

## Coarse and fine-tuning of lasing transverse electromagnetic modes in coupled all-inorganic perovskite quantum dots

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Supporting information to <https://doi.org/10.1007/s12274-020-3051-y>

### Chemicals

High-quality CsPbBr<sub>3</sub> QDs were synthesized at room temperature using a top-down fabrication approach which is easy to reproduce and highly cost-effective. First, we synthesized the bulk CsPbBr<sub>3</sub> by dissolving PbBr<sub>2</sub> in hydrobromic acid and then subsequent addition of Cs<sub>2</sub>CO<sub>3</sub> gives the yellowish orange precipitate. The precipitate was washed with copious amount of acetone to get the bulk CsPbBr<sub>3</sub>. The bulk product and octylammonium bromide were dissolved in DMF and then toluene was added to it resulting in the yellow colloidal solution. The colloidal product was separated by centrifugation and dried at 60 °C to get the CsPbBr<sub>3</sub> QDs. PbBr<sub>2</sub> (98%), HBr (48% in water) and Cs<sub>2</sub>CO<sub>3</sub> (99%) were purchased from Sigma-Aldrich. N,N-Dimethylformamide (DMF, 99%), toluene (99%), and acetone (99%) were purchased from Samchun.

### Synthesis of bulk CsPbBr<sub>3</sub> and quantum dots

PbBr<sub>2</sub> (1 gm, 2.72 mmol) was dissolved in HBr (5 ml) and then Cs<sub>2</sub>CO<sub>3</sub> (650 mg, 2 mmol) was added to it under stirring conditions. A vigorous reaction follows from which CO<sub>2</sub> gas was emitted and a yellowish-orange product appeared immediately. The product was washed with acetone and dried in an oven at 60 °C. CsPbBr<sub>3</sub> QDs were synthesized by a top-down approach at room temperature. First, bulk CsPbBr<sub>3</sub> (164 mg, 0.282 mmol) and octylammonium bromide (70 mg, 0.127 mmol) were dissolved in DMF (10 ml) whilst being stirred and then filtered to remove any undissolved particles. Then toluene (20 ml) was added to the solution to precipitate out the yellow colloidal nanocrystals. DMF has a higher dielectric constant than toluene. The change in solubility caused precipitation of CsPbBr<sub>3</sub> QDs. Octylammonium acts as capping ligand to arrest the precipitation of the CsPbBr<sub>3</sub> QDs. The nanoparticles were separated by a centrifuge operating at 5000 rpm for 5 minutes. The CsPbBr<sub>3</sub> QDs are soluble in toluene and exhibit bright green fluorescence under UV light.

### Optical system

The microphotoluminescence system uses a Mitutoyo 0.7 N.A. 100× long working distance microscope objective with an effective focal length of 2 mm to collect the emission. This is matched in a confocal configuration by a 50 mm focal length lens focusing into a 25 μm multi-mode fiber to maintain a spatial resolution at 534 nm on the sample surface of ~0.71 ± 0.02 μm (as measured directly from fitting the profile in figure S6(a) below). The fiber forms the effective entrance slit of our 0.3 m spectrograph, giving a spectral resolution of 0.15 nm at the focal plane imaged by an Andor EMCCD with 16×16 μm pixels using a 1200 l/mm 500 nm blazed grating.

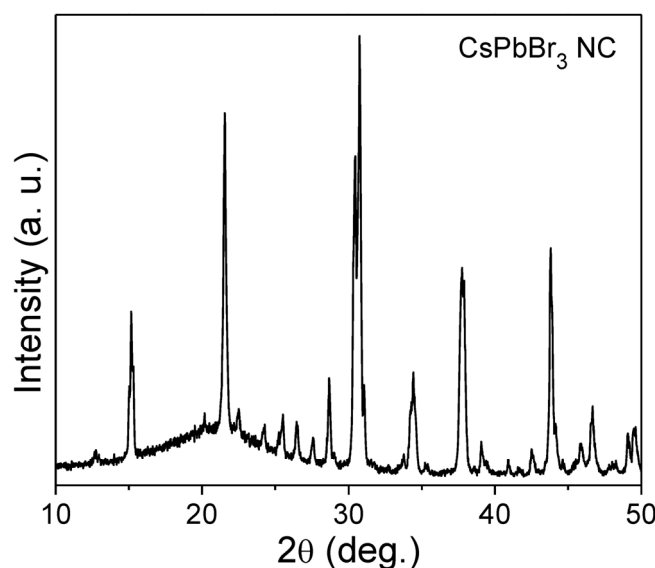


**Figure S1** The diffraction peaks correspond to the orthorhombic phase structure similar to bulk  $\text{CsPbBr}_3$ . Distinguishing this phase structure from the cubic phase is difficult because of the similarity in their diffraction peaks [1-4].

