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Bridging the Green Gap: Monochromatic InP-Based Quantum-Dot-on-Chip LEDs with over 50% Color Conversion Efficiency

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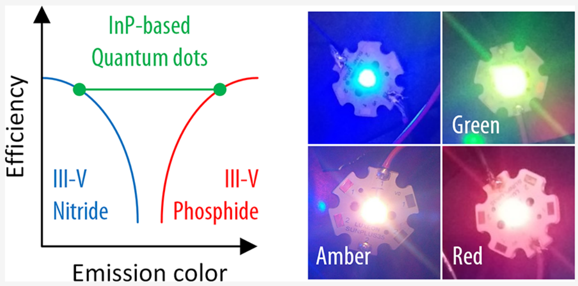

ABSTRACT: Solid-state light-emitting diodes (LEDs) emit nearly monochromatic light, yet seamless tuning of emission color throughout the visible region remains elusive. Color-converting powder phosphors are therefore used for making LEDs with a bespoke emission spectrum, yet broad emission lines and low absorption coefficients compromise the formation of small-footprint monochromatic LEDs. Color conversion by quantum dots (QDs) can address these issues, but high-performance monochromatic LEDs made using QDs free of restricted, hazardous elements remain to be demonstrated. Here, we show green, amber, and red LEDs formed using InP-based QDs as on-chip color convertor for blue LEDs. Implementing QDs with near-unity photoluminescence efficiency yields color conversion efficiency over 50% with little intensity roll-off and nearly complete blue light rejection. Moreover, as the conversion efficiency is mostly limited by package losses, we conclude that on-chip color conversion using InP-based QDs can provide spectrum-on-demand LEDs, including monochromatic LEDs that bridge the green gap.

KEYWORDS: *solid-state lighting, nanocrystals, photoluminescence, color conversion, InP/ZnSe*

The introduction of blue light-emitting diodes (LEDs) based on InGaN made solid-state lighting the preferred technology for lighting and display applications, for which different spectral requirements could be met by combining blue LEDs with color-converting phosphors.[1] This approach was preferred over the incorporation of LEDs emitting different colors within a single package because realizing energy-efficient green to amber LEDs using nitride- or phosphide-based LEDs proved difficult; an issue described as the green gap.[2,3] Still, the wide emission lines and low absorption coefficients of powder phosphors cause a trade-off between color rendering and efficacy in the case of white LEDs,[4] result in limited color purity for displays, and prevent the formation of thin-film color convertors for microLEDs.

Colloidal quantum dots (QDs) have been used over the last 10 years as alternative phosphors for color conversion. QDs stand out by their narrow-band emission and high absorption coefficients for blue light,[5,6] such that less material is required to obtain a more monochromatic color-converted spectrum.[7] From the initial implementation of red- and green-emitting QDs as remote phosphors in liquid crystal displays,[8−10] interest shifted to QD-on-chip configurations, which offer more design freedom at lower cost. Despite exposure to higher temperature and a higher flux of blue light, this approach led to the demonstration of high color rendering, efficient white LEDs through the addition to the phosphor coating of red-emitting QDs,[11] or LEDs for display backlighting using red- and green-emitting QDs.[12,13] In this context, CdSe-based QDs are typically considered the best performing QDs, offering a more narrow emission with a higher quantum yield and a better stability. Even so, cadmium is a strongly regulated, toxic heavy metal, a situation that makes the successful implementation of nonrestricted alternatives imperative for QD-on-chip LEDs to become a viable technology. In this context, InP-based QDs are widely seen as the most promising alternative, as demonstrated by first reports on white InP QD-on-chip LEDs with high color rendering.[14]

Recently, monochromatic QD-on-chip LEDs were introduced as a unique technology for realizing full-color microLED displays,[15,16] an application requiring the integration of separate red, green and blue LEDs. For optimal color purity, the color-converted emission spectrum of such LEDs should be free of residual blue light, which has been accomplished by adding Bragg reflectors,[17] or using QD films with a high optical

