# Peer Review File

# Development of Fin-LEDs for Next-generation Inorganic Displays using Face-selective Dielectrophoretic Assembly

Corresponding Author: Professor Young Rag Do

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

This is a careful study presenting two developments: 1) incorporating MQWs on the large facet of a several-micron long cuboid GaN structure (finLED). Authors show an 8 times larger active region relative to nanorodLEDs, leading to brighter fin LEDs. 2) Alignment of finLEDs which includes fin axial direction and rotation and placement of the desired facet on the underlying electrodes using dielectrophoretic (DEP) method. Authors also state "... our study attained a 91.3% vertical assembly percentage". This represents the fraction of the fins that face down correctly. The facet-controlled placement is introduced to enable a brighter pixel with 8640 cd/m2, while EQE stands at 9.1%. In a sense a better light extraction is obtained relative to other types of nanorod LEDs.

Authors, as far as I understand, do not disclose a distinct advantage in their DEP assembly method (which is the main point of this work) relative to what has been previously reported, for instance, in ref. 26. In other words, their contribution seems gradual and not in a stark difference relative to the prior methods. They don't show if their torque-induced rotation is different from the earlier reports or what are the new aspects! It should be noted that the manipulation and placement of micro-finLEDs, several microns long, seems to be a new aspect (relative to ref 26), however, as discussed in my comments below, there are some questions and ambiguities around that. As such this work is not suitable for publication in Nature Comm. Below are my questions and comments:

Line 223: "We define pixels with 15 or fewer fin-LEDs aligned in a 42x42 µm2 area as bad cells, and out of a total of 2940 pixels, only two defective subpixels were found." It appears that their method has no deterministic control over the number of finLEDs per pixel and process seems stochastic; is that true? Based on what criteria, value 15 is chosen as the threshold of normal or bad pixel? The 99.93% assembly accuracy is based on how many experimental runs? Also based on Figure 5f, there seems to be a noticeable variation in the emission intensity of different pixels with the large array. How is this variation contrasted with the # of finLEDs per pixel? Does the poor emission indicate poor contact with the metal electrodes?

Line 237 In sentence:"..., namely the triple-shell, SiO2-shell, and TiO2-shell, in terms of brightness", the "triple-shell" seems redundant.

Line 247-249: Figure 5e precedes Figure 5d.

Line 254: "Figures 5f-g illustrate the array cell containing 588 pixels...". Figure 5g in comparison with 5f, does not look like the right one as it shows a small pixel.

Line 43. "Upon transferring nanorod-LED display technology by our team, the Samsung Display Company (SDC) reported the successful fabrication of a nanorod-LED-based electroluminescent (EL) device through the FSA-DEP process, highlighting the various advantages of the DEP process". This sounds a causal sentence implying the SDC work was based on the work of this team. Rewording recommended. Perhaps replace "Upon" with "following".

Page 2. "These nanorod-LEDs autonomously align on subpixel electrodes using fluidic assembly". If they align autonomously, then what is the purpose of the fluidic assembly? Word "autonomously" should be reworded or omitted.

Line 96: What are the superior optical characteristics that author refer to on line 96. Do they mean a stronger light extraction efficiency? Please clarify.

Line 50: What is the goal of discussing nanorod LEDs in this work? It is understandable that finLEDs with a larger active region are better than nanorod LEDs. Perhaps authors should consider eliminating this section as it seems a distraction from the main point.

Figure 1c. Why the emissions related to the EBL and MQW layers are not apparent in the nanorod LEDs. In CL, at least, due to its high sensitivity, it should have some signature?

Page 4: line 110: how is the 20-fold CL enhancement calculated? Is it based on analysis of individual structures or their groups?

Lastly, fin LED design is not a new concept and several teams have reported them with aspect ratios that look like a fin (in contrast to the small aspect ratio cuboid objects of this study). It is recommended that authors site their work for a fair and accurate description of the field, for instance:

1.Selective-Area Growth of III-Nitride Core-Shell Nanowalls for Light-Emitting and Laser Diodes. In 2014 Conference on Lasers and Electro-Optics, IEEE: 2014; pp 1-2.

2. Molecular beam epitaxial growth and characterization of AIN nanowall deep UV light emitting diodes. Appl. Phys. Lett. 2017, 111.

3. High-brightness lasing at submicrometer enabled by droop-free fin light-emitting diodes (LEDs). Sci. Adv. 2020, 6, eaba4346.

#### Reviewer #2

## (Remarks to the Author)

In this manuscript, fin-LEDs that overcome the weaknesses previously reported in nanorod-LEDs have been demonstrated. These fin-LEDs show marked improvements in transfer efficiency and pixel assembly yield compared to former nanorod-LEDs. The increased volume of the MQW (more than 8 times) and the thickening of the EBL (electron-blocking layer) contribute to the observation of strong blue light (as shown in PL, CL, and EL graphs). Additionally, a triple-shell fin-LED EL device was developed, which demonstrates superior performance in terms of passivation, brightness, and emission uniformity.

By enhancing the performance of fin-LEDs, significant contributions to the advancement of next-generation inorganic displays are anticipated. Please refer to the following comments.

1. The size distribution of fin-LEDs should be provided. The authors used millions of chips for these experiments. However, it seems that the size of the fin-LEDs is not uniform, as shown in Figures 1b and 2f. The size distribution of fin-LEDs might change the EL distribution. All these effects should be addressed.

2.In addition, each fin-LED contains a large number of defects at the edges, as shown in Figure 2f. It is questionable whether the large number of defects in each fin-LED might affect the uniformity of brightness and, consequently, the uniformity of the pixel. This effect should also be addressed.

3.In the electric field simulation part, FDEP and TDEP are calculated as functions of  $\theta x$ , as shown in Figures 3b and 3d. However, further explanation is required for Figures 3b and 3d in conjunction with Figures 4a to 4h. It is not clear how such a high yield is achieved from the DEP force or DEP torque differences.

4. The bad pixels and good pixels were defined. If a bad pixel is defined as a pixel with 15 or fewer fin-LEDs, it is anticipated that using a large number of chips for alignment will lead to fewer bad pixels. Is it possible to quantitatively explain the relationship between the number of bad pixels and the number of input chips?

5. The production yield of 99.93% is unclear. Please explain the calculation method of the production yield.

6.In Figure 5d, the threshold voltage of the triple-shell LED shifts positively with a steep increase in current density as the driving voltage increases. However, the mechanism of the increasing EQE is unclear in the manuscript. This should be further addressed.

7.In the conclusion, it is mentioned that the EQE is slightly lower than in prior results. However, the method for improving the EQE was not properly addressed. This should also be further addressed.

#### Reviewer #3

#### (Remarks to the Author)

The authors presented a novel, FIN-based approach for micro-LEDs. This is a very timely, important topic, and the results are interesting and of significance. I recommend publication pending the following revisions.

## 1. The Comparison of different transfer methods

From the introduction part, the idea of this Fin-LEDs with DEP is to reduce the cost and increase the yield. In table S1, DEP method is compared with other transfer methods. But the transfer yield of LASER and Stamp is higher than DEP. LASER also has a small chip size. In the main text, it is mentioned that LASER method may generate more damages and has high requirement of parameters thus increasing cost. However, the analysis of damages is very limited in the following part. For the comparison cost, the parameter requirement is also not very low from your following discussions, so it is a bit confusing about the cost comparison and how did the authors lower the cost by DEP. In addition, the assembly speed is lower in your method.

2. The introduction of DEP method and its advantages is limited. The authors only mentioned that it has several advantages in the line 44 of the page 2, but the advantages are not listed clearly. It is a bit unclear about the motivation of choosing DEP as the method. Did someone else use this method for transfer? Some more literature reviews in this part may be helpful to depict the picture of your idea and let readers know this method.

3. As table S2 shows, the face selective alignment ratio of this work is higher than other nanorod LEDs. But the comparison with other transfer methods is lacked from here. What is the alignment performance of other methods?

4. The authors mentioned that the EQE of DEP Fin-LED is 9.1%, which is a bit lower than nanorod LEDs. Because it is short-wavelength LED, the 9.1% EQE performance is not dominant in the current industry. It seems that there is a trade-off between efficiency and other factors.

5. In figure S1(a), there is a typo. It should be "emission" instead of "emmision".

6. From the section "Properties of vertically oriented Fin-LEDs vs Horizontally Oriented Nanorod-LEDs", the comparison focuses on the Fin-LED and nanorod-LED. The authors mentioned that the MQW volume is an advantage of Fin-LED compared to nanorod LED, but how about the comparison with other structures or methods? Is the MQW and front emission still an advantage?

7. In the Line 103 of page 3, the authors mentioned the decreased surface-defect density and its reason. But it is better if the authors can add some references here to support this conclusion, and the decreased surface-defect density can be reflected by analyzing the EL or PL spectra such as the full-width at half-maximum. The analysis of EL and PL spectra is a bit over-simplified.

8. From the line 111-112, the authors attribute the increasing of emission intensity of Fin-LED to the increased MQW and the decreased defect density. But in the following paragraph, only the influence of MQW on the emission intensity is discussed, the defect density is not mentioned any more. Maybe the explanation of defect or damage is required since this is one of the key factors showing DEP is better than LASER transfer method as the authors discussed in the previous par

9. The Maxwell stress tensor and the finite element method are used to simulate the force and torque. The authors list a reference to verify the dipole approximation method is not suitable, but there is no reference to support the suitability of MST+FEM. Did anyone use these methods to simulate before?

10. In line 176-183 at page 6, the authors summarized the limitation of this simulation, and the observation is observed in the experiments. However, it seems that it is not mentioned any more in the following discussions.

11. In the "Face-Selective Assembly of Fin-LEDs" section, the very detailed comparison among different structures, different shell materials and solvents are conducted. But it looks like this method has a very high demanding of materials used, and different materials have very different performance which may limit the potential wide application. Hence, how do the authors control the cost of this method if there is a high requirement of parameters and materials, and how do the authors show its universality? As the introduction part, the authors attributed the high-cost problem of LASER to the high requirement of the laser parameters and bonding methods, but the authors also need a relatively high demanding in you method. In addition, how do the authors keep the stability of this method if the performance varies so much among different parameters or materials.

12. In line 238-239, the high performance of triple-shell LED is attributed to the effective passivation from face-selective effect, how do the authors prove this conclusion?

13. The optical performance of Fin-LED is only compared with nanorod-LEDs, but the comparison with other structures and methods is also important to show the advantage of the reported method.

14. The luminance of 8640 cd/m2 is one advantage of Fin-LED compared to nanorod-LED, but what about this value in other structures or methods?

15. There are some recent developments of nanowire (or nanorod) LEDs by other growth methods, e.g., MBE, showing significantly improved efficiency for device sizes in the sub-micron regime. These studies should be referenced to provide a

more comprehensive overview of the micro-LED fields.

Version 1:

Reviewer comments:

Reviewer #1

## (Remarks to the Author)

The response prepared by authors answers the question and concerns to a reasonable extent. The advantages of the reported approach and the scope of the fabricated devices and results reported makes this work appropriate for consideration in this journal.

One minor comment is on lines 57-58. "Following the transfer of nanorod-LED display 58 technology from our team, the Samsung Display Corporation (SDC) has successfully demonstrated...". What do authors mean by "transfer". Is this a technology transfer. The relevance of this sentence is not clear.

## Reviewer #2

(Remarks to the Author)

The manuscript has been revised according to the reviewer's comments. I would like to recommend this manuscript for publication in Nature Communications as is.

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## **1 REVIEWER COMMENTS**

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	8

## 9 Dear Reviewer,

In response to the reviewers' comments, we have revised the introduction as follows. We would
appreciate it if you could refer to our responses.

