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Soft holographic interference lithography microlens for enhanced organic light emitting diode light extraction

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Abstract
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Keywords
Ames Laboratory, Physics and Astronomy, Electrical and Computer Engineering, Microelectronics Research Center

Disciplines
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Comments
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Abstract: Very uniform 2 μm-pitch square microlens arrays (μLAs), embossed on the blank glass side of an indium-tin-oxide (ITO)-coated 1.1 mm-thick glass, are used to enhance light extraction from organic light-emitting diodes (OLEDs) by ~100%, significantly higher than enhancements reported previously. The array design and size relative to the OLED pixel size appear to be responsible for this enhancement. The arrays are fabricated by very economical soft lithography imprinting of a polydimethylsiloxane (PDMS) mold (itself obtained from a Ni master stamp that is generated from holographic interference lithography of a photoresist) on a UV-curable polyurethane drop placed on the glass. Green and blue OLEDs are then fabricated on the ITO to complete the device. When the μLA is ~15 × 15 mm^2, i.e., much larger than the ~3 × 3 mm^2 OLED pixel, the electroluminescence (EL) in the forward direction is enhanced by ~100%. Similarly, a 19 × 25 mm^2 μLA enhances the EL extracted from a 3 × 3 array of 2 × 2 mm^2 OLED pixels by 96%. Simulations that include the effects of absorption in the organic and ITO layers are in accordance with the experimental results and indicate that a thinner 0.7 mm thick glass would yield a ~140% enhancement.

OCIS codes: (080.3630) Lenses; (230.3990) Microstructure devices.

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1. Introduction

Despite their high electrical power-to-light conversion efficiency, there is an innate limitation to light extraction from LEDs and organic LEDs (OLEDs) due to the interface between the light emitting materials and air. The fraction of light which escapes in the forward direction is

$$\eta_{ext} \sim (1-\cos\theta_c) \sim \frac{1}{2n_{org}{}^2}, \qquad (1)$$

where θ_c is the organic-to-air critical angle and n_{org} ~1.7 is the refractive index of the organic layer(s) [1]. Hence, for typical OLEDs η_{ext} ~0.17; for GaN with n_{GaN} ≈3.5, η_{ext} ~0.04. The majority of light is either reabsorbed by the materials or leaks out from the device edges where photon recapturing for useful emission has proven problematic.

The use of microlens arrays (μLAs) is a notable approach to overcome the foregoing outcoupling limits [2–11]. However, fabrication of such arrays is either not economical or the resulting outcoupling enhancement is reported to be < 80%, especially if the array is either confined to an area directly under the pixel [2–5,7] or that area under the pixel is excluded [8]. In this work we show that if the microlens array is of high quality and much larger than any of the pixels in the OLED array, the outcoupling enhancement will be ~100% in the forward direction, i.e., higher than any reported to date. We also demonstrate a very low-cost procedure to fabricate such high-quality arrays. Simulations indicate that a thinner 0.7 mm thick glass substrate would yield a ~140% enhancement. In addition, the μLA yields more diffuse light, which is advantageous for some applications.

2. Experimental procedure

The highly ordered, uniform, and economical μLAs are fabricated by soft lithography imprinting of 2 μm-pitch square arrays, embossed on the blank side of a 1.1 mm thick indium-tin-oxide (ITO)-coated glass [11]. Thus, this technique does not interfere with the device fabrication process. In this method, we generate a structure with a polydimethylsiloxane (PDMS) mold, which has the desired inverse relief pattern. A suitable material, in this work a tiny drop of UV-curable Summers Optical Type J-91 polyurethane (PU), is then applied to the desired surface, and the PDMS mold is simply stamped on it, creating the desired pattern. Any excess of PU was easily removed. Once the PU was cured in a UV chamber, the PDMS mold was lifted off and the μLA pattern was formed. For a feature size of a few microns, PDMS is an excellent choice because of its flexibility and non-wetting properties [12]. A master stamp is used for generating the PDMS mold; we fabricated it using two-beam laser (interference) holography patterning on a photoresist, as detailed elsewhere [13] and described briefly below. As long as the master stamp is not physically damaged, it can be used repeatedly for making more molds, each of which can also be used many times until it wears out.

