

Beyond Spin Coating: Homogeneous All-Inorganic Perovskite Films via High-Pressure Recrystallization

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Abstract

Metal halide perovskite semiconductors have garnered attention for their exceptional optical and electronic properties, making them promising for optoelectronic applications. Among these, all-inorganic halide perovskites such as CsPbBr_3 are particularly attractive due to their superior thermal and chemical stability. However, achieving high-quality CsPbBr_3 thin films through solution-based methods remains challenging because of the precursor's low solubility, with most approaches relying on additive or solvent engineering that are often complex and difficult to reproduce. High-pressure recrystallization has recently emerged as a viable method for improving film quality, but comprehensive studies on the effects of pressure on film properties are limited. In this work, we conducted a detailed investigation into the morphological, structural, and optical properties of CsPbBr_3 thin films, comparing non-recrystallized films to those fabricated using high-pressure recrystallization but without any additive or solvent engineering. Optimized recrystallization at 300 bar yields smooth, pinhole-free, single-phase 3D perovskite layers with sub-nanometer roughness. Film thickness is precisely controlled by adjusting the precursor concentration, and high-pressure recrystallization increases both grain size and crystallite size. Optical analysis reveals that the recrystallized films exhibit amplified spontaneous emission with a lower excitation threshold, and more photo-stability. Furthermore, temperature-dependent X-ray diffraction reveals the orthorhombic-to-tetragonal-to-cubic phase transition in the recrystallized films, consistent with reports on single crystals in the literature. Our study provides valuable insights into the effects of high-pressure recrystallization on CsPbBr_3 thin films, highlighting a reproducible and scalable solution-based approach for achieving high-quality films in optoelectronic devices.

Keywords

All-Inorganic perovskites, perovskite thin film, recrystallization, high pressure, homogeneous, crystallinity, amplified spontaneous emission

Introduction

Metal halide perovskites have emerged as leading materials for a broad range of optoelectronic applications, including solar cells,¹ light-emitting diodes (LEDs),² lasers,³ photodetectors,^{4,5} and scintillators.⁶ Their exceptional optical and electronic properties have positioned them at the forefront of technological advancement in these fields. Early research efforts primarily focused on hybrid organic–inorganic perovskites, which demonstrated remarkable features such as high absorption coefficients, tunable bandgaps, and low-cost, solution-processable fabrication, yielding impressive device efficiencies.^{7–9} However, the long-term operational stability of these materials is hindered by the degradation of organic components and halide corrosion under environmental stressors such as moisture and heat.^{10–12} While strategies including passivation, surface engineering, and structural tuning have been employed to mitigate these issues,^{13,14} they often add fabrication complexity and do not fully resolve the intrinsic stability challenges.

To overcome these limitations, all-inorganic perovskites—particularly cesium lead bromide (CsPbBr_3) have garnered increasing attention due to their superior thermal and chemical stability.¹⁵ Nonetheless, the fabrication of high-quality CsPbBr_3 thin films via solution processing remains challenging. The inherently low solubility of CsPbBr_3 precursors in common solvents hampers the formation of uniform, pinhole-free films with good coverage and crystallinity.^{16–19} Techniques such as solvent engineering and anti-solvent dripping have been employed to control nucleation and film growth, thereby improving film quality.^{20–22} However, these approaches often suffer from poor reproducibility, limited scalability, and environmental concerns due to the use of toxic anti-solvents.²³

As a promising alternative to additive or solvent engineering, pressure-assisted processing has recently shown potential to enhance both the structural and optoelectronic properties of CsPbBr_3 films. For instance, Pourdavoud et al. demonstrated that applying 100 bar of pressure during annealing led to smooth films with large crystal grains and improved lasing performance,²⁴ while Tatarinov et al. reported enhanced photostability under ambient con-

ditions without encapsulation using a pressure-assisted recrystallization strategy.²⁵ Despite these promising results, prior studies have primarily emphasized qualitative improvements, leaving key questions unanswered regarding the systematic influence of pressure on film morphology (e.g., roughness, coverage), phase purity, and crystallinity.

In this work, we present a detailed and systematic investigation of high-pressure recrystallization for the fabrication of CsPbBr₃ thin films using an automated imprinting platform. This method allows precise and reproducible control of pressure and temperature, enabling a robust exploration of how both processing pressure and precursor concentration affect film characteristics. Morphological analysis reveals that optimized recrystallization yields smooth, dense, and pinhole-free films with nanometer-scale surface roughness. Optical characterization shows enhanced amplified spontaneous emission (ASE) and larger photostability at high excitation fluence. Structural analysis confirms the typical orthorhombic–tetragonal–cubic phase transitions known in CsPbBr₃ single crystals, in contrast with non-recrystallized thin films. Our study provides new insights into the pressure–morphology–optical property relationship and demonstrates a scalable, additive-free, and environmentally benign route for producing high-quality all-inorganic perovskite films suitable for next-generation optoelectronic devices.

Experimental Methods

Sample Preparation

The perovskite solutions were prepared by mixing different precursor concentrations of CsBr and PbBr₂ in the same molar ratio (1.5:1) in dimethylsulfoxide (DMSO). In particular, three precursor PbBr₂ concentrations have been studied: 0.23 M, 0.3 M and 0.4 M. The prepared solutions were stirred at 60°C in a nitrogen-filled glovebox overnight. The solutions were then filtered using a 0.2 µm PTFE filter before perovskite film deposition. In parallel, the Si/SiO₂ substrates were ultrasonically cleaned in acetone, ethanol, and isopropanol for 15 minutes in

each solvent and subsequently dried with N_2 gas. The substrates were treated with UV-ozone for 15 minutes to remove residual organic molecules and to improve hydrophilicity. Finally, the substrates were loaded into an N_2 -filled glovebox for perovskite deposition.

Figure 1 illustrates the fabrication process used to achieve high-quality $CsPbBr_3$ films with full coverage and high crystallinity, involving two crystallization steps. The pristine layer (Figures 1(a) and 1(b)) was first crystallized through a simple annealing process. Subsequently, the film was recrystallized (Figures 1(c) and 1(d)) under both high pressure and a controlled crystallization temperature.

