

Advances in metal halide perovskite lasers: synthetic strategies, morphology control, and lasing emission

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Abstract. In the past decade, lead halide perovskites have emerged as potential optoelectronic materials in the fields of light-emitting diode, solar cell, photodetector, and laser, due to their low-cost synthesis method, tunable bandgap, high quantum yield, large absorption, gain coefficient, and low trap-state densities. In this review, we present a comprehensive discussion of lead halide perovskite applications, with an emphasis on recent advances in synthetic strategies, morphology control, and lasing performance. In particular, the synthetic strategies of solution and vapor progress and the morphology control of perovskite nanocrystals are reviewed. Furthermore, we systematically discuss the latest development of perovskite laser with various fundamental performances, which are highly dependent on the dimension and size of nanocrystals. Finally, considering current challenges and perspectives on the development of lead halide perovskite nanocrystals, we provide an outlook on achieving high-quality lead perovskite lasers and expanding their practical applications.

Keywords: perovskite; nanocrystal; morphology; stimulated emission; laser.

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1 Introduction

Research related to perovskites can be traced back to 1970s,^{1–3} but systematic research was lacking due to technology limitations in that period. In 2009, Kojima et al.⁴ first added organic–inorganic hybrid perovskites as semiconductor materials in dye-sensitized solar cells, achieving a power conversion efficiency (PCE) of 3.8%. Since that breakthrough, the development of perovskites with large absorption coefficient, low defect state density, long carrier diffusion length, and bipolar carrier transport property has made them uniquely suitable for photovoltaic applications.^{4–16} Currently, the PCE of single-junction pure perovskite-based solar cells has reached 25.5% for small-area devices and 24.2% for large area over 1 cm².^{15,17} According to the Shockley–Queisser limit, a type of high-quality conversion

material in solar cells is also efficient luminescent materials in light-emitting devices such as LEDs and lasers.^{18–20} In 2004, the first evidence of optical gain in lead halide perovskites was reported, which is amplified spontaneous emission (ASE) from microcrystalline films of CsPbCl₃ recrystallized from the amorphous phase.^{21,22} In 2014, ASE and lasing were realized from MAPbX₃ polycrystalline thin films at room temperature. Ultralow threshold could benefit from the excellent optical absorption of MAPbX₃ with a coefficient greater than 2×10^4 cm⁻¹.^{23–27}

In addition, the research about micro/nanolasers based on perovskite with high coherence, low threshold, and high-quality factor has increased rapidly. The advances in lasing performance mainly benefit from the excellent optical properties such as high photoluminescence quantum yield (PLQY), narrow linewidth, large absorption coefficient, and widely tuned band.^{28–32} In addition, the shape and size of perovskite could be flexibly adjusted, which can affect their physical and chemical properties and the performance of optoelectrical devices.^{33–36} Hence, various

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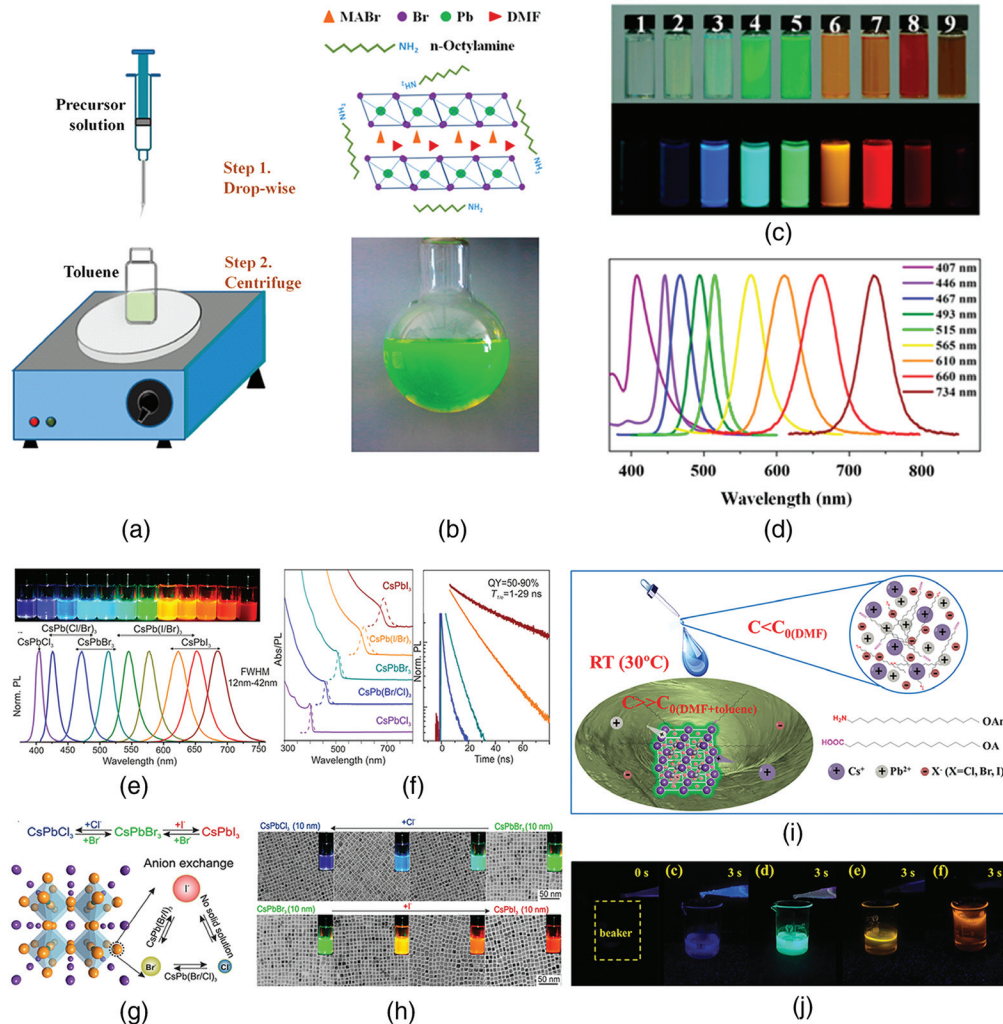


Fig. 2 (a) Schematic of LARP technique. Figures reproduced from Ref. 25. (b) Schematic of precursor and optical image of MAPbBr₃ solution. Figures reproduced from Ref. 25. (c) Optical images of MAPbX₃ solution under natural light and under 365 nm excitation. Figures reproduced from Ref. 25. (d) PL spectra of MAPbX₃ QDs. Figures reproduced from Ref. 25. (e) PL optical images and PL spectra of CsPbX₃ QDs. Figures reproduced from Ref. 42. (f) Time-resolved PL decays for CsPbX₃ QDs. Figures reproduced from Ref. 42. (g) Schematic of the anion-exchange of CsPbX₃. Figures reproduced from Ref. 41. (h) TEM images of CsPbX₃ QDs with various PL. Figures reproduced from Ref. 41. (i) Schematic of room-temperature fabrication of CsPbX₃ QDs. Figures reproduced from Ref. 39. (j) Optical images of CsPbX₃ QDs after the addition of precursor ion solutions for 3 s. Figures reproduced from Ref. 39.

the precursors, the authors tuned the emission in the range of 407 to 734 nm [Figs. 2(c) and 2(d)].²⁵ In the case of all-inorganic ones, the emission spectra of CsPbX₃ QDs fabricated by the high temperature method could be tunable over 410 to 700 nm with narrow half maximum of 12 to 42 nm and radiative lifetime of 1 to 29 ns [Figs. 2(e) and 2(f)].⁴² Subsequently, they proposed an anion-exchange process to tune the emission of colloidal CsPbX₃ QDs via postsynthetic reactions with different compounds [Figs. 2(g) and 2(h)].⁴¹ Besides the hot-injection technique, the room-temperature synthesis for perovskite QDs was also studied.³⁹ In 2016, Zeng and coworkers developed a room-temperature method to fabricate CsPbX₃ QDs via supersaturated recrystallization. In this process, the crystallization process occurred in the transform of Cs⁺, Pb²⁺, and X⁻ ions from soluble to insoluble solvents in the absence of inert gas within a few

seconds, as shown in Figs. 2(i) and 2(j).³⁹ Although crystallized at room temperature, these CsPbX₃ QDs held superior optical properties with PLQYs above 70%, and PLs can remain at ~90% after aging 30 days in the air. Except for being regulated by changing composition, the bandgap of perovskite QDs also can be tuned by the size regulation of QDs. Chen et al.⁶⁶ fabricated CsPbBr₃ QDs with an average diameter from 7.1 to 12.3 nm by modifying the temperature, and the corresponding PL emission peaks could be tuned from 493 to 531 nm. Fang et al.⁶⁷ synthesized MAPbBr₃ QDs with tunable average diameter from 2.82 to 5.29 nm by varying the additive amount of surfactant, and the corresponding PL emission peaks could shift from 436 to 520 nm due to the quantum confinement effect. Most recently, there have been much more researches about perovskite QDs by composition engineering for wider optoelectronic applications.⁶⁸⁻⁷⁰

2.3 Perovskite Nanowires and Nanorods

Perovskite 1D NWs and NRs are more applicable in the field of optoelectronic applications due to their special anisotropic structures. In the growth of 1D perovskite structures, reaction temperature, reaction time, and organic ligands are critical factors for crystallization.^{71–74} Deng et al.⁷¹ first fabricated MAPbI₃ NWs via the one-step solution method. In this process, the precursor solution containing PbI₂ and CH₃NH₃I was dropped onto a substrate and then heated at different temperatures. Finally, uniform MAPbI₃ NWs were obtained after heating at 80°C for 10 min. In 2017, they fabricated Cs_x(MA)_{1-x}PbI₃ NWs through a two-step solution method.⁷² As shown in Fig. 3(a), PbI₂ powder was dissolved in water at 75°C initially, then PbI₂ separated out when the solution cooled down to room temperature. With the addition of CsI and MAI, perovskite NWs could be formed after shaking for a few seconds. The length and diameter of obtained perovskite NWs could reach 10 μm and several hundred nanometers. Moreover, the amount of perovskite NWs was related to the concentration of PbI₂ separated out from aqueous solution.⁷² Zhu et al. developed a direct conversion of MAPbI₃ film into NWs through a recrystallization process [Fig. 3(b)]. The first step was the formation of perovskite film from a mixture of PbCl₂ and CH₃NH₃I.⁷³ Then, a mixture solution containing DMF and isopropyl was dropped onto the as-grown perovskite film. Along with the evaporation of the solvent, NWs could be formed [Fig. 3(b)].⁷³ Furthermore, they found the content of DMF in isopropyl, and the rotation speed could affect the sizes of prepared MAPbI₃ NWs.

As for all-inorganic perovskite, in 2015, Yang and coworkers used a solution method to synthesize single-crystalline CsPbX₃ NWs first. The reaction temperature was set as 150°C to 250°C.⁵¹ They found that the reaction time was critical to the growth of NWs. As shown in Fig. 3(c), the SEM of prepared CsPbBr₃ with different reaction times showed perovskite nanocubes formed initially, then NS and NW formed at 90 min [Fig. 3(c)].⁵¹ In the formation of Cs-based perovskite NWs, surface ligands could affect the width and size. Imran et al.⁵⁶ tuned the width of CsPbX₃ NWs from 10 to 20 nm by regulating the ratio of octylamine to oleylamine and varying the reaction time. They found that the width of NWs can be decreased below

~5 nm by introducing carboxylic acids with short aliphatic chains. Correspondingly, the emission spectra of CsPbBr₃ NWs could be tuned from 524 to 473 nm [Fig. 3(d)].⁵⁶ Amgar et al.⁵⁸ found that various hydrohalic acids (HX, X = Cl, Br, and I) affect the length of CsPbBr₃ NWs efficiently. With the increasing amount of HX, the length of NWs would be shortened [Fig. 3(e)]. Using this method, the emission of the CsPbBr₃ NWs could be tunable in the range of 423 to 505 nm [Fig. 3(f)].⁵⁸ CsPbBr₃ NWs/NRs can also be synthesized by a low-temperature method. Dong's group⁷⁴ fabricated CsPbBr₃ perovskite NRs with controllable size in a polymer matrix. Then, Liu et al. fabricated single-crystalline CsPbBr₃ NWs without inert gas at room temperature. By increasing the reaction time, the length of NWs could be increased from nanometers to micrometers, and the diameter could be tuned from 2.5 to 32.0 nm. Moreover, using this method, the emission spectra of CsPbX₃ NWs could be tuned from 434 to 681 nm.⁵⁷

