2.3: Monte Carlo Simulations and Opto-Electronic Properties of Polymer Light-Emitting Devices on Flexible Plastic Substrates

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Abstract

A Monte Carlo method for modeling the light transport phenomena in organic polymer light-emitting devices (PLEDs) has been reported by our group. [1] The unique advantage of this method is its ability to model bulk absorption events, thin film coatings, and rough surfaces while keeping track of the photon polarization state in a realistic geometry of the devices. We have applied this method to analyze the wavelength output distribution and out-coupling efficiency of the PLEDs. In this paper we further expand this method to model the Fresnel interactions at rough surfaces. We have found that the interfacial roughness between the polymer light emissive layer and hole transport layer increases the probability of the photon out-coupling and wave-guiding parts of the internally generated light.

1. Introduction

The external quantum efficiency of the organic polymer light emitting devices (PLEDs) are limited by four major losses: the charge injection at the contacts and charge transport within bulk, electron and hole radiative recombination, photo-luminescent efficiency, and light out-coupling efficiency [2]. It is estimated using a standard refraction theory that the photon out-coupling efficiency is about one-fifth of the total internally generated photons [3]. Therefore, several methods for improving the light out-coupling efficiency have been employed to overcome this limitation set by the escape cone of the substrate. These methods include introducing rough or textured surfaces or interfaces [4], use of ordered microlens arrays or microsphere medium [5], use of reflecting surfaces or distributed Bragg reflectors [6], [7], and use of a thin silica aerogel layer [8]. Several models have also been proposed for modeling organic light-emitting devices, such as half-space optical model [9], one-dimensional ray-tracing model [5], and quantum mechanical micro cavity model [10], [11].

The Monte Carlo approach was developed in our laboratory to analyze the PLED light out-coupling efficiency [1]. This statistical method has the flexibility for modeling events such as absorption, wave-guiding, scattering, out-coupling, and trapping happening in the devices. The unique advantage of the Monte Carlo simulation method is that it takes into account the realistic geometry of the devices. Also, a Monte Carlo simulation includes the angular and spectral distributions of the emitted photons, the point-spread function, the specular and diffuse reflection coefficients, and a summary of scattering events statistics. In summary, the unique advantage of this method is its ability to model bulk absorption events, thin film coatings, and rough surfaces while keeping track of the photon polarization state in a realistic geometry of the devices. In this paper, we applied a Monte Carlo simulation to investigate the influence of different light emitting material and different degree of interfacial roughness on the PLED light out-coupling efficiency.

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2. Methods

2.1 Optical modeling

In this work, we used a Monte Carlo method for the simulation of light transport processes in PLEDs [1], [12], [13]. The Monte Carlo method makes use of the generation of photons with random direction according to a distribution function describing the nature of the light emission. In this analysis, the light source within the organic polymer layer is considered isotropic from a single point situated in the center of the device, Figure 1. We assumed the light source being described by the photo-luminescence spectra of the light-emiting materials [14]. The photon histories are then followed through a sequence of interactions that includes absorption and Fresnel refraction. At the optical boundaries, an analysis is performed depending on the surface type and material properties using Fresnel's equations and considering the polarization of the incoming photon [1]. When the film thickness is comparable to the photon wavelength, we use modified Fresnel coefficients to describe the interference effects of optically thin-films. The reflection and transmission coefficients are then interpreted as probabilities. The simulation outcome is calculated by a statistical average of the fate of all histories according to the desired quantity to be evaluated for each experiment.

In our simulation, we introduce out-coupling efficiency (η_{coup}) to represent the probability that a photon generated at the luminescent center within the PLED, emerges through the front surface of the device, thereby contributing to electroluminance. The η_{coup} depends strongly on the device structure and on the optical constants of the materials and surface properties, and is always less than unity due to light absorption, edge emission, and light trapping. We can summarize the relevant physical processes that occur as

$$\eta_{coup} = 1 - \eta_{wav} - \eta_{abs} - \eta_t$$

where η_{wav} is the fraction of photons that are wave-guided within the device structure and exit through the device edge (contribution

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from all layers is included in this term), η_{abs} is the bulk absorbed fractions within device absorptive layers, and η_t is the trapped fraction in the ITO optical thin-film. In this simulation, we have neglected the light leakage through the cathode side, and the photo-luminescence (PL) quenching due to polymer composition variations and the presence of carrier flow within the PLEDs. Electric-field-induced photoluminescence quenching in conjugated polymers has experimentally been confirmed in this study, but cannot be implemented easily in this calculation.

