



# Tuning magneto-electroluminescence in organic light emitting diodes by controlling the competition between singlet fission and triplet fusion



H. Liu, W.Y. Jia, Y. Zhang, Q.M. Zhang, Y.L. Lei, C.L. Lu, Y.Z. Ling, Z.H. Xiong\*

School of Physical Science and Technology, MOE Key Laboratory on Luminescence and Real-Time Analysis, Southwest University, Chongqing 400715, PR China

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## ABSTRACT

We report a tunable magneto-electroluminescence (MEL) in doped organic light-emitting diodes (OLEDs) with narrow energy gap molecule of 4-dicyanomethylene-2-t-butyl-6-1,1,7,7-tetramethyljulolidyl-9-4H-pyran (DCJTB) doped rubrene as active layers. It is noted that the MEL at 500 mT alters significantly from 19.3% to -1.0% by controlling the doping concentration of DCJTB in the active layer at room temperature. The results might be caused by the conversion from singlet fission to triplet fusion depending on the competition between energy transfer and charge trapping channels in doped layer. Moreover, the MEL can also be tuned by changing the working temperatures for the devices with certain doping concentration. Therefore, this work provides a feasible pathway to tune the MEL by controlling the competition between singlet fission and triplet fusion in OLEDs.

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## 1. Introduction

Since the magnetic field effects (MFEs) on luminescence and conductance were found in nonmagnetic organic semiconducting materials and devices [1], they have attracted tremendous scientific research interest [2–25]. As an important part, magneto-electroluminescence (MEL), the relative changes of electroluminescence induced by an applied magnetic field, has been a powerful tool to reveal the excited processes occurring in organic optoelectronic devices [2–19]. For engineering purpose, tuning the MEL signal by changing the internal microscopic excited process is promising for realizing the practical application of controllable MFEs in organic devices. Previously, Chen et al. [3] and Wu et al. [4] had investigated the variation of MEL after adjusting the density of triplet excitons and mixing the molecules with different spin-orbit coupling strengths, respectively. However, the ways of tuning MEL is still not enough, and seeking more practical methods is still a valuable work for development of integrated magnetic-optic-electronic multifunctional devices.

Singlet exciton fission (SF) [5–12] and triplet exciton fusion (TF) [12–20] are two important processes in organic devices. Both of them have clear impacts on the electroluminescence (EL) of the organic light-emitting diodes (OLEDs) [5–19]. Apparent SF process will seriously reduce the light-emission of device since it constitutes an ultrafast decay pathway for singlet excitons [5–7]. In contrast, the EL intensity of the OLED would be effectively enhanced if strong TF is able to occur in the emission layer [7, 15–19]. Both SF and TF are suggested to be magnetic field dependent. They are reversal processes for each other [7]. Thus, the MEL of the devices would be affected by the competition between SF and TF.

In this work, red light-emission dye molecule DCJTB was doped into rubrene layer in order to realize the coexistence of SF and TF in a single device. And the other process like intersystem crossing would be too slow to happen due to its large time scale ( $10^{-8}$ – $10^{-3}$  s) compared with that of SF ( $10^{-14}$ – $10^{-11}$  s) [26], which will not be concerned in this work. Energy match is very important in the excitonic interaction, SF requires  $E_S \geq 2E_T$ , while TF requires  $E_S \leq 2E_T$ , where  $E_S$  and  $E_T$  represent the singlet and triplet energy levels, respectively. In rubrene, the singlet and triplet energy are in resonance, i.e., ( $E_S \approx 2E_T$ ) [5,7,13,21,27], which is fit for that SF requires. What is more, it has been estimated that the ~90% of the singlet excitons undergo SF at room temperature [6]. When DCJTB

\* Corresponding author. Tel.: +86 23 68254377.

E-mail address: [zhxiong@swu.edu.cn](mailto:zhxiong@swu.edu.cn) (Z.H. Xiong).

molecules were mixed in rubrene layer, the excitons and charge carriers would transfer from rubrene to DCJTB through two channels including energy transfer (ET) and charge trapping (CT) [22], resulting in the luminescence changing from rubrene to DCJTB. Consequently, the SF occurring on rubrene molecules will be suppressed whereas the TF on DCJTB molecules will be strengthened. Therefore, controlling the doping concentration of DCJTB is able to modulate the competition between the SF and TF, and then obtain the tunable MEL. Besides, the MEL can also be tuned by changing the operating temperatures for a certain doping concentration device. Moreover, each device with different doping concentration is temperature-dependant. The present study on the transition process between SF and TF occurring at different molecules exhibits a reliable way to achieve controllable MEL for the magnetic application of OLEDs.

## 2. Experimental

The OLEDs with structure of ITO (120 nm)/copper phthalocyanine (CuPc) (15 nm)/*N,N'*-bis(naphthalene-1-yl)-*N,N'*-bis(phenyl) benzidine (NPB) (60 nm)/rubrene: doped DCJTB (30 nm)/bathocuproine (BCP) (50 nm)/lithium fluoride (LiF) (1 nm)/Al (120 nm) were fabricated, as shown in Fig. 1(a). 15 nm CuPc and 60 nm NPB are used as buffer layer and the hole transporting layer, respectively. 50 nm BCP acts as the electron transporting and hole blocking layer. LiF (1 nm) is used to improve electron injection efficiency. The device area is approximately  $2 \times 2 \text{ mm}^2$ . The energy level diagram is shown in Fig. 1(a). All organic layers were deposited by thermal evaporation under high vacuum ( $\sim 10^{-7}$  Pa). The samples were mounted on the cold finger of the close cycle cryostat located between the pole pieces of an electromagnet, the magnetic field of maximum strength of 500 mT was applied parallel to the device surface and measured by Hall probe Gauss Meter (lakeshore 421) placed close to the sample [3,7,17–19].