Besides the above-mentioned solution-process, plenty of works have been reported on synthesizing perovskite NWs and NRs by vapor-phase growth.⁴⁹ More than ever, the vapor-phase process can control the morphology and crystalline phase of perovskite NCs efficiently. It has been demonstrated that the growth temperature and the substrates are critical for the orientation of perovskite NWs in vapor-growth. Xing et al.⁴⁹ first used the vapor-phase technique to fabricate perovskite NWs. First, PbI₂ NWs were deposited on SiO₂ substrates by the chemical vapor deposition (CVD) method [Fig. 4(a)].⁴⁹ Consequently, PbI₂ was converted into MAPbX₃ after the reaction with MAX through a CVD process. As shown in Fig. 4(b), the prepared MAPbI₃ wires had a length about tens of micrometers and a diameter of ~500 nm. In Figs. 4(c) and 4(d), they indicated that MAPbI₃ NW grows along the [100] direction.⁴⁹ Due to the thermal decomposition of organic hybrid perovskite occurring easily at high temperatures, direct vapor-phase growth of hybrid perovskites is more challenging. However, the vapor-phase technique is an attractive method for all-inorganic perovskites, which have better thermostability. Zhou et al.⁷⁵ prepared CsPbX₃ NRs with high crystallization quality and regular triangular morphology through a vapor deposition method [Fig. 4(e)]. As shown in the SEM image [Fig. 4(f)], prepared CsPbX₃ NRs had a triangular cross section, smooth surfaces, and a length of 2 to 20 μm. They

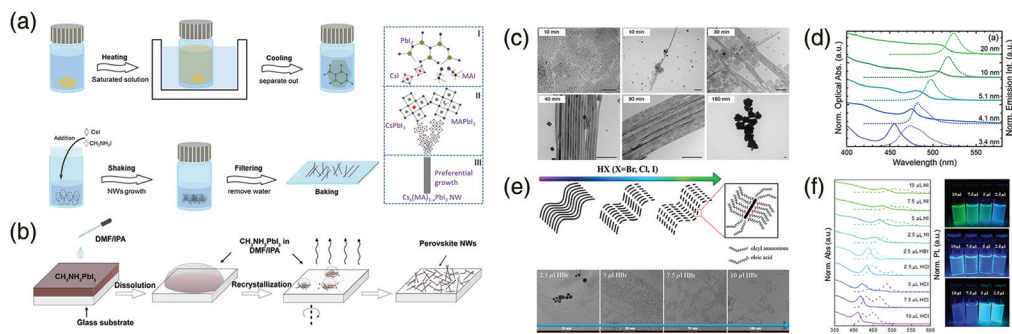


Fig. 3 (a) Schematic of the fabrication process for the Cs_x(CH₃NH₃)_{1-x}PbI₃ NWs. Figures reproduced from Ref. 72. (b) Schematic of the formation of the MAPbI₃ NWs by recrystallization process. Figures reproduced from Ref. 73. (c) TEM images of as-grown CsPbBr₃ NCs with increasing times. Figures reproduced from Ref. 51. (d) Absorption and PL spectra of CsPbBr₃ NWs. Figures reproduced from Ref. 56. (e) Schematic of the passivation effect by HX on the length of CsPbX₃ NWs and TEM images of the synthesized CsPbX₃ NWs. Figures reproduced from Ref. 58. (f) Normalized absorption, PL spectra, and photographs of CsPbX₃ NWs. Figures reproduced from Ref. 58.

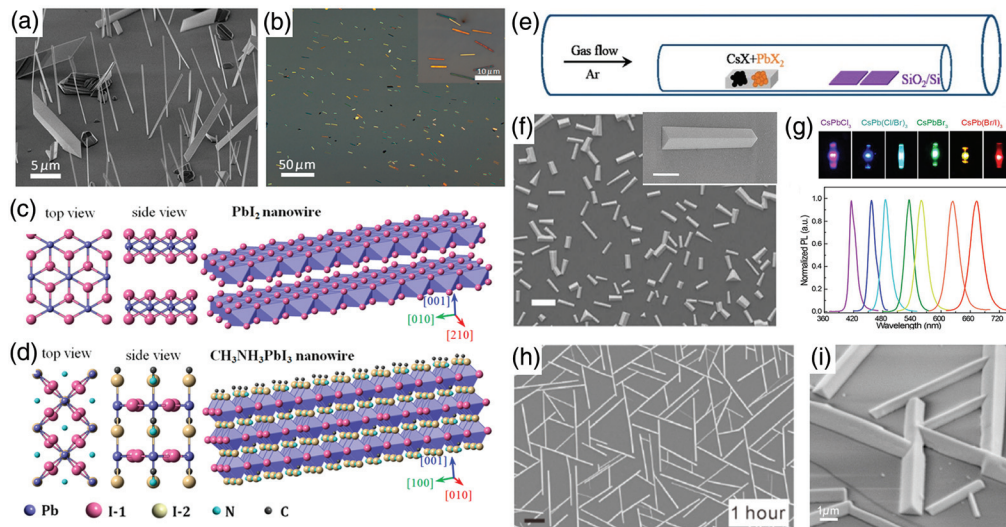


Fig. 4 (a) SEM image of PbI_2 NWs. Figures reproduced from Ref. 49. (b) Optical microscopy image of MAPbI_3 NWs. Figures reproduced from Ref. 49. Structure simulation images of (c) PbI_2 NW and (d) MAPbI_3 NW. Figures reproduced from Ref. 49. (e) Schematic of the CsPbX_3 triangular micro/NRs. Figures reproduced from Ref. 75. (f) SEM image of CsPbBr_3 triangular rods. Figures reproduced from Ref. 75. (g) Real-color image and PL spectra of CsPbX_3 triangular rods. Figures reproduced from Ref. 75. (h), (i) SEM images of the CsPbBr_3 NWs. Figures reproduced from Ref. 62.

demonstrated that the reaction temperature was critical for the control of perovskite NCs during the growth of triangular CsPbBr_3 NRs. Moreover, the emission of these as-grown CsPbX_3 NRs also can be tuned from 415 to 673 nm by halide component regulation [Fig. 4(g)].⁷⁵

In addition, it has been confirmed that the substrate can affect the grain orientation growth of perovskite NWs.⁷⁶ Chen et al.⁶² fabricated CsPbX_3 wires on mica by the CVD method. During the growth of CsPbBr_3 NWs, heteroepitaxial matching occurred in the interface between CsPbBr_3 NCs and mica substrate. Then, the formation of NWs was caused by the asymmetric lattice mismatch with the mica substrate. As shown in Figs. 4(h) and 4(i), the obtained CsPbBr_3 wires were well-aligned, surface-bound, and formed a network with a length about tens of μm and width of $\sim 1 \mu\text{m}$, respectively.⁶² Moreover, various nanostructures could be formed by controlling the deposition time, such as single NWs, Y-shaped branches, and interconnected NW or MW networks.⁶²

2.4 2D Metal Halide Perovskite Nano/Microstructures

2.4.1 Perovskite nano/microplates

The unique and excellent properties of 2D structured perovskite such as NSs, NPs, and microdisks (MDs) make them promising for potential optoelectronic devices.⁷⁷ Sichert et al.⁶¹ synthesized MAPbBr_3 NPs and investigated the quantum size effect of NPs via the solution method. They found that the thickness of MAPbBr_3 NPs was reduced with the increase of the content of OA [Figs. 5(a) and 5(b)]. Correspondingly, the PL emission was tuned from the green to violet region [Fig. 5(c)].⁶¹ Qin et al. prepared MAPbI_3 NPs via a two-step solution method. First, PbI_2/DMF solvent was spin-coated onto a substrate to form PbI_2 thin films. Then, the formed PbI_2 thin film was immersed into MAI solution, in which MAPbI_3 single NCs were formed.⁸¹

CsPbBr_3 NPs were prepared by Bekenstein et al.³⁷ through a hot-injection method. They demonstrated that the reaction temperature is critical for the shape and thickness of CsPbBr_3 NPs. As the temperature decreased from 150°C to 130°C , the shape of CsPbBr_3 NCs evolved from nanocubes to NPs. Correspondingly, the PL emission was shifted from 512 to 405 nm [Figs. 5(d) and 5(e)].³⁷ When the temperature decreased to 90°C and 100°C , the thin CsPbBr_3 NPs were obtained with lengths of about 200 to 300 nm [Fig. 5(d)].³⁷ Except for the reaction temperature, surface ligands also affect the formation of CsPbX_3 NPs, which was demonstrated by Pan et al. in 2016. During the growth of NPs, CsPbX_3 NPs were obtained at a relatively lower reaction temperature (120°C to 140°C). They obtained thinner CsPbX_3 NPs with shorter chain amines.⁸² In addition, the reaction time was also found to be critical for the formation of perovskite NPs.^{78,79} By adding the PbBr_2 concentration and increasing the reaction time above 1 h (135°C), a CsPb_2Br_5 microplate (MP) with a micrometer order size and regular end faces could be obtained [Fig. 5(f)].⁷⁸ In 2018, Li et al. demonstrated that 2D CsPbX_3 NPs and NSs can be obtained by varying the reaction time [Fig. 5(g)]. The thickness can be controlled in the range of 3 to 6 nm and the width in the range of 0.1 to 1 μm .⁷⁹ Huang et al. reported a method for spontaneous crystallization of perovskite NCs in nonpolar organic solvent by mixing precursor ligand complexes without any heat treatment. By varying the ratio of monovalent to Pb^{2+} cation–ligand complexes, the shape of the NCs can be controlled from 3D nanocubes to 2D nanoplatelets.⁸³

Similar to the NWs, perovskite NPs can also be formed by the vapor synthesis method. Xiong and coworkers⁸⁰ reported the CVD growth of MAPbI_3 NPs. These NPs exhibited triangular or hexagonal platelet shapes, with thickness of 10 to 300 nm and lateral dimensions of 5 to 30 μm [Figs. 5(h) and 5(i)]. PbX_2 platelets were first grown on mica via van der Waals epitaxy and then converted to MAPbX_3 NPs with the existence of MAX. In

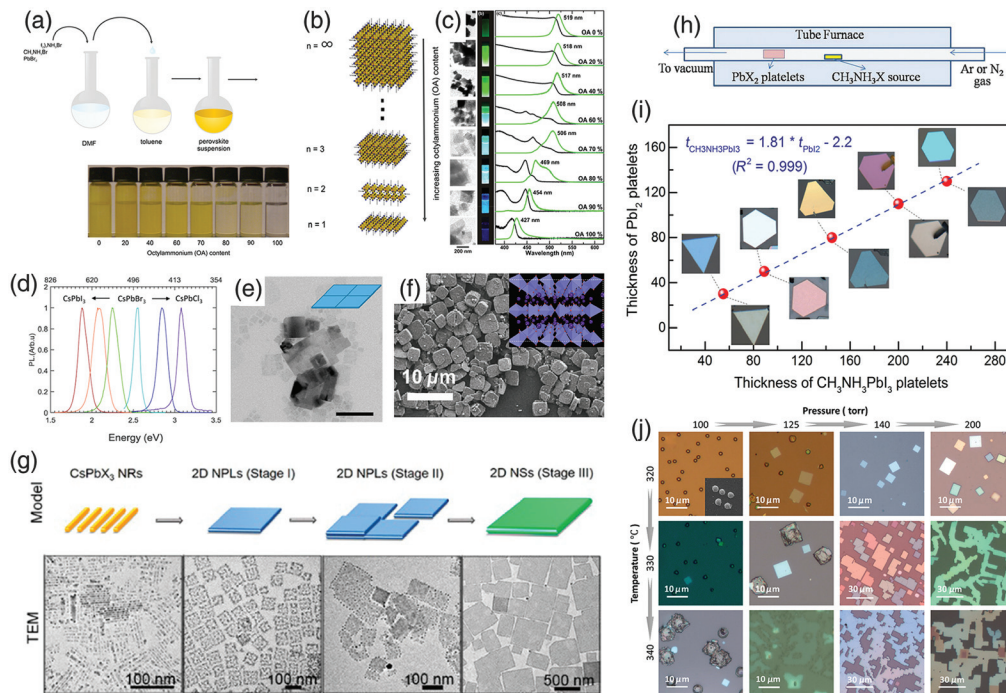


Fig. 5 (a) Schematic of the synthesis of MAPbBr₃ NPs. Figures reproduced from Ref. 61. (b) Quantum size effect in MAPbBr₃ NPs. Figures reproduced from Ref. 61. (c) Bandgap tuning in MAPbBr₃ NPs and micro/NRs via size or compositional control. Figures reproduced from Ref. 61. (d) PL spectra of the halide–anion exchanged CsPbX₃ NPs. Figures reproduced from Ref. 37. (e) 2D CsPbBr₃ NSs. Figures reproduced from Ref. 37. (f) SEM images of CsPb₂Br₅ MP. Figures reproduced from Ref. 78. (g) Top: Schematic of the growth of 2D CsPbX₃ NPs and NSs from CsPbX₃ NRs. Bottom: TEM images of CsPbBr₃ NCs for different times. Figures reproduced from Ref. 79. (h) Schematic of the fabrication of MAPbI₃ NCs using a vapor-transport system. Figures reproduced from Ref. 80. (i) Thickness of PbI₂ platelets before and after being converted to MAPbI₃. Figures reproduced from Ref. 80. (j) Optical images of as-grown MAPbI₃ NCs with different temperature and pressure. Figures reproduced from Ref. 64.

