

Supporting Information

Temperature dependent amplified spontaneous emission in CsPbBr₃ thin films deposited by single-step RF-magnetron sputtering

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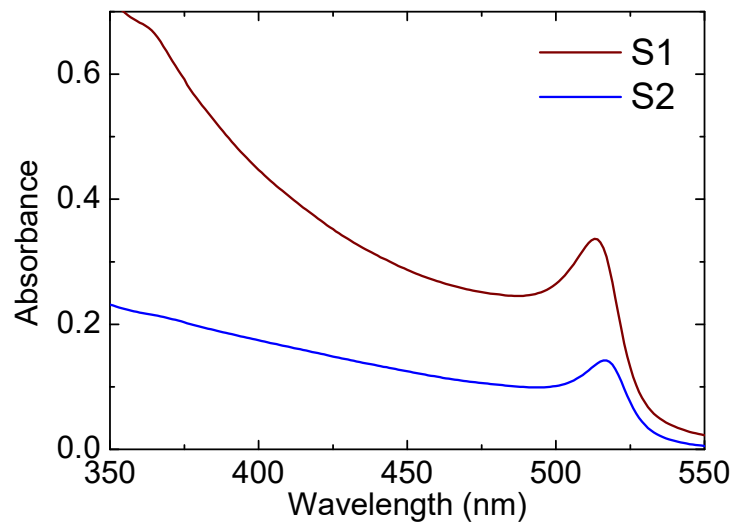


Figure S1: Absorption spectra of the two samples studied recorded at room temperature.

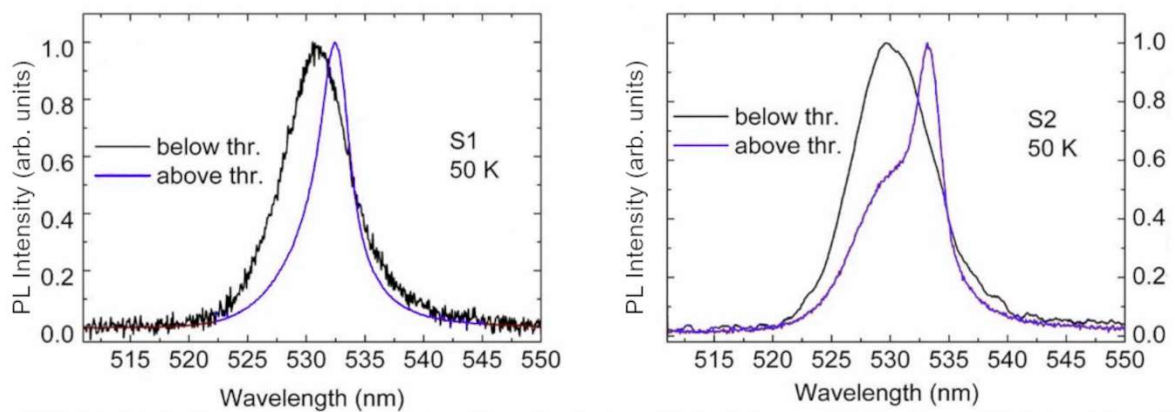


Figure S2: Comparison of the normalized emission spectra obtained at an excitation below and above threshold for both the samples studied, recorded at 50 K.

