Supporting Information

Pyramid-Shaped Quantum Dot Superlattice Exhibiting Tunable Room-Temperature Coherent emission via Oriented Attachment

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CONTENTs

1. EXPERIMENTAL SECTION

1.1 Materials

Lead(II) bromide ($PbBr_2$, 99.999%), zinc bromide ($ZnBr_2$, 99.999%), cesium carbonate (Cs_2CO_3, 99.9%), oleylamine (OAm, 70%), oleic acid (OA, 90%), 1-octadecene (ODE, 90%), ethyl acetate (≥99.5%) and hexane (≥95%) were purchased from Sigma-Aldrich. All chemicals were used without further purification.

1.2 Preparation of precursors

Pb-oleate precursor: The Pb-oleate precursor solution was prepared by dissolving $PbBr_2$ (75 mg) and $ZnBr_2$ (414 mg) in OAm (2 mL), OA (2 mL) and ODE (5 mL), followed by degassing at 100℃ for 30 min, then heating to 120°C under an Ar atmosphere for 1 h.

Cs-oleate precursor: The Cs-oleate precursor solution was prepared by dissolving Cs_2CO_3 (0.25 g) in OA (0.8 mL) and ODE (7 mL), followed by degassing at 120℃ for 30 min, then heating to 150°C under an Ar atmosphere for 1 h, then keep the temperature at 100°C.

1.3 Synthesis of $CsPbBr_3$ QDs nanocrystal solution

Pb-oleate precursor were loaded into a three-necked flask and the solution was heated to 190°C under N_2 atmosphere. 0.4 mL Cs-oleate precursor be injected at 190°C. The reaction mixture was cooled to room temperature after 10 min, and the crude product

was purified by ethyl acetate, and re-dispersed in hexane.

1.4 Self-assembly of QD superlattices.

TEM carbon grids in a glass vial 200 μL of QD nanocrystal solution and TEM carbon grids were added in a glass vial. The vial was then placed at an angle and the toluene solvent toluene was allowed to slowly evaporate. After 2 days, self-assembled QD nanocrystal superlattices were obtained. A certain amount of oleic acid was added to the QD nanocrystal solution, and the prepared solution was then dropped onto the Si wafer with some drops of toluene solvent around the wafer. Subsequently, a culture dish was used to cover the wafer. After 8 hours, a mesocrystalline superlattice was obtained after undergoing thermal annealing treatment.

1.5 Characterization

Transmission electron microscope (TEM) characterization: TEM and selected area electron diffraction images were obtained using a JEM-2100HR operating at 200 kV. TEM and HAADF-STEM images were obtained using a FEI Talos F200X operating at 200 kV.

Scanning electron microscopy (SEM) characterization. SEM imaging of mesocrystalline QD superlatticess on Si/SiOx devices was performed with a ZEISS Gmeini 500 at 20 kV.

Time-Resolved Photoluminescence (TRPL). TRPL experiments were performed using a confocal microscope (WITec, alpha-300) as the collection device, and the

emission signal was reflected into astreak camera (C10910, Hamamatsu) by Ag mirrors. Ti:sapphire laser pulses at 400 nm (with a repetition rate of 80 MHz and a pulse width of 80 fs) were used as the light source. The 400 nm output was generated by an 800 nm laser from a mode-locked oscillator (Tsunami 3941-X1BB, Spectra-Physics) positioned after a BBO crystal. The laser beam was focused onto the sample with a spot diameter of~3μm from the top by an objective lens (50×, Zeiss, 0.75 NA).

Confocal fluorescence imaging. The fast and completely integrated fluorescence lifetime imaging microscopy (FLIM) confocal platform was performed using Leica STELLARIS 8 FALCON, provided by Leica Microsystems (Shanghai) Trading Co., Ltd.

