

Enhancing the Brightness of Cesium Lead Halide Perovskite Nanocrystal Based Green Light-Emitting Devices through the Interface Engineering with Perfluorinated Ionomer

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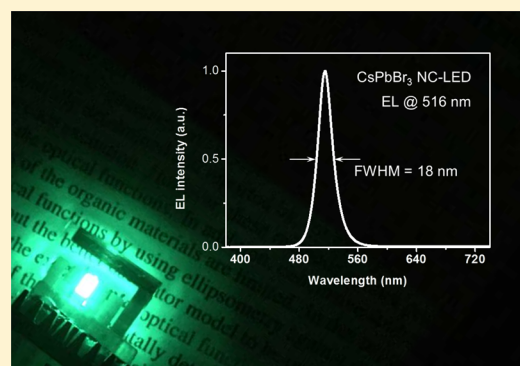
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ABSTRACT: High photoluminescence quantum yield, easily tuned emission colors, and high color purity of perovskite nanocrystals make this class of material attractive for light source or display applications. Here, green light-emitting devices (LEDs) were fabricated using inorganic cesium lead halide perovskite nanocrystals as emitters. By introducing a thin film of perfluorinated ionomer (PFI) sandwiched between the hole transporting layer and perovskite emissive layer, the device hole injection efficiency has been significantly enhanced. At the same time, PFI layer suppressed charging of the perovskite nanocrystal emitters thus preserving their superior emissive properties, which led to the three-fold increase in peak brightness reaching 1377 cd m^{-2} . The full width at half-maximum of the symmetric emission peak with color coordinates of (0.09, 0.76) was 18 nm, the narrowest value among perovskite based green LEDs.

KEYWORDS: Perovskites, CsPbBr_3 nanocrystals, light-emitting device, interface engineering, perfluorinated ionomer



Perovskite nanocrystals (NCs) synthesized by solution-phase chemistry approach have been recently reported by several groups.^{1–7} Notably, inorganic cesium lead halide perovskite NCs (CsPbX_3 , $X = \text{Cl, Br, and I}$ or mixed halide system Cl/Br and Br/I) exhibited high photoluminescence (PL) quantum yield (QY) reaching 90% in solution, with narrow emission peaks and wide color gamut.¹ Following the recent breakthroughs in synthetic chemistry, a number of applications of perovskite NCs has been proposed literally within the last months, including their use as single photon emitters,^{8,9} lasing,^{10,11} and as active layer components in light-emitting devices (LEDs).¹² Related properties of perovskite NCs have been also addressed, such as ultrafast interfacial charge transfer,¹³ suppressed PL blinking,¹⁴ and the possibility for a fast anion exchange resulting in emission color tuning.^{15,16} Emission color purity and high PL QY of CsPbX_3 NCs make them especially attractive for generating electroluminescence (EL), with first LEDs using CsPbX_3 NC emitters reported by Zeng and co-workers.¹² However, the turn-on voltages (V_{on}) of their devices were much higher than the band gap energy of the NCs used, suggesting that the carrier injection from charge-transporting layers (CTL) into NCs was inefficient. The existence of charge injection barriers is a universal issue, which has to be taken into account when designing CTL and emissive layers of LEDs.^{17–19} In our study, an inefficient hole injection in the conventional CsPbBr_3 NC based LEDs has been

confirmed by estimation of absolute values for NC energy levels obtained via ultraviolet photoelectron spectroscopy (UPS) measurements. This issue can be addressed by introducing interfacial layers, the strategy widely used in optoelectronic devices.^{20–24} For solar cells, an interface layer between a photoactive layer and electrodes significantly influences both the built-in potential and the charge carrier extraction.^{25–27} For LEDs, it allows to change the work function of the modified CTL, which enables charge carriers to be easily injected, and ensures efficient radiative recombination of excitons in the emitting layer.^{19,28,29} We incorporated a perfluorinated ionomer (PFI) interlayer between the hole-transporting layer (HTL) and the perovskite NC emissive layer of our LEDs, which resulted in 0.34 eV increase of the valence band maximum of HTL. Besides, the introduction of PFI layer was also found to maintain charge balance of NC emitters and to preserve their superior emissive properties in the film. As a result, our LEDs achieved a peak brightness of 1377 cd m^{-2} , the highest reported value for CsPbX_3 NC based LEDs so far. Pure green light emission has been observed under a voltage as low as 2.5 V, indicating that an efficient and barrier-free charge injection into the NC emitters

