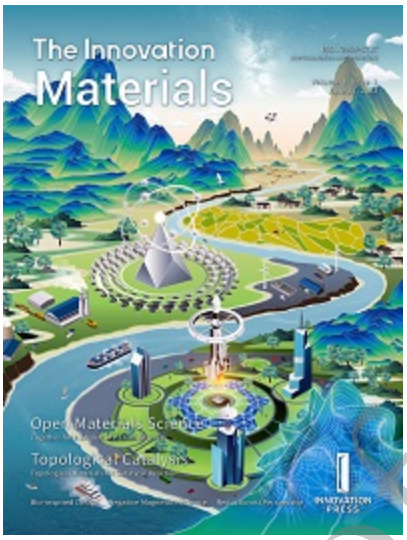

Polymerization-orchestrated crystallization for efficient blue PeLEDs

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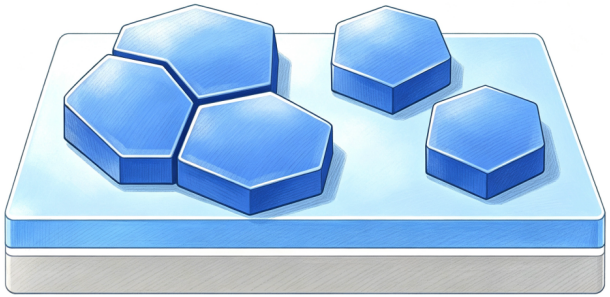
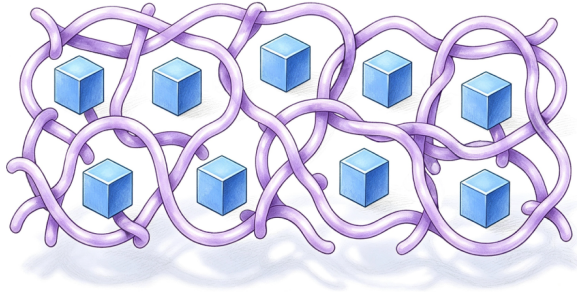
Manuscript Type:	Commentary
Keywords:	Blue perovskite LEDs, Polymer-assisted crystallization, Ligand engineering

Pristine film



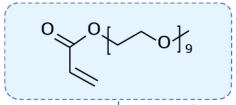
- Large grains (> 250 nm)
- Stronger e-ph coupling
- Orthorhombic phase

OEGA in situ polymerization



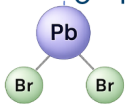
- Small nanocrystals (11±3 nm)
- Weaker e-ph coupling
- Cubic phase

1 Precursor coordination



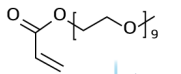
OEGA

O•••Pb coordination



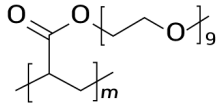
$PbBr_2$

2 Thermal annealing



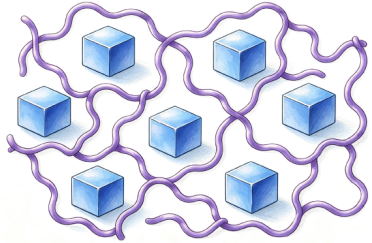
OEGA

(AIBN 60 °C)

