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HIGH-SEED OPERATION OF ORGANIC ELECTROLUMINESCENT DIODES AND PHOTO-DETECTORS FOR APPLICATION OF OPTICAL INTEGRATED DEVICES

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Organic electroluminescent diode (OELD) has been investigated for use as a light source of polymeric optical integrated devices. The OLED was fabricated by organic molecular beam deposition (OMBD) technique. The OLEDs were fabricated on both glass and polymeric substrates. The device fabricated on a polymeric substrate shows similar device characteristics to those on a glass substrate. Optical signals of faster than 100 MHz has been created by applying pulsed voltages directly. Optical photo detectors (OPDs) utilizing phthalocyanine superlattice structure provide increased pulse response with optical signals and have response for optical pulse of faster than 1 MHz.

Keywords: optical integrated circuits; organic electroluminescent diode; organic photo-detector; polymeric waveguide; rubrene

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INTRODUCTION

Organic electroluminescent diodes (OLEDs) [1] utilizing fluorescent dye or conducting polymer have attracted great interest because they have advantages for thin film flat-panel display. An additional advantage is that they are simple for fabrication on various kinds of substrates, including polymeric substrates. Metal phthalocyanines are well-known materials which show good stability, photo activity and high mobility. Among them, fluorinated zinc phthalocyanine shows high electron mobility and will be suitable for fabrication high speed organic photo-detectors (OPD).

On the other hands, polymeric waveguide devices have attracted great attentions with regard to their use for flexible optical circuits and optical components. The combination of polymer waveguide [2] and optical devices (OLED and OPD) and will realize a flexible optical integrated circuits [3,4]. Since the polymeric waveguides have a low transmission loss in the near-infrared region, a light-emitting device with longer wavelength range, as a light source, will provide an efficient tool for optical integrated circuit utilizing polymeric waveguides.

In this paper, we discuss the characteristics of OLEDs fabricated on the glass and polymeric substrates. OLED with various emission wavelengths including red light, yellow light, green light and blue light emissions has been investigated. Among them, the OLED, with which emissive layer consists of rubrene (5,6,11,12-tetraphenylnaphthacene) doped in Alq_3 (8-hydroxyquinoline aluminum), emits yellow-light with high power intensity and high frequency response. We also discuss photo-response of OPDs, which consist of the multilayered p-type and n-type phthalocyanine thin films.

EXPERIMENTAL

The molecular structures of the materials used for the OLEDs with typical device structure and OPDs with device structure are shown in Figure 1(a) and (b), respectively. The organic layer was vacuum deposited by organic molecular beam deposition (OMBD) system at a background pressure of 10^{-5} Pa. For substrates used for the experiments, both glass substrates and polymeric substrates were used. Polyimide films were employed as polymeric substrates, because they have high thermal stability, high optical transparency, and they are one of the promising candidates for polymeric waveguide devices. After fabricating the OLEDs and OPDs, they are sealed in an inert gas (Ar gas) as is also shown in Figure 1. Typical OLED consists of indium-tin-oxide coated glass or polymeric substrate, 4,4-bis[N-(1-napthyl)-N-phenyl-amino]-biphenyl (α-NPD) hole transporting layer,

