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Supporting Information

Luminescent Down-Shifting Enables UV-Stable and Efficient ZnO Nanowire-Based PbS Quantum dot Solar Cells with J_{SC} Exceeding 33 mA cm^{-2}

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S1. Absorption and PL spectra for PbS QDs and PL of CdSe/ZnS QDs

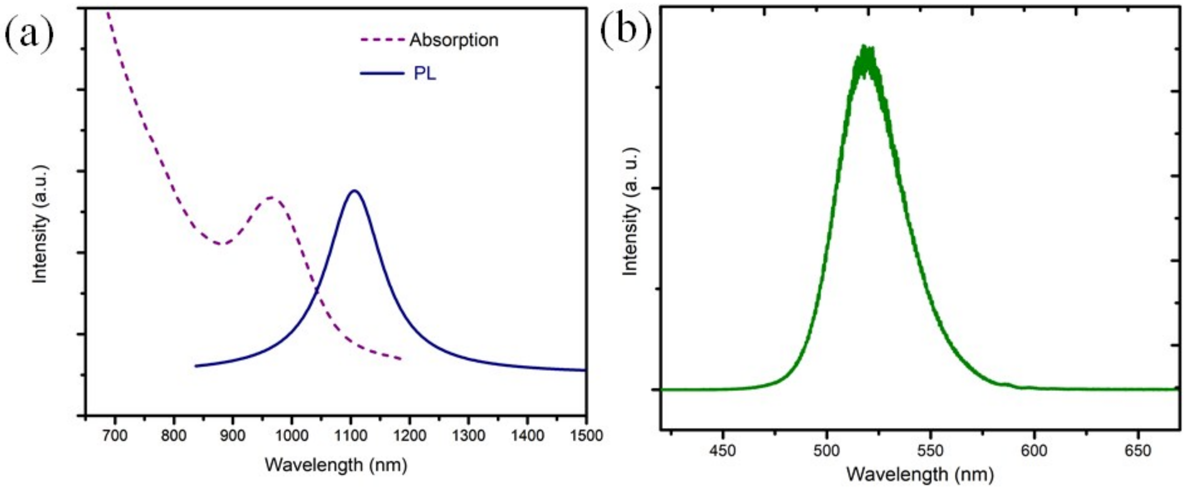


Fig. S1. a) Absorption and PL spectra for PbS QDs with first absoprtion peak at 950 nm. b) PL of CdSe/ZnS QDs.

S2. Impact of UV exposure on PL and absorption spectra of PbS/TBAI films with and without CdSe/ZnS QDs UV protective layer

PL measurements were performed on films: PbS/TBAI before (reference) and after UV irradiation, and PbS/TBAI with CdSe/ZnS QDs protective layer on the back of the glass substrate after UV exposure. As shown in Fig. S2 (a), compared to the reference sample, which was not exposed to UV irradiation, the PL peaks of the other two samples redshifted ~11 nm and ~25 nm upon UV illumination for the PbS/TBAI film with and without CdSe/ZnS QDs protective layer, respectively. Together with the XPS results (Fig. 3b) that suggests the removal of Pb-I bonds, we speculate that the ligand damage upon UV irradiation is likely due to decreasing the interdot spacing. The full width half maximum (FWHM) for the PbS/TBAI film with QDs coating is 121 nm, which is very similar to that of reference sample (119 nm). However, the FWHM for the film without QDs coating is ~129 nm, which is likely due to increased non-radiative recombinations in QD surface states as a result of reduced number of surface-passivating ligands. Moreover, a relatively weak and broad emission with the peak position at around ~1455 nm is observed which can be attributed to defects [1]. The intensity of the defect peak is increased for the thin film without CdSe/ZnS QDs coating compared to the sample with CdSe/ZnS QDs coating. In addition, for the reference sample (no UV exposure) we could not detect the peak under the same measurement settings used for other samples. We attribute this to the effective surface passivation with short TBAI ligands for the reference device.

