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Review Article

Colloidal quantum dot light-emitting devices

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Abstract

Colloidal quantum dot light-emitting devices (QD-LEDs) have generated considerable interest for applications such as thin film displays with improved color saturation and white lighting with a high color rendering index (CRI). We review the key advantages of using quantum dots (QDs) in display and lighting applications, including their color purity, solution processability, and stability. After highlighting the main developments in QD-LED technology in the past 15 years, we describe the three mechanisms for exciting QDs – optical excitation, Förster energy transfer, and direct charge injection – that have been leveraged to create QD-LEDs. We outline the challenges facing QD-LED development, such as QD charging and QD luminescence quenching in QD thin films. We describe how optical downconversion schemes have enabled researchers to overcome these challenges and develop commercial lighting products that incorporate QDs to achieve desired color temperature and a high CRI while maintaining efficiencies comparable to inorganic white LEDs (>65 lumens per Watt). We conclude by discussing some current directions in QD research that focus on achieving higher efficiency and air-stable QD-LEDs using electrical excitation of the luminescent QDs.

Keywords: *nanocrystals; optoelectronics; displays; lighting*

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In the past decade, colloiddally synthesized quantum dots (QDs) – efficient, spectrally pure emitters that can be optically or electrically excited – have emerged as a competitive choice for the emissive component of light-emitting devices (LEDs). Here we review the advantages of QD emitters, the current state-of-the-art in QD-LED technologies, and the key challenges facing the development of QD-based displays and lighting solutions.

Why quantum dots for light-emitting devices (LEDs)?

Saturated colors

The electronic structure of colloidal QDs, which typically range from 3 to 12 nm in diameter, is dominated by quantum size effects (1). This gives colloidal QDs their signature narrowband emission that can be spectrally

positioned by controlling the nanocrystal size during synthesis. For example, CdSe QDs of different sizes provide emission from the blue through the red, while QDs made of a smaller band gap material, such as PbSe, PbS, or CdTe, offer spectral tunability in the near-infrared spectral region (Fig. 1a). Typical emission spectra of QD solutions show a full-width at half-maximum (FWHM) of 30–40 nm, depending on the degree of monodispersity achieved during the colloidal synthesis.

To illustrate the benefit of QD emitters, Fig. 1b shows a Commission International de l'Eclairage (CIE) chromaticity diagram, which maps color in terms of hue and saturation. The boundary of the CIE diagram is defined by the different saturated hues that can be perceived by the human eye, which range in wavelength from $\lambda = 380$ to 780 nm. The purer the color, the closer to the boundary of the CIE diagram it is mapped. The color gamut enabled by a display with red, green, and blue (RGB) pixels is the triangle defined by the coordinates of the individual pixels. The triangle that can be defined for a display using RGB-emitting QDs (black circles) is larger than that of the International Telecommunication Union HDTV standard (dashed black line), highlighting the benefit of QD emitters.

Furthermore, QDs can be used to tune the quality of white lighting, which can be evaluated by color temperature and color rendering index (CRI). The color temperature of a light source is the temperature of an ideal black-body radiator that radiates light of the same hue (i.e. coordinate location on the CIE diagram). For solid-state lighting applications, warmer white with a color temperature ranging from 2,700 to 3,000 K is preferred over bluish white ($>5,000$ K). The CRI defines how well

a particular artificial light source illuminates an object compared to illumination by natural light, with a CRI of 100 indicating that the artificial light source reproduces the lighting conditions achieved by a black-body light source (such as the sun) with the designated temperature. As shown in Fig. 2a, most solid-state lighting sources consist of a blue inorganic LED backlight coated with a yellow phosphor [e.g. cerium-doped yttrium aluminum garnet (Ce^{3+} : YAG)]. While these inorganic white LEDs suffer from poor color temperature and low CRI (typically <75), QD Vision Inc. has shown that by adding QD lumophores to the phosphor, it is possible to achieve white lighting with a color temperature of 2,700 K and a CRI >90 while maintaining an efficiency of 65 lumens per Watt.

Solution processable

Since colloidal nanoparticles are synthesized from organometallic precursors, they retain a passivating layer of ligands, making them solution processable. This facilitates a variety of low cost, large-area deposition techniques, such as phase separation, inkjet printing, and microcontact printing (2–5). The ligands used in the QD synthesis can be exchanged to make the QDs compatible with aqueous solutions in addition to standard organic solvents (6). Furthermore, ligands can be selected to enable deposition of subsequent layers with orthogonal solvents (7) or to permit post-deposition cross-linking for the creation of stable QD films (8).

