

Nano-patterned Quantum Dot - LED structures with enhanced photoluminescence emission directionality

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Planar light-emitting structures like displays usually suffer from poor out-coupling efficiency reducing their external quantum efficiency. LED displays based on the electrical injection of charges into colloidal Quantum Dots (QDs) have been predicted to be the next step in display technology. In this work we show that nano-patterning of QD-LED structures can be used to increase the photoluminescence emission directionality and hence improve the out-coupling efficiency and brightness.

Introduction

It is a common problem of planar light-emitting structures that a poor out-coupling efficiency causes the External Quantum Efficiency (EQE) to be only a fraction of the Internal Quantum Efficiency (IQE). Extensive research has been conducted to improve the efficiency of classic liquid crystal displays, as well as for the newer Organic Light Emitting Diode (OLED) technology. LED displays based on the electrical injection of charges into colloidal Quantum Dots (QDs) have been predicted to be the next step in display technology. But while color saturation and IQE can be improved with this technology step, the problem of poor out-coupling efficiency remains an issue.

One root cause is the increased reflection occurring at glass-air interface for larger emission angles. This effect is described by the Fresnel equations and merely dependent on the refractive index contrast, so one pathway that is commonly used to improve the out-coupling efficiency is the modification of the glass surface, by applying methods to achieve a more gradual change of the refractive index.

Also, the high refractive index of the conductive oxides commonly used as transparent electrode causes a slab mode into which a significant fraction of the emitted light can be collected. There is a limit to how thin the electrodes can be due to the requirement of a highly conductive electrode for an electrically efficient device. Thus, introducing a grating by nano-patterning the LED to scatter light from this slab mode, can be another pathway to increased efficiency.

QD-LED design

The layer stack for the nano-patterned QD-LED structure presented here was adapted from an efficient hybrid inorganic-organic design found in literature [1]. Electron injection into the colloidal CdSe/CdS QDs occurs from Indium Tin Oxide (ITO) and a layer of ZnO. Evaporated small molecule organics enable efficient hole injection due to a favourable band alignment. However, due to a temporary unavailability of the organics deposition we replaced these layers with evaporated Al_2O_3 to mimic the refractive index value. Figures 1a and b illustrate the device structure including the grating introduced by the nano-patterned QDs and SiO_2 . The patterned SiO_2 serves a dual purpose, firstly it introduces the necessary refractive index contrast with respect to the layer of QDs. Secondly in a device with organic layers it would also funnel current to the area with QDs, ensuring an electrically efficient device.

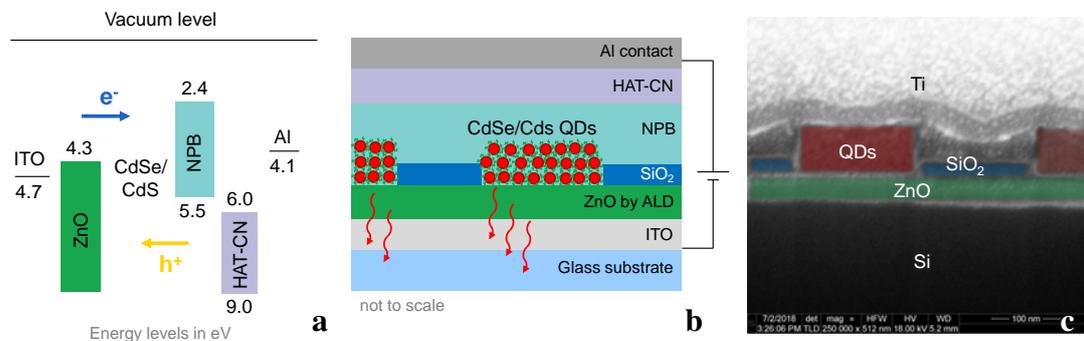


Figure 1: The band alignment of the layer stack in (a) shows the working principle of the hybrid inorganic-organic QD-LED design, electrons are injected from a ZnO layer, holes from an evaporated layer of small molecule organics. The schematic cross-section in (b) illustrates that patterned SiO_2 forms a grating and also focusses the current to the areas of QDs. The FIB/SEM cross-section of a dummy sample in (c) with false colors to highlight the different layers, shows a to scale image of the nano-patterned QD layer.