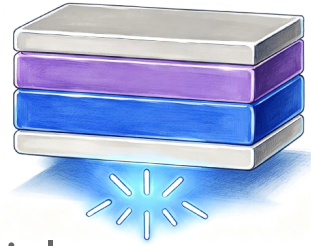


POEGA

3 Polymer-network confinement



4 Efficient blue PeLEDs



Grain size:
11 ± 3 nm

PLQY:
83% with
PEA passivation

EQE:
21.8%
@ 491 nm

T_{50} :
69.4 min
@ 100 cd m^{-2}

Polymerization-orchestrated crystallization for efficient blue PeLEDs

Metal halide perovskites have emerged as promising emitters for light-emitting diodes (LEDs), owing to their tunable bandgaps, narrow emission linewidths, high colour purity and compatibility with low-temperature solution processing. Green and red perovskite LEDs (PeLEDs) have already achieved external quantum efficiencies (EQEs) exceeding 30%, highlighting the rapid progress of perovskite electroluminescence.[1] Blue PeLEDs, by contrast, still lag behind. One important reason is related to a general challenge in perovskite nanocrystals: efficient emission requires nanoscale domains to strengthen carrier confinement and radiative recombination, but also high crystalline quality to suppress defect-mediated non-radiative losses.[2,3] This size-crystallinity balance is relevant to perovskite nanocrystals in general, but it becomes particularly demanding for blue PeLEDs, where wide-bandgap compositions, stronger confinement requirements and defect sensitivity make high-efficiency emission more difficult to achieve.[2,4]

Conventional crystallization-control strategies, including solvent engineering, thermal annealing and additive-assisted crystallization, primarily improve crystallinity by moderating nucleation and facilitating lattice reorganization. However, these approaches often promote grain growth, thereby weakening carrier confinement. Conversely, strong size restriction can produce nanoscale domains, but often introduces structural disorder, surface defects and reduced crystallinity. Developing strategies that simultaneously regulate nanocrystal dimensions and crystallization dynamics has therefore become a central challenge for high-performance blue PeLEDs.[3,4]

Liu and co-workers now report an in situ polymerization-driven nanocrystal confinement strategy that addresses this long-standing trade-off in the in situ synthesis of perovskite nanocrystals.[2] Rather than treating ligands solely as static surface passivators, their work integrates ligand polymerization directly into the crystallization process, enabling dynamic regulation of nanocrystal formation (Fig. 1). The strategy is built upon oligo (ethylene glycol) methyl ether acrylate (OEGA), a polymerizable monomer that serves both as a passivation ligand and as a precursor for an in situ-formed polymer network. Through carbonyl and ether oxygen sites, OEGA coordinates with perovskite precursors, stabilizing intermediate clusters and retarding premature aggregation. Upon thermal annealing, the acrylate groups undergo radical polymerization, forming poly (OEGA) networks that continuously confine crystal growth, thereby enabling the nanocrystals to retain their nanoscale confined dimensions.

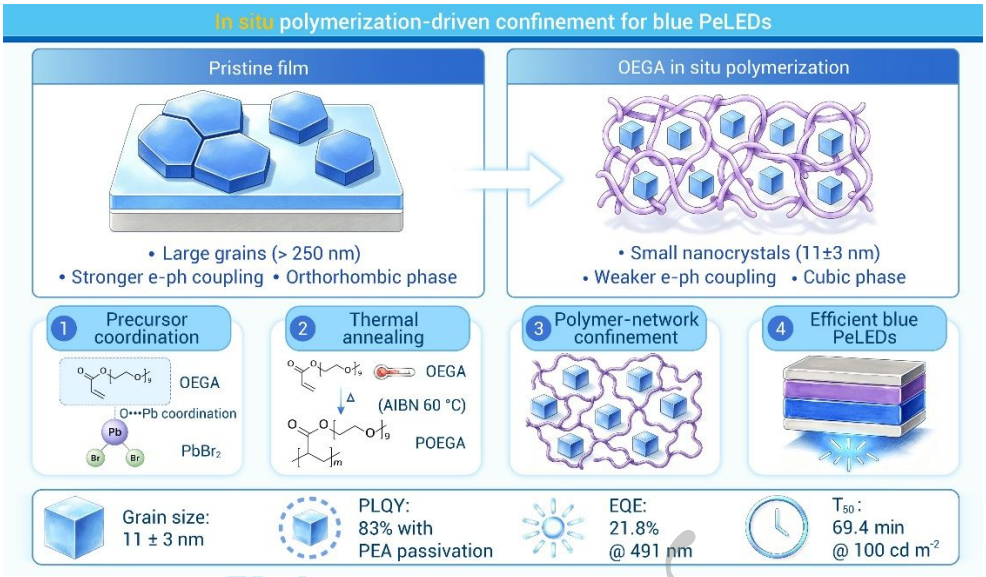


Figure 1. Schematic illustration of OEGA-driven in situ polymerization confinement for blue PeLEDs. Unlike conventional ligand passivation, OEGA participates directly in the crystallization process of perovskite nanocrystals through precursor coordination and subsequent in situ polymerization to form a POEGA network. This dynamic confinement suppresses nanocrystal overgrowth, stabilizes the cubic phase, weakens electron-phonon coupling and enables efficient blue electroluminescence.

