Efficient green light-emitting diodes based on quasi-two-dimensional composition and phase engineered perovskite with surface passivation

Xiaolei Yang1,2, Xingwang Zhang1,3, Jinxiang Deng2, Zema Chu1, Qi Jiang1,3, Junhua Meng1,3, Pengyang Wang1,3, Liuqi Zhang1,3, Zhigang Yin1,3 & Jingbi You1,3

Perovskite light-emitting diodes (LEDs) are attracting great attention due to their efficient and narrow emission. Quasi-two-dimensional perovskites with Ruddlesden-Popper-type layered structures can enlarge exciton binding energy and confine charge carriers and are considered good candidate materials for efficient LEDs. However, these materials usually contain a mixture of phases and the phase impurity could cause low emission efficiency. In addition, converting three-dimensional into quasi-two-dimensional perovskite introduces more defects on the surface or at the grain boundaries due to the reduction of crystal sizes. Both factors limit the emission efficiency of LEDs. Here, firstly, through composition and phase engineering, optimal quasi-two-dimensional perovskites are selected. Secondly, surface passivation is carried out by coating organic small molecule triocetylphosphine oxide on the perovskite thin film surface. Accordingly, green LEDs based on quasi-two-dimensional perovskite reach a current efficiency of 62.4 cd A\(^{-1}\) and external quantum efficiency of 14.36%.
solution-processed halide perovskite semiconductor materials show good photoluminescence quantum yields (PLQYs) \(^\sim 1\)\(^\text{2–4}\), which motivates applications in displays and lighting\(^\text{5}^\text{6}\). The perovskite light-emitting diodes (PeLEDs) have been reported with moderate electroluminescence (EL) efficiencies as well\(^\text{7–15}\). Their external quantum efficiencies (EQEs) have reached about \(10\%\) for near-infrared \(^\text{13–15}\) and also in green emission\(^\text{11,12,13}\). Hybrid organic–inorganic perovskites are better known for their free charge carriers with long diffusion length, which is good for solar cells. However, opposite properties are desired when they are used as emitting materials in LEDs\(^\text{11,16}\). It is known that quasi-2D perovskite materials usually contain a mixture of phases and the phase impurity could lead to low emission efficiency, and thus fine control of the composition/ phases is critical for efficient emission. In addition, solution processed thin films of quasi-2D perovskite show reduced crystal size compared to those of 3D perovskite\(^\text{13,15}\). This could increase the concentration of the defects and traps on the film surface and grain boundaries, which will act as non-radiative recombination centers and thus decrease the emission efficiency. Surface passivation has been proved an efficient way for reducing the defects in perovskite solar cells\(^\text{22–24}\).

In this article, \(\text{PEA}_n(\text{FAPbBr}_3)_m\text{PbBr}_4\) \((n = 1, 2, ..., \infty)\) is investigated as the emitting layer for quasi-2D perovskite LEDs, where PEA and FA are phenylethylammonium (C\(_6\)H\(_5\)C\(_2\)H\(_4\)NH\(_3^+\)) and formamidinium (HC(NH\(_2\))\(_2\)\(^+\)), respectively. Firstly, we optimize the films phase engineering/phase engineering. We find that the perovskite with \(n = 3\) composition shows decent phases and the obtained films show the highest photoluminescence (PL)

**Fig. 1** Properties of perovskite films with different compositions. **a** Scheme of \(\text{PEA}_n(\text{FAPbBr}_3)_m\text{PbBr}_4\) phases. The black part is PEA, the blue square is PbBr\(_4\)-octahedron and the gray dot is FA. **b** X-ray diffraction patterns of \(\text{PEA}_n(\text{FAPbBr}_3)_m\text{PbBr}_4\) films with different \(n\)-compositions. Diffraction patterns of \(n = 1\) phase (black vertical lines), \(n = 2\) phase (pink vertical lines) and \(n = \infty\) phase (marked with (100)) were labeled. The analysis details can be found in the Supplementary Table 2. **c** Photoluminescence of \(\text{PEA}_n(\text{FAPbBr}_3)_m\text{PbBr}_4\) with different \(n\)-compositions. **d** Photoluminescence intensity and peak of \(\text{PEA}_n(\text{FAPbBr}_3)_m\text{PbBr}_4\) films with different compositions with the data collected from **c**. **e** Photoluminescence image of \(\text{PEA}_n(\text{FAPbBr}_3)_m\text{PbBr}_4\) films with different compositions under ultraviolet lamp excitation. Figure 1a adapted from ref. 13 (copyright 2016 Macmillan Publishers)
yield. Secondly, we present surface passivation of quasi-2D perovskite films by trioctylphosphine oxide (TOPO) treatment. Accordingly, the PeLEDs based on \( \text{Pb}_{3} \text{Cl}_{6} \) \( \text{PbBr}_{3} \) with \( n = 3 \) composition show green emission with current efficiency (CE) of 62.4 cd A\(^{-1}\) and EQE of 14.36%, which is the highest efficiency for PeLEDs reported so far.

