.6 mmol) of CdO and 1.85 g (6.5 mmol)of OA were used for the growth of CdS shell.

For ligand exchange, 1 ml of QDs (PbS or PbS/CdS core/shell) dispersed in octane (10mg/ml) was added into 1 ml of dimethylformamide (DMF) solution containing 117 mg of $PbI_2$and 50 mg of MAI (CH_3NH_3I).[13] After stirring for 30 minutes, QDs had transferred from the top octane phase to the bottom DMF, see Fig. 1a. After removing the octane, the QD solution was washed three more times using octane to remove the organic residue. Subsequently, the QDs were precipitated by the addition of toluene. After removing all the liquid, QDs were dried under vacuum for several hours then dispersed in the solution of PS.

Perovskite solution: 622 mg (1.35 mmol) of PbI_2 were added in the solution containing 1ml of DMF and 95 µl of dimethyl sulfoxide (DMSO). This solution was heated at 65°C to dissolve PbI_2, then cooled down to room temperature. Subsequently, the PbI_2 solution was poured into a vialcontaining 215 mg (1.35 mmol) of MAI to obtain the final solution for the perovskite deposition.[27]

Perovskite-QDs solution: QDs ligand exchanged were dispersed in the solution of PS with the concentration varying from 200 to 400 mg/ml.

Film deposition: PS and PS-QDs layers were spin coated on TiO_2substrates from the corresponding PS and PS-QDs solutions, at different spin rates to obtain different layer thicknesses. Diethylether was added to the film when the spin coater was running. PS and PS-QDs films were annealed during1 minute at 65°Cand then,2 minutes at 100°C.

Spiro OMeTAD and Gold: Spiro OMeTAD solution was prepared by dissolving 72.3 mg of spiro-OMeTAD(2,2′,7,7′-tetrakis(N,N-di-p-methoxyphenylamine)-9,9-spirobifluorene) in 1 ml of chlorobenzene, then mixed with 28.8 μl of 4-tert-butylpyridine and 17.5 μl of a stock Li^+ solution (which contained 520 mg/ml bis-trifluoromethylsulfonylamine lithium salt in acetonitrile). Spiro OMeTAD layer was spin coated on PS or PS-QDs films at 4000 rpm for 30s. Finally, 60 nm of gold were thermally evaporated in an ultrahigh vacuum chamber on top of Spiro OMeTAD layer to make complete devices.[6]

Characterization of samples in solution, on film, and devices: The morphology of the films was analyzed by scanning electron microscopy (SEM) using a JSM7001F (Field emission scanning electron microscope).The absorbance and photoluminescence (PL) spectra of the QDs in solution, before and after ligand exchange, were obtained by using a spectrophotometer based on a CCD detector (Andor-iDUSDV420A-OE) coupled with a spectrograph as a diffraction grating (Newport 77400). A commercial continuous laser diode (650 nm, 5 mW) was used as an excitation source. Photoluminescence (PL) experiments for films were carried out by pumping the surface of the samples with a 533 nm DSPP (diode pumped solid state) laser and a 780 nm semiconductor diode. Back scattered PL was then collected with the aid of a microscope objective and dispersed in a Ocean Optics HR4800 and NIRQUEST512 spectrographs at visible (400-1000 nm) and infrared wavelengths (900-1700 nm), respectively. EL experiments were performed by applying an electric field in the PS-QD layer integrated in a diode configuration and collecting the emission of the film with a similar set-up (objective lens and spectrograph). Measurements have been carried out using a non-sealed sample holder. Sample holder has a gas connection to flowN_2continuously during the EL measurements.

Quantum yield calculation. QY in solution was calculated from the equation below:

$$QY_s = QY_{Ref}\frac{Abs_{ref}\, I_s}{Abs_s\, I_{ref}}\left(\frac{n_s}{n_{ref}}\right)^2$$

In which, *s* and *ref* mean sample and reference respectively. *Abs* is the value of absorbance at 650 nm, and *I* is the integrated PL of solution which is excited by 650 nm – laser diode. *n* is the refractive index of solvent.

