Transient property of optically pumped organic film of different fluorescence lifetimes

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(Received 27 February 2007; accepted 11 May 2007; published online 4 June 2007)

The authors have investigated the direct relationship between the fluorescence lifetime (FL) and the transient photoluminescence (PL) response of organic materials to examine the applicability of organic light-emitting diodes as light sources for optical communications. Transient PL responses of organic materials with different FLs were measured as a frequency dependence of PL intensity pumped by a modulated violet laser diode. The authors have revealed that the cutoff frequency of PL intensity is significantly related to FL of an organic film. The −3 dB cutoff frequency of 1,4-bis[2-[4-[*N*,*N*-di(*p*-tolyl)amino]phenyl]vinyl]benzene (DSB) has reached about 160 MHz, which is much higher than that of other organic materials. This is because the FL of DSB is shortest among all the organic materials used in this study. © *2007 American Institute of Physics*.
[DOI: 10.1063/1.2746083]

In recent years, organic light-emitting diodes (OLEDs) have attracted many researchers because of some advantages, such as a low fabrication cost, color selectivity, thickness, and flexibility.[1,2] By now, OLEDs have been mainly researched for display applications to make the best use of their advantages. Furthermore, many practical uses of OLED displays have been developed.

Some research groups have reported application of OLEDs as light sources for optical communications.[3–6] Several factors affecting response time of OLEDs have been already investigated, particularly capacitance determined by device area and thicknesses of organic layers,[3,4] fluorescence lifetime (FL) of the light-emitting materials,[5] and carrier mobility of electron/hole transport materials.[7–9] In optical communication applications, modulation speed of OLEDs has been estimated as the −3 dB cutoff frequency of electroluminescence intensity and the −3 dB cutoff frequency of more than 10 MHz has been achieved with a small OLED.[10] However, the reported cutoff frequency was not always sufficient to apply these OLEDs as light sources for optical communications. It has been recognized that several hundred megahertz or over gigahertz modulation speed is desirable to expand an application field.

In these parameters, it has been known that the modulation speed can increase by utilizing the short FL material.[5] However, there is little quantitative data on the relationship between the −3 dB cutoff frequency and the FL.

In this letter, we have investigated the direct influence of the FL on the transient property of photoluminescence (PL) intensity. The quantitative relationship between the −3 dB cutoff frequency and the FL has been found by investigating PL responses of thin organic films excited by a high-frequency violet laser diode.

We fabricated organic films on glass substrates by conventional thermal evaporation at a base pressure below 5.0×10^{-6} Torr. The glass substrates were cleaned in deionized water, detergent, and isopropyl alcohol sequentially under ultrasonic waves, and then treated with 50 W oxygen plasma for 5 min just before use. We used organic materials of 1,4-bis[2-[4-[*N*,*N*-di(*p*-tolyl)amino]phenyl]vinyl]benzene (DSB), 4,4′-(bis(9-ethyl-3-carbazovinylene)-1,1′-biphenyl, 4,4-bis(2,2-ditolylvinyl)biphenyl (DPVBi), tris(8-hydroxyquinoline) aluminum) (Alq_3), 0.5 mol % (3-(2-benzothiazolyl)-*N*,*N*-diethylumbelliferylamine (coumarin 6) doped Alq_3, 0.5 mol % 5,6,11,12-tetraphenyltetracene (rubrene) doped Alq_3, and 1.0 mol % 4-(dicyanomethylene)2-methyl-6-(julolidin-4-yl-vinyl)-4*H*-pyran (DCM 2) doped Alq_3. The deposition rate was monitored using a quartz sensor and maintained at 0.5 nm/s. Thicknesses of all the films were 100 nm.

We have measured FLs of all the organic films by a femtosecond pulse laser. The center wavelength and the pulse width of the femtosecond pulse laser were 390 nm and 112 fs, respectively. All the organic films radiated PLs when the femtosecond pulse laser was irradiated. The radiated PL was captured with a streak camera, and then time-resolved PL spectra were measured. Finally, monoexponential fitting was employed to derive the FL from the measured time-resolved PL intensity.

Furthermore, we have also measured the frequency dependence of PL intensity to investigate the direct relationship between the −3 dB cutoff frequency of PL intensity and the FL. A schematic configuration the an experimental setup is shown in Fig. 1. The organic film was excited by the violet laser diode (NDHV220APAE1-E, Nichia Corp.), whose center wavelength was 405 nm. By utilizing the programmable FM/AM standard signal generator (SG-7200, Kenwood), the violet laser diode was operated by a high-frequency sine wave voltage. The amplitude of the sine wave voltage was ±0.5 V and the bias voltage was 9.5 V, which was applied by the dc power supply (IPS3610D, ISO-TECH). Then, PL intensity was observed by using the avalanche photodiode

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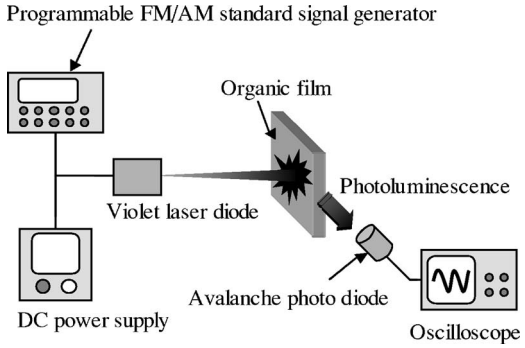

FIG. 1. Schematic configuration of an experimental setup for the frequency dependence of PL intensity under a violet laser diode.