2016, Bao and coworkers developed a combined method containing a solution process and a vapor-phase conversion process to prepare MAPbI₃ NSs. First, PbI₂ flakes were dropped on a silica substrate and then heated. In this process, the temperature plays a crucial role in the nucleation and growth of 2D PbI₂ NSs, since the amount of nucleation sites is controlled by temperature. Subsequently, MAPbI₃ NSs were formed after the conversion reaction with MAI.⁸⁴ During the vapor-phase growth, the growth pressure and temperature both could affect the formation of perovskite NCs. Liu et al.⁶⁴ fabricated 2D MAPbBr₃ platelets (001) via the CVD method. As shown in Fig. 5(j), the square-shaped platelets could not form, as the growth pressure and temperature were low. By increasing the pressure, 2D platelets and 3D spheres could be observed. The average thickness of MAPbBr₃ platelets increased from 29 to 73 nm, and the lateral size increased from 6 to 10 μm with the pressure increasing from 140 to 200 Torr.⁶⁴

As for all-inorganic 2D perovskite NCs, Zeng and coworkers⁸⁵ synthesized ultrathin CsPbBr₃ NPs (thickness ~148.8 nm) on a mica substrate by van der Waals epitaxy through heating the PbBr₂ and CsBr mixture. Zheng et al.⁸⁶ synthesized 2D CsPbI₃ perovskite NSs with high quality, controllable morphology, and ultrathin thickness (~6.0 nm) via a space-confined vapor-phase epitaxial growth. In 2020, Yang and coworkers developed a facile method to pattern CsPbX₃ plate arrays with crystal size (200 nm

to 1 μm) and spacing (2 to 20 μm). These plate arrays were confined by prepatterned hydrophobic/hydrophilic surfaces.⁶⁵ The method can evade the restriction of lattice matching between perovskite and substrates, enabling a large-area growth of 2D perovskite NCs with excellent crystalline quality.⁸⁵

2.4.2 Metasurface

A metasurface is a type of 2D optical element composed of units with subwavelength scale size, producing resonant coupling between electric and magnetic components of the incident electromagnetic fields.^{87–91} Several functionalities were demonstrated on all-dielectric metasurfaces, such as optical encoding, optical wavefront molding, polarization beam splitter, and enhanced PL.^{92–94} Perovskite-based metasurfaces demonstrated potential for nonlinear absorption and optical encoding.⁹⁵ Metasurface structures can be realized by nanopatterning thin film. Many conventional nanofabrication techniques have been used for the fabrication of perovskite metasurfaces, such as nanoimprinting, electron beam lithography (EBL), focused ion beam milling (FIB), and inductively coupled plasma etching (ICP).^{90,94,96–98}

Gholipour et al.⁹⁷ first used the FIB technique to fabricate MAPbI₃ metasurfaces (thickness ~200 nm), which consisted of nanogratings and nanoslit metamolecules. Moreover, they demonstrated that the emission and quality factor of the

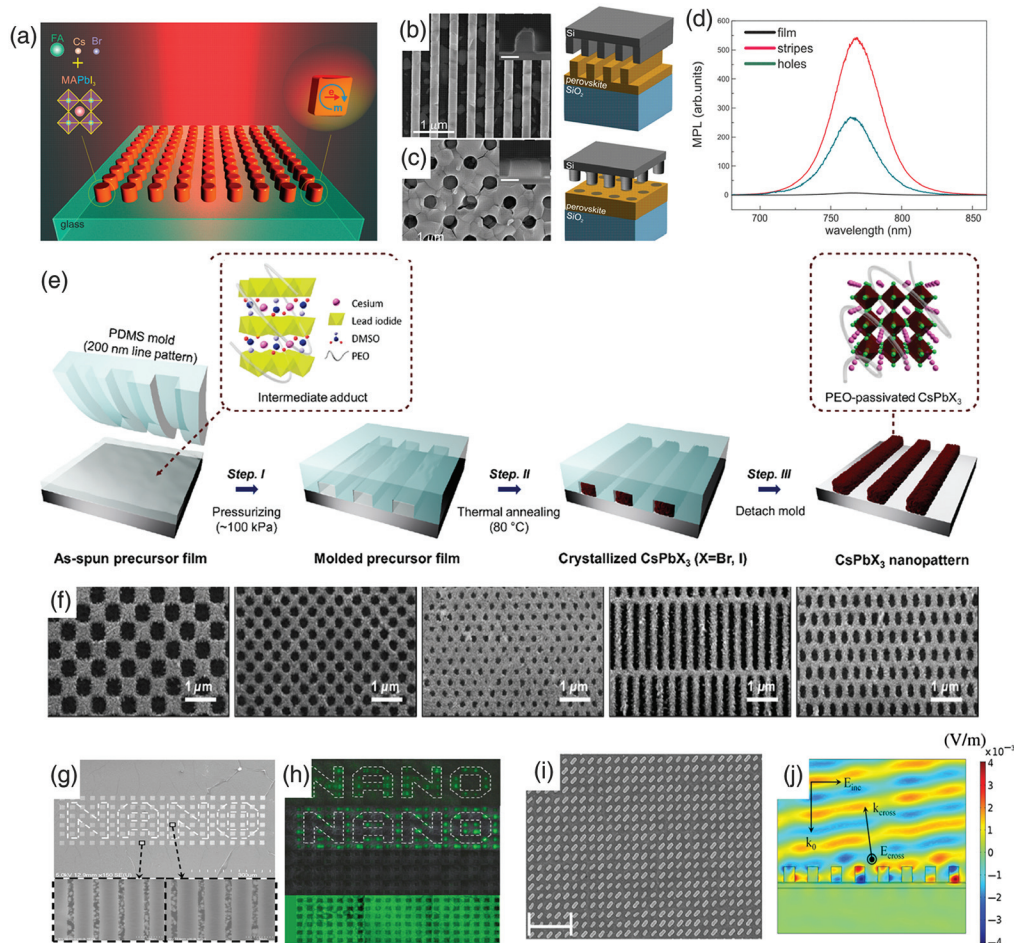


Fig. 6 (a) Perovskite metasurfaces with enhanced emission. Figures reproduced from Ref. 98. SEM images of perovskite with (b) nanostripe and (c) nanohole structures. Figures reproduced from Ref. 98. (d) Enhanced PL spectra from perovskite metasurfaces with different structures. Figures reproduced from Ref. 98. (e) Schematics of the polymer-assisted nanoimprinting process for perovskite nanopatterns. Figures reproduced from Ref. 94. (f) SEM images of various perovskite nanopatterns. Figures reproduced from Ref. 94. (g) SEM images of MAPbBr₃ metasurface for nonlinear imaging. Figures reproduced from Ref. 90. (h) The nonlinear PL and linear PL images of MAPbBr₃ metasurfaces. Figures reproduced from Ref. 90. (i) SEM image of MAPbBr₃ metasurface. Figures reproduced from Ref. 96. (j) The field distributions of MAPbBr₃ perovskite metasurface. Figures reproduced from Ref. 96.

reflection resonances can be tuned by varying the grating period.⁹⁷ Makarov et al.⁹⁸ developed nanoimprinting technology for patterning Cs_αFA_βMA_γPb(I_xBr_y)₃ metasurfaces, enabling them to enhance their linear and nonlinear PL [Figs. 6(a)–6(d)]. After the spin-coating of the perovskite film with thickness of ~200 nm, nanoimprinting with nanopillar and nanostripe molds was performed on perovskite thin film to form metasurfaces. They demonstrated that these metasurfaces can enhance linear PL eight times and nonlinear PL 70 times.⁹⁸ Jeong et al.⁹⁴ presented a polymer-assisted nanoimprinting method for fabricating large-area CsPbX₃ nanopatterns. As shown in Fig. 6(e), during their nanoimprinting process, a precursor solution was spin-coated on a substrate initially, and then the nanoimprinting mold was pressed on the precursor film with thermal treatment subsequently. Thus, CsPbX₃ was crystallized within the confines of molds [Fig. 6(f)]. This method could be

easily extended to large-area perovskite patterns on different substrates.⁹⁴ In addition, Fan et al.⁹⁰ used the EBL flowed ICP technique to prepare near-infrared MAPbBr₃ perovskite metasurfaces [Fig. 6(g)]. Based on these metasurfaces, many types of nonlinear processes and enhanced PL could be observed [Fig. 6(h)].⁹⁰ The authors presented the application of perovskite metasurfaces on optical encryption.⁹⁰ The perovskite metasurface also can be used in optical phase control, which was confirmed by Zhang et al. in 2019. They also used the EBL flowed ICP technique to prepare MAPbX₃ cut-wire metasurfaces on metal substrates [Figs. 6(i) and 6(j)].⁹⁶ They found that these MAPbX₃ metasurfaces can generate a full phase control from 0 to 2π and high-efficiency and broadband polarization. Finally, they proved the potential application in holographic images based on the unique property of perovskite metasurfaces.⁹⁶

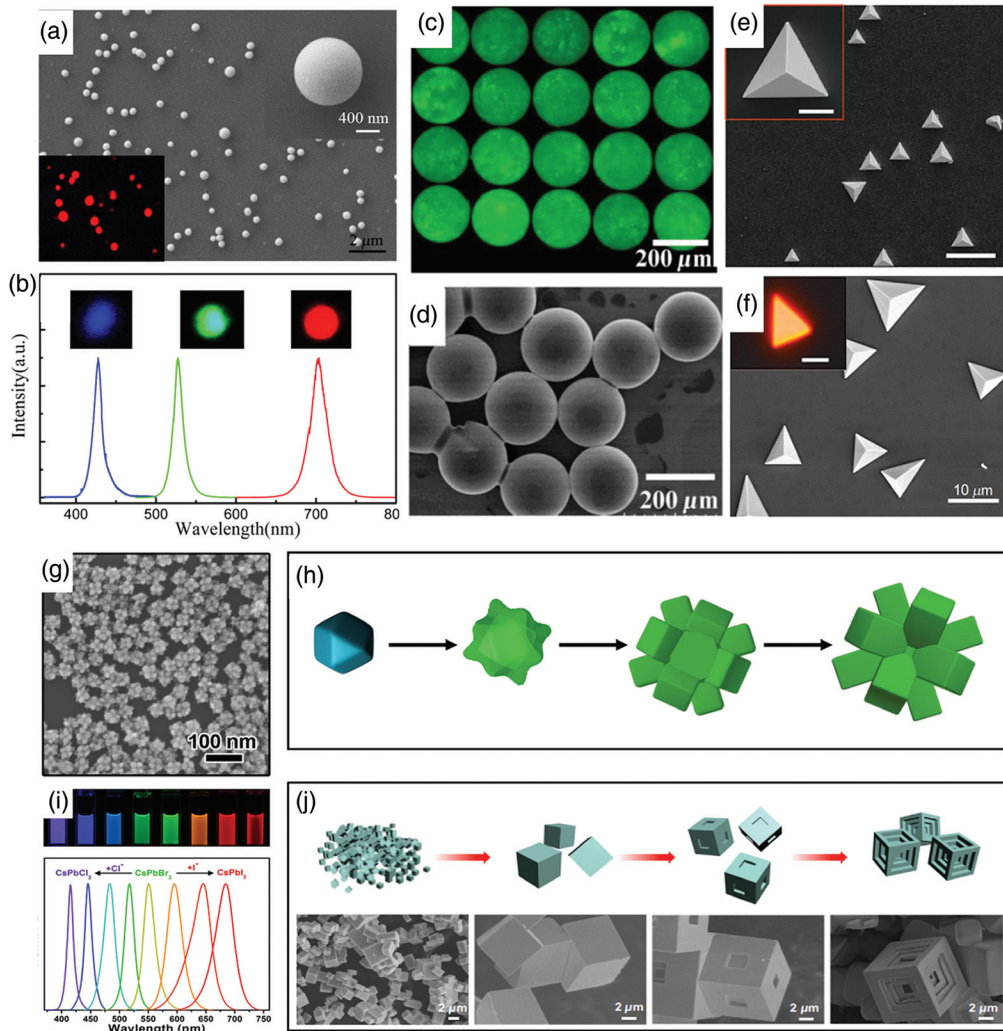


Fig. 7 (a) SEM image of the CsPbI₃ MSs. Figures reproduced from Ref. 100. (b) PL spectra of CsPbCl₃, CsPbBr₃, and CsPbI₃ MSs. Figures reproduced from Ref. 100. (c) Monodispersed CsPbBr₃ spheres under the excitation of UV light. Figures reproduced from Ref. 101. (d) SEM image of the monodispersed CsPbBr₃ spheres. Figures reproduced from Ref. 101. (e) SEM image of the MAPbBr₃ triangular pyramids. Figures reproduced from Ref. 102. (f) SEM image of the CsPbI₃ triangular pyramids on a Si/SiO₂ substrate. Figures reproduced from Ref. 103. (g) SEM image and (h) schematic of the formation of CsPbX₃ nanoflowers. Figures reproduced from Ref. 104. (i) Photograph (upper) and PL emission spectra (bottom) of CsPbX₃ nanoflowers. Figures reproduced from Ref. 104. (j) Crystal growth of MAPbBr₃ cuboids (top) and SEM images of MAPbBr₃ perovskite under different reaction time (bottom). Figures reproduced from Ref. 105.

