Journal of Materials Chemistry C



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Cite this: J. Mater. Chem. C, 2023, 11, 11730

Received 24th May 2023, Accepted 6th August 2023 DOI: 10.1039/d3tc01797d

rsc.li/materials-c

Introduction

Semiconductor lasers have been widely used in many optoelectrical areas, such as optical communication and consumer optoelectrical devices, due to their efficiency, long life, and compact advantages. Traditional semiconductor lasers use inorganic semiconductor materials as gain media, but involve complicated processes and incur high production costs. By contrast, some new gain media, including organic semiconductors,¹ colloidal quantum dots,² and halide perovskite semiconductors, have been developing rapidly in recent years to meet the developing needs for miniaturization, portability, and highly integrated laser devices. These materials can be prepared under vacuum thermal deposition or solution processing. Among them, organic semiconductors have great potential for obtaining functional materials by chemical modification of the molecular structure. Since Rhodamine 6G doped in polymethyl methacrylate (PMMA) was first used as an organic gain

Solvent atmosphere-assisted crystallization of perovskites for room-temperature continuouswave amplified spontaneous emission[†]

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Low-threshold, solution-processable quasi-two-dimensional (quasi-2D) perovskites hold great significance in the development of compact, solution-processable lasers. However, defects such as pinholes, caused by the rapid crystallization process and non-uniform distribution of mixed phases, contribute to increased optical losses and decreased energy transfer. In this study, we developed a simple strategy involving DMSO atmosphere-assisted crystallization for preparing quasi-2D perovskite films. This approach allows effectively controlling the surface crystallization process, resulting in a smooth surface morphology and improved phase distribution. Consequently, the non-radiative recombination rate decreases, while the fluorescence quantum yield increases. As a result, using the $PEA_2FA_{n-1}Pb_nBr_{3n+1}$ (n = 6) film with DMSO atmosphere treatment, we achieved a low pulse optically pumped amplified spontaneous emission (ASE) threshold of 12.9 μ J cm⁻², which was 3.6 times lower than the case without atmosphere treatment. Furthermore, by incorporating a microcavity structure to enhance optical feedback, we successfully achieved continuouswave (CW) optically pumped ASE at room temperature with an extremely low threshold of 3.8 W cm⁻².

> medium³ in 1967, many organic gain media with semiconductor properties, such as Alq3:DCM¹ and poly(*p*-phenylenevinylene) (PPV),⁴ have been reported. At present, the research into laser technology is mainly focused on the optimization of the resonant cavity⁵ and the molecular design of gain media.⁶ Colloidal quantum dot lasers offer good solution processability, temperature stability, and a wide range of tunability in terms of the emission wavelength. By controlling the size and composition of the quantum dots, the laser output can be tailored to cover a broad spectral range, from ultraviolet to infrared, making them versatile for many diverse applications. Great progress has been made in colloidal quantum dot synthesis, device architectures, and lasing properties.² Perovskites are new kinds of potential gain materials, which are wavelength tunable, solution-processable, and low cost. Owing to the characteristics of their high fluorescence quantum efficiency, high light absorption coefficient, and long carrier diffusion length, they have shown excellent laser performance and great potential in the fields of laser and other optoelectronic devices.7-13

> Compared to traditional three-dimensional (3D) perovskites, quasi-two-dimensional (quasi-2D) perovskites possess spontaneous quantum-well structures that facilitate rapid energy transfer from the low-*n* phase to the high-*n* phase. This characteristic effectively enhances the utilization of excitons and population inversion for achieving lasing, indicating their significant potential as laser gain materials.^{14–18} The quality of quasi-2D perovskite

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[†] Electronic supplementary information (ESI) available. See DOI: https://doi.org/

^{10.1039/}d3tc01797d

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films, including their smoothness, density, and thickness, plays a crucial role in the output of amplified spontaneous emission (ASE).¹⁹ The smoothness and density of the film partly determine the optical gain length, while the film thickness affects waveguide construction. Vacuum thermal evaporation offers a precise control over the thickness of perovskite films, making it an effective method for obtaining more-uniform thin films.^{20,21} The combination of film surface modification and vacuum thermal evaporation has yielded positive results in terms of optimizing the quality of perovskite crystals.²² However, vacuum thermal evaporation, as a method for preparing thin films, is expensive and complex. Additionally, controlling the evaporation of perovskites containing methylammonium is challenging due to the relatively high vapor pressure of the source materials.^{23,24}

