A theoretical and experimental investigation of light extraction from polymer light-emitting diodes

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(Received 24 September 2004; accepted 10 January 2005; published online 9 March 2005)

Using numerical modeling techniques we predict the emission into air, substrate, polymer/indium tin oxide (ITO), and surface-plasmon modes of a polymer light-emitting diode. The results give good insight into the possible efficiency increases that can be expected for various light extraction techniques. In addition, the effects of various optical properties and layer thicknesses on the optical performance of the device are reported. We show how modification of the substrate can be used to concentrate light into mode types from which light can be easily extracted. We then report experimental results for two very different light extraction techniques. First, we demonstrate how Bragg gratings can be used to extract light from waveguide modes in the polymer/ITO layers. With an optimized Bragg grating, we have increased the external power efficiency by 25% at high brightness levels. In addition, we have used substrate-shaping techniques to extract light from waveguide modes in the glass substrate. In this case, we were able to increase the efficiency by over 60%. Finally, we compare our numerical and experimental results to summarize the possible benefits of various light extraction strategies. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1866487]

I. INTRODUCTION

The majority of the light generated in the emissive layer of a polymer light-emitting diode (LED) is trapped by the total internal reflection and waveguided along the device until it is eventually absorbed and ultimately wasted.[1] A number of techniques have been demonstrated to extract the waveguided light in order to increase the external efficiency. These include microcavities that alter the directionality of emission,[2,3] substrate modifications that enable extraction of light trapped in the substrate,[4–10] and the introduction of refractive index perturbations that outcouple trapped light from the organic/indium tin oxide (ITO) layers.[11–15] While many techniques have been proposed, there is still no general consensus on the best method(s) for extracting trapped light. Optical models have been developed to calculate how the emission is modified by the LED structure.[16–20] Yet there is also a great deal of confusion regarding what fraction of light is actually trapped and what enhancements can be expected from the various extraction techniques. This is due in part to the wide variety of structures that have been investigated, each with different photonic structures, emitting layers, and extraction techniques.

In this paper, we model an optimized LED structure to calculate how much power is emitted into the waveguide modes. We then present experimental results for two extraction techniques with this structure. First, we incorporate Bragg gratings into the anode to extract light from waveguide modes confined in the polymer and ITO layers. We discuss the issues necessary for scattering light out of the waveguide before absorption occurs. Next we use lenses to extract light from waveguide modes in the substrate. We compare and contrast the performance, features, and issues associated with each approach, along with the possible methods of low-cost fabrication.

II. EXPERIMENT

In this paper we show the results of numerically modeling the coupling of power into various optical modes using classical electrodynamics.[18–20] In general terms, we calculated how the emission pattern of an excited dipole is altered by the surrounding multilayer structure. From the modified emission pattern, we calculated what fraction of the total power is emitted into each mode type. For details we refer the readers to the papers cited above. The complex refractive indices of each layer were included as a function of wavelength. The anisotropy of the polymer layers was also taken into account. The results were computed over the entire emission spectrum of the polymer. The internal emission spectrum of the emissive layer was calculated from the transmission and photoluminescence spectrum of a thin film on glass. All the numerical modeling was done in the MATLAB environment on a desktop computer.

The reference LED structure is shown in Fig. 1. The molecular structure of the LUMATION Green 1304 light-emitting polymer (LEP), available from the Dow Chemical Company, has not been published, but the properties of similar materials have been reported.[21] LEDs were fabricated in a nitrogen environment on soda-lime glass substrates coated with 150-nm-thick ITO films. The conducting polymer poly(3,4-ethylenedioxythiophene)–poly(styrenesulfonate) (PEDOT–PSS Bayer CH 8000 from H.C. Starck) was filtered before spincoating using a 0.45-μm polyvinylidene flouride (PVDF) filter. The LUMATION Green LEP was deposited by spincoating from a *p*-xylene solution followed by thermal evaporation of 5 nm of calcium and 50 nm of silver.

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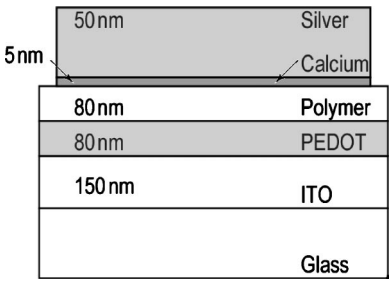

FIG. 1. The LED structure studied in this paper with the optimized thickness for each layer.

