

Analysis of Low-Threshold 1D Perovskite Photonic Crystal Laser

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Abstract. By using the bandgap effect and light localization effect, photonic crystal lasers can decrease the optical mode volume and lower the laser threshold, so satisfying the requirements for device shrinking and photonic integration. Given their wide range of potential applications in fields including light-emitting diodes and sensors, they are attracting considerable interest. The laser threshold is the minimum energy density required to cause laser oscillation and is a crucial specification for the practical application of laser devices. Perovskite materials, equipped with their exceptional optical gain characteristics, are well-suited for the development of low-threshold lasers. By combining them with photonic crystal lasers, it is possible to realize low-threshold or even threshold-free pumped lasers. This paper categorizes photonic crystal lasers according to their photonic crystal structures and examines the advancements in research on perovskite photonic crystal lasers in attaining low minimum thresholds. This work investigates the prospective capabilities of perovskite photonic crystal lasers, with the objective of achieving laser output that is low-threshold or near-zero-threshold, high-power, and of high quality.

Keywords: Photonic crystal lasers; Low-threshold lasers; Perovskite materials.

1. Introduction

In recent decades, semiconductor lasers have played an essential role in various applications, including telecommunications, medical diagnostics, and optical data storage. One of the critical parameters that affect the performance of semiconductor lasers is the excitation threshold, which refers to the minimum energy required to initiate laser emission. Reducing this threshold has become a major focus of research, as lower thresholds can lead to more energy-efficient devices, enhanced performance, and the potential for novel applications in integrated photonics and miniaturized systems.

In recent years, hybrid perovskite materials, particularly methylammonium lead iodide ($\text{CH}_3\text{NH}_3\text{PbI}_3$, MAPbI_3), have gained significant attention due to their immense potential in photovoltaic applications [1]. These materials possess advantages such as long carrier lifetimes and diffusion lengths, high internal quantum efficiency, and the ability to easily tune their wavelength through chemical composition adjustments, making them ideal candidates for laser gain media capable of achieving low-threshold lasing. Since 2014, after Xing et al. from Nanyang Technological University in Singapore reported low-threshold, stable amplified spontaneous emission in perovskite films at room temperature, research on perovskite lasers based on various morphologies, such as nanowires and nanosheets, has surged [2]. Combining perovskite materials with optimized laser structures could further reduce the laser threshold.

This paper explores the fundamental concepts of laser excitation thresholds, discusses recent advancements in reducing the threshold in semiconductor lasers, and examines innovative techniques, such as photonic crystal integration, that offer promising avenues for future development.

2. Basic of threshold of semiconductor lasers

2.1. Stimulated Absorption, Spontaneous Emission, and Stimulated Emission

When light interacts with a gain medium, three phenomena can occur: stimulated absorption, spontaneous emission, and stimulated emission. The amplification of light depends on whether the

process of stimulated emission predominates [3]. The physical processes in laser operation includes stimulated absorption, population inversion and increased spontaneous radiation as shown in Fig.1.

