

Cite as: Appl. Phys. Lett. 118, 173501 (2021); https://doi.org/10.1063/5.0047854 Submitted: 17 February 2021 • Accepted: 09 April 2021 • Published Online: 26 April 2021

Yang Li, 🗓 Jin Tao, Qiang Wang, et al.

COLLECTIONS

EP

This paper was selected as an Editor's Pick







ARTICLES YOU MAY BE INTERESTED IN

Development of microLED

Applied Physics Letters 116, 100502 (2020); https://doi.org/10.1063/1.5145201

Comparison of size-dependent characteristics of blue and green InGaN microLEDs down to 1 μm in diameter

Applied Physics Letters 116, 071102 (2020); https://doi.org/10.1063/1.5144819

Beyond solid-state lighting: Miniaturization, hybrid integration, and applications of GaN nanoand micro-LEDs

Applied Physics Reviews 6, 041315 (2019); https://doi.org/10.1063/1.5096322





Cite as: Appl. Phys. Lett. **118**, 173501 (2021); doi: 10.1063/5.0047854 Submitted: 17 February 2021 · Accepted: 9 April 2021 · Published Online: 26 April 2021







Yang Li,^{1,2} Jin Tao,^{1,a)} 🕞 Qiang Wang,¹ Yongzhou Zhao,^{1,2} Yifang Sun,^{2,3} Panyuan Li,^{1,2} Jinguang Lv,¹ Yuxin Qin,¹ Weibiao Wang,¹ Qinghui Zeng,³ and Jingqiu Liang¹

AFFILIATIONS

- ¹State Key Laboratory of Applied Optics, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun, Jilin 130033, China
- ²University of Chinese Academy of Sciences, Beijing 100049, China
- ³State Key Laboratory of Luminescence and Applications, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun, Jilin 130033, China

ABSTRACT

In this study, quantum dot color conversion layers (QDCCLs) for full-color micro-LED display were successfully fabricated using microfluidics to conduct red and green perovskite quantum dots to the position of the pixel array. The QDCCL with full-color pixel size of $200 \times 200 \,\mu\text{m}$ and sub-pixel size of $140 \times 50 \,\mu\text{m}$ was achieved. Perovskite quantum dots with high quantum yield and narrow full width at half-maximum were used to achieve a wide color gamut, which was 131% of National Television Systems Committee standard. The proposed microfluidics-based QDCCL featured easy fabrication, low cost, high performance, and good integration prospects.

Published under license by AIP Publishing. https://doi.org/10.1063/5.0047854

In recent years, micro-LED displays have attracted increasing research and commercial attention because of their unique advantages, including high resolution, low power consumption, high brightness, flexibility, fast response, and high reliability. However, there are still some remaining issues that hinder the practical implementation of full-color micro-LED displays. It is nearly impossible to realize monolithic integration of RGB micro-LEDs via high-efficiency epitaxial RGB fabrication on a single wafer because of incompatibility of the material systems. Transfer printing, as a commonly used technology to realize full-color micro-LEDs, has undergone a lot of research, including elastomer stamping, electrostatic transfer, electromagnetic transfer, laser-assisted transfer, and fluid self-assembly. Yet, the high cost and low yield of transfer printing are the main obstacles to the commercialization of full-color micro-LED display. 6.7

To overcome these issues, a compromise that combines a blue or ultraviolet (UV) monolithic micro-LED array with a quantum dot color conversion layer (QDCCL) has drawn interest.^{6,8} Quantum dots (QDs) are usually synthesized via solution-based methods and have unique properties, such as high quantum yield (QY), size-dependent emission wavelength, and narrow bandwidth emission.^{9–12} As reported in previous studies, there are mainly two schemes to pattern QDCCLs: one is to spray QDs on the micro-LED array or on a

transparent substrate by ink-jet printing, ^{13–15} and the other is to pattern the QDs via mixing the QDs with the photoresist in a certain ratio and using multiple photolithography methods. ^{16,17} However, ink-jet printing depends on the nozzle, which is more suitable for QD pixels with larger sizes, and it is difficult to control the edge morphology of the QDs using this method. Photolithography has a good control on the size and morphology of the QDs; however, a large amount of QDs is wasted because they need to be mixed with composite materials, such as a photoresist for lithography and development, and the photoresist has a large influence on the fluorescence efficiency of the QDs.