Commercial Indocyanine green (reference) and original QDs (capped with OA ligand) were dispersed in methanol and toluene, respectively. After ligand exchange, PS capped QDs were dispersed in DFM without PS precursors (QDs_PS solution) and with PS precursors with the concentration varying from 200 to 400 mg/ml (PS-QDs solutions). Very low volume of each solution was taken and diluted in its corresponding solvent to obtain the value of absorbance close to 0.1 at 650 nm. PL spectra was measured using a commercial continuous laser diode (650 nm, 5 mW).

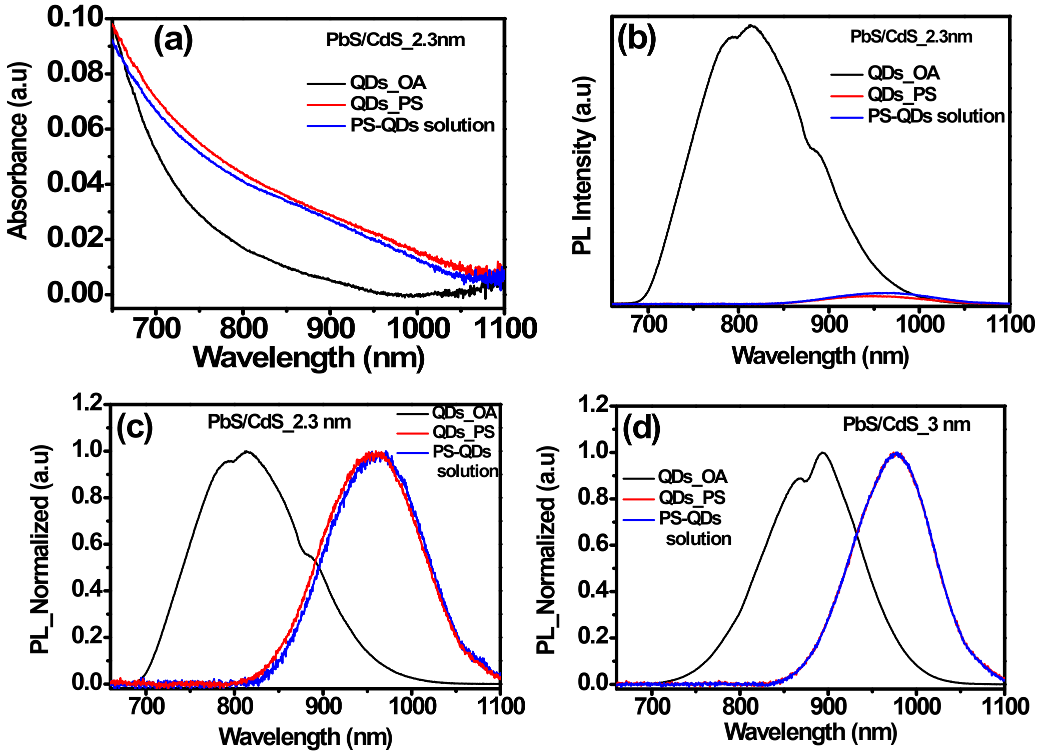

Fig S1. Solution absorbance**(a)** and Photoluminescence (PL) spectra**(b)**forPbS/CdS core/shell QDs2.3 nm diameter size.Normalized PL for PbS/CdS core/shell QDs of 2.3 nm **(c)** and 3 nm size**(d)**.Spectra before (OA capped) and after ligand exchange with perovskite precursors (PS capped) and of the final PS-QDs solutions are plotted (λ_{excited} = 650 nm).

