Low-Dimensional Metal Halide Perovskite Crystal Materials: Structure Strategies and Luminescence Applications

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Replacing methylammonium (MA\textsuperscript{+}), formamidine (FA\textsuperscript{+}), and/or cesium (Cs\textsuperscript{+}) in 3D metal halide perovskites by larger organic cations have built a series of low-dimensional metal halide perovskites (LDMHPs) in which the inorganic metal halide octahedra arranging in the forms of 2D layers, 1D chains, and 0D points. These LDMHPs exhibit significantly different optoelectronic properties from 3D metal halide perovskites (MHPs) due to their unique quantum confinement effects and large exciton binding energies. In particular, LDMHPs often have excellent broadband luminescence from self-trapped excitons. Chemical composition, hydrogen bonding, and external factors (temperature and pressure etc.) determine structures and influence photoelectric properties of LDMHPs greatly, and especially it seems that there is no definite regulation to predict the structure and photoelectric properties when a random cation, metal, and halide is chosen to design a LDMHP. Therefore, this review discusses the construction strategies of the recent reported LDMHPs and their application progress in the luminescence field for a better understanding of these factors and a prospect for LDMHPs’ development in the future.

1. Introduction

As promising photoelectric materials, metal halide perovskites (MHPs) have been widely studied and applied to photovoltaic devices, photodetectors, light emitting diodes (LEDs), etc.\textsuperscript{[1]} In particular, MHPs have unique structure tunability, the FA\textsuperscript{+}, MA\textsuperscript{+}, or Cs\textsuperscript{+} in 3D MHPs replaced by larger sized organic cations can build various low-dimensional MHPs (LDMHPs) at the molecular level, such as 2D, 1D, and 0D metal halide crystal materials.\textsuperscript{[2]} 3D MHPs (ABX\textsubscript{3}, A stands for monovalent cation, such as FA\textsuperscript{+}, MA\textsuperscript{+}, Cs\textsuperscript{+}; M represents metal ions, such as Pb\textsuperscript{2+}, Sn\textsuperscript{2+}, Ge\textsuperscript{2+}, etc., and X stands for halogen ion Cl\textsuperscript{−}, Br\textsuperscript{−}, I\textsuperscript{−})\textsuperscript{[3]} with the advantages of high absorption coefficient, adjustable optical bandgap, low exciton binding energy, and long carrier diffusion distance have been widely used in the field of solar cells with a certified power conversion efficiency (PCE) of world recorded over 25\%.\textsuperscript{[4]} In general, 3D perovskites have small exciton binding energies (\textlesseqqless 20–50 meV),\textsuperscript{[5]} which lead to their low radiation recombination efficiency and photoluminescent quantum efficiency (PLQE), and limit their application in luminescence.\textsuperscript{[6]} On the contrary, LDMHPs commonly exhibit excellent luminescence due to their quantum confined effect and large exciton binding energies.\textsuperscript{[7]} Different from nanosheets, nanowires, and quantum dots at the structural level, LDMHPs discussed in this work refer to the low-dimensional crystals at the molecular level in particular.

The selection of cations, metals, and halogens play a key role in determining chemical composition and crystal structures of LDMHPs. Crystals with different dimensions were constructed by selecting the size of organic cations, and regulating metal cations could also construct LDMHPs with different dimensions.\textsuperscript{[8]} In addition to single metal, double perovskite, and quadruple perovskites could be formed by mixing metal cations.\textsuperscript{[9]} The change of halogen could regulate the configuration and the distortion degree of LDMHPs, which is the key to realize the broadband white luminescence.\textsuperscript{[10]} Generally, controlling growth of all-inorganic Cs-Pb-X perovskites with different dimensions could be easily realized by regulating the stoichiometric ratio of the precursor solution during the crystal growth progress.\textsuperscript{[11]} However, for organic–inorganic hybrid perovskites, a kind organic cation usually only constructed a structure even the stoichiometric ratio of every elements in the precursor solution was changed unless it formed different valence states or interacted with other molecules. On the other hand, manipulating the hydrogen bonding interactions between the inorganic skeletons and organic cations could significantly affect the orientation and conformation of inorganic skeleton.\textsuperscript{[12]} Furthermore, external pressure and temperature will provide thermodynamic energies to change the structures and photoelectric properties of LDMHPs.\textsuperscript{[13,14]}

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Recently, it has spawned the “diamond fever” on the family of LDMHPs, and they have been intensely studied and applied as promising new photoelectric functional materials due to excellent luminescence properties and good stabilities. In particular, LDMHPs exhibit typical luminescent properties of large octahedrons, which is conforming to the theoretical formula of $R_{\text{octahedrons}}$, where $R_A$, $R_M$, and $R_X$ are the maximum values ($R_{\text{Pb}} = 1.19 \, \text{Å}$, $R_i = 2.20 \, \text{Å}$), the maximum $R_\text{A}$ approximately is 2.6 Å. Therefore, only small cations which maximum lengths are shorter than 2.6 Å can satisfy the condition to build 3D APb$_i$$_j$$_k$$_l$ perovskites. Correspondingly, replacing the FA$^+$, MA$^+$, or Cs$^+$ with larger size organic cations, abundant LDMHPs can be obtained, including layered 2D, linear 1D and points distributed 0D metal halide hybrids (Figure 1b–d).

The size of cations affects the connection modes of lead halide octahedrons. There are three types of connection modes for metal halide perovskites: corner-sharing, edge-sharing, and face-sharing as shown in Figure 2a. Although most configurations of 2D perovskites are corner-sharing, a few 2D perovskites have broken the conventional connection mode and formed edge-sharing and face-sharing structures. The types of connection modes can be regulated by changing the length of amines. For example, as shown in Figure 2b–e, C$_6$H$_5$(CH$_2$)$_n$NH$_3^+$, where $n = 1, 2, 3, 4$ represents organic phenylammonium (PMA), phenylethylammonium (PEA), phenylethylammonium (PPE), and phenylethylammonium (PBA), respectively. The 2D structures constructed by PMA and PEA are corner-sharing, while the 2D structures constructed by the longer PPA and PBA are corner-sharing and face-sharing coexistence. Recently, the post-perovskite with edge-sharing structure formed by organic cation trans-2, 5-dimethylpiperezine has been reported. Due to the distorted inorganic lead halogen structure, it exhibits white emission with a high PLQE. Furthermore, the connection mode influences the orbital overlap between metals and halogens, thus affecting the bandgap values of 2D perovskites, where it is common that face-sharing $>$ edge-sharing $>$ corner-sharing for bandgap values.