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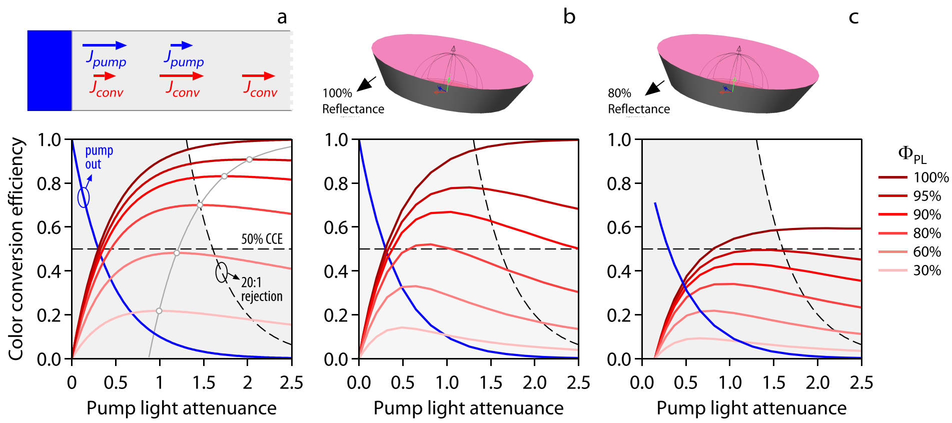

Figure 1. Color conversion efficiency $\Phi_{ext,c}$ simulated for QD-on-chip LEDs assuming nonscattering QDs exhibiting a 5:1 ratio of the absorption cross section at the pump and the converted wavelength and Φ_{PL} as indicated. (a) Analytical simulation results assuming a one-dimensional convertor layer, neglecting any reflection of pump and converted light at the convertor surface and assuming perfect reflection of converted light on the blue LED. The increasing pump light attenuance reflects either an increased QD loading or convertor film thickness. The gray line indicates the locus of maximal $\Phi_{ext,c}$ as a function of the PLQY. (b) Numerical simulations for a specific LED cup, fully filled with a convertor layer assuming perfectly reflecting sidewalls. The increasing pump light attenuance reflects an increased QD loading. (c) Same as (b), assuming sidewalls with 80% reflectance. In all cases, the white area marks combinations of $\Phi_{ext,c}$ and pump light attenuance where the ratio of extracted converted versus blue photons exceeds 20:1, and the blue trace is the normalized flux of outgoing pump photons.

density at blue wavelengths.[18] While the latter approach requires less processing steps, increasing the absorbance may reduce the external quantum efficiency of the convertor film by exacerbating losses caused by QD self-absorption.[19] Despite such problems, research has mostly focused on enhancing color quality.[20−22] Best-reported external conversion efficiencies remain at 45% for red and 23% for green QD-on-chip LEDs made using CdSe-based materials,[19,23] while few, if any, studies made use of InP-based QDs.

Here, we report on monochromatic QD-on-chip LEDs formed by depositing a color convertor coating containing InP-based QDs with a near-unity photoluminescence quantum yield (PLQY) on blue LEDs. Using green-emitting QDs as an example, we discuss the main characteristics of InP-based QD-on-a-chip LEDs, and we address the factors limiting the optical and power conversion efficiency. Highlighting the impact of the PLQY and package-related losses, we demonstrate a green-emitting QD-on-chip LED with a package-loss limited color conversion efficiency of 52%, corresponding to a wall plug efficiency of 30%, that is almost independent of the LED power. Finally, we show monochromatic QD-on-chip LEDs using amber-and red-emitting InP-based QDs with similar characteristics. Such QD-on-chip LEDs outperform monochromatic green and red QD-on-chip LEDs made using CdSe-based QDs described in the literature and all amber LEDs available on the market. We therefore conclude that InP-based QD-on-chip color conversion can provide spectrum-on-demand LEDs, including monochromatic LEDs that address the restrictions on Cd-based compounds and bridge the green gap.

