

Colloidal Quantum Dot Enhanced Color Conversion Layer for Micro LEDs

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SUMMARY In this paper, we introduce our latest progress in the colloidal quantum dot enhanced color conversion layer for micro LEDs. Different methods of how to deploy colloidal quantum dots can be discussed and reviewed. The necessity of the using color conversion layer can be seen and color conversion efficiency of such layer can be calculated from the measured spectrum. A sub-pixel size of 5 micron of colloidal quantum dot pattern can be demonstrated in array format.

key words: *colloidal quantum dots, micro LEDs, mini LEDs, heterogeneous integration*

1. Introduction

In the past decade, we saw a rising trend of the information displays which provide not only the entertainment but also functionality not dreamt before. A monitor can be applied in TV sets, computers, and even kiosks in train station. A variety of technologies have been brought to this field, including plasma display panel, liquid crystal, and organic light emitting devices etc. Among these technologies, micro/mini light emitting diodes (micro/mini LEDs) based on semiconductors have attracted much attention in recent years. Because of their potentially mass-production capability and high performances, they are regarded as the product for the future generation [1]–[3]. The use of self-emissive and independently addressable pixels in the display can provide a highly dynamic and vivid-colored viewing experience and the high contrast ratio can surpass most of the existing technologies. For the development of advanced displays, in addition to highly saturated color, a fine pixel or high resolution is very much desirable. In the micro/mini LED domain, it becomes clear that the size of the chip can be directly related to the pixel per inch (PPI) index, a usual key number for display resolution. Usually we will treat chip size smaller than 100 micron as the micro-LEDs and otherwise as the mini-LEDs. To accomplish a full-color scheme, two different methods can be adapted: mass-transfer and color conversion [4], [5]. In mass-transfer, LED chips with different colors can be mechanically removed from their original

substrate and moved into a common substrate. Such heterogeneous process can reduce the requirements on the epitaxial growth and optimize the chip design for each color. However, limitations exist for mass-transfer. One of the key concerns rises from the difficulties for smaller chip transfer. Although it is possible to fabricate micro LED chips with sizes smaller than 5 μm [6], [7] these days, it is difficult to mass-transfer tiny devices onto different substrates, not to mention the high precision that requires in this job. In the past, 8 μm by 15 μm devices have been demonstrated with an estimated array size of 100 by 100 [8]. We demonstrated full-color micro LEDs with sizes of several tens of microns transferred onto the printed circuit board [4], [5] and a functional full-color display (30 cm by 30 cm) was shown in CES 2020. One of the solutions that could possibly alleviate this situation is to use color-conversion layer to convert high energy photons (blue) into lower energy ones (red and green). In this way, the micro LED arrays can be maintained as single-color and monolithic fabrication. No mass-transfer will be needed in this scheme. Several groups (including ITRI) have also demonstrated such technologies [9]–[11]. As for the material for color conversion, a strong illuminative capability is necessary. Several candidates have been brought up for this purpose: fluorescent dye molecules, conjugated polymer, and colloidal quantum dots (CQDs) [12]–[19]. Among them, colloidal quantum dot has been in research focus for decades. They are synthesized by chemical reactions and usually made of semiconductor in a nano-meter scale. The extraordinary capability in photon emission makes them very promising for the future generation of display devices. In the past, most of the researches focused on mixing them with individual LED chips for solid-state lighting application [20]. We saw an increasing demand to use them for display purposes these days [16], [21]–[23]. However, patterning these nano-sized particles remains a difficult task to be conquered. In this study, we will demonstrate our latest results on the color conversion layer for micro LEDs. As small as 5-micron QD pattern can be fabricated for the micro display application and shall be useful for the next generation of high definition display technology.

2. Design and Theory

In this section, we will discuss several basic ideas and theories about these color conversion layer designs. It is

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essential to have a feasible structure and optimize it to meet the display requirement. Due to the fragile nature of the color conversion material, the substrate becomes an important part. The method to characterize these conversion layer will also be reviewed.

