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Device Reflectivity as a Simple Rule for Predicting the Suitability of Scattering Foils for Improved OLED Light Extraction

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ABSTRACT

A general challenge in Organic Light Emitting Diodes (OLEDs) is to extract the light efficiently from waveguided modes within the device structure. This can be accomplished by applying an additional scattering layer to the substrate which results in outcoupling increases between 0% to >100% in external quantum efficiency. In this work, we aim to address this large variation and show that the reflectivity of the OLED is a simple and useful predictor of the efficiency of substrate scattering techniques without the need for detailed modeling. We show that by optimizing the cathode and anode structure of glass based OLEDs by using silver and an ITO free high conductive Agfa Orgacon™ PEDOT:PSS we are able to increase the external quantum efficiency of OLEDs with the same outcoupling substrates from 2.4% to 5.6%, an increase of 130%. In addition, Holst Centre and partners are developing flexible substrates with integrated light extraction features and roll to roll compatible processing techniques to enable this next step in OLED development both for lighting and display applications. These devices show promise as they are shatterproof substrates and facilitate low cost manufacture.

Keywords: Light Extraction, Reflectivity, Flexible, Large Area, Solution Processed, OLED

1. INTRODUCTION

At the moment extensive experimental investigation and modeling is required to predict how to improve light extraction from a given device structure. In this work we aim to show that the effectiveness of adding a scattering layer to a glass substrate at improving light efficiency can be simply and accurately predicted using just a measure of the OLEDs reflectivity.

In a typical bottom emission organic light emitting diode light is emitted through a transparent substrate (typically glass) while the device is capped with a reflective cathode that serves as an electrode and to redirect emitted light in the desired direction. In these structures light is refracted as it passes from the higher index organic layers and transparent ITO anode into the lower index glass layers, as shown in Figure 1. For light emitted at higher angles within the emissive layer(s) this results in light that is totally internally reflected in these internally guided modes. The same effect can occur at the substrate air interface. These two sets of wave guided never escape in the forward direction unless some additional light extraction techniques are introduced and therefore represent losses in the system. Using typical refractive indices these light extraction losses are of the order of 80% of the initially generated light [1].

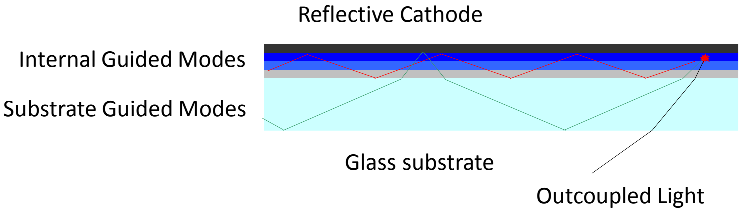

Figure 1. The substrate guided and internally guided modes of an OLED devices. In a typical glass based OLED these take up approximately 80% of the light resulting in significant performance losses.

With the use of phosphorescent emitters in optimized devices with optimized materials OLED devices can achieve 100% internal quantum efficiency [2-5]. Furthermore doped transport layers have reduced driving voltages to the point where

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device turn on occurs just above the bandgap of the materials [6]. This means that any further significant increase in OLED efficiency must come from improvements in extracting all the light from the organic layers and substrate where it can be waveguided and lost. Light extraction also increases the light output of devices of a fixed size driven with a certain amount of electrical power and so offers assistance with device lifetime and cost per lumen: the two other main challenges on the path to commercial OLED lighting.

While it is possible to extract light from the device layers [1, 7-9] it is easier to extract the substrate guided modes than the internally guided modes as there are high requirements on the electrical properties and smoothness of the OLED device layers themselves. By contrast it is comparatively easy to roughen the substrate or add a scattering film on top of it in order to extract the substrate modes [1, 10]. For full simulation of the consequences of changing device layers integrated models of charge transport, re-combination, light emission and light extraction need to be coupled together in order to relate the device structure to the location of the emission region in the device [11] and finally to the angle dependent emission [12] in order to validate the model. The typically used experimental approach to understanding the light emission within the substrate is to attach an index matched glass hemisphere to the substrate so the angle dependent properties of the modes in the glass can be analyzed and used to validate the models being used.