The thickness of the metal layers was monitored during deposition with a quartz-crystal monitor. The devices were encapsulated using glass and UV-cured epoxy so that measurements could be performed in air.

Bragg gratings were fabricated by holographic lithography using 150-nm-thick films of Shipley S1805 resist and the 325-nm line from a helium–cadmium (HeCd) laser. We transferred the grating pattern into the ITO by reactive-ion etching (RIE) in a Freon-22 ($CHClF_2$) plasma. The grating periods were between 380 and 400 nm and the depth was 65 nm. The gratings were characterized with both scanning electron (SEM) and atomic force (AFM) microscopes.

We fabricated our extraction lenses by placing small drops of UV-curable epoxy (Norland Optical Adhesive 81) on the substrate above our devices. Surface tension causes the droplets to form small hemispheres above the active area of the LED. The LEDs were rectangular stripes 5 mm long and 1 mm wide. The lenses were about 2 mm in diameter, and two or three drops were used to cover the length of the LED stripe. The epoxy was cured in air with exposure to 365-nm light for 5 min. The UV light did not degrade the performance of the devices.

External efficiencies were obtained by collecting the emission from the LEDs with an integrating sphere and measuring with a photodiode. The entire system was calibrated by coupling the integrating sphere to a spectrometer with an optical fiber and comparing the emission from the LEDs to the emission spectrum of a calibrated white light source. Emission from the sides and back of the glass substrates was prevented by blackening the edges.

The angular emission profiles were obtained by collecting the emission with a silicon photodiode. The devices were placed on a rotation stage to vary the emission angle. The collection angle ($<1°$) was controlled by placing an iris in front of the photodiode.

III. RESULTS AND DISCUSSION

A. Numerical modeling

In order to properly choose an extraction technique for a specific device structure, it is important to know what fraction of the emitted light is trapped in the waveguide modes. Most light extraction techniques are only valid for a specific mode type. For example, roughening the substrate scatters out the light trapped in the substrate, but it does not affect the light waveguided in the polymer/ITO layers. Comparing the fraction of the light in each mode type lends insight into which extraction method could provide the largest improvements in external efficiency. In addition, the calculated power can be used as a benchmark for judging the performance of a particular extraction technique.

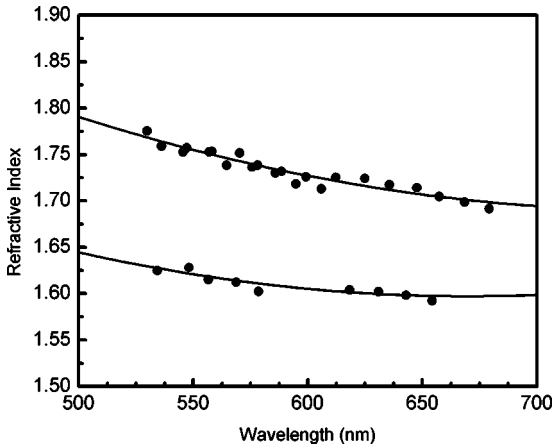

FIG. 2. The refractive index of a LUMATION Green 1304 LEP film as a function of wavelength. The refractive index is uniaxial, with a larger index in the plane ($n_{\parallel}$) and a smaller index normal to the plane of the film ($n_{\perp}$).

In this paper, we have calculated how the emitted power is distributed among four mode types. *Air* modes contain all the light that escapes the device as useful radiation. *Substrate* modes contain all the light that is trapped and waveguided in the substrate. *Polymer/ITO* modes contain all the light that is trapped and waveguided in the active layers and ITO. *Plasmon* modes contain all the light that is emitted into guided optical modes at the polymer/cathode interface (surface-plasmon polaritons)[22] plus light that is emitted into lossy surface waves.[23]