A key feature of this strategy is the synchronization of polymer formation with perovskite crystallization, allowing ligand chemistry to actively regulate nanocrystal growth rather than merely passivate the final product. This coordinated sequence reduces the perovskite grain size from more than 250 nm to 11 ± 3 nm, while simultaneously maintaining high crystallinity.[2] A combination of complementary characterizations supports the proposed crystallization pathway. Proton nuclear magnetic resonance confirms the thermal polymerization of OEGA through the disappearance of vinyl proton signals after annealing, whereas X-ray photoelectron spectroscopy reveals electronic interactions between OEGA and the perovskite framework. In situ absorption and photoluminescence measurements further show a delayed onset of perovskite emission and a more gradual optical evolution in the presence of OEGA, consistent with moderated precursor aggregation and prolonged lattice relaxation. Direct visualization by in situ liquid-phase electron microscopy reveals distinct growth behaviours, with pristine precursors rapidly evolving into large aggregates, whereas OEGA-containing precursors remain as stable nanoscale particles throughout the observation period. The results also show that confinement depends strongly on the additive structure. Small polymerizable monomers can polymerize but provide limited spatial restriction; very long polymerizable ligands tend to self-aggregate and lose effective contact with the perovskite

lattice. OEGA falls in a more suitable range, where precursor coordination, polymer-network formation and charge transport can be balanced. This provides a practical guideline for future ligand design: the ligand should not only bind to precursors, but should also polymerize on a timescale that matches perovskite crystallization.

The influence of in situ polymer confinement extends beyond nanocrystal size control to the structural and photophysical properties of the perovskite. High-resolution transmission electron microscopy and X-ray diffraction consistently indicate that OEGA-confined $Cs_{0.7}EA_{0.3}PbBr_3$ nanocrystals adopt a cubic phase at room temperature instead of the orthorhombic phase observed in the pristine film. Because the cubic phase exhibits reduced octahedral distortion, the confined nanocrystals display weaker electron–phonon coupling and consequently reduced thermally activated non-radiative recombination. Polymer confinement therefore modifies not only nanocrystal morphology but also the lattice structure governing excited-state dynamics. Subsequent surface passivation with phenethylammonium bromide further increases the photoluminescence quantum yield to 83% and prolongs the carrier lifetime, illustrating that dynamic crystallization control and conventional surface passivation play complementary roles. This strategy yields perovskite films with confined dimensions, enhanced crystallinity, stabilized phase structure and improved radiative efficiency.

At the device level, these materials advances translate into high-performance blue PeLEDs. The optimized OEGA/PEA devices achieve an external quantum efficiency of 21.8% at an electroluminescence peak of 491 nm, together with a narrow full width at half maximum of 23 nm and CIE coordinates of (0.07, 0.29). Compared with the pristine device, the OEGA/PEA device shows an approximately 2.7-fold increase in maximum luminance and a 6.7-fold extension of operational half-lifetime (T_{50}). More importantly, these improvements are consistent with suppressed ion migration in the polymer-confined films, as reflected by the increase in ion-migration activation energy from 0.27 to 0.77 eV, suggesting that polymer-network confinement contributes not only to nanocrystal growth control but also to improved device robustness. Beyond these performance metrics, the broader significance of this work lies in redefining the role of organic ligands in perovskite synthesis. Here, the ligand is embedded within a reaction pathway that shapes the nucleation, growth, phase evolution and defect landscape of the perovskite nanocrystals.[2,3]

