Ultrapure Green Light-Emitting Diodes Using Two-Dimensional Formamidinium Perovskites: Achieving Recommendation 2020 Color Coordinates

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*Supporting Information

ABSTRACT: Pure green light-emitting diodes (LEDs) are essential for realizing an ultrawide color gamut in next-generation displays, as defined by the recommendation (Rec.) 2020 standard. However, because the human eye is more sensitive to the green spectral region, it is not yet possible to achieve an ultrapure green electroluminescence (EL) with a sufficiently narrow bandwidth that covers >95% of the Rec. 2020 standard in the CIE 1931 color space. Here, we demonstrate efficient, ultrapure green EL based on the colloidal two-dimensional (2D) formamidinium lead bromide (FAPbBr3) hybrid perovskites. Through the dielectric quantum well (DQW) engineering, the quantum-confined 2D FAPbBr3 perovskites exhibit a high exciton binding energy of 162 meV, resulting in a high photoluminescence quantum yield (PLQY) of ~92% in the spin-coated films. Our optimized LED devices show a maximum current efficiency (ηCE) of 13.02 cd A−1 and the CIE 1931 color coordinates of (0.168, 0.773). The color gamut covers 97% and 99% of the Rec. 2020 standard in the CIE 1931 and the CIE 1976 color space, respectively, representing the "greenest" LEDs ever reported. Moreover, the device shows only a ~10% roll-off in ηCE (11.3 cd A−1) at 1000 cd m−2. We further demonstrate large-area (3 cm2) and ultraflexible (bending radius of 2 mm) LEDs based on 2D perovskites.

KEYWORDS: Colloidal 2D perovskites, formamidinium lead bromide, dielectric quantum wells, Rec. 2020, ultrapure green, light-emitting diodes

The color gamut offered by a display, which is defined the coverage in the Commission Internationale de l’Eclairage (CIE) color space, is determined by the emission properties of the pure red (R), green (G), and blue (B) light sources. The newly defined International Telecommunication Union (ITU) Recommendation BT 2020 (Rec. 2020) standard requires the monochromatic RGB primaries with extremely narrow bandwidth, which, in particular, sets a significant challenge for the green emitters. This is due to the fact that the human eye is more sensitive in the green spectral region and allows the green emitters. This is due to the fact that the human eye is more sensitive to the green spectral region, it is not yet possible to achieve an ultrapure green electroluminescence (EL) with a sufficiently narrow bandwidth that covers >95% of the Rec. 2020 standard in the CIE 1931 color space. Here, we demonstrate efficient, ultrapure green EL based on the colloidal two-dimensional (2D) formamidinium lead bromide (FAPbBr3) hybrid perovskites. Through the dielectric quantum well (DQW) engineering, the quantum-confined 2D FAPbBr3 perovskites exhibit a high exciton binding energy of 162 meV, resulting in a high photoluminescence quantum yield (PLQY) of ~92% in the spin-coated films. Our optimized LED devices show a maximum current efficiency (ηCE) of 13.02 cd A−1 and the CIE 1931 color coordinates of (0.168, 0.773). The color gamut covers 97% and 99% of the Rec. 2020 standard in the CIE 1931 and the CIE 1976 color space, respectively, representing the “greenest” LEDs ever reported. Moreover, the device shows only a ~10% roll-off in ηCE (11.3 cd A−1) at 1000 cd m−2. We further demonstrate large-area (3 cm2) and ultraflexible (bending radius of 2 mm) LEDs based on 2D perovskites.
formation of the local ferroelectric domains\textsuperscript{15,16} and large polarons\textsuperscript{17} screens the Coulombic interactions between the charged carriers and the charged defects. Clearly, the fact that the hybrid perovskite systems show higher tolerance to materials defects provides a fundamental basis for the high-performance light-emitting devices.\textsuperscript{18–21} Another practical motivation is that, compared to the inorganic QD technology, the perovskite emitters generally possess a narrower bandwidth (fwhm of $\sim 20–25$ nm) in the green spectral range, thereby standing a better chance to approach the Rec. 2020 color coordinates. Indeed, since the first demonstration of the perovskite-based light emitting diodes (LEDs) at room temperature in 2014,\textsuperscript{22} considerable research effort has been generated to develop the colloidal emitters with high photoluminescence quantum yields (PLQYs) and the device architectures that facilitate radiative recombination.\textsuperscript{23–29} Nevertheless, to date, although the green perovskite LEDs with a high current efficiency (up to 42.9 cd A$^{-1}$) have been demonstrated,\textsuperscript{13} the emission chromaticity is not sufficiently close to the Rec. 2020 standard.

