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Enhanced Direct White Light Emission Efficiency in Quantum Dot Light-Emitting Diodes via Embedded Ferroelectric Islands Structure

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1 Abstract

White light emission is of great importance in our daily life as it is the primary source of light 2 3 indoor and outdoor as well as day and night. Among various materials and lighting technologies, intensive efforts have been made to quantum dots based-light-emitting diode (QLEDs) because 4 5 of outstanding optical properties, facile synthesis, and bandgap tunability of QDs. Despite the 6 fact that QLEDs are able to present various colors in a visible range, realizing efficient direct white light emission has been a challenge as white light emission can only be achievable 7 8 through stacking and patterning of QD films or mixing of different sizes of QDs. This 9 inevitably involves energy band mismatch at interfaces, leading to degradation of device performance. Here, we introduce a new effective method to improve white QLED 10 11 performances through embedding a ferroelectric islands structure which induces an electric field to effectively modulate the energy band at the junction interface. The formation of 12 favorable energy landscape leads to efficient charge transport, improved radiative 13 recombination, and consequently high EQE in the white QLEDs. In addition, we demonstrate 14 that our new approach is proved to be effective in different color temperature ranging from 15 16 3000 to over 120,000 Kelvin.

17 Introduction

In 2014, Nobel prize was given to researchers who invented gallium nitride (GaN)-based blue 18 light emitting diodes (LEDs). This opened up great advancement in white light emission, which 19 contributed to saving massive amount of energy as one fourth of electricity is being consumed 20 for the lighting purpose over the world.^[1] Ever since, tremendous research efforts have been 21 paid to improve the efficiency of white LEDs.^[2-9] However, white light emission was mostly 22 achieved by wavelength conversion because direct white light emission required the LEDs to 23 simultaneously emit different color spectra. Therefore, over the past decades, synthesis of the 24 active materials with the appropriate bandgaps was one of the major challenges to achieve 25 26 direct white light emission.

Colloidal quantum dots (QDs), in particular, cadmium selenide-zinc sulfide core shell 27 QDs (CdSe@ZnS), are considered to be the prospective active materials in lightings and 28 displays as they possess outstanding optical properties, such as color tunability, narrow 29 emission width, high photoluminescence quantum yield (PLQY), high stability, and solution-30 processability for facile fabrication.^[10-13] The QDs can be driven electrically in QD-LEDs (in 31 short, QLEDs hereafter) and they have demonstrated high brightness, high external quantum 32 efficiency (EQE), device stability, and low power consumption.^[14-18] In particular, white 33 QLEDs have attracted considerable attention because the QDs are technologically favorable to 34 35 achieve direct white light emission by simply mixing red (R), green (G), and blue (B) color QDs.^[19-22] More importantly, for future displays with high definition, industries have put 36 intensive efforts in integrating white light into a pixel together with RGB. The solution 37 processability of the mixed white QD provides cost-effective and facile fabrication process, 38 which is attractive and desirable to industrial applications. Nevertheless, the existence of 39 different sizes of QDs in a mixed RGB QD solution can limit the overall performance of white 40

QLEDs due to the difficulties in predicting the energy band alignments that govern charge balance, radiative recombination, and driving current. This has limited to develop an effective method to improve the performance of a white QLED. As a result, a new universal approach is required to add a driving force to tailor the energy band alignment so that charge transport and luminescence within the LEDs can be improved to achieve high performance in direct white light emission and high resolution in future display technology.

Here, we introduce a ferroelectric islands layer using poly(vinylidene fluoride-47 trifluoroethylene), P(VDF-TrFE), to effectively modulate energy band levels and induce an 48 additional electric field which facilitates charge carrier transport. This structural modification 49 provides direct white light emission via a much simpler way to fabricate white LEDs with 50 higher device performance. In addition, our new method is applicable to devices with different 51 color temperature for direct white light emission without a wavelength conversion. White 52 QLEDs with the P(VDF-TrFE) islands layer exhibit 37 and 48% improvement in EQE 53 compared to reference devices for warm white and daylight white color, respectively. In 54 addition, we demonstrate universality of our new method in various white color temperatures 55 56 ranging from 3,000 to 120,000 Kelvin (K), corresponding to from warm to cool white color. The white QLEDs with different color temperatures all exhibit significant improvement in 57 device performances, which suggests that our new method is a facile and effective way to 58 enhance direct white light emission efficiency. 59

60

61 **Results and Discussion**

Figure 1(a) illustrates the structure of a ferroelectric effect QLED (FE-QLED), consisting of
indium tin oxide (ITO), poly(3,4-ethylenedioxythiophene) polystyrene sulfonate
(PEDOT:PSS), Poly(9,9-dioctylfluorene-alt-benzothiadiazole) (TFB), CdSe/ZnS QDs, zinc

65 oxide nanoparticles (ZnO NPs), and aluminum (Al) electrode where P(VDF-TrFE) is deposited 66 either between the TFB and QD layer or between the QD and ZnO NP layer. To fabricate a FE-QLED, we used a spin-coating method to demonstrate the solution processability of the FE-67 QLED (details of fabrication method are shown in Methods). As the transmission electron 68 69 microscopy (TEM) cross sectional image of the FE-QLED shows in Figure 1(b), the thickness of each layer was approximately 45 nm for both PEDOT:PSS and TFB, 15 nm for CdSe/ZnS 70 ODs, and 30 nm for ZnO NPs. The TEM image in Figure 1(c) shows RGB mixed CdSe/ZnS 71 72 QDs in a ratio of 1:2:9 without any aggregation where the photoluminescence quantum yield (PLQY) of red, green, and blue QDs was 84%, 81%, and 83%, respectively. Figure 1(d) shows 73 PL characteristics of the RGB mixed QD solution with 75% PLQY, exhibiting three distinct 74 wavelengths at ~625 nm, ~525 nm, and ~450 nm. A slight decrease in PLQY of the mixed QD 75 solution compared to the PLQY of each RGB QD solution is due to Förster resonance energy 76 77 transfer (FRET) from large bandgap ODs to small bandgap ODs, namely from blue to green and red light emissive QDs.^[21-25] This phenomenon becomes more apparent by exhibiting the 78 decrease in the PLQY in the QD film where the distance between QDs is reduced as shown in 79 80 Figure S1 in Supporting Information (SI).^[26] The PL characteristics of QD films showed an opposite trend compared to those of the QD solutions: the intensity of PL in the films is $I_{red} >$ 81 $I_{green} \approx I_{blue}$ whereas the PL intensity in the solutions is $I_{blue} > I_{green} \approx I_{red}$. This resulted from the 82 increased FRET process in the film state due to reduced distance between QDs.^[23,27] As a result, 83 84 a much higher volume of blue QDs were required than that of green and red QDs in the mixed QD solution for white light emission evidenced by the RGB mixing ratio 1:2:9, which is well 85 consistent with previous reports that employed larger fraction of the QDs with higher bandgap 86 than the QDs with lower bandgap.^[19,23,28] 87



