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# Over a Decade of Progress in Metal-Halide Perovskite Light-Emitting Diodes

*Tae-Woo Lee*

Metal-halide perovskites have emerged as a highly promising material for light-emitting diodes (LEDs), driving significant progress in the performance of perovskite LEDs (PeLEDs), particularly in terms of external quantum efficiency (EQE), which has reached as high as 30% due to rapid progress over the past 10 years. This marks a dramatic improvement from the low EQEs of ≈0.1% observed using bulk polycrystalline films with large grains, as reported online in August 2014[1] and November 2014[2] at room temperature. Despite the inherent challenges posed by the low exciton binding energy and long exciton diffusion length of bulk perovskites,[1,2] which hinder efficient radiative recombination, material strategies to overcome those limitations have been developed to confine charge carriers within nanoscale structures, referred to as nanocrystalline perovskites.[3] This approach enabled the first high-efficiency PeLEDs, thereby triggering a surge in research by demonstrating the potential for achieving commercially viable efficiencies comparable to those exhibited by organic light-emitting diodes (OLEDs) or inorganic quantum dot light-emitting diodes (QLEDs). Three main categories of nanocrystalline perovskites have been identified: nanoscale polycrystalline perovskites, quasi-2D perovskites, and perovskite nanocrystals (PNCs). Each perovskite has undergone distinct material engineering strategies, contributing significantly to improved device efficiencies (**Figure** 1A).

Simultaneously, significant progress has been made in the development of charge-transporting layers (CTLs) specifically designed for perovskite materials. Research has extensively focused on optimizing the band alignment and charge mobility of CTLs, as well as their interactions with perovskite crystallization and chemical properties, leading to improved device efficiency.

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Additionally, several strategies have been explored to enhance the outcoupling efficiency. Perovskites, in particular, offer the unique advantage of photon recycling and scattering structures, which further contribute to increasing outcoupling efficiency (Figure 1A).

While the efficiency and operational lifetime of PeLEDs have improved, several challenges persist. Notably, the operational lifetime must exceed $10^6$ h, but the current status remains below $10^5$ h only for green PeLEDs. In response to concerns about lead toxicity, lead-free perovskite LEDs have achieved EQE over 20% recently, but their emissions are mainly focused on red, and their brightness and stability are still limited. Additionally, achieving high efficiency in the deep-blue emission region (<465 nm) remains an ongoing challenge. Furthermore, PeLEDs emitting in the short-wave infrared (SWIR) range are still relatively rare.

Beyond electroluminescence in PeLEDs, strategies for commercialization are increasingly important. Down-conversion applications are promising, emphasizing the need for the synthesis of highly stable perovskite light emitters suitable for mass production. Additionally, large-area printing and patterning techniques must be developed to support this scalability. Recently, optical phenomena in perovskites, such as amplified spontaneous emission, superfluorescence, and single-photon emission, have garnered significant interest for potential applications in lasers and quantum communication systems.

To commemorate the 10th anniversary of this breakthrough, this Special Issue of *Advanced Materials* covers the remarkable progress achieved across materials development, device engineering, and optical characterization, featuring 29 articles from world-leading experts.

## 1. Nanoscale Polycrystalline Perovskites

Polycrystalline perovskites typically exhibit grain structures ranging from hundreds of nanometers to micrometers. Due to their relatively low exciton binding energy, excitons can thermally ionize into free carriers, which predominantly emit via bimolecular recombination at higher carrier densities. However, defects at grain boundaries often act as charge traps, significantly limiting the EQE. In 2015, a strategy was proposed to reduce the grain size below 100 nm as a crucial step to improve efficiency.[3] This led to the development of nanoscale polycrystalline perovskites, which utilize nanocrystal pinning, and additive-controlled crystallization to precisely modulate nucleation, and growth, resulting in perovskites with nanoscale grain sizes. The more advanced format is in situ core-shell perovskite, which directly forms crystals around ≈10 nm in size, while simultaneously passivating, forming a shell with short organic ligands (Nature, 2022, 611, 688). This approach effectively minimizes grain size, and enhances

