Metal halide perovskite materials hold great potential as a core element in full-color display and lighting applications due to their unique optoelectronic properties, such as high photoluminescence efficiency, good color purity, tunable bandgap, and low-temperature solution processability. However, the performance of blue perovskite light-emitting diodes (PeLEDs) lags far behind their red and green counterparts, impeding their practical use. Herein, the recent progress on blue PeLEDs based on different restrictions and some feasible strategies to improve luminescence efficiency and stability are summarized. The methods to optimize blue-emission perovskite materials with different dimensions are addressed. The major factors affecting luminescence stability, the approaches to balance charge injection and minimize the optical loss are also discussed. Several problems in blue PeLEDs, particularly the toxicity of lead-based perovskites and the alternative solutions of lead-free perovskites, are emphasized. Finally, the perspective of future development in blue PeLEDs is provided.

1. Introduction

In recent years, metal-halide perovskite materials have been regarded as the promising candidate for the use in light-emitting diodes (LEDs) due to their unique characteristics such as high photoluminescence quantum yield (PLQY), excellent color purity, tunable bandgap, and simple solution processability. According to the dimensions of perovskite materials, they can be categorized as 0D, 1D, 2D, and 3D, among which quasi-2D perovskites are most widely used at present. For the potential applications in full-color displays and lightings, perovskite LEDs (PeLEDs) not only overcome the shortcomings of inorganic LEDs that cannot be used for flexible displays but also show excellent color purity as compared with that of organic LEDs. Since the first report of red and green PeLEDs at room temperature in 2014 by Tan et al., rapid progress in external quantum efficiencies (EQEs) has been made. PeLEDs with green, red and near-infrared emissions have gained tremendous developments with EQEs exceeding 20%. However, the performance of blue PeLEDs still lags far behind the other counterparts in the aspects of electroluminescent (EL) efficiency and spectral stability. Moreover, blue PeLEDs suffer from the poor lifetime as compared with that of traditional inorganic, organic, and quantum dot LEDs. Therefore, the effective schemes toward high efficiency and long lifetime of blue PeLEDs are highly desirable to promote their commercialization in the new-generation full-color displays.

In this Review, we focus on the strategies that improve the EL efficiency and stability of blue PeLEDs. To be specific, the proposed strategies are summarized based on the problems in blue PeLEDs and the corresponding solutions (Figure 1). We first begin with a summary of the methods to optimize blue-emission perovskite materials based on their crystal dimension, including 3D, quasi-2D, and nanocrystals (NCs). Then, we present the factors that affect the spectral stability and luminance stability (or lifetime) based on different degradation mechanisms. The origins of unbalanced charge injection and optical loss are discussed, and the possible solutions to charge transport and light outcoupling are provided. The current progress made on eliminating the toxicity problem of Pb-based perovskites is also analyzed along with the alternative solution of Pb-free perovskites. Finally, the perspectives for future development in blue PeLEDs are provided.
Figure 1. Schematic illustration of the strategies to improve luminescence efficiency and stability of blue PeLED.
2. Status Quo of Blue Perovskite Materials

2.1. 3D/Quasi-2D Perovskites

Metal halide perovskites have a typical chemical formula of ABX₃, where A is a monovalent organic or inorganic cation (e.g., methylammonium [MA⁺], formamidinium [FA⁺], or Cs⁺), and B is a bivalent metal cation (e.g., Pb²⁺ or Sn²⁺), and X is a halide anion (Cl⁻, Br⁻, or I⁻). Usually, a mixture of different ions (e.g., mixed MA⁺, FA⁺, and Cs⁺, mixed Pb²⁺ and Sn²⁺, and mixed halide) is used to form each site in the perovskite crystals, leading to a large tunability of electronic and optical properties.[37] 3D perovskites possess superior advantages of long carrier diffusion length, excellent charge mobility, low trap states, and so on. Increasing the chlorine content in chloride/bromide (Cl/Br)-mixed perovskites can tune the emission wavelength toward blue-light region (Table 1). Since 2015, as shown in Table 1, the indium tin oxide (ITO) is usually used as the transparent anode, a layer of zinc oxide nanoparticles (ZnO-NPs) is used as an electron-transfer layer, and poly (2,3-dihydrothieno-1,4-dioxin)-poly styrenesulfonate (PEDOT:PSS), N,N'-bis-(4-butylphenyl)-N,N'-bis(phenyl)-benzil (TPD), and N,N-bis(1-naphthyl)-N,N-diphenylbenzidine (NPD), poly[bis(4-phenyl)-2,6-trimethylphenyl]amine (PTAA) are used as a hole-transfer layer, respectively. Kumawat et al. used the MAPbBr₂$_{0.8}$Cl$_{0.2}$-based 3D perovskite as an emitter to achieve the first blue PeLED with an EL peak (λ$_{EL}$) at 482 nm.[16] However, this device exhibits a low EQE of only 0.0003% due to inadequate film coverage and high defect density of the Cl/Bristored mixed perovskite. To solve the problem of low efficiency in blue PeLEDs, Kim et al. synthesized the Cs$_{10}$(MA$_{0.17}$FA$_{0.83}$)$_{100-x}$PbBr$_x$Cl$_{1.5}$ perovskite by doping Cs⁺ cations into the MA/FA-based perovskite, improving the surface coverage of traditional 3D perovskite films and reducing the surface defects.[38] A blue PeLED (λ$_{EL}$ = 475 nm) was obtained with a maximum luminance (L$_{max}$) of 3567 cd m⁻² and a maximum EQE (EQE$_{max}$) of 1.7%. However, the phase separation and ion migration behaviors were observed in this halogen-mixed 3D perovskite, impeding the luminescence stability. The luminance increased to 948.5 cd m⁻² during the first 25 min and then decreased rapidly to 88.5 cd m⁻² after 150 min operation. To improve the operational stability of blue PeLEDs, Yuan et al. developed a cocktail strategy that incorporated multiple cations Rb/FA/phenethylamine (PEA)/K into the inorganic CsPb(Cl/Br)$_x$ perovskite, and the synergetic effect of multiple cations on the ion-induced crystallization ameliorated the film quality with the improved radiative recombination and operational stability.[39] When further using an insulator/perovskite/insulator structure to reduce hole-caused leakage current and inhibit exciton quenching, blue PeLEDs (λ$_{EL}$ = 484 nm) achieved a L$_{max}$ of 4015 cd m⁻², an EQE$_{max}$ of 2.01%, and a half-lifetime (T$\frac{1}{2}$) of >300 min under continuous operation conditions. However, the organic components contained in mixed perovskites inevitably impose the intrinsic drawbacks in thermal stability of perovskite materials and spectral stability of blue-light emission. Du et al. successfully prepared all-inorganic CsPb(Br/Cl)$_x$ perovskite films with the emission wavelengths ranging from 450 to 480 nm using the dual-source coevaporation of CsCl and PbBr$_2$ (Figure 2a,b).[40] The vacuum deposition method can avoid the disadvantages of the limited solubility of cesium salt in the solution method, and the Cl/Br ratio can be continuously adjusted to precisely achieve the standard blue emission. Moreover, the thermally evaporated perovskite film exhibited good film morphology, and an EQE$_{max}$ of 0.38% was realized for deep blue PeLEDs (λ$_{EL}$ = 468 nm) (Figure 2c-e).

