# UV and Visible range Electroluminescence from MOS Devices Fabricated by Spin-Coating of Gd/Dy Organic Compound Films on Silicon

Toshihiro Matsuda<sup>1</sup>, Shogo Saito<sup>1</sup>, Hideyuki Iwata<sup>1</sup>, and Takashi Ohzone<sup>2</sup>

 <sup>1</sup> Toyama Prefectural Univ. Kurokawa, Imizu, Toyama 939-0398, Japan
Phone: +81-766-56-7500 E-mail: matsuda@pu-toyama.ac.jp
<sup>2</sup> Dawn Enterprise Co., Ltd. Nagoya 467-0808, Japan

#### Abstract

UV and visible range EL from MOS devices with spin-coated Gd silicate-related oxide having additive elements of Dy is presented. The devices emitted UV and visible wavelength range EL, which originated from the intrashell transitions in Gd<sup>3+</sup> and Dy<sup>3+</sup>, respectively. The surface silicate layer is analyzed by the XPS depth profiles and an EL mechanism is discussed.

# 1. Introduction

Si-based light-emitting devices, which have superior process compatibility to Si metal-oxide-semiconductor (MOS) LSIs, are very attractive for realizing microdisplays in portable systems as well as optical interconnections reducing metal wire delay. Recently, EL in ultraviolet (UV) through IR range from MOS devices with rare-earth elements were reported [1,2]. MOS devices fabricated with the spin-coated organic compound films of Tb/Eu showed the green and red EL [3,4].

In this study, we demonstrate UV and visible range EL from MOS devices with spin-coated gadolinium (Gd) silicate-related oxide having additive elements of dysprosium (Dy). The surface silicate layer is analyzed by the X-ray photoelectron spectroscopy (XPS) depth profiles and a model of photoemission mechanism is also discussed.

# 2. MOS EL Devices and Measurement Setup

Fig. 1(a) shows a schematic cross section of a MOS device with a [Gd/(Gd+Dy)-Si-O] layer. Organic compound liquids of Gd and Dy were spin-coated on n<sup>+</sup>-Si. Device #1 was fabricated with a single liquid source of Gd. The atomic ratios (Gd : Dy) in mixture liquids for device #2 was prepared to be (10 : 1), where Gd and (Gd : Dy)

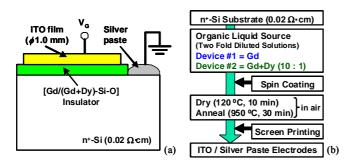


Fig. 1 (a) A schematic cross section of a MOS device and (b) fabrication process steps.

mixture liquids were two fold diluted with the thinner liquid. The fabrication process steps are shown in Fig.2 (b).

The EL spectra were measured by a back-illuminated CCD camera stabilized at -70 °C with a monochromator under a constant current supply. The wavelength range from 290 to 1000 nm can be detected at once [3,4].

# 3. Results and Discussion

Fig. 2(a) shows the forward  $+I_G vs +V_G$  characteristics. The rising voltage  $+V_R$ , where  $+I_G$  increased steeply, was about +11 V for both devices #1 and #2. The linear relation of the Fowler-Nordheim (FN) plot in Fig. 2(b) confirms the FN tunnel current and the injection of hot electrons into the insulator from Si substrate.

Fig. 3 shows microphotographs of (a) an ITO electrode in #2, and (b) an EL color image of devices #2. Since EL could not be observed from the devices with the single element liquid source of Dy, the devices with strong white EL were realized by mixing the Gd solution with the Dy solutions.

Fig. 4 shows EL spectra of #1 and #2 in the wavelength range of (a)  $300 \sim 1000$  nm and (b)  $300 \sim 330$  nm (UV range). Device #2 emitted strong visible light with narrow peaks but less UV light. Device #1 had smaller visible components, which contained the broad peaks superimposed on narrow peaks.

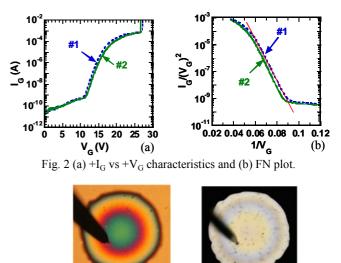


Fig. 3 Microphotographs of (a) an ITO electrode in #2, and (b) an EL color image of devices #2.

(b)

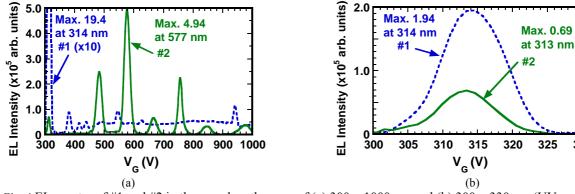


Fig. 4 EL spectra of #1 and #2 in the wavelength range of (a)  $300 \sim 1000$  nm and (b)  $300 \sim 330$  nm (UV range).

