

Toward Full-Color Electroluminescent Quantum Dot Displays

Jiwoong Yang,[†] Moon Kee Choi,[†] U Jeong Yang, Seo Young Kim, Young Seong Kim, Jeong Hyun Kim, Dae-Hyeong Kim,* and Taeghwan Hyeon*



Cite This: *Nano Lett.* 2021, 21, 26–33



Read Online

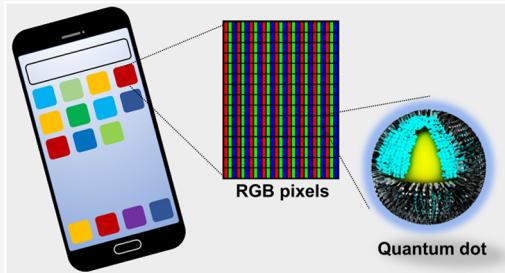
ACCESS |

Metrics & More

Article Recommendations

ABSTRACT: Colloidal quantum dots (QDs) exhibit unique characteristics such as facile color tunability, pure color emission with extremely narrow bandwidths, high luminescence efficiency, and high photostability. In addition, quantum dot light-emitting diodes (QLEDs) feature bright electroluminescence, low turn-on voltage, and ultrathin form factor, making them a promising candidate for next-generation displays. To achieve the overarching goal of the full-color display based on the electroluminescence of QDs, however it is essential to enhance the performance of QLEDs further for each color (e.g., red, green, and blue; RGB) and develop novel techniques for patterning RGB QD pixels without cross-contamination. Here, we present state-of-the-art material, process, and device technologies for full-color QLED-based displays. First, we highlight recent advances in the development of efficient red-, green-, and blue-monochromatic QLEDs. In particular, we focus on the progress of heavy-metal-free QLEDs. Then, we describe patterning techniques for individual RGB QDs to fabricate pixelated displays. Finally, we briefly summarize applications of such QLEDs, presenting the possibility of full-color QLED-based displays.

KEYWORDS: quantum dot, light-emitting diode, full-color display, high-resolution patterning, electroluminescence



Quantum-confined semiconductor nanocrystals, known as quantum dots (QDs), exhibit superior properties to conventional light-emitting semiconductors.¹ For instance, QDs show tunable band gaps owing to their quantum confinement effect, superior color purity with narrow emission spectra, bright photoluminescence with high quantum yield, and high photo/air stability.^{2–5} These advantages support the development of high-performance quantum dot light-emitting diodes (QLEDs) in which QDs serve as light-emitting layers by electroluminescence (EL).^{6–12} Since the first demonstration of EL from CdSe QDs,¹² immense effort has been made to develop efficient QLEDs both in academia and industry. Displays using QDs have already been commercialized, although they are based on the photoluminescence mechanism, and the EL-based QD devices could not be mass-produced yet. Recently, however, notable progress on EL-based QLEDs has been made. For example, Cd-chalcogenide QLEDs,^{13–15} blue-emitting ZnSeTe QLEDs,¹⁶ and red-emitting InP QLEDs¹⁷ showed high external quantum efficiency (EQE) similar to their theoretical limits as well as high brightness with low turn-on voltages. Moreover, QLEDs could be fabricated as an ultrathin-type device whose entire thickness is $<3\text{ }\mu\text{m}$ including encapsulation layers ($\sim 200\text{ nm}$ without the encapsulation layer). Such ultrathin devices could be fabricated due to the material stability of QDs and enabled various form factors of QLEDs including bendable and foldable devices.¹⁸ These distinctive features promise the potential of QLEDs for future displays, which can become key

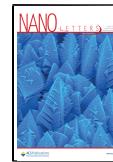
components of next-generation electronics, including wearable/mobile ones.¹⁹

Meanwhile, displays consist of multiplexed pixel arrays, and the unit pixel consists of subpixels with different colors such as red, green, and blue (RGB). Recently, the resolution of commercial displays has been rapidly increasing. For example, a modern 8K television has 7680×4320 pixels, corresponding to 160 pixels per inch (ppi) for 50" panels, and the latest mobile phones employ high-resolution displays (e.g., 563 ppi for Galaxy S20). High-resolution displays are key components for virtual reality (VR) and/or augmented reality (AR) applications. General human eyes can resolve ~ 60 pixels per degree with a field of view of $>120^\circ$, indicating that at least 7200×7200 pixels are needed to match the resolution of human eyes in VR/AR applications. Approximately, a resolution of 3000 ppi is required to fabricate these megapixels on a 2.5" glass. Therefore, the development of high-resolution QD patterning methods, which would enable the fabrication of fine pixels for EL devices, is essential. Moreover,