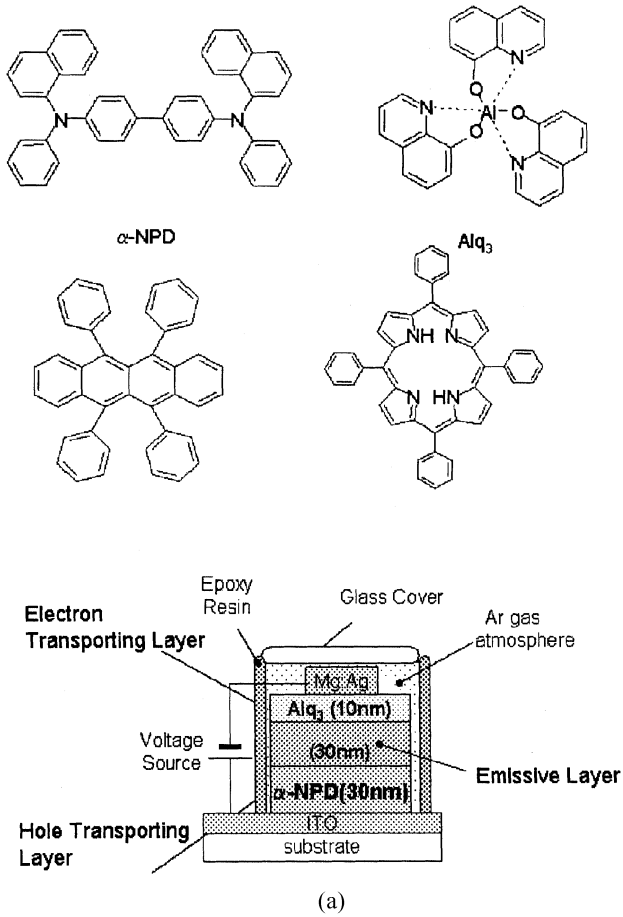
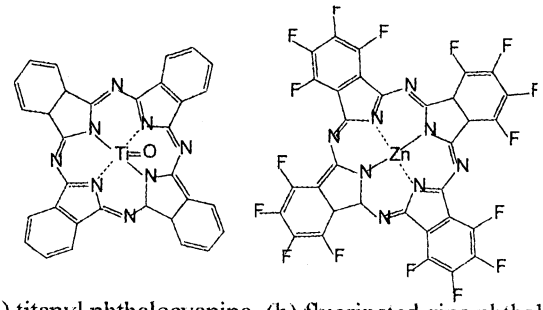


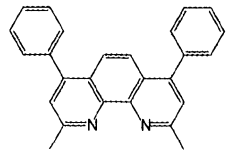
FIGURE 1 Molecular structure and typical device structures used in the experiments. (a) organic electroluminescent diodes(OLED), (b) organic photo-detectors (OPD).

8-hydroxquinoline aluminum (Alq_3) or dye doped in Alq_3 emissive layer, terminated with silver-containing magnesium (Mg:Ag) cathode. 5,6,11,12-tetraphenyl-naphthacene (rubrene) or 5,10,15,20 tetraphenyl-21H,23H-porphine (TPP) molecules were doped in Alq_3 as an emissive material, respectively. In order to reduce the straight capacitance of the diode, the active size of the device were employed as 0.01 mm^2.

The OPDs were fabricated by the same OMBD system. The device consists of alternating layers of TiOPc, $F_{16}ZnPc$ and bathocuproine (BCP) as is shown in Figure 1. The total layer thickness of photo-absorption layer of



(a) titanyl phthalocyanine (b) fluorinated zinc phthalocyanine



(c) bathocuproine

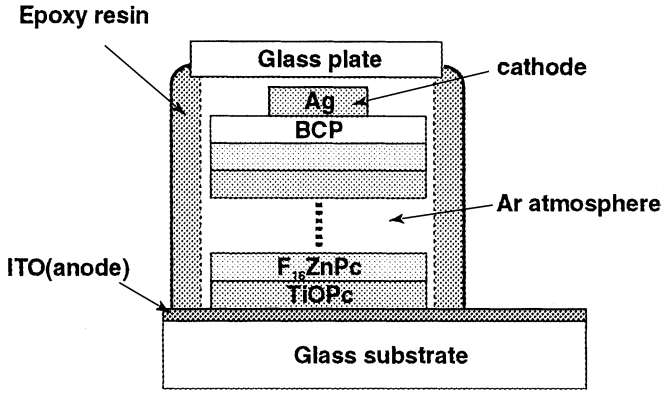


FIGURE 1 (Continued).

TiOPc and F_{16}ZnPc has been kept at 40 nm. The number of periods of photo-absorbing layers was chosen from single heterostructure to 10 periods of alternate layers. The 20 nm-thick BCP layer acts as a carrier blocking layer, and also prevents the organic materials from the damage during metal deposition. This is followed by the deposition of metal cathode evaporated. Finally, the device was sealed in an inert Ar gas. The measurement of photo response has been performed with and without applied reverse bias to the photo detectors under an illumination of pulsed light of red LED (640 nm).