Fig. 5 shows the extraction of Gaussian distribution functions  $G_i$  for EL of #2. The spectra are normalized to the maximum intensity and fitted to the following equation: G<sub>i</sub> = M<sub>i</sub> exp[ - (E - E<sub>i</sub>)<sup>2</sup>/2( $\sigma_i$ )<sup>2</sup>], where M<sub>i</sub>, E<sub>i</sub>, and  $\sigma_i$  are the maximum EL intensity, the center energy, and the standard deviation, respectively. E<sub>4</sub> in #2 is the intrashell transitions of  ${}^{6}P_{7/2} - {}^{8}S_{7/2}$  [3.97 eV] in Gd<sup>3+</sup> ions [1]. Energy levels E<sub>1</sub>~  $E_3$ , and  $E_5$  observed in #2 correspond well to the intrashell transitions of  ${}^{4}F_{9/2}$  -  ${}^{6}H_{J/2}$  (J = 13, 15, 9, and 11) in the Dy<sup>3+</sup> ions [2].  $E_6$  and  $E_7$  appear to correspond to the transitions of  ${}^{4}F_{9/2}$  -  ${}^{6}H_{J/2}$  (J = 5 and 7) in Dy<sup>3+</sup>. The higher energy electrons accelerated by the electric fields in #2 are transferred to the Gd<sup>3+</sup> ions and excite the state from the  ${}^{8}S_{7/2}$  to  ${}^{6}P_{7/2}$ . Then, the electron energy is transferred from Gd<sup>3+</sup> to Dy<sup>3+</sup>, followed by the radiative transitions from  ${}^{4}F_{9/2}$  to  ${}^{6}H_{J/2}$  in the Dy $^{3+}$  ions. As for #1, the peak around 310 nm corresponds to the transition in  $Gd^{3+}$ . The origins of the other peaks in the visible range, which may be related to Gd and/or defects in the oxide, are not clear as of now.

Fig. 6 shows the atomic concentration profiles of Gd, O and Si in addition to (a) C for #1, (b) Dy for #2 analyzed by XPS. The Gd concentration became the highest around 3 nm from the surface, and then gradually decreased toward the Si substrate. The profile of Dy was similar to that of Gd. Inside the cross point of Gd and Si concentrations, the Si/O ratios of about 1/2 suggested that the layers were SiO<sub>x</sub> rich oxides with the Gd and/or Dy composites. The schematic cross sections of the devices estimated from XPS spectrum analyses are shown in Fig. 7. The first surface 6 nm thick layer consists of  $[Gd_2O_3/(DyO_x)]$  and Gd/(Gd+Dy)-Si-O]. The second 23 nm thick layer is the SiO<sub>x</sub> rich oxides with the Gd and/or Dy composites.

#### 4. Conclusions

UV and visible range EL characteristics are reported for (ITO)/[(Gd/(Gd+Dy)-Si-O] insulator/n<sup>+</sup>-Si substrate MOS devices, which were fabricated from the spin-coated films with organic liquid sources of (Gd) or (Gd+Dy). The devices with (Gd+Dy) emitted UV and visible wavelength range EL, which originated from the intrashell transitions in Gd<sup>3+</sup> and Dy<sup>3+</sup>, respectively. The insulator films consist of [Gd<sub>2</sub>O<sub>3</sub>/(DyO<sub>x</sub>) and Gd/(Gd+Dy)-Si-O], and the SiO<sub>x</sub> rich oxide with the Gd and/or Dy composites.

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#### References

- [1] J. M. Sun et al., J. Appl. Phys. 99 (2006) 103102.
- [2] N. Miura et al., Jpn. J. Appl. Phys., 31 (1992) 51.
- [3] T. Ohzone et al., Jpn. J. Appl. Phys. 49 (2010) 084102.
- [4] T. Ohzone et al., Jpn. J. Appl. Phys. 50 (2011) 064102.

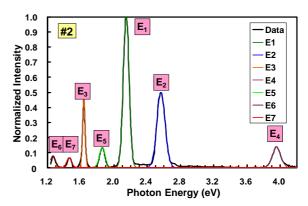


Fig. 5 Extraction of Gaussian distribution functions G<sub>i</sub> for EL of #2

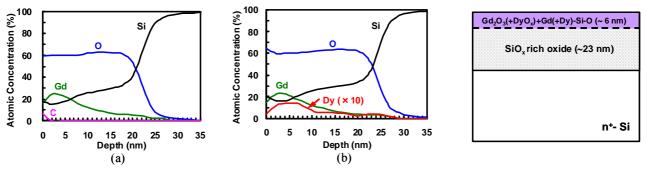


Fig. 6 Atomic concentration profiles of Gd, O, Si, (a) C for #1, (b) Dy for #2.

Fig. 7 Estimated schematic cross sections of #1/#2.