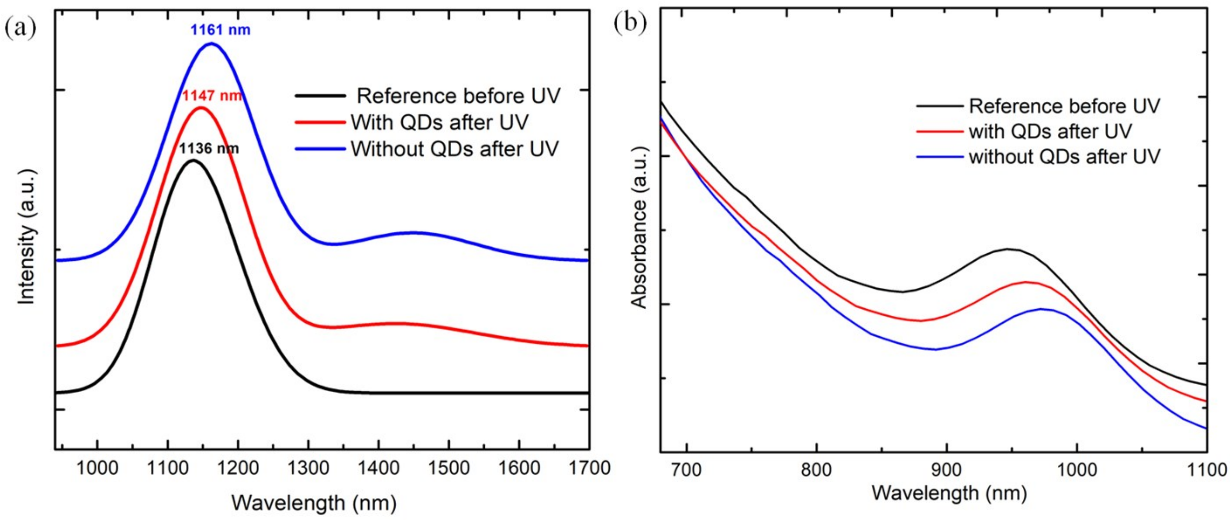


Fig. S2. a) PL spectra of PbS/TBAI films with and without CdSe/ZnS QDs UV protective layer before and after UV exposure. The relatively weak and broad peak at ~1450 nm observed in UV-exposed films is related to defect peaks. The intensity of the defect peak is less for the device with CdSe/ZnS QDs coating, suggestive of UV protection role of the coating. b) Absorption spectra of PbS/TBAI films with and without CdSe/ZnS QDs UV protective layer before and after UV exposure. The red shift of the spectra suggests reduced bandgap upon UV exposure. After UV exposure, the device with CdSe/ZnS QDs coating shows less red shift compared to the reference device.

S3. PV performance of PbS QDs PV with ZnO NWs

Table S3. PV performance metrics of solar cells with ZnO NWs and different concentrations of the energy down shifting QDs.The mean and standard deviations are reported for six devices on each set

CdSe/Zn QDs concentration of NW PV	V_{OC} **(mV)**	J_{SC} **(mA/cm^2)**	**FF (%)**	**PCE (%)**

Without QDs coating	585±3	31.1±0.3	0.55±0.01	9.8±0.2
20 mg/mL	590±5	32.1±0.3	0.55±0.01	10.4±0.1
40 mg/mL	585±8	32.8±0.5	0.55±0.01	10.6±0.1
60 mg/mL	580±5	30.6±0.1	0.55±0.02	9.7±0.3

