Review

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Advances in inorganic and hybrid perovskites for miniaturized lasers

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Abstract: The rapid advancement of perovskite-based optoelectronics devices has caught the world's attention due to their outstanding properties, such as long carrier lifetime, low defect trap density, large absorption coefficient, narrow linewidth and high optical gain. Herein, the photonic lasing properties of perovskites are reviewed since the first stimulated emission of perovskites observed in 2014. The review is mainly focused on 3D structures based on their inherently active microcavities and externally passive microcavities of the perovskites. First, the fundamental properties in terms of crystal structure and optical characteristics of perovskites are reviewed. Then the perovskite lasers are classified into two sections based

on the morphology features: the ability/inability to support lasing behaviors by themselves. Every section is further divided into two kinds of cavities according to the light reflection paths (Standing wave for the Fabry–Pérot cavity and travelling wave for the Whispering-Gallery-Mode cavity). The lasing performance involves fabrication methods, cavity sizes, thresholds, quality factors, pumping sources, etc. Finally, some challenges and prospects for perovskite lasers are given.

Keywords: perovskite; ASE; laser; semiconductor; Fabry–Pérot cavity; Whispering Gallery Mode.

1 Introduction

The demands of miniaturization and integration of photonic components have opened up and pushed the development of a wide variety of new application areas [1–4]. There has been a long-standing and continuous pursuit of ultrasmall laser sources due to the rapid progress of the optical integrated systems. The initial miniaturized laser could be traced back to the introduction of the vertical-cavity surface-emitting laser (VCSEL) in the last century [5]. The cavity lengths of the VCSEL were several times those of the emitted light wavelength. Subsequently, the microdisk lasers [6], photonic crystal lasers [7], nanowire lasers [8], and so on, further shrank the laser sizes to several microns. However, the optical loss began to increase with the further miniaturization of laser devices, thereby leading to high lasing threshold and high energy consumption. Thus, finding ways to achieve high optical gain and low losses is an urgent concern in the field of semiconductors and ultra-compact microcavities, as these could help approach and break the diffraction limit for nanophotonic laser.

In 2009, hybrid organic-inorganic perovskites with an efficiency of 3.8% were first adopted as visible-light sensitizers in photovoltaic cells [9], attracting great research attention worldwide. According to the Shockley-Queisser limit, in order to achieve the limit of the solar cell efficiency, the external luminous efficiency of the device must be close to 100%. Therefore, a high-quality solar cell material is also considered a high-quality optical emission

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material. The first demonstration of low-threshold, stable and tunable amplified spontaneous emissions (ASEs) in perovskite thin films was reported in 2014 [10]. The optical gain was measured up to $\sim 3200 \text{ cm}^{-1}$ of MAPbI_3 perovskite thin film [11], which was comparable to that of single-crystalline GaAs. Furthermore, the stimulated emission lifetime could sustain as long as $\sim 200 \text{ ps}$. Since then, studies on miniaturized lasers from perovskite semiconductors have experienced explosive growth and have led to great advances in lasing performance [12–16]. Owing to their ultracompact physical size and highly localized coherent output, perovskite nanostructures, such as nanowires (NWs) and nanoplate (NPs), are promising building blocks for fully integrated nanoscale photonic and optoelectronic devices.

For example, Zhang et al. demonstrated that the remarkable whispering-gallery-mode (WGM) nanolasers of perovskite NPs can be easily integrated onto conductive platforms, such as Si, Au and indium tin oxide (ITO), which have shown great promise for the related on-chip integration [13]. In addition to the nanolasers based on the individual nanostructured devices, a photonic cavity can also utilize external optical cavities, including SiO_2 spheres [17, 18], distributed Bragg reflector (DBR) [14, 19], photonic crystal (PC) [7, 20] and so on, which increased the versatility of nanolaser configurations. Due to the rapid progress, substantial reviews on the perovskite lasing have been carried out, including those on quantum dots, NW and NP lasers [15, 21–25].

Herein, we first summarize the important and fundamental properties of perovskites from their component-engineering for the bandgap, optical gain, and exciton binding energy in Section 2. Next, we review the advances of perovskite photonic nanolasers, which we divided into active and passive cavities, and then discuss the lasing properties in Sections 3 and 4. Finally, the challenges and perspectives of future research areas in perovskite lasers are highlighted in Section 5.

2 The fundamental properties of perovskite

2.1 Crystal structure and tunable bandgap

Three-dimensional (3D) metal halide perovskites with the general chemical formula ABX_3 (Figure 1A) have received extensive attention, where A and B are cations, bound to an anion X [26]. Among them, the A ion could be organic (e.g. CH_3NH_3^+ (MA^+), $\text{NH}_2\text{CH}=\text{NH}_2^+$ (FA^+)) and

inorganic cations (e.g. Cs^+ , Rb^+ , etc.). The B ion could be a metallic cation, such as Pb^{2+} , Sn^{2+} , etc., the X ion could be halogen, such as Cl^- , Br^- and I^- [28]. The B together with six X atoms consists of an $[\text{BX}_6]^{4-}$ octahedron, which then form a 3D octahedral network. In addition, the octahedral units could be cut into layers as 2D perovskites and structurally distorted into 0D perovskites [29]. These can affect the physical properties, such as the exciton binding energy and bandgap.

Figure 1B describes the emission spectra from perovskites [15], which can be tuned by halogen stoichiometry and serve as substitute of the A or B site. For example, the bandgaps decrease with the increase of effective ionic radius (R) of A site ($R_{\text{Cs}^+} < R_{\text{MA}^+} < R_{\text{FA}^+}$) due to the lattice expansion [30]. Furthermore, through compositional modulations from either a mixture of Cl^- and Br^- or Br^- and I^- , the bandgap energies and emission spectra of all-inorganic CsPbX_3 are readily tunable over the entire visible spectral region of 410–700 nm (Figure 1C) [27]. Aside from the facile tuning of the bandgap, the narrow FWHM (12–42 nm), tunable optical absorption and high PLQY (Figure 1D–E) would be beneficial in developing multicolor or multi-wavelength light-emitting devices, including LEDs, optical amplifiers and lasers.

2.2 Optical gain

There are three essential elements of a laser: gain medium, pumping source and the resonant cavity. The gain medium provides transition energy level that induces specific particles transiting from higher energy levels to a lower energy level to form stimulated emission under the excitation of pump source. The generated photons are confined and provided feedback by the resonant cavity, which would output the spontaneous emission with certain frequency, and subsequently form a laser oscillation above a certain pump intensity. For a simple laser resonator that is a Fabry–Pérot (F–P) cavity, the minimum cavity length must satisfy $L_{\min} = -\ln(R_1 R_2)/G_m$ [31], where R_1 , R_2 and G_m are the end mirror reflectivities and the modal gain related to the material gain, respectively. Therefore, the high optical gain is highly beneficial for miniaturized laser.

The high optical gain indicates the high ability of light amplification and lowers the requirement of strong pump to overcome the optical losses for the lasing behaviors. The net optical gain could be acquired by variable stripe length (VSL) technique in which the emission intensity is recorded as a function of the “stripe length” of the excitation laser spot shaped by a cylindrical mirror

Figure 1: The optical properties of perovskites.

(A) The perovskite crystal structure. A is usually the MA, FA or Cs. B is Pb or Sn. X is halogen. Reproduced with permission [26]. Copyright © 2014, Springer Nature. (B) Schematics of the bandgaps of MAPbX₃, FAPbX₃, CsPbX₃ and CsSnX₃. Reproduced with permission [15]. Copyright © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (C) Top: the colloidal solutions in toluene under UV lamp ($\lambda = 365$ nm). Bottom: the PL spectra of CsPbX₃. Reproduced with permission [27]. Copyright © 2015, American Chemical Society. (D) The typical optical absorption and PL spectra. Reproduced with permission [27]. Copyright © 2015, American Chemical Society. (E) The time-resolved PL decays for all samples shown in (D) except CsPbCl₃. Reproduced with permission [27]. Copyright © 2015, American Chemical Society.

(Figure 2A) [18]. The current highest net optical gain is as high as $\sim 3200 \pm 830$ cm⁻¹ from MAPbI₃ perovskite thin film via atomic layer deposition, comparable to that of single-crystalline GaAs [11]. Employing the VSL method, the net optical gains are determined to be ~ 980 , ~ 580 , ~ 502 , ~ 480 , ~ 450 , ~ 250 and ~ 125 cm⁻¹ for CsPbBr₃ nanorods [36], CsPbBr₃ nanocrystals [32], CsPbBr₃ nanocuboids [33], FAPbBr₃ nanocrystal [37], CsPbBr₃ nanocrystals [18], MAPbI₃ film [10] and MAPbI₃ nanocrystals [17], respectively. Although there is no definite value, the gain of the Cs-based perovskites is expected to be higher than those of the MA-based ones.

