Supporting Information

Photon-Induced Reshaping in Perovskite Material Yields Nanocrystals with Accurate Control of Size and Morphology

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Experimental Procedures

***Preparation of the $CsPbBr_3$ nanocrystals*:** $PbBr_2$ (99.99% metals basis), and CsBr (99.9% metals basis) were custom-made from Macklin Biochemical Co., Ltd. Unless otherwise expressly noted, all reagents and solvents were obtained from commercial sources and used directly without further purification. Acetone and toluene were from Sinopharm Chemical Reagent Co., Ltd. The $CsPbBr_3$ nanocrystals were prepared by the traditional hot injection growth method: the Cs-oleate precursor solution was prepared in a 50 mL three-necked round-bottomed flask by dissolving Cs_2CO_3 (0.25 g) in a mixture of oleic acid (OA, 0.8 g) and 1-octadecene (ODE, 7g) at 150 ℃ for over 10 min under N_2 atmosphere. The temperature of the Cs-oleate solution was maintained above 100 ℃ to avoid precipitation. The precursor solution of Pb and Br was prepared by dissolving 75 mg $PbBr_2$ and 5 mL ODE, OA (2 mL), and oleylamine (OAm, 2 mL) in a 50 mL three-necked round-bottomed flask under N_2 atmosphere at 120 ℃ for 10 min. After the setting of the temperature of the precursor solution of Pb and Br at a chosen reaction temperature, 0.4 mL of Cs precursor solution was injected to initiate the reaction. The reaction was quenched after a short period of time (60 s) by cooling the flask in an ice bath. The product was centrifuged at 3500 rpm to remove the unreacted salts as the precipitate, and the quantum dots (QDs) dispersed in the supernatant were collected. The QDs in the recovered supernatant solution was subsequently precipitated by adding ~ 8 mL of acetone.

Preparation of the $CsPbBr_3$ micron powders: The $CsPbBr_3$ micron powders were prepared by the traditional crystallization method at room temperature: excessive $PbBr_2$ and CsBr was

dissolved in the dimethyl sulfoxide (DMF). The solution was then injected into the toluene. The orange flocculent sediment was then collected after centrifuging and drying.

Irradiation experiments: All irradiation experiments were carried out using 50 mL round-bottomed flask. 0.03 g $CsPbBr_3$ micron powders were dispersed in 40 mL toluene with 200 μL OA and 200 μL OAm.

Laser equipment: Laser pulses were generated by a neodymium-doped (Nd): yttrium aluminum garnet (YAG) laser (1064 nm, 532 nm, 355 nm in wavelength, 10 Hz in frequency, and 8 ns in pulse duration)**.** During the irradiation experiments, all the samples were stirred at a speed of 300 rpm using a magnetic bar.

Characterization: The XRD patterns of the as-synthesized products were obtained by a Bruker D8 Advance XRD system using CuK radiation. The SEM images were obtained on a Quant 250 FEG SEM instrument. TEM measurement was carried out on an FEI Tecnai G20 with a Cu grid. The photoluminescence and absorption spectra were detected with a Varian Cary Eclipse instrument and Shimadzu UV-3600, respectively. The fluorescence decay was recorded with time-correlated single-photon counting (TCSPC) technique on an Edinburgh FLS920 phosphorescence lifetime system equipped with a 375 nm laser at room temperature.

Finite Element Method (FEM): We use the Finite Element Method (FEM) to solve the following heat transport equation:

$$\rho C_p \frac{\partial}{\partial t} T = \nabla \cdot (\kappa \nabla T) + Q$$

where ρ is the density, C_p is the specific heat, κ is the thermal conductivity and Q is the heat source. Values for ρ, C_p and κ can be found in the literature for both the Perovskite quantum dot and the solvent. The heat source Q is defined as:

$$Q = \frac{P_0 \sigma_{abs}}{a^3} = \frac{P_0 2\pi}{a^3\ \lambda} a^3 \eta\ = \eta P_0 \frac{2\pi}{\lambda}$$

where P_0 is the power density (W/m^2) of the incoming laser beam, λ is the wavelength, σ_{abs} is the absorption cross-section of the quantum dot and a is the side length. Im(α) is a dimensionless parameter closely related to the optical constant of the quantum dot. For simplicity, we set it as 0.5 in our simulation and the heat source in the quantum dot region is simply set to be $2\pi P_0/\lambda$ (W/m^3).