2. SUPPLEMENTARY FIGURES

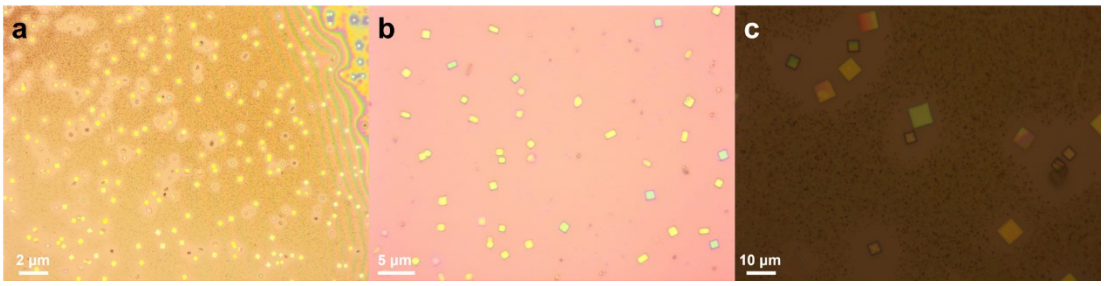

Figure S1 Optical microscope image of cube-shaped QD superlattices before annealing at a magnification of a) 10x, b) 20x, c) 50x.

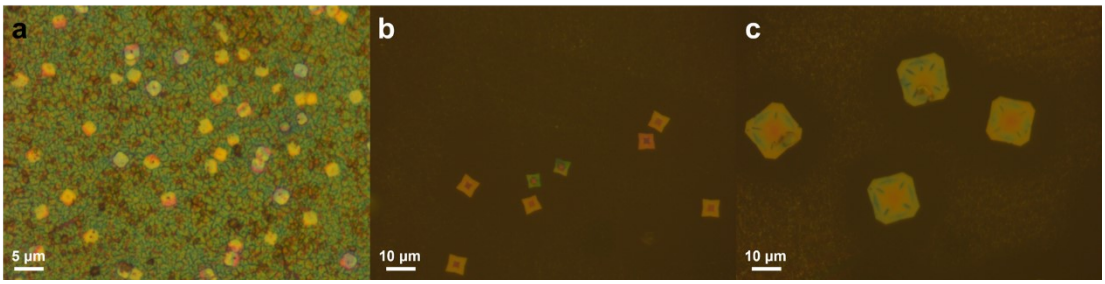

Figure S2 Optical microscope image of pyramid-shaped mesocrystalline QD superlattices after annealing at a magnification of a) 20x, b) 50x, c) 50x.

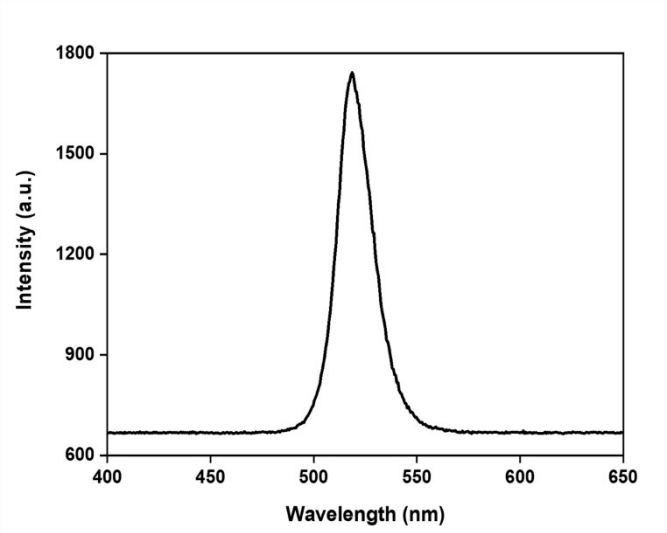

Figure S3 The PL spectrum of a single mesocrystalline QD superlatticess under an excitation density of 340 µJ/cm^2 at 300K.

