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Article

# A Simple Approach to Achieving Ultrasmall III-Nitride Microlight-Emitting Diodes with Red Emission

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4 **ABSTRACT:** The microdisplays for augmented reality and visual reality require ultrasmall 5 microlight-emitting diodes ( $\mu$ LEDs) with a dimension of  $\leq 5 \mu$ m. Furthermore, the 6 microdisplays also need three kinds of such  $\mu$ LEDs each emitting red, green, and blue 7 emission. Currently, in addition to a great challenge for achieving ultrasmall  $\mu$ LEDs mainly 8 based on III-nitride semiconductors, another fundamental barrier is due to extreme difficulty 9 in growing III-nitride-based red LEDs. So far, there has not been any effective approach to 10 obtain high indium content InGaN as an active region required for a red LED while 11 maintaining high optical performance. In this paper, we have demonstrated a selective 12 epitaxy growth approach on a template featuring microhole arrays. This allows us to not 13 only obtain the natural formation of ultrasmall  $\mu$ LEDs but also achieve InGaN with 14 enhanced indium content at an elevated growth temperature, at which it is impossible to 15 obtain InGaN-based red LEDs on a standard planar surface. By means of this approach, we 16 have demonstrated red  $\mu$ LEDs (at an emission wavelength of 642 nm) with a dimension of 17 2  $\mu$ m, exhibiting a high luminance of  $3.5 \times 10^7$  cd/m<sup>2</sup> and a peak external quantum



18 efficiency of 1.75% measured in a wafer form (i.e., without any packaging to enhance an extraction efficiency). In contrast, an LED 19 grown under identical growth conditions but on a standard planar surface shows green emission at 538 nm. This highlights that our 20 approach provides a simple solution that can address the two major challenges mentioned above.

21 KEYWORDS: InGaN, microLED, selective epitaxy growth, patterned template, MOVPE, EQE

### 1. INTRODUCTION

22 There is a growing interest for developing microdisplays with 23 compact screens of  $\leq 1/4''$  diagonal length, which have a wide 24 range of applications in smart watches, smart phones, smart 25 bands, and augmented reality and virtual reality (AR & VR) 26 devices.<sup>1-5</sup> Their individual pixel elements typically consist of 27 a large number of microscale visible LEDs mainly based on III-28 nitride semiconductors, which are referred to as microLEDs  $_{29}$  (µLEDs). For instance, the microdisplays for AR and VR 30 require  $\mu$ LEDs with an ultrasmall dimension of <5  $\mu$ m.<sup>6-8</sup> 31 Such devices are typically utilized in a scenario where spaces 32 are small or the devices need to be close to the eyes. Therefore, the devices require high resolution, high contrast ratio, high 33 34 luminance, and high external quantum efficiency (EQE).<sup>9,10</sup> Of 35 course, a microdisplay needs three kinds of individual  $\mu$ LEDs 36 as a single pixel each emitting red, green, and blue emission 37 (i.e., RGB), respectively.

InGaN semiconductors have direct bandgaps across their whole content ranging from 0.7 eV for InN to 3.43 eV for GaN, covering part of the infrared region, the full visible part of the ultraviolet (UV) region. So far, InGaN-based  $\mu$ LEDs with reasonably good performance in the blue and green spectral region have been reported. However, Hered LEDs still rely on AlGaInP materials. Although a large area AlGaInP red LED with a high efficiency of >50% can be 45 obtained,<sup>11</sup> the efficiency reduces dramatically when its 46 dimension is reduced to the microscale, namely,  $\mu$ LEDs. 47 This is due to an enhancement in the surface recombination 48 rate and the long diffusion lengths of carriers.<sup>12–15</sup> Moreover, 49 the efficiency of AlGaInP red LEDs is sensitive to their 50 junction temperature,<sup>16,17</sup> and thus, AlGaInP red LEDs 51 generally suffer from a severe leakage current at a high 52 temperature, generating a severe efficiency thermal drop. All 53 these fundamental issues indicate that it is indispensable to 54 develop III-nitride-based red LEDs to meet the requirements 55 for the fabrication of a full color microdisplay. 56

InGaN with high indium content (>20%) is necessary for 57 obtaining long wavelength emission. Unfortunately, it is greatly 58 challenging to achieve high indium content InGaN while 59 maintaining high optical performance.<sup>18,19</sup> A typical method to 60 achieve high indium content in InGaN is to lower the growth 61

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62 temperature for InGaN. However, it is clear that this method is63 not ideal because it causes a significant degradation in crystal64 quality.

