Development of 2D carrier transport model for random dopant effect and exciton coupling model

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***Abstract*—A novel 2D simulation solver for doped OLEDs, incorporating a random doping model, overcomes 1D constraints. Validated experimentally, it shows low doping creates current-limiting traps (via deeper guest levels) rather than altering mobility. The model pinpoints EML/ETL recombination and identifies host triplet-triplet annihilation and guest hole-polaron quenching as key losses at high currents, offering routes for OLED optimization.**

Keywords—OLED, 2D random dopant model, Carrier Transport, Exciton Diffusion, Efficiency Optimization

I. INTRODUCTION

Organic optoelectronic materials are foundational to modern display, lighting, and communication technologies. However, a persistent challenge limiting their emission efficiency is exciton spin statistics. In conventional closed-shell materials, electrical excitation predominantly generates non-emissive triplet excitons (75%) over emissive singlet excitons (25%), as only the latter undergo spin-allowed radiative decay [1]. While thermally activated delayed fluorescence (TADF) and phosphorescent materials offer pathways to harness triplet excitons, they often contend with issues like long exciton lifetimes, significant efficiency roll-off, and complex fabrication [1, 2].

An innovative approach involves open-shell radical materials, which feature doublet ground and excited states. This configuration permits rapid, spin-allowed radiative decay via $D_1 \rightarrow D_0$ transitions [2]. Crucially, recent advancements have demonstrated that in well-designed host–guest systems, triplet excitons generated in the host can be efficiently converted to doublet excitons in the radical emitter through a process known as triplet-to-doublet energy transfer [3-7]. This mechanism effectively bypasses the spin blockade.

The process itself can involve distinct energy transfer pathways: Förster resonance energy transfer (FRET), typically mediating $S_1 \rightarrow D_1$ transitions and dependent on spectral overlap and dipole-dipole coupling, and Dexter energy transfer, facilitating $T_1 \rightarrow D_1$ transitions through direct orbital overlap and energy level alignment [3, 4]. Since the $D_1 \rightarrow D_0$ decay in doublet excitons is spin-allowed, the circumvents the need for intersystem crossing (ISC) or reverse ISC (RISC). Combined with the nanosecond-scale lifetimes of radical excitons, this leads to significantly faster emission kinetics than traditional TADF materials [5, 6, 8], making the luminescence mechanism highly attractive for applications demanding rapid responses, such as optical communication [4, 7].

Detailed kinetic modeling of these exciton transitions is essential to fully understand and optimize the efficiency of these systems and elucidate the dominant pathways and rate-limiting steps. This study will present a comprehensive model to simulate these dynamic processes.

II. METHODOLOGY

A: Carrier transport:

Our laboratory developed a finite element method to solve the 2D Poisson and drift-diffusion equations at steady state, a widely used approach for modeling the electrical characteristics of organic light-emitting diodes (OLEDs). In this study, we combine the Poisson-drift-diffusion equations with a Gaussian density of states (DOS) model and Poole-Frenkel field-dependent mobility to simulate carrier transport in OLED host-guest systems, incorporating the effects of random doping. To address the influences of doping effect, a 2D random doping model is used [9]. In this approach, the doping region is divided into small grid units to represent the positions of molecules. The Gaussian weighting method is used to determine it dopant concentration.

A: Exciton diffusion and energy transfer model for singlets, triplets, and doublets:

When electrons and holes recombine, they form excitons, which can then diffuse and decay within the material. In this study, the emission mainly relies on the conversion of triplets to doublets, with triplet excitons making up 75% of exciton generation. The exciton dynamics are modeled by the following diffusion equation:

$$\frac{dS}{dt} = D^S\nabla^2 S - (k_r^S + k_{nr}^S + k_e^S n + k_h^S p + k_{ISC} + k_{FRET})S - k_{RISC}T + \alpha G_S \qquad (1)$$

$$\frac{dT}{dt} = D^T\nabla^2 T - (k_r^T + k_{nr}^T + k_e^T n + k_h^T p + k_{RISC} + k_{DET})T - k_{TT}T^2 + k_{ISC}S + \beta G_T \qquad (2)$$

$$\frac{dD}{dt} = D^D\nabla^2 D - (k_r^D + k_{nr}^D + k_e^D n + k_h^D p)D - k_{DD}D^2 + k_{FRET}S + k_{DET}T + (1-\alpha-\beta)\,G_D \qquad (3)$$

Here, S, T and D represents singlet, triplet and doublet exciton density, D is the exciton diffusion coefficient, and γ is the exciton annihilation coefficient. α and β are the generation fraction coefficients of excitons. The term G corresponds to the initial distribution of exciton density. Additionally, n and p refer to the densities of electron and hole carriers, respectively.

III. RESULT AND DISSUSION

To validate the accuracy of the proposed model, OLED devices, as well as electron-only (EOD) and hole-only (HOD) devices with a 3% doping concentration, were fabricated by

Prof. Cho. The devices utilized CzDBA as the host material and TPA-PyBTM' as the guest material. The experimental results demonstrate that at low doping concentrations, the guest molecules introduce trap states that hinder carrier transport, reducing current density. This observation is consistent with the simulated result presented in Fig. 1(a–c). The decrease in current density is primarily attributed to the formation of deeper energy states by the guest material rather than a direct change in charge carrier mobility.

