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Abstract. We focus on the determination of the internal luminescence quantum efficiency of a green-emitting organic light-emitting diode (OLED). By considering different geometrical configurations of OLED thin-film stacks, we elucidate the role of the internal luminescence quantum efficiency of the emitter in the thin-film microcavity. Combining optical simulations with experimental results, a comprehensive efficiency analysis is performed. Here the electroluminescence of a set of OLEDs is characterized. Additionally, the devices are characterized using time-resolved photoluminescence measurements. The experimental data are analyzed using optical simulations. This analysis leads to a quantification of internal luminescence quantum efficiency and allows conclusions about competing mechanisms resulting in nonradiative recombination of charge carriers. © 2011 Society of Photo-Optical Instrumentation Engineers (SPIE). [DOI: [10.1117/1.3528274](https://doi.org/10.1117/1.3528274)]

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1 Introduction

Today's displays based on organic light-emitting diodes (OLEDs) are integrated in various applications such as cell phones, MP3 players, or digital cameras. At the moment, the OLED technology is on the verge of creating commercial applications in the lighting sector. The remarkable advantages of OLEDs will drive innovative products and open up new fields of application: They are thin, flat, and lightweight. Furthermore, the technology allows for the production of large-area lighting panels in a cheap and simple process. One evident drawback of state-of-the-art OLEDs is the fact that only a small amount of the internally generated light is finally emitted to the outside medium air. The mismatch of the refractive index between air and the organic layers ($n \approx 1.8$) leads to most of the generated light being lost by total internal reflection into wave-guided modes and self-absorption. As a consequence, only $\sim 20\%$ of the generated light is commonly extracted from the device.¹ Light extraction can be enhanced by modifying the substrate surface (i.e., the light-emitting surface of the device), disturbing the total internal reflection at the substrate–air interface. By this means, almost 50% of the generated light can be extracted by well-known substrate surface modifications, such as microlens arrays² or diffusers.³ However, a mirrorlike off-state appearance of the OLED device is desired for certain fields of application. In this case, substrate surface modification, usually leading to a milky

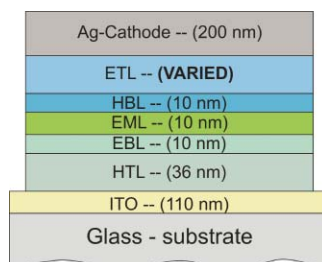


Fig. 1 Structure of the green-emitting electrophosphorescent OLEDs used in this study. The ETL thickness was varied in the range between 34 and 284 nm.

off-state appearance, has to be avoided. The optimum device configurations for OLEDs with and without outcoupling enhancement differ from each other.¹ However, for both configurations the internal luminescence quantum efficiency q —describing the radiative decay probability of an excited emitter molecule and thus the conversion of electrical into optical power—is of crucial importance.

The external quantum efficiency (EQE) η_{EQE} is defined as⁴

$$\eta_{\text{EQE}} = \gamma \eta_{\text{S/T}} q_{\text{eff}}(q) \eta_{\text{out}}, \quad (1)$$

with γ being the fraction of injected charge carriers that form excitons (charge balance factor). The singlet-triplet factor $\eta_{\text{S/T}}$ takes into account the material system specific fraction of excitons with transitions into the ground state being allowed by selection rules. (For phosphorescent emitters used in this study, this factor can be assumed to be unity).⁵ η_{out} is the classical outcoupling efficiency of the device. q_{eff} denotes the effective internal luminescence quantum efficiency of the emitter material. It differs from the internal luminescence quantum efficiency q , (i.e., the quantum efficiency of the emitter material in an unbounded space filled by the light-emitting dye and its host material) because of modifications due to the presence of reflecting interfaces in an OLED structure. The optimum emitter position in the optical microcavity formed by the OLED thin-film stack depends on the value of q ,^{6–9} as will be discussed further later. In the present paper, methods for the determination of the internal luminescence quantum efficiency q from photoluminescence (PL) experiments (giving q_{PL}) and from the electroluminescence (EL) measurements (yielding q_{EL}) are presented. Furthermore, we will show that these two quantities are significantly less than unity and that q_{EL} is further reduced with increasing current density j . Finally, the impact of the q value on the optimum device design is discussed. The efficiency analysis is applied on a set of green-emitting phosphorescent OLEDs with systematically varied thickness of the electron transport layer.

