Highly Efficient Visible Colloidal Lead-Halide Perovskite Nanocrystal Light-Emitting Diodes

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Supporting Information

ABSTRACT: Lead-halide perovskites have been attracting attention for potential use in solid-state lighting. Following the footsteps of solar cells, the field of perovskite light-emitting diodes (PeLEDs) has been growing rapidly. Their application prospects in lighting, however, remain still uncertain due to a variety of shortcomings in device performance including their limited levels of luminous efficiency achievable thus far. Here we show high-efficiency PeLEDs based on colloidal perovskite nanocrystals (PeNCs) synthesized at room temperature possessing dominant first-order excitonic radiation (enabling a photoluminescence quantum yield of 71% in solid film), unlike in the case of bulk perovskites with slow electron–hole bimolecular radiative recombination (a second-order process). In these PeLEDs, by reaching charge balance in the recombination zone, we find that the Auger nonradiative recombination, with its significant role in emission quenching, is effectively suppressed in low driving current density range. In consequence, these devices reach a maximum external quantum efficiency of 12.9% and a power efficiency of 30.3 lm W⁻¹ at luminance levels above 1000 cd m⁻² as required for various applications. These findings suggest that, with feasible levels of device performance, the PeNCs hold great promise for their use in LED lighting and displays.

KEYWORDS: Colloidal nanocrystal, lead-halide perovskite, light-emitting diode

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nonradiative recombination, highly efficient emission at low charge densities is desired. By effectively suppressing Auger nonradiative recombination with balanced charge at low driving charge density levels, these PeLEDs exhibit a high EQE of 12.9% and a high power efficiency level of 30.3 lm W⁻¹ at high brightness level above 1000 cd m⁻², which indicates the feasibility of colloidal PeNCs in LED applications in terms of efficiency requirements.

**Luminescence Dynamics of Lead-Halide Perovskite Emitters.** The radiation of bulk perovskites originates from slow bimolecular recombination. The Langevin theory predicts that this bimolecular recombination rate could be four orders of magnitude higher in perovskites, which is one of the main merits ensuring superior performance in photovoltaics. The carrier kinetics in bulk lead-halide perovskites can be described by

\[
\frac{dn(t)}{dt} = G - nk_1 - n^2k_2 - n^3k_3
\]

(1)

where \(n\) is the carrier density, \(t\) is the time, \(G\) is the generation rate of carrier density, \(k_1\) is the first-order trap-mediated monomolecular recombination rate constant, \(k_2\) is the electron–hole bimolecular recombination rate constant, and \(k_3\) is the many-body Auger recombination rate constant. Thus, under steady-state electric excitation, the luminous QY (\(A(n)\)) is given by

\[
A(n) = \frac{nk_3}{k_1 + nk_2 + n^2k_3}
\]

(2)

The emission efficiency (Figure 1a) first increases with increasing \(n\) due to the domination of bimolecular recombination over the trap-mediated nonradiative recombination. However, at high carrier densities, where the Auger recombination dominates over the bimolecular recombination, the emission efficiency will decrease with increasing \(n\). Since \(k_2\) (\(\sim 10^{10}\) cm³ s⁻¹) and \(k_3\) (\(\sim 10^{28}\) cm³ s⁻¹) are the intrinsic parameters of the bulk perovskites, the emission efficiency strongly depends on \(k_1\) and \(n\). By ignoring other quenching factors, with a typical reported value of \(k_1\) (\(\sim 10^7\) s⁻¹, 100 ns), the internal efficiency of the PeLEDs is constrained to <10%, even when driving at a high carrier density of \(10^{16}\) cm⁻³ (Figure 1a).

The bulk perovskite films have shown a long carrier lifetime (i.e., low trap states density) and a high mobility. Thus, the delocalized charge carriers are also possible to transport through the thin active layer without recombination. When the PeLEDs are driven at a lower carrier density level or under a higher trap density regime (trap-related recombination time <100 ns), the nonradiative recombination could always dominate over the bimolecular recombination, which also leads to an internal QY of <10%. Therefore, the slow bimolecular recombination rate in bulk perovskites limit the PeLEDs working at a relatively low carrier density. The quadratic power dependence of the recombination rate on the carrier densities of MAPbBr₃ bulk crystal (Figure 1b) confirms the bimolecular recombination in the bulk perovskite. The experimentally measured curves match well with the theoretical prediction at the low charge carrier density level (Figure 1a,c). It is noted that the first-order excitonic emission could compete effectively with the trap-mediated nonradiative recombination in the low carrier density range of \(n \leq 10^{14}\) cm⁻³ with trap states density (\(k_1 \approx 10^7\) s⁻¹) as shown in Figure 1a. Therefore, small-size PeNCs can enhance the radiative excitonic recombination with increased exciton confinement and exciton binding energy compared to bulk crystals with dominating free-carrier recombination. Moreover, the surrounded passivation of ligands can decrease the surface defects of PeNCs, which also leads to an increment of QY at low excitation density level.

