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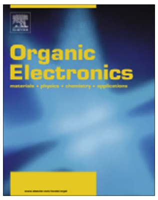

Transient electroluminescence in heavy metal complex-based phosphorescent organic light-emitting diodes

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ABSTRACT

In phosphorescent organic light-emitting diodes (PHOLEDs), both the rise time and decay time decrease with increasing amplitude of the applied voltage pulse. The rise time τ_r of the transient electroluminescence (TEL) increases linearly with increasing value of the ratio of voltage V to the current j, that is, with V/j. Using the equations for the dynamics of charge carriers an expression is derived for the rise time τ_r of the TEL in OLEDs. It is shown that τ_r should increase with increasing values of the ratio (V/j), dielectric constant ε, and area of cross-section of the emission layer, however, it should decrease with the thickness of emission layer. For higher values of the applied voltage nonlinearity occurs in the τ_r versus V/j plot because the increase in mobility of carriers at high electric field causes increase in the current flowing through the OLEDs. In fact, the rise time of TEL is related to the product of capacitance and effective resistance of the OLED. Considering the rate of generation and decay of radiative triplet excitons in the emission layer, an expression is derived for the decay time of TEL in PHOLEDs and it is shown that, for higher values of the time-constant of OLED, the decay time should be equal to the time-constant, however, for lower values of the time-constant, the decay time should be equal to the lifetime of radiative triplet excitons in the emission layer. A good agreement is found between the theoretical and experimental results.

1. Introduction

Now-a-days, phosphorescent organic light-emitting diodes (PHOLEDs) are attracting the interest of a large number of researchers because of their applications in displays and environmentally friendly solid state lighting devices [1–4]. The possibility of utilization of triplet excitons in organic light emitting diodes (OLEDs) was proposed by Kido et al. [5] and it was successfully demonstrated by Baldo et al. [6]. In fact, the OLEDs prepared with heavy metal complexes, particularly those containing $\mathrm{Ir^{III}}$, are the most efficient OLEDs reported, in which the theoretical internal quantum efficiency attains a value of 100% due to the harvest of both singlet and triplet excitons. One of the concerns in using phosphorescent OLEDs is the response time [7]. For the application of such OLEDs in display sufficiently short response time, usually in the millisecond range or shorter is required. Therefore, it is important to investigate the transient electroluminescence (TEL) of phosphorescent OLEDs. Although the TEL of fluorescent OLEDs has been investigated extensively [8–30], the TEL of phosphorescent OLEDs is least studied and the dependence of rise time τ_r and decay time τ_d on different parameters is not known satisfactorily till now. Moon et al. [31] have investigated the phosphorescent OLEDs including 4,4-*N*,*N*′-dicarbazolylbiphenyl (CBP) doped with iridium(acetylacetonate) [Ir(acac)], CBP:Ir(acac), as an emission layer (EML), in which the rise time and decay time are found to decrease monotonically with the applied

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voltage pulse amplitude. Teramura et al. [32] have determined the TEL response of CBP:Ir(ppy)-based phosphorescent OLEDs, in which both the rise time and decay time are found to decrease exponentially with the amplitude of applied voltage pulse. Youn et al. [33] have reported that, when the applied voltage pulse in an PHOLED having $Bepp_2$ or CBP:Ir $(ppy)_3$ emission layer decreases, then an overshoot or satellite is observed in the TEL decay. Baldo et al. [7] have reported the exponential decay of the TEL in the PHOLEDs in which the decay time gives the lifetime of the radiative triplet excitons.

In the present paper, using the dynamic equations for charge carriers, expression is derived for the dependence of the rise time of PHOLEDs on different parameters. It is shown that the rise time τ_r of TEL in PHOLEDs, should increase linearly with the increasing ratio of the applied voltage V to the current j flowing through the OLED, that is with V/j. The expression derived for τ_r indicates that, in addition to V/j, the rise time τ_r should also depend on other factors such as the dielectric constant ε, the area of cross-section S of the emission layer and thickness d of the emission layer. An expression is also derived for the decay time τ_d of PHOLEDs, which indicates that for higher values of the time-constant of the OLED, the decay time should be equal to the time-constant, however, for lower values of the time-constant, the decay time should be equal to the lifetime of radiative triplet excitons in the emission layer. Furthermore, the comparison is made between the theoretical and experimental results, in which a good agreement is found. It is shown that the expressions derived for τ_r and τ_d may be useful in tailoring the PHOLEDs having desired values of rise time and decay time.