Stability

Because they have inorganic semiconductor cores, QDs are often more resistant than organic dyes to degradation caused by effects such as photobleaching. Overcoating

Fig. 1. (a) Photoluminescence spectra of CdSe/ZnS and PbS/CdS core/shell colloidal QDs showing the possibility for narrowband emission across the visible and near-IR wavelength regions. The inset schematic shows a colloidal quantum dot (QDs). (b) CIE chromaticity diagram showing that the spectral purity of QDs enables a color gamut larger than the HDTV standard.

Fig. 2. (a) Plot showing the efficiency and color rendering index for different lighting solutions. The first commercial QD-based solid-state lighting solution developed by QD Vision and Nexxus Lighting has a high color rendering index while maintaining high luminescence efficiency. The Quantum Light platform consists of a quantum dot optic (see inset photograph in upper right) that is backlit by a blue LED. (b) Photographs comparing the illumination of objects with an incandescent lamp, a cool white LED, and the Quantum Light solution. (c) Long-term efficiency testing data on QD Vision/Nexxus Lighting LED shows the stability of the QD optic. (Figures courtesy of QD Vision Inc.)

the QD core with a wider bandgap shell of a material such as CdS or ZnS (Fig. 1a) can further improve the stability of the nanocrystal (9). Recent developments in QD synthesis techniques have given rise to extremely stable QDs. For example, thick-shell QDs have been reported to sustain a high degree of thermal stability and maintain their luminescence even when the passivating ligands are removed (10). Furthermore, these thick-shell

QDs suppress blinking (10, 11), which has been linked to suppressed Auger recombination (12), a key source of inefficiency in electrically excited QDs (13). Recent work replacing organic aliphatic ligands on QDs with metal chalcogenide ligands has enabled QD films that are entirely inorganic and exhibit record electronic transport properties (14).

Challenges in quantum dot light-emitting devices (QD-LED) development

There are two key challenges facing the electrical excitation of colloidal QDs: QD charging and QD luminescence quenching in thin film. QD charging can occur whenever dc current passes through a QD film. As QDs become charged, it becomes increasingly difficult to pass current through the device and maintain QD electroluminescence (EL). Time scales associated with QD charging range from minutes to days (15), making it challenging to obtain consistent luminescence from a QD film that experiences significant QD charging.

Second, while QDs suspended in solution routinely have photoluminescence quantum yields of 95%, when the QDs are deposited in a close-packed thin film, the luminescence efficiency decreases by approximately an order of magnitude (to 5 or 10%). Embedding QDs in an insulating polymer matrix decreases the amount of QD luminescence quenching observed in close-packed QD structures (4, 16); however, dc electrical conductivity through these QD-polymer composites is impeded by the low conductance of the wide band gap polymers.

Quantum dot light-emitting devices (QD-LEDs) based on optical downconversion

The small Stokes shift of QDs makes them an ideal system for optical excitation. In contrast to organic dyes, QDs can be optically excited by light anywhere to the blue of the absorption band. The first demonstration of optical downconversion with QDs used blue gallium nitride (GaN) LEDs to excite QDs in poly(lauryl)methacrylate (PLMA) and generate point sources of saturated color light (16). More recently, a full color, flexible electroluminescent display was achieved using inkjet-printed thin films of QDs in polyisobutylene, optically excited by the blue EL from a commercial phosphor powder (4).

Using optical excitation of QDs, both challenges of QD charging and luminescence quenching can be avoided. Electrical current need not pass through the QD emissive layer, so the QDs do not experience charging and can be dispersed in a polymer to preserve their high luminescent efficiencies. Consequently, the first commercial products that utilize QDs employ optical excitation schemes: following the first demonstrations by Lee et al.

(16), Evident Corporation developed strings of Christmas lights in which blue LEDs excite QDs in an epoxy to achieve monochromatic color emission across the visible spectrum, including colors such as purple and aqua that can typically only be achieved by color filtering of white light. Also, QD Vision Inc., in partnership with Nexxus Lighting, is currently commercializing a solid-state lighting solution based on optical excitation of QDs. In their technology, QDs in a transparent matrix are dispersed onto a substrate to form a QD optic with record solid-state QD photoluminescence efficiency of $(95\% \pm 5)$, and are excited by an array of efficient blue LEDs (Fig. 2) to provide mixed color emission across the visible spectrum. This Nexxus/QD Vision Quantum Light™ platform demonstrated chromatic light quality with a CRI of 90 at a color temperature of 2,700 K (soft white), a quality similar to that of an incandescent luminaire but with power to light conversion efficiency that is four times higher, 60 lumens per Watt.

