Comparison of different RGB InP-quantum-dot-on-chip LED configurations

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Abstract: InP/ZnSe/ZnS quantum dots (QDs) offer a cadmium-free solution to make white LEDs with a narrow blue, green and red emission peak. Such LEDs are required for display and lighting applications with high color gamut. An important phenomenon that hampers the efficiency of such quantum-dot-on-chip LEDs is re-absorption of already converted light by the QDs. Proposed solutions to remedy this effect often rely on complex or cost-ineffective manufacturing methods. In this work, four different RGB QD-on-chip LED package configurations are investigated that can be fabricated with a simple cavity encapsulation method. Using accurate optical simulations, the impact of QD re-absorption on the overall luminous efficacy of the light source is analyzed for these four configurations as a function of the photo-luminescent quantum yield (PLQY) of the QDs. The simulation results are validated by implementing these configurations in QD-on-chip LEDs using a single set of red and green emitting InP/ZnSe/ZnS QDs. In this way, the benefits are demonstrated of adding volume scattering particles or a hemispherical extraction dome to the LED package. The best configuration in terms of luminous efficacy, however, is one where the red QDs are deposited in the recycling cavity, while the green QDs are incorporated in the extraction dome. Using this configuration with green and red InP/ZnSe/ZnS QDs with a PLQY of 75% and 65% respectively, luminous efficacy of 102 lm/W was realized for white light with a CCT of 3000 K.

1. Introduction

Quantum dots (QDs) are considered as a potential alternative to the inorganic phosphors used in wavelength-converted LEDs. Some even describe them as the ultimate down-conversion material [1]. A main advantage of QDs over inorganic phosphors is their narrow and size-tunable emission spectrum that can be varied over the full visible spectrum. This feature allows the usage of the QDs for different applications; e.g., to make an LED with discrete blue, green and red emission peaks. Such an LED is beneficial for the display applications to achieve a wide color gamut at high luminous efficacy (LE) [1], but also for lighting applications that target high color gamut (i.e. high Gamut Index (R_g) according to the TM-30 rendering metric [2]). Of course, such discrete spectral characteristics can also be realized with multi-chip RGB LEDs. This approach, however, faces some important technological issues: inefficient green LEDs (green gap), different temperature-dependent characteristics and aging rates for the three LED chips, and complex mixing optics [3]. A better alternative is offered by using green and red emitting QDs on top of a blue LED chip. However, despite the high photo-luminescent quantum yield (PLQY) of certain QDs: some reported even values close to 100% [4–6], to date, such RGB QD-on-chip LEDs have not reached luminous efficacy values as provided by current phosphor-converted LEDs [7].

Two main physical phenomena hamper the efficacy of QD-on-chip LEDs: (1) Förster resonance energy transfer (FRET) and (2) re-absorption effects [7–9]. The first problem, FRET, occurs when QDs are in very close proximity (less than 10 nm) and results in a red-shift of the emission spectrum and decrease of the PLQY [8]. Tightly packed QDs (agglomerates) may be formed during the transition from liquid (dispersed in toluene) to solid (inclusion in a polymer matrix) [10, 11]. Secondly, the efficacy can be seriously hampered by re-absorption effects [12]. Because the emission of QDs overlaps partly with their absorption spectrum, there is always a certain fraction of already emitted photons by the QDs that is re-absorbed by these QDs. Each re-absorption event increases the probability that the photon is lost if the PLQY of the QDs is non-unity. Such re-absorption events become particularly pronounced when QDs with different output spectra, such as green and red, are mixed together. In such a case, there is a complete overlap of the emission spectrum of the green QDs with the absorption spectrum of the red QDs. The main focus of this work is to analyze and reduce the impact of QD re-absorption on the efficacy of QD-on-chip LEDs.

