Review

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Quasi-2D lead halide perovskite gain materials toward electrical pumping laser

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Abstract: Quasi two-dimensional (2D) lead halide perovskite materials have shown outstanding performance in various photoelectric devices, including perovskite light-emitting diodes (LEDs) and perovskite optical pumping lasers. Due to the structure diversity of bulky organic cation, the photoelectric property for quasi-2D perovskite materials is flexible to be tuned. The spontaneously formed quantum-well structures allow rapid and efficient energy funneling from low- n domains to high- n domains, contributing to high exciton utilization for perovskite LEDs and low threshold for amplified spontaneous emission (ASE) and optical pumping perovskite lasers. Moreover, the hydrophobic bulky organic cations benefit to improve the environmental and operating stability owing to the better moisture tolerance and defects passivation ability. In this review, we will primarily introduce the quasi-2D lead halide perovskite materials from the structure to their optical and electrical properties. Then, we will focus on the advances of optical pumping lasers based on quasi-2D lead halide perovskite materials as gain mediums. Especially, more attention will be paid to perovskite lasers using distributed feedback (DFB) and distributed Bragg reflector (DBR) cavities. Furthermore, the key issues to realize quasi-2D perovskite-based electrical pumping lasers will be discussed.

Keywords: ASE; DBR; DFB; lasers; quasi-2D lead halide perovskite materials.

1 Introduction

Lead halide perovskite materials have attracted considerable attention for various photoelectric application. Owing to the high absorption coefficient, high carrier mobility and long carrier diffusion length, 3D metal halide perovskite materials have been widely used as light absorption and carrier separation layers for solar cells [1–4]. The recorded certified power converse efficiency of single-junction perovskite solar cell has improved to 25.5%. Combined with silicon to construct tandem solar cells, the power converse efficiency has further increased to 29.15%. Recently, quasi-2D perovskites with flexibly tunable photoelectric property are widely used to obtain perovskite LEDs with high performance. By introducing bulky organic cations in 3D perovskites, the dielectric shield and quantum effects are enhanced in 2D and quasi-2D perovskites, which benefit to accelerate the radiation recombination of exciton [5–12]. The spontaneously formed mixed domains allow energy funneling from low- n domains to high- n domains in the order of picosecond in quasi-2D perovskite films [13–16]. Then, the excitons concentrated on high- n domains can efficiently radiate to recombination. Up to now, the EQE of perovskite LEDs based on quasi-2D perovskites has exceeded 20% in the near-infrared emission region [17]. What's more, the stability of quasi-2D can be improved due to the hydrophobicity of bulky organic cation [17–21]. For instance, FAPbI₃-based near-infrared quasi-2D perovskite LEDs with a half-time of 46 h has been obtained at its peak EQE of 20.1% [21].

Perovskite materials also show huge potential as gain medium for lasers due to the high optical gain coefficients [22–24]. The tailorable bandgap and facile low-temperature solution processing are appealing for obtaining lasing from visible to near-infrared region and low-cost nonepitaxial fabrication. At present, low-threshold ASE and optical pumping lasing have been observed in 3D, 2D and quasi-2D Ruddlesden–Popper (RP) perovskite single crystals, nanocrystals, microrods, quantum dots and polycrystalline thin films combining with resonator geometries of whispering gallery mode (WGM) microspheres and

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nanoplatelets, Fabry–Pérot (FP) mode nanowires, DFB and DBR cavities under diverse conditions [25–35]. The energy transfer from low- n domains to high- n domains in quasi-2D perovskites is considered as an advantage to achieve low threshold ASE and laser [36–39]. Recently, the first stable continuous-wave (CW) lasing at room-temperature based on quasi 2D perovskite films using DFB optical cavities has been reported by Qin et al., showing the potential for quasi-2D-based perovskites achieving electrical pumping lasers in the future [40]. Typically, the low threshold for perovskite optical pumping laser is still in the order of nJ cm^{-2} . Thus, the corresponding threshold current density for perovskite electrical pumping laser diodes will be in the order of at least A cm^{-2} . Under such intense electrical excitation, both Joule heating- and electric-field-induced effects will be much more obvious than in common perovskite LEDs, leading in emission quenching and enhanced laser threshold [41, 42]. In addition, the broken carrier balance and enhanced Auger recombination could also be the barriers to obtain perovskite electrical pumping laser [42, 43]. Strategies to improve the poor stability for perovskite LEDs are also helpful for perovskite electrical pumping laser.

