

Efficient Amplified Spontaneous Emission from Solution-Processed CsPbBr₃ Nanocrystal Microcavities under Continuous Wave Excitation

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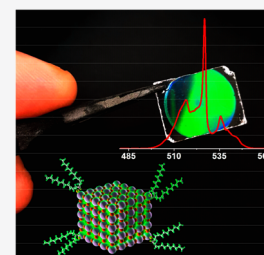
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Supporting Information

ABSTRACT: Solution-processed lasers are cost-effective, compatible with a vast range of photonic resonators, and suited for a mass production of flexible, lightweight, and disposable devices. The emerging class of lead halide perovskite nanocrystals (LHP NCs) can serve as a highly suitable active medium for such lasers, owing to their outstanding optical gain properties and the suppressed optical nonradiative recombination losses stemming from their defect-tolerant nature. In this work, CsPbBr₃ NCs are embedded within polymeric Bragg reflectors to produce fully solution-processed microcavities. By a systematic parametric optimization of the polymer mirrors, resonators with *Q*-factors up to 110 can be produced in the green, supporting amplified spontaneous emission (ASE) under continuous wave excitation, with a threshold as low as 140 mW/cm². Angle-dependent reflectivity and luminescence studies performed below the ASE threshold demonstrate the strong spectral and angular redistribution of the CsPbBr₃ NC spontaneous emission when coupled to the cavity mode. Under resonance, amplification of the output intensity by a factor of 9 in the vicinity of the cavity mode and by a factor of 5 in the whole integrated emission along with an increase of the radiative rate accounted by a Purcell factor of 2 is obtained with respect to NCs deposited in reference microcavity structures.

KEYWORDS: lead halide perovskites, nanocrystals, polymer photonic crystals, microcavities, amplified spontaneous emission



Solution-processed, optically pumped lasers combine facile and low-cost fabrication with the prospect of flexible, lightweight, and disposable devices suitable for a range of applications that stretch from optical modulation to on-chip characterization, to biological sensing and security tagging.^{1–10} Fabry–Perot-type cavities appear as an obvious simple and versatile resonator choice for such lasers owing to the simple design and the demonstrated ability to effectively control light propagation and amplification.^{11–13} Optical feedback in such structures is provided by two facing, solution-processed, distributed Bragg reflectors (DBRs) produced by alternating casted layers of materials with refractive index contrast, processed using orthogonal solvents. Based on such a design, relatively high *Q*-factor cavities supporting stimulated emission with low threshold and high beam directionality have been reported. The majority of the reported work has been based on conjugated polymers that exhibit high and broad optical gain and large absorption cross-section, but suffer from losses due to polaron¹⁴ and triplet exciton absorption.¹⁵ Such losses are less significant in lead halide perovskite nanocrystals (LHP NCs), which are also less prone to stimulated emission competing mechanisms affecting other colloidal nanocrystal media such as carrier trapping and surface recombination. The ability of perovskite poly- and nanocrystalline materials including LHP NCs to sustain lasing oscillation even under continuous wave excitation has been recently demonstrated, via the implementation of various types of optical resona-

tors.^{16–22} Such attributes make LHP NCs promising contenders for practical, optically pumped lasers,^{23–30} yet there is limited reported work combining LHP NCs active media with solution-processed resonators.^{31–34}

Herein, we demonstrate the monolithic fabrication of all-solution-processed microcavities based on green-emitting, weakly confined CsPbBr₃ NCs and polymer-based DBRs, produced from alternating spin-casted layers of cellulose acetate (CA) and polyvinylcarbazole (PVK) polymers. Parametric studies to optimize the performance of such DBRs are initially performed, followed by the fabrication of fully processed microcavities and detailed characterization based on steady-state and time-resolved emission combined with angle-dependent reflectivity and photoluminescence (PL). The photonic structures exhibit *Q*-factors up to ~110, in line with state-of-the-art polymer-based resonators, allowing the generation of bright, green efficient amplified spontaneous emission (ASE) under continuous wave excitation at room temperature. The onset of ASE upon excitation with a

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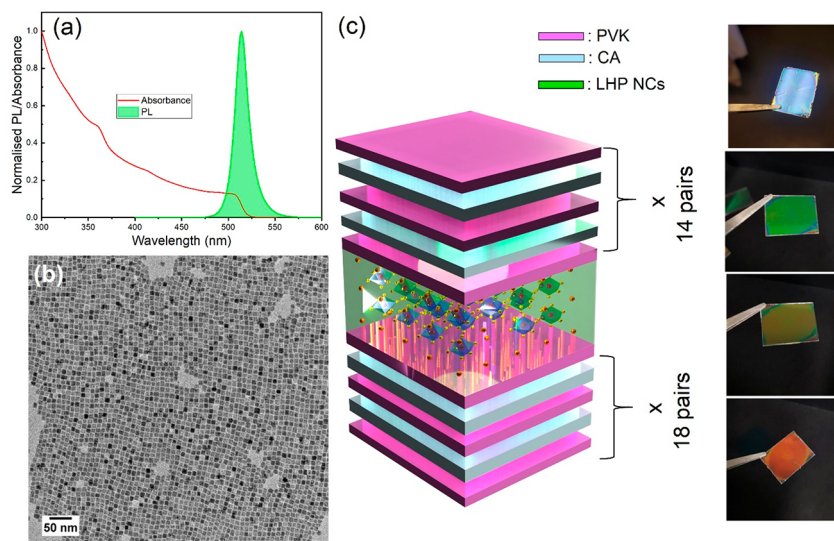