While QD-LEDs based on optical downconversion have made it possible to begin the commercialization of QD-containing products, electrically excited QD films would eliminate the need for multicomponent integration of a QD optic with point-source standard III-V semiconductor LEDs. Demonstrations of such electrically excited planar-format QD-LED technologies could be utilized in novel-format lighting applications at high brightness, or alternatively could be the foundation of a new technology for high color quality, paper-thin flat panel emissive displays that would be structurally simpler than the present LCD display technologies.

Quantum dot light-emitting devices (QD-LEDs) based on organic charge transport layers

Since the first report of electrically driven QD-LEDs in 1994, significant progress has been made toward efficient electrical excitation of QDs. The first electrically driven QD-LEDs incorporated CdSe QDs and polymers between a transparent indium tin oxide (ITO) anode and a metal cathode. Colvin et al. (17) used a layered CdSe QD and p-paraphenylene vinylene (PPV) architecture, and in the work of Dabbousi et al. (18), QDs were dispersed in polyvinylcarbazole and an oxadiazole derivative. Both these initial efforts yielded QD-LEDs with low turn-on voltages and substantial EL from the polymers as well as the QDs.

In 2002, Coe et al. (2) developed a phase separation technique that enabled formation of a close-packed monolayer of QDs on a molecular organic hole transport layer (HTL) [N,N'-diphenyl-N,N'-bis(3-methylphenyl)-N,N'-bis(phenyl)benzidine (TPD)] from a single spin-cast step. The device was completed with a thermally evaporated electron transporting organic molecule layer and exhibited minimal emission from the organic layers, a low turn-on voltage, and an external quantum efficiency (EQE) of 0.52%.

Since then, QD-LEDs incorporating a monolayer of QDs between an organic electron transport layer (ETL) and an HTL, as shown schematically in Fig. 3a, have continuously held the record for QD-LED efficiency, with numerous studies devoted to improving their performance and understanding the mechanism behind their operation.

Fig. 3. (a) Photographs and spectra of multicolor QD-LEDs that sandwich the QD monolayer using the same organic charge transport layers on either side of the having the cross-sectional geometry shown in the inset. Microcontact printing of QDs enables mixing of red, green, and blue QDs within a single monolayer to create a single white light-emitting active layer (b) or side-by-side deposition of $25 \times 25 \mu\text{m}$ red, green, and blue pixels (c). (Figure courtesy of Polina Anikeeva.)

Experimental data and numerical modeling have confirmed that the high efficiency of these organic-based QD-LEDs can be attributed to the concomitant excitation of the luminescent QD film via Förster energy transfer and direct charge injection (19). In the former method for generating QD EL, an exciton forms on the wide band gap organic thin film, non-radiative energy transfers to a QD and radiatively recombines. In the latter, an electron and a hole are injected into the QD, form an exciton, and radiatively recombine.

As shown in Fig. 3a, careful material selection to optimize these two excitation methods enabled EL from blue, cyan, green, orange, and red QD monolayers sandwiched between the same two organic layers [hole-transporting spiro-N,N'-diphenyl-N,N'-bis(3-methylphenyl)-(1,1'-biphenyl)-4,4'-diamine (spiroTPD) and electron-transporting 2,2',2''-(1,3,5-benzenetriyl)-tris(L-phenyl-1-H-benzimidazole) (TPBi)] (20). At the Materials Research Society Fall Meeting in December 2008, QD Vision Inc. reported a red QD-LED with an EQE of 7.5%, which is competitive with the efficiencies of commercially available organic LEDs (OLEDs).

These high efficiency, multicolor QD-LEDs were made possible by the development of QD contact printing, in which single monolayers of QDs can be transferred, solvent free, from a parylene-C coated poly(dimethylsiloxane) (PDMS) stamp onto an organic charge transport layer (5). Fig. 3b shows a display with side-by-side $25 \times 25 \mu\text{m}$ RGB pixels, which was created in only two contact printing steps. First, lines of red QDs at $25 \mu\text{m}$ pitch were printed, followed by orthogonal lines of green QDs. The blue emission comes from the HTL, TPD.

Unlike most white light sources that require stacking of different color pixels or downconversion schemes, Anikeeva et al. (21) demonstrate that it is possible to

fabricate a white LED by incorporating a monolayer of mixed RGB QDs between HTL and ETL. Fig. 3c depicts this white QD-LED, which has an EQE of 0.36% at 10 V applied bias and a CRI of 87.

While these results clearly illustrate the promise of electrically driven QDs for displays and large-area lighting, one principle limitation of these QD-LEDs with organic transport layers is that, like OLEDs, they require environmental packaging to prevent degradation of the organic transport layers in the presence of atmospheric O_2 and water vapor.