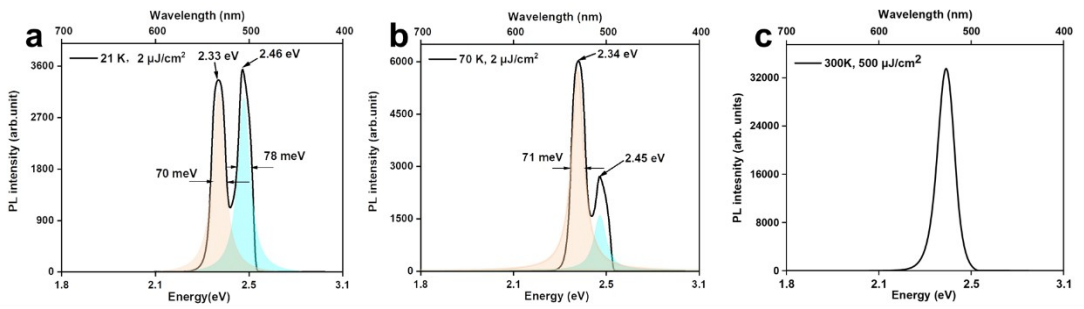

Figure S4 The PL spectrum of QD superlattices at a) 21 K , b) 70 K, and c) 300 K.

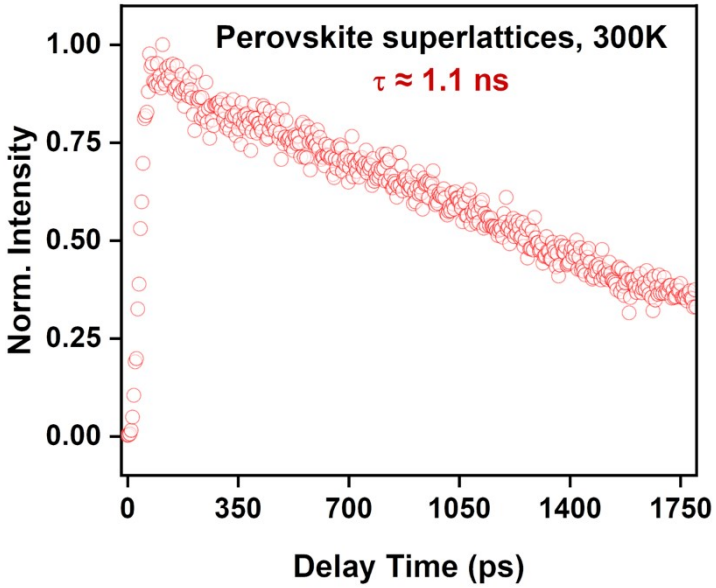

Figure S5 TRPL dynamics of the QD superlattices feature at 300 K.

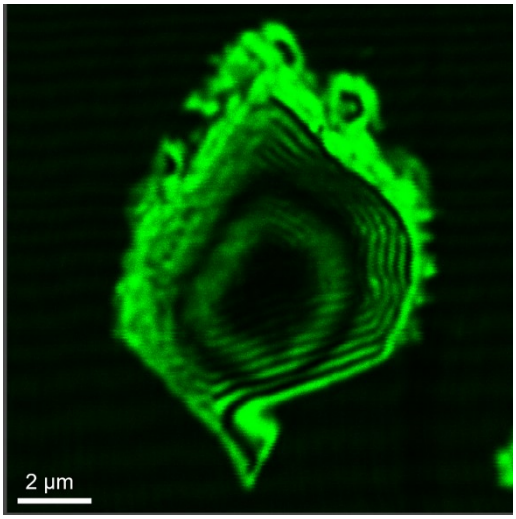

Figure S6 Confocal fluorescent imaging of mesocrystalline QD superlattices.