In addition, the gain lifetime is also investigated by ultrafast transient absorption spectroscopy (TAS). In 2015, Sutherland et al. observed the gain of MAPbI₃ perovskite film via TAS, which could sustain as long as 200 ps and large gain bandwidth at high pump fluence (Figure 2B) [11]. Subsequently, Xu et al. found that the lifetime of optical gain was ~ 35 ps and that the multiexciton effect plays an important role in the development

of optical gain in CsPbBr₃ nanocrystals [32], as depicted in Figure 2C. Meanwhile, Liu et al. used the TAS to study the optical gain of CsPbBr₃ nanocubes under different pump intensities and observed the lifetime of 19.7 ps, which was also confirmed by the output pulse width of laser and ascribed to the electron-hole plasma [33]. Furthermore, the time-resolved photoluminescence (TRPL) measurement is another strong tool to study the stimulated emission. In 2015, Zhu et al. reported that the stimulated emission process was less than 20 ps for both MAPbBr₃ and MAPbI₃ NWs (Figure 2E) and was assigned to a correlated electron-hole plasma, which formed in a broad carrier density range of 10^{16} – 10^{19} cm⁻³ [34]. Fu et al. also observed the ultrafast decay of stimulated emission of <20 ps in FAPbI₃ NWs [38]. As shown in Figure 2F, similar rapid decay corresponding to the stimulated emission process of <30 ps was measured by TRPL for CsPbX₃ NWs under strong excitation, and the electron-hole plasma mechanism was responsible for the stimulated emission [35]. More recently, Schlaus et al. further

Figure 2: The optical gain and lifetime of stimulated emission process.

(A) The VSL experiment for estimation of modal net gain. Reproduced with permission [18]. Copyright © 2015, Springer Nature. (B) Gain spectra at 3 ps of MAPbI₃ thin film for various pump fluences. Reproduced with permission [11]. Copyright © 2014 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (C) The TA spectroscopic data of CsPbBr₃ nanocrystals film with two-photon excitation at 1.5 mJ/cm². SE: stimulated emission. Inset: the TA kinetics probed at 527 nm with components of the SE and excited state absorption. Reproduced with permission [32]. Copyright © 2016, American Chemical Society. (D) The TA spectroscopic data of CsPbBr₃ nanocuboids film with two-photon excitation below (top) and above (bottom) threshold. Reproduced with permission [33]. Copyright © 2018, American Chemical Society. (E) The TRPL decay kinetics after photoexcitation with pump fluence below and above the threshold. Reproduced with permission [34]. Copyright © 2015, Springer Nature. (F) The TRPL from a CsPbBr₃ nanowire below (red) and above (blue) the lasing threshold. The emergence of a rapid decay component (<10 ps) suggests rapid carrier quenching due to stimulated emission. Reproduced with permission [35]. Copyright © 2016, National Academy of Sciences of the United States of America.

reported that the CsPbBr₃ NW lasing originated from the stimulated emission of an electron-hole plasma by TRPL and transient reflectance [39]. They observed an anomalous blue-shifting of the lasing gain profile with time up to 25 ps and assigned this as a signature for lasing involving plasmon emission. The fast stimulated emission process leads to the low threshold for population inversion, which is faster than the Auger process (nonradiative loss), and then avoids being disturbed by the Auger recombination for the lasing process [40]. Therefore, the

excellent optical gain characteristics of perovskites offer great promise for light-emitting devices.

2.3 Exciton binding energy

Understanding the nature of excitons is a critical key in the optical emission performance of semiconductors. An exciton consists of an electron and a hole bounded by Coulomb forces. The Coulomb interactions between the

electron and hole could be denoted by the exciton binding energy (E_b), which determines the existence or dissociation of excitons and then influences the exciton recombination dynamics. At room temperature, when the E_b is smaller than the thermal fluctuation energy ($k_b T \sim 26$ meV), the generated exciton is likely to dissociate into free carriers (electron and hole), which is highly desired for photovoltaic device. On the contrary, the large E_b signifies the existence and stability of excitons, which is more favorable for light-emitting devices due to the efficiently radiative recombination achieved through the excitons at relatively lower carrier densities [41].

Considering the importance of E_b , many experimental and theoretical efforts have been carried out in recent years. For example, temperature-dependent PL [42–45], optical absorption [46, 47] and magnetoabsorption spectra [48–50] have been widely used to obtain the values in the perovskite family. MAPbI₃ has attracted extensive attention as a common and important light absorption layer in perovskite solar cells. The initial E_b values of MAPbI₃ have been reported in a wide range of 2–50 meV [41, 49, 50]. Furthermore, the E_b of the analogues of MA-based perovskites has been studied and shown to increase with substitution of halide elements from I, Br to Cl. For example, the E_b values of MAPbCl₃ and MAPbBr₃ have been reported as ranging from ~50–106 meV [51, 52] and 21–100 meV [51, 53, 54], respectively, higher than that of MAPbI₃. Similar trends have been shown in FAPbX₃ (~8.1–110 meV) and CsPbX₃ (~20–72 meV) [41, 47]. The perovskite E_b values from several to hundreds of meV are favorable for high-performance and versatile optoelectronic devices. Especially, the large E_b (over 26 meV) values of (MA/FA)PbBr₃, (MA/FA)PbCl₃ and CsPbX₃ have been demonstrated to stabilize excitons at room temperature and boost the developments of miniaturized coherent photonic sources, such as micro/nano lasers.

3 Perovskite lasers without external cavity

Although the halide perovskites have been investigated for a long time, the first observation of perovskites for stimulated emission as ASE was only achieved in 2014 by Xing et al [10]. Subsequently, the perovskite ASE and lasers have a big growth spurt. Based on the composition, structure and morphology engineering, perovskite micro-nano lasers have achieved rapid advancements. For example, due to the flexible structure of halide perovskites, their shapes and sizes can be conveniently manipulated as

QDs, NWs, NPs and so on. Particularly, the NWs and NPs have regular and smooth end faces, which can naturally serve as an active resonant cavity for F–P/WGM oscillation and optical gain media by themselves. The ultracompact physical sizes from several to tens of microns can confine the generated photons located in the nano-structured cavities, and when the photons satisfy the resonance conditions of microcavities and obtain sufficient gain, the lasing will happen. Thus, perovskite laser is highly promising for integration onto on-chip circuitry because of its small modal volume.

3.1 Perovskite F–P lasers

Benefitting from the relatively high difference between the refractive indexes of perovskite gain medium and air, the interfaces can be regarded as high reflectivity mirrors for microcavities. The simplest cavity is the F–P cavity geometry, which consists of only two end mirrors. In 2015, the first perovskite NW lasing from hybrid organic-inorganic perovskites (MAPbX₃) was reported by Zhu et al. [34]. The single-crystal NWs from low-temperature solution processing possessed high optical quality and suitable dimension with long carrier lifetimes and low non-radiative recombination rates, which are ideal for F–P lasing. As shown in Figure 3A, the remarkable performance of perovskite NW lasing are demonstrated under optical pumping (402 nm, ~150 fs, 250 kHz) with small size (length: 8.5 μm), low lasing thresholds (0.22 μJ/cm²), high quality factors (~3600) and broad tunability lasing covering the near-infrared to visible wavelength region (~500–790 nm). Soon after that, Xing et al. also demonstrated optical-pumped (400 nm, 50 fs, 1 kHz), room-temperature MAPbI₃ nanowire lasers (~10 μm length and ~200 nm diameter) with wavelength of 777 nm, low threshold of 11 μJ/cm², FWHM of 1.9 nm and a quality factor of 405 (Figure 3B) [55]. The free-standing perovskite nanowires were synthesized by a two-step vapor phase synthesis method with good optical properties and long electron hole diffusion length. Tunable lasing has also been observed from MAPbBr₃ (551 nm) and MAPbI_xCl_{3-x} (744 nm) with thresholds of 20 and 60 μJ/cm², respectively. Notably, the relatively short lifetimes for the NWs can be attributed to the special morphology that is not compatible with its intrinsic lattice structure and large surface to volume ratio. To improve the thermal and photo stability of hybrid perovskites, Fu et al. substituted MA with FA to prepare the FAPbX₃ NWs with lengths ranging from ~6–30 μm via low-temperature solution growth [38]. Under 402 nm pulsed laser excitation (150 fs, 250 kHz), the FAPbX₃ NWs were

Figure 3: The perovskite NW lasers.