RESULTS AND DISCUSSIONS FOR ORGANIC LIGHTEMITTING DIODES

The emission characteristics of OLEDs are summarized in Table 1, which are made with the materials shown in Figure 1. In case of a device in which α-NPD as an emissive layer, the device consists an ITO-coated glass substrate, 60 nm-thick α-NPD as a hole transporting emissive layer, 5 nm-thick 4,4-bis(carbazol-9-yl)-biphenyl (CBP) and 10 nm-thick 2,9-dimethy-4,7-diphenyl-1,10-phenanthroline (BCP) as hole-blocking layers, and 15 nm-thick (Alq_3) as an electron transporting layer, terminated with a silver containing magnesium (Mg:Ag) cathode. The device emits clear blue emission centered at about 435 nm, and the turn-on voltage of the device is about 3 V. The maximum emission intensity reaches to 40 mW/cm^2. The light pulses were created by direct modulation of the OLED with an active area of 0.01 mm^2 under the applied pulse of 100 MHz.

The OLED made with Alq_3 as an emissive material emits clear green emission centered at about 520 nm. The device consists of an ITO-coated glass substrate, 50 nm-thick α-NPD as a hole transporting layer, 30 nm-thick Alq_3 as an emissive and electron transporting layer, terminated with a Mg:Ag cathode. The maximum emission intensity reaches to 30 mW/cm^2, at an applied voltage of 10 V and an injection current density of 6 A/cm^2. However, the modulation characteristics are rather poor compared with α-NPD device, and its high frequency modulation limited to 60 MHz. It will be due the fluorescence lifetime of Alq_3, which is more than 10 ns.

The device which consists of 9.1 vol% of rubrene doped in Alq_3 layer as an emissive layer [5], emits clear yellow emission centered at about 560 nm. The device consists of 50 nm-thick α-NPD as a hole transporting layer, 30 nm-thick rubrene doped in Alq_3 as an emissive layer and 10 nm-thick Alq_3 as an electron transporting layer, terminated with a Mg:Ag cathode. The emission reaches 30 mW/cm^2, at an applied voltage of 9 V and the

TABLE 1 Summary of Emission Characteristics of Various Kind Organic Light Emitting Diodes

Emissive Materials	α-NPD	Alq_3	Rubrene	TPP
Central Wavelength	430 nm	520 nm	560 nm	655 nm 720 nm
Emission Intensities	More than 40 mW/cm^2	More than 30 mW/cm^2	More than 30 mW/cm^2	More than 4 mW/cm^2
Frequency Limitation	More than 100 MHz	~60 MHz	More than 100 MHz	More than 100 MHz

current density of 10 A/cm^2. The device also creates more than 100 MHz modulation.

The red light emitting device consists of 50 nm-thick α-NPD as a hole transporting layer, 30 nm-thick TPP doped in Alq_3 as an emissive layer and 10 nm-thick Alq_3 as an electron transporting layer, terminated with a Mg:Ag cathode. The mole fraction of TPP is only 1%, and it's enough to emit red light. The emission of TPP has two peaks centered at 650 nm and 720 nm. The modulation frequency reaches at 100 MHz, which is similar to the device with α-NPD as an emissive layer. However, the emission intensity is rather poor compared with other materials due to the poor emission efficiency, and the maximum emission intensity is 4 mW/cm^2 at a current density of 2 A/cm^2.

In Figure 2, modulation characteristics of the OLED with rubrene doped in Alq_3 layer is shown. The optical pulses are created directly by applying voltage pulses to the rubrene doped OLED as shown in Figure 2. By applying 9 V pulsed voltage to the OLED, the modulation ratio output light is about 30%. In order to enhance the response of the OLED, it is effective to apply bias voltage to the OLED in addition to the pulsed voltage. The response of more than 100 MHz was obtained by applying pulsed voltage directly. The active size of the device was 0.01 mm^2.

The emission intensity reaches 30 mW/cm^2 at an applied voltage of 10 V for both the OLED fabricated on a glass and a polyimide substrates.

In Figure 3, emission characteristics are compared as a function of injection current for the OLED with 30-nm thick Alq_3 and 50-nm thick

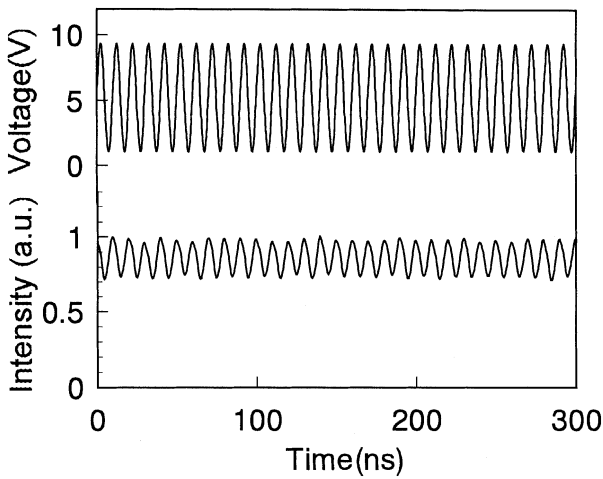


FIGURE 2 Direct modulation of OLED with applying pulsed voltage.