In general, vapor-solid thermodynamic equilibrium can be 65 66 modified by stress, making the solid-phase epitaxial composi-67 tion reduce toward lattice-matched conditions. This is the  $_{68}$  major reason why it is difficult to increase indium  $_{69}$  incorporation into GaN. $^{20-24}$  Therefore, the growth of 70 InGaN on a relaxed layer is beneficial for obtaining high 71 indium content in InGaN. However, bear in mind that the 72 formation of a relaxed layer is often associated with the 73 generation of extra defects if a heterostructure with a large 74 lattice mismatch is used to generate a relaxed layer. This leads 75 to degradation in optical performance. Furthermore, the stress 76 status of an underlying layer plays a critical role in determining 77 indium incorporation into GaN. Generally speaking, tensile 78 stress tends to enhance indium incorporation into GaN, 79 offering a unique advantage for growing red LEDs on silicon 80 substrates as GaN on Si suffering tensile stress.<sup>25,26</sup> In contrast, 81 GaN grown on sapphire substrates exhibits compressive strain. The growth of InGaN-based red LEDs has been reported by 82 83 means of inserting a thin AlN or an AlGaN layer into each 84 InGaN quantum well as an emitting region, leading to an 85 enhancement in strain that pushes the emission wavelength of 86 InGaN quantum wells toward longer wavelength.<sup>27,28</sup> So far, 87 this approach has become a popular method for the growth of 88 long wavelength emitters, in particular, red LEDs.<sup>25-30</sup> 89 Furthermore, by combining the idea of inserting a thin AlN or AlGaN and further adjusting the in-plane strain of a GaN 90 template by tuning the GaN thickness, 633 nm-wavelength red 91 92 LEDs with an external quantum efficiency (EQE) of 1.6% has 93 been reported, where an extremely thick GaN (8–10  $\mu$ m) <sup>95</sup> been reported, where an extremely link Garv (6–10  $\mu$ m) <sup>94</sup> template has been employed.<sup>26,29</sup> A more recent work has <sup>95</sup> demonstrated that a peak EQE as high as 4.5% has been <sup>96</sup> achieved on red InGaN  $\mu$ LEDs.<sup>31</sup> However, it is worth noting 97 that the approach of strain enhancing also leads to a reduction 98 in internal quantum efficiency.

99 We expect that an enhanced relaxation can be achieved by 100 using a selective epitaxy growth approach on a microhole-101 patterned template, which we have developed recently, where 102  $\mu$ LEDs can be naturally formed but without employing any 103 dry-etching techniques because selective epitaxy growth can 104 take place only within these microholes.<sup>7,8</sup> In this work, we are 105 proposing to employ this approach to achieve ultrasmall red 106 µLED arrays with enhanced quantum efficiency but without 107 inserting any thin AlN or AlGaN into InGaN quantum wells as 108 an emitting region. It is expected that no lateral confinement 109 during the selective epitaxy growth process leads to strain 110 relaxation effectively and naturally. By this mechanism, 642 nm 111 red  $\mu$ LEDs with a dimension of 2  $\mu$ m have been achieved by 112 our selective epitaxy growth conducted at an elevated 113 temperature, at which a red LED cannot be achieved on a 114 standard planar GaN surface. The resultant external quantum 115 efficiency is 1.75%. For comparison, only 538 nm green LEDs 116 on a standard planar GaN template can be obtained even 117 under identical growth conditions. Our X-ray diffraction 118 measurements have confirmed that a significant enhancement 119 in indium content in InGaN has been achieved by our 120 approach.

### 2. RESULTS AND DISCUSSION

121 In this work, two different InGaN-based LED samples have 122 been designed and then grown, aiming to study the influence of selective epitaxial growth on the optical performance of III- 123 nitride LEDs grown on a prepatterned template featuring 124 microhole arrays. A  $\mu$ LED array sample is obtained by our 125 selective epitaxy growth on the prepatterned n-GaN template 126 as mentioned above and is denoted as LED A. The other one is 127 a normal LED sample grown under identical growth but on a 128 standard planar n-GaN template without any features and is 129 denoted as LED B. 130

Silicon-doped n-GaN epiwafers are first grown on *c*-plane 131 (0001) sapphire substrates using the standard two-step 132 approach by a metalorganic vapor phase epitaxy (MOVPE) 133 technique. Initially, a 25 nm GaN nucleation layer is prepared 134 at a low temperature after the substrate is subject to a 135 thermally annealing process at a high temperature of 1150 °C, 136 followed by a 1  $\mu$ m GaN buffer layer, and then another 500 nm 137 silicon-doped n-GaN layer both grown at a high temperature of 138 1120 °C. For LED A, the n-GaN template is further patterned 139 into microhole arrays using SiO<sub>2</sub> masks on its top, which is 140 then used as a prepatterned template for our selective epitaxial 141 growth.

