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Transient electroluminescence of fluorescent organic light- emitting diodes

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Abstract

When a voltage pulse is applied to a fluorescent OLED, then after a certain time, 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. Both the rise time τ_r and decay time τ_d decrease with increasing value of the voltage applied to the fluorescent OLEDs. The rise time τ_r of the TEL of fluorescent OLEDs increases linearly with increasing value of the ratio of voltage V to 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 fluorescent OLEDs. 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 fluorescent OLEDs. A good agreement is found between the theoretical and experimental results.

1. INTRODUCTION

Organic light-emitting diode (OLED) is an optoelectronic device consisting of a single layer or bilayer or multiple layer of organic materials sandwiched between two electrodes, at least one of which is transparent. Although significant experimental measurements have been made on the TEL of fluorescent OLEDs**,** the dependence of the rise time τ_r and decay time τ_d of fluorescent OLEDs on different parameters are not satisfactory known till now. In the present paper, expressions are derived for the dependence of τ_r and τ_d on different parameters and it is shown that there is a good agreement between the theoretical and experimental results.

2.THEORY

2.1 Rise time of EL intensity

Chandra and Chandra [1] have reported previously that the time dependence of EL intensity can be expressed as

$$I = ZJ_0[1-\exp(-t/\tau_r)] = I_s[1-\exp(-t/\tau_r)] \quad (1)$$

where Z is the correlation factor between I and $J_r(t)$, $I_s = ZJ_0$, is the steady state EL intensity, $\tau_r = 1/Rn$, R is the Langevin recombination coefficient and n is the density of electrons in the emissive layer of OLED. From Eq. [1], the rise time of EL intensity can be written as

$$\tau_r = \frac{1}{Rn} = \left(\frac{\varepsilon}{q\mu_e n_e}\right) = \left(\frac{\varepsilon E}{J_0}\right) = \frac{\varepsilon SV_E}{jd} = \frac{(f\varepsilon SV)}{jd} \quad (2)$$

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.

Because $(\varepsilon S/d)$ is related to the capacitance C of the OLED and (V/j) is related to the differential resistance r_d of the OLED, Eq. (2) can be expressed as

$$\tau_d = \phi C r_e \quad (3)$$

where ϕ is the normalizing factor.

If the voltage dependence of the current j is given by the following equation

$$j = j_0 \exp[(\beta V) - 1] \quad (4)$$

where J_0 and β is a constant.

From Eq. (4) the differential resistance r_d can be expressed as

$$r_d = \frac{dV}{dj} = \frac{\exp(-\beta V)}{\beta J_0} \quad (5)$$

From Eqs. (3) and (5), τ_r is given by

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$$\tau_r = \frac{\phi C}{\beta J_0}\exp(-\beta V) \qquad (6)$$

2.1 Decay time of EL intensity

The decay of TEL starts from the time t_c, at which the applied voltage pulse starts decreasing. If the relaxation of charges Q across the OLED starts at t = t_c, then, 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 differential 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 rate equation

$$\frac{dN_{ex}}{dt} = B\alpha Q_0 \exp[-\alpha(t-t_c)] - \frac{N_{ex}}{\tau_{ex}}$$

$$= B\alpha Q_0 \exp[-\alpha(t-t_c)] - \beta N_{ex} \qquad (7)$$

where τ_{ex} is the lifetime of excitons, $\beta = 1/\tau_{ex}$, B is a constant, and N_{ex} is the number excitons at any t time after t_c

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

$$N_{ex} = \frac{B\alpha Q_0}{(\beta-\alpha)}\exp[-\alpha(t-t_c)] +$$

$$N_{ex}^0 \exp[-\beta(t-t_c)] - \frac{B\alpha Q_0}{(\beta-\alpha)}\exp[-\beta(t-t_c)] \qquad (8)$$

If η is the efficiency for the radiative decay of excitons then the EL intensity is, $I = \eta\beta N_{ex}$, and from Eq. (8), we get

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

Now, the following three cases arise:

Case I: β >> α

For β>>α, Eq. (9) becomes

$$I = \eta DB\alpha Q_0 \exp[-\alpha(t-t_c)] \qquad (10)$$

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

Case II: α>>β

For α>>β, Eq. (9) gives

$$I = \eta\beta N_{ex}^0 \exp[-\beta(t-t_c)] + \frac{\eta\beta B\alpha Q_0}{(\alpha-\beta)}\exp[-\beta(t-t_c)]$$

or, $$I = \eta[\beta N_{ex}^0 + \beta B Q_0]\exp[-\beta(t-t_c)] \qquad (11)$$

As the rate of generation of excitons at t = t_c, is $B\alpha Q_0$ and lifetime of excitons is 1/β; we get, $N_{ex}^0 = (B\alpha Q_0)/\beta$. Thus, Eq. (11) can be expressed as

$$I = \eta\beta Q_0 \alpha\left[1+\frac{\beta}{\alpha}\right]\exp[-\alpha(t-t_c)] \qquad (12)$$

For β >> α, Eq. (12) is given by

$$I = I_s \exp[-\beta(t-t_c)] \qquad (13)$$

where $I_s = \eta B\alpha Q_0$

Equation (13) indicates that for α >> β, the decay time τ_{ex} = 1/β and thus the decay time of TEL should be equal to the lifetime of excitons, and it should not depend monotonically on the amplitude of the applied voltage pulse.

