

## *Supporting Information*

### **Enlarging Stokes Shift of CuInS<sub>2</sub> Quantum Dots Using Thiol-Enes Polymers for Efficient Large-area Luminescent Solar Concentrators**

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## ***Section 1. Experimental details***

***Chemicals:*** Zinc acetate ( $\text{Zn}(\text{Ac})_2$ , 99.99%), Zinc stearate ( $\text{Zn}(\text{St})_2$ , Purum), copper(I) iodide ( $\text{CuI}$ , 99.99%), indium acetate ( $\text{In}(\text{Ac})_3$ , 99.99%), sulfur powder (S, 99.99%), 1-dodecanethiol (DDT, 99.9%), oleic acid (OA, 97%), oleylamine (OLA, 97%), octadecene (ODE, 90%), trioctylphosphine (TOP, 97%), butyl 3-mercaptopropionate (MPE, 97%), Pentaerythritol Tetra(3-mercaptopropionate) (thiol monomers), pentaerythritol tetrakis (3-mercaptopbutylate) (PE1), 1,4-Bis(3-mercaptopbutyryloxy) butane (BD1), triallyl-1,3,5-triazine-2,4,6(1H,3H,5H)-trione, 1-hydroxycyclohexyl phenyl ketone (Irgacure-184, 99%).

***Synthesis of  $\text{CuInS}_2/\text{ZnS}$  (CIS/ZnS) QDs (Thin ZnS shell-1):*** To synthesize CIS core QDs, 0.5 mmol of  $\text{CuI}$  (for QDs with copper to indium ratios of 1:4 was 0.125 mmol), 0.5 mmol of  $\text{In}(\text{Ac})_3$ , and 5 ml of DDT were placed in a round bottom three-necked flask, with a magnetic stir in the Schlenk line system under  $\text{N}_2$  atmosphere. The mixture was then heated to 120 °C until the solid powder was completely dissolved. The temperature was then increased to 235 °C and the reaction was carried out for 20 min to obtain the required QDs. Finally, the mixture was cooled to room temperature for subsequent use.

For the synthesis of CIS/ZnS QDs, 8 mL of ODE, 4 mmol of  $\text{Zn}(\text{Ac})_2$  and 8 mmol of DDT were placed in a round bottom three-necked flask. The mixture was evacuated and heated to 120°C under  $\text{N}_2$  to prepare Zn-DDT. After the solution was clarified, the prepared CIS core was injected into the solution. The temperature was then quickly raised to 240 °C and kept for 1h. If no further coating of the ZnS shell is required, the reaction was allowed to cool naturally. The QDs were purified by repeated precipitation and dispersion with ethanol

and hexane. Finally, the QDs were dispersed in hexane.

***Synthesis of CIS/ZnS QDs (Thin ZnS shell-2):*** In another 100 mL round-bottom flask, a zinc oleate solution was prepared by mixing 8 mL of ODE, 4 mmol of Zn(Ac)<sub>2</sub> and 8 mmol of OA were mixed. The mixture was heated to 120 °C under N<sub>2</sub> protection to prepare Zn-OA. After complete dissolution, Zn-OA was slowly injected into the reaction solution of the QDs with thin ZnS shell-1 (1 ml/min), and kept for 1 h to form CIS/ZnS QDs with gradient alloy shells. Then, the reaction was allowed to cool naturally. The QDs were purified by repeated precipitation and dispersion with ethanol and hexane. Finally, the QDs were dispersed in hexane.

***Synthesis of Thick Shell CIS/ZnS:*** QDs with thin ZnS shell-2 obtained in the previous step was dispersed into 10 ml of ODE, 2 mmol of Zn(St)<sub>2</sub> and 1 mL aliquot of CIS/ZnS in ODE was placed in a 100 mL three-neck flask and was further diluted with 9 mL of ODE. The mixture was heated to 100 °C under N<sub>2</sub> protection, then the mixture was brought to 230 °C. Then, 2 mL of 1 M TOP/S was swiftly injected. Subsequently, the reaction temperature was raised to 300 °C and then held for 12 min for the growth of the thick shell. Then, the reaction was allowed to cool naturally. The QDs were purified by repeated precipitation and dispersion with ethanol and hexane. Finally, the QDs were dispersed in hexane.

***Ligand exchange with MPE:*** Previously obtained QDs (Thin ZnS shell-2) were further purified by repeated precipitation and dispersion with ethanol and hexane for 3~4 times to remove excess ligand, then 60 mg of CIS/ZnS QDs with thin shell-2 dispersed in 5 ml of ODE was placed in a three-neck flask. The mixture was vacuumed for 30 min and then

heated to 120 °C under N<sub>2</sub> atmosphere. 2 ml of MPE was injected into the mixture and kept for 20 min. The mixture was then cooled to room temperature and purified multiple times using a combination of hexane and ethanol. Finally, the QDs were dispersed in hexane.

***Fabrication of LSC:*** Take 8 ml of the exchanged QDs, and after ethanol precipitation and centrifugation, the obtained QD powder was dispersed in 13 g of trimethylolpropane triacrylate (alkene monomers) to obtain a clear and transparent mixture. Then, 37 g of tetrakis(3-mercaptopropionate) pentaerythritol ester (thiol monomers) and 0.4 g of Irgacure-184 (initiators) was added to the solution. Subsequently, the mixture was mixed thoroughly by ultrasonication for 10 minutes. Finally, place the mixture in a vacuum desiccator to remove bubbles in the solution. Slowly pour the mixture solution into a prepared glass mold, and cure it with 360 nm ultraviolet light for 30 seconds to initiate the thiol-ene polymerization reaction. To obtain a uniform nanocomposite interlayer, the UV light intensity should be kept below 0.1 mW·cm<sup>-2</sup>. The polymerization is completed after storing the sample in the dark for 1 hour.

***Transmission Electron Microscopy (TEM):*** TEM measurements were performed on an TECNAI G2 20 LaB6 microscope (Schottky emitter), and operated at an accelerating voltage of 200 kV.

***Powder X-Ray Diffraction (XRD):*** Powder XRD patterns were acquired in Bragg–Brentano geometry with Cu K $\alpha$  radiation (Bruker-AXS D8 Advance powder diffractometer).

***Optical Measurements:*** The PL spectra of the QDs were obtained using a spectrometer

(Cary Eclipse, Agilent, Austria). The UV–vis light absorption spectra were obtained by Shimadzu UV-3150 UV–vis–NIR spectrometer. Time-resolved PL was determined using a Hamamatsu C11367 Quantaaurus-Tau system with an excitation wavelength of 405 nm. The absolute PLQY was measured using a Horiba Fluorolog system equipped with a single grating and a Quanta-Phil integration sphere coupled to the Fluorolog system. FTIR measurements were performed using JASCO-FT/IR-4700, and the FTIR spectra of the QDs were recorded in the attenuated total reflectance (ATR) mode ( $4\text{ cm}^{-1}$ ).

***Femtosecond transient absorption spectroscopy:*** The femtosecond transient absorption is based on a regenerative amplified Ti: Sapphire femtosecond laser system (Light Conversion, 100 kHz repetition rate,  $\sim 190$  fs pulse duration and  $200\text{ }\mu\text{J/pulse}$  1027 nm fundamental pulse energy). The data collection used system from Time-Tech Spectra, Inc. In short, the fundamental pulse was split in two parts with a 90:10 beam splitter. A 1027 nm pulse with  $\sim 200\text{ }\mu\text{J}$ , after adjusted by a variable ND filter and an iris, was focused into a crystal window to generate a white light continuum (WLC) probe. The probe light for our TA tests was produced by a 1 cm thick YAG crystal, which has a spectral range of 500-950 nm. In this experiment, the transient absorption data within 7 ns time window was studied by two crystals with different probe ranges. Then the WLC was split by a 70:30 beam splitter to provide the probe for the sample and reference to correct for the intrinsic fluctuation. The probe beam was focused onto the sample and eventually focused into an optical fiber entrance. The optical fiber is coupled with visible spectrometer and then a 1024 elements CMOS camera. A chopper modulated the pump by 5000 Hz to cut off every other pump pulse to provide transient absorption signal. The diameter on the sample for pump and probe, is  $248\text{ }\mu\text{m}$  and  $157\text{ }\mu\text{m}$ , respectively. The typical instrument response is

well fitted by a Gaussian function with  $\sim 200$  fs FWHM. The chirp was corrected by fitting the solvent response in all data set. The nanosecond transient absorption data was collected using system from Time-Tech Spectra, Inc. In short, the pump pulse was provided the same way as in the femtosecond setup. The probe pulse was generated by 1 kHz WLC laser (Disco, Leukos). The sample was contained in 1 mm quartz cell and was constantly stirred during the experiment to avoid photodegradation. The sample was loaded in glovebox and was air tight during experiments. All experiments were performed under room temperature. Each curve on transient absorption spectra is the average over all the spectra within the corresponding time window.

