

Co-Interlayer Engineering for Homogenous Phase Quasi-2-D Perovskite and High-Performance Deep-Blue Light-Emitting Diodes

Ning Zhou, Guangren Na, Dongyu Li, Zhengyan Jiang, Xiangtian Xiao, Hong Lin, Zhijun Zhang, and Wallace C.H Choy*

Organic and inorganic hybrid perovskites are a class of solution-processable semiconductors that hold significant promise for light-emitting diode applications. However, the development of efficient blue perovskite light-emitting diodes (PeLEDs) has remained a critical challenge. Although significant advancements have been achieved in the development of sky-blue PeLEDs, the performance of pure-blue and deep-blue PeLEDs still lags. In this manuscript, by incorporating 1-cyclohexene-1-ethanamine cation (CEA⁺) into the host 1-phenylbutylamine cation (PBA⁺) spacer and fabricating mixed-cations quasi-2D perovskites, a homogeneous phase distribution in quasi-2D perovskite films is achieved through precise tuning of the phase formation energy. The resulting PeLEDs exhibit remarkable performance with an external quantum efficiency (EQE) of 5.8% at 467 nm, which is one of the highest EQEs among the deep-blue light quasi-2D PeLED devices with emission wavelengths below 470 nm. Additionally, the device demonstrates a half-lifetime of ≈ 25 min. Furthermore, the PeLEDs utilizing mixed-cation perovskite exhibit a stable electroluminescence (EL) spectrum under varying voltage biases, even when the chlorine content in the halogen sites reaches as high as 30%. Overall, this work offers a novel and promising avenue to achieve both high EQE and a stable EL spectrum in blue-light quasi-2D PeLEDs.

1. Introduction

Organic and inorganic perovskites have garnered significant attention in the fields of LEDs because of their outstanding optoelectronic properties such as adjustable bandgap, high color purity, and high photoluminescence quantum yield (PLQY).^[1,2] Meanwhile, perovskite materials can be easily fabricated by low-cost and simple manufacturing processes. Owing to their excellent performances, PeLEDs have a great potential for light display applications. The external quantum efficiencies (EQE) of PeLEDs, including near-infrared, red-emitting, and green-emitting devices, have surpassed 20%, indicating a promising future for practical applications.^[3–7] However, the efficiency of deep-blue light emission, characterized by a wavelength shorter than 470 nm still lags behind that of red and green light-emitting devices. It is challenging to fabricate deep-blue PeLEDs with good operational stability against continuous bias current and spectral stability under different applied voltages with high EQE.

Several strategies have been applied to realize blue light-emitting PeLEDs. One effective approach is incorporating chlorine into bromine-based perovskites to form mixed-halide perovskite with an enlarged bandgap.^[8] However, it should be noted that Br-Cl mixture perovskites can exhibit phase separation when exposed to light or under continuous electrical bias, leading to peak shifts or multi-peaks during light emission. It has been reported that the electroluminescence (EL) spectra of corresponding PeLEDs gradually shift toward the green light region due to halide migration, which is observed in almost all mixed-halide (Cl and Br) perovskite-based LEDs.^[9,10]

Another efficient strategy to achieve blue light-emitting films is to introduce large organic cations to obtain 2D or quasi-2D perovskites ($A_2B_{n-1}Pb_nX_{3n+1}$, where $A=PEA^+$, BA^+ , PBA^+ ; $B=CH_3NH_3^+$, $CH(NH_2)_2^+$, Cs^+ ; $X=Cl^-$, Br^- , I^-). By reducing the phase number n (i.e., the number of perovskite layers) of 2D perovskites to a low value, the emission spectra exhibit a noticeable blue shift. However, these 2D or quasi-2D blue PeLEDs typically have low EQE, accompanied by a wide EL peak and inferior

N. Zhou, D. Li, Z. Jiang, X. Xiao, H. Lin, W. C. Choy
Department of Electrical and Electronic Engineering
The University of Hong Kong
Hong Kong, China
E-mail: chchoy@eee.hku.hk

G. Na, Z. Zhang
State Key Laboratory on Integrated Optoelectronics
Key Laboratory of Automobile Materials of MOE, and College of Materials
Science and Engineering
Jilin University
Changchun 130012, China

