

Review

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Review

Metal Halide Perovskite: A Promising Materials for Light Emitting Diodes

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Abstract: Metal halide perovskites have shown excellent optoelectronic properties, including high photoluminescence quantum yield, tunable emission wavelength, narrow full-width at half-maximum and low-cost solution-processed fabrication, which make it exhibit great potential as the emission layer materials of light emitting diodes. With the joint efforts of researchers from different disciplines, there has been a significant progress in the improvement of external quantum efficiency (EQE) and stability of perovskite light emitting diodes (PeLEDs) in recent few years, especially in green PeLEDs with EQE over 30%. In this review, we firstly introduce the basic device structure of PeLEDs, as well as the factors influencing EQE and stability of PeLEDs. Secondly, the development of lead-based and lead-free PeLEDs are summarized systematically. Thirdly, challenges of PeLEDs are discussed in detail, including low EQE of blue PeLEDs, poor device stability and EQE roll-off. Finally, some suggestions and perspectives of the future research directions for PeLEDs are proposed.

Keywords: metal halide perovskites; light emitting diodes; external quantum efficiency; stability; photonics

1. Introduction

Metal halide perovskites refer to a class of materials with a generic formula ABX_3 where A is Cs^+ , MA^+ or FA^+ , B is Pb^{2+} or Sn^{2+} , and X is Cl^{-1} , Br^{-1} or I^{-1} (Figure 1a), these materials not only have the benefits of high photoluminescence quantum yield (PLQY), tunable bandgaps and high color purity, but also can be fabricated easily via a sample chemical vapor deposition (CVD) method or solution-processing [1–3]. These merits enable the metal halide perovskite as the excellent emission layer in light emitting diodes to satisfy the new International Telecommunication Union Rec. 2020 display standard [4–6]. Since the impressive work of perovskite light emitting diodes (PeLEDs) reported by Tan et al. in 2014 [7], PeLEDs have achieved prosperous development with impressive peak EQE values of blue, green, red, and near-infrared PeLEDs of 18.65%, 30.84%, 25.8%, and 23.8% respectively [8–11]. The common device structure of PeLEDs is shown in Figure 1b, which include substrate, charge injection layers, charge blocking layers, perovskite emission layer and electrode (Figure 2a) [12]. When charges are injected from charge injection layers into the perovskite emission layer, the electron-hole pairs are generated. These electron-hole pairs recombine radiatively, and the photons are emitted [13].

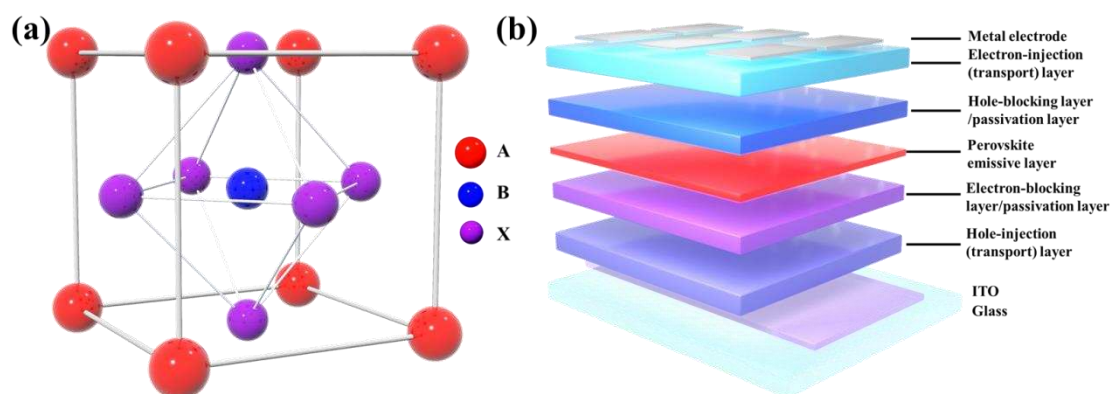


Figure 1. Illustrations of (a) crystal structure of perovskites, (b) device structure of a perovskite LED.

One of important performance parameter of PeLEDs is EQE [12], which is expressed as: $EQE = \eta_{inj} \times \eta_{rad} \times \eta_{out}$. The η_{inj} refer to the efficiency of charges injection from device electrodes into the perovskite emission layer. A bad energetic alignment and high trap state density at the interface will cause the nonradiative recombination of carriers and unbalanced injection of electron and hole, leading to a lower η_{inj} [14–16]. η_{rad} is the radiative recombination efficiency of electron-hole pairs, which are directly related to the intrinsic property of emission layer materials, namely their PLQY. As for the η_{out} , it is the outcoupling efficiency and reflects the ability of emitting photons generated from perovskite emission layer into free space by PeLEDs device. The value of η_{out} is affected by the degree of mismatch of refractive index between different device layers, the large mismatch would yield severe optical losses via substrate and waveguide modes [17]. Hence, a perfect energy energetic alignment, high-quality perovskite emission layer and interface, and well-matched refractive index between different device layers are essential to develop ideal PeLEDs with large EQE [18].

Besides, the lifetime and stability of PeLEDs are also important for their commercial application. Compared with the nucleation and crystallization barriers of silicon semiconductors (470 kJ mol⁻¹ and 280 kJ mol⁻¹ respectively), the crystallization of perovskites materials can be achieved at lower temperature due to smaller crystallization barriers (56.6–97.3 kJ mol⁻¹) [19]. Because of the low formation energy and soft ionic property of perovskites, the quality of perovskite films fabricated by solution-processing, however, are poor owing to many types of defects at inter-grain boundaries, intra-grain boundaries, and the surface of perovskites [20–23]. Under the influence of temperature, electric field, moisture, UV light and other external factors, these defects will lead to phase segregation of mixed-halides perovskites, phase transition, broadening and shift of the peak emission, etc. [24–31]. Therefore, some reasonable strategies, including additive engineering, crystallization kinetic regulation, dimensionality control and interface modification, would be recommended to get a low defect density of the perovskite emission layer, which is in favor of the long-term and stable running of PeLEDs [16,32–36].

At the beginning of this review, the basic information of perovskites materials and device structures of PeLEDs as well as the factors influencing EQE and stability of PeLEDs are discussed systematically. Then the state-of-the-art for lead-based PeLEDs are summarized, from bulk, quasi-2D to 0D perovskites materials as the emission layer, due to lead-based perovskites superior optoelectronic properties. Some novel PeLEDs based on co-friendly compositions, such as tin-based and double metal perovskites, are also presented in section 3. Subsequently, we discuss the challenges of PeLEDs that remain to overcome, including low EQE of blue PeLEDs, poor device stability and EQE roll-off. In the end, a summary and outlook about PeLEDs in the future are provided.

2. Progress in lead-based PeLEDs

2.1. Progress in PeLEDs based on 3D lead-based perovskites

3D CsPbX_3 perovskites are commonly utilized as the emitter in PeLEDs owing to easy synthesis, composition regulation and good charge-transport characteristics without using organic ligands [6,37–39]. Their film can be fabricated by spin coating, blade coating, slot-die coating and inkjet printing [40–44]. These solution-processing methods, however, generally produce inferior quality perovskite films. Hence, some strategies, ranging from bulk passivation to surface/interface modification, are developed to improve the film quality of perovskites [37,45,46]. Cao et al. [47] introduced 5-aminovaleric acid into perovskite precursor solutions. With the optimization of the feed ratio of 5-aminovaleric acid in the precursor solution, the additive not only passivated the surface defect but also increased the outcoupling efficiency by the submicrometre-scale structures formed in perovskite films (Figure 2a). Finally, the PeLEDs achieved an exceeding 20% EQE at 800 nm. Although organic molecules can passivate defects effectively via Lewis bases bearing amino, carboxyl, sulfonate, phosphate, thiol and other functional groups [48–54], the inferior conductivity of these organic additives weakens the transportation of carriers and discounts the EQE of PeLEDs [55,56]. A multifunctional organic semiconductor 9,9'-bis(2-(2-(2-aminoethoxy)ethoxy)ethyl)fluorene (FI-OEGA) was unitized as an additive by Song and coworkers [57]. The additive was mainly distributed at grain boundaries, which released the residual stress and inhibited the ion migration (Figure 2b). The corresponding green PeLEDs device obtained a champion EQE of 21.3%. Mixing Bromide/chloride is a facile and common strategy to realize blue emission [58,59], the low ion migration energy of Cl^- , however, inevitably causes the generation of halide vacancies and phase separation [60,61], besides, the achievement of emission wavelength < 470 nm base on 3D mixed-Br/Cl perovskites is difficult due to the low solubility of chloride [62]. Wang et al. [63] designed an in situ anion exchange approach, where high Cl ratio and homogeneous of $\text{CsPbCl}_x\text{Br}_{1-x}$ perovskites were obtained via treating CsPbBr_3 film with solution of tetraphenylphosphonium chloride (Figure 2c). By additionally introducing the organic ammonium halide salts passivator, they fabricated deep-blue PeLEDs with desired EQE.