Despite the use of conventional 3D perovskites for blue emission, quasi-2D perovskites have attracted tremendous attention in recent years due to their high chemical and light stability as well as large exciton binding energy caused by quantum confinement for the promoted radiative recombination efficiency (Table 1).[41–45] By controlling the number (n) of corner-sharing octahedra (PbCl₆) layers, Yuan et al. reported multilayered quasi-2D PeLED operating at near-infrared wavelengths with an EQE$_{max}$ of 8.8%.[41] In 2018, Hu et al. fabricated an efficient deep-blue PeLED (λ$_{EL}$ = 468 nm) with a quasi-2D perovskite of (BA)₂(MA)$_{n-1}$PbBr$_n$Cl, yielding an EQE$_{max}$ of 0.01% and a L$_{max}$ of 21 cd m⁻².[42] The variation of the n number in quasi-2D perovskites can not only tune the energy transfer (ET) process, but also another process of charge transfer (CT). Shang et al. engineered the (PEA)$_2$(MA)$_{n-1}$PbI$_n$I$_{1-n}$ Ruddlesden-Popper perovskite structure, demonstrating the effective CT in low pinhole-density perovskite films.[43] Alternatively, Chen et al. successfully used PA$_2$(CsPbBr$_3$)$_{n-1}$PbBr$_2$ to fabricate blue-green PeLEDs (λ$_{EL}$ = 505 nm) with an EQE$_{max}$ of 3.6% and a L$_{max}$ to 7320 cd m⁻², which was attributed to the efficient CT by adopting an electron transport material of TmPyPB.[44] In 2020, Pang et al. rearranged the low-dimensional phase distribution by incorporating sodium ions (Na⁺) to partially substitute the large organic molecules in the Cl/Br-mixed quasi-2D perovskites (Figure 2f), which induced the suppression of the formation of n = 1 phase but the significant elevation of other small-n phases (i.e., n = 2, 3, and 4).[45] This rearrangement of phase distributions gave rise to effective radiative recombination and exciton ET in the quasi-2D perovskite films, achieving the sky-blue PeLED (λ$_{EL}$ = 488 nm) with an EQE$_{max}$ of 11.7% (Figure 2g).

Table 1. Summary of blue PeLEDs based on 3D and quasi-2D perovskites.

| Perovskite material | Device structure | EL [nm] | EQE$_{max}$ [%] | L$_{max}$ [cd m⁻²] | Ref. |
|---------------------|-----------------|---------|-----------------|-------------------|-----|
| MAPbBr$_{1.68}$Cl$_{0.32}$ | ITO/PEDOT:PSS/3D perovskite/PbM/Ag | 482 | 0.0003 | 1.72 | [16] |
| Cs$_{10}$(MA$_{0.17}$FA$_{0.83}$)$_{100-x}$PbBr$_x$Cl$_{1.5}$ | ITO/ZnO-NPs/3D perovskite/ε-NPD/MoO$_3$/Al | 475 | 1.7 | 3567 | [38] |
| (Cs/Rb/FA/PEA/K)Pb(Cl/Br)$_x$ | ITO/LiF/3D perovskite/LiF/Bphen/LiF/Al | 484 | 2.01 | 4015 | [39] |
| CsPbBr$_{3-x}$Cl$_x$ | ITO/NiO$_2$/3D perovskite/TPBi/LiF/Al | 468 | 0.38 | 121 | [40] |
| (BA)$_2$(MA)$_{n-1}$PbBr$_n$Cl | ITO/PEDOT:PSS/poly-TPD/quasi-2D/TPBi/LiF/Al | 468 | 0.01 | 21 | [42] |
| PA$_2$(CsPbBr)$_{n-1}$PbBr$_x$ | ITO/PEDOT:PSS/quasi-2D/TmPyPB/CsCO$_3$/Al | 505 | 3.6 | 7320 | [44] |
| – | ITO/NiO$_2$/PTAA/PVK/quasi-2D/TPBi/LiF/Al | 488 | 11.7 | – | [45] |
Quasi-2D PeLEDs usually exhibit multiple EL peaks and unstable colors due to the different ET efficiencies inside perovskite materials. To solve this problem, the anion mixing and quantum confinement engineering have been proposed. However, the EL spectra show the instability with the variation of the driving bias, which was caused by phase separation or the coexistence of multiple crystal domains. To further boost the spectral stability of quasi-2D perovskite-based blue PeLEDs, Xing et al. partially replaced the long ligand PEA with the short ligand IPA for reducing the van der Waals force between bulk organic cations and suppressing the formation of $n = 1$ phase, which was accompanied with the strengthened phase ($n = 2, 3, 4$) monodispersity. The optimized PeLED yielded a single emission peak ($\lambda_{EL} = 490$ nm), an average EQE of 1.0%, and a $L_{\max}$ of 2480 cd m$^{-2}$.