Currently, the solution method remains the prevalent approach for preparing perovskite films. However, due to the rapid evaporation of the solvent, the surface of the perovskite film often ends up with holes in it and precipitations of lead halide.²⁵ Regulation of the perovskite crystal quality can be achieved through various means, such as controlling the long-chain/short-chain organic cations,²⁶⁻³⁰ selecting appropriate precursor solutions,^{19,31} surface modification,³² incorporating additives,^{12,33–39} choosing the proper substrates,⁴⁰⁻⁴² and implementing recrystallization techniques.43-45 Most of the aforementioned processes involve introducing additional ions into the perovskite precursor solution or the substrate interface to induce crystallization, or necessitate a reprocessing of the crystallized film. This is particularly true for solvent recrystallization, which can take from 1 h to 2 weeks.⁴⁴ The relationship between crystallization and the surrounding atmosphere is often overlooked. Typically, perovskites begin to crystallize as a result of solvent evaporation, causing the precursor solution to become oversaturated.³⁰ Jin's team recognized that a partial pressure of DMSO vapor in the atmosphere plays an important role in determining the rate of solvent evaporation. This, in turn, has a significant impact on the duration and degree of supersaturation during the wet film stage in the solution processing of perovskite materials.⁴⁶ The atmosphere has a significant impact on the nucleation and growth of perovskite crystals. In the case of guasi-2D perovskites, rapid crystallization is unfavorable for controlling the composition of mixed phases and can affect energy transfer between different quantum-well components. Therefore, rational control of the crystallization rate holds great importance in the preparation of high-quality perovskite optical gain media.

Here, we present an effective processing method for enhancing the crystallization of quasi-2D perovskite films by utilizing a DMSO atmosphere during the film-formation process. This approach can lead to significant improvements in the surface morphology, optical properties, and lasing performance of perovskite films. When compared to perovskite films prepared without DMSO atmosphere assistance (w/o DMSO), the films treated with DMSO atmosphere (with DMSO) exhibited a remarkable reduction in the pulse optically pumped ASE threshold to 12.9 μ J cm⁻². Furthermore, by incorporating an optimized optical microcavity structure, an extremely low ASE threshold of 3.8 W cm⁻² was achieved under continuous-wave (CW) optical pumping at room temperature.

Results and discussion

In this study, we employed a DMSO atmosphere to deliberately slow down solvent evaporation during the spin-coating process of the quasi-2D perovskite $PEA_2FA_{n-1}Pb_nBr_{3n+1}$ (n = 6, P2F6). The Lewis basic DMSO formed a coordination complex with Pb^{2+} , resulting in the formation of $[PbBr_6]^{4-}$ intermediate complexes. These intermediate complexes gradually released Pb^{2+} ions during the crystallization process.^{44,47} As a result, a perovskite film with improved surface smoothness, enhanced orientation, enhanced crystallinity, and larger grain sizes was obtained. We conducted characterizations of the films to examine the potential impact of decelerating the crystallization process on the film thickness.

Fig. 1a and d display the cross-sectional scanning electron microscopy (SEM) images of the quasi-2D perovskite P2F6 films with and without DMSO atmosphere treatment, while their top surface morphology is depicted in Fig. 1b and e. The DMSOtreated P2F6 film exhibited a smoother structure with fewer pinholes (Fig. S1, ESI[†]). The film thickness of the P2F6 film induced by the DMSO atmosphere-assisted crystallization was approximately 100 nm, which was comparable to the film without DMSO treatment. Fig. 1c and f present the atomic force microscopy (AFM) images of these films, revealing that the P2F6 film undergoing DMSO atmosphere crystallization had a smoother surface with fewer pinholes. The root-meansquare (RMS) value of the P2F6 films decreased from 6.02 nm to 1.75 nm before and after the atmosphere treatment. Hence, it can be concluded that an appropriate amount of DMSO atmosphere can significantly enhance the crystallization quality of quasi-2D perovskite P2F6 films. The crystallization of perovskites was induced at the interface between air and the film,⁴⁴ resulting in smoother films and effectively reducing scattering losses.

