Metal Halide Perovskite Light Emitting Devices for Optical Communications: A Spotlight on Speed, Brightness, Wavelength Tunability, Bidirectionality, and Integration

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#### **ABSTRACT**

Metal halide perovskites are an emerging class of non-epitaxial semiconductor that have garnered the attention of the photovoltaic and light emitting device (LED) research communities due to their remarkable optoelectronic properties and wide wavelength tunability. In addition to displays and lighting, recent studies have shown the promise of perovskite LEDs (PeLEDs) in other applications such as optical communications. In this Spotlight article, we focus on the uniqueness and recent progress of perovskite light emitters and their applications in optical communications. We spotlight on the progress of their operational speed, light-emitting brightness, wide wavelength tunability, bidirectional communication, and feasibility of monolithic integration with silicon Complementary Metal-Oxide-Semiconductor (CMOS) technology. We believe that metal halide perovskites are an important class of materials that hold the potential to revolutionize the next generation of low-cost, high-performance communication links in both free space and on-chip optical interconnect applications.

### 1. Introduction

The remarkable optoelectronic characteristics and broad wavelength tunability of metal halide perovskites, a relatively new class of non-epitaxial semiconductors, have attracted the interest of the photovoltaic and light emitting device (LED) research communities. <sup>1–6</sup> Over the last ten years, perovskite-based devices have experienced rapid improvement, with solar cells and LEDs surpassing 25% power conversion efficiency and 90% internal quantum efficiency, respectively. <sup>7,8</sup> Furthermore, these materials are also considered as a new class of promising non-epitaxial gain media for lasers, <sup>9–11</sup> especially due to the perception that perovskites feature all the optical and electrical qualities that make practical, high-performance laser sources viable (e.g., low defect

density of  $5 \times 10^{16} \ cm^{-3}$ , balanced electron/hole charge carrier mobility in the range of 10 - 300 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup> depending on specific compositions, high photoluminescence quantum yield approaching 100%, and a high material gain coefficient exceeding 3000 cm<sup>-1</sup>).

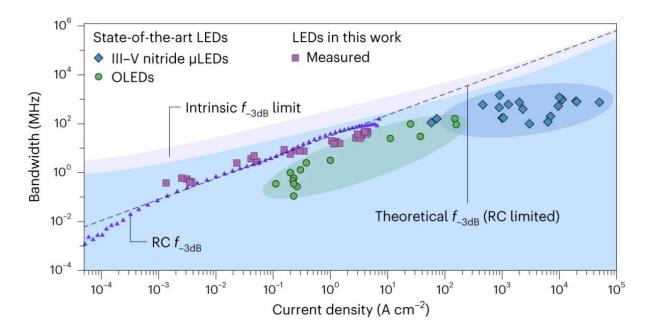
In addition to displays and lighting, recent studies have shown the promise of perovskite LEDs (PeLEDs) in other applications such as optical communications. 12-15 The inherent properties of metal-halide perovskites, particularly their versatility and high optoelectronic quality, make them ideal candidates for this application. Recent studies suggest that PeLED bandwidth can achieve a gigahertz range, exceeding the current state-of-the-art high-speed LEDs. 16 Furthermore, these materials can be solution-processed on various substrates, potentially allowing for easier and more cost-effective manufacturing processes.<sup>17</sup> Their compatibility with low-temperature processing is especially advantageous for integrating light emitters on silicon chips, the workhorse of microelectronics applications, at the backend-of-the-line. Additionally, optical simulation reveals that a significant enhancement in light extraction efficiency, reaching 73.6%, <sup>18</sup> is attainable through a dual-stage light extraction mechanism employing nano-dome light couplers and nanowire optical antennas on the nanophotonic substrate. These findings indicate that the integration of nanophotonic architectures offers a promising strategy for enhancing the performance of perovskite light-emitting diodes and makes them suitable candidate for efficient optical communication.<sup>18</sup> This is particularly relevant in the era of the Internet of Things (IoT), where multiple devices are connected, and there is a growing need for efficient, low-cost communication methods. 19-21

In this Spotlight article, we focus on the recent progress of perovskite light emitters and their applications in optical communications. In particular, we spotlight on the progress of their operational speed, light-emitting brightness, wide wavelength tunability, bidirectional

communication, and feasibility of monolithic integration with silicon Complementary Metal-Oxide-Semiconductor (CMOS) technology. We believe that metal halide perovskites are a new and useful materials platform and hold the potential to revolutionize the next generation of low-cost, high-performance communication links in both free space and optical interconnect applications.

## 2. High Speed Operation

A sufficiently high operational speed, or high modulation bandwidth of a light emitter is a prerequisite for optical communications. Unlike other thin film emitters such as phosphorescent organic molecules and thermally activated delayed fluorescence (TADF) molecules, which are intrinsically slow due to long photoluminescence/excitonic lifetimes, metal halide perovskite emitters can be fast if well-engineered. Recent analysis suggests that the bandwidth of perovskite light emitters may exceed gigahertz levels, although this high speed can only be achieved when current density exceeds 100 A/cm<sup>2</sup> (**Figure 1**). Detailed discussions on high-current operation of PeLEDs can be found in Section 3. Experimentally, however, the bandwidth of perovskite LEDs demonstrated so far is limited to 42.6 MHz (**Figure 1**). Both extrinsic factors (such as parasitic capacitance) and intrinsic factors (such as charge carrier recombination lifetime) can limit the speed of PeLEDs. which is discussed in detail in Section 2.1 and 2.2.



**Figure 1**. Bandwidth versus current density of the record PeLEDs, organic LEDs and III–V nitride micro-LEDs. Solid triangles indicate the calculated  $RCf_{-3dB}$  of PeLEDs. The dashed line shows the theoretical  $f_{-3dB}$  curve of PeLEDs limited by  $\tau_{RC}$ , estimated from the measured results. Reprinted with permission from ref. <sup>16</sup>Copyright [Springer Nature] [2023].

# 2.1. Device Configuration Limited Operational Speed

Perovskite light-emitters are generally composed of multiple functional layers that collaborate to generate and emit light (**Figure 2a**). The device begins with a substrate, which functions as the mechanical supporting structure upon which subsequent layers are fabricated. Notably, perovskite materials are amenable to solution-based processing, allowing for their deposition on a wide range of substrates such as silicon, glass, and flexible plastics. This versatility sets them apart from III-V semiconductor light emitters, which typically require more specialized deposition techniques such as molecular beam epitaxy (MBE) or metalorganic chemical vapor deposition (MOCVD).9 Common methods for depositing perovskites include spin-coating, vapor deposition, and inkjet printing,<sup>22</sup> making them highly adaptable for various applications. On top of the substrate, a common PeLED structure typically consists of a perovskite emissive layer sandwiched between electron and hole transport layers (ETLs & HTLs), which in turn are connected to the anode and cathode electrodes. While applying voltage, electrons from the cathode are injected through the

ETL into the perovskite layer, and holes from the anode are injected through the HTL into the perovskite layer, where they recombine, emitting photons. These ETLs and HTLs are designed to facilitate efficient charge carrier transport and minimize energy barriers at the interfaces.<sup>23</sup>

A typical PeLED structure is shown in **Figure 2a-b**. The overlap between the two electrodes defines the active device area of the PeLED. Resistance and capacitance associated with the device structure inevitably affect the device's operational speed. In essence, a typical equivalent circuit model for an LED can be simplified into a network comprising a diode resistor ( $R_d$ ) and capacitor (C), along with a series resistor ( $R_s$ ), as shown in **Figure 2c.**<sup>12,15</sup> The resistance of the diode displays a nonlinear response to the electric field applied to it. Conversely, the series resistance represents the collective resistance from various sources including the contacts, wiring, and driving circuitry, and is generally considered to be a fixed value. The capacitor aspect of the LED stems from its dielectric properties and the accumulation of charge at the junctions of its different functional layers. Consequently, there's an inherent delay in the LED's response, attributable to the combined effects of the RC time constant, the movement of charges, and the charge carrier recombination activities.

Mathematically, the RC time constant  $\tau_{RC}$  can be written as

$$\tau_{RC} = \frac{C}{\frac{1}{R_d} + \frac{1}{R_s}}$$

The frequency response of an LED is given by

$$f_{-3dB} = \frac{1}{2\pi\tau}$$

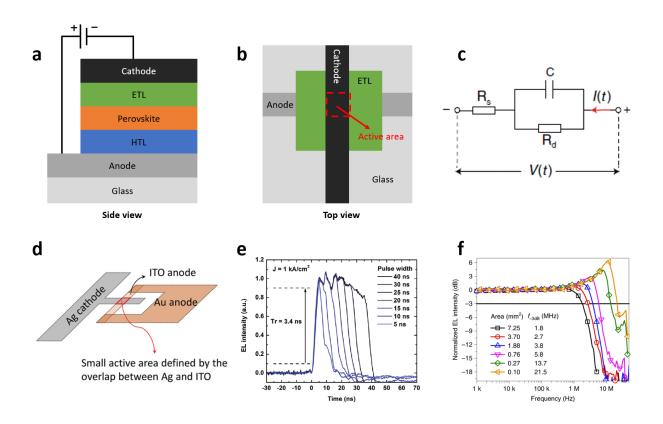
where  $\tau$  could represent either the carrier lifetime  $\tau_c$  (intrinsic factor limited) or  $\tau_{RC}$  (extrinsic factor limited), depending on which factor predominantly limits the modulation performance.<sup>12</sup>

A significant result of the *RC* effects is the delay between the turn-on/turn-off of the driving voltage and the rise/fall of electroluminescence. This delay has a pronounced impact on its ability to modulate emitted light at high frequencies. Essentially, it represents a critical factor influencing the temporal characteristics and modulation capabilities of such light-emitting devices. <sup>13,15,24–26</sup> To mitigate this issue, reducing the associated resistance and capacitance in the device becomes key.

Reducing series resistance. Reducing the series resistance has shown to be an effective way to reduce  $\tau_{RC}$ . For example, Indium tin oxide (ITO), the commonly used transparent electrode in PeLEDs, represents significant series resistance involved in the electrical injection process. An effective strategy is to connect the ITO electrode outside of the device active area to a metal electrode (e.g., Au or Ag) (**Figure 2d**). A high-speed PeLEDs with nanosecond rise time is reported using this device configuration (**Figure 2e**). Another source of significant series resistance comes from the low carrier mobilities of the organic charge transport layers (for example,  $10^{-6}$  -  $10^{-4}$  cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup>). As a result, doping in the charge transport layer is shown to be important in PeLEDs.<sup>27</sup> It will improve the mobility of the carriers and therefore decrease resistance.

