

# Ultrafast Dynamics of Pressure-Activated Energy Funneling and Amplified Spontaneous Emission in Microscale Quasi-2D Perovskites

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## Article

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**Ultrafast Dynamics of Pressure-Activated Energy Funneling and Amplified  
Spontaneous Emission in Microscale Quasi-2D Perovskites**

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## Abstract

Quasi-2D perovskites are attractive for optical gain because of their large exciton binding energies, built-in quantum wells, and efficient energy funneling. However, for micro- and nanolasers, these same properties create a fundamental problem: the energy funnel relies on a statistical mix of different phases, and miniaturize the device inevitably disrupts this delicate distribution—causing the optical gain to collapse. Here we show that high-pressure engineering, a clean and controllable approach, breaks this size-imposed limitation. Applying hydrostatic pressure to microscale  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  films rebuilds efficient energy funneling in microscale  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  perovskites, activating amplified spontaneous emission (ASE) that is otherwise absent. A modest 0.14 GPa turns on ASE, and at 0.41 GPa the ASE threshold drops by 50%. *In situ* structural and femtosecond transient absorption measurements reveal a two-step compression-mode transition that directly resolves accelerates energy transfer from low- $n$  to high- $n$  phases and extends the high- $n$  carrier lifetime by 60%. This work uncovers the ultrafast dynamics and photophysical mechanism behind pressure-tuned energy transfer and opens a new route to chip-scale coherent light sources from quasi 2D perovskite film, with direct implications for ultrafast nonlinear optical probes of quantum materials.

**Keywords:** quasi-2D perovskites, ASE, high-pressure, energy funneling, carrier dynamics

## Introduction

Quasi-two-dimensional (quasi-2D) perovskites are attractive gain materials for optical amplification because of their large exciton binding energy, improved stability, and natural multiple quantum well structures<sup>1-3</sup>. In these materials, low- $n$  phases (wide bandgap) absorb light and transfer excitons to high- $n$  phases (narrow bandgap) via Förster resonance energy transfer (FRET), allowing population inversion at relatively low excitation densities<sup>4,5</sup>. This “energy cascade” is often seen as an inherent advantage.

However, this same advantage creates a fundamental problem upon device miniaturization. Efficient energy funneling relies on a statistical mix of phases with different  $n$  values spread across the film. As the lateral size is scaled down to the micro-/nanoscale, the probability of preserving enough low- $n$  domains to feed the high- $n$  emitters drop sharply. Below a certain size threshold, the cascade breaks down and optical gain collapses, which is not merely a practical nuisance, but a fundamental limitation of the multiphase cooperation. As a result, maintaining stable amplified spontaneous emission (ASE) and optical gain in micro-/nanoscale continuous films have remained out of reach, despite successful demonstrations in nanowires, microplatelets, and single crystals<sup>6-8</sup>. A natural question arises: can an external stimulus break this size-imposed limit?

Here, we show that high-pressure engineering can do exactly that. We start with a  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  film that shows no ASE at ambient pressure. Applying a modest compression of 0.14 GPa turns on ASE. At 0.41 GPa, the ASE threshold drops by half. This is not a minor improvement; it goes against the expected trade-off between size and light amplification performance. In fact, the pressure compressed microscale film performs as well as or even better than a large-area film at ambient pressure.

What underlying mechanism drives this counterintuitive recovery? Using multiple *in situ* spectroscopic probes, we identify a two-step compression mode transition. Under mild pressure, the interlayer spacing contracts first, reducing the donor–acceptor distance and reverse the hindered energy transfer from low- $n$  phases to high- $n$  phases by nearly an order of magnitude. More unexpectedly, the exciton lifetime in high- $n$  phases increases by 60% at 0.41 GPa, which points to suppressed Auger

recombination, a major obstacle for establishing population inversion at high carrier densities.

Together, these two effects create a synergy: faster energy delivery into the gain region and slower exciton loss there. Pressure turns the “curse of miniaturization” into a “blessing of compression.” Our work introduces mechanical tuning of the energy cascade as a new design dimension for micro-/nanoscale coherent light sources, on par with composition, thickness, and cavity geometry, and opens a practical route to high-performance, addressable chip-scale gain media from quasi-2D perovskite films.