Figure 1. (a) Normalized absorbance and PL spectra from CsPbBr₃ NCs capped with DDAB/PbBr₂ ligands. (b) TEM image of the CsPbBr₃ NCs. (c) Schematic illustration of the all-solution-processed vertical microcavity. The inset photos present polymer DBR mirrors, operating at different parts of the visible spectrum region.

continuous wave laser is evidenced by a clear threshold-type behavior and spectral narrowing on the excitation-variable PL. Below the ASE threshold, coupling of the NC spontaneous emission with the cavity mode results in directional emission and amplification within the mode resonance by an order of magnitude along with a factor of 2 increase in the radiative NC rate.

CsPbBr₃ NCs capped with a dimethyldioctadecylammonium bromide (DDAB) ligand have been selected as the active gain medium based on the structural robustness, the good photostability, and their high PL quantum yield,³⁵ reaching up to ~90% in the form of spin-casted films.^{36–39} The NCs exhibit bright emission, centered at ~518 nm, and full width at half-maximum (fwhm) of ~20 nm, as shown in Figure 1(a). The corresponding TEM image in Figure 1(b) reveals a uniform and cuboidal shape of the NCs with mean sizes of ca. 10 nm. The DBR mirrors were fabricated via an alternate deposition of cellulose acetate (Mn = 30 000, $n_{ca} \approx 1.46$) dissolved in acetone (14 mg/mL) and polyvinylcarbazole (Across Organic, MW = 90 000, $n_{PVK} \approx 1.68$) dissolved in toluene (35 mg/mL). A detailed parametric study for the optimization of the mirror characteristics was carried out by exploring the deposition speed, number of pairs, polymer concentration, and the impact of thermal annealing; the study results are presented in Figures S1–S4 of the SI. Optimized mirrors consisted of 18 quarter-wave pair polymer layers produced via dynamic dispense spin-coating at 6000 rpm with peak reflectivity at ~525 nm. This condition gives the largest possible first-order photonic band gap (PBG) based on the refractive index contrast of the polymer materials used,³² with the second order appearing in the deep-UV region of the spectrum, which is not detectable due to absorption by the PVK polymer. Each DBR pair was subjected to thermal treatment at 70 °C for 1 min, promoting the efficient evaporation of trapped solvent. The used polymers were chosen to give the best possible refractive index contrast, while satisfying the orthogonal solubility condition. The final structure of the microcavity is schematically illustrated in Figure 1(c), while the inset images demonstrate the ability of

polymer-based DBRs to form highly uniform mirrors covering the whole visible spectrum.

Following fabrication of the bottom DBR mirror, a mixture of 25 mg/mL CsPbBr₃ NCs and 30 mg/mL polystyrene (PS) in toluene, in 1:1 volume ratio, was spin casted at 1000 rpm for 30 s, followed by a drying step of 4000 rpm for another 30 s. The microcavity was completed with the deposition of a top polymer DBR mirror with similar characteristics to those of the bottom mirror. The active layer thickness was measured at ~140 nm, via single-point ellipsometry. The polystyrene matrix provides both improved ambient and optical stability^{40,41} by encapsulating the NCs, while allowing the formation of sufficiently thick active layers, to fulfill the $\lambda/2$ condition for suitable photon field confinement. The surface morphology of the polystyrene-NC active region as well as that of a polystyrene-NC layer overcoated by a pair of CA and PVK polymer layers forming the DBRs of the cavity was studied using AFM; representative images are displayed in Figure S5. The microscopy indicates a relatively smooth surface topography of the two samples that becomes slightly rougher as the mirror pair is deposited, with an RMS roughness of ~2.4 and ~3.9 nm prior to and after the deposition of the DBR pair, respectively.

The optical performance of the microcavities was evaluated by comparative studies with two reference structures. The first reference was an “open cavity”, with the active NC-polystyrene medium being deposited on a transparent quartz substrate and capped by a single, top DBR mirror. The sample was used to evaluate the optical performance of NCs in the absence of a complete optical cavity stack. The second “off-resonance” reference was produced out of an identical active layer deposited in between polymer DBRs, which were intentionally mismatched to the green NC emission, by exhibiting a PBG in the orange range. The off-resonance cavity aimed to mimic the dielectric environment of the active gain medium, allowing a quantitative analysis of the changes in spontaneous emission induced in the on-resonance cavity.

Photoexcitation of the microcavities was performed through the backside of the quartz substrate, using a continuous wave (CW) 405 nm diode laser, with the emission coaxially

