

High reverse intersystem crossing efficiency of exciplex states

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1. Introduction

The internal electroluminescence (EL) quantum efficiency (η_{int}) based on fluorescent materials as an emission center is limited to only 25%. Recently, without phosphorescent materials we proposed the novel EL mechanism to enable the η_{int} of 100% by using up-conversion from triplet (T_1) to singlet (S_1) excited states¹. In this mechanism, in order to realize the high up-conversion efficiency, the energy gap between S_1 and T_1 (ΔE_{ST}) should be close to zero. The ΔE_{ST} is determined by the exchange energy between the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO). Therefore, the separation of the HOMO from the LUMO can enhance the reverse intersystem crossing from T_1 to S_1 (RISC). In this paper, we demonstrate breakthrough of the theoretical limitation of fluorescent materials by using exciplex states which are formed between electron-donating and -accepting molecules.

2. Result and Discussion

First, we examined the property of photoluminescence on 50mol%-4,4',4''-tris[3-methylphenyl(phenyl)amino]triphenylamine(m-MTDATA) : 2-(biphenyl-4-yl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole (t-BuPBD) film. Figure 1 shows the PL spectra of m-MTDATA, t-Bu-PBD, and 50mol%-m-MTDATA:t-BuPBD film. We observed the red-shifted emission of the co-deposited film which is different from each neat film. This is due to the formation of exciplex between m-MTDATA and t-Bu-PBD. Figure 2 shows the PL transient decay curve of the co-deposited film. The curve is well fitted by a double exponential decay model, indicating that the emission from the co-deposited film has two different relaxation processes. We assigned the prompt component the exciplex fluorescence. On the other hand, the delayed component can be assigned to be delayed fluorescence originated from RISC. Inset of fig. 2 shows the temperature dependences of the PL quantum efficiencies (PLQE) for the prompt and delayed components, respectively. The PLQE of the prompt component increases with temperature decreasing. This is because of the suppression of non-radiative decay process from the singlet exciplex state with temperature decreasing. However, the delayed component shows different dependency. Above 150K, the PLQE steeply increases with decreasing temperature, since the non-radiative decay process from the triplet exciplex states is suppressed, increasing the RISC efficiency. Below 150K, the PLQE is decreased with decreasing temperature. This is due to the suppression of the RISC. Therefore, we conclude the delayed component is delayed fluorescence originated from RISC.

Next, we fabricated the device based on the exciplex as an emission center. The device structure is consisted of an ITO / m-MTDATA (20nm) / 50 mol% m-MTDATA:t-Bu-PBD (60nm) / t-Bu-PBD (20nm) / LiF / Al. The maximum external EL quantum efficiency (η_{ext}) of 2% was obtained. The PLQE of a 50mol%-m-MTDATA:t-Bu-PBD film was 20%. Assuming that the single-exciton production efficiency under electrical excitation is 25% and the light out-coupling efficiency is 20 ~ 30%, the η_{ext} is theoretically limited to approximately 1 ~ 1.5%. Therefore, this result means that the η_{ext} exceeds the theoretical limitation based on fluorescent materials.

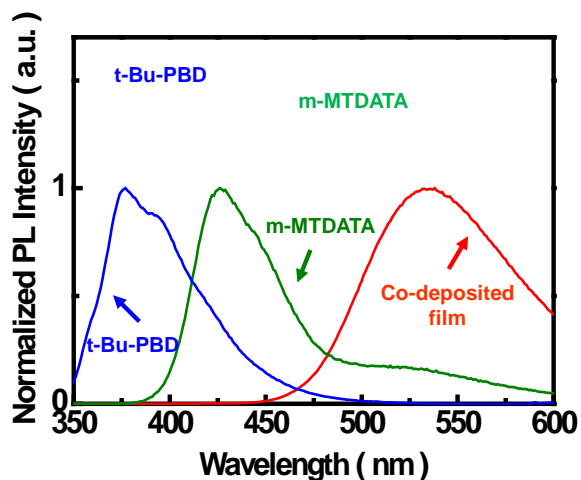


Fig. 1: PL spectra of m-MTDATA, t-Bu-PBD neat films, and a 50 mol% m-MTDATA:t-Bu-PBD co-deposited film.

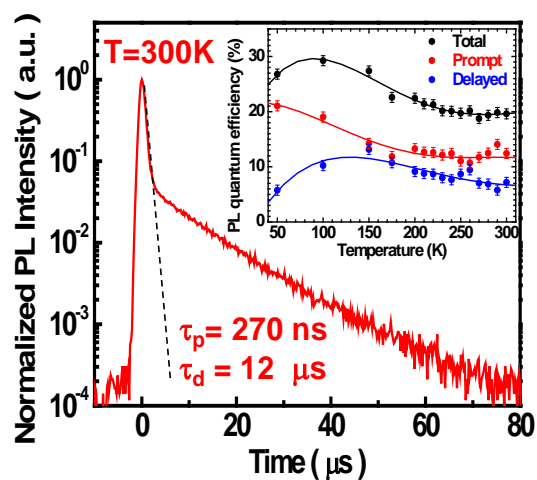


Fig. 2: PL transient decay curve of a 50 mol% m-MTDATA:t-Bu-PBD co-deposited film. (Inset) temperature dependence of PL quantum efficiency in a 50mol% m-MTDATA:t-Bu-PBD film.

3. References

- [1] A. Endo, M. Ogasawara, A. Takahashi, D. Yokoyama, Y. Kato, and C. Adachi, *Adv. Mater.* **21** (2009) 4802.

4. Acknowledgement

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