

High-Resolution, Intermixing-Free Quantum Dot Patterning Technology for Electroluminescent Display Applications

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Abstract

Patterning of the quantum dot (QD) layer with resolutions of 4,200 pixels per inch (ppi) is achieved by using the transfer printing technique which effectively prevents intermixing between QDs and the underlying hole transport layer. As a result, the peak external quantum efficiencies of QD light-emitting diodes (QLEDs) are significantly improved: 16.0% for red, 5.1% for green, and 5.8% for blue, outperforming those achieved by using spin-coated QDs. The result demonstrates that the QD transfer printing process is a promising fabrication process for highly efficient, high-resolution QLEDs.

Keywords Quantum dots (QDs); Quantum dot light emitting diode (QLED); High-definition displays; Transfer printing; Cd-free InP QDs; Intermixing-free QLEDs.

1. Introduction

Cadmium-free quantum dots (QDs), such as indium phosphide (InP) and zinc selenide telluride (ZnSeTe), have emerged as core materials for next-generation display technologies due to their outstanding properties, including high photoluminescence quantum yield (PLQY), narrow emission spectra, and environmentally friendly composition.[1-2] Thanks to these advantages, various products such as QD-OLEDs or QD-LCDs, utilizing QDs as color enhancement layer or color conversion layer (CCL), are commercialized. However, these photoluminescent QD-based displays still have several challenges including low brightness and high power consumption. Addressing these challenges necessitates the development of electroluminescent (EL) QD-based displays, QLEDs, where electrons and holes are directly injected into QDs via external bias to achieve high emission efficiency and accurate color representation.

To realize full-color QLEDs, precise patterning of red, green, and blue QD subpixels is essential. In general, photolithography and inkjet printing are widely utilized for patterning QD pixels [3]. Inkjet printing, which offers cost competitiveness and scalability for large-area production, has been successfully applied to pixelate QD CCLs for QD-OLED and QD-LCD. Samsung Display exhibited all-inkjet-printed 18.2-inch 202 ppi QLED panel during SID Display Week 2024 [4]. However, high-definition pixel formation over 1,000 ppi and intermixing issue between QDs and underneath hole transport layer (HTL) are still remained [5-7].

Herein, we present a dry transfer process for the high-definition, intermixing-free QLEDs. Cd-free QD films are picked up from donor substrate and released to the intaglio trench for the pixel formation. The remained pixelated QDs on the elastomeric stamp can be transferred onto the target substrate in film state, resulting in QD pixels with 4,200 ppi. As the patterned QD subpixels are transferred in a film state, there is a distinct interface between the QD layer with HTL, without intermixing. Moreover, the pressure

applied during the transfer printing process reduces the inter-QD distance and improves the surface morphology with better packing of QDs, thereby reducing the leakage current of the QLED and increasing brightness and efficiency compared to spin coated counterpart.

2. Results

Dry transfer printing enables high-resolution RGB pixelation of heavy-metal-free QDs – red and green InP QDs and blue ZnSeTe QDs. Fig. 1 shows the transfer printing procedure for QDs. QD solutions are spin-coated onto a silicon substrate treated with a self-assembled monolayer (SAM) to reduce adhesion between the substrate and the QD layer. Subsequently, elastomer stamp is used to detach the QD layer from the substrate. The detached QD layer is then patterned using intaglio trenches and transferred to the target substrate. Transfer printing utilizes differences in surface energy to achieve patterning, avoiding the use of photoresists, organic solvents, and high-energy light. This minimizes potential damage to the QDs during the patterning process, providing more gentle and effective transfer printing approach.

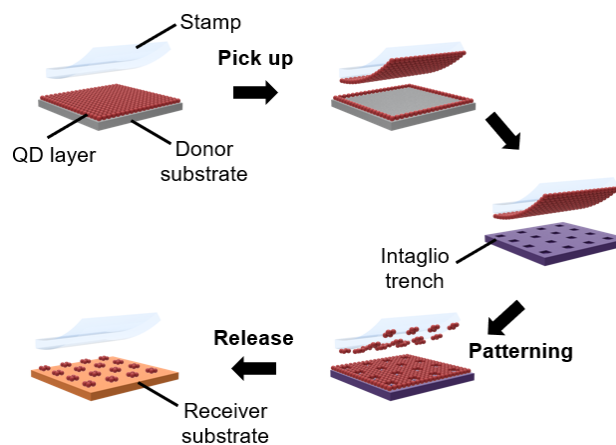


Figure 1. Schematic illustration of transfer printing process for the high-definition patterning of QDs

This transfer printing process exhibits a high patterning yield of 99.9%, enabling QD patterning from feature sizes as large as 200 μm down to ultra-fine patterns of 1.5 μm (Fig. 2). Using trenches with a 1.5 μm pattern size, ultra-high-resolution patterning of 4,200 ppi was achieved, maintaining a patterning yield of 99.9%. Optical microscopy images confirm the sharp and well-defined edges of the patterned QD layers (Fig. 3a), while fluorescence microscopy verifies the stable photoluminescence of the patterned QD layers (Fig. 3b).