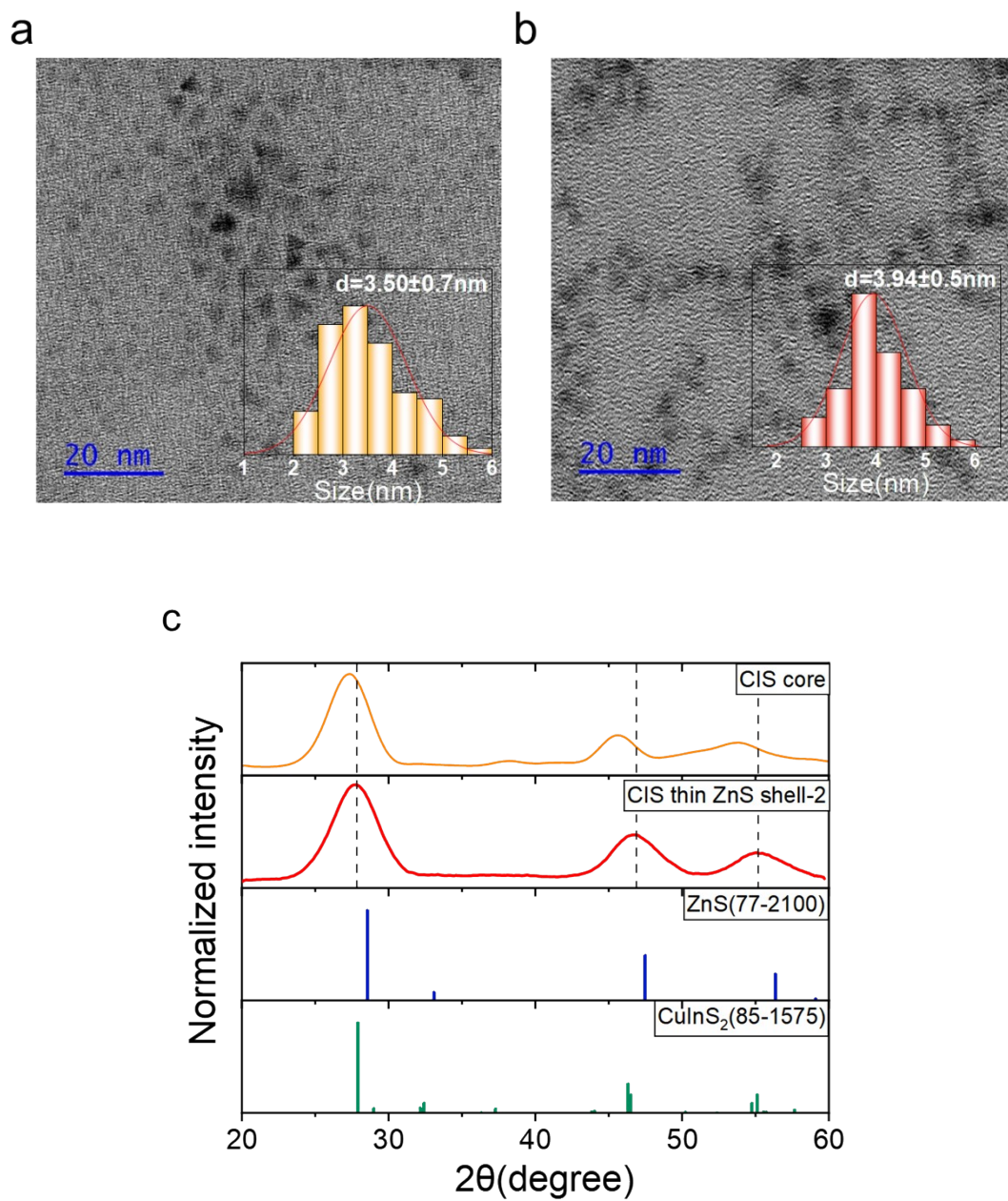
***Photovoltaic Measurements:*** The photovoltaic performance of the LSCs was investigated using a large-area LED-based solar simulator (Sunbrick from G2V optics, 350–1100nm spectral range, ASTM E927 class AAA+,  $25 \times 25$  cm<sup>2</sup>) and carried out by applying an external potential bias (from 0 to 6V) to the device while recording the generated photocurrent with a Keithley model 2450 digital source meter. Excess parts on the solar cells were covered by black tape. For LSC-S series, all four edges of the device were attached with solar cells (IXYS, IXOLARTM, SM141K08L,  $88 \times 15$  mm), which were connected in parallel. For I-V measurements using the back reflector, a piece of white paper was placed under the LSCs at a distance of 6 cm. For LSC-L, all four edges of the device were attached with solar cells (IXYS, IXOLARTM, SM182K01L,  $285 \times 8$  mm), which were connected in tandem, and I-V measurements was conducted at a third-party certification institution, the Shanghai Institute of Microsystem and Information Technology (SIMIT), using a class AAA long-pulse solar simulator (Wacom Electric Co. Ltd., WPSS-2.0  $\times$  1.5H  $-50 \times 6$ ), the test light intensity equivalent to a standard sunlight.

For device with four edges connected to a solar cell, the PCE value can be calculated by the following equation.

$$PCE = \frac{I_{sc} \cdot V_{oc} \cdot FF}{I_0 \cdot A_{top}} \quad S1$$

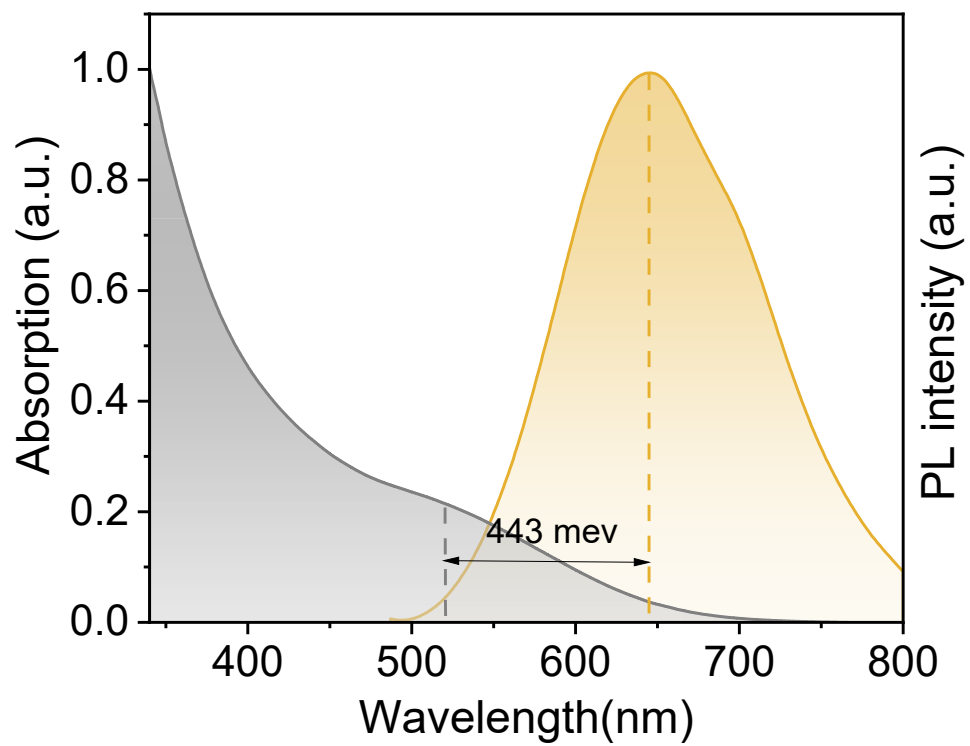
where  $I_{sc}$ ,  $V_{oc}$ , and  $FF$  were obtained directly from the measured  $I-V$  curves,  $I_0$  is the light intensity of AM1.5 G, which is 0.1 W/cm<sup>2</sup>, and  $A_{top}$  is the surface area of the LSCs.

