Low-dimensional perovskite materials and their optoelectronics

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Abstract
Three-dimensional (3D) organic–inorganic metal halide perovskite materials possess great potential applications for approaching efficient optoelectronics due to the unique optoelectronic properties of perovskite materials and cost-effective manufacturing possibilities of optoelectronics. However, the scientific and technical challenges of 3D perovskite materials were their inferior long-term stability, which hampered their practical applications. The low-dimensional perovskite materials composed of alternating organic and inorganic layers are one of the most credible paths toward stable perovskite photovoltaics and optoelectronics. In this short review, we first present a discussion of the crystal structure and nontrivial optoelectronic properties of the low-dimensional halide perovskites. The synthetic methods for the preparation of the low-dimensional halide perovskites are reviewed. After that, we focus on the recent development of perovskite photovoltaics, light-emitting diodes, and lasers by the low-dimensional halide perovskites. Finally, we outline the challenges of the low-dimensional halide perovskites and their applications.

KEYWORDS
lasers, light emitting diodes, low-dimensional, perovskite, photodetectors, solar cells

1 | INTRODUCTION

Three-dimensional (3D) halide perovskites have shown outstanding optoelectronic properties, such as strong light-harvesting capability, high light absorption coefficient (~10^5 cm^-1), low trap densities, high photoluminescence quantum yield (PLQY), narrow-band emission, and low exciton binding energies (<50 meV). These unique features enable 3D perovskite materials to have great potential applications in solar cells, photodetectors (PDs), light-emitting diodes (LEDs), and lasers. The growth of photovoltaics by 3D perovskites was unprecedented in the past decade. So far, over 25.2% power conversion efficiencies (PCEs) from perovskite solar cells (PSCs) by 3D perovskites with a small device area (<1 cm^2) have been demonstrated and photo-detectivities of ~10^15 Jones (1 Jones = 1 cm Hz^1/2 W^-1) have been reported from perovskite PDs (PPDs) by 3D perovskites. Moreover, a PLQY of over 90%, which was an ultra-high value, was reported from 3D perovskite quantum dots. The laser exhibited ultralow-threshold (220 nJ cm^-2) and high-quality factors (Q ≈ 3600) at a charge carrier density of 1.5 × 10^16 cm^-3 from 3D...
perovskite single-crystal were also demonstrated.\textsuperscript{29} However, the instability of 3D perovskite materials against moisture, light, and heat was still a key challenge, which hindered practical applications of 3D perovskite materials in photovoltaics and other optoelectronics.\textsuperscript{30–32} Many efforts have been devoted to enhancing the stability of 3D perovskite materials over the past several years. These approaches included interfacial modification,\textsuperscript{33,34} additives processing,\textsuperscript{35} lowering dimensions,\textsuperscript{36} tolerance factor adjustment,\textsuperscript{37} and encapsulation techniques.\textsuperscript{38} Among them, the concept of utilization of 3D perovskites embedded with low-dimensional perovskites has been demonstrated to be a promising approach for suppressing degradation of perovskite materials.\textsuperscript{39–43}

The most commonly studied low-dimensional perovskites were Ruddlesden–Popper (RP)-phase two-dimensional (2D) perovskites (Scheme 1(A)), which was composed of alternating organic and inorganic layers with a general formula, \(A_2A'_{n-1}B_nX_{3n+1}\), in which \(A\) is the longer organic cation (e.g., \(C_8H_{17}NH_3^+\) or PEA\(^+\)), \(A'\) is the shorter organic cation (e.g., \(CH_3NH_3^+\) or MA\(^+\)), \(B\) is the metal cation (e.g., \(Pb^{2+}\) or Sn\(^{2+}\)) and \(X\) is the halide anion (e.g., \(I^-\) or \(Cl^-\)), \(n\) is the numbers of metal halide layer between organic spacers (OSs) and organic layers.\textsuperscript{44} Alternatively, organic cations with two amino groups on both ends thereby removing the van der Waals gap between the layers (Scheme 1(B)), which could form a more stable Dion–Jacobson (DJ)-phase 2D perovskites with a general formula, \(AA'_{n-1}B_nX_{3n+1}\).

For alternating cations in the interlayer space (ACI) phase 2D perovskite with a formula, \(A_2A'_{n}B_nX_{3n+1}\), the shorter organic cation (MA\(^+\)) fills in both corner-sharing \([BX_6]\) metal halide network and interlayer (Scheme 1(C)).

Different from shorter organic cations (e.g., \(CH_3NH_3^+\) (MA\(^+\))) in 3D perovskite materials, longer organic cations (e.g., \(C_8H_{17}NH_3^+\) (PEA\(^+\))) with hydrophobic properties in 2D perovskite materials could slow down the degradation process in (2) step described below:\textsuperscript{45}

\[
PbI_2 + MAI \Leftrightarrow MAPbI_3 \quad (1)
\]

\[
MAPbI_3 + H_2O \Leftrightarrow MAPbI_3 \cdot H_2O \quad (2)
\]

\[
(4-n)MAPbI_3 + nMAPbI_3 \cdot H_2O + (2-n)H_2O \Leftrightarrow (MA)_4PbI_6 \cdot 2H_2O + 3PbI_2 \quad (3)
\]

\[
(MA)_4PbI_6 \cdot 2H_2O \Leftrightarrow PbI_2 + 4MAI + 2H_2O \quad (4)
\]

In addition, the electrically insulating nature of a long OS and perovskite conductor layers could lead to the natural multiple-quantum-well structures, as shown in Scheme 2(A,B). The OS layers serve as the potential “wall” and the perovskite layers serve as the potential “well”. In this case, the excitons are formed in the low-dimensional perovskite materials instead of free charge carriers due to

\begin{SCHEME}
\centering
\includegraphics[width=\textwidth]{scheme1.png}
\caption{Schematic of (A) Ruddlesden–Popper (RP) phase, (B) Dion–Jacobson (DJ) phase, and (C) alternating cations in the interlayer space (ACI) phase two-dimensional (2D) perovskite, where Gua is guanidinium.}
\end{SCHEME}
increased binding energy induced by quantum confinement and the generated electroluminescence in the low bandgap regions, which was confined effectively by higher energy gap regions, resulting in an efficient radiative charge carrier recombination. These unique properties could make the low-dimensional perovskite materials to be great candidates for LEDs and laser applications. As the organic layer thickness is increased, the quantum confinement effects are prominent, consequently resulting in blue-shifted absorption and emission. The large bandgap of pure 2D perovskite materials ($n = 1$) certainly renders themselves less desirable for solar cell applications. Therefore, the low-dimensional perovskites with a larger $n$ ($n > 3$) are proposed to be candidate materials for approaching efficient PSCs and PPDs.

In this short review, we firstly give a discussion of the crystal structures and optoelectronic properties of the low-dimensional perovskites. Second, the synthetic methods for the preparation of the low-dimensional halide perovskites are summarized. After that, the overview of the applications of solar cells, PDs, LEDs, and lasers are discussed. Finally, summary and key challenges toward high-performance optoelectronic applications of the low-dimensional perovskites are outlined.

2 | CRYSTAL STRUCTURE AND NONTRIVIAL OPTOELECTRONIC PROPERTIES

2.1 | Multiple quantum well structures in 2D perovskites

The low-dimensional perovskite materials are composed of a corner-sharing [BX$_6$] metal halide network sandwiched between the organic barrier planes of A. The variable $n$ is the stacking number of the [BX$_6$] network layers between two organic barrier layers. In the case of $n = 1$, the thickness of the [BX$_6$] network layers decreases to the scale of de Broglie wavelength, leading to a strong quantum confinement effect on the charge carrier's behaviors. As a result, the charge carriers transport in 2D perovskites with $n = 1$ is expected to be highly confined within the multiple quantum well (MQWs) structures. It is prevalently agreed that the electrons under this condition are freely movable along the in-plane direction but are restricted in the out-of-plane direction. Theoretically, 2D perovskites with $n = 1$ are ideal materials for LEDs application as it has natural the out-of-plane self-termination and is expected to exhibit 100% PLQY. Unfortunately, recent studies indicated that a low quantum yield observed from the low-dimensional perovskites suggested that the quantum losses were taken place within these materials. So far, this is still an open question because of the lack of fundamental knowledge, such as exciton reduced mass and the spatial extension of the charge carriers wavefunctions.

Various macroscopic techniques have been applied for the investigation of the MQWs structures of 2D perovskites. Mohite et al. demonstrated the importance of the Coulombic interactions in the low-dimensional perovskites. They proposed a generic formulation of the scaling of the exciton binding energy with the thickness of the [BX$_6$] layer, which is described by:

$$E_{b,1s} = \frac{E_0}{\left(1 + \frac{n-3}{2}\right)^2}$$

where $n = 3 - \gamma e^{-L_w/a_0}$, $a_0$ (4.6 nm) is the Bohr radius of 3D perovskites and the exciton ground state binding energy, $E_0$ (16 meV) is 3D Rydberg energy, $L_w$ is the physical width of quantum wells (QWs) for an infinite QW potential barrier (i.e., 0.641 for (BA)$_2$Pb$_2$I$_4$, 1.255 for
(BA)$_2$(MA)Pb$_2$I$_7$, where BA is butylammonium. In their model, the exciton is considered isotropic in an $n$-dimensional space ($1 < n < 3$) and the $\gamma$ is an empirical correction factor (i.e., 1.76) for the deviations from the pure quantum confinement regime. The factor $\gamma$ contains the electron and hole densities and dielectric confinement effects. Based on this model, the decrease in the value of $n$ would lead to a compression of the exciton wavefunction in the QW due to the dielectric confinement, which results in enhanced exciton binding energy. In the case of large $n$ values, they further predicted that the exciton binding energy in the low-dimensional perovskites is higher than the thermal fluctuation ($k_B T = 25.7$ meV) at room temperature as $n$ values are larger than 20 ([$\text{BX}_6$] layers of $\sim 12.6$ nm). Certainly, this work provided a fundamental step toward the design of novel low-dimensional perovskite materials for optoelectronic applications, such as solar cells, PDs, LEDs, and lasers.