2 Methods

For this study, a series of green bottom-emitting devices with an active area of 4 mm² was fabricated. The structure of the devices was glass-substrate/110 nm indium tin oxide (ITO)/36-nm hole-transporting layer (HTL)/10-nm electron-blocking layer (EBL)/10-nm green-emitting layer (EML) consisting of an electron conductive host and a green phosphorescent dye [fac-tris(2-phenyl-pyridyl)iridium (Ir(ppy)₃)]/10 nm HBL/electron-transporting layer (ETL)/200 nm Ag-cathode. The thickness of the ETL was varied in the range from 34 to 284 nm. The organic layers were deposited onto an ITO-coated glass substrate using standard evaporation techniques. For better electron and hole transport, doped ETL and HTL were used, where the dopant was applied by coevaporation. This technique was also used for depositing the green dye inside the host. The evaporation rate was 0.5 Å/s at a base pressure of 10^{−7} mbar. After evaporating the Ag-cathode, all devices were encapsulated with a glass lid and getter. A scheme of the stack is shown in Fig. 1.

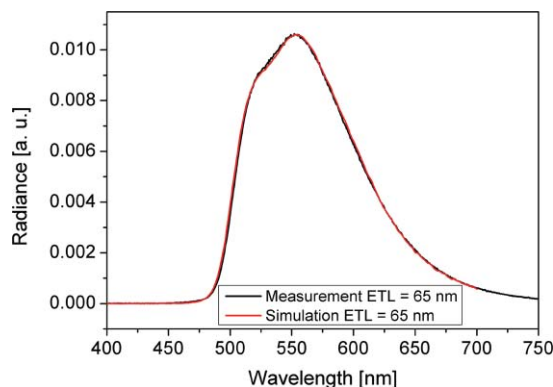


Fig. 2 Measured (black) and simulated (red) 0-deg emission spectrum of the device with an ETL thickness of 65 nm showing the good agreement between measurement and simulation.

In order to provide reliable data for this study, the layer thicknesses of the diodes were checked using optical simulations. The complete series was produced on one substrate using a common mask for all the layers except the ETL. For the deposition of the ETL, different shadow masks were used to fabricate different ETL thickness only on several devices on the substrate. With this method, the thickness of the whole stack needs to be checked only once and, for all further devices, only the ETL needs to be changed in the simulations. For each diode, the spectrum of the emission in the 0-deg direction (i.e., the direction normal to the OLED substrate) and the spectra at different angles were simulated and the layer thicknesses were varied to fit the measured spectra. Figure 2 depicts the measured EL spectrum at an angle of 0 deg for a device with an ETL thickness of 65 nm. In order to enable for accurate simulation, the complex refractive indices as a function of wavelength were determined by means of reflection-transmission spectroscopy of single supported films for all materials used.¹⁰ The current density–voltage (j – V) characteristic of the OLEDs was also characterized, showing no significant difference between the devices with varying ETL thicknesses. This can be attributed to appropriate n-doping,⁹ yielding the possibility to neglect influences such as carrier injection or transport properties. Thus, only optical effects need to be considered in this study.

The setup for the time-resolved PL measurements was based on a Nd:YAG Laser (Ekspla PL2210) with a wavelength of 355 nm. The samples were excited under an angle of 20 deg to the surface normal. The pulse length was 30 ps with energy of 10 μ J per pulse. The measurement was performed with a repetition rate of 50 Hz. The time-dependent PL decay was recorded normal to the surface with a streak-camera system (Hamamatsu C5680).

The EL measurements were performed in an integrating sphere, which was connected to a source-measurement unit (Keithley 2400) and a fiber spectrometer (Instrument Systems CAS 140). Prior to the measurement a calibration with a halogen lamp was performed with and without the device in order to correct for OLED self-absorption. The substrate edges were blackened in order to avoid an influence from side emission. The Keithley 2400 and the CAS 140 spectrometer were also used for the measurement of the emission spectra.

