Efficient emission of quasi-two-dimensional perovskite films cast by inkjet printing for pixel-defined matrix light-emitting diodes

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Abstract
Quasi-two-dimensional (quasi-2D) perovskites are promising materials for potential application in light-emitting diodes (LEDs) due to their high exciton binding energy and efficient emission. However, their luminescent performance is limited by the low-\(n\) phases that act as quenching luminescence centers. Here, a novel strategy for eliminating low-\(n\) phases is proposed based on the doping of strontium bromide (SrBr\(_2\)) in perovskites, in which SrBr\(_2\) is able to manipulate the growth of quasi-2D perovskites during their formation. It was reasonably inferred that SrBr\(_2\) readily dissociated strontium ions (Sr\(^{2+}\)) in dimethyl sulfoxide solvent, and Sr\(^{2+}\) was preferentially adsorbed around [PbBr\(_6\)]\(^{4-}\) through strong electrostatic interaction between them, leading to a controllable growth of quasi-2D perovskites by appropriately increasing the formation energy of perovskites. It has been experimentally proved that the growth can almost completely eliminate low-\(n\) phases of quasi-2D perovskite films, which exhibited remarkably enhanced photoluminescence. A high electroluminescent efficiency matrix green quasi-2D perovskite-LED (PeLED) with a pixel density of 120 pixels per inch fabricated by inkjet printing technique was achieved, exhibiting a peak external quantum efficiency of 13.9\%, which is the most efficient matrix green quasi-2D PeLED so far to our knowledge.

Supplementary material for this article is available online

Keywords: quasi-2D perovskite, inkjet printing, SrBr\(_2\) doping, phase distribution, pixel-defined matrix LEDs

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1. Introduction

Metal halide perovskites exhibited excellent optoelectronic properties, including high emission efficiency, high color purity, tunable band gap, etc [1–3]. These characteristics have made them extremely promising candidates for LEDs for potential display applications. So far, the external quantum efficiency (EQE) of green and red PeLEDs has both exceeded 25% based on a spin-coating technology [4–6]. However, the spin-coating method cannot match the industrial manufacturing requirements of large-area, fine-patterned and high-density light-emitting pixel arrays, as well as colorful images for display applications. Instead, inkjet printing is the most promising technique for full-color display applications [7–9]. However, the electroluminescent (EL) device performance of pixel-defined matrix PeLEDs via inkjet printing technology is significantly inferior to those of spin-coating devices [10–13]. This is due to the more sophisticated processes of inkjet printing to fabricate high-quality pixel-defined matrix perovskite films and to control favorable film structures. These processes need to consider the ink formulations, substrate surface properties, inkjet processing and droplet drying conditions. This is because the rheological properties and solvent evaporation processes of ink drops can seriously influence the surface morphology and crystallization of perovskites, and then the EL performance of the devices based on perovskites.

Quasi-two-dimensional (Quasi-2D) perovskites with Ruddlesden–Popper (RP) structure have the general formula of $\text{L}_2\text{A}_{n-1}\text{MX}_{3n+1}$, where L is a large-size organic spacer cation, with low conductivity including butylammonium (BA) or phenylethylammonium (PEA), A is Cs, methylammonium cation, with low conductivity including butylammonium (BA) promising light-emitting materials with superior luminescent properties and thus become even more promising light-emitting materials [16, 17]. However, among quasi-2D perovskites, low-$n$ phases are unfavorable to luminescence and need to be suppressed [18]. Unfortunately, the formation of low-$n$ phases is more favorable because of their lower formation energy [19]. Substantial effort has been made to diminish the low-$n$ phase [4, 19–22]. One way is to mix spacer cations in the perovskite precursor to suppress the formation of undesired low-$n$ phases [19–22]. However, the low-$n$ phases cannot be removed completely. Another way is to introduce the tris(4-fluorophenyl)phosphine oxide (TFPPO) to produce an interaction with PEA spacer cations through strong hydrogen bonding [4]. Unfortunately, this method requires an anti-solvent process while casting the films. It is difficult to adopt anti-solvent technology in fabricating high-quality small-sized pixel films for full-color display applications via inkjet printing. Other ways, univalent metal cations, such as sodium ion (Na$^+$) or potassium ion (K$^+$), can also reduce the low-$n$ phases due to the strong electrostatic interaction between Na$^+$, K$^+$ and [PbBr$_6$]$^{4-}$ [23, 24]. However, the low-$n$ phases cannot be removed completely. Divalent metal cations, such as barium ion (Ba$^{2+}$), strontium ion (Sr$^{2+}$) and calcium ion (Ca$^{2+}$) were demonstrated to function in improving the surface morphology, passivating the grain boundary defects, and expanding the band gap of perovskite films by partially substituting Pb [25–27]. However, the influence of doping divalent metal cations on phase distribution in quasi-2D perovskite films is rarely discussed.

