



Cite as

Nano-Micro Lett.

(2026) 18:183

Received: 21 August 2025

Accepted: 6 November 2025

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Scalable Manufacturing and Precise Patterning of Perovskites for Light-Emitting Diodes

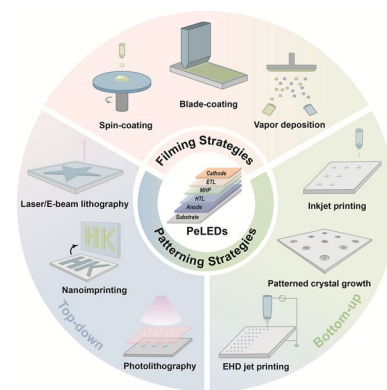
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HIGHLIGHTS

- This review provides a comprehensive exploration of advanced film and patterning fabrication techniques for high-performance perovskite light-emitting diodes (PeLEDs).
- This review examines both top-down and bottom-up techniques, such as photolithography and inkjet printing to achieve precise patterning of PeLEDs for full-color displays.
- This review discusses critical challenges, including device stability, scalable manufacturing, and microscale pixel patterning, as well as promising strategies to overcome these obstacles for the commercialization of PeLEDs.

ABSTRACT Owing to the exceptional optoelectronic properties, metal halide perovskites have emerged as leading semiconductor materials for next-generation display technologies, providing perovskite light-emitting diodes (PeLEDs) great potential for high-quality color displays with a wide color gamut and pure color emission. Although laboratory-scale PeLEDs have achieved near-theoretical efficiencies, challenges such as achieving uniform large-area films, improving material stability, and enhancing patterning precision remain barriers to commercialization. This review presents a systematic analysis of scalable manufacturing and precision patterning strategies for PeLEDs, focusing on their applications in large-area lighting and full-color displays. Fabrication methods are categorized into film deposition techniques (spin-coating, blade-coating, and thermal evaporation) and patterning strategies, including top-down (photolithography, laser/e-beam lithography, and nanoimprinting) and bottom-up (patterned crystal growth, inkjet printing, and electrohydrodynamic jet printing) approaches. In this review, we discuss the advantages and limitations of each strategy, highlight current challenges, and outlook possible pathways towards scalable, high-performance PeLEDs for advanced optoelectronic applications.

KEYWORDS Perovskite materials; Scalable manufacturing; Precise patterning; Light-emitting diodes



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Published online: 05 January 2026



SHANGHAI JIAO TONG UNIVERSITY PRESS

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1 Introduction

Metal halide perovskites (MHPs) are a class of emerging semiconductor materials with exceptional potential for next-generation optoelectronic technologies. The typical structure of MHPs follows the ABX_3 formula, where A represents monovalent cations (such as Cs^+ , Rb^+ , methylammonium (MA^+), and formamidinium (FA^+), B represents divalent metal cations (such as Sn^{2+} and Pb^{2+}), and X denotes halide anions (such as Cl^- , Br^- , and I^-) [1–6]. MHPs exhibit unique optoelectronic properties, primarily including high absorption coefficients, broad bandgap tunability, long carrier diffusion lengths, and excellent defect tolerance [7–9], which make them highly promising for a variety of applications, especially in the field of solar cells [10–15]. Over just a decade, perovskite solar cells (PSCs) have achieved impressive power conversion efficiencies (PCE) of 27.3% (perovskite single junction) [16], 33.89% (perovskite/silicon) [17], 29.1% (all perovskite tandem) [18], marking a significant breakthrough and driving forward the research and commercialization of MHP materials in the photovoltaic industry.

Perovskite light-emitting diodes (PeLEDs), a promising next-generation display technology, have also attracted widespread attention as the photovoltaic application. Compared to traditional liquid crystal displays (LCDs) and organic light-emitting diodes (OLEDs), PeLEDs offer numerous advantages, including narrower emission linewidths (15–20 nm), wide color gamuts, and high contrast ratios [5, 19–21]. These features enable PeLEDs to reach up to 140% of the national television system committee (NTSC) color standard, making them highly promising for applications in displays [3, 21, 22]. Moreover, the diverse chemical compositions of MHPs allow their optical properties to be highly tunable, enabling continuous spectral tuning from the blue-violet to near-infrared regions, further enhancing their appeal in optoelectronic applications [2, 3, 23, 24]. In 2014, Tan et al. [25] successfully demonstrated the first room-temperature green and near-infrared emitting PeLEDs, opening a new chapter in LED research. Since then, the performance of PeLEDs has rapidly improved, with external quantum efficiencies (EQE) for green, red, and near-infrared emitting devices exceeding 25%, and the EQE of blue PeLEDs surpassing 20% [26–31]. These breakthroughs have positioned PeLEDs on par with the most advanced organic and quantum dot LEDs. Beyond display technologies, PeLEDs

hold significant promise for flexible and wearable electronics due to the inherent mechanical flexibility of perovskite materials [32, 33]. Flexible PeLEDs could enable innovations such as foldable displays [34–36] and biocompatible imaging devices [21, 32, 37].

Despite the remarkable progress in PeLED technology, several challenges remain before PeLEDs can be widely adopted in consumer products. First, translating high-performance perovskite films from small laboratory devices to large-area substrates introduces significant non-uniformities in thickness, crystallinity, and defect density, which degrade device efficiency and shorten operational lifetime. Ensuring reproducible film quality across such scales is therefore critical for achieving consistent, industry-scale performance of PeLEDs [38–41]. Second, the development of high-resolution patterning techniques for full-color displays adds further complexity. Achieving precise pixel control down to tens of micrometers while maintaining the optoelectronic properties of the perovskite material is critical for the successful integration of PeLEDs into practical applications [42, 43]. Moreover, integrating pure red, green, and blue (RGB) emitting units into subpixels with precise control of color and pattern remains a critical challenge for commercialization, since full-color displays with high resolution and wide color gamut are essential for consumer electronics such as smartphones and virtual reality (VR) devices [21, 34]. This requires microscale alignment of subpixels and suppression of cross-talk between adjacent emission layers, both of which are hindered by material instability and fabrication complexity [43]. Together, these dual requirements of wide-area uniformity and precise pixel-level patterning define the engineering benchmark for scalable, high-resolution PeLED manufacturing. Moreover, other key issues including long-term operational stability, light outcoupling efficiency, and resistance to thermal and environmental degradation, also need to be addressed to extend the applicability of PeLEDs [44–46].

In this review, we aim to provide a comprehensive overview of representative fabrication strategies of perovskite materials, with a particular focus on large-area film fabrication and high-resolution patterning strategies in light-emitting diodes. According to the target morphology and processing requirements of MHP, we classify the fabrication techniques for perovskite into two main sections: film fabrication and patterning techniques. The first section covers three typical film fabrication methods, including

spin-coating, blade-coating, and thermal evaporation. In the second section, we focus on patterning techniques, which are further categorized into two main approaches based on their processing mechanisms. Top-down methods include photolithography, laser/e-beam lithography, and nanoimprinting, whereas bottom-up methods involve patterned crystal growth, inkjet printing, and electrohydrodynamic (EHD) jet printing. The principles, advantages, and drawbacks of each fabrication method are analyzed, with specific emphasis on their applications in full-color displays. Finally, we discuss future directions for large-area fabrication and patterning technologies, highlighting their roles in advancing high-performance PeLEDs toward industry-level commercialization.

2 Film Fabrication Strategies

Deposition of high-quality and scalable MHP emission layers is critical for PeLED manufacturing. Figure 1a, b compares three main deposition strategies: spin-coating, blade-coating, and thermal evaporation, for small- and large-area PeLEDs. In this work, we define small-area PeLEDs as devices with active area $< 10 \text{ mm}^2$ and large-area PeLEDs as those with active area $\geq 100 \text{ mm}^2$. This empirical partition follows the distribution of published device areas (Fig. 1c), where most reports cluster below $\sim 10 \text{ mm}^2$ (laboratory studies) or above $\sim 100 \text{ mm}^2$ (scale-up demonstrations), while the intermediate range ($\sim 10\text{--}100 \text{ mm}^2$) remains comparatively underexplored. The scarcity of studies in the $10\text{--}100 \text{ mm}^2$ window is mainly due to (i) the incompatibility of many lab-scale deposition/patterning methods with intermediate-area substrates without substantial re-engineering, (ii) scale-dependent drying and flow dynamics that change nucleation/uniformity when moving away from small pixels, and (iii) practical applications focus either to small pixel-scale demonstrations or to large-area proof-of-concept panels [44]. Spin-coating is highly effective for small-area films, offering excellent quality, controllability, and low cost. This is attributed to rapid and uniform solvent evaporation driven by centrifugal forces, which ensures controlled nucleation and homogeneous crystallization. However, when extended to large areas, spin-coating faces challenges in scalability and uniformity, resulting in less consistent film quality and thickness, due to uneven solvent evaporation and centrifugal force distribution at larger radii. Blade-coating, by contrast,

is cost-effective for large-area applications, providing good scalability and controllability. Its effectiveness at large scale arises from continuous blade movement that facilitates controlled film spreading. But variations in solvent evaporation rates and local concentration gradients during coating lead to less uniform nucleation and crystallization, thus sacrificing crystallinity and uniformity over small area compared to spin-coating. Thermal evaporation stands out for its superior scalability and film uniformity, making it ideal for industrial-scale production. The high uniformity originates from the precisely controlled vapor-phase deposition process under vacuum conditions, ensuring consistent molecular flux and film thickness distribution. Nevertheless, its low material utilization and the requirement for vacuum environments increase complexity and costs.

We further compare three methods in EQE, device area, and half-operation lifetime ($@ 100 \text{ cd m}^{-2}$) aspects in Fig. 1c, d and summarized key metrics of large-area PeLEDs in Table 1. Spin-coating, the most commonly employed technique for perovskite thin-film preparation, achieves remarkably high EQE and long lifetime due to its advanced defect passivation strategies [66–68]. However, its scalability to larger areas is limited, reducing its practicality for industrial-scale manufacturing. Blade-coating offers a more balanced approach, enabling cost-effective deposition over medium-to-large areas. While its EQE and operational lifetime are typically lower than those of spin-coated small-area devices, which mainly reasoned by reduced uniformity and crystallinity, its superior scalability makes it more suitable for larger-scale applications [69, 70]. In contrast, thermal evaporation excels in large-area perovskite film fabrication, offering outstanding scalability and precise thickness control. However, devices produced by this method typically exhibit relatively low EQEs (mostly below 10%). This limited performance primarily arises from the inconsistent deposition temperatures of bromide precursors, which lead to a high density of defects. Additionally, the inherently weak surface interactions during vapor-phase deposition often result in sparse nucleation and rough film morphologies, further diminishing optoelectronic performance [40]. Overall, spin-coating is best suited for high-performance, small-area devices, while thermal evaporation is advantageous for large-scale production, and blade-coating provides a practical middle ground between performance and scalability.



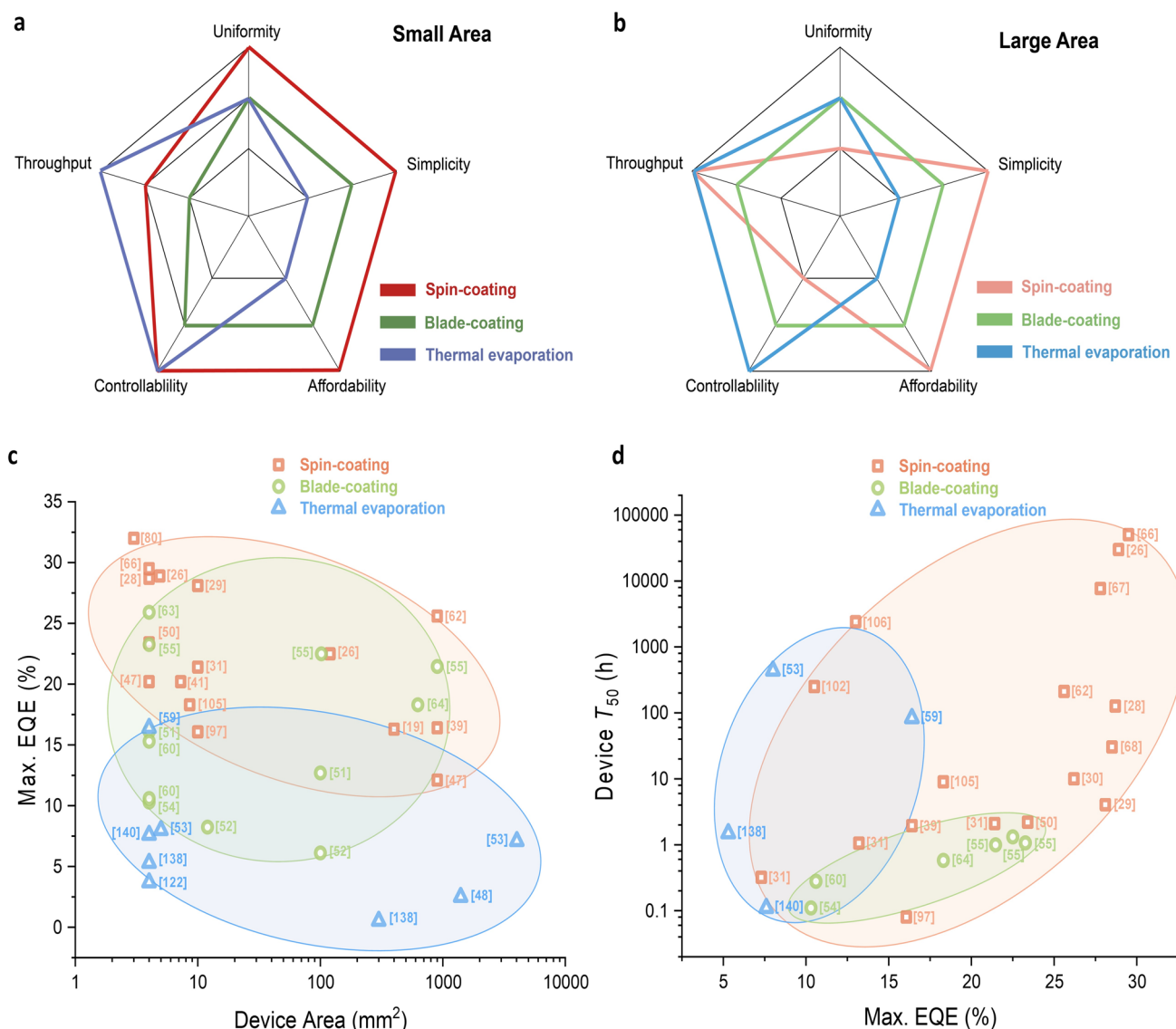


Fig. 1 Perovskite filming strategies. **a** Radar plots comparing different perovskite film deposition techniques for small-area devices ($< 10 \text{ mm}^2$). **b** Radar plots comparing different perovskite film deposition techniques for large-area devices ($\geq 100 \text{ mm}^2$). “Affordability” refers to the cost-effectiveness of fabrication strategy. “Controllability” refers to the ability to achieve the desired thickness, area, and quality of perovskite films. **c** Statistical scatter plot showing the maximum EQE versus device area for PeLEDs studies conducted over the past 5 years. **d** Statistical scatter plot showing the device half-operation lifetime ($@ 100 \text{ cd m}^{-2}$) versus maximum EQE for PeLEDs studies conducted over the past 5 years

2.1 Spin-coating

Spin-coating is a widely used technique where a small volume of solution is deposited onto a substrate fixed on a rotating stage. It spreads the solution radially onto the surface of a rotating substrate through centrifugal force (proportional to the angular velocity and plate radius), and its uniformity depends on the dynamic balance between the viscosity gradient caused by solvent evaporation and the distribution

of centrifugal force. Although the centrifugal force at the edge is greater, rapid solvent evaporation can suppress the radial thickness difference and form a uniform gel-like layer. By adjusting the rotational speed (controlling centrifugal force), time (affecting evaporation rate), and antisolvent addition (promoting uniform nucleation), the morphology, crystallinity, and thickness uniformity of the film can be optimized [71–75]. Beyond solvent/antisolvent control, spin-coated PeLED films benefit markedly from rational

additive engineering: (i) Lewis-base/ligand additives (e.g., H₂O microadditives, phosphine-oxide, or amide donors) coordinate Pb²⁺ and stabilize solvate intermediates, slowing crystallization to yield denser, more uniform films over centimeter scales [76, 77]; (ii) ionic/bulky A-site additives (e.g., PEACl, MACl, FABr; pseudohalides) regulate halide homogenization and promote vertically oriented quasi-2D/3D stacks, suppressing phase segregation and non-radiative traps [78, 79]. Synergistic dual-additive strategies can simultaneously accelerate radiative recombination and passivate defects—particularly valuable for red/near-infrared (NIR) compositions where exciton–phonon coupling is stronger and spectral broadening is more pronounced [80, 81]. At the interfaces, self-assembled monolayers (SAMs) and ultrathin passivation layers provide complementary levers for scalable uniformity: (i) SAMs (e.g., PACz-type or

co-adsorbed ionic SAMs) tune surface energy and dipoles to improve wetting, increase nucleation density, and reduce interfacial trap-assisted quenching; (ii) SAM-assisted blue PeLEDs, for instance, show order-of-magnitude EQE gains by suppressing non-radiative loss at the perovskite/hole transport layer (HTL) interface [82]. Through multiple optimization strategies, spin-coating is particularly effective for producing MHP films with uniform structures and high crystallinity at small scales, making it a standard approach for fabricating PeLEDs at the laboratory [5, 30, 50, 83–85]. However, it is limited by edge flow effects and radial thickness gradients, making it difficult to directly extend to large-scale applications [69].