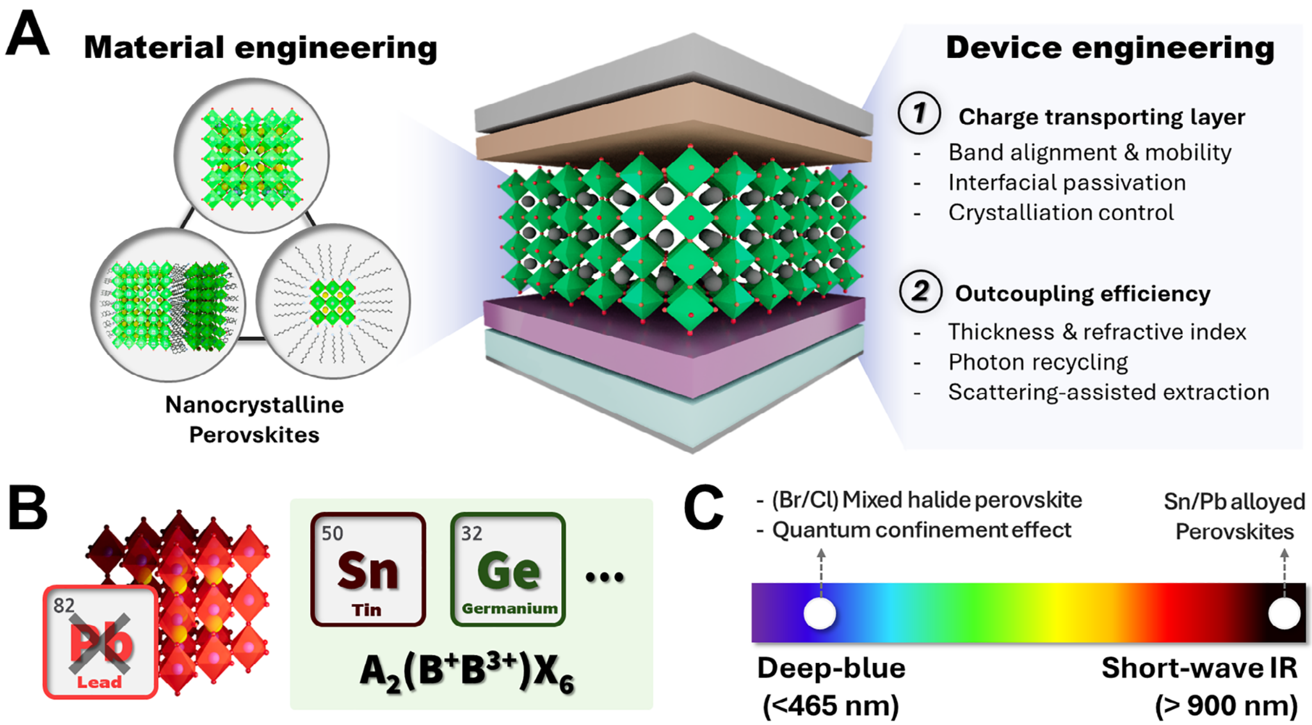


**Figure 1.** A) Material and device engineering strategies to enhance the performance of PeLEDs. B) Remaining challenges in lead-free perovskite development and potential candidates. C) Emission tuning strategies for deep-blue and short-wave IR region.

defect passivation, leading to significantly improved EQE, and operational stability. Tae-Woo Lee, co-workers (adma.202415648) have provided a comprehensive summary of the development of these nanoscale polycrystalline perovskites, as well as the overall progress in PeLEDs.

## 2. Quasi-2D Perovskites

Quasi-2D perovskites exhibit a layered structure consisting of octahedral slabs separated by organic spacer cations. This arrangement facilitates strong exciton confinement, significantly enhancing radiative excitonic recombination. Typical quasi-2D perovskites involve multiple $n$-phases, creating an energy cascade that transfers excitons toward lower bandgap $n$-phases. Precisely managing the distribution and cascading path of these $n$-phases is crucial for achieving highly efficient perovskite LEDs (E. H. Sargent, D. Ma et al., adma.202410633; L. Dou et al., adma.202411998). Maintaining the stability of these layered structures remains challenging due to thermal and structural instabilities at edges and surfaces. W. Nie and co-workers (adma.202413412) investigated the thermal instability of quasi-2D phases, attributing it to weak interfacial interactions between the perovskite and organic spacer layers, which lead to phase segregation and reduced optical performance. Additionally, M. Yuan, Y. Jiang, and co-workers (adma.202412041) emphasized the detrimental effects of exciton-phonon coupling from vibrations of edge-dangling octahedra, demonstrating that anchoring these octahedra with appropriate ligands mitigates lattice vibrations, thereby improving optical stability and efficiency.