2. Theory

2.1. Rise of transient EL in phosphorescent OLEDs

Neglecting the spatial distribution of carriers in the emission layer and diffusion term for simplicity, the density of holes after the delay time t_d can be expressed as [9].

$$\frac{\partial p}{\partial t} = -\mu_h \frac{\partial}{\partial x}(pE) - Rnp \tag{1}$$

where E is the electric field and R is the Langevin recombination coefficient.

Assuming the recombination constant being determined by the Langevin recombination coefficient R, and taking the electron mobility to be greater than the hole mobility, that is, $\mu_e >> \mu_h$ (as in the case for the PHOLEDs used by Moon et al. [31] and Teramura et al. [32]), we can write

$$R = \frac{q}{\varepsilon}(\mu_e + \mu_h) \approx \frac{q\mu_e}{\varepsilon} \tag{2}$$

where q is the electronic charge and ε is the dielectric constant of the organic material of EML.

For simplicity we can assume that μ_h and μ_e are independent of the electric field. The recombination current J_r is directly related to the integral of the carrier density product, and therefore, it can be expressed as [9].

$$J_r = q\int_0^d Rnpdx = J_0\left[1 - \exp\left(-\frac{t}{\tau_r}\right)\right] \tag{3}$$

where d is the thickness of the emission layer, $\tau_r = 1/Rn$, is the rise time of J_r, $J_0 = J_p(0) – J_p(d) = J_n(d) – J_n(0)$, J_h and J_n are hole current density and electron current density, respectively.

The EL intensity I is directly proportional to the recombination current density $J_r(t)$. If Z is the correlation factor between I and $J_r(t)$, then from Eq. (3),we get

$$I = ZJ_0[1 - \exp(-t/\tau)] = I_s[1 - \exp(-t/\tau)] \tag{4}$$

where, $I_s = ZJ_0$, is the steady state EL intensity.

In this case, the rise time of EL intensity can be expressed as

$$\tau_r = \frac{1}{Rn} = \left(\frac{\varepsilon}{q\mu_e n_e}\right) = \left(\frac{\varepsilon E}{J_0}\right) = \frac{\varepsilon S V_E}{jd} = \frac{(f\varepsilon S V)}{jd} \tag{5}$$

where E is the electric field in the emission layer, $J_0 = n_e q \mu_e E$, is the steady state current density, V_E is the voltage across the two sides of the EML, j is the current flowing through PHOLED, f is the fraction of the applied voltage V developed across the two ends of EML, μ_e is the electron mobility in the emission layer and n_e is the concentration of electrons in the emission layer.

Eq. (5) indicates that τ_r should increase linearly with increasing value of (V/j).It is seen from Eq.(5) that, in addition to V/j, τ_r should also depend on the dielectric constant ε, thickness d of the emission layer, and on the cross-sectional area S of the emission layer.

As $(\varepsilon S/d)$ is related to the capacitance C of the OLED and (V/j) is related to the effective resistance r_e of the OLED, Eq. (5) can be written as

$$\tau_r = \phi C r_e \tag{6}$$

where ϕ is the normalizing factor.

It is to be noted that, at higher amplitude of the voltage pulse, more numbers of charge carriers are injected, and therefore, r_e decreases and consequently, τ_r decreases (see Eq. (6)).

