

Ionic Liquid Passivation for High-Performance Sky-Blue Quasi-2D Perovskite Light-Emitting Diodes

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Great progress has been made in high-performance perovskite light-emitting diodes (PeLEDs). However, the external quantum efficiency (EQE) of blue PeLEDs still lags. Particularly, defects are inevitable in solution-processed perovskite films, which inhibit the efficient radiative recombination in blue PeLEDs. Herein, a facile ionic liquid (IL, 1,3-dimethyl-3-imidazolium hexafluorophosphate) post-treatment on the as-fabricated quasi-2D perovskite film is reported, to passivate the uncoordinated Pb through the strong interaction with IL. The IL-assisted perovskite film demonstrates a great enhancement of radiative recombination efficiency, and the photoluminescence quantum yield of the film increases from 35.7% to 74.6%. IL post-treatment also adjusts the energy level of perovskite film and enhances the hole injection in PeLED. As a result, the peak EQE of the sky-blue quasi-2D PeLED is improved from 4.0% to 9.0%, while the turn-on voltage is reduced from 3.3 to 3.0 V. This work contributes to a feasible and effective approach for improving the quality of blue quasi-2D perovskites and enhancing the efficiency of blue PeLEDs.

1. Introduction

Metal halide perovskites are attracting a lot of attention due to their promising optoelectrical properties, for example, high quantum efficiency, the narrow full width of half maximum

(FWHM), tunable bandgap, and solution process compatibility, including spin-coating, thermal evaporation, and inkjet-printing. These merits enable the perovskite light-emitting diodes (PeLEDs) to be an emerging light source with high performance and low cost.^[1] Recently, there has been significant development in PeLED performances. Green, red, and near-infrared PeLEDs have been reported with a peak external quantum efficiency (EQE) of over 20%.^[2] Substantial progress has been made to improve the performance of blue PeLEDs. However, the maximum EQE of blue PeLEDs lags far behind the analogous PeLEDs.^[3] To achieve full-color displays in future applications, there is an urgent need to improve the performance of blue PeLEDs.

Recently, low-dimensional perovskites have risen interest for efficient blue emission with enhanced radiative recombination, including perovskite nanocrystals,^[4] perovskite nanoplatelets,^[5] and quasi-2D perovskites.^[3a,6] Quasi-2D perovskites have strong exciton binding energy due to the quantum and dielectric confinement, which favors the efficient generation of excitons and radiative recombination.^[7] A typical structure of quasi-2D Ruddlesden–Popper (RP) perovskites has a formula of $L_2(\text{APbBr}_3)_{n-1}\text{PbBr}_4$, where L is the organic spacer cations (e.g., phenylethylammonium: PEA^+ ; *n*-propylammonium: PA^+), A is a monovalent cation (e.g., Cs^+ , methylammonium: MA^+ or formamidinium: FA^+), and *n* represents the number of lead halide octahedral layers. The thickness of the lead halide octahedral layers determines the bandgap of the quantum-well-like domains.^[8] The quasi-2D perovskite film usually contains a mixture of domains with different thicknesses and energy bandgaps, which forms the self-organized energy funneling. The self-organized multiple-quantum-wells in quasi-2D perovskites facilitate an efficient carrier transfer and radiative recombination.^[9] However, defects, particularly ionic defects, such as halide and ammonium spacer vacancies, are unavoidable during the solution-processed crystallization of quasi-2D perovskites.^[10] Deep-level trap states will cause severe nonradiative recombination and thus low emission efficiency in blue perovskite with a large bandgap.^[10b,11] Therefore, defect passivation is essential for improving blue PeLED performance.

Recently, ionic liquids (IL) have been adopted to enhance the performance of perovskite solar cells, benefiting from

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DOI: 10.1002/adom.202202721

defect passivation, interface modification, crystallization control, and stability improvement.^[12] The wide selection of cations and anions in IL provides massive possibilities and huge potential for various targeted functions. For example, Liu et al. used methyltriethylammonium trifluoromethanesulfonate to passivate defects and to fill the surficial boundaries at the perovskite-electron transport layer interface for efficient electron extraction in perovskite solar cells.^[12e] H. Snaith et al. used 1-butyl-3-methylimidazolium tetrafluoroborate (BMIMBF₄) and 1-butyl-1-methylpiperidinium tetrafluoroborate (BMPBF₄) as the additive to retard the formation of impurity phases and thus enhance the long-term operational stability of perovskite solar cells.^[12a,b] Besides, IL shows effectiveness as the charge transport layer modifier and the precursor solution additive^[13] in PeLEDs. Sun et al. incorporated imidazolium IL into green PeLEDs, in which the IL was employed to modify the interface between the hole transport layer of PEDOT:PSS and perovskite to passivate the interfacial halogen vacancies and suppress the interfacial exciton quenching.^[13a] Peng et al. added BMIMBF₄ into green perovskite precursor solution and achieved a peak EQE of 22.9% because the big π bond of N-C = N in the cation passivates the Pb-related defects.^[13b] Luo et al. adopted 1-butyl-3-methylimidazolium trifluoromethanesulfonate (BMIMOTf) in green PeLEDs for defects passivation through bonding [BMIM]⁺ cations to uncoordinated Br, OTf⁻ anions to uncoordinated Pb²⁺ defects.^[13c] Nevertheless, these approaches add ionic liquid additives into precursor solution, which rise challenges including storage stability of precursor solutions,^[14] precise control of additives amount,^[12a,14] and different crystallization kinetics.^[12a,d,15] By contrast, post-treatment is separated from the fabrication of common perovskite films, which provides a general strategy to improve the quality of film and device.^[15a,16]

