Efficient quantum dot light-emitting diodes with ultra-homogeneous and highly ordered quantum dot monolayer

Denglin Zhao^{1,2}, Yueting Zheng¹, Tingtao Meng¹, Yangbin Zhu¹, Jipeng Jing¹, Xiang Chen¹, Hongjin Gao¹, Chaomin Mao¹, Wenchen Zheng¹, Hailong Hu¹, Tailiang Guo^{1,2} and Fushan Li^{1,2*}

ABSTRACT Regarding conventional quantum dot light-emitting diodes (QLEDs) fabricated by using the spin-coating (SC) technique, voids and interstitial spaces are inevitable due to unordered quantum dots (QDs) stacking, generating device leakage current under an external bias. In the present study, we fabricated an ultra-homogeneous and highly ordered QD monolayer by adopting the Langmuir-Blodgett (LB) technique. The QD monolayer was transferred as a emissive layer with a horizontal lifting (HL) method to a red QLED, which exhibited high performance with an external quantum efficiency (EQE) of 19.0% and lifetime ($T_{95}@100\text{ cd m}^{-2}$) of 13,324 h. When compared with the SC-based device, the EQE and lifetime were improved by 15% and 183% due to the compact and ordered QD monolayer that lowered the leakage current. Moreover, white QLEDs with stacked QD monolayers could be obtained at a low voltage of 4 V because LB technique is an organic-solvent-free approach avoiding interlayer mixing and controlling the QD layer thickness precisely. In addition, we successfully fabricated an ultra-homogeneous large-area QD monolayer on a rectangular substrate with a size of $9\text{ cm} \times 5\text{ cm}$, indicating the promising size scalability of the LB-HL strategy.

Keywords: quantum dots, Langmuir-Blodgett, quantum dot light-emitting diodes

INTRODUCTION

Colloidal quantum dots (QDs) have remarkable application prospects in lighting and display fields due to their simple preparation, low cost, high spectral purity and precise color tunability [1–8]. Over the past decade, the studies on optimization of QD syntheses and device structures have been intensively performed to enhance the performance of QD light-emitting diodes (QLEDs) [9–12]. Generally, QDs are spin-coated on indium-tin-oxide (ITO) substrate to form a thin film as the emissive layer. Unfortunately, the uncontrollable QD stacking during the spin-coating (SC) process makes it easy to produce voids, causing large leakage current under an external bias voltage and unsatisfactory device lifetime. Therefore, a critical issue of controlling the QD stacking during solution processes needs to be addressed.

Self-assembly of microstructures is ubiquitous in nature [13,14]. In order to better adapt to the environment, the above-

mentioned microstructures tend to be more compact and ordered to form more assemblies in a smaller space. QDs are analogous to the microstructures in biology, and it is of necessity to fabricate compact and ordered films to avoid the generation of voids and interstitial spaces, which may significantly enhance the efficiency of QLEDs [15]. The Langmuir-Blodgett (LB) technique is a suitable choice for the fabrication of a compact and ordered QD monolayer due to its cost-efficient, large-area fabrication and precise control of a monolayer formation process, which has become one of the most promising techniques of fabricating self-assembly films [16–20]. In the current work, a strategy of realizing a highly ordered, ultra-homogeneous QD monolayer based on the LB technique is proposed to achieve efficient QLEDs. Significantly, QDs at the air-liquid interface are confined to a horizontal two-dimensional space, resulting in QDs that can only migrate in the horizontal direction. Afterwards, barriers (usually of Teflon) are closed slowly to compress a monolayer in order that surface pressure of it is adjusted to realize a compact and ordered one. Moreover, we report a QLED containing a QD monolayer which was fabricated by the LB technique and transferred to the device directly. Excitingly, the QD monolayer based on the LB technique is compact and ordered, resulting in low leakage current, high efficiency and long lifetime of QLEDs. Compared with classical SC-based QLEDs, the maximum external quantum efficiency (EQE) values for LB-based QLEDs is improved from 16.5% to 19.0%. The LB technique is an organic-solvent-free method so that the previously deposited layers will not be damaged upon the transfer of the following monolayer from the air-liquid interface. Moreover, it is adopted for the fabrication of well-defined layered structures with molecular level precision. Therefore, QD films with different thicknesses (different numbers of QD layers) can be fabricated by repeatedly transferring from the air-liquid interface to study the impact of the thickness of QD monolayers on the device efficiency. According to the results, the red QLEDs exhibit the highest efficiency when the QD emissive layer is a single monolayer. In addition, the blue, green, and red QD monolayers can be successively transferred to the underlying hole transport layers, and subsequently the QD monolayers with different colors can be adjusted to obtain white QLEDs. Because the LB technique involved in this work is a method without the influence of organic solvent, there exists no buffer layers between the QD monolayers, which significantly simplifies the fabrication process of QLEDs and lowers the turn-on voltage of