2.2 Edge states in 2D perovskites

In addition to considering the bulk physics in the low-dimensional perovskite materials, the layer edge states (ES) with nontrivial conducting phenomena in the low-dimensional perovskites have also been studied. The ES in organic MQWs have untrivial properties. Fu et al. indicated that the ES states in HgTe QWs enable the dissipationless 1D electrical conduction. Blancon et al. reported that the free charge carriers at the ES within 2D perovskite $[(\text{C}_4\text{H}_9\text{NH}_3)\text{Pb}_2 \text{I}_{3n + 1}]$ can accelerate the exciton dissociation, which is favorable for solar cells application. Recently, Priya et al. provided the direct observations of the distinct conductive layer ES in the low-dimensional perovskite single crystals ($\text{C}_4\text{H}_9\text{NH}_3\text{Pb}_2\text{I}_4$ ($n = 1$) by conducting atomic force microscopy mapping techniques. As shown in Figure 1 (A), it was acceptable that the out-of-plane current in the low-dimensional perovskite materials would behave as an insulating feature due to MQWs. It was found that the out-of-plane current in the bulk terrace region was pretty low, but an unexpected current along the contour of the layer edges was observed. As shown in Figure 1(B), the current in the bulk terrace region was negligible but it became sharp at the layer edges. The huge difference between the bulk and the layer edge indicated that there were a number of free charge carriers presented at the ES layer and the bulk terrace region behavior as an insulating characteristic due to the insulating organic layers. The total collected mobile charges per area across the layer edges are calculated by:

$$F = \frac{4}{31 - \nu^2} \sqrt{R \delta}$$

$$\int I \cdot dy = \int \frac{dQ}{dt} \cdot dy = \int u \cdot dQ = u \cdot \int dQ$$

where $F$ is the force applied by the tip, $E$ is the Young’s modulus, $\nu$ is the Poisson’s ratio of the samples, $\delta$ is nanoindentation from the contact geometry, $R$ is the radius of tip curvature, $I$ is the detected current, $dQ$ is the differential of free charge carriers, and $u$ is the local scan rate, respectively. Thus, the contact area determined by the nanoindentation $\delta$ and the total free charge carriers is estimated from Equation (7). The charge carrier density at the ES was calculated to be $1.1 \times 10^{21}$ cm$^{-3}$, which is close to that of metals ($10^{21}$ cm$^{-3}$). Noted that the conducting nature at the layer edges would not change with the scan rates. For the insulating behavior of the bulk terrace region in the low-dimensional perovskite single crystals ($\text{C}_4\text{H}_9\text{NH}_3\text{Pb}_2\text{I}_4$ ($n = 1$), the electrons and holes are heavily bonded with a binding energy of $\sim 470$ meV, leading to the formation of the exciton or further recombined through either photogeneration or
injection from the electrodes. Such an investigation on the ES layer of the low-dimensional perovskite materials offers great opportunities for designing novel photovoltaic materials and their applications.

2.3 Crystal structure and bandgap in 2D perovskites

The crystal growth in the low-dimensional halide perovskite materials was also a significant difference compared to 3D halide perovskite materials. The latter one generally possesses multiple crystal planes, including the <100>, <110>, and <111> planes. However, in the case of pure 2D perovskite materials (n = 1), there are only the <100> groups that favor the continuous growth, giving rise to a poor charge carrier transport in the vertical direction. As n is increased to over 1, the <110> and <111> planes start to grow, which are favorable for charge carrier transport in the vertical direction since the oriented crystallization is parallel to the substrates, assisting in the growth of the charge-transport channels.

Compared to 3D perovskite materials, the low-dimensional halide perovskite materials show significantly different optical properties, which are dependent on dimensionality and size. The bandgap (E_g) of a semiconductor refers to the minimum energy required for an electron to be excited from the ground state in the valence band (VB) to the conduction band (CB). Owing to the quantum confinement effect, the E_g of the low-dimensional halide perovskites can be varied by the values of n (the thickness of [BX_n]). For example, increasing the number of [PbI_4] layer affords a narrowing of the E_g from 2.24 eV for n = 1 to 1.60 eV for n = 4, in the case of BA_2MA_n - 1Pb_nI_3n + 1 perovskite.

2.4 Influence of OSs on 2D perovskites

The long-chain OSs, as the crucial constituent, are essential to the optical and electronic properties of 2D perovskites. The commonly used OS cations are listed in Scheme 3.

It was reported that the methyl groups anchoring on the ammonium N atom have a significant influence on the device performance of PSCs by 2D perovskites. For example, Li et al. reported 2D perovskites based on phenylammonium (PA, primary ammonium), N-phenylmethylammonium (PMA, secondary ammonium), N,N-dimethyldiphenylammonium (tertiary ammonium), and phenyltrimethylammonium (PTA, quaternary ammonium) as OSs, and then fabricated PSCs by these 2D perovskites. They found that PSCs by 2D perovskites based on PTA OS with three methyl groups exhibited the highest PCEs, which was possibly due to the altered rigidity, size, and dielectric constant of the organic interlayer spacer.

In addition, it was found that the larger and more hydrophobic cations were beneficial for improving perovskite stability against moisture, although larger cations can adversely influence the device performance. It was further found that the organic cations with flexible aliphatic hydrocarbons exhibited a better stereochemical configuration than rigid aromatic hydrocarbons.

Studies have also demonstrated that controlling the QW width distribution was crucial to overcoming the performance-stability compromise in devices by 2D perovskites. Sargent et al. investigated how the OS influences QW width distribution. They investigated 2D perovskites based on allylammonium (ALA) (OS (OS) 16), PEA (OS 4), and BA (OS 17) organic cations and found that ALA could induce the formation of higher-n with monodisperse QWs in 2D perovskite film, which was ascribed to its single C=C bond could promote the intermolecular interactions between QWs. However, PEA + and BA + have higher formation energies due to the van der Waals and π-stacking interactions between cations, favoring the initial formation of low-n and polydisperse distribution QWs. Noted that polydisperse distribution QWs in 2D perovskites may act as ultrafast shallow traps for holes in materials, resulting in stronger recombination.

3 SYNTHETIC METHODS FOR THE PREPARATION OF THE LOW-DIMENSIONAL HALIDE PEROVSKITES

The first observation of quantum-confined 2D perovskite materials was reported by Tyagi et al. They obtained a single-layer thick crystalline 2D methylammonium lead bromide (MAPbBr_3) perovskite by colloidal synthesis, where the octylammonium bromide was used as the long-chain ligand. The synthetic procedures involved adding lead bromide, methylammonium bromide, and octylammonium bromide into a stirring solution of oleic acid (OLA) and 1-octadecene at 80°C. The final 2D perovskite nanocrystals were obtained by further dilution, filtration, precipitation, and redispersion. The nanostructures with various morphologies of perovskites can be obtained by controlling the purification of 2D MAPbBr_3 nanocrystals. The as-synthesized 2D MAPbBr_3 exhibited an obvious blue-shifted (~0.5 eV) absorption compared to 3D bulk MAPbBr_3, which is owing to the quantum confinements.
3.1 Solution phase method

Yuan et al.\textsuperscript{81} reported a one-pot synthetic method to prepare low-dimensional perovskite microdisks, \((R_1NH_3)_2[(R_2NH_3)_2PbBr_4]_{(n-1)}PbBr_4\), where \(R_1\) is a long octyl chain, \(R_2\) is an aromatic alkyl group. The synthesis details are shown in Scheme 4. Hydrobromic acid was added into a dimethylformamide (DMF) solution containing aromatic methylamine (benzylamine or thiophenemethylamine), octylamine, and lead bromide to generate a yellow pale precursor solution, which was then injected into vigorous hexane and then stirred for 5 min at room temperature, followed by acetone quenching. These 2D perovskites were only a few micrometers in lateral size with a thickness of 100–150 nm. They found that the as-synthesized perovskites exhibited significantly improved photophysical properties over 3D bulk perovskites, such as narrow deep blue emissions peaked at 403–413 nm, high PLQY of 53%, and good stability.

Dou et al.\textsuperscript{82} further reported a direct growth of atomically thin 2D hybrid perovskite \((C_4H_9NH_3)\text{PbBr}_4\) from the solution. In detail, a very dilute precursor solution \((C_4NH_9NH_3\text{Br} + \text{PbBr}_2 + \text{DMF})\) was dropped on the pre-cleaned surface of Si/SiO\(_2\) substrate and dried under 75°C. After that, CB was selected as a co-solvent to reduce the solubility of \((C_4H_9NH_3)\text{PbBr}_4\) in DMF, promoting crystallization. It was found that the crystallization process was uniform across the whole substrate due to the similar boiling point and evaporation rate of DMF and CB. This method overcame the limitations of the conventional exfoliation and chemical vapor deposition methods, which generally produced a thick perovskite plate. They found that the photoluminescence of the as-
synthesized 2D perovskites was changed with the structural relaxation or the lattice-constant expansion. Aharon et al. presented a facile low-temperature synthesis of 2D perovskite nanorods (NRs). The 2D NRs showed a shift to higher energies in the absorption and photoluminescence compared to 3D bulk perovskite. The formation mechanism of 2D NRs was analyzed by varying different ligands (from octylammonium to OLA). They further found that the bandgaps can be tuned from 1.90 to 2.26 eV by adjusting the halide from iodide to bromide.

Vybornyi et al. reported another synthetic method for the preparation of low-dimensional CH$_3$NH$_3$PbX$_3$, which was without polar solvents (DMF). The reaction between methylamine and PbX$_2$ was conducted in a non-polar solvent such as 1-octadecene ODE at an elevated temperature in the presence of oleylamine (OLM) and OLA as ligands in the precursor solution. The tetrahydrofuran solution containing methylamine and OLA was injected into the above precursor solution under vigorous stirring. The nanocrystals could be formed within seconds. They found that the low-dimensional perovskite nanocrystals exhibited either blue emitting or green luminescent by adjusting the amount of OLM. The platelets with $n = 3$ (approximately value) exhibited an absorption peaked at 450 nm and PL emission at 465 nm with a PLQY of 18%.