Here, we propose a strategy to manipulate the phase distribution of quasi-2D perovskite films with the divalent metal cation of strontium from strontium bromide (SrBr$_2$). It is found that SrBr$_2$ can control the growth of quasi-2D perovskites by appropriately increasing the formation energy of perovskites. In this way, the low-$n$ phases of quasi-2D perovskites can be completely suppressed. As a result, the quasi-2D perovskite films with SrBr$_2$ exhibited PL efficiency of 55%. The matrix green quasi-2D PeLED based on the films exhibited a peak EQE of 13.9% via inkjet printing, which is the most efficient matrix green PeLED so far. Moreover, the EQE was over 9% for large-area EL devices (10.0 cm$^2$) and 8.6% for flexible PeLEDs.

2. Methods

2.1. Materials

PEABr (99.5%), CsBr (99.9%), PbBr$_2$ (99.99%) and Poly-TPD were purchased from Xi’an Polymer Light Technology Corp., PFN-Br from Organtecsolar Materials Inc., PEG (average Mw ≈ 10 000) from Aladdin, dimethyl sulfoxide (DMSO) (>99.0%) from TCI, SrBr$_2$ (99%) and LiF from Alfa Aesar and TPBi from Lumtec. All chemicals were used as received.

2.2. Perovskite inks

The green perovskite ink was prepared by dissolving 0.08 M CsBr, 0.08 M PbBr$_2$, 0.032 M PEABr, 0.5 mg ml$^{-1}$ PEG and 6% SrBr$_2$ in DMSO. All inks were stirred vigorously at 40 °C for 12 h, then filtered by 0.22 μm polytetrafluoroethylene (PTFE) prior to inkjet printing.
2.3. Device fabrication

The pixel-defined matrix substrates were provided by Guangzhou New Vision Opto-Electronic Technology Co., Ltd (China). The pixel size was 50 μm in width and 190 μm in length. The thickness of the pixel definition layer was 1.5 μm. The patterned substrates were first ultrasonically cleaned with DI water, then dried at 75 °C in a baking oven. After a 10 min oxygen plasma treatment, a Poly-TPD layer was spin-coated (in chlorobenzene, 15 mg ml⁻¹) onto the ITO substrate at 3000 rpm for 30 s, and the resulting coated substrate was annealed at 140 °C for 20 min. A PFN-Br layer was spin-coated (in methanol, 0.6 mg ml⁻¹) at 3000 rpm for 30 s. The perovskite films were then coated via a high-precision Jet-Lab II printer (Micro-Fab Technologies Inc.) equipped with a 30 μm diameter piezoelectric-driven inkjet nozzle in the air. The coated substrates were then dried in a vacuum for 10 min and then annealed at 70 °C for 5 min. Finally, the samples were transferred from the N₂-filled glove box into a high-vacuum deposition system (base pressure ≈1 × 10⁻⁴ Pa) for thermal evaporation of TPBi (40 nm) and LiF (1 nm)/Al (100 nm) through shadow masks to produce a 0.1 cm² active area.

