Extended Abstracts of the 2013 International Conference on Solid State Devices and Materials, Fukuoka, 2013, pp1120-1121

Detection of pre-electrical breakdown phenomena of IZO/α-NPD/Alq3/Al light-emitting diodes by electric-field-induced optical second-harmonic generation measurement

Dai Taguchi, Ryo Nakamoto, Takaaki Manaka, Mitsumasa Iwamoto
Department of Physical Electronics, Tokyo Institute of Technology
2-12-1 S3-33, O-okayama, Meguro-ku, Tokyo 152-8552, Japan
Phone: +81-3-5734-2191 E-mail: iwamoto@pe.titech.ac.jp

Abstract

By using the electric-field-induced optical second harmonic generation (EFISHG) measurement, we directly measured interfacial charge accumulation in IZO/α-NPD/Alq3/Al OLEDs, whilst electroluminescence is being activated. EFISHG pulses indicating excess electron charging were detected as signals of pre-electrical breakdown phenomena. The EFISHG is available for the evaluation of life-time of OLEDs.

1. Introduction

Organic light-emitting diodes (OLEDs) have been intensively studied for practical use as displays in electronics [1], where the improvement of device life time is one of main research subject. As a result, the luminance decay is measured to estimate the OLED life time. Lots of origins are considered to account for the decaying mechanism, on the basis of the luminescence-time (*L-t*) curves recorded. Electrical breakdown during the device operation is considered as one of the most important origins. In the field of electrical insulation engineering, “Electro-luminance (EL)” is well-known as pre-electrical breakdown phenomena of insulators which are being subjected to high electric fields [2,3]. Therefore detection of EL related to “pre-electrical breakdown phenomenon” will be helpful to study the OLED life time. In our previous study using indium-zinc-oxide (IZO)/tetracene/Al diodes [4], we showed that EL is activated via intrinsic EL process through electron-hole recombination process in bulk, but EL can be also activated via interfacial states. Interestingly, these two ELs were enhanced at different wavelengths. Further we could show that there are two EL modes in IZO/α-NPD/Alq3/Al diodes, depending on charge accumulation at the double-layer interface, and so forth, by using the electric-field-induced optical second-harmonic generation (EFISHG) measurement that is capable of directly probing electron and hole behaviors [5]. Results motivated us to detect pre-electrical breakdown phenomenon induced in OLEDs by using the EFISHG measurement.

In our previous study using the EFISHG measurement, we showed that excessive holes are being accumulated at the α-NPD/Alq3 interface during the ordinary EL emission is activated [6-8]. In this study, we focused on the detection of pre-electrical breakdown in OLEDs by using the EFISHG measurement.

2. Experimental

Figure 1 illustrates an optical arrangement for the EFISHG measurement and IZO/α-NPD/Alq3/Al diode used here. The diode was prepared in a manner as described in [6], using the vacuum evaporation method. The thickness of α-NPD layer and Alq3 layer was 50 nm. Al electrode was 100 nm in thickness. *L-t* and EFISHG measurements were carried out by applying DC voltage to the IZO electrode with respect to the Al electrode grounded. For the EFISHG measurement, a laser pulse generated from the third-order harmonic light of Nd:YAG laser coupled with an optical parametric oscillator was used as an optical probe (average power 1 mW, repetition rate 10 Hz, duration 4 ns). *p*-polarized laser pulses were irradiated on the diode from the IZO electrode side at an incident angle of 45°. The wavelength of the laser beam was set at 820 nm (the wavelength of EFISHG at 410 nm) to selectively probe the electric field in the α-NPD layer [6]. The laser pulses impinged onto the OLED sample, and the EFISHG radiated from the α-NPD layer was measured. Note that there is no generation of EFISHG from IZO, Alq3, and Al. The EFISHGs were detected by the photomultiplier tube (PMT), where laser pulses reflected from the diode were eliminated using optical filters. The EFISHG radiated from the layer is given by [9]

$$I(2\omega) \propto |\chi^{(3)} \vdots E(0)E(\omega)E(\omega)|^2. \quad (1)$$

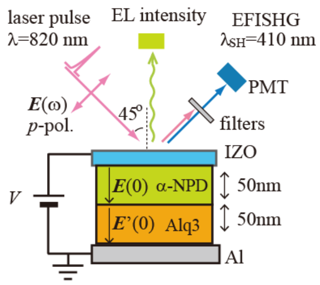

Figure 1: Optical arrangement and IZO/α-NPD/Alq3/Al OLEDs used here.

Here $\chi^{(3)}$ is the third order susceptibility, $E(0)$ is a local DC field, and $E(\omega)$ is electric field of the laser pulse. The electrostatic field $E(0)$ in α-NPD layer is described as sum of an external field by charges Q_m on electrode and a space charge field caused by charge Q_s accumulated at the α-NPD/Alq3 interface due to the Maxwell-Wagner effect [8,9]. That is, modeling the IZO/α-NPD/Alq3/Al diodes as a double-layer dielectric system, the electric field $E(0)$ is given using charge Q_m induced on electrode, and interfacial charges Q_{s1} and Q_{s2} as

$$E(0) = Q_m/\varepsilon_1 \varepsilon_0 - 1/d_1 \cdot (Q_{s1} + Q_{s2})/(C_1+C_2), \quad (2)$$

where ε_1 is the relative permittivity of the α-NPD, ε_0 is the dielectric constant of vacuum, d_1 is the α-NPD layer thickness, and C_1 (C_2) is the capacitance of the α-NPD (Alq3) layer. Q_{s1} is accumulated charge in steady state, and Q_{s2} is interface charge leading to the pre-electrical breakdown and it is assumed to appear as a trigger of pre-breakdown.