The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/adom.202302273>

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DOI: 10.1002/adom.202302273

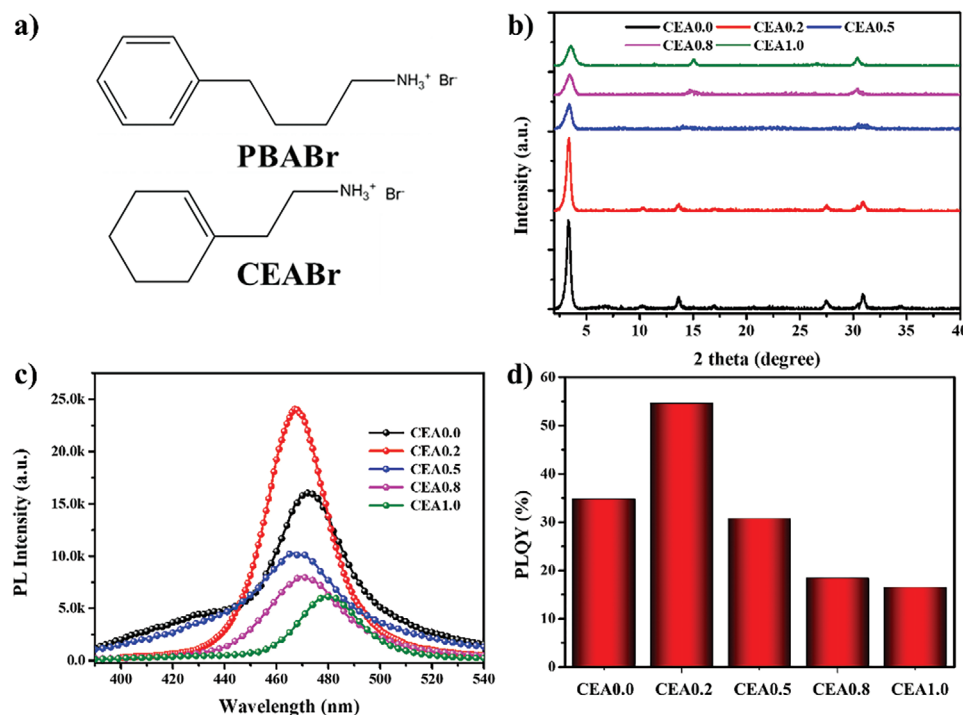


Figure 1. a) The chemical structures of PBABr and CEABr molecules; b) XRD pattern of quasi-2D perovskite thin films with different CEA ratios; c) steady-state PL spectra of quasi-2D perovskite layers on glass with varying ratios of CEA; d) PLQY of the quasi-2D perovskite films with varying ratios of CEA.

operational stability. These issues are primarily attributed to Ostwald ripening, broad 2D phase diffusion, and the presence of insulative organic additives.^[11–13] Sargent and his team have achieved a significant milestone by successfully fabricating sky-blue PeLEDs with good stability and a maximum EQE of $\approx 1.5\%$ using a homologous quasi-2D approach.^[14] Subsequently, pure blue and deep-blue photoluminescent (PL) emission-stable PeLEDs have been reported by incorporating different spacer cations.^[15–20] Moreover, mixed spacer cation engineering has proven to be an effective strategy for manipulating the optoelectronic properties and enhancing the performance of perovskite-based LEDs and solar cells.^[21,22]

Herein, we present a novel approach to develop highly stable and efficient deep-blue PeLEDs by simultaneously incorporating two different ligands, PBABr (phenylbutylammonium bromide) and CEABr (2-(1-cyclohexenyl)ethylaminium bromide), into the perovskite precursor solution. Our findings demonstrate that by introducing an appropriate ratio of CEABr to PBABr in quasi-2D perovskites, the photoluminescence quantum yield (PLQY) of the perovskite film can be significantly enhanced. Furthermore, this dual-ligand strategy leads to the formation of perovskite films with homogeneous phase distribution and a compact, smooth morphology. The PeLEDs fabricated using these dual-ligand incorporated perovskite films exhibit excellent performance. They achieve a peak EQE of 5.8% at 467 nm and a T50 lifetime of 25 min, which represents one of the highest EQEs among deep-blue light quasi-2D PeLED devices with emission wavelengths shorter than 470 nm. Notably, the EL spectrum remains unchanged even under different applied voltages and during device operation.