For common 2D perovskites, organic cations which are suitable for 2D interlayer frames often contain a positive charge (e.g., RNH$_3^+$, R$_n$NH$_2^+$, R$_n$NH$^+$, R$_n$N$^+$ (R is a paraffinic chain)); hydrogen atoms of the organic amine can interact with halide atoms on metal halide octahedrons by hydrogen bonding. The distance between the inorganic layers in 2D perovskites can be adjusted by organic cations with short or long paraffinic chains, and perovskites in lower dimensions can be obtained by adjusting longer paraffinic chains. For example, 2D perovskites (R$_n$NH$_2$)$_2$MX$_4$ and (NH$_4$R$_n$NH$_3$)MX$_4$ constructed by monoamine and dianmine organic cations are shown in Figure 4. For (R$_n$NH$_2$)$_2$MX$_4$, when $n = 1$, methylamine cation construct the common 3D perovskite MAPbX$_4$ and when $n = 2$, ethylamine, it construct the 2D perovskites EA$_4$Pb$_3$X$_{10}$. EA has two configurations in two positions, as some of the cations are filled into 3D perovskite cavity and another part of the EA that splits into inorganic layer to form a 2D perovskite. In addition, when $n > 2$, the organic cations will construct single layer 2D perovskites. For (NH$_4$R$_n$NH$_3$)MX$_4$, the dianmine organic cations are long enough to link the two inorganic layers as the $n$ values increases. For example, when $n \geq 4$, such as [NH$_4$(CH$_2$)$_n$NH$_3$]Pb$_4$ ($n = 4, 6, 8$), the dianmine organic cations are long enough to construct regular 100-oriented 2D perovskites. The other good example is that, for butyric diamine ($n = 4$), propylene diamine ($n = 3$), and ethylenediamine ($n = 2$), as the length of the amine chain decreases, the layered
perovskite changes from a 100-oriented flat layer to a distorted layer, and then when \( n = 2 \), the layer broken to construct a 1D ribbon structure.\(^{[25]}\)

Recently, Ma’s group synthesized multiple perovskites with 1D and 0D structures by diamine cation with the \( N, N' \)-2-dimethylethlyenediamine (organic cation 17 in Figure 4).

Such as the 1D \( \text{C}_4\text{N}_2\text{H}_{14}\text{PbBr}_4 \) with efficient bluish white-light emission,\(^{[19]}\) as shown in Figure 3a. The \( \text{C}_4\text{N}_2\text{H}_{14}\text{PbBr}_4 \) achieved white light emission with PLQE of 28% by doping \( \text{Mn}^{2+} \) (Figure 3b).\(^{[26]}\) The author also reported 1D perovskite \( \text{C}_4\text{N}_2\text{H}_{14}\text{PbCl}_4 \) (Figure 3c) and studied its structure distortion.\(^{[27]}\) As shown in Figure 3d, from no luminescence property to

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**Figure 2.** (a) 2D structure of corner-sharing, edge-sharing and face-sharing. Reproduced with permission.\(^{[7]}\) Copyright 2019, American Chemical Society. (b–e) 2D perovskites constructed by PMA, PEA, PPA, and PBA, respectively. Reproduced with permission.\(^{[20]}\) Copyright 2016, American Chemical Society.

**Figure 3.** (a) Structure of 1D organic lead halide perovskites \( \text{C}_4\text{N}_2\text{H}_{14}\text{PbBr}_4 \) constructed by \( \text{C}_4\text{N}_2\text{H}_{14}^{2+} \). Reproduced with permission.\(^{[19]}\) Copyright 2017, Springer Nature. (b) Preparation of 28% white light emission system with 1D \( \text{C}_4\text{N}_2\text{H}_{14}\text{PbBr}_4 \) doping \( \text{Mn}^{2+} \). Reproduced with permission.\(^{[26]}\) Copyright 2017, American Chemical Society. (c) Structure of 1D organic lead halide perovskites \( \text{C}_4\text{N}_2\text{H}_{14}\text{PbCl}_4 \). Reproduced with permission.\(^{[27]}\) Copyright 2018, American Chemical Society. (d) 1D Sn-based \( \text{C}_4\text{N}_2\text{H}_{14}\text{SnBr}_6 \) transformed into a 0D structure \( (\text{C}_4\text{N}_2\text{H}_{14}\text{Br})_4\text{SnBr}_6 \). Reproduced with permission.\(^{[28]}\) Copyright 2017, Wiley-VCH. (e) Structure of core–shell 0D perovskites \( (\text{C}_4\text{N}_2\text{H}_{14}X)_4\text{SnX}_6 \) (\( X = \text{Br} \) or \( \text{I} \)). Reproduced with permission.\(^{[29]}\) Copyright 2018, Royal Society of Chemistry.
yellowish-white emission, 1D Sn-based $C_4N_2H_14SnBr_6$ is easier to transform into a 0D of $(C_4N_2H_14Br)_4SnBr_6$.[28] And the Sn-based 0D $(C_4N_2H_14X)_4SnX_6 (X^- = Br^-, I^-)$ has a PLQE of nearly unity (Figure 3e).[29]

Although the organic cations in 2D structure are not as small as the cations in 3D perovskites, they still need to fit into the frame of 2D inorganic layers, and the cross-sectional area of organic molecule must be suitable to the square range constructed by the four corner-sharing lead halide octahedrons, allowing the organic molecule to tilt and interlace, where the side length of the square is greater than or equal to twice the average bond length of the Pb-X. The space cannot accommodate the too large adjacent organic molecules, because the larger cations more possibly construct lower dimensional structures, such as 1D and 0D structures. For organic molecules with large cross-sectional area, it’s more likely to break up the 2D inorganic layers to form a 1D or 0D structure. For example, the large circular organic molecules $C_9NH_{20}^+$ (organic cation 2 in Figure 4) constructed a variety of 0D MHPs, such as $(C_9NH_{20})_7(PbCl_4)Pb_3Cl_{11}$ with a blue emission,[31] $(C_9NH_{20})_2SnBr_4$ with a deep-red emission,[32] $(C_9NH_{20})_2SbCl_5$ with a yellow emission, and $(bpy)_9[ZnCl_4]_2[Pb_3Cl_{11}]$ with a green emission, etc. Organic cations with large cross-sectional area in 1D and 0D structures are summarized in Figure 4, and the optical properties of these LDMHPs constructed by corresponding molecules are shown in Table 1. In summary, the size of the organic cations plays a key role in constructing LDMHPs’ structures.
Most 2D perovskites are in \((NH_4RNH_4)MX_2\) and \((RNH_4)_2MX_4\) structures, where M is usually a divalent metal, e.g., \( Pb^{2+}, Sn^{4+}, Ge^{4+}, Mn^{2+}, Fe^{2+}, Co^{2+}, Cu^{2+}\). Here, \( Sn^{2+}\) and \( Ge^{2+}\) are often introduced to replace toxic \( Pb^{2+}\) in perovskites, but their stability is very poor, as the \( Sn^{2+}\) and \( Ge^{2+}\) ions are easily oxidized to \( Sn^{4+}\) and \( Ge^{4+}\).[49] Recently, the \( Cu^{2+}\) based perovskites have attracted attention due to their good stability, such as the 2D 100-oriented perovskite \( MA_2CuCl_4\), prepared by MA cations.[54]

