A Simple Multiple Centrifugation Method for Large-area Homogeneous Perovskite $CsPbBr_3$ Films with Optical Lasing

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1 Experimental Section

1.1. Chemicals. All chemical reagents were of analytical grade and were used as received without further purification. oleic acid(OA), oleylamine (OAm, 80-90%), lead bromide ($PbBr_2$, 98.00%) and cesium bromide (CsBr, 99.99%) were purchased from Aladdin. dimethylsulfoxide (DMSO), toluene and alcohol were purchased from Sinopharm Chemical Reagent Company of china.

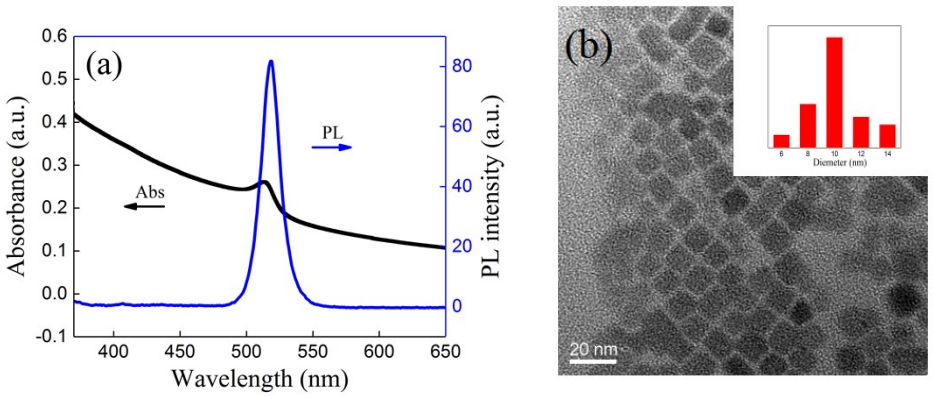
1.2. Preparation of the $CsPbBr_3$ solution. First, dissolved the CsBr (0.085 g) and the $PbBr_2$ (0.146 g) in the dimethylsulfoxide (DMSO, 10 mL) solution. Second, mixed the prepared solution with oleic acid (OA, 1 mL) and oleylamine (OAm, 0.5 mL) sufficiently. Finally, added the prefabricated solution (1 mL) into the toluene (10 mL) solution and stir continuously, the $CsPbBr_3$ solution was prepared.

1.3. Preparation of the $CsPbBr_3$ films. (1)Fixed the substrate in the centrifuge tube, and the centrifuge time and speed was kept constant 2 minutes and 8500 r/min respectively. Then took the centrifuge tube out when the centrifugation was finished and removed the liquid wastes, added the new $CsPbBr_3$ solution to repeat centrifugate until the films meet the desired thickness. (2)Took the substrate out and waited the evaporation of the toluene on the film, and then put the sample in the center of the cuvette on the heating stage (70 ℃) and drop the reagent solution (50 μL) mixed with the alcohol and toluene at a rate of 5:2 surrounded the film (about 15 minutes). The enclosed environment make it possible that the steam interact with the film sufficiently, which make the films compact and small roughness.

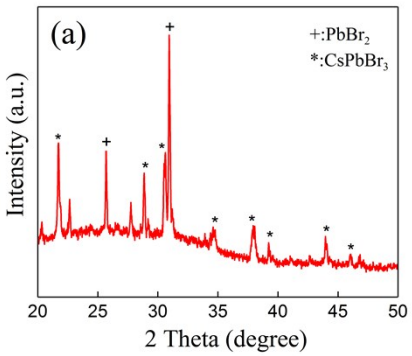
1.4. ASE and lasing Measurements. For the ASE measurements, all experiments were conducted at room temperature. The excitation source used for this work was a Ti: sapphire oscillator/amplifier. The latter produced ~120 fs duration, 800 nm wavelength laser output with a repetition rate of 1 kHz. The output divergence angle after collimation was around 0.65 mrad. By using a beta barium borate (BBO) crystal, the output wavelength was converted to 400nm. The pump beam was focused by a cylindrical lens on the sample, and the emission from the edge

of the film serving as the optical waveguide was vertically collected by a sepectrograph (spectrapro-300i, Acton research corporation). For the time-resolved PL measurements, the conventional backscattering configuration was adopted for the PL collection, it is the same as used in ASE measurements. The emission was detected and analyzed by a streak camera (c5680-04/M, Hamamastu).