2.1 Device Design for Color-Converted Display

The design of the color conversion layer usually has two concerns: conversion efficiency and process integration. In the conversion efficiency concern, it depends on the quantum dot material we choose. At present, most of the commercially available products are cadmium based (i.e. II-VI group compound semiconductors) metal quantum dots. They are composed of CdS, CdSe, or CdTe, etc., and have a wide coverage of emission colors spanning from blue to infrared. As for the process integration, the main focus is to facilitate the assembly processes and to enhance the effectiveness of the conversion layer, and photon extraction. In a basic concept, as shown in Fig. 1 (a), a background white light source, which can be underneath or channeled by a light guide from the side of panel, properly filtered by the color conversion layer on the surface and three colors can be obtained. This basic design can be seen in current LCD technology by installing color filters as the conversion layers and it is already used for production. However, in the current LCD scheme, the surface color filter can reduce the radiance greatly, and thus it is not preferable for energy-saving purposes. By introduction of quantum dot, the color filter can be more effective and the resultant RGB output can be enhanced. Quantum dots can also be applied to obtain a better

white light source before the filtering due to their highly saturated colors [16].

Meanwhile, the color conversion layer can combine with extra optical layer to increase the utilization of pumping photons and QD outputs. This idea can be visualized in Fig. 1 (b). The optical layer is usually made of reflective coating such as Distributed Bragg Reflector (DBR) to bounce back the high energy photons while the QD photons can escape. The glass substrate can mechanically support the conversion layer and provide a path for blue or UV photons from the bottom emitters. The third design, shown in Fig. 1 (c), flips the color conversion layer to face the emitter array. This design facilitates the photonic pumping and shortens the optical path of blue photons before they hit the color conversion layer. A traditional or improved color filter layer can be placed beforehand in this design to reduce the residual blue lights after the color conversion. In the past, many groups demonstrated similar designs like Figs. 1 (b) and (c) [10], [24], and a variety of designs can be found in [25]. Some adjustments could be necessary because of the differences in details of the devices.

2.2 Theory

After putting all components together, it is essential for us to properly characterize the effectiveness of the color conversion layer. Several related calculation procedures are required to figure out the best design of the color conversion layer. One of the important concerns is the proper thickness of the quantum dot mixture. The total thickness of the QD layer can affect the light conversion efficiency and also the color quality of the display and thus attention needs to be paid. Whether the QD is directly dispensed or pre-mixed with polymer and resin for other purpose, the strong absorption of quantum dots and emission efficiency have to be considered. Here a simple 1D model is set in Fig. 2 to provide the preliminary evaluation. In a QD-filled texture, assumed the thickness of d , the incoming blue photons are from left hand side, and we would expect certain decline of the blue photon intensity in the QD layer due to absorption (α_{blue}). At position x , we have an infinitesimal small QD region of dx which absorbs blue photons and re-emits red photons with the percentage of $A\%$. Meanwhile, the emitted red-photons also slightly get absorbed by quantum dots themselves, which corresponds to an absorption coefficient of α_{red} . The combination of these two effects can be

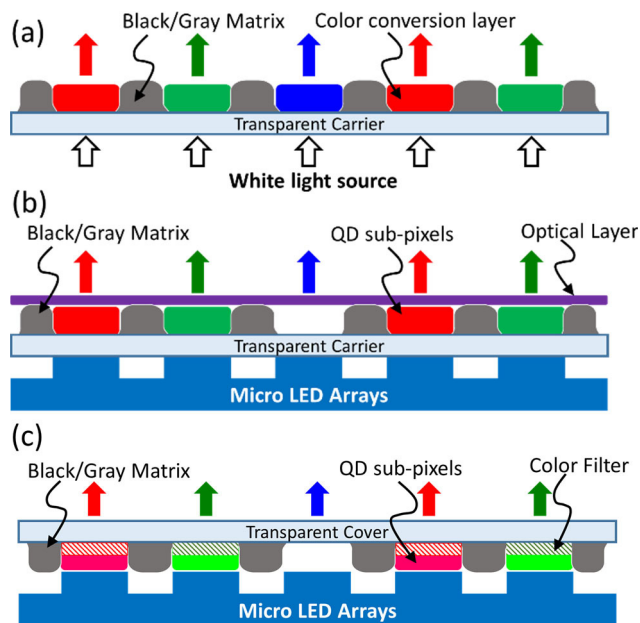


Fig. 1 (a) A basic structure of a color-converted device having color conversion layers, black matrices and background white light sources. (b) A design with glass substrate between QDs and micro LEDs. (c) A design with QDs facing directly to micro LEDs.

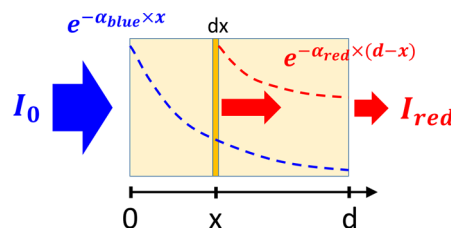


Fig. 2 The schematic diagram of a 1D QD layer absorption and emission model.

considered in the expression as:

$$I_{red} = \int_0^d \frac{I_0 \nu_{red}}{2\nu_{blue}} A\% \times \alpha_{blue} e^{-\alpha_{blue}x} \left(e^{-\alpha_{red}(d-x)} \right) dx, \quad (1)$$

where I_{red} is the intensity of red photons, ν_{red} and ν_{blue} are the frequencies of red and blue photons, respectively.