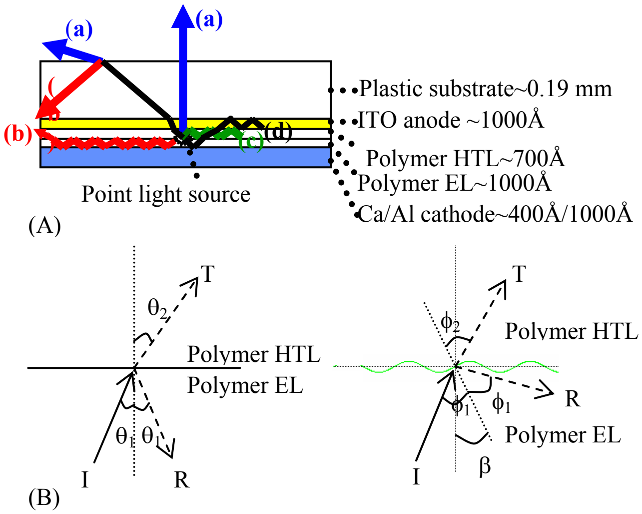


Figure 1. (A) Schematic representation of the PLED structure used in this work. A point light source was specified at the interface between the polymer hole transport layer (HTL) and polymer emissive layer (EL). The generated light has four fates: (a) emitted through the front surface, (b) waveguided within the device and exit through device edges, (c) absorbed in the absorptive materials, and (d) trapped in the ITO thin film, (B) Two-dimensional representation of the rough surface model. The rough surface is specified at the interface between polymer EL and polymer HTL. θ_1 and θ_2 are given by Snell's law. On the left, the flat surface case is depicted. The surface normal is rotated within an angle β on the right. After the boundary analysis is performed in the rotated system, T or R are expressed in the original coordinate system.

To describe Fresnel interactions at uneven surfaces of the type found in light emissive displays, an algorithm that randomly perturbs the surface normal was developed. This description is appropriate for surfaces having undulating profiles with low aspect ratio. The departure from a smooth surface is specified by defining a maximum cone within which surface normal tilting can occur. The tilting angle β, Figure 1(B), is sampled uniformly from a cosine distribution within the allowed range β_M. The tilting angle used in this simulation represents a surface slope that can be related to surface roughness, although a direct relationship between this description and the roughness is not possible. In the method used, a Fresnel analysis is then performed using angles ϕ_1 and ϕ_2. The Fresnel equations are solved with the incident photon vector expressed in a rotated coordinate system. The out-coming vector is then rotated back to the initial coordinate system. Additionally, a check-point is performed: $\phi_1 + \beta < \frac{\pi}{2}$, which prevents a reflection to be directed back into the surface. For a reflection occurring in a valley of the surface, the reflected photon may strike a neighboring hill. This is not accounted in the model, and therefore, surface normal distributions should be limited to modest cone angles ($\beta_M < 30°$). This algorithm is a first approximation to the problem of uneven surfaces found in actual devices, and it is used only to investigate general trends in the simulation data.

2.2 Experiment

The organic polymer light-emitting device (PLED) structure used in this work is published in [14]. The PLEDs front out-coupling and edge wave-guiding light emissions were measured using an integrating sphere that was calibrated to measure the device total optical flux [13]. In this specific experiment, the PLEDs were placed inside the integrating sphere. When the PLEDs front light-emission measurements were done, we masked all sides of the PLEDs (anode contact, cathode contact, and two remaining edges) with a black mask, and collected only the PLEDs luminous flux from the front at different current density levels, Figure 2(A). When we measured the PLEDs front + two-edge light-emission, we only masked the anode and cathode contacts with a black mask, and then collected the light emitted from the PLED front and two other edges at different current density levels, Figure 2(B). From these results, we were able to calculate the PLEDs 4-edge emission values and PLED front to 4-edge emission ratios.