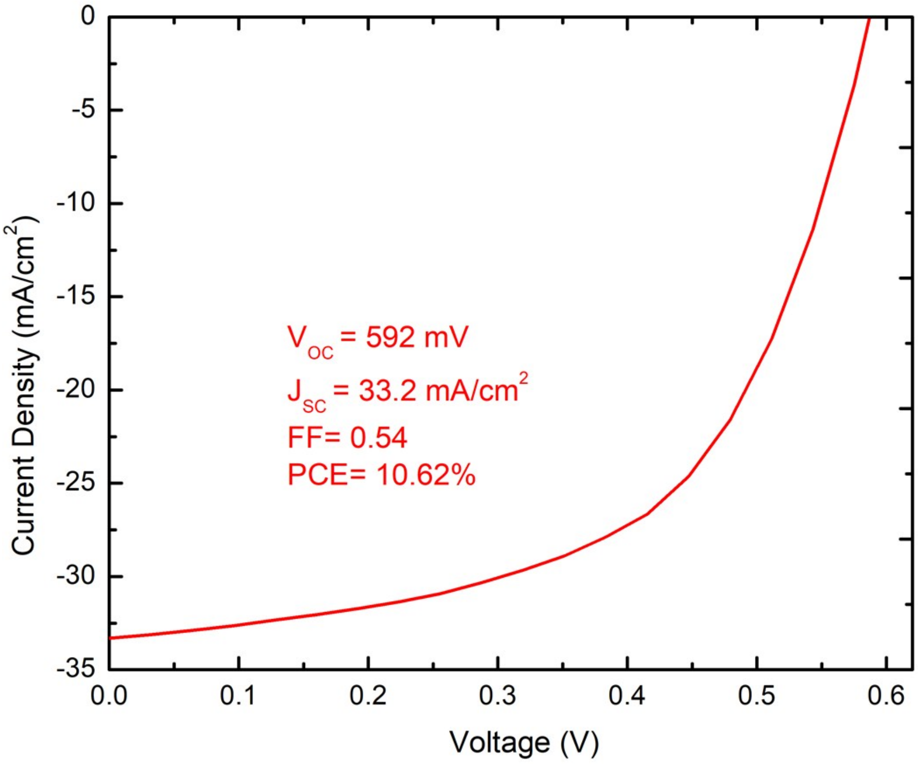


Fig. S3. *J*-*V* characteristics of champion PbS QDs/ZnO NWs PV device (highest J_{SC}) with CdSe/ZnS QDs coating of 40 mg/mL concentration measured under 100 mW cm^{-2} AM1.5G illumination.

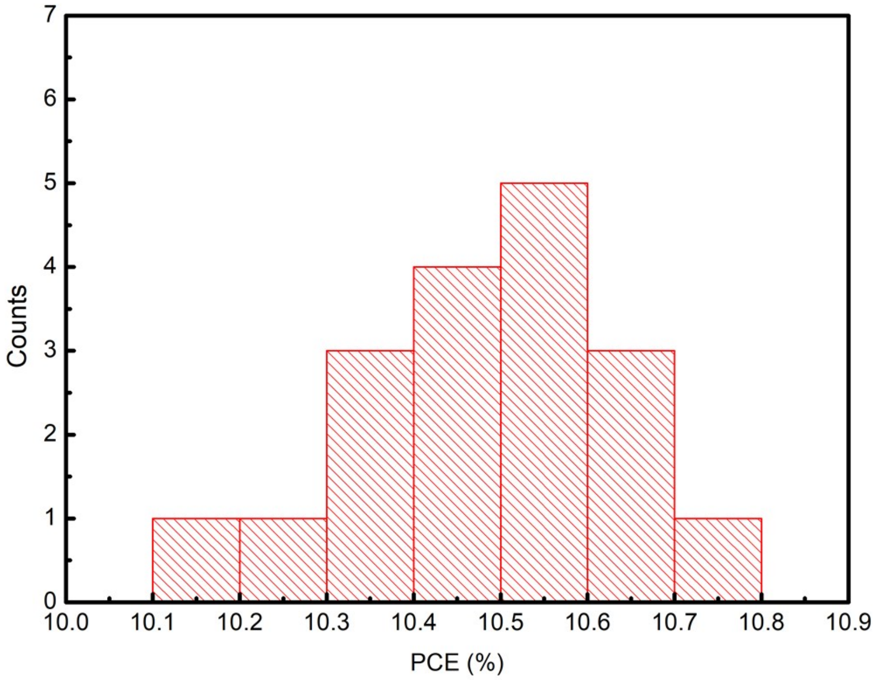
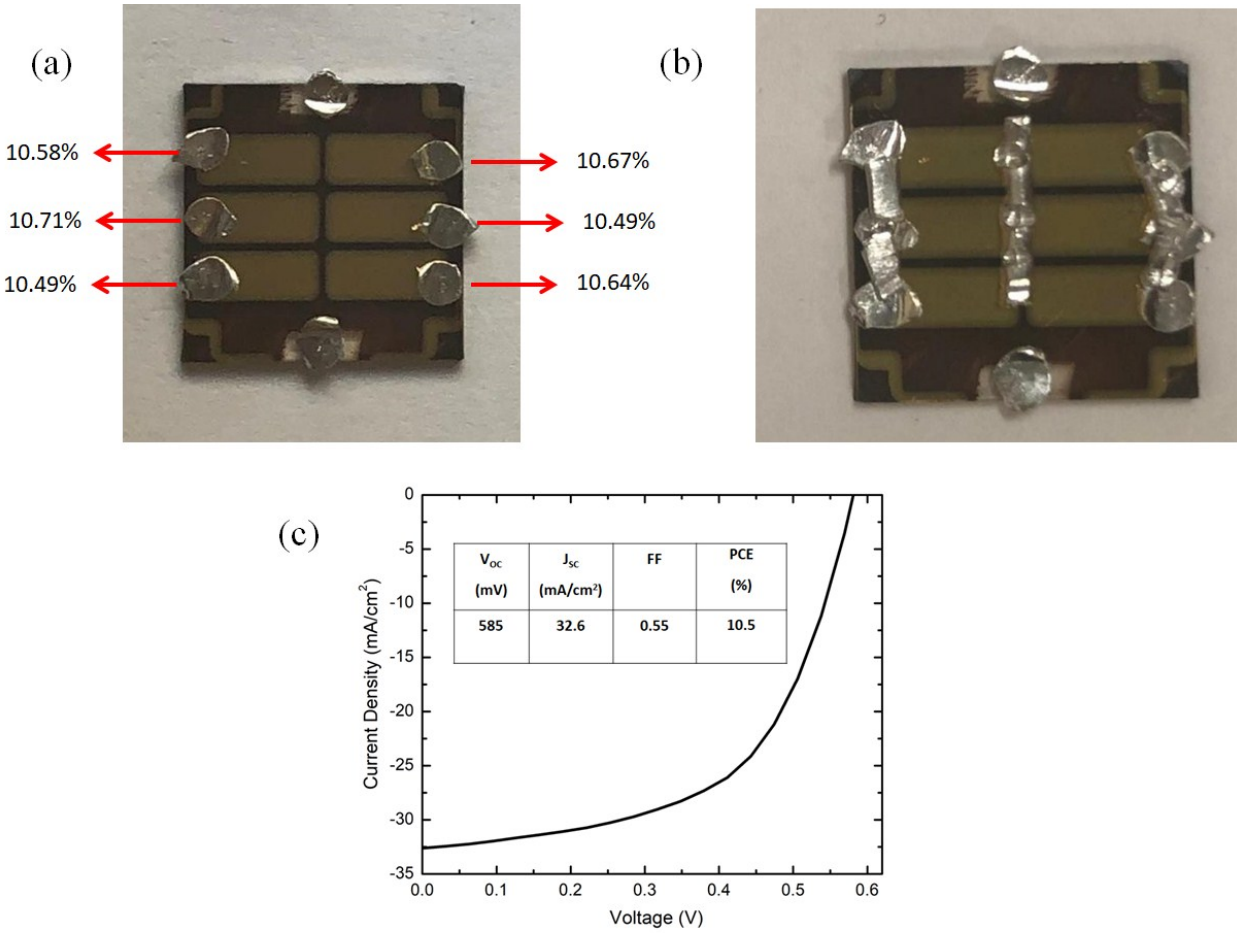