2.5 3D Metal Halide Perovskite Nano/Microstructures

Besides 1D and 2D structured-perovskite, perovskite-based 3D structures have also been investigated. In 2017, a two-step method for the fabrication of CsPbX₃ microcubes with sub-wavelength size was developed by Hu et al.⁹⁹ These CsPbX₃ microcubes had a regular cube shape and smooth end faces, displaying tunable emission and excellent structure stability for several months under ambient conditions. In the same year, Zhang and coworkers used the CVD method on the prepared CsPbX₃ MSs with controlled diameter of ~1 μm and tunable PL ranging from 425 to 715 nm [Figs. 7(a) and 7(b)].¹⁰⁰ Wei et al.¹⁰¹ developed an automated microreactor system to fabricate an inorganic perovskite NCs sphere by UV photoinitiated

polymerization in flow-focusing microfluidics [Figs. 7(c) and 7(d)]. These obtained CsPbBr₃ spheres had a large diameter around 100 μm, and the diameter could be influenced by flow rates.¹⁰¹ Mi et al.¹⁰² used the CVD method to fabricate high-quality single MAPbBr₃ crystals with a cube-corner pyramids shape and lateral dimension in the range of 2 to 10 μm on mica substrates [Fig. 7(e)]. Then, Yang et al.¹⁰³ also used the CVD method to fabricate CsPbI₃ triangular pyramids with a spontaneous emission of ~719 nm at room temperature on a Si/SiO₂ substrate [Fig. 7(f)].

Except for the regular morphologies, complex perovskite structures have also been investigated. Chen et al.¹⁰⁴ used a seed-mediated solvothermal method to fabricate monodisperse CsPbX₃ NCs with nanoflower morphology [in Figs. 7(g)–7(i)].

Figure 7(h) shows the growth process of CsPbBr₃ nanoflowers, which is formed by the structure transformation from Cs₄PbBr₆ to CsPbBr₃. It is obtained that CsPbX₃ dodecapods contained 12 well-defined branches, with a PLQY of about ~50%. Moreover, the PL emission could be tuned from 415 to 685 nm. They prepared a white LED device based on using CsPbBr₃ nanoflowers, exhibiting the 135% National Television System Committee (NTSC) standard.¹⁰⁴ In 2019, Li et al. fabricated single crystal microcuboid-MAPbBr₃ and multistep-MAPbBr₃ NCs via the solvothermal method at 120°C. In this process, microcuboid-MAPbBr₃ was formed initially, and then the center of the surface was etched after long-time reaction, inducing the formation of multisteps. By adjusting the reaction temperature and time, the morphology and size of microcuboid-MAPbBr₃ [Fig. 7(j)] could be adjustable, with performing potential in perovskite nanolaser and other optoelectronic devices.¹⁰⁵

3 Perovskite-Based Laser

3.1 Nonlinear Optical Properties

Nonlinear optics describes the nonlinear state of the interaction between light and matter.^{106–111} The researches on optical nonlinear materials are fundamental to nonlinear optics devices such as optical storage, optical switches, optical amplifiers, and lasers.^{112–114} Due to the multifariousness of physical and chemical properties, halide perovskites have been demonstrated as promising materials as nonlinear optics materials, which are related to the component and crystal structure of perovskite NCs.¹⁰⁶

In 2015, Sargent and coworkers investigated two-photon absorption in MAPbBr₃ single crystals, under ultrashort pulses 800 nm excitation [Figs. 8(a)–8(d)]. They observed two-photon PL around ~572 nm with an absorption coefficient of $8.6 \pm 0.5 \text{ cm GW}^{-1}$ at 800 nm.¹¹⁷ Later, Heiko et al. performed

temperature-dependent PL measurements on MAPbBr₃ single crystals under 810 nm excitation. They observed obvious wavelength shifts of PLs with variable temperatures, which was attributed to discrete transitions between several stable crystalline phases of MAPbBr₃ single crystals.¹¹⁵ In 2016, Kalanoor et al. studied the nonlinear optical responses of MAPbI₃ films by the Z-scan technique, under nanosecond and femtosecond pulsed lasers. The nonlinear refractive index under femtosecond excitation was $\sim 69 \times 10^{-12}$ and $\sim 34.4 \times 10^{-9} \text{ cm}^2/\text{W}$ for resonant nanosecond excitation, which was equivalent to conventional semiconductors.¹¹⁸ The Z-scan study of MAPbX₃ (X = Cl, Br, I) perovskite film under the 800 nm, 40 fs pulse indicated that MAPbI₃ films have a relatively large nonlinear optical coefficient compared with the MAPbCl₃ and MAPbBr₃ films.¹¹⁹ In the case of inorganic perovskites, Sun and coworkers discovered nonlinear optical properties of CsPbX₃ NCs for the first time [Figs. 8(e)–8(g)]. They observed strong two-photon absorption from 9-nm-sized CsPbBr₃ NCs, with a large absorption cross-section of $\sim 1.2 \times 10^5 \text{ GM}$.¹¹⁶ The nonlinear optical properties of CsPbX₃ perovskite are highly correlated with their morphology. Jiang and coworkers¹²⁰ investigated nonlinear optical properties of CsPbBr₃ NSs with a dependence on their thickness. When the thickness of CsPbBr₃ NS was adjusted from ~104.6 to ~195.4 nm, PL intensity increased nearly three times. They demonstrated that the two-photon absorption coefficient is inversely proportional to the thickness of CsPbBr₃ NSs.¹²⁰ Krishnakanth et al. investigated nonlinear optical properties from nanocubes and NRs by Z-scan technology, under femtosecond 600, 700, and 800 nm lasers. They obtained large two-photon absorption cross sections of $\sim 10^5 \text{ GM}$ and strong nonlinear optical susceptibility of $\sim 10^{-10} \text{ esu}$ in these films.¹²¹

The laser is a process of amplifying optical signals and generating high-intensity coherent light through stimulated radiation and is usually composed of three parts: energy pumping

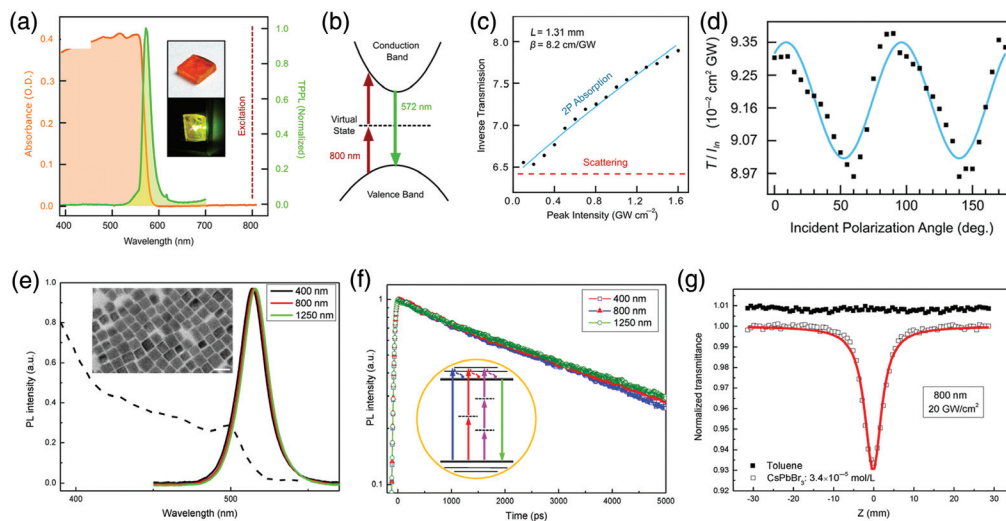


Fig. 8 (a) Absorption spectrum and normalized two-photon PL spectra of single MAPbBr₃ NCs. Figures reproduced from Ref. 115. (b) Schematic of two-photon absorption at 800 nm in perovskite. Figures reproduced from Ref. 115. (c) Two-photon absorption coefficient. (d) Inverse transmission versus peak intensity for typical single MAPbBr₃ NCs. Figures reproduced from Ref. 115. (e)–(g) Nonlinear optics of CsPbX₃ NCs: (e) linear absorption spectrum and normalized PL spectra from CsPbBr₃ NCs, (f) PL decay of CsPbBr₃ NCs, and (g) Z-scan responses of the CsPbBr₃ NC solution and the pure solvent. Figures reproduced from Ref. 116.

source, gain medium, and optical resonator. The amplification of the laser can be quantified as the resonance ability of gain media.^{122,123} For gain media, the optical gain of the semiconductor is similar to the optical absorption, which is suitable for perovskite.^{124,125} At the same time, optical losses are generated in optical cavities, which mainly come from nonradiative recombination, phonon scattering, edge scattering, and field leakage in the interface of cavities.¹²⁶ Perovskite materials have ultralow density, inducing high optical gain and low optical losses for resonance in perovskite, enabling promising potential in perovskite lasers with low threshold. The optical gain of semiconductors can be calculated by the variable-stripe-length measurement, which is related to the dependence of the amplified luminescence on the length of the slit width of the excitation. Xing et al.^{23,124} performed variable stripe length measurements on MAPbI₃ with a gain coefficient of $\sim 250 \text{ cm}^{-1}$, which was close to that of conventional semiconductor materials. The obtained optical gain coefficients of MAPbBr₃ and MAPbCl₃ were ~ 300 and $\sim 110 \text{ cm}^{-1}$, respectively.^{127,128} Liu et al.¹²⁹ demonstrated that the optical gain coefficient of CsPbBr₃ nanocuboids can be calculated to be $\sim 502 \text{ cm}^{-1}$ under the 800 nm laser. Then, Zhao et al. reported efficient two-photon ASE from CsPbBr₃ single crystals with a millimeter size and an optical gain of 38 cm^{-1} .¹³⁰

3.2 Perovskite QDs Laser

In the case of perovskite QDs without an external cavity, the amplification was generated from multiple scattering between QDs, enabling random fluctuations of lasing modes.¹²⁷ In 2015, Kovalenko and coworkers reported low-threshold ASE from colloidal CsPbX₃ NCs with an optical gain coefficient of $\sim 450 \text{ cm}^{-1}$ and threshold of $\sim 5 \mu\text{J}/\text{cm}^2$.¹²⁷ In Figs. 9(a)–9(c), the ASE from CsPbX₃ NCs could be tuned from 440 to 700 nm. Finally, they obtained random lasing from CsPbX₃ films without the resonant cavity and whispering gallery mode (WGM) lasing using a silica sphere as the resonant cavity [Fig. 9(c)].¹²⁷ Besides, coating perovskite QDs onto an external cavity, Zeng and coworkers developed another method to form resonant cavities for perovskite QDs. They obtained enhanced random lasing from strong scattering in the perovskite/SiO₂ composite with low threshold of $\sim 40 \mu\text{J}/\text{cm}^2$ [Figs. 9(d)–9(f)].¹³¹ Similarly, Yang et al.¹³⁶ realized upconversion random lasing from FAPbBr₃/A-SiO₂ composites with a threshold of $\sim 413.7 \mu\text{J}/\text{cm}^2$. Liu et al.¹³⁷ obtained WGM and random lasing with a threshold of $\sim 430 \mu\text{J}/\text{cm}^2$ under 800 nm excitation by embedding CsPbBr₃ QDs into a single silica sphere. In addition, microcapillary tubes can also be used to build WGM cavities for perovskite QDs. In 2015, Zeng and coworkers observed lasing emission from CsPbBr₃ QDs by filling the CsPbBr₃ QDs into a capillary tube, which acted as a WGM cavity for perovskite QDs film around the inner wall.¹³⁸ Later, stable two-photon pumped WGM lasing was realized by coupling CsPbBr₃ and FAPbBr₃ perovskite QDs into microtubules with thresholds of ~ 0.8 and $\sim 0.31 \text{ mJ}/\text{cm}^2$ [Figs. 9(g)–9(j)], respectively.^{132,139}

Besides the realization of perovskite QDs lasing-based silica sphere and microcapillary tube, the well-designed distributed Bragg reflector (DBR) can also be used to achieve a vertical-cavity surface-emitting laser (VCSEL).^{133–135} In 2017, Zeng and coworkers first fabricated VCSELs with a sandwiched structure of DBR/CsPbBr₃ QDs/DBR, which exhibited a low

threshold $\sim 9 \mu\text{J}/\text{cm}^2$ directional output and favorable stability [Fig. 9(k)].¹³³ The lasing emission of CsPbX₃-based VCSELs can be tuned in the visible light range.¹³³ In the same year, Huang et al.¹³⁴ fabricated CsPbBr₃ QDs VCSELs with ultralow threshold of $\sim 0.39 \mu\text{J}/\text{cm}^2$ [Fig. 9(l)]. Organic hybrid perovskites-based VCSELs have also been performed. Chen and Nurmikko¹³⁵ developed FAPbBr₃-based VCSELs by embedding FAPbBr₃ solid thin films in two DBRs [Fig. 9(m)] with a threshold of $\sim 18.3 \mu\text{J}/\text{cm}^2$ under subnanosecond pulse excitations. They also demonstrated that the VCSEL device fabrication process can be applicable to flexible substrates, as shown in Fig. 9(m), which extended further practical applications for perovskite-based laser devices.¹³⁵ Most recently, Li et al.¹⁴⁰ fabricated a two-photon-pumped MAPbBr₃ VCSEL by intergrading MAPbBr₃ with DBR and Ag mirrors with a threshold of $\sim 421 \mu\text{J}/\text{cm}^2$, a Q factor of ~ 1286 , and a small divergence of $\sim 0.5 \text{ deg}$.