12

## 13 Revised introduction

14 Micro-LEDs have emerged as a star player in next-generation display technology due to their efficient performance, extended lifespan, wide color gamut, modular capabilities, 15 and stability.<sup>1-8</sup> Notably, several companies have introduced TVs featuring micro-LED 16 technology.<sup>9-10</sup> Apple meanwhile planned to incorporate micro-LED displays in its smartwatch 17 scheduled for release in the future.<sup>11-12</sup> Micro-LEDs are positioned as a possible platform 18 technology for a wide range of applications, from compact screens to giant displays. However, 19 20 critical challenges remain in micro-LED display production. A significant concern is the need 21 to improve the yield of the pixel manufacturing process while addressing the substantial fabrication costs associated with micro-LED chip production.<sup>13-16</sup> 22

The production cost of micro-LEDs depends on the chip size, assembly speed, and transfer yield. The smaller the chip size is, the faster the assembly speed is, and the higher the transfer yield is, the lower the unit cost is. Table S1 provides an overview of chip size, assembly speed, and transfer yield for various micro-LED transfer methods. It is crucial to note that the transfer yield depends on precise positioning of micro-LEDs within pixel grooves. Fluidic selfassembly (FSA) exhibits lower selective transfer rates and requires larger micro-LED chips.<sup>10</sup>

In contrast, stamp transfer printing offers the advantages of smaller chip sizes and faster 1 transfer speeds than FSA, albeit with lower repeatability.<sup>10</sup> Despite presenting small chip sizes, 2 rapid transfer speeds, and high accuracy, LASER-based transfer is susceptible to LED chip 3 damage when the chip size becomes smaller.<sup>17</sup> Additionally, it relies on specific laser 4 parameters and bonding methods, with the significant drawback of higher initial costs.<sup>7-10</sup> 5 6 Conventional FSA, laser transfer, and stamp transfer have respective strengths and weaknesses, but each method has critical issues that must be solved prior to commercializing low-cost 7 micro-LEDs. 8

9 One of the micro-LED transfer methods with various sizes, ranging from nano to micrometer, reported to date combines the FSA and dielectrophoretic (DEP) methods. DEP is 10 11 a method of moving materials by controlling the movement of particles under a non-uniform 12 electric field. This is promising for effectively aligning under 10 micron micro-LEDs as it can quickly move particles to a selective location depending on the strength of the electric field.<sup>28</sup> 13 14 LG Electronics (LGE) recently expanded the FSA-DEP approach by aligning ~40µm micro-15 LEDs into subpixel grooves and assembling them using DEP force within the groove through 16 a positive DEP field and secondary magnetic field to improve the site and face-selectivity of 17 micro-LEDs, which cannot be achieved in conventional FSA with high production yield. In 18 contrast, our team introduced an innovative display device and manufacturing process using 19 nanorod-LEDs to effectively address the high material cost of micro-LEDs and the low assembly speed of conventional FSA processes.<sup>19-20</sup> Following the transfer of nanorod-LED 20 21 display technology from our team, the Samsung Display Corporation (SDC) has successfully 22 demonstrated the practical possibility of the DEP process by fabricating a nanorod-LED-based 23 electroluminescent (EL) device through the FSA-DEP process and inkjet printing process. 24 These reports demonstrates the various advantages of inkjet-DEP process, such as low-cost 25 nanorod materials, fast assembly speed matched with commercial inkjet speed, and high possibility of transfer yield, promising a bright future for micro-LED manufacturing.<sup>26-27</sup> 26 27 However, the horizontal orientation of nanorod-LEDs leads to lateral light emission spread, causing limited MQW volume and diminished forward-directed light emission <sup>19-20</sup>, as depicted 28 in Figure 1a. 29

This study suggests adopting fin-LEDs to overcome the challenges presented by nanorod-LEDs and the high costs associated with micro-LED displays. Fin-LEDs are similar in size to nanorod-LEDs and are distinguished by the orientation of the p-GaN/MQW/n-GaN

structure, aligned parallel to the long axis of fin-LEDs. Featuring a vertical LED device 1 2 structure, the fin-LED shows a substantial ~eight-fold increase in MQW volume compared to 3 nanorod-LEDs, leading to enhanced forward-directed light emission (Figure 1a). This effect intensifies when fin-LEDs are face-selectively arranged on lower assembly electrodes, 4 concentrating light emission primarily on the front side. The vertical structure of fin-LEDs 5 6 demands precise alignment for the p-GaN or n-GaN side with the lower assembly electrode to ensure their optical functionality. The DEP field induces motion in fin-LEDs through positive-7 DEP (p-DEP) movement, attracting them perpendicularly between interdigitated 8 electrodes.<sup>20,25,28-30</sup> Achieving the desired face-selective assemblies involves critical rotation 9 around the long axis and optimizing DEP assembly parameters, such as shell and electrode 10 11 material, solvent type, applied frequency, and voltage, for precise placement on bottom 12 electrodes within subpixels. As indicated in prior reports (Table S2), the alignment yield for nanorod-LEDs reached a maximum of 74.4% using DEP assembly.<sup>20</sup> In contrast, our study 13 attained a 91.3% vertical assembly percentage with face-selective DEP, enabling the creation 14 of brighter, vertically structured fin-LED devices and displays. Therefore, the DEP method of 15 16 nanorod-LEDs was enhanced by a combined approach involving rotation around the short axis 17 by the asymmetric field for horizontal alignment and inkjet printing for site-selective assembly. 18 In the case of larger micro-LEDs, the DEP approach was improved by using chip-shaped 19 grooves for site selectivity and additional magnetic fields for face-selective vertical alignment. Here, the DEP of fin-LEDs is enhanced by rotating around a long axis with specific design 20 21 adjustments for face-selective vertical alignment without secondary force. This technique 22 appears complex but has been successfully demonstrated in this study and remains unreported.

23 As shown in Table S2, much higher brightness can be achieved when fin-LEDs are aligned vertically while having a high assembly percentage, whereas horizontally aligned 24 25 nanorod-LEDs have either low brightness or are not mentioned. As previously reported, 26 employing DEP assembly techniques featuring double or triple-shelled nanorod-LEDs with dimensions measuring 600 nm in diameter and 4~5 µm in length, they fabricated 27 electroluminescent (EL) test devices that achieved external quantum efficiency (EQE) levels 28 ranging from 20.2% to 22.2%.<sup>26,27</sup> Previous papers claiming the highest EQE seldom report 29 brightness data. On the other hand, our fin-LED, emitting blue light at a wavelength of 448nm, 30 achieved a reasonable EQE of 9.1% and a peak luminance of 8640  $cd/m^2$  at 5V. This EOE is 31 comparable to the best EQE of nanorod-LEDs reported without using a high indium doped 32 wafer, especially at wavelengths below 455nm.<sup>27</sup> In addition, the brightness of fin-LEDs is 33

## 1 much higher than that of other reported nanorod data.<sup>19,20,26,27,28</sup>

2 Fin-LEDs and nanorod-LEDs, due to their similar shapes and sizes, are well-suited for a color-by-blue approach using quantum dots (QDs). The integration of inkjet and DEP 3 techniques allows us to achieve a full-color display with fin-LEDs, as depicted in Figure S1.<sup>31-</sup> 4 <sup>34</sup> Additionally, we can use inkiet technology to apply a QD-based color conversion layer onto 5 6 red (R) and green (G) subpixels, achieving a wide-gamut RGB color spectrum. The inkjet-DEP 7 process for fin-LEDs can effectively overcome transfer challenges typically encountered with 8 micro-LEDs, even though the conventional FSA method used in this paper allows the assembly 9 of fin-LEDs with somewhat high assembly speed and transfer yield. Similar to the nanorod-10 LEDs, the tiny fin-LED can use inkjet printing to improve site selectivity, transfer yield, and speed and reduce material cost.<sup>26,35</sup> This face-selective DEP-based fin-LED assembly process 11 12 offers notable advantages, including preventing chip damage, achieving rapid assembly speed, and attaining a high pixelation fabrication yield exceeding 99.93%. Our results position this 13 14 process as a promising technology for creating next-generation inorganic displays.

- 1 Reviewer #1 (Remarks to the Author):
- 2

3 This is a careful study presenting two developments: 1) incorporating MQWs on the large facet of a several-micron long cuboid GaN structure (finLED). Authors show an 8 times larger active 4 region relative to nanorodLEDs, leading to brighter fin LEDs. 2) Alignment of finLEDs which 5 includes fin axial direction and rotation and placement of the desired facet on the underlying 6 electrodes using dielectrophoretic (DEP) method. Authors also state "... our study attained a 7 91.3% vertical assembly percentage". This represents the fraction of the fins that face down 8 9 correctly. The facet-controlled placement is introduced to enable a brighter pixel with 8640 cd/m2, while EQE stands at 9.1%. In a sense a better light extraction is obtained relative to 10 11 other types of nanorod LEDs.

12

Authors, as far as I understand, do not disclose a distinct advantage in their DEP assembly 13 14 method (which is the main point of this work) relative to what has been previously reported, for instance, in ref. 26. In other words, their contribution seems gradual and not in a stark 15 16 difference relative to the prior methods. They don't show if their torque-induced rotation is 17 different from the earlier reports or what are the new aspects! It should be noted that the 18 manipulation and placement of micro-finLEDs, several microns long, seems to be a new aspect 19 (relative to ref 26), however, as discussed in my comments below, there are some questions and ambiguities around that. As such this work is not suitable for publication in Nature Comm. 20 21 Below are my questions and comments:

22 Previous research has explored methods for aligning nanorod LEDs horizontally using DEP 23 force that attracts across two-assembled electrodes and torque that rotates around an axis perpendicular to the long axis (a short axis) of the nanorod.<sup>1-6</sup> However, our research takes a 24 novel approach to aligning fin-LEDs vertically using only DEP force and specific torque that 25 rotates around a long axis of the fin-LED, a method not previously reported. Other researchers 26 27 have reported approaches for aligning micro-LEDs vertically by combining DEP with additional forces, focusing on selectively aligning the p-GaN or n-GaN faces in specific 28 directions.<sup>7</sup> Notably, this approach recently used a magnetic field in combination with DEP 29 force to achieve face-selective vertical alignment of micro-LEDs, ensuring that the p-GaN 30 faces align in the same direction.<sup>7</sup> However, the challenge we address is the high cost and 31 complexity of fabricating vertically aligned micro-LED devices. This complexity arises from 32 the need to control DEP and additional forces while fabricating micro-LEDs responsive to these 33

1 forces.

In response to the challenges faced in previous methods, our paper introduces a simple and 2 viable method for aligning ultra-small and ultra-thin devices, such as fin-LEDs, vertically in a 3 face-selective manner using only DEP force and torque. Our approach eliminates the need for 4 5 additional complex forces and control systems, making the alignment process more 6 straightforward. We also demonstrate that fin-LEDs with a structure sensitive to DEP force and 7 torque can be fabricated by adjusting the shell material on the sides and the electrode material on the top. This enables face-selective vertical alignment with DEP alone, confirming the 8 9 advantage of simplicity of our method and facilitating the creation of vertically structured fin-10 LED light-emitting devices and displays.

Our approach is significant because it simplifies the vertical alignment process, reducing the need for complex external adjustments. This concept makes precise vertical alignment feasible and contributes to the advancement of vertical micro/nano-LED devices. By paving the way for more cost-effective and efficient micro/nano-LED displays in the future, our research holds potential for a significant impact in micro/nano-LED display technology.

16 Please refer to the attached revised introduction based on the reviewers' comments.

17

1. Line 223: "We define pixels with 15 or fewer fin-LEDs aligned in a 42x42  $\mu$ m<sup>2</sup> area as bad 18 cells, and out of a total of 2940 pixels, only two defective subpixels were found." It appears 19 20 that their method has no deterministic control over the number of finLEDs per pixel and process 21 seems stochastic; is that true? Based on what criteria, value 15 is chosen as the threshold of 22 normal or bad pixel? The 99.93% assembly accuracy is based on how many experimental runs? 23 Also based on Figure 5f, there seems to be a noticeable variation in the emission intensity of 24 different pixels with the large array. How is this variation contrasted with the # of finLEDs per 25 pixel? Does the poor emission indicate poor contact with the metal electrodes?

26 *Our response*: Thank you for your valuable comments.

We fabricated and evaluated five fin-LED devices, each containing 588 subpixels, and found only two bad subpixels. On average, with one ink drop, approximately 30 fin-LEDs are aligned in the 42  $\times$  42  $\mu$ m<sup>2</sup> area, which we have defined as the optimal LED count for a pixel. This suggests that deterministic control over the number of aligned fin-LEDs is achievable. The threshold of 15 fin-LEDs was chosen as it represents half of the average number of aligned fin-

LEDs, providing a margin for non-operational fin-LEDs while maintaining uniform emission 1 intensity within the  $42 \times 42 \ \mu\text{m}^2$  area. However, if more than 60 fin-LEDs are aligned within a 2 3 pixel, this not only increases the cost but also risks inducing leakage current during device manufacturing, and we define such pixels as defective. To compensate for LEDs that do not 4 turn on due to contact failures, maintaining the number of fin-LEDs between 15 and 60 is 5 6 critical to avoid performance degradation and manufacturing defects. The 99.93% assembly accuracy is derived from five experimental runs, each evaluating 588 pixels, resulting in a total 7 evaluation of 2940 pixels. The variation in emission intensity observed in Figure 6f is closely 8 9 related to the number of fin-LEDs per pixel; as the number of fin-LEDs in a pixel increases, the emission intensity also increases. While poor contact with metal electrodes can indeed 10 11 cause lower emission intensity, other factors, such as a particle issue from using a classroom 12 for a clean room and a misalignment issue of the manual fabrication process, which can cause leakage in pixel lines and affect micro-scale patterns, must also be considered. 13

14 This paper presents a method for area-selective alignment of vertically structured fin-LEDs 15 that entails dropping a fin-LED solution and using DEP assembly. This verifies that fin-LEDs 16 can be vertically aligned within subpixel areas and somewhat evenly distributed across each 17 subpixel. Looking ahead, we plan to utilize inkjet technology to deposit a consistent number 18 of fin-LEDs into the subpixels. Unlike traditional pipette-based dropping methods, the inkjet 19 technique is expected to enhance the distribution of fin-LED uniformity. As previously reported, 20 inkjet printing is already recognized for its ability to precisely place a uniform number of horizontally aligned nanorod LEDs in each pixel.<sup>3</sup> 21

- 22 Based on the comments, the text has been modified as follows.
- 23

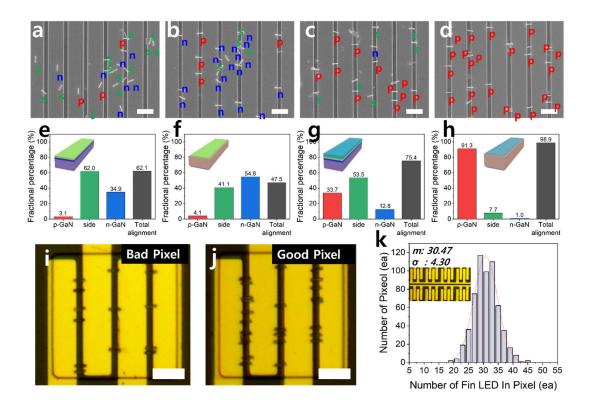
## 24 Before) Page 7, Line 13

Figures 4i-k illustrate images of bad pixels and normal pixels. We define pixels with 15 or fewer fin-LEDs aligned in a  $42x42 \ \mu m^2$  area as bad cells, and out of a total of 2940 pixels, only two defective subpixels were found. Based on this assessment, the assembly accuracy reached 99.93%. Furthermore, the aligned fin-LEDs also show a ~91% face-selective alignment ratio, primarily with the p-GaN side facing over the bottom electrode.