In this review, the structures, unique optical and electrical properties of quasi-2D lead halide perovskite materials will be briefly demonstrated at first. Then, we will mainly focus on the advances of optical pumping lasers based on quasi-2D perovskites as gain mediums. At last, the obstacles that hinder to obtain electrical pumping lasers will be discussed. Studies on emission behaviors under intense optical and electrical excitation and strategies to reduce Joule heating effect will be presented.

2 Quasi-2D RP lead halide perovskite materials

The general chemical structure of 3D lead halide perovskite materials is APbX_3 , where $[\text{PbX}_6]^{4-}$ octahedra skeletons are surrounded by small monovalent cations. X are halide ions of Cl^- , Br^- and I^- . Monovalent cations contain inorganic cation of Cs^+ and organic cations of methylammonium (MA^+) and formamidinium (FA^+). By introducing bulky organic cations, such as butylamine (BA^+), octylamine (OA^+), 1-naphthylmethylamine (NMA^+) and phenylethylamine (PEA^+), in 3D perovskite precursors, the crystal grow in some directions could be hindered. Then, 2D and quasi-2D RP perovskites form with the formula of $\text{L}_2\text{A}_{n-1}\text{Pb}_n\text{X}_{3n+1}$, where n value represents the number of inorganic PbX_6^-

perovskite sheets between bulky organic cations. With increasing the ratio of bulky organic cations, n -value of quasi-2D metal halide perovskites decreases (Figure 1a). 3D and 2D perovskites refer to the situations of $n = \infty$ and $n = 1$. Here, the different dimensionality of lead halide perovskites is related to their crystal structures, not defined from the physical size/shape. As known, the unique optical and electrical characteristics of photoelectrical materials are affected by the properties of their frontier orbitals. For lead halide perovskite materials, the conduct band (CB) and valence band (VB) are mainly decided by the inorganic skeleton $[\text{PbX}_6]^{4-}$. The bandgaps can be turned by selecting different halides. Emissions from ultraviolet to visible region have been obtained with halides varying from Cl to Br and I. Besides, the bandgaps can also be influenced by forming quasi-2D perovskites with different ratio of bulky organic cations. For instance, the bandgaps of perovskites based on cations of Cs and PEA range from 2.56 eV (0% PEABr) to 2.71 eV (120% PEABr) (Figure 1b) [16]. Moreover, the bulky organic cations were found to heavily influence the Pb-halide orbitals coupling and lattice distortion, leading in the bandgap variation. As demonstrated in Figure 1c, Chu et al. found that the VB energy levels could be reduced from -5.24 to -5.48 eV due to the decreased shortest Pb–Br bond by inserting a relative larger organic cation of ethylammonium (EA^+) in the CsPbBr_3 lattice [44]. As a result, the emission peak blue-shifted to the sky-blue region at 488 nm. In addition, Mao et al. investigated the tunable semiconducting properties by altering the stereochemistry of organic cations [45]. The relationship between bandgap and Pb–Br–Pb distorted angles was given in Figure 1d. From the single-crystal X-ray diffraction, they observed that the Pb–Br–Pb angles tended to decrease by introducing bulky organic cation of 4-(aminomethyl) piperidinium) (4AMP), whereas it tended to increase with more FA. Apart for influencing the bandgap, the insulating bulky organic cations could impede the carrier transport and lower thermal conductivity, which may cause charge accumulation and serious Joule heating under high current density [46]. Quasi-2D perovskites with high orientation may be one of the solutions to settle this issue [47, 48].

3 Advances of optical pumping lasers based on quasi-2D lead halide perovskite materials

Generally, a laser device consists three parts: pumping source, gain medium and optical cavities. When stimulated

Figure 1: (a) The schematic crystal structures of 3D, quasi-2D ($n = 2, 3$) and 2D ($n = 1$) RP perovskites with different ratio of bulky organic cations. (b) The VB and CB energy levels of quasi-2D perovskite based on $\text{CsPbBr}_{0.9}\text{Br}_{2.1}$ with different ratio of PEABr. Reproduced with permission [16]. Copyright © 2019, Springer Nature. (c) The variation of energy levels on insertion of a relative larger organic cation of EA in CsPbBr_3 lattice. Reproduced with permission [44]. Copyright © 2020, Springer Nature. (d) The trend of bandgap with changing Pb-Br-Pb angles on addition of different organic cations. Reproduced with permission [45]. Copyright © 2020, American Chemical Society.