Herein, we report a facile and effective post-treatment by spin-coating IL, 1,3-dimethyl-3-imidazolium hexafluorophosphate ([DMIM]⁺PF₆⁻), onto as-fabricated sky-blue quasi-2D perovskite films. IL can effectively passivate the defects of uncoordinated Pb through strong interaction with both cation and anion. Simultaneously, IL post-treatment benefits the hole injection in PeLED through the energy level adjustment. As a result, we realized an increased photoluminescence quantum yield (PLQY) of 74.6% from 35.7%. The IL-assisted sky-blue PeLEDs exhibit an improved peak EQE of 9.0%, with a 0.3 V lower turn-on voltage of 3.0 V.

2. Results and Discussion

To avoid severe phase separation in the mix-halide perovskite,^[17] the bandgap is enlarged only by the phase modulation of the quasi-2D bromide perovskite to achieve blue emission. The pristine perovskites are constructed based on the quasi-2D perovskite of PEA_xPA_{2-x}(CsPbBr₃)_{n-1}PbBr₄, where PEA⁺ and PA⁺ act as spacer ligands. Dual spacer ligands are adopted here to suppress the formation of low-order phases with large bandgap (e.g., $n = 1$ phase) which hinder carrier injection and energy transfer.^[18] The perovskite films are fabricated through the spin-coating process in which ethyl acetate (EA) is quickly dripped onto the film for rapid crystallization of perovskite. In preparation of the ionic liquid-modified pristine perovskite films (denoted as pristine/IL perovskite), [DMIM]⁺PF₆⁻ is adopted for the post-treatment on the pristine perovskite films. The whole process scheme is shown in **Figure 1a**. After annealing of perovskite film, the solution of IL dissolved in EA is dripped onto the pristine perovskite via a dynamic way

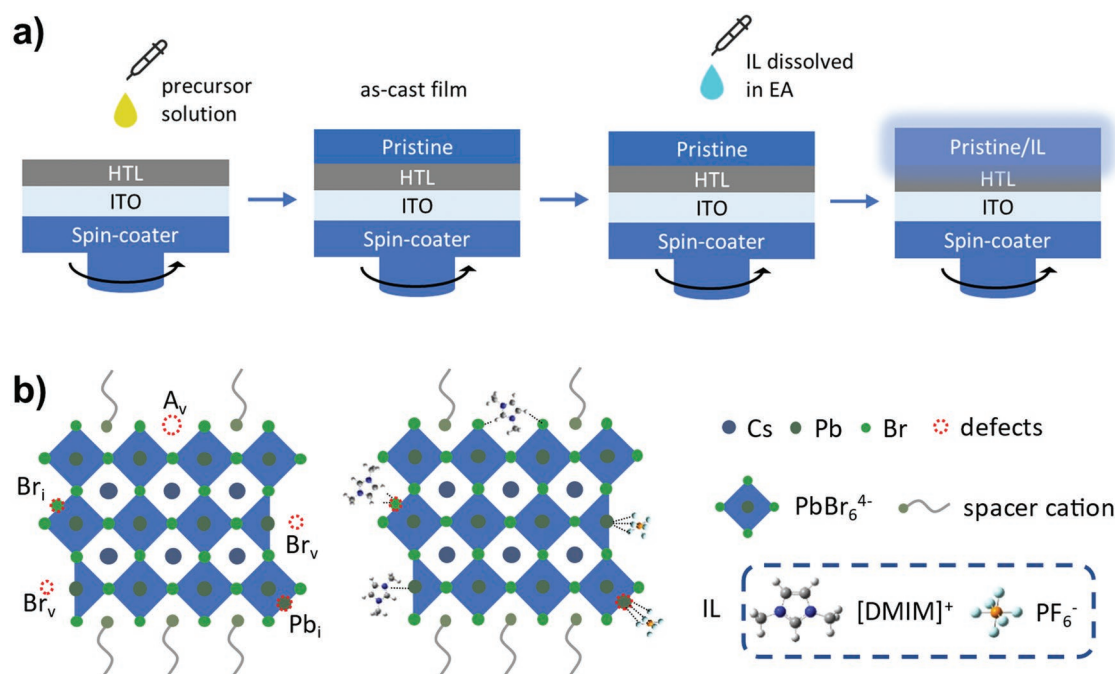


Figure 1. a) Schematic illustration of the IL treatment on the pristine perovskite film; b) schematic illustration of the passivation functions of anions and cations of IL in perovskite films. A_v: A-site atom vacancy, Br_i: interstitial bromide, Br_v: bromide vacancy, Pb_i: interstitial lead.

