Supporting Informations

Temperature Dependence of the Amplified Spontaneous Emission from $CsPbBr_3$ Nanocrystal Thin Films

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Materials and Synthesis

Materials and chemicals. Cesium carbonate (Cs2CO3, Aldrich, 99.9%), oleic acid (OA, SigmaAldrich, 90%), 1-octadecene (ODE, Sigma-Aldrich, 90%), oleylamine (OAm, Acros Organics, 80-90%), lead bromide (PbBr2, ABCR, 98%), lead iodide (PbI2, ABCR, 99.999%), toluene (Fischer Scientific, HPLC grade), hexane (Sigma-Aldrich, ≥95%). All the chemicals were used as received without any additional purification steps, excepting the ODE and OA which were degassed for 1h at 120 °C under vacuum and stored in an Ar-filled glovebox.

Preparation of Cs-oleate: Cs2CO3 (0.407 g, Aldrich, 99.9%), OA (1.25 mL, Sigma-Aldrich, 90%) and ODE (20 mL, Sigma-Aldrich, 90%) were added into a 50 mL 3-neck flask, degassed for 1h at 120 °C under vacuum. Since Cs-oleate precipitates out of ODE at room-temperature, it must be preheated to 100 °C before injection.

Synthesis of CsPbBr3 NCs: PbBr2 (0.376 mmol, 138 mg), pre-dried OA (1 mL), OAm (1 mL) and pre-dried ODE (5 mL) were loaded into a 25 mL 3-neck flask and kept for 10 minutes at 120 °C under vacuum with vigorous stirring. The reaction mixture was switched on N2 atmosphere, heated up to 180 °C and 3.2 mL Cs-oleate was swiftly injected. 10 seconds later the reaction was stopped by immersing the flask into a water-ice bath. The CsPbB3 NCs were further purified by a fast centrifugation (5 minutes at 20133 rcf) and redispersion of the obtained precipitate into hexane (0.5 mL). The NCs were centrifuged again at high speed and the supernatant was disposed. The precipitated NCs were redispersed in toluene (4 mL) and after one last centrifugation (about 2 minutes at 2200 rcf), the supernatant was collected and further used for our investigations.

Nanocrystal characterization

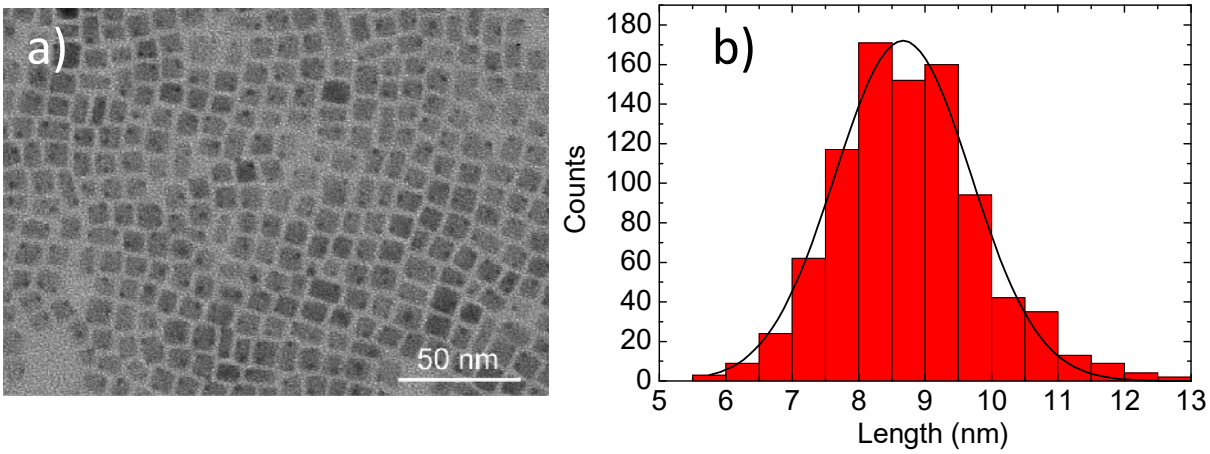

Fig S1: a) TEM image of the $CsPbBr_3$ nanocrystals. b) distribution of the nanocrystal side length (the line is the best fit curve with a single gaussian peak).

The photoluminescence Quantum Yield (PLQY) of the NCs in toluene solution is 64.7 %, evidencing a high quality of the nanocrystals.

