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Electron-Hole Recombination Near the Interface of Bilayer OLEDs

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Abstract: The easy processing, flexibility, low operating voltage, thin size, low cost and other advantages of organic light emitting diodes (OLEDs) make them promising for flat panel technologies, and therefore, they are being widely investigated in recent years. Organic electroluminescent displays, monochromatic as well as multicolor have been reported. Understanding the operating principles of the device is necessary to achieve good performance of OLEDs. Study of electroluminescence provides information on the event controlling charge carriers and recombination process leading to light emission. The present paper reports the effect of interfacial barrier of charge carrier recombination in bilayer organic light emitting diodes. Considering the equation for the drift current in hole transport layer (HTL) equation is derived for the electric field at the interface, and then, assuming that the accumulation layer in HTL is like a charge-sheet, another expression relating charge and electric field is obtained through application of Gauss's law at the interface. Then the simplified expressions are obtained for the electric field near the HTL and electron transport layer (ETL). Subsequently, the expression for the ratio of currents in HTL and ETL is obtained. Finally, the expression is obtained for the dependence of the ratio of the recombination rates at HTL and ETL on interfacial heights, which shows that the ratio of the recombination rates of charge carriers at HTL and ETL should increase exponentially with the difference in ΔE_c and ΔE_v, where ΔE_c is electron barrier height and ΔE_v is hole barrier height. As the recombination rate controls the brightness of OLEDs, the present study may be useful in enhancing the brightness and efficiency of OLEDs.

Keywords: Recombination, OLEDs, electroluminescence, hole transport layer, electron transport layer.

I. INTRODUCTION

During the last decade, organic light emitting diodes (OLEDs) have attracted considerable interest from the research community due to their easier process to fabricatelower cost and the applications in multicolor flat-panel displays and solid state lighting devices [1]. OLED is an optoelectronic device in which a single layer, double layer or multilayer of organic materials is sandwiched between two electrodes, at least one of which is transparent. Electroluminescence (EL) in organic solids requires several steps, including the injection, transport, capture, and radiative recombination of holes and electrons inside an organic layer with suitable energy gap, to yield visible light. A very successful approach to optimize these individual steps separately is the concept of multilayer OLEDs with heterostructures between different organic materials [2,3]. A bilayer OLED consists of active luminescent layers, hole transporting layers (HTL) and electron transporting layers (ETL) to achieve a balanced charge-carrier injection and enhanced luminescence efficiency [4]. Due to the work function differences between the cathode-metal and organic layer, there is a potential energy barrier that limits electron injection at the cathode-organic layer interface.Several studies have been conducted to investigate the operating principles of bilayer devices [5]. These studies have led to a greater understanding of the role of interfacial barrier heights in achieving current balance despite large differences in hole and electron mobilities and injection barriers at the contacts. Althoughrecombination in bilayer devices takes place predominantly in a region very close to the organic interface, the recombination peak can occur either in HTL or ETL depending on device parameters. The first report on the efficient bright emission was obtained from bilayer OLEDs. The present paper reports the electron-hole recombination near the interface of bilayer OLEDs.

II. THEORY FOR RECOMBINATION PROFILE

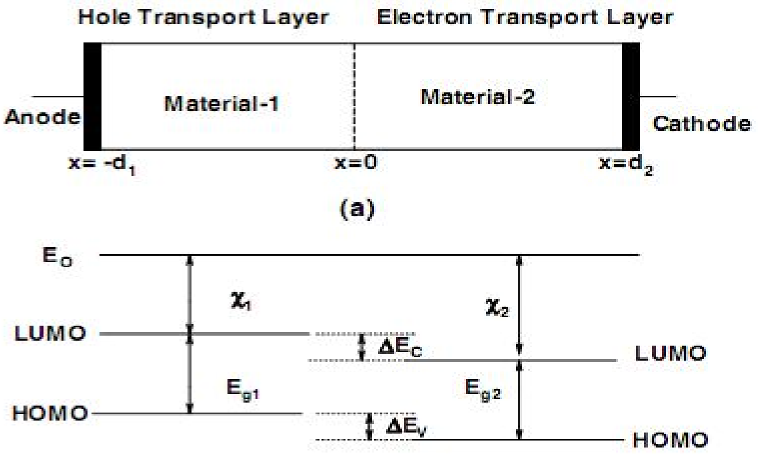
Assuming that, due to the large electric field at the interface, the drift component is predominant, the electron current in HTL at the organic interface can be expressed as

$$J \cong q\mu_{n1}\, E(0)\, n(0^-) \qquad (1)$$

where$J$is the current density,q is the charge of carrier,$E(0)$is the electric field at the organic interface, μ_{n1} is the mobility of electrons in HTL layer and $n(0^-)$ is the density of electrons in HTL layer at the organic interface.