3 Theory

Near-field phenomena and the photonic mode density must be considered when computing the optical behavior of emissive sites inside the thin-film structure formed by the layers of an OLED. The algorithm used to perform the optical calculations in this study is based on an early work of Sommerfeld,¹¹ who investigated the propagation of radiowaves due to dipole antennas above the Earth's surface by means of a Hertz-Vector representation. Later, this model was adapted by Chance *et al.*¹² [Chance, Prock, and Silbey (CPS)] for molecular fluorescence and energy transfer near interfaces. This rigorous approach uses dyadic Green's functions to describe effects

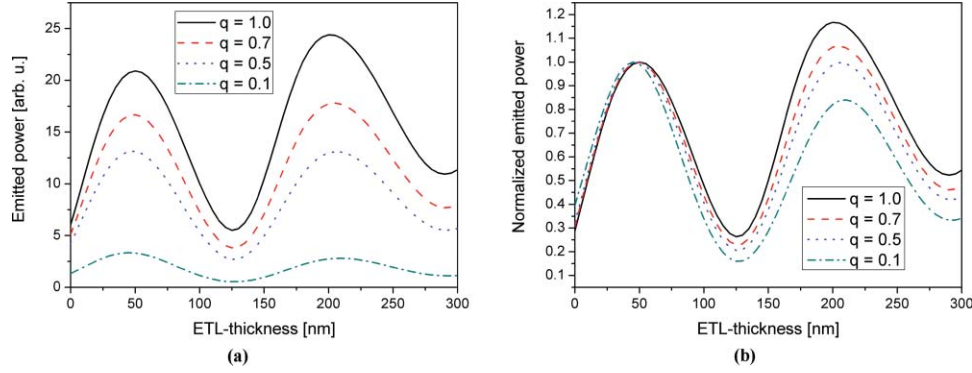


Fig. 3 Plot of the integral emission into air as a function of the ETL thickness for different assumptions of the internal luminescence quantum efficiency: (a) absolute values and (b) normalized to the value of the first maximum.

classically. Danz *et al.*¹³ extended the CPS model to the general case of arbitrary multilayer systems. According to CPS,¹² the electric field $\vec{E}(\vec{r}) \exp(-i\omega t)$ due a harmonic oscillating current distribution $\vec{j}(\vec{r}) \exp(-i\omega t)$ in a thin-film system can be calculated by the use of the following Green's function $G = G_0 + s$:

$$\vec{E}(\vec{r}) = i\omega\mu_0 \int G(\vec{r}, \vec{r}') \vec{j}(\vec{r}') d^3\vec{r}', \quad (2)$$

where μ_0 is the free-space permeability, ω is the angular light frequency, and t , \vec{r} denote time and space coordinates. Here, the total Green's function is decomposed into the Green's function G_0 of the homogeneous emitter medium and a scattering contribution G_s due to the multilayer system. The decay rate b (inverse lifetime) of a dipole emitter at the position \vec{r}_0 with a dipole moment \vec{p} inside a cavity can be derived from the rate of energy dissipation and is calculated using

$$\frac{b}{b_0} = 1 + q \frac{\text{Im}[\vec{p} G_s(r_0, r_0) \vec{p}]}{\text{Im}[\vec{p} G_0(r_0, r_0) \vec{p}]}, \quad (3)$$

where q is the internal luminescence quantum efficiency and b_0 is the decay rate of the infinite emitter medium. A more detailed analysis is given by Danz *et al.*,¹³ and the references therein. In result, the relative change in transition rate of the dipole normalized to the spontaneous emission rate in the infinite emissive medium without boundaries can be calculated. The geometric configuration of the microcavity alters the radiative rate and the spatial distribution of the emitted radiation.⁹ The emitter cathode distance mainly determines the interference conditions at the position of the emissive sites. Consequently, the external output of an OLED structure (i.e., the light that is not total internally reflected but directed into the escape cone) severely depends on the thickness of the ETL defining the distance of the emission layer to the cathode mirror. The model outlined above allows calculating the extracted power as a function of ETL thickness by integrating the angular emission into the ambient over the entire half sphere.

Our method for the determination of q_{EL} is based on the computation of the fraction of light extracted from the device as a function of ETL thickness. Figure 3 depicts the calculated emitted power of the green OLED series as a function of ETL thickness for different values of q assuming a constant emission zone and isotropic emitter orientation distribution. These assumptions were experimentally verified by utilizing approaches presented elsewhere.¹⁴ Figure 3(a) shows the absolute emitted power calculated for four different q values. The emitted power strongly depends on the distance to the cathode mirror leading to two maxima at ETL thicknesses of about 60 and 200 nm for this green OLED series.