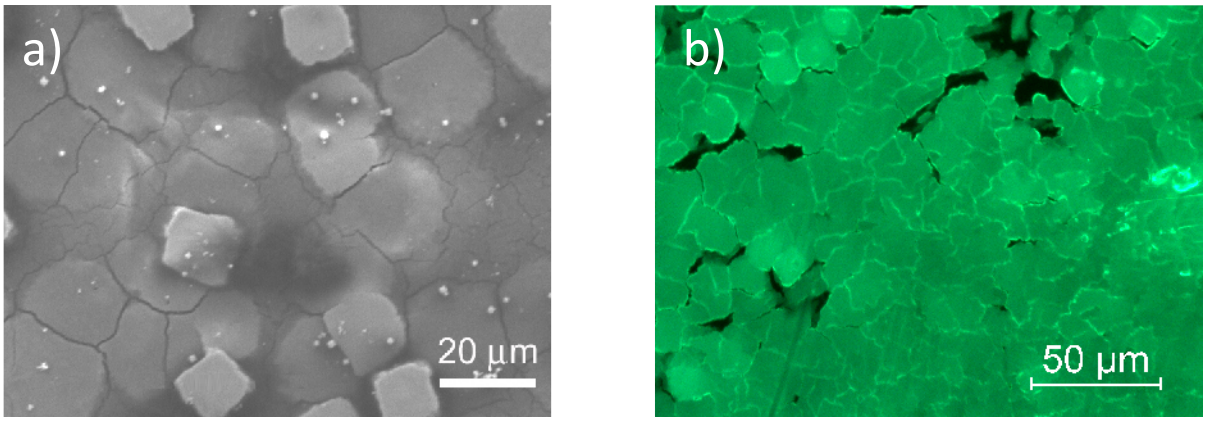

Fig S2: a) SEM image of the film. b) Fluorescence map of the film in the 540±20 nm spectral range.

The TEM image of the nanocrystals evidences a typical slightly rectangular shape (see Fig. S1 a). The distribution of the lateral size (Fig. S1 b) can be reproduced by a single Gaussian peaked at 8.67±0.04 nm and with a standard deviation of 1.02±0.04 nm, evidencing the presence of a single family of nanocrystal with a narrow size distribution.

The film morphology evidences an almost uniform substrate coverage (see Fig. S2 a) with interconnected domains with a typical lateral size of about 20 µm, and almost square aggregates with a size of about 15 µm.

The local emission properties of the film have been investigated by fluorescence microscopy, evidencing a uniform emission intensity (see Fig. S2 b), with brighter emission at the grain boundaries likely due to waveguiding along the film and partial light scattering at the grain boundaries.

Multigaussian fitting of the PL spectra below ASE threshold

In order to understand the complex PL temperature dependence discussed in the main text we performed a multigaussian fitting of the PL spectra at low excitation density.

The number of employed gaussians has been fixed as the minimum number necessary to reproduce the spectra leading to random fluctuation of the differences between the best fit curve and the experimental data.

The PL spectra between 10 K and 170 K can be well reproduced by the linear combination of two gaussian peaks, while a third peak is necessary to reproduce the experimental lineshape above 190 K (see Fig. S3).

The following figures report the temperature dependence of the best fit gaussians intensity (see Fig. S4), the relative contribution to the total intensity of each gaussian (see Fig. S5) and the best fit peak wavelength, compared with the PL and the ASE peak (see Fig S6).

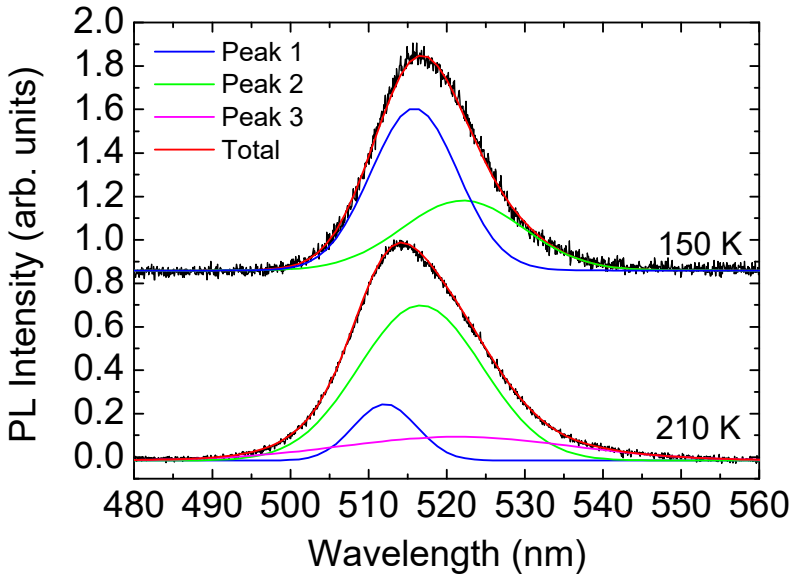

Fig S3: example of multigaussian fit of the PL spectra with 2 peaks at 150 K and with 3 peaks at 210 K.