### 3.2 Vapor-assisted method

The low-dimensional perovskite materials prepared by vapor-assisted methods generally have better crystallinity and fewer impurities. Vapor-assisted methods have been demonstrated in 3D CH$_3$NH$_3$PbI$_3$ perovskites by Liu et al. They used a dual-source thermal evaporation system to evaporate PbCl$_2$ and CH$_3$NH$_3$I to produce 3D CH$_3$NH$_3$PbI$_3$, which was much uniform than solution-processed films. The first vapor-assisted method for preparing low-dimensional halide perovskite materials was reported by Guo et al. They fabricated 2D/3D mixed hybrid perovskite thin films by low-pressure vapor-assisted solution process. The solid-vapor reaction was conducted between spin-coated PEAI-doped PbI$_2$ thin film and MAI vapor under controlled pressure. Due to the weak van der Waals interaction between organic PEAI$^+$ cation and inorganic octahedral [PbI$_6$], the vapor MA$^+$ cation can accessibly intercalate into the octahedral [PbI$_6$] to form 2D perovskites. They found that the PEAI-doped perovskite films by MAI vapor treatment exhibited a similar absorption with a cut-off at 780 nm, suggesting that the predominant structure of MAI-vapor-CH$_3$NH$_3$PbI$_3$ was 3D CH$_3$NH$_3$PbI$_3$ rather than 2D CH$_3$NH$_3$PbI$_3$.

### 4 PSCs BY THE LOW-DIMENSIONAL HALIDE PEROVSKITES

#### 4.1 PSCs by low n quasi-2D perovskites ($n < 5$)

The low-dimensional halide perovskite materials have been intensively investigated in solar cells application over the last few years. The first PSCs by 2D halide perovskites was reported by Smith et al. In this study, the authors systematically investigated the origins of instability in traditional 3D CH$_3$NH$_3$PbI$_3$ and proposed a new concept of 2D layered perovskites, which was crafted by using PEAI molecules in place of MAI, as shown in Figure 2(A). The 2D layered perovskites with a structure of (PEA)$_2$(MA)$_2$Pb$_3$I$_{10}$ ($n = 3$) exhibited a dramatically enhanced moisture stability compared to 3D...
CH$_3$NH$_3$PbI$_3$. However, as indicated in Figure 2(B,C), the PCEs were lower than 5% from PSCs with a device structure of FTO/compact TiO$_2$/(PEA)$_2$(MA)$_2$Pb$_3$I$_{10}$ ($n=3$)/spiro-OMeTAD/Au, due to low absorption coefficient and poor carrier transport of (PEA)$_2$(MA)$_2$Pb$_3$I$_{10}$ ($n=3$). Interestingly, Sargent et al. found that the 2D layered perovskite with (PEA)$_2$(MA)$_2$Pb$_n$I$_{3n+1}$ ($n=60$) possessed superior long-term stability, and PSCs by this novel 2D perovskites exhibited decent PCEs. PCEs observed from PSCs based on (PEA)$_2$(MA)$_2$Pb$_n$I$_{3n+1}$ ($n=60$) were declined from initial 17.21 to 12.80% after 2 weeks, while PCEs from PSCs by 3D CH$_3$NH$_3$PbI$_3$ was reduced from initial 16.47 to 0.72% after 2 weeks in humidity air (RH 55%). However, these device performances were relatively poor since the insulating OS cations hindered charge transport.

Motivated by Smith's work, many researchers have recently focused on the development of alternative and effective spacer cations for balancing PCEs and stability of PSCs. Kanatzidis et al. reported a highly oriented 2D perovskite thin films, where n-buthylammonium (n-BA) cation was substituted by PEA cation. The 2D (n-BA)$_2$(MA)$_2$Pb$_n$I$_{3n+1}$ family of perovskite compounds ($n=1$ to $n=4$) was synthesized from a stoichiometric reaction between PbI$_2$, MAI, and n-BA. As shown in Figure 3(A,B), (n-BA)$_2$(MA)$_2$Pb$_n$I$_{3n+1}$ perovskite thin film showed a highly remarkable orientation. In the case of $n=1$, the (n-BA)$_2$(MA)$_2$Pb$_1$I$_4$ has a preferential growth along the (110) direction, and thus revealed the (00l) reflection (crystallizing along the [hk0] plane, thereby showing only the [00l] reflection). As the $n$ was larger ($n>1$), the n-BA cations have an intention to confine the perovskite growth within the planar layer, but the MA cations have an intention to expand the perovskite growth outside the layer. For compound as the $n=2$, the (0k0) reflections were split into the (111) and (202) reflections, which indicated that the vertical growth of the compound was pronounced. This effect was more serious for the compounds as $n=3$ and 4. Noted that the 2D (n-BA)$_2$(MA)$_2$Pb$_3$I$_{10}$ thin films remained stable after 2 months as it was evident from the X-ray diffraction (XRD) spectra shown in Figure 3(C,D). Such enhanced stability was attributed to the hydrophobicity of the long BA cation chain, preventing direct contact of adventitious water within perovskite. PSCs by 2D (n-BA)$_2$(MA)$_2$Pb$_3$I$_{10}$ with a device structure of FTO/TiO$_2$(compact)/TiO$_2$(mesoporous)/(n-BA)$_2$(MA)$_2$Pb$_3$I$_{10}$/spiro-OMeTAD/Au yielded a PCE of 4.02%. Such a low PCE was majorly attributed to poor charge carrier transport in the vertical transportation since the BA cation with a long chain serves as the potential barriers for charge carrier transporting. To circumvent the above issues, Liang et al. designed an alternative 2D perovskites with a short branched-chain, butylamine (iso-BA) spacer cations. The 2D (iso-BA)$_2$(MA)$_3$Pb$_4$I$_{13}$ ($n=4$) were prepared from precursor solution by mixing PbI$_2$, C$_3$H$_7$NH$_2$, HI, and CH$_3$NH$_3$I at a stoichiometric ratio of 4:2:2:3 in DMF. As shown in Figure 4(A), compared to the long-linear-chain of n-BA in 2D (n-BA)$_2$(MA)$_3$Pb$_4$I$_{13}$ ($n=4$), the short-branched-chain of iso-BA in 2D (iso-
BA$_2$(MA)$_3$Pb$_4$I$_{13}$ ($n = 4$) could facilitate charge carrier transporting efficiently in the vertical direction. As a result, PSCs by 2D (iso-BA)$_2$(MA)$_2$Pb$_3$I$_{10}$ with a device structure of FTO/C$_{60}$/iso(-BA)$_2$(MA)$_2$Pb$_3$I$_{10}$/spiro-OMeTAD/Au yielded a PCE of 8.82%, which was over 60% enhancements compared to that (5.38%) by 2D (n-BA)$_2$(MA)$_2$Pb$_3$I$_{10}$ with the same device structure. Moreover, the highest PCEs of 10.63% were observed

**FIGURE 3** The X-ray diffraction (XRD) patterns of thin film of (A) BA$_2$PbI$_4$ and (B) MAPbI$_3$, with respective diffraction planes. (C) XRDs of fresh and aged (BA)$_2$(MA)$_3$Pb$_4$I$_{13}$ film. (D) Images of different perovskite films before and after exposure to humidity. Reproduced with permission from Reference 62. Copyright 2015. American Chemical Society

**FIGURE 4** Schematic crystal structure of (A) (n-BA)$_2$(MA)$_3$Pb$_3$I$_{13}$ and (B) (n-BA)$_2$(MA)$_3$Pb$_3$I$_{13}$. The grazing-incidence wide-angle X-ray scattering (GIWAXS) images of (n-BA)$_2$(MA)$_3$Pb$_3$I$_{13}$ perovskite processed under (C) room temperature and (D) hot casting method. Reproduced with permission from Reference 89. Copyright 2017. Wiley

BA$_2$(MA)$_3$Pb$_3$I$_{13}$ ($n = 4$) could facilitate charge carrier transporting efficiently in the vertical direction. As a result, PSCs by 2D (iso-BA)$_2$(MA)$_2$Pb$_3$I$_{10}$ with a device structure of FTO/C$_{60}$/iso(-BA)$_2$(MA)$_2$Pb$_3$I$_{10}$/spiro-OMeTAD/Au yielded a PCE of 8.82%, which was over 60% enhancements compared to that (5.38%) by 2D (n-BA)$_2$(MA)$_2$Pb$_3$I$_{10}$ with the same device structure. Moreover, the highest PCEs of 10.63% were observed
from PSCs if the 2D (iso-BA)$_2$(MA)$_2$Pb$_3$I$_{10}$ was cast from a hot solution. Such enhanced PCEs were ascribed to the out-of-plane orientation of 2D (iso-BA)$_2$(MA)$_2$Pb$_3$I$_{10}$ thin films from the hot casting method, as indicated in Figure 4(B,C).

It was reported that the low-dimensional perovskite materials tend to be growth in the direction parallel to the substrates, which hinders the charge transport in the vertical direction. The two-step method in preparation of the low-dimensional perovskite materials was proposed to address the above issues. Mhaisalkar et al. reported a sequential deposition method to fabricate low-dimensional perovskite thin films.\textsuperscript{90} The fabrication method was presented in Figure 5(A). (IC$_2$H$_4$NH$_3$)$_2$PbI$_4$ thin film was first prepared on the substrates by spin-coating method, then the CH$_3$NH$_3$I solution was immersed on the surface of (IC$_2$H$_4$NH$_3$)$_2$PbI$_4$ to form (IC$_2$H$_4$NH$_3$)$_2$(CH$_3$NH$_3$)$_n$Pb$_{n+1}$I$_{3n+1}$ perovskite thin films. The 2D grazing-incidence wide-angle X-ray scattering (GIWAXS) was used to investigate the crystal orientation of (IC$_2$H$_4$NH$_3$)$_2$(CH$_3$NH$_3$)$_n$Pb$_{n+1}$I$_{3n+1}$ thin films, concerning the dipping duration. As indicated in Figure 5(B,C), the crystal orientation of perovskite thin films exhibited preferential crystal orientation in the out-of-plane direction as the dipping duration was last for 5 min. As a result, the highest PCE of 9.03% was observed from PSCs with a device structure of FTO/TiO$_2$ blocking layer/mesoporous TiO$_2$/[IC$_2$H$_4$NH$_3$)$_2$(CH$_3$NH$_3$)$_n$Pb$_{n+1}$I$_{3n+1}$/spiro-OMeTAD/Au.