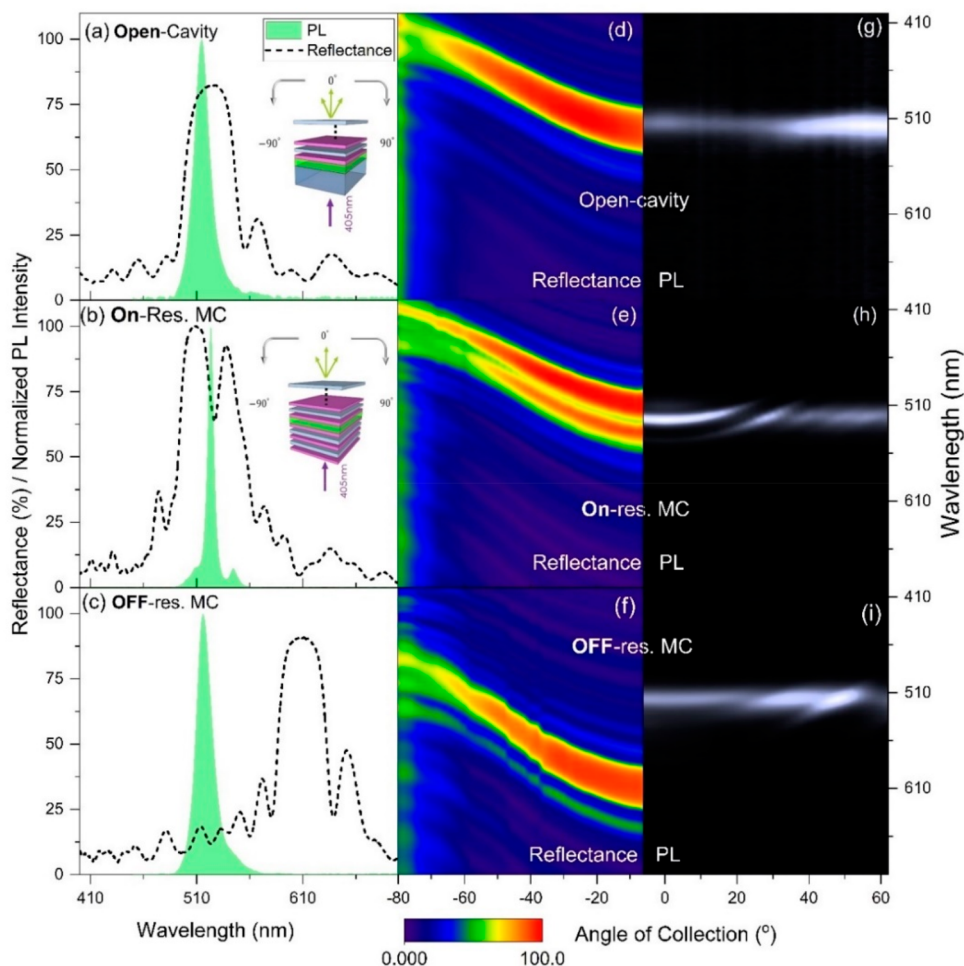


Figure 2. Normalized PL and reflectance spectra measured at 4 degrees normal to surface for an (a) open, (b) on-resonance, and (c) off-resonance cavity. Inset schematics illustrate the cavities' structure and the experimental geometry used. The angular distribution of the reflectance and the emission from the three cavities is plotted in (d)–(f) and (g)–(i), respectively.

collected in transmission with a long-pass filter, blocking the leakage excitation laser. The broadband reflectance along with the respective PL spectra, collected near the surface normal ($\sim 4^\circ$) from the three types of cavities, is presented in Figure 2(a) to (c). The open cavity shows a maximum reflectance of $\sim 78\%$, centered at ~ 526 nm, assigned to the PBG of the single polymer DBR. The large fwhm of the PBG (~ 50 nm) arises from the relatively large refractive index contrast ($\Delta n \approx 0.22$) between the PVK and CA polymers. In the absence of full optical feedback, no significant spectral modification of the spontaneous emission can be observed, with the PL spectra of NCs being centered at ~ 518 nm with a fwhm of ~ 18 nm, similar to the respective characteristics of the pristine NC film presented in Figure 1(a). In contrast, the data from the complete, on-resonance microcavity contain signatures of the cavity resonance and the emission modification, with spectral overlapping reflectance dip and emission peak in the vicinity of ~ 527 nm, as seen in Figure 2(b). The high, for such solution-processed structures, cavity finesse is further witnessed by the reflectance peak intensity of $\sim 99\%$, the wide PBG of ~ 65 nm, and the estimated quality factor Q of ~ 110 . The Q -factor was obtained by eq 1 using the PL characteristics at 527 nm at low excitation fluence below the threshold of emission amplification, being comparable to the state-of-the-art reported for monolithic polymer microcavities:^{33,42}

$$Q = \frac{\lambda}{\Delta\lambda} \quad (1)$$

The PL and reflectance spectra of the off-resonance microcavity are presented in Figure 2(c), with the PBG centered at ~ 610 nm, exhibiting an fwhm of ~ 60 nm. The mismatch between the CsPbBr₃ NCs emission and the cavity stop band results in negligible modification in the spontaneous emission from the nanocrystal layer, yielding PL characteristics similar to those obtained from the open-cavity reference. The angular dispersion of the PBG from the three cavity samples is presented in Figures 2(d)–(f), over a wide range of azimuthal angles, showing a characteristic blue-shift at wider angles, described by the Bragg–Snell law below.

$$m\lambda_{\text{PBG}} = 2d\sqrt{n_{\text{eff}}^2 - \sin^2(\theta_{\text{ext}})} \quad (2)$$

The steady-state PL spectra of the microcavities collected at different angles are presented in Figure 2(g) to (i), highlighting angular profiles of the resonators under investigation. Typically, the microcavity emission is enhanced along the axis of the resonator, while being suppressed at wider angles due to the existence of the PBG. However, the emission of the open-cavity and off-resonance cavity as presented in Figure 2(g) and (i) show a Lambertian angular profile due to the absence of optical feedback and mismatch with the PBG,

respectively. The modulation of the PL spectra, observed at wider angles for the off-resonance cavity, arises from the interference fringes of the cavity. On the contrary, the NC luminescence couples efficiently to the cavity mode on the on-resonance cavity as evidenced by (i) the strong PL enhancement at the cavity resonance of ~ 527 nm, (ii) the efficient suppression of the emission that does not couple to the cavity resonance, and (iii) the similar blue-shift of the reflectivity and the emission as collection angle increases, resulting in a directional emission profile.

