Multi-Objective Optimization of Microcavity OLEDs with DBR Mirror

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Abstract

In this work, the emission efficiency and spectral shift with respect to viewing angle were optimized by optimizing the design of the multi-layer top mirror of a microcavity OLED device. We first established criteria for the emission side mirror in order to optimize light intensity and spectral shift with viewing angle. Then we designed mirror using metallic and dielectric layers based on the target defined. The electroluminescence emission spectra of a microcavity OLED consisting of widely used organic materials, N,N′-di(naphthalene-1-yl)-N,N′-diphenylbenzidine (NPB) as a hole transport layer and tris (8-hydroxyquinoline) (Alq3) as emitting and electron transporting layer was then calculated. Silver was used as the anode and back reflection mirror for the microcavity OLED. The simulation was performed for both the conventional LiF/Al cathode/top mirror and the optimized 5-layered top mirror. Our results indicate that by following the design procedure outlined, we simultaneously optimize the device for better light intensity and spectral shift with viewing angle.

Keywords: Microcavity OLEDs, Mirror Design, Optimization

1. INTRODUCTION

OLEDs have emerged as potential candidates for applications in display devices due to their many prominent advantages such as small size, high brightness, low manufacturing cost and wide viewing angle. Since the development of first prototype of OLED by Tang and Van Slyke from Kodak, intense efforts have been seen in past decade on improving device performance thus leading to commercializing of small size OLED panels on market today. In order to further improve and optimize this device for use in practical applications, device modeling of OLED characteristics is required to better understand the physical processes affecting the device performance. Furthermore, a comprehensive design procedure that allows for multi-objective optimization will be required so that enhancing a particular display property is not achieved at the expense of degrading other characteristics. Although work based on electrical and optical modeling have been documented separately in numerous studies, comprehensive device simulation that includes both electrical and optical models, let alone an in depth multi-objective device design procedure, has been. In this work, we aim to investigate into the simulation models in order to gain insight into the requirements for multi-objective optimization and perform microcavity OLED design based on these requirements.

The metallic mirror optical microcavity is a simple and effective device structure for enhanced light extraction and color purity via modified spontaneous emission. However, the metallic electrode (which acts as the mirror) used typically display poor carrier injection characteristics when compared to LiF/Al. In order to maintain both the efficient injection of LiF/Al and the desired mirror properties of metals such as Ag, several groups had experimented with top mirror consisting of LiF/Al cathode enhanced by metallic and dielectric capping layers. However, as the number of layers in the mirror increases, device optimization becomes increasingly difficult and time consuming. Furthermore, the method of randomly choosing a number of metallic and dielectric layers and then optimize accordingly does not provide us with a systematic procedure for effective microcavity OLED mirror design. Another problem with metallic microcavity OLEDs is that it typically displays spectral shift with change in viewing angle, an undesirable feature that must be suppressed for practical applications.
In this work, we begin by examining a popularly used optical model\textsuperscript{6-8} to find out the ideal mirror characteristics for improving light emission and reducing spectral shift with view angle. We then design mirrors containing metallic and dielectric layers based on the criteria established. The paper first describes the optical and electrical simulation models used including the simulation parameters. Then the design criteria and the model used for mirror design and optimization are described. Finally, the optimized device is compared with the conventional LiF/Al device.

2. DESCRIPTION OF THE MODEL

2.1. Electrical model

Inside the organic semiconductor the electrical transport is modeled by the one-dimensional time-independent drift-diffusion model\textsuperscript{15-17}, which solves for a self-consistent solution of electron density, $n$, hole density, $p$ and potential $\psi$ using the semiconductor solver Atlas\textsuperscript{18}. The output of the electrical model (recombination rate) forms part of the input for the optical model. The model includes:

The continuity equation for $n$ (electrons) and $p$ (holes)

\begin{align}
\frac{d}{dx}(-\mu_n n \frac{d\psi}{dx} + D_n \frac{dn}{dx}) &= R \\
\frac{d}{dx}(\mu_p p \frac{d\psi}{dx} + D_p \frac{dp}{dx}) &= R
\end{align}

where $\mu_n$ and $\mu_p$ are the electron and hole mobilities and $D_n$ and $D_p$ are diffusion constants and $R$ is the recombination rate. The $\mu$ and $D$ are related by the well known Einstein relation. The carrier mobilities are modeled by the field-dependent form:

\begin{align}
\mu_n(E) &= \mu_{n0} \exp \left( \frac{E}{E_0} \right) \\
\mu_p(E) &= \mu_{p0} \exp \left( \frac{E}{E_0} \right)
\end{align}

where $\mu_{n0}$ and $\mu_{p0}$ are the zero field mobilities, $E$ is the electric field and $E_0$ is the constant known as characteristic field.

