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<th>Modifications of the exciton lifetime and internal quantum efficiency for organic light-emitting devices with a weak/strong microcavity</th>
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Organic light-emitting devices (OLEDs) with various device configurations have been the subject of intensive research due to their applications in display and lighting.\textsuperscript{1–4} Considering the microcavity effect, OLEDs can be roughly categorized into two types, i.e., weak microcavity OLEDs and strong microcavity OLEDs. Conventional bottom-emitting OLEDs are weak microcavity devices, while OLEDs with distributed Bragg reflectors or two metallic electrodes are considered as strong microcavity devices. Light emission properties, including the internal quantum efficiency ($\eta_{\text{int}}$), external quantum efficiency, exciton lifetime, and angular dependence, are distinct in the two types of OLEDs due to the Purcell effect.\textsuperscript{5–8}

Exciton lifetimes of emitters in planar dielectric microcavity structures,\textsuperscript{9} near a partially reflecting surface\textsuperscript{10,11} in weak microcavity OLEDs,\textsuperscript{12,13} and above metallic gratings\textsuperscript{14} have been investigated either theoretically or experimentally. In this letter, a comprehensive analysis is given on the modifications of exciton lifetime for OLEDs with various device structures. A linear relation between the exciton lifetime and $\eta_{\text{int}}$ will be derived, which means that $\eta_{\text{int}}$ can be obtained indirectly through the measurement of the exciton lifetime. In addition, we investigate both experimentally and theoretically the exciton lifetime for OLEDs with a weak or a strong microcavity.

The theoretical analysis of exciton lifetime is based on a classical approach where the emitter is considered as an electric dipole running at a fixed current and with a random orientation.\textsuperscript{5,15,16} As a consequence of Fermi’s golden rule, the radiative decay rate $\Gamma_{\text{r}}$ at a wavelength $\lambda$ in an OLED device is modified to\textsuperscript{5,15,16}

$$\Gamma_{\text{r}}(\lambda) = F(\lambda) \cdot \Gamma_0(\lambda),$$

where $\Gamma_0(\lambda)$ is the radiative decay rate in the infinite medium. Here, $F(\lambda)$, the so-called Purcell factor, is the normalized total emission power of the electric dipole with random orientation (normalized by the total power of the dipole in the infinite medium). It should be noted that here, the emission power coupled to the metal electrode is also included in $F$. We consider such decay channel as radiative decay since this part of emission can potentially be coupled out from the device through, for example, patterning the substrate.\textsuperscript{17} In addition, we typically assume that the nonradiative decay rate $\Gamma_{\text{nr}}$ does not change by varying the thickness of the hole-transport layer (HTL) or the electron transport layer (ETL) in the device. The initial quantum efficiency $\eta_0$, $\eta_{\text{int}}$, and exciton lifetime $\tau_{\text{OLED}}$ in the OLED are given by

$$\eta_0 = \int_{\lambda_1}^{\lambda_2} \Gamma_0(\lambda) d\lambda / \left[ \int_{\lambda_1}^{\lambda_2} \Gamma_0(\lambda) d\lambda + (\lambda_2 - \lambda_1) \Gamma_{\text{nr}} \right],$$

$$\eta_{\text{int}} = \int_{\lambda_1}^{\lambda_2} F(\lambda) \cdot \Gamma_0(\lambda) d\lambda / \left[ \int_{\lambda_1}^{\lambda_2} F(\lambda) \cdot \Gamma_0(\lambda) d\lambda + (\lambda_2 - \lambda_1) \Gamma_{\text{nr}} \right],$$

$$\tau_{\text{OLED}} = \tau_0 / \left[ \eta_0 \int_{\lambda_1}^{\lambda_2} F(\lambda) \Gamma_0(\lambda) d\lambda / \int_{\lambda_1}^{\lambda_2} \Gamma_0(\lambda) d\lambda + 1 - \eta_0 \right],$$

where $(\lambda_1, \lambda_2)$ is the wavelength range of the emission and $\tau_0$...
is the exciton lifetime of the bulk emitting material. Equation (4) can be written as

$$\tau_{\text{OLED}} = \tau_0(1 - \eta_{\text{int}})/(1 - \eta_0).$$  

From Eq. (5), one observes that $\tau_{\text{OLED}}$ linearly depends on $\eta_{\text{int}}$, which means that $\eta_{\text{int}}$ can be determined from $\tau_{\text{OLED}}$. As shown above, the Purcell factor $F(\lambda)$ is a crucial quantity, which can be evaluated with the knowledge of the electric field at the location of the dipole. The electric field can be efficiently calculated by integrating the plane wave component of the field along a proper integration path.

