Recombination Efficiency in Bilayer Organic Light Emitting Diodes

Seminar Paper[*]

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Abstract: Bilayer Organic Light Emitting Diodes (OLEDs) consists of two thin organic layers sandwiched between two electrodes, with the structure, ITO (anode)/ hole transporting layer (HTL)/ electron transporting layer (ETL)/Al (cathode). In this paper we assume the analytical model and using the numerical simulations to calculate efficiency of carrier recombination in bilayer OLEDs. It is shown that the most important factor that determines efficiency is the ratio of hole and electron mobility in the ETL. When this ratio is much less than unity, recombination efficiency is 100% even without the presence of either an electron or hole barrier at the organicorganic interface. However, if the hole mobility is much higher than electron mobility, then a hole barrier at the organic interface is necessary to achieve a high recombination efficiency. The minimum interfacial hole barrier height required for high efficiency increases with the ratio of hole and electron mobilities in electron transport layer. The present study may be useful for the increasing recombination efficiency in bilayer OLEDs.

Keywords: Recombination efficiency, Hole transporting layer, Electron transporting layer.

1.   Introduction

Organic light emitting diode (OLED) displays are being increasingly viewed as the flat panel technology of the future due to their several advantages including easy manufacturability, low cost, wide viewing angle, fast response time and thin size [1–3]. The simplest bilayer OLED suffers from poor recombination efficiency [4,5], because in most organic materials, the hole mobility is much larger than electron mobility so that most of the carriers recombine in a region close to cathode where there is much greater probability of non-radiative recombination. By incorporating an additional suitably chosen organic layer, recombination can be shifted predominantly to a region close to the organic interface. As a result of the shift in recombination peak away from the quenching sites next to the cathode, much higher radiative recombination efficiency can be obtained. Since they were first reported [1], several studies have been conducted to investigate the operating principles of these bilayer OLEDs [6–13]. However, the impact of hole and electron mobility in hole and electron transport layers and energy offsets at the organic–organic interface on recombination efficiency has not been clearly elucidated.

In this paper we described an analysis of carrier recombination in bilayer OLED and constraints that must be obeyed to ensure that all injected electrons and holes recombine within the bulk of the organic layers. While, numerical simulations are

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used to provide more accurate quantitative results, simple analytical models are used to provide insight into the operation of the device. The present paper reports the analytical model for recombination efficiency in bilayer OLEDs.

2.   Mathematical Approach For Recombination Efficiency

A bilayer device consisting of an organic material-1 which serves as a HTL and an organic material-2 which serves as an ETL is shown in Fig. 1. The associated band diagrams are shown in Fig. 2. ΔE_L (ΔE_H) is the discontinuity in the lowest unoccupied molecular orbit (LUMO) [highest occupied molecular orbit (HOMO)] level at the organic-organic interface. In order to isolate the effect of electron and hole mobilities on efficiency the barrier height between the anode (cathode) metal and HTL (ETL) is taken such that both hole and electron current in the device are bulk-limited. Like in most bilayer devices, the hole mobility in HTL is assumed to be much larger than electron mobility in ETL Since carrier recombination close to electrodes may be largely non-radiative, recombination efficiency in this work is determined by considering recombination in the entire device except for regions of width 100 $\overset{0}{\text{A}}$ close to both the electrodes.

$$\eta = \frac{J_P(L_1) - J_P(L_2)}{J} \tag{1}$$

Where η is the recombination efficiency, J is the total current density, J_P is the current density of hole, $L_1 = 100 \overset{0}{\text{A}}$ and $L_2 = L - 100 \overset{0}{\text{A}}$. Fig. 3 shows the impact of hole and electron mobility respectively in ETL on carrier recombination efficiency. The hole and electron mobility in HTL were kept constant and the internal barriers at the organic interface were assumed to be absent so as to illustrate the impact of carrier mobility in ETL alone. It can be seen that carrier recombination efficiency improves with reduction in hole mobility and increase in electron mobility in ETL. In the inset of Fig. 3 the results are re-plotted as a function of mobility ratio μ_{P2}/μ_{N2}.