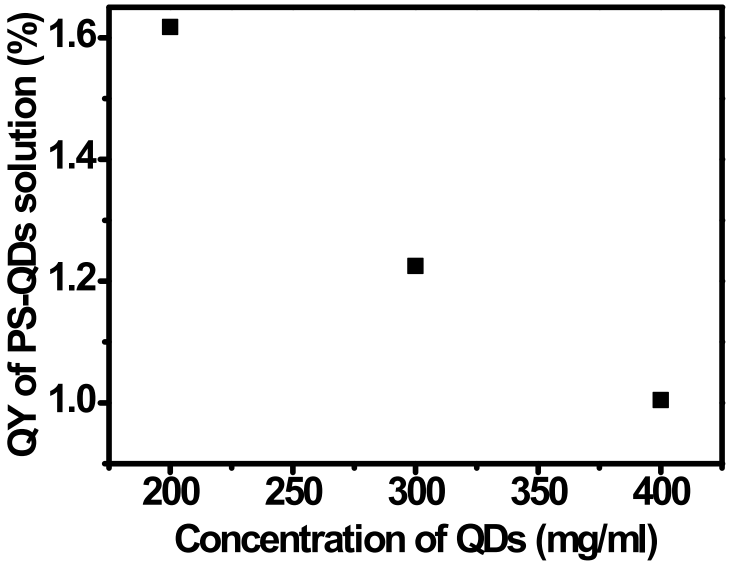

Fig S2. Quantum Yield of perovskite solution containing different QD concentration.

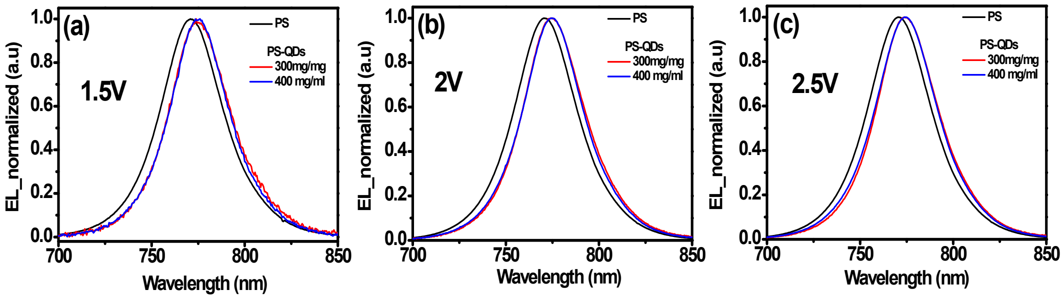

Fig S3. Normalized EL at PS emission of LED devices without QDs (PS)and with PbS/CdS core/shell 2.3 nm QDs (PS-QDs) with the concentration of 300 mg/ml and 400 mg/ml at different applied bias. PS and PS-QDs films were spin coated at 9000 rpm and annealed at 65°C for 1 minute then at 100°C during 2 minutes.

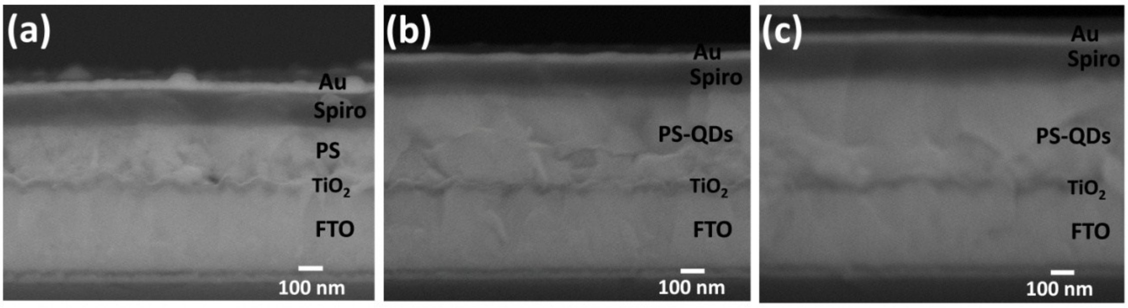

Fig S4. Cross section SEM images from backscattered electrons of LED devices **(a)**without QDs and with different concentrations of PbS/CdS core/shell QDs 2.3 nm: **(b)**300 mg/ml and **(c)**400 mg/ml. Concentration of perovskite precursors was kept constant, all PS and PS-QDs films were spin coated at 9000 rpm and annealed at 65°C in 1 minute then at 100°C in 2 minutes