2. Results and Discussion

To obtain a low phase number with an average stoichiometry of $\langle n \rangle = 3$ in the quasi-2D perovskites family $(\text{PBA}_{1-x}\text{CEA}_x)_2(\text{Cs}_{0.8}\text{FA}_{0.2})_2\text{Pb}_3(\text{Br}_{0.5}\text{Cl}_{0.5})_{10}$, where $x = 0, 0.2, 0.5, 0.8$, and 1.0 , we strategically replace CEA^+ with PBA^+ according to the specified stoichiometric ratio. The detailed fabrication process is described in the experimental section. For simplicity in representing the chemical formula, we use CEA0.0, CEA0.2, CEA0.5, CEA0.8, and CEA1.0 to denote $(\text{PBA}_{1-x}\text{CEA}_x)_2(\text{Cs}_{0.8}\text{FA}_{0.2})_2\text{Pb}_3(\text{Br}_{0.5}\text{Cl}_{0.5})_{10}$, where $x = 0, 0.2, 0.5, 0.8$, and 1.0 , respectively, unless stated otherwise. The chemical structures of PBABr and CEABr are provided in Figure 1a. The crystal phase and orientation of the $(\text{PBA}_{1-x}\text{CEA}_x)_2(\text{Cs}_{0.8}\text{FA}_{0.2})_2\text{Pb}_3(\text{Br}_{0.5}\text{Cl}_{0.5})_{10}$ perovskite films were investigated using X-ray diffraction (XRD) measurement. The dominant 2D diffraction peak was observed at a diffraction angles (2θ) of 3.28° for the CEA0.0 film, as shown in Figure 1b. When CEA^+ partially or completely replaced PBA^+ in the perovskite films, the diffraction angle shifted toward higher angles. This shift can be attributed to the reduction in crystal plane spacing due to the incorporation of CEA^+ . Specifically, for diffraction angles near 3° , the peak position shifted from 3.28° to 3.58° as the CEA ratio increased from CEA0.0 to CEA1.0. This observation indicates that the presence of CEA^+ affects the crystallinity of the perovskite films compared to films dominated by PBA^+ . The decreased crystallinity suggests that CEA likely influences the crystallization kinetics during the film formation process. The results suggest that the incorporation of CEA^+ leads to alterations in the crystal structure and

potentially affects the optoelectronic properties of the perovskite films.

Regarding the optical properties, the photoluminescence (PL) emission properties of $(\text{PBA}_{1-x}\text{CEA}_x)_2(\text{Cs}_{0.8}\text{FA}_{0.2})_2\text{Pb}_3(\text{Br}_{0.5}\text{Cl}_{0.5})_{10}$ perovskite films were studied. The films were prepared using a spin-coating technique on glass substrates, as detailed in the experimental section. Under laser beam excitation, the perovskite films exhibited distinct PL emission peaks as depicted in Figure 1c. The PL emission peak observed a blue-shift from 472 to 467 nm when transitioning from the CEA0.0 to CEA0.2 films, respectively. When the ratio of CEA incorporation further increased beyond 0.2, a gradual red shift in the PL emission was observed. This red shift indicates a reduction in the bandgap of the perovskite films due to the introduction of CEA in amounts exceeding 20%. This effect is likely a consequence of the shorter spacer layer caused by the smaller ionic radius of the CEA^+ cation. Moreover, the observed blue shift of the PL emission peak in the CEA0.2 film can be ascribed to the disturbance of the phase distribution caused by the incorporation of a small ratio of CEA while the primary spacer cations are still PBA^+ , which will be described in the following in details.

Figure S1 (Supporting Information) illustrates the absorption spectra of the perovskite films. The absorption peaks observed in the spectra correspond to quasi-2D perovskites with different n numbers. The presence of these peaks indicates the existence of domains from $n = 1$ to effectively 3D perovskites within the films. The pure PBA-based film (CEA0.0) exhibits a strong tendency to form $n = 1$ and $n = 2$ phases due to the π - π stacking behavior between PBA molecules.^[14,23] This behavior is known to promote the formation of low “ n ” phases. When a significant portion of the bulky organic cations is consumed, leaving behind short-chain precursors, high n phases are favored during nucleation. As a result, mixed phases with an inhomogeneous phase distribution may form. The high- n components display longer wavelength emissions due to efficient carrier funneling toward the phase with the smallest-bandgap. This phenomenon can explain the blue shift of the PL emission peak in the CEA0.2 film. Interestingly, the CEA0.2 film exhibits a significantly reduced presence of $n = 1$ and $n = 2$ phases. This can be attributed to the disruptive effect of CEA on the stacking behavior between PBA molecules, leading to a more homogeneous phase distribution throughout the film. In films dominated by CEA, there is a tendency to form $n = 2$ dominated phases but less $n = 3$ phases. For instance, the CEA0.5 film lacks $n = 1$, but exhibits an increased presence of $n = 2$ phases due to the tendency to form $n = 2$ phase for high CEA ratio films. To further understand the influence of CEA on the formation of different “ n ” value phases in quasi-2D perovskites, Density functional theory (DFT) simulations were conducted. The formation energy of different n values for the combination of PBA and CEA was calculated (Figure S2a, Supporting Information). The simulation revealed that the energies required for forming 2D CEA0.2 ($n = 1$) and CEA0.2 ($n = 2$) were notably higher compared to those for higher CEA ratios and pure PBA or CEA-based perovskites. This suggests a smaller ratio of low “ n ” value phases in CEA0.2-based quasi-2D perovskite structures, resulting in a more homogeneous phase film. However, a higher ratio of CEA may promote the formation of low n -value phases and contribute to a less homogeneous film. It implies that

