

## Electroluminescence of a red fluorescent dye doped in an Alq<sub>3</sub>:rubrene Mixed Host

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### Abstract

The electroluminescence (EL) properties were studied in organic light-emitting diodes with a red fluorescent dye, 4-(dicyanomethylene)-2-tert-butyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4H-pyran (DCJTb) doped into tris-(8-hydroxyquinoline)aluminum (Alq<sub>3</sub>), rubrene and the mixed matrix of Alq<sub>3</sub> and rubrene. The device with DCJTb doped into the Alq<sub>3</sub>:rubrene mixed host shows an efficient red emission from DCJTb with a negligible EL emission from Alq<sub>3</sub> and a lower EL onset voltage compared to the device with DCJTb doped into the Alq<sub>3</sub> only host. The quantum efficiency is almost temperature-independent for the device with the Alq<sub>3</sub>:rubrene mixed host. The results indicate that recombination of injected electrons and holes occurs on rubrene and subsequent energy transfer to DCJTb dominates in the device with the Alq<sub>3</sub>:rubrene mixed host.

### 1. Introduction

Doping the light-emitting layer with fluorescent or phosphorescent dyes has been widely used to enhance the electroluminescence (EL) efficiency or to tune the emission color of organic light-emitting devices (OLEDs), since the first demonstration by Tang *et al.* [1]. Another advantage of doping is the enhancement of the device stability by facilitating radiative recombination of injected electrons and holes and thereby reducing unstable excited states [2-4]. In addition, it was recently reported that the color coordinates and the EL efficiency of red fluorescent dyes doped into the tris(8-hydroxyquinoline) aluminum (Alq<sub>3</sub>) host can be further improved by doping additional dopant of rubrene which assists the energy transfer from Alq<sub>3</sub> to the red dopants [5, 6].

In this work we studied the energy transfer and charge transport processes in the devices with 4-(dicyanomethylene)-2-tert-butyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4H-pyran (DCJTb) doped into either the Alq<sub>3</sub> alone host or the

Alq<sub>3</sub>:rubrene (1:1) mixed host layer. Fig. 1 shows the device structure, the molecular structure of DCJTb and rubrene and the schematic energy band diagram with the energy levels of the highest occupied molecular orbitals (HOMO) and the lowest unoccupied molecular orbitals (LUMO).