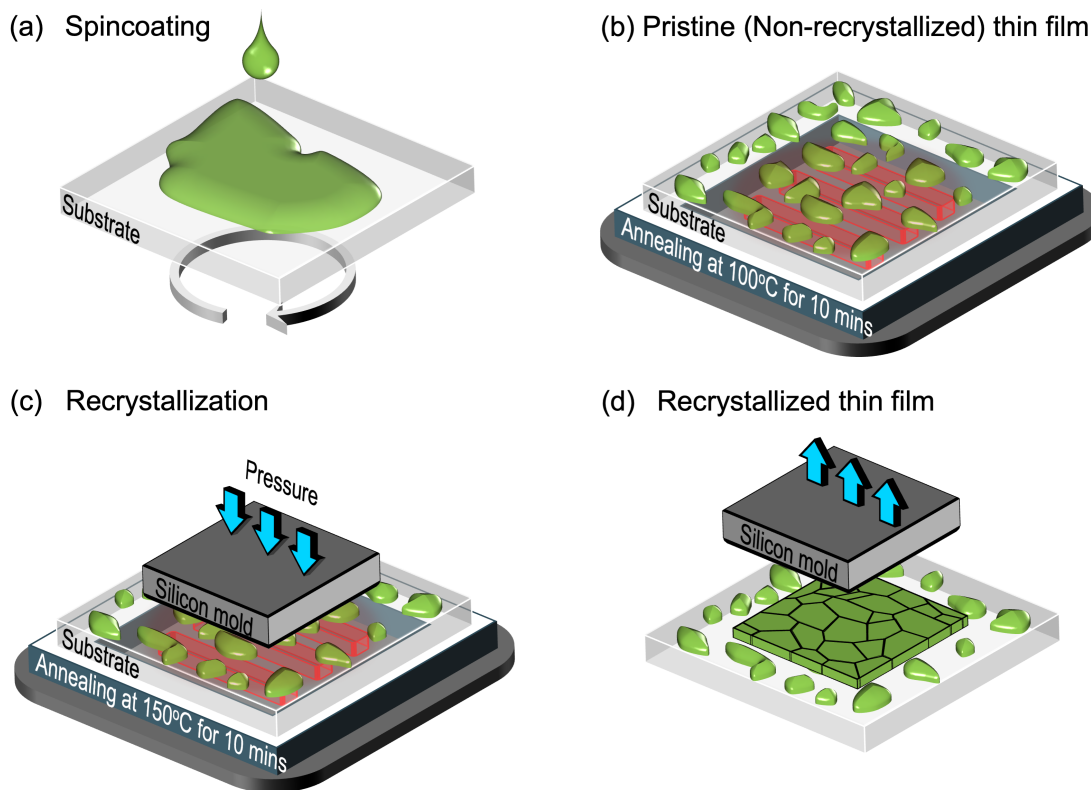


Figure 1: Schematic of the two-step fabrication process: the pristine spin-coat includes (a) spin coating of the precursor solution, followed by (b) annealing to induce crystallization, also called non-recrystallized thin film. (c) Recrystallization involves applying high pressure and a controlled crystallization temperature, resulting in (d) a flattened perovskite layer with larger crystal sizes and improved surface coverage.

During the first crystallization step, the precursor solution was spin-coated onto a Si/SiO_2

substrate at 3000 rpm for 80 seconds, as shown in Figure 1(a). Subsequently, the samples were annealed at 100°C for 15 minutes to evaporate the solvent, allowing the perovskite to crystallize into a nanocrystalline layer, also called a non-recrystallized layer, as illustrated in Figure 1(b).

Recrystallization was performed using an imprint system (NPS300 – Smart Equipment Technology Corporation, France), which simultaneously applied high pressure to the crystallized layer using a flat silicon mold while maintaining a temperature of 150°C, as depicted in Figure 1(c). The temperature was held constant for 10 minutes while the pressure was gradually released as the system returned to room temperature. After removing the silicon mold, the perovskite layer was flattened, resulting in larger crystal sizes and improved surface coverage, as shown in Figure 1(d). This recrystallization process lasted a total of 35 minutes, bringing the entire fabrication process to under one hour. Furthermore, the area of the recrystallized layer depends solely on the size of the silicon mold, demonstrating that this method enables simple and fast fabrication of perovskite layers over a large area. In this study, we focused on examining the impact of pressure on the recrystallized layer. To ensure consistency, the silicon mold size was fixed at 1 cm $\times$ 1 cm with a thickness of 0.5 mm.

Characterization Tools

In this work, multiple standard techniques were employed to investigate the fabricated layers. The structural properties of the perovskite layers were analyzed using X-ray diffraction (XRD) measurements performed with a Rigaku Smartlab diffractometer. This instrument is equipped with a 9 kW rotating anode and a two-reflection Ge (400) crystal monochromator, which selects the Cu $K_{\alpha 1}$ radiation ($\lambda = 1.54056 \text{ \AA}$). Film morphologies were examined using a scanning electron microscope (SEM) and atomic force microscopy (AFM).

For optical characterization, the absorption spectra of the layers were measured with a CARY UV-VIS-NIR spectrometer. Home-built macro-photoluminescence (macro-PL) setup was used to study photoluminescence (PL) and amplified spontaneous emission (ASE). We

used an amplified TiSa laser (800 nm, 4 kHz, 80 fs, ~ 1.5 mJ) as the excitation source, which is further frequency-doubled to 400 nm and focused on the sample with a focal length of 50 or 100 mm (corresponding beam section: 3.4×10^{-4} and 1.4×10^{-4} , respectively). The PL was collected at 90° or 45° and focused on an optical fiber spectrometer using a set of lenses. For the 90° configuration, the excitation was set right at the edge of the sample to limit re-absorption effects.

Results and discussions

Morphological and Structural Characterizations

The Pristine Spin-coated (Non-Recrystallized) Thin Film

The pristine (non-recrystallized) surface morphology of the CsPbBr₃ thin layers for all three concentrations exhibits randomly distributed grains with a significant density of pinholes, as shown in Figures 2(a) and 2(b) for the precursor concentration of PbBr₂ of 0.23 M. This low coverage after the first fabrication step is attributed to insufficient wetting and slow solvent evaporation during nucleation. The same properties are observed for the higher concentrations of 0.3 M (Figure S1 (a)) and 0.4 M (Figure S1 (d)). Notably, the CsPbBr₃ films prepared with lower precursor concentrations display smaller pinholes than those with higher concentrations. This can be explained by the lower viscosity of the solution, which promotes better grain distribution and enhanced substrate coverage. Furthermore, Figure 2 (b) is the AFM characterization of the non-recrystallized layer of the concentration 0.23 M, revealing its high roughness corresponding with the root mean square (RMS) of 25 nm. Both layers of 0.3 M (Figure S1(b)) and 0.4 M (Figure S1(d)) exhibit similar surface morphology, with RMS roughness values of 23.3 nm and 33.4 nm, respectively, measured over the AFM-scanned area of $4.0 \mu\text{m} \times 4.0 \mu\text{m}$ (see Table 1). These results indicate significant surface irregularities across the layers, which are consistent with the SEM observations.

