Electronic Supplementary Information

Room-Temperature and Gram-Scale Synthesis of $CsPbX_3$ (X=Cl, Br, I) Perovskite Nanocrystals with 50-85% Photoluminescence Quantum Yields

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Experimental Section

Chemcials

Cs_2CO_3 (99.9%), $CsOH{\cdot}H_2O$ (99.9%), PbO (99.9%), tetraoctylammonium bromide (98%), tetrabutylammonium bromide (99%), cetyltrimethylammonium bromide (CTAB, 99%), dihexadecyldimethylammonium bromide (97%), tetrabutylammonium chloride (97%), tetrabutylammonium iodide (99%), butyric acid (99%), hexanoic acid (99%), octanoic acid (98%), decanoic acid (99%), myristic acid (99%), and γ-butyrolactone (99%) were purchased by Aladdin Inc. Oleic acid (90%) was obtained from Sigma-Aldrich Inc. Toluene, hexane, chloroform, xylene, and dichloromethane

were bought from Beijing Chemical Works, and all solvents were of analytical grade and were used as received without further purification.

Preparation of mixed Cs-oleate and Pb-oleate precursor solution

First, 1.0 mmol $CsOH{\cdot}H_2O$ (or 0.5 mmol Cs_2CO_3), 1.0 mmol PbO, and 5.0 mL of oleic acid were loaded into a 20 mL of glass vial, and the mixture was magnetically stirred on a hot plate at 160 ^oC until a transparent solution was obtained. Then, the glass vial was putted into a 120 ^oC oven for 30 min to remove the water. Next, Cs and Pb precursor solution was diluted to be 0.1 M by adding 5 mL of toluene. Finally, the mixed Cs and Pb precursor solution was sealed and stored in air for further use. Note that all experimental procedures in this paper were conducted in the open air. To investigate the impact of the solvent on the synthesis of $CsPbBr_3$ quantum dots, chloroform, dichloromethane, xylene, or hexane was used to replace toluene. In addition, butyric acid, hexanoic acid, octanoic acid, decanoic acid, or myristic acid was used to substitute oleic acid for the synthesis of $CsPbBr_3$ quantum dots.

Small-scale synthesis of $CsPbBr_3$ quantum dots

In a typical procedure, 1.0 mL of Cs and Pb precursor solution and 15.0 mL of toluene were mixed in a 20 mL of glass vial with vigorously stirring at room temperature (25~30^oC). A Br precursor solution containing 0.1 mmol tetraoctylammonium bromide, 0.5 mL oleic acid, and 2.0 mL of toluene was swiftly added into the glass vial. A clear and green $CsPbBr_3$ quantum dot solution was formed immediately. After 10 s, the $CsPbBr_3$ quantum dots were precipitated by adding

γ-butyrolactone. Finally, the $CsPbBr_3$ quantum dots were centrifigued and redispersed in toluene for various characterizations. Note that the crude solution of quantum dots was used for long-term storage.

Synthesis of alloyed $CsPbX_3$ (X=Cl, Br, I) perovskite quantum dots

First, the oleic acid-capped $CsPbBr_3$ quantum dots were prepared in chloroform. Subsequently, the different volumes of chloroform solution of tetrabutylammonium chloride (0.02 M) or tetrabutylammonium iodide (0.2 M) were dropped into the crude solution of $CsPbBr_3$ QDs until the desired emission peak position was achieved.

Large-Scale synthesis of $CsPbBr_3$ quantum dots

First, 50 mL of oleic acid was used to dissolve 2.5 mmol Cs_2CO_3 and 5.0 mmol PbO in a 1.0 L of beaker, followed by a drying process in a 120 ^oC oven for 30 min. 900 mL of toluene was added into Cs and Pb precursor solution. Under magnetic stirring, a Br precursor solution which was prepared by dissolving 5.0 mmol tetraoctylammonium bromide into 40 mL of toluene and 10 mL of oleic acid was added into the Cs and Pb precursor solution. Finally, ~1.0 L of the quantum dot solution was achieved. The yield was estimated by precipitating 2.0 mL of QD crude solution in a pre-weighted centrifuge tube.