¹ Institute of Optoelectronic Technology, Fuzhou University, Fuzhou 350116, China

² Fujian Science & Technology Innovation Laboratory for Optoelectronic Information of China, Fuzhou 350116, China

* Corresponding author (email: fsli@fzu.edu.cn)

white QLEDs with stacked QD monolayers [21]. The Commission Internationale de l'Eclairage (CIE) coordinates of the white QLEDs are located extremely close to the ideal white point of (0.34, 0.33). Finally, an ultra-homogeneous large-area monolayer with a size of $9\text{ cm} \times 5\text{ cm}$ can be achieved with the application of the LB technique. These results prove that the LB technique is a promising approach for achieving high-performance QLEDs with a compact and ordered QD monolayer.

EXPERIMENTAL SECTION

LB-based QD monolayer fabrication

CdSe/ZnS QDs were acquired from Suzhou Xingshuo Nano (China). A Langmuir trough (Powereach JML04C1) was adopted to assemble the LB-based QD monolayer. Typically, $50\text{ }\mu\text{L}$ QD solution dispersed in hexane was added to deionized water and spread at the air-liquid interface. After evaporation of the organic solvent, two Teflon barriers approached at a speed of 0.5 mm s^{-1} until a certain surface pressure (monitored by Wilhelmy plate) was achieved in order to form a monolayer. Subsequently, the QD monolayer was transferred from the water surface to the target substrate *via* a horizontal lifting (HL) method. Multilayer transferring could be realized by simply repeating this step.

QLEDs

ITO glass substrate was precleaned by ultrasonication in isopropanol, acetone, and deionized water in sequence for 10 min each before use. Poly(ethylenedioxythiophene):polystyrene (PEDOT:PSS, from Xi'an Polymer Light Technology Co., Ltd.)

and poly(9,9-dioctylfluorene-*co*-N-(4-butylphenyl) diphenylamine) (TFB, from Xi'an Polymer Light Technology Co., Ltd.) were spin-coated successively at 3000 r min^{-1} for 50 s and later annealed at 130°C for 30 min. Subsequently, an LB-based QD monolayer was transferred on top of TFB. Followed by annealing at 100°C for 15 min, ZnMgO (from Guangdong Poly OptoElectronics Co. Ltd) was spin-coated at 3000 r min^{-1} for 50 s. Finally, an Ag electrode was evaporated on the ZnMgO layers.

Characterizations

The morphologies of the QD films were observed by a scanning electron microscope (SEM, Verios G4). Time-resolved photoluminescence (TRPL) spectra were acquired by a fluorescence lifetime measurement instrument (Horiba TempPro-01). Electroluminescence (EL) spectra were measured using an Ocean Optics USB2000+ spectrometer. The EL characteristics were measured on a Keithley 4200-SCS semiconductor characterization system.

RESULTS AND DISCUSSION

As shown in Fig. 1a, in order to adapt to the environment, compact and ordered arrays of microstructures exist widely in nature, which can thus enhance the performance of themselves [14,22,23]. Fig. 1b, e schematically present the distribution of QDs in ordered and disordered monolayers, respectively. If the stacking of QDs is disordered, this layer may be non-continuous with a number of tiny voids and cracks. By contrast, the compact and ordered arrangement for QDs can effectively avoid voids and interstitial spaces, thus decreasing the leakage current pas-

Figure 1 (a) Examples of a wide range of compact and ordered microstructures in nature. (b, e) Schematic illustration of the QDs in ordered and disordered monolayers, respectively. (c) π -A isotherm of QD spreading on the surface of deionized water. (d, g) SEM images of SC-based QD films and a homogeneous QD monolayer by adopting the LB technique, respectively. (f) Fluorescence microscopy image of a homogeneous QD monolayer.

sing through the QD film under the external bias.