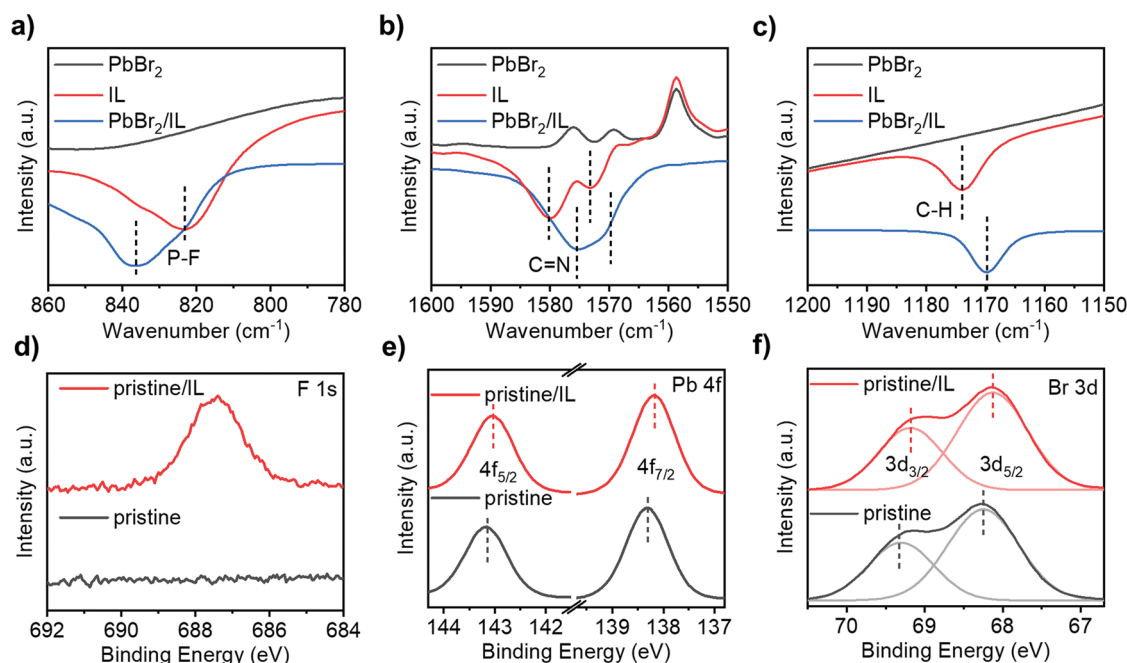


Figure 2. a–c) FTIR spectra of PbBr_2 , IL, and their mixture; XPS spectra of d) F 1s, e) Pb 4f, and f) Br 3d signals of pristine and pristine/IL perovskite films.

followed by depositing the following charge transport layer. Figure 1b illustrates the chemical structure of IL and the IL-assisted quasi-2D perovskites. It is considered that IL will not introduce new perovskite components with no additional x-ray diffraction (XRD) peaks after IL treatment (Figure S1, Supporting Information). IL cation and anion are supposed to locate at the perovskite grains and the surface of perovskite film to passivate the defects. Details will be discussed in the following part.

To explore the mechanism of IL passivation on the pristine perovskite films, PbBr_2 powder, IL powder, and their mixture were prepared and measured through Fourier transform infrared (FTIR) spectroscopy first. As shown in Figure 2a, the IL exhibits an intense absorption peak at $\approx 823 \text{ cm}^{-1}$, which corresponds to the P-F asymmetric stretch of the IL.^[19] While the P-F absorption peak shifts toward a higher wavenumber of $\approx 836 \text{ cm}^{-1}$ for IL/ PbBr_2 , which indicates the strong interaction between PF_6^- and PbBr_2 .^[13a,20] The C=N stretching vibration peak (Figure 2b) of pure IL in the imidazole ring shifted from 1580 and 1573 cm^{-1} to 1575 and 1568 cm^{-1} , respectively, after mixing with PbBr_2 powders, due to the strong interaction between imidazole and undercoordinated Pb defects through the electron delocalization of π bond on N-C=N.^[13b,21] In addition, the peak of C-H (Figure 2c) slightly shifted from 1173 to 1169 cm^{-1} , probably due to the formed hydrogen bond between C-H and Br atoms.^[13b] Moreover, X-ray photoelectron spectroscopy (XPS) of the pristine and the pristine/IL perovskite films was performed to clarify the interaction between IL and perovskite. As shown in Figure 2d, the peak at 687 eV of the F 1s signal derives from PF_6^- ions, indicating that IL remains on the perovskite film after IL treatment. Figure 2e shows that the peaks of the Pb $4f_{5/2}$ and Pb $4f_{7/2}$ signal at 143.2 and 138.3 eV for the pristine perovskite film shift toward the lower binding energies after IL treatment. The reduced binding energy of

both Pb 4f peaks indicates the lowered oxidation state of lead, that is to say, the surrounding electron cloud density of Pb^{2+} increases after IL treatment. This is ascribed from the donation from the lonely electron pair on the nitrogen to the empty 6p orbital of Pb^{2+} .^[13a,c,22] The lower wavenumber of the C=N peak in FTIR stands for a weaker bond of C=N and a lower electron cloud density around nitrogen, which donate electrons to Pb^{2+} . The red shift of the C=N peak in FTIR and the reduced binding energy of Pb demonstrate the interaction between nitrogen and undercoordinated Pb^{2+} ions. Similarly, the peaks of Br $3d_{3/2}$ and $3d_{5/2}$ signals of the pristine/IL perovskite film also shift toward lower binding energy from 69.3 and 68.3 eV of the pristine perovskite film (Figure 2f), because of the increased electron cloud density and hydrogen bond from the imidazole cation to bromine.^[13b,23] Consequently, we demonstrate the strong interaction between the IL and the perovskite through both the imidazole cation and PF_6^- , which provide a strong passivation effect to unsaturated Pb^{2+} in the perovskite film, while the hydrogen bond also offers passivation to point defects like interstitial bromide and A-site vacancy, as illustrated in Figure 1b, benefitting the improved radiative recombination efficiency.

