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Quantum Dot Display Patterning Technology for Ultra-High Pixel Density

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Abstract. Colloidal quantum dot light-emitting diodes (QD-LEDs) stand out as a premier technology for next-generation displays, particularly in smartphones and near-eye AR/VR systems. This enthusiasm is largely driven by their remarkable optical traits, such as near-perfect color purity, a massive color gamut, and high photoluminescence quantum yield. Yet, moving ultra-high-resolution QD-LEDs from the lab to mass production hits a severe roadblock. In this paper, six leading strategies for QD patterning—photolithography, inkjet printing, laser processing, transfer printing, self-assembly, and optical microcavities are review. Then, it points out that each patterning technology has its own advantages and limitations. The future development direction may focus on the collaborative integration of multiple patterning technologies, such as combining the high-precision structure definition ability of lithography with the material deposition advantages of printing and transfer technologies, to achieve the large-scale manufacturing of ultra-high PPI quantum dot micro-display devices. By mapping out these critical bottlenecks, this paper offers a strategic roadmap for developing the high-density patterning solutions necessary to finally realize the commercial promise of advanced QD-LEDs.

Keywords: quantum dots, QLED, patterning technology, micro-display, ultra-high pixel density

1. Introduction

1.1. Background

Since the first time Quantum Dot (QD) light-emitting diodes were synthesized in a single-dispersed manner through chemical methods in 1994, these nanocrystals have attracted widespread attention due to their excellent photoelectric properties. In the development of display technology, quantum dots initially entered the market as conversion light-emitting materials, such as Quantum Dot Liquid Crystal Display (QD-LCD), absorbing blue light from the backlight to convert it into high-purity red and green light, significantly enhancing the color gamut of liquid crystal displays. However, the goal of the display industry is to achieve self-luminous display of quantum dot electroluminescent devices (QLED). Compared with mature organic light-emitting diodes (OLED), QLED has potential significant advanages in terms of thermal stability, material lifespan, and color purity [1-3].

Currently, the display industry is entering the "micro-display" era. With the rise of devices such as augmented reality (AR) and virtual reality (VR), users' requirements for display resolution have jumped from the traditional hundreds of PPI (Pixels Per Inch) to above 1000 PPI, and in near-eye display, it is necessary to exceed 3000 PPI [4]. The requirement for ultra-high resolution means that the pixel size in the display panel must be reduced to the level of several micrometers, while maintaining extremely high array accuracy and fidelity. Compared with traditional OLED materials or micro-LEDs, quantum dots do not show significant degradation in core photoelectric performance at small sizes, which gives them a significant advantage in the field of ultra-high resolution micro-display.

1.2. Recent research progress

The key foundation for achieving full-colorization and commercialization of QLEDs lies in the ultra-high pixel density patterning technology of quantum dots. This demand for ultra-high pixel density has posed significant physical challenges for the traditional vacuum evaporation combined with fine metal mask (FMM) process when dealing with quantum dot displays. Issues such as difficulty in processing solution-based materials and high costs have emerged. Poor alignment accuracy and severe waste of evaporation materials.

To overcome this technical bottleneck, researchers worldwide have developed various solution-based or direct physical transfer patterning technologies of quantum dots in the past few years. Lithography technology, leveraging its accumulated ultra-high resolution genes in semiconductor processes, has achieved micro-micron or even sub-micron pixel arrays through the introduction of photosensitive ligands and non-destructive developing schemes [3, 5]. Meanwhile, transfer printing technology, through composite stamp design, has successfully achieved high-density array transfer with pixel fidelity up to 94%, maintaining excellent device efficiency at 2565 PPI [6, 7]. Additionally, electrohydrodynamic printing (EHD Printing) as an advanced version of inkjet printing has broken through the physical limitations of piezoelectric nozzles and achieved resolutions above 500 PPI [8]; while the emerging technology of femtosecond laser-induced forward transfer (FsLIFT) demonstrates the possibility of rapidly preparing arbitrary shape full-color arrays without masks [9]. Moreover, self-assembly technology utilizes thermodynamic driving to achieve spontaneous ordered arrangement of quantum dots, avoiding chemical damage to luminescence properties caused by etching reagents [10]. Meanwhile, optical microcavity technology has opened up a new path of "indirect patterning", by adjusting the thickness of the non-luminescent layer of the lithography, achieving ultra-high resolution full-color extraction while significantly amplifying the device's light output efficiency without directly damaging the quantum dots [11, 12]. The cross-development of these technologies has laid the technical foundation for achieving commercialization and high-performance ultra-high-density displays.