## ***Section 2. Basic characterization of CIS/ZnS QDs***



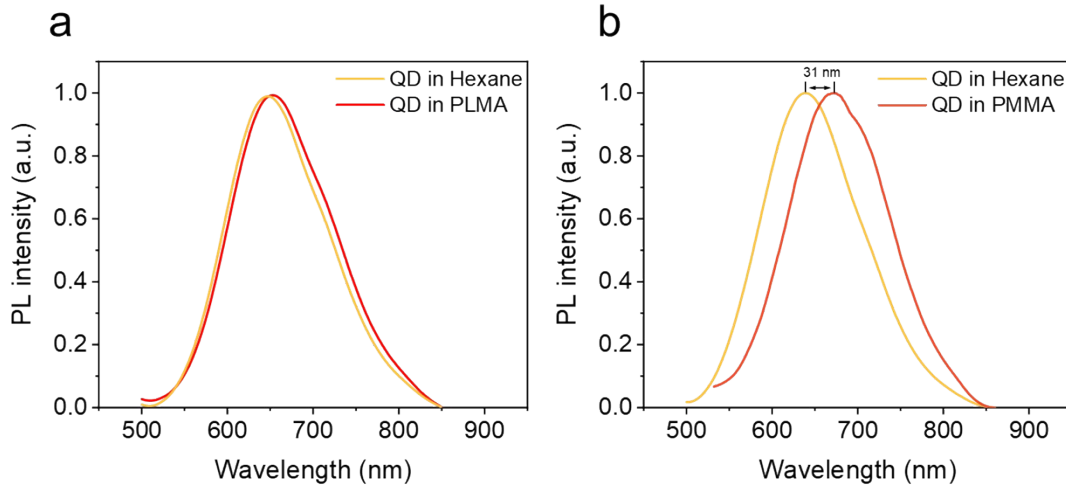
**Figure S1.** TEM images of CIS/ZnS QDs before a) and after b) ligand exchange with particle size distribution inserted. c). XRD of the as-synthesized CIS/ZnS QDs.





**Figure S2.** UV-Visible absorption and PL spectra of the CIS/ZnS core/shell QDs.

**Section 3. Optical properties of QD solution and QD/polymer nanocomposites**



**Figure S3.** a) PL spectra of QDs-OA in PLMA and hexane. b) PL spectra of QDs-MMA in PMMA and hexane.

**PL lifetime fitting:** The excitation wavelength of 405 nm was used for all samples, and the monitoring wavelengths were the respective emission peak positions of the samples, and the data were fitted according to equation S3

$$I(t) = \sum_{i=1}^n A_i \exp\left(-t/\tau_i\right) \quad \text{S2}$$

**Table S1.** Fitting parameters of PL lifetime for CIS/ZnS QDs in different solvent.

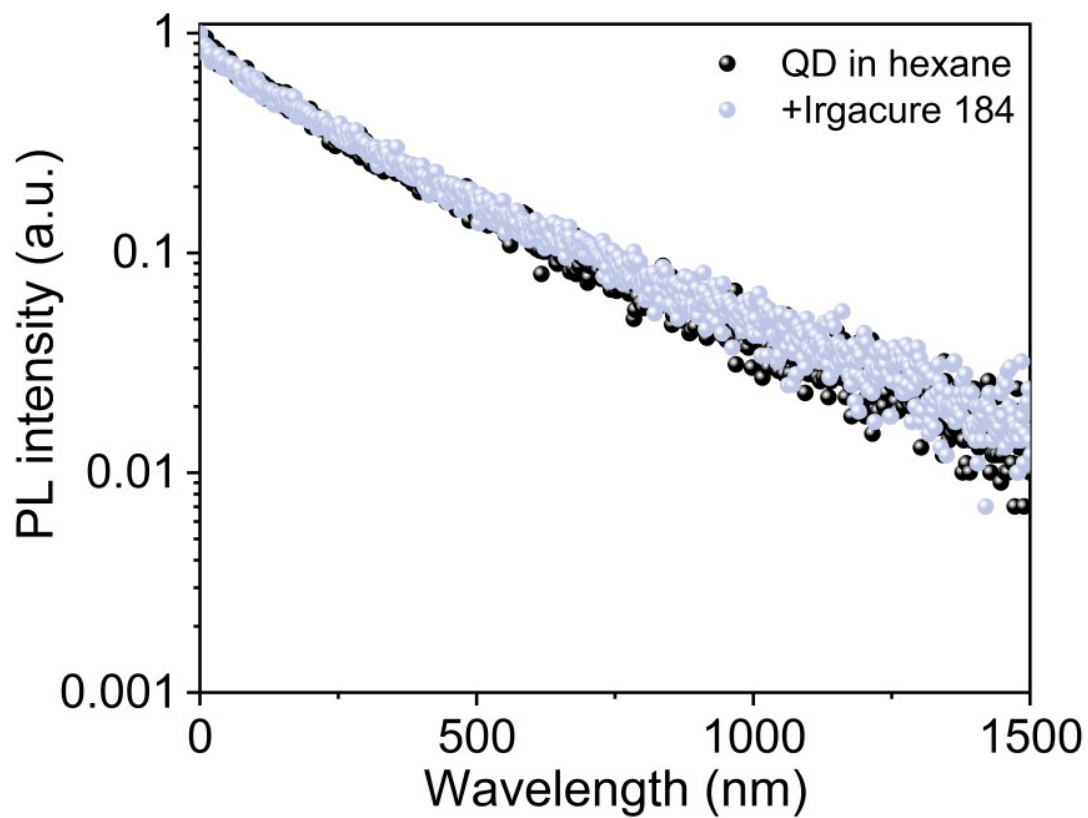
Solvent	A <sub>1</sub>	A <sub>2</sub>	A <sub>3</sub>	τ <sub>1</sub>	τ <sub>2</sub>	τ <sub>3</sub>	τ <sub>avg</sub>	F <sub>1</sub>	F <sub>2</sub>	F <sub>3</sub>
hexane	0.070	0.43	0.48	26.1	131.8	373.4	339.6	0.008	0.238	0.754
thiol	0.34	0.481	0.21	41.5	121.3	310.8	203.3	0.102	0.424	0.474
OSTE	0.032	0.52	0.45	1.6	114.7	325.7	259.4	0.0002	0.2892	0.7106

$$\tau_{avg} = \frac{\sum A_i \tau_i^2}{\sum A_i \tau_i}$$

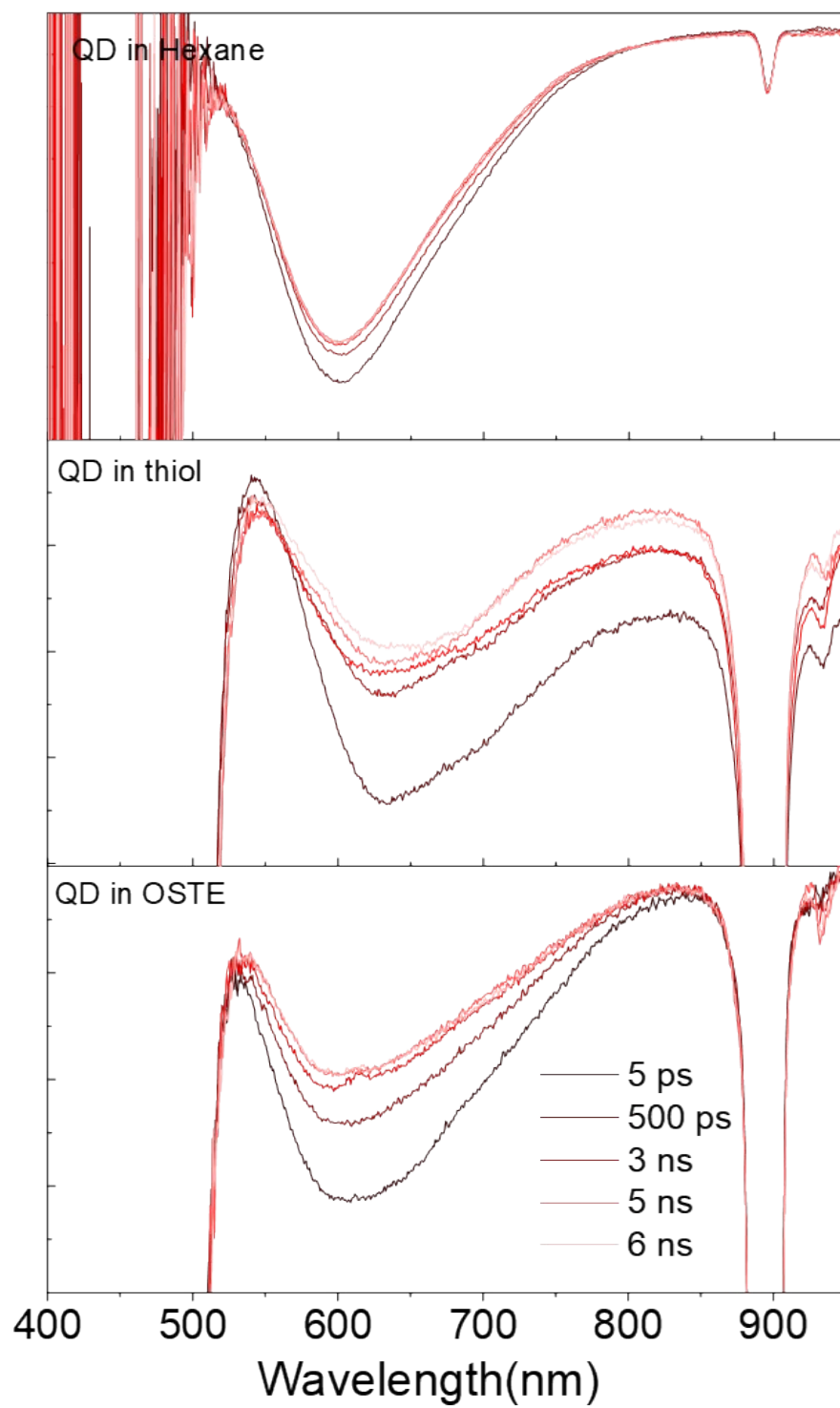
The average PL lifetime  $\tau_{avg}$  is calculated according to