The electroluminescent (EL) linewidth of PeLEDs remains distinctly narrow in the visible yet broadens toward the near-infrared, with trends that follow both intrinsic

Table 1 The summary of main characteristics of large-area ($\geq 100 \text{ mm}^2$) PeLEDs over the past five years

Year	Strategy	Emission layer (wavelength)	Maximum device area (mm^2)	Maximum EQE (% , @maximum area)	T_{50} (h, @maximum area, 100 cd m^{-2})	Refs
2020	Spin-coating	FAPbI ₃ (799 nm)	900	12.1	10 (@10 mA cm^{-2})	[47]
2020	Spin-coating	FAPbBr ₃ (531 nm)	400	16.3	2.9 (T_{80})	[19]
2020	Thermal evaporation	CsPbBr ₃ (520 nm)	1400	~2	7 (@500 cd m^{-2})	[48]
2021	Spin-coating	PEA ₂ (FA _{0.7} Cs _{0.3}) ₂ Pb ₃ Br ₁₀ (520 nm)	900	16.4	1.95	[39]
2021	Spin-coating	CsPb(Br/Cl) ₃ (490 nm)	400	6.1	0.16	[49]
2021	Spin-coating	FA _{0.9} GA _{0.1} PbBr ₃ PeNCs (535 nm)	900	-	-	[50]
2021	Blade-coating	MAPbI ₃ (725 nm)	2800	-	10.1 (@3 mA cm^{-2})	[51]
2021	Blade-coating	(PEA) ₂ Cs _{n-1} Pb _n Br _{3n+1} (530 nm)	1225	-	-	[52]
2021	Thermal evaporation	CsPbBr ₃ (508 nm)	4020	7.1	7.3	[53]
2022	Spin-coating	(FA _{0.7} MA _{0.1} GA _{0.2}) _{0.87} Cs _{0.13} PbBr ₃ (540 nm)	120	22.5	-	[26]
2022	Blade-coating	CsPb(Br _{0.84} Cl _{0.16}) ₃ (489 nm)	2800	-	-	[54]
2022	Blade-coating	FA _x GA _{1-x} PbBr ₃ (530 nm)	900	21.46	1	[55]
2022	Thermal evaporation	MAPbBr ₃ (517 nm)	7850	-	-	[56]
2023	Spin-coating	CsPbBr _{3-x} Cl _x /CsPbBr ₃ /CsPbBr _{3-x} I _x (478/512/630 nm)	1050	-	-	[57]
2023	Spin-coating	(PEA) ₂ Cs _{n-1} Pb _n Br _{3n+1} (516 nm)	1225	12.1	-	[58]
2023	Thermal evaporation	CsPbBr ₃ (523 nm)	7995	-	-	[59]
2024	Spin-coating	CsPbI ₃ (645 nm)	900	28.7	126.8	[28]
2024	Blade-coating	CsPbI ₃ (650 nm)	2800	-	-	[60]
2025	Spin-coating	CsPbBr _{2.5} Cl _{0.5} (508 nm)	400	2.81	2.25	[61]
2025	Spin-coating	CsPbI ₃ (630 nm)	900	25.6	211	[62]
2025	Blade-coating	(PEA,Cs)PbBr ₃ (495 nm)	3500	-	-	[63]
2025	Blade-coating	FAPbI ₃ (780 nm)	625	18.3	0.58	[64]
2025	Thermal evaporation	FA _x Cs _{1-x} PbI _y Br _{3-y} (675 nm)	2500	-	-	[65]

(exciton–phonon) and extrinsic (compositional/structural) broadening mechanisms [86, 87]. State-of-the-art green PeLEDs based on bromide 3D/NC films routinely show full width at half maximum (FWHM) ~ 18 – 20 nm, while blue/deep-blue emitters (typically Br/Cl mixed or low-dimensional phases) are usually ~ 20 – 30 nm, which suffer color drift and line broadening from field-accelerated halide segregation and phase redistribution unless halide homogenization and defect passivation are enforced [44, 67, 88–91]. Red devices (iodide-rich) commonly exhibit ~ 28 – 35 nm, though exceptional systems (e.g., highly passivated α -CsPbI₃) can approach ~ 21 – 28 nm [92]. By contrast, NIR PeLEDs generally display broader lines ~ 40 – 50 nm (representative reports ~ 41 – 46 nm) owing to stronger exciton–phonon coupling at lower bandgaps and greater inhomogeneity from grain/strain distributions [93, 94].

In 2014, Tan et al. [25] demonstrated the first room-temperature PeLED device fabricated using the spin-coating method. The perovskite layer in their devices was designed to be extremely thin (~ 15 nm) to spatially confine electrons and holes, thereby enhancing radiative recombination efficiency. Despite achieving a modest EQE of only 0.1%, the low cost and versatility of this approach sparked significant research interest for further exploration. A subsequent breakthrough was achieved by Cho et al., who improved both the EQE and current efficiency through stoichiometric adjustments and a nanocrystal pinning (NCP) process (Fig. 2a) [95]. To further reduce the grain size, they incorporated an organic molecule of 2,2',2''-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBI) as an additive in the NCP solvent. By using a self-organized conducting polymer (SOCP) as the anode, they successfully fabricated a flexible large-area PeLED device with a 20 mm by 20 mm pixel for the first time (Fig. 2b, c). This method facilitated the in situ formation of MAPbBr₃ nanograins, which confined excitons within the nanograins, thereby suppressing exciton quenching. As a result, their green PeLEDs achieved a maximum EQE value of 8.53%.

To enhance LED performance, two strategies subsequently emerged: direct spin-coating of colloidal perovskite nanocrystals (PeNCs) [98, 99] and engineering perovskite precursor solutions for bulk films [83, 100, 101]. The first approach utilized the high photoluminescence quantum yield (PLQY) of PeNCs, along with their tunable optical properties achieved through compositional adjustments and control of crystal size. In contrast, the second strategy

focuses on depositing bulk perovskite films by optimizing the composition of precursor solutions, allowing for precise tailoring of the material properties to enhance film quality and device performance. Beyond these two approaches, a significant advancement was reported by Lin et al. in 2018, who achieved an impressive EQE of 20.3% using a CsPbBr₃/MABr quasi-core/shell structure [96]. The MABr shell could passivate the non-radiative defects that often present in CsPbBr₃ and enhance the charge injection balance. This quasi-core/shell structure, formed by blending pre-synthesized CsPbBr₃ perovskite powder with an MABr additive (Fig. 2d), demonstrated great benefits for crystallinity improvement and defect passivation of the emitting layer, making it a promising approach for enhancing the performance and longevity of PeLEDs. The cross-section TEM image (Fig. 2e) evidenced well-aligned layer-by-layer structure and the boundaries formed by MABr. Another remarkable milestone was reached by Kim et al., who combined a three-dimensional (3D) perovskite film formed via an additive-based nanocrystal pinning (A-NCP) technique with in situ formation of a core/shell structure using benzylphosphonic acid (BPA) solution combined with tetrahydrofuran (THF) post-treatment [26]. Specifically, the in situ particle structure was created by forming covalent bonds between the surficial undercoordinated Pb²⁺ of the 3D perovskite matrixes with BPA additives. The strong acidity and small size of the BPA additive enable the penetration and intercalation of BPA molecules into larger perovskite crystals, leading to the cleavage of the large crystal domains and formation of a nanosized core/shell structure. This was achieved by exposing the particle structure to a BPA/THF solution, which reduced the large crystals into nanosized cores and shells surrounded by BPA (Fig. 2f). As a result, the grain size distribution of the perovskite structure was significantly minimized, decreasing from 205 ± 97 nm (for the 3D structure) to 10 ± 2 nm (for the in situ core/shell). Ultimately, this innovative approach yielded a maximum EQE of 28.9% and a peak current efficiency of 151 cd A^{-1} , while maintaining over 20% efficiency under ultra-high brightness conditions exceeding $400,000 \text{ cd m}^{-2}$. Furthermore, leveraging the core/shell structure, they successfully fabricated a 120 mm^2 PeLED that exhibited high brightness, excellent uniformity, and a maximum EQE of 22.5% (Fig. 2g).

Compared with green PeLEDs, although several works achieved great improvement in EQE of red PeLEDs, it is still challenging to realize pure-red (620–650 nm) LEDs with

good stability [102–106]. To enhance the performance of PeLEDs in the red spectrum, Kong et al. [28] developed a method to stabilize the I–Pb–I octahedron by introducing 3-methoxyphenethylammonium (MOPA) into reduced-dimensional perovskite structures. Unlike conventional ammonium ligands, which provide weak hydrogen bonding to anchor the lattice, MOPA features a double-ended anchored ligand that includes both ammonium and methoxy groups. This structure enhances hydrogen bonding interactions and effectively coordinates lead ions, thereby suppressing I^- migration under an electric field and reducing non-radiative recombination. These enhancements led to the ultralong operation lifetime (7610 min @ 100 cd m⁻²) for the fabricated PeLEDs with a maximum EQE of 28.7%, achieving bright and uniform pure red emission at 638 nm over an active area of 30 mm × 30 mm (Fig. 2h).

Despite significant advancements in the performance of green- and red-emitting PeLEDs using bromine- and iodine-based perovskites, achieving efficient and stable PeLEDs with blue emission (between 450 and 490 nm) using chlorine-based materials remains challenging due to poor charge injection, high amounts of non-radiative recombination, and random phase distribution [49, 107–109]. Addressing the issue of inefficient charge injection, Chu et al. incorporated CsCl into the hole injection layer (HIL), poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), resulting in the improving of charge-carrier transport and better band alignment with the perovskite emissive layers (Fig. 2i). Based on such strategy, fabricated sky-blue PeLEDs centered at 486 nm achieved an EQE of 16.07%. Moreover, Yuan et al. recently developed efficient blue-emitting LEDs using a reduced-dimensional mixed-halide perovskite with the additive bis(triphenylphosphine)iminium chloride (PPNCl), which improves phase tuning toward highly emissive quasi-3D structures and reduces defects by forming bonds with organic and lead components during crystallization. The resulting films achieve over 88% PLQY, enabling blue PeLEDs with a maximum EQE of 21.4%, 13.2%, and 7.3 peaking at 483, 474, and 464 nm, respectively (Fig. 2j). This work offers a promising approach for fabricating next-generation high-performance blue PeLEDs.

However, large-area PeLEDs fabricated using the spin-coating method typically exhibit degraded EQEs, brightness, current efficiency, and uniformity compared to their

small-area counterparts [19]. This performance shortfall is primarily due to the decreased uniformity in film flatness and the uncontrollable crystallization process that leads to inhomogeneous diffusion and crystallization between the edge and center of the film. Although the increased defect density and reduced homogeneity pose challenges for scaling spin-coating techniques to larger PeLED devices, it remains a valuable approach for developing perovskite precursors and additive decorations for large-area films.

Without the need for further purification or ligand-exchange steps, spin-coating can yield high-quality quasi-2D films, which have been successful in small-area PeLEDs thanks to their self-assembled multiple-quantum-well structure [39]. However, the simple one-step spin-coating deposition strategy, even when enhanced by “antisolvent-assisted” techniques—shown to be highly effective for fabricating emitting quasi-2D films—faces challenges in producing large-area homogeneous films. Investigating the underlying issues, Sun et al. discovered that the antisolvent does not diffuse uniformly to the edge regions, where bulk 3D and layered 2D phases dominate. To address this, they introduced L-norvaline (NVAL) into the precursor solution to replace the PEA⁺ ligand, creating a COO⁻-coordinated intermediate phase with lower formation enthalpy. This approach enabled the production of a large-area (2500 mm²) quasi-2D film with high homogeneity, bright emission, and high PLQY. Consequently, the large-area green PeLED achieved a maximum EQE of 16.4% with an active device area of up to 900 mm².

Rather than tackling the non-uniformity of large-area PeLED devices, Zhao et al. [47] concentrated on enhancing hole injection efficiency, a significant factor affecting performance. By incorporating 5-aminovaleric acid (5-AVA) as an additive, their fabricated PeLEDs exhibited passivated traps and improved microstructure. Additionally, they replaced the hole transport layer with poly-TPD (poly(N,N'-bis(4-butylphenyl)-N,N'-bisphenylbenzidine)), which has a shallow ionization potential. This strategy led to high-efficiency small-area PeLEDs measuring 4 mm², achieving an EQE of 20.2% with a narrow standard deviation, as well as a large-area PeLED of 900 mm² that demonstrated outstanding brightness, uniformity across the entire device, and an EQE of 12.1% (Fig. 2k).



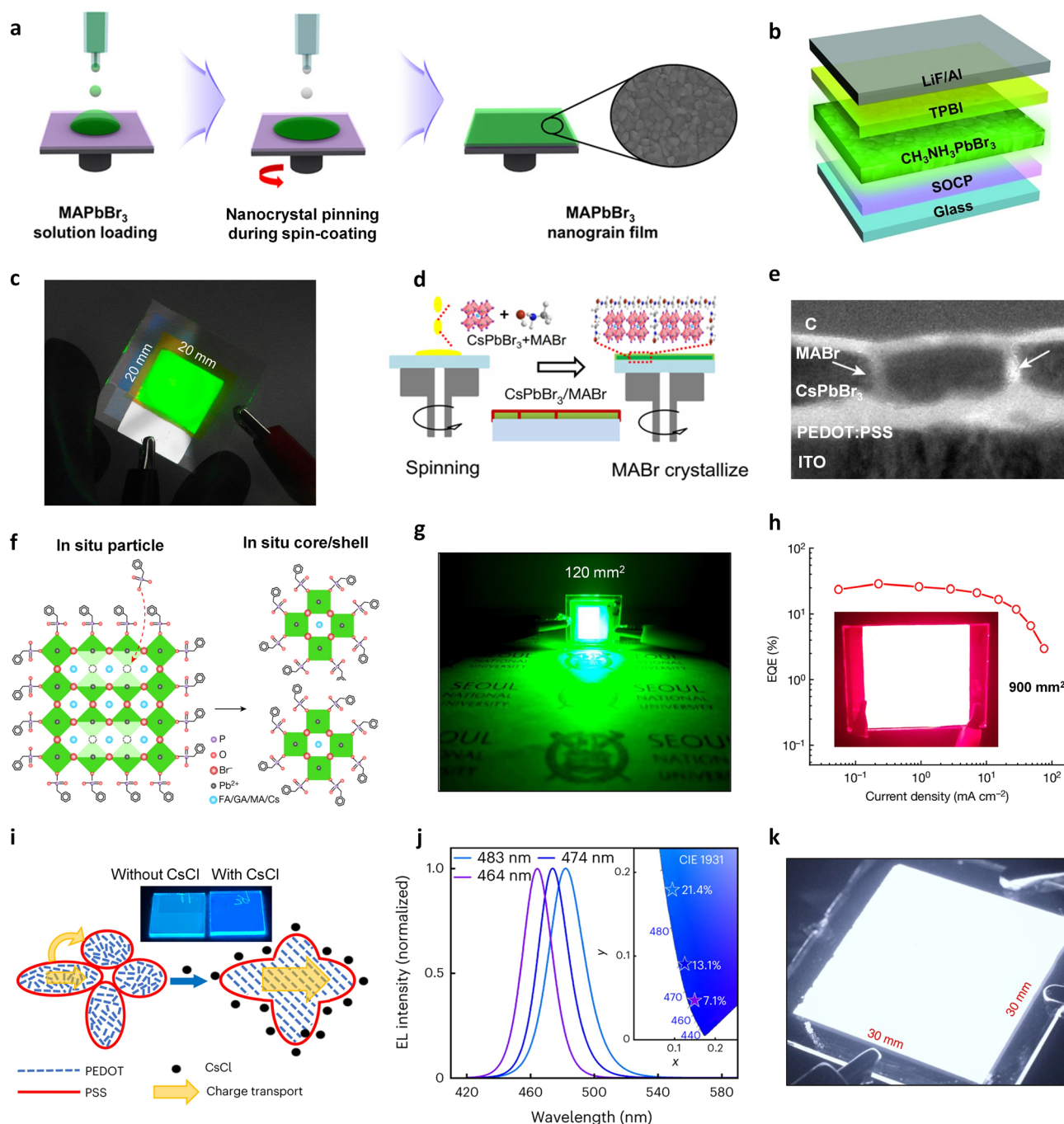


Fig. 2 Spin-coating strategy for PeLEDs. **a** Schematics of NCP process during spin-coating to fabricate MAPbBr₃ nanograin films, **b** the corresponding device structure, and **c** a photograph of large-area (20 mm by 20 mm pixel) PeLEDs [95]. **d** Schematic formation process of the CsPbBr₃/MABr quasi-core/shell structure. **e** Cross-sectional TEM image of the quasi-core/shell CsPbBr₃/MABr structure. White arrows indicate the MABr shell (the grain boundary) [96]. **f** Schematic illustration of the transformation process of in situ particle into in situ core/shell structures by BPA treatment. **g** Photograph of an operating large-area device (pixel size: 120 mm²) [26]. **h** EQE versus current density curve of PeLEDs based on MM-MOPA perovskite; the inset is an optical photograph of a 900 mm² PeLED under 4 V voltage [28]. **i** Proposed schematic of the structural evolution of PEDOT:PSS after CsCl incorporation. The inset shows the emission images under ultraviolet excitation (365 nm) [97]. **j** EL spectra of PPNCI-containing PeLEDs with different Cl⁻/Br⁻ ratios (driven by 4.5 V). The inset shows the corresponding CIE coordinates [31]. **k** NIR image of the 900 mm² large-area PeLED on glass [47]

2.2 Blade-coating

Compared to spin-coating, which is restricted by limited substrate size, blade-coating provides a scalable method for producing large-area perovskite films and optoelectronic devices like PSCs and PeLEDs [69, 110–115]. The coating process involves depositing the precursor solution uniformly across the surface of substrate by controlling the movement of blade or substrate (Fig. 3a) [114]. Fundamentally, this process is governed by shear-driven fluid dynamics, where the blade-induced shear force aligns precursor molecules and suppresses uncontrolled convection, enabling homogeneous solution spreading [114]. The quality and thickness of the formed films depend not only on the setup parameters, including travel speed, placement angle, and gap distance, but also on the viscosity of the precursor solution, the wettability of the substrate, and the adhesion interface between them [111]. The balance between viscous forces and capillary flow dictates the film uniformity, while solvent evaporation kinetics during the sol–gel transition critically influence crystallization pathways and defect formation [115]. Thanks to its rapid and efficient formation of uniform thin films over large surface area, low equipment cost, high compatibility, and high material utilization rate, blade-coating is generally considered as one of the most favorable strategies for the large-area PeLEDs fabrication [52].