Optimization of the structure

The 2D geometry found in Figure 1b was implemented and optimised using a commercial FDTD solver (Lumerical). The QDs were simulated by a dielectric layer with an refractive index value extracted from ellipsometry and a single tilted dipole source to obtain a superposition of different polarization orientations. The source was simulated with a Gaussian line-shape centered at 650 nm and a Full Width at Half Maximum (FWHM) of 50 nm, closely resembling the emission spectrum of the colloidal QDs. To reduce the simulation complexity the monitors to record the far field were placed inside the glass substrate and thus to obtain the correct far field emission angles accessible to experiments without an oil immersion objectives one would still need to correct for the glass-air interface. However, since the main purpose of the simulations was only to optimize the design for enhanced emission directionality this was omitted.

Theoretically, a large number of parameters can be varied in the design, however in prac-

tice there are limitations to what can be fabricated and will still work for electrical injection. Also, many parameters were identified to have barely any effect on the emission directionality. The significant parameters for the optimization were the grating period, the fill factor of the grating and the thickness of the organic/ Al_2O_3 layer. A maximum of emission can be found for a fill factor of 0.3 in the simulation, nonetheless a higher fill factor means a higher fraction QD emitters and will therefore lead to a brighter source.

The thickness of the organic/ Al_2O_3 layer also plays a vital role by determining the distance of the aluminium mirror and should be set to achieve constructive interference, while ensuring that device is still able to function electrically. The period of the grating has as expected the dominating effect on the far field, as seen in Figure 2. The optimal values of approximately 80 nm for the organic/ Al_2O_3 layer thickness and of 430 nm for the grating period can be extracted from Figure 2a.

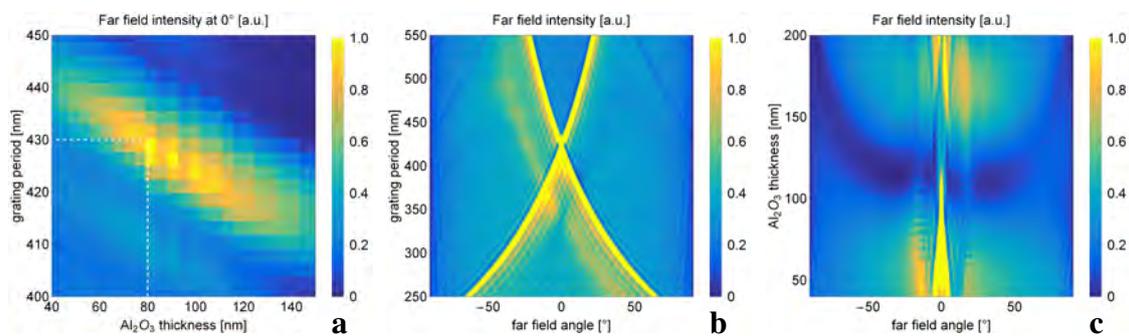


Figure 2: Optimal values for organic/ Al_2O_3 layer thickness and grating period can be extracted from the plot of the far field intensity at 0° in (a). The other plots show the simulated angular dependence of the far field on the grating period (b) organic/ Al_2O_3 layer thickness of 110 nm and the dependence on the organic/ Al_2O_3 layer thickness for a grating period of 430 nm (c).

Fabrication

To fabricate the nano-patterned grating structures we implemented a self-aligned electron-beam lithography processes. For that purpose we used only one lithography step combining Reactive Ion Etching (RIE) of the SiO_2 layer and subsequently the lift-off of spin-coated colloidal QDs. For mass-production more scalable nano-imprint lithography could be applied instead of the electron-beam lithography step. [2] In the structures we used colloidal CdSe/CdS core-shell QDs with a Photo-Luminescence (PL) emission centred around a wavelength of $\lambda = 630$ nm, which red-shifts to $\lambda = 640$ nm for the case of Electro-Luminescence (EL). Figure 1c shows a FIB/SEM cross-section of the nano-patterned QD structure on a dummy sample. The QDs were subsequently covered with a 100 nm Al_2O_3 layer and an aluminium mirror.

Fourier Plane imaging

Since we fabricated the device with an Al_2O_3 layer instead of the organics necessary to obtain EL emission, we investigated the directionality of the PL emission only. Therefore, we excited the QDs with an UV-LED emitting around 405 nm. After passing a 500 nm

short pass filter we used an objective with x50 magnification and a numerical aperture of 0.8 for excitation and collection of the QD emission. A 600 nm long pass filter was inserted to block the reflected excitation light before imaging the Fourier plane on a Thorlabs camera. Comparing the brightness of the emission for different grating periods in the image plane in Figure 3a suggests that a grating period between 375 nm and 400 nm should be ideal. Also the corresponding unprocessed Fourier plane images in Figure 3b show an increased intensity for emission angles near $\theta = 0^\circ$.