In Fig. 3(b), the calculated emitted power for the different q values were normalized to their first maximum. It can be seen that the value of q alters the shape of the power plot as a function of ETL thickness and is not acting as a multiplicative factor only [see Eq. (1)]. For high q values (i.e., $q > 0.5$), the second maximum is higher than the first maximum, which has been observed before.¹⁵ On the contrary, for lower q values, the first maximum is higher. Consequently, the q value corresponding to the specific emitter system must be considered when designing OLEDs with optimum light output. The method for the determination of q_{PL} uses the phosphorescence lifetime measured on the same OLED structures under PL excitation, as will be discussed in detail in Sec. 4.

4 Results and Discussion

4.1 Photoluminescence

Time-resolved measurements of the PL intensity allow for the determination of the phosphorescence lifetime τ_{PL} . Figure 4 shows an example of the measured decay time for the device with an ETL thickness of 34 nm, including a monoexponential fit to the long-living tail of the decay signal. The initial deviation from the monoexponential decay can be attributed to bimolecular annihilation processes.¹⁶ Here, the PL lifetime τ_{PL} is fitted to a value of $\tau_{\text{PL}} = 0.47 \mu\text{s}$. The fitting was performed for all measured devices, and the results are plotted in Fig. 4(b). In the following, we describe how q_{PL} (extracted from PL) can be quantified based on the measurement of the lifetime τ_{PL} . The measured lifetime is a superposition of the radiative (R) and nonradiative (NR) lifetimes,

$$\frac{1}{\tau_{\text{PL}}} = b_{\text{PL}} = b_{\text{R}} + b_{\text{NR}}. \quad (4)$$

The radiative lifetime depends not only on the internal luminescence quantum efficiency q of the emitter but also on the geometric configuration of the microcavity.¹⁷ By contrast, the nonradiative lifetime is not affected by the optical environment, as assumed for deriving the rate (inverse lifetime) relation given in Eq. (3). Recalling the definition of q ,

$$q = \frac{b_{\text{R}}}{b_{\text{R}} + b_{\text{NR}}}. \quad (5)$$

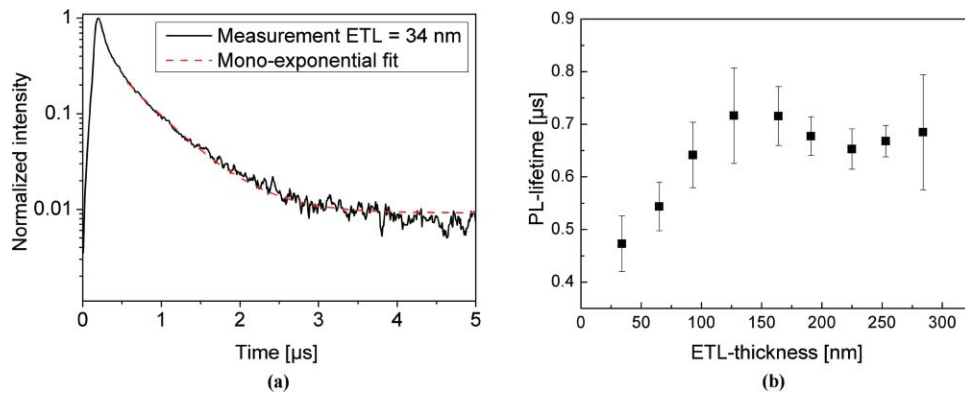


Fig. 4 (a) Measured (black) phosphorescence intensity after short-pulse PL excitation for the device with an ETL thickness of 34 nm. Best monoexponential fit for the long-living tail of the decay signal (red) yields a PL lifetime of $0.47 \mu\text{s}$. (b) Fitted phosphorescence lifetimes for all devices under study as a function of ETL-thickness. The huge error bars at 126 and 284 nm ETL thickness originate from the weak PL signals due to emitter position in outcoupling minima to air (see Fig. 7).