2.2. Decay of transient EL in phosphorescent OLEDs

When the applied voltage is turned off at $t = t_{c,}$ then the relaxation of charges Q across the OLED takes place and thus, we get, $Q = Q_0\exp[-\alpha(t - t_c)]$, where Q_0 is the charge at $t = t_c$, and $\alpha = 1/Cr_{d,}$ in which $r_d = dV/dj$, is the dynamic resistance of the OLED. The rate of generation of excitons in EML will be proportional to dQ/dt, and therefore, we can write the following equation

$$\begin{aligned}\frac{dN_{ex}}{dt} &= B\alpha Q_0 \exp[-\alpha(t - t_c)] - \frac{N_{ex}}{\tau_{ex}} \\ &= B\alpha Q_0 \exp[-\alpha(t - t_c)] - \beta N_{ex}\end{aligned} \tag{7}$$

where τ_{ex} is the lifetime of excitons, $\beta = \frac{1}{\tau_{ex}}$, and B is a constant.

Integrating Eq. (7) and taking $N_{ex} = N^0_{ex}$, at $t = t_c$, we get

$$N_{ex} = \frac{B\alpha Q_0}{(\beta - \alpha)} \exp[-\alpha(t - t_c)] + N^0_{ex} \exp[-\beta(t - t_c)] - \frac{B\alpha Q_0}{(\beta - \alpha)} \exp[-\beta(t - t_c)] \tag{8}$$

If η is the efficiency for the radiative decay of excitons, then the EL intensity can be expressed as

$$I = \eta\beta\left[\frac{B\alpha Q_0}{(\beta - \alpha)} \exp\{-\alpha(t - t_c)\} + N^0_{ex} \exp\{-\beta(t - tc)\} - \frac{B\alpha Q_0}{(\beta - \alpha)} \exp\{-\beta(t - t_c)\}\right] \tag{9}$$

Now, the following three cases arise:

Case I: $\beta >> \alpha$

For $\beta >> \alpha$, Eq. (9) becomes

$$I = \eta B\alpha Q_0 \exp[-\alpha(t - t_c)] \tag{10}$$

In this case, the decay time $\tau_d = 1/\alpha = Cr_d$, should decrease with increasing value of the voltage as r_d decreases with increasing voltage.

Case II: $\alpha >> \beta$

For $\alpha >> \beta$, Eq. (7) gives

$$I = \eta\beta N^0_{ex} \exp[-\beta(t - t_c)] \tag{11}$$

In this case, the decay time $\tau_{ex} = 1/\beta$ should be independent of the applied voltage.

Case III: Overshoot in the TEL decay

In certain OLEDs, overshoot occurs in the TEL decay. Such peak may be due to the delayed fluorescence [34], an electrical distribution of charge carriers [8,10,29,30] or it may be due to the phosphorescence in which light emission is delayed about triplet lifetime after turning off the applied voltage pulse [35].

3. Correlation between the theoretical and experimental results

Moon et al. [31] have measured the TEL in two OLEDs, hereby called, OLED-1 and OLED-2. The structure of OLED-1 is: Ni(200 nm)/2-TNATA(20 nm)/α-NPD(30 nm)/CBP:BtpIr (acac)(5%, 35 nm): BCP(10 nm)/Ca(10 nm)/Ag(10 nm), and the structure of the OLED-2 is: Ni (200 nm)/2-TNATA(20 nm)/α-NPD (30 nm)/CBP:BtpIr(acac) (5% 35 nm)/BCP(10 nm)/Alq_3 (10 nm)/BCP(5 nm)/Ca(10 nm)/ Ag (10 nm). In order to study the time evolution of TEL, rectangular voltage pulses of different amplitudes varying from 4.5 to 9.5 V were applied to the OLED. The TEL was detected by placing a fast silicon photo-diode directly onto the surface of the OLED. Fig. 1(a) shows the typical example of an applied voltage pulse and the TEL of OLED-1 and OLED-2 are shown in Fig. 1(b) and Fig. 1(c), respectively. Fig. 2 indicates the voltage pulse amplitude dependence of the rise, decay and delay times. The results for OLED -1 and OLED-2 are displayed in layers (a) and layer (b) of Fig. 2, respectively. The rise and decay times decrease monotonically with the applied pulse amplitudes, in which these variations are of two orders of magnitude in a voltage range between 4 and 10 V. Interestingly, changes in OLED-1 are more significant, in which a substantial variation of the decay time of about 18% has been observed as compared to only 8% variation of the decay time in OLED-2.

