Luminescent Cavity Design for High Ambient Contrast Ratio, High Efficiency Displays

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Abstract

A key display characteristic is its efficiency (emitted light power divided by input power). While display efficiencies are being improved through emissive (e.g., quantum dot and organic light emitting display (OLED) designs\textsuperscript{1,2}, which remove the highly inefficient color filters found in traditional liquid crystal displays (LCDs)\textsuperscript{3,4}, polarization filters, which block about 50\% of the light, remain required to inhibit ambient light reflection. We introduce a luminescent cavity design to replace both the color and polarization filters. Narrow-band, large Stokes shift, CdSe/CdS quantum dot emitters are embedded in a reflective cavity pixel element with a small top aperture. The remainder of the top surface is coated black reducing ambient light reflection. A single pixel demonstrates an extraction efficiency of 40.9\% from a cavity with an 11\% aperture opening. A simple proof-of-concept multi-pixel array is demonstrated.

Main Text

Avoiding use of color filters through use of diffraction, interference or surface plasmon effects to manipulate the spectrum of a broadband backlight source has been widely discussed\textsuperscript{5–9}. These solutions, however, require features on the order of the wavelength of light (< 500 nm) and often possess undesirable angular sensitivity\textsuperscript{10}. As such, emissive quantum dot and OLED display concepts have become popular. However, even these emissive display concepts alone do not provide a high ambient contrast ratio\textsuperscript{11,12}. To suppress light reflection from the display, many displays employ a circular polarizer on the display face\textsuperscript{13,14}. In brief, the circular polarizer circularly polarizes external irradiation incident on the display. Upon reflection of this light from an internal surface, its polarization state reverses so that the reflected beam is now blocked by the circular polarizer. While this increases the ambient contrast ratio, light from the display also passes through the circular polarizer, which absorbs about half the emitted light.

In this letter, we present a display pixel design that uses quantum dots (QD) embedded in a polymer matrix in conjunction with a reflective cavity to provide a combined high photon extraction efficiency and ambient contrast ratio. Photoluminescence from the QDs is mostly trapped in the reflective cavity via total internal reflection (TIR) until emitting from a small aperture at the top of the device (the optical physics is similar to that of a luminescent solar concentrator, see the following paragraph). The cavity design takes advantage of the narrow bandwidth emission\textsuperscript{15,16} of QDs to enable photon recycling, and eliminate the need for absorptive color filters. The ambient contrast ratio is maximized by placing a black absorbing layer on top of the pixel (except for over the emission aperture).

Luminescent solar concentrators (LSCs) have been studied since first introduced in 1973 by Lerner\textsuperscript{17,18}. A LSC utilizes a luminophore embedded in a polymer or glass waveguide to absorb incident solar irradiance from all angles (i.e., direct and diffuse sunlight) to create and then transport downshifted photons to a solar cell; the photon transport relies on TIR of the emitted light within waveguide. The decrease in photon entropy required for concentration is provided by the Stokes shift (a loss in photon energy) of the luminophore. Work continues to develop and increase optical efficiencies\textsuperscript{19}, and concentration ratios
greater than 30 have been demonstrated using designs that minimize parasitic losses, including luminophore reabsorption, and maximize the waveguide efficiency\(^{20}\). Here, we take concepts developed for LSCs and “reverse” the design into an emissive display technology. Instead of concentrating photons from a large area towards a small solar cell, here photons from a large area are directed towards a small aperture placed at the top of an optical microcavity.

Figure 1a displays the pixel design. The back surface consists of a distributed Bragg reflector (DBR) designed to transmit excitation light into the cavity while reflecting the Stokes shifted emission. The remaining interior surfaces are highly reflective to support photon recycling. The pinhole on the front surface serves as the extraction point of light from within the cavity. A luminescent material, QD-containing poly(lauryl methacrylate) (PLMA), sits inside the cavity. Figure 1b shows absorption and emission of the QDs used in this study. These core-shell CdSe/CdS QDs absorb strongly in the blue and ultraviolet (black, Fig. 1b) and emit at a center wavelength of 630 nm (red, Fig. 1b). Importantly, these QDs have a very little absorption over the range where they emit, leading to few reabsorption events (reabsorption leads to optical loss). Additionally, the QD emission has a narrow linewidth (full-width at half-maximum, FWHM, of 31.6 nm) eliminating the need for a color filter.

