Optical "NOT" Operation in an Organic Film Device Combining OLED and Current Photo-controllable Organic Film

<u>Masaaki Yokoyama</u>, Masahiro Hiramoto, Yoshihito Motohashi, and Norio Nagayama Material and Life Science, Graduate School of Engineering, Osaka University, Yamadaoka, Suita, Osaka 565-0871, Japan

Abstract

An organic multi-layered optical logic device for twodimensional optical computing which can perform 'NOT' operation, was successfully demonstrated in the light transducer composed of a high-gain organic photoresponsive heterojunction between copper phthalocyanine (CuPc) and perylene pigment (Me-PTC) layers and an organic electroluminescent (EL) diode. EL output by the irradiation of red light exciting CuPc was effectively erased upon the superimposed irradiation of blue light exciting Me-PTC because of the effective suppression of multiplied photocurrent reaching 10^4 -fold at the CuPc/Me-PTC heterojunction. When a spatial pattern image was used for the superimposed blue light, EL output was erased as patterned with a high spatial resolution smaller than 280 µm.

Introduction

Recent remarkable progresses in the researches on organic electroluminescent diodes (OLED) are about to realize their practical use to flat panel display.¹⁴ Aiming to expand the potential of OLED to the advanced opto-electronic devices, we have already proposed novel applications including light transducers, in principle, based on the combination of antipodal functions of the electric-to-photo conversion in organic EL diode and the photo-to-electric conversion in photoconductive films such as amorphous silicon carbide⁵⁻⁷ and photoresponsive organic pigments.⁸⁻¹⁰ In the latter case, light amplification of the gain reaching 25-fold in photon conversion was accomplished.^{9,10} Such high photon conversion in the devices was achieved by the photocurrent multiplication phenomena exceeding 10⁵-fold occurring at the interface between a metal electrode and the photoresponsive organic films such as pervlene pigment,¹¹ quinacridone pigment¹² and naphthalene tetracarboxylic anhydride.¹³ The mechanism for the photocurrent multiplication at the metal/organic interface has been shown to be reasonably explained by tunneling electron injection from a metal electrode to the organic layer with the assistance of the high electric field built up by photo-accumulated space charges of trapped holes near the organic/metal interface.¹⁵

On the other hand, we have found later that similar photocurrent multiplication occurs even at organic/organic heterojunction between copper phthalocyanine (CuPc) and perylene pigment (Me-PTC). In this case, a very interesting phenomenon of the effective suppression of multiplied photocurrent was observed by superimposing the second light.¹⁴ This phenomenon provides a new aspect of photocontrol of the multiplied photocurrent. It seems, therefore, to be applicable to the photo-control of light output in the above-mentioned light transducer *via* the photo-modulation of the photocurrent multiplication at the organic heterojunction.

In this paper, we would like to report a successful demonstration of effective erasing of the EL output with a high spatial resolution in the light transducer composed of a high-gain CuPc/Me-PTC heterojunction multiplication device and an organic EL diode by means of the superimposition of blue light on red light.

Experimental

The device structure of the present light transducer is illustrated in Fig. 1. All films were deposited by vacuum evaporation technique under 1×10^{-3} Pa. An EL diode composed of hole transporting layer of aromatic amine (TPD) and light emitting layer of red fluorescent perylene derivative (t-BuPh-PTC) was successively deposited on an indium tin oxide (ITO) glass substrate coated with thin magnesium. Then, a high-gain organic double-layer composed of CuPc (500 nm) and Me-PTC (100 nm) were deposited. Finally, a semitransparent Au electrode was provided on the organic multi-layer. In order to confirm the photocurrent multiplication behavior in the CuPc/Me-PTC heterojunction, the double layered CuPc/Me-PTC heterojunction device without the EL layer was prepared separately on ITO glass substrate as shown in Fig. 2(a).

Measurements were made in an optical cryostat (Janis, Model VPF-475) evacuated to 0.1 Pa at room temperature. Monochromatic light from Xe lamp (Ushio, UI-501C) through monochromator (Shimadzu, SPG-100ST) and 488 nm-line of the Ar-ion laser (NEC, GLG3050) were used as light sources. Voltage application and current monitoring were performed using a source measure unit (Keithley, Model 238). Images of EL output from the device were detected using the CCD camera (Suruga Seiki Co., Ltd., Model VCS5720).