**Reducing device area.** Since parasitic capacitance is inversely dependent on device area, reducing the active device area is the most straightforward way to improve the device modulation bandwidth. For example, the 3dB cutoff frequency is increased from 1.9 MHz for a 7.25-mm<sup>2</sup> device to 21.5 MHz for a 0.1-mm<sup>2</sup> (**Figure 2f**).<sup>15</sup>

Increasing operating voltages. Since the device differential resistance decreases with increasing carrier density, another strategy to reduce the  $\tau_{RC}$  is to increase the voltage bias or current density. It has been reported that the bandwidth of PeLEDs increases from 2.1 MHz to 37.6 MHz as the bias increases from 3.5 to 7.5 V.<sup>16</sup>



**Figure 2.** (a-b) A schematic PeLED structure in side-view (a) and top view (b). (c) a typical equivalent circuit model for an LED. Reprinted with permission from ref. <sup>16</sup> Copyright [Springer Nature] [2023]. (d) Alternative PeLED device structure for higher speed operation. Reprinted with permission from ref. <sup>13</sup> Copyright [Wiley] [2021]. (e) Fast electroluminescence rise time of several nanosecond due to reduced *RC* time constant in the device architecture. Reprinted with permission from ref. <sup>13</sup> Copyright [Wiley] [2021]. (f) Bandwidth of PeLEDs with various device area. Reprinted with permission from ref. <sup>15</sup>Copyright [Springer Nature] [2020].

# 2.2. Charge Carrier Recombination Limited Operational Speed

The charge carrier recombination lifetime in perovskites sets the intrinsic speed limitations of PeLEDs. The device cannot be turned on and off faster than the carrier recombination lifetime.

Thus, a faster carrier recombination process enables faster modulation. There are three major

carrier recombination processes in perovskites, assuming that 3D perovskites are not excitonic owing to a small binding energy of <50 meV: (1) first-order defect-assisted recombination, (2) second-order bimolecular radiative recombination, and (3) third-order Auger recombination.<sup>28</sup> The change of carrier density n versus time t can then be described by the rate equation:

$$-\frac{dn}{dt} = -G + An + Bn^2 + Cn^3,$$

where G is the carrier generation rate, and A, B, C are the rate coefficients describing defect-assisted monomolecular recombination, radiative bimolecular recombination, and Auger recombination, respectively. The defect-assisted recombination lifetime can be defined as  $\tau_A = \frac{1}{A}$ , the radiative recombination lifetime as  $\tau_B = \frac{1}{Bn}$ , and the Auger recombination lifetime as  $\tau_C = \frac{1}{Cn^2}$ .

The defect-assisted monomolecular recombination lifetime is constant, while the other two decrease with increasing carrier density. Therefore, one could expect a faster charge carrier recombination rate at high excitation intensity (either optically or electrically). In other words, the intrinsic response speed of PeLEDs will be faster when operating at high current densities.

Perovskite films containing a high nonradiative defect density generally have a short carrier recombination lifetime due to a large defect-assisted monomolecular recombination coefficient A. However, this will lead to low light-emission efficiency and is not desirable for LED applications. Most defect passivation strategies developed so far can reduce the defect-assisted nonradiative recombination process, which will increase the overall carrier recombination lifetime due to longer  $\tau_A$ . However, longer carrier radiative lifetime is not desirable for high-speed PeLEDs. Electrical doping might be a promising strategy that can reduce carrier recombination lifetime by promoting

radiative recombination and shortening  $\tau_B$  and at the same time maintain efficient light emission. The Auger recombination coefficient C is typically on the order of  $10^{-28}$  cm<sup>6</sup>s<sup>-1</sup>,  $^{30-32}$  which is on average 25 times higher than the recombination coefficient for GaAs. This means perovskite light emitters suffer more severe Auger recombination loss at high carrier densities (n>10<sup>17</sup> cm<sup>-3</sup>). Although a higher Auger recombination rate has theoretical benefits to increase the bandwidth at high current injection levels, it impedes PeELDs from reaching high current density operating regimes for high brightness and fast radiative bimolecular recombination.<sup>16</sup>

Notably, despite the charge carrier recombination rate sets the material's intrinsic bandwidth, all reported studies so far have been constrained by extrinsic *RC* factors. Based on experimentally measured *A*, *B*, *C* rate coefficients, a recent study suggested that PeLED bandwidth can achieve a gigahertz range, exceeding the current state-of-the-art high-speed LEDs.<sup>16</sup>

# 3. High Brightness Operation

For optical communications, the signal-to-noise ratio (SNR) of a communication system increases with the power of the carrier signals, and based on Shannon's theory, the capacity of a communication channel increases as well with SNR.<sup>33</sup> Therefore, the brightness (power) of light emitters is a critical property for optical communications.

Despite the rapid improvement in performance,<sup>1</sup> PeLEDs with brightness comparable to inorganic LEDs have yet to be reported. One source of this difficulty is that the external quantum efficiency (EQE) of PeLEDs typically decreases at high current densities (or brightness), an effect known as EQE roll-off. Factors that contribute to EQE roll-off include Joule heating,<sup>34</sup> charge imbalance,<sup>35</sup> Auger recombination,<sup>36</sup> and electrical field-induced quenching.<sup>37</sup> Furthermore, operational lifetime decreases dramatically at high current densities,<sup>38</sup> making it difficult to achieve both high

brightness and long lifetime. Nevertheless, the ability to maintain operation at high current density is a prerequisite for PeLEDs to achieve high brightness. **Figure 3** shows the timelines for the maximum current densities of PeLEDs achieved over the years.<sup>39–50</sup> Notably, driven by nanosecond electrical pulses, the maximum current density of PeLEDs has exceeded 20 kA/cm<sup>2</sup>,<sup>41</sup> and the maximum current density, driven continuously, has achieved 25 A/cm<sup>2</sup>.<sup>39</sup> These reports show the promise for high-power PeLED operations. The following strategies have been demonstrated to be effective to improve the brightness and stability of PeLEDs, particularly operating at high current densities:

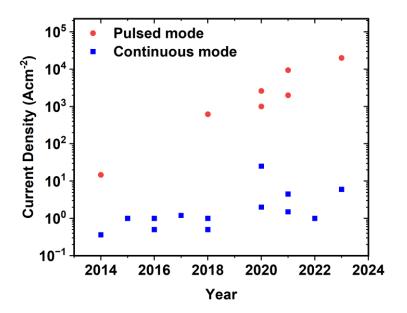
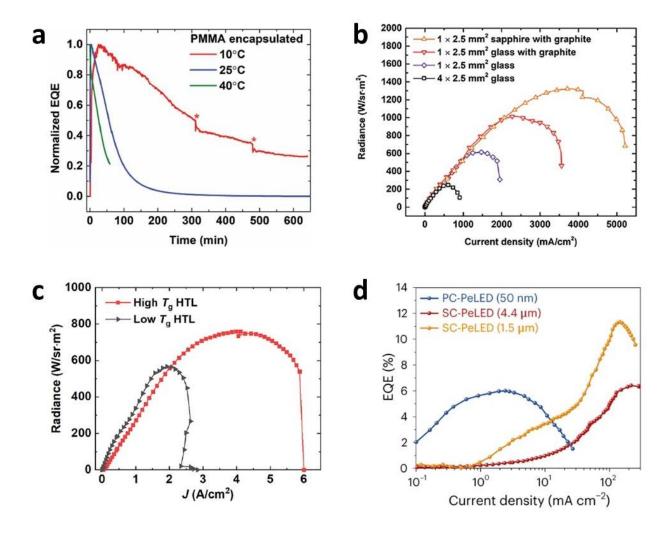


Figure 3. Timelines showing maximum current density for PeLEDs, driven continuously or by electrical pulses. Data adopted from literature. 37,39–50

**Thermal management.** It has been reported that PeLEDs are sensitive to temperature, mainly due to the ionic nature of perovskites. For example, the lifetime of PeLEDs decreases by an order of magnitude when the environmental temperature increases from 10 to 40 °C (**Figure 4a**). Hence, thermal management is crucial to achieve a stable device performance. Several strategies have been demonstrated to be effect in controlling heat in PeLEDs, such as the use of heat sinks and

thermally conductive substrates. It has been reported that using a thermally conductive sapphire substrate and a graphite as a heat spreader significantly improves device performance in the high injection level (**Figure 4b**).<sup>27</sup> Furthermore, utilizing doped charge transport layers can be instrumental in enhancing electrical conductivity. <sup>34</sup> Consequently, it reduces Joule heating in PeLEDs.<sup>34</sup> In addition to better heat dissipation, another strategy to support high-power high-brightness operation of PeLEDs is to improve the thermal stability of PeLEDs, such as adopting more thermally stable organic charge transport layers in PeLEDs (**Figure 4c**).<sup>51</sup>

Reducing Auger recombination. Efficient perovskite light emitters typically adopt polycrystalline perovskite films with nanometer sized grains or low-dimensional perovskites.<sup>52</sup> These materials exhibit a higher exciton binding energy (E<sub>b</sub>) compared to bulk 3D perovskites due to the influence of quantum and dielectric confinement.<sup>53</sup> These features result in severe Auger recombination in polycrystalline based PeLEDs and limit their maximum attainable brightness. To address this issue, in a recent report, relatively thick single-crystal (SC) perovskite thin films are used as the light emission layer in PeLEDs.<sup>36</sup> At high current densities, the 4.4-μm thick SC PeLEDs exhibit significantly reduced EQE roll-off compared to polycrystalline PeLEDs (**Figure 4d**), and the authors attribute this improved performance to reduced Auger recombination.



**Figure 4.** (a) Operational stability of PeLEDs working at various environmental temperatures operating at a constant current density of 10 mA/cm<sup>2</sup>. Reprinted with permission from ref.<sup>38</sup> Copyright [Wiley] [2020]. (b) Radiance of PeLED with various thermal management strategies. Reprinted with permission from ref.<sup>34</sup> Copyright [Wiley] [2020]. (c) Radiance of PeLEDs using polymer HTLs with different glass-transition temperatures. Reprinted from ref.<sup>54</sup> Copyright [2023] American Chemical Society. (d) Efficiency of PeLEDs with single-crystal or polycrystalline perovskite emission layers. Reprinted with permission from ref.<sup>36</sup> Copyright [Springer Nature] [2023].

