Development of time-dependent Exciton diffusion solver for modeling Triplet-Triplet Fusion Mechanism in OLEDs

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***Abstract*—In this work, we developed a both stable and time-dependent exciton diffusion model including singlet and triplet exciton coupled with a modified Poisson & drift-diffusion solver to demonstrate the mechanism of triplet-triplet fusion (TTF) OLEDs. Using this modified simulator, we can demonstrate the characteristics of OLEDs including current-voltage curve, quantum efficiency performance, time-resolved electroluminescence spectrum, and electric profile...etc. Also, this solver can be used to explain the mechanism of hyper-TTF OLEDs, and analysis the loss from different exciton mechanism. Furthermore, we can do further optimization for TTF-OLEDs to achieve an internal quantum efficiency increasing of 23% (from 29% to 36%).**

***Index Terms*—Poisson-Drift-Diffusion, TTF-OLEDs, Device Modeling, Organic Material, Exciton Diffusion Model**

I. INTRODUCTION

Since Prof. Tang fabricated the first organic light-emitting diodes (OLEDs), OLEDs have attracted the attention of researchers and consumers , because OLEDs devices are able to emit light which is the most similar to natural light [1]. Recently, several OLEDs-based electronics (TV, smart phone, wearable devices, ...etc) have a great achievement on consumer electronics. But the efficiency of OLEDs is still lower than traditional solid-state lighting devices, especially blue OLEDs.

The candidates of the next generation OLEDs have in common utilizing both of singlet and triplet exciton as much as possible. Hence, TTF-OLEDs is regarded as the most potential candidate for next generation blue-OLEDs [2]. Figure 1 shows the process of TTF-OLEDs, this kind of OLEDs utilizes singlet exciton to emit prompt fluorescence (PF), next stage is that the triplet-triplet annihilation upconversion (TTA-UC) would be triggered, and then the singlet converting from TTA-UC process would emit delayed fluorescence (PF).

However, there is still concern in TTF-OLEDs, the main reason is that it's really hard to fully utilize both singlet and triplet exciton to emit light, because triplet exciton which has longer lifetime is likely to be annihilated or quenched by polaron and singlet exciton in emitting layer (EML) after triplet exciton transforming to singelt exciton. In order to overcome this problem, a TCAD program which can demonstrate exciton interaction behavior is needed. In this work, a complete exciton diffusion model considering both singlet and triplet exciton and their interaction (triplet singlet annihilation, TSA and triplet-triplet annihilation, TTA and TTF) is developed to demonstrate the performance of TTF-OLEDs.

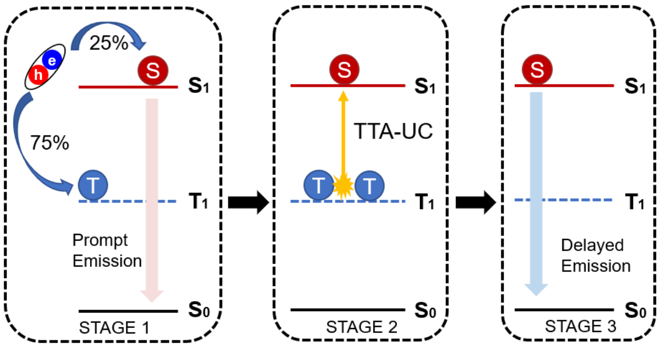

Fig. 1. The schematic illustration of mechanism of triplet-triplet fusion OLEDs emission process from initial stage to final stage.

II. METHODOLOGY

In this work, the simulation program is based on Poisson-DD solver which can demonstrate organic-based devices by using a Gaussian density of state and filed dependent mobility model, the detail can refer our previous work [3]. And exciton diffusion model considering most exciton behavior and the interaction of singlet and triplet exciton is developed in this study. The exciton diffusion model supports stable and time-dependent modes, the stable solver can be used to calculate the internal quantum efficiency (IQE) of device. And time-dependent model can be utilized to demonstrate some transient spectrum of device, which can extract the parameters of exciton by calibrating with experimental data.

To demonstrate the performance of TTF-OLEDs, considering both of singlet and triplet excitons is needed. Singlet and

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triplet exciton diffusion equations are shown in the following:

$$\frac{dn_{ex}^{S}}{dt} = D_{ex}^{S}\nabla_r^2 n_{ex}^{S} - \left(k_r^S + k_{nr}^S + k_e^S n + k_h^S p + k_{TS} n_{ex}^{T}\right) n_{ex}^{S} + \frac{1}{2}\gamma n_T^2 + G_{ex}^{S}, \tag{1}$$

$$\frac{dn_{ex}^{T}}{dt} = D_{ex}^{T}\nabla_r^2 n_{ex}^{T} - \left(k_r^T + k_{nr}^T + k_e^T n + k_h^T p + k_{TS} n_{ex}^{S}\right) n_{ex}^{T} - \gamma n_T^2 + G_{ex}^{T}, \tag{2}$$

where n_{ex} is the exciton density distribution, D_{ex} is exciton diffusion coefficient, k_r and k_{nr} are the radiative and non-radiative exciton quenching constants, respectively, k_e and k_h are the TPQ rate constant for electrons and holes, respectively, and G_{ex} is the initial exciton density distribution. And superscript (S,T) represents singlet and triplet excitons, respectively. γ is the exciton annihilation coefficient, n and p are the carrier density of electron and hole, respectively.