The fact that the efficiency curves corresponding to hole and electron mobilities overlap when plotted against the mobility ratio μ_{P2}/μ_{N2} clearly shows that this ratio is very important from the point of recombination efficiency. It can be seen that recombination efficiency is close to ideal (100%) when this ratio is much less than unity. It also show that hole mobility in HTL is large, it has no significant impact on efficiency. Similarly, it was found that energy offset in the LUMO level at the organic- organic interface also has a negligible impact on recombination efficiency. In contrast, the energy discontinuity in the HOMO level was found to have a large impact as illustrated in Fig.4. There are two important results to note from fig. 4. First, the impact of HOMO level discontinuity (or hole barrier height) decreases as mobility ratio μ_{P2}/μ_{N2} becomes smaller. Second, the minimum magnitude of hole barrier height required to achieve efficiency larger than 99% increases with increase in mobility ratio μ_{P2}/μ_{N2}.
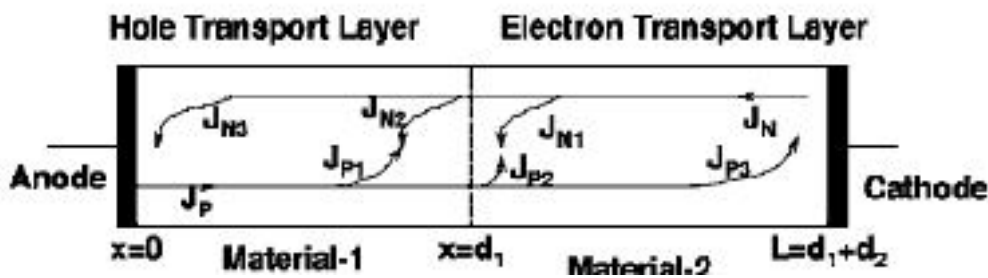

Fig. 1: Schematic diagram of a bilayer OLED, illustrating the different component of hole and electron currents.

In Fig. 1 we find the different components of hole and electron current in a bilayer device. The hole current can be broken into three distinct components, Component-1 which results from recombination with electrons in HTL, component-2 which result from recombination with electrons in ETL and component-3 which results from leakage of holes to cathode. Similarly, the electron current can be broken into three components. The recombination efficiency can be defined as

$$\eta = \frac{J - (J_{P3} + J_{N3})}{J} \tag{2}$$

where J_{N3} is the leakage current of electron, J_{P3} is the leakage current of hole. In a well designed OLED, all the holes injected from the anode recombine with electrons injected from the cathode in a region close to the organic-organic interface. Using Equation (2) a criterion for high recombination efficiency can therefore be written as

$$J_{P3} + J_{N3} \langle\langle J \tag{3}$$

Since hole mobility in HTL is much higher than electron mobility in ETL the probability of electrons reaching anode without recombining is very small, so J_{N3} can be ignored in Equation (3), the criterion for high efficiency then becomes,

$$J_{P3} \langle\langle J \cong J_N(ETL) \tag{4}$$

where J_N (ETL) is the electron current in ETL. The assumption that hole mobility in HTL (μ_{P1}) is much larger than electron mobility in ETL (μ_{N2}) effectively means that almost all the applied voltage drops across ETL as a result the electron current, described by space charge limited conduction in ETL can be expressed as

$$J_N(ETL) = 9/8 \times \varepsilon_0 \varepsilon_r \times \mu_{N2} \times (V - V_{bi})^2 / d_2^3 \tag{5}$$

Where V is the applied voltage of bilayer OLED, ε_0 is the permittivity of the free space, ε_r is the dielectric constant, V_{bi} is the built-in voltage of bilayer OLED and d_2 is the thickness of ETL.
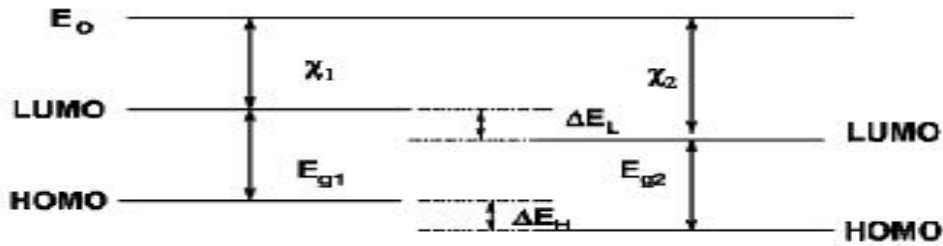

Fig. 2: Energy band diagram of OLEDs (E_0 is a electric field, χ_1 is electron affinity of material-1 and χ_2 is electron affinity of material-2).

Consider a situation where electron injection into the device is suppressed so that only hole current flows in the device. In this case the entire hole current will be due to flow of holes to the cathode and as a result

$$J_P^{SC} \langle\langle J_N(ETL) = 9/8 \times \varepsilon_0 \varepsilon_r \times \mu_{N2} \times (V - V_{bi})^2 / d_2^3 \tag{6}$$