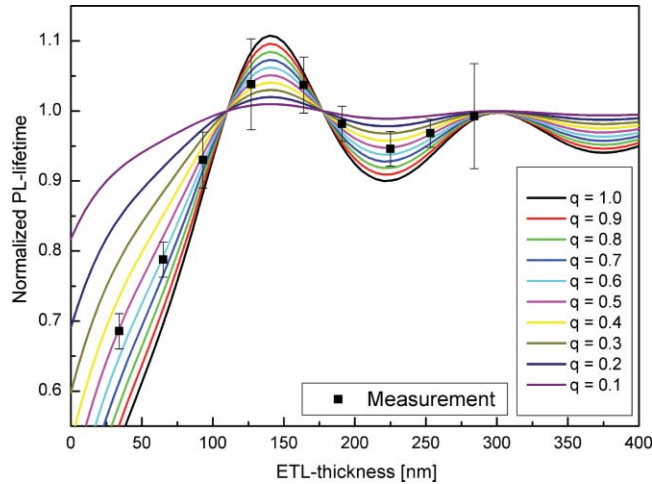


Fig. 5 Black squares: measured PL lifetime as a function of ETL thickness. Solid lines: simulated change in PL lifetime as a function of ETL thickness. All values are normalized to the free-space PL lifetime $\tau_0 = 0.69 \mu\text{s}$.

points out that changes of the radiative lifetime correspond to changes in the resulting q value.

In Fig. 5, the resulting normalized PL lifetimes τ_{PL} obtained from these calculations are plotted as a function of ETL thickness for the different q values from 0.1 to 1 (solid lines). According to our simulations, the free-space lifetime yields $\tau_0 = 0.69 \mu\text{s}$. This allows fitting Eq. (3) to the measurement data. The measured values of the phosphorescence lifetime τ_{PL} (black squares) have also been normalized on τ_0 . The data are plotted in Fig. 5.

It can be clearly seen that the change of the PL lifetime depends on both the thickness of the ETL and the value of the internal luminescence quantum efficiency q , especially close to the mirror. For very thin ETL layers, the internal luminescence quantum efficiency strongly affects the lifetime because of strong coupling into surface plasmon polaritons⁸ close to the metal cathode. The comparison of measurement and simulation leads to a value of $q_{\text{PL}} = 0.5 \pm 0.1$. Hence, only half of the excitons generated by optical excitation are transferred into radiation, while the other half is lost by nonradiative transitions.

4.2 Electroluminescence

The determination of the internal luminescence quantum efficiency q_{EL} under electrical operation is also based on a comparison of measured values with simulation results. Here the measurements are performed in an integrating sphere operating the OLEDs under different current densities. The measurements were performed in two modes: First, the OLEDs are measured without an outcoupling structure and, in a second measurement, with a half-ball lens optically coupled to the substrate surface. The lens has a 25-mm diameter and a refractive index of $n = 1.52$, matching the substrate refractive index exactly. Thus, the light that is trapped inside the substrate can be coupled out when applying the half-ball lens atop the device. The advantage of these two measurements is a good control, because both measurements have to yield to consistent results for q_{EL} . In Fig. 6, the optical output power obtained from measurements with the half-ball lens at a current density of $j = 1.0 \text{ mA/cm}^2$ is plotted (right axis). Additionally, the corresponding results from the simulation of the light output into the substrate are given in the same graph. In the simulations, q has been varied between 0.2 and 1.0, allowing the extraction of q_{EL} by comparing the measurement to the simulation. Both experimental and simulation data have