Our polymer LED structure is shown in Fig. 1. This high-efficiency device contains a LUMATION Green 1304 LEP sandwiched between a calcium/silver cathode and a PEDOT/ITO anode. We typically achieve 3.5% external quantum efficiencies and 6–7 lm/W power efficiencies with this structure. We have carefully measured the refractive index of the LUMATION Green 1304 LEP with a grating outcoupling technique described previously.[24] The anisotropy and dispersion of the refractive index are plotted in Fig. 2 and must be taken into account in order to accurately model the device. In our model, we assumed that all the dipoles were oriented in the plane. This approximation is reasonable since the anisotropic refractive indices indicate that the conjugated polymer chains lie in the plane.[18,19,25–27] In contrast, the dipoles in a small-molecule LED are randomly distributed. As a result, the coupling into the four mode types is very different for a small-molecule LED. In general, the vertically oriented dipoles in a small-molecule emitter cause even more waveguiding and surface-plasmon coupling.[28]

1. Effect of emitting dipole location on emitted power distribution

In Fig. 3(a), we plot the fraction of the power emitted into each type of mode as a function of the dipole position within the emitting layer. This plot shows the importance of the emitting dipole's location on the efficiency of the device.

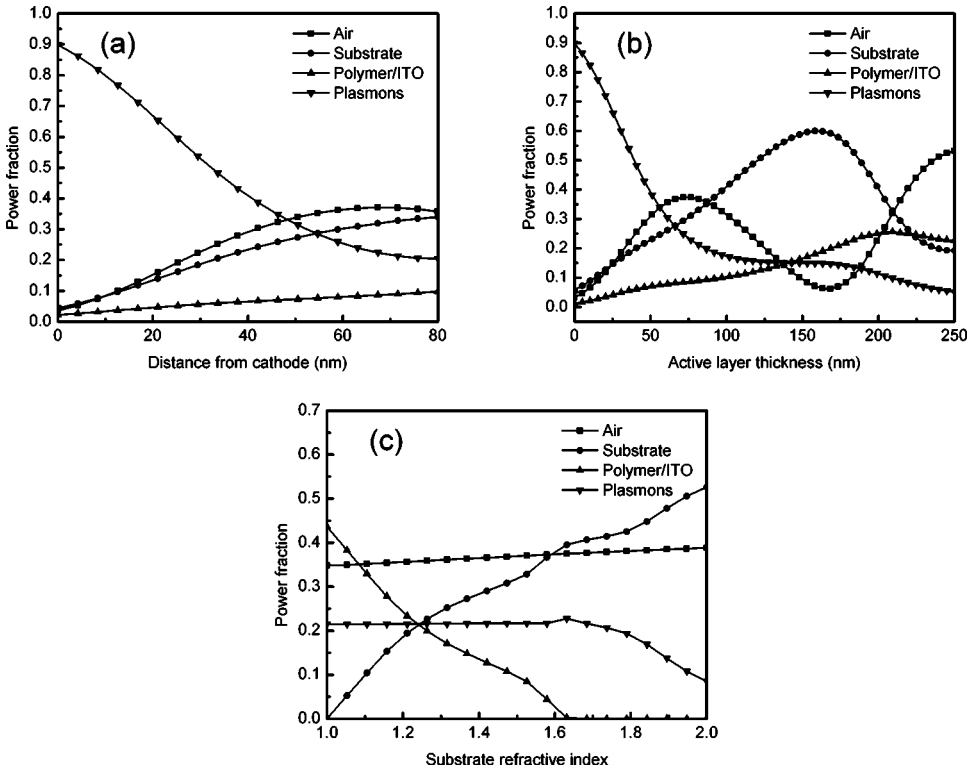

FIG. 3. (a) The fraction of the emitted power coupled into the four optical mode types as a function of the emitting dipole location within the active polymer film. (b) The fraction of the emitted power coupled into the four optical mode types as the active layer thickness is varied. (c) The fraction of the emitted power coupled into the four optical mode types as the substrate index is varied. The dipoles are assumed to be very close to the anode and the power has been integrated over all emission wavelengths. The intrinsic emission spectrum of the LUMATION Green 1304 LEP is used to weigh the results over all emission wavelengths.

If the dipoles are very close to the cathode, the emission is strongly quenched by the metal layers and most of the power is coupled into surface-plasmon modes. In contrast, if the dipoles are very close to the anode, the coupling into surface-plasmon modes is reduced and the coupling into air modes is maximized. For this particular polymer LED, the electron mobility is higher than the hole mobility and the electron injection is more efficient.[29] Therefore, the recombination zone is located close to the anode.