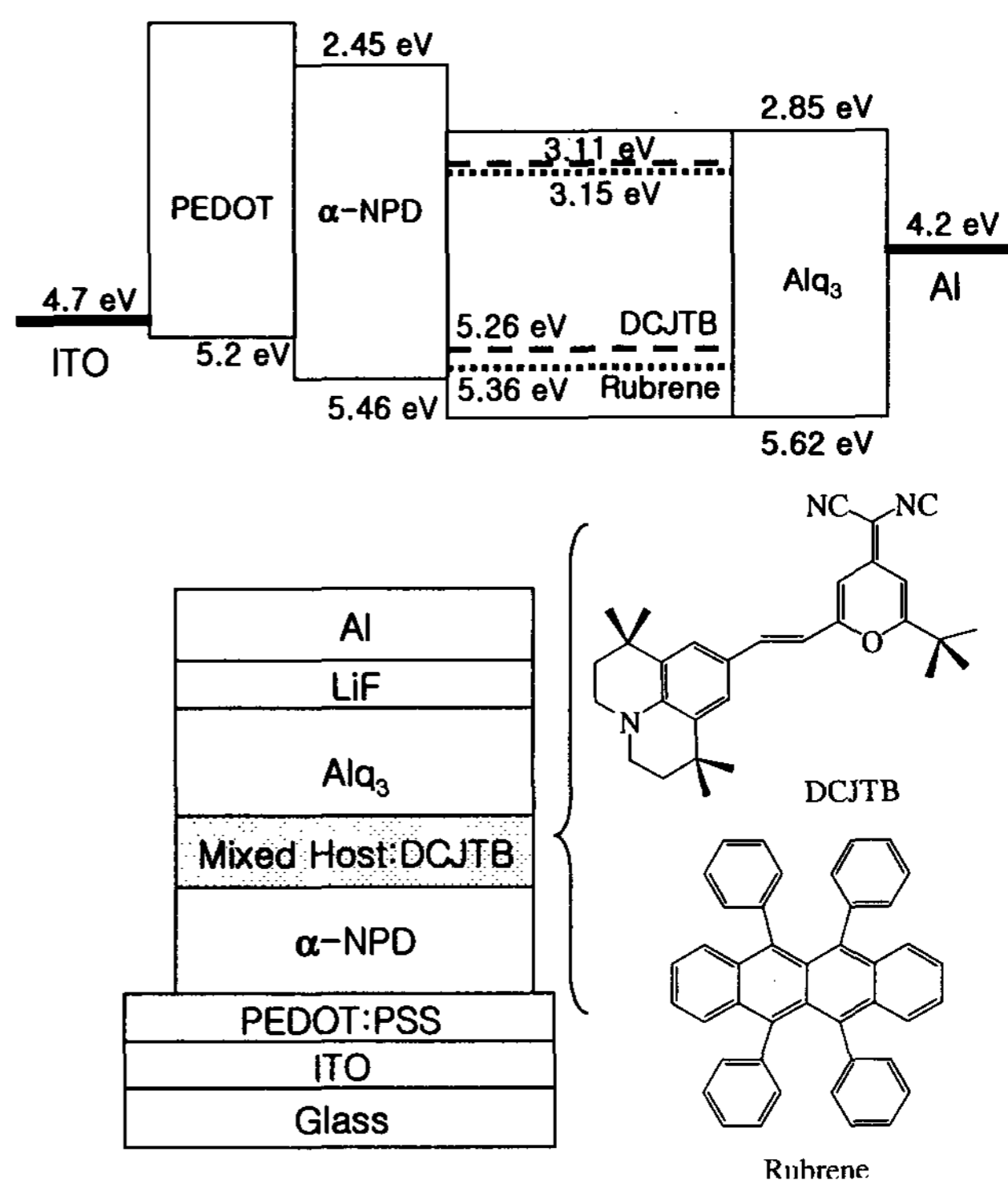


Figure 1. The schematic energy band diagram, device structure of ITO/PEDOT-PSS/ $\alpha$ -NPD/(Alq<sub>3</sub>:rubrene):DCJTb/Alq<sub>3</sub>/LiF/Al and the molecular structure of DCJTb and rubrene.

### 2. Experimental

The devices were fabricated on precleaned ITO glass substrates with a sheet resistance of about 10  $\Omega/\square$ . To enhance the device performance, poly(3,4-ethylene dioxithiophene):poly(styrenesulfonate)

(PEDOT-PSS, Baytron P4083) was deposited by spin coating from solution at 4000 rpm for 30 s, followed by drying at 100 °C for 1 hour in vacuum. Organic layers were deposited on top of the PEDOT:PSS layer by successive vacuum-deposition of  $\alpha$ -NPD (500 Å), the emitting layer (300 Å), Alq<sub>3</sub> (400 Å), LiF (5 Å), and Al electrode, without breaking vacuum. The emitting layer was deposited by simultaneously evaporating DCJTB, Alq<sub>3</sub>, and rubrene with appropriate evaporation rates. The doping concentration of DCJTB is about 0.5 % and the mixing ratio of Alq<sub>3</sub> and rubrene is 1:1. We also fabricated the reference device with DCJTB doped into the emitting layer of Alq<sub>3</sub> alone and keeping all the other parameters the same. The overlap area of the Al and ITO electrodes is about 2.5 mm<sup>2</sup>.