A schematic diagram illustrating the quasi-2D perovskite crystal structure is presented in Fig. 2a. As the solvent evaporated, nucleation was initiated within the perovskite, ultimately resulting in the formation of a thin film with multiple mixed phase distributions. Fig. 2b shows the X-ray diffraction (XRD) patterns of P2F6 films with and w/o DMSO atmosphere treatment. Two dominant diffraction peaks were observed at 14.8° and 29.8°, corresponding to the [100] and [200] crystal planes, respectively, confirming the horizontal alignment of perovskite crystals.^{8,48} The absence of any deviation from the diffraction peaks indicated that the P2F6 film treated under the DMSO atmosphere retained the original crystal structure of the untreated film. Compared to the P2F6 film without DMSO atmosphere treatment, the DMSO atmosphere-treated P2F6 film exhibited a narrowing of the full width at half maximum (FWHM) for the 14.8° diffraction peak from 0.8° to 0.7° , and for the 29.8° diffraction peak from 3.3° to 2.6°. According to Scherrer's formula $(D = K\lambda/(\beta \cos \theta))$, where D denotes the average crystallite size, K is a constant of 0.9, λ is the X-ray wavelength of 0.154 nm for Cu K α , β represents the FWHM of the diffraction peak in radians, and θ indicates the Bragg diffraction angle), the reduced FWHM in the DMSOtreated film suggests an increase in the grain size of the film.

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Fig. 1 Cross-sectional SEM images of P2F6 films (a) w/o and (d) with DMSO atmosphere treatment. SEM images of P2F6 films (b) w/o and (e) with DMSO atmosphere treatment. AFM images of P2F6 films (c) w/o and (f) with DMSO atmosphere treatment.

The aforementioned narrowed diffraction peaks were accompanied by a noticeable increase in intensity, indicating that the P2F6 film treated under the DMSO atmosphere had a larger grain size and improved crystallinity.

Fig. 2c shows the UV-vis absorption spectra and photoluminescence (PL) spectra of the P2F6 films w/o and with DMSO treatment. The untreated pristine P2F6 film showed a bandedge absorption peak at 524 nm, corresponding to phases with

 $n \ge 5$. By contrast, the P2F6 film treated with DMSO showed an additional absorption peak at 439 nm, corresponding to the n = 2 phase. Moreover, two less pronounced absorption peaks, corresponding to n = 3 and n = 4, at 473 nm and 494 nm,⁴⁸ respectively, could be observed in the first-order differential absorption spectrum (Fig. 2c inset). These differences in the absorption spectra suggest that, under the influence of the DMSO atmosphere, the perovskite film undergoes a slow



Fig. 2 (a) Schematic diagram of the quasi-2D perovskite crystal structure. (b) XRD, (c) absorption and PL spectra, (d) PLQY, and (e) fluorescence lifetime of the P2F6 films with and w/o DMSO treatment. Inset in (c) shows the first-order differential absorption spectrum of the P2F6 film with DMSO treatment.

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top-to-bottom crystallization process, resulting in a higher proportion of smaller *n*-phase structures with the inclusion of n = 2, 3, and 4. This leads to a more reasonable gradient in the distribution of perovskite mixed phases. Under the same experimental conditions, the PL intensity of the P2F6 film with DMSO treatment was approximately 1.47 times stronger than that of the untreated film, attributed to the fewer defects and suppressed non-radiative recombination rates. Furthermore, the PL quantum yield (PLQY) of the P2F6 films increased from the original 23% to 28% (Fig. 2d), and the fluorescence lifetime (τ_{avg}) increased from 47.09 ns to 65.66 ns (Fig. 2e) after the optimized crystallization process. The non-radiative recombination rate of the P2F6 films can be calculated using the following equations, and where the relationship between τ_{avg} and PLQY is given too as follows:

$$\tau_{\rm avg} = \frac{1}{K_{\rm r} + K_{\rm nr}} \tag{1}$$

$$PLQY = \frac{K_{\rm r}}{K_{\rm r} + K_{\rm nr}}$$
(2)

$$K_{\rm nr} = \frac{1 - PLQY}{\tau_{\rm avg}} \tag{3}$$

where K_r is the radiative recombination rate, and K_{nr} is the non-radiative recombination rate.