To overcome the re-absorption problem, configurations with separated red and green QD layers were demonstrated [13–17]. In such a layer-by-layer assembly, it is possible to reduce the amount of green light that passes the red QD layer, thereby limiting re-absorption losses. As an alternative, Li et al. demonstrated that the pore structure of SBA-15 particles acts as an induced waveguide for the down-converted photons, which also reduces QD re-absorption [7]. Another approach to reduce re-absorption losses is by considering QDs with large Stokes shift, e.g. by considering dot-in-rod transitions [18]. The overall extraction efficiency of the QD converted light can furthermore be enhanced by positioning the QDs at a certain distance from the LED chip (so-called remote "phosphor" configurations) [19, 20], and/or by adding scattering particles [21, 22]. Although many of these studies offer inventive solutions to improve optical efficiency, it is also crucial that the proposed configuration can be produced in a straightforward and affordable manner. For this reason, on-chip phosphor LED configurations have prevailed over remote-phosphor systems, despite possible efficiency gains of the latter [23].

The most prominent industrial packaging method for the luminescent material(s) on top of the (blue) source LED is cavity encapsulation. With this approach, an LED chip is mounted on a silver-coated metal lead and surrounded by a plastic or ceramic recycling cavity. This cavity is then filled with a slurry of transparent polymer that contains the luminescent materials [24]. A dome-like structure (hemispherical lens) is often added at the top of the package to enhance the extraction efficiency [24–26]. This type of packaging is simple, cheap, and has a well-developed production technology [25, 26].

In this work, we investigate the overall efficacy and impact of re-absorption of white QD-on-chip LED configurations that can be realized with a simple cavity encapsulation method. In doing so, we try to combine some of the earlier-mentioned ideas with practical manufacturing requirements. Four different configurations are considered: 1) a blue LED with green and red QDs in the recycling cavity, 2) the same configuration with additional scattering particles in the cavity, 3) green and red QDs in the recycling cavity with a dome structure on top, and 4) red QDs in the cavity and green QDs in the dome. For each of these four configurations, the overall luminous efficacy and angular color uniformity are systematically analyzed with reliable optical simulations and experiments. The experimental results are obtained by using green and red InP/ZnSe/ZnS core-shell QDs in a compatible polymer.

2. Materials and methods

2.1. Considered RGB quantum-dot-on-chip LED package configurations

We investigate four different RGB QD LED configurations that can be fabricated with a simple cavity encapsulation method. For each configuration in this study, we start from an empty blue LUXEON 3535 LED package from Lumileds [27] that is filled with QDs dispersed in a photocurable resin. The emission peak of the blue LED chip is at 450 nm. A schematic of the four considered configurations is shown in Figure 1. In the first configuration, the LED package

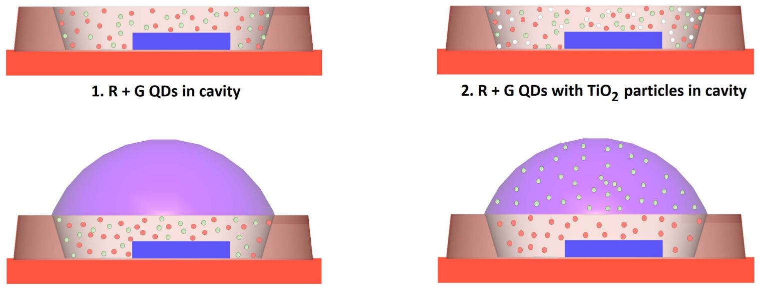

Fig. 1. Schematic representation of the four investigated QD LED configurations that can be fabricated with a cavity encapsulation method.

is filled with a mixture of green and red QDs that are directly applied on top of the blue LED chip. In a second configuration, TiO_2 micro-particles are added to this mixture to induce scattering and thus facilitate the extraction of photons from the package. This can also reduce the likelihood of QD re-absorption. The third configuration is similar to the first but with an additional resin dome on top of the LED package. The function of this dome is also to improve the extraction efficiency. Due to the curvature of this resin dome, the angle of incidence of the light onto the resin/air exit surface, is on average smaller than in the case of a planar exit surface. This reduces the amount of Fresnel reflection and total internal reflection for the light that emerges from the LED package. Finally, in a fourth configuration, we separate the green and red QDs. Red QDs are deposited in the recycling cavity, while green QDs are embedded in the extraction dome on top of the LED package. By doing so, the re-absorption of green light by the red QDs can be significantly reduced. Also in this configuration, the dome helps to reduce total internal reflection.