Microfluidics has been widely used in chemical and biological analysis because of its advantages for transporting small amounts of reagents. ^{18–20} The microchannels are usually fabricated via lithography using various materials, including silicon, glass, and polymers. The handling sizes of microfluidics are about several to hundred micrometers, which are suitable for the pixel sizes of micro-LED display. Herein, we propose a much easier way to fabricate a low-cost and high-resolution QDCCL by using microfluidics. Benefiting from the characteristic that the patterns of QDs are determined by the precision of photolithography, high precision of several micrometers for microchannels of microfluidics becomes possible. Moreover, the large amount of QD losses can be easily avoided as the QDs only exist in

a) Author to whom correspondence should be addressed: taojin@ciomp.ac.cn

the microchannels, which is challenging for the photolithography method.

The structure of a single pixel of the proposed full-color micro-LED display is shown in Fig. 1(a). The device is composed of a blue micro-LED array and a QDCCL with red and green QDs. Blue micro-LEDs are used as the excitation light source to irradiate the red and green QDs to convert blue light to red and green, respectively. The QDCCL is made by bonding a glass substrate and a PDMS mold with microchannels. The converted red and green light from the QDs and blue light through the transparent QDCCL form the three primary colors required to realize the full-color display. The QDs are conducted into the microchannels in solution via syringes, as shown in Fig. 1(b). The solvent (hexane) is easily volatilized to form the solid QDs. After the solvent is volatilized, the outlet and inlet of the microchannels are sealed to protect the QDs from degradation by air and water vapor. The PDMS mold was fabricated via SU-8 mold photolithography [Fig. 1(c)], PDMS casting, PDMS and SU-8 mold separation, PDMS drilling, and PDMS bonding with transparent glass substrates [Fig. 1(d)], as reported in previous work. Figure 1(e) shows the magnified image of the microchannels. The pixel size was $200 \times 200 \,\mu\text{m}$, which is approximate to the resolutions of display monitors, and contained red and green QD sub-pixels and a blue sub-pixel. The sub-pixel size was $140 \times 50 \,\mu\text{m}$, as shown in Fig. 1(e). Further details are provided in the supplementary material.

Compared with the traditional QDs, perovskite QDs (PQDs) have the advantages of a low cost and good luminescent properties, such as high quantum yield, narrow excitation spectrum, narrow emission, and adjustable emission spectrum.^{23,24} In this work, CsPbX3 PQDs (CsPbBr3 for green light and CsPbI2Br for red light) were synthesized via thermal injection at high temperature, as previously reported.¹⁰ Further details on the QD synthesis are provided in the supplementary material. The stability of most fluorescent quantum dots is the main technical bottleneck in practical applications, especially when exposed to high temperatures.^{25,26} Some fluorescent QDs decompose when the temperature gets 90 °C.²⁷ Baking is necessary for the photolithography process, usually at 100 °C, which may cause the

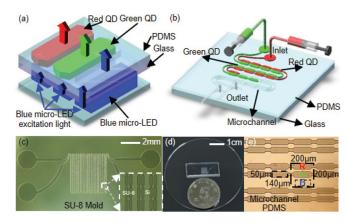


FIG. 1. (a) Schematic diagram of a single pixel of the full-color micro-LED display. (b) Red and green QDs are injected in the micro-channels by syringes. (c) SU-8 mold on the silicon substrate. (d) PDMS microchannel bonding with the glass substrate. (e) Magnified image of the micro-channels of the red and green QDs. The RGB pixel size is $200 \times 200 \ \mu m$ and the sub-pixel size is $50 \times 140 \ \mu m$.

degradation of QDs. On the other hand, it is a huge waste of QDs during the development process. In this paper, the QDs array is fabricated by microfluidics at room temperature, which has no harm to QDs. And there can be a space between the blue LED and the QDs color conversion layer that prevents the thermal transfer from the LED to the QDs. Therefore, the proposed device shows obvious advantage less requirement on the temperature-resistance of QDs.

