

OPTICAL MODELLING OF AN Alq₃-BASED ORGANIC LIGHT-EMITTING DIODE

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A theoretical model that features analytical formulas is developed for the evaluation of the light intensity that is radiated from a layered structure containing optically-active layers. The model is applied to an Alq₃-based organic light-emitting diode to study the convenience of using a LiF/Al cathode and find optimal thicknesses of some of the device layers.

Key words: organic light-emitting diode, Alq₃, lithium fluoride, thin film.

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Introduction

Since 1987, when the pioneering work of Tang and VanSlyke [1] demonstrated the possibility to design high-luminance low-voltage driven devices based on tris(8-hydroxyquinoline)aluminum, Alq₃, thin films, significant progress has been achieved in the development of efficient organic light-emitting diodes (OLEDs) for flat-panel display and lighting. The initially reported typical OLED structure consists of two organic layers, a hole-transport layer involving a diamine-like molecule and an electron-transport layer based on Alq₃, which are sandwiched between a metallic cathode and a semi-conductor anode grown on a glass substrate. Multilayered devices in which the emission wavelength can be tuned and/or the charge injection and carrier transport are increased are under development and their optimization is critically important towards their technological success.

The theory of light emission from a point source, e. g. a single atomic or molecular system, sandwiched between two plane parallel mirrors – either metallic or Bragg reflectors – is well assessed in both its quantum [2–7] and classical [8–15] interpretations, which give equivalent results. Among the many available approaches, the classical one by Benisty and co-workers is based on elementary source terms that depend on dipole alignment and radiate linearly polarized fields that interact with surrounding mirrors

[13, 14]. A suitable superposition of the dipole alignment and polarization states is applied to model the electromagnetic field that is radiated into the external medium [13, 14].

Recently, Benisty's model has been extended to the case of a volume source consisting of a dense collection of component point sources [16], which can be assumed to mutually interact within a certain spatial range and give rise to cooperative emission [16] or coherent phenomena [17]. The convenience of this extended model stems from the analyticity of the mathematical expressions that describe the radiated field in those cases where the Fourier transform of the source density distribution is analytic [16]. One of such cases is that of a homogeneous optically-active layer. As a consequence, the extended model can be used to theoretically foresee the luminescence spatial distribution of a layered active device.

Theory

Benisty's model

Benisty's source terms are the electric-field components that are radiated from a single dipole, defined as [13, 14]

$$\begin{aligned} A_{\uparrow}^{h,s} &\equiv A_{\downarrow}^{h,s} = \sqrt{\frac{3}{16\pi}}, \quad A_{\uparrow}^{h,p} \equiv A_{\downarrow}^{h,p} = \sqrt{\frac{3}{16\pi}} \cos\theta_0, \\ A_{\uparrow}^{v,p} &\equiv -A_{\downarrow}^{v,p} = \sqrt{\frac{3}{8\pi}} \sin\theta_0. \end{aligned} \quad (1)$$

In the above formulas, referring to Fig. 1, the arrow in the subscript indicates the field propagation direction (upwards or downwards), while the superscript letters stand for the dipole alignment and the field polarization state, respectively: $h(v)$ means that the dipole is horizontal (vertical) – that is, parallel (perpendicular) to the layered structure – while $s(p)$ means that the propagating electric-field vector is perpendicular (parallel) to the propagation plane. The propagation angle θ_0 is measured at the dipole position, therefore within the material the medium is embedded in, with respect to the layered structure. As far as the s -polarized field from a vertically-aligned dipole is concerned, its contribution is null ($A_{\uparrow}^{v,s} \equiv A_{\downarrow}^{v,s} = 0$).

Benisty's model for the evaluation of the electric fields and intensities that are outcoupled in the external media is dealt with in detail elsewhere [13, 14, 16]. **Here it is worthwhile recalling** that a point source, such as a single atomic or molecular system, is modelled by means of a randomly-oriented dipole, which, in turn, is considered as the superposition of the elementary alignment and polarization states corresponding to Benisty's source terms [13, 14, 16].