the incorporation of specific CEA cations can influence the formation energy of different n -value phases in quasi-2D perovskite, resulting in a distinct phase distribution. In addition, DFT calculations were used to determine the interlayer interaction energy between PBA and CEA, as shown in Figure S2b (Supporting Information). The results showed that the interlayer interaction for different n -value phases can be tuned by CEA. To investigate the energy level values of the valence band maximum (VBM) and conduction band minimum (CBM) in the quasi-2D perovskites with different CEA ratios, UV photoelectron spectroscopy (UPS) measurements were conducted, which was depicted in Figure S3 (Supporting Information). Table S1 (Supporting Information) provides a summary of the photoemission parameters. The altered energy bands observed in the UPS measurements indicate changes in the crystal structure and optoelectronic properties. In Figure 1d, the PLQY of the five different perovskite films was measured. Despite the presence of the $n = 2$ phase in the films, a significant enhancement in PLQY was observed. This enhancement can be attributed to the high exciton binding energy of the low n -value phase. Specifically, the CEA0.2 sample exhibited a PLQY of $\approx 54.6\%$, which is considerably higher than that of the CEA0.0 sample (34.8%) and the CEA0.5 sample (30.7%). Based on the promising results obtained, the CEA0.2 film was chosen as the target film for further investigation of its optoelectronic behavior. Additionally, blue light PeLEDs were fabricated using the CEA0.2 film as a key component.

Figure S4 (Supporting Information) demonstrates that both the CEA0.0 and CEA0.2 films exhibit a smooth morphology with densely packed grains. This observation is consistent with the strong space-confined effect typically observed in quasi-2D perovskite. The densely packed grains and smooth morphology indicate a well-organized and high-quality film structure. Furthermore, Figure S5 (Supporting Information) presents the atomic force microscopy (AFM) images of perovskite films. The root-mean-square (RMS) roughness values of CEA0.0 and CEA0.2 films were determined to be 0.462 and 0.357 nm, respectively, indicating a smoother and more uniform surface compared to the CEA0.2.

To investigate carrier recombination and energy transfer dynamics, transient absorption (TA) spectrum measurement is carried out in two samples: CEA0.0 perovskite film (Figure 2a) and CEA0.2 perovskite film (Figure 2b). A series of ground state bleach peaks at 436, 454, and 472 nm corresponding to the single phase of $n = 2$, 3, and higher than 3 (∞), respectively, are observed in the CEA0.0 perovskite film (Figure 2a). This indicates the presence of different phases within the perovskite film. In contrast, the CEA0.2 perovskite film (Figure 2b) shows only two obvious bleaching peaks at 450 and 467 nm, implying a relatively purer perovskite phase. This could be attributed to the interruption of stacking between PBA molecules, which tend to group together due to π - π interactions, by incorporating the CEABr ligand. The overall peak locations in the TA spectra are consistent with the steady-state absorption spectrum shown in Figure S1 (Supporting Information). By analyzing TA spectra with different delay times as depicted in Figure 2a, it is observed that photoexcited carriers are initially generated in the $n = 2$ and 3 domains of the perovskite material. The carriers in the $n = 2$ phase rapidly diminish, while those in the $n = 3$ phase diminish at a slower rate. This observation suggests that there is a transfer from $n = 2$ to