(APD) (S5343, Hamamatsu Photonics), which was located perpendicular to the optical axis of the laser diode, as shown in Fig. 1.

As a result, the frequency dependence of PL intensity was estimated by changing the modulation frequency of the violet laser diode. Moreover, PL spectra were measured by the spectrophotometer (USB 2000, OceanOptics Company) also located perpendicular to the optical axis of the laser diode.

Figure 2(a) shows the logarithmic plot of PL intensity versus the frequency of the violet laser diode for two organic materials, DSB and Alq_3. For both organic films, PL intensity decreases with increasing the frequency of the violet laser diode due to the decay time of the PL. The −3 dB

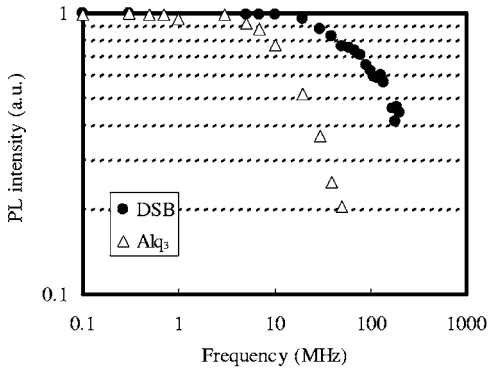

FIG. 2. (a) Frequency dependence of PL intensity of DSB and Alq_3 and (b) −3 dB cutoff frequency of PL response to the violet laser diode as a function of the FL.

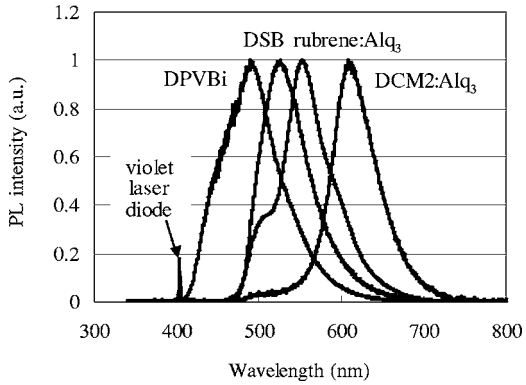

FIG. 3. PL spectra of organic films excited by the high-frequency violet laser diode.

cutoff frequency of PL intensity was estimated as the frequency when PL intensity was 0.5 relative to that of the low-frequency region. This experimental result yielded that −3 dB cutoff frequencies were 160 and 20 MHz for DSB and Alq_3, respectively. Moreover, FLs of DSB and Alq_3 were 0.2 and 16.0 ns, respectively.

Figure 2(b) shows the −3 dB cutoff frequency of PL intensity as a function of the FL for seven species organic films. As is evident from Fig. 2(a), FL dependence is found to be linear and yields good fits to power law, $F_C=72.996L_F^{-0.486}(R^2=0.97)$, where F_C and L_F are the −3 dB cutoff frequency and the FL, respectively. An additional remark, which should be made here, is that the −3 dB cutoff frequency can reach 160 MHz for DSB due to the short FL of 0.2 ns. So, the longer FL of Alq_3 causes the decrease of the −3 dB cutoff frequency as shown in Fig. 2(a).

This result indicates only an effect of the FL without influences of capacitance and carrier mobility, which are known to affect modulation speed.[7–9] So, we can estimate the direct influence of FL on transient property of an OLED. The transient property of PL is strongly dependent on the FL, and the modulation speed is considered to increase by utilizing the short FL organic material as a light-emitting layer of an OLED.

Figure 3 shows PL spectra of four organic films excited by a violet laser diode (wavelength of 405 nm). Organic films were DSB, DPVBi, 0.5 mol % rubrene doped Alq_3, and 1.0 mol % DCM2 doped Alq_3. Organic films of 100 nm thicknesses fully absorbed the violet laser emission and effectively converted into PL. As a result, little blue emission was detected with an APD. Therefore, we can surely conclude that the measured PL response was mainly contributed to the excited emission by the violet laser diode. Furthermore, organic materials have different peak emission wavelengths due to the energy band gap between the highest occupied molecular orbital and the lowest unoccupied molecular orbital levels. In optical communication applications, several OLEDs with different peak emission wavelengths can realize the wavelength division multiplexing system to increase the transmission capacity.

In conclusion, we have investigated the direct relationship between the FL and the −3 dB cutoff frequency against an applied sine wave voltage for optical communication applications. By measuring the frequency dependence of PL intensity from pumped organic films with different FLs, we found that the −3 dB cutoff frequency correlates closely to the FL. The highest −3 dB cutoff frequency of PL intensity

can reach about 160 MHz using one substituted phenyl/vinyl compound, DSB, of which the FL was 0.2 ns.

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