In addition, a series of 2D perovskites with different organic cations including \( (C_4H_9NH_3)_2CuBr_4\), \( (C_5H_9NH_3)_2CuBr_4\), and \( (C_6H_5CH_2CH_2NH_3)_2CuCl_4\), \( (C_4H_9NH_3)_2MCl_4\), and \( (C_3H_7NH_3)_2CuBr_4\), \( (C_5H_9NH_3)_2CuBr_4\), and \( (C_6H_5CH_2CH_2NH_3)_2CuCl_4\) were successfully synthesized by MA cations.[54] The synthesis and characterization of \( (C_6H_5NH_3)_2CuCl_4\) and \( (C_6H_5NH_3)_2CuBr_4\) have been reported.[54]

The layered double perovskite \( Cs_3CuSbCl_6\), with two metal ions \( (Cu^{2+} and Sb^{4+})\) was synthesized for the first time.[50] As shown in Figure 5a, the number of inorganic layers regulation of this material has been realized, which initiated a new chemical component design strategy to regulate the layers of 111-oriented 2D perovskites. It’s worth mentioning that Bi metals formed the 0D structures, such as \( (C_3N_3H_11O)_2PbBr_6\) (Figure 5b) with \( Bi^{3+}\) and \( ABIBr_6\) (A = \( Cs^+\), \( MA^+\), \( FA^+\), \( C_5H_7N^+, \( C_7H_7N^+, \( C_8H_8N^+, \( N\)-EtPy) + with \( Bi^{3+}\) (Figure 5c).[51] It has been reported that for 0D double perovskite \( (C_5H_7NH_3)_2BiCl_3\) \( [BiCl_3]^+\) and \( [SbBr_6]^3-\) octahedrons are completely isolated and surrounded by large organic \( C_5H_7N^+\), and so they have exhibited a great stability.

The diversified options of metal ions can not only construct the double perovskites,[9] but also theoretically design the structure of quadruple perovskites. For example, Lin et al. propose a strategy to design quadruple perovskites by introducing heterovalent cation to form double perovskites. And these two stable quadruple perovskite halides of \( Cs_3CdSb_2Cl_{12}\) and \( Cs_4CdBi_3Cl_{12}\) with vacancy-ordered structures were successfully synthesized by solvent heat method and exhibited broadband emissions.[61] The figures (1–12) highlighted in blue refer to the corresponding organic cations shown in Figure 4.

### Table 1. Summary of reported organic cations of LDMHPs.

| Dimension | Formula                  | Connecting type | Fabrication              | Peak [nm] | FWHM [nm] | PLQE [%] |
|-----------|--------------------------|-----------------|--------------------------|----------|-----------|---------|
| 1-0D[20]  | \((Ph_4P)_2SbCl_3\)      | N/A             | Antisolvent method       | 648      | 136       | 87      |
| 2-0D[20]  | \((C_3N_3H_11O)_2PbBr_6\)| N/A             | Antisolvent method       | 590      | 119       | 98±2 2  |
| 2-0D[31]  | \((C_9NH_20)_2S\)        | N/A             | Antisolvent method       | 470      | 84        | 83      |
| 2-0D[27]  | \((C_9NH_20)_7(PbCl_4)\) | N/A             | Solution method          | 695      | 146       | 46      |
| 3-0D[33]  | \((C_4H_8N_2H_4)_2PbBr_6\)| Corner-sharing  | Cooling crystallization   | N/A      | N/A       | N/A     |
| 4-0D[24]  | \((C_3NH_2H(NH_3)_2Pb_4\) | N/A             | Antisolvent method       | 460      | 66        | 40      |
| 5-0D[35]  | \((C_2H_7NH_3)_2SnCl_6\) | N/A             | Solution method          | 450      | 125       | 8.1     |
| 6-0D[22]  | \((C_3N_3H_11)_2Pb_4\)   | N/A             | Antisolvent method       | 568      | 200       | 9.6     |
| 7-1D[26]  | \((C_9NH_20)_2SbCl_5\)   | N/A             | Antisolvent method       | 590      | 119       | 98      |
| 8-1D[27]  | \([Nmethyldabconium]PbI_3\) | N/A             | Antisolvent method       | 470      | 84        | 83      |
| 9-1D[37]  | \((C_4H_9NH_3)_2PbBr_10\) | N/A             | Antisolvent method       | 580      | 158       | 7       |
| 10-1D[39] | \((C_5H_9NH_3)_2PbBr_7\) | N/A             | Antisolvent method       | 469      | N/A       | N/A     |
| 11-1D[45] | \((C_4H_9NH_3)_2PbCl_6\) | N/A             | Antisolvent method       | 412      | 617       | N/A     |
| 12-1D[41] | \((C_6H_14N)_2PbBr_4\)   | N/A             | Antisolvent method       | 675      | N/A       | N/A     |

*The figures (1–12) highlighted in blue refer to the corresponding organic cations shown in Figure 4.*

### 2.2. Metal Ions

Halogenes have great influence on the structures of LDMHPs, too. For example, Dohner et al. reported 2D perovskites \( (EDBE)[PbX_4] \) \( (X = Cl^-, Br^-, I^-) \) with white emissions. Crystal structures of these materials are shown in Figure 7, in which the Pb-Cl is a 100-oriented structure. When the lead halide octahedron is deformed due to the different coordination environment of Pb\(^{2+}\), the 2D perovskites of Pb-Br and Pb-I became 110-oriented structures. Kanatzidis et al. reported 2D hybrid perovskites of \( EA_4PbX_{10} \) \( (X = Cl^-, Br^-) \) and when the halogen is I\(^-\), it did not form an analogue with \( EA_4PbX_{10} \) \( (X = Cl^-, Br^-) \), but a 1D face-sharing perovskite.

Halogenes affect structure distortions of LDMHPs significantly. As the ion radius of Cl\(^-\) (1.67 Å) is less than that of Br\(^-\) (1.82 Å), it does not accommodate the EA\(^+\) cation well and so the \( EA_4PbCl_{10} \) has highly distorted structure.[62] For \( EA_4PbBr_{10} \), the intermediate layer of Pb-Br is of regular structure, while the outer layer Pb-Br has obvious structural distortion (Figure 8a). The distortion of intrinsic structure affects the luminescence characteristics of LDMHPs. That the effect of halogen replacement on structure distortion and luminescence properties have many examples and broadband emission is strongly associated with distorted degrees of lattices in LDMHPs. Mao et al.[10] found that different distortion levels in \( EA_4PbCl_{10} \) (large distortion) versus \( EA_4PbBr_{10} \) (small distortion) and \( EA_4PbCl_{10} \) has a broadband white-light emission, while \( EA_4PbBr_{10} \) has a narrow blue emission. By further tuning the ratio of chlorine/bromine for \( EA_4PbBr_{10-x} \) \( (x = 0, 2, 4, 6, 8, 9.5, 10) \), they found that two of the
intermediate compounds \((x = 8 \text{ and } 9.5)\) have more optimized white-light emissions than that of pure \(\text{EA}_3\text{Pb}_3\text{Cl}_{10}\) (Figure 8b). Yangui et al.\(^{[63]}\) studied \((\text{C}_6\text{H}_{11}\text{NH}_3)_2[\text{PbBr}_4]\) with similar structures, in which the \([\text{PbI}_4]^{4-}\) is more regular than \([\text{PbBr}_4]^{4-}\) octahedron. Figure 8c shows the PL variation of \((\text{C}_6\text{H}_{11}\text{NH}_3)_2[\text{PbBr}_4 - x\text{I}_x]\) with mixed halogens. The full width at half-maximum (FWHM) of PL gradually widened with the decrease of \(\text{I}^-\) content due to the difference of structure distortions. Furthermore, halogens can also tune structure distortion through hydrogen bonding formed between organic cations and halide anions. Here, different electronegative properties of halogens lead to different strength of hydrogen bonding force (where \(\text{H-Cl} > \text{H-Br} > \text{H-I}\)), determining the orientation of organic cation\(^{[18]}\) in interlayer of metal halides and caused different distortion degrees of lead halide octahedron. Furthermore, the halogens can be replaced by other anions, such as the \(\text{SCN}^-\) which has a similar radius with \(\text{I}^-\). The 100-oriented 2D perovskite \((\text{C}_6\text{H}_{11}\text{NH}_3)_2\text{Pb(SCN)}_2\text{I}_2\)\(^{[64]}\) can be obtained by partially replacing the \(\text{I}^-\) with \(\text{SCN}^-\). Its structure is shown in Figure 8d, in which the lead coordination octahedron is constructed by four equatorial \(\text{I}^-\) and two axial \(\text{SCN}^-\) coordination. Due to the polarity of \(\text{SCN}^-\), the octahedron is distorted by \(\text{Pb-S}\) bonds\(^{[65]}\).