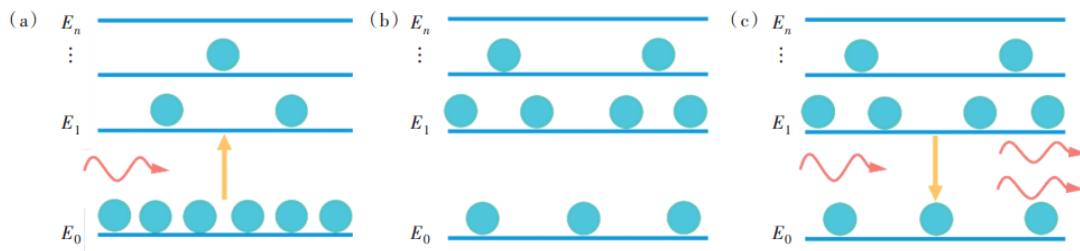


Fig 1. Physical Processes in Laser Operation

For most materials, electrons have two energy states: E_0 as the ground state and E where $n=1, 2, 3, \dots$, as the excited state. At room temperature, the number of electrons in the ground state is greater than those in the excited state. When a photon with energy $h\nu_0 = E_{Sn} - E_{S0}$, where h is Planck's constant, $h\nu_0$ represents the energy of a photon, E_{Sn} represents the excited state energy, and E_{S0} represents the ground state energy, enters an atomic system, the electron in the ground state S_0 absorbs the photon's energy and transitions to the excited state E_n . This transition process is called stimulated absorption as shown in Figure 1(a). In stimulated absorption, the transition of electrons between different energy levels requires stimulation by the energy of incoming photons. The position of electrons in energy levels and the energy of incoming photons influence the transition behavior of electrons between levels. Excited state electrons Free electrons are inherently unstable and frequently undergo spontaneous transitions back to the ground state E_0 , resulting in the emission of a photon with energy $h\nu_0$. This phenomenon is referred to as spontaneous emission. The light generated by spontaneous emission is somewhat dispersed, exhibiting several phases and directions of propagation. Additionally, during spontaneous emission, some energy transitions occur without radiation, primarily in the form of heat. When an electron in the excited state is stimulated by a photon with energy $h\nu_0$, it can transition from the high-energy excited state E_n back to the low-energy ground state E_0 , emitting a photon that has identical properties (direction, frequency, and energy) to the incoming photon. This process is called stimulated emission [4].

2.2. Population Inversion

To achieve laser emission, stimulated emission must dominate, and population inversion is a necessary condition for producing a large amount of stimulated emission. Under sufficiently strong excitation light, many electrons in the ground state undergo stimulated absorption and transition to the excited state, resulting in more electrons occupying the high-energy excited state as shown in Fig.2(b). This increases the likelihood of incoming excitation light encountering electrons in the excited state, making stimulated emission more likely, thereby amplifying the light and forming a laser [4-5]. Thus, the key to a laser emitting high-energy light lies in the condition where the number of electrons in the excited state E_n exceeds the number of electrons in the ground state E_0 , a state known as "population inversion" [5].

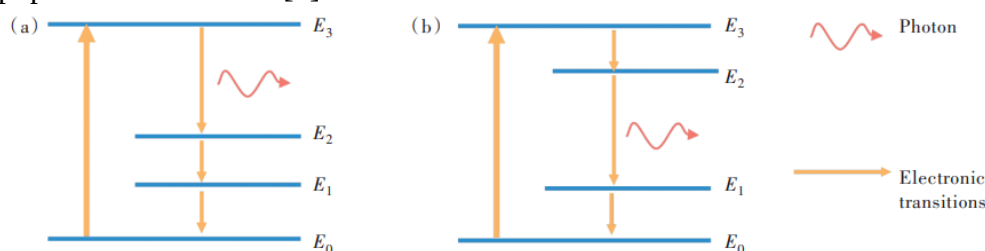


Fig 2. Illustration of two variations of the four-level structural system

2.3. Amplification of Spontaneous Emission

When the gain medium achieves population inversion under the illumination of a photon stream $h\nu_0$, light amplification can occur [6]. Even without the photon stream $h\nu_0$, photons generated by spontaneous emission can act as excitation light, causing electrons in the high-energy state to undergo stimulated emission, releasing a photon with identical properties to the spontaneous emission photon. This photon, along with the original, continues to induce further stimulated emissions, eventually producing outgoing light with a certain directionality and good monochromaticity, known as ASE (Amplified Spontaneous Emission), as shown in Fig.1 (c).