## Results

### Characterization of $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$ thin films under ambient conditions

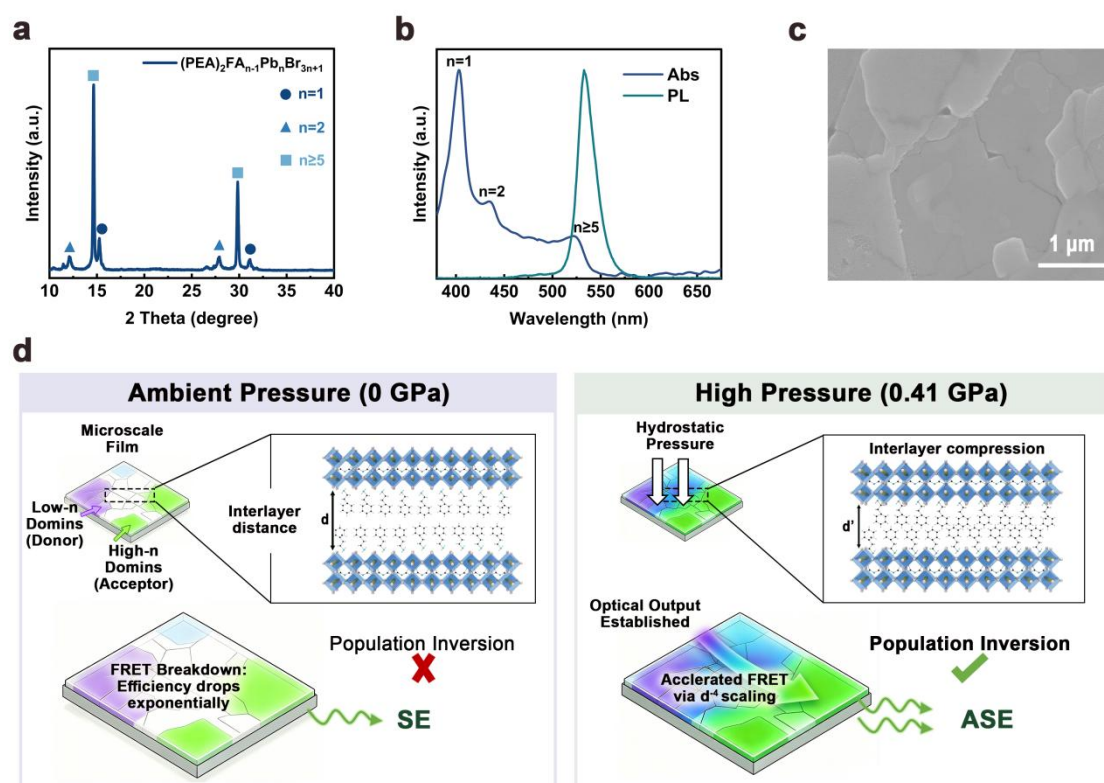


**Fig. 1 Structural and optical characterizations.** **a** XRD patterns, **b** steady-state absorption and PL spectra, and **c** SEM image of the  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  thin films. **d** Schematic illustration of the evolution of the structural and ASE behavior of the  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  thin films upon ambient condition and compression.

Quasi-2D  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  thin films were fabricated using a one-step spin-coating method in ambient atmosphere (**Experimental details**). The X-ray diffraction

(XRD) patterns in **Fig. 1a** exhibit two dominant peaks at  $14.9^\circ$  and  $30.1^\circ$ , corresponding to perovskite phases with  $n \geq 5$ . Other lower-intensity peaks located around these two main peaks provide evidence for 2D perovskite phase generation ( $n < 5$ ), indicating the coexistence of both 3D and 2D phases arising from the steric hindrance of long-chain  $\text{PEA}^+$  cations<sup>9,10</sup>. The steady-state absorption and photoluminescence (PL) spectra of the films are shown in **Fig. 1b**. Multiple strong absorption peaks at  $\sim 405$ ,  $436$ , and  $520$  nm correspond to  $n = 1$ ,  $2$ , and  $n \geq 5$  phases, respectively, confirming the presence of a mix of low- $n$  and high- $n$  phases within the  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  thin films. The PL spectra exhibit a single emission peak centered at  $532$  nm, derived from free-exciton recombination in  $n \geq 5$  phases. The scanning electron microscope (SEM) images (**Fig. 1c**) reveal a uniform and compact morphology, thereby eliminating the potential interference of film quality defects in the subsequent high-pressure investigations. Based on this mixed-phase structural platform, **Fig. 1d** schematically illustrates the core strategy of this work: using high pressure to promote efficient energy funneling in microscale quasi-2D perovskite films, thereby reversing the hindered energy transfer from low- $n$  phase donors to high- $n$  phase acceptors. Under moderate compression, the reduced interlayer spacing accelerates interphase exciton transfer, providing a clean and controllable approach for the reactivation and optimization of ASE.

### ***Pressure-induced ASE reactivation***

The significant energy cascade effect resulting from the multi-phase mixing in quasi-2D perovskite films necessitates that, in order to harness this advantage for practical micro/nanoscale optical amplification, quasi-2D perovskite films with minimal physical dimensions must still possess excellent performance as gain media to achieve efficient optical amplification. However, a strong size dependence of ASE behavior, mediated by the energy transfer among these multiple quantum wells, is observed in the present work. **Fig. 2a** shows the emission spectra of the  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  thin films with different size ranging from  $225 \text{ mm}^2$  to  $0.04 \text{ mm}^2$  at a constant pump fluence of  $750 \mu\text{J cm}^{-2}$ . Indeed, we observe a clear dependence of optical gain on the film size; the ASE threshold increases monotonically from  $52.3 \mu\text{J cm}^{-2}$  to  $182.6 \mu\text{J cm}^{-2}$  (**Fig. 2b** and **Fig. S1**). As film size decreases below a certain area threshold of  $0.04$

mm<sup>2</sup>, the ASE eventually disappears. This phenomenon can be attributed to the random distribution of quantum wells with varying  $n$ -values in quasi-2D perovskite thin films as plotted in **Fig. 1d**. When the lateral dimensions are reduced to the microscale, the statistical distribution of different  $n$ -phase domains is disrupted, causing the physical distance between donors and acceptors to exceed the effective energy transfer range. Consequently, the efficient energy cascading from low- $n$  to high- $n$  phases becomes fragmented, resulting in suppressed exciton funneling and unstable optical gain<sup>11,12</sup>.