The UV-Vis absorption (dash line) and photoluminescence (PL) spectra (solid line) of the prepared PQDs were characterized, as shown in Figs. 1(a) and 1(b). Two kinds of PQDs in hexane solution were measured via UV excitation. The insets in Figs. 2(a) and 2(b) are images of the red and green PQDs in hexane excited by 395 nm UV light. The emission peaks of the green and red PQDs were measured as 517 and 647 nm, respectively, with full width at half-maximum (FWHM) of 17.3 and 35.2 nm, respectively. The quantum yield (QY) of green PQDs is 90%, and that of red PQDs is 51% (see supplementary material). Trap states in the PQDs significantly influence their optical properties, and surface passivation of the PQDs can increase their QY up to nearly 100%. 28 The prepared red and green PQDs were dried and characterized via scanning electron microscopy (SEM), as shown in Figs. 2(c) and 2(d). It can be seen that the prepared green and red PQDs presented a cubic shape and had good homogeneity. The sizes of the red and green PQDs were $\sim 20.9 \pm 4.5$ and 13.9 ± 2.6 nm, respectively (see supplementary material).

To form the QDCCL, the PQD solution was injected into the microchannels using syringes actuated by micro-pumps (LD-P2020). As shown in Fig. 3(a), first, the green PQDs were slowly injected into the green microchannel from the inlet, and it then flowed from the corresponding outlet on the other side. The injection of a 20×20 array can be completed within several seconds, and the injection speed could be tuned using the micro-pumps. Then, the red PQDs were slowly injected into the microchannels, as shown in Fig. 3(b). Because the red and green PQD microchannels do not cross each other, the two kinds of PQDs could be simultaneously injected into reduce preparation time of the proposed device.

The minimum pixel resolution of the proposed QDCCL was approximately three times that of the width of the microchannels, which is determined by photolithography (several micrometers or

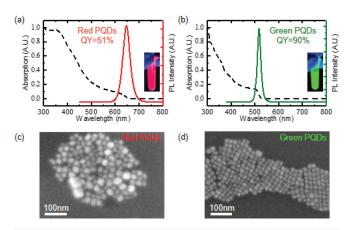


FIG. 2. Absorption and emission spectra of red (a) and green (b) PQDs. (c) SEM image of the red PQDs and (d) green PQDs.

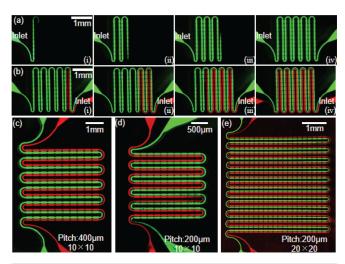


FIG. 3. (a) Fabrication process for the green QDCCL and (b) red QDCCL. QDCCLs with full-color pixel pitch and a pixel number of (c) 400 μ m and 10 \times 10, (d) 200 μ m and 10 \times 10, and (e) 200 μ m and 20 \times 20.

even sub-micron). However, considering the aspect ratio of the microchannels, when the channel width decreased, its height decreased accordingly, resulting in a thinner solidified PQDs layer. Thinner red and green PQD layers could not adequately absorb the blue backlight, which led to a lower electro-photo efficiency and leakage of blue light from red and green sub-pixels. Therefore, a balance exists between the resolution and efficiency. In this work, QDCCLs with different pitches (resolution) and pixel numbers were demonstrated, as shown in Figs. 3(c)-3(e). The pitches, pixel number, and light emitting areas of the QDCCLs were 400 μ m, 10 × 10 and 4 × 4 mm [Fig. 3(c)], 200 μ m, 10×10 and 2×2 mm [Fig. 3(d)], and $200 \mu m$, 20×20 and 4×4 mm [Fig. 3(e)], respectively. There was no obvious crosstalk between the adjacent red pixels or between adjacent red and green pixels. Additionally, the luminescence of the QDCCLs was uniform, and their fabrication via the microfluidic method is very economical. For a 2 K (2048×1080) full-color display, a total \sim 1 mg of red and green PQDs is required, assuming a concentration of 2 mg/ml and a microchannel height of 30 μ m.