Characterizations

UV-vis absorption spectra were measured by Metash 5200 spectrophotometer. Photoluminescence (PL) spectra were taken using Shimadzu RF 5301PC. The relative photoluminescence quantum yields (PLQYs) of the quantum dots were determined by

comparing the integrated emission of the QD samples with a standard fluorescence dye (coumarin 6 in ethanol, QY=78% or 9, 10-diphenylanthracene in cyclohexane, QY=90%). The luminescence decay curves were obtained from a Lecroy Wave Runner 6100 digital oscilloscope (1 GHz) using a tunable laser (pulse width=4 ns, gate=50 ns) as the excitation source (Continuum Sunlite OPO). The temperature dependent PL spectra were monitored by a Maya 2000 Pro CCD-based fiber optic spectrometer (Ocean Optics Inc.) in air, and the temperature was programmed to increase from 20 ^{o}C to 100 ^{o}C. The XRD patterns were recorded using a Bruker D8 FOCUS X-ray diffractometer. High-resolution transmission electron microscopy (HR-TEM) images were taken on a FEI Tecnai G2 F20 with an accelerating voltage of 200 kV. Energy disperse spectroscopy (EDS) spectrum was recorded by using a scanning electron microscope (Hitachi S-4800) equipped with a Bruker AXS XFlash detector 4010.

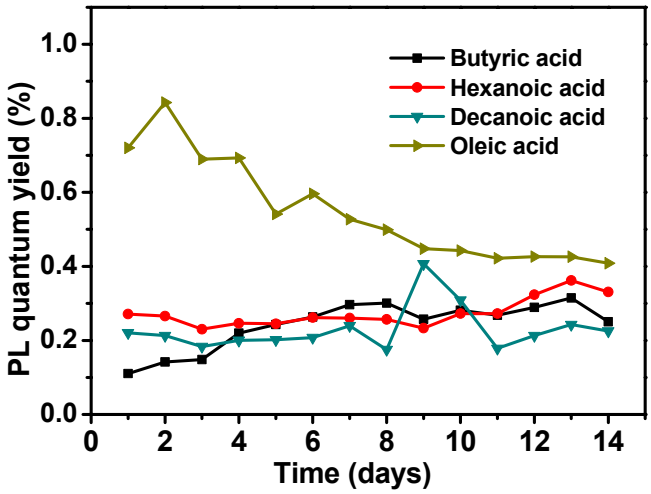

Figure S1. The PL quantum yields and stability tests of different fatty acids-capped $CsPbBr_3$ nanocrystal solution stored in ambient environment.

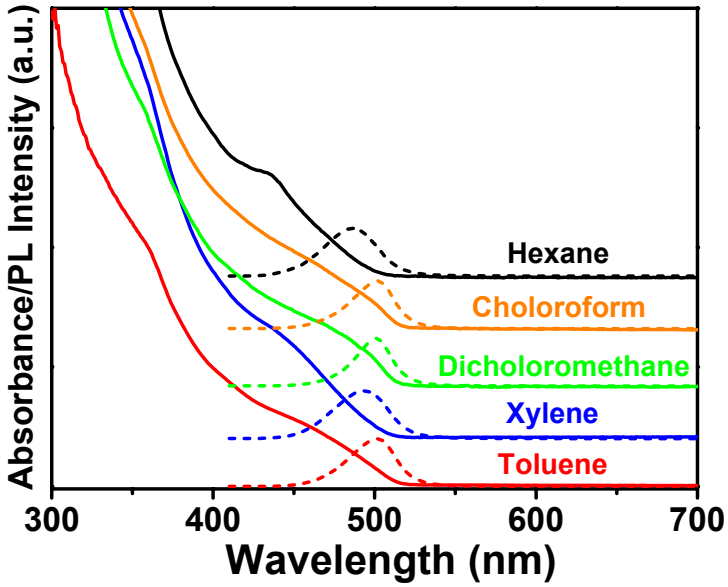

Figure S2. UV-vis absorption (solid) and PL (dash) spectra of $CsPbBr_3$ nanocrystals synthesized in different organic solvents.