Figure 2. Fabrication of all-inorganic CsPbBr$_{x}$Cl$_{3-x}$ perovskite films. a) Schematic illustration of dual-source coevaporation of CsCl and PbBr$_2$. b) Spatially resolved PL peak and intensity of the coevaporated CsPbBr$_{x}$Cl$_{3-x}$ perovskite films. c) EQE versus current density for PeLEDs based on samples A, B, and C as indicated in (b). d) Thermal stability of all-inorganic and hybrid perovskites under the aging conditions of 100 °C continuous heating in N$_2$. e) Ultraviolet (UV) irradiation stability under 10 mW cm$^{-2}$, 365 nm ultraviolet lamp irradiation in N$_2$. a–e Reproduced with permission. Copyright 2019, American Chemical Society. f) Schematic illustration of the phase rearrangement by adding Na$^+$ in quasi-2D perovskites. g) EQE versus luminance of PeLEDs without and with 20% NaBr. f,g Reproduced with permission. Copyright 2020, American Chemical Society.
2.2. Perovskite NCs

Perovskite NCs have also attracted considerable research interests due to their unique properties such as low-temperature synthesis, controllable crystal size, and high dupability. An amount of colloidal synthesis strategies have been developed for perovskite NCs in recent years, including template-assisted synthesis, ligand-assisted precipitation (LARP), hot-injection method, anion exchange reaction, microwave-assisted synthesis, and so on. The performance of blue-emission perovskite NCs mentioned in this section are shown in Table 2.

2.2.1. Template-Assisted Synthesis

In 2014, Schmidt et al. used the template-assisted method to synthesize luminous 6 nm-sized MAPbBr₃ perovskite NCs by choosing thin Al₂O₃ film as a mesoporous medium, which could be maintained stable in the solid state with a high quantum yield of 20%. However, many toxic organic solvents were consumed in the process of synthetic and purification processes, delivering the risk to human health and environmental pollution. Correspondingly, Wang et al. used the in situ formed mesoporous Al₂O₃ as a template to prepare highly emissive CsPbBr₃ perovskite NCs through a solvent-free high-temperature solid-state template confined growth strategy (Figure 3a). The obtained CsPbBr₃/Al₂O₃ powders possessed the PLQY up to 70%, narrow emission peak with a full width at half maximum of 25 nm, and outstanding thermal stability. This versatile method can be used for the synthesis of a large amount of perovskite NCs, and holds great potential for other CsPbX₃ NCs by controlling the ratio of the halide ions.

2.2.2. Ligand-Assisted Precipitation

Due to the advantages of convenient operational temperatures, receptive equipment requirements, and ease of extensibility, the LARP method for synthesis of perovskite NCs is an affordable and potential pathway. It was found that perovskite NCs could exist stably in nonpolar organic solvents with the help of amine ions, whereas their optical properties were significantly influenced by several parameters, such as the ratios of precursor chemicals, temperature, capping agents, and solvent system of the precipitation process. On this basis, Zhang et al. successively proposed the LARP method which could synthesize bright MAPbX₃ NCs directly. However, the perovskite NCs were synthesized by dropping the clear perovskite solution containing co-ligands in the nonpolar aprotic solvents. The resulting solution still contained the solvents that could dissolve perovskites, such as N,N-dimethylformamide (DMF), dimethylsulfoxide (DMSO), and ligands. To overcome this problem, Park et al. developed a modified LARP method that used DMF as a good solvent and n-hexylamine (HxA) as a co-ligand to synthesize organic–inorganic hybrid perovskite NCs (Figure 3b). The remained DMF would dissolve the MAPbX₃ NC-solid-films if the amount of DMF could not reduce as much as possible. By controlling the ratio of DMF, the solid perovskite NC film were formed under the drying process.

2.2.3. Hot-Injection Method

Compared with other colloidal synthesis methods, the hot-injection method provides a versatile way to prepare high-luminescence colloidal perovskite NCs with tunable size, shape, and surface passivation. Proteescu et al. injected Cs-oleic acid quickly into a noncoordinating solvent containing lead halide and ligand under the protection of N₂ via the hot-injection method. By changing the halide composition, the particle size, bandgap energy, and emission spectra of these perovskite NCs can be adjusted in the entire visible spectrum of 400–780 nm, implying a good solution to realize blue-emission perovskite NCs.

2.2.4. Anion Exchange Reaction

Post-synthetic halide exchange reactions in perovskite NCs, including cation exchange and anion exchange reaction, were regarded as a promising tool to manipulate the composition of perovskite NCs. In recent years, plentiful attempts have been carried out to exchange Cs⁺ cations or Pb²⁺ cations in CsPbX₃ (X = Cl, Br, I) NCs, where the anion exchange reaction led to a blue shift for the routes of I⁻ → Br⁻ and Br⁻ → Cl⁻. Unfortunately, this method usually formed a new halide along with the decomposition of CsPbX₃ NCs. In 2015, Akkerman et al. tuned the optical properties of CsPbBr₃ NCs via anion exchange reaction method with the preservation of the pristine crystal structure. Similarly, Nedelcu et al. presented a