Received: September 30, 2020

Revised: November 23, 2020

Published: December 1, 2020



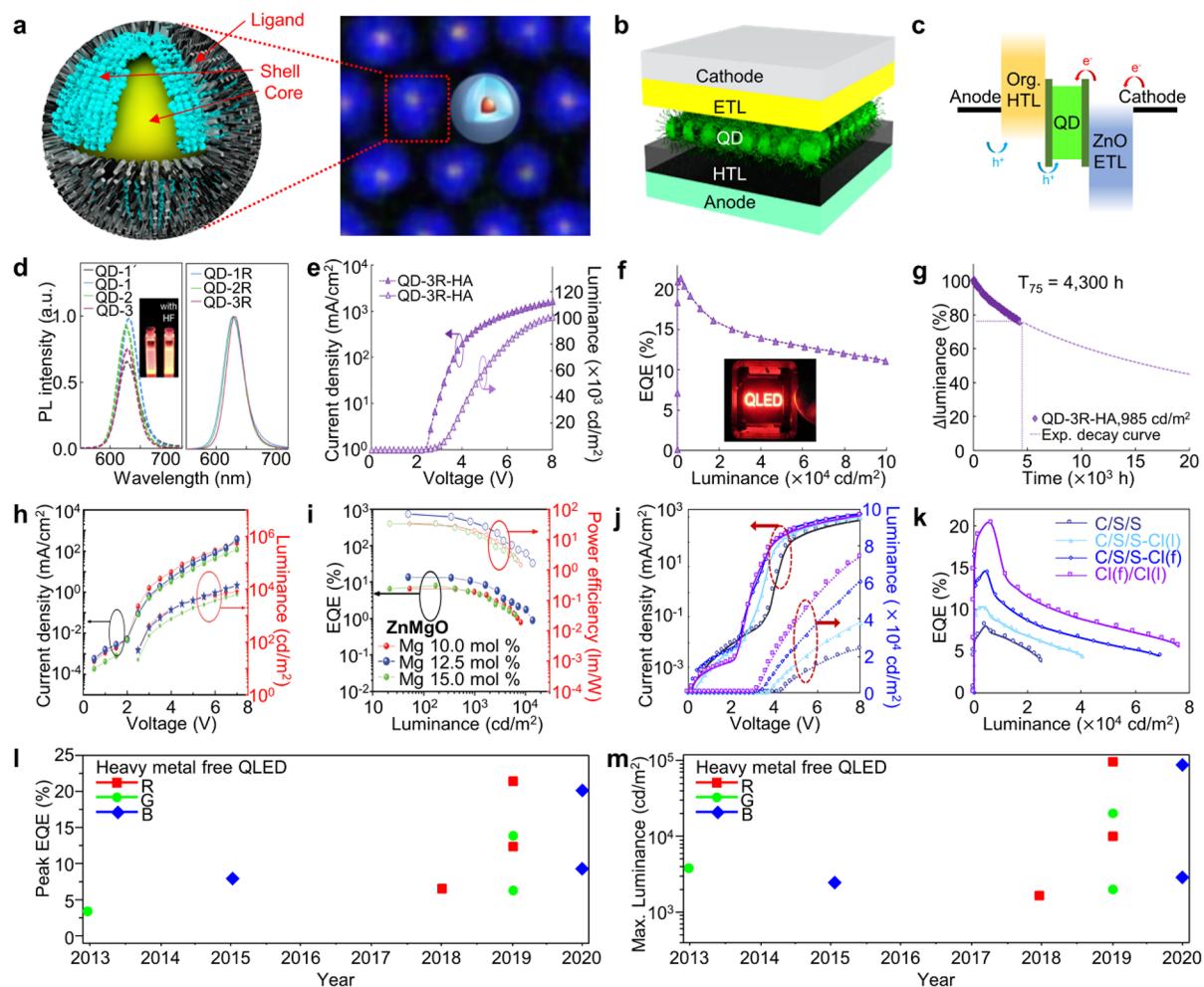


Figure 1. Material and device designs for efficient QLEDs. (a) Schematic and electron diffraction spectroscopy mapping of core–shell structured QDs. Right image adapted with permission from ref 17. Copyright 2019 NPG. (b,c) Device structure and energy diagram of QLEDs. (d–g) Photoluminescence spectra, J – V – L curve, EQE versus luminance, and lifetime measurement of InP/ZnSe/ZnS red QLEDs. Reprinted with permission from ref 17. Copyright 2019 NPG. (h,i) J – V – L characteristics and EQE and power efficiency of InP-based green QLEDs according to ZnMgO ETL modifications. Reprinted with permission from ref 31. Copyright 2019 the RSC. (j,k) J – V – L characteristics and EQE of ZnSeTe blue QLEDs. Reprinted with permission from ref 16. Copyright 2019 NPG. (l,m) Progress in the performance of heavy-metal-free QLEDs in terms of peak EQE and maximum luminance. Data for panels l and m are taken from the following references: (i) red QLEDs, 17, 29, 30; (ii) green QLEDs, 31–33; and (iii) blue QLEDs, 16, 34, 35.

it becomes important to develop device techniques that efficiently utilize pixelated QD layers.

Here, we present the recent progress on material, process, and device technologies for QLEDs toward full-color EL displays. First, we underline the advancement of high-efficiency QLEDs with monochromatic emissions (i.e., red-, green-, and blue-emitting QLEDs). Particularly, we discuss the recent progress of heavy-metal-free QLED technologies. Then, we review various approaches for patterning individual RGB QDs on pixelated display panels. Finally, we summarize various application examples of multicolored and/or pixelated QLEDs and then assess the feasibility of full-color QD displays.

■ DEVELOPMENT OF EFFICIENT QLEDS

One of the most important prerequisites to establish full-color EL-based QD displays is the development of efficient single color QLEDs (e.g., R, G, B QLED). Since the first report on EL devices using CdSe QDs,¹² tremendous efforts have been made to discover new ways for synthesizing QDs and designing device structures to fabricate highly efficient and

stable QLEDs. Because of the comprehensive optimization of both QD synthesis and device structures, outstanding EQEs from Cd-chalcogenide-based QLEDs were reported, similar to their theoretical limits.^{13–15} A wide range of analysis methods has been adopted to understand the operation and degradation mechanism of QLEDs.

High-quality type I core/shell QDs^{20,21} (Figure 1a) are preferred for EL devices because of their excellent stability and high-photoluminescence quantum yield (PLQY, typically 70–100% for CdSe/ZnS QDs), which is directly related to the EQE of EL emissions (EQE \propto PLQY). In addition to PLQY, carrier injection and recombination dynamics should be taken into consideration. For example, the thickness and composition of the shell materials heavily affect the characteristics of QLEDs.^{20,22,23} An unbalanced carrier injection easily charges the QD layers in QLEDs, thus decreasing the light emission from QDs because of various nonradiative recombination pathways such as Auger recombination. QDs with thick shell are less influenced by the charge fluctuation and interparticle energy transfer. As another important design factor, the

composition of the core/shell interfaces severely affects the charge carrier injection and recombination process. For example, core/shell QDs with gradually alloyed intermediate shell are preferred because of the low energy barrier for charge carrier injection.^{20,23} Recently, the role of the ligand molecules during EL operations has been studied.^{17,24} It has been suggested that short-chain ligands can improve charge injection and electrochemically stable ligands can enhance device lifetime.

The design of a device structure also plays a significant role in fabricating highly efficient QLEDs. Figure 1b describes the general structure of QLEDs, which consist of light-emitting QD layers sandwiched between charge transport layers (electron and hole) and electrodes (transparent anode and cathode). ZnO nanoparticle (NP)-based electron transport layers (ETLs) and the organic hole transport layers (HTLs) are commonly employed. To design highly efficient and stable QLEDs, it is important to optimize the composition and the thickness of the charge transport layers; the injection of both electrons and holes into QDs should be finely controlled for efficient light emission (Figure 1c). The ETLs and HTLs should exhibit high electron and hole mobilities, respectively, to efficiently transport each charge carrier into QDs. Moreover, it turns out that not only the injection rate but also the balance²² and sequences²⁵ between the injection of electrons and holes are critical. Unbalanced injection rates in QDs result in the luminance efficiency drop by charging QDs. In addition, recent works on the QLED degradation mechanism suggest that the unbalanced injection of holes and electrons can damage charge transport layers.^{26,27}