Improving Electron-Hole Balance. The transport layers should ensure a balanced injection of both holes and electrons into the emissive layer.<sup>55</sup> This balance is important for maximizing quantum efficiency. However, an uneven injection of electrons and holes into the perovskite layer is fairly common due to different energy barriers at interfaces in the device stack.<sup>56</sup> For example, electrons from common electron transport materials like 2',2'-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) can easily enter the perovskite layer due to a favorable energy level

alignment, while holes struggle to do the same because of an energy mismatch between the HTL and perovskite layer. This imbalance leads to inefficiencies in PeLED performance. Furthermore, the energy level difference at the HTL-perovskite interface can quench excitons before they emit light. This quenching effect is particularly significant for green and blue-emitting PeLEDs. To mitigate this issue, several attempts have been made. One strategy is to introduce energy barriers to reduce electron injection, which consequently improves the device performance and helps to achieve a peak external quantum efficiency (EQE) of 16.7%.<sup>57</sup> Alternatively, strategies have been proposed to enhance hole injection.<sup>58–60</sup> It is evident that device efficiency can be enhanced by selecting HTLs with proper highest occupied molecular orbital (HOMO) energies that minimize the energy barriers between HTLs and perovskites.<sup>55</sup> When an energy barrier is inevitable, building a stepwise "energy ladder" with multiple HTLs (e.g., inserting additional HTLs of TFB and PVK between PEDOT:PSS and perovskites) is favorable to enhance hole.<sup>55</sup> This strategic choice helps to mitigate carrier imbalance issues, ensuring a more balanced injection of electrons and holes, ultimately leading to improved device performance and efficiency.

Improving the Morphology of Perovskites: The surface morphology of perovskite films plays a crucial role in achieving optimal performance in PeLEDs, as it directly influences their optical, electrical characteristics, and overall device efficiency. Nonuniform perovskite films with pinholes are commonly caused by the dynamic crystallization and grain-growth process from perovskite precursors to perovskite polycrystalline films during the fabrication process (e.g., spin coating),<sup>61</sup> which is appliable to multiple perovskite compositions.<sup>62</sup> The pinholes can lead to a shunt pathway between the perovskite layer and the charge transport layer, which induce reduction in device performance.<sup>63–65</sup> Anti-solvent (e.g., chlorobenzene, chloroform, toluene, or other nonpolar solvent) quenching treatment has been widely used to control the crystallization process, leading

to pin-hole free perovskite thin films.<sup>62</sup> Another effective approach to address the morphological challenges of perovskite films is by regulating the crystallization rate of perovskites. For example, after incorporating an acid (HBr) into the MAPbBr<sub>3</sub> precursor solution,<sup>66</sup> the solubility of the inorganic component rises, leading to a reduction in the crystallization rate of the perovskite film and a more uniform film is formed.<sup>66</sup> Furthermore, to achieve efficient radiative recombination, it's essential to confine charge carriers effectively. Studies have demonstrated that the efficiency of PeLEDs can be increased by introducing quasi-2D perovskite structure so that radiative efficiency can be improved by concentrating charge carriers.<sup>66</sup> Another example is the use of dielectric polymer: perovskite composite films, which shows a substantial increase in both photoluminescence (PL) efficiency and lifetime for CsPbBr<sub>3</sub> perovskites due to the suppression of trap states and reduction in grain boundaries.<sup>67</sup>

Improvement in light outcoupling: The EQE of an LED is a product of the IQE and light outcoupling efficiency ( $\eta_{out}$ ). With IQEs of nearly 100% being achieved in PeLEDs, realizing high EQEs would rely on the improvement of light outcoupling or the extraction of generated photons. In planar LEDs, like organic LEDs, approximately 70 to 80 percent of the emitted light from the emitters becomes confined within the device.<sup>68,69</sup> Various techniques, such as employing diffraction gratings, low-index grids, introducing polymers and buckling patterns, have been utilized to extract the light trapped within LEDs.<sup>70–73</sup> However, most of the methods have some complex fabrication difficulty which paves the way for new ideas to be implemented and make the device practically realizable.

## 4. Wavelength Tunability

Wavelength division multiplexing (WDM) is a widely employed technique in optical communication systems. It enables the simultaneous transmission of multiple signals over a single optical path by utilizing distinct wavelengths of light for each signal. By adjusting the composition of the perovskite material (e.g., the mixed halide stoichiometry), PeLEDs can emit light across the visible spectrum, including challenging colors such as blue and green (figure 5a).<sup>74</sup> This wavelength tunability of perovskite light emitters makes it promising for WDM applications. Furthermore, there is currently a so-called "green gap", a deficiency in the performance of III-V semiconductor based optical sources within the green spectral range. This underperformance stems from fundamental limitations within these materials in this energetic range, which is at the very edge of their tolerance. After only a few years of investigation, perovskite devices offer a compelling alternative in the green, with a current EQE record of 29.5%, despite that more light is trapped in thin film LEDs compared to their III-V counterparts. In other words, PeLEDs possess significant potential to surpass established technologies in green light generation.

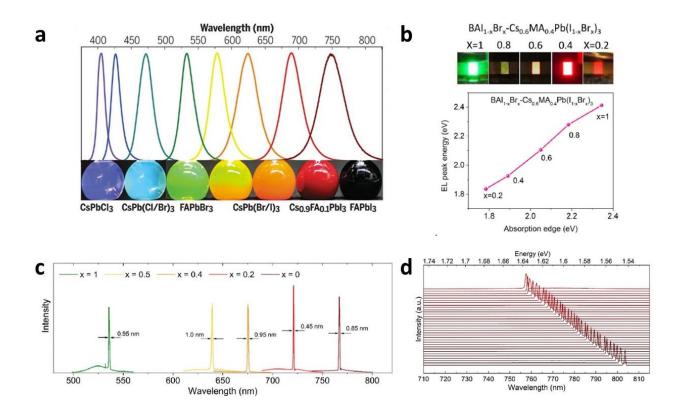
Metal halide perovskites possess a highly tunable bandgap, which directly dictates the wavelengths of light they absorb and emit. 48,76,77 At the heart of this tunability lies the perovskite's crystal structure and its flexible composition. The general formula for halide perovskites is ABX<sub>3</sub>, where A-site cations are typically organic cations like methylammonium (MA<sup>+</sup>) or formamidinium (FA<sup>+</sup>), or inorganic cations like cesium (Cs<sup>+</sup>), B-site cations are usually divalent metal ions, most commonly lead (Pb<sup>2+</sup>) and tin (Sn<sup>2+</sup>), and X-site ions are halide anions such as chloride (Cl<sup>-</sup>), bromide (Br<sup>-</sup>), or iodide (I<sup>-</sup>). By carefully manipulating the elements occupying these specific sites within the crystal lattice, the bandgap, and thus the light-emitting properties of the perovskites can be engineered.

The size and nature of the A-site cation influence the overall geometry of the perovskite lattice. Larger cations expand the lattice, leading to a smaller bandgap (red shifting the emission color), while smaller cations cause the opposite effect. This allows for a degree of bandgap control and color tuning. For example, doping with Cs<sup>+</sup> can reduce the lattice constant and enlarge bandgap in MAPbBr<sub>3</sub> crystal, which in lead to blue-shifted light emission,<sup>78</sup> while replacing Cs<sup>+</sup> with FA<sup>+</sup> leads to a wide range of tunable luminescence.<sup>79</sup> Substituting the B-site cation lead with tin is another powerful bandgap tuning method. Tin-based perovskites have smaller bandgaps than their lead counterparts, allowing for a wider range of accessible emission wavelengths. Changing the halide anions has the most dramatic impact on the bandgap. Iodide-based perovskites possess smaller bandgaps than bromide-based perovskite counterparts, while bromide-based perovskites have smaller bandgaps than chloride-based perovskites. By creating mixed-halide perovskites, the bandgap of perovskites can be controlled so that the emitted light can cover the full visible spectrum.<sup>80–82</sup>

However, simply mixing different halide species to form mixed-halide perovskites typically leads to unstable light emitting wavelength.<sup>83</sup> This wavelength instability issue originates from halide phase separation. The phase separation process leads to the formation of separate iodide-rich domains and bromide-rich domains.<sup>83</sup> Consequently, this results in a challenging "red gap" problem, wherein the emission wavelength becomes fixed to that of the lower bandgap iodide-rich phase, hindering the achievement of red-emitting PeLEDs at room temperature.

Introducing cesium (Cs) cations into perovskite materials containing methylammonium (MA) or formamidinium (FA) cations can help mitigate the lattice mismatch between bromine (Br) and iodine (I) components. This adjustment in the cation composition leads to a reduction, though not a complete elimination, of the segregation of halide species within the mixed-halide perovskite

structure. This strategy helps improve the homogeneity and stability of mixed-halide perovskite materials.<sup>84</sup> On the other hand, introducing bulky organo-ammonium ligands seems to be key to achieving stable emission wavelength for perovskite emitters. Both wavelength tunable PeLEDs (**Figure 5b**) and optically pumped lasers (**Figure 5c**, **d**) have been demonstrated using this strategy, 11,83,85,86 showing the potential of perovskite light emitters in wavelength division multiplexing.



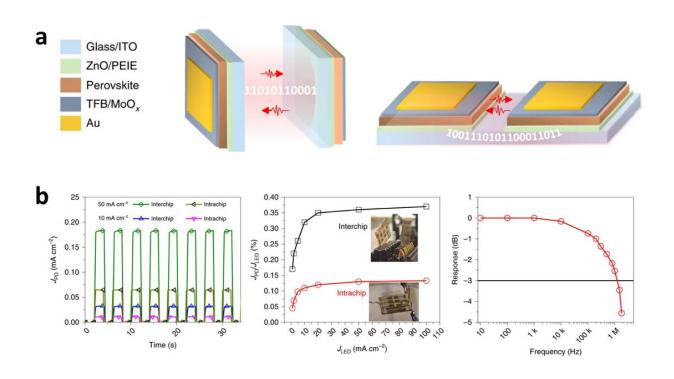
**Figure 5.** (a) Wavelength tunability of perovskite light emitters by changing their compositions. Reprinted with permission from ref.<sup>74</sup> Copyright [American Association for the Advancement of Science] [2017]. (b) Photo images and electroluminescence spectra of mixed-halide perovskite LEDs. Reprinted from ref.<sup>83</sup>. Copyright [2017] American Chemical Society. (c) Single-mode lasing emission in a wide range of wavelengths from mixed-halide perovskite thin films. Reprinted from ref.<sup>85</sup> Copyright [2019] American Chemical Society. (d) Single-mode lasing spectra from a single perovskite composition with varied distributed feedback (DFB) grating periodicities. Reprinted from ref.<sup>86</sup> Copyright [2021] American Chemical Society.