For a ferroelectric layer, we employed ferroelectric polymer P(VDF-TrFE) due to the

fascinating material properties of P(VDF-TrFE), such as chemical inertness, low fabrication 89 temperatures, photostability, and its large electric polarization at even one nanometer 90 thickness.^[29,30] Ferroelectric P(VDF-TrFE) polymer was directly deposited on either the TFB 91 or QD layer. Well-distributed P(VDF-TrFE) islands were formed when it was spin-casted on 92 93 both films as shown in Figure 1(e) and Figure S2(a) and (b) in SI, which is due to the similar surface energy of the TFB and QD layer as indicated by the contact angle (Figure S3 in SI). 94 The size and coverage of the P(VDF-TrFE) island structure on TFB and OD layer was 95 96 controlled by employing either spin-coating or spin-casting method as well as changing the 97 concentration of P(VDF-TrFE) solutions and the optimum condition was attained at the concentration of 0.2 wt% with the spin-casting method as shown in Figure S4 in SI. The 98 average height of the P(VDF-TrFE) islands was approximately 7 nm as shown in Figure S2(c) 99 in SI. It is worth noting that P(VDF-TrFE) does not absorb light in the visible range due to its 100 high bandgap, ~ 6 eV, which holds the same for the ZnO layer (~3.4 eV).^[31,32] As shown in 101 Figure S5 in SI, the transmittance of the P(VDF-TrFE) and P(VDF-TrFE) on the ZnO layer in 102 the visible range was almost similar, which demonstrates that there was negligible light 103 104 reflection from the P(VDF-TrFE) islands structures. In order to confirm the ferroelectric 105 properties of the islands layer, X-ray diffraction (XRD) was performed and monitored its distinctive peaks. As shown in Figure 1(f), the P(VDF-TrFE) islands layer deposited on the 106 107 ZnO/ ITO layer exhibited the peak near $2\theta \sim 20^\circ$, indicating that the P(VDF-TrFE) islands layer formed the β -phase, i.e., a ferroelectric phase.^[30] To further confirm that the island structures 108 are formed by the P(VDF-TrFE) solution, we further analyzed X-ray photoelectron spectra 109 110 (XPS). Figure S6(a) shows four prominent C states in C 1s deconvoluted spectra: ~ 284.50 eV of organic contaminants, ~ 286.00 eV of saturated hydrocarbon CH₂, ~ 288.40 eV of C-F-H, 111 and ~ 290.50 eV of CF₂. In addition, F 1s spectrum with the peak position ~ 687.40 eV was 112

identified in Figure S6(b).^[33,34] These results indicate that the island structure is formed by
coating the P(VDF-TrFE) solution.

115 When negative and positive voltages are applied to the Al and ITO electrodes, respectively, 116 electric dipoles are aligned in a way that negative charges to the ITO side and positive charges 117 to the Al side in the β -phase P(VDF-TrFE) layer, which modulates local energy band levels 118 near the P(VDF-TrFE) layer. By judiciously using this phenomenon, we observed significant 119 enhancement in the QLED performances, which will be discussed shortly.

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Figure 2 illustrates the schematics of energy band diagram in three different types of QLEDs 121 used in this study. The energy level of each layer was referred to literature reported previously. 122 ^[20,21] We define each FE-QLED with respect to the location of P(VDF-TrFE) islands layer as 123 follows: (1) a reference device without the P(VDF-TrFE) layer (Figure 2(a) and (d)); (2) a T/Q 124 FE-QLED with the P(VDF-TrFE) layer between the TFB and QD layer (Figure 2(b) and (e)); 125 and (3) a Q/Z FE-QLED with the P(VDF-TrFE) between the QD and ZnO layer (Figure 2(c) 126 and (f)), respectively. At low forward bias (Figure 2(a)-(c)), electric dipoles in the P(VDF-TrFE) 127 128 islands layer start aligning towards negative to ITO (anode) and positive to Al (cathode). Due to the low forward bias condition, a dipole polarization field is relatively weaker than the 129 polarization field at high forward bias condition (Figure 2(e) and (f)). As a result, energy levels 130 131 near the P(VDF-TrFE) islands layer are slightly bent whereas the reference cell does not show 132 any band bending effect as shown in Figure 2(a)-(c).

When higher potential is applied to the QLEDs, there are two mechanisms causing energy band shift: (1) shift of conduction and valence levels of the TFB and ZnO layers by the applied bias; and (2) the energy band bending at the edge between the TFB and QD layer (Figure 2(e)) or between the QD and ZnO layer (Figure 2(f)) due to the ferroelectric field of the P(VDF-

TrFE) layer. As shown in Figure 2(f), the energy band bending in the Q/Z FE-QLED is 137 138 favorable for electron transport, and therefore, charge carrier transport is facilitated in this device structure. However, the T/Q FE-QLED (Figure 2(e)) forms a higher barrier for holes at 139 the interface between the TFB and the QD layer, which limits the efficient transport of charge 140 141 carriers. Moreover, the energy band bending in the T/Q FE-QLED (Figure 2(e)) causes a leakage current and consequently non-radiative recombination at the TFB and QD interface 142 due to the proximity of energy levels between the valence level of TFB and the conduction 143 level of the QDs, leading to the degraded device performance. 144

145

To confirm our conjecture, we characterized the performance of each device type. Figure 3 146 illustrates the device characteristics of warm and daylight white FE-QLEDs where the terms, 147 warm and daylight white, were used based on the color temperature of the device.^[35] As it was 148 predicted, the device performance was modulated depending on the existence of the P(VDF-149 TrFE) islands layer and its inserted location in the device. All the devices were measured by 150 applying from low to high potentials to effectively investigate the dipole polarization effect in 151 the QLEDs.^[36-38] For both warm and daylight white Q/Z FE-QLEDs (Figure 2(c) and (f)), 152 higher luminance (approximately 20% and 30% improvement, respectively) and EQE 153 (approximately 37% and 48% improvement, respectively) were observed compared to the 154 155 reference cells as shown in Figure 3(a) and Table 1. It is worth noting that the turn-on voltage (Von) of the Q/Z device was 0.1 V higher than that of the T/Q one (Table 1). This is because of 156 a notch formed between the conduction band level of QD and ZnO layer at the low applied bias 157 condition (Figure 2(c)). The significant enhancement in the device performance is attributed to 158 the energy band level modification by the dipole polarization of the P(VDF-TrFE) islands layer, 159 constructing a favorable energy band structure for the efficient charge carrier transport as well 160