Gain media in lasers mainly include two types of energy-level systems: three-level systems and four-level systems. Three-level systems can achieve laser emission, as seen in ruby lasers [5]. However, in a three-level system, more than 50% of the particles must be transported from the ground state E_0 to the excited state E_1 to achieve population inversion, resulting in high laser thresholds and low efficiency. Fluorescent dyes, semiconductor quantum dots, perovskites, organic small molecules, and CDs are examples of four-level systems. There are two main forms of four-level systems: The first type as shown in Fig.2(a) consists of a ground state E_0 , a pump level E_3 , a laser upper level E_2 , and a laser lower level E_1 . During laser operation, the working particles are pumped from the ground state E_0 to E_3 by the pump source. Due to the short lifetime of the excited state E_3 , the particles quickly relax to E_2 . E_2 is a metastable state with a longer lifetime compared to the excited state E_3 , allowing electrons to gradually accumulate in E_2 , forming population inversion between E_2 and E_3 . Finally, electrons transition from the laser upper level back to the ground state, emitting laser light. The second type as shown in Fig.2 (b) operates similarly, but with the difference that E_1 is a metastable state. After the working particles are pumped to E_3 , they relax to E_1 and accumulate, leading to population inversion between E_1 and E_2 [6].

3. 1D Perovskite Photonic Crystal Lasers

Photonic crystals are a type of special artificial micro-nano structures made by arranging two or more media periodically, which modulate light. Since the concept of "photonic crystals" was proposed in 1987, their unique optical properties, such as photonic bandgaps, the Purcell effect, photon localization, and slow group velocity, have introduced new strategies for controlling electromagnetic wave transmission [7]. Incorporating photonic crystals into semiconductor lasers offers the following advantages: 1) constructing high-reflectivity mirrors; 2) creating high-Q microcavities; 3) enhancing the spontaneous emission coupling factor; and 4) increasing the gain of optical modes, which has significant advantages in reducing the laser threshold [8]. Combining photonic crystals with semiconductor lasers can result in ultra-compact or micro-scale lasers with high Q-values, small mode volumes, and low or even zero thresholds. Introducing perovskite materials into photonic crystal lasers, leveraging their high optical gain characteristics, can effectively compensate for optical losses, thereby achieving low-threshold lasers. This section will review the research progress in reducing laser thresholds based on one-dimensional (1D) photonic crystal lasers, classified according to their structures. 1D photonic crystals are structures typically composed of materials with alternating dielectric constants, where the dielectric constant exhibits periodic variation in only one direction. These structures are commonly used as high-reflectivity mirrors and anti-reflective coatings, and in lasers, they can replace traditional mirrors. Due to their nearly 100% reflectivity, they provide strong optical confinement, with advantages such as low absorption loss, resistance to mirror surface damage, and easier photonic integration. The specific structures can be classified into dielectric multilayer structures, grating structures, and air-bridge structures.

3.1. Dielectric Multilayer Structures

Dielectric multilayer structures consist of alternating thin layers of materials with different dielectric constants. In lasers, dielectric multilayer structures are typically used in two ways: 1) A multilayer distributed Bragg reflector (DBR) formed by stacking two different dielectric materials

alternately is considered a 1D photonic crystal and is used as a mirror; 2) A photonic crystal laser can be created by diffusing laser dyes into a medium with refractive index [9]. The dielectric multilayer perovskite photonic crystal lasers are usually restricted to DBR configurations due to the constraints imposed by material physical features.

Multilayer DBRs, with their periodically distributed dielectric constants, can be regarded as 1D photonic crystals. Compared to traditional mirrors like metal mirrors, they have higher reflectivity and lower optical loss, which can effectively reduce the optical loss in the resonant cavity and significantly contribute to lowering the laser threshold.

In 2017, Huang et al. from the University of Washington constructed a DBR microcavity laser using a 240 nm-thick CsPbBr₃ quantum dot (QD) thin film as the gain medium [10]. Fig.3 (a)–(b) demonstrate that the laser achieved an exceptionally low threshold of 0.39 $\mu\text{J}\cdot\text{cm}^{-2}$ when subjected to pulsed laser pumping at 400 nm for 50 fs. Due to the quantum confinement effect, QDs have a high photoluminescence quantum yield, which helps to improve the efficiency of light-emitting devices [11]. For comparison, the team conducted the same experiment using a pure CsPbBr₃ QD thin film, which resulted in a laser threshold of 5.6 $\mu\text{J}\cdot\text{cm}^{-2}$. This demonstrates that adding a high-reflectivity DBR ($R>99\%$) can effectively reduce the laser threshold and improve laser performance. However, this laser still has the issue of a relatively low Q-factor, approximately 73, which is related to the fact that DBR lasers only provide optical confinement in one direction, an inherent problem with this type of laser.