The nanocrystal emission modification is quantified via the PL enhancement factor (G_e^{PL}),^{3,43} an accurate estimation of this factor can be obtained for the open- and on-resonance cavities by computing the intensity ratio of their emission to the respective emission of the off-resonance photonic structure. The factor is displayed for the two cavities for various detection angles in Figure 3(a) to (j). The green and red shaded areas denote the position of the PBG for the respective angles, while values of G_e^{PL} greater (smaller) than 1 indicate emission enhancement (suppression), respectively. In the open cavity, the PL is suppressed within the PBG range compared to the off-resonance cavity, regardless of the emission angle probed. In contrast, the emission of the on-

resonance cavity is enhanced by almost an order of magnitude at the cavity mode, i.e., $G_e^{\text{PL}} \approx 8.9$, when collected near the surface normal. As wider detection angles are probed, the photonic band gap blue-shifts with respect to the nanocrystal emission, gradually decreasing the cavity-induced PL enhancement. For detection angles greater than 45° , no emission enhancement or spectral redistribution is observed as PBG and NC emission no longer overlap, as evident in the spectra of Figure 2. The angular distribution of the emission from the open and on-resonance cavities at the cavity mode (~ 527 nm) is presented in Figure 3(k), demonstrating the highly directional emission of the latter structure, with a beam divergence of $\sim 8^\circ$. The emission enhancement observed at $\sim 45^\circ$ in both cavities is due to the angular blue-shift of the PBG, minimizing the PBG overlap with the NC emission as observed in the angle-resolved PL and reflectance data displayed in Figure S6; as the optical reflectivity of the structure decreases, extraction of the NC luminescence becomes more efficient. For wider angles than $\sim 45^\circ$ the light is totally internally reflected, and the collected emission is diminished from both structures.

The emission enhancement factor (G_e^{Cavity})⁴⁴ normal to the sample surface at the resonance wavelength can also be quantified by the cavity characteristics and the radiative recombination dynamics via

$$G_e^{\text{Cavity}} = \frac{\xi(1 + \sqrt{R_{\text{bottom}}})^2(1 - R_{\text{top}}) \tau_{\text{cav}}}{2(1 - \sqrt{R_{\text{bottom}}R_{\text{top}}})^2 \tau_{\text{ref}}} \quad (3)$$

where R_{bottom} and R_{top} are the reflectivity of the bottom and top DBR and τ_{cav} and τ_{ref} are the PL lifetimes of the NCs inside and outside the cavity; the quantity ξ is known as the antinode enhancement factor, taking values between 0 and 2. When the active region is placed exactly at a node (antinode) of the produced standing wave, then ξ becomes 0. If the active region is smeared out over many periods of the optical wave, as in the case of our cavities, ξ averages to a value of 1.⁴⁴ For the bottom DBR containing 18 polymer layer pairs, R_{bottom} was estimated at $\sim 90\%$, as presented in Figure S7.

On the other hand, the reflectivity of the top DBR of 14 pairs was obtained via reflectance measurements on the mirror side of the open cavity, yielding a value of $\sim 78\%$, shown in Figure 2(a). Finally, the parameters τ_{cav} and τ_{ref} were extracted by multiexponential fittings of the respective PL transients of the on-resonance and off-resonance cavities, respectively. The time-resolved PL decays for different excitation energies are displayed in Figure S8, along with the respective average PL lifetimes for the three types of studied cavities. The PL dynamics of the off-resonance cavity provide the reference lifetime τ_{ref} allowing to mimic the dielectric environment surrounding the active region. The PL lifetimes used for the calculation correspond to the decays obtained at the highest excitation density of 5 mW/cm^2 that lies closer to the excitation regime of the steady-state PL experiments based on which G_e^{PL} was estimated.

By applying the aforementioned parameters listed in Table 1 in eq 3, an enhancement factor G_e^{Cavity} of ~ 9.2 is estimated, which agrees well with the 8.9 value of the factor $G_e^{\text{PL}}(\theta)$. The two PL enhancement factors are estimated at the resonance cavity wavelength.

For practical devices, the overall emission enhancement obtained by evaluating the enhancement across the whole spectral range of the emission is a more relevant figure of merit.