161 as increased radiative recombination. In addition, the FE-QLEDs exhibited high long-term stability, maintaining 90 % of the L/L0 ratio with an initial luminance (L0) of $\sim 1000 \text{ cd/m}^2$ for 162 more than 250 min as shown in Figure S7 in SI. QLEDs were encapsulated using an epoxy and 163 a glass coverslip. Measurement was conducted in ambient air condition at room temperature. 164 165 On the contrary, increased leakage current was observed in the T/Q FE-QLEDs as shown in Figure S8 and S9 in SI, which is due to the lowered energy level difference between the valence 166 band level of TFB and conduction band level of QDs caused by the electric dipoles. This led 167 to the inefficient charge carrier transport and accordingly higher rates of non-radiative 168 recombination. As a result, both luminance and EQE of the devices were deteriorated. Details 169 of device performance are shown in Figure S8, S9, and Table 1. It is worth noting that there is 170 a trade-off between the induced ferroelectric field and more coverage of the P(VDF-TrFE) 171 layer on either TFB or QD layer due to the insulating nature of P(VDF-TrFE). Therefore, the 172 173 introduction of the P(VDF-TrFE) island structure is crucial to open an enough channel for charges to be transported (Figure S4 in SI). In addition, the height of P(VDF-TrFE) was 174 carefully controlled for our study so that the P(VDF-TrFE) island structure did not penetrate 175 176 through the layer deposited on it.

Figure 3(b) and (e) show the distinct EL characteristics of the Q/Z devices for warm and 177 daylight white devices, respectively. Correlated color temperature (CCT) at maximum 178 179 luminance (at 8 V of applied bias) was found to be 3000K for the warm white and 6500K for 180 daylight white QLED. Interestingly, to tune the color temperature, far larger amount of blue QDs were needed: for warm white, the ratio of RGB QDs was 1:3.5:3.5 whereas RGB ratio 181 was 1:2:9 for daylight white emission. This is due to the FRET from blue to green and red QDs, 182 which is well consistent with the PLQY measurement, i.e. when the mixed QDs formed a QD 183 layer, PLQY of blue QDs significantly decreased as FRET increased due to the reduced dot-to-184

dot distance which was discussed in Figure 1.^[21-26,28] However, the severe imbalance of the 185 RGB ratio leads to degradation of white QLED performance because a turn-on voltage of a 186 QLED would increase due to the high ratio of blue QDs with the larger bandgap as evidenced 187 by the turn-on voltage of warm and daylight white QLEDs in Table 1. Therefore, smaller 188 189 amount of blue QDs is desirable with the same CCT value. In this regard, for daylight white emission, we employed QDs with a deep blue color, or shorter wavelength, to avoid 190 performance degradation. Commission internationale de l'éclairage (CIE) chromaticity is 191 192 shown in Figure 3(c) and (f) where CIE coordinate for warm and daylight white was found to be (0.4473, 0.4165) and (0.3151, 0.3039) at maximum luminance, respectively. Gradual color 193 modulation from red to white color in CIE chromaticity was observed as the applied potential 194 increased. 195

196

197 To demonstrate the universality of our new approaches for enhanced white light emission, white QLEDs with various CCT values with and without the ferroelectric islands layer were 198 fabricated. The details of RGB mixing ratio are shown in Table S1 in SI. Figure 4(a) illustrates 199 200 EQE improvement of various FE-QLEDs where higher CCT values indicate that the light emission includes higher portion of blue light. Luminance-EQE plots and EL characteristics 201 are shown in S10-S12 in SI and device parameters are summarized in Table S2 in SI. It was 202 203 found that the improvement in EQE was more prominent in devices with higher CCT values. 204 We ascribed this to larger amount of surface traps in blue QDs due to the high surface area to volume ratio than those of red and green QDs.^[39,40] Because larger amount of blue QDs was 205 206 required to have higher CCT values as shown in Table S1, the RGB mixed QD solution with more blue QDs have more surface traps that lead to non-radiative recombination, deteriorating 207 intensity of light emission, power efficiency, and EQE.^[41-44] This phenomenon was also 208

209 observed in PLQY measurement of the mixed QD solutions with higher volume ratio of blue 210 QDs. As shown in Figure S13, addition of blue QDs to the mixed solution exhibited relatively small increase in PL at blue wavelength whereas decrease in PL at green and red were 211 noticeable, which led to decrease in PLQY from 26.35% to 24.14%. Therefore, the EQE 212 213 improvement was more prominent in devices with higher CCT values by the employment of P(VDF-TrFE) layer which facilitated charge carrier transport and consequently reducing non-214 radiative recombination at the surface traps in small size blue QDs. Corresponding CIE 215 coordinates were marked in the Figure 4(b), demonstrating the universality of the ferroelectric 216 effect-enhanced QLEDs performance for direct white light emission with various color 217 temperatures. Finally, performances of white QLEDs in this work were compared with other 218 219 works reported in recent 10 years as shown in Table S3 in Si.

220

221 In summary, we have demonstrated an enhanced direct white light emission by judiciously employing a ferroelectric polymer P(VDF-TrFE) islands layer. Electric dipoles in the 222 embedded P(VDF-TrFE) layer were polarized by the application of bias, which resulted in 223 224 energy band bending at the local area. Favorable energy band bending, formed by the P(VDF-TrFE) layer between QD and ZnO layer, facilitated charge carrier transport and consequently 225 enhanced device performance with high luminance. Our new strategy further demonstrated its 226 227 versatile ability to improve performances of white QLEDs with different CCT values, ranging 228 from warm to cool temperature. The facile but effective and versatile new approach would bring great opportunities in advancing direct white light emission which is necessary for 229 230 pursuing our daily lives.