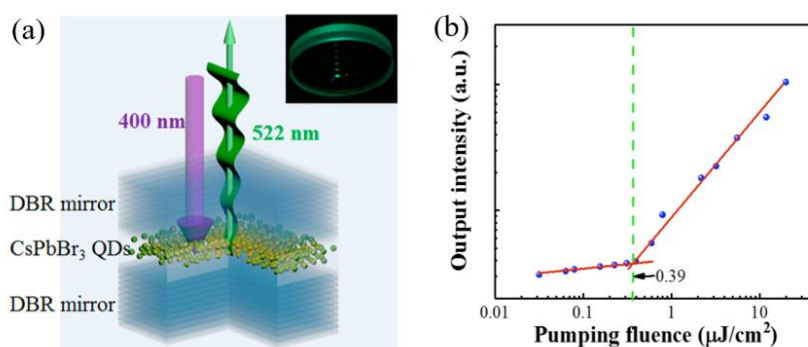


Fig 3. (a) Diagram and inset of the CsPbBr₃ quantum dot VCSEL device; (b) Analysis of output power in relation to pump energy density indicates a specific threshold of 0.39 $\mu\text{J}/\text{cm}^2$ [10]

In addition to fully inorganic perovskite materials, low-threshold laser emission has also been observed in DBR/organic-inorganic hybrid perovskite/DBR structures at room temperature, with some even achieving continuous-wave pumped laser emission. In 2019, Neda et al. introduced a vertical-cavity surface-emitting laser (VCSEL) that achieved an extremely low threshold of 2.2 $\mu\text{J}\cdot\text{cm}^{-2}$, as shown in Fig.4 (a)–(b), with a pump pulse of 300 ps, functioning at ambient temperature [12].

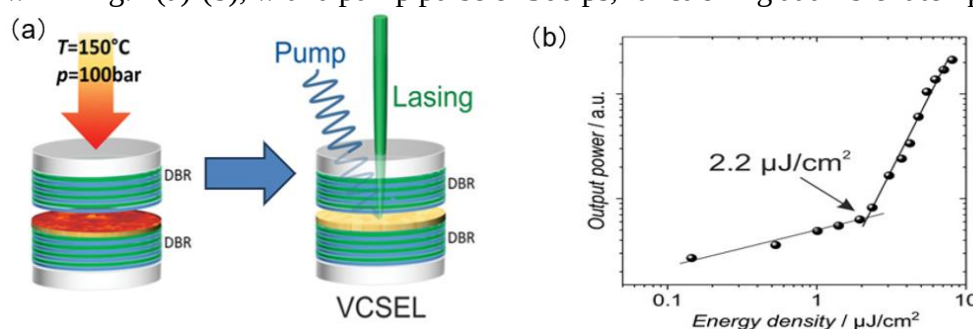


Fig 4. (a) Diagram of VCSEL configuration;(b) Analysis of output power in relation to pump energy density. [12]

Notably, this laser did not depend on CsPbX₃ nanoparticles (NPs). Instead of covering the dielectric substrate with layers of nanocrystals or quantum dots, they applied a layer of cesium lead bromide. Next, the identical mirrors were employed as stamps in a hot-pressing/recrystallization procedure at 150°C and 100 bar. Following this procedure, the stamp was positioned on the surface

of the stabilized perovskite layer, hence creating the complete VCSEL structure. Neda et al.'s CsPbBr thin-film VCSEL was pumped with relatively "long" 300 ps pulses, in contrast to Huang et al.'s VCSEL which used CsPbBr₃ nanocrystals as the gain medium with thresholds of 0.39 $\mu\text{J cm}^{-2}$ under femtosecond pulse pumping and 98 $\mu\text{J cm}^{-2}$ under nanosecond pulse pumping [10]. Hence, their exceptionally low threshold level of 2.2 $\mu\text{J cm}^{-2}$ is even more noteworthy. The relatively low threshold of fully inorganic perovskite DBR lasers may be attributed to the better thermal and moisture stability of fully inorganic perovskites in atmospheric environments.