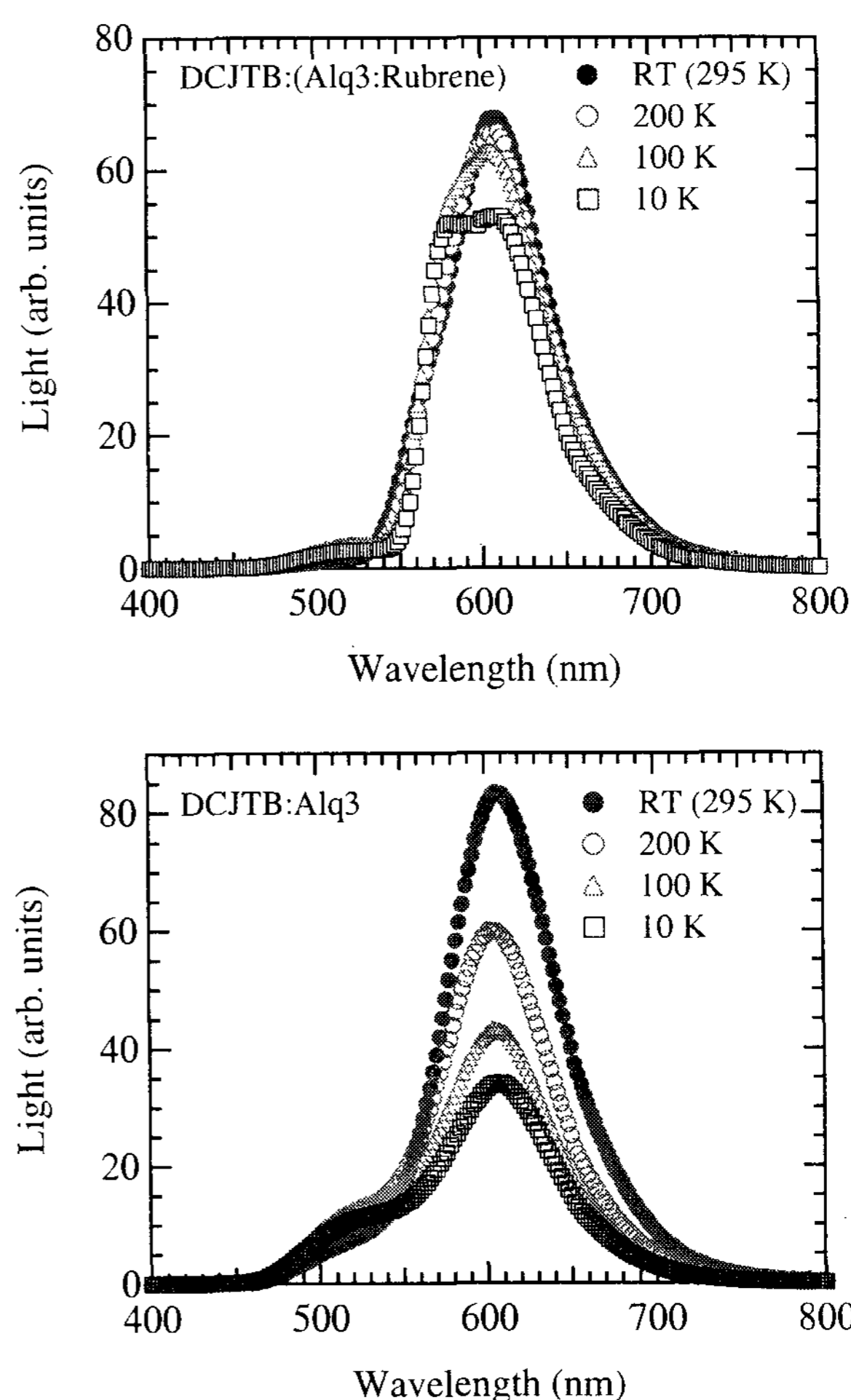
The current-voltage-luminance (I-V-L) characteristics and the EL spectra were studied in the wide temperature range between 10 and 320 K. The I-V-L characteristics were measured with a Keithley 236 source-measure unit and a Keithley 2000 multimeter equipped with a PMT through an ARC 275 monochromator. The external quantum efficiency (QE) of the EL, defined as the ratio of the emitted photons to the injected charges, was calculated from the EL intensity measured by the calibrated Si photodiode.

### 3. Results and discussion

Fig. 2 compares the EL spectra measured under the current density of 20 mA/cm<sup>2</sup> at various temperatures for the devices with the Alq<sub>3</sub>:rubrene mixed host (top) and the Alq<sub>3</sub> alone host (bottom). At room temperature, the device with DCJTB doped into the Alq<sub>3</sub>:rubrene mixed host shows a strong red emission with a negligible EL emission from Alq<sub>3</sub>. In contrast, there is some EL emission from Alq<sub>3</sub> due to the incomplete energy transfer from Alq<sub>3</sub> to DCJTB in the device with DCJTB doped into Alq<sub>3</sub> alone. The EL spectra of the two devices show different temperature dependences.