The aforementioned efficient QLEDs are mostly based on Cd-chalcogenide QDs. However, the development of heavy-metal-free QLEDs is essential for their wide and practical implementation.^{16,17,28–35} In most developed countries, the use of Cd-based compounds in consumer electronics is prohibited. Consequently, III–V InP-based QDs have emerged as the most promising alternatives to toxic Cd-chalcogenide QDs because their band gaps cover most of the visible spectra.^{17,28–33} Fortunately, most of the strategies used for Cd-chalcogenide QDs can be successfully adapted for heavy-metal-free QDs. Particularly, Samsung reported highly efficient red-emitting QLEDs using InP QDs with engineered core/shell structures and surface passivation (Figure 1d–g).¹⁷ The device showed an outstanding EQE of 21.4%, which is comparable to that of the best Cd-chalcogenide-based QLEDs (Figure 1f). In addition, auspicious progress has been made in the development of green-emitting InP QLEDs (Figure 1h,i).³¹ High-efficiency green-emitting InP QLEDs have been reported by multiple research groups.^{31–33} However, the fabrication of highly efficient blue-emitting QLEDs using InP QDs is challenging, presumably because of the difficulties in synthesizing high-quality blue-emitting InP QDs and the energy-level alignment of these QDs. To overcome this limitation, ZnSe- or ZnSeTe-based QDs have been introduced to obtain blue-light emission from heavy-metal-free QDs.^{16,34,35} Recently, blue-emitting heavy-metal-free QLEDs with an EQE of ~20% have been reported using ZnSeTe/ZnS QDs (Figure 1j,k).¹⁶ To fabricate efficient heavy-metal-free QLEDs, it is also important to improve charge injections into QDs within the device structure. For instance, Mg-doped ZnO NPs have been suggested in place of ZnO NPs as ETLs in heavy-metal-free QLEDs to match the charge balance in the QD layers.³¹ The peak EQE and maximum luminance values of several

remarkable heavy-metal-free QLEDs are summarized in Figure 1l,m. The representative characteristics of state-of-the-art heavy-metal-free RGB QLEDs are also shown in Table 1.

Table 1. Performance of Heavy-Metal-Free QLEDs Reported in the Literature

color	core QD	peak wavelength (nm)	QY (%)	max. EQE (%)	max. luminance (cd m ⁻²)	reference
red	InP	630	100	21.4	100 000	17
green	InP	531	82	13.6	13 900	31
blue	ZnSeTe	457	100	20.2	88 900	16

PATTERNING QDS FOR FABRICATION OF THE PIXEL ARRAYS

The highly efficient QLEDs discussed above are mostly for monochromatic EL devices.^{12–17} Because QDs are usually suspended in the solution, the spin-casting method is commonly used to fabricate QD films, either for single-color QDs or for their mixed state. However, for EL-based displays individual RGB QDs should be patterned on pixelated panels without cross-contamination between QDs of different colors. In addition, the resolution of commercial displays has been progressively increased, and the required resolution for future displays will be even higher. Therefore, it is important to develop a novel patterning technology that can fabricate submicron-sized pixels with high color purity. Various approaches have been developed to fabricate RGB QD pixel arrays, such as (i) photolithography, (ii) inkjet printing, and (iii) transfer printing.

The photolithography has been applied to fabricate RGB QD pixels.³⁶ In this case, QDs and photoresists are mixed in the solution phase and coated on the substrate as a thin film. Then, it is exposed to UV light through photomasks to generate patterns, and the unexposed parts are washed away by the solvent. This procedure is repeated to form RGB pixels on a single substrate. The primary advantage of this technique is that it can utilize most of the benefits of the conventional photolithography that is already well established by the modern semiconductor industry. However, due to the presence of the photoresist, the generated patterns inevitably contain organic residues, which can act as electrically insulating layers for charge transport into the QDs. To minimize the organic contents in the final QD patterns, novel photolithography processes that use inorganic ligand-photo acid generator complexes³⁷ (Figure 2a) or ligand cross-linkers³⁸ (Figure 2b,c) have been introduced.

Inkjet printing has been suggested as an alternative method to directly form a desired pattern using QD-based inks without any waste products (Figure 2d–g).^{39–41} For example, electrohydrodynamic jet printings can produce high-resolution QD patterns (Figure 2e).³⁹ The printed patterns using red and green QDs exhibit a uniform line thickness and a narrow width (~5 μm). Furthermore, by combining inkjet printing with 3D printing technique, QD pixels can be integrated on curved objects such as a contact lens (Figure 2g).⁴¹ However, the inks used in this process usually contain organic additives for improving the printing quality (e.g., better wettability and uniformity without the coffee-ring effect), and these additives hinder efficient charge injection into QDs. In addition, high-resolution inkjet printing of QDs is very challenging because