30 After) Page 8, line 19

Figure 5a-b shows images of bad and good pixels. We defined a bad cell as a pixel with 15 or less but 60 or more fin-LEDs aligned in an area of  $42 \times 42 \ \mu m^2$ . This is because fin-LEDs with less than 15 fin-LEDs produce uniform light intensity within the pixel, and fin-LEDs with over 60 fin-LEDs will act as dead pixels due to overlapped alignment of the fin-LEDs. This overlapped alignment can cause leakage current during device processing. The fin-LEDs produced five cells with a total of 588 pixels, and only two defective sub-pixels were found out of a total of 2940 pixels.

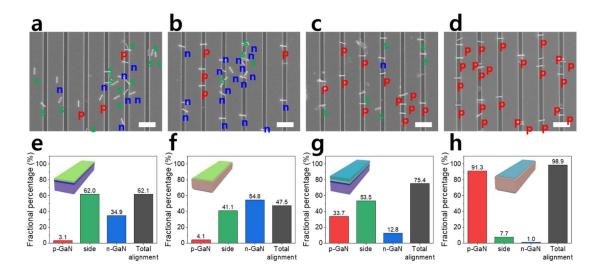
8



9 Before) Figure 4 To reduce confusion, we divided it into two figures.

Figure 4. Self-assembly results of differently structured fin-LEDs by DEP force. a–d, SEM image of fin-LED, fin-LED@SiO<sub>2</sub>, ITO/fin-LED, and ITO/fin-LED@SiO<sub>2</sub> structures. To align the fin-LEDs of all structures, a sinusoidal function with a voltage of 20 V<sub>pp</sub> and a frequency of 10 kHz was applied to the electrodes. To indicate the contact surface between the fin-LEDs and electrodes, different colored letters were marked on the individual LEDs: red p, p-GaN contact; blue n, n-GaN contact; green s, side contact. e–h, Fractional ratios of different contacts of fin-LED, fin-LED@SiO<sub>2</sub>, ITO/fin-LED, and ITO/fin-LED@SiO<sub>2</sub> structures. i–k, Optical

- 1 microscope (OM) image of Fin LED Array Cell assembled using DEP method i. Bad pixel :
- 2 fin LED assembled in pixel (number of fin LEDs : 14 ea) j. Good pixel : fin LED asembled in
- 3 pixle (number of fin LEDs : 25 ea) k. Distribution of fin LED count per pixel , Insect image :
- 4 Fin LED pixel array image (pixel size:  $40 \times 40 \ \mu m^2$ ). All scale bars represent  $10 \ \mu m$ .
- 5



## 6 After) Figure 4 and figure 5

Figure 4. Self-assembly results of differently structured fin-LEDs by DEP force. a–d, SEM image of fin-LED, fin-LED@SiO<sub>2</sub>, ITO/fin-LED, and ITO/fin-LED@SiO<sub>2</sub> structures. To align the fin-LEDs of all structures, a sinusoidal function with a voltage of 20 V<sub>pp</sub> and a frequency of 10 kHz was applied to the electrodes. To indicate the contact surface between the fin-LEDs and electrodes, different colored letters were marked on the individual LEDs: red p, p-GaN contact; blue n, n-GaN contact; green s, side contact. e–h, Fractional ratios of different contacts of fin-LED, fin-LED@SiO<sub>2</sub>, ITO/fin-LED, and ITO/fin-LED@SiO<sub>2</sub> structures.

15

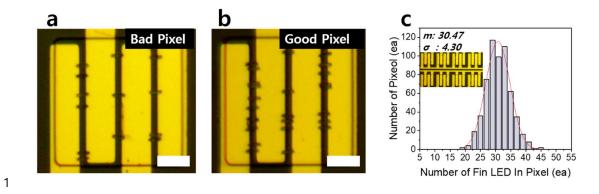
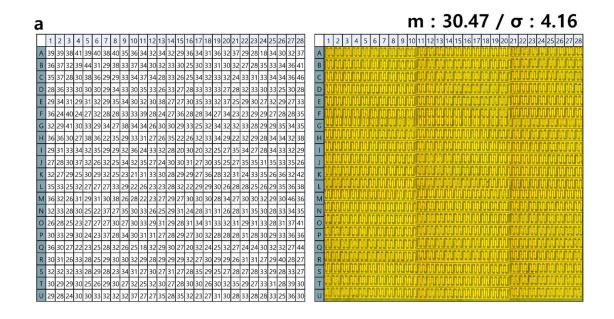
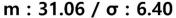
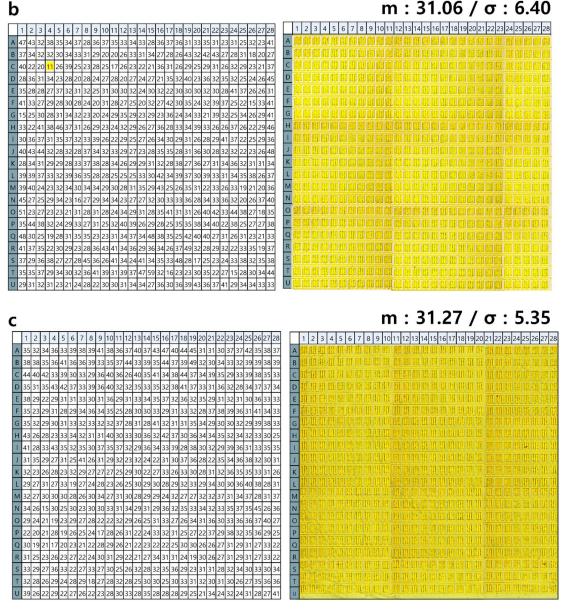


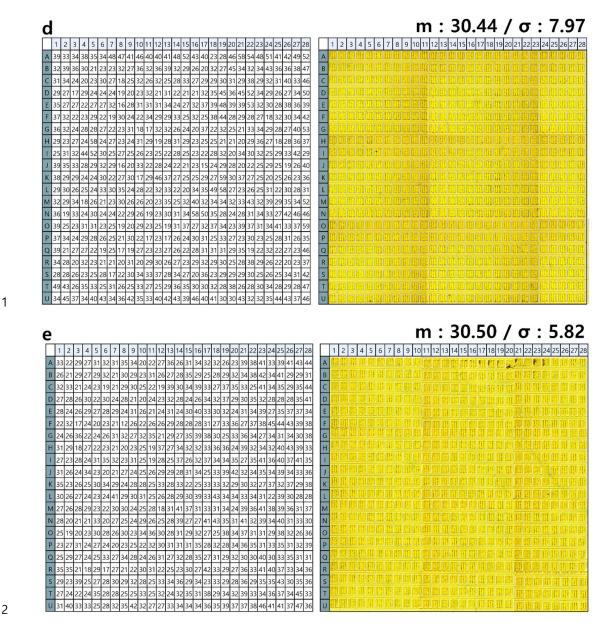
Figure 5. DEP alignment of fin-LEDs in the pixel a-b, Optical microscope (OM) image of
Fin LED Array Cell assembled using DEP method a. Bad pixel: fin LED assembled in pixel
(number of fin LEDs: 14 ea) b. Good pixel: fin LED asembled in pixle (number of fin LEDs:
25 ea) c. Distribution of fin LED count per pixel , Insect image : Fin LED pixel array image
(pixel size: 42 x 42 µm<sup>2</sup>). All scale bars represent 10 µm.

8 After: Supporting Figure 9, data for five devices with 588 pixels has been added.









3 Figure S9. Optical microscope (OM) image of Fin LED Array Cell assembled using DEP method (Left) The number of fin-LEDs that fit into one pixel in the 588 Array Cell. (Right) 4 Full OM image of 21\*28 array cell 5

- 6 2. Line 237 In sentence:"..., namely the triple-shell, SiO2-shell, and TiO2-shell, in terms of brightness", the "triple-shell" seems redundant. 7
- Our response: Thank you for your valuable comments. 8
- 9 We sincerely apologize for any confusion. The explanation of line 237 has been modified.

```
1
      Before) Page 7 Line 27
 2
 3
      Figures 5a and 5b confirm that the triple-shell fin-LED EL device outperforms its counterparts,
      namely the triple- SiO<sub>2</sub>-shell and TiO<sub>2</sub>-shell, in terms of brightness and uniformity of light
 4
      emission.
 5
 6
      After) Page 9, Line 4
 7
 8
      Figures 6a and 6b confirm that the triple-shell fin-LED EL device outperforms its counterparts,
 9
      namely the SiO<sub>2</sub>-shell and TiO<sub>2</sub>-shell, in terms of brightness and uniformity of light emission
10
11
      3. Line 247-249: Figure 5e precedes Figure 5d.
12
      Our response: Thank you for your valuable comments.
      We sincerely apologize for any confusion. The explanation of line s247-249 has been modified.
13
14
      (The order has been changed.)
15
      Before) Page 8, Line 2
16
17
      Figure 5e shows that, compared to micro-LEDs, the fin-LED displays a relatively modest blue
18
      shift with increasing applied voltage. This behavior is attributed to the smaller dimensions of
19
      fin-LEDs, which mitigate the MQW polarization effect. Figure 5d shows the current density-
      voltage (J-V) curve of the fin-LEDs. The turn-on voltages for SiO<sub>2</sub>, TiO<sub>2</sub>, and triple-shelled
20
21
      fin-LEDs are 2.6 V, 2.7 V, and 2.6 V, respectively, with brightness at the turn-on voltage of
22
      10.78 cd/m<sup>2</sup>, 5.1171 cd/m<sup>2</sup>, and 47.495 cd/m<sup>2</sup>, respectively. Among these, the triple-shelled fin-
23
      LED exhibits the lowest leakage current density. This substantial reduction in leakage current
      is thus achieved through improved passivation and face-selective alignment.
24
25
26
      After) Page 9, Line 16
27
28
      Figure 6d shows the current density-voltage (J-V) curve of the fin-LEDs. The turn-on voltages
29
      for SiO<sub>2</sub>, TiO<sub>2</sub>, and triple-shelled fin-LEDs are 2.6 V, 2.7 V, and 2.6 V, respectively, with
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30 brightness at the turn-on voltage of 10.78 cd/m<sup>2</sup>, 5.1171 cd/m<sup>2</sup>, and 47.495 cd/m<sup>2</sup>, respectively.

Among these, the triple-shelled fin-LED exhibits the lowest leakage current density. This substantial reduction in leakage current is thus achieved through improved passivation and face-selective alignment. Figure 6e shows that, compared to micro-LEDs, the fin-LED displays a relatively modest blue shift with increasing applied voltage. This behavior is attributed to the smaller dimensions of the fin-LEDs, which mitigate the MQW polarization effect.

6

4. Line 254: "Figures 5f-g illustrate the array cell containing 588 pixels...". Figure 5g in
comparison with 5f, does not look like the right one as it shows a small pixel.

9 *Our response*: Thank you for your valuable comments.

10 We sincerely apologize for any confusion. The image in figure 5f image has been modified.

11 Before) Figure 5

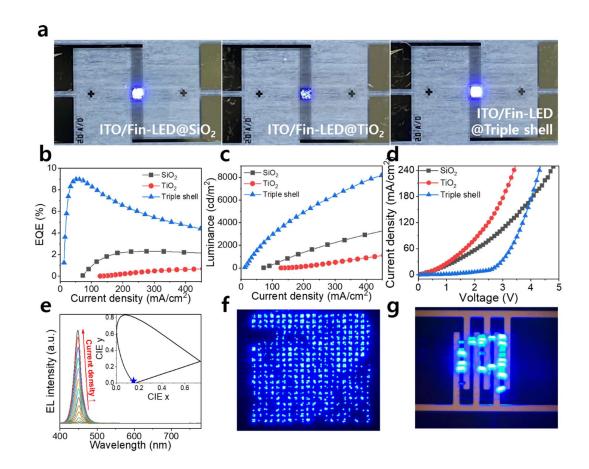


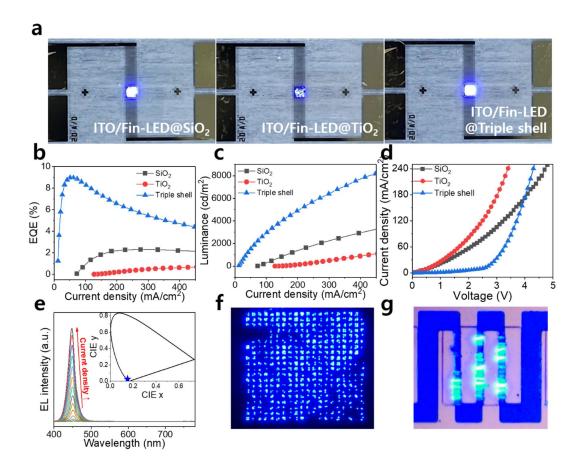


Figure 5. EL characteristics of fin-LED devices according to fin-LED shell materials. a,
Photographs of EL emission images. A voltage of 3.5 V was applied to all devices and the light

emitting area was 1 × 1 mm<sup>2</sup>. b, External quantum efficiency (EQE)-current density curve. c,
Luminance-current density (L-J) curve. d, Current-voltage (I-V) curve in 0 to +5 V range. e,
EL spectra and CIE color coordinates according to current density. f, Fin-LED array cell
emission images of fin-LED array. g, Magnified optical microscope image of fin-LED pixel
(fin-LED array : 21x28 array /total 588 pixels, pixel size: 40 x 40 µm<sup>2</sup>)

6

## 7 After: Figure 6



8

Figure 6. EL characteristics of fin-LED devices according to fin-LED shell materials. a,
Photographs of EL emission images. A voltage of 3.5 V was applied to all devices and the light
emitting area was 1 × 1 mm<sup>2</sup>. b, External quantum efficiency (EQE)-current density curve. c,
Luminance-current density (L-J) curve. d, Current-voltage (I-V) curve in 0 to +5 V range. e,
EL spectra, and CIE color coordinates according to current density. f, Fin-LED array cell
emission images of fin-LED array. g, Magnified optical microscope image of fin-LED pixel
(fin-LED array: 21 × 28 array / total 588 pixels, pixel size: 42 × 42 µm<sup>2</sup>)

5. Line 43. "Upon transferring nanorod-LED display technology by our team, the Samsung Display Company (SDC) reported the successful fabrication of a nanorod-LED-based electroluminescent (EL) device through the FSA-DEP process, highlighting the various advantages of the DEP process". This sounds a causal sentence implying the SDC work was based on the work of this team. Rewording recommended. Perhaps replace "Upon" with "following".

8 *Our response*: Thank you for your valuable comments.

9 We sincerely apologize for any confusion. The explanation in line 43 has been modified.

10