■ REQUIREMENTS FOR EFFICIENT MONOCHROMATIC QD-ON-CHIP LEDS

To understand the performance-limiting factors of QD-based color conversion, Figure 1 represents the simulated external quantum efficiency of color conversion $\Phi_{ext,c}$ (color conversion efficiency (CCE) in short) of a QD-on-chip LED. Here, we define this quantity as the ratio between the flux of incoming pump photons ($J_{in,pump}$) and outgoing converted photons ($J_{out,conv}$):

$$\Phi_{ext,c} = \frac{J_{out,conv}}{J_{in,pump}} \tag{1}$$

Moreover, we characterize a given convertor film by the attenuance A_{pump} of pump light, defined in terms of the incoming and outgoing flux of pump photons as

$$A_{pump} = -\log \frac{J_{out,pump}}{J_{in,pump}} \tag{2}$$

As detailed in Suporting Information Section S1, we obtained $\Phi_{ext,c}$ as a function of the attenuance using increasingly realistic models. Interestingly, while A_{pump} will depend on the QD loading and, possibly, light scattering, we show in Suporting Information Section S1 that the eventual color conversion efficiency is, to a good approximation, a unique function of A_{pump}. Hence, we will characterize in this study color-converted QD-on-chip LEDs by A_{pump}, rather than by QD loading. Furthermore, we assumed a 5:1 ratio between the absorption cross section at the pump and the converted wavelength for all simulations, a figure that is in agreement with experimental values for the different QD batches used here; see Supporting Information S2.

In the most simple case of a one-dimensional, scatter-free converter layer atop a perfectly reflecting pump source, $\Phi_{ext,c}$ can be written as an analytical function determined by the single-event photoluminescence quantum yield (PLQY, Φ_{PL}) of the QDs and the attenuance of the pump light.[19] As shown in Figure 1a, such a model predicts that $\Phi_{ext,c}$ will be systematically smaller than Φ_{PL}, apart from the limiting case of Φ_{PL} = 100%, an outcome caused by self-absorption of the converted light by the QDs. The ensuing trade-off between absorbing more pump light and minimizing self-absorption results in an optimal attenuance, where $\Phi_{ext,c}$ is maximal. Still, for the given 5:1 ratio between the cross section at the pump and the converted wavelength, a PLQY of ~65% suffices to obtain a CCE over 50% in combination with a 20:1 rejection

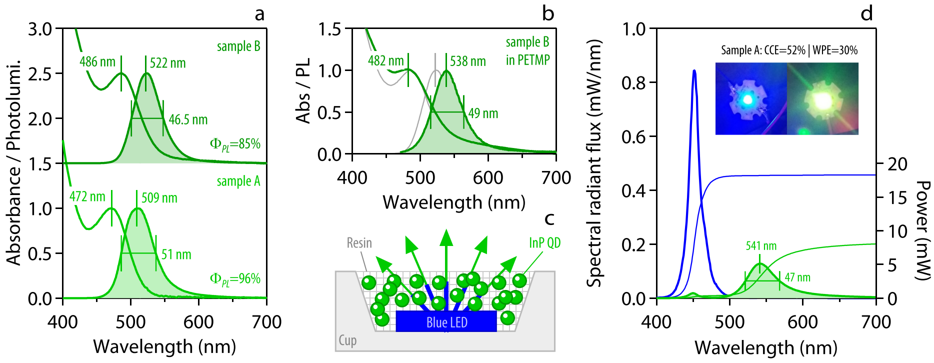

Figure 2. Formation of green QD-on-chip LEDs from InP-based QDs. (a) Example of absorbance and photoluminescence spectra of a dispersion in toluene of green-emitting InP/Zn(Se,S)$_{25:75}$/ZnS core/shell/shell QDs used in this study. (b) Same as (a) for the QD-in-resin obtained after photocuring. (c) Cartoon image of a fabricated green QD-on-chip LEDs. (d) Spectral radiant flux of (blue) the blue pump LED and (green) a green QD-on-chip LED both driven by a current of 10 mA, together with the integrated power. The peak emission wavelength and line width of the green emission band have been indicated, and so are the color conversion efficiency and the wall plug efficiency. The inset depicts the blue pump LED and the green QD-on-chip LED driven by a 10 mA current.

ratio between the residual pump and converted photons in the output spectrum.