Obviously, the K_{nr} of a film is inversely proportional to its PLQY and τ_{avg} . Therefore, a higher PLQY and longer fluorescence lifetime of the P2F6 film signifies a lower non-radiative recombination rate. The calculated K_{nr} of the P2F6 film without

DMSO treatment was $1.6 \times 10^7 \text{ s}^{-1}$, while that of the treated film was 1.1×10^7 s⁻¹, indicating a decrease of about 31%. A longer fluorescence lifetime indicates a decrease in the sum of $K_{\rm r}$ and $K_{\rm nr}$. Additionally, the calculated ratio of $K_{\rm r}$ to $K_{\rm nr}$ increased by 30%, indicating that the non-radiative recombination of P2F6 films can be effectively reduced by employing the DMSO treatment method. To verify the applicability of the method, the DMSO atmosphere treatment was applied to a series of quasi-2D perovskite materials with different n values, including P2F7, P2F8, and P2F9. For each perovskite material, the enhancement of the PL emission intensity (Fig. S2-S4, ESI[†]) and the general extension of the fluorescence lifetime (Fig. S5-S7, ESI⁺) observed under the DMSO atmosphere treatment were not accidental occurrences (Table S1, ESI⁺). The increase in PL intensities could be attributed to the effective suppression of non-radiative recombination defects through DMSO-assisted crystallization (Table S1, ESI⁺). Moreover, the rational distribution of *n*-phases facilitates an efficient energy funnel, thereby improving the energy-transfer process and enhancing the radiative recombination rate of quasi-2D perovskite films.

To investigate the difference in energy transfer between perovskite films with and w/o DMSO treatment, we conducted comparative tests to obtain ultrafast transient absorption (TA) spectra at high pump intensities. Fig. 3a and d show the 2D pseudo-color contour mapping of the TA spectra for the P2F6 films w/o and with DMSO treatment, respectively, both displaying two distinct negative signals attributed to ground-state bleaching (GSB). Notably, in the case of the DMSO-treated film, two additional GSB signals appeared at 473 and 494 nm. This observation confirmed that the phase distribution of quasi-2D perovskites could be controlled by slowing down the crystallization using the



Fig. 3 2D pseudo-color contour mapping of the TA spectra of the P2F6 films (a) w/o DMSO and (d) with DMSO treatment. TA spectra of the P2F6 films (b) w/o DMSO and (e) with DMSO at selected time scales. TA kinetics probe of the P2F6 films (c) w/o DMSO and (f) with DMSO at different wavelengths, corresponding to different *n*-phases.

DMSO atmosphere treatment. Furthermore, Fig. 3b and e illustrate the extracted P2F6 TA spectra at different delay times, where the GSB signals exhibited a good correspondence with the steady absorption spectra shown in Fig. 2c.