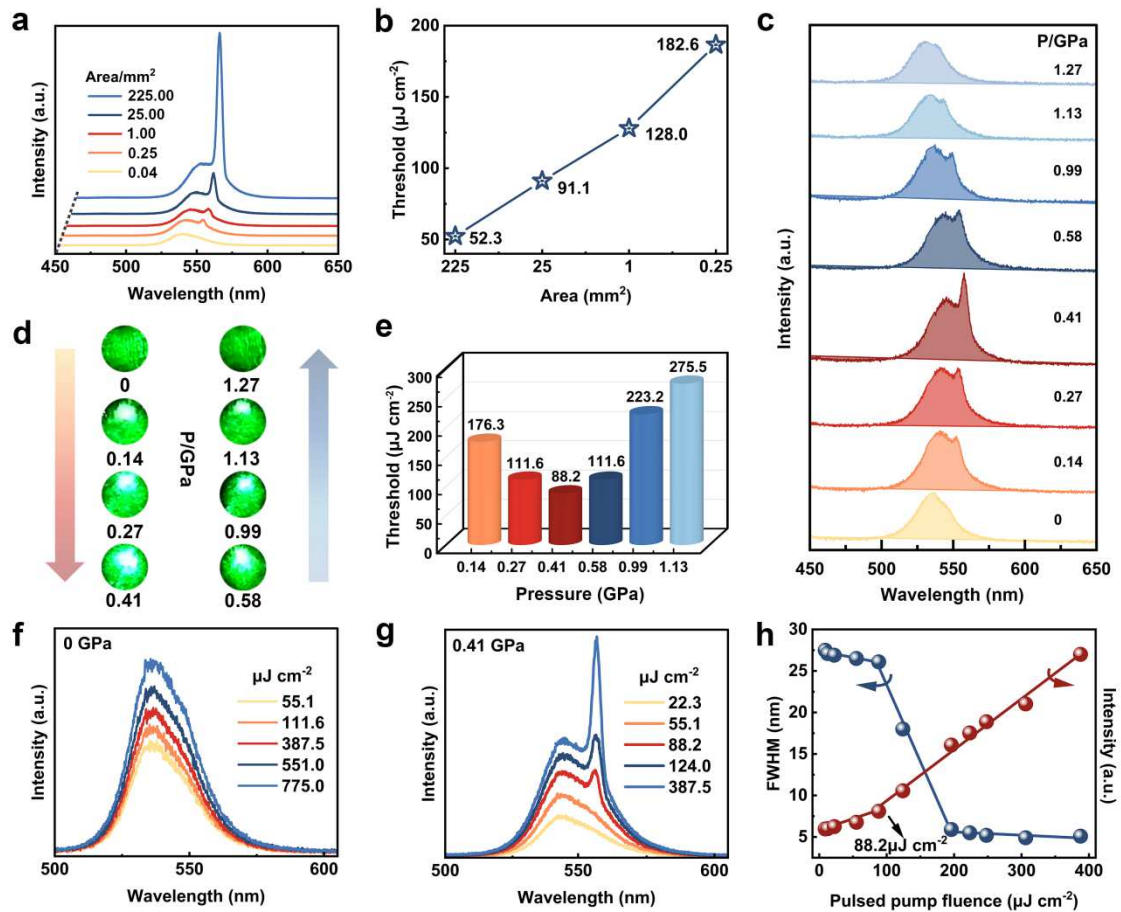


**Fig. 2 Size effect and pressure modulation of ASE in (PEA)<sub>2</sub>FA<sub>n-1</sub>Pb<sub>n</sub>Br<sub>3n+1</sub> thin films. a**

Size-dependent ASE emission spectra of (PEA)<sub>2</sub>FA<sub>n-1</sub>Pb<sub>n</sub>Br<sub>3n+1</sub> thin films. **b** The variation of ASE threshold with film area size. **c** Pressure-dependent ASE spectra, and **d** corresponding micrographs of excite area upon compression. **e** Evolution of the ASE threshold with applied pressures. Power-dependent emission spectra at **f** ambient condition and **g** 0.41 GPa. **h** Output intensity and FWHM versus the pumped density at 0.41 GPa

143 This leaves us with a critical problem: shrink the film below  $0.04\text{ mm}^2$ , and the  
144 energy cascade falls apart. One might think this simply an inherent limitation of  
145 quasi-2D perovskites that the statistical mix of phases cannot be preserved at small  
146 scales. However, we approached this from a different angle: can we force the distinct  
147 phases to interact efficiently, even when the film is miniaturized? Our answer is high-  
148 pressure engineering. Instead of altering the chemical composition or the macroscopic  
149 film structure, we utilized mechanical compression to squeeze the layers together. The  
150 concept is straightforward: reduce the interlayer distance, bring low- $n$  and high-  
151  $n$  domains closer, and reactivate the energy transfer that the small size has disrupted.  
152 We tested this hypothesis on a  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  film of only  $0.01\text{ mm}^2$ , a size that  
153 exhibits absolutely no ASE at ambient pressure, and obtained striking results.

154 A film sample with an area of  $0.01\text{ mm}^2$  was loaded into a symmetrical diamond  
155 anvil cell (DAC) for high-pressure investigation as shown in **Fig. S2**. The dynamic  
156 variation of emission spectra with pressure is shown in **Fig. 2c**. At atmospheric  
157 pressure, the emission spectra exhibit broad spontaneous emission (SE) features,  
158 consistent with the steady-state PL characteristics. However, upon applying a pressure  
159 exceeding  $0.14\text{ GPa}$ , an ASE phenomenon characterized by a sharp increase in emission  
160 intensity and significant full width at half-maximum (FWHM) narrowing emerges,  
161 clearly indicating the reactivating of ASE in microscale film. The ASE intensity initially  
162 increases with continuous pressurization until a turning point is reached at  $0.41\text{ GPa}$ ;  
163 further compression leads to a gradual decrease in ASE intensities, accompanied by the  
164 emission peak transitioning from a redshift to a blueshift. Finally, the ASE completely  
165 quenches at  $1.27\text{ GPa}$ . The corresponding ASE micrographs recorded at different  
166 pressures inside the DAC clearly pronounced variations in emission brightness (**Fig.**  
167 **2d**), which closely match the pressure-dependent evolution of ASE intensity. The ASE  
168 thresholds of the  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  thin films under different pressures exhibit a  
169 consistent pattern. As plotted in **Figs. 2e-h** and **Fig. S3**, the ASE threshold gradually  
170 decreases from  $176.3\text{ }\mu\text{J cm}^{-2}$  at  $0.14\text{ GPa}$  to a minimum of  $88.2\text{ }\mu\text{J cm}^{-2}$  at  $0.41\text{ GPa}$ ,  
171 representing a remarkable reduction of 50%. This notable optimization underscores the  
172 potential of pressure modulation for achieving low-threshold ASE. Upon compression

beyond 0.41 GPa, the threshold progressively increases until ASE is completely quenched at 1.27 GPa. It should be emphasized that such continuous regulation of light amplification behavior is achieved for the first time in perovskite thin films. The in situ pressure-dependent PL evolution of the  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  thin films was simultaneously investigated and exhibited the same evolutionary trend as the ASE, as evidenced by the micrographs and spectra upon compression (Figs. S4-S6).