$$F_i = \frac{A_i \tau_i}{\sum_i A_i \tau_i}$$

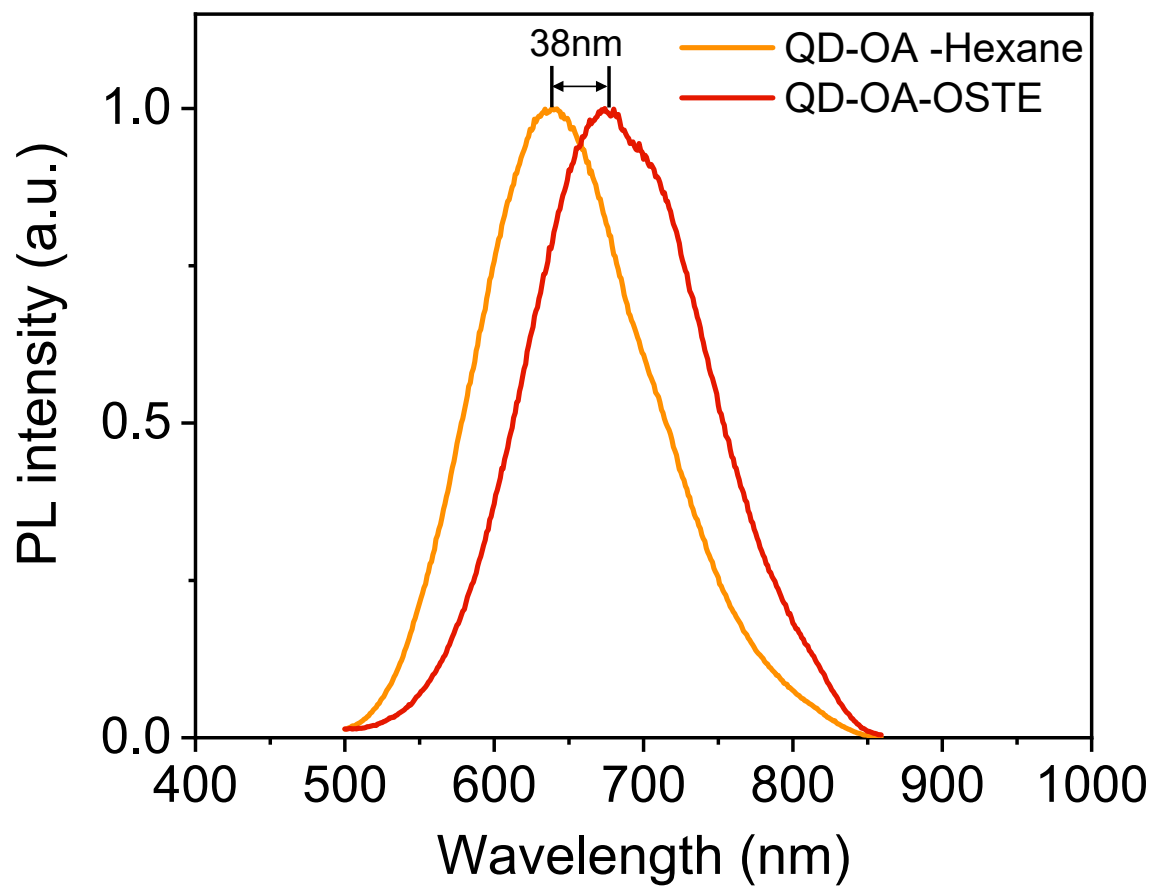
$F_i$  refers to fraction of the lifetime component is calculated according to



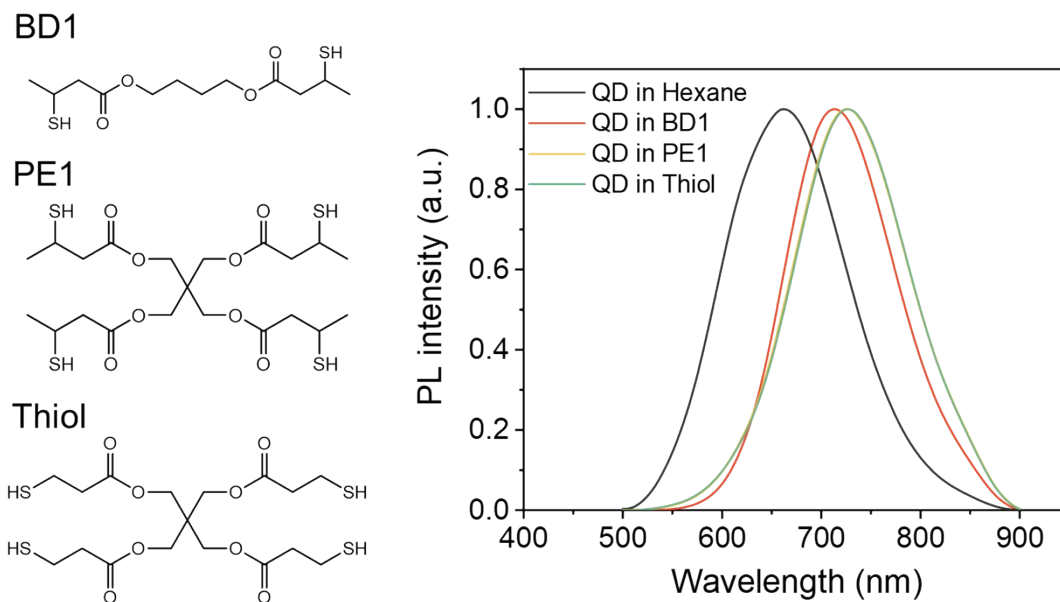
**Figure S4.** Transient PL decay curves of quantum dot solution after UV exposure with and without the addition of Irgacure 184.



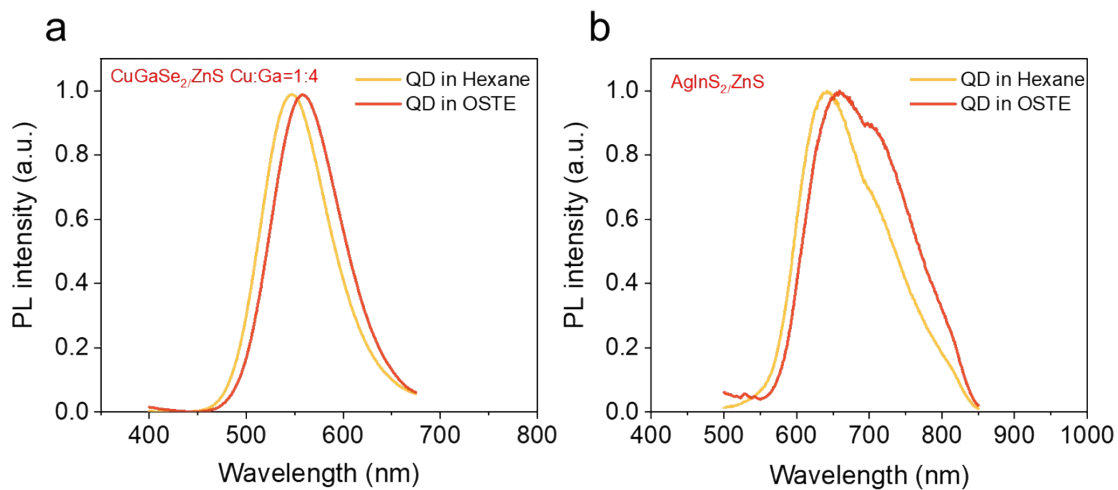
**Figure S5.** TA spectra of the QDs in different dispersion media from 400nm-950nm, excited at 450 nm.