by pumping source beyond the threshold, population inversion to achieve ASE occurs on the gain mediums. Then, the light transport in the optical cavities arouses optical feedback for laser oscillation. Coherent lasing emission can also origin from the steady-state leakage of exciton-polariton condensate [25, 49]. In addition to act as gain mediums, perovskite crystals with high gain coefficient can also form intrinsic optical cavities themselves. Benefiting from the microfabrication techniques, such as nanoimprint lithography, perovskite laser devices are facile to fabricate with high reproduction due to the soft lattice structure. Since Xing et al. reported the first ASE phenomenon based on 3D perovskites at room temperature in 2014 [50], lasing behavior has been reported for varied 3D perovskite-based gain mediums combined with the common optical cavities and well summarized in review articles [51–53]. For example, Xing et al. realized single-mode perovskite laser arrays based on CsPbBr_3 nanocrystals by using a top-down self-healing lithographic

patterning technique [29]. The single-mode microdisk laser demonstrated a low threshold of $3.8 \mu\text{J cm}^{-2}$ with a narrow full width at half maximum (FWHM) of 0.24 nm. Zhong et al. found that temperature was a key parameter to continuously grow CsPbBr_3 single-crystal films with excellent crystallinity and high quality [31]. By using chemical vapor epitaxial technology, they obtained large-scale CsPbBr_3 single-crystal ~ 300 nm grown on sapphire substrates. Benefiting from the high optical gain of 1255 cm^{-1} , low ASE threshold of $8 \mu\text{J cm}^{-2}$ was achieved. Further, they prepared microdisk array via a focused ion beam etching method. As a result, a single-mode laser with low threshold of $1.6 \mu\text{J cm}^{-2}$ was achieved on a $3 \mu\text{m}$ diameter disk. Tian et al. sandwiched single-crystalline perovskite thin films based on MAPbCl_3 between a pair of DBR mirrors constructed of alternating $\text{SiO}_2/\text{Ta}_2\text{O}_5$ to form strong optical confinement [30]. By changing the applied pressure on the substrates to adjust the thickness of the perovskite gain films, a deep-blue perovskite laser in the

range of 414–435 nm with threshold of $211 \mu\text{J cm}^{-2}$ was obtained. They owed such high threshold to the lower crystal quality and the pumping condition of a nanosecond pulsed pumping laser.

Compared to the 3D perovskite materials, quasi-2D perovskite materials with insulating and hydrophobic bulky organic cations are widely considered as candidates to improve both the PL efficiency and environmental stability. The first ASE behavior from quasi-2D perovskite materials was reported by Li et al. in 2018 [36]. Combined bulky organic cation of NMA with FA, they constructed quasi-2D RP perovskites with the formula of $(\text{NMA})_2(\text{FA})\text{Pb}_2\text{Br}_x\text{I}_{7-x}$. From the absorption and PL spectra, they observed mixed domains with different n -values in the formed perovskite films. To identify the possible cascade energy transfer process between different domains (Figure 2a), they used the femtosecond transient absorption measurement to track the excited-state dynamics. As shown in Figure 2b, the ground state bleaching (GSB) signs of $(\text{NMA})_2(\text{FA})\text{Pb}_2\text{Br}_x\text{I}_6$ -based perovskite thin films at 558 and 611 nm (corresponding to the emission peak of $n = 2$ and $n = 4$ domains) showed ultrafast decay, with the time constants of 0.42 ± 0.06 and 1.3 ± 0.1 ps. While the GSB sign at 710 nm (corresponding to the emission from $n > 5$ domains) exhibited a obvious rising kinetics with the time constant of 0.24 ± 0.02 ps and subsequent biexponential decay with the time constant of 36.9 ± 2.6 and 581 ± 63 ps. Due to the comparable timescale of bleaching recovery at low- n domains and excited-state formation at $n > 5$ domains, they confirmed the ultrafast energy transfer from low- n domains to $n > 5$ domains and considered this process would help to establish population inversion at $n > 5$ domains for achieving low ASE threshold. The ASE properties were measured from the PL intensity and spectra under different pumping density. As seen in Figure 2c and 2d, when the pumping intensity exceeded the threshold of $19 \pm 2 \mu\text{J cm}^{-2}$, the FWHM was narrowed from 34 to 6 nm. According to the intensity-dependent value of 1 under the ASE threshold, they further confirmed the energy transfer process from domains with different n values. Furthermore, the quasi-2D films showed excellent optical stability at the ambient conditions (25 °C and 40% moisture). After excitation under continue pulsed laser for 32 h, the ASE intensity maintained almost same.