The charge carrier transport along the out-of-plane direction is more difficult than that from the inorganic layer due to the insulating organic layer. It is accepted that additives could assistant the crystal growth of 2D perovskite films in the vertical direction.\textsuperscript{91,92} The first additive used in the preparation of the low dimensional perovskites was demonstrated by Chen et al.\textsuperscript{93} They added ammonium thiocyanate (NH$_4$SCN) into precursor solution containing BAI, PbI$_2$, and MAI, and then prepared perovskite thin films by a one-step method at room temperature.\textsuperscript{93} The boundary-free with larger grains oriented in the vertical direction was found in the (BA)$_2$(MA)$_2$Pb$_4$I$_{10}$ ($n = 3$) thin films processed with 1SCN additive (1 is the mole-ratio of NH$_4$SCN to (BA)$_2$(MA)$_2$Pb$_4$I$_{10}$). The perovskite thin films prepared by

![Figure 5](image-url)
the above method exhibited enhanced charge carrier mobility. As shown in Figure 6(A–D), they further fabricated PSCs based on (PEA)$_2$(MA)$_4$Pb$_5$I$_{16}$ ($n = 5$) film processed by different concentrations of NH$_4$SCN, and observed the optimal PCE of 11.01%. Such enhanced PCE was ascribed to the improved charge carrier mobility, vertically orientated 2D perovskite thin films. The crystallization and charge carrier mobility were further improved by NH$_4$SCN cooperated with NH$_4$Cl. A PCE of 14.1% was observed from PSCs by (PEA)$_2$(MA)$_4$Pb$_5$I$_{16}$ ($n = 5$).

It was also reported that solvent engineering was an effective method in controlling the morphology of quasi-2D perovskite films for achieving high crystallinity in the vertical direction. Kanatzidis et al. first demonstrated that the mixed DMF and DMSO solvent had a significant influence on the crystallinity, crystal orientation, grain size, and film quality of quasi-2D perovskite films. Gao et al. further explored its working mechanism. It was found that for DMF only solvent, perovskite, and intermediate complex co-exist in the film after anti-solvent treatment, and therefore it has a tendency to grow in different directions after thermal annealing. However, by using DMF:DMSO mixed solvent, there was only one intermediate complex formed after anti-solvent treatment. As a result, the quasi-2D perovskites from DMF:DMSO mixed solvent exhibited highly oriented crystallization. In addition, alternative low polarity and suitable boiling point solvent dimethylacetamide also exhibited a remarkable effect on crystallization kinetics. A mixture of DMF:DMSO: hydriodic acid (HI) has been reported to assist (3AMP)(MA$_{0.75}$FA$_{0.25}$)$_3$Pb$_4$I$_{13}$ film (where AMP is 3-(aminomethyl)piperidinium) with much more preferred perpendicular orientation and better crystalline quality, yielding PSCs with a PCE of 12.04%.

The cooperation of different large spacer cations was further demonstrated by Chen et al. Another spacer cation PEAI was added into BA$_3$MA$_4$Pb$_5$I$_{16}$ ($n = 5$) perovskite precursor solution. They found that PEAI in BA$_3$MA$_4$Pb$_5$I$_{16}$ ($n = 5$) perovskite precursor solution can assist preferential nucleation and reduce the nucleation density, resulting in perovskite with large grains, and consequently PSCs with high PCEs (a PCE of 14.09%).

By substitution of the para position of PEA with fluorene, Zhang et al. introduced new OS F-PEA in 2D perovskites. They found that F-PEA can assist perovskite sheets oriented well and enhance π orbital overlapping in the out-of-plane direction, leading to higher out-of-plane conductivity (Figure 7(A–D)). PSCs by F-PEA-based 2D perovskites with a device structure of FTO/c-TiO$_2$/(F-PEA)$_2$MA$_4$Pb$_5$I$_{16}$/spiro-OMeTAD/Au exhibited a PCE of 13.64% (Figure 7(E)).

The development of efficient OSs and understanding the charge transport mechanism are very important to further boost PCEs of PSCs. Xu et al. developed two multiple-ring spacer cations, 1-naphthalenemethylammonium (NpMA) and 9-anthracenemethylammonium (AnMA) for approaching high PCEs from PSCs. As shown in Figure 8 (A,B), the absorption and PL spectra of 2D perovskite thin films confirmed that 2D perovskite thin films based on either AnMA or NpMA OSs showed similar bandgaps and 3D-like phases. The slightly blue-shifted PL peak observed from 2D perovskite thin films based on NpMA OSs, as compared with that based on AnMA OS, indicated that the
formation of more layered perovskite phases in the NpMA-based 2D perovskite thin films. PSCs with a device structure of glass/ITO/PEDOT:PSS/perovskite/PCBM/BCP/Ag (n is 4) as shown in Figure 8(C) exhibited an ultrahigh open-circuit voltage ($V_{OC}$) of 1.24 V, a large short-circuit current density ($J_{SC}$) of 20.89 mA cm$^{-2}$, a fill factor (FF) of 66.35% and an overall PCE of 17.25% (Figure 8(D)). However, it was found that PSCs fabricated by AnMA-based 2D perovskite thin films exhibited a much low PCE of 14.47%. Such poor PCE was attributed to the decreased charge carrier lifetime, enlarged trap density, and increased charge carrier nonradiative recombination loss in AnMA-based 2D perovskite thin films.

It was further reported that high-quality 2D perovskite thin films with oriented out-of-plane direction can be achieved through the incorporation of a suitable amount of FA$^+$.101 As shown in Figure 9(A–D), Zhou et al. found that pristine (BA)$_2$MA$_3$Pb$_4$I$_{13}$ possess highly oriented crystal grain with the (111) planes, which is paralleled to the substrate surface, whereas the (BA)$_2$(MA$_{0.8}$FA$_{0.2}$)$_3$Pb$_4$I$_{13}$ thin films exhibited even stronger intensity in the out-of-plane direction. The PSCs by (BA)$_2$MA$_3$Pb$_4$I$_{13}$ thin films showed a PCE of 12.81%.

Zhang et al. reported PSCs by 2D BA$_2$(Cs$_{0.02}$MA$_{0.64}$FA$_{0.34}$)$_4$Pb$_5$I$_{16}$ ($n = 5$), which was prepared by spin coating a mixture solution of PbI$_2$ and PEI/C$_1$HI, and then followed with deposition of an MAI layer through spin-casting method. They found that a small amount of PEI$_2$PbI$_4$ can adjust the film morphology and crystallization of perovskite thin film. A PCE of over 15% was demonstrated from (PEI$_2$PbI$_4$)$_x$(MAPb$_3$I$_3$)$_{1-x}$ ($x = 2\%$) perovskite thin film. The enhanced device performance was ascribed to the improved film morphology and crystallinity.

The ACI-type 2D perovskites exhibited a reduced bandgap compared to RP perovskites with the same n values, which was due to larger crystal symmetry and different stackings.103 The first PSCs by ACI 2D perovskite (Gua)(MA)$_n$Pb$_{n+1}$I$_{3n}$ ($n = 1–3$) was reported by Kanatzidis et al. (where Gua is guanidinium), which showed a PCE of 7.26%.103 The film morphology of (Gua)(MA)$_3$Pb$_4$I$_{10}$ ($n = 3$) and QW’s distribution were further tuned with the assistance of MACl additive. As a result, an impressive PCE (18.48%) was observed.104

Recent advancement of PSCs by low n quasi-2D mixed perovskites ($n < 5$) is summarized in Table 1.

4.2 | PSCs by 2D/3D mixed perovskites

PSCs based on low n quasi-2D perovskite ($n < 5$) exhibited superior long-term operational stability but possessed poor PCEs compared to pure 3D perovskites. Toward the ends, PSCs based on 2D/3D mixed perovskites were developed to enhance PCEs and boost long-term stability as well.

Zhou et al. reported a two-step method in fabrication of (PEI$_2$PbI$_4$)$_x$(MAPb$_3$I$_3$)$_{1-x}$ perovskite thin film (where $x$ is 2%).112 which was prepared by spin coating a mixture solution of PbI$_2$ and PEI-HI, and then followed with deposition of an MAI layer through spin-casting method. They found that a small amount of PEI$_2$PbI$_4$ can adjust the film morphology and crystallization of perovskite thin film. A PCE of over 15% was demonstrated from (PEI$_2$PbI$_4$)$_x$(MAPb$_3$I$_3$)$_{1-x}$ ($x = 2\%$) perovskite thin film. The enhanced device performance was ascribed to the
FIGURE 8  (A) The absorption and (B) photoluminescence (PL) spectra of the 1-naphthalenemethylammonium (NpMA) and 9-anthracenemethylammonium (AnMA) films. (C) Device structure of NpMA and AnMA-based perovskite solar cells (PSCs). (D) The J–V curves of the two-dimensional (2D) PSCs. Reproduced with permission from Reference 72. Copyright 2020. American Chemical Society

FIGURE 9  Grazing-incidence wide-angle X-ray scattering (GIWAXS) patterns of the perovskite films of (BA)2(MA1-xFAx)3Pb4I13 x = (A) 0, (B) 0.2, (C) 0.4, (D) 0.6 films, respectively. (E) GIXRD patterns of BA2(MA)3Pb4I13 and (BA)2(MA0.8FA0.2)3Pb4I13 thin films. (F) The best J–V curve of BA2(MA)3Pb4I13 and (BA)2(MA0.8FA0.2)3Pb4I13-based perovskite solar cells by adopting invert device structure. Reproduced with permission from Reference 101. Copyright 2018. American Chemical Society
PEI$_2$PbI$_4$ doping at the molecular level rather than the merits from the low-dimensional structure.

Later on, Huang et al. reported MAPbI$_3$ incorporated with diethylammonium iodide ((CH$_3$CH$_2$)$_2$NH$_2$I) as an OS.$^{113}$ As shown in Figure 10(A), it was found that the DA mixed perovskite film exhibited stronger diffusion peaks at the (110) and (220) plans, with a smaller full width at half maximum as compared with those from MAPbI$_3$, indicating that these novel perovskite thin films possessed better crystallinity. PSCs based on (DA$_2$PbI$_4$)$_{0.05}$MAPbI$_3$ perovskite thin film exhibited a PCE of 19.05%, which was much higher than that (15.73%) by MAPbI$_3$ thin film (15.73%). The results are shown in Figure 10(B). The authors believed that enhanced PCEs were mainly ascribed to the large grains and fewer grain boundaries.