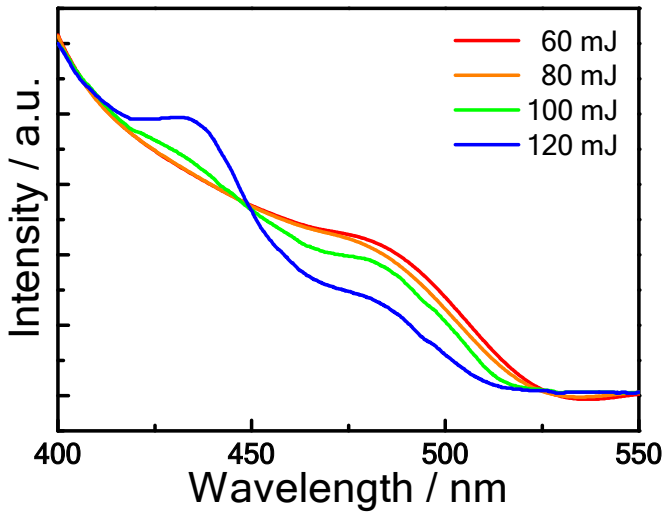

Figure S1. The UV-Vis spectrum of 0.03 g $CsPbBr_3$ micron powders dispersed in 40 mL toluene with 200 μL OA and 200 μL OAm under 355 nm laser irradiation with different power intensity for 1 hour.

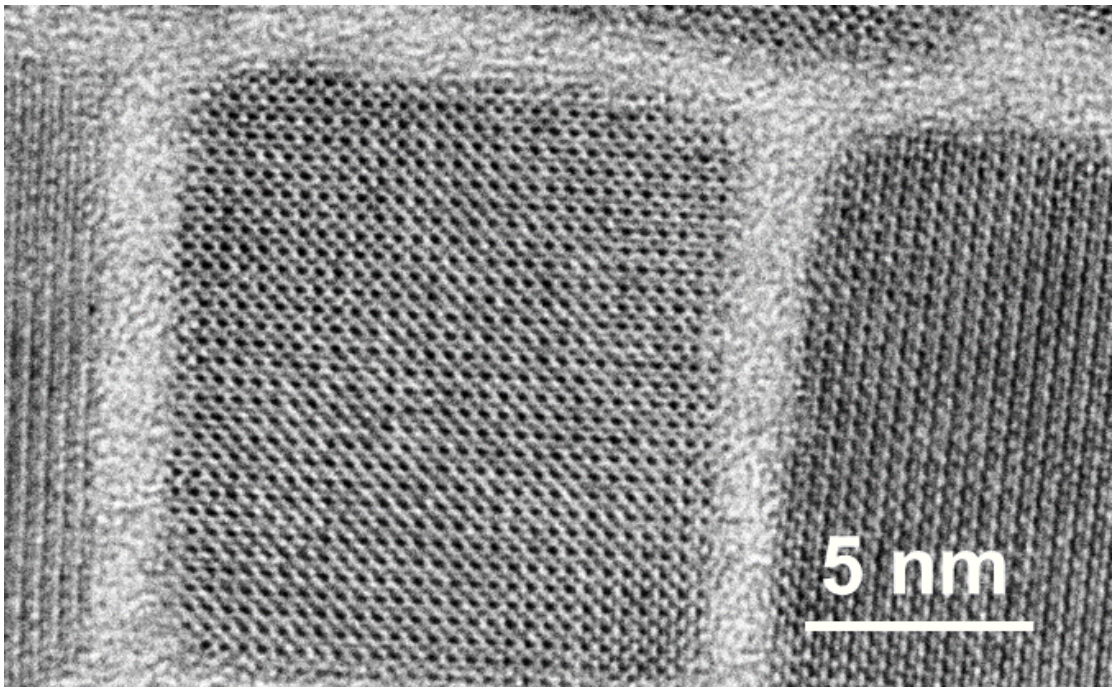

Figure S2. The TEM image of $CsPbBr_3$ micron powders dispersed in toluene with OA and OAm under 532 nm laser irradiation for 3h.