| Table 2. Summary of blue-emission perovskite NCs with various synthesis methods. |
|---|
| Synthesis method | Perovskite | NC size [nm] | PLQY [%] | Lifetime [ns] | FWHM [nm] | Ref. |
| Template-assisted synthesis | CH₃NH₃PbBr₃ | 6 | 20 | – | – | [47] |
| Template-assisted synthesis | CsPbBr₃ | 6 | 70 | 9.8 | 25 | [51] |
| LARP | CH₃NH₃PbX₃ | 3.3 | 20 | 6.6 | ≈20 | [52] |
| LARP | MAPbX₃ | 5 | 49 | 4.83 | ≈20 | [53] |
| Hot-injection method | CsPbX₃ | 4–15 | 50–90 | 1–29 | 50 | [54] |
| Anion exchange reaction | CsPbX₃ | 8.5 | 24 | – | 26 | [49] |
| Anion exchange reaction | CsPbX₃ | 4–15 | 20–80 | – | 10 | [50] |
| Anion exchange reaction | CsPbBrCl | 8 | 81 | – | 18 | [55] |
| Microwave-assisted synthesis | CsPbX₃ | 15.1–23.7 | 90 | 1.9–135.7 | 14 | [56] |
| Microwave-assisted synthesis | CsPbBr₂ | 15.58 | 92 | 27 | 26 | [57] |
one-step synthesis method of CsPbX$_3$ NCs to form monodisperse colloidal nanotubes via anion exchange reaction, demonstrating the spectrally narrow and bright photoluminescence (PL) over the whole visible region.

In 2020, Ye et al. used tetra-tert-butylammonium p-toluenesulfonate (TBSA) to trigger anion exchange reaction (Br$^-$/$\text{Cl}^-$) due to the anion and cation synergistic effects of benzenesulfonates, leading to a blue shift from 456 to 409 nm. The resulting mixed-halide CsPbBr$_x$Cl$_{3-x}$ blue PeLEDs achieved an EQE$_{\text{max}}$ of 2.6%.

2.2.5. Microwave-Assisted Synthesis

The microwave-assisted synthesis method can facilitate formation of the uniformly distributed cube-shaped CsPbX$_3$ NCs because of the homogeneous heating and rapid temperature growth, making it promising for achieving high-quality perovskite NCs. In 2017, Liu et al. prepared all-inorganic CsPbX$_3$ NCs with high PLQY over 90% by optimizing the microwave-assisted synthesis conditions, gaining blue PeLEDs with an EQE$_{\text{max}}$ of 0.09% and color coordinate of (0.15,0.26). In 2020, Thesika and Murugan used microwave-solvothermal synthesis method to improve the heterogeneous reaction at the interfaces for the formation of all-inorganic CsPbX$_3$ NCs with PLQY of 92% and narrow emission line widths. A unique feature of this method is that the synthesis time is dramatically reduced within 6 min as compared with traditional hot-injection method that requires more than 3 h.

3. Challenges and Strategies for Blue PeLEDs

Despite rapid progress made in device efficiency, blue PeLEDs are still suffering from the inferior performance, including undesirable stability of color and luminance, imbalanced charge injection, inevitable optical loss, and toxicity. To date, the development of efficient and stable blue PeLEDs is urgently desired to be given high priority. The performances and device architectures of representative blue PeLEDs mentioned in this section are shown in Table 3.

3.1. Structural Modification for Efficient Emission

To solve the bottleneck of the inferior charge transport, Wang et al. recently introduced PEACL (C$_6$H$_2$C$_2$H$_4$NH$_3$Cl) and 2% YCl$_3$ into 3D CsPbBr$_3$ perovskite films (Figure 4a,b) to improve the PLQY of the layered perovskite from 1.1% to 49.7% and to enhance the operational stability of perovskites on account of the suppression of ion migration. A sky-blue PeLED ($\lambda_{EL} = 485$ nm) with an EQE$_{\text{max}}$ of 11.0% and a blue PeLED ($\lambda_{EL} = 477$ nm) with an EQE$_{\text{max}}$ of 4.8% were achieved by changing the content of YCl$_3$ in the perovskite films (Figure 4c). Moreover, the sky-blue PeLEDs with the incorporation of YCl$_3$ exhibited the highly enhanced spectrum and luminance stabilities compared with 3D perovskite materials, which were ascribed to the dramatically suppressed ion migration and phase segregation in these mixed-halide perovskite films under electric field. Soon
afterward, Chu et al. refreshed the EQE values of sky-blue PeLEDs by importing a large cation of CH$_3$CH$_2$NH$_2^+$ (EA') to PEAO$_2$(CsPbBr$_3$)$_2$PbBr$_4$ perovskite (Figure 4d), which inhibited the orbit coupling of Pb—Br and enlarged the optical bandgap for the emission changing from green (508 nm) into blue (466 nm). By optimizing the replacement of Cs$^+$ cations with larger EA$^+$ cations, a sky-blue PeLED ($\lambda_{\text{EL}} = 488$ nm) with a high EQE$_{\text{max}}$ of 12.1% was obtained, exhibiting excellent stability under light soaking and heating (Figure 4e).

Although remarkable development has been gained for sky-blue PeLEDs with EQE over 10%, efficient and stable deep-blue devices are rarely reported. A common way to fabricate deep-blue PeLEDs is to reduce dimensional domain by adding larger organic ammonium (spacer). Yuan et al. used P-PDAPbBr$_4$ to ameliorate the poor coordination between traditional spacer (PEABr) and inorganic octahedron, leading to the optimized pure-bromide PeLED with a deep-blue emission ($\lambda_{\text{EL}} = 465$ nm) and an EQE$_{\text{max}}$ of 2.6%. However, the introduction of larger organic ammonium would be adverse to the electrical conductivity of perovskite films. To avoid the influence of the incorporated organic additives on the transport properties of perovskite films, Shen et al. modified the substrate with potassium (K') cations to improve radiative recombination and hole-transport capabilities, arising from strong dipole interaction between K$^+$ and halide ions.