**Results**

**Properties of quasi-2D perovskite with different compositions.** The schematics of \( n \)-phase \( \text{Pb}_{3} \text{Cl}_{6} \) \( \text{PbBr}_{3} \) are drawn in Fig.1a, illustrating that the \( n \)-phase unit cell contains \( n \) sheets of \( \text{PbBr}_{3} \)-octahedra and two PEA tiers. The perovskite films are prepared by spin-coating a precursor solution using a crystallizing process. Their compositions are controlled by the ratio \( \text{PEA}_{3} \text{Br}_{2} \text{Pb}_{3} \) (Supplementary Table 1), and compositions \( n \) ranging from 2 to 6 are discussed. Previous results suggested that the quasi-2D perovskite films are not only a single phase but contain a variety of \( n \)-phases. For example, if the precursor solution with \( n = 2 \) composition is spin-coated, the generated films might include \( n = 2 \) phase but could also contain other phases such as \( n = 1 \) or \( n > 2 \). To avoid misunderstanding, we distinguish the composition and phase by \( n \)-composition and \( n \)-phase, respectively. Methylammonium chloride (MACl) is also added into the precursor to improve the morphology and enhance the emission efficiency of the formed films (Supplementary Figs. 1, 2 and 12), but most MACl escape from the film during annealing. The role of MACl has been well investigated in solar cells. First, the MACl can slow down perovskite crystallization and improve film morphology. Second, the incorporation of a very small amount of elemental Cl could passivate the grain boundary, and then enhance the emission efficiency (Supplementary Fig. 2).

X-ray diffraction (XRD) measurements are carried out to determine the accurate phases in our obtained quasi-2D perovskite films. As shown in Fig. 1b, all the perovskite films with \( n \geq 2 \) compositions show diffraction peaks at 14.8°, which is the same as the (100) diffraction patterns of the 3D perovskite \( \text{FAPbBr}_{3} \) (refs. 7, 35). However, the peaks become much more broadened, indicating that the diffraction peak could be from a series of \( \text{PEA}_{2} \text{FAPbBr}_{3} \) phases with large \( n \) (\( n \rightarrow \infty \)) or 3D \( \text{FAPbBr}_{3} \) with nano-size grains (Supplementary Fig. 3). This result shows that all the quasi-2D perovskite films always contain large \( n \)-phases (\( n \rightarrow \infty \)) or 3D perovskite regardless of the composition of the precursor. Atomic force microscopy (AFM) images confirm the reduction of the crystal size during the transformation of 3D perovskite into quasi-2D perovskites (Supplementary Fig. 3).

In addition to the diffraction peak at 14.8°, a series of Bragg reflections at low angles (2θ < 14.8°) are observed for these quasi-2D perovskites (Fig. 1b). This indicates that the PEA group with large size is incorporated, and the size of unit cells is enlarged compared with 3D perovskite. According to the analysis shown in Supplementary Table 2 and the unit cells of \( n \)-phases shown in Fig. 1a, it is found that the diffraction peaks (2θ < 14.8°) are attributed to the \( n \)-phases (diffraction patterns from \( n \geq 3 \) phases are not observed here). Specifically, for \( n = 1 \) composition, only the \( n = 1 \) phase exists; for \( n = 2 \) composition, the \( n = 1 \) and \( n = 2 \) phases are both presented in the films. It is unexpected that diffraction peaks of the \( n = 1 \) phase is absent in the \( n = 3 \) composition, while obvious diffraction peaks from the \( n = 2 \) phase are observed. For \( n \geq 4 \) compositions (\( n > 4 \) compositions are not shown here due to very weak small \( n \)-phases diffractions), the diffraction peaks from small \( n \)-phases become weak or absent.