Fig. 1 The structure of organic multilayered optical logic device consisting of an organic EL diode and a high-gain organic photoresponsive heterojunction of CuPc and Me-PTC layers and the compounds used in the present study.

Results and Discussion

1. Photocurrent Multiplication at the Organic/Organic Heterojunction

First, the photocurrent multiplication at the organic/ organic interface is briefly described here since the present novel optical logic function comes from the unique characteristics of the high gain photo-to-electric conversion double-layer having organic/organic heterojunction between CuPc and Me-PTC pigments (Fig. 2(a)). An useful superiority of CuPc/Me-PTC double layer is that the selective excitation of each pigment can be possible with the different wavelength lights because of different absorption region of CuPc and Me-PTC films i.e., the irradiation of red light of 680 nm can excite only CuPc and the irradiation of blue light of 480 nm only Me-PTC.

When the ITO electrode was biased negatively, the photocurrent multiplication phenomenon occurring CuPc/Me-PTC interface was observed by the irradiation of red light (680 nm) exciting CuPc only. Multiplication rate,

which is defined as the ratio of the number of carriers flowing through the device to the number of photons absorbed by CuPc film, reached 10⁴-fold at 40 V. This means that 10^4 carriers flowed across the device by one input photon on the average. Interestingly, as clearly demonstrated in Fig. 2(b), under the occurrence of photocurrent multiplication by the irradiation of red light exciting CuPc, the superimposition of blue light (480 nm) exciting Me-PTC suppressed the multiplied photocurrent effectively. Response was quite reversible. Suppression was observed in the wavelength region of the superimposed second light from 400 to 540 nm, which corresponds well to the absorption region of Me-PTC film. Slow recovery of multiplied photocurrent after removing the blue light is a time progression necessary for space charge accumulation at the organic/organic heterojunction.



Fig. 2. The structure of organic heterojunction device (a) and typical photoresponse of miltiplied photocurrent by superimposed irradiation of the second blue light (b). In this case, photocurrent miltiplication of 2400-fold was obtained by irradiating red light for selectively excitation of CuPc pigment.

This photocurrent multiplication phenomenon is explained reasonably based on the tunneling injection of electrons from the valence band of CuPc to the conduction band of Me-PTC under a high electric field across the Me-PTC layer, which is built up by the photo-accumulation of trapped electrons in the CuPc pigment near the interface. Multiplication suppression is considered to be caused by the extinction of accumulated electrons due to the recombination with holes photogenerated by superimposed blue light in the Me-PTC film. Detailed multiplication and its suppression mechanism have already been reported in Ref. 14. The multiplied photocurrent thus observed at the organic/organic heterojunction reached several tens milliampere enough to drive organic EL devices and the effective suppression of more than 70% is sufficient to switch EL emission off.

2. Optical "NOT" Operation in the Light Transducer

In the present light transducer shown in Fig. 1, when Au electrode is positively biased with respect to ITO electrode under the irradiation of red input-light, a large number of holes are generated in CuPc due to the photocurrent multiplication process occurring at the CuPc/Me-PTC heterojunction. These holes are injected to t-BuPh-PTC through TPD. On the other hand, electrons are injected to t-BuPh-PTC from negatively biased Mg/ITO electrode. Recombination of injected electrons and holes in t-BuPh-PTC generates the red EL output of the present device.

Figure 3 illustrates the experimental sequence for superimposition of two lights in the light transducer along with the images of EL output detected by a CCD camera. (1) When the monochromatic red light of 680 nm was irradiated on the whole area of the device, red EL output was emitted from the whole active area of the opposite side. The active area is determined by the area of Au electrode, (2) Then, the short pulse of the second light of blue spot from Ar-ion laser was fed on the device. (3) The irradiation of blue light spot left a dark spot, which did not emit EL output, i.e., extinction of EL emission only in the portion irradiated with the second blue light. In addition, when the second blue spot was scanned quickly from left to right across the electrode, the stripe-shaped erase of EL emission was clearly observed following the track of the spot passed. The state (3) restored to the stage (1) gradually. Thus, in the present device, the EL output caused by the first red inputlight was erased only in the corresponding part superimposed with the second blue light. From the standpoint of logic operation, the present result can be regarded as an optical "NOT" operation since the illuminated part was darkened. Optical image processing devices require high spatial resolution. A typical result concerning the spatial resolution of the present device is demonstrated in Fig. 4. The collimated second blue light was irradiated through the photomask having the slim linepair patterns instead of a spot. EL output pattern was recorded with a CCD camera. Obviously, original pattern of second blue light was reproduced in the erased pattern of EL output. In a preliminary experiment, we confirmed the spatial resolution of 280 µm line-pair. Resolution is, however, limited by the spatial resolution of detection system, especially due to the performances of CCD camera and image edition of video frames by computer. Since the total film thickness of organic multi-layers in the present device was less than 1 μ m, a spatial resolution of near 1 μ m would be expected if the carrier diffusion length lateral to the organic film of about 1 µm was taken into account.