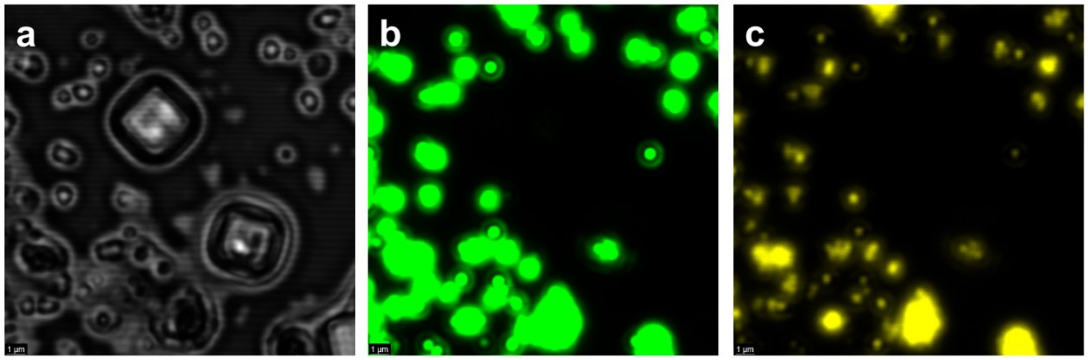

Figure S7 a) Optical images observed under dark field conditions, and confocal PL images in b) 2.2 eV and c) 2.4 eV channels of the same mesocrystalline QD superlattices were acquired using a confocal PL microscope.

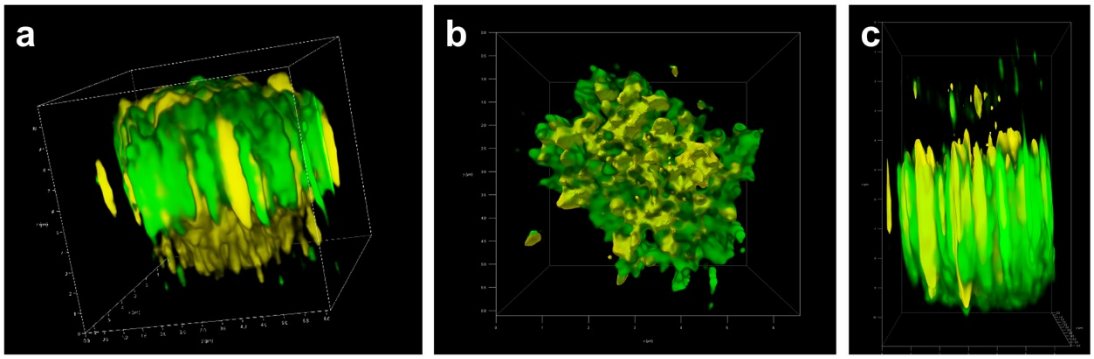

Figure S8 a) Stereo view, b) top view and c) left view of the 3D construction of a single mesocrystalline QD superlattice' confocal microscopy imaging.

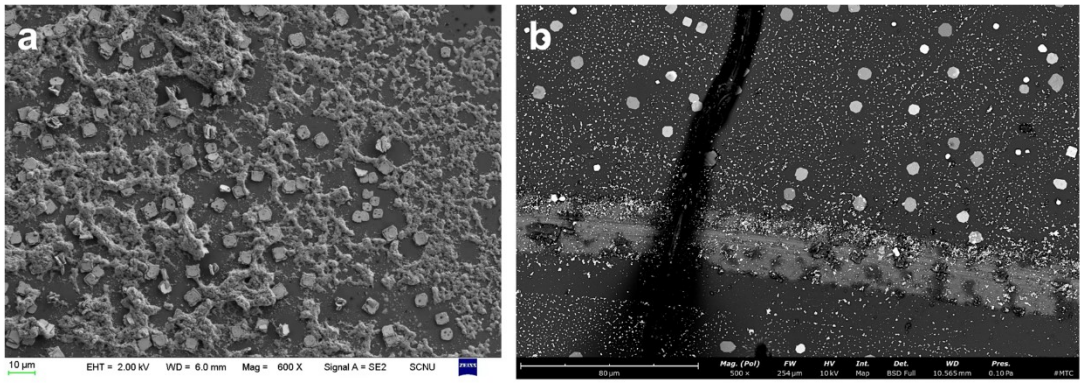

Figure S9 SEM images of mesocrystalline QD superlattices annealing at 60 degrees Celsius for a) 2 and b) 10 minutes.

