

Strategies Toward Efficient Blue Perovskite Light-Emitting Diodes

Zhenwei Ren, Kai Wang, Xiao Wei Sun,* and Wallace C. H. Choy*

Substantial progress has been made in blue perovskite light-emitting diodes (PeLEDs). In this review, the strategies for high-performance blue PeLEDs are described, and the main focus is on the optimization of the optical and electrical properties of perovskites. In detail, the fundamental device working principles are first elucidated, followed by a systematical discussion of the key issues for achieving high-quality perovskite nanocrystals (NCs) and quasi-2D perovskites. These involve ligand optimization and metal doping in enhancing the carrier transport and reducing the traps of perovskite NCs, as well as the perovskite phase modulation and defect passivation in improving energy transfer and emission efficiency of quasi-2D perovskites. The strategies for efficient 3D mixed-halide perovskite and lead-free perovskite blue LEDs are then briefly introduced. After that, other strategies, including effective charge transport layer, efficient perovskite emission system, and effective device architecture for high light outcoupling efficiency, are further discussed to boost the blue PeLED performances. Meanwhile, the testing standard of blue PeLED lifetime is suggested to enable the direct comparisons of the device operational stability. Finally, challenges and future directions for blue PeLEDs are addressed.

Specifically, high external quantum efficiencies (EQEs) exceeding 20% have been achieved for green, red, and near-infrared perovskite LEDs (PeLEDs),^[5–7] which are comparable with the organic LEDs (OLEDs) and quantum dot LEDs (QLEDs), showing a great potential toward practical applications. However, the blue PeLED performances still lag far behind with the green and red PeLED counterparts,^[8–12] which undoubtedly hinder the PeLED applications in full-color displays. Therefore, there is an urgent need to improve blue PeLED performance.

Recently, substantial efforts have been made to develop blue light emitted perovskites, such as perovskite nanocrystals (NCs), 2D perovskite nanoplatelets (NPLs), quasi-2D perovskites, and 3D mixed-halide polycrystalline perovskites.^[13–15] Among the blue perovskite emitters, there is only moderate development of the blue PeLED performance based on 2D perovskite NPLs. The room-temperature

2D nanoplate PeLEDs with an electroluminescence (EL) peak at 410 nm were first reported in 2016 by Liang et al. with use of (PEA)₂PbBr₄ NPLs, where phenethylamine (PEA) is 2-phenylethylammonium.^[9] The 2D NPL PeLEDs showed a narrow EL peak with a full width at half-maximum (FWHM) of 14 nm, while the efficiency of 0.04% is unsatisfied which can be attributed to the inefficient charge injection derived from the insulating ammonium ligands and the fast non-radiative decay induced by the strong electron–phonon interactions of 2D perovskite.^[9,16] In addition, difficulties are usually encountered

1. Introduction

Light-emitting diodes (LEDs) have been widely adopted in daily used illumination and displays. Meanwhile, the market is looking for new display technologies with high color purity and wide color gamut. In recent years, metal–halide perovskite materials have emerged as very competitive candidates for LEDs due to their excellent optical properties, such as the narrow emission linewidths, flexible and wide bandgap tuning, broad color gamut, and high photoluminescence quantum yield (PLQY).^[1–4]

Dr. Z. Ren, Prof. W. C. H. Choy
Department of Electrical and Electronic Engineering
The University of Hong Kong
Pokfulam Road, Hong Kong, China
E-mail: chchoy@eee.hku.hk

Prof. K. Wang, Prof. X. W. Sun
Key Laboratory of Energy Conversion and Storage Technologies
Ministry of Education and Department of Electrical and Electronic Engineering
Southern University of Science and Technology
Shenzhen 518055, China
E-mail: sunxw@sustech.edu.cn

Prof. K. Wang, Prof. X. W. Sun
Guangdong University Key Laboratory for Advanced Quantum Dot Displays and Lighting
Guangdong-Hong Kong-Macao Joint Laboratory for Photonic-Thermal-Electrical Energy Materials and Devices
Southern University of Science and Technology
Shenzhen 518055, China
Prof. W. C. H. Choy
Guangdong-Hong Kong-Macao Joint Laboratory for Photonic-Thermal-Electrical Energy Materials and Devices
Shenzhen 518055, China

 The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/adfm.202100516>.

DOI: 10.1002/adfm.202100516

Table 1. Summary of blue PeLED performances based on 2D NPI perovskites.

Type ^{a)}	Perovskites	Device structure	EL Peak [nm]	Luminance [cd m ⁻²]	EQE [%]	Year	Ref.
2D NPIs	PEA ₂ PbBr ₄	ITO/PEDOT/Pero/TPBi/Ca/Al	410	–	0.04	2016	[9]
	MAPbBr ₃ NPIs	ITO/PEDOT/PVK/Pero/TPBi/LiF/Al	456	<2	0.03	2016	[22]
			492	<10	0.3		
	CsPbBr ₃ NPIs	ITO/PEDOT/PolyTPD/Pero/TPBi/Ca/Ag	464	38	0.057	2018	[18]
	CsPbBr ₃ NPIs	ITO/PEDOT/PolyTPD/Pero/TPBi/LiF/Al	463	62	0.124	2018	[21]
	CsPbBr ₃ NPIs	ITO/PEDOT/PolyTPD/Pero/TPBi/LiF/Al	480	25	0.1	2018	[23]
	CsPbBr ₃ NPIs	ITO/PEDOT/PolyTPD/TFB/Pero/TPBi/Ca/Ag	464	40	0.3	2019	[24]
	CsPbBr ₃ NPIs	ITO/PEDOT/PolyTPD/Pero/TPBi/LiF/Al	469	41.8	1.42	2019	[19]
	PEA ₂ PbBr ₄	ITO/PEDOT/PVK/TAPC/Pero/TPBi/Ca/Ag	410	147.6	0.31	2019	[17]
	FAPbBr ₃ NPIs	ITO/PEDOT/PolyTPD/Pero/TmPyPB/LiF/Al	439	–	0.14	2020	[20]
	CsPbBr ₃ NPIs	ITO/PEDOT/PolyTPD/Pero/TPBi/LiF/Al	465	631	0.8	2021	[25]

^{a)}Abbreviations used: perovskite emission layer (Pero); PEDOT:PSS (PEDOT).

to prepare high-quality NPIs thin films from dilute suspension through the deposited by spin-coated approach. Deng et al. adopted an electric-field-deposition (EFD) for a dense and smooth NPI film, in which the (PEA)₂PbBr₄ NPIs were directionally driven to the positive poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS)/indium tin oxides (ITO) electrode under the applied electric field. With this effective deposition, the NPI PeLED efficiency increased to 0.31% with a luminance of 147.6 cd m⁻².^[17] In addition, the CsPbBr₃ NPIs were also employed to fabricate blue PeLEDs and the EL peaks can be tuned from 432 and 497 nm by varying the NPI thickness.^[18–20] Although the high PLQY near 100% was reported for CsPbBr₃ NPI solution via the PbBr₆^{4–} octahedra passivation strategy, the devices show a poor efficiency of 0.124%, which may be ascribed to the unfavorable device structure and insulated surface ligands causing inefficient charge injection.^[21] The blue PeLED performances based on 2D NPIs are summarized^[9,17–25] and listed in **Table 1**; more efficient ways are needed to make larger breakthroughs in the device performances.

Very recently, the blue emitters focused on perovskite nanocrystals, quasi-2D perovskites, and 3D mixed-halide perovskites have experienced extensive attention, which render boom in device efficiencies of NC,^[10,11] quasi-2D,^[26–30] and 3D mixed-halide^[8,31,32] blue PeLEDs. This is especially for the low-dimensional perovskites of perovskite NCs and quasi-2D perovskites, which have larger exciton binding energies and stronger charge carrier confinement than the 3D mixed-halide perovskite analogs due to the dielectric property of the organic ligands or spacer cations, which favor high radiative recombination rate and thus the efficient light-emission.^[33–35] Moreover, it is relatively easy to achieve good film morphology with high coverage and low roughness for perovskite NCs and quasi-2D perovskites comparing with the 3D polycrystalline perovskite owing to the organic ligands or organic ammonium cations assisted film formation, which benefits from low leakage current for high device performance.^[36,37]

In this review, we mainly focus on the very competitive blue emitters of perovskite NCs and quasi-2D perovskites and systematically discuss the strategies for high NC and quasi-2D

blue PeLED performances. We first briefly elucidate the device working principle. Then, we comprehensively describe the evolutions and improvements of perovskite NC and quasi-2D perovskite optoelectronic properties and elaborate the key issues including (1) the ligand optimization and metal doping for high-quality perovskite NCs with efficient charge transport and suppressed traps, and (2) the perovskite phase modulation and passivation for bright blue-emissive quasi-2D perovskite with high color purity, fast energy transfer, and bright emission. Meanwhile, we also discuss the strategies for efficient 3D mixed-halide perovskite and lead-free perovskite blue LEDs. After that, we further discuss other important perspectives including effective transport layers, efficient emission systems, and effective device structure for light out-coupling, for promoting high-efficient PeLEDs in the future researches. In addition, we also suggest a testing standard of blue PeLED lifetime to enable the direct comparisons of the device operational stability. Finally, challenges and future directions of blue PeLEDs for the practical applications are further addressed.

2. Blue PeLED Performance Improvement Strategies

2.1. Device Working Principle

The typical structure of blue PeLEDs is shown in **Figure 1a** with perovskite emission layer sandwiched between hole-injecting layer (HIL) and electron-injecting layer (EIL), which is similar to that of OLED and QLED analogues.^[38,39] When applying forward bias on the electrodes, holes and electrons are first injected into HIL and EIL, respectively. Then the injected charge carriers are drifted into the perovskite layer under the applied field, and the electroluminescence emerges when the charge carriers recombine radiatively in perovskite layer (**Figure 1b**). The PeLED performances are typically characterized by some factors of merit including the key parameters of EQE, luminance, and current efficiency,^[40] which can be measured by the integrating sphere system (**Figure 1c**).

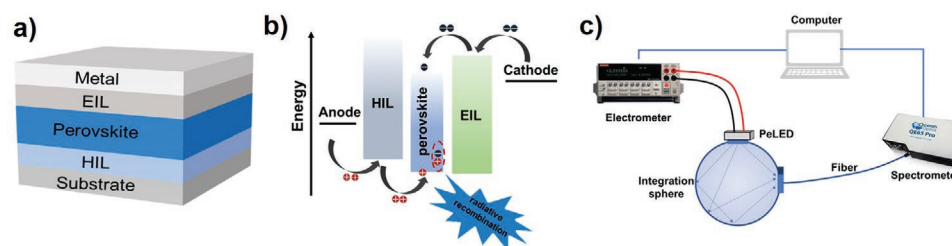


Figure 1. a) The typical device structure of blue PeLEDs, b) the working principle of blue PeLEDs, and c) the widely used integrating sphere system for PeLED characterizations.

2.2. NC Blue PeLEDs

2.2.1. Perovskite NC Blue Emitters

Generally, the perovskite NC blue emitters can be obtained through the perovskite composition and crystal size modulation. In 2015, Kovalenko and co-workers fabricated monodisperse CsPbX_3 ($X = \text{Cl}, \text{Br}, \text{I}$) NCs with tunable emission from the entire visible spectral region (Figure 2a,b), in which the blue emission can be reached by increasing the ratio of Cl ion in $\text{CsPbBr}_{3-x}\text{Cl}_x$ NCs.^[3] Donega and co-workers reported to partially replace Pb^{2+} of CsPbBr_3 NCs with smaller divalent cations of M^{2+} (Sn^{2+} , Cd^{2+} , and Zn^{2+}) for $\text{CsPb}_{1-x}\text{M}_x\text{Br}_3$ NCs and achieved a blue emission due to the lattice contraction (Figure 2c,d).^[41] In addition, exchanging Pb^{2+} with Al^{3+} and Sb^{3+} ions, could also obtain blue perovskite emitters in which the Al^{3+} doping resulted in an enlarged bandgap because of

the lattice contraction,^[42] while the Sb^{3+} doping could slow the growth of small CsPbBr_3 NCs due to the higher lattice energy of Sb^{3+} doped CsPbBr_3 NCs^[43]. Besides, a blue-shift of the emission peak was observed for Bi^{3+} doped CsPbBr_3 NCs at high-doping concentrations due to the introduced higher trap state energy level above conduction band.^[44] Furthermore, reducing the perovskite crystal size to near the Bohr radius is another facile strategy to realize the blue emission of perovskites due to the strong quantum confinement effect. Lee and co-workers showed a distinct quantum-size effect of MAPbBr_3 NCs when their sizes decreased to lower than 10 nm (Figure 2e,f). They reported that the PL spectra could be tuned to the blue area by reducing the NC size to below 10 nm and the PL peaks at 484 and 470 nm were obtained by reducing the NC size to 5 and 3 nm, respectively.^[45] Recently, adopting the strategy of size-tunable emission property has shown great potential toward bright blue perovskite NC emitters for efficient PeLEDs.

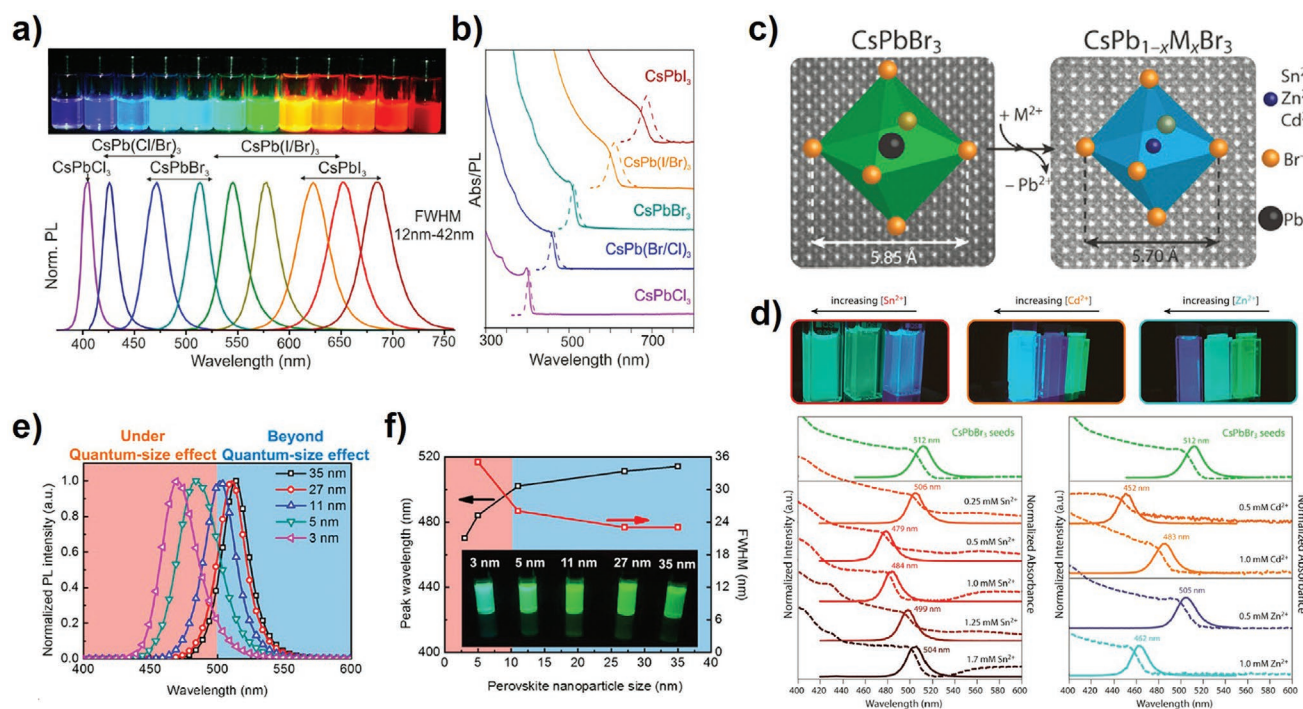


Figure 2. a) Colloidal perovskite CsPbX_3 NC ($X = \text{Cl}, \text{Br}, \text{I}$) dispersions in toluene under UV lamp ($\lambda = 365$ nm) and the corresponding PL spectra ($\lambda_{\text{exc}} = 400$ nm for all but 350 nm for CsPbCl_3 samples), b) Typical optical absorption and PL spectra. Reproduced with permission.^[3] Copyright 2015, American Chemical Society. c) Schematic diagram of partial cation exchange in CsPbBr_3 NCs, and d) The changes corresponding to their spectra. Reproduced with permission.^[41] Copyright 2017, American Chemical Society. e) PL spectra of perovskite NCs, f) PL peak wavelengths and FWHM of perovskite NCs and photograph of perovskite NPs under a $\lambda = 350$ nm Xe lamp (inset). Reproduced with permission.^[45] Copyright 2017, American Chemical Society.

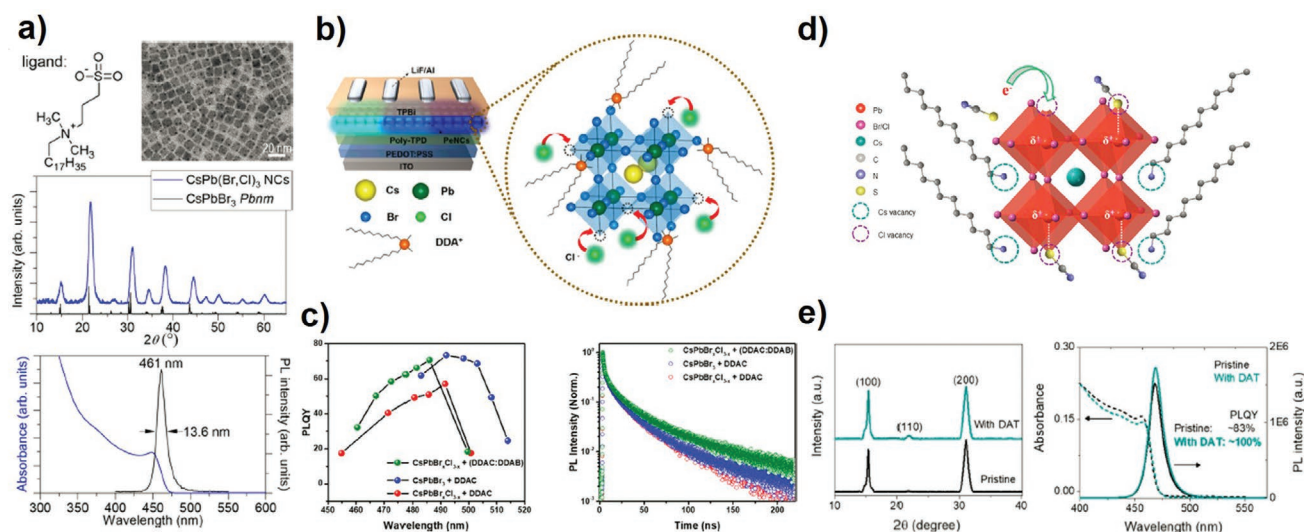


Figure 3. a) Structure of the long-chain zwitterionic sulfobetaine molecule, transmission electron microscopy (TEM) image, X-ray diffraction (XRD) pattern, and the corresponding absorption (blue line) and PL (black line) spectra of CsPb(Br,Cl)₃ NCs. Reproduced with permission.^[51] Copyright 2019, American Chemical Society. b) Illustration of the schematic CsPbBr₃Cl_{3-x} NCs-based blue PeLEDs and c) the PLQY and time resolved PL lifetime characteristics. Reproduced with permission.^[52] Copyright 2019, American Chemical Society. d) Illustration of organic thiocyanate passivation of perovskite NCs and e) XRD patterns, steady-state optical absorption, and PL spectra of pristine CsPbBr₃Cl_{3-x} NCs and DAT-treated NCs. Reproduced with permission.^[54] Copyright 2020, American Chemical Society.