The photoluminescence (PL) results of the quasi-2D perovskite films with different compositions are shown in Fig. 1c, and the 2D (\( n = 1 \)) and 3D (\( n \rightarrow \infty \)) perovskite films are also included for comparison. The 2D perovskite shows an emission at 412 nm (3.01 eV) and the long tail at a longer wavelength, which could be due to the self-trapped exciton emission or the disordered structure. The 3D perovskite showed a band edge emission peak at 542 nm (2.29 eV). All quasi-2D perovskite films have two main emission peaks: one in the green region (approximately 532 nm, approximately 2.33 eV) and another in the blue region (at 440 nm, 2.82 eV). Comparing the PL with the absorption spectra (Fig. 1c and Supplementary Fig. 4), it can be found that the stronger green emission at approximately 532 nm comes from large \( n \)-phases (\( n \rightarrow \infty \)) or 3D perovskite, and the weaker blue emission at 440 nm comes from the \( n = 2 \) phase. The emission phases we find are nearly almost consistent with the XRD results. There are some exceptions: for the \( n = 2 \) composition, we have not observed the \( n = 1 \) phase emission, while the \( n = 4 \) composition shows an emission at 405 nm from the 2D perovskite phase (\( n = 1 \)), and the reason for this is not clear at present.

The relative PL intensity and emission peak wavelength in the green emission from the quasi-2D perovskites and 3D perovskite are plotted in Fig. 1d. It is found that the \( n = 3 \) composition film shows the brightest green emission. The significant blue-shift of the green emission from quasi-2D perovskites is observed compared with the 3D perovskite. In addition, a smaller \( n \)-composition showed a larger blue-shift, indicating that the blue-shift is due to the quantum confinement effect. The emission images of these quasi-2D perovskite films under ultraviolet excitation (Fig. 1e) also confirms that the \( n = 3 \) composition shows the best PL.

Compared with 3D perovskites, significant enhancement of the PL is observed for quasi-2D perovskites (Fig. 1c). This is due to the quantum confinement effect via the formation of quantum well structures between large bandgap phases (small \( n \)-phases) and small bandgap phases (\( n \rightarrow \infty \) phase or 3D perovskite). The different emissions from various compositions observed in Fig. 1c could be explained by the competition between confinement phase (small \( n \)-phase) and emission phase (large \( n \)-phase). Here, we use \( n = 2, 3, 4 \) compositions as examples. According to XRD results shown in Fig. 1b, the \( n = 2 \) confinement phase and 3D emission phase dominate in \( n = 2 \) and \( n = 4 \) composition, respectively, and both of these will cause weak green emission. While the amount of \( n = 2 \) phase and 3D phase are adequate in the \( n = 3 \) composition, this could be the reason of the highest PL of \( n = 3 \) composition. The PLQY of the \( n = 3 \) shows a value as high as 57.3% under low density excitation (excitation wavelength of 400 nm, power density of 3.5 mW cm\(^{-2}\)). The quantum well of \( n = 3 \) composition is mainly formed by the \( n = 2 \) and \( n \rightarrow \infty \) phases (Supplementary Fig. 6).

**Passivation of quasi-2D perovskite.** Keeping in mind that the \( n = 3 \) composition of \( \text{Pb}_{3} \text{Cl}_{6} \) \( \text{PbBr}_{3} \) perovskite could be a good candidate for an emitting material, we further improve its emission by surface passivation. An organic small molecule TOPO is spin-coated onto the perovskite surface to form a thin passivation layer. It is found that the PLQY of the \( n = 3 \) composition film was significantly increased from 57.3% to 73.8% after coating with the TOPO layer (Fig. 2a), confirming the passivation effect of the TOPO layer. The average fluorescence lifetime of the \( n = 3 \) composition film is also increased from 0.17 μs to 0.36 μs after TOPO treatment (Fig. 2b). Although a small red-shift in PL spectra has been observed for the TOPO-treated 3D perovskite film previously, no obvious modulation of PL due to TOPO treatment is observed in this study (Supplementary Fig. 7).
The passivation of halide perovskites is ascribed to the chemical interaction between molecular ligands and incomplete PbI$_6$-octahedra in previous study$^{39}$. To confirm this, Fourier transform infrared (FTIR) spectroscopy measurements were carried out, and the result is shown in Fig. 2c. An absorption peak located at 1150 cm$^{-1}$ is observed for TOPO, which corresponds to P=O bond stretching vibrations$^{40}$. This P=O bond absorption peak shifts to approximately 1100 cm$^{-1}$ in the film comprised of TOPO and PbBr$_2$, indicating that the bonding between perovskite and TOPO could be formed$^{41,42}$.