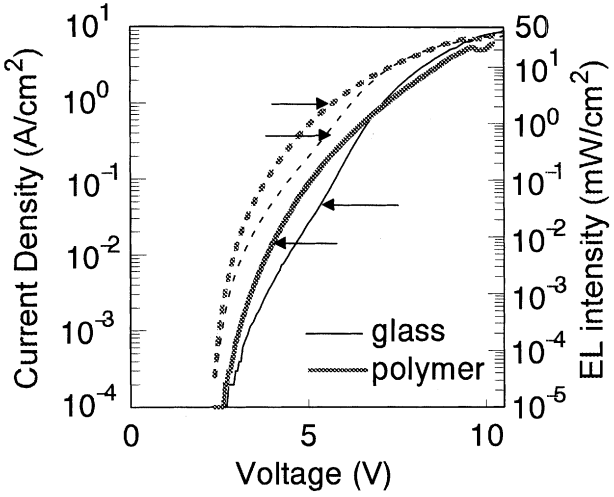


FIGURE 3 Emission characteristics and emission efficiency of OLED fabricated on a glass substrate and polymeric substrate.

α-NPD fabricated on a glass and polyimide substrate. The emission intensity is rather high in the device with polyimide substrate compared with that with glass substrate. The difference of the thickness of the substrate and the refractive index enhances the emission efficiency. At this stage of experiments, the emission characteristics of both the OLED fabricated on a glass substrate and a polymer substrate will be the same.

RESULTS AND DISCUSSIONS FOR ORGANIC PHOTOFETECTORS

TiOPc shows the absorption band region of 950–550 nm with the peak wavelength at 729 nm and a shoulder at 652 nm, and for F_{16}ZnPc there are the absorption band of 900–500 nm with the peaks at 645 nm and 810 nm. These results show that these materials have photo sensitive to the red LED (640 nm) used for this experiment.

A typical photo response of the multilayered TiOPc/F_{16}ZnPc hetero-structured device at a wavelength of 640 nm is shown in Figure 4. It is found that the device could respond to the light pulse at a few kHz of frequency even without applying reverse bias voltage. The cut-off frequency of the multilayered metal-phthalocyanine hetero-junction device reached to the limit at about 5 kHz without the external reverse field. The magnitude of current density was increased with the increase of the applying reverse bias

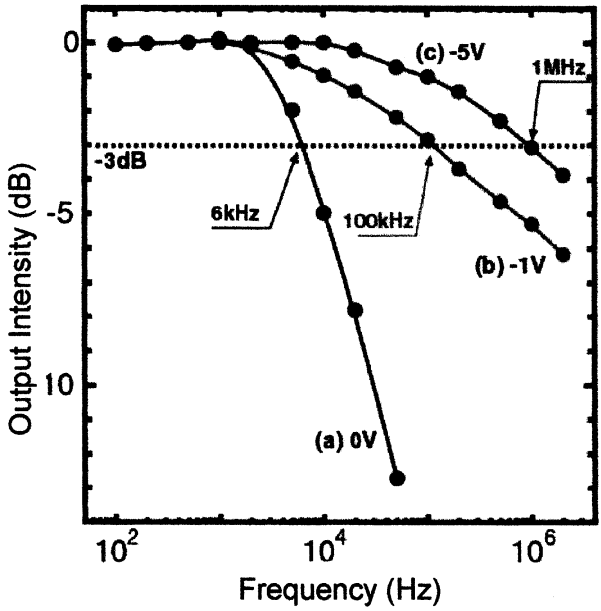


FIGURE 4 Frequency response of TiOPc/F_{16}ZnPc 5-period-multilayered photo-detectors with (a) no bias, (b) −1 V, and (c) −5 V.

voltage. This is the reason why the dissociation of photo-generated exciton in each interface in multilayer structure of metal-phthalocyanine thin films is accelerated by applying reverse bias to the device.