(A) Top: optical image (left) of single NW with a length of 8.5 μm . The middle and right images show the NW emission below and above lasing threshold (scale bar, 10 μm), respectively. Middle: The NW emission spectra around the lasing threshold. Inset: The integrated emission intensity and FWHM as a function of pump intensity. Bottom: The tunable lasing from mixed perovskite NWs. Reproduced with permission [34]. Copyright © 2015, Springer Nature. (B) Top: The evolution from spontaneous emission to lasing in a typical MAPbI₃ nanowire. Inset: The threshold behavior with a knee at $\sim 11 \mu\text{J}/\text{cm}^2$. Bottom: The lasing spectra of MAPbI₃, MAPbBr₃ and MAPbI_xCl_{3-x} with the corresponding thresholds. Reproduced with permission [55]. Copyright © 2015, American Chemical Society. (C) Top: The broad wavelength-tunable lasing from single-crystal lead perovskite NWs. Bottom: The lasing stability measurement of FAPbI₃, MABr-stabilized FAPbI₃ and MAPbI₃ NW. Reproduced with permission [38]. Copyright © 2016, American Chemical Society. (D) Left: The power-dependent emission spectra from the CsPbBr₃ NW. Inset: The optical image of NW under high pump intensity. Right: The integrated emission intensity for CsPbBr₃ NW under constant pulsed excitation while exposed to N₂ and air. Reproduced with permission [35]. Copyright © 2016, National Academy of Sciences of the United States of America. (E) Top: The schematic diagram of the aligned CsPbBr₃ NW lasers. Bottom: The real-color optical images of the in-plane directional CsPbBr₃ NWs under optical pumping above the lasing threshold. Reproduced with permission [56]. Copyright © 2018, American Chemical Society. (F) Left: The PL spectra of a 20 μm -long NW obtained with increasing excitation power densities. Inset: The integrated intensity plotted against the power density. Right: The fluorescence images above threshold with different lengths. Reproduced with permission [57]. Copyright © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

demonstrated to have low lasing thresholds of several $\mu\text{J}/\text{cm}^2$ (~ 6 – $30 \mu\text{J}/\text{cm}^2$) and high quality factors of about ~ 1500 – 2300 . As depicted in Figure 3C, through cation and anion substitutions, the NW lasers can be tuned from 490 to 820 nm, and can fill in the gap of lasing wavelength that are previously unavailable with MA-based perovskites. In particular, both FAPbI₃ and MABr-stabilized FAPbI₃ NW lasers demonstrated better photostability than MAPbI₃ NW lasers due to the better hydrogen bonding between FA and the I anions in the former.

In addition to the hybrid perovskite NWs, the first all-inorganic CsPbX₃ NW lasers (lengths: ~ 2 – $40 \mu\text{m}$) were reported by Eaton et al. with low-temperature, solution-phase growth in 2016 [35]. The NWs F–P lasing was pumped by 400 nm pulsed source (150–200 fs, 295 kHz) and displayed an excellent performance with a threshold of $\sim 5 \mu\text{J}/\text{cm}^2$ and maximum quality factor of $\sim 1009 \pm 5$. Remarkably, the lasing can be maintained for over 1 h ($\sim 10^9$ excitation cycles) and persist upon exposure to ambient atmosphere, showing much better performance

than the hybrid perovskite NW lasers (Figure 3D). Fu et al. also reported the facile solution growth of single-crystal CsPbX_3 NWs for lasing [58]. They examined CsPbBr_3 NWs with different lengths ranging from 5–21 μm and observed either single or multiple lasing with thresholds ranging from 2.8–9 $\mu\text{J}/\text{cm}^2$. For example, under optical excitation with 402 nm, 150 fs and 1 kHz, lasing action at a peak wavelength of ~ 538 nm with low threshold of ~ 6.2 and a high quality factor of 2069 (FWHM ~ 0.26 nm) was achieved from an individual CsPbBr_3 NW with a length of 12 μm and width of 0.7 μm . Using continuous pumping, the NW laser can output stable lasing emissions for longer than 8 h or over 7.2×10^9 pulse excitations.

Due to the large E_b and sub-wavelength scale of NWs, the strong light–matter interactions of NWs were also revealed by the exciton-polariton model with the reduced oscillator strength [40, 59–61]. Park et al. synthesized the CsPbX_3 NWs with lengths of ~ 2 –15 μm by using the chemical vapor transport method and realized the lasers with low thresholds of 6, 3 and 7 $\mu\text{J}/\text{cm}^2$, with high quality factors of 1200, 1300, and 1400 for CsPbI_3 , CsPbBr_3 and CsPbCl_3 , respectively [40]. They also discovered the nonclassical and nonidentical spacing of the lasing modes, thereby suggesting that the general photonic model was inadequate in explaining the F–P modes in the CsPbBr_3 nanowires. This can be attributed to the strong light–matter interactions in the NWs using the exciton-polariton model. Zhang et al. also comprehensively investigated strong exciton–photon coupling in micro/nanowires system with low-threshold polariton lasing at room temperature [62]. As shown in Figure 3E, large-area CsPbX_3 NW laser arrays (length: ~ 10 –20 μm) were achieved by vapor growth method with low threshold (~ 4 $\mu\text{J}/\text{cm}^2$) and high quality factor (~ 2256) under excitation by 400 nm, 100 fs and 1 kHz laser [56]. The room-temperature Rabi splitting energy determined by coupling strength in these NWs were 210 ± 13 , 146 ± 9 and 103 ± 5 meV for the CsPbCl_3 , CsPbBr_3 , and CsPbI_3 NWs, respectively. Zhu et al. further realized the CW lasing in CsPbBr_3 NWs with a threshold of ~ 6 kW/ cm^2 (Figure 3F) [57]. A vacuum Rabi splitting of 0.20 ± 0.03 eV was achieved from the polariton modes near the bottleneck region on the lower polariton branch. These findings suggested that lead halide perovskite NWs have great potential in the production of excellent and low-threshold coherent light sources.

Except for the NWs, other nanostructures of perovskites, such as the micro/nanorods and cubes were also naturally F–P cavities with optical gain. Zhou et al. realized the effective F–P cavities in CsPbX_3 micro/nanorods with lengths ranging from ~ 2 –20 μm with low thresholds of (~ 14.1 $\mu\text{J}/\text{cm}^2$), high quality factor (~ 3500) and tunable

lasing wavelength (428–668 nm) under 360 nm laser with 100 fs and 1 kHz (Figure 4A) [63]. Subsequently, Wang et al. reported the multiphoton-pumped lasing from CsPbX_3 nanorods with thresholds of ~ 0.6 and 1.7 mJ/ cm^2 under excitations of 800 and 1200 nm with 80 fs and 1 kHz [65]. Meanwhile, Hu et al. demonstrated F–P lasing in CsPbBr_3 microcubes with high quality factor of 1150 under optical excitation (800 nm, 35 fs, 1 kHz), as described in Figure 4B [64]. The microcube cavities were synthesized by a solution process with a side length of ~ 500 nm; tunable ASE and enhanced stability were observed. Further, Liu et al. succeeded in reducing the laser size to subwavelength scale in three dimensions using an individual CsPbBr_3 perovskite nanocuboid with only ~ 400 nm³, as depicted in Figure 4C [33]. Although such a small size can introduce relatively large optical losses, the single-mode F–P lasing at room temperature has been achieved with low laser thresholds of 40.2 and 374 $\mu\text{J}/\text{cm}^2$ under 400 and 800 nm excitation, respectively, with the corresponding high quality factors of 2075 and 1859. The physical volume of the CsPbBr_3 nanocuboid laser is only ~ 0.49 λ^3 . The 3D subwavelength size, excellent stable lasing performance, frequency up-conversion ability and temperature-insensitive properties of nanocuboids were also demonstrated, which may lead to a miniaturized platform for nanolasers and integrated on-chip photonic devices in the nanoscale. Moreover, Mi et al. and Yang et al. demonstrated the F–P lasing in MAPbBr_3 (edge length ~ 8 μm) and CsPbI_3 (bottom side ~ 4 –10 μm) triangular pyramid with low thresholds (~ 92 $\mu\text{J}/\text{cm}^2$, 53.15 $\mu\text{J}/\text{cm}^2$ at 223 K) and narrow FWHM (~ 0.63 nm, < 0.5 nm), respectively [66, 67].