**Device structure and performance.** We configure PeLEDs as the structure glass/ITO/PEDOT:PSS/PEA$_2$(FAPbBr$_3$)$_n$PbBr$_4$ (n = 3 composition) perovskite films with and without TOPO passivation. Time-resolved photoluminescence (TRPL) of the PEA$_2$(FAPbBr$_3$)$_n$PbBr$_4$ (n = 3 composition) film with and without TOPO passivation layer. Fourier transform infrared (FTIR) spectroscopy measurement for TOPO, PbBr$_2$ and TOPO-PbBr$_2$ films prepared on silicon wafers.

Fig. 2 Passivation effect of TOPO on the perovskite with n = 3 composition. a Photoluminescence quantum yield (PLQY) of the PEA$_2$(FAPbBr$_3$)$_n$PbBr$_4$ (n = 3 composition) perovskite films with and without TOPO passivation. b Time-resolved photoluminescence (TRPL) of the PEA$_2$(FAPbBr$_3$)$_n$PbBr$_4$ (n = 3 composition) film with and without TOPO passivation layer. c Fourier transform infrared (FTIR) spectroscopy measurement for TOPO, PbBr$_2$ and TOPO-PbBr$_2$ films prepared on silicon wafers.

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phase. Correspondingly, the brightness and CE of $n=2$ composition-based devices are both the lowest (Fig. 4a, b). The compositions of $n=3$ and $n=4$ based PeLEDs show the maximum brightness of about 7000 cd m$^{-2}$ and 8700 cd m$^{-2}$, respectively, while $n=3$ composition shows a higher CE (40.2 cd A$^{-1}$) than $n=4$ composition (37.6 cd A$^{-1}$). For the $n=5$ and $n=6$ compositions, consistent with their PL efficiency (Fig. 1c, d), their EL also could not compare with that of $n=3$ composition. The detailed device performances for various compositions are summarized in Table 1.

We next discuss the role of surface passivation in performance improvement for $n=3$ composition. It is found that the turn-on voltage increases and the injection current decreases after introducing a TOPO layer (Supplementary Fig. 10). This is due to the insulating property of TOPO and indicates that the TOPO is successfully coated on the perovskite surface. Figure 4c shows the brightness curve of the PeLEDs with and without TOPO. The maximum brightnesses of the PeLEDs with and without passivation are at the same level, while the devices with passivation showed higher CE (Fig. 4d). The best performing device without TOPO passivation shows a CE of 52.5 cd A$^{-1}$, while TOPO-treated devices show a CE as high as 62.4 cd A$^{-1}$, and the power efficiency also increased from 49.4 lm W$^{-1}$ to 53.3 lm W$^{-1}$ (Supplementary Fig. 11). Accordingly, a high EQE of 14.36% is obtained (Fig. 4e). We also find that our devices show good reproducibility. The EQE histogram for 60 devices from five batches are presented, showing an average EQE of 13.1% with a low relative standard deviation of 4.4% (Fig. 4f).

**Discussion**

Device stability is still a critical and common issue in PeLEDs. The stability of our PeLEDs with encapsulation was tested under a constant injection current in dry air (Supplementary Fig. 14). The luminance and CE increased at the beginning for several minutes, which could be due to trap filling$^{15}$ that results in the improvement of electrical contacts. After that, the device performance gradually decreased, and finally the devices work for approximately 120 min. We also noticed that larger injection current induces faster degradation, which could be due to heat accumulation or ion movements. Our device showed the almost same level stability of I-related perovskite with the stability in several 100 min$^{15}$. Compared with the Br-related perovskite with several 10 min stability in previous reports$^{15,44}$, our devices showed a little bit improvement.

Further improvement of PeLED stability is necessary, and we assume that there are several issues leading to PeLED instability. One major issue could be ion migration under voltage bias, and another issue could be the phase stability of perovskite materials under heating or moist conditions. Several strategies could be adopted to improve PeLED stability such as finding stable perovskite-emitting materials$^{19,45}$, incorporating suitable groups to avoid or suppress the ion movements$^{12,19}$, seeking effective ligands$^{17,46}$ to bond with the grain surface and also the film surface to reduce Joule heating induced by non-radiative recombination and finding stable charge injection layers$^{17}$.