An easy way to improve the conversion efficiency from blue to red and green light is by changing the concentration of the PQDs solution. Figures 4(a) and 4(b) shows the emission spectra of the red and green PQDs, respectively, at six different concentrations ranging from 0.3 to 20 mg/ml in a hexane solution under excitation with a blue LED source (peak wavelength of 457 nm). The PL emission peaks and FWHMs of the red (red line) and green (green line) PQDs were found to display a unidirectional change as the concentrations increased, as shown in Fig. 4(c). Because of Förster resonant energy transfer or reabsorption, the PL peak wavelength redshifted as the concentration increased for both the red and green PQDs. ^{29,30} In contrast, the FWHM decreased with increasing concentration. The concentration dependent peak wavelength and FWHM provide a potential way to adjust the optical characteristics of RGB micro-LED pixels to realize improved display performance.

Figure 4(d) shows the relative intensity of the PL emission peaks of the red and green PQDs at different concentrations ranges from 0.1

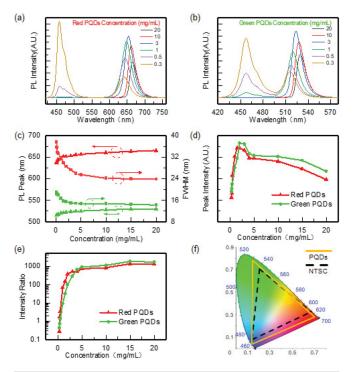


FIG. 4. Emission spectra of (a) red and (b) green PQDs with different concentrations. (c) Emission peak (left) and FWHM (right) of red and green PQDs at different concentrations. (d) Emission peak intensity of red and green PQDs at different concentrations. (e) The ratio of the peak intensity of the red and green PQDs to blue peak intensity at different concentrations. (f) Color gamut range of red and green QDCCLs and blue LED.

to 20 mg/ml. It can be seen that the peak intensities of the red and green PQDs increase first and then decrease. The peak intensities of the red and green PQDs reach a maximum at 1.5 and 2 mg/ml, respectively, indicating full absorption of the blue light. When the concentration of the PQDs exceeds the limits, the output photoluminescence recedes because of scattering and absorption effects. Figure 4(e) shows the ratio of the peak intensities of the red and green lights to the blue light. Full blue light absorption by the PQDs is important, and full conversion from blue to red/green light not only enhances the efficiency of the proposed device but also allows the blue filter or Bragg reflection layers to be omitted, which reduces fabrication complexity and cost.

On the other hand, the geometry configuration of the red and green QD color conversion layers and the blue LED is another important factor to avoid blue light leakage and optical crosstalk in the subpixel LEDs. The divergence angle of the blue LED, the shapes and the refractivity of the QD dots, and the distance from the blue LED to QD dots synergistically determine the performance of the full-color micro-display. More geometry optimization of the QDs-based micro-LEDs is needed to get better performance in the future.

The color gamut is an important index for displays and indicates the ability to reproduce natural colors. PQDs have been demonstrated as favorable materials to realize a wide color gamut as a color filter in LCDs, ^{32,33} color converters for LEDs, ⁸ or electroluminescent layers. ⁹ In this work, the color gamut was measured based on red and green

PQDs and blue LED, as shown in Fig. 4(f). The color space coverage is as high as 131% of the National Television Systems Committee (NTSC) standard, which is better than most previous reports. The high efficiency and wide color gamut of the QDCCL-based micro-LED will be beneficial in future practical applications.