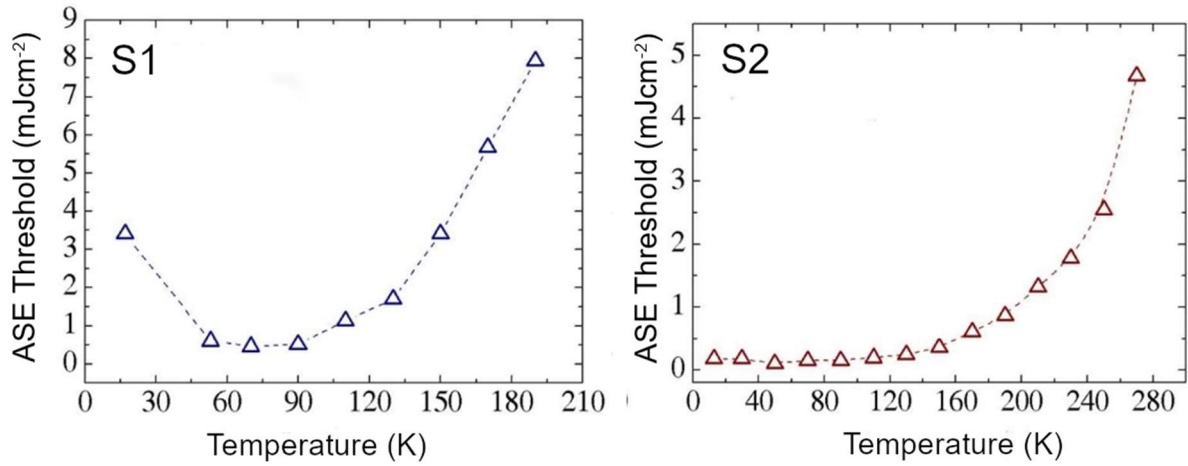


Figure S3: Complete dataset of the extracted thresholds in the samples studied, as a function of the temperature.

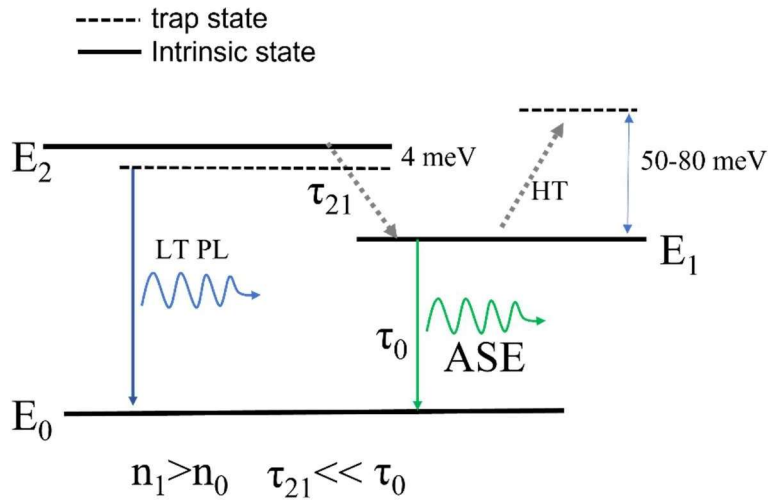


Figure S4: 3-level scheme, showing the main processes occurring during ASE action at different temperature ranges.

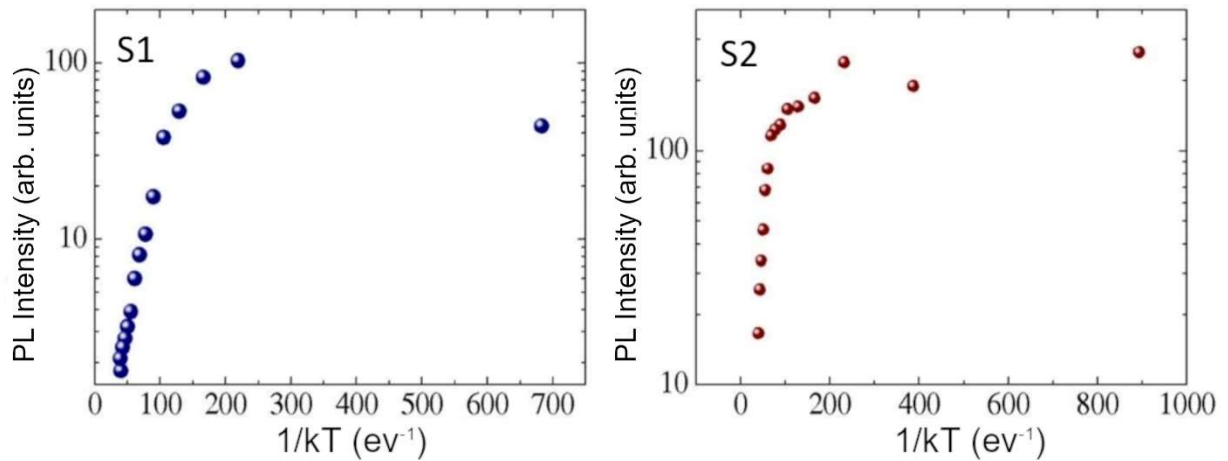


Figure S5: complete dataset of the PL intensities vs. temperature.