The K$^+$-triggered nucleation and grain growth promoted the formation of well-packed (Cs/FA/p-F-PEA) PbCl/Br$_3$ perovskite film with a PLQY of 34.8%, and thereby a deep-blue PeLED ($\lambda_{\text{EL}} = 469$ nm) was obtained with an EQE$_{\text{max}}$ of 4.14%. Yan et al. synthesized a stable pure layered (benzimidazole)PbBr$_4$ (BI/PbBr$_4$) perovskite via the strong π–π interaction between benzene rings of the BI ligand, leading to the 2D-layered perovskite film with a PLQY of 80% and the deep-blue PeLED ($\lambda_{\text{EL}} = 445$ nm) with an EQE$_{\text{max}}$ of 3.08% and a $L_{\text{max}}$ of 1315 cd m$^{-2}$. [61]

### 3.2. Luminescence Stability

Regardless of the significant boost in device efficiency, the luminescence stability of blue PeLEDs is still a considerable challenge for the practical use in full-color display and lightings. Particularly, perovskite films incline to degrade under some special circumstances, such as light illumination, moisture exposure, heat and electrical bias, and the degradation pathways arise from various physical and chemical factors. Here, the origins for the luminescence instability and the key challenges are discussed in terms of composition engineering and surface passivation.[50,62–76]

#### 3.2.1. Composition Engineering

It has been known that most defects occurring during the growth of perovskites are shallow defects, which do not contribute to carrier trapping and nonradiative recombination. As a result, perovskite materials have a high defect tolerance that enables huge freedom in the composition engineering with a variety of component choices to intentionally tune the electronic and optical properties. To improve the luminescence stability of perovskites, A-site and B-site doping in the ABX$_3$ formula have been attempted. Swarnkar et al. presented a new strategy that used other metal ions as a substitute to replace Pb$^{2+}$ (B-site cation) for the phase stabilization of CsPbX$_3$ perovskites, achieving stable optoelectronic devices with stable emission.[69] Benefiting from high diffusion rate of halides, postsynthetic halide exchange reactions at room temperature have been widely used for the growth of perovskites. However, the postsynthetic exchange of Pb$^{2+}$ ions is not a simple and direct way. Mondal et al. prepared blue-violet-emitting ($\lambda = 406$ nm) CsPbCl$_3$ perovskite NCs by treating CsPbCl$_3$ with CdCl$_2$ at room temperature, resulting in the easy replacement of Pb$^{2+}$ ions with Cd$^{2+}$ ions.[73] The nonradiative carrier trapping centers were suppressed to facilitate radiative recombination process, and the Cd-doped CsPbCl$_3$ NCs exhibited near-unity PLQY, high air stability, and high photostability. Meanwhile, Mn-doped perovskite NCs have been proved to be highly efficient emitters, arising from the quick ET. Hou et al. added a small amount of Mn dopant into the perovskite NCs, which gave rise to the three-fold increase in PLQY and maintained robust deep-blue emission ($\lambda_{\text{EL}} = 466$ nm) with an EQE$_{\text{max}}$ of 2.12%.[74]

| Structure | $\lambda_{\text{EL}}$ [nm] | EQE$_{\text{max}}$ [%] | $L_{\text{max}}$ [cd m$^{-2}$] | $T_{50}$ [min] | Ref. |
|-----------|-----------------|-----------------|-----------------|-------------|-----|
| ITO/PEDOT:PSS/TFB/PFI/CsPbBr$_3$·PEACl·YCl$_3$/TPBi/LiF/Al | 485 | 11.0 | 9040 | $\approx$80 | [19] |
| ITO/PEDOT:PSS/PEAO$_2$(Cs$_{1-x}$EA$_x$PbBr$_3$)$_2$PbBr$_4$/TPBi/LiF/Al | 477 | 4.8 | – | – | [19] |
| ITO/PVK/PFI/P-PDA-PbI$_2$/PEAO$_2$/TPYMB/Liq/Al | 465 | 2.6 | 211 | 13.3 | [59] |
| ITO/PEDOT:PSS/(Cs/FA/p-F-PEA)PbCl/Br$_3$/[TPBi/LiF/Al] | 469 | 4.14 | 451 | 14 | [60] |
| ITO/PEDOT:PSS/TPD/Bl$_2$PbBr$_4$/TPBi/LiF/Al | 445 | 3.08 | 1315 | 210 | [61] |
| ITO/NiO$_2$/PEAO$_2$...PA$_2$(CsPbBr$_3$)$_{n-1}$...PbBr$_4$/TPBi/LiF/Al | 466 | 7.51 | 1765 | 66 | [68] |
| ITO/PEDOT:PSS/TFB/PFI/CsMn$_{n_1}$Pb$_{1-x}$Br$_x$Cl$_{3-n}$/TFBi/LiF/Al | 488 | – | 245 | – | [74] |
| ITO/PEDOT:PSS/PATA/CsPbBr$_3$(NCs)/MoO$_3$/Ag | – | 12.3 | – | 20 | [76] |
| ITO/NiO$_2$/PSSNa$_x$/PA$_3$(CsPbBr$_3$)$_{n-1}$...PbBr$_4$/TPBi/LiF/Al | 492 | 1.45 | 4359 | 220 | [77] |
| ITO/NiO/TFB/PVK/(FABr/CsPbBr$_3$)/TPBi/LiF/Al | 483 | 9.5 | 54 | 250 | [79] |
| ITO/p-NiO/Cs$_5$Cu$_{14}$(NCs)/TPBi/LiF/Al | 445 | 1.12 | 263.2 | 6480 | [78] |
3.2.2. Surface Passivation

The poor film quality of perovskites inevitably hinders the realization of efficient and stable blue PeLEDs. The rapid crystallization of the Cl/Br mixed halide perovskites usually induces inhomogeneous and inconsecutive film morphology, leading to high leakage current and poor luminescence. Particularly, quasi-2D perovskites possess some disadvantages such as low-order phases and defect-induced nonradiative recombination, which severely restrict the luminescence efficiency and stability. By introducing 2D PEA$_2$PbBr$_4$ into quasi-2D P$_{A2}$(CsPbBr$_3$)$_{n-1}$PbBr$_4$ solution, Ren et al. fabricated a profitable blue-emission quasi-2D perovskite of PEA$_{2-x}$P$_x$(CsPbBr$_3$)$_{n-1}$PbBr$_4$ with the low-$n$ phase suppression.