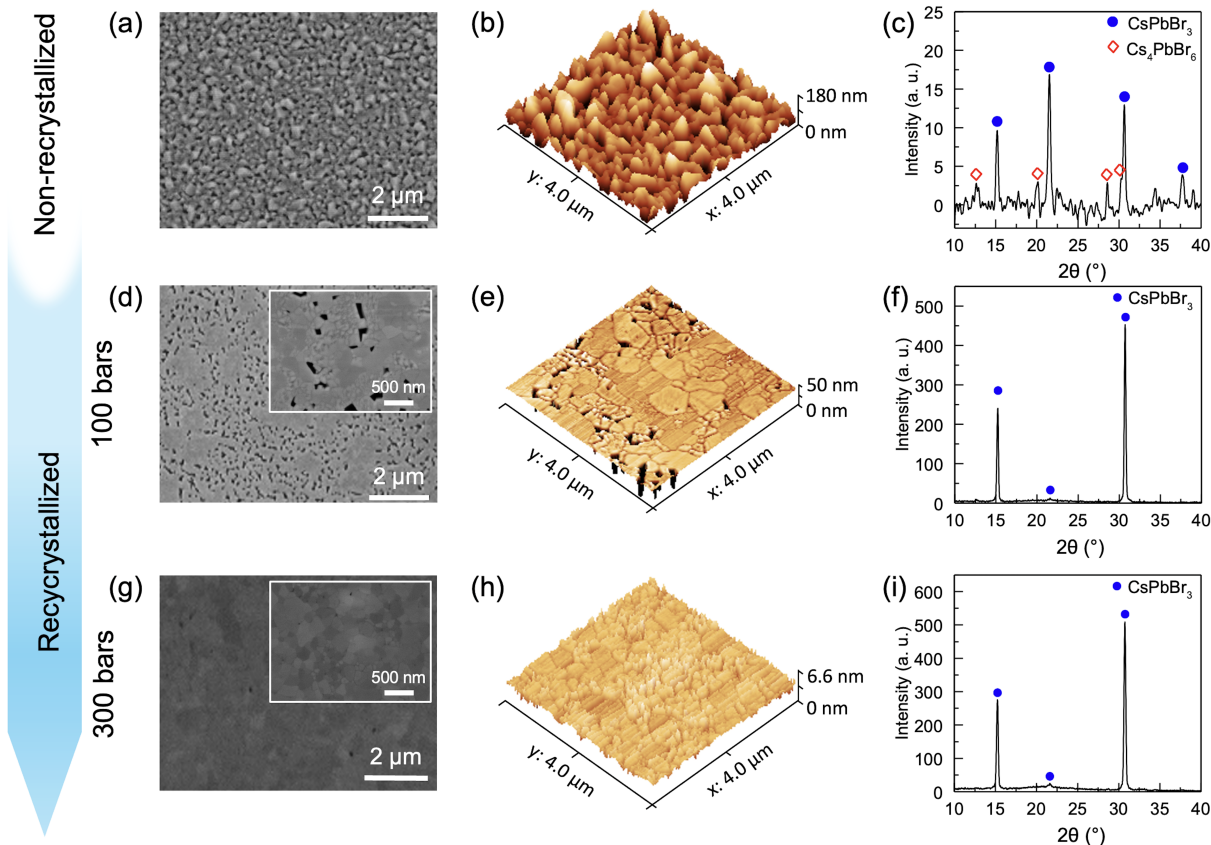


Figure 2: (a-c) are the SEM image, AFM image, and XRD spectrum of the non-recrystallized perovskite film, respectively. (d-f) and (g-i) are the SEM images, AFM images, and XRD spectra of the recrystallized perovskite film under 100 bar and 300 bar, respectively. The precursor concentration of PbBr_2 is 0.23 M.

Additionally, the grain size ranges from 100 to 300 nm with an average value of $\sim 200 \text{ nm} \pm 90 \text{ nm}$ among the three concentrations.

The crystal structure of the non-recrystallized perovskite layers typically exhibits different phases, including 3D (CsPbBr_3), 2D (CsPb_2Br_5), and 0D (Cs_4PbBr_6) phases.²⁶ Figure 2 (c) is the XRD spectra of the layer of 0.23 M precursor showing a mixed phase which comprises both the 3D phase (CsPbBr_3) and the 0D phase (Cs_4PbBr_6). This is indicated by the intense diffraction peaks at $2\theta = 15.21^\circ, 21.50^\circ, 30.60^\circ$, and 37.7° for the 3D phase and the weaker peaks at $2\theta = 12.6^\circ$ and 25.4° for the 0D phase. At a higher concentration of 0.3 M, an additional diffraction peak at $2\theta = 35.1^\circ$ reveals the presence of a 2D phase (CsPb_2Br_5) alongside the 3D and 0D phases,²⁷ as shown in Figure S1 (c). While the 2D phase is gener-

ally considered undesirable, the 0D phase has been reported to improve radiative efficiency in optoelectronic applications, particularly in LEDs, by enhancing luminescence.²⁷ At the highest concentration of 0.4 M, the XRD spectrum in Figure S1 (f) displays only diffraction peaks corresponding to the 3D phase ($CsPbBr_3$), indicating complete crystallization into the single 3D phase. This result suggests that a concentration of 0.4 M is optimal for the one-step fabrication of single-phase $CsPbBr_3$ films.

However, despite the improved phase purity, the crystallinity of the layers remains low, and the high density of pinholes limits their suitability for optoelectronic applications. To address these limitations and enhance the morphological and crystal quality of the $CsPbBr_3$ films, the layers were recrystallized under high pressure at a crystallization temperature of 150°C.

The Recrystallized Thin Film

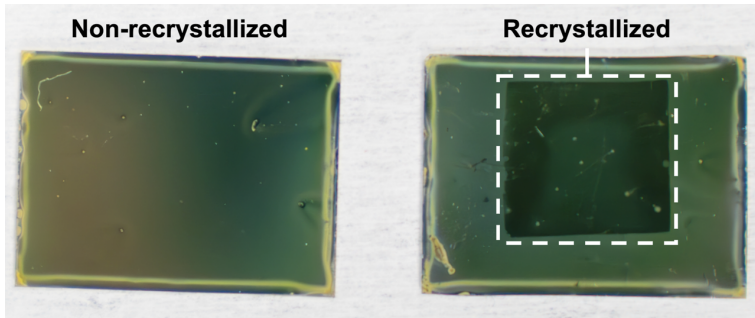


Figure 3: Optical image of the non-recrystallized (left) and the recrystallized (right) $CsPbBr_3$ thin film under 300 bar of pressure, both samples correspond to the precursor concentration of 0.23 M.