In the device with the Alq<sub>3</sub>:rubrene mixed host, the EL emission from rubrene increases significantly in addition to the EL emission from DCJTB as the temperature decreases. The emission from Alq<sub>3</sub> remains very weak at all temperatures. The overall EL intensity is almost same at all temperatures. Since the HOMO and LUMO energy levels of rubrene are in between those of Alq<sub>3</sub> and DCJTB (see Fig. 1),

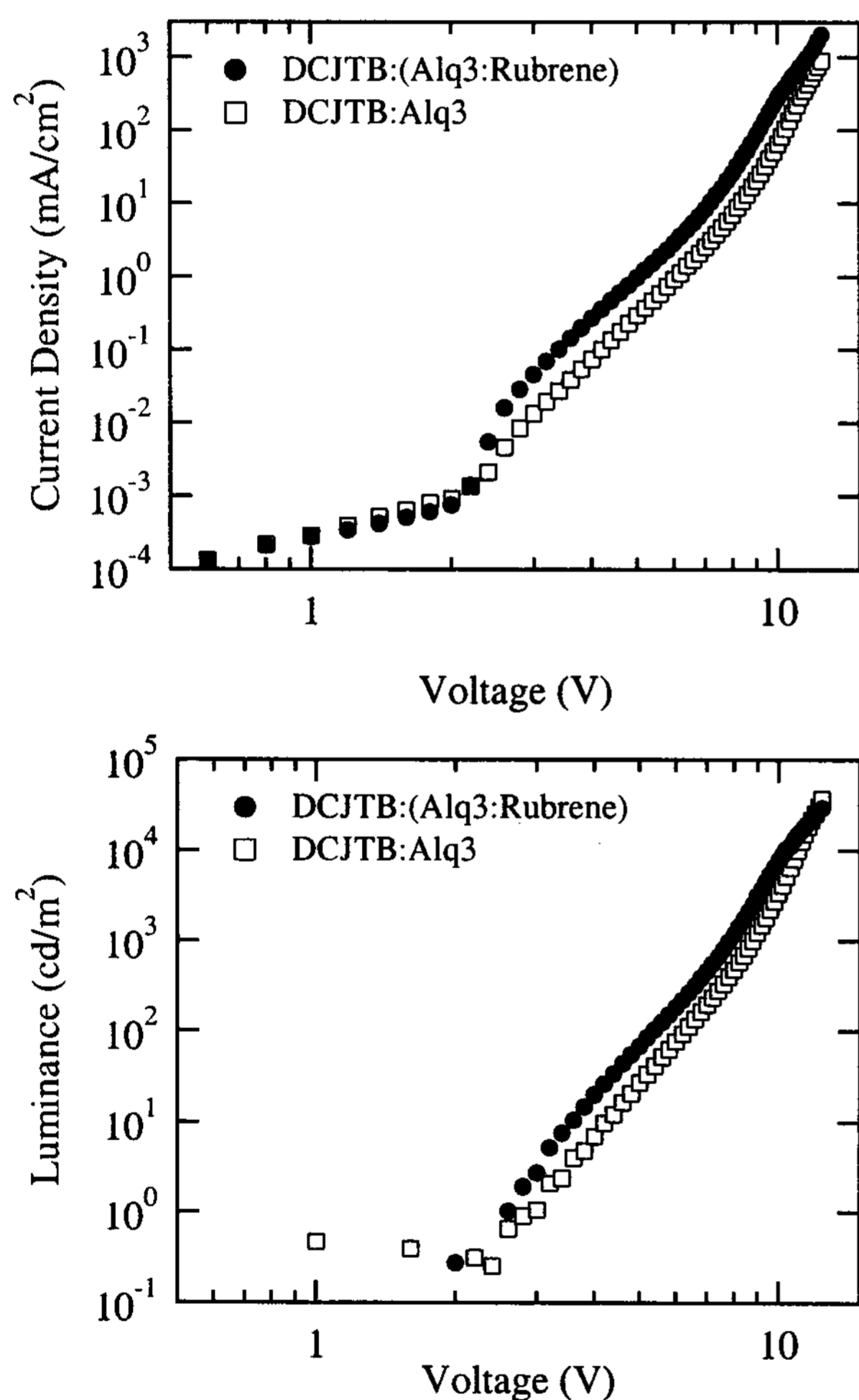
rubrene can play an intermediate state for the cascade energy transfer process from Alq<sub>3</sub> to DCJTB in the device with the Alq<sub>3</sub>:rubrene mixed host, as previously reported by Hamada *et al.* [5]. However, the observation of significant EL emission from rubrene but negligible EL from Alq<sub>3</sub> at low temperature implies that direct trapping and recombination of injected electrons and holes at rubrene and then energy transfer to DCJTB dominates in the Alq<sub>3</sub>:rubrene mixed host. As the temperature decreases, the energy transfer process becomes limited due to the decreased exciton diffusion process, resulting in a significant emission from rubrene.