### 2.4. Hydrogen Bonding

The all-inorganic \(\text{Cs-M-X}\) structures \((\text{M} = \text{Pb}^{2+}, \text{Sn}^{4+}, \text{Ge}^{2+} \text{etc., } \text{X} = \text{Cl}^-, \text{Br}^-, \text{I}^-)\) are mainly constructed by electrostatic effects
between negatively charged metal halide octahedrons and positively charged Cs⁺. For organic–inorganic hybrid perovskites, organic cations contain one or more terminal amines in which H atoms interacting with halogens on inorganic octahedrons by hydrogen bonding. For example, the 110-oriented 2D perovskites have been widely studied recently due to their excellent white emission properties, and their unique corrugated structures are stabilized by hydrogen bonding. For example, Kanatzidis et al.⁴⁵ reported the corrugated 2D perovskite (DMEN)PbBr₄, as is shown in Figure 9a. Its 3 × 3 corrugated layer configuration is stabilized by hydrogen bonding. With H atoms on the primary and secondary amines of organic cations forming hydrogen bonding with Br atoms, the inorganic layer folds ≈90°, producing the strongly corrugated structure (Figure 9b). In addition, many other corrugated 2D structures, such as (N-MEDA)[PbBr₄]⁴⁻ which is shown in Figure 9c have been reported. It exhibits a white emission upon UV light with a high PLQE of 9% (Figure 9d). (Epz)PbBr₄ (Figure 9e)⁶⁶ is another example to demonstrate the role of hydrogen bonding in stabilizing the 110-oriented 2D perovskites (Figure 9f).

In fact, the formation of hydrogen bonding plays a decisive role in regulating the orientation and conformation of inorganic skeleton. Different amines, such as primary amines, secondary amines, tertiary amines all can construct metal halides 1D structures with different interlinkage modes (Figure 10a).⁴³ and the organic cations 1, 2, and 3 containing different N-H sites which are adjacent, interphase and pair sites constructed 2D, 1D, and 0D configurations, respectively (Figure 10b).⁶⁷ The structures of (GUA)₂PtI₆ and (FA)₂PtI₆⁶⁸ are shown in Figure 10c, and here the discrete [PtI₆]⁴⁻ octahedrons are connected by 3D hydrogen...
bonding network constructed by FA$^{2+}$ and the 2D network constructed by GUA$^{2+}$ cations, indicating that the hydrogen bonding force of organic cations affect the arrangement of inorganic [PtI$_6$]$^{4-}$ octahedrons. Recently, Cui et al. reported the structure transition from the corrugated 1D structure to 0D by adjusting hydrogen bonding (Figure 10d,e). By employing a unique urea-amide cation containing H$_2$N─C═O, which can form multiple hydrogen bonds between adjacent organic cations and inorganic skeleton. The corrugated 1D structure was stabilized by the hydrogen bonding of Pb─Br···H─N. When increase the concentration of amides in the growth process of crystals, the amides will form hydrogen bonds with H$_2$O in precursor solution and adjacent cations to construct a large network (Figure 10f), which completely separated the [PbBr$_4$]$^{2-}$ octahedrons to form a 0D perovskite. The 1D and 0D perovskites both exhibited stable white emission, and the dimension reduction from 1D to 0D increased the PLQE as much as five times, providing a new strategy to regulate the structure and improve the luminescence performance by regulating hydrogen bonding forces in MHPs.$^{[12]}$

2.5. Stoichiometric Ratio of Precursor Solution

For all-inorganic Cs-Pb-X perovskite crystals, different structure dimensions can be easily regulated by changing the stoichiometric ratio of precursor solution. For example, a ternary phase diagram of 1D Cs$_5$PbBr$_{14}$, 2D Cs$_3$Pb$_2$Br$_7$, and 3D Cs$_3$PbBr$_5$ is shown in Figure 11a, in which dimension regulation is achieved by stoichiometric ratio (CsBr:PbBr$_2$) control. As shown in Figure 11b, the traditional 3D structure of Cs$_3$PbBr$_5$ has two phases, cubic phase (Pm3m) and orthorhombic phase (Pnma). When PbBr$_2$ is abundant, 2D structure Cs$_3$Pb$_2$Br$_7$ is formed,$^{[69]}$ and Cs$^+$ ions appear between the inorganic layers. In contrast, when CsBr is excess, 0D structure Cs$_5$PbBr$_{14}$ is formed, of which individual lead halide octahedrons are completely separated by Cs$^+$.$^{[70]}$ The bandgaps of these Cs-Pb-Br perovskites increase with the dimension decreasing (Figure 11c). Both Cs$_3$PbBr$_5$ and Cs$_5$PbBr$_{14}$ showed direct bandgap, while Cs$_3$Pb$_2$Br$_7$ showed indirect bandgap. The Cs-Pb-I perovskites can also construct different dimensions by controlling the stoichiometric ratio of CsI versus PbI$_2$.