Remarkably, applying high pressure results in a glossy layer compared to the diffusive surface of the non-recrystallized layer (Figure 3). Two distinct areas can be observed in the recrystallized sample. The area under pressure of 1 cm² corresponding to the size of the silicon mold shows a homogeneous and mirror-like surface, while the area without applied pressure remains opaque as the non-recrystallized layer. The results suggest that high pressure has flattened the perovskite crystals, leading to a continuous layer.

The application of high pressure significantly improves the uniformity and coverage of the recrystallized CsPbBr₃ layers. Figures 2(d) and 2(g) present SEM images of films prepared with a precursor PbBr₂ concentration of 0.23 M under pressures of 100 and 300 bar, respectively. Although films recrystallized at 100 bar show substantial improvements in coverage compared to the non-recrystallized layers, undesired pinholes persist. In contrast, the film prepared at 300 bar exhibits an almost pinhole-free surface, indicating markedly enhanced coverage. However, it should be noted that prolonged exposure to e-beams during SEM imaging can damage perovskite films and may introduce artifacts such as pinholes.

AFM measurements (Figures 2(e) and (h)) further reveal that increasing the applied pressure results in larger grain sizes and smoother surfaces. At 100 bar, the average grain size is approximately $407 \text{ nm} \pm 147 \text{ nm}$, about twice that of the pristine layer. As the pressure increases at 300 bar, the grain size grows modestly to approximately $463 \text{ nm} \pm 153 \text{ nm}$ at 300 bar. Notably, the recrystallized film at 300 bar achieves an RMS roughness of around 0.6 nm, which is more than 40 times lower than that of the non-recrystallized layer (Table 1). Similar trends were observed for films prepared with precursor concentrations of 0.3 M (RMS = 2.5 nm) and 0.4 M (RMS = 2.2 nm) (Figure S2 and Table 1). Overall, the application of high pressure reduces the surface roughness by up to an order of magnitude (Figure S4(b)), indicating that 300 bar is optimal for fabricating high-coverage, smooth CsPbBr₃ layers.

In addition to enhancing surface coverage, the application of high pressure significantly improves crystallinity and induces single-phase perovskite structures. Figures 2(f) and 2(i) display the XRD spectra of recrystallized layers under various pressures, each exhibiting two prominent peaks at $2\theta = 15.21^\circ$ and 30.70° , corresponding to the (110) and (220) planes of the orthorhombic phase of radiative 3D CsPbBr₃ perovskite.²⁸ Crystallinity was quantified by evaluating the full width at half maximum (FWHM) of the (220) diffraction peak at $2\theta = 30.70^\circ$. For the pristine spin-coated film (Figure 2(c)), the FWHM was 0.340° , whereas for the recrystallized film at 300 bar (Figure 2(i)), it decreased to 0.240° . Using the Scherrer

equation,

$$L = \frac{K\lambda}{B \cos \theta},$$

where $K = 0.94$ is the Scherrer constant, $\lambda = 0.154$ nm is the X-ray wavelength, B is the FWHM, and θ is the diffraction angle, the estimated crystallite size increased from approximately 28 nm for the pristine film to 40 nm for the recrystallized film at 300 bar. This enhancement confirms that high-pressure recrystallization promotes the formation of larger crystals. Furthermore, the XRD peak intensity of the recrystallized layers increased by up to one order of magnitude relative to that of the pristine film, indicating markedly improved crystal organization. High-pressure recrystallization combined with the annealing of 150° likely reduces structural defects and enhances grain density by promoting atomic diffusion and reorganization of interatomic bonds, thereby optimizing bond angles and distances and facilitating uniform recrystallization.

Recrystallized layers prepared with precursor concentrations of 0.3 M and 0.4 M also display high crystallinity and single-phase structures under 300 bar (Figures S2(c) and S2(f), respectively), with significant improvements compared to non-recrystallized films. Interestingly, the XRD peak intensities of these layers are approximately twice as high as those of the 0.23 M layer, and impurity peaks are no longer observed. These results suggest that higher precursor concentrations enhance crystallinity while preserving full surface coverage and the dominance of the 3D perovskite phase.

In conclusion, our findings demonstrate that recrystallization at 300 bar is the optimal condition to achieve the desirable morphological and structural properties required for optoelectronic applications.²⁹ Among the three precursor concentrations investigated, all recrystallized layers exhibit low roughness, large grains, high crystallinity, and a single 3D phase, with the layers prepared from 0.3 M and 0.4 M solutions showing particularly superior crystallinity.

Table 1: Thickness and RMS values for varying precursor concentrations. Note that RMS values are extracted from AFM over the scanned surface, whereas thickness and its standard deviation are measured from SEM at different cleaved-edge positions; therefore, the thickness deviation does not directly correspond to the RMS roughness.

Precursor concentration (M)	Non-recrystallization		Recrystallization (300 bar)	
	Thickness (nm)	RMS (nm)	Thickness (nm)	RMS (nm)
0.23	120 ± 28	25.0	61 ± 9	0.6
0.30	143 ± 25	23.3	75 ± 9	2.5
0.40	163 ± 22	33.4	115 ± 8	2.2

Impact of Recrystallization on Thin Film Thickness

The thickness of the recrystallized layer is a critical parameter to investigate due to its significant influence on the performance of optoelectronic devices such as LEDs and solar cells. We measured the thickness of both the pristine spin-coated and recrystallized layers using 45°-tilted SEM images in (Figure S3), with the results summarized in Table 1. A general trend is observed where the thickness increases linearly with precursor concentration, as shown in Figure S4(a) for both types of layers.

For pristine spin-coated layers, the thickness increases by only 36%, from 120 nm at the lowest precursor concentration to 163 nm at the highest. In contrast, the recrystallized layers show a much more pronounced increase, with the thickness nearly doubling from 61 nm to 115 nm across the same concentration range.