Sargent et al. prepared strongly confined CsPbBr₃ NC solids by resurfacing the nanocrystals with bipolar-shell, in which the NCs exhibited efficient emission with tunable EL peaks from green to blue by reducing the NC size from 7 to 4 nm^[11] (details will be discussed in Section 2.2.2). Similarly, Tian et al. reported to achieve pure-blue emission (EL peak: 470 nm) using a small sized CsPbBr₃ NCs of around 4 nm. The NCs showed good optoelectronic properties in preparation of efficient PeLEDs with high brightness and robust durability^[46] (details will be discussed in Section 2.2.2). In addition, Wang et al. reported an antisolvent treatment assisted in situ formation of perovskite nanocrystals with a facile spin-coating method, in which small Cs_xMA_{1-x}PbBr₃ NCs (≈4.1 nm) with efficient deep-blue emission of 464 nm (PL peak) were achieved by a delay time of the antisolvent treatment^[47] (details will be discussed in Section 3.2).

2.2.2. Perovskite NC Ligand Optimization

The first NC blue PeLEDs were fabricated by Zeng and co-workers in 2015 with the use of CsPb(Cl/Br)₃ NCs. The unoptimized PeLEDs show a poor performance with an EQE of 0.07% at 455 nm,^[10] which needs further improvements. For NC PeLEDs, the perovskite NC surface-capping ligands play a crucial role for the device performance.^[48] On the one hand, mass ligands are needed to provide sufficient surface passivation for less surface defects and high PLQY. While excessive ligands, such as oleylamine (OAm) and oleic acid (OA) can form an insulating layer around the perovskite NC surface that degrades electric conductivity and thus, device efficiency. Consequently, the optimization of perovskite NC ligands for less traps and efficient charge transport is essential to achieve high device performances.

Ligand Optimization for Less Traps: In the synthetic process of perovskite NCs, OAm and OA are widely employed as surface-capping ligands.^[48–50] Although excessive amounts of OAm and OA are used to passivate the perovskite NC surface, there are still ligand losses during the purification process owing to the weak interaction between OAm (or OA) and perovskite NCs. This induces mass defect sites on perovskite NC surface for nonradiative recombination and deteriorates the device efficiency. To inhibit the ligand loss from the perovskite NCs, the chelating ligands with high affinity toward perovskite NC surface have been adopted recently. For example, Kovalenko and co-workers chose zwitterionic sulfobetaine of 3-(N,N-dimethyl-octadecylammonio) propanesulfonate as the surface ligand to fabricate CsPb(Br/Cl)₃ NCs (Figure 3a). The strong interaction between sulfobetaine and CsPb(Br/Cl)₃ NCs allowed for thorough purification of the NCs while leaving good PL intensity and colloidal integrity. Based on the zwitterionic sulfobetaine capped NCs, a device performance with an EQE of 1.2% at 463 nm was obtained.^[51] Besides capping perovskite NCs with high affinity ligands in the synthetic process, the post-treatment of perovskite NCs with compensatory ligands is another efficient way to suppress the perovskite NC surface defects for improved device performance. Shin et al. employed the mixed halide ion-pair ligands of di-dodecyl dimethyl ammonium bromide (DDAB) and di-dodecyl dimethyl ammonium chloride (DDAC) to post-treat the perovskite NCs and revealed that the perovskite NC surface ion vacancies can be effectively passivated with halide anions adsorbing into positive sites (Pb²⁺ and Cs⁺) and didodecyl dimethyl ammonium (DDA⁺) absorbing into the negative sites (Br⁻ and Cl⁻) (Figure 3b). Then, the PLQY and PL lifetime of the post-treated NCs were improved accordingly (Figure 3c), which enables blue PeLEDs with an EQE of 0.86% at 479 nm.^[52] Similarly, the post-treatment of the benzenesulfonate ligand (e.g., sodium dodecylbenzenesulfonate,

SDSA) was also adopted to passivate the $\text{CsPbBr}_x\text{Cl}_{3-x}$ NC surface defects through filling the halogen vacancies with the ionic sulfonate group. The post-treatment of SDSA greatly improves the PLQY of $\text{CsPbBr}_x\text{Cl}_{3-x}$ NC solution from 7% to 81% and promotes an EQE of 2.6% at 496 nm for the PeLEDs.^[53] Bakr and co-workers reported a decent passivation strategy by directly adding proper amount of *n*-dodecylammoniumthiocyanate (DAT) into the perovskite NC dispersion. The Cs and Cl vacancies of $\text{CsPb}(\text{Br}_x\text{Cl}_{1-x})_3$ NCs could be saturated by *n*-dodecylammonium and thiocyanate groups, respectively (Figure 3d). As a result, a high PLQY near 100% was achieved for DAT-treated $\text{CsPb}(\text{Br}_x\text{Cl}_{1-x})_3$ NCs (in solution), which benefited the PeLEDs of a high EQE of 6.3% at 470 nm.^[54] Recently, Tian et al. introduced the novel strategy of acid etching-driven ligand exchange for the preparation of bright emissive pure-blue CsPbBr_3 NCs. They employed hydrogen bromide (HBr) to etch imperfect octahedrons with its strong acidity to promote the protonation of OAm and desorption of OA^- . In the etching process, the imperfect octahedrons and poorly crystalline NCs were removed while leaving halide-rich condition on the NCs surface for the suppression of halide vacancies. Then, didodecylamine and phenethylamine were successively introduced to further exchange with the original organic ligands and bond the residual uncoordinated sites for low trap density and high PLQY up to 97%. As a result, the fabricated PeLEDs exhibited decent performance with an EQE of 4.7%, a high luminance of 3850 cd m^{-2} at 470 nm, and a very long half-lifetime exceeding 12 h under continuous operation.^[46]

Ligand Optimization for Efficient Charge Transport: The insulating nature of long-chain organic ligands (e.g., OAm, OA) degrades the charge transport in PeLEDs. In order to facilitate carrier transport in $\text{CsPbBr}_3\text{Cl}_{3-x}$ NC film, Pan et al. first adopted a short-chain ligand of DDAB to replace the long-chain OAm and OA ligands through a two-step ligand-exchange procedure (Figure 4a) to desorb protonated OAm while avoiding the formation of 2D quantum wells of $(\text{OAm})\text{PbBr}_4$.^[55] Benefitting from the increased carrier transport in the DDAB-treated perovskite NC film, blue PeLEDs with an EQE of 1.9% and a luminance of 35 cd m^{-2} at 490 nm were achieved (Figure 4b). Nevertheless, the strategy of direct replacement of long-chain ligands with short-chain counterparts may lead to notable changes in NC's sizes and photophysical properties.^[56,57] In view of this, Shynkarenko et al. reported direct synthesis of CsPbX_3 NCs with the short-chain ligand of DDAC and achieved a uniform cuboid morphology of $\text{CsPb}(\text{Br}_{1-x}\text{Cl}_x)_3$ NCs with an average crystal size of $\approx 9 \text{ nm}$. With the sole short-chain ligand for efficient charge transport, they achieved an EQE of 3.5% with a luminance of 2063 cd m^{-2} at 490 nm.^[58] Besides, Zhang et al. demonstrated a well-controlled mild ligand exchange process on the NC's surface through the reaction of thionyl halides (SOCl_2 , SOBr_2) with the carboxylic and amine groups, in which most organic ligands were removed and simultaneously preserved the nanocrystal emission characteristics (Figure 4c).^[59] Consequently, they fabricated organic-ligand-lacking halide perovskite NCs and achieved well-performed deep blue PeLEDs with an EQE of 1.35% at 460 nm and an extremely narrow FWHM of 14.6 nm. In order to further reduce the insulating ligands, Sargent and co-workers have developed a bipolar-shell-resurfacing approach to substitute the long organic ligands with

atomic ligands, which improved perovskite NC coupling and charge transport, while keeping an excellent colloid stability of NCs.^[11] The ligand exchange process mainly consisted of two steps. The isopropylammonium (IPA) bromide was first added into the purified NC dispersion to form a Br^- -rich surface with the IPA cation left in dimethylformamide (DMF) solvent as an outer shell. Consistent with this, a high electrokinetic positive (ζ potential) of $\approx 90 \text{ mV}$ was exhibited due to the positive charge of IPA cation. Then, the IPA group was further replaced with Na ion by adding NaBr-saturated DMF solution for another ligand exchange, and Na ion enriched outer shell was finally formed as indicated by the positive ζ potential of $\approx 40 \text{ mV}$. Based on the advanced ligand exchange strategy for excellent perovskite NC properties, the carrier mobility reached over $0.01 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. Finally, they achieved efficient PeLED performance with an EQE of 12.3% at 479 nm, which represents the most efficient NC blue PeLED at present.

2.2.3. Metal Ion Doping Perovskite NCs

Although fine optimization of the synthetic conditions (e.g., precursors, reaction temperature, and stoichiometry) can reduce the defects, the imperfect assembly of constituents still occurs during the formation of perovskite NCs. Especially for chloride based perovskite NCs (e.g., $\text{CsPb}(\text{Br}/\text{Cl})_3$ NCs), the low formation energy of defects further aggravates the imperfections of crystals and increases the population of defects during crystallization process. Metal doping is another efficient strategy to achieve high-quality perovskite NCs with less defects, in which the doped ions can merge into the crystal lattice to suppress the structure disorder (octahedra distortion) without changing the crystals' size.^[60] The metal ions doped perovskite NCs, such as $\text{Mn}^{2+}/\text{Zn}^{2+}$ -doped CsPbI_3 , $\text{Mn}^{2+}/\text{Sn}^{2+}/\text{Ce}^{3+}$ -doped CsPbBr_3 have shown great potential for high red and green PeLED performances, respectively.^[61–64] Meanwhile, substantial works of metal doped blue emissive perovskite NCs have also been made. For example, in order to suppress the defects produced during CsPbCl_3 NC formation process, Sun and co-workers introduced nickel (Ni) ions into the NCs to form Ni-doped CsPbCl_3 NCs, which increases the defect formation energy of the NCs and improves the short-range order of the perovskite lattice. As a result, the intrinsic defects such as halide vacancies of Ni- CsPbCl_3 NCs were substantially eliminated with the nonradiative recombination rate reduced from 378.29 to $1.90 \mu\text{s}^{-1}$ and a maximum solution PLQY increased up to 96.5%.^[65] Then, the Ni ion doped $\text{CsPbCl}_x\text{Br}_{3-x}$ NCs were employed as an emission layer of the PeLEDs and an EQE of 2.4% and a maximum luminance of 612 cd m^{-2} at 470 nm were obtained.^[66] Besides, manganese (Mn) ion is another efficient dopant to improve photoluminescence yield of the perovskite NCs for bright emitters. However, it is noted that Mn^{2+} doped NCs are prone to show an additional PL peak at orange-red region due to energy transfer from the exciton to Mn^{2+} ion for Mn emission (Figure 5a).^[67,68] In this case, Hou et al. carefully tuned the Mn-doping concentration to increase the NC solution PLQY by over threefold while maintaining a pure emission peak (Figure 5b,c). Benefitting from the bright emitter of Mn^{2+} -doped $\text{CsPb}(\text{Br}_x\text{Cl}_{1-x})_3$ NCs, deep blue PeLEDs with

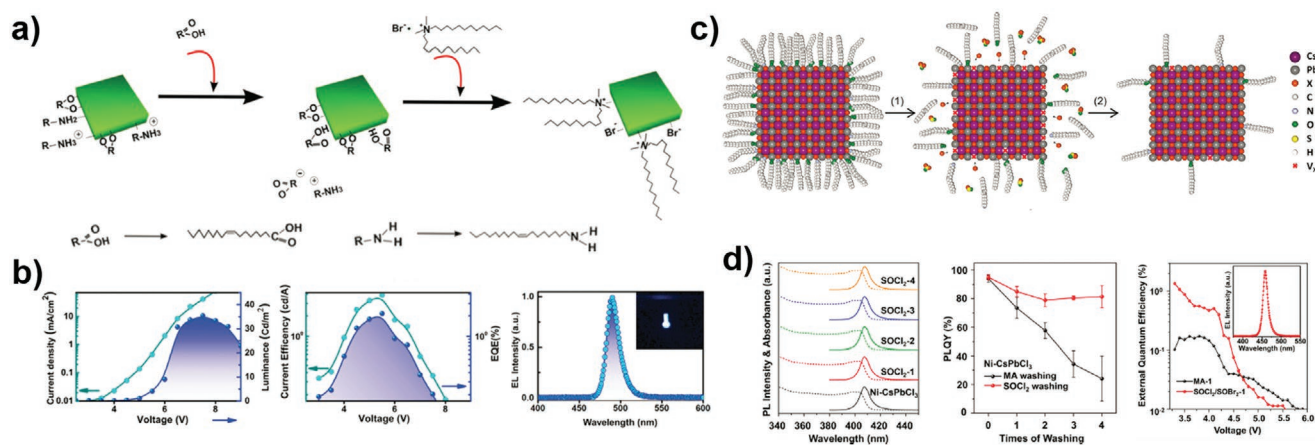


Figure 4. a) The ligand-exchange mechanism on perovskite NC surfaces, and b) the PeLED device performance: Current density and luminance, current efficiency, and EQE versus driving voltage characteristics, and EL spectrum at 5.5 V, and inset, a photograph of a device. Reproduced with permission.^[55] Copyright 2016, Wiley-VCH. c) Schematic of the mild reaction and ligand-exchange process. d) Normalized absorption and PL spectra of Ni-CsPbCl₃ NC dispersion after different times of ligand exchange; PLQYs of Ni-CsPbCl₃ NCs solution after washing by methyl acetate or SOCl₂; EQEs of the methyl acetate and SOCl₂/SOBr₂ LEDs. Inset is the EL spectrum at 4 V. Reproduced with permission.^[59] Copyright 2019, American Chemical Society.

an increased EQE from 0.50% (undoped) to 2.12% at 466 nm were fabricated (Figure 5d).^[68] Nevertheless, for mixed halide CsPb(Br_xCl_{1-x})₃ NCs, there is a concern about the phase separation in the mixed halide perovskites due to the ion migration, especially for the operating device; the chance of ion migration increases under the applied electric field and thus degrades the device's spectral stability.^[69,70] Recently, Zhang et al. has shown that the ion migration can effectively be alleviated by doping CsPb(Br_xCl_{1-x})₃ NCs with lanthanum (La) ion owing to strong interaction between La and Cl ions. Moreover, the contraction of octahedral occurs once the Pb ion (ionic radius, 1.19 Å) is partially replaced by smaller La ion (1.032 Å), which further enhances the Pb-Cl interaction for the anchoring of Cl ions (Figure 5e). Encouragingly, the red-shifts of EL spectra under higher voltage were largely suppressed for La³⁺-doped CsPb(Br_xCl_{1-x})₃ PeLEDs where merely little red-shift of 3 nm

was observed with varying the voltages from 5 to 8 V much less than that of 12 nm for the undoped NCs (Figure 5f). Meanwhile, La³⁺-doped CsPb(Br_xCl_{1-x})₃ NCs also show higher carrier temperature and exciton binding energy, which favors a high NC solution PLQY of 84.3% and thus an improved device efficiency of 3.25% at 489 nm.^[71]

2.3. Quasi-2D Perovskite Blue LEDs

2.3.1. Quasi-2D Perovskites

Similar to the rapid development of NC blue PeLEDs (Table 2),^[10,11,47,51-55,66,58,59,68,72-77] quasi-2D perovskite blue LEDs have also experienced substantial progress recently (Table 3). Quasi-2D perovskites have good properties of

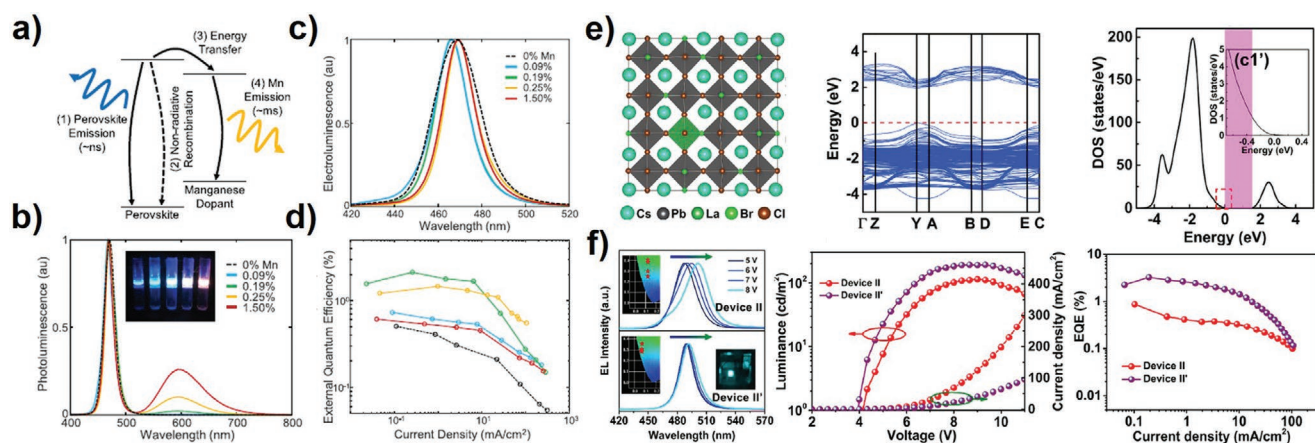


Figure 5. a) Energy diagram of the Mn²⁺ doped nanocrystal system, b) PL and c) EL spectra with nanocrystals of varying Mn content, and d) the EQE characteristics. Reproduced with permission.^[68] Copyright 2018, Elsevier Inc. e) Schematic diagram, energy band structure, and DOS of La³⁺-doped CsPb(Br_xCl_{1-x})₃ NCs. f) EL spectra, CIE, the device lighting photographs, current-voltage-luminescence (J-V-L), and EQE of Device II (undoped), II' (La³⁺-doped) CsPb(Br_xCl_{1-x})₃ NCs. Reproduced with permission.^[71] Copyright 2020, Elsevier Ltd.

Table 2. Summary of blue PeLED performances based on perovskite NCs.