Where J_P^{SC} is the magnitude of hole current with single carrier mode. Equation (6) implies Equation (4) and thus represent a more conservative criteria for achieving high efficiency. Equation (4) can be used to show that when $\mu_{P2}/\mu_{N2} \langle\langle 1$ Then
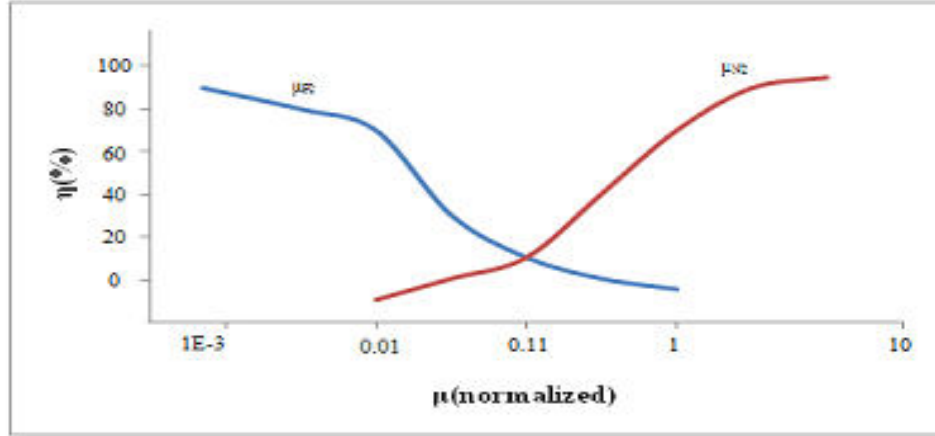
recombination efficiency is very high. The condition $\mu_{P2}/\mu_{N2} \langle\langle 1$ also implies that $\mu_{P2}/\mu_{P1} \langle\langle 1$. A typical hole density profile in the device under this condition is shown in Fig. 5. Result shown are for a device with $E_{g1} = E_{g2} = 2.4eV$, $\chi_1 = \chi_2 = 2.9eV$, $\mu_{P1} = 0.5 \times 10^6 cm^2/Vs$ and $d_1 = d_2 = 500\overset{0}{A}$ and applied voltage was 5eV.

Fig. 3: Recombination efficiency as a function hole (blue line) and electron (red line) mobility in ETL.

There are two important results to note from Fig. 3. First, the effect of HOMO level discontinuity (or hole barrier height) decreases as mobility ratio μ_{P2}/μ_{N2} becomes smaller. Second, the minimum magnitude of hole barrier height required to achieve efficiency larger than 99% increases with increase in mobility ratio μ_{P2}/μ_{N2} .
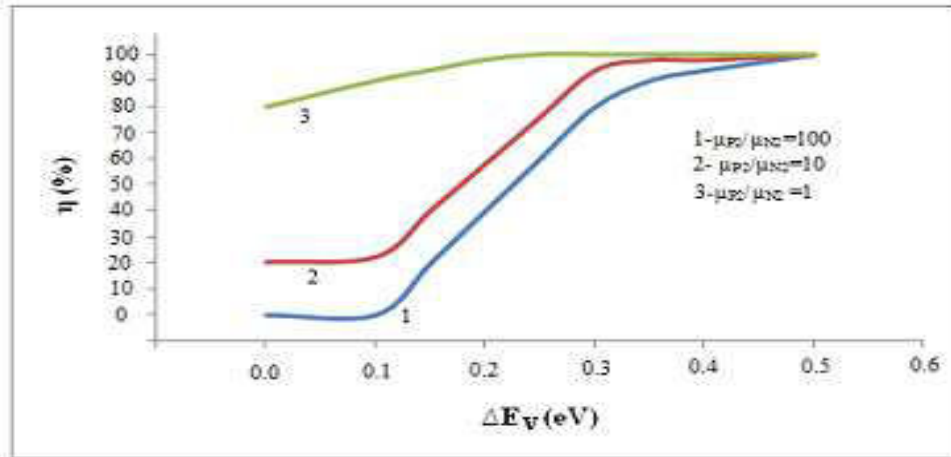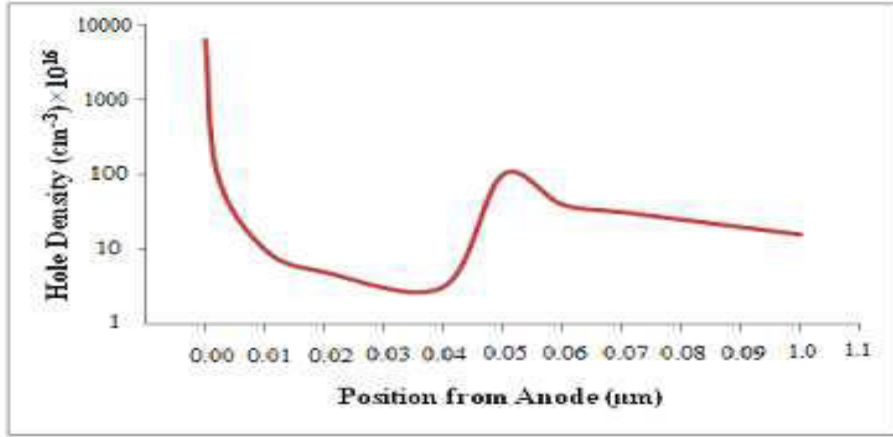

Fig. 4: Recombination efficiency as a function of HOMO level discontinuity for different values of mobility ratio μ_{P2}/μ_{N2}

Fig. 5: A typical hole density profile in a bilayer OLEDs.

