Supporting information

Amplified spontaneous emission and random lasing using $CsPbBr_3$ quantum dots glass through controlling crystallization

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1. Experimental

1.1 Materials

All the raw materials were from Aladdin and used without further purification. They include the following: SiO_2 (AR), B_2O_3 (99.9%), ZnO (99%), Cs_2CO_3 (99%), $PbBr_2$ (99%), and NaBr (AR, 99%).

1.2 Fabrication method for the $CsPbBr_3$ QDs glass

$CsPbBr_3$ QDs based on a borosilicate glass (B-Zn-Si) was synthesized via melt-quenching and glass crystallization methods. First, the weighed material was placed in a mortar and mixed well. Second, the mixed powder was added to an alumina crucible, placed in a muffle furnace and heated up to 1220 ℃ at a heating rate of 5 ℃/min. This temperature was maintained 10 min. Third, the melt was placed into a mold and the solidified glasses were quickly moved into a 410 ℃ annealing furnace for 150 min. Finally, the as-prepared glasses were cut into the pieces, and the $CsPbBr_3$ QDs were obtained via glass crystallization at different heat treatment (440~530 ℃) temperatures for 5 h. And all the prepared $CsPbBr_3$ QDs glasses were cut into the thickness of 0.8

nm pieces for further characterization.

1.3 Measurements and Characterization

PL spectra were used to characterize the optical properties of the as-prepared $CsPbBr_3$ QDs glass and were obtained using a spectrofluorometer (FLS920) with pulsed xenon lamps. The UV-Vis absorption spectra of the $CsPbBr_3$ QDs glass were obtained using a UV-Vis spectrometer (PerkinElmer Lambda 750). The PL decay time of the $CsPbBr_3$ QDs glasses were monitored via time-correlated single-photon counting (TCSPC) with a nanosecond-pulsed diode laser (nanoLED-370). XRD patterns within the range of 10°~80° were obtained to identify the crystal structure of the glass samples using XRD (XRD, D8 Advance, Bruker, Germany) with Cu Kα radiation operating at 40 kV and 40 mA. An FEI Tecnai F20 instrument operating at 200 kV was used to obtain the TEM, HR-TEM, and SAED. To detect the valence states of the elements, an AXI ULTRA DLD spectrometer with a monochromator and Al Kα as the X-ray source was used to obtain the XPS spectra. A QE-2100 spectrophotometer from Otsuka Japan Photal Electronics were used to measured PLQY.

The UC lasing emission of the $CsPbBr_3$ QDs glasses at different annealing temperature was recorded under 800 nm femtosecond (fs) pulsed laser excitation. Here, a Ti: sapphire laser (coherent Libra) was used as the excitation source and used to generate the 800 nm fs pulses (1kHz). A laser beam with an 8 mm diameter was focused by a convex lens with a 10 cm focal lens on the surface of the QDs glasses, which was located inside a low temperature chamber (Linkam DSC 600 temperature-controlled stage) purged with N_2 gas. The emission from the $CsPbBr_3$ QDs glass surface was recorded by an optical fiber (600 μm in diameter). The optical fiber was coupled to a monochromator (Princeton Spectra Pro 2750 integrated with a Pro EM EMCCD camera with a spectral resolution of less than 0.1 nm) for spectral analysis.

The optical gain was measured using the variable stripe length (VSL) method. The 800 nm fs pulsed laser with the diameter of 8 mm was focused by a cylindrical lens to a line on the $CsPbBr_3$ QDs glasses surface, which was located inside a low temperature chamber (Linkam DSC 600 temperature-controlled stage) purged with N_2 gas. An adjustable slit was placed between the cylindrical lens and the $CsPbBr_3$ QDs glasses surface to accurately modulate the length of the stripe. By changing the length of the strips on the $CsPbBr_3$ QDs glasses surface, the corresponding emission from the edge of the sample was collected by an optical fiber (600 μm in diameter). The optical fiber was coupled to a spectrometer (Ocean Optics USB 2000+VIS-NIR).

2. Supporting Tables

Sample	**t_1 (ns)**	**t_2 (ns)**	**A_1**	**A_2**	**τ_{avg} (ns)**
440 ℃	11.28	45.11	0.2	0.8	43.12
470 ℃	19.45	77.79	0.2	0.8	74.36
500 ℃	25.26	101.05	0.2	0.8	96.59
530 ℃	34.88	139.54	0.2	0.8	133.39

Table S1 Detailed information for calculating the decay time with the biexponential fit.