Figure 1a shows the schematics of our selective epitaxy 143 fl growth approach, allowing us to naturally achieve  $\mu$ LED arrays 144



**Figure 1.** (a) Schematic of selective epitaxy growth and (b) plan-view SEM image for the  $\mu$ LED array epiwafer, showing a diameter of 2  $\mu$ m and an interpitch of 1.5  $\mu$ m.

without involving any dry-etching process, i.e., LED A. For the 145 detailed information on fabricating the prepatterned templates, 146 refer to the Experimental Methods section. 147

Afterward, a standard III-nitride LED structure is selectively 148 grown on the micropatterned template by MOVPE, namely, a 149 silicon-doped n-GaN layer is first prepared, followed by an 150  $In_{0.05}Ga_{0.95}N/GaN$  superlattice (SLS) structure as a prelayer, 151 five periods of InGaN/GaN multiple quantum wells (MQWs) 152 as an emitting region, then a 20 nm *p*-type  $Al_{0.2}Ga_{0.8}N$  as an 153 electron blocking layer, and a final 150 nm *p*-type GaN layer. 154 The total thickness of the overgrown layers is 500 nm, which 155 matches the thickness of the SiO<sub>2</sub> masks. Due to the SiO<sub>2</sub> 156 masks, the growth of the LED structure takes place within the 157 microholes only, naturally forming regularly arrayed  $\mu$ LEDs. 158

A Raith 150 scanning electron microscopy (SEM) system 159 has been used to characterize the surface morphology of our 160 regularly arrayed  $\mu$ LEDs. Figure 1b shows a typical plan-view 161 162 SEM image of our regularly arrayed  $\mu$ LEDs wafer (i.e., LED 163 A), exhibiting a nice circular shape with an excellent high 164 uniformity in shape, diameter, and interpitch. All  $\mu$ LEDs are 2 165  $\mu$ m in diameter and only 1.5  $\mu$ m in interpitch. Such a small 166 diameter and an interpitch are crucial for manufacturing a 167 high-resolution microdisplay in a compact manner. Further-168 more, the  $\mu$ LED pixels share a common *n* contact while all the 169 *p* contacts are left open. As a result, our regularly arrayed 170  $\mu$ LED epiwafers well match any existing manufacturing 171 technique of microdisplays, for instance, the pick-and-place 172 technology, which has been widely used,<sup>32</sup> and the integrating 173 technique using driving transistors based on the silicon CMOS 174 IC to achieve individually addressable  $\mu$ LED-based microdis-175 plays.<sup>33</sup>

176 A high-resolution X-ray diffractometer (HRXRD) (Bruker 177 D8) has been employed to determine the indium content of 178 the InGaN MQWs by performing  $\omega$ -2 $\theta$  scan measurements 179 along the (002) direction, together with a fitting using the 180 Bruker JV-RADS simulation software. Figure 2a,b shows the



**Figure 2.** HRXRD  $\omega$ -2 $\theta$  scan curves of the  $\mu$ LED array sample on a patterned template, i.e., (a) LED A, and the LED sample grown on a standard planar template, i.e., (b) LED B under identical conditions. Fittings have also been provided to determine the indium content in InGaN MQWs.

181 HRXRD  $\omega$ -2 $\theta$  scan curves of our regularly arrayed  $\mu$ LED wafer 182 (i.e., LED A) and the standard LED wafer (i.e., LED B), 183 respectively. In both cases, the satellite peaks with up to 4 or 5 184 orders from the InGaN/GaN MQWs have been clearly 185 observed. The satellite peaks from the SLS structure as a 186 prelayer have also been observed. Based on a detailed fitting, it 187 can be determined that the indium content of the InGaN 188 MQWs of LED A is 31% and that the thicknesses of the 189 InGaN quantum well and the barrier are 2.2 and 13.8 nm, 190 respectively. In contrast, LED B exhibits 24% indium content 191 in the InGaN MQWs with a 2.6 nm quantum well and a 14.1 192 nm barrier. The XRD fittings are conducted based on fully strained InGaN MQWs for both LEDs. It is well known that 193 strain relaxation will reduce the strain-induced quantum-194 195 confined Stark effect (QCSE), leading to a blue-shift in the 196 emission. It means that if the InGaN MQWs are assumed to be 197 strain-relaxed, the indium content should be even higher. In 198 consideration of a higher chance of strain relaxation for the 199  $\mu$ LEDs, the fitted values of indium contents represent the least 200 difference between the two LEDs. This direct comparison 201 indicates that enhanced indium content in InGaN MQWs can

be obtained by using our selective epitaxy growth approach on 202 a prepatterned template featuring microhole arrays. 203