2. Effect of active layer thickness on emitted power distribution

In Fig. 3(b) we plot the coupling into each type of mode as the thickness of the LUMATION Green 1304 LEP layer is varied. The results show that the light extraction efficiency of the LEDs should be highest when the active layer is either 80 or 250 nm thick. Since the operating voltage scales with thickness, the power efficiency should peak at 80 nm. We have varied the thickness and confirmed that this is indeed true. An oscillation of LED efficiency with film thickness has been observed previously in another polymer LED structure.[30] The oscillation of power going into the air modes with film thickness is due to the dipoles moving in and out of phase with their image dipole in the metallic cathode. As the thicknesses increases above 80 nm, the coupling into air modes decreases while the coupling into polymer/ITO waveguide modes increases. Consequently, extracting light from these modes is very important for thick devices. This result explains why 100% increases in external efficiency have been achieved using gratings in thick LEDs.[11,12]

3. Effect of the substrate index on emitted power distribution

In Fig. 3(c) we illustrate that changing the index of a layer can change how the power is coupled from the dipole. In this case, we vary the refractive index of the substrate to illustrate how the trapped light may actually be concentrated into a specific type of mode. For example, polymer/ITO modes can be suppressed by increasing the refractive index of the substrate above the index of the ITO and polymer layers. This places most of the trapped power in the substrate modes. The concentration of power in the substrate modes could be combined with substrate- shaping[6,9] or scattering techniques[7,8,10] to significantly increase the external efficiencies of the devices.

4. The emitted power distribution of an actual device

In this study we are specifically interested in extracting light from the device structure introduced in Fig. 1 where the active layer is 80 nm thick and the substrate is soda-lime glass ($n=1.52$). To calculate these results, we assumed an exponential distribution of emitting dipoles that peaks at the polymer/PEDOT interface with a decay length of 10 nm and integrated over the entire emission spectrum of the polymer. The calculated fraction of the dipole power emitted into each

TABLE I. The power fraction (PF) that goes into each mode as well as the possible increase in efficiency factor (1) from the efficient extraction of light in air, polymer/ITO (Poly), substrate (Sub), and plasmon (SP) modes.

	Air	Poly	Sub	SP	Tot
PF	0.36	0.09	0.32	0.23	1.00
1	1.00	1.25	1.89	1.64	2.78

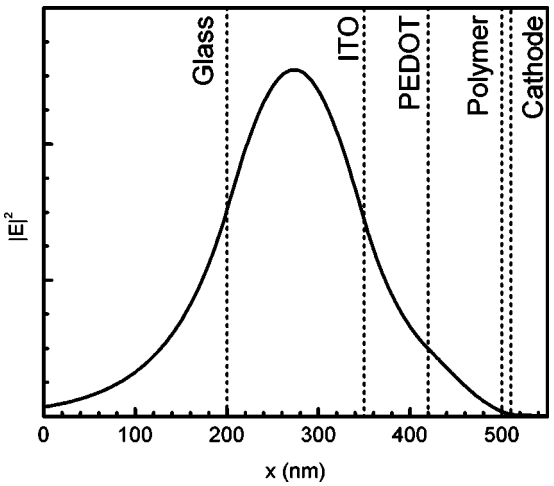

FIG. 4. The distribution of the in-plane electric-field intensity as a function of location within a LUMATION Green 1304 LED at $\lambda = 550$ nm. This particular mode is a TE_0 mode, which means the electric field is entirely in the plane of the LED.

mode type is summarized in Table I. As expected the emission into air modes accounts for less than half of the total amount of power radiated by the emitting dipoles. However, the distribution of power between the trapped modes is somewhat surprising. The substrate modes contain the largest fraction of the trapped light, and only a very small fraction is trapped in the polymer/ITO modes.

The fraction of power coupled into plasmon modes is also larger than expected. Plasmon losses are generally considered to be important only for small-molecule LEDs due to dipole orientation issues.[28] However, these results suggest that plasmon modes can also be significant sources of loss for polymer LEDs as well. In our calculations, we have combined surface-plasmon polariton (SPP) modes[22] with other lossy surface modes.[23] The SPP modes are guided optical modes, and some of the power lost to these modes may be recovered with an appropriate extraction technique.[31–34]

5. Controlling spontaneous emission

The large changes in the coupling into the four mode-types with changes in the active layer thickness and substrate refractive index highlight how sensitive the emission properties are to the device structure. In fact, modification of the emission pattern by addition of a single dielectric layer[4,5] or addition of many layers to form a microcavity structure[2,3] is one option for increasing efficiency. In this paper, we instead focus on how light can be extracted from the trapped modes using various extraction techniques that have the potential to be implemented at low cost.