```
11 Before) Page 2, Line 8
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12

Upon transferring nanorod-LED display technology by our team,<sup>21-25</sup> the Samsung Display Company (SDC) reported the successful fabrication of a nanorod-LED-based electroluminescent (EL) device through the FSA-DEP process, highlighting the various advantages of the DEP process.<sup>26-27</sup>

17

18 After) Page 2, Line 22

19

Following the transfer of nanorod-LED display technology from our team, the Samsung Display Corporation (SDC) has successfully demonstrated the practical possibility of the DEP process by the fabrication of a nanorod-LED-based electroluminescent (EL) device through the FSA-DEP process and inkjet printing process. These reports suggested the various advantages of the inkjet-DEP process, such as low-cost nanorod materials, fast assembly speed matching commercial inkjet speed, and high possibility of transfer yield, promising a bright future for micro-LED manufacturing.<sup>26-27</sup>

6. Page 2. "These nanorod-LEDs autonomously align on subpixel electrodes using fluidic
assembly". If they align autonomously, then what is the purpose of the fluidic assembly? Word
"autonomously" should be reworded or omitted.

30 *Our response*: Thank you for your valuable comments.

We sincerely apologize for any confusion. The overall introduction has changed. Please refer
 to the revised introduction provided on the page with Reviewer comment 1.

3

4 7. Line 96: What are the superior optical characteristics that author refer to on line 96. Do they

5 mean a stronger light extraction efficiency? Please clarify.

6 *Our response*: Thank you for your valuable comments.

7 We sincerely apologize for any confusion. The explanation of line 96 has been modified.

8 Before) Page 3, Line 23

9

10 Figure 1b presents SEM images that compare the fabricated fin-LEDs with nanorod-LEDs.

11 The fin-LEDs exhibit a volume and surface area similar to those of nanorod-LEDs but show 12 superior optical characteristics.

13 After) Page 4, Line 12

14

Figure 1b shows an SEM image comparing the fabricated fin-LED and nanorod LED. Fin-LEDs have similar volumes and surface areas as nanorod LEDs, but exhibit superior optical properties due to their vertical emission and large MQW area. Vertical emission corresponds to the direction perceived by the eye.

19

8. Line 50: What is the goal of discussing nanorod LEDs in this work? It is understandable that
finLEDs with a larger active region are better than nanorod LEDs. Perhaps authors should
consider eliminating this section as it seems a distraction from the main point.

23 *Our response*: Thank you for your valuable comments.

24 The discussion of nanorod LEDs in line 50 provides a clear context for the advantages of fin-

25 LEDs in our study. By comparing nanorod LEDs and fin-LEDs, we aim to highlight that fin

26 LEDs offer superior emission efficiency and overall performance due to their larger active

27 MQW areas.

We understand that this discussion may diverge from the main point of the study. However,since most sub-micron LEDs currently being developed are nanorod LEDs, this comparison is

- essential to emphasize the advantages of fin-LEDs, thereby strengthening the importance and
   impact of our work.
- 3

9. Figure 1c. Why the emissions related to the EBL and MQW layers are not apparent in
the nanorod LEDs. In CL, at least, due to its high sensitivity, it should have some signature?

6

7 *Our response*: Thank you for your valuable comments.

8 Table R1 compares the volumes of MQW, EBL, and n-GaN between nanorod LEDs and fin-9 LEDs. In the case of fin-LEDs, the CL spectrum shows a high intensity of the MQW and EBL layers located at the top, similar to an EL device with the P-GaN surface at the top. However, 10 the sensitivity of the n-GaN layer located at the bottom is relatively low. On the contrary, in 11 nanorod LEDs, the CL spectrum, detected from a smaller surface area, shows lower emission 12 13 from the EBL and MQW layers. In contrast, the n-GaN layer, which occupies a larger area, shows higher intensity. Additionally, nanorod LEDs are exposed to plasma for extended periods 14 15 during the fabrication process. This can cause damage to the MQW and AlGaN layers, resulting 16 in lower intensity. The same trend is observed in micro PL measurements (Figure 2b).

	MQW (µm <sup>3</sup> )	EBL (µm <sup>3</sup> )	GaN (µm <sup>3</sup> )
Nanorod LED	0.04	0.01	0.90
Fin LED	0.42	0.14	10.64

<sup>17</sup> **Table R1**. Volume comparison of nanorod and fin-LEDs for MQW, EBL, and GaN

18

19 10. Page 4: line 110: how is the 20-fold CL enhancement calculated? Is it based on analysis
20 of individual structures or their groups?

21

1.Selective-Area Growth of III-Nitride Core-Shell Nanowalls for Light-Emitting and Laser
Diodes. In 2014 Conference on Lasers and Electro-Optics, IEEE: 2014; pp 1-2.

24

25 2.Molecular beam epitaxial growth and characterization of AlN nanowall deep UV light
26 emitting diodes. Appl. Phys. Lett. 2017, 111.

3.High-brightness lasing at submicrometer enabled by droop-free fin light-emitting diodes
 (LEDs). Sci. Adv. 2020, 6, eaba4346.

3 *Our response*: Thank you for your valuable comments.

We calculated the 20-fold CL enhancement based on the values extracted from the CL spectra of nanorod LEDs and fin-LEDs separated from the substrate. This calculation was based on the analysis of individual structures. The papers attached by the reviewer discuss structures similar to fin-LEDs, but they were fabricated using different methods and analyzed in different contexts. Our method specifically focuses on a comparison of individual nanorod and fin-LED structures to accurately determine the CL enhancement.

#### 1 Reviewer #2 (Remarks to the Author):

2

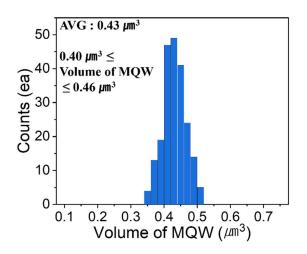
3 In this manuscript, fin-LEDs that overcome the weaknesses previously reported in nanorod-LEDs have been demonstrated. These fin-LEDs show marked improvements in transfer 4 5 efficiency and pixel assembly yield compared to former nanorod-LEDs. The increased volume 6 of the MQW (more than 8 times) and the thickening of the EBL (electron-blocking layer) 7 contribute to the observation of strong blue light (as shown in PL, CL, and EL graphs). 8 Additionally, a triple-shell fin-LED EL device was developed, which demonstrates superior 9 performance in terms of passivation, brightness, and emission uniformity. By enhancing the 10 performance of fin-LEDs, significant contributions to the advancement of next-generation 11 inorganic displays are anticipated. Please refer to the following comments.

12

13 1. The size distribution of fin-LEDs should be provided. The authors used millions of chips for
14 these experiments. However, it seems that the size of the fin-LEDs is not uniform, as shown in
15 Figures 1b and 2f. The size distribution of fin-LEDs might change the EL distribution. All these
16 effects should be addressed.

17 *Our response*: Thank you for your valuable comments.

Figure 2(f) shows that the average size of the fin-LEDs is  $4.0 \times 0.7 \times 1.1 \ \mu$ m, with slight 18 19 variations in length, width, and height within the error range. It has the following aspect ratio 20 structure to ensure the stability of the EL device. Figure 1b shows fin-LEDs randomly 21 distributed on a silicon wafer. Since fin-LEDs have no orientation without an external electric 22 field, they appear to be oriented differently. Because of this, they appear to be of different sizes. 23 Figure R1 provides the size distribution data of fin-LEDs produced on a 4-inch wafer. Since 24 the fin-LED size and MQW do not vary significantly within micrometers (±7.98%), the EL 25 emission wavelength does not differ depending on the size and MQW variation. Additionally, 26 since multiple LEDs are mixed within one pixel, the impact of size and MQW variations on 27 the overall EL distribution is minimized.



2 Figure R1. volume of MQW distributions of fin-LEDs fabricated on 4-inch wafer

1

2. In addition, each fin-LED contains a large number of defects at the edges, as shown in Figure
2f. It is questionable whether the large number of defects in each fin-LED might affect the
uniformity of brightness and, consequently, the uniformity of the pixel. This effect should also
be addressed.

8 *Our response*: Thank you for your valuable comments.

9 The pores on the n-GaN surface that may appear to be as defects are actually a result of the 10 precise electrochemical etching (ECE) method used to separate the fin LED from the substrate. 11 This method, employed in the manufacturing of thin LEDs, creates tiny pores that can increase 12 the internal quantum efficiency (IQE) by relieving the stress of the n-GaN layer and enhancing light extraction. While these pores may seem to affect the uniformity of brightness, our results 13 14 show that the benefits of reduced stress and improved light extraction outweigh any potential 15 negative impact. Furthermore, we have optimized the pore size and distribution to ensure uniformity of the pixel brightness.<sup>8</sup> 16

17

3.In the electric field simulation part, FDEP and TDEP are calculated as functions of θx, as
shown in Figures 3b and 3d. However, further explanation is required for Figures 3b and 3d in
conjunction with Figures 4a to 4h. It is not clear how such a high yield is achieved from the
DEP force or DEP torque differences.

- 1 *Our response*: Thank you for your valuable comments.
- 2 To supplement the answer to the comment, the text has been modified.
- 3

## 4 Before) Page6, Line 14

We fabricated two groups of fin-LED configurations featuring different fin structures 5 (fin-LED, fin-LED@SiO2, ITO/fin-LED, and ITO/fin-LED@SiO2) and different shell 6 materials (ITO/fin-LED@TiO2, ITO/fin-LED@SiNx, ITO/fin-LED@SiO2/Al2O3/SiO2) to 7 8 compare face-selective assembly (SEM images in Figure S6). We conducted DEP assembly 9 tests on four distinct fin-LED structures, visually represented in Figure 4 and elaborated further 10 in Figure S7. Baseline conditions were established at a frequency of 10 kHz and a voltage of 11 20 V<sub>pp</sub>. As seen in Figure 4, various fin-LEDs dispersed in acetone underwent self-assembly 12 on the electrode surface. Observations of the fin-LED structure indicated a substantial presence 13 of non-assembled fin-LEDs, with random placement predominantly on the shell face and at the 14 n-GaN face contacts. In contrast, the ITO/fin-LED structures yielded consistent simulation 15 results, demonstrating an enhanced tendency for p-GaN face contact. Further substantiating 16 these findings, SEM images of the ITO/fin-LED@SiO<sub>2</sub> structure revealed that approximately 17 91.3% of fin-LEDs exhibited selective assembly with a downward orientation, establishing contact between the ITO layer of p-GaN and the bottom electrodes. It was also noted that most 18 19 ITO/fin-LED@SiO<sub>2</sub> displayed alignment tendencies spanning two interdigitated electrodes.