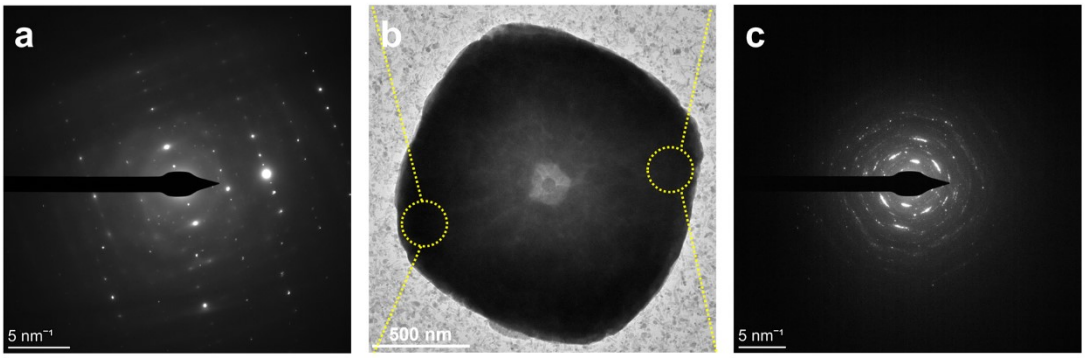

Figure S10 a) SAED patterns obtained from the left area of b). b) TEM image of mesocrystalline QD superlattices. c) SAED patterns obtained from the right area of b).

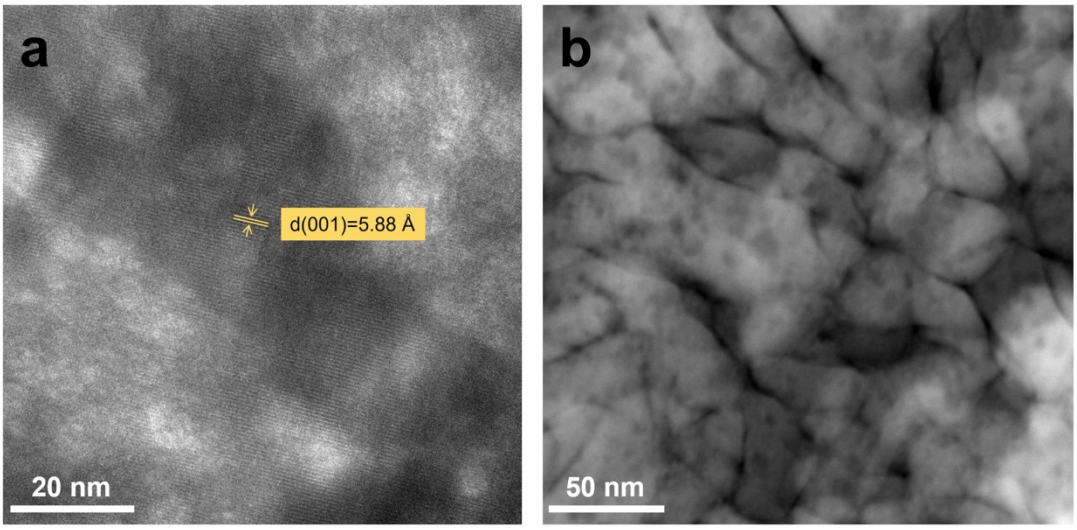

Figure S11 a) HR-TEM and b) STEM patterns obtained from the central area of the same mesocrystalline QD superlattice.

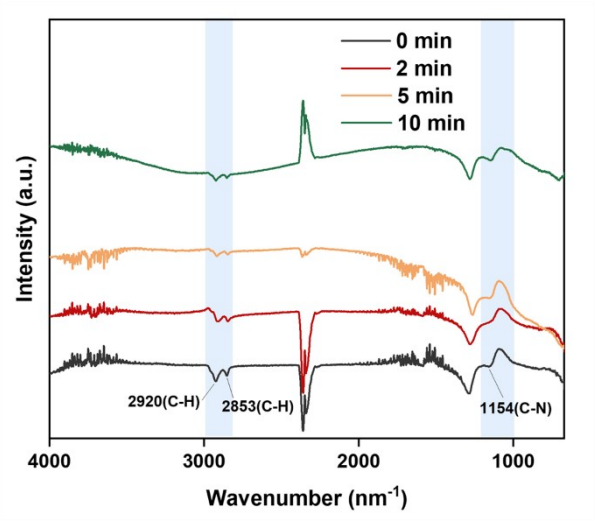

Figure S12 IN-SITU FT-IR change of mesocrystalline QD superlattices with annealing time at 60 ℃.