The recombination rate is taken to be optical only and modeled by the Langevin recombination coefficient $\gamma$\textsuperscript{15,16}:

\begin{align}
R_{\text{opt}} &= \gamma (pn - n_i^2) \\
\gamma &= \frac{4\pi e \mu_R}{\varepsilon_{r}\varepsilon_0}
\end{align}

where $n_i$ is the intrinsic concentration and $\mu_R$ is effective recombination mobility, taken to be the larger of the electron and hole mobilities in the material, $\varepsilon_{r}\varepsilon_0$ is the permittivity of the material. The effect of traps in the organic layers is not included in the current electrical model as the literature indicates the inclusion of traps has no significant effect on the simulation results obtained\textsuperscript{15}.

2.1.1. Poisson’s equation

\begin{align}
\frac{d^2\psi}{dx^2} = -\frac{e}{\varepsilon_r\varepsilon_0} \left[ N_D(x) - N_A(x) + N_D - N_A \right]
\end{align}

where $N_D$ and $N_A$ are the ionized donor and acceptor dopant concentrations.

These equations are solved for the p-n junction structure using Schottky contact boundary conditions between a metal (which also serves as the reflecting surface for optical modeling) and the organic layer at the anode and the cathode. The barrier heights governing carrier injections are: $\phi_m$ for electrons and $\phi_p$ for holes and are related to the metal work function $\phi_m$ of the electrodes and the electron affinity of the organic material $\chi_e$:
The continuity equations and the Poisson equation are solved to obtain the carrier concentrations, electric field distributions and recombination rate. The thickness of recombination region can be determined from the recombination rate, which can be used to estimate the width of emission region (taking into account of exciton diffusion) to be included into the optical model.

2.1.2. Parameters used in simulation models

The material parameters used for modelling of carrier transport are obtained from literature\textsuperscript{15-17}. The devices were simulated with forward bias of 5 volts. The mobility of majority carriers in the organic materials has been set to be two orders of magnitude higher than its minority carriers. Therefore NPB and Alq\textsubscript{3} are assumed to act as hole transport layer (HTL) and emission (EML) layer respectively. Where the simulation model requires a large number of material parameters only a few critical parameters including barrier heights to carrier injection, bandgaps of the organic materials and carrier concentrations have marked effects on the simulated result\textsuperscript{15}.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>NPB</th>
<th>Alq\textsubscript{3}</th>
</tr>
</thead>
<tbody>
<tr>
<td>Relative Permittivity</td>
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<td>3.0</td>
</tr>
<tr>
<td>$\mu_{no}$ (cm\textsuperscript{2}/Vs)</td>
<td>6.1·10\textsuperscript{-6}</td>
<td>1.9·10\textsuperscript{-6}</td>
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<tr>
<td>$\mu_{po}$ (cm\textsuperscript{2}/Vs)</td>
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<td>7.1·10\textsuperscript{4}</td>
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<td>1·10\textsuperscript{21}</td>
</tr>
<tr>
<td>$N_v$ (cm\textsuperscript{-3})</td>
<td>1·10\textsuperscript{21}</td>
<td>1·10\textsuperscript{21}</td>
</tr>
<tr>
<td>$N_A$ (cm\textsuperscript{-3})</td>
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<td>--</td>
</tr>
<tr>
<td>$N_D$ (cm\textsuperscript{-3})</td>
<td>--</td>
<td>1·10\textsuperscript{9}</td>
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<tr>
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<td>2.7</td>
</tr>
<tr>
<td>$\chi_c$ (eV)</td>
<td>2.4</td>
<td>3.0</td>
</tr>
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</table>

Table 1 Electrical simulation parameters

2.2. Optical model

The resonant modes of a microcavity have to satisfy the condition that the phase change during one round trip is a multiple of 2\textpi. In other words, for normal incidence the following equation holds:

$$\sum d_i n_i(\lambda) - \phi_{top}(0, \lambda) - \phi_{bot}(0, \lambda) = 2m\pi,$$

where $\lambda$ is the emission wavelength, $\phi_{top}(\lambda)$, $\phi_{bot}(\lambda)$ are the wavelength dependent phase changes upon reflection from top and bottom mirrors, respectively, $m$ is an integer which defines the mode number, and the summation is performed over all the layers inside the cavity with thicknesses $d_i$ and refractive indices $n_i(\lambda)$. The phase shift upon reflection from the mirrors is calculated using matrix method\textsuperscript{19}.

The intensity of the light output in the form of the electroluminescence (EL) spectrum was calculated using the method based on the equivalence between the probability of photon emission and the power radiated by a classical dipole antenna\textsuperscript{20-23}.