### Multispectral probe reveals structural evolution under pressure

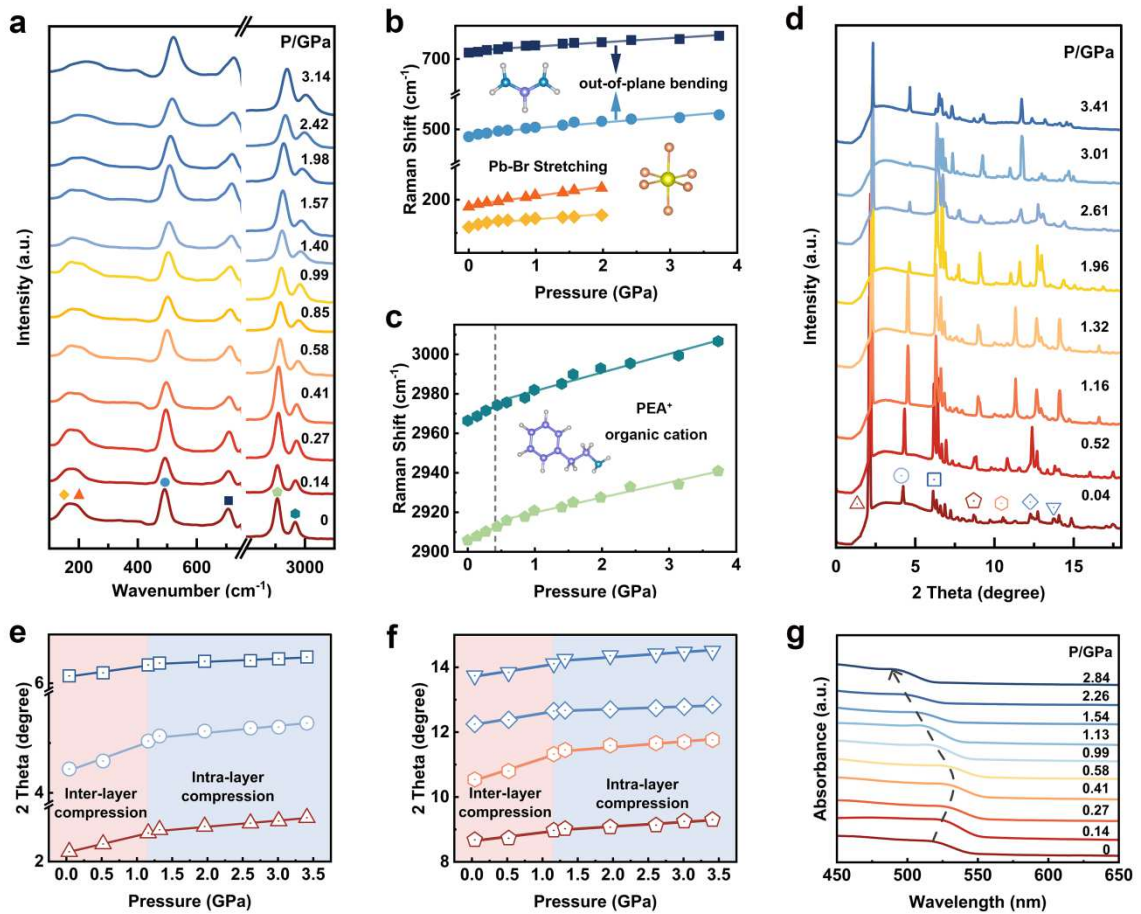


**Fig. 3** *In situ* spectroscopic and structural evolutions under compression for layered perovskites. **a** Raman spectra of the  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  thin films at selected pressures. Pressure-dependent shifting rates of the **b** low-frequency and **c** high-frequency Raman peaks. **d** *In situ* synchrotron XRD patterns of the  $\text{PEA}_2\text{FAPb}_2\text{Br}_7$  single crystals at selected pressures. Shifting rate of the **e** first three and **f** last four diffraction peaks versus applied pressure. **g** *In situ* optical absorption spectra of the  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  thin films at selected pressures.

To investigate the relationship between the aforementioned pressure-induced ASE emission behavior and microstructural evolution, a series of *in situ* high-pressure characterizations were systematically performed. Pressure-dependent Raman spectroscopy, which reflects the structural evolution of the organic layer and its coupling with the inorganic sublattice, was first conducted (**Fig. 3a**). For the (PEA)<sub>2</sub>FA<sub>n-1</sub>Pb<sub>n</sub>Br<sub>3n+1</sub> thin films, six Raman peaks are observed. The low-frequency vibrational modes in the ranges of 150-200 cm<sup>-1</sup> and 500-700 cm<sup>-1</sup> are attributed to the lattice vibrations of the Pb-Br inorganic framework and the out-of-plane bonding of FA<sup>+</sup>, respectively. In contrast, the high-frequency vibrational features in the 2900-3000 cm<sup>-1</sup> region originate from the molecular vibrations of the PEA<sup>+</sup> organic cations. With increasing pressure, all Raman vibrational modes exhibit significant blueshifts, primarily due to pressure-induced bond hardening resulting from lattice contraction. **Fig. 3b** and **3c** present the pressure-dependent evolution of the six Raman mode wavenumbers. The shifting rate of each mode, quantitatively described by the pressure coefficient  $d\omega/dP$  and listed piecewise in **Table. S1**, reflects the compressibility of different vibrational modes. Notably, the interlayer organic cation PEA<sup>+</sup> exhibits a higher  $d\omega/dP$  in the low-pressure regime, specifically, 14.5 and 19.2 cm<sup>-1</sup>/GPa below 0.41 GPa, compared to 7.9 and 9.1 cm<sup>-1</sup>/GPa above 0.41 GPa. This distinct contrast indicates a significantly greater compressibility along the interlayer direction<sup>13</sup>.