3. EXPERIMENTAL SUPPORT TO THE PROPOSED THEORY

Barth et al**.** [2] have studied the voltage dependence of the transient EL from an ITO/CuPc ~20 nm/NPB ~45 nm/ Alq3 ~50 nm /Mg:Ag multilayer OLED , in which the pulse width was 5 ms and the repetition rate was 1 kHz . It is found that when a voltage is applied to the OLED, then initially the EL intensity increases with time, attains a saturation value, and when the applied voltage is turned off, the EL intensity decreases with time and finally disappears. Whereas the EL intensity increases with the applied voltage, the delay time of the OLED decreases with increasing value of the applied voltage. The value of rise time is determined from the time required for the rise of EL intensity from 0 to 62% and the value of decay time is determined from the time required for the decrease of EL intensity from 100 % to 38 %. It is found that both the rise time and the decay time decrease with increasing value of the applied voltage. Fig. 1 shows the semilog plots of the rise time versus applied voltage and semilog plot of the decay time versus applied voltage for the ITO/CuPc ~20 nm/NPB ~45 nm/ Alq3 ~50 nm /Mg:Ag multilayer OLED OLED. It is seen that the plots are straight lines with a negative slopes. This result is in accord with Eqs**.** (6) and (12).

Barth et al. [2] have also studied the voltage dependence of the time-resolved EL from an ITO/Alq_3 ~100 nm/Mg:Ag single-layer OLED, in which the pulse length was 10 ms and the repetition rate was 1 kHz. The results for rise and decrease of EL intensity for ITO/Alq_3 ~100 nm/Mg:Ag single-

layer OLED, are similar to that obtained for ITO/CuPc ~20 nm/NPB ~45 nm/ Alq3 ~50 nm /Mg:Ag multilayer OLED, except that the values of the rise time and decay time in a single layer OLED are different as compared to that for the multilayer OLED.

Fig. 2 shows the dependence of the rise time τ_r on the differential resistance for ITO/α-NPD (30 nm)/rubrene (9.1 vol%) doped in Alq_3 (30 nm)/Alq_3 (10 nm)/Mg:Ag with the active area of 0.02mm^2 size, in which the data were taken from ref. [3]. It is seen that the plot is a straight line with a positive slope. This result is in accord wit h Eq. (2).

Fig. 3 shows the decay of EL at the falling edge of an applied 10 V pulse on a semilogrithmic scale for ITO/PPV/Al OLED][4]. The decay signal is non-exponential, and it can be described as the sum of two expontial decays with time constants of approximately 1 μs (component C) and 80 μs (component D), respectively. From photoluminescence measurement it is shown that the radiative recombination of the singelet recombition it is kown that the the singelet exciton occurs with a characteristic decay time of 100 ps. Therefore ,the measured decay time of component C is dominated by the RC time constant of the OLED and the setup. However, the component D shows the typical behaviour of a delayed luminescence. A delayed luminescence has been observed in undoped anthracene crystals. In these crystals, the exponential tail in the has been interpreted as delayed fluorescence caused by triplet-triplet annihilation./. In this model, the decay of EL (component D) is related to the lifetime of the triplet excitons.

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

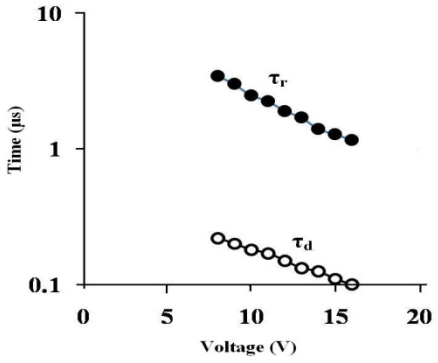

Fig. 1: Semilog plot of rise time and decay time of an ITO/CuPc (20 nm)/NPB(45 nm/Alq3 (50 nm)/Mg:Ag multilayer OLED. The pulse width was 5 μs and the repletion rate 1 kHz.

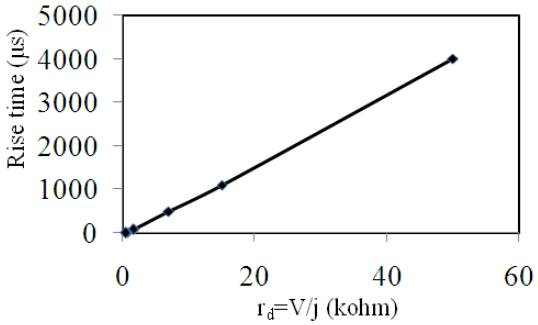

Fig. 2: Dependence of the rise time on the differential resistance for ITO/α-NPD (30 nm)/rubrene (9.1 vol%) doped in Alq_3 (30 nm)/Alq_3 (10 nm)/Mg:Ag with the active area of 0.02mm^2 size. The data were taken from the j-V curve given in ref. [3].

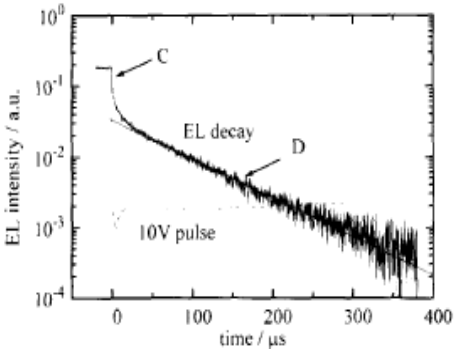

Fig. 3: EL decay at the falling edge of 10 V pulse showing a prompt EL (lifetime less than 1μs) and a delayed EL (lifetime about 80 μs) (: ____, EL intensity, and ------, voltage) (after Karg et al., ref. [4]).

4. CONCLUSION

The value of rise time and decay time of fluorescent OLEDs increases linearly with increasing value of the ratio between the applied voltage and current, that is, with (V/j). There is a good agreement between the theoretical and experimental results.

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