Consider the structure shown below.
This cavity consists of two multilayer film DBR mirrors with a spacer layer sandwiched between them. In a layer with complex refractive index $N_i$, the amplitude of the wave vector is given by:

$$k_i = \frac{2\pi}{\lambda} N_i,$$  

where $\lambda$ is the emission wavelength. The complex refractive index for each layer of the device was determined using Variable Angle Spectroscopic Ellipsometry (VASE). The complex refractive index is then determined from Ellipsometry data by using the Lorentz-Drude model\textsuperscript{24-26}. Lorentz-Drude model parameter extraction was performed by using simulated annealing algorithm.

When considering wave propagation through thin film layers, it is convenient to resolve the wave vector into two components: the component normal to the direction of propagation and the component along the direction of propagation (we use the $z$-axis as the axis of propagation), which is given by:

$$\kappa = k_i \sin(\alpha_i),$$ 

and

$$k_{z,i} = k_i \cos(\alpha_i) = \sqrt{k_i^2 - \kappa^2},$$

respectively, where $\alpha_i$ is the angle the wave makes with the $z$-axis.

The total power $F$ emitted by a dipole antenna located within the multilayer structure normalized to the power output of the same dipole in an infinite medium is given by:

$$F = \int_0^\kappa K(\kappa) d\kappa^2,$$  

where $K$ is the power density per unit $d\kappa^2$.  

![Figure 1: Microcavity multilayer structure](http://proceedings.spiedigitallibrary.org/)
The power density \( K \) can be resolved into the TM and TE component, with each component separated into power densities for dipoles oriented parallel and perpendicular to the z-axis. With this in mind, the power densities can be defined as:

\[
K_{\perp}^{TM} = \frac{3}{4} \Im \left[ \frac{k_z^2 k_{z,e}^3}{k_e^6} \left( \frac{1 - a_z}{1 - a_{TM}} \right) \left( \frac{1 - a_{TM}}{1 - a_z} \right) \right],
\]

\[
K_{\perp}^{TE} = 0, \tag{11}
\]

\[
K_{\parallel}^{TM} = \frac{3}{8} \Im \left[ \frac{k_z}{k_e} \left( \frac{1 + a_z}{1 - a_{TM}} \right) \left( \frac{1 + a_{TM}}{1 - a_z} \right) \right],
\]

\[
K_{\parallel}^{TE} = \frac{3}{8} \Im \left[ \frac{1}{k_z k_{z,e}} \left( \frac{1 + a_{TM}}{1 - a_{TE}} \right) \left( \frac{1 + a_{TE}}{1 - a_{TM}} \right) \right], \tag{15}
\]

where \( e \) denotes the emissive layer (sandwiched between top and bottom mirrors) and \( a \) is the reflection coefficient of the mirror with respect to the location of the dipole, defined as:

\[
a_{\pm}^{TM/TE} = r_{\pm}^{TM/TE} \exp(-2 j k_{z,e} z_{\pm}) \quad , \tag{16}
\]

\[
a^{TM/TE} = a_{+}^{TM/TE} a_{-}^{TM/TE} \quad , \tag{17}
\]

where \( z_{+} \) is the dipoles’ distance from the top mirror, \( z_{-} \) is the dipoles’ distance from the bottom mirror and \( r \) is the amplitude reflection coefficient of the top and bottom mirrors calculated using the modified transfer matrix approach of Katsidis and Siapkas\(^{27,28} \).

For randomly oriented dipole antenna (equal probability for all orientation direction), the power density is given by:

\[
K_{TM,TE} = \frac{1}{3} K_{\perp}^{TM,TE} + \frac{2}{3} K_{\parallel}^{TM,TE}. \tag{18}
\]

The overall power density will then be the average of the TM and TE component.

### 3. MIRROR DESIGN AND OPTIMIZATION PROCEDURE

Starting from our optical model, we first maximized the light emission from microcavity OLED by varying mirror parameters namely: mirror reflectance \((R)\), transmittance \((T)\) absorptance \((A)\) and phase \((\varphi)\). Once these optimal values have been determined we proceeded to determine the multilayer mirror structure (step two) that will provide the required set of \( R, T, A \) and \( \varphi \) and therefore the maximum light extraction for the device.
For the reduction of spectral shift with viewing angle, we note that the spectral shift is approximately given by\textsuperscript{13, 14}:

\[
\Delta \lambda = \sum_i \sum_j \frac{4 \pi i}{\lambda} n_i \left[ \cos \vartheta_j - 1 \right] + \Delta \varphi_{\text{top}} + \Delta \varphi_{\text{bot}},
\]

where \( \vartheta \) is the viewing angle. It can be seen that by reducing the phase shift on reflection for all angles, we can reduce the spectral shift with viewing angle.