In the following, we will briefly summary the advances of optical pumping lasers based on quasi-2D perovskite single-crystals and thin films. The key parameters, including threshold, working condition, FWHM, morphology, excited condition, and so on, were demonstrated in Table 1.

3.1 Optical pumping lasers based on quasi-2D perovskite single-crystals

High-quality single-crystal with the advantages of pure phase and low defect density is useful to understand the fundamental physical properties of perovskites and realize low-threshold perovskite lasers. In 2018, Raghavan et al. obtained millimeter-sized homologous single-crystals of $(\text{BA})_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}$ -based quasi-2D RP perovskites with different n values of 1, 2, 3 (Figure 3a) using the slow evaporation at a constant-temperature solution-growth technique [54]. With increasing the pumping energy, they found that PL spectra with narrowed FWHMs occurred for all the perovskite single-crystals (Figure 3b). Although the lasing thresholds for perovskite single-crystal with higher n value tended to increase, the estimated lasing thresholds for them were low as $2.85 \mu\text{J/cm}^2$ ($n = 1$), $3.02 \mu\text{J/cm}^2$ ($n = 2$) and $3.21 \mu\text{J/cm}^2$ ($n = 3$), respectively. They owed such low lasing thresholds to the step-pyramid-like morphology (Figure 3c and d) for these perovskite single-crystals and large difference in the refractive indices between perovskites and air. Thus, light scattering and reflection would form coherent feedback loops inside the single-crystals, leading to strikingly low-threshold lasing action at room temperature. Furthermore, in another work published in 2020, their group used the same crystal-growth method and reported random lasing at room temperature for 2D perovskite single-crystal microrods [55]. Besides the narrow FWHM of 0.1 nm and low threshold of $0.5 \mu\text{J/cm}^2$, the lasing emission exhibited negligible degradation after at least 2 h under continuous illumination, proving the potential of 2D perovskites in improving the lasing stability.

In addition, Liang et al. investigated the influences of Auger recombination and exciton-phonon interaction on the lasing behavior in mechanically exfoliated quasi-2D microflakes based on $(\text{BA})_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}$ single-crystals [56]. They found the thickness of as-exfoliated microflakes was important to obtain laser. Owing to the low radiative losses and good optical confinement, they established lasing in the nanometer-thick microflakes with thicknesses of 100–300 nm. Although the lasing threshold was also found to enhance with increasing the n value (Figure 4a), they only observed lasing emission from $n = 3$, 4, and 5 perovskites. There was no stimulated emission from $n = 1$ and 2 perovskites even at the low temperature of 78 K (Figure 4b), which is different from the phenomenon for homologous bulk single-crystals mentioned above. One reason should be their different sizes, morphology and crystal quality. Thus, the light confinement and laser

Figure 2: (a) The schematic diagram of cascade energy transfer from low- n domains with wide-bandgap to high- n domains with narrow bandgap. (b) Normalized bleaching kinetics of $(\text{NMA})_2(\text{FA})\text{Pb}_2\text{Br}_1\text{I}_6$ -based perovskite at the peaks of 558, 611, and 738 nm corresponding to $n = 2, 4$, and >5 domains. (c) Pump-fluence-dependent PL spectra for $(\text{NMA})_2(\text{FA})\text{Pb}_2\text{Br}_1\text{I}_6$ -based perovskite thin films with a thickness of ≈ 75 nm under the pulsed excitation laser (400 nm, 150 fs pulses). (d) Pump-fluence-dependent emission intensity for $(\text{NMA})_2(\text{FA})\text{Pb}_2\text{Br}_1\text{I}_6$ -based perovskite thin films. From the abrupt narrowing FWHM and increased emission intensity as a function of the pump fluence, ASE property for $(\text{NMA})_2(\text{FA})\text{Pb}_2\text{Br}_1\text{I}_6$ -based perovskite thin films can be identified. Reproduced with permission [36]. Copyright © 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

behavior showed difference. Since the Auger recombination and exciton-phonon interaction can be enhanced with decreasing the thickness of inorganic layer, they considered the increased laser threshold should be another reason for prohibiting laser in $n = 1$ and 2 perovskite microflakes. To verify this hypothesis, they carried out time resolved and temperature-dependent PL spectra. As seen in Figure 4c, for perovskite microflakes of $n = 3, 4$, and 5, the decay curves under high pumping fluence demonstrated faster decay constants of 1.9, 2.7, and 3.4 ns than in the case of low pumping fluence with the decay constants of 2.1, 3.8, and 3.6 ns. However, for $n = 2$ perovskite microflakes, the decay constants under high and low pumping fluence were similar, showing a low Auger recombination threshold

even smaller to the lasing threshold. Moreover, with increasing the temperature from 78 to 298 K, they observed that the PL spectra got broaden and more asymmetric for $n = 2$ perovskite microflakes, indicating the enhanced phonon scattering.