2.4. Characterization

The crystalline structures of the perovskite films were investigated using an x-ray diffractometer (PANalytical X’pert PRO) equipped with a Cu–Kα x-ray tube. UV–vis absorption spectra were obtained by a UV–vis spectrophotometer (HP 8453E). SEM images of the perovskite films were acquired by an electron microscope (ZEISS Merlin) operating at 10 kV. UV–vis absorption spectra (190–2100 nm) and photoluminescence (PL) spectra of the films fabricated by the three inks differ substantially, and were 30%, 24% and 55% for the pristine film, PbBr₂-incorporated film and SrBr₂-incorporated film, respectively (figures 1(b) and S1). These results suggest that the elimination of low-n phases in SrBr₂-incorporated film can provide the highest PL efficiency, which is consistent with reported works [20]. The PbBr₂-incorporated film with a large number of low-n phases exhibited the lowest PL efficiency. Density functional theory calculations (figure S2) showed that the value of octahedral formation energy of [PbBr₆]⁴⁻ (−13.808 eV) was significantly lower than that of [SrBr₆]²⁻ (−12.522 eV), suggesting that CsPbBr₃ unit cells are easier to form than CsSrBr₃ unit cells. This speculation can be proved by comparing the delay time (the time elapsed from the beginning of spin-coating to the appearance of light emission) during the film-forming process (figure S3). The pristine film began to produce PL emission within 27 s, which was significantly faster than that of the SrBr₂-incorporated film (36 s), given that the 0.2 M perovskite precursor was spin-coated on a quartz substrate at 4000 rpm. The remarkably delayed crystallization time indicated higher formation energy of quasi-2D perovskites with SrBr₂ [31]. In addition, it was found that other alkaline earth metal bromides (MgBr₂, CaBr₂, BaBr₂) can also delay crystallization time, suggesting that alkaline earth metal bromide can increase the formation energy of quasi-2D perovskites and eliminate low-n phases (figures S3 and S4) [19]. The SrBr₂-incorporated film exhibited the highest PLQY, which was attributed to an appropriate increase in the formation energy of quasi-2D perovskite doped with Sr²⁺ and its ionic radius similar to that of Pb²⁺ in perovskites [32].

The XRD patterns with obvious diffraction peaks at 5.23° and 10.62°, respectively, suggested that the PbBr₂-incorporated film contained more n = 1 phases (figure 1(d)) [29, 33]. After doping 6% concentration of SrBr₂, the full width at half maximum of the diffraction peak at 15.25° decreased from 0.70° to 0.46°, indicating an increase in the grain size of perovskites, which was consistent with the results of the SEM and atomic force microscope images (figures 1(e), S5 and S6). However, there was almost no change in the perovskite peak position due to the similar ionic radiiues of Sr²⁺ (1.41 pm) and Pb²⁺ (1.26 pm) [32]. In the absorption spectra of SrBr₂ and PbBr₂ solutions (figure S7), clear absorption at about 345 nm was detected from [PbBr₆]⁴⁻ frames in PbBr₂ solution, while no signal appeared in SrBr₂ solution. Based on the results that almost no [SrBr₆]²⁻ frames in the SrBr₂ solution, it was deduced that the SrBr₂ solution was in an ionic solution state in the DMSO solvent. Therefore, we may speculate on the crystallization processes of PbBr₂ and SrBr₂-based quasi-2D perovskites (figure 2). During the growth of quasi-2D perovskites, the imbalance of the crystallization rate of different phases led to severe phase separation. For PbBr₂-incorporated ink, PbBr₂ exists in the form of [PbBr₆]⁴⁻ in DMSO, which provides more components of

3. Results

The peaks at 403, 434, 464 and 483 nm in the absorption spectra of the quasi-2D perovskite with RP films were accordingly assigned to the low-n (n = 1, 2, 3, 4) phases, respectively, which deteriorate the luminescence efficiency due to the strong exciton–phonon coupling, and low charge transport properties in low-n phases, especially n < 4 (figure 1(a)) [28–30]. In order to suppress the low-n phases, SrBr₂ was specifically introduced into the perovskites. For comparative investigation, three kinds of ink were prepared, including perovskite ink without additive (Ink 1), perovskite ink with 6% (mole ratio between PbBr₂ and CsBr) PbBr₂ added (Ink 2), and perovskite ink with 6% (mole ratio between SrBr₂ and CsBr) SrBr₂ added (Ink 3). The absorption spectra exhibited that the low-n peaks of SrBr₂-incorporated film fabricated using Ink 3 almost disappeared, and so did the strongest low-n peaks of PbBr₂-incorporated film using Ink 2 as well as the slightly weak low-n peaks of pristine film using Ink 1 (figure 1(a)). The PLQYs of the films fabricated by the three inks differ substantially, and were 30%, 24% and 55% for the pristine film, PbBr₂-incorporated film and SrBr₂-incorporated film, respectively (figures 1(b) and S1). These results suggest that the elimination of low-n phases in SrBr₂-incorporated film can provide the highest PL efficiency, which is consistent with reported works [20]. The PbBr₂-incorporated film with a large number of low-n phases exhibited the lowest PL efficiency. Density functional theory calculations (figure S2) showed that the value of octahedral formation energy of [PbBr₆]⁴⁻ (−13.808 eV) was significantly lower than that of [SrBr₆]²⁻ (−12.522 eV), suggesting that CsPbBr₃ unit cells are easier to form than CsSrBr₃ unit cells. This speculation can be proved by comparing the delay time (the time elapsed from the beginning of spin-coating to the appearance of light emission) during the film-forming process (figure S3). The pristine film began to produce PL emission within 27 s, which was significantly faster than that of the SrBr₂-incorporated film (36 s), given that the 0.2 M perovskite precursor was spin-coated on a quartz substrate at 4000 rpm. The remarkably delayed crystallization time indicated higher formation energy of quasi-2D perovskites with SrBr₂ [31]. In addition, it was found that other alkaline earth metal bromides (MgBr₂, CaBr₂, BaBr₂) can also delay crystallization time, suggesting that alkaline earth metal bromide can increase the formation energy of quasi-2D perovskites and eliminate low-n phases (figures S3 and S4) [19]. The SrBr₂-incorporated film exhibited the highest PLQY, which was attributed to an appropriate increase in the formation energy of quasi-2D perovskite doped with Sr²⁺ and its ionic radius similar to that of Pb²⁺ in perovskites [32].