The results also reveal that recrystallization under high pressure systematically reduces the film thickness. This reduction can be attributed to the compaction and densification of the perovskite structure induced by high pressure. Furthermore, the extent of this thickness reduction depends on the precursor concentration. For instance, at a concentration of 0.23 M of precursors, the film thickness is halved (~ 60 nm) after recrystallization, whereas for 0.4 M, the reduction is less pronounced, with the thickness decreasing by approximately 30% (~ 50 nm). This difference can be explained by the initial film density and porosity. At lower concentrations, the spin-coated films are less dense and more porous, making them more susceptible to compaction under pressure. In contrast, higher precursor concentrations

result in denser initial films, allowing less room for compaction and structural reorganization during recrystallization.

These findings highlight the significant impact of high-pressure recrystallization on both film thickness and densification. Importantly, given the consistently high quality and surface coverage of recrystallized layers across all concentrations, this characterization provides valuable guidance for optimizing fabrication parameters to suit specific device applications.

Fiber Texture and Anisotropic Behavior in Recrystallized CsPbBr₃ Films

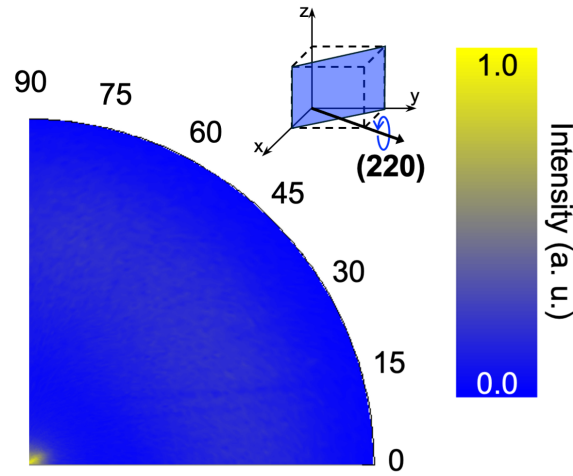


Figure 4: (220) XRD pole figure of the recrystallized layer (0.4 M, 300 bar)

To investigate the distribution of crystalline grain orientations, a pole figure measurement was performed on the recrystallized CsPbBr₃ layer. Figure 4 presents the pole figure of the (220) plane for the recrystallized layer with a precursor concentration of 0.4 M under 300 bar, providing insight into the sample texture. The measurement reveals an intense peak at a polar angle $\theta = 0^\circ$, corresponding to an out-of-plane orientation (perpendicular to the substrate surface). This observation aligns well with the θ - 2θ scan, which exclusively shows (hh0) diffraction peaks, confirming that the CsPbBr₃ perovskite layer exhibits a fiber texture with a predominant out-of-plane alignment direction.

The mechanism driving this specific orientation is not yet fully understood but may be

attributed to the minimization of surface energy and/or the anisotropic mechanical strain applied during the high-pressure recrystallization process. As noted by Wang et al.,³⁰ anisotropic strain and controlled fabrication processes can play critical roles in aligning crystal orientations, particularly in perovskite thin films where quasiepitaxial growth mechanisms often emerge. This alignment not only enhances structural coherence but could also improve charge transport properties and light extraction efficiency, as previously reported for well-aligned perovskite layers.

Interestingly, single-crystal CsPbBr_3 has been shown to exhibit unique excitonic features responsible for giant birefringence effects.³¹ While our recrystallized films do not achieve single-crystal quality, the predominant out-of-plane alignment observed in the fiber texture suggests that excitonic features in our layers are likely to exhibit high anisotropic behavior. Such anisotropy could enable applications leveraging directional optical properties, such as polaritonics, lasers, and advanced light management in optoelectronic devices.

Optical characteristics and amplified spontaneous emission

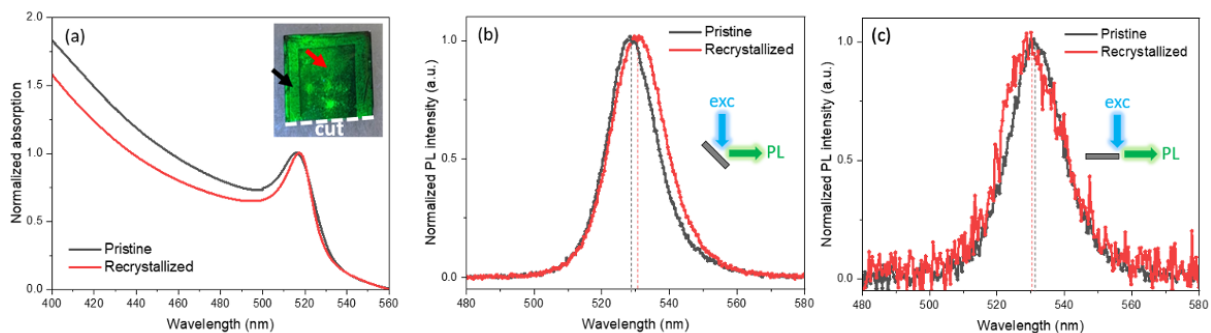


Figure 5: (a) Absorption spectrum of CsPbBr_3 thin film before (black) and after recrystallization (red) on transparent fused silica substrate, the inset is an image of the sample on Si/SiO₂ substrate used for ASE measurement. (b,c) Low excitation fluence photoluminescence (PL) spectra of the non-recrystallized (black) and recrystallized (red) part of the same CsPbBr_3 thin film sample prepared with 0.4 M of precursors on Si/SiO₂ substrate, measured in the (b) 45° and (c) 90° excitation-detection configuration, as schematized in the insets.

Understanding the optical properties of the thin layers of CsPbBr_3 perovskite is a crucial

step in optimizing their efficiency for optoelectronic devices.³² Figure 5 (a) shows the optical absorption spectra of both pristine and recrystallized parts of a film that exhibits a similar profile with a distinct excitonic peak, as reported in single crystals³³ and polycrystalline films.³⁴ The excitonic peak is at around 516 nm for the non-recrystallized part and 517 nm for the recrystallized one. We note a small narrowing of this excitonic peak accompanying the small red shift for the recrystallized part.

Under above-bandgap photoexcitation, both the non-recrystallized and recrystallized regions display bright green photoluminescence, as shown in the inset of Figure 5(a) and Figure S5(b). The apparent higher brightness of the non-recrystallized region arises from its rough surface, which scatters light more efficiently toward the observation direction. In contrast, the recrystallized region, characterized by a much flatter surface, promotes in-plane light confinement through total internal reflection. As a result, part of the emitted light is guided within the film rather than radiated out of plane, giving the recrystallized area a comparatively dimmer appearance at this viewing angle.