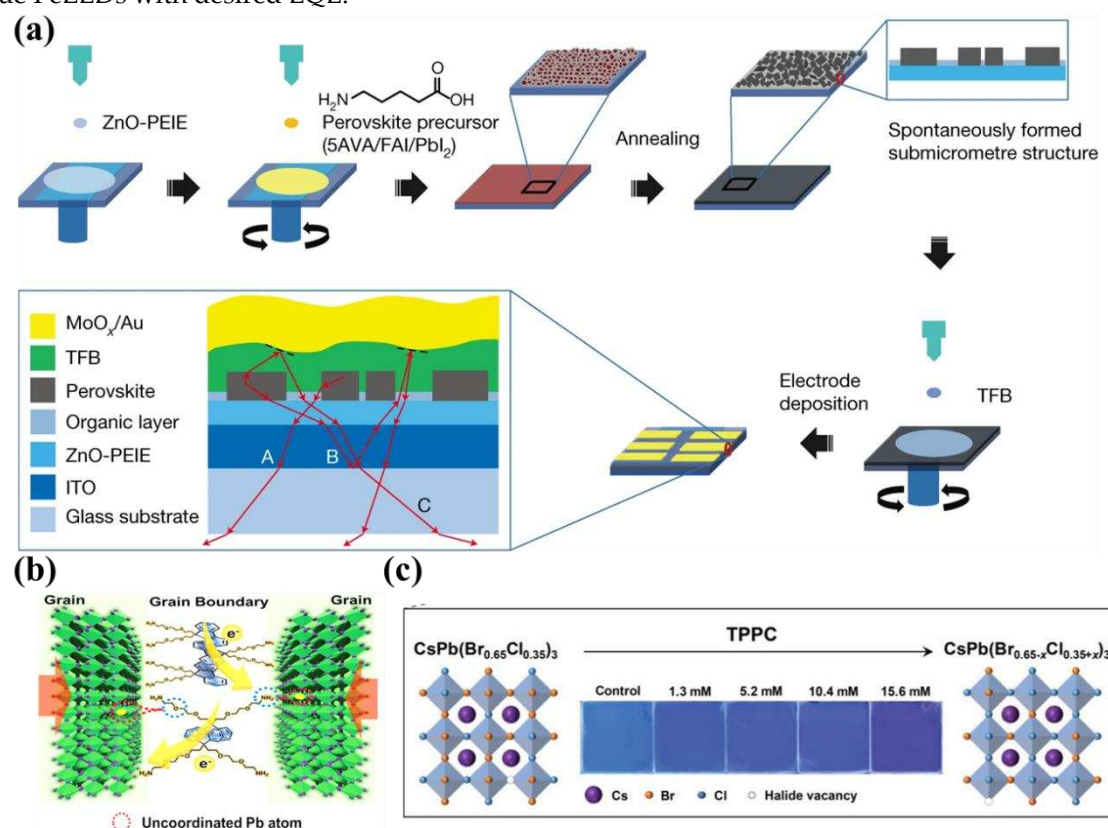


Figure 2. (a) The scheme of device fabrication with spontaneously formed submicrometre structure, the arrows A, B and C represent light trapped in devices with a continuous emitting layer, can be extracted by the submicrometre structure. crystal structure of perovskites. [12], (b) Illustration of the

defect passivation at grain boundary for perovskite film via FI-OEGA [57], (c) Mixed-halide perovskites film formed by an in situ halide ions exchange process [63].

A benign interface quality is also essential for efficient PeLEDs, it can improve band alignment and passivate surface defects, leading to rapid and balanced charge injection, restrict ion migration and nonradiative recombination, as well as ameliorate EQE roll-off and the stability of PeLEDs [64–67]. Wei et al. [68] dropped a phosphine oxide (PO-T2T) additive solution onto perovskites, the $-P=O$ bond in the additive bonded tightly with undercoordination Pb^{2+} by dative bond at both grain boundaries and surface (Figure 3a), besides, PO-T2T also formed a well energetic alignment to accelerated electron transportation between perovskites layer and electron transport layer (Figure 3b). Finally, a peak EQE of 22.06% for green PeLEDs was gained with satisfied maximum luminance (103286 cd m^{-2}). Ion migration driven by the electric field is a notorious problem for PeLEDs, especially under high current density, resulting in EQE roll-off for 3D perovskite-based PeLEDs [61]. Zhao et al. [69] washed the top surface of perovskites with chloroform to eliminate residual PbI_2 and restrain ion migration. By further inserting an ultrathin poly(methyl methacrylate), the optimized PeLEDs device realized the balance carriers injection and increasing EQE. The device could maintain $> 70\%$ of the original EQE at 1400 mA cm^{-2} (Figure 3c). In recent years, the buried interface between the hole transport layer and the perovskites layer has drawn increasing attention, because it affects directly the growth of perovskites. Despite the existence severe deep-level trap-state at the buried interface, it is usually more difficult to modify than that of the top surface of perovskites. Carbazole-phosphonic acid derivatives are desirable hole transport layer materials with fast hole transfer rates and low interfacial trap state density [70,71]. Di and coworkers [72] utilized [2-(9H-carbazol-9-yl)ethyl]phosphonic acid as the hole transport layer, in which the group of phosphonic acid could interact with Pb^{2+} to fill up halide vacancies and obtain a perfect buried interface. The corresponding blue PeLEDs devices had a peak EQE of 10.3 and 13.6% at 478 and 487 nm respectively with fine maximum luminance (Figure 3d). Polymers are desirable modification materials for the defect passivation and suppression the decomposition induced by humidity of perovskites [73–75]. Halpert and coworkers [76] reported a poly [(phenylglycidyl ether)-co-formaldehyde] (PCF)-protected perovskites layer as the emitter for PeLEDs (Figure 3e). The ether in PCF could form hydrogen bond with methylammonium cation and replace halide vacancy via coordination bond to increase perovskite film quality. Meanwhile, PCF also weakened the invasion of moisture under a $> 70\%$ RH environment, which was beneficial for the practical application of PeLEDs.