231 Methods

232 Material preparation: Pixelated ITO substrates, PEDOT:PSS solution and TFB powder were purchased from Ossila. CdSe@ZnS QDs were synthesized following previous works with 233 some modifications.^[45,46] ZnO nanoparticles (NPs) were synthesized using previously reported 234 recipe by our group.^[47,48] Pixelated ITO substrates were cleaned using acetone, 1-propanol, and 235 warm deionized water for 10 minutes, respectively. Cleaned ITO substrates were dried by 236 blowing nitrogen, and then, UV-Ozone treatment was carried out on the substrates for 5 237 minutes. A concentration of a TFB, P(VDF-TrFE), CdSe/ZnS QDs, and ZnO NPs solution was 238 8 mgml⁻¹, 0.2 wt%, 12.5 mgml⁻¹, and 25 mgml⁻¹, respectively. 239

Device fabrication: PEDOT:PSS was spin-coated at the speed of 3000 rpm for 45 seconds and 240 then baked at 150 °C in ambient air for 20 minutes. After the annealing, the PEDOT:PSS coated 241 substrates were transferred to a nitrogen-filled glove box, and then annealed again at 150 °C 242 243 for 15 minutes. TFB (8 mgml⁻¹) in chlorobenzene was spin-coated on the PEDOT:PSS layer at the speed of 4000 rpm for 45 seconds, which was followed by thermal annealing at 130 °C for 244 30 minutes. CdSe@ZnS QDs (12.5 mgml⁻¹) in hexane were spin-casted on the TFB layer at a 245 speed of 4000 rpm for 30 seconds, and then thermally annealed at 90 °C for 10 minutes. ZnO 246 NPs (25 mgml⁻¹) in ethanol were spin-coated on the QD layer at the speed of 3000 rpm for 30 247 seconds, which was followed by thermal annealing at 80 °C for 10 minutes. P(VDF-TrFE) (0.2 248 wt%) in 2-bunatone was spin-casted on either TFB layer or QD layer at 4000 rpm for 30 249 250 seconds. Then, thermal annealing was performed to form β -phase at 130 °C for 20 minutes. Finally, Al electrode (100 nm) was deposited using a thermal evaporator. The active area of a 251 252 QLED was 0.045 cm^2 .

253 *Device characterization*: All QLEDs were passivated in the glove box after deposition of the 254 Al electrode, and then, measurements were performed using HAMAMATSU PMA-12 connected with Keithley 2400 source meter. The cross-sectional TEM images of the QLEDs
were characterized by a high-resolution (HR)-STEM (FEI Tecnai F20 FEGTEM), and the
samples were sliced using a focused ion beam (FIB) system (Dual-Beam FIB, FEI Helios
Nanolab SEM/FIB).

259

260 Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

263

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269

270 Author Contributions

271 Y.C. and S.P. fabricated devices and performed materials as well as device characterizations.

272 B.H. synthesized materials. B.L. performed contact angle measurement. The manuscript was

written by Y.C., S.P., and S.C. and revised by all authors.

274

275 Appendix A. Supporting information

276 Supplementary data associated with this article can be found in the online version.

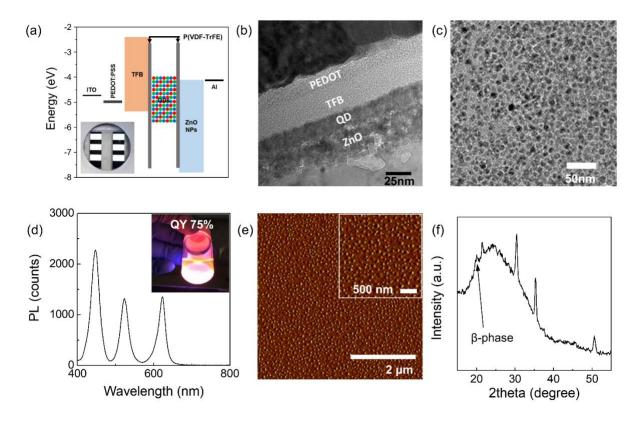
277 **References**

- 278 [1] Y. Nanishi, Nat. Photon. 2014, 8, 884.
- 279 [2] S. Pimputkar, J. S. Speck, S. P. DenBaars, S. Nakamura, *Nat. Photon.* 2009, *3*, 180.
- 280 [3] H. Jia, L. Guo, W. Wang, H. Chen, Adv. Mater. 2009, 21, 4641.
- 281 [4] E. Jang, S. Jun, H. Jang, J. Lim, B. Kim, Y. Kim, Adv. Mater. 2010, 22, 3076.
- 282 [5] K. Kim, J. Y. Woo, S. Jeong, C. -S. Han, Adv. Mater. 2011, 23, 911.
- 283 [6] H. Zhang, Q. Su, S. Chen, Nat. Commun. 2020, 11, 2826.
- [7] X. Dai, Z. Zhang, Y. Jin, Y. Niu, H. Cao, X. Liang, L. Chen, J. Wang, X. Peng, *Nature* 2014, 515, 96.
- [8] Q. Lin, L. Wang, Z. Li, H. Shen, L. Guo, Y. Kuang, H. Wang, L. S. Li, *ACS Photonics* 2018,
 5, 939.
- [9] K. Ding, H. Chen, L. Fan, B. Wang, Z. Huang, S. Zhuang, B. Hu, L. Wang, *ACS Appl. Mater*.
 Interfaces 2017, 9, 20231.
- 290 [10] X. -B. Li, C. -H. Tung, L. -Z. Wu, Nat. Rev. Chem. 2018, 2, 160.
- 291 [11] M. Yuan, M. Liu, E. H. Sargent, *Nat. Energy* **2016**, *1*, 16016.
- [12] H. Shen, Q. Lin, H. Wang, L. Qian, Y. Yang, A. Titov, J. Hyvonen, Y. Zheng, L. S. Li, ACS
- 293 Appl. Mater. Interfaces **2013**, *5*, 12011.
- [13] H. Shen, Q. Gao, Y. Zhang, Y. Lin, Q. Lin, Z. Li, L. Chen, Z. Zeng, X. Li, Y. Jia, S. Wang,
- 295 Z. Du, L. S. Li, Z. Zhang, Nat. Photon. 2019, 13, 192.
- [14] J. Song, O. Wang, H. Shen, Q. Lin, Z. Li, L. Wang, X. Zhang, L. S. Li, *Adv. Funct. Mater.*2019, 29, 1808377.
- 298 [15] M. Ban, Y. Zou, J. P. Rivett, Y. Yang, T. H. Thomas, Y. Tan, T. Song, X. Gao, D.
- 299 Credington, F. Deschler, *Nat. Commun.* **2018**, *9*, 3892.