Finally, both the regularly arrayed  $\mu$ LED wafer (i.e., LED A) 204 and the standard LED wafer (i.e., LED B) have been fabricated 205 into LED devices with an area of 330  $\times$  330  $\mu$ m<sup>2</sup>. For the 206 detailed information about device fabrication, refer to the 207 Experimental Methods section. For LED A, each LED device 208 consists of a few thousands of 2  $\mu$ m  $\mu$ LEDs connected. In this 209 work, the  $\mu$ LEDs in LED A share a common p contact and n 210 contact, which are driven simultaneously in all electro- 211 luminescence (EL) measurements. However, it is worth noting 212 that our arrayed  $\mu$ LEDs are designed to make the *p* contacts of 213 each  $\mu$ LED left open, providing an opportunity in the future to 214 allow indium bumps to be bonded to an active matrix driving 215 transistors. This means that our regularly arrayed  $\mu$ LED 216 structure entirely matches any existing individually addressable 217 µLED microdisplays. 218

For a direct comparison, the LED B wafer has also been 219 processed under identical conditions in the same batch. All the 220 characteristics of our  $\mu$ LED chips in the present study have 221 been carried out on bare chips, meaning that we did not use 222 coating or passivation or epoxy or reflector for improving 223 extraction efficiency. Current–voltage (*I*–*V*) characteristic and 224 EL measurements have been performed at room temperature 225 in continuous wave (CW) mode using a Keithley 2400 226 sourcemeter on a probe station equipped with an optical 227 microscopy system.

The EL spectra have been measured on the two LED devices  $^{229}$  under identical conditions aiming to make a direct comparison.  $^{230}$  For instance, Figure 3a,b shows the EL spectra of the two LED  $^{231}$  f3 devices measured at a current density of 10 A/cm<sup>2</sup>,  $^{232}$  respectively. Both spectra exhibit a single emission peak. The  $^{233}$ 



**Figure 3.** EL spectra measured at 10 A/cm<sup>2</sup> for the  $\mu$ LED array device, i.e., (a) LED A and (b) LED B, where the insets show their respective emission images. EL spectra measured at increased current densities from 10 to 80 A/cm<sup>2</sup> for (c) LED A and (d) LED B, respectively.



**Figure 4.** Emission images of the  $\mu$ LED array device taken using an optical microscopy system as a function of injection current density (4, 8, and 12 A/cm<sup>2</sup>) under (a-c) a low magnification and (d-f) a high magnification, respectively.

234 µLED array device shows a strong emission at an emission wavelength of 642 nm in the red spectral region. The inset of 235 236 Figure 3a exhibits an emission image of the  $\mu$ LED array chip, 237 demonstrating red light. In contrast, Figure 3b displays a strong green emission at 538 nm from the LED B device also 238 measured at 10  $A/cm^2$ , and the inset displays its emission 239 image. This means that the selective epitaxial growth on a 240 prepatterned template featuring regularly arrayed microholes 241 242 results in a red-shift of about 100 nm in emission wavelength in comparison with the LED grown on a standard planar GaN 243 surface, although both are grown under identical growth 244 conditions. As discussed earlier, the growth of InGaN on a 245 246 relaxed layer is beneficial for obtaining high indium content in 247 InGaN. Due to no lateral confinement during the overgrowth within the microholes, the overgrown n-GaN is very likely 248 249 strain-relaxed, which leads to an enhancement in the indium 250 content in the overlying InGaN MQWs. Combined with the 251 XRD results, it has been confirmed that our selective epitaxy 252 growth approach can enhance indium incorporation into GaN 253 significantly. Figure 3c,d shows the EL spectra of LED A and 254 LED B, both measured as a function of injection current 255 density ranging from 10 to 80  $A/cm^2$ , respectively.