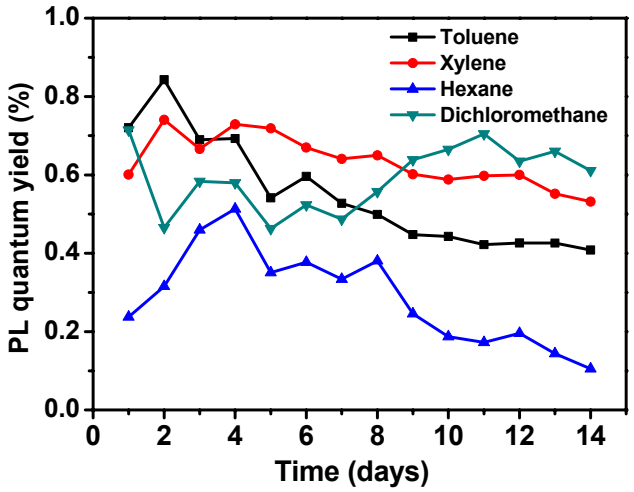

Figure S3. PL quantum yields and PL stability of oleic acid-capped $CsPbBr_3$ quantum dots synthesized in different organic solvents.

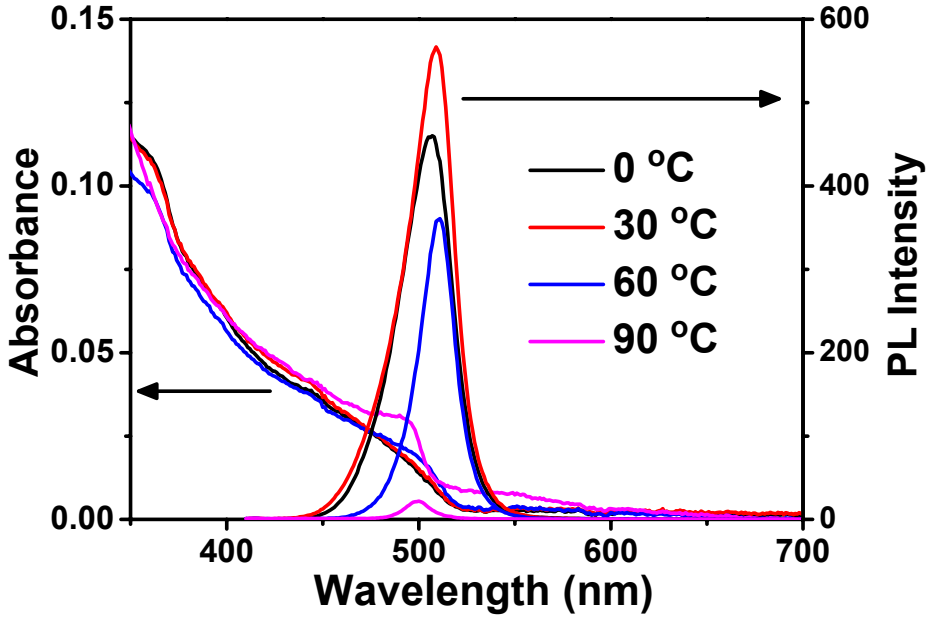

Figure S4. UV-vis absorption and PL spectra of $CsPbBr_3$ nanocrystals synthesized at different temperatures.

