Efficiency enhancement of organic based Light Emitting Diodes using a scattering layer.

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This paper presents an investigation of organic LED extraction efficiency enhancement using a low refractive index scattering layer. A scattering model is developed based on rigorous electromagnetic modelling techniques. The model accounts for proportions of scattered guided and radiation modes as well as the efficiencies with which emitted and scattered light are extracted. Constrained optimisation techniques are implemented for a single operation wavelength to maximise the extraction efficiency of a generic OLED device. Calculations show that a 2 fold efficiency enhancement is achievable with a correctly engineered scattering layer. The detailed analysis of the enhancement mechanism highlights ways in which this scheme could be improved.

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I. INTRODUCTION

An important consideration in the design of any opto-electronic device is the efficiency with which light can be extracted. A fundamental limitation on all light emitting materials with refractive indices greater than the collection medium is that light emitted outside the critical angle with respect to the collection medium will be trapped by total internal reflection. One successful approach that overcomes this problem is to texture the surface of devices so that trapped light can be scattered into the critical angle of extraction \[ n \approx n_{ITO} \approx 1 \]. This approach works well for inorganic devices where the scattering surface is very close to the emission region \[ n \approx n_{ITO} \approx 1 \]. In organic based LEDs the textured surface is generally separated from the emission zone by the thickness of the substrate (\( \approx 1 \text{ mm} \)). Although light trapped in the substrate can be extracted \[ n \approx n_{ITO} \approx 1 \], the most significant improvements require high refractive index substrates that are matched to the emission region \[ n \approx n_{ITO} \approx 1 \]. Introducing wavelength size texturing closer to the active layers is generally not feasible as this would compromise the device’s electrical properties.

The use of a microcavity to redistribute emission geometrically has been demonstrated in both organic \[ n \approx n_{ITO} \approx 1 \] and inorganic material systems \[ n \approx n_{ITO} \approx 1 \]. Despite their effectiveness, microcavities only provide enhancement over a limited spectral range and are significantly more elaborate in design. A more recent demonstration has shown that low refractive index hydrophobic aerogel materials can be used to successfully enhance extraction efficiency in organic based LEDs \[ n \approx n_{ITO} \approx 1 \]. In this approach, trapped light from an emitting region \( n = 1.8 \) is encouraged to couple evanescently through the higher index Indium Tin Oxide (ITO) anode \( n_{ITO} \approx 1.9 \) to a thick aerogel layer \( n < 1.1 \). Angular resolved intensity enhancements near a factor of 1.5 were reported; likely to be less in terms of actual power enhancement. A theoretical study of volumetric light scattering in such low refractive index substrates shows that significant improvements can result in combining these two enhancement mechanisms \[ n \approx n_{ITO} \approx 1 \]. Here a similar approach is examined which combines the successful texturing / scattering approach with the use of low refractive index layers. A scattering layer, consisting of high refractive index dielectric spheres embedded in a low refractive index material, is placed beside the anode of an organic device as depicted in Fig. 1. This is possible because of the surface flatness and uniformity of the scattering layer, developed by Mitsubishi Chemicals Corporation, that does not interfere with the electrical properties of the device. The distance between the emission region and this layer is comparable to the wavelength of light allowing efficient scattering of low order guided modes that constitute a large proportion of trapped light. Scattered light is effectively redistributed within the lower refractive index medium from where it can be extracted with greater efficiency.

Consider the channels through which emission is extracted or trapped within the device. Light originates via spontaneous emission from the emission region in either radiation modes which can escape the device with efficiency \( \eta_{c}(\lambda) \), or guided modes that are trapped, \( (1 - \eta_{c}(\lambda)) \). Proportions of radiation, \( \gamma_{R}(\lambda) \), and guided, \( \gamma_{G}(\lambda) \), modes are then redistributed into the same set of radiation and guided modes inside the scattering layer. Scattered radiation modes are extracted from the scattering layer with efficiency \( \eta_{r}(\lambda) \). Figure 1 identifies the 6 emission channels whose descriptions and resulting efficiencies are given below.