**Figure S6.** PL spectra of QDs-OA in OSTE and Hexane.

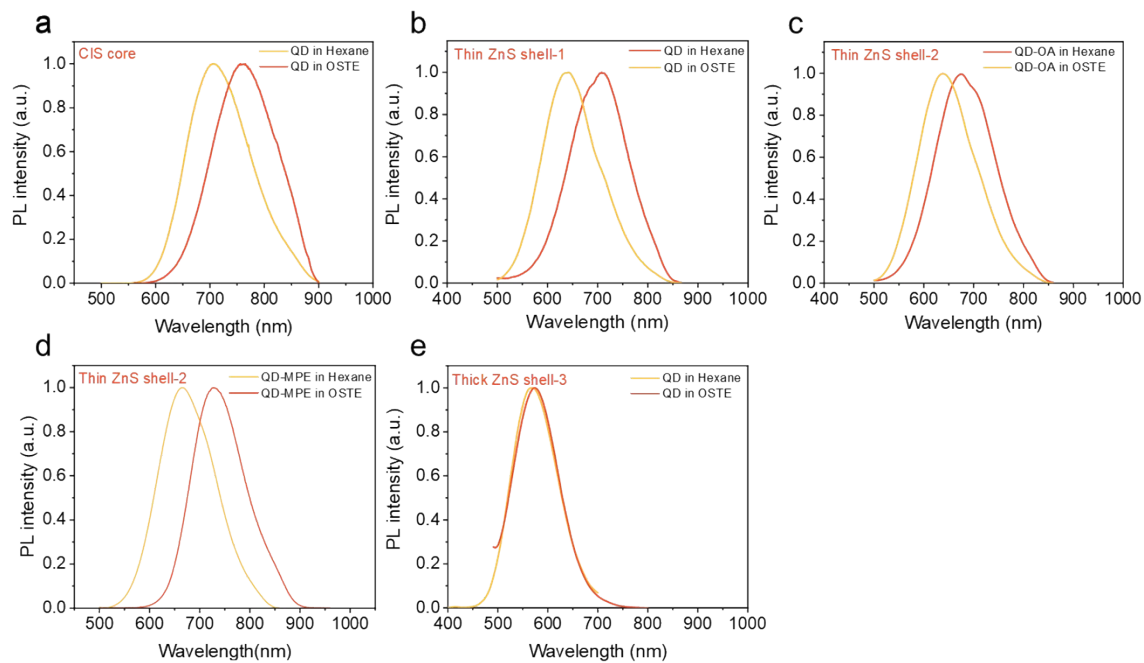


**Figure S7.** Molecular structure of BD1 and PE1, the thiol monomers mainly used in this work and PL spectra of QDs-MPE in different solvents.



**Figure S8.** a) PL spectra of  $\text{CuGaSe}_2/\text{ZnS}$  QDs in OSTE and hexane b) PL spectra of  $\text{AgInS}_2/\text{ZnS}$  QDs in OSTE and hexane.





**Figure S9.** a) PL spectra of CIS core in OSTE and hexane. b-e) PL spectra of CIS/ZnS QDs with different ZnS shell thickness in OSTE and hexane.

**Table S2.** PL peak wavelength (derived from Figure S7) and PLQY of CIS QDs with different ZnS shell coating thicknesses in hexane and OSTE.

QDs	Ligand	PL peak wavelength (hexane) (nm)	PL peak wavelength (OSTE) (nm)	Red-shift (nm)	PLQY (hexane) (%)	PLQY (OSTE) (%)
Core	DDT	706	761	55	21	22
Thin ZnS shell-1	DDT	639	697	58	84	72
Thin ZnS shell-2	OA	638	676	38	93	93
Thin ZnS shell-2	MPE	665	730	65	85	85
Thick ZnS shell-3	stearic acid	568	574	6	90	92

As shown in Section 1, we obtained QDs with thin ZnS shell-1 and thin ZnS shell-2 by coating the CIS core with precursor solutions of Zn-DDT and Zn-OA, and no extra S precursor was added during the coating process. In this process, cation-exchange reactions dominate and a thin ZnS shell growth occurred, resulting in negligible changes in the particle size of QDs with thin ZnS shell compare to CIS core. However, when we used zinc stearate and highly active TOP-S as the precursors, a thick ZnS shell layer was formed on the QDs.

#### ***Section 4. Optical and I-V measurements of LSCs***

***Absorption - PL overlap integral:*** The effective overlap integral of LSC can be calculated by the following equations (S3),

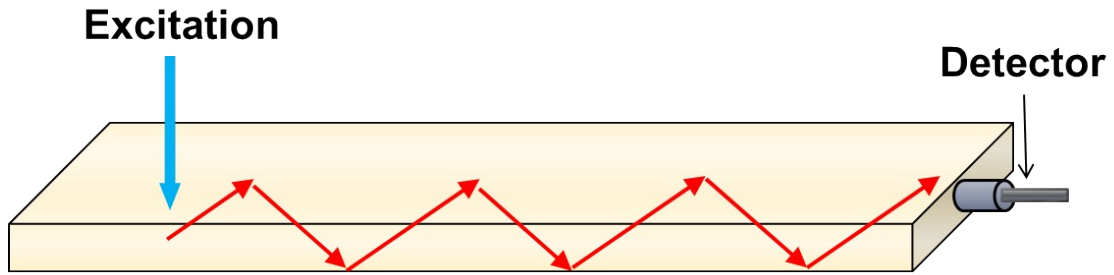
$$\alpha_{ab-PL} = \alpha_{ab-PL} \cdot \frac{d}{d + 2H} \quad \text{S3}$$

where  $d$  is the thickness of the QD/OSTE nanocomposite interlayer, and  $H$  is the glass thickness. In this work, the glass thickness was 2 mm, and the interlayer of LSC-Hexane and LSC-OSTE was 5 mm.

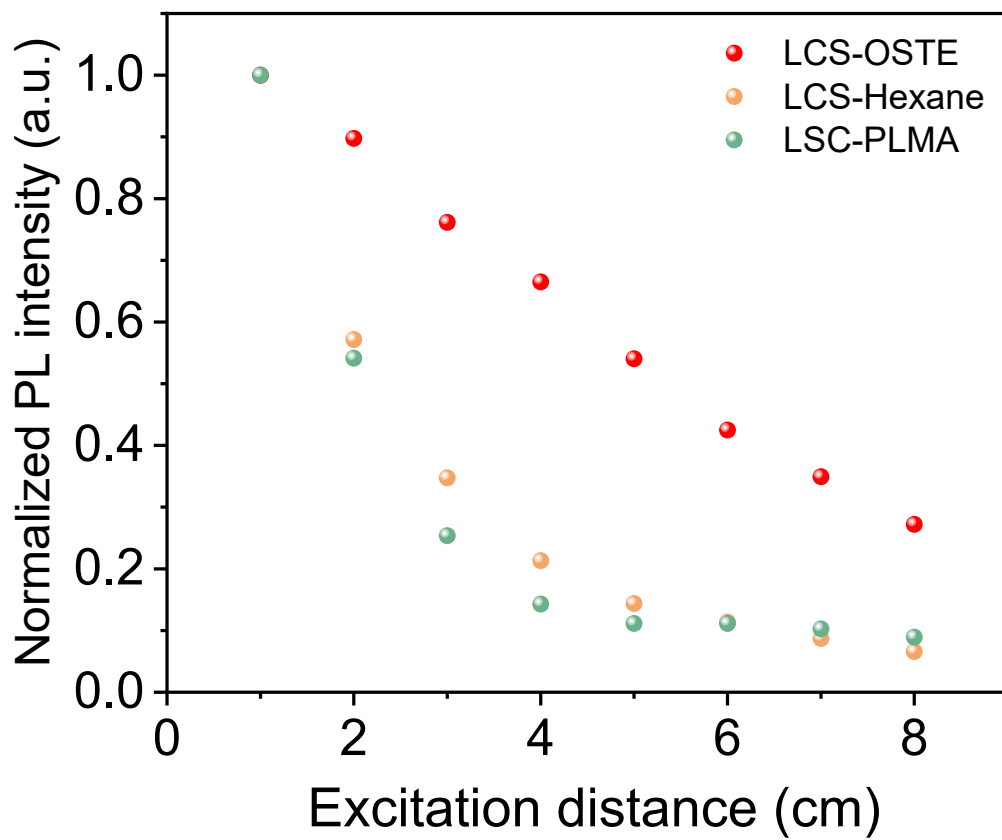
**Table S3.** correction factor( $\beta$ ) and reabsorption coefficient of different LSCs.