A central challenge toward achieving the Rec. 2020 color coordinates with the hybrid perovskites is to precisely fine-tune the emission wavelength without sacrificing the PLQY. Specifically, considering the emission bandwidth of the hybrid perovskites, we estimate the optimal emission wavelength to be $\sim 525–530$ nm, which is slightly lower than those for the bulk methylammonium (MA), formamidinium (FA), and cesium (Cs) lead bromide perovskites ($\sim 540–560$ nm).\textsuperscript{30} To this end, although the chloride doping seems to be a straightforward approach to introducing a degree of blue-shifting, it often results in a drop in the PLQY (e.g., ref 31). In addition, the solubility for the chloride precursors in the regularly used polar solvents (e.g., N,N-dimethylformamide, DMF) is low,\textsuperscript{32} thereby increasing the process complexity in the bulk-film or nanocrystal synthesis. In this respect, the colloidal two-dimensional (2D) perovskites\textsuperscript{33–41} might be the ultimate solution due to the following reasons. First, a precise control over the layer number enables a quantized and sizable bandgap due to the quantum confinement effect.\textsuperscript{42} Second, the quantum confinement effect in turn enhances the exciton binding energy, $E_B$, which, in principle, boosts the PLQY.\textsuperscript{30}

In this work, we demonstrate the ultrapure green LEDs based on the colloidal 2D FAPbBr$_3$ hybrid perovskites, with the stacking number of perovskite unit cells, $n$, to be around 7 to 10. Through the dielectric quantum well (DQW) engineering, the quantum-confined 2D FAPbBr$_3$ perovskites exhibit a high exciton-binding energy of 162 meV, resulting in a high photoluminescence quantum yield (PLQY) of 92% in the spin-coated films. Our optimized device shows a maximum current efficiency of 13.02 cd A$^{-1}$, which is among one of very few reports that reach double digits. Moreover, the EL emission locates at 529 nm with a fwhm of 22.8 nm, reaching the CIE 1931 color coordinates of (0.168, 0.773). Accordingly, for the first time, we report an ultrawide color gamut that covers 97% and 99% of the Rec. 2020 standard in the CIE 1931 and the CIE 1976 color space, respectively, representing the “greenest” LED ever reported. We further demonstrate large-area (3 cm$^2$) and ultraflexible (bending radius of 2 mm) LEDs based on the 2D perovskites.

The 2D FAPbBr$_3$ and MAPbBr$_3$ perovskites are synthesized by using modifications of the synthesis method developed in our previous report (for details, see the Supporting Information).
In short, the first precursor, formamidinium bromide (FABr) or methylammonium bromide (MABr), was dissolved in a polar solvent, and the second precursor, lead bromide (PbBr₂), was dissolved in DMF. A pair of precursor solutions were next added drop-wise to a toluene solution containing two surfactants, oleic acid (OLA) and octylamine (OA). Colloidal crystallization was triggered immediately due to a low solubility of the precursors in toluene. The resulting precipitates were then collected and dispersed in toluene to obtain colloidal dispersions. Before device fabrication, a small amount of a low-dielectric-constant (low k) compound, poly(methyl methacrylate), PMMA, was added into the FAPbBr₃ colloidal solution to take advantage of DQW effect, as will be discussed later.