In this work we seek to find an industrially feasible approach to optimizing device design that will allow broadly applicable predictions to be made about the effectiveness of light outcoupling substrates on a variety of OLED stacks without the need for detailed modeling or analysis. This includes the difficult case of proprietary structures which have so far been optimized for charge transport and recombination and for which changing the OLED stack for light extraction purposes is not desirable. The final result of this effort will be a series of flexible substrate and device designs which can be produced on a large scale for good performance in combination with a variety of OLED structures before final optimisations are made.

Holst Centre has decided to focus on flexible substrates coated with our own moisture barriers because of their compatibility with low cost roll to roll processing [13]. However this choice also has significant potential for allowing improved light extraction by re-engineering the substrate itself to better couple light from the device layers and out couple substrate guided modes while retaining a thin flexible form factor.

It has been estimated that in 2005 19% of the world's electricity use was consumed by lighting applications and a significant fraction of that is in the form of low efficiency light sources [14]. Therefore there is significant scope to reduce both energy consumption and corresponding greenhouse gas emissions through low cost, efficient, lighting. Organic light emitting diodes (OLEDs) are a candidate for such a technology but they have significant other advantages such as their thin from factor and potential for flexible, conformable and light weight solutions for lighting which could prove especially valuable in vehicles and aircraft.

2. EXPERIMENTAL

OLED devices were fabricated on glass substrates by spin casting Livilux white light emitting polymer (LEP) provided by Merck and either Orgacon G6 from Agfa, Clevios Al.4083 or Clevios PH1000 poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate) (PEDOT:PSS). Light extraction foils (DTF #20100510) containing high index particles were provided by Dupont Teijin Films.

Current-voltage-light cycles were measured with a custom built characterization tool, where a voltage is supplied to the device by a Keithley 2440 source meter which also measures the current running through the device. The light was detected using a photodiode connected to a Keithley 2400 multimeter and was calibrated with a luminance meter LS110 from Konica Minolta. The reflectance of the OLEDs was measured with a Perkin Elmer Lambda-950 with integrating sphere containing openings that enabled a distinction between the specular and diffuse reflectance's.

A Veeco Daktak 6 stylus profiler with an accuracy of 5 nm was used to measure the thicknesses of the functional layers. The complex refractive indices of ITO, Orgacon PEDOT:PSS and Livilux light emitting polymer were determined by ellipsometry (Woollam variable-angle spectroscopic elipsometer). The intrinsic emission spectrum of this material was measured with a Perkin Elmer LS55 on a separate layer on quartz plates coated with carbon black to reduce interference effects.

Angle dependent measurements of the spectrum were performed using a Display Metrology System (DMS, Autronic Melchers GmbH) with an index match hemisphere in order to study the substrate guided modes in a manner similar to the publications of Gather et al. [15], Flämmich et al. [16] and Van Mensfoort et al. [17]. The angle dependent emission was modeled and analyzed using the modeling package Setfos 3.2 using the methods now published by B. Perucco [11] and Ruhstaller [18].

In order to investigate this effect systematically the reflectivity of a set of single layer white polymer OLEDs was varied by changing the cathode and anodes used as show in Figure 2. The device structures are listed in Table 1. Barium/aluminum and barium/silver cathodes were used for lower and higher reflective cathodes. Increasing barium thickness resulted in reduced reflectivity. ITO and Clevios Al.4083 PEDOT and high conductive Clevios PH 1000 or Orgacon G6 PEDOT with shunting lines were used to provide the anode [19]. The ITO/PEDOT anode produced lower reflectivity samples than the high conductive PEDOT. All experiments were performed on ordinary low cost soda lime glass substrates. Though coupling of the light into the substrate could have been significantly improved by using a high refractive index substrate [20], high refractive index glass is considered to be too expensive to be industrially applicable.