The specific DBR used is composed of a deposited multilayer stack of transparent oxides and is designed to be highly transparent at the excitation wavelength (440 nm) and almost 100% reflective at the emission wavelength of the QDs (630 nm, Fig. 1c). The interior surfaces of an aluminum cap are sputter-coated with silver to create the rest of the reflective cavity, exploiting the high reflectivity of silver at the wavelength range of interest\(^{21}\).

To minimize the ambient reflectivity, an absorbing material consisting of a thin membrane of poly(dimethylsiloxane) (PDMS) mixed with iron oxide nanoparticles (black PDMS, bPDMS) is placed on the external surface of the device. Figure S1b in the Supplementary Information shows the absorbance spectrum of a thin (ca. 50 \(\mu\)m) bPDMS membrane. Figure S3 in the Supplementary Information shows images of such a single-pixel prototype, both in the dark, and illuminated with ambient (laboratory) lighting.

Each pixel can be individually addressed by introducing a blue backlight behind the DBR. In a full-color (i.e., RGB) display, green- and red-emitting QDs would be used to provide the other two colors. Here, we use red QDs with emission at a center wavelength of 630 nm, but the unique optical design principles should be translatable to extended materials sets (i.e., QDs with varying emission) to create a fully emissive display, eliminating absorptive components.

Previous work on LSCs suggests that multiple physical properties of the luminescent element are important\(^{22,23}\). These include a large Stokes shift to prevent reabsorption, a high quantum yield, and low absorption at the emission wavelength. The luminescent element should also not excessively scatter light. Figure 1b shows the large Stokes shift of the CdSe/CdS QDs we used, which minimizes reabsorption. We determine the quantum yield of the QDs after embedding in the polymer matrix is 78%.
These core-shell QDs are similar to those previously used by the team for LSCs; we use these QDs rather than commercial QDs because of the importance of a large Stokes shift for our design, as illustrated in Figure S2\textsuperscript{20}.

\[
\eta_{\text{extract}} = \frac{I_E}{I_S}
\]

We define the extraction efficiency ($\eta_{\text{extract}}$) as follows:

where $I_E$ is the intensity of the emission of the assembled device and $I_S$ is the intensity of the source excitation transmitted through a device (i.e., a reflective cavity) not containing the luminescent film. An integrating sphere fluorimeter equipped with a CCD camera was used to measure light intensities; the measurement setup is explained in Methods. $I_E$ and $I_S$ were calculated by integrating the measured photon counts at the wavelength range of interest after a calibration correction. Control experiments were performed to validate the characterization setup. Bare caps with varying exit aperture areas, defined as the area of the opening over the total area of the top surface, of 2.8% and 11% showed 3.0% and 9.9% of the excitation passing through the cap, respectively. As an additional control, we measured devices otherwise identical to the luminescent devices (reflective Al cap, DBR) containing a QD-free PLMA film and observed 3.0% and 9.9% of the excitation source exiting the device, illustrating the high transparency of the PLMA at the excitation wavelength. Both sets of values agree well with the aperture areas, as expected.

Figure 2a shows the measured spectra of the excitation source (shown in black) and sample photoluminescence (shown in red) from a pixel having an 11% aperture area. The extraction efficiency of the sample is calculated to be 40.9%, which we note is significantly better than the theoretical maximum possible efficiency of an absorptive color filter of 33.3\textsuperscript{%}\textsuperscript{4}. The leakage of the source light out of the pixel was 0.7\%. In the case of a device with even only a 0.17\% aperture area, an extraction efficiency of 35.8\% was achieved with negligible leakage of the source light. Should leakage of the source light be significant, we suggest a bandpass filter could be added on top of the cavity to absorb the source (we did not add such a filter). Images of a single-pixel device containing a black top layer, in the presence and absence of ambient light are shown in Figure S3. Here, the black layer is a bPDMS film placed on top of the pixel.

