Low-dimensional metal halide perovskites and related optoelectronic applications

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Abstract
Low-dimensional materials have pivotal significance in modern photonic, electronic, and optoelectronic areas due to their unique properties of the scale effect. Metal halide perovskites have revived in the optoelectronic fields recently, drawing intensive attention in photovoltaic devices, light-emitting diodes, lasers, photodetectors, and so on. Compared to their three-dimensional counterparts, the role of low-dimensional perovskites is becoming crucial, requiring a comprehensive understanding and exploration unceasingly. In this review, we examine low-dimensional perovskite of different forms and clarify various synthesis methods with morphological and dimensional control. Additionally, we also summarize potential optoelectronic applications based on their advantageous optical/electrical properties and enhanced mechanical integrity and stability. Finally, we propose a future perspective and possible developing directions in the exploration of novel perovskite-derived materials, new physics, and promising applications.

KEYWORDS
applications, low-dimension, metal halide perovskites, synthesis

1 | INTRODUCTION

Perovskite, which refers to specific compounds that possess a formula of $ABX_3$ (A and B sites are cations, and X site is anion) and the identical structure as $\text{CaTiO}_3$, has been investigated for more than a century. In recent years, organic-inorganic metal halide perovskites, as one of the various types, have attracted much attention due to their unique properties and remarkable device performances. In 2009, Kojima et al. utilized lead halide perovskite as sensitizers in dye-sensitized solar cells, achieving a solar-to-electricity power conversion efficiency (PCE) of 3.8%. This research marks the new renaissance of metal halide perovskites first in PV devices. From that time on, the development of perovskite solar cells has dramatically ascended, achieving a certified PCE up to 25.2% (Best Research-Cell Efficiencies (NREL, 2019) https://www.nrel.gov/pv/assets/pdfs/best-research-cell-efficiencies.20190802.pdf). Due to the unique optoelectronic merits like high absorption coefficient, good defect-tolerant properties, ease of fabrication, and so on, metal halide perovskites have extensive applications not only in PV field but also in other optoelectronic devices such as light-emitting diodes (LEDs), photodetectors, nanolasers, and so on.

Bulk three-dimensional (3D) metal halide perovskites have a general structural formula of $ABX_3$, where A site
is a cation, typically the organic methylammonium (MA$^+$), formamidinium (FA$^+$) or inorganic cesium (Cs$^+$). 22-24 B site is metal, typically lead (Pb$^{2+}$), or tin cations (Sn$^{2+}$), and X site is the halide like chloride (Cl), bromide (Br), and iodide (I). 25-27 The term “3D” here focuses on the materials themselves and emphasizes the connected ways where $[\text{BX}_6]$$_4^-$ octahedra consisting of one B cation, and six X anions are combined through corner sharing to form a 3D network. The A cation occupies the site in the middle of eight octahedra, and each element here needs to owe the proper valence state to keep a whole charge balance. 28-30

To predict the 3D structure forming possibility, Goldschmidt tolerance factor ($t$) and octahedral factor ($\mu$) are used as empirical rules. The definitions of $t$ and $\mu$ are as following: 

$$t = \frac{R_A + R_X}{\sqrt{2(R_B + R_X)}}$$

$$\mu = \frac{R_B}{R_X}$$

where $R_A$, $R_B$, and $R_X$ are the effective radii of the A, B, and X ions, respectively. 31,32 For a well-established 3D network, $t$ and $\mu$ are usually in the range of 0.8-1.0 and 0.44-0.9, respectively. 10 Non-perovskite structures are often observed for improper-sized ions, for example, large A cations like CH$_3$CH$_2$NH$_3^+$ ($t$ > 1.0) and small A cations like Rubidium ($t$ < 0.8) will both lead to the collapse of the 3D structured network. 33-37 As observed empirically, distinguished advantages suitable for optoelectronics often vanish when switching to the non-perovskite structures. 38-40

“Low-dimensional” perovskites refer to the perovskite compounds with at least one reduced dimension, in contrast to that with three unlimited extensions. 41-45 The term “low-dimensional” often leads to confusion due to its diverse meanings in different situations. In this review, we classify low-dimensional perovskites from two aspects: “material-level” and “structure-level” low dimensionalities.

The “material-level” low dimensionality emphasizes the perovskite material itself, underlining the building blocks of the perovskites that the individual components should have at least one dimension down to the molecular level. Specifically, these consisting parts made of $[\text{BX}_6]$$_4^-$ octahedra usually exist in several forms, such as two-dimensional (layers), one-dimensional (wires), or separately zero-dimensional (polyhedrons) styles (Figure 1A). They are isolated by organic molecules and possess the dimension to one or several molecular units in one direction, at least. No matter they are assembled into nanostructures or even bulk films, the inherent properties remain nearly unchanged relative to the basic blocks. 44-50

Differently, “structure-level” low-dimensional perovskites emphasize the final morphologies and commonly refer to those nanostructures of nanoplatelets/nanosheets, nanowires/nanorods, and nanocrystals (Figure 1B). 51-53 Mostly, these “structure-level” low-dimensional perovskites are made up of the 3D networks of corner-sharing
[BX₆]⁴⁻ octahedra, the same as in bulk ABX₃ perovskites. In some specific cases, these nanostructures also consist of “material-level” low-dimensional individual species (layers, wires, or polyhedrons) as well.⁵⁴ From the above discussion, “structure-level” and “material-level” low dimensionalities are used to elaborate on different aspects and are not on opposite sides due to the partial overlap. In the following section, when referring to “structure-level” low-dimensional perovskites, we also use expressions such as nanoplatelets, nanowires, and nanocrystals to distinguish from “material-level” low-dimensional perovskites.

Compared to 3D counterparts commonly used, low-dimensional perovskites possess additional properties, mainly originating from their shape diversity and quantum confinement effects.⁵⁵-⁵⁸ For example, due to the widely adjustable dimensions and sizes, low-dimensional materials usually own feasibly tunable bandgaps, enriching the wavelength range of absorption and emission.⁶⁹-⁷² Owing to the enlarged Coulombic force between electrons and holes, these electron-hole pairs have much higher binding energy and consequent high photoluminescent efficiency, which are desired for luminescent applications.⁶²-⁶⁵ Also, strong anisotropic nature, together with enhanced surface-volume-ratio, could provide better contacts and smooth charge transport channel.⁶⁶,⁶⁷ Finally, better crystallinity with reduced trap density and enlarged mechanical stability are often observed in low-dimensional material, greatly improving the optoelectronic device longevity and beneficial for flexible electronics.⁶⁸-⁷¹

In this review, we will focus on the “structure-level” low-dimensional perovskites regardless of the consisting individuals (bulk, layers, wires, or polyhedrons). Meanwhile, some perovskite bulk forms consisting of “material-level” low-dimensional components are also selectively discussed in the following section.

2 | 2D METAL HALIDE PEROVSKITES

As the thickness reduces to several unit cells, the quantum confinement effect typically leads to larger binding energy, higher photoluminescence (PL), and increased bandgaps.⁷¹ Perovskite nanosheets or nanoplatelets can be thought by minimizing the thickness of bulk 3D ABX₃ perovskite to a large extent. When they are extremely thin, the formula is no longer the ABX₃ but with a finite value of n: Aₙ₋₁BₓX₃n₊₁.⁴⁶ These thin nanosheets typically consist of similar networks as 3D counterparts, with two A cations terminated on the top and bottom sides.

In addition to these nanosheets, there is another “material-level” low type of two-dimensional (2D) perovskite, Ruddlesden-Popper (RP) phase perovskites, which have a chemical formula of L₂Aₙ₋₁BₓX₃n₊₁ (L is the long-chain cation and A is the smaller cation that can be fitted into the 3D lattice, like MA⁺ or FA⁺). The L normally has ammonium cations that can react with the terminated [BX₆]⁴⁻ corner-shared layer and fit the periphery of these octahedra. These RP-type perovskites can be assembled into a bulk crystal or other nanostructures through the van der Waals interaction of each layer, keeping quantum confinement effects unchanged.⁴⁴,⁵⁴,⁷²-⁷⁴ In this section, we concentrate on the synthesis methods and corresponding applications of nanoplatelets based on 3D networks and partially discuss the 2D RP-phase assembled bulk perovskites when referring to some optoelectronic applications.