For mirror design and optimization, we used optical thin film design software OptiLayer\textsuperscript{29}. OptiLayer uses the matrix formalism for optical multilayer calculations\textsuperscript{19} and employs an efficient multi-objective optimization algorithm proposed by Furman and Tikhonravov\textsuperscript{30}. The software allows the user to specify "target functions" in the form of \( R, T, A \) and \( \varphi \), then the design can be refined by the needle optimization procedure\textsuperscript{30}.

### 4. RESULTS AND DISCUSSION

Using LiF/Al as cathode and 70nm Ag as anode, optimum thickness of the cavity is determined to be approximately 120 nm with cavity order of zero. After the thickness of the device was determined, the electrical properties were simulated as shown in Fig. 2. When a forward bias voltage of 5 V was applied, holes were injected from anode into NPB layer and accumulated near the NPB/Alq\textsubscript{3} interface due to offsets in the HOMO bands. Alq\textsubscript{3} layer acted as a hole blocking layer in this case.

![Figure 2: a) simulated carrier density and electric field and b) Simulated recombination rate.](image)

Figure 2a) shows the simulated carrier recombination rate in the same device. It is clearly observed that majority of the recombination occurred within 5 nm from the NPB/Alq\textsubscript{3} interface in the Alq\textsubscript{3} layer. This information becomes the input to our optical model.

The next step is to establish the target function for mirror design. Starting from our optical model, we modified the model so that instead of having layer thicknesses and complex refractive indices as input parameters, the model now takes mirror parameters reflectance, transmittance, absorptance and phase shift on reflection as input.

Figure 3a) shows the output optical intensity versus top mirror R and T plot. It can be seen that the intensity increases with increasing R and decreasing T, provided the absorption \((1-(R+T))\) is close to zero. This conclusion is in agreement with the design criteria proposed by Benisty \textit{et al.}\textsuperscript{9}. In their paper, Benisty \textit{et al.} also remarked that it is technologically more realistic to aim for \( R \) of 80~90\% and minimal absorption.

Figure 3b) is the contour plot of optical output versus the real and imaginary part of the reflection coefficient of the top mirror (bottom mirror is 70nm Ag and is fixed), the transmittance is assumed to be 10\%. This shows that we want the
imaginary term to be within the region 0~0.2 and the real term to be within the region 0.8~0.9. We performed numeric optimization to determine the exact combination that would yield maximum light out, which turns out to be at (real, imaginary) equal to (0.91502, 0.146239). This corresponds to $R=0.93$ and $\varphi=0.16$ radians (approximate value). Thus we will use $R=0.9$, $T=0.1$, $A=0$ and $\varphi=0.16$ radians (or 9 degrees) as our target for mirror design.

![Figure 3: a) Light intensity with respect to T and R and b) light intensity with respect to real and imaginary part of reflection coefficient of emission side mirror. Black denotes minimum and white denotes maximum in both figures.](image1)

The materials used for the mirror design procedure include Al, Ag, LiF (as the low index dielectric) and NPB (as the high index dielectric). The starting design we used is:

Device 1 => LiF(0.5nm)/Al(5nm)/Ag(10nm)/LiF(20nm)/NPB(20nm),

the optimization algorithm modified and/or adds metallic and dielectric layers to the cathode attempting to modify the mirror characteristics to conform with that of the target specified. Our optimization procedure produced the following modified design:

Device 2 => LiF(0.5nm)/Al(5nm)/Ag(10nm)/NPB(5nm)/Al(10nm).

![Figure 4: a) Normalized EL relative to device 1 and b) Wavelength shift from zero degrees versus viewing angle](image2)
Figure 4a) shows the Electroluminescence (EL) comparison between device 1 (basic device) and device 2 (5 layered mirror). Device one is a generic metallic mirrored microcavity OLED with LiF/Al as the cathode and Ag as anode. It displays good optical properties compared to conventional device with ITO anode and has better carrier injection properties than devices using metallic cathode (such as Ag or Au). In this work we use this metallic mirror microcavity OLED as the basis of our design. It can be seen that the optimized device (device 2) shows a 2.4 times improvement in peak optical output intensity. The spectral peak (wavelength at peak intensity) shift from the peak spectral position at viewing angle to zero degrees is shown in figure 4b). It can be seen that for small viewing angle (< 20 degrees), the spectral shift is small for both devices. However, as the viewing angle is increased, the optimized device shows less spectral shift compared to device 1.

5. CONCLUSION

We have established criteria for optimizing both the output optical intensity and spectral shift with viewing angle for top emitting microcavity OLEDs. The criteria is used for mirror design to come up with a relatively simple 5-layered mirror that improves light emission and reduces spectral shift with viewing angle compared to a conventional device with LiF/Al cathode. Simulation was performed on both devices and the results confirms that the device with 5-layered mirror performs better than the basic two layered mirror device both in terms of peak output optical intensity and spectral shift with respect to viewing angle.

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