This interlayer compression directly promotes energy transfer by reducing the donor-acceptor separation between distinct *n*-value phases. In quasi-2D perovskite systems, FRET efficiency exhibits a strong distance dependence, typically scaling as “ $d^{-4}$ ”, where *d* represents the interlayer distance between different *n* phases<sup>14-16</sup>. The pressure induced contraction along the stacking axis shortens the *d* value, thereby enhancing exciton funneling from low-*n* (wide-bandgap, donor) domains to high-*n* (narrow-bandgap, acceptor) domains<sup>17</sup>. This reactivated energy cascade restores efficient gain buildup in the microscale films, accounting for the reemergence of ASE under moderate pressure and the pronounced threshold reduction observed at 0.41 GPa. When the pressure exceeds 0.41 GPa, the compression rate along the stacking axis slows considerably, leading to a competitive interplay between residual interlayer

contraction (i.e., the “two-step” structural compression process) and octahedral distortion, with the latter gradually becoming dominant<sup>18,19</sup>. The increasing structural disorder gives rise to enhanced carrier scattering and nonradiative recombination, leading to a gradual attenuation of emission intensity<sup>20,21</sup>. Consequently, the optical gain progressively deteriorates, and the ASE threshold increases continuously beyond 0.41 GPa, eventually quenching at higher pressures. Moreover, the low-frequency Raman modes progressively broaden and become less resolved, providing direct evidence of this pressure-induced structural disordering. These findings indicate that (PEA)<sub>2</sub>FA<sub>n-1</sub>Pb<sub>n</sub>Br<sub>3n+1</sub> thin films do not undergo a first-order structural phase transition under pressure; instead, they experience a transition from interlayer to intralayer two-step compression. This distinct sequence provides a fundamental physical understanding of how energy transfer reconstruction underlies the reactivation of ASE from a structural perspective.

The structural revolution of the (PEA)<sub>2</sub>FA<sub>n-1</sub>Pb<sub>n</sub>Br<sub>3n+1</sub> perovskite framework under compression was further validated through *in situ* high-pressure synchrotron XRD diffraction. To ensure optimal signal acquisition quality, PEA<sub>2</sub>FAPb<sub>2</sub>Br<sub>7</sub> ( $n = 2$ ) single crystals, synthesized via the solution temperature-lowering (STL) method (**Fig. S7** for the fresh single crystal’s XRD pattern), were employed for the *in situ* XRD characterizations. The  $n = 2$  composition was specifically chosen because it preserves the representative structural characteristics of quasi-2D perovskites. *In situ* high-pressure XRD analysis of the PEA<sub>2</sub>FAPb<sub>2</sub>Br<sub>7</sub> single crystals (**Fig. 3d**) further confirms the absence of any structural phase transition within the investigated pressure range from 0 to 3.41 GPa. **Fig. 3e** and **Fig. 3f** present the shifting rates of seven selected XRD diffraction peaks versus applied pressure, where all peaks exhibit two distinct pressure regimes with markedly different slopes. The steeper slope below 1.16 GPa corresponds to predominant interlayer compression, whereas the more gradual slope above this pressure indicates a shift toward dominant intralayer compression<sup>22</sup>. This transition occurs at 1.16 GPa, a value higher than the 0.41 GPa inflection point observed in the mixed-phase (PEA)<sub>2</sub>FA<sub>n-1</sub>Pb<sub>n</sub>Br<sub>3n+1</sub> thin film. This discrepancy arises because the low- $n$  phase possesses a larger compressive elastic regime compared to the high- $n$  phases,

resulting in a delayed inflection point relative to the mixed-phase thin film<sup>23</sup>. Nevertheless, these XRD results robustly corroborate the two-step structural compression behavior observed in the Raman spectra. Furthermore, the *in situ* high-pressure steady-state absorption spectra of the (PEA)<sub>2</sub>FA<sub>n-1</sub>Pb<sub>n</sub>Br<sub>3n+1</sub> thin film, as displayed in **Fig. 3g**, demonstrate that the absorption edge (**Fig. S9**) exhibits a shifting trend consistent with the structural compression. This correlation between optical behavior and microscopic structural evolution underscores the structure-property relationship involved in the pressure-induced activation and modulation of ASE within the (PEA)<sub>2</sub>FA<sub>n-1</sub>Pb<sub>n</sub>Br<sub>3n+1</sub> framework, providing solid evidence for structure-mediated FRET engineering via pressure modulation.