1.3. Research motivation

Although there have been continuous advancements in ultra-high pixel density patterning technologies, it remains a significant challenge to maintain the photoelectric properties of quantum dots while ensuring extremely small pixel sizes. Issues such as solvent erosion during the patterning process also pose constraints on the commercialization of high-density QLEDs.

This paper aims to systematically and comprehensively review the mainstream technical solutions for quantum dot patterning with ultra-high pixel density, analyze the physical and chemical mechanisms of each process during miniaturization, and explore how to collaboratively optimize pattern quality and device performance. The overall discussion framework for future development is as follows: Chapter 2 presents the basic composition and working principle of quantum dot microdisplay. Chapter 3 reviews the research progress of quantum dot patterning technologies, including several mainstream process schemes: lithography technology, transfer printing technology, inkjet printing technology, laser processing technology, self-assembly technology, and optical microcavity technology. Chapter 4 discusses the limitations of current technical solutions and looks forward to future technological evolution trends and application scope expansion. Chapter 5 summarizes this paper.

2. Basic composition and working principle of quantum dot microdisplay

The performance of quantum dots in the field of microdisplay not only depends on the quality of the material itself, but also on the reasonable design of the device structure and the precise processing technology. Therefore, understanding the working principle of QLED quantum dots and the technological challenges brought by miniaturization is very important.

The luminescence mechanism of quantum dots in the display panel mainly includes two modes: photoluminescence and electroluminescence. In photoluminescence, the quantum dot film acts as a color conversion layer, absorbing the short-wavelength excitation light from the underlying blue backlight and converting it into high-purity red or green light. In the electroluminescence mode, that is QLED, the device achieves true self-luminous display through the radiative recombination of injected electrons and holes in the quantum dots [1].

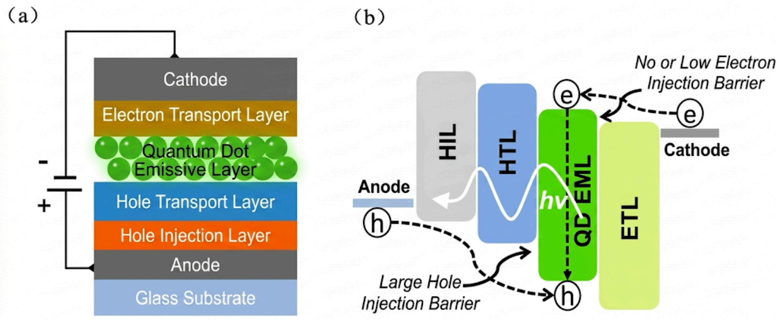

Figure 1. Structure and luminous principle of QLED. (a) Multi-layer heterojunction sandwich structure of QLED; (b) basic luminous mechanism of QLED [1]

To achieve efficient charge injection and balance, as shown in Fig. 1, standard QLEDs typically employ a multi-layer heterojunction sandwich structure, with the layers from bottom to top being: a transparent anode such as ITO, a hole injection layer, a hole transport layer or HTL such as PEDOT:PSS or TFB, a quantum dot light-emitting layer or EML, an electron transport layer or ETL such as ZnO nanoparticles, and a metal cathode such as Al or Ag [1]. This layered structure has very high requirements for the flatness of the interface and energy level matching. The emission of quantum dots comes from the radiative recombination of excitons. A single quantum dot itself is an independent light-emitting center, and its emission characteristics are mainly determined by the size of the quantum dot and the surface ligands, and have no direct correlation with the macroscopic pixel size scaled down to several micrometers or even sub-micrometers. This means that the quantum dot emission is not affected by macroscopic cutting. This self-emission structure is the advantage of quantum dots in microdisplay. It does not have the complex physical stacking and optical crosstalk in LCD technology, and at the same time, because it is solution processing, it also avoids the physical limits in OLED evaporation process and does not have the side wall effect caused by etching when the size of inorganic Micro-LED shrinks, resulting in a significant loss of non-radiative recombination and low luminous efficiency [2, 3].