Fig. S4. Histogram of the reproducibility of PbS QDs PV with ZnO NWs and CdSe/ZnS QDs coating (40 mg/mL).

S5. PV performance of a device with larger area

In order to show the potential of our CdSe/ZnS-coated PbS QDs PV with ZnO NWs for large scale, we measured the efficiency of each of the six devices on one sample as shown in the Fig. S5(a). We find that all devices give us efficiencies of over 10.4%, indicating the possibility of the fabrication of large-scale NW-based devices. Moreover, we connected all six pads (active area for each pad: 0.054 cm^2) in one device to obtain a device with 0.324 cm^2 active area (Fig. S5(b)) and measured the *J-V* curve of this device. Fig. S5 (c) shows the *J-V* curve of the corresponding device. This device with relatively large area gave an efficiency of 10.5%.



V_{oc} (mV)	J_{sc} (mA/cm^2)	FF	PCE (%)
585	32.6	0.55	10.5

Fig. S5. Photographs of PbS QDs PV with ZnO NWs and Cdse/ZnS QDs (40 mg/mL) with six pads and device area of 0.054 cm^2 for each pad before (a) and after (b) connection of all the pads. (c) *J-V* curve of the device with 0.324 cm^2 active area by connecting all the pads.

References:

[1] Chuang, C.-H. M. et al. Open-circuit voltage deficit, radiative sub-bandgap states, and prospects in quantum dot solar cells. Nano Lett. 15, 3286-3294 (2015).