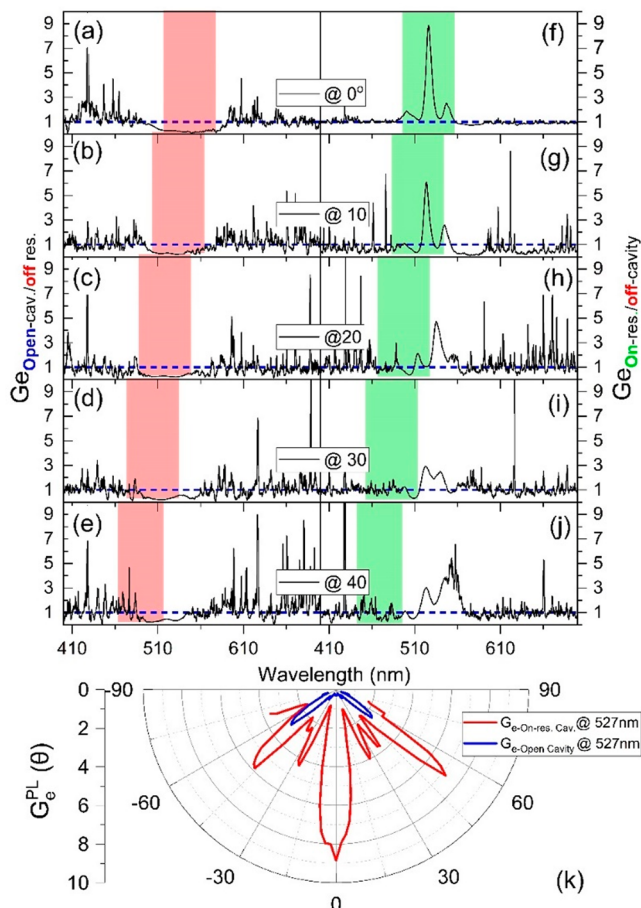


Figure 3. (a–j) PL enhancement factor $G_e^{\text{PL}}(\theta)$ of the open/on-resonance cavities at different collection angles, with respect to the off-resonance cavity. Green and red boxes highlight the position of the PBG, while the blue dashed line is set at value 1, designating the boundary between the suppression and enhancement of the PL. (k) Polar plot of the emission enhancement factor as a function of detection angle at the resonance wavelength, 527 nm.

Table 1. Parameters Used for the Calculation of the Theoretical Spontaneous Emission Enhancement and Comparison of the Respected Experimental and Theory Emission Enhancement Factors^a

R_{bottom}	R_{top}	τ_{cav}	τ_{ref}	ξ	$G_{\text{e}}^{\text{Cavity}}(0)$	$G_{\text{e}}^{\text{PL}}(0)$	$G_{\text{int}}^{\text{Cavity}}$	$G_{\text{int}}^{\text{PL}}$	$G_{\text{tot}}^{\text{exp}}$
~ 0.90	~ 0.78	2.6 ± 0.2	4.5 ± 0.1	1	9.2	8.9	5.3	5.1	3.1

^a R_{bottom} and R_{top} correspond to bottom and top DBR reflectivities, τ_{cav} and τ_{ref} are the PL decay times of the on- and off-resonance microcavities and ξ is the antinode enhancement factor. The emission enhancement factor at the resonance wavelength is denoted as $G_{\text{e}}^{\text{Cavity}}(0)$ and for PL $G_{\text{e}}^{\text{PL}}(0)$ normal to the sample surface. The cavity-integrated PL enhancement factor ($G_{\text{int}}^{\text{Cavity}}$) is estimated via assuming a Gaussian emission model of LHP NCs, while the steady-state PL enhancement factor $G_{\text{int}}^{\text{PL}}$ is obtained via integrating over the entire spectrum. The overall PL enhancing factor integrated over the wavelength range of emission and the detection angle is denoted as $G_{\text{tot}}^{\text{exp}}$.