Perovskites	Device structure	EL Peak [nm]	Luminance [cd m ⁻²]	EQE [%]	Year	Ref.
CsPb(Cl/Br) ₃	ITO/PEDOT/PVK/Pero/TPBi/LiF/Al	455	742	0.07	2015	[10]
CsPbBr _{1.5} Cl _{1.5}	ITO/ZnO/Pero/TFB/MoO ₃ /Al	480	1559	0.0074	2016	[74]
CsPbBr _x Cl _{3-x}	ITO/PEDOT/PVK/Pero/TPBi/LiF/Al	490	35	1.9	2016	[55]
CsPbBr _x Cl _{3-x}	ITO/NiO _x /Pero/TPBi/LiF/Al	470	350	0.07	2017	[75]
CsPbBr _x Cl _{3-x}	ITO/PEDOT/TFB/PFI/Pero/TPBi/LiF/Al	469	111	0.5	2018	[76]
CsMn _y Pb _{1-y} Br _x Cl _{3-x}	ITO/PEDOT/TFB/PFI/Pero/TPBi/LiF/Al	466	245	2.12	2018	[68]
(Rb _{0.33} Cs _{0.67}) _{0.42} FA _{0.58} PbCl _{1.25} Br _{1.75}	ITO/Poly-TPD/Pero/TPBi/Ba/Al	466	40	0.61	2019	[72]
CsPbBr _x Cl _{3-x}	ITO/PEDOT/Poly-TPD/Pero/TPBi/LiF/Al	479	29.9	0.86	2019	[52]
CsPb(Br/Cl) ₃	ITO/PEDOT/PolyTPD/CBP/Pero/B3PYMPM/LiF/Al	463	318	1.2	2019	[51]
CsPbBr _x Cl _{3-x}	ITO/PEDOT/pTPD/Pero/B3PYMPM/TPBi/LiF/Al	490	2063	3.5	2019	[58]
Ni ²⁺ -CsPbCl _{1.7} Br _{1.3}	ITO/PEDOT/Poly-TPD/Pero/TPBi/LiF/Al	460	—	1.35	2019	[59]
CsPbBr _x Cl _{3-x}	ITO/PEDOT/Poly-TPD/ PFN/Pero/TPBi/LiF/Al	470	46.7	1.34	2020	[77]
K ⁺ -CsPb(Br/Cl) ₃	ITO/PEDOT/Poly-TPD/ PVK/Pero/POT2T/LiF/Al	477	86.95	1.96	2020	[73]
Ni ²⁺ -CsPbCl _{0.99} Br _{2.01}	ITO/PEDOT/TFB/PFI/Pero/TPBi/LiF/Al	470	612	2.4	2020	[66]
Cs ₃ MA _{1-x} PbBr ₃	ITO/PEDOT/Poly-TPD/ PVK/Pero/TmPyPB/LiF/Al	465	<20	2.4	2020	[47]
CsPbBr _x Cl _{3-x}	ITO/PEDOT/Poly-TPD/ Pero/TPBi/Ca/Ag	496	603	2.6	2020	[53]
CsPb(Br _x Cl _{1-x}) ₃	ITO/TFB/PFI/Pero/3TPYMB/Liq/Al	470	465	6.3	2020	[54]
CsPbBr ₃	ITO/PEDOT/PTAA/Pero/TPBi/LiF/Al	479	—	12.3	2020	[11]
CsPbBr ₃	ITO/PEDOT/PVK/Pero/ZnO/Ag	470	3850	4.7	2021	[46]

uniform film morphology, cascade energy transfer, and high quantum efficiency, which will benefit for high-efficient blue PeLEDs. Generally, the quasi-2D perovskites have a formula of $B_2(APbX_3)_{n-1}PbX_4$, where B is an organic spacer cation (e.g., aliphatic, aromatic ammonium cation), A is a monovalent cation (e.g., Cs⁺, methylammonium (MA⁺), or formamindinium, (FA⁺)), X represents the halide anion (Br⁻, Cl⁻), and n is the number of lead halide octahedral layers ([PbX₆]⁴⁻).^[36] The octahedral layers ([PbX₆]⁴⁻) are sandwiched by adjacent two organic spacer cation layers to form the multiple-quantum-well structures, in which the [PbX₆]⁴⁻ sheet functions as the potential well, while organic spacer cation layers serve as the potential barriers (Figure 6). The generated excitons are confined within the octahedral layers due to the consequences of quantum and dielectric confinement effects. In quasi-2D perovskites, the value of n determines the bandgap (E_g), that is, high- n (low- n) corresponds to small (large) bandgap. Due to energy funneling effect, the energy transfer process is driven from a larger bandgap phase (i.e., lower- n) to a smaller bandgap (i.e., higher- n), and subsequently the excitons recombine in higher- n perovskite.^[35,78] To obtain blue emission in quasi-2D perovskites, the n number and component should be carefully controlled.

2.3.2. Quasi-2D Perovskite Phase Modulation for Efficient Blue PeLEDs

Quasi-2D perovskite phase modulation consists of the optimization of perovskite quantum-well and spacer (i.e., potential barrier) component collocation, which determines the perovskite optoelectrical properties, such as color purity, spec-

tral stability, and energy/charge transfer efficiency, and thus the device performance. Therefore, the efficient strategies for fine modulation of the perovskite phases are essential to achieve high PeLED performances.

Quantum-Well Component Modulation for Blue Emission: As for preparation of quasi-2D perovskite blue emitters, a lot of organic ammonium cations are used to narrow the potential well of octahedral layers. To gain deep blue emission, more organic ammonium cations are needed to obtain lower- n perovskites, because the lower- n perovskite phases consume more spacer cations than that of higher- n phases. For example, Yip and co-workers improved the concentration of 2-phenoxyethylamine (POEA) into CH₃NH₃PbBr₃ precursor to fabricate (POEA)₂(MA)_{*n*-1}Pb_{*n*}Br_{3*n*+1} quasi-2D perovskites with continuous blue-shifted PL spectra from 532 (without POEA) to 480 nm (40% POEA) and 462 nm (60% POEA). (Figure 7a).^[79] Congreve et al. doubled the stoichiometric ratio of butylammonium (BA) in BA₂(MAPbBr₃)_{*n*-1}PbBr₄ quasi-2D perovskite and reduced the $n > 3$ phase to $n = 1$ and $n = 2$ dominated phases. Consistent with this trend, the EL peaks were shifted from 500 to 440 nm (Figure 7b).^[80] Similar strategy was also adopted for ethylammonium bromide (EABr) based quasi-2D perovskite. The EL peaks of EA₂MA_{*n*-1}Pb_{*n*}Br_{3*n*+1} PeLEDs were shifted from 540 nm to two individual peak of 473 and 485 nm driven by the phase transition from bulk to $n = 3$ dominated phases with continuously increasing the ratio of EABr. (Figure 7c).^[27]

However, the strategy of mass organic spacers for blue quasi-2D emitters not only impedes the energy transfer causing multiple emission peaks, but also decreases carrier mobility resulting in charge accumulation and radiative recombination

Table 3. Summary of blue PeLED performances based on quasi-2D perovskite system.

Perovskites	Device structure	EL Peak [nm]	Luminance [cd m ⁻²]	EQE [%]	Year	Ref.
(4-PBA) ₂ PbBr ₄	ITO/ZnO/PEIE/Pero/TFB/MoO ₃ /Al	491	186	0.015	2017	[26]
BA ₂ (MA) _{n-1} Pb _n Br _{3n+1}	ITO/PEDOT/Pero/TmPyPB/LiF/Al	450	1	0.0054	2017	[80]
POEA ₂ MA _{n-1} Pb _n Br _{3n+1}	ITO/PEDOT/Pero/TPBi/LiF/Al	480, 494 462	19.25 1.26	1.1 0.06	2017	[79]
EA ₂ (MA) _{n-1} PbnBr _{3n+1}	ITO/PEDOT/Pero/TmPyPB/CsF/Al	473, 485	200	2.6	2017	[27]
IPA/PEA ₂ MACs _{n-1} Pb _n Br _{3n+1}	ITO/PEDOT/Pero/TPBi/LiF/Al	490	2480	1.5	2018	[85]
PEA ₂ Cs _{n-1} Pb _n (Br/Cl) _{3n+1}	ITO/NiO _x /LiF/Pero/TPBi/LiF/Al	490	1446	0.52	2019	[103]
PEA ₂ (Rb _x Cs _{1-x}) ₂ Pb ₃ Br ₁₀	ITO/PEDOT/Pero/TmPyPB/LiF/Al	475	100.6	1.35	2019	[81]
PA ₂ (CsPbBr ₃) _{n-1} PbBr ₄	ITO/NiO _x /PSSNa/PVK/Pero/TPBi/LiF/Al	492	5737	1.45	2019	[109]
P-PDA/PEA ₂ Cs _{n-1} PbnBr _{3n+1}	ITO/PVK/PFI/Pero/3TPYMB/Liq/Al	465	211	2.6	2019	[86]
PEA ₂ Cs _{n-1} Pb _n (Cl _x Br _{1-x}) _{3n+1}	ITO/PEDOT/Pero/TPBi/LiF/Al	480	3780	5.7	2019	[28]
BA ₂ Cs _{n-1} Pb _n (Br/Cl) _{3n+1}	ITO/PEDOT/PVK/Pero/TPBi/LiF/Al	487	3340	6.2	2019	[29]
PBABr _{1,1} (Cs _{0.7} FA _{0.3} PbBr ₃)	ITO/NiO _x /TFB/PVK/Pero/TPBi/LiF/Al	483	700	9.5	2019	[104]
PEACl:YCl ₃ :CsPbBr ₃	ITO/PEDOT/Pero/TPBi/LiF/Al	485	9040	11.0	2019	[82]
(BA _{0.5} PEA _{0.5}) ₂ MAPb ₂ Br ₇	ITO/NiO _x /F6TCNNQ/PVK/Pero/TPBi Cs ₂ CO ₃ /Al	456	—	0.015	2020	[87]
POEA-CsPbBr _{1.65} Cl _{1.35}	ITO/PEDOT/Pero/TPBi/LiF/Al	468	122	0.71	2020	[97]
(Cs/Rb/FA/PEA/K)Pb(Cl/Br) ₃	ITO/LiF/Pero/LiF/Bphen/LiF/Al	484	4015	2.01	2020	[102]
EDA/PEA ₂ Cs _{n-1} PbnBr _{3n+1}	ITO/PEDOT/Pero/TPBi/LiF/Al	474	290	2.17	2020	[91]
PEA ₂ DMA _m Cs ₂ Pb ₃ Br _{10+m}	ITO/PEDOT/Pero/TPBi/LiF/Al	490	2825	2.4	2020	[105]
PEA ₂ NPA ₁ Cs ₂ Pb ₃ Br ₁₂	ITO/PVK/Pero/PO-T2T/Liq/Al	482	1200	2.62	2020	[88]
PEA ₂ FA ₂ Pb ₂ Br ₇ Cl ₃	ITO/MPS2-TBA/Pero/TPBi/LiF/Al	489	311	2.64	2020	[106]
(BDAD)MA _{n-1} PbnBr _{3n+1}	ITO/PEDOT/Pero/TmPyPB/CsF/Al	494	440	2.7	2020	[107]
DPPA ₂ Cs _{n-1} Pb _n (Br/Cl) _{3n+1}	ITO/PEDOT/TFB/Pero/PMMA/TPBi/LiF/Al	475	547.5	3.03	2020	[108]
PPA ₂ MAPb ₂ Br ₇	ITO/NiO _x /F6TCNNQ/PVK/Pero/TPBi Cs ₂ CO ₃ /Al	480	286	3.35	2020	[93]
(Cs/FA/p-F-PEA)Pb(Cl/Br) ₃	ITO/PEDOT/Pero/PMMA/TPBi/LiF/Al	469	451	4.14	2020	[101]
PBA ₂ Cs _{n-1} Pb _n Br _{3n+1}	ITO/PEDOT/Pero/PO-T2T/LiF/Al	493	1000	5.08	2020	[110]
PEA ₂ Cs _{1.6} MA _{0.4} Pb ₃ Br ₁₀	ITO/PEDOT/Poly-TPD/Pero/TPBi/LiF/Al	479	468	5.2	2020	[98]
GABA-PEA ₂ Cs _{n-1} Pb _n Br _{3n+1}	ITO/PEDOT/PVK/Pero/TPBi/LiF/Al	478	200	6.3	2020	[94]
PEA _x PA _{2-x} (CsPbBr ₃) _{n-1} PbBr ₄	ITO/NiO _x /Pero/TPBi/LiF/Al	488	1711	7.22	2020	[89]
(BA _x PEA _{1-x}) ₂ Cs _{n-1} Pb _n (Br _{0.7} Cl _{0.3}) _{3n+1}	ITO/PEDOT/Pero/TPBi/Ca/Al	485	1130	7.84	2020	[90]
[(PEA) _{0.75} (GA) _{0.25}] ₂ CsPb ₂ (Br/Cl) ₇	ITO/PVK/F4TCNQ/Pero/TPPO/TPBi/LiF/Al	492	1003	8.2	2020	[99]
DPPA/PEA-CsPbClBr ₂	ITO/PEDOT/TFB/Pero/TPBi/LiF/Al	473	177	8.8	2020	[92]
Na/PEA ₂ Cs _{n-1} Pb _n (Cl _x Br _{1-x}) _{3n+1}	ITO/NiO _x /PTAA/PVK/ Pero/TPBi/LiF/Al	488	2060	11.7	2020	[30]
PEA ₂ (Cs _{1-x} EA _x PbBr ₃) ₂ PbBr ₄	ITO/PEDOT/Pero/TPBi/LiF/Al	488 495	2191 2790	12.1 13.3	2020	[12]
NEA-FAPbBr _{3-x} Cl _x	ITO/PEDOT/ Pero/B3PYMPM/LiF/Al	474	2810	3.1	2021	[111]
(ABA/PEA) _x PA _{2-x} (CsPbBr ₃) _{n-1} PbBr ₄	ITO/PVK/Pero/TPBi/LiF/Al	486	513	10.11	2021	[112]
PEACl:CsPbBr ₃ :YCl ₃	PEN/ITO/PEDOT/Pero/TPBi/LiF/Al	493	6600	12.8	2021	[113]

loss.^[27,79,80] In view of this, the simultaneous strategy of combining organic spacers with alloyed “A-site” or mixed halides was proposed to obtain the blue emissive quasi-2D perovskites. For example, the alkali metal ion of Rb⁺ was introduced to form rubidium–cesium alloyed PEA₂(Rb_xCs_{1-x})_{n-1}Pb_nBr_{3n+1} quasi-2D perovskite which can generate blue emissions in the whole blue spectral region ranging from 454 to 492 nm due

to the octahedral tilting of the perovskite structure by Rb ion (Figure 8a). The alloyed PEA₂(Rb_xCs_{1-x})_{n-1}Pb_nBr_{3n+1} showed an efficient energy transfer within the scale of ps from donor domains (low-*n*) to the emitting domain (high-*n*) phase. Together with the excess RbBr for higher perovskite film PLQY, the PeLEDs with an EQE of 1.35% at single EL peak of 475 nm and a narrow FWHM of ≈20 nm were achieved (Figure 8b).^[81]

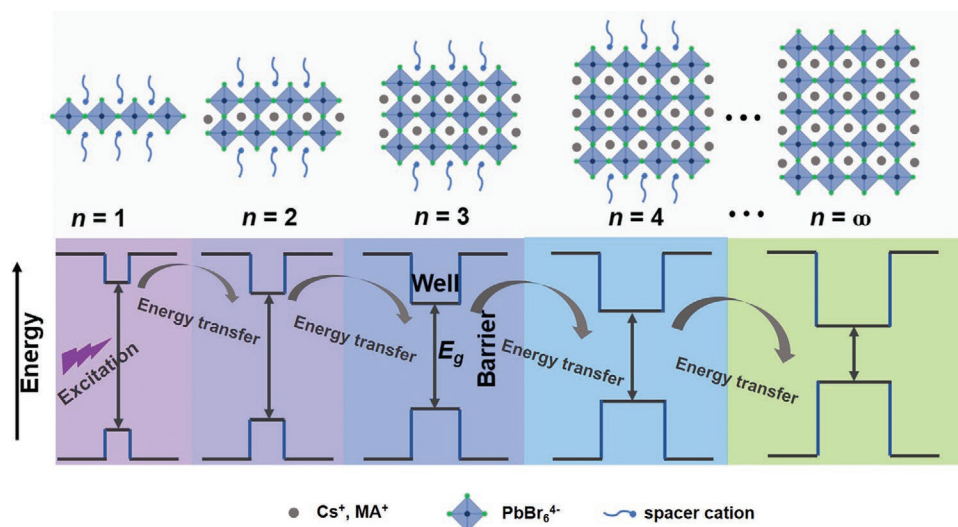


Figure 6. Schematic illustration of quasi-2D perovskites and the energy transfer process.

Recently, You and co-workers reported an enlarged bandgap of hybrid “A-site” $\text{PEA}_2(\text{Cs}_{1-x}\text{EA}_x\text{PbBr}_3)_2\text{PbBr}_4$ quasi-2D perovskite through introducing large cation $\text{CH}_3\text{CH}_2\text{NH}_2^+$ (EA^+) into $\text{PEA}_2(\text{CsPbBr}_3)_2\text{PbBr}_4$ perovskite. The first-principle calculations show that the formed $\text{Cs}_{1-x}\text{EA}_x\text{PbBr}_3$ sheet in the quasi-2D perovskite can affect the shortest Pb–Br orbital coupling and induces a lower valence band maximum (VBM) for a larger bandgap and thus, blue shifts of the PL peaks (Figure 8c). In addition, the introduction of EA^+ induces an increase of perovskite film PLQY from 42% to 70% due to the suppression of non-radiative recombination which contributes efficient sky-blue PeLEDs with a high EQE of 12.1% at 488 nm.^[12] Besides the “A-site” modulation, the mixed halide quasi-2D perovskites were also demonstrated as promising materials for high efficient

blue PeLEDs. For example, the mixed halides of Cl/Br ions were introduced to construct $\text{PEA}_2\text{Cs}_{n-1}\text{Pb}_n(\text{Cl}_x\text{Br}_{1-x})_{3n+1}$ perovskite in which the blue-emission region below 480 nm can be easily reached (Figure 8d). Meanwhile, there is an efficient energy transfer from the lower- n layered perovskite (2D perovskite) to the higher- n perovskite (3D perovskite) in the quasi-2D system (Figure 8e). With further modulation of the recombination zone to enlighten the majority of perovskite crystals, a high efficiency of 5.7% with the luminance of 3780 cd m^{-2} at single EL peak of 480 nm was achieved.^[28] Similar strategy was adapted to the butylammonium (BA) based quasi-2D $\text{BA}_2\text{Cs}_{n-1}\text{Pb}_n(\text{Br/Cl})_{3n+1}$ system, in which the number of CsPbBr_3 sheets was optimized at $\approx 24 \pm 7$ with an emission peak of 505 nm for superior conduction properties (Figure 8f). Then, the emission

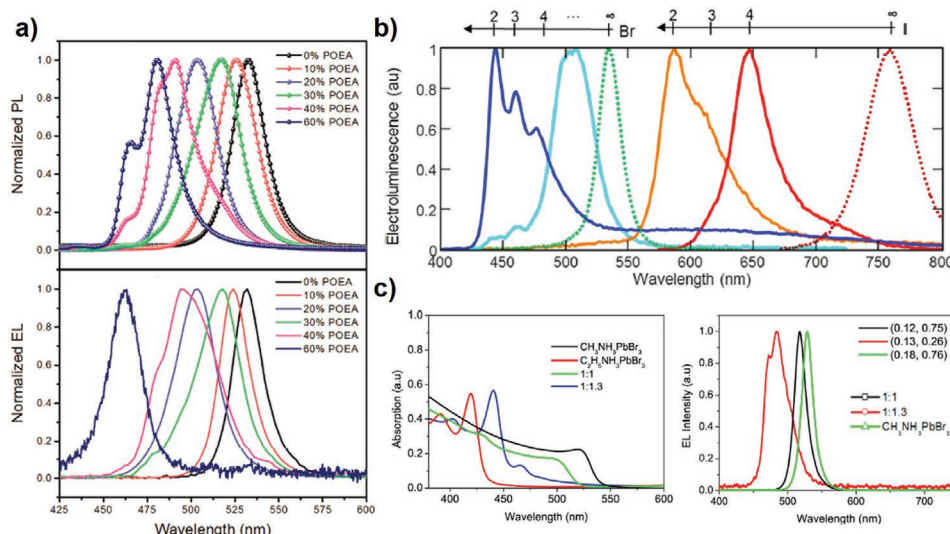


Figure 7. a) PL and EL spectra of $\text{CH}_3\text{NH}_3\text{PbBr}_3$ perovskite thin films incorporated with different concentrations of POEA. Reproduced with permission.^[79] Copyright 2017, Wiley-VCH. b) EL spectra of the PeLEDs based on $\text{BA}_2(\text{MAPbX}_3)_{n-1}\text{PbBr}_4$ ($X = \text{Br}$ or I) with different ratios of BA. Reproduced with permission.^[80] Copyright 2017, American Chemical Society. c) UV-vis spectra $\text{CH}_3\text{NH}_3\text{PbBr}_3$ films with incorporation of different ratios of EABr and the corresponding device EL spectra. Reproduced with permission.^[27] Copyright 2017, American Chemical Society.