**Figure 2.** The comparison of the EL spectra for the devices with the Alq<sub>3</sub>:rubrene mixed host (top) and the Alq<sub>3</sub> only host (bottom) under current density of 20 mA/cm<sup>2</sup> at various temperatures.

In the device with the Alq<sub>3</sub> alone host, the EL from

$\text{Alq}_3$  remains almost same and the EL emission from DCJTB increases significantly as the temperature increases. Therefore, it is considered that the injected electrons and holes recombine on  $\text{Alq}_3$  and then the energy transfer from  $\text{Alq}_3$  to DCJTB occurs in the device with the  $\text{Alq}_3$  alone host. As the temperature increases, the energy transfer process becomes more efficient due to increasing exciton diffusion length. Since the HOMO and LUMO levels of DCJTB locate deep inside the  $\text{Alq}_3$  energy band gap, direct recombination of electrons and holes on DCJTB may also increase at high temperature due to the increased mobility of charge carriers.

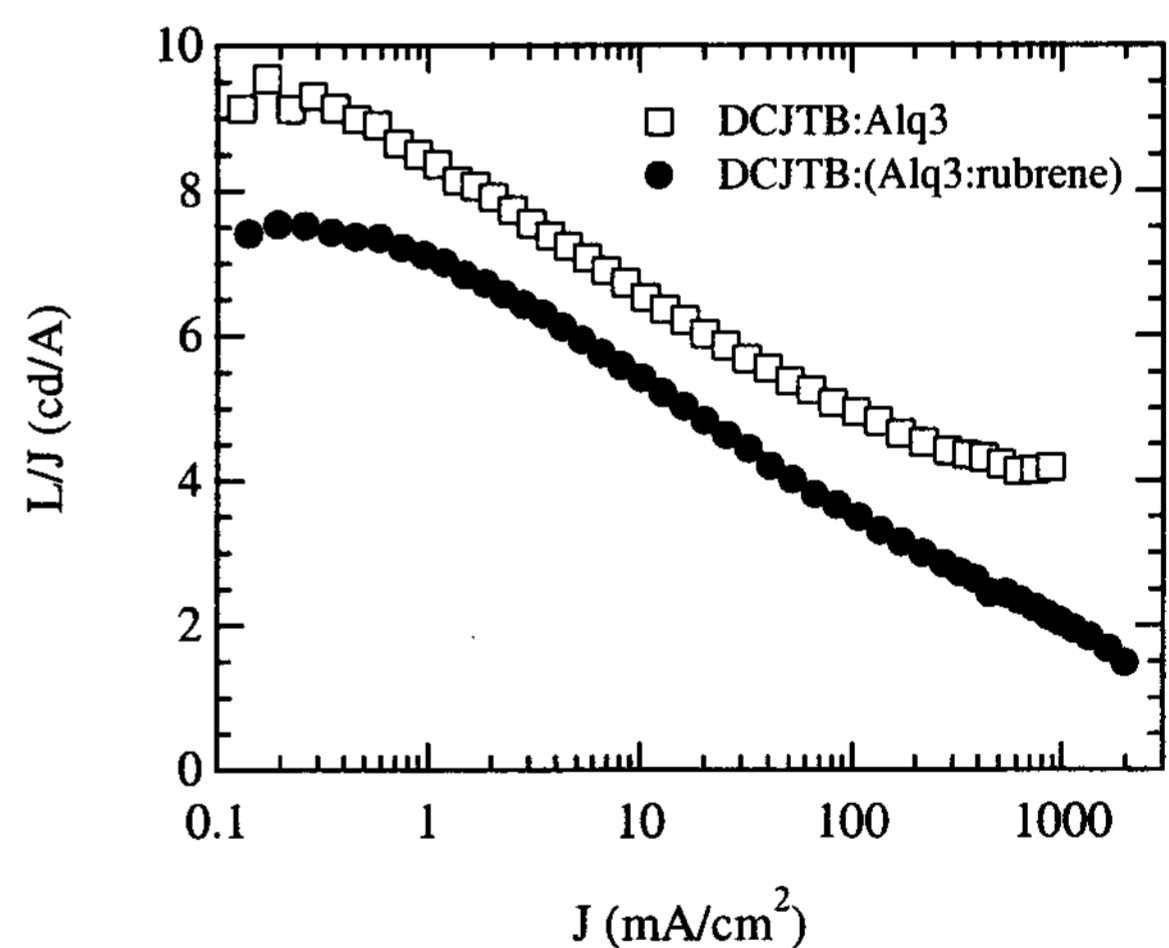


**Figure 3.** The current-voltage (I-V) (top) and the luminance-voltage (L-V) characteristics of the devices with the  $\text{Alq}_3$ :rubrene mixed host (solid circle) and the  $\text{Alq}_3$  only host (open square) at

room temperature.

Fig. 3 compares the current-voltage (I-V) and luminance-voltage (L-V) characteristics of both devices at room temperature. The EL onset voltage and the operating voltages at the same luminance level are lower by about 1 V for the  $\text{Alq}_3$ :rubrene mixed host compared with the  $\text{Alq}_3$  only host. Since the HOMO and LUMO energy levels of rubrene are within those of  $\text{Alq}_3$  [5], the recombination of injected electrons and holes occurs on rubrene, and therefore, resulting in the lower voltage compared with the recombination on  $\text{Alq}_3$  for the device with the  $\text{Alq}_3$  only host. Therefore, the additional rubrene doping enhances the power efficiency of the EL.