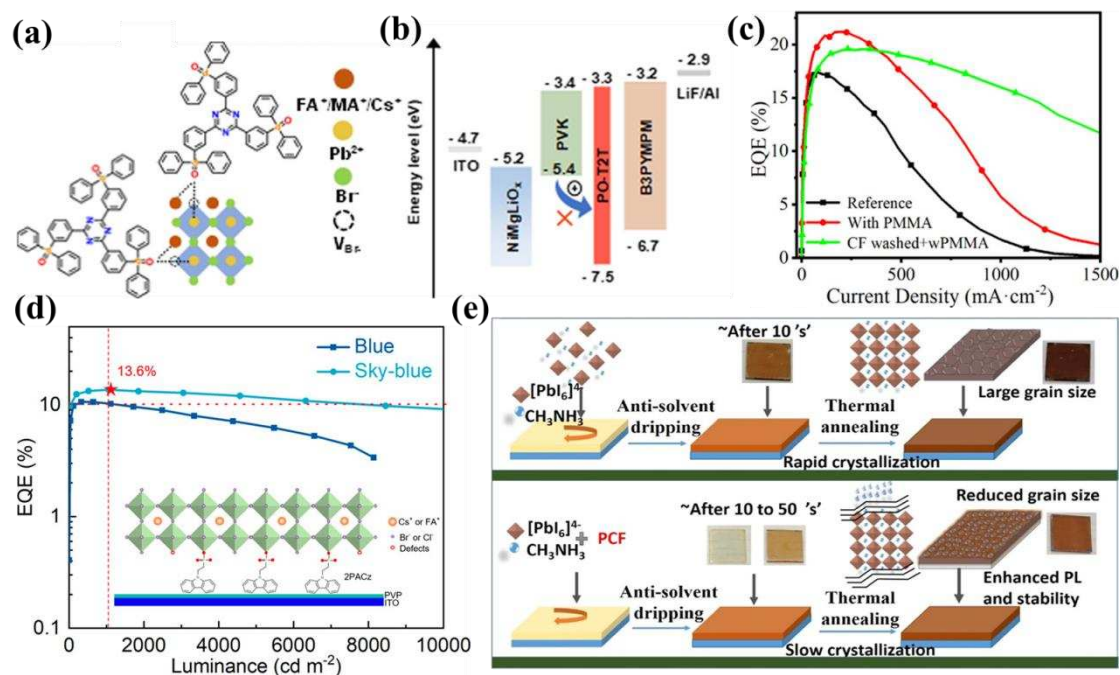


Figure 3. (a) The interaction between PO-T2T and perovskites [68], (b) The energy level alignment in PeLEDs [68], (c) EQE-current density relations of the reference, with PMMA layer and CF washed + PMMA device [69], (d) The evolution curves of EQE with the change of luminance (inset: the modification of carbazole-phosphonic acid at buried interface) [72], (e) Illustration of MAPbI₃ formation through one-step method without (top) and with (bellow) protection of PCF [76].

2.2. Progress in PeLEDs based on quasi-2D lead-based perovskites

Since the pioneering work reported by Calabrese et al [77], quasi-2D perovskites have been one of the promising materials for photoelectronic applications resulting from their unique structural characteristics [78]. They are formed through inserting bulky organic cations into the A site of 3D perovskites lattice. The general chemical formula of quasi-2D perovskites is $A'_2A_{n-1}B_nX_{3n+1}$, the element composition of A, B and X is similar to that of 3D perovskite, in addition, A' refer to the organic spacer cation and n indicate the layer number of [BX₆]⁴⁻ octahedral slab sandwiched organic spacer cations [79]. Because of the small value of dielectric constants for organic spacer cations, a "quantum-well" structure can be produced for quasi-2D perovskites [80,81]. The quantum- and dielectric-confinement effects originating from "quantum-well" structure contribute to the large exciton binding energy (E_b), combination with the tunable bandgap by changing the thickness of "quantum-well" and high PLQY due to efficient energy transfer from low n-phase to high n-phase, the quasi-2D perovskites is very suitable as the emission layer materials for PeLEDs [82].

A fine quality of quasi-2D perovskite films is the basic requirement for ideal PeLEDs. Tang et al. [82] added the bifunctional benzoic acid potassium into the precursor solution of mixed-halide quasi-2D perovskites, in which benzoic acid (BA⁻) group could as Lewis-base to interact with undercoordinated Pb²⁺, and K⁺ could bind tightly with halide atoms by ionic bond, leading to the throughout modification of quasi-2D perovskites with the reduction of defect-induced nonradiative recombination and halide ion migration (Figure 4a). A record value of EQE for sky-blue PeLEDs was achieved, and the work guided the selection of appropriate additives employed in perovskite materials. Though PLQY of quasi-2D perovskites is well resulting from their special properties previously mentioned, the undesirable electrical conductivity of organic spacer cations will retard the charge injection and undermine the EQE of PeLEDs [83]. Sirringhaus and co-workers [84] displaced organic spacer cations with sodium cation. The fabricated perovskites layer had a fine orientation, besides, an amorphous NaPbBr₃ was formed with the incorporation of sodium salt, resulting in the generation of the quasi-2D perovskites. By optimizing the feed ratio of sodium salt and a little organic additive, the green PeLEDs obtained 15.9% of EQE and a good operational lifetime (Figure 4b).

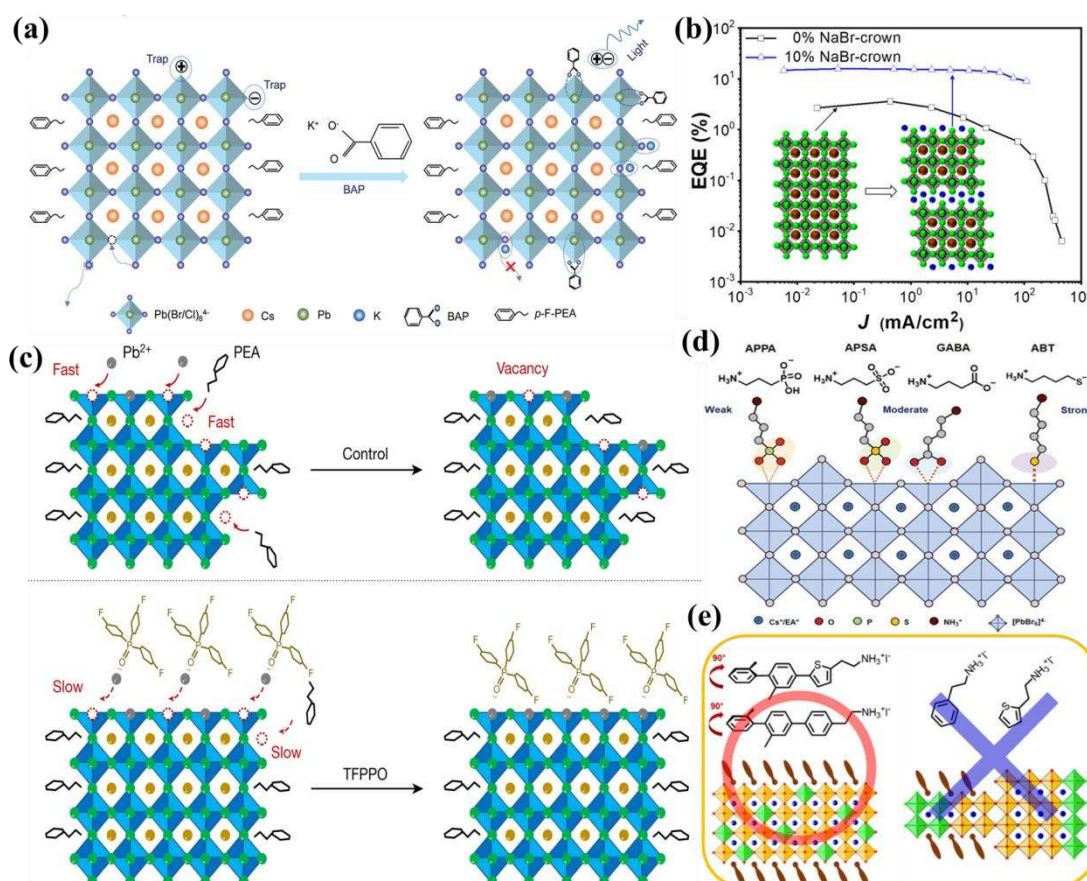


Figure 4. (a) The passivation function of additive in quasi-2D perovskites [82], (b) EQE-J curves of PeLEDs with and without addition of NaBr [84], (c) The process of crystallization for quasi-2D perovskites after the treatment of antisolvent with and without TFPPO [89]. (d) The interaction intensity between different zwitterions and perovskites [90], (e) Phase distribution in mixed-halide quasi-2D perovskites with different organic cation spacer [91].