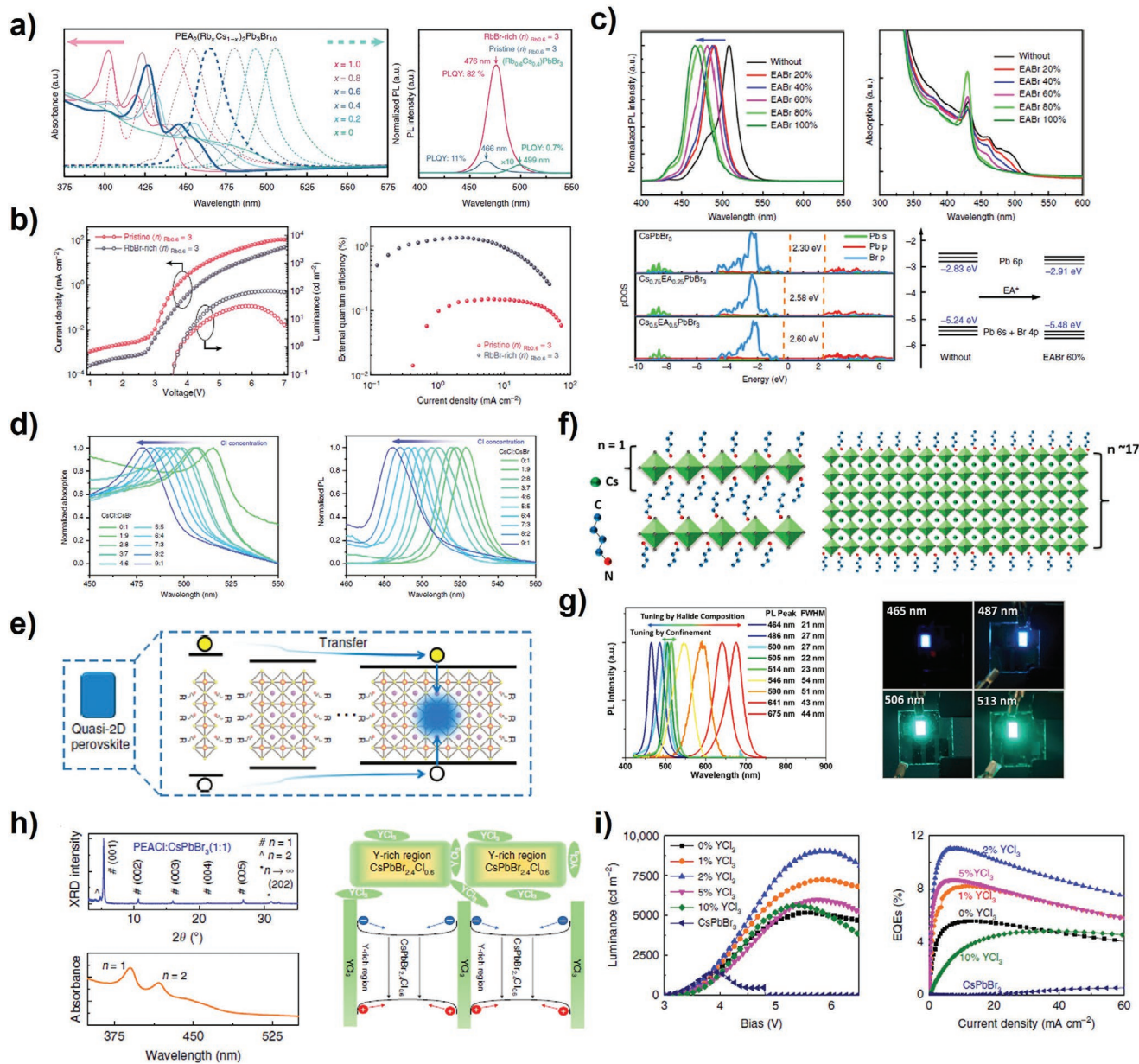


Figure 8. a) UV-vis absorption, steady-state PL spectra of pristine $\text{PEA}_2(\text{Rb}_x\text{Cs}_{1-x})_2\text{Pb}_3\text{Br}_{10}$ perovskites, and steady-state PL spectra and PLQYs of $\text{Rb}_{0.6}\text{Cs}_{0.4}\text{PbBr}_3$, pristine, and RbBr-rich perovskite films. b) J - L - V curves, and EQE characteristics of pristine and RbBr-rich devices. Reproduced with permission.^[81] Copyright 2019, Springer Nature. c) Normalized steady PL and UV-vis absorption spectra of perovskite films with different ratios of EABr, the calculated electronic density of states (DOS), and schematic of energy levels of the quasi-2D perovskite with insertion of EA cation. Reproduced with permission.^[12] Copyright 2020, Springer Nature. d) Normalized absorption and PL spectra of $\text{CsPbCl}_x\text{Br}_{3-x}$ thin films with different molar ratios of CsCl to CsBr. e) The energy transfer and/or charge transfer occurs in the $\text{PEA}_2\text{Cs}_{n-1}\text{Pb}_n(\text{Cl}_x\text{Br}_{1-x})_{3n+1}$ quasi-2D perovskite. Reproduced with permission.^[28] Copyright 2019, Springer Nature. f) Illustrated structure of $\text{BA}_2\text{Cs}_{n-1}\text{Pb}_n\text{X}_{3n+1}$ perovskite for thin ($n = 1$) and thick nanosheet layers. g) Normalized PL spectra and photos of the PeLEDs. Reproduced with permission.^[29] Copyright 2019, American Chemical Society. h) XRD and absorbance of CsPbBr_3 :PEACl film, and schematic illustration of the yttrium gradient distribution in the film. i) Luminance-bias and EQE-current density curves of the devices with different ratios of YCl_3 . Reproduced with permission.^[82] Copyright 2019, Springer Nature.

spectra were further tuned to deep blue by increasing the ratio of Cl ion (Figure 8g). This strategy realized a color tunable $\text{BA}_2\text{Cs}_{n-1}\text{Pb}_n(\text{Br/Cl})_{3n+1}$ quasi-2D perovskite in the blue region with good conduction properties, which benefited the PeLED performances to reach peak EQEs of 2.4% and 6.2% at 465 and 487 nm, respectively.^[29] Based on the mixed halide

quasi-2D perovskite system of $\text{PEA}_2\text{Cs}_{n-1}\text{Pb}_n(\text{Cl}_x\text{Br}_{1-x})_{3n+1}$, Huang and co-workers further introduced yttrium (III) chloride (YCl_3) into the perovskite to confine the charge carriers inside grains for efficient radiative recombination. With the incorporation of YCl_3 , the perovskite film PLQYs greatly increased from 19.8% to 49.7%. In detail, there was a gradient distribution of

yttrium in perovskite crystals with larger proportion on the face than crystal center, which increased the bandgap of grain shell and formed an energy barrier for charge confinement and thus reduced the nonradiative recombination (Figure 8h). Benefitting from this strategy, efficient PeLEDs with EQE and maximum luminance up to 11.0% and 9040 cd m⁻² at 485 nm were achieved (Figure 8i).^[82]

Spacer (Potential Barrier) Component Modulation for Phase Redistribution: Spacer (potential barrier) component modulations were adopted for the optimization of quasi-2D perovskite phase distribution. Typically, quasi-2D perovskites contain a mixture of layered perovskites with wide perovskite phase distribution ranging from the low-*n* (e.g., *n* = 1, 2) to high-*n* (e.g., *n* = 4, 5, ...) phases. This is because, there is a complex mixture of colloids with random size distribution in the precursor solution.^[83] Especially for the small colloids with several unit cells, they cannot be removed by the filter, which can act as the nucleation

center in forming 2D perovskites with different layers, thus resulting in wide perovskite phase distribution. However, the mass high-*n* (e.g., *n* ≥ 5) phases induce red-shift of the spectra to over the blue region, while the formation of low-*n* phases, especially *n* = 1 phase, causes retarded energy transfer and severe nonradiative recombination.^[81,84,85] Thus, modulation of the phase distribution with controlled high-*n* and suppressed low-*n* phases is essential to obtain efficient blue PeLEDs. Xing et al. reported a strategy to realize the phase modulation through developing mixed-ligand (PEA/IPA)₂A_{*n*-1}Pb_{*n*}Br_{3*n*+1} (IPA = isopropylammonium, A = MA and Cs) quasi-2D perovskite. In the mixed-spacer perovskite system, the formation of the pure *n* = 1 phase was slowed, while the purity and monodispersity of *n* = 2, 3, 4 phases were enlarged as shown in the transient absorption (TA) spectroscopy (Figure 9a). This is because compared to the *n* = 2, 3, and 4 phases, the *n* = 1 phase for the mixed-spacer perovskite is destabilized as the formation energy increases from -7.2

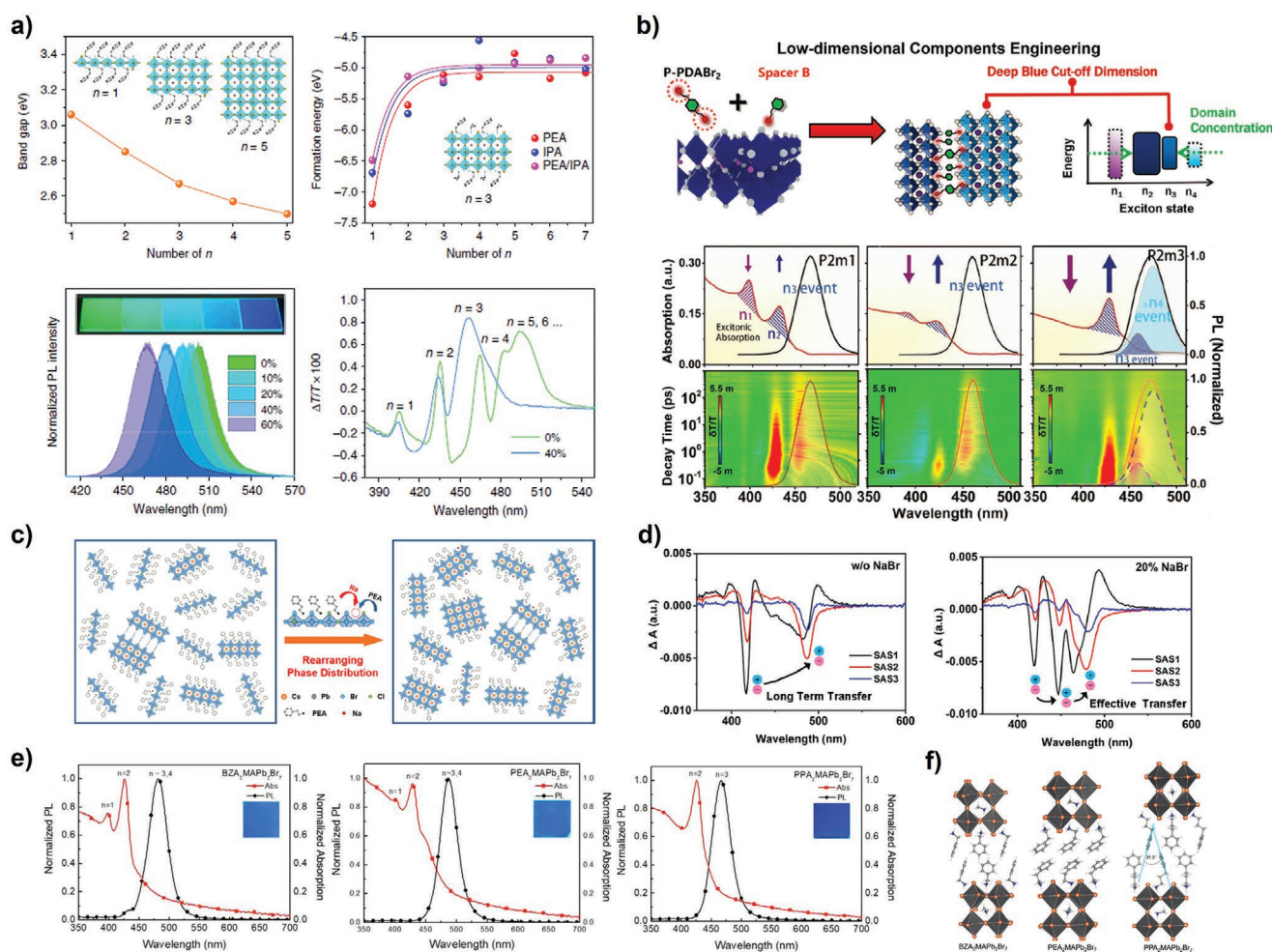


Figure 9. a) Bandgap and formation energy calculations of quasi-2D perovskite, PL spectra of perovskite PEA₂A_{1.5}Pb_{2.5}Br_{8.5} with 0–60% IPABr additive, and TA spectra of PEA₂A_{1.5}Pb_{2.5}Br_{8.5} with 0% and 40% IPABr. Reproduced with permission.^[85] Copyright 2018, Springer Nature. b) Schematic diagram of low-dimensional components engineering, absorption, and PL spectra of three perovskite films, where different amounts of P-PDABr₂ are replaced by PEA and marked as P2m1, P2m2, and P2m3, and the corresponding transient absorption spectroscopy. Reproduced with permission.^[86] Copyright 2019, Wiley-VCH. c) Schematic illustrating the rearrangement of phase distribution by adding Na⁺ in quasi-2D perovskites and d) the TA measurements for exciton transfer dynamics. Reproduced with permission.^[30] Copyright 2020, American Chemical Society. e) Absorption and PL spectra of BZA₂MAPb₂Br₇, PEA₂MAPb₂Br₇, and PPA₂MAPb₂Br₇ perovskites, and f) The corresponding perovskite structures. Reproduced with permission.^[93] Copyright 2020, The Royal Society of Chemistry.

(stable) to -6.5 eV (less stable) by the density functional theory theoretical simulations. Together with the stoichiometry constraints, the quasi-2D perovskites only showed dominated low numbers of inorganic layers ($n = 2, 3$, and 4 phases) for blue emission. Along with the optimization of the film thickness, sky-blue PeLEDs with a maximum luminance of 2480 cd m^{-2} at 490 nm were achieved.^[85] Besides the PEA/IPA mixed cations for phase modulation, the coordination of aromatic polyamines of 1,4-Bis(aminomethyl)benzene bromide (P-PDABr₂) and PEA was also used to pin the domain distribution of $n = 3$ phase by improving $n = 3$ phase stability through forming a strong binding force between P-PDABr₂ and [PbBr₆⁴⁻] octahedron (Figure 9b). The exceptional domain distribution of $n = 3$ phase led to a fast and complete exciton transfer in the perovskite film, which benefited the devices of high EQE of 2.6% at a deep blue emission of 465 nm .^[86] Moreover, other mixed spacer cation combinations, such as PEA⁺ and BA⁺,^[87] PEA⁺ and (*N*-(2-Bromoethyl)-1,3-propanediamine) cation NPA⁺,^[88] PEA⁺ and propylamine cation (PA⁺),^[89] PEA⁺ and isobutylammonium cation (*i*-BA⁺),^[90] PEA⁺ and ethylenediammonium cation (EDA⁺),^[91] and PEA⁺ and 3,3-diphenylpropylamine cation (DPPA⁺)^[92] were also demonstrated as effective groups to redistribute the quasi-2D phase for bright blue emission. Besides the organic spacer cation combinations, the incorporation of sodium ion into quasi-2D perovskites for perovskite phase rearrangement has also been reported.^[30] The perovskite phase redistribution after adding Na⁺ was attributed to the change of crystallization process, in which Na⁺ would competitively occupy the A-sites with PEA⁺ on the perovskite surfaces during the perovskite crystallization process (Figure 9c). However, the $n = 1$ phase of Na⁺ incorporated perovskite showed short thermodynamic stability due to the induced larger formation energy, which resulted in the reduction of $n = 1$ phase while allowing more chance for PEA⁺ to form other larger phases such as $n = 2$ and 3 phases. Benefitting from the suppression of $n = 1$ phase for less nonradiative transition and the enhancement of $n = 2, 3$ phases for efficient exciton energy transfer (Figure 9d), a high EQE of 11.7% with a stable emission peak at 488 nm was achieved.

The alkylammonium tail length in phenyl-alkyl spacer cations also had a distinct effect on the quasi-2D perovskite phase distribution. In detail, the arylamines of benzylammonium (BZA), PEA, and phenylpropylammonium (PPA) with a phenyl ring and one, two, and three carbon atoms in the alkyl chains, respectively, were adopted as spacer cations for comparison. The proportion of $n = 1$ phase decreased following the spacer cation order of BZA, PEA, and PPA (Figure 9e). This indicated that the increase of the alkyl chain length can inhibit the formation of $n = 1$ phase, which could be attributed to the change of the spacer cation packing arrangement in the phase structure where the parallel packing is for BZA and PEA perovskites while non-parallel packing is for PPA to maintain the minimum energy (Figure 9f). Consequently, high phase purity dominated by $n = 2$ phases was achieved for PPA perovskite, which facilitated energy funneling from dominant $n = 2$ phase to $n = 3, 4$ phases and contributed to efficient blue PeLEDs with an EQE of 3.35% at 480 nm .^[93] In addition, other strategies for modulation of quasi-2D perovskite phase distribution have been also developed. For example, Wang et al. adopted chelating-agent-assisted

quantum well growth to control the distribution of quasi-2D perovskite phases. The chelating amino acid of γ -aminobutyric acid (GABA) could coordinate with lead bromide in the vicinity of perovskite and prevented the subsequent binding of PbBr₂ to the perovskite surface, which inhibited the further growth of thick layers and left $n = 2$ and 3 dominated phases for GABA incorporated PEA₂Cs_{*n*-1}Pb_{*n*}Br_{3*n*+1} perovskite. Furthermore, the incorporation of GABA into perovskites improved the film morphology with a greatly reduced roughness from 13.1 to 0.3 nm , which enabled efficient blue PeLEDs with an EQE of 6.3% at 478 nm .^[94]

2.3.3. Quasi-2D Perovskite Phase Passivation for Improved Blue PeLED Performance

Inevitable trap states emerge during crystallization of quasi-2D perovskites, which undoubtedly increase nonradiative recombination and deteriorate the emission efficiency and operational stability. Despite the unclear origin of the traps, they were considered to relate to the ionic defects such as halide and ammonium spacer vacancies.^[95,96] In order to suppress the traps, the ionic vacancies should be effectively passivated.