1. Unscattered radiation modes: \( \eta_{c}(\lambda)(1 - \gamma_{R}(\lambda)) \).
in a slightly different form. Eqn. (1) expresses this inequality
\[ \eta > \eta(0) \]
which is given by the sum of those emission channels that re-
\[ \gamma \eta(0) \]
\[ \gamma \eta(0) \]
\[ \gamma \eta(0) \]
\[ \gamma \eta(0) \]
\[ \gamma \eta(0) \]

Let \( \eta(0) \) be the extraction efficiency when the scat-
\[ \eta(0) \]
\[ \eta(0) \]
\[ \eta(0) \]
\[ \eta(0) \]
\[ \eta(0) \]

where \( n_c \) and \( n_s \) are the refractive indices of the emitting
\[ \gamma \eta(0) \]
\[ \gamma \eta(0) \]
\[ \gamma \eta(0) \]
\[ \gamma \eta(0) \]

By substituting \( g\gamma G(\lambda) = g\gamma(\lambda) = \lambda R(\lambda) \) into
\[ \gamma \eta(0) \]
\[ \gamma \eta(0) \]
\[ \gamma \eta(0) \]
\[ \gamma \eta(0) \]
\[ \gamma \eta(0) \]

Here, the distribution of emitted and scattered light is assumed
to be uniform and \( \theta_s \) and \( \theta_c \) are the critical
coupling angle for the scattering layers. As an example, consider
\[ \eta(0) \]
\[ \eta(0) \]
\[ \eta(0) \]
\[ \eta(0) \]
\[ \eta(0) \]

II. ORGANIC LED DESIGN.

In the following study, a simplified generic OLED design is
\[ \eta(0) \]
\[ \eta(0) \]
\[ \eta(0) \]
\[ \eta(0) \]
\[ \eta(0) \]

where the refractive indices and layer thicknesses for a typical
\[ \eta(0) \]
\[ \eta(0) \]
\[ \eta(0) \]
\[ \eta(0) \]
\[ \eta(0) \]

The basic structure of the device is designed to enhance the extraction of radiation modes to a first order scattering approximation at a wavelength

FIG. 1: Identification of the channels of emission for an OLED with a low refractive index scattering layer.

1. Emission
2. Scattering
3. Guided Mode
4. Radiation Mode
5. Glass
6. Scatter
7. ITO
8. Emitter
9. Al

2. Radiation modes scattered into radiation modes: \( \eta(1) = (1 - \eta(0)) \eta(0) \).
3. Guided modes scattered into radiation modes: \( \eta(1) = (1 - \eta(0)) \eta(0) \).
4. Radiation modes scattered into guided modes: \( \eta(1) = (1 - \eta(0)) \eta(0) \).
5. Guided modes scattered into guided modes: \( \eta(1) = (1 - \eta(0)) \eta(0) \).
6. Unscattered guided modes: \( \eta(1) = (1 - \eta(0)) \eta(0) \).

Let \( \eta(0) \) be the extraction efficiency when the scattering
strength is zero. The first order efficiency, \( \eta(1) \), is
given by the sum of those emission channels that result
in the extraction of radiation modes. When \( \eta(1) \) is greater
than \( \eta(0) \), the scattering layer provides an improvement in efficiency. Eqn. (1) expresses this inequality in a slightly different form.

\[ \frac{\gamma G(\lambda)}{\gamma R(\lambda)} > \frac{\eta(1) \eta(0) (1 - \eta(0))}{\eta(0) (1 - \eta(0))} \]
(1)

Given that the extraction efficiency from a dielectric medium into air is approximated by \( 1 - \cos \theta_c \) where \( \theta_c \) is the critical angle with respect to air. This assumes both uniform emission and scattered light distributions. Eqn. (1) can be approximated by,

\[ \frac{\gamma G(\lambda)}{\gamma R(\lambda)} > \frac{\frac{n_c}{\sqrt{n_c^2 - 1}} - 1}{\frac{n_s}{\sqrt{n_s^2 - 1}} - 1} \]
(2)
of $\lambda = 450$ nm. The augmentation of the efficiency due to the scattering layer will be investigated by controlling three parameters, specified in Table I. The parameters are the scattering layer thickness $d_s$, refractive index $n_s$ and the hole conduction layer thickness, $d_h$. During the optimisation, the scattering coefficient, $\alpha_s$, is kept constant. Currently, Mitsubishi have successfully engineered scattering layers with $\alpha_s d_s = 0.33$ so $\alpha_s = 0.33/d_s$. The constrained optimisation problem is formally stated in Eqn. 4.