Despite these advances, several important challenges remain. First, high-brightness operation

remains a key limitation for QD/NC-based blue LEDs. Strong nanocrystal confinement and ligand/polymer-rich matrices help preserve blue emission and suppress defects, but they can also impede charge transport, causing charge accumulation, efficiency roll-off and faster degradation at high current density. Future designs must therefore balance confinement, passivation and transport to sustain efficiency at practical luminance.[2,4,5] Second, the reported emission peak of 491 nm corresponds to sky-blue electroluminescence. Moving toward deeper and more saturated blue emission will likely require further bandgap engineering, including chloride-rich compositions or reduced-dimensional architectures, which may introduce additional phase instability and defect-related losses.[4] Third, as with most high-performance PeLEDs, achieving high efficiency, high spectral purity and improved operational stability within the same blue-emitting device remains challenging. The present work makes important progress toward this goal, but further optimization is still needed to address the intrinsic difficulties associated with blue perovskite emission.

Looking forward, this work can be viewed alongside the synthesis-on-substrate strategy for perovskite quantum dot solids reported by Jiang and co-workers.[5] Both approaches aim to form and retain nanoscale emissive units in solid films, but they do so through different routes: the earlier work directly formed ultrasmall $CsPbBr_3$ quantum dot solids on substrates, whereas the present study confines nanocrystal growth through OEGA polymerization during film formation. This comparison suggests that polymerization-driven confinement could be further explored in other blue-emitting perovskite systems, including chloride-rich emitters and reduced-dimensional perovskites.[4] A polymerizable ligand strategy may provide a complementary route to regulate phase purity, defect formation and film transport during crystallization. Another opportunity lies in engineering the polymer network as a multifunctional component. In the present study, the in situ-formed POEGA network confines nanocrystal growth, suppresses ion migration and contributes to improved device performance. Future ligand designs could further tune polymer chain length, coordination geometry and polymerization kinetics to optimize the interplay between crystallization control and charge transport. Such molecular-level design would shift polymer additives from empirical processing aids to programmable components of the perovskite optoelectronic architecture.[2,3] Equally important will be the development of a deeper mechanistic understanding of polymer-mediated crystallization. The current work provides compelling evidence that OEGA coordinates with precursors, delays aggregation and confines nanocrystal overgrowth, but the

precise temporal sequence of coordination, polymerization, nucleation and lattice ordering remains a rich topic for future study. Resolving this sequence would help identify when confinement is most decisive and guide the rational design of next-generation polymerizable ligands with tailored functionality.

In summary, Liu et al. have introduced a compelling strategy for overcoming the crystallinity–confinement trade-off in blue perovskite nanocrystals. By synchronizing in situ polymerization with perovskite crystallization, they transform OEGA from a molecular additive into a dynamic architect of nanoscale order. The resulting improvements in grain-size control, phase stabilization, photoluminescence efficiency and device performance mark an important advance for blue PeLEDs.

REFERENCES

1. Li, Y., Gao, T., He, Z., et al. (2025). Flexible perovskite light-emitting diodes: recent progress, applications and challenges. *npj Flex. Electron.* **9**, 32. DOI:10.1038/s41528-025-00408-5
2. Liu, S., Pols, M., Zhang, Z., et al. (2026). In situ nanocrystal confinement for efficient blue perovskite LEDs. *Nature* **654**, 375-382. DOI:10.1038/s41586-026-10596-3
3. Shamsi, J., Rainò, G., Kovalenko, M. V., et al. (2021). To nano or not to nano for bright halide perovskite emitters. *Nat. Nanotechnol.* **16**, 1164-1168. DOI: 10.1038/s41565-021-01005-z
4. Yuan, S., Dai, L., Sun, Y. et al. (2024). Efficient blue electroluminescence from reduced-dimensional perovskites. *Nat. Photon.* **18**, 425-431. DOI:10.1038/s41566-024-01382-6
5. Jiang, Y., Sun, C., Xu, J., et al. (2022). Synthesis-on-substrate of quantum dot solids. *Nature* **612**, 679-684. DOI:10.1038/s41586-022-05486-3