3.2. Grating Structures

The application of grating structures in perovskite photonic crystal lasers is primarily seen in distributed feedback (DFB) lasers, where perovskite materials are typically introduced into the laser structure through methods like spin coating and nanoimprinting [8]. Ultra-low thresholds in these lasers can be attained by precisely regulating the equilibrium between feedback coupling and output coupling.

In 2017, Gong et al. from Fudan University fabricated a perovskite DFB laser with a grating structure on a quartz substrate, as shown in Fig. 5 (a) [13]. The team first used nanoimprinting technology to create a DFB resonant cavity with a periodicity of 360 nm on a polymer film coated on a quartz substrate. Then, an 80 nm thick perovskite film (CsPbBrI₂-PEO) was deposited on top as the gain medium. As shown in Fig. 5 (b), under laser pumping with a wavelength of 355 nm and a pulse width of 90 ps, the laser successfully achieved a threshold of 33 $\mu\text{J cm}^{-2}$ and emitted a 654 nm single-mode laser. The low laser threshold is attributed not only to the DFB cavity but also to the high optical gain coefficient (161.2 cm^{-1}) of the CsPbBrI₂-PEO film, which exhibits a photoluminescence intensity an order of magnitude higher than the CsPbBrI₂ film. This improvement can be credited to PEO and the nanoimprinting process, which enhanced the quality of the CsPbBrI₂ film and reduced scattering loss. However, the team noted that the relatively broad laser emission (4.9 nm) was due to an inappropriate grating duty cycle, indicating that there is room for further optimization of the FWHM and Q-factor of the laser.

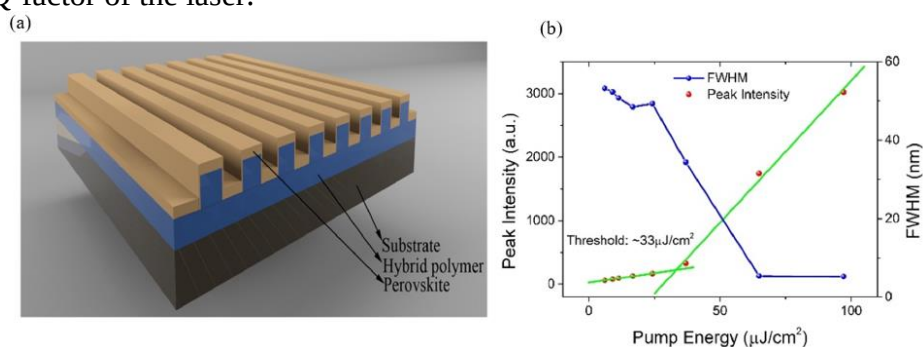


Fig 5. (a) Illustrative representation of a DFB resonator utilizing a completely inorganic perovskite gains medium;(b) Analysis of the relationship between peak intensity and full width at half maximum (FWHM) with pump energy density [13]

In 2018, Pourdavoud's team from the University of Wuppertal in Germany disclosed a DFB laser that utilised a MAPbBr₃ film. The laser achieved a remarkably low threshold of 3.4 $\mu\text{J cm}^{-2}$, as depicted in Fig. 6 [14].

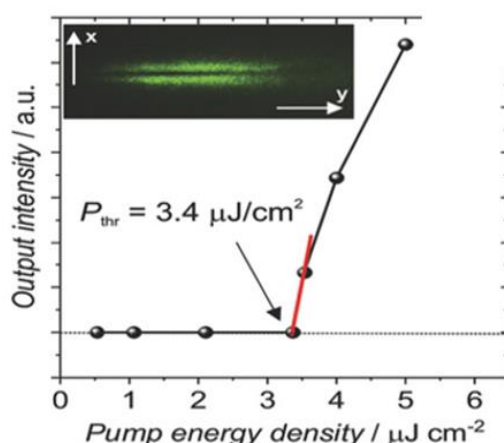


Fig 6. An analysis of output power vs excitation energy density indicates a specific threshold of 3.4 $\mu\text{J/cm}^2$ [14]

This threshold is significantly lower than that of previously reported inorganic perovskite DFB lasers. Unlike the optimization method used by Gong et al., Pourdavoud et al. did not introduce any additional materials but instead optimized the MAPbBr₃ film using a thermal pressing method. This process increased the grain size in the film, improving its stability in air and reducing the surface roughness to 0.6 nm, which is an order of magnitude lower than the roughness in Gong et al.'s work. This significant reduction in scattering loss resulted in a Q-factor exceeding 3900, contributing to the lower laser threshold.