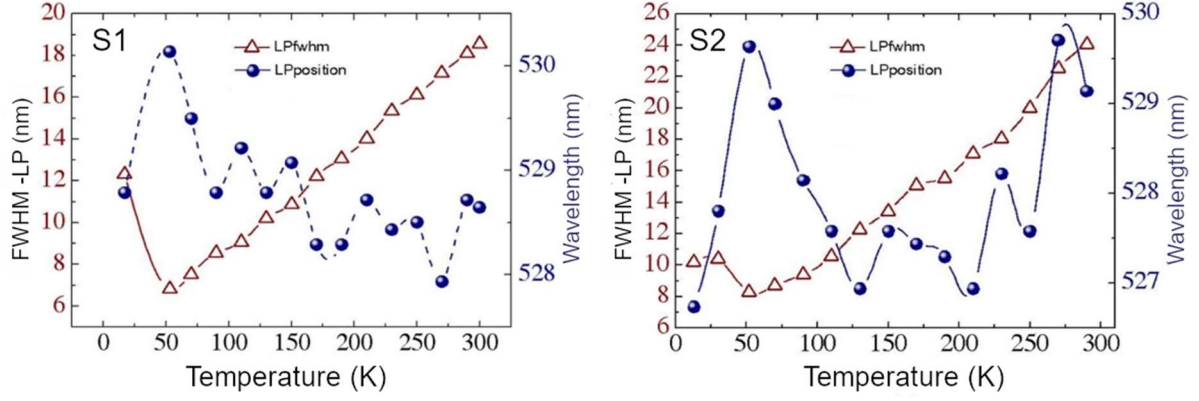


Figure S6: PL position and FWHM as a function of the temperature in both the samples S1 and S2, when excited at one half of the threshold density.

Model of temperature dependence of ASE Thresholds in a 3-level system

If N is the whole population (volume density, cm^{-3}) and n_0 and n_1 are the populations of the lower and upper states, respectively (with $N = n_0 + n_1$), the condition for population inversion is reached when a half of the carriers ($N/2$) occupies the upper state, i.e. $n_1 = N/2$.

The amplification process starts when

$$\sigma(2n_1 - N) > \alpha \Leftrightarrow 2n_1 - N = \frac{\alpha}{\sigma} \quad (1)$$

where α represents the losses coefficient and σ is the gain cross-section. Let n_{1th} be the population level at the threshold condition; the consequent depopulation rate at the threshold pumping can be expressed as

$$\frac{n_{1th}}{\tau} = \frac{1}{2} \left(\frac{\alpha}{\sigma} + N \right) \frac{1}{\tau} \quad (2)$$

Here, $\frac{1}{\tau} = \frac{1}{\tau_0} + \frac{1}{\tau_{nr}}$ is the total decay rate at the threshold condition, τ_0 is the intrinsic transition lifetime and τ_{nr} is the lifetime of a non-radiative process, a parameter accounting for eventual thermally activated processes, such as thermally induced carrier trapping/detrapping, exciton thermal dissociation, exciton-exciton scattering, carrier thermal escape from the material, all of them detrimental for efficient ASE.^[32] In fact, for a defined thermally induced non-radiative process (characterized by the lifetime τ_T and an activation energy E_a), the rate $1/\tau_{nr}$ is expressed as

$$\frac{1}{\tau_{nr}} = \frac{1}{\tau_T} e^{-\frac{E_a}{kT}} \quad (3)$$

Where k is the Boltzmann constant.

In steady state regime (our pump pulse is much longer than the ASE lifetime) the excited state depopulation rate $\frac{n_{1th}}{\tau}$ is equal to the excitation rate g_{0th} .

As the pump rate is directly proportional to the excitation density we finally have $D_{th} = Cg_{0th}$ and,

substituting Equation 3 in Equation 2 and compacting the temperature independent terms in $D_0 =$

$$C \frac{1}{2} \left(\frac{\alpha}{\sigma} + N \right) \frac{1}{\tau_0}, D_1 = C \frac{1}{2} \left(\frac{\alpha}{\sigma} + N \right) \frac{1}{\tau_T}, \text{ and } D_{th} = C \frac{n_{2th}}{\tau}:$$

$$D_{th} = D_0 + D_1 e^{-\frac{E_a}{KT}}$$