Fig. 3 Illustration of optical "NOT" operation by irradiating two different wavelength lights in the device combining organic EL and organic photocurrent multiplication double-layers and video frames of EL output image for each stage taken with CCD camera.



Fig. 4. The EL output image(b) for the irradiation of patterned second light through a photo-mask (a).

Conclusion

In conclusion, a new type of light transducer using organic EL diode coupled with a high-gain organic photoresponsive heterojunction of CuPc and Me-PTC was fabricated. Red EL output by the irradiation of red light exciting CuPc was reversibly erased by the superimposed irradiation of blue light exciting Me-PTC due to the effective suppression of photocurrent multiplication at CuPc/Me-PTC heterojunction. High spatial resolution of about 300 μ m for the optical image was confirmed. The present device is expected to

process an optical image directly and used as a new type of optical logic device for two-dimensional optical computing which can perform 'NOT' operation.

References

- 1. C. W. Tang and S. A. VanSlyke, Appl. Phys. Lett., **51**, 913 (1987).
- C. W. Tang, S. A. VanSlyke, and C. H. Chen, J. Appl. Phys., 65, 3610 (1989).
- G. Gustafsson, Y. Cao, G. M. Treacy, F. Klavetter, N. Colaneri, and A. J. Heeger, Nature, 357, 477 (1992).
- 4. T. Tsutsui, Oyo Buturi, **66**, 109 (1997).
- 5. M. Hiramoto, T. Miyao, and M. Yokoyama, Appl. Phys. Lett., **57**, 1625 (1990).
- 6. M. Hiramoto, K. Yoshimura, T. Miyao, and M. Yokoyama, Appl. Phys. Lett., **58**, 1146 (1991).
- 7. M. Hiramoto, K. Yoshimura, and M. Yokoyama, Appl. Phys. Lett., **60**, 324 (1992).
- T. Katsume, M. Hiramoto, and M. Yokoyama, Appl. Phys. Lett., 64, 2546 (1994).
- 9. M. Hiramoto, T. Katsume, and M. Yokoyama, Optical Review, 1, 82 (1994).
- T. Katsume, M. Hiramoto, and M. Yokoyama, Appl. Phys. Lett., 66, 2992 (1995).

- 11. M. Hiramoto, T. Imahigashi, and M. Yokoyama, Appl. Phys. Lett., **64**, 187 (1994).
- M. Hiramoto, S. Kawase, and M. Yokoyama, Jpn. J. Appl. Phys., 35, L349 (1996).
- 13. T. Katsume, M. Hiramoto, and M. Yokoyama, Appl. Phys. Lett., **69**, 3722 (1996).
- M. Hiramoto, H. Kumaoka, and M. Yokoyama, Proceedings of IS&T's NIP13 International Conference on Digital Printing Technologies, pp248-251, Nov. 1997, Seattle, USA.
- K. Nakayama, M. Hiramoto, and M. Yokoyama, J. Appl. Phys., 84, 6154(1998).
- M. Hiramoto, K. Nakayama, T. Katsume, and M. Yokoyama, Appl. Phys. Lett., 73, 2627(1998).

Biography

Prof. Masaaki Yokoyama received the B.Sc. in 1966, the M.Sc. in 1968 and the Ph. D. in 1971 from Osaka University in polymer science. Since 1971 he has worked in the Faculty of Engineering, Osaka University, and in 1987 became professor. His current interests involve the researches in the field of material science including photoconducting organics and their application to new optoelectronic and photonic devices.