The optical properties of perovskite films are changed with IL assistance. The UV-vis absorption spectra of the perovskite films are shown in Figure 3a, in which both of the perovskite films present two obvious excitonic absorption shoulders at around 430 and 458 nm, corresponding to $n = 2$ and $n = 3$ phases, respectively.^[18] It is also observed that there is an absorption tail extended to 490 nm which can be assigned to $n \geq 4$ phases.^[24] The increased absorbance of pristine/IL perovskite film benefits from the improved crystallinity, which is coordinated with XRD patterns (Figure S1, Supporting Information). The improvement of crystallinity may be due to the coordination between IL and exposed defect sites. From the UV-vis absorption spectra, the optical bandgap can be estimated by Tauc plot,^[25] as shown

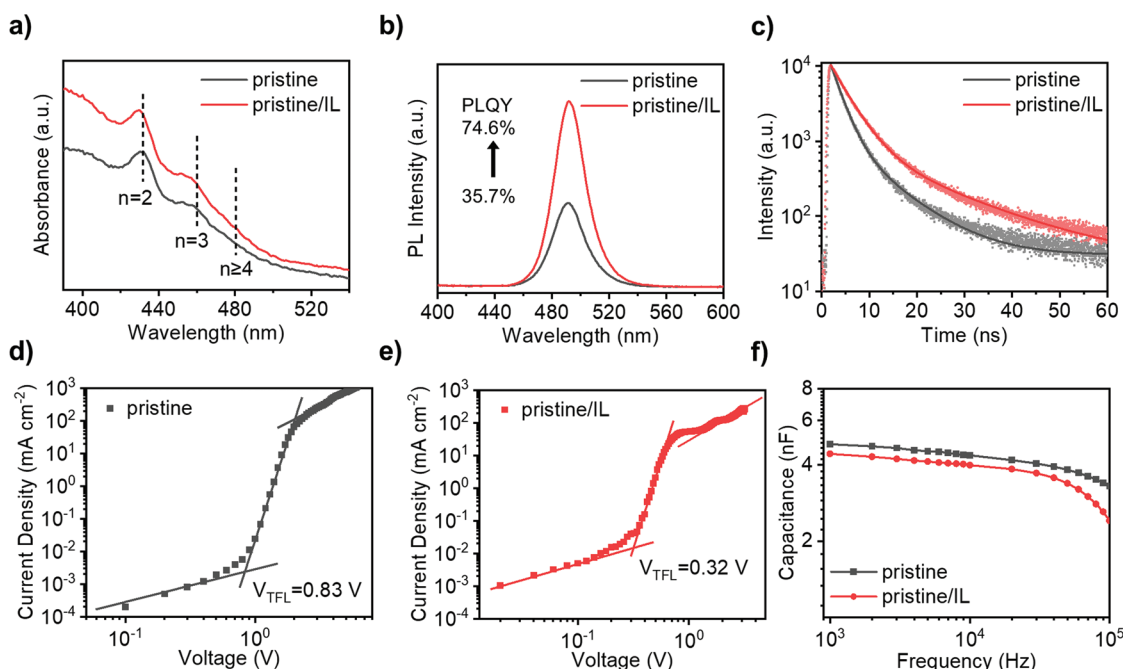


Figure 3. a) The UV–vis absorption spectra, b) steady-state PL spectra, c) TRPL decay spectra of pristine and pristine/IL perovskite films; d,e) space charge limited current (SCLC) measurements; f) capacitance–frequency (C – f) curve of pristine and pristine/IL perovskite devices.

in Figure S2, Supporting Information. The calculated bandgap of the perovskite film has no obvious change after IL treatment, which means that the IL will not change the phase distribution in the film. Figure 3b shows the PL spectra of the pristine and the pristine/IL perovskite films. The perovskite films show the same PL peak at 490 nm with an FWHM of 25 nm due to the unchanged phase distribution of the perovskite films after IL treatment, as verified by the same absorption peaks (Figure 3a). While the pristine/IL perovskite film displays stronger PL intensity than that of the pristine perovskite film, indicating more radiative recombination of the carriers in the pristine/IL perovskite films. Accordingly, the pristine/IL perovskite film shows an enhanced PLQY of 74.6% which is over two-fold larger than 35.7% for the pristine perovskite film, indicating the reduced non-radiative recombination after IL treatment. Pure EA post-treatment on pristine film is performed through UV–vis absorption and PL spectra to exclude the effect of solvent, and the results show that pure EA post-treatment hardly influences the absorbance and PLQY of perovskite films (Figure S4a,b, Supporting Information). Furthermore, the time-resolved photoluminescence (TRPL) measurement was performed to study the effect of IL treatment on the perovskite film carrier lifetime. As shown in Figure 3c, the TRPL decay curves are fitted by a biexponential function of $I(t) = I_0 + a_1 \exp(-t/\tau_1) + a_2 \exp(-t/\tau_2)$, where a_1 , a_2 are the amplitudes, τ_1 , τ_2 , are the time constants of fast and slow decay lifetimes. The fast time constant of τ_1 reflects the non-radiative recombination due to the trap states, while the slow time constant, τ_2 , reflects the radiative recombination.^[26] The fitted parameters are extracted in Table S1, Supporting Information. The pristine/IL perovskite film has both longer fast decay and slow decay lifetimes of 3.60 and 15.79 ns than those of the pristine perovskite film of 2.16 and 8.23 ns, respectively, which promotes the average fluorescence