Now, using the relation, $n(0^-) = n(0^+)\exp(-\Delta E_c/kT)$, we can re-write Eq.(1) in the following way

$$J \cong q\mu_{n1} \times E(0) \times n(0^+)\exp(-\Delta E_c/kT) \qquad (2)$$



(b)

Fig.1: (a) Schematic diagram of a bilayer OLEDs and(b) Its associated energy band diagram.

Since the current remains relatively constant with increase in the magnitude of LUMO energy offset, the product of electric field and electron density at the organic interface must increase proportionately according to Eq. (2). To obtain an analytical estimate of the increase in electric field or electron density separately, a relationship between electron density and electric field next to the interface is required. The results from simulations indicate that carrier density decays rapidly away from the interface in a complicated manner [6]. To capture thesharp variation in electron density, we assume forsimplicity that electron density in ETL decays inan exponential manner away from the organicinterface and thus we can write

$$n(x \geq 0) = n(0^+)\exp(-\delta_2 x) \qquad (3)$$

MultiplyEq. (3) by q, we get

$$qn(x \geq 0) = qn(0^+)\exp(-\delta_2 x) \qquad (4)$$

Now, the integration of Eq. (4) net accumulated electron density in ETL close to the interface is given by

$$\int qn(x \geq 0)dx = \int qn(0^+)\exp(-\delta_2 x)\ dx \qquad (5)$$

Using$\int qn(x \geq 0)dx = Q_n^{int.}$in Eq. (5), we get

$$Q_n^{int.} = qn(0^+)\int \exp(-\delta_2 x)dx$$

or,

$$Q_n^{int.} = qn(0^+)\delta_2^{-1} \qquad (6)$$

The electron drift current density J_e(drift)in HTLis given by the expression

J_e(drift)= $nq\mu_e E(0)$ (7) The electron diffusion current density J_e (diff.) in HTL is given by the expression

J_e (diff.) = $qD_e\frac{dn}{dx}$ (8)

where$\frac{dn}{dx}$ is the rate of change of carrier concentration per unit length or concentration gradient.

The accumulation of electrons close to the interface also means that electron drift and diffusion currents must be almost equal and in opposition to each other,thus, from Eqs. (7) and (8), we obtain the following expressions a term of electric field

$$nq\mu_e E(0) = -qD_e\frac{dn}{dx}$$

$$n\mu_e E(0) = -D_e\frac{dn}{dx}$$

$$n\mu_e E(0) = -(\mu_e kT/q) \times (-\delta_2 n)$$

Using the Einstein correlation $D_e = \mu_e kT/q$, where D_e is the diffusion constant, μ_e is the mobility of electron, k is Boltzmann's constant and T is the absolute temperature and we get from Eq. (3) $\frac{dn}{dx} = -\delta_2 n$, and thus Equation

$$E(0) = \frac{kT}{q} \times \delta_2 \qquad (9)\text{or,}$$

$$q = \frac{kT}{E(0)} \times \delta_2 \qquad (10)$$

Substituting the value of q in Eq. (6), then from Eq. (10) we obtain

$$Q_n^{int.} = n(0^+) \times \frac{kT}{E(0)} \qquad (11)$$

Assuming that the accumulation layer in HTL is like a charge-sheet, another expression relating charge and electric field can be obtained through application of Gauss's law at the interface is given by

$$E(0) = \frac{Q_n^{int.}}{\varepsilon} + E_{b2} \qquad (12)$$

where E_{b2} is electric field in the bulk of ETL close to the interface. Substitution of Eq. (11) in Eq.(12) gives the following relationship between electron density and electric field at the organic interface:

$$E(0) = \frac{n(0^+)kT}{\varepsilon E(0)} + E_{b2}$$

or,

$$\varepsilon(E(0))^2 - \varepsilon E_{b2}E(0) - n(0^+)kT = 0 \qquad (13)$$

Equation (13) is a quadratic equation and solving it for the value of $E(0)$ gives

$$E(0) = \frac{\varepsilon E_{b2} \pm \sqrt{\varepsilon^2 E_{b2}^2 - 4\,(\varepsilon)(-n(0^+)kT)}}{2\varepsilon}$$

$$E(0) = \frac{E_{b2}}{2\varepsilon}\left[\epsilon(1 + \sqrt{1 + \frac{4\varepsilon n(0^+)kT)}{\varepsilon E_{b2}^2}}\right]$$