In reality, part of the pump and converted light will be internally reflected upon exiting the convertor film. To evaluate the impact of the resulting enhanced light recycling, we switched from an analytical model to ray-tracing simulations for the actual LED cup to calculate $\Phi_{ext,c}$. As shown in Figure 1b, the lengthening of the optical path brought about by internal reflections magnifies the impact of self-absorption. Unless the QDs have unity PLQY, the CCE deteriorates, and the rejection of blue light when $\Phi_{ext,c}$ is maximal diminishes. As a result, Φ_{PL} must exceed ∼85% to combine a +50% CCE with a 20:1 rejection ratio. Furthermore, the surfaces of the LED package and/or the LED dye are not ideal reflectors. In a previous study, we estimated the reflectance for the LEDs used in this work rather at 80−90%.[14] As shown in Figure 1c, reducing the sidewall reflectance to 80% in the simulations mostly affects the convertor films with the highest PLQY since photons can still escape from such films in a loss-free package after multiple reflections at sidewalls or the convertor top surface. For the simulation parameters used, a CCE over 50% and a 20:1 rejection ratio are obtained only if Φ_{PL} exceeds 95%. We thus conclude that realizing efficient, monochromatic QD-on-chip LEDs requires both high PLQY QDs and low-loss LED packages.

■ GREEN QD-ON-CHIP LEDS BY INP-BASED QUANTUM DOTS

To form such QD-on-chip LEDs, we made use of InP-based QDs synthesized by reacting tris(diethylamino)phosphine and an indium halide in oleylamine according to established procedures;[24,25] see Supporting Information Section S2 for details on synthetic protocols and material characteristics. As reported recently,[25] shelling such InP core QDs using a ZnSe/ZnS or Zn(Se,S)/ZnS double shell results in QDs with near-unity PLQY across the visible spectrum from cyan/blue to red. Figure 2a shows absorbance and photoluminescence spectra of thus obtained green-emitting InP/Zn(Se,S)$_{25:75}$/ZnS QDs that feature an emission line centered around 522 nm with a full width at half-maximum of 46.5 nm and a PLQY of 85%. To form color convertor layers, these QDs were dispersed in a photocurable thiol−ene resin consisting of pentaerythritol tetrakis(3-mercaptopropionate) (PETMP); see Supporting Information Section S3 for details. We found that the incorporation of the QDs in such a cross-linked resin significantly enhanced the photostability of the QDs in ambient, while inducing a considerable redshift of the emission in the QD-in-resin composites used for on-chip color conversion; see Figure 2b. On the other hand, we observed no shift of the absorption line of the same composites, while also the emission line of dilute QD-in-resin composites stayed put; see Supporting Information Section S3. We therefore assigned the redshift in the actual QD-in-resin composites used for on-chip color conversion to self-absorption of the emitted light. Moreover, upon analyzing dilute QD-in-resin composites, we found that the several steps involved in the formation of this resin did not affect the initial PLQY. Hence, we will use this initial PLQY, measured on QD dispersions using an integrating sphere, as representative for the single-point PLQY of the QD-resin.

As outlined in Figure 2c and Supporting Information Section S3, we fabricated QD-on-chip LEDs by filling the cup of a blue LED (LumiLeds Luxeon 3535L) with the uncured QD-resin and induced photopolymerization using a midpower 400 nm LED. Figure 2d represents the spectral radiant power of a QD-on-a-chip LED thus obtained. Here, a resin stock solution of 30 wt % QDs was diluted such that an attenuance A_{pump} = 1.65 was obtained, which implies that a mere 2% of the blue pump light is transmitted through the color convertor. Anticipating a spectral red shift upon resin incorporation, we used InP/Zn(Se,S)$_{25:75}$/ZnS QDs (sample A, see Figure 2a and Supporting Information Section S2) emitting at 509 nm in this case. As shown in Figure 2d, the thus obtained QD-on-chip LED exhibited a distinct green color, which corresponded to a color-converted spectrum with a peak emission at 541 nm, a line width of 47 nm, and a blue pump rejection ratio of 23:1.