2.2. *Material synthesis and QD LED sample preparation*

2.2.1. QD synthesis procedure

Red InP/ZnSe/ZnS and green InP/Zn(Se,S)25:75/ZnS QDs were formed using established colloidal synthesis protocols described in detail in previous work [28, 29].

2.2.2. QD integration into the polymer matrix

Purified and degassed 124 mg of InP-based QDs, made using the synthesis procedure referenced above, are redispersed in 0.2 mL of anhydrous toluene in a glovebox. After stirring and obtaining a homogeneous dispersion, 0.2 mL of the thiol polymer precursor pentaerythritol tetrakis(3-mercaptopropionate) (PETMP) is added. The mixture is stirred. Next, the QDs are precipitated using 0.5 mL of anhydrous acetone and then redispersed in 0.2 mL of anhydrous toluene. After that, 131 μL of PETMP is added. After stirring and obtaining a homogeneous dispersion, the mixture is placed for 1 hour under a vacuum to remove toluene. The resulting mixture is subsequently transferred for 4 hours into an ultrasonic bath at 60°C. Finally, the obtained mixture is exposed for 2 minutes to a UV curing light.

2.2.3. Preparation of QD LEDs

In order to obtain a specific correlated color temperature (CCT) for the final white QD LEDs, the concentration of QDs in the PETMP polymer matrix can be fine-tuned by diluting it with an additional transparent polymer. For this purpose, the clear photopolymer resin from Formlabs [30] was used. After homogenization of this polymixture, it is deposited into the LED cavity with a micropipette and cured. The photo-initiator in the Formlabs resin enables the mixture to be

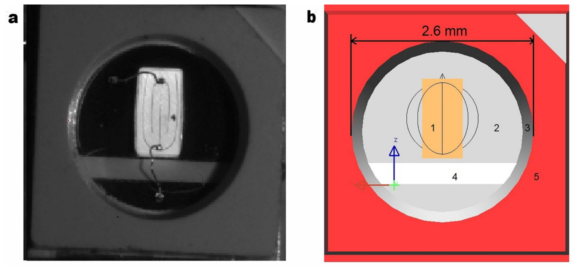

Fig. 2. a) Image of the empty LUXEON 3535 LED module. b) Simulation model of the LED package : (1) LED chip, (2) bottom reflector, (3) inner side of recycling cavity, (4) diffusing bar, (5) package.

cured at around 420 nm. This is helpful since light absorption by the QDs can prevent UV curing at shorter wavelengths. The additional dome structure is made by the so-called 'dip-transfer' technique [31, 32]. Using a vertically oriented hydrophilic strip, photopolymer resin is deposited on top of the LED chip, creating a symmetrical dome shape that is immediately cured with UV light.

2.3. Experimental characterisation

The spectral radiant flux of the resulting LEDs is measured with a custom-made integrating sphere [33]. The LEDs are operated with a low drive current to prevent thermal quenching of the quantum dots (I_f = 10 mA and V_f = 2.615 V). At this drive current, the blue LED chip inside the LUXEON 3535 package has a wall-plug efficiency of around 70% (±2%) when operated at room temperature. In order to characterize the angular color uniformity of the emitted light by the various QD LEDs, an Ocean Optics QE65000 Scientific-grade spectrometer was attached to a near-field goniophotometer for measuring LEDs (RIGO801 from TechnoTeam). The precise rotation mechanism of this goniometer allows the measurement of the LED spectrum from different viewing angles. The obtained spectra are used to analyze the color uniformity. All color-related calculations were performed in the LuxPy toolbox [34].

2.4. Optical modeling and material characterization

The optical performance of all four investigated QD LED configurations was simulated using the commercially available ray-tracing software LightTools [35]. LightTools has built-in algorithms that allow the accurate simulation of an optical system with multiple luminescent materials, including re-absorption. The simulation model includes the blue LED chip (with external quantum efficiency = 70%), the LED package and the luminescent materials (i.e. the quantum dots). The diameter and depth of the LED package are 2.6 mm and 0.5 mm, respectively, and it contains a 580 ×1143× 200 μm blue LED chip. The specular reflectance of the back reflector and LED chip have an average value of ∼ 95%, while an average diffuse reflectance of ∼ 90% is attributed to the sidewalls of the LED package. Detailed measurements and validation of this simulation model can be found in previous work [36]. The LUXEON 3535 LED module and corresponding simulation model are shown in Figure 2.