#### 5. Bidirectional communication

Optical communication is indeed a cornerstone of modern communication technology because of its numerous advantages including high-speed data transmission, long-range connectivity, robust resistance to interference, and exceptional security features. To establish any successful optical communication system, three components are needed: an emitter, a transmission channel, and a detector. The information is coded by the modulation of light from the emitter, converting the electrical signal into optical signal, which is detected by a detector, where the optical signal is converted back to the electrical signal at the receiving end. As our reliance on data communication is increasing with time, the entire system needs to be capable of dealing with high modulation bandwidth. The advantages of PeLEDs over currently available optical communication systems are intriguing, such as the potential for high-speed modulation (challenging for phosphorescent and TADF molecule-based emitters), cost effectiveness, fabrication simplicity, and easy integration with silicon chips (difficult for III-V semiconductors-based emitters). Notably, visible light communication (VLC) represents an emerging technology employing light-emitting diodes (LEDs) for both illumination and data communication simultaneously. It's anticipated to play a significant role in addressing existing bottlenecks in data and wireless communication. VLC boasts numerous advantages over lower-frequency communication methods like Wi-Fi and Bluetooth. These include superior energy efficiency, utilization of an unregulated communication spectrum, environmental friendliness, compact security, and immunity to radio frequency interferences.<sup>87–90</sup> However, conventional LED bulbs are typically based on blue inorganic LEDs with a phosphor coating as a down converter to generate white light. While the modulation speed of the inorganic LED is fast, the response speed of the phosphor coating is slow, which significantly constrained

the modulation bandwidth, hindering the realization of VLC's full potential.<sup>91</sup> Perovskite LEDs, if well engineered, can be fast, which hold great potential for VLC applications.

Interestingly, bidirectional optical communication has been demonstrated using identical perovskite based optoelectronic devices as both the emitter and the receiver (Figure 6). 15,92 Bidirectional optical signal transmission offers significant potential for miniaturized, monolithic optoelectronic systems. While achievable with III-V semiconductors, their integration remains complex. Solution-processable semiconductors promise advantages like flexibility and simplified integration but face intrinsic material and design limitations. For example, organic semiconductors suffer from a large Stokes shift, hindering efficient signal reception. Perovskites, however, demonstrate remarkable versatility in both light emission and detection. Their small Stokes shift, impressive LED efficiencies, and the sensitivity and fast response times of their photodetectors make them exceptional candidates for dual-functional devices capable of bidirectional optical signal transmission.



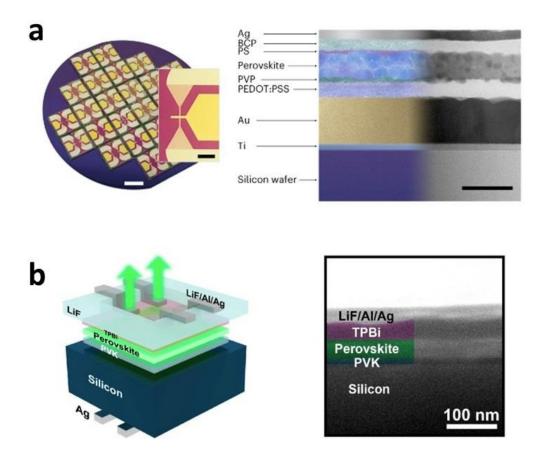
**Figure 6:** (a) PeLEDs used as inter-chip (Left) and intra-chip (Right) data communication. (b) Perovskite diode biased at -0.5V working both as photodetector and LED. Reprinted with permission from ref. <sup>15</sup> Copyright [Springer Nature] [2020].

## 6. Monolithic Integration with Silicon Microelectronics

Optoelectronic devices are ubiquitous in modern life, powering displays across our devices, enabling diverse lighting solutions, and driving lasers for healthcare, spectroscopy, communications, and security. However, a key limitation hindering their broader adoption is silicon's inability to efficiently emit light. Silicon remains the backbone of microelectronics, and the need to integrate conventional III-V semiconductor light sources adds substantial complexity and cost. Metal halide perovskites offer a compelling solution. They possess the optoelectronic properties necessary for efficient LEDs (e.g., balanced electron/hole mobility, high photoluminescence quantum yield) and, crucially, their solution-based deposition allows integration with virtually any substrate, including silicon. Monolithic integration of perovskite-based VIS-NIR light emitters on silicon would revolutionize silicon photonics, offering unparalleled scalability compared to current III-V integration approaches.

Despite a comprehensive investigation on the monolithic integration theme of perovskite emitters on silicon is yet to complete, there have been a few promising reports on PeLEDs fabricated on silicon wafers. One straightforward integration strategy would be fabricating complete PeLED stacks including the electrodes on electrically isolated silicon wafers (e.g., silicon CMOS wafers covered with an insulating oxide layer). However, direct deposition of the perovskite layer on bare c-Si substrate would result in poor film quality. As a result, a thin hole transport layer (PVK) is pre-deposited in top of the c-Si layer before spin coating Perovskite layer. For light outcoupling, a semitransparent electrode using an ultrathin metal layer (e.g., 15 nm Ag) is typically adopted in the top-emitting structure (**Figure 7a**). This integration theme can essentially be applied to any

substrates including glass and plastics,<sup>93</sup> taking advantage of the low-temperature, solution-processing capability of PeLEDs. Another study uses a p-type silicon wafer as both the substrate and hole injection layer (**Figure 7b**),<sup>94</sup> which shows the possibility of direct electrical interactions between silicon and perovskites. This integration technique provides better heat dissipation as well.



**Figure 7.** (a) Photographs of PeLEDs fabricated on a 100-mm-diameter silicon wafer, and the corresponding cross-sectional image of the PeLED. Reprinted with permission from ref. <sup>16</sup> Copyright [Springer Nature] [2023]. (b) Configuration of a PeLED on a silicon substrate and corresponding cross-sectional image of the device active stack. Reprinted from ref. <sup>94</sup> Copyright [2020] American Chemical Society.

#### 7. Conclusions and Outlook

In this Spotlight article, we have offered insight into the unique optical and electrical properties that make perovskite light emitting devices promising for next-generation optical communication applications. While significant progress has been made since the first demonstration of room-temperature electroluminescence from perovskites in 2014,95 there are still many challenges that must be overcome for this technology to reach its full potential. We summarize the primary challenges here.

- Speed: It is predicted that the bandwidth of PeLEDs can achieve a gigahertz range, exceeding the current state-of-the-art high-speed LEDs.  $^{16}$  However, all reported studies so far have been constrained by extrinsic RC factors, limiting their speed to megahertz range. To lower RC time-constant, the device area needs to be reduced. Additionally, challenges such as unbalanced charge injection, outcoupling loss, and inadequate thermal management must be tackled. The increased Joule heating and efficiency decline pose significant hurdles in sustaining continuously operating devices under high injection conditions. To achieve high speed operation, it is essential to simultaneously attain a combination of an emitter featuring fast radiative recombination and high charge-carrier mobility, alongside a low  $\tau_{RC}$  device architecture and an efficient light-outcoupling structure.
- **Stability:** One of the most significant challenges faced by PeLEDs, regardless of their specific applications, is their instability. This instability is closely related to many unique properties of halide perovskites, such as ion migration and many chemical reactions that perovskites may participate in during device operation. As a relatively new material system, there are still a lot to learn about the dynamics of the ions inside the active layer, such as the mechanism

governing the halide separation upon long-time illumination, what might be their diffusion length and most importantly, how to stabilize them. Notably, iodide transport through the charge transport layers is identified as one significant degradation channel for perovskite-based devices. Since hole transport layers efficiently transport holes in an iodide-based perovskite system, it can also be oxidized by the free iodides and hence aid the entire process of iodide separation. As a result, a charge transport layer that can efficiently block iodide transport is critically in need. One effective strategy to meet this criterion is mixing MoO<sub>3</sub> to oxidize the hole transport layer m-MTDATA (4,4',4"-tris[(3-methylphenyl)-phenylamino] triphenylamine) to avoid further oxidation with iodides.

- Efficiency roll-off: While PeLEDs can achieve high peak efficiencies, the efficiency often drops off rapidly at higher brightness levels, a phenomenon known as "efficiency roll-off". Strategies to overcome the efficiency roll-off of PeLEDs are highly desirable for many relevant applications. 100 Factors that contribute to EQE roll-off include Joule heating, 34 charge imbalance, 35 Auger recombination, 36 and electrical field-induced quenching. 37 Joule heating can be controlled by introducing thermal management strategies, such as using heat sinks alongside, or increasing the charge mobility inside the device so that the resistance can be lowered. On the other hand, quantum and dielectric confinement increases the probability of many body interactions such as Auger recombination under high injection level. 37 Using 3D perovskite single crystals can be a potential solution in this regard.
- Manufacturing and scaling: In the last ten years, PeLEDs have made consistent strides in their electroluminescent (EL) performance, with key metrics nearing the thresholds for commercial viability. However, developing industry-relevant, cost-effective, reproducible manufacturing processes for large-scale production is a challenge. While solution processing of

perovskites holds promise for low-cost production, it also introduces challenges regarding achieving high-quality films and devices with high uniformity and reproducibility, and local area patterning of different colors and compositions for wavelength division multiplexing. Inkjet printing of perovskites is a promising method due to its compatibility with large-area, flexible substrates, and its ability to pattern different materials. Recently, additive manufacturing has also come into the picture for PeLEDs. Perovskite QDs are distributed in the polymer (Polypropylene) matrix during the fabrication process which provides excellent quantum yield and stability.

• Toxicity: Most high-performing perovskite materials contain lead, which is toxic and poses environmental and health risks. Research is ongoing to find lead-free alternatives that still deliver high performance.

These are only five of the many remaining challenges facing PeLEDs for optical communications. Nevertheless, as research continues, improvements to device performance, stability, and manufacturing techniques, the potential for commercialization and widespread adoption of PeLEDs for optical communications becomes increasingly likely.