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low-\(n\) phases due to the lower formation energy. However, for SrBr\(_2\)-incorporated ink, SrBr\(_2\) existed in the form of Sr\(^{2+}\) in DMSO. Compared with Cs\(^{+}\) and PEA\(^{+}\), Sr\(^{2+}\) was relatively likely to be adsorbed on [PbBr\(_6\)]\(^{4-}\) frames because of strong electrostatic interactions, due to its smaller size and larger amount of charge [23, 24, 34]. The adsorption of Sr\(^{2+}\) on the surface of [PbBr\(_6\)]\(^{4-}\) frames might increase the formation energy of perovskites and balance the crystallization rates of different phases, resulting in slow and homogeneous growth of perovskite crystals.

The pristine film showed five distinct bleach peaks in the transient absorption spectra, corresponding to \(n = 1, 2, 3, 4,\) and \(n \geq 5\) phases, respectively (figures 3(a) and (c)). The PbBr\(_2\)-incorporated film showed stronger peaks at 404, 435, 465 and 483 nm, indicating the formation of more low-\(n\) phases (figures S8(a) and (b)), whereas the SrBr\(_2\)-incorporated
film showed a prominent peak at 508 nm dominated by $n > 5$ phases (figures 3(b) and (d)). The intensity evolution of the ground-state bleach (GSB) peaks with time and the redshift of the GSB peaks of $n \geq 5$ phases were consistent with an energy transfer from low-$n$ to high-$n$ phases (figures 3(d) and S9(a)). We extracted the fast component decay constants ($\tau_1$) of each phase by tracing the bleaching signals on different timescales to estimate the energy transfer kinetics (figure S9) [35]. After doping SrBr$_2$, the decrease in $\tau_1$ for $n \geq 5$ phases from 56.66 to 40.54 ps indicated faster energy transfer and more efficient radiation recombination, which were precisely in line with the TRPL results, as illustrated in figure 1(c). Compared to pristine film, the excited state lifetime of the SrBr$_2$-incorporated perovskite film increased from 26.2 to 39.5 ns, manifesting remarkably suppressed non-radiative recombination (figure S10). According to previous reports, a SCLC technique was utilized to characterize the film’s trap densities [35]. Figure 1(f) displays the dark current density–voltage ($J$–$V$) curves of the hole-only devices with the device structure of ITO/Poly-TPD/PFN-Br/perovskite/MoO$_x$/Ag. The defect density was calculated according to the equation $n_t = \frac{2V_{TFL}\varepsilon_0\varepsilon_0}{eL^2}$, where $n_t$ is the trap density, $\varepsilon$ is the relative dielectric constant, $\varepsilon_0$ is the vacuum permittivity, $L$ is the thickness of the perovskite layer and $e$ is the elementary charge. Based on the equation, the calculated defect density of the 6% SrBr$_2$ doped sample was $5.28 \times 10^{16}$ cm$^{-3}$, comparably lower than that of the pristine sample ($9.08 \times 10^{16}$ cm$^{-3}$), which was ascribed to the retarded crystallization rate and likely to form high-quality films with lower defects [31].