We conducted additional assembly experiments based on the ITO/fin-LED@Shell structure with various shell materials (Figure S7) under different conditions. Face-selective assembly was only possible using low dielectric constant  $SiN_x$  and  $SiO_2$ -shelling ITO/fin-LEDs. In contrast, the high dielectric constant shell of the ITO/fin-LED@TiO\_2 structure did not lead to face-selective alignment, as shown in Figure S7. These results suggest that face-selective assembly is achievable when the dielectric constant of the shelling material surrounding the fin-LED is low.

We also compared alignment based on the ITO/fin-LED@SiO<sub>2</sub> structure as a function of the solvent and determined the optimal face-selective alignment. We performed experiments by comparing their dielectric constants for each medium (hexane, acetone, and IPA). **Figure S8a** illustrates the results of face-selective alignment as a function of applied voltage when the medium is hexane. In addition to the lack of p-GaN or n-GaN face-selective 1 alignment, many fin-LEDs remained unaligned between the electrodes. However, Figures S8a,c demonstrate that acetone and IPA enable face-selective alignment of over 90% between 2 3  $20 \sim 40 \sin(\omega t)$  voltage as the dielectric constant of the medium increases. These figures indicate that face-selective alignment is more achievable when the medium solvent has a higher 4 dielectric constant (e.g., acetone and IPA). Figure S8d confirms that face-selective alignment 5 achieves the highest values at applied frequencies between  $10^1$  and  $10^2$  kHz under an acetone 6 medium using a 20  $sin(\omega t)$  voltage. The decrease in fin-LED rotation at a high frequency of 7  $10^3$  kHz can be attributed to the relatively lower T<sub>DEPx</sub> values of the fin-LED. 8

- Video S1 demonstrates the rapid and precise alignment of fin-LEDs under an electric 9 field, moving precisely toward the positions of the pixels. LEDs that do not align correctly with 10 11 the pixels remain in the solvent during assembly. After completion, any misaligned fin-LEDs can be recovered, leaving no LEDs outside the designated pixel areas. Figures 4i-k illustrate 12 13 images of bad pixels and normal pixels. We define pixels with 15 or fewer fin-LEDs aligned in a 42x42  $\mu$ m<sup>2</sup> area as bad cells, and out of a total of 2940 pixels, only two defective subpixels 14 15 were found. Based on this assessment, the assembly accuracy reached 99.93%. Furthermore, the aligned fin-LEDs also show a ~91% face-selective alignment ratio, primarily with the p-16 GaN side facing over the bottom electrode. Figure S9 illustrates the numbers of assembled fin-17 18 LEDs on each pixel using DEP and ink-dropping processes within an array cell of 588 sub-19 pixels. On average, approximately 26 fin-LEDs are assembled on the designated electrodes and 20 aligned within each subpixel, with a standard deviation of 4.07.
- 21

## After) Page 7, Line 4

We fabricated a group of fin-LED configurations featuring different fin structures 23 24 (fin-LED, fin-LED@SiO<sub>2</sub>, ITO/fin-LED, and ITO/fin-LED@SiO<sub>2</sub>) to compare face-selective 25 assembly (SEM images in Figures S6a-S6d). We conducted DEP assembly tests on four 26 distinct fin-LED structures, visually represented in Figure 4. Baseline conditions were 27 established at a frequency of 10 kHz and a voltage of 20 V<sub>pp</sub>. Various fin-LEDs dispersed in 28 acetone underwent self-assembly on the electrode surface. Self-assembly results for the basic 29 fin-LED structure revealed that a significant number of LEDs were not assembled, and the assembled LEDs were mainly randomly assembled on the side and n-GaN face contacts 30 (Figures 4a and 4e). Adding a SiO<sub>2</sub> shell to the side of the fin-LED (fin-LED@SiO<sub>2</sub>) reduced 31

1 the total alignment of the LEDs compared to the assembled result of the basic fin-LED, but 2 increased the fraction of LEDs aligned to the n-GaN side (Figures 4b and 4f). These 3 differences occurred because the electrical conductivity of SiO<sub>2</sub> used as the shell material was lower than that of the medium. As simple examples, spherical particles composed of  $SiO_2$ 4 experience negative-DEP (n-DEP) in a similar low-frequency range as used for self-assembly 5 6 of fin-LEDs, while spherical particles composed of GaN experience p-GaN in that frequency range.<sup>39,40</sup> Therefore, it is intuitively clear that when the fin-LED@SiO<sub>2</sub> with coexisting p- and 7 n-GaN and SiO<sub>2</sub> experiences DEP, the face on which the LED is aligned on the electrodes is 8 9 determined by the exposed material. Because the magnitude of the electric field increases 10 closer to the electrode, the n-GaN side, which has the highest electrical conductivity among the 11 constituent materials of the fin-LED@SiO<sub>2</sub>, tends to face the electrode. For this reason, it is 12 obvious that ITO/fin-LED in which an ITO layer having much higher electrical conductivity than n-GaN is added on p-GaN has a greater proportion of LEDs aligned with the p-GaN side 13 14 than the n-GaN side on electrodes (Figures 4c and 4g). Similar to the basic fin-LED alignment results, the high proportion of ITO/fin-LED aligned on the side is due to the exposure of the 15 16 side (GaN epi- and ITO layers) of the LED. Ultimately, further substantiating these findings, 17 the SEM image of the ITO/fin-LED@ $SiO_2$  structure revealed that approximately 91.3% of fin-18 LEDs exhibited selective assembly with a downward orientation, establishing contact between 19 the ITO layer of p-GaN and the bottom electrodes (Figure 4d). It was also noted that most ITO/fin-LED@SiO2 displayed alignment tendencies spanning two interdigitated electrodes 20 21 (Figure 4h).

22 We conducted additional assembly experiments based on the ITO/fin-LED@shell 23 structure with various shell materials (ITO/fin-LED@SiO2, ITO/fin-LED@TiO2, ITO/fin-24 LED@SiNx, and ITO/fin-LED@SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub>) under identical conditions (Figures S6d-25 **S6g**). Face-selective assembly was only possible using low dielectric constant  $SiN_x$  and  $SiO_2$ -26 shelling ITO/fin-LEDs. In contrast, the high dielectric constant shell of the ITO/fin-LED@TiO2 27 structure did not lead to face-selective alignment, as shown in Figure S7. These results suggest 28 that face-selective assembly is achievable when the dielectric constant of the shelling material 29 surrounding the fin-LED is lower than that of the medium.

30

4. The bad pixels and good pixels were defined. If a bad pixel is defined as a pixel with 15 or
fewer fin-LEDs, it is anticipated that using a large number of chips for alignment will lead to

fewer bad pixels. Is it possible to quantitatively explain the relationship between the numberof bad pixels and the number of input chips?

3 *Our response*: Thank you for your valuable comments.

4 The definition of a defective pixel is contingent upon pixel size. Assuming 6 to 14 LEDs per 5 row, we have defined the optimal LED count for a  $42x42\mu m^2$  pixel as an average of 30. To 6 compensate for LEDs that do not turn on due to contact failures, we have established a 7 threshold: if the average LED count is less than 15, applying current to more LEDs can negatively impact performance. Additionally, using high-concentration ink limits the number 8 9 of fin-LEDs per pixel, often resulting in discarded LEDs. Arranging more than 60 fin-LEDs 10 within a pixel not only increases the cost but also induces leakage current during device 11 manufacturing, thus defining such pixels as defective. Therefore, the number of fin-LEDs must 12 be appropriately maintained between 15 and 60 to avoid performance degradation and manufacturing defects. This research focuses on utilizing DEP to selectively align fin-LEDs on 13 a surface by controlling the surrounding solvent and fin-LED shell. We have identified the 14 potential for pixelation. 15

16 Please refer to the revised text attached to reviewer 1's question 1.

17

5.The production yield of 99.93% is unclear. Please explain the calculation method of theproduction yield.

20 *Our response*: Thank you for your valuable comments.

We fabricated and evaluated five fin-LED array devices containing 588 pixels to determine the production yield. On average, about 30 fin LEDs (ranging from 27 to 33) are aligned into a pixel measuring  $42x42 \ \mu m^2$  with one ink drop. As noted in the above answer regarding a threshold, we established a bad pixel: the average LED count is less than 15. In evaluating the five fin-LED array devices, only two pixels had fewer than 15 fin-LEDs aligned. Thus, out of 2940 pixels, 2938 pixels had more than 15 fin-LEDs aligned, resulting in a production yield of 99.93%.

28 Please refer to revised **figure S9** attached to reviewer 1's question 1.

6. In Figure 5d, the threshold voltage of the triple-shell LED shifts positively with a steep
increase in current density as the driving voltage increases. However, the mechanism of the
increasing EQE is unclear in the manuscript. This should be further addressed.

4 *Our response*: Thank you for your valuable comments.

The increase in EQE can be attributed to two main factors. First, the triple shell passivates defects on the fin-LED surface, as reported in previous studies. Second, the outermost shell of the triple shell is SiO<sub>2</sub>, which significantly increases the face-selective alignment of fin-LEDs when aligned, similar to the single SiO<sub>2</sub> shells. This minimizes electrical leakage and improves EQE. As a result, despite potential causes of leakage current, such as dust and low-grade lithography facilities, the overall EQE is enhanced by the improved face-selective alignment and passivation provided by triple shelling.

12 We have added an additional explanation to the text as follows.

13

## 14 Before) Page7, Line 23

15 We fabricated three fin-LED electroluminescent (EL) devices using TiO<sub>2</sub>, SiO<sub>2</sub>, and a triple-16 shell SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> configuration. These differently aligned EL devices were manufactured 17 using fin-LEDs, employing SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> triple-shells to reduce surface defects and 18 maintain a high face-selective ratio. Figure 5a visually compares emission images captured from the three types of fin-LED EL devices operated at 5 V. Figures 5a and 5b confirm that 19 20 the triple-shell fin-LED EL device outperforms its counterparts, namely the triple-shell, SiO<sub>2</sub>-21 shell, and TiO<sub>2</sub>-shell, in terms of brightness and uniformity of light emission. The exceptional 22 performance within the triple-shell configuration is attributed to the effective passivation that 23 results from the face-selective effect.

## 24 After) Page 8 Line 34

25 We fabricated three fin-LED electroluminescent (EL) devices using TiO<sub>2</sub>, SiO<sub>2</sub>, and a triple-

shell SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> configuration. These differently aligned EL devices were manufactured

27 using fin-LEDs, employing SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> triple-shells to reduce surface defects and

28 maintain a high face-selective ratio. Additionally, Al<sub>2</sub>O<sub>3</sub> also serves as an etch block layer

29 during device fabrication, and the passivation further complements the E-beam deposition

30 passivation, providing more effective overall passivation.<sup>26-27</sup> Figure 6a visually compares

emission images captured from the three types of fin-LED EL devices operated at 5 V. Figures 6a and 6b confirm that the triple-shell fin-LED EL device outperforms its counterparts, namely the SiO<sub>2</sub>-shell and TiO<sub>2</sub>-shell, in terms of brightness and uniformity of light emission. The difference between the SiO<sub>2</sub>-shell and the TiO<sub>2</sub>-shell appears to be due to face-selective alignment. In addition, since the triple shell shows alignment similar to that of the SiO<sub>2</sub> shell, the difference can be attributed to the passivation effect as well as the superiority of surfaceselective alignment.

8

9 7.In the conclusion, it is mentioned that the EQE is slightly lower than in prior results. However,
10 the method for improving the EQE was not properly addressed. This should also be further
11 addressed.

12 *Our response*: Thank you for your valuable comments.

Several methods with great potential have been proposed to enhance the maximum EQE of ultra-small LEDs. These include growth techniques that significantly reduce defects by controlling indium (In) levels during nano LED fabrication, etching methods that greatly minimize defects of 3.5  $\mu$ m-LED (EQE = ~ 28.5%) through neutral beam etching<sup>9</sup>, and shell coating technologies of nanorod LEDs that perfectly passivate surface defects (EQE = 20.2 ~ 22.2%) using sol-gel SiO<sub>2</sub> coating method.<sup>3</sup>

19 In this paper, we achieved an EQE of  $\sim 9.1\%$  or more, not by using the highest reported EQE 20 methods but by employing conventional dry etching and passivation techniques. We use 21 passivation technology with a SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> triple-shell coating through conventional 22 PECVD and ALD coatings to passivate the surface defects. Face-selective DEP alignment 23 technology is mainly applied to decrease the electrical leakage of misaligned fin-LEDs and 24 increase EQE. While not matching the best previously reported EQEs, our approaches achieve 25 a reasonably high EQE despite triple-shell passivation and low leakage by the face-selective 26 assembly. These challenges include defects and leakage caused by low cleanliness and lower-27 grade lithography equipment typical of academic fabrication facilities.

In future, we can realize a significantly high EQE by obtaining the best-qualified wafer and high indium wafer, reducing etched defects, improving the process environment, and achieving optimal passivation in harmony. This potential for improvement instills hope that, even in the face of challenges, we can enhance the overall performance of DEP fin-LEDs.