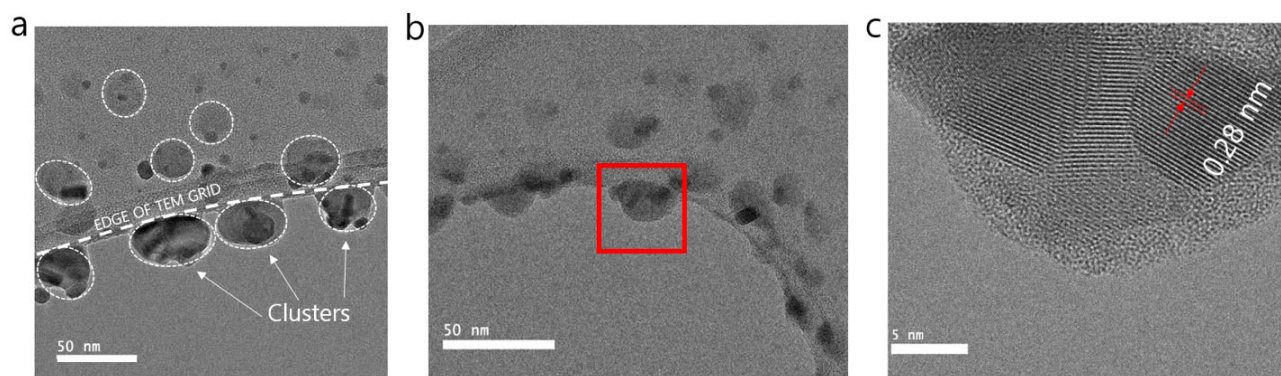


**Figure S2** Transmission electron microscopy image of  $\text{CsPbBr}_3$  nanocrystals. (a), (b) Formation of nanocrystals from quantum dots. The morphology of the QDs is consistent with that reported elsewhere, such as in Di et al [5], and should not be confused with lead precipitates that can result from Transmission electron microscopy beam damage to the sample. c Zoomed-in image of the nanocrystal from the red squared region in (b) to show the quantum dot lattice structure. The lattice fringes are clearly observed and the interplanar spacing is found to be 0.28 nm, which is comparable to that of  $\text{CsPbBr}_3$  [5,6]. There is strong coupling between neighboring small QDs in the nanocrystal.

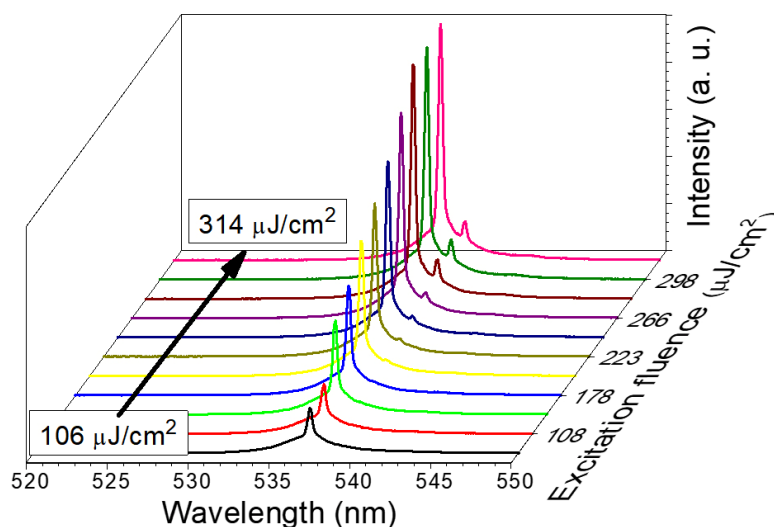


**Figure S3** Power-dependent PL spectra for the same QD cluster demonstrated in figure 1(d and e) at high excitation fluence regimes from  $\sim 106$  to  $\sim 314$   $\mu\text{J}/\text{cm}^2$ . For the intensity comparison, the spectrum measured at  $\sim 106$   $\mu\text{J}/\text{cm}^2$  was also shown in figure 1d.