$$\max_{d_s, n_s, d_h} \eta_c(1)(d_s, n_s, d_h)$$

s.t. \hspace{1cm} 200 \leq d_s \leq 800 \\
\hspace{1cm} 1.1 \leq \Re\{n_s\} \leq 1.4 \\
\hspace{1cm} 50 \leq d_h \leq 200 \hspace{1cm} (4)$$

Here, the objective function is the efficiency from a first order scattering enhancement. Although multiple scattering has been considered later in the report, introducing it in the objective function seriously increases the computation time of local solutions. A global solution was determined by successive optimisation searches using distributed starting points spanning the parameter space. A total of 125 device configurations were considered resulting in three local maximum solutions. The global maximum solution is shown in Table I and is used throughout the paper.

The operation of optical devices in the spontaneous emission regime involves spectral, spatial and angular parameterisations. For example, Figure 2 (a) shows the electric field intensity at normal incidence and 30° with respect to air. Notice in particular the field overlap at the shaded emission region. (b) The radial plot shows the strength of the electric field intensity in the emission region as a function of emission angle. Points corresponding to the field plots in Fig. 2 (a) are indicated by circular markers.

In the present study, the four efficiency parameters are calculated by integrating over solid angles leaving only the wavelength as the independent variable. This simplifies the problem greatly, but, it is important to remain mindful of the internal angular and spatial variations. In addition, the polarisation is eliminated from the study by averaging over dipole orientations. For details of the techniques and models incorporated here, the reader is referred to Ref. 13.

### III. EVALUATION OF MODEL COMPONENTS.

#### A. Evaluation of the underlying extraction efficiency, $\eta_c^{(0)}$

The extraction efficiency, $\eta_c^{(0)}$, of the OLED device is calculated by integrating angular emission results, such as those in Fig. 2 (b), over solid angle for $\alpha_s = 0$. 

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**Table I: Table specifying the OLED design under investigation.** Since the glass substrate is 0.7 mm thick, it is modelled as an incoherent layer. Here the refractive indices are given for 450 nm. * indicates a refractive index value that has been either determined or estimated experimentally.
Consider, the impact of the optimum solution on $\eta_c^{(0)}$: Notice that interference effects due to the large contrast between the ITO and scattering layer will result in weak cavity effects that enhance $\eta_c$. This is observed in Fig. 2 (b), which shows that emission at 17.3° is preferred in the optimised design. Lobed emission like this is a result of the weak cavity being detuned from the preset wavelength of 450 nm. This is preferential as there is a larger density of off-axis emission states [7]. The tuning wavelength of the weak cavity is given approximately by $\lambda_0 = 450/\cos \theta_{\text{max}}$ where $\theta_{\text{max}}$ is the angle of maximum emission. Here, $\lambda_0 = 456.3$ nm.

Evidently, interference effects are important in the determination of this model component as the thickness of the scattering layer is a multiple of $\lambda_0/4$. Here, $3\lambda_0/4n_s \approx 311.1$ nm $\approx d_s$.

**B. Evaluation of the scattered light extraction efficiency, $\eta_s$**

The extraction efficiency of scattered light, $\eta_s$, can be calculated provided the scattered light distribution within the layer is known. Here, the distribution of scattered light is assumed to be uniform. This is a good approximation provided scatterers are much smaller than the wavelength of light. This can be shown for small spherical dielectric scatterers using Mie theory [10].

Following a scattering event, light is collected from the low refractive index layer. Since subsequent scattering events are considered separately in Section III E, the complex part of the scattering layer refractive index is dropped for the calculation of $\eta_s$. Under these conditions, $\eta_s$ represents the upper limit on extraction from the scattering layer. As expected from the discussion in the introduction, the optimum choice of refractive index in the scattering layer corresponds to the lower bound.