Fig. S8 (ESI[†]) shows that the GSB signal of the small *n*-phase gradually decreased within the interval of 1 ps to 1.8 ps, while the GSB signal of the large n-phase gradually increased. This phenomenon suggests the presence of an energy funnel structure in the quasi-2D perovskite that facilitates energy transfer from the small *n*-phase to the large *n*-phase. Representative GSB kinetics corresponding to different *n*-phases of the P2F6 films are illustrated in Fig. 3c and 3f. The GSB kinetic fitting results are summarized in Table S2 (ESI⁺).⁴⁹⁻⁵¹ For the P2F6 film w/o DMSO treatment, the n = 2 phase exhibited a relaxation time of approximately 0.27 ps, followed by a slow decay of ~ 15 ps, indicating an incomplete energy-transfer process.⁵²⁻⁵⁴ Additionally, a fast GSB decay within 1-10 ps was observed in the $n \ge 5$ phase in Fig. 3c, which was not present in the perovskite film treated with DMSO. This difference could be attributed to the improved crystallization and higher quality crystallinity of the optimized film, which reduces the impact of defect traps.⁵⁵ Fig. 3f presents the kinetic curve at the selected time for the P2F6 film with DMSO treatment, with relaxation times of approximately 0.36, 0.43, and 0.32 ps for the n = 2, n = 3, and n = 4 phases, respectively. It is worth noting that the rise time for the n = 2 phase was significantly reduced by about 1.7 times when using the DMSO treatment method compared to the case without DMSO treatment (Table S2, ESI⁺). Furthermore, the rise time for the $n \ge 5$ phase was shortened from ~ 0.95 ps to ~ 0.54 ps, representing a nearly 43% faster rise time after employing the DMSO treatment method. Upon optical excitation, excitons are initially generated at a high energy level. These excitons tend to populate the n = 2 phase first. The fast rising time observed in the n = 2 phase indicates that the DMSO treatment expedited the relaxation process from the higher energy level to the lowest excited state of the n = 2phase. The presence of a graded band structure among the mixed *n*-phases enables efficient energy transfer from the n = 2 to the $n \ge 5$ phase. The shortened rise time in the $n \ge 5$ phase suggests an accelerated exciton accumulation process and indicates a faster energy transfer occurred from the high-bandgap n = 2phase to the lowest bandgap $n \ge 5$ phase. Consequently, the DMSO treatment method facilitated a more favorable distribution of *n* values, leading to an improved graded energy-band distribution. Thus, optimization of the energy-transfer pathway was achieved. The above results provide evidence that by employing the DMSO treatment method to slow down the crystallization process, it is possible to obtain a perovskite film with an accelerated energy-transfer process and improved crystal quality.

Furthermore, we investigated the ASE performance of the P2F6 films. The quasi-2D perovskite films were pumped using a 355 nm nanosecond pulsed laser with a stripe size of 200 μ m × 4 mm. The ASE properties of the P2F6 film without DMSO treatment are shown in Fig. 4a and b. At low pump fluences, broad spontaneous emission spectra centered around 535 nm could be observed, with a FWHM of 39.7 nm. When the pump

fluence exceeded 40 μ J cm⁻², the emission intensity started to increase nonlinearly, accompanied by a gradual decrease in the FWHM. This behavior is a characteristic of the ASE phenomenon, with a pump threshold ($P_{\rm th}$) of about 46.7 µJ cm⁻². Above the threshold, the narrowest ASE spectrum showed a peak at around 545 nm and a minimum FWHM of 3.6 nm. For comparison, Fig. 4c and d demonstrate that the P2F6 film treated with DMSOatmosphere-assisted crystallization exhibited a wider PL spectrum, with an emission peak at 532 nm below the $P_{\rm th}$. Notably, a significantly reduced ASE threshold of 12.9 µJ cm⁻² was also observed, accompanied by an ASE emission peak at 547 nm. The gain-induced linewidth was reduced significantly from 38.3 nm to 1.6 nm in the DMSO-atmosphere-treated perovskite film. This film exhibited improved photophysical properties, including a lower $P_{\rm th}$, and a narrower ASE FWHM. To further assess the optical gain characteristics, the variable stripe length (VSL) method was employed to measure the net gain of the perovskite films. Fig. S9 (ESI⁺) depicts the evolution of the PL intensity as the stripe length of the films increased. The effective optical gain (g) is determined by the following equation relating the output intensity (I) and the length of the excited strip (L), ^{56,57}

$$I = I_0 (e^{gL} - 1)/g$$
 (4)

where the term I_0 represents the rate of spontaneous emission per unit volume. The net gain coefficient for the pristine perovskite film was thus estimated to be ~86 cm⁻¹, whereas this value increased to $\sim 205 \text{ cm}^{-1}$ for the film treated with DMSO, indicating an enhancement of 2.38 times compared to the pristine film. In comparison to the ASE threshold of the films without DMSO treatment, the series of perovskite films treated with DMSO showed a significant reduction (Fig. S10-S12 and Table S1, ESI[†]). For the P2F9 film, the ASE threshold decreased from 31.1 μ J cm⁻² to 9.8 μ J cm⁻² before and after DMSO treatment (Fig. S12, ESI[†]). The observed increase in fluorescence (Fig. 2c and Fig. S5-S7, ESI⁺) and the extended fluorescence lifetime (Table S1, ESI⁺) were in line with a reduction in the nonradiative recombination rate and the ASE threshold. These findings suggest that the utilization of DMSO atmosphere treatment would be universally effective for the perovskite film series. This treatment decelerates the crystallization process, decreases defect density, and consequently enhances the properties of spontaneous emission and stimulated emission. As a result, the ASE threshold is significantly reduced.