Pump density (mJ/cm^2)	**g (cm^{-1})/ 440 ℃**	**g (cm^{-1})/ 470 ℃**	**g (cm^{-1})/ 500 ℃**	**g (cm^{-1})/ 530 ℃**
0.017	-12.10	1.66	5.18	9.71
0.053	-3.40	8.31	12.58	10.90
0.088	-0.35	12.47	17.67	20.04
0.124	0.66	19.60	26.29	36.11

Table S2 Optical gain versus excitation power for the $CsPbBr_3$ QDs glasses at different heat treatment temperatures at ambient pressure and a temperature of -180 °C. The excitation wavelength was set to 800 nm.

3. Supporting Figures

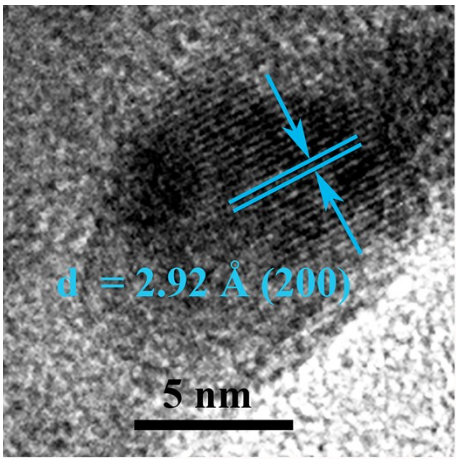

Fig. S1 HR-TEM image of the $CsPbBr_3$ QDs glass.