To make the QD LEDs, green and red InP/ZnSe/ZnS quantum dots and TiO_2 scattering particles were used in a resin with a refractive index of 1.54 (@650 nm) after curing [36]. The scattering properties of the TiO_2 particles are modeled using Mie theory, for which, refractive index and particle size distribution are specified into a built-in LightTools model for Mie scattering. The TiO_2 particles used in this work have an average diameter of 50 nm. Following the specifications given in [37], we set the refractive index equal to 2.5 in our simulation model. The QDs are considered scatter-free in the simulations [38]. The required optical properties for the simulation of the QDs are the absorption coefficient as a function of wavelength, the emission spectrum for different excitation wavelengths, and the PLQY. The absorption spectra of the QDs were obtained with a Perkin Elmer spectrophotometer [39]; emission spectrum and PLQY were measured with the integrating sphere system [40]. The results are shown in Figure 3. We notice a dependence of the emission spectrum on the excitation wavelength for the green QDs, while such an effect was almost imperceptible for the red QDs. The PLQY of the red QDs was measured to be around 65% (in polymer), while 75% was obtained for the green QDs.

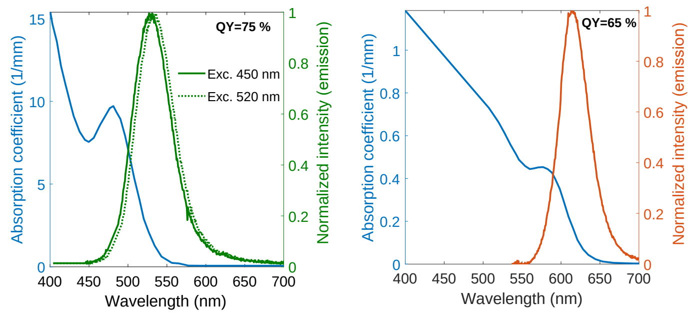

Fig. 3. Emission/absorption spectrum of green and red InP/ZnSe/ZnS quantum dots.

3. Results

3.1. Consequences of QD re-absorption

In order to investigate the impact of re-absorption on the performance of a QD-on-chip LED, we deposited resins with increasing concentration of green or red QDs on top of the blue LED chip and measured the resulting spectral radiant flux. These experimental results are shown in Figure 4(a) for the red QDs. One can see that the amount of blue light decreases when the red QD concentration increases: i.e., more blue light is absorbed by the red QDs. As the amount of blue light decreases, the amount of red light increases, but only for low to moderate QD concentrations. At higher QD loading, one notices a reduction of both the blue and red emission. This decline is a direct result of the re-absorption by the red QDs. In addition to the reduction of the red emission, one also notices a clear red-shift of this emission. This is also a direct consequence of re-absorption, as shorter emitted wavelengths are more susceptible to self-absorption. Similar effects can be seen for the QD-on-chip LED with green QDs, shown in Figure 4(b). The red-shift of the emission spectrum is even more apparent in this case. In both cases, it is clear that the light conversion efficiency is seriously reduced when higher QD concentrations are needed.

Re-absorption effects become even more pronounced in QD LEDs with multiple types of quantum dots. A relevant example are white QD LEDs for lighting or display applications that rely on a combination of green and red QDs on top of a blue LED chip. In this case, there is also a significant part of the green light that is absorbed by the red QDs. This type of re-absorption is often called inter-absorption [41].