#### ***Fs-TA probing the ultrafast photophysical processes of ASE pressure-induced activation***

While the aforementioned discussion provides critical insights and a structural framework for understanding the structure-property relationships that govern pressure-induced ASE activation and optical gain modulation in microscale (PEA)<sub>2</sub>FA<sub>n-1</sub>Pb<sub>n</sub>Br<sub>3n+1</sub> thin films, the underlying photophysical processes and the specific role of pressure in mediating energy transfer remain less understood. *In situ* high-pressure femtosecond transient absorption (fs-TA) spectroscopy offers a powerful approach to probe these ultrafast dynamical processes. As shown in **Fig. 4a** and **4d**, the fs-TA spectra of (PEA)<sub>2</sub>FA<sub>n-1</sub>Pb<sub>n</sub>Br<sub>3n+1</sub> thin films at 0 GPa (ambient pressure) and 0.41 GPa both exhibit multiple ground-state bleaching (GSB) signals, confirming the coexistence of multiple perovskite phases. The absorption peaks are assigned to the  $n = 1$  phase (404 nm and 410 nm),  $n = 2$  phase (430 nm and 444 nm), and the higher- $n$  phases ( $n \geq 5$ , 520 nm and 530 nm), respectively. The TA spectra at different delay times derived from **Fig. 4a** and **4d** (plotted in **Fig. 4b** and **4c**), together with the representative GSB kinetics for different  $n$  phases (**Fig. 4e** and **4f**), clearly reveal the typical energy transfer pathway in (PEA)<sub>2</sub>FA<sub>n-1</sub>Pb<sub>n</sub>Br<sub>3n+1</sub> films: excitons are generated in the low- $n$  phases, leading to a rapid sequential rise in photobleaching at the absorption peaks of the  $n = 1$  and  $n = 2$  phases. Subsequently, within a delay time ( $\leq 1$  ps), charge carriers transfer from low- $n$  phases to  $n \geq 5$  phase, progressively intensifying the resonance signals

associated with the high- $n$  phases. This dynamic is indicative of a cascade energy transfer process originating from the low- $n$  phase, and ultimately populating the phases with  $n \geq 5^{24}$ .

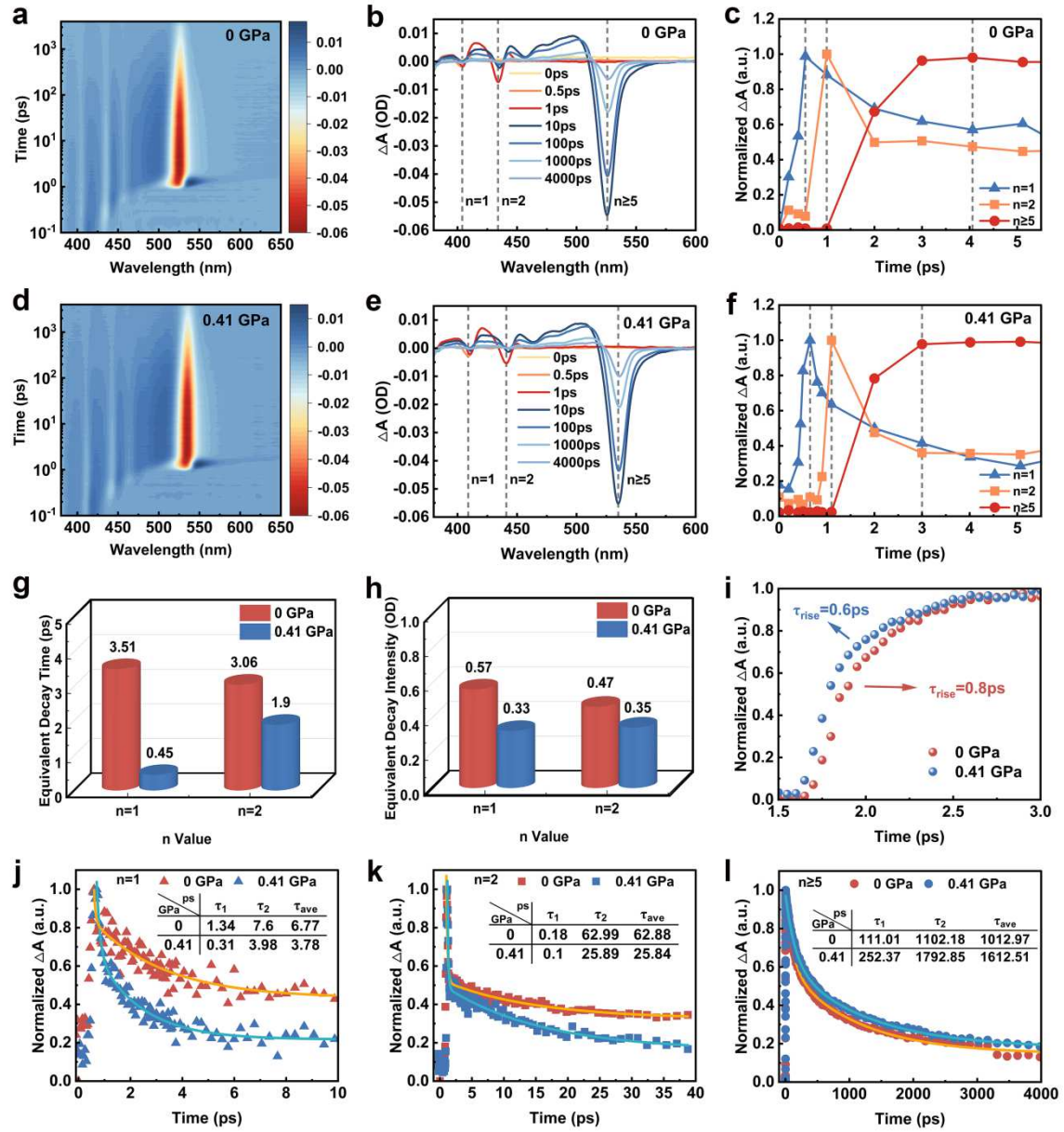


**Fig. 4 Pressure-dependent femtosecond transient-absorption (Fs-TA) dynamics of  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  thin films.** Fs-TA spectra color maps of the  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  thin films at **a** 0 GPa and **d** 0.41 GPa. TA spectra at different decay times at **b** 0 GPa and **e** 0.41 GPa. TA kinetics at different wavelengths at **c** 0 GPa and **f** 0.41 GPa. **g** Equivalent decay time of the  $n = 1$  and  $n = 2$  phases at different pressures. **h** Equivalent decay intensity of the  $n = 1$  and  $n =$

2 phases at different pressures. **i** Fast rise time of the  $n \geq 5$  phase at different pressures. Exciton lifetimes of the **j**  $n = 1$ , **k**  $n = 2$ , and **l**  $n \geq 5$  phases at different pressures.