3.2 Perovskite WGM lasers

Different from the F–P cavity between two mirrors, the WGM laser is another type of cavity in which the light propagates as a ring to form travelling wave amplification. The interface between the gain medium and air can provide total internal reflection due to the large difference between their refractive indexes. In 2014, Zhang et al. first demonstrated the perovskite WGM laser from hybrid organic-inorganic perovskite $\text{MAPbI}_{3-x}\text{X}_x$ nanoplatelets, which they fabricated using a two-step vapor-phase deposition method [13]. As shown in Figure 5A, the triangular or hexagonal shapes can naturally serve as WGM cavities. A typical MAPbI_3 ($\text{MAPbI}_{3-x}\text{X}_x$) triangular (hexagonal) nanoplatelet with a thickness of ~ 150 nm and an edge length of ~ 32 μm realized the near-infrared lasing with a threshold of ~ 37 $\mu\text{J}/\text{cm}^2$ (~ 128 $\mu\text{J}/\text{cm}^2$), quality factor of ~ 650 (~ 900), and FWHM of ~ 1.2 nm (~ 0.9 nm) when they were

Figure 4: The Perovskite microrod and microcube lasers.

(A) Left: The triangular rod emission spectra around the lasing threshold. Inset: The optical images of a single triangular rod. Right: The wavelength-tunable lasing at room temperature from the CsPbX_3 nanorod lasers. Reproduced with permission [63]. Copyright © 2017, American Chemical Society. (B) The intensity-dependent emission spectra from CsPbBr_3 microcubes around the lasing threshold. Inset: A photograph of a single CsPbBr_3 microcube above the lasing threshold. Reproduced with permission [64]. Copyright © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (C) Left: The SEM image of the CsPbBr_3 nanocuboids. The scale bar is 500 nm. Middle: Schematics of the crystalline structure (below) and stand-wave in F-P cavity (upper). Right: The pump intensity-dependent emission spectra from a single CsPbBr_3 nanocuboid under two-photon excitation. Reproduced with permission [33]. Copyright © 2018, American Chemical Society.

pumped by a 400 nm, 50 fs and 1 kHz laser. Furthermore, the output lasing mode can be tuned to red-shift by different edge lengths from 28 to 47 μm owing to the intrinsic self-absorption of the excitons. Most remarkably, the WGM nanolaser can be easily integrated onto conductive platforms (Si, Au, ITO and so forth). These results suggested the potential of perovskite WGM nanolasers as suitable on-chip integrated laser sources.

In contrast, Liao et al. used the one-step self-assembly method and reported the square microdisk laser from MAPbBr_3 with a side length of $\sim 1\text{--}10\ \mu\text{m}$ and thickness of 0.1–0.25 μm [68]. As shown in Figure 5B, single-mode lasing at 557.5 nm was achieved in a $2.0 \times 2.0 \times 0.6\ \mu\text{m}^3$ square MD with a threshold of $\sim 3.6\ \mu\text{J}/\text{cm}^2$ and quality factor of 430, under excitation source of 400 nm, 150 fs and 1 kHz. Based on the side length of the MDs, the lasing mode can be tuned from single- to multi-mode. Figure 5C displays the WGM laser in the cross-sections of the MAPbBr_3 micro-rods with widths ranging from a few hundred nanometers to a few microns [69]. With pulsed excitation (400 nm, 100 fs, 1 kHz), the smallest lasing FWHM at the threshold is around 0.1 nm, giving a quality factor over 5000. In contrast, due to much better stability than hybrid perovskites, the all-inorganic perovskite CsPbX_3 has gained

great research attention as well. In 2016, Zhang et al. prepared high-quality, single-crystalline CsPbX_3 NPs by using the vapor phase van der Waals epitaxy method and first demonstrated the all-inorganic WGM laser [47]. The laser threshold was as low as $\sim 2.0\ \mu\text{J}/\text{cm}^2$ under 400 nm excitation with 50 fs and 1 kHz. As shown in Figure 5D, the lasing emission can cover the whole visible regime and the lasing FWHM was narrowed down to $\sim 0.14\ \text{nm}$. The excellent lasing performance can further promote the nanophotonic applications of the WGM.

The optical losses are inevitable due to the field leakage from the cavity facet and edge scattering, which can induce relatively high pump intensity and low quality factor. Changing the types of WGM microcavities is an effective method to improve lasing performance. Similar with NPs, spheres can also naturally form the WGM cavity. Meanwhile, Tang et al. prepared CsPbX_3 submicron spheres ($\sim 0.2\text{--}10\ \mu\text{m}$) by dual-source chemical vapor deposition method and realized tunable single-mode lasing from 425 to 715 nm under optical excitation of 400 nm, 40 fs and 1 kHz, as shown in Figure 5E [70]. The lasing was demonstrated in an individual sphere with a diameter of around 1.0 μm and showed a low threshold of $\sim 0.42\ \mu\text{J}/\text{cm}^2$ and a high quality factor of ~ 6100 with narrow FWHM

Figure 5: The perovskite WGM lasers.

(A) Top: The schematic of the hexagonal NP laser. Bottom: The lasing spectrum from NP. Insets: The optical images of NPs. Right: The lasing spectra of the perovskite NP laser devices on mica, Si, ITO and Au from the bottom to top panels. Reproduced with permission [13]. Copyright © 2014 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (B) Left: The emission spectra of a square microdisk with pump intensity. Inset: The locally amplified spectra with distinct blue shift. Right: The optical images of a microdisk under different pump intensity. Reproduced with permission [68]. Copyright © 2015 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (C) Left: The SEM image of the big “cross” in MAPbBr₃ microrod. The area under the yellow dashed rectangular box is the optical pumping area. Right: The laser spectrum and fitted FWHM at the threshold. Reproduced with permission [69]. Copyright © 2017, Royal Society of Chemistry. (D) Left: The lasing spectra and emission images of individual CsPbX₃ perovskite nanoplatelets with different halide ions. Right: The zoom-in spectrum of a lasing mode of CsPbBr₃I_{x-3-x} with FWHM of 0.14 nm. Reproduced with permission [47]. Copyright © 2016 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (E) Left: The schematic of an individual CsPbBr₃ microsphere on silicon substrate pumped by a 400 nm laser excitation. Right: The excitation power-dependent lasing spectra from a single CsPbBr₃ microsphere. Inset: The PL image of microsphere above the lasing threshold. Reproduced with permission [70]. Copyright © 2017, American Chemical Society.

(~0.09 nm). They further improved the quality factor to 15,000 with FWHM of 0.037 nm. They concluded that the ultrahigh quality factor can be attributed to the regular spherical structure with smooth surface, the high gain and high spontaneous emission coupling efficiency [71]. These high-performance perovskite WGM lasers have huge potential to pioneer a new field of ultracompact, low-threshold and high quality factor laser sources for photonic integrated systems.

3.3 Perovskite random laser

Unlike regular lasers, random lasers do not rely on additional resonators, thus ensuring low production cost and simple technological requirement. In a random laser, the optical cavity is absent, but multiple scattering between

particles in the disordered gain medium can keep the photons from travelling long enough for the efficient amplification. Figure 6A presented a schematic diagram of the random laser principle [72]. In 2014, Dhanker et al. first reported a random laser under an optical pump (355 nm, 0.8 ns, 1 kHz) in the MAPbI₃ perovskite microcrystal networks [73]. The laser threshold was less than 200 μJ/cm² and the FWHM was less than 0.5 nm (Figure 6B). Moreover, the spectral mode was unevenly distributed under different power excitations and large areas of microcrystals were excited. In the same year, Kao et al. also observed random lasers due to polycrystalline boundaries scattering in the MAPbI₃ perovskite thin film synthesized by the solution [77].

In 2016, Liu et al. observed the random laser in self-assembled MAPbBr₃ perovskite nanoparticles with a threshold of 60 μJ/cm² and quality factor of 500 [78].

Figure 6: The perovskite random laser.