The sample was cleaved (white dashed line in Figure 5a, inset) to access both the recrystallized and pristine regions under 90° edge detection. Under low-fluence 400 nm excitation, the recrystallized thin film preserves the single-peak PL spectrum of the non-recrystallized film, as confirmed in both the 45° (Figure 5b) and 90° (Figure 5c) configurations. At 45° detection, the recrystallized film shows a slight redshift of the PL peak (531 nm) and a modest broadening (FWHM = 19 nm) compared with the pristine film (529 nm, FWHM = 17 nm), consistent with crystallite growth that relaxes microstrain and narrows the bandgap.³⁵ Interestingly, in the 90° geometry, the pristine film exhibits a ~ 2 nm redshift relative to 45°, resulting from photon recycling during in-plane propagation across its rough, scattering-dominated surface. Reabsorption of higher-energy photons within the PL-absorption overlap leads to a spectrally filtered, red-shifted edge emission. In contrast, the recrystallized film, with its flat and dense morphology, acts as a planar waveguide that supports low-loss in-plane propagation and minimal reabsorption, yielding nearly identical PL spectra at 45° and 90°.

Overall, the observed PL features are consistent with excitonic emission in polycrystalline films, showing a slight blue-shift relative to single crystals.³³

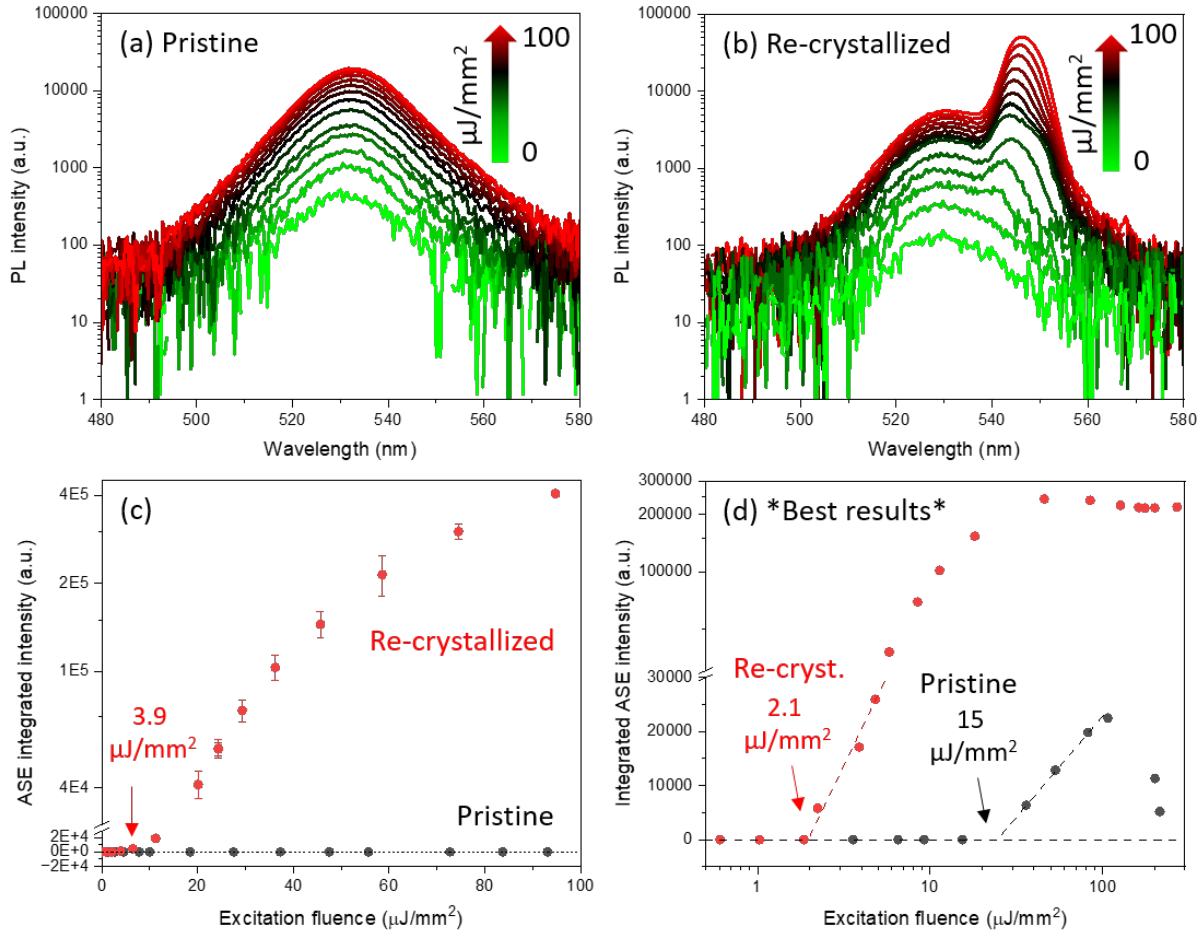


Figure 6: PL spectra of (a) the non-recrystallized part and (b) the recrystallized part (0.4 M) with increasing excitation fluence. (c) Integrated ASE intensity plotted versus the excitation fluence, for the corresponding samples. (d) Integrated ASE intensity obtained for the best samples.

Under high excitation fluence (up to 200 $\mu\text{J}/\text{mm}^2$), the recrystallized area exhibits distinct amplified spontaneous emission (ASE) behavior, which is hardly observed in the non-recrystallized part. Indeed, Figure 6 (a) demonstrates typical PL spectra evolution of the non-recrystallized part according to increasing exciting fluence (up to 100 $\mu\text{J}/\text{mm}^2$), showing an identical profile of the spontaneous emission with PL peak around 529 nm. However, apart from the typical spontaneous emission peaking at 530 nm, a second sharp peak (546

nm) appears in the PL spectra of the recrystallized layer with fluence above $5 \mu\text{J}/\text{mm}^2$, as shown in Figure 6 (b), which is a characteristic of ASE. The only rare non-recrystallized area found to exhibit ASE behavior (Figure S6 (a)) is at the edge of the substrate, which is specifically higher in concentration as a result of the spin-coating process. The ASE peak of this non-recrystallized part is recorded at around 539 nm as the fluence reaches $20 \mu\text{J}/\text{mm}^2$. Notably, all ASE measurements are performed with 90° configuration to maximize the contribution of the ASE relative to the spontaneous emission.³⁶

To describe the ASE spectra in more detail, the PL spectra are fitted with two Gaussian peaks, one of which corresponds to the spontaneous emission at shorter wavelength and the other corresponds to the ASE contribution at the longer wavelength, as illustrated in Figures S6 (c) and (d) in the SI. In general, the difference in energy between the center of the spontaneous emission and the position of the rising ASE is about 40 meV, which is attributed to the inter-exciton interactions.³⁶ Further redshift in ASE is observed at higher fluence, which can be attributed to the negative contribution of reabsorption in the optical gain.