In conclusion, the proposed microfluidics-based QDCCLs could enable preparation of full-color micro-LEDs without mass transfer, which is the current bottleneck for their broad application. Compared with ink-jet printing and photolithography methods, microfluidics has the advantages of high precision and economic QD utilization, which can further promote commercialization of micro-LEDs. PQDs with a high luminescence efficiency were synthesized via thermal injection. The QY of the red and green PQDs reached up to 51% and 90%, respectively, and their FWHMs were narrow at 23.8 and 14.2 nm, respectively. By adjusting the concentration, a high fluorescence conversion efficiency and a wide color gamut of 131% NTSC could be realized.

See the supplementary material for the fabrication of the micro-fluidics and the synthesis and characterization of PQDs.

This work was supported by the National Key Research and Development Program of China (Nos. 2018YFB1801901, 2018YFB1801902, and 2019YFB2006000), the Science and Technology Development Project of Jilin Province (Nos. 20190302062GX and 20200401056GX), and the National Natural Science Foundation of China (No. 62004195).

DATA AVAILABILITY

The data that support the findings of this study are available within the article and its supplementary material.

REFERENCES

- ¹Y. Huang, E. L. Hsiang, M. Y. Deng, and S. T. Wu, Light Sci. Appl. **9**, 105 (2020).
- ²S. Zhang, Z. Gong, J. J. D. McKendry, S. Watson, A. Cogman, E. Xie, P. Tian, E. Gu, Z. Chen, G. Zhang, A. E. Kelly, R. K. Henderson, and M. D. Dawson, IEEE Photonics J. 4, 1639 (2012).
- ³K. Chung, J. Sui, B. Demory, and P.-C. Ku, Appl. Phys. Lett. 111, 041101 (2017)
- ⁴H. Zhang and J. A. Rogers, Adv. Opt. Mater. 7, 1 (2019).
- ⁵T. Wu, C. W. Sher, Y. Lin, C. F. Lee, S. Liang, Y. Lu, S. W. H. Chen, W. Guo, H. C. Kuo, and Z. Chen, Appl. Sci. 8, 1557 (2018).
- ⁶X. Zhou, P. Tian, C.-W. Sher, J. Wu, H. Liu, R. Liu, and H.-C. Kuo, Prog. Quantum Electron. 71, 100263 (2020).
- ⁷K. Ding, V. Avrutin, N. Izyumskaya, Ü. Özgür, and H. Morkoç, Appl. Sci. 9, 1206 (2019).