Generalized model

Benisty and co-workers have addressed the case of a single irradiating atomic or molecular system sandwiched between two plane-parallel mirrors, which can be Fabry-Pérot reflectors [13, 14]. **Their theory is here extended to a collection of atomic systems** that, in case their volume density is large enough, can be thought of as a finite and continuous radiating volume. A typical example is an optically-active layer embedded within an optical multilayer.

In a dense collection of atomic systems, the emission features of each of them can be triggered by its neighbouring systems so that cooperative emission takes place. For space reasons, we are not addressing here the topic of cooperative emission from within a layered structure, which is however dealt with elsewhere in detail and with analytical results under suitable conditions [16, 17]. **It is worth pointing out that cooperation effects for light emission from a volume source are likely to be more pronounced when the radiating volume is placed within a cavity with high enough Q parameter** [18] because therein emission modes broader than in the free space are present. For low- Q cavities, such as the device

discussed in this paper, neglecting the otherwise small cooperation effects seems reasonable.

Set equal to $\mu(\mathbf{r})$ the density of atomic systems that form the active volume and $w_{\uparrow\downarrow}(\mathbf{r})$ the intensity radiated from a volume element in \mathbf{r} , where $\mathbf{r} = (x, y, z)$ is the position vector, the global light intensity that is radiated from the structure is

$$W_{\uparrow\downarrow} = \int \mu(\mathbf{r}) w_{\uparrow\downarrow}(\mathbf{r}) d^3r, \quad (2)$$

which, once an observation angle $\theta_{\uparrow\downarrow}$ is set, is true for any dipole alignment and polarization state. Here, the subscript double-arrow is a shorthand notation to represent two equations with one: one corresponding to the first arrow and the other corresponding to the second arrow. As for the source terms of Eq. (1), the direction the arrow points to – either upwards or downwards – indicates the propagation direction.

If the structure consists of a layered medium with an optically-active layer, Eq. (2) gives an analytical result provided the Fourier transform of $\mu(\mathbf{r})$ is analytical. For instance, let us consider a uniform active layer of thickness d and refractive index n_0 bounded by multilayers whose intensity reflection and transmission coefficients are $R_{\uparrow\downarrow}$ and $T_{\uparrow\downarrow}$ for incidence from within the active layer (here too, the double-arrow subscripts are introduced, with obvious meaning, to keep a compact notation). Let the axis z coincide with the symmetry axis of the structure, with z growing upwards and $z = 0$ placed at the centre of the active layer (Fig. 1). An element in $z = z_0$ con-

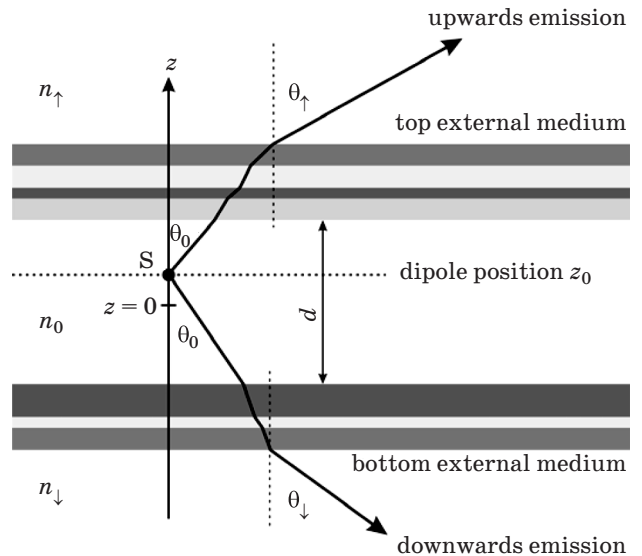


Fig. 1. Light emission from a point source S , represented by a randomly-oriented dipole embedded in a layered medium. The symbols are explained in the text.

