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Effect of the applied bias voltage on exciton profile and efficiency roll-off in organic light emitting diodes

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Abstract- Despite organic light emitting diode (OLED) devices reach more than 60% external quantum efficiency and in thermal activated delayed fluorescence (TADF) type ones, reach near 100%, efficiency roll-off in higher bias voltage due to the excitation quenching and annihilation is unsolved. Numerical simulation helps researchers to find fundamental parameters in excitonic profile of OLED in higher bias voltage. In this paper, we present an electro-optical modeling of single layer phosphorescent OLED to describe excitonic processes and loss mechanisms. We investigated electro-optical processes include radiative and non-radiative recombination of excitons, triplet-polaron quenching, triplet-triplet annihilation. Simulation results show triplet-triplet annihilation at high bias increases due to the quadratic relation with exciton density and the contribution of triplet-polaron quenching decreases due to the larger effective electron mobility than the hole mobility at high bias voltage. The triplet-triplet annihilation process is the main reason for decreasing the radiation efficiency and efficiency roll-off at high bias voltage which is in good agreement with published experimental data. The results show how OLED efficiency and radiative recombination of excitons decrease due to the roll-off process.

Keywords: organic light emitting diode (OLED); excitonic processes; efficiency roll-off.

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1. Introduction

Efficient and low-cost production, physical flexibility, and non-toxic property of organic semiconductors make them a good alternative to common inorganic semiconductors devices [1]. In recent years, the development of high efficiency organic luminescent semiconductors makes them the best candidate for modern applications. Organic light emitting diode (OLED) devices became key devices in modern optoelectronic devices, from displays to organic laser and lightning [2]. Phosphorescent OLEDs reach more than 60% external quantum efficiency (EQE) and newly introduced thermal activated delayed fluorescence (TADF) type, reach near 100 EQE [3]. Despite high-efficiency OLED devices, decreasing device luminous efficiency due to the loss mechanisms in high bias and high injection current is a major challenge. Loss mechanisms that cause efficiency roll-off categorized two main processes: losses of excitons and losses of radiative photons. Losses of excitons such as triplet-polaron quenching (TPQ) and triplet-triplet annihilation (TTA) lead to a considerable decrease in the high current density [2,3]. As mentioned, excitonic processes play a key role in the OLED device working principle, and study these processes helps researchers to develop high efficiency devices [4]. To describe the main features of charge transport in organic LEDs four processes have to be considered as illustrated in the schematic energy level diagram in Figure 1.

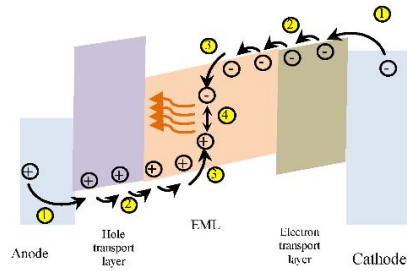


Fig. 1. Schematic energy level diagrams of the OLED with four main processes.

In the step (1), charge carriers have to be injected into the organic material from anode and cathode, in the step (2) they will be transported, and in the step (3) they recombine to an exciton and then, the excitons decay radiatively or non-radiatively (4). The radiative decay of such an entity or following its evolution successive excited states produces light called recombination radiation. Main excitonic processes occur in the emission layer (EML) and in this study, we investigated the excitonic and charge transport process in EML. Although lots of experimental progress have attained in the development of new materials and device architecture; numerical simulation methods have received much attention includes both electrical and optical modeling via, Monte Carlo, Finite Element Methods (FEM), etc. [5]. In this study, by electro-optical modeling, excitonic processes are described by solving the exciton and charge continuity equations. The FEM has been taken into account in calculations.

2. Models and Methods

We used the following exciton continuity equation [5]:

$$\frac{d}{dt}T(t, x) = G.R(t, x) + \frac{d}{dx}J_s(t, x) - F(x)k_{rad}T(t, x) - k_{nonrad}T(t, x) - k_{TPQh}T(t, x)p(t, x) - k_{TPQe}T(t, x)n(t, x) - k_{ann}T(t, x)^2 \quad (1)$$