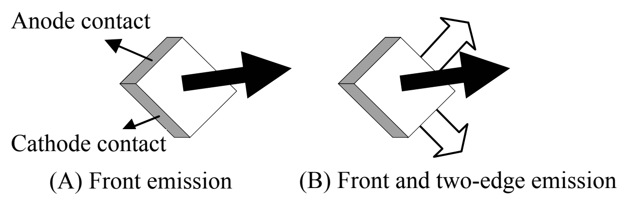
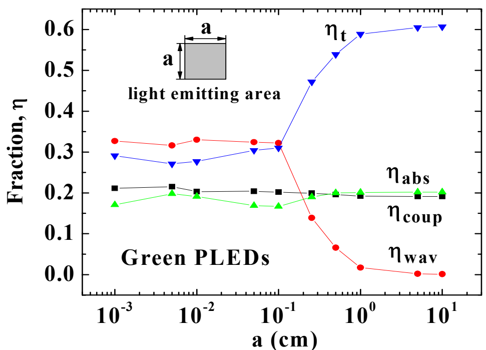


Figure. 2. Schematic representation of the PLED cross-sections used for the measurement of the device front and edge light emissions (A) front light emission and (B) front and two-edge light emission

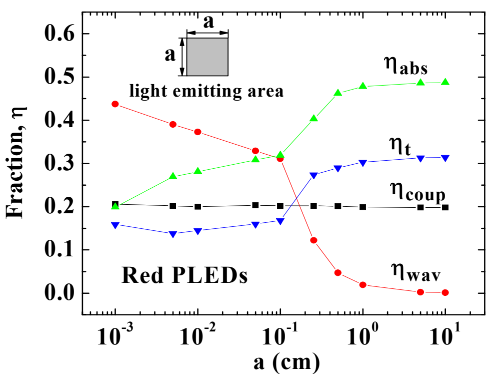
3. Results

The refractive indices and absorption coefficients with photon wavelength of the red and green polymers used in the simulation and experimental measurement were published in [14]. The refractive indices of hole transport layer and ITO thin film were obtained from literature [15]. The refractive index of the 0.19 mm thick transparent plastic substrate was 1.5. In this analysis, we used the photo-luminescent spectra of the green and red polymers as the input light source spectra [14]. The input photons go through a series of events, such as refraction, reflection, and absorption, and finally exit out or are killed in the device.

In Figure 3, we present results showing the change in the values of photon out-coupling η_{coup}, wave-guiding η_{wav}, absorption η_{abs}, and trapping η_t parts with the device sizes ranging from $10^{-3}\times10^{-3}$ cm^2 to $10^1\times10^1$ cm^2. As can be seen in Figure 3, the calculated photon out-coupling η_{coup}, wave-guiding η_{wav}, absorption η_{abs}, and trapping η_t parts are 0.20, 0.14, 0.19, and



(A)



(B)

Figure 3. Photon out-coupling (■), wave-guiding (●), absorption (▲), and trapping (▼) fractions of the green (A) and red (B) PLEDs with the different device sizes.

0.47 for 0.254×0.254 cm^2 green PLED and 0.20, 0.09, 0.43, and 0.29 for 0.254×0.254 cm^2 red PLED, respectively. The photon out-coupling efficiency is not affected by the wavelength distribution of the photon source for the green and red polymers used in this simulation. However, the absorption part in polymer layers (η_{abs} =0.43) of red PLED is larger than that (η_{abs} =0.19) of green PLED, and trapping / wave-guiding parts in ITO layer (η_t / η_{wav} = 0.29 / 0.09) of red PLED are smaller than those (η_t / η_{wav} = 0.47 / 0.14) of green PLED. As also can be seen in Figure 3, the calculated out-coupling efficiencies for both green and red PLEDs are approximately the same and do not change significantly when PLED sizes range from $10^{-3}\times10^{-3}$ cm^2 to $10^{1}\times10^{1}$ cm^2, while the wave-guiding part decreases significantly from ~30 to ~0% when PLED sizes increase from $10^{-3}\times10^{-3}$ cm^2 to $10^{1}\times10^{1}$ cm^2. The decrease in wave-guiding is due to the increase of absorption in the polymer layers and trapping part in the ITO thin-film when PLED size increases. Since the refractive indices at 633 nm for the ITO, plastic substrate, and air are ~1.8, ~1.5, and ~1, respectively, the photons reflected from the substrate-air interfaces will enter the ITO layer freely. Therefore, the increasing travel path of photons will (1) decrease probability of wave-guiding in plastic substrate, (2) increase probability of absorption in the polymer layer, and (3) increase probability of trapping in the ITO layer. The quantitative enhancement in the probability of absorption and trapping will depend on the material properties, such as refractive index and absorption coefficients, of each layer.