Figure 1a presents the photograph of the 2D FAPbBr₃ solution and spin-coated thin film after adding PMMA under ultraviolet (UV) excitation. The PLQY of the spin-coated film composed of the 2D FAPbBr₃−PMMA complex is determined to be 92%, which to our knowledge, represents the highest value ever reported in the perovskite thin films. The absorption and photoluminescence (PL) spectra of 2D MAPbBr₃ and FAPbBr₃ dispersions are shown in Figure S1, with the emission wavelength, λPL, centered at 521 and 530 nm, respectively. A degree of blue-shifting, which results from the quantum confinement effect, is observed and allows us to estimate the thickness of the 2D perovskite dispersions to be n ≈ 7−10. The 2D nature of the perovskite dispersions is endorsed by the X-ray diffraction (XRD) and transmission electron microscopy (TEM), as shown in Figures 1b,c. In the XRD spectra for both compounds, the three dominant peaks at around 15°, 30°, and 45° are assigned as the (100), (200), and (300) planes for the cubic perovskite structure. The lattice constant is therefore determined to be 5.9 and 6.0 Å for 2D MAPbBr₃ and FAPbBr₃, respectively, consistent with those reported in the bulk counterparts. Nevertheless, the peak intensity associated with other planes, such as (110) at ~21° and (210) at ~34°, is very weak, suggesting that the crystalline symmetry possibly not be 3D. In the grazing-incidence wide-angle X-ray scattering (GIWAXS) pattern (Figures 1b inset and S2), the Debye–Scherrer ring for the (100) plane, corresponding to q = 10.5 nm⁻¹, becomes the strongest on the qz axis, consistent with our previous observations in the spin-coated solids of 2D perovskite dispersions. Rectangular colloidal 2D nanocrystals were clearly identified on the TEM grids (Figures 1c and S3). In the FAPbBr₃ case, its emission wavelength locates at 529 ± 1 nm, corresponding to a blue shift of ~20 nm relative to that in the bulk counterpart (n > 10). As revealed by the TEM image (Figure 1c), we observed nearly monodispersed nanocrystals with vertical and horizontal orientations, giving the average lateral length and thickness of 14.0 and 4.9 nm, respectively. Considering the unit cell thickness (~0.6 nm), n ≈ 7−10 is therefore determined. Note that the observed dark spots within the 2D structures have been suggested to be the degradation product of perovskites caused by the focused electron beam. Figure S4 presents the particle size distribution (PSD) characterized using the dynamic light scattering (DLS). A narrow distribution with an average hydrodynamic diameter of 18.2 nm was observed, confirming the colloidal monodispersity (for details, see the Supporting Information).

The concept of the dielectric confinement is realized by surrounding the 2D perovskite dispersions with a wide-bandgap, low-k compound (PMMA is used here), acting as the barrier. The resulting complex consists of a matrix of the dielectric quantum wells in which the exciton binding energy is boosted due to a reduction of the effective dielectric constant. Taking the 2D FAPbBr₃ as an example, we illustrate that the DQW engineering significantly changes the photophysical properties of the perovskite films based on the following observations: (i) an increase of PLQY from 88% (solution) to 92% (complex), as shown in Table 1; (ii) a decrease of the exciton lifetime (τavg) from 42.3 (neat film) to 24.2 ns (complex), as determined by the time-resolved (TR) PL spectroscopy in Figure 1d; and (iii) an increase of the exciton binding energy (E₀) from 149.3 (neat film) to 161.6 meV (complex), as determined by the temperature-dependent PL measurements in Figure 1e (for details, see the Supporting Information and Figure S5). Compared to the pure hybrid perovskites, in which the E₀ and the thermal energy at room temperature (~25 meV) are in the same order of magnitude, the synthesized 2D perovskites are much more excitonic, although the degree of quantum confinement is small. We therefore infer that the dielectric confinement effect, which has been proven to be long-range, is responsible for the binding energy enhancement because the colloidal nanocrystals are dispersed in low-dielectric-constant media (toluene or PMMA). The orientation, packing, and self-assembly behavior of the surrounding molecules determines the effective dielectric constant of the environment, which may be worth further investigation. In principle, a lower surrounding dielectric constant leads to a higher E₀, thereby decreasing the lifetime and increasing the PLQY, which is qualitatively supported by the photophysical characteristics of our 2D FAPbBr₃ samples (see Table 1).

Table 1. Photophysical Characteristics of 2D Perovskites-Based Solutions and Films

<table>
<thead>
<tr>
<th>material</th>
<th>sample type</th>
<th>λPL (nm)</th>
<th>fwhm (nm)</th>
<th>ΦPLQY (%)</th>
<th>τavg (ns)</th>
<th>E₀ (meV)</th>
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<tbody>
<tr>
<td>FAPbBr₃</td>
<td>solution</td>
<td>531</td>
<td>21.8</td>
<td>88</td>
<td>62.7</td>
<td>-</td>
</tr>
<tr>
<td>FAPbBr₃</td>
<td>thin film</td>
<td>530</td>
<td>22.6</td>
<td>90</td>
<td>42.3</td>
<td>149.3</td>
</tr>
<tr>
<td>FAPbBr₃ + PMMA</td>
<td>solution</td>
<td>530</td>
<td>21.6</td>
<td>86</td>
<td>77.7</td>
<td>-</td>
</tr>
<tr>
<td>MAPbBr₃</td>
<td>thin film</td>
<td>530</td>
<td>22.6</td>
<td>92</td>
<td>24.2</td>
<td>161.6</td>
</tr>
<tr>
<td>MAPbBr₃</td>
<td>solution</td>
<td>521</td>
<td>23.6</td>
<td>79</td>
<td>83.8</td>
<td>-</td>
</tr>
<tr>
<td>MAPbBr₃</td>
<td>thin film</td>
<td>522</td>
<td>22.5</td>
<td>85</td>
<td>29.4</td>
<td>152.6</td>
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</tbody>
</table>