Given the low ASE threshold observed in the 2D perovskites with DMSO treatment under pulsed optical pumping, they possess the potential to serve as continuous-wave (CW) optically pumped gain media. We were unable to observe ASE in perovskite films under CW optical pumping. We attribute this absence of ASE to two primary factors: First, the required power threshold for ASE was quite high, making it difficult to find a suitable highpower CW laser for pumping; second, when the perovskite film samples were optically pumped in the presence of air, their fluorescence intensity rapidly decreased. Fig. S13 (ESI[‡]) shows the decline in intensity under constant optical pumping power, suggesting that this decline may be faster with a higher pumping power. Continuous optical pumping can cause photochemical



Fig. 4 Emission spectra of P2F6 films (a) without and (c) with DMSO treatment at different pump fluences. Output intensity and FWHM of the P2F6 films (b) without and (d) with DMSO treatment as a function of the pump fluence.

reactions due to the oxygen in the air, leading to material degradation. Additionally, the heat generated by CW pumping is not effectively dissipated through the substrate, causing heat accumulation and the non-radiative recombination of perovskite excitons. Using a microcavity structure, in which the perovskite film is sandwiched between two distributed Bragg reflectors (DBRs), could serve as an effective means of shielding the film from the influences of oxygen and water molecules. Moreover, the microcavity can enhance the radiation transition rate of the perovskite through the Purcell effect, significantly reducing the ASE threshold. By designing a suitable planar microcavity structure, we successfully achieved CW optically pumped ASE in quasi-2D perovskites at room temperature. The P2F6 film treated with DMSO atmosphere served as the intracavity gain medium.

Fig. 5a presents a schematic diagram of the designed microcavity structure. The device comprised a bottom DBR, a 150 nm-thick layer of P2F6 film treated with DMSO, and a top DBR. The reflectance spectra of the top DBR, bottom DBR, and the monolithic microcavity device are displayed in Fig. 5b. Notably, a strong cavity mode appeared at 559 nm (Fig. 5b). This cavity mode enabled the effective feedback of light with resonant wavelength within the microcavity, which was crucial

for achieving effective light gain in the perovskite layer. A CW laser with a wavelength of 405 nm was employed to excite the microcavity sample, with an excitation spot diameter of approximately 200 µm. It is important to note that when using a pulsed laser as the pump light source, the short pulse duration and long interval time between pulses allow the device sufficient time to dissipate residual heat to the substrate. However, in the case of CW laser pumping, heat accumulation becomes more significant, leading to the non-radiative recombination of perovskite excitons and an increase in the threshold. By leveraging the quantum electrodynamics effect, a microcavity structure effectively reduces the device loss and threshold, thereby facilitating the achievement of continuous optical pumping gain at room temperature. Fig. 5c and d show the emission properties of the microcavity under CW optical pumping. An emission peak was observed at 560 nm, resulting from the cavity mode induced by the modulation of the photon state density. At lower pump intensities, the emission spectrum showed a FWHM of 3.3 nm. As the pump intensity gradually increased, the output intensity showed a nonlinear rise, and the FWHM narrowed further to approximately 2 nm, indicating the occurrence of the ASE phenomenon. The threshold for CW optically pumped ASE was about 3.8 W cm⁻².



Fig. 5 (a) Schematic diagram of the microcavity structure. (b) Reflectance spectra of the microcavity and DBRs. (c) Emission spectra of the microcavity at different CW optical pump intensities. (d) Dependence of the output intensity and FWHM of the microcavity on the CW pump intensity.

Conclusions

In summary, we propose a facile atmosphere-assisted crystallization method for the preparation of quasi-2D perovskite films with a larger grain size and improved crystallinity. Using P2F6 as a representative material, we observed a 2.4 times decrease in RMS, a 31% reduction in the non-radiative recombination rate, a 22% increase in PLQY, and a significantly faster energy-transfer process with the DMSO atmosphere treatment. In consequence, the ASE threshold of the P2F6 film was lowered by 72%. By incorporating a high Q microcavity structure, we successfully achieved continuouswave optically pumped ASE at room temperature with a remarkably low threshold of 3.8 W cm⁻². These findings support the proposed method as a promising strategy for fabricating highquality perovskite thin films suitable for low-threshold perovskite lasers, thereby advancing the research and development of electrically pumped perovskite laser devices.