	$\alpha_{ab-PL}$	correction factor( $\beta$ ) $\frac{d}{d + 2H}$	effective overlap integral
LSC-PLMA	0.312	1	0.312
LSC-Hexane	0.355	5/9	0.197
LSC-OSTE	0.126	5/9	0.07

*Excitation distance dependent PL measurement for LSC:* The PL spectrum of the light reaching the edge is detected by a spectrometer probe fixed at the edge of the LSC, and the distance of the excitation point from the edge of the device is varied to obtain distance dependent PL spectrum.



**Figure S10.** Schematic diagram of showing output PL intensity of LSC under different optical paths.



**Figure S11.** Normalized output PL intensity of the LSCs detected under different excitation distances.

## Section 5. Calculation of waveguiding efficiency

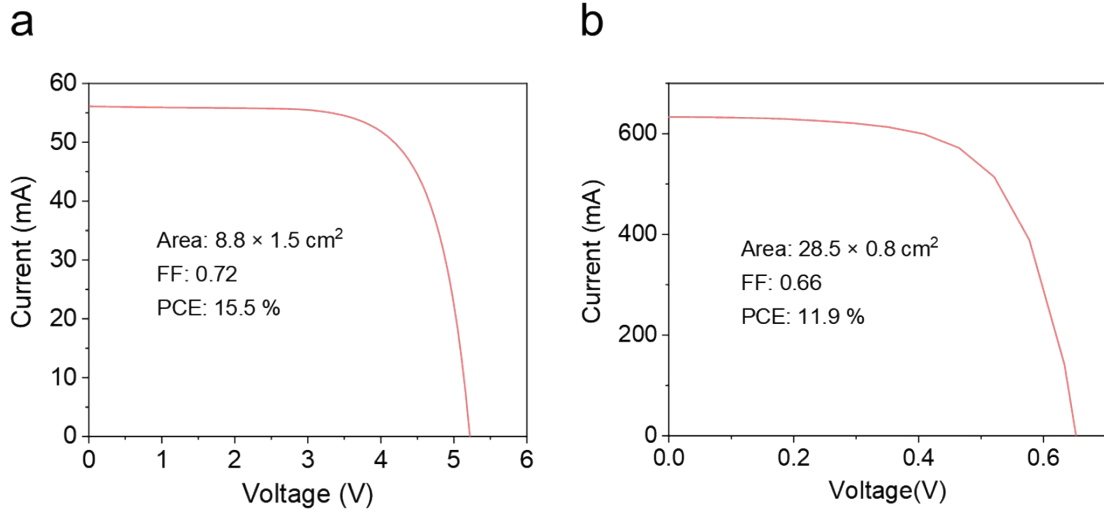
$\eta_{wvgd}$  is a waveguiding efficiency, which is one of the most important parameters in LSC.

For a square shape LSC with a side length  $l$ , it can be evaluated as<sup>1,2</sup>

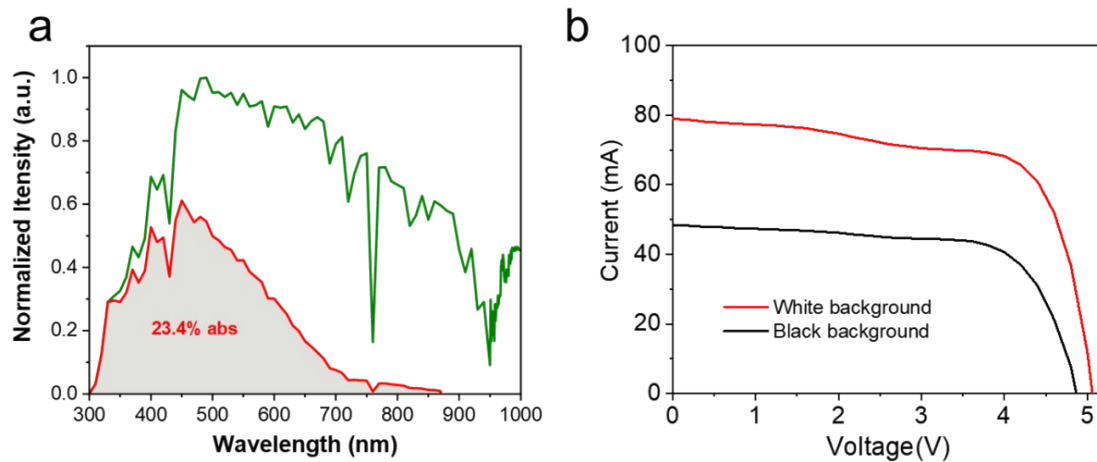
$$\eta_{wvgd} = \frac{M_1(2\alpha lk/\sqrt{\pi}) \cdot \alpha \cdot \sqrt{\pi}}{\delta(\alpha_{sc} + QY \cdot \alpha_{re}) \left( lk\alpha + M_1\left(\frac{2\alpha lk}{\sqrt{\pi}}\right) \cdot \sqrt{\pi} \right) - lk\alpha^2} \quad S4$$

where  $\alpha$  [ $\text{cm}^{-1}$ ] is the sum of scattering  $\alpha_{sc}$  [ $\text{cm}^{-1}$ ], reabsorption  $\alpha_{re}$  [ $\text{cm}^{-1}$ ], and matrix absorption  $\alpha_{mx}$  [ $\text{cm}^{-1}$ ] coefficients:  $\alpha = \alpha_{sc} + \alpha_{re} + \alpha_{mx}$ ;  $\delta$  is a waveguiding fraction ( $\sim 75\%$  for  $n = 1.5$ , which is unity minus escape cone losses of 25%).  $k$  is a coefficient for 3D geometry:  $k \approx 1.14$  for  $n = 1.5$ ;  $M_1(\xi)$  is a modified Struve function of the second kind.

**Section 6. Device performance**

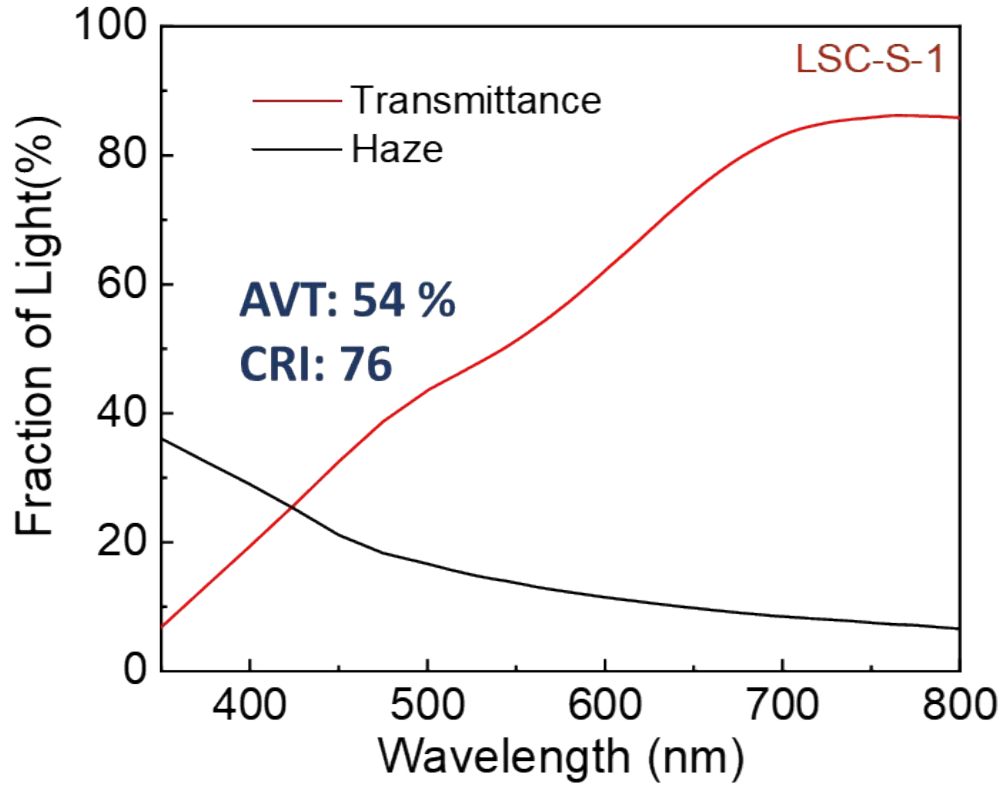


**Figure S12.**  $I-V$  curves of the silicon solar cells used for LSC-S(left) and LSC-L(right) under the solar illumination with power of  $100 \text{ mW}\cdot\text{cm}^{-2}$ .