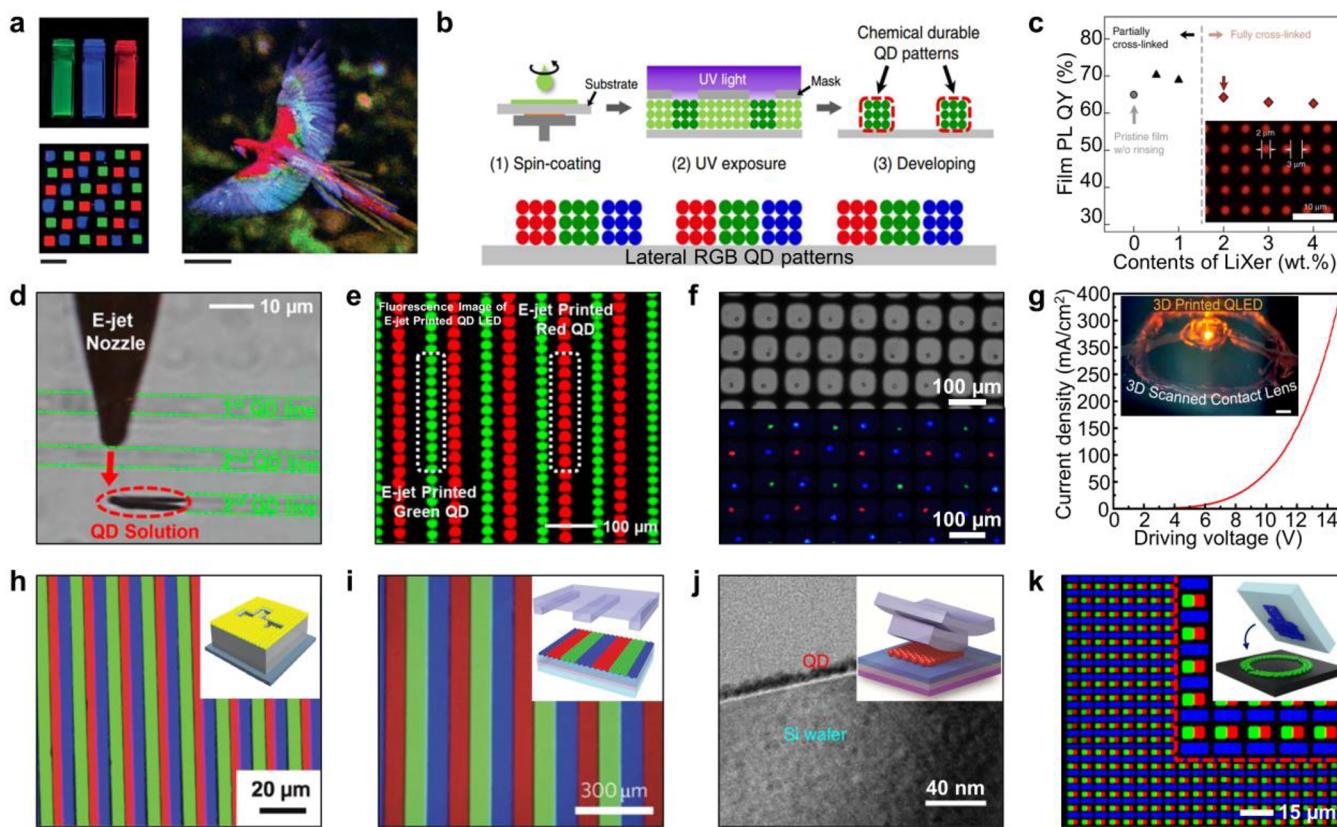


Figure 2. Patterning techniques for the full-color QLEDs. (a) Photopatterned all-inorganic QDs using direct optical lithography. Reprinted with permission from ref 37. Copyright 2017 AAAS. Scale bars, 5 mm. (b,c) Scheme illustration and film quantum yield of photo cross-linked QDs. Reprinted with permission from ref 38. Copyright 2020 NPG. (d,e) Optic image of electrohydrodynamic jet printing nozzle and fluorescence image of the printed red and green QDs.³⁹ (f) Optic and fluorescence image of inkjet printed salt-encapsulated RGB QDs.⁴⁰ (g) J - V characteristics of 3D printed QLEDs on contact lens.⁴¹ (h) Fluorescence image of RGB QD patterns fabricated with direct spin-casted QDs onto a structured stamp. Reprinted with permission from ref 44. Copyright 2012 Wiley. (i) Fluorescence image of RGB QD patterns using the pick-and-release transfer method. Reprinted with permission from ref 45. Copyright 2011 NPG. (j) SEM image of monolayer QD film transferred with sacrificial layer. Reprinted with permission from ref 46. Copyright 2013 NPG. (k) Fluorescence image of RGB QD pixels using intaglio transfer printing. Reprinted with permission from ref 18. Copyright 2015 NPG.

controlling the fluid dynamics of QD inks in the several-micrometer scale is difficult.

Transfer printing is a promising method for high-resolution QD patterning, particularly for EL device applications,^{18,42–46} because this method does not involve additional organic additives for the patterning procedure (Figure 2h–k). Viscoelastic stamps with low surface energy (e.g., perfluoropolyether⁴⁴ and poly(dimethylsiloxane)^{18,45,46} stamps) have been widely used for the transfer printing of QD films (Figure 2h).⁴⁴ The pick-and-release process, in which the QD film coated on the self-assembled monolayer-treated donor substrate is rapidly picked up with the structured stamp and transferred to the target substrate, is typically employed for patterning of RGB QDs (Figure 2i).⁴⁵ In addition, monolayered or vertically stacked multilayered QDs can be successfully transferred to the target substrate with the help of a sacrificial layer including a poly(vinyl alcohol) film (Figure 2j).⁴⁶ Recently, the intaglio transfer printing method has been introduced for the formation of submicron- and several-micron-sized precise QD pixels (Figure 2k).¹⁸ In the intaglio transfer printing, the QD patterns are formed when a QD film picked up from a donor substrate using a flat stamp is transferred on the intaglio trench having high surface energy. The crack propagation of QD film is inhibited at the edges of

the intaglio trench, which results in high yield and accurate pattern formation.

PIXELATED/PATTERNEDE QLEDS

For full-color displays, the RGB subpixels needs to be individually addressed. Although most previous research on QLEDs has focused on the improvement of the EL efficiency for the monochromatic light emission, there is an increasing demand for the development of full-color QLED devices for their practical applications. Accordingly, there are ongoing efforts to implement full-color displays using the QD patterning techniques discussed in the previous chapter.

In 2013, for the first time Samsung Advanced Institute of Technology demonstrated a 4"-scale flexible full-color QD display by combining 413 ppi RGB QLEDs with an oxide transistor array (Figure 3a).⁴⁵ Each monochromatic QD pixel was transfer-printed onto a 320×240 array of hafnium–indium–zinc oxide transistors. Because of the external pressure applied during the printing process, the transfer-printed QD layers exhibit low voids and excellent morphology, which enhances the facile charge transport into QDs. Accordingly, the flexible QLEDs show stable luminescence efficiency and current density even at various bending radii of curvature (Figure 3b). There have been continuous efforts to achieve the multicolor display by other QD patterning methods including