In 2016, Bade et al. [116] pioneered the use of the blade-coating method for fabricating PeLEDs. However, due to a lack of control over crystallization kinetics, the resulting perovskite films exhibited micrometer-scale grain sizes and excessive thickness (1–2 μm), leading to imbalanced charge carrier transport, enhanced non-radiative recombination, and poor light outcoupling efficiency. Consequently, the devices achieved an EQE of only 1.1%. Unlike PSCs, which benefit from large grains and thick films, PeLED active layers require small grain sizes and ultrathin films to realize charge carrier confinement and enable efficient charge injections [117, 118]. Blade-coated MHP films also retain more residual solvent compared to spin-coated films, leading to slow and uneven solvent evaporation during the sol–gel stage. This often results in high surface roughness and inhomogeneous crystallization. To accelerate solvent evaporation, the substrate is usually heated during the blade-coating process [112, 119]. However, elevated substrate temperatures also induce rapid solute migration and aggregation, complicating the formation of uniform and smooth films [69].

Consequently, the blade-coating method faces considerable challenges in achieving high-performance PeLEDs, primarily due to the high surface roughness and undesirable crystal morphology caused by uncontrolled crystallization during the deposition process.

To address these challenges, researchers have explored innovative strategies to improve solvent evaporation and film morphology during the blade-coating process. Among these, utilizing a gas knife, such as N_2 or other inert gases, offer a significant advantage over the single solid blade in accelerating solvent evaporation. By employing an N_2 knife-assisted low-temperature blade-coating approach, Chu et al. successfully prepared uniform, large-area perovskite films with great uniformity in film thickness and roughness, and improved optoelectronic properties [51]. They modified the blade-coating process by diluting precursor concentration, lowering the substrate temperature down to 50 $^\circ\text{C}$, and incorporating 4-fluorophenylmethylammonium iodide (FPMAl) as an additive to overcome the inherent inhomogeneous crystallization at the sol–gel stages. The diluted precursor with excess organoammonium dramatically reduced the whole sol–gel period, promoting dense nucleation centers, and accelerating phase transformation. This approach significantly decreased the roughness of perovskite films, yielding ultra-flat large-area films with roughness as low as 1 nm (Fig. 3b). The resulting doctor-bladed red-emission PeLEDs achieved a peak EQE of 16.1%. This strategy also enabled the fabrication of an ultra-large-area PeLED (2800 mm^2) with uniform EL emission, demonstrating its compatibility with large-scale manufacturing processes (Fig. 3c). Chu et al. further extended this blade-coating method to the fabrication of large-area PeLEDs with stable sky-blue emission. By partially replacing dimethyl sulfoxide (DMSO) with dimethylformamide (DMF) to obtain a more volatile supersaturated solution, they successfully modulated the nucleation process from gas-solution interface to whole solution phase, enabling much higher nucleation sites, and a faster crystallization rate [54]. The peak EQE of their blade-coated PeLEDs reached 10.3% with a blue emission wavelength of 489 nm, and the large PeLED device also showed a bright and uniform sky-blue emission, demonstrating quite small fluctuations of both radiative and non-radiative recombination rates over the entire film (Fig. 3d, e).

Chen et al. demonstrated the vacuum quenching-assisted blade-coating process at room temperature for the fabrication of large-area green-emission PeLEDs [52]. To avoid the



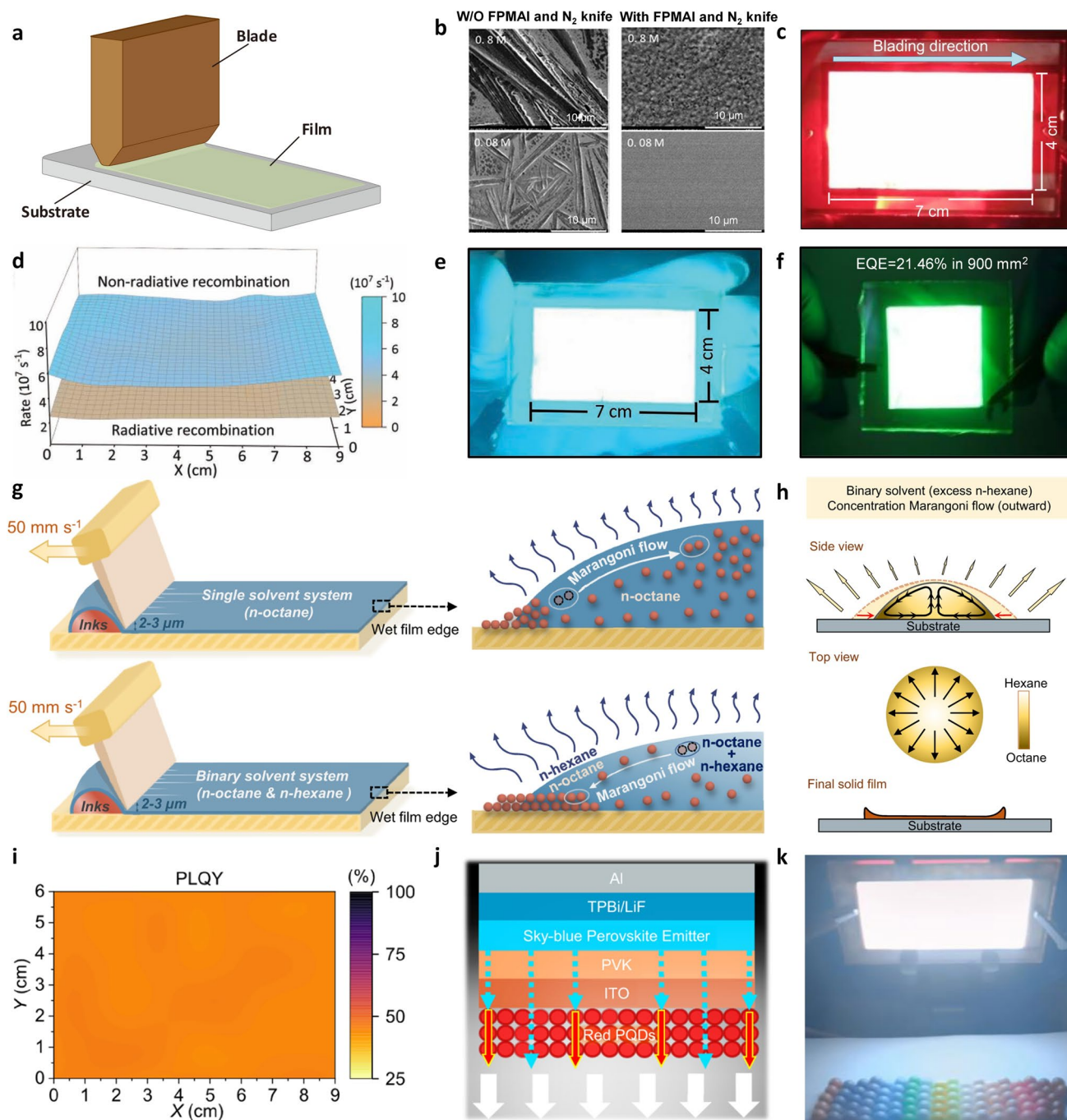


Fig. 3 Blade-coating strategy for perovskite films and PeLEDs. **a** Schematics of the blade-coating setup. **b** SEM images of the doctor-bladed films fabricated with/without FPMAl or an N_2 knife. **c** Photograph image of a red large-area PeLED (40×70 mm²) [51]. **d** Radiative and non-radiative recombination rate mapping of large-area films. **e** Photograph image of a sky-blue large-area PeLED (40×70 mm²) [54]. **f** Photograph image of a m-bar-coated PeNC film-based large-area PeLED (900 mm²) [55]. **g** Schematics of the fabrication process of PeQD films by blade-coating with the single solvent and binary-solvent system. **h** Schematic illustration of solution droplet evaporation and final film patterns with excess ratios of n-hexane. **i** PLQY mapping of a large-area PeQD film (60×90 mm²). **j** Device structure of white PeLEDs. **k** Photograph image of a white large-area PeLED (40×70 mm²) [60]

solution migration and aggregation induced by nonuniform heat-transfer, they creatively put the newly coated precursor wet films into a vacuum chamber to evaporate excess solvent, followed by an annealing post-treatment. The fabricated LED devices achieved high EQEs of 8.24% and 6.12% on emitting areas of 0.12 and 1 mm², respectively. Meanwhile, a large LED device with an emitting area 35 × 35 mm² was also fabricated, which exhibited bright and uniform emission characteristics. As mentioned in the spin-coating section, colloidal PeNCs hold great potential for manufacturing controllable high-quality perovskite films without suffering from rapid crystallization or nonuniform phase distribution. This is because PeNCs are pre-synthesized and surrounded by organic ligands, which shield them from the adverse effects typically associated with the film formation process—such as high-temperature annealing, solvent-induced degradation, and uncontrolled crystallization—that can otherwise deteriorate the material's structural and optoelectronic properties [120]. Besides, with efficient charge carrier confinement and fast exciton recombination, PeNCs films exhibit bright luminescence with narrow widths, making them promising candidates for the fabrication of large-area PeLEDs. Kim et al. exploited the full advantages of colloidal PeNCs for the large-area efficient PeLEDs with improved EL efficiency, high uniformity, and reproducibility [55]. They printed the pre-prepared FAPbBr₃ NCs with a modified bar-coating method in which a tilted substrate by more than 50° was adopted to accelerate the evaporation of resident solvent. The surface of FAPbBr₃ NCs was capped with organic ligands (n-decylamine and oleic acid), which could effectively suppress ion migration and charge trapping during the operation of PeLEDs. By this way, they demonstrated a highly efficient green PeLED with an EQE of 21.46% across the 900 mm² device area, which is comparable to the best efficiency of small-area PeLEDs fabricated by spin-coating (Fig. 3f). Their efforts also indicate that colloidal PeNCs might be more suitable than polycrystalline films for applying printed large-area, high-efficiency PeLEDs for industrial displays and solid-state lighting.

Similar to colloidal PeNCs, premade perovskite quantum dots (PeQDs) can also decouple crystallization from the film formation process, making them a promising material for large-area PeLEDs. However, due to their smaller size (less than 10 nm) and higher surface-to-volume ratio, PeQDs are more ionic and sensitive to polar solvents, posing challenges in their dispersion and deposition [2]. To address this,

Shi et al. [60] developed a blade-coating approach using a binary-solvent system (n-octane and n-hexane) to control the solvent fluidic dynamics and achieve uniform PeQD film deposition. As illustrated in Fig. 3g, the introduction of n-hexane into n-octane solvent prevents the accumulation of PeQDs by inducing an outward Marangoni flow, leading to uniform and dense film formation. Meanwhile, the proper amount of n-hexane is also of great significance; otherwise, faster evaporation strengthens the outward flow and leads to premature drying during spreading with excess n-hexane (Fig. 3h). This method enabled the fabrication of large-area film with uniform PLQY (Fig. 3i) and red-emitting PeLEDs with a peak EQE of 15.3%. To exploit the full advantages of the binary-solvent approach of making uniform perovskite films, they also fabricated a white PeLED (EQE > 10.6%) by combining a sky-blue PeLED with a red PeQD layer as the downconverter (Fig. 3j) and successfully produced a large-area (2800 mm²) white PeLED with excellent brightness and uniformity (Fig. 3k).

2.3 Thermal Evaporation

Thermal evaporation is generally considered as a mature technology, commonly used in electrode coating, semiconductor deposition, and large-scale OLED production [40, 124–126]. In recent years, thermal evaporation technique has attracted great interest for the fabrication of MHPs in PSCs and PeLEDs [127–130]. This strategy is also referred as vapor deposition, which involves the controlled deposition of vaporized precursor materials onto a substrate in a vacuum. As illustrated in Fig. 4a, the vapor deposition process starts with the co-evaporation of two halide precursors (typically an alkali-metal halide and a lead halide) which react on the substrate to form the perovskite [40]. In LED-oriented vacuum deposition, inorganic salts are preferred over organic ammonium halides (e.g., MABr/FABr) because the latter partially decompose under high vacuum at deposition temperatures, complicating flux control and stoichiometry, whereas CsBr/PbBr₂ co-evaporation reproducibly yields dense CsPbBr₃ films [131–133]. Nucleation occurs as vapor condenses, forming small clusters that grow into crystals. The nucleation density and subsequent crystal growth are strongly influenced by parameters such as deposition rate, substrate temperature, and surface energy: slower deposition rates and optimized substrate heating promote



adatom surface diffusion, leading to larger crystal domains and reduced defect density. Post-deposition annealing often further improves crystallinity by facilitating atom rearrangement and grain coalescence [82].

However, the thermal evaporation process also introduces several challenges. In vacuum co-evaporation of CsPbBr_3 from CsBr and PbBr_2 , the two precursors have markedly different vapor pressures/sublimation temperatures (PbBr_2 reaches useful vapor pressures in the $\sim 340\text{--}360\text{ }^\circ\text{C}$ range, whereas CsBr generally requires $\sim 600\text{--}700\text{ }^\circ\text{C}$), so even small drifts in source temperature or crucible loading can bias the Cs/Pb flux ratio, leading to non-stoichiometry [134, 135]. These regions often form deep-level defects that act as non-radiative recombination centers, increasing trap-assisted recombination and reducing PLQY [40]. In addition, because adatom–substrate interactions for halide perovskites in vacuum are relatively weak, adatom diffusion lengths are large and nucleation densities can be sparse, yielding coarser grains and higher RMS roughness than the rapid, supersaturation-driven nucleation typical of antisolvent solution routes. This difference in the early growth regime explains why vacuum films often require additional measures to reach the morphology and defect levels routinely achieved by solution processing [136, 137]. To mitigate these issues, approaches include: closed-loop flux control with $\text{PbBr}_2/\text{CsBr}$ ratio tuning, substrate-temperature windows that promote reaction yet avoid decomposition, post-annealing to complete solid-state conversion and heal defects, hybrid thermal evaporation assists to strengthen interfacial reaction/wetting, and nucleation-seeding interlayers to raise nucleus density [65, 128]. Despite these limitations, thermal evaporation offers excellent scalability and precise thickness control due to its directional, collision-free vapor transport in vacuum, which minimizes scattering and contamination [128]. The ability to form well-defined films with tunable composition makes it a compelling choice for large-area manufacturing, provided that the crystallization and stoichiometry challenges can be effectively addressed [138].

To the best of our knowledge, Hu et al. [139] pioneered the use of the thermal evaporation method for fabricating an all-inorganic PeLED in 2017, employing a dual-source co-evaporation technique with an equimolar of CsBr and PbBr_2 . However, due to the low crystallinity of the films, their evaporation-based PeLEDs achieved an EQE of only 1.55%, significantly lower than that of solution-processed

PeLEDs during the same period. The poor crystallinity of MHP films was the main culprit, hindering the performance of these evaporated PeLEDs. Countering this issue, Chen et al. [48] in 2020 discovered that applying a moderate in situ annealing temperature ($60\text{ }^\circ\text{C}$) during the deposition process improved crystallinity and produced better grain orientation. By utilizing this strategy, they successfully fabricated the first thermally evaporated large-area PeLED ($20\text{ mm} \times 70\text{ mm}$) with excellent uniformity (Fig. 4b).

On the other hand, while Hu et al. focused on optimizing the stoichiometric ratio through chemical composition engineering, the effects of composition variation on crystallization dynamics and phase evolution were not deeply explored in their study. To address this gap, Li et al. [121] employed an all-vacuum deposition process to fabricate CsPbBr_3 -based PeLEDs and systematically investigated the influence of the Cs/Pb ratio on phase purity and device performance. By using a nonrotating substrate, they achieved a gradient composition in the perovskite film, as illustrated in Fig. 4c. The Cs/Pb ratio increased from the region above the PbBr_2 source to that above the CsBr source, leading to the formation of mixed CsPb_2Br_5 and CsPbBr_3 phases, which transitioned to CsPbBr_3 and Cs_4PbBr_6 phases. By replacing LiF with NiO_x for the hole-injection layer, all-vacuum deposition PeLEDs reached an EQE of 3.26%. Building on these findings, Li et al. [53] further demonstrated in 2021 that zero-dimensional (0D) Cs_4PbBr_6 could be incorporated into a CsPbBr_3 film via simply modifying the Cs/Pb ratio. This led to the formation of a $\text{Cs}_4\text{PbBr}_6/\text{CsPbBr}_3$ core–shell structure, where Cs_4PbBr_6 acted as the matrix and CsPbBr_3 as nanoinclusions. The size of the CsPbBr_3 nanoinclusions determines the strength of quantum confinement effect (Fig. 4d). This spatial confinement significantly enhanced carrier recombination efficiency, enabling the fabrication of an ultra-large PeLED (4020 mm^2) with an EQE of up to 7.1% and uniform green emission across the entire device area (Fig. 4e), showcasing the potential of thermally evaporated PeLEDs for next-generation large-area displays. In 2022, Li et al. [122] extended this approach by replacing CsBr with CsI in the chamber crucible, fabricating yellow-emitting PeLEDs via a similar all-vacuum evaporation method. Owing to the great importance of controlling the grain size and thickness of perovskite layer, they regulated the growth kinetics by modifying the co-evaporation rate. Consequently, the PeLEDs based on the optimal perovskite film demonstrated the yellow EL (574 nm) with a maximum