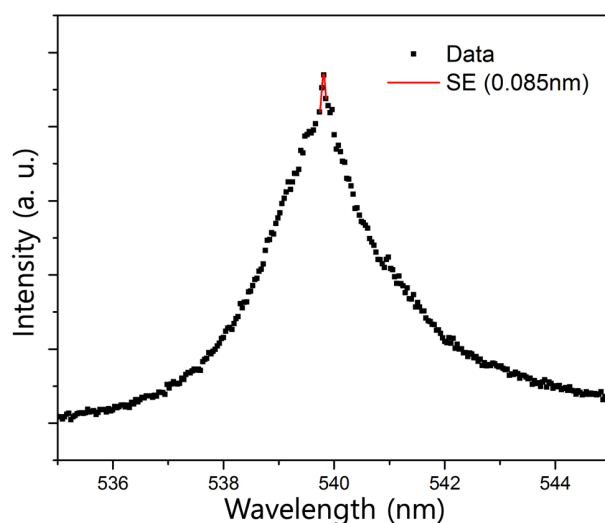


**Figure S4** PL taken at sub-threshold excitation of less than  $\sim 45 \mu\text{J}/\text{cm}^2$ , the FWHM of the mode is 0.085 nm.

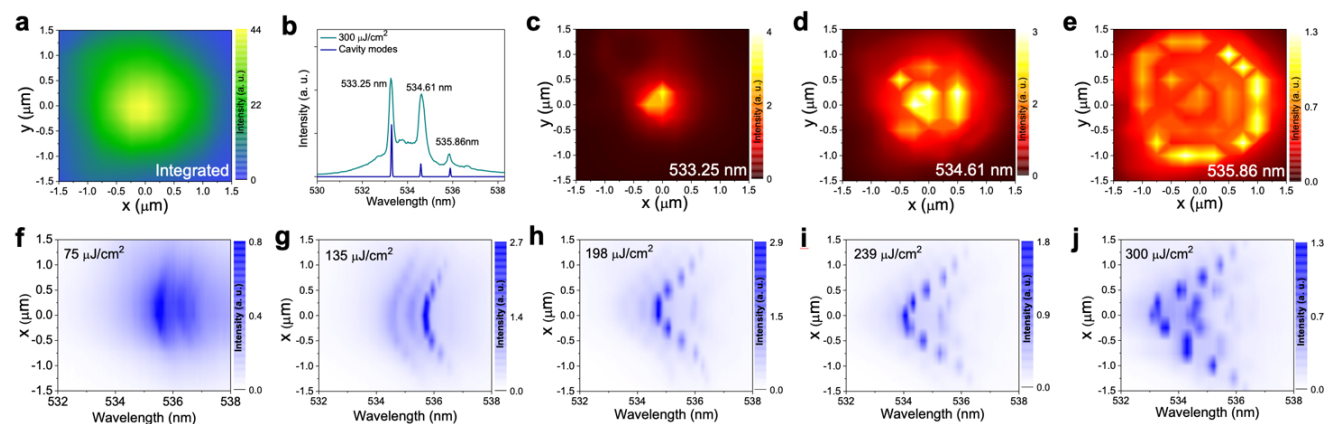


**Figure S5** (a) Integrated luminescence map over the region of the QD cluster for an excitation power  $\sim 300 \mu\text{J}/\text{cm}^2$ . (b) A single spectrum recorded near the center in a. (light green). Frequencies of the longitudinal electromagnetic ( $\text{TEM}_{00}$ ) and transverse electromagnetic ( $\text{TEM}_{i+j \geq 1}$ ) modes follow from Eq. 2 (dark blue). (c)-(e) cross-sections of the lasing beam waist revealing the TEM mode structure with intensity variations at three distinct mode wavelengths. (f)-(j) Power-dependent spectra maps taken along a slice through  $y = 0$ .