B. Extraction of light from polymer/ITO waveguide modes

Light trapped in the polymer/ITO waveguide modes will be concentrated in three layers with a total thickness of less than 400 nm. As a result, the light is well confined with a finite number of modes. In fact, for our specific LED structure, there is exactly one mode for each polarization and wavelength. One of the most common methods for coupling light out of confined, finite modes is to use a Bragg grating to scatter the light out into the air modes. The Bragg equation is given by

$$k_0 \sin\theta = \beta - \frac{2\pi}{\Lambda}, \tag{1}$$

where k_0 is the free-space wave vector $2\pi/\lambda$, θ is the angle at which the waveguide mode is outcoupled into the air, β is the in-plane wave vector of the waveguided mode, and Λ is the period of the grating. Since β/k_0 is typically between 1.5 and 2 for these structures, the grating periods necessary for efficient scattering must be slightly less than λ. From the Bragg equation, we see that the outcoupling of the waveguide modes of a specific wavelength occurs at well-defined angles. As a result, it should be possible to use multiple Bragg gratings to change the shape of the emission spectrum as a function of angle.

The fundamental physics of outcoupling by Bragg gratings was demonstrated for polymer LEDs a few years ago.[11,12] The initial results were extremely promising, with a factor of 2 improvement in efficiency over a reference LED with no grating. However, the device structures were somewhat unconventional. In addition to a thick active layer, the external emission was collected after traveling through gold and photoresist layers. As a result, the absolute efficiencies of the devices were extremely low.

Recently, we have reported on our attempts to incorporate Bragg gratings into LEDs with a more traditional

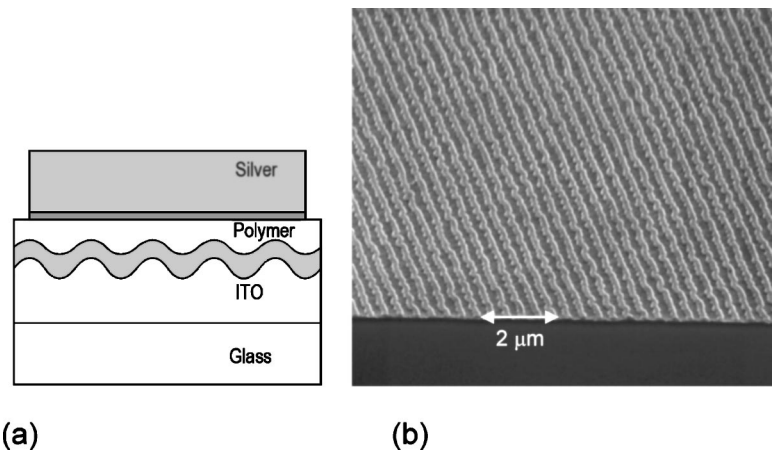

FIG. 5. (a) The LED structure when the Bragg grating is incorporated into the ITO layer. (b) A SEM image of a Bragg grating etched into the ITO layer.

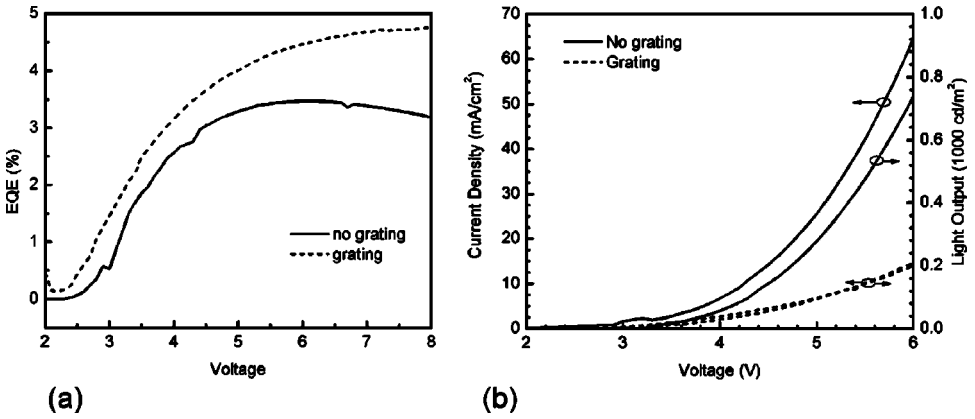

FIG. 6. (a) The external quantum efficiency as a function of the applied voltage for a LED with and without a Bragg grating. The grating period was 380 nm and the grating depth was 65 nm. (b) Current density and luminance as a function of applied voltage for LEDs with and without a Bragg grating.