For new types of display devices, their resolution needs to exceed 1000 PPI, which means that the physical size of the pixels must be compressed to the micrometer or sub-micrometer scale. This extreme spatial miniaturization poses three core challenges for patterning techniques:

The first is the limitation of spatial resolution. Traditional mask-based evaporation (FMM) or conventional inkjet printing technologies are limited by physical obstructions and particle volume, making it difficult to break through the geometric size bottleneck corresponding to extremely high PPI. New micro-nano processing methods that can break through the diffraction limit or fluid limit need to be developed.

Second, the requirement for lossless processing characteristics. Colloidal quantum dots have a very high surface volume ratio, and their optical properties and the integrity of the surface ligands are highly related. During patterning, quantum dots are easily eroded by oxygen, light, photoresist solvents, etc. These factors can cause the detachment of surface ligands, introducing a large number of non-radiative recombination centers on the quantum dot surface, resulting in severe fluorescence quenching, and ultimately causing a sharp decline in the photoluminescence quantum yield (PLQY). Therefore, the patterning process must also be sufficiently gentle, achieving the preparation of patterns while exhibiting the optical properties of the material as much as possible [2].

Third, the electrical compatibility of multi-layer structures is also very important. The electroluminescence of QLED and the close contact between layers are highly related. When preparing high-density pixels, the quantum dot array after patterning must maintain close contact with the upper and lower charge transport layers. The electrical isolation ability of the multi-layer structure at the pixel edge also affects the photoelectric properties of quantum dots. The patterning technology may cause damage to the pixel edge, which will lead to leakage current in non-luminous areas, causing electrical interference between adjacent pixels. This will affect various display characteristics including contrast, color purity, etc. Therefore, industrialized patterning technology also needs to pay attention to introducing methods such as charge blocking layers to solve the leakage problem [5].

3. Research status of ultra-high pixel density quantum dot patternization technology

3.1. Lithography patternization technology

Lithography technology is a classic method for achieving patterization. As shown in Fig. 2, its patterization process mainly relies on photochemical reactions. First, a layer of photo-sensitive photoresist is uniformly coated on the substrate surface. Then, using ultraviolet (UV) light passing through a mask with a preset pattern, the local area of the substrate is irradiated, causing cross-linking or degradation of the exposed photoresist. Finally, the soluble part is washed away with a developing solution, thereby transferring the precise geometric patterns on the mask to the material surface [2].

Thanks to this mature optical projection and selective dissolution mechanism, lithography technology has advantages in extreme resolution. It is also similar to existing semiconductor production aspects, but traditional photoresist is prone to causing quantum dot fluorescence quenching [3]. Currently, the proposed "direct lithography technology" is a possible solution to quantum dot fluorescence quenching. Hahm et al. developed a "dual-ligand passivation system" to achieve direct lithography, manufacturing devices with a resolution of over 15,000 PPI, and retaining the carrier transport characteristics of quantum dots [2]. At the same time, Gao et al. used the ultraviolet-induced ligand exchange mechanism to achieve micrometer-scale arrays, and inserted PMMA as an electric charge blocking layer in the pixel gap, which inhibited leakage current and improved the efficiency of the device [5].

In lithography technology, quantum dot materials are relatively sensitive to ultraviolet exposure, solvents, and etching environments. The multi-step processing process is prone to introducing interface defects and degradation of fluorescence performance. At the same time, this technology requires complex process flows and high-cost equipment, which leads to a relatively high process cost of lithography technology.