Because the formation energy of different n phase of quasi-2D perovskites is similar, there is a wide n phase distribution in quasi-2D perovskites materials [85]. Although the cascade energy transfer from high- n phase to low- n phase enable the improvement of PLQY, a too wide distribution of n phase is unfavorable due to the low color purity and possible energy loss in energy transfer, in addition, low- n ($n = 1$) have strong electron-phonon coupling interaction, which cause the nonradiative recombination [86–88]. Hence, some strategies have been developed to narrow the n phase distribution of quasi-2D perovskites. Sargent and coworkers [89] synthesized a bifunctional additive tris(4-fluorophenyl)phosphine oxide (TFPPO) and dissolved it into antisolvent to treat perovskites during the LED devices fabrication. The strongly electronegative fluorine atoms in the additive interacted with the organic ammonium cations through hydrogen bond to restrict the low- n phase formation and achieve a monodispersed “quantum-well” structure, in addition, the phosphate could fill up the unsaturated sites to reduce the trap state density (Figure 4c). Finally, they realize a desired EQE value of 25.6% for green PeLEDs with exceeding 2 hours half-lifetime. The interaction intensity between organic spacer cation and Pb^{2+} influence directly the crystallization kinetics and n -phase distribution of quasi-2D perovskites. Yan et al. [90] investigated the effect of Pb^{2+} coordination ability of a series of zwitterions spacer on the different n -phase formation (Figure 4d). The results shown that the stronger coordination ability with Pb^{2+} zwitterions have, the easier low- n phase will obtain. Therefore, the zwitterions with moderate coordination ability could produce concentrated n -phase distribution of quasi-2D perovskites, which were used to fabricated a high-performance blue PeLEDs with a 15.9% of EQE. Apart from the control the n -phase distribution, the inhibition of phase separation induced by halide migration in mixed-halide quasi-2D perovskites is

also necessary. Yang et al. [91] comprehensively considered the heteroatom, conjugation length, and molecular twist of organic cation spacer, and they found twisted and extended conjugated organic cation spacer enabled the narrow n-phase distribution and negligible segregation of mixed-halide (Figure 4e). Eventually, they reported a number of stable and high-efficiency PeLEDs devices with emission wavelengths range from red to NIR area.

Interfacial engineering between quasi-2D perovskites and charge injection layers is an efficient strategy to ameliorate the performance of PeLEDs [33]. You group [92] utilized CsCl to modify the hole injection layer poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS). The incorporation of CsCl could achieve oriented alignment of PEDOT chains and change the position of the valance band, these modifications were beneficial to the improvement of carrier injection and transportation (Figure 5a). Moreover, according to the results of XRD and transient absorption spectra, the formation of the low-n phase was also restrained by the modified-hole injection layer (Figure 5b). The optimized sky-blue quasi-2D perovskite PeLEDs obtained 16.07% of EQE and a fine lifetime. Chueh et al. [93] added a conductive interlayer between the electron transport layer and perovskites emission layer, where the undesirable energy transfer and nonradiative recombination were inhibited (Figure 5c). They found the modification can significantly improve the brightness of PeLEDs and alleviate the EQE roll-off at high current density. The refractive index for most organic hole injection layer materials is very small compared with that of perovskite materials, and the large difference will cause the photons capture by hole injection layers [94,95]. To overcome the photon loss, Xie et al. [9] utilized NiO_x as the hole injection layer materials, which possessed the merits of adjustable energy levels and refractive index [96,97]. By a doping suitable ratio of Mg in NiO_x film and inserting a polymer layer before the deposition of the quasi-2D perovskite layer, the optimized green PeLEDs effectively improved the outcoupling efficiency (41.82%) as well as (Figure 5d), got a rapid and balance the charge carrier injection. Finally, a record EQE of 30.84% was achieved for green PeLEDs (Figure 5e).

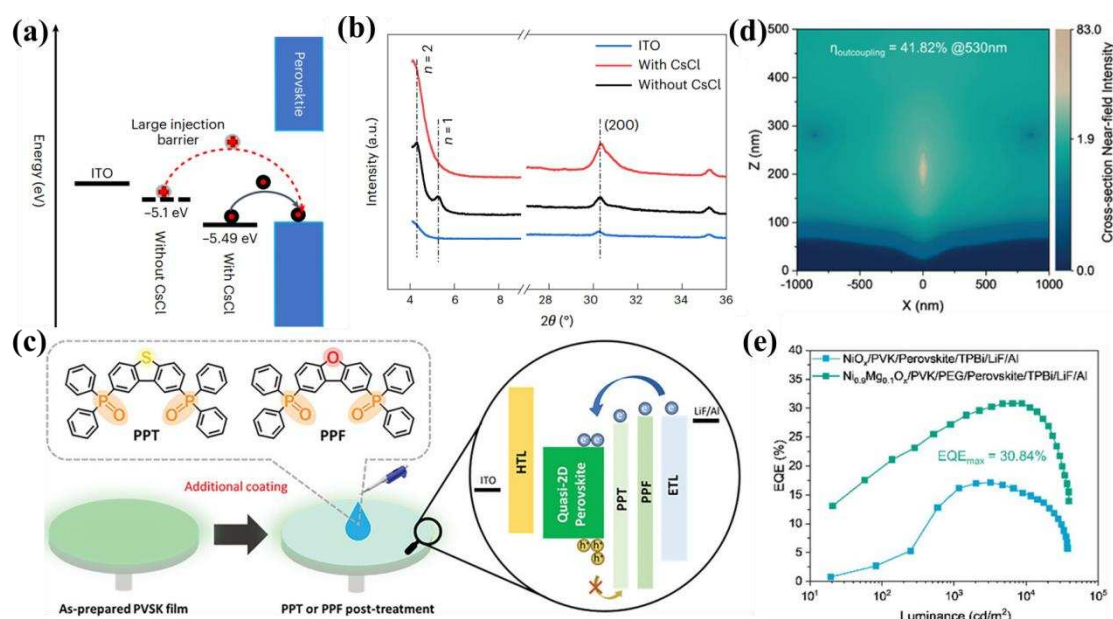


Figure 5. (a) Band alignment in PeLEDs using PEDOT:PSS with and without CsCl addition [92], (b) XRD results of perovskite films deposited on PEDOT:PSS substrates with and without CsCl addition [92], (c) Band alignment in PeLEDs with interfacial post-treatment [93], (d) Cross-section near-field intensity distribution of the optimized PeLEDs device for light outcoupling at 530 nm [9], (e) EQE-luminance curves of reference and modified PeLEDs [9].

2.3. Progress in PeLEDs based on 0D lead-based perovskites

The 0D perovskites, namely perovskite quantum dot (PsQD), can be viewed as a core of stoichiometric ABX_3 covered by a BX_2 inner shell and an $\text{A}'\text{X}'$ outer shell, where A' refers to either Cs^+

or ligand cation (normally alkylammonium ions), and X' are consist of halide anion and ligand anions (normally alkyl carboxylate ion) [98]. With the confinement of the surface ligand, the PeQD possesses some unique virtues, including adjustment emission wavelength via size and composition control, high color purity and near-unity PLQY [99–101]. Since the first report of the synthesis of PeQD colloidal solution by ligand-assisted reprecipitation method (LARP) in 2014 [102], some significant progress has been gained in PeQD-based LEDs (PeQD LEDs), such as exceeding 20% of EQE for red and green LEDs as well as 10% of EQE for blue LEDs [39,103,104].