- 300 [16] Z. Zhang, Y. Ye, C. Pu, Y. Deng, X. Dai, X. Chen, D. Chen, X. Zheng, Y. Gao, W. Fang,
- 301 *Adv. Mater.* **2018**, *30*, 1801387.
- 302 [17] H. Zhang, S. Chen, X. W. Sun, ACS Nano 2017, 12, 697.
- 303 [18] B. R. Lee, J. C. Yu, J. H. Park, S. Lee, C. Mai, B. Zhao, M. S. Wong, E. D. Jung, Y. S.
- 304 Nam, S. Y. Park, ACS Nano 2018, 12, 5826.
- [19] W. K. Bae, J. Lim, D. Lee, M. Park, H. Lee, J. Kwak, K. Char, C. Lee, S. Lee, *Adv. Mater*. **2014**, *26*, 6387.
- 307 [20] M. K. Choi, J. Yang, K. Kang, D. C. Kim, C. Choi, C. Park, S. J. Kim, S. I. Chae, T. -H.
- 308 Kim, J. H. Kim, T. Hyeon, D. -H. Kim, Nat. Commun. 2015, 6, 7149.
- 309 [21] H. Zhang, Q. Su, Y. Sun, S. Chen, Adv. Optical Mater. 2018, 6, 1800354.
- 310 [22] B. Li, M. Lu, J. Feng, J. Zhang, P. M. Smowton, J. I. Sohn, I. K. Park, H. Zhong, B. Hou,
- 311 J. Mater. Chem. C 2020, 8, 10676.
- 312 [23] S. -Y. Yoon, J. -H. Kim, K. -H. Kim, C. -Y. Han, J. -H. Jo, D. -Y. Jo, S. Hong, J. Y.
- 313 Hwang, Y. R. Do, H. Yang, *Nano Energy* **2019**, *63*, 103869.
- [24] P. Shen, F. Cao, H. Wang, B. Wei, F. Wang, X. W. Sun, X. Yang, ACS Appl. Mater. *Interfaces* 2019, 11, 1065.
- 316 [25] B. S. Mashford, M. Stevenson, Z. Popovic, C. Hamilton, Z. Zhou, C. Breen, J. Steckel, V.
- 317 Bulovic, M. Bawendi, S. Coe-Sullivan, P. T. Kazlas, *Nat. Photon.* 2013, 7, 407.
- 318 [26] H. Zhang, Q. Su, S. Chen, *Adv. Optical Mater.* **2020**, *8*, 1902092.
- 319 [27] Y. J. Choi, D. Hwang, H. Chung, D. Y. Kim, D. Kim, NPG Asia Mater. 2015, 7, e202.
- 320 [28] Y. Zhu, R. Xu, Y. Zhou, Z. Xu, Y. Liu, F. Tian, X. Zheng, F. Ma, R. Alsharafi, H. Hu, T.
- 321 Guo, T. W. Kim, F. Li, Adv. Optical Mater. 2020, 8, 2001479.
- 322 [29] Y. Yuan, T. J. Reece, P. Sharma, S. Poddar, S. Ducharme, A. Gruverman, Y. Yang, J. Huang,
- 323 Nat. Mater. 2011, 10, 296.

- 324 [30] Y. Cho, P. Giraud, B. Hou, Y. -W. Lee, J. Hong, S. Lee, S. Pak, J. Lee, J. E. Jang, S. M.
- 325 Morris, J. I. Sohn, S. Cha, J. M. Kim, *Adv. Energy Mater.* **2018**, *8*, 1700809.
- 326 [31] M. N. Almadhoun, M. A. Khan, K. Rajab, J. H. Park, J. M. Buriak, H. N. Alshareef, Adv.
- 327 *Electron. Mater.* **2019**, *5*, 1800363.
- [32] A. Arrigoni, L. Brambilla, C. Bertarelli, G. Serra, M. Tommasini, C. Castiglioni, *RSC Adv.*2020, *10*, 37779.
- 330 [33] X. Zhao, H. Xuan, C. He, *RSC Adv.* 2015, 5, 81115.
- 331 [34] Y. -Y. Choi, J. Hong, D. -S. Leem, M. Park, H. W. Song, T. -H. Sung, K. No, J. Mater.
- 332 *Chem.* **2011**, *21*, 5057.
- 333 [35] Q. Du, J. Zheng, J. Wang, Y. Yang, X. Liu, *RSC Adv.* 2018, *8*, 19585.
- [36] J. Kim, J. H. Lee, H. Ryu, J. -H. Lee, U. Khan, H. Kim, S. S. Kwak, S. -W. Kim, *Adv. Funct. Mater.* 2017, *27*, 1700702.
- [37] J. -H. Lee, R. Hinchet, T. Y. Kim, H. Ryu, W. Seung, H. -J. Yoon, S. -W. Kim, *Adv. Mater.*2015, *27*, 5553.
- [38] K. Y. Lee, S. K. Kim, J. -H. Lee, D. Seol, M. K. Gupta, Y. Kim, S. -W. Kim, *Adv. Funct. Mater.* 2016, *26*, 3067.
- 340 [39] C. Xiang, L. Wu, Z. Lu, M. Li, Y. Wen, Y. Yang, W. Liu, T. Zhang, W. Cao, S. -W. Tsang,
- 341 B. Shan, X. Yan, L. Qian, *Nat. Commun.* **2020**, *11*, 1646.
- 342 [40] A. Veamatahau, B. Jiang, T. Seifert, S. Makuta, K. Latham, M. Kanehara, T. Teranishi, Y.
- 343 Tachibana, *Phys. Chem. Chem. Phys.* **2015**, *17*, 2850.
- 344 [41] X. Li, Y. -B. Zhao, F. Fan, L. Levina, M. Liu, R. Q. -Bermudez, X. Gong, L. N. Quan, J.
- 345 Fan, Z. Yang, S. Hoogland, O. Voznyy, Z. -H. Lu, E. H. Sargent, Nat. Photon. 2018, 12, 159.33
- 346 [42] H. Moon, H. Chae, *Adv. Optical Mater.* **2020**, *8*, 1901314.
- 347 [43] W. Chen, X. Tang, P. Wangyang, Z. Yao, D. Zhou, F. Chen, S. Li, H. Lin, F. Zeng, D. Wu,

- 348 K. Sun, M. Li, Y. Huang, W. Hu, Z. Zang, J. Du, Adv. Optical Mater. 2018, 6, 1800007.
- 349 [44] D. Hahm, J. H. Chang, B. G. Jeong, P. Park, J. Kim, S. Lee, J. Choi, W. D. Kim, S. Rhee,
- 350 J. Lim, D. C. Lee, C. Lee, K. Char, W. K. Bae, Chem. Mater. 2019, 31, 3476.
- 351 [45] B. Hou, D. Parker, G. P. Kissling, J. A. Jones, D. Cherns, D. J. Fermín, J. Phys. Chem.
- 352 *C* **2013**, *117*, 6814.
- 353 [46] D. Benito-Alifonso, S. Tremel, B. Hou, H. Lockyear, J. Mantell, D. J. Fermin, P. Verkade,
- 354 M. Berry, M. C. Galan, Angew. Chem. Int. Ed. 2013, 53, 810.
- 355 [47] Y. Cho, B. Hou, J. Lim, S. Lee, S. Pak, J. Hong, P. Giraud, A. -R. Jang, Y. -W. Lee, J. Lee,
- J. E. Jang, H. J. Snaith, S. M. Morris, J. I. Sohn, S. Cha, J. M. Kim, *ACS Energy Lett.* 2018, *3*,
 1036.
- [48] B. Hou, Y. Cho, B. S. Kim, J. Hong, J. B. Park, S. J. Ahn, J. I. Sohn, S. Cha, J. M. Kim, *ACS Energy Lett.* 2016, *1*, 834.