In order to demonstrate emitting  $\mu$ LED pixels, optical 256 257 microscopy images have been taken using a micro-EL measurement system where emissions are collected through 258  $_{259}$  two objective lenses (one  $10 \times$  magnification lens with NA = 260 0.28 and another  $50 \times$  magnification lens with NA = 0.43). 261 Figure 4a-c displays the emission images of our  $\mu$ LED array 262 chip taken under 4, 8, and 12 A/cm<sup>2</sup> current density, respectively, while Figure 4d-f provides their corresponding 263 emission images taken under a high magnification, showing 264 strong red emissions from individual 2  $\mu$ m  $\mu$ LED pixels even 265 266 under low current densities. It is worth mentioning that such 267 low current densities used for the operation of our  $\mu$ LEDs are 268 lower than a typical current density  $(22 \text{ A/cm}^2)$  for the operation of a conventional broad area LED. 269

f4

f5

<sup>270</sup> Both light output powers and luminous flux have been <sup>271</sup> measured on the bare-chip LEDs bonded on TO5-headers in <sup>272</sup> CW mode using a LCS-100 integrating sphere equipped with a <sup>273</sup> CCD APRAR spectrometer. Figure 5a-c shows the output <sup>274</sup> power, luminance, and EQE of the  $\mu$ LED array device (i.e.,



**Figure 5.** (a) Output power, (b) luminance, (c) EQE, and (d) current–voltage characteristics of the  $\mu$ LED array device (i.e., LED A).

LED A) as a function of injection current density. This <sup>275</sup> demonstrates that the output power and luminescence increase <sup>276</sup> monolithically with increasing current density up to 450 A/cm<sup>2</sup> <sup>277</sup> and that a high luminance of  $3.5 \times 10^7$  cd/m<sup>2</sup> has been <sup>278</sup> achieved. The peak EQE is about 1.75%. It is worth <sup>279</sup> highlighting that although there are not any heat-sink <sup>280</sup> components used, our ultrasmall  $\mu$ LEDs can still sustain a <sup>281</sup> high current density of above 450 A/cm<sup>2</sup>, also confirming the <sup>282</sup> high crystal quality of our  $\mu$ LED array sample achieved by our <sup>283</sup> selective epitaxy growth approach. Figure 5d displays the <sup>284</sup> typical *I–V* characteristics of LED A measured as a function of <sup>285</sup> bias, which is similar to that of the LED B device. This also <sup>286</sup> shows the good electrical property of our  $\mu$ LED array device. <sup>287</sup>

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### 3. CONCLUSIONS

288 In summary, we are proposing to employ a selective epitaxy 289 growth approach on a microhole patterned template to 290 significantly enhance strain relaxation, allowing us to not 291 only obtain the natural formation of regularly arrayed  $\mu$ LEDs 292 but also achieve enhanced indium content in the InGaN/GaN 293 MQWs used as an active region for the  $\mu$ LEDs. By means of 294 this approach, we have demonstrated red InGaN-based  $\mu$ LED <sup>295</sup> arrays with a dimension of 2  $\mu$ m and an interpitch of 1.5  $\mu$ m. A 296 high luminance of 3.5  $\times$   $10^7$  cd/m² and a peak EQE of 1.7% 297 have been achieved for the red  $\mu$ LED array chip in a wafer 298 form without any packaging. In contrast, the standard LED 299 grown under growth conditions but on a standard planar GaN 300 template demonstrates green emission. This means that our 301 approach paves the way for achieving long wavelength InGaN-302 based  $\mu$ LEDs with ultrasmall dimensions at an elevated growth 303 temperature, at which it is impossible to obtain InGaN-based 304 red LEDs on a standard planar template.

### 4. EXPERIMENTAL METHODS

4.1. Fabrication of Prepatterned Templates. A 500 nm SiO<sub>2</sub> 305 306 dielectric film is deposited on the n-GaN template by a plasma-307 enhanced chemical vapor deposition (PECVD) technique, followed 308 by employing a standard photolithography and then a dry etching 309 technique to selectively etch the SiO<sub>2</sub> dielectric layer down to the n-310 GaN surface by inductively coupled plasma (ICP), forming regularly 311 arrayed microholes with a diameter of 2  $\mu$ m and an interpitch of 1.5 312  $\mu$ m. This prepatterned template will then be used for further selective 313 epitaxy growth. Finally,  $\mu$ LEDs will be selectively grown only within 314 SiO<sub>2</sub> microhole regions, naturally forming regularly arrayed  $\mu$ LEDs.