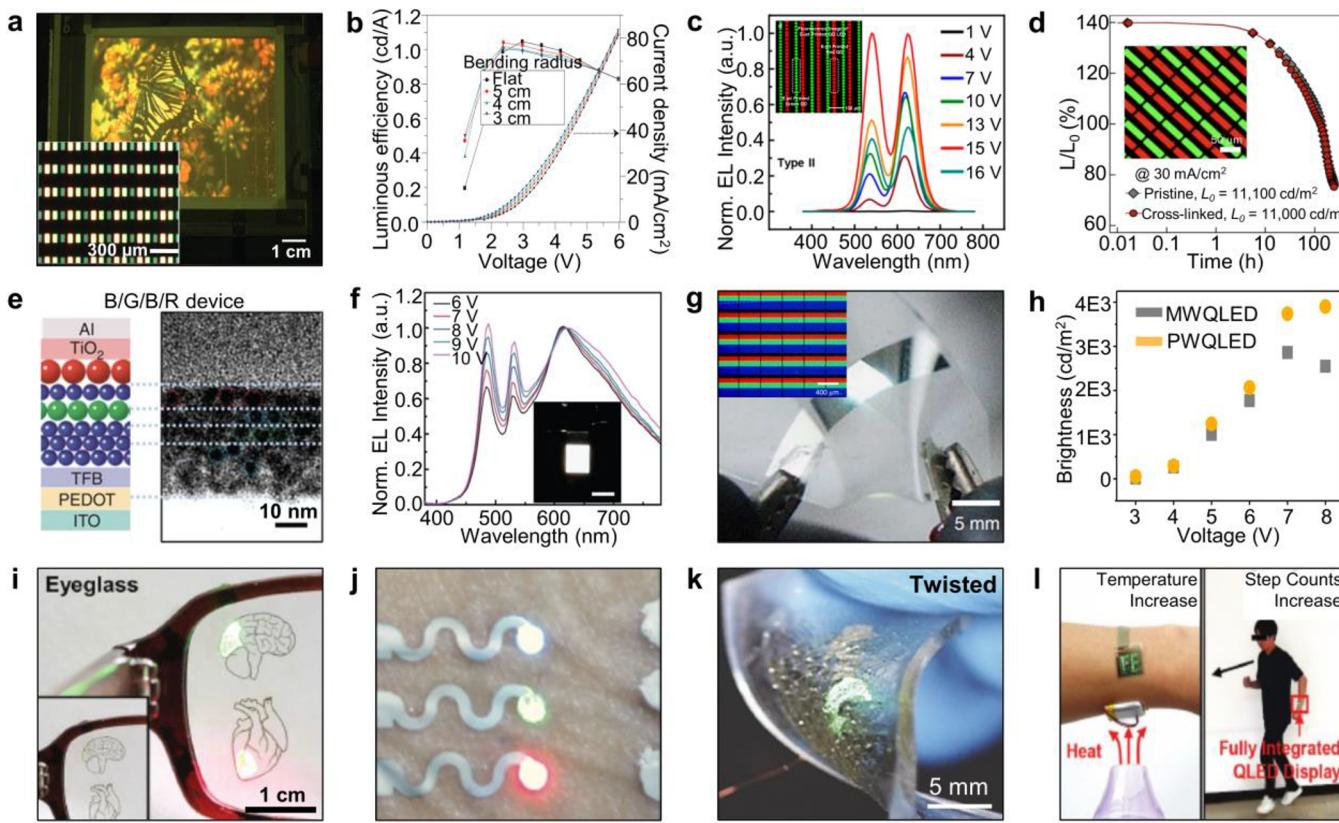


Figure 3. Patterned QLEDs for the next generation display. (a,b) Optic image and $J-V-L$ characteristics of full-color QLEDs. Inset of (a) shows a magnified image of RGB pixels. Reprinted with permission from ref 45. Copyright 2011 NPG. (c) EL spectra of inkjet-printed QLEDs according to the applied bias.³⁹ (d) Lifetime of photopatterned QLEDs. Reprinted with permission from ref 38. Copyright 2020 NPG. (e,f) Schematic, cross-sectional TEM image, EL spectra of white QLED by layer-by-layer transfer printing. Reprinted with permission from ref 46. Copyright 2013 NPG. (g) Photograph and optic image of flexible pixelated white QLEDs. Reprinted with permission from ref 18. Copyright 2015 NPG. (h) Brightness versus voltage of mixed white QLEDs and pixelated white QLEDs. Reprinted with permission from ref 18. Copyright 2015 NPG. (i,j) Ultrathin QLEDs attached on eyeglass (i) and human skin (j). Reprinted with permission from ref 48. Copyright 2018 Wiley (i). Reprinted with permission from ref 49. Copyright 2015 Wiley (j). (k) Ultrathin QLEDs integrated with wearable transparent touch sensors. Reprinted with permission from ref 50. Copyright 2017 Wiley. (l) Ultrathin QLEDs integrated with a temperature sensor and a step counting sensor. Reprinted with permission from ref 19. Copyright 2017 Wiley.

inkjet printing and/or photolithography. Figure 3c,d present pixelated QLEDs based on electrohydrodynamic jet printing³⁹ and organic-cross-linker based photolithography,³⁸ respectively. As shown in Figure 3c, an electrohydrodynamic jet-printed QLED exhibits various colors from green to red depending on the applied bias because emissions from heterogeneously patterned green and red QDs are superimposed.³⁹ In addition, photo-cross-linked pixelated QLEDs show stable device operation, which is comparable to the pristine single-colored QLEDs.³⁸

Efficient white QLEDs for indoor lighting or display backlight have been demonstrated based on patterned RGB QDs. When QDs with different band gaps are mixed into a single thin film to emit white light, the resulting emission can be red-shifted due to the nonradiative interparticle energy transfer.¹⁸ Energy transfer occurs through adjacent particles of different colors. Thus, to compensate for the light loss caused by interparticle energy transfer, the proportion of blue QDs with low luminous efficiency needs to be dramatically increased for white emission. Figure 3e,f show true-white QLEDs based on vertically stacked⁴⁶ and spatially patterned¹⁸ RGB QDs. Pixelated white QLEDs can exhibits higher luminance with a much lower proportion of blue QDs due to minimized interparticle energy transfer (Figure 3g).

Pixelated/patterned QLEDs can be manufactured on flexible and stretchable substrates instead of rigid ones, which facilitates wearable QLEDs.^{18,47–51} Choi et al. reported the tattoo-like wearable QLEDs using 1 μm thick polyethylene-epoxy encapsulation layers; active layers of QLEDs are located on a neutral mechanical plane and demonstrate stable emission even with external deformation.¹⁸ Patterned ultrathin QLEDs can easily be attached on the surfaces of various curvatures, including paper, eyeglasses, and human skin (Figure 3h–j).^{48,49} Furthermore, pixelated QLED arrays can be integrated with diverse electronic components (i.e., sensors, memories, actuators, and communication units),^{19,50,51} and can be utilized as signal output devices to display various information (Figure 3k,l).¹⁹

CONCLUSION

In summary, we have presented recent advancements of QLED technologies for full-color EL-based QD displays. Their unique properties, such as high color purity, high efficiency, and ultrathin form factors, promise the great potential of QLEDs. Moreover, the ultimate goal of the next-generation full-color QLED-based display has been getting closer by the recent development of the QD synthesis, device fabrication, and QD patterning technologies.

Rapid improvement of monochromatic QLEDs has led to Cd-chalcogenide-based red-, green-, and blue-emitting QLEDs with outstanding EQEs and stability.^{13–15} In addition, recent studies reported blue-emitting¹⁶ and red-emitting¹⁷ heavy-metal-free QLEDs with maximum EQEs, which are close to their theoretical limits. Various QD patterning methods have also been developed to fabricate fine RGB pixel arrays. These high-performance QLED technologies and ultrahigh-resolution patterning techniques reassure the high feasibility of their diverse applications such as AR/VR displays or 3D displays. Furthermore, some of the recent studies have started to consider the use of pixelated or patterned QD layers in QLEDs, which is also a significant step toward the successful fabrication of the EL-based full-color QD displays. Finally, QD displays can be seamlessly integrated with various electronic devices for system-level applications due to their high efficiency and ultrathin form factors. We envision that full-color EL-based QD displays will not only replace conventional displays but also facilitate the advancement of next-generation technologies such as VR/AR, the Internet of Things, and ubiquitous computing.

However, several challenges still remain for full-color QLED-based displays. For example, the efficiency of heavy-metal-free green-emitting QLEDs should be further improved, and the high-efficiency of heavy-metal-free QLEDs has only been achieved by the specific company (i.e., Samsung). In addition, it is ideal to develop heavy-metal-free QLEDs based on earth-abundant materials considering the rarity of indium (~0.1 ppm in the crust of the Earth). Fundamental understandings of the efficiency roll-off mechanisms are needed for the development of highly efficient and stable QLEDs with a longer device lifetime. In addition, most of the current QD patterning techniques are not optimized for large-scale EL devices. Developing organic additive-free printing techniques with large-scale uniformity is highly required. Finally, gaining further knowledge about the operation principles of high-resolution pixelated QLEDs is also needed.