Herein, a compact and ordered QD monolayer is fabricated by adopting the LB technique that can precisely control the assembly process of QDs. A known quantity of QDs are dispersed in hexane and spread on the surface of deionized water. At this time, a QD monolayer is in a two-dimensional gaseous phase without an external force, where QDs can randomly distribute on the surface of deionized water. Then, its surface pressure increases gradually when the barriers are closed to compress this monolayer with a very slow speed that supports enough time for QDs to assemble. The QD monolayer gradually transforms a gaseous phase to a two-dimensional liquid phase when the QDs touch each other. Admittedly, compact and ordered arrangement allows more particles to exist in a smaller space. Therefore, the QDs will spontaneously assemble into compact and ordered arrays when the space between two barriers becomes smaller. Further compression results in a two-dimensional solid phase where the QDs cannot move anymore due to the lack of sufficient space. However, the monolayer will collapse when the surface pressure exceeds the range that the QDs can withstand [24]. The formation of the QD monolayer was observed *in situ* during compression by an optical microscope (see Fig. S1 in the Supplementary information). The typical surface pressure-surface area (π -A) isotherm curve obtained during compression allows monitoring of this process (Fig. 1c). There exists no interaction between the QDs in the beginning, whereas further compression promotes closing of the gaps as well as formation of the compact and ordered monolayer. The abscissa of the tangent line of the curve shows that a 11-nm-diameter QD yields an area of $\sim 105 \text{ nm}^2$, which is close to the projected area of it (95 nm^2). Additionally, this also serves

as further evidence of the compact arrangement between the QDs (the high-resolution transmission electron microscopy image of the QDs is shown in Fig. S2). Obviously, the monolayer collapses at a surface pressure of 38 mN m^{-1} . Therefore, a surface pressure of 25 mN m^{-1} is chosen to transfer the QDs to a substrate in order to form a compact monolayer without collapse. Since the surface organic ligands covering the QDs are hydrophobic, the transfer from water to a polymer substrate is energetically favorable. The optical microscopy image of the QD monolayer is presented in Fig. 1f, showing a voidless and homogeneous morphology over the entire area. An atomic force microscopy (AFM) image of the as-prepared QD monolayer reveals a smooth and homogeneous film fabricated by using the LB technique (the root-mean-square surface roughness is estimated to be 1.88 nm , Fig. S3). Besides, the AFM image and profile of the QD monolayer provide further evidence for monolayered film formation (Fig. S4). Moreover, SEM images of an SC-based QD film and a homogeneous QD monolayer by adopting the LB technique are presented in Fig. 1d, g, respectively. Apparently, the SEM observations demonstrate that voids and interstitial spaces are present for the QDs film based on SC, while the monolayer formed with the LB technique exhibits an ordered and homogeneous surface morphology without obvious voids and interstitial spaces.

Fig. 2a presents the schematic illustration of the fabrication process of an LB-based QD monolayer. Because the LB technique is an organic-solvent-free method, QD multilayers can be achieved through repetitive transferring of a substrate. Fig. 2b illustrates the PL spectra of LB-based QD mono- and multi-layers whose emission peak does not shift as the thickness increases. The inset of Fig. 2b shows a linear relationship

Figure 2 (a) Schematic illustration of the fabrication process of an LB-based QD monolayer. (b) PL spectra of QD films with different numbers of QD layers. Inset: PL emission peak intensity scales linearly with the number of deposited monolayers. (c) UV-vis absorption spectra and (d) TRPL of the LB-based QD films with different thicknesses.

between PL intensity and the number of deposited layers, revealing that the QD monolayers are well homogeneous at a given surface pressure [19,25]. Meanwhile, ultraviolet-visible (UV-vis) absorption and TRPL spectra of LB-based QD films with different thicknesses presented in Fig. 2c, d exhibit the same trend with their PL spectra. Slight shift of peak positions was observed with different thicknesses in UV-vis absorption spectra, due to that the increase in thickness facilitates the interaction between QD layers. Based on the homogeneity of the LB-based QD monolayer, its optical properties change linearly with the increase of the number of layers.