4.2. Device Fabrication. Indium-tin-oxide (ITO) is deposited 315 316 and then undergoes an annealing process in air at 600 °C for 1 min, 317 forming transparent p-type contact, while Ti/Al/Ni/Au alloys are 318 prepared as *n*-type contact. Ti/Au alloys are used as *p*-type and *n*-type 319 electrodes. All the characteristics of the LEDs in this paper are 320 conducted on bare chips, namely, no coating, no passivation, no 321 epoxy, or no reflector, which are often employed for obtaining 322 enhanced extraction efficiency.

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# **Author Contributions**

<sup>+</sup>P.F., C.X., and J.B. contributed equally to this work. 349 350

# **Author Contributions**

T.W. conceived the idea and organized the project. T.W. and 351 J.B. prepared the manuscript. P.F., X.C., and C.Z. grew all the 352 samples. P.F., X.C., and I.F. performed the material character- 353 ization. J.B. and G.M.D.A. fabricated the prepatterned 354 templates and carried out the device fabrication. J.B. conducted 355 the device characterization. 356

#### Notes 357

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### REFERENCES

(1) Meng, W.; Xu, F.; Yu, Z.; Tao, T.; Shao, L.; Liu, L.; Li, T.; Wen, 364 K.; Wang, J.; He, L.; Sun, L.; Li, W.; Ning, H.; Dai, N.; Qin, F.; Tu, 365 X.; Pan, D.; He, S.; Li, D.; Zheng, Y.; Lu, Y.; Liu, B.; Zhang, R.; Shi, 366 Y.; Wang, X. Three-dimensional Monolithic Micro-LED Display 367 Driven by Atomically Thin Transistor matrix. Nat. Nanotechnol. 2021, 368 16, 1231. 369

(2) Park, J.; Choi, J. H.; Kong, K.; Han, J. H.; Park, J. H.; Kim, N.; 370 Lee, E.; Kim, D.; Ki, J.; Chung, D.; Jun, S.; Kim, M.; Yoon, E.; Shin, J.; 371 Hwang, S. Electrically Driven Mid-Submicrometre Pixelation of 372 InGaN Micro-Light-Emitting Diode Displays for Augmented-Reality 373 Glasses. Nat. Photonics 2021, 15, 449. 374

(3) Huang, Y.; Hsiang, E.-L.; Deng, M.-Y.; Wu, S.-T. Mini-LED, 375 Micro-LED and OLED Displays: Present Status and Future 376 Perspectives. Light: Sci. Appl. 2020, 9, 105. 377

(4) Han, H.-V.; Lin, H.-Y.; Lin, C.-C.; Chong, W.-C.; Li, J.-R.; Chen, 378 K.-J.; Chen, T.-M.; Chen, H.-M.; Lau, K.-M.; Kuo, H.-C. Resonant- 379 enhanced full-Color Emission of Quantum-Dot-Based Micro LED 380 Display Technology. Opt. Express 2015, 23, 32504-32515. 381

(5) Green, R. P.; McKendry, J. J. D.; Massoubre, D.; Gu, E.; 382 Dawson, M. D.; Kelly, A. E. Modulation Bandwidth Studies of 383 recombination Processes in Blue and Green InGaN Quantum Well 384 Micro-Light-Emitting Diodes. Appl. Phys. Lett. 2013, 102, 385 No. 091103. 386

(6) Ley, R. T.; Smith, J. M.; Wong, M. S.; Margalith, T.; Nakamura, 387 S.; DenBaars, S. P.; Gordon, M. J. Revealing the Importance of Light 388 Extraction Efficiency in InGaN/GaN MicroLEDs via Chemical 389 Treatment and Dielectric Passivation. Appl. Phys. Lett. 2020, 116, 390 251104. 391

(7) Bai, J.; Cai, Y.; Feng, P.; Fletcher, P.; Zhao, X.; Zhu, C.; Wang, 392 T. A Direct Epitaxial Approach To Achieving Ultrasmall and 393 Ultrabright InGaN Micro Light-Emitting Diodes (µLEDs). ACS 394 Photonics 2020, 7, 411. 395

(8) Bai, J.; Cai, Y.; Feng, P.; Fletcher, P.; Zhu, C.; Tian, Y.; Wang, T. 396 Ultrasmall, Ultracompact and Ultrahigh Efficient InGaN Micro Light 397 Emitting Diodes ( $\mu$ LEDs) with Narrow Spectral Line Width. ACS 398 Nano 2020, 14, 6906. 399

(9) Zhan, T.; Yin, K.; Xiong, J.; He, Z.; Wu, S.-T. Augmented Reality 400 and Virtual Reality Displays: Perspectives and Challenges. iScience 401 2020, 23, No. 101397. 402