Re-absorption losses are the largest for white light with a low correlated color temperature (CCT), as larger QD concentrations are needed in this case to reach the required conversion of blue light. The large possible impact of re-absorption on the overall conversion efficiency of such white QD LEDs is perfectly illustrated in Figure 5(a). It shows the measured spectral radiant flux of two fabricated QD-on-chip LEDs: one with a relative low concentration of green and red QDs in the recycling cavity (resulting in a CCT of 10438 K), and another with a higher concentration

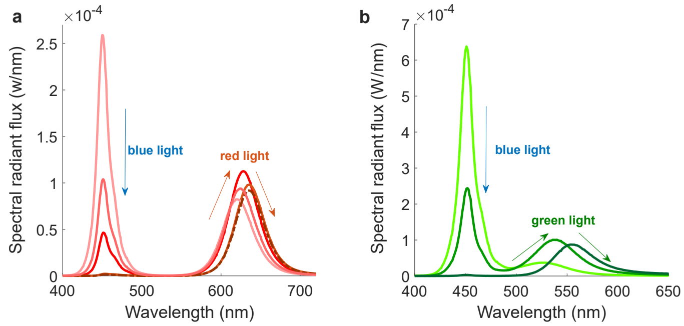

Fig. 4. Resulting spectral radiant flux for QD-on-chip LEDs with increasing amount of red (a) and green (b) QDs. The impact of re-absorption on the amount of down-converted light for higher QD loading is apparent, and clearly results in lower conversion efficiency at higher QD loading. The absorption percentages of the blue pump light with increasing QD concentration are 42%, 67%, 84%, 98.5 %, 99% for the red QDs, and 28%, 73%, 99.5% for the case with green QDs.

of green and red QDs (resulting in a CCT of 2685 K). It is quite striking that the increase of the QD concentration does not result in an increase of the green and red emission; in fact it slightly reduces. The fact that the lower amount of blue light is not accompanied by an increasing amount of green and red light, clearly reduces the total output power and LED efficacy.

Not only the concentration and type of QDs determine the amount of re-absorption; also the specific packaging and ordering of the QD materials on top of the blue LED chip can have an important effect. Figure 5(b) shows the measured spectra of two fabricated white QD LEDs with a different order for the QD deposition: in one LED the red QDs are deposited in the recycling cavity, while the green QDs are positioned in the extraction dome on top of the LED; in the other LED it is the opposite. Positioning the green QDs above the red improves the optical efficiency of the white LED by almost 70% in this case. It is important to note that the PLQY of the used QDs for obtaining the results in Figure 5 was only 50%. This PLQY is also a quite decisive factor for the impact of re-absorption, as will be demonstrated in the next section.

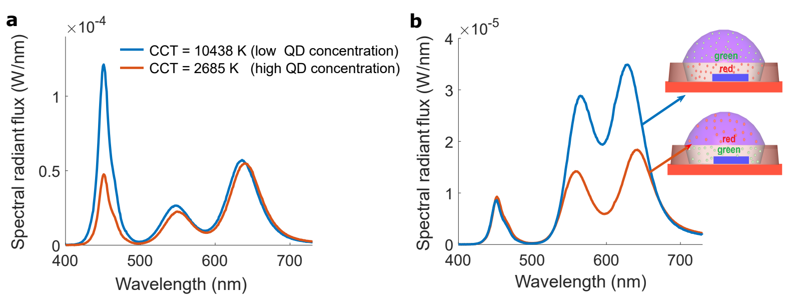

Fig. 5. (a) Measured spectral radiant flux of two white QD LEDs with a relative low and high concentration of green and red QDs. (b) Measured spectral radiant flux of two white QD LEDs with a different order for the green and red QD position in the cavity and dome, respectively. In all cases the PLQY of the QDs is only 50%.

3.2. Simulation results

The four considered QD LED package configurations (see Methods section) are simulated and analyzed in the ray-tracing software LightTools. The different configurations are compared with each other on the basis of the obtained luminous efficacy of the light source. For all configurations, the same optical properties are assumed for the green and red QDs, and the LED package (see Methods section). For the second configuration, the concentration of the TiO_2 particles is optimized to reach maximal output power. In all cases, the green and red QD concentrations are optimized such that the simulated spectrum results in white light with chromaticity coordinates on the Planckian locus and a CCT equal to 3000 K or 5000 K. The simulations are performed for QDs with a PLQY of respectively 50, 75 and 95%. The results for the various configurations are shown in Figure 6.