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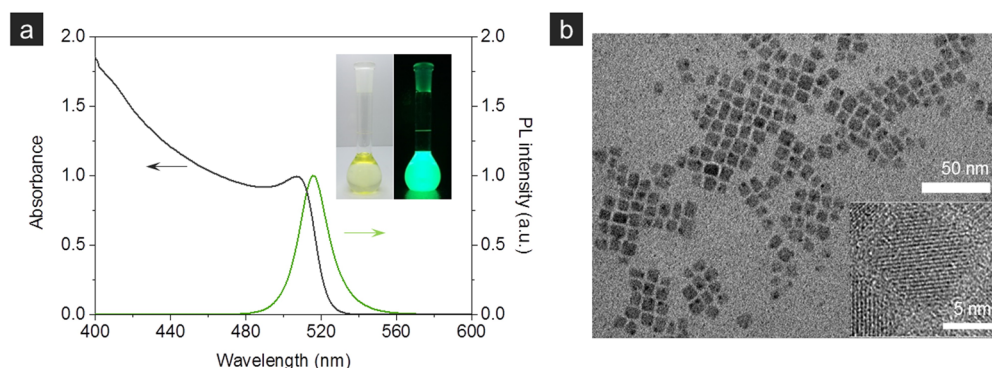


Figure 1. (a) UV-vis absorption and photoluminescence (365 nm excitation wavelength) spectra of CsPbBr₃ NCs dissolved in toluene, with insets showing the photograph of NC solutions under ambient light (left) and UV irradiation (right). (b) TEM image of CsPbBr₃ particles, with an inset showing HRTEM image of a single nanocrystal.