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The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

#### **Notes**

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#### REFERENCES

- (1) Fakharuddin, A.; Gangishetty, M. K.; Abdi-Jalebi, M.; Chin, S.-H.; bin Mohd Yusoff, Abd. R.; Congreve, D. N.; Tress, W.; Deschler, F.; Vasilopoulou, M.; Bolink, H. J. Perovskite Light-Emitting Diodes. *Nat Electron* **2022**, *5* (4), 203–216. https://doi.org/10.1038/s41928-022-00745-7.
- (2) Sutherland, B. R.; Sargent, E. H. Perovskite Photonic Sources. *Nature Photonics*. 2016, pp 295–302. https://doi.org/10.1038/nphoton.2016.62.
- (3) Green, M. A.; Ho-Baillie, A.; Snaith, H. J. The Emergence of Perovskite Solar Cells. *Nat Photonics* **2014**, *8* (7), 506–514. https://doi.org/10.1038/nphoton.2014.134.
- (4) Schmidt-Mende, L.; Dyakonov, V.; Olthof, S.; Ünlü, F.; Lê, K. M. T.; Mathur, S.; Karabanov, A. D.; Lupascu, D. C.; Herz, L. M.; Hinderhofer, A.; Schreiber, F.; Chernikov, A.; Egger, D. A.; Shargaieva, O.; Cocchi, C.; Unger, E.; Saliba, M.; Byranvand, M. M.; Kroll, M.; Nehm, F.; Leo, K.; Redinger, A.; Höcker, J.; Kirchartz, T.; Warby, J.; Gutierrez-Partida, E.; Neher, D.; Stolterfoht, M.; Würfel, U.; Unmüssig, M.; Herterich, J.; Baretzky, C.; Mohanraj, J.; Thelakkat, M.; Maheu, C.; Jaegermann, W.; Mayer, T.; Rieger, J.; Fauster, T.; Niesner, D.; Yang, F.; Albrecht, S.; Riedl, T.; Fakharuddin, A.; Vasilopoulou, M.; Vaynzof, Y.; Moia, D.; Maier, J.; Franckevičius, M.; Gulbinas, V.; Kerner, R. A.; Zhao, L.; Rand, B. P.; Glück, N.; Bein, T.; Matteocci, F.; Castriotta, L. A.; Di Carlo, A.; Scheffler, M.; Draxl, C. Roadmap on Organic-Inorganic Hybrid Perovskite Semiconductors and Devices. *APL Mater* 2021, *9* (10). https://doi.org/10.1063/5.0047616.
- (5) Yin, W.-J.; Shi, T.; Yan, Y. Unusual Defect Physics in CH3NH3PbI3 Perovskite Solar Cell Absorber. *Appl Phys Lett* **2014**, *104* (6), 63903. https://doi.org/10.1063/1.4864778.
- (6) Deshmukh, P.; Zhao, L.; Satapathy, S.; Khatoniar, M.; Datta, B.; Rand, B. P.; Menon, V. Radiative Pumping of Exciton-Polaritons in 2D Hybrid Perovskites. *Opt Mater Express* **2023**, *13* (6), 1655–1662. https://doi.org/10.1364/OME.485398.
- (7) Yoo, J. J.; Shin, S. S.; Seo, J. Toward Efficient Perovskite Solar Cells: Progress, Strategies, and Perspectives. *ACS Energy Lett* **2022**, 7, 2084–2091. https://doi.org/10.1021/acsenergylett.2c00592.
- (8) Fakharuddin, A.; Gangishetty, M. K.; Abdi-Jalebi, M.; Chin, S.-H.; bin Mohd Yusoff, Abd. R.; Congreve, D. N.; Tress, W.; Deschler, F.; Vasilopoulou, M.; Bolink, H. J. Perovskite Light-Emitting Diodes. *Nat Electron* **2022**, *5*, 203–216. https://doi.org/10.1038/s41928-022-00745-7.
- (9) Gunnarsson, W. B.; Roh, K.; Zhao, L.; Murphy, J. P.; Grede, A. J.; Giebink, N. C.; Rand, B. P. Toward Nonepitaxial Laser Diodes. *Chemical Reviews*. 2022, pp 7548–7584. https://doi.org/10.1021/acs.chemrev.2c00721.
- (10) Cegielski, P. J.; Giesecke, A. L.; Neutzner, S.; Porschatis, C.; Gandini, M.; Schall, D.; Perini, C. A. R.; Bolten, J.; Suckow, S.; Kataria, S.; Chmielak, B.; Wahlbrink, T.; Petrozza, A.; Lemme, M. C. Monolithically Integrated Perovskite Semiconductor Lasers on Silicon

- Photonic Chips by Scalable Top-Down Fabrication. *Nano Lett* **2018**, *18* (11), 6915–6923. https://doi.org/10.1021/acs.nanolett.8b02811.
- (11) Kim, H.; Roh, K.; Murphy, J. P.; Zhao, L.; Gunnarsson, W. B.; Longhi, E.; Barlow, S.; Marder, S. R.; Rand, B. P.; Giebink, N. C. Optically Pumped Lasing from Hybrid Perovskite Light-Emitting Diodes. *Adv Opt Mater* **2020**, *8* (1). https://doi.org/10.1002/adom.201901297.
- (12) Ren, A.; Wang, H.; Zhang, W.; Wu, J.; Wang, Z.; Penty, R. V; White, I. H. Emerging Light-Emitting Diodes for next-Generation Data Communications. *Nat Electron* **2021**, *4* (8), 559–572. https://doi.org/10.1038/s41928-021-00624-7.
- (13) Zhao, L.; Roh, K.; Kacmoli, S.; Al Kurdi, K.; Liu, X.; Barlow, S.; Marder, S. R.; Gmachl, C.; Rand, B. P. Nanosecond-Pulsed Perovskite Light-Emitting Diodes at High Current Density. *Advanced Materials* **2021**, *33* (44), 2104867. https://doi.org/10.1002/adma.202104867.
- (14) Lian, Y.; Lan, D.; Xing, S.; Guo, B.; Ren, Z.; Lai, R.; Zou, C.; Zhao, B.; Friend, R. H.; Di, D. Ultralow-Voltage Operation of Light-Emitting Diodes. *Nat Commun* 2022, *13* (1), 3845. https://doi.org/10.1038/s41467-022-31478-y.
- (15) Bao, C.; Xu, W.; Yang, J.; Bai, S.; Teng, P.; Yang, Y.; Wang, J.; Zhao, N.; Zhang, W.; Huang, W.; Gao, F. Bidirectional Optical Signal Transmission between Two Identical Devices Using Perovskite Diodes. *Nat Electron* **2020**, *3* (3), 156–164. https://doi.org/10.1038/s41928-020-0382-3.
- (16) Ren, A.; Wang, H.; Dai, L.; Xia, J.; Bai, X.; Butler-Caddle, E.; Smith, J. A.; Lai, H.; Ye, J.; Li, X.; Zhan, S.; Yao, C.; Li, Z.; Tang, M.; Liu, X.; Bi, J.; Li, B.; Kai, S.; Chen, R.; Yan, H.; Hong, J.; Yuan, L.; Marko, I. P.; Wonfor, A.; Fu, F.; Hindmarsh, S. A.; Sanchez, A. M.; Lloyd-Hughes, J.; Sweeney, S. J.; Rao, A.; Greenham, N. C.; Wu, J.; Li, Y.; Cheng, Q.; Friend, R. H.; Penty, R. V; White, I. H.; Snaith, H. J.; Zhang, W. High-Bandwidth Perovskite Photonic Sources on Silicon. *Nat Photonics* **2023**, *17* (9), 798–805. https://doi.org/10.1038/s41566-023-01242-9.
- (17) Liu, X. K.; Xu, W.; Bai, S.; Jin, Y.; Wang, J.; Friend, R. H.; Gao, F. Metal Halide Perovskites for Light-Emitting Diodes. *Nature Materials*. 2021, pp 10–21. https://doi.org/10.1038/s41563-020-0784-7.
- (18) Zhang, Q.; Tavakoli, M. M.; Gu, L.; Zhang, D.; Tang, L.; Gao, Y.; Guo, J.; Lin, Y.; Leung, S.-F.; Poddar, S.; Fu, Y.; Fan, Z. Efficient Metal Halide Perovskite Light-Emitting Diodes with Significantly Improved Light Extraction on Nanophotonic Substrates. *Nat Commun* 2019, 10 (1), 727. https://doi.org/10.1038/s41467-019-08561-y.
- (19) De Almeida, A.; Santos, B.; Paolo, B.; Quicheron, M. Solid State Lighting Review Potential and Challenges in Europe. *Renewable and Sustainable Energy Reviews* **2014**, *34*, 30–48. https://doi.org/https://doi.org/10.1016/j.rser.2014.02.029.

- (20) Commission, E.; Centre, J. R.; Zissis, G.; Bertoldi, P.; Serrenho, T. *Update on the Status of LED-Lighting World Market since 2018*; Publications Office, 2021. https://doi.org/doi/10.2760/759859.
- (21) Minotto, A.; Haigh, P. A.; Łukasiewicz, Ł. G.; Lunedei, E.; Gryko, D. T.; Darwazeh, I.; Cacialli, F. Visible Light Communication with Efficient Far-Red/near-Infrared Polymer Light-Emitting Diodes. *Light Sci Appl* **2020**, *9* (1), 70. https://doi.org/10.1038/s41377-020-0314-z.
- (22) Zou, Y.; Cai, L.; Song, T.; Sun, B. Recent Progress on Patterning Strategies for Perovskite Light-Emitting Diodes toward a Full-Color Display Prototype. *Small Science* **2021**, *1* (8), 2000050. https://doi.org/https://doi.org/10.1002/smsc.202000050.
- (23) Zou, Y.; Yuan, Z.; Bai, S.; Gao, F.; Sun, B. Recent Progress toward Perovskite Light-Emitting Diodes with Enhanced Spectral and Operational Stability. *Mater Today Nano* **2019**, *5*, 100028. https://doi.org/https://doi.org/10.1016/j.mtnano.2019.100028.
- (24) Kim, H.; Zhao, L.; Price, J. S.; Grede, A. J.; Roh, K.; Brigeman, A. N.; Lopez, M.; Rand, B. P.; Giebink, N. C. Hybrid Perovskite Light Emitting Diodes under Intense Electrical Excitation. *Nat Commun* **2018**, *9* (1), 4893. https://doi.org/10.1038/s41467-018-07383-8.
- (25) Zou, C.; Liu, Y.; Ginger, D. S.; Lin, L. Y. Suppressing Efficiency Roll-Off at High Current Densities for Ultra-Bright Green Perovskite Light-Emitting Diodes. ACS Nano 2020, 14 (5), 6076–6086. https://doi.org/10.1021/acsnano.0c01817.
- (26) Elkhouly, K.; Gehlhaar, R.; Genoe, J.; Heremans, P.; Qiu, W. Perovskite Light Emitting Diode Characteristics: The Effects of Electroluminescence Transient and Hysteresis. *Adv Opt Mater* **2020**, *8* (23), 2000941. https://doi.org/https://doi.org/10.1002/adom.202000941.
- (27) Pei, F.; Li, N.; Chen, Y.; Niu, X.; Zhang, Y.; Guo, Z.; Huang, Z.; Zai, H.; Liu, G.; Zhang, Y.; Bai, Y.; Zhang, X.; Zhu, C.; Chen, Q.; Li, Y.; Zhou, H. Thermal Management Enables More Efficient and Stable Perovskite Solar Cells. *ACS Energy Lett* **2021**, *6* (9), 3029–3036. https://doi.org/10.1021/acsenergylett.1c00999.
- (28) Qin, J.; Liu, X. K.; Yin, C.; Gao, F. Carrier Dynamics and Evaluation of Lasing Actions in Halide Perovskites. *Trends Chem* **2021**, *3* (1), 34–46. https://doi.org/10.1016/j.trechm.2020.10.010.
- deQuilettes, D. W.; Koch, S.; Burke, S.; Paranji, R. K.; Shropshire, A. J.; Ziffer, M. E.; Ginger, D. S. Photoluminescence Lifetimes Exceeding 8 Ms and Quantum Yields Exceeding 30% in Hybrid Perovskite Thin Films by Ligand Passivation. *ACS Energy Lett* **2016**, *I* (2), 438–444. https://doi.org/10.1021/acsenergylett.6b00236.
- (30) Wehrenfennig, C.; Liu, M.; Snaith, H. J.; Johnston, M. B.; Herz, L. M. Charge-Carrier Dynamics in Vapour-Deposited Films of the Organolead Halide Perovskite CH3NH3PbI3–xClx. *Energy Environ Sci* **2014**, 7 (7), 2269–2275. https://doi.org/10.1039/C4EE01358A.