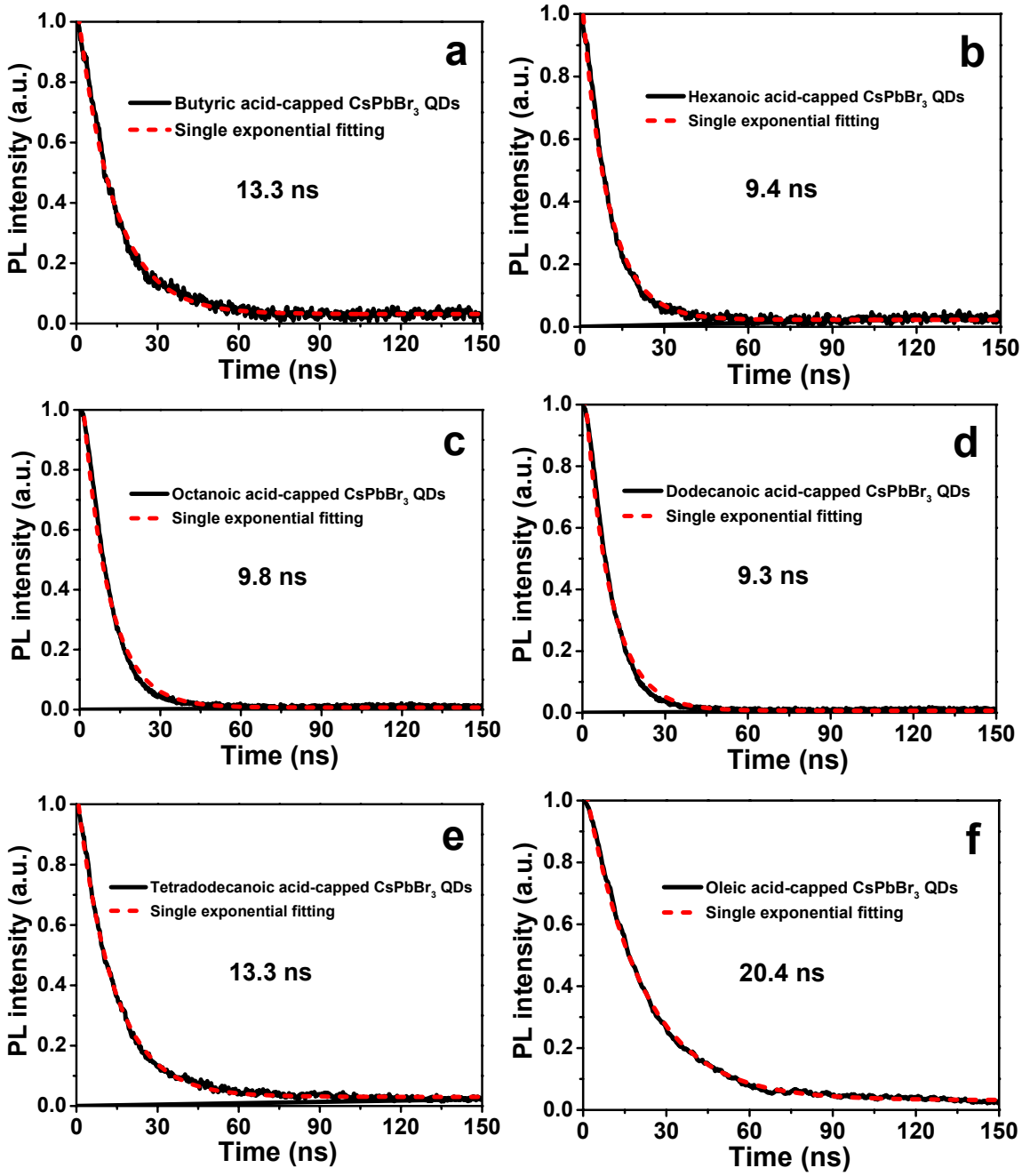

Figure S5. PL decay curves of different fatty acids-capped $CsPbBr_3$ nanocrystals.