We further compared the experimental front out-coupling part over edge wave-guiding part of the light emission with the theoretical value as tabulated in Table 1 for 0.254 × 0.254 cm^2 and 0.508 × 0.508 cm^2 green PLEDs and 0.254 × 0.254 cm^2 red PLEDs with the device configuration shown in Figure 1. As can be seen in Table 1, a discrepancy between the simulated result without considering the effect of substrate absorption and experimental result exists. Please note that the effect of absorption or trapping of the plastic substrates with a multilayer structure cannot be easily quantified by experiments [16]. Therefore, we further simulate cases by assuming that the plastic substrate has an absorption coefficient of 7 cm^{-1}. In these cases we find excellent agreement between the experimental and theoretical results. As can be seen in Table 1, the front to edge emission ratio increases with the increasing device size experimentally and theoretically. Also, the experimental out-coupling part over the wave-guiding part is ~3.6 ± 0.6 and ~3.8 ± 0.7 for green and red PLEDs, respectively, which agrees qualitatively with the theoretical trend predicted by the Monte Carlo method.

Table 1: The experimental front out-coupling / edge wave-guiding emission ratio, and theoretical front out-coupling /edge wave-guiding emission ratio for 0.254 × 0.254 cm^2 and 0.508 × 0.508 cm^2 green PLEDs and 0.254 × 0.254 cm^2 red PLEDs

Sample	Theoretical front / edge emission ratio without substrate absorption	Experimental front / edge emission ratio	Theoretical front / edge emission ratio with substrate absorption, α=7cm^{-1}
Green B			
0.254 × 0.254 cm^2	1.43	3.6 ± 0.6	3.2
0.508 × 0.508 cm^2	3.06	15.0 ± 2.9	16
Red B			
0.254 × 0.254 cm^2	2.22	3.8 ± 0.7	3.5

Figure 4 (A) and Figure 4 (B) show the effect of interfacial roughness between the polymer emissive layer and hole transport layer on the photon out-coupling, wave-guiding, absorption, and trapping parts of the 0.254 × 0.254 cm^2 green and red PLEDs, respectively. As can be seen in Figure 4, the sum of the out-coupling and wave-guiding parts of the internally generated light of green and red PLEDs gradually increases from 0.34 to 0.57 and from 0.29 to 0.53 when the maximum surface tilting angle β_M increases from 0° to 30°. The interfacial roughness between the polymer emissive layer and hole transport layer increases the probability of the light transmission part at the polymer emissive layer and hole transport layer at the expense of the light reflection part. These results suggest that randomly roughening the interface light emission region of PLEDs will enhance both the light out-coupling efficiency and wave-guiding efficiency.

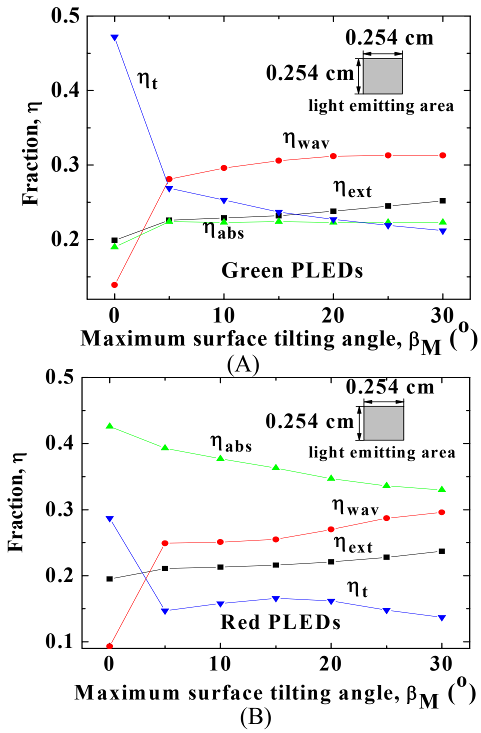


Figure 4. Photon out-coupling (■), wave-guiding (●), absorption (▲), and trapping (▼) fractions of the green (A) and red (B) PLEDs with different degree of maximum surface tilting angle β_M between the polymer emissive layer and hole transport layer

4. Conclusions

We have demonstrated that a light transport Monte Carlo code can be used for modeling the realistic geometry of the PLEDs. This method takes into account the absorption in the polymer layers, multiple refractions within the device structure, back-reflection from the cathode, and interference effect of the optically ITO thin-film. We showed that red (n_D~1.84 at 633 nm) and green (n_D ~1.7 at 633 nm) polymer PLEDs have similar simulated photon out-coupling efficiency, which is about 20%. We also showed that the interfacial roughness between the polymer light-emissive and hole transporting layers increases the probability of the photons out-coupling and wave-guiding fractions of the internally generated light.

5. Acknowledgements

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