- (31) Rehman, W.; Milot, R. L.; Eperon, G. E.; Wehrenfennig, C.; Boland, J. L.; Snaith, H. J.; Johnston, M. B.; Herz, L. M. Charge-Carrier Dynamics and Mobilities in Formamidinium Lead Mixed-Halide Perovskites. *Advanced Materials* **2015**, *27* (48), 7938–7944. https://doi.org/https://doi.org/10.1002/adma.201502969.
- (32) Wehrenfennig, C.; Eperon, G. E.; Johnston, M. B.; Snaith, H. J.; Herz, L. M. High Charge Carrier Mobilities and Lifetimes in Organolead Trihalide Perovskites. *Advanced Materials* **2014**, *26* (10), 1584–1589. https://doi.org/https://doi.org/10.1002/adma.201305172.
- (33) Wu, B.; Shastri, B. J.; Prucnal, P. R. System Performance Measurement and Analysis of Optical Steganography Based on Noise. *IEEE Photonics Technology Letters* **2014**, *26* (19), 1920–1923. https://doi.org/10.1109/LPT.2014.2341917.
- (34) Zhao, L.; Roh, K.; Kacmoli, S.; Al Kurdi, K.; Jhulki, S.; Barlow, S.; Marder, S. R.; Gmachl, C.; Rand, B. P. Thermal Management Enables Bright and Stable Perovskite Light-Emitting Diodes. *Advanced Materials* **2020**, *32* (25), 2000752. https://doi.org/10.1002/adma.202000752.
- (35) Fakharuddin, A.; Qiu, W.; Croes, G.; Devižis, A.; Gegevičius, R.; Vakhnin, A.; Rolin, C.; Genoe, J.; Gehlhaar, R.; Kadashchuk, A.; Gulbinas, V.; Heremans, P. Reduced Efficiency Roll-Off and Improved Stability of Mixed 2D/3D Perovskite Light Emitting Diodes by Balancing Charge Injection. *Adv Funct Mater* **2019**, *29* (37), 1904101. https://doi.org/10.1002/adfm.201904101.
- (36) Chen, W.; Huang, Z.; Yao, H.; Liu, Y.; Zhang, Y.; Li, Z.; Zhou, H.; Xiao, P.; Chen, T.; Sun, H.; Huang, J.; Xiao, Z. Highly Bright and Stable Single-Crystal Perovskite Light-Emitting Diodes. *Nat Photonics* **2023**. https://doi.org/10.1038/s41566-023-01167-3.
- (37) Zou, W.; Li, R.; Zhang, S.; Liu, Y.; Wang, N.; Cao, Y.; Miao, Y.; Xu, M.; Guo, Q.; Di, D.; Zhang, L.; Yi, C.; Gao, F.; Friend, R. H.; Wang, J.; Huang, W. Minimising Efficiency Roll-off in High-Brightness Perovskite Light-Emitting Diodes. *Nat Commun* **2018**, *9* (1), 608. https://doi.org/10.1038/s41467-018-03049-7.
- (38) Zhao, L.; Lee, K. M.; Roh, K.; Khan, S. U. Z.; Rand, B. P. Improved Outcoupling Efficiency and Stability of Perovskite Light-Emitting Diodes Using Thin Emitting Layers. *Advanced Materials* **2019**, *31* (2), 1805836. https://doi.org/10.1002/adma.201805836.
- (39) Zhao, L.; Roh, K.; Kacmoli, S.; Al Kurdi, K.; Jhulki, S.; Barlow, S.; Marder, S. R.; Gmachl, C.; Rand, B. P. Thermal Management Enables Bright and Stable Perovskite Light-Emitting Diodes. *Advanced Materials* **2020**, *32* (25), 2000752. https://doi.org/10.1002/adma.202000752.
- (40) Zhao, L.; Roh, K.; Kacmoli, S.; Al Kurdi, K.; Liu, X.; Barlow, S.; Marder, S. R.; Gmachl, C.; Rand, B. P. Nanosecond-Pulsed Perovskite Light-Emitting Diodes at High Current Density. *Advanced Materials* 2021, 33 (44), 2104867. https://doi.org/https://doi.org/10.1002/adma.202104867.
- (41) Zhao, L.; Astridge, D. D.; Gunnarsson, W. B.; Xu, Z.; Hong, J.; Scott, J.; Kacmoli, S.; Al Kurdi, K.; Barlow, S.; Marder, S. R.; Gmachl, C. F.; Sellinger, A.; Rand, B. P. Thermal

- Properties of Polymer Hole-Transport Layers Influence the Efficiency Roll-off and Stability of Perovskite Light-Emitting Diodes. *Nano Lett* **2023**, *23* (11), 4785–4792. https://doi.org/10.1021/acs.nanolett.3c00148.
- (42) Yang, Y.; Xu, S.; Ni, Z.; Van Brackle, C. H.; Zhao, L.; Xiao, X.; Dai, X.; Huang, J. Highly Efficient Pure-Blue Light-Emitting Diodes Based on Rubidium and Chlorine Alloyed Metal Halide Perovskite. *Advanced Materials* **2021**, *33* (33), 2100783. https://doi.org/https://doi.org/10.1002/adma.202100783.
- (43) Sun, R.; Lu, P.; Zhou, D.; Xu, W.; Ding, N.; Shao, H.; Zhang, Y.; Li, D.; Wang, N.; Zhuang, X.; Dong, B.; Bai, X.; Song, H. Samarium-Doped Metal Halide Perovskite Nanocrystals for Single-Component Electroluminescent White Light-Emitting Diodes. *ACS Energy Lett* **2020**, *5* (7), 2131–2139. https://doi.org/10.1021/acsenergylett.0c00931.
- (44) Zou, S.; Liu, Y.; Li, J.; Liu, C.; Feng, R.; Jiang, F.; Li, Y.; Song, J.; Zeng, H.; Hong, M.; Chen, X. Stabilizing Cesium Lead Halide Perovskite Lattice through Mn(II) Substitution for Air-Stable Light-Emitting Diodes. *J Am Chem Soc* **2017**, *139* (33), 11443–11450. https://doi.org/10.1021/jacs.7b04000.
- (45) Byun, J.; Cho, H.; Wolf, C.; Jang, M.; Sadhanala, A.; Friend, R. H.; Yang, H.; Lee, T.-W. Efficient Visible Quasi-2D Perovskite Light-Emitting Diodes. *Advanced Materials* **2016**, 28 (34), 7515–7520. https://doi.org/https://doi.org/10.1002/adma.201601369.
- (46) Li, J.; Bade, S. G. R.; Shan, X.; Yu, Z. Single-Layer Light-Emitting Diodes Using Organometal Halide Perovskite/Poly(Ethylene Oxide) Composite Thin Films. *Advanced Materials* **2015**, 27 (35), 5196–5202. https://doi.org/https://doi.org/10.1002/adma.201502490.
- (47) Zhao, L.; Lee, K. M.; Roh, K.; Khan, S. U. Z.; Rand, B. P. Improved Outcoupling Efficiency and Stability of Perovskite Light-Emitting Diodes Using Thin Emitting Layers. *Advanced Materials* **2019**, *31* (2), 1805836. https://doi.org/https://doi.org/10.1002/adma.201805836.
- (48) Fakharuddin, A.; Gangishetty, M. K.; Abdi-Jalebi, M.; Chin, S.-H.; bin Mohd Yusoff, Abd. R.; Congreve, D. N.; Tress, W.; Deschler, F.; Vasilopoulou, M.; Bolink, H. J. Perovskite Light-Emitting Diodes. *Nat Electron* **2022**, *5* (4), 203–216. https://doi.org/10.1038/s41928-022-00745-7.
- (49) Tan, Z.-K.; Moghaddam, R. S.; Lai, M. L.; Docampo, P.; Higler, R.; Deschler, F.; Price, M.; Sadhanala, A.; Pazos, L. M.; Credgington, D.; Hanusch, F.; Bein, T.; Snaith, H. J.; Friend, R. H. Bright Light-Emitting Diodes Based on Organometal Halide Perovskite. *Nat Nanotechnol* 2014, 9 (9), 687–692. https://doi.org/10.1038/nnano.2014.149.
- (50) Kim, H.; Zhao, L.; Price, J. S.; Grede, A. J.; Roh, K.; Brigeman, A. N.; Lopez, M.; Rand, B. P.; Giebink, N. C. Hybrid Perovskite Light Emitting Diodes under Intense Electrical Excitation. *Nat Commun* **2018**, *9* (1), 4893. https://doi.org/10.1038/s41467-018-07383-8.
- (51) Zhao, L.; Astridge, D. D.; Gunnarsson, W. B.; Xu, Z.; Hong, J.; Scott, J.; Kacmoli, S.; Al Kurdi, K.; Barlow, S.; Marder, S. R.; Gmachl, C. F.; Sellinger, A.; Rand, B. P. Thermal Properties of Polymer Hole-Transport Layers Influence the Efficiency Roll-off and Stability

