A Universal High-Resolution Patterning Technology for Quantum Dot Color Converters in Micro-LED Displays

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Abstract

We report a photolithography-based technology for patterning quantum dot color converters for micro-LED displays. A patterning resolution of ~1 µm is achieved. The method can be applied to any color converter materials. Integration of perovskite quantum dots and CdSe/ZnS quantum dots is demonstrated to show the versatility of the technology.

Author Keywords

Micro-LED; quantum dot; color converter; perovskite; photolithography; micro-patterning.

1. Objective and Background

Micro-LED technologies are being actively pursued by display R&D to deliver unprecedented optical quality, including resolution, brightness and contrast, that current technologies cannot achieve. The demand is propelled by the continuous drive for better screens ranging from smartphones/watches to large panel displays, and the emerging augmented reality (AR)/virtual reality (VR) market where current display technologies are inadequate for, in particular AR. Conventional micro-LED arrays require processing each color of LED, blue, green, and red, on individual semiconductor wafers. These LEDs must then be separated and assembled on a display substrate through the very manual and error-prone process of picking and placing pixel groups, resulting in a very high cost for the display. Micropatterned color converters can convert shorter-wavelength backlight, such as blue, to longer wavelengths such as green and red. Therefore, only one single-color backlight micro-LED array needs to be manufactured, which can drastically reduce the overall cost of micro-LED displays.

Typical color converters are solution-processed materials, mostly quantum dots (QDs), which are challenging to micro-pattern with high resolution because the chemicals used in industry-standard photolithography processes often damage the materials or greatly deteriorate their optical properties. Various QD photoresist have been developed in research labs and industry R&D to achieve direct patterning of QD color converters (1-3). For this approach, different photoresist chemistries need to be designed for different types of QDs. The picture gets more complex with UV absorption by the QDs during patterning, which can restrict chemistries and concentrations of QDs. It can be difficult to ensure full removal of ODs during development of the resist layer, thereby leaving residual converters and degrading overall display contrast. There is also significant waste of QDs through the process. Inkjet printing has been the industry's method for patterning color converters, with resolutions limited to TV display standards. Electrohydrodynamic (EHD) printing is being developed in response to the limited resolution of conventional inkjet printing. The new technique borrows many concepts; however, it introduces an electric field to further reduce the drop size emitted from the print head to micron or sub-micron resolution (4-6). There are challenges with EHD printing, including ink formulations that work with its mechanism (7).

We developed a photolithography-based process to micro-pattern

solution-processed metal-halide perovskite thin films (8). The process utilizes dry lift-off of an intermediary layer which protects the perovskite layer from solvents used in photolithography. Here, we report our progress on demonstrating the compatibility of this technology with micro-LED display manufacturing. This includes micro-patterning perovskite QDs on the substrate of a blue GaN LED array, demonstration of a wafer-scale process result, and improving the patterning resolution to ~1 μm. Finally, since the industry has not reached consensus on the ultimate QD for micro-LEDs, it's desirable that a patterning technology can be applied universally to any type of QDs. We demonstrate this by integrating green perovskite QDs and red CdSe QDs on the same substrate with high resolution. The process can be extended to other types of QDs such as InP QDs and in general any types of solution-processed materials.

2. Results

Micro-Patterning Process: Our micro-patterning process for color converter materials relies on the limited adhesion of parylene to various substrates. The type of parylene employed in this process is parylene-C as it has a very low permeability to solvents. Figure 1 depicts the principle of this method. First, a ~2.5 µm-thick parylene film is deposited onto a clean substrate by room-temperature chemical vapor deposition (CVD). Subsequently, standard photolithography is used to fabricate the desired trenches in the photoresist layer. The patterned trenches are then transferred to the parylene film by reactive ion etching (RIE). The remaining photoresist is stripped by O₂ plasma or acetone. Then, the QDs are deposited on the patterned parylene film. (Figure 1(a)) Finally, the underlying parylene film can be peeled off mechanically, and this dry lift-off is a one-step process for the entire substrate. (Figure 1(b)) The peeled-off QDs can be recycled, which can reduce the material cost and is an added benefit of this process. This process can be applied to all types of solution-processed materials.

QD Patterning on a Blue LED Array: To validate that our patterning process can be applied to a micro-LED backplane, we acquired commercial GaN/AlGaN epitaxial wafers on sapphire substrates and fabricated large-size blue LED arrays. The epitaxial wafer structure consists of active layers defined by 10 pairs of InGaN (3.5 nm) / GaN (5 nm) multiple quantum wells, sandwiched between n-type GaN/AlGaN layers at the bottom and p-type GaN/AlGaN at the top. Blue LEDs were fabricated through etching the wafer down to the heavily doped n-type GaN layer, followed by oxide deposition to passivate the sidewalls. Contact windows through the oxide layer were opened through dry etching. Finally, electrical contacts to the top p-type GaN/AlGaN layer and the bottom GaN layer were completed through metal deposition. Figure 2(a) shows a fabricated 4 × 4 blue LED array on a chip area of 1 cm × 1 cm. Figure 2(b) shows an optical image of one of the LEDs being turned on under a forward bias.