In summary, by composition/phase engineering, we find that a quasi-2D perovskite of PEA$_2$(FAPbBr$_3$)$_n$PbBr$_4$ with $n=3$ composition showed good PL. We also introduce surface passivation to reduce the non-radiative recombination on the perovskite surface or grain boundaries. Combining composition/phase engineering and surface passivation, high performance of quasi-2D PeLEDs with a CE of 62.4 cd A$^{-1}$ and an external quantum efficiency of 14.36% is achieved.

**Methods**

**Materials**

The modified PEDOT:PSS (m-PEDOT:PSS) solution is a mix of normal PEDOT:PSS (AI 4083) aqueous solution and 100 mg ml$^{-1}$ PSS Na (Sigma Aldrich) aqueous solution by a volume ratio of 6:5. TOPO, PbBr$_2$, dimethyl sulfoxide (DMSO) and chlorobenzene (CB) were purchased from Sigma Aldrich. 2,2',7',2"-
Device fabrication. The indium tin oxide (ITO)-coated glass substrates were sequentially cleaned in detergent, distilled water, acetone and isopropanol by sonication and used as anode. The cleaned substrates were ultraviolet ozone treated for 15 min to make the surface hydrophilic, then m-PEDOT:PSS (or normal PEDOT:PSS) aqueous solution was spin-coated at 9000 rpm (or 2500 rpm) for 40 s and baked at 160 °C for 15 min in ambient air. Thereafter, the substrates were transferred into a nitrogen-filled glove box, and the 0.6 mol l$^{-1}$ (Pb$^2+$ concentration) perovskite solution in DMSO was spin-coated onto the m-PEDOT:PSS (or normal PEDOT:PSS) films at 3000 rpm for 2 min, and after spin coating for 40 s, 100 µl of CB was poured onto the films for pinning the perovskite crystallization, followed by annealing on a hot plate at 90 °C for 1 h. The perovskite precursor solution was prepared according to Supplementary Table 1. For surface passivation, 2 mg ml$^{-1}$ TOPO in CB was dripped onto the perovskite films for a 1 min to let TOPO passivate the films surface enough and then spin-coated at 7000 rpm for 1 min. Finally, the fabrication of PeLEDs was completed by depositing TPBi, LiF (less than 1 nm) and Al electrode layer by layer through a shadow mask in a high vacuum thermal evaporator. The device area was 0.108 cm$^2$ as defined by the overlapping area of the ITO and Al electrode. The thickness of ITO and Al are 130 nm and 60 nm, respectively.

Material and device characterizations. XRD measurements were performed with a Rigaku D/max 2500H equipment with a conventional Cu target X-ray tube (Cu
The work function of the perovskite samples was measured by the Kelvin probe method using a scanning Kelvin probe microscope (HVResearch, S7000). The bandgap values of 3.00 and 0.35 nm for the excitation and emission slits, respectively, were used for PL measurements: excitation wavelength of 400 nm; bandpass of 3.00 nm and 0.35 nm for the emission and excitation slits, respectively; and increments of 0.5 nm and integration time of 0.3 s per point. We measured the light intensity with a laser power meter and the estimated light power density incident on the samples was about 3.5 mW cm−2. We calculated the absorbance spectra, which were recorded on an ultraviolet–visible spectrophotometer (Cary 5000, Varian). The FTIR spectra were recorded on a Fourier-transform infrared spectrophotometer (NIR 680 FTIR, Thermo Electron). The excitation wavelength for PL and TRPL is 340 nm, and the bandpass is 10 nm. The THz absorbance of the samples was about 3.5 mW cm−2. The ultraviolet–visible absorbance spectra were recorded on an ultraviolet–visible spectrophotometer (Cary 5000, Varian). The FTIR spectra were recorded on a Fourier-transform infrared spectrophotometer (NIR 680 FTIR, Thermo Electron). The excitation wavelength for PL and TRPL is both 340 nm, and the bandpass is 10 nm. The THz absorbance of the samples was about 3.5 mW cm−2.

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

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Author contributions
J.Y. conceived the idea, designed the experiment and analyzed the data. X.Y. fabricated devices and collected all data. X.Z., J.D., Z.C., Q.I., J.M., P.W., L.Z. and Z.Y. were involved in data analysis. J.Y. and X.Y. co-wrote the manuscript. J.Y. directed and supervised the project. All authors contributed to discussions and finalization of the manuscript.

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