To further enhance both PCEs and stability, Chen et al. reported mesoscopic PSCs based on highly stable 2D/3D mixed perovskite (PEA$_2$PbI$_4$)$_x$(MAPbI$_3$) (where $x$ = 0.0017) thin film. As shown in Figure 11(A), no shift in the 2θ values and no new diffraction peaks from the (PEA$_2$PbI$_4$)$_x$(MAPbI$_3$) thin film were observed, indicating that a tiny of 2D perovskite was hard to alter the crystal structure of MAPbI$_3$ thin film. Moreover, (PEA$_2$PbI$_4$)$_x$(MAPbI$_3$) thin-film exhibited blue-shifted absorption and PL spectra (Figure 11(B,C)) compared to that of pure MAPbI$_3$ thin film, which is due to the presence of 2D sheet in (PEA$_2$PbI$_4$)$_x$(MAPbI$_3$) thin film. As shown in Figure 11(D), PSCs by 2D/3D mixed perovskite (PEA$_2$PbI$_4$)$_x$(MAPbI$_3$) (where $x$ = 0.0017) showed a PCE of 19.84% with a $V_{OC}$ of 1.146 V, $J_{SC}$ of 22.69 mA cm$^{-2}$, and FF of 0.7632, respectively. However, PSCs by PEA$_2$PbI$_4$ incorporated with a small amount of MAPbI$_3$ exhibited lower $J_{SC}$.

Most of the work on 2D/3D mixed perovskites was focused on MA cation at the A site. Lately, such an approach was extended to FA as well as to Cs$^{+}$ cation, and the final goal is to replace toxic lead as well as to maximize the device stability. Wu et al. reported Cs$_{0.1}$(FA$_{0.83}$MA$_{0.17}$)$_{0.9}$Pb(I$_{0.83}$Br$_{0.17}$)$_3$ incorporated Gua$^+$ to form CsGuaFAMA mixed cation perovskite.$^{115}$ As shown in Figure 12(A), all of the samples have a similar band edge at ~770 nm, which indicated that a tiny amount of Gua$^+$ does not affect the bandgap of perovskite thin films. However, 10% Gua$^+$ in perovskite thin-film showed the highest PL intensity and then PL intensities were gradually decreased as the concentration of Gua$^+$ was over 10%, which is due to the formation of 1D

### TABLE 1 Recent advancement of solar cells based on low n quasi-2D mixed perovskite ($n < 5$)

| Active layer | Device structure | PCE (%) | Stability | Year (ref) |
|--------------|-----------------|---------|-----------|------------|
| (Gua)(MA)$_3$Pb$_3$I$_{10}$ ($n = 3$) | ITO/PEDOT:PSS/active layer/PCBM/Al | 7.26 | N/A | 2017$^{103}$ |
| (3AMP)(MA)$_3$Pb$_4$I$_{13}$ ($n = 4$) | FTO/PEDOT:PSS/active layer/C60/BCP/Ag | 7.32 | N/A | 2018$^{105}$ |
| (PDA)(MA)$_3$Pb$_4$I$_{13}$ ($n = 4$) | ITO/PEDOT:PSS/active layer/C60/BCP/Ag | 13.0 | 90% of initial PCE after 1000 h in dark, 85% RH | 2018$^{106}$ |
| (F-PEA)$_2$(MA)$_3$Pb$_4$I$_{16}$ ($n = 5$) | FTO/m-TiO$_2$/active layer/spiro-OMeTAD/Au | 13.64 | 65% of PCE after 76 h, dark, 70°C | 2019$^{67}$ |
| (F-PEA)$_2$(MA)$_3$Pb$_4$I$_{16}$ ($n = 5$) | ITO/PEDOT:PSS/active layer/PCBM/BCP/Ag | 14.5 | 90% of initial PCE after 40 days, dark, 40–50% RH | 2019$^{68}$ |
| (Gua)(MA)$_3$Pb$_3$I$_{10}$ ($n = 3$) | FTO/c-TiO$_2$/active layer/spiro-OMeTAD/Au | 16.84 | 88% of PCE after 240 days, dark, 30–40% RH | 2019$^{107}$ |
| (Gua)(MA)$_3$Pb$_3$I$_{10}$ ($n = 3$) | FTO/c-TiO$_2$/active layer/spiro-OMeTAD/Au | 18.48 | 95% of PCE after 131 days, in dark, 40% RH | 2019$^{104}$ |
| (BDA)MA$_3$Pb$_4$I$_{13}$ ($n = 4$) | ITO/PEDOT:PSS/active layer/PCBM/BCP/Ag | 12.81 | 60% of PCE after 23 days, in dark, 50–60% RH | 2020$^{108}$ |
| (BA)$_2$(MA)$_3$Pb$_4$I$_{13}$ ($n = 4$) | ITO/PEDOT:PSS/active layer/PCBM/PEI/Al | 15.08 | N/A | 2020$^{109}$ |
| (BA)$_{1.6}$(MA)$_{3.4}$Pb$_4$I$_{13}$ ($n = 4$) | FTO/PTAA/active layer/C$_{60}$/BCP/Ag | 15.44 | 97% of PCE after 85 days, N$_2$, in dark | 2020$^{110}$ |
| (ThMA)$_2$(FA)$_3$Pb$_3$I$_{16}$ ($n = 5$) | ITO/PEDOT:PSS/active layer/PCBM/BCP/Ag | 19.06 | 96% of PCE after 576 h, N$_2$, in dark | 2020$^{111}$ |

**Abbreviation:** PCE, power conversion efficiency.
GuaPbI₃, as illustrated in Figure 12(B). The time-resolved PL studies (Figure 12(C)) demonstrated that the bimolecular radiative recombination was gradually enhanced by embedding larger Gua⁺ into perovskite thin film. In addition, by incorporating Gua⁺, the suppressed defect, elongated PL lifetime, reduced energy disorder in the band edge, enlarged charge recombination resistance, and suppressed trap state density were found in the

**FIGURE 10** (A) The X-ray diffraction (XRD) patterns of perovskite without and with 7, 10, and 15% diethylammonium iodide (DAI) contents. (B) The J–V characteristics of the perovskite devices without and with 10% DAI contents. Reproduced with permission from Reference 113. Copyright 2019. Royal Society of Chemistry

**FIGURE 11** The X-ray diffraction (XRD) patterns of MAPbI₃ and (PEA₂PbI₄)₀.₀₁₇(MAPbI₃) films spin-coated on FTO-coated glass substrate. (B) UV–visible absorption and (C) the steady-state photoluminescence (PL) spectra of MAPbI₃ and (PEA₂PbI₄)₀.₀₁₇(MAPbI₃) films spin-coated on glass. (D) The J–V curves of the best-performing devices employing MAPbI₃ and (PEA₂PbI₄)₀.₀₁₇(MAPbI₃). Reproduced with permission from Reference 114. Copyright 2017. American Chemical Society
resultant perovskite thin film. As a result, enlarged $V_{OC}$ from 1.11 to 1.19 V and consequently an enhanced PCE of 21.12% was observed.

Recent advancement of PSCs by 2D/3D mixed perovskites is summarized in Table 2.

### 4.3 PSCs by 2D/3D perovskites with a bilayer structure

It has been reported that the effective mitigation of defects in perovskite thin films is essential to further enhance the device performance of PSCs. Therefore, the formation of 2D/3D heterojunction structure is a promising method to improve the perovskite absorber layer with lower defect densities and longer charge carrier lifetimes.

Zhao et al. explored a series of diammonium iodides, NH$_3$I(CH$_2$)$_4$NH$_3$I (C4), NH$_3$I(CH$_2$)$_8$NH$_3$I (C8), and NH$_3$I( CH$_2$)$_2$O(CH$_2$)$_2$NH$_3$I (EDBE) (Figure 13(A)) to passivate perovskite surface and grain boundaries.$^{121}$ It was found that the molecular structure of diammonium salts has a profound effect on the surface morphology and phase purity of perovskite thin films (Figure 13(B)). Also, C4- and EDBE-caped perovskite thin films showed phase transformation during treatment, which is unfavorable for charge carrier transportation. The C8-caped perovskite thin-film could efficiently passivate perovskite thin film and thus, the C8 salt was used to dope the electron transport layer PCBM. As a result, PSCs by the C8-caped perovskites thin film exhibited a PCE of over 17.60%.

Chen et al. reported an innovative facile way to prepare PEAM$_4$Pb$_3$ capping layer on the top of 3D Cs$_{0.05}$(FA$_{0.83}$MA$_{0.17}$)$_{0.95}$Pb(I$_{0.83}$Br$_{0.17}$)$_3$ 3D perovskite thin film.$^{122}$ It was found that the PEAM$_4$Pb$_3$ layer upon 3D perovskite thin film can simultaneously improve the device performance and stability of PSCs by 2D/3D perovskites, as shown in Figure 14. PSCs by the 2D/3D perovskites treated by 1 mg ml$^{-1}$ PEAI solution showed a PCE of 18.51%, which was ~10% enhancement compared to that by 3D perovskites. The enhanced device performance was attributed to the reduced nonradiative recombination loss in the 2D/3D perovskites. Similar work was also reported by Jiang et al.$^{123}$ They prepared a PEAM$_4$Pb$_3$ layer on top of FA$_{1-x}$MA$_x$Pb$_3$ thin film to suppress the surface defects of perovskite thin films for approaching efficient PSCs, as shown in Figure 15(A,B). It was found that a thin PEAI layer can not only stabilize the α phase of FAPbI$_3$, but also can slow down the degradation of underneath 3D perovskite, which was confirmed by both XRD and XPS studies (Figure 15(C,D)). The PSCs by 2D/3D perovskites with a device structure of ITO/SnO$_2$/

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**FIGURE 12** (A) UV–vis spectra, (B) photoluminescence, and (C) the time-resolved photoluminescence (TRPL) spectra of the solid perovskite films with different Gua/CsFAMA ratios. (D) Energy level diagrams for different materials. (E) The J–V curves. (F) The power conversion efficiency (PCE) distribution of perovskite solar cells (PSCs). Reproduced with permission from Reference 115. Copyright 2019. Royal Society of Chemistry
perovskite/spiro-OMeTAD/Au shown a PCE of 23.56% with a $V_{OC}$ of 1.16, a $J_{SC}$ of 24.9 mA-cm$^{-2}$, and an FF of 81.4%.

Recent advancement of PSCs by 2D/3D perovskites with a bilayer structure is summarized in Table 3.