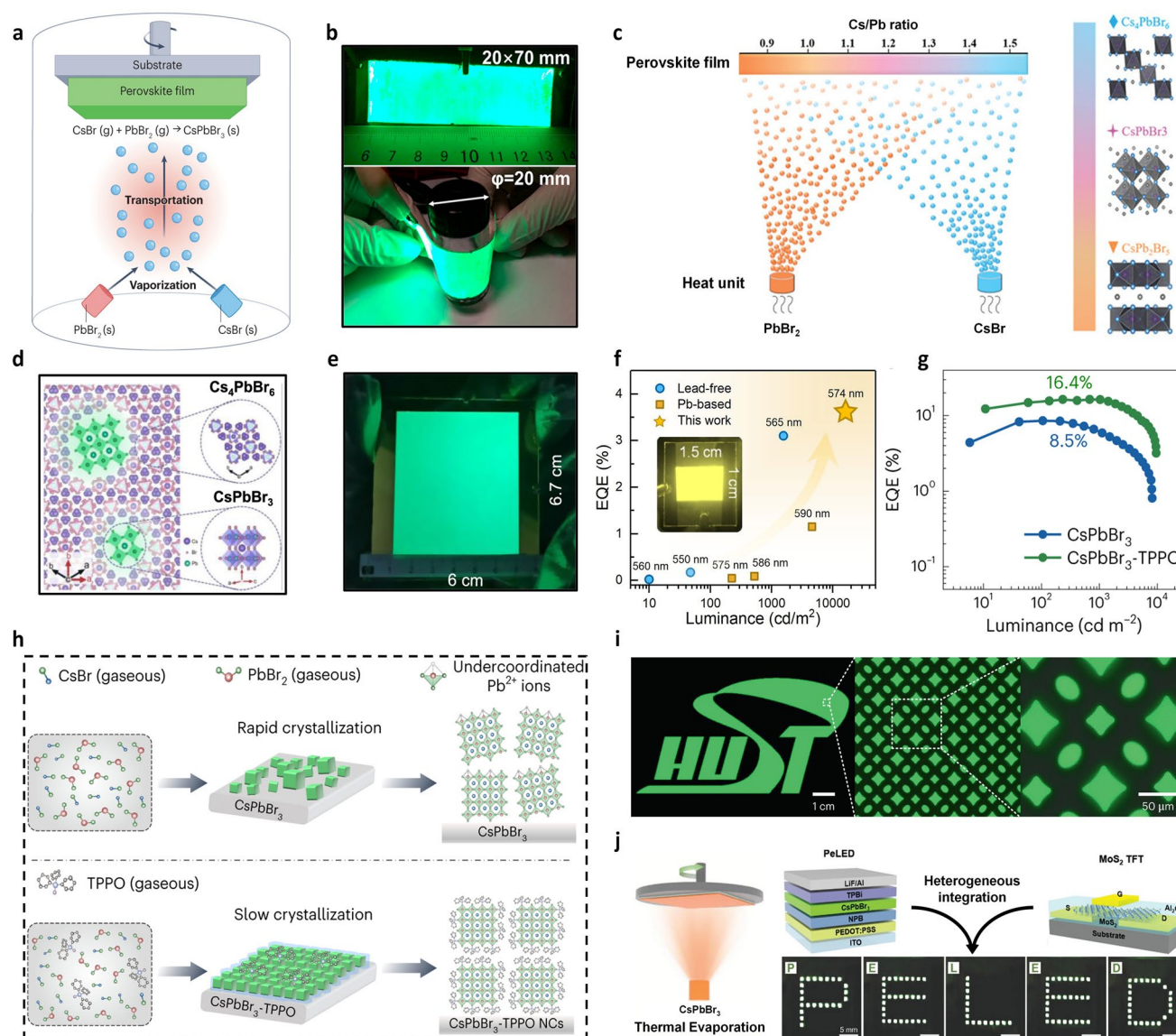


Fig. 4 Thermal evaporation strategy for PeLEDs. **a** Schematic of a dual-source thermal evaporation of perovskite CsPbBr_3 films [40]. **b** Photograph of a large-scale flexible PeLED (70 mm × 20 mm) [48]. **c** Schematic of dual-source thermal evaporation for perovskite film, right insets are structures of Cs_4PbBr_6 , CsPbBr_3 , and CsPb_2Br_5 [121]. **d** Schematic diagram of CsPbBr_3 embedded in the Cs_4PbBr_6 matrix, illustrating the separation of $3\text{D} [\text{PbBr}_6]^{4-}$ of CsPbBr_3 induced by $0\text{D} [\text{PbBr}_6]^{4-}$ of Cs_4PbBr_6 . **e** Photograph of a thermally evaporated 4020 mm^2 PeLED [53]. **f** Statistical charts of EQE versus luminance of the yellow PeLEDs with emission in the range of 550–600 nm. The inset shows a photograph image of CsPbBr_2I film-based $15 \times 10 \text{ mm}^2$ yellow PeLED [122]. **g** EQE versus luminance curves for CsPbBr_3 and $\text{CsPbBr}_3\text{-TPPO}$ -based devices. **h** Schematics of crystallization process for thermally evaporated CsPbBr_3 and $\text{CsPbBr}_3\text{-TPPO}$ films. **i** Display of “HUST” pattern and its arrays of emissive pixel units after magnification by microscope [59]. **j** Left shows schematic illustration of the deposition of CsPbBr_3 powder. Right shows heterogenous integration of CsPbBr_3 PeLED display with a MoS_2 TFT-based backplane and photographs of the operating PeLED display representing each letter of “PELED” [123]

EQE of 3.7%, and the large-area PeLED (150 mm^2) exhibited great uniformity and brightness (Fig. 4f).

In addition to in situ annealing and spatial confinement, Kim et al. and Song et al. further improved the EQE of thermally evaporated PeLEDs by introducing innovative

passivation techniques. Kim et al. [140] introduced a polyethylene oxide (PEO) interlayer doped with MgCl_2 beneath vacuum-evaporated CsPbBr_3 films, increasing the EQE to approximately 7.6%, making it one of the highest-performing vacuum-processed PeLEDs at the time. In contrast, Song

et al. [141] implemented a bilateral interfacial passivation strategy—treating both sides of the perovskite layer—which resulted in a significantly higher EQE, exceeding 15%, along with enhanced operational stability. Recently, Li et al. [122] achieved a milestone breakthrough by developing a tri-source co-evaporation strategy incorporating triphenylphosphine oxide (TPPO) as a Lewis-base additive, which increased the EQE of thermally evaporated PeLEDs to a maximum of 16.4% (Fig. 4g). As illustrated in Fig. 4h, TPPO, with the P=O band, acts as an electron donor that binds to PbBr_2 , serving as a surface ligand to constrain crystal growth. This not only impedes the crystallization of CsPbBr_3 but also promotes the formation of small crystal grains with enhanced charge carrier confinement, effectively passivating the perovskite crystals. Based on this strategy, they fabricated active-matrix PeLED displays by integrating top-emitting PeLEDs onto a 6.67-inch thin-film transistor backplane. These displays demonstrated high-definition patterns and videos with a resolution of $1,080 \times 2,400$ and continuous greyscale information (Fig. 4i), showcasing the potential of thermally evaporated PeLEDs for advanced display technologies.

Further advancing the field, Ji et al. [123] explored heterogeneous integration by combining single-source evaporation-synthesized perovskite films with molybdenum disulfide (MoS_2)-based thin-film transistors (TFTs) to create a unified optoelectronic system. The thermal evaporation process utilized a mixture of CsBr and PbBr_2 , with HBr incorporated to generate uniform large-area films through vapor deposition (Fig. 4j). Although limited to a maximum EQE of 2.21%, Ji et al. successfully fabricated an 8×8 active-matrix luminescent device using ALD-coated MoS_2 TFTs, which demonstrated stable operation, precise brightness control, and negligible response delay. Five characters, “P,” “E,” “L,” “E,” and “D,” were successfully displayed by integrated device (Fig. 4j), highlighting the potential of heterogeneous integration for developing functional electronic systems with enhanced performance and scalability.

3 Patterning Strategies

Achieving high-quality perovskite thin films is essential for optoelectronic performance; however, the transition from uniform films to micro- and nanopatterned architectures represents a critical frontier—where crystal engineering for

emerging photoelectronics intersects with integrated device array technology. Precise patterning of perovskite materials is particularly vital for full-color PeLED displays, as it directly impacts pixel resolution, color purity, and device performance. When pixel lateral size L is reduced (typically $< 50 \mu\text{m}$), PeLED efficiency often declines because the perimeter-to-area ratio rises, so etched/processed sidewalls dominate non-radiative recombination. Simultaneously, to reach a given luminance, the required current density increases, aggravating Auger/Joule heating and current crowding, and field fringing at the contact edges may introduce leakage paths [43]. In fact, the size penalty is not intrinsic to perovskites and can be mitigated by process/device design. For example, active-matrix PeLED arrays show little efficiency loss relative to their large-area prototypes, and localized-contact micro/nano-PeLEDs maintain $\sim 20\%$ EQE while downscaling pixel length from 650 to $3.5 \mu\text{m}$ by preventing boundary non-radiative losses [142]. Practical levers for patterned integration include: (i) damage-minimizing pattern transfer and conformal sidewall passivation to suppress edge recombination; (ii) charge-balance engineering and current-spread control; (iii) high-PLQY, uniform emitters via additive/interface control to preserve film quality in micropatterned pixels [30, 43, 143]. Together, these measures substantially narrow the performance gap between small-area, patterned pixels and their larger-area counterparts.

Patterning techniques for perovskites can be broadly classified into top-down and bottom-up approaches, depending on their underlying processing mechanisms. Top-down methods (Fig. 5a-c)—including photolithography, laser/e-beam lithography, and nanoimprinting—typically rely on subtractive processes such as etching, ablation, or imprinting to define high-resolution features. These techniques offer exceptional spatial precision and geometric control, making them attractive for research-scale device prototyping. However, their inherent serial processing nature, especially in laser and e-beam lithography, limits throughput and scalability. Moreover, perovskite materials often exhibit sensitivity to the solvents, heat, and high-energy exposure used in these techniques, posing material compatibility challenges.

In contrast, bottom-up methods (Fig. 5d, e), such as pre-patterned crystal growth, inkjet printing, and EHD jet printing, are based on the solution processability and low formation energy of perovskites, enabling material assembly under ambient or low-temperature conditions. These techniques

exhibit superior material utilization and reduced fabrication waste, making them attractive for sustainable manufacturing. However, challenges remain in achieving nanoscale resolution, and crystallization dynamics must be precisely controlled to ensure pattern uniformity and reproducibility. Recent advancements in PeLED patterning strategies have significantly improved scalability, resolution, and multi-color integration, paving the way for high-performance and full-color displays (Table 2). In the following section, we systematically examine and compare these six key patterning techniques, analyzing their working principles, technical advantages, and practical applications in PeLED manufacturing.

3.1 Top-down Methods

3.1.1 Photolithography

Photolithography is a well-established semiconductor fabrication technique used to create fine patterns on films or substrates by direct photolithography or combined photomasks [175, 176]. The process typically involves spin-coating a photoresist onto the surface, followed by selective exposure to ultraviolet (UV) light through a patterned photomask [174, 177]. Depending on the type of photoresist used, exposure induces chemical changes that either increase solubility (positive photoresist) or promote polymerization and hardening (negative photoresist) [178, 179]. Commonly utilized resist types and corresponding parameters are summarized in Table 3 for comparison. Despite its precision and scalability, applying photolithography to perovskite materials presents significant challenges due to their intrinsic chemical and thermal sensitivity [180, 181]. Traditional lithographic processes rely on organic solvents and alkaline developers, which can dissolve perovskite layers, disrupt their structural integrity, and induce undesirable ion migration [182]. Meanwhile, high-temperature processing steps, typically around 100 °C, required for photoresist curing and resolution enhancement pose thermal stability challenges, as organic–inorganic hybrid perovskites tend to undergo decomposition or phase transitions within the 100–150 °C range.

To mitigate the detrimental effects of solvents and plasma exposure during photolithography, a stable protective layer—such as inorganic materials (e.g., SiO₂ or SU-8) or

resin-based coatings—is typically deposited or transferred onto the perovskite thin film prior to lithography and etching processes [183]. These encapsulation layers help preserve the integrity of the perovskite’s optoelectronic properties. Harwell et al. demonstrated SU-8 as a negative epoxy-based resist used to facilitate photolithography and polymethyl methacrylate (PMMA) as a removable protecting layer for the perovskite layer [169]. This two-layer protective strategy minimized damage to the perovskite layer during photolithography. The process involved exposing and developing the SU-8 to create a patterned template, followed by the deposition of different colored perovskite materials in the unexposed regions. This process is repeated multiple times, with each new layer of perovskite being selectively deposited in the unexposed areas, resulting in a multicolor pixel array (Fig. 6a). Figure 6b indicates that, with this multilayer protective structure, the PLQY of the perovskite film experienced minimal loss during processing (from 55 to 40%), demonstrating that the photolithography process does not significantly affect the optical properties of the perovskite. With this dual-layer protection structure, they achieved multicolor patterns (green and blue). Despite this success, the requirement for thick protective layers complicates the multicolor photolithography process, posing a significant challenge for fabricating high-quality multicolor patterns.

An alternative strategy involves the application of a photoresist or sacrificial layer on the substrate before perovskite deposition [176]. The perovskite is then grown or deposited on top of this protective layer, and patterning is achieved through a lift-off process or other similar techniques. This approach effectively avoids the direct etching of the perovskite layer, thereby minimizing the risk of material degradation and preserving the integrity of the perovskite’s optoelectronic properties. The key distinction between these two methods mentioned above lies in the sequence of deposition and patterning: the first method involves depositing the perovskite layer before setting the pattern on the template, while the second method deposits the perovskite after the template has been patterned. Using the lift-off approach, Zou et al. successfully fabricated multicolor patterns, including a “University of Washington” logo and red-green pixel arrays (Fig. 6c, d) [147]. However, during the lift-off process, the perovskite layer may adhere to the boundaries of the patterned material, leading to potential damage or loss. This is evident in the observed pattern defects at the boundaries, likely caused



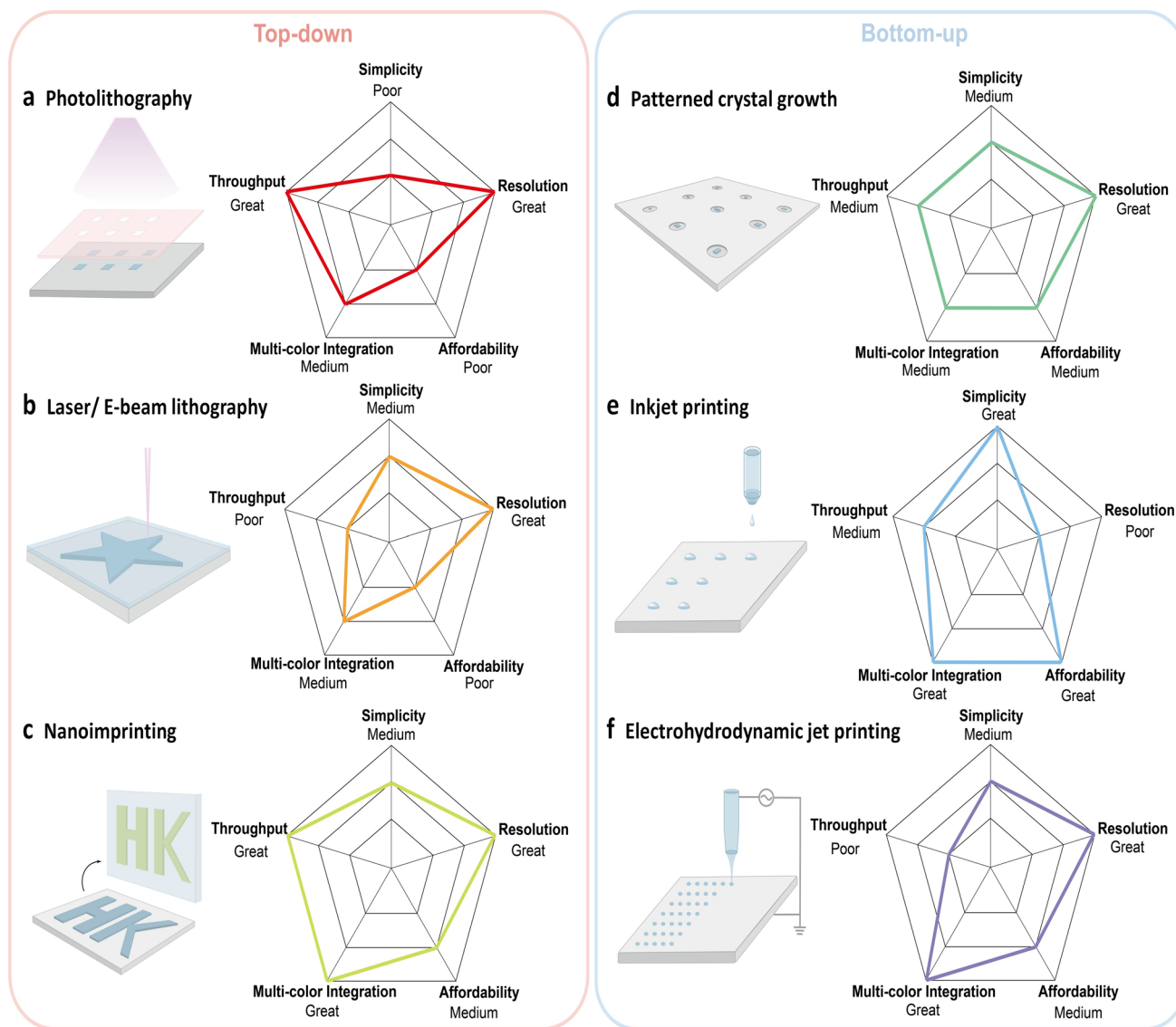


Fig. 5 Perovskite patterning strategies. **a–c** Schematics and radar plots for top-down perovskite patterning techniques. **d–f** Schematics and radar plots for bottom-up perovskite patterning techniques. “Multi-color Integration” refers to the controllability of RGB emission units within a single pixel

by the photolithography or lift-off process. The resulting devices achieved a maximum EQE of 1.24% (Fig. 6e), a current efficiency of 3.85 cd A^{-1} , and a luminance of $13,043 \text{ cd m}^{-2}$, which is relatively low compared to other perovskite patterning strategies. This performance gap can be attributed to two coupled factors. First, chemical incompatibility and residuals contamination: conventional resists, developers, and strippers can dissolve or reconstruct halide perovskites, leaving surface residues that quench PL [147]. A common workaround is to pattern the template first, then deposit the perovskite into the features,

and finally peel or lift off the template, so the perovskite never contacts photoresist solvents or developers. Second, process-induced damage: oxygen-plasma cleans, plasma-enhanced chemical vapor deposition (PECVD) steps, and ambient $\text{O}_2/\text{H}_2\text{O}$ exposure can extract halides, form Pb-oxide and vacancy defects, suppress PLQY, and thus reduce EQE [184]. Accordingly, oxygen plasma could be avoided, and all wet steps could be carried out under an inert atmosphere. For multicolor arrays, repeated photolithography and lift-off cycles, together with protective interlayers to shield prior perovskite layers from solvents

Table 2 The summary of main characteristics of patterned PeLEDs over the past five years