(A) The schematic diagram of the random laser principle. Reproduced with permission [72]. Copyright © 2000, Springer Nature. (B) Left: The emission spectra collected below, near and above lasing threshold. Right: The micro-PL images showing the spatial distribution of the emission at a certain pump intensity. Reproduced with permission [73]. Copyright © 2014, AIP Publishing. (C) Left: The tunable ASE via compositional modulation. Right: The random lasing from CsPb(Br/Cl)₃ nanocrystal film. Inset: The path-length distribution averaged over 256 pump laser shots. Reproduced with permission [18]. Copyright © 2015, Springer Nature. (D) Random laser from the CsPbBr₃ QDs/A-SiO₂. Inset: The isolation effect of QDs on the silica sphere surface. Reproduced with permission [74]. Copyright © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (E) The schematic illustration of the excitation and emission of a flexible thin film perovskite laser at a bending condition. Reproduced with permission [75]. Copyright © 2019, American Chemical Society. (F) The intensity-dependent emission spectra of CsPbBr₃ QDs/TiO₂. Reproduced with permission [76]. Copyright © 2019, Optical Society of America Publishing.

When the excited position changed slightly, all laser characteristics changed correspondingly, thus confirming that the laser was formed by the scattering between internal nanoparticles rather than the overall boundaries. Further theoretical simulation and experimental results also verified this gain source of multiple scattering. Subsequently, Shi et al. observed random lasers in MAPbI₃ films with a threshold of 102 $\mu\text{J}/\text{cm}^2$, thickness of 450 nm and particle size of ~ 200 nm; the lasing mode of the MAPbI₃ films changed when detected at different angles [79]. This can be attributed to the high optical gain and strong

multiple scattering provided by the polycrystalline grain boundaries.

Compared to the hybrid organic-inorganic perovskite, the CsPbX₃ of bulk materials has been studied as early as 1958 [80]. Meanwhile, the development of all-inorganic CsPbX₃ perovskite colloidal quantum dots is relatively later than this. In 2015, Protesescu et al. synthesized for the first time some high-quality CsPbX₃ quantum dots [27] with a quantum efficiency of up to 90% and a spectrum covering around 410–700 nm. Yakunin et al. first realized the all-inorganic CsPbX₃ random

laser with narrow line width (0.14 nm) and tunable ASE spectra (440–700 nm) from perovskite nanocrystal thin film under laser excitation of 400 nm, 100 fs and 1 kHz (Figure 6C) [18]. The optical gain coefficient of the perovskite CsPbBr_3 was as high as $450 \pm 30 \text{ cm}^{-1}$, and its ASE threshold was less than $5 \mu\text{J}/\text{cm}^2$. Furthermore, the lasing mode was fully stochastic due to the unique and irreproducible path. In 2017, Li et al. reported the random laser formed by amino-mediated anchoring perovskite quantum dots on the surface of monodisperse SiO_2 spheres by one-step synthesis (Figure 6D) [74]. By dispersing the quantum dots on the SiO_2 sphere, the “aggregation induced quenching” of the quantum dots was effectively suppressed and their spatial distribution on the SiO_2 sphere was regulated to form a suitable light scattering loop. This led to the formation of a low threshold ($\sim 40 \mu\text{J}/\text{cm}^2$) and highly stable random emission under the optical pump (400 nm, 100 fs, 1 kHz).

Meanwhile, the use of disordered gain materials has emerged as a strategy for fabricating random laser sources. Recently, Wang et al. demonstrated a random laser in curved perovskite MAPbBr_3 thin films on flexible substrates by using the solvent-engineered method (Figure 6E) [75]. Under 355 nm excitation with 0.5 ns and 1 kHz, the lasing threshold was $2.5 \text{ mJ}/\text{cm}^2$ with emission FWHM of $\sim 1.8 \text{ nm}$, which can be further reduced by increasing the local curvature of the film arising from the variation of the scattering strengths of the bent thin film. Meanwhile, the curved perovskite lasers can be extremely robust with repeated deformations. Moreover, Tang et al. reported a successful room-temperature up-conversion random lasing by distributing uniformly the CsPbBr_3 QDs into TiO_2 nanotubes, as depicted in Figure 6F [76]. Under 800 nm and 35 fs laser excitation, the ASE and random lasing showed thresholds of $2.33 \text{ mJ}/\text{cm}^2$ and $9.54 \text{ mJ}/\text{cm}^2$, respectively. Due to the small diameter of the nanotubes, the lasing emission of the $\text{CsPbBr}_3/\text{TiO}_2$ film was assigned to the random laser with FWHM of $\sim 0.49 \text{ nm}$. These results indicated that the random lasers can be expected to be adopted in practical applications in durable speckle-free light sources for various applications, including spectroscopy, bioimaging, medical diagnosis and illumination.

4 Perovskite lasers with external cavity

Aside from the above-mentioned active cavities, the perovskites can serve as a gain medium only, and couple

with other external optical components, including DBR, distributed Feedback (DFB), PC and so on, to form laser devices. Xing et al. first reported in 2014 the initial stimulated emission for the ASE of the MAPbX_3 perovskite thin film, which they synthesized through the low-temperature solution process [10]. The pristine, 65 nm-thick film underwent photo-excitation by 600 nm, 150 fs and 1 kHz; it showed a low threshold of $\sim 12 \mu\text{J}/\text{cm}^2$ with large absorption coefficient of $\sim 5.7 \times 10^4 \text{ cm}^{-1}$ and tunable ASE peaks ranging from 390–790 nm. This cavity-free configuration of perovskites can manifest the ability of photon amplification and can be embedded into a wide range of cavity resonators for the realization of on-chip coherent light sources.

4.1 Perovskite VCSEL and DFB lasers

In 2014, Deschler et al. demonstrated the first perovskite F–P mode vertical laser based on DBR [81]. The laser cavity consisted of a middle gain layer of hybrid perovskite $\text{MAPbI}_{3-x}\text{Cl}_x$ thin film with 500 nm thickness, a top layer of PMMA and Au, and a bottom layer of DBR (Figure 7A). Under the excitation of 532 nm and 0.4 ns pulse width, the lasing threshold was as low as $0.2 \mu\text{J}/\text{pulse}$. Notably, the luminescence in the perovskite $\text{MAPbI}_{3-x}\text{Cl}_x$ film was ascribed to the bimolecular-free carrier electron–hole recombination by TAS. Chen et al. succeeded in micro-fabricating a VCSEL by embedding the perovskite MAPbI_3 thin film ($\sim 300 \text{ nm}$) into two high-reflectivity ($\sim 99.5\%$) DBRs [19]. The low threshold ($\sim 7.6 \mu\text{J}/\text{cm}^2$) and high quality factor (~ 1100) vertical laser was achieved under optical excitation at 532 nm with 340 ps and 1 kHz. Further optimization was carried out by using other lead halide precursors or by changing the organic cation. They subsequently used the FAPbBr_3 and $\text{Cs}_{0.17}\text{FA}_{0.83}\text{PbBr}_3$ as gain layers together with the optimized DBR structures, and good lasing performance was demonstrated with the following characteristics: high quality factors of 1420 and 1350; low thresholds of 18.3 and $13.5 \mu\text{J}/\text{cm}^2$, respectively; and the corresponding stable lifetimes of 20 h and $>35 \text{ h}$ [85, 86]. Room temperature CW pumped VCSEL-based MAPbBr_3 and MAPbI_3 were reported at pump thresholds of 89 and $13 \text{ W}/\text{cm}^2$, respectively [87, 88]. This finding is a huge step towards achieving more efficient CWs and electric pumped perovskite lasers.

In 2017, Wang et al. realized the first all-inorganic perovskite VCSEL [82]. As shown in Figure 7B, the high reflective dielectric mirror was composed of 25 pairs of $\text{SiO}_2/\text{TiO}_2$ quarter-wave layers, and the all inorganic perovskite CsPbBr_3 nanocrystals gain layer was sandwiched

Figure 7: Perovskite VCSEL and DFB lasers.