To quantitatively evaluate the effect of pressure on the interphase energy transfer rate, we define an equivalent decay time as the time required for the normalized TA change ( $\Delta A$ ) of the GSB signal to decay to a value approaching stability. At ambient pressure, the equivalent decay times of the  $n = 1$  and  $n = 2$  phases in the  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  thin film are 3.51 ps and 3.06 ps, respectively. In contrast, at 0.41 GPa, the time required to reach the equivalent normalized intensity is significantly shortened to 0.45 ps and 1.90 ps, respectively (**Fig. 4g**), representing a decay time reduction of 87% and 38%. In addition to the accelerated decay kinetics, exciton accumulation in the low- $n$  phases under high pressure is also notably reduced compared to ambient conditions. Specifically, the residual normalized  $\Delta A$  values for the  $n = 1$  and  $n = 2$  phases decrease from 0.57 and 0.47 at ambient pressure to 0.33 and 0.35 at 0.41 GPa (**Fig. 4h**), respectively, indicating a further reduction in the number of accumulated excitons in the low- $n$  phases. Consistent with the accelerated signal decay in the low- $n$  phases under high pressure, the fast rise time (defined as the time required for the normalized  $\Delta A$  to rise from 0 to 1) of the GSB signal for the  $n \geq 5$  phases is shortened from 0.8 ps at 0 GPa to 0.6 ps at 0.41 GPa (**Fig. 4i**), representing a 25% reduction. These ultrafast interphase dynamics demonstrate that pressure significantly accelerates exciton decay in the low- $n$  phases and enhances the energy transfer efficiency toward the higher- $n$  phases. This mechanism is evidenced by the accelerated depletion of the low- $n$  phases and the faster exciton population buildup in the high- $n$  phases, with the most pronounced effect observed for the  $n = 1$  phase<sup>25</sup>.

The exciton lifetimes of the different  $n$ -value phases, which characterize the excited-state relaxation process and determine the duration of population inversion, can be extracted from the fs-TA kinetics fitting (**Figs. 4j-4l**). These kinetic traces are adequately fitted using a double-exponential function:

$$I(t) = I_0 + A_1 \exp\left(-\frac{t}{\tau_1}\right) + A_2 \exp\left(-\frac{t}{\tau_2}\right)$$

where  $I(t)$  represent the  $\Delta A$  of the GSB signal,  $I_0$  is the constant offset in the fitting function,  $A_1$  and  $A_2$  are the relative amplitudes of the two decay components, and  $\tau_1$  and  $\tau_2$  correspond to the fast and slow decay time constants, respectively. The amplitude-weighted average lifetime ( $\tau_{ave}$ ) is calculated by  $\tau_{ave} = (A_1\tau_1^2 + A_2\tau_2^2)/(A_1\tau_1 + A_2\tau_2)$ . The fs-TA kinetics of quasi-2D perovskites consistently exhibit two distinct decay components: a fast decay component associated with interphase exciton energy transfer, and a slow decay component ascribed to radiative and nonradiative exciton recombination<sup>26,27</sup>. At 0 GPa, owing to the incomplete cascade energy transfer, the  $n = 1$  and  $n = 2$  phases exhibit weak GSB signal amplitudes and prolonged relaxation processes, with average lifetimes of 6.77 ps and 62.88 ps, respectively<sup>28-30</sup>. In contrast, at 0.41 GPa, the low- $n$  phases show markedly accelerated decay dynamics, with average lifetimes of 3.98 ps ( $n = 1$ ) and 25.89 ps ( $n = 2$ ), both of which are significantly shorter than the corresponding values at ambient pressure. For the high- $n$  phase ( $n \geq 5$ ), the average exciton lifetime reaches 1612.51 ps at 0.41 GPa, which is 60% longer than the 1012.97 ps obtained at 0 GPa. This prolonged lifetime suggests that Auger recombination, a dominant nonradiative loss channel under high carrier density, is effectively suppressed at 0.41 GPa<sup>31-33</sup>. Ultimately, this pressure-induced synergistic modulation — specifically, the accelerated exciton depletion in low- $n$  phases for efficient energy injection, coupled with the prolonged exciton lifetime in high- $n$  phases for stable population inversion accumulation — provides the fundamental photophysical origin for the pressure-activated ASE behavior in quasi-2D perovskite thin films.

### Discussion

Combining the above pressure induced structure and photophysical results, the underlying mechanism of pressure-enhanced ASE activation can be elucidated, as illustrated in **Fig. 5a**. Upon the application of pressure, the  $(\text{PEA})_2\text{FA}_{n-1}\text{Pb}_n\text{Br}_{3n+1}$  perovskite films undergo preferential compression along the interlayer direction, owing to the substantially higher compressibility of the organic spacer layers. This leads to a rapid reduction of interlayer distance, thereby decreasing the spatial separation between quantum wells built from low- $n$  to high- $n$  phases. Given the strong  $d^{-4}$  dependence of FRET on donor-acceptor distance in quasi-2D system, modest interlayer contraction