Grating structures modulate light using horizontally oriented Bragg gratings, resulting in a much smaller longitudinal size compared to DBR lasers [15]. However, the perovskite gain medium is directly exposed to air, which is detrimental to the long-term stability of the device, necessitating protective encapsulation. Additionally, compared to the multilayer structure of DBRs, grating structures are more intricate and require more precise fabrication processes [8]. Moreover, grating structures can only confine light in one direction, providing limited confinement.

3.3. Nanobeam Structures

Nanobeam structures consist of narrow dielectric strips with periodically distributed air holes. The gain medium is directly loaded onto these structures, allowing the use of the photonic bandgap effect and the high-density electromagnetic field at the photonic band edge to achieve laser emission [16]. In lasers, nanobeam structures typically use sinusoidally distributed air holes at the center to form a photonic well, effectively confining light and reducing scattering losses from the photonic crystal cavity (PhC) to the outside [17]. The periodic air holes at both ends of the microcavity act as photonic crystal mirrors due to the photonic bandgap effect, ensuring that the wavelength range of interest is highly reflected, thus increasing the Q-factor and enabling low-threshold lasing. Sinusoidal photonic crystal nanobeam structures are usually constructed by linearly varying the size and periodicity of the air holes near the central position [18].

The integration of sinusoidal photonic crystal nanobeam structures with perovskite materials can significantly augment the spontaneous emission of the material. This approach effectively tackles the issue of low quantum efficiency resulting from the polycrystalline nature of perovskite films. Consequently, it leads to an increase in optical gain and an improvement in laser performance, specifically in terms of device footprint, laser threshold, and FWHM. In 2019, Fong et al. from Nanyang Technological University successfully made a connection at ambient temperature between completely inorganic CsPbBr₃ perovskite nanocrystals and a silicon nitride nanobeam photonic crystal cavity is encased in PMMA glass [19]. This coupling led to an increase in the photoluminescence of the CsPbBr₃ perovskite nanocrystals. The DBR mirrors at both ends of the optimized resonant cavity were composed of 40 elliptical holes with a periodicity (a) of 157 nm (long axis of 202 nm, short axis of 100 nm), as shown in Fig. 7.

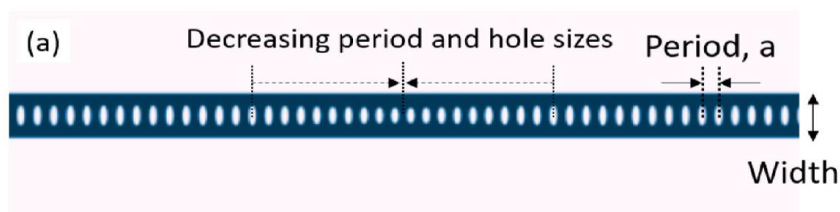


Fig 1. Illustration of a nanobeam including elliptical holes, annotated with the parameters period, a , and width [19]

The cavity itself consisted of 20 symmetrically distributed air holes with periodicity and elliptical hole sizes increasing regularly from the center, forming a sinusoidal distribution. Thus, this resonant cavity is referred to as a sinusoidal nanobeam resonant cavity. The simulated Q-factor of the cavity was about 9000, with a mode volume of approximately $3(\lambda/n)^3$, and its resonant wavelength was around 520 nm, matching the photoluminescence wavelength of CsPbBr₃ perovskite nanocrystals. An investigation of time-resolved photoluminescence decay showed a 1.4-fold increase in the spontaneous emission of CsPbBr₃ perovskite nanocrystals when the concentration of CsPbBr₃ nanocrystals in the PMMA layer was manipulated [19]. This results in the promising connection of perovskite nanocrystals with photonic crystals (PhC). The localization of waves in small-mode volume cavities might be utilized as light sources in integrated photonic circuits or for the miniaturization of photonic devices.