We fabricated QLEDs by using the LB technique and investigated the impact of QDs stacking on the EL performance. We attempted to transfer a QD monolayer directly to the underlying layers by employing an HL method. The underlying layers can avoid the damage from deionized water because the residual water volatilizes immediately in a vacuum after transferring. The schematic architecture of QLEDs is presented in Fig. 3a (the following QLEDs have the same architecture except for emission layer). Fig. 3b, c show the corresponding energy level alignment and CIE coordinates of the devices. The CIE coordinate of the LB-based red QLEDs is (0.69, 0.31), indicating that the transferring does not change the wavelength of QDs' emission. The EL spectra for the LB-based and SC-based QLEDs presented in Fig. S5 suggest that the EL spectra of them have the same peak position. Noteworthily, the full width at half maximum (FWHM) for the LB-based QLEDs is narrower than that obtained by adopting the SC technique due to the improvement

of the device color purity caused by more compact and ordered QDs stacking preventing emission from occurring outside the QD monolayer. The current density-voltage (J - V), luminance-voltage (L - V) and the external quantum efficiency-voltage (EQE- V) curves for the SC-based QLEDs and LB-based QLEDs are depicted in Fig. 3d, e. In comparison with the SC-based device, it should be pointed out that, the leakage current of LB-based QLEDs is significantly reduced due to the compact and ordered QD monolayer. The device fabricated by the LB technique, whose peak EQE and current efficiency (CE) reached 19.0% and 23.0 cd A⁻¹, respectively, has approximately 15% improvement in comparison with those (16.5% and 21.6 cd A⁻¹) of the SC-based device (CE- L curve is presented in Fig. S6 and the performances of these devices are listed in detail in Table S1). In addition, Fig. 3f indicates that the lifetime of the LB-based QLEDs is nearly 1.8 times that of the SC-based device. Besides, the lifetime of the device is calculated by the following expression:

$$L_0^n \times T_{95} = \text{constant},$$

where, the 95% lifetime value (T_{95}) of the device represents the time it takes for the actual luminance to decay to 95% of the initial luminance (L_0). n , as an acceleration factor, is set to be 1.7. Calculated by using the above expression, the T_{95} lifetime at 100 cd m⁻² for the LB-based and SC-based QLEDs are 13,324 and 7276 h, respectively.

In terms of QLEDs, the suitable thickness of QD layers is important for achieving carrier recombination balance and high device performance [7]. The LB technique, as a precise method

Figure 3 Schematic illustration of (a) the QLED structure and (b) energy band alignment. (c) CIE coordinates for the LB-based QLEDs. (d) J - V and L - V curves. Inset: leakage current for the SC-based QLEDs and LB-based QLEDs. (e) EQE- L and (f) lifetime curves for the SC-based QLEDs and LB-based QLEDs. (g) EL spectra, (h) EQE- V , (i) J - V and L - V curves for QLEDs with different thicknesses of QD layers.

for controlling the number of QD layers, is a favorable approach to investigate the impact of the thickness of QD layers on device performance. According to Fig. 3g, the EL spectra of the LB-based QLEDs indicate that the emission peak keeps the same position when the QD thickness increases. The EQE-*V* curves for QLEDs with different thicknesses of QD layers are presented in Fig. 3h. The EQE values of the device with three QD monolayers reach the maximum (14.6%) at 2.8 V, while those with single and two QD monolayers show higher EQE values of 18.3% and 17.3%, respectively. For compact QD stacking and low efficiency of ‘dot-dot’ charge transport, it is difficult for carriers to transport between QD monolayers, which lowers the probability of carrier recombination, thus obtaining the low efficiency for the device with large QD thickness [26]. Fig. 3i shows the corresponding *J-V* and *L-V* curves for these devices. At the same luminance, larger current is needed for QLEDs with thicker QD layers due to an increase in resistance. As a result, for the device with a QD monolayer it is not difficult to reach high luminance (over 1000 cd m⁻²) at 2 V, while a bias of 3.6 V is required for the device with three QD monolayers to achieve the same luminance. In addition, the device with larger thickness of emission layer needs to be driven at higher bias to realize carrier recombination in QDs, generating the increase of turn-on voltage (the performances of these devices are listed in detail in Table S2).