3. Results and Discussion

Figure 2 shows the *L-t* characteristic of the IZO/α-NPD/Alq3/Al diode, under applying a DC voltage of 25 V. Results showed that the luminance decays in a 3-step process; (I) initial rising process in the time 0-1 s, (II) luminance decay process, in a time 1-320 s, (III) luminance interrupted process at 320 s, due to electrical breakdown. Similar results were observed for other OLED devices we fabricated, where the interval of the initial rising and decaying times decrease as the applied DC voltage increases.

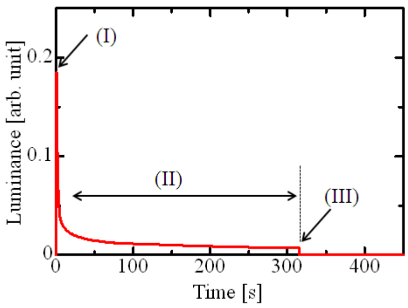

Figure 2: Luminance-time curve of the IZO/α-NPD/Alq3/Al OLED.

Figure 3 shows the EFISHG of the OLED with time. Results showed that the EFISHG intensity is nearly constant in the initial rising and luminance decay processes (see Fig. 2), suggesting that Q_m and Q_{s1} are nearly constants during these processes. On the other hand, as a pre-breakdown phenomenon, we can see EFISHG peaks as indicated by arrows in Fig. 3. The appearance of the EFISHG peaks suggests additional space charge field $E_s > 0$ is created, possibly due to accumulation of electrons $Q_{s2} < 0$ at the α-NPD/Alq3 interface. With taking into account the results of Fig. 2 and Fig. 3, we may argue that EFISHG pulses are signals of charge accumulation which triggers electrical breakdown, i.e., pre-electrical breakdown phenomenon. Noteworthy that the EL is used for detecting pre-breakdown phenomena in insulators, but our EFISHG results suggested that pre-breakdown phenomenon can be probed more sensitive as electron charging phenomena, before EL emission is activated.

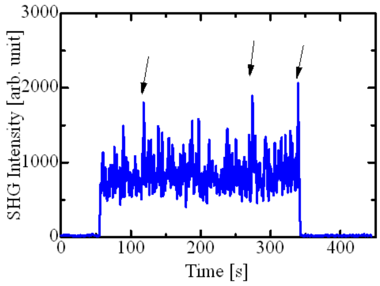

Figure 3: EFISHG intensity of the IZO/α-NPD/Alq3/Al OLED. DC voltage was applied from 50 s, and the OLED was electrically broken down at 340 s.

4. Conclusions

By using the EFISHG measurement, pre-breakdown phenomena of α-NPD/Alq3 OLEDs was studied. Luminance decayed in a 3-step process (1) initial EL rising process, (2) luminance decaying process, and (3) electrical breakdown process. EFISHG measurement showed that interfacial charge Q_{s1} at the α-NPD/Alq3 is nearly constant, but additional electron charging Q_{s2} leading to the pre-electrical breakdown is detectable by means of the EFISHG.

Acknowledgements

A part of this work was financially supported by a Grant-in-Aid for Scientific Research (Nos. 22226007, 25709022) from the Japan Society for the Promotion of Science.

References

[1] T. Tsujimura: *OLED Display Fundamentals and Applications* (Wiely, Weinheim, 2012).

[2] N. Shimizu, H. Katsukawa, M. Miyauchi, M. Kosaki and K. Horii: IEEE Trans. Electr. Insul. **14** (1979) 256.

[3] T. Mizuno, Y. S. Liu, W. Shionoya, K. Yasuoka, S. Ishii, H. Miyata and A. Yokoyama: IEEE Trans. Dielectr. Electr. Insul. **4** (1997) 433.

[4] A. Sadakata, Y. Ohshima, D. Taguchi, M. Fukuzawa, T. Manaka and M. Iwamoto: Mol. Cryst. Liq. Cryst. **538** (2011) 112.

[5] A. Sadakata, K. Osada, D. Taguchi, T. Yamamoto, M. Fukuzawa, T. Manaka and M. Iwamoto: J. Appl. Phys. **112** (2012) 083723.

[6] D. Taguchi, M. Weis, T. Manaka and M. Iwamoto: Appl. Phys. Lett. **95** (2009) 263310.

[7] D. Taguchi, S. Inoue, L. Zhang, J. Li, M. Weis, T. Manaka and M. Iwamoto: J. Phys. Chem. Lett. **1** (2010) 803.

[8] D. Taguchi, L. Zhang, J. Li, M. Weis, T. Manaka and M. Iwamoto: J. Phys. Chem. C **114** (2010) 15136.

[9] M. Iwamoto, T. Manaka, M. Weis and D. Taguchi: J. Vac. Sci. Technol. B **28** (2010) C5F12.