Despite the promising potential of PeQDs in LEDs application, the highly dynamic adsorption and desorption of surface ligands for PeQD will influence its stability and PLQY during the process of purification, film formation and long-term storage [105]. Besides, long alkyl chains of ligands, such as commonly used oleic acid (OA) and oleylamine (OAm), have lower conductivity and poor interaction between PeQDs [106]. These shortages limit the practical application of PeQD LEDs. Hence, the synthesis of bright and stable PeQD colloidal solution is the primary requirement for corresponding LEDs and other optoelectronic device applications. Considering the facile ligand desorption due to the proton transfer between OA and OAm, Sargent et al. [107] designed a novel amine-free synthetic approach, in which CsOA and PbOA reacted with tetraoctylammonium halides, and obtained PeQD colloidal solution exhibited satisfying stability during purification process (Figure 6a). Based on the PeQD, the fabricated green and blue LEDs had several-fold improvement of EQE compared with traditional OA/OAm capped-PeQD LEDs. Kovalenko and coworkers [108] also changed the ligand type used in the hot injection method synthesis of PeQD. They added didodecyl-di-methylammonium halides, a constantly applied ligand in the post-treatment process of PeQD colloidal solution, into precursor solution as the singly ligand to get $\text{CsPb}(\text{Br}_{1-x}\text{Cl}_x)_3$ PeQD (Figure 6b). Benefiting from shorter alkyl chains compared with OA/OAm and optimized device structure, efficient pure blue LEDs were achieved. Zeng et al. [109] found dodecylbenzene sulfonic acid could act as a “Br-equivalent” ligand to bind tightly with Pb^{2+} due to the strong ionic nature of the sulfonate group, this inhibited not only the desorption of ligands but also the formation of halide vacancy (Figure 6c). Using the perfect ligand, the synthesized PeQD colloidal solution could achieve > 90% of PLQY easily and maintain desired stability even after 8 purification cycles or exceeding 5 months of storage, which shown promising application in LEDs.

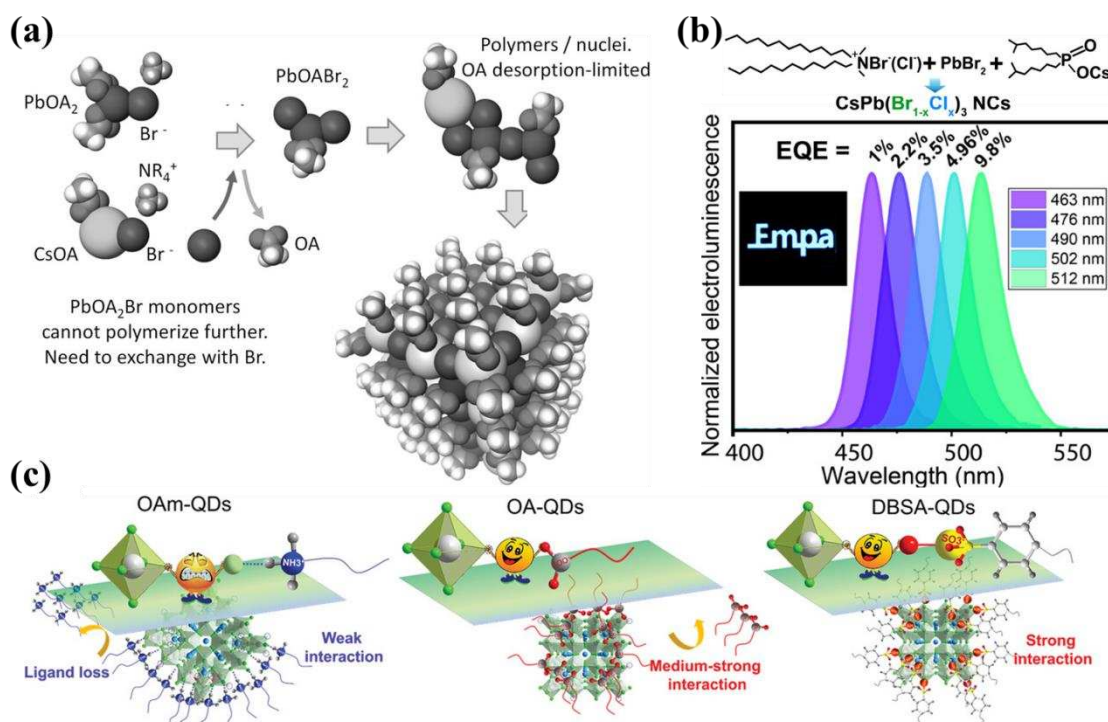


Figure 6. (a) Molecular dynamics simulation of CsPbBr_3 nucleation and growth steps without amine ligand by Density functional theory [107], (b) Normalized PL spectra of $\text{CsPb}(\text{Br}_{1-x}\text{Cl}_x)_3$ PeQDs with

didodecyldi-methylammonium ligand [108], (c) The interaction intensity between PeQD with ligand with different binding motif [109].

Ligand exchange is also an effective strategy to modify the stability and photoelectric property of PeQD colloidal solution [39,110,111]. Bakr and coworkers [112] added OA and di-dodecyl dimethyl ammonium bromide into purified PeQD solution. They proposed the OA could induced the desorption of protonated OAm, and then the PeQD surface would be reconstruction by the further substitution of OA with di-dodecyl dimethyl ammonium bromide. With the success of ligand exchange, they were able to achieve a green LEDs based on the halide-ion-pair-capped PeQD (Figure 7a). Zhang et al. [113] found dimethyl butylamine and ethyl bromide reacted spontaneously at room temperature via nucleophilic substitution to yield alkylammonium bromide, therefore, they added the two additives into purified PeQD solution, the alkylammonium and Br^- filled up the defect at A and X site respectively, and replaced partial OA/OAm ligand, lead to the improvement of PLQY, stability and electroconductivity of PeQD (Figure 7b). This in situ ligand compensation approach enabled the corresponding green PeQD LEDs to acquire 23.45% of EQE and 109427 cd m^{-2} of maximum luminance. As mentioned above, PeQD prefers to bind with strong polar ligands due to the soft ion nature of perovskites [114–117], these ligands cannot dissolve in non-polar solvents, high polar solvents, however, undermine the stability of PeQD [39,118–120]. Based on this situation, a liquid Iodotrimethylsilane ligand was used to treat the CsPbI_3 colloidal solution [121], the ligand possessed a fine solubility in non-polar solvents, and it can react with oleate to in situ etch PeQD surface by produced HI, resulting in the reduction of defect density and improvement of stability due to the strong-binding ligands. A 23% of EQE for red PeQD LEDs was eventually reported (Figure 7c), and this ligand has similar effect for other compositional PeQD. Compared with the organic cap layer, the inorganic shells have more advantages, for example, good conductivity and stability [111]. Lee et al. [122] utilized CsPbBr_3 PeQD as seed, through blended the seed with ZnCl_2 , ZnBr_2 and S-ODE, the $\text{CsPb}(\text{Br}_{1-x}\text{Cl}_x)_3$ PeQD covered with a ZnS shell was obtained due to low lattice mismatch (Figure 7d). The ZnS shell protected the $\text{CsPb}(\text{Br}_{1-x}\text{Cl}_x)_3$ core from the external effect, accelerated the carrier injection and restricted the ion migration of mixed-halide. Finally, they achieved a 1.32% of EQE for deep-blue LEDs with satisfying operational lifetime.