The operational stability of the QD-on-chip LEDs reported here is directly linked to the radiant power of the blue pump LED. As outlined in Supporting Information Section S4, the power emitted by the QD-on-chip LEDs remained constant during continuous operation when a driving current of 10 mA was used, which corresponds to a blue irradiance of 2.75 W/cm^2. On the other hand, increasing the driving current to 150 mA, i.e., 37.7 W/cm^2, resulted in a progressive reduction of the converted light intensity with time. Unless mentioned otherwise, all characterization therefore involved an LED

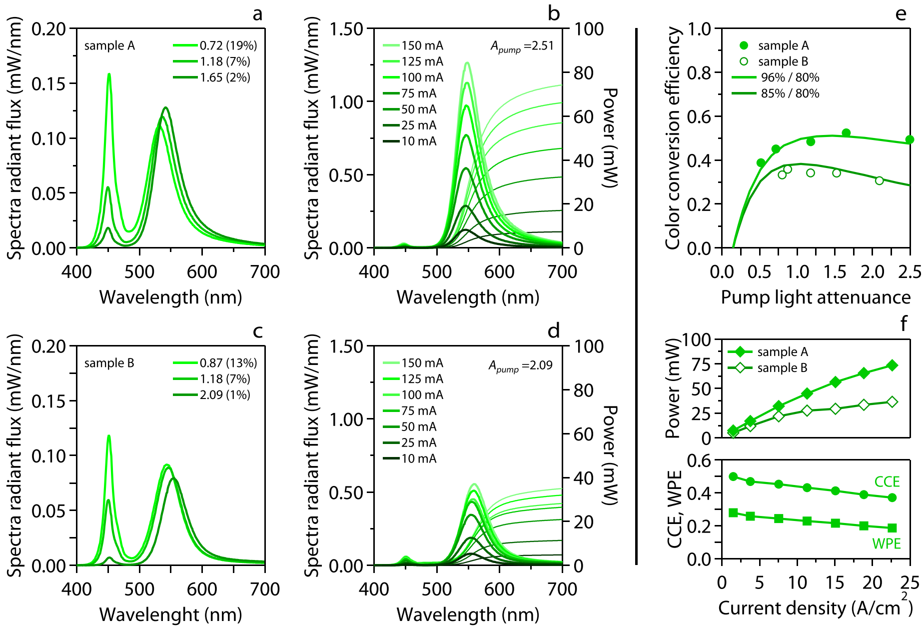

Figure 3. Identification of performance-limiting factors of QD-on-chip LEDs. (a) Spectral radiant flux for QD-on-chip LEDs made with convertor layers containing a different loading of sample A green-emitting QDs, as characterized by the resulting attenuance A_{pump} and the transmitted fraction of blue light. (b) Spectral radiant flux for a QD-on-chip made using sample A QDs featuring A_{pump} = 2.51 as a function of the current driving the blue LED. Thin lines represent a running integration of the emitted power. (c,d) Same as (a,b) for sample B green-emitting QDs. (e) Color conversion efficiency as deduced from the spectral radiant flux of the sample A and sample B QD-on-chip LEDs with different convertor layers and the uncoated blue LEDs. The full lines represent simulations assuming combinations of PLQY and sidewall reflection as indicated. (f) Representation of (light green, solid markers) the emitted power, CCE and WPE as a function of the driving current density of the blue LED die, as deduced from the spectral radiant flux shown of the sample A QD-on-chip LED shown in (b) and the uncoated blue LED. (dark green, open markers) the same for the sample B QD-on-chip LED. Only the output power is shown in this case.