For organic–inorganic hybrid perovskites, a kind of organic cation usually constructed a kind of structure even the stoichiometric ratio of every element in the precursor solution changed. However, when cations form different valence states in crystals, multiple dimensional structures, such as the 2D µ-IPA$_3$Sn$_2$I$_7$ and 1D θ-IPA$_3$SnI as shown in Figure 12a can be built.$^{[71]}$ On the other hand, different single crystal structures can also be obtained when H$_2$O molecules grow into unit cells. For example, Cui et al. synthesized the 1D (C$_3$N$_3$H$_9$O)$_2$(C$_3$N$_3$H$_{11}$O)$_2$Pb$_2$Br$_9$ with the ratio of C$_3$N$_3$H$_9$O:2HBr : PbO = 3 : 2, and when the ratio increased to 2 : 1, H$_2$O entered into the unit cell to form a 0D [(C$_3$N$_3$H$_{11}$O)$_2$Pb$_2$Br$_9$-4H$_2$O.$^{[12]}$ Recently, Thomas T. M. et al. reported the well-known 100-oriented 2D (C$_6$H$_5$CH$_2$NH$_3$)$_2$PbI$_4$ phase with a corner-sharing structure, and a new compound 1D edge-sharing (C$_6$H$_5$CH$_2$NH$_3$)$_4$Pb$_5$I$_{14}$·2H$_2$O was generated when H$_2$O incorporated into the crystal structure (Figure 12b).$^{[72]}$ Furthermore, H$_2$O molecules improved LDMHPs’ luminescent properties. Lin et al. has synthesized two new 0D [(C$_6$N$_2$H$_{16}$)SbCl$_5$ and (C$_6$N$_2$H$_{16}$)SbCl$_5$·H$_2$O, in which H$_2$O molecules grew into
lattices of \((C_6N_2H_{16})SbCl_5\cdot H_2O\) leading to the larger spatial distance between the \([SbCl_5]\) dimers (Figure 12c) and improving the PLQE of 57%.\(^{[71]}\) In addition, dimethylsulfoxide (DMSO) molecules also can be introduced to crystals to build different dimensional structures, such as the 0D \((PDI)_2PbI_2\) and 1D \((PDI)_2PbI_2\cdot 2DMSO\) (Figure 12d).\(^{[74]}\)

### 2.6. Temperature Effect

Perovskite materials usually undergo phase transition when the temperature changes, which are mainly caused by changes in configurations of organic cations. For example, the organic cations were disorderly arranged as the temperature increased (> 300 K) and the disordered point was called the melting temperature.\(^{[75]}\) The disordered cations led to sudden change of 2D interlayer distance and lattice parameters along the direction of organic cations.\(^{[76]}\) Recently, Zeb et al.\(^{[77]}\) synthesized a 1D \((C_6H_{13}NH_3)_2PbI_4\) as shown in Figure 13a. With the temperature decreasing, \((C_6H_{13}NH_3)_2PbI_4\) underwent three structure phase transitions at 320, 298, and 262 K, respectively. The second phase transition at 298 K was related to the rearrangement of \((C_6H_{13}NH_3)_2PbI_4^+\) and this phase transition caused a significant change in the dielectric constant. As shown in Figure 13d, the interlayer spacing between the inorganic layers increased near the \(T_c = 202 \text{ K}\) due to the ordered-disordered transition of 1-methylpiperidinium cation (Figure 13a). In addition, Liao’s group\(^{[78]}\) synthesized a 1D \((C_6H_{13}NH_3)_2PbI_4\cdot CdCl_4\) with high temperature dielectric response, due to it undergoes the phase transition at 367 K caused by the change of relative position of Cd atoms (Figure 13b).

Temperature not only affects LDMHPs’ structures, but also causes difference in their photoelectric properties. For example, Eric et al. studied the exciton characteristics of 2D \((C_4H_9NH_3)_2PbI_4\) in a temperature range of 10–300 K.\(^{[79]}\) Under the drive of \((C_4H_9NH_3)_2PbI_4^+\) rearrangement, phase transition occurred at around 250 K, corresponding to the decrease of lattice spacing and the increase of exciton binding energy. In addition, Sieradzki et al.\(^{[14]}\) synthesized a 2D \((MHy)_2PbI_4\) as shown in Figure 13c. With the temperature decreasing, \((MHy)_2PbI_4\) underwent three structure phase transitions at 320, 298, and 262 K, respectively. The second phase transition at 298 K was related to the rearrangement of \((MHy)_2PbI_4^+\), and this phase transition caused a significant change in the dielectric constant. As shown in Figure 13d, the interlayer spacing between the inorganic layers increased.
Figure 11. a) The ternary phase of Cs-Pb-Br, proportion of precursor solution (CsBr and PbBr₂). b) Conversion between (CsPbBr₃, CsPb₂Br, and Cs₄PbBr₆). c) Electron band structure of CsPbBr₃, CsPb₂Br, and Cs₄PbBr₆. Reproduced with permission.[11] Copyright 2018, American Chemical Society.

with the temperature increasing. In addition, the emission of MHy₂PbI₄ exhibited blueshift during cooling temperature from 300 to 80 K. It exhibited light yellow emission at 300 K and turned to yellow-green emission at 80 K (Figure 13e). The few-layer exfoliated 2D (C₄H₉NH₃)₂PbI₄ underwent phase transition at ≈175 K, producing a new blueshift PL peak (Figure 14a).[80] It is worth mentioning that temperature has a great influence on the emission from relaxed states of LDMHPs. These materials usually have emission from relaxed states caused by lattice distortion or self-trapped excitons (STEs). The PL intensity of relaxed states is more efficient than the free exciton (FE) at low temperatures.

Recently, RPP perovskite (CH₃(CH₂)₃NH₃)₂(CH₃NH₃)ₙ⁻¹PbnI₃⁺¹ (n = 1–4) has been reported,[81] and it undergoes structure distortion under laser irradiation, resulting in a relaxed redshifted emission (Figure 14b). The relaxation peak increased sharply with the temperature decreasing (Figure 14c). In addition, the STE emission intensity also increased as the temperature decreased. Some 2D 100-oriented perovskites can observe STE emissions at low temperature (Figure 14d).[82,48] When temperatures are high, the STEs are easy to detraps from STE states to FE states, and at low temperatures, the detrapping process is suppressed.[82] Therefore, STEs emission gradually appears and increases at lower temperature.

2.7. Pressure Effect

Exerting high pressure to the material is an effective way to regulate structures and photoelectric properties of LDMHPs. For example, Liu et al.[83] reported that 2D (BA)₄AgBiBr₈ under high pressure. As shown in Figure 15d, under high pressure, the bandgap of Cs₃Bi₂I₉ continued to become narrow, and finally reached the optimal value. In addition, at atmospheric pressure, the isolated [Bi₂I₉]⁻ octahedron exhibited only weak emission upon excitation. But it exhibited significant enhancement of PL intensity about ten times under relatively high pressure (< 1 GPa) due to the increased exciton binding energy.

Shi’s team[13] synthesized a unique 1D organic tin bromide perovskite C₄N₂H₁₄SnBr₄. The 1D inorganic chain deformed under the transition from monoclinic phase to triclinic phase (Figure 15b), and the exciton binding energy increased, resulting in the enhancement of PL intensity (Figure 15c). This work revealed a potential application of supercharging to improve the luminescence performance of LDMHPs. Zhang et al.[85] studied the PL properties of 0D Cs₃Bi₂I₉ under high pressure. As shown in Figure 15d, under high pressure, the bandgap of Cs₃Bi₂I₉ continued to become narrow, and finally reached the optimal value. In addition, at atmospheric pressure, the isolated [Bi₂I₉]⁻ octahedron exhibited only weak emission upon excitation. But it exhibited significant enhancement of PL intensity about ten times under relatively high pressure (< 1 GPa) due to the increased exciton binding energy.