Year	Strategy	Emission layer (wavelength)	Minimum pattern size (μm^2)	Maximum area (mm^2)	Maximum EQE (%)	T_{50} (h, @ 100 cd m^{-2})	Refs
2020	E-beam lithography	MAPbBr ₃ (521 nm)	~300	4	0.1	-	[145]
2020	Inkjet printing	FA _{0.3} Cs _{0.7} PbBr ₃ (520 nm)	~9000	-	2.8	0.12 (@ 90 cd m^{-2})	[146]
2020	Photolithography	PEA-CsPbI ₃ /PEA-CsPbBr ₃ /PEA-CsPbBr _{1.5} Cl _{1.5} (670/523/482 nm)	~300	4	1.24	-	[147]
2021	Inkjet printing	CsPbBr ₃ (515 nm)	~10000	-	3.03	-	[148]
2021	Inkjet printing	PEA-CsPbI ₃ /PEA-CsPbBr ₃ /PEA-CsPbBr ₃ (645/515/486 nm)	~9000	10	3.5(R), 3.4(G), 1.0(B)	0.03 (@ 128 cd m^{-2})	[149]
2021	Inkjet printing	MAPbBr ₃ (536 nm)	~25000	-	0.804	-	[150]
2021	Inkjet printing	CsPbBr ₃ (518 nm)	~6075	1288	9	0.13 (@ 120 cd m^{-2})	[151]
2022	Laser lithography	CsPbBr ₃ (512 nm)	~900	25	-	-	[152]
2022	Transfer printing	Cs _{0.7} MA _{0.3} Pb(I _{0.8} Br _{0.2}) ₃ and CsPb(Br _{0.84} Cl _{0.16}) ₃ (680/493 nm)	25	225	10.5	0.75	[153]
2022	Nanoimprinting	CsPbBr _{3-x} I _x (650.9 nm)	~100	4	5.9	0.1	[154]
2022	Nanoimprinting and transfer printing	CsPbBr _{0.6} Cl _{0.4} /CsPbBr ₃ /CsPbI ₃ (480/516/670 nm)	3	~9	15.3(R), 14.8(G), 2.5(B)	0.35	[34]
2022	Inkjet printing	CsPbBr ₂ /CsPbBr ₃ /CsPbCl _{1.56} Br _{1.44} (646/514/465 nm)	~675	625	0.83(R), 0.42(G), 0.05(B)	0.13(R), 0.04(G), 0.01(B)	[155]
2022	Inkjet printing	PEA ₂ SnI ₄ (633 nm)	-	32	1	~3	[156]
2022	Inkjet printing	CsPbBr ₃ (517 nm)	~6075	555	8.54	1.1	[157]
2022	Inkjet printing	CsPb(I/Br) ₃ (640 nm)	~9000	-	9.6	0.19	[158]
2023	Inkjet printing	MAPbBr ₃ /PEG (530 nm)	-	1600	0.5	-	[159]
2023	Crystal growth	CsPb 6-4Cl-Br/4-6Cl-Br/CsPbBr ₃ /I-Br (478/491/512/630 nm)	40,000	1050	9.97(R), 26.09(G), 16.49(sky-B), 12.41(B)	0.39	[57]
2024	Nanoimprinting	PEA-CsPbBr _{3-x} Cl _{3-x} (471.2 nm)	~75	4	6.1	-	[160]
2024	Inkjet printing	CsPbBr ₃ (505 nm)	-	360	5.47	0.14	[161]
2024	Inkjet printing	FA _{0.8} Cs _{0.2} PbI ₃ /CsPbBr ₃ /Cs _{0.75} EA _{0.25} PbBr ₃ (769/515/490 nm)	-	2800	14.3	2.59	[162]
2024	Crystal growth	CsPbI _{3-x} Br _x /CsPbBr ₃ /CsPbBr _{3-x} Cl _x (625/512/490 nm)	-	400	7.6(R), 11.6(G), 4.3(B)	720(R), 1400(G), 1000(B)	[163]
2024	Inkjet printing	PEA ₂ MA ₃ Pb ₄ I ₁₃	~7500	400	-	-	[164]
2025	Photolithography/ Focused ion beam etching	(FA, Cs)PbI ₃ /(FA, Cs)Pb(Br,Cl) ₃ /(FA, Cs)PbBr ₃ /(Cs, FA, Rb)Pb(Br,Cl) ₃ (800/645/525/488 nm)	0.008	-	20.6(NIR), 12(R), 20.6(G), 12(sky-B)	40(G, @ 10 mA cm^{-2})	[142]
2025	Photolithography	CsPbBr ₃ (516 nm)	~6.5	625	13.09	0.5	[165]
2025	Crystal growth	CsPbBr ₃ /CsPb(Br,I) ₃ (516/640 nm)	20	100	-	-	[166]
2025	Nanoimprinting	FA _{0.15} Cs _{0.85} PbBr ₃ (517 nm)	~2.8	225	15.79	74.4	[167]
2025	Nanoimprinting	CsPbBr ₃ (460 nm)	~20	225	5.04	-	[168]

and etchants, further increase process complexity and cross-contamination risk. While these techniques offer precise patterning capabilities, their limitations in terms of efficiency and process complexity underscore the need for further innovation in perovskite patterning strategies.

Photolithography also plays a critical role in the patterning of PeQDs, facilitating their spatially controlled formation and integration [185–187]. This technique enables both the direct conversion of perovskite precursor solutions into well-defined PeQD patterns and the realization of ligand- or polymer-assisted synthesis and patterning

strategies, thereby achieving high-resolution and ordered structures suitable for optoelectronic applications [188, 189]. Recently, Zhou et al. [170] proposed a groundbreaking method for directly synthesizing perovskite quantum dot photoresists, significantly simplifying the photolithographic process. In this method, isobornyl acrylate (IBOA) effectively dissolved perovskite precursors and triggered photopolymerization upon UV exposure, producing high-brightness, high-stability PeQD patterns after development (Fig. 6f). Figure 6g demonstrates that the PL intensity of both the perovskite film and the perovskite photoresist



Table 3 Summary of typical perovskite-compatible photoresist types and critical metrics

Resist type	Developer/process solvent	Typical bake/solvent exposure	PLQY retention (after a full cycle)	Typical minimum feature size of perovskite	Refs
Positive novolac/DNQ i-/g-line PRs (S1813, AZ series)	TMAH aqueous developers (AZ 300 MIF, MF-319)	~90 °C soft-bake; ~110 °C PEB (tool-dependent)	Significant weaken (directly exposed); safe (pattern-first, deposit-later)	≥ 10 μm	[147]
Negative epoxy PR (SU-8)	PGMEA (SU-8 Developer)	95 °C soft-bake; 150–200 °C PEB/hard-bake	Significant weaken (directly process on perovskite); generally used as mask	~1–3 μm (tool-dependent)	[172]
PMMA/ZEP EBL stacks (perovskite-compatible)	Non-polar / weakly polar developers (o-xylene, toluene; ZED-N50/amyl acetate; hexane rinse)	~90–100 °C	No weakening (orthogonal solvent set avoids perovskite attack)	~50 nm	[173]
Orthogonal photolithography	Chlorobenzene:hexane (≈1:3) or fluoruous HFEs with fluorinated resists	≤ 100 °C	No weakening (minimal PL degradation reported when fully orthogonal)	~1–5 μm routinely; 5 μm RGB PeQD	[171, 172, 174]

solution remained mostly unchanged after the lithography, and the emission peak wavelength of the cured film matched that of the original PeQD photoresist. However, the precision of this method can be influenced by factors such as precursor concentration, photoresist thickness, and exposure conditions. Additionally, post-exposure development may introduce defects, limiting the overall quality of the patterned PeQD. In contrast, the ligand/polymer-assisted synthesis method enhances deposition selectivity and PeQD stability by incorporating functionalized ligands and polymer layers. This strategy not only improves the selective deposition of quantum dots but also stabilizes them against environmental degradation [170, 190].

A notable advancement is the development of a non-destructive technique that directly patterns perovskite precursors without exposing them to high-energy UV light or harsh solvents. Instead, PeQDs are generated in situ through annealing, ensuring uniform distribution and high optical quality (Fig. 6h) [171]. UV-activated lead bromide complexes act as catalysts for thiol-ene free-radical polymerization. Sulfur radicals generated during the process initiate polymer chain formation, while oxygen facilitates the regeneration of lead bromide complexes, ensuring sustained catalytic activity. Additionally, it supports the fabrication of multi-color PeQD patterns with customizable thicknesses up to 10 μm, making it suitable for red and green color displays (Fig. 6i). Furthermore, polymer encapsulation provides robust stability by protecting PeQDs from ion migration, crystal aggregation, and environmental factors such as moisture, oxygen, and heat (Fig. 6j). Despite these advantages, the ligand/polymer-assisted synthesis method introduces additional complexity due to the need for functionalized ligands and polymer layers. These components can affect the adhesion and distribution of quantum dots on the substrate, potentially leading to uneven deposition and impacting the final patterning quality and optical performance. Nevertheless, this method represents a significant step forward in achieving high-resolution, multicolor PeQD patterns for advanced display applications.

3.1.2 Laser/E-beam Lithography

Instead of utilizing conventional UV light, electron beam (e-beam) and laser irradiation have been explored as alternative patterning techniques for perovskite optoelectronic

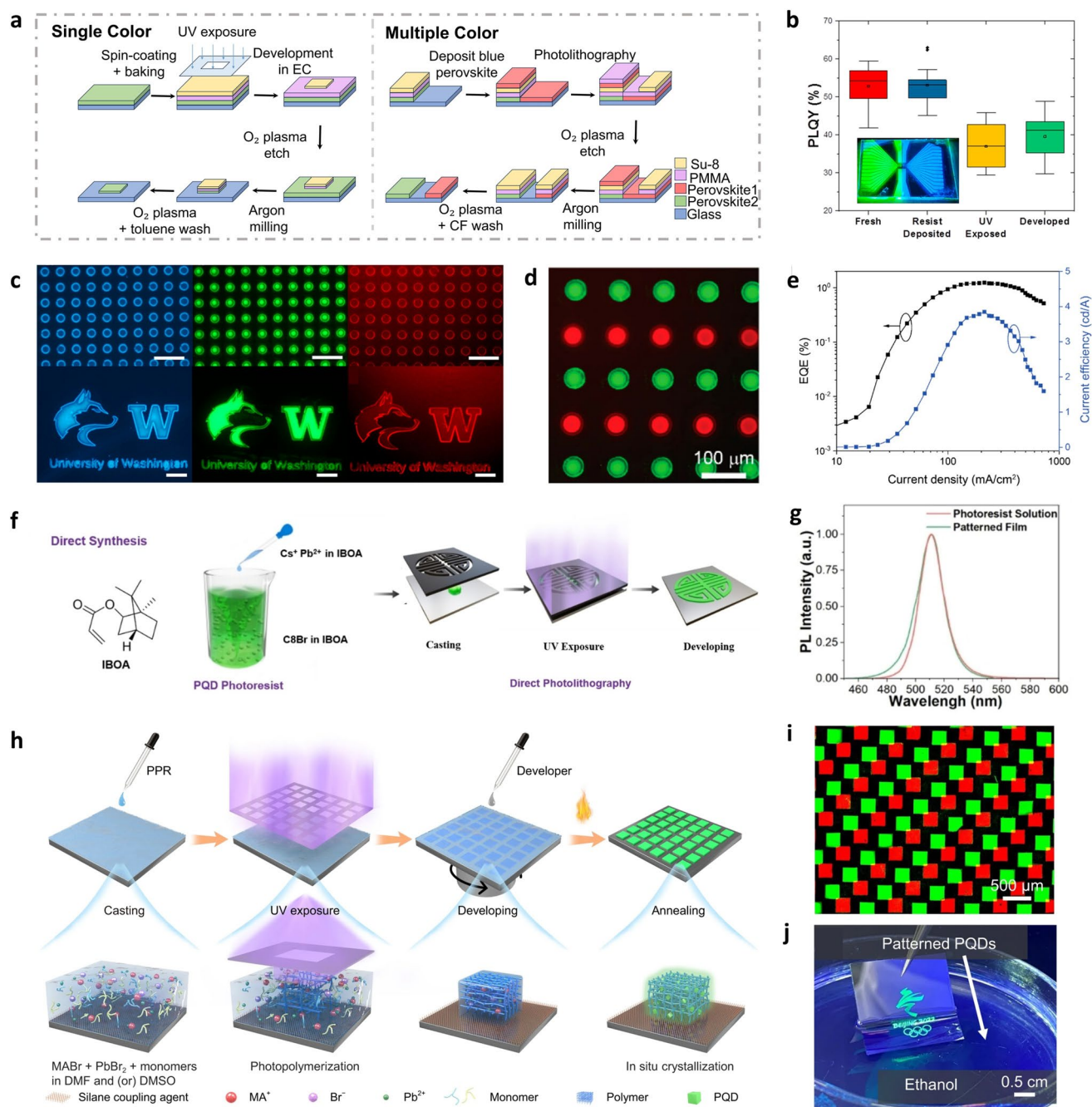


Fig. 6 Photolithography strategy for perovskite patterns and PeQDs. **a** Multicolor patterning process by photolithography. **b** PLQY of perovskite sample under different processing conditions. Inset: green and blue color patterns [169]. **c** PL images of multicolor patterning (scale bar: 50 μm) and "University of Washington" logo (scale bar: 200 μm). **d** PL image of multicolor (red and green) pattern. **e** EQE and current efficiency versus current density of PeLEDs [147]. **f** The process of the direct synthesis method for PeQDs. **g** The PL spectra of photoresist solution and patterned film [170]. **h** Schematic of the direct in situ photolithography method using PPR, where annealing involves heating the samples to a precise temperature. **i** PL images of red and green patterns. **j** Photograph of a green logo immersed in ethanol under UV light [171]

devices [194–200]. These methods eliminate the need for photomasks, enabling programmable, high-resolution, and highly flexible patterning processes on both flat and curved

surfaces [199]. In e-beam lithography, a focused e-beam is used to expose the resist layer, creating a nanoscale pattern that serves as a mask for subsequent etching or material

deposition, thereby defining the geometry of the perovskite layer [201]. While e-beam lithography offers excellent resolution down to the nanometer scale, its high cost and low throughput limit its practicality for large-area applications [202]. Laser lithography, on the other hand, replaces the electron beam with a focused laser and typically employs conventional photoresists instead of specialized e-beam resists, offering a faster and more cost-effective alternative [203]. However, its resolution is constrained by the laser spot size, typically on the micrometer scale [204, 205]. Depending on the interaction between the e-beam/laser and the perovskite material, these lithography strategies can be classified into two main categories: direct ablation, which physically removes material to create patterns, and photo-thermal-induced crystallization, which enables localized perovskite crystallization [206, 207]. Besides of perovskite patterning, these techniques can also be employed for fabricating structured substrates, photoresist molds, and even inducing selective crystallization in perovskite films [208].

In 2017, Chen et al. [191] introduced an innovative application of laser direct writing (LDW) for patterning all-inorganic perovskite quantum dots (CsPbBr_3), marking its first use in this context. Without the requirements of vacuum environments or complex post-processing, LDW simplifies the fabrication process into three efficient steps: spin-coating quantum dots, laser writing, and solvent washing (Fig. 7a). By adjusting parameters such as laser spot size, scanning speed, and energy, LDW achieves controllable pattern resolutions ranging from 3.3 to 100 μm , making it suitable for both microscale and large patterns. Additionally, LDW not only defines patterns but also optimizes the morphology and PL intensity of QDs, enhancing both brightness and morphology quality. A fabricated large-scale 100 mm \times 100 mm pattern demonstrates its potential for applications in quantum dot displays and optical storage.

Due to its high precision, rapid processing, and programmable patterning capabilities, femtosecond laser ablation (FsLA) has emerged as a promising method for fabricating perovskite films with resolutions down to the micrometer scale and 3D patterning of PeQDs [209]. Compared to conventional laser writing, which typically uses continuous wave or nanosecond pulsed lasers, FsLA achieves higher spatial resolution and minimizes thermal damage by confining energy deposition within extremely short pulses. This enables precise ablation without affecting surrounding areas, preserving perovskite material

quality [210]. Based on this strategy, Liang et al. [211] utilized a focused NIR femtosecond laser (1030 nm) to selectively ablate perovskite material through lattice melting and Coulomb explosion, achieving fine 2D perovskite patterns with resolution as high as 1.78 μm . These patterns featured diverse geometries, including portraits, microline arrays, and microsquare grids, all fabricated without introducing additional pinholes or cracks. Utilizing this FsLA strategy, they developed fluorescent anti-counterfeiting labels capable of retaining their properties even in environments with 96% relative humidity, highlighting their potential for high-security authentication applications. To further enhance device integration, they introduced a sandwich-laminated structure, in which perovskite films were enclosed between two glass layers to prevent debris contamination during processing (Fig. 7b) [152]. As shown in Fig. 7c, this approach yielded complex perovskite patterns with well-defined edges and uniform PL properties, which were successfully incorporated into PeLED devices, demonstrating high-quality emission characteristics.

In contrast to conventional thermal annealing, which lacks spatial control over crystallization, laser-induced photothermal heating provides a precise means of inducing local perovskite crystallization [212]. Femtosecond lasers, known for their ability to create three-dimensional photonic structures with high spatial resolution, achieve this through rapid atomic redistribution enabled via nonlinear optical processes [213]. Using femtosecond laser-induced liquid nanophase separation, Sun et al. achieved 3D direct lithography of PeNCs embedded in glass with tunable photoluminescence from 480 to 700 nm [192]. As illustrated in Fig. 7d, localized laser heating and pressure gradients drive halide migration, initially forming Br-rich regions (~ 520 nm green emission), with prolonged exposure inducing I^- incorporation, shifting the red emission (~ 690 nm). The integration of Cl^- ions further enables precise emission tuning, enabling the creation of multicolor perovskite patterns like $\text{CsPb}(\text{Br}_{1-x}\text{I}_x)_3$ and $\text{CsPb}(\text{Cl}_{1-x}\text{Br}_x)_3$ NCs, suitable for diverse optoelectronic applications (Fig. 7e, f). Utilizing 3D laser printing, Huang et al. also demonstrated the reversible in situ formation of MHP QDs embedded within a transparent glass matrix [193]. Compared to solution-processed QDs, this approach offers enhanced stability and seamless device integration. The multiphoton absorption effect of the laser effectively induces nucleation within localized regions, enabling precise 3D patterning. As illustrated in Fig. 7g, patterns were

selectively erased and re-formed through laser-induced decomposition and subsequent low-temperature annealing, allowing for multiple processing cycles while maintaining the QD structure and luminescent properties. This method paves the way for high-precision, reconfigurable optoelectronic devices.