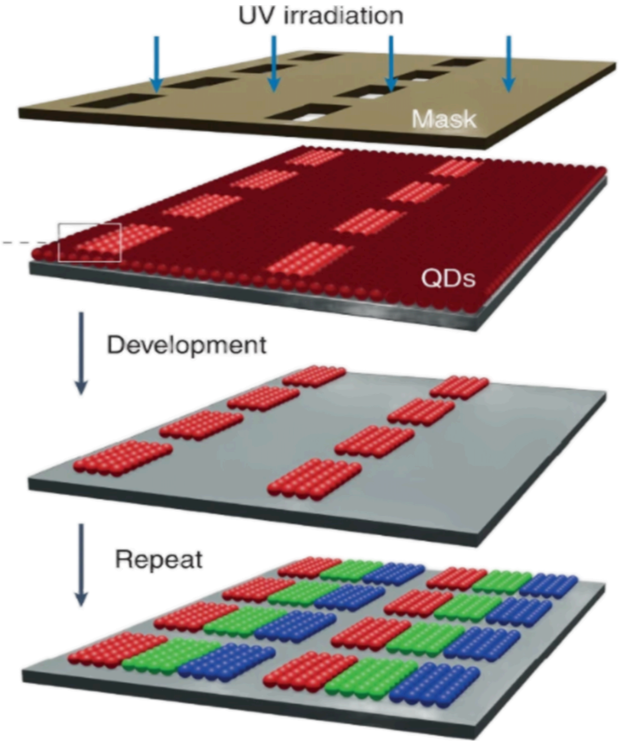

Figure 2. The patterning technology of quantum dots based on lithography processing [2]

3.2. Transfer printing technology

The transfer printing technology employs the "pick-up - placement" micro-nano processing technique. As shown in Fig. 3, the basic patterning process mainly relies on an elastic body stamp to achieve, typically with polydimethylsiloxane or PDMS. First, the physical adhesion force between the stamp and the film is utilized to "pick up" the quantum dot layer that has been pre-deposited on the donor substrate. Then, through high-precision mechanical alignment, the stamp with quantum dots is pressed onto the target receiving substrate. Finally, by regulating the peeling speed or interface energy, the micro-nano pattern is released and "placed" at the target position without any damage [7].

With the "dry method" operation, transfer printing can effectively avoid the erosion of quantum dots by chemical solvents. The transfer technology requires the use of a stamp, but traditional soft stamps are prone to deformation during the transfer of tiny pixels. Zhou et al. designed a composite stamp made of soft and hard PDMS, achieving a pattern fidelity of 94.0% at a density of 6350 PPI, with pixel diameters of only 1.78 μm [7]. Yoo et al. proposed an ultra-high-definition double-layer transfer technology, optimizing the interface charge injection, achieving synchronous transfer of the QD layer and ZnO transport layer, and preparing a full-color array with 2565 PPI, and preparing a green light QLED with an external quantum efficiency (EQE) of 23.3% [6].

The transfer printing technology has high requirements for stamp adhesion force, transfer pressure, and alignment accuracy. Under high PPI conditions, transfer may still be incomplete, pixel misalignment, and uniformity degradation occur, and there is still a considerable distance to reach industrialized mass production.

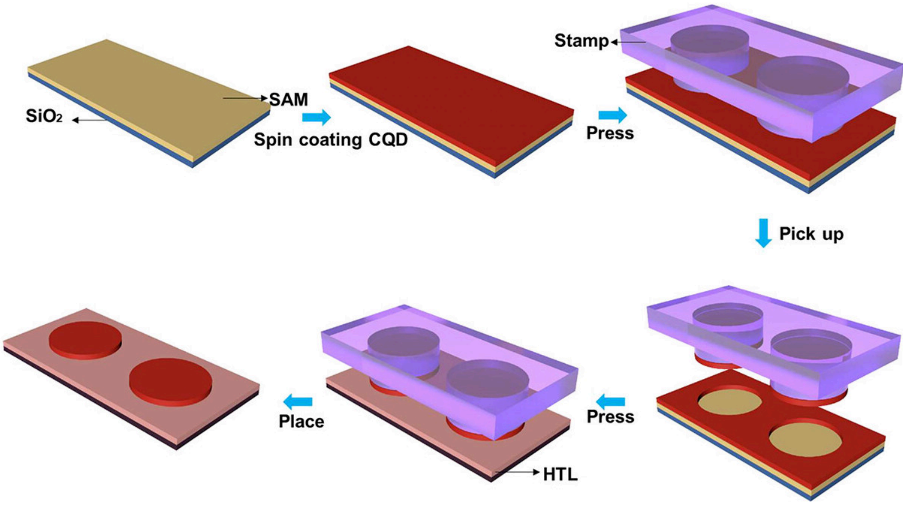

Figure 3. Quantum dot patterning technology based on transfer printing [7]

3.3. Inkjet printing technology

Inkjet printing technology differs from lithography technology. Its basic patterning process mainly relies on the on-demand droplet dispensing mechanism. First, quantum dots are uniformly dispersed in a specific organic solvent to prepare functional ink. Through precise piezoelectric actuators to control the nozzle, tiny volume ink droplets are sprayed to the target substrate at the required position according to the preset program. As the solvent evaporates and the film solidifies, quantum dots are directly deposited in situ to form the required pattern [1].