3.3 Perovskite Nanowire/Nanorod Laser

Owing to the difference between the refractive index of perovskite material and air, the reflection can occur at the output interface easily, acting as optical reflector.^{141,142} Hence, different from QDs, single perovskite crystals structures such as rods, wires, plates, cubes, and spheres can act as Fabry–Pérot (F-P) or WGM cavities by themselves, since the light can be confined in the resonant cavity with regular morphology and smooth end faces.¹²⁵ For the 1D NWs structure, light will propagate along 1D and form resonance between two end-facets.¹⁴³ Hence, perovskite NWs and NRs have been confirmed as potential structures in optoelectronic devices and nanoscale-integrated photonics due to their unique optical properties, such as highly coherent output and efficient waveguide effect.¹⁴³

Zhu et al.¹⁴⁴ demonstrated perovskite NW lasers using high-quality MAPbX₃ NWs, which had a regular shape with rectangular cross section [Fig. 10(a)]. Tunable F-P lasing could be observed from single MAPbX₃ NWs with low threshold of $\sim 0.22 \mu\text{J}/\text{cm}^2$ and Q factor of ~ 3600 at room temperature [Figs. 10(b) and 10(c)]. In the same year, Xing et al.⁴⁹ realized F-P lasing from MAPbI₃ NWs with rectangular morphology and length of $\sim 20 \mu\text{m}$. The obtained NW laser exhibited low threshold of $\sim 11 \mu\text{J}/\text{cm}^2$ and Q factor of ~ 405 , and the lasing wavelength could be tuned in the range of 551 to 777 nm.⁴⁹ In case of lasing from all-inorganic perovskite NWs, Yang and coworkers realized F-P lasing from CsPbBr₃ NWs with a threshold of $\sim 5 \mu\text{J}/\text{cm}^2$ and a Q factor of ~ 1009 .⁵⁵ Fu et al. realized wavelength widely tunable F-P lasing from CsPbX₃ NWs. The lasing wavelength could be tuned in the visible spectral region from 420 to 710 nm [Figs. 10(e)–10(g)].¹⁴⁵ In 2017, lasing emission from triangular CsPbX₃ micro/NRs with an ultrasmooth surface by the vapor-phase approach was reported. The obtained lasing could be tuned in the range from 428 to 628 nm, with low threshold of $\sim 14.1 \mu\text{J}/\text{cm}^2$ and high Q factor of ~ 3500 .⁷⁵ Efficient multiphoton pumped lasing in a wide excitation wavelength range (700 to 1400 nm) was realized.¹⁴⁷ Most of the single perovskite NWs mainly exhibited single-band lasing emission. In 2020, Tang et al. fabricated a single CsPbCl_{3–3x}Br_{3x} alloy NW via a solid–solid anion–diffusion process. They realized continuous F-P lasing in single as-prepared NWs, which could be tuned from 480 to 525 nm [Figs. 10(i) and 10(j)].¹⁴⁶

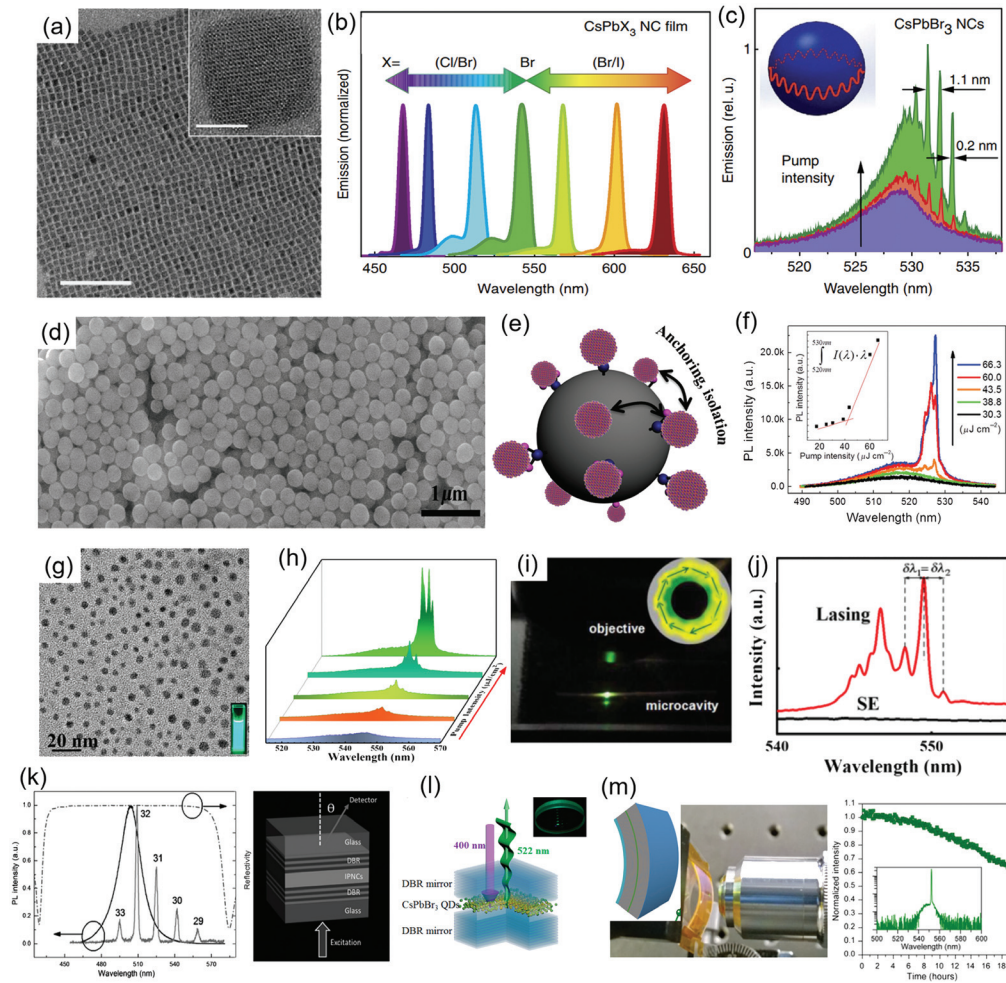


Fig. 9 (a) TEM images of CsPbBr₃ QDs. Figures reproduced from Ref. 127. (b) Spectral tunability of ASE of CsPbX₃ via compositional modulation. Figures reproduced from Ref. 127. (c) Evolution from PL to lasing in an MS resonator with increasing pump intensity. Figures reproduced from Ref. 127. (d) SEM image and (e) isolation effect of CsPbBr₃ QDs/A-SiO₂ composites. Figures reproduced from Ref. 131. (f) PL spectra from CsPbBr₃ QDs/A-SiO₂ composite with increasing pump intensity. Figures reproduced from Ref. 131. (g) TEM image of FAPbBr₃ QDs. (h) Two-photon PL spectra from FAPbBr₃ NCs in a microcapillary tube. (i) Optical image and (j) lasing emission spectra from FAPbBr₃ NCs in a microcapillary tube. Figures reproduced from Ref. 132. (k) Left: PL spectra from CsPbBr₃ film within/without microcavity. Right: Schematic of the CsPbBr₃ VCSEL. Figures reproduced from Ref. 133. (l) Schematic of the CsPbBr₃ VCSEL. Figures reproduced from Ref. 134. (m) Photograph and PL stability of flexible FAPbBr₃ VCSEL. Figures reproduced from Ref. 135.

3.4 Perovskite Nano/Microplate Laser

Different from the F-P cavity formed by NWs/NRs, the perovskite 2D structure such as NPs will result in the WGM optical resonant cavity, which has a higher Q factor than the F-P cavity. In 2014, Zhang et al. first realized WGM lasing from MAPbI₃ NPs with well-defined hexagonal and triangular shapes under femtosecond-pulsed laser excitation. The lasing wavelength was located at ~ 780 nm with a threshold of ~ 37 $\mu\text{J}/\text{cm}^2$ [Figs. 11(a)–11(d)].⁸⁰ Liao et al.¹⁵⁰ obtained single-mode WGM lasing from single MAPbBr₃ MDs peaked at ~ 557.5 nm with a threshold of ~ 3.6 $\mu\text{J}/\text{cm}^2$ and Q factor of ~ 430 . Liu et al. realized WGM lasing from MAPbI₃ MP arrays with low threshold of

~ 11 $\mu\text{J}/\text{cm}^2$ and Q factor of ~ 1210 .¹⁵¹ Moreover, they observed single mode lasing by shortening the size of MPs.¹⁵¹ Qi et al.¹⁵² demonstrated that the threshold of the MPs laser decreases linearly depending on the later size, and the cavity mode density increases with the size. In 2019, WGM lasing from a triangular MAPbI₃ perovskite NP with a lateral length of 27 μm and thickness of 80 nm was realized at room temperature. The threshold of the WGM laser was ~ 18.7 $\mu\text{J}/\text{cm}^2$ and Q factor was ~ 2600 [Figs. 11(e)–11(i)].¹⁴⁸

As for the 2D all-inorganic perovskite-based laser, Zhang et al.¹⁴⁹ obtained WGM excitonic lasing from single-crystalline CsPbX₃ NPs with micron-size length and subwavelength thickness [Figs. 11(j)–11(m)]. Multicolor lasing from 410 to

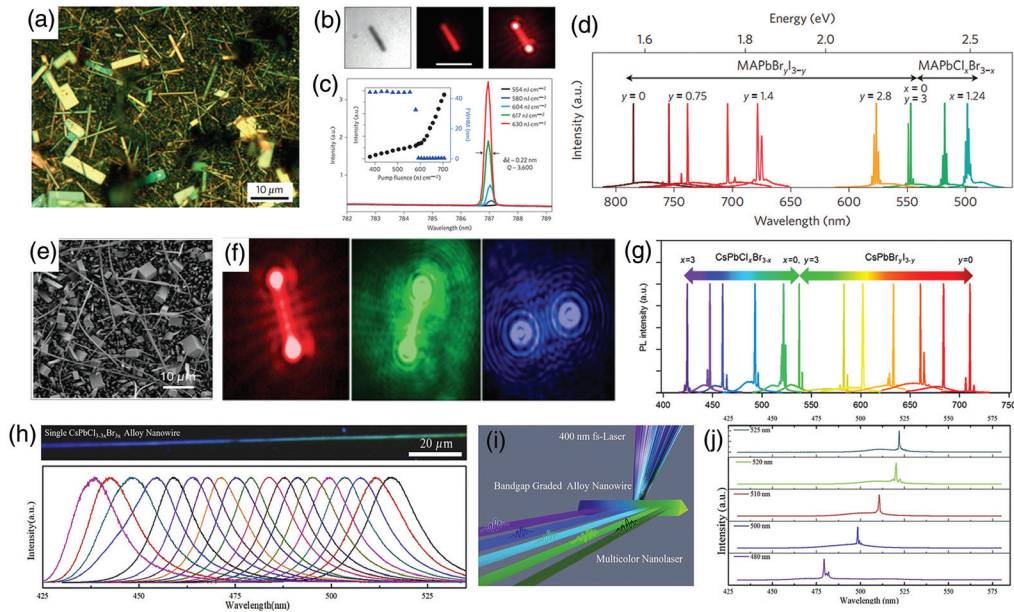


Fig. 10 (a) SEM of MAPbI₃ nanostructures. Figures reproduced from Ref. 144. (b) Optical image of single MAPbI₃ NW. Figures reproduced from Ref. 144. (c) PL spectra of MAPbI₃ NW around the lasing threshold. Figures reproduced from Ref. 144. (d) Broad tunable lasing from single-crystal MAPbX₃ NW. Figures reproduced from Ref. 144. (e) SEM image of CsPbBr₃ nanostructures. Figures reproduced from Ref. 145. (f) Fluorescence images of red/green/blue CsPbX₃ NWs above lasing threshold. Figures reproduced from Ref. 145. (g) Broad tunable lasing from single-crystal CsPbX₃ NWs. Figures reproduced from Ref. 145. (h) The photograph and PL spectra of a single CsPbCl_{3-3x}Br_{3x} NW. Figures reproduced from Ref. 146. (i) The schematic of optically pumping lasing from a single CsPbCl_{3-3x}Br_{3x} NW. Figures reproduced from Ref. 146. (j) Typical lasing spectra from a single CsPbCl_{3-3x}Br_{3x} NW. Figures reproduced from Ref. 146.

700 nm was realized in these NPs at room temperature [Fig. 11(l)]. The lasing threshold of the CsPbX₃ NP was as low as $\sim 2.0 \mu\text{J}/\text{cm}^2$, and the linewidth of the WGM modes was ~ 0.14 to 0.15 nm [Fig. 11(m)].¹⁴⁹ Zheng et al.⁸⁶ demonstrated that CsPbI₃ perovskite NSs possess WGM lasing under both one- and two-photon pumps with low-threshold-pumped excitation [Figs. 11(n)–11(q)]. The thresholds of lasing were ~ 0.30 and $\sim 2.6 \text{ mJ}/\text{cm}^2$ under one- (470 nm) and two-photon (1200 nm) excitation, and the Q factors were ~ 1489 and ~ 1179 , respectively, which is three times higher than the reported values of organic–inorganic lead halide perovskite NS. Most recently, Liu et al.¹⁵³ realized two lasing modes (F-P and WGM) in the all-inorganic perovskite CsPb₂Br₅ MPs with subwavelength thickness and uniform square shape under two-photon pump. Remarkably, low-threshold F-P multimode lasing with Q factor of ~ 3551 and single-mode WGM lasing with Q factor of ~ 3374 from the same MP at room temperature have been achieved successfully.