Fig. 8 illustrates the fabrication of a hybrid laser by He et al. from Sun Yat-sen University in 2020 [19].

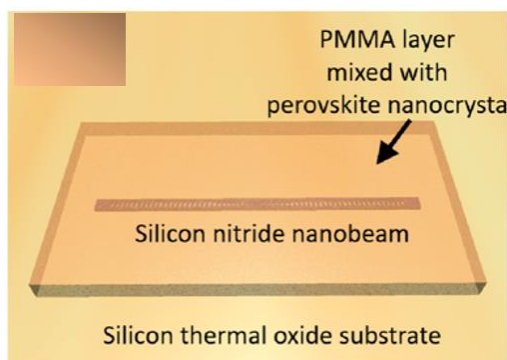


Fig 8. Illustrative representation of a photonic cavity consisting of a perovskite nanocrystal connected SiN nanobeam [19]

The laser utilizes CsPb(Br/I)₃ perovskite nanocrystals and a silicon nitride nanobeam microcavity. The achievement of high-performance coherent emission with a laser threshold of $5.62 \mu\text{J}/\text{cm}^2$, a full width at half maximum (FWHM) of 0.045 nm, and a tiny mode volume of $2.5(\mu/n)^3$ was demonstrated by dispersing perovskite nanocrystals in the PMMA layer as emitters, as depicted in Fig. 9 [20].

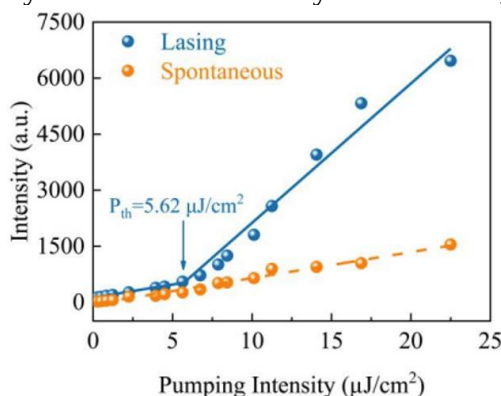


Fig 9. L–L curve showing nonlinear lasing emission and linear spontaneous emission, with a lasing threshold of $5.62 \mu\text{J}/\text{cm}^2$ [20]

Lacking grating couplers or adiabatic tapers, the experiment augmented the number of air holes in the Bragg mirrors at both ends of the nanobeam, leading to a Q-factor resonant cavity that was ten times more than in prior studies. The measured Q-factor of this laser was 17000, with one reason for the high Q-factor being the increased effective refractive index ratio between the PMMA layer and the nanobeam. Moreover, it has been demonstrated that photonic crystal nanocavities produced on solid substrates have rather superior thermal and mechanical stability in comparison to conventional suspended cavities [19]. The use of a PMMA encapsulating layer on the SiN surface can greatly improve the Q-factor of the cavity mode, offering a novel method for producing nanobeam microcavities with high Q-factor. Reducing the dimensionality of photonic crystals to 1D can improve the Q-factor of the microcavity and lower the laser threshold to some extent, but optical confinement is only achieved in one direction.

4. Conclusion

In recent years, significant progress has been made in lowering the excitation threshold of semiconductor lasers through the introduction of novel materials and structural designs. The combination of hybrid perovskite materials and photonic crystal structures has opened new pathways for achieving low-threshold lasing. Due to their excellent optical gain properties, perovskite materials, when integrated with one-dimensional photonic crystal structures, have demonstrated great potential for further reducing laser size and enhancing efficiency.

In the future, these technologies are expected to drive the development of more energy-efficient lasers and play a key role in integrated photonics and miniaturized systems. Future research should focus on further optimizing these materials and structures to address existing issues such as optical loss and stability, ultimately paving the way for the realization of truly low-threshold or even threshold-free lasers.