In order to realize commercial application for QLEDs in

general illumination and displays, white QLEDs should be developed [21,27–29]. Till present, there are two main methods to fabricate white QLEDs, namely, red-green-blue (RGB) QDs mixing and RGB QD layers stacking. However, compared with the stacking strategy, the former is difficult to optimize the mixing ratio of RGB-QDs because of the different sizes of QDs and the variation of their quantum yields. Moreover, QLEDs with mixed-RGB QD films can only obtain white emission at a relatively high bias due to the existence of Förster resonance energy transfer (FRET) between QDs with different sizes. The CIE coordinates of the white device would gradually change because of the different lifetimes of red, green and blue QDs. The QLEDs with stacked QD monolayers are capable of obtaining white emission in a wide range of brightness from hundreds to tens of thousands cd m⁻² [30]. Generally, for white QLEDs with stacked QD layers, buffer layers are needed to be inserted between QD layers in order to avoid interlayer mixing [21]. Due to the distinct advantage of LB technique, namely, organic-solvent-free environment and precise control of the number of layers and layering sequence, buffer layers are removed and white QLEDs with stacked QD monolayers could be realized based on repetitive transferring of QD monolayers with different colors. As shown in Fig. S7, we meticulously optimized the thickness and sequence of the emission layers to acquire the optimal white device. The optimal device architecture and the energy level alignment of the white device are

Figure 4 Schematic illustration of (a) white QLEDs structure and (b) energy band alignment. (c) CIE coordinates for LB-based QLEDs. (d) EL spectra, (e) *J-V* and *L-V* and (f) EQE-*V* and CE-*V* characteristics for white QLEDs. (g, h) Photographs of the large-size and homogeneous red and green QD monolayers, respectively.

Table 1 Device performances of the white QLEDs at various voltages

V_T (V)	L (cd m ⁻²)	EQE (%)	CE (cd A ⁻¹)	CIE (x, y)
2.5	259	6.4	10.2	(0.42, 0.42)
3.0	1394	6.7	10.7	(0.38, 0.38)
3.5	3123	6.6	10.6	(0.35, 0.34)
4.0	7588	6.4	10.3	(0.34, 0.33)
4.5	11505	6.2	10.0	(0.34, 0.32)
5.0	17719	5.9	9.4	(0.33, 0.32)
5.5	22004	5.6	9.1	(0.33, 0.32)
6.0	27803	5.3	8.5	(0.32, 0.32)
6.5	31585	5.1	8.2	(0.32, 0.32)
7.0	37020	4.8	7.8	(0.32, 0.33)

presented in Fig. 4a, b, respectively. Fig. 4c, d show the corresponding CIE coordinates and EL spectra of the white QLEDs with stacked QD monolayers, respectively. The device presents white emission with CIE coordinate of (0.34, 0.33) at a low voltage of only 4 V, which is close to the ideal white point (0.33, 0.33). Low turn-on voltage is achieved because there is no buffer layer between the QD layers. The J - V and L - V curves of this white device are depicted in Fig. 4e. The EQE- V and CE- V curves of it presented in Fig. 4f show that the maximum EQE and CE are 6.7% and 10.7 cd A⁻¹, respectively. The device performances of the white QLEDs at various voltages are detailed in Table 1. In addition, a large-area monolayer was readily realized by adopting the LB technique with precise control of the surface pressure during the packing process of a monolayer. As shown in Fig. 4g, h, we successfully fabricated large-size and ultra-homogeneous red and green QD monolayers on glass substrates with the size of 9 cm × 5 cm.

CONCLUSIONS

To conclude, a compact and ordered QD monolayer with an ultra-homogeneous surface morphology and highly ordered QD stacking was fabricated with the LB technique. We successfully transferred an LB-based QD monolayer from the surface of water to the underlying hole transport layer of the device by a direct HL method. The LB-based device exhibited high performances with EQE of 19.0% and lifetime ($T_{95}@100$ cd m⁻²) of 13,324 h. In comparison with the SC-based device, the EQE and lifetime of the device were improved by 15% and 183% because the compact and ordered QD monolayer significantly decreased the leakage current. The results imply that a single QD monolayer is the most suitable thickness for red QLEDs. In addition, as an organic-solvent-free method, the LB technique could be used for fabricating white QLEDs with stacked RGB QD monolayers. Besides, pure white emission located in CIE coordinate of (0.34, 0.33) could be obtained at a low voltage of 4 V. Finally, we fabricated an ultra-homogeneous large-area QD monolayer on a rectangular glass substrate with a size of 9 cm × 5 cm. The LB-HL strategy for realizing an ultra-homogeneous and highly ordered QD monolayer could pave a promising way for commercialization of QLEDs in lighting and display fields.