Experimental

Materials

Lead bromide (PbBr₂), phenylethylammonium bromide (PEABr), and formamidinium bromide (FABr) were purchased from Xi'an Polymer Light Technology Corp. Zinc sulfide, yttrium fluoride, silicon dioxide, and titanium oxide were purchased from Zhong Nuo Advanced Material (Beijing) Technology Co., Ltd. All materials were used directly without further purification.

Preparation and characterization of the perovskite films

The powders of PEABr, FABr, and PbBr₂ (molar ratio of 1:3:3) were added into anhydrous DMF solvent and stirred at 50 °C for 4 h. A transparent perovskite precursor solution with a concentration of 0.4 mM was obtained. Next, 600 µL DMSO solution was pre-sprayed in to an almost airtight spinner. The mixtures were then spin-coated on the glass at 4500 RPM for 30 s. During spin-coating, 0.25 mL of absolute ether was dropped onto the perovskite precursor layer at the fifth second. The substrates were baked on a hotplate at 120 °C for 10 min. The morphology of the films was characterized using an Hitachi 4800 scanning electron microscope. The surface roughness was analyzed with a Shimadzu SPA-9700 atomic force microscope. The absorption spectra of the films were measured with a Shimadzu UV-3101PC spectrophotometer. The PL spectra of the films were measured using an Hitachi F-7000 fluorescence spectrometer. The timeresolved PL spectra of the films were obtained with an Edinburgh FLS920 fluorescence spectrometer, on which fluorescent quantum yield measurements were performed with a calibrated integrating sphere. The XRD patterns of the perovskite films

were obtained using a Rigaku SmartLab X-ray diffractometer. The reflectance spectra were measured using a UV-visible-nearinfrared spectrophotometer (Lambda 1050, PerkinElmer). For the CW optical pumping microcavity experiments, the samples were pumped using a 405 nm CW laser. The laser light was focused and incident on the sample from the substrate side at a 45° angle relative to the microcavity normal. The emission signal from the sample was collected from the top DBR side, in the normal direction of the microcavity. Pulse optical pumping ASE measurements were performed using a CryLas GmbH Nd:YAG laser at 355 nm with a pulse width of 1 ns and a repetition rate of 50 Hz. The stripe pumping beam was focused by a cylindrical lens and the pump fluence was tuned by using calibrated neutral density filters. TA spectra experiments were carried out on a Newport TAS-1 transient absorption spectrometer. All the measurements were performed at room temperature under ambient conditions.

Device fabrication and characterization

The optimized microcavity device had a structure comprising glass/bottom DBR/P2F6(150 nm)/top DBR. The bottom DBR consisted of 15.5 pairs of periodic TiO₂/SiO₂ layers with a Bragg wavelength of 530 nm. The TiO₂ and SiO₂ layers were deposited by electron beam evaporation at a substrate temperature of 200 °C in an oxygen pressure of 2 \times 10⁻² Pa with deposition rates of 0.4 and 0.35 nm s⁻¹, respectively. The precursor solution with a concentration of 0.6 mmol mL⁻¹ was spincoated on the bottom DBR at 4500 RPM for 30 s. During spincoating, 0.25 mL of absolute ether was dropped onto the perovskite precursor layer. The substrates were baked on a hotplate at 120 °C for 10 min. The top DBR consisted of 8.5 pairs of periodic ZnS/YF_3 with a Bragg wavelength of 515 nm. The ZnS and YF₃ layers were deposited in a vacuum at 2 \times 10⁻³ Pa by electron beam evaporation with deposition rates of 0.3 and 0.35 nm s^{-1} , respectively.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

This work was supported by the National Science Foundation of China No. 62035013, 62175235, 51973208, 61975256, 52211530094, and the Jilin Province Science and Technology Research Project (20220201091GX, 20220201064GX), and the Perovskite Thin-Film Innovation Technology Centre (Project number: YZCXPT2022104).

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