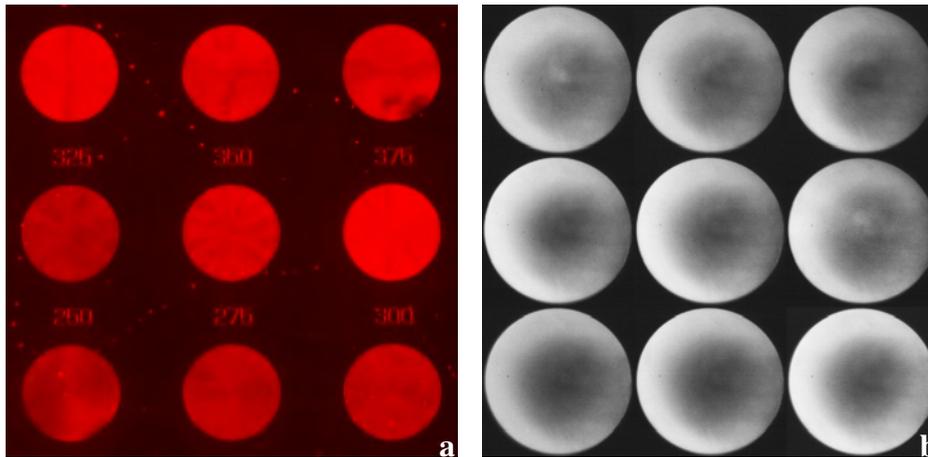


Figure 3: The red color channel of the sample image plane visualized with a x10 objective in (a) shows increased brightness of the structures with a grating period of 375 nm and 400 nm. Also the corresponding unprocessed Fourier plane images from a x50 0.8 NA objective in (b) show an increased intensity for emission angles near $\theta = 0^\circ$.

After correcting for the microscope transmission function using the Fourier plane image collected from a part of the sample where there was only the aluminium mirror and no QDs, the picture becomes even clearer. Figure 4 shows the angular dependence of the far field intensity $I(\theta)$ after integration over $d\phi$. Again, a period between 375 nm and 400 nm will clearly give the highest intensity near $\theta = 0^\circ$ and also the emission directionality for other periods becomes visible. The mismatch of these findings in comparison with the simulation results can be explained by the shift of the QD emission wavelength and Al_2O_3 layer thickness in our fabricated samples.

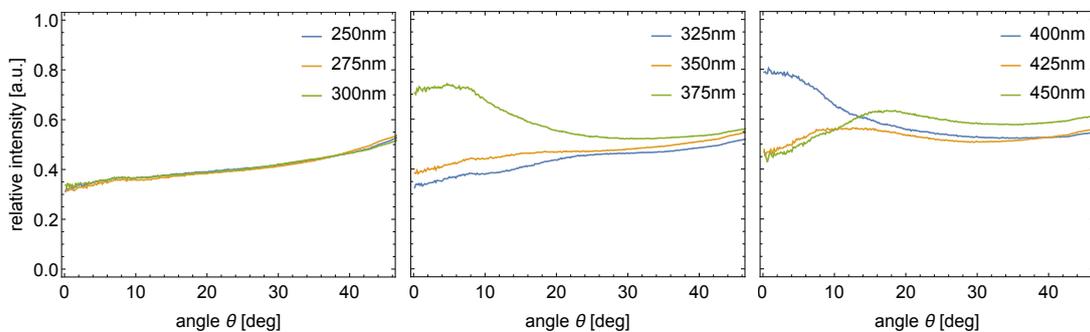


Figure 4: Angular dependence of the far field intensity $I(\theta)$ after correction for the transmission function of the microscope system and integration over $d\phi$.

Conclusion

We designed and numerically optimized nano-patterned QD-LED structures for enhanced emission directionality to improve the external quantum efficiency of these devices. Comparing the brightness of the photoluminescence emission and doing Fourier plane imaging for different grating periods, we showed that there is a maximum of emission directionality for the QD-LED structures with the optimal grating period, in agreement with our simulations.

References

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