**Synthesis of PeNCs and Their Luminescence Dynamics.** The colloidal MAPbBr₃ PeNCs were synthesized by using antisolvent precipitation method at room temperature. The as-synthesized solution was preaged for 6 days before being used for LEDs fabrication. In this duration, the absorption edge and the full-width-at-half-maximum (fwhm) of PL spectra keeps narrowing synchronously (Figure 1d, Supplementary Figure 1). Notably,
the PLQY was significantly enhanced from 70% in solution and 35% in film to 89% in solution and 71% in film. From nuclear magnetic resonance (NMR), the composition of the MAPbBr₃ NCs was found to remain stable during the aging time (Supplementary Table 1). However, as observed in X-ray diffraction (XRD) patterns, the MAPbBr₃ NCs show increased crystallization during this process (Figure 1e). The high-resolution transmission electron microscopy (HR-TEM) measurements (Figure 1f) illustrate that the aged PeNCs still exhibit a small crystal size (<8 nm). The lattice fringes of the MAPbBr₃ PeNCs are 2.93 and 2.65 Å, which correspond to the (200) and (210) facets of the cubic phase MAPbBr₃, respectively. With the passivation of ligands and solvent molecules, the low-crystallinity MAPbBr₃ NCs in colloidal solution possess a high PLQY. However, the passivation effect decreases drastically in film (Supplementary Table 1); thus, the defect sites become significant in quenching emission through the competition with radiation process. Though with similar passivation, the highly crystallized NCs with a low defect sites display low trap-assisted nonradiative recombination, which ensures a high PLQY in film. Even with such passivation effects, the PeNCs still show high mobility of 0.22 cm² V⁻¹ s⁻¹ for holes and 0.48 cm² V⁻¹ s⁻¹ for electrons (Supplementary Figure 2).

As illustrated in Figure 1b, for MAPbBr₃ PeNCs, \( I_{PL} [t = 0] \) is nearly linearly dependent on the carrier density from \( 5 \times 10^{14} \) to \( 2 \times 10^{17} \) cm⁻³, which suggests that the emission is mainly from the first-order excitonic radiation process, in agreement with a recent report in formamidinium lead halide nanocrystals. Below \( 5 \times 10^{14} \) cm⁻³, the trap-mediated nonradiative recombination dominates over the excitonic emission. Above \( 2 \times 10^{17} \) cm⁻³, the multiparticle Auger recombination prevails. Therefore, MAPbBr₃ PeNCs feature a high PLQY level under the charge carrier density from \( 10^{13} \) to \( 10^{16} \) cm⁻³, which matches well with the device working regime in PeLEDs (Figure 1c). Therefore, the highly efficient first-order excitonic emission suggests the MAPbBr₃ PeNCs as suitable candidates for efficient PeLEDs.

Investigation of PeLEDs. As the top most important parameter for EL device characterization, EQE, which is defined as the number of emitted photons per injected electrons, is given as the product of four different parameters:

\[
EQE = E_{in} \times E_{s/t} \times E_{rad} \times E_{out}
\]  

(3)

Therein the first parameter \( E_{in} \) indicates the charge balance factor in recombination zone. The second one \( E_{s/t} \) gives the fraction of excitons that are allowed to radiate by spin statistics. The third factor \( E_{rad} \) depicts the fraction of the intrinsic radiative efficiency of the emitter. The last one \( E_{out} \) determines the photon extraction efficiency in a device. In this work with the proposed devices using normal structure without any additional measurement for spin injection of charge carriers or enhanced outcoupling efficiency of radiation, thus, all attention was paid onto the charge balance factor in recombination zone to reach the high device efficiency promised by MAPbBr₃ PeNCs. Moreover, because of the significant role of Auger nonradiative recombination (Figure 1a,c), which is to be effectively suppressed in the proposed MAPbBr₃ PeNCs devices, a high EQE in low driving current density level was also targeted. In addition to EQE, the power efficiency, especially at high brightness level, is also a critical parameter for evaluating the potential applications of LEDs; therefore, a low driving voltage is essential.