DOI: 10.1021/acs.nanolett.7b01544
Nano Lett. 2017, 17, 5277–5284
The LED devices were then fabricated using the synthesized 2D MAPbBr₃ and FAPbBr₃, which yield EL with the emission wavelength at 524 and 529 nm, respectively. Because the color coordinates for 2D FAPbBr₃ are closer to the Rec. 2020 standard; thereafter, we focus on the FAPbBr₃-based devices. The color coordinates of the 2D FAPbBr₃ LED presented here (this work) in the CIE 1931 color space are shown in Figure 2b. The Rec. 2020, National Television System Committee (NTSC), and Rec. BT 709 standards are also included. Accordingly, we demonstrate an ultrapure green EL with the color coordinates of (0.168, 0.773), which, to our knowledge, represents the purest green emission ever reported, relative to the Rec. 2020 standard (0.170, 0.797). The color coordinates for other technologies, including organic LED (OLED), QD-Led, InGaN LED, and perovskite LED (PeLED), are also attached for comparison. Indeed, the chromaticity reported here results in an ultrawide gamut that covers 97% and 99% of the Rec. 2020 standard in the CIE 1931 and CIE 1976 color space, respectively, as summarized in Table 2, which has never been achieved in LED devices thus far.

The device architecture and energy diagram for our LED device is shown in panels a and b of Figure 3 (for details, see the Supporting Information). The energy levels of valence band maximum (VBM) for 2D FAPbBr₃ and MAPbBr₃ perovskites were obtained using the ultraviolet photoelectron spectroscopy (UPS) (for details, see the Supporting Information and Figure S6). The conduction band minimum (CBM) energy levels were then back calculated by subtracting the optical bandgap values from the PL emission wavelengths. We use a typical three-layered structure, which consists of a hole-transport layer (HTL), an emission layer (EML), and an electron-transport layer (ETL) (for molecular structures, see Figure S7). A number of experiments were carried out to optimize the HTL and ETL materials in the FAPbBr₃ devices, as summarized in Table 3. The control device, without PMMA in the EML (ITO/PEDOT·PSS/2D FAPbBr₃/TPBi/LiF/Al), shows a maximum current efficiency (ηₑₑₑ) of 1.58 cd A⁻¹ and a maximum luminance (Lmax) of 229 cd m⁻², corresponding to the maximum power efficiency (ηₚₑ) of 1.29 lm W⁻¹ and the external quantum efficiency (ηₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑ_userdata
other FAPbBr₃ perovskite-based LEDs, e.g., ref 55. More importantly, the optimal device only showed a ∼10−15% roll-off in the η_CE (11.3 cd A⁻¹), η_PE (10.2 lm W⁻¹), and η_ext (2.74%) at 1000 cd m⁻². We attribute the superior device performance to a balanced charge injection and efficient exciton recombination resulting from the following mechanisms: (i) a low highest occupied molecular orbital (HOMO) level (−5.4 eV) in the poly-TPD HTL and a high lowest unoccupied molecular orbital (LUMO) level (−3.3 eV) in the 3TPYMB ETL,⁵⁶,⁵⁷ thereby enabling cascade carrier injection; (ii) comparable carrier mobilities in HTL and ETL;⁵⁶,⁵⁸ and (iii) an effective carrier confinement due to a high LUMO level (−2.0 eV) in HTL and a low HOMO level (−6.8 eV) in ETL.⁵⁶,⁵⁷ Figure S11b presents a histogram of η_CE based on the fabricated 52 devices with the optimal conditions, exhibiting an average η_CE of 10.31 cd A⁻¹. A higher L_max of 4425 cd m⁻² is further achieved by spin-coating the EML twice, but the efficiencies are slightly reduced. We also demonstrate high-efficiency 2D MAPbBr₃-based devices (η_CE = 8.43 cd A⁻¹, η_PE = 7.64 lm W⁻¹, η_ext = 2.06%, and L_max = 2735 cd m⁻²) (Figures S12 and S13), as described in detail in the Supporting Information.