**C. Evaluating the proportion of scattered radiation mode power, $\gamma_R$**

$\gamma_R$ is a measure of the strength of radiation mode scattering within the device. It is calculated by considering the difference between a device with $(\alpha_s \neq 0)$ and without $(\alpha_s = 0)$ scattering. Where there is no scattering, the extraction efficiency is $\eta_c^{(0)}$. With scattering, the zero order extraction efficiency is reduced to $\eta_c = (1 - \gamma_R)\eta_c^{(0)}$. Therefore, $\gamma_R$ is given by Eqn. 6.

$$\gamma_R = \frac{\eta_c^{(0)} - \eta_c}{\eta_c^{(0)}}$$

Figure 3 illustrates important features of the $\gamma_G(\lambda)$ calculation. Here, the absorption, emission and scattering of the TE1 mode is shown, where the Solid Line corresponds to $|E(\lambda, \alpha_s = 0)|^2$ and the broken line to $|E(\lambda, \alpha_s \neq 0)|^2$. $k_{ai}(\lambda)$ allows the calculation of mode attenuation due to scattering. Absorption, $k_{ai}(\lambda)$, is strongest at the metal surface.

**D. Evaluating the proportion of scattered guided mode power, $\gamma_G$.**

The proportion of scattered guided modes, $\gamma_G$ is evaluated by examining the internal fields of the device. Despite this, the calculation approach is similar to the evaluation of $\gamma_R$. The optical field intensity, $|E(\lambda, \theta_c, \alpha_s = 0)|^2$, within the emission region in the absence of scattering $(\alpha_s = 0)$ can be directly compared with the field intensity, $|E(\alpha_s, \theta_c \neq 0)|^2$, for $\alpha_s \neq 0$ to give a value for $\gamma_G$. Note that this does not take into account the lateral extent of the device, although this will be considered shortly. Fig. 8 illustrates the relevant components in these guided mode scattering calculations for the TE1 mode corresponding to a particular internal angle, $\theta_c$. $\gamma_G$ is evaluated using the expression in Eqn. 6 by integrating over the internal emission solid angle, $\Omega_c$.

$$\gamma_G = \frac{\int_{\Omega_c} |E(\alpha_s = 0)|^2 - |E(\alpha_s \neq 0)|^2 d\Omega}{\int_{\Omega_c} |E(\alpha_s = 0)|^2 d\Omega}$$

Figure 4 plots $|E(\lambda, \theta_c, \alpha_s = 0)|^2$ and $|E(\alpha_s, \theta_c \neq 0)|^2$ highlighting the effect of the scattering layer on the optical field within the device against the internal angle, $\theta_c$. Here, 90° corresponds to propagation in the plane of the device with respect to the light emitting region ($\alpha = 1.8$). TE emission is shown in Fig. 4 (a) while TM emission is shown in Fig. 4 (b). The arrows indicating the peaks correspond to the solutions of guided modes considered later; the effective mode angles (angles at peaks in optical field) correspond to those given in Table 4. The amount of scattering can be gauged by the difference between the broken lines $(\alpha_s = 0)$ and the solid lines $(\alpha_s \neq 0)$. In this...
FIG. 4: Examination of the scattering of guided modes in an effective plane wave angle representation for (a) TE and (b) TM polarisations. $|E(\lambda, \alpha_s = 0)|^2$ are represented by solid lines and $|E(\lambda, \alpha_s \neq 0)|^2$ by broken lines showing the change in guided mode coupling with the OLED due to scattering at $\lambda = 450$ nm.

A mode $i$ propagating within the device has a propagation constant, $\beta_i$. $\beta_i$ is in general complex and is calculated using the Argument Principle Method [14, 15]. The complex propagation constant of a mode describes the phase velocity of propagation in the plane of the device and the mode attenuation as it propagates, given by the real and imaginary parts respectively. The imaginary components of the propagation constant $k_{si} + k_{ai}$ for the $i^{th}$ guided mode will indicate mode attenuation due to scattering and absorption respectively. $k_{ai}$ can be evaluated by subtracting modal propagation constants calculated with and without scattering, $\alpha_s$.