Following Eq. (1), we are able to find out the red photon intensity under different optical density (OD) values of the quantum dot layer at blue wavelength. The OD value of an absorptive layer can be defined as the inverse of the transmitted power ratio:

$$OD = \log_{10} \left(\frac{I_0}{I_t} \right), \quad (2)$$

where I_0 and I_t are the incident and transmitted power, respectively. As shown in Fig. 3, a peak value can be found at different OD values. The higher OD value is, the stronger blue absorption in the QD layer will be. In the calculation, the red photon's absorption coefficient was set as 38.4 cm^{-1} in the calculation of Eq. (1), and the conversion rate $A\%$ was set at 50%. In Fig. 3, a trend on the peak thickness, which represents the highest red photons at certain OD value, can be observed. As the blue absorption in QD layer gets higher, the peak intensity of the red photons will get higher and the optimal thickness will become thinner as well. Thus it is not always beneficial to increase the thickness of color conversion layer in order to increase the QD emission. For each different QD concentration of the color conversion layer, it is important to find out their best thickness to balance the absorption and re-emission of the specific color of photons.

Another important parameter is the color conversion efficiency (CCE) which compares before and after conversion layer was placed. The difference of the spectral intensity at blue (or UV) and QD-emission bands can be used to figure out the overall conversion efficiency [17], [26]:

$$CCE = \frac{\int_{\text{green or red}} \left(\frac{\lambda}{hc} \right) \times [I_{em}^{QD}(\lambda) - I_{em}^{ref}(\lambda)] d\lambda}{\int_{UV} \left(\frac{\lambda}{hc} \right) \times [I_{ex}^{ref}(\lambda) - I_{ex}^{QD}(\lambda)] d\lambda}, \quad (3)$$

where I_{ex}^{QD} and I_{ex}^{ref} are the integrated intensities of the

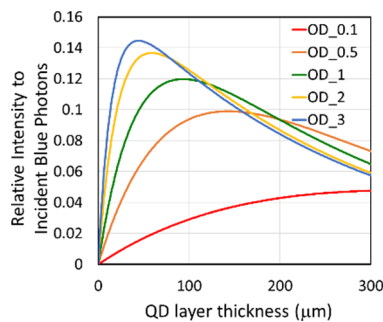


Fig. 3 The emitted red photons versus QD layer thickness under different OD values of the layer.

UV LED excitation source with and without the CQD layers, and I_{em}^{QD} and I_{em}^{ref} are the intensities of the visible band (but excluding the UV band) with and without CQD layers, respectively. The high CCE value can be viewed as an indicator of highly utilized pumping photons in the system, while the low CCE number can mean the loss of photons in the structure is high. A traditional PDMS (polydimethyl-siloxane)/QD packaged LED can usually reach 30% or 40% [26].

3. Formation of QD Arrays for Color Conversion Layer

In this section, we will discuss various types of technologies that could help to form a large QD array on the substrate. It is essential that the full-color pixels in a display can be fabricated in a fast and efficient fashion. Different methods have been developed in the past to facilitate this process and some of them might be able to go through the commercialization and become prevailing in the future.

3.1 Direct QD Spray

One of the most popular methods is to cast CQD solution directly onto the substrate. The quantum dots are usually stored in solvent or water, and both of the media are good to drop-cast QDs directly onto the target location. However, due to the requirement of patterning, a simple drop-and-spin method will not suffice. A sequential and precise dispense of CQD materials will be needed to achieve this goal.