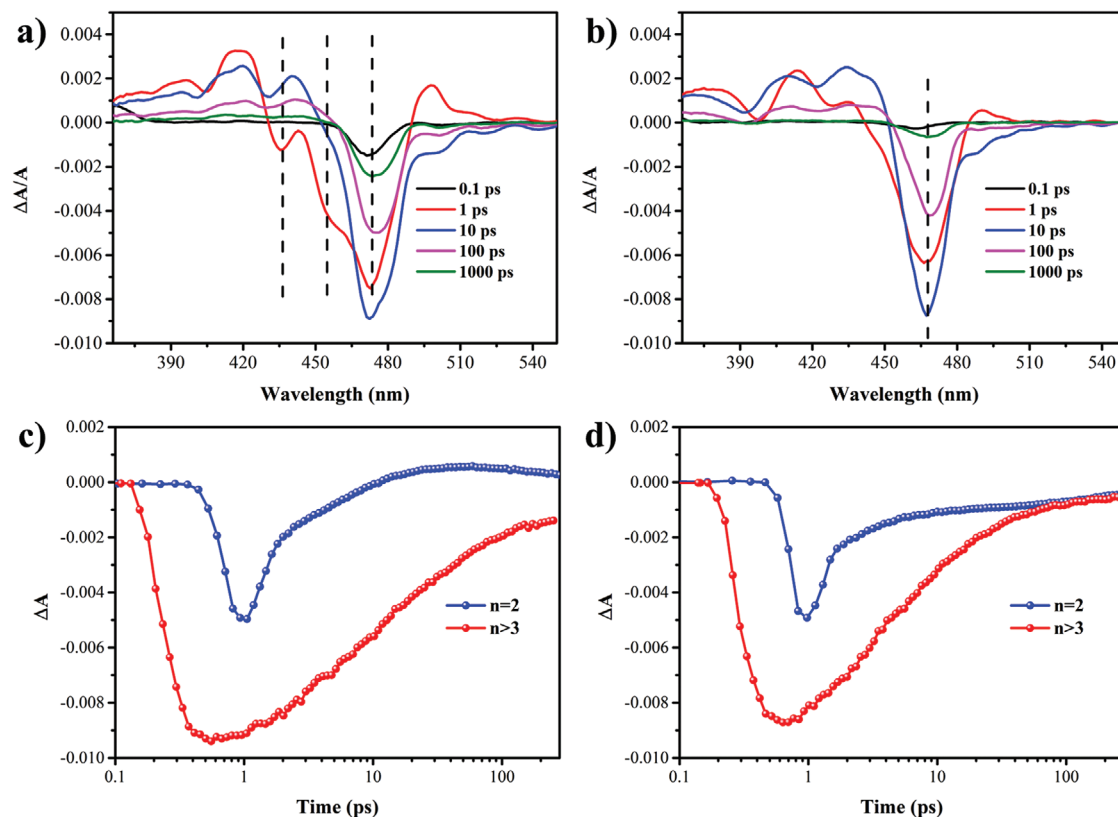


Figure 2. a) The obtained TA spectra of CEA0.0 and b) CEA0.2 quasi-2D perovskite film at different pump–probe delays in the response of the probe wavelength. c) Decay kinetics at $n = 2$ and $n > 3$ domains of CEA0.0 and d) CEA0.2 quasi-2D perovskite films.

$n = 3$ occurred during this process. A similar transfer process is also observed in the CEA0.2 film. (Figure 2b). The TA measurements provide insights into the dynamics of exciton bleaching in the CEA0.0 and CEA0.2 samples (Figure 2c,d). The evolution of ground-state bleach intensity was analyzed by fitting it with a multiexponential function, and the resulting fitting parameters are listed in Table 1. In the $n > 3$ phase of the perovskite material, the behavior of energy transfer and carrier recombination can be described by three distinct processes. Among these processes, τ_1 is associated with the energy acceptance from low n phases, τ_2 corresponds to the overlap of energy acceptance and energy decay within the $n > 3$ phases, and τ_3 represents the recombination of exciton in the $n > 3$ phases.^[24] The fitting results indicate that the τ_1 of the CEA0.2 sample (0.36 ps) is ≈ 2 times shorter than that of the CEA0.0 film (0.71 ps), indicating faster energy transfer into high- n phases for the CEA0.2 sample. The accelerated energy transfer process leads to reduced energy loss and enhances the concentration of carriers in the emitting center. The elevated carrier concentration induces a higher-order recombina-

tion process for luminescence, particularly bimolecular radiative recombination, resulting in a faster depletion of carriers. These findings align with the TRPL analysis depicted in Figure S6 (Supporting Information).

Meanwhile, temperature-dependent PL measurements were conducted to investigate the carrier behavior of CEA0.0 and CEA0.2 perovskite films. The results are presented in Figure S7 (Supporting Information), and the temperature-dependent PL mapping is displayed in Figure 3a,b. By examining the temperature dependence of the integral PL intensity and analyzing the resulting emission quenching behavior, it is possible to extract the exciton binding energy. The exciton binding energy is a crucial factor that significantly impacts the efficiency of exciton recombination and charge transfer. Figure 3c illustrates the variation of the integral photoluminescence (PL) intensity with temperature. The fitting procedure follows the Equation (1):

$$I_T = \frac{I_0}{1 + Ae^{-E_B/k_B T}} \quad (1)$$

where I_T represents the integral PL intensity, I_0 is the intensity at 0 K, E_B denotes the exciton binding energy, and K_B represents the Boltzmann constant. By fitting the experimental data to Equation 1, the E_B is determined to be 105.4 meV for the CEA0.0 film and 123.8 meV for the CEA0.2 film, respectively. This implies that the interruption of π – π interaction, which occurs due to the incorporation of the CEABr ligand, may affect the exciton

Table 1. The fitted TA (Experimental) Lifetime for $n = 2$ and $n > 3$ phase.