tributes to the intensity of light emitted from the active layer with [16]

$$w_{\uparrow\downarrow}(z_0) = \frac{n_{\uparrow\downarrow}^2 \cos \theta_{\uparrow\downarrow}}{n_0 \cos \theta_0} F_{\uparrow\downarrow} \left| A_{\uparrow\downarrow} + r_{\uparrow\downarrow} A_{\downarrow\uparrow} \exp[-ikn_0(d \pm 2z_0) \cos \theta_0] \right|^2, \quad (3)$$

where n_{\uparrow} (n_{\downarrow}) is the refractive index of the top (bottom) external medium,

$$F_{\uparrow\downarrow} = \frac{T_{\uparrow\downarrow}}{\left| 1 - r_{\uparrow\downarrow} \exp(-i2kn_0 d \cos \theta_0) \right|^2} \quad (4)$$

is the Airy function, k is the amplitude of the wavevector in vacuum, and r_{\uparrow} (r_{\downarrow}) is the

complex-amplitude reflectance of the layers above (below) the active layer for incidence from inside the latter. It is worth recalling that the above equations need to be specialized to specific polarization state and dipole alignment.

By integrating according to Eq. (2), the outcoupled power is found to be

$$W_{\uparrow\downarrow} = \frac{n_{\uparrow\downarrow}^2 \cos \theta_{\uparrow\downarrow}}{n_0 \cos \theta_0} F_{\uparrow\downarrow} \left\{ \left| A_{\uparrow\downarrow} \right|^2 + R_{\uparrow\downarrow} \left| A_{\downarrow\uparrow} \right|^2 \right\} N + 2A_{\downarrow} A_{\uparrow} \operatorname{Re} \left[r_{\uparrow\downarrow} \exp(-ikn_0 d \cos \theta_0) \int \mu(\mathbf{r}) \exp(\mp i2kn_0 z \cos \theta_0) d^3 r \right], \quad (5)$$

where

$$N = \int \mu(\mathbf{r}) d^3 r \quad (6)$$

is the total number of dipoles in the distribution. For volume formed by randomly-aligned dipoles, Eq. (5) has to be specialized to the three possible alignment and polarization states and then evaluated through a suitable linear combination [13, 14, 16]. The integral in Eq. (5) is a Fourier transform that can be analytically evaluated in some cases of interest. One of such cases is that of a cylinder distribution of height h (along z) and diameter D , centred around $z = z_0$,

$$\mu(\mathbf{r}) = \begin{cases} \frac{4N}{\pi D^2 h} & \text{if } \sqrt{x^2 + y^2} \leq D/2 \text{ and } |z - z_0| \leq h/2 \\ 0 & \text{otherwise.} \end{cases} \quad (7)$$

This distribution is fully contained within the central layer of thickness d provided that $|z_0| \leq (d - h)/2$. As one can verify, the integral appearing in Eq. (5) can be analytically evaluated in this case and is equal to

$$\begin{aligned} & \int \mu(\mathbf{r}) \exp(\mp i2kn_0 z \cos \theta_0) d^3 r = \\ & = N \operatorname{sinc} \left(2 \frac{n_0 h}{\lambda} \cos \theta_0 \right) \exp(\mp i2kn_0 z_0 \cos \theta_0), \end{aligned} \quad (8)$$

where $\lambda = 2\pi/k$ being the wavelength in vacuum and the sinc function being defined as

$$\operatorname{sinc}(z) = \frac{\sin(\pi z)}{\pi z}. \quad (9)$$

It should be noted how the radiated intensity does not depend on D . This fact is strictly related to the invariance of far-field emission from a single dipole for translation over the Oxy plane and to the assumed lack of cooperation among the distribution elements [16]. It also ensues that the found results keep true for any transversal section of the cylinder, here assumed to be circular for convenience.