To precisely spray a little amount of CQDs into the location of each pixel is an intuitive way to pattern the QDs. Especially for the low viscosity solution such as solvents, we have a lot of room to adjust the possible spray area for the patterning purpose. In the past, we have demonstrated a fine pitched pattern of QDs in different colors. Such as those in [27], a $35 \mu\text{m}$ by $35 \mu\text{m}$ squared shape of QD pattern was successfully cast onto the surface of a micro LED with similar size. Its process flow can be seen in Fig. 4. Further control in terms of air flow and viscosity of the machine can provide improved resolution. In 2019 and 2020, we showed the feasibility of the CQD arrays whose sub-pixel sizes are 10 and 5 microns [28], [29]. With proper isolation

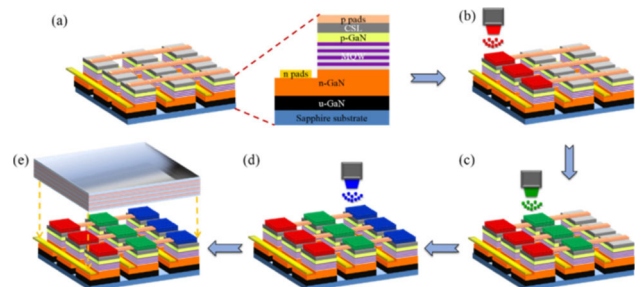


Fig. 4 A generic procedure of direct spray of QD patterns on top of micro LED devices, and the addition of the optical layer. Reprinted with permission from [27]. © The Optical Society.

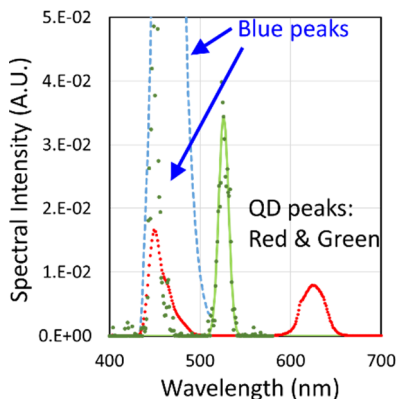


Fig. 5 The spectra of pumping blue light and emitted QD light on 30 μm QD pixels of different colors. Residual blue intensity can be seen in different strengths.

of the QD arrays from the environment, a continuous operation close to 100 hours can be demonstrated [29]. From the comparison in the spectra between blue pump source and the quantum dot sub-pixel emission in Fig. 5, we are able to calculate the CCE by the formula mentioned in Sect. 2. The calculated results showed that the best conversion efficiencies are 8.45% for 30 μm and 1.94% for 10 μm patterns. The drop-cast method is simple but the overall CQD thickness can be limited to micron-scale because of the inherent tiny volume of CQDs after solvent evaporation, and the self-aggregation concern will rise once the amount of nanoparticles is high. Thus it is difficult to construct a very thick and effective CQD layer through direct drop-cast method.

3.2 QD in Photo-Sensitive Resin

Although the method of direct spray is simple, the overall throughput is questionable when we need to work on a large number of elements in the QD array. This can happen frequently when the resolution of the micro display rises. To become more efficient on QD patterning, using photo-sensitive resin to mix with QD is necessary. These resins, sometimes called quantum-dot photoresist (QDPR), can be exposed and patterned by a UV source in one shot with large area of patterns accomplished. It is the ultimate way for the semiconductor processing lab to cope with quantum dot materials. However, difficulties remains as several factors hindering its prevailing. The first one is the uniformity of dispersed QD in the resin. The QD distribution in solution such as toluene and hexane is very good but it will take extra efforts to have a good dispersed QD in more viscous liquid like photo-resist.

Second is the resolution of the quantum dot pattern (or sub-pixel size). Because of the strong absorbance in quantum dots, the regular UV exposure which can define the pattern size of the photo-resist become less effective and thus the resolution of the pattern can be greatly affected. This situation will become worse when the aspect ratio (thickness to the side length) of the QDPR pattern increases. From the previous section and Fig. 3, although we don't need infinite

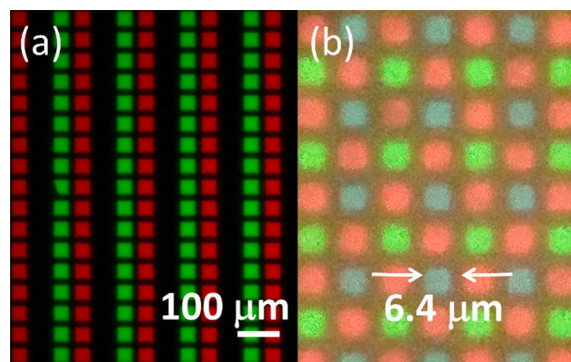


Fig. 6 The QDPR patterns of (a) 30 micron under FLOM (fluorescent optical microscope), and (b) 5 microns under uniform blue pumping lights.