Quantum dot light-emitting devices (QD-LEDs) with inorganic charge transport layers

The need for environmental packaging of QD-LEDs using organic charge transport layers may unnecessarily increase the cost and complexity of device manufacturing. As discussed previously, one of the benefits of using QD emitters is that, as inorganic particles, they are often more stable than many organic dyes. It is therefore rational to investigate whether replacing the organic charge transport layer with inorganic charge transport layers would enable air-stable devices that do not require packaging.

The first promising effort at building air-stable colloidal QD-LEDs with inorganic charge transport layers used n- and p-type doped GaN transport layers deposited by energy neutral atomic beam lithography/epitaxy (ENABLE) surrounding colloidal QDs (22). The external quantum efficiencies of the devices ranged from 0.001 to 0.01%, although much of the observed EL was from GaN.

Subsequent work showed that it is possible to use metal oxide charge transport layers deposited at room temperature to build an air-stable QD-LED that exhibits pure QD emission and an efficiency of 0.1% (Fig. 4) (23). The

Fig. 4. (a) Electroluminescence spectra of the first QD-LED to use metal oxide charge transport layers (23). (b) Plot of the QD-LED external quantum efficiency (EQE) measured from the front face of the device as a function of current density. The inset shows a photograph of a bright and uniform pixel at 6 V applied bias operating in ambient conditions, unpackaged.

structure consisted of an ITO anode, a p-type NiO layer, a QD luminescent layer, an n-type zinc tin oxide layer, and a metal cathode. Radio frequency (RF) magnetron sputtering was selected as the deposition method for the metal oxide transport layers since it is a physical vapor deposition (i.e. non-chemically reactive) technique that can be performed at low power and at room temperature to minimize damage to the QD luminescence.

While it has been possible to make systematic improvements to the efficiency of this structure (13), one explanation for the relatively low efficiency of these QD-LED structures with inorganic charge transport layers is that unlike QD-LEDs with organic-based charge transport layers, inorganic QD-LEDs cannot benefit from Förster energy transfer. While excitons form readily in organic thin films, they do not form on the metal oxide thin films and therefore cannot transfer to the QDs. Thus, inorganic QD-LEDs operate solely via direct charge injection, requiring device designs to be based largely on energy band alignment considerations.

Two main approaches to improving the efficiency of inorganic QD-LED structures are being investigated. One approach is to enable Förster energy transfer excitation of colloidal QDs within an inorganic structure. Work by Achermann et al. (24) focused on using InGaN/GaN quantum well structures in close proximity to the QD layer to enable both energy transfer and carrier injection as a means of exciting QDs in an all inorganic structure. Other efforts focus on hybrid QD-LEDs with both inorganic and organic transport layers. By retaining one organic layer, these devices benefit from the dual excitation mechanisms of Förster energy transfer and direct charge injection while gaining stability from the one ceramic transport layer. For example, a solution-deposited ZnO nanocrystal-based ETL was successfully implemented in work by Stowdam et al. (7). Another group explored cross-linking of the QD layer followed by spin-cast deposition of a sol-gel TiO₂ ETL using DuPont precursor tyzol BTP (8).

A second approach is to investigate new mechanisms of QD excitation. We recently reported the development of a unipolar light-emitting device architecture, which highlights the possibility for a paradigm shift away from direct charge injection into QDs as a means for EL in inorganic-based QD-LED structures (25). These devices are the first reported example of DC-field-driven EL in QDs, and we demonstrate that we are able to eliminate charging of the QD film, showing constant luminance from the device over 20 h of continuous operation in air, unpackaged.

Future work

The field of QD optoelectronics has made tremendous strides in the past 15 years, and the need for lower cost, higher efficiency devices with increased functionality will

continue to drive innovation. As the first colloidal-QD-based products to see commercialization, QD-LEDs are playing a crucial role in demonstrating the potential for nanostructured materials in display and lighting technologies as well as other solid-state devices, such as solar cells, photodetectors, and imagers. At the same time, the challenges facing QD-LED development clearly highlight that understanding and controlling QD physics and chemistry are critical to advancing the technology. Multiple research groups have shown that with careful experimental design and systematic investigations, QD-LEDs themselves can be used as platforms for investigating these basic properties. There are unique opportunities for both industry and basic research involvement in QD-LED development that will undoubtedly dramatically change the landscape of colloidal synthesized QD usage in large-area optoelectronic nanotechnologies in the years to come.

Conflict of interest and funding

The authors declare that Vladimir Bulović is a founder of QDVision.

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