Case study: Alq₃-based organic light-emitting diode

In principle, the discussed volume-source model can be applied to any multilayer structure containing active layers. The analytical formulas make the theory convenient to study how the optical properties of such devices depend on physical parameters such as layer thicknesses and optical constants. The device to which our model is applied in this paper is an OLED based on an Alq₃ electroluminescent layer (Fig. 2). The Alq₃ layer is sandwiched between a TPD organic film – grown on an ITO anode deposited over a SiO₂ substrate – and a cathode that is either a single layer of Al (Fig. 2a) or a LiF/Al bi-layer (Fig. 2b). Electroluminescence from the Alq₃ layer is detected at the substrate side of the device. Here, we analyze from an optical point of view the convenience of using the LiF/Al cathode in place of the Al one, a configuration that proved to be useful in lowering the threshold voltage in Alq₃-

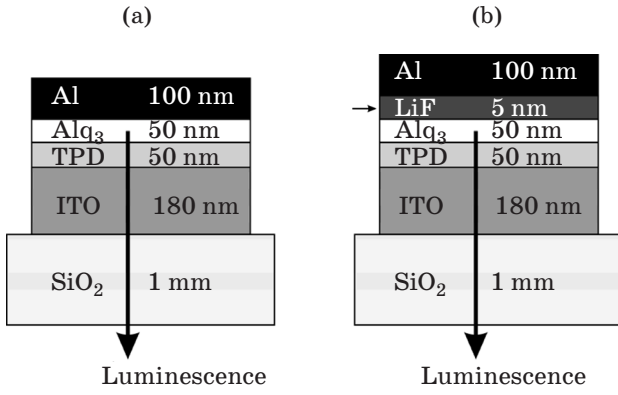


Fig. 2. OLED based on an active Alq_3 layer. Design with an Al cathode (a) and a LiF/Al cathode (b).

based OLEDs [19, 20]. Moreover, we also check if the thickness of some layers of the device can be optimized to improve the electroluminescence intensity.

Effect of a LiF/Al cathode

As already mentioned, a thin LiF layer placed between the active layer and the Al cathode has proved to improve the electrical performances of an Alq_3 -based OLED [19, 20]. Our purpose is to check how the insertion of the LiF layer modifies the electroluminescence intensity at the representative emission wavelength $\lambda = 530$ nm. The complex refractive indices utilized for the simu-

lations are: $0.867 - i 6.42$ for Al [21], 1.70 for Alq_3 [22], $2.00 - i 0.0085$ for ITO [23], 1.393 for LiF [24], 1.547 for SiO_2 [21], and 1.85 for TPD [22]. By applying Eqs. (5–9), one can evaluate the power W_{\downarrow} that is outcoupled downwards (i. e. to the substrate side) vs. the propagation angle, both for a simple Al and a LiF/Al cathode. The obtained results are plotted in Fig. 3. It should be noticed how the introduction of a LiF layer as thin as 5 nm increases the outcoupled power by about 21% along the normal direction and 14% over the whole solid angle (that is, for a unitary numerical aperture). Such a performance increase is due to the phase change introduced by the LiF layer rather than to reflectivity changes [16].

It is worthwhile considering also the situation where, instead of the whole Alq_3 layer, only a portion of it, thin 10 nm and close to the TPD layer, is responsible for the generation of electroluminescence [25]. Our theoretical model can easily deal with such a configuration by setting $h = 10$ nm and $z_0 = -(d - h)/2 = -20$ nm in Eqs. (5) and (8). The resulting angular distributions of outcoupled power are plotted in Fig. 4; note how they are similar to those shown in Fig. 3. Quantitatively speaking, in the case of Fig. 4 the introduction of the LiF/Al cathode increases the outcoupled power by about 17% along the normal direction and 11% over the whole solid angle, that is, just a few percent less than the whole active Alq_3 layer case reported in Fig. 3.