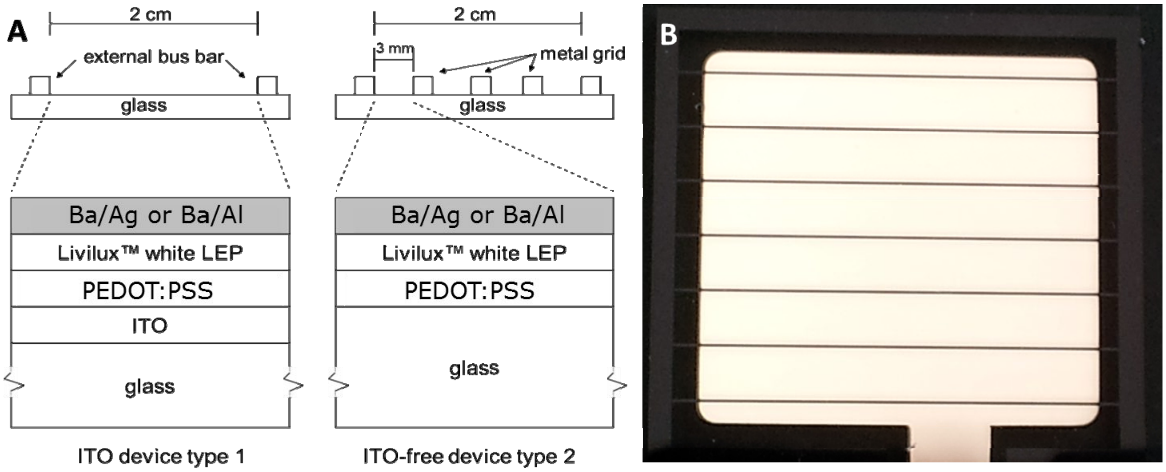

Figure 2.(A) OLED device structures with either silver or aluminum barium cathodes used in combination with either ITO or high conductive Orgacon or Clevios PH1000 PEDOT:PSS with metal lines (B).

3. RESULTS AND DISCUSSION

Table 1: OLED device structures. Layer thicknesses shown are as used in Setfos to reproduce the experimentally determined angle dependent emission. These matched closely with surface profilometry results. The measured spectrally averaged reflectivity $\mathbf{R}_{\mathbf{av}}$ is also shown.

Device #	**ITO**	**PEDOT:PSS**		**LEP**	**Cathode**	$\mathbf{R}_{\mathbf{av}}$
1	135 nm	Clevios Al 4083	90 nm	77 nm	Ba/Al/Ag	41%
2	140 nm	Clevios Al 4083	130 nm	76 nm	Ba/Al	51%
3	None	Clevios PH 1000	140 nm	83 nm	Ba/Al	55%
4	None	Orgacon G6	80 nm	92 nm	Ba/Al	65%
5	138 nm	Clevios Al 4083	90 nm	79 nm	Ba/Ag	77%
6	140 nm	Orgacon G6	104 nm	81 nm	Ba/Ag	82%
7	None	Orgacon G6	120 nm	90 nm	Ba/Ag	86%

The EQEs of these devices with and without light extraction foils are shown in Figure 3 along with the quantum efficiency available in the substrate guided plus the air modes, and thus available for scattering. At lower reflectivities adding a light extraction foil does not increase the amount of outcoupled light at all with the EQE remaining at 2.4% in the case of device 1 with a barium/aluminum cathode and Clevios Al. 4083/ITO anode. Only above 60% reflectivity is there an increase in light extraction on adding a light extraction foil. Below 60% reflectivity, raising reflectivity raises the EQE with or without foil. However, above 60% reflectivity the EQE plateaus at 3.8% for devices without outcoupling foil. In this regime however the EQE of devices with outcoupling foil improves with increasing reflectivity reaching a peak with device 7 at 5.6% EQE with a barium/silver cathode and high conductive Orgacon G6 anode.