**Figure S13.** Calculated solar absorption and  $I$ - $V$  curves for the LSC-S-1 with area of  $9 \times 9$   $\text{cm}^2$ .





**Figure S14.** Aesthetic quality of the LSC-S-1, with measured transmittance spectrum (black), haze spectra (red). The relatively high haze value of the device was due to the special two-interlayer structure, in which two interlayers of QD/OSTE nanocomposites were sandwiched in three layers of glass.

AVT was calculated as the integral of the transmission spectrum and AM 1.5G photon flux weighted against the photopic response of the human eye:

$$AVT = \frac{\int T(\lambda) \cdot V(\lambda) \cdot AM1.5G(\lambda) d\lambda}{\int V(\lambda) \cdot AM1.5G(\lambda) d\lambda}$$

S5

where  $T(\lambda)$  is the transmission spectrum of the TQDG and  $V(\lambda)$  is the human eye photopic

response. All aesthetical parameters were calculated with a plug-in developed from a reported method<sup>3</sup>.

The External Quantum Efficiency (EQE) refers to the ratio of the number of photons emitted from the edge of a photovoltaic device to the number of photons incident upon it. it could be calculated as<sup>4</sup> :

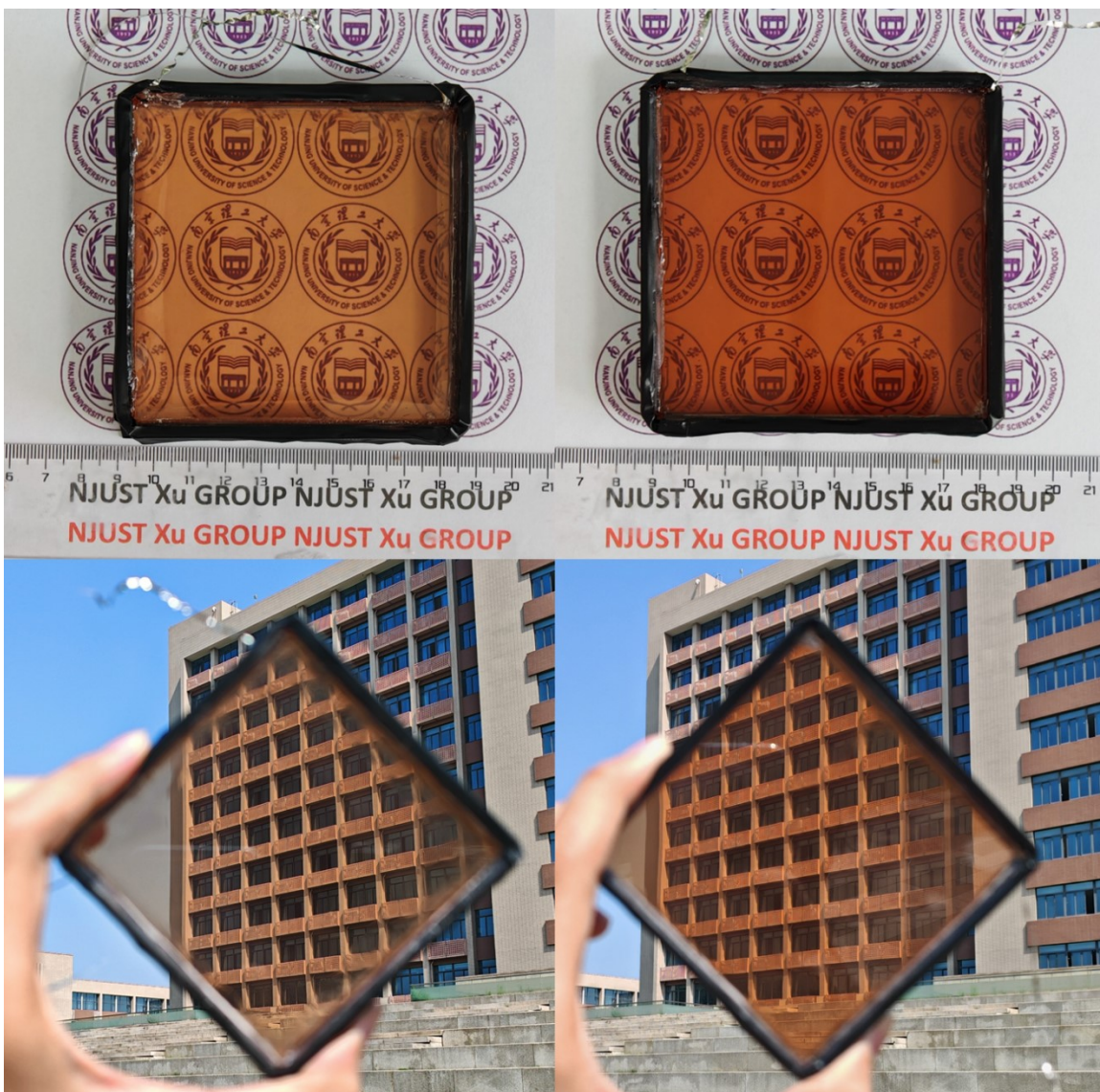
$$EQE = \frac{P_{LSC - PV}}{P_{PV} S_{LSC} G} \quad S6$$

where  $P_{PV}$  and  $P_{LSC - PV}$  are power output of a calibrated crystalline silicon solar cell when it is placed first in front of the LSC surface and then at its edge,  $S_{LSC}$  is the LSC spectral reshaping factor defined as the ratio of the PV EQE averaged over the PL spectrum of the LSC fluorophore and  $G$  is the geometric gain factor.