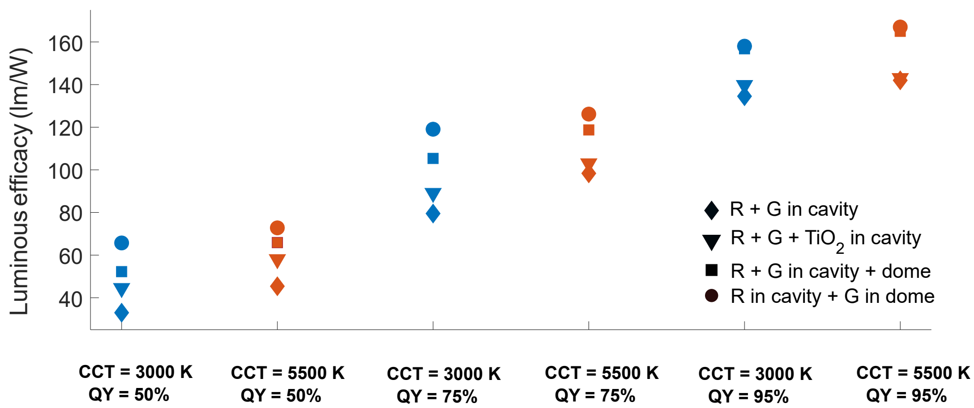

Fig. 6. Simulated luminous efficacy of the four QD-on-chip LED package configurations (shown in Figure 1), for different values of the QD PLQY and CCT of the emitted light.

As could be expected, the luminous efficacy is significantly higher when the PLQY of the green and red QDs is higher. Furthermore, the efficacy is also higher for larger CCT, as this requires lower QD concentrations, resulting in a lower impact of QD re-absorption. When considering the different QD LED configurations, the same tendencies are observed for different PLQY and CCT: the best performance is systematically obtained when the green and red QDs are separated in the cavity and dome, followed by the QD LED with the extraction dome, the QD LED with additional scattering particles and finally the most basic QD LED configuration. The positive effect of adding scattering particles or a dome structure to the QD LED package is more or less similar, irrespective of the PLQY, while the positive effect of separating the green and red QDs is more important when the PLQY of the QDs is lower.

Table 1 summarizes the main loss mechanisms and resulting optical efficiency for each of the simulated configurations, for the case with CCT = 3000 K and PLQY = 75%. The optical efficiency is calculated as the optical power of the emitted light from the QD LED divided by the emitted power by the blue LED chip. The needed concentrations of green and red QDs in order to reach a CCT = 3000 K are also specified by giving the absorption coefficient (in 1/mm) at 450 nm. As can be seen from this table, configuration 4 with green QDs in the dome has relative low PLQY + Stokes losses, and therefore requires less green and red QDs. Also, configuration 2 with TiO_2 particles comes with a relatively low concentration of QDs, because the enhanced scattering in the package increases the light interaction with the QDs. Unfortunately, this scattering also increases the light interaction with the partially absorbing surfaces of the package, thereby increasing the overall package losses. The use of an extraction dome (3 and 4) clearly minimizes package losses by enhancing the extraction efficiency from the package. Given the large differences in the required QD concentrations, it is certainly worth mentioning that configurations with lower QD concentration have an economic advantage for high-volume manufacturing.

Simulation: loss mechanisms in QD LEDs CCT = 3000 K & PLQY = 75%				
QD LED configuration	**1**	**2**	**3**	**4**
μ_a (@450 nm) [1/mm] Green QDs	5.9	1.5	5.8	2.4
μ_a (@450 nm) [1/mm] Red QDs	0.96	1.92	0.65	1.05
PLQY + Stokes losses	56.5%	48.9%	52.9%	45.4 %
Package losses	11%	13.8%	6.4%	8%
Polymer absorption	0.7%	1.4%	0.8%	1.8%
Optical efficiency	**31.8%**	**35.9%**	**39.9%**	**44.8%**

Table 1. Overview of main loss mechanisms for the considered QD LED configurations.

3.3. Experimental results

The four QD-on-chip LED configurations that were simulated and analyzed in the previous section, have also been fabricated in practice using green and red QDs with a measured PLQY of 75% and 65%, respectively. For all configurations, a CCT of 3000 K was targeted. To demonstrate the reliability of our simulation model, in Figure 7(a), we show the good match between the simulated and measured spectrum for the configuration with green and red QDs mixed together in the recycling cavity.