Received 6 August 2021; accepted 9 September 2021;
published online 19 October 2021

- Colvin VL, Schlamp MC, Alivisatos AP. Light-emitting diodes made from cadmium selenide nanocrystals and a semiconducting polymer. *Nature*, 1994, 370: 354–357
- Shirasaki Y, Supran GJ, Bawendi MG, *et al.* Emergence of colloidal quantum-dot light-emitting technologies. *Nat Photon*, 2013, 7: 13–23
- Murray CB, Norris DJ, Bawendi MG. Synthesis and characterization of nearly monodisperse CdE (E = sulfur, selenium, tellurium) semiconductor nanocrystallites. *J Am Chem Soc*, 1993, 115: 8706–8715
- Kim TH, Chung DY, Ku JY, *et al.* Heterogeneous stacking of nanodot monolayers by dry pick-and-place transfer and its applications in quantum dot light-emitting diodes. *Nat Commun*, 2013, 4: 2637–2645
- Zhao J, Zhang J, Jiang C, *et al.* Electroluminescence from isolated CdSe/ZnS quantum dots in multilayered light-emitting diodes. *J Appl Phys*, 2004, 96: 3206–3210
- Coe S, Woo WK, Bawendi M, *et al.* Electroluminescence from single monolayers of nanocrystals in molecular organic devices. *Nature*, 2002, 420: 800–803
- Sun Q, Wang YA, Li LS, *et al.* Bright, multicoloured light-emitting diodes based on quantum dots. *Nat Photon*, 2007, 1: 717–722
- Liu Y, Han F, Li F, *et al.* Inkjet-printed unclonable quantum dot fluorescent anti-counterfeiting labels with artificial intelligence authentication. *Nat Commun*, 2019, 10: 2409–2417
- Won YH, Cho O, Kim T, *et al.* Highly efficient and stable InP/ZnSe/ZnS quantum dot light-emitting diodes. *Nature*, 2019, 575: 634–638
- Tian F, Zhu Y, Xu Z, *et al.* Achieving highly efficient and stable quantum dot light-emitting diodes with interface modification. *IEEE Electron Device Lett*, 2020, 41: 1384–1387
- Zhu Y, Hu H, Liu Y, *et al.* All-solution-processed high-performance quantum dot light emitting devices employing an inorganic thiocyanate as hole injection layer. *Org Electron*, 2019, 70: 279–285
- Liu Y, Li F, Xu Z, *et al.* Efficient all-solution processed quantum dot light emitting diodes based on inkjet printing technique. *ACS Appl Mater Interfaces*, 2017, 9: 25506–25512
- Bae WG, Kim HN, Kim D, *et al.* Scalable multiscale patterned structures inspired by nature: The role of hierarchy. *Adv Mater*, 2014, 26: 675–700
- Sanchez C, Arribart H, Giraud Guille MM. Biomimetic and bioinspiration as tools for the design of innovative materials and systems. *Nat Mater*, 2005, 4: 277–288
- Luo D, Qin X, Song Q, *et al.* Ordered superparticles with an enhanced photoelectric effect by sub-nanometer interparticle distance. *Adv Funct Mater*, 2017, 27: 1701982–1701990
- Ariga K, Yamauchi Y, Mori T, *et al.* What can be done with the Langmuir-Blodgett method? Recent developments and its critical role in materials science. *Adv Mater*, 2013, 25: 6477–6512
- Bourvon H, Le Calvez S, Kanaan H, *et al.* Langmuir-Schaeffer monolayers of colloidal nanocrystals for cost-efficient quantum dot light-emitting diodes. *Adv Mater*, 2012, 24: 4414–4418
- Wang J, Yan H, Liu Z, *et al.* Langmuir-Blodgett self-assembly of ultrathin graphene quantum dot films with modulated optical properties. *Nanoscale*, 2018, 10: 19612–19620
- Lambert K, Capek RK, Bodnarchuk MI, *et al.* Langmuir-Schaeffer deposition of quantum dot multilayers. *Langmuir*, 2010, 26: 7732–7736
- Crawford NF, Leblanc RM. CdSe and CdSe(ZnS) quantum dots in 2D: A Langmuir monolayer approach. *Coord Chem Rev*, 2014, 263–264: 13–24
- Lee KH, Han CY, Jang EP, *et al.* Full-color capable light-emitting diodes based on solution-processed quantum dot layer stacking. *Nanoscale*, 2018, 10: 6300–6305
- Liu K, Jiang L. Bio-inspired design of multiscale structures for function integration. *Nano Today*, 2011, 6: 155–175
- Sun M, Watson GS, Zheng Y, *et al.* Wetting properties on nanostructured surfaces of cicada wings. *J Exp Biol*, 2009, 212: 3148–3155
- Lambert K, Justo Y, Kamal JS, *et al.* Phase transitions in quantum-dot Langmuir films. *Angew Chem Int Ed*, 2011, 50: 12058–12061
- Xu J, Ji X, Gattás-Asfura KM, *et al.* Langmuir and Langmuir-Blodgett films of quantum dots. *Colloids Surf A-Physicochem Eng Aspects*, 2006, 284–285: 35–42