driving current of 10 mA. Interestingly, from the spectral radiant power of the blue pump LED and the color-converted output spectrum under these conditions, we obtained a CCE for the LED shown in Figure 2d of 52%, which corresponded to a wall plug efficiency (WPE) of 30%. As we use blue LEDs with a WPE of ~69%, such figures correspond to an external quantum efficiency (EQE), i.e., the ratio between converted photons transmitted and charges injected in the blue LED, of 36%. This figure exceeds the highest EQE of 23.2% reported for a green QD-on-chip LED in the literature, which was formed by placing a liquid dispersion of CdSe-based QDs atop a blue LED,[23] and indicates that monochromatic, InP-based QD-on-chip LEDs can have a performance on par or better than InGaN green LEDs.[1,26]

■ PERFORMANCE ANALYSIS OF GREEN QD-ON-CHIP LEDS

To assess the factors limiting the performance of the QD-on-chip LEDs introduced here, we prepared LEDs with a progressively higher QD loading in the convertor layer. Characterizing this loading by the attenuance A_{pump}, Figure 3a indicates that a higher QD loading shifts the QD emission line to longer wavelengths, an observation typically assigned to self-absorption,[7] while enhancing the intensity of converted green light. Quantifying these spectra, we find that $\Phi_{ext,c}$ increases from 39 to 52%, when A_{pump} is raised from 0.52 to 1.65; see Figure 3e. Hence, while self-absorption does occur in the convertor layer, the process does not reduce the converted light intensity in this range of concentrations. Referring to the initial simulations shown in Figure 1, such a situation points toward a combination of a high PLQY and significant package loss. As illustrated in Figure 3e, the observed variation of the CCE as a function of A_{pump} is indeed well described by the model simulation when combining the measured PLQY of 96% for sample A in combination with 80% reflective sidewalls.

Referring to the simulation results depicted in Figure 1, increasing the CCE of a QD-on-chip LED by absorbing more pump light is not a trivial feat. To underscore this point, Figure 3c represents a similar output spectrum analysis of QD-on-chip LEDs made using somewhat less efficient green-emitting QDs (sample B; see Figure 2a). At lower QD loading, yielding A_{pump} = 0.87, this LED has a CCE of 35%, a figure reflecting the lower PLQY of the sample B QDs. In this case, however, we found that increasing the QD loading so as to raise A_{pump} above 1, decreases the power of the green light extracted from the LED; see Figure 3c. As a result, $\Phi_{ext,c}$ systematically decreases with increasing loading; a variation that is well described by the model simulation when combining the measured PLQY of 85% for sample B and 80% reflective sidewalls; see Figure 3e.

Assuming pump and converted wavelengths of 450 and 540 nm, respectively, the heat generated by radiative or non-radiative recombination of electron−hole pairs amounts to 0.45 and 2.75 eV per absorbed photon. Hence, even when neglecting self-absorption, a decrease of the PLQY from 96 to 85% will increase the heat generated by conversion losses during LED operation by almost by half. The resulting temperature increase can be an issue since the band-edge photoluminescence of InP-based QDs shifts to longer wavelengths and becomes less efficient with increasing temperature. In the case of the green-emitting InP/Zn-$(Se,S)_{25:75}$/ZnS QDs used here, for example, increasing temperature from room temperature to 100 °C shifts the