In addition to lithography and transfer technologies, printing technology has a high material utilization rate and the advantage of convenient large-scale manufacturing, and has also become an important research direction for the patterning of quantum dots. However, the resolution of traditional piezoelectric inkjet printing is usually difficult to exceed 20 μm and cannot well meet the requirements of ultra-high PPI display devices. To further improve the accuracy of patterning manufacturing, As shown in Fig. 4, Wang et al. introduced fluid dynamics to the printing technology. By driving droplet ejection with a high-voltage electric field, they achieved a pixel array with a feature size of approximately 5 μm and prepared efficient QLED devices that meet the requirements of high PPI microdisplay [8].

On the other hand, in addition to the direct spatial resolution, the stability of the quantum dot ink used in printing also affects the performance of the device. Wang et al. designed a core-dual-shell coating structure for inorganic lead halide perovskite quantum dots, which enhanced the UV resistance and moisture stability of red light quantum dot ink, and improved its adaptability and device reliability in industrial printing processes [4].

In printing technology, the evaporation process of droplets is prone to cause coffee ring effect and uneven film thickness phenomena. At the same time, the resolution of printing is also limited by droplet diffusion and nozzle size.

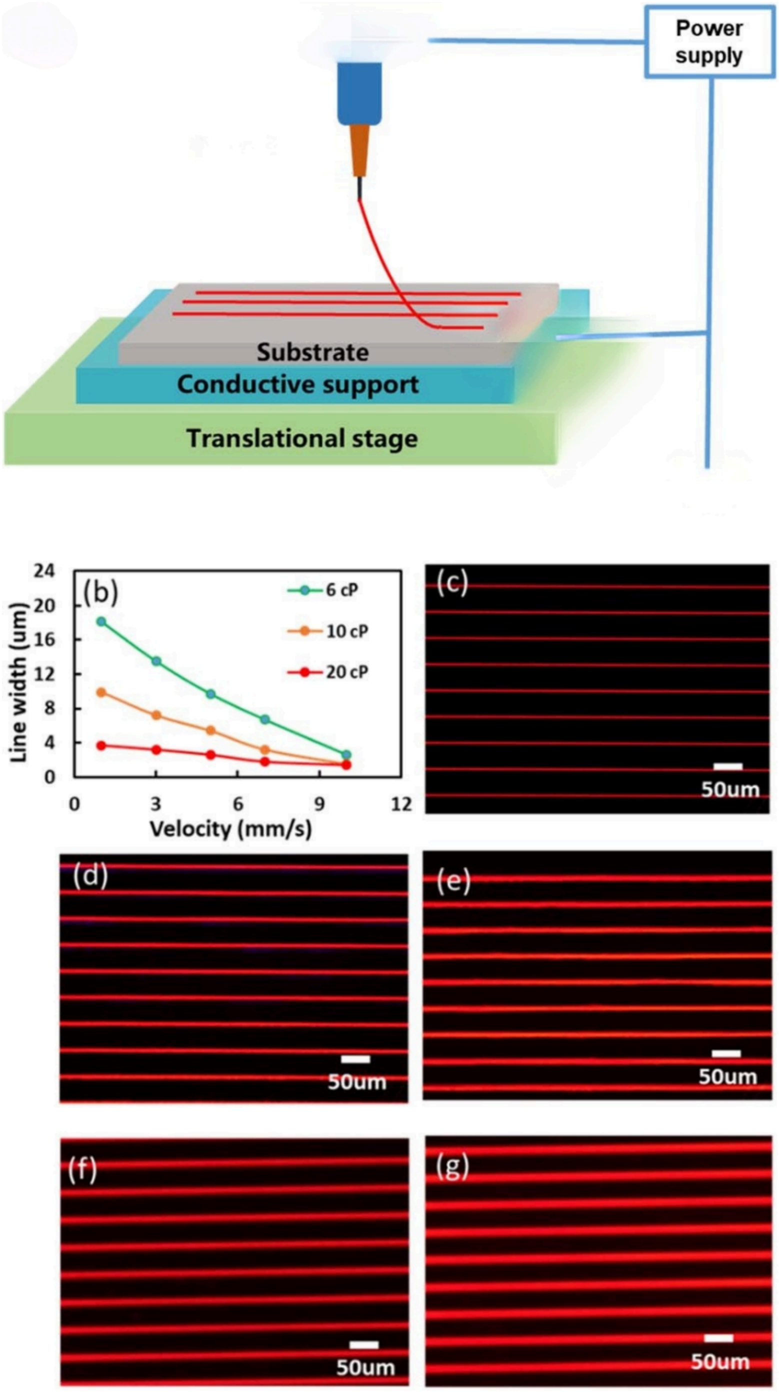

Figure 4. Quantum dot patterning technology based on inkjet printing [8]

3.4. Femtosecond laser processing technology

Laser processing technology, as an advanced manufacturing method, has been extensively studied in the field of micro-nano device fabrication in recent years [9]. Among them, the laser-induced forward transfer (LIFT) technology used for the array integration of nanomaterials such as quantum dots mainly relies on the mechanism of kinetic energy conversion and controlled ejection. As shown in Fig. 5, the operation begins by fixing the transparent donor substrate, which usually features a high-absorption sacrificial layer, and the target receiving substrate face-to-face and closely parallel.Then, a focused pulsed laser beam is used to irradiate the specific area of the absorption interface from the backside. In this micro-area, after absorbing intense laser energy instantaneously, it undergoes local vaporization or plasma expansion, generating extremely high transient gas thrust on the film's backside. This localized high pressure ultimately drives the micron-scale quantum dot film to overcome the microscopic forces and eject in a discrete sheet or droplet form towards the front, and deposit on the receiving substrate, thereby achieving in-situ spatial patterning arrangement [9].