Efficient Organic Passivators: The organics with functional group of nitrogen or oxygen atom serve as good passivators to reduce the defects by bonding the undercoordinated Pb sites. Tang et al. adopted POEA to passivate the perovskite surface defects.^[97] They proved that POEA had better passivating effect than PEA because the oxygen atoms within POEA could polarize the amino groups through the inductive effect, thus enhancing coordination of the amino groups with the perovskite defect sites. Based on POEA passivated perovskites, the blue PeLEDs exhibited an EQE of 0.71% at 468 nm which was twofold higher than that of PEA based ones. Ma et al. introduced organic chloride to passivate the halide vacancies. Diphenylphosphinic chloride (DPPOCl) could react with trace water in the precursor solution to form DPPOOH which bound with the unsaturated lead dangling bonds and halogen ions on the perovskite edge through coordination bond ($\text{P}=\text{O}:\text{Pb}$) and hydrogen bonds ($\text{O}-\text{H}\cdots\text{Cl}$ and $\text{O}-\text{H}\cdots\text{Br}$), respectively (Figure 10a). The interaction between DPPOOH and perovskite passivated surface halide vacancies and reduced non-radiative recombination, subsequently, improved the perovskite film PLQY and radiative lifetime to 76% and 14.6 ns , respectively. Together with the optimized device structure, high PeLED performance with an EQE of 5.2% at 479 nm and an operating half-life of 90 min at 100 cd m^{-2} were achieved.^[98] Besides, introducing the organic ammonium and halide ions to saturate the ion vacancies is another effective way to passivate the defects for bright blue emission. Ren et al. reported to passivate the quasi-2D perovskite by incorporating excess PEABr. The studies of density of states showed that the pristine perovskite had an obvious trap state at the band edge induced by the ion vacancies. However, the ion vacancies could be saturated by excess PEABr, which could provide the ionic compensation with bromide and ammonium spacer ions for reduction of vacancy sites (Figure 10b). Consistent with this, the trap state density of excess PEABr-decorated perovskite was dramatically reduced to $1.43 \times 10^{17} \text{ cm}^{-3}$ from $3.85 \times 10^{17} \text{ cm}^{-3}$

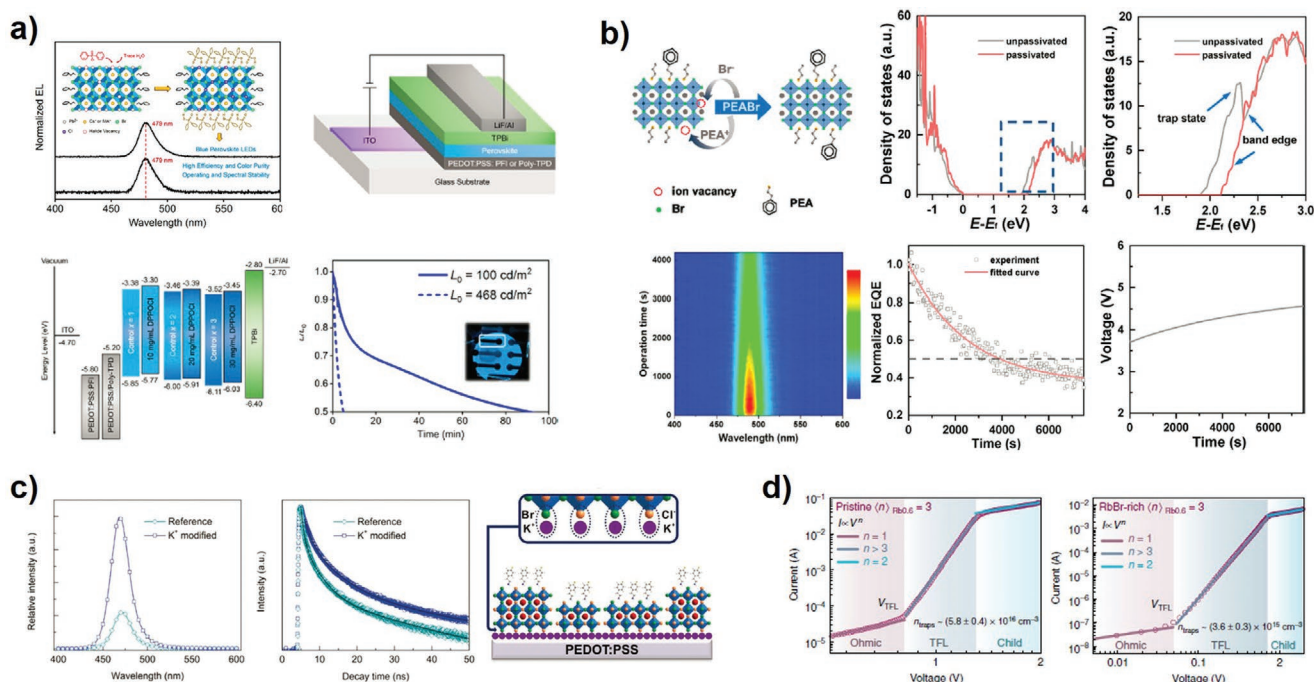


Figure 10. a) Illustration of the passivation mechanism, the device structure and energy band diagram, and operating lifetime of the sky-blue LEDs under different constant driving currents; the inset shows a photograph of the operating device at 468 cd m^{-2} . Reproduced with permission.^[98] Copyright 2020, American Chemical Society. b) Schematic diagram of the passivation role of excess PEABr within the quasi-2D perovskites and the theoretical results of density of states. And, the EL spectra, time-dependent stability, and the corresponding bias voltage traces during the lifetime test. Reproduced with permission.^[89] Copyright 2020, American Chemical Society. c) Steady-state PL, transient PL decay curves of the perovskite film grown on K^+ modified substrate, and schematic illustration of the K^+ interaction with halides. Reproduced with permission.^[101] Copyright 2020, Wiley-VCH. d) Current-voltage (J - V) response of pristine and RbBr-rich perovskite films in SCLC measurement.^[81] Copyright 2019, Springer Nature.

of the pristine perovskite, following a significant increase of perovskite film PLQY from 39.5% to 49.6%. Consequently, they achieved blue PeLEDs with a high EQE of 7.51% at 488 nm and a long operational stability of 66 min (Figure 10b).^[89] Zeng et al. introduced guanidine hydrobromide (GABr) into the perovskite films and improved the carrier decay lifetime (56.66 ns) threefold longer than the untreated films (18.07 ns). The incorporation of GABr led to rich halides on perovskite surface to effectively passivate the halogen defects, which promoted efficient carrier radiative recombination and thus a considerable enhancement of the efficiency from 1.5% to 8.2% at 492 nm.^[99]

Efficient Inorganic Passivators: The inorganic metal salt, such as potassium salt, is a good passivator to reduce the perovskite defects. The potassium ion can passivate the halide defects at the grain boundaries and surfaces due to the interaction between potassium and halide ions to weaken the halide ion migration and thus, reduce the halide vacancies.^[100] Tang et al. employed potassium ion modified substrate to prepare the quasi-2D perovskite films which showed bright blue emission at 467 nm and longer PL lifetime with PLQY increasing from 12.3% to 34.8% (Figure 10c). Meanwhile, the potassium ion modified substrate also assisted the crystal-growth for controlled crystal orientation and uniform perovskite films which led to enhanced radiative recombination. As a result, the fabricated PeLEDs showed an improved EQE of 4.14% at 469 nm.^[101] Yuan et al. reported a Rubidium-Cesium alloyed, quasi-2D perovskite, in which they demonstrated excess RbBr could effectively passivate the perovskite defect states by affecting

the crystallization process as well as crystal orientation. The density of trap states of RbBr-rich perovskite could be lowered more than one order of magnitude than the pristine perovskite (Figure 10d), which extended the carrier lifetime by nearly five times longer than the pristine perovskite and favored a high PLQY of 82% at 476 nm.^[81] Similarly, Wu et al. incorporated RbBr and KBr into the perovskite precursor to form $(\text{Cs/Rb/FA/PEA/K})\text{Pb}(\text{Cl/Br})_3$ multiple cations quasi-2D perovskites. The collaboration of RbBr and KBr showed better passivation effect than the separate use, which led to a distinct enhancement of PL lifetime from 3.61 to 9.17 ns and PLQY from 0.94% to 6.51%. With further incorporation of PEA and FA for the fabrication of uniform film morphology, they achieved blue PeLEDs with an EQE of 2.01% at 484 nm and robust durability with half-lifetime over 300 min under continuous operation.^[102]

2.4. 3D Mixed-Halide Perovskite Blue LEDs

For the 3D mixed-halide perovskite blue emitters, they generally have small exciton binding energies close to the room-temperature thermal energy, suggesting that the produced excitons can easily dissociate into free charge carriers.^[114] The carrier recombination dynamics mainly consist of trap-mediated monomolecular nonradiative recombination, bimolecular radiative recombination, and trimolecular Auger recombination. In order to improve the performance of 3D perovskite blue LEDs, the strategies of fabricating high perovskite film quality (e.g.,

Table 4. Summary of blue PeLED performances based on 3D mixed-halide and lead-free perovskites.

Type	Perovskites	Device structure	EL peak [nm]	Luminance [cd m ⁻²]	EQE [%]	Year	Ref.
3D mixed halides	MAPbBr _{1.08} Cl _{1.92}	ITO/PEDOT/Pero/PC ₆₁ BM/LiF/Ag	482	2	0.0003	2015	[8]
	Cs ₁₀ (MA _{0.17} FA _{0.83}) _{100-x} PbBr _{1.5} Cl _{1.5}	ITO/ZnO/Pero/NPD/MoO ₃ /Al	475	3567	1.7	2017	[121]
	CsPbBr _x Cl _{3-x}	ITO/NiO _x /Pero/TPBi/LiF/Al	468	121	0.38	2019	[117]
	Cs _x Rb _{1-x} PbBr ₃	ITO/PEDOT/Pero/TmPyPB/LiF/Al	492	244	0.17	2019	[115]
	Rb _{0.13} Cs _{0.87} PbBr _{2.22} Cl _{0.78}	ITO/PEDOT/RbCl/Pero/TmPyPB/LiF/Al	484	9243	1.66	2020	[116]
	CsPb(Br _{0.65} Cl _{0.35}) ₃	ITO/PEDOT/Pero/TmPyPB/LiF/Al	482	9352	4.13	2020	[31]
	(Rb-Cs-FA)PbBr _x Cl _{3-x}	ITO/NiO _x /PVK/PVP/Pero/TPBi/LiF/Al	477	2180	11.0	2021	[32]
Lead-free emitters	Cs ₃ Cu ₂ I ₅ NCs	ITO/ZSO/Pero/NPD/MoO _x /Al	440	10	–	2018	[126]
	Cs ₃ Cu ₂ I ₅ NCs	ITO/NiO/Pero/TPBi/LiF/Al	445	263.2	1.12	2020	[127]
	Cs ₃ Sb ₂ Br ₉ NCs	ITO/ZnO/PFI/Pero/TCTA/MoO _x /Al	408	29.6	0.206	2021	[128]

pin-hole free, low defect density) and spatially confining the charge carriers in perovskite emitters can be adopted to reduce the nonradiative recombination and promote the radiative recombination in the 3D perovskites.

The mixed-halide polycrystalline blue LEDs were first reported in 2015 by Kumawat et al. based on the mixed-halide MAPb(Br_{1.08}Cl_{1.92})₃ (MA: methylammonium) perovskites. Nevertheless, the devices show a low luminance of 2 cd m⁻² and a tiny EQE of 3 × 10⁻⁴% at room temperature^[8] due to the weak charge confinement of the polycrystalline perovskite and the poor film morphology for low radiative recombination and high leakage current. In order to improve the film quality, Wang et al. incorporated the dopants of RbX (X = Cl or Br) into polycrystalline CsPbBr₃ and fabricated a dense and uniform film with the film coverage increased from 80% to 99%, which benefitted in terms of an improved device performance with a luminance of 244 cd m⁻² and an EQE of 0.17% at 492 nm.^[115] Subsequently, they further decorated the Rb–Cs polycrystalline perovskite film morphology with isopropanol treatment and interface modification, and improved the efficiency to 1.66% at 484 nm.^[116] Alternatively, Tang et al. adopted thermal evaporation to fabricate compacted CsPbBr_xCl_{3-x} polycrystalline films with a continuous coverage and low surface roughness, which favored the vacuum processed PeLEDs of an EQE of 0.38% at 468 nm.^[117] In addition, the 3D perovskite LED performance can be enhanced by spatially confining the carriers into perovskite emitters through constraining the perovskite size into nanoscale grains. The strategy has demonstrated effective ways for high-efficient green PeLEDs in which nanocrystal pinning process,^[118] large organic cations,^[119] and polymer matrixes^[120] have been adopted to obtain MAPbBr₃ nanograins with strong carrier confinement. Very recently, Wang et al.^[31] and Gao et al.^[32] have incorporated the large organic molecules with the perovskite precursor to form 3D nano-structure mixed-halide perovskite blue emitters which can also promote the confinement of charge carriers into the perovskites, and they achieved EQEs of 4.13% and 11.0% at 482 and 477 nm, respectively. Besides, they also found that the homogeneous halide perovskite benefitted spectral-stable blue PeLEDs which will be discussed in the spectra stability part. Despite the rapid development of 3D perovskite blue PeLEDs (Table 4),^[8,31,32,115–117,121] there is much room for further improvement in future works.

2.5. Lead-Free Perovskite Blue LEDs

The direct way to prepare the lead-free perovskite blue emitters is to replace lead with tin to form CsSnX₃ perovskite (e.g., CsSnCl₃Br_{3-x}, CsSnCl₃). However, the CsSnX₃ perovskites were found to show weak emission with PLQY below 1% due to the susceptible oxidation of Sn²⁺ to Sn⁴⁺.^[122] While Sn⁴⁺-based perovskites (e.g., Cs₂SnCl₆) exhibit much improved stability, Tang et al. synthesized Cs₂SnCl₆ perovskite as a host and then employed Bi³⁺ as a dopant to synthesize Bi³⁺-doped Cs₂SnCl₆ with a high PLQY of 78.9% at 455 nm, in which Bi³⁺ ions were shown to partially replace Sn⁴⁺ and acted as luminescent centers for the bright emission.^[123] Besides, the Bi-based lead-free perovskite of MA₃Bi₂Br₉ NCs with a deep-blue emission at 422 nm was also synthesized. While, the large surface-to-volume ratio of MA₃Bi₂Br₉ NCs resulted in mass defects and a weak emission, with a PLQY of 13.5%. Leng et al. adopted the strategy of halogen surface passivation to suppress MA₃Bi₂Br₉ NC surface defect states and improved the PLQY to 54.1%.^[124] In addition, Song et al. reported the Sb-based lead-free perovskite of Cs₃Sb₂Br₉ NCs which can achieve a high PLQY of 46% at 410 nm by the modified synthesis process of ligand-assisted reprecipitation strategy.^[125] As another promising lead-free blue emitter, Cs₃Cu₂I₅ has also attracted much attention. It was proved that Cs₃Cu₂I₅ had 0D electronic structure. The generated excitons in Cs₃Cu₂I₅ could be spatially confined around the [Cu₂I₅]³⁻ sites which promoted a large exciton binding energy of 492 meV and a high PLQY of 60% at 445 nm.^[126]

For the lead-free blue emitters based LEDs, Hosono et al. explored employing Cs₃Cu₂I₅ thin film as the emission layer and obtained a luminance of 10 cd m⁻² at 440 nm. One of the reasons for the low performance could be attributed to the inefficient charge injection caused by the unfavorable energy level alignment.^[126] In order to prove the promising emission layer of high-quality Cs₃Cu₂I₅ NCs for efficient deep-blue LEDs, Shan et al. adopted an effective device structure of Al/LiF/TPBi/Cs₃Cu₂I₅/NiO/ITO to improve charge carrier injection efficiency. The fabricated deep-blue LEDs exhibited a great enhancement of performance with a luminance of 263.2 cd m⁻² and an EQE of 1.12% at 445 nm. Besides, the

devices also showed a very long half-lifetime of ≈ 108 h under continuous operation.^[127] Recently, Shan et al. also demonstrated the application of $\text{Cs}_3\text{Sb}_2\text{Br}_9$ quantum dots for electrically driven violet emission. The device showed an EQE of $\approx 0.206\%$ at 408 nm and a good working stability with a small emission decay of $\approx 10\%$ after continuous running for 6 h,^[128] indicating the potential candidate of $\text{Cs}_3\text{Sb}_2\text{Br}_9$ for stable violet LEDs. In order to further improve the performance of lead-free emitters based LEDs, it is necessary to match the energy level of carrier transport layers with the large bandgap of lead-free emitters for efficient charge injection in future works.

3. Other Effective Strategies for Further Improvements

Although rapid developments have witnessed a boom for blue PeLEDs, there is still much room for further improvement with the following aspects:

3.1. Strategies for Effective Hole Transport/Injection layers (HTL/HIL)

The HTL/HIL not only has major impacts on the charge injection ability, but also influences the film morphology. The HTL/HIL of PEDOT:PSS, Al 4083 has been widely used in blue PeLEDs. However, it can either result in the aggregation of perovskite NCs or wider phase distribution of the quasi-2D perovskite likely due to its stronger acid nature.^[85] Meanwhile, the shallow HOMO (highest occupied molecular orbital, ≈ -5.1 eV) energy level also causes a large hole-injection barrier and thus high turn-on voltage. In view of this, the neutral organic polymers with deeper HOMO energy level were used as HTL. Song et al. replaced PEDOT:PSS with poly(fluorene-co-phenylene)-based anionic conjugated polyelectrolytes (CPEs) which have deep HOMO level ≈ -5.8 eV to guarantee efficient hole injection from the CPEs into the perovskite (valence band: -5.95 eV).^[106] Similarly, Liao et al. adopted Poly(9-vinylcarbazole) (PVK)^[88] and PVK/PFI (perfluorinated ionomer)^[86] as HTLs to reduce the hole injection barrier for efficient hole injection. It could be difficult to obtain uniform 3D/quasi-2D perovskite films on the surface of the hydrophobic polymers via spin-coating process due to the poor wettability. To improve the wettability of perovskite precursor on the HTL, Yao et al. adopted inorganic HTL of NiO_x/LiF film as a HTL which showed a high affinity with the precursor solution which benefitted for a good film morphology and reduced the quenching effect for bright blue emission of 1446 cd cm^{-2} at 490 nm.^[103] Recently, Ren et al. introduced the poly(sodium-4-styrene sulfonate) (PSSNa) modified NiO_x ($\text{NiO}_x/\text{PSSNa}$) film as HTL which was proven to simultaneously reduce the hole injection barrier by the increased work function and suppress the nonradiative recombination by reducing the pin-holes in the perovskite films and thus favoring very bright blue PeLEDs of 5737 cd m^{-2} .^[109] This indicated a great potential of inorganic HTL/HIL for fabrication of high-performing PeLEDs.