lifetime (τ_{av}) of 7.33 ns (pristine/IL perovskite), near a two-fold longer than 4.13 ns of the pristine perovskite film. These results clearly show that IL treatment effectively suppresses the non-radiative recombination and enhances the radiative recombination,^[27] benefitting from the diminished trap states of the perovskite films. To evaluate the effect of IL treatment on exciton dynamics and carrier energy transfer, transient absorption (TA) spectra are performed, as shown in Figure S7, Supporting Information. However, there is no obvious difference in TA spectra in different delay times and kinetic fitting, which is predictable due to the similar UV–vis absorption spectra. It can be considered that the IL post-treatment will not affect the phase distribution and thus the exciton dynamics and energy transfer inside the perovskite film. To further verify the effect of ionic liquid on reducing the defect density of perovskite films, the space charge limited current (SCLC) is conducted. The current–voltage (J – V) curves of the devices were measured with a structure of ITO/perovskite/MoO₃/Ag, as shown in Figure 3d,e. The trap state density of the perovskite film can be calculated by the equation of $N = (2V_{TFL}\epsilon_r\epsilon_0)/(eL^2)$, where V_{TFL} is the trap-filled limiting voltage, L is the thickness of the perovskite film, e is the elementary charge, ϵ_r and ϵ_0 are the relative permittivity and the vacuum permittivity, respectively. In general, a lower V_{TFL} means a lower defect density in the films. The values of defect density can be further calculated. The value of ϵ_r is estimated by the equation of $\epsilon_r = (CL)/(\epsilon_0 S)$, where the geometrical capacitance of C can be determined from the capacitance–frequency (C – f)–curves at a high frequency of around 10^3 – 10^5 Hz (Figure 3f).^[28] According to the calculation according to the capacitance at 1k Hz, the trap state density of the pristine/IL perovskite film is reduced to 8.83×10^{16} from $2.54 \times 10^{17} \text{ cm}^{-3}$, (the pristine perovskite film), which further confirms the effective passivation of IL.

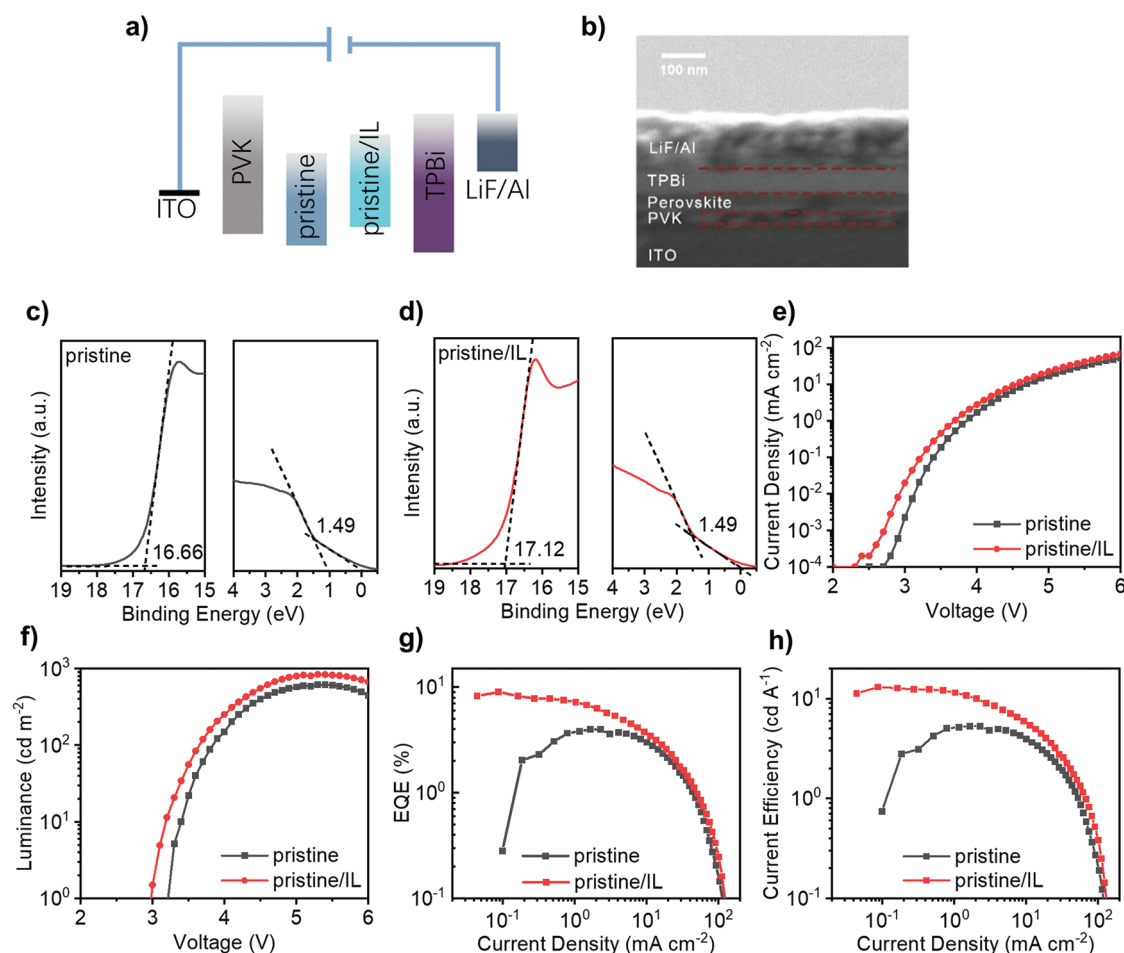


Figure 4. a) Schematic energy levels alignment of the PeLED structure, b) the cross-section scanning electron microscopy (SEM) images of PeLEDs; UV photoelectron emission spectra (UPS) spectra of c) the pristine perovskite film and d) the pristine/IL perovskite film; e) current density–voltage (J – V), f) luminance–voltage (L – V) curves, g) external quantum efficiency–current density (EQE– J), and h) current efficiency–current density (CE– J).