(A) The emission spectra of MAPbI_{3-x}Cl_x with different pump fluence. Inset: The vertical microcavity structure. Reproduced with permission [81]. Copyright © 2014, American Chemical Society. (B) Left: The structure of the VCSEL with the CsPbBr₃ nanocrystals. Right: The blue (up) and red (bottom) laser emissions from the CsPb(Br/Cl)₃ and CsPb(I/Br)₃ VCSEL, respectively. Reproduced with permission [82]. Copyright © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (C) The CsPbBr₃ thin film VCSEL with a FWHM of 0.07 nm. Inset: The far-field beam characteristics of the VCSEL above threshold. Reproduced with permission [83]. Copyright © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (D) Left: The schematic diagram of a DFB cavity. Right: The emission fluence-dependent PL spectra of the MAPbI₃ DFB cavity. Reproduced with permission [84]. Copyright © 2016 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (E) Left: The structure of the perovskite CsPbBr₃ DFB laser. Right: The laser spectrum from the DFB structure with a line width of 0.14 nm. Inset: The photograph of far-field emission profile. Reproduced with permission [83]. Copyright © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

between two DBRs. Under the excitation of the fs laser (400 nm, 100 fs, 1 kHz), the thresholds of the F–P mode laser were as low as ~19, ~9 and ~25.5 μJ/cm² for the stable red, green and blue lasing, respectively. Recently, Pourdavoud et al. implemented the CsPbBr₃ thin film featuring large crystals with micrometer lateral extension as a gain layer to demonstrate the high performance VCSEL [83]. The threshold was only ~2.2 μJ/cm² and the FWHM was narrowed down to 0.07 nm (Figure 7C). In contrast to the conventional photon laser, the lasing in a strong coupling cavity, such as VCSEL, could occur without the need for population inversion due to fact that the coherent light emission originated from the steady-state leakage of an exciton-polariton condensate [89]. Exciton-polaritons are bosonic quasiparticles that exist inside semiconductor microcavities; these consist of a superposition of an exciton and a cavity photon rather than the independent eigenmodes of the system. Su et al. reported the experimental realization of room-temperature polariton lasing with threshold of ~12 μJ/cm² based on an epitaxy-free CsPbCl₃ perovskite nanoplatelet microcavity [90]. A 373 nm-thick nanoplatelet was embedded in the bottom

and top DBRs made of 13 and 7 HfO₂/SiO₂ pairs, respectively. The superlinear power dependence, macroscopic ground-state occupation, blueshift of the ground-state emission, narrowing of the line width and the buildup of the long-range spatial coherence verified the polariton lasing. They also demonstrated the capability of long-range nonresonantly excited polariton condensate flow at room temperature in a CsPbBr₃ perovskite microwire microcavity [91]. A microwire with a length of ~30 μm, a thickness of ~120 nm and a width of ~2 μm was embedded in two DBRs to form the microcavity. Under 400 nm laser (100 fs and 1 kHz) excitation, the threshold of the polariton lasing was as low as 0.8 μJ/cm². Such strong coupling polariton lasing can serve as ideal candidates for the commercialization of electrically driven laser devices.

In 2016, Saliba et al. first introduced Bragg gratings as reflective devices in perovskite lasers [84]. They deposited a 120 nm-thick organic-inorganic perovskite MAPbI₃ onto two gratings with different periods (380–420 nm) and achieved low threshold lasing (~0.32–2.11 μJ/cm²) and tunable lasing (770–793 nm) under the excitation of

a 532 nm, 1 ns, and 1 kHz laser. The DFB single-mode laser provides a good idea for the development of all-electrically pumped perovskite lasers, thus demonstrating the potential commercial application of perovskite lasers. In 2017, Jia et al. first realized the CW-pumped MAPbI₃ DFB laser with threshold 17 kW/cm² for over an hour and at temperatures below the tetragonal-to-orthorhombic phase transition ($T < 160$ K) [92]. They deposited the MAPbI₃ film onto a period grating etched into an alumina layer on a high-thermal-conductivity sapphire substrate to avoid the thermal damage. They reported the continuous gain originating from tetragonal-phase inclusions, which were photogenerated by the pump within the bulk orthorhombic host matrix on a submicrosecond timescale. Soon, Li et al. reported room-temperature CW lasing action in a surface-emitting MAPbI₃ perovskite DFB laser on a silicon substrate [88]. With the thermal nanoimprint lithography technique, the perovskite thin film was directly patterned into a high-Q cavity with large mode confinement and enhanced emission intensity. Consequently, the ultralow lasing threshold of 13 W/cm² with narrow FWHM of ~ 0.7 nm was achieved, thus demonstrating a major step toward electrically pumped perovskite lasing. In 2019, Pourdavoud et al. used a DFB structure, which was formed by a recrystallized perovskite CsPbBr₃ thin film, to achieve a low threshold (7.2 μ J/cm²) and narrow FWHM (0.14 nm) laser emission (Figure 7E) [83]. PC refers to an artificial periodic dielectric structure with photonic band gap characteristics and has a wavelength selection function. A typical 1D PC uses the DBR or DFB structure to hold a perovskite in the middle in order to form a simple F–P resonant cavity, thereby achieving a single-mode and tunable laser. In 2016, Chen et al. embedded hybrid perovskite MAPbI₃ thin films, which they synthesized via solution-processed method into the 2D PC [20]. When the pump density reached 68.5 ± 3.0 μ J/cm², a single-mode lasing was achieved at 788.1 nm with a FWHM of 0.24 nm and power conversion efficiency of $13.8\% \pm 0.8\%$. Furthermore, the single-mode laser output can be tuned by changing the PC pitch range. Meanwhile, Schünemann et al. reported 3D PC DFB laser in perovskite MAPbBr₃, which they prepared by using the all-solution process [93]. Under the excitation of pulsed laser (532 nm, 0.5 ns, 2 kHz), they achieved highly stable lasing with a threshold of 1.6 mJ/cm² and FWHM of 0.15 nm.

4.2 Microsphere and microcapillary for perovskite lasers

Apart from the vertical cavity, the perovskites can be coupled with sphere and microcapillary as the WGM laser

resonator. In 2014, Sutherland et al. deposited a thin layer of MAPbI₃ film onto a silica sphere via the atomic-layer deposition technique and successfully achieved WGM lasing at 80 K (Figure 8A) [17]. Under 355 nm excitation with 2 ns and 100 Hz, the threshold of ~ 75 μ J/cm² and quality factor of ~ 1000 were achieved. Similarly, Yakunin et al. demonstrated the WGM laser from CsPbX₃ nanocrystal with narrow FWHM (~ 0.15 – 0.20 nm) using silica microspheres with a diameter of ~ 15 μ m as high-finesse resonators (Figure 8B) [18]. The feasibility of lasing in perovskite films indicates the high-quality optical gain in perovskites. Almost simultaneously, Wang et al. also observed the WGM laser from the CsPbBr₃ QDs, as depicted in Figure 8C [94]. After the evaporation of the solvent, they filled the CsPbBr₃ QDs into a capillary tube with solid film around the inner wall. The capillary tube with the inner diameter of ~ 50 μ m provided optical feedback, after which multi-mode WGM lasing was demonstrated with threshold of ~ 11 mJ/cm² under excitation at 400 nm with 5 ns and 20 Hz. They also observed the low-threshold, wavelength-tunable and stable ASE from the CsPbX₃ perovskite QDs, which could be ascribed to the biexciton emission.

Xu et al. incorporated the perovskite CsPbBr₃ nanocrystals into optical resonators of glass microcapillary tubes with diameter of ~ 60 μ m (Figure 8D) [32]. Under 800 nm fs excitation, the lasing threshold was around 900 μ J/cm² with FWHM of ~ 0.15 – 0.3 nm, indicating the quality factors of lasing modes to be in the range of 1700–3500. Such high quality factors could be ascribed to the high refractive index contrast between the glass tube (1.46) and nanocrystal solids (~ 2.30), which can well-confine the light emission from nanocrystals supporting the WGM oscillation. Liu et al. embedded the CsPbBr₃ QDs into the sub-micro silica sphere to improve the stability and observed the compounded mode of random and WGM under 800 nm excitation with 35 fs and 1 kHz [95]. Then, they incorporated the CsPbBr₃-SiO₂ spheres into a microtubule with a diameter of ~ 40 μ m, as shown in Figure 8E. The multi-mode lasing with a threshold of 430 μ J/cm² and FWHM of ~ 0.35 nm was sustained for over 140 min under continuous excitation. The lasing mode was analyzed by the WGM model. Furthermore, they also coupled the FAPbBr₃ nanocrystals into a hollow capillary tube and demonstrated the WGM laser with a low threshold of ~ 310 μ J/cm² under same pump condition [37]. Kurahashi et al. reported the MAPbBr₃ WGM laser in microcapillary with different inner diameters of 2–40 μ m [97]. Under excitation at 397 nm with 200 fs and 1 kHz, the mode number and the lasing threshold fluence decreased with reduced inner diameter. Notably, the single mode was realized at a

Figure 8: The perovskite microsphere and microcapillary laser.