3.2. Strategies for Developing Efficient Low-Dimensional Perovskite Emitter System

As for perovskite nanocrystal blue emitters, the nanocrystals are pre-synthesized first, then deposited on the substrate, in which several purification processes are needed to discard excess ligands before use. However, inevitable trap states are produced owing to the ligand loss during purification, which results in severely nonradiative recombination and thus low emission efficiency. In view of this, Wang et al. reported to in situ fabricate the perovskite nanocrystal films by a facile spin-coating method. They achieved small $\text{Cs}_x\text{MA}_{1-x}\text{PbBr}_3$ nanocrystals with average sizes decreasing from 4.5 ± 1.1 to 4.1 ± 1.0 nm by a delay dropping time from 7 to 9 s, which led to effective blue-shifts of the PL peaks from 475 to 464 nm. Based on the high-quality nanocrystalline perovskite nanocrystals, the fabricated PeLEDs exhibited an efficient deep-blue emission of 465 nm with an EQE of 2.4%.^[47] Besides, Zhong et al. demonstrated the in situ fabrication of CsPbClBr_2 nanocrystal films on substrate by collocating the mixed ligands of 2-phenylethanamine bromide and 3,3-diphenylpropylamine bromide. They showed the use of dual-ligand an effective strategy for controlling the compositional domains in CsPbClBr_2 nanocrystal films, in which 2-phenylethanamine bromide was adopted to form small- n domains while 3,3-diphenylpropylamine bromide was for large- n domains. By carefully optimizing the ratio of the two ligands, the quantum-well width distribution with central domination of $n = 4$ was achieved, which facilitated efficient energy transfer for bright blue emission and promoted decent device performance with an EQE of 8.8% at 473 nm.^[92] In addition, Jin et al. proved in situ antisolvent processing scheme to develop the emitter system of quantum-confined perovskite nanoparticles embedded within quasi-2D phases for efficient blue emission. In the formation process of the hybrid perovskite system, they demonstrated that the use of ethyl acetate antisolvent was critical to remove excess phenylbutylammonium cations, improve charge injection in the perovskite films, and strengthen energetic coupling between nanoparticles and quasi-2D phases for high EL efficiency. As a result, they obtained strong emissive perovskite films with PLQY over 60% and achieved good device performance with an EQE up to 9.5% at 483 nm.^[104]

3.3. Strategies for Light Out-Coupling Device Structure

In planar LEDs, it is a big challenge to enhance the light out-coupling efficiency due to the various permittivities of dielectric and metal thin film multilayers, which result in near 80% of the light generated by the emitters being trapped inside the device.^[129,130] Thus, there is a strong aspiration to improve the device performance by enhancing the light out-coupling efficiency. The construction of periodic micro-nanostructures on the device was accepted as an efficient way to improve the light out-coupling efficiency. Fan et al. fabricated efficient MAPbBr_3 light-emitting diodes on nanophotonic substrates which comprised the nanodome light couplers and the nanowire arrays optical antennas. The optical modeling showed that a high light extraction efficiency of 73.6% can be achieved with the nanophotonic substrates. The formed photonic crystal optical antennas could convert

waveguide modes to leaky modes and resulted in the enhanced light out-coupling in the nanostructured device. As a result, a great enhancement of EQE from 8.19% to 17.5% was achieved with the nanophotonic substrates.^[131] In addition, Tang et al. showed that a bioinspired moth-eye nanostructure could enhance the out-coupling efficiency of waveguided light in PeLEDs. The moth-eye nanostructured patterns were embedded at the front electrode/perovskite interface which out-coupled the waveguided light, while maintaining spectral and angular independence. Moreover, the patterns induced a broadband increase of optical transmittance over the spectral range of 350–800 nm due to the broad distribution of periodicity and the gradient refractive index profiles of the subwavelength nanostructures. Consequently, the based CsPbBr₃ PeLEDs showed a high EQE of 20.3%, which was 1.5 times higher than that of the reference flat device.^[132]

4. Challenges

Despite the achieved substantial progress in perovskite NC and quasi-2D perovskite blue PeLEDs, there are still challenges for the blue PeLEDs toward practical applications. For example, only moderate device performances were achieved for deep-blue (EL peak ≤ 470 nm) PeLEDs, in which the highest efficiency for NC blue PeLEDs is 6.3% at 470 nm,^[54] while only 5.5% at 467 nm for quasi-2D blue PeLEDs.^[32] Besides, there is poor stability for blue PeLEDs, especially for the mixed-halide perovskite emitters; the poor spectral stability induced by trap states and/or halide ion migration, is an obstacle for the practical applications, which needs to be solved urgently.

4.1. The Strategies for Improving the Spectra Stability of Blue PeLEDs

In blue PeLEDs, perovskite phase segregation in mixed halide perovskite blue emitters is the key issue to cause unstable

spectra. To improve the spectral stability of blue PeLEDs, we first need to protect the devices from the surrounding environment erosion by either sealing the devices or carrying out the test into glove box. Then, the mixed halide perovskite phase segregation induced by ion migration should be suppressed.^[133] In order to inhibit the ion migration, Kim et al. optimized the spacer cation of diphenylpropylammonium to form Br/Cl mixed halide perovskite. They found that the benzene rings in diphenylpropylammonium cation promoted the positive charge concentrated on the single N atom which provided better electrostatic interaction with Cl[−] to prevent the halide migration. As a result, the based PeLEDs exhibited improved spectral stability with 2 nm redshifts from 464 to 466 nm with increasing the bias voltage from 4 to 10 V and 4 nm redshifts during 20 min of operation at 5 V.^[108] Ma et al. adopted the strategy of immobilizing halogen ions to prevent the halide migration for spectral stable PeLEDs, in which the halogen ions were anchored by diphenylphosphinic acid through hydrogen bonds (O–H···Cl and O–H···Br) (Figure 10a). The PeLEDs showed decent spectral stability at 479 nm without shifts of the EL spectra under operation for 90 min.^[98] Recently, the heterogeneous distribution of halides in the perovskite precursor was revealed to account for the phase separation by Wang's group.^[31] They showed that the phase separation issue can be effectively suppressed in CsPbBr_xCl_{3−x} perovskites through introducing the cationic surfactants of tetraphenylphosphonium bromide (TPPB) and polyoxyethylene sorbitan monolaurate (Tween) to improve the homogeneity of the halides in precursor solution (Figure 11a). The homogenized CsPb(Br_{1−x}Cl_x)₃ perovskites were proved to exhibit much more stable structures than the phase-separation ones by the theoretical calculations. Subsequently, the blue PeLEDs showed good color stability at 482 nm and the shapes of EL spectra are consistent during the lifetime measurement (half-lifetime of 41 min). The PeLEDs also exhibited decent color stability under various bias voltages

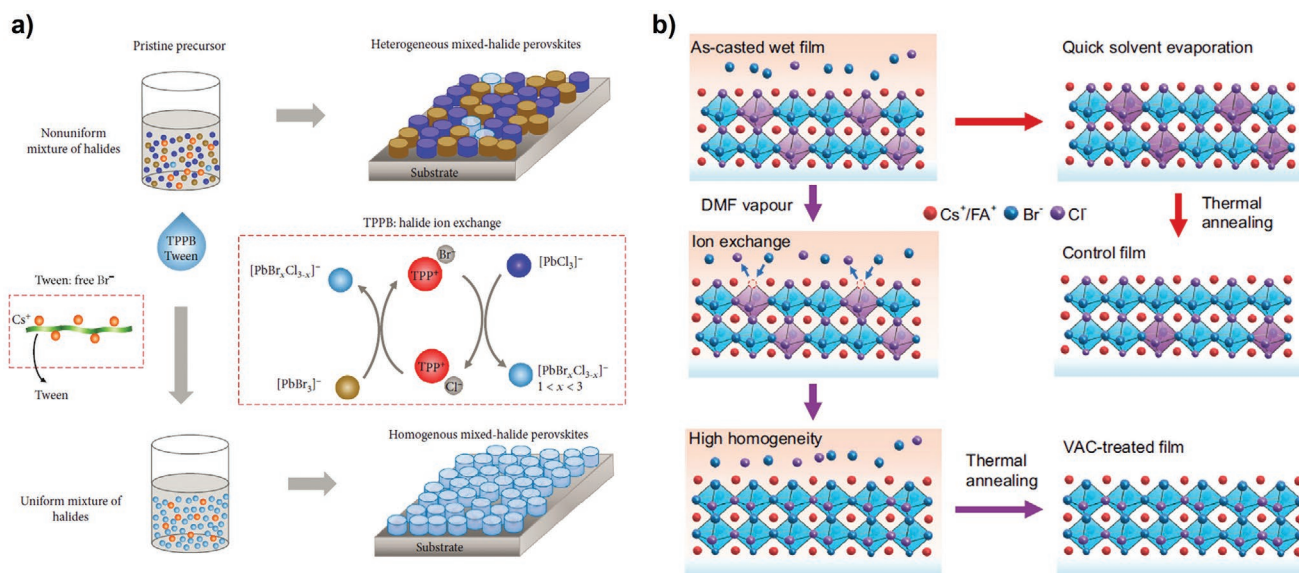


Figure 11. a) Schematic illustration of phase homogenization process through the assistance of Tween and TPPB. Reproduced with permission.^[31] Copyright 2020, American Association for the Advancement of Science. b) Schematic illustration of the proposed mechanism for halide redistribution via vapor-assisted crystallization technique. Reproduced with permission.^[32] Copyright 2021, Springer Nature.

Table 5. The operational stability of NC and quasi-2D blue PeLEDs.

Type	Perovskites	EL Peak [nm]	EQE [%]	Operational stability testing conditions		Lifetime [T_{50}]	Year	Ref.
				Constant current /voltage	Luminance/EQE decay			
NC blue PeLEDs	CsPbCl _{1.7} Br _{1.3}	460	1.35	3.7 V	<10 cd m ⁻²	51.5 s	2019	[59]
	CsPbBr _x Cl _{3-x}	470	1.34	3.5 V	<50 cd m ⁻²	257 s	2020	[77]
	CsPb(Br/Cl) ₃	477	1.96	4 V	26.7 cd m ⁻²	4.5 min	2020	[73]
	CsPb(Br _x Cl _{1-x}) ₃	470	6.3	4.5 V	<100 cd m ⁻²	99 s	2020	[54]
	CsPbBr ₃	479	12.3	—	100 cd m ⁻²	20 min	2020	[11]
	CsPbBr ₃	470	4.7	12.5 mA cm ⁻²	102 cd m ⁻²	12 h	2021	[46]
Quasi-2D system blue PeLEDs	IPA/PEA ₂ MACs _{n-1} Pb _n Br _{3n+1}	490	1.5	constant current	10 cd m ⁻²	600s	2018	[85]
	PEA ₂ (Rb _x Cs _{1-x}) ₂ Pb ₃ Br ₁₀	475	1.35	4.5 V	15 cd m ⁻²	14.5 min	2019	[81]
	P-PDA/PEA ₂ Cs _{n-1} Pb _n Br _{3n+1}	465	2.6	0.35 mA cm ⁻²	2.6%	13.5 min	2019	[86]
	PEA ₂ Cs _{n-1} Pb _n (Cl _x Br _{1-x}) _{3n+1}	480	5.7	4.4 V	≈5.5%	10 min	2019	[28]
	PBABr _{1.1} (Cs _{0.7} FA _{0.3} PbBr ₃)	483	9.5	1 mA cm ⁻²	100 cd m ⁻²	250 s	2019	[104]
	PEACl:YCl ₃ :CsPbBr ₃	485	11.0	3.2 V	100 cd m ⁻²	≈120 min	2019	[82]
	(Cs/Rb/FA/PEA/K)Pb(Cl/Br) ₃	484	2.01	25 mA cm ⁻²	—	300 min	2020	[102]
	EDA/PEA ₂ Cs _{n-1} Pb _n Br _{3n+1}	474	2.17	5.4 V	20 cd m ⁻²	150 s	2020	[91]
	PEA ₂ NPA ₁ Cs ₂ Pb ₃ Br ₁₂	482	2.62	3.0 V	15 cd m ⁻²	8.8 min	2020	[88]
	DPPA ₂ Cs _{n-1} Pb _n (Br/Cl) _{3n+1}	475	3.03	5.0 V	—	420 s	2020	[108]
	(Cs/FA/p-F-PEA)Pb(Cl/Br) ₃	469	4.14	1 mA cm ⁻²	≈30 cd m ⁻²	14 min	2020	[101]
	PBA ₂ Cs _{n-1} Pb _n Br _{3n+1}	493	5.08	—	15 cd m ⁻²	232 s	2020	[110]
	PEA ₂ Cs _{1.6} MA _{0.4} Pb ₃ Br ₁₀	479	5.2	constant current	100 cd m ⁻²	90 min	2020	[98]
	GABA-PEA ₂ Cs _{n-1} Pb _n Br _{3n+1}	478	6.3	—	200 cd m ⁻²	150 s	2020	[94]
	PEA _x PA _{2-x} (CsPbBr ₃) _{n-1} PbBr ₄	488	7.22	1.5 mA cm ⁻²	6.02%	3961s	2020	[89]
	(BA _x PEA _{1-x}) ₂ Cs _{n-1} Pb _n (Br _{0.7} Cl _{0.3}) _{3n+1}	485	7.84	—	100 cd m ⁻²	311 s	2020	[90]
	[(PEA) _{0.75} (GA) _{0.25}] ₂ CsPb ₂ (Br/Cl) ₇	492	8.2	5 V	21 cd m ⁻²	274 s	2020	[99]
	Na/PEA ₂ Cs _{n-1} Pb _n (Cl _x Br _{1-x}) _{3n+1}	488	11.7	constant current	100 cd m ⁻²	≈970 s	2020	[30]
	(ABA/PEA) _x PA _{2-x} (CsPbBr ₃) _{n-1} PbBr ₄	486	10.11	0.3 mA cm ⁻²	8.65%	81.3 min	2021	[112]

(3.4–5.0 V) with emission peaks from 489 to 460 nm. Similarly, Gao et al. showed that the phase segregation induced by halide ion migration and compositional heterogeneity could be remarkably inhibited by an efficient perovskite film deposition way of vapor-assisted crystallization (VAC) technique.^[32] The VAC treatment provided a favorable diffusive environment for halide rearrangement within the films which resulted in an equilibrating crystallization process for homogeneous constituents (Figure 11b). Consequently, the VAC-prepared blue PeLEDs showed stable EL spectra at 479 nm with a negligible shift of the CIE coordinates up to around 400 mA cm⁻² (6–6.5 V). In addition, the spectrally stable PeLEDs with emission colors from sky-blue to deep-blue were demonstrated.

4.2. Testing Standard for the Operational Stability

Meanwhile, there is no testing standard to directly compare the performances of PeLED operational stability by considering the different testing conditions (Table 5). The lifetime testing modes of PeLEDs under constant current and voltage are illustrated in Figure 12a,b, respectively. In the constant

current mode, the applied voltage increases with extending the operating time, while the current decreases in the constant voltage mode, with both showing an increased resistance during operation. The growing resistance can be induced by the increases of material resistance (e.g., material damages induced by the decomposition of perovskite emitters and transport layers) and interface resistance (e.g., the interface degradations caused by interface electrochemical reactions, oxygen, and moisture erosion) in the device working process. Comparing with the constant current mode, the constant voltage mode cannot exactly show the information of injected charges in the device due to the voltage loss induced by the enlarged resistance during operation. In addition, the wire resistance and the contact resistance affect the applied voltage on the devices and thus, the working reliability. Therefore, we suggest the lifetime tests of blue PeLEDs under continuous driving current. The lifetime can be evaluated by T_{50} (defined as the elapsed time of PeLED decaying to 50% of its initial luminance), in which the initial luminance can be fixed at 100 cd m⁻² in view of the presently moderate performances of blue PeLEDs, while higher initial values (e.g., 500 cd m⁻², 1000 cd m⁻²) can be adopted once breakthroughs of PeLED performances are achieved in the

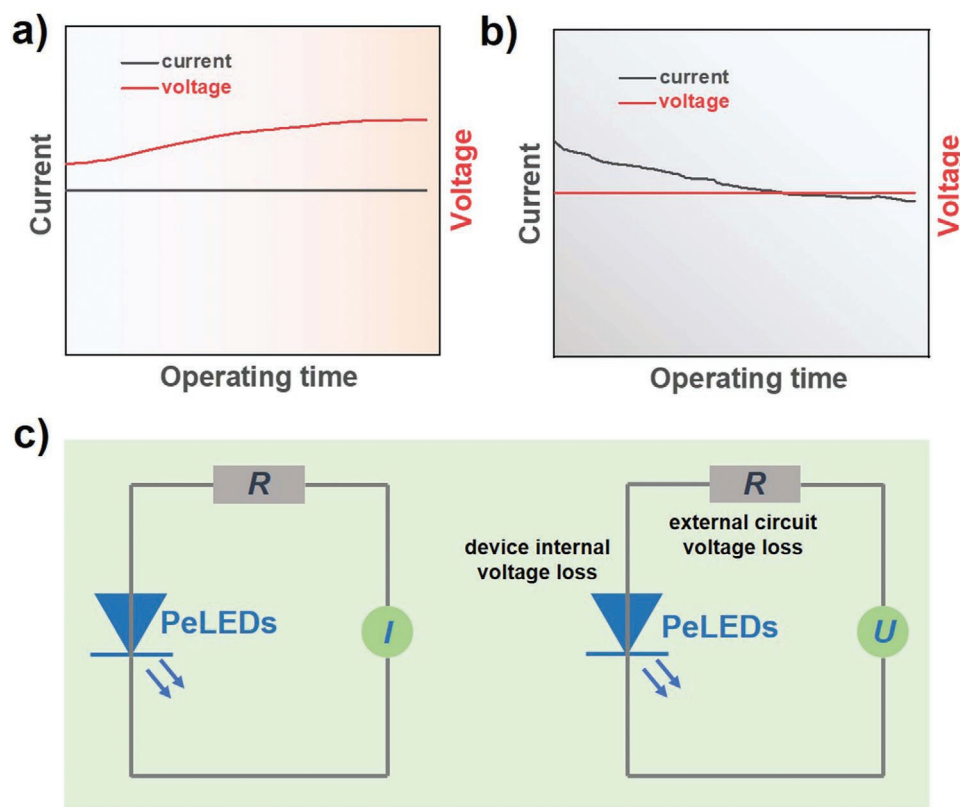


Figure 12. The lifetime testing modes of PeLEDs under a) constant current with bias voltage traces, b) constant voltage with current traces, and c) the corresponding circuit diagram.

near future. Besides, the traces of the EL spectra should better be provided to monitor the device spectral stability during lifetime test.

5. Conclusion and Outlook

Substantial progress achieved in perovskite blue emitters has promoted a boom in their blue PeLED efficiencies. In this review, we have summarized the strategies mainly focusing on the optimization of the optoelectronic properties of perovskite NCs and quasi-2D perovskites for the achievements of high blue PeLED performances. We have systematically discussed the key strategies to obtain high-quality perovskite NCs with less traps and good conductance through the ligand optimization and metal doping and thus for efficient NC PeLEDs. For quasi-2D perovskite, the key issues of phase composition, distribution, and defects are discussed in detail, in which we introduce the fundamental strategies of perovskite phase modulation and passivation for efficient quasi-2D PeLEDs with improved color purity, fast energy transfer, and long operational stability. Meanwhile, we also briefly introduce the strategies of improving the perovskite film quality and confining the charge carriers in perovskites for efficient 3D mixed-halide perovskite, as well as optimizing the energy level alignment for efficient lead-free perovskite blue LEDs. After that, other strategies, such as effective charge transport/injection layer of inorganic HTLs,

efficient perovskite emitters of in situ fabrication of perovskite emitter system, and effective device structure for high light out-coupling efficiency are proposed to further improve blue PeLED performance. Besides, we also discuss the challenges existing in blue PeLEDs and suggest a testing standard of blue PeLED lifetime to enable the direct comparisons of the device operational stability.