III. RESULT AND DISCUSSION

In this work, the characteristics of three different devices are demonstrated by our solver, and the experimental data is referred from this study [4]. Figure 2(a) shows the normalized quantum efficiency calculated by this solver. As shown in figure 2(b), our solver also can demonstrate the loss from different mechanism including electron-, hole-induced TPQ, and TSA, which can be utilized to find the way to optimize the performance of device. Also, our solver can demonstrate the time-resolved electroluminescence spectrum which is shown in figures 2(c)-(d), which can be applied to extract and fit the parameters used in exciton diffusion model by comparing with experimental data.

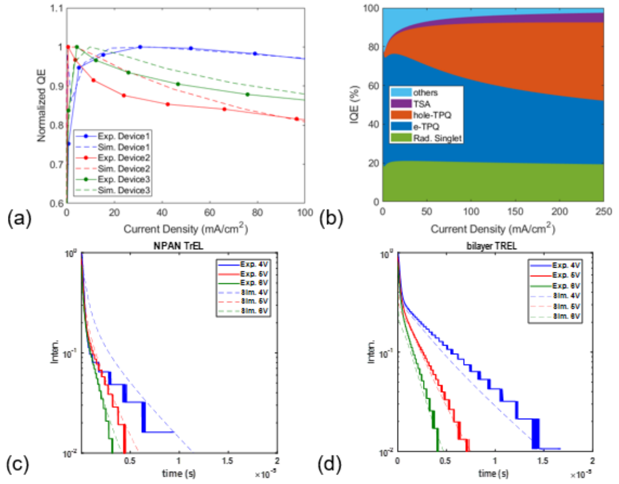

Fig. 2. (a) The normalized quantum efficiency curves for device A, B, and C. (b) The loss results from radiative singlet exciton, electron-induced TPQ, hole-induced TPQ, and TSA. (c)-(d) The transient electriclumiance spectrum for device B, and device C, respectively. The solid- and dash- lines are experimental and simulated results, respectively.

According to the modeling results which we have mentioned before, a significant loss resulting from both electron- and hole-induced TPQ can be observed. Hence, the optimized properties of TTF layer is implemented to improve the performance of device. To reduce the electron- and hole-induced TPQ, we try to demonstrate the different properties of materials including carrier mobility, band alignment of device, and Dexter energy transfer of exciton. Figure 3 shows that the IQE versus current density for cases of reference and optimized device. The maximum efficiency can be increased from 29.5% to 36.3%, which can achieve a 23% of improvement. And figure 3 shows that the losses from different mechanism, the reason of efficiency improvement is because these changing can reduced both electron- and hole-induced TPQ, which can achieve a higher delayed singlet emission.

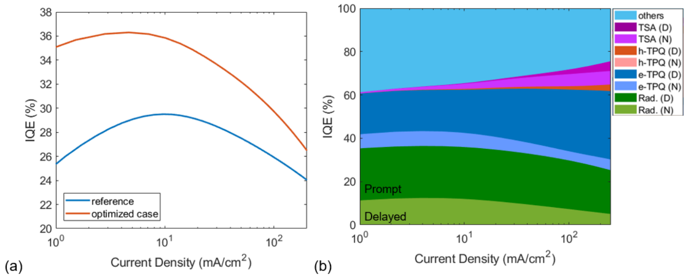

Fig. 3. (a) The IQE curves for reference and optimized cases. (b) The loss results from radiative singlet exciton, electron-induced TPQ, hole-induced TPQ, and TSA for optimized case.

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REFERENCES

[1] Tang, C. W., VanSlyke, S. A. (1987). Organic electroluminescent diodes. Applied physics letters, 51(12), 913-915.

[2] Lee, J. H., Chen, C. H., Lee, P. H., Lin, H. Y., Leung, M. K., Chiu, T. L., Lin, C. F. (2019). Blue organic light-emitting diodes: current status, challenges, and future outlook. Journal of Materials Chemistry C, 7(20), 5874-5888.

[3] Huang, J. Y., Lee, J. H., Chen, T. Y., ... Wu, Y. R. (2020). Revealing the mechanism of carrier transport in host-guest systems of organic materials with a modified Poisson and drift-diffusion solver. Physical Review Materials, 4(12), 125602.

[4] Chen, C. H., Lee, P. H., Lin, H. Y., Lin, B. Y., Leung, M. K., Chiu, T. L., Lee, J. H. (2022). Hyper triplet-triplet fusion blue fluorescent organic light-emitting diode.