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The μLA master stamp was fabricated using an AZ 6612KE photoresist that was spin coated at 1000 rpm on a glass substrate and then placed on a 110°C hot plate for 60 s to remove all solvent. It was next mounted on the sample holder of a 2-beam UV laser lithography system. The first exposure created a 1-D pattern; the desired 2-D pattern was achieved by a second exposure after rotating the sample by 90°. The photoresist was then developed for 60 s in an MIF 300 developer. To generate a spherical array structure, the sample was further heated to 140°C on a hot plate for 60 seconds. Finally, a 100 nm gold layer was deposited on top of the photoresist. This gold film acted as an electrode for subsequent nickel electroplating. A total thickness of 10 μm of nickel was electroplated (Caswell Plating) and the whole nickel film was detached from the glass by dissolving the photoresist. This thin nickel sheet with the microlens pattern facing up was glued to a glass substrate for molding purpose. Next, PDMS was poured onto the master stamp, to generate a mold with the desired inverse relief pattern. After the PDMS was cured and solidified, it was peeled off of the master stamp.

OLED pixels were fabricated on the ITO side of the glass. The ITO was patterned and etched to form anode stripes. It was then thoroughly cleaned (prior to the μLA fabrication) with detergent and organic solvents and treated in a UV/ozone oven to increase the ITO work function. Following the μLA fabrication, it was rinsed with isopropanol. This step was followed by thermal deposition of the organic layers, CsF buffer layer, and Al cathode in a vacuum evaporation chamber (background pressure $< 5 \times 10^{-6}$ torr) inside an Ar-filled glove box. The organic layers consisted of a copper phthalocyanine (CuPc) hole injecting layer, an N,N'-diphenyl-N,N'-bis(1-naphthylphenyl)-1,1'-biphenyl-4,4'-diamine (NPD) hole transport layer, an emitting layer of various materials, and tris(8-hydroxyquinoline) Al (Alq_3) electron transport layer. Two different emitting materials were used: green emitting Alq_3 and blue emitting 4,4'-bis(2,2'-diphenylvinyl)-1,1'-biphenyl (DPVBi). The final Al cathode was deposited through a shadow mask with 3 mm-wide stripe openings; the Al stripes were perpendicular to the ITO stripes. As a result, the OLED pixels were defined by the overlap of the ITO and Al stripes. Half of the pixels were on the portion of the plain glass substrate and the rest were on the glass covered with the μLA. The devices were encapsulated by placing a similar size glass slide over them and sealing with Torr Seal epoxy around the edges of the glass slides. Wires were connected to the electrodes of each pixel.

Measurements were performed by placing the devices on the d_a = 5, 10, or 25 mm opening of a 75 mm diameter Sphere Optics integrating sphere. The signal collected from the sphere was transmitted through an optical fiber to a Model S2000PCI Ocean Optics spectrometer for analysis.

3. Results and discussion

Figure 1(a) shows the fabrication process of the microlenses on the glass surface. Figure 1(b) shows the SEM image of the photoresist pattern after developing. The patterned area can be quite large (several cm^2), limited by the geometry of the optics used for pattern generation and the incidence angle [13]. As seen in Fig. 1(b), the top of the structure is not quite as spherical as that for an optimal microlens, but this is remedied by heating the sample on a hot plate to 140°C for 60 seconds. Note that such an inverse pattern was reported as a light extraction layer for an InGaN LED [14]. Figure 1(c) is an SEM image of the μLA having spherical shapes. This array covers only a portion of the ITO/glass. OLED fabrication on the ITO side of the glass is detailed elsewhere [15–17]. Measurements were performed as shown in the supporting information.

Figure 2 shows two energized (a) green Alq_3- and (b) blue DPVBi-based OLED pixels lit at the same current density. Only the left pixels in each case are under a μLA. Note the much larger size of the μLA in comparison to the OLED pixels. As seen, the left pixels appear much brighter, but the light around them is diffuse. The pixels on the right appear much sharper with a defined square shape. The defocused images and flaring intensity of the left pixels demonstrate the extraction enhancement due to the microlenses. Furthermore, by closely examining the right green and blue pixels, it is seen that these pixels also contribute to the

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enhanced light extraction due to the adjacent μLA. This indicates that the μLA extracts light from the glass substrate as intended. Turning to the images' outer rims, it is seen that the right rims are much brighter than the left; the uneven contour around the edges is due to the uneven encapsulating epoxy (the two horizontal black strips in each image are the Al stripes). The dark left rims are clearly due to the extraction of the waveguided light by the μLA.