3.5 Perovskite Laser with 3D Structure

A single perovskite spherical 3D structure has also usually been demonstrated as a WGM cavity. In comparison with other nano/microstructure resonant cavities, the coupling between the sphere cavity and substrate was relatively weak, which resulted in less optical losses. Zhang and coworkers realized single-mode lasing in CsPbX₃ MSs with regular sphere shape and submicron size at room temperature [Figs. 12(a)–12(d)].¹⁰⁰

The line width of WGM lasing was $\sim 0.09 \text{ nm}$, the threshold was $\sim 0.42 \mu\text{J}/\text{cm}^2$, and Q factor was ~ 6100 [Fig. 12(c)]. In addition, the single-mode lasing can be tuned in the whole visible region through element modulation and size control of perovskite MSs [Fig. 12(d)].¹⁰⁰ Furthermore, they achieved two-photon single-mode lasing with linewidth of $\sim 0.037 \text{ nm}$ and Q factor of $\sim 1.5 \times 10^4$ from a single CsPbBr₃ MS at room temperature, which are the best values obtained in perovskite-based micro/nanocavities until now.¹⁵⁴ Moreover, these perovskite MS lasers showed uniform lasing emission, which could be observed in the range from -30 deg to 30 deg.^{154,155}

Another 3D structure generally used for perovskite lasing is the nano/microcube. Liu et al.¹²⁹ obtained F-P lasers from an individual CsPbBr₃ nanocuboid with subwavelength scale for the first time [Figs. 12(e)–12(h)]. They realized single-mode F-P lasing from a CsPbBr₃ nanocuboid with low thresholds of ~ 40.2 and $\sim 374 \mu\text{J}/\text{cm}^2$ and Q factors of ~ 2075 and ~ 1859 under one- and two-photon pumps, respectively.¹²⁹ The physical volume of the obtained laser is $\sim 0.49 \lambda^3$. Moreover, the pulse duration is only $\sim 22 \text{ ps}$, which is consistent with the resulting fast decay of SE observed by fs transient absorption spectroscopy [Fig. 12(h)].¹²⁹ Cube-corner pyramid cavities could also act as microretroreflectors. In 2018, Mi et al. realized F-P lasing in cube-corner MAPbBr₃ pyramids at room temperature [Figs. 12(i)–12(m)].¹⁰² Furthermore, the threshold of lasing could be reduced from ~ 92 to $26 \mu\text{J}/\text{cm}^2$ by coating a thin layer of Ag film on a mica substrate [Figs. 12(l) and 12(m)].¹⁰² Most recently, Yang et al.¹⁰³ also realized F-P lasing from a single

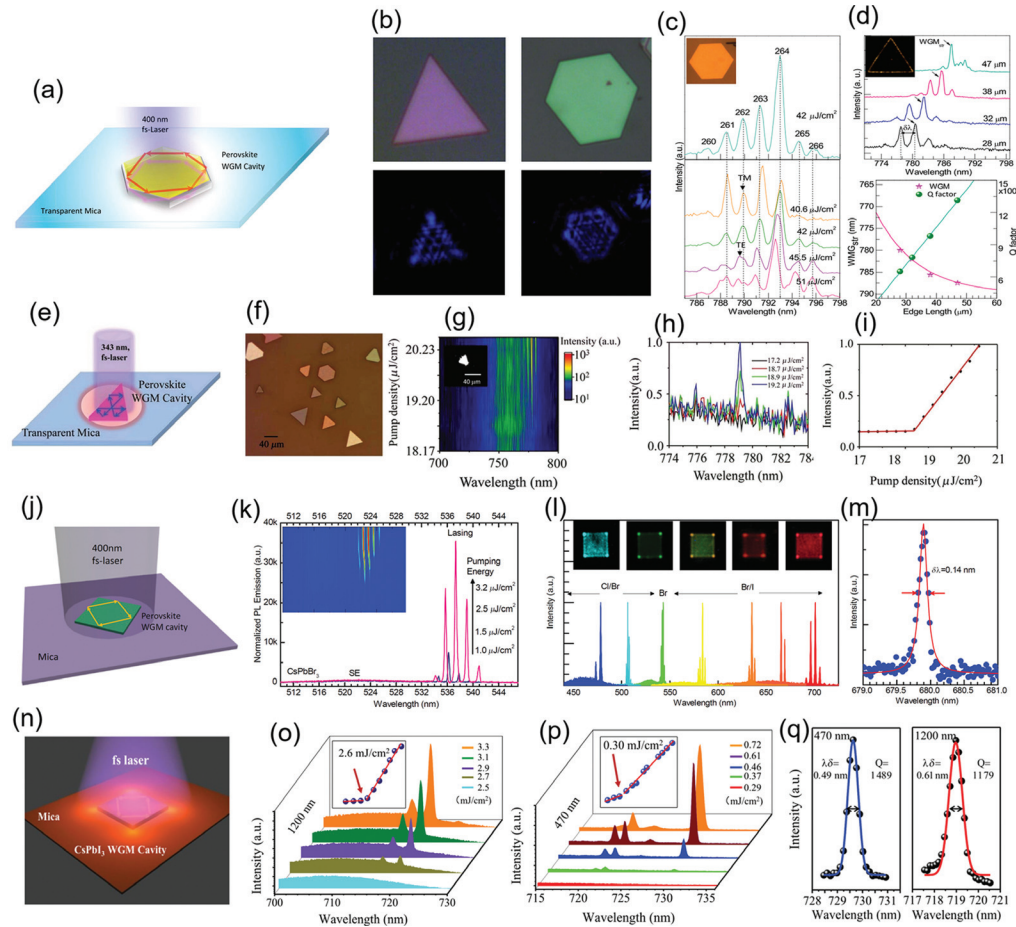


Fig. 11 (a) Schematic of an MAPbX_3 NP pumped by a pulsed laser. Figures reproduced from Ref. 80. (b) Optical image of MAPbI_3 NPs under white light and laser excitation. Figures reproduced from Ref. 80. (c) Lasing spectra of hexagonal MAPbI_3 NPs (upper) and the lasing mode evaluation with pumping fluence (bottom). Figures reproduced from Ref. 80. (d) Upper: Lasing spectra of triangular MAPbI_3 NPs with different edge length. Bottom: The wavelength of lasing modes and Q -factor as a function of the triangular cavity edge length. Figures reproduced from Ref. 80. (e) Schematic of triangular MAPbI_3 NPs pumped by a 343 nm laser. Figures reproduced from Ref. 148. (f) Optical image of triangular MAPbI_3 NPs. Figures reproduced from Ref. 148. (g) 2D plot of a triangular MAPbI_3 NP emission under different pump densities. Figures reproduced from Ref. 148. (h) The emission spectra from MAPbI_3 NPs around the lasing threshold. Figures reproduced from Ref. 148. (i) Output emission intensity as a function of pump densities. Figures reproduced from Ref. 148. (j) Schematic of a CsPbX_3 plate under a 400 nm laser. Figures reproduced from Ref. 149. (k) Emission spectra at different pump intensities. Figures reproduced from Ref. 149. (l) Tunable lasing spectra and images of individual CsPbX_3 perovskite NPs. Figures reproduced from Ref. 149. (m) Single-mode lasing of $\text{CsPbBr}_x\text{I}_{3-x}$. Figures reproduced from Ref. 149. (n) Schematic of a CsPbI_3 NS on mica substrate. Figures reproduced from Ref. 86. Excitation intensity-dependent emission spectra under (o) 470 nm and (p) 1200 nm excitation. Figures reproduced from Ref. 86. (q) Gaussian fitting of a lasing mode under 470 and 1200 nm laser. Figures reproduced from Ref. 86.

CsPbI_3 triangular pyramid with a microsize at low temperature. They demonstrated that the temperature-dependent lasing threshold can be reduced from ~ 53.15 to $21.56 \mu\text{J}/\text{cm}^2$ with corresponding temperature from 223 to 148 K.¹⁰³

3.6 Perovskite Nanolaser Array

In comparison with single perovskite lasers, laser arrays with high-density patterns and high-precision arrangements are more

necessary for mass-produced, compact on-chip optoelectronic circuit integration. In 2016, Wang et al. fabricated MAPbBr_3 microwire arrays and realized high density perovskite lasers from these microwire arrays [Figs. 13(a) and 13(b)], in which all of the subunits generated the same lasing emission.¹⁵⁶ The minimum unit period was 800 nm, presenting an integration density of nanolasers as high as 1250 mm^{-1} . In 2017, Fu and coworkers prepared MAPbBr_3 NW arrays with the width from 460 to 2500 nm, height from 80 to 1000 nm, and length from

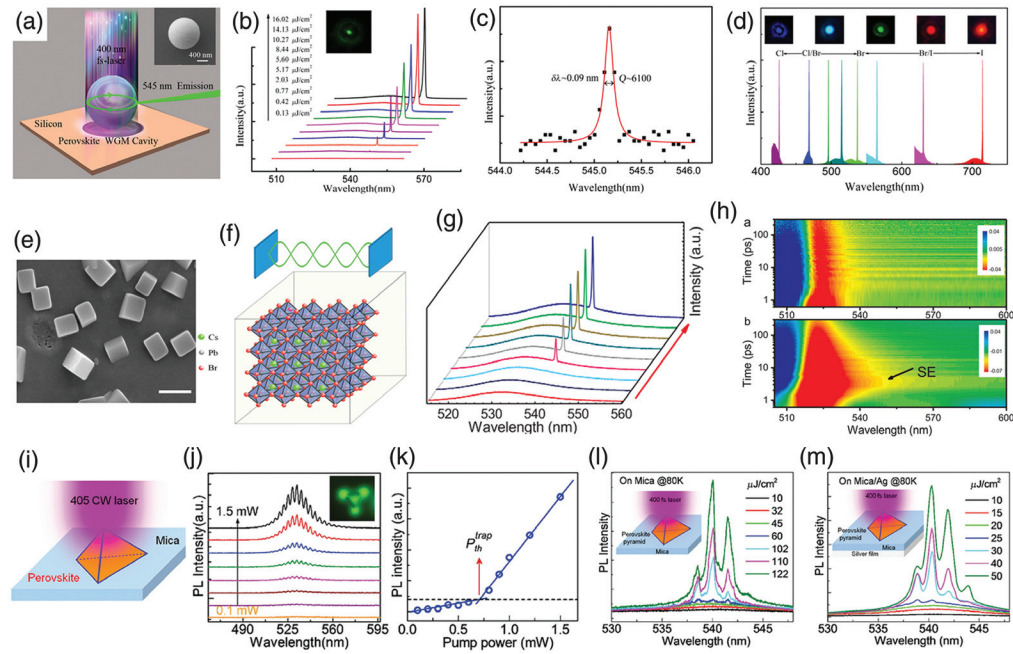


Fig. 12 (a) Schematic of a single CsPbBr₃ MS under 400 nm laser. Figures reproduced from Ref. 100. (b) Lasing PL spectra from a single CsPbBr₃ MS under different pump intensities. Figures reproduced from Ref. 100. (c) Lorentzian fitting of a lasing mode. Figures reproduced from Ref. 100. (d) Photograph and lasing emission of multicolor CsPbX₃ MS lasers. Figures reproduced from Ref. 100. (e) SEM image and (f) schematics of F-P cavity of CsPbBr₃ nanocuboids. Figures reproduced from Ref. 129. (g) Single-mode lasing spectra and (h) TA spectroscopic data of CsPbBr₃ nanocuboids under two-photon excitation. Figures reproduced from Ref. 129. (i) Schematic of a cube-corner MAPbBr₃ pyramid under 405 nm laser. Figures reproduced from Ref. 102. (j) PL spectra of a cube-corner MAPbBr₃ and (k) output intensity as a function of excitation power. Figures reproduced from Ref. 102. (l), (m) Multimode lasing spectra of a cube-corner pyramid of MAPbBr₃ on (l) mica and (m) mica/Ag. Figures reproduced from Ref. 102.