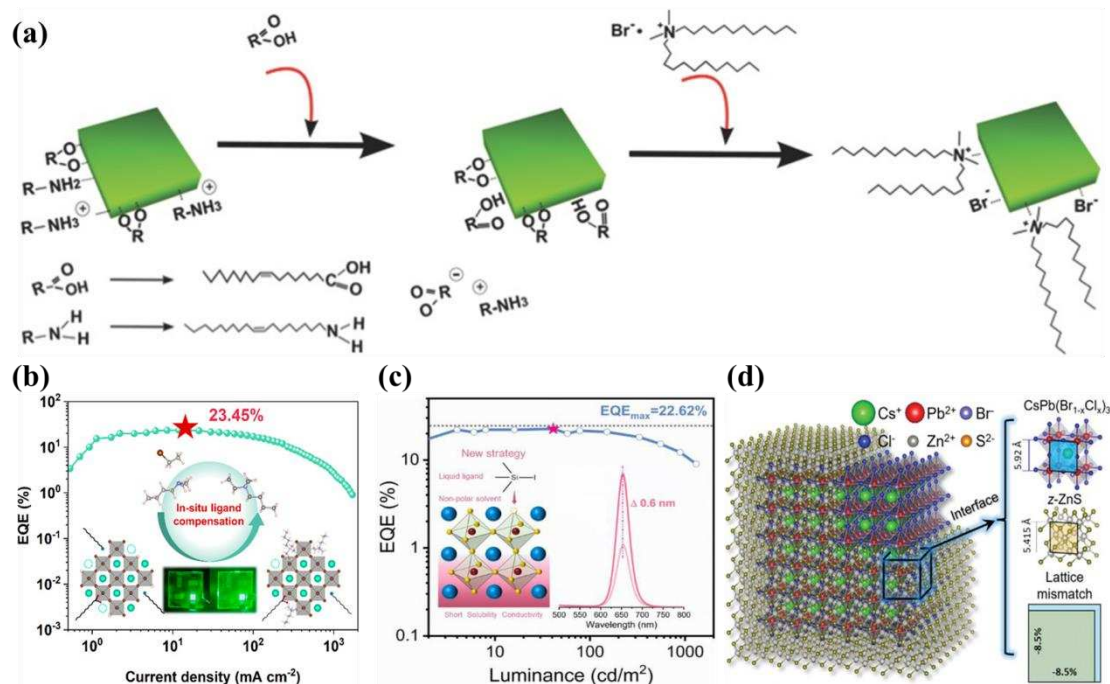


Figure 7. (a) The mechanism of ligand-exchange on surface of PeQD [112], (b) Illustration of in situ ligand compensation for PQDs and the EQE evolution with current density of optimized PeLEDs [113], (c) EQE-Luminance curves of PeLEDs device based on Iodotrimethylsilane ligand-treated PeQD [121], (d) Simulated 3D atomic model of CsPbBr_3 PeQD covered by ZnS [122].

Although PeQD films have lower defect state density than that of polycrystalline bulk films with the existence of surface ligands, there are also several undercoordinated Pb^{2+} at the surface of PeQD films due to the easy desorption and steric effects of ligands, so a post-treatment of PeQD films is necessary to get a high-efficiency EQE device [123,124]. Lee et al. [103] first doped guanidinium into FAPbBr_3 to increase the stability and charge confinement effect of PeQD. On the other hand, passivation interlayers (TBTB) were deposited on the top surface of PeQD films to further heal the bromide vacancy defect, which could ameliorate the photoluminescence properties of PeQD films and carrier injection balance of PeLEDs (Figure 8a). A 23.4% of EQE and 108 cd A^{-1} current efficiency were obtained in the optimized PeLEDs device (Figure 8b). Brovelli et al. [125] investigated the influence of emission layer thickness in the balancing charge transport, by further inserting the NiO_x layer both at top the surface and buried interface to optimize the energetic alignment (Figure 8c), they fabricated a stable green PeQD LEDs with EQE as high as 26.7%. The selection of suitable charge transportation layer materials is crucial for the formation of benign interfaces without inserting extra and complex modification materials. TPBi is the commonly used electron transportation layer material, it, however, has lower electron mobility than hole mobility of generally used hole transportation layer materials (such as PTAA and poly-TPD) [126–129]. Considering the case, Xu et al. [130] synthesized new electron transportation layer materials, denoted as B2, to facilitate the electron injection. This could perfectly balance the carriers injection and suppress nonradiative recombination at the top surface (Figure 8d), and a 13.17% of EQE for blue PeQD LEDs was achieved with an $8656.67 \text{ cd}\cdot\text{m}^{-2}$ of maximum luminance.

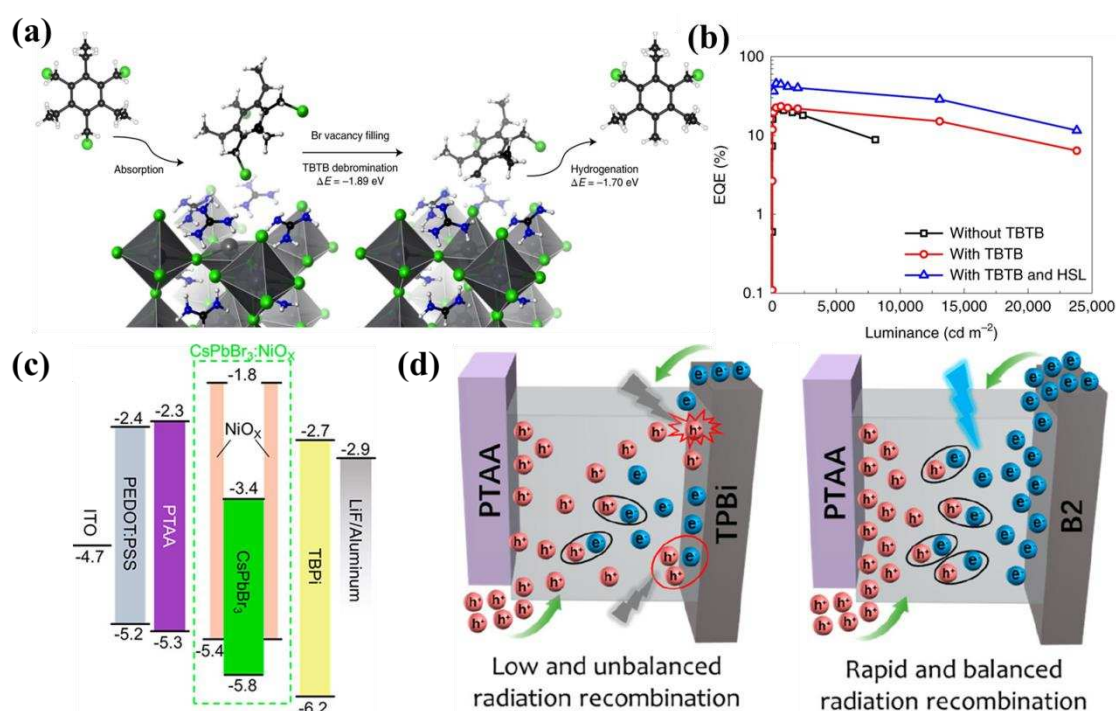


Figure 8. (a) The healing process of bromide vacancy by TBTB molecules on guanidinium-terminated FAPbBr_3 surface simulated by Density functional theory [103], (b) EQE-Luminance curves of PeLED with or without a TBTB interlayer and a hemispherical lens (HSL) [103], (c) Energy band alignment of PeLEDs with treatment of NiO_x [125], (d) Illustration of the charge recombination in PeLED devices based on TPBi (left) and B2 (right) [130].

Excepted the commonly used hot injection and ligand-assisted reprecipitation methods, in situ synthesis of PeQD in the substrate is simpler and more favorable to decrease the cost of LEDs device fabrication [131–133]. Yuan et al. [134] manipulated the chemical structure of phenylethyl ammonium ligands and in situ successfully produced monodispersed PeQD on the hole transportation layers. On the one hand, extra methyl substitution at head group inhibited the yield of layered perovskites, besides, halide substitution at ortho-site of phenyl ring could effectively increase the interaction

between ligand and perovskites (Figure 9a). Therefore, the size of PeQD could be adjusted continuously through the change of ligand concentration, and the short and conjugated ligand enabled the suitable coupling between PeQDs (Figure 9b). This strategy achieved high-performance LEDs with diverse emission wavelength based on different size and composition of PeQD (Figure 9c).