AUTHOR INFORMATION

Corresponding Authors

Taeghwan Hyeon – Center for Nanoparticle Research, Institute for Basic Science (IBS), Seoul 08826, Republic of Korea; School of Chemical and Biological Engineering, Institute of Chemical Processes, Seoul National University (SNU), Seoul 08826, Republic of Korea;  orcid.org/0000-0001-5959-6257; Email: thyeon@snu.ac.kr

Dae-Hyeong Kim – Center for Nanoparticle Research, Institute for Basic Science (IBS), Seoul 08826, Republic of Korea; School of Chemical and Biological Engineering, Institute of Chemical Processes, Seoul National University (SNU), Seoul 08826, Republic of Korea;  orcid.org/0000-0002-4722-1893; Email: dkim98@snu.ac.kr

Authors

Jiwoong Yang – Department of Energy Science and Engineering, Daegu Gyeongbuk Institute of Science and Technology (DGIST), Daegu 42988, Republic of Korea;  orcid.org/0000-0002-2346-8197

Moon Kee Choi – Department of Materials Science and Engineering, Ulsan National Institute of Science and Technology (UNIST), Ulsan 44919, Republic of Korea;  orcid.org/0000-0003-4536-3393

U Jeong Yang – Department of Materials Science and Engineering, Ulsan National Institute of Science and Technology (UNIST), Ulsan 44919, Republic of Korea

Seo Young Kim – Department of Energy Science and Engineering, Daegu Gyeongbuk Institute of Science and Technology (DGIST), Daegu 42988, Republic of Korea

Young Seong Kim – Center for Nanoparticle Research, Institute for Basic Science (IBS), Seoul 08826, Republic of Korea; School of Chemical and Biological Engineering, Institute of Chemical Processes, Seoul National University (SNU), Seoul 08826, Republic of Korea

Jeong Hyun Kim – Center for Nanoparticle Research, Institute for Basic Science (IBS), Seoul 08826, Republic of Korea; School of Chemical and Biological Engineering, Institute of Chemical Processes, Seoul National University (SNU), Seoul 08826, Republic of Korea

Complete contact information is available at:

<https://pubs.acs.org/10.1021/acs.nanolett.0c03939>

Author Contributions

All authors contributed to the writing of the manuscript.

Author Contributions

[†]J.Y. and M.K.C. contributed equally to this work.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

T.H. acknowledges financial support by IBS-R006-D1. D.-H.K. acknowledges financial support by IBS-R006-A1. J.Y. and M.K.C. acknowledge financial support by the NRF grant funded by the Korean government (MSIT) (2020M3D1A2102677). J.Y. also acknowledges financial support by the NRF grant funded by the Korean government (MSIT) (2020R1F1A1061505). M.K.C. also acknowledges financial support by the NRF grants funded by the Korean government (MSIT) (2020M3D1A1110502, 2020M3E5D8108314).

REFERENCES

- (1) Murray, C. B.; Norris, D. J.; Bawendi, M. G. Synthesis and characterization of nearly monodisperse CdE (E = sulfur, selenium, tellurium) semiconductor nanocrystallites. *J. Am. Chem. Soc.* **1993**, *115* (19), 8706–8715.
- (2) Alivisatos, A. P. Semiconductor clusters, nanocrystals, and quantum dots. *Science* **1996**, *271*, 933–937.
- (3) Yu, H.; Li, J.; Loomis, R. A.; Wang, L. W.; Buhro, W. E. Two-versus three-dimensional quantum confinement in indium phosphide wires and dots. *Nat. Mater.* **2003**, *2* (8), 517–520.
- (4) Kagan, C. R.; Lifshitz, E.; Sargent, E. H.; Talapin, D. V. Building devices from colloidal quantum dots. *Science* **2016**, *353* (6302), No. aac5523.
- (5) Ravi, V. K.; Saikia, S.; Yadav, S.; Nawale, V. V.; Nag, A. CsPbBr₃/ZnS core/shell type nanocrystals for enhancing luminescence lifetime and water stability. *ACS Energy Lett.* **2020**, *5* (6), 1794–1796.
- (6) Rogach, A. L.; Gaponik, N.; Lupton, J. M.; Bertoni, C.; Gallardo, D. E.; Dunn, S.; Pira, N. L.; Paderi, M.; Repetto, P.; Romanov, S. G.; O'Dwyer, C.; Torres, C. M. S.; Eychmüller, A. Light-emitting diodes with semiconductor nanocrystals. *Angew. Chem., Int. Ed.* **2008**, *47* (35), 6538–6549.
- (7) Shirasaki, Y.; Supran, G. J.; Bawendi, M. G.; Bulović, V. Emergence of colloidal quantum-dot light emitting technologies. *Nat. Photonics* **2013**, *7* (1), 13–23.