10 to 50 μm . These perovskite NW arrays were demonstrated as almost identical optical resonance cavities with a low threshold of $\sim 10.2 \mu\text{J}/\text{cm}^2$ [Figs. 13(c) and 13(d)].¹⁵⁷ In 2016, Liu et al. realized WGM lasing from patterned MAPbI₃ microplatelets arrays with a threshold of $\sim 11 \mu\text{J}/\text{cm}^2$ and Q factor up to ~ 1210 . The wavelength tunability and single mode lasing could be selected by changing platelet sizes or breaking the symmetry of the designed laser pattern.¹⁵¹ In the same year, Feng et al.¹⁵⁸ demonstrated that the MAPbBr₃ square MP array shows high-performance WGM lasing with tunable mode and low lasing threshold [Figs. 13(e) and 13(f)]. Single mode lasing was obtained from a 2.1- μm MAPbBr₃ square. Lin et al.¹⁵⁹ fabricated a large-area CsPbX₃ QDs array by a photolithographical approach, which could be used as efficient lasing structures and emitting pixel arrays [Figs. 13(g)–13(i)]. They realized WGM lasing from the QD arrays with high Q factor and demonstrated that this patterning technique can be used in large-area perovskite laser arrays with multicolor pixels [Fig. 13(g)].¹⁵⁹ Most recently, Wang et al.¹⁶⁰ fabricated large-area MAPbX₃ MD arrays via a screen-printing technique [Figs. 13(j)–13(m)]. They obtained tunable WGM lasing from these MAPbX₃ MD arrays with a threshold of $\sim 21.3 \mu\text{J}/\text{cm}^2$ and a Q factor of ~ 1570 successfully. Multicolor WGM lasing emission could be tuned from ~ 510 to 650 nm [Fig. 13(l)].¹⁶⁰ In 2020, Song and coworkers employed the topologically protected optical bounded states in the continuum (BICs) and demonstrated the ultrafast control of perovskite-based vortex microlasers at room temperature.

They proved that vortex beam lasing based on perovskite metasurfaces could be switched to linearly polarized beam lasing with switching time of 1 to 1.5 ps. The energy consumption was several orders of magnitude lower than that of previously reported all-optical switching.¹⁶¹

3.7 Others

Surface-plasmon (SP) is an excited state with large enhancement of the electromagnetic field localized at the metal–dielectric interface, which provides confinement on the subwavelength scale, overcoming the diffraction limit of light.¹⁶² In perovskite micro/nanolasers, SPs have been demonstrated as an effective method to tailor the properties of lasers. In general, SPs could be generated by the metal layer, such as Au or Ag, and transfer along the semiconductor–metal interface. Kao et al.¹⁶³ reduced the lasing threshold of perovskite by strong exciton–plasmon coupling between the Ag and perovskite films [Fig. 14(a)],¹⁶³ in which the confined optical fields between Ag and perovskite films could be enhanced about 19.3 and 7.7 times in comparison with bare perovskites and perovskites coated by Ag thin film, respectively. In 2017, Wang et al. deposited Al nanoparticles onto the surface of CsPbBr₃ perovskites. The lasing thresholds of CsPbBr₃ perovskite microrods were significantly reduced by $>20\%$, and the output intensities were significantly enhanced via the plasmonic resonances.¹⁷² In 2019, Wu et al. reported a method to enhance ASE performance of MAPbI₃ films by

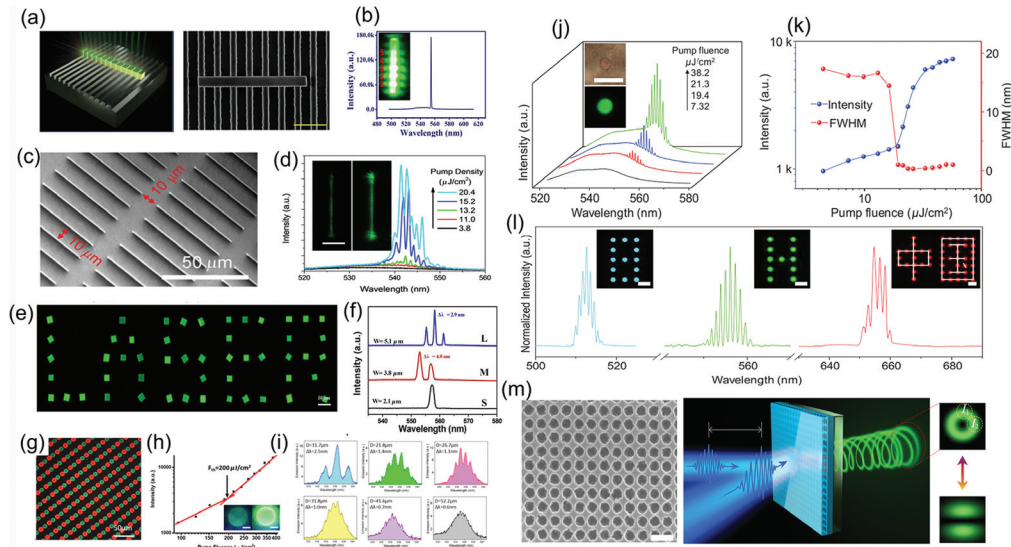


Fig. 13 (a) SEM image of the MAPbBr₃ microwire on silicon grating. Figures reproduced from Ref. 156. (b) Laser spectrum of MAPbBr₃ microwire. Figures reproduced from Ref. 156. (c) Optical image of MAPbBr₃ NW arrays. Figures reproduced from Ref. 157. (d) PL spectra of a single MAPbBr₃ NW under 400 nm laser. Figures reproduced from Ref. 157. (e) “LASER” patterned perovskite square MPs. Figures reproduced from Ref. 158. (f) Lasing spectra from perovskite MPs with different sizes. Figures reproduced from Ref. 158. (g) PL image of green and red QD arrays. Figures reproduced from Ref. 159. (h) Emission intensity versus excitation fluence measured from a CsPbBr₃ MD. Figures reproduced from Ref. 159. (i) Lasing spectra from CsPbBr₃ MDs with different diameters. Figures reproduced from Ref. 159. (j) PL spectra from a typical MAPbBr₃ MD with different power energies and (k) output intensity and FWHM as a function of pump intensity. Figures reproduced from Ref. 160. (l) Widely tunable lasing from MAPbX₃ arrays. Figures reproduced from Ref. 160. (m) Left: SEM image of the fabricated perovskite meta-surface; right: ultrafast control of the quasi-BIC microlasers. Figures reproduced from Ref. 161.

adding Au NRs-doped PMMA on MA₃PbI₃ perovskite films. The ASE threshold was significantly reduced by ~36%, and the output intensity increased by 13.9-fold with the plasmon resonance enhancement of Au NRs.¹⁷³ Yang et al.¹⁷⁴ also reduced the lasing threshold of CsPbBr₃ perovskite nanocubes significantly by ~33% via the surface plasmonic effect of Au nanoparticles. In 2021, single-mode upconversion plasmonic lasing from MAPbBr₃ perovskite NCs was realized by Lu et al.,¹⁶⁴ exhibiting low threshold ~10 μJ/cm² and small mode volume ~0.06 λ³ at 6 K, where TiN was used as a promising resonance adjustable plasmonic platform [Fig. 14(b)]. Hsieh et al.¹⁶⁵ realized continuous-wave (CW) lasing from a single CsPbBr₃ QD in a plasmonic gap-mode nanocavity with low threshold of ~1.9 W/cm² and small mode volume of ~0.002λ³ [Fig. 14(c)]. Most recently, Li et al.¹⁶⁶ proposed a hybrid nanocavity composed of CsPbBr₃ nanoparticles and a thin Au film, which could realize optically controlled quantum size effect by the reversible phase transition from polycrystalline to monocrystalline [Fig. 14(d)]. These results demonstrated that SPs could not only modulate the performance of perovskite lasers but also can realize deep subdiffraction plasmonic lasers.

Perovskite is also an ideal candidate to realize a room temperature exciton polariton laser, which mainly results from the strong exciton–photon coupling between the gain media and nanocavity. In perovskite laser researches, room temperature exciton polaritons have been realized with various

nanostructures.^{167,168,175,176} Perovskite NCs with self-assembled morphology can provide optical resonators due to the confinement of exciton–photon coupling. On the other hand, a planar optical cavity composed of two mirrors can be used as an F-P cavity conventionally. In 2018, Liu et al.¹⁶⁷ observed strong exciton–photon coupling in single CsPbBr₃ micro/NWs and MAPbBr₃ micro/NWs, respectively [Fig. 14(e)]. Moreover, polariton lasing was realized at room temperature with exceptionally large vacuum Rabi splitting of ~656 and 390 meV.^{167,175} Shang et al.¹⁷⁶ proved light could propagate as an exciton–photon in CsPbBr₃ NWs at room temperature, increasing optical absorption and emission in comparison with bulk crystals. They demonstrated that the decrease of CsPbBr₃ dimensions could enhance the exciton–photon coupling strength, which increased the exciton fraction. Furthermore, they found that the increase of temperature could significantly decrease the exciton fraction of exciton–photons, causing high thresholds and restraining CW lasing above 100 K. They successfully realized CW-pumped lasing from CsPbBr₃ nanoribbons by reducing the height to ~120 nm on sapphires with low threshold of ~0.13 kW/cm² [Fig. 14(f)].¹⁶⁸ Then, they coupled MAPbBr₃ NWs with a hybrid plasmonic microcavity to enhance exciton–photon interaction.¹⁷⁷ They observed a Rabi-splitting up to ~564 meV in a hybrid perovskite/SiO₂/Ag waveguide microcavity at room temperature. In 2017, Su et al.¹⁷⁸ reported room-temperature polariton lasing based on an epitaxy-free all-inorganic CsPbCl₃ nanoplatelet embedded in DBRs,

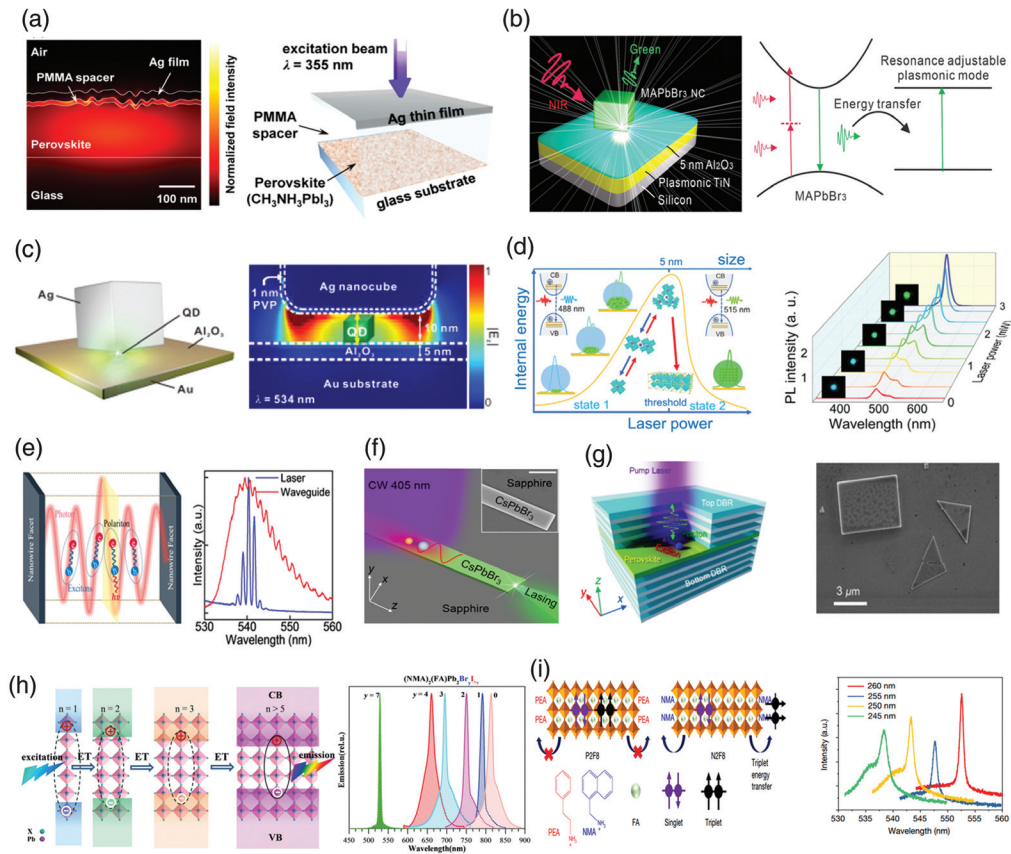


Fig. 14 (a) Field intensity distributions and schematic structure of Ag/PMMA/perovskite. Figures reproduced from Ref. 163. (b) Schematic and working process of plasmonic nanolaser of MAPbBr₃/Al₂O₃/TiN. Figures reproduced from Ref. 164. (c) Schematic and calculated electric field distribution of plasmonic nanolaser based on CsPbBr₃ QDs. Figures reproduced from Ref. 165. (d) Schematic of phase transition from polycrystalline to monocrystalline CsPbBr₃ nanoparticles by adjusting the laser power and the PL spectrum under different laser power. Figures reproduced from Ref. 166. (e) Schematic polaritons in a micro/NW cavity and lasing spectrum of CsPbBr₃ NWs. Figures reproduced from Ref. 167. (f) Schematic of CW lasing of CsPbBr₃ nanoribbons. Figures reproduced from Ref. 168. (g) Schematic structure of CsPbBr₃ flakes/DBR microcavity and SEM image of CsPbBr₃ flakes. Figures reproduced from Ref. 169. (h) Cascade energy transfer in quasi-2D perovskite and tunable ASE from solution-processed (NMA)₂(FA)Pb₂Br_yI_{7-y} films. Figures reproduced from Ref. 170. (i) Chemical structures of quasi-2D perovskite with different organic cations and CW lasing characteristics of quasi-2D perovskite films. Figures reproduced from Ref. 171.

supporting F-P oscillations. The polariton lasing exhibited a threshold of $\sim 12 \mu\text{J}/\text{cm}^2$. Zhang et al.¹⁶⁹ investigated the trapping of polaritons in micron-sized CsPbBr₃ flakes embedded in DBRs as a microcavity [Fig. 14(g)]. They demonstrated quantized polariton states arising from the optical confinement of flakes.