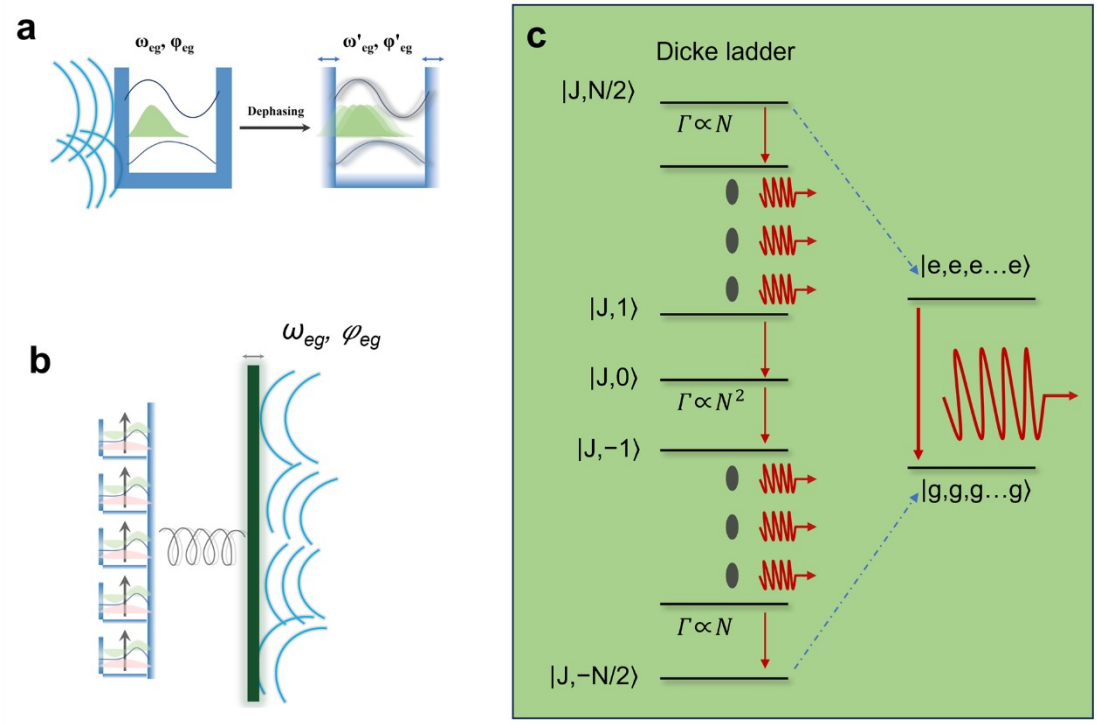

Figure S13 a) Multiple boxes attached to the strongest-coupled lattice distortion mode that promotes coherence between the dipoles. b) This leads to a macroscopic state, forming a 'giant dipole' that radiates a superfluorescent burst. c) The coherent population forms a giant atom, with its relaxation path following the states in a Dicke ladder