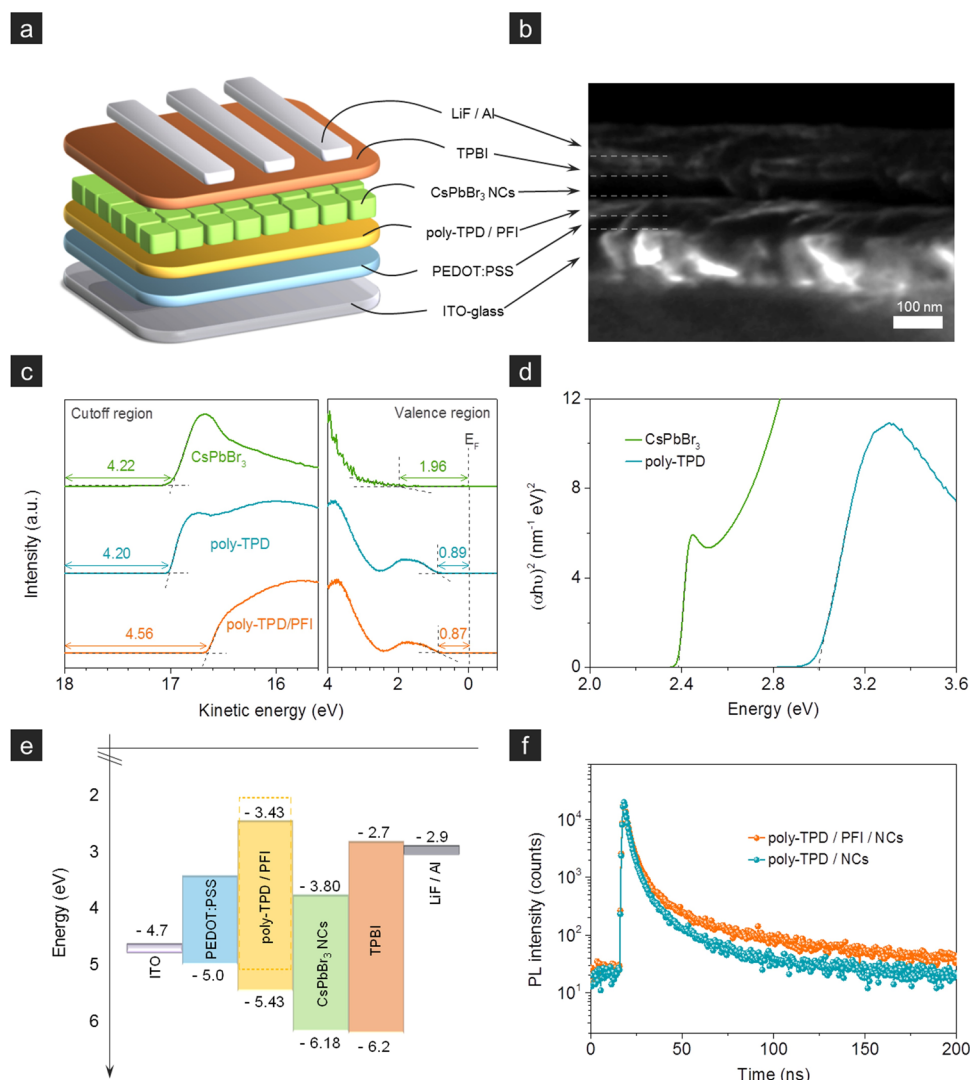


Figure 2. (a) Device structure and (b) cross-sectional SEM image of the CsPbBr₃ NC LED. (c) UPS spectra of CsPbBr₃ NC film, poly-TPD film, and poly-TPD/PFI film deposited on ITO glass substrates. (d) Tauc plot of CsPbBr₃ NC films and poly-TPD films on quartz substrates. (e) Overall energy band diagram of the LED structure. The poly-TPD and NC emitter energy bands were determined from UPS and optical absorption measurements, while others were taken from refs 39–41. (f) PL decay curves of a CsPbBr₃ NC film on a poly-TPD/glass substrate, and as a film on a PFI/poly-TPD/glass substrate.

was realized. The full width at half-maximum (fwhm) of the EL peak was 18 nm, the narrowest value compared with previously reported perovskite film based green LEDs.^{30–35}

Results and Discussion. UV-vis absorption and PL emission spectra of CsPbBr₃ NCs dissolved in toluene are shown in Figure 1a, together with a photograph of NC solution

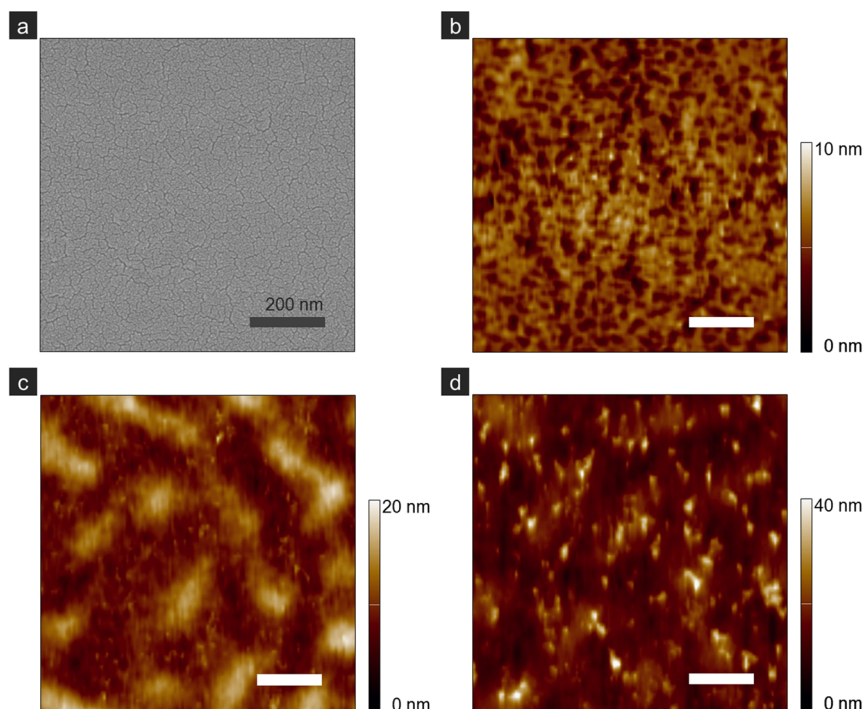


Figure 3. (a) SEM image showing the top view of the ITO/PEDOT:PSS/poly-TPD/PFI device. AFM images of poly-TPD layer (b, $R_{\text{rms}} = 1.3$ nm), PFI layer (c, $R_{\text{rms}} = 2.5$ nm), and CsPbBr₃ NC layer (d, $R_{\text{rms}} = 4.8$ nm). All white scale bars are 1 μm .

exhibiting bright green emission (365 nm excitation wavelength) with high color purity (fwhm = 18 nm) as an inset. The absorption and PL peaks are located at 507 and 516 nm, respectively. Figure 1b presents a typical transmission electron microscopy (TEM) image of CsPbBr₃ NCs, showing the presence of rather monodisperse cubic-shaped NCs with an edge length of 10–11 nm.