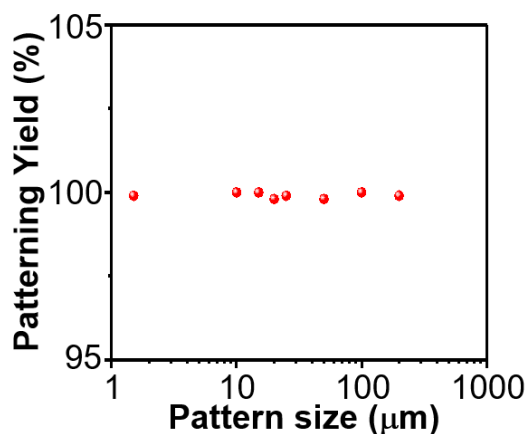


Figure 2. Patterning yield of transfer-printed QD patterns with various pattern size (from 1.5 to 200 μm)

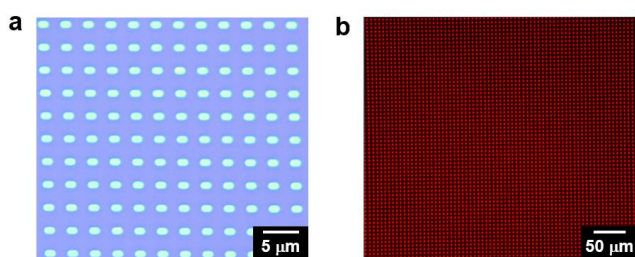


Figure 3. High resolution pixelated red QDs. (a) Optical microscope image and (b) fluorescence microscope image of 4,200 ppi QD patterns.

We incorporated transfer printing method into the device fabrication process to develop high-resolution, high-performance, and eco-friendly QLEDs. The device structure, designed for achieving electron-hole balance for high external quantum efficiency (EQE), consists of ITO/PEDOT:PSS/TFB/QDs/ZnMgO/Al (Fig. 4). The QD light-emitting layer was transfer-printed onto the spin-coated hole transport layer of TFB, poly[(9,9-dioctylfluorenyl-2,7-diyl)-co-(4,4'-(N-(4-s-butylphenyl)diphenylamine))].

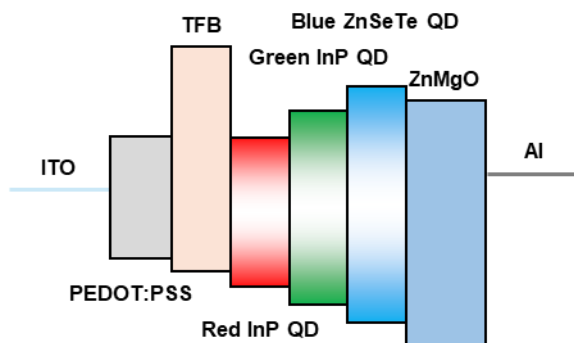


Figure 4. Energy diagram of transfer printed QLEDs.

The dry transfer process used for the fabrication of QLEDs prevents intermixing of nanoparticle layers during the stacking process, as confirmed by EDAX analysis (Fig. 5a). Magnesium (Mg), a component of the ZnMgO electron transport layer (ETL), was absent in the QD and underlying layers. Similarly, selenium (Se), which constitutes the shell of InP QDs, was absent in the

underlying hole transport and injection layers. This indicates that the transfer printing process minimizes layer-to-layer interference during device fabrication. Additionally, cross-sectional transmission electron microscopy (TEM) images confirmed the formation of smooth interfaces between the layers due to the vertical pressure applied during the transfer printing (Fig. 5b). Grazing-incidence small-angle X-ray scattering (GI-SAXS) analysis revealed a reduction in interparticle distance between QDs in the transfer-printed layer (0.0840 \AA^{-1} , corresponding to 7.47 nm) compared to the spin-coated layer (0.0825 \AA^{-1} , corresponding to 7.62 nm) (Fig. 5c). This reduced interparticle distance contributes to decreased leakage current, enabling the fabrication of QLEDs with higher EQE.

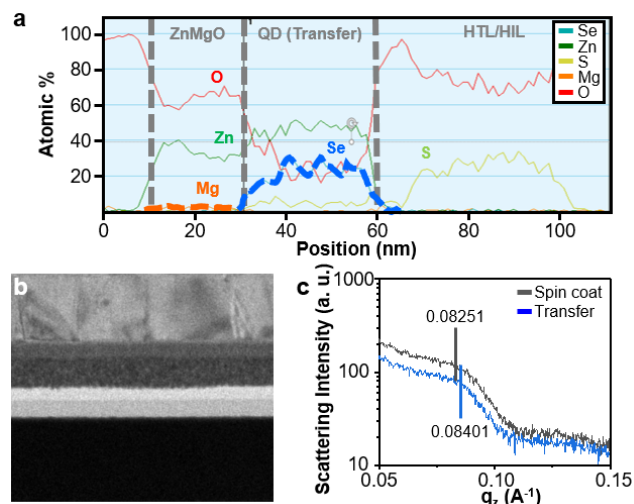


Figure 5. Intermixing-free QLEDs (a) Energy dispersive x-ray spectroscopy (EDAX) and (b) Transmission electron microscopy (TEM) of transfer-printed QLEDs. (c) Out-of-plane interparticle distance of QDs with spin-coated and transferred QDs.