3.2 Optical pumping lasers based on quasi-2D perovskite thin films

Combined the anti-solvent engineering with diverse bulky organic cations, quasi-2D perovskite thin films with good morphology and reduced defect density are widely used as emitting layers for perovskite LEDs to obtain carrier and

Table 1: Summary of the key parameters for quasi-2D perovskites-based optical pumping lasers.

Quasi-2D perovskite	<i>N</i>	Threshold ($\mu\text{J cm}^{-2}$)	Working condition	FWHM (nm)	Morphology	Excited condition	Ref.
$(\text{BA})_2(\text{FA})_{n-1}\text{Pb}_n\text{Br}_{3n+1}$	3	27–31	RT	0.3	Microwire laser arrays	400 nm	[32]
$(\text{BA})_2(\text{MA})_{n-1}\text{Pb}_n\text{Br}_{3n+1}$	6	12–15	RT	0.21	Microring laser arrays	400 nm, 150 fs, 1000 Hz	[33]
$(\text{OA})_2(\text{MA})_{n-1}\text{Pb}_n\text{Br}_{3n+1}$	9	8.5	RT	0.3–0.6	Microplatelets	400 nm, 150 fs, 1000 Hz	[37]
$(\text{BA})_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}$	1	2.85	RT	0.22	Single-crystal	374 nm, 55 ps, 400 MHz	[54]
	2	3.02		~0.22			
	3	3.21		~0.22			
	4	–		–			
$\text{FA}-(\text{N-MPDA})\text{PbBr}_4$	1	0.5	RT	0.1	Single-crystal microrods	374 nm, 55 ps, 400 MHz	[55]
$(\text{BA})_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}$	3	2.6	78 K	0.9	Single-crystal	400 nm, 80 fs, 1 kHz	[56]
	4	–		–	microflakes		
	5	–		0.5–0.7	–		
$(\text{NMA})_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}$	64	36	RT	0.68	Thin films	337 nm, 0.8 ns, 10 Hz	[58]
$(\text{PEA})_2(\text{Cs})_{n-1}\text{Pb}_n\text{Br}_{3n+1}$	3	500	RT	0.8	Thin films	355 nm, 8 ns, 1000 Hz	[39]
$(\text{PEA})_2(\text{FA})_{n-1}\text{Pb}_n\text{Br}_{3n+1}$	8	32.8	RT	0.45	Thin films	337 nm, 3 ns	[40]
		59 (W cm^{-2})		1		CW	
$(\text{NMA})_2(\text{FA})_{n-1}\text{Pb}_n\text{Br}_{3n+1}$	8	4.7		0.45		337 nm, 3 ns	
		45 (W cm^{-2})		0.8		CW	

Figure 3: (a) The millimeter-sized quasi-2D RP perovskite single-crystals based on $(\text{BA})_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}$ with *n* values of 1, 2, 3. (b) Lasing behavior from the high-quality quasi-2D RP perovskite single-crystals. (c) and (d) Optical images of the bottom and top surface of the as-grown single-crystal, showing the formation of a “step-pyramid-like” morphology. Reproduced with permission [54]. Copyright © 2018, American Chemical Society.

exciton confinement. By the means of the proven technique, ASE and lasing behaviors have also been reported for the quasi-2D perovskite thin films with DFB and DBR cavities.