The experimental data was compared with Monte Carlo ray tracing simulations. Monte Carlo models are regularly used to simulate LSCs to predict device performance\textsuperscript{24,25} and here, we use a modified version of an LSC model used in previous works\textsuperscript{20}. Using the Monte Carlo simulations, we explore factors that affect the efficiency of the devices, the first of which is the aperture area. A larger aperture area results in higher efficiencies due to fewer necessary reflections within the pixel, ultimately minimizing losses associated with multiple non-unity reflections. Larger apertures, however, directly affect how much of the top surface area is covered by the absorptive layer, resulting in a reduced ambient contrast. Hence, there is an inherent trade-off between $\eta_{\text{extract}}$ and the ambient contrast ratio. We measured efficiencies of
devices with six different aperture areas, from 0.17% to 84%, and these results are shown in Figure 2b. The diamonds refer to experimental results and the simulation results are shown as a blue line. Experimentally $\eta_{\text{extract}}$ varied from 36% to 51% as the aperture area increases. The simulation matches the general trend of the experimental data, but there were some discrepancies. We partially attribute this difference to the machined aluminum caps, which introduce deviations from the desired reflectance. We suspect that reflective caps fabricated with high quality, flat optical surfaces would reduce the discrepancy between simulation and experiment. The reflectivity of the caps is shown in Figure S1a. We fabricated multiple caps, and only those that exhibited a reflectivity of at least 90% at 630 nm were chosen, as high reflectivity was a requirement for a high extraction efficiency.

In addition to the aperture area, another important factor affecting $\eta_{\text{extract}}$ of the device is the quantum yield of the luminescent layer. Higher quantum yields result in a higher efficiency due to suppression of non-radiative processes. Figure 2c illustrates the dependence of $\eta_{\text{extract}}$ on quantum yield for a device with an 11% aperture opening. In the case of unity quantum yield, the simulation predicts an efficiency of 57%, vs 39% when the experimentally determined quantum yield of 78% was used in the simulation.

Loss of efficiency in the extraction of photons from the aperture also stems from imperfect reflectance on the interior surfaces of the cavity. A fraction of the emitted light will be absorbed by the metal layer, reducing $\eta_{\text{extract}}$. The simulations assume a reflectance of 96% for the silver-coated interior surfaces, which is comparable to the measured value (Figure S1, Supplementary Information). Figure 2d shows the simulation results of devices with varying interior surface reflectance for three different quantum yields. As expected, $\eta_{\text{extract}}$ increases with surface reflectance due to minimized absorption losses by the metal layer.

One of the most important aspects of a display is its viewing angle. Wide viewing angles are usually preferred for large displays and televisions whereas small, portable electronics typically employ narrower viewing angles focused on directing emission normal to the surface for single-user purpose. We measured the luminance of the device at every 5° and compared the results to a ray tracing simulation (Figure S4, Supplementary Information). The measured luminance shows a broad viewing angle with a dip in the normal direction whereas the simulation predicts the highest luminance at 70°. Since the luminesced light trapped within the waveguide is traveling laterally, the photons having a higher emission angle are more likely to escape the device, explaining the higher luminance at high angles. The deviation from the simulation might be explained by surface roughness on the QD film (by AFM, the surface had a peak-to-valley roughness of ~200nm over a distance of a few micrometers). It is important to note that a fully assembled display is composed of multiple layers, which introduces complexity into the angular emission pattern and additional optics could be utilized to modify the viewing angle.

Finally, the lifetime of this design is important to consider for potential use in an electronic display. Unprotected QDs (i.e., QDs in solution without protective ligands) are sensitive to air, however, incorporating protective ligands and further, dispersing the QDs inside of a polymer matrix minimizes degradation from exposure to oxygen and moisture. To verify the longevity of the proposed device, we
tested the performance of a freshly prepared device and compared its efficiency to the same device after being exposed to ambient conditions (air and white light) for 50 days. The change in emission intensity can be found in Figure S5 in the Supplementary Information. The decrease in extraction efficiency is found to be less than 3% with no change in the leakage of the device. This preliminary stability data is promising for a device based on our QD films.