	τ_1 [$n = 2$]	τ_1 [$n > 3$]	τ_2 [$n = 2$]	τ_2 [$n > 3$]	τ_3 [$n = 3$]	τ_3 [$n > 3$]
CEA 0.0	0.15 ps	0.71 ps	7.6 ps	14.3 ps	25.3 ps	259 ps
CEA 0.2	0.27 ps	0.36 ps	10.9 ps	11.8 ps	46.2 ps	187 ps

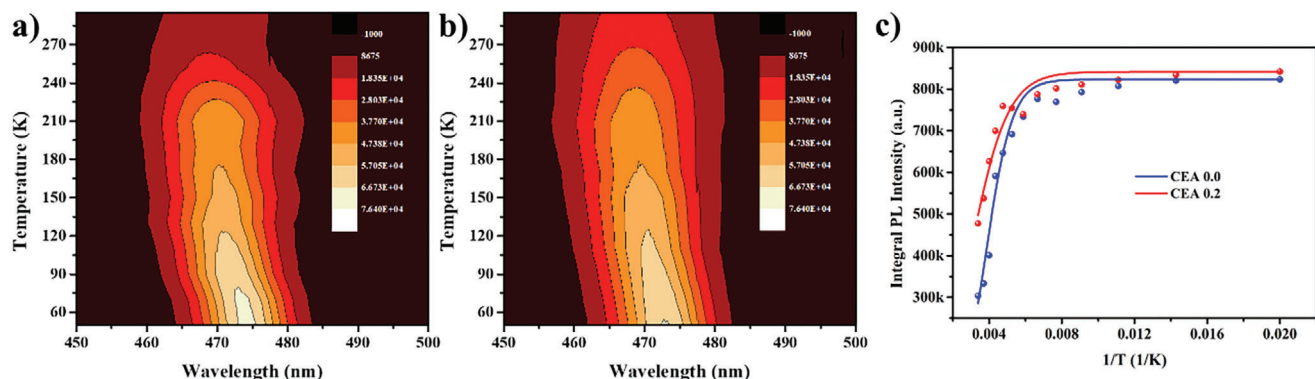


Figure 3. a) and b) Temperature-dependent PL mapping for CEA0.0 and CEA0.2, respectively. c) Temperature-dependent data of integrated intensity for CEA0.0 and CEA0.2, respectively.

behavior in the perovskite film. The larger binding energy of excitons implies that carrier recombination is more efficient, which may contribute to a higher PLQY.

PeLED devices with various perovskite layers were fabricated using the ITO/PEDOT:PSS/TFB/PVK/perovskite/TPBi/LiF/Al structure, as depicted in Figure 4a. The CEA0.0-based devices exhibited an EL spectrum with a peak at 472 nm, while the CEA0.2-based device showed stable blue emission with a peak at 467 nm (Figure 4b). The corresponding CIE coordinates were (0.128, 0.135) and (0.128, 0.084), respectively (Figure 4c). The CEA0.0-based devices generally had lower performance compared to the CEA0.2-based devices. At 6 V, the devices utilizing CEA0.0 as the perovskite layer achieved a luminance of 51.5 cd m⁻² and a

peak EQE of 4.0%. On the other hand, the CEA0.2-based devices demonstrated a higher luminance of 163.2 cd m⁻² at the bias voltage of 6 V (Figure 4d), and a peak EQE of 5.8% (Figure 4e). Figure 4f shows statistical EQEs of the CEA0.0 and CEA0.2-based devices, which clearly demonstrates the efficiency enhancement by incorporating 20% CEA in the perovskites. Figure S8 (Supporting Information) shows a typical EL image at different voltages for both devices. Notably, the CEA0.2 devices exhibited good spectral stability, as the EL spectrum maintained consistent peak location and full width at half-maximum (≈ 25 nm) even with an increase in bias from 2 to 8 V (Figure S8, Supporting Information). Additionally, no obvious changes were observed in the shape of the EL spectrum. Regarding operational stability,