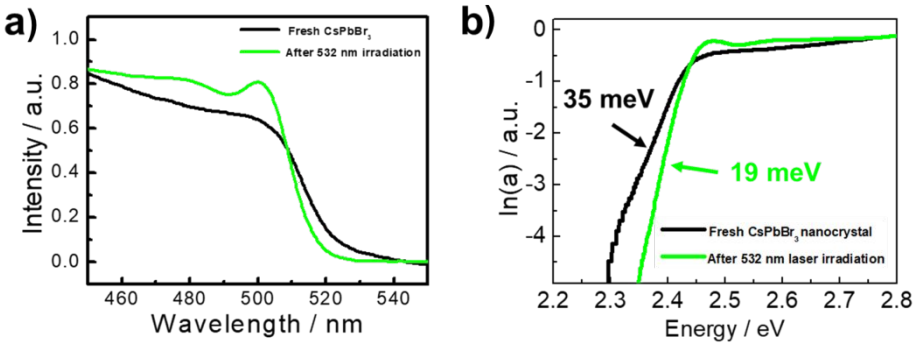

Figure S3. a) the effective absorption coefficient of $CsPbBr_3$ with/without laser irradiation and b) their corresponding Urbach Energy.

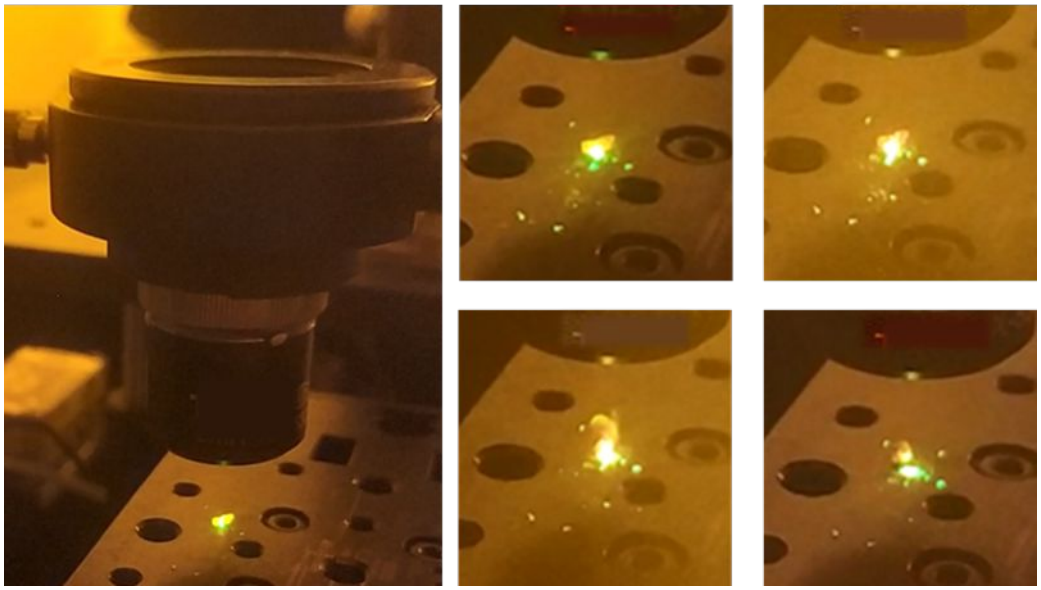

Figure S4. The images of laser-induced the fracture in $CsPbBr_3$ single crystal.

