

Zwitterionic additive for high-performance pure-blue perovskite light-emitting diodes

Hongling Yu and Feng Gao*

Linköping University, Department of Physics, Chemistry and Biology (IFM), Linköping, Sweden

Metal halide perovskites have emerged as promising candidates for light-emitting diodes (LEDs) due to their cost effectiveness, solution processability, and impressive luminescence.¹ At present, the external quantum efficiencies (EQEs) of sky-blue,² green,³ red,⁴ and near-infrared perovskite LEDs (PeLEDs) have exceeded 20%,⁵ demonstrating significant potential for future display and communication technologies. However, achieving high-performance pure-blue PeLEDs with a peak wavelength of 460–470 nm, which is an indispensable color, has been challenging.⁶

The emission color of PeLEDs can be tuned through halide mixing or contraction of the quantum-confinement effect. In the pursuit of pure-blue PeLEDs, the most straightforward approach is mixing bromide with a high load of chloride. However, the prevalent challenge arising from halide ion migration and phase segregation significantly hinders color purity during device operation.^{7,8} Additionally, chloride perovskites exhibit a higher density of deep-level traps and lower intrinsic defect tolerance compared to their mixed bromide and iodine counterparts, posing obstacles to achieving a breakthrough EQE exceeding 10%.⁹

Reduced-dimensional (quasi-2D) pure bromide perovskites have emerged as a promising alternative for high-performance PeLEDs, yet challenges persist for achieving pure-blue LEDs.^{10,11} Leveraging the

quantum-confinement effect, quasi-2D perovskites facilitate enhanced radiative recombination rates, leading to higher photoluminescence quantum yields (PLQYs). However, the incorporation of large organic cations triggers severe charge injection issues, resulting in poor EQE and consequently low luminance. Additionally, the inferior distribution across multiple dimensions can lead to broad emission peaks, ultimately hindering the color purity of the devices. State-of-the-art quasi-2D PeLEDs have demonstrated high EQE of 28.1% and luminance of 36,500 cd m⁻², attributed to defect passivation and structure regulation;¹² however, when targeting higher bandgaps for pure-blue LEDs, the requirement for additional large organic cations can exacerbate charge injection issues, consequently leading to poor device performance.

Employing a combination of strategies utilizing mixed Br/Cl and quasi-2D configurations holds promise for maximizing the benefits of both approaches, pushing the emission color towards pure-blue while minimizing the charge injection problem. Recently, Hou et al. reported a color-stable pure-blue 466-nm PeLED based on mixed-halide quasi-2D, achieving a record luminance of 1806 cd m⁻² and a significantly increased EQE of 9.25% (Fig. 1).¹³ This enhanced performance is attributed to the multifunctional zwitterionic molecule additive, 3-(benzyltrimethylammonio)propanesulfonate (3-BAS; see the molecular structure in Fig. 1).

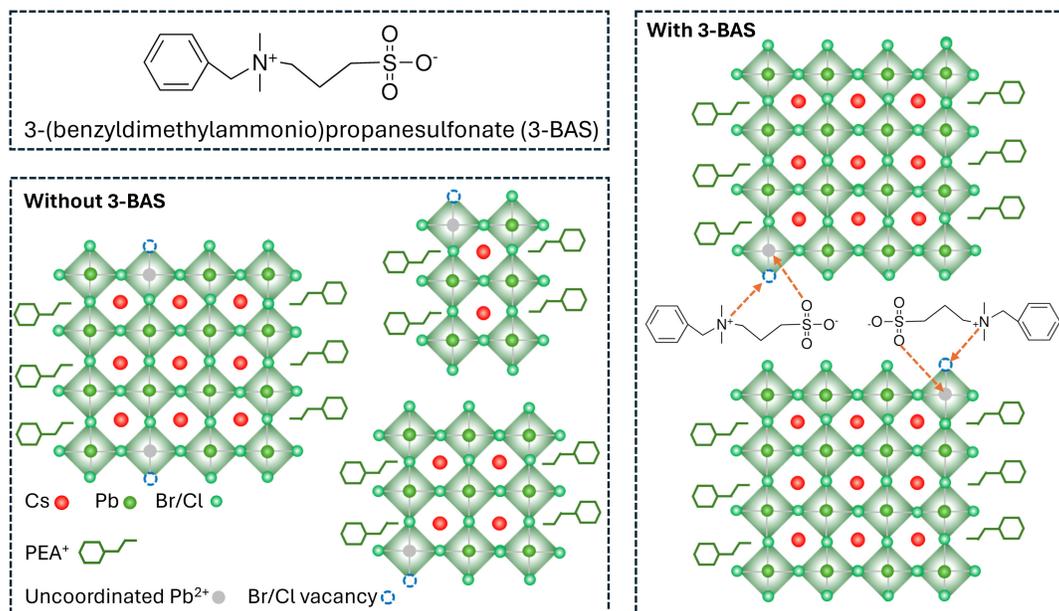


Fig. 1 The molecular structure of 3-BAS and the schematics of surface defect passivation and crystallization regulation via 3-BAS incorporation.