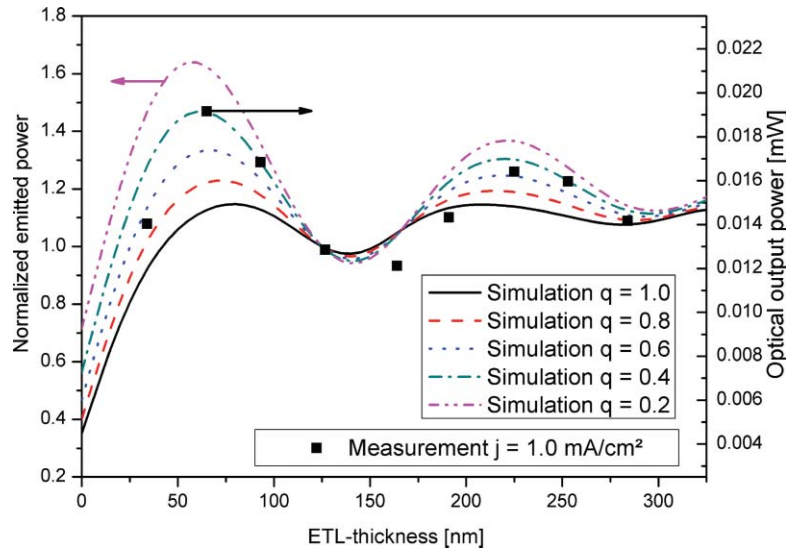


Fig. 6 Black squares: integrating sphere measurements of total emitted power with optically coupled half-ball lens. Lines: simulated emitted power as a function of ETL thickness for different values of the internal luminescence quantum efficiency q . The simulation data are normalized to an ETL thickness of 126 nm.

been normalized to the light output of the device with 126-nm ETL thickness (right axis). This representation of the data allows comparing the curve progression of both data sets.

It can be seen that the results from the measurement follow the predicted variation in emitted power obtained from the simulations. Also in the case of electrical driven measurements, the curve progression of the power plot as a function of ETL thickness depends on the value of q . According to Fig. 6, the simulation assuming $q_{EL} = 0.4$ reproduces the experimental data well. Moreover, the assumption of an internal luminescence efficiency of $q = 1$, often associated with phosphorescent emissive materials, will lead to wrong OLED efficiency predictions.

Furthermore, the dependency of q_{EL} on the current density can be investigated by performing these measurements at different current densities. In our devices, appropriate blocking materials are used and a charge balance factor of $\gamma = 1$ can be assumed [see Eq. (1)]. When the measured quantum efficiency is brought into correlation with optical power data (obtained from the simulation), one should keep in mind that the outcoupled spectra depend on the ETL thickness, leading to ETL-dependent nonlinearity between EQE and power efficiency. By analyzing our measurement data, we have found a deviation of $<4\%$ between the values of the EQE and the corresponding values of power contribution of the green-emitting devices.

In Fig. 7, the results for measurements at current densities from 0.1 to 1000 mA/cm² with and without the half-ball lens as a function of ETL thickness are shown and compared to the simulation results.

The decrease of the EQE with increasing current density can clearly be seen. In Fig. 7(a), the measurements and simulations with the half-ball lens are shown. In this case, the first maximum is always higher than the second. Consequently, the emissive sites should be placed in the first maximum when an outcoupling structure is used. This is in contrast to the measurements shown in Fig. 7(b). It can be seen that for $q = 0.6$ (and higher values, not shown in the graph), the second maximum is higher than the first one. Thus, if the efficiency of the emitter could be more than $q = 0.5$, then the emitter should be placed in the second maximum. In both cases, the comparison of the measurements to the simulation leads to the same value of the current dependent internal luminescence quantum efficiency. From these measurements, the current dependency of q_{EL} can be extracted and is depicted in Fig. 8.

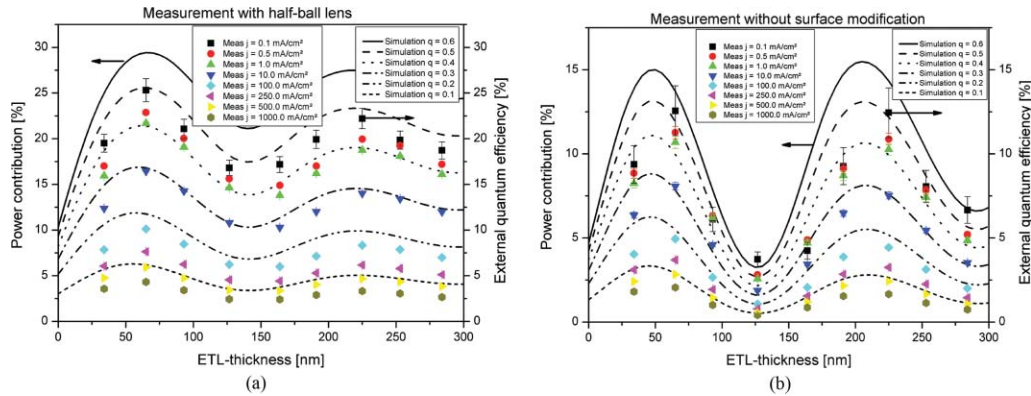


Fig. 7 Measured EQE (dots) and simulated (lines) power contribution as a function of the ETL thickness for the measurement in an integrating sphere (a) with and (b) without the optically coupled half-ball lens.