A schematic diagram and the cross-sectional scanning electron microscope (SEM) image of the conventional LED device with multilayers of patterned indium tin oxide (ITO) as the anode, poly(ethylenedioxythiophene):polystyrenesulfonate (PEDOT:PSS, 25 nm) film, poly(*N,N'*-bis(4-butylphenyl)-*N,N'*-bis(phenyl)-benzidine) (poly-TPD, 40 nm) film as the HTL, CsPbBr₃ NC film (40 nm) as the emitting layer, 1,3,5-tris(*N*-phenylbenzimidazol-2-yl) benzene (TPBI, 40 nm) film as the electron-transporting layer (ETL), and LiF/Al as the cathode are shown in Figure 2a,b, respectively. PEDOT:PSS was used as a buffer layer on top of ITO to obtain stable and pinhole-free electrical conduction across the device and to increase the anode work function. The hole mobilities of poly-TPD and electron mobilities of TPBI are both around $1 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$,^{36,37} which makes it easy to achieve charge transport balance through optimization of CTL thickness. The charge injection is one of the key points to realize efficient LEDs.³⁸ To gain a better understanding of the injection process in our devices, UPS measurements (Figure 2c) have been performed on CsPbBr₃ NC films in order to map the valence band maximum (VBM) and the conduction band minimum (CBM). The Tauc plot of a CsPbBr₃ NC film on quartz substrate (Figure 2d) reveals a bandgap of 2.38 eV. Thus, we derived the VBM and CBM values for CsPbBr₃ NC as -6.18 and -3.80 eV, respectively. In a similar way, the highest occupied molecular orbital (HOMO) of poly-TPD was determined to be -5.09 eV.

Figure 2e shows a schematic of the flat-band energy level diagram of our LED device, with energy level values for ITO,

PEDOT:PSS, TPBI, and LiF/Al taken from refs 39–41. High CBM of TPBI allows for the exciton energy transfer from TPBI molecules to NCs, while the back transfer is negligible. Therefore, when using TPBI as ETL, we do not need to introduce any additional hole blocking layer to realize narrow NC-LED EL spectra dominated by NC emission.⁴² The commonly used poly-TPD HTL has a favorable spectral overlap with NC absorption spectra (Figure 2d), indicating that poly-TPD could also transfer its excitons to NC emitters, but the transfer efficiency is expected to be low for the big difference (1.09 eV) of the HTL and NC VBM values. We have reduced this large injection barrier by coating the poly-TPD film with a thin layer (~ 5 nm) of perfluorinated ionomer (PFI). The PFI has been a widely used material in optoelectronic devices, such as the blended PFI/PEDOT:PSS films used as hole extraction layers in solar cell applications.^{43,44} PFI has also been used as the interfacial layer on the top and the bottom surface of HTLs⁴⁵ and mixed into other HTLs such as metal oxides.⁴⁶ Benefiting from the self-organization ability of the polymers used, the surface of such blended films is enriched of PFI, resulting in an increase of the surface work function (a molecularly thin PFI overlayer would set up a surface dipole that provides the high work function),²⁹ which is beneficial for the device power conversion efficiency. Here, we utilized a simple physisorption of a thin PFI film on a poly-TPD layer, which also increases the VBM of this HTL material, alleviating the issue of optimizing the ratio of constituting components if fabricating blended films. Figure 2c displays UPS spectra taken from poly-TPD film and PFI coated poly-TPD film on top of ITO. Considering the 3.0 eV optical energy bandgap of poly-TPD film (Figure 2d), we calculated the resulting CBM and VBM values, which are summarized in Figure 2e. An increase of VBM by 0.34 eV has been achieved after the PFI modification, indicating that the hole injection barrier was successfully reduced.