- of Perovskite Light-Emitting Diodes. *Nano Lett* **2023**. https://doi.org/10.1021/acs.nanolett.3c00148.
- (52) Xiao, Z.; Kerner, R. A.; Zhao, L.; Tran, N. L.; Lee, K. M.; Koh, T. W.; Scholes, G. D.; Rand, B. P. Efficient Perovskite Light-Emitting Diodes Featuring Nanometre-Sized Crystallites. *Nat Photonics* **2017**, *11* (2), 108–115. https://doi.org/10.1038/nphoton.2016.269.
- (53) Lei, L.; Dong, Q.; Gundogdu, K.; So, F. Metal Halide Perovskites for Laser Applications. *Adv Funct Mater* **2021**, *31* (16), 2010144. https://doi.org/https://doi.org/10.1002/adfm.202010144.
- (54) Zhao, L.; Astridge, D.; Gunnarsson, W.; Xu, Z.; Hong, J.; Scott, J.; Kacmoli, S.; Al Kurdi, K.; Barlow, S.; Marder, S.; Gmachl, C.; Sellinger, A.; Rand, B. Thermal Properties of Polymer Hole-Transport Layers Influence the Efficiency Roll-off and Stability of Perovskite Light-Emitting Diodes. *Nano Lett 0* (0), null-null. https://doi.org/10.1021/acs.nanolett.3c00148.
- (55) Zou, C.; Liu, Y.; Ginger, D. S.; Lin, L. Y. Suppressing Efficiency Roll-Off at High Current Densities for Ultra-Bright Green Perovskite Light-Emitting Diodes. *ACS Nano* **2020**, *14* (5), 6076–6086. https://doi.org/10.1021/acsnano.0c01817.
- (56) Lu, M.; Zhang, X.; Bai, X.; Wu, H.; Shen, X.; Zhang, Y.; Zhang, W.; Zheng, W.; Song, H.; Yu, W. W.; Rogach, A. L. Spontaneous Silver Doping and Surface Passivation of CsPbI3 Perovskite Active Layer Enable Light-Emitting Devices with an External Quantum Efficiency of 11.2%. *ACS Energy Lett* **2018**, *3* (7), 1571–1577. https://doi.org/10.1021/acsenergylett.8b00835.
- (57) Jin, X.; Chang, C.; Zhao, W.; Huang, S.; Gu, X.; Zhang, Q.; Li, F.; Zhang, Y.; Li, Q. Balancing the Electron and Hole Transfer for Efficient Quantum Dot Light-Emitting Diodes by Employing a Versatile Organic Electron-Blocking Layer. *ACS Appl Mater Interfaces* **2018**, *10* (18), 15803–15811. https://doi.org/10.1021/acsami.8b00729.
- (58) Xiao, X.; Wang, K.; Ye, T.; Cai, R.; Ren, Z.; Wu, D.; Qu, X.; Sun, J.; Ding, S.; Sun, X. W.; Choy, W. C. H. Enhanced Hole Injection Assisted by Electric Dipoles for Efficient Perovskite Light-Emitting Diodes. *Commun Mater* **2020**, *1* (1), 81. https://doi.org/10.1038/s43246-020-00084-0.
- (59) Shin, Y. S.; Yoon, Y. J.; Heo, J.; Song, S.; Kim, J. W.; Park, S. Y.; Cho, H. W.; Kim, G.-H.; Kim, J. Y. Functionalized PFN-X (X = Cl, Br, or I) for Balanced Charge Carriers of Highly Efficient Blue Light-Emitting Diodes. *ACS Appl Mater Interfaces* **2020**, *12* (31), 35740–35747. https://doi.org/10.1021/acsami.0c09968.
- (60) Wang, J.; Song, C.; He, Z.; Mai, C.; Xie, G.; Mu, L.; Cun, Y.; Li, J.; Wang, J.; Peng, J.; Cao, Y. All-Solution-Processed Pure Formamidinium-Based Perovskite Light-Emitting Diodes. *Advanced Materials* 2018, 30 (39), 1804137. https://doi.org/https://doi.org/10.1002/adma.201804137.

- (61) Kerner, R. A.; Zhao, L.; Xiao, Z.; Rand, B. P. Ultrasmooth Metal Halide Perovskite Thin Films via Sol–Gel Processing. *J Mater Chem A Mater* **2016**, *4* (21), 8308–8315. https://doi.org/10.1039/C6TA03092K.
- (62) Sun, J.; Li, F.; Yuan, J.; Ma, W. Advances in Metal Halide Perovskite Film Preparation: The Role of Anti-Solvent Treatment. *Small Methods* **2021**, *5* (5), 2100046. https://doi.org/https://doi.org/10.1002/smtd.202100046.
- (63) Wang, J.; Wang, N.; Jin, Y.; Si, J.; Tan, Z.-K.; Du, H.; Cheng, L.; Dai, X.; Bai, S.; He, H.; Ye, Z.; Lai, M. L.; Friend, R. H.; Huang, W. Interfacial Control Toward Efficient and Low-Voltage Perovskite Light-Emitting Diodes. *Advanced Materials* **2015**, *27* (14), 2311–2316. https://doi.org/https://doi.org/10.1002/adma.201405217.
- (64) Stranks, S. D.; Hoye, R. L. Z.; Di, D.; Friend, R. H.; Deschler, F. The Physics of Light Emission in Halide Perovskite Devices. *Advanced Materials* **2019**, *31* (47), 1803336. https://doi.org/https://doi.org/10.1002/adma.201803336.
- (65) McMeekin, D. P.; Wang, Z.; Rehman, W.; Pulvirenti, F.; Patel, J. B.; Noel, N. K.; Johnston, M. B.; Marder, S. R.; Herz, L. M.; Snaith, H. J. Crystallization Kinetics and Morphology Control of Formamidinium—Cesium Mixed-Cation Lead Mixed-Halide Perovskite via Tunability of the Colloidal Precursor Solution. *Advanced Materials* 2017, 29 (29), 1607039. https://doi.org/https://doi.org/10.1002/adma.201607039.
- (66) Yu, J. C.; Kim, D. Bin; Jung, E. D.; Lee, B. R.; Song, M. H. High-Performance Perovskite Light-Emitting Diodes via Morphological Control of Perovskite Films. *Nanoscale* 2016, 8 (13), 7036–7042. https://doi.org/10.1039/C5NR05604G.
- (67) Wu, C.; Zou, Y.; Wu, T.; Ban, M.; Pecunia, V.; Han, Y.; Liu, Q.; Song, T.; Duhm, S.; Sun, B. Improved Performance and Stability of All-Inorganic Perovskite Light-Emitting Diodes by Antisolvent Vapor Treatment. *Adv Funct Mater* **2017**, *27* (28), 1700338. https://doi.org/https://doi.org/10.1002/adfm.201700338.
- (68) Lee, Y.-J.; Kim, S.-H.; Huh, J.; Kim, G.-H.; Lee, Y.-H.; Cho, S.-H.; Kim, Y.-C.; Do, Y. R. A High-Extraction-Efficiency Nanopatterned Organic Light-Emitting Diode. *Appl Phys Lett* **2003**, *82* (21), 3779–3781. https://doi.org/10.1063/1.1577823.
- (69) Bulović, V.; Khalfin, V. B.; Gu, G.; Burrows, P. E.; Garbuzov, D. Z.; Forrest, S. R. Weak Microcavity Effects in Organic Light-Emitting Devices. *Phys Rev B* **1998**, *58* (7), 3730–3740. https://doi.org/10.1103/PhysRevB.58.3730.
- (70) Ziebarth, J. M.; Saafir, A. K.; Fan, S.; McGehee, M. D. Extracting Light from Polymer Light-Emitting Diodes Using Stamped Bragg Gratings. *Adv Funct Mater* **2004**, *14* (5), 451–456. https://doi.org/https://doi.org/10.1002/adfm.200305070.
- (71) Koo, W. H.; Jeong, S. M.; Araoka, F.; Ishikawa, K.; Nishimura, S.; Toyooka, T.; Takezoe, H. Light Extraction from Organic Light-Emitting Diodes Enhanced by Spontaneously Formed Buckles. *Nat Photonics* **2010**, *4* (4), 222–226. https://doi.org/10.1038/nphoton.2010.7.

- (72) Sun, Y.; Forrest, S. R. Enhanced Light Out-Coupling of Organic Light-Emitting Devices Using Embedded Low-Index Grids. *Nat Photonics* **2008**, *2* (8), 483–487. https://doi.org/10.1038/nphoton.2008.132.
- (73) Matterson, B. J.; Lupton, J. M.; Safonov, A. F.; Salt, M. G.; Barnes, W. L.; Samuel, I. D. W. Increased Efficiency and Controlled Light Output from a Microstructured Light-Emitting Diode. *Advanced Materials* 2001, 13 (2), 123–127. https://doi.org/https://doi.org/10.1002/1521-4095(200101)13:2<123::AID-ADMA123>3.0.CO;2-D.
- (74) Kovalenko, M. V; Protesescu, L.; Bodnarchuk, M. I. Properties and Potential Optoelectronic Applications of Lead Halide Perovskite Nanocrystals. *Science* (1979) **2017**, 358 (6364), 745–750. https://doi.org/10.1126/science.aam7093.
- (75) Ding, S.; Wang, Q.; Gu, W.; Tang, Z.; Zhang, B.; Wu, C.; Zhang, X.; Chen, H.; Zhang, X.; Cao, R.; Chen, T.; Qian, L.; Xiang, C. Phase Dimensions Resolving of Efficient and Stable Perovskite Light-Emitting Diodes at High Brightness. *Nat Photonics* **2024**. https://doi.org/10.1038/s41566-023-01372-0.
- (76) Schmidt-Mende, L.; Dyakonov, V.; Olthof, S.; Ünlü, F.; Lê, K. M. T.; Mathur, S.; Karabanov, A. D.; Lupascu, D. C.; Herz, L. M.; Hinderhofer, A.; Schreiber, F.; Chernikov, A.; Egger, D. A.; Shargaieva, O.; Cocchi, C.; Unger, E.; Saliba, M.; Byranvand, M. M.; Kroll, M.; Nehm, F.; Leo, K.; Redinger, A.; Höcker, J.; Kirchartz, T.; Warby, J.; Gutierrez-Partida, E.; Neher, D.; Stolterfoht, M.; Würfel, U.; Unmüssig, M.; Herterich, J.; Baretzky, C.; Mohanraj, J.; Thelakkat, M.; Maheu, C.; Jaegermann, W.; Mayer, T.; Rieger, J.; Fauster, T.; Niesner, D.; Yang, F.; Albrecht, S.; Riedl, T.; Fakharuddin, A.; Vasilopoulou, M.; Vaynzof, Y.; Moia, D.; Maier, J.; Franckevičius, M.; Gulbinas, V.; Kerner, R. A.; Zhao, L.; Rand, B. P.; Glück, N.; Bein, T.; Matteocci, F.; Castriotta, L. A.; Di Carlo, A.; Scheffler, M.; Draxl, C. Roadmap on Organic–Inorganic Hybrid Perovskite Semiconductors and Devices. APL Mater 2021, 9 (10), 109202. https://doi.org/10.1063/5.0047616.
- (77) Quan, L. N.; Rand, B. P.; Friend, R. H.; Mhaisalkar, S. G.; Lee, T.-W.; Sargent, E. H. Perovskites for Next-Generation Optical Sources. *Chem Rev* **2019**, *119* (12), 7444–7477. https://doi.org/10.1021/acs.chemrev.9b00107.
- (78) Xu, B.; Wang, W.; Zhang, X.; Cao, W.; Wu, D.; Liu, S.; Dai, H.; Chen, S.; Wang, K.; Sun, X. Bright and Efficient Light-Emitting Diodes Based on MA/Cs Double Cation Perovskite Nanocrystals. *J Mater Chem C Mater* **2017**, *5* (25), 6123–6128. https://doi.org/10.1039/C7TC01300K.
- (79) Chen, D.; Chen, X.; Wan, Z.; Fang, G. Full-Spectral Fine-Tuning Visible Emissions from Cation Hybrid Cs1–MFAmPbX3 (X = Cl, Br, and I,  $0 \le m \le 1$ ) Quantum Dots. *ACS Appl Mater Interfaces* **2017**, 9 (24), 20671–20678. https://doi.org/10.1021/acsami.7b05429.
- (80) Akkerman, Q. A.; D'Innocenzo, V.; Accornero, S.; Scarpellini, A.; Petrozza, A.; Prato, M.; Manna, L. Tuning the Optical Properties of Cesium Lead Halide Perovskite Nanocrystals by Anion Exchange Reactions. *J Am Chem Soc* **2015**, *137* (32), 10276–10281. https://doi.org/10.1021/jacs.5b05602.