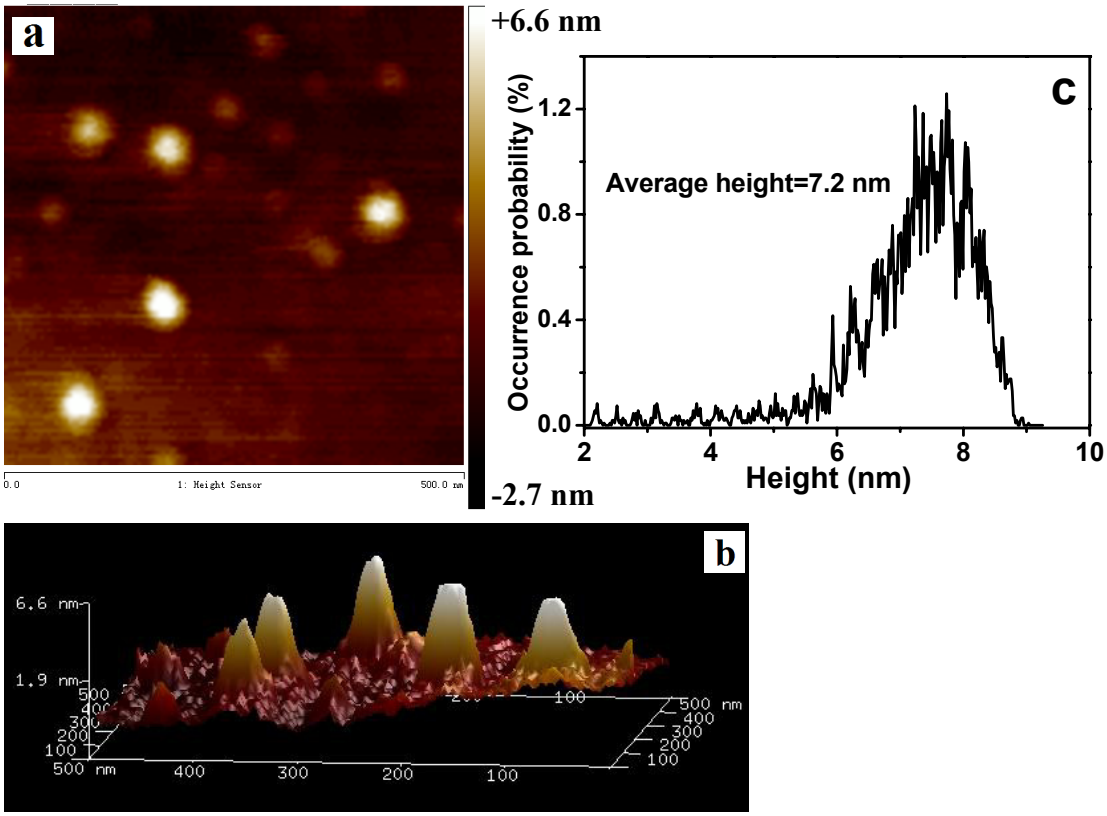

Figure S6. (a) AFM image of oleic acid-capped $CsPbBr_3$ QDs; (b) AFM 3D image of $CsPbBr_3$ QDs; (c) height distribution curve of $CsPbBr_3$ QDs.

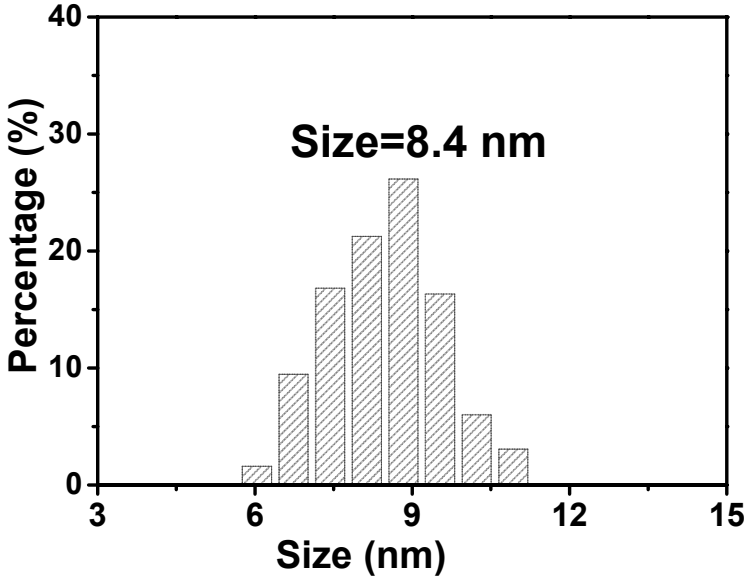

Figure S7. Size distribution of large-scale prepared $CsPbBr_3$ nanocrystals.

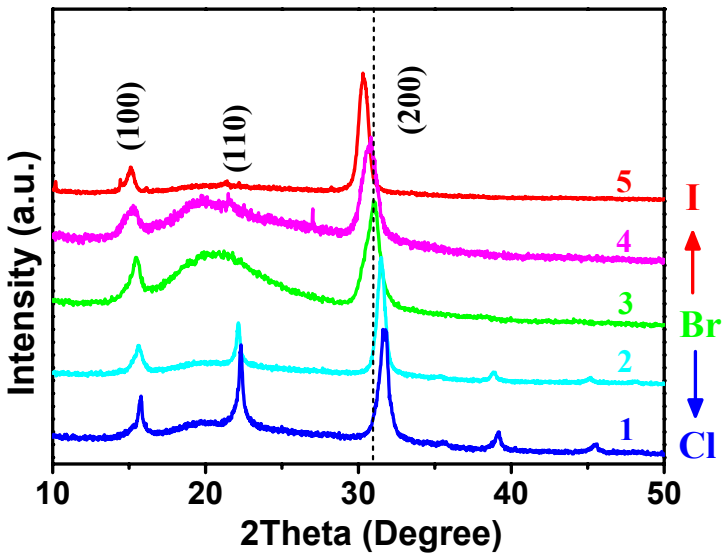

Figure S8. XRD patterns of alloyed $CsPbX_3$ (X=Cl, Br, I) nanocrystals.