For the future directions of blue PeLEDs: (1) High efficiencies at high brightness will be expected for the practical applications of blue PeLEDs. Since the severe device efficiency roll-off at high brightness relates closely with Auger recombination,^[134] the effective strategies of diminishing Auger recombination (e.g., modulation of quantum confinement effect of blue emitters) are urgently needed. (2) Fundamentally revealing the origins of the halide segregation in the mixed halide perovskites for the guidance of achieving stable and efficient deep-blue PeLEDs. It remains elusive of the origins of halide segregation, which are likely due to the free energy differences, the perovskite lattice strain, or the electric fields induced halide segregation. (3) Other available emitters for stable blue PeLEDs can also be explored. For example, the single crystal perovskites with low density of trap states exhibit good stability against heat and environmental oxygen/moisture showing great potential toward stable PeLEDs.^[135,136] Consequently, this review not only provides a comprehensive vision of blue PeLEDs but also contributes an instructive prospect for blue PeLED community.

Acknowledgements

The authors acknowledge Fanghao Ye and Jiayun Sun for their help in proof-reading. This research was supported by Equipment Fund, Platform Research Fund, and Feed fund (Grant Nos. 2019157209, and 201811159147), from the University Grant Council of the University of Hong Kong, the General Research Fund (Grant Nos. 17211220, 17201819, 17204117, and 17200518), Collaborative Research Fund (Grant No. C7035-20G) from Hong Kong Special Administrative Region, the Ministry of Science and Technology of China (No. 2016YFB0401702), Key-Area Research and Development Program of Guangdong Province (Project No. 2019B010925001, 2019B010924001), Guangdong University Key Laboratory for Advanced Quantum Dot Displays and Lighting (No. 2017KSYS007), and Shenzhen Peacock Team Project (No. KQTD2016030111203005).

Conflict of Interest

The authors declare no conflict of interest.

Keywords

blue perovskite light-emitting diodes, efficient strategies, perovskite nanocrystals, quasi-2D perovskites

Received: January 18, 2021

Revised: March 23, 2021

Published online:

- [1] L. N. Quan, B. P. Rand, R. H. Friend, S. G. Mhaisalkar, T.-W. Lee, E. H. Sargent, *Chem. Rev.* **2019**, 119, 7444.
- [2] Z. K. Tan, R. S. Moghaddam, M. L. Lai, P. Docampo, R. Higler, F. Deschler, M. Price, A. Sadhanala, L. M. Pazos, D. Credgington, F. Hanusch, T. Bein, H. J. Snaith, R. H. Friend, *Nat. Nanotechnol.* **2014**, 9, 687.
- [3] L. Protesescu, S. Yakunin, M. I. Bodnarchuk, F. Krieg, R. Caputo, C. H. Hendon, R. X. Yang, A. Walsh, M. V. Kovalenko, *Nano Lett.* **2015**, 15, 3692.
- [4] X. Li, Y. Wu, S. Zhang, B. Cai, Y. Gu, J. Song, H. Zeng, *Adv. Funct. Mater.* **2016**, 26, 2435.
- [5] K. Lin, J. Xing, L. N. Quan, F. Pelayo García de Arquer, X. Gong, J. Lu, L. Xie, W. Zhao, D. Zhang, C. Yan, W. Li, X. Liu, Y. Lu, J. Kirman, E. H. Sargent, Q. Xiong, Z. Wei, *Nature* **2018**, 562, 245.
- [6] B. Zhao, S. Bai, V. Kim, R. Lamboll, R. Shivanna, F. Auras, J. M. Richter, L. Yang, L. Dai, M. Alsari, X.-J. She, L. Liang, J. Zhang, S. Lilliu, P. Gao, H. J. Snaith, J. Wang, N. C. Greenham, R. H. Friend, D. Di, *Nat. Photonics* **2018**, 12, 783.
- [7] Y. Cao, N. Wang, H. Tian, J. Guo, Y. Wei, H. Chen, Y. Miao, W. Zou, K. Pan, Y. He, H. Cao, Y. Ke, M. Xu, Y. Wang, M. Yang, K. Du, Z. Fu, D. Kong, D. Dai, Y. Jin, G. Li, H. Li, Q. Peng, J. Wang, W. Huang, *Nature* **2018**, 562, 249.
- [8] N. K. Kumawat, A. Dey, A. Kumar, S. P. Gopinathan, K. L. Narasimhan, D. Kabra, *ACS Appl. Mater. Interfaces* **2015**, 7, 13119.
- [9] D. Liang, Y. Peng, Y. Fu, M. J. Shearer, J. Zhang, J. Zhai, Y. Zhang, R. J. Hamers, T. L. Andrew, S. Jin, *ACS Nano* **2016**, 10, 6897.
- [10] J. Song, J. Li, X. Li, L. Xu, Y. Dong, H. Zeng, *Adv. Mater.* **2015**, 27, 7162.
- [11] Y. Dong, Y.-K. Wang, F. Yuan, A. Johnston, Y. Liu, D. Ma, M.-J. Choi, B. Chen, M. Chekini, S.-W. Baek, L. K. Sagar, J. Fan, Y. Hou, M. Wu, S. Lee, B. Sun, S. Hoogland, R. Quintero-Bermudez, H. Ebe, P. Todorovic, F. Dinic, P. Li, H. T. Kung, M. I. Saidaminov, E. Kumacheva, E. Spiecker, L.-S. Liao, O. Voznyy, Z.-H. Lu, E. H. Sargent, *Nat. Nanotechnol.* **2020**, 15, 668.
- [12] Z. Chu, Y. Zhao, F. Ma, C.-X. Zhang, H. Deng, F. Gao, Q. Ye, J. Meng, Z. Yin, X. Zhang, J. You, *Nat. Commun.* **2020**, 11, 4165.
- [13] T. Fang, F. Zhang, S. Yuan, H. Zeng, J. Song, *InfoMat* **2019**, 1, 211.
- [14] S. Peng, S. Wang, D. Zhao, X. Li, C. Liang, J. Xia, T. Zhang, G. Xing, Z. Tang, *Small Methods* **2019**, 3, 1900196.
- [15] N. K. Kumawat, X.-K. Liu, D. Kabra, F. Gao, *Nanoscale* **2019**, 11, 2109.
- [16] X. Gong, O. Voznyy, A. Jain, W. Liu, R. Sabatini, Z. Piontkowski, G. Walters, G. Bappi, S. Nokhrin, O. Bushuyev, M. Yuan, R. Comin, D. McCamant, S. O. Kelley, E. H. Sargent, *Nat. Mater.* **2018**, 17, 550.
- [17] W. Deng, X. Jin, Y. Lv, X. Zhang, X. Zhang, J. Jie, *Adv. Funct. Mater.* **2019**, 29, 1903861.
- [18] B. J. Bohn, Y. Tong, M. Gramlich, M. L. Lai, M. Döblinger, K. Wang, R. L. Z. Hoyer, P. Müller-Buschbaum, S. D. Stranks, A. S. Urban, L. Polavarapu, J. Feldmann, *Nano Lett.* **2018**, 18, 5231.
- [19] C. Zhang, Q. Wan, B. Wang, W. Zheng, M. Liu, Q. Zhang, L. Kong, L. Li, *J. Phys. Chem. C* **2019**, 123, 26161.
- [20] S. Peng, Z. Wen, T. Ye, X. Xiao, K. Wang, J. Xia, J. Sun, T. Zhang, G. Mei, H. Liu, B. Xu, X. Li, R. Chen, G. Xing, K. Wang, Z. Tang, *ACS Appl. Mater. Interfaces* **2020**, 12, 31863.
- [21] Y. Wu, C. Wei, X. Li, Y. Li, S. Qiu, W. Shen, B. Cai, Z. Sun, D. Yang, Z. Deng, H. Zeng, *ACS Energy Lett.* **2018**, 3, 2030.
- [22] S. Kumar, J. Jagielski, S. Yakunin, P. Rice, Y. C. Chiu, M. Wang, G. Nedelcu, Y. Kim, S. Lin, E. J. G. Santos, M. V. Kovalenko, C. J. Shih, *ACS Nano* **2016**, 10, 9720.
- [23] D. Yang, Y. Zou, P. Li, Q. Liu, L. Wu, H. Hu, Y. Xu, B. Sun, Q. Zhang, S.-T. Lee, *Nano Energy* **2018**, 47, 235.
- [24] R. L. Z. Hoyer, M.-L. Lai, M. Anaya, Y. Tong, K. Gałkowski, T. Doherty, W. Li, T. N. Huq, S. Mackowski, L. Polavarapu, J. Feldmann, J. L. MacManus-Driscoll, R. H. Friend, A. S. Urban, S. D. Stranks, *ACS Energy Lett.* **2019**, 4, 1181.
- [25] W. Yin, M. Li, W. Dong, Z. Luo, Y. Li, J. Qian, J. Zhang, W. Zhang, Y. Zhang, S. V. Kershaw, X. Zhang, W. Zheng, A. L. Rogach, *ACS Energy Lett.* **2021**, 6, 477.
- [26] L. Cheng, Y. Cao, R. Ge, Y.-Q. Wei, N.-N. Wang, J.-P. Wang, W. Huang, *Chin. Chem. Lett.* **2017**, 28, 29.
- [27] Q. Wang, J. Ren, X.-F. Peng, X.-X. Ji, X.-H. Yang, *ACS Appl. Mater. Interfaces* **2017**, 9, 29901.
- [28] Z. Li, Z. Chen, Y. Yang, Q. Xue, H.-L. Yip, Y. Cao, *Nat. Commun.* **2019**, 10, 1027.
- [29] P. Vashishtha, M. Ng, S. B. Shivarudraiah, J. E. Halpert, *Chem. Mater.* **2019**, 31, 83.
- [30] P. Pang, G. Jin, C. Liang, B. Wang, W. Xiang, D. Zhang, J. Xu, W. Hong, Z. Xiao, L. Wang, G. Xing, J. Chen, D. Ma, *ACS Nano* **2020**, 14, 11420.
- [31] L. Cheng, C. Yi, Y. Tong, L. Zhu, G. Kusch, X. Wang, X. Wang, T. Jiang, H. Zhang, J. Zhang, C. Xue, H. Chen, W. Xu, D. Liu, R. A. Oliver, R. H. Friend, L. Zhang, N. Wang, W. Huang, J. Wang, *Research* **2020**, 2020, 9017871.
- [32] M. Karlsson, Z. Yi, S. Reichert, X. Luo, W. Lin, Z. Zhang, C. Bao, R. Zhang, S. Bai, G. Zheng, P. Teng, L. Duan, Y. Lu, K. Zheng, T. Pullerits, C. Deibel, W. Xu, R. Friend, F. Gao, *Nat. Commun.* **2021**, 12, 361.
- [33] Y. Chen, Y. Sun, J. Peng, J. Tang, K. Zheng, Z. Liang, *Adv. Mater.* **2018**, 30, 1703487.
- [34] J. Almutlaq, J. Yin, O. F. Mohammed, O. M. Bakr, *J. Phys. Chem. Lett.* **2018**, 9, 4131.
- [35] M. Yuan, L. N. Quan, R. Comin, G. Walters, R. Sabatini, O. Voznyy, S. Hoogland, Y. Zhao, E. M. Beauregard, P. Kanjanaboos, Z. Lu, D. H. Kim, E. H. Sargent, *Nat. Nanotechnol.* **2016**, 11, 872.
- [36] L. Cheng, T. Jiang, Y. Cao, C. Yi, N. Wang, W. Huang, J. Wang, *Adv. Mater.* **2020**, 32, 1904163.

- [37] H.-C. Wang, Z. Bao, H.-Y. Tsai, A.-C. Tang, R.-S. Liu, *Small* **2018**, *14*, 1702433.
- [38] X. Dai, Y. Deng, X. Peng, Y. Jin, *Adv. Mater.* **2017**, *29*, 1607022.
- [39] X. Yang, X. Xu, G. Zhou, *J. Mater. Chem. C* **2015**, *3*, 913.
- [40] M. Lu, Y. Zhang, S. Wang, J. Guo, W. W. Yu, A. L. Rogach, *Adv. Funct. Mater.* **2019**, *29*, 1902008.
- [41] W. van der Stam, J. J. Geuchies, T. Altantzis, K. H. W. van den Bos, J. D. Meeldijk, S. Van Aert, S. Bals, D. Vanmaekelbergh, C. Donega, *J. Am. Chem. Soc.* **2017**, *139*, 4087.
- [42] M. Liu, G. Zhong, Y. Yin, J. Miao, K. Li, C. Wang, X. Xu, C. Shen, H. Meng, *Adv. Sci.* **2017**, *4*, 1700335.
- [43] X. Zhang, H. Wang, Y. Hu, Y. Pei, S. Wang, Z. Shi, V. L. Colvin, S. Wang, Y. Zhang, W. W. Yu, *J. Phys. Chem. Lett.* **2019**, *10*, 1750.
- [44] R. Begum, M. R. Parida, A. L. Abdelhady, B. Murali, N. M. Alyami, G. H. Ahmed, M. N. Hedhili, O. M. Bakr, O. F. Mohammed, *J. Am. Chem. Soc.* **2017**, *139*, 731.
- [45] Y.-H. Kim, C. Wolf, Y.-T. Kim, H. Cho, W. Kwon, S. Do, A. Sadhanala, C. G. Park, S.-W. Rhee, S. H. Im, R. H. Friend, T.-W. Lee, *ACS Nano* **2017**, *11*, 6586.
- [46] C. Bi, Z. Yao, X. Sun, X. Wei, J. Wang, J. Tian, *Adv. Mater.* **2021**, *33*, 2006722.
- [47] W. Xu, R. Ji, P. Liu, L. Cheng, L. Zhu, J. Zhang, H. Chen, Y. Tong, C. Zhang, Z. Kuang, H. Zhang, J. Lai, K. Wen, P. Yang, N. Wang, W. Huang, J. Wang, *J. Phys. Chem. Lett.* **2020**, *11*, 10348.
- [48] J. Li, L. Xu, T. Wang, J. Song, J. Chen, J. Xue, Y. Dong, B. Cai, Q. Shan, B. Han, H. Zeng, *Adv. Mater.* **2017**, *29*, 1603885.
- [49] Z. Chen, Y. Hu, J. Wang, Q. Shen, Y. Zhang, C. Ding, Y. Bai, G. Jiang, Z. Li, N. Gaponik, *Chem. Mater.* **2020**, *32*, 1517.
- [50] J. Wang, J. Wang, N. Li, X. Du, J. Ma, C. He, Z. Li, *ACS Appl. Mater. Interfaces* **2020**, *12*, 31477.
- [51] S. T. Ochsenein, F. Krieg, Y. Shynkarenko, G. Rainò, M. V. Kovalenko, *ACS Appl. Mater. Interfaces* **2019**, *11*, 21655.
- [52] Y. S. Shin, Y. J. Yoon, K. T. Lee, J. Jeong, S. Y. Park, G.-H. Kim, J. Y. Kim, *ACS Appl. Mater. Interfaces* **2019**, *11*, 23401.
- [53] F. Ye, H. Zhang, P. Wang, J. Cai, L. Wang, D. Liu, T. Wang, *Chem. Mater.* **2020**, *32*, 3211.
- [54] X. Zheng, S. Yuan, J. Liu, J. Yin, F. Yuan, W.-S. Shen, K. Yao, M. Wei, C. Zhou, K. Song, B.-B. Zhang, Y. Lin, M. N. Hedhili, N. Wehbe, Y. Han, H.-T. Sun, Z.-H. Lu, T. D. Anthopoulos, O. F. Mohammed, E. H. Sargent, L.-S. Liao, O. M. Bakr, *ACS Energy Lett.* **2020**, *5*, 793.
- [55] J. Pan, L. N. Quan, Y. Zhao, W. Peng, B. Murali, S. P. Sarmah, M. Yuan, L. Sinatra, N. M. Alyami, J. Liu, E. Yassitepe, Z. Yang, O. Voznyy, R. Comin, M. N. Hedhili, O. F. Mohammed, Z. H. Lu, D. H. Kim, E. H. Sargent, O. M. Bakr, *Adv. Mater.* **2016**, *28*, 8718.
- [56] J. H. Park, A.-Y. Lee, J. C. Yu, Y. S. Nam, Y. Choi, J. Park, M. H. Song, *ACS Appl. Mater. Interfaces* **2019**, *11*, 8428.
- [57] W. Chen, X. Tang, P. Wangyang, Z. Yao, D. Zhou, F. Chen, S. Li, H. Lin, F. Zeng, D. Wu, K. Sun, M. Li, Y. Huang, W. Hu, Z. Zang, J. Du, *Adv. Opt. Mater.* **2018**, *6*, 1800007.
- [58] Y. Shynkarenko, M. I. Bodnarchuk, C. Bernasconi, Y. Berezovska, V. Verteletskyi, S. T. Ochsenein, M. V. Kovalenko, *ACS Energy Lett.* **2019**, *4*, 2703.
- [59] B.-B. Zhang, S. Yuan, J.-P. Ma, W. Zheng, H. Shen, X.-C. Wang, B. Sun, O. M. Bakr, L.-S. Liao, H.-T. Sun, *J. Am. Chem. Soc.* **2019**, *141*, 15423.
- [60] X. Zheng, Y. Hou, H.-T. Sun, O. F. Mohammed, E. H. Sargent, O. M. Bakr, *J. Phys. Chem. Lett.* **2019**, *10*, 2629.
- [61] S. Zou, Y. Liu, J. Li, C. Liu, R. Feng, F. Jiang, Y. Li, J. Song, H. Zeng, M. Hong, X. Chen, *J. Am. Chem. Soc.* **2017**, *139*, 11443.
- [62] X. Shen, Y. Zhang, S. V. Kershaw, T. Li, C. Wang, X. Zhang, W. Wang, D. Li, Y. Wang, M. Lu, L. Zhang, C. Sun, D. Zhao, G. Qin, X. Bai, W. W. Yu, A. L. Rogach, *Nano Lett.* **2019**, *19*, 1552.
- [63] J. Deng, H. Wang, J. Xun, J. Wang, X. Yang, W. Shen, M. Li, R. He, *Mater. Des.* **2020**, *185*, 108246.
- [64] J.-S. Yao, J. Ge, B.-N. Han, K.-H. Wang, H.-B. Yao, H.-L. Yu, J.-H. Li, B.-S. Zhu, J.-Z. Song, C. Chen, Q. Zhang, H.-B. Zeng, Y. Luo, S.-H. Yu, *J. Am. Chem. Soc.* **2018**, *140*, 3626.
- [65] Z.-J. Yong, S.-Q. Guo, J.-P. Ma, J.-Y. Zhang, Z.-Y. Li, Y.-M. Chen, B.-B. Zhang, Y. Zhou, J. Shu, J.-L. Gu, L.-R. Zheng, O. M. Bakr, H.-T. Sun, *J. Am. Chem. Soc.* **2018**, *140*, 9942.
- [66] G. Pan, X. Bai, W. Xu, X. Chen, Y. Zhai, J. Zhu, H. Shao, N. Ding, L. Xu, B. Dong, Y. Mao, H. Song, *ACS Appl. Mater. Interfaces* **2020**, *12*, 14195.
- [67] D. Rossi, D. Parobek, Y. Dong, D. H. Son, *J. Phys. Chem. C* **2017**, *121*, 17143.
- [68] S. Hou, M. K. Gangishetty, Q. Quan, D. N. Congreve, *Joule* **2018**, *2*, 2421.
- [69] I. L. Braly, R. J. Stoddard, A. Rajagopal, A. R. Uhl, J. K. Katahara, A. K.-Y. Jen, H. W. Hillhouse, *ACS Energy Lett.* **2017**, *2*, 1841.
- [70] Z. Xiao, L. Zhao, N. L. Tran, Y. L. Lin, S. H. Silver, R. A. Kerner, N. Yao, A. Kahn, G. D. Scholes, B. P. Rand, *Nano Lett.* **2017**, *17*, 6863.
- [71] S. Zhang, H. Liu, X. Li, S. Wang, *Nano Energy* **2020**, *77*, 105302.
- [72] F. Meng, X. Liu, X. Cai, Z. Gong, B. Li, W. Xie, M. Li, D. Chen, H.-L. Yip, S.-J. Su, *Nanoscale* **2019**, *11*, 1295.
- [73] F. Yang, H. Chen, R. Zhang, X. Liu, W. Zhang, J. Zhang, F. Gao, L. Wang, *Adv. Funct. Mater.* **2020**, *30*, 1908760.
- [74] G. Li, F. Wisnivesky Rocca Rivarola, N. J. L. K. Davis, S. Bai, T. C. Jellicoe, F. de la Peña, S. Hou, C. Ducati, F. Gao, R. H. Friend, N. C. Greenham, Z.-K. Tan, *Adv. Mater.* **2016**, *28*, 3528.
- [75] E.-P. Yao, Z. Yang, L. Meng, P. Sun, S. Dong, Y. Yang, Y. Yang, *Adv. Mater.* **2017**, *29*, 1606859.
- [76] M. K. Gangishetty, S. Hou, Q. Quan, D. N. Congreve, *Adv. Mater.* **2018**, *30*, 1706226.
- [77] Y. S. Shin, Y. J. Yoon, J. Heo, S. Song, J. W. Kim, S. Y. Park, H. W. Cho, G.-H. Kim, J. Y. Kim, *ACS Appl. Mater. Interfaces* **2020**, *12*, 35740.
- [78] N. Wang, L. Cheng, R. Ge, S. Zhang, Y. Miao, W. Zou, C. Yi, Y. Sun, Y. Cao, R. Yang, Y. Wei, Q. Guo, Y. Ke, M. Yu, Y. Jin, Y. Liu, Q. Ding, D. Di, L. Yang, G. Xing, H. Tian, C. Jin, F. Gao, R. H. Friend, J. Wang, W. Huang, *Nat. Photonics* **2016**, *10*, 699.
- [79] Z. Chen, C. Zhang, X.-F. Jiang, M. Liu, R. Xia, T. Shi, D. Chen, Q. Xue, Y.-J. Zhao, S. Su, H.-L. Yip, Y. Cao, *Adv. Mater.* **2017**, *29*, 1603157.
- [80] D. N. Congreve, M. C. Weidman, M. Seitz, W. Partimongkol, N. S. Dahod, W. A. Tisdale, *ACS Photonics* **2017**, *4*, 476.
- [81] Y. Jiang, C. Qin, M. Cui, T. He, K. Liu, Y. Huang, M. Luo, L. Zhang, H. Xu, S. Li, J. Wei, Z. Liu, H. Wang, G.-H. Kim, M. Yuan, J. Chen, *Nat. Commun.* **2019**, *10*, 1868.
- [82] Q. Wang, X. Wang, Z. Yang, N. Zhou, Y. Deng, J. Zhao, X. Xiao, P. Rudd, A. Moran, Y. Yan, J. Huang, *Nat. Commun.* **2019**, *10*, 5633.
- [83] D. P. McMeekin, Z. Wang, W. Rehman, F. Pulvirenti, J. B. Patel, N. K. Noel, M. B. Johnston, S. R. Marder, L. M. Herz, H. J. Snaith, *Adv. Mater.* **2017**, *29*, 1607039.
- [84] X. Yang, X. Zhang, J. Deng, Z. Chu, Q. Jiang, J. Meng, P. Wang, L. Zhang, Z. Yin, J. You, *Nat. Commun.* **2018**, *9*, 570.
- [85] J. Xing, Y. Zhao, M. Askerka, L. N. Quan, X. Gong, W. Zhao, J. Zhao, H. Tan, G. Long, L. Gao, Z. Yang, O. Voznyy, J. Tang, Z.-H. Lu, Q. Xiong, E. H. Sargent, *Nat. Commun.* **2018**, *9*, 3541.
- [86] S. Yuan, Z.-K. Wang, L.-X. Xiao, C.-F. Zhang, S.-Y. Yang, B.-B. Chen, H.-T. Ge, Q.-S. Tian, Y. Jin, L.-S. Liao, *Adv. Mater.* **2019**, *31*, 1904319.
- [87] T. L. Leung, H. W. Tam, F. Liu, J. Lin, A. M. C. Ng, W. K. Chan, W. Chen, Z. He, I. Lončarić, L. Grisanti, C. Ma, K. S. Wong, Y. S. Lau, F. Zhu, Ž. Skoko, J. Popović, A. B. Djurišić, *Adv. Opt. Mater.* **2020**, *8*, 1901679.
- [88] Y. Jin, Z.-K. Wang, S. Yuan, Q. Wang, C. Qin, K.-L. Wang, C. Dong, M. Li, Y. Liu, L.-S. Liao, *Adv. Funct. Mater.* **2020**, *30*, 1908339.
- [89] Z. Ren, L. Li, J. Yu, R. Ma, X. Xiao, R. Chen, K. Wang, X. W. Sun, W.-J. Yin, W. C. H. Choy, *ACS Energy Lett.* **2020**, *5*, 2569.