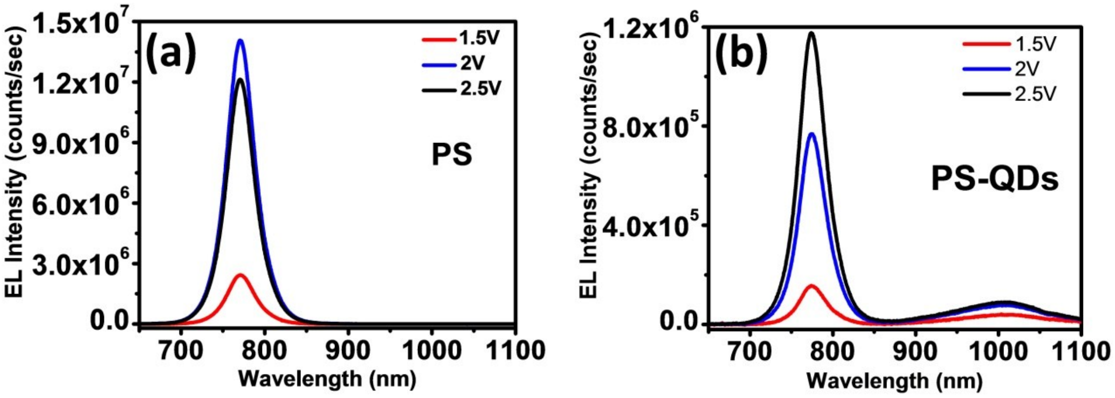

Fig S5. Electroluminescence of LED devices **(a)**without QDs **(b)** and with PbS/CdS core/shell 2.3 nm QDs with the concentration of 300 mg/ml at different applied bias. PS and PS-QDs films were spin coated at 9000 rpm and annealed at 65°C for 1 minute then at 100°C during 2 minutes.