$$E(0) = \frac{E_{b2}}{2}\left[(1 + \sqrt{1 + \frac{n(0^+)}{n_2}}\right]$$

$$E(0) = 0.5\,E_{b2}\left[(1 + \sqrt{1 + \frac{n(0^+)}{n_2}}\right] \qquad (14)$$

where $n_2 = \frac{\varepsilon E_{b2}^2}{4kT}$. Eq. (14) can be written in a form which is more amenable to verification

$$\frac{2E(0)}{E_{b2}} = 1 + \sqrt{1 + \frac{n(0^+)}{n_2}}$$

$$\frac{2E(0) - E_{b2}}{E_{b2}} = \sqrt{1 + \frac{n(0^+)}{n_2}} \qquad (15)$$

Squaring in Eq. (15), we get

$$\left(\frac{2E(0) - E_{b2}}{E_{b2}}\right)^2 = 1 + \frac{n(0^+)}{n_2} \qquad (16)$$

Substitution of Eq. (16) in Eq. (2), gives

$$J \cong 0.5 q\mu_{n1} \times E_{b2} \times \left[\sqrt{1 + \frac{n(0^+)}{n_2}}\right] \times n(0^+)\exp\left({}^{-\Delta E_c}/_{kT}\right) \qquad (17)$$

Now, Eq. (17) can be rewritten as

$$\left(\frac{J}{\exp\left({}^{-\Delta E_c}/_{kT}\right)}\right)^2 \propto n(0^+)^2 \times \left(1 + \frac{n(0^+)}{n_2}\right) \qquad (18)$$

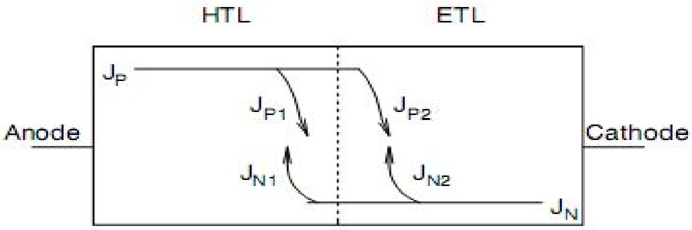


Fig.2: Schematic diagram of a bilayer device illustratingcomponent of electron and hole currents.

An accumulation layer of holes is formed in HTL close to the organic interface due to the presence of a hole barrier. This accumulated hole density can be modeled in a manner similar to the accumulated electron density in ETL and a set of expressions similar to Eqs. (3) – (12) can be written to obtain the following expression relating the electric field to the hole density in HTL immediately to the left of the interface

$$E(0) = 0.5\,E_{b1} \times \left[(1 + \sqrt{1 + \frac{p(0^-)}{p_1}}\right] \tag{19}$$

where $p_1 = \frac{\varepsilon E_{b2}^2}{4kT}$ and E_{b1} is the field in the bulk of HTL close to the organic interface. Similar to the case of electron density injected into HTL, now the hole density injected into ETL can be express as

$$p(0^+) = p(0^-) \times \exp(-\Delta E_V / kT)$$

or

$$p(0^+) = 4p_1 \times [\frac{E(0)-E_{b1}}{E_{b1}/E(0)}]\, p(0^-) \times \exp(-\Delta E_V / kT) \tag{20}$$

For smaller values of interfacial electron barrier height, the current component J_{N1} is dominant and most of the recombination takes place in HTL. However, as discussed earlier, with increase in the barrier height, the hole injection into ETL also increases so that the fraction of electron current carrier by J_{N2} also increases. In light of the above discussions, the ratio of these two currents $J_{N1}$and J_{N2} can be expressed as

$$\frac{J_{N2}}{J_{N1}} \propto \frac{p(0^+)}{n\,(0^-)} \tag{21}$$

The expressions for hole and electron density obtained from Eqs. (20) and(14), respectivelycan be used to rewrite the Eq. (21) and thus, we obtain

$$\frac{J_{N2}}{J_{N1}} \propto \left[\frac{E(0)-E_{b1}}{E(0)-E_{b2}}\right] \times \exp(\frac{\Delta E_C - \Delta E_V}{kT}) \tag{22}$$

As the current components $J_{N1}$and $J_{N2}$are also proportional to recombination rates $U(0^+)$and$U(0^-)$, respectively, an expression relating recombination rates and interfacial barrier heights can be written as

$$\frac{U(0^+)}{U(0^-)} \propto \exp(\frac{\Delta E_C - \Delta E_V}{kT}) \tag{23}$$

III. CONCLUSIONS

The expressions are derived for hole and electron density as the current components $J_{N1}$and $J_{N2}$are also proportional to recombination rates $U(0^+)$ and$U(0^-)$, respectively, expression relating recombination rates and interfacial barrier heights is obtained for bilayer OLEDs, which clearly indicates the role of interfacial barriers in the control of recombination currents in bilayer OLEDs. It is to be noted that the recombination currents are responsible for the brightness of OLEDs.