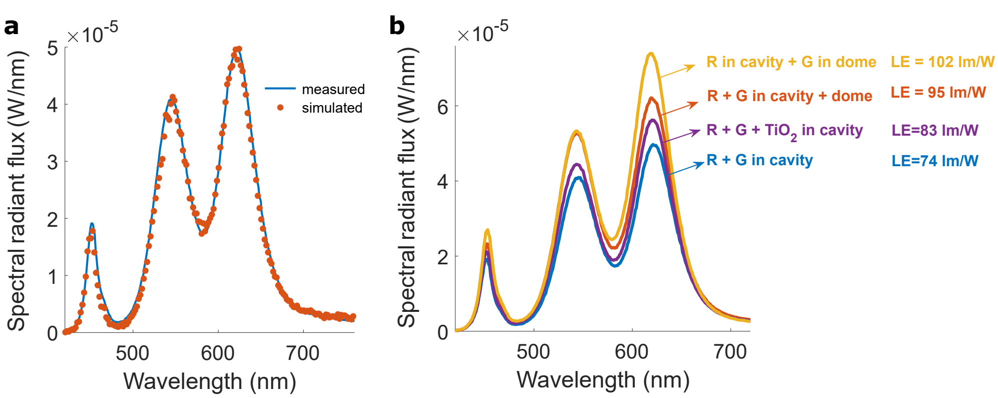

Fig. 7. (a) Good match between the simulation and measurement for the QD LED configuration with green and red QDs mixed in the recycling cavity. (b) Measured spectral radiant flux of four QD LED configurations with a CCT ≈ 3000 K.

In Figure 7(b) the measured spectral power distributions of the different QD LED configurations are shown. For each, the corresponding luminous efficacy of the light source is given in Table 2, together with the corresponding simulated values. The measured luminous efficacies are between 74 lm/W when the red and green QDs are mixed in the recycling cavity, and 102 lm/W when the green QDs are in the dome and the red QDs in the cavity. The main tendencies for these experimental results are in line with the simulation results, although there are some deviations when considering the actual values; especially for the configurations with a dome structure. We see two main reasons for this discrepancy: (1) the shape of the manufactured domes deviates a bit from the hemispherical shape that is considered in the simulations. This is a consequence of the manual dip-transfer technique that was used to make these domes. (2) The QD resin is not completely scatter-free. So even for the configurations without TiO_2 particles, there is some volume scattering in the LED package. This assumption is confirmed by the fact that less TiO_2 particles needed to be used in practice to reach optimal performance for the second configuration, than in simulation. This intrinsic scattering of the QD resin could also explain why the difference in luminous efficacy between configuration 1 and 2 is less in practice than in simulations. Furthermore, if we take some intrinsic scattering into account in the simulations of the QD LEDs with dome, we indeed see that the luminous efficacy drops.

	Simulations		**Measurements**	
	LE (lm/W)	**CCT (K)**	**LE (lm/W)**	**CCT (K)**
R + G in cavity	76	3060	74	3170
R + G + TiO_2 in cavity	87	3180	83	3160
R + G in cavity + dome	110	2950	95	3240
R in cavity + G in dome	114	3150	102	2980

Table 2. Luminous efficacy of white QD LEDs : Simulated vs. experimental results

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For certain LED applications, angular color-uniformity or color-over-angle is an important property. The measured angular color uniformity for all four configurations is shown in Figure 8. The (u',v') chromaticity coordinates are calculated for the measured spectra under different viewing angles along two orthogonal directions (shown with arrows on the LED figure) with a 10° interval. As a reference, a circle with a radius of 0.0055 is drawn on the CIE 1976 (u',v') chromaticity diagram, corresponding to a five-step MacAdam ellipse [42]. The only QD LED configuration that has a color-over-angle within this interval is the one with added scattering particles.