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Figure 1. (a) Energy band diagram of the white FE-QLED without an applied bias. (b) TEM cross section image of the white FE-QLED. (c) TEM image of RGB mixed QDs. (d) PLQY of the RGB mixed QD solution where the inset shows the light emission of the mixed QDs under the UV light. (e) AFM image of the P(VDF-TrFE) islands layer on the QD film. (f) XRD patterns of FE-QLED.

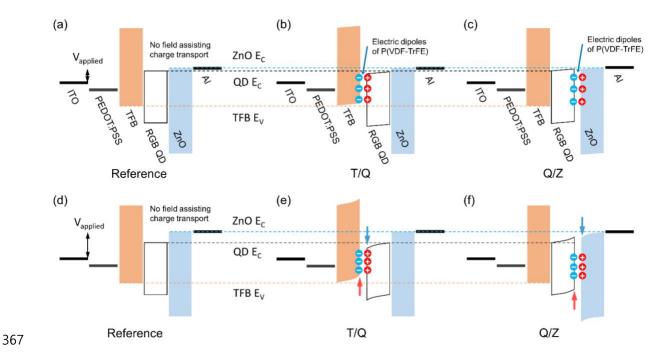


Figure 2. Energy band diagram of QLEDs for (a) a reference cell, (b) a T/Q cell, and (c) a Q/Z cell at low forward bias. (d)-(f) Energy band shift of QLEDs at high forward bias where (d), (e), and (f) corresponds to the devices shown in (a), (b), and (c), respectively. Negative and positive dipoles of P(VDF-TrFE) are depicted as blue and red circles in the figures.

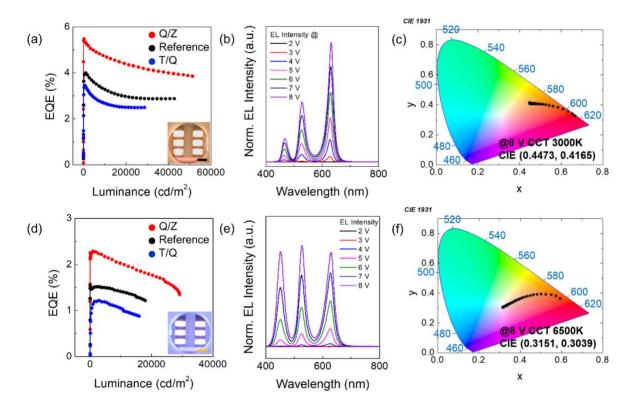


Figure 3. (a)-(c) Performance of warm white FE-QLED. (a) EQE as a function of Luminance,

(b) EL spectra of FE-QLED and (c) CIE coordinate and CCT value of FE-QLED of the red line
in Figure 3(a). (d)-(e) Performance of daylight white FE-QLED. (d) EQE as a function of

- 376 Luminance, (e) EL spectra of FE-QLED and (f) CIE coordinate and CCT value of FE-QLED
- of the red line in Figure 3(d). The scale bar in (a) and (d) is 3 mm.

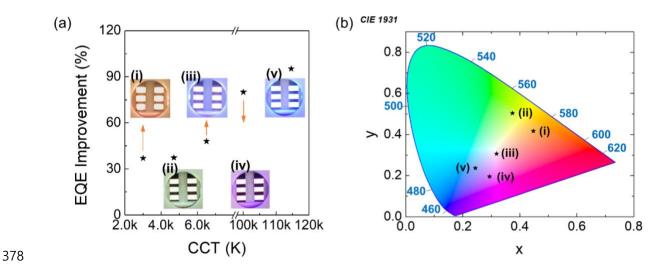


Figure 4. (a) CIE coordinates with respect to the FE-QLEDs with CCT values and corresponding digital images. (b) EQE improvement in FE-QLEDs with different CCT values using the P(VDF-TrFE) islands structure.

		Turn-on	Max. Luminance	Max. EQE	CCT	CIE coordinate	Avg. EQE#
		[V]	[cdm ⁻²]	[%]	[K]	[x, y]	[%]
	T/Q*	2.0	28680	3.454	~2000	0.5263, 0.4071	3.28 ± 0.21
Warm white	Ref	2.0	42670	3.998	~2000	0.5242, 0.4092	3.91 ± 0.26
	Q/Z**	2.1	51220	5.476	~3000	0.4473, 0.4165	5.10 ± 0.35
	T/Q*	2.6	15900	1.218	~25000	0.2404, 0.2630	1.07 ± 0.10
Daylight white	Ref	2.6	17338	1.542	~19000	0.2490, 0.2686	1.41 ± 0.15
	Q/Z**	2.7	22710	2.284	~6500	0.3151, 0.3039	2.16 ± 0.12

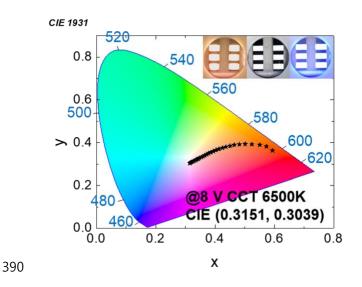
Table 1. Device characteristics of warm and daylight white QLEDs.

*T/Q – ITO/ PEDOT:PSS/ TFB/ P(VDF-TrFE)/ QD/ Al

**Q/Z - ITO/ PEDOT:PSS/ TFB/ QD/ P(VDF-TrFE)/ Al

#Average EQE from 10 QLEDs of each type of the devices

Table of Contents



Short summary

Efficient direct white light emission has been a challenge due to inevitable energy band mismatch at interfaces between quantum dots with different bandgaps. The embedded ferroelectric island structure effectively modulates the energy band at the junction interface, and this leads to improved direct white light emission efficiency with various color temperatures.