The ASE threshold is a key parameter in characterizing amplified spontaneous emission, typically defined as the intersection point of the linear fits to the integrated PL intensity as a function of excitation fluence before and after the onset of ASE. As demonstrated in Figures 6(a) and 6(b), the recrystallized CsPbBr₃ layer displays a clear threshold at $3.9 \mu\text{J}/\text{mm}^2$, a feature absent in the pristine layer of the same sample (Figure 6 (c)). Taking into account the measurement in different positions, Figure 6 (d) indicates that the best threshold of the pristine part is found to be approximately $15 \mu\text{J}/\text{mm}^2$ (or $1500 \mu\text{J}/\text{cm}^2$), which is more than 7 times higher than the best value achieved by the re-crystallized part ($2 \mu\text{J}/\text{mm}^2$ or $200 \mu\text{J}/\text{cm}^2$). This significant reduction in threshold is attributed to the enhanced crystallinity and smoother morphology of the recrystallized layer, which minimizes both scattering and defect states at grain boundaries, thereby increasing optical gain within the excitation volume. However, the overall thickness of the pristine part is larger than the recrystallized

one. This affects the ASE threshold, in addition to sample density and quality (presence of defect, scattering surface). The threshold values are consistent with previous reports for CsPbBr₃ nanocrystal thin films,^{37,38} CsPbBr₃ colloidal nanocrystals,³⁹ and bulk CsPbBr₃ single crystals.^{40–42}

Not only being much more efficient in generating ASE, the recrystallized layer also shows enhanced photo-stability at high excitation fluence (a few tens of $\mu J/mm^2$). As shown in Figure 6 (d), the integrated PL reaches its saturation at the fluence around 60 $\mu J/mm^2$ and maintains almost constant intensity under the excitation beyond 100 $\mu J/mm^2$. On the contrary, the ASE from the pristine part quickly degraded as the fluence exceeds 100 $\mu J/mm^2$. This could be explained by the absence of "hot spots" in the recrystallized part that can occur in rough surfaces and the better crystallinity, hence allowing an easier heat dissipation mediated by phonons for the recrystallized CsPbBr₃.

Overall, the recrystallized film, characterized by superior film quality and well-defined grains, exhibits enhanced ASE process. The difference with the pristine part not only confirms the high optical quality of the recrystallized film but also underscores the critical role of processing conditions in dictating the emission properties of perovskite thin films.

Phase Transition Dynamics in Recrystallized Thin Films

Although phase transition behavior has been extensively reported on CsPbBr₃ single crystal,^{43–47} it was scarcely studied in CsPbBr₃ perovskite polycrystalline layers^{48,49} due to their low crystallinity. Given the high crystallinity of our recrystallized layers, we anticipated that phase transition behavior could be observed. To investigate this, we performed temperature-dependent XRD measurements on our recrystallized CsPbBr₃ layers. Such study would be challenging in non-recrystallized thin layers due to insufficient crystallinity and the coexistence of multiple phases, including 0D and 2D perovskites.

Figure 7(a) shows XRD patterns of the recrystallized layer under 300 bars with a pre-