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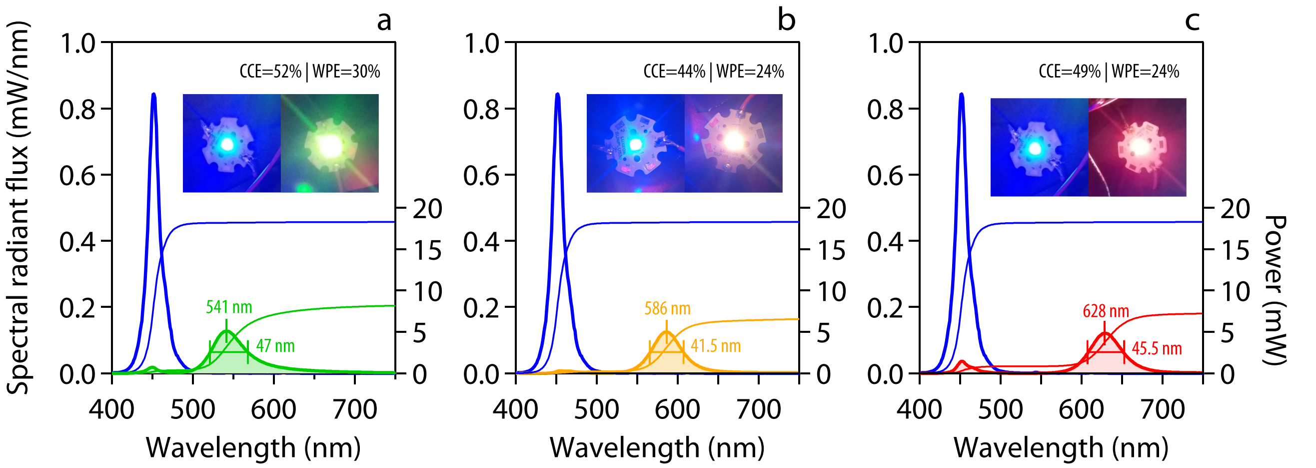

Figure 4. Bridging the green gap by on-chip color conversion with InP-based QDs. (a-c) Spectral radiant flux and emitted power measured on QD-on-chip LEDs formed using (a) green-, (b) amber-, and (c) red-emitting InP-based QDs. In each case, the color conversion efficiency and the wall plug efficiency have been indicated. Note that (a) repeats Figure 2d to enable a direct comparison with the amber-and red-emitting QD-on-chip LEDs.

peak emission by 7−8 nm and reduces the PLQY by 26%; see Supporting Information Section S5. Given the pronounced impact of the PLQY on the light output, this reversible thermal quenching can not only cause an emission line shift but also a considerable output intensity drop with increasing driving current, a known issue for nitride and phosphide-based LEDs.[27−29]

Figure 3b,Figures 3d represent the spectral radiant flux of samples A and B QD-on-chip LEDs, both fabricated to transmit less than 1% of the pump light, as a function of the driving current. The difference between both LEDs is striking. Whereas the sample A LED emits progressively more green light with increasing driving current, the output power of the sample B LED levels off. In addition, the peak emission wavelength shifts by 2.5 and 5.1 nm to the red for samples A and B, respectively. Both observations are in line with the assumed link between a lower PLQY and higher temperature in the convertor film. Still, the minor shift of the LED spectrum and the limited CCE roll-off of the sample A LED (see Figure 3f) indicate that monochromatic QD-on-chip LEDs can combine variable brightness with a stable color point provided that converter coatings with high PLQY QDs are used.

■ BRIDGING THE GREEN GAP BY MONOCHROMATIC QD-ON-CHIP LEDS

InP/Zn(Se,S)/ZnS core/shell/shell QDs can be made with near-unity quantum yield across the visible spectrum from cyan/blue to red.[25] In Figure 4a−c, we show how such QDs enable the results obtained for green-emitting QD-on-chip LEDs to be extended to amber-and red-emitting LEDs; see Supporting Information Sections S2 and S6 for details on the InP-based QDs used and the QD-on-chip LED characterization. For this set of monochromatic LEDs, we thus obtained color-converted spectra featuring an ∼45 nm wide emission line, a CCE in the range 44−52%, and a luminous efficacy varying from 35 to 116 and 161 lm/W for QD-on-chip LEDs with emission lines centered at 628, 586, and 541 nm, respectively. For the blue pump LEDs used here, the CCE corresponds to a wall plug efficiency of (green) 30%, (amber) 24%, and (red) 24%. Using the external quantum efficiency, i.e., the ratio between the number of converted photons emitted and electrons injected, as a benchmark, these monochromatic LEDs outperform electroluminescent QD-based LEDs across the full spectral range analyzed here and (In,Ga)N-based LEDs in the green-gap spectral range; see Supporting Information Section S7. Moreover, the results are outlined in Supporting Information Section S6; the amber and red LEDs have the same limited intensity droop with increasing driving current as shown for the green LEDs in Figure 3f.