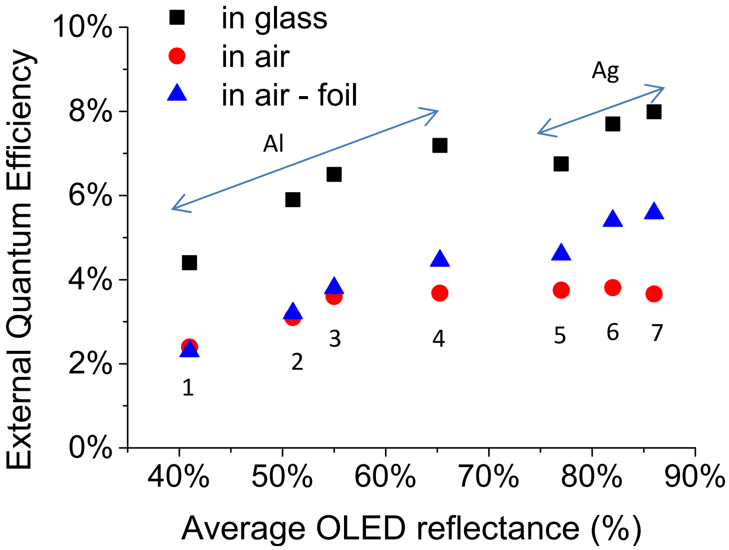

Figure 3. The external quantum efficiency of fluorescent white OLED devices with (blue triangles) and without (red circles) light extraction foil versus spectrally averaged reflectance. The quantum efficiency of light emitted into the substrate guided modes and in air modes, and thus available for light extraction, is shown by the black squares.

In order to understand the light extraction in these devices angle dependent measurements of the spectrum were performed and the different fractions of light coupled into the various modes of the device were calculated. To illustrate the effects of different anode choices with a constant silver cathode the angle dependent emission for silver cathode devices 5, 6 and 7 are shown in Figure 4 along with the calculated values. The fraction of light coupled into the different modes calculated based on the angle dependent measurements is shown in Figure 5.

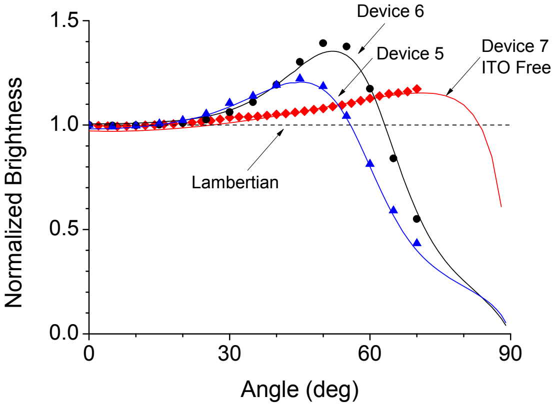

Figure 4. Normalized emission luminance into the substrate for silver cathode devices 5, 6 and 7 as a function of angle. The calculated curves are shown as solid lines. The experimental data, obtained with the Autronic Melchers DMS, are plotted as separate data points.

The angle dependent emission of ITO free device 7 in Figure 4 is clearly different from the ITO containing devices 5 and 6. This change in angular dependence results from the lower refractive index of the high conductivity PEDOT:PSS compared to the ITO which couples more of the light into the substrate guided modes where it can be outcoupled. By comparison the angular emission of devices 5 with Clevios Al. 4083 PEDOT:PSS on ITO and 6 with lower absorption Orgacon PEDOT:PSS are less different. Higher angle emission in device 5 is weaker because of stronger absorption in the Clevios Al. 4083 PEDOT:PSS layer.