- (8) Yang, J.; Choi, M. K.; Kim, D.-H.; Hyeon, T. Designed assembly and integration of colloidal nanocrystals for device applications. *Adv. Mater.* **2016**, *28* (6), 1176–1207.
- (9) Dai, X.; Deng, Y.; Peng, X.; Jin, Y. Quantum-dot light-emitting diodes for large-area displays: towards the dawn of commercialization. *Adv. Mater.* **2017**, *29* (14), 1607022.
- (10) Panfil, Y. E.; Oded, M.; Banin, U. Colloidal quantum nanostructures: Emerging materials for display applications. *Angew. Chem., Int. Ed.* **2018**, *57* (16), 4274–4295.
- (11) Choi, M. K.; Yang, J.; Hyeon, T.; Kim, D.-H. Flexible quantum dot light-emitting diodes for next-generation displays. *npj Flex. Electron.* **2018**, *2* (1), 10.
- (12) Colvin, V. L.; Schlamp, M. C.; Alivisatos, A. P. Light-emitting diodes made from cadmium selenide nanocrystals and a semiconducting polymer. *Nature* **1994**, *370* (4), 354–357.
- (13) Dai, X.; Zhang, Z.; Jin, Y.; Niu, Y.; Cao, H.; Liang, X.; Chen, L.; Wang, J.; Peng, X. Solution-processed, high-performance light-emitting diodes based on quantum dots. *Nature* **2014**, *515* (6), 96–99.
- (14) Manders, J. R.; Qian, L.; Titov, A.; Hyvonen, J.; Tokarz-Scott, J.; Acharya, K. P.; Yang, Y.; Cao, W.; Zheng, Y.; Xue, J.; Holloway, P. H. High efficiency and ultra-wide color gamut quantum dot LEDs for next generation displays. *J. Soc. Inf. Disp.* **2015**, *23* (11), 523–528.
- (15) Wang, L.; Lin, J.; Hu, Y.; Guo, X.; Lv, Y.; Tang, Z.; Zhao, J.; Fan, Y.; Zhang, N.; Wang, Y.; Liu, X. Blue quantum dot light-emitting diodes with high electroluminescent efficiency. *ACS Appl. Mater. Interfaces* **2017**, *9* (44), 38755–38760.
- (16) Kim, T.; Kim, K.-H.; Kim, S.; Choi, S.-M.; Jang, H.; Seo, H.-K.; Lee, H.; Chung, D.-Y.; Jang, E. Efficient and stable blue quantum dot light-emitting diode. *Nature* **2020**, *586* (7829), 385–389.
- (17) Won, Y.-H.; Cho, O.; Kim, T.; Chung, D.-Y.; Kim, T.; Chung, H.; Jang, H.; Lee, J.; Kim, D.; Jang, E. Highly efficient and stable InP/ZnSe/ZnS quantum dot light-emitting diodes. *Nature* **2019**, *575* (7784), 634–638.
- (18) Choi, M. K.; Yang, J.; Kang, K.; Kim, D. C.; Choi, C.; Park, C.; Kim, S. J.; Chae, S. I.; Kim, T.-H.; Kim, J. H.; Hyeon, T.; Kim, D.-H. Wearable red-green-blue quantum dot light-emitting diode array using high-resolution intaglio transfer printing. *Nat. Commun.* **2015**, *6*, 7149.
- (19) Kim, J.; Shim, H. J.; Yang, J.; Choi, M. K.; Kim, D. C.; Kim, J.; Hyeon, T.; Kim, D.-H. Ultrathin quantum dot display integrated with wearable electronics. *Adv. Mater.* **2017**, *29* (38), 1700217.
- (20) Yang, Y.; Zheng, Y.; Cao, W.; Titov, A.; Hyvonen, J.; Manders, J. R.; Xue, J.; Holloway, P. H.; Qian, L. High-efficiency light-emitting devices based on quantum dots with tailored nanostructures. *Nat. Photonics* **2015**, *9* (4), 259–266.
- (21) Oh, N.; Kim, B. H.; Cho, S.-Y.; Nam, S.; Rogers, S. P.; Jiang, Y.; Flanagan, J. C.; Zhai, Y.; Kim, J.-H.; Lee, J.; Yu, Y.; Cho, Y. K.; Hur, G.; Zhang, J.; Trefonas, P.; Rogers, J. A.; Shim, M. Double-heterojunction nanorod light-responsive LEDs for display applications. *Science* **2017**, *355* (6325), 616–619.
- (22) Bae, W. K.; Park, Y.-S.; Lim, J.; Lee, D.; Padilha, L. A.; McDaniel, H.; Robel, I.; Lee, C.; Pietryga, J. M.; Klimov, V. I. Controlling the influence of Auger recombination on the performance of quantum-dot light-emitting diodes. *Nat. Commun.* **2013**, *4*, 2661.
- (23) Lim, J.; Park, Y.-S.; Wu, K.; Yun, H. J.; Klimov, V. I. Droop-free colloidal quantum dot light-emitting diodes. *Nano Lett.* **2018**, *18* (10), 6645–6653.
- (24) Pu, C.; Dai, X.; Shu, Y.; Zhu, M.; Deng, Y.; Jin, Y.; Peng, X. Electrochemically-stable ligands bridge the photoluminescence-electroluminescence gap of quantum dots. *Nat. Commun.* **2020**, *11*, 937.
- (25) Deng, Y.; Lin, X.; Fang, W.; Di, D.; Wang, L.; Friend, R. H.; Peng, X.; Jin, Y. Deciphering exciton-generation processes in quantum-dot electroluminescence. *Nat. Commun.* **2020**, *11*, 2309.
- (26) Chang, J. H.; Park, P.; Jung, H.; Jeong, B. G.; Hahm, D.; Nagamine, G.; Ko, J.; Cho, J.; Padilha, L. A.; Lee, D. C.; Lee, C.; Char, K.; Bae, W. K. Unraveling the origin of operational instability of quantum dot based light-emitting diodes. *ACS Nano* **2018**, *12* (10), 10231–10239.
- (27) Chen, S.; Cao, W.; Liu, T.; Tsang, S.-W.; Yang, Y.; Yan, X.; Qian, L. On the degradation mechanisms of quantum-dot light-emitting diodes. *Nat. Commun.* **2019**, *10*, 765.
- (28) Ramasamy, P.; Ko, K.-J.; Kang, J.-W.; Lee, J.-S. Two-step “seed-mediated” synthetic approach to colloidal indium phosphide quantum dots with high-purity photo- and electroluminescence. *Chem. Mater.* **2018**, *30* (11), 3643–3647.
- (29) Cao, F.; Wang, S.; Wang, F.; Wu, Q.; Zhao, D.; Yang, X. A layer-by-layer growth strategy for large-size InP/ZnSe/ZnS core-shell quantum dots enabling high-efficiency light-emitting diodes. *Chem. Mater.* **2018**, *30* (21), 8002–8007.
- (30) Li, Y.; Hou, X.; Dai, X.; Yao, Z.; Lv, L.; Jin, Y.; Peng, X. Stoichiometry-controlled InP-based quantum dots: Synthesis, photoluminescence, and electroluminescence. *J. Am. Chem. Soc.* **2019**, *141* (16), 6448–6452.
- (31) Moon, H.; Lee, W.; Kim, J.; Lee, D.; Cha, S.; Shin, S.; Chae, H. Composition-tailored ZnMgO nanoparticles for electron transport layers of highly efficient and bright InP-based quantum dot light emitting diodes. *Chem. Commun.* **2019**, *55* (88), 13299–13302.
- (32) Lim, J.; Park, M.; Bae, W. K.; Lee, D.; Lee, S.; Lee, C.; Char, K. Highly efficient cadmium-free quantum dot light-emitting diodes enabled by the direct formation of excitons within InP@ZnSeS quantum dots. *ACS Nano* **2013**, *7* (10), 9019–9026.
- (33) Zhang, H.; Hu, N.; Zeng, Z.; Lin, Q.; Zhang, F.; Tang, A.; Jia, Y.; Li, L. S.; Shen, H.; Teng, F.; Du, Z. High-efficiency green InP quantum dot-based electroluminescent device comprising thick-shell quantum dots. *Adv. Opt. Mater.* **2019**, *7* (7), 1801602.
- (34) Wang, A.; Shen, H.; Zang, S.; Lin, Q.; Wang, H.; Qian, L.; Niu, J.; Li, L. S. Bright, efficient, and color-stable violet ZnSe-based quantum dot light-emitting diodes. *Nanoscale* **2015**, *7* (7), 2951–2959.
- (35) Han, C.-Y.; Lee, S.-H.; Song, S.-W.; Yoon, S.-Y.; Jo, J.-H.; Jo, D.-Y.; Kim, H.-M.; Lee, B.-J.; Kim, H.-S.; Yang, H. More than 9% efficient ZnSeTe quantum dot-based blue electroluminescent devices. *ACS Energy Lett.* **2020**, *5* (5), 1568–1576.
- (36) Park, J.-S.; Kyhm, J.; Kim, H. H.; Jeong, S.; Kang, J.; Lee, S.-e.; Lee, K.-T.; Park, K.; Barange, N.; Han, J.; Song, J. D.; Choi, W. K.; Han, I. K. Alternative patterning process for realization of large-area, full-color, active quantum dot display. *Nano Lett.* **2016**, *16* (11), 6946–6953.
- (37) Wang, Y.; Fedin, I.; Zhang, H.; Talapin, D. V. Direct optical lithography of functional inorganic nanomaterials. *Science* **2017**, *357* (6349), 385–388.
- (38) Yang, J.; Hahm, D.; Kim, K.; Rhee, S.; Lee, M.; Kim, S.; Chang, J. H.; Park, H. W.; Lim, J.; Lee, M.; Kim, H.; Bang, J.; Ahn, H.; Cho, J. H.; Kwak, J.; Kim, B. S.; Lee, C.; Bae, W. K.; Kang, M. S. High-resolution patterning of colloidal quantum dots via non-destructive, light-driven ligand crosslinking. *Nat. Commun.* **2020**, *11*, 2874.
- (39) Kim, B. H.; Onses, M. S.; Lim, J. B.; Nam, S.; Oh, N.; Kim, H.; Yu, K. J.; Lee, J. W.; Kim, J.-H.; Kang, S.-K.; Lee, C. H.; Lee, J.; Shin, J. H.; Kim, N. H.; Leal, C.; Shim, M.; Rogers, J. A. High-resolution patterns of quantum dots formed by electrohydrodynamic jet printing for light-emitting diodes. *Nano Lett.* **2015**, *15* (2), 969–973.
- (40) Ho, S.-J.; Hsu, H.-C.; Yeh, C.-W.; Chen, H.-S. Inkjet-printed salt-encapsulated quantum dot film for UV-based RGB color-converted micro-light emitting diode displays. *ACS Appl. Mater. Interfaces* **2020**, *12* (29), 33346–33351.
- (41) Kong, Y. L.; Tamargo, I. A.; Kim, H.; Johnson, B. N.; Gupta, M. K.; Koh, T.-W.; Chin, H.-A.; Steingart, D. A.; Rand, B. P.; McAlpine, M. C. 3D printed quantum dot light-emitting diodes. *Nano Lett.* **2014**, *14* (12), 7017–7023.
- (42) Kim, L.; Anikeeva, P. O.; Coe-Sullivan, S. A.; Steckel, J. S.; Bawendi, M. G.; Bulović, V. Contact printing of quantum dot light-emitting devices. *Nano Lett.* **2008**, *8* (12), 4513–4517.
- (43) Nam, T. W.; Kim, M.; Wang, Y.; Kim, G. Y.; Choi, W.; Lim, H.; Song, K. M.; Choi, M.-J.; Jeon, D. Y.; Grossman, J. C.; Jung, Y. S. Thermodynamic-driven polychromatic quantum dot patterning for