To obtain high quality PeNCs film, a classical PEDOT:PSS layer was selected as hole injection/transport layer (HIL/HTL) for all devices in this work (Figure 2a,b and Supplementary Figures 3 and 4). As a good HIL/HTL, PEOPT:PSS may lead to emission quenching of PeNCs at HTL/PeNCs interface, but we did not observe a significant weakness of emission comparing to the PeNCs film deposited onto other polymer substrates, like poly-TPD, at an excitation wavelength of 365 nm. The insertion of an insulating thin film can improve charge balance by blocking the majority carriers, but this would probably be accompanied by high driving voltage, which would lead to high power consumption and low power efficiency.

To balance the injected hole, a blended layer of B3PYMPM:TPBi (1:X w/w) with adjustable electron transport capability by changing the ratio of two components served as an electron transport layer (ETL) (Figure 2a,b). The deeper LUMO level (~3.4 eV) and high electron conductivity \( (4.1 \times 10^{-7} \) S cm⁻¹) of B3PYMPM can lower down the barrier for...
effective electron injection and transport with low driving voltage,20,33,34 Because of the significant difference of the electron conductivity in TPBi and B3PYMPM,33 the electron transport capability of the blended ETL can be tuned in a large range (Figure 2c). According to Miller−Abrahams hopping model, electrons transport through a hopping process between the localized occupied and unoccupied state. In the blended ETL, TPBi (B3PYMPM) molecules play the role of an energy barrier (trap) to scatter the electron transfer between two B3PYMPM (TPBi) molecules, leading to a tunable electron transport capability.35 In the single carrier device with TPBi ETL, as the low electron conductivity ($4.2 \times 10^{-10} \text{ S cm}^{-1}$) and high energy barrier for electron injection caused by the high lying LUMO level of TPBi, the driving voltage is much higher than that of single carrier device with B3PYMPM ETL, which can offer a much higher level of electron conductivity and a deeper LUMO level (Figure 2a,c).33,34 For the single carrier device with B3PYMPM:TPBi (1:X, w/w) ETL, the blended layer shows a tunable electron transport capability depending on the varied component ratio (Figure 2c). To reach a high power efficiency, a n-type doped electron injection layer was used for a low driving voltage.20

Control device I with pure TPBi ETL and control device II with pure B3PYMPM ETL were selected as the baseline. As seen in Figure 2d, PeLEDs show a green color emission with a nearly ideal Lambertian profile (Supplementary Figure 5). Control device I exhibits overall performance of 15 130 cd m$^{-2}$ in $L_{\text{max}}$, 5.11% in EQEmax, and 10.82 lm W$^{-1}$ in $\eta_{\text{max}}$ (Figures 2, 3, Table 1, and Supplementary Table 2). On the other hand, control device II displays a higher $L_{\text{max}}$ of 24 410 cd m$^{-2}$ and a drastically decreased driving voltage, but a lower $\eta_{\text{max}}$ of 5.96 lm W$^{-1}$ caused by a lower EQEmax of 2.39% compared to control device I (Table 1 and Figures 2 and 3). With B3PYMPM:TPBi (1:2, w/w) ETL, control device III shows the best overall performance up to now, with $L_{\text{max}}$ of 33 570 cd m$^{-2}$, EQEmax of 7.91%, and $\eta_{\text{max}}$ of 20.18 lm W$^{-1}$ (Figures 2 and 3, Table 1, and Supplementary Table 3). In all devices, the influence of $E_{\text{inj}}$, $E_{\text{rad}}$, and $E_{\text{out}}$ can be neglected because the emitters are identical, and the difference in refractive index between TPBi and B3PYMPM is also negligible. Therefore, the significant