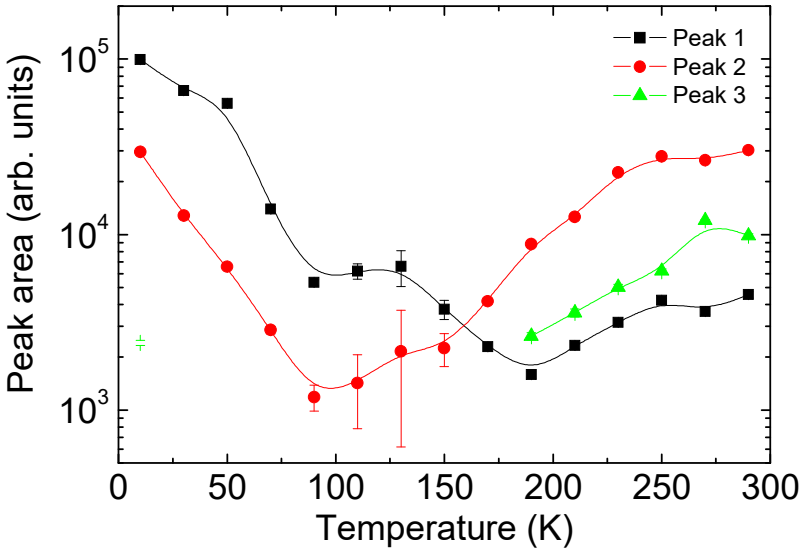

Fig S4: temperature dependence of the intensity of the three gaussian peaks used for the PL spectra fitting.

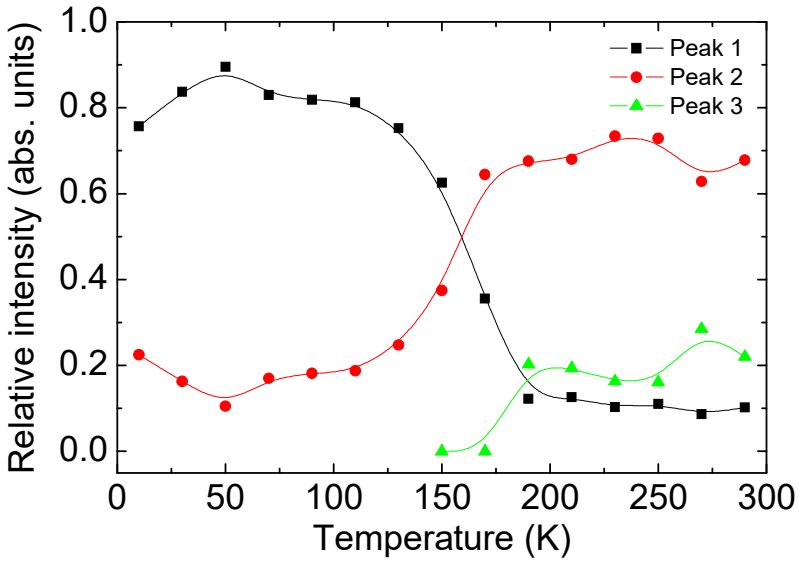

Fig S5: temperature dependence of the relative intensity of the three gaussian peaks used for the PL spectra fitting.

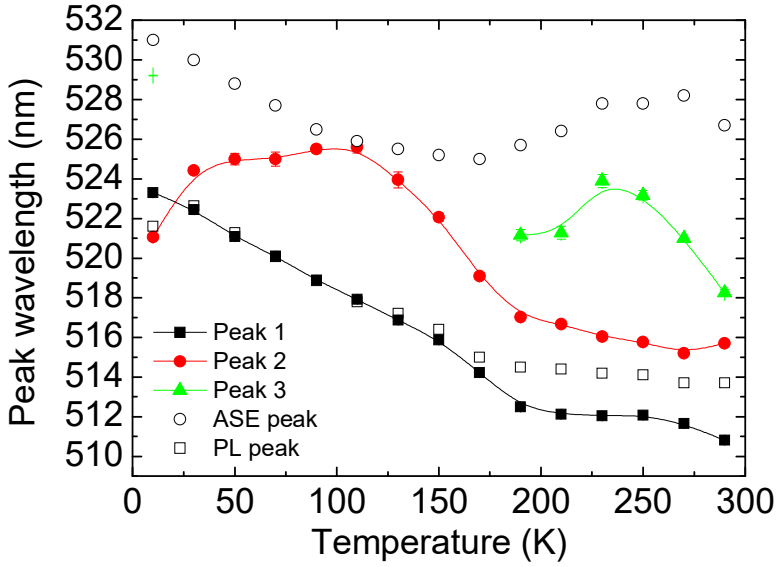

Fig S6: temperature dependence of the peak wavelength of the three gaussian peaks used for the PL spectra fitting.