In the experiment, europium (Eu) organometallic complexes were used in the OLEDs as the emitters. As shown in Fig. 1, four sets of OLEDs, i.e., devices A, B, C, and D, were fabricated to study the exciton lifetime of Eu ion based emitters in weak microcavity OLEDs (devices A and C) and strong microcavity OLEDs (devices B and D). The Eu complexes of Eu(2,2-bis(benzimidazole)-diphenyl-benzidine), Eu(TTA)$_3$(TPPO)$_2$, and Eu(dibenzoylemethanato)$_3$(bathophenanthroline) Eu(DBM)$_3$, bath were used as the emitting layer in devices A and B and devices C and D, respectively. The widely used material of bis(3-methylphenyl)-diphenyl-benzidine was used as the HTL. UV emissive (1,3,5-benzinetril)-tris(1-phenyl-1-H-benzimidazole) was used as the ETL. All the materials were evaporated by thermal vacuum deposition at a pressure of $10^{-6}$ Torr. The thickness of each layer is indicated in the inset of Fig. 2. The calculated Purcell factors $F(\lambda)$ of the two devices are also shown in the inset. According to Eq. (4) and the experimental results in Fig. 2, the initial internal quantum efficiency $\eta_0$ and the lifetime $\tau_0$ for Eu(TTA)$_3$(TPPO)$_2$ are determined to be 0.6 and 0.55 ms, respectively.

From Eq. (5), we observe that the internal quantum efficiency ($\eta_{\text{int}}$) depends linearly on the normalized exciton lifetime ($\tau_{\text{OLED}}/\tau_0$). In Fig. 3, we plot the linear relation as dashed, solid, and dash-dotted lines for $\eta_0=0.8, 0.6, 0.4$, respectively. All the lines pass a fixed point ($\tau_{\text{OLED}}/\tau_0=1$) and the slopes only depend on $\eta_0$. Therefore, after the determination of $\eta_0$ and $\tau_0$, $\eta_{\text{int}}$ of an OLED device can be obtained by measuring the exciton lifetime, which is much easier as compared with the direct measurement. In Fig. 3, the open circle and square are for devices A and B ($t=115$ nm) studied in Fig. 2, respectively. Device B has a larger $\eta_{\text{int}}$ than device A due to a larger Purcell effect in device B.

Figure 4(a) shows the experimental (symbols) and theoretical (lines) exciton lifetimes of devices A and B as the functions of the total thickness of the organic layers $t$. From the figure, we observe a good agreement between the simulated and experimental results. For device A, the lifetime only changes slightly when $t$ varies since the weak cavity effect has less influence on the spontaneous emission property. However, for device B (strong microcavity OLEDs), the exciton lifetime changes significantly as $t$ varies due to the strong microcavity effect and the surface plasmon effect. In device B, when the emissive layer is close to the silver film (small $t$), the emission has a good coupling to the surface plasmon modes which is absent in device A. Thus, the exciton lifetime is significantly shortened at small $t$ for device B. As $t$ increases, the surface plasmon effect is quickly diminished and microcavity effect gradually becomes pronounced. When the resonant condition of the microcavity is satisfied, the exciton lifetime of device B will reach a minimum.
device D, the simulated and experimental results agree well in trend but have small discrepancy in absolute values. This may be due to some deviation in measuring the thickness of the silver film.

In conclusion, a comprehensive analysis has been given on the modifications of the exciton lifetime ($\tau_{\text{OLLED}}$) and internal quantum efficiency ($\eta_{\text{int}}$) in OLEDs. We have shown that $\tau_{\text{OLLED}}$ has a linear relation with $\eta_{\text{int}}$. This means that $\eta_{\text{int}}$, which is important but difficult for a direct measurement, can be estimated easily by measuring $\tau_{\text{OLLED}}$. The modifications of exciton lifetimes in various OLEDs have been well explained through the microcavity effect and surface plasmon effect.

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5. E. M. Purcell, Phys. Rev. 69, 681 (1946).