

Current Density Dependence of Transient Properties in Green Phosphorescent Organic Light-Emitting Diodes

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1. Introduction

Organic semiconductors have attracted considerable attention due to their simple and low-cost processes and potential electronic and optoelectronic applications. Organic light-emitting diodes (OLEDs) utilizing fluorescent dyes or conducting polymers are capable of emission over a wide visible range, high efficient, and require only a low driving voltage. OLEDs have been realized which have a long lifetime and excellent durability for flat-panel display applications. There are some requirements of OLEDs for use not only in display applications but also as various light sources.

Employing phosphorescent materials exhibits high efficiencies because breaking the spin conservation rule indicates that both the singlet and triplet excitons contribute to emission. However, the response time of phosphorescent materials such as Ir complexes is slower than that of fluorescent ones due to their long emission lifetime. The study of the transient electroluminescence of OLEDs has been also reported. [1-3] The applied voltage dependence of fundamental transient properties in these devices are mostly investigated.

In this study, we studied the current density dependence of transient characteristics of green phosphorescent OLEDs with Ir complex as the emissive layer. We discussed the transient electroluminescence of green phosphorescent OLEDs using pulses of alternating current sine-waves with various frequencies.

2. Experimental

The device structure used in this study is shown in the inset of Fig. 1. The substrate was degreased with solvents and cleaned in a UV ozone chamber. First, poly(ethylene dioxythiophene):poly(styrene sulfonic acid) (PEDOT:PSS) hole injection layer with 40 nm was spin-coated on the indium tin oxide (ITO)-coated glass substrate and baked in air at 120°C for 20 min. Next, the device consisted of an indium-tin-oxide (ITO) - coated glass substrate, PEDOT:PSS hole injection layer, 4,4'-bis[N-(1-naphthyl)-N-phenyl-amino]-biphenyl (α -NPD) hole transporting layer, tris(2-phenylpyridine)iridium(III) ($\text{Ir}(\text{ppy})_3$) doped in 4,4',4''-tris(carbazolyl-9-yl)triphenylamine (TCTA) and 2,2',2''-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) double phosphorescent emissive layers, and bis(2-methyl-8-quinolino)-4-(phenylphenolato)aluminum (BALq) hole blocking layer, terminated with a 0.5 nm-thick lithium fluoride (LiF) and Al/Ag electrodes cathode. The

doping concentration of $\text{Ir}(\text{ppy})_3$ was fixed at 10 vol% relative to the double hosts of TPBi and TCTA. The typical active area was 4 mm².

3. Results and discussion

The J-V-L characteristics for a device of ITO/PEDOT:PSS (40 nm)/ α -NPD (20 nm)/TCTA: $\text{Ir}(\text{ppy})_3$ (15 nm)/TPBi: $\text{Ir}(\text{ppy})_3$ (15 nm)/BALq (40 nm)/LiF/Al/Ag are presented in Fig.1. The luminance increased significantly in the forward bias direction above a threshold voltage of approximately 2.6 V. The maximum luminance of approximately 45,000 cd/m² and maximum current efficiency of 34 cd/A were obtained.

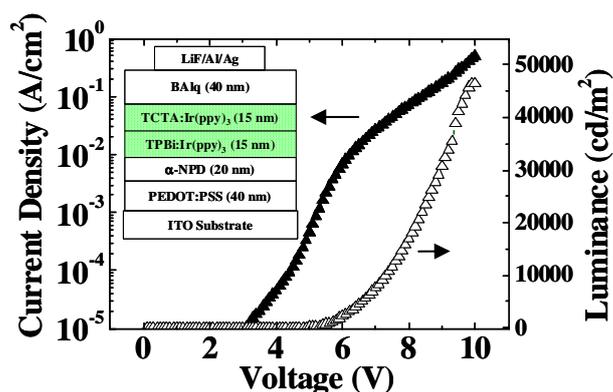


Fig. 1 J-V-L characteristics of a green phosphorescent OLED. The inset shows the device structure.

Figure 2 shows current density dependence of the rise and decay times of a green phosphorescent OLED with $\text{Ir}(\text{ppy})_3$ as an emissive dopant by applying the rectangle voltages. The device was driven at a 0.1 or 1 ms period and duty ratio of 1/10 pulse voltage. The decay (rise) time is defined as the time required to change the optical response from 90 (10) to 10 % (90 %) of its total intensity change. There were two parts in the behavior of rise time. At a lower current density, the rise time was longer than the phosphorescence lifetime of $\text{Ir}(\text{ppy})_3$. For a green OLED, the rise time gradually decreased with increasing applied voltage and was approximately 1 μs at higher current density. On the other hand, the decay time has fast and slow components, which are attributed to the photoluminescence lifetime of emissive materials or the RC time constant of devices and delayed EL from residual carriers in the emissive layer, respectively. The decay time of a green OLED

was approximately 2 μs at less than $40\text{mA}/\text{cm}^2$. While, both the rise and decay times of photoluminescence (PL) of host:Ir(ppy)₃ film were approximately 2 μs by measuring the transient response time in PL with a 408 nm blue semi-conducting laser diode. The decay time of EL is almost same as the that of PL in Ir(ppy)₃ at lower current densities. The decay time decreased with the current efficiency. Then, at higher current densities, the rise time was almost the same as the decay time.