Figure 5a represents the three different QD-on-chip LEDs proposed in Figure 4a−c in a CIE1931 chromaticity diagram. In addition, that diagram shows a locus of color points that can be reached by changing the central wavelength of such QD-on-chip LEDs, which we calculated by assuming a 45 nm wide emission line. One sees that the QD-on-chip LEDs are

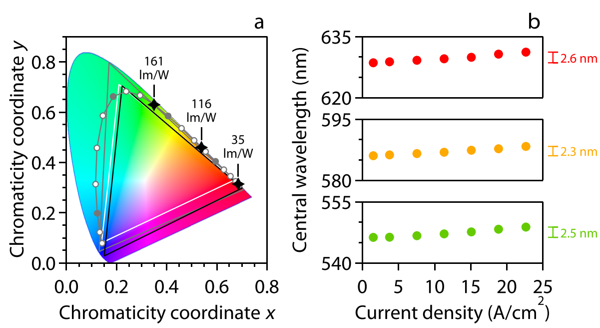

Figure 5. Chromaticity of InP-based QD-on-chip LEDs. (a) CIE1931 chromaticity diagram, including (black markers) the color point of the QD-on-chip LEDs shown in Figure 4, (open gray markers) locus of color points corresponding to a Gaussian emission spectrum with a 45 nm full width at half-maximum and changing peak emission wavelength. Color points are indicated in steps of 10 nm, with central wavelengths at 480, 520, 560, and 600 nm printed as filled markers. The triangles represent (white) the NTSC standard, (gray) the REC2020 standard, and (black) the largest color gamut that can be reached using LEDs with a 45 nm wide emission spectrum and a cutoff filter at 490 nm. (b) Variation of the peak emission wavelength for the QD-on-chip LEDs shown in Figure 4, including the wavelength shift between the highest and lowest operating current densities.

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Nano Lett. XXXX, XXX, XXX−XXX

retrieved on this locus, provided that we eliminate the residual blue LED emission through a 490 nm cutoff filter for the green QD-on-chip LED. As indicated in Figure 5a, we find that two such filtered QD-on-chip LEDs with peak emission wavelengths at 525 and 630 nm can provide a maximum color gamut of 112% of the NTSC standard. Moreover, not unlike the green sample A QD-on-chip LED, the red and amber QD-on-chip LED only exhibit a minor shift of the central emission wavelength with increasing power; see Figure 5b. As a result, the color triangle shown in Figure 5a will hardly change when increasing the LED power; a considerable advantage of these QD-on-chip LEDs given the strong power dependence of the emission wavelength of green InGaN LEDs.[30]

In summary, we report the formation of monochromatic LEDs using on-chip color conversion by InP-based QDs. Regardless of the emission wavelength, these QD-on-chip LEDs have a consistent output spectrum that is about 45 nm wide and show little intensity roll-off or wavelength shift for increasing driving currents. These feats are accomplished by using QDs with a near-unity PLQY, which results in an over 50% color conversion efficiency in the best case. Since the color conversion efficiency is, in large part, limited by package losses, improving light extraction from the convertor layer can further increase the efficiency of these QD-on-chip LEDs. Even so, wall plug efficiencies of up to 30% make the QD-on-chip LEDs demonstrated here competitive with commercial LEDs in the green-to-amber spectral range. This result confirms that QD-on-a-chip color conversion provides a genuine pathway to bridge the green gap in LED technology.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.3c00652.

Additional data on color-conversion efficiency simulations, QD synthesis and characterization, QD-in-resin formation, QD-on-chip characterization, temperature-dependent photoluminescence, characterization of amber and red QD-on-chip LEDs, and comparison of LED performance (PDF)

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Notes

The authors declare the following competing financial interest(s): Ghent University is a shareholder of QustomDot BV. Z.H. is a member of the board of directors of QustomDot BV, representing Ghent University.

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