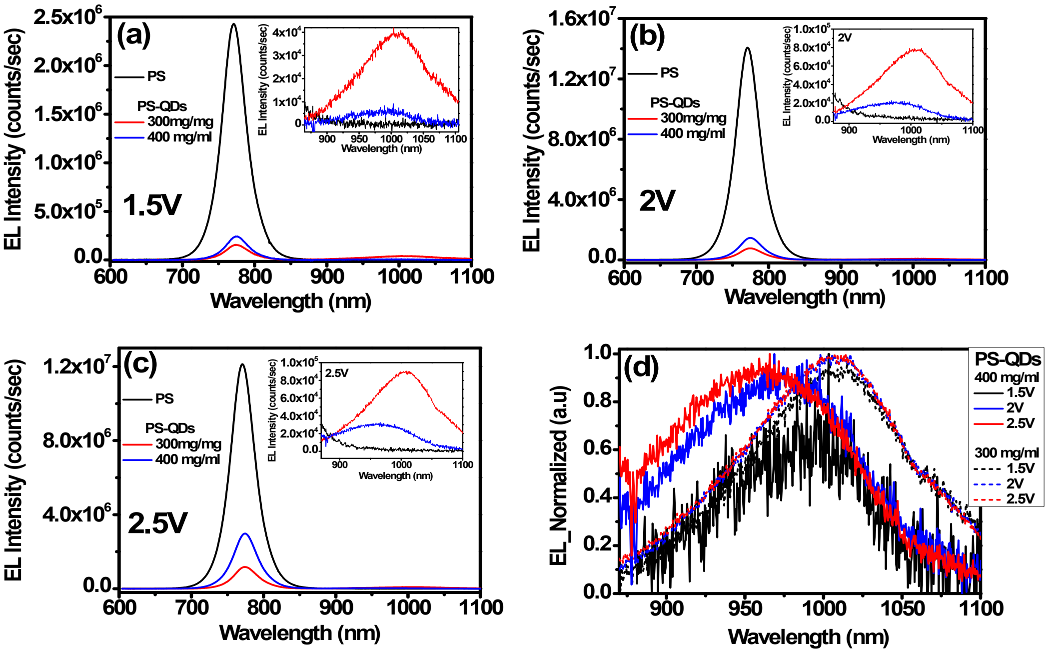

Fig S6. Electroluminescence of LED devices based on $CH_3NH_3PbI_3$ perovskite and PS-QDs absorbers at different applied voltages**(a, b, c)** and normalized EL intensity of QD emission signal **(d)**. In which PS capped QDs were dispersed in PS solution with concentration of 300 and 400 mg/ml.PS and PS-QDs films were spin coated at 9000 rpm and annealed at 65°C for 1 minute then at 100°C during 2 minutes.

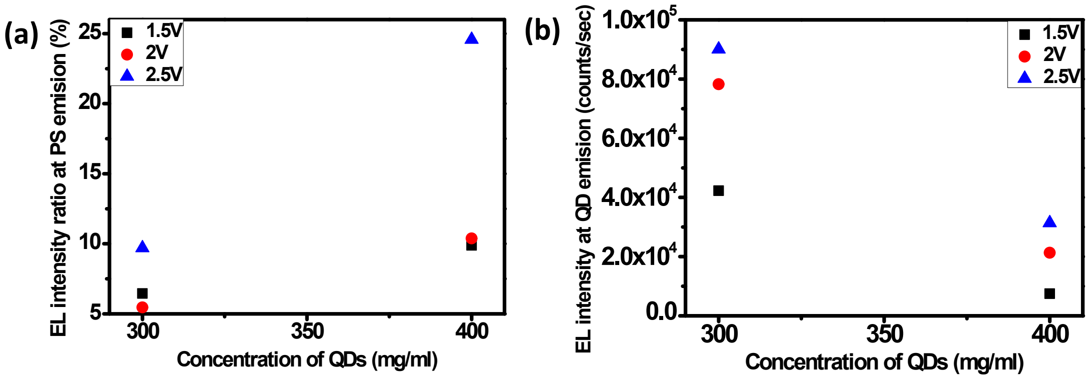

Fig S7.(a) The EL intensity ratio at the PS emission maximum of PS-QDs samples versus reference samples(without embedded QDs), whose EL intensity was normalized to 100%. **(b)** EL intensity of PS-QD devices at the emission maximum of QDs as a function of QD concentration.

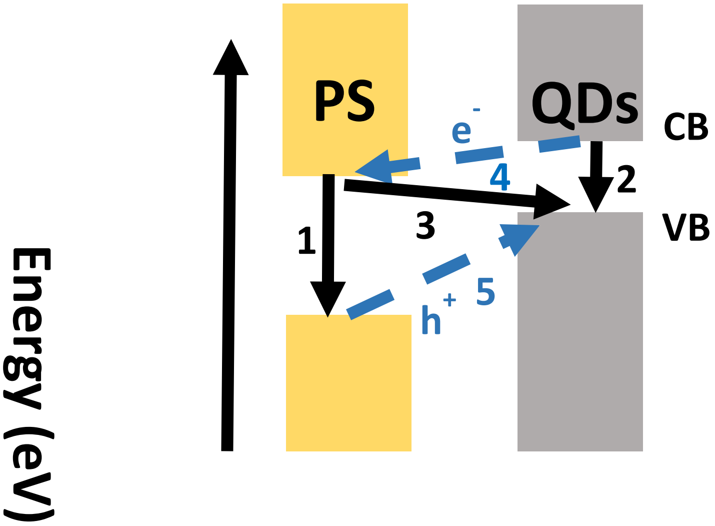

Fig S8. Proposed relative energy diagram. Solid arrows indicate the emission and dotted arrows indicate the charge carrier transfers.