To evaluate the effect of IL treatment on device electroluminescent performance, the sky-blue PeLEDs based on the pristine perovskite and the pristine/IL perovskite were assembled. The PeLEDs were fabricated with a structure of ITO/poly(9-vinylcarbazole) (PVK)/perovskite/1,3,5-tris(1-phenyl-1H-benzimidazol-2-yl)benzene (TPBi)/LiF/Al, where the pristine perovskite or the pristine/IL perovskite act as emission layer, PVK and TPBi act as the hole transport layer and electron transport layer, respectively. The energy levels alignment of the functional layer in PeLEDs is summed up in **Figure 4a**. The energy levels of the functional layers are obtained from the previous reports,^[3b] while the energy levels and band gaps of the pristine perovskite and pristine/IL perovskite films are calculated from the Tauc plot and UV photoelectron emission spectra (UPS) (Figure 4c,d and Figure S2, Supporting Information), respectively. The results show that the pristine/IL perovskite film possesses a shallower valence band than that of the pristine perovskite films, which reduces the hole injection barrier and facilitates the hole injection. The work function of pristine/IL perovskite decreases to 4.00 from 4.56 eV. Considering the relative position of the work function (WF) in the energy band, the reduced WF is not mainly caused by the reduced n-type or p-type trap

density.^[29] A surface dipole may exist at the top surface of perovskite film, which reduces the work function.^[25,46] The smaller PF_6^- anions may penetrate and coordinate with the Pb^{2+} defect or partially replace bromide, while the larger imidazole cations accumulate at the surface, which forms surface dipoles spontaneously (Figure S5, Supporting Information). The surface work function measurements of pristine and pristine/IL film are also performed through a Kelvin probe after calibration by highly oriented pyrolytic graphite (HOPG) and confirming the reduced work function after IL treatment (Figure S6). From a typical scanning electron microscopy (SEM) image of the cross-section of the pristine/IL PeLED in Figure 4b, the thicknesses of PVK, perovskite film, TPBi, LiF/Al are around 25, 50, 45, and 100 nm, correspondingly. The J – V and luminance–voltage (L – V) curves of the PeLEDs are shown in Figure 4e,f, respectively. The current density of the pristine/IL PeLED is larger than the pristine PeLED, which benefits from the lower hole injection barrier from PVK to perovskite film, according to the decreased work function and valence band. Thus, the turn-on voltage (defined as the voltage where $L = 1 \text{ cd m}^{-2}$) of the pristine/IL PeLED decreases from 3.3 to 3.0 V due to improved hole injection. The maximum luminance (L_{max}) of the pristine perovskite

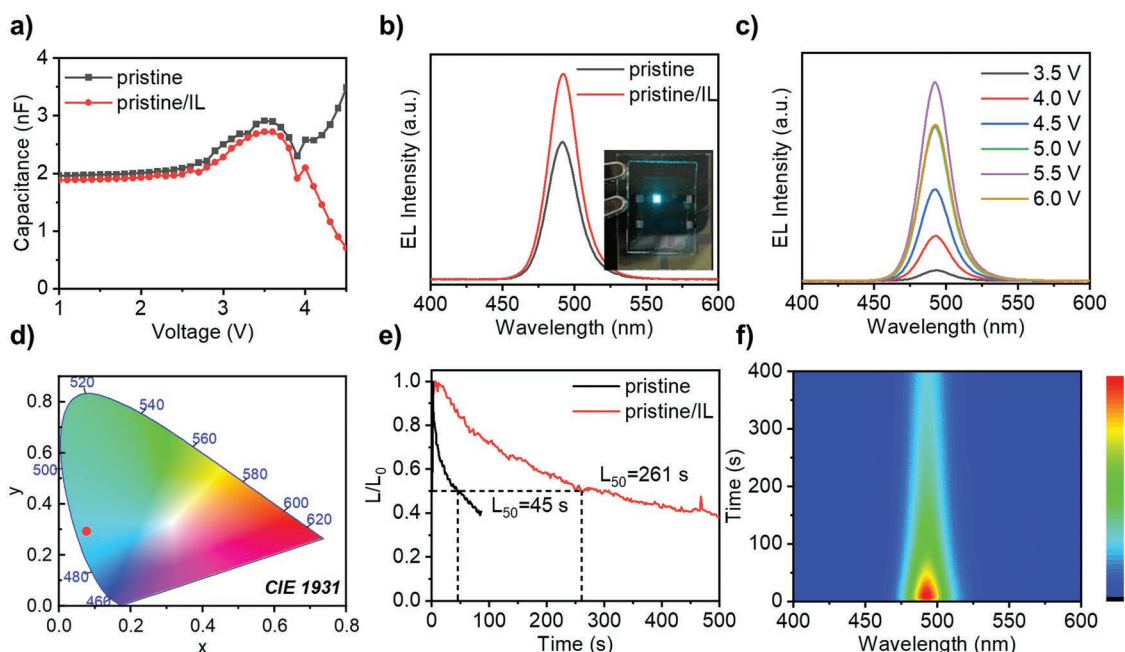


Figure 5. a) The capacitance–voltage (C – V) curves of pristine and pristine/IL PeLED; b) electroluminescence (EL) spectra at 5.0 V (insert: pictures of lighting pristine/IL PeLED); c) EL spectra of pristine/IL PeLED at different bias voltage; d) the CIE coordinate of the pristine/IL PeLED; e) time-dependent stability lifetime measurements under a current density of 0.75 mA cm^{-2} of the pristine PeLED and the pristine/IL PeLED; f) the traced EL spectra of pristine/IL PeLED during operation.