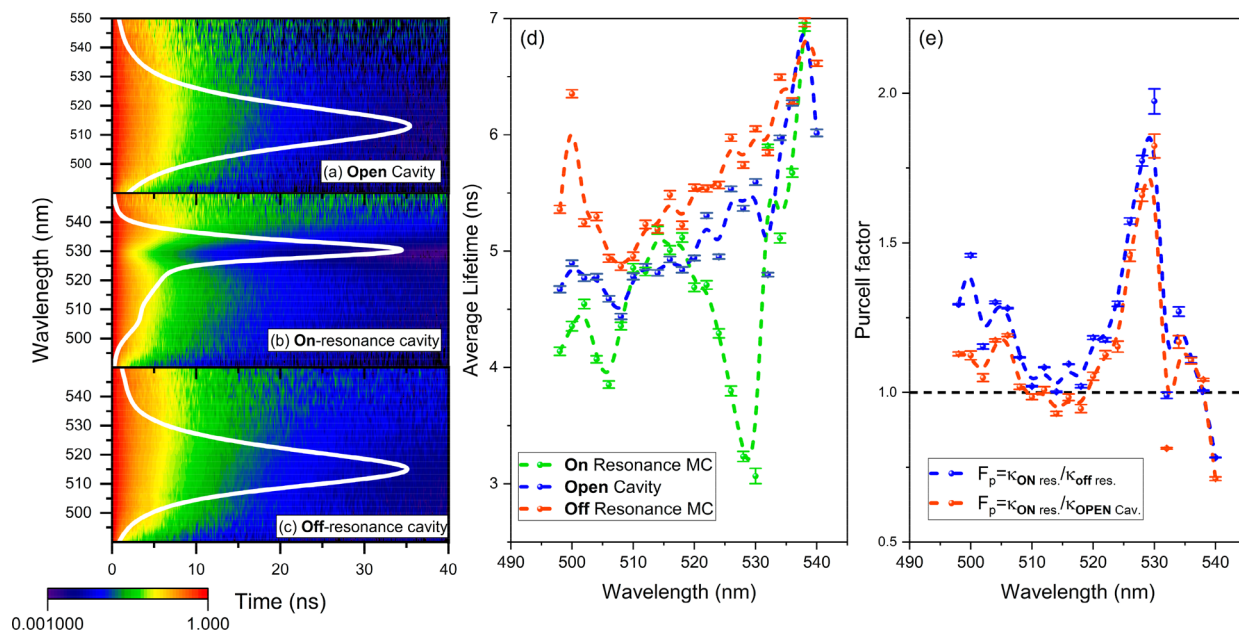


Figure 4. (a–c) Time-resolved emission spectra (TRES) of the open and the on- and off-resonance cavities measured normal to the sample surface. The white line presents the time-integrated PL spectra obtained from the measurements. (d) Average PL lifetimes of the three samples plotted against detection wavelength. (e) Purcell factor versus emission wavelength, using the open and off-resonance cavities as reference, respectively. The black dashed line is set to unity for clarity.

To this extent, the cavity-integrated PL enhancement ($G_{\text{int}}^{\text{Cavity}}$) can be evaluated by assuming a Gaussian emission model of the LHP NCs, given by eq 4:^{43,44}

$$G_{\text{int}}^{\text{Cavity}} = G_{\text{e}}^{\text{Cavity}} \sqrt{\pi \ln(2)} \frac{\Delta\lambda_{\text{on-res}}}{\Delta\lambda_{\text{ref}}} \quad (4)$$

where $\Delta\lambda_{\text{on-res}}$ and $\Delta\lambda_{\text{ref}}$ are the fwhm of the PL from LHP NCs within the cavity (~ 7 nm) and in free space (~ 18 nm), respectively. The calculation yields a factor $G_{\text{int}}^{\text{Cavity}}$ equal to ~ 5.3 . The respective estimation of the steady-state PL enhancement factor is obtained by integrating over the entire spectrum:

$$G_{\text{int}}^{\text{PL}} = \frac{\int \text{PL}_{\text{on-res}}(\lambda, \theta) d\lambda}{\int \text{PL}_{\text{off-res}}(\lambda, \theta) d\lambda} \quad (5)$$

The calculated $G_{\text{int}}^{\text{PL}}$ along the direction normal to the surface of the cavity is ~ 5.1 , well matching the 5.3 value of the $G_{\text{int}}^{\text{Cavity}}$ factor. The dependence of the $G_{\text{int}}^{\text{PL}}$ factor with detection angle is presented in detail in Figure S9 for angles up to 45° . The overall PL-enhancing factor $G_{\text{tot}}^{\text{PL}}$ integrated over the wavelength range of emission and the detection angle is estimated from eq 6 below:

$$G_{\text{tot}}^{\text{PL}} = \frac{\iint \text{PL}_{\text{on-res}}(\lambda, \theta) d\lambda d\theta}{\iint \text{PL}_{\text{off-res}}(\lambda, \theta) d\lambda d\theta} \quad (6)$$

By considering the integrated emission of the on-resonance cavity over the off-resonance cavity up to 45° angle, an overall PL enhancement of $G_{\text{tot}}^{\text{PL}} \approx 3.1$ is calculated. This indicates that the overall intensity of the on-resonance cavity is greatly enhanced over the reference cavity at all angles, further confirming the quality of the produced photonic structures.

The ability of the cavity to enhance the radiative emission rate of the NC emitters by increasing the photonic density of states (PDOS) at the cavity modes is described by the Purcell factor, within eq 7 below.^{45–47}

$$\frac{\kappa_{\text{cav}}}{\kappa_{\text{ref}}} = F_p = \frac{3\lambda^3 Q}{4\pi^2 n_{\text{eff}}^3 V} \quad (7)$$

where κ_{cav} and κ_{ref} are the decay rates ($\sim 1/\tau$) of the CsPbBr₃ NCs within and out of the cavity, respectively, Q is the cavity quality factor, and V is the mode volume. To estimate the Purcell factor, κ_{cav} and κ_{ref} were obtained via time-resolved emission spectra (TRES) measurements with PL decays recorded across the whole emission range with a step of 2 nm and a bandpass of 1 nm. TRES spectra of the incomplete cavity and on- and off-resonance cavity are plotted in Figure 4(a) to (c), superimposed to the respective PL emission