Beyond direct lithography, e-beam lithography has been employed to fabricate perovskite-based display components. Using an e-beam lithography patterned PMMA resist, Wang et al. reported a wettability-guided screen-printing technique to produce multicolor perovskite microdisk arrays with precise geometry [145]. This technique exploits wetting/dewetting mechanisms, where the surface energy contrast created by the EBL-patterned resist directs the placement of perovskite ink, enabling controlled deposition of microdisk arrays. The resulting microstructures exhibited whispering gallery mode (WGM) lasing with tunable colors across the visible-to-near-infrared spectrum, making them suitable for high-resolution display applications. Besides, they successfully fabricated micro-LEDs with a residual PMMA-induced "current-focusing" architecture within the transporting layer of TFB as illustrated in Fig. 7h, enhancing charge injection efficiency for practical display integration (Fig. 7i). However, poor crystallization control and insufficient grain boundary passivation in perovskite films remained significant challenges, ultimately limiting the EQE of fabricated PeLEDs to ~0.1%.

3.1.3 Nanoimprinting

Nanoimprinting is a cost-effective, high-resolution, and high-throughput lithography technique that enables direct replication of nanoscale features onto substrates using a soft stamp with an inverse pattern [214, 215]. This method is widely employed in perovskite optoelectronic devices, either by imprinting patterns onto an electron transport layer (e.g., TiO_2 or ZnO) or directly onto the perovskite active layer [42, 215–218]. In the case of perovskite films, a thin layer of perovskite precursor material is first deposited onto a substrate, followed by pressing a pre-patterned mold—typically made of chemically inert, low surface energy materials such as silicon, quartz, or polydimethylsiloxane (PDMS)—under controlled pressure and temperature conditions [219, 220]. Solvent evaporation and directional deformation guide crystallization into the mold geometry, followed by thermal

or chemical curing. Imprint quality depends on precursor viscosity, mold-substrate surface energy, applied pressure, temperature, and solvent evaporation rate. Pattern formation is governed by capillary filling and viscous flow, followed by rapid solidification to retain shape. Feature resolution can reach ultra-high values (about 20 nm), but is limited by mold fidelity, material relaxation, and capillary-driven reflow [221]. In contrast with photolithography, nanoimprinting enables high-resolution pattern formation without the diffraction limit of light, making it particularly suited for fabricating surface-relief quasi-3D structures and compatible with flexible substrates due to its low-temperature and low-pressure process conditions [214, 222]. However, nanoimprinting strategy still suffers from several problems, including materials compatibility issues, as perovskite precursors are highly sensitive to temperature variations. Additionally, controlling the material flow within the mold is difficult, often resulting in non-uniform pattern formation. Large-area uniformity and alignment remain critical issues, and scaling up the process for industrial applications requires precise control over imprinting parameters, multiple processing steps, and optimization of curing conditions [223]. Despite these issues, nanoimprinting remains a promising technique for fabricating high-resolution perovskite films, offering excellent patterning fidelity for advanced optoelectronic applications.

Although nanoimprinting has been extensively utilized in fabricating patterned OLED [224], QDLED [225], solar cell [223, 226], photodetector [227], and other optoelectronic devices [154, 228–230], its application to PeLEDs began only recently in 2019. Shen et al. [144] employed bioinspired moth-eye nanostructures (MEN) at the front electrode/perovskite interface to enhance the outcoupling efficiency of waveguided light, by utilizing nanoimprinting to fabricate CsPbBr_3 -based green-emitting PeLEDs. As illustrated in Fig. 8a a sol-gel-derived ZnO precursor was spin-coated onto an ITO glass, followed by imprinting with a PDMS mold containing the MEN pattern. After conformal pressing and post-annealing at 150 °C, subsequent layers of PEDOT: PSS, CsPbBr_3 perovskite, TPBi, LiF, and Al were sequentially deposited to complete the PeLED device. The cross-sectional scanning electron microscope (SEM image) in Fig. 8b shows the patterned ZnO layer with an inverse-patterned CsPbBr_3 layer. This nanoimprinted structure significantly reduced Fresnel reflections, enabling the PeLED to achieve an EQE of 20.3%, which increased to 28.2% with



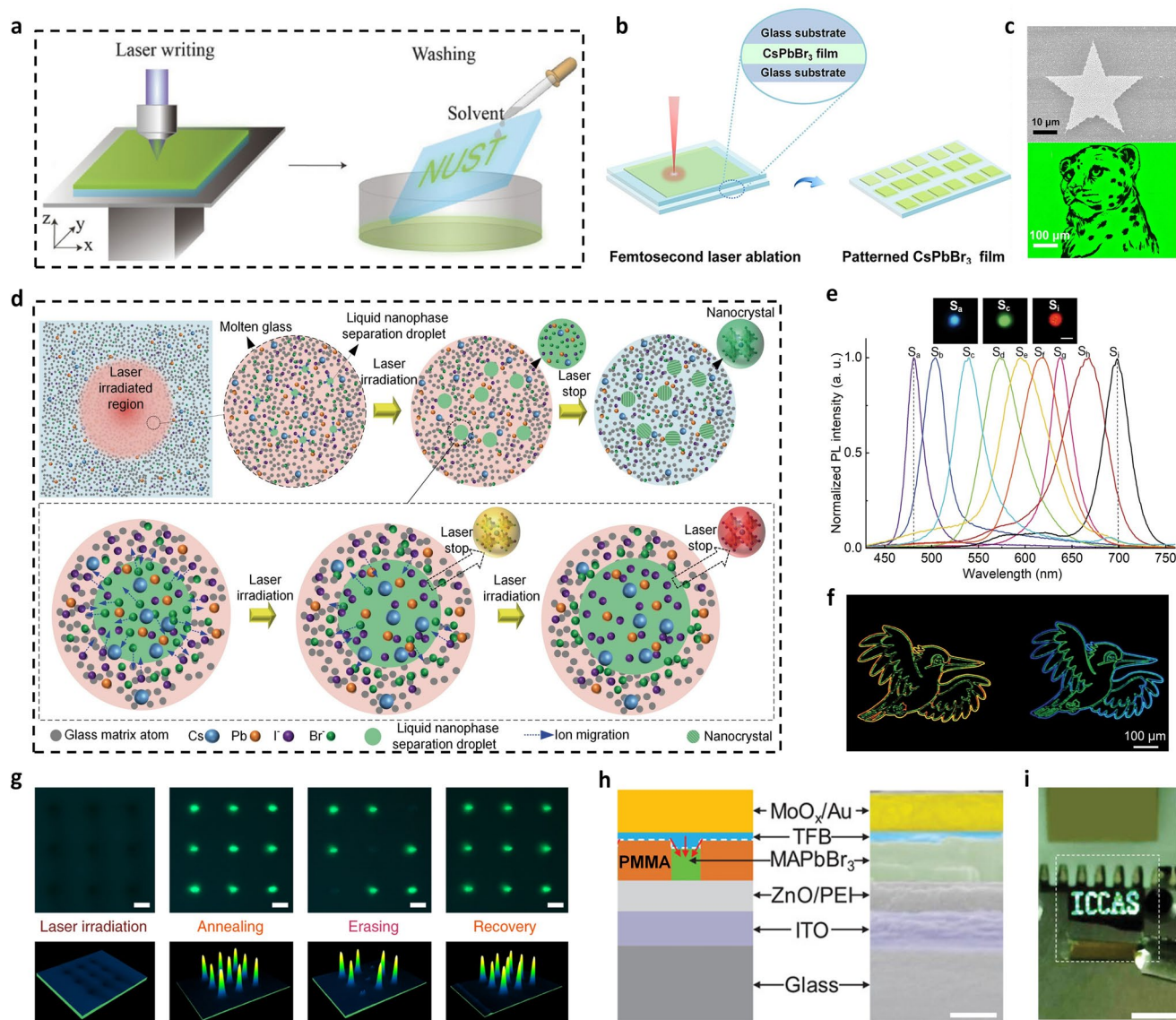


Fig. 7 Laser/e-beam lithography strategy for perovskite patterns and luminescent applications. **a** Schematic illustration of laser-induced patterning process [191]. **b** Schematic illustration of patterned CsPbBr₃ films fabricated via lamination-assisted femtosecond laser ablation (LA-FsLA). **c** High-magnification SEM image and PL image of the patterned CsPbBr₃ film [152]. **d** Schematic of ultrafast laser-induced liquid nanophase separation and formation of CsPb(Br_{1-x}I_x)₃ NCs in the Br⁻-I⁻-doped glass. **e** PL mappings and PL spectra of PeNCs written in the Cl⁻-Br⁻-I⁻ co-doped glass. S_a to S_i represent the PeNC samples written in the Cl⁻-Br⁻-I⁻ co-doped glass with different laser parameters. **f** PL images of the multicolor patterns produced with CsPb(Br_{1-x}I_x)₃ NCs in the Br⁻-I⁻ doped glass and CsPb(Cl_{1-x}Br_x)₃ NCs in the Cl⁻-Br⁻ doped glass [192]. **g** Optical images (upper) and readout signal intensity mapping images (lower) of a CsPbBr₃ QD array during the erasing–recovery processes under UV light (scale bars, 100 μm) [193]. **h** Schematic structure and cross-sectional SEM image of fabricated perovskite micro-LED device (scale bars, 100 nm). **i** Display of “ICCAS” characters using perovskite micro-LED arrays (scale bars, 10 mm) [145]

a half-ball lens. To address the challenge of fabricating ultra-high-resolution display, Kim et al. developed a micropatterning method utilizing capillary force lithography, achieving 10-μm-scale micropatterns [154]. This method incorporated poly(ethylene oxide) (PEO) and an O₂ plasma-treated PDMS

mold to suppress self-aggregation and enhance the heterogeneous nucleation of the perovskite precursor, resulting in highly uniform microarrays. The fabricated micro-PeLEDs exhibited a characteristic red emission at 650.9 nm and an EQE of 5.9%.

While nanoimprinting offers high-precision patterning, it faces inherent challenges in controlling the nucleation and crystallization dynamics of perovskite materials. The confinement of precursors within nanoscale cavities can lead to non-uniform grain growth and defect formation due to uneven solvent evaporation and stress accumulation. Moreover, nanoimprinting is typically limited to single-layer patterning and lacks the flexibility for spatially selective deposition, making it unsuitable for multicolor pixel fabrication [231]. To address these limitations, transfer printing has emerged as a complementary technique, allowing crystallization and patterning to be performed independently. By first developing high-quality perovskite films or nanocrystal layers and then transferring them onto target substrates, this technique minimizes defects and enables advanced pixel architectures [232]. Transfer printing typically employs soft, viscoelastic stamps (e.g., PDMS), which facilitate conformal contact and/or separation through controlled adhesion modulation [233]. Recent studies have demonstrated laser- or thermally triggered release protocols that tune interfacial adhesion, enabling precise pattern transfer with sub-micron alignment accuracy [232, 234, 235]. For example, double-layer printing with organic charge transport layers has achieved ultrahigh resolution up to $\sim 3 \mu\text{m}$ RGB pixels ($\approx 2,550$ pixels per inch) without internal cracking and 100% pattern transfer yield on flexible substrates [34]. The underlying physics involves carefully balancing the work of adhesion, viscoelastic stamp mechanics, and stamp–substrate separation kinetics to favor either pickup or release. Thermal and laser triggers temporarily alter adhesion properties, while peeling dynamics define whether the film remains on the stamp or transfers to the substrate [231, 236–240]. By decoupling film quality control from pattern alignment, transfer printing surmounts nanoimprinting's limitations—enabling large-area multicolor integration, high-resolution patterning, and compatibility with flexible or curved substrates—making it a versatile and scalable route for next-generation PeLED devices [241].

Li et al. developed a reliable mass transfer printing technique for perovskite films and nanostructures, achieving comparable results to optimized spin-coated films in terms of surface morphology, composition, and optoelectronic properties [153]. The method enabled high-resolution patterning

up to 1270 pixels per inch (PPI). A key innovation was the introduction of an ultrathin branched-polyethyleneimine(B-PEI) chemical bonding layer, which effectively resolved electrical contact issues between the transferred perovskite films and the receiving substrate (Fig. 8c). Using this method, they fabricated PeLEDs with decent EQEs of 10.5% (red) and 6.7% (sky-blue) (Fig. 8d). Additionally, they demonstrated a novel white PeLED structure by laterally aligning red and sky-blue perovskite microstripes via multiple transfer steps (Fig. 8e). To further improve performance, alignment accuracy can be improved using an automated alignment system that utilizes an optical microscope with computer vision algorithms and a motorized microstage to achieve placement accuracy below 40 nm, or microstripes can be transferred onto the TFT to independently control the drive voltage for each color, eliminating the need for a PMMA layer [242].

A key limitation of conventional dry transfer printing is the formation of internal cracks in the PeNC film during transfer. To address this, Kwon et al. demonstrated a double-layer transfer printing approach, incorporating both PeNCs and an organic charge transport layer [160]. Specifically illustrated in Fig. 8f, this method begins by simultaneously picking up spin-coated PeNCs and thermally evaporated TPBi using a viscoelastic PDMS stamp. The assembly is then gently placed onto an intaglio trench, allowing selective removal of the double-layer structure due to the surface energy contrast between the PDMS stamp and the trench. After construction of RGB subpixels by repeating the printing method, solvent treatment is conducted to enhance the junction properties between the emitting and charge transport layers, optimizing performance for LED applications. This double-layer transfer printing technique enabled the fabrication of RGB pixelated PeNC patterns with an ultrahigh resolution of 2550 PPI and a near-perfect transfer yield ($\sim 100\%$) (Fig. 8g, h). Moreover, PeLEDs with transfer-printed active layers demonstrated outstanding EQEs of 15.3% for red devices and 14.8% for green devices, as well as synthesis of ultrathin multicolor PeLEDs that can operate on human skin. These results indicate double-layer transfer printing holds significant promise for next-generation high-resolution displays and wearable optoelectronic devices.



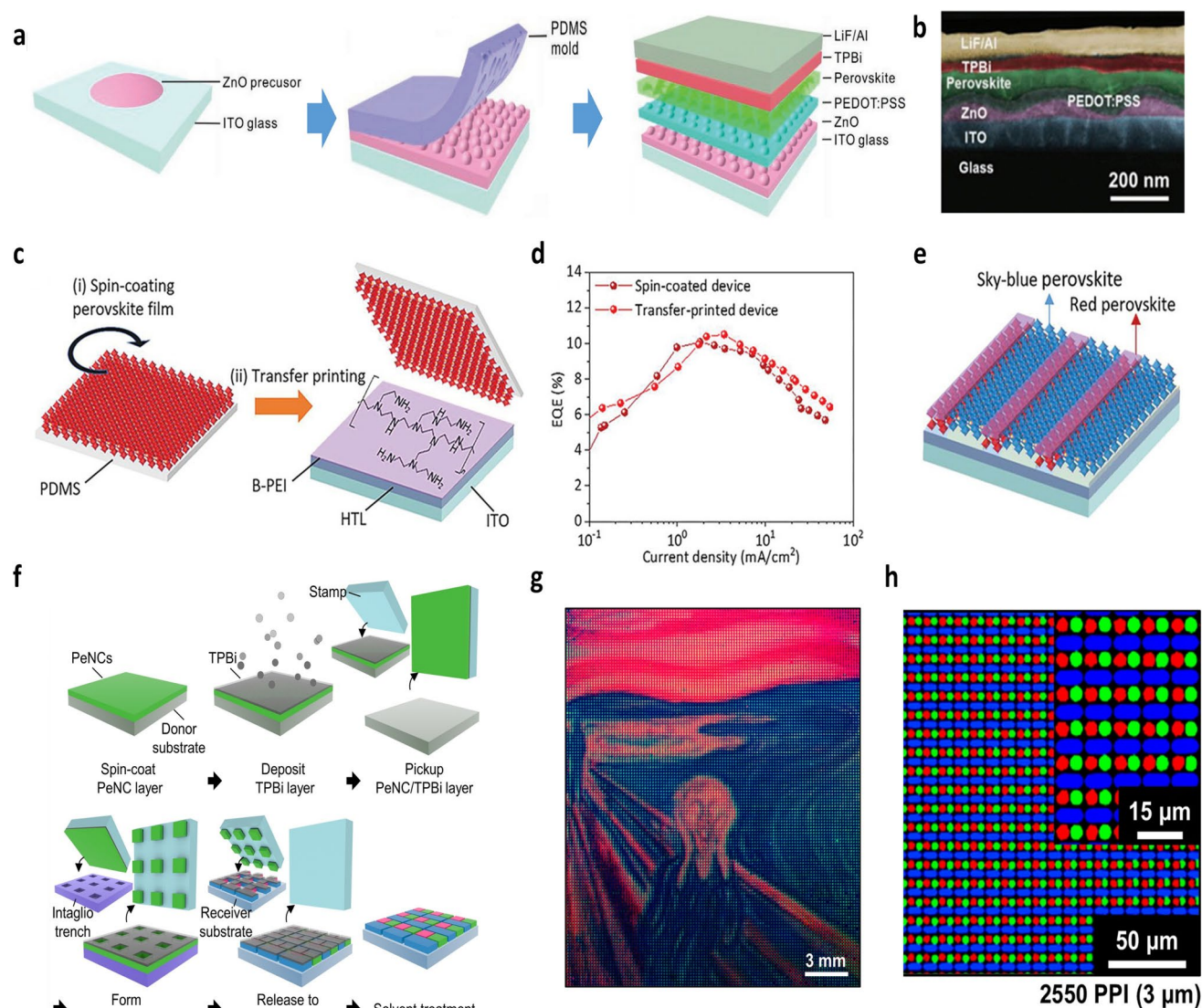


Fig. 8 Nanoimprinting for PeLEDs. **a** Schematic illustration of the fabrication process of CsPbBr₃ PeLEDs with imprinted nanostructure. **b** Cross-sectional SEM image of a nanoimprinted PeLED [144]. **c** Schematic illustration of transfer-printing process of perovskite films. **d** EQE of transfer-printed and spin-coated red PeLEDs. **e** Schematic illustration of the structure of a transfer-printed white emission perovskite film [153]. **f** Schematic illustration of the double-layer transfer printing process with RGB pixelated arrays of PeNCs. **g** PL image of transfer-printed RGB PeNC pixelated patterns displaying *The Scream* by Edvard Munch. **h** Fluorescence microscopic image of the pixelated RGB PeNC patterns with 2550 PPI resolution [34]

3.2 Bottom-up Methods

3.2.1 Patterned Crystal Growth

Among the various approaches for synthesizing perovskite films, crystal growth methods have emerged as crucial strategies for optimizing material performance [247, 248], stability [249, 250], and scalability [251, 252]. From a thermodynamic perspective, crystal growth is driven by

the minimization of the system's free energy. During the transition from a disordered precursor state to an ordered crystalline structure, the reduction in Gibbs free energy facilitates nucleation and subsequent crystal growth. Controlling surface energy, crystallization rate, and phase stability—particularly through modifications that increase the Goldschmidt tolerance factor or reduce crystal size—further optimizes the energy landscape, enabling the formation of high-quality films with suppressed defect densities

and enhanced photovoltaic performance [253, 254]. The growth of perovskite patterns is typically achieved by using templates with specific shapes and structures to guide the perovskite material to grow in a particular morphology and arrangement on the template surface or structure, resulting in highly oriented displays [255]. Alternatively, by hydrophilic/hydrophobic treatments to the substrate and large-area film fabrication methods such as blade-coating, growth can be localized to specific regions, enabling the formation of arrayed perovskite crystals for patterning in display applications [256]. These strategies enable the fabrication of high-quality perovskite materials with fewer defects, reduced non-radiative recombination, and improved carrier mobility [257]. By precisely controlling growth conditions, the size and morphology of the crystals can be tailored, leading to more efficient and stable devices [258].