- (81) Zhang, D.; Yang, Y.; Bekenstein, Y.; Yu, Y.; Gibson, N. A.; Wong, A. B.; Eaton, S. W.; Kornienko, N.; Kong, Q.; Lai, M.; Alivisatos, A. P.; Leone, S. R.; Yang, P. Synthesis of Composition Tunable and Highly Luminescent Cesium Lead Halide Nanowires through Anion-Exchange Reactions. *J Am Chem Soc* 2016, 138 (23), 7236–7239. https://doi.org/10.1021/jacs.6b03134.
- (82) Imran, M.; Caligiuri, V.; Wang, M.; Goldoni, L.; Prato, M.; Krahne, R.; De Trizio, L.; Manna, L. Benzoyl Halides as Alternative Precursors for the Colloidal Synthesis of Lead-Based Halide Perovskite Nanocrystals. *J Am Chem Soc* **2018**, *140* (7), 2656–2664. https://doi.org/10.1021/jacs.7b13477.
- (83) Xiao, Z.; Zhao, L.; Tran, N. L.; Lin, Y. L.; Silver, S. H.; Kerner, R. A.; Yao, N.; Kahn, A.; Scholes, G. D.; Rand, B. P. Mixed-Halide Perovskites with Stabilized Bandgaps. *Nano Lett* **2017**, *17* (11). https://doi.org/10.1021/acs.nanolett.7b03179.
- (84) McMeekin, D. P.; Sadoughi, G.; Rehman, W.; Eperon, G. E.; Saliba, M.; Hörantner, M. T.; Haghighirad, A.; Sakai, N.; Korte, L.; Rech, B.; Johnston, M. B.; Herz, L. M.; Snaith, H. J. A Mixed-Cation Lead Mixed-Halide Perovskite Absorber for Tandem Solar Cells. *Science* (1979) 2016, 351 (6269), 151–155. https://doi.org/10.1126/science.aad5845.
- (85) Roh, K.; Zhao, L.; Gunnarsson, W.; Xiao, Z.; Jia, Y.; Giebink, N.; Rand, B. Widely Tunable, Room Temperature, Single-Mode Lasing Operation from Mixed-Halide Perovskite Thin Films. *ACS Photonics* 6 (12), 3331–3337. https://doi.org/10.1021/acsphotonics.9b01501.
- (86) Roh, K.; Zhao, L.; Rand, B. Tuning Laser Threshold within the Large Optical Gain Bandwidth of Halide Perovskite Thin Films. *ACS Photonics* 8 (8), 2548–2554. https://doi.org/10.1021/acsphotonics.1c00910.
- (87) Shen, C.; Ng, T. K.; Leonard, J. T.; Pourhashemi, A.; Oubei, H. M.; Alias, M. S.; Nakamura, S.; DenBaars, S. P.; Speck, J. S.; Alyamani, A. Y.; Eldesouki, M. M.; Ooi, B. S. High-Modulation-Efficiency, Integrated Waveguide Modulator–Laser Diode at 448 Nm. *ACS Photonics* **2016**, *3* (2), 262–268. https://doi.org/10.1021/acsphotonics.5b00599.
- (88) Zhang, Z.; Guo, K.; Li, Y.; Li, X.; Guan, G.; Li, H.; Luo, Y.; Zhao, F.; Zhang, Q.; Wei, B.; Pei, Q.; Peng, H. A Colour-Tunable, Weavable Fibre-Shaped Polymer Light-Emitting Electrochemical Cell. *Nat Photonics* **2015**, *9* (4), 233–238. https://doi.org/10.1038/nphoton.2015.37.
- (89) Behabtu, N.; Young, C. C.; Tsentalovich, D. E.; Kleinerman, O.; Wang, X.; Ma, A. W. K.; Bengio, E. A.; ter Waarbeek, R. F.; de Jong, J. J.; Hoogerwerf, R. E.; Fairchild, S. B.; Ferguson, J. B.; Maruyama, B.; Kono, J.; Talmon, Y.; Cohen, Y.; Otto, M. J.; Pasquali, M. Strong, Light, Multifunctional Fibers of Carbon Nanotubes with Ultrahigh Conductivity. *Science* (1979) **2013**, 339 (6116), 182–186. https://doi.org/10.1126/science.1228061.
- (90) Wang, Z.; Wang, Z.; Lin, B.; Hu, X.; Wei, Y.; Zhang, C.; An, B.; Wang, C.; Lin, W. Warm-White-Light-Emitting Diode Based on a Dye-Loaded Metal-Organic Framework for Fast White-Light Communication. *ACS applied materials & amp; interfaces* **2017**, *9* (40), 35253–35259. https://doi.org/10.1021/acsami.7b11277.

- (91) Dursun, I.; Shen, C.; Parida, M. R.; Pan, J.; Sarmah, S. P.; Priante, D.; Alyami, N.; Liu, J.; Saidaminov, M. I.; Alias, M. S.; Abdelhady, A. L.; Ng, T. K.; Mohammed, O. F.; Ooi, B. S.; Bakr, O. M. Perovskite Nanocrystals as a Color Converter for Visible Light Communication. ACS Photonics 2016, 3 (7), 1150–1156. https://doi.org/10.1021/acsphotonics.6b00187.
- (92) Shan, Q.; Wei, C.; Jiang, Y.; Song, J.; Zou, Y.; Xu, L.; Fang, T.; Wang, T.; Dong, Y.; Liu, J.; Han, B.; Zhang, F.; Chen, J.; Wang, Y.; Zeng, H. Perovskite Light-Emitting/Detecting Bifunctional Fibres for Wearable LiFi Communication. *Light Sci Appl* **2020**, *9* (1), 163. https://doi.org/10.1038/s41377-020-00402-8.
- (93) Miao, Y.; Cheng, L.; Zou, W.; Gu, L.; Zhang, J.; Guo, Q.; Peng, Q.; Xu, M.; He, Y.; Zhang, S.; Cao, Y.; Li, R.; Wang, N.; Huang, W.; Wang, J. Microcavity Top-Emission Perovskite Light-Emitting Diodes. *Light Sci Appl* 2020, 9 (1), 89. https://doi.org/10.1038/s41377-020-0328-6.
- (94) Xu, H.; Wang, X.; Li, Y.; Cai, L.; Tan, Y.; Zhang, G.; Wang, Y.; Li, R.; Liang, D.; Song, T.; Sun, B. Prominent Heat Dissipation in Perovskite Light-Emitting Diodes with Reduced Efficiency Droop for Silicon-Based Display. *J Phys Chem Lett* **2020**, *11* (9), 3689–3698. https://doi.org/10.1021/acs.jpclett.0c00792.
- (95) Tan, Z. K.; Moghaddam, R. S.; Lai, M. L.; Docampo, P.; Higler, R.; Deschler, F.; Price, M.; Sadhanala, A.; Pazos, L. M.; Credgington, D.; Hanusch, F.; Bein, T.; Snaith, H. J.; Friend, R. H. Bright Light-Emitting Diodes Based on Organometal Halide Perovskite. *Nat Nanotechnol* **2014**, *9* (9), 687–692. https://doi.org/10.1038/nnano.2014.149.
- (96) Dong, Q.; Lei, L.; Mendes, J.; So, F. Operational Stability of Perovskite Light Emitting Diodes. *Journal of Physics: Materials* **2020**, *3* (1), 12002. https://doi.org/10.1088/2515-7639/ab60c4.
- (97) Zhao, L.; Kerner, R.; Xiao, Z.; Lin, Y.; Lee, K. M.; Schwartz, J.; Rand, B. Redox Chemistry Dominates the Degradation and Decomposition of Metal Halide Perovskite Optoelectronic Devices. *ACS Energy Lett 1* (3), 595–602. https://doi.org/10.1021/acsenergylett.6b00320.
- (98) Xu, Z.; Astridge, D. D.; Kerner, R. A.; Zhong, X.; Hu, J.; Hong, J.; Wisch, J. A.; Zhu, K.; Berry, J. J.; Kahn, A.; Sellinger, A.; Rand, B. P. Origins of Photoluminescence Instabilities at Halide Perovskite/Organic Hole Transport Layer Interfaces. *J Am Chem Soc* **2023**, *145* (21), 11846–11858. https://doi.org/10.1021/jacs.3c03539.
- (99) Hong, J.; Xu, Z.; Lungwitz, D.; Scott, J.; Johnson, H. M.; Kim, Y.-H.; Kahn, A.; Rand, B. P. Mitigating Iodine Diffusion by a MoO3–Organic Composite Hole Transport Layer for Stable Perovskite Solar Cells. ACS Energy Lett 2023, 8 (12), 4984–4992. https://doi.org/10.1021/acsenergylett.3c01873.
- (100) Han, T.-H.; Jang, K. Y.; Dong, Y.; Friend, R. H.; Sargent, E. H.; Lee, T.-W. A Roadmap for the Commercialization of Perovskite Light Emitters. *Nat Rev Mater* **2022**, *7* (10), 757–777. https://doi.org/10.1038/s41578-022-00459-4.

- (101) Liu, Y.; Li, F.; Qiu, L.; Yang, K.; Li, Q.; Zheng, X.; Hu, H.; Guo, T.; Wu, C.; Kim, T. W. Fluorescent Microarrays of in Situ Crystallized Perovskite Nanocomposites Fabricated for Patterned Applications by Using Inkjet Printing. *ACS Nano* **2019**, *13* (2), 2042–2049. https://doi.org/10.1021/acsnano.8b08582.
- (102) Solodov, A. N.; Shayimova, J.; Balkaev, D.; Nizamutdinov, A. S.; Zimin, K.; Kiiamov, A. G.; Amirov, R. R.; Dimiev, A. M. High-Throughput, Low-Cost and "Green" Production Method for Highly Stable Polypropylene/Perovskite Composites, Applicable in 3D Printing. *Addit Manuf* **2022**, *59*, 103094. https://doi.org/https://doi.org/10.1016/j.addma.2022.103094.