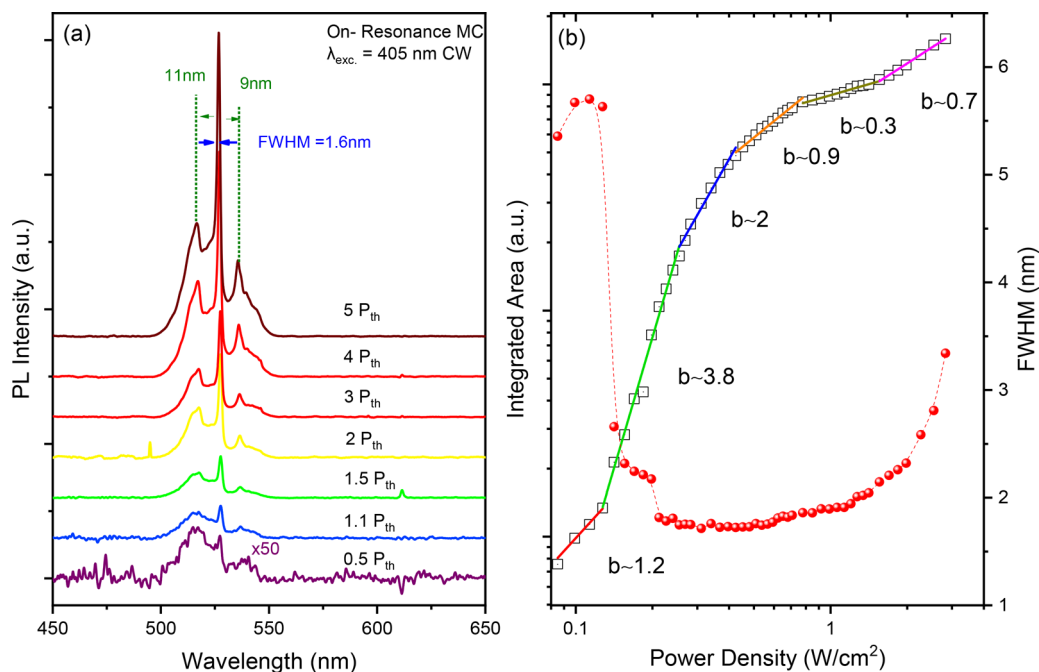


Figure 5. (a) Power-dependent PL spectra of the on-resonance microcavity. The spectra were offset on the y-axis for clarity. (b) Light output intensity integrated over the whole emission spectrum, and fwhm as a function of the optical pumping power density; the output intensity and the pumping power are displayed in logarithmic scales. The integrated emission is fitted by allometric functions with the respective fitted exponents displayed. The ASE onset is clearly defined by the linear to superlinear change of slope and the concomitant reduction of the emission line width.

obtained from time integration of the TRES data; the respective average PL lifetimes from the three samples are plotted against detection wavelength in Figure 4(d). For both the incomplete and the off-resonant cavity, the NC emission rate varies smoothly within the PL line shape as a result of the NC size distribution, resembling the respective PL dynamics of the NCs in the absence of the cavity.⁴⁸ On the contrary for the on-resonance sample, a pronounced reduction of the average PL lifetime from ~ 5 ns to ~ 3 ns is readily visible at the vicinity of the cavity resonance wavelength at ~ 528 nm.

The calculated Purcell factor from eq 7 is plotted against emission wavelength in Figure 4(e). The Purcell factor was found to exhibit a maximum value of $F_p = 1.97 \pm 0.1$ and $F_p = 1.82 \pm 0.1$, considering off-resonance and open cavity as the reference, respectively. The greater than unity values near the cavity resonance confirm the structure-induced enhancement in the spontaneous emission rate from the CsPbBr₃ NC active media. The impact of the cavity modes in the recombination dynamics is further analyzed via angle-dependent TRES measurements presented in Figure S10. The data demonstrate that the PL lifetime minimum recorded at the cavity resonance exhibits the characteristic blue-shift of the photonic structure mode described by the Bragg–Snell law of eq 2. At 45°, the cavity mode does not spectrally overlap with the PL emission of the NCs, diminishing the cavity impact on the recombination dynamics.

The ability of the cavity to sustain stimulated emission is examined via amplified spontaneous emission (ASE) studies performed at elevated power densities using a continuous wave excitation by a 405 nm laser diode. The evolution of the emission with excitation power density is presented in Figure 5(a). As the excitation power increases, ASE is activated via the efficient coupling of the emission to the cavity mode, overtaking the background spontaneous emission from the active NC medium. An ASE threshold of $P_{th} \approx 0.14$ W cm⁻² is

estimated by the distinct change of slope from linear to superlinear of the power dependence and the concurrent collapse of the fwhm from ~ 6 nm to ~ 1.6 nm. The ASE is sustained at higher powers with emission intensity superlinear increasing up to 0.4 W cm⁻² before exhibiting saturating effects due to Auger recombination and local heating^{11,49,50} with a visible increase of the emission line width above 1 W cm⁻². Comparative studies performed on the open and the off-resonance cavity, presented in Figure S11, show no evidence of a threshold-type behavior or emission narrowing that would evidence the activation of ASE. Furthermore, similar experiments were carried out on films of the CsPbBr₃ NCs deposited on quartz substrates. ASE was observed only when excitation via a pulsed femtosecond or nanosecond laser source was employed. The ASE performance of the NC film under 355 nm excitation with 5 ns pulses is summarized in Figure S12, yielding an ASE peak red-shifted by ~ 16 nm with respect to the PL spectra (~ 518 nm) with a threshold of ~ 300 μ J/cm².^{24,25,49,51–55} Based on the reference experiments, it is quite clear that continuous wave ASE can only be sustained for the on-resonance cavity via the efficient coupling of the NC emission to the cavity modes formed by the polymer-based optical resonator, due to the increase of the local photonic density of states.^{56–60} We note that the observed threshold-type behavior on the emission intensity and line width, the light directionality, and the small FWHM of the resonant emission peak, i.e., ~ 1.6 nm, are characteristics consistent with lasing operation, as well. However, a proper justification of lasing requires further proof of spatial, temporal, and spectral coherence.⁶¹ Furthermore, the great majority of lasers exhibit strongly polarized emission. In our study, we find no experimental evidence of either coherent or optical polarization effects. The polarization of the resonant peak in the on-resonance cavity is negligible and similar in magnitude to that of the reference off-resonance and open cavities. In addition,

the output emission does not exhibit a well-defined beam shape that would indicate spatial coherence. Such findings further support our interpretation that the threshold behavior marks the activation of CW-excited ASE rather than lasing; further optimization of the cavity design and quality factor may potentially allow laser oscillation to be sustained at the (quasi) CW-operating regime from similar type photonic structures.