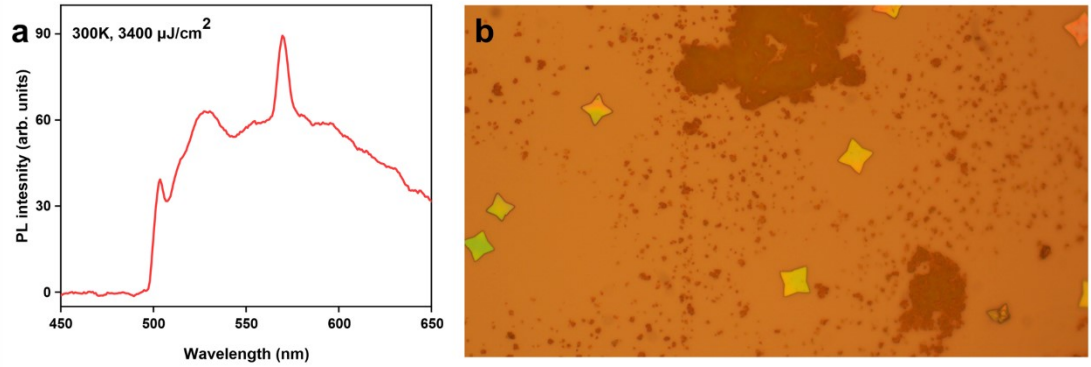

Figure S14 a) Fluorescence spectra and b) morphology of over-aged mesocrystalline QD superlatticesline QD superlattices.

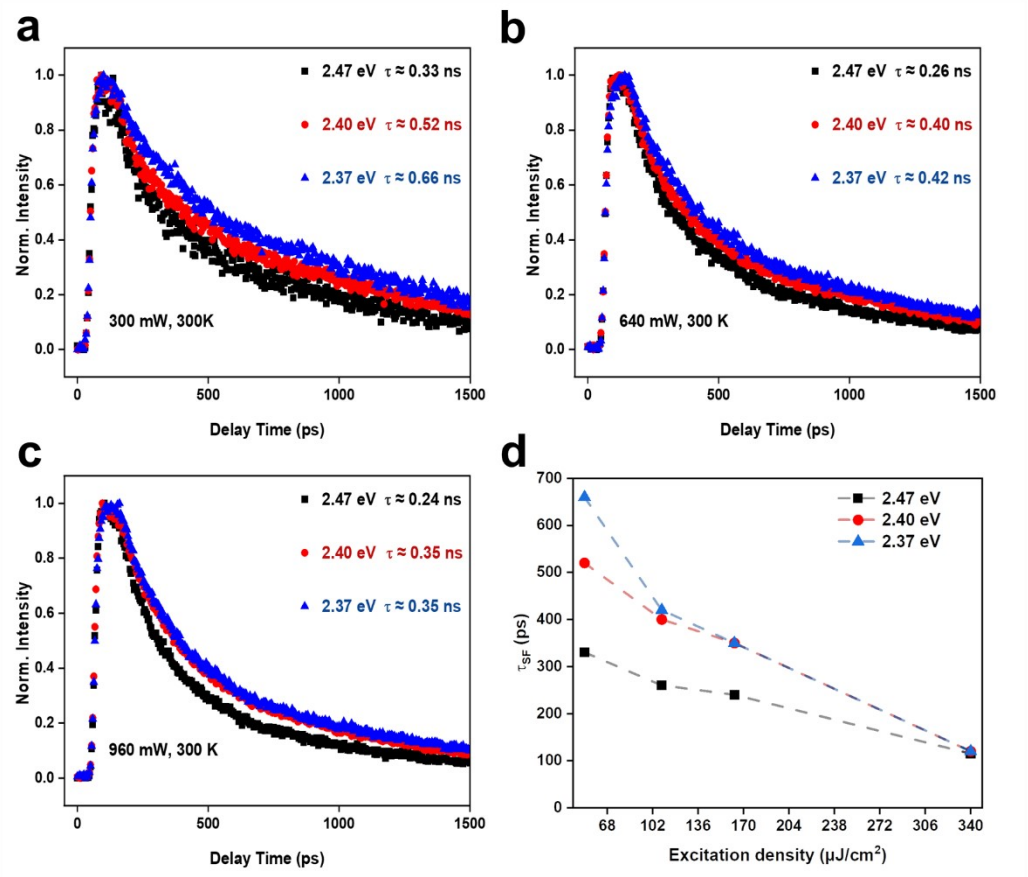

Figure S15 Coherent Emission decay dynamics as the change of the excitation power density in the same mesocrystal.