2.1 | Synthesis methods

2.1.1 | All vapor-phase synthesis method

The bottom-up process is found to be feasible to achieve the desired 2D nanostructures. Considering the van der Waals layered nature of lead halides, it appears an effective way to acquire perovskite nanosheets with a start from PbX₂ nanoflakes (X = Cl/Br/I).⁷⁵-⁷⁷ In 2014, Ha et al. developed a two-step method, combining chemical vapor deposition (CVD) and gas-solid reaction, to grow perovskite nanoplatelets (Figure 2A).⁷⁵ The authors first grew PbI₂ nanoplatelets on the muscovite mica substrates by van der Waals epitaxy in a vapor transport chemical deposition system. Then, they converted the as-formed PbI₂ nanoplatelets into perovskites by a gas-solid reaction with methylammonium iodide (MAI) molecular gas, slightly increasing the thickness after the incorporation of MAI molecular (Figure 2B).⁷⁵ The prepared perovskite nanoplatelets exhibit a lateral dimension of 5-30 μm and a thickness from dozens of to several hundreds of nanometers. By replacing the muscovite mica by SiO₂/Si substrates and altering the synthesis temperature and time, Niu et al. could precisely tune the thickness of perovskite nanoplatelets from few layers to several nanometers.⁷⁶ Besides, when employing rough-surfaced substrates, free-standing and large-scale PbI₂ nanosheets were synthesized by Lan et al. through the manipulation of microenvironment (Figure 2C).⁷⁷ Complete conversion to free-standing perovskite nanoflakes by CVD provides the promising potential for photon-detected applications.
2.1.2 Combined solution- and vapor-phase synthesis method

Thin PbI₂ nanoplatelets with the thickness down to monolayer can also be obtained by a facile solution process (Figure 2D). Saturated PbI₂ aqueous solution was cast on a substrate, and 2D PbI₂ nanosheets would nucleate at an elevated temperature. Similar to the vapor-conversion process above, 2D MAPbI₃ perovskite nanosheets were formed by intercalating the MAI molecules into PbI₂ nanosheets under a gas atmosphere. Later, an impressive work by Wang et al. demonstrated well-patterned perovskite microplate arrays on a periodic substrate (Figure 2E). Specifically, the substrates were partially treated with periodic hydrophilic patterns, where the PbI₂ seeds emerged after the seeding solution flew over.
Afterward, a common vapor reaction process by MAI gas converted PbI₂ microplates into the desired perovskite plates. The successful realization of the periodic perovskite patterns proves the potential applications in integrated electronics.

2.1.3 Exfoliation method

Different from the inherent ionic bond of nanoplatelets, the weak van der Waals interaction between neighboring layers makes it possible for RP type perovskite to be exfoliated into ultrathin nanosheets.⁸⁰-⁸² Niu et al. produced one to a few layers of thin flakes from (C₆H₉C₂H₄NH₃)₂PbI₄ crystals (Figure 3A-C).⁸⁰ Hexagonal RP type (C₆H₉C₂H₄NH₃)₂PbI₄ perovskite microcrystals with 30 μm lateral size were first synthesized, followed by a micromechanical exfoliation technique to create ultrathin flakes. This method is rather suitable for the investigation of fundamental properties but inconvenient for high production due to the uncontrollable size and thickness.

2.1.4 Direct solution growth method

In 2015, Dou et al. proposed a direct solution growth method to synthesize atomically thin RP type BA₂PbI₄ (BA = C₄H₉NH₃⁺) perovskite nanosheets. A much diluted precursor in ternary organic solvents was cast on flat substrates, where acetonitrile (ACN) and chlorobenzene were used to reduce the solubility of BA₂PbI₄ in dimethylformamide (DMF) and promote the crystallization (Figure 3D).⁸³ Thus, a single-unit layer, with a thickness of ~1.6 nm, was successfully demonstrated. Based on this work, Chen et al. further studied the critical factors systematically, including crystallization temperature, solvent ratio, and polarity on the growth dynamics (Figure 3E).⁸⁴ They assumed that the formation of large-size thin sheets was governed by the effects of diffusion-mechanism-dominated branched growth, together with CAN-induced c-axis suppression. Under the optimized condition, 2D BA₂PbBr₄ perovskites with the lateral sizes up to 40 μm and thicknesses down to a few nanometers were obtained. These works above suggest that the solution growth method can be suitable to achieve high-quality perovskite nanosheets with controllable thickness.

2.1.5 Colloidal growth method

Due to the great success in the synthesis of inorganic quantum dots like CdSe and ZnSe,⁸⁵-⁸⁸ the colloidal growth method has vital importance in the perovskite nanostructure fabrication.⁶⁰,⁸⁹ In 2015, Tyagi et al. adapted a precipitation method used for perovskite nanoplatelet...
synthesis, where octylammonium bromide (OABr) was chosen as the long-chain ligands. Later, Sichert et al. synthesized MAPbBr₃ nanoplatelets by a modified process where they added a mixed solution of PbBr₂, OABr, and MABr in DMF into toluene dropwise (Figure 4A). The thicknesses of these nanoplatelets were well controlled from one to several units by varying the adding amount of OABr. Bekenstein et al. reported the colloidal synthesis of highly luminescent (photoluminescent quantum yield [PLQY] 84%) inorganic CsPbX₃ nanoplatelets with the thickness of 1-5 unit cells, showing adjustable PL emissions due to strong quantum size effect (Figure 4B). A facile anion exchange process further promoted the PL emissions covering the entire visible spectrum. The systematic investigations facilitated the following development of the CsPbX₃ nanoplatelet synthesis, with a more precise thickness control (Figure 4C) and an enlarged lateral size up to micrometer range (Figure 4D-E).

2.2 Applications of 2D metal halide perovskites

Solar cells employing 3D perovskites have achieved considerable improvements in the past few years, yielding a certified PCE up to 25.2%. Along with the rapid PCE development, the instability problem has been proposed...
as a considerable issue. Compared to the traditional 3D ABX₃ perovskite, 2D RP layered perovskite has a much more enhanced moisture tolerance and can improve the stability to a certain extent. In addition to this benefit, the confinement in 2D perovskite will bring up large binding energy and improve the photoluminescent efficiency, which is promising for the application of emissions like LEDs and lasers. In the following section, we will discuss the optoelectronic applications based on 2D perovskites, including not only nanosheets or nanoplatelets but also some unique forms of RP type perovskites.
# Table 1
Summary of key parameters of solar cells based on low-dimensional perovskites