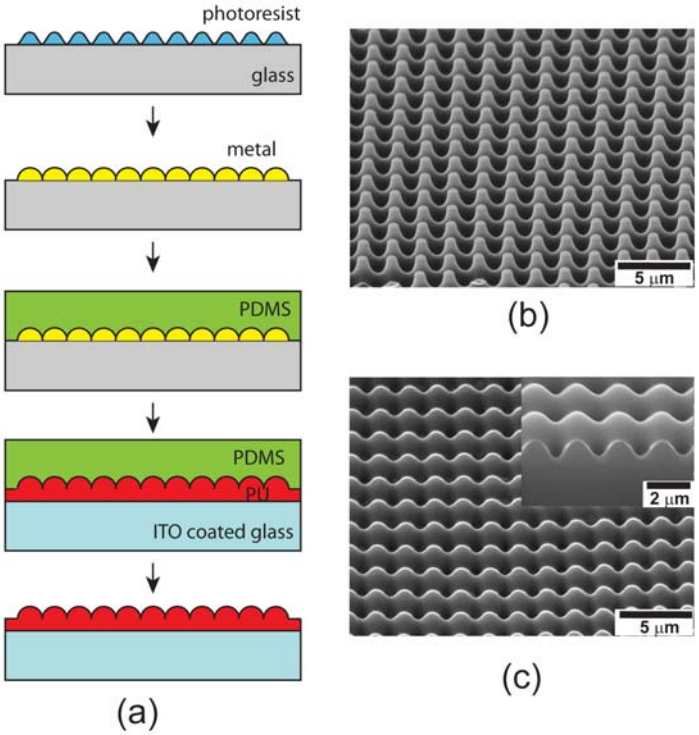

Fig. 1. (a) Schematic of μLA fabrication on glass. A master template is covered with PDMS. The PDMS is removed from the master and then pressed against a PU drop on another glass substrate. The PDMS is lifted off and the PU microlens array remains on the glass substrate. (b) SEM image of the 2D patterns of photoresist and (c) the resulting PU microlens array.

Figure 2 also shows the EL spectra of the (c) Alq_3 and (d) DPVBi OLEDs, each taken with only one pixel energized and with different d_a. We note that by comparing the normalized spectra (not shown) we confirmed that the lineshapes without and with the μLA were nearly identical. Hence, the enhancement of the integrated EL intensity is proportional to the enhancement of the peak EL amplitude, and since it is measured with an integrating sphere that collects all of the emission in the forward direction, so is the external quantum efficiency. We also integrated the intensity over the EL spectrum and, as expected, confirmed that the enhancements are those that occur at the peak wavelength. As seen, the EL intensity I_{EL} from the OLED pixel under the μLA increases with increasing d_a, as more of the light extracted by the μLA from outside the pixel area is collected, whereas it is unaffected by d_a for the reference pixels. When d_a = 25 mm, it collects essentially all the light extracted by the μLA and the enhancement is ~100%. That is, the large μLA area outside the OLED pixel area accounts for the increased enhancement. However, as seen in Fig. 2(c), increasing d_a from 5 to 10 mm increased I_{EL} less than four fold. The reason is that the waveguided light within the glass substrate is not uniform: Due to the extraction by the μLA, the intensity of the waveguided light, integrated over the circumference of radius r (where r is the distance from the pixel), decreases as r increases. The EL intensities of the reference sample did not vary with d_a. Figure 2(d) shows the same enhancement for a blue DPVBi OLED using the same PDMS mold, demonstrating that the enhancement is wavelength-independent. Moreover, the

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EL spectra with the plain glass substrate and the μLA-covered glass are identical after normalized at peak intensities, demonstrating the dispersionless behavior of the microlenses.