The surface-confined crystal growth typically involves a confinement depth of around 100 nm, which is relatively small compared to macroscopic dimensions and can therefore be regarded as a surface treatment. In this strategy, perovskite precursors are confined to pre-defined surfaces or voids on the substrate, often leading to superior crystallinity relative to thin films produced by spin-coating. Fan et al. reported the fabrication of crystalline all-inorganic perovskite quantum wire arrays with ultrahigh density and exceptional uniformity using porous alumina membranes (PAMs) with ultra-small pore sizes (6.4 nm) as templates, eliminating the need for organic ligands and antisolvents (Fig. 9a) [57]. The PeLEDs based on these quantum wire arrays, incorporating a dual-functional molecule as both a passivation layer and a hole transport layer, achieved EQEs of 9.97%, 12.41%, 16.49%, and 26.09% for pure-red, blue, sky-blue, and green emissions, respectively (Fig. 9b). Flexible ($15 \times 15 \text{ mm}^2$) and large-area ($30 \times 35 \text{ mm}^2$) PeLED device based on PAMs demonstrated stable EL emission. Since commonly employed patterning and etching techniques are designed to obtain perovskite patterns on two-dimensional surfaces, achieving perovskite patterns on curved substrates represents a significant challenge. To address this, perovskite quantum wires (PeQWs) have been integrated into aluminum fibers through a roll-to-roll solution-coating technique, which allows for the growth of patterned perovskite material on the curved surface of the fibers [163]. As illustrated in Fig. 9c, the thin aluminum fibers, due to their malleability and plasticity, are well-suited for both 2D and 3D architectures. This is exemplified by a 3D multicolor “night

scene” of Victoria Harbour, demonstrating their versatility in creating complex freestanding and multicolor display structures (Fig. 9d). The PeQWs exhibited nearly 90% PLQY, and the resulting fiber LEDs (Fi-LEDs) achieved a high EQE of 15.2% (Fig. 9e). These Fi-LEDs maintained excellent stability under strong external interference, such as twisting and water immersion, demonstrating excellent stability. Despite these advancements, the need for pre-treatment of the Al_2O_3 substrate to form PAM templates adds complexity to the process, potentially hindering industrialization. Additionally, SEM images indicate that the hole arrangement in the PAMs was less regular and uniform than depicted in the schematic (Fig. 9a), which could impact device performance.

Focusing on perovskite crystal growth with different morphologies, space-confined perovskite crystal growth utilizes preconfigured templates to guide perovskite growth within defined spaces, forming structures with predetermined shapes. Kędziora et al. [243] demonstrated a template-assisted method for fabricating large-scale waveguide perovskite crystals with arbitrary predefined geometries. As illustrated in Fig. 9f, a PDMS mold is created by polymerizing on a GaAs master template, transferred to a new substrate, and filled with a perovskite solution under cooled conditions to control crystal growth. Heating facilitates solvent evaporation and crystallization, after which the mold is removed. SEM images show that different predesigned crystal geometries with a height of 600 nm can be achieved. This crystallization method reduces production costs and process complexity by eliminating the need for atomically flat substrates or complex chemical treatments. The precision and shape of the template design are crucial to the final morphology of the crystals, and inaccuracies or defects in the template can lead to uneven crystal growth or shape deviations. However, the process of fabricating and using templates remains relatively complex, particularly when dealing with large-area or uniquely shaped crystals. Additionally, residual material from template removal may affect crystal quality and device performance.

Another approach is the surface dewetting-assisted method, which utilizes hydrophobic substrate treatment to induce patterned perovskite crystallization [244, 259, 260]. During annealing or solvent evaporation, surface dewetting causes phase separation, forming discrete droplets or isolated patches [261]. Wang et al. [244] reported this method by treating a SiO_2 substrate with octadecyltrichlorosilane (OTS) to create hydrophilic and hydrophobic regions,



enabling controlled nucleation and growth of perovskite crystals (Fig. 9g). The nucleation and growth of PbI_2 microplates were precisely controlled in pre-defined patterns. Xu et al. [245] further advanced this technique by combining space confinement with antisolvent-assisted crystallization (SC-ASC) to fabricate high-quality perovskite single-crystal arrays on-chip. The SC-ASC method, which integrates

traditional photolithography with antisolvent crystallization, achieves precise control over crystal position, size, and orientation, enabling high-resolution arrays with tunable pixel dimensions (ranging from 2 to 8 μm) and less than 10% positional deviation (Fig. 9h). This method allows flexible pattern customization and uniformity across large arrays (Fig. 9i). However, the need for hydrophobic substrate

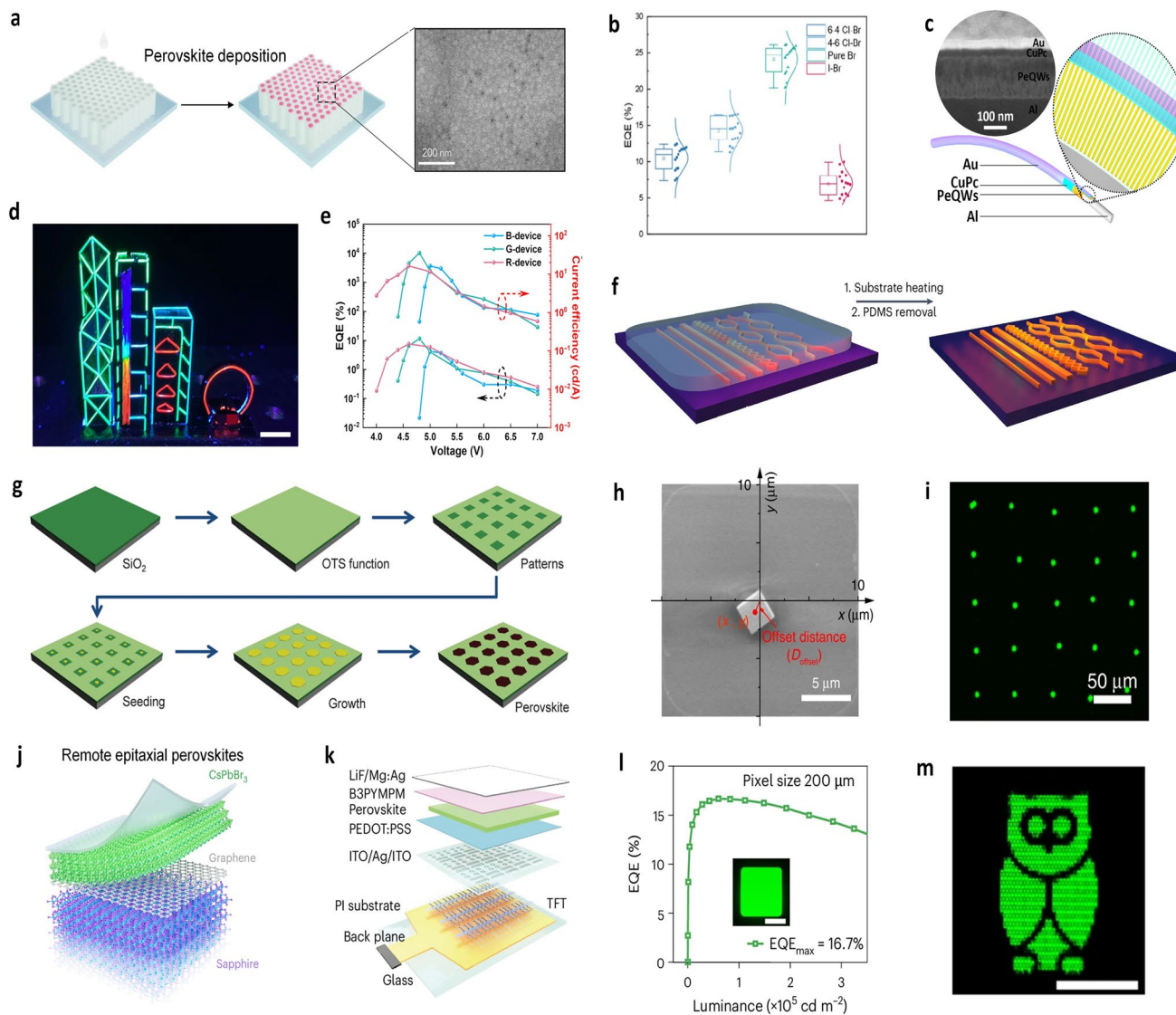


Fig. 9 Crystal growth strategy for PeLEDs. **a** Scheme of perovskite growth process on PAM template. **b** Distribution of EQEs of four different colored PeQWs-based LEDs [57]. **c** Schematic structure images of Fi-LEDs fabricated by crystal growth using PAM template. **d** Three colors of Fi-LEDs with freestanding structures. **e** EQE–current efficiency–voltage curves of RGB LED devices [163]. **f** Schematic illustration of patterned perovskite structures by the pseudomorphosis under decreased crystallization rate [243]. **g** Scheme of perovskite crystallization on a hydrophobically treated substrate [244]. **h** Coordinate system of a perovskite microplate in a hydrophilic window. **i** Fluorescence micrograph of MAPbBr_3 array under UV light [245]. **j** Schematic of perovskite epitaxial growth on a sapphire substrate with a graphene interlayer, resulting in a freestanding film after release. **k** A schematic of an active-matrix perovskite micro-LED display. **l** EQE–luminance curves of perovskite micro-LEDs with 200 μm pixel size. **m** Static images displayed by perovskite micro-LEDs (scale bar: 2 mm) [246]

treatment adds complexity, and achieving uniform crystallization remains a challenge.

In addition to surface confinement, remote epitaxy growth has been proposed as an innovative technique. Wu et al. [246] introduced a sub-nanometer graphene interlayer between the substrate and the perovskite film to achieve remote epitaxial growth with relaxed strain (Fig. 9j). By remote epitaxial crystalline, large-area (400 mm^2), grain-boundary-free, and purely epitaxially oriented perovskite films were fabricated, which were seamlessly integrated into micro-LED devices. Photolithography was employed to isolate each perovskite pixel, approaching $10\text{ }\mu\text{m}$ resolution without detectable damage. Figure 9k illustrates the device configuration, where thin-film transistors control each perovskite pixel independently. These devices demonstrated a high EQE of 16.7% with $200\text{ }\mu\text{m}$ pixels and a brightness of $4.0 \times 10^5\text{ cd m}^{-2}$, with a minimum pixel size of $4\text{ }\mu\text{m}$ (Fig. 9l). The display can show static images with uniform brightness (Fig. 9m), demonstrating a promising route for achieving high-performance micro-LED displays with fine resolution and exceptional brightness.

3.2.2 Inkjet/EHD Jet Printing

Inkjet printing offers a versatile and scalable approach for depositing perovskite patterns at desired locations, making it suitable for LED displays and large-area devices fabrication [155, 262–266]. Unlike conventional deposition methods, inkjet printing generates minimal material waste, as the precursor ink is directly ejected from the nozzle onto the target substrate, where in situ crystallization occurs to form a perovskite film or pattern [267]. Additionally, inkjet printing is compatible with flexible electronics, facilitating the fabrication of bendable or foldable devices [150]. Inkjet printing enables layer-by-layer printing, allowing for the precise assembly of multiple device components, from LED electrodes to the perovskite emission layer [268]. There are 2 main droplet generation mechanisms in inkjet printing: continuous inkjet (CIJ) printing and drop-on-demand (DOD) inkjet printing [269]. In CIJ printing, a liquid column is formed under pressure and breaks apart into a continuous stream of droplets due to Rayleigh–Plateau instability (Fig. 10a) [270].

CIJ printing faces the risk of ink contamination due to the recycling of droplets and has lower resolution [271].

In contrast, DOD printing utilizes a printhead connected to an ink reservoir, where small, precise droplets (ranging from 1 to 100 picoliters) are ejected on demand via thermal or piezoelectric actuation [272, 273]. Miniaturized thermal DOD systems are commonly used for home applications, while piezoelectric DOD systems are preferred in industrial and material science printing due to their ability to modulate droplet volume and velocity. This is essential for stable, high-frequency inkjet processes with consistent droplet sizes. Most studies on inkjet printing for perovskite devices also use piezoelectric DOD systems, where the piezo actuator deforms to modulate chamber pressure, ejecting ink filaments via nozzle contraction [270, 274, 275]. Droplet detachment occurs through “end-pinching,” which is influenced by the rheology of the fluid, governed by the Ohnesorge, Reynolds, and Weber numbers [276, 277]. The governing dimensionless groups are the Reynolds number $Re = \rho v L / \mu$, the Weber number $We = \rho v^2 L / \sigma$, and the Ohnesorge number $Oh = \mu / \sqrt{\rho \sigma L}$ [278, 279]. Neck thinning and final pinch-off follow capillary–inertial scaling at low Oh and viscous–capillary scaling at high Oh , while stable DOD operation is typically achieved in transitional windows of Re and We with $Oh \approx 0.1$ – 1 [269]. A stable end-pinching process requires fluid properties within transitional regimes, balancing viscosity, surface tension, and inertial forces.

Since the pioneering works of Hermerschmidt et al. (2020) using ionic perovskite precursors and Li et al. (2020) employing pre-synthesized PeQDs in inkjet printing formulations, both strategies have been established as fundamental approaches for PeLED fabrication [146, 283]. However, a major challenge in inkjet printing is the “coffee ring” effect, which impacts film morphology and uniformity. When a droplet is deposited onto a wetting substrate, solvent evaporation occurs more rapidly at the edges than at the center, generating microflows that transport solutes to the droplet perimeter. As the solvent continues to evaporate, solute accumulation at the edges leads to ring-like deposition patterns, deteriorating film uniformity and device performance.

To address this issue, many studies have focused on multisolvent strategies to suppress the coffee ring effect and improve PeLEDs performance. As shown in Fig. 10b, Xiao et al. [162] developed highly efficient, flexible, and large-area PeLEDs via inkjet printing by engineering surface wettability and optimizing solvent compositions. A double-hole transport layer and a wetting interface layer (PVP) were introduced to improve substrate wettability,



while a binary-solvent system was employed to suppress solute migration and weaken capillary flow, thereby minimizing coffee ring formation. Additionally, the binary-solvent system created a surface tension gradient that induced recirculating Marangoni flow, leading to more uniform solute deposition. This approach enabled the fabrication of inkjet-printed PeLEDs with an EQE of 14.3%, representing a significant improvement in efficiency. The method also demonstrated scalability, achieving a uniform 2800 mm² PeLED device and patterned structures such as the "USTC" logo. Further advancing ink formulation, Zeng et al. [157] introduced a ternary halogen-free solvent ink composed of naphthane, n-tridecane, and n-nonane, significantly improving printing performance and film quality compared with a binary-solvent system (Fig. 10c). The Marangoni effect, as a key interfacial phenomenon governing fluid dynamics in inkjet-printed droplets, deserves further elaboration on its intrinsic mechanism and regulatory pathway in PeLED fabrication [284]. Specifically, the surface tension gradient driving this effect originates from the differential evaporation rates of multi-solvent components: in binary systems, the preferential volatilization of the low-surface-tension solvent creates a local surface tension discrepancy between the droplet edge and center [285, 286]. This gradient triggers Marangoni flow directed from the low-tension edge to the high-tension center, which counteracts the outward capillary flow that dominates coffee ring formation—effectively redistributing aggregated PeQDs from the droplet periphery back to the bulk, thereby homogenizing the film [157]. Notably, the intensity of Marangoni flow is synergistically determined by the surface tension difference between solvents ($\Delta\gamma$) and their volatility mismatch (ΔP_{vap}): a larger $\Delta\gamma$ enhances the driving force, while an appropriate ΔP_{vap} avoids overly rapid gradient decay caused by excessive volatilization of the low-surface-tension component [287]. In ternary halogen-free systems like naphthane/n-tridecane/n-nonane inks, the introduction of a third solvent enables more precise modulation of this effect: by tuning the content of n-nonane, the dynamic evolution of the surface tension gradient can be regulated—slowing the rate of gradient establishment to match the PeQD crystallization kinetics, and preventing the attenuation of the Marangoni effect due to premature depletion of volatile components [21, 288]. By using this ternary solvent system, green PeLEDs exhibited an EQE of 8.54% and a maximum brightness of 43,883.39 cd m⁻², far surpassing the binary solvent-based PeLEDs (EQE of 2.26%)

(Fig. 10d). Moreover, this approach enabled the fabrication of high-resolution perovskite arrays with feature sizes as small as 45 μm (Fig. 10e).