Finally, we demonstrate large-area and ultra-flexible LEDs based on the 2D FAPbBr₃ perovskites. Figure 4a presents the EL spectra of a large-area (3 cm²) device under different driving voltages, showing emission wavelengths of 530 ± 1 nm and a

Table 3. Electroluminescence Characteristics of 2D-Perovskites-Based LEDs

<table>
<thead>
<tr>
<th>HTL</th>
<th>EML</th>
<th>ETL</th>
<th>V_on (V)</th>
<th>η_CE (cd A⁻¹)</th>
<th>η_PE (lm W⁻¹)</th>
<th>η_ext (%)</th>
<th>L_max (cd m⁻²)</th>
<th>λ_EL/fwhm (nm)</th>
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<tbody>
<tr>
<td>FAPbBr₃</td>
<td>3TPYMB (35 nm)</td>
<td>2.7</td>
<td>4.53</td>
<td>4.10</td>
<td>1.14</td>
<td>301</td>
<td>S29/22.5</td>
<td></td>
</tr>
<tr>
<td>poly-TPD</td>
<td>FAPbBr₃</td>
<td>2.8</td>
<td>6.46</td>
<td>5.80</td>
<td>1.57</td>
<td>2608</td>
<td>S29/22.4</td>
<td></td>
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<tr>
<td>poly-TPD</td>
<td>FAPbBr₃ + 15 wt % PMMA</td>
<td>2.8</td>
<td>10.00</td>
<td>7.71</td>
<td>2.31</td>
<td>3267</td>
<td>S29/22.6</td>
<td></td>
</tr>
<tr>
<td>poly-TPD</td>
<td>FAPbBr₃ + 15 wt % PMMA</td>
<td>3.0</td>
<td>8.22</td>
<td>7.38</td>
<td>1.99</td>
<td>4425</td>
<td>S30/22.5</td>
<td></td>
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<tr>
<td>FAPbBr₃</td>
<td>3TPYMB (45 nm)</td>
<td>2.75</td>
<td>12.76</td>
<td>12.95</td>
<td>3.02</td>
<td>575</td>
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<td>2.75</td>
<td>13.02</td>
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<td>3.04</td>
<td>2939</td>
<td>S29/22.8</td>
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<td>FAPbBr₃ + 8 wt % PMMA</td>
<td>3.0</td>
<td>1.58</td>
<td>1.29</td>
<td>0.38</td>
<td>229</td>
<td>S29/23.5</td>
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<tr>
<td>poly-TPD</td>
<td>TBPi (35 nm)</td>
<td>3.0</td>
<td>4.05</td>
<td>3.54</td>
<td>1.01</td>
<td>717</td>
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<td>2.30</td>
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<td>1.72</td>
<td>1.54</td>
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<td>3.4</td>
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<td>TBPi (35 nm)</td>
<td>2.6</td>
<td>1.28</td>
<td>1.15</td>
<td>0.31</td>
<td>529</td>
<td>S30/24.3</td>
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<tr>
<td>poly-TPD</td>
<td>TBPi (35 nm)</td>
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<td>8.43</td>
<td>7.64</td>
<td>2.06</td>
<td>2735</td>
<td>S24/23.5</td>
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Figure 3. Device characteristics of the 2D FAPbBr₃ perovskite-based LEDs. (a) Schematic device architecture. (b) Energy diagram for the materials used in the optimized device. (c) Current density and luminance as a function of voltage. (d) Current efficiency and EQE as a function of current density.
LED at di FAPbBr3 2D perovskites. (a) EL spectra of a large-area (20 mm × 15 mm) LED at different driving voltages. Inset: Photographs of a large area device operating at 4.0 V (top) and 5.0 V (bottom). Logo reproduced with permission. Copyright 2017 ETH Zürich, Switzerland. (b) EL spectra of a flexible LED on the polyimide substrate at different driving voltages. Inset: Photographs of a flexible perovskite LED at different bending radii (scale bar: 1 cm).