device structure.[13] In our work, we incorporated gratings in the PEDOT layer using soft lithography. This technique was attractive because of the potential for low-cost fabrication. We reported a dependence of the grating effect on the thickness of the light-emitting polymer poly[2-methoxy-5-(3′,7′-dimethyloctyloxy)-*p*-phenylene-vinylene] (OC_1C_{10}–PPV). For an optimum OC_1C_{10}–PPV thickness of 100 nm, the grating did not improve the external efficiency. However, as we increased the thickness of the active layer, which causes more light to be emitted into the polymer/ITO modes, we were able to see an increase in the efficiencies due to the grating. We concluded that waveguide absorption was limiting the effect of our gratings and showed that using a thin layer of calcium capped with silver was better than just using calcium in the cathode since silver does not absorb light as well.

In our LUMATION Green 1304 LEP devices, waveguide absorption is also extremely strong. The origin of the strong waveguide absorption is the concentration of the waveguided light in highly absorbing ITO and PEDOT layers. For Fig. 4, we have used a transfer-matrix technique[35] to solve Maxwell's equations and plotted the electric-field intensity of a polymer/ITO waveguide mode as a function of position within the device. This plot clearly shows that most of the power in the waveguide mode is concentrated in the PEDOT and ITO layers. While these layers are transparent for thicknesses of a few hundred nanometers, the waveguided light travels *along* the layers and is completely absorbed in less than 10 μm. The waveguide absorption must be overcome by increasing the coupling strength of the grating.

The coupling strength of a Bragg grating depends on the strength of the fields in the grating region.[36] From Fig. 4, we can see that the field intensities at the PEDOT/polymer interface are relatively low. Combined with the strong waveguide absorption, it is not surprising that the PEDOT gratings were not completely effective. By contrast, the fields are much higher at the ITO/PEDOT interface. Therefore, the coupling strength of the grating should increase substantially by simply putting the grating in the ITO.

An improved Bragg grating structure is shown schematically in Fig. 5(a) along with a SEM image of a grating in the ITO layer in Fig. 5(b). We plot the external quantum efficiency as a function of voltage for devices with and without ITO gratings in Fig. 6(a). With gratings in the ITO, we were able to increase the peak external efficiency of our LEDs from 3.5% to 4.7%. This corresponds to a 34% increase in the quantum efficiency; greater than the amount that we calculated was in the polymer/ITO modes in Sec. III A. Since these were one-dimensional gratings, we are confident that an even larger increase could be achieved with a two-dimensional grating. In addition to the polymer/ITO modes, the grating may be scattering out some of the light waveguided in the substrate. The higher than expected increase may also be due to the changes in the device structure. For example, the grating has introduced significant variations in the thickness of the ITO, PEDOT, and polymer layers.

The effect of the thickness variations on the *IV* curves is shown in Fig. 6(b). The devices with the gratings have much lower current densities, and the operating voltage necessary to achieve 1000 cd/m^2 increases. Therefore, while the quantum efficiencies have increased by 34%, the power efficiencies at 1000 cd/m^2 increased by just 25%, from 6.9 to 8.6 lm/W. In addition to the thickness variations, the physical etching may have changed the optical or electronic properties of the ITO. These effects are undesirable, but we can propose two possible solutions. The first would be to move the grating to the ITO/glass interface. The field intensities plotted in Fig. 4 are equally high at the glass/ITO interface and the grating effect should be strong. This approach has been successfully demonstrated for small-molecule LEDs,[14,15] and it has the advantage of separating the optical extraction structure from the electronic device structure. An-