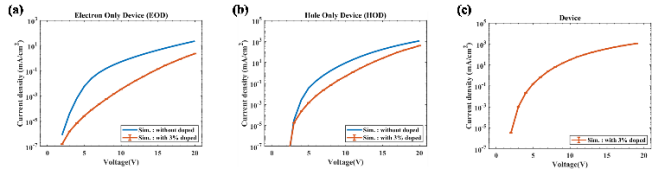

Fig. 1. (a), (b), and (c) are EOD, HOD and OLED, respectively, along with the J-V curves for different cases with 3% dopant concentration.

Figures. 2(a) and 2(b), show that the current density decreases as the doping concentration increases from 0% to 3%. In contrast, when the doping concentration exceeds 3%, the current density begins to increase. This trend can be attributed to the energy gap between the LUMO or HOMO levels of the host and guest materials, which affects carrier injection and transport efficiency.

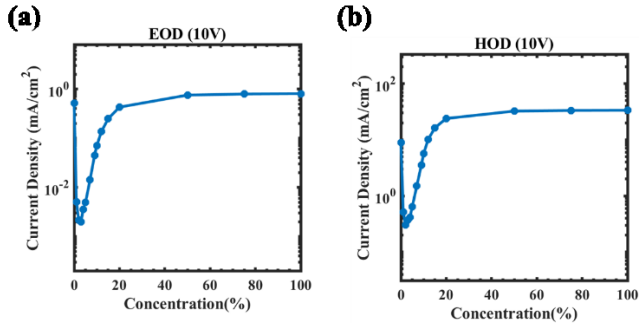

Fig. 2. (a-d) The current density under voltages of 5V and 10V for electron-only and hole-only devices.

Owing to the low dopant concentrations, the guest material becomes isolated. As a result, carriers passing through these localized guest states are more likely to become trapped. This behavior is clearly illustrated in Fig. 3(b) and 3(c), where clusters of high carrier density indicate the presence of trap sites. Such trapping impacts the charge balance and recombination efficiency within the device.

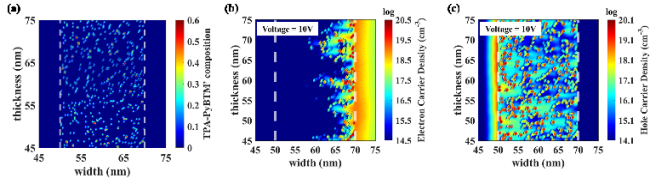

Fig. 3. (a) The composition of the EML with random doping. (b) and (c) are electron and hole carrier density distribution at 10 V.

Fig. 4(a) and (b) illustrate the two-dimensional distribution of the radiative recombination region under different applied voltages, providing an initial indication of exciton generation locations. As the voltage increases from 5 V to 10 V, a clear trend is observed: the recombination region becomes more concentrated and is primarily located at the interface between the emission layer (EML) and the electron transport layer (ETL).

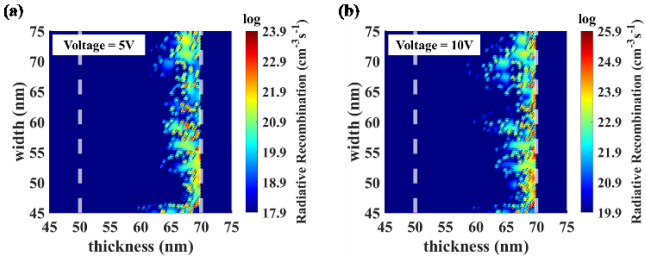

Fig. 4. (a) and (b) are radiative recombination at 5.0 V and 10.0 V, respectively.

To gain deeper insight into the spatial dynamics of exciton behavior under high-bias conditions, the exciton concentration and various loss processes were mapped at 10 V, as shown in Fig. 5, which further highlights the diffusion of excitons from the host to the guest material, where energy transfer plays a crucial role in determining emission efficiency. The loss mechanism in this system is relatively complicated, as it is decided by doublet and triplet exciton carrier quench mechanism. The details will be discussed at this conference presentation.

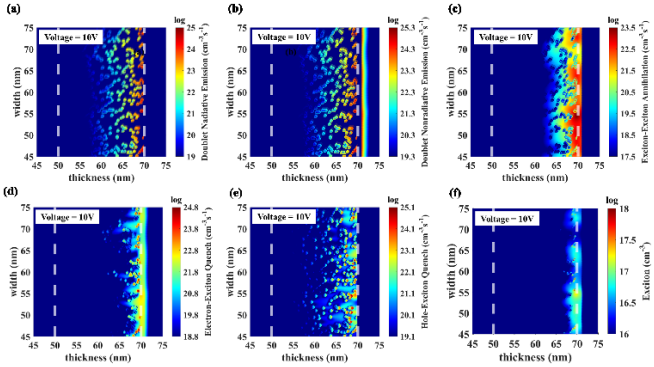

Fig. 5. (a-f) Two-dimensional distributions distribution of exciton density, radiative emission, nonradiative emission, annihilation, electron quenching, and hole quenching at 10.0 V.

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