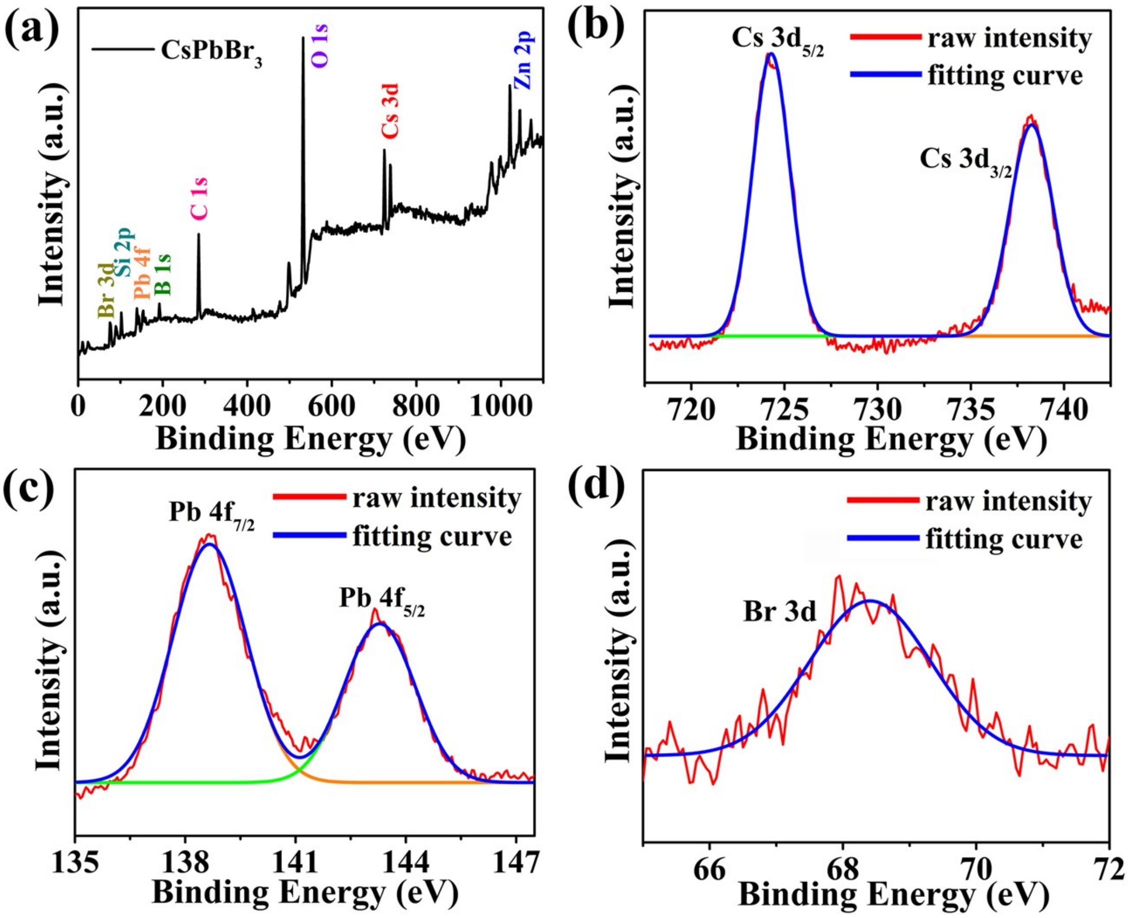

Fig. S2 (a) The survey XPS spectra of the $CsPbBr_3$ QDs glass. The high resolution XPS spectrum of **(b)** Cs 3d, **(c)** Pb 4f, **(d)** Br 3d.

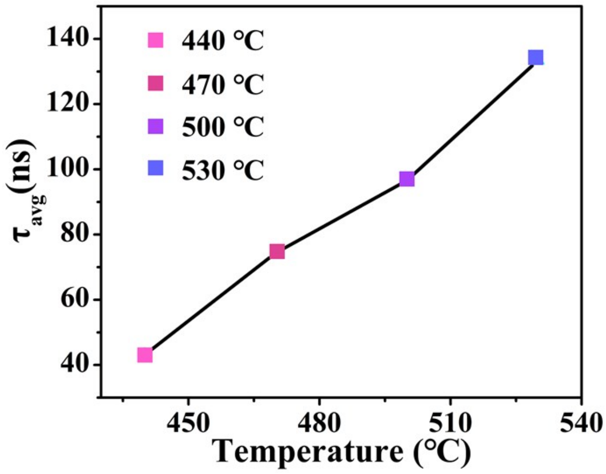

Fig. S3 The line graph shows the relationship between the τ_{avg} and the heat treatment temperature.