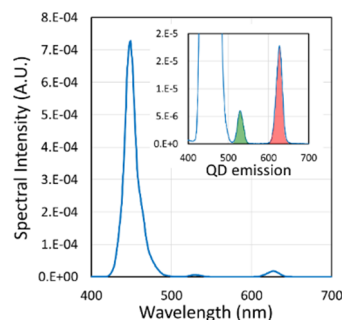


Fig. 7 The emission spectrum of a full-color 5-micron QD enabled pixel array. The inset is the same spectrum but with magnified red and green scales.

thickness of QDPR, certain thickness of several tens of microns will be good to provide enough converted photons in the micro-display. But when the feature size of each sub-pixel shrinks, the aspect ratio, which is the ratio of thickness to the side length, increases. To overcome this difficulty, many research groups have devoted their efforts and great results can be seen. Previously research group demonstrated multiple exposure is feasible to implement dual color conversion on the same substrate and a 20-micron pixel can be realized [25]. Researchers also showed the 10 micron patterned quantum dot arrays [30] fabricated by photolithography and the glass substrate was used as the carrier. In our lab, we also adapt glass substrate to carry the finished QDPR arrays. As shown in Fig. 6 (a), a complete multiple color QDPR array with sub-pixel size of 30 micron can be demonstrated. The thickness of this pattern is 6 μm . Further reduction in size will lead to more reduction of the QD emission due to less quantity of emissive particles and shorter optical path for pumping blue photons through QDPR regions. In Fig. 6 (b), a 1.8-micron thick QDPR array for red and green color is demonstrated. The pitch between QD patterns is 6.4 μm and the actual size of the QD is around 5 μm . The measured optical spectra of a 5 micron sub-pixel size QDPR pattern can be seen in Fig. 7. From these data, we can deduct that the integrated intensity ratio among blue, green and red is 31.56 : 0.27 : 1. However, due to the color filter layer pre-installed in the structure, it is difficult to

estimate the CCE values of red and green QDs since the residual blue photons are filtered by this color filter layer. Currently the conversion efficiency of a 5-micron QD pattern is still small compared to large devices [26], but we would expect much better improvement in the future by the material and also the new optical design.

3.3 Nano-Imprint QD Patterns

Another important method to implement CQD arrays is to use nano-imprint technology. As shown in previous papers of other groups [31], a mold with pre-determined patterns can be fabricated by semiconductor processes. Then by distributing QD layer evenly on a donor substrate, the nano-imprint mold can pick up these QDs slowly and transfer these nanoparticles to another location or wafer. This action, like “stamping”, has made the QD pattern as small as 6 μm in the past and also multiple and aligned operation to produce flexible electrically driven QD LEDs [31]. By this method, researchers also demonstrated a 4-inch full color QD display on a TFT backplane [32].

3.4 The Optical Layer for Enhancement

Another usual components in the color converted design is the optical layer which can help to increase (or enhance) the light extraction of the quantum dots. In many cases, the thickness of the color conversion layer is little compared to generic optical coating. The spray of quantum dots can usually coat a layer of 1 micron or less. With multiple times of spray, we were able to achieve several microns [27]. The thickness of color conversion layer can be thicker in the photo-sensitive resin case, but it still ranges in 10 microns or less. Without other optical scheme, the blue (or UV) photons can penetrate this thin film very easily and remain strong in the viewers’ end. To increase the usage of pumping photons, we need to “recycle” these photons by deflecting the unused ones back to the QD patterns. One of the best choices is the distributed Bragg reflector (DBR) structure. It can be formed by multiple pairs of dielectric materials with two different refractive indices. If their layer thickness is designed correctly (to the quarter wavelength of interests), the combination of the reflected EM waves to become strong and most of the traveling EM waves (or photons) can return to the QD pattern. From the past experience [18], it is possible to increase the QD emission by two times if the DBR is correctly installed. In many cases, the central wavelength of DBR is targeted at blue or UV band, and high reflection needs to be avoided in the red and green bands.

4. Conclusion

In this paper, we introduce our latest progress in the color conversion layer made of colloidal quantum dots. These nanocrystals can render very high quality colors while the technologies for their patterning and integration need to be investigated. Two major methods were implemented:

direct spray of QD and QD mixed with photo-sensitive resin. Both of them showed satisfied results in terms of miniaturized patterning, and a 5-micron multiple color QD array can be demonstrated by these two methods. Although the CCE is not very high at current stage, it is possible to enhance their performances via optical reflection or improved material quality. We hope the implementation of color conversion layer onto micro LEDs can be a great thrust to their eventual commercialization, and the future generation micro-display can be fully developed.

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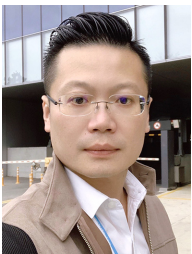
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