In summary, we have demonstrated monolithic integration of green-emitting CsPbBr₃ NCs and polymeric DBRs into high-quality, solution-processed microcavities. In the low-excitation regime below the onset of stimulated emission, coupling to the photonic mode of the cavity results in an overall integrated emission enhancement by a factor of ~ 3 with an amplification by an order of magnitude for the emission part that is resonant to the cavity mode. The observation of high directionality in the emission and the estimation of an appreciable Purcell effect in the NC radiative rate further confirm the efficient coupling of the nanocrystal exciton emission to the cavity photonic modes. At higher excitation fluences, efficient continuous-wave excited ASE is observed with a threshold of 0.14 W/cm² and a narrow fwhm of ~ 1.6 nm. The presented results demonstrate the high potential of simple, solution-processed photonic structures based on LHP NC gain media, for practical, scalable, and low-cost laser applications.

METHODS

Nanocrystal Synthesis. Synthesis of CsPbBr₃ DDAB PbBr₂ nanocrystals is similar to the method described in ref 62.

Substrate Preparation. Quartz substrates with dimensions of 2 × 1.5 cm were used, for the deposition of all-solution-processed microcavities. Prior to the deposition process, the substrates underwent a sequential cleaning with *n*-butyl acetate, acetone, and isopropyl alcohol (IPA). The substrates were then kept in nitric acid for 8 h, allowing the formation of a highly hydrophilic surface. The samples were then washed with deionized water, followed by a sequential cleaning with *n*-butyl acetate, acetone, and IPA, and dried under compressed air.

Polymer-Based DBRs. The DBR mirrors were fabricated via an alternate deposition of CA ($M_n = 30\,000$, $\eta_{ca} \approx 1.46$) dissolved in acetone (14 mg/mL) and PVK (Across Organic, MW = 90 000, $\eta_{PVK} \approx 1.68$) dissolved in toluene (35 mg/mL). The polymer materials (~ 15 μ L) were deposited via a dynamic spin coating approach at 6000 rpm. Each pair of polymer layers was annealed at 70 °C, prior to the deposition of additional layers.

Nanocrystal Film Deposition. LHP NCs in a polystyrene matrix was used as described above (20–30 μ L, ~ 30 mg/mL). Initially the solution was deposited on top of the sample, allowing it to rest for 10 s. Then it was spin-casted under static spin-casting conditions at 1000 rpm for 30 s followed by a drying step at 4000 rpm for another 30 s, producing smooth films. The active layer thickness was determined via the single-point ellipsometry method, measured by a thetametris ellipsometer (FR-PRO).

Optical Spectroscopy. Optical absorbance of films was acquired using a PerkinElmer Lambda 1050 spectrophotometer. Steady-state PL was excited by a 405 nm diode laser and detected via a combination of a 0.75 m Acton750i Princeton spectrometer and a 1024 × 256 pixel PIXIS charge-coupled device camera. Angle-dependent reflectivity and PL measurements were performed in a custom-made goniometer. For the

reflectance measurements, a tungsten-halogen white light source was used as our broadband light source coupled into a 600 μ m core multimode fiber. The white light was then focused onto the sample in a 2 mm spot diameter. The reflected beam was collected by a 100 μ m core multimode fiber at approximately 10 cm distance from the sample with steps of 2°. The collected beam was analyzed by an Ocean Optics spectrometer with a spectral resolution of ~ 1.5 nm. Angle-dependent photoexcitation of the microcavities was performed through the backside of the quartz substrate, using a CW 405 nm diode laser, with the emission coaxially collected every 2° in transmission with a long-pass filter used to block the leakage excitation laser.

Time-resolved PL and TRES were recorded on a FluoroLog FL3 Horiba Jobin Yvon spectrofluorimeter using a monochromator-based time-correlated single-photon-counting method. A 405 nm Delta diode laser with a pulse width of ~ 70 ps was used to excite the samples through the quartz substrate with emission coaxially collected normal to the sample surface. For the TRES measurements the PL decay was recorded every 2 nm with the detection bandwidth set to 2 nm. The measured PL decay curves were fitted by a triple-exponential function, allowing the production of good fits of the PL kinetics. The dynamics can be overall quantified by the average transient PL lifetime τ_{av} , calculated by the equation below:

$$\tau_{av} = \frac{\sum_i A_i \tau_i^2}{\sum_i A_i \tau_i} \quad (8)$$

where τ_i are the decay times extracted from triple-exponential fitting of the PL transients and A_i their corresponding decay amplitudes.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsp Photonics.1c00565>.

Optimization and characterization of all-solution-processed DBRs; AFM characterization of microcavity layers; power-dependent transient PL studies; estimation of PL enhancement factor as a function of detection angle; angle-dependent TRES measurement of the on-resonance cavity; power-dependent studies of reference samples under CW excitation; ASE studies of pristine NC films under nanosecond excitation (PDF)

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Notes

The authors declare no competing financial interest.

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