Fig. 4 displays the luminous efficacy (cd/A) as a function of the current density for the two devices. The device with the  $\text{Alq}_3$ :rubrene mixed host exhibits a slightly lower luminous efficacy although it has a lower operating voltage. The reason is attributed to the concentration quenching effect of rubrene in the  $\text{Alq}_3$ :rubrene mixed host. The luminous efficacy decreases with increasing current density for both devices. This phenomenon is common for devices with a dye doped emitting layer. It is attributed to the quenching of the singlet-excited state of the dopant by a cationic species [7].



**Figure 4.** The luminous efficacy (L/J) as a function of the current density for both devices shown in Fig. 3 at room temperature.

#### 4. Conclusion

We have studied the energy transfer and charge

carrier trapping processes in organic light-emitting diodes with DCJTB doped into the Alq<sub>3</sub>:rubrene mixed host and the Alq<sub>3</sub> only host. The device with the Alq<sub>3</sub>:rubrene mixed host shows an efficient red emission from DCJTB, a negligible EL emission from Alq<sub>3</sub>, and a lower EL drive voltage compared to the device with the Alq<sub>3</sub> only host. Therefore, the Alq<sub>3</sub>:rubrene mixed host is promising for the development of an efficient red OLEDs. We find that the recombination of injected electrons and holes takes place on rubrene and subsequent energy transfer to DCJTB dominates in the device with the Alq<sub>3</sub>:rubrene mixed host. In contrast, the electron-hole recombination occurs on Alq<sub>3</sub> and subsequent energy transfer to DCJTB occurs in the device with the Alq<sub>3</sub> only host.

### 5. Acknowledgements

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### 6. References

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