It is worth mentioning that external pressure could distort the structure of the LDMHP to produce its STE emission. Recently, it has been reported that the pressure effect on regulating the structure distortion achieving the STE emission of (BA)₄AgBiBr₈.[86] With the increasing pressure, the emission of (BA)₄AgBiBr₈ changed from a nonluminescent state under atmospheric pressure to a state with obvious fluorescence at 2.5–25.0 GPa (Figure 16a). The sample completely transformed to a
new structure at 2.1 GPa (Figure 16b). In addition, the new structure gradually distorted and its crystallinity reduced upon compression. Finally, a completely lower crystallinity phase formed at around 25.0 GPa, which led to the quenching of the luminescence. The luminescence mechanism is shown in Figure 16c. Under atmospheric conditions, the STEs were easy to detrap and return to FE states. When the pressure increases, the structure distortion of (BA)₄AgBiBr₈ deepens its self-trapped states, which enhanced the energy barrier to avoid the detrap of excitons. And the bright emission from self-trapping realized.

3. Luminescence Applications

3.1. Light-Emitting Diodes (LEDs)

Due to the size effect and significant differences in dielectric constants between the organic and inorganic parts, excitons in LDMHPs are spatially confined. This suppresses the separation of excitons and greatly improves the radiation recombination efficiency. Therefore, organic–inorganic LDMHPs are considered to be good candidate materials for stable LEDs with high-efficiency. Most 2D perovskite electroluminescent layers can be fabricated via the one or two-step fast crystallization spin-coating method using precursors. Table 2 shows a performance summary of LEDs based on recent 2D MHPs. However, 1D and 0D fail to be fabricated by solution method, which is the main reason for no 1D and 0D electroluminescent diodes. Therefore, unlike “structure-level” 1D nanowire/nanorod perovskites and 0D nanocrystal perovskites, 1D and 0D organic–inorganic MHPs at the molecular level have not been used in the electroluminescent layer.⁹⁷

In order to prepare color-adjustable LEDs, two main strategies have been tried so far. One is to rely on mixed halides,⁹⁸ and the other is to control quantum well structures.⁹⁹ However, ion migration and phase separation are prone to occur under light and electric fields in LED devices with mixed halide perovskites.¹⁰⁰ resulting in the changes in electroluminescent color during device operation.¹⁰¹ Controlling the quantum well structure to obtain quasi-2D perovskite is another effective method to obtain the target color. The LEDs produced by this method have a very stable
Figure 13. a) Phase transition and dielectric response of 1D MPIP perovskite structural phase transition induced by temperature. Reproduced with permission.[77] Copyright 2018, Royal Society of Chemistry. b) Structural phase transition of [C₆H₁₁NH₃]₂CdCl₄ at 367 K. Reproduced with permission. [78] Copyright 2014, American Chemical Society. c) MH₃PbI₄ underwent three structural phase transitions and the tilt degree of octahedral PbI₆[4]− at each phase.[14] d) The interval between lead iodide sheets varies with temperature.[14] Copyright 2019, American Chemical Society. e) CIE coordinates of MH₃PbI₄ at different temperatures. Reproduced with permission.[14] Copyright 2019, American Chemical Society.

Figure 14. a) Changes of PL spectrum of stripped monolayer (C₄H₉NH₃)₂PbI₄ with temperature. Reproduced with permission.[80] Copyright 2018, American Chemical Society. b) 2D perovskite (n = 2) under laser irradiation, PL redshift, and then blueshift reversible process after laser annealing. Reproduced with permission.[81] Copyright 2018, Springer Nature. c) Temperature-dependent changes in PL intensity, highlighting changes in the relative intensity of relaxed peak. Reproduced with permission.[81] Copyright 2018, Springer Nature. d) The broadband emission appears at low temperature in 100-oriented 2D perovskite. Reproduced with permission.[81] Copyright 2017, Royal Society of Chemistry.
Figure 15. a) Schematic diagram of Pb—I—Pb bond under compression in (BA)$_2$PbI$_4$. Reproduced with permission.\cite{86} Copyright 2018, American Chemical Society. b) 1D structure distortion caused by pressure. c) The relationship between PL spectra of C$_4$N$_2$H$_4$SnBr$_4$ and pressure, and luminescence photographs at different pressure points. b,c) Reproduced with permission.\cite{13} Copyright 2019, American Chemical Society. d) Optical absorption spectrum of Cs$_3$Bi$_2$I$_9$ at high pressure. Reproduced with permission.\cite{87} Copyright 2018, Wiley-VCH.

Figure 16. a) The photomicrograph shows that the strength of PL varies with the increase of pressure. b) The compressive structural transformation of (BA)$_4$AgBiBr$_8$ crystals (the pink and green octahedrons represent [AgBr$_6$] and [BiBr$_6$] octahedrons, respectively). c) Schematic diagram of the pressure-induced luminescence mechanism of self-trapped exciton. Reprinted with permission.\cite{86} Copyright 2019, Wiley-VCH.
electroluminescence spectrum. For example, Xing et al.\[92]\ introduced a short ligand (iso-pro-pyrammonium, IPA) to partially replace the long ligand (PEA) to synthesize a quasi-2D perovskites. The growth of n = 2, 3, 4 phases become dominant, thereby adjusting the crystallization of the quasi-2D perovskites. The fabricated LEDs devices reached a maximum brightness of 2480 cd m\(^{-2}\) at 490 nm and showed stable sky blue emission under high operating voltage (Figure 17a). In addition, Tian et al.\[93]\ prepared a quasi-2D perovskites/poly(ethylene oxide) (PEO) composite films by blending metal halide perovskites with polymer, and used it as a light-emitting layer to prepare a good spectral stability bright red perovskite LEDs (Figure 17b).

However, the insulating properties of the large cations limit the carrier transport in 2D perovskites, thereby reducing the radiation recombination rate of quasi-2D perovskite based LEDs.\[94]\ It has been reported that inorganic alkali metals with small ionic radius such as Na\(^+\) was used to replace large organic cations partly in perovskite layers to improve the carrier transport.\[95]\ Another way is to ensure the good connection of the inorganic layer through the optimized nanocrystalline pinning (NCP) process, which can effectively promote the carrier transfer in the LED devices of quasi-2D perovskites. For example, Lee et al.\[96]\ used the optimized NCP process to modulate the Ruddelsden–Popper (RP) phase of (PEA)\(_2(CH_3NH_3)_{n-1}PbBr_{3n+1}\) nanostructure, and their RP phase units oriented randomly. This modulated nanostructure can increase the contact between adjacent quasi-2D perovskite layers, and so carriers can be effectively transported through the connection interface between the inorganic layers, greatly improving the efficiency of RP quasi-2D perovskite LEDs. When n = 3, they developed RP quasi-2D perovskite LED devices with a maximum current efficiency of 20.18 cd A\(^{-1}\) (Figure 17c) and a maximum EQE of 4.98% (Figure 17d).