The emission spectrum of Alq_3 ranges about from 400 nm to 800 nm and is peaked at about 530 nm [20, 26]. For space reason, we can only

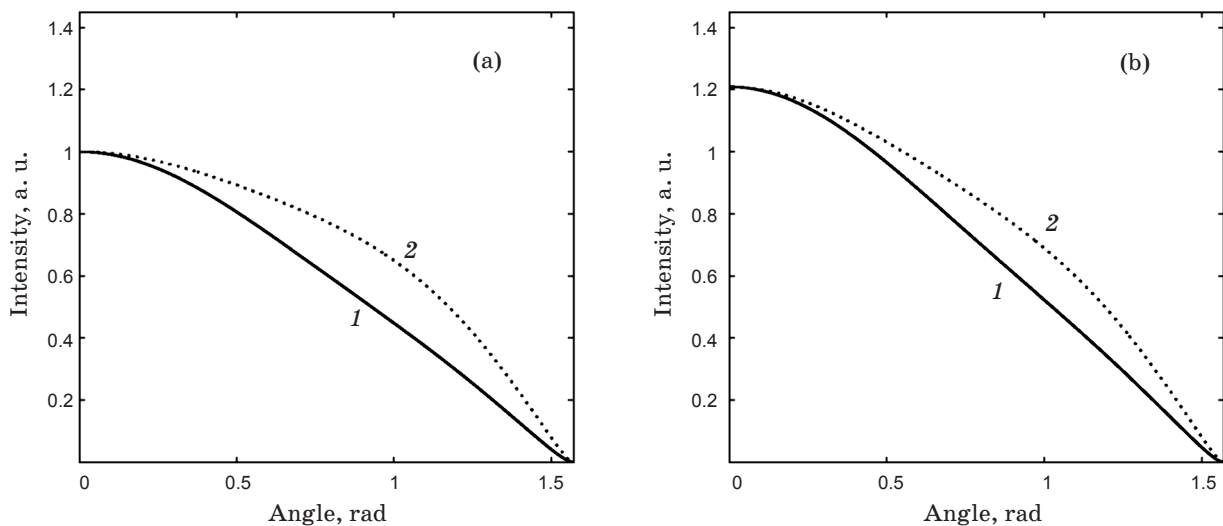


Fig. 3. Theoretical intensity at $\lambda = 530$ nm radiated by the OLED vs. the propagation angle θ_{\downarrow} in the bottom external medium for an Al cathode (a) and a LiF/Al cathode (b). The two curves in each plot correspond to the two polarization states s (1) and p (2). The entire Alq_3 layer is assumed to be optically active.