References

- [1] Gao R, Li G H, Han Y, et al. Carrier lifetime exceeding 81 ns in single crystalline perovskite nanowires enable large on-off ratio photodetectors. *Organic Electronics*, 2020, 83: 105744.
- [2] Xing G C, Mathews N, Lim S S, et al. Low-temperature solution-processed wavelength-tunable perovskites for lasing. *Nature Materials*, 2014, 13(5): 476-480.
- [3] Amann M C. Semiconductor lasers. *Encyclopedia of Condensed Matter Physics*. Elsevier, 2005: 281-289.
- [4] Yariv A. *Quantum Electronics*. 3rd ed. New York: John Wiley & Sons, 1989: 12-22.
- [5] Scully M O, Zubairy M S. *Quantum Optics*. Cambridge: Cambridge University Press, 1997: 168-172.
- [6] Keppler S, Sävert A, Körner J, et al. The generation of amplified spontaneous emission in high-power CPA laser systems. *Laser & Photonics Reviews*, 2016, 10(2): 264-277.
- [7] Chen K, Sheng Q Q, Han J, et al. Study on photonic crystal and its application. *Optoelectronic Technology*, 2003, 23(1): 16-23.
- [8] Tang H X, Wang Q M. Research progress of semiconductor photonic crystal lasers. *Semiconductor Optoelectronics*, 2005, 26(3): 165-171, 192.
- [9] Kong J Y, Chu S, Huang J, et al. Use of distributed Bragg reflectors to enhance Fabry-Pérot lasing in vertically aligned ZnO nanowires. *Applied Physics A*, 2013, 110(1): 23-28.
- [10] Huang C Y, Zou C, Mao C Y, et al. CsPbBr₃ perovskite quantum dot vertical cavity lasers with low threshold and high stability. *ACS Photonics*, 2017, 4(9): 2281-2289.
- [11] Wang Y, Li X, Zhao X, et al. Nonlinear absorption and low-threshold multiphoton pumped stimulated emission from all-inorganic perovskite nanocrystals. *Nano Letters*, 2016, 16(1): 448-453.
- [12] Pourdavoud N, Haeger T, Mayer A, et al. Room-temperature stimulated emission and lasing in recrystallized cesium lead bromide perovskite thin films. *Advanced Materials*, 2019, 31(34): 1903717.
- [13] Gong J, Wang Y, Liu S, et al. All-inorganic perovskite-based distributed feedback resonator. *Optics Express*, 2017, 25(24): A1154-A1161.

- [14] Pourdavoud N, Mayer A, Buchmüller M, et al. Distributed feedback lasers based on MAPbBr₃. *Advanced Materials Technologies*, 2018, 3(4): 1700253.
- [15] Zhai T R, Zhang X P, Dou F. Microscopic excavation into the optically pumped polymer lasers based on distributed feedback. *Chinese Physics Letters*, 2012, 29(10): 104204.
- [16] Niu N, Woolf A, Wang D Q, et al. Ultra-low threshold gallium nitride photonic crystal nanobeam laser. *Applied Physics Letters*, 2015, 106(23): 231104.
- [17] Karnadi I, Son J, Kim J Y, et al. A printed nanobeam laser on a SiO₂/Si substrate for low-threshold continuous-wave operation. *Optics Express*, 2014, 22(10): 12115-12121.
- [18] Li Y Z, Zhang J X, Huang D D, et al. Room temperature continuous-wave lasing from monolayer molybdenum ditelluride integrated with a silicon nanobeam cavity. *Nature Nanotechnology*, 2017, 12(10): 987-992.
- [19] Fong C F, Yin Y, Chen Y, et al. Silicon nitride nanobeam enhanced emission from all-inorganic perovskite nanocrystals. *Optics Express*, 2019, 27(13): 18673-18682.
- [20] He Z, Chen B, Hua Y, et al. CMOS compatible high-performance nanolasing based on perovskite-SiN hybrid integration. *Advanced Optical Materials*, 2020, 8(15): 2000453.