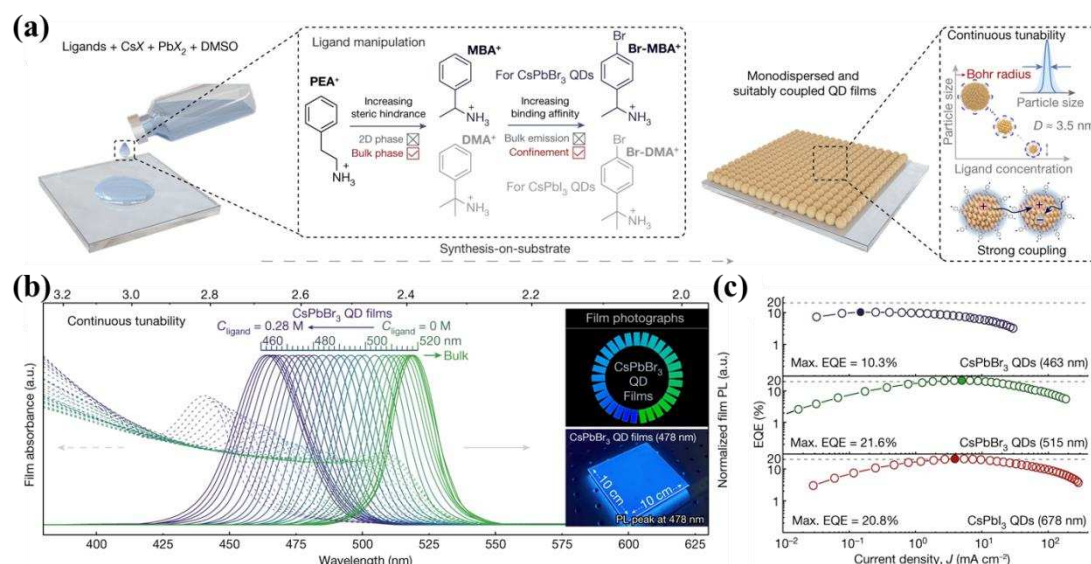


Figure 9. (a) Ligand designment for the in situ synthesis Monodispersed and suitably coupled PeQD on substrate, (b) Photoluminescence and absorption spectra of CsPbBr₃ QD films with different ligands concentrations, (c) The evolution of EQE with current density diverse color PeLEDs [134].

3. Progress in lead-free PeLEDs

Although near the milestone of 30% of EQE for lead-based PeLEDs has been reported, the toxicity of lead for humans and ecosystems has been a concern for a long time. Researchers have contributed tremendous efforts to find the reasonable substitution elements of lead without discounting the EQE and stability of PeLEDs [135]. As the same main group element, the ionic radius and valence electron configuration are similar to lead, hence, tin-based perovskites are viewed as a very ideal and eco-friendly substitution materials of emission layer for PeLEDs [136,137]. Friend et al. [138] demonstrated the application of CH₃NH₃SnI₃ for PeLEDs emitting at NIR in 2016 due to the smaller bandgap of CH₃NH₃SnI₃ than CH₃NH₃PbI₃. Meanwhile, with the increase of the Br ratio, the emission wavelength of PeLEDs could blue shift down to 667 nm, which widened the application of PeLEDs in sensing, medical devices and optical communication (Figure 10a). Despite some merits of tin-based perovskites mentioned above, the performance of their LEDs nowadays is far behind that of lead-based perovskite LEDs, the major reasons are the rapid crystallization kinetic and the easy oxidization of Sn²⁺ into Sn⁴⁺, leading to inferior films quality [139,140]. Huang et al. [141] investigated the crystallization process of tin-based perovskites via in-situ PL spectra, and they found fast clusters aggregation at the primary growth procedure was effectively retarded through the strong chemical interactions between SnI₂ and additives (PEAI and Vitamin B1). And then a good film quality was obtained with improved crystallinity and luminescence efficiency (Figure 10b). Finally, the best EQE of 8.3% was achieved in tin-based PeLEDs emitting at 894 nm (Figure 10c). Wang and coworkers [142] introduce cyanuric acid (CA) into the precursor solution of quasi-2D tin-based perovskites. The C=N in CA could strongly coordinate with Sn²⁺ to restrict the oxidization of Sn²⁺, and the hydrogen bond interaction between -OH and I⁻ reduced the loss of I⁻. In addition, the dimers or trimers for mixed keto and enol tautomers of CA were arranged orderly at the surface of perovskites, facilitating the perpendicular growth of perovskites (Figure 10d, e). These advantages enabled to gain of a 20.29% EQE of tin-based PeLEDs, which were comparable with the high-performance lead-based PeLEDs (Figure 10f).

The other alternative of lead-based perovskites is double perovskites, whose universal chemical formula of it is $A_2B^I B^{III} X_6$, where Pb^{2+} is replaced by a combination of monovalent metal cation (Na^+ , K^+ , Ag^+ , etc.) and trivalent metal cation (In^{3+} , Sb^{3+} , Bi^{3+} , etc.) [143,144]. The most widely studied double perovskites, like $Cs_2(Ag/Na/K)(Bi/In)X_6$, have not only lower toxicity, but also higher stability and carrier lifetime than those of lead-based perovskites [135,145]. Besides, these double perovskites can produce broad emission wavelength originating from the interaction between carrier and soft octahedron lattice, showing the prospective application of white LEDs based on a single emitter [146–148]. Tang et al. [149] employed the theory calculations to study the luminescent property of $Cs_2AgInCl_6$, and they found the parity-forbidden cause of the extremely small value of PLQY. To break the parity-forbidden, the Na^+ was incorporated into $Cs_2AgInCl_6$, which achieved a remarkable improvement of PLQY. By further little doping of bismuth, the fabricated electroluminescence device obtained $86 \pm 5\%$ of quantum efficiency with a long operational lifetime (exceeding 1000 h) (Figure 10g, h). Qu et al. [150] performed density functional theory calculations and found the Bi-doping also enabled the break of the parity-forbidden transition of $Cs_2AgInCl_6$ to ameliorate the PLQY, therefore, $Cs_2AgIn_{0.9}Bi_{0.1}Cl_6$ quantum dots were synthesized and used the materials as the emitter for LEDs. The optimized white LEDs had a 0.08% EQE with competitive brightness and long-term stability (Figure 10i).