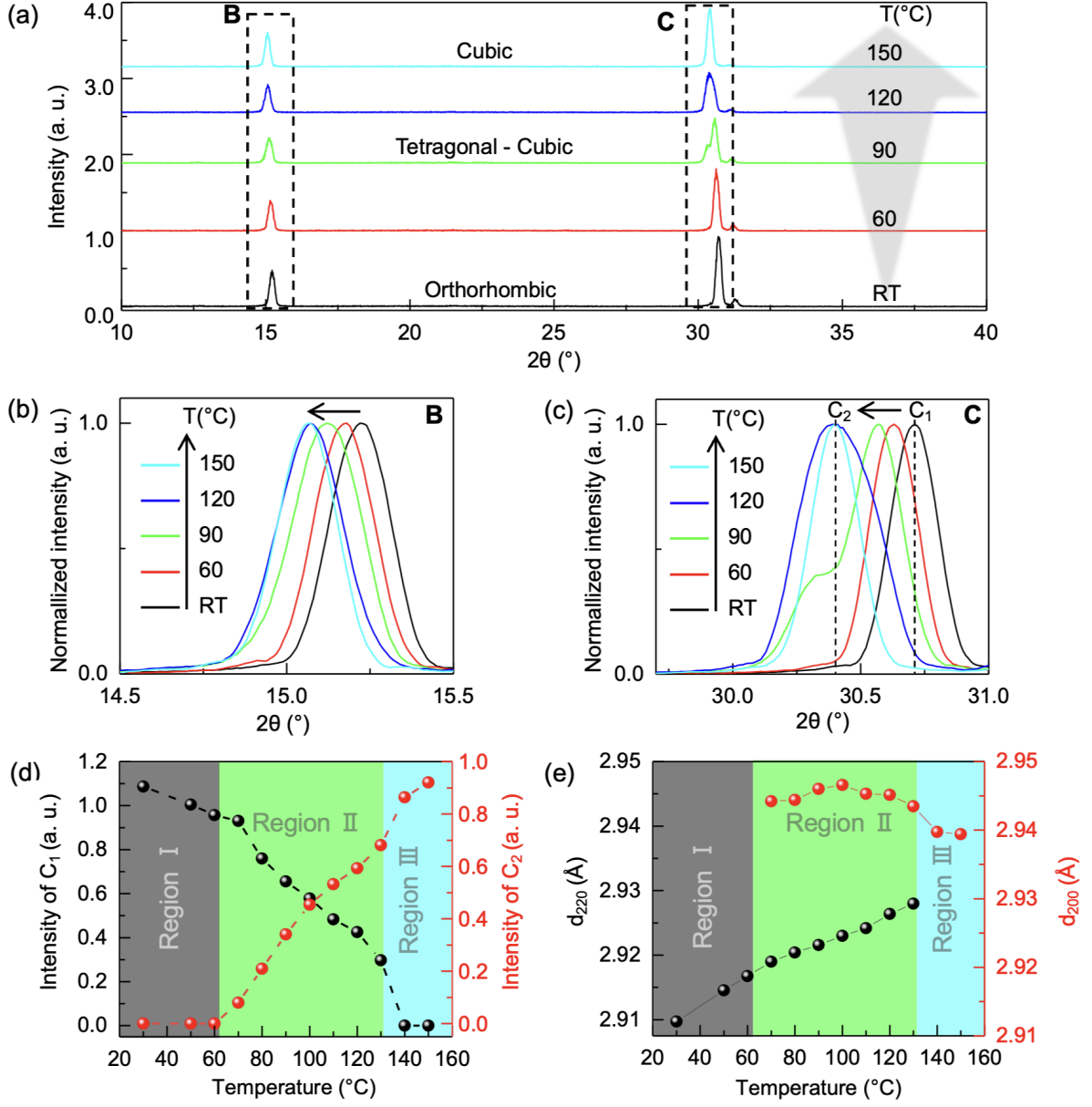


Figure 7: (a) XRD patterns of prepared CsPbBr₃ thin films with the higher precursor concentration at room temperature, 60, 90, 120 and 150°C. (b) and (c) presents the enlargement normalized XRD spectrum of the two principal peaks B and C at $2\theta = 15.21^\circ$ and 30.70° , respectively. (d) The intensity of XRD peak C_1 and C_2 which respectively correspond to (220) and (200) plane as a function of temperature. (e) The interplanar distance of (220) and (200) plane as a function of temperature.

cursor concentration of 0.4 M at room temperature, 60°C, 90°C, 120°C, and 150°C. The data reveal a clear shift of the Bragg peaks (B) at $2\theta = 15.21^\circ$ and (C) at $2\theta = 30.70^\circ$ to

lower angles as the temperature increases. These shifts, better emphasized in Figures 7(b) and 7(c), suggest a structural transformation from the orthorhombic to the cubic phase. Specifically, at $T < 60^\circ\text{C}$, peak B at 15.21° corresponds to the (110) plane, and peak C at 30.70° corresponds to the (220) plane, consistent with the orthorhombic phase. As the temperature increases to $T = 150^\circ\text{C}$, peak B shifts to 15.11° and peak C shifts to 30.40° , corresponding to the (100) and (200) planes of the cubic phase. A similar phenomenon was previously reported for epitaxially grown CsPbBr_3 thin films by Yifan et al.⁵⁰

More specifically, for peak C in Figure 7(c), a shoulder peak appears at 90°C , marking the evolution of the phase transition. This phase transition is further characterized by examining the intensity evolution and interplanar distances of the corresponding diffraction planes (hkl), as shown in Figures 7(d) and 7(e). Based on the 2θ positions, we indexed the diffraction peaks as follows: (220) for the orthorhombic and tetragonal phases (peak C_1) and (200) for the cubic phase (peak C_2). The analysis reveals three distinct regions of interest:

1. **Region I ($T < 60^\circ\text{C}$):** A linear evolution of the interplanar distance of the diffraction peak at $2\theta \approx 30.70^\circ$, corresponding to (220) of the orthorhombic phase.
2. **Region II ($60^\circ\text{C} < T < 140^\circ\text{C}$):** A coexistence of the tetragonal and cubic phases, marked by the evolution of interplanar distances and intensities of diffraction peaks at $2\theta \approx 30.50^\circ$ (tetragonal, (220)) and $2\theta \approx 30.40^\circ$ (cubic, (200)). A significant shift in interplanar distances during this region highlights the phase transition from the orthorhombic phase to the coexistence of tetragonal and cubic phases.
3. **Region III ($T > 140^\circ\text{C}$):** Suppression of the orthorhombic peak and stabilization of the interplanar distance corresponding to (200), marking the complete transition to the cubic phase.

To further support our findings, we evaluated the thermal expansion coefficient α by fitting the linear evolution of the interplanar distance d_{220} in Figure 7(e). The analysis reveals two distinct values of α , corresponding to Region I and Region II: $\alpha_I = 0.8096 \times 10^{-4}$

K^{-1} and $\alpha_{II} = 0.5008 \times 10^{-4} \text{ K}^{-1}$. These values are significantly higher than those reported for single-crystal CsPbBr_3 ,⁵¹ likely due to the differences in structure and thermal behavior between recrystallized thin films and single crystals.

Similar phase transition behavior was also observed for recrystallized layers with lower precursor concentrations of 0.23 M and 0.3 M, as shown in Supporting Information (Figure S7). On average, the cubic phase (region III) remains as the temperature is higher than $\approx 130^\circ\text{C}$ which is in good agreement with the literature.^{43–46,48} On the other hand, the transition point between the orthorhombic and tetragonal phase is around 70°C which is slightly lower than the reported value.^{43–46,48} Interestingly, the recrystallized films reverted to the orthorhombic structure upon cooling to room temperature, demonstrating the reversibility of the phase transition.

Conclusion

In conclusion, our study demonstrates that recrystallization under optimized pressure associated with an annealing 150°C is a highly effective method for enhancing the morphological, structural, and optical properties of CsPbBr_3 thin films. Through a systematic exploration of various pressures and precursor concentrations, we identified 300 bar as the optimal condition, achieving high surface coverage, a pure 3D-phase crystal structure, and sub-nanometric surface roughness. Moreover, the ability to precisely control the film thickness—from 60 nm to 110 nm by varying the PbBr_2 precursor concentration from 0.23 M to 0.4 M, further highlights the versatility of this approach while maintaining consistent film quality.

Beyond the morphological improvements, the textural orientation of the recrystallized thin films was thoroughly characterized, offering valuable insights into their potential for optoelectronic applications. Optical measurements revealed that the recrystallized films exhibit enhanced ASE under optical excitation. Notably, this ASE originates from free excitons in CsPbBr_3 , which underscores the films' suitability not only for high-performance

LEDs and lasers but also for exploring the strong coupling regime between excitons and photons—a key phenomenon in the development of advanced polaritonic devices.

In addition to their optical properties, we systematically investigated the temperature-dependent phase transition behavior of the recrystallized films. Our results revealed a reversible structural transformation between orthorhombic, tetragonal, and cubic phases, providing valuable insights into the thermal stability of these materials. This reversible phase behavior further highlights the potential of recrystallized CsPbBr₃ thin films as phase-change materials for advanced applications.

Overall, recrystallization via pressure represents a fast, scalable, and effective method for producing high-quality all-inorganic CsPbBr₃ thin films. By providing detailed insights into their morphological, optical, and phase-transition properties, this study paves the way for the development of pressure-recrystallized perovskite thin films in a wide range of optoelectronic devices and fundamental photonic applications.

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Supporting Information Available

In the Supplemental Material, additional details and results are provided, including 1) morphological and structural characterizations; 2) analysis methods for ASE results; and 3)

phase transition behavior of recrystallized films with PbBr_2 precursor concentrations of 0.23 M and 0.3 M

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