### TABLE 2 Recent advancement of solar cells based on 3D/2D mixed perovskites

| Active layer | Device structure | PCE (%) | Stability | Year (ref) |
|--------------|------------------|---------|-----------|------------|
| (MAPbI$_3$)$_{0.98}$(PEI)$_{0.02}$ | ITO/PEDOT:PSS/active layer/PC$_{61}$BM/LiF/Ag | 15.2 | 84% of PCE after 14 days, dark, 50% RH | 2015$^{112}$ |
| (AVA)$_{0.03}$(MAPbI$_3$)$_{0.97}$ | FTO/TiO$_2$(compact)/TiO$_2$(meso)/active layer/ZnO$_2$/Carbon | 10.10 | 100% of PCE after 10,000 h, 55°C, continuous light soaking | 2017$^{75}$ |
| BA$_{0.09}$(FA$_{0.8}$Cs$_{0.2}$)$_{0.91}$Pb$_4$(I$_{0.6}$Br$_{0.4}$)$_3$ | ITO/SnO$_2$/PC$_{61}$BM/active layer/spiro-OMeTAD/Au | 17.2 | 80% of PCE after 1000 h, air, dark | 2017$^{116}$ |
| CA$_2$MA$_{30}$Pb$_4$I$_{121}$ | ITO/SnO$_2$/active layer/spiro-OMeTAD/MoO$_3$/Ag | 6.6 | 59% of PCE after 264 h, dark, 77% RH | 2018$^{117}$ |
| (DA)$_{3}$Pb$_4$I$_{0.05}$(MAPbI$_3$)$_{0.95}$ | ITO/SnO$_2$/active layer/spiro-OMeTAD/MoO$_3$/Ag | 19.05 | 80% of PCE after 60 days, dark | 2019$^{113}$ |
| (PEA)$_{3}$Pb$_{1.017}$(MAPbI$_3$)$_{0.983}$ | FTO/c-TiO$_2$/m-TiO$_2$/active layer/spiro-OMeTAD/Ag | 19.84 | 96% of PCE after 100 h, dark | 2014$^{114}$ |
| Gu$_{0.1}$(FA$_{0.8}$MA$_{0.2}$)$_{0.17}$Pb$_4$(I$_{0.6}$Br$_{0.4}$)$_3$ | ITO/SnO$_2$/active layer/spiro-OMeTAD/MoO$_3$/Ag | 21.12 | N/A | 2019$^{115}$ |
| (HEA)$_{0.9}$CBA$_{0.1}$(C$_{0.5}$I$_{0.5}$)$_{12}$ | FTO/c-TiO$_2$/m-TiO$_2$/active layer/spiro-OMeTAD/Au | 18.75 | 90% of PCE after 1500 h, air, dark | 2020$^{118}$ |
| (HEA)$_{0.9}$FBA$_{0.1}$(C$_{0.5}$I$_{0.5}$)$_{12}$ | FTO/c-TiO$_2$/m-TiO$_2$/active layer/spiro-OMeTAD/Au | 17.67 | 85% of PCE after 1500 h, air, dark | 2020$^{118}$ |
| iBA$_{0.1}$MA$_{0.9}$PbI$_3$ | ITO/NiOx/active layer/PCBM/ZrAcac/Ag | 17.48 | 75% of PCE after 35 days, air, dark | 2020$^{119}$ |
| PEABr-treated MAPbI$_3$ | ITO/TAPC/PEABr/active layer/PC$_{61}$BM/Bphen/Ag | 19.46 | 86.1% of PCE after 1300 h, N$_2$, dark | 2020$^{120}$ |

Abbreviations: 2D, two-dimensional; 3D, three-dimensional; PCE, power conversion efficiency.

**5 | PEROVSKITE PDS BY THE LOW-DIMENSIONAL HALIDE PEROVSKITES**

PDs refer to light-responsive devices that convert optical signals into electric signals, which is central to modern science and technology because of their great applications, including imaging, vision, and digital display technology.

PDs could catalog as the two-terminal and three-terminal devices based on device architecture. The two-terminal devices are composed of photodiode and photoconductor, and the three-terminal devices refer to phototransistors with source, drain, and gate electrodes. PDs with a two-terminal device structure generally provide a low driving voltage and fast photoresponse owing to a narrow electrode spacing (~100 nm). Noted that the photodiodes-based PDs cannot exceed 100% external quantum efficiency (EQE) since no additional charge injection occurs under the reverse bias, but it happens in photoconductor-based PD. The photoconductive gain ($G$) is given by the ratio of trapped carriers ($\tau$) to the transit time of the transported carriers ($\tau_{transit}$) through the device. The $G$ is described as $G = \frac{\tau}{\tau_{transit}}$. Generally, phototransistor-based PDs need a high driving voltage to obtain decent device performance due to their wide electrode space.

Currently, PDs market was mainly dominated by photodiodes based on crystalline inorganic semiconductors, such as silicon, Ge, and InGaAs. However, the preparation of these inorganic PDs was not a cost-effective process. Moreover, some of these inorganic-based PDs are required to be operated at extremely low temperatures, which substantially limit their applications. Thus, it is necessary to develop high-performance PDs with easy manufacturing and cost-effective techniques.

In recent years, the emerging of the low-dimensional halide perovskite single-crystal has attracted tremendous attention in the fabrication of PDs owing to their
remarkable optical and electronic properties, such as reduced defects and enhanced charge transfer, compared with the low-dimensional halide perovskite polycrystalline films. However, the challenges in reducing dark current and enhancing photocurrent along the direction of the charge transporting need to be addressed.

Feng et al. developed a series of perovskite nanowire of (BA)2(MA)n−1PbnI3n+1 with a pure (101) crystallographic orientation. They found that (BA)2(MA)n−1PbnI3n+1 single-crystal exhibited fewer surface defects and grain boundaries, which allow efficient charge transport in the inorganic layer [PbI6]. In addition, the BA+ as an insulating organic barrier is responsible for suppressing the dark current and the exotic crystal edges in perovskites are a benefit for efficient exciton dissociation. Thus, (BA)2(MA)n−1PbnI3n+1 single-crystal-based PDs exhibited a high responsivity of 1.5 × 105 A W−1 and a specific detectivity of over 7 × 1015 Jones (cm Hz1/2 W−1).

Zhu et al. reported large-scale low-dimensional halide perovskite (C4H9NH3)n(CH3NH3)n−1PbnI3n+1 single crystals by the potassium ions assisted controllable crystal growth during the precipitation process. They found that the (C4H9NH3)n(CH3NH3)n−1PbnI3n+1 single crystals were prepared from the water–air interface. The crystallization process occurred through nucleation and
crystal growth. The nucleation rate $J$, is exponentially related to the energy barrier for nucleation, $\Delta G$, which is expressed by an Arrhenius-type equation:

$$J = \Lambda \cdot \exp \left( - \frac{\Delta G}{k_B T} \right)$$

where $\Lambda$ depends on supersaturation, $k_B$ is the Boltzmann constant, and $T$ is temperature. The energy barrier for nucleation is different in bulk solution and at the water–air interface. The energy barrier for nucleation in bulk solution is given by:

$$\Delta G_{\text{bulk}} = \frac{16}{3} \pi \sigma^3 \left( \xi - E_C - \xi_A + k_B T \cdot \ln \left( \frac{M_p}{M_A} - J^2 M_S^{-1} \right) \right)^{-2}$$

where $\xi$ is the cohesive energy of precursor molecules in the cluster, $\xi_A$ is the energy of precursor molecules, $\sigma$ is the surface tension coefficient, and $M_p$ and $M_A$ are the total molar concentrations of solvent and precursor molecules, $\chi$ is the increased energy of precursor molecules induced by extra tensile elastic stress at the water–air interface as indicated in Figure 16(C). Thus, a higher nucleation probability at the water–air interface is expected in contrast to that in the bulk solution. Therefore, in the process of crystallization, the self-assembly $\text{C}_4\text{H}_9\text{NH}_3^+$ precursor cation at the water–air interface acts as a soft template helping the growth of nanostructures. The higher solvation energy of precursor molecules at the asymmetric water–air interface offers higher chemical potentials, leading to a low energy barrier and faster in-plane growth. Consequently, inch-size freestanding quasi-2D perovskite single crystals have been achieved at the water–air interface. The PDs based on the quasi-2D perovskite single crystals with the smallest QW thickness ($n = 1$) exhibited a strikingly low dark current of $\sim 10^{-13}$ A, higher on/off ratio of $\sim 10^4$, and faster rise time of $\sim 1.7 \mu$s and drop time of $3.9 \mu$s. Similar work was reported by Liu et al. They synthesized high-quality 36 mm sized 2D (PEA)$_2\text{PbI}_4$ by inducing extra surface tension. As shown in Figure 17(B), the XRD patterns indicated that the top plane and side plane was well-defined with the (001) series and the
series of reflections, respectively. They found that PDs by the (001) plane of (PEA)$_2$PbI$_4$ exhibited better photoresponse compared to that by the (010) plane, with a dramatically low dark current of $3.06 \times 10^{-12}$ A under a bias of 5 V. Such low dark current was ascribed to boundary-free of (PEA)$_2$PbI$_4$ single crystals. The (PEA)$_2$PbI$_4$-based PDs possessed the highest detectivity of $1.89 \times 10^{15}$ Jones under a bias of 5 V, which was a record-high sensitivity for PPDs.

Recent advancement of PPDs by the low-dimensional perovskites is summarized in Table 4.

6 | PEROVSKITE LEDs BY THE LOW-DIMENSIONAL HALIDE PEROVSKITES

It was reported that perovskite LEDs based on quasi-2D perovskites could display better device performance than that by 3D counterparts. Emma et al. reported a solution-state synthetic route to fabricate well-defined and single-phase white light emitters. They demonstrated that layered PbBr$_2$ perovskites crystals of N1-methylethane-1,2-diammonium (N-MEDA)[PbBr$_4$] and N1-methylpropane-1,3-diammonium (N-MPDA)[PbBr$_4$] formed the <110> and <001> oriented structures. The <001> sheets displayed sharp blue emission at approximately 420 nm, whereas the <110> sheets showed broad white-light emission (maximum at 558 nm, PLQY $\approx 0.5\%$) covering the whole visible spectrum, as shown in Figure 18(A–D). Thus, a broader emission within the entire visible spectrum was observed from corrugated lead halide sheets in (N-MEDA)[PbBr$_4$] 2D perovskite. Through changing the ligands from N-MEDA to EDBE, they reported “warm” and “cold” white-light-emitting crystals with quantum efficiencies of 2 and 9% from perovskite LEDs based on perovskites with Cl and Br anions. Their observations offered a...
flexible platform for studying the phenomenon of broadband emission from 2D perovskite materials.