markedly accelerates exciton transfer from the low- $n$  to high- $n$  phases. Fs-TA directly validate this mechanism, whereby pressure significantly accelerates interphase energy funneling from low- $n$  to high- $n$  domains while prolonging the exciton lifetime of high- $n$  phases. The resulting synergy enables efficient carrier accumulation and robust population inversion in the emissive phase. It is precisely under this synergistic enhancement mechanism that ASE re-emerges at 0.14 GPa and reaches its lowest threshold at 0.41 GPa (**Fig. 5b**). These mechanistic insights demonstrate that mechanical compression can activate strong optical gain in a microscale perovskite film, which cannot sustain on its own in ambient pressure. However, a practical question raises: how can such compression be integrated into a real device? Given the relatively low optimal pressure range demonstrated here, a DAC is not strictly necessary. For instance, laminating the perovskite film onto a thin polymer sheet and bending it with a controlled radius would effectively compress the crystal layers. With proper design, such a structure could be built into chip-scale coherent light sources, turning the “blessing of compression” into a viable technological tool. More generally, any integrated platform that allows local compression, such as piezoelectric actuators or prestrained membranes, could dynamically switch on or tune the ASE from a microscale perovskite domain. Building on these concepts, our work points toward engineered micro/nanoscale gain media where the emission properties are adjusted on demand, without changing the material composition or cavity geometry. This engineering paradigm facilitates versatile applications, including low-pressure-responsive fluorescent probes and on-chip laser sources, and ultimately addresses the critical bottlenecks in the miniaturization and practical deployment of quasi-2D perovskite thin films.

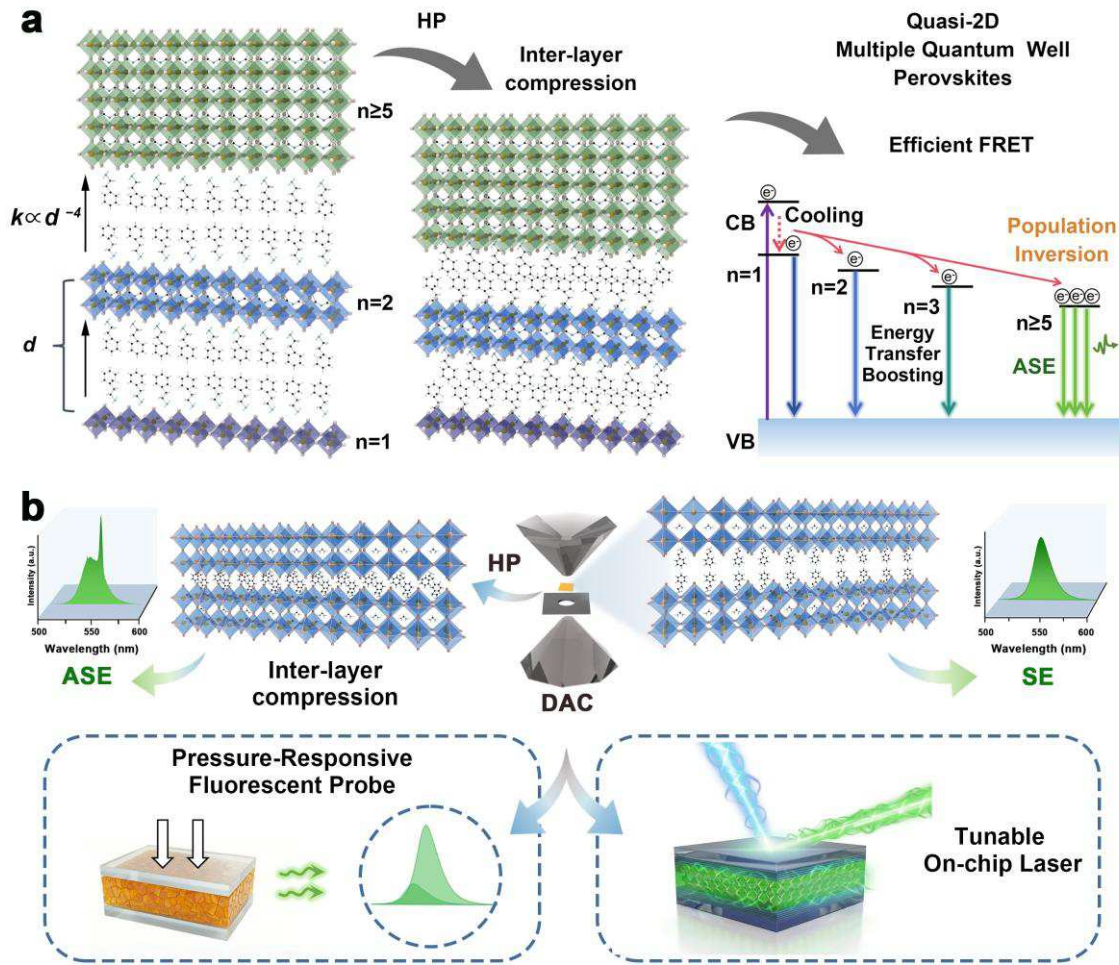


**Fig. 5 Schematic illustration of energy-cascade optimization and pressure-activated ASE in microscale quasi-2D perovskite films for multi-purpose applications. a** The underlying mechanism of energy cascade optimization, and **b** the pressure-induced ASE activation in microscale quasi-2D perovskite films and its application for multiple purposes.

### Conclusion

In summary, this work demonstrates that high-pressure engineering works as a dual-function strategy: it not only activates ASE in microscale quasi-2D perovskites but also allows for its precise modulation. Applying an optimal pressure switch on ASE in otherwise inactive microscale films by accelerating the energy cascade. Multiple *in situ* probes reveal the structure-property relationship linking the interlayer-to-intralayer compression transition to the ASE modulation effect. Mechanistically, fs-TA spectroscopy directly resolves a faster exciton transfer from the low- $n$  to high- $n$  phases and a 60% longer excited-state lifetime in the high- $n$  phases, both of which facilitate

the buildup of population inversion and lower the ASE threshold. Importantly, the ASE threshold can be tuned continuously with pressure, providing a clean and reversible control mechanism. This work clarifies the photophysical mechanism behind pressure-controlled energy transfer and establishes mechanical compression as a practical tool for the development of compact quasi-2D perovskite gain media.

#### ***Data Availability Statement***

Data available with the paper or supplementary information.

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#### ***Conflict of Interest***

The authors declare no conflict of interest.

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