- 1 Reviewer #3 (Remarks to the Author):
- 2

The authors presented a novel, FIN-based approach for micro-LEDs. This is a very timely,
important topic, and the results are interesting and of significance. I recommend publication

5 pending the following revisions.

6 1. The Comparison of different transfer methods from the introduction part, the idea of this 7 Fin-LEDs with DEP is to reduce the cost and increase the yield. In table S1, DEP method is compared with other transfer methods. But the transfer yield of LASER and Stamp is higher 8 9 than DEP. LASER also has a small chip size. In the main text, it is mentioned that LASER method may generate more damages and has high requirement of parameters thus increasing 10 11 cost. However, the analysis of damages is very limited in the following part. For the 12 comparison cost, the parameter requirement is also not very low from your following 13 discussions, so it is a bit confusing about the cost comparison and how did the authors lower the cost by DEP. In addition, the assembly speed is lower in your method. 14

15 *Our response*: Thank you for your valuable comments.

For a 55" 4K TV, the required micro-LED chip size is 9 micrometers, while for a 5.8" QHD display, it must be 3 micrometers, with a minimum internal quantum efficiency (IQE) of 25%.<sup>10,11</sup> The largest cost contributor in micro-LED production is the epitaxy process (chip fabrication). Additionally, transferring micro-LEDs using an interposer (groove) increases the overall cost. Therefore, aligning fin-LEDs via FSA and DEP without the use of an interposer and directly transferring the LEDs to achieve a high transfer yield represents a promising technology for the commercialization of micro-LEDs.

Considering these factors, we have revised the introduction and Table S1 accordingly,
representing chip costs relative to chip size. In the case of LLO (Laser Lift-Off), the chip cost
increases as a buffer layer is required to reduce damage to the chip.<sup>12-14</sup> Although many sources
suggest that LLO incurs high costs, specific figures are rarely mentioned.

27

28 Before) Table S1

Ours	Fluid assembly	LASER	Stamp transfer
		Transfer	

		(Dielectricphoresis)			
	Chip size	$0.196 \sim 2.28$	> 400	> 1	> 100
	(µm²)				
	Assembly speed	$\approx$ 1 million per hour	≈50 million per hour	≈100 million per hour	≈1 million p
	-			•	hour
	Transfer yield (%)	99.93	65.00	99.99	99.99
1	Table S1. Co	omparison for micro-LE	D transfer method.		
2					
2 3	1. Lin,	YC., Chen, LY. & Cl	hiu, FC. Lossy Mod	le Resonance-Ba	sed Glucose Se
		YC., Chen, LY. & Cl Dielectric Film. <i>Crystal</i>	-	le Resonance-Ba	sed Glucose Se
3	with High-κ		s <b>9</b> , 450 (2019).		
3 4	with High-κ 2. Zhou	Dielectric Film. Crystal	s <b>9</b> , 450 (2019).	lour conversion	techniques tow
3 4 5	with High-κ 2. Zhou full-colour m	Dielectric Film. <i>Crystal</i> 1, X. et al. Growth, tran	s <b>9</b> , 450 (2019). Insfer printing and co ress in <i>Quantum Elec</i>	lour conversion etronics <b>71</b> , 1002	techniques tow 63 (2020).
3 4 5 6	with High-κ 2. Zhou full-colour m	Dielectric Film. <i>Crystal</i> 1, X. et al. Growth, tran 1icro-LED display. Prog 1, J. E., Park, S., Park, Y.	s <b>9</b> , 450 (2019). Insfer printing and co ress in <i>Quantum Elec</i>	lour conversion etronics <b>71</b> , 1002	techniques tow 63 (2020).
3 4 5 6 7	with High-κ 2. Zhou full-colour m 3. Ryu, 2023, 22049	Dielectric Film. <i>Crystal</i> 1, X. et al. Growth, tran 1icro-LED display. Prog 1, J. E., Park, S., Park, Y.	s <b>9</b> , 450 (2019). nsfer printing and co ress in <i>Quantum Elec</i> ., Ryu, S., Hwang, K	lour conversion <i>etronics</i> <b>71</b> , 1002 ., Jang, H. W. , 2	techniques tow 63 (2020). 4 <i>dvanced Mate</i>
3 4 5 6 7 8	<ul> <li>with High-κ</li> <li>2. Zhou</li> <li>full-colour m</li> <li>3. Ryu,</li> <li>2023, 22049</li> <li>4. Anw</li> </ul>	Dielectric Film. <i>Crystal</i> 1, X. et al. Growth, tran hicro-LED display. Prog J. E., Park, S., Park, Y. 47 (2023).	s <b>9</b> , 450 (2019). Insfer printing and co ress in <i>Quantum Elec</i> ., Ryu, S., Hwang, K P., <i>Laser photoics Re</i>	lour conversion etronics <b>71</b> , 1002 ., Jang, H. W. , 2 ev. <b>16</b> , 2100427 (	techniques tow 63 (2020). 4 <i>dvanced Mate</i> (2022).
3 4 5 6 7 8 9	<ul> <li>with High-κ</li> <li>2. Zhou</li> <li>full-colour m</li> <li>3. Ryu,</li> <li>2023, 22049</li> <li>4. Anw</li> </ul>	Dielectric Film. <i>Crystal</i> 1, X. et al. Growth, tran nicro-LED display. Prog J. E., Park, S., Park, Y. 47 (2023). var, A.R., Lepkowski, S.	s <b>9</b> , 450 (2019). Insfer printing and co ress in <i>Quantum Elec</i> ., Ryu, S., Hwang, K P., <i>Laser photoics Re</i>	lour conversion etronics <b>71</b> , 1002 ., Jang, H. W. , 2 ev. <b>16</b> , 2100427 (	techniques tow 63 (2020). 4 <i>dvanced Mate</i> (2022).

Assamble	Transfer method	Chip size (µm²)	Chip cost	Transfer	Transfer	Transfer	Ref
Assembly				Cost	speed	yield	
Pick up	Elastomer stamp	~1256	Middle	High	1 M/hr	99.50	[1,2]
	LASER	~1	Low~Middle	Middle~High	100M/Hr	99.80	[1,2]
Fluidic assembly	Wave energy	~1256.00	Middle	Middle	NR	97.00	[3]
	Gravity and capillary force	~1256.00	Middle	Middle	1 M/hr	99.90	[4]
	Vander walls force	1256.00	Middle	Middle	54K/hr	99.99	[5]

Molten solder	706.50	Middle	Middle	1 M/hr	99.88	[6]
magnetic-force-assisted dielectrophoretic self- assembly technology	~980.20	Middle	Middle	NR	99.50	[7]
	~0.193	Low	Low	NR	99.98	[8~13]
Dielectricphoresis	2.80	Low	Low	1M/hr	99.93	Our works

2 **Table S1.** Comparison for micro-LED mass transfer method.

3

4 1. Anwar, A.R., Lepkowski, S. P., *Laser photoics Rev.* **16**, 2100427 (2022).

5 2. Chen, D., Chen, Y.-C., Zeng, G., Zhang, D. W. & Lu, H.-L. Integration Technology of
6 Micro-LED for Next-Generation Display. *Research* 6, 0047 (2023).

Ryu, J. J. *et al.* Wave energy-assisted fluidic self-assembly of LED chips for display
applications. *Journal of Information Display* 23, 267–272 (2022).

9 4. Paul J. et al. Display with surface mount emissive elements US patent
10 US009825202B2 (2017)

Hwang, J. *et al.* Wafer-scale alignment and integration of micro-light-emitting diodes
using engineered van der Waals forces. *Nat Electron* 6, 216–224 (2023).

13 6. Lee, D. *et al.* Fluidic self-assembly for MicroLED displays by controlled viscosity.
14 *Nature* 619, 755–760 (2023).

15 7. Chang, W. et al. Concurrent self-assembly of RGB microLEDs for next-generation
16 displays. *Nature* 617, 287–291 (2023).

Park, H. K. et al. Horizontally assembled green InGaN nanorod LEDs: scalable
polarized surface emitting LEDs using electric-field assisted assembly. *Sci. Rep.* 6, 28312
(2016)

Eo, Y. J. et al. Enhanced DC-Operated Electroluminescence of Forwardly Aligned
 p/MQW/n InGaN Nanorod LEDs via DC Offset-AC Dielectrophoresis. *ACS Appl. Mater. Interfaces* 9, 37912–37920 (2017).

<sup>1</sup> 

Sheen, M. et al. Highly efficient blue InGaN nanoscale light-emitting diodes. *Nature* 608, 56–61 (2022).

Cho, H., Kim, D., Lee, S., Yoo, C. & Sim, Y. Efficiency enhancement of submicronsize light-emitting diodes by triple dielectric layers. *J. Soc. Inf. Disp.* 31, 289–297 (2023).

Kim, S. *et al.* Self-array of one-dimensional GaN nanorods using the electric field on
dielectrophoresis for the photonic emitters of display pixel. *Nanoscale Adv.* 5, 1079–1085
(2023).

8 13. Kim, T., Uthirakumar, P., Cho, Y.-H., Nam, K. H. & Lee, I.-H. Enhanced quantum
9 efficiency of horizontally aligned individual InGaN/GaN nanorod LEDs by self-assembled Ag
10 nanoparticles. *Applied Surface Science* 656, 159706 (2024).

11

12 Before) Page 1, Line 31

Table S1 provides an overview of chip size, assembly speed, and transfer yield for various 13 14 micro-LED transfer methods. It is crucial to note that the transfer yield depends on the precise positioning of micro-LEDs within pixel grooves. Fluidic self-assembly (FSA) exhibits lower 15 selective transfer rates and requires larger micro-LED chips.<sup>10</sup> In contrast, stamp transfer 16 printing offers the advantages of smaller chip sizes and faster transfer speeds than FSA, albeit 17 with lower repeatability.<sup>10</sup> Despite presenting small chip sizes, rapid transfer speeds, and high 18 accuracy, LASER-based transfer is susceptible to LED chip damage.<sup>17</sup> Additionally, it relies 19 20 on specific laser parameters and bonding methods, with the significant drawback of higher initial costs.7-10 21

22

After) Page 1, Line 34

The production cost of micro-LED depends on the chip size, assembly speed, and transfer yield. The smaller the chip size is, the faster the assembly speed is, and the higher the transfer yield is, the lower the unit cost is. **Table S1** provides an overview of chip size, assembly speed, and transfer yield for various micro-LED transfer methods. It is crucial to note that the transfer yield depends on the precise positioning of micro-LEDs within pixel grooves. Fluidic selfassembly (FSA) exhibits lower selective transfer rates and requires larger micro-LED chips.<sup>10</sup>

In contrast, stamp transfer printing offers the advantages of smaller chip sizes and faster 1 transfer speeds than FSA, albeit with lower repeatability.<sup>10</sup> Despite presenting small chip sizes, 2 rapid transfer speeds, and high accuracy, LASER-based transfer is susceptible to LED chip 3 damage when the chip size becomes smaller.<sup>17</sup> Additionally, it relies on specific laser 4 parameters and bonding methods, with the significant drawback of higher initial costs.<sup>7-10</sup> 5 6 Conventional FSA, laser transfer, and stamp transfer have respective strengths and weaknesses, but each method has critical issues that must be solved prior to commercializing low-cost 7 micro-LEDs. 8

9

10 2. The introduction of DEP method and its advantages is limited. The authors only mentioned 11 that it has several advantages in the **line 44 of the page 2**, but the advantages are not listed 12 clearly. It is a bit unclear about the motivation of choosing DEP as the method. Did someone 13 else use this method for transfer? Some more literature reviews in this part may be helpful to 14 depict the picture of your idea and let readers know this method. 15 More superiors Luminance properties

16 *Our response*: Thank you for your valuable comments.