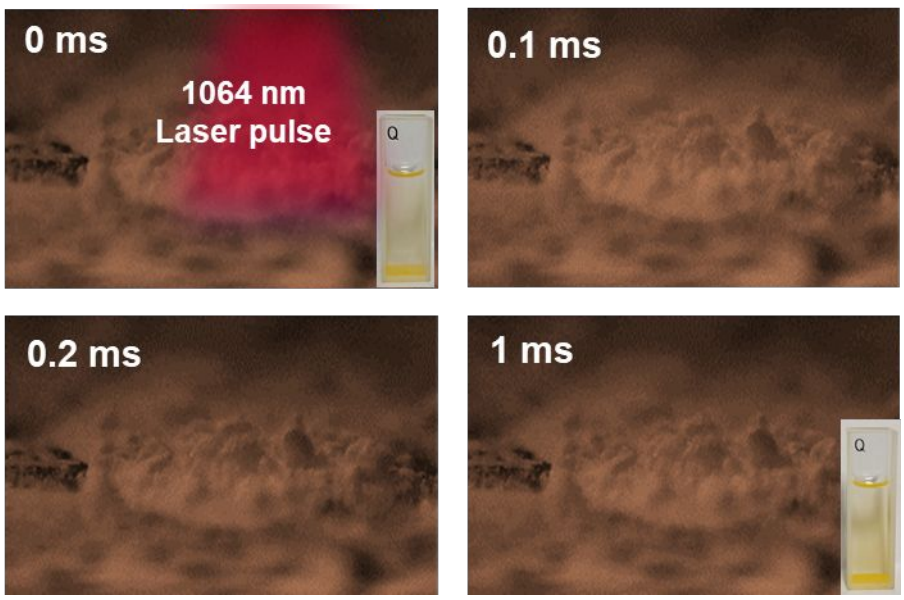

Figure S5. The high-speed camera images of $CsPbBr_3$ powder dispersed in toluene with OA and OAm under 1064 nm laser irradiation.

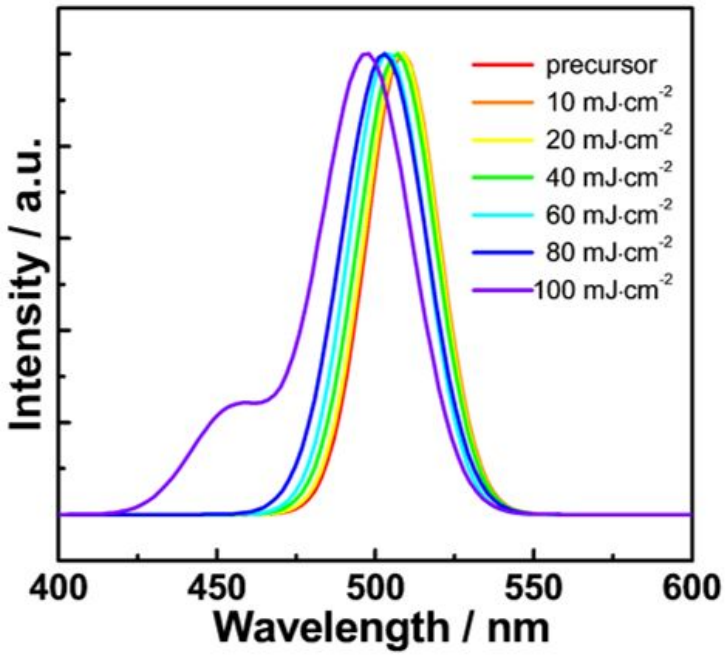

Figure S6. the PL emission of the $CsPbBr_3$ nanocrystals under different laser irradiation power intensity at a fixed wavelength of 355 nm for 1 hour.