Figure 4. Cation incorporation in the blue-emission perovskites. a) A photograph of the YCl$_3$-incorporated perovskite films under UV lamp. b) PLQYs versus light intensity of the perovskite films with different YCl$_3$ compositions. c) EQE versus current density of sky-blue PeLEDs with YCl$_3$. a–c) Reproduced under the terms of the CC-BY 4.0 license.© 2019, The Authors, published by Springer Nature. d) Device performance of PeLEDs with different EABr ratios. d,e) Reproduced under the terms of the CC-BY 4.0 license.© 2020, The Authors, published by Springer Nature. f) Schematic of the EA cation doping in quasi-2D perovskite. e) Device performance of PeLEDs with different EABr ratios. d,e) Reproduced under the terms of the CC-BY 4.0 license.© 2020, The Authors, published by Springer Nature. f) Schematic of bipolar-shell resurfacing of neutral and sterically stabilized perovskite NCs. g) PL peak shift of bipolar-shell-stabilized CsPbBr$_3$ perovskite NCs for monitoring the anion exchange rate at different reaction times. h) PLQYs of bipolar-shell-stabilized perovskite NCs and NC solid films. i) EQE of blue PeLED based on perovskite NC solid film. f–i) Reproduced with permission.© 2020, Springer Nature.
and the efficient ET. As a result, a superfluous amount of PEABr could efficaciously passivate the quasi-2D perovskite, and the blue PeLED ($\lambda_{\text{EL}} = 488$ nm) showed an $\text{EQE}_{\text{max}}$ of 7.51%, a $L_{\text{max}}$ of 1765 cd m$^{-2}$, and the improved $T_{50}$ of 66 min. Meanwhile, Dong et al. presented a different approach that utilized a solution-based ligand exchange to passivate the film of close-packed perovskite NCs with a bipolar shell, consisting of an interior anion shell, an external cation shell and solvents of polar molecules (Figure 4f). This strategy could simultaneously control the colloidal stability and short-ligand passivation, whereas the quantum confinement was retained in bipolar-shell-stabilized CsPbBr$_3$ perovskite NCs (with a diameter of 4 nm) (Figure 4g). An excellent blue PeLEDs based on perovskite NCs were fabricated with a near-unity PLQY (surpassing 90%) (Figure 4h) and an $\text{EQE}_{\text{max}}$ of 12.3% (Figure 4i). In addition, the $T_{50}$ reached 20 min at an initial luminance of 90 cd m$^{-2}$, demonstrating that the bipolar-shell perovskite NCs could improve the device stability.

### 3.3. Imbalanced Charge Injection

In addition to the abundant nonradiative recombination centers in the perovskite films, the mismatched energy-level alignment between perovskites and the adjacent charge-transport layers will cause the charge accumulation at the perovskite interfaces, giving rise to the enhancement of Auger nonradiative recombination and the accelerated decomposition of the perovskite films. Correspondingly, the operational lifetime and spectral stability of blue PeLEDs will be decreased significantly. This situation is more serious for blue PeLEDs than that of green and red counterparts, as blue-emission perovskites possess the deeper valence band maximum (VBM) and higher conduction band minimum (CBM). For example, the VBM of blue-emission perovskites are commonly deeper than the highest occupied molecular orbital (HOMO) of the typical hole-transport layer (HTL), leading to a large hole-injection barrier as compared with the injection barrier for electrons. Furthermore, the different carrier mobilities of electron-transport layer (ETL) and HTL cause the discrepant charge-transport rate and thus an imbalanced charge injection. The electron leakage toward HTL will cause irreversible physical damage to HTL. To balance the injection of electrons and holes, it is highly desirable to optimize the device architecture of blue PeLEDs by adopting the suitable ETL and HTLs with well-matched energy-level alignment and charge mobilities.

To balance the charge injection and suppress the nonradiative decays, Ren et al. designed a bi-layer HTL structure using the dipole characteristics of PSSNa on NiO$_x$ to improve hole transport. In this way, the hole-injection barrier was minimized (Figure 5a), and the nonradiative recombination at the interface between NiO$_x$ and quasi-2D perovskite film was significantly suppressed (Figure 5b), yielding a sky-blue PeLED ($\lambda_{\text{EL}} = 492$ nm) with an $\text{EQE}_{\text{max}}$ of 1.45% and a $L_{\text{max}}$ of 4359 cd m$^{-2}$. More importantly, the $T_{50}$ operational lifetimes were enhanced to 220 and 120 min, at the initial luminance of 150 and 415 cd m$^{-2}$ (Figure 5c), respectively. Zheng et al. used the solution-processed poly[9,9-dioctyl-fluorene-co-N-(4-butylphenyl) diphenylamine] (TFB)/Li-doped poly(9,9-dioctyl-fluorene-co-N-(4-butylphenyl) diphenylamine) (TFB)/Li-doped poly(9,9-dioctyl-fluorene-co-N-(4-butylphenyl) diphenylamine) (TFB)/Li-doped polyvinylcarbazole) (PVK) bilayered HTL in blue-emission quantum-dot LEDs, and the bilayer HTL provided a stepwise energy-level structure for not only reducing the hole-injection barrier but also facilitating the hole transport. The TFB/Li-doped PVK-based quantum-dot LEDs record a $L_{\text{max}}$ of 5829 cd m$^{-2}$ and an $\text{EQE}_{\text{max}}$ of 5.37%. Shin