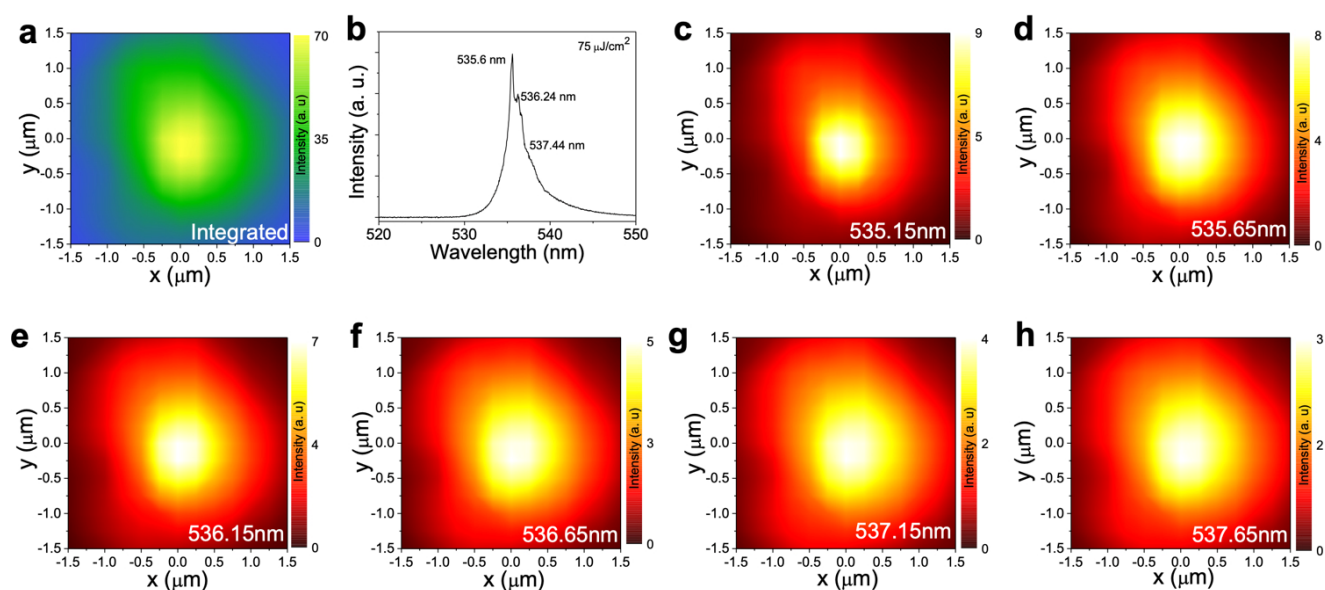


**Figure S6** (a) Integrated luminescence map. (b) A single spectrum taken at a fixed position of  $x=0$  and  $y=0$ . (c)-(h) For the SPE, no transverse optical modes appear.

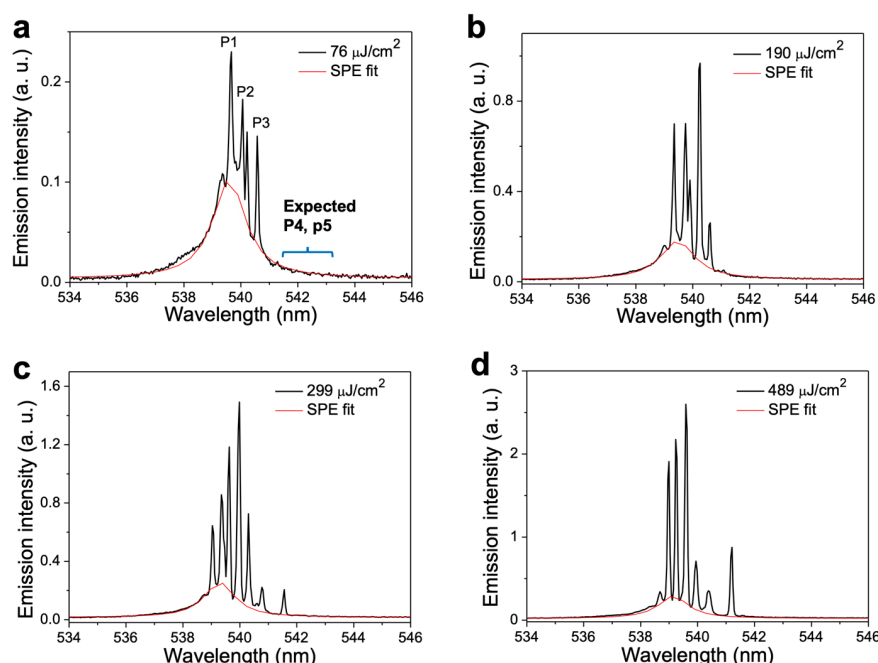


**Figure S7** Emission spectra at increasing excitation fluence with fitting of the SPE emission shown in red. The black spectra are the measured spectra and the red are fitted by Gaussian function. The excitation fluence is (a)  $76 \mu\text{J}/\text{cm}^2$ , (b)  $190 \mu\text{J}/\text{cm}^2$ , (c)  $299 \mu\text{J}/\text{cm}^2$ , and (d)  $489 \mu\text{J}/\text{cm}^2$ .

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