In addition, the conversion of 3D perovskites to quasi-2D perovskites will introduce more defects on the surface or grain boundaries due to the reduction of crystal size, resulting in non-radiative recombination rate, which will also reduce the electroluminescence efficiency 2D perovskite LEDs.\[97]\ The intrinsic defects in the perovskite films can be passivated by introducing additives, or the interface modification methods can effectively reduce the defects in the perovskite films, thereby improving the radiation recombination rate in 2D perovskite LEDs.\[98]\ For example, Zhao et al.\[1\] reported quasi-2D and 3D (2D/3D) perovskite-polymer (poly (2-hydroxyethyl methacrylate)) heterostructure LEDs. The introduction of the polymer effectively eliminated the nonradiative recombination pathway, and the external quantum efficiency of the LED devices was as high as 20.1% (current density is 0.1–1 mA cm\(^{-2}\), Figure 17e). And Yang et al.\[99]\ investigated LEDs with high efficiency based on quasi-2D PEA\(_2(FAPbBr_3)_{n-3}PbBr_4\) (n = 3) as the emitting layer. External quantum efficiency (EQE) of the champion device with
trioctylphosphine oxide (TOPO) passivation layer is up to 14.36% (Figure 17f).

3.2. Phosphors in Solid Lighting

LDMHPs have larger exciton binding energies, which greatly increases the exciton recombination rate, resulting in LDMHPs with high photoluminescence quantum yield (PLQE). The Ma’s group\(^{[115]}\) has reported an efficient broadband yellow-emitting phosphors composed of 0D tin mixed halide perovskites \((\text{C}_4\text{H}_{14}\text{N}_2\text{Br})_4\text{SnBr}_x\text{I}_{6-x}\) \((x = 3)\). The phosphors had a full width half maximum of 126 nm (FWHM) and a photoluminescence quantum efficiency (PLQE) of about 85% due to the structural reorganization of the excited state. By mixing the yellow phosphors and commercial Eu-doped barium magnesium aluminate blue phosphors \((\text{BaMgAl}_{10}\text{O}_{17}:\text{Eu}^{2+})\) with the weight ratio 1:4, a near-perfect white emission with CIE coordinates of \((0.32, 0.32)\), the color rendering index (CRI) of 84 and the correlated color temperature (CCT) value of 6160 K can be fabricated (Figure 18a). UV-pumped white LEDs was as high as 85. Zhong’s group\(^{[116]}\) used the HBr-assisted slow cooling method (SCM) to grow centimeter-sized \(\text{Cs}_x\text{PbBr}_y\) crystals with embedded \(\text{CsPbBr}_3\) nanocrystals, showing excellent green PL emission. Great green light-emitting performance up to 97% of PLQE and excellent thermal stability made the materials an attractive candidate for white light-emitting diodes (WLEDs) manufacturing. They also fabricated WLED devices by using green-emitting crystals \(\text{Cs}_x\text{PbBr}_y\) with \(\text{CsPbBr}_3\) nanocrystals, red-emitting phosphor \(\text{K}_2\text{SiF}_6:\text{Mn}^{4+}\) (KSF), and blue-emitting GaN chips. The optimized devices had a luminous efficiency of 151 lm W\(^{-1}\) at 20 mA and a chromaticity coordinate value of \((0.331, 0.331)\) (Figure 18b–d).

Actually, white-light-emitting from a single emitter layer is very important for solid-state lighting applications because it simplifies the device structure and avoids problems, such as self-absorption and color instability in hybrid emitters and multiple emitters.\(^{[46]}\) Recently, a series of LDMHPs with 2D, 1D, and 0D structures have been reported. There is a variety of low-dimensional single crystal growth strategies including but not limited to cooling crystallization method, antisolvent method and solution method. For 2D and 1D, adjacent octahedrons can be connected by corner-sharing, edge-sharing, and face-sharing. They all exhibit broadband white-light-emitting and are applied to a single white-emitting WLED phosphor. For example, The Ma’s group\(^{[19]}\) also reported an 1D organic halide perovskites \(\text{C}_6\text{N}_2\text{H}_{14}\text{PbBr}_4\) with the edge-sharing octahedral lead bromide chains \([\text{PbBr}_6]^{2-}\) that emitted bluish white-light with a PLQE
Figure 18. a) Emission stability and luminous performance of 0D (C₄N₂H₁₄Br)₄SnBr₆−ₓIₓ LED. Reproduced with permission. Copyright 2017, American Chemical Society. b) Schematic diagram of the configuration of the prototype device. c) A picture of surface mounted device (left) and the WLED operated at a forward bias current (right). d) EL spectrum of the prototype WLED device. b-d) Reproduced with permission. Copyright 2018, Wiley-VCH. e) CIE coordinates of (EDBE)(PbBr₄) and sunlight at noon. Inset is a photograph of (EDBE)(PbBr₄) under 365 nm irradiation. Reproduced with permission. Copyright 2014, American Chemical Society. f) CIE coordinates and emission spectrum of white-light emitters and sunlight at noon. Inset is a photograph of (N-MEDA)(PbBr₄) under 380 nm irradiation. Reproduced with permission. Copyright 2014, American Chemical Society. g) CIE coordinate of (EDBE)PbBr₄. Inset is an image of a UV-pumped WLED. Reproduced with permission. Copyright 2016, Wiley-VCH.

Table 3. Summary of reported LDMHPs phosphors used in white LED devices.

| Dimension | Formula | Connecting type | Fabrication | Peak [nm] | FWHM [nm] | PLQE [%] |
|-----------|---------|-----------------|-------------|-----------|-----------|----------|
| 21-110-2D⁴⁵ | (DMEN)PbBr₄ | Corner-sharing | Cooling crystallization | ≈520 | ≈180 | N/A |
| 22-110-2D⁴⁶ | (N-MEDA)(PbBrₓCl₁₋ₓ) | Corner-sharing | Solution method | ≈530 | 220 | <1.5 |
| 23-110-2D⁴⁷ | C₆H₁₃Cl₄N₃Pb | Corner-sharing | Solution evaporation | 573 | 220 | <1 |
| 25-110-2D²⁵ | (PDA)₂PbBr₂ | Corner-sharing | Antisolvent method | 671 | 265 | 6.7 |
| 27-110-2D¹² | (EDBE)(PbBr₄) | Corner-sharing | Solution method | 573 | 215 | 9 |
| 27-100-2D¹² | (EDBE)(PbCl₄) | Corner-sharing | Solution method | 538 | 208 | 2 |
| 20-100-2D¹⁰ | (CH₃NH₂)₂PbBr₆−ₓClₓ | Corner-sharing | Solution method | 359–450 | 150–228 | N/A |
| 24-100-2D⁴⁸ | (2meptH₂)PbBr₄ | Corner-sharing | Solution method | 417 | ≈50 | 3.37 |
| 18-1D¹² | (C₃N₃H₁₀O)(C₃N₃H₁₁O)₂Pb₂Br₉ | Corner-sharing | Antisolvent method | 530 | 218 | 1.7 |
| 25-1D²⁵ | (EDA)₂PbBr₆ | Corner-sharing | Antisolvent method | 523 | 138 | 9.1 |
| 17-0D¹² | (C₄N₂H₁₄I)₂SnI₆ | N/A | Antisolvent method | 620 | 188 | 75±4 |
| 17-0D¹² | (C₄N₂H₁₄Br)₂SnBr₆ | N/A | Antisolvent method | 570 | 103 | 95±5 |
| 6-0D¹² | (C₄N₂H₁₁O)₂PbBr₂·4H₂O | N/A | Antisolvent method | 568 | 218 | 9.6 |