(A) Left: The schematic of the laser emission of MAPbI_3 perovskite-coated microsphere. Right: The spectra of the MAPbI_3 perovskite-coated microsphere laser emission at 80 K for three different pump fluences. Reproduced with permission [17]. Copyright © 2014, American Chemical Society. (B) The emission spectra with different pump intensity. Inset: A microsphere resonator covered by a film of the CsPbBr_3 nanocrystals. Reproduced with permission [18]. Copyright © 2015, Springer Nature. (C) The pump intensity-dependent PL spectra from the CsPbBr_3 QDs microcapillary laser. Left inset: The suddenly increasing emission at the pump threshold. Right inset: The optical images of the CsPbBr_3 QDs capillary tube cavity below and above the lasing threshold. Reproduced with permission [94]. Copyright © 2015 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (D) The two-photon-pumped PL spectra of the CsPbBr_3 nanocrystals microcapillary laser. Inset: The emission intensity as a function of excitation fluence and threshold can be calculated to be $\sim 900 \mu\text{J}/\text{cm}^2$. Reproduced with permission [32]. Copyright © 2016, American Chemical Society. (E) Left: The lasing image of a cylindrical microtube incorporated with $\text{CsPbBr}_3/\text{SiO}_2$ QDs. Inset: The schematic of WGM cavity with the micro-ring resonator. Right: The emission spectra with increasing pump intensity. Reproduced with permission [95]. Copyright © 2019, Optical Society of America Publishing. (F) Intensity-dependent emission spectra from $\text{CsPbBr}_3/\text{CdS}$. Inset: The 3D schematic representation of core/shell structured $\text{CsPbBr}_3/\text{CdS}$ QDs. Reproduced with permission [96]. Copyright © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

pump threshold at $\sim 4.7 \mu\text{J}/\text{cm}^2$ in a microcapillary cavity with a diameter of $2 \mu\text{m}$.

Recently, Tang et al. succeeded in preparing a single core/shell structured perovskite semiconductor QDs with $\text{CsPbBr}_3/\text{CdS}$, which exhibited ultrahigh chemical

stability and nonblinking photoluminescence with high PLQY due to the reduced electronic traps within the core/shell structure [96]. Lasing action was demonstrated from the core/shell perovskite QDs in a cylindrical microcapillary with a threshold of $87 \mu\text{J}/\text{cm}^2$ and FWHM of $\sim 0.44 \text{ nm}$.

These flexible and changeable perovskites lasing can simplify the requirements of microcavities and enable the perovskite to be used in versatile photoelectric integrated devices.

4.3 Perovskite laser array

The high-density laser arrays have great potential for compact on-chip optoelectronic circuit integration. The nanomaterial arrays highly depend on the nanofabrication and template technology. In 2016, Liu et al. utilized a novel bottom-up growth technology for synthesizing high-quality patterned perovskite arrays on Si with pre-patterned single layer hexagonal boron nitride (h-BN) buffer layer [98]. The fabricated MAPbI₃ perovskite microplatelet arrays displayed a hexagon with an edge length of ~15 μm and a natural WGM cavity. Under optical pump by 400 nm laser excitation (~50 fs, 1 kHz), the multi-mode WGM lasing was realized with a pump threshold of ~11 μJ/cm² and FWHM of ~0.64 nm, corresponding to a quality factor of 1210. Notably, modal selectivity for single-mode lasing could be achieved with different cavity sizes or by simply breaking the structural symmetry of the cavity through designing the pattern.

Wang et al. successfully achieved high density nanolaser arrays by positioning a MAPbBr₃ perovskite microwire onto a silicon grating with threshold of ~6 μJ/cm² [99]. The transverse single-crystalline microwire were divided by a silicon grating into tiny subunit with the smallest subunit period of 800 nm. Only the suspended parts could hold high quality resonances and generate laser emissions. They further transferred the MAPbBr₃ nanoribbon onto a gold grating and successfully achieved laser arrays with threshold of 2.5 μJ/cm², as shown in Figure 9B [100]. Notably, the smallest size of the gap for light confinement is optimized to around 350–400 nm.

Liu et al. developed a large-scale perovskite MAPbX₃ nanowire array by combining the “top-down” fabricated polydimethylsiloxane rectangular groove template with the “bottom-up” solution self-assembly [101]. As exhibited in Figure 9C, not only the dimensions of individual NWs (width 460–2500 nm; height 80–1000 nm, and length 10–50 μm) but also the interwire distances could be precisely adjusted. Under the excitation of pulsed laser with wavelength of 400 nm (150 fs, 1 kHz), the nanowire laser was achieved with almost identical optical modes and similarly low-lasing thresholds from 9.5 to 12.7 μJ/cm², together with good photostability of an operation duration exceeding 4×10^7 laser pulses. He et al. adopted the polydimethylsiloxane cylindrical-hole-template confined

solution-processed growth method to obtain the large-area CsPbX₃ microdisk laser arrays (up to 1×1 cm²) [102]. The microdisks were rectangle shapes with a side length of 2.5 ± 0.3 μm and a thickness of 0.6 ± 0.2 μm, supporting WGM cavity in Figure 9D. The lasing output wavelengths could be well tuned in the blue-green region from 425 to 540 nm by adjusting their compositions of CsPbCl_{3-x}Br_x from x=0 to 3, respectively, with similarly low lasing thresholds of 3–12 μJ/cm². Furthermore, they integrated the multicolored lasing arrays on the same substrate.

Duan et al. also fabricated well-controlled and uniform MAPbBr₃ microdisks by a simple solution-based one-step anti-solvent method [103]. The size and mode numbers of the self-assembled microdisk cavities could be well adjusted by modifying the sizes of the SiO₂ microdisks on the substrate or changing the concentration of perovskite precursor. Recently, Zhizhchenko et al. proposed a laser-printed perovskite MAPbBr₃I_y microdisks by laser ablation of glass films directly using a donut-shaped femtosecond laser beam, as shown in Figure 9E [16]. The prepared microdisks were 760 nm thickness and diameters ranging from 2 to 9 μm precisely controlled by a topological charge of the vortex beam. The tunable lasing from different compositions could be realized from 500 to 800 nm under optical excitation at a wavelength of 532 nm (0.56 ns, 1.5 kHz) with a quality factor up to 5500. These results would further push the potential application of these perovskite-based lasers in fully integrated optoelectronic circuitry.

4.4 2D perovskite laser

The inherent poor stability of 3D perovskites resulting from their high moisture and heat sensitivity impedes their commercialization [104]. Perovskites with reduced dimensionality can promote the radiative recombination, improve the stability and enhance the exciton binding energy and PLQYs [28, 105–107]. Recently, scientists have demonstrated the great potential of quantum well (QW) quasi-2D Ruddlesden–Popper perovskites (RPPs) to enhance the stability and exciton binding energy by adopting the chemical formula of R₂M_{n-1}Pb_nX_{3n+1} with R as a long-chain ligand [106]. As shown in Figure 10A, the inorganic lead-halide layers are sandwiched between two hydrophobic organic layers formed by R bulky cations [108]. The large dielectric constant difference between the two layers can lead to a high exciton binding energy, thus indicating stable excitons at room temperature.

In 2018, Li et al. reported for the first time the intrinsic gain properties of 2D-RPP thin films by demonstrating

Figure 9: The perovskite laser array.

(A) Left: The SEM image of the as-prepared MAPbX₃ platelet array. Right: The intensity-dependent emission spectra of microplatelet laser. Inset: The 2D pseudo color plot of emission spectra under different pump fluence. Reproduced with permission [98]. Copyright © 2016 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (B) Left: The top-view SEM image of the nanoribbon onto a gold grating; the scale bars were 50 and 5 μm , respectively. Right: The laser spectra from nanoribbons with different pumping densities. Reproduced with permission [100]. Copyright © 2017, American Chemical Society. (C) The MAPbBr₃ perovskite nanowire laser emission and laser arrays (insets). Reproduced with permission [101]. Copyright © 2017, American Chemical Society. (D) The intensity-dependent emission spectra of a microdisk laser. Inset: The optical image of the microdisk lasers above the lasing threshold. Reproduced with permission [102]. Copyright © 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (E) Left: The schematic of laser printing perovskite microdisks. Middle: The close-up false-color SEM image (top) as well as a photograph of a $1 \times 1 \text{ cm}^2$ array of perovskite microlasers. Right: The lasing spectra of the MAPbI₃ microdisk with diameter of 3.8 μm . Inset: The optical image of microdisk above the threshold. Reproduced with permission [16]. Copyright © 2019, American Chemical Society

Figure 10: The 2D perovskite laser.