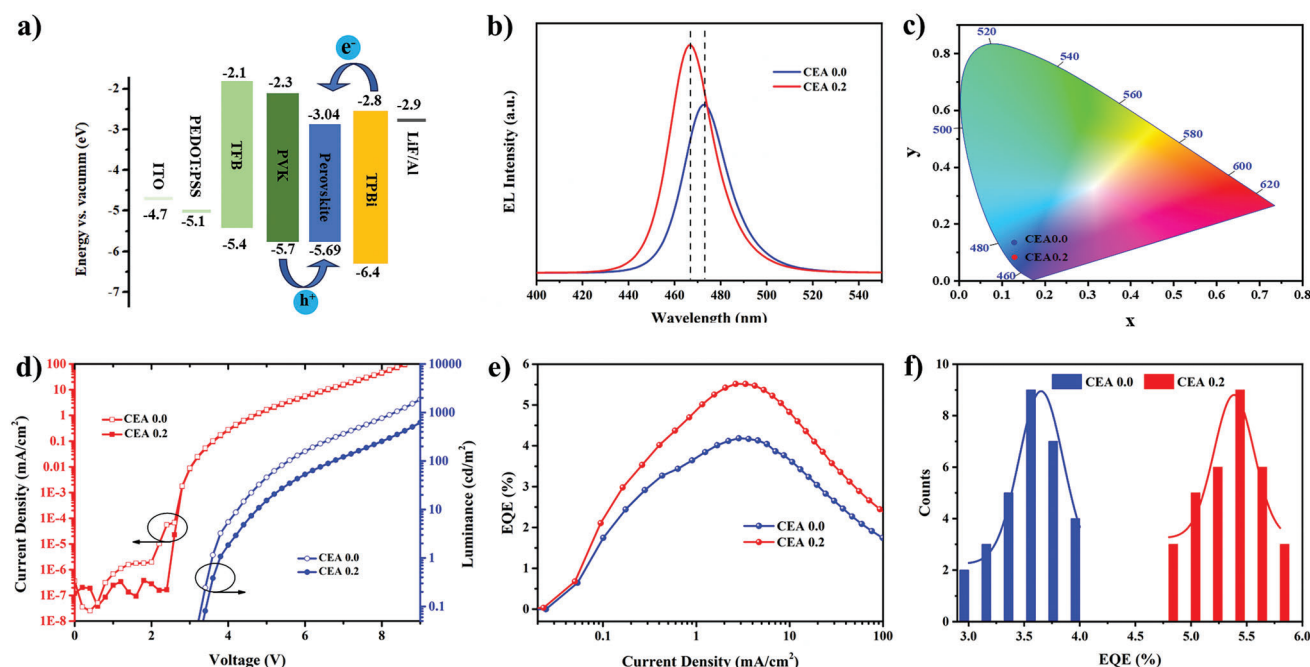


Figure 4. a) Energy-level diagram of the as-fabricated perovskite LED. b) Electroluminescence spectra of the three as-fabricated PeLEDs. c) CIE coordinates of CEA0.0 and CEA0.2 based perovskite LEDs. d) L–V and J–V curves of the best-performing perovskite LED for CEA0.0 and CEA0.2 based PeLEDs. e) EQE curves of the best-performing perovskite LED for CEA0.0 and CEA0.2 based PeLEDs. f) The histogram of the maximum EQE replace; by CEA0.0 and CEA0.2 based PeLEDs.

Figure S9 (Supporting Information) shows that the CEA0.0 devices, starting with an initial brightness of 29.5 cd m^{-2} , had a half lifetime (T_{50}) of $\approx 18 \text{ min}$ under continuous operation. In comparison, the CEA0.2 devices showed a higher initial brightness of 99.6 cd m^{-2} at the same bias condition as the CEA0.0 devices, and the T_{50} extend to $\approx 25 \text{ min}$.

3. Conclusion

In summary, this study presented a significant advancement in the fabrication of quasi-2D perovskite films using a co-interlayer approach. This method enabled the successful creation of deep-blue PeLEDs with excellent efficiency and remarkable spectral and operational stabilities. The incorporation of the appropriate ratio of CEA significantly enhanced the PLQY of the perovskite film. Additionally, the dual-ligand treatment resulted in a compact perovskite film with a smooth surface and a blue-shifted PL emission, with a peak wavelength below 470 nm . The investigation of energy transfer and exciton recombination dynamics revealed that the dual-ligand treatment facilitated efficient carrier transfer and radiative recombination, thereby promoting the emission performance of the PeLEDs. The fabricated blue PeLEDs exhibited outstanding device performance, including a peak EQE of 5.8% and a T_{50} lifetime of 25 min . Moreover, the devices exhibited remarkable spectral stability, with no observable changes in the EL spectra across a wide range of applied biases, from 2 to 8 V . This work presents a novel and promising approach for the fabrication of stable and efficient deep-blue PeLEDs, offering potential applications in display technologies.

4. Experimental Section

Materials: All the chemicals used in this study were utilized as received without undergoing additional purification processes. Formamidinium iodide (FABr), cesium bromide (CsBr), lead bromide (PbBr_2), lead chloride (PbCl_2), phenylbutylammonium bromide (PBABr) and 2-(1-cyclohexenyl)ethylammonium bromide (CEABr) were purchased from Xi'an Polymer Light Technology Corp. Dimethyl sulfoxide (DMSO) and ethyl acetate (EA) were purchased from Sigma-Aldrich. Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) was obtained from Heraeus, while Poly(9,9-dioctylfluorene-co-N-(4-(3-methylpropyl)) diphenylamine) (TFB) with an average molecular weight of $\approx 50,000 \text{ g mol}^{-1}$ was purchased from Luminescence Technology Corp. Poly(9-vinylcarbazole) (PVK) were obtained from Xi'an Polymer Light Technology Corp.