PeLED is 615 cd m^{-2} , which is improved to 840 cd m^{-2} of the pristine/IL perovskite PeLED. The EL peaks of the pristine and pristine/IL PeLED are posited at the same wavelength of 492 nm (Figure 5b), which matches up with previous PL results. The picture of the operating pristine/IL PeLED is shown in the insert picture in Figure 5b. As shown in Figure 4 g,h, the pristine PeLED reached the highest EQE of 4.0% and the highest CE of 5.3 cd A^{-1} , while the pristine/IL PeLED reached the highest EQE of 9.0% and the highest CE of 13.1 cd A^{-1} . The EQE exhibits a significant two-fold increase, and the CE exhibits a 2.47-fold increase. The improvement of PeLED performance after IL treatment can be attributed to the effective defect passivation and improved carrier injection. To further study the carriers' behavior in the PeLEDs, the capacitance–voltage (C – V) measurements of the pristine and pristine/IL PeLEDs were performed (Figure 5a). It is observed that the capacitances of the pristine and pristine/IL PeLEDs show a similar trend during the bias voltage scanning. The capacitance increases from 2.6 to 3.5 V, which indicates a strong injection of majority carriers, corresponding to the increased current density in the J – V curve. When the bias voltage increases, the capacitance of the pristine/IL PeLED reaches the highest value and then decreases sharply with a larger slope as a result of the largely reduced charges caused by massive electron and hole recombination.^[30] The capacitance of pristine PeLED increases again at a higher voltage of 4.0 V, which indicates a charge accumulation due to the poor hole injection. Therefore, it is verified that the IL benefit both the hole injection and radiative recombination to achieve a high-efficiency blue emission in PeLEDs.

The excellent spectra stability for sky-blue PeLEDs is essential for practical application. The EL peaks and FWHM of

pristine/IL PeLED maintain the same, ensuring good color consistency at different voltages (Figure 5c). The pristine/IL PeLED emits with an FWHM of around 24 nm, allowing for a high color purity with Commission Internationale de L'Eclairage (CIE) chromaticity coordinate at (0.077, 0.293) (Figure 5d). The EL intensity of the PeLED increases from 3.5 to 5.5 V during the voltage scanning and then decreases at the higher voltage, which may blame on Auger recombination in quasi-2D perovskite.^[14,31] As shown in Figure 5e, the operation lifetime of the pristine PeLED and pristine/IL PeLED was also measured at a constant current density of 0.75 mA cm^{-2} with an initial luminance of 49 and 66 cd m^{-2} separately. The pristine/IL PeLED shows a half-lifetime (L_{50} , defined as the time it takes for a decrease to 50% of the initial value) of 261 s, which is nearly five times longer than that of the pristine PeLED (45 s). The tracing EL spectra during the operation display that the pristine/IL PeLED also keeps excellent spectral stability during operation (Figure 5f). Overall, our results show that the pristine/IL PeLEDs show excellent spectral and operational stability, indicating a great potential for blue PeLEDs in practical applications.

3. Conclusion

We introduced an IL, [DMIM]PF₆, to assist high-efficiency sky-blue quasi-2D perovskite PeLEDs, through a facile post-treatment method of spin-coating onto as-fabricated perovskite film. Through the strong interaction of cation and anion in IL with undercoordinated Pb defects from the electron donation of imidazole ring and F atoms, the defects are well

passivated, which concentration decreased from 2.54×10^{17} to $8.83 \times 10^{16} \text{ cm}^{-3}$. Besides, the IL post-treatment also reduces the hole injection barrier between the charge transport layer and perovskite film for efficient carrier injection by energy level adjustment. As a result, a peak EQE of 9.0%, and a low turn-on voltage of 3.0 V are achieved in sky-blue quasi-2D PeLEDs, with good spectra stability. This work proposes a feasible strategy of IL post-treatment on as-fabricated perovskite film for effect defect passivation for high-efficiency blue quasi-2D PeLEDs.

4. Experimental Section

Materials: N-Propylammonium bromide (PABr, 99.999%), and phenethylammonium bromide (PEABr, 99.999%) were purchased from Greatcell Solar; Cesium bromide (CsBr, 99.999%), lead bromide (PbBr_2 , 99.999%), anhydrous DMSO, and hydrobromic acid (HBr, 48% wt. in H_2O) were bought from Sigma-Aldrich. Poly(9-vinylcarbazole), tris(1-phenyl-1H-benzimidazol-2-yl)benzene (TPBi, 99.5%), and lithium fluoride (LiF, 99.99%) were purchased from Lumtec. $[\text{DMIM}]\text{PF}_6$ and ethyl acetate (Superdry, 99.9%) were purchased from J&K Chemical Ltd.