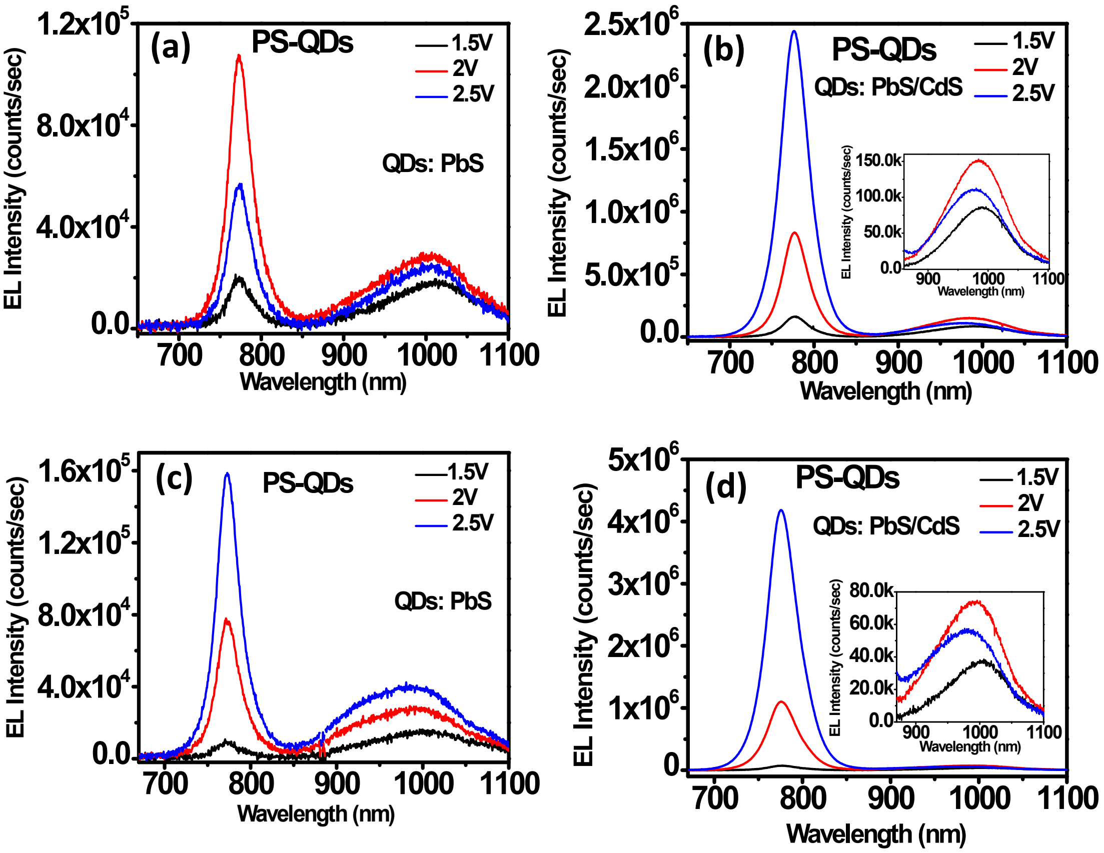

Fig S9. Electroluminescence PS-QDs devices at different applied voltages in **(a, c)** withPbS core 2.3nm QDs and **(b, d)**PbS/CdS core/shell 2.3 nm QDs embedded.**(a, b)** QD concentration of 300 mg/ml and **(c, d)**400 mg/ml. All the PS-QDs films were spin coated at 5000 rpm and annealed at 65°C for 1 minute then at 100°C during 2 minutes.