| Materials                               | Structures | Device configuration                      | $V_{oc}$ (V) | $J_{sc}$ (mA cm$^{-2}$) | FF (%)  | PCE (%)  | References |
|-----------------------------------------|------------|-------------------------------------------|--------------|--------------------------|--------|---------|------------|
| PEA$_2$MA$_2$Pb$_3$I$_{10}$             | Thin film  | FTO/c-TiO$_2$/PVSK/spiro-OMeTAD/Au        | 1.18         | 6.72                     | 60.0   | 4.73    | 93         |
| PEA$_2$MA$_2$Pb$_3$I$_{16}$             | Thin film  | ITO/PEDOT:PSS/PVSK/PCBM/BCP/Ag            | 1.19         | 15.8                     | 75.0   | 14.1    | 100        |
| PEA$_2$MA$_x$Pb$_{3x}$I$_{16}$          | Thin film  | FTO/c-TiO$_2$/PVSK/spiro-OMeTAD/Au        | 1.12         | 16.42                    | 67.0   | 12.29   | 101        |
| (iso-BA)$_2$MA$_x$Pb$_{3x}$I$_{13}$     | Thin film  | FTO/C$_{60}$/PVSK/spiro-OMeTAD/Au         | 1.20         | 16.54                    | 53.54  | 10.63   | 102        |
| (3BBA)$_x$MA$_{1.5}$Pb$_{3n+1}$ (3 < n < 4) | Thin film  | ITO/PTAA/PVSK/PCBM/Cr/Au                 | 1.23         | 18.22                    | 81.2   | 18.2    | 103        |
| BA$_2$MA$_2$Pb$_{10}$                   | Thin film  | FTO/c-TiO$_2$/m-TiO$_2$/PVSK/spiro-OMeTAD/Au | 0.929       | 9.42                     | 46.0   | 4.02    | 94         |
| BA$_2$MA$_3$Pb$_{13}$                   | Thin film  | FTO/PEDOT:PSS/PVSK/PCBM/Ag                | 1.01         | 16.76                    | 74.13  | 12.51   | 98         |
| BA$_2$MA$_3$Pb$_{13}$                   | Thin film  | ITO/PEDOT:PSS/PVSK/PCBM/BCP/Ag            | 0.97         | 12.79                    | 55.0   | 6.82    | 104        |
| BA$_2$MA$_3$Pb$_{13}$                   | Thin film  | ITO/PEDOT:PSS/PVSK/PCBM/BCP/Ag            | 0.98         | 14.71                    | 61.0   | 8.79    | 104        |
| BA$_2$MA$_3$Pb$_{13}$                   | Thin film  | ITO/PEDOT:PSS/PVSK/PCBM/PEIE/Ag           | 1.14         | 18.8                      | 69.5   | 14.9    | 105        |
| BA$_2$MA$_3$Pb$_{13}$                   | Thin film  | ITO/PTAA/PVSK/C$_{60}$/BCP/Ag             | 1.24         | 19.86                    | 70.44  | 17.26   | 106        |
| BA$_2$MA$_4$Pb$_{16}$                   | Thin film  | ITO/PEDOT:PSS/PVSK/PCBM/Al                | 0.986        | 15.5                      | 65.5   | 10.0    | 107        |
| BA$_2$(MA$_{0.5}$FA$_{0.5}$)$_{3}$Pb$_{13}$ | Thin film  | ITO/PEDOT:PSS/PVSK/PCBM/BCP/Ag            | 0.999        | 18.12                    | 70.79  | 12.81   | 108        |
| BA$_2$(Cs$_{0.05}$MA$_{0.95}$)$_{3}$Pb$_{13}$ | Thin film  | FTO/c-TiO$_2$/PVSK/spiro-OMeTAD/Au       | 1.08         | 19.95                    | 63.47  | 13.68   | 109        |
| [(BA)$_{0.94}$(F$_3$EA)$_{0.06}$]$_2$MA$_3$Pb$_{13}$ | Thin film  | ITO/PEDOT:PSS/PVSK/PCBM/BCP/Ag            | 1.02         | 16.08                    | 76.3   | 12.51   | 110        |
| FA$_x$PEA$_{1-x}$Pb$_3$ (FAI/PEAL = 40) | Thin film  | ITO/NiO$_3$/PVSK/PCBM/bis-C$_{60}$/Ag    | 1.04         | 22.08                    | 77.13  | 17.71   | 111        |
| PEA$_2$MA$_{0.1}$Pb$_{3n+1}$ (n = 60)   | Thin film  | FTO/c-TiO$_2$/PVSK/spiro-OMeTAD/Au        | 1.09         | 19.12                    | 73.70  | 15.36   | 99         |
| PEA$_2$MA$_{0.1}$Pb$_{3n+1}$ (PEAI/PhI = 0.05) | Thin film  | FTO/c-TiO$_2$/m-TiO$_2$/PVSK/spiro-OMeTAD/Ag | 1.08    | 21.91                    | 80.36  | 19.10   | 112        |
| BA$_{0.05}$(FA$_{0.85}$Cs$_{0.17}$)$_{0.91}$Pb (I$_6$Br$_{0.4}$) | Thin film  | FTO/SnO$_2$/PCBM/PVSK/spiro-OMeTAD/Au     | 1.18         | 19.8                      | 73.0   | 17.12   | 113        |
| BA$_{0.05}$(FA$_{0.85}$Cs$_{0.17}$)$_{0.91}$Pb (I$_6$Br$_{0.2}$) | Thin film  | FTO/SnO$_2$/PCBM/PVSK/spiro-OMeTAD/Au     | 1.14         | 22.7                      | 80.0   | 20.6    | 113        |
| (EDBEPbI)$_{0.03}$ | Thin film  | FTO/SnO$_2$/PVSK/spiro-OMeTAD/Au          | 1.13         | 23.53                    | 79.2   | 21.06   | 114        |
| ThMA$_2$MA$_{0.1}$Pb$_{3n+1}$           | Thin film  | ITO/SnO$_2$/PVSK/spiro-OMeTAD/MoO$_3$/Ag  | 1.16         | 22.88                    | 81.0   | 21.49   | 115        |
| MAPbI$_3$                               | Nanowires  | FTO/c-TiO$_2$/m-TiO$_2$/PVSK/spiro-OMeTAD/Au | 1.052       | 19.12                    | 72.1   | 14.71   | 116        |
| MAPbI$_3$                               | Nanowires  | ITO/PEDOT:PSS/PVSK/spiro-OMeTAD/MoO$_3$/Ag | 1.01         | 23.39                    | 79.74  | 18.83   | 117        |

(Continues)
2.2.1 Solar cells

The first generation of solar cells utilizing an RP type perovskite PEA$_2$MA$_3$Pb$_3$I$_{10}$ (PEA = C$_6$H$_5$[CH$_2$]$_2$NH$_3$$^+$) as the absorber was fabricated by Smith et al. (Figure 5A) exhibiting a PCE of 4.73%.\textsuperscript{93} Although this efficiency falls behind that based on 3D perovskites, the moisture tolerance, which is essential for large-scale manufacture and broad commercialization, shows much superior performance than its bulk counterpart.\textsuperscript{93} Later, Cao et al. reported another RP type perovskite-based solar cell by using a relatively shorter organic cations BA (Figure 5B). Due to the self-assembly nature, the 2D compounds with n > 2 values (BA$_2$MA$_{n-1}$Pb$_n$I$_{3n+1}$) were formed with preferential growth orientation, beneficial for charge transport and formation of smooth thin films.\textsuperscript{94} Interestingly, when adopting a planar structure, the photovoltaic (PV) parameters were much poorer compared to the mesoporous structure, attributed to the short diffusion length of photocarriers in the 2D RP type perovskite.\textsuperscript{94} Tsai et al. proposed a similar hypothesis that insufficient charge transport could lead to worse performance arose from the nonideal layer orientation, where organic cations acted like insulating spacing layers between the conducting inorganic slabs. By applying a proposed hot-casting method here (Figure 5C), they produced BA$_2$MA$_{n-1}$Pb$_n$I$_{3n+1}$ thin films of near-single-crystalline quality, where the crystallographic planes of the inorganic perovskite had a preferential orientation perpendicular to both contacts.\textsuperscript{98} In a planar structure employing BA$_2$MA$_3$Pb$_4$I$_{13}$ as absorber, they obtained a PCE of 12.52%, among the highest efficiency of low-dimensional perovskite solar cells at that time (Figure 5D). Unencapsulated and encapsulated devices both exhibited much-enhanced stability when exposed to humidity and constant illumination. As the layer number n can effectively affect the device performance and stability, a systematical exploration to understand this

| Materials | Structures | Device configuration | $V_{oc}$ (V) | $J_{sc}$ (mA cm$^{-2}$) | FF (%) | PCE (%) | References |
|-----------|------------|----------------------|--------------|------------------------|--------|---------|-------------|
| MAPbI$_3$ | Nanowires  | FTO/c-TiO$_2$/PVSK/spiro-OMeTAD/Au | 1.04 | 23.8 | 74.6 | 18.42 | 118 |
| MAPbI$_3$ | Nanowires  | FTO/c-TiO$_2$/PVSK/spiro-OMeTAD/Au | 1.12 | 22.47 | 70 | 17.62 | 119 |
| CsPbBr$_3$ | Nanowires | FTO/c-TiO$_2$/PVSK/spiro-OMeTAD/Au | 0.851 | 2.96 | 44.5 | 1.21 | 120 |
| CsSnI$_3$ | Nanocrystals | ITO/c-TiO$_2$/PVSK/spiro-OMeTAD/Au | 0.86 | 23.2 | 0.65 | 12.96 | 121 |
| MAPbBr$_3$ | Nanocrystals | FTO/c-TiO$_2$/m-TiO$_2$/PVSK/PTAA/Au | 1.110 | 14.07 | 73.0 | 11.40 | 122 |
| FAPbI$_3$ | Nanocrystals | ITO/SnO$_2$/PVSK/spiro-OMeTAD/Au | 1.10 | 11.83 | 64.42 | 8.38 | 123 |
| FAPbI$_3$ | Nanocrystals | ITO/SnO$_2$/PVSK/spiro-OMeTAD/Au | 1.10 | 15.4 | 74.8 | 12.7 | 124 |
| CsPbI$_3$ | Nanocrystals | FTO/c-TiO$_2$/PVSK/spiro-OMeTAD/MoO$_3$/Al | 1.23 | 13.47 | 65.0 | 10.77 | 125 |
| CsPbI$_3$ | Nanocrystals | FTO/c-TiO$_2$/PVSK/spiro-OMeTAD/MoO$_3$/Al | 1.20 | 14.37 | 78.0 | 13.4 | 126 |
| CsPbI$_3$ | Nanocrystals | FTO/c-TiO$_2$/µ-GR/PVSK/PTAA/Au | 1.18 | 13.59 | 72.6 | 11.40 | 127 |
| CsPbI$_3$ | Nanocrystals | FTO/c-TiO$_2$/PVSK/PTB7/MoO$_3$/Ag | 1.27 | 12.39 | 80.0 | 12.55 | 128 |
| Cs$_{0.25}$FA$_{0.75}$PbI$_3$: CsPbI$_3$ | Nanocrystals | ITO/c-TiO$_2$/PVSK/spiro-OMeTAD/MoO$_3$/Ag | 1.20 | 18.91 | 76.0 | 17.39 | 129 |
| CsPbI$_3$: FAPbI$_3$ | Nanocrystals | FTO/c-TiO$_2$/PVSK/PTAA/MoO$_3$/Ag | 1.22 | 17.26 | 74.0 | 15.6 | 130 |