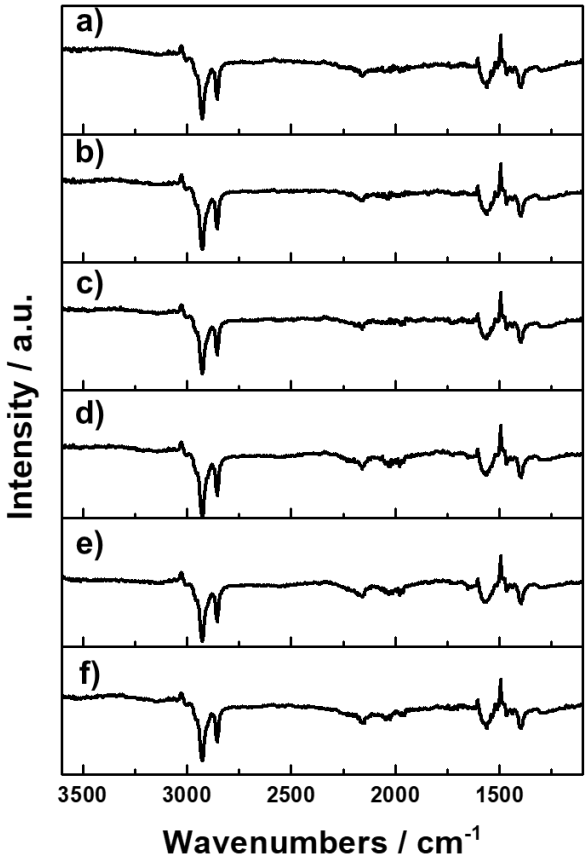

Figure S7. The FTIR spectrum in different experimental conditions of the laser irradiation process in toluene. a) OA and OAm without laser irradiation. b) OA and OAm irradiated by 355 nm and c) 532 nm laser for 1 hour. d) OA, OAm and $CsPbBr_3$ nanocrystals irradiated by 355 nm and e) 532 nm laser for 1 hour. f) OA, OAm and $CsPbBr_3$ powders irradiated by 355 nm laser for 1 hour.

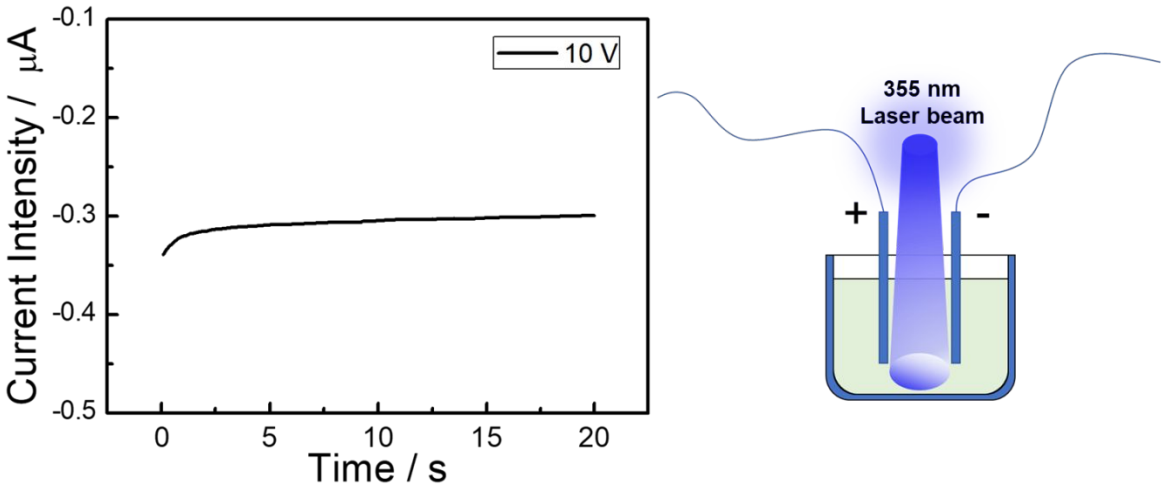

Figure S8. The I-V test and schematic diagram of the $CsPbBr_3$ nanocrystals dispersed in toluene during laser irradiation with the bias voltage of 10 V.