The single pixel demonstration is useful to optimize the device optics and evaluate the key parameters controlling emission, however, these single pixels are much larger than display pixels, and demonstrate only one, single pixel. Here we apply our concept to a micrometer sized pixel array as outlined in the schematic of Figure 3a. Individual pixels are fabricated by polymerizing QD films inside microfabricated elements in a silicon wafer and are individually addressed with light sources placed underneath each pixel. Figure 3b illustrates the microfabrication process. First, Si$_3$N$_4$ is deposited on the Si wafer and used as a mask layer for a subsequent KOH wet etch. Holes in the Si$_3$N$_4$ are patterned via standard photolithography. An anisotropic etchant, KOH, is used to etch Si, resulting in pixels with angled sidewalls. Holes that pass through the Si wafer are laser drilled. Figures 3c and 3d show SEM micrographs of a fabricated pixel array after laser drilling. The pixel array has square pixels 240 µm on each side and the diameter of the holes are around 80 µm, giving an overall aperture opening of 9%. The pixel array is then sputter-coated with a reflective silver layer. Polymerization of the QD-PLMA inside the holes (Fig. 3e) is performed by using a PDMS slab as a temporary substrate on the back side to prevent leakage of the QD-LMA monomer solution prior to polymerization. The final step is to apply an absorptive material over the external surface of the device; here, we use a thin membrane of bPDMS. The pixel size of our fabricated device is similar to that used in a LCD display, for example, in a common laptop, the display has a pitch of 231 µm which is approximately three times that of our single pixel size (i.e., 80 µm).

A 69-pixel array was used to measure $\eta_{\text{extract}}$ of the optical microcavity array. For the micropixel array sample, we define $I_S$ as the intensity of light emitting from the microcavity array containing empty pixels (prior to polymerization) at the excitation wavelength. The output, $I_E$, is then measured after pixels are filled with polymerized QD-PLMA. $\eta_{\text{extract}}$ is then calculated from equation 1, as before. Figure 3f shows $\eta_{\text{extract}}$ and the excitation source light that transmitted through (defined as the leakage). The $\eta_{\text{extract}}$ of this micropixel array is determined to be 52% with a leakage of 20%. As this leakage is too high for a display application, two strategies to reducing the leakage are to either add a bandpass filter to the top of the display (under the black layer), or to increasing the optical density of the QD-PLMA film. We did attempt to increase the optical density of the QD-PLMA film by increasing the QD concentration, however, undesirable aggregation of the QDs occurred. We did not attempt to apply a bandpass filter to the top of this display.

Images of a 4x3 micropixel array under various ambient conditions are shown in Figure 3g. The image on the left (Fig. 3g-i) is taken when the backlight is on at low ambient light and the light emission is clearly seen from 8 of the 12 pixels (alignment of the black PDMS on the small apertures was challenging, and 4
of the pixels were covered with the PDMS). The image on the right (Fig. 3g-ii) shows the device when the backlight is on under conventional room light. Light emission is still observed from each active pixel (the absorptive bPDMS provides sufficient contrast under ambient light). Once the fabrication process is further improved, performance under ambient light could be quantified.

A prototype of a display utilizing the micropixel array and a custom-made printed circuit board with individually addressable pixels is shown in Figure 4. A 9-pixel device, with pixels matching those in Fig. 3, was formed. In this device, the LED backlights for each pixel can be individually addressed. Since the optical density of the QD film in this demonstration is low, some of the excitation from the blue LEDs passes through the pixel array and a bandpass filter was physically placed on the surface of the micropixel array to block this leaked blue light. The video in the Supplementary Information shows each pixel being sequentially addressed.