### Table 1. Key Parameter Comparison of PeLEDs

<table>
<thead>
<tr>
<th>device</th>
<th>structure</th>
<th>$V_{\text{on}}$ (V)</th>
<th>$L_{\text{max}}$ (cd m$^{-2}$)</th>
<th>EQEmax (%)</th>
<th>$\eta_{\text{max}}$ (lm W$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>control device I</td>
<td>ITO/PEDOT:PSS (40 nm)/MAPbBr$_3$ PeNCs (40 nm)/TPBi (30 nm)/TPBi: Cs$_2$CO$_3$ (10:1 w/w, 20 nm)/Al (100 nm).</td>
<td>3.3</td>
<td>15 130</td>
<td>5.11</td>
<td>10.82</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.4</td>
<td>16 470</td>
<td>5.06</td>
<td>7.03</td>
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<td></td>
<td></td>
<td>3.6</td>
<td>14 190</td>
<td>4.81</td>
<td>7.49</td>
</tr>
<tr>
<td>control device II</td>
<td>ITO/PEDOT:PSS (40 nm)/MAPbBr$_3$ PeNCs (40 nm)/B3PYMPM (30 nm)/B3PYMPM: Cs$_2$CO$_3$ (10:1 w/w, 20 nm)/Al (100 nm).</td>
<td>2.7</td>
<td>24 410</td>
<td>2.39</td>
<td>5.96</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.6</td>
<td>23 290</td>
<td>2.28</td>
<td>6.21</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.7</td>
<td>23 120</td>
<td>2.19</td>
<td>5.62</td>
</tr>
<tr>
<td>control device III</td>
<td>ITO/PEDOT:PSS (40 nm)/MAPbBr$_3$ PeNCs (40 nm)/B3PYMPM:TPBi (1:2 w/w, 30 nm)/B3PYMPM: Cs$_2$CO$_3$ (10:1 w/w, 20 nm)/Al (100 nm).</td>
<td>3.0</td>
<td>33 570</td>
<td>7.91</td>
<td>20.18</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.9</td>
<td>30 380</td>
<td>6.75</td>
<td>17.92</td>
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<td></td>
<td></td>
<td>3.0</td>
<td>32 730</td>
<td>7.25</td>
<td>18.47</td>
</tr>
<tr>
<td>champion device</td>
<td>ITO/PEDOT:PSS (40 nm)/MAPbBr$_3$ PeNCs (80 nm)/B3PYMPM:TPBi (1:2 w/w, 30 nm)/B3PYMPM: Cs$_2$CO$_3$ (10:1 w/w, 20 nm)/Al (100 nm).</td>
<td>3.1</td>
<td>22 830</td>
<td>12.9</td>
<td>26.07</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.0</td>
<td>43 400</td>
<td>7.53</td>
<td>17.56</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.9</td>
<td>25 410</td>
<td>10.67</td>
<td>30.3</td>
</tr>
</tbody>
</table>

$V_{\text{on}}$, turn on voltage @ 1 cd m$^{-2}$; $L_{\text{max}}$, maximum luminance; EQEmax, maximum external quantum efficiency; $\eta_{\text{max}}$, maximum power efficiency.
difference in EQE$_{\text{max}}$ among control devices is primarily caused by different charge balance in the recombination zone.

Generally, the brightness in a LED is correlated with the amount of activated emitters in the recombination zone. In all control devices, the nanocrystals distribution should be the same; hence, the higher brightness means more activated emitters in the recombination zone as the driving current density increases, more emitters were excited, leading to increment of brightness. With the highest electron supply capability of B3PYMPM, control device II, instead, gave the lowest luminous efficiency among all devices, even though its $L_{\text{max}}$ is higher than that of control device I by 61%. As the emitters were the same for all devices, the possible reason is the imbalanced charge in the recombination zone of control device II. Additionally, the red emission in Ir(piq)$_2$acac indicator device with TPBi ETL suggested the injection of remaining hole into ETL as the maximum effective radius of Förster energy transfer is 6 nm. It was thus deduced that hole is the major charge carrier in the recombination zone of control device I (Supplementary Figure 6). Otherwise, the pure PeNCs green color emission in Ir(piq)$_2$acac indicator device with B3PYMPM ETL would have suggested there was less or no hole injection into ETL compared to TPBi devices (Supplementary Figure 6). With the same hole injection, the highest EQE$_{\text{max}}$ suggested the best charge balance in control device III with medium electron supply ETL comparing to I and II (Figure 2, 3). Therefore, the lowest EQE$_{\text{max}}$ of control device II was ascribed to the overwhelming majority of electrons in the PeNCs emissive layer as the highest electron supply of B3PYMPM.