As a result of discontinuity in hole mobility, an accumulation of holes at the interface occurs so that quasi-equilibrium can be assumed to exist. It allows modeling of current transports in ETL as if it is single layer device with accumulated hole density at the interface serving as the injecting contact and cathode forming the second contact. As a result, the hole current can be described by space charge limited conduction in ETL. The assumption that almost all the applied voltage drops across ETL allows the hole current to be expressed as space charge limited current:

$$J_P^{SC} = 9/8 \times \varepsilon_0\varepsilon_r \times \mu_{p2} \times (V^2/d_2^3) \tag{7}$$

Substitution of Equation ([7]) in Equation ([6]), and we get

$$\mu_{P2}/\mu_{N2} \langle\langle 1 \tag{8}$$

Equation ([8]) reveals that recombination efficiency will be very high when the ratio of hole and electron mobility in ETL is much smaller than unity. It shows that energy offsets at organic-organic interface for blocking of hole injection is unnecessary when this condition is satisfied. The criteria that hole mobility be much smaller than electron mobility in ETL for obtaining high efficiency in the absence of energy offsets can be understood also through the model shown in Fig. 6.
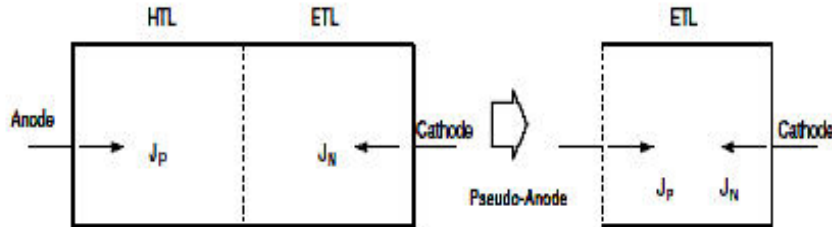

Fig. 6: Schematic diagram showing that the bilayer OLEDs shown on the left can be modeled as a single layer device (on the right) consisting only of electron transport layer. The anode together with hole transport layer can be modeled as a pseudo-anode.

The bilayer OLEDs is modeled as a single layer device consisting of only ETL layer with the usual cathode forming one contact and HTL serving as a pseudo anode. If it is assumed that hole barrier height at anode is small and HTL mobility is high then the pseudo anode can be considered as a good injector of holes and hole current in ETL can be considered to bulk limited. It is well known that recombination profile in a single layer OLED is determined by ratio of hole and electron mobility. When hole mobility is much higher, recombination takes place close to cathode which is undesirable. If on the other hand hole mobility becomes much smaller than electron mobility then most of the recombination would take place at the anode which would also normally undesirable because of the close proximity of metal which increase losses. However, shifting of recombination peak to pseudo- anode is what is required because efficient radiative recombination can take place near the organic-organic interface.

For the case where $\mu_{P2}/\mu_{N2} \rangle\rangle 1$, recombination peak can still be situated close to pseudo- anode to obtain high efficiency by employing a discontinuity in the HOMO level at the organic interface to reduce hole injection into ETL. The presence of a discontinuity in HOMO level of magnitude ΔE_H is expected to reduce hole injection into ETL by a factor $\exp(-\Delta E_H/kT)$.

As a result the injected hole current can be expressed as

$$J_P^{SC} \propto \mu_{P2} \times \exp(-\Delta E_H/kT) \tag{9}$$

Where k is Boltzmann's constant and T is the absolute temperature. Substitution of above expression in Equation (6) provides an estimate of minimum interfacial hole barrier height required to achieve high recombination efficiency:

$$\Delta E_H \rangle kTIn(\tfrac{\mu_{P2}}{\mu_{N2}}) + \text{constant} \tag{10}$$

Because of the approximations inherent in the derivation of Equation (9), the essential insight offered by it is that the magnitude of HOMO energy level offset required to achieve 100% recombination efficiency increase with increase in mobility ratio μ_{P2}/μ_{N2}.

3.   Conclusion

In present study, numerical simulations and analytical model show that the ratio of hole and electron mobility in electron transport layer is a key factor that determines recombination efficiency in bilayer devices. When this ratio is considerably less than unity, efficiency is high even without the presence of either an electron or hole barrier at the organic–organic interface. For the case of materials where the above mentioned constraint may not be satisfied, a hole interfacial barrier at the organic–organic interface is necessary and that its minimum required magnitude for obtaining high recombination efficiency increases with the ratio of hole and electron mobility in the electron transport layer. The present study may be useful for the increasing recombination efficiency in bilayer OLEDs.

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