Note: $J_{sc}$, short-circuit current; $V_{oc}$, open-circuit voltage; FF, fill factor; PCE, power conversion efficiency.
correlation and inherent mechanism is in a request. Quan et al. fabricated quasi-2D perovskite films that exhibited enhanced stability without sacrificing the high performance of 3D MAPbI₃ perovskites (Figure 5F). Confirmed by density functional theory, the instability issue of 3D perovskites originates from the low formation energy and becomes aggravated when taking high humidity into account (Figure 5E). The addition of larger organic cation to form quasi-2D perovskite could enlarge the formation energy owing to the appreciable van der Waals forces that conferred enhanced stability. This work provides one possible elucidation of why 2D or quasi-2D perovskites own improved stability, especially when exposed to high moisture. For clear comparison, we summarize the key parameters of solar cells based on 2D perovskites in Table 1.

2.2.2 Light-emitting diodes

Enlarged binding energy and strong PLQY from quantum confinement make 2D perovskites suitable for the applications of LEDs. The first 2D RP perovskite-based LEDs can date back to the mid-1990s, where Era et al. demonstrated the device by incorporating a layered perovskite compound PEA₂PbI₄, showing an emission peak at 520 at liquid-nitrogen temperature. However, when driven at room temperature, the electroluminescence (EL) efficiency became rather low, which could be explained by the thermal quenching of photoluminescent in the perovskite film. Thus, fabricating efficient LEDs with certain EL efficiency at room temperature is a demanding trend. Liang et al. improved the crystal quality by converting poly-crystalline PEA₂PbBr₄ thin films into single-crystalline micro-sized nanoplates via solvent vapor annealing and showed efficient room-temperature violet EL at 410 nm (Figure 6A). Compared to the pure phase RP perovskite components (one value of n only), poly-crystalline RP perovskite films containing multiple phases (multiple values of n) have superior light-emission performance. Yuan et al. and Huang et al. independently reported LEDs incorporated with mixed layered perovskite materials, displaying impressive performance and stability. The mixed compounds act like a photocarrier concentrator, funneling the charge carriers from high-bandgap materials to the lowest-bandgap ones and boosting the external quantum efficiency (EQE) to 8.8% and 11.7% (Figure 6B-C), respectively. Jiang et al. tuned the “A-site” cation by using a Rubidium-Cesium alloyed perovskite, demonstrating a spectra-stable blue LED with a peak EQE of 1.35% (Figure 6D). In addition to the composition engineering, other strategies like surface passivation, crystallite tailoring, structure engineering, and so forth were also applied for performance and longevity improvement. Yang et al. utilized triocylphosphine oxide treatment to passivate the surface of quasi-2D PEA₂(FAPbBr₃)ₓPbBr₄ films, achieving an EQE of 14.36%. Xiao et al. added large-group ammonium as a surfactant to constrain the growth of 3D grains, producing smooth films consisting of crystallites with a size of about 10 nm. Through the incorporation of small ions such as sodium (Na⁺) to substitute the large organic molecules, Wu et al. overcame the charge transport issue resulting from the insulating long-chain cation and achieved a high EQE of 15.9% (Figure 6E,F). To further boost the EL efficiency of LEDs, Zhao et al. demonstrated a perovskite-polymer bulk heterostructure (PPBH) to suppress non-radiative recombination channels and obtained an EQE up to 20.1% (Figure 6G). Bipolar host materials like PVK: PBD were also incorporated for the protection, enabling glove-box free fabrication of LEDs, with efficient green photoluminescence and long-term stability. Detailed key parameters of 2D perovskite-based LEDs are summarized in Table 2.

2.2.3 Photodetectors

Owing to the large absorption coefficient, high carrier mobility, and good compatibility with flexible substrates, perovskite nanosheets are also appropriate for photodetectors. In 2015, Liu et al. demonstrated a thin MAPbI₃ nanosheet-based photodetector through a combined solution process and vapor-phase conversion method (Figure 7A). Under the incidence at 405 and 532 nm, the current density was significantly increased, showing photo responsivities of 22 and 12 AW⁻¹ (at 1 V) and fast response time (Figure 7B,C). Wang et al. grew 2D perovskite microcrystals on patterned electrodes (Figure 7D), creating independently addressable photodetector arrays and functional field-effect transistors (Figure 7E,F). In addition, photodetector based on inorganic perovskite nanosheets (like CsPbX₃, X = I/Br/Cl) has analogous performances compared to their organic counterparts. Due to the ultra-smooth surface and high optical response, a flexible detector based on monolayer and few-layer CsPbBr₃ nanosheets showed superior electrical stability even after 10 000 times of bending (Figure 7G,H). The following optimization to increase carrier extraction and transport by a combination of CNTs and CsPbBr₃ nanosheets (Figure 7I) enabled a flexible photodetector with an EQE and responsivity as high as 7488% and 31.1 AW⁻¹ (Figure 7J). A summary of the key parameters of photodetectors based on 2D perovskites is exhibited in Table 3.
FIGURE 6  A, Normalized photoluminescence (PL) and electroluminescence (EL) (at 6 V) spectra of an light-emitting diode (LED) device based on the (PEA)$_2$PbBr$_4$ nanoplatelets. Reproduced with permission. Copyright 2016 American Chemical Society. B, External quantum efficiency (EQE) values of two-dimensional (2D) Ruddlesden-Popper (RP) perovskite-based LEDs vs current density. Reproduced with permission. Copyright 2016 Nature Publishing Group. C, EQE and energy conversion efficiency vs current density for the NFPI$_7$ and NFPI$_6$B-based LED. Reproduced with permission. Copyright 2016 Nature Publishing Group. D, EQE values of 2D RP perovskite-based LEDs vs current density. Reproduced with permission. Copyright 2019 Nature Publishing Group. E, View of the unit cell of Na$_2$Cs$_{n-1}$Pb$_n$Br$_{3n+1}$ perovskites with different values of $n$ (1 and 2). F, EQE-J values of devices based on 2D-3D perovskite films with different NaBr molar ratios. Reproduced with permission. Copyright 2019 American Chemical Society. G, EQE-current density characteristics of the best perovskite-polymer bulk heterostructure (PPBH) LED. Reproduced with permission. Copyright 2018 Nature Publishing Group.
Lasers