⁴⁵ The figures highlighted in blue refer to the corresponding organic cations shown in Figure 4.
of 20%. To utilize the phosphor-converted WLEDs for artificial lighting requirement, the CRI points of 95,[117] the CCT of 5564 K,[118] and the PLQE of 90%[119] should be the standards. Table 3 gives a summary of LDMHPs phosphors used in WLED devices. Karunadasa and his colleagues[17,46] published 2D hybrid perovskites with 110-oriented broadband white-light-emitting, namely (EDBE)PbBr$_4$ (Figure 18e) and (N-MEDA)PbBr$_4$ (Figure 18f), which had broadband white-light-emitting due to the formation of self-trapped excitons in the deformable lattice. Recently, Ma’s group[120] reported a high-luminance micrometer-scale corrugated 2D lead bromide perovskite (EDBE)PbBr$_4$, its emission almost cover the entire visible spectrum, and its PLQE up to 18%. This perovskite phosphor can produce white-light-emitting under the irradiation of ultraviolet lamp. The CCT
pumped by the 365 nm UV-LED chip is 6519 K, and the CIE coordinates are (0.30, 0.42) (Figure 18g).

3.3. Scintillator and Transducer

It is generally believed that LDMHPs have higher PCEs, faster decay and larger stokes shifts compared with 3D ones. These advantages demonstrate the potential of LDMHPs for large-area and low-cost scintillator devices in the field of medical imaging, nondestructive detection, space exploration, etc. For example, Birowosuto et al. reported three X-ray scintillator characteristics of 3D MAPbI₃ and MAPbBr₃ and 2D (EDBE)PbCl₄ hybrid perovskite crystals. Comparing to 3D perovskites, 2D (EDBE)PbCl₄ has less thermal quenching due to the large exciton binding energy so that moderate light yield of 9000 photons MeV⁻¹ can be obtained even at room temperature (Figure 19a). Moreover, Masanori et al. researched 2D organic–inorganic hybrid perovskite-type scintillator single crystal of 5 × 6 × 1 mm³ of (PEA)₂PbBr₄ (PEA⁺ = C₆H₅(CH₂)₂NH₃⁺). Under 662 keV gamma-ray excitation, the crystal shows a substantial light yield of 10 000 photons MeV⁻¹, with a main decay time component of 9.4 ns. Subsequently, they further optimized the crystal and investigated scintillation properties of (PEA)₂PbBr₄ under gamma-ray and X-ray irradiations. They fabricated 17 × 23 × 4 mm³ bulk sample, which shows a remarkably high light yield of 14 000 photons MeV⁻¹ and very fast decay time of 11 ns under gamma-rays.

In addition, Xie et al. demonstrated Li-dopant (PEA)₂PbBr₄ crystal and explored the application of Li-(PEA)₂PbBr₄ scintillator in alpha particle detection, X-ray imaging and discrimination between alpha particle and gamma-ray. It is worth mentioning that they obtained first satisfactory X-ray imaging pictures of a ubiquitous safety pin using Li-(PEA)₂PbBr₄ film (Figure 19b,c). However, the toxicity of lead in these lead-based halide perovskites may restrict its potential commercial applications. Thereby, Cao and his colleagues developed a lead-free 2D layered (C₈H₁₇NH₃)₂SnBr₄ perovskite scintillators, which not only exhibit a high PCE of 98% and a large Stokes shift of 246 nm but also provide nontoxicity, good PL intensity, and stability under X-ray illumination. The results make the novel perovskite scintillators suitable for X-ray imaging applications (Figure 19d). And Hardhienata et al. studied the optical and scintillation properties of manganese-based 2D organic–inorganic hybrid perovskite crystals X₂MnCl₄ (X = PEA, PPA). Both of them show promising PL properties, reasonable decay time (about 3–4 µs) and small bandgaps (about 2 eV), making them applicable for scintillator in X-ray imaging application. Viktoria et al. reported isostructural 0D halide complexes of Bmpip₂SnBr₄ and Bmpip₂PbBr₄ (Bmpip = 1-buty1-1-methylpyperidinium cation) that can exhibit potent X-ray fluorophores being comparable to that of a commercial inorganic NaI:Tl X-ray scintillator (Figure 19e,f).

At the same time, LDMHPs have also shown potential in transducer field due to their adjustable structural phase transition and fluorescence emissions. For example, Zhou and his colleagues reported a novel 0D all-inorganic perovskite single crystal Cs₂InBr₃·H₂O for sensitive water detection. And Li et al. have carefully researched the temperature-dependent phase transition and photoluminescence properties of 0D organic–inorganic hybrid metal halide (Cs₉NH₂O)₂MnBr₄. Based on this compound, they further developed a fluorescent sensor for acetone. By simplified gas-sensitive detecting device with (Cs₉NH₂O)₂MnBr₄ rapid fluorescence quenching shows when the organic solvent is acetone, which cannot be found when applying other organic solvents (Figure 19g,h).

4. Prospects and Outlook

As a new perovskite-type photovoltaic material system, LDMHPs exhibit excellent stability to water, heat and light, and are expected to solve the instability problem of traditional 3D perovskites, which plays a crucial role for the final industrialization of perovskite materials. By adjusting the dimensions and crystal structures, LDMHPs are expected to provide more opportunity for various optoelectronic devices. However, the LDMHP are still a new type material which has not been fully developed yet, and there are still some problems to be solved:

a) How to decrease environmental pollution. Although the stability of organic–inorganic hybrid perovskites has been greatly improved, the potential toxicity problem of Pb²⁺ has not been solved. At present, some people try to use other metal elements to replace or partially replace Pb to prepare lead-free or lead-less perovskite solar cells, however, the PCE is not ideal. Public health concerns about lead toxicity will promote the development of lead-free LDMHPs.

b) Currently, LDMHPs with inherent white emission have many attractive advantages, but their PLQEs are still too low when used in WLEDs. Moreover, there is a lack of systematic researches and understanding of the design, synthesis, and luminescence process mechanism of these LDMHPs. In addition, 1D and 0D perovskite materials still need a lot of work to explore especially for the application to LED devices.

c) Although more and more LDMHPs-based optoelectronic devices show multifunctional performance and long-term stability, the properties of intrinsic materials and the composite dynamic process need further research. At the same time, an in-depth understanding of the structure-performance relationship is very necessary for the continued development of LDMHPs.

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

Y.H. and S.Y. contributed equally to this work. All the authors contributed to the discussion of the content, writing and editing of manuscript prior to submission.
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