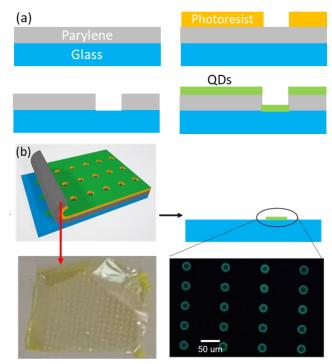


Figure 1. A universal photolithography-based micropatterning process for color converters. (a) Photolithography is performed on an intermediary parylene layer, followed by deposition of QDs. (b) Lifting-off the parylene layer completes patterning of QDs.

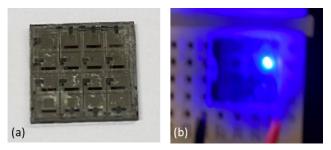


Figure 2. Blue backlight LED array. (a) A fabricated 4 x 4 InGaN/GaN blue LED array on a sapphire substrate. (b)

One of the blue LEDs is turned on.

Green perovskite QDs synthesized by ourselves were used for micro-patterning experiments on the blue LED array chip. While there have been many reports on synthesis methods for perovskite QDs, most of the perovskite QDs with high photoluminescence quantum yield (PLQY) were synthesized through a hot-injection method which requires precisely controlled elevated temperatures and an inert gas environment, therefore may not be suitable for mass manufacturing. Instead, we adopted a room-temperature triple-ligand surface engineering strategy that does not require inert gas to synthesize green-emitting CsPbBr₃ QDs (9). The synthesized perovskite QDs achieved >94% PLQY and a narrow emission linewidth of ~20 nm.

We then applied the process depicted in Figure 1, with the addition of an Al black matrix layer prior to parylene-C deposition (see next paragraph for further explanation), and fabricated patterned green perovskite QD color converters on the

sapphire substrate of the blue LEDs. Figure 3(a) shows a photograph of perovskite QDs patterned on the sapphire substrate of a blue LED array chip, and Figure 3(b) shows the fluorescence of the perovskite QDs under UV excitation. Figure 3(c) shows the close-up optical microscope image of the patterned parylene-C layer, and Figure 3(d) shows the patterned perovskite QDs after depositing the QDs and lifting off the parylene-C layer. The results show that perovskite QDs were patterned on the blue LED substrate with good uniformity using parylene-C-mediated dry lift-off photolithography.

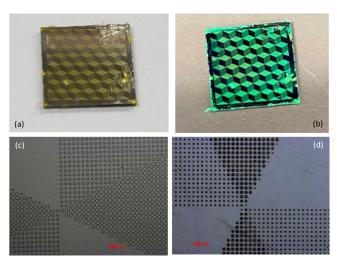


Figure 3. Integrating perovskite QD color converters with a blue LED chip. Optical images of (a) perovskite QDs patterned on the sapphire substrate of the blue LED array chip, and (b) fluorescence of the green-emitting QDs under UV excitation. Optical microscope images of (c) the patterned parylene-C/Al layer, and (d) patterned perovskite QDs after depositing the QDs and lifting off the parylene-C layer. The metallic gray area in between QDs is the patterned Al layer for blocking blue backlight.

The LED chip covered with patterned perovskite QDs was placed under a fluorescence microscope, and one of the blue backlight LEDs was turned on. Figure 4 shows the fluorescence microscope image. Part of the image is out of focus because the blue LED array chip with wires connected to it was slightly tilted under the microscope. In order to block the blue backlight coming through the area without QD coverage, an Al layer was deposited on the sapphire substrate prior to parylene-C deposition. This Al layer was patterned simultaneously with the parylene-C layer and served as a black-matrix layer in between QDs. It appears as the metallic gray region in Figure 3(d).

Wafer-Scale Process with High Resolution: In order to assert standard semiconductor-fabrication compatibility, wafer-level processing is essential. Through a whole-wafer process, shown by Figure 5(a), we patterned green perovskite QDs with various feature sizes and pitches on a glass wafer. Processing on this size scale is more applicable to commercial level processing and validates that the method is easily scaled. Figure 5(b) and Figure 5(c) show the fluorescence microscope images of part of the patterned perovskite QDs. The smallest feature size was designed to be 1 μ m. Due to slight over-development during photolithography, the final feature sizes were enlarged, and the highest resolution achieved was ~3 μ m, seen in Figure 5(c). Figure 5(d) shows the PL spectrum of green emission from the

perovskite QDs. The narrow linewidth was not affected by the fabrication process.