In the $\text{PEA}_2(\text{MA})_{n-1}\text{Pb}_n\text{Br}_{3n+1}$ system, Byun et al. demonstrated that the quasi-2D perovskites displayed a much higher current efficiency and luminescence than 3D $\text{MAPbBr}_3$ and 2D $\text{PEA}_2\text{PbBr}_4$. They reported a series of precursor solutions with different MA:PEA ratios to prepare $\text{PEA}_2(\text{MA})_{n-1}\text{Pb}_n\text{Br}_{3n+1}$ thin films. As indicated in Figure 19, it was found that perovskite LEDs based on the quasi-2D perovskite thin film with $\text{MAPbBr}_3:(\text{PEA})_2\text{PbBr}_4$ of 1:16 presented the highest current efficiency and luminance of 4.90 cd A$^{-1}$ and 2935 cd m$^{-2}$, respectively. Perovskite LEDs were with a device structure of ITO/Buf-HIL/perovskite/TPBI/LiF/Al, where Buf-HIL is poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate):perfluorinated ionomer and TPBI is 1,3,5-tris($N$-phenylbenzimidazol-2-yl)benzene.

The iodide-based analog $\text{PEA}_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}$ ($n = 5$) has been demonstrated by Sargent et al. to outperform 3D $\text{MAPbI}_3$ ($n = \infty$) for near-infrared emission, with an EQE of 8.8% and radiance of 82 W sr$^{-1}$ m$^{-2}$. Similarly, they attributed the superior performance to the funneling mechanism, which occurs within sub-ns timescales and outcompetes nonradiative exciton quenching and increases radiative recombination.

Liao et al. developed perovskite LEDs exhibiting a deep blue emission with commission international de L’Eclairage (CIE) coordinate at $(0.14, 0.05)$ by designing a new aromatic polyammonium bromide spacer [1, 4-bis (aminomethyl)benzene bromide (P-DABr)] with double functional amid terminal, which provided a strong binding force with PbBr$_6$, leading to stable low-dimensional domains. A mixed spacer system (PEABr + P-DABr) was further executed to modulate the domain distribution in perovskite film. It was stated that the nonradiative recombination and retarded exciton transfer were significantly suppressed by this method. The device presented a peak EQE of 2.6%, which was an

| Table 3: Recent advancement of solar cells based on 3D/2D perovskites with a bilayer structure |
|-----------------------------------------------|-------------------------------|-----------------|-----------------|
| Active layer                                   | Device structure               | PCE (%)         | Stability        | Year (ref)      |
| $\text{MAPbI}_3$/CAI                           | ITO/PEDOT:PSS/active layer/PCBM/ rhodamine 101/LiF/Ag | 13.86           | 54% of PCE after 220 h, dark, >50% RH | 2016$^{124}$ |
| $\text{Cs}_{0.1}\text{FA}_{0.74}\text{MA}_{0.13}\text{Pb}_2\text{Br}_{0.39}/\text{A34}$ | FTO/c-TiO$\text{2}/m$-TiO$\text{2}/active layer/spiro-OMeTAD/Au | 20.13           | N/A              | 2018$^{125}$ |
| ($\text{FAPbI}_3)_{0.88}$(CsPbBr$_3)_{0.12}$/5-AVA | FTO/c-TiO$\text{2}/m$-TiO$\text{2}/active layer/ CuSCN/Au | 16.75           | 98% of PCE after 63 days, dark, 10% RH | 2018$^{126}$ |
| $\text{Cs}_{0.1}\text{FA}_{0.74}\text{MA}_{0.13}\text{Pb}_2\text{Br}_{0.39}$/PEAI | FTO/c-TiO$\text{2}/m$-TiO$\text{2}/active layer/spiro-OMeTAD/Au | 20.1            | 90% of PCE after 800 h, 50°C, continuous light soaking | 2018$^{127}$ |
| $\text{Cs}_{0.05}(\text{MA}_{1.12}\text{FA}_{0.88}\text{Br}_{0.27})/(\text{PEA})_{0.83}\text{Pb}_{0.17}/(\text{PEAI})$ | FTO/TiO$\text{2}/active layer/spiro-OMeTAD/Au | 18.51           | 90% of PCE after 1000 h, dark, 60 RH | 2018$^{122}$ |
| $\text{Cs}_{0.05}(\text{MA}_{1.12}\text{FA}_{0.88}\text{Br}_{0.27})/4$-PEAI | FTO/TiO$\text{2}/active layer/spiro-OMeTAD/Au | 20.54           | 99% of PCE after 36 d, dark, 10–30% RH | 2019$^{128}$ |
| ($\text{FAPbI}_3)_{0.83}$(CsPbBr$_3)_{0.17}$/MDACL$_2$ | FTO/TiO$\text{2}/active layer/spiro-OMeTAD/Au | 23.77           | 90% of PCE, over 600 h, continuous light soaking | 2019$^{129}$ |
| PTAI-$\text{MAPbI}_3$ | ITO/SnO$\text{2}/active layer/spiro-OMeTAD/ MoO$_\text{3}$/Ag | 21.16           | 93% of PCE, over 500 h, continuous light soaking | 2020$^{130}$ |
| MAPbI$_3$/SBLC | ITO/PTAA/active layer/PCBM/BCP/Ag | 20.14           | N/A              | 2020$^{131}$ |
| $\text{Cs}_{0.05}\text{Rh}_{0.05}(\text{FA}_{0.83}\text{MA}_{0.17}\text{Br}_{0.39})/(\text{PEAI})_{0.83}\text{Pb}_{0.17}/3$/PTAA | ITO/SnO$\text{2}/active layer/spiro-OMeTAD/Ag | 21.38           | 70.4% of PCE, over 1000 h, N$_2$, 85°C | 2020$^{132}$ |
| $\text{Cs}_{0.08}\text{FA}_{0.77}\text{MA}_{0.12}\text{Pb}_2\text{Br}_{0.35}$/BnAI | ITO/m-TiO$\text{2}$/SnO$\text{2}/active layer/spiro-OMeTAD/Au | 20.79           | 91% of PCE, over 6 months, air, 85°C | 2020$^{133}$ |

Abbreviations: 2D, two-dimensional; 3D, three-dimensional; PCE, power conversion efficiency.
FIGURE 16  Crystallization of quasi-two-dimensional (2D) perovskite SCMs. (A) Schematic illustration of the alignment of butylammonium cation surfactant at the water–air interface for templating the nucleation. (B) Experimental setup during nucleation. (C) Molecular interaction between precursor molecule (red) and water molecules (yellow). Lower interaction energy is expected for the surface layer molecules due to the surface tension effect compared to those in bulk solution. (D) Experimental setup during crystal growth. Graphic illustrations on the (E) lower nucleation barrier, (F) larger free energy changes during crystal growth, and (G) higher growth rate of the precursor molecules at the water–air interface compared to those in bulk solution. Reproduced with permission from Reference 60. Copyright 2018. American Chemical Society

FIGURE 17  (A) Photograph of a well-shaped 36 × 6 × 6-mm³ bulk two-dimensional (2D) (PEA)₂PbI₄ perovskite solar cell (PSC). (B) X-ray diffraction (XRD) patterns of a 2D (PEA)₂PbI₄ PSC recorded from (001) and (010) planes. (C,D) Optimized crystal structures on (001) (C) and (010) (D) facets of (PEA)₂PbI₄. SEM image of (PEA)₂PbI₄ PSC for the (001) (E) and (010) (F). Reproduced with permission from Reference 137. Copyright 2019. Elsevier
invigorative result for deep blue perovskite LED on basis of pure bromide perovskite.

To enhance radiative emission processes in LEDs, Di et al. reported LEDs with a record EQE of 20.1%, by embedding the quasi-2D perovskites (NMA)$_2$(FA)Pb$_2$I$_7$ into a high bandgap polymer poly(2-hydroxyethyl methacrylate) (poly-HEMA) forming a bulk heterojunction (BHJ) composite, where NMA is 1-naphthylmethylammonium. The GIWAXS indicated that the 2D/3D perovskite crystallites were iso-tropically oriented in the perovskite-polymer (PP) BHJ thin film (Figure 20(A)). The LED was constructed from a solution-processed method with a structure of ITO/MZO/PEIE/PPBHJ/poly(9,9-dioctylfluorene-alt-N-(4-sec-butylphenyl)-diphenylamine) (TFB)-poly(9,9-di-η-octylfluorenyl2,7-diyl) (PFO)/MoO$_x$/Au, where MZO is magnesium-alloyed zinc oxide, PEIE is polyethyleneimine, TFB-PFO is TFB blended with PFO. As indicated in Figure 20(B), the peak of EQE reached 20.1%, which was a record for perovskite LEDs so far.

### Table 4

| Active layer                  | Morphology | $R$ (A/W) | $D^*$ (Jones) or dark current (A) | Year (ref) |
|-------------------------------|------------|-----------|----------------------------------|------------|
| (BA)$_2$PbBr$_4$              | Single crystal | $\sim$2100 A W$^{-1}$ at 470 nm | $10^{-10}$ A at 470 nm | 2016$^{138}$ |
| (BA)$_2$PbBr$_4$              | Nanobelt   | —         | $1.97 \times 10^{-8}$ A at 405 nm | 2017$^{135}$ |
| (BA)$_2$(MA)$_3$Pb$_4$I$_{13}$ | Nanowire   | $\sim1.5 \times 10^5$ A W$^{-1}$ at 10 Hz | $\sim4.7 \times 10^{15}$ Jones at 10 Hz | 2018$^{134}$ |
| (BA)$_4$Pb$_3$I$_{13}$        | Single crystal | —        | $\sim1.6 \times 10^{13}$ Jones at 0.4 V | 2018$^{60}$ |
| (PEA)$_2$PbBr$_4$             | Single crystal | 139.6 A W$^{-1}$ at $-5$ V | $1.89 \times 10^{15}$ Jones at $-5$ V | 2019$^{137}$ |
| Sn-doped (PEA)$_2$Pb$_4$      | Polycrystal | 6.43 A W$^{-1}$ at 10 V, 520 nm | $6.77 \times 10^{13}$ Jones at 10 V, 520 nm | 2020$^{139}$ |
| (ThMA)$_2$(MA)$_2$Pb$_4$I$_{10}$ | Nanowire | $1.1 \times 10^4$ A at 30 Hz | $9.1 \times 10^{15}$ Jones at 120 Hz | 2020$^{140}$ |
| (PEA)$_2$(MA)$_{n-1}$Pb$_n$I$_{3n-1}$ | Polycrystal | 149 A W$^{-1}$ at 598 nm | $2.0 \times 10^{12}$ Jones at 598 nm | 2020$^{141}$ |
| (PEA)$_2$(MA)$_4$Pb$_3$I$_{16}$ | Microcrystal | 182.63 A W$^{-1}$ at 15 V | $2.52 \times 10^{14}$ Jones at 15 V | 2020$^{142}$ |
Recent advancement in perovskite LEDs based on the low-dimensional perovskites is summarized in Table 5.