- ⁸Z. Liu, C. H. Lin, B. R. Hyun, C. W. Sher, Z. Lv, B. Luo, F. Jiang, T. Wu, C. H. Ho, H. C. Kuo, and J. H. He, Light Sci. Appl. **9**, 1 (2020).
- ⁹Q. Shan, C. Wei, Y. Jiang, J. Song, Y. Zou, L. Xu, T. Fang, T. Wang, Y. Dong, J. Liu, B. Han, F. Zhang, J. Chen, Y. Wang, and H. Zeng, Light Sci. Appl. 9, 163 (2020).
- 10 Y. Su, X. Chen, W. Ji, Q. Zeng, Z. Ren, Z. Su, and L. Liu, ACS Appl. Mater. Interfaces 9, 33020 (2017).
- ¹¹K. Dohnalová, A. N. Poddubny, A. A. Prokofiev, W. D. De Boer, C. P. Umesh, J. M. Paulusse, H. Zuilhof, and T. Gregorkiewicz, Light Sci. Appl. 2, e47 (2013).
- ¹²X. Zhu, L. Bian, H. Fu, L. Wang, B. Zou, Q. Dai, J. Zhang, and H. Zhong, Light Sci. Appl. 9, 73 (2020).
- ¹³S.-W. Huang Chen, C.-C. Shen, T. Wu, Z.-Y. Liao, L.-F. Chen, J.-R. Zhou, C.-F. Lee, C.-H. Lin, C.-C. Lin, C.-W. Sher, P.-T. Lee, A.-J. Tzou, Z. Chen, and H.-C. Kuo, Photonics Res. 7, 416 (2019).
- ¹⁴H.-V. Han, H.-Y. Lin, C.-C. Lin, W.-C. Chong, J.-R. Li, K.-J. Chen, P. Yu, T.-M. Chen, H.-M. Chen, K.-M. Lau, and H.-C. Kuo, Opt. Express 23, 32504 (2015).
- 15H.-Y. Lin, C.-W. Sher, D.-H. Hsieh, X.-Y. Chen, H.-M. P. Chen, T.-M. Chen, K.-M. Lau, C.-H. Chen, C.-C. Lin, and H.-C. Kuo, Photonics Res. 5, 411 (2017).
- ¹⁶H. M. Kim, M. Ryu, J. H. J. Cha, H. S. Kim, T. Jeong, and J. Jang, J. Soc. Inf. Disp. 27, 347 (2019).
- 17 X. Li, D. Kundaliya, Z. J. Tan, M. Anc, and N. X. Fang, Opt. Express 27, 30864 (2019).
- ¹⁸W. Xiong, Y. S. Zhou, X. N. He, Y. Gao, M. Mahjouri-Samani, L. Jiang, T. Baldacchini, and Y. F. Lu, Light Sci. Appl. 1, 1 (2012).
- ¹⁹V. Bianco, B. Mandracchia, V. Marchesano, V. Pagliarulo, F. Olivieri, S. Coppola, M. Paturzo, and P. Ferraro, Light Sci. Appl. 6, e17055 (2017).
- ²⁰L. Shang, Y. Cheng, and Y. Zhao, Chem. Rev. 117, 7964 (2017).
- ²¹J. C. Mcdonald, D. C. Duffy, J. R. Anderson, and D. T. Chiu, Electrophoresis 21, 27 (2000).
- ²²J. Friend and L. Yeo, Biomicrofluidics 4, 026502 (2010).
- 23 J. Song, J. Li, X. Li, L. Xu, Y. Dong, and H. Zeng, Adv. Mater. 27, 7162 (2015)
- ²⁴C. H. Kang, I. Dursun, G. Liu, L. Sinatra, X. Sun, M. Kong, J. Pan, P. Maity, E. N. Ooi, T. K. Ng, O. F. Mohammed, O. M. Bakr, and B. S. Ooi, Light Sci. Appl. 8, 94 (2019).
- 25 P. Fulmek, C. Sommer, P. Hartmann, P. Pachler, H. Hoschopf, G. Langer, J. Nicolics, and F. P. Wenzl, Adv. Opt. Mater. 1, 753 (2013).
- ²⁶P. Fulmek, J. Nicolics, W. Nemitz, and F. P. Wenzl, Mater. Chem. Phys. 196, 82 (2017).
- ²⁷J. Tang, F. Li, G. Yang, Y. Ge, Z. Li, Z. Xia, H. Shen, and H. Zhong, Adv. Opt. Mater. 7, 1 (2019).
- ²⁸F. Liu, Y. Zhang, C. Ding, S. Kobayashi, T. Izuishi, N. Nakazawa, T. Toyoda, T. Ohta, S. Hayase, T. Minemoto, K. Yoshino, S. Dai, and Q. Shen, ACS Nano 11, 10373 (2017).
- ²⁹M. Lunz, A. L. Bradley, W. Y. Chen, V. A. Gerard, S. J. Byrne, Y. K. Gun'Ko, V. Lesnyak, and N. Gaponik, Phys. Rev. B 81, 1 (2010).
- ³⁰Y. Wang, Y. Yang, P. Wang, and X. Bai, Optik **139**, 56 (2017).
- ³¹C. Sommer, P. Hartmann, P. Pachler, M. Schweighart, S. Tasch, G. Leising, and F. P. Wenzl, Opt. Mater. 31, 837 (2009).
- ³²H.-W. Chen, J.-H. Lee, B.-Y. Lin, S. Chen, and S.-T. Wu, Light Sci. Appl. 7, 17168 (2018).
- ³³H. W. Chen, R. D. Zhu, J. He, W. Duan, W. Hu, Y. Q. Lu, M. C. Li, S. L. Lee, Y. J. Dong, and S. T. Wu, Light Sci. Appl. 6, e17043 (2017).