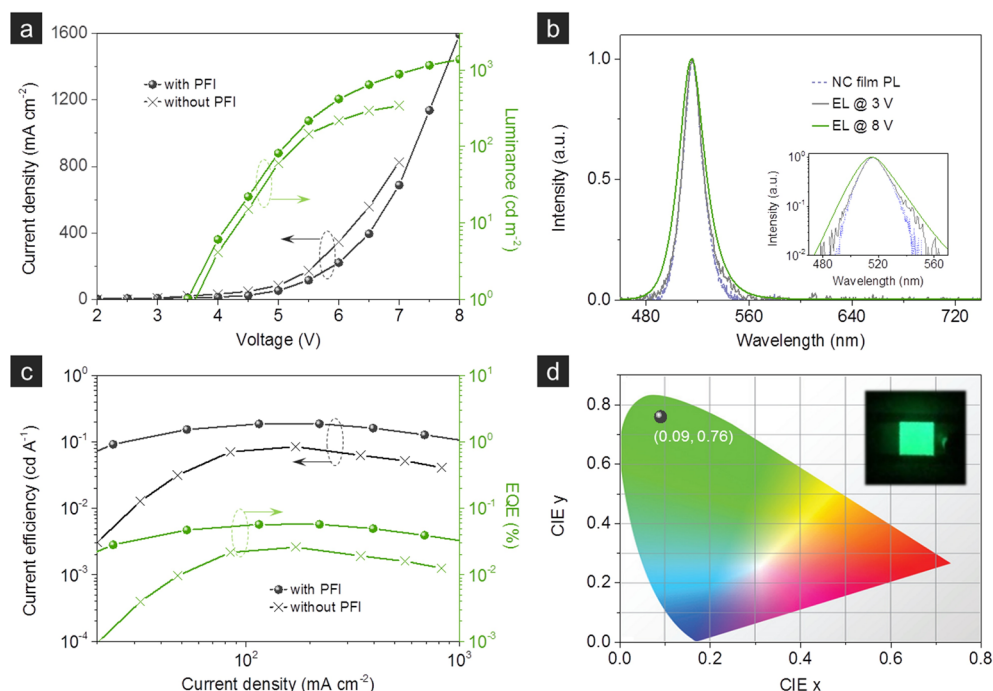


Figure 4. (a) Current density and brightness vs driving voltage of devices with or without PFI interface modifier. (b) PL spectrum of a NC film, and EL spectra of the LED using interface engineering under different applied voltages, together with their log-scale spectra given as an inset. (c) External quantum efficiency and current efficiency vs current density of devices with or without PFI interface modifier. (d) CIE coordinates for the EL spectrum under an applied voltage of 8 V in (b). The photograph in (d) shows a working device (the PFI modified one with an emitting area of $5 \times 5 \text{ mm}^2$) at applied voltage of 5 V.

The PFI layer plays yet another important role in achieving bright CsPbBr₃ NC LEDs, helping to maintain strong EL of perovskite NCs in the film due to prevention of NC charging, known to be a common source of luminescence quenching.^{47–49} In the devices studied here, for perovskite NC emitters in a direct contact with the poly-TPD film, a spontaneous charge transfer process occurs due to the large energy level difference, leading to charged NCs with lower emission efficiency. As shown in Figure 2f, perovskite NCs from the samples with a PFI layer have much slower PL decay (15 ns) compared with those without PFI (8 ns), indicating that PFI prevents NCs from getting charged.

Although the thickness of PFI layer was only $\sim 5 \text{ nm}$, we found those films complete and homogeneous. The surface SEM image of ITO/PEDOT:PSS/poly-TPD/PFI device shows that the spin-coated PFI layer possesses a uniform surface morphology after washing by butanol solvent (Figure 3a). Atomic force microscope (AFM) image of the same PFI layer provided a root-mean-square roughness (R_{rms}) value of 2.5 nm, which is slightly larger than that of the ITO/PEDOT:PSS/poly-TPD structure (1.3 nm, Figure 3b). The spin-coated layer of CsPbBr₃ NCs was also uniformly formed on top of the PFI film, with R_{rms} value of 4.8 nm (Figure 3d).