For example, Fu’s group reported quasi-2D perovskite lasing with the similar energy transfer process [32, 33]. One was for $(\text{BA})_2(\text{FA})_{n-1}\text{Pb}_n\text{Br}_{3n+1}$ -based quasi-2D perovskite thin films [32]. The best ASE performance with low threshold of $21.5 \mu\text{J cm}^{-2}$ was obtain for *n* = 3 perovskite film. They owned this to the suitable ratio of low and high-*n* values domains. Similarly, they fabricated nanowire laser arrays with each quasi-2D nanowire as high-quality FP

lasers showing identical optical modes and similar lasing thresholds ($27\text{--}31 \mu\text{J cm}^{-2}$). Another one was for $(\text{BA})_2(\text{MA})_{n-1}\text{Pb}_n\text{Br}_{3n+1}$ -based quasi-2D perovskite thin films [33]. The low ASE threshold of $13.6 \mu\text{J cm}^{-2}$ was obtained for *n* = 6 perovskite film. By using a facile polydimethylsiloxane template confined solution-growth method, they fabricated high-density large area microring laser arrays with individual quasi-2D perovskite microrings as high-quality WGM lasers showing similar low threshold ($12\text{--}15 \mu\text{J cm}^{-2}$) and identical optical modes. In addition, Lei et al. obtained optical pumping lasers from quasi-2D RP perovskite thin films with highly efficient Förster

Figure 4: (a) Temperature-dependent lasing threshold for $n = 3, 4, 5$ perovskite microflakes. (b) Lasing spectra of $n = 3, 4$, and 5 perovskite microflakes at $78 K$. No lasing was observed for $n = 2$ perovskite microflakes. (c) The transient PL decay curves for $n = 2, 3, 4, 5$ perovskite microflakes under low (red) and high (blue) excitations. (d) PL spectra for $n = 2$ perovskite microflakes under temperatures from 78 to $298 K$. Reproduced with permission [56]. © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

resonance energy transfer [38]. From GIWAXS results, they determined that the crystal structure of quasi-2D perovskites using dimethyl sulfoxide (DMSO) as solvent showed random orientation (Figure 5a), while the crystals of quasi-2D perovskites using N-methyl-2-pyrrolidone (NMP) as solvent preferred to demonstrate a horizontal orientation compared to the substrate (Figure 5b). Since the preferential horizontal orientation could reduce the distance between domains and transition dipole orientation, they considered a faster energy funneling for perovskites based on NMP, which were confirmed by the exciton dynamic obtained from the transient absorption measurement (Figure 5b and e). As a result, they observed low ASE threshold of $4.16 \mu J/cm^2$ for perovskite thin films using NMP (Figure 5f), while perovskite thin films using DMSO exhibited a relative high ASE threshold of $51.08 \mu J/cm^2$ (Figure 5c). Combined with a DFB cavity (Figure 5g), they eventually reported NMP-based quasi-2D RP perovskite lasers with low lasing threshold of $10 \mu J/cm^2$ (Figure 5h) and narrow FWHM of $1.7 nm$ (Figure 5i).

Moreover, Zhai et al. reported optical pumping DBR laser at room temperature from the segregated quasi-2D perovskite microcrystals forming in a sandwich structure consisting of perovskite/PMMA/perovskite [39]. Given the optical microscopy images, X-ray diffraction, absorption and PL spectra, they confirmed the self-organized segregated patterns of the spin-coated quasi-2D perovskites (region III in Figure 6a) consisted mixed domains with different n values. The cavity structure was shown in Figure 6b. With increasing the pump fluence above $500 \mu J cm^{-2}$, lasing occurred at the peak of $532 nm$ with narrow FWHM of $0.8 nm$ (Figure 6c) under pulsed excitation at $355 nm$ with pulse width of $8 ns$.

Since 2018, Lyeden et al. reported a series of researches on quasi-2D perovskites-based ASE and lasing. For example, they showed the ASE behavior with low threshold of $1.6 \mu J/cm^2$ in quasi-2D RP perovskite based on bulky organic cation of PEA in 2018 [57]. Later in 2019, they investigated the feasibility of NMA-based low-D perovskite using for electrically driven lasers [58]. Compared to the

Figure 5: (a), (b), and (c) Diagram, photobleaching dynamic and pumping fluence-dependent emission intensity of quasi-2D perovskite with random orientation using DMSO as solvent. (d), (e), and (f) Diagram, photobleaching dynamic and pumping fluence-dependent intensity of quasi-2D perovskite with preferential orientation using NMP as solvent. (g), (h) and (i) SEM morphology, lasing threshold and PL spectra of DFB lasers based on well-orientation quasi-2D perovskite thin films. Reproduced with permission [38]. © 2020 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

Figure 6: (a) Optical microscopy image of the segregated quasi-2D perovskites. (b) Schematic diagram of DBR cavity structure. (c) Pumping fluence-dependent emission intensity and FWHM for the segregated quasi-2D perovskites. Reproduced with permission [39]. © 2019 AIP Publishing.