Key elements in the luminescent cavity design are high quantum yield quantum dots with a large Stokes shift to reduce reabsorption losses within the luminescent cavities and highly reflective cavity sidewalls. These sidewalls are especially vital to optimize as the luminescence is trapped via TIR modes within the cavity which reflect of the sidewalls of the cavity. As we show, reflectance values < 90% result in low extraction efficiencies and if reflectances can be increased to >98%, $\eta_{\text{extract}} > 50\%$ is possible. This luminescent cavity design has potential to enhance the efficiency of both conventional LCD designs, by eliminating both color filters and circular polarizers, and emissive displays, where it eliminates the circular polarizer.

**Methods**

*Silicon Pixel Array Fabrication*

The microscale pixel array was fabricated from a 200 µm thick (100)-oriented silicon wafer. First, a 1 µm Si$_3$N$_4$ layer was deposited on the Si wafer via LPCVD; this layer served as a mask layer for a subsequent KOH wet etch. The holes (pixels) were patterned via standard photolithography and the Si$_3$N$_4$ layer is then selectively etched exposing Si via inductively coupled plasma reactive ion etching (ICP-RIE). An anisotropic etchant, potassium hydroxide (KOH), was used to etch Si, resulting pyramid-shaped etch pits. After etch completion, holes that permeate from the bottom of the etch pit to the other side of the Si wafer were drilled using a tightly focused laser (YAG laser, Potomac Laser, Baltimore, MD). During the laser drilling, some of the evaporated Si was deposited at the sidewalls. To smooth the sidewalls, a second KOH etch was performed. The pixel array was then sputter-coated with 50 nm of silver as the reflective layer (AJA Orion 3 magnetron sputtering system).

*Extraction Efficiency Measurements*

The extraction efficiency of the device was measured using a custom setup on an optical table. The setup includes a light source (Acton Research Corp 75W Xenon lamp with an Opti-Quip 1200 power supply), a monochromator (Jarrel-Ash M-20, slit size 0.5 mm) and an integration sphere (Labsphere RTC-060-SF). A
200 μm optical fiber (Thorlabs FT400EMT) was used to couple the output light from the integration sphere. The light was measured by Acton Research SpectroPro3001 spectrometer equipped with a calibrated Acton Pixis 100 CCD camera. To compensate the different spectral response of the detection system, it was calibrated with a Labsphere halogen standard light source IRF G3 (NIST traceable). At the start of the experiment, the dark background reading was collected and deducted from the subsequent readings. The input intensity was calculated by measuring the excitation light set at 440 nm when the light path is unobstructed. The output intensity is measured after introducing the device on the light path. Emission and leakage spectra were taken between 370 nm and 710 nm. The area under the spectrum reading between 615 and 690 nm was defined as emission intensity whereas the area around 440 nm was defined as leakage. The ratio between emission and input defines the extraction efficiency.

*Angular Fluorescence Detection*

An Argon laser emitting at 488 nm was used as excitation source. The laser beam went through a beam expander to increase the spot size and hit the sample stationed at the center of a custom-made swivel arm. At the end of the arm (10” from the center) sits a fiber optic cable connected to a Si spectrometer. Intensity measurements were recorded at every 5 degrees and fluorescence intensities were integrated using a computer script.

*Quantum Dot Synthesis*

Quantum dots were synthesized as described in detail previously.

*Quantum Dot Film Fabrication*

A solution of the monomer, lauryl methacrylate (LMA), is prepared with the cross-linker, ethylene glycol dimethacrylate (EGDMA), in a 10:1 volume ratio LMA:EGDMA. Trioctylphosphine, the ligand on the CdSe/CdS QDs, is also added at 4 vol.% to aid in stabilizing the QDs within the matrix and avoid aggregation. QDs in hexanes (5 mg/mL) are then added to the monomer solution in a glovebox at 15 vol.%. The hexanes are allowed to evaporate for > 1 hour. Concurrently, two quartz plates are prepared by treating them with Repel Silane ES (GE Healthcare). Silica beads (SPI, 500 μm) are then used as spacers and placed at the corners of the quartz plates. This assembly is then brought into the glovebox where the initiator, 2-hydroxy-2-methylpropiophenone, is then added at 0.05 vol.% The monomer solution is then filled via capillary action between the two quartz plates and the QD-LMA solution is UV-cured (365 nm, 4 W) for 40 minutes.