It is well known that well-faceted nanowires with suitable diameters support predominantly axial Fabry-Pérot waveguide modes in nanolasers.\textsuperscript{174} Apart from this, whispering-gallery mode existing in micro/nanodisks can utilize successive total internal reflection along the circumference and provide high cavity quality factor.\textsuperscript{175-178} In 2014, Zhang et al. realized room-temperature near-infrared nanoplatelet lasers using MAPbI$_3$-$x$A$_{x}$ (X = I, Br, Cl) nanoplatelets (Figure 8A).\textsuperscript{176} This wavelength-tunable nanolaser (Figure 8B) could be easily integrated onto conductive platforms like Si, Au, and indium tin oxide, promising for the integrity of existing Si technology. Later, the same group fabricated all inorganic nanoplatelet lasers, where the whispering-gallery mode cavities showed multicolor lasing (Figure 8C) with a low threshold ($\sim 2 \mu$J cm$^{-2}$) and high spectra coherence ($\sim 0.14-0.15$ nm).\textsuperscript{179} 2D RP type perovskites widely integrated in LEDs are also desirable for the application of lasers for its high PL efficiency. Zhang et al. demonstrated a novel microring laser array based on the (BA)$_2$(MA)$_n$PbnBr$_{3n+1}$ perovskites (Figure 8D).\textsuperscript{180}

| Materials | Structures | EL $\lambda_{\text{max}}$ (nm) | Max EQE (%) | Max CE (cd A$^{-1}$) | Max PE (lm W$^{-1}$) | Max L (cd m$^{-2}$) | References |
|-----------|------------|-------------------------------|-------------|---------------------|---------------------|--------------------|------------|
| PEA$_2$MA$_{n-1}$Pb$_n$I$_{16}$ | Thin film | ~760 | 8.8 | N/A | N/A | N/A | 134 |
| NMAI:FABr: PbI$_2$ = 2:1:2 | Thin film | 763 | 11.7 | N/A | N/A | N/A | 135 |
| PEA$_2$(FAPbI$_3$)$_2$PbBr$_4$ | Thin film | 532 | 14.36 | 62.43 | 53.3 | 9120 | 140 |
| PEA$_2$(RbC$_{n-1}$I$_n$)$_2$PbBr$_{10}$ | Thin film | 475 | 1.35 | N/A | N/A | N/A | 100.6 | 136 |
| BA$_2$MA$_3$Pb$_{11}$I$_3$ | Thin film | 733 | 0.2 | N/A | N/A | N/A | 143 |
| BA$_2$MA$_4$Pb$_3$I$_{16}$ | Thin film | 744 | 0.5 | N/A | N/A | N/A | 143 |
| PEA$_2$(FAPbI$_3$)$_2$PbBr$_4$ | Thin film | 532 | 15.4 | 67.5 | N/A | 15 765 | 144 |
| Na$_2$C$_{n-1}$Pb$_n$Br$_{3n+1}$ | Thin film | 518 | 15.9 | 50.3 | 45.1 | 11 560 | 137 |
| BAI:MAPbI$_3$ = 1:5 | Thin film | 748 | 10.4 | 0.09 | 0.10 | N/A | 141 |
| BA$_2$Br$_2$MAPbI$_3$ = 1:5 | Thin film | 513 | 9.3 | 17.1 | 13.0 | N/A | 141 |
| BA$_2$C$_{n-1}$Pb$_n$I$_{3n+1}$ | Thin film | 680 | 6.23 | 1.74 | 1.37 | 1392 | 145 |
| NMA$_2$(FAPbI$_3$)$_{n-1}$Pb$_n$I$_{3n+1}$ | Thin film | 795 | 20.1 | N/A | N/A | N/A | 138 |
| NMA$_2$(CsPbI$_3$)$_n$.Pb$_n$(Cl) | Thin film | 688 | 3.7 | N/A | 0.24 | 440 | 146 |
| PEA$_2$(CsPbI$_3$)$_{n-1}$Pb$_n$(Cl) | Thin film | 480 | 5.7 | 6.1 | N/A | 3780 | 147 |
| MAPbBr$_3$ | Nanoplatelets | 530 | 0.48 | N/A | 1.0 | 10 590 | 142 |
| PEAPbBr$_4$ | Nanoplatelets | 410 | 0.31 | 0.19 | 0.10 | 147.6 | 148 |
| PEAPbBr$_4$ | Nanoplatelets | 410 | 0.038 | N/A | N/A | N/A | 132 |
| MapbBr$_3$ | Nanorods | 533 | N/A | N/A | N/A | N/A | 149 |
| MAPbBr$_3$ | Nanocrystals | 532 | N/A | 9.2 | N/A | 3187 | 150 |
| FA$_2$(CsPbI$_3$)$_2$PbBr$_3$ | Nanocrystals | ~525 | 2.8 | 10.09 | N/A | 55 005 | 151 |
| CsPbBr$_3$ | Nanocrystals | 516 | 0.12 | 0.43 | 0.18 | 946 | 152 |
| CsPbI$_3$ | Nanocrystals | 512 | 6.72 | 13.3 | 5.24 | 15 185 | 153 |
| CsPbBr$_3$ | Nanocrystals | 515 | 3.0 | N/A | N/A | N/A | 330 | 154 |
| CsPbI$_3$ | Nanocrystals | 510 | 0.325 | N/A | N/A | N/A | 934 | 155 |
| CsPbBr$_3$ | Nanocrystals | 518 | 16.48 | 66.7 | 65.9 | 76 940 | 156 |
| CsPbBr$_4$I$_{1-x}$ | Nanocrystals | 653 | 21.3 | 10.6 | N/A | 500 | 157 |
| CsPbI$_3$ | Nanocrystals | 698 | 5.7 | N/A | N/A | 206 | 158 |

Note: EL, electroluminescence; CE, current efficiency; PE, power efficiency; L, luminance; EQE, external quantum efficiency.
population inversion for stimulated emission. Thanks to the well-defined geometries and inherent carrier concentration effect, the microring laser behaved well as an efficient whispering-gallery-mode laser with a very high quality factor (~2600) and low lasing threshold (~13.6 μJ cm⁻²) simultaneously.

**FIGURE 7** A, Schematic of the perovskite nanosheet-based photodetector. B-C, Time-dependent photocurrent and dark current of this perovskite detector. Reproduced with permission.⁷⁸ Copyright 2016 American Chemical Society. D, Optical image of photodetector arrays with a U-shaped mask. E, Schematic illustration of the photodetector arrays with a U-shaped mask under blue LED illumination (wavelength, 463 nm; power density, 600 mW cm⁻²). F, Photocurrent mapping of photodetector arrays with a U-shaped mask under blue LED illumination. Reproduced with permission.⁷⁹ Copyright 2015 AAAS. G, Schematic of a flexible photodetector based on CsPbBr₃ nanosheets. H, I-t curves of the CsPbBr₃ nanosheets-based flexible photodetector bent at different curvatures at a voltage of 5.0 V. The insets are the photographs of the device at different bending states. Reproduced with permission.¹⁶¹ Copyright 2016 Wiley-VCH. I, The schematic of the photodetector consisted of CsPbBr₃ nanosheets and CNTs. J, I-V curves of the photodetector with different amounts of CNTs in dark conditions. Reproduced with permission.¹⁶² Copyright 2017 American Chemical Society
As another basic building block, perovskites nano-wires or nanorods have been widely investigated for their tunable bandgaps, anisotropic electrical/optical properties, and outstanding photoluminescent efficiency. In this section, we will go through the progress on the 1D perovskites, including nanowires and nanorods.

### 3.1 Synthesis methods

#### 3.1.1 Solution-phase synthesis method

The first synthesis of metal halide perovskite nanowires (MAPbI₃) was demonstrated by the Horváth group in 2014 (Figure 9A-E). Two sets of MAPbI₃ nanowires with mean diameters of 50 and 400 nm and lengths up to 10 μm were fabricated by a solution-mediated crystallization process (Figure 9F-I). A thin saturated solution film...
of MAPbI$_3$ in DMF was confined by two glass slides, and the nanowires came into crystallization while gradually sliding the upper glass to expose the liquid to air. It was assumed that the nanowire growth was due to the directional effect of DMF that an internal complex with the methylamine group was formed during the crystallization process.$^{116,167,188,189}$ A great breakthrough in halide perovskite nanowire synthesis was put forward by