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**Figure 5.** Balanced charge injection via interfacial engineering. a) Schematic of dipole moment formation on NiO$_x$ by PSSNa and the increased work function of NiO$_x$-PSSNa film. b) TRPL decay profiles of perovskites on NiO$_x$ and NiO$_x$-PSSNa, and perovskite with KBr additive on NiO$_x$-PSSNa. c) Luminance decay of blue PeLEDs at a constant current. a–c) Reproduced with permission. Copyright 2019, Wiley-VCH. d) Molecular structure of PFN-Cl, PFN-Br, and PFN-I. e) TRPL decay profiles of perovskite NCs on different glass/poly-TPD with PFN-Cl, PFN-Br, and PFN-I. f) Luminance decay of blue devices with different interface modifications. d–f) Reproduced with permission. Copyright 2020, American Chemical Society.
et al. demonstrated an interfacial engineering strategy with conjugated polyelectrolytes PFN-X (X = Cl, Br, or I) (Figure 5d), functionalized PFN (poly[(9,9-bis(3’-N,N-dimethylamino)propyl)-2,7-fluorene-alt-2,7-(9,9-dioctylfluorene)]) with halide anions, between HTL and perovskite NCs. The PFN-X exhibited the effective electron blocking capability, leading to the well-balanced charge recombination with the reduced hole-injection barrier through a high interfacial dipole moment (Figure 5e). Thus, the blue PeLEDs ($\eta_{\text{EL}} = 470 \text{ nm}$) show an $\eta_{\text{EL}}$ of 9.5% and $T_{\text{0}}$ of 250 min. It has also been observed that incomplete internal ET may lead to multiple EL peaks due to spatial and dielectric confinements in quasi-2D perovskite. Therefore, Wang et al. presented a deep-blue PeLED ($\eta_{\text{EL}} = 445 \text{ nm}$) based on Cs$_2$Cu$_2$I$_4$ perovskite NCs with a high PLQY of 87% and an $\eta_{\text{EL}}$ of 1.12%. More importantly, this device exhibited the unchanged EL peak position regardless of the applied bias, indicating a novel way to improve the luminescence stability of the deep-blue PeLEDs.

### 3.4. Optical Loss

For a LED built in a standard substrate emitting architecture, the EQE is determined by the internal quantum efficiency and light outcoupling efficiency ($\eta_{\text{out}}$) according to the equation $\eta_{\text{out}} = \eta_{\text{max}} \times \eta_{\text{in}}$, in which $\eta_{\text{in}}$ is the internal quantum efficiency. Upon the utilization of composition engineering, dimensional modulation, and surface passivation, the PLQY of perovskite materials has been dramatically boosted and the IQE of PeLEDs is approaching 100%. However, the EQE is severely limited by the low $\eta_{\text{out}}$ that is approx. 20%. The majority of internally generated light is confined in the waveguide mode due to largely different refractive indices between perovskite and glass substrate as well as in the substrate mode due to total internal reflection (TIR) at the glass/air interface. In addition to the waveguide and substrate modes, the optical loss induced by surface plasmon mode should be considered, which originates from the longitudinal oscillation of electrons between metal and dielectric layers. Meng et al. demonstrated that the device structure consisting of transparent electrode/high-index transport layer/perovskite emissive/low-index transport layer reflective electrode could raise $\eta_{\text{out}}$ to 12.0%, 17.5%, and 31.2% for blue, green, and red PeLEDs by reducing the optical loss in surface plasmon and waveguide mode. Therefore, the low $\eta_{\text{out}}$ leaves considerable opportunity for a substantial boost in EQE of blue PeLEDs if the optical loss can be efficiently suppressed, including waveguide mode, surface plasmon mode, and substrate mode.

Many techniques have been implemented for the light outcoupling enhancement for various LED technologies, including the use of photonic structure at the appropriate device interfaces, refractive-index coupling layers, microlens arrays, diffraction gratings, and so on. Jeon et al. used a randomly distributed nanohole array (NHA) embedded in a Si$_n$ layer between ITO and glass substrate to facilitate the outcoupling of the waveguide mode (Figure 6a). High-efficiency red/near-infrared PeLEDs with an $\eta_{\text{EL}}$ of 14.6% were realized (Figure 6b). Shen et al. presented a moth-eye nanostructures (MEN)-based outcoupling structure at the front electrode/perovskite interface by inserting a nanoimprinted ZnO layer to release the light trapped in the waveguide mode (Figure 6c). A half-ball lens was further used to outcouple the light trapped in the substrate mode, resulting in an extremely efficient green-emission CsPbBr$_3$ PeLEDs with an $\eta_{\text{EL}}$ of 28.2% (Figure 6d). Cao et al. proposed the light extraction from the waveguide mode in all directions by preventing the leakage current with thin organic insulating layer between perovskite submicrometric platelets, resulting in an $\eta_{\text{EL}}$ of 20.7% for infrared PeLEDs.

### 3.5. Toxicity

Although blue PeLEDs possess broad prospects, their application is still limited by two aspects: chemical/thermal instability and toxicity. The chemical/thermal instability will affect the operational lifetime of the devices, whereas the use of Pb ions in perovskites may have the potential influence on health and the environmental pollution. The ecofriendly Pb-free halide perovskites have drawn considerable attention as a promising alternative for the practical applications. Similar to Pb$^{2+}$, Sn$^{2+}$ has a $\sigma^2$ outmost electronic configuration, and the antibonding hybridization with halide anion is favorable to the formation of shallow defect states for CT and carrier extraction. In addition, the Pb$^{2+}$ ions in the 3D perovskite framework of corner-sharing octahedral mode can be replaced by cation pair of monovalent metals (such as Na$^+$, Ag$^+$) and trivalent metals (such as Sb$^{3+}$, Bi$^{3+}$) based on the principle of cation transformation.