From the PL measurements, a value of $q_{PL} = 0.5 \pm 0.1$ was determined. For the electrical driven devices, this value is only reached for very low current densities. With increasing current density, the difference between q_{EL} and q_{PL} increases. One reason for this difference is the missing current flow during the PL measurements. Because the host of the dye is predominantly electron conductive, the generated excitons will mainly be located at the interface between the emitter and the EBL.¹⁸ This leads to a relatively narrow emission zone. In PL measurements, excitons are generated in the whole emitting layer and can recombine in the whole layer. This leads to lower exciton densities associated with weaker exciton quenching.¹⁹ With increasing current density, the difference between q_{PL} and q_{EL} increases strongly. This is expected because with increasing current density the rate of competing nonradiative processes, such as triplet-triplet or triplet-polaron, annihilation increases.^{20,21} Especially at very high current density, the effect of Joule heating leads to significant singlet-heat annihilation and exciton dissociation that can no longer be neglected.²² The knowledge of the current dependency of q_{EL} is a very important fact for the design of the devices. The assumption of 100% internal luminescence quantum efficiency will lead to wrong simulation predictions, and also, the distance to the cathode has to be adapted to the desired current density. The internal luminescence quantum efficiency of the green emitter considered in the present study is about $q = 0.5$ for lower current

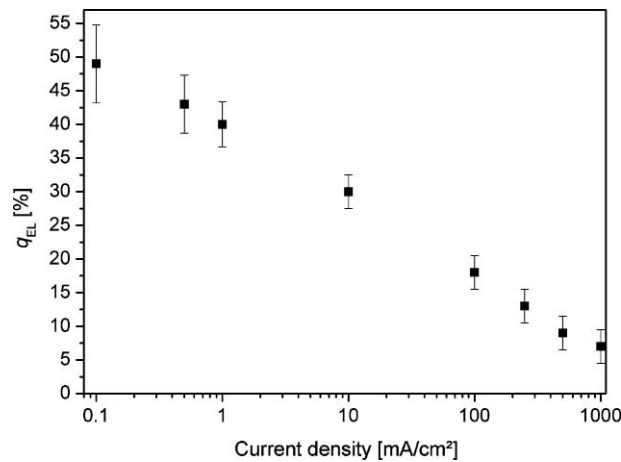


Fig. 8 Determined internal luminescence quantum efficiency of the green emitter as a function of current density.

densities. According to the simulation results (Fig. 3) and to our measurements of the devices with 65 and 225 nm ETL at low current densities, these two ETL thickness values will lead to devices of the same efficiency: From the plots in Figs. 3 and 7, it can be seen that for a q value of 0.5, the first and second maximum are at the same height. However, increasing the current densities the first maximum will lead to a higher efficiency because q_{EL} is reduced to <0.5 due to triplet-triplet or triplet-polaron annihilation. On the other hand, for an emitter with $q > 0.5$, it will be more favorable to place it in the second maximum—provided that q_{EL} is not significantly reduced by competing nonradiative processes. This is especially important for the design of high-brightness OLEDs, which are driven at higher current densities, resulting in a lower internal luminescence quantum efficiency under operation.

5 Conclusion

In conclusion, we have discussed two different methods to determine the internal luminescence quantum efficiency q of OLED emissive materials. The PL-measurement-based method allows one to get a rough estimation of the q value without the need to fabricate complete OLEDs. This is important for a rapid characterization of new emitter materials. The EL-based method enables for determining the internal luminescence quantum efficiency at different current densities. This is of great importance if OLEDs are designed for significantly higher or lower brightness levels. Consequently, the assumption of efficiency close to unity often used can lead to wrong efficiency predictions and layer thicknesses. At least a rough quantification of q from the PL-based measurements should be performed before designing OLEDs. In this study, the internal luminescence quantum efficiency of the emitter used is only half the value of the theoretical maximum of $q = 1.0$. Thus, the determination of the internal luminescence quantum efficiency gives the material and device developers feedback about the potential for further improvement of the emitter systems.

Acknowledgments

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