Figure 4 shows the photo response of 5 periods of multilayered TiOPc/F_{16}ZnPc devices with active area of 0.015 mm^2 at (a) no bias, (b) applied bias voltage of −1 V, and (c) −5 V, respectively. The 3 dB bandwidth of the OPD still remains at 6 kHz without applying any bias voltage. However, the response time of the photo detectors increased drastically by applying a reverse bias field. Under the reverse biased condition, the photo response of 10 periods of TiOPc/F_{16}ZnPc device shows the remarkable increase in cut-off frequency up to more than 1 MHz. The incident photon reached to the TiOPc and F_{16}ZnPc photo-absorption layers and the photo-generated excitons were effectively dissociated to free electrons and holes by external field applied, and then recombine in the thin film multi-layers resulting unto photocurrent in the external circuits. Furthermore, the acceleration of photo-generated carriers by applying reverse bias voltage improves the cut-off frequency of the multilayered device.

In Figure 5, the comparison of 3 dB bandwidth of OPDs are shown for (a) 1 period (individual layer thickness, $d =$ 20 nm), (b) 5 periods ($d = 4$ nm),

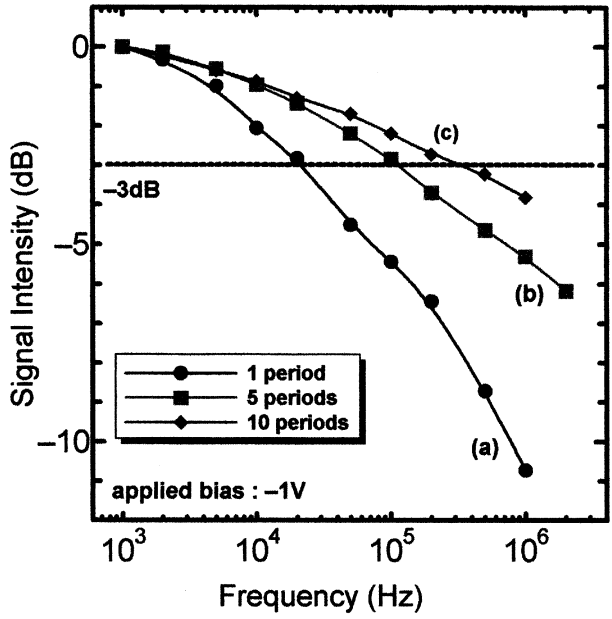


FIGURE 5 Frequency response of TiOPc/F_{16}ZnPc multilayered photo-detectors with (a) 1 period, (b) 5 periods and (c) 10 periods at an applied bias of −1 V.

and (c) 10 periods ($d = 2$ nm) of multilayered TiOPc/F_{16}ZnPc devices at an applied bias of −1 V. The cut-off frequency of 1 period, 5 periods and 10 periods of multilayered photo detector is 20 kHz, 100 kHz and 300 kHz, respectively. As decreasing the layer thickness, the response speed for the input light increased. It was found that the cut-off frequency remarkably increases with decreasing the layer thickness under the same total layers thickness. The results will be due to fast dissociation and recombination of photo-generated carriers in thin layers with decreasing the layer thickness and increasing the numbers of interfaces of the organic layers [6].

SUMMARY

In summary, emission characteristics and the modulation frequency are summarized for various kinds of emissive materials.

As far as emission intensity, OLEDs with α-NPD, Alq_3 and rubrene doped in Alq_3 as an emissive layer emit more than $30 \sim 40\,mW/cm^2$, which cover from blue to yellow light. Red light emitting TPP doped in Alq_3 shows about one order of magnitude poor compared with other emissive materials. As far as modulation frequency, the OLEDs with α-NPD, rubrene doped in Alq_3

and TPP doped in Alq_3 shows more than 100 MHz with direct modulation of voltage pulse. However, in case of the OLED with Alq_3 as an emissive layer, the modulation frequency limited to 60 MHz due to the fluorescence lifetime of the material.

The pulse response of photo detectors, which consists of multilayered metal phthalocyanines with TiOPc/F_{16}ZnPc has been discussed. To increase the number of interface is effective to the improvement of the cut-off frequency of OPD, and the cut-off frequency of 10 periods multilayered TiOPc/F_{16}ZnPc hetero-structured device shows more than 1 MHz at an applied bias of 5 V.

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