Beyond the major challenge, coffee-ring effect, micro-scale patterning introduces additional constraints. Even when using 1–3 pL nozzles, the printed wet spot typically spans about 10–30 μm on low-energy surfaces, and subsequent spreading and merging push the practical pixel pitch above roughly 20–30 μm unless strong confinement is applied [289]. Serial, pass-by-pass printing also accumulates overlay errors across HTL, perovskite emission layer, and ETL stacks, and these errors become more pronounced on banked topographies [290]. Hygroscopic precursors and fast-evaporating co-solvents can crystallize at the nozzle tip, intermittent printing aggravates this crusting [275, 291]. These interactions swell seals or leave metal-salt residues that poison the orifice, which causes drift in drop volume and velocity and ultimately degrades pixel fidelity, yield, and device metrics.

EHD jet printing is an advanced printing technique capable of producing high-resolution perovskite patterns beyond the limits of conventional inkjet printing [292–297]. Unlike inkjet printing, which relies on thermal or piezoelectric mechanisms, EHD jet printing utilizes an electric field established between the nozzle and the substrate to control ink ejection and deposition [295, 298]. When the field is sufficiently strong, the liquid meniscus at the nozzle tip deforms into a Taylor cone [299, 300]. At the cone apex, the normal Maxwell stress ($\propto \epsilon E^2$) counterbalances surface tension ($\propto \gamma/R$), and once the electric pressure exceeds the capillary pressure, a thin, charged microjet or a train of fine droplets is emitted [301, 302]. In the steady cone-jet regime, the stability limit is fully determined by the surface tension γ , the electric stress ϵE^2 and 2 geometric parameters of the droplet, so the printed droplet size or line width is set primarily by field strength and ink properties rather than by the nozzle size [303, 304]. As a result, features 10–100 \times smaller than the nozzle are routinely achieved, and perovskite patterns with $\approx 1\ \mu\text{m}$ resolution have been reported [155, 305]. Field-driven scaling further implies that increasing voltage, increasing ink conductivity, and reducing flow rate each tend to shrink the emitted jet or droplet, while viscosity and surface tension set stability windows for maintaining a steady cone-jet (Fig. 10f) [280, 295, 306].

For micro-LEDs with pixel pitches of roughly 10–30 μm , EHD jet printing enables deterministic placement of emitters

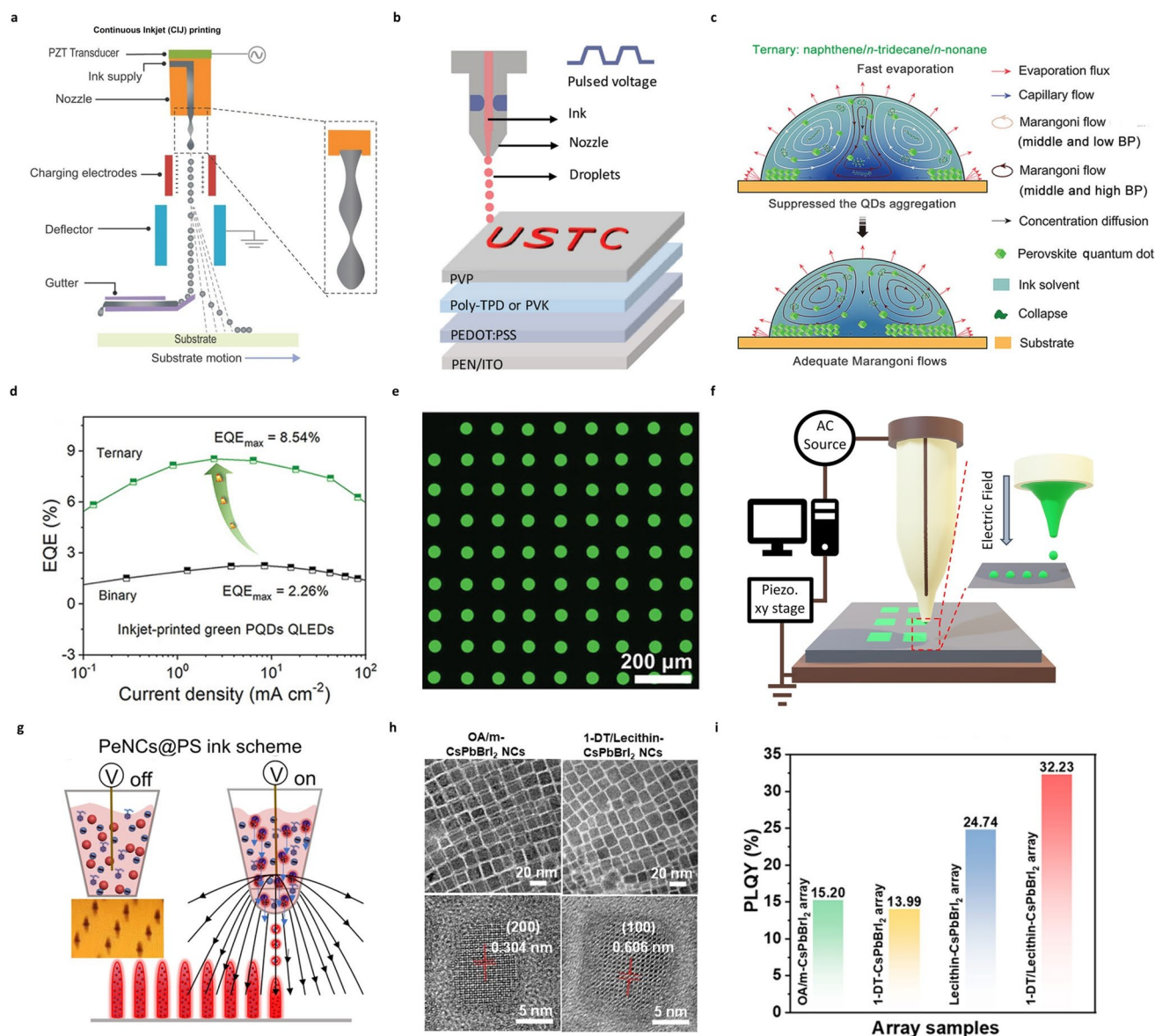


Fig. 10 Inkjet and EHD jet printing strategy for perovskite and light-emitting applications. **a** Schematic diagram of CIJ printing process [270]. **b** Structure setup of perovskite inkjet printing system [162]. **c** Schematic illustration of solvent flow, evaporation process, and self-assembled formation. **d** EQE of an inkjet printed PeLED based on ternary ink [157]. **e** PL spectra of PeQDs patterns [155]. **f** Structure illustration of EHD jet printing system [280]. **g** Schematic illustration of printed PeNC arrays from PeNCs@PS ink. The inset displays microscope of printed arrays [281]. **h** TEM images of PeNCs with corresponding ligands. **i** PLQY of OA/m-CsPbBr₂ array, 1-DT-CsPbBr₂ array, lecithin-CsPbBr₂ array, and 1-DT/lecithin-CsPbBr₂ array [282]

with narrow linewidths and sharp edges, which suppresses optical and electrical crosstalk across pixel boundaries [297]. The electrically focused ejection reduces splashing and coffee-ring artifacts because the charged cone jet and electric field guided landing in EHD lower the effective Weber number We and suppress rim detachment, thereby improving uniformity within confined apertures [307, 308]. Because

the electric field lines can be shaped by the device stack and biasing, EHD jet printing remains effective over passivation layers and mildly nonplanar topographies, aiding integration with dielectric isolation and thin-film interconnects. At the nanoscale, the decoupling of feature size from nozzle diameter allows sub-micrometer emitters, nanoribbons, and dense interconnects to be written directly, supporting

ultra-high-PPI layouts and compact pixel apertures for next-generation displays and on-chip optoelectronics.

The earliest report of EHD jet printing can be traced back to 2007, showcasing the feasibility of using electric fields to generate droplets significantly smaller than nozzle diameters [292]. This capability enables ultra-fine perovskite patterning, making EHD jet printing a promising alternative for high-resolution optoelectronics. To enhance brightness and environmental stability in PeNC arrays, Chen et al. introduced a novel EHD-printed colloidal ink composed of PeNCs mixed with polystyrene (PS) in a nonpolar xylene solvent [281]. The addition of PS significantly improved the brightness and stability of red light-emitting PeNC arrays, addressing common issues such as low emission intensity and degradation. Unlike traditional 2D perovskite microdisk arrays, this study demonstrated the formation of 3D PeNC micropillars (Fig. 10g), which enhanced color conversion efficiency and brightness, making them highly effective as color conversion layers (CCLs) for micro-LED displays. Another advancement in EHD jet printing was made by Yang et al. [282] who developed a dual-ligand strategy to enhance the stability of PeNCs and reduce ion migration during the printing process. By incorporating lecithin and dodecanethiol (1-DT) as dual ligands, they significantly mitigated spectral shifts and emission instability caused by electric field-induced ion migration. Figure 10h compares OA/m-CsPbBr₂ NCs, which exhibited irregular morphologies and weaker crystallinity, to dual-ligand CsPbBr₂ NCs, which displayed well-defined square shapes, enhanced uniformity, and superior PLQY (Fig. 10i).

Despite its advantages, EHD jet printing faces several challenges when applied to PeLED fabrication. One major limitation is the high viscosity and low electric field sensitivity of typical electrode materials, which hinders the formation of stable and uniform jets, leading to inconsistent deposition. These issues make it difficult to achieve the precise electrode patterns necessary for optimal LED performance [309]. Additionally, poor adhesion between the printed patterns and electrode materials can lead to delamination or peeling during device operation, disrupting charge transport and increasing electrical resistance. This not only degrades the LED efficiency and operational lifespan but also compromises overall device stability and reliability [310]. Beyond these materials and interface constraints, the intrinsic serial, pixel-by-pixel nature of EHD jet printing limits raw throughput when scaling to large areas because each pixel must be

written individually [311]. To mitigate this scalability bottleneck, multi-nozzle arrays have been introduced to boost print speed. However, close-packed nozzles can suffer electric-field crosstalk that deflects jets and produces non-uniform deposition [312, 313]. To counter crosstalk, researchers have optimized nozzle layouts and implemented auxiliary electrodes around nozzle arrays to electrostatically shield and stabilize the jets to enhance pattern fidelity and consistency [314–317]. Consequently, while EHD jet printing holds promise for high-resolution perovskite patterning, further advancements are needed to optimize material formulations and deposition techniques to enhance its applicability for PeLED manufacturing.

4 Summary and Perspectives

MHPs have attracted extensive attention in both academia and industry due to their outstanding optoelectronic properties, including high color purity, narrow emission linewidth, high photoluminescence quantum efficiency, and excellent defect tolerance. As a promising candidate for next-generation display technology, PeLEDs offer significant advantages such as a wide color gamut, excellent contrast, and cost-effective fabrication. This review systematically explores perovskite film fabrication and patterning strategies for high-performance PeLEDs, classifying them into top-down and bottom-up approaches. In the top-down approach, methods such as photolithography, laser/e-beam lithography, and nanoimprinting enable precise pattern formation on perovskite films, often using PDMS molds or lithographic techniques. Conversely, the bottom-up approach focuses on precise control of perovskite crystal growth at the atomic or molecular level. Additive manufacturing strategies also provide greater flexibility in device geometry design and potential applications, enabling three-dimensional perovskite structures [318–320].

To realize their full potential in large-area, high-resolution displays, the scalable fabrication of uniform, high-quality perovskite thin films remains a foundational requirement. Although spin-coated small-area PeLEDs have demonstrated high external quantum efficiencies and operational lifetimes, large-area devices still lag behind due to challenges in achieving uniform thickness, controlled crystallization, and interfacial consistency across

wide substrates [40]. Additionally, variations in film morphology and defect densities can significantly impact device reproducibility and long-term performance. To address these limitations, future research may focus on developing deposition techniques that combine scalability with fine control over film formation dynamics—such as solvent-engineered blade-coating, slot-die coating with in situ crystallization control, and hybrid vacuum-solution methods [321–323]. Moreover, integration with flexible or unconventional substrates requires careful interfacial engineering to maintain mechanical reliability and optoelectronic performance.

Building upon this, high-resolution patterning technologies are also essential for achieving full-color PeLED displays. While perovskite materials offer excellent optoelectronic properties, their inherent instability under common photolithographic and laser-based processing conditions (e.g., exposure to heat, UV light, and moisture) presents challenges in fabricating high-quality RGB microarrays with optimal optical and electrical performance [324]. Inkjet printing, particularly with multi-nozzle configurations, is a promising technique for fabricating full-color PeLEDs. However, differences in the crystallization kinetics of the 3 RGB perovskite precursors complicate uniform patterning. Thermal evaporation, a mature and scalable process, is typically restricted to single-color pixelation due to its limited compositional control. In contrast, emerging techniques such as EHD jet printing, nanoimprinting, and transfer printing offer high-resolution, non-destructive patterning strategies. EHD jet printing achieves submicron scale resolution governed by electric-field-driven droplet formation. Nanoimprinting and transfer printing, on the other hand, bypass optical and solvent-related resolution limits and are particularly advantageous for preserving film integrity. Furthermore, transfer printing enables decoupling of film crystallization and pixel alignment, offering a practical route for multicolor integration on both rigid and flexible substrates. Utilizing these approaches, it will be possible to fabricate high-precision, full-color perovskite displays with excellent optical and electrical properties.

Despite progress in efficiency optimization, the long-term operational stability of PeLEDs remains a critical bottleneck. Compared to commercial OLEDs, which exhibit high EQE (> 30%) and operation lifetimes exceeding 100,000 h, PeLEDs currently demonstrate much shorter lifetimes, often

only a few hours—far below commercial viability [22, 325]. As demonstrated in perovskite photovoltaics, the instability of PeLEDs is closely linked to factors such as halide ion migration, interfacial degradation at charge transport layers, and the intrinsic chemical instability of the perovskite material [326–328]. However, PeLEDs operate under higher electric fields and harsher conditions (e.g., elevated temperatures), leading to degradation mechanisms more complex than those observed in photovoltaic devices [329]. Therefore, stability-enhancing strategies that are successful in photovoltaics may not directly translate to PeLEDs. To mitigate instability, one promising approach is the development of core/shell heterostructures, where the perovskite emitter is encapsulated within a wide-bandgap matrix, effectively passivating surface defects and suppressing trap-induced ion migration [330]. For instance, Kim et al. [26] reported BPA-capped core/shell PeNCs with a projected device half-life exceeding 30,000 h at 100 cd m⁻². Additionally, Guo et al. introduced dipolar molecular stabilizers to suppress field-induced phase degradation in α -FAPbI₃ PeLEDs, achieving operational stability over 660,000 h [331]. These developments highlight the feasibility of overcoming intrinsic instability in perovskite emitters.

In addition to stability challenges, concerns over lead (Pb) toxicity have motivated ongoing efforts to develop lead-free or low-lead perovskite alternatives [332–335]. A promising strategy involves substituting Pb²⁺ with alternative B-site cations such as Sn²⁺, Ge²⁺, Cu⁺ (usually represented as Cs₃Cu₂X₅, CsCu₂X₃, and Cu-I cluster hybrids) or Bi³⁺ to mitigate environmental and health risks [336–342]. However, lead-free perovskites currently exhibit lower photoelectric conversion efficiency, reduced stability, and inferior processability compared to Pb-based counterparts [334, 339, 343]. Moreover, Sn²⁺, a commonly used substitute, readily oxidizes to Sn⁴⁺, accelerating device degradation and limiting performance [344]. To bridge this gap, future research may investigate correlations between structural, optical, and charge transport properties in lead-free perovskites, supported by theoretical predictions and compositional screening. At the same time, Pb risk may be managed on two fronts: preventing release during use and recovering Pb in a closed loop from fabrication and end-of-life. Firstly, encapsulation with Pb-sequestering layers (e.g., ionogel sorbents; mesoporous “cage-trap” adsorbents) can immobilize leaked Pb under impact/weathering—critical because unencapsulated devices can leach > 60% of their Pb within minutes



of rain exposure [345]. Secondly, closed-loop recycling proven in PSCs could be ported to PeLED pilot lines: cation exchange/adsorbent capture of Pb from process solvents with conversion back to 99% purity PbI_2 ; benzoic acid-assisted routes that recover > 98–99% Pb from modules or precursor mixes, and demonstrated reuse of recovered PbI_2 in high-efficiency devices (> 20%) [346, 347]. Practically, pilot lines should add in-line Pb monitors and secondary containment, segregate Pb-bearing waste streams for adsorption/recovery, and qualify barrier stacks for Pb capture alongside moisture/oxygen ingress tests [348].

Overall, the convergence of high-quality film deposition, precise and scalable patterning, stability enhancement, and material sustainability will define the roadmap toward practical, full-color perovskite light-emitting display technologies.

Acknowledgements This work was supported by 14th Five-Year Plan Key R&D Plan, Ministry of Science and Technology of the People's Republic of China, 2024YFB3409002, National Natural Science Foundation of China, 12302142, HKUST-HKUST(GZ) Collaborative Research Scheme, G035, Yangcheng Scholars Research Project-Leading Talent Training Project, 2024312156, Guangzhou-HKUST(GZ) Joint Funding Scheme, 2023A03J0157, Guangzhou Basic and Applied Basic Research Project, 2024A04J4765, Shenzhen Basic Research Project, JCYJ20220530114417040.

Author Contributions Shuaiqi Liu and Hao Jiang helped in (conceptualization: equal; data curation: equal; formal analysis: equal; investigation: equal; methodology: equal; resources: equal; software: equal; visualization: equal; writing—original draft: equal); Jizhuang Wang was involved in (investigation: supporting; methodology: supporting; project administration: supporting; writing—review & editing: supporting); Li Liu helped in (funding acquisition: supporting; project administration: supporting; writing review & editing: supporting); Zhiwen Zhou contributed to (conceptualization: lead; investigation: lead; methodology: lead; project administration: lead; writing—review & editing: lead); Mojun Chen helped in (conceptualization: lead; project administration: lead; supervision: lead; validation: lead; writing—review & editing: lead).

Declarations

Conflict of Interest The authors declare no conflict of interest. They have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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