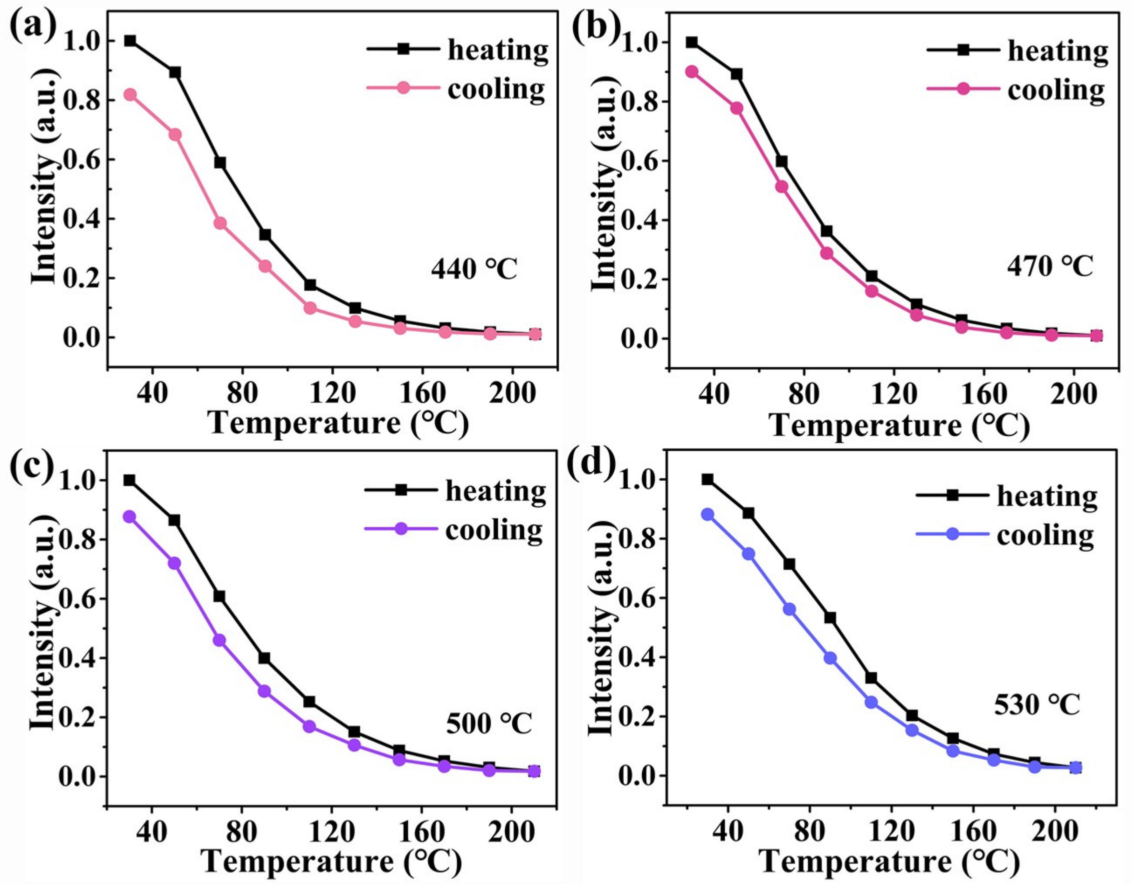

Fig. S4 The relationship between the integral emission intensities and thermal cycle of $CsPbBr_3$ QDs glasses heat-treated at **(a)** 440 ℃, **(b)** 470 ℃, **(c)** 500 ℃ and **(d)** 530 ℃. The emission intensity of all the samples were maintained above 80% of the original intensity, although they have experienced a 30 ~ 210 °C heating and cooling cycles process.

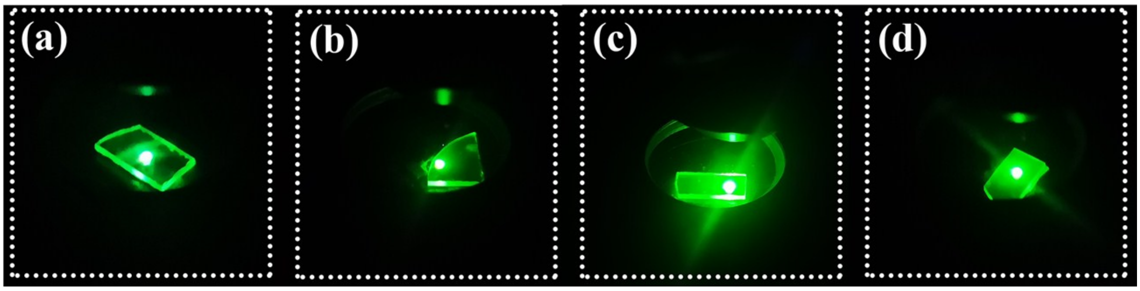

Fig. S5 Photographs of various $CsPbBr_3$ QDs glasses with different heat treatment temperature under of 800 nm fs pulsed laser excitation. **(a)** 440 °C, **(b)** 470 °C, **(c)** 500 °C, and **(d)** 530 °C.

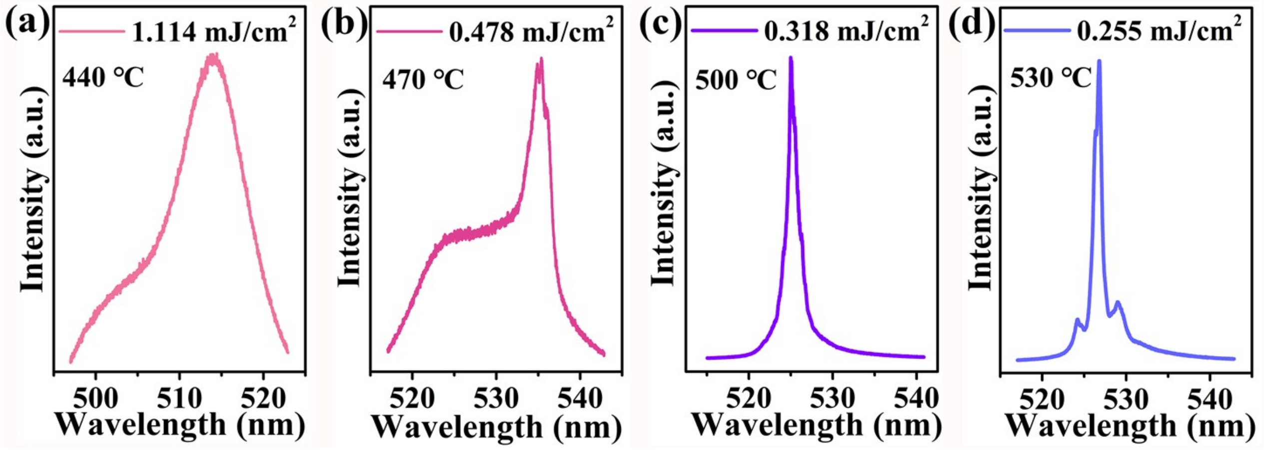

Fig. S6 The UC emission for the $CsPbBr_3$ QDs glasses heat-treated at **(a)** 440 °C, **(b)** 470 °C, **(c)** 500 °C, and **(d)** 530 °C. The results show that a higher heat treatment temperature makes it easier to achieve random lasing. The random lasing spectra shows a shark peak.