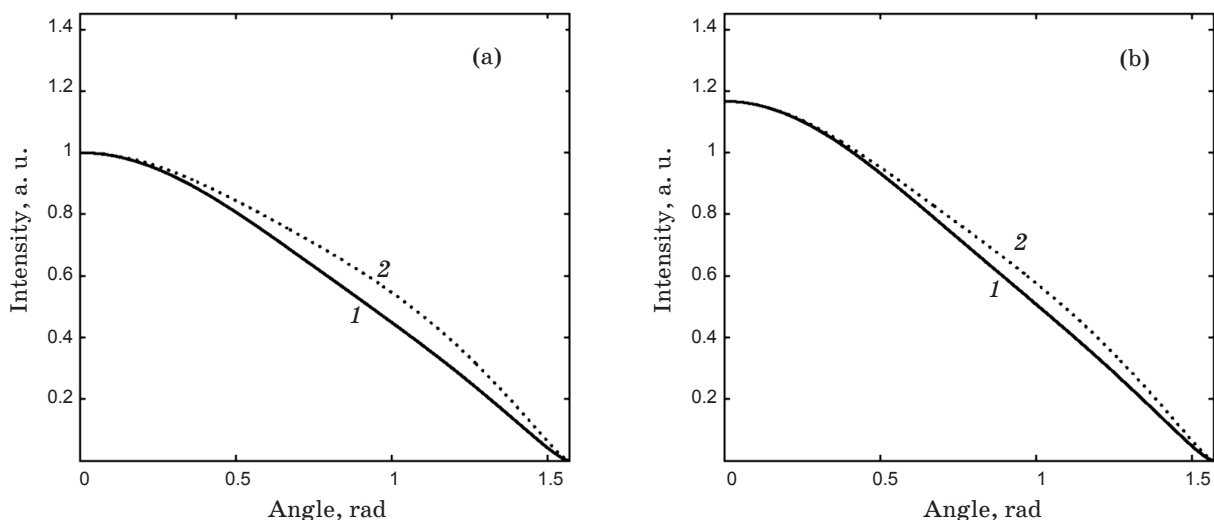


Fig. 4. Theoretical intensity at $\lambda = 530$ nm radiated by the OLED vs. the propagation angle θ_{\downarrow} in the bottom external medium for an Al cathode (a) and a LiF/Al cathode (b). The two curves in each plot correspond to the two polarization states s (1) and p (2). Only a portion of the Alq_3 layer, thin 10 nm and close to the TPD layer, is assumed to be optically active.

mention that the use of a LiF/Al cathode in place of a simple Al one increases the outcoupled electroluminescence intensity rather uniformly over the emission spectrum by an amount equal to that found at the wavelength $\lambda = 530$ nm. Moreover, both for the Al and LiF/Al cathode cases, the reflection and phase properties of the layered structure surrounding the Alq_3 layer cause a small blue shift, quantifiable as about 10–15 nm, of the emission spectrum peak of Alq_3 .

Layer thickness influence on Alq_3 -based OLED performances

Fig. 2 depicts an Alq_3 -based OLED with typical design thickness values for the layers [20]. One could wonder if it is possible to obtain better performances of the device, in terms of outcoupled intensity, by changing some thickness values, for instance those of the Alq_3 and TPD layers. Such a parametric study can be performed quite effortlessly thanks to the analytical formulas of our volume-source model. To that purpose, we consider the more performing design of Fig. 2b, where an ultrathin LiF layer is placed in contact to the Al cathode. Analyzing, by means of Eqs. (5–9), light emission along the normal direction $\theta_{\downarrow} = 0$ at the wavelength $\lambda = 530$ nm, for the two cases of either a fully active Alq_3 layer or a 10 nm active portion of it close to the TPD layer [25], the contour maps shown in Figs. 5a and b are obtained, respectively.

For a fully active Alq_3 layer (see Fig. 5a) the largest outcoupled intensity is found within the examined thickness domain when the thicknesses of Alq_3 and TPD are set to (200 ± 2) nm and (100 ± 2) nm, respectively. For those thicknesses, the calculated intensity is about 5.6 times larger than that extracted from the design in Fig. 2b. This value is obviously influenced by a 4 times thicker Alq_3 layer (200 nm vs. 50 nm) and hence a 4 times larger number of Alq_3 radiating molecules. Disregarding the higher number of molecules, the efficiency increase for a single delocalized molecule can be evaluated as $5.6/4 = 1.4$, that is, 40% better than the design in Fig. 2b.

From a practical viewpoint, the thickness ranges that are considered in the simulation of Fig. 5a are too wide and should be restricted to about [20, 70] nm for both the layers [27]. Limiting the analysis domain in Fig. 5a within such range, the maximum intensity is then found in correspondence of a thickness of (70 ± 2) nm for both the Alq_3 and TPD layers and is about 2.54 times larger than the design value. Again, this increase is partly due to the larger number of Alq_3 radiating molecules within the thicker layer; the efficiency increase for a single delocalized molecule amounts to $2.54/(70/50) = 1.81$, which is 81% better than the design one.

One can conduct the same analysis as above for the case of a 10 nm thin active portion of the Alq_3 layer close to the TPD layer [25]. The simu-