In amorphous organic semiconductor, the LUMO/HOMO level distribution was broad, of which the tail extended into the band gap as gap states. Moreover, as $\sigma = n\mu_e$ (where $\sigma$ is electron conductivity, $n$ is the electron concentration, $\mu_e$ is the electron mobility) indicates, a comparable electron mobility but significantly high electron conductivity suggest high electron concentration. Thus, here it was deduced that the amount of gap states in B3PYMPM is much higher comparing to TPBi, which can be supported by the smaller distance between the Fermi level and LUMO level of B3PYMPM comparing to TPBi (Supplementary Figure 7). In consequence, with a deeper LUMO level, there were more electrons injected into B3PYMPM layer even when the applied bias was less than the threshold voltage of 2.2 V compared to TPBi (Figure 2e). By the law of charge conservation, in a closed-loop circuit, the electrons injected into B3PYMPM layer will transit to the valence band of PeNCs by a nonradiative process, leading to leakage current (Figure 2a,e). Moreover, under high driving voltage, the excessive electrons injected into the conductive band of PeNCs will transit to the Fermi level of PEDOT:PSS by a nonradiative process as current overflow, which is another way of leakage current for control device II working in high driving current density range. The leakage of carriers that flow through the device without recombining drives down the EQE directly. Moreover, the imbalanced charge also leads to an enhanced Auger nonradiative recombination. Thus, control device II presented the lowest EQE$_{\text{max}}$ among all devices. As exhibited in Figure 2e, with blended ETL$_n$, the leakage current density was suppressed significantly by 3–4 magnitudes. In most practical LEDs, such leakage current was ascribed to device inherent structure. Therefore, we believe that, in control device III by decreasing the amount of electron injected into the recombination zone with a blended ETL, the leakage current was suppressed and the charge balance in recombination zone was improved simultaneously.

Furthermore, a thicker PeNCs layer containing more emitters but fewer pinholes will be helpful to obtain a broader recombination zone, higher luminance, and lower leakage current density. As the comparable mobility of hole and electron, the thickness change in PeNCs layer will not influence the charge balance in the recombination zone significantly. Therefore, a thicker (80 nm) PeNCs layer was deposited using a mixed THF:CB (Y:1 v/v, Y = 16) PeNCs colloidal solution, by means of which the champion device reached a record EQE$_{\text{max}}$ of 12.9% with the lowest leakage current (Figures 2e and 3a, Table 1). Assuming the light outcoupling efficiency of around 20%, the estimated internal quantum yield reaches 64.5%. The EQE$_{\text{max}}$ histogram for 15 devices from four batches describes the distribution of EQE$_{\text{max}}$ of the fabricated PeLEDs (Figure 3d). Because of the significant difference between CB and THF colloidal solution in PeNCs film deposition (Supplementary Figures 8 and 9), the reproducibility of PeNCs film deposition using THF-CB (Y:1 v/v, Y = 16) colloidal solution is moderate. To balance the EQE$_{\text{max}}$ and $L_{\text{max}}$ the peak value of 43 440 cd m$^{-2}$ in $L_{\text{max}}$ 12.9% in EQE, and 30.3 lm W$^{-1}$ in $\eta$ (at a luminance level of 2069 cd m$^{-2}$, which meets the requirement for lighting application of 1000 cd m$^{-2}$) were observed from different champion devices (Table 1, Figure 3). Furthermore, we measured the device half-lifetime to be around 5 min (Supplementary Figure 10), which is still a daunting problem for PeLEDs, and there is a considerable margin to improve. Moreover, with the same device structure except a slightly changed ratio of B3PYMPM:TPBi (1:1.5, w/w), a PeLEDs with emitter of CaPbBr$_3$ nanocrystals also offered a high performances, including a EQE$_{\text{max}}$ of 12.4%, for which details will be discussed in the following work.

For all devices, it was found that a higher EQE$_{\text{max}}$ corresponds to a lower driving current density ($j$) (Figure 3a,e). As the device structures were different, the exponential dependence was probably related to the luminescence dynamics of PeNCs. The rising of driving current density EQEs increases from very low initial values (Figure 3a), which is caused by the competition of traps. Additionally, because of the high carrier mobilities of PeNCs holes and electrons meet and recombine with low probability, which is also a reason for the negligible EQE value in the low driving current density. With the increasing driving current density, the radiative excitonic recombination takes a dominating role gradually. With the further increase of the driving current density, the Auger nonradiative recombination dominates over the excitonic recombination. However, the imbalanced charges in the recombination zone, which do not exist in the photoexcitation process, boosts the nonradiative Auger process even at the low driving current density level (Figure 3a,f). As shown by the electron single carrier PeNCs device, the excessive electrons injected into the PeNCs layer lead to a significantly quenched PL even at a low electron density ($n$) level. Though the charge balance factors are the same in the recombination zone for control device III and champion device, the thicker emissive layer of champion device can decrease the charge density, allowing for a weak Auger nonradiative recombination compared to control device III.