(10) Liu, Z.; Lin, C.-H.; Hyun, B.-R.; Sher, C.-W.; Luo, B.; Lv, Z.; 403 Jiang, F.; Wu, T.; Ho, C.-H.; Kuo, H.-C.; He, J.-H. Micro-light- 404 emitting diodes with quantum dots in display technology. Light: Sci. 405 Appl. 2020, 9, 83. 406

(11) Krames, M. R.; Ochiai-Holcomb, M.; Höfler, G. E.; Carter- 407 Coman, C.; Chen, E. I.; Tan, I. H.; Grillot, P.; Gardner, N. F.; Chui, 408 H. C.; Huang, J. W.; Stockman, S. A.; Kish, F. A.; Craford, M. G.; 409 410 Tan, T. S.; Kocot, C. P.; Hueschen, M.; Posselt, J.; Loh, B.; Sasser, G.; 411 Collins, D. High-Power Truncated-Inverted-Pyramid 412 ( $Al_xGa_{1-x}$ )<sub>0.5</sub>P/GaP Light-Emitting Diodes Exhibiting >50% 413 External quantum efficiency. *Appl. Phys. Lett.* **1999**, 75, 2365–2367. 414 (12) Boroditsky, M.; Gontijo, I.; Jackson, M.; Vrijen, R.; 415 Yablonovitch, E.; Krauss, T.; Cheng, C. C.; Scherer, A.; Bhat, R.; 416 Krames, M. Surface Recombination Measurements on III–V 417 Candidate Materials for Nanostructure Light-Emitting Diodes. *J.* 418 *Appl. Phys.* **2000**, 87, 3497.

(13) Royo, P.; Stanley, R. P.; Ilegems, M.; Streubel, K.; Gulden, K.
H. Experimental Determination of the Internal Quantum Efficiency of
AlGaInP Microcavity Light-Emitting Diodes. J. Appl. Phys. 2002, 91,
2563.

423 (14) Oh, J.-T.; Lee, S.-Y.; Moon, Y.-T.; Moon, J. H.; Park, S.; Hong, 424 K. Y.; Song, K. Y.; Oh, C.; Shim, J.-I.; Jeong, H.-H.; Song, J.-O.; 425 Amano, H.; Seong, T.-Y. Light Output Performance of Red AlGaInP-426 Based Light Emitting Diodes with Different Chip Geometries and 427 Structures. *Opt. Express* **2018**, *26*, 11194.

428 (15) Wong, M. S.; Kearns, J. A.; Lee, C.; Smith, J. M.; Lynsky, C.; 429 Lheureux, G.; Choi, H.; Kim, J.; Kim, C.; Nakamura, S.; Speck, J. S.; 430 Denbaars, S. P. Improved Performance of AlGaInP Red Micro-Light-431 Emitting Diodes with Sidewall Treatments. *Opt. Express* **2020**, *28*, 432 5787–5793.

433 (16) Yadav, A.; Titkov, I. E.; Sokolovskii, G. S.; Karpov, S. Y.; 434 Dudelev, V. V.; Soboleva, K. K.; Strassburg, M.; Pietzonka, I.; 435 Lugauer, H.-J.; Rafailo, E. U. Temperature Effects on Optical 436 Properties and Efficiency of Red AlGaInP-Based Light Emitting 437 Diodes under High Current Pulse Pumping. *J. Appl. Phys.* **2018**, *124*, 438 No. 013103.

(17) Oh, C.-H.; Shim, J.-I.; Shin, D.-S. Current- and TemperatureDependent Efficiency Droops in InGaN-Based Blue and AlGaInPBased Red Light-Emitting Diodes. Japanese. J. Appl. Phys. 2019, 58,
SCCC08.

443 (18) El-Masry, N. A.; Piner, E. L.; Liu, S. X.; Bedair, S. M. Phase 444 Separation in InGaN Grown by Metalorganic Chemical Vapor 445 Deposition. *Appl. Phys. Lett.* **1998**, *72*, 40.

446 (19) Wakahara, A.; Tokuda, T.; Dang, X.-Z.; Noda, S.; Sasaki, A. 447 Compositional Inhomogeneity and Immiscibility of a GaInN Ternary 448 Alloy. *Appl. Phys. Lett.* **1997**, *71*, 906–908.

449 (20) Inatomi, Y.; Kanagawa, Y.; Ito, T.; Suski, T. Theoretical Study 450 of the Composition Pulling Effect in InGaN Metalorganic Vapor-451 Phase Epitaxy Growth. *Jpn. J. Appl. Phys.* **2017**, *56*, No. 078003.