## 3. Colloidal Perovskite Nanoparticles

PNCs are colloidal nanoparticles surrounded by organic ligands, in which nanometer-scale perovskite crystals are embedded. L. Manna, Z. Shi, J. Dai, and co-workers (adma.202415606) summarized recent advances in PNCs for PeLEDs, including optimization of synthesis, ion doping, ligand passivation, and the development of emerging lead-free PNCs. Y. Kim, B. R. Lee, J. Choi, and co-workers (adma.202410128) reported dual ligand passivation strategies that effectively prevent the dissociation of PNCs into reduced-dimensional perovskites, thereby improving both stability, and efficiency. Z.-K. Tan, T. G. Hoang, co-workers (adma.202409564) introduced novel cross-linkable ligands that enable high-resolution photopatterning by enhancing solvent resistance, increasing the film density of PNC films. Additionally, W. C. H. Choy, and co-workers (adma.202415211) developed multidentate aromatic ligand strategies to significantly improve mechanical stability against repeated folding, thereby enhancing the adhesion between PNCs, adjacent layers, which addresses key challenges in foldable PeLED application.

## 4. Improving External Quantum Efficiency of PeLEDs

Material engineering of nanocrystalline perovskites plays a crucial role in enhancing device performance, but optimizing charge-transporting layers (CTLs) is equally essential. In addition to achieving proper charge balance throughout the device, functional groups on the CTLs can interact with the perovskite surface, significantly reducing surface defects and influencing crystallization through wettability and surface interactions. Z. Wei, J. Lu, and co-workers (adma.202410535) emphasized the multifunctionality of CTLs in PeLEDs.

Despite advances that have led to internal quantum efficiencies approaching 100%, approximately 75% of photons generated in PeLED devices remain trapped due to low outcoupling efficiency. To address this issue, extensive studies on both intrinsic and extrinsic light-outcoupling strategies have been conducted by S. Kumar, C.-J. Shih, and co-workers (adma.202413622).

## 5. Lead-Free Perovskites

Concerns about lead toxicity have driven the development of lead-free perovskites (Figure 1B). As substantial progress has been made in enhancing device performance, D. Di, B. Zhao, C. Zou, and co-workers (adma.202411020) summarized recent advancements in various lead-free perovskite emitters, crystallization processes, optimized device architectures, and strategies for improving stability and efficiency. While iodide-based Sn-perovskites have achieved efficiencies of over 20%, progress with Sn/Br-based perovskites remains slower. Y. Jin and co-workers (adma.202414841) developed an interfacial reaction-assisted crystallization method, significantly improving the film quality and emission performance of $CsSnBr_3$-based PeLEDs. H. Zhou and co-workers (adma.202413895) further investigated 2D Sn/Br-based blue-emitting perovskites by manipulating electron-phonon coupling through lattice rigidity engineering. Additionally, alternative lead-free options, such as Sb-based perovskites, have been explored. B. Ma and co-workers (adma.202412239) utilized self-trapped exciton emissions in Sb-based perovskites to develop solution-processed bilayer white PeLEDs.

## 6. Deep-Blue & SWIR PeLEDs

A significant challenge in PeLED development is extending spectral coverage toward both deep-blue (<465 nm) and short-wave infrared (SWIR, >900 nm) regions while maintaining high efficiency (Figure 1C). Chloride incorporation is a common strategy for inducing bandgap widening in deep-blue emission, but higher $Cl^-$ content often introduces chloride-related defects and internal halide heterogeneity, reducing spectral stability. To address this, N. Zhao and co-workers (adma.202414788) proposed a surface engineering approach using acetate-rich treatments to effectively passivate defects and suppress halide redistribution, achieving uniform mixed-halide compositions and improving spectral stability in the blue region. Besides, J. You and Z. Chu (adma.202409867) emphasized the development of pure-bromide-based PeLEDs to avoid halide segregation issues, demonstrating improved spectral stability in the blue region.

Regarding SWIR emission, there has been growing interest due to its broad range of applications, including biomedical imaging, non-invasive physiological sensing, night vision, surveillance, and optical communication. However, most PeLEDs currently operate in the near-infrared (NIR) range (700–900 nm), and stable emission above 900 nm remains rare. M. A. Loi and co-workers (adma.202415958) reported PeLEDs based on Sn–Pb alloyed perovskites that successfully emit at 988 nm, representing one of the few demonstrations of PeLEDs operating in the SWIR region.