**Conclusion.** In summary, we have demonstrated highly efficient PeLEDs using colloidal organometal halide PeNCs. The high-quality colloidal MAPbBr$_3$ PeNCs with efficient one-
order excitonic emission were synthesized at room temperature. As the significant Auger nonradiative recombination caused by high driving current density and imbalanced charge, the PeLEDs with optimized charge balance in low driving current density level were developed and reached an EQE\textsubscript{max} of 12.9% accompanied by high $\eta$\textsubscript{max} of 30.4 lm W$^{-1}$ at high brightness above 1000 cd m$^{-2}$. This work demonstrates a breakthrough advancement toward the ultimate goal of application of PeLEDs and lays down a systematic constructive approach for researchers on halide PeLEDs in their future works.

**Methods. Materials and Chemicals.** Methylamine (MA) solution in ethanol, formamidine acetate, hydrobromic acid (HBr), hydroiodic acid (HI), lead bromide (PbBr\textsubscript{2}), lead iodide (PbI\textsubscript{2}), octylamine, and anhydrous N,N-dimethylformamide (DMF), γ-butylactone, tetrahydrofuran (THF), and chlorobenzene (CB) were all purchased from Sigma-Aldrich. PEDOT:PSS (AI 4083) was purchased from Heraeus. 1,3,5-Tris(N-phenylbenzimidazole-2-yl)benzene (TPBi), cesium carbonate (Cs\textsubscript{2}CO\textsubscript{3}), 4,6-bis(3,5-di(pyridin-3-yl)phenyl)-2-methylyprimidine (B3PYMPM), poly(4-butylyldiphenylene) (poly-TPD), and bis[1-phenylisoquinoline)-(acetylacetonate)iridium(III) (Ir(piq)\textsubscript{2}(acac)) as purchased from Lumtec. The colloidal zinc oxide (ZnO) nanoparticles (N-11) were purchased from Avantama. All chemicals were used as received.

**Lead-Halide Perovskite Preparation.** MABr. CH\textsubscript{3}NH\textsubscript{2} solution in ethanol was reacted with HBr in water with excess methylamine at 0 °C. The precipitate was recovered via evaporation at 60 °C. The yellowish raw product was washed with diethyl ether five times and dried at 60 °C in a vacuum oven for 24 h. MABr. CH\textsubscript{3}NH\textsubscript{2} solution was spin-coated onto PEDOT:PSS to form the PeNCs solution with 5 mg/mL concentration for LEDs fabrication. MAPbBr\textsubscript{3} Bulk Crystal. The 1 mmol MABr and 1 mmol PbBr\textsubscript{2} were dissolved in 0.5 mL of anhydrous DMF and 8.5 mL of γ-butylactone mixed solution with 15 μL of octylamine to form a halide perovskite precursor solution. A 0.3 mL sample of the perovskite precursor solution was dropped non-linear absorption and negligible sample damage, the relative PLQY measurements of colloidal samples was obtained by normalizing the collected PL intensity with the corresponding excitation light intensity.

**Transistor Testing.** The transistors performance was measured to extract mobility with a Keithley 4200 semiconductor parameter analyzer in air at room temperature. A Si/SiO\textsubscript{2} substrate with patterned gold electrodes was selected as a substrate to make a bottom gate field effect transistor, and a PeNCs film was deposited on top as the active layer by spin-casting.