Enhanced Direct White Light Emission Efficiency in Quantum Dot 408

Light-Emitting Diodes via Embedded Ferroelectric Islands 409 **Structure** 410

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- **Keywords:** Quantum dots, White light, Light-emitting diodes, Ferroelectricity, P(VDF-TrFE) 432 433

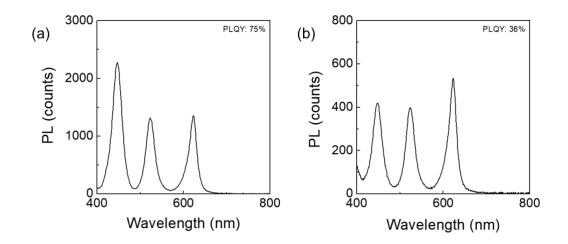


Figure S1. PLQY of (a) a QD solution and (b) a QD film.

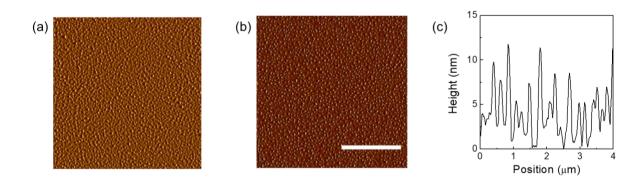


Figure S2. P(VDF-TrFE) on (a) a TFB layer and (b) a QD layer. (c) Morphology of a P(VDF-

439 TrFE) islands structure.

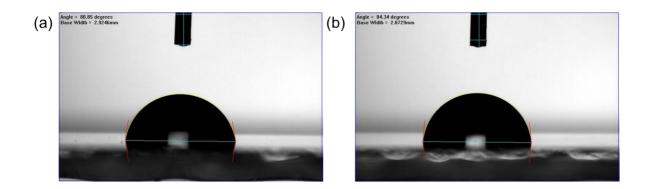


Figure S3. Contact angle on (a) TFB (80.85°) and (b) QD (84.34°).

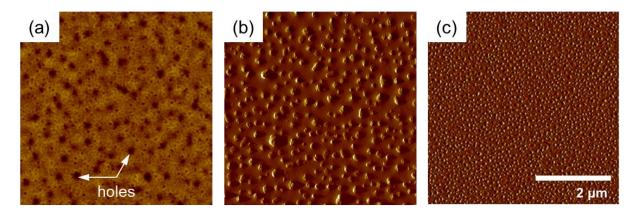


Figure S4. AFM images of a P(VDF-TrFE) layer prepared by (a) spin-coating 1 wt%, (b) spincasting 1 wt%, and (c) spin-casting 0.2 wt% P(VDF-TrFE) solution. (a)-(c) are in the same
scale with the scale bar of 2 μm.

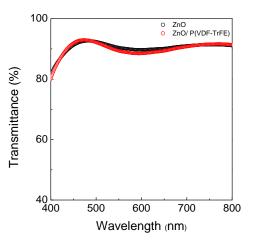


Figure S5. Transmittance (%) of the ZnO nanoparticle film and the P(VDF-TrFE) island layer

454 on the ZnO nanoparticle film.

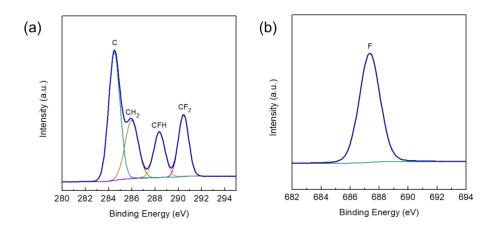


Figure S6. Core-level X-ray photoelectron spectra (XPS) of each composition in P(VDF-TrFE)

458 island layer (a) C 1s and (b) F 1s.

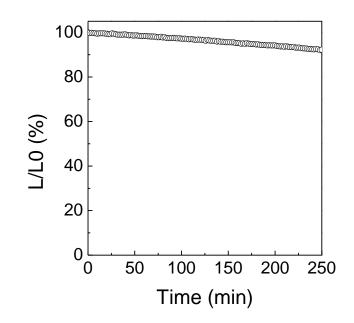
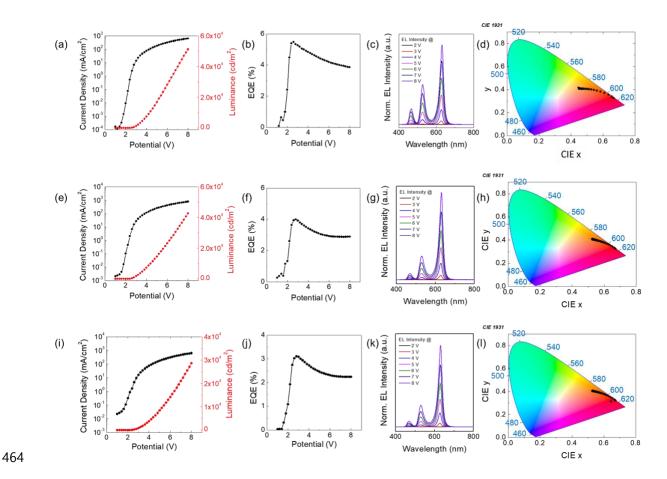
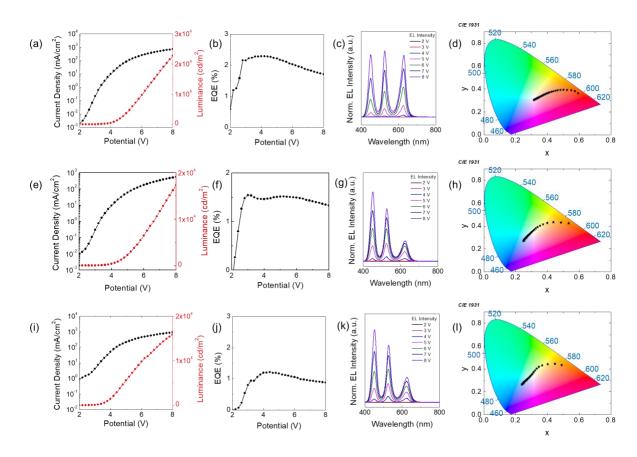




Figure S7. Long-term stability measurement of FE-QLED under continuous DC bias.



465 Figure S8. Characteristic behaviors of warm white QLEDs. (a)-(d) Q/Z FE-QLEDs, (e)-(h)
466 references, and (i)-(l) T/Q FE-QLEDs.



469 Figure S9. Characteristic behaviors of daylight white QLEDs. (a)-(d) Q/Z FE-QLEDs, (e)-(h)
470 references, (i)-(1) T/Q FE-QLEDs.