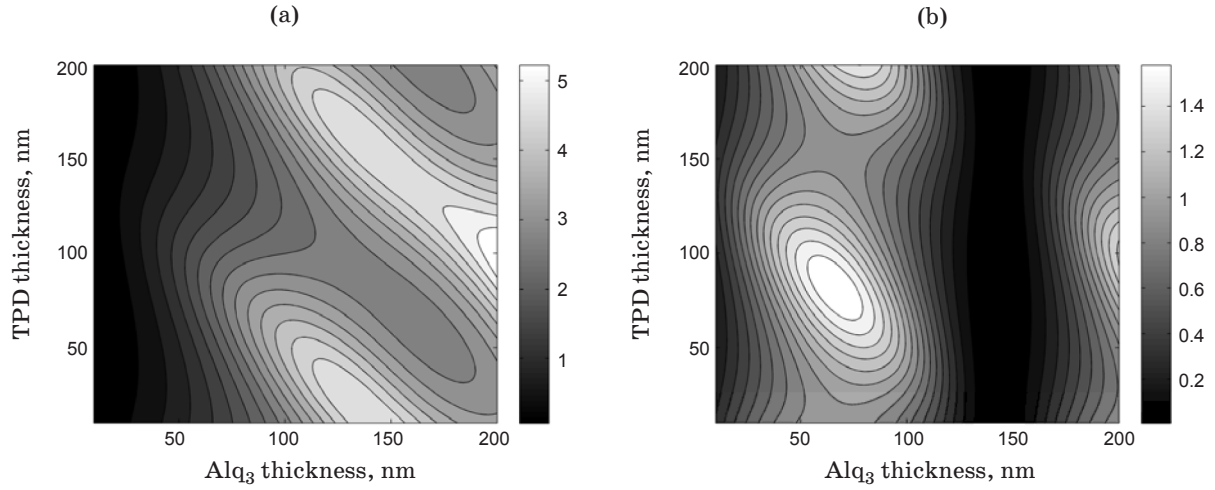


Fig. 5. Contour maps of the theoretical intensity at $\lambda = 530$ nm radiated by the OLED with LiF/Al cathode along the normal direction $\theta_{\downarrow} = 0$ in the bottom external medium for varying thicknesses of the TPD and Alq₃ layers. The units are such that intensity is unitary when the thicknesses of TPD and Alq₃ are both 50 nm, as in the device shown in Fig. 2. In the simulations, the whole Alq₃ layer (a) or a portion of it (b), thin 10 nm and close to the TPD layer, is assumed to be optically active.

lation results for this case are shown in Fig. 5b. Note how they differ from the results shown in Fig. 5a. As a matter of fact, in Fig. 5b the maximum outcoupled intensity is found for the thicknesses of (66 ± 2) nm and (82 ± 2) nm for the Alq₃ and TPD layers, respectively, and the intensity increase amounts to about 68%. Restricting the analysis, as previously done, to the thickness range of [20, 70] nm for both the layers [27], here too the thickness giving the optimal outcoupled intensity is equal to (70 ± 2) nm for both Alq₃ and TPD, which corresponds to an increase of about 63% with respect to the design.

It can be concluded that by increasing the thickness of Alq₃ and TPD from their typical design value of 50 nm to about 70 nm should significantly improve the performances of the Alq₃-based OLED of Fig. 2, at least as far as the device efficiency in outcoupling photons generated in the Alq₃ layer is concerned. A study of the consequences of such thickness changes from an electrical point of view is outside the purposes of this paper.

Conclusions

A generalization to volume sources of a theoretical model for light emission from inside a layered medium allows evaluating the outcoupled intensity by using analytical expressions in many cases of interest. This finding can be exploited for the quick evaluation of the performances of

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optically-active layered devices, such as light-emitting diodes.

As an application of the model, an Alq₃-based OLED has been theoretically analyzed. First, the effect of an ultrathin LiF layer placed at the device cathode has been investigated from an optical point of view, demonstrating that it increments the intensity of electroluminescence by about 15–20%. Second, starting from a consolidated design of the device, the possibility of changing the thickness of some layers has been investigated leading to the conclusion that, from a bare optical point of view, the device would outcouple light about 40–80% more efficiently if the thickness of the Alq₃ and TPD layers were increased from their typical design value of 50 nm to the calculated optimal value of 70 nm.

Thanks to its analyticity, the developed model has demonstrated to be useful for the quick analytical evaluation of the light intensity radiated from a layered device containing an optically active layer. Such evaluations would have otherwise required the application of lengthy and time-consuming numerical codes. Possible developments of the model include its generalization to non-planar structures.

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