Perovskite Emission Layer Preparation: CsPbBr_3 powder was first synthesized according to the previous method.^[10c] The precursor of $\text{PEA}_x\text{PA}_{2-x}(\text{CsPbBr}_3)_{n-1}\text{PbBr}_4$ perovskite was prepared by dissolving 0.15 M CsPbBr_3 powder, 0.15 M PABr, and 0.015 M $\text{PEA}_2\text{PbBr}_4$ into DMSO. Then the pristine $\text{PEA}_x\text{PA}_{2-x}(\text{CsPbBr}_3)_{n-1}\text{PbBr}_4$ perovskite films were obtained by spin-coating the precursor at 3000 rpm for 60 s in which 300 μL of EA was quickly dripped on the substrate at 26 s after the spin-procedure starting, followed by annealing at 70 °C for 10 min. For the IL-treated perovskites, a 30 μL of IL-dissolved EA solution (0.30 mg mL^{-1}) was dynamically spin-coated on the films at 5000 rpm for 45 s.

Device Fabrication: The indium tin oxide (ITO)-coated glass substrates were sequentially cleaned by sonication in detergent, deionized water, acetone, and isopropyl alcohol and then dried with N_2 . After an oxygen plasma treatment of the ITO for 5 min, the PVK solution (4 mg mL^{-1} in chlorobenzene) was spin-coated onto the ITO substrate at 4000 rpm for 45 s and baked at 120 °C for 10 min. The 0.15 M precursor perovskite solution was spin-coated onto the PVK substrates as described. The TPBi layer (45 nm) and LiF/Al electrodes (1 nm/100 nm) were deposited using a thermal evaporation system through a shadow mask under a high vacuum of 5×10^{-4} Pa. The device's active area was 4 mm^2 as defined by the overlapping area of the ITO and Al electrode. A Lambertian emission profile was assumed in calculating the brightness. The devices were encapsulated with a UV-curable resin (LOCTITE 3335) before testing in the atmosphere.

Characterization: UV-vis absorption spectra were carried out on the spectrophotometer of Perkin Elmer 950. The PLQYs were obtained using the equipment of Hamamatsu Quantaurus-QY, model no. C11347. The PL and TRPL spectra of the perovskites were performed on PicoQuant FluorTime 300 equipped with a picosecond pulse laser (360 nm, LDH-P-C-360). X-ray diffraction analysis was carried out with an X-ray diffractometer (Bruker Advance D8 Ew Germany) with $\text{Cu K}\alpha$ radiation. The C-V measurements were recorded using a Keithley 4200A parameter analyzer at 1 kHz with an AC amplitude of 50 mV. The capacitance-frequency (C-f) measurements were performed using a Keithley 4200A parameter analyzer with a frequency range from 1k to 100k Hz and an AC amplitude of 50 mV. The value of absolute dielectric permittivity is $8.85 \times 10^{-12} \text{ F m}^{-1}$, the device area is 0.04 cm^2 , and the thickness of the perovskite film is 50 nm. The geometrical capacitances for pristine, pristine/IL are 4.83, and 4.42 nF, corresponding to the calculated ϵ of 6.81, and 6.23, respectively. The J-V-L curves were performed on an Ocean Optics system equipped with a Keithley 2400 source meter, an integrating sphere (FOIS-1), and a QE65 Pro spectrometer. The integrating sphere was calibrated with a radiometric calibration light source (HL-3plus) before use. The SEM images were obtained using ZEISS Gemini 300. He (I) UV radiation source (21.22 eV) from a He discharge lamp was used

in the UPS measurements. The work functions and the highest occupied molecular orbital (HOMO) regions were extracted from the UPS spectra using the equations $\text{WF} = 21.22 \text{ eV} - E_{\text{cutoff}}$ and $\text{HOMO} = \text{WF} + E_{\text{onset}}$. The E_{cutoff} and the E_{onset} were determined by the intercepts of the tangents of the peaks with the extrapolated baselines, as depicted in the relevant plots. The surface work function through a Kelvin probe was done by SKP5050 scanning Kelvin Probe System with a resolution of 1–3 meV from KP Technology Ltd. The Kelvin Probe measurement used a new layer of HOPG as a reference (4.60 eV) to calculate the absolute work function values of different perovskite films. For the transient absorption measurements system, the perovskite films were pumped with a femtosecond 365 nm laser pulse generated from an optical parametric amplifier (Lyra, Light Conversion). The probe pulses ranging from 380 to 800 nm were derived from the fundamental 800 nm laser pulses by a Ti:Sapphire regenerative amplifier (Orpheus). The pump and probe pulses were detected by a commercial TA spectrometer (Time-tech Spectra, Dalian).

Supporting Information

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

Acknowledgements

This work was supported by National Key Research and Development Program of China (No. 2022YFB3606504), National Natural Science Foundation of China (No. 62122034), Guangdong Basic and Applied Basic Research Foundation (No. 2022A1515011071), and Shenzhen Basic Research General Program (No. JCYJ20210324104413036, No. JCYJ20190809152411655), the General Research Fund (grant Nos. 17201819, 17211220, and 17200021) and Collaborative Research (grant No. C5035-20G) from the Research Grants Council of Hong Kong Special Administrative Region, China, as well as Shenzhen Stable Support Research Foundation (No. 20220717215521001) and Natural Science Foundation of Top Talent of Shenzhen Technology University (SZTU) (No. GDRC202110).

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

defect passivation, ionic liquids, sky-blue perovskite light-emitting diodes

Received: November 15, 2022
Revised: February 10, 2023
Published online: March 20, 2023

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