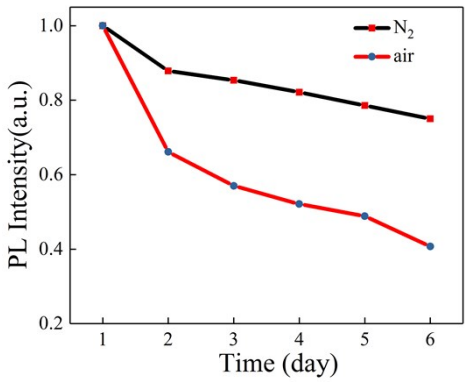
1.5 Device Measurements. The X-ray diffraction (XRD) patterns were recorded using a XRD measurement instrument (Smartlab, RIGAKU). Transmission electron microscopy (TEM) images were taken with an accelerating voltage of 200 kV (Tecnai G20, FEI). X-ray photoelectron spectroscopy (XPS) was performed using a PHI550 spectrometer (ULVAC) with Mg Kα excitation (1253.6 eV). The atomic force microscope (AFM) were recorded using a microscope (Bioscope Resolve, Bruker). We measured the thickness through cutting a line on the film with a knife, the depth of the line reached the substrate, thus, the height of the scratch section is the thickness of the film. Scanning electron microscopy (SEM) images were taken with an accelerating voltage of 20 kV (Inspect F50, FEI). The absorption and PL intensity spectra were measured through a spectrophotometer (UV-3600, SHIMADZU) and spectrofluorophotometer (RF-5301PC, SHIMADZU) respectively.



S1. The absorption/photoluminescence (PL) spectrum of the $CsPbBr_3$ QDs solution (figure (a)) displays an obvious peak at 514/518 nm. The transmission electron microscope (TEM) of the $CsPbBr_3$ QDs is illustrate in figure (b), and the QDs with an average size of 10.1 nm.



S2. The XRD pattern of the $CsPbBr_3$ film,The strong intensity of the diffraction peaks depicts the good crystallinity of the film. Most peaks are matching well with the standard (rhombic) $CsPbBr_3$ (PDF#18-0364). The peaks at degree of 24.4 and 30.9 come from $PbBr_2$ (PDF#18-0679), which may be caused by the incomplete reaction during fabrication process.



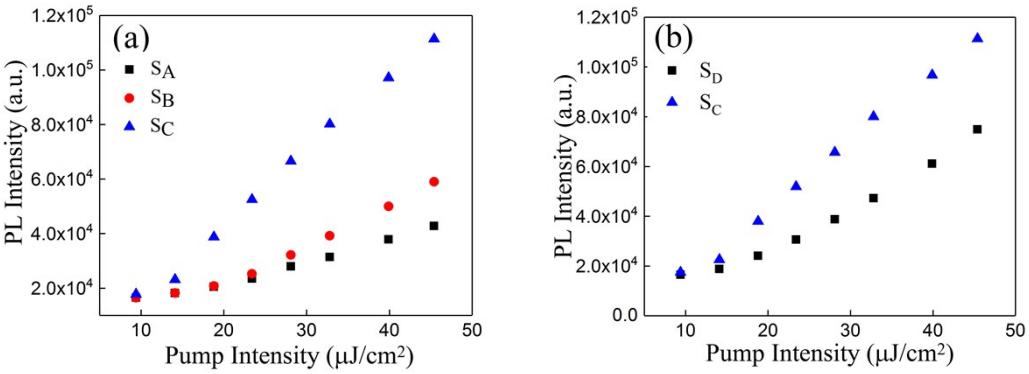
S3. The stability of the PL intensity versus the time

Tabe S1: The ASE threshold and the gain coefficient of the $CsPbBr_3$ film before and after solvent annealing with the thickness of 880 nm.