17 We sincerely apologize for any confusion. The explanations of page 1 line 35 to page 2 line 318 have been modified.

19 Before) Page 2, Line 4

20 LG Electronics (LGE) recently expanded the FSA-DEP approach, aligning ~40 µm micro-LEDs to subpixel grooves, achieving high assembly production yields with a positive DEP 21 field and a secondary magnetic field.<sup>18</sup> In contrast, to effectively address the challenges of 22 micro-LEDs and FSA processes, our team introduced an innovative display device and 23 manufacturing process using nanorod-LEDs.<sup>19-20</sup> Upon transferring nanorod-LED display 24 technology by our team,<sup>21-25</sup> the Samsung Display Company (SDC) reported the successful 25 26 fabrication of a nanorod-LED-based electroluminescent (EL) device through the FSA-DEP process, highlighting the various advantages of the DEP process.<sup>26-27</sup> These nanorod-LEDs 27 autonomously align on subpixel electrodes using fluidic assembly, including inkjet printing and 28 DEP assembly processes, although no reported transfer yield data are available.<sup>26-28</sup> However, 29 the horizontal orientation of nanorod-LEDs leads to lateral light emission spread, causing 30 limited MQW volume and diminished forward-directed light emission<sup>19-20</sup>, as depicted in 31

## 1 Figure 1a.

## 2 After) Page 2, Line 12

The micro-LED transfer method under 10 micrometers reported to date is a method that 3 combines the FSA and DEP method. DEP is a method of moving materials by controlling the 4 5 movement of particles under a non-uniform electric field. This is promising for effectively 6 aligning under 10 micron micro-LEDs as it can quickly move particles to a selective location depending on the strength of the electric field.<sup>[28]</sup> LG Electronics (LGE) recently expanded the 7 FSA-DEP approach by aligning ~40µm micro-LEDs into subpixel grooves and assembling 8 9 them using DEP force within the groove through a positive DEP field and secondary magnetic field to improve the site and face-selectivity of micro-LEDs, which cannot be achieved in 10 11 conventional FSA with high production yield. In contrast, our team introduced an innovative 12 display device and manufacturing process using nanorod-LEDs to effectively address the high material cost of micro-LEDs and the low assembly speed of conventional FSA processes.<sup>19-20</sup> 13 Following the transfer of nanorod-LED display technology from our team, the Samsung 14 Display Corporation (SDC) has successfully demonstrated the practical possibility of the DEP 15 16 process by fabricating a nanorod-LED-based electroluminescent (EL) device through the FSA-17 DEP process and inkjet printing process. These reports demonstrates the various advantages of inkjet-DEP process, such as low-cost nanorod materials, fast assembly speed matched with 18 commercial inkjet speed, and high possibility of transfer yield, promising a bright future for 19 micro-LED manufacturing.<sup>26-27</sup> 20

3. As table S2 shows, the face selective alignment ratio of this work is higher than other nanorod
LEDs. But the comparison with other transfer methods is lacked from here. What is the
alignment performance of other methods?

24

## 25 *Our response*: Thank you for your valuable comments.

Only one transfer method, excluding DEP, has been reported for chip sizes of 10 µm<sup>2</sup> or less.
This method involves separating nanorod LEDs from the wafer using laser lift-off (LLO). The
transfer yield for this method is reported to be 99.5%. However, while LLO shows promise for

29 separating nano-LEDs, the performance of the devices after the transfer was not reported.<sup>15</sup> In

30 contrast, the reported DEP transfer of nanorod LEDs shows promise for high transfer yields

and device performance in sub-micron scale LEDs. On this basis, fin-LEDs with face-selective
 alignment show significant potential.

3

4 4. The authors mentioned that the EQE of DEP Fin-LED is 9.1%, which is a bit lower than
5 nanorod LEDs. Because it is short-wavelength LED, the 9.1% EQE performance is not
6 dominant in the current industry. It seems that there is a trade-off between efficiency and other
7 factors.

8 *Our response*: Thank you for your valuable comments.

In the case of the reported nanorod LED, a wafer with a high indium content was used.<sup>4</sup>As the 9 10 indium content increases, non-radiative recombination due to defects occurring during dry 11 etching decreases. Additionally, the relatively low EQE of our DEP fin-LEDs can be attributed 12 to the research being conducted in a school-level clean room environment, where particle 13 contamination (index of particles larger than  $0.1 \mu m$  in  $1 m^3$  air) can cause leakage sources. 14 Furthermore, passivation using the sol-gel method involves the formation of nanoparticles, which can generate particles that affect the performance evaluation when applied to actual fin-15 LEDs.<sup>3</sup> We can realize a significantly high EQE by obtaining the best-qualified wafer and high 16 17 indium wafer, reducing etched defects, improving the process environment, and achieving 18 optimal passivation in harmony. This potential for improvement instills hope that, even in the 19 face of challenges, we can enhance the overall performance of DEP fin-LEDs.

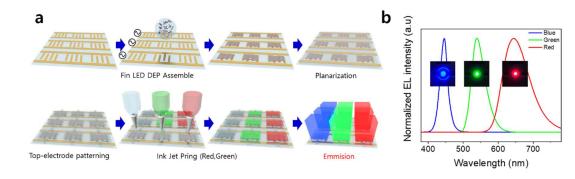
20

5. In figure S1(a), there is a typo. It should be "emission" instead of "emission".

22 *Our response*: Thank you for your valuable comments.

23 We sincerely apologize for any confusion. Figure S1 (a) has been modified.

24 Before)



1

2 **Figure S1.** (a) Schematic of Implementing a Full-Color Fin LED Display. (b) Fin LED Color

- 3 Conversion RGB EL Spectrum (Blue: Fin LED, Green: Fin LED@InP QDs Color Film, Red:
- 4 Fin LED@R6832 Phosphor Color Film.
- 5
- 6
- 7 After)

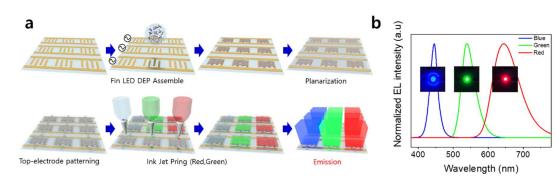


Figure S1. (a) Schematic of Implementing a Full-Color Fin LED Display. (b) Fin LED Color
Conversion RGB EL Spectrum (Blue: Fin LED, Green: Fin LED@InP QDs Color Film, Red:

- 11 Fin LED@R6832 Phosphor Color Film.
- 12

8

6. From the section "Properties of vertically oriented Fin-LEDs vs Horizontally Oriented
Nanorod-LEDs", the comparison focuses on the Fin-LED and nanorod-LED. The authors
mentioned that the MQW volume is an advantage of Fin-LED compared to nanorod LED, but
how about the comparison with other structures or methods? Is the MQW and front emission
still an advantage?

18 *Our response*: Thank you for your valuable comments.

1 As summarized in Table R2, most sub-micron-scale LEDs, excluding nanorod LEDs, currently 2 adopt a front emission structure. Compared to the side emission in nanorod LEDs, front 3 emission does not require additional reflective processes within the device to enhance light 4 extraction efficiency. Additionally, it can be confirmed that the light is emitted in the direction 5 recognized. This makes front emission particularly advantageous in sub-micron LEDs. 6 Furthermore, the slightly larger MQW volume in fin-LEDs remains an advantage compared to 7 other structures or methods. The increased active region in fin-LEDs can contribute to higher 8 emission efficiency and overall performance.

9

Process	SS		Luminance (cd/m²)	EQE (%)	Ref	
Top down	Mesa	Front	12.25	Non-reported	26	[9]
Top down	SAMs	Front	0.44	1070	6.21	[8]
Top down	LLO	Front	0.785	Non-reported	Non- reported	[15]
Bottom up	MBE	Front	16	~10 <sup>6</sup>	0.2	[14]
Top down	DEP	Front	2.8	8640	9.1	Our works

10 Table R2. Sub-micron LED transfer method and luminance properties.

11

12 7. In the Line 103 of page 3, the authors mentioned the decreased surface-defect density and 13 its reason. But it is better if the authors can add some references here to support this conclusion, 14 and the decreased surface-defect density can be reflected by analyzing the EL or PL spectra 15 such as the full-width at half-maximum. The analysis of EL and PL spectra is a bit over-16 simplified.

17 *Our response*: Thank you for your valuable comments.

18 In the case of LEDs fabricated from InGaN/GaN MQW wafers grown on the c-plane using a

top-down approach, surface dangling bonds generated during the dry etching process act as non-radiative recombination centers. Due to their long length, nanorod LEDs require a longer etching time. This prolonged etching time results in extensive exposure to plasma, generating a large amount of Ga-O vacancies.<sup>3,9</sup> In contrast, fin-LEDs have a lower height, resulting in a shorter etching time than nanorod LEDs. This can be confirmed through a XPS analysis, which shows that the Ga-O bond ratio for nanorod LEDs is 21.62%, whereas for fin-LEDs, it is 15.46%, indicating fewer defects.

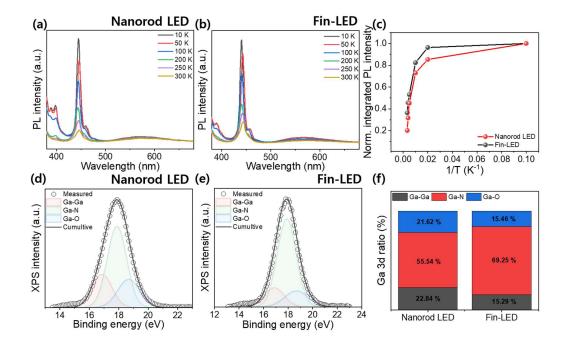
8 Additionally, these defects can be further understood through the internal quantum efficiency

9 (IQE). Nanorod LEDs have an IQE of 20.2 0%, while fin-LEDs have a higher IQE of 36.44%,

10 indicating fewer defects in fin-LEDs.

11 Regarding EL characteristics, horizontal methods face challenges in achieving contact at the 12 laboratory stage, as 2–3 micrometer scale is challenging to achieve without external factors 13 such as dust. Reported nanorods efficiencies, such as those using SDC, show high efficiency 14 but are produced in highly controlled environments and do not mention luminance.<sup>3,4</sup> Moreover, 15 nanorod LEDs reported by *Kims,s et al.* show a low EL efficiency of 2%.<sup>5</sup> Therefore, it can be 16 concluded that fin-LEDs have fewer defects compared to nanorod LEDs during the 17 manufacturing process.

18



1 Figure R2. (a-c) Temperature dependence photoluminescence (TDPL) spectra of nanorod LED

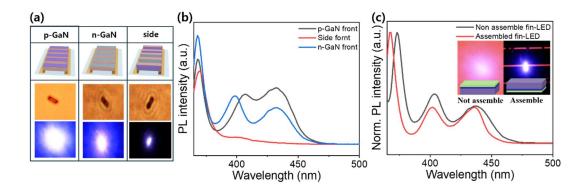
2 and fin-LED (d-e) Ga 3d X-ray photoelectron spectroscopy (XPS) spectra of nanorod LED and

3 fin-LED (f) Ga 3d ratio of nanorod LED and fin-LED obtained XPS spectra

8. From the line 111-112, the authors attribute the increasing of emission intensity of Fin-LED to the increased MQW and the decreased defect density. But in the following paragraph, only the influence of MQW on the emission intensity is discussed, the defect density is not mentioned any more. Maybe the explanation of defect or damage is required since this is one of the key factors showing DEP is better than LASER transfer method as the authors discussed in the previous par

10 *Our response*: Thank you for your valuable comments.

In the case of LASER translocation, defects occur in the MQW due to screw dislocation in GaN from the LASER.<sup>12</sup> To reduce this damage, an additional buffer layer is added, increasing the thickness of the chip.<sup>13,14</sup> This buffer layer incurs additional costs. However, in the case of DEP, there is no damage because the LED is separated from the wafer using ECE. In addition, when measuring the micro PL of LEDs aligned using DEP, there is no significant difference in the excitation intensity, and thus it can be stated that the DEP method causes less damage.



17

Figure R3. (a) Excitation image and micro PL spectra of fin-LED direction (c) micro PL
spectra before and after alignment

20 9. The Maxwell stress tensor and the finite element method are used to simulate the force and

21 torque. The authors list a reference to verify the dipole approximation method is not suitable,

22 but there is no reference to support the suitability of MST+FEM. Did anyone use these methods

23 to simulate before?

24 *Our response*: Thank you for your valuable comments.

2

## 3 Before) Page 6 Line 14

We fabricated two groups of fin-LED configurations featuring different fin structures 4 5 (fin-LED, fin-LED@SiO2, ITO/fin-LED, and ITO/fin-LED@SiO2) and different shell materials (ITO/fin-LED@TiO<sub>2</sub>, ITO/fin-LED@SiN<sub>x</sub>, ITO/fin-LED@SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub>) to 6 7 compare face-selective assembly (SEM images in Figure S6). We conducted DEP assembly 8 tests on four distinct fin-LED structures, visually represented in Figure 4 and elaborated further 9 in Figure S7. Baseline conditions were established at a frequency of 10 kHz and a voltage of 20 V<sub>pp</sub>. As seen in Figure 4, various fin-LEDs dispersed in acetone underwent self-assembly 10 11 on the electrode surface. Observations of the fin-LED structure indicated a substantial presence 12 of non-assembled fin-LEDs, with random placement predominantly on the shell face and at the 13 n-GaN face contacts. In contrast, the ITO/fin-LED structures yielded consistent simulation 14 results, demonstrating an enhanced tendency for p-GaN face contact. Further substantiating 15 these findings, SEM images of the ITO/fin-LED@SiO<sub>2</sub> structure revealed that approximately 16 91.3% of fin-LEDs exhibited selective assembly with a downward orientation, establishing contact between the ITO layer of p-GaN and the bottom electrodes. It was also noted that most 17 18 ITO/fin-LED@SiO2 displayed alignment tendencies spanning two interdigitated electrodes.

We conducted additional assembly experiments based on the ITO/fin-LED@Shell structure with various shell materials (Figure S7) under different conditions. Face-selective assembly was only possible using low dielectric constant  $SiN_x$  and  $SiO_2$ -shelling ITO/fin-LEDs. In contrast, the high dielectric constant shell of the ITO/fin-LED@TiO\_2 structure did not lead to face-selective alignment, as shown in Figure S7. These results suggest that face-selective assembly is achievable when the dielectric constant of the shelling material surrounding the fin-LED is low.