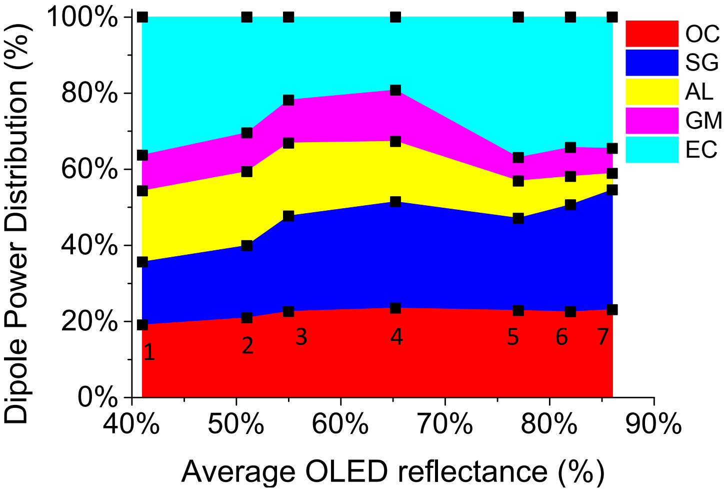

Figure 5. The calculated proportion of light for devices 1-7 directly outcoupled into air (OC), substrate wave guided (SG) and the loss modes associated to absorption (AL), evanescent coupling (EC) and internal wave guiding (GM).

The increase in higher angle emission into the substrate boosts the amount of light available for light extraction (as shown in Figure 5) and so we might expect a steady increase in the effectiveness of light extraction foils from device 5 to 6 to 7 corresponding to the greater availably of substrate guided light. However, despite the obvious changes in the light emission profile within the substrate between device 6 and 7 the amount of light that is emitted is outcoupled both with and without the light extraction foil in Figure 3 is similar. Instead the biggest difference is between devices 5 and 6 where the change to lower absorbing PEDOT has resulted in an increase in the reflectivity but a comparatively small change in the angular dependent emission.

Similarly from Figure 3 the amount of available substrate guided light and outcoupled light, thus the amount available for extraction, is actually greater in device 4 than in device 5. However device 5 shows better performance with light extraction foil. These results show that without detailed analysis the reflectivity of the device is actually a better predictor of the effectiveness of light extraction foils than working out the amount of extractable light in substrate guided modes.

So far we have used an approach to reporting the effectiveness of various light extraction strategies in OLEDs followed by many other workers and based on reporting the amount of light coupled into the air compared to a reference device. While this is a direct measure of the applicability of a particular light out coupling strategy to a particular OLED this approach is highly sensitive to the amount of light that is initially coupled into the air and in substrate guided modes. This approach can be misleading when comparing dissimilar devices. For example, a device with low external quantum efficiency (EQE) where, due to microcavity effects, the majority of light has been coupled into the substrate where it could be extracted by a scattering substrate, would show a very high enhancement. In contrast, a structure that had originally been optimized to out couple light directly into the air would show a much more modest improvement with the application of the same scattering layer.

The reason for the importance of OLED reflectivity in scattering light from the device can be understood intuitively by the scattering layer re-distributing light between the modes that would have been outcoupled and substrate guided in an unmodified glass device. Some of the light that would have been outcoupled directly is scattered into substrate guided modes as well as the foil scattering substrate, from which light is guided into the air. In many cases the scattered light must be reflected by the OLED, possibly multiple times, before it can be extracted. Therefore the reflectivity of the OLED is a highly important parameter.

For assessing the effectiveness of substrate scattering approaches, in the general case it is better to use integrated light outcoupling (ILO) which is a measure of the proportion of total light in the substrate guided and air modes in the reference device that are out coupled in the device with scattering substrate [21]. This means that the effectiveness of a scattering solution can be investigated in a way that is much less dependent on the particular emission profile of the device itself. As this approach neglects light in the organic modes, which cannot be extracted by substrate scattering, this means that it is not suitable for discussing modifications to the micro cavity in an OLED or scattering in the device layers themselves.

Empirically there is a general trend between the reflectivity of a complete OLED stack and the effectiveness of scatter foils and microlens arrays [21]. This holds for multiple device types with different structures and has been reported in the past as a general rule of thumb that the maximum ILO of a device is approximately equal to its average reflectance over the relevant spectral range. This trend is empirically illustrated in Figure 6 where the ILO values for numerous devices we have fabricated are plotted against their spectrally averaged reflectivity. In further work we will aim to better understand this relationship.