**PeLEDs Fabrication.** The patterned ITO-glass substrates were cleaned with deionized water, acetone, and isopropanol in sequence, followed by oven-drying at 80 °C for 2 h and O\textsubscript{2} plasma treatment (FEMTO) for 15 min. PEDOT:PSS was deposited onto the substrate by spin-casting at 4000 rpm for 60 s, and annealed at 120 °C for 30 min in air. Colloidal PeNCs solution was spin-coated onto PEDOT:PSS to form the PeNCs layer. The remaining functional layers and aluminum electrodes were fabricated by thermal evaporation in a single run at a base pressure of <2 × 10$^{-4}$ Pa without breaking the vacuum. All the organics were evaporated at a rate of around 0.1−0.2 nm s$^{-1}$, and the aluminum electrodes were evaporated at a rate of 0.8−1 nm s$^{-1}$.
B3PYMPM films serve as electron transport layer (ETL). The TPBi:Cs2CO3 and B3PYMPM:Cs2CO3 films serve as the n-type doped electron injection layer (EIL), which can enhance the electron injection and decrease the driving voltage effectively.20 All blended function layers were deposited using multiosource coevaporation. The lowest unoccupied molecular orbital (LUMO) and the highest occupied molecular orbital (HOMO) of all materials were obtained from the literature.20,34,37 The valence band and conductive band of PeNCs were obtained by ultraviolet photoelectron spectroscopy (UPS) and absorption spectrum (Supplementary Figures 1 and 3).

Electron-Only Devices Fabrication. The devices structures are as follows. Device 1: ITO/ZnO (40 nm)/TPBi (30 nm)/TPBi:Cs2CO3 (10:1 w/w, 20 nm)/Al (100 nm). Device 2: ITO/ZnO (40 nm)/TPBi:B3PYMPM (2:1 w/w, 30 nm)/B3PYMPM:Cs2CO3 (10:1 w/w, 20 nm)/Al (100 nm). Device 3: ITO/ZnO (40 nm)/TPBi:B3PYMPM (1:2 w/w, 30 nm)/B3PYMPM:Cs2CO3 (10:1 w/w, 20 nm)/Al (100 nm). Device 4: ITO/ZnO (40 nm)/B3PYMPM (30 nm)/B3PYMPM:Cs2CO3 (10:1 w/w, 20 nm)/Al (100 nm). Device with PeNCs layer: ITO/ZnO (40 nm)/MAPbBr3 PeNCs (40 nm)/B3PYMPM:Cs2CO3 (10:1 w/w, 20 nm)/Al (100 nm). ZnO nanoparticle film was deposited onto the substrate by spin-casting at 3000 rpm for 60 s, and annealed at 150 °C for 10 min in air. Except the deposition of ZnO nanoparticles films, the device fabrication were the same with PeLEDs.

MAPbBr3 PeNCs Layer Deposition. Control Devices. A THF solvent dispersed PeNCs colloidal solution was selected for the PeNCs layer deposition with an optimized rotational speed of 1500 rpm (Supplementary Table 2 and Supplementary Figure 6).

Champion Devices. A mixed solvent of THF:CB (16:1 v/v) was used to disperse PeNCs to obtain a uniform colloidal solution. A programmable spin-casting was developed for deposition of PeNCs film, which consists of two steps: step 1 was chosen as 400 rpm for 3 s, and step 2 was 1500 rpm for 60 s.

PeLEDs Characterization. With a nearly ideal Lambertian emission, the voltage independent EL spectra of the PeLEDs were measured using a Photo Research Spectra Scan spectrometer PR705 at different brightness, while the luminance—current—voltage characteristics of the devices were measured simultaneously with a programmable Agilent B2902A sourcemeter and a Konica-Minolta LS-110 luminance meter in air at room temperature (Supplementary Figure Sb). The lifetime test was carried out by a programmable sourcemeter of Agilent B2902A and a calibrated Si-photo-detector (Thorlabs, FDS-1010CAL).

PL of Electron Single Carrier Device. PL spectra were recorded through a micro-Raman spectrometer (Horiba, LABRAM HR800) with a single-grating setup and a 442 nm solid state laser were used as the excitation laser. The bias was applied by Agilent B1500A semiconductor device parameter analyzer.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.8b00789.

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Author Contributions

H.V.D., Q.H.X., and E.H.S. planned and guided the work. F.Y. designed and fabricated the devices, performed the device experiments, analyzed the data with assistance of S.T.T., and discussed the results with H.V.D. J.X. designed and synthesized the perovskite materials and performed the material experiments and data analyses with assistance of J.X.Z., L.L.Z., and L.N.Q, and discussed the results with Q.H.X. The luminescence lifetime measurement was made by R.S. The luminescence dynamics study was performed by G.C.X. The TEM measurement was made by Y.W.Z. The UPS measurement was made by S.C. and A.H. All authors discussed and commented on the manuscript.

Notes

The authors declare no competing financial interest.

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