RGB Volume Ratio	CCT [K]	CIE (X , Y)
1:3.5:3.5	3000	0.4473 0.4165
1:2:4	4700	0.3743, 0.5042
1:2:9	6500	0.3183 0.3065
1:1:10	100,000	0.2938 0.1951
1:1.5:15	114,600	0.2440 0.2367

Table S1. CCT values and CIE coordinates with respect to the RGB QDs mixing ratio.

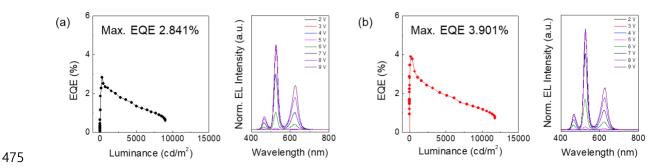


Figure S10. QLEDs with 4700 K of the CCT value (a) without P(VDF-TrFE) island layer and

477 (b) with P(VDF-TrFE) island layer between the QDs and ZnO layer.

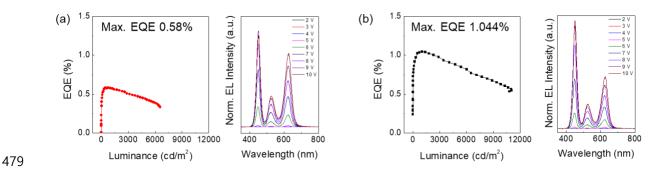


Figure S11. QLEDs with 100,000 K of the CCT value (a) without P(VDF-TrFE) island layer

and (b) with P(VDF-TrFE) island layer between the QDs and ZnO layer.

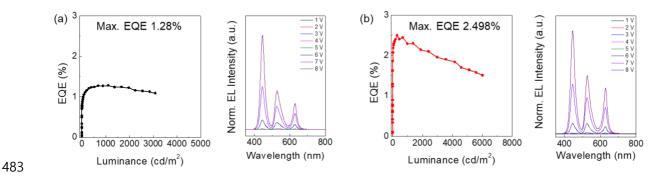
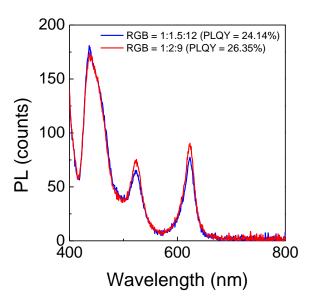


Figure S12. QLEDs with 114,600 K of the CCT value (a) without P(VDF-TrFE) island layer

and (b) with P(VDF-TrFE) island layer between the QDs and ZnO layer.

CCT [K]		Turn-on [V]	Max. Luminance [cdm ⁻²]	Avg. Luminance [cdm ⁻²]	Max. EQE [%]	Avg. EQE [%]	CIE coordinate [x, y]
3000	Ref	2.0	42670	39278 ± 2193	3.998	3.91 ± 0.26	0.5242, 0.4092
	Q/Z	2.1	51220	49023 ± 3753	5.476	5.10 ± 0.35	0.4473, 0.4165
4700	Ref	3.6	8995	8306 ± 792	2.831	2.96 ± 0.15	0.3743, 0.5042
	Q/Z	3.8	11750	10885 ± 1208	3.901	4.17 ± 0.28	0.3509, 0.5136
6500	Ref	2.6	17338	17477 ± 1426	1.542	1.41 ± 0.15	0.2490, 0.2686
	Q/Z	2.7	22710	22638 ± 1929	2.284	2.16 ± 0.12	0.3151, 0.3039
100,000	Ref	3.4	6656	6117 ± 420	0.580	0.50 ± 0.12	0.2938, 0.1951
	Q/Z	3.4	11040	10045 ± 1313	1.044	0.93 ± 0.18	0.3006, 0.2054
114,600	Ref	3.4	3094	2332 ± 360	1.280	1.13 ± 0.12	0.2387, 0.2387
	Q/Z	3.4	6016	5539 ± 423	2.498	2.26 ± 0.17	0.2440, 0.2367

Table S2. The summary of device parameters of white QLEDs with various CCT values.



491 Figure S13. PLQY measurement with the structure of Glass/ PEDOT:PSS/ TFB/ QDs/ ZnO

results of the different ratio of RGB in the mixed QD solutions: one with R:G:B = 1:2:9 and

493 the other with higher blue QD ratio, R:G:B = 1:1.5:12.

Ref	Material	Туре	V _{on} [V]	Max L. [cd/m ²]	EQE [%]	CIE	Year
[1]	CdSe/ZnS	BY mixed	5.3	6390	1.0	0.28, 0.33	2014
		RGB mixed	4.3	1440	1.3	0.39, 0.40	
		BCYR mixed	6.1	5340	0.9	0.29, 0.29	
[2]	CdSe/ZnS	RGB mixed	5	23352	10.9	0.20, 0.17	2015
[3]	CdZnSeS/ZnS	RGB mixed	3.1	60810	6.39	0.33, 0.32	2018
		Three-unit tandem	9.0	65690	23.9	0.33, 0.34	
[4]	CdSe/ZnS	RGB mixed	8.5	2953	5.0	0.28, 0.31	2018
[5]	CdSe/ZnS	RGB mixed	4.0	58361	10.6	0.38, 0.35	2020
		Light outcoupling	3.2	74363	28.4	0.33, 0.34	
This work	CdSe/ZnS	RGB mixed,	2.1	51220	5.48	0.44, 0.42	2021
		ferroelectric coupling	2.7	22710	2.23	0.31, 0.30	2021

495 **Table S3.** Comparison of Cd-based white QLEDs performances.

- 497 [1] W. K. Bae, J. Lim, D. Lee, M. Park, H. Lee, J. Kwak, K. Char, C. Lee, S. Lee, *Adv. Mater.*
- **2014**, *26*, 6387.
- 499 [2] K. -H. Lee, C. -Y. Han, H. -D. Kang, H. Ko, C. Lee, J. Lee, N. Myoung, S. -Y. Yim, H.
- 500 Yang, ACS Nano 2015, 9, 10941.
- 501 [3] H. Zhang, Q. Su, Y. Sun, S. Chen, *Adv. Optical Mater.* **2018**, *6*, 1800354.
- 502 [4] P. Shen, X. Li, F. Cao, X. Ding, X. Yang, J. Mater. Chem. C 2018, 6, 9642.
- 503 [5] Y. Zhu, R. Xu, Y. Zhou, Z. Xu, Y. Liu, F. Tian, X. Zheng, F. Ma, R. Alsharafi, H. Hu, T.
- 504 Guo, T. W. Kim, F. Li, Adv. Optical Mater. 2020, 8, 2001479.