Figure 4. Fluorescence microscope image of patterned perovskite QDs after the blue backlight LED is turned on.

After further process refinement and experimentation, we were able to improve the resolution of patterned QDs. We also applied the process to CdSe/ZnS core-shell QDs to verify that this method is agnostic to the material to be patterned. Figure 6 (a) and 6(b) show the optical microscope and fluorescence microscope images of patterned QDs. A patterning resolution of ~1 μm was achieved. The PL spectrum of the red-emitting CdSe/ZnS QDs is shown in Figure 5(e). Figure 6(c) shows the atomic-force microscope (AFM) image of a QD color converter pattern with ~ 2×3 μm^2 feature size. A cross-sectional height measurement scanned across the long axis of the pattern is shown in Figure 6(d). The thickness of the QD color converter pattern is close to 200 nm for the inner part of the pattern.

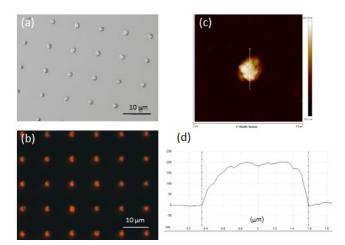


Figure 6. (a) Optical microscope and (b) fluorescence microscope image of patterned CdSe/ZnS core-shell QDs. (c) AFM image of a QD color converter pattern. (d) Cross-sectional height characterization of patterned QD color converters, showing a film thickness close to 200 nm for the inner part of the pattern.

Integration of Different QD Types: To achieve multi-color patterning and to demonstrate the universal patterning capability of this method, we integrated green perovskite QDs and red CdSe/ZnS QDs on the same substrate. Figure 7(a) shows fluorescence microscope image of part of a pattern formed by both colors, and Figure 7(b) shows a zoom-in fluorescence image. This part of the masks contains larger feature sizes, and the images show pattern size of ~5 µm.

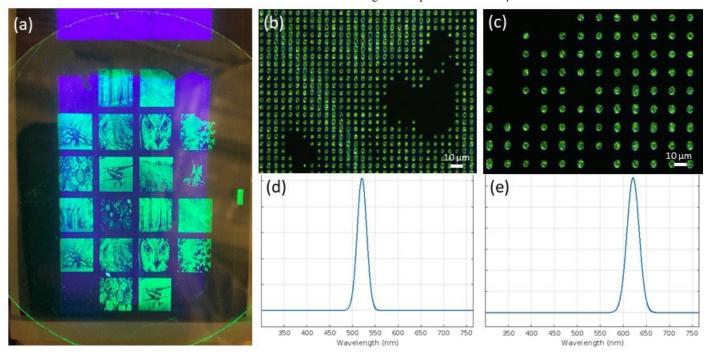
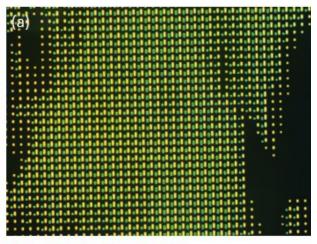


Figure 5. Wafer-scale patterning of QD color converters. (a) A full-wafer process is employed for patterning green perovskite QDs. (b) and (c) Fluorescence microscope images of parts of the patterns. The smallest feature size ~3 µm was achieved. (d) PL spectrum of the green perovskite QD emission. (e) PL spectrum of the red CdSe/ZnS core-shell QDs from Figure 6(b).



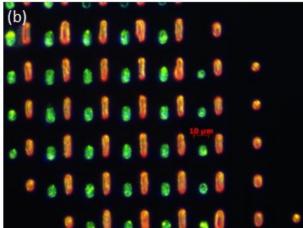


Figure 7. Fluorescence microscope images of multicolor patterning through integrating green perovskite QDs and red CdSe/ZnS QDs. (a) A larger sub-region of a pattern. (b) Zoom-in of the pattern. A feature size of \sim 5 μ m was achieved

3. Conclusion

We demonstrated a universal micro-patterning method for QD color converters. Utilizing a dry liftoff process through an intermediary parylene-C layer, the optical quality of the QDs is preserved during the fabrication process, and the method can be applied broadly to different types of QDs and all solutionprocessed materials in general. We first verified that the process can be performed on the substrate of a fabricated blue LED array chip, thus suitable for integrating the QD color converters with backlight micro-LEDs. Through a wafer-scale process, we achieved a patterning resolution of 1 µm. We then integrated green perovskite QDs with red CdSe/ZnS QDs on the same substrate. To our best knowledge, this is the first time integration of micro-patterned color converters consisting of different types of QDs was achieved. The results demonstrate the versatility of our micro-patterning method, and present it as a strong candidate approach for enabling high-resolution micro-LED displays through utilizing color converters.

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