The femtosecond laser-induced forward transfer technology can integrate material deposition, patterization, and pixel alignment in a single transfer step, avoiding thermal diffusion damage [9]. This technology can flexibly construct any shape RGB full-color dot matrix with a resolution of 1.78 μm, and the transferred film shows extremely high flatness and continuity. Compared with previous technologies, the femtosecond laser-induced forward transfer technology has a significant advantage in maintaining the photoelectric properties of quantum dots, and may become the basis for future patterned manufacturing. However, its processing process may introduce local defects and surface roughening, affecting the long-term stability of the obtained equipment.

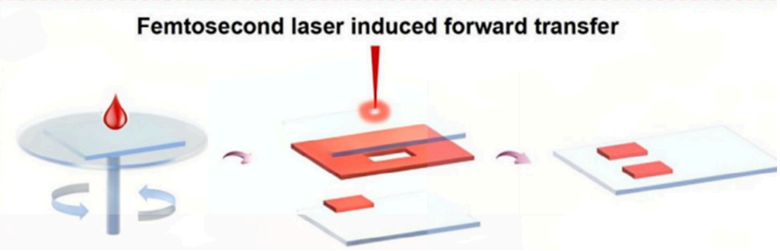

Figure 5. Quantum dot patterning technology based on femtosecond laser processing [9]

3.5. Self-assembly patterning technology

Unlike lithography, transfer printing, and laser processing, self-assembly technology (Self-assembly Technology) is a bottom-up method for constructing quantum dot arrays [10]. Its core principle is to utilize weak interaction forces such as van der Waals forces, electrostatic interactions, hydrogen bonds, and ligand interactions between quantum dots, driving colloidal quantum dots to spontaneously arrange themselves from the solution under no external force conditions, ultimately forming an ordered structure.

Hao et al. pointed out that colloidal quantum dots have a relatively high surface-to-volume ratio, and their surfaces are prone to numerous defects. Therefore, they often exhibit poor chemical stability in complex environments, and disordered stacking also limits the carrier transport efficiency [10]. Self-assembly technology can guide quantum dots to form regular structures on the substrate surface by regulating parameters such as solvent evaporation rate, surface charge state, and interaction forces. This ordered stacking can shorten the distance between adjacent quantum dots, enhance the electron and exciton interactions between nanocrystals, thereby reducing leakage current and non-radiative losses caused by disorderly arrangement, and improving the photoelectric performance and structural stability of the quantum dot film to a certain extent.