Where $G.R(t, x)$ is the generation term for the time (t) and position (x) dependent triplet exciton density $T(t, z)$, which is calculated from the exciton formation ratio $G = 1$ and the Langevin recombination rate R in the EML. We studied only the triplet exciton equation because any singlet excitons quickly relax to triplet states. $J_s(t, x)$ is the exciton diffusion term, and $F(x)k_{rad}T(t, x)$ describes radiative recombination of excitons with a rate constant k_{rad} modified by the position-dependent Purcell factor $F(x)$. The term $k_{nonrad}T(t, x)$ represents non-radiative recombination with a rate constant k_{nonrad} . The terms $k_{TPQh}T(t, x)p(t, x)$ and $k_{TPQe}T(t, x)n(t, x)$ are the position-dependent triplet-polaron quenching terms, depending on the hole- and electron density with k_{TPQh} and k_{TPQe} being the respective rate constants $k_{ann}T(t, x)^2$ is the position-dependent triplet-triplet annihilation term. The current continuity equations are:

$$\begin{aligned} \frac{d}{dt}n(x) &= \frac{1}{e} \frac{d}{dx}J_n(x) - R(x) \\ \frac{d}{dt}p(x) &= \frac{1}{e} \frac{d}{dx}J_p(x) - R(x) \end{aligned} \quad (2)$$

In these equations, J_n , J_p are electron current density, hole current density, respectively. In our study, we solve these equations via FEM by applying the boundary conditions and 35 nm active layer thickness.

3. Results and Discussion

Fig. 2 (a) shows the applied voltage dependence of the above-mentioned parameters in steady-state conditions in EML layer. As expected, by increasing the applied bias voltage, exciton generation (black line or GR) increases. Following total exciton's contribution to triplet-polaron quenching of holes (dashed magenta line $k_{TPQH}T(t, x)p(t, x)$) in lower voltages and strongly bias-dependent triplet-triplet annihilation ($k_{ann}T(t, x)^2$). They followed by radiative recombination (solid green line). Non-radiative recombination (blue line) and electrons triplet-polaron quenching with ($k_{TPQE}T(t, x)n(t, x)$) are only minor contributions to the exciton balance.

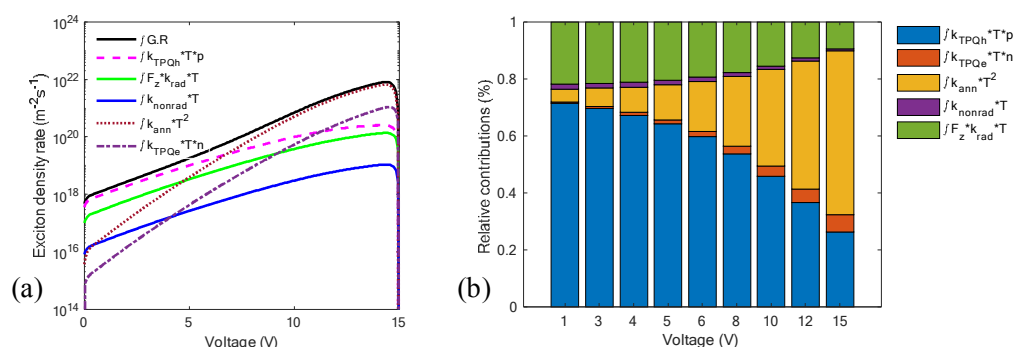


Fig. 2: (Color online) (a) Integrated terms of the exciton continuity equation for different bias voltages and (b) relative contribution of each process.

For a better investigation of excitonic processes, the relative contribution of each process in terms of bias voltage in EML layer has shown in Fig. 2 (b). In OLED devices, Due to the larger field-dependence of mobility for electrons than for holes, the effective electron mobility is larger than the hole mobility, and therefore, the contribution of triplet-polaron quenching decreases. Contribution of triplet-triplet annihilation losses at high bias voltage increases due to the quadratic relation with exciton density ($\propto T^2$) and this process is the main reason for decreasing the radiation efficiency and efficiency roll-off at high bias voltage which is in good agreement with published experimental data [4]. The results show how OLED efficiency and radiative recombination of excitons decrease due to the roll-off process.

4. Conclusion

In conclusion, the influence of the applied bias voltage on the exciton profile and efficiency roll-off in high voltages in organic light emitting diode has been investigated. In this study, by electro-optical modeling, excitonic processes are described by solving the exciton and charge continuity equations and the FEM has been taken into account in our calculations. Numerical simulation results show triplet-triplet annihilation at high bias increases due to the quadratic relation with exciton density and the contribution of triplet-polaron quenching decreases due to the larger effective electron mobility than the hole mobility at high bias. The triplet-triplet annihilation process is the main reason for decreasing the radiation efficiency and efficiency roll-off at high bias voltage which is in good agreement with published experimental data. The results show how OLED efficiency and radiative recombination of excitons decrease due to the roll-off process.

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