Table S1. EDS results (atomic percent) of alloyed $CsPbX_3$ (X=Cl, Br, I) nanocrystals in Figure S8.

Sample	Cs%	Pb%	Cl%	Br%	I%
1	21.75	20.55	36.41	21.29	------
2	20.36	20.80	23.49	35.35	------
3	20.64	22.25	------	57.11	------
4	21.09	22.16	------	46.62	10.13
5	21.54	23.64	------	35.55	19.27

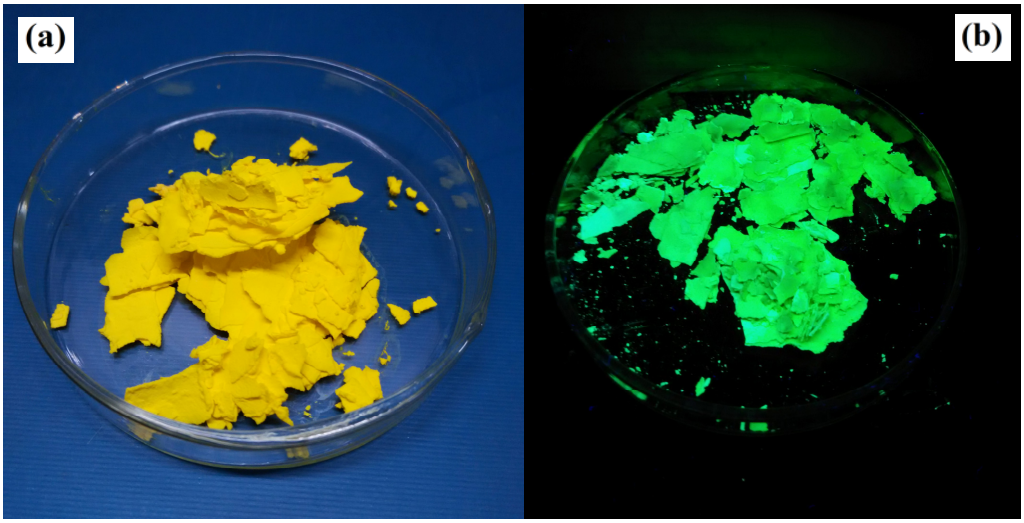

Figure S9. The photographs of the aggregated $CsPbBr_3$ nanocrystals under normal indoor light (a) and UV light (b) illumination after 30 days of air storage.

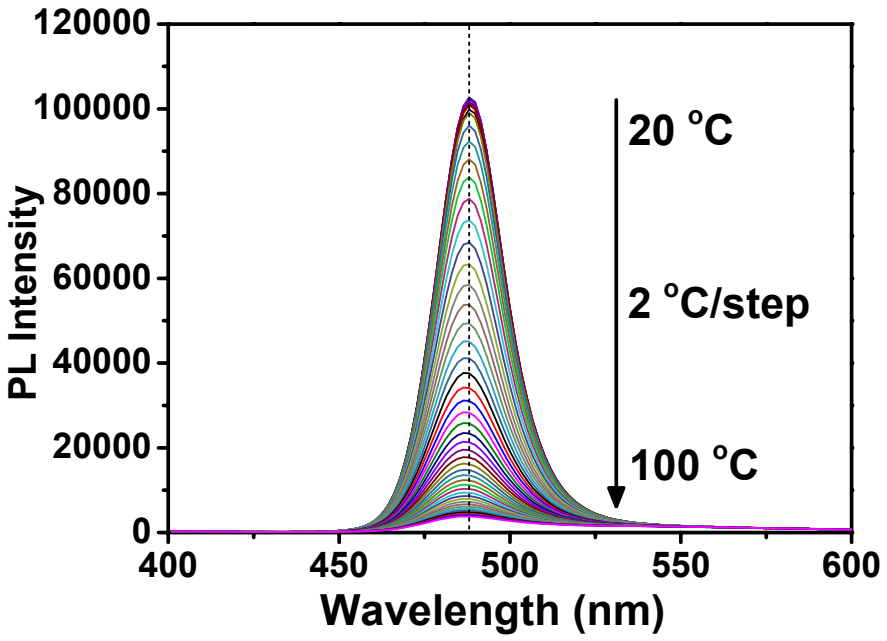

Figure S10. Temperature-dependent PL spectra of $CsPbBr_3$ nanocrystal thin film.