**FIGURE 8**  
A, Schematic of the MAPbX$_3$ perovskite nanoplatelet-based laser. The whispering-gallery-mode is illustrated in these nanoplatelets. B, Optical images of two CH$_3$NH$_3$PbI$_3$ nanoplatelets under the illumination of white light (upper) and incidence laser (bottom). Clear diffraction patterns can be seen inside the whispering-gallery-mode cavity. Reproduced with permission.$^{176}$ Copyright 2014 American Chemical Society. C, Lasing spectra from CsPbX$_3$ nanoplatelets with different compositions. The insets are photographs of the corresponding lasers. Reproduced with permission.$^{179}$ Copyright 2016 Wiley-VCH. D, Photoluminescence (PL) images of 3 × 3 microring laser arrays. Spatial interference patterns from all the microrings indicate the coherent outputs. The right figure shows the PL spectra of the ring labeled as 8 vs pump intensity. Reproduced with permission.$^{180}$ Copyright 2018 Wiley-VCH.
Fu et al. (Figure 9J)\textsuperscript{184} They proposed a dissolution-recrystallization model to grow single-crystalline MAPbI\textsubscript{3} nanowires. The pre-deposited poly-crystalline lead iodide (PbI\textsubscript{2}) film was immersed into MAI/isopropanol solution with different concentrations, where high concentration will lead to the formation of thermodynamically...
FIGURE 10  A-B, Optical images and scanning electron microscope (SEM) images of lead halide nanostructures from the vapor-phase synthesis method. Reproduced with permission.\textsuperscript{75} Copyright 2014 Wiley-VCH. C, Optical images of CsPbBr\textsubscript{3} wire networks grown on phlogopite mica from the vapor-phase synthesis method (Scale, 10 μm). The inset is the corresponding 2D Fast Fourier Transform image. Reproduced with permission.\textsuperscript{173} Copyright 2016 American Chemical Society. D-F, SEM images of the vapor-synthesized ultralong CsPbBr\textsubscript{3} nanowires grown along M-plane sapphire at different magnifications. Reproduced with permission.\textsuperscript{172} Copyright 2017 American Chemical Society. G, Transmission electron microscope (TEM) images of MAPbI\textsubscript{3} nanowires obtained through a colloidal synthesis method. Reproduced with permission.\textsuperscript{199} Copyright 2015 American Chemical Society. H-M, TEM images of the shape evolution of CsPbBr\textsubscript{3} nanostructures at different reaction stages (Scale bar, 100 nm). Reproduced with permission.\textsuperscript{190} Copyright 2015 American Chemical Society. N-P, CsPbBr\textsubscript{3} nanowires with different diameters synthesized by controlling the precursor ratio in a colloidal synthesis method. Reproduced with permission.\textsuperscript{200} Copyright 2016 American Chemical Society
favourable $\text{PbI}_4^{2-}$ complex ions after reacting with superfi-
cial MAPbI$_3$ and PbI$_2$. As PbI$_4^{2-}$ ions became over-
saturated, they would react with MA$^+$ and facilitate the
anisotropic growth of nanostructures (Figure 9K). Starting from this method, different kinds of perovskite
nanowires were successfully synthesized. For example, stabilized FAPbI$_3$ nanowires (Figure 9L) and robust
inorganic CsPbX$_3$ nanowires (Figure 9M) were both suc-
cessfully demonstrated by modifying the precursor com-
positions and concentrations feasibly.

3.1.2 Vapor-phase synthesis method

In the past years, the vapor-phase synthesis method
was widely used to fabricate high-quality all inorganic
nanowires like silicon, tin oxide, and zinc oxide and
was then modified for the perovskite nanowire synthe-
sis. In 2014, Ha et al. successfully demonstrated MAPbX$_3$ perovskite nanowires through a two-step
vapor phase method, where lead halide (PbX$_2$) nano-
wires were first deposited by CVD (Figure 10A-B) and
then converted into perovskite through the gas-solid
reaction with amine halide molecules. Compared to
this with two processes, direct vapor-phase growth is
also effective for nanowire fabrication. It is commonly
used for traditional inorganic nanowire synthesis and
unsuitable for organic halide perovskite with low
decomposition temperature. However, when applied to
all inorganic thermal-stable halide perovskite, this
approach will become more feasible. Chen et al. re-
ported a vapor-phase epitaxial growth of horizontal-
tal single-crystalline CsPbX$_3$ nanowires and microwires
with controlled crystallographic orientations. The
asymmetric lattice mismatch between CsPbX$_3$ and mica
substrates leads to strict growth along the [110] direc-
tion and unlimited growth along the [001] direction,
forming different morphologies such as single nano-
wires, Y-shaped branches, interconnected nanowire,
or microwire networks (Figure 10C). Later, based on a
similar direct vapor-phase synthesis, Shoaib et al. fabricated ultralong CsPbBr$_3$ nanowires (Figure 10D-F), induced by graphoepitaxial effect on M-plane
sapphire substrates, with diameters of several hundred nanometers and lengths up to several
millimeters.

3.1.3 Colloidal synthesis method

The successful appliance of the colloidal method on the
synthesis of perovskite quantum dots promotes it to the
fabrication of perovskite nanowires. The first colloidal
synthesis was based on an antisolvent precipitation
method. Zhu et al. dissolved PbX$_2$, MAX, and long-chain
CH$_3$(CH$_2$)$_2$NH$_3$X (X = Cl/Br/I) in a polar solvent (such
as ACN) and added the mixture into a solvent with lower
polarity (such as toluene), obtaining perovskite nano-
wires with precise control (Figure 10G). When
switching to inorganic nanowires, representative works
employing the hot-injection method were performed by
Zhang et al. Through injecting the cesium precur-
sor into the PbX$_2$ solution in the presence of oleylamine
and oleic acid at 150°C-250°C, they demonstrated the
fabrication of inorganic CsPbX$_3$ nanowires for the first
time and controlled the compositions and diameters in their subsequent work, enabling these
nanowires potential candidates for optoelectrical appli-
cations. Afterward, colloid synthesis methods were
further investigated with more precise control. For
instance, Imran et al. prepared size-tunable CsPbBr$_3$
nanowires by introducing carboxylic acids with short
aliphatic chains (Figure 10N-P). High PL quantum
yield up to 77% and color tunability from blue to green
were both obtained in these colloidal synthesized
nanowires.

3.1.4 Templated synthesis method

Using a template that confines the precursor to form the
desired structures seems to be straightforward. Ashley
et al. fabricated MAPbI$_3$ nanowires by selecting an AAO
membrane as the template (Figure 11A). They drop-
cast perovskite precursor into the AAO pores and
followed with an annealing process to enable the for-
amation. One obvious advantage here is the amenable diam-
ter control by selecting varisized AAO templates. In their
demonstration, MAPbI$_3$ nanowires with diameters rang-
ing from 50 to 200 nm, with a dispersity less than 10%,
were synthesized readily. Later, Waleed et al. combined
the vapor-phase process with this template confinement
method, fabricating lead-free and inorganic perovskite
nanowires with a start from pure metals (Figure 11B). To
be specific, AAO membranes were still chosen as the template, and metal tin or lead was
electrochemically deposited in the bottom of the
nanopores. MAI or CsI gas, produced by heating the
corresponding powders, were transported by the carrier
gas and reacted with the as-deposited metal, facilitating
the forming of final MASnI$_3$ or CsPbI$_3$ nanowires
(Figure 11B,C). Oener et al. devised a nanowire extru-
sion method where precursor was first dropped on the
AAO templates with double-sided pores. Then a reduced
pressure by a syringe was applied to enable the precursor
to fill in the pores of the template and extrude out on the
3.2 | Applications of 1D metal halide perovskites

3.2.1 | Lasers

Anisotropic nanowires can serve as a waveguide along the axial direction, and the two end facets can form a Fabry-Perot cavity for optical amplification. Therefore, the single-crystalline perovskite nanowires with ideal facets are good candidates for the optically pumped lasers. The first lead halide perovskite nanowire-based laser was demonstrated by Zhu et al. (Figure 12A) from a room-temperature solution method, showing wavelength-tunable lasing with very low thresholds (220 nJ \( \text{cm}^{-2} \)) and a high-quality factor (\( Q \approx 3600 \)) (Figure 12B). The further analysis estimated that the lasing quantum yield approaches 100% due to little charge carrier trapping in these nanowires. Using a similar solution method, nanowire lasers based on FAPbI\(_3\) and their stabilized alloys were proposed by the same group (Figure 12C). The lasers exhibited durable lasing under...
FIGURE 12  Legend on next page.
-10⁸ shots of sustained illumination of pulsed laser excitation, substantially exceeding the stability of MAPbI₃ (~10⁷ shots) (Figure 12D). In addition to the solution process, vapor-phase synthesized nanowires were also suitable for nanolasers. The optically pumped MAPbI₃ nanowire lasers exhibited a wavelength of 777 nm, a threshold of 11 μJ cm⁻², and a quality factor of 405 (Figure 12E-G). In consideration of the stability issue, Eaton et al. first moved their eyesight onto inorganic CsPbX₃ nanowire-based nanolasers (Figure 12H). Compared to the organic counterparts, the inorganic metal halide nanowires showed robust property without sacrificing high emitting performance (Figure 12I). Low lasing threshold, high-quality factors, and robust stability under ambient conditions render the inorganic nanowires a promising platform for nano-photonic integrity.