7 | PEROVSKITE LASERS BY THE LOW-DIMENSIONAL HALIDE PEROVSKITES

A laser refers to a device that can emit coherent light with strong intensity and perfect directionality. By taking into consideration of high absorption coefficient and low density of defects, low-dimensional halide perovskites are excellent gain materials for the development of high-performance lasing devices.

Li et al. demonstrated that FA-based 2D RR perovskite thin films of \((\text{NMA})_2(\text{FA})_n/\text{Co}_1\text{Pb}_n\text{X}_{3n+1}\) showed superior optical gain properties. Different from the stimulated emission mechanism of 3D perovskites with electron–hole plasma at room temperature, 2D RR perovskites exhibited the feature of strongly bound electron–
hole pairs (excitons) and naturally form an energy cascade. It was observed that amplified spontaneous emission (ASE) from these perovskites with a low threshold ($<20.0 \pm 2 \mu\text{J cm}^{-2}$), tunable wavelengths from visible to the near-infrared spectral range ($530$–$810$ nm), and good photostability with an operation duration exceeding $1.2 \times 10^8$ laser pulses.

After that, Zhang et al. demonstrated room-temperature ASE and lasing from mixed multiple QWs in 2D-RPPs of $(\text{BA})_2(\text{MA})_n - 1\text{Pb}_n\text{Br}_{3n+1}$. They performed femtosecond transient absorption characterization to reveal an ultrafast population transfer along the energy cascade from small-$n$-QW to large-$n$-QW, concentrating photo excitations at the lowest bandgap QWs ($n \approx \infty$), enabling the population inversion for stimulated emission. They found that the 2D-RPPs of $(\text{BA})_2(\text{MA})_5\text{Pb}_6\text{Br}_{19}$ ($n = 6$) show an ASE threshold of $13.6 \mu\text{J cm}^{-2}$ and a high gain coefficient ($G$) of $112 \text{ cm}^{-1}$. Their findings reveal that 2D RPPs can be potentially applied for electrically driven lasers in on-chip integration of photonics and electronic circuits.

Recent advancement of perovskite lasers based on the low-dimensional perovskites is summarized in Table 6.

### Table 5 Recent advancement of LEDs based on the low-dimensional perovskites

| Active layer | Wavelength (nm) | EQE (%) | CE (cd/A) | Luminance (cd/m²) | Year (ref) |
|--------------|----------------|---------|-----------|-------------------|------------|
| PEA$_2$PbBr$_4$ | 410 | 0.04 | — | — | 2016$^{149}$ |
| PEA$_2$(MA$_n$ $-$ $1\text{Pb}_n\text{Br}_{3n+1}$)(MA$_n$ $-$ $1\text{Pb}_n\text{I}_{3n+1}$) | 760 | 8.8 | — | — | 2016$^{145}$ |
| (NMA)$_2$(MA$_n$ $-$ $1\text{Pb}_n\text{Br}_{3n+1}$) | 763 | 11.7 | — | — | 2016$^{146}$ |
| PEA$_2$(MA$_n$ $-$ $1\text{Pb}_n\text{Br}_{3n+1}$) | 526 | 7.4 | 4.9 | 8400 | 2017$^{150}$ |
| PEA$_2$(FAPbBr$_4$)$_2$PbBr$_4$ | 532 | 14.36 | 62.43 | 9120 | 2018$^{151}$ |
| (BAB)(FA$_n$ $-$ $1\text{Pb}_n\text{I}_{3n+1}$) | 776 | 4.2 | — | — | 2019$^{152}$ |
| PEA$_2$Cs$_2$Pb$_3$Br$_{10}$ | 508 | — | — | — | 2020$^{153}$ |
| NPA$_1$Cs$_2$Pb$_3$Br$_{10}$ | 480 | 0.086 | 0.1 | 8 | 2020$^{153}$ |
| PEA$_2$NPA$_1$Cs$_2$Pb$_3$Br$_{12}$ | 485 | 2.62 | 15 | 1200 | 2020$^{153}$ |
| PEA$_2$PbBr$_4$FAPbBr$_3$ | 527 | 12.4 | 52.1 | 5200 | 2020$^{154}$ |
| NMABr-FAPbBr$_3$ | 520 | 3.4 | 16.3 | 500 | 2020$^{154}$ |

Abbreviations: EQE, external quantum efficiency; LEDs, light-emitting diodes.

### Summary and Outlook

The emergence of low-dimensional perovskites as semiconductors has revolutionized next-generation optoelectronics owing to their extraordinary optical and electronic properties. In this short review, we discussed the optoelectronic properties of the low-dimensional
halide perovskites. From this point of view, we highlighted the properties of 2D perovskite MQWs structure. The decrease in the value of $n$ could lead to a compression of exciton wavefunction in the QW due to dielectric confinement, which results in enhanced exciton binding energy within 2D perovskites. In addition, the observation of charge carrier density at the ES is close to that of metal, which implies that the low-dimensional perovskites offer great opportunities for designing novel optoelectronics. Subsequently, we reviewed synthetic methods for the preparation of the low-dimensional halide perovskites, including the one-pot synthetic method, direct growth method, polar solvents free method, and vapor-assisted method. Despite the great progress in the synthetic methods for the preparation of low-dimensional halide perovskites, more work needs to be done in the future toward the understanding of the crystal nucleation and growth mechanism, aiming to achieve ultra-uniform low-dimensional perovskite thin films.

The low-dimensional perovskites with nontrivial optical and electronic properties display tremendous potential applications in solar cells, FDS, LEDs, and lasers. To further explore the unique characteristics of the low-dimensional perovskites for all these applications, more research is demanded to focus on materials design and the fundamental physical and chemical properties of the low-dimensional perovskites.

In terms of PSCs by the low-dimensional perovskites, even though enormous efforts have been devoted to approaching the vertical orientation of perovskite thin films, aiming to accelerate the charge carrier transport in the vertical direction, the efficiencies were still hard to compete with those by 3D perovskites. To overcome it, we think that we should establish the judicious selection criteria for OS’s design principles for 2D perovskites, for example, the effect of different spacer lengths, functional units, and the substituent groups. It is necessary to develop low-dimensional perovskite structures with pure phases, like adopting mechanical exfoliation process, to avoid the drawbacks of the anisotropic properties. The synthetic techniques and characterization are required to study the optical and electronic properties of 2D perovskites, and a deep understanding of structure–property relationships of different 2D perovskite phases needs to be explored. To substitute the insulating OSs with conjugated OSs would be a fascinating direction for boosting PCEs of PSCs. In addition, forming low bandgap organic donor (or acceptor)/2D perovskite bulk heterostructure would not only contribute to additional absorption of the 2D perovskite but also minimize the energy required for dissociating exciton into free charge carriers.

For PPDs by the low-dimensional perovskites, great efforts are still demanded to achieve high device performance. To minimize the dark current in PPDs by the low-dimensional perovskites, surface passivation between the low-dimensional perovskites and charge transport layers, utilization of Lewis acid and/or Lewis base to passivate $\text{PbX}_3^-$ and/or $\text{Pb}_2^{2+}$ anti-site defects, and synthesis of the low-dimensional perovskite single crystals (no grain boundary effect) are crucial importance to reduce trap states, which is also essential to achieve high response speed.

Although low-dimensional perovskites are being considered for making next-generation LEDs and lasers in the future, there are some significant challenges. For example, the stronger exciton–lattice coupling and randomly oriented polycrystalline in the low-dimensional perovskite thin films are responsible for fast nonradiative exciton quenching. Toward the ends, large-scale size single-crystalline perovskite films with higher carrier mobility, lower defect density, are expected to replace the conventional polycrystalline perovskite films. It has been reported that the additives can not only effectively passivate perovskite surface traps, but also tune the film morphology, which thus ensures reduced nonradiative recombination.$^{161}$ Another effort should be focused on increasing PLQY by metal ions doping and defect passivation with different ligands. Through these novel designs, we would expect to develop devices with high performance by the low-dimensional perovskites in the future.

**Table 6** Recent advancement of lasers based on the low-dimensional perovskites

| Active layer | Morphology | Crystallinity | Pump source | Lifetime (laser shots) | Year (ref) |
|--------------|------------|---------------|-------------|------------------------|-----------|
| (BA)$_2$(MA)$_3$Pb$_3$Br$_{19}$ | Microring | Polycrystal | 400 nm, 150 fs, 1 kHz | $>3.6 \times 10^4$ | 2018$^{156}$ |
| (BA)$_2$(FA)$_2$Pb$_3$Br$_{10}$ | Nanowire | Polycrystal | 400 nm, 150 fs, 1 kHz | $>2.16 \times 10^7$ | 2018$^{157}$ |
| (OA)$_2$(MA)$_n$ – $\text{Pb}_n\text{Br}_{3n+1}$ | Microplatelet | Polycrystal | 400 nm, 150 fs, 1 kHz | $>4 \times 10^7$ | 2018$^{158}$ |
| (NMA)$_2$(FA)Pb$_2$Br$_6$ | Film | Polycrystal | 400 nm, 150 fs, 1 kHz | $>1.2 \times 10^8$ | 2018$^{155}$ |
| PEABr-FAPbBr$_3$ | Film | Polycrystal | 488 nm | Unchanged after 1 h, air | 2020$^{159}$ |
| NMABr-FAPbBr$_3$ | Film | Polycrystal | 488 nm | Unchanged after 1 h, air | 2020$^{159}$ |
| (PEA)$_2$(FA)$_3$Pb$_2$Br$_{13}$ | Film | Polycrystal | 400 nm, 150 fs, 1 kHz | — | 2020$^{160}$ |
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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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