Both transfer-printed and spin-coated QLEDs exhibited identical EL spectrum, with an emission peak at 634 nm and a full width at half maximum (FWHM) of 40 nm (Fig. 6). This confirms that the transfer printing process does not adversely affect the properties of QDs. Furthermore, the reduced interparticle distance and improved interface smoothness of the transfer-printed layers resulted in QLEDs with higher efficiency compared to their spin-coated counterparts.

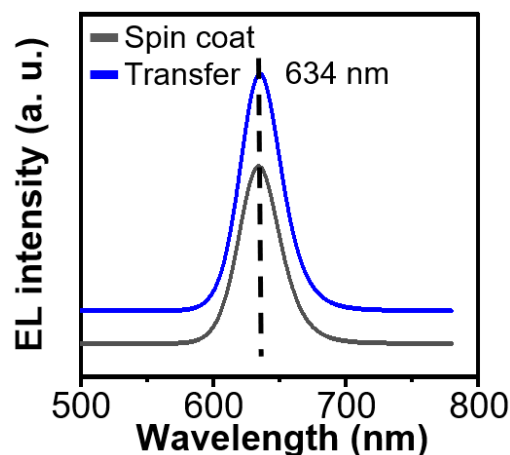


Figure 6. The EL spectra of QLEDs with spin-coated and transfer-printed QDs.

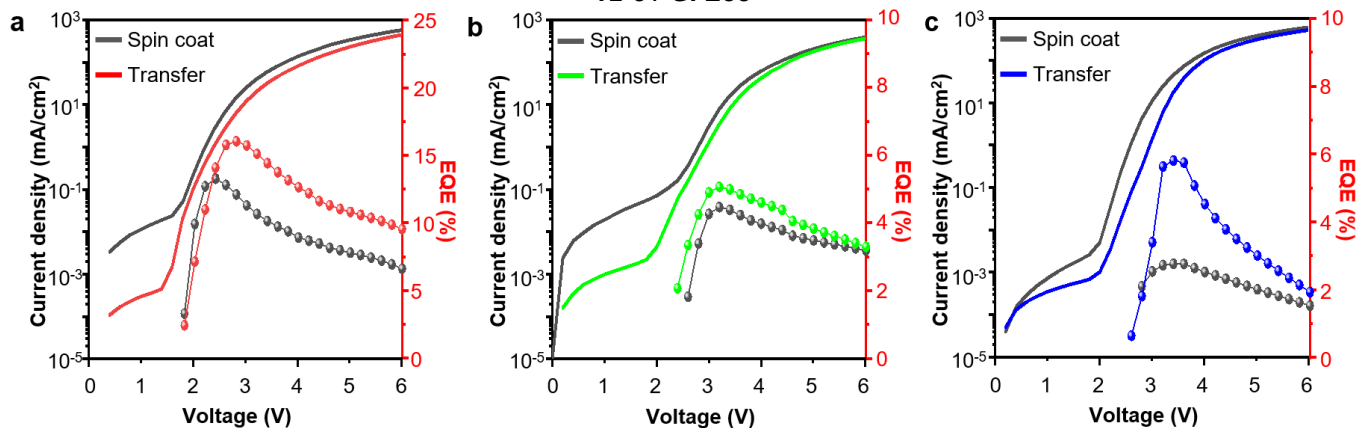


Figure 7. Current density and EQE of (a) red, (b) green, and (c) blue spin-coated and transfer-printed QLEDs.

Transfer-printed red, green, and blue QLEDs demonstrated significantly lower leakage currents below operating voltages compared to spin-coated devices. As a result, the EQE of the transfer-printed devices (red: 16.0%, green: 5.1%, blue: 5.8%) was substantially higher than that of the spin-coated devices (red: 13.2%, green: 4.5%, blue: 2.8%) (Fig. 7a, 7b, and 7c). This highlights the advantages of transfer printing in producing high-efficiency QLEDs with reduced power consumption and superior performance.

3. Conclusion

We have presented a universal quantum dot patterning technique for QLEDs. By using PDMS elastomer stamps in a dry transfer process, we successfully fabricated QLEDs with no intermixing problems at interface. This method can be widely applied not only to various types of QDs but also to general solution-processed nanoparticle thin films. Through this technique, we successfully achieved QLEDs with high performance, showing EQE superior to solution-processed devices, and demonstrated 4,200-PPI pixel patterning, proving the feasibility of ultra-high-resolution QLEDs. These results suggest that the transfer-printing-based pixel formation method has excellent applicability and is a promising approach for enabling the realization of ultra-high-resolution QLEDs.

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5. References

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