Table II shows the complex propagation constants, $\beta_i(\alpha_s)$, for the optimised OLED device specified in Tab I for the scattering layer with $\alpha_s = 0$ and $\alpha_s \neq 0$. Notice that the internal mode angles, $\theta_i$, relative to the emission region are shown and correspond directly to the indicated modes in Fig. 4.

The complex eigenvalues in Tab. II correspond to a guided mode eigenvector within the OLED. The electric field distribution for the modes are shown in Fig. 5. Note that the eigenvalues of the mode functions calculated here correspond to the peaks in emission distribution in Figs. 4 (a) and 4 (b). The mode functions indicate both the degree of absorption and the scattering in the different regions of the device. Firstly, the two polarisations are
TABLE II: Propagation constants, $\beta_i$ for the most important modes of an OLED. The two sets of results (with and without scattering) allow calculation of $k_{s,i}$. In addition, the internal mode angles, $\theta_i$, with respect to the emitting region are also given.

| Mode | $\beta_i(\alpha_s = 0)$ [nm$^{-1}$] | $\beta_i(\alpha_s \neq 0)$ [nm$^{-1}$] | $k_{s,i}$ | $\theta_i$ | $L$ |
|------|----------------------------------|----------------------------------|---------|---------|-------|
| TM$_1$ | $1.550 \times 10^{-2} - 5.398 \times 10^{-5}$ | $1.550 \times 10^{-2} - 5.990 \times 10^{-4}$ | $-1.54 \times 10^{-4}$ | 38.07° | 0.7 µm |
| TE$_2$ | $1.953 \times 10^{-2} - 1.601 \times 10^{-4}$ | $1.953 \times 10^{-2} - 2.592 \times 10^{-4}$ | $-9.90 \times 10^{-5}$ | 51.99° | 10.1 µm |
| TM$_2$ | $2.321 \times 10^{-2} - 3.205 \times 10^{-4}$ | $2.321 \times 10^{-2} - 4.134 \times 10^{-4}$ | $-9.29 \times 10^{-5}$ | 67.44° | 10.8 µm |
| TE$_1$ | $2.535 \times 10^{-2} - 1.986 \times 10^{-4}$ | $2.535 \times 10^{-2} - 2.188 \times 10^{-4}$ | $-2.03 \times 10^{-5}$ | $> 90°$ | 49.3 µm |
| TM$_1$ | $2.678 \times 10^{-2} - 3.427 \times 10^{-4}$ | $2.678 \times 10^{-2} - 3.471 \times 10^{-4}$ | $-4.40 \times 10^{-6}$ | $> 90°$ | 227.3 µm |

So far the internal components of the calculations have been examined in detail. Now consider the behaviour of the model components as a function of the scattering strength. The strength of scattering ultimately governs the degree of mixing between the radiation and guided modes of the device. As the scattering is increased, there will also be an increase in enhancement factor of the underlying device. This behaviour is seen in Fig. 6.

Fig. 6 (a) shows the variation of the model components as a function of the scattering strength. The enhancement factor has been compared to the efficiency $f(\alpha_s d_s)$, for the case of multiple scattering. Here, the enhancement factor has been compared to the efficiency of a device without a scattering layer, $\eta_{c,0} = 27.8\%$. The enhancement factor $f(\alpha_s d_s)$ is given by the following expression related to Eqn. 3:

$$f(\alpha_s d_s) = \frac{\eta_c(0)}{\eta_{c,0}} + \left(\eta_s - (1 + g(1 - \eta_s))\frac{\eta_c(0)}{\eta_{c,0}}\right) \gamma + \mathcal{O}(\gamma^2)$$

(9)

In the absence of absorption, there is an enhancement of about 20% due to the presence of the low index scattering layer, $c.f.$ the first term in Eqn. 3. This is comparable to enhancements obtained by Tsutsui et al in

E. Calculating multiple scattering.

The device efficiency due to first order scattering can be expressed as,

$$\eta_c^{(1)} = M_1\eta_c^{(0)} + C_1$$

(7)

where $M_1 = 1 - \gamma_R + \eta_s(\gamma_R - \gamma_G)$ and $C_1 = \gamma_G \eta_s$. The efficiency for higher order scattering can similarly be expressed as,

$$\eta_c^{(n)} = M_n\eta_c^{(n-1)} + C_n$$

(8)