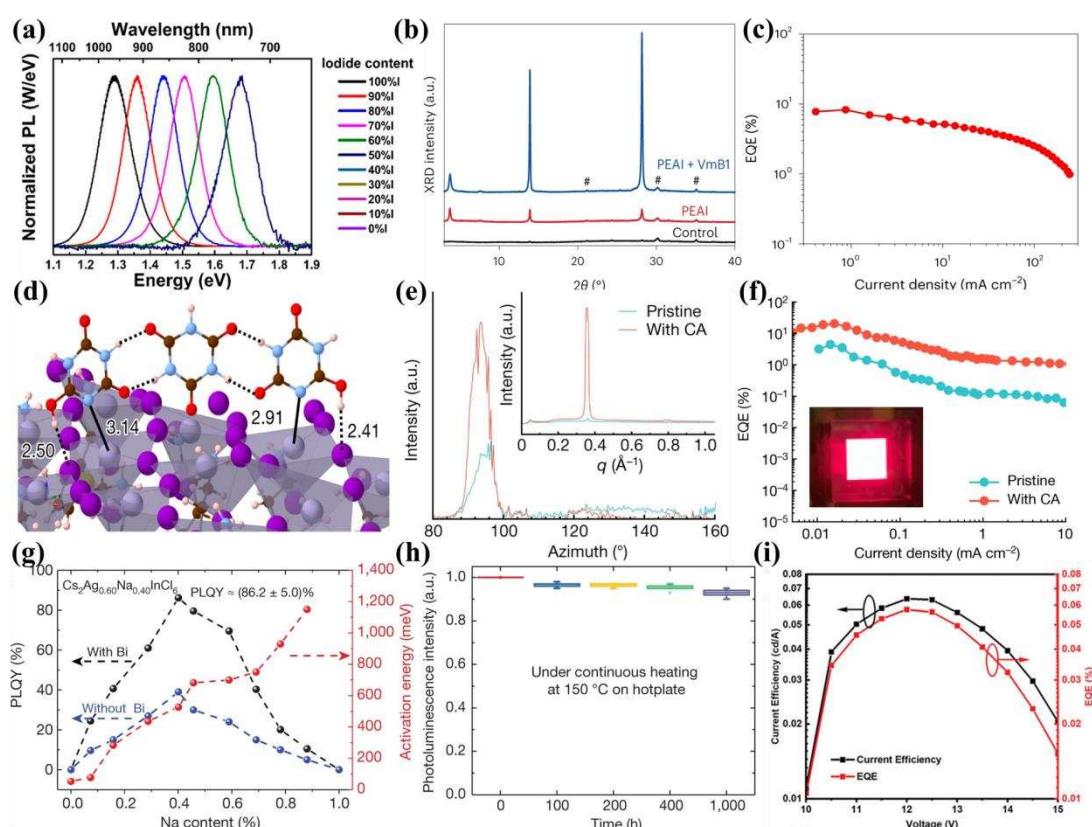


Figure 10. (a) The evolution of PL spectra of perovskite with change of iodide content [138], (b) XRD patterns of perovskites film with different additives. # indicates the diffraction data from the ITO substrate [141], (c) The curve of EQE with the change of current density for PeLEDs [141], (d) The simulated stable configurations for the tautomeric CA trimer on the surface of tin-based perovskite [142], (e) Intensity curves of GIWAXS patterns along the (002) ring for the samples with and without CA (Inset: integrated intensity of GIWAXS patterns for the samples) [142], (f) The curve of EQE with the change of current density for PeLEDs with and without CA [142], (g) Dependence of activation energy and PLQY of $Cs_2Ag_xNa_{1-x}InCl_6$ with and without doping of Bi on Na content [149], (h) The evolution of PL intensity of $Cs_2Ag_{0.60}Na_{0.40}InCl_6$ under continuous heating at 150 °C on a hotplate, measured after cooling to room temperature [149], (i) The evolution of EQE and current efficiency on the increase of driving voltage [150].

4. Challenges for PeLEDs

4.1. Poor devices operational stability

The perfect operational stability is the basic requirement for the practical application of PeLEDs, compared with the lifetime of organic LEDs ($T_{50} > 9500000$ h at 1000 cd m^{-2}) and quantum dots LEDs ($T_{50} = 125000000$ h at 100 cd m^{-2}) [151,152], the record T_{50} of PeLEDs at original brightness of 100 cd m^{-2} is only 2500 h [153]. The instability mainly originates from the perovskite layer as well as its top and buried interface [154]. The bulk perovskite materials are sensitive to some external factors. The temperature of PeLEDs will constantly rise with the consecutive running of devices, the increased temperature may cause the dissociation of excitons into free carriers, even worse, too high temperature can decompose the perovskites, especially for organic and inorganic hybrid perovskite with organic cation at A site [155–157]. The water and oxygen in the ambient may also influence the stability of PeLEDs. The water can migrate into the inside of the perovskite lattice via surface defect sites to form hydrates, the hydrates enable the formation of deep-level defects and decomposition of perovskites [31,158,159]. The oxygen can trap hole carriers to produce reactive superoxide, which may cause the oxidization of halide anion and degradation of organic A sites cation [27]. The electric field has a significant influence on the stability of PeLEDs by halide segregation, band bending, electrode corrosion, etc. [160–162]. Besides, a poor interface quality will also hurt the stable operation of PeLEDs, for example, high trap-state density at the interface leads to the restriction of charge transportation, and the accumulative charge at the interface may cause the degradation of perovskites layer [163].

4.2. EQE roll-off

The EQE roll-off is a general problem for all types of LED devices, especially in PeLEDs. It refers to the rapid decrease of EQE when LEDs are operated at a high current density, the major factors of EQE roll-off are unbalanced charge injection, Joule heating and Auger recombination [164–167]. The unbalanced charge injection can be ascribed to the inferior interface quality and inappropriate band alignment, and the unbalanced charge injection causes the carrier accumulation, leading to the aggravation of Auger recombination and Joule heating [68,154]. Besides, Auger recombination probability is proportional to excitons binding energy, so there is severe Auger recombination in strong-confinement PeQD and quasi-2D perovskites [168,169].

4.3. Low EQE for blue PeLEDs

Although red and green PeLEDs nowadays have achieved exceeding 20% of EQE, the EQE for blue counterparts (emission wavelength $< 470 \text{ nm}$) is very low ($\sim 12\%$). Mixed Cl-Br perovskites are the most straightforward materials for blue emission, they, however, take place notorious ion migration and phase separation, especially under high bias voltages, leading to the reduction of EQE and the shift of emission wavelength [131,170,171]; Another candidate is low-n phase ($n=1, 2$) quasi-2D perovskites due to their special “quantum well” structure, whereas, there are strong electron-phonon coupling interaction in low-n phase quasi-2D perovskites, which undermine the luminescence efficiency of PeLEDs [172]; In addition, CsPbBr₃ quantum dots with ultra-small size can also realize the blue light emission according to the quantum confinement effect, nevertheless, the synthesis of the ultra-small-size quantum dots is difficult without compromise the stability and efficiency, because the smaller the quantum dots are, the larger the surface-to-volume ratio is, so higher ligand density are needed, resulting in the inefficient carrier injection and undesirable EQE for PeLEDs [39,173,174].

5. Summary and outlook

This review summarizes the recent progress in the application of lead-based perovskite materials on LEDs. Various strategies have been developed to improve the performance and long-term storage and operational stability of PeLED devices, including additive engineering, interfacial engineering, dimensionality control, device structure optimization, etc., which make PeLEDs show a

comparable EQE with advanced organic and quantum dot LEDs. Here we provide some perspectives on the future development in this interesting field.

1. Considering the commercial application, the further breakthrough in the performance of PeLEDs is needed, especially in operational stability. Polymer materials may act as good additives for the enhancement of optoelectronic properties and stability of the perovskite emission layer [175,176]. On the one hand, the polymers can fill the vacancy at the grain boundaries of perovskites due to their rich functional groups, and the interaction between these functional groups and halide anion or Pb^{2+} enables the modulation of orientation and crystallization kinetics of perovskites [177–179]. Besides, the polymer network can also protect the perovskites against ion migration and degradation induced by moisture, temperature, and other external stimulation [76]. The extra insertion of the passivator layer is a prevalent method to modify the charge injection and interface quality, it, however, increases the complexity of device fabrication. Therefore, the reasonable design of novel charge transportation materials is desirable to achieve efficient carrier injection and reduce trap-state density at the surface of perovskites, in addition, a suitable selection of charge transportation materials benefits heat dissipation and outcoupling efficiency improvement [72,130].

2. Despite the significant success in red and green PeLEDs, there is still a challenge in the blue counterparts. To broaden the application of PeLEDs in display and luminance, further optimization of blue PeLEDs with a satisfying performance is necessary. Firstly, suitable additives are key to inhibiting the formation of Cl vacancy defect and phase separation in the mixed Br/Cl perovskites emitter. Alkali metal ions are potential candidates to fix the halide anion via strong ion bond [35,180], and conjugated benzene ring with sulfonate and phosphate group can bind with undercoordinated Pb^{2+} effectively and modulate the crystallization process of perovskites [134,181]. Hence, the choice of additives based on these considerations is recommended. As for pure bromide-based PeQD emitter, some surface ligands with the characteristics mentioned above are also beneficial for efficient charge injection. Besides, there is a requirement to develop novel hole transportation materials due to inefficient hole injection originating from unmatched band alignment [182–184].

3. Lead is toxic to humans, animals and the environment, hence, finding safe and high-performance lead-free PeLEDs is also important. Nowadays, tin-based PeLEDs are most likely alternatives for lead-based counterparts, however, rapid crystallization and easy oxidation of tin-based perovskites tend to cause the formation of deep-level defects (like Sn^{4+}). Organic molecules possessing appropriate coordination ability with Sn^{2+} are always used to retard the crystallization kinetics and suppress oxidation of Sn^{2+} of tin-based perovskites, combined with the little addition of reductive reagent, fine tin-based perovskite films will be obtained to achieve efficient PeLEDs [141,142].

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