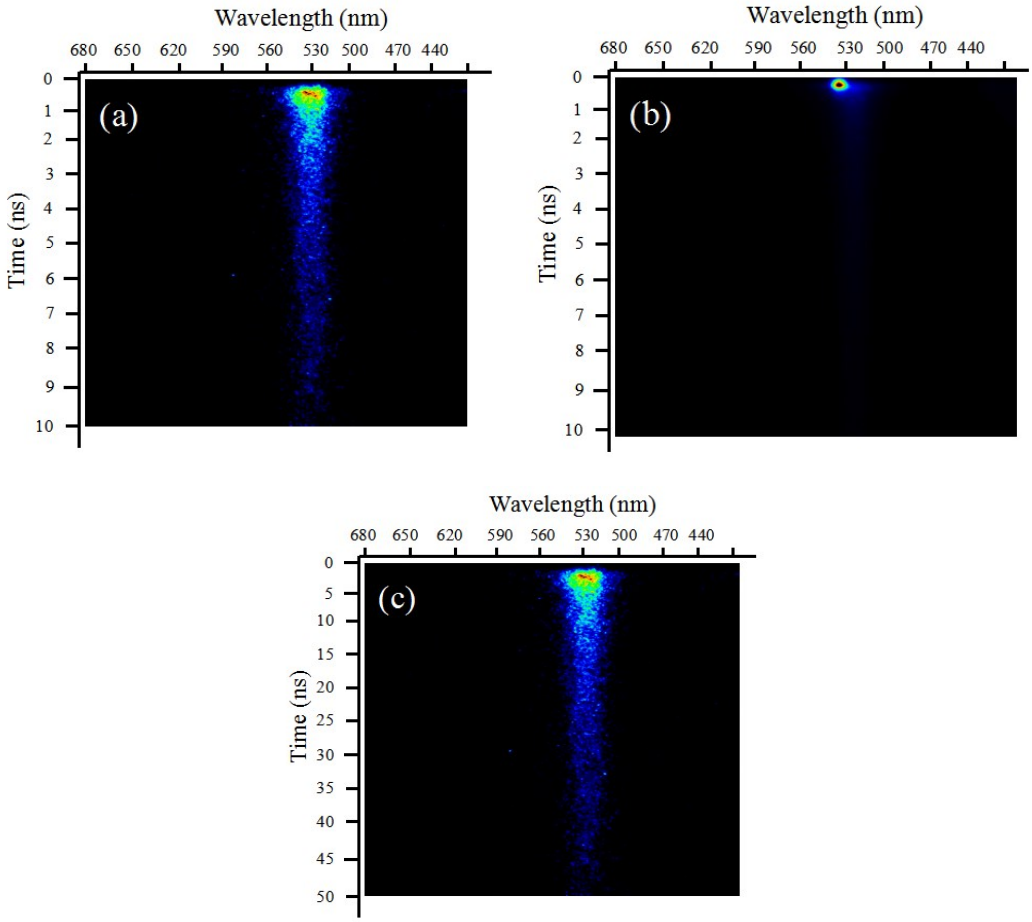
Parameter	Before solvent annealing	After solvent annealing
ASE threshold/μJ. cm^{-2}	16.2	14.2
Gain coefficient /cm^{-1}	112.7	122.7

Table S2: The ASE threshold and the gain coefficient of the $CsPbBr_3$ film with the thickness of 180 nm, 500 nm and 880 nm, respectively.

Thickness/nm	180 nm	500 nm	880 nm
ASE threshold/μJ. cm^{-2}	18.1	17.4	14.2
Gain coefficient /cm^{-1}	78.5	96.3	122.7



S4: The PL intensity versus the pump intensity of (a) S_A, S_B and S_C (b) S_C and S_D.



S5: The PL decay of the $CsPbBr_3$ film with the pump intensity (a) 10.7 μJ/cm^2 and (b) 25.0 μJ/cm^2 (c) The PL decay of the $CsPbBr_3$ QDs solution.

Table S3: The comparison of the threshold with different synthesis method.

Method	Threshold (μJ. cm^{-2})	reference
Spin-coating	207, 192	1,2
Thermal evaporation	6.2	3
MCDSA	14.1	

reference

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