## 7. Understanding and Characterization of Perovskite Materials

In addition to material engineering developments, substantial progress has been made in the understanding and characterization of perovskite materials. W. A. Tisdale and co-workers (adma.202415757) provide a comprehensive summary of exciton and charge transport dynamics in perovskites. L. Nienhaus, S. Wieghod, and Y. Luo (adma.202411916) introduce advanced spectro-microscopy techniques, enabling spatially resolved insights into the properties of perovskites, thereby offering a deeper understanding of their local behavior. Furthermore, B. Hu, Y. Ma, and co-workers (adma.202411913) explore the impact of spin–orbit coupling on perovskite light-emitting characteristics, including amplified spontaneous emission (ASE) and circularly polarized emission.

## 8. Toward Commercialization of Perovskite Light-Emitter

Beyond the laboratory-scale development of PeLEDs using spin-coating, scaling up to large-area and mass-production processes remains a key challenge for commercialization. Solution-processable techniques, such as blade coating, inkjet printing, and vacuum thermal evaporation, have been explored as promising fabrication strategies. Z. Xiao and co-workers (adma.202410154) reviewed these methods. In addition, color-conversion applications offer a viable route for early commercialization. J. Tang, J. Luo, Y. Duan, and co-workers (adma.202410194) reviewed the application of PNCs as color-converting materials in displays, highlighting advances in ligand design, patterning techniques, and encapsulation strategies to improve long-term stability and integration with mini-/micro-LEDs. To further improve the stability and scalability of perovskite, H. Zhong and co-workers (adma.202412276) introduced in-situ fabrication of perovskites directly within organic/inorganic matrices or porous hosts. Additionally, L. Quan and co-workers (adma.202406274) reported the development of water-stable "perovskitoid" crystals, facilitating the application of perovskite emitters in ambient environments.

## 9. Emerging Optical Phenomena and Applications of Perovskites

In addition to their conventional application for display as LED, perovskite materials exhibit several unique optical phenomena, including ASE, superfluorescence, and single-photon emission. These properties significantly broaden the scope of perovskite applications, extending into laser, optical communication, and quantum optics. T. C. Sum and M. Feng (adma.202413836) summarized the photophysics of perovskites, particularly their potential in laser and quantum applications. Q. Xiong, Q. Zhang, and colleagues (adma.202413559) further provided a comprehensive review of perovskite laser, categorizing various laser architectures and discussing material engineering strategies to enhance lasing performance. A. Petrozza, H. Wang, and co-workers (adma.202407652) offered new insights into modulating perovskite materials for more efficient lasers. Additionally, H.-L. Yip and colleagues (adma.202414745) highlighted the promising applications of perovskites in visible light communication systems, emphasizing their potential for high-speed data transmission.

## 10. Summary and Outlook

Research on PeLEDs has now entered a stage of maturity, and significant progress has been achieved. In particular, the development of nanocrystalline perovskites and optimized device architectures has enabled EQEs exceeding 30%. These achievements, demonstrating a wider color gamut than conventional OLEDs and QLEDs, clearly highlight the potential of PeLEDs as a next-generation display technology that satisfies the Rec. 2020 color standard.

However, several challenges remain. Operational stability is currently the most critical obstacle to commercialization. Most PeLEDs still do not meet the operational lifetimes required for practical applications. The issue is further critical in red and blue emission devices, where stability is particularly limited. Maintaining high EQE at high brightness, commonly referred to as low-efficiency roll-off, is essential but has not received sufficient attention in the research community. Achieving high brightness at low voltage and with high efficiency is especially important for realizing augmented reality (AR) and virtual reality (VR) displays that enable a fully immersive and interactive user experience.

In parallel, there is growing industrial interest in using perovskite materials as color conversion layers or color enhancement films. These approaches offer a promising route to commercialization due to their compatibility with existing backlight technologies such as LEDs, mini LEDs, and micro (O)LEDs. However, these applications demand even higher reliability, as the perovskite layers are continuously exposed to intense illumination and elevated thermal and humidity stress. Enhancing stability under ambient conditions is therefore increasingly critical. At this point, it is important to think about how to achieve further progress in the field. The focus should shift from achieving peak performance before degradation to ensuring long-term stability and reliability in harsh environments. Additionally, high-resolution patterning is becoming increasingly important, especially for applications in AR/VR. Pixel resolutions above 3000 PPI are necessary to prevent pixelation and enhance visual realism. To achieve this, it is crucial to attain high optical density with a thickness below 2 μm. At the same time, maintaining high color conversion efficiency and stability is also essential.

Through this special issue, we aim to highlight the major advances in PeLED research over the past decade and to offer perspectives on key directions for future development. We hope this collection provides a useful basis for identifying practical strategies to address current limitations and accelerate the transition of PeLED technologies toward real-world applications.

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## Conflict of Interest

The authors declare no conflict of interest.

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