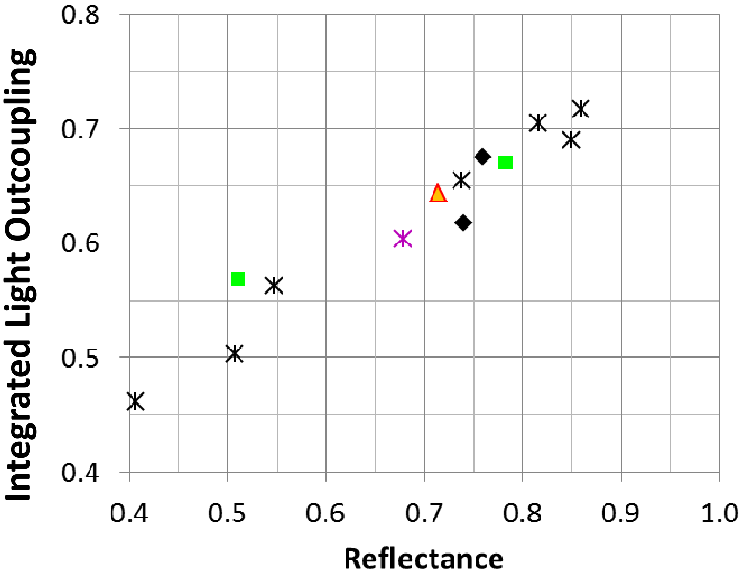

Figure 6. Integrated light outcoupling (ILO) as a function of device reflectance for a variety of emitters and device designs. The experiments described in this work are represented by black barred crosses.

These results show that introducing scattering to an OLED substrate does not automatically result in an increase in device efficiency even if the same scattering layer was effective when applied to other devices. However the success or failure of substrate scattering at improving light extraction can be estimated from simple measurements of the devices reflectivity with higher device reflection directly relating to higher light extraction enhancements. This also illustrates the importance of optimizing both anode and cathode reflectivity and minimizing absorption when designing OLEDs. As light out coupling will be a necessary feature of commercial OLED lighting products in order to improve lifetime and efficiency it is important to consider these factors even if their effects are not clearly visible in glass test devices without any light extraction features.

Although the discussion above has focused on glass based devices the relationship between the amount of substrate guided light that can potentially be outcoupled and the reflectivity of the cathode still holds for flexible devices based on plastics. These substrates are interesting for their low cost and flexibility but also because of the fact that they can have additional features incorporated into them to improve the OLED device in a way that is not cost effective on glass. For example metal grid lines (necessary for large area applications) can be embedded into them in a way that leaves a negligible surface topology. Similarly light extraction features can also be directly incorporated into the substrates.

Finally, although the OLED architectures used in this study do not meet the performance requirements for practical applications in the lighting industry, the approach used can also be applied to high efficiency OLED stacks. We have already demonstrated the effectiveness of these scattering solutions on flexible, phosphorescent small molecules white OLEDs made using the Solvay and Plextronics material sets, for which the use of a light extracting foil supplied by DuPont Teijin Films resulted in a power efficacy of 42 lm/W for a pixel area of 69 cm^2.

4. CONCLUSION

In this work we have shown that by optimizing only the anode and cathode structures it is possible to improve the EQE of an OLED by 130%. We have shown that the effectiveness of scatter foils can be quickly estimated by measuring the reflectivity of OLED devices. This allows the suitability of OLED designs for light extraction strategies and points to high reflectivity structures using silver and high conductivity PEDOT:PSS, rather than aluminum and ITO, offering better prospects for highly efficient phosphorescent OLED lighting devices.

5. ACKNOWLEDGEMENTS

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REFERENCES

[1] K. Saxena, V. K. Jain, and D. S. Mehta, “A review on the light extraction techniques in organic electroluminescent devices,” Optical Materials, 32(1), 221-233 (2009).

[2] C. Adachi, M. A. Baldo, M. E. Thompson *et al.*, “Nearly 100% internal phosphorescence efficiency in an organic light-emitting device,” Journal of Applied Physics, 90(10), 5048 (2001).