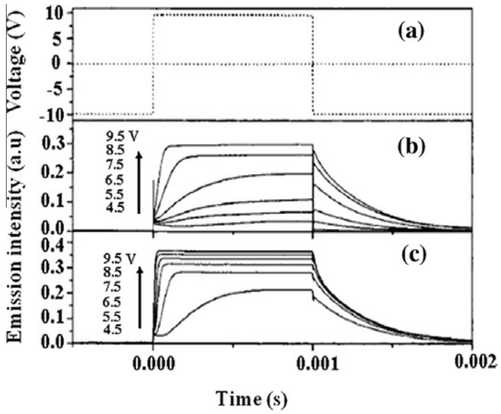

Fig. 1. Time evolution of electroluminescence as function of the voltage pulse amplitude. (a) Typical example of an applied voltage pulse, (b) transient electroluminescence in the OLED-1, and (c) transient electroluminescence in OLED-2. (after Moon et al. Ref. [31]).

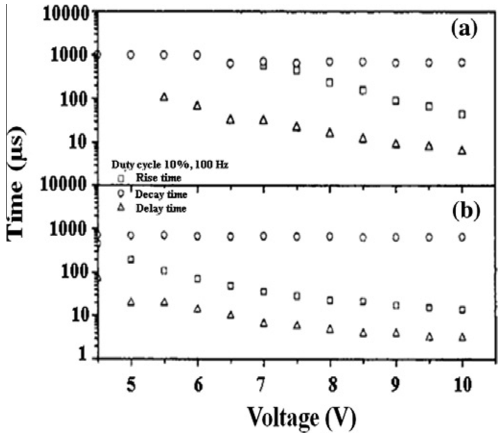

Fig. 2. Rise, decay, and delay times as function of the voltage pulse amplitude (a) for OLED-1 and (b) for OLED-2. (after Moon et al. Ref. [31]).

Teramura et al. [32] have fabricated two kinds of phosphorescent OLEDs, namely, OLED-A and OLED-B. The structure of OLED-A is: glass substrate/ITO/PEDOT:PSS/α-NPD (HTL) (20 nm)/EBL (20 nm)/CBP:Ir $(ppy)_3$ (40 nm) EML/BCP (10 nm)/Alq_3 (ETL) (20 nm)/Ag:Mg (150 nm). The structure of OLED-B is: glass substrate/ITO/PEDOT:PSS/α-NPD (HTL) (20 nm)/EBL (20 nm)/[CBP (20 nm) (CBP:Ir $(ppy)_3$ (20 nm)] EML/BCP)10 nm/Alq_3 (ETL (20 nm)/Ag:Mg (150 nm). Device A was uniformly doped in EML and device B was a cathode-side-doped OLED. The approximate doping concentration of $Ir(ppy)_3$ in the host CBP was 4%. The light emitting area was 4×4 mm^2. The ITO and cathode strip electrodes of 4×7 mm^2 area were arranged orthogonally. The voltages of OLED-A and OLED-B for current of 1 μA were 5.6 V and 7.2 V, respectively. The spectra observed in OLED-A were centered around 510 nm, showing typical emission spectra arising from $Ir(ppy)_3$. Although an additional luminescence peak appeared around 430 nm in OLED-B together with the main peak at 510 nm, the intensity ratio of the second peak compared with the main peak was as low as 0.1. The TEL of OLEDs was measured by applying a pulse voltage. The response time of the driving was as short as 15 ns, and TEL was detected with a photomultiplier tube whose response time was 0.8 ns.

Fig. 3 shows the current–voltage characteristics of OLED-A and OLED-B. The inset in Fig. 3 shows the semilog plot of current versus voltage plots for OLED-A and OLED-B. It is evident that the plots are nonlinear. Fig. 4 illustrates the luminance versus currents for OLED-A and OLED-B. The optical intensity ratio of OLEDs A:B was 1:0.25. The TEL of OLED-B is shown inside the figure for a pulse voltage of $V_p = 9$ V applied to the device. It is seen that the rise time τ_r is markedly longer than the decay time t_d.