## 3. Results and discussion

### 3.1. Devices structure and the mechanism analysis

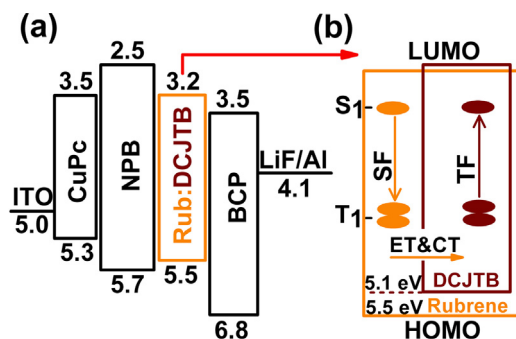
Fig. 1(b) shows the simplified kinetic scheme for rubrene: DCJTB doped layer. SF and TF processes may happen in the host molecular rubrene and the guest molecular DCJTB, respectively. Although they can coexist in the doped layer, the two processes are not independent. They were actually affected by both ET and CT [22]. The ET means that the excitons are primarily formed in the host molecules and then transfer their energy to the guest exciting the excitons in the guest. The ET contains Förster ET (FET) and

Dexter ET (DET) [22]. The FET can transfer the energy of singlet exciton in donor to the acceptor, depending on the overlap between the fluorescence spectrum of the donor and absorption spectrum of the acceptor. However, the DET process is important to the energy transfer of triplet excitons. In the DCJTB-doped devices, the intensity of singlet and triplet excitons of DCJTB will be increased by the FET and DET, respectively. In the CT channel, the carriers are directly trapped in the guest molecules to form excitons [7,22], rather than acquiring energy from the host excitons. Since the highest occupied molecular orbital (HOMO) of the guest DCJTB ( $\sim 5.1$  eV) is higher than that of rubrene ( $\sim 5.5$  eV), hole-trapping by DCJTB is possible and the electrons will consequently be attracted by the trapped holes. Therefore, the ET and CT processes can facilitate the process of TF whereas suppress the process of SF. As a consequence, controlling competition between the SF of rubrene and TF of DCJTB can be realized by changing the doping concentration of DCJTB in rubrene layer, achieving an effective method for tuning the MEL of devices.

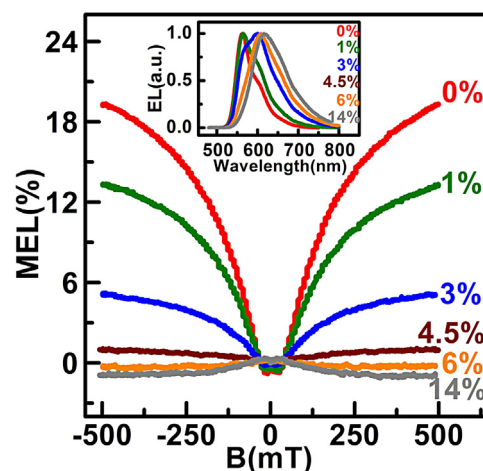
### 3.2. Tuning MEL at room temperature and the normalized spectrum analysis

We fabricated multiple devices with different doping concentration of DCJTB from 0% to 14%, and measured the MEL with the same injection current of  $150 \mu\text{A}$ . The quantity of MEL is defined as  $\text{MEL} = \Delta\text{EL}/\text{EL} = [\text{EL}(\text{B}) - \text{EL}(0)]/\text{EL}(0)$ , where  $\text{EL}(\text{B})$  and  $\text{EL}(0)$  are the intensities of electroluminescence with and without magnetic field B, respectively. As shown in Fig. 2, the MEL curves for undoped devices shows a slight decrease within  $\sim \pm 20$  mT and a large increase up to 19.3% at 500 mT. When DCJTB was doped into rubrene, the magnitude of MEL at 500 mT became smaller. The decrease of MEL was more apparent with larger doping concentrations, a decrease magnitude down to  $-1.00\%$  can be observed when the doping concentration gets 14%. Additionally, the value of MEL from positive to negative indicates that the originally dominant SF process in pure rubrene has been gradually taken over by the TF process after more and more DCJTB molecules were doped into emissive layer.

Generally, the fluorescence intensity  $F_{\text{EL}}$  of an OLED can be expressed as



**Fig. 1.** (a) Energy-level diagrams for rubrene: DCJTB doped OLED. (b) A schematic representation of singlet fission and triplet fusion processes in rubrene-based OLEDs. Singlet excitons are formed in rubrene. Triplet fusion can occur in DCJTB by the two channels, ET and CT. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)



**Fig. 2.** The MELs ( $\Delta\text{EL}/\text{EL}$ ) and normalized EL spectras (the inset figures) for devices changed with different doping concentration OLED (0%, 1%, 3%, 4.5%, 6%, 14%) with same injection current of  $150 \mu\text{A}$  at room temperature. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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