(A) The crystal structures of the 2D perovskites with n from 1 to 4. Reproduced with permission [108]. Copyright © 2016, American Chemical Society. (B) The tunable ASE spectra from the $(\text{NMA})_2(\text{FA})\text{Pb}_2\text{Br}_{7-y}\text{I}_y$ thin films. Reproduced with permission [109]. Copyright © 2018 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (C) Left: The μ -PL image recorded above lasing threshold with distinct spatial interference patterns. Right: The intensity-dependent PL spectra recorded from microring 8 labeled in Left. Inset: The lasing spectrum above the threshold with a Gaussian fitting. Reproduced with permission [110]. Copyright © 2018 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (D) The pump-intensity-dependent PL spectra of a single MPL. Left inset: The optical image with scale bar of 5 μm . Right inset: The optical image of MPL above the lasing threshold. Reproduced with permission [111]. Copyright © 2018 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (E) The evolution of spontaneous emission to normalized lasing spectra under various excitation fluences for $n=3$ RPP. Right panels: The corresponding dark field images below and above the lasing threshold and simulated electric field distribution ($|\mathbf{E}|^2$) from the top to bottom panels, respectively. Scale bar: 5 μm . Reproduced with permission [112]. Copyright © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

the ASE behavior in a cavity-free configuration [109]. The self-organized multiple QWs 2D-RPP thin films were prepared by a solution process with gain coefficients as high as $>300 \text{ cm}^{-1}$. Under excitation at 400 nm with 150 fs and 1 kHz, room-temperature ASE was achieved at a low

pump threshold of $\sim 8.5 \text{ J/cm}^2$ from 75 nm-thick quasi-2D $(\text{NMA})_2(\text{FA})\text{Pb}_2\text{Br}_7$ thin film. The tunable ASE wavelengths from visible to near-infrared spectral range (530–810 nm) by adjusting the halide compositions were presented in Figure 10B. Remarkably, an energy cascade between the

QWs in the quasi-2D perovskites was revealed, which enabled an ultrafast energy transfer process from higher energy-bandgap QWs to lower energy-bandgap QWs to build up the population inversion.

Zhang et al. fabricated high-density, large-area microring arrays of 2D $(\text{BA})_2(\text{MA})_{n-1}\text{Pb}_n\text{Br}_{3n+1}$ RPPs for WGM cavities by using a facile PDMS template confined solution-processed method [110]. The microring exhibited smooth outer surfaces and regular cross-sections with a width of $\sim 2.5 \mu\text{m}$ and height of $\sim 1.6 \mu\text{m}$ along the diameter of $19 \mu\text{m}$. As a consequence, a multi-mode WGM laser was observed with high-quality factor of ~ 2600 and low threshold of $\sim 12.2 \mu\text{J}/\text{cm}^2$ under 400 nm, 150 fs and 1 kHz pulsed excitation, as shown in Figure 10C. Using a similar method, they further reported high-density large-area NW arrays in 2D-RPPs of $(\text{BA})_2(\text{FA})_{n-1}\text{Pb}_n\text{Br}_{3n+1}$ with width of $\sim 2 \mu\text{m}$, height of $\sim 420 \text{ nm}$ and length of $17 \mu\text{m}$ [113]. The NW laser arrays exhibited high photo-stability, high quality factor (~ 1800) with FWHM of 0.3 nm and excitation threshold of $27.2 \mu\text{J}/\text{cm}^2$ under a 400 nm laser pump.

Raghavan et al. reported the growth of high-quality, millimeter-sized single crystals of $(\text{BA})_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}$ by a slow evaporation at a constant-temperature solution growth [114]. Under the 374 nm pulse laser excitation with a 40 MHz and 55 ps, the multi-mode lasers were achieved in the inorganic layers 1, 2 and 3 with low thresholds of 2.85, 3.02 and $3.21 \mu\text{J}/\text{cm}^2$, respectively; FWHM values of 0.22, 0.5 and 0.4 nm, respectively; and corresponding lasing wavelengths of 522.3, 577.6, and 623.6 nm. The lasing modes could be assigned to the possible existence of some cavities or microdomains inside the crystals. Li et al. realized the solution-processed room-temperature microplatelet lasers from multilayered $(\text{OA})_2(\text{MA})_{n-1}\text{Pb}_n\text{Br}_{3n+1}$ RPPs [111]. Figure 10D showed the multi-mode microplatelet laser with threshold of $\sim 8.5 \mu\text{J}/\text{cm}^2$ and narrowed FWHM of $\sim 0.3\text{--}0.6 \text{ nm}$ pumped by a 400 nm laser excitation ($\sim 150 \text{ fs}$, 1 kHz). Simultaneously, highly stable low-threshold (down to $\sim 7.8 \mu\text{J}/\text{cm}^2$) linearly polarized lasing was achieved from the microplatelets by adjusting the layers. Combining the femtosecond microarea PL with TAS revealed that the mixed lower-dimensional layers of the RPPs can cause enhanced exciton and photon confinement in the higher-dimensional layers.

Liang et al. reported the lasing and loss mechanisms of homologous 2D $(\text{BA})_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}$ RPP micron-sized thin flakes, which they mechanically exfoliated from the bulk crystal [112]. Wavelength-tunable lasing was achieved from the large- n RPPs ($n \geq 3$) but not from small- n RPPs ($n \leq 2$) even down to 78 K. Figure 10E displayed the pump

dependent PL spectra of an $n=3$ RPP microflake (length: $\sim 11.2 \mu\text{m}$; width: $\sim 11.1 \mu\text{m}$; thickness: $\sim 150 \text{ nm}$) at 78 K, exhibiting an excitation threshold at $\sim 2.6 \mu\text{J}/\text{cm}^2$ pumped by 400 nm laser with 80 fs and 1 kHz. Temperature-dependent and time-resolved PL spectroscopy together with TAS revealed that the non-radiative recombination pathways were mainly induced by the Auger recombination and exciton-phonon interaction as a result of the n -dependent lasing behaviors. These 2D-RPPs possessing improved environmental stability, enhanced exciton confinement and solution processable application-level ability have great potential in the electrically driven semiconductor lasers.

5 Summary and challenges

The perovskite nanomaterials have received worldwide attention due to their excellent properties. Especially in the field of micro/nanolasers, perovskite-based ones have gone through rapid development and remarkable improvements. In less than 5 years, plenty of perovskite lasers have been reported, all exhibiting great potential as miniaturized coherent light source for integrated circuit. In this review, we have summarized the development and recent processes of 3D perovskite-based photonic lasers. These are divided into nanostructured perovskite lasers, such as NW, NP, nanocube and so on, and perovskite lasers with external cavity, such as DBRs, DFB, etc. Specifically, such advantages as low cost, large optical gain, high PLQY, long carrier lifetime, low trap state density, large exciton binding energy, tunable wavelength covering ultraviolet to the near-infrared regime and so on, all support the use of metal halide perovskites as a new class of strong emitters for a wide array of applications, such as laser display, optical interconnection, 3D sensing and imaging, etc. [16, 28, 102, 115, 116]. The excellent properties and flexible structures of perovskites also ensure their strong compatibility for integration.

The high-performance perovskite nanolasers have achieved low threshold, high quality factor and tunable wavelengths under optical excitation. The ultracompact volume could confine the light field in a small region and enhance the light-matter interactions. However, the current volumes of perovskite photonic lasers, and even some plasmonic lasers, still range from several to tens of microns only. Due to the optical diffraction limit, microcavities with physical size of less than 1 micron, especially for the wavelength scale, have rarely been reported. Simultaneously, the power density is inversely related

to the laser size. Nevertheless, the high optical gain and large absorption section of perovskites and their effective structural engineering provide an inspiring opportunity to realize subwavelength scale nanolasers for photoelectric integrated systems.

From the current research status, some challenges involving perovskites laser devices must be addressed. First, stable operation is the premise of practical application. The poor stability as well as the toxicity (Pb) of the perovskites under atmosphere are topics that have been studied for a long time. Encapsulation is an effective solution to improve the stability, but this could lead to the big device volume and low efficiency. The fabrication of high-quality perovskites and the substitution of Pb could thus be considered urgent topics for future works. Next, the photophysical properties of perovskites are especially crucial for the future optoelectronic applications. Discovering the exciton and carrier behaviors can contribute to controlling the Auger losses and charge transport for designing functional devices. Moreover, the compatibility and volume of devices can influence the high-density integrated circuit across the board.

Finally, the huge challenge of realizing electrically driven laser devices is impeding the broader practical prospect. Therefore, at the present stage, pushing the development of optical pumped lasing, especially for room-temperature continuous-wave pumped lasing, is essential, as this can provide a key step for electrically driven lasers. In the next decade, with the advancement of technology and more efforts from researchers worldwide, the high-performance, electrically driven perovskite lasers can be fully realized.

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