### 3.2.2 Solar cells

Due to the enhanced mechanical property and better conduction in specific directions, perovskite nanowires are also explored as absorber in solar cells. Park et al. first incorporated MAPbI₃ nanowires into the solar cell (Figure 13A) and investigated its optical and electrical properties. Compared to the nano-cubic counterparts, higher lateral conductivity was observed in nanowires, revealing a better connecting pathway. Time-resolved fluorescence spectroscopy confirmed that hole extraction from the nanowires was more efficient than that in the bulk case, mainly due to the better contact with hole transport materials from increased surface area. As a result, an improved PV performance with the best PCE of 14.71% was proved in nanowire-based solar cells (Figure 13B). Since then, a series of solar cells based on MAPbI₃ perovskite nanowires have emerged with modified fabrication processes (Figure 13C). A PCE of 18.83% with long-term stability was achieved through an integrated approach by Chang et al. (Figure 13D). In 2017, Kuang et al. fabricated solar cells based on CsPbX₃ nanowires with enhanced thermal and humidity stability compared to organic counterparts. The CsPbI₃ and CsPbBr₃ nanowire-based solar cells yielded the PCEs of 0.11% and 1.21%, respectively (Figure 13E). An impressive work was done in lead-free perovskites where Chen et al. synthesized high-quality CsSnX₃ (X = Cl/Br/I) nanorods for solar cell absorbers (Figure 13F). A PCE up to 12.96% under AM 1.5G has been achieved, revealing the potential of Sn-based perovskite nanorods in solar cell applications (Figure 13G). A summary of the key parameters of solar cells based on 1D perovskites is exhibited in Table 1.

### 3.2.3 Light-emitting diodes

In 2015, through a solution method, Wong et al. successfully synthesized MAPbBr₃ perovskite nanorod arrays (Figure 14A), which were further converted into MAPbI₃ nanorods by an anion exchange reaction. Both types of nanorod arrays were applied as the active layer in LED devices (Figure 14B) and exhibited electroluminescent at room temperature (Figure 14C). Compared to thin films, the vertically oriented nanorod arrays offers several advantages, such as large active surface area, high carrier injection efficiency, and improved strain relaxation from thermal expansion. In their work, the MAPbBr₃ nanorod-based LED showed green EL at 532 nm with a full width at half maximum (FWHM) of 26 nm (Figure 14D) and the converted MAPbI₃ arrays showed infrared EL at 782 with an FWHM of 41 nm. This work demonstrates the potential of large-scale nanorod fabrication in LED applications.

### 3.2.4 Photodetectors

In 2014, Horvath did the pioneering work of perovskite nanowire-based photodetectors (Figure 15A). Although the responsibility of this detector is 5 mA W⁻¹, 4 orders of magnitude smaller than that of state-of-art photodetectors.
detectors made from monolayer graphene, the response times (rise and decay times) were within 500 μs, ~10^4 faster than that of graphene-based detectors (Figure 15B).213-216 The photocurrent-dark current ratio and EQE at low voltages (<0.5 V) both showed superior performance than that based on their nanoparticle form, mainly arising from the reduced grain boundaries, which permit smooth carrier flowing channels in transport. In addition to these merits, enhanced absorption and mechanical integrity are other outstanding advantages. Zhu et al. demonstrated a MAPbI₃ nanowire-based photodetector on a flexible substrate from a post-solution treatment, revealing a 10% increase in absorption and enhanced mechanical property compared to thin-film based one (Figure 15C).167 Utilizing the morphological anisotropy of nanowires, Gao et al. demonstrated a photodetector with high polarization sensitivity and detectivity (2 × 10^{13} Jones) for the first time.166 Considering the instability issues, several groups moved interests onto the inorganic counterparts.192,203 Waleed et al. used the AAO templates to fabricate CsPbI₃ nanowire-based photodetector, showing high stability when stored in organic polar solvents for 30 days.203 Zhang et al. formed a
thin CsPbBr₃-nanowires based photodetector, showing a sensitive photoresponse (Figure 15D). For practical spatial image applications, individual photodetectors should be integrated into arrays for graphic detection. Deng et al. demonstrated photodetector arrays for image mapping by synthesizing perovskite networks on poly(ethylene terephthalate) substrates. These arrays consisted of 49 individual detectors and could obtain a clear mapping of light source (Figure 15E). In the later work, Gu et al. increased the number of pixels to 1024 in each image sensor using well-aligned nanowires by a templated method discussed above (Figure 15F). The still images and videos could be well captured by these high-resolution nanowire arrays (Figure 15G). For further comparison, we summarize the key parameters of photodetectors based on 1D perovskites in Table 3.

4 | 0D METAL HALIDE PEROVSKITES

4.1 | Synthesis methods

4.1.1 | Ligand-assisted precipitation method

In 2014, for the first time, Schmidt et al. synthesized MAPbBr₃ nanocrystals (Figure 16A) by reacting the mixture of MABr and a long-chain alkyl ammonium bromide with PbBr₂ in the presence of oleic acid and octadecene. The nanoparticles were then precipitated by the addition of acetone and centrifugation. The as-synthesized nanoparticles exhibited diameters of ~6 nm and a strong PL intensity around 525 nm. Later, this method was adopted by several groups with further modification. For example, Zhang et al. produced brightly luminescent MAPbBr₃ nanocrystals with absolute PLQY up to 70% by the incorporation of n-octylamine and oleic acid as ligands (Figure 16B-D). The nanocrystals show an average size of 3.3 nm and binding energy to ~350 meV. Wei et al. synthesized perovskite nanocrystals by a homogeneous reaction in various nonpolar organic solvents (Figure 16E-G). Notably, this approach could be conducted in the open air and further extended to a gram level.

4.1.2 | Hot-injection method

In addition to ligand-assisted precipitation, hot-injection is another efficient method to synthesize monodisperse perovskite nanocrystals. Inorganic CsPbX₃ nanocrystals were achieved (Figure 17A-C) by the injection of cesium oleate into a mixed solution of PbX₂, oleic acid, and oleylamine in a high boiling solvent (octadecene) at
The synthesized nanocrystals showed stable, narrow, and broadly tunable photoluminescence with a quantum yield of 50%-90%. Subsequently, a low-temperature, partial/complete anion-exchange in CsPbX\textsubscript{3} nanocrystals was demonstrated by the same group (Figure 17D).\textsuperscript{221} The emission could cover the visible spectral region (410-700 nm) by reacting with suitable reagents in the precursors (Figure 17E). An inter-nanocrystal anion-exchange was further proposed, which could achieve CsPbCl\textsubscript{3-x}Br\textsubscript{x} or CsPbBr\textsubscript{3-x}I\textsubscript{x} compositions by readily mixing CsPbCl\textsubscript{3}, CsPbBr\textsubscript{3}, and CsPbI\textsubscript{3} nanocrystals in proper ratios (Figure 17F).

4.1.3 | Template-assisted method

Similar to 2D or 1D nanostructure fabrication, the template-assisted method is also suitable for perovskite nanocrystal synthesis. Kojima et al. demonstrated the MAPbBr\textsubscript{3} nanocrystal synthesis using a mesoporous alumina oxide in 2012.
(Figure 18A).\textsuperscript{224} The MAPbBr\textsubscript{3} nanocrystals were formed on porous Al\textsubscript{2}O\textsubscript{3} film due to a rapid self-organized process after the dry of solvents. In 2015, Longo et al. utilized Al\textsubscript{2}O\textsubscript{3} nanocrystals, with which MABr and PbBr\textsubscript{2} were mixed, to prepare perovskite nanocrystals with high PLQY.\textsuperscript{227} Spin-coating the mixture on a glass/quartz substrate and annealing the sample, the perovskite nanocrystals were formed with Al\textsubscript{2}O\textsubscript{3} particles surrounding. The dimensions of perovskite nanocrystals were precisely governed by the sizes and concentrations of alumina nanoparticles. In addition, porous silica could also be used as templates. Malgras (Figure 18B-D) and Dirin et al. (Figure 18E-F) proposed a similar approach that they infiltrated perovskite solution into mesoporous silica followed by drying, synthesizing desired perovskite nanoparticles by the confinement of templates.\textsuperscript{225,226} Size distributions could be adjusted by the pores of silica to even 3.3 nm without sacrificing the high luminescent efficiency.