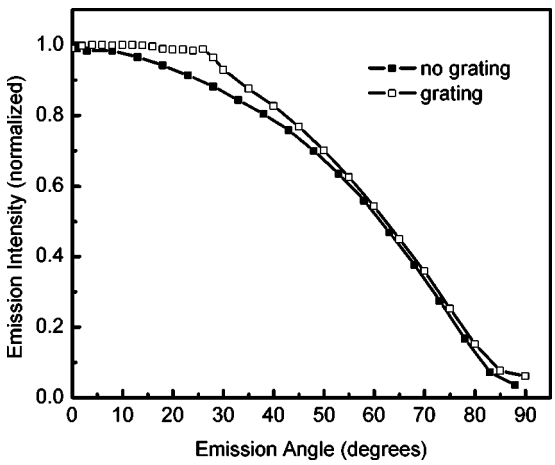

FIG. 7. The emission intensity as a function of emission angle for a LED with and without a grating.

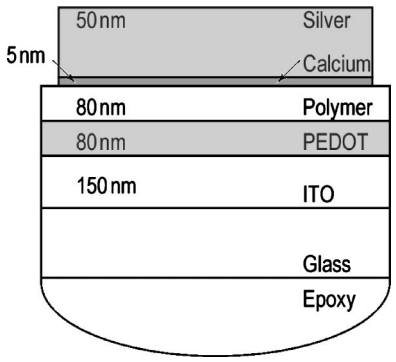

FIG. 8. The LED structure when a UV-cured epoxy lens is formed on the glass substrate. The index of the epoxy was 1.52, which is matched to the index of the soda-lime glass.

other alternative would be to planarize the anode by spin-coating a thicker PEDOT layer, which would eliminate any variation in the thickness of the LUMATION Green 1304 LEP layer.

The angular emission patterns of the devices are shown in Fig. 7. It is interesting to note that the grating does not substantially change the emission pattern. To see if the color varied with emission angle, we calculated the Commision Internationale de I'Eclairage (CIE) coordinates and found that the color did not vary significantly.

In summary, we have improved the extraction of light waveguided in the polymer/ITO layers by changing the location of the grating within the device structure. While the relative increase we have observed is much smaller than some of the increases that have been previously reported, our results are based on optimized thicknesses and we have achieved one of the highest efficiencies to date with a Bragg grating in a polymer LED. In addition, our modeling indicates that we have extracted the majority of the light waveguided in these layers with our grating.

C. Extracting light from the substrate waveguide modes

Since the modeling suggests that an even larger fraction of the trapped light is waveguided in the substrate, we have also investigated the extraction of waveguided light from substrate modes. The glass substrate is typically much thicker than the other device layers (0.7–1 mm). As a result the waveguide modes are not well confined and there are a much larger number of modes at a given wavelength. Many techniques have been proposed to extract light from the substrate. These can be categorized into scattering[7,8,10] and substrate-shaping techniques.[6,9] By modifying the planar nature of the air/glass interface with small epoxy lenses we strove to reduce the total internal reflection at the air/glass interface and increase the coupling of light into the air. The structure we fabricated is shown in Fig. 8. The lenses were not designed to focus or collimate the light, and from simple ray-tracing calculations we estimated that about 50% of the light trapped in the substrate would be outcoupled by the lens. We were able to test the same device with and without the lenses. Typical results are shown in Fig. 9(a). The peak external quantum efficiency increased by 43% from 3.5% to 5.0% and the power efficiency at 1000 cd/m^2 increased from 6 lm/W to 10.1 lm/W, corresponding to an over 60% increase. The expected outcoupling fraction and the measured increase match well with the amount predicted by the model. Figure 9(b) shows that the lens significantly changes the emission pattern. The effect of the lens depends on the relationship between the lens height and the radius of curvature. By patterning the lenses using soft lithography it should be possible to design a lens that will be even more effective at extracting light from the substrate. However, the increase we have observed is encouraging and lends more support to our modeling results.