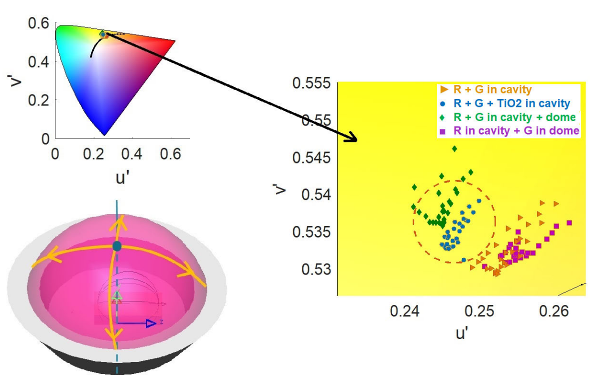

Fig. 8. The chromaticity points of the emission spectrum measured from different viewing angles in the CIE 1976 (u',v') chromaticity diagram, for the different QD LED configurations. The 'dashed' circle has a radius of 0.0055 and corresponds with a five-step MacAdam ellipse.

4. Discussion

Both experiments and simulations in this paper clearly illustrate the large impact that QD re-absorption can have on the luminous efficacy of white QD-on-chip LEDs. While it is almost impossible to completely eliminate this effect, a careful design of the QD LED package configuration can clearly minimize the reduction of the optical efficiency by re-absorption. When comparing the simulation results for a QD PLQY of 75%, we found a difference in luminous efficacy of up to 40% between the best and worst QD LED configuration. By adding volume scattering particles to the QD LED package, some gain in luminous efficacy can be realized, while also improving the color-over-angle performance. The addition of an extraction dome to the package and separation of the green and red QDs, can further increase the luminous efficacy. At higher values (near-unity) for the PLQY, the impact of the specific package configuration becomes smaller. In this case, the external quantum efficiency of the blue LED chip and the package losses dominate the overall efficacy of the white QD LED.

The answer to the question if white quantum-dot-on-chip LEDs can have a higher efficacy than current color-mixed RGB LEDs, despite the re-absorption losses, is definitely yes. If we take into account that the best color-mixed RGB LEDs at this moment have a luminous efficacy around 120 lm/W [43], it is clear from Figure 6 that this value can be easily surpassed by using QDs with a PLQY of around 90-95%. Already at this moment, this is feasible with state-of-the-art synthesis procedures for InP/ZnSe/ZnS QDs, as has been demonstrated in [29].

5. Conclusion

In this paper, the performance of four different white quantum-dot-on-chip LED configurations has been analysed in detail using both optical simulations and experimental demonstrators. In doing so we have demonstrated that the impact of QD re-absorption losses on the luminous efficacy of white QD LEDs can be significantly reduced with a clever design of the LED package configuration. All investigated QD LED configurations in this paper can be manufactured with a simple cavity encapsulation method. The configuration with red QDs in the recycling cavity and green QDs in the extraction dome gives the highest luminous efficacy, both in simulation and practice. To make the experimental demonstrators, LUXEON 3535 LED packages were used with a wall-plug-efficiency of 70% for the blue LED chip. By filling this LED package with green and red InP/ZnSe/ZnS QDs, with a PLQY of 75% and 65% in resin, a luminous efficacy of 102 lm/W was reached for a CCT of 3000 K. This is the highest reported value in the literature for LEDs with Cd-free QDs (i.e. InP/ZnSe/ZnS core-shell QDs in our case), when considering the relative low CCT of 3000 K. For obtaining such low CCT values, higher QD loading is needed and re-absorption effects are even more prominent [7, 44]. Our optical simulations indicate that a luminous efficacy above 150 lm/W can be reached when the PLQY of the used QDs is equal to 90-95%. To increase this luminous efficacy even further, an improvement of the wall-plug-efficiency of the blue LED chip or a further reduction of the package losses would be needed.

Funding.

This work was financially supported by the SBO-project QDOCCO within the SIM SoPPoM+ program of the Flemish Strategic Initiative for Materials (SIM) and Ghent University (GOA no. 01G01019).

Acknowledgments. We are grateful to Lumileds for providing us with a large batch of empty 3535 LED packages and relevant information about this specific package.

Disclosures.

The authors declare no conflicts of interest.

Data Availability Statement.

Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon request.

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