Solution Preparation: The perovskite emissive layers were prepared using a perovskite precursor solution consisting of PBABr, CsBr, FABr, PbBr_2 , and PbCl_2 with a molar ratio of $1.1:0.8:0.2:0.6:0.4$ dissolved in DMSO. To form the co-interlayer perovskite precursor, CEABr was introduced to replace PBABr at a specific molar ratio. The molar concentration for $\text{PbBr}_2 + \text{PbCl}_2$ in the precursor solution was 0.15 M . The device structure of the PeLED was as follows: ITO/PEDOT:PSS/TFB/PVK/perovskite/TPBi/LiF/Al. The ITO-coated glass substrates were prepared by spin-coating them with a layer of PEDOT:PSS. The spin-coating process was performed at a speed of 4000 rpm for a duration of 30 s . Afterward, the coated substrates were annealed in air at a temperature of 150°C for a period of 10 min . The substrates were then transferred into an N_2 glovebox. Next, TFB dissolved in toluene at a concentration of 4 mg mL^{-1} was spin-coated onto the PEDOT:PSS layer at a speed of 5000 rpm for 30 s . The coated films were then annealed at 100°C for 10 min . Subsequently, a solution of PVK dissolved in chlorobenzene with a concentration of 8 mg mL^{-1} was spin-coated onto the TFB layer. The

spin-coating process was performed at a speed of 5000 rpm for a duration of 30 s . The resulting coated films were then annealed again at 100°C for 10 min . The perovskite films were prepared by spin-coating the precursor solution at a speed of 5000 rpm for 90 s . At the end of the 30 s , $200 \mu\text{L}$ of ethyl acetate was added as anti-solvent. Finally, TPBi (40 nm), LiF (1 nm), and Al electrodes (100 nm) were deposited onto the perovskite film by a thermal evaporation system through a shadow mask. The deposition process was carried out under a base pressure of 10^{-5} Pa .

Characterizations: The surface morphologies of the films were characterized using two different techniques: field emission scanning electron microscopy (FE-SEM) with a Hitachi S-4800 scanning electron microscope and atomic force microscopy (AFM) with an NT-MDT NTEGRA system. To measure the PL and TRPL spectra of the perovskite films, a PicoQuant FluoTime 300 system equipped with a picosecond pulse laser (360 nm , LDH-P-C-360) was used. UV-vis absorption spectrum was measured with a Shimadzu UV-vis Spectrophotometer (UV-2600). The integrated sphere method was employed to determine the PLQY of the perovskite films. During the measurement, an excitation power density of $3.63 \mu\text{W cm}^{-2}$ was utilized. TA spectra were acquired using the ExciPro XL Femtosecond Transient Absorption Pump-Probe Spectrometer from CDP Systems. For this measurement, the perovskite films were excited using a femtosecond laser pulse at 365 nm . To evaluate the device's performance, various measurements were conducted using a spectrophotometer coupled with a Keithley 2400 instrument, including current-voltage (I - V) characteristics, luminescence, current efficiency, EQE, and EL spectra. All of these measurements were carried out within a glove box full of nitrogen. The formation energy of the perovskites was calculated using ground state DFT with the Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation (GGA) exchange-correlation functional. The calculations were performed using the Vienna ab initio simulation package (VASP). To obtain the formation energy, the following formula was used:

$$E_F = (E_{(\text{PBA}_{1-x}\text{CEA}_x)_2(\text{Cs})_{n-1}\text{Pb}_{n+1}} - nE_{\text{PbBr}_2} - xE_{\text{CEABr}} - (1-x)E_{\text{PBABr}} - (n-1)E_{\text{CsBr}}) / n \quad (2)$$

To calculate the interlayer interaction energy of the perovskites, it can be determined using the following formula:

$$E_I = E_{\text{RP}(\text{PBA}_{1-x}\text{CEA}_x)_2(\text{Cs})_{n-1}\text{Pb}_{n+1}} - E_{2\text{D}(\text{PBA}_{1-x}\text{CEA}_x)_2(\text{Cs})_{n-1}\text{Pb}_{n+1}} \quad (3)$$

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

This work has received generous support from various funding sources: University Grant Council of the University of Hong Kong Grants (Grant# 202011159254; Grant# 202111159113), General Research Fund (GRF) (Grant# 17211220; Grant# 17200021; Grant# 17200823), and Collaborative Research Fund (CRF) from the Research Grants Council (RGC) of Hong Kong Special Administrative Region, China (Grant No. C7035-20G).

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

deep blue light, light-emitting diodes, perovskites

Received: January 17, 2024
Revised: March 20, 2024
Published online: April 9, 2024

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