452 (21) Wang, T. Topical Review: Development of Overgrown 453 Semipolar GaN for High Efficiency Green/Yellow Emission. *Semi*-454 cond. Sci. Technol. **2016**, 31, No. 093003.

455 (22) Pereira, S.; Correia, M. R.; Pereira, E.; O'Donnell, K. P.; Alves, 456 E.; Sequeira, A. D.; Franco, N.; Watson, I. M.; Deatcher, C. J. Strain 457 and Composition Distributions in Wurtzite InGaN/GaN Layers 458 Extracted from X-ray Reciprocal Space Mapping. *Appl. Phys. Lett.* 459 **2002**, *80*, 3913.

460 (23) Shimizu, M.; Kawaguchi, Y.; Hiramatsu, K.; Sawaki, N. 461 Metalorganic Vapor Phase Epitaxy of Thick InGaN on Sapphire 462 Substrate. *Ipn. J. Appl. Phys., Part 1* **1997**, *36*, 3381.

463 (24) Sonderegger, S.; Feltin, E.; Merano, M.; Crottini, A.; Carlin, J. 464 F.; Sachot, R.; Deveaud, B.; Grandjean, N.; Ganière, J. D. High Spatial 465 Resolution Picosecond Cathodoluminescence of InGaN Quantum

466 Wells. Appl. Phys. Lett. 2006, 89, 232109.

467 (25) Tawfik, W. Z.; Hyun, G. Y.; Ryu, S.-W.; Ha, J. S.; Lee, J. K.
468 Piezoelectric Field in Highly Stressed GaN-Based LED on Si (1 1 1)
469 Substrate. *Opt. Mater.* 2016, 55, 17–21.

470 (26) Iida, D.; Zhuang, Z.; Kirilenko, P.; Velazquez-Rizo, M.; Najmi, 471 M. A.; Ohkawaa, K. High-color-Rendering-Index Phosphor-Free 472 InGaN-Based White Light-Emitting Diodes by Carrier Injection 473 Enhancement via V-Pits. *Appl. Phys. Lett.* **2020**, *116*, 162101.

474 (27) Hwang, J.-I.; Hashimoto, R.; Saito, S.; Nunoue, S. Development 475 of InGaN-Based Red LED Grown on (0001) Polar Surface. *Appl.* 476 *Phys. Express* **2014**, 7, No. 071003. (28) Hashimoto, R.; Hwang, J.; Saito, S.; Nunoue, S. High-Efficiency 477 Yellow Light-Emitting Diodes Grown on Sapphire (0001) Substrates. 478 Phys. Status Solidi C 2014, 11, 628. 479

(29) Zhuang, Z.; Iida, D.; Ohkawa, K. InGaN-based red light- 480 emitting diodes: from traditional to micro-LEDs. *Jpn. J. Appl. Phys.* 481 **2022**, *61*, SA0809. 482

(30) Li, P.; Li, H.; Zhang, H.; Lynsky, C.; Iza, M.; Speck, J. S.; 483 Nakamura, S.; DenBaars, S. P. Size-Independent Peak External 484 Quantum Efficiency (>2%) of InGaN Red Microlight-Emitting 485 Diodes with An Emission Wavelength over 600 nm. *Appl. Phys.* 486 *Lett.* **2021**, *119*, No. 081102. 487

(31) Li, P.; Li, H.; Zhang, H.; Yang, Y.; Wong, M. S.; Lynsky, C.; Iza, 488 M.; Gordon, M. J.; Speck, J. S.; Nakamura, S.; DenBaars, S. P. Red 489 InGaN micro-light-emitting diodes (>620 nm) with a peak external 490 quantum efficiency of 4.5% using an epitaxial tunnel junction contact. 491 *Appl. Phys. Lett.* **2022**, *120*, 121102. 492

(32) Wong, M. S.; Hwang, D.; Alhassan, A. I.; Lee, C.; Ley, R.; 493 Nakamura, S.; DenBarrs, S. P. High efficiency of III-nitride micro- 494 light emitting diodes by sidewall passivation using atomic layer 495 deposition. *Opt. Express* **2018**, *26*, 21324–21331. 496

(33) Day, J.; Li, J.; Lie, D. Y. C.; Bradford, C.; Lin, J. Y.; Jiang, H. X. 497 III-Nitride full-scale high-resolution microdisplays. *Appl. Phys. Lett.* 498 **2011**, *99*, No. 031116.