*Address all correspondence to Feng Gao, feng.gao@liu.se

© The Authors. Published by SPIE and CLP under a Creative Commons Attribution 4.0 International License. Distribution or reproduction of this work in whole or in part requires full attribution of the original publication, including its DOI. [DOI: [10.1117/1.AP.6.2.020501](https://doi.org/10.1117/1.AP.6.2.020501)]

3-BAS incorporation can effectively passivate defects and regulate the crystallization of mixed halide quasi-2D perovskites (Fig. 1). The achieved performance during device operation stems from: 1) the passivation of surface defects facilitated by Lewis base-acid reaction between the sulfonic group ($-\text{SO}_3^-$) of 3-BAS and the uncoordinated Pb^{2+} ions and 2) the inhibition of halide migration via the formation of electrostatic interactions between the electropositively quaternary ammonium group ($-\text{R}_4\text{N}^+$) of 3-BAS and the electronegatively halide ions. The suppression of the two primary factors contributing to the phase segregation observed in mixed-halide perovskites ensures the stable pure-blue LED. Furthermore, the incorporation of 3-BAS molecule can inhibit the formation of small- n phases during the perovskite crystallization because of the strong interaction of 3-BAS with Pb^{2+} and halide ions, resulting in well-oriented perovskite layers and higher quality film morphology. This will enhance the energy transfer process, leading to higher radiative recombination and thereby enhanced luminance and EQE in the device.

Hou et al. further explored the potential application of the resultant pure-blue PeLEDs in visible light communication (VLC).¹³ The 3-BAS-treated PeLED served as a high-speed light source for a low-loss VLC system, exhibiting superior signal restoration ability and demonstrating effective electrical-optical-electrical signal conversion. These findings highlight the potential application for high-performance wireless communication. However, perovskite LEDs for optical communication are still at the early stage of development, falling short of necessary standards. While conventional inorganic thin-film technologies dominate, we see potential for solution-processed perovskite-based LEDs to offer complementary capabilities.^{14,15}

Continued efforts in the development of pure-blue PeLEDs should prioritize not only further narrowing the EQE gap with other counterparts but also extending long-term operational stability (lifetime) and minimizing the efficiency roll-off. As previously discussed, a combination of different strategies, such as mixed halides, the quantum confinement effect, and other materials composition engineering, might be needed for advancing the EQE of pure-blue PeLEDs.¹ Furthermore, despite enhancements in EQEs, state-of-the-art blue PeLEDs frequently exhibit insufficient long-term operational stability from several seconds to minutes.^{2,9} Device lifetime is further complicated by the stability of not only the perovskite film but also the interlayers within the entire device during operation. Exploring new device structures, such as the screening of all-inorganic charge injection layers, could aid in achieving durable pure-blue perovskite LEDs.

A common key issue for LEDs, while receiving less discussion in pure-blue PeLEDs, is the efficiency reduction at high current densities, known as efficiency roll-off.¹⁶ Addressing this challenge requires a heightened focus on achieving a balanced carrier injection regime, minimizing Auger losses, and exploring thermal management in future device developments.¹⁷ We believe that as research continues to advance, existing barriers will be overcome, leading to advancements in full display and optical communication technologies.

Disclosures

The authors declare no competing interests.

Acknowledgments

H.Y. is supported by the Swedish Research Council Vetenskapsrådet (Grant No. 2021-06668).

References

1. X.-K. Liu et al., "Metal halide perovskites for light-emitting diodes," *Nat. Mater.* **20**(1), 10 (2021).
2. S. Yuan et al., "Efficient blue electroluminescence from reduced-dimensional perovskite," *Nat. Photonics* (2024).
3. J. S. Kim et al., "Ultra-bright, efficient and stable perovskite light-emitting diodes," *Nature* **611**(7937), 688 (2022).
4. J. Jiang et al., "Red perovskite light-emitting diodes with efficiency exceeding 25% realized by co-spacer cations," *Adv. Mater.* **34**(36), 2204460 (2022).
5. W. Xu et al., "Rational molecular passivation for high-performance perovskite light-emitting diodes," *Nat. Photonics* **13**(6), 418 (2019).
6. K. Zhang et al., "Blue halide perovskite materials: preparation, progress, and challenges," *Laser Photonics Rev.* **17**(2), 2200689 (2023).
7. E. T. Hoke et al., "Reversible photo-induced trap formation in mixed-halide hybrid perovskites for photovoltaics," *Chem. Sci.* **6**(1), 613 (2015).
8. H. Yu et al., "Color-stable blue light-emitting diodes enabled by effective passivation of mixed halide perovskites," *J. Phys. Chem. Lett.* **12**(26), 6041 (2021).
9. X. Luo et al., "Effects of local compositional heterogeneity in mixed halide perovskites on blue electroluminescence," *Matter* **7**(3), 1054 (2024).
10. H. Gu et al., "Phase-pure two-dimensional layered perovskite thin films," *Nat. Rev. Mater.* **8**(8), 533 (2023).
11. Z. Chu et al., "Large cation ethylammonium incorporated perovskite for efficient and spectra stable blue light-emitting diodes," *Nat. Commun.* **11**(1), 4165 (2020).
12. Z. Liu et al., "Perovskite light-emitting diodes with EQE exceeding 28% through a synergetic dual-additive strategy for defect passivation and nanostructure regulation," *Adv. Mater.* **33**(43), 2103268 (2021).
13. C. Shen et al., "High performance and stable pure-blue quasi-2D perovskite light-emitting diodes by multifunctional zwitterionic passivation engineering," *Adv. Photonics* **6**(2) 026002 (2024).
14. A. Liu, C. Bi, and J. Tian, "All solution-processed high performance pure-blue perovskite quantum-dot light-emitting diodes," *Adv. Funct. Mater.* **32**(44), 2207069 (2022).
15. A. Ren et al., "Emerging light-emitting diodes for next-generation data communications," *Nat. Electron.* **4**(8), 559 (2021).
16. W. Zou et al., "Minimising efficiency roll-off in high-brightness perovskite light-emitting diodes," *Nat. Commun.* **9**(1), 608 (2018).
17. X. Shen et al., "Passivation strategies for mitigating defect challenges in halide perovskite light-emitting diodes," *Joule* **7**(2), 272 (2023).