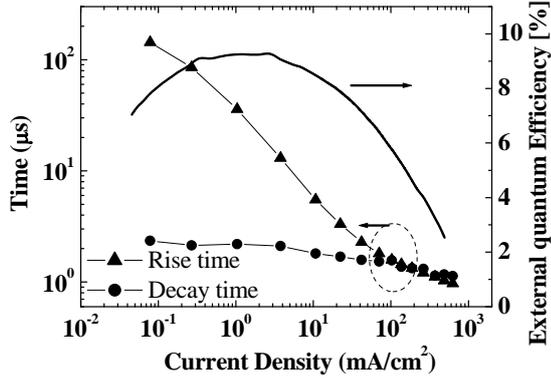


Fig. 2 Current density dependence of the rise and decay times, and current efficiency of a green phosphorescent OLED with Ir(ppy)₃ as an emissive dopant

The rate equation [4] of the phosphorescence OLEDs is indicated as follows:

$$\frac{d[{}^3M^*]}{dt} = -\frac{[{}^3M^*]}{\tau} - \frac{1}{2}k_{TT}[{}^3M^*]^2 + \frac{J}{qd},$$

where τ is the phosphorescence recombination lifetime, $[M^*]$ is the concentration of triplet excitons, k_{TT} is the triplet-triplet annihilation constant, J is the current density, d is the exciton formation zone, and q is the electron charge. From this rate equation, the transient properties are strongly affected by phosphorescence recombination lifetime. The rate of phosphorescence recombination lifetime (τ) is represented by the inverse of radiative recombination lifetime (τ_r) and nonradiative recombination lifetime (τ_{nr}): $1/\tau = 1/\tau_r + 1/\tau_{nr}$.

At higher current densities, the increase of the nonradiative process results in the decrease in τ . This is because exciton interaction within an emissive layer and between an emissive layer and a hole or an electron transporting layer increases the probability of the nonradiative process. The device exhibited gradual decrease in quantum current efficiency due to the triplet-triplet annihilation at the high current density as shown in Fig. 2. At higher current density, the reduced rise and decay times may also be due to high-density triplet excitons related to the enhanced triplet-triplet annihilation. Therefore, both the rise and decay times were observed to be approximately 1 μs . This green phosphorescent OLED can be expected to be utilized as one of the light sources driven at less than 0.5 MHz.

Figure 3 demonstrates the typical transient current density, voltage, and EL signal of a device at a current density

of $100\text{mA}/\text{cm}^2$ with a repetition of 10 kHz, and the frequency dependence of the EL intensity of a device using pulses of alternating current sine-waves, where EL intensities at modulation frequencies are normalized by the EL intensity at 0.1 kHz. It should be noted that the transient EL signal was almost synchronized with the input current sine-waves as shown in Fig. 3(a). The frequency response of the device was improved by increasing applied current densities. The cut-off frequencies of the device were approximately 10 and 100 kHz at 1 and $100\text{mA}/\text{cm}^2$, respectively.

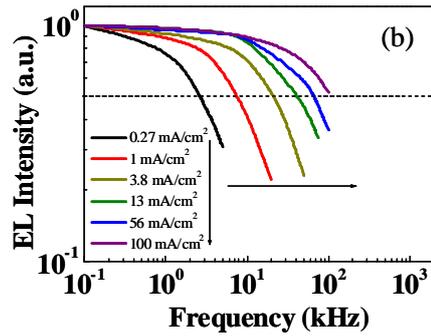
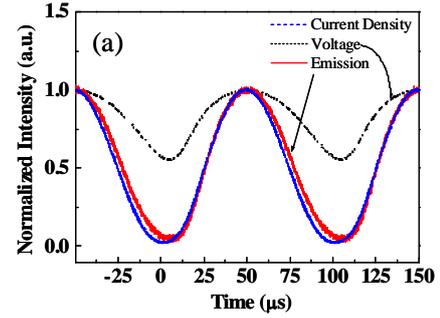


Fig. 3 (a) Typical transient current density, voltage, and EL signal of a device at a current density of $100\text{mA}/\text{cm}^2$ with a repetition of 10 kHz, and (b) frequency dependence of the EL intensity in a device using pulses of alternating current sine-waves.

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