26

# 27 After) Page 7, Line 4

We fabricated a group of fin-LED configurations featuring different fin structures
(fin-LED, fin-LED@SiO<sub>2</sub>, ITO/fin-LED, and ITO/fin-LED@SiO<sub>2</sub>) to compare face-selective
assembly (SEM images in Figures S6a–S6d). We conducted DEP assembly tests on four

distinct fin-LED structures, visually represented in Figure 4. Baseline conditions were 1 established at a frequency of 10 kHz and a voltage of 20 V<sub>pp</sub>. Various fin-LEDs dispersed in 2 3 acetone underwent self-assembly on the electrode surface. Self-assembly results for the basic fin-LED structure revealed that a significant number of LEDs were not assembled, and the 4 assembled LEDs were mainly randomly assembled on the side and n-GaN face contacts 5 (Figures 4a and 4e). Adding a SiO<sub>2</sub> shell to the side of the fin-LED (fin-LED@SiO<sub>2</sub>) reduced 6 the total alignment of the LED compared to the assembled result of the basic fin-LED, but 7 increased the fraction of LEDs aligned to the n-GaN side (Figures 4b and 4f). These 8 differences occurred because the electrical conductivity of SiO<sub>2</sub> used as the shell material was 9 10 lower than that of the medium. As simple examples, spherical particles composed of  $SiO_2$ 11 experience negative-DEP (n-DEP) in a similar low-frequency range as used for self-assembly of fin-LEDs, while spherical particles composed of GaN experience p-GaN in that frequency 12 range.<sup>38,39</sup> Therefore, it is intuitively clear that when the fin-LED@SiO<sub>2</sub> with coexisting p- and 13 n-GaN and SiO<sub>2</sub> experiences DEP, the face on which the LED is aligned on the electrodes is 14 determined by the exposed material. Because the magnitude of the electric field increases 15 16 closer to the electrode, the n-GaN side, which has the highest electrical conductivity among the 17 constituent materials of the fin-LED@SiO<sub>2</sub>, tends to face the electrode. For this reason, it is 18 obvious that ITO/fin-LED in which an ITO layer having much higher electrical conductivity 19 than n-GaN is added on p-GaN has a greater proportion of LEDs aligned with the p-GaN side 20 than the n-GaN side on electrodes (Figures 4c and 4g). Similar to the basic fin-LED alignment 21 results, the high proportion of ITO/fin-LED aligned on the side is due to the exposure of the 22 side (GaN epi- and ITO layers) of the LED. Ultimately, further substantiating these findings, 23 the SEM image of the ITO/fin-LED@SiO<sub>2</sub> structure revealed that approximately 91.3% of fin-24 LEDs exhibited selective assembly with a downward orientation, establishing contact between 25 the ITO layer of p-GaN and the bottom electrodes (Figure 4d). It was also noted that most ITO/fin-LED@SiO<sub>2</sub> displayed alignment tendencies spanning two interdigitated electrodes 26 27 (Figure 4h).

We conducted additional assembly experiments based on the ITO/fin-LED@shell structure with various shell materials (ITO/fin-LED@SiO<sub>2</sub>, ITO/fin-LED@TiO<sub>2</sub>, ITO/fin-LED@SiN<sub>x</sub>, and ITO/fin-LED@SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub>) under identical conditions (Figures S6d– S6g). Face-selective assembly was only possible using low dielectric constant SiN<sub>x</sub> and SiO<sub>2</sub>shelling ITO/fin-LEDs. In contrast, the high dielectric constant shell of the ITO/fin-LED@TiO<sub>2</sub> structure did not lead to face-selective alignment, as shown in Figure S7. These results suggest that face-selective assembly is achievable when the dielectric constant of the shelling material
 surrounding the fin-LED is lower than that of the medium.

3 References

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9 10. In line 176-183 at page 6, the authors summarized the limitation of this simulation, and the
10 observation is observed in the experiments. However, it seems that it is not mentioned any
11 more in the following discussions.

12 *Our response*: Thank you for your valuable comments.

To address this, we would like to clarify and expand on the implications of these limitations in our study. The limitations mentioned specifically—the simplified geometric assumptions made during the simulation, such as modeling the fin-LED as a straight cuboid shape and not accounting for potential deformations of the n-GaN layer caused by the electrochemical etching (ECE) process—were crucial considerations in our analysis. These simplifications were necessary to manage the complexity of the simulation but inevitably introduced some discrepancies between the simulated and observed behaviors.

Despite these limitations, our experimental results were largely consistent with the simulation predictions. For example, the periodic variations in DEP torque observed in the simulation could not fully explain the selective alignment of the fin-LEDs. However, in the experiments, we observed that the actual alignment was influenced by additional factors, such as the physical properties of the n-GaN layer, which may have been altered by the ECE process, leading to lower electrical conductivity and affecting the DEP behavior.

While we briefly mentioned these points in the manuscript, we realize that further discussion would have strengthened the interpretation of our results. Therefore, we will ensure that the final manuscript includes a more detailed discussion on how these limitations impacted our findings and how they were addressed in our experimental procedures. This will help provide a more comprehensive understanding of the relationship between our simulations and

#### 1 experimental outcomes.

2

11. In the "Face-Selective Assembly of Fin-LEDs" section, the very detailed comparison 3 among different structures, different shell materials and solvents are conducted. But it looks 4 5 like this method has a very high demanding of materials used, and different materials have very 6 different performance which may limit the potential wide application. Hence, how do the 7 authors control the cost of this method if there is a high requirement of parameters and materials, and how do the authors show its universality? As the introduction part, the authors attributed 8 9 the high-cost problem of LASER to the high requirement of the laser parameters and bonding methods, but the authors also need a relatively high demanding in you method. In addition, 10 how do the authors keep the stability of this method if the performance varies so much among 11 12 different parameters or materials.

13 *Our response*: Thank you for your valuable comments.

The employed materials are currently used in submicron LEDs. Referring to Table R3, most micro-LEDs below 10 micrometers incorporate SiO<sub>2</sub> passivation. Additionally, PEG is a suitable solvent for inkjet, and its properties allow for face-selective alignment of fin-LEDs. Laser-assisted transfer technology successfully transfers numerous micro-LEDs by selectively irradiating a UV laser. However, it is not easy to apply in mass production due to precise laser power and resolution control, consideration of complex material absorption coefficient, and

strict requirements.<sup>14</sup> Although the requirements for materials may be relatively high, the
materials used are those used for micro LEDs of 10 micrometers or less.

22 Utilizing the SiO<sub>2</sub> shell commonly used for LEDs below 10 micrometers, the process for 23 shelling fin-LEDs is remarkably simple. Unlike other LEDs, they do not require additional 24 shelling for DEP alignment and only require shelling for device stability. This simplicity in 25 fabrication adds to the appeal of fin-LEDs, although many different materials appear to be 26 needed in different stages when making fin-LEDs. Additionally, inkjet-based patterning using 27 PEG solvent allows for control of the dielectric constant and viscosity without additional high 28 cost. As the optimum number of LEDs within a pixel decreases, alignment speeds can increase, making inkjet-based transfer faster.<sup>3,16</sup> 29

30

Chip size (µm)		Asse mble	Passivation		Properties			Ref	
Chip size (µm)	Chip size(µm 2)		Passivation	Materials	EQE (%)	Brightness (cd/m <sup>2</sup> )	Wavelength (nm)		
3.5*3.5	12.25	Mesa	PECVD	SiO2 150 nm	38	NR	NR	[9]	
3.5*3.5	12.25	Mesa	PECVD	SiO <sub>2</sub> 150 nm	28.50	NR	NR	[9]	
0.53	0.22	DEP	Solgel	SiO <sub>2</sub> 20nm Al <sub>2</sub> O <sub>3</sub> 40 nm	20.2	NR	450	[3]	
0.53	0.22	DEP	PEALD	SiO <sub>2</sub> 20nm Al <sub>2</sub> O <sub>3</sub> 40 nm	8.9	NR	450	[3]	
0.58	0.26	DEP	PEALD/Thermal ALD	SiO <sub>2</sub> 20nm Al <sub>2</sub> O <sub>3</sub> 40 nm	19.6	NR	464	[4]	
0.58	0.26	DEP	Thermal ALD/PEALD/Thermal ALD	HfO <sub>2</sub> 2 nm SiO <sub>2</sub> 20nm Al <sub>2</sub> O <sub>3</sub> 40 nm	22.2	NR	464	[4]	
0.58	0.26	DEP	Thermal ALD/PEALD/Thermal ALD	SiO <sub>2</sub> 2 nm SiO <sub>2</sub> 20nm Al <sub>2</sub> O <sub>3</sub> 40 nm	18.8	NR	464	[4]	

# 1 Table R3. Table of sub-micron LED passivation

2 12. In line 238-239, the high performance of triple-shell LED is attributed to the effective

3 passivation from face-selective effect, how do the authors prove this conclusion?

4 *Our response*: Thank you for your valuable comments.

5 In Figure 4d, there is no significant difference in alignment between the  $SiO_2$  single shell and the triple shell. This suggests that surface-selective alignment is achieved due to the presence 6 of SiO<sub>2</sub> on the outermost layer of the triple shell. The middle layer, Al<sub>2</sub>O<sub>3</sub>, acts as an etching 7 block layer during device fabrication, enhancing device efficiency by preventing etching 8 9 damage. Additionally, Al<sub>2</sub>O<sub>3</sub> passivation further complements the E-beam-deposited 10 passivation, providing more effective overall passivation. A fin-LED with a SiO<sub>2</sub> single shell 11 was etched and failed during device manufacturing, and thus a single-shell and a triple-shell 12 fin-LED device cannot be compared. A previous paper already reported that the SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> double shell has a better passivation effect than the SiO<sub>2</sub> single shell to obtain maximum 13 efficiency in nanorods.<sup>3,4</sup> See also our response to Reviewer 2's question 6. 14

15

16 13. The optical performance of Fin-LED is only compared with nanorod-LEDs, but the 17 comparison with other structures and methods is also important to show the advantage of the 18 reported method.

19 *Our response*: Thank you for your valuable comments.

20 Referring to Table R2, most groups that report LED efficiencies below 10 micrometers

mention EQE but not luminance. Additionally, the mesa structure requires two electrodes in
 one plane, and thus the presence of the electrodes reduces the effective light extraction area.
 On the other hand, fin-LEDs with a top-down contact structure have a larger light extraction
 area, making this technology more advantageous.

5

14. The luminance of 8640 cd/m<sup>2</sup> is one advantage of Fin-LED compared to nanorod-LED, but
what about this value in other structures or methods?

8 *Our response*: Thank you for your valuable comments.

9 Referring to **Table R2**, most groups reporting the efficiency of LEDs smaller than 10 10 micrometers mention EQE but do not provide information on luminance. The dot LED, which 11 has a similar structure to fin-LEDs, exhibits a luminance of approximately 1070 cd/m<sup>2</sup>. 12 Additionally, LEDs with a mesa structure require an electrode on the top, which reduces the 13 light-emitting area compared to the actual chip size, potentially resulting in lower luminance. 14 Therefore, the competitive and relatively high luminance of 8640 cd/m<sup>2</sup> for fin-LEDs using a 15 top-down contact is a testament to the contribution and possibility of our research.

16

17 15. There are some recent developments of nanowire (or nanorod) LEDs by other growth 18 methods, e.g., MBE, showing significantly improved efficiency for device sizes in the sub-19 micron regime. These studies should be referenced to provide a more comprehensive overview 20 of the micro-LED fields.

21 *Our response*: Thank you for your valuable comments.

Referring to **Table R2**, Most micro-LED mesa structures report efficiency but not luminance. Additionally, in the mesa structure, the luminance area is small compared to the chip size due to the electrode portion for contact, resulting in low luminance. LEDs grown using MBE have the advantage of high brightness but have the disadvantage of low efficiency. This means controlling growing conditions is difficult. Therefore, the methods of separating chips from a wafer using ECE and transferring the separated chips using DEP can still be considered promising technologies.

29

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1	Response letter to the first reviewer
2	Reviewer #1 (Remarks to the Author):
3	
4	1. One minor comment is on lines 57-58. "Following the transfer of nanorod-LED display
5	58 technology from our team, the Samsung Display Corporation (SDC) has successfully
6	demonstrated". What do authors mean by "transfer". Is this a technology transfer. The
7	relevance of this sentence is not clear.
8	Our response: Thank you for your valuable comments.
9	Before) Page 2, Line 20
10	Following the transfer of nanorod-LED display technology from our team, the Samsung
11	Display Corporation (SDC) has successfully demonstrated the practical possibility of the DEP
12	process by fabricating a nanorod-LED-based electroluminescent (EL) device through the FSA-
13	DEP process and inkjet printing process.
14	
15	After) Page 2, line 34
16	Following the license and technical assistance of nanorod-LED display technology from our
17	team, the Samsung Display Corporation (SDC) has successfully demonstrated the practical
18	possibility of the DEP process by fabricating a nanorod-LED-based electroluminescent (EL)
19	device through the FSA-DEP process and inkjet printing process.
20	
21	Reviewer #2 (Remarks to the Author):
22	
23	The manuscript has been revised according to the reviewer's comments. I would like to
24	recommend this manuscript for publication in Nature Communications as is.