$M_n$ and $C_n$ are given by expressions similar to $M_1$ and $M_2$, however, use the new model components $\gamma^{(2)}_G$ and $\gamma^{(2)}_R$ to quantify the proportions of light that are scattered a second time and subsequent time. These parameters have been calculated using similar techniques to those described in Sections III (a) and III (b). In the case of the optimisation calculation, where $\alpha_s d_s = 0.33$, multiple scattering contributes 5% of the efficiency.

IV. RESULTS.

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their experiments with aerogel layers. As the scattering strength is increased, there is an initial reduction in efficiency, mainly due to second order scattering loss in the low refractive index region. Above a scattering strength of 0.05, however, $f(\alpha_s d_s) > 1$ and increases monotonically. For the current design an enhancement that is 95% of the limiting value of $\eta_s/\eta_0$ is attained. This is still almost a two-fold enhancement in the efficiency. With the current engineering capabilities of Mitsubishi Chemicals Corporation, nearly 75% of the upper limit can be achieved, corresponding to a 60% enhancement.

The first order scattering provides the largest contribution to the enhancement factor. Indeed the first order scattering coefficient in Eqn. (9) encapsulates the limitations of this enhancement approach. The primary limitations, in order of importance, are the extraction efficiency from the low refractive index medium, $\eta_s$, the strength of guided mode scattering, $\gamma_g$, and the ratio of radiation to guided mode scattering, $g$. $\eta_s$ must be maximised, requiring a scattering layer with as low a refractive index as possible. Here, the optimum value of $\eta_s = 1.1$ is at the lower bound of what can be achieved in the fabrication process. Alternative materials such as aerogels, which have refractive indices as low as 1.01, would show even larger enhancements. However, the choice of material must be compatible with the formation of dielectric spheres to provide the required scattering strength, $\gamma$. Maximising $\gamma$ allows the device to attain a larger fraction of the limiting enhancement factor. Finally, the ratio of guided to radiation modes, $g$, must be minimised.

In Sec. IV, the calculation of the guided mode scattering component, $\gamma_g$, is detailed. The reader is therefore referred to this section for an in depth appraisal of the factors that effect the value of $g$. One of the most significant factors in the guided mode scattering strength is the strong absorption in the high refractive index anode region (ITO) to which modes are confined. At $\alpha_s d_s = 0.33$, guided mode absorption is approximately 3/2 times larger than scattering. In contrast, scattering is approximately 4 times larger than the absorption for the radiation modes. Low absorption near the active components of the device is clearly crucial for minimising $g$ and maximising the efficiency enhancement.

The principles of operation presented here suggest enhancements of up to a factor of 2 could be achieved with a carefully designed device incorporating a scattering layer. Although this is comparable to microcavity enhancements, here, the enhancement is achievable across the visible spectrum. The design of an enhancement layer for a broad spectral range would be limited by the scattering strength drop-off at red wavelengths and the difficulty associated with maximising coherent reflections. Despite this, calculation of the device structure investigated here using scattering data from Mitsubishi at green and red wavelengths show $f(\lambda = 550) = 1.5$ and $f(\lambda = 630) = 1.3$. In addition, the device design could be optimised for overall broad spectral performance.

V. CONCLUSIONS.

A perturbative model was developed for the description of low refractive index scattering layers that enhance the extraction efficiency of light from organic LEDs. Components of the model were calculated using rigorous electromagnetic techniques. The scattering model was used to optimise an OLED design incorporating a scattering layer. The calculations show that a two-fold enhancement in the extraction efficiency is attainable.

Three parameters were highlighted as crucial to the enhancement mechanism. Most importantly a low refractive index medium that supports high refractive index...
scattering particles is necessary to set the limiting enhancement factor and the scattering strength required to attain it. Finally, the ratio of radiation to guided modes must be minimised. Optimisation of the first two parameters is difficult as they depend on complex fabrication techniques. In contrast, careful device design could allow the ratio of radiation guided mode scattering to be reduced.

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