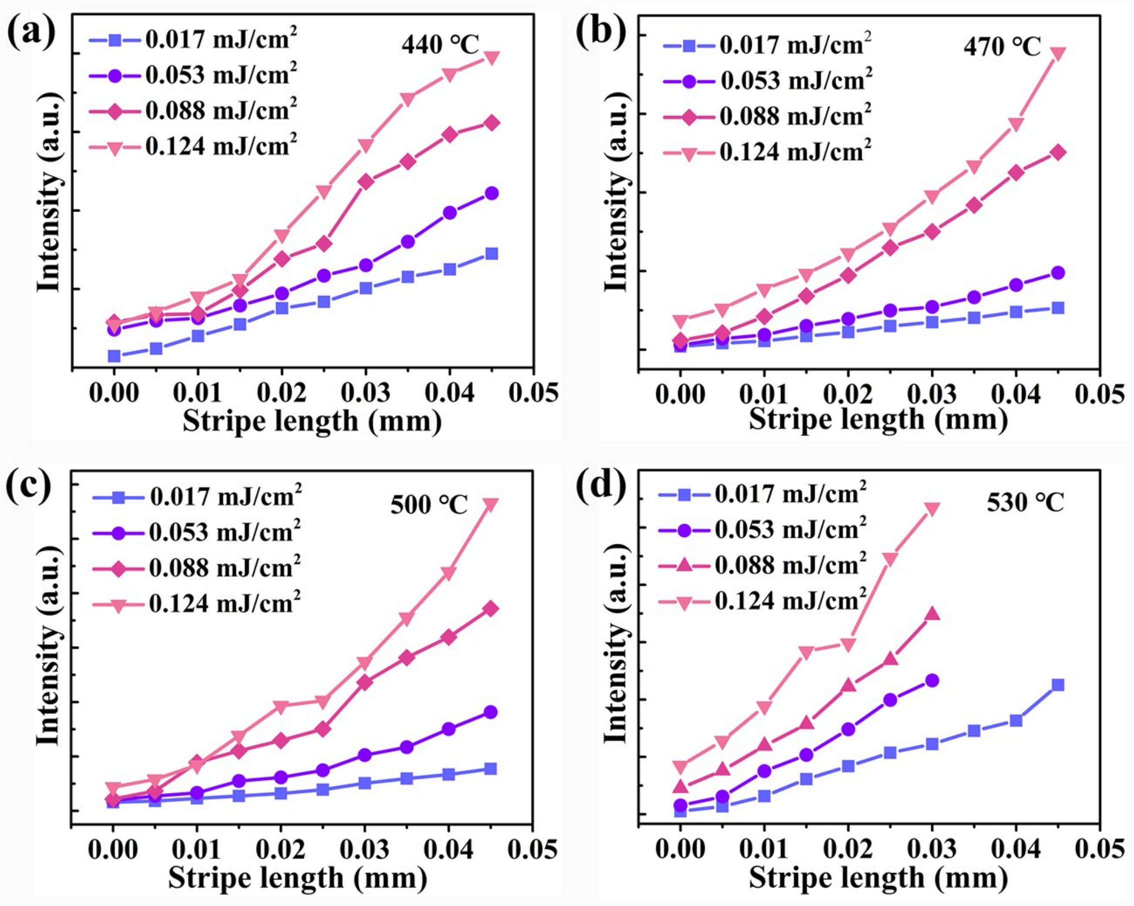

Fig. S7 Stripe-length dependent emission spectra with a pump intensity of 0.017 mJ/cm^2, 0.053 mJ/cm^2, 0.088 mJ/cm^2, and 0.124 mJ/cm^2 for $CsPbBr_3$ QDs glasses heat-treated at different temperatures. **(a)** 440 ℃, **(b)** 470 ℃, **(c)** 500 ℃, and **(d)** 530 ℃. The experiments were carried out at ambient pressure and at a temperature of -180 ℃.