light-emitting diodes beyond eye-limiting resolution. *Nat. Commun.* **2020**, *11*, 3040.

(44) Sung, S. H.; Yoon, H.; Lim, J.; Char, K. Reusable stamps for printing sub-100 nm patterns of functional nanoparticles. *Small* **2012**, *8* (6), 826–831.

(45) Kim, T.-H.; Cho, K.-S.; Lee, E. K.; Lee, S. J.; Chae, J.; Kim, J. W.; Kim, D. H.; Kwon, J.-Y.; Amaralunga, G.; Lee, S. Y.; Choi, B. L.; Kuk, Y.; Kim, J. M.; Kim, K. Full-colour quantum dot displays fabricated by transfer printing. *Nat. Photonics* **2011**, *5* (3), 176–182.

(46) Kim, T.-H.; Chung, D.-Y.; Ku, J.; Song, I.; Sul, S.; Kim, D.-H.; Cho, K.-S.; Choi, B. L.; Kim, J. M.; Hwang, S.; Kim, K. 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.

(47) Kim, T.-H.; Lee, C.-S.; Kim, S.; Hur, J.; L, S.; Shin, K. W.; Yoon, Y.-Z.; Choi, M. K.; Yang, J.; Kim, D.-H.; Hyeon, T.; Park, S.; Hwang, S. Fully stretchable optoelectronic sensors based on colloidal quantum dots for sensing photoplethysmographic signals. *ACS Nano* **2017**, *11* (6), 5992.

(48) Choi, M. K.; Yang, J.; Kim, D. C.; Dai, Z.; Kim, J.; Seung, H.; Kale, V. S.; Sung, S. J.; Park, C. R.; Lu, N.; Hyeon, T.; Kim, D.-H. Extremely vivid, highly transparent, and ultrathin quantum dot light-emitting diodes. *Adv. Mater.* **2018**, *30* (1), 1703279.

(49) Choi, M. K.; Park, I.; Kim, D. C.; Joh, E.; Park, O. K.; Kim, J.; Kim, M.; Choi, C.; Yang, J.; Cho, K. W.; Hwang, J.-H.; Nam, J.-M.; Hyeon, T.; Kim, J. H.; Kim, D.-H. Thermally controlled, patterned graphene transfer printing for transparent and wearable electronic/optoelectronic system. *Adv. Funct. Mater.* **2015**, *25* (46), 7109–7118.

(50) Song, J. K.; Son, D.; Kim, J.; Yoo, Y. J.; Lee, G. J.; Wang, L.; Choi, M. K.; Yang, J.; Lee, M.; Do, K.; Koo, J. H.; Lu, N.; Kim, J. H.; Hyeon, T.; Song, Y. M.; Kim, D.-H. Wearable force touch sensor array using a flexible and transparent electrode. *Adv. Funct. Mater.* **2017**, *27* (6), 1605286.

(51) Son, D.; Chae, S. I.; Kim, M.; Choi, M. K.; Yang, J.; Park, K.; Kale, V. S.; Koo, J. H.; Choi, C.; Lee, M.; Kim, J. H.; Hyeon, T.; Kim, D.-H. Colloidal synthesis of uniform-sized molybdenum disulfide nanosheets for wafer-scale flexible nonvolatile memory. *Adv. Mater.* **2016**, *28* (42), 9326–9332.

■ NOTE ADDED AFTER ASAP PUBLICATION

This paper was published ASAP on December 1, 2020, with an incorrect reference citation in Table 1. The corrected version was reposted on December 2, 2020.