In recent years, researchers have developed various implementation methods such as gravity sedimentation, spontaneous evaporation, electric field induction, and self-assembly of colloidal solutions. Among them, the vertical deposition method driven by capillary force and solvent evaporation can form an ordered quantum dot film with fewer defects and uniform thickness on the substrate surface. The electric field-assisted self-assembly can improve the deposition uniformity of different-sized colloidal particles by regulating the applied electric field. In addition, the quantum dot superlattice structure formed by self-assembly can exhibit collective photoelectric properties different from individual quantum dots, showing potential application value in solar cells, photodetectors, and new light-emitting devices.

However, self-assembly technology is highly sensitive to temperature, solvent concentration, and surface charge conditions, and the assembly process is difficult to control and stabilize and repeat. At the same time, it is still difficult to achieve uniform large-area preparation and high-precision pixel definition. Therefore, its practical application in ultra-high PPI full-color QLED microdisplay still faces significant challenges.

3.6. Optical microcavity technology

Optical Microcavity Technology is a non-contact, precise, and light-controlled method based on optical interference. At the micro-nano scale, the light-emitting layer is sandwiched between a high-reflection bottom electrode and a semi-transparent top electrode, forming a resonant cavity. After the light waves reflect multiple times within the cavity, only the specific wavelengths that meet the resonance conditions can undergo constructive interference and be output and enhanced, thereby achieving precise control over the emission spectrum, directionality, and color purity [1]. In the field of colloidal quantum dot display, this technology provides an indirect method for adjusting colors without directly adjusting the quantum dot layer. Researchers can adjust the thickness of the electrodes or functional layers to change the cavity length, thereby selectively obtaining different colors of light emission and achieving a high-color-purity, high-brightness micron-level pixel array [11].

In the scheme based on color-converting optical microcavities (Color-Converting Cavities), Chen et al. proposed a top-emitting microcavity QLED architecture [11]. As shown in Fig. 6, Researchers first spin-coated a patternless white light quantum dot emitting layer composed of red, green, and blue quantum dots in proportion, over the entire device area. Then, they used photolithography to pattern the bottom indium zinc oxide (IZO) phase-change layer. By changing the thickness of IZO in different sub-pixel areas, the resonant conditions for red, green, and blue light emission were respectively constructed. Under the selective filtering effect of microcavity resonance and interference enhancement, the white light emission was selected, and finally, high-saturation RGB monochromatic light was output. A resolution of approximately 1700 PPI and a color gamut coverage of 111% NTSC were achieved. Under a 5.5 V voltage drive, the brightness of the red, green, and blue sub-pixels reached 22170, 51930, and 3064 cd·m^{-2}, respectively.

In addition to achieving full-colorization without etching, the optical microcavity structure can also enhance the light extraction efficiency and color purity of the top-emitting QLED. Lai et al. constructed a top-emitting active matrix quantum dot light-emitting diode (TE-AMQLED) array with a strong microcavity enhancement effect for a micro QLED display chip [12]. This device uses an ITO/Ag/ITO composite anode as the bottom electrode and combines a semi-transparent Ag cathode to form a microcavity structure. By optimizing the thickness of the ZnO layer and the quantum dot layer, the device brightness successfully exceeded 100,000 cd·m^{-2}, and the current efficiency reached 28 cd·A^{-1}. The research team also successfully fabricated a 1.49-inch top-emitting AMQLED array on a LTPS active backplane, verifying the compatibility of the microcavity top-emitting structure with the existing OLED-LTPS process chain, providing an important reference for the large-scale integration of ultra-high PPI micro displays.

Overall, optical microcavity technology provides a new path for QLED micro displays to avoid direct processing of quantum dots, with advantages such as high color purity, high directionality, and high process compatibility. However, it seems that this technology may cause a blue shift in the resonant wavelength and color deviation when the observation angle increases. At the same time, because the microcavity structure is sensitive to the thickness errors of each functional layer, the uniformity of large-scale manufacturing also has very high requirements.