- [90] F. Wang, Z. Wang, W. Sun, Z. Wang, Y. Bai, T. Hayat, A. Alsaedi, Z. Tan, *Small* **2020**, 16, 2002940.
- [91] M. Worku, Q. He, L. Xu, J. Hong, R. X. Yang, L. Z. Tan, B. Ma, *ACS Appl. Mater. Interfaces* **2020**, 12, 45056.
- [92] C. Wang, D. Han, J. Wang, Y. Yang, X. Liu, S. Huang, X. Zhang, S. Chang, K. Wu, H. Zhong, *Nat. Commun.* **2020**, 11, 6428.
- [93] H. W. Tam, T. L. Leung, W. Sun, F. Liu, C. Ma, K. S. Wong, I. Lončarić, L. Grisanti, J. Ovc̆ar, Ž. Skoko, J. Popović, A. B. Djurišić, *J. Mater. Chem. C* **2020**, 8, 11052.
- [94] Y.-K. Wang, D. Ma, F. Yuan, K. Singh, J. M. Pina, A. Johnston, Y. Dong, C. Zhou, B. Chen, B. Sun, H. Ebe, J. Fan, M.-J. Sun, Y. Gao, Z.-H. Lu, O. Voznyy, L.-S. Liao, E. H. Sargent, *Nat. Commun.* **2020**, 11, 3674.
- [95] S. Yang, J. Dai, Z. Yu, Y. Shao, Y. Zhou, X. Xiao, X. C. Zeng, J. Huang, *J. Am. Chem. Soc.* **2019**, 141, 5781.
- [96] M. Abdi-Jalebi, Z. Andaji-Garmaroudi, S. Cacovich, C. Stavarakas, B. Philippe, J. M. Richter, M. Alsari, E. P. Booker, E. M. Hutter, A. J. Pearson, S. Lilliu, T. J. Savenije, H. Rensmo, G. Divitini, C. Ducati, R. H. Friend, S. D. Stranks, *Nature* **2018**, 555, 497.
- [97] Z. Tan, J. Luo, L. Yang, X. Li, Z. Deng, L. Gao, H. Chen, J. Li, P. Du, G. Niu, J. Tang, *Adv. Opt. Mater.* **2020**, 8, 1901094.
- [98] D. Ma, P. Todorović, S. Meshkat, M. I. Saidaminov, Y.-K. Wang, B. Chen, P. Li, B. Scheffel, R. Quintero-Bermudez, J. Z. Fan, Y. Dong, B. Sun, C. Xu, C. Zhou, Y. Hou, X. Li, Y. Kang, O. Voznyy, Z.-H. Lu, D. Ban, E. H. Sargent, *J. Am. Chem. Soc.* **2020**, 142, 5126.
- [99] F. Zhang, B. Cai, J. Song, B. Han, B. Zhang, H. Zeng, *Adv. Funct. Mater.* **2020**, 30, 2001732.
- [100] M. Abdi-Jalebi, Z. Andaji-Garmaroudi, S. Cacovich, C. Stavarakas, B. Philippe, J. M. Richter, M. Alsari, E. P. Booker, E. M. Hutter, A. J. Pearson, S. Lilliu, T. J. Savenije, H. Rensmo, G. Divitini, C. Ducati, R. H. Friend, S. D. Stranks, *Nature* **2018**, 555, 497.
- [101] Y. Shen, K.-C. Shen, Y.-Q. Li, M. Guo, J. Wang, Y. Ye, F.-M. Xie, H. Ren, X. Gao, F. Song, J.-X. Tang, *Adv. Funct. Mater.* **2020**, 31, 2006736.
- [102] F. Yuan, C. Ran, L. Zhang, H. Dong, B. Jiao, X. Hou, J. Li, Z. Wu, *ACS Energy Lett.* **2020**, 5, 1062.
- [103] K.-H. Wang, Y. Peng, J. Ge, S. Jiang, B.-S. Zhu, J. Yao, Y.-C. Yin, J.-N. Yang, Q. Zhang, H.-B. Yao, *ACS Photonics* **2019**, 6, 667.
- [104] Y. Liu, J. Cui, K. Du, H. Tian, Z. He, Q. Zhou, Z. Yang, Y. Deng, D. Chen, X. Zuo, Y. Ren, L. Wang, H. Zhu, B. Zhao, D. Di, J. Wang, R. H. Friend, Y. Jin, *Nat. Photonics* **2019**, 13, 760.
- [105] S. Zeng, S. Shi, S. Wang, Y. Xiao, *J. Mater. Chem. C* **2020**, 8, 1319.
- [106] C. H. Jang, A. K. Harit, S. Lee, S. H. Kim, J.-E. Jeong, J. H. Park, E. D. Jung, J. M. Ha, S. K. Kwak, H. Y. Woo, M. H. Song, *ACS Nano* **2020**, 14, 13246.
- [107] L. He, Z. Xiao, X. Yang, Y. Wu, Y. Lian, X. Peng, X. Yang, *J. Mater. Sci.* **2020**, 55, 7691.
- [108] Y. C. Kim, H. J. An, D. H. Kim, J.-M. Myoung, Y. J. Heo, J. H. Cho, *Adv. Funct. Mater.* **2021**, 31, 2005553.
- [109] Z. Ren, X. Xiao, R. Ma, H. Lin, K. Wang, X. W. Sun, W. C. H. Choy, *Adv. Funct. Mater.* **2019**, 29, 1905339.
- [110] N. Yantara, N. F. Jamaludin, B. Febriansyah, D. Giovanni, A. Bruno, C. Soci, T. C. Sum, S. Mhaisalkar, N. Mathews, *ACS Energy Lett.* **2020**, 5, 1593.
- [111] F. Zhang, X. Zhang, C. Wang, M. Sun, X. Luo, Y. Yang, S. Chang, D. Zhang, L. Duan, *Nano Energy* **2021**, 79, 105486.
- [112] Z. Ren, J. Yu, Z. Qin, J. Wang, J. Sun, C. C. S. Chan, S. Ding, K. Wang, R. Chen, K. S. Wong, X. Lu, W.-J. Yin, W. C. H. Choy, *Adv. Mater.* **2021**, 33, 202005570.
- [113] Y. Liu, Z. Yu, S. Chen, J. H. Park, E. D. Jung, S. Lee, K. Kang, S.-J. Ko, J. Lim, M. H. Song, B. Xu, H. J. Snaith, S. H. Park, B. R. Lee, *Nano Energy* **2021**, 80, 105511.
- [114] X.-K. Liu, W. Xu, S. Bai, Y. Jin, J. Wang, R. H. Friend, F. Gao, *Nat. Mater.* **2021**, 20, 10.
- [115] H. Wang, X. Zhao, B. Zhang, Z. Xie, *J. Mater. Chem. C* **2019**, 7, 5596.
- [116] H. Wang, Y. Xu, J. Wu, L. Chen, Q. Yang, B. Zhang, Z. Xie, *J. Phys. Chem. Lett.* **2020**, 11, 1411.
- [117] P. Du, J. Li, L. Wang, J. Liu, S. Li, N. Liu, Y. Li, M. Zhang, L. Gao, Y. Ma, J. Tang, *ACS Appl. Mater. Interfaces* **2019**, 11, 47083.
- [118] H. Cho, S.-H. Jeong, M.-H. Park, Y.-H. Kim, C. Wolf, C.-L. Lee, J. H. Heo, A. Sadhanala, N. Myoung, S. Yoo, S. H. Im, R. H. Friend, T.-W. Lee, *Science* **2015**, 350, 1222.
- [119] Z. Xiao, R. A. Kerner, L. Zhao, N. L. Tran, K. Lee, T.-W. Koh, G. D. Scholes, B. P. Rand, *Nat. Photonics* **2017**, 11, 108.
- [120] G. Li, Z.-K. Tan, D. Di, M. L. Lai, L. Jiang, J. H.-W. Lim, R. H. Friend, N. C. Greenham, *Nano Lett.* **2015**, 15, 2640.
- [121] H. P. Kim, J. Kim, B. S. Kim, H.-M. Kim, J. Kim, A. R. b. M. Yusoff, J. Jang, M. K. Nazeeruddin, *Adv. Opt. Mater.* **2017**, 5, 1600920.
- [122] T. C. Jellicoe, J. M. Richter, H. F. J. Glass, M. Tabachnyk, R. Brady, S. E. Dutton, A. Rao, R. H. Friend, D. Credgington, N. C. Greenham, M. L. Böhm, *J. Am. Chem. Soc.* **2016**, 138, 2941.
- [123] Z. Tan, J. Li, C. Zhang, Z. Li, Q. Hu, Z. Xiao, T. Kamiya, H. Hosono, G. Niu, E. Lifshitz, Y. Cheng, J. Tang, *Adv. Funct. Mater.* **2018**, 28, 1801131.
- [124] M. Leng, Y. Yang, Z. Chen, W. Gao, J. Zhang, G. Niu, D. Li, H. Song, J. Zhang, S. Jin, J. Tang, *Nano Lett.* **2018**, 18, 6076.
- [125] J. Zhang, Y. Yang, H. Deng, U. Farooq, X. Yang, J. Khan, J. Tang, H. Song, *ACS Nano* **2017**, 11, 9294.
- [126] T. Jun, K. Sim, S. Iimura, M. Sasase, H. Kamioka, J. Kim, H. Hosono, *Adv. Mater.* **2018**, 30, 1804547.
- [127] L. Wang, Z. Shi, Z. Ma, D. Yang, F. Zhang, X. Ji, M. Wang, X. Chen, G. Na, S. Chen, D. Wu, Y. Zhang, X. Li, L. Zhang, C. Shan, *Nano Lett.* **2020**, 20, 3568.
- [128] Z. Ma, Z. Shi, D. Yang, F. Zhang, S. Li, L. Wang, D. Wu, Y. Zhang, G. Na, L. Zhang, X. Li, Y. Zhang, C. Shan, *ACS Energy Lett.* **2020**, 5, 385.
- [129] L. Gu, K. Wen, Q. Peng, W. Huang, J. Wang, *Small* **2020**, 16, 2001861.
- [130] Q. Zhang, D. Zhang, Y. Fu, S. Poddar, L. Shu, X. Mo, Z. Fan, *Adv. Funct. Mater.* **2020**, 30, 2002570.
- [131] Q. Zhang, M. M. Tavakoli, L. Gu, D. Zhang, L. Tang, Y. Gao, J. Guo, Y. Lin, S.-F. Leung, S. Poddar, Y. Fu, Z. Fan, *Nat. Commun.* **2019**, 10, 727.
- [132] Y. Shen, L.-P. Cheng, Y.-Q. Li, W. Li, J.-D. Chen, S.-T. Lee, J.-X. Tang, *Adv. Mater.* **2019**, 31, 1901517.
- [133] H. Cho, Y.-H. Kim, C. Wolf, H.-D. Lee, T.-W. Lee, *Adv. Mater.* **2018**, 30, 1704587.
- [134] W. Zou, R. Li, S. Zhang, Y. Liu, N. Wang, Y. Cao, Y. Miao, M. Xu, Q. Guo, D. Di, L. Zhang, C. Yi, F. Gao, R. H. Friend, J. Wang, W. Huang, *Nat. Commun.* **2018**, 9, 608.
- [135] Y. Lei, Y. Chen, Y. Gu, C. Wang, Z. Huang, H. Qian, J. Nie, G. Hollett, W. Choi, Y. Yu, N. Kim, C. Wang, T. Zhang, H. Hu, Y. Zhang, X. Li, Y. Li, W. Shi, Z. Liu, M. J. Sailor, L. Dong, Y.-H. Lo, J. Luo, S. Xu, *Adv. Mater.* **2018**, 30, 1705992.
- [136] H. Chen, J. Lin, J. Kang, Q. Kong, D. Lu, J. Kang, M. Lai, L. N. Quan, Z. Lin, J. Jin, L. Wang, M. F. Toney, P. Yang, *Sci. Adv.* **2020**, 6, eaay4045.



Zhenwei Ren is currently a postdoctoral fellow in the Department of Electrical and Electronic Engineering at the University of Hong Kong. He received his Ph.D. in 2018 from East China University of Science and Technology. His current research interests mainly focus on the design and synthesis of nanocrystals, perovskites, and metal oxides as well as their application in light-emitting diodes, photodetectors, and solar cells.



Kai Wang is an Associate Professor at the Southern University of Science and Technology, China. His research focuses on quantum dots, perovskites, and their applications in light-emitting diodes and photodetectors. He has published over 150 peer-reviewed papers with an H-Index of 33, 1 academic book, and authorized 25 granted invention patents in China and 2 in USA. His research achievement was being awarded the second prize of the National Award for Technological Invention and the first prize of the National Ministry of Education Award for Technological Innovation.



Xiao Wei Sun is a Chair Professor in the Southern University of Science and Technology, China. He was previously a Full Professor at Nanyang Technological University, Singapore. He is an Honorary Doctor of the Belarusian State University of Informatics and Radioelectronics. He is an academican of the Asia-Pacific Academy of Materials, and Fellow of OSA, SPIE, SID, and Institute of Physics. He is a distinguished lecturer of IEEE. His main research presently is on semiconductor nanocrystals for power-saving high-quality displays. Professor Sun has more than 500 publications with an H-index of 77. He is an Elsevier Highly Cited Scholar.



Wallace C. H. Choy is a Full Professor in the Department of Electrical and Electronic Engineering at the University of Hong Kong. His research interests cover organic/inorganic optoelectronic devices, plasmonic structures, metal oxides, and nanomaterial devices. He has published more than 220 peer-reviewed papers, several book chapters, patents, and edited one book. He was recognized as being in the top 1% of most-cited scientists in Thomson Reuter's Essential Science Indicators from 2014 to 2019. He is named as one of the "Highly Cited Researchers" by Clarivate Analytics in 2018. He is an elected fellow of OSA.