IV. CONCLUSION

The results of our work on light extraction are summarized in Table II. By modeling the LED structure, we were able to predict the fraction of the emitted power coupled into each type of mode. For this particular device structure, the modeling predicts a 25% increase in external efficiency by extracting light from waveguide modes in the polymer/ITO layers. We have demonstrated a 34% increase by using Bragg gratings in ITO to scatter the waveguided light out of the device. The larger increase can be attributed to the variations in the active layer thickness and the possibility that some substrate modes are also scattered by the grating. In order to effectively extract light from these modes, the grating must be properly located to interact with the high fields of the waveguide modes. We have also investigated light extraction from waveguided modes in the substrate. Our nu-

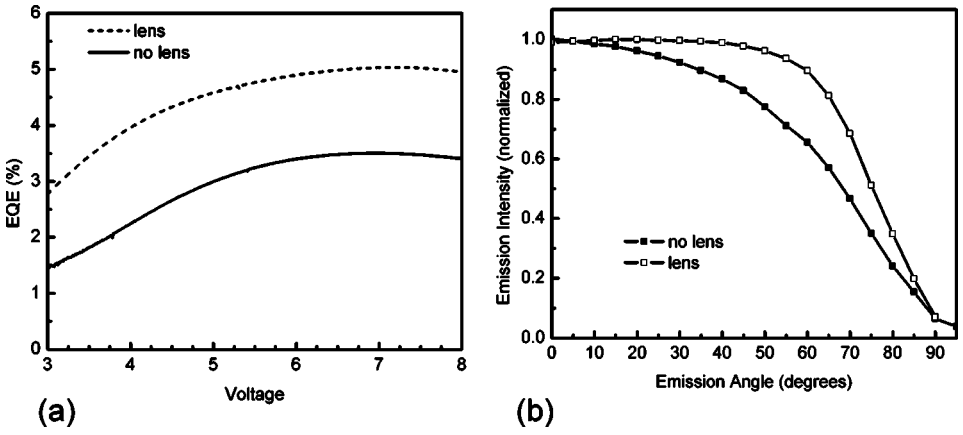

FIG. 9. (a) The external quantum efficiency as a function of the applied voltage for a LED with and without the epoxy lenses. (b) The emission intensity as a function of emission angle for a LED with and without lenses.

TABLE II. Our results for polymer/ITO (Poly), substrate (Sub), and plasmon (SP) waveguide modes. The peak external quantum efficiencies of reference devices (QE_{ref}) are compared to the extracted peak efficiencies (QE_{ext}). Also the power efficiencies of reference LEDs (PE_{ref}) are compared to the extracted efficiencies (PE_{ext}) at 1000 cd/m^2. The experimental increase in quantum efficiency due to the extraction (ΔQE) can also be compared to the maximum increase predicted by the numerical model (Δ_{max}). The experimental increase in the power efficiencies (ΔPE) has also been listed.

	QE_{ref} (%)	QE_{ext} (%)	PE_{ref} (lm/W)	PE_{ext} (lm/W)	ΔQE	ΔPE	Δ_{max}
Poly	3.5	4.7	7.0	8.6	0.34	0.25	0.25
Sub	3.5	5.0	6.0	10.1	0.43	0.60	0.89
SP	···	···	···	···	···	···	0.64[a]

[a]Note that the maximum increase by SP extraction cannot be achieved due to absorption by lossy surface waves.

merical model predicts an increase of up to 89% by extracting light from these modes. We have demonstrated a 43% increase in external efficiency and an over 60% increase in the power efficiency by using lenses to change the shape of the substrate. This technique is particularly attractive because the lenses could be formed at low cost using soft lithography or ink-jet printing. Our modeling also suggests that a significant fraction of the light is trapped in surface-plasmon modes. While much of the power is lost into these surface modes, only light in the surface-plasmon polariton modes is actually extractable. However, effective extraction of light from these modes is certainly an important issue. Surface-plasmon polaritons are extremely well confined to the polymer/cathode interface, and periodic structures at this interface could be used to scatter these modes out of the device.[31,32,34] Using soft lithography to pattern the polymer layer before thermal evaporation of the cathode is one possibility for inexpensive extraction of these modes. However, a grating would increase the interfacial area between the polymer and the cathode and may significantly increase metal quenching and *reduce* rather than increase the external efficiency of these devices.

Careful modeling of a LED structure combined with well-designed extraction techniques can significantly enhance the external efficiencies of organic LEDs. In this report, we have studied a polymer LED structure, but the same techniques could be applied to small-molecule LEDs. We have demonstrated methods for effectively extracting light from two of the three general types of waveguide modes. By using soft lithography or ink-jet printing, the light extraction could be included without significantly increasing the fabrication costs.

ACKNOWLEDGMENTS

We would like to thank the Dow Chemical Company for providing the light-emitting polymer. Financial support for this work was provided by the National Science Foundation (Grant No. ECS-0093502).

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