*Fabrication of bPDMS*

Thin bPDMS membranes are prepared on bulk PDMS substrates for support. All clear PDMS is prepared by mixing Sylgard 184 (Dow Corning) Parts A (base) and B (initiator) in a 10:1 ratio, respectively. Bulk clear PDMS is first prepared and the mixture of Parts A and B is degassed and cured in a 70°C oven for > 3 hours. The bulk PDMS substrate is then treated with No-Stick [trichloro(1H,1H,2H,2H-
perfluorooctyl)silane, Sigma Aldrich] and a thin layer of clear PDMS is then spin-coated on top at 1500 rpm. This layer is cured for one hour at 70°C. Black PDMS (bPDMS) is then prepared by combining the Sylgard 184 base with Fe₃O₄ powder (Earth Pigments) in a 2:1 mass ratio. This mixture is combined with a centrifugal mixer (THINKY MIXER, ARE-310) at 2000 rpm for 2 minutes and then manually mixed thereafter. After the mixture homogenizes, the Sylgard 184 initiator is added in the same 10:1 part A:B ratio used for clear PDMS. This solution is then mixed and degassed. The bPDMS is then spin-coated at 5000 rpm and cured at 70°C for one hour. The membrane is encapsulated with a final layer of clear PDMS spin-coated at 4000 rpm and cured at 70°C for one hour. The finished membrane can then be peeled off of the bulk PDMS substrate. Holes in the bPDMS can then be manually punched out to be placed atop the single pixel or multipixel demonstrations.

**Declarations**

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**Author Contributions**

O.S.C. and M.A.Y., under the supervision of P.V.B. and R.G.N., respectively, designed the study. O.S.C. and M.A.Y. performed the experiments, analyzed and interpreted data and co-wrote the manuscript. B.A.K, Z.N., J.K.S., and A.P.A. provided quantum dots. H.C. helped with the optical characterization setup. C.J.B. designed the circuit board for illumination. J.H. advised on modeling and experimental setup and interpretation. L.X advised and helped developed procedures for QD encapsulation in the polymer. All authors reviewed the manuscript.

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**Figures**
Figure 1

(a) Top and cross-sectional schematic of a single-pixel luminescent cavity. (b) CdSe/CdS QD absorbance (black) and emission (red). (c) DBR reflectance vs. wavelength.
Figure 2

(a) Excitation intensity (black) and photoluminescence intensity (red and inset) of the single pixel luminescent cavity. (b) Extraction efficiency vs. aperture areas of the single pixel luminescent cavity. Experimental datapoint are in black and a Monte Carlo simulation is shown in blue. (c) Simulation of $\eta_{\text{extract}}$ for QDs with varying quantum yields. (d) Simulation of $\eta_{\text{extract}}$ for varying cap wall reflectance and quantum yields (QY).
Figure 3

(a) Schematic of a micropixel array and a micropixel. (b) Micropixel fabrication process. (c) SEM image of the Si micropixel array after laser drilling. (d) SEM image of a single micropixel after laser drilling. (e) Photograph of the micropixel array containing 69 QD-containing polymer filled pixels (f) Input excitation intensity (black) and photoluminescence intensity (red) emanating from the micropixel array. (g) Images
of a 4x3 pixel micropixel array (i) in the dark when the excitation is on and (ii.) under substantial ambient illumination while the excitation is on (main figure) and off (inset).

**Figure 4**

Images of a 3x3 micropixel array where individual pixels are illuminated to create patterns including (a) four separate pictures depicting a U, I, and C, spelling out UIUC (b) a diagonal line (c) an ‘x’ and (d) a ‘+’

**Figure 4**

Images of a 3x3 micropixel array where individual pixels are illuminated to create patterns including (a) four separate pictures depicting a U, I, and C, spelling out UIUC (b) a diagonal line (c) an ‘x’ and (d) a ‘+’
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