[3] Y. Kawamura, K. Goushi, J. Brooks *et al.*, “100% phosphorescence quantum efficiency of Ir(III) complexes in organic semiconductor films,” Applied Physics Letters, 86(7), 071104 (2005).

[4] S.-O. Jeon, K. S. Yook, C. W. Joo *et al.*, “100% internal quantum efficiency and stable efficiency roll-off in phosphorescent light-emitting diodes using a high triplet energy hole transport material,” Applied Physics Letters, 93(6), 063306 (2008).

[5] J. H. Seo, S. J. Lee, B. M. Seo *et al.*, “White organic light-emitting diodes showing nearly 100% internal quantum efficiency,” Organic Electronics, 11(11), 1759-1766 (2010).

[6] T. Matsushima, and C. Adachi, “Extremely low voltage organic light-emitting diodes with p-doped alpha-sexithiophene hole transport and n-doped phenyldipyrenylphosphine oxide electron transport layers,” Applied Physics Letters, 89(25), 253506 (2006).

[7] Y. R. Do, Y.-C. Kim, Y.-W. Song *et al.*, “Enhanced light extraction efficiency from organic light emitting diodes by insertion of a two-dimensional photonic crystal structure,” Journal of Applied Physics, 96(12), 7629 (2004).

[8] Y.-J. Lee, S.-H. Kim, J. Huh *et al.*, “A high-extraction-efficiency nanopatterned organic light-emitting diode,” Applied Physics Letters, 82(21), 3779 (2003).

[9] C. J. Yates, I. D. W. Samuel, P. L. Burn *et al.*, “Surface plasmon-polariton mediated emission from phosphorescent dendrimer light-emitting diodes,” Applied Physics Letters, 88(16), 161105 (2006).

[10] J. J. Shiang, “Experimental demonstration of increased organic light emitting device output via volumetric light scattering,” Journal of Applied Physics, 95(5), 2889 (2004).

[11] B. Perucco, N. A. Reinke, D. Rezzonico *et al.*, “On the exciton profile in OLEDs-seamless optical and electrical modeling,” Organic Electronics, 13(10), 1827-1835 (2012).

[12] J. M. Ziebarth, and M. D. McGehee, “A theoretical and experimental investigation of light extraction from polymer light-emitting diodes,” Journal of Applied Physics, 97(6), 064502 (2005).

[13] J. van den Brand, J. de Baets, T. van Mol *et al.*, “Systems-in-foil – Devices, fabrication processes and reliability issues,” Microelectronics Reliability, 48(8–9), 1123-1128 (2008).

[14] P. Waide, and S. Tanishima, [Light's Labour's Lost] International Energy Agency, (2005).

[15] M. C. Gather, M. Flämmich, N. Danz *et al.*, "Measuring the profile of the emission zone in polymeric organic light-emitting diodes," Applied Physics Letters*,* 94(26), 263301 (2009).

[16] M. Flämmich, M. C. Gather, N. Danz *et al.*, "Orientation of emissive dipoles in OLEDs: Quantitative in situ analysis," Organic Electronics*,* 11(6), 1039-1046 (2010).

[17] S. L. M. van Mensfoort, M. Carvelli, M. Megens *et al.*, "Measuring the light emission profile in organic light-emitting diodes with nanometre spatial resolution," Nat Photon*,* 4(5), 329-335 (2010).

[18] B. Ruhstaller, E. Knapp, B. Perucco *et al.*, [Advanced Numerical Simulation of Organic Light-emitting Devices], (2011).

[19] S. Harkema, S. Mennema, M. Barink *et al.*, "Large area ITO-free flexible white OLEDs with Orgacon PEDOT:PSS and printed metal shunting lines," 74150T-74150T (2009).

[20] S. Reineke, F. Lindner, G. Schwartz *et al.*, "White organic light-emitting diodes with fluorescent tube efficiency," Nature*,* 459(7244), 234-8 (2009).

[21] H. Greiner, "Light Extraction from Organic Light Emitting Diode Substrates: Simulation and Experiment," Japanese Journal of Applied Physics*,* 46(7A), 4125-4137 (2007).