roughness of MAPbI_3 -based 3D perovskite, they thought low-D perovskite films with more uniform morphology would benefit to achieve lasing emission. In addition, they demonstrated an effective method to obtain perovskite DFB lasers with perovskite materials as both gain medium and optical cavities [59]. The illustration of fabrication process was shown in Figure 7a. After perovskite films were spin-coated on the master grating of silicon oxide substrate and annealed, they were placed on the low-cost flexible substrates of polyethylene terephthalate (PET) and vacuum sealed in a plastic bag. Then, hot and press were treated on them, leading to the complete separation of perovskite films from the master grating. Finally, perovskite films with the desired DFB structure were constructed. The atomic force microscopy given in Figure 7b shows the well-defined grating patterns. Based on the transferred perovskite DFB films, lasing emission with FWHM ~ 0.7 nm (Figure 7c) and low threshold $\sim 14 \mu\text{J cm}^{-2}$ was achieved for the 385 nm grating (Figure 7d). Compared to the threshold $\sim 19 \mu\text{J cm}^{-2}$ for the control laser devices, this identified that the performance of transferred films can be well maintained with high repetition.

As CW optical pumping lasers is the footstone toward electrical pumping lasers, much efforts have been made to obtain CW optical pumping lasers [40, 60–64]. Recently, Qin et al. reported stable room-temperature CW optical

pumping lasers combined quasi-2D perovskite thin films with DFB cavities [40]. In the meantime, they studied the cause of “lasing death” and considered the singlet-triplet exciton annihilation as one of the possible intrinsic mechanism. The chosen bulky organic cations were PEA and NMA due to their different triplet energy levels compared to the formed quasi-2D perovskites. According to their early study [65], triplet exciton with long lifetime of nearly $1 \mu\text{s}$ exist in PEA-based perovskites due to its relative high triplet level, while the triplet excitons in NMA-based perovskites can be transferred to NMA. Thus, they proposed the long lifetime triplet exciton in PEA-based perovskites would impede population inversion for ASE and lasing, leading to the lasing death phenomenon. Given by the oxygen-sensitive characteristic for triplet, they carried out the ambient atmosphere-dependent ASE intensity and confirmed this perspective according to the declined ASE intensity in nitrogen for PEA-based perovskites. Since NMA can act as triplet quencher, the ASE intensity of NMA-based perovskites demonstrated almost same in both oxygen and nitrogen environment. They also observed lasing intensity for PEA-based films under continue excitation sharply decreased in nitrogen, indicating that singlet-triplet annihilation may be one of the intrinsic reasons causing lasing death. The low-threshold pump densities were 32.8 and $4.7 \mu\text{J cm}^{-2}$ for PEA- and NMA-based quasi-2D perovskite on

Figure 7: (a) The diagram of perovskite DFB films fabrication process. (b) Atomic force microscopy of a transferred perovskite DFB film. (c) PL spectra of perovskite DFB films with different transferred gratings. (d) Comparison of PL intensities under different pump fluences for a transferred and original films. Reproduced with permission [59]. Copyright © 2019, AIP Publishing.

DFB cavity with grating period of 250 nm under pulsed laser. However, given the similar thresholds under continue excitation for both PEA- and NMA-based quasi-2D thin films in air (Figure 8a), they considered the long lifetime triplet exciton may contribute to lasing emission for PEA through highly efficient up-converted to singlet exciton.

4 Challenges toward electrical pumping lasers based on quasi-2D perovskite materials as gain mediums

According to the great advances made in quasi-2D perovskites-based LEDs and CW optical pumping lasers, quasi-2D perovskite materials are promising as gain mediums in realizing electrical pumping lasers. As mentioned above, electrical pumping laser diodes need to operate under intense current density to reach population inversion and optical feedback. Compared to the common working current density $<100 \text{ mA cm}^{-2}$ for perovskite LEDs, the working current density for electrical pumping laser diodes in the order of at least A cm^{-2} is much higher. Although the structure of electrical pumping laser diodes can refer to the structure of perovskite LEDs, the sharp EQE roll-off and poor stability in perovskite LEDs should be more serious in electrical pumping laser diodes. Joule heating and electric-field induced effects will be more obvious in electrical pumping laser diodes, resulting in both severe emission quenching and increased threshold for optical gain. In this part, we will briefly discuss the remained challenges to

achieve quasi-2D perovskites-based electrical pumping lasers.