Figure 4a presents the voltage-dependent variations of luminance and current density for two CsPbBr₃ NC based LEDs, with and without PFI layers. The turn-on voltage (which is commonly defined in literature as the voltage necessary to detect luminescence of 1 cd m^{-2}) of devices with PFI was 3.5 V, slightly smaller than for devices without PFI (3.6 V). The current densities of the devices with PFI were substantially lower throughout the entire bias range, for instance, 688 and 826 mA cm^{-2} at 7 V for LEDs with and without PFI, respectively. The peak luminances of 1377 cd m^{-2} (at 8 V) and 340 cd m^{-2} (at 7 V) from devices with and without PFI, respectively, were detected.

All the above results indicate that for devices with PFI layer, the charge injection into the emitting layer becomes easier, resulting in higher injection efficiency. Notably, pure green emission could be observed from the PFI modified devices at a voltage as low as 2.5 V, indicating that an efficient and barrier-free charge injection into the NC emitters was achieved.³⁸ As a direct consequence of lower current densities along with higher luminance values, the efficiency of devices with PFI layer increased. As shown in Figure 4c, the peak current efficiency (CE) and the external quantum efficiency (EQE) of a device without PFI reached 0.08 cd A^{-1} and 0.026%, respectively, while a device with PFI showed device efficiency of the peak values of CE of 0.19 cd A^{-1} and EQE of 0.06% with an enhancement of 1.4 and 1.3 times, respectively.

Figure 4b shows normalized PL spectrum of a CsPbBr₃ NC film, and EL spectra of a CsPbBr₃ NC LED with PFI layer at different applied voltages. Both the EL spectra measured under a voltage lower than V_{on} (3 V) and at the luminescence maximum (8 V) were located at the same wavelength of 516 nm. The bandwidths of the EL spectrum broadens only slightly, from 18 to 20 nm, along with the increase of the applied voltage, which may originate from an increased longitudinal optical-phonon interaction accompanied by a large degree of exciton polarization by higher electric field.^{50,51} For most semiconductor quantum dot based LEDs, their EL spectra exhibit a red-shift comparing to the PL peaks of corresponding NC films, which originates from the change of the dielectric function of the surrounding medium⁵⁰ and/or from the energy transfer from smaller to larger NCs in the ensemble.^{52,53} The PL spectra of CsPbBr₃ NC films had their maxima at 516 nm with a bandwidth of 18 nm, which fully matched the EL spectra of the respective LEDs at 3 V, evidencing on the efficient suppression of the energy transfer.⁵⁴ The LED displayed EL solely from CsPbBr₃ NCs without any noticeable contribution from any charge transport materials,

indicating that the NC emitters serve as the primary exciton recombination centers during device operation and further suggesting that the balanced charge carrier transport has been achieved. Their symmetric emission corresponds to Commission Internationale de l'Eclairage (CIE) color coordinates of (0.09, 0.76) (Figure 4d), which fully meets the demands for display applications. The reproducibility of the optimized devices was very high; over 80% of the LEDs provided the brightness over 1300 cd m^{-2} .

The EL stability of the devices under continuous operation at a constant voltage of 5 V has been evaluated in a N_2 filled glovebox at room temperature. As shown in Figure 5, the EL signal slightly increases during the first 2 min and decays to 50% of its initial value after 10 min. No change in the shape of the EL curves was observed.

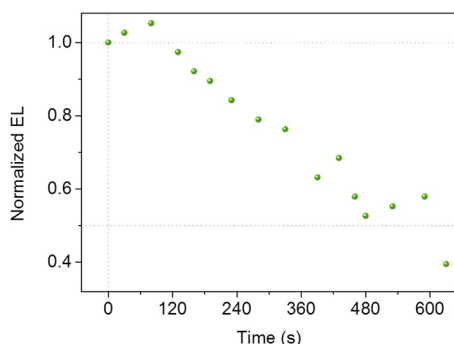


Figure 5. Evolution of the normalized EL signal of a working LED under nitrogen atmosphere at a constant voltage of 5 V.

Conclusions. Because of the intrinsically deep lying VBM of CsPbBr_3 NCs, a considerable hole injection barrier at their interface with HTL exists in such NC-based LEDs. We addressed this issue through proper interface engineering applying an easy processable layer of PFI, which facilitates the hole injection and prevents the NC emitters from charging. By optimizing charge injection and transport, we demonstrated devices with the highest luminescence value among perovskite NC based LEDs, and the narrowest emission bandwidth among perovskite based green LEDs reported so far.