In 2016, Leng et al. fabricated blue-emission Pb-free perovskite NCs of MA$_3$Bi$_2$X$_6$ (X = Cl, Br, I) through a collaborative LARP method, and the PL peaks could be tuned from 360 to 540 nm by controlling the anion composition ratios. The deep-blue PeLEDs were achieved with an EL peak at 430 nm and the PLQY of 12%. Subsequently, Sb-based perovskite has been proved to possess higher absorption coefficient than that of Bi-based counterparts. Zhang et al. successfully fabricated Cs$_3$Sb$_2$Br$_9$ perovskite NCs with an emission at 410 nm (Figure 7a), which reached a PLQY of 46% and FWFM of 41 nm (vs a FWHM of MA$_2$Bi$_2$Br$_6$ is 62 nm). Liu et al. prepared Cs$_2$AgInCl$_6$ and Bi-doped Cs$_2$AgInCl$_6$ NCs by the thermal injection method with oleic acid, oleylamine, and octadecene as ligands and solvents. Undoped Cs$_2$AgInCl$_6$ NCs showed blue emission at 470 nm, whereas Bi-doped Cs$_2$AgInCl$_6$ NCs showed wide orange emission around 580 nm with the greatly improved PLQY up to 11.4%. Recently, Huang et al. developed a colloidal solution-phase synthetic approach to obtain homogeneous size-distribution single-crystalline Cs$_3$EuCl$_3$ NCs, which exhibited a PLQY of 2% at 435 nm with a narrow FWHM of 19 nm (Figure 7b). In addition to the improvement in color purity and the degree of saturation, the embedding of Cs$_3$EuCl$_3$ NCs into a polymer matrix could increase the device stability under continuous laser irradiation. Chen et al. reported sky-blue Cu-treated halide
perovskite NCs by adding CuCl$_2$ to Cs$_x$FA$_{1-x}$PbBr$_3$ NCs, exhibiting an EQE$_{\text{max}}$ of 5.02%.

4. Conclusion

This Review has systematically summarized the factors that restrict the EL and stability of blue PeLEDs and the recent progress to improve luminescence efficiency and stability. The methods to optimize blue-emission perovskite materials, enhance luminescence stability, balance charge injection, and minimize the optical loss are discussed.

First, the fabrication of homogeneous perovskite films is of utmost importance to achieve high-performance blue PeLEDs with high luminance and robust stability. Several methods are usually utilized to manipulate rough film morphology and extensive pinholes by controlling the perovskite crystallization rate, optimizing the deposition method, and mixing the polymeric additives into perovskite precursors. In addition, it is a crucial challenge to improve the stability of perovskite NC films, as the degradation of perovskite NC films can lead to a deterioration
of the optical properties, such as low PLQY, reduced device efficiency, and spectral variation. In addition, the phase transition of all-inorganic perovskite NCs can take place between three photoactive black phases (e.g., α phase [cubic], β phase [tetragonal], γ phase [orthorhombic]), and the photo-inactive yellow δ phase (orthorhombic). To address this problem, replacing Pb\(^{2+}\) with B-site dopants such as Mn\(^{2+}\) can provide an effective method to enhance the stability in blue-emission perovskite NCs.

Second, the precise regulation of the emission wavelength is necessary for the fabrication of blue PeLEDs. Two methods are widely used to obtain the blue emission. One is the composition engineering by controlling the halogen components to tune the bandgap of mixed halide perovskite. Unfortunately, the phase segregation will occur in the mixed halide perovskites due to the electric field-induced migration of halogen ions, causing an undesirable wavelength shift from blue to green. The other is the dimensional engineering for quasi-2D perovskite by modulating the low-dimensional domain with the controlled layer number of inorganic octahedrons. However, this approach would attenuate the color stability because of multiple emission peaks that arise from mixed phases.

Third, the mismatched energy-level alignment in PeLEDs is detrimental to the device performance. Interlayers or interfacial solutions provide the effective pathways to facilitate charge injection into the perovskite emitters. The selection of the suitable ETL and HTL with well-matched charge-transport capabilities are commonly used to balance charge injection and suppress the nonradiative recombination due to the interfacial charge accumulation. Although 2\(^{-}\)2\(^{1}\)(1,3,5-benzoic acid triyl)tris(1-phenyl-1-H-benzimidazole (TPBi) is widely used as a ETL, its carrier mobility is one order of magnitude higher than that of the widely used HTLs, leading to the imbalanced charge injection between electrons and holes. In addition, the incorporation of inorganic charge-transport materials holds the great potential to improve the operational stability because of their high moisture resistance and oxygen resistance.

Fourth, there is huge room for improvement of \(\eta_{\text{out}}\). To unlock the full potential of blue PeLEDs, minimizing the optical loss is required without distorted output spectra and limited enhancement at special viewing angles. However, the light outcoupling enhancement is rarely reported for blue PeLEDs. Many techniques have been investigated to enhance \(\eta_{\text{out}}\) for other LED technologies with the substrate surface modifications and the integration of nanostructures inside the devices for the wave-guided light outcoupling. It is thus reasonable to imitate these design guidelines in blue PeLEDs as a potential direction to improve device performance and stability.

Finally, the intrinsic toxicity of Pb-based perovskite materials is of great concern for blue PeLEDs. Replacing Pb\(^{2+}\) cations with the less toxic group IV metal ion is a simple method to fabricate Pb-free perovskites. Meanwhile, the double perovskites with a general formula of \(A_3M^+M^{2+}X_6\) have emerged as a promising alternative to Pb-based halide perovskites by replacing two toxic Pb\(^{2+}\) ions with one M\(^{2+}\) cation and one M\(^{3+}\) cation. Recently, rare-earth-metal halide perovskites are regarded as potential perovskite candidates on account of their superior luminescence properties. For example, the bulk crystal of hybrid europium halide perovskite consisting of \((C_6H_5NH_3)_2EuI_4\) and CsEuBr\(_3\) processes strong blue emission with a narrow FWHM of 25–35 nm. However, it is still difficult to control the uniformity and morphology of europium halide perovskite, which is synthesized by solid-state methods. Therefore, research directions should pay more attention to improve the synthetic route of europium or other rare-earth halide perovskites in the future to form perovskite films of uniformity and homogeneity. Moreover, the solubility of rare-earth precursors remains a challenge for the fabrication of blue PeLEDs.

Therefore, as a crucial component of display techniques, blue PeLEDs are still suffering from poor device performance and severe instability both in long-term operation and emission spectrum. More efforts are still needed to continuously improve the EL efficiency and stability toward the practical applications of blue PeLEDs.

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**Conflict of Interest**

The authors declare no conflict of interest.
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