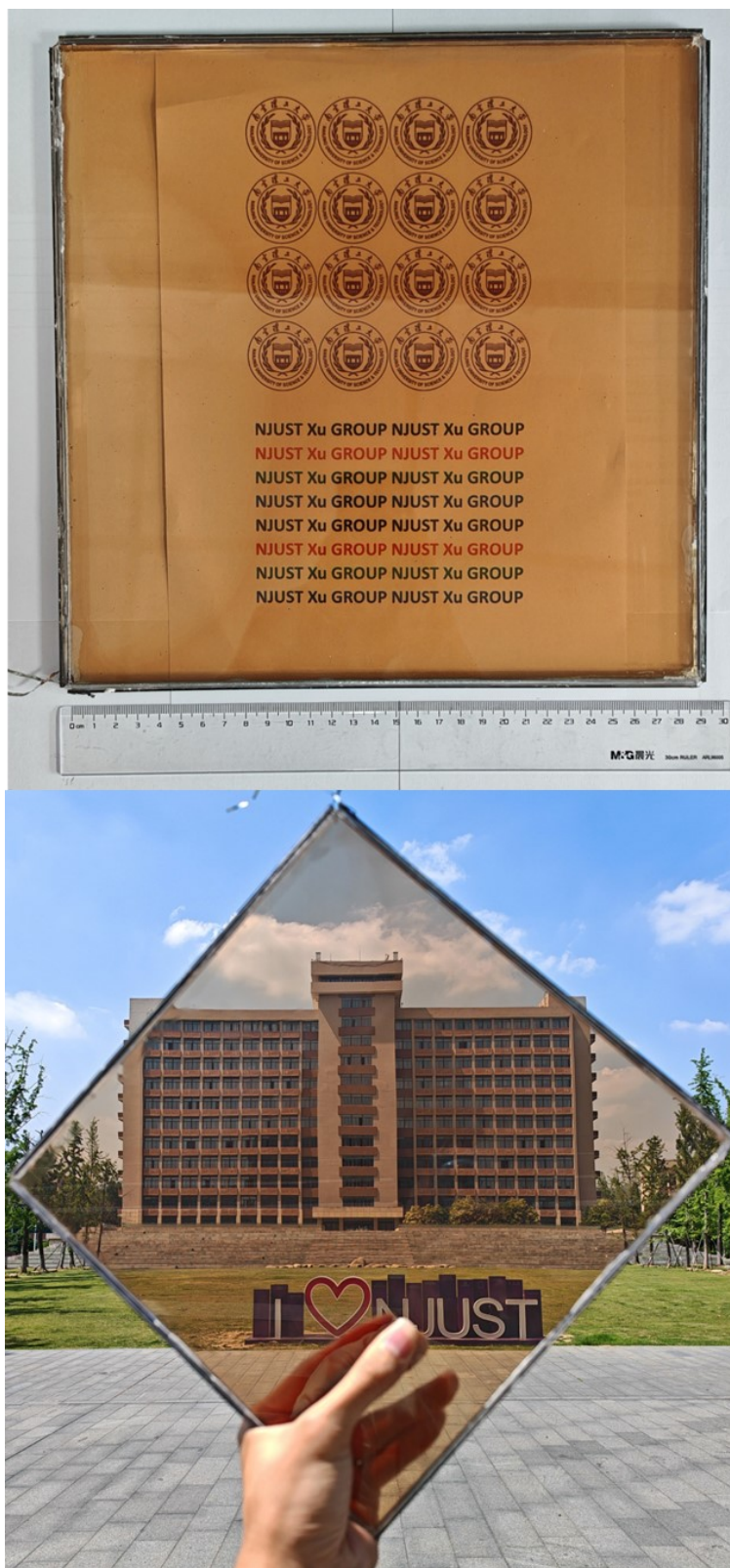
The Internal Quantum Efficiency (IQE) is a critical parameter for evaluating the performance of photonic devices. It is defined as the ratio of the number of photons emitted from the edge of the device to the number of photons absorbed by the device. It could be calculate as:

$$IQE = \frac{EQE}{A_{QD}} \quad S7$$

Where  $A_{QD}$  is solar photon absorption ratio of the device.



**Figure S15.** Photos of LSCs ( $9 \times 9 \text{ cm}^2$ ) under room light (top) and sunlight (bottom) (Left: LSC-S-1, Right: LSC-S-2).



**Figure S16.** Photos of LSCs ( $29 \times 29 \text{ cm}^2$ ) under room light (top) and sunlight (bottom).

**Table S4.** Light intensity of the I-V measurements, haze value at 730 nm (scattering coefficient), as well as PV parameters extracted from I-V measurements of LSCs. “W/O” standards for without back reflection and “W” refers to with back reflection.

<b>Samples</b>	<b>Light Intensity (mW/cm<sup>2</sup>)</b>	<b>Haze (scattering coefficient) cm<sup>-1</sup></b>	<b>Back reflection</b>	<b><math>I_{sc}</math> (mA)</b>	<b><math>V_{oc}</math> (V)</b>	<b>FF</b>	<b>PCE (%)</b>
LSC- S-1	100	0.083	W/O	48	4.85	0.69	2.09
			W	79	5.07	0.69	3.56
LSC- S-2	100	0.069	W/O	60	4.93	0.70	2.67
			W	90	5.09	0.72	4.25
LSC- L	100	0.027	W/O	397	2.64	0.63	0.82
			W	616	2.75	0.65	1.36

## Section 7. Third-party certification report



中国认可  
国际互认  
检测  
TESTING  
CNAS L8490

Test and Calibration Center of New Energy Device and Module,  
Shanghai Institute of Microsystem and Information Technology,  
Chinese Academy of Sciences (SIMIT)

# Measurement Report

Report No. 24TR051501

<b>Client Name</b>	Nanjing University of Science and Technology
<b>Client Address</b>	200 Xiaolingwei, Nanjing, Jiangsu Province, China
<b>Sample</b>	Transparent solar glass
<b>Manufacturer</b>	Nanjing University of Science and Technology
<b>Measurement Date</b>	15 <sup>th</sup> May, 2024

<b>Performed by:</b>	Qiang Shi <i>Qiang Shi</i>	<b>Date:</b> 15/05/2024
<b>Reviewed by:</b>	Wenjie Zhao <i>Wenjie Zhao</i>	<b>Date:</b> 15/05/2024
<b>Approved by:</b>	Yucheng Liu <i>Yucheng Liu</i>	<b>Date:</b> 15/05/2024

<b>Address:</b> No.235 Chengbei Road, Jiading, Shanghai	<b>Post Code:</b> 201800
<b>E-mail:</b> solarcell@mail.sim.ac.cn	<b>Tel:</b> +86-021-69976905

The measurement report without signature and seal are not valid.  
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**Sample Information**

Sample Type	Transparent solar glass, AVT 51%
Serial No.	2-1#
Lab Internal No.	24051501-1#
Measurement Item	I-V characteristic
Measurement Environment	24.6±2.0°C, 59.1±5.0%R.H

**Measurement of I-V characteristic**

Reference cell	PVM 1121
Reference cell Type	mono-Si, WPVS, calibrated by NREL (Certificate No. ISO 2098)
Calibration Value/Date of Calibration for Reference cell	143.95mA/ Feb. 2024
Measurement Conditions	Standard Test Condition (STC): Spectral Distribution: AM1.5 according to IEC 60904-3 Ed.3, Irradiance: 1000±50W/m <sup>2</sup> , Temperature: 25±2°C
Measurement Equipment/ Date of Calibration	Long pulse Solar Simulator (WPSS-2.0*1.5H-50*6) / July.2023 IV test system (ADC 4601) / June. 2023 SR Measurement system (CEP-25ML-CAS) / May.2024
Measurement Method	I-V measurement: Irradiance adjusted to 1000W/m <sup>2</sup> according to the reference cell calibration value. Linear sweep in direct direction (Isc to Voc) based on IEC 60904-1:2020.
Measurement Uncertainty	Isc: 2.0%(k=2); Voc: 1.0%(k=2); Pmax: 2.4%(k=2)







====Measurement Results====

Area [cm <sup>2</sup> ]	Isc [A]	Voc [V]	Pmax [W]	FF [%]	Eff [%]	Comments
812.25	0.610	2.736	1.102	66.04	1.36	With a white backsheet
812.25	0.371	2.631	0.620	63.56	0.76	With a black absorbing paper

- A white backsheet or a black absorbing paper attached to the back of the sample during the measurement.  
- Test results listed in this measurement report refer exclusively to the mentioned measured samples.  
- The results apply only at the time of the test, and do not imply future performance.

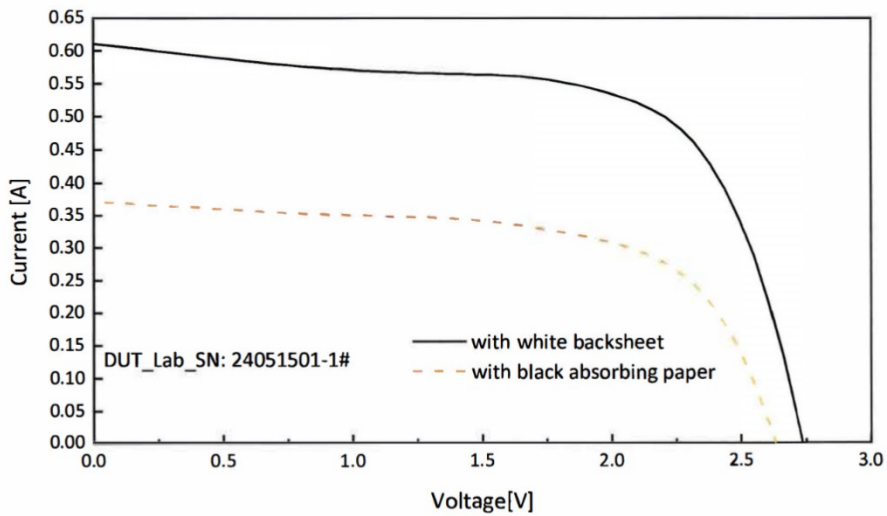


Fig.1 I-V curves of the measured sample



-----End of Report-----

## References

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