\section*{4.2 Applications}
\subsection*{4.2.1 Solar cells}

The pioneering work by Mayasiki in 2009 successfully demonstrated a perovskite nanocrystal-based solar cell,
exhibiting a PCE of 3.8% (Figure 19A). The performance was further improved by the Park group to 6.54% (Figure 19B-E), a record efficiency among perovskite nanocrystal-based PV devices. The research focus then moves onto inorganic counterparts like CsPbI3 quantum dots owing to the consideration of tandem devices. Swarnkar et al. in 2016 developed a novel process to purify the as-formed quantum dots by the selection of methyl acetate, an antisolvent that removes excess unreacted reagents. The stability was significantly enhanced for the purified sample, which was stable in the cubic phase for months (Figure 19F). Solar cells based on these stable quantum dots showed an impressive PCE exceeding 10%, with a high open-circuit voltage up to 1.23 V. Further investigation, like utilizing AX salts like FAI to tune the coupling between perovskite quantum dots and increase charge transport properties between quantum dots arrays, boosted a record efficiency of quantum dots-based solar cell up to 13.4% (Figure 19H-J). A summary of the performance of perovskite nanocrystal-based solar cells is listed in Table 1.

4.2.2 | LEDs

High binding energy, strong PLQY together with tunable color gamut made perovskite quantum dots rather promising for LED fabrication. Song et al. used inorganic CsPbX3 quantum dots as emitters in LEDs for the first time (Figure 20A). It showed a wide color tunability from blue to orange by simply changing the anionic composition, but a low luminance and EQE accompanied simultaneously (Figure 20B). The moderate performance...
is possibly attributed to the excess long ligand on quantum dots’ surface that hampered the efficient carrier transporting. Replacing these long ligands with shorter one would address this issue effectively. Pan et al. realized efficient LEDs based on CsPbX$_3$ quantum dots, which were capped by relatively short ligand (e.g., diodecyl dimethyl ammonium bromide) (Figure 20C), and achieved much improved EQE (1.9% for the blue, and 3.0% for the green) (Figure 20D-E).\textsuperscript{154} Li et al. achieved a 50-fold EQE enhancement by balancing surface passivation and carrier injection of perovskite quantum dots (Figure 20F). An EQE up to 6.27% in the CsPbBr$_3$ nanocrystal-based LED has been obtained by precisely controlling the ligand density on the surface. Besides, the current efficiency was 30-fold improved up to 13.3 cd A$^{-1}$, as well (Figure 20G).\textsuperscript{153} In addition to these solutions, other strategies like surface engineering and so on were also proposed to address the remaining problem,
significantly accelerating the development of nanocrystals based LEDs. Recently, some types of 0D perovskites were demonstrated to possess broadband emission, which are promising candidates for the applications of down-conversion white LEDs. Zhou et al. replaced the element lead by tin and selected different ligands, demonstrating 0D perovskites with tunable broad emission. Through the manipulation of locally collective hydrogen bonding, Cui et al. successfully tailored the assembling behavior and synthesized 0D perovskite single crystal with “warm” white emission and enhanced PL efficiency. By combining the hybrid

**FIGURE 19**  
A, J-V curves of perovskite solar cells based on MAPbI$_3$ quantum dots (dashed line) and MAPbBr$_3$ quantum dots (solid line). Reproduced with permission. Copyright 2009 American Chemical Society.  
B, Photographs of MAPbI$_3$ quantum dot-sensitized 5.5 μm thick TiO$_2$ films fabricated with different precursor concentrations. From left to right the concentrations are 10.05, 20.13, 30.18, and 40.26 wt %.  
C, J-V curves of solar cells based MAPbI$_3$ quantum dot-sensitized 5.5 μm thick TiO$_2$ films with different concentrations.  
D, Absorption spectra of MAPbI$_3$ quantum dot-sensitized 1.4 μm thick TiO$_2$ films with different concentrations.  
E, The corresponding external quantum efficiency (EQE) spectra of devices in figure C. Reproduced with permission. Copyright 2011 The Royal Society of Chemistry.  
F, Device structure of a CsPbI$_3$ quantum dot-based solar cell and a corresponding cross-sectional scanning electron microscope (SEM) image of the device.  
G, J-V curves of the CsPbI$_3$ quantum dot-based solar cell measured at different times. Reproduced with permission. Copyright 2016 AAAS.  
H, J-V curves of the CsPbI$_3$ quantum dot-based solar cell with a certified efficiency.  
I, I-t curve of the corresponding device in figure H, showing a stabilized PCE of 13.43%.  
J, EQE spectrum of the corresponding device in figure H. Reproduced with permission.
with the commercial phosphor in various ratios, Worku et al. fabricated UV-pumped white LEDs with tunable color gamut, from “cold” to “warm” white. More detailed parameters of LEDs based on perovskite nanocrystals are listed in Table 2.

4.2.3 Lasers

In 2015, Yakunin demonstrated the spontaneous emission and lasing in blue and green spectral regions from ~10 nm monodispersed CsPbX₃ nanocrystals. Low pump thresholds down to 5 ± 1 μJ cm⁻², together with high values of a modal net gain of at least 450 ± 30 cm⁻¹, were demonstrated in this system (Figure 21A). Two lasing modes were observed. One was the whispering-gallery-mode where silica microspheres as high-finesse resonators were coated with CsPbX₃ nanocrystals and the other was random lasing in films of CsPbX₃ nanocrystals. Later work by Wang et al. demonstrated a CsPbX₃ nanocrystal-based laser, with a low threshold of 22 μJ cm⁻², wide color tunability, and good stability (Figure 21B). Based on this, the same group investigated the manifested nonlinear absorption and multiphoton pumped stimulated emission from 9 nm-sized CsPbBr₃ nanocrystals (Figure 21C). Low-threshold and tunable frequency-
upconverted stimulated emission by two-photon absorption and green stimulated emission by three-photon were both obtained from the CsPbClₓBr₃₋ₓ nanocrystals, providing promising opportunities in nonlinear photonics (Figure 21D).

5.1 | Precise control of the size and dimensionality

As one of the most valuable merits with reduced dimensionality, size and dimensional control for various bandgaps attract tremendous attention. Nowadays, although some groups can manipulate the perovskites with size and shape control to some degree, most synthesis methods for low-dimensional perovskites are trial-and-error, without a precise regulation. With the emerging machine learning techniques recently, Lu et al. can predict enormous perovskite structures and even material stability, according to the former database. We are inquisitive whether this approach can be utilized as the guidelines for the synthesis of

5 | OUTLOOK

Although 3D perovskite has been investigated intensively, the exploration of low-dimensional perovskites has not caught the pace of 3D counterparts in multiple aspects. It can be expected that advantages arising from low dimensionality can be further magnified both fundamentally and practically.
perovskites with accurate structures and dimensions we desire. Based on these, more explorations of in-depth physical mechanism and wide applications will be duly concomitant.

5.2 Understanding of the physics in low-dimensional perovskites

Since tremendous optoelectronic applications have been demonstrated, the investigations focusing on fundamental properties of low-dimensional perovskites have not attracted much attention. With the reduced dimensions, some inherent natures like band structure and photon transport will behave differently. For example, reduced thermal conductance from the enhanced boundary scattering, together with the improved Seebeck coefficient from the modified energy band, makes low-dimensional perovskites potential materials for thermoelectrics. Recently, Wang et al. have investigated the cation dynamics governed by thermal properties in different types of perovskite nanowires. However, the study on this aspect is limited and more efforts should be devoted to further exploration. In addition, the low dimensionality could also bring about confined charge transport in the selected directions, leading to unique electrical properties such as 2D electron gas, which is the basis for field-effect devices. However, these fundamental investigations rather lag behind the application and is supposed to draw more attention in the near future.

5.3 Exploration of new applications

In addition to the applications in solar cells, LEDs, lasers, and photodetectors, metal halide perovskites may possess other suitable uses. For example, Liao et al. have demonstrated ferroelectricity in bulk 3D perovskite. It is expected that the strong quantum and dielectric confinements of nanostructures may bring up much-enhanced ferroelectricity, which calls for more systematic investigations in low-dimensional nanoplatelets or nanowires.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest.

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