Methods. Materials. Oleic acid (OA, 90%) and 1-octadecene (ODE, 90%) were purchased from Alfa Aesar. Oleylamine (OLA, 80–90%) was purchased from Aladdin. Poly(N,N' -bis(4-butylphenyl)- N,N' -bis(phenyl)-benzidine) (poly-TPD) was purchased from 1-Material. Trioctylphosphine (TOP, 97%), perfluorinated ionomer (PFI, Nafion 1100EW), 1,3,5-tris(N -phenylbenzimidazol-2-yl) benzene (TPBI), LiF, Cs_2CO_3 , and PbBr_2 were purchased from Sigma-Aldrich. Poly(ethylenedioxythiophene):polystyrenesulfonate (PEDOT:PSS) was purchased from Clevios.

Synthesis of CsPbBr_3 NCs. The synthesis procedures were carried out following the previously published method.¹ For the synthesis of Cs-oleate, Cs_2CO_3 (0.8 g), OA (2.5 mL), and ODE (30 mL) were added into a 100 mL 3-neck flask, degassed, and dried under vacuum for 1 h at 120°C . The mixture was heated to 150°C under N_2 until a clear solution was obtained. For the synthesis and purification of CsPbBr_3 NCs, ODE (10 mL) and 0.188 mmol PbBr_2 (0.138 g) was loaded into a 50 mL three-neck flask, degassed, and dried by applying vacuum for 1 h at 120°C . Dried OLA (1 mL) and dried OA (1 mL) were injected to the flask at this temperature. After the solution became clear, the

temperature was raised to 180°C and Cs-oleate solution (0.8 mL, 0.1 M in ODE, preheated to 100°C before injection) was quickly injected. Five seconds later, the reaction mixture was cooled down to room temperature by an ice–water bath. The reaction mixture was separated by centrifuging for 10 min at 5000 rpm. After centrifugation, the precipitate was redispersed in 2 mL of toluene and centrifuged again for 10 min at 12,000 rpm. After repeating this step one more time, the supernatant was discarded, and the precipitate was redispersed in 2 mL of toluene.

Device Fabrication. Patterned ITO coated glass was cleaned with soap, deionized water, ethanol, chloroform, acetone, and isopropanol successively and treated in UV-ozone for 15 min. PEDOT:PSS was spin-coated onto ITO glass at 3000 rpm for 40 s, and annealed in air at 120°C for 20 min. The substrate was transferred into a glovebox, and a solution of poly-TPD (dissolved in chlorobenzene with a concentration of 15 mg mL^{-1}) was spin-coated onto the PEDOT:PSS film at a speed of 3000 rpm for 40 s and annealed at 110°C for 30 min. PFI was diluted in a component solvent (0.5 mg mL^{-1}), spin-coated onto poly-TPD at 3000 rpm for 40 s, and washed by butanol before annealed at 100°C for 10 min. The component solvent contains 97% of butanol, 2.4% of lower-weight aliphatic alcohols, and 0.6% water (volume ratios). Perovskite NC active layers were spin-cast from their colloidal solution at 1000 to 2000 rpm. TPBI, LiF, and Al layers were sequentially deposited by thermal evaporation in a vacuum deposition chamber (1×10^{-7} Torr). The Al cathode was deposited through a shadow mask defining device area of 0.06 cm^2 .

Characterization. Transmission electron microscopy (TEM) images were obtained on a FEI Tecnai F20 microscope. Scanning electron microscopy (SEM) images were taken with a JEOL JSM-7500F system. Atomic force microscopy (AFM) images were recorded on a VEECO DICP-II microscope. Ultraviolet photoelectron spectroscopy (UPS) spectra are collected using a PREVAC system. Absorption spectra were measured on a PerkinElmer Lambda 950 UV–vis–NIR spectrometer and photoluminescence (PL) spectra on a Cary Eclipse spectrofluorimeter. Time-resolved PL measurements were performed on a time-correlated single-photon counting (TCSPC) system with a 320 nm laser as the excitation light source. The current–voltage–luminance characteristics were measured using a Keithley 2635 source measure unit in conjunction with a Newport 818-UV Si photodiode centered over the light-emitting pixel at a fixed distance. The LED brightness was determined from the fraction of light that reaches the photodetector. The EL spectra were recorded with an Ocean Optics QE65000 spectrometer coupled to an optical fiber.

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Notes

The authors declare no competing financial interest.

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