# Drift-diffusion simulations of thermally activated delayed fluorescence OLEDs

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***Abstract*—In this work we present drift-diffusion simulations of a blue-emitting TADF OLED. We exploited the ability of the multi-particle drift-diffusion model to explicitly calculate the charge transport for the different sub-populations of the TADF emitter, including the singlet and triplet exciton states. The inclusion of proper models concerning the band-to-band transition, the most bimolecular recombination mechanism and above all the inter-system crossing, allowed to investigate the device operation under different conditions. Moreover we conclude on the role that TTA and TPQ losses have over the IQE roll-off effect that typically occurs in TADF OLEDs.**

## I. Introduction

To date, organic light emitting diodes (OLEDs) represent the most promising and attractive technology for full-colour displays and lighting application. Their success is due to significant features as large emitting area, ultra thin and flexible substrate compatibility, offering high colour quality and contrast ratio at low power consumption, unlike conventional LEDs. Efficient OLEDs at least three important issues have to be guaranteed: (i) low operation voltage, (ii) high internal quantum efficiency (IQE) and (iii) high light extraction efficiency (LEE). Since the first OLED fabrication by Tang and Van Slyke[1], over the years, the research has been focused on the device design and performance improvement. In first-generation OLEDs based on fluorescent emitters the ideal IQE was 25% due to the limitation imposed by spin statistics on singlets and triplets ratio (1:3). Second-generation OLEDs based on phosphorescent emitters, instead, they could ideally reach IQE=100% by exploiting the high spin-orbit coupling (SOC) given by the inclusion of organometallic complexes.

The use of the RGB technology in OLED displays makes the the blue emitters design definitely the most significant issue, because they require wide bandgaps, and exhibit limited lifetime and stability problems[2]. In 2012, Uoyama et al.[3] demonstrated that the use of thermally activated delayed fluorescence (TADF) materials can successfully overcome these limitations. TADF OLEDs require a $\Delta E_{ST}$<100meV between exciton states of fluorescent material, and can ideally harvest both singlets ($S$) and triplets ($T$) allowing IQE=100% and a EQE of nearly 20%. Under long triplets decay lifetime condition, a reverse inter-system crossing (RISC) mechanisms takes place in TADF material promoting $T$ into $S$ states. As result, the radiative emission will be fostered by delayed fluorescence in addition to the prompt emission. Given the complexity of mechanisms involved, a versatile and fast simulation tool is crucial for device design and optimization.

## II. The multi-particle drift-diffusion model

We use a multi-particle drift-diffusion model (mp-DD)[4] capable to calculate the transport explicitly accounting for both charged carriers and excitons. The model is based on the coupling of Poisson (1a) and transport equations (1b), and allows to define any number of carriers, each one with its own properties as charge, spin and DOS.

$$\nabla \cdot (\varepsilon_0 \varepsilon_r \nabla \varphi) = -q \sum_i z_i n_i + qC \tag{1a}$$

$$\nabla \cdot (\mu_i n_i \nabla \phi_i) = R_i \ , \ \forall i. \tag{1b}$$

The modeling approach is based on the generalization to more than two carrier populations ($n_i$), each individually assumed in a local thermal equilibrium by local quasi-Fermi levels ($\phi_i$), and a strictly thermodynamically consistent formulation of the transitions between populations, which are implemented as recombination-generation terms ($R_i$). The full system is discretized with finite elements method (FEM) and solved by the Newton method, as implemented in TiberCAD simulation tool.

## III. Results and Discussion

In order to model the blue-TADF OLED operation we perform simulation DD simulations on the structure model depicted in Figure 1, where the emitter (EML) consists of a host-guest (80:20) system matrix. Simulations explicitly account for electron and hole sub-populations, and for localized exciton states in the EML. Here, we place $E_S^g = 2.56eV$ and $E_T^g = 2.49eV$ for guest, and $E_T^h = 2.84eV$ for host. Notably, the resulting $\Delta E_{ST}$=70 meV of guest guarantees the TADF mechanism activation. According with organic materials features, we provide molecular disorder using the Gaussian model for each charged and neutral sub-population of the system.

The exciton generation is controlled by constant rates of $10^{-31}cm^3s^{-1}$ and $10^{-30}cm^3s^{-1}$, respectively in host and guest material, and guaranteeing the (1:3) ratio. Consistently with the TADF OLED typical operation, we set non-radiative and radiative exciton decay lifetimes of $5 \times 10^{-5}s^{-1}$ and $1 \times 10^{-8}s^{-1}$. Simulations performed are aimed to investigate the IQE roll-off which typically takes place at high current density[5]. To do this, we include in the model charge transfer