Pixel-defined matrix green PeLEDs with 120 PPI resolution were constructed based on multi-type perovskite films prepared by in situ inkjet printing with the unified device architecture of glass/ITO/Poly-TPD/PFN-Br/perovskites/TPBi/LiF/Al. In this structure, the indium tin oxide (ITO) acted as an anode, the Poly-TPD as the hole-transport layer and the ~5 nm thick PFN-Br, prepared by the spin-coating method, proved an effective interface layer [36]. The green perovskite pixel layer was prepared by inkjet printing, and 2,2’,2”-(1,3,5-benzinetriyl)-tris(1-phenyl-1H-benzimidazole) (TPBi) acted as the electron-transport layer, LiF and Al layer, prepared by vacuum evaporation (figure 4(a)). The energy level diagram of the PeLED is shown in figure S11, and all the data are from the literature [37]. The PeLED doped with SrBr$_2$ reached a maximum brightness and peak EQE of 15194 cd m$^{-2}$ and 13.9%, respectively (figures 4(b)–(d) and S12, S13), which is the most efficient PeLED via ink-jet printing technique to date to our knowledge (table S1). Uniform EL emission was observed over the entire pixel area under a driven voltage of 3.5–5.5 V, indicating the even thickness of perovskite films (figures S14 and S15). A histogram of the maximum EQE values from 22 devices (figure 4(e)) shows an average EQE of 11.3% and good reproducibility. The SrBr$_2$-incorporated device (initial luminance ($L_0$) was 796 cd m$^{-2}$) had a prolonged lifetime (defined as $T_{50}$, luminance decay to half of the initial value) of 681 s, while the lifetime of the reference device without SrBr$_2$ ($L_0$ was 765 cd m$^{-2}$) was only 338 s (figure 4(f)).

In order to further prove the robustness and scalability of inkjet printing technology, relatively large-area films were...
Figure 4. Matrix device structure and performance of PeLEDs without and with 6% SrBr₂. (a) PeLED architecture, (b) current $J$–$V$–luminance curves, (c) current efficiency-current density curves, (d) EL spectra, and the inset shows the corresponding photo of a working PeLED, (e) histogram of device performance and (f) lifetime measurement ($T_{50}$) of the PeLEDs.

Figure 5. Performances of PeLEDs with a 10.0 cm² active area and flexible PeLEDs. (a) PL photo of perovskite film, (b) current $J$–$V$–luminance curves, (c) current efficiency-current density curves, and the inset shows the corresponding photo of a working PeLED device operated under voltage of 4.5 V, with an active area of 10.0 cm², (d) performance of flexible PeLEDs under different bending times, and the inset shows the corresponding photo of a working flexible PeLED device operated under voltage of 4.0 V.

prepared with an area of $4.0 \times 2.5$ cm² (figure 5(a)). The peak EQE of the PeLEDs with an active area of 10.0 cm² reached 9.3%, which is the most efficient large-area PeLED via inkjet printing technology to date (figures 5(b), (c) and S16). As schematically shown in figure 5(d), corresponding flexible PeLEDs were prepared on a structure of polyimide (PI)/ITO/Poly-TPD/PFN-Br/perovskites/TPBi/LiF/Al, and their bending resistance was tested by evaluating device
performance with different bending cycles (figure S17). The EQEs decreased to 68.6% of the initial EQE after 200 cycles of bending, which proved that the defined-pixel matrix PeLED via inkjet printing has great potential in the application of flexible devices.

4. Conclusion

We systematically and comparatively investigated the effects of PbBr$_2$ and SrBr$_2$ on the crystallization of quasi-2D perovskites. Compared with PbBr$_2$ additive, SrBr$_2$ can effectively dissociate into Sr$^{2+}$ in DMSO and preferentially adsorb surrounding [PbBr$_6$]$^{4-}$ through strong electrostatic interaction between them. This leads to slow and homogeneous growth of quasi-2D perovskites due to the appropriately increased formation energy of the perovskites. The manipulation of the growth of quasi-2D perovskites contributes to a more homogeneous n-phase distribution and more efficient radiative recombination. Based on these results, an inkjet-printed green matrix PeLED with a pixel density of 120 PPI was realized using a divalent metal ion strategy for manipulating the phase fitting from the robustness of inkjet printing technology, large-area (10 cm$^2$) PeLEDs with a peak EQE of 9.3% and flexible PeLEDs with a peak EQE of 8.6% were achieved. Therefore, using a divalent metal ion strategy for manipulating the phase distribution may provide a route to fabricate efficient PeLED displays via the inkjet printing technique.

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Conflict of interest

The authors declare that they have no conflict of interest.

Author contributions

Junjie Wang and Junbiao Peng designed the experiments; Junjie Wang performed the experiments; Junjie Wang and Danyang Li helped with the measurements. Junjie Wang and Danyang Li helped with the inkjet printing operation. Junjie Wang wrote the paper with support from Junbiao Peng. Jian Wang gave valuable advice. All authors contributed to the general discussion.

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