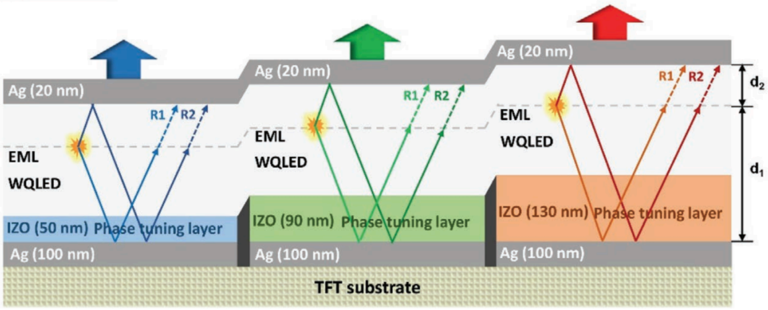

Figure 6. The structural design and optical resonance mechanism of red, green, and blue microcavity devices constructed by using different thicknesses of ITO phase-tuning layers [11]

4. Challenges and future prospects

4.1. Current technological challenges

Although the quantum dot microarray technology has made significant progress in recent years, there are still many key bottlenecks preventing the large-scale industrialization of ultra-high-resolution microdisplay. Among them, the major problems are large-scale high yield rate for large areas and precise alignment at large scales. When manufacturing near-eye display devices, it is necessary to integrate hundreds of millions or even billions of red, green, and blue pixels within a small area while ensuring their precise arrangement. Under this premise, whether it is lithography, transfer printing, inkjet printing, etc., maintaining nanometer-level alignment accuracy over a large area is extremely difficult.

For lithography technology, multiple exposures and overlaying will introduce cumulative errors. As the pixel size continues to decrease, this error will be further amplified. For transfer printing technology, the stamp is prone to deformation during mechanical contact, thereby reducing the transfer accuracy and yield. And for printing technology, the liquid droplet spreading, coffee ring effect, and substrate influence make it difficult to precisely control the landing and morphology of quantum dot materials.

There are also issues related to side wall edge damage and photoelectric crosstalk at the limit pixel size. When the pixel size is reduced to the micrometer or sub-micrometer scale, the area at the device edge is relatively large, making defects more likely to occur during the processing. For example, in plasma etching, laser processing, etc., suspended bonds and surface trap states are easily introduced at the pixel edge, leading to enhanced non-radiative recombination and further reducing the luminous efficiency. At the same time, the very small spacing between adjacent pixels is prone to optical leakage and current leakage, which will reduce the display contrast and color purity.

4.2. Technological evolution trend

In response to the technical challenges faced, the realization of ultra-high PPI micro displays in the future is likely to involve multiple technologies. By leveraging the respective advantages of different manufacturing processes and avoiding their individual limitations, it is hoped that process collaboration can balance resolution, yield, material utilization, and device performance. For instance, the pixel isolation structure, driving circuit, and bottom optical microcavity can be first constructed using lithography technology, ensuring the dimensional accuracy and electrical consistency of the device. Then, inkjet printing, electrohydrodynamic (EHD) printing, or self-assembly techniques can be combined to supplement quantum dot luminescent materials in the predetermined area. Finally, the microcavity structure, photonic crystals, or nano-optical structures can be used to enhance the control of the light field, improving the light output efficiency and directionality.

In addition, there is also artificial intelligence-assisted manufacturing, and machine vision alignment may also provide significant assistance for future manufacturing. By optimizing the printing path using AI algorithms and reducing errors, stability and consistency in large-scale production can be improved.

5. Conclusion

In conclusion, this paper reviews the quantum dot patterning technologies for high-resolution micro-display applications in recent years and their research progress, focusing on the mechanisms, characteristics, and current development status of printing, transfer, and lithography process routes. Each patterning technology has its own advantages and limitations. Currently, there is still no process scheme that simultaneously meets ultra-high resolution, high performance, and low cost. The future development direction may focus on the collaborative integration of multiple patterning technologies, such as combining the high-precision structure definition ability of lithography with the material deposition advantages of printing and transfer technologies, to achieve the large-scale manufacturing of ultra-high PPI quantum dot micro-display devices. The development of quantum dot patterning technology is not only a key manufacturing foundation for achieving ultra-high-resolution micro-display devices but also provides important support for the development of new optoelectronic devices, integrated photonics, and advanced display technologies, with broad research value and application prospects.

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