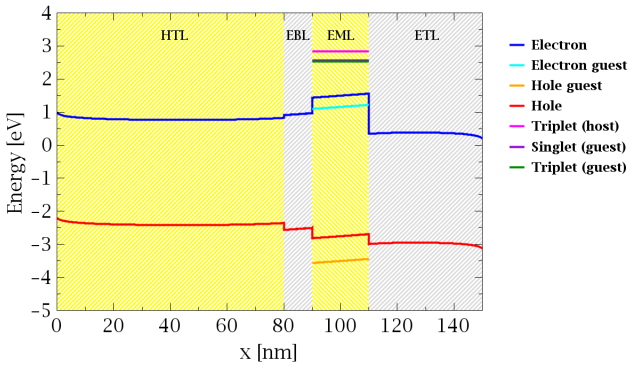


Fig. 1. LUMO and HOMO of carrier sub-populations, and average exciton state energy levels included in the TADF OLED system simulated.

between HOMOs and LUMOs levels in EML, and the RISC between exciton states in guest as follow

$$R_{nn^*} = C_{nn^*} n \left(1 - \frac{n^*}{N^*}\right) \left[1 - \exp\left(\frac{E_{f,n^*} - E_{f,n}}{k_B T}\right)\right] \tag{2a}$$

$$R_{ISC} = C_{ISC} n_S \left(1 + \frac{n_T}{N_T}\right) \left[1 - \exp\left(\frac{E_{f,T} - E_{f,S}}{k_B T}\right)\right]. \tag{2b}$$

Equation (2a) models the charge transfer between the host-guest system energy levels taking into account of the different electrochemical potentials ($E_{f,n}$ and $E_{f,n^*}$) and available states $N^*$. Equation (2b) models the ISC, where the crossing direction (direct or reverse) depending on $E_{f,T}$ and $E_{f,S}$. We estimate the RISC contribution to the internal radiative emission of EML simulating the device with $C_{ISC}$=$10^{-13} cm^3 s^{-1}$ and then neglecting it.

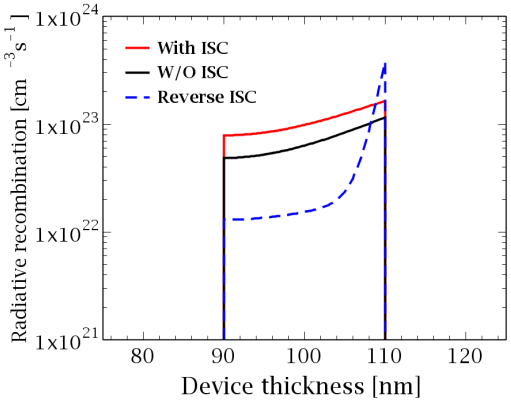


Fig. 2. Radiative emission profile either accounting and not the ISC (dashed-blue) at 6 V operation.

Figure 2 shows that the RISC increase the average radiative emission of 65%, with a peak contribution at EML/ETL interface where $E_{f,T} - E_{f,S}$ is significantly reduced. In addition we observe that the mp-DD model is capable to calculate the radiative emission profile taking into account the different charge carrier injection from EBL/EML and EML/ETL interfaces. By including the most important bi-molecular degradation processes that affect the luminescent efficiency, such as triplet-to-triplet annihilation (TTA)[5] and triplet-to-polaron quenching (TPQ)[6], we estimated the IQE at different current density operation.

$$R_{TTA} = C_{TTA} n_T^2 \left[1 - \exp\left(-\frac{2E_{f,T}}{k_B T}\right)\right] \tag{3a}$$

$$R_{TPQ} = C_{TPQ} n_T n_Q \left[1 - \exp\left(-\frac{E_{f,T}}{k_B T}\right)\right]. \tag{3b}$$

Equation (3a) models the interaction between two triplets ($n_T{}^2$) leading to the loss of one of them, while Eq. (3b) the triplet vanishing interacting with a polaron quencher, i.g. holes.

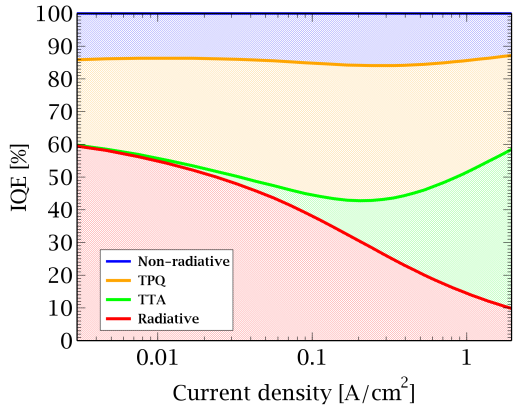


Fig. 3. Quantum efficiency contributions from non-radiative recombination, TPQ, TTA and internal radiative emission at different current densities.

In Figure 2 we report the plot of all contributions to the quantum efficiency calculated under different operation conditions. According with experimental observations in TADF OLEDs[5], TTA losses determine the IQE roll-off at high current densities. This effect is due to the increasing significantly of triplets density at high current density, which mainly reduce the triplet-to-singlet recycling that normally occurs via RISC.

## References


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