The fwhm of 22.6 nm, which are also consistent with the PL spectrum (Figure 2a). More importantly, with the optimized device architecture, the device performance does not compromise significantly (\(\eta_{\text{ce}} = 8.4 \text{ cd A}^{-1}, \eta_{\text{fl}} = 6.6 \text{ lm W}^{-1}\), and \(\eta_{\text{ext}} = 2.0\%\)) (Figure S14), and to our knowledge, it is the largest perovskite LED ever reported, with record high efficiency.26,59 Figure 4b exhibits an ultra-flexible device on a thin polyimide substrate (50 μm). The insets show photographs of a working device under different degrees of bending. More details about device fabrication, characteristics, and bending tests can be found in section 3.2 and Figure S15 in the Supporting Information. Similarly, the EL spectra under different driving voltages are consistent and stable. Because of the ultra-flexible substrate considered here, a minimum bending radius down to 2 mm was achieved (Figure S15e). The turn-on voltage (\(V_{\text{on}}\)) for the flexible device is slightly higher due to a higher resistivity of the ITO layer deposited on the polyimide surface. The large-area, ultra-flexible devices demonstrated here give promise of the low-cost, large-scale production of the ultrapure green LEDs for the next generation of displays.

In summary, we demonstrate that in the colloidal 2D FAPbBr3 perovskite system, one can introduce a small degree of quantum confinement that results in a proper bandgap and bandwidth for achieving the Rec. 2020 standard. In combination with its monodispersity in solution, the emission wavelength in the final LED devices is nearly identical to that in the diluted solution, clearly outperforming the OLED and QD technologies. Through the DQW and interface engineering, the LED devices exhibit ultrahigh color purity and high efficiency, which allow us to achieve the Rec. 2020 color coordinates for the first time. The results presented here open an avenue toward the realization of large-area, low-cost LEDs with an ultra-wide gamut. We believe that further optimization of device performance and the development of perovskite-based optoelectronics will be greatly facilitated by the original concepts proposed here.

ASSOCIATED CONTENT

Supporting Information
The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.7b01544.

Additional details on synthetic procedures, characterization techniques and parameters, materials, fabrication, and characterization of perovskite LED devices. Supplementary figures including absorption and PL spectra of 2D perovskites, GIWAXS diffraction patterns, additional STEM images, temperature-dependent PL spectra, additional TRPL spectra, UPS spectra of 2D perovskites, molecular structures of other materials used in LEDs, AFM and SEM images for emissive layer, a cross-section SEM image of LED, device performance and architecture for small- and large-area devices, and photographs and EL characteristics of flexible perovskite LEDs. Tables showing comparative device performance. (PDF)

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*S.K. and J.J. contributed equally to this work. S.K., J.J., and C.J.S. conceived and designed the experiments. S.K. designed, fabricated, and characterized the LED devices. J.J. synthesized and characterized the colloidal 2D perovskites. K.N. helped in LED fabrication, and F.J. helped with the colloidal 2D perovskite synthesis. Y.H.K. performed the TRPL and temperature-dependent PL measurement in the guidance of T.W.L. and characterized the colloidal 2D perovskites. C.J.S. performed the STEM analysis. S.K. and C.J.S. performed the AFM and SEM analysis. S.K. and C.J.S. performed the STEM analysis with the supervision of T.W.L. and characterized the colloidal 2D perovskites. K.N. helped in LED fabrication, and F.J. helped with the colloidal 2D perovskite synthesis. Y.H.K. performed the TRPL and temperature-dependent PL measurement in the guidance of T.W.L. C.W. carried out the UPS analysis. Y.C.C. performed the GIXD analysis. S.K. and T.T. performed the AFM and SEM analysis. C.J.S. performed the STEM analysis with the supervision of W.J.S. T.T. made the schematic device figure. S.K. and C.J.S. prepared all the graphics and co-wrote the paper. All authors discussed the results and commented on the manuscript.

Notes
The authors declare no competing financial interest.

ACKNOWLEDGMENTS
S.K., J.J., and C.J.S., are grateful for financial support from ETH startup funding. The authors thank Professor Gerhard Tröster and Mr. Giuseppe Cantarella in ETH Zurich, Switzerland for providing the flexible polyimide substrates. The authors thank Dr. Frank Krumeich for HRTEM analysis. This work was also supported by the National Research Foundation of Korea.

Nano Letters
2017, 17, 5277–5284
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