Fig. 5 shows the dependence of τ_r and τ_d on the pulse voltage V_p, in which the pulse-width was 10 ms. It is evident that the rise time and decay time of OLED-1 and OLED-2 decrease with increasing value of Vp. The rise time is much longer than the decay time. The EL delay time t_d appearing before the onset of EL was nearly 700 ns at $V_p = 6$ V for both the devices. Both τ_r and τ_d of OLED-B are longer than those of OLED-A at the same V_p. The transient response times of photoluminescence (PL) measured using 405 nm blue semiconductor laser diode are 1.5 μs for OLED-A and 0.9 μs for OLED-B.

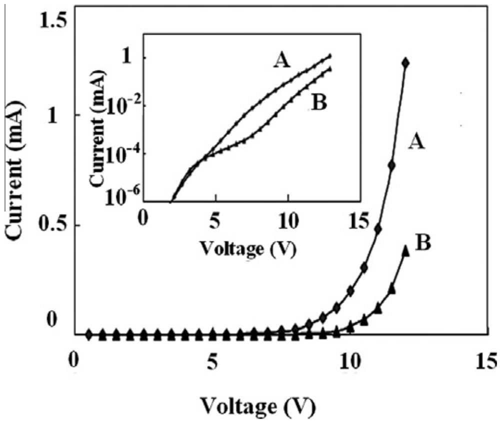

Fig. 3. Current–voltage characteristics of OLED-A and OLED-B. The voltage of OLED-A for a current of 1 mA was 5.6 V, and that for OLED-B was 7.2 V. Inset: Semilog plot of current versus voltage. The semilog plot is almost linear at voltages of more than 8 V. (after Teramura et al. Ref. [32]).

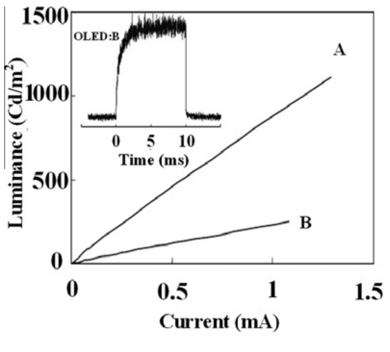

Fig. 4. Luminance versus current plots for OLEDs-A and -B. The optical intensity ratio of A:B was 1:0.25. TEL of B is shown inside the figure, when a pulse voltage of $V_p = 9$ V, is applied to the device (after Teramura et al., Ref. [32]).

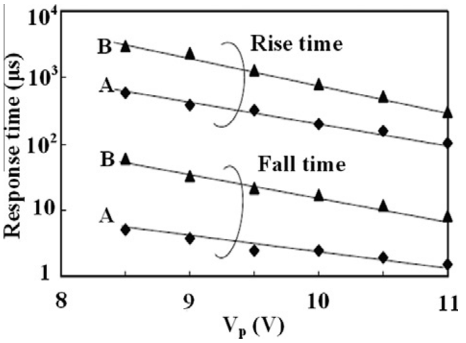

Fig. 5. Dependence of the rise time and fall time on the voltage pulse amplitude V_p. (after Teramura et al., Ref. [32]).

Fig. 6 shows the dependence of the rise times of OLED-A and OLED-B on the current flowing through the devices. It is seen that the plots almost follow the same curve, indicating that the rise time for OLED-A and OLED-B are determined by the magnitude of the current.