In comparison with perovskite NCs with 3D structure, quasi-2D perovskites have a quantum well (QW) structure with the advantages of large exciton binding energy and low nonradiation loss and are more easily coupled with a resonant cavity. In 2018, Fieramosca et al. observed strong exciton–photon coupling from hybrid 2D perovskite flakes. The organic ligands efficiently affected the out-of-plane exciton–photon coupling, suggesting that the organic interlayer plays a significant role in the anisotropy of the exciton and exciton polariton.¹⁷⁹ Then, they observed highly interacting polaritons in (PEA)₂PbI₄

with an excitonic interaction constant as $\sim 3 \mu\text{eV} \mu\text{m}^2$, which was two orders higher than that of organic excitons.¹⁸⁰ Zhang et al.¹⁸¹ investigated cavity polariton modes in 2D perovskite (PEA)₂PbBr₄ sheets. The perovskite layer naturally could act as an F-P cavity and exhibited evident cavity polariton modes with Rabi splitting energy of $\sim 259 \text{ meV}$. Li et al.¹⁷⁰ first reported room temperature optical gain from 2D perovskite (NMA)₂FA_{n-1}Pb_nX_{3n+1} (NMA = C₁₀H₇CH₂NH₃⁺). In these layered perovskite nanostructures, multiple QW phases naturally form an energy cascade, enabling an ultrafast energy transfer process from higher energy bandgap QWs ($n < 5$) to lower energy bandgap QWs ($n > 5$). They obtained tunable ASE ranging from 530 to 810 nm with low ASE threshold ($< 20.0 \mu\text{J}/\text{cm}^2$) [Fig. 14(h)]. Later, lasing based on these quasi-2D perovskite nanostructure has also been realized by researchers, e.g., Liang et al. investigated multicolor lasing from (BA)₂(MA)_{n-1}

Table 1 Lasing performance of perovskite.

Materials	Nanostructure	Laser mode	Emission wavelength	Threshold	Year	Ref.
CsPbX ₃	QD on silica sphere	WGM	400 to 700 nm	5 to 22 $\mu\text{J}/\text{cm}^2$	2015	127
CsPbBr ₃	CsPbX ₃ /SiO ₂ composite	Random	520 to 530 nm	40 $\mu\text{J}/\text{cm}^2$	2017	131
FAPbBr ₃	FAPbX ₃ /SiO ₂	Random	540 nm	413.7 $\mu\text{J}/\text{cm}^2$	2020	136
CsPbBr ₃	QD in silica sphere	Random/WGM	530 nm	430 $\mu\text{J}/\text{cm}^2$	2019	137
CsPbBr ₃	QD in capillary tube	WGN	530 to 540 nm	11 $\mu\text{J}/\text{cm}^2$	2015	138
CsPbBr ₃	QD in capillary tube	WGM	535 nm	0.9 mJ/cm ²	2016	139
FAPbBr ₃	QD in capillary tube	WGM	540 to 550 nm	0.31 mJ/cm ²	2019	132
CsPbBr ₃	DBR/CsPbBr ₃ QD/DBR	F-P	460 to 650 nm	9 $\mu\text{J}/\text{cm}^2$	2017	133
CsPbBr ₃	DBR/CsPbBr ₃ QD/DBR	F-P	520 nm	0.39 $\mu\text{J}/\text{cm}^2$	2017	134
FAPbBr ₃	Flexile DBR/FAPbBr ₃ film/DBR	F-P	552.7 nm	18.3 $\mu\text{J}/\text{cm}^2$	2017	135
MAPbBr ₃	DBR/MAPbBr ₃ film/Ag	F-P	552 nm	421 $\mu\text{J}/\text{cm}^2$	2020	140
MAPbX ₃	NWs	F-P	500 to 790 nm	0.22 $\mu\text{J}/\text{cm}^2$	2015	144
MAPbX ₃	NWs	F-P	551, 750, 777 nm	11 $\mu\text{J}/\text{cm}^2$	2015	49
CsPbX ₃	NWs and NPs	F-P	430, 532, 550 nm	5 $\mu\text{J}/\text{cm}^2$	2016	55
CsPbX ₃	NWs	F-P	420 to 710 nm	6.2 $\mu\text{J}/\text{cm}^2$	2016	145
CsPbX ₃	Micro/NRs	F-P	428 to 628 nm	14.1 $\mu\text{J}/\text{cm}^2$	2017	75
CsPbCl _{3-3x} Br _{3x}	NWs	F-P	480 to 525 nm	11.7 to 35.0 $\mu\text{J}/\text{cm}^2$	2020	146
MAPbI ₃	NPs	WGM	780 nm	37 $\mu\text{J}/\text{cm}^2$	2014	80
MAPbCl _x Br _{3-x}	Microdisks	WGM	525 to 558 nm	3.6 $\mu\text{J}/\text{cm}^2$	2015	150
MAPbI ₃	Microplatelets	WGM	780 nm	12 $\mu\text{J}/\text{cm}^2$	2016	151
MAPbBr ₃	Microplates	WGM	550 nm	20 $\mu\text{J}/\text{cm}^2$	2017	152
MAPbI ₃	Triangular nanoplatelets	WGM	780 nm	18.7 $\mu\text{J}/\text{cm}^2$	2019	148
CsPbX ₃	Nanoplatelets	WGM	400 to 700 nm	2.0 to 10.0 $\mu\text{J}/\text{cm}^2$	2016	149
CsPbI ₃	NSs	WGM	702 to 725 nm	0.3 mJ/cm ²	2018	86
CsPb ₂ Br ₅	Microplates	F-P	530, 540 nm	230 $\mu\text{J}/\text{cm}^2$	2020	153
		WGM		180 $\mu\text{J}/\text{cm}^2$		
CsPbX ₃	MSs	WGM	425 to 715 nm	0.42 $\mu\text{J}/\text{cm}^2$	2017	100
CsPbBr ₃	MSs	WGM	520 to 542 nm	203.7 $\mu\text{J}/\text{cm}^2$	2018	154
CsPbBr ₃	Nanocuboids	F-P	531 nm	40.2 $\mu\text{J}/\text{cm}^2$	2018	129
MAPbBr ₃	Pyramids	F-P	530 nm	26 $\mu\text{J}/\text{cm}^2$	2018	102
CsPbI ₃	Pyramids	F-P	720 nm	21.56 to 53.15 $\mu\text{J}/\text{cm}^2$	2019	103
MAPbBr ₃	Microwire array	F-P	554 nm	5.9 $\mu\text{J}/\text{cm}^2$	2016	156
MAPbX ₃	NW array	F-P	543 nm	12.3 $\mu\text{J}/\text{cm}^2$	2017	157
MAPbX ₃	Microplate array	WGM	510 to 780 nm	3.5 $\mu\text{J}/\text{cm}^2$	2016	158
CsPbX ₃	QDs array	WGM	534 nm	200 $\mu\text{J}/\text{cm}^2$	2018	159
MAPbX ₃	Microdisk array	WGM	510 to 650 nm	21.3 $\mu\text{J}/\text{cm}^2$	2019	160
CsPbBr ₃	CsPbBr ₃ microrod/Al nanoparticle	SP	540 nm	7.24 $\mu\text{J}/\text{cm}^2$	2017	172
CsPbBr ₃	CsPbBr ₃ /PEDOT:PSS/Au nanoparticle	SP	542 nm	157.6 $\mu\text{J}/\text{cm}^2$	2018	174
MAPbBr ₃	MAPbBr ₃ /Al ₂ O ₃ /TiN	SP	550 nm	10 $\mu\text{J}/\text{cm}^2$	2021	164
CsPbBr ₃	Ag/CsPbBr ₃ /Al ₂ O ₃ /Au	SP	534 nm	1.9 W/cm ²	2021	165
CsPbBr ₃	CsPbBr ₃ /Au	SP	495 to 520 nm	2.0 mW	2021	166
CsPbBr ₃	NWs	F-P	520 nm	8 $\mu\text{J}/\text{cm}^2$	2018	167
MAPbBr ₃	Micro/NWs	F-P	550 nm	15 $\mu\text{J}/\text{cm}^2$	2018	175
CsPbBr ₃	Nanoribbons	F-P (CW lasing)	2.34 eV	0.13 kW/cm ²	2020	168
CsPbCl ₃	DBR/CsPbCl ₃ /DBR	F-P	2.9 eV	12 $\mu\text{J}/\text{cm}^2$	2017	178

Materials	Nanostructure	Laser mode	Emission wavelength	Threshold	Year	Ref.
CsPbBr ₃	DBR/CsPbBr ₃ /DBR	F-P	2.3 eV	0.25 μJ/cm ²	2020	169
(BA) ₂ (MA) _{n-1} Pb _n I _{3n+1}	Quasi-2D perovskite flakes	F-P	630, 663, 687 nm	4.8 μJ/cm ²	2019	182
PEA ₂ A _{n-1} Pb _n Br _{3n+1} (A: MA, Cs)	UV glue/quasi-2D perovskite/glass	F-P	539 nm	10.5 μJ/cm ²	2021	183
PEA-FAPb _x Br _y	Quasi-2D perovskite on DFB	CW lasing	553 nm	32.8 μJ/cm ²	2020	171
NMA-FAPb _x Br _y			555 nm	4.7 μJ/cm ²		

Pb_nI_{3n+1} (BA = C₄H₉NH₃⁺) in 2019.¹⁸² Most recently, Liu et al. shrank the quasi-2D perovskites laser to the deep-subwavelength scale with 50 nm, which was the smallest room temperature all-dielectric laser.¹⁸³ They revealed the contribution from excitons and polarons to the high optical gain, which provided an insight into the design of next-generation integrated laser sources. Qin et al.^{171,184} found that the triplet excitons in hybrid quasi-2D perovskite have a lifetime up to 1 μs, which might cause the disappearance of the laser. Then, using a distributed-feedback (DFB) cavity with a high *Q* and triplet management strategies, they realized stable room-temperature CW lasing in quasi-2D perovskite films [Fig. 14(i)]. The representative works about perovskite lasers in recent years are summarized in Table 1. All of these progresses prove the potential of perovskite materials in micro/nano-lasers.

4 Conclusion and Outlook

Over the last few years, tremendous investigations have been carried out on metal halide perovskite materials, especially studies of the corresponding physicochemical properties and exploration of relevant applications in optoelectronic devices. In this review, we summarized the recent developments of the synthesis strategies, the morphological control, and lasing application of metal halide perovskite materials. The various synthetic methods for the fabrication of perovskite NCs have been investigated in previous researches, including the solution method and chemical deposition method. Moreover, the morphology of perovskite NCs can be controlled with different dimensions via adjusting the reaction conditions. Their structure-related optical properties were investigated on the single-particle with various structures as 0D, 1D, 2D, and 3D, enabling their potential in LEDs, solar cells, photodetectors, and lasers.

In spite of the tremendous advances in perovskite materials and perovskite-based lasers so far, there are still many issues to be further solved. The central issue of perovskite materials is their instability, which is the biggest obstacle for their industrialization. Although enormous work has been performed to enhance the stability of perovskites, such as the surface ligand modification or encapsulation method, the instability characteristic of the perovskites still limits their commercial applications. So far, the mechanisms of their decomposition are not yet clearly understood, hindering their further performance improvements. Another important issue related to lead halide perovskite materials is the urgent trend of reducing or removing the lead element due to its toxicity. For this purpose, some strategies have been proposed to constitute lead-free perovskites by possible substitutes using either homovalent elements such as Sn and Ge or heterovalent elements such as Bi and Sb.^{185,186} Unfortunately, the optoelectronic properties of lead-

free perovskites have not been effectively improved. Furthermore, the nucleation and growth mechanisms of perovskite NCs are yet to be revealed clearly, which is helpful to accurately control the morphology of the perovskite NCs for better understanding structure–property relationships. Last, but not least, theoretical explanation about the photophysics of perovskite NCs is necessary to better explain the quantum size effects of perovskite crystals, which could guide the research directions to regulate and control their electronic, optical, and defect properties.

The potential of perovskite materials in laser applications has been abundantly demonstrated. We reviewed a variety of laser cavities and summarized the dependence between the resonant cavity and the structure of perovskite NCs. Various linear and nonlinear perovskite lasers with an ultralow threshold have been realized in single perovskite NCs with different dimensions. Owing to the large gain coefficient and long-distance ambipolar carrier-transport, perovskites have great potential in electrically driven lasers, which have huge application value in integrated optoelectronic devices. But until now, all of the obtained perovskite lasers are pumped by laser excitation. The research about electrically pumped perovskite lasers has not been realized. Further investigation of resonant cavities, together with further reduction of the lasing threshold under optical excitation via optimization of the material properties, will boost the realization for electrically driven lasers of perovskites. The future trend of the perovskite-based laser is to integrate with optoelectronic components for further waveguide and signal processing. More importantly, the resonance and gain of perovskite materials and perovskite-based lasers need photophysical theory, which will inspire exploring the carrier relaxation and charge transfer processes of high-performance devices.

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