- 26 Chaudhary S, Ozkan M, Chan WCW. Trilayer hybrid polymer-quantum dot light-emitting diodes. *Appl Phys Lett*, 2004, 84: 2925–2927
- 27 Zhu Y, Xu R, Zhou Y, *et al.* Ultrahighly efficient white quantum dot light-emitting diodes operating at low voltage. *Adv Opt Mater*, 2020, 8: 2001479–2001486
- 28 Acharya KP, Titov A, Hyvonen J, *et al.* Full color-tunable vertically stacked quantum dot light emitting diodes for next-generation displays and lighting. *Nanoscale*, 2020, 12: 24403–24410
- 29 Wang L, Pan J, Qian J, *et al.* A highly efficient white quantum dot light-emitting diode employing magnesium doped zinc oxide as the electron transport layer based on bilayered quantum dot layers. *J Mater Chem C*, 2018, 6: 8099–8104
- 30 Zhang H, Su Q, Sun Y, *et al.* Efficient and color stable white quantum-dot light-emitting diodes with external quantum efficiency over 23%. *Adv Opt Mater*, 2018, 6: 1800354–1800361

Acknowledgements This work was supported by the National Natural Science Foundation of China (62075043), and Fujian Science & Technology Innovation Laboratory for Optoelectronic Information of China (2021ZZ126).

Author contributions Li F, Hu H and Guo T conceived and supervised the project; Zhao D and Zhu Y wrote the draft of the manuscript; Zheng Y and Meng T prepared some of the samples; Jing J performed the lifetime measurements; Chen X carried out the UV-Vis absorption measurements; Gao H, Mao C and Zheng W participated in the discussion and analysis of the data.

Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Supporting data are available in the online version of the paper.

Denglin Zhao is currently a master degree candidate under the supervision of Prof. Fushan Li at Fuzhou University and Fujian Science & Technology Innovation Laboratory for Optoelectronic Information of China. His current research interests include QDs and their optoelectronic applications.

Fushan Li has been a distinguished professor at the School of Physics and Information Engineering of Fuzhou University since October, 2009. Prof. Li's research speciality is nanoelectronic devices and fabrication technology, especially on the optoelectronic properties of various nanomaterials for application in next-generation light-emitting devices, photovoltaic, and memristors.

具有超均匀和高度有序量子点单层的高效量子点发光二极管

赵等临^{1,2}, 郑悦婷¹, 孟汀涛¹, 朱阳斌¹, 井继鹏¹, 陈祥¹, 高宏锦¹, 毛超民¹, 郑文晨¹, 胡海龙¹, 郭太良^{1,2}, 李福山^{1,2*}

摘要 对于采用旋涂法制备量子点层的传统量子点发光二极管(QLEDs)而言, 由于量子点有序性低, 量子点薄膜不可避免地出现空隙, 导致器件在偏压下产生漏电流. 本研究中, 我们利用Langmuir-Blodgett(LB)技术制备了超均匀、高度有序的量子点单层, 并且采用水平提拉法(HL)将量子点单层作为发光层转移到红色QLEDs上, 器件表现出了高性能, 其外量子效率(EQE)为19.0%, 寿命($T_{95}@100\text{ cd m}^{-2}$)为13,324 h. 由于致密有序的量子点单层减少了漏电流, EQE和寿命分别提高了15%和183%. 此外, LB技术是一种无有机溶剂气氛的方法, 避免了层间互溶, 同时也可以精确控制量子点薄膜的厚度, 因此所制备的叠层白光QLEDs可以在4 V的低电压下获得白光. 最后, 我们成功地在 $9\text{ cm} \times 5\text{ cm}$ 的矩形基板上制备了超均匀大面积量子点单层, 表明LB-HL方法具有良好的尺寸可扩展性.