Figs. 7 and 8 show the dependence of the rise time τ_r on the ratio of V and j, that is, on V/j for OLED-A and OLED-B. It

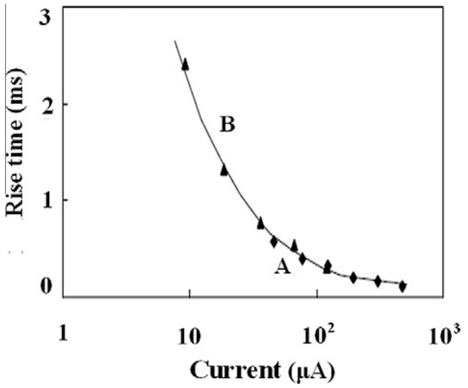

Fig. 6. Dependence of the rise times for OLED-A and OLED-B on current. The plots almost follow the same curve, indicating that the rise times for OLED-A and OLED-B are determined by the magnitude of the current. ♦: A, ▼: B (after Teramura et al., Ref. [32]).

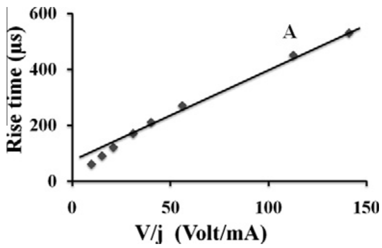

Fig. 7. Dependence of the rise time τ_r of OLED-A on V/j.

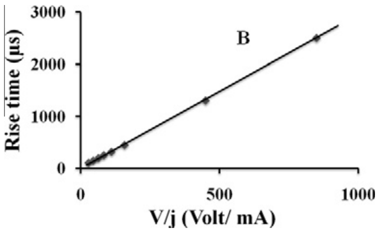

Fig. 8. Dependence of the rise time τ_r of OLED-B on *V*/*j*.

is evident that τ_r for OLED-B is linear with *V*/*j* for all the values of applied voltages, however, in the τ_r versus *V*/*j* plot of OLED-A nonlinearity occurs for the higher values of applied voltage. The linear relation between τ_r and *V*/*j* supports Eq. (5). The nonlinearity, in τ_r versus *V*/*j* plot occurs in OLED-A for higher values of the applied voltage because of the reduction of τ_r with increasing value of the mobility μ at higher electric field, whereby higher current flows through the OLEDs. (see Eq. (5)]. In the OLED-A fabricated by Teramura et al. [32], in the voltage range from 8.5 to 12 V, when *V*/*j* increases 2.5 times, the rise time τ_r, also increases nearly 2.5 times, and in the OLED-B fabricated by Teramura et al. [32], in the voltage range from 8.5 to 10.5 V, when A/j increases 9 times, the rise time τ_r also increases nearly 9 times. Furthermore, for a given voltage, the rise time is less in the OLED having higher value of the current, for example, for 8.5 V the current flowing through OLED-A is 0.06 mA and the current flowing through OLED-B is 0.01 mA, and the rise time τ_r for OLED-A is 530 µs and τ_r for OLED-B is 2500 µs. The expression derived in the present investigation indicates that, in addition to *V* and *j*, the rise time τ_r should also depend on other factors such as the dielectric constant ε, the area of cross-section *S* of the emission layer and thickness d of the emission layer.

It is evident from Eq. (5) that the rise time τ_r depends on the ratio (*V*/*j*), dielectric constant ε, on the area of cross-section of the emission layer and thickness *d* of the emission layer. Thus, by controlling these factors, an OLED of desired τ_r can be tailored.

The exponential decay of the TEL follows Eq. (10). The exponential decrease of τ_d with increasing voltage indicates that the dynamic resistance r_d decreases exponentially with increasing value of the applied voltage. As the current through the OLEDs increases exponentially with the applied voltage [36], the exponential decrease of r_d with applied voltage is expected.