At present, only several reports are about the intense electrical excitation in perovskite LEDs. For instance, Kim et al. investigated the lasing and EL behaviors based on quasi-2D ($n_{\text{BAI}}:n_{\text{MAPbI}_3} = 20:100$) driven under short pulsed [34]. As seen in Figure 9a, the EL spectra in high energy region were broaden with current density increasing from 0.039 to 203 A cm^{-2} . They considered Joule heating as the cause for this phenomenon is similar to the variation in EL spectra depending on temperature ranging from 293 to 364 K under a direct current of 1 mA cm^{-2} . Moreover, they observed emission from transport layer under high current density (Figure 9b) and deduced the unbalance carrier originating from leakage current could affect the lasing generation under electrical excitation. Another study on quasi-2D-based perovskite LEDs under intense current density was carried out by Xu et al. [43]. As seen in Figure 9c, the EL intensity was enhanced with reducing the duty cycle from 1 to 0.1%, leading to the suppression of EQE roll-off. Compared to the EL intensity variation from 300 to 77 K, they also attributed this to the reduction of Joule heating under lower temperature. However, when the temperature was further reduced to 6 K, they considered the increased EL intensity roll-off originating from the enhanced Auger recombination. In addition, the threshold variation with increasing the temperature (corresponding to more Joule heating) and electric-field intensity were studied (Figure 9e and f) [41, 60]. As seen, the pulsed threshold fluence (F_{th}) increased under high temperature and the ASE intensity decreased with increasing the electric-field intensity.

Although there are still debates on the factors limiting ASE under intense current density, Joule heating is widely

Figure 8: (a) Lasing intensity variation for PEA-based perovskites thin films under continue illumination in air and nitrogen. (b) Lasing threshold for PEA- and NMA-based perovskites thin films (corresponding to P2F8 and N2F8) under CW laser excitation (Insert: schematic diagram of DFB cavity).

Figure 9: (a) EL spectra under different current density and temperature. (b) EL spectra under high current density, showing emission from transport layer and possible unbalanced carriers. Reproduced with permission [42]. Copyright © 2018, Springer Nature. (c) and (d) EL intensity-current density curves under different duty circle and environment temperature. Reproduced with permission [43]. Copyright © 2019, AIP Publishing. (e) Pulsed threshold fluence variation with increasing the temperature. Reproduced with permission [60]. Copyright © 2019, Springer Nature. (f) Evolution of emission as the applied electric-field intensity increased, showing quenching of ASE intensity. Reproduced with permission [41]. Copyright © 2015, AIP Publishing.

accepted as a major obstacle toward electrical pumping laser [66]. Operation at low temperature can significantly decrease this unwanted effect. However, the extrinsic carrier frozen may bring the problem of high driven

voltage. Therefore, strategies to efficiently reduce the thermal effect are desired and have been reported by several groups [67, 68]. For instance, Rand et al. combined doping charge transport layers, optimizing device

Figure 10: (a) EQE-current density curves of perovskite LEDs with different active areas, substrates and heat spreaders. (b) EQE-current density curves of perovskite LEDs with different substrates, spreaders under different electrical pulses. Reproduced with permission [68]. Copyright © 2020, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

geometry and heat spreaders and sinks to help heat dissipation and enhance EQE and stability at high current density [68]. As shown in Figure 10a, high EQE > 10% was achieved at the current density as high as 2 A cm^{-2} . Using the electrical pulses, they further improved the EQE at superhigh current density of 1 kA cm^{-2} to 1% (Figure 10b). Although no ASE sign was observed yet, the high EQE at intense electrical excitation shows perovskite materials promising toward electrically driven lasers. In the further, more effort should be made to develop strategies to relieve the thermal effect and study the essential factors limiting EQE and stability under high excitation intensity.

5 Conclusion

In this review, we primarily introduced the quasi-2D lead halide perovskites from the crystal structure to the optical and electrical properties. Due to the advantages, such as facile solution-fabrication engineering, nature quantum-well effect and high gain coefficient, quasi-2D perovskite materials show great potential toward nonpolar low threshold laser. Next, the advances for optical pumping lasers based on quasi-2D perovskite as gain medium were presented. Finally, we briefly discussed the challenges toward electrical pumping laser and the recent study on high optical and electrical excitation. To obtain electrical pumping lasers, more attention should be paid to investigate the behaviors under intense excitation and develop strategies to reduce their negative influence. Given the rapid development on perovskite materials and CW optical pumping lasers, we hold the confidence in perovskite-based electrical pumping lasers.

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