IV. ACKNOWLEDGEMENT

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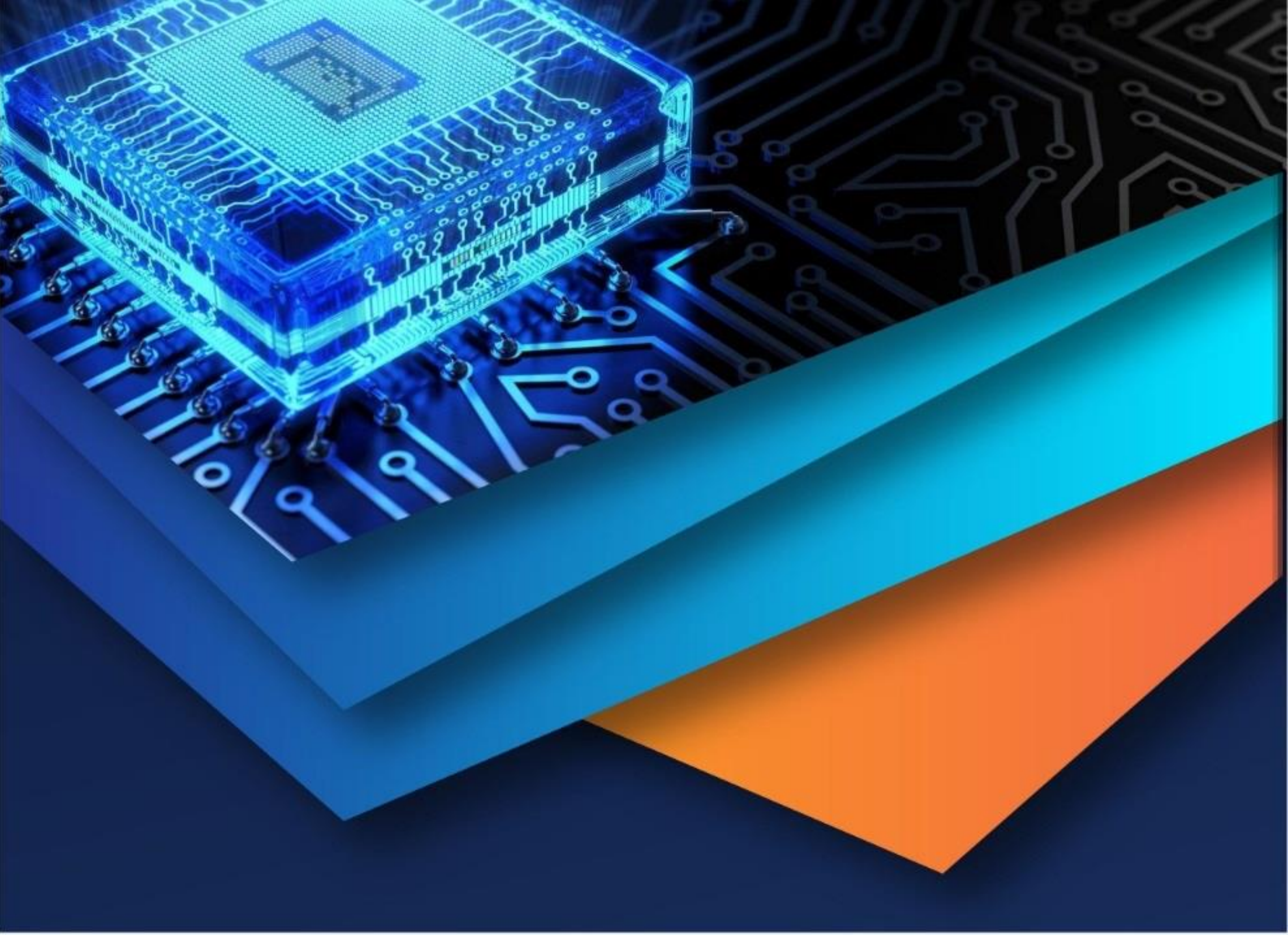

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