Baldo et al. [7] have measured the lifetime of radiative triplet excitons in two kinds of PHOLEDs hereby called OLED-1 and OLED-2. The structure of OLED-1 is: ITO/α-NPD (HTL)(50 m)/Alq_3 (ETL and excitons diffusion layer/ 8% PtOEP in Alq_3 (EML) (10 nm)/Alq_3 (ETL) (20 nm)/Mg:Ag and the structure of OLED-2 is: ITO/NPD (HTL) (40 nm)/8% Ir $(ppy)_3$ in (CBP (10 nm)/BCP (12 nm)/Alq_3 (ETL) (30 nm)/ Mg:Ag. Fig. 9(a) shows the TEL response of 8% Ir $(ppy)_3$ in TPD. The device contains no diffusion layer yet the lifetime of Ir $(ppy)_3$ in TPD is significantly higher (15 µs) than the natural radiative lifetime of Ir $(ppy)_3$ ($\sim$1 µs). The initial peak in the response is principally due to fluorescence from TPD. Fig. 9(b) shows the photoluminescence response of 8% Ir $(ppy)_3$ in TPD at T = 292 K and T = 200 K. It is seen that the lifetime increases at low temperature which is consistent with a thermally activated process. However, unlike the TEL response the initial transient in the photoluminescence response is comprised entirely of emission from photoexcited Ir $(ppy)_3$. In this case, the TEL measurements were obtained by applying a narrow (200 ns) voltage pulse to the device under test and coupling the emission into a streak camera. The pulse width is chosen to be less than the radiative rate of the phosphors and larger than the charging time of the OLED, which for a 50 Ω load and a typical capacitance of 1 nF is $\sim$50 ns. In this case, the time-constant Cr_d is much less, hence, the inverse of the slope of the in EL intensity versus $(t - t_c)$ plot (Fig. 9a) gives the lifetime of the radiative triplet excitons. This result is in accord with Eq. (11).

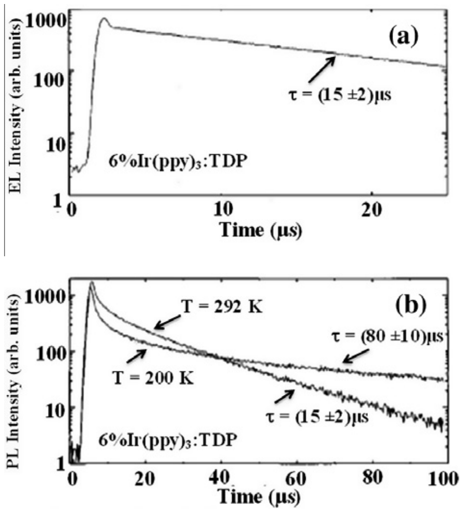

Fig. 9. (a) The transient electroluminescence response of 8% Ir $(ppy)_3$ in TPD, and (b) the photoluminescence response of 8% Ir $(ppy)_3$ in TPD at T = 292 K and T = 200 K (after Baldo et al. Ref. [7]).

Thus, there is a good agreement between the theoretical and experimental results.

4. Conclusions

When a voltage pulse is applied to a phosphorescent OLED, then initially the EL intensity increases with time, attains a saturation value and later on it decreases with time, whereby the decrease in EL intensity starts from the time at which the applied voltage begins decreasing. In phosphorescent organic light-emitting diodes, both the rise time and decay time decrease with increasing amplitude of the applied voltage pulse. The rise time τ_r of the TEL of PHOLEDs increases linearly with increasing value of the ratio of voltage *V* to current *j*, that is, with *V*/*j*. For higher values of the applied voltage nonlinearity occurs in the τ_r versus *V*/*j* plot because increase in mobility of carriers at high electric field causes increase in the current flowing through the OLEDs. Using the equations for the dynamics of charge carriers an expression is derived for the rise time τ_r of the TEL in PHOLEDs. The equation derived for τ_r is as given below: $\tau_r = (f\varepsilon SV)/jd = \phi CR_e$. It is evident from this equation that τ_r should increase with increasing values of the ratio (V/j), dielectric constant ε, and area of cross-section of the emission layer, however, it should decrease with the increasing value of the thickness of emission layer. In fact, the rise time is related to the product of capacitance and effective resistance of OLED.

Considering the rate of generation and the rate of decay of radiative triplet excitons in the emission layer, an expression is derived for the decay time of the TEL in PHOLEDs. The expression derived indicates that, for higher values of the time constant for OLED, τ_d should be equal to the time-constant (Cr_d), however, for lower values of the time-constant, decay time τ_d should be equal to the lifetime of the radiative excitons in the emission layer.

A good agreement is found between the theoretical and experimental results.

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