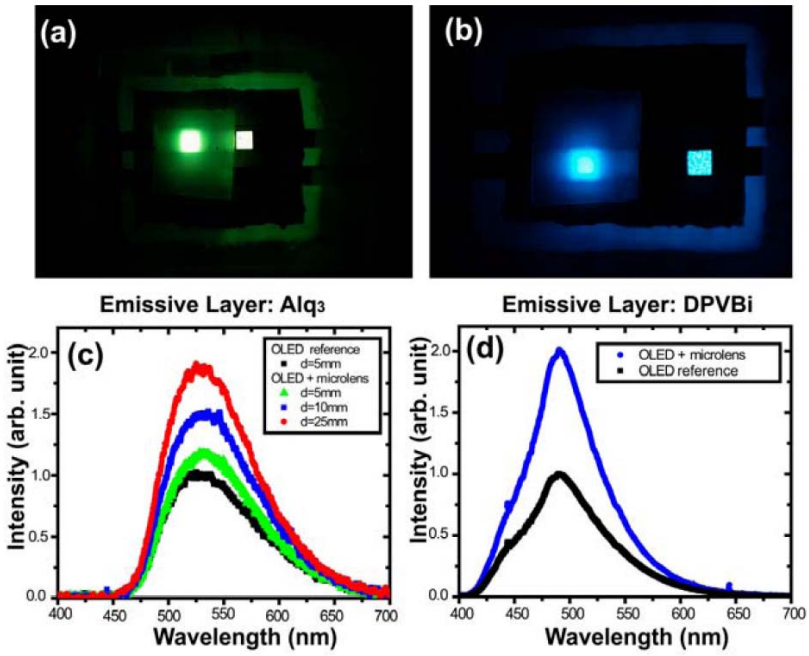

Fig. 2. Images of two OLED arrays with (a) green emitting Alq_3 and (b) blue emitting DPVBi. The left side pixels in each image are under a microlens array and the right ones are reference pixels. The surrounding (rim) lines are the epoxy sealant used for OLED encapsulation. (c) EL spectra of the Alq_3-based OLED with a PU microlens array measured with different apertures of an integrating sphere. (d) EL spectra of a DPVBi-based OLED with microlenses measured with a 25 mm diameter integrating sphere aperature. The black lines in (c) and (d) are the reference spectra of nominally identical OLED pixels without the microlenses.

We used a three dimensional (3D) ray tracing method to trace a large number of rays generated at random positions, polarizations, and angles in the pixel region. At each material interface, the reflectance and transmittance were calculated from the Fresnel equations. Then we let a ray randomly choose reflection or transmission with probability equal to the reflectance or transmittance, respectively. The effects of absorption in the aluminum electrode, organic and ITO layers were included in the simulations. For conventional OLEDs with no μLA, we obtained η_{ext} ~15%, which is close to the 17% predicted from geometric optics without considering absorption in the system. Next we simulated the case of a μLA with 2 μm period, 1.2 μm height, and 1.6 μm diameter. Once the light is trapped inside the glass, it can escape through the side of the glass unless absorbed. The μLA changes the incidence angle and extracts the trapped light (see Fig. 3(a)). The forward emission intensity, normal to the glass surface, is the sum of the extra extraction due to the μLA and the pixel (diffuse) emission (see Fig. 3(b)). The forward intensity of an OLED pixel without the μLA is shown for comparison in Fig. 3(c); η_{ext} increases as the μLA area increases and it saturates when all guided light in the glass is extracted. A μLA area of 25 × 25 mm^2 can extract most of the light trapped inside the glass. Further increasing the μLA area increases η_{ext} by only another 3%.

For a given microlens patch, the glass thickness can be varied to optimize the extraction efficiency; if it decreases from 1.1 to 0.7 mm, the calculated enhancement increases from 121% to 140%. The number of reflections in the μLA increases when using a thinner glass, and hence the increased light extraction. Table 1 summarizes the simulations results for different values of the μLA area and glass thickness. The simulation results agree well with

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experiments except for the 10 mm aperture case. The discrepancy lies in the fact that the microlenses in the experiment are semi-ellipsoids whereas the simulations assume perfect closely packed semispherical lenses.

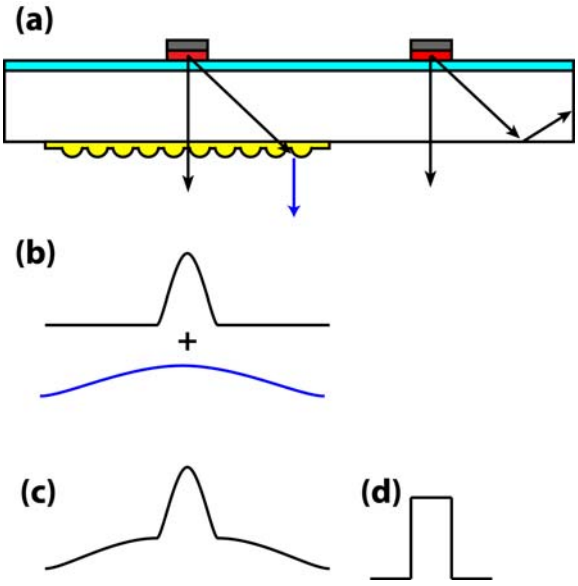

Fig. 3. Schematic of light extraction model with microlenses: (a) left: extraction enhancement by the microlens array due to incidence angles change that reduces the total internal reflection inside the glass; right: light waveguiding in the glass in the absence of the microlenses. (b,c) Forward EL intensity of an OLED pixel with the microlens array that is the sum of the slightly diffused emission at the pixel area and the extra emission due to the microlens. (d) Forward intensity of an OLED pixel without microlenses.

The images and the results shown in Fig. 2 demonstrate that for any OLED array size, a μLA that is large relative to the size of a single OLED pixel will enhance η_{ext} by ~100%. To demonstrate this statement, 9 pixels with and without a μLA were made side by side on a same substrate and their EL spectrum was measured (see Fig. 4). Each pixel area was 2 × 2 mm^2, as it was defined by the overlap between the 2 mm Al cathode stripes and the 2 mm ITO anode stripes, as shown in Fig. 4 insert. The μLA area was 19 × 25 mm^2 and EL spectra were measured with d_a = 25 mm. As clearly seen, the intensity with μLA was 175/90 = 1.94 fold that of the reference, i.e., the μLA enhanced the emission of the OLED array by 94%.

Table 1. (a)–(c) Calculated η_{ext} Enhancements with Various μLA Areas, Glass Thickness, and Integrating Sphere Apertures[a]

d_a (mm)	5	10	25
(a)15 × 15 mm^2 μLA, 1.1 mm thick glass (calc.)	21%	73%	97%
15 × 15 mm^2 μLA, 1.1 mm thick glass (exp.)	18% (Alq_3)	54% (Alq_3)	92% (Alq_3) 100% (DPVBi)
(b) 25 × 25 mm^2 μLA, 1.1 mm thick glass (calc.)	23%	75%	121%
(c) 25 × 25 mm^2 μLA, 0.7 mm thick glass (calc.)	48%	96%	140%

[a]Measured EL enhancements for Alq_3- and DPVBi-based OLEDs are included in (a) for the comparison.

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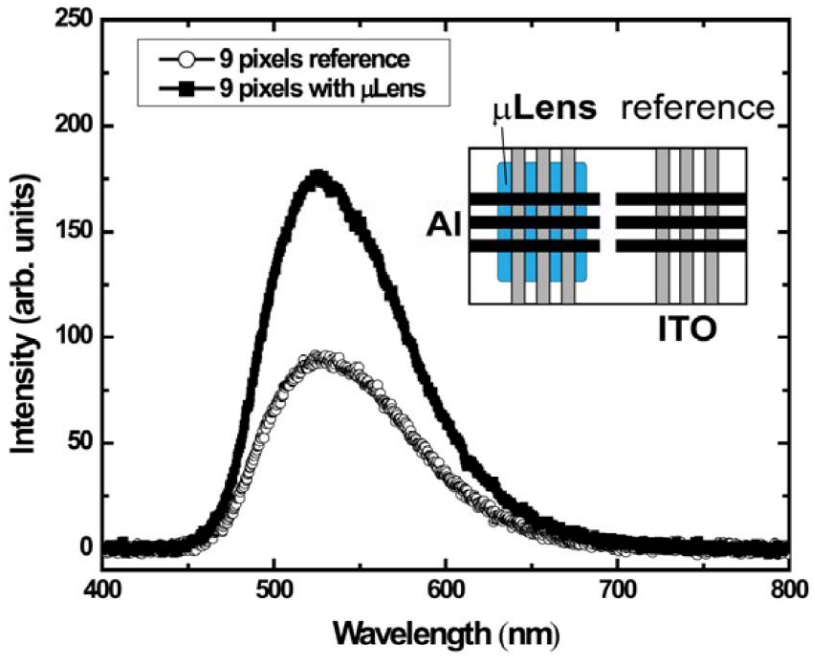

Fig. 4. The EL spectra of the 9 pixels without (open circles) and with (solid squares) the 19 × 25 mm^2 PU μLA, and the EL spectrum of the 9 pixels with the μLA (open triangles), obtained from an integrating sphere with 25 mm aperture. All devices were driven at 8 V. Each OLED pixel is 2 × 2 mm^2, and adjacent pixels are separated by a 2 mm gap.

4. Summary and concluding remarks

In conclusion, we have shown that a uniform 2 μm-pitch square PU μLA originating from soft lithography delivers a η_{ext} enhancement of ~100% from a single OLED pixel if the μLA area significantly exceeds that of the OLED pixel. Similarly, a ~94% enhancement is obtained for an OLED pixel array covered by a ~5 fold larger area μLA. We note that the minimal size ratio required to achieve this larger enhancement has not yet been determined. By a suitable choice of the glass substrate, such as thin glass substrate with high refractive index, an even higher η_{ext} enhancement can be achieved. Other than the observed high η_{ext} enhancement, the fabrication technique of the high-quality holographic interference lithography μLA is very economical and may provide